

UNIVERSITI PUTRA MALAYSIA

DISTRIBUTION AND ASSESSMENT OF METALS AND METHYL MERCURY IN INTERTIDAL SURFACE SEDIMENT AND SNAIL, Nerita lineata (CHEMNITZ, 1774) IN PORT KLANG, SELANGOR, MALAYSIA

HAZZEMAN HARIS

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

December 2015

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Doctor of Philosophy



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DISTRIBUTION AND ASSESSMENT OF METALS AND METHYL MERCURY IN INTERTIDAL SURFACE SEDIMENT AND SNAIL, *Nerita lineata* (CHEMNITZ, 1774) IN PORT KLANG, SELANGOR, MALAYSIA

By

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December 2015

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Coastal mangrove and estuarine areas are important to the environment. However, these areas are also vulnerable to the accumulation of metals in its sediment, which can be detrimental to the health of the aquatic ecosystem. Therefore, this study was conducted with the objectives of determining the distribution, enrichment and the factors affecting metals retention in sediment. Other than that, this study also aims to explore Nerita lineata potential as a biomonitor of metals in sediment and the speciation of Hg in both of these matrixes. This is because currently there are only a few studies on Hg speciation in the Malaysian environment and no study was ever conducted to determine if field collected N. lineata is suitable as a biomonitor for Co, Cr, Hg, MeHg and Mn. In order to achieve these objectives, 30 intertidal sediment samples were collected from sampling points along the coast of Port Klang, which covers the Lumut Strait, parts of the South Klang Strait and the estuaries of both the Klang River and the Langat River. The samples were then prepared according to the standard used when studying metals and mercury speciation in order to ensure data accuracy and reliability. The water and sediment samples were measured for their pH, salinity and electrical conductivity. The total dissolved solids in water and sediment particle size were also determined. Apart from that, samples of N. lineata (15 individuals per sampling station) were also collected from five predetermined sampling stations based on preliminary studies conducted. Their soft tissue was also measured for metal concentration. In general, the mean sediment metal concentration in descending order were; Fe>Mn>Zn>Cr>Pb>Ni>Cu>Co>Cd>Hg. The metal's concentration pattern in this study differs than those previously reported in the Port Klang coastal area. This suggests that the pollution sources for this study area are also different. Accumulation of metals in the sediment was found to occur near or within the Klang and the Langat River estuaries and along the Lumut Strait. Cd, Cu, Hg, Pb and Zn were found to be enriched in the sediment as indicated by the various indices calculated. The concentration of Cr, Cu and Hg were found to exceed some of the sediment quality guidelines used around the world and the adverse effects index (AEI) also indicated that Cu and Hg at several stations could produce adverse effects to the adjacent biota. Areas within the Lumut Strait and the Klang River estuary were identified as experiencing higher ecological and toxicity risk due to the accumulation of metals. Based on investigation via the use of analytical analysis of sediment, site observation, the use of geochemical analysis and multivariate analyses, it can be



concluded that the anthropogenic sources (i.e. steel related industries, shipyard, marina and jetty) from the upstream of both the Klang River and the Langat River as well as those within the study area play a crucial part in the enrichment of metals in the sediment of Port Klang. The presence of organic matter and fine grain sediment such as clay and silt were found to favour metal retention in the sediment. The soft tissue of N. lineata was found to have a mean metals concentration in the order of Fe>Zn>Mn>Ni>Cu>Pb>Co>Cr>Hg. The concentration of Cu, Mn and Pb in the soft tissue were significantly (p<0.05) positively correlated with the corresponding metals in sediment. The result of biota-sediment accumulation factor (BSAF) also indicated that Cu, Ni, Hg and MeHg were bio accumulated in N. lineata. All of this suggests that N. lineata has the potential to be used as a bioindicator for Cu, Mn, Ni, Pb, Hg and MeHg. Speciation result for Hg found that MeHg in sediment can make up from 0.06% to 94.95% of total Hg, while in N. lineata MeHg can be from 3.97% to 88.33% of total Hg. Through this study, the influence of anthropogenic activity and geographical feature on the distribution and enrichment of metals in sediment and the current state of Hg speciation in the Malaysian environment were further understood. Apart from confirming the reported ability of *N. lineata* as a biomonitor for Cu, Ni and Pb, this study also provides new information on its potential as a biomonitor for Hg, MeHg and Mn.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

TABURAN DAN PENILAIAN LOGAM SERTA METIL MERKURI DI DALAM SEDIMEN PERMUKAAN DI KAWASAN PASANG SURUT DAN SIPUT, *Nerita lineata* (CHEMNITZ, 1774) DI PELABUHAN KLANG, SELANGOR, MALAYSIA

Oleh

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Kawasan paya bakau dan muara sungai adalah penting kepada ekosistem. Tetapi kawasan ini juga terdedah kepada pengumpulan logam di dalam sedimen yang boleh menyebabkan kesan buruk kepada ekosistem. Oleh yang demikian, kajian ini telah dijalankan dengan objektif untuk menentukan taburan logam, kadar pengayaan dan faktor yang mempengaruhi kandungannya di dalam sedimen. Selain dari itu, kajian ini juga bertujuan untuk meneroka potensi Nerita lineata sebagai penunkuk biologi kepada logam di dalam sedimen dan juga penspesiesan Hg di dalam kedua-dua matrik ini. Ini kerana pada masa kini hanya terdapat beberapa kajian sahaja mengenai penspesiesan Hg di persekitaran Malaysia dan tiada sebarang kajian pernah dilakukan untuk menentukan sama ada *N. lineata* yang diambil dari lapangan sesuai sebagai penunjuk biologi untuk Co, Cr, Hg, MeHg dan Mn. Demi mencapai objektif ini, tiga puluh sampel sedimen dari kawasan pasang surut telah diambil dari kawasan persampelan di sepanjang pesisir pantai Pelabuhan Klang yang turut meliputi Selat Lumut, sebahagian dari Selat Klang Selatan dan juga muara Sungai Klang dan Sungai Langat. Sampel ini kemudian disediakan mengikut kaedah piawai yang digunakan untuk kajian logam dan penspesiesan Hg bagi memastikan ketepatan dan kebolehpercayaan data. Sampel air dan sediment dianalisis untuk menentukan nilai pH, kemasinan dan kekonduksian elektrik. Kandungan pepejal terlarut di dalam air dan saiz partikel sedimen turut ditentukan. Selain dari itu, sampel N. lineata (15 individu per stesen persampelan) dikutip dari lima stesen persampelan yang telah ditentukan berdasarkan kepada kajian awal yang dilakukan. Tisu lembut dari N. lineata ini kemudian dianalisis untuk menentukan kandungan logam. Secara amnya, purata kandungan logam di dalam sedimen adalah mengikut turutan menurun seperti berikut: Fe>Mn>Zn>Cr>Pb>Ni>Cu>Co>Cd>Hg. Pengumpulan logam di dalam sedimen didapati berlaku di kawasan muara Sungai Klang dan muara Sungai Langat serta di sepanjang Selat Lumut. Pengiraan pelbagai indeks mendapati pengayaan Cd, Cu, Hg, Pb dan Zn pada sedimen turut berlaku. Kandungan Cr, Cu dan Hg di dalam sedimen turut melebihi beberapa garis panduan kualiti sedimen yang digunakan di seluruh dunia dan indeks kesan buruk (AEI) turut menunjukkan yang kandungan Cu dan Hg pada beberapa stesen boleh menyebabkan kesan buruk kepada biota yang berdekatan. Kawasan Selat Lumut dan muara Sungai Klang juga mempunyai risiko ekologi dan

toksikologi yang tinggi akibat pengumpulan logam. Berdasarkan penyiasatan menggunakan kaedah analisis sedimen, pemerhatian lapangan, analisis geologi dan multivariat, maka dapat disimpulkan bahawa pencemaran logam oleh sumber antropogenik (seperti industri berkaitan besi, limbungan, marina dan jeti) di bahagian hulu Sungai Klang dan Sungai Langat serta di dalam kawasan kajian tersebut memainkan peranan penting dalam pengayaan logam di dalam sedimen di Pelabuhan Klang. Kehadiran bahan organik dan partikel sedimen yang halus seperti tanah liat dan lempung dapat meningkatkan kadar penyerapan logam oleh sedimen. Tisu lembut N. lineata mempunyai purata kandungan logam mengikut turutan Fe>Zn>Mn>Ni>Cu>Pb>Co>Cr>Hg. Kandungan Cu, Mn dan Pb di dalam tisu lembut *N. lineata* didapati mempunyai korelasi positif yang signifikan (p<0.05) dengan logam yang sepadan di dalam sedimen. Keputusan faktor pengumpulan biota-sedimen (BSAF) juga menunjukkan yang Cu, Ni, Hg dan MeHg telah dibio-akumulasi oleh N. lineata. Kesemua ini menunjukkan yang N. lineata mempunyai potensi untuk digunakan sebagai penunjuk biologi bagi Cu, Mn, Ni, Pb, Hg dan MeHg. Keputusan penspesiesan Hg menunjukkan yang 0.06% hingga 94.95% daripada keseluruhan Hg dalam sedimen adalah MeHg, manakala MeHg mewakili antara 3.97% hingga 88.33% daripada jumlah Hg dalam tisu N. lineata. Melalui kajian ini, pengaruh terhadap taburan dan pengayaan logam pada sedimen serta penspesiesan Hg di persekitaran Malaysia dapat lebih difahami. Selain dari mengesahkan keupayaan N. lineata sebagai penunjuk biologi untuk Cu, Ni dan Pb, kajian ini juga memberikan maklumat baharu mengenai potensinya sebagai penunjuk biologi untuk Hg, MeHg dan Mn.

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I certify that a Thesis Examination Committee has met on 30 December 2015 to conduct the final examination of Hazzeman bin Haris on his thesis entitled "Distribution and Assessment of Metals and Methyl Mercury in Intertidal Surface Sediment and Snail, *Nerita lineata* (Chemnitz, 1774) in Port Klang, Selangor, Malaysia" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Doctor of Philosophy.

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LIST OF ABRREVIATIONS

AEI	Adverse Effects Index
AMAP	Artic Monitoring and Assessment Programme
ANZECC	Australian and New Zealand Environment and
	Conservation Council
ARMCANZ	Agriculture and Resource Management Council of
	Australia and New Zealand
ATSDR	Agency for Toxic Substances and Disease Registry
BSAF	Biota-Sediment Accumulation Index
С	Carbon
CCME	Canadian Council of Ministers of the Environment
Cd	Cadmium
CF	Contamination Factor
Со	Cobalt
Cr	Chromium
Cu	Cuprum or Copper
EC	Electrical conductivity
ECHA	European Chemical Agency
EF	Enrichment Factor
Eh	Redox Potential
EPA	Environmental Protection Agency
Er	Risk Factor
ERL	Effect Range Low
ERM	Effect Range Medium
FDEP	Florida Department of Environmental Protection
Fe	Ferrum or iron
Н	Hydrogen
HCA	Hierarchical Cluster Analysis
Hg	Mercury
ICM	Integrated Coastal Management
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
I _{geo}	Geo-accumulation index
LEL	Lowest Effect Level
LOI	Loss on ignition
ISQG	Interim Sediment Quality Guidelines
ISQV	Interim Sediment Quality Value
mCd	Modified Degree of Contamination
MeHg	Methylmercury
MMNRE	Malaysia Ministry of Natural Resources and
	Environment
Mn	Manganese
MOE	Ministry of Environment
m-PEL-Q	Mean Probable Effect Level Quotient
MSPI	Marine Sediment Pollution Index
	Nickel National Oceania and Atmospheric Administration
NUAA O	National Oceanic and Atmospheric Administration
U Dh	Uxygen Lood or Dhumhum
	Leau of Plumbum Dringing Component Anglesis
PUA	Principal Component Analysis

Probable Effect Level Partnerships in Environmental Management for the Seas of East Asia
Pollution Load Index
Particle Size Analysis
Polyvinyl chloride
Risk Index
Sulfur
Severe Effect Level
Statistical Package for Social Science
Sediment Quality Objective
State Water Resources Control Board
Total Dissolved Solids
Threshold Effect Level
United Nation Environment Programme
United State Department of Agriculture
United State Environmental Protection Agency
World Register of Marine Species
Zinc

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LIST OF UNITS

%	percent	
<	less than	
\leq	less or equal to	
>	more than	
\geq	more than or equal to	
° C	degrees Celsius	
μS/cm	micro Siemens per centimeter	
μm	micrometer	
cm	centimeter	
g	gram	
ha	hectare	
km	kilometer	
km2	square kilometer	
m	meter	
mg/kg	milligrams per kilogram	
mg/L	miligram per liter	
mV	milivolt	
p.a.	per annum or per annual	
ppt	part per thousand	
meq	miliequivalent	
meq/100g	milliequivalents per 100 grams	
cmol _c	centimoles of charge	
cmol _c /kgcentimoles of charge per kilogram		

CHAPTER 1

INTRODUCTION

1.1 Background Study

The mangrove and estuarine areas have an important role in the natural environment. They act as a shelter for coastal area from erosion that is cause by waves. They also act as a breeding ground for aquatic organisms. Mangroves and estuarine area also play an important role in conserving rare and endangered wildlife species such as proboscis monkeys and migrating birds by acting as a sanctuary for these animals. Currently the total mangrove forest area in Malaysia covers 566,856 ha.

Mangrove area is also economically important because it provided a suitable environment for economic activities such as aquaculture, tourisms and logging of mangrove trees. A study by Bennett and Reynolds (1993) on the economic value of the Sarawak Mangroves Forest Reserve found that the mangroves there support marine fisheries worth US\$21.1 million p.a., timber products worth US\$123,217 p.a., tourism industry worth US\$3.7 million p.a. and providing up to 3000 jobs.

Due to the various functions and services provided by the mangrove area, any factors that influence the mangrove ecosystems and its productivity is of a great interest since it will have a broad impact affecting not just the natural environment and organisms that lives in the mangrove area but also humans.

One of the threats to the mangrove and estuarine ecosystems is the enrichment of metals in the surface sediment. This is because mangrove and estuarine sediment are also known to be a good sink for metal pollution (Tam and Yao 1998; Kamaruzzaman et al. 2004; Praveena et al. 2008, 2010). Consequently, mangrove and estuarine areas are vulnerable to the influence of anthropogenic activities. Studies by several researchers have found that accumulations of metals are prone to occur in mangrove areas that are located near sources of metal pollutants, such as industrial areas (Pekey 2006; Praveena et al. 2008, 2010). However, generally, various anthropogenic activities (e.g. water drainage, discharge of wastewater from urban and industrial areas) and natural processes (e.g. soil, coastal and seafloor erosion, biological activities, riverine and atmospheric input) influence the distribution of metals in aquatic environments (Ip et al. 2007; Leivouri 1998; Christophoridis et al. 2009).

The ability of the sediment of wetlands to treat wastewater containing high metal content has been extensively studied by Sobolewski (1999), Carleton et al. (2001), Mays and Edwards (2001), Walker and Hurl (2002), and Nelson and Gladden (2008). They concluded that most waterborne metals are strongly retained by the wetland sediment. Among the factors that influence metal retention in sediment are grain size distribution, the presence of iron and manganese oxides, particulate organic matter and types of minerals (Horowitz 1985; Horowitz and Elrick 1987).

The main concern with metal enrichment in mangrove and estuarine sediment is due to the fact that they are elements and therefore cannot be broken down, unlike organic pollutants that can degrade to carbon dioxide and water (Gupta et al. 2001; Khan 2004). As the metal accumulates in the environment, it will create a condition that is detrimental to the ecosystems. This is because metals can affect aquatic organisms and mangrove plants as toxic substance in water and sediment, or as a toxicant in the food chain (Sorensen 1991; Rainbow 1996; Yim and Tam 1999; Maret et al., 2003).

However, the total concentration of metal is not a good indicator of its bioavailability, toxicity and transportability. It has been concluded that metal toxicity, bioavailability, bioactivity, transport in the organism, bio-geological distribution and transportation and its impact to organisms or environment depends on its speciation or form (Sanz-Medel 1998). Because of this, it is imperative to separate, quantify and identify the species of metal available in the environment (Sanz-Medel 1998).

1.2 The Importance of Sediment Quality Monitoring

Sediment quality is an important aspect of the aquatic ecosystem as sediment can mediate their pollution of nutrient uptake, storage, release and transfer between environmental compartments (Ongley 1996). Due to this, not only sediment can be influenced the pollution in the overlying waters, sediment can also influence the quality of the overlying waters. Apart from that, sediment quality will also have an impact on the benthic community (e.g. worms, mollusks and amphipods) and organisms that source food from the sediment. Therefore, a change in sediment quality may signal a change in the aquatic ecosystem and the pollution inputs from point or non-point sources.

Sediment quality monitoring is normally conducted with the aims to assess the risk associated with the sediment or/and to identify the spatial distribution of pollution in the sediment of an area. Spatial monitoring of sediment quality can indicate the status of contamination in a horizontal spread of an area and possibly enabling the identification of the pollution sources (Brils 2008).

Monitoring of sediment quality near ports or other rapidly developed coastal area is important as activities such as dredging for development (e.g. airport and port development, land formation, infrastructure improvements) or maintaining of waterway can cause the release of pollutant that had accumulated in the sediment (Jones et al. 1979; Chapman et al. 1999). Information regarding sediment quality in these areas will enable the relevant authorities to make an informed decision on method that can be used to minimize the impact of pollution remobilization due to dredging and also the best means to dispose the dredge sediment (Chapman et al. 1999).

1.3 Problem Statement

Port Klang plays an important part in the economic development of Malaysia. This is due to it being one of the main gateways for trade between Malaysia and the rest of the world.

Apart from having economic importance, the estuarine and mangrove areas around Port Klang are well known among the locals as fishing hotspots. This is where the coastal fishermen and the *Orang Asli* (indigenous people) from the nearby settlement catch fish, shrimp, crabs and collect mollusk for their own consumption or trade purposes. The mangrove areas around Port Klang also serve as resting and feeding place for migratory birds. However, the socioeconomic benefit and ecological integrity of the mangrove and estuarine area in Port Klang can be compromised by metal pollution from point and nonpoint sources located near the area or further upstream within the Klang and the Langat River catchment areas.

Port related activities such as the operation of ferry terminal, marina and ship maintenance (Turner 2010, Turner et al. 2009; Schiff et al. 2004), as well as industrial activities such as manufacturing, smelting and metal related industries can contribute to metal pollution (Kabir et al. 2012). Apart from that, the discharge of greywater from residential areas (Eriksson and Donner, 2009), wastewater from animal farm, sewage, illegal dumping of domestic and construction waste and surface runoff from the urban, industrial and agricultural area within the catchment area of both Klang and Langat River can contribute to the metal pollution in the Port Klang area. Therefore, there is a need for constant monitoring so that any threat to this area can be detected and address before the condition escalate further.

The usage of sediment and biota to determine metal pollution in the aquatic environment in recent literature were determined using the Needs, Approach, Benefits and Challenges (NABC) analysis (Table 1.1). This analysis facilitates in the assessment of the conditioned that occurred in previous literatures and helped this study to overcome the problems or limitation faced by other researchers.

A lot of previous studies paid more attention to the metal pollution in the Klang River (Naji and Ismail 2012, 2011; Naji et al. 2010), the Langat River (Lim et al. 2011; Shafie et al. 2013) and the area within the South Klang Strait (Tavakoly Sany et al. 2013b, 2013c, 2012a, 2012b) in Port Klang. However, not many studies were focused on the area within Lumut Strait where discharge from both the Klang and the Langat River converge. Due to its function as a source of fisheries for the local communities and significant role in ecology and tourism (eg. recreational fishing, bird watching), it is important to study this area as metal enrichment will negatively influence its function, productivity and the lives of peoples that depends on it. The Lumut Strait and its surrounding area have the highest possibilities of experiencing high metal accumulation due to inputs from the two rivers and from activities near and within the area. The presence of mangroves and weak water currents in the area will naturally encourage sedimentation of suspended particle, which may contain high metal content. The weak current also meant increased contact time for the sediment to adsorb metal ion from the overlying water. Therefore, the sediment in Lumut Strait has a higher risk of experiencing metal enrichment compared to the other area.

Table 1.1 The Needs, Approach, Benefits and Challenges (NABC) outputs for recent literatures on metal pollution in the aquatic environment.

	Outputs
Needs	• There are inadequate information on metal pollution, especially on mercury and methylmercury in the Malaysian environment.
	• Information on metal pollution and its distribution in the sediment is needed to assess the degree of contamination or toxicity levels and its effect on both marine organisms and human population.
Approach	• Analize the sediment for metals concentration as it will reflect

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	the historical variation and effects of anthropogenic and
	lithogenic inputs into the marine environment.
	• The use of various organisms (e.g. fish, gastropod, insect and etc.) as bioindicator for metal pollution.
Benefits	• Address the lack of information on metal distribution in the Port
	Klang area (especially for the Lumut Strait) and also on mercury speciation.
	• Help the authorities to better understand the current degrees of metal pollution so that appropriate action can be taken.
	• Information on metals contamination in sediment helps in the assessment of potential environmental effect associated with
	bio-toxicity, environmental stability and bioaccumulation in the food chain.
Challenges	• There is an information gap on the viability of certain organism
C	as a bioindicator for metals especially for Co, Hg, MeHg and Mn.
	• The studies done were inadequate to represent the true
	distribution of metals in the studied area.

Based on previous studies, the sediment in the Port Klang area was reported to have a considerable amount of Hg (Law 1987; Sakamoto et al., 2004; Tavakoly Sany et al., 2013c, 2012a,b), however, most of these studies only cover the South Klang Strait while no comprehensive study on mercury distribution were done in the Lumut Strait. The studies regarding Hg in Port Klang were mostly done based on the total Hg, but few were on the mercury speciation especially on methylmercury (MeHg) which, is the most common organic mercury in the environment (UNEP 2002) and has a higher bioavailability and toxicity compared to elemental mercury (UNEP 2002; Gochfeld 2003). This shows a gap in the information regarding the concentration of MeHg in the sediment and in the organism living in the area.

Therefore, this study will address the gap in information regarding metals concentration in the Lumut Strait by identifying the distribution of Cd, Co, Cu, Cr, Hg, Mn, Ni, Pb, Zn and MeHg concentration in the sediment and mollusk (*Nerita lineata*) in the area. The gastropods species *N. lineata* was chosen for this study due to its habitat which is between the tide mark on the muddy shores of mangrove swamp, river and sea such as found in the study area and also for its feeding behavior where it grazed on micro algae growing on sediments, rocks, shells, roots and trunks of mangrove plants or other larger plants (Hughes 1986; Cheng 2008). Therefore, there is a higher possibility that the concentration of metals accumulated in the sediment will be reflected by the concentration of metals in the soft tissue of *N. lineata*. Other than that, *N. lineata* was also chosen due to its importance in the marine food chain and as a human food source especially those living near the study area.

1.4 Objectives

In general, the aims of this study were to understand the distribution of metals in the Port Klang mangrove and coastal intertidal surface sediment and its influence on metals concentration in the soft tissue of *Nerita lineata*. Apart from that, the speciation

of mercury in both sediment and *N. lineata* soft tissue was also investigated. The specific research objectives are as follows:

- 1. To determine the distribution and enrichment of metals in the sediments of Port Klang.
- 2. To determine factors that affects metal retention in the sediment and the correlation between metals in sediment with metals in the soft tissue of *Nerita lineata*.
- 3. To elucidate the speciation of mercury in sediment and soft tissue of *Nerita lineata*.
- 4. To elucidate the relationship of mercury and methylmercury accumulation in sediment and *Nerita lineata*.

1.5 Scopes of Study

Essentialy this study covers the following scopes:

- 1. This study focused on the distribution of elements such as Cd, Co, Cr, Cu, Fe, Hg, Mn, Ni, Pb and Zn in the surface sediment and the soft tissue of *Nerita lineata*. Mercury speciation in both sediment and *N. lineata* soft tissue was also conducted. The physico-chemical parameters of water and sediment were measured to enable better understanding of the dynamics of metal distribution and Hg speciation in the aquatic environment of the mangrove and coastal area.
- 2. Various indices (i.e. enrichment factor, geoaccumulation index, pollution load index, contamination factor, modified degree of contamination, risk factor, potential risk index, marine sediment pollution index, mean probable effect level quotient and adverse effect index) were calculated to assess the enrichment, pollution and toxicity of metals concentration in the sediment. These indices also help to determine the threat level to biota and the ecology in general. The application of several statistical techniques aids in the determination of possible sources for metals accumulating in the study area and the factors affecting its retention in the sediment.
- 3. This study also looks into the relationship between metals concentration in the sediment with metals in the soft tissue of *N. lineata*. This helps to determine the most suitable elements that can be monitored using *N. lineata*'s soft tissue. Other than that, factors that influence the concentration and accumulation of metals and Hg speciation in *N. lineata* can also be known.

1.6 Thesis Outline

This thesis is divided into five (5) chapters that consist of an Introduction (Chapter 1), Literature Review (Chapter 2), Materials and Methods (Chapter 3), Results and Discussion (Chapter 4) and Conclusions and Recommendations. Descriptions of each chapter are as follows:

- 1. Chapter 1 of this thesis gives a brief introduction on the importance of mangrove, estuarine and coastal areas in terms of economic value, ecological function and the role they play as a sink of pollutants, especially metals. This chapter also explains the need for this study and its objectives.
- 2. The second chapter reviews the relevant literature related to this study. This includes background information on metals and the biota used in this study.

Factors influencing metals transport and accumulation in the sediment were also elaborated. Previous studies on metal accumulation in sediment and biota (especially *Nerita lineata*) were also reviewed.

- 3. The third chapter covers the materials and methods used in this study. The research methodology was based on the analysis of water, sediment and biota (i.e. *Nerita lineata*) collected from the study area in Port Klang. This chapter is divided into several parts such as site description, sample collection, sample analysis, data analysis and data interpretation. Methods for calculating various indices to assess sediment and biota metal concentration were also explained.
- 4. Chapter 4 covers the results and discussion for all findings after samples were analysed. Some parts of the findings in this chapter were published in journals, book chapter and conference proceeding (see the list of publications at the end of this thesis). In general, this chapter provides an overview on the recent status of sediment in the study area in regards to metal concentration. The metals distribution, potential source and factors influencing its retention in sediment were elaborated further. The risk posed by metals in the sediment and the speciation of Hg was also discussed. Other than that, the relationships between metals in sediment with metals in the soft tissue of *N. lineata* were further explored.
- 5. The fifth chapter summarize and concludes the findings of this study, which addressed all objectives stated in the first chapter. Some recommendations that may be useful for future research and gives some ideas to the authority on the importance of continuous monitoring were also made. This is to ensure that the various functions and services provided by the mangrove and coastal area are preserved.

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APPENDICES

Appendix A1

Values of K for use in equation for computing diameter of particle in hydrometer analysis.

Temperature	Specific Gravity of Soil Particles								
(°C)	2.45	2.50	2.55	2.60	2.65	2.70	2.75	2.80	2.85
16	0.01510	0.01505	0.01481	0.01457	0.01435	0.01414	0.01394	0.01374	0.01356
17	0.01511	0.01486	0.01462	0.01439	0.01417	0.01396	0.01376	0.01356	0.01338
18	0.01492	0.01467	0.01443	0.01421	0.01399	0.01378	0.01359	0.01339	0.01321
19	0.01474	0.01449	0.01425	0.01403	0.01382	0.01361	0.01342	0.01323	0.01305
20	0.01456	0.01431	0.01408	0.01386	0.01365	0.01344	0.01325	0.01307	0.01289
21	0.01438	0.01414	0.01391	0.01369	0.01348	0.01328	0.01309	0.01291	0.01273
22	0.01421	0.01397	0.01374	0.01353	0.01332	0.01312	0.01294	0.01276	0.01258
23	0.01404	0.01381	0.01358	0.01337	0.01317	0.01297	0.01279	0.01261	0.01243
24	0.01388	0.01365	0.01342	0.01321	0.01301	0.01282	0.01264	0.01246	0.01229
25	0.01372	0.01349	0.01327	0.01306	0.01286	0.01267	0.01249	0.01232	0.01215
26	0.01357	0.01334	0.01312	0.01291	0.01272	0.01253	0.01235	0.01218	0.01201
27	0.01342	0.01319	0.01397	0.01277	0.01258	0.01239	0.01221	0.01204	0.01188
28	0.01327	0.01304	0.01283	0.01264	0.01244	0.01255	0.01208	0.01191	0.01175
29	0.01312	0.01290	0.01269	0.01249	0.01230	0.01212	0.01195	0.01178	0.01162
30	0.01298	0.01276	0.01256	0.01236	0.01217	0.01199	0.01182	0.01165	0.01149

Hydrometer 152H				
Actual	Effective	Actual	Effective	
Hydrometer	Depth, L	Hydrometer	Depth, L	
Reading	(cm)	Reading	(cm)	
0	16.3	31	11.2	
1	16.1	32	11.1	
2	16.0	33	10.9	
3	15.8	34	10.7	
4	15.6	35	10.6	
5	15.5	36	10.4	
6	15.3	37	10.2	
7	15.2	38	10.1	
8	15.0	- 39	9.9	
9	14.8	40	9.7	
10	14.7	= 41	9.6	
11	14.5	42	9.4	
12	14.3	43	9.2	
13	14.2	44	9.1	
14	14.0	45	8.9	
15	13.8	46	8.8	
16	13.7	47	8.6	
17	13.5	48	8.4	
18	13.3	49	8.3	
19	13.2	50	8.1	
20	13.0	51	7.9	
21	12.9	52	7. 8	
22	12.7	53	7.6	
23	12.5	54	7.4	
24	12.4	55	7.3	
25	12.2	56	7.1	
26	12.0	57	7.0	
27	11.9	58	6.8	
28	11.7	59	6.6	
29	11.5	60	6.5	
30	11.4			

Values of effective depth based on hydrometer and sedimentation cylinder of specific size

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Temperature Correction Factors (C_T).

Temperature, °C	Factor C _T
15	1.10
16	-0.90
17	-0.70
18	-0.50
19	-0.30
20	0.00
21	+0.20
22	+0.40
23	+0.70
24	+1.00
25	+1.30
26	+1.65
27	+2.00
28	+2.50
29	+3.05
30	+3.80

 \mathbf{G}

Specific Gravity (g/cm ³)	Correction Factor (a)
2.95	0.94
2.90	0.95
2.85	0.96
2.80	0.97
2.75	0.98
2.70	0.99
2.65	1.00
2.60	1.01
2.55	1.02
2.50	1.03
2.45	1.05

C

Values of Correction Factor (a) for different specific gravities of soil particles.



Appendix B

Water parameters.

G

Station	pН	Salinity (ppt)	Eh (mV)	EC (mS/cm)	TDS (mg/L)
PK 1	7.40±0.00	25.80±0.08	-9.17±0.21	40.40±0.14	19795.00±70.60
PK 2	7.39 ± 0.00	23.07±0.09	-8.63±0.12	36.47±0.09	17875.33±60.27
PK 3	7.42 ± 0.00	24.03±0.05	-10.43±0.05	37.93±0.12	18587.67±44.73
PK 4	7.39 ± 0.00	25.73±0.05	-8.77±0.26	40.27±0.09	19733.67±39.14
PK 5	7.01 ± 0.00	14.93±0.05	14.17±0.09	24.58±0.11	12043.67±54.93
PK 6	7.42 ± 0.00	23.06±0.09	-10.42 ± 0.25	36.44±0.10	16110.83±109.16
PK 7	7.01 ± 0.00	10.60 ± 0.00	14.03±0.12	17.97±0.02	8807.00±9.20
PK 8	6.89 ± 0.01	1.80 ± 0.00	20.83±0.40	3.38 ± 0.02	1654.33±7.13
PK 9	6.93±0.01	0.20 ± 0.00	18.50±0.78	0.49 ± 0.00	242.33±0.47
PK 10	6.87 ± 0.00	0.20 ± 0.00	22.07±0.05	0.51±0.00	250.33±0.47
PK 11	7.66±0.00	26.17±0.05	-24.80±0.22	40.87±0.05	20433.50±23.57
PK 12	7.04±0.00	23.53±0.05	12.17±0.17	37.13±0.05	18196.00±36.48
PK 13	7.34 ± 0.00	24.87±0.09	-5.67±0.12	39.03±0.19	19129.67±92.16
PK 14	7.37 ± 0.00	23.17±0.09	-6.77±0.12	36.63±0.12	17953.67±66.76
PK 15	7.35 ± 0.01	24.73±0.05	-6.10±0.43	38.87±0.09	19044.33±49.98
PK 16	7.23 ± 0.00	23.97±0.09	0.93±0.12	37.77±0.17	185 <mark>19</mark> .67±81.79
PK 17	7.48 ± 0.00	24.60±0.08	-14.13±0.17	38.73±0.12	18964.00±58.88
PK 18	7.36 ± 0.00	22.40±0.22	-7.30±0.08	35.53±0.26	17418.67±125.85
PK 19	7.15 ± 0.00	17.03±0.05	5.53±0.19	27.76±0.09	13602.00±46.68
PK 20	7.03 ± 0.00	12.23±0.05	12.87±0.09	20.51±0.08	10048.67±41.96
PK 21	7.54 ± 0.00	25.40±0.16	-18.30±0.22	39.90±0.14	19550.00±68.44
PK 22	7.13 ± 0.01	17.83±0.05	6.77±0.31	28.94±0.10	14182.33±49.29
PK 23	7.29 ± 0.00	23.43±0.05	-2.90±0.14	37.00±0.08	18127.67±22.31
PK 24	7.34 ± 0.00	21.23±0.05	-5.33±0.09	33.87±0.09	16595.67±50.45
PK 25	7.29 ± 0.00	22.20±0.22	-2.60±0.08	35.30±0.36	17292.33±165.18
PK 26	7.39 ± 0.00	19.23±0.05	-8.47±0.21	31.00±0.08	15190.67±34.62
PK 27	7.55 ± 0.00	26.63±0.12	-17.97±0.12	41.53±0.12	20765.00±62.36
PK 28	7.28 ± 0.00	24.03±0.12	-2.37±0.12	37.87±0.17	18871.33±423.41
PK 29	7.33 ± 0.01	23.30±0.10	-4.83±0.41	36.80±0.12	17031.00±212.53
PK 30	7.43 ± 0.00	21.03+0.12	-11.00+0.14	33.57+0.17	16462.33+86.10

Appendix C

Sediment parameters.

Station	pН	Salinity (ppt)	Eh (mV)	EC (mS/cm)	LOI (%)
PK 1	7.31 ± 0.00	14.03 ± 0.12	-11.47±0.17	23.30±0.22	8.0 ± 0.1
PK 2	5.69 ± 0.00	13.20 ± 0.08	83.50±0.00	22.01±0.12	6.9±0.1
PK 3	6.78 ± 0.00	13.13±0.12	19.70±0.08	21.90±0.20	7.6±0.0
PK 4	3.97 ± 0.00	12.80 ± 0.08	184.30 ± 0.08	21.43±0.14	7.4 ± 0.1
PK 5	3.44 ± 0.00	10.73 ± 0.05	215.13±0.26	18.26±0.13	4.4 ± 0.0
PK 6	5.67 ± 0.00	6.97 ± 0.05	84.87 ± 0.05	12.22±0.03	6.5±0.0
PK 7	7.10 ± 0.00	12.67 ± 0.05	0.73±0.05	21.22±0.07	5.8±0.0
PK 8	6.09 ± 0.00	9.53±0.09	60.40 ± 0.00	16.34±0.16	5.1±0.2
PK 9	4.50 ± 0.00	10.03 ± 0.05	153.17±0.17	17.09±0.09	7.6±0.0
PK 10	5.12±0.00	14.10±0.22	116.93±0.05	23.31±0.27	10.3±0.0
PK 11	7.68 ± 0.00	12.37±0.09	-33.17±0.12	20.74±0.15	4.6±0.2
PK 12	7.20±0.00	15.90±0.78	-4.97±0.05	26.13±1.21	4.6±0.1
PK 13	4.00 ± 0.00	10.73±0.12	182.53±0.05	18.20±0.18	3.5±0.1
PK 14	7.72±0.00	8.07±0.09	-35.67±0.26	14.02±0.12	3.0 <u>±</u> 0.0
PK 15	7.12±0.00	7.40 ± 0.00	-0.10±0.00	12.97±0.02	3.2±0.0
PK 16	3.83 ± 0.00	11.73±0.17	192.47±0.29	19.80±0.25	4.6±0.0
PK 17	7.21±0.01	10.33±0.05	-5.57±0.37	17.62±0.06	4.2±0.0
PK 18	6.98 ± 0.00	14.7 <mark>0±0</mark> .00	8.10±0.08	24.36±0.02	5.2±0.1
PK 19	7.01±0.00	6.93±0.12	6.07±0.05	12.19±0.14	3.8±0.0
PK 20	3.49 ± 0.00	7.53±0.05	212.23±0.17	13.19±0.10	2.3±0.0
PK 21	4.28 ± 0.01	14.70±0.00	166.20±0.64	24.32±0.02	7.2 ± 0.5
PK 22	5.26 ± 0.00	6.30±0.08	108.83±0.05	11.13±0.11	3.6±0.1
PK 23	7.63 ± 0.00	16.40±0.08	-29.90±0.08	26.88±0.09	8.2±0.2
PK 24	7.46±0.01	9.33±0.17	-20.07±0.46	16.01±0.27	6.2±0.3
PK 25	6.30 ± 0.00	13. <mark>97±0.09</mark>	48.10±0.22	23.22±0.19	7.7±0.1
PK 26	5.98 ± 0.00	13.50 ± 0.08	66.50±0.22	22.51±0.09	8.7±0.4
PK 27	5.17±0.09	10.87±0.05	114.30±5.02	18.44±0.04	6.7±0.1
PK 28	6.90±0.01	13.80±0.00	12.17±0.74	22.94±0.04	8.0±0.2
PK 29	6.15±0.01	10.33±0.09	56.63±0.45	17.59±0.14	9.7±0.2
PK 30	7.65 ± 0.00	19.13±0.12	-31.20±0.00	30.90±0.16	5.5 ± 0.1

BIODATA OF STUDENT

Hazzeman Haris received this Bachelor degree in International Tropical Forestry from Universiti Malaysia Sabah (UMS) in 2004. He then furthers his study at Universiti Sains Malaysia (USM) and graduated with a Master of Science in 2010. Currently he is pursueing his doctorate in Universiti Putra Malaysia (UPM). His doctorate studies involves the evaluation of metal pollution in sediment of Port Klang coastal and mangrove area and its accumulation in *Nerita lineata* with special attention given to mercury and its prevalent organic form which is methylmercury. This study was funded by the Research University Grants Scheme (RUGS) number 91895. Several papers from this study have been published. He also took part in several conferences as a speaker.



LIST OF PUBLICATIONS

This thesis is partly based on the following publications:

Journals

- Haris H, Aris A (2013) The geoaccumulation index and enrichment factor of mercury in mangrove sediment of Port Klang, Selangor, Malaysia. Arabian Journal of Geosciences 6(11):4119-4128 doi:10.1007/s12517-012-0674-7
- Haris H, Aris A (2015) Distribution of metals and quality of intertidal surface sediment near commercial ports and estuaries of urbanized rivers in Port Klang, Malaysia. Environmental Earth Sciences 73(11):7205-7218 doi:10.1007/s12665-014-3900-7

Chapter in book

Haris H, Aris A (2014) Mercury Distribution in Port Klang Mangrove and Estuarine Sediment. In: Aris AZ, Tengku Ismail TH, Harun R, Abdullah AM, Ishak MY (eds) From Sources to Solution. Springer Singapore, pp 187-190. doi:10.1007/978-981-4560-70-2_35

Proceedings

Haris H, Aris AZ (2011) The geo-accumulation index and enrichment factor of mercury in mangrove sediment of Port Klang, Selangor, Malaysia. Paper presented at the 24th Malaysian Symposium of Analytical Sciences (SKAM 24), Langkawi, Malaysia, 21-23 November 2011