COMPARISON OF HEAVY METAL CONCENTRATIONS IN SEDIMENTS, LITTER AND PLANTS FROM THREE DIFFERENT MANGROVE HABITATS IN SELANGOR

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ABSTRACT

In recent years, mangroves have become a massive pollution sink which receives a number of pollutants from various sources. An experiment was conducted to determine the heavy metal concentrations in sediments, litter and plants at different mangrove habitats at the Klang Islands. Three different land use types of mangrove ecosystems were selected: natural mangrove forest (Pulau Kelang) and two types of disturbed mangrove habitats due to agriculture (Pulau Carey) and industry (Telok Gong). This study focuses on correlating the concentrations of heavy metals in sediment between tidal gradients and identifying pollution level using the geoaccumulation index. The heavy metal concentrations were also compared between different land use types. Other aims of this study were to investigate the ability of plants to absorb heavy metals in sediments in terms of an accumulation coefficient, compare the accumulation and cycle of heavy metals in mangrove sediments, fresh tissues, and litter tissues, as well as correlate the concentrations of heavy metals between sediments and different tissues of mangrove plants. The results showed that the concentrations of Pb, Hg, Cd and As of the sediments were in the order of Pb > As > Cd > Hg. At the same time, the concentrations of these heavy metals in the sediments showed some correlation with the tidal gradient but this was not significant. According to the geoaccumulation Index (I_{geo}), the four heavy metals in all samplings were uncontaminated to moderately contaminated (Class 1). In addition, the concentrations of heavy metals were different between land use types, whereby Telok Gong mangrove had significantly higher heavy metal concentrations than Pulau Carey mangrove. In terms of heavy metals in sediments, the accumulation coefficient values for the plant branch were Hg> Pb> Cd> As, while those for the leaves were Hg > Pb > As > Cd. The concentrations of Pb, Hg, Cd and As in mangroves were as follows: sediments > litter (branch and leaves) > fresh specimens (branch and leaves). Finally, this study showed that the concentrations of Pb, Hg and Cd

were higher in branch as compared to leaves, while that of As was similar between branch and leaves.

ABSTRAK

Kebelakangan ini, hutan paya bakau telah menjadi sinki pencemaran dari pelbagai sumber. Eksperimen ini dijalankan untuk menentukan kepekatan logam berat dalam mendapan, sampah sarap, dan tumbuh-tumbuhan di habitat paya bakau yang berbeza di Kepulauan Klang. Tiga jenis penggunaan tanah ekosistem paya bakau telah dipilih: hutan bakau semula jadi (Pulau Klang) dan dua jenis habitat paya bakau terganggu oleh aktiviti pertanian (Pulau Carey) dan industri (Telok Gong). Kajian ini bertujuan untuk mengkaji perhubungan antara kepekatan logam berat dalam mendapan merentasi kecerunan pasang surut, mengenal pasti tahap pencemaran melalui indeks geoakumulasi dan membandingkan kepekatan logam berat di tempat penggunaan tanah yang berbeza. Kajian ini juga bertujuan untuk mengetahui keupayaan tumbuh-tumbuhan untuk menyerap logam berat dalam mendapan melalui pekali pengumpulan, membandingkan pengumpulan dan kitaran logam dalam mendapan di paya bakau, tumbuh-tumbuhan, dan sesampah, serta untuk menghubungkaitkan logam berat dalam mendapan dengan bahagian tumbuhan yang berbeza. Dapatan kajian menunjukkan bahawa kepekatan Pb, Hg, Cd dan As dalam mendapan adalah dalam urutan Pb > As > Cd > Hg. Pada masa yang sama, kepekatan logam berat dalam mendapan boleh dikaitkan dengan kecerunan pasang surut, biarpun tidak ketara. Menurut indeks geoakumulasi, empat logam berat dalam semua sampel dari paya bakau tersebut berjulat dari tidak tercemar ke sederhana tercemar (Kelas 1). Di samping itu, kepekatan logam berat berbeza-beza antara jenis penggunaan tanah; paya bakau di Telok Gong mempunyai kandungan logam berat yang lebih tinggi daripada paya bakau di Pulau Carey. Nilai pekali pengumpulan logam berat di dahan pokok untuk adalah Hg > Pb > Cd > As, manakala untuk daun, nilai tersebut adalah seperti berikut: Hg > Pb > As > Cd. Kepekatan Pb, Hg, Cd dan As adalah seperti berikut: mendapan> sesampah (ranting dan daun)> daun segar (ranting dan daun). Akhir sekali, kajian ini menunjukan bahawa Pb,

Hg dan Cd banyak didapati di bahagian dahan pokok, manakala As menunjukkan kepekatan yang sama di dahan dan daun.

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LIST OF SYMBOLS AND ABBREVIATIONS

AC	:	Accumulation Coefficient
Adj.	:	Adjust
Ag	:	Silver
Al	:	Aluminium
ANOVA	:	Analysis of Variance
As	:	Arsenic
ASEAN	:	Association of Southeast Asian Nations
Cd	:	Cadmium
Co	:	Cobalt
Cr	:	Chromium
Cu	:	Copper
DWF	:	Dry Weight Factor
EF	:	Enrichment Factor
Fe	:	Ferrum
FIMS	:	Flow Injection Mercury System
g/cm^3	:	Gram Per Cubic Centimeter
GLM	:	Generalized Linear Model
Ha	:	Hectare
Hg	:	Mercury
IARC	:	International Agency for Research on Cancer
ICP-MS	:	Inductively Coupled Plasma Mass Spectrometer
ICP-OES	:	Inductively Coupled Plasma Optical Emission Spectrometer
I_{geo}	:	Geoaccumulation Index

ISQGs	:	Interim Sediment Quality Guidelines
kg	:	Kilogram
KIMFR	:	Klang Islands Mangrove Forest Reserve
mg/kg	:	Milligram Per Kilogram
ml	:	Milliliter
Mn	:	Manganese
Ni	:	Nickel
Pb	:	Lead
PRFs	:	Permanent Reserved Forests
RI	:	Potential Ecological Risk Index
Sig.	:	Significant
SPSS	:	Statistic Package of Social Science
Std.	:	Standard
t/ha	:	Tonne Per Hectare
ug/g	:	Microgram Per Gram
ug/kg	:	Microgram Per Kilogram
USEPA	:	United States Environmental Protection Agency
$wt.m^{-2}$	:	Weight Per Square Metre
yr	:	Year
Zn	:	Zinc

CHAPTER 1: INTRODUCTION

1.1 Mangroves

Mangroves are predominantly found at intertidal zones and are mainly made up of plants that occur worldwide in the tropical and subtropical latitudes between the land and sea (Nagelkerken et al., 2008). Animals and plants living in mangroves are able to adapt well to high salinity, temperature, wind speed, anaerobic soils and tidal disturbances (Sandilyan & Kathiresan, 2012). Mangrove plants have unique adaptations such as breathing and interlocking aerial root systems, salt regulation, viviparous mode of reproduction, and nutrient retention (Kathiresan & Bingham, 2001). Mangroves provide a perfect habitat for the life cycles of animals, such as mammals, birds, insects and reptiles. Underwater animals and plants, or epibionts (such as sponges and algae) have a tendency to overgrow near the mangrove roots. The soft substratum space between plant roots provides shelter and food for prawns, crabs and fishes (Nagelkerken et al., 2008).

Mangrove forests are important for human and coastal communities. The mangrove ecosystem can be evaluated by ecological sustainability, economic prosperity and environmental security (Sandilyan & Kathiresan, 2012). Mangrove forests are important in protecting the shore from natural disasters. For example, they can mitigate the effects of tidal waves and typhoons (Kathiresan & Rajendran, 2005). Mangroves can also stabilize the coastal systems as healthy mangroves are a repository of heavy metals and other pollutants which enter the system from industrial effluents and runoffs (Vannucci, 2001). Moreover, some mangrove plants have medicinal values: some are used to treat diarrhea (*Bruguiera gymnorrhiza)* as well as prevent viral and bacterial infections (*Sesuvium portulacastrum*) (Sandilyan & Kathiresan, 2012).

The mangrove ecosystem in Malaysia is a vital resource for the estuarine, coastal and riverine communities. These communities have either been living at the edges of inland mangroves or within the mangrove swamps for livehood. The aquatic resources (fishes, gastropods) in mangrove swamps are very important to fishermen. Also, the mangrove forests provide wood which can act as fuels and other building materials for local communities (Latiff & Faridah - Hanum, 2014). Jusoff (2009) reported that mangroves are the major type of wetlands in Peninsular Malaysia (Kedah, Perak, Selangor and Johor, Sabah, as well as northern and southwestern Sarawak). Most of the mangroves in Selangor (Kelang islands) and Johore (Pulau Kukup) are near the shores of islands, while other small areas of mangroves are located at rocky shores. At the East Coast, mangroves predominantly occur along the Kemaman River and Behar (Jusoff, 2009).

1.2 Status of mangroves

Mangroves are largely restricted to latitudes between 30°N and 30°S, and they occupy around 15.2 million hectares worldwide. They are mainly distributed in Asia (39%) and Africa (21%). The rest are present in North and Central America (15%), South America (12.6%), and Oceania (12.4%) (Forestry Economics and Policy Division, 2007). During 1980 - 2000s, approximately 35% of the global mangroves area has been lost. Meanwhile, the percentage of protected mangroves was only 6.9% (Sandilyan & Kathiresan, 2012). From the year 2000 to 2005, mangroves disappeared at a rate of 0.66% per year (Forestry Economics and Policy Division, 2007). Mangroves are the primary coastal community in Asia, and are predominantly distributed in the Malaysia - Indonesian region. Indonesia has 3,112,989 hectares of mangrove forests, followed by Malaysia with 505,386 hectares comprising of 104 mangrove species (Mazlan et al., 2005). In Peninsular Malaysia, 74 percent of mangrove forests have been gazetted as Permanent Reserved Forests (PRFs) in 2006. These included Matang in Perak, Kelang in Selangor, and Johor (Jusoff, 2009). The details of mangrove areas by state are shown in Table 1.1. Since the 1960s, the destruction and reclamation of mangroves in Selangor have been going on owing to unabated industrialization. A large of forest reserves (around 62%) has been lost in light of vast and massive development (Haliza, 2004) (Table 1.2).

Table 1.1: Current area (ha) of mangrove forests (stateland and PRFs) in Peninsular Malaysia in 2006 (Jusoff, 2009)

Sate	Stateland mangroves	Mangrove Forest Reserves	Total
Perak	1,885	40,466	42,351
Johor	13,561	16,127	29,688
Selangor	4,650	14,897	19,547
Kedah	1,916	6,202	8,118
Pahang	1,813	2,387	4,200
Terengganu	692	1,295	1,987
Penang	494	376	870
Kelantan	744	-	744
N.Sembilan	-	204	204
Malacca	-	80	80
Perlis	13	-	13
Total	25,768	82,091	107,802

Table 1.2: Changes in the state and area of Permanent Mangrove Forest Reserves in Selangor beginning 1975 (Haliza, 2004)

Forest Reserve	1975	1984	1991	1999	Percentage loss
Banjar Utara	2,535	268	268	268	89.400
Banjar Selatan	1,261	111	111	111	91.200
Kuala Bernam	4,799	3,004	2,915	2,915	39.200
Kapar	5,429	3,836	1,722	410	95.400
Pulau Klang	8,785	8,785	5,370	5,371	38.900
Pulau Che Mat Zin	1,450	1,450	1,450	1,450	0.000
Pulau Tonggok	13	13	13	13	0.000
Pulau Selat Kering	954	954	954	954	0.000
Pulau Selat Meriam	56	56	56	56	0.000
Pulau Selat Mahang	149	149	149	149	0.000
Pulau Pintu Gedong	807	807	807	807	0.000
Telok Gadong	2,389	1,056	1,056	800	66.500
Sepang Kecil	559	414	414	302	46.000
Kuala Sepang	245	244	244	244	0.400

1.3 Heavy metal pollution in mangroves

Mangroves, as primary producers, have a major role in the estuarine ecosystems whereby they provide a habitat for a variety of species. In recent years, the loss of mangroves has been very significant, more so owing to the fact that existing mangroves are affected by heavy metals (Nazli & Hashim, 2010). Mangroves have become massive pollution sinks which received many pollutants from different sources. The main origins of the heavy metals in the mangrove ecosystems are from manufacturing industrial wastes, agro - based industries, and urbanization. Moreover, the use of fertilizers and pesticides in agricultural activities is a normal practice in Malaysia (Hashim & Hughes, 2010). The purification of fertilizers usually cannot completely remove impurities, including heavy metals (Zarcinas et al., 2004). Heavy metals, which are toxic compounds, have been reported to be present in high concentrations in mangroves due to the immobilization of heavy metals by the soil and plants. Some studies illustrated that most of the Asian mangroves have accumulated heavy metals primarily due to urban and coastal developments. For example, around 37,000 tons of industrial waste (which predominantly contained lead and mercury resulting from tanning activities) was dumped into the coast of Karachi (Sandilyan & Kathiresan, 2014).

1.4 Problem statement

Mangroves in Peninsular Malaysia are now facing an increasing threat of heavy metal pollution. The location of the mangrove forest and the type of land use in the surrounding area influences the variation of heavy metal deposits in mangrove sediments (Nazli & Hashim, 2010). In a plant - sediment system, strong adsorption and heavy metal fixation by the soil can lead to residual accumulation in soil, resulting in over-absorption of heavy metals in growing plants (Lian et al., 1999). These plants' litter are bad for the health of humans and intertidal zone animals. Mangroves in Selangor occupy the Straits of Malacca, which is an important shipping lane in the world. As the area is important for industrial and agriculture activities, a study on heavy metal concentrations in different land use types have some scientific significance. A good understanding of the current mangrove ecosystem pollution status is important to the Malaysian seafood industry, apart from determining public health as well as achieving sustainable management of mangrove ecosystems. This experiment was conducted on natural and disturbed mangrove habitats in the Klang Islands. At least 3 different land use types were present in this area: natural mangrove (Pulau Kelang), agricultural mangrove (Pulau Carey) and industrial mangrove (Telok Gong). This study reported the concentrations of Pb, Hg, Cd and As as they are toxic when present in humans and animals.

1.5 Objectives

(1) To determine and correlate the heavy metal concentrations (Pb, Hg, Cd and As) in sediments, litter and fresh tissues in three different mangrove habitats across the tidal gradient.

(2) To identify pollution levels via the geoaccumulation index, and determine the ability of plants to absorb heavy metals in sediments via the accumulation coefficient.

(3) To compare the accumulation and cycle of heavy metals in mangrove soils, fresh tissues and litter tissues.

CHAPTER 2: LITERATURE REVIEW

2.1 Definition of heavy metal

Heavy metals are commonly defined as substances having a specific density of more than 5 g/cm^3 (Järup, 2003). Heavy metals are persistent and non - biodegradable in the environment (Nazli & Hashim, 2010). Heavy metals are of two categories that are essential and non - essential elements. Toxicity depends on the properties and level of concentrations, which usually arise due to the formation of complex organic compounds (Rahimah, 2012). Heavy metals are released into the environment in many different ways, including (1) combustion and release into the air, as well as (2) runoff or release from transport to the surface water and groundwater to the soil. Human beings can be exposed to several agents (chemical, biological and physical) in the air, water, soil, or food. Heavy metals like lead, mercury, cadmium and arsenic pose a major threat to the human health (Järup, 2003).

2.2 Heavy metals occurrence and property

2.2.1 Lead (Pb)

According to USEPA (2015), Pb is a natural element that is found in the Earth's crust. Even though there are some applications of Pb, it can be toxic to humans and animals. Lead is found in the air, soil, water, and even indoors. Human are exposed to lead mainly from the usage of fossil fuels and leaded gasoline. Earlier, the use of lead in industrial activities and the domestic use of lead - based paint contributed to lead toxicity in humans. Lead and lead compounds have been used in products such as paints, ceramics, pipes and plumbing materials, solders, gasoline, batteries, ammunition, and cosmetics. Lead can enter the marine environment from industrial discharges, sewage effluents, highway runoffs, as well as from atmosphere (Rahimah, 2012).

2.2.2 Cadmium (Cd)

Cadmium is also a natural Earth element. Physically, cadmium is a soft and silver-colored white metal. Cadmium is rarely found as a free metal, rather it is usually combined with other elements (oxygen or sulfur) as cadmium oxide and cadmium sulfide. These compounds are solids that may dissolve in water but will not disappear from the environment. All soils and rocks, including coal and mineral fertilizers, contain cadmium (USEPA, 2015). Cadmium can be applied in consumer products and industries, including batteries, pigments, plastics, and metal coatings. Ore mines, metallurgical industries, and sewage sludge act as primarily anthropogenic sources of cadmium (Rahimah, 2012). Cadmium is released to the air from household wastes and metal mining. It can also dissolve in water during disposal of waste water. The use of fertilizers can contribute to the penetration of cadmium into the soil. In addition, spills and leaks from hazardous waste sites can also cause cadmium to enter the water or soil (USEPA, 2015).

2.2.3 Mercury (Hg)

Mercury occurs naturally in the environment in many forms. It can combine with other elements to form organic and inorganic compounds. All forms of mercury are considered to be poisonous. It is worth mentioning that one organic form of mercury, named methyl mercury, can build up in certain fish. Methyl mercury may be present in these fishes in rather low levels (USEPA, 2015). It is usually used in light bulbs, switches, thermometers, dental fillings, and batteries. Metallic mercury is also used in producing chlorine gas and sodium hydroxide. Methyl mercury is converted by microorganisms in the soil and water, and is also a bio-accumulating toxin (Martin & Griswold, 2009). Wood from the forests are widely used, the soil erosion flowing excessive deforestation causes inorganic and alkylated mercury to enter the aquatic system, so the concentrations of mercury in the fishes will be increased (Florea &

Büsselberg, 2006). Mercury can enter the atmosphere from the deposition of ore, the burning of fossil fuels or garbage, and the emission of gas and waste from the factories. Inorganic mercury might enter the water or soil from the rocks. This can also occur following the release of water from treatment plants or the disposal of wastes. Organic mercury may be released into the soil through the use of mercury- containing fungicides (USEPA, 2015).

2.2.4 Arsenic (As)

Arsenic is a naturally - occurring metalloid. Arsenic has the properties of both metal and non-mental, but due to its toxicity, it is usually referred to as a heavy metal. It can exist as organic and inorganic compounds by combining with other elements. Physically, arsenic has no color and odor. It is not easy to be identified in food, water, and air. These properties of arsenic pose a real danger to human health (Jomova et al., 2011). Arsenic is a universal contaminant in the environment and comes not only from polluted water and soil, but also from food that is rich in arsenic, such as marine food and garlic. Moreover, another source of arsenic is related to occupational activities in which involve wood preservatives, paper production, and pesticides. Though it is highly toxic, arsenic is used for curing trypanosomiasis (Florea & Büsselberg, 2006). Arsenic is not biodegradable under normal conditions. It can change from organic to inorganic forms and react with other chemicals by movement between/through air, water, and soil. Some arsenic compounds can end up in waterways, but most arsenic substances eventually accumulate in the sediment system (USEPA, 2015).

2.3 Hazards of heavy metal contamination

2.3.1 Toxic effects on plants

In general, plants are sensitive to the quantity of essential heavy metal ions, either a lack or excess of them. At the same concentration, these four ions (Pb, Hg, Cd and As) are strongly toxic towards some metabolic activities in plants (Nagajyoti et al., 2010).

Cadmium can affect plants' living cells and cause impaired photosynthesis, disrupted water imbalance, and altered mineral nutrition (Huang & Wang, 2010). In addition, cadmium is reported to be toxic to animals and microorganisms. In case of latter, cadmium significantly influences leaf litter decomposition. Hg is a unique heavy metal because of its various forms. One of the forms (Hg^{2+}) can cause physiological disorders in plant cells when it is present at high toxic level. More specifically, it can close the stomata of leaves and prevent water from flowing to other organs. Pb is a common toxic element in the soil and comes from various activities such as mining and smelting. The existence of lead at high concentrations can inhibit enzymatic activities and cause water imbalance (Yadav, 2010). As is used in smelting, mining, agricultural chemical, and wood preservative industries. Arsenic exists in both organic and inorganic forms. Generally, the inorganic form is more toxic than the organic forms. Being inorganic, As^{III} has higher toxicity than As^{v}. As^{v} can break the energy flow in plant cells, while As^{III} can inhibit cellular functions and even kill the plants (Akter et al., 2005).

2.3.2 Toxic effects on humans and animals

Lead can induce various symptoms which are related to the nervous system. Children may have difficulties in learning and concentration. Humans may have loss of memory, slowness of reaction time, and decreased understanding ability if exposed to lead for a long period of time (Järup, 2003). As for mercury, its inorganic and organic forms can affect health. For instance, acute toxicity of inorganic mercury can cause lung damage, while chronic poisoning is characterized by neurological and psychological symptoms, including tremors, restlessness, and sleep disturbances. Moreover, methyl mercury, an organic mercury, can cause nervous system damage after acute exposure, whereby there will be different symptoms at different stages. The hands and feet will be numbed in the early stages, followed by difficulties in coordination and subsequent death (Järup, 2003). With long term exposure to high concentrations of cadmium, kidneys and bones will be damaged. In addition, it is also a factor that leads to cardiovascular disease in animals. Furthermore, the IARC has deduced that cadmium can cause cancers both in humans and animals, such as lung cancer, prostate cancer, and kidney cancer. The toxicity of arsenic is of two types: acute and chronic. Acute toxicity occurs through ingestion of contaminated food and drinks by humans or animals. The symptoms include stomach bugs, anemia, diarrhea, and bloody urine. It can even cause shock and death in severe acute toxicity. Some of the human body systems are influenced by exposure to the inorganic form of As over a long time. These systems are skin, nervous and endocrine. Cancers of the liver, kidney, and lung are also related to As (Akter et al., 2005).

2.4 Importance of mangroves

Mangroves are vital to humans who live at the coastal areas and are dependent on its resources. They serve as commercial products, fishery resources, biomass, and litter production sites, apart from being sites for eco - tourism and education purposes. The benefits are as following:

(a) Social economic

It is demonstrated that the number of human activities in mangroves are of more than 70 types, ranging from fuel-wood collection to fishery (Dixon, 1989; International Union for Conservation of Nature and Natural Resources, 2006). In general, the yearly area of mangroves is valued from USD 2,000 to USD 10,000 per hectare (Kathiresan, 2012). According to the marine fisheries landing for the state of Selangor, the average landing was RM 376,645,139.000 yr^{-1} from 1995 to 1997 (Sasekumar & Chong, 2012). Marine fisheries are considered as off-shore fisheries, and some of them are dependent on mangroves. Based on the 50% mangrove - dependency assumption, the marine fisheries' productivity and value for the state of Selangor were calculated to be

56,646.330 tonnes yr^{-1} and RM 188,322,569.500 yr^{-1}, respectively. With a total mangrove area of 15,093 ha in Selangor, the productivity was estimated to be RM 12,477.480 $ha^{-1}yr^{-1}$ (Sasekumar & Chong, 2012). In some areas, mangroves can attract honey bees so people can undertake bee-keeping. Most of honey comes from *Ceriops* in India Sundarbans where every year, around 35,000 kg the former can be collected by the local people (Kathiresan, 2012). In addition, it is estimated that the Matang mangroves in Perak, Malaysia provide direct employment for about 1,400 workers and indirect employment for around 1,000 workers in the processing industries and timber extraction (Mansor, 2012).

(b) **Nursery grounds (fisheries)**

Mangroves serve as a habitat for fishery species where fishes stay for at least one period in their life cycle. There are many fishes which use mangroves as a nursery site when they are in the early stage of life (like juveniles). After that they will migrate to other places such as coral reefs. With the abundance of fish in the early life stages of life, mangroves also attract carnivorous fishes that undergo feeding migrations to mangrove areas. In addition, crabs, shrimps, prawns, and other crustaceans also use mangroves as their habitat. Mangrove species have some significant contributions to the commercial catch of fish. In eastern Australia, the commercial catch was 67 percentage, in the southern part of the Strait of Malacca, the benthic fish resources was 49 percent, and in ASEAN countries, the rate of fish and shrimp catch were 30 percent and almost 100 percent, respectively (Walters et al., 2008).

There are few studies conducted in Peninsular Malaysia which show that the mangrove system is crucial for sustaining the production of fishery resources, although fish and prawns rely on mangroves as a nursery ground. For instance, it was illustrated that the quantity of plant detritus accounted for 11% of the diet of fish at the Angsa Bank which is next to the mangroves of Selangor (Mansor, 2012).

(c) **Eco-tourism and education**

Mangroves can serve as public education sites due to the presence of salt - tolerant animals and plants. For example, the Kuala Selangor Nature Park has 13 species of plants and 157 species of birds, hence it is suitable site to conduct education programs for schools and the general public (Latiff, 2012). Moreover, eco - tourism is one of the ways to show the mangroves' beautiful and unique ecosystem to tourists, whereby economic returns can also be generated through jungle - trekking, bird - watching and fireflies - watching (Latiff, 2012).

(d) **Biomass and litter production**

Mangroves have a crucial role in the global carbon cycle. It is estimated that the total dry weight of mangrove biomass is around 8.700 gigatons, of which mangroves contribute about 4 gigatons of carbon to the carbon cycle (Kathiresan, 2012). The range of litter fall is from 130 - 1,870 g/m every year. There are studies reporting that the significant difference in litter production is due to habitat conditions, species composition, and production capacity of individual mangroves. As an example, litter production in the mangroves of Kenya was 0.011 t/ha/year, while in Bermuda, it was 9.400 t/ha/year, and in Australia, it was 23.690 t/ha/year (Kathiresan, 2012). Litter productions in the south Banjar reserve in Selangor in the *Avicennia*, *Sonneratia* and *Rhizophora* zones were 4.220, 3.380 and 4.320 g dry wt. m^{-2} day^{-1}, respectively. Leaves constituted the largest component, contributing 40 - 67% of the total production while twigs contributed 16 - 18% (Sasekumar & Chong, 2012).

(e) **Other functions of mangroves**

Mangroves act as sinks for waste water - borne pollutants. It has been reported that mangrove soil can trap and immobilize heavy metals and nutrients from waste water. It is believed that mangroves function as a filter for pollutants (Conley et al., 1991). Mangroves which are located in industrialized states in Peninsular Malaysia are no exception to this function (Latiff, 2012). Mangroves can also serve as a sediment removal system. Water flows slower in streams and rivers of mangrove areas than those at non-mangrove areas. In the former, sediments tend to settle down during their flow so the sea will be free from sediment (Wolanski, 1995). In addition, mangroves can prevent coastal erosions. Strong roots and buttress systems of the mangrove plants form a natural buffer between the land and the sea. Sometimes, they also break strong wind and wave actions. Mangroves also contribute to land - building through accretion (Othman, 1994).

2.5 Responses of mangroves to stressors

2.5.1 Sea level rise

Global warming is the main cause of sea level rise. The rise in sea level throughout the 20th century was recorded at 12 - 22 cm (Cazenave & Nerem, 2004). Another related term is called relative sea level change, in which the change in the sea level is influenced by regional and local factors. Mangroves can respond to changes in the relative sea level. There are three trends that contribute to sea level rise. One of them is the stable site - specific sea level (position of the mangrove is the same, while the sea level remains constant relative to the mangrove surface). Another one is the specific fall in the relative sea level (the mangrove margin migrates toward the sea while sea level falls relative to mangrove surface). Moreover, the seaward and landward margins of the mangrove will shift towards the land as the mangrove species maintain their preferred hydroperiod. This happens when the sea level rise is relative to the elevation of the mangrove sediment surface, and this can result in the land to move to higher areas (Gilman et al., 2008).

2.5.2 Oil spills

Spilled oil and its products can affect the growth of mangroves by befouling the gas exchange surfaces, and inducing chronic stress flowing exposure to residual amounts. Mangroves are sensitive to oil pollution and response as such: total defoliation will cause the death of trees; moderate defoliation will cause some elements to increase which include substrate temperature and salinity; toxic residue will induce leaf size changes and then decrease the photosynthetic area; mangrove tree functions will also be impaired through interference with the root and microbial substrate processes (Cintrón-Molero & Schaeffer - Novelli, 1992).

2.5.3 Sewage and solid waste disposal

The responses of mangroves to solid waste and sewage disposal depend on the amounts involved. This is due to the mangroves which are tolerant to nutrient enrichment. They are not harmed by indirect sewage input that has undergone some dilution before entering the mangrove ecosystem (Cintrón - Molero& Schaeffer - Novelli, 1992). Sewage has a high concentration of nutrients (nitrogen and phosphorus) and low salinity that can enhance mangrove growth and productivity. It is reported that mangroves can be used in waste water treatment (Mansor, 2012).

2.5.4 Heavy metals

Heavy metals come from various sources and can enter the mangrove system. They can accumulate in mangrove sediments. Following uptake by plants, high concentrations of the heavy metals can be extruded from plants via senescent tissues that fall on the surface sediments (Abohassan, 2013). Although mangrove plants have high relative immunity to the toxic effects of heavy metals, this is not the case for mangrove animals that can be more exposed to the risk of toxicity. Some of the heavy metals such as mercury and cadmium can cause physiological stress and reduce reproduction. In addition, heavy metals that accumulate in fish, shrimp, or edible mollusks will produce some health problems in the human population through consumption as part of the food chain (Mansor, 2012).

2.6 Heavy metals in mangroves

Elemental contamination of coastal areas caused by rapid industrialization and urbanization is now a global concern because they accept wastes which include anthropogenic and industrial ones (Udechukwu et al., 2015). These wastes, which are characterized by bioaccumulation, persistence, and toxicity, include heavy metals. Heavy metals enter the aquatic environment and accumulate in sediments via several pathways, such as natural process and anthropogenic process (Akoto et al., 2008). These processes include the disposal of liquid effluents, leachates, runoffs, surface soil erosion, water drainages, atmospheric depositions, abuse of heavy metal containing fertilizers and the use of pesticides in agricultural fields (Soares et al., 1999; Yang & Rose, 2005; Nouri et al., 2008). Mangroves are the most affected ecosystems of coastal areas as their sediments are significantly polluted by heavy metals (Loring & Rantala, 1992). These heavy metals are taken up by plants and finally enter the animals' bodies when the litter fall on the surface sediments (Lian et al., 1999).

2.6.1 Heavy metals in sediments

Heavy metals can be found in various water sources (tidal waters, freshwater rivers, and storm water runoffs) and can get deposited into the sediments. Sediments are the main reception tank for heavy metals in mangroves due to their anaerobic conditions, organic matter components, and richness in sulfide, all of which lead to the retention of water-borne heavy metals (Tam & Wong, 2000).

For tracing the concentrations of metals of sediments, the mangroves in United Kingdom, Australia, Mexico, China, Malaysia, Philippines, Brazil and Tanzania have been studied (Lewis et al., 2011). The concentrations of 22 metals (especially Pb, Cu, Zn) have been reported in mangrove sediments. The ranges of metal concentrations are from 0.010 - 87 μg/g (Cd), 0.010 - 845 μg/g (Cu), 0.610 - 125 μg/g (Cr), 2 - 1,120 μg/g (Hg), 1.200 - 3,253 μg/g (Mn), 0.080 - 1,950 μg/g (Pb), 0.300 - 102 μg/g (Ni) and 0.300

-2,372 μg/g (Zn). The relative concentrations of a specific metal usually varied with different geographical factors. For example, Fe > Mn > Zn > Pb > Cu > Ni > Cr > Cd in 2005, while in the following year, the order was Fe > Mn > Cr > Ni > Co > Cu > Pb > Hg (Lewis et al., 2011).

Mangroves in China are mainly distributed in six provinces (Guangdong, Guangxi, Hainan, Fujian, Taiwan and Zhejiang) and two special administrative regions(Macao and Hong Kong). Related studies primarily concentrated on Cu, Mn, Cd, Zn, Cr, Pb, As, Ni, Hg and Co. In most studies, the heavy metal concentrations were higher than the related background values, which illustrated that the mangroves have been polluted by heavy metals. Among these heavy metals, the concentrations of Pb, Cd, Zn and Cu in Guangdong, Fujian and Hong Kong mangrove sediments were more than those in the other provinces (Hainan and Guangxi). It may be due to the fact that the aforementioned cities are more developed and have large populations, higher levels of industrialization, and are known as international metropolises. In contract, Hainan and Guangxi are relatively less developed. In general, anthropogenic activities are the main reason behind the input of heavy metal contaminants in mangroves. However, Cr and Ni are of relatively high concentrations in the mangrove sediments in Hainan province. This might be due to great storage (Zhang et al., 2014). In a study on Hong Kong mangrove swamps, it is illustrated that heavy metals in fine-grained sediments have higher concentrations than those in sand-sized sediments. However, the concentrations of heavy metals in these two fractions were less significant in the mangrove swamps which are more heavily contaminated (Tam & Wong, 2000).

Tanzania, a developing country which has less industrial activities, is also facing environmental degradation due to an increase in development which produces a lot of pollutants, including heavy metals. Mangroves are affected by pollution caused by drainages from the cities towards the ocean and clearance for salt extraction. In a few heavy metal sediment analyses, As and Cd exceeded the detection limits. There is significance different between the sites and the presence of these elements (Ag, Fe, Mn and Zn); of all elements detected, the concentrations of Fe, Mn and Zn declined from a polluted to a clean area. From the year 1988 to 1999, the concentrations of the three elements have increased tremendously. For Al, it was 530 - 6,375 μg/g dry weight. For Fe and Cr, the concentrations increased from 630 - 3,539 μg/g and 2.700 - 10.100 μg/g, respectively (De Wolf et al., 2001).

Malaysia is a developing country that is transforming into a modern and advanced country by the year 2020. The west coast of Peninsular Malaysia has the most industrial areas, and at the same time, its offshore area is one of the busiest shipping lanes in the world. In addition, the mangroves of Peninsular of Malaysia are found mainly at the sheltered west coast. A study at the west coast of the Peninsular illustrated that the total Cu and Pb concentrations were 0.400 - 315 μg/g and 0.960 - 69.800 μg/g, respectively. Anthropogenic activities were identified as the contributor to the heavy metal concentrations at intertidal areas. For example, these activities contributed about 54% of the total Pb concentration in the sediments, while that for Cu was 46.3% (Yap et al., 2002). For the studies on Cd and Zn, it is reported that the concentrations of these two elements at the offshore area of the west coast were 0.100 - 1.420 μg/g and 4.000 - 79.050 μg/g, respectively. For the intertidal area, the concentrations of Cd was 0.030 - 1.980 μg/g and that for Zn was 3.120 - 306.200 μg/g between the year 1999 and 2001. By referring to different environmental quality guidelines on the contamination levels in sediments, these two elements did not serious contamination (Yap et al., 2003a). As for the studies on Hg, it is reported that its concentration in the surface sediments along the west coast of Peninsular Malaysia was caused by anthropogenic activities (Yap et al., 2003b). Law (1987) found that the mean concentration of mercury in the wet sediments at the Klang estuary was 0.200 mg/kg. Sakamoto et al. (1999) also reported the concentration of mercury in the sediments at Port Klang was 163 μg/kg based on a dry weight. For the detailed distribution of mercury at the Port Klang area, Haris and Aris (2013) showed that mercury was concentrated in the sediments along the Lumut Straits. Meanwhile, the stations located farther from Klang River and Langat River did not reveal enriched sediments. Furthermore, in East Malaysia, the mangrove surface sediments at the Mengkabong Lagoon in Sabah were detected to have heavy metals due to activities like local fishing, fish seed rearing as well as ecology tourism. The result was that the concentrations of the heavy metals (Fe, Cu, Pb, Zn and Al) increased significantly between different tides (from high tide to low tide), especially for Pb that was found to be uncontaminated (Praveena et al., 2008).

Some studies compared the concentrations of heavy metals in different mangrove sediments. Hong Kong researches Tam and Wong (2000) found that the concentrations of copper, zinc and lead were high in Nai Chung, while in Ho Chung, zinc and chromium levels were high. Tolo Pond as well as Sam Mun Tsai had significantly high concentrations of lead. These concentrations in sediments were considered to be moderately to seriously contaminated. The reason for this was the different geographical locations. For example, the high concentration of Cr in Ho Chung was because of surrounding dyeing operations. Zn came from waste dumping at the upper part of river while the heavy metals in Nai Chung were from nearby houses and restaurants. The high level of lead in Sam Mun Tsai was probably due to the fact that the area that was very close to the mariculture zone and seafood restaurants.

The correlations of heavy metal concentrations in sediments at the different locations in Malaysia was studied by Mokhtar et al. (2015). Mokhtar et al. (2015) used the one - way ANOVA method and showed that the concentrations of the selected metals were strongly different at various locations, especially at the downstream area of the Langat River.

In addition, the levels of heavy metal contamination can be evaluated using guidelines, such as the Interim sediment quality guidelines (ISQGs) and indexes such as the geoaccumulation index (I_{geo}), potential ecological risk index (RI), and enrichment factor (EF) (Cheng & Yap, 2015; Wang et al., 2015). Using the interim sediment quality guidelines, Khodami et al. (2017) reported that concentrations of metals (Cd and Pb) in the sediments of Bayan Lepas ranged from below low level to above high level at different stations. The contamination levels of the sampling stations were unpolluted to strongly polluted using the geoaccumulation index, and Bayan Lepas free industrial zone was at low risk using potential ecological risk index. Udechukwu et al. (2015) assessed the Cd, Pb and Zn pollution levels in the sediments of the Sg. Puloh mangrove using the interim sediment quality guidelines and it was reported that Zn and Pb may cause some environmental concern. According to different evaluation methods, Haris and Aris (2013) compared the enrichment factor with the geoaccumulation index by testing the mercury level in the Port Klang mangrove sediment. It was illustrated that the geoaccumulation index was more effective to categorize the enrichment level as compared to EF since the result of EF is highly influenced by the concentration of the reference element that was used in the calculation.

2.6.2 Heavy metals in plants

Plants can absorb various elements from the soil. Some of the absorbed elements are essential and required for plants which requires them as nutrients to complete the life cycle, such as copper, zinc and nickel. However, plants also absorb elements which can be toxic even at very low concentrations. They are referred to as non - essential elements, which include arsenic, cadmium, mercury and lead (Peralta - Videa et al., 2009).

2.6.2.1 Heavy metals in fresh tissues

Heavy metals have been detected in many mangrove plant tissues at different locations, such as the roots, leaves, branch and propagules (Zhang et al., 2014). So far, Zhang et al. (2014) reported that at least 15 species as study materials, including *Sonneratia caseolaris*, *S. hainanensis*, *S. ovata*, *S. apetala*, *Kandelia candel*, *Bruguiera gymnorrhiza*, *B. sexangula*, *Aegicera corniculatum*, *A. marina*, *A. ilicifolius*, *Ceriops tagal*, *Rhizophora stylosa*, *R. apiculata*, *Excoecaria agallocha* and *Lumnitzera apiculatal*. The concentrations of heavy metals illustrated that different species and tissues have different abilities for accumulating, for instance, *K. candel* accumulated more Ni, while *A. marina* tended to accumulated more Pb. By comparing the accumulative abilities of heavy metals (Cu, Pb, Ni, Cr, and Zn) among the three species, the highest value was seen in *S. caseolaris*, followed by *S. apetala*, and *K. candel* (Zan et al., 2002). For the following five plant species, the abilities to store Cu, Zn and Pb was in the order of *C. tagal* > *B. sexangula* > *K. candel* > *A. corniculatum* > *B. gymnorrhiza* (Li et al., 2013). Mangrove plant species have their own abilities for accumulating heavy metals, although there are also significant differences in plant tissues for same species, for example, Zn was abundant in the leaves of *S. apetala* while its branch have the highest concentration of Cu. For Hg, the concentration in different *K. candel* tissues declined in order of leaf > bark > root > xylem (Liang et al., 2011). In addition, there was a common and regular detection in tissue of nine plant species in Hainan, where Hg was generally abundant in the leaves; Zn and Cu in the fruits; As in the roots; Pb, Cd and Cr in the branch (Zhang et al., 2014).

Heavy metals were also found in *Rhizophora stylosa*. There was a study conducted for mangrove at Yingluo Bay, China, whereby the results showed that the branch had a high level concentration of Cu, Pb and Cr. The flowers were rich in Zn, while the roots and leaves had Cd and Mn respectively. In addition, the concentrations of Pb and Mn increased as the leaves developed, while the concentrations of Cu and Zn did not show much difference between young and mature leaves (Wen - jiao et al., 1997). In Malaysia, *Rhizophora apiculata* is used as sterilizers, deodorizers and fertilizers. This species was studied at the Setiu mangrove in Terengganu, and the results showed that the concentration of Cu was in the order of bark > leaf. On the contrary, the concentration of Pb was slightly higher in the leaf as compared to the bark (Kamaruzzaman et al., 2009).

MacFarlane et al. (2003) reported that although mangroves are a heterogeneous group as they adapt to anoxic and saline environments using differing strategies, metals (Pb, Cu and Zn) accumulation and partitioning were found to be similar across families and genera.

2.6.2.2 Heavy metals in litter

Mangrove ecosystems produce large amounts of litter in the form of falling leaves, branch and other debris. Organic matters are produced through the decomposition of litters. Invertebrates activities can accelerate the speed of decomposition of mangrove litter. After several weeks of decomposition, mangrove leaves can also attract crabs, shrimps and fishes (Kathiresan, 2012). The release of nutrients is dependent on the rate of loss of litter mass. Some studies reported that most heavy metals are not released, on the contrast, their accumulation slow down the rate of decomposition of the litter (Berg & McClaugherty, 2008). Mangrove litter may washed into rivers or streams, or remain at intertidal areas of mangroves. High concentrations of heavy metals in plants may be released by senescent leaves falling, then the litter fall which include heavy metals can be released into sediment, and when accompanied by high tidal activities, metals can be exported to adjacent systems (Abohassan, 2013). Heavy metals can also remain in mangroves, approximately 61% of which are removed by crabs (Nordhaus et al., 2006).

It is reported that non - essential metals can be accumulated in leaves and then removed from falling leaves. The regular of concentrations of metals (Cu, Pb, Zn and Cr) in litter fall in *Rhizophora stylosa* at the Yingluo Bay mangrove in China were the highest in the branch and lowest in litter fall leaves (Wen - jiao et al., 1997). It was also showed that high mercury areas raise some health concern of the mangrove ecosystem in the Port Klang (Haris & Aris, 2013). Mercury absorbed by mangrove plants from sediment can be released back into the environment through litterfall (Ding et al., 2011). The litterfall will decay and become food for the aquatic organisms, thus entering the food chain. Eventually, the mercury enrichment will not only adverse effect the flora and fauna, but also the people who harvest fish, crustaceans, and mollusks from the mangrove areas (Klekowski et al., 1999).

2.7 Comparison of heavy metal concentrations between sediments and different tissues

Avicennia marina is a species which can potentially be used for phytoremediation, as per the concentrations of metals that were detected in the natural and artificial mangroves at the Iranian coasts (Pakzadtoochaei, 2013). The results illustrated that all the studied metals were present in high concentrations in the sediments. Also, Cd was enrich in the leaves, Cu and Ni in the roots, and Zn in the flowers due to the fact that the plants required it for growth. As compared to the artificial forest (Kharchang Bay), the concentrations of metals in the natural forest (Gwatr) were higher, hence resulting in upstream runoff (Pakzadtoochaei, 2013). Moreover, *Avicennia marina* was studied at Futian in Shenzhen, China. The concentrations of the metals were compared between the tissues of living plants and that of the litter tissues. Result showed that Cu, Pb, Zn and Ni in litter leaves and branch have higher concentrations than those in living leaves and branch except for Cr were similar in two parts (Peng et al., 1997a). However, for the *A. marine* at the Newington North wetlands, Cu and Pb concentrations were higher level in the roots than in the sediments, while Zn showed a similar concentration between the roots and sediments. As Cu and Zn are essential micro - nutrients plants, some restricted mobility was seen. On the other hand, Pb is a non - essential element, so that have litter transported mobility which showed only 3 percent was taken up by the roots. In the leaf litter, the concentrations of Pb and Zn were similar while Cu was lower (MacFarlane et al., 2003).

There was a study on the accumulation and distribution of heavy metal in *Avicennia marina* and *Rhizophora apiculata* from the Balok mangrove forest in Pahang, Malaysia. It is reported that the concentrations of lead and copper in the roots of both species were higher than those in the leaves and bark, but were lower than those in the sediments (John & Waznah, 2011). The reason might be that the root system has direct contact with the sediment, unlike other tissues of the plants. In addition, there was an increase trend that are close to the estuary (John & Waznah, 2011). However, in Peninsular Malaysia, the roots of *Sonneratia caseolaris* had higher concentrations of Cr, Pb and Zn than in the sediments, also the aforementioned concentrations were higher in the leaves than in the roots, except for Cd (Nazli & Hashim, 2010).

CHAPTER 3: MATERIALS AND METHOD

3.1 Introduction

This chapter describes the materials and methods used in the detection of heavy metals in the mangrove systems at Pulau Kelang, Pulau Carey and Telok Gong. The study areas are also described. The study areas, sampling methodology, methods of analysis are carefully described and explained in detail.

3.2 Description of study sites

The Klang Islands Mangrove Forest Reserve (KIMFR), consists of Pulau Kelang, Pulau Ketam, Pulau Carey, Selat Kering, Pulau Gedong, Che Mat Zain, Jugra and Telok Gong (Norhayati et al., 2009). The Klang Islands are located within the Klang Straits in the western tropical coastal region of Malaysia. The Klang Strait at the west coast of Peninsular Malaysia is a busy shipping route with an average number of 2.8 vessels per day. The Klang Strait is also a popular site for fishing and recreational activities. These activities inevitably bring about anthropogenic inputs into the waterways. The principal pollutants are petrogenic, like petroleum hydrocarbons, biogenic - like *Escherichia coli* from domestic sewage, total suspended solids from sedimentation and siltation, as well as chemicals from industrial waste discharge (Sasekumar & Chong, 2012).

This study was conducted at natural and disturbed mangrove habitats in the Klang Islands (Figure 3.1). Specifically, the habitats include natural mangrove (Pulau Kelang N 03° 01′12.3″, E 101° 20′09″), an agricultural mangrove (Pulau Carey N 02° 49′41.0″, E 101° 21′56.7″) and an industrial mangrove (Telok Gong N 02° 56′54.1″, E 101° 22′14.0″).

Figure 3.1: The locations of sampling mangroves in Klang Islands

Pulau Kelang is a natural mangrove forest island. The Pulau Kelang mangrove forest reserve is gazetted as a virgin reserve forest by the Selangor Forestry Department and is located 11 km from Port Klang. Telok Gong as an industrial and human settlement area, and its mangrove forest reserve is located near to roads, factories, and houses. In 2001, 80.900 ha of the Telok Gong mangrove forest reserve have been degazetted for development. Pulau Carey has the biggest mangrove area among the Klang Islands, and is separated from the Selangor coast by the Langat River at its east and the Klang River at its north (Motamedi et al. 2014). Oil palm cultivation is the primary economic activity of the island. Oil palm plantations occupy approximately 65% of the total area of the island. This is followed by the mangrove forest reserve, while the rest consist of state land and settlements. Since the opening of the land for agricultural activities, the area of the mangrove has suffered an obvious loss. Assuming the mangrove area was 16,187.450 hectares in the 1900s, only approximately 1,876.850 ha currently remains, which accounts to 88% of mangrove loss due to agricultural activities and development of residential areas (Baharuddin et al., 2013).

3.3 Sampling and preparation

Samples were collected at low tide within four plots with tidal gradient intervals of 25 m between the plots (from the sea to the land at each site). Two replications were done for each plot. For soil sampling, the samples were collected using a soil sampler of depth 15 cm. For litter (branch and leaves) sampling, the samples were collected using a 1 m x 1 m quadrat. Fresh tissues (leaves and branch) were collected using a pair of scissors. All samples were immediately sealed within plastic bag and transported back to the laboratory on the same day. The vegetation samples were washed with tap water to remove the dust and were dried naturally.

3.4 Analytical procedures

3.4.1 Apparatus

In this experiment, the concentrations of the heavy metals were determined by Inductively Coupled Plasma Optical Emission Spectrometry 5100 series (Agilent), Flow Injection Mercury System 400 and Inductive Coupled Plasma Mass Spectrometry 7500 series (Agilent).

3.4.2 Soil

For soil analysis, the fresh soil sample was divided into two processes namely process A and process B. Process A involved instrumental analysis while process B was used to determine the dry weight factor (DWF). The weights of the soil for processes A and B were 1.500 g and 10 g respectively. The samples were then subjected to the following procedures:

Process A: 1.500 g of soil was weighed and placed in a beaker prior to the digestion process. Several types of acids were added into the soil sample. This include 4 ml of dilute nitric acid solution (2 ml of 65% nitric acid + 2 ml of deionized water), 2 ml of 65% nitric acid, and 2 ml of 37% hydrochloric acid. The acid - soil mixture was then heated using an 85°C hot block for 30 minutes. Once the heating process was completed, the soil sample was then placed in a desiccator to cool down. Then, the volume of the sample was made to be 50 ml by adding deionized water. The digested sample was aspirated into the Inductively Coupled Plasma Optical Emission Spectrometer (ICP - OES) for arsenic, cadmium, and lead analysis. Mercury analysis was done by the CVASS (FIMS - 400). Once the readings were obtained, the concentrations of the heavy metals in terms of dry weight were determined using the following formula:

$$\text{Heavy metal concentration} = (R_1 \times \text{Markup Vol})/(W \times DWF)$$

Where:

R_1= Instrument reading

Markup Vol=50 ml

W=Sample weight

DWF=Dry weight factor

Process B: In order to calculate the dry weight factor (DWF) in process A, 10 g of soil was weighed and placed in a crucible cup prior to heating in oven for 2 hours at 103 - 105°C. Once the heating process was completed, the soil sample was then placed in the desiccator to cool down before being weighed again. The DWF was determined using the following formula:

$$DWF = 1 - (W_1 - W_2)/(W_1 - W_{cup})$$

Where:

W_1= Weight of sample and crucible before heating

W_2= Weight of sample and crucible after heating

W_{cup}=Weight of crucible cup

3.4.3 Vegetation (fresh tissues and litter)

The plants samples were grinded and weighed to 1 g before being placed in a beaker. 20 ml of 65% nitric acid was added to the ground samples. The acid - ground sample mixture was then heated using an 85°C hot block for 2 hours. Once the heating process was completed, the grinded sample was then placed in desiccator to cool down. Then, the volume of the sample was made to be 50 ml by adding deionized water. The digested sample was aspirated into the Inductively Coupled Plasma Mass Spectrometer (ICP - MS) for arsenic, cadmium, lead and mercury analysis. Once the readings were obtained, the concentrations of heavy metals in terms of dry weight were determined using the following formula:

$$\text{Heavy metal concentration} = (R_1 \times \text{Markup Vol})/W$$

Where:

R_1= Instrumental reading

Markup Vol=50 ml

W=Sample weight

3.5 Geoaccumulation index (for sediments)

In this study, the geoaccumulation index (I_{geo}) was calculated to determine if there was any enrichment of heavy metals in these three mangrove sediments. It is an effective and meaningful way to explain the sediment quality (Fernandes et al., 2012). The calculated I_{geo} determines the level of contamination by comparing the current metal concentration with those that are accepted as background levels. The background metal contamination is multiplied with 1.500 to take into account the natural fluctuations of the element in the background. I_{geo} is calculated using the Müller (1969) expression:

$$I_{geo} = \log2(C_n/1.5B_n)$$

Where:

Cn = metal content in analyzed sediment (in milligrams per kilogram)

Bn = Background content of metal (in milligrams per kilogram). In this study, the selected background heavy metal concentrations at Port Klang were Pb (39.800 mg/kg), Hg (0.080 mg/kg), Cd (0.186 mg/kg), and As (18.790 mg/kg) (Yap, 2005).

The I_{geo} results were then used to determine the metal contamination levels. Generally, the results can be categorized into seven classes (Table 3.1).

Table 3.1: The degree of metal contamination based on the I_{geo} contamination Classification (Haris & Aris, 2013)

I_{geo} value	Class	Designation of sediment quality
$I_{geo} \geq 5$	6	Extremely contaminated
$4 < I_{geo} < 5$	5	Strong to extremely contaminated
$3 < I_{geo} < 4$	4	Strongly contaminated
$2 < I_{geo} < 3$	3	Moderately to strongly contaminated
$1 < I_{geo} < 2$	2	Moderately contaminated
$0 < I_{geo} < 1$	1	Uncontaminated to moderately contaminated
$I_{geo} \leq 0$	0	Uncontaminated

3.6 Accumulation coefficient of heavy metals

The ability of a plant to absorb heavy metals in the sediments is generally evaluated by an accumulation coefficient (Wen - jiao et al., 1997). The accumulation coefficient (AC) was calculated using the following equation (Al - Farraj et al., 2009):

$$AC = C_{branch\ or\ leaves}/C_{soil}$$

Where:

$C_{branch\ or\ leaves}$ = The concentration of heavy metal (mg/kg) in the branch or leaves

C_{soil} = The concentration of the heavy metal (mg/kg) in the soil

3.7 Statistical analysis

Statistical analyses were performed using Microsoft Excel and SPSS version 22 software. Pearson correlation (level of significance was 0.050) was used to determine the relationship between the heavy metal concentrations in sediments and the tidal gradient. According to Cohen (1988), absolute r values of 0.100, 0.300, and 0.500 are considered to be weak, moderate and strong respectively. Owing to the non - normal of the data of this variable, the generalized linear model and Bonferroni test were employed to compare the heavy metal concentrations between land use types. The relationships between heavy metal accumulation in plant tissues and individual sediment metals were examined using the bivariate correlation analyses. Nonparametric tests (Kendall's coefficient of concordance) were used to compare the significance of the concentration differences between the tissues of the three mangroves. In order to predict the adverse biological effects in the contaminated sediments, this study used the New York Sediment Criteria and Provincial Sediment Quality Guidelines which included the effect range low and effect range high. Sany et al. (2011) reported that the effect range low indicates that the sediment contaminants do not have adverse effects on the aquatic organisms, while the effect range high shows that the sediment contaminants have adverse effects on the said organisms. In addition, metal concentrations which are between effect range low and effect range high indicate that the contaminants are considered have potential adverse effects.

CHAPTER 4: RESULTS

4.1 Introduction

Heavy metals can be found naturally in the aquatic environment, but their levels have increased tremendously due to human activities such as industrial waste dumping, as well as agricultural and mining activities. Heavy metals enter the water runways through land surface runoffs, rainfall precipitations and factory waste outlet point discharges. These trace elements may remain within the water bodies or soil and are taken up by plants or animals. This can eventually affect humans, which are consumers. Therefore it is important that a baseline study be conducted to determine the heavy metal concentrations. This study reports the concentrations of Pb, Hg, Cd and As in the sediments, litter and plant tissues obtained from the mangroves of three land use types (Pulau Kelang mangrove, Pulau Carey mangrove and Telok Gong mangrove) in the Klang Islands. These metals were selected because of their toxicity to humans and animals.

4.2 Heavy metal in sediment

4.2.1 Heavy metal concentrations in sediment

The concentrations of heavy metals at Pulau Kelang, Pulau Carey and Telok Gong are summarized in Figure 4.1. At Pulau Kelang, the ranges of the traced heavy metal concentrations were 15.600 - 18.310 mg/kg (Pb), 0.051 - 0.104 mg/kg (Hg), 0.170 - 0.430 mg/kg (Cd) and 4.42 - 8.45 mg/kg (As). At Pulau Carey, the ranges of traced heavy metal concentrations were 9.845 - 13.670 mg/kg (Pb), 0.050 - 0.093 mg/kg (Hg), 0.100 - 0.240 mg/kg (Cd) and 5.890 - 10.580 mg/kg (As). At Telok Gong, the ranges of traced heavy metals concentrations were 10.310 - 21.200 mg/kg (Pb), 0.085 - 0.119 mg/kg (Hg), 0.360 - 0.400 mg/kg (Cd) and 5.920 - 15.000 mg/kg (As). In all the sampled mangroves, the concentrations of Pb and As were higher than those of Hg and Cd. In other word, the concentration order was Pb > As > Cd > Hg.

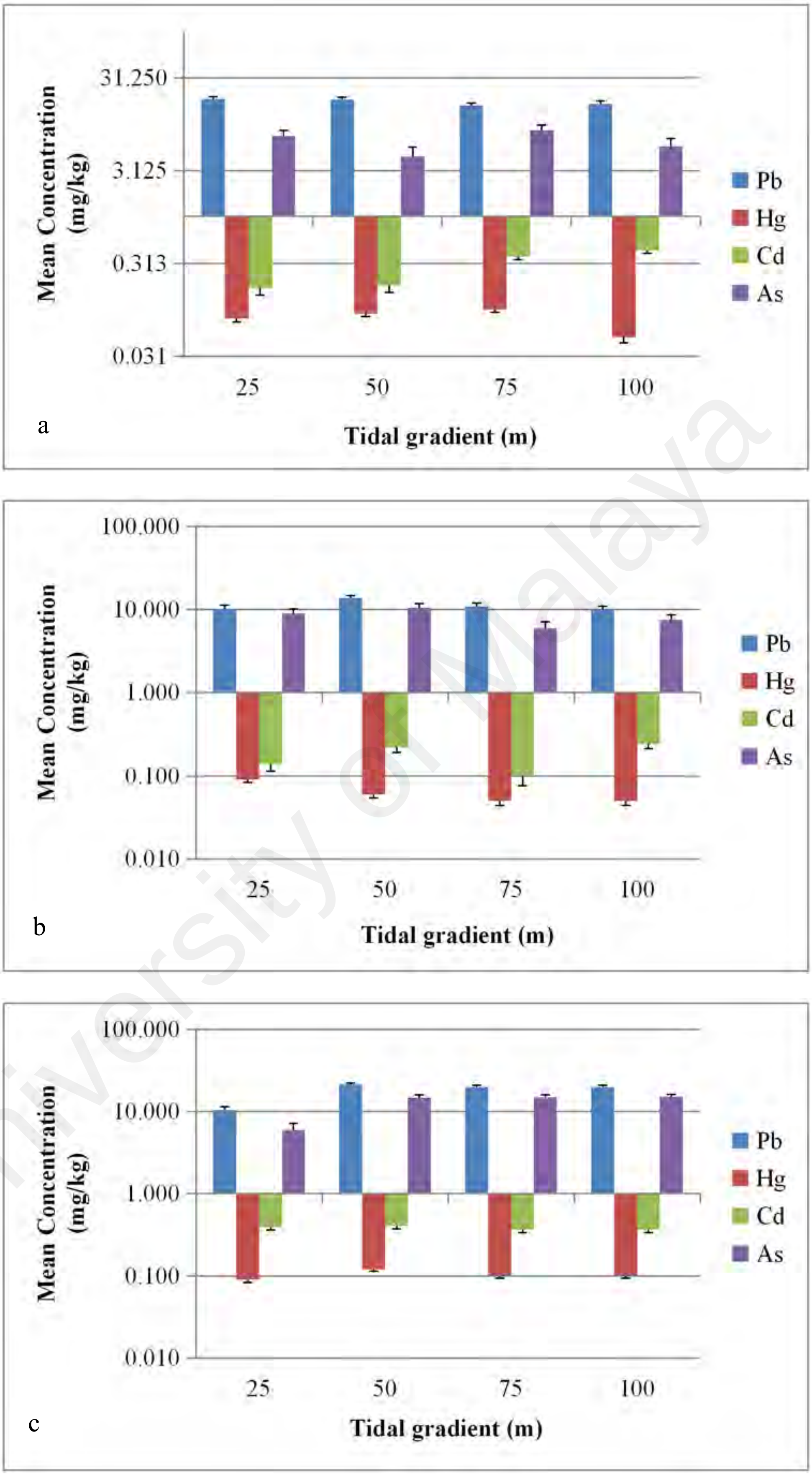


Figure 4.1: Mean concentrations (mg/kg) of heavy metals from seaward to landward in three mangroves (a: Pulau Kelang mangrove; b: Pulau Carey mangrove; c: Telok Gong mangrove)

4.2.2 Correlation of heavy metal concentrations in sediment with tidal gradient

Based on Figure 4.1, the heavy metal concentrations varied with increasing tidal gradients from seaward to landward. Table 4.1 shows the correlation between the concentrations of the traced heavy metals in sediments and tidal gradient. The results indicated that the concentrations had either positive or negative relationship with tidal gradient, but there were no significant differences ($P > 0.050$).

Table 4.1: Correlation of heavy metal concentrations in sediments with tidal gradient in Pulau Kelang, Pulau Carey and Telok Gong mangroves

Site	Element	R value	P value
Pulau Kelang	Pb	-0.824	0.176
	Hg	-0.351	0.649
	Cd	0.947	0.053
	As	-0.050	0.950
Pulau Carey	Pb	-0.271	0.729
	Hg	-0.928	0.072
	Cd	0.352	0.648
	As	-0.590	0.410
Telok Gong	Pb	0.694	0.306
	Hg	-0.009	0.991
	Cd	-0.814	0.186
	As	0.795	0.205

Levels of significance are indicated as $P < 0.050$.

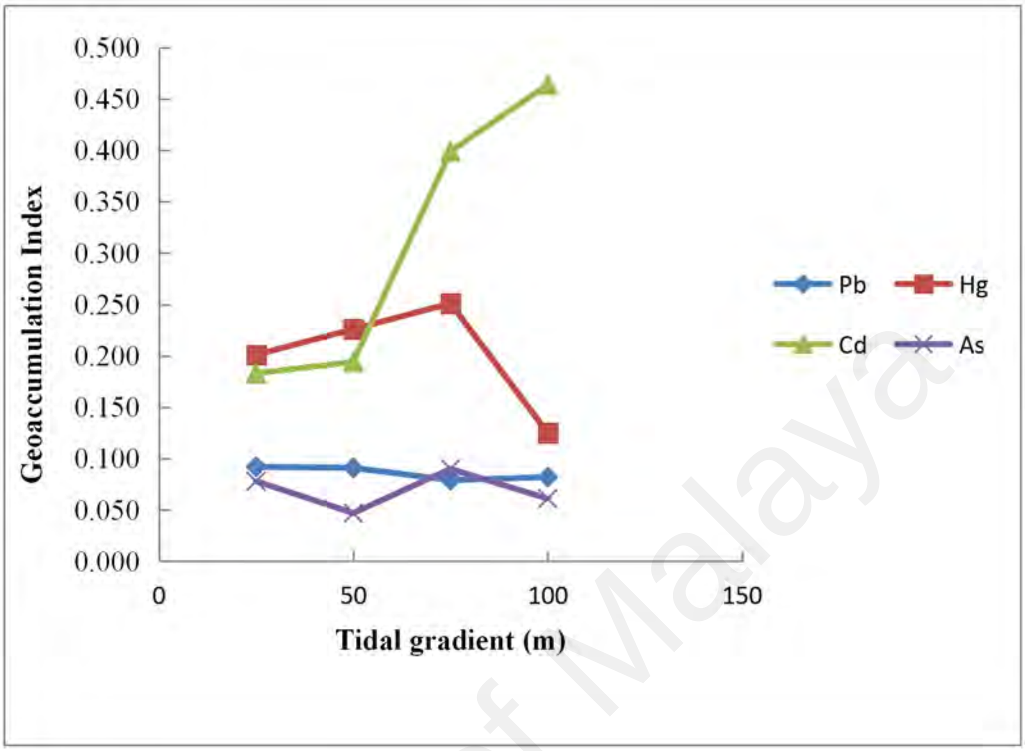
At the Pulau Kelang mangrove, Pearson correlation indicated that the concentrations of all the traced heavy metals in the sediments were negatively correlated with the tidal gradient, except for cadmium. However, the levels of correlation were different, with Pb and Cd showing a strong relationship ($|r| > 0.500$) between concentration in sediment and tidal gradient. Meanwhile, Hg had a moderate correlation ($0.300 < |r| < 0.500$). In addition, with the changing of tidal gradient, the concentrations of all the elements did not have a significant statistical correlation ($p > 0.050$) in sediment with the changes in the tidal gradient. Similarly, at Pulau Carey mangrove, Pearson correlation indicated that the concentrations of all the traced heavy metals in the sediments were negatively correlated with increased tidal gradient, except for cadmium. As for correlation analysis, Pb showed a weak correlation ($0.100 < |r| <$

0.300), Cd showed a medium correlation ($0.300 < | r | < 0.500$), as well as Hg and As a strong correlation ($| r | > 0.500$). In addition, the concentrations of all the elements did not have a significant statistical correlation ($p > 0.050$) with changes in the tidal gradient. At the Telok Gong mangrove, the Pearson correlation indicated that the concentrations of heavy metals (Hg and Cd) in the sediments were negatively correlated with the increased tidal gradient, while of the converse was true for Pb and As. At this mangrove, the results showed that Pb, Cd and As were strongly correlated ($| r | > 0.500$) with the tidal gradient, while Hg weakly correlated ($0.100 < | r | < 0.300$). In addition, the changing tidal gradient did not result in significant ($p > 0.05$) difference in the concentrations of the elements in the sediments.

4.2.3 Geoaccumulation Index (I_{geo})

Geoaccumulation index is a method to determine the geological substrates and metal pollution of the environment. Many researchers on mangrove sediments have also applied this index to explain the sediment quality. According to the I_{geo} equation, the background concentrations of metals at Port Klang value were Pb: 39.800 mg/kg, Hg: 0.080 mg/kg, Cd: 0.186 mg/kg and As: 18.790 mg/kg (Yap, 2005). The degree of tracing heavy metal contamination is shown in the following figures (Figure 4.2, Figure 4.3 and Figure 4.4).

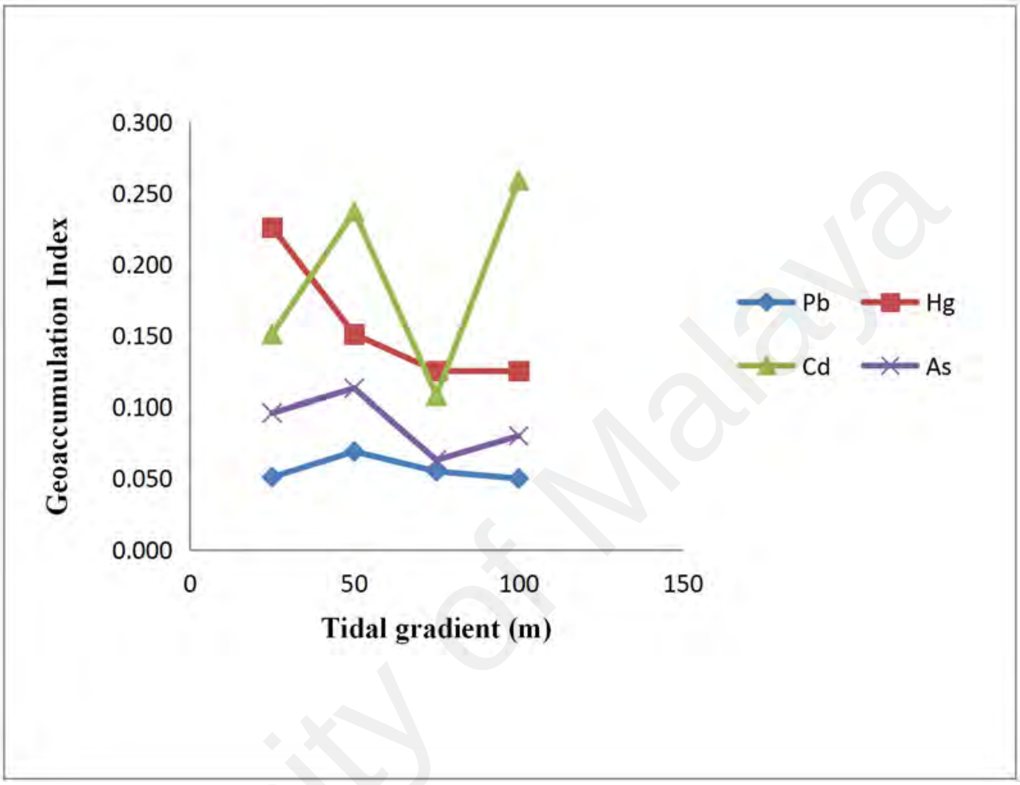
At the Pulau Kelang mangrove (Figure 4.2), the geoaccumulation indexes of these four heavy metals in the sediments were between 0 and 0.500. This indicated that the I_{geo} values of Pulau Kelang were $0 < I_{geo} < 1$. It showed that the sediment quality was uncontaminated to moderately contaminated, and was rated as class 1. As compared to the other three heavy metals, cadmium pollution of the in Pulau Kelang sediment was relatively higher, especially when going from seaward to landward. According to the polynomial trend - lines, the contamination of cadmium increased with the increase in tidal gradient, while that of mercury decreased when going landward. In addition, the geoaccumulation indexes of lead and arsenic remained constant.



Element	Model (Polynomial trend - line)	R^2
Cd	$y = 2E\text{-}05x^2 + 0.0015x + 0.1155$	0.909
Hg	$y = -6E\text{-}05x^2 + 0.0067x + 0.0627$	0.872
Pb	$y = 2E\text{-}06x^2 - 0.0004x + 0.1015$	0.732
As	$y = 8E\text{-}07x^2 - 0.0001x + 0.0735$	0.004

Figure 4.2: Geoaccumulation indexes of sediments from seaward to landward in Pulau Kelang mangrove

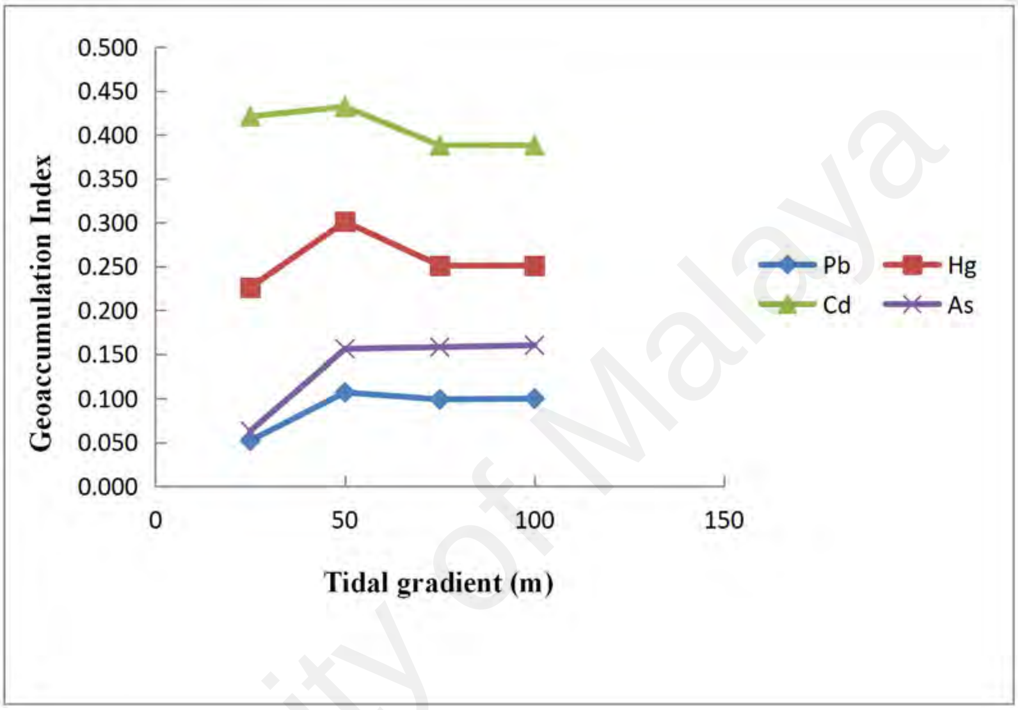
At the Pulau Carey mangrove (Figure 4.3), the geoaccumulation indexes of these four heavy metals were between 0 and 0.300, so the I_{geo} values of Pulau Carey were $0 < I_{geo} < 1$. Hence, Pulau Carey was rated as class 1. It showed that the sediment quality was uncontaminated to moderately contaminated. As compared to the other heavy metals, cadmium pollution of the Pulau Carey sediment was relatively higher, especially when going from seaward to landward. According to the polynomial trend-lines, the contamination of cadmium increased with the increase in tidal gradient, while those of mercury, arsenic and lead decreased when going landward.



Element	Model (Polynomial trend - line)	R^2
Cd	$y = 3E\text{-}05x^2 - 0.0025x + 0.2213$	0.195
Hg	$y = 3E\text{-}05x^2 - 0.0051x + 0.3328$	0.996
As	$y = - 0.0004x + 0.1125$	0.349
Pb	$y = -9E\text{-}06x^2 + 0.0011x + 0.0317$	0.636

Figure 4.3: Geoaccumulation indexes of sediments from seaward to landward in Pulau Carey mangrove

At the Telok Gong mangrove (Figure 4.4), the geoaccumulation indexes of these four heavy metals were between 0 and 0.500, so the I_{geo} values of Telok Gong were $0 < I_{geo} < 1$. The status of Telok Gong was class 1, which showed that the sediment quality as uncontaminated to moderately contaminated. As compared with other heavy metals, cadmium pollution of the Telok Gong sediment was relatively higher. The geoaccumulation indexes of all the traced heavy metals (from seaward to landward) were in the order of Cd > Hg > As > Pb. According to the polynomial trend - lines, the contamination of cadmium slightly decreased from seaward to landward, while those of the other three heavy metals increased gently going landward.



Element	Model (Polynomial trend - line)	R^2
Cd	$y = -4E\text{-}06x^2 - 2E\text{-}05x + 0.4292$	0.682
Hg	$y = -3E\text{-}05x^2 + 0.0039x + 0.1573$	0.484
As	$y = -4E\text{-}05x^2 + 0.0057x - 0.0528$	0.939
Pb	$y = -2E\text{-}05x^2 + 0.0032x - 0.012$	0.865

Figure 4.4: Geoaccumulation indexes of sediments from seaward to landward in Telok Gong mangrove

Generally, all the sampled mangroves were uncontaminated to moderately contaminated (Class 1) by the four heavy metals. In these three mangroves, there were relative hot spots from seaward to landward for the accumulation of heavy metals .

4.2.4 Comparison of heavy metal concentrations between different land use types

Owing to the non- normal distribution of this variable, the generalized linear model (GLM) was employed for the analysis. Table 4.2 indicated that there are significant differences in the heavy metal scores ($p < 0.010$) between the locations, which showed that the means of the heavy metal score changes were different between the locations.

Table 4.2: Summary of analysis of generalized linear model for heavy metals

Element	Wald Chi-Square	P value
Pb	22.680	<0.001
Hg	13.243	0.001
Cd	20.710	<0.001
As	13.851	0.001

In addition, the Bonferroni post hoc test was conducted to identify the specific means were different. Figure 4.5 showed the mean concentrations of the heavy metals at the three mangrove sediments. All the tested heavy metals had the highest concentrations in the Telok Gong mangrove sediment. Table 4.3 indicated that Pb concentrations were significantly different between Pulau Kelang and Pulau Carey mangroves, with the former being 5.940 mg/kg higher than the latter. There was also a significantly difference between Pulau Carey and Telok Gong. Pulau Carey was lower 6.624 mg/kg than Telok Gong, while Pb concentration was not significantly different ($p > 0.050$) between Pulau Kelang and Telok Gong. In terms of the Hg concentrations, there was a significant difference between Pulau Carey and Telok Gong mangroves, with the former being 0.035 mg/kg lower than the latter. As for the Cd concentrations, there was a significant difference between Pulau Kelang and Pulau Carey mangroves, with the former being 0.113 mg/kg higher than the latter. Pulau Carey and Telok Gong mangroves also showed a significant difference, with the former being 0.203 mg/kg lower than the latter. In terms of the As concentrations, there were also significant differences between Pulau Carey and Telok Gong (the former being 4.370 mg/kg lower than the latter), as well as Pulau Kelang and Telok Gong (the former being 6.118 mg/kg lower than the latter).

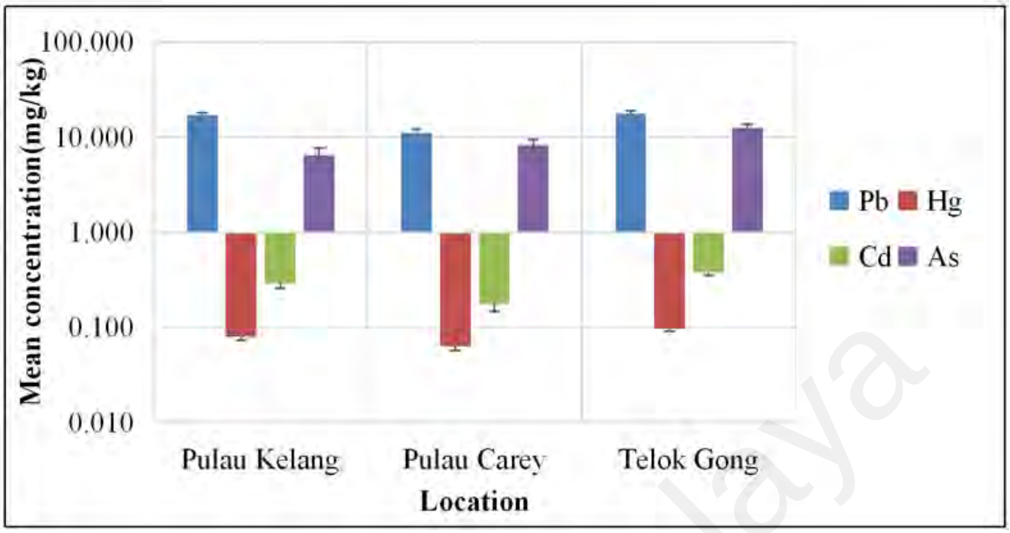


Figure 4.5: Mean concentrations (mg/kg) and standard deviations of heavy metals in three mangrove sediments

Table 4.3: Results of Pos-Hoc tests for heavy metal scores between locations

Element	(I) Location	(J) Location	Mean Difference (I-J)	Std. Error	df	P value	95%Wald Confidence Interval for Difference	
							Lower	Upper
Pb	Pulau Kelang	Pulau Carey	5.940[a]	1.530	1.000	<0.001	2.278	9.602
	Pulau Carey	Telok Gong	-6.624[a]	1.530	1.000	<0.001	-10.286	-2.961
	Pulau Kelang	Telok Gong	-0.684	1.530	1.000	1.000	-4.346	2.979
Hg	Pulau Kelang	Pulau Carey	0.016	0.010	1.000	0.274	-0.007	0.039
	Pulau Carey	Telok Gong	-0.035[a]	0.010	1.000	0.001	-0.058	-0.012
	Pulau Kelang	Telok Gong	-0.019	0.010	1.000	0.154	-0.042	0.004
Cd	Pulau Kelang	Pulau Carey	0.113[a]	0.045	1.000	0.035	0.006	0.219
	Pulau Carey	Telok Gong	-0.203[a]	0.045	1.000	<0.001	-0.309	-0.096
	Pulau Kelang	Telok Gong	-0.09	0.045	1.000	0.131	-0.197	0.017
As	Pulau Kelang	Pulau Carey	-1.748	1.693	1.000	0.906	-5.801	2.306
	Pulau Carey	Telok Gong	-4.370[a]	1.693	1.000	0.030	-8.424	-0.316
	Pulau Kelang	Telok Gong	-6.118[a]	1.693	1.000	0.001	-10.171	-2.064

a. The mean difference is significant at the 0.050 level between sites.

4.2.5 Comparison of heavy metal concentrations with back ground values and sediment quality guidelines

The concentrations of the heavy metals at the three sampling locations were compared with the background value of Port Klang and the sediment quality guidelines to assess the environmental conditions and effects of industrial and economic activities on the aforementioned locations. Table 4.4 showed the guidelines that used in this study, which were the New York Sediment Criteria and Provincial Sediment Quality Guidelines for metals.

Table 4.4: Comparison of heavy metal concentrations with Port Klang back ground value and sediment quality guidelines (SQG)

Concentration of heavy metal(mg/kg)	Pb	Hg	Cd	As
Pulau Kelang (present study)	17.066± 1.313	0.080± 0.022	0.288± 0.132	6.470± 1.766
Pulau Carey (present study)	11.126± 1.749	0.063± 0.021	0.175± 0.066	8.218± 2.010
Telok Gong (present study)	17.750± 5.007	0.098± 0.015	0.378± 0.021	12.588± 4.447
Back ground value in the port klang (Yap, 2005)	39.800	0.080	0.186	18.790
[a]Lowest effects range (New York Sediment Criteria)	32.000	0.150	0.600	6.000
[a]Sever effects range (New York Sediment Criteria)	110.000	1.300	9.000	33.000
[a]Lowest effects range (ISQG-low) (ISQG1992)*	31.000	0.200	0.600	6.000
[a]High effects range (ISQG-high) (ISQG 1992)*	250.000	2.000	10.000	33.000

*Interim Sediment quality criteria guideline (ISQG) (1992)
a. (Sany et al., 2011)

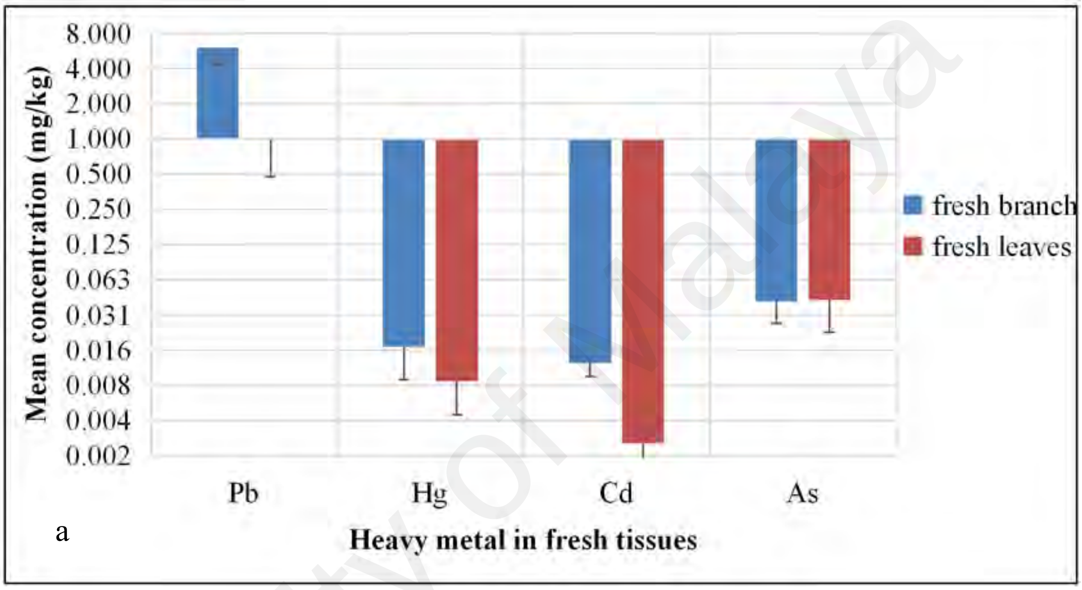
4.3 Heavy metal in mangrove fresh tissues

4.3.1 Heavy metal concentrations

In these three mangroves, the concentrations of heavy metals in the fresh tissues varied in different fractions (Table 4.5). In general, the mean concentrations of Pb, Hg, Cd and As were higher in fresh branch as compared to fresh leaves. It was expected that the concentrations of As in the leaves would be higher than those of the branch at both Pulau Kelang and Telok Gong mangroves. For both fresh branch and fresh leaf tissues, the concentrations of the heavy metals were in the order of Pb > As > Hg > Cd. In addition, the results indicated that the concentrations of Pb in fresh branch and leaves were much higher than those of the other three heavy metals (Figure 4.6).

Table 4.5: The ranges of heavy metal concentrations (mg/kg) in fresh tissues in three mangroves

Site	Tissue	Pb	Hg	Cd	As
Pulau Kelang	Fresh branch	2.931-8.714	0.000-0.037	0.008-0.016	0.021-0.068
	Fresh leaves	0.182-2.548	0.002-0.020	0.000-0.007	0.008-0.081
Pulau Carey	Fresh branch	1.246-2.203	0.006-0.028	0.006-0.010	0.078-0.109
	Fresh leaves	0.196-0.580	0.006-0.017	0.000-0.001	0.015-0.042
Telok Gong	Fresh branch	1.235-3.415	0.026-0.053	0.000-0.037	0.040-0.080
	Fresh leaves	0.578-0.858	0.015-0.064	0.000-0.003	0.060-0.194



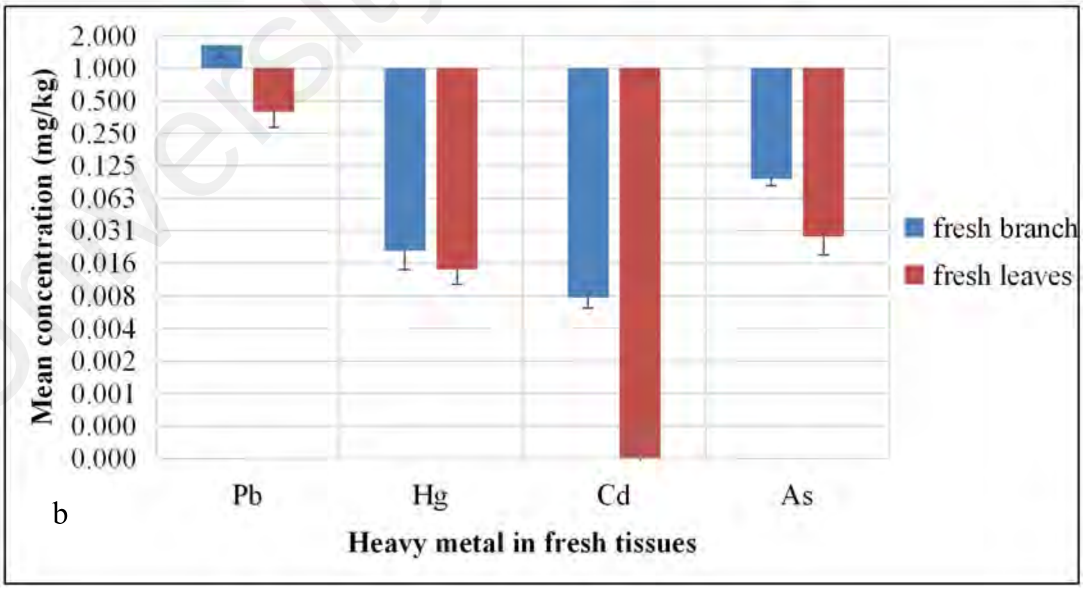


Figure 4.6: The mean concentrations (mg/kg) and standard deviations of heavy metals in fresh tissues in mangroves (a: Pulau Kelang mangrove; b: Pulau Carey mangrove; c:Telok Gong mangrove)

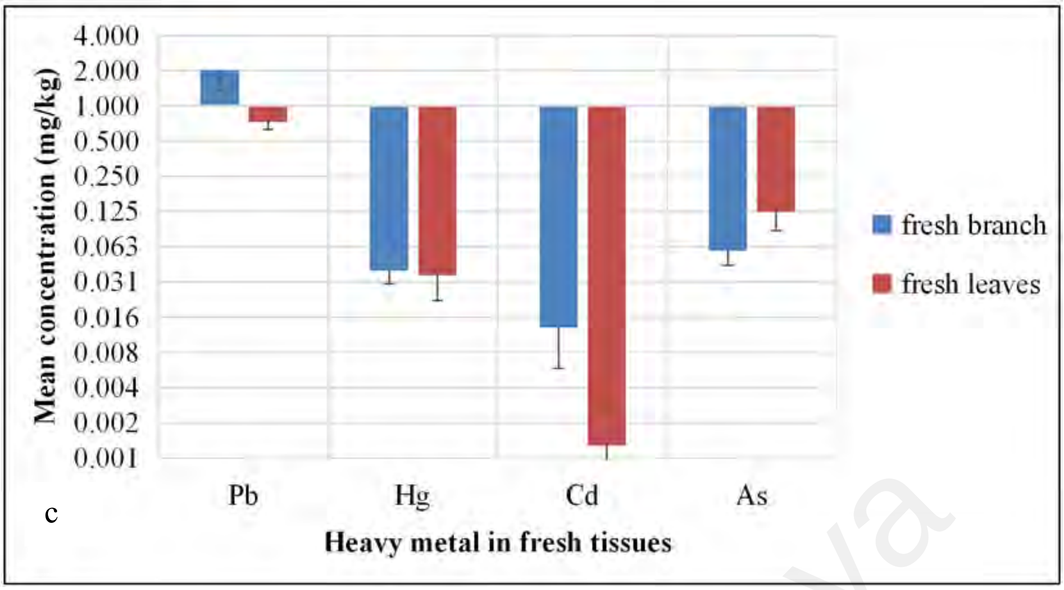


Figure 4.6: Continued

4.3.2 Heavy metal concentrations in different land use types

4.3.2.1 Fresh branch

Owing to the non - normally distribution of this variable, the generalized linear model (GLM) was employed for the analysis. Table 4.6 showed that there were significant differences in the heavy metal scor ($p < 0.010$) between the locations, which indicated that the means of heavy metal scores was different between locations, except for that of Cd which did not significantly difference between the locations ($p > 0.010$).

Table 4.6: Summary of analysis of generalized linear model for heavy metals

Element	Wald Chi-Square	P value
Pb	40.010	<0.001
Hg	35.367	<0.001
Cd	1.676	0.433
As	42.627	<0.001

In addition, the Bonferroni post hoc test was conducted to identify the specific means which were different. Figure 4.7 showed the mean concentrations of the heavy metals in the fresh branch of the three mangroves, which were of the following orders: Pulau Kelang > Telok Gong > Pulau Carey (Pb), Telok Gong > Pulau Carey > Pulau Kelang (Hg), Telok Gong > Pulau Kelang > Pulau Carey (Cd), Pulau Carey > Telok Gong > Pulau Kelang (As). Table 4.7 showed the differences in the significance levels between the individual location points. It showed that Pb concentrations at Pulau Kelang and Pulau Carey mangroves were significantly different, with the former being 4.383 mg/kg higher than the latter. There was also a significant difference in the Pb concentration between Pulau Kelang and Telok Gong mangroves, with the former being 3.999 mg/kg higher than the latter. There was no significant difference ($p > 0.050$) in the Pb concentration between Pulau Carey and Telok Gong mangroves. For the Hg concentrations, there was significant difference between Pulau Carey and Telok Gong mangroves, with the former being 0.019 mg/kg lower than the latter. There was also a significant difference between Pulau Kelang and Telok Gong mangroves, with the latter being 0.022 mg/kg higher than the former. For Hg concentration, there was no significant difference ($p > 0.050$) between Pulau Kelang and Pulau Carey mangroves. In terms of As concentrations, there was a significant difference between Pulau Kelang and Pulau Carey mangroves, with the former being 0.054 mg/kg lower than the latter. There was also a significant difference between Pulau Carey and Telok Gong mangroves, where the former being 0.037 mg/kg higher than the latter. Meanwhile, the As concentration was not significantly different between Pulau Kelang and Telok Gong mangroves.

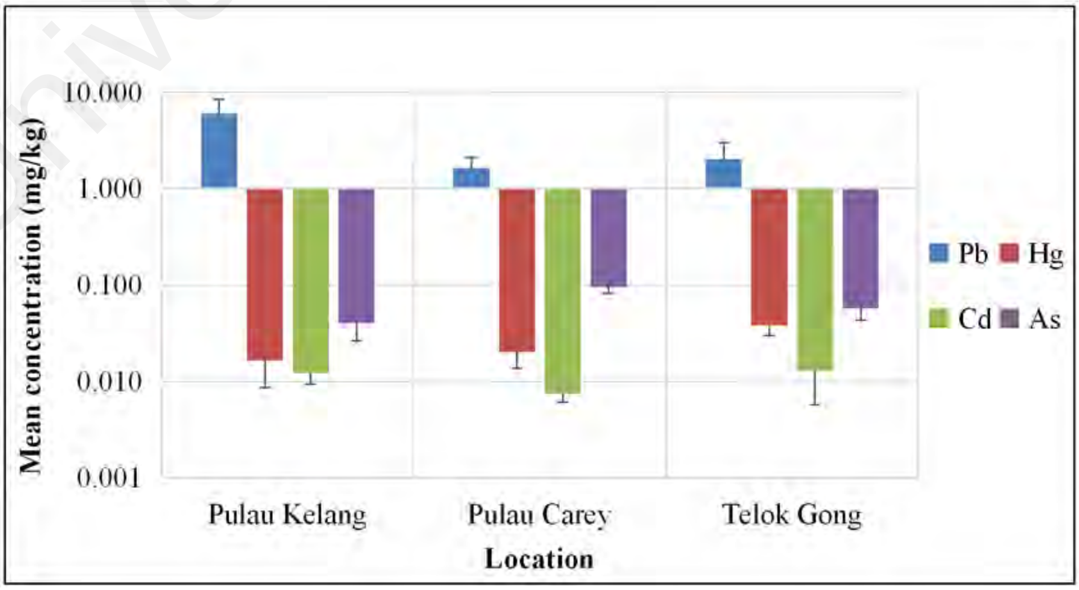


Figure 4.7: Mean concentrations (mg/kg) and standard deviations of heavy metals in fresh branch in three mangroves

Table 4.7: Results of Pos-Hoc tests for heavy metal scores between locations

Element	(I) Location	(J) Location	Mean Difference (I-J)	Std. Error	df	P value	95%Wald Confidence Interval for Difference	
							Lower	Upper
Pb	Pulau Kelang	Pulau Carey	4.383[a]	0.767	1.000	<0.001	2.546	6.220
	Pulau Carey	Telok Gong	-0.384	0.767	1.000	1.000	-2.221	1.453
	Pulau Kelang	Telok Gong	3.999[a]	0.767	1.000	<0.001	2.161	5.836
Hg	Pulau Kelang	Pulau Carey	-0.004	0.004	1.000	1.000	-0.013	0.006
	Pulau Carey	Telok Gong	-0.019[a]	0.004	1.000	<0.001	-0.028	-0.009
	Pulau Kelang	Telok Gong	-0.022[a]	0.004	1.000	<0.001	-0.032	-0.013
Cd	Pulau Kelang	Pulau Carey	0.005	0.005	1.000	0.908	-0.006	0.016
	Pulau Carey	Telok Gong	-0.006	0.005	1.000	0.698	-0.017	0.006
	Pulau Kelang	Telok Gong	-0.0007	0.005	1.000	1.000	-0.012	0.010
As	Pulau Kelang	Pulau Carey	-0.054[a]	0.008	1.000	<0.001	-0.074	-0.034
	Pulau Carey	Telok Gong	0.037[a]	0.008	1.000	<0.001	0.016	0.057
	Pulau Kelang	Telok Gong	-0.017	0.008	1.000	0.121	-0.037	0.003

a. The mean difference is significant at the 0.050 level between sites.

4.3.2.2 Fresh leaves

Owing to the non - normal distribution of this variable, the generalized linear model (GLM) was employed for the analysis. Table 4.8 showed that there were significant differences in the Hg and As scores ($p < 0.010$) between the locations, which indicated that the means of these score changes differed between locations, except for those of Pb and Cd which did not significantly difference between the locations ($p > 0.010$).

Table 4.8: Summary of analysis of generalized linear model for heavy metals

Element	Wald Chi-Square	P value
Pb	3.302	0.192
Hg	13.077	0.001
Cd	4.383	0.112
As	26.652	<0.001

In addition, the Bonferroni post hoc test was conducted to identify the specific means which were different. Figure 4.8 showed the mean concentrations of the heavy metals in the fresh leaves of the three mangroves, which were of the following orders: Pulau Kelang > Telok Gong > Pulau Carey (Pb), Telok Gong > Pulau Carey > Pulau Kelang (Hg), Pulau Kelang > Telok Gong > Pulau Carey (Cd), Telok Gong > Pulau Kelang > Pulau Carey (As). Table 4.9 indicated the differences in significance levels between the individual location points. It showed that the Hg concentration was significantly different between Pulau Carey and Telok Gong mangroves, with the latter being 0.022 mg/kg higher than the former. There was also a significant difference in the Hg concentration between Pulau Kelang and Telok Gong mangroves, with the former being 0.027 mg/kg lower than the latter. In terms of As concentrations, there was a significant difference between Pulau Carey and Telok Gong mangroves, with the former being 0.097 mg/kg lower than the latter. There was also a significant difference between Pulau Kelang and Telok Gong mangroves, with the latter being 0.083 mg/kg higher than the former. As for Pb and Cd, there were no significant differences ($p > 0.050$) between the land use types of the mangroves.

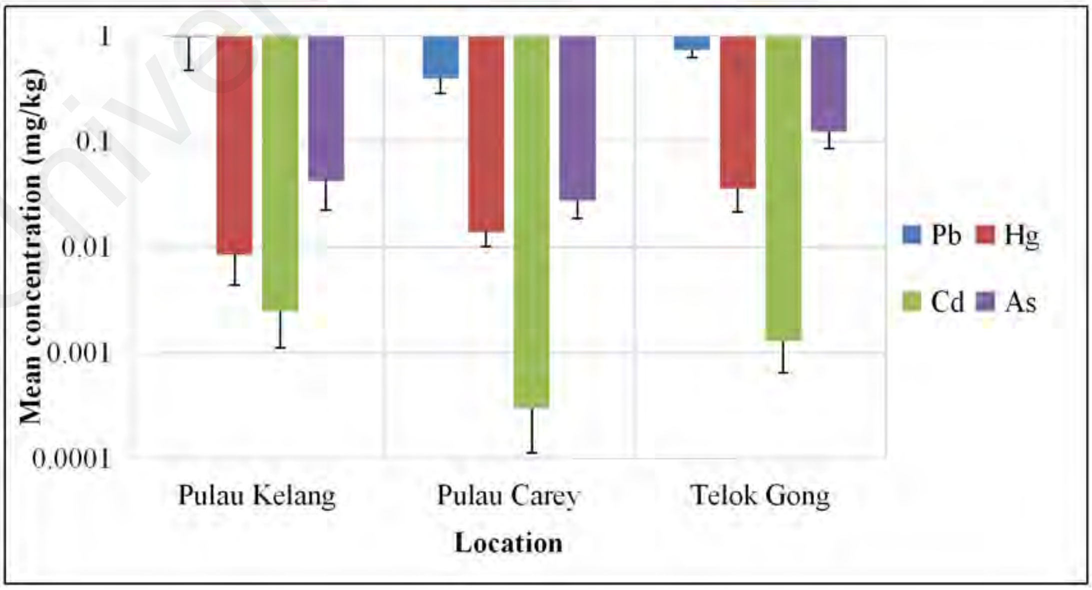


Figure 4.8: Mean concentrations (mg/kg) and standard deviations of heavy metals in fresh leaves in three mangroves

Table 4.9: Results of Pos-Hoc tests for heavy metal scores between locations

Element	(I) Location	(J) Location	Mean Difference (I-J)	Std. Error	df	P value	95%Wald Confidence Interval for Difference	
							Lower	Upper
Pb	Pulau Kelang	Pulau Carey	0.592	0.327	1.000	0.210	-0.190	1.373
	Pulau Carey	Telok Gong	-0.336	0.327	1.000	0.910	-1.118	0.446
	Pulau Kelang	Telok Gong	0.256	0.327	1.000	1.000	-0.526	1.037
Hg	Pulau Kelang	Pulau Carey	-0.005	0.008	1.000	1.000	-0.024	0.014
	Pulau Carey	Telok Gong	-0.022[a]	0.008	1.000	0.018	-0.041	-0.003
	Pulau Kelang	Telok Gong	-0.027[a]	0.008	1.000	0.002	-0.046	-0.008
Cd	Pulau Kelang	Pulau Carey	0.003	0.001	1.000	0.202	-0.001	0.006
	Pulau Carey	Telok Gong	-0.0007	0.001	1.000	0.601	-0.005	0.002
	Pulau Kelang	Telok Gong	0.002	0.001	1.000	1.000	-0.003	0.004
As	Pulau Kelang	Pulau Carey	0.015	0.020	1.000	1.000	-0.034	0.063
	Pulau Carey	Telok Gong	-0.097[a]	0.020	1.000	<0.001	-0.146	-0.048
	Pulau Kelang	Telok Gong	-0.083[a]	0.020	1.000	<0.001	-0.131	-0.034

a. The mean difference is significant at the 0.050 level between sites.

4.3.3 Accumulation coefficient (AC)

As per Table 4.10, the accumulation coefficient of different elements and mangroves were diverse, with all of them being less than 0.500. For the Pulau Kelang mangrove, the value of Pb was highest in the branch, followed by Hg, Cd and As, while the values of four heavy metals in the leaves were in the order of Hg > Pb > Cd > As. For the Pulau Carey and Telok Gong mangroves, the values in the branch and leaves shared the same trend with the leaves of the Pulau Klang mangrove, except that the value of As in the leaves was higher than that of Cd. For all the three mangroves, the weighted average values in the branch and leaves were Hg > Pb > Cd > As and Hg > Pb > As > Cd, respectively. The results showed that all the sampled mangroves had different absorption and taken up abilities according to the type of heavy metal. The weighted average results for the all mangroves showed that the AC values of Hg was the highest as compared to the other three heavy metals, due to its physical property as a semi - volatile metal.

Table 4.10: Accumulation coefficient of heavy metals in mangrove fresh tissues

Site	Tissue	Pb	Hg	Cd	As
Pulau Kelang	Fresh branch	0.353	0.209	0.043	0.006
	Fresh leaves	0.058	0.106	0.009	0.006
Pulau Carey	Fresh branch	0.147	0.321	0.043	0.011
	Fresh leaves	0.036	0.218	0.001	0.003
Telok Gong	Fresh branch	0.114	0.395	0.034	0.005
	Fresh leaves	0.041	0.362	0.003	0.010
Weighted average	Fresh branch	0.205	0.308	0.040	0.007
	Fresh leaves	0.045	0.229	0.004	0.006

4.4 Heavy metal in litter

4.4.1 Heavy metal concentrations

In these three mangroves, the concentrations of heavy metals in litter varied in different fractions (Table 4.11). In general, the concentrations of Pb, Hg, Cd and As were higher in litter branch than in litter leaves, except that the concentrations of Hg in litter leaves was higher than that of litter branch at the Pulau Kelang mangrove. For both branch and leaves, the concentrations of the heavy metals were in the order of Pb > As > Hg > Cd, except that the concentrations of Hg and Cd in litter branch were similar between both Pulau Kelang and Pulau Carey mangroves (Figure 4.9).

Table 4.11: The ranges of heavy metal concentrations (mg/kg) in litter tissues in three mangroves

Site	Tissues	Pb	Hg	Cd	As
Pulau Kelang	Litter branch	0.775-14.855	0.003-0.050	0.001-0.062	0.291-2.005
	Litter leaves	0.906-1.202	0.020-0.083	0.001-0.006	0.060-0.423
Pulau Carey	Litter branch	5.292-10.824	0.009-0.059	0.014-0.046	0.294-0.808
	Litter leaves	1.596-1.786	0.012-0.020	0.003-0.006	0.110-0.432
Telok Gong	Litter branch	1.700-3.375	0.048-0.071	0.005-0.043	0.469-1.663
	Litter leaves	0.689-1.653	0.020-0.048	0.002-0.019	0.228-0.739

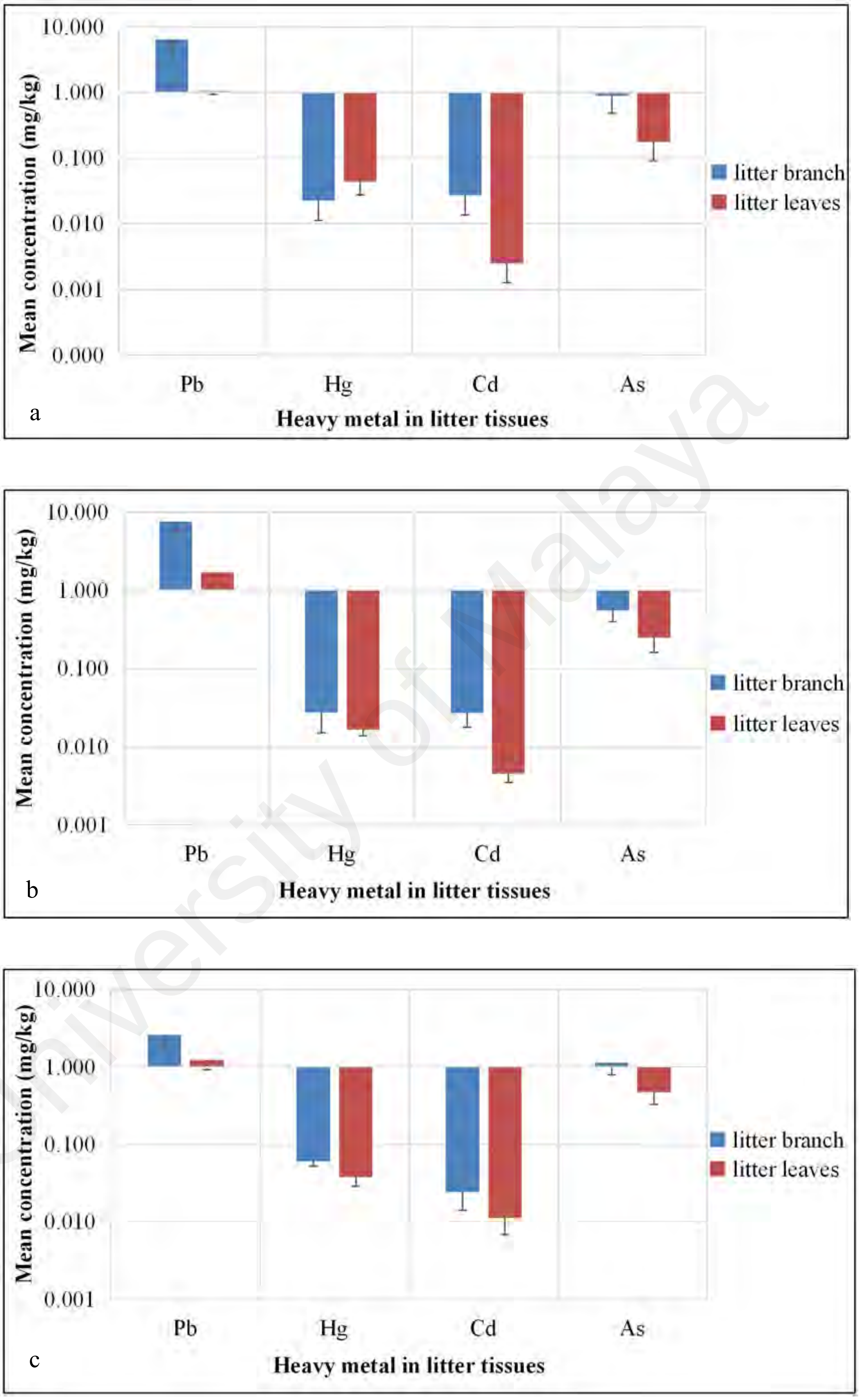


Figure 4.9: The mean concentrations (mg/kg) and standard deviations of heavy metalsin litter tissues in mangroves (a: Pulau Kelang mangrove; b: Pulau Carey mangrove; c: Telok Gong mangrove)

4.4.2 Heavy metal concentrations in different land use types

4.4.2.1 Litter branch

Owing to the non - normal distribution of this variable, the generalized linear model (GLM) was employed in the analysis. Table 4.12 showed that there were significant differences in the heavy metal scores ($p < 0.050$) between the locations, which indicated that the means of the heavy metal score changes differed between locations, except that of Cd that did not significantly difference between locations ($p > 0.050$).

Table 4.12: Summary of analysis of generalized linear model for heavy metals

Element	Wald Chi-Square	P value
Pb	7.953	0.019
Hg	15.892	<0.001
Cd	0.087	0.957
As	9.326	0.009

In addition, the Bonferroni post hoc test was conducted to identify the specific means which were different. Figure 4.10 showed the mean concentrations of heavy metals in the litter branch of the three mangroves, which were of the following orders: Pulau Carey > Pulau Kelang > Telok Gong (Pb), Telok Gong > Pulau Carey > Pulau Kelang (Hg), Pulau Kelang = Pulau Carey > Telok Gong (Cd), Telok Gong > Pulau Kelang > Pulau Carey (As). Table 4.13 indicated the differences in significance levels between the individual location points. It showed that Pb concentrations were significantly difference between Pulau Carey and Telok Gong mangroves, with the former being 4.983 mg/kg higher than the latter. For the Hg concentrations, there were significant difference between Pulau Carey and Telok Gong mangroves, with the former being 0.034 mg/kg lower than the latter, and also there were significant difference between Pulau Kelang and Telok Gong mangroves, with the former being 0.039 mg/kg lower than the latter. For the Cd concentrations, there was no significant difference between any of the two mangroves ($p > 0.050$). In terms of the As concentrations, there were significant difference between Pulau Carey and Telok Gong mangroves, with the former being 0.578 mg/kg lower than the latter.

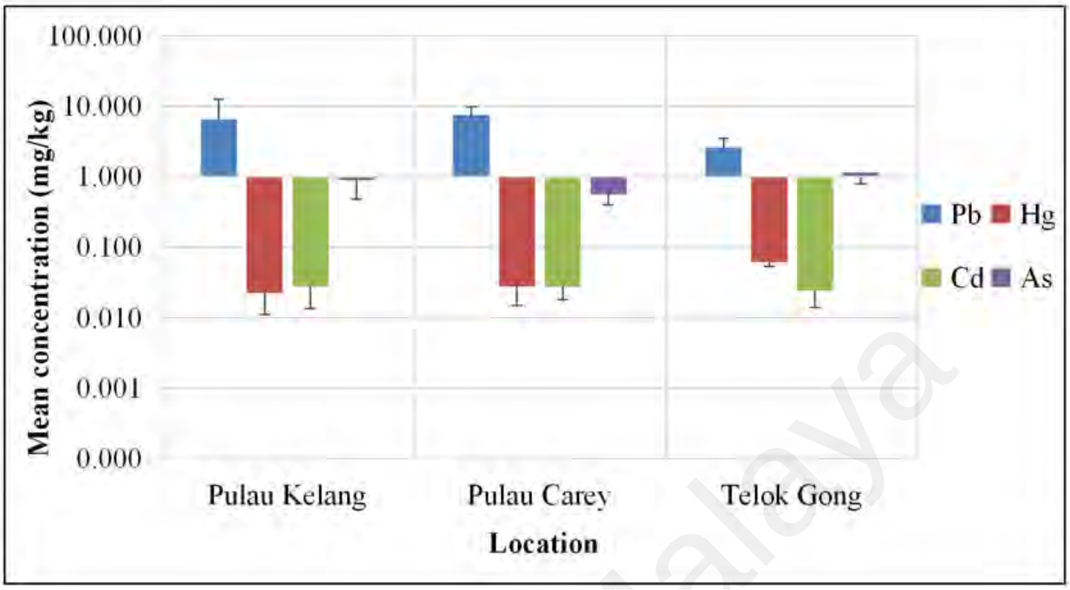


Figure 4.10: Mean concentrations (mg/kg) and standard deviations of heavy metals in litter branch in three mangroves

Table 4.13: Results of Pos-Hoc tests for heavy metal scores between locations

Element	(I) Location	(J) Location	Mean Difference (I-J)	Std. Error	df	P value	95%Wald Confidence Interval for Difference	
							Lower	Upper
Pb	Pulau Kelang	Pulau Carey	-1.071	1.860	1.000	1.000	-5.524	3.382
	Pulau Carey	Telok Gong	4.983[a]	1.860	1.000	0.022	0.530	9.435
	Pulau Kelang	Telok Gong	3.911	1.860	1.000	0.106	-0.542	8.364
Hg	Pulau Kelang	Pulau Carey	-0.005	0.011	1.000	1.000	-0.030	0.020
	Pulau Carey	Telok Gong	-0.034[a]	0.011	1.000	0.004	-0.059	-0.008
	Pulau Kelang	Telok Gong	-0.039[a]	0.011	1.000	0.001	-0.064	-0.013
Cd	Pulau Kelang	Pulau Carey	0.000	0.011	1.000	1.000	-0.026	0.026
	Pulau Carey	Telok Gong	0.003	0.011	1.000	1.000	-0.023	0.028
	Pulau Kelang	Telok Gong	0.003	0.011	1.000	1.000	-0.023	0.028
As	Pulau Kelang	Pulau Carey	0.337	0.190	1.000	0.229	-0.118	0.793
	Pulau Carey	Telok Gong	-0.578[a]	0.190	1.000	0.007	-1.034	-0.123
	Pulau Kelang	Telok Gong	-0.241	0.190	1.000	0.616	-0.696	0.214

a. The mean difference is significant at the 0.050 level between sites.

4.4.2.2 Litter leaves

Owing to the non - normal distribution of this variable, the generalized linear model (GLM) was employed for the analysis. Table 4.14 showed that there were significant differences in the heavy metal scores ($p < 0.010$) between the locations, which indicated that the means of heavy metal scores changes differed between the locations.

Table 4.14: Summary of analysis of generalized linear model for heavy metals

Element	Wald Chi-Square	P value
Pb	26.841	<0.001
Hg	14.129	0.001
Cd	17.938	<0.001
As	10.669	0.005

In addition, the Bonferroni post hoc test was conducted to identify specific means were differed. Figure 4.11 showed the mean concentrations of heavy metals in litter leaves of the three mangroves. For the following three mangroves, the concentrations declined with Pulau Carey > Telok Gong > Pulau Kelang (Pb), Pulau Kelang > Telok Gong > Pulau Carey (Hg), Telok Gong > Pulau Carey > Pulau Kelang (Cd), Telok Gong > Pulau Carey > Pulau Kelang (As). Table 4.15 indicated the significance level for the differences between the individual location points. It showed that Pb concentrations were significantly different between Pulau Kelang and Pulau Carey mangroves, with the former being 0.645 mg/kg lower than the latter, and also there was a significant difference between Pulau Carey and Telok Gong mangroves, with the former being 0.462 mg/kg higher than the latter. As for the Hg concentrations were significant difference between Pulau Kelang and Pulau Carey mangroves, with the former being 0.028 mg/kg higher than the latter. There was also a significant difference between Pulau Carey and Telok Gong mangroves, with the former being 0.022 mg/kg lower than the latter. With reference to the Cd concentrations, there were significant difference between Pulau Carey and Telok Gong mangroves, with the former being 0.007 mg/kg lower than the latter, also there was a significant difference between Pulau

Kelang and Telok Gong mangroves, with the former being 0.009 mg/kg lower than the latter. In terms of the As concentrations, there was only a significant difference between Pulau Kelang and Telok Gong mangroves, with the former being 0.295 mg/kg lower than the latter.

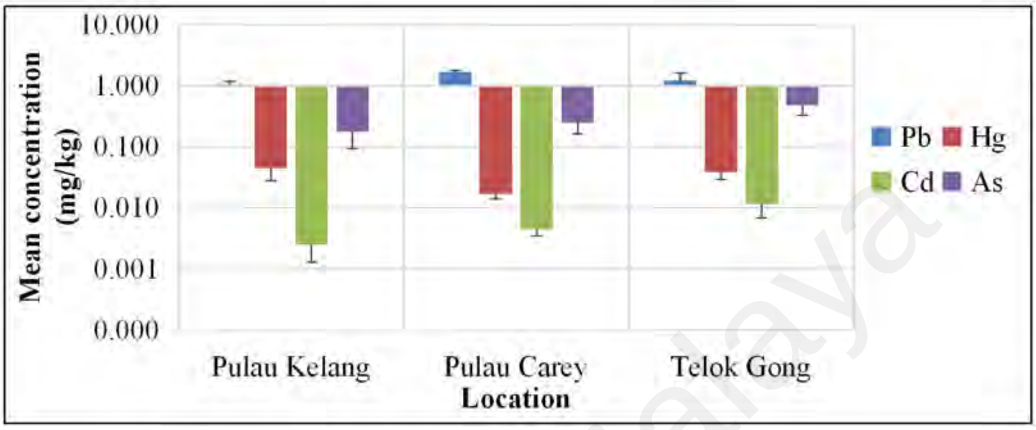


Figure 4.11: Mean concentrations (mg/kg) and standard deviations of heavy metals in litter leaves in three mangroves

Table 4.15: Results of Pos-Hoc tests for heavy metal scores between locations

Element	(I) Location	(J) Location	Mean Difference (I-J)	Std. Error	df	P value	95%Wald Confidence Interval for Difference	
							Lower	Upper
Pb	Pulau Kelang	Pulau Carey	-0.645[a]	0.128	1.000	<0.001	-0.953	-0.338
	Pulau Carey	Telok Gong	0.462[a]	0.128	1.000	0.001	0.154	0.769
	Pulau Kelang	Telok Gong	-0.184	0.128	1.000	0.458	-0.491	0.124
Hg	Pulau Kelang	Pulau Carey	0.028[a]	0.008	1.000	0.001	0.009	0.046
	Pulau Carey	Telok Gong	-0.022[a]	0.008	1.000	0.014	-0.040	-0.003
	Pulau Kelang	Telok Gong	0.006	0.008	1.000	1.000	-0.013	0.024
Cd	Pulau Kelang	Pulau Carey	-0.002	0.002	1.000	1.000	-0.007	0.003
	Pulau Carey	Telok Gong	-0.007[a]	0.002	1.000	0.005	-0.012	-0.002
	Pulau Kelang	Telok Gong	-0.009[a]	0.002	1.000	<0.001	-0.014	-0.004
As	Pulau Kelang	Pulau Carey	-0.071	0.094	1.000	1.000	-0.296	0.155
	Pulau Carey	Telok Gong	-0.224	0.094	1.000	0.052	-0.449	0.002
	Pulau Kelang	Telok Gong	-0.295[a]	0.094	1.000	0.005	-0.520	-0.069

a. The mean difference is significant at the 0.050 level between sites.

4.4.3 Accumulation coefficient (AC)

As per Table 4.16, the accumulation coefficient of different elements and mangroves were diverse, with all of them being less than 1. For the Pulau Kelang mangrove, the value of Pb was the highest in the branch, followed by Hg, As and Cd, while the values of the four heavy metals in the leaves were in the order of Hg > Pb > As > Cd. For the Pulau Carey mangrove, the values of the heavy metals in the branch were in the order of Pb > Hg > Cd > As, while that in the leaves was Hg > Pb > As > Cd. For the Telok Gong mangrove, the accumulation coefficients of litter branch and litter leaves shared same trend, that was Hg > Pb > As > Cd. For all the three mangroves, the values in the branch and in leaves were in the sequence of Hg > Pb > Cd > As and Hg > Pb > As > Cd, respectively. Among the metals, Hg was the easiest to be absorbed.

Table 4.16: Accumulation coefficient of heavy metals in mangrove litter fraction

Site	Tissue	Pb	Hg	Cd	As
Pulau Kelang	Litter branch	0.381	0.278	0.094	0.138
	Litter leaves	0.061	0.550	0.009	0.027
Pulau Carey	Litter branch	0.680	0.431	0.154	0.067
	Litter leaves	0.152	0.261	0.026	0.030
Telok Gong	Litter branch	0.146	0.618	0.064	0.090
	Litter leaves	0.069	0.389	0.030	0.037
Weighted average	Litter branch	0.402	0.442	0.104	0.098
	Litter leaves	0.094	0.400	0.022	0.031

4.5 Comparison of heavy metal concentrations in mangrove fraction

The mean concentrations of the heavy metals in the sediments, fresh branch, fresh leaves, liter branch and litter leaves in three mangrove forests are shown in Figure 4.12, Figure 4.13 and Figure 4.14. For all the three mangroves, the concentrations of the heavy metals were highest in the sediments, while there were various sequences in tissues among different heavy metal element. In general, it can be illustrated that the concentrations of heavy metals in litter tissues were higher than those in fresh tissues. At the Pulau Kelang mangrove, the contents of Pb, Hg, Cd and As in litter branch were higher by 7.4%, 24.7%, 54.6% and 95.5% respectively as compared to those in fresh branch. The concentrations of Pb, Hg and As in litter leaves were higher by 5.6%, 80.7% and 76.3% respectively as compared to those in fresh leaves. However, the concentration of Cd in litter leaves and fresh leaves shared the same value. At the Pulau Carey mangrove, the concentrations of Pb, Hg, Cd and As in litter branch were higher by 78.4%, 25.7%, 72.2% and 83% respectively as compared to those in fresh branch. The aforementioned concentrations in litter leaves were also higher by 76.6%, 16.7%, 94.4% and 88.9% respectively as compared to those in fresh leaves. The same trend was also seen at the Telok Gong mangrove, with the said concentrations litter branch being higher than those of fresh branch by 21.9%, 36.2%, 1.1% and 94.9% respectively. Meanwhile, the concentrations in litter leaves were higher than those in fresh leaves by 40.5%, 7.2%, 88.9% and 73.6% respectively.

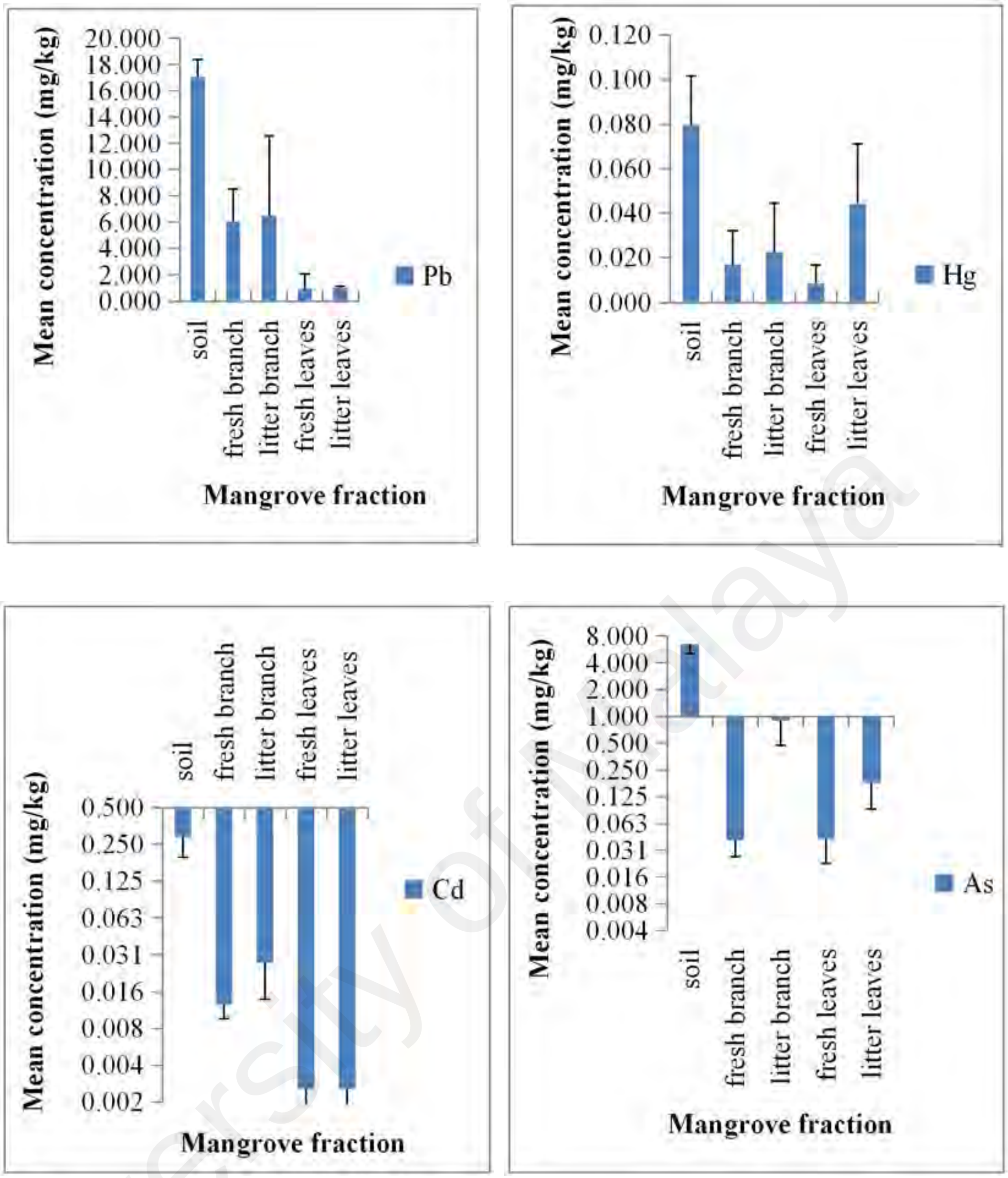


Figure 4.12: Mean concentrations (mg/kg) and standard deviations of heavy metals in Pulau Kelang mangrove

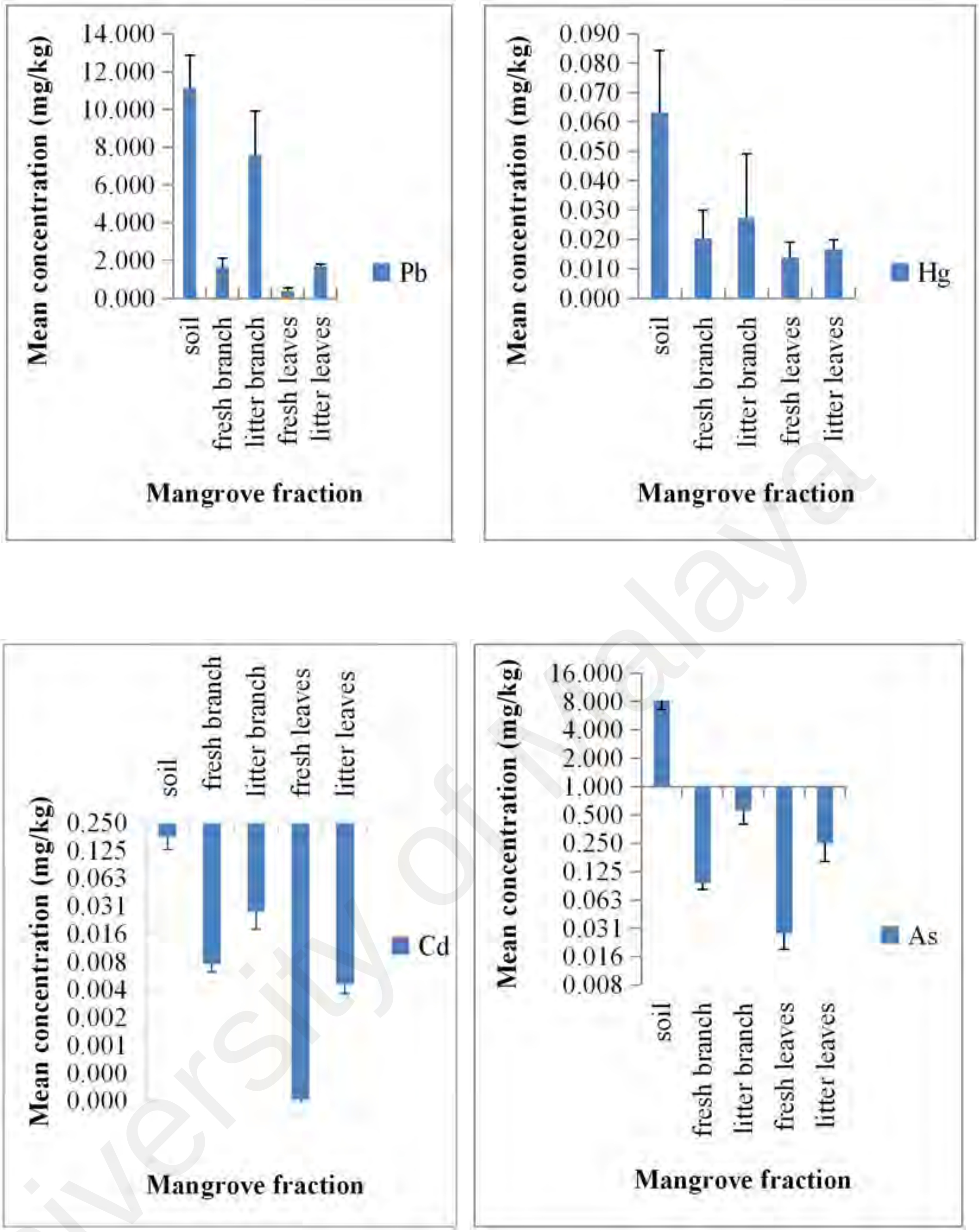


Figure 4.13: Mean concentrations (mg/kg) and standard deviations of heavy metals in Pulau Carey mangrove

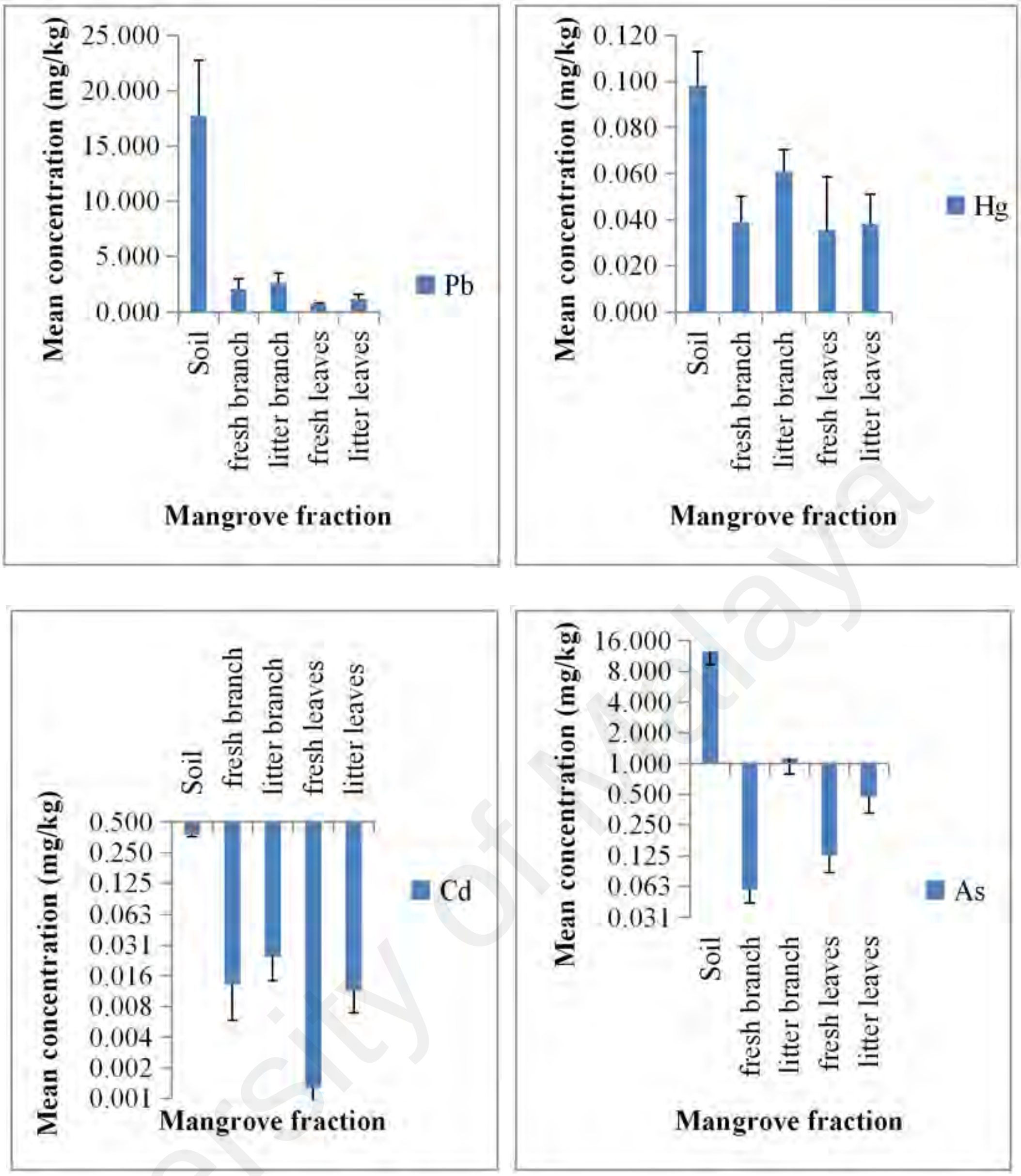


Figure 4.14: Mean concentrations (mg/kg) and standard deviations of heavy metals in Telok Gong mangrove

4.6 Accumulation and cycle of heavy metals in mangrove

The biological cycle in an ecosystem refers to the circulation of materials among soil, plants and animals. This study only deals with the part of the cycle between the soil and plants that includes 3 processes: uptake, retention and return. The concentrations of heavy metals varied between different fractions of the mangroves (Table 4.17, Figure 4.15). In general, the concentrations of the heavy metals followed the pattern of sediments > litter tissues > fresh tissues in all the sampled mangroves. Mangrove sediment have a large capacity to retain heavy metals. The average concentrations of the heavy metals (Pb, Hg, Cd and As) in the soil which were taken up by the plants at the three mangroves were about 12.8%, 27.5%, 2.1% and 0.7% respectively, then falling on the surface of soil. The percentages of these four heavy metals in the litter tissues were higher than those of the fresh tissues by 42.8%, 37.1%, 62.5% and 88.9% respectively.

Table 4.17: The mean concentrations (mg/kg) and standard deviations of heavy metals in fractions

Site	Fraction	Pb	Hg	Cd	As
Pulau Kelang	Sediment	17.066±1.313	0.080±0.022	0.288±0.132	6.470±1.766
	Fresh tissues	3.503±1.016	0.013±0.017	0.007±0.004	0.041±0.061
	Litter tissues	3.772±4.630	0.033±0.022	0.015±0.016	0.534±0.401
Pulau Carey	Sediment	11.126±1.749	0.063±0.021	0.175±0.066	8.218±2.010
	Fresh tissues	1.016±0.878	0.017±0.005	0.004±0.005	0.061±0.047
	Litter tissues	4.630±4.1566	0.022±0.008	0.016±0.016	0.401±0.217
Telok Gong	Sediment	17.750±5.007	0.098±0.015	0.378±0.021	12.588±4.447
	Fresh tissues	1.376±0.912	0.037±0.002	0.007±0.008	0.091±0.047
	Litter tissues	1.908±0.960	0.045±0.016	0.018±0.009	0.802±0.467
Average	Sediment	15.314±3.643	0.080±0.018	0.280±0.102	9.092±3.151
	Fresh tissues	1.965±1.344	0.022±0.013	0.006±0.002	0.064±0.025
	Litter tissues	3.436±1.392	0.035±0.014	0.016±0.002	0.579±0.204

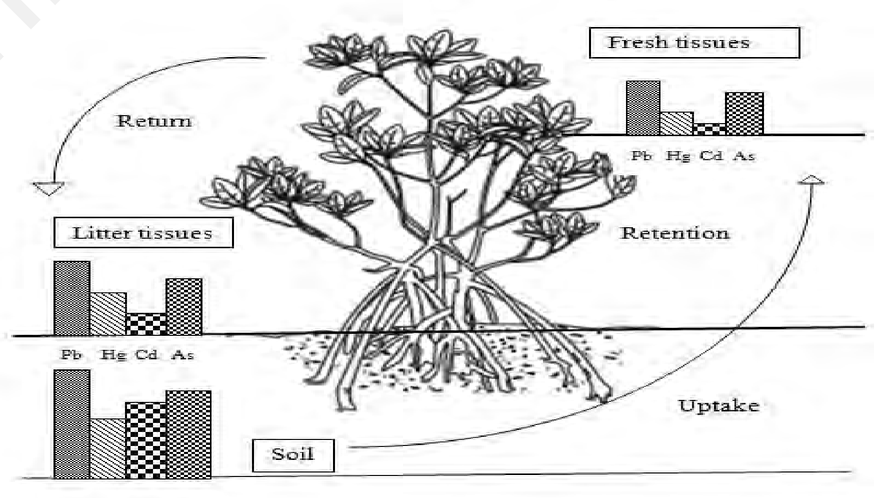


Figure 4.15: Accumulation and cycle of heavy metals (mg/kg) in three mangroves

4.7 Correlations of heavy metal concentrations in sediments and different plant tissues at three mangrove forests

The assessment of the potential uses of the different tissues of mangroves, including branch and leaves, as a materials for the bio - monitoring of Pb, Hg, Cd and As was analyzed using Pearson correlation. The relationships between heavy metal concentrations in different tissues and sediments are presented in Table 4.18, Table 4.19, Table 4. 20 and Table 4.21. The results show that the concentrations of all heavy metals in the tissues were not significantly correlated with those in the sediments ($p > 0.050$) of the Pulau Kelang mangrove (Table 4.18). The correlations between the concentrations of Pb, Hg, Cd and As in the sediments and those in the branch and leaves were not significant ($P > 0.050$), except for the concentrations of Hg and Cd in litter branch which were significantly correlated with those in the sediments ($P < 0.050$) of the Pulau Carey mangrove (Table 4.19). In addition, the concentration of Cd in litter leaves was significantly correlated with that in the sediment ($P < 0.050$) of the Telok Gong mangrove (Table 4.20). The results for all the three mangroves shows that As concentration in the fresh leaves was significantly correlated with that in the sediment ($P < 0.050$), and that the Pb concentration in the litter leaves was also significantly correlated with that in the sediment ($P < 0.050$) (Table 4.21).

Table 4.18: Correlations of heavy metals between sediment and different tissues in Pulau Kelang mangrove

Site			Pb	Hg	Cd	As
Pulau Kelang	Sediment	Fresh branch	R=0.233 P=0.767	R=0.417 P=0.583	R=-0.103 P=0.897	R=0.416 P=0.584
	Sediment	Fresh leaves	R=0.728 P=0.272	R=-0.380 P=0.620	R=-0.523 P=0.477	R=0.031 P=0.969
	Sediment	Litter branch	R=-0.684 P=0.316	R=-0.801 P=0.199	R=-0.109 P=0.891	R=-0.612 P=0.388
	Sediment	Litter leaves	R=0.159 P=0.841	R=-0.078 P=0.922	R=0.641 P=0.359	R=-0.159 P=0.841

Levels of significance are indicated as $p<0.050$.

Table 4.19: Correlations of heavy metals between sediment and different tissues in Pulau Carey mangrove

Site			Pb	Hg	Cd	As
Pulau Carey	Sediment	Fresh branch	R=-0.553 P=0.447	R=0.390 P=0.610	R=-0.786 P=0.214	R=-0.151 P=0.849
	Sediment	Fresh leaves	R=0.789 P=0.202	R=0.355 P=0.645	R=0.454 P=0.546	R=-0.902 P=0.098
	Sediment	Litter branch	R=0.813 P=0.187	R=0.958 P=0.042	R=-0.975 P=0.025	R=-0.376 P=0.624
	Sediment	Litter leaves	R=0.394 P=0.606	R=0.385 P=0.615	R=-0.742 P=0.258	R=0.633 P=0.367

Levels of significance are indicated as p<0.050.

Table 4.20: Correlations of heavy metals between sediment and different tissues in Telok Gong mangrove

Site			Pb	Hg	Cd	As
Telok Gong	Sediment	Fresh branch	R=0.411 P=0.589	R=-0.868 P=0.132	R=0.513 P=0.487	R=0.502 P=0.498
	Sediment	Fresh leaves	R=0.251 P=0.749	R=0.918 P=0.082	R=-0.610 P=0.390	R=0.763 P=0.237
	Sediment	Litter branch	R=0.709 P=0.291	R=0.702 P=0.298	R=-0.883 P=0.117	R=0.879 P=0.121
	Sediment	Litter leaves	R=-0.667 P=0.333	R=0.513 P=0.487	R=-0.955 P=0.045	R=0.041 P=0.959

Levels of significance are indicated as p<0.050.

Table 4.21: Correlations of heavy metals between sediment and different tissues in three mangroves

		Pb	Hg	Cd	As
Sediment	Fresh branch	R=0.355 P=0.258	R=0.409 P=0.186	R=0.202 P=0.529	R=0.178 P=0.579
Sediment	Fresh leaves	R=0.404 P=0.193	R=0.533 P=0.074	R=-0.074 P=0.820	R=0.714 P=0.009
Sediment	Litter branch	R=-0.279 P=0.380	R=0.457 P=0.135	R=-0.254 P=0.425	R=0.316 P=0.317
Sediment	Litter leaves	R=-0.760 P=0.004	R=0.332 P=0.292	R=0.350 P=0.264	R=0.509 P=0.091

Levels of significance are indicated as p<0.050.

4.8 Comparison of heavy metals between mangrove fresh tissues and litter

4.8.1 Lead

In Table 4.22, the ranges of Pb concentrations in fresh branch, litter branch, fresh leaves and litter leaves were 1.235 - 8.714, 0.775 - 14.855, 0.182 - 2.548 and 0.689 - 1.786 mg/kg respectively. Figure 4.16 shows the mean concentrations of Pb in different tissues, which were in the order of litter branch > fresh branch and litter leaves > fresh leaves. The concentrations of Pb in litter branch and leaves were higher than those in the fresh tissues by 41.9% and 46.7% respectively. For fresh tissues, the concentration of Pb in the branch was higher than that in the leaves, which mean Pb was enrich in branch. The results of the non - parametric analysis showed that the concentrations of Pb in fresh leaves and fresh branch were significant, which mean their existence in fresh leaves and litter branch (Table 4.23).

Table 4.22: Concentrations of Pb (mg/kg) in four sampled tissues in three mangroves

Tissues	Minimum	Maximum	Mean	Std. Deviation
Fresh branch	1.235	8.714	3.226	2.508
Litter branch	0.775	14.855	5.551	4.095
Fresh leaves	0.182	2.548	0.705	0.617
Litter leaves	0.689	1.786	1.322	0.363

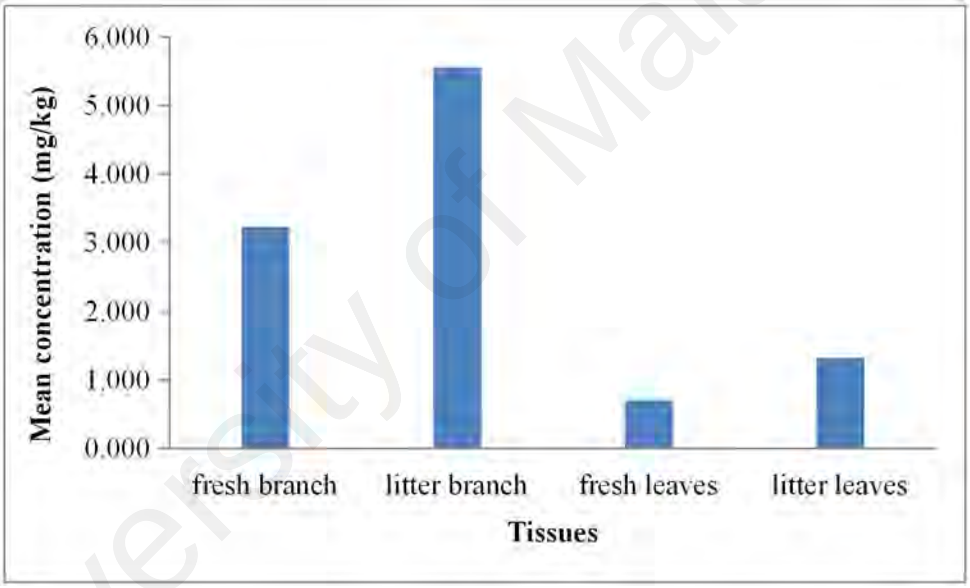


Figure 4.16: The mean concentrations (mg/kg) of Pb in four sampled tissues in three mangroves

Table 4.23: The non-parametric test of Pb in four sampled tissues in three mangroves

Sample1	Sample2	Test Statistic	Std. Error	Std. Test Statistic	Sig.	Adj.Sig.
Fresh leaves	Litter leaves	-1.000	0.527	-1.897	0.058	0.347
Fresh leaves	Fresh branch	1.750	0.527	3.320	0.001	0.005
Fresh leaves	Litter branch	2.250	0.527	4.269	<0.001	<0.001
Litter leaves	Fresh branch	0.750	0.527	1.423	0.155	0.928
Litter leaves	Litter branch	1.250	0.527	2.372	0.018	0.106
Fresh branch	Litter branch	-0.500	0.527	-0.949	0.343	1.000

The significance level is 0.050.

4.8.2 Mercury

In Table 4.24, the ranges of Hg concentrations in fresh branch, litter branch, fresh leaves and litter leaves were 0.000 - 0.053, 0.003 - 0.071, 0.002 - 0.064 and 0.012 - 0.083 mg/kg respectively. Figure 4.17 shows the mean concentrations of Hg in different tissues, which were in the order of litter branch > fresh branch and litter leaves > fresh leaves. The concentrations of Hg in litter branch and leaves were higher than those in the fresh tissues by 30.6% and 42.4% respectively. For fresh tissues, the concentration of Hg in the branch was higher than that in the leaves, which meant that Hg was enrich in branch. The results of the non - parametric analysis showed that the concentrations of Hg in fresh leaves and litter branch were significant (Table 4.25).

Table 4.24: Concentrations of Hg (mg/kg) in four sampled tissues in three mangroves

Tissues	Minimum	Maximum	Mean	Std. Deviation
Fresh branch	0.000	0.053	0.025	0.015
Litter branch	0.003	0.071	0.036	0.025
Fresh leaves	0.002	0.064	0.019	0.018
Litter leaves	0.012	0.083	0.033	0.020

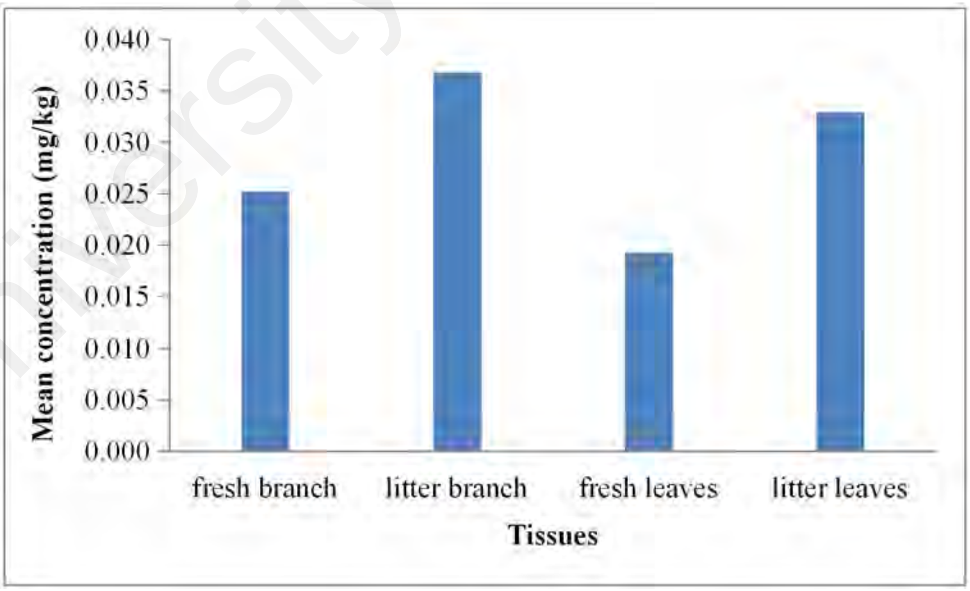


Figure 4.17: The mean concentrations (mg/kg) of Hg in four sampled tissues in three mangroves

Table 4.25: The non-parametric test of Hg in four sampled tissues in three mangroves

Sample1	Sample2	Test Statistic	Std. Error	Std.Test Statistic	Sig.	Adj.Sig.
Fresh leaves	Fresh branch	0.917	0.527	1.739	0.082	0.492
Fresh leaves	Litter leaves	-1.083	0.527	-2.055	0.040	0.239
Fresh leaves	Litter branch	1.667	0.527	3.162	0.002	0.009
Fresh branch	Litter leaves	-0.167	0.527	-0.316	0.752	1.000
Fresh branch	Litter branch	-0.750	0.527	-1.423	0.155	0.928
Litter leaves	Litter branch	0.583	0.527	1.107	0.268	1.000

The significance level is 0.050.

4.8.3 Cadmium

In Table 4.26, the ranges of Cd concentrations in fresh branch, litter branch, fresh leaves and litter leaves were 0.000 - 0.037, 0.001 - 0.062, 0.000 - 0.007 and 0.001 - 0.019 mg/kg, respectively. Figure 4.18 shows the mean concentrations of Cd in different tissues, which were in the order of litter branch > fresh branch and litter leaves > fresh leaves. The concentrations of Cd in litter branch and leaves were higher than those in the fresh tissues by 57.7% and 83.3% respectively. For fresh tissues, the concentration of Cd in the branch was higher than that in the leaves, which meant that Cd was enrich in branch.The results of the non - parametric analysis showed that the concentrations of Cd in fresh leaves and fresh branch were significant. At the same time, the results were also significant between fresh leaves and litter branch, as well as between litter leaves and litter branch (Table 4.27).

Table 4.26: Concentrations of Cd (mg/kg) in four sampled tissues in three mangroves

Tissues	Minimum	Maximum	Mean	Std. Deviation
Fresh branch	0.000	0.037	0.011	0.009
Litter branch	0.001	0.062	0.026	0.018
Fresh leaves	0.000	0.007	0.001	0.002
Litter leaves	0.001	0.019	0.006	0.006

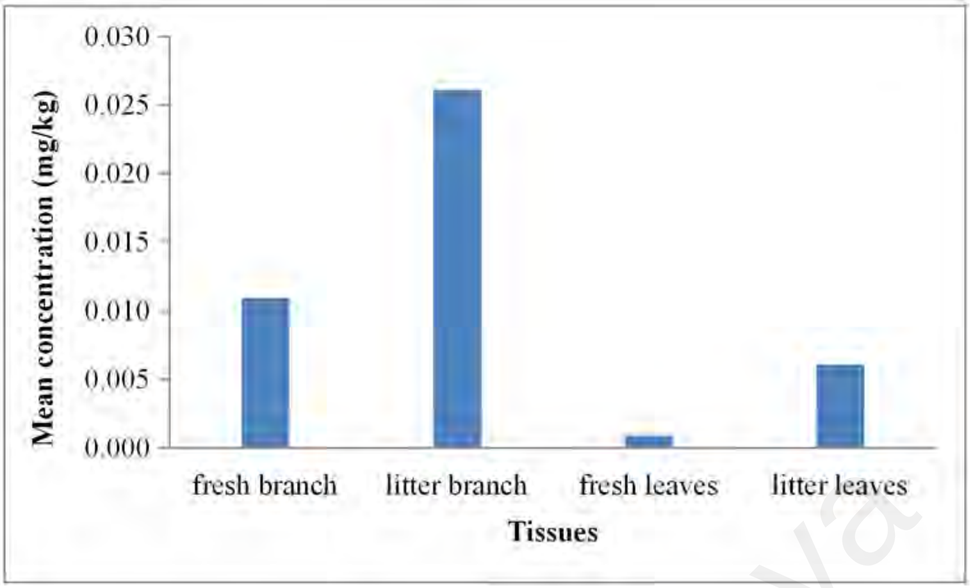


Figure 4.18: The mean concentrations (mg/kg) of Cd in four sampled tissues in three mangroves

Table 4.27: The non-parametric test of Cd in four sampled tissues in three mangroves

Sample1	**Sample2**	**Test Statistic**	**Std. Error**	**Std.Test Statistic**	**Sig.**	**Adj.Sig**
Fresh leaves	Litter leaves	-0.958	0.527	-1.818	0.069	0.414
Fresh leaves	Fresh branch	1.667	0.527	3.162	0.002	0.009
Fresh leaves	Litter branch	2.375	0.527	4.506	<0.001	<0.001
Litter leaves	Fresh branch	0.708	0.527	1.344	0.179	1.000
Litter leaves	Litter branch	1.417	0.527	2.688	0.007	0.043
Fresh branch	Litter branch	-0.708	0.527	-1.344	0.179	1.000

The significance level is 0.050.

4.8.4 Arsenic

In Table 4.28, the ranges of As concentrations in fresh branch, litter branch, fresh leaves and litter leaves were 0.021 - 0.109, 0.291 - 2.005, 0.008 - 0.194 and 0.060 - 0.739 mg/kg respectively. Figure 4.19 shows the mean concentrations of As in different tissues, which were in the order of litter branch > fresh branch and litter leaves > fresh leaves. The concentrations of As in litter branch and leaves were higher than those in the fresh tissues by 92.5% and 78.3% respectively. For fresh tissues, the concentration of As in the leaves was slightly higher than that in the branch, although both values were similar. The results of the non - parametric analysis showed that the difference in concentrations of As in fresh tissues and litter tissues were significant (Table 4.29).

Table 4.28: Concentrations of As (mg/kg) in four sampled tissues in three mangroves

Tissues	Minimum	Maximum	Mean	Std. Deviation
Fresh branch	0.021	0.109	0.064	0.029
Litter branch	0.291	2.005	0.859	0.555
Fresh leaves	0.008	0.194	0.065	0.058
Litter leaves	0.060	0.739	0.299	0.204

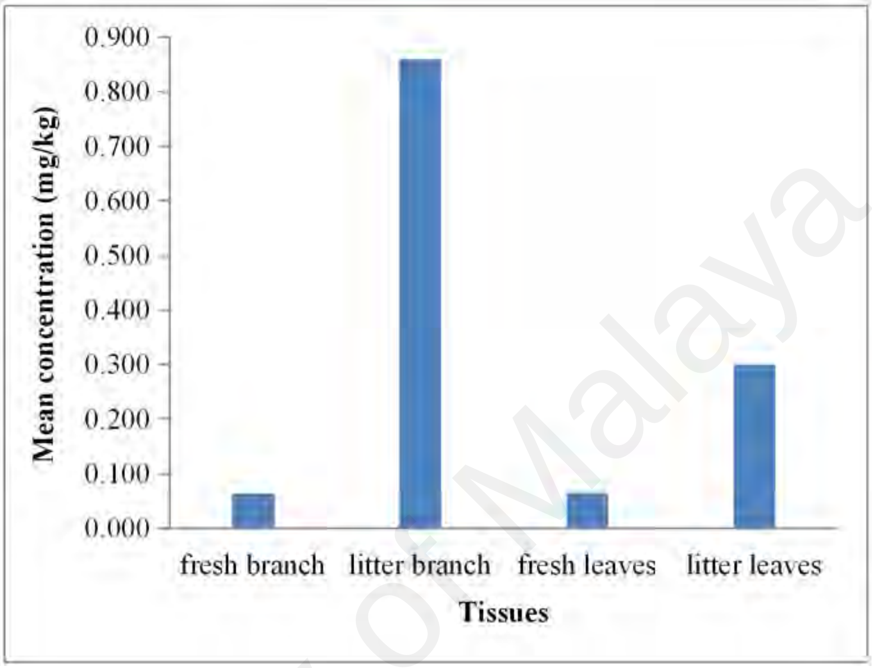


Figure 4.19: The mean concentrations (mg/kg) of As in four sampled tissues in three mangroves

Table 4.29: The non-parametric test of As in four sampled tissues in three mangroves

Sample1	Sample2	Test Statistic	Std. Error	Std. Test Statistic	Sig.	Adj.Sig.
Fresh branch	Fresh leaves	0.000	0.527	0.000	1.000	1.000
Fresh branch	Litter leaves	-1.583	0.527	-3.004	0.003	0.016
Fresh branch	Litter branch	-2.417	0.527	-4.585	<0.001	<0.001
Fresh leaves	Litter leaves	-1.583	0.527	-3.004	0.003	0.016
Fresh leaves	Litter branch	2.417	0.527	4.585	<0.001	<0.001
Litter leaves	Litter branch	0.833	0.527	1.581	0.114	0.683

The significance level is 0.050.

CHAPTER 5: DISCUSSION

5.1 Heavy metal concentrations in sediments

Marine sediments are always regarded as the final sink for numerous pollutants, including trace metals. Trace metals in sediments may pose a hazard to deposit feeders through their direct digestion or release into the aquatic environment. Many metals are essential elements which are potentially toxic only if their values exceed a certain threshold level. However, there are non - essential elements (such as Pb, Hg, Cd and As) that are toxic even at low levels. In this study, the results showed that the concentrations of heavy metals in sediments had positive or negative relationship with the tidal gradient (from seaward to landward) in all sampled mangroves, but the differences were not significant. Similar findings were reported by Soto - Jiménez and Páez - Osuna (2001), who stated that heavy metals (Al, Fe, Ni, Pb and Cd) had strong or weak concentration gradients reducing from the upper lagoon of the Mazaltlan Harbor. Tam and Wong (2000) found that heavy metal contamination occurred at the hot spots within each swamp. Previously, Tam and Wong (1996) also reported that Cu concentrations in the sediments fluctuated within the range of 1 - 42 mg/kg from seaward to landward regions at the Sai Keng mangrove.

This study used the New York Sediment Criteria and Provincial Sediment Quality Guidelines to predict the adverse biological effects on the contaminated sediments. In Figure 5.1, the mean concentrations of Pb and As in the sediments of the three mangrove were lower than the back ground values (39.800 mg/kg and 18.790 mg/kg, respectively). Meanwhile, the mean concentration of Hg in Telok Gong was higher than the back ground value (0.080 mg/kg), while the concentrations in Pulau Kelang is equal with the back ground value (0.080 mg/kg). The mean concentrations of Cd in Pulau Kelang and Telok Gong were higher than the back ground value (0.186 mg/kg). According to the New York Sediment Criteria and the Provincial Sediment Quality

Guidelines, Pb, Hg and Cd in the sediments of the three mangroves were below the lowest effects range in both guidelines which showed that the levels of these three elements were safe for organisms. The levels of As in sediments of the three mangroves were higher than the lowest effects range and lower than high effects range, which showed that this element could probably have adverse effects on organisms. As these three mangroves are located at Port Klang, Pb, Hg, Cd and As pollutions may due to industrial activities, like combustion of fossil fuels, production of paper and glass, manufacturing of cement, and mining. The metals which are released into Port Klang also originate from the wastes of the palm oil industry, electrical based power generation, and cement manufacturing. In addition, these heavy metals might be from atmospheric depositions, and terrestrial runoffs, which are the primary routes to the marine environment. Moreover, other sources of contamination may be from organic insecticides (lead - arsenate), pesticides, and fertilizers which are applied in agricultural activities. Sany et al. (2013) reported that the concentrations of Pb, Hg, Cd and As were higher than the normal concentrations in the sediments of Port Klang, which can be regarded as a serious threat to the health of marine organisms and humans , especially the concentration of As that was significantly higher than value of PEL (effect range medium). Moreover, the concentrations of these four heavy metals were also studied at West Port in Malaysia, whereby the As value was considered to be able to result in adverse effects on the sediment organisms (Sany et al., 2011). Based on the I_{geo} index, all the sampled mangroves in this study were considered as uncontaminated to moderately contaminated (Class 1) by these four trace heavy metals. However, Sany et al. (2013) reported that the potential contamination of Port Klang by these four heavy metals were between moderate and high contaminated in sediments. With reference to the overall picture, the concentrations of heavy metals in these three mangroves were relatively low. Using the same index (I_{geo}), Ruilian et al. (2008) reported that

Quanzhou Bay, China was heavily polluted by Cd. In addition, the contamination in Quanzhou Bay sediment suggested that the input of different anthropogenic sources due to serious pollution of some metals only existed at localized areas (Ruilian et al., 2008). A study by Christophoridis et al. (2009) stated that the sediments of the Bay of Thessaloniki were unpolluted to moderately polluted by Zn, Cu and Cr. Meanwhile samples from some areas were moderately to strongly polluted by Pb. Defew et al. (2005) also reported that the concentrations of Zn and Pb were high enough to cause moderate to serious contamination in Punta Mala Bay in Panama, thus giving rise to harmful effects on the growth and regeneration of the mangroves. According to a study by Tam and Wong (2000), the sediments in Hong Kong were moderately to seriously contaminated by heavy metals (Pb, Cr and Cu). It also showed that the degree of anthropogenic contamination can be reflected by the concentrations of heavy metals in mangrove swamps at different geographical locations.

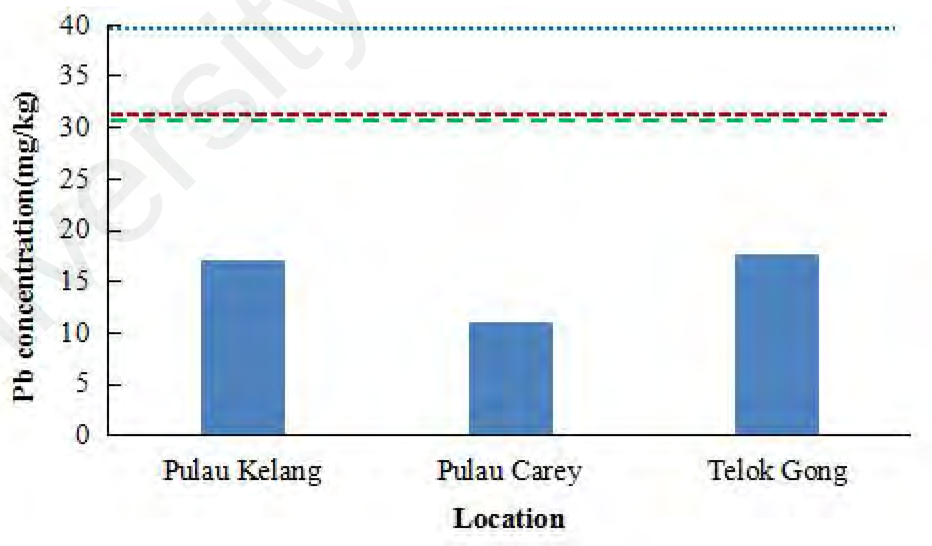


Figure 5.1: Comparison of heavy metal concentrations at the Port Klang with back ground value and sediment quality guidelines (SQG)
……… Back ground value in Port Klang, ……… Lowest effects range (New York Sediment Criteria, – – – – Lowest effects range (ISQG-low) (ISQG 1992), – – Sever effects range (New York Sediment Criteria, High effects range (ISQG-high) (ISQG 1992)

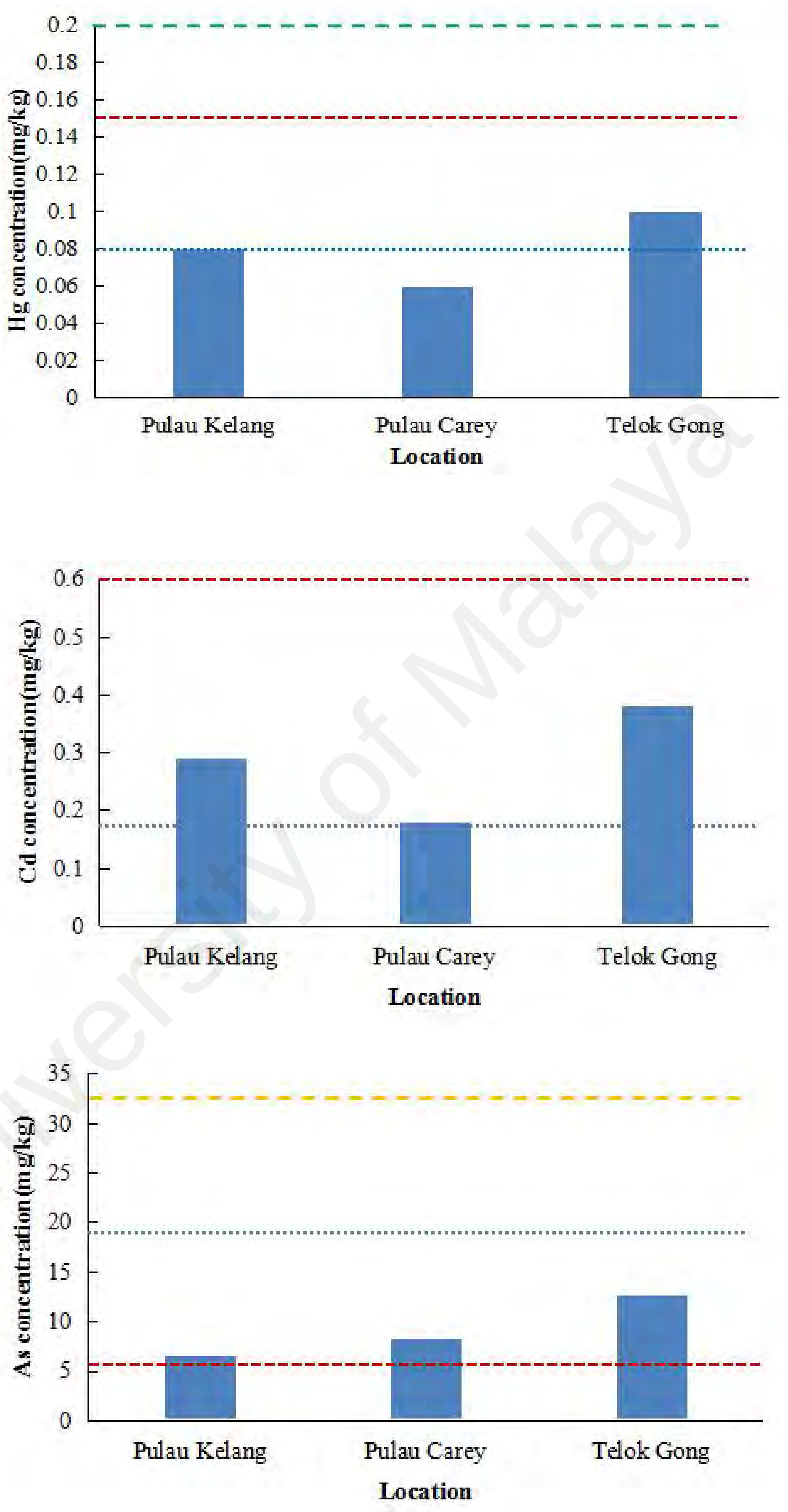


Figure 5.1: Continued

Table 5.1 summarizes the concentrations of same trace heavy metals in other mangroves around the world. Sany et al. (2013) showed that the heavy metal concentrations in Port Klang, Malaysia were 59.450, 0.230, 0.826 and 60.360 mg/kg for Pb, Hg, Cd and As, respectively, which are much higher than those in the present study. A different study found that the concentrations varied at different sampling stations at Port Klang (Sany et al., 2013). The concentrations of these four heavy metals at Donghai Haibor and Sanya Bay in Hainan, China were slightly higher and lower than those in this study (Qiu et al., 2011). However, the Cd level at Wenchang in Hainan, China was not detected (Qiu et al., 2011). Moreover, the concentrations of Pb and Cd at Maipo, Hong Kong and Punta Mala Bay, Panama were much higher than those in this study, although the concentrations of Hg and As were not detected (Tam & Wong, 2000; Defew et al., 2005). At the same time, the concentrations of Hg and As were also not detected in Singapore, India, and Australia. The values of Pb and As were not significantly different as compared to those in this study (Cuong et al., 2005; Ramanathan et al., 1999; Preda & Cox, 2002).

Table 5.1: The average concentrations of tracing heavy metals (mg/kg) in mangrove sediments of Pulau Kelang, Pulau Carey , Telok Gong and some mangroves around the world

Location	Pb	Hg	Cd	As
[a]Pulau Kelang, Selangor,Malaysia	17.070	0.080	0.290	6.470
[a]Pulau Carey , Selangor,Malaysia	11.130	0.060	0.180	8.220
[a]Telok Gong, Selangor,Malaysia	17.750	0.100	0.380	12.590
[b]West port(WC1000),Port Klang, Malaysia	51.310	0.200	0.890	68.130
[b]West port(WL1000),Port Klang, Malaysia	58.230	0.310	0.620	50.310
[b]West port(WT1000), Port Klang, Malaysia	71.550	0.280	1.260	78.300
[b]North port, Port Klang,Malaysia	68.500	0.200	0.890	76.200
[b]South port, Port Klang,Malaysia	52.400	0.190	0.570	40.200
[b]Port Klang, Malaysia	59.450	0.230	0.826	60.360
[c]Donghai Haibor, Hainan, China	19.000	0.080	0.110	13.000
[c]Sanya Bay, Hainan, China	18.000	0.060	0.130	7.000
[c]Wenchang, Hainan, China	30.000	0.060	ND	15.000
[d]Maipo, Hong Kong	79.200	ND	2.620	ND
[e]Sungei Buloh, Singapore	12.280	ND	0.181	ND
[f]Pichavaram, India	11.200	ND	6.600	ND
[g]Punta Mala Bay,Panama	78.200	ND	<10	ND
[h]Queensland, Australia	36.000	ND	0.600	ND

a: this study b: (Sany et al., 2013) c: (Qiu et al., 2011) d: (Tam & Wong, 2000)
e: (Cuong et al., 2005) f: (Ramanathan et al., 1999) g: (Defew et al., 2005)
h: (Preda & Cox, 2002) ND: not detected

By comparing the heavy metal concentrations in the sediments between different land use types, the results of this study showed that the concentrations of all the trace heavy metals were different between mangroves. Pb and Cd levels were significantly different between Pulau Kelang and Pulau Carey (the concentration in Pulau Kelang was higher than that in Pulau Carey), as well as between Pulau Carey and Telok Gong (the concentration in Pulau Carey was lower than that in Telok Gong). The concentrations of As were significantly different between Pualu Kelang and Telok Gong (the concentration in Telok Gong was higher than that in Pulau Kelang), as well as between Pulau Carey and Telok Gong (the concentration in Pulau Carey was lower than that in Telok Gong). However, the different in Hg levels was only significant between Pulau Carey and Telok Gong (the concentration in Telok Gong was higher than that in Pulau Carey) (Table 4.3). Nazli and Hashim (2010) reported that the mangrove location and land use type can influence the concentrations of heavy metals in the mangrove sediments. These three mangroves are in the Port Klang area, which can receive pollution from the surrounding area due to agricultural, industrial, shipping and shipyard activities. This area is also connected with two main rivers which are Klang River and Langat River in the state of Selangor (Haris & Aris, 2013). The concentrations of heavy metals (Pb, Hg, Cd and As) were significantly different between Pulau Carey and Telok Gong (the concentrations were higher in Telok Gong than those in Pulau Carey). This was because Pulau Carey is located at an oil palm plantation area while the Telok Gong mangrove is near an industrial area. These four heavy metals mostly come from industrial activities, such as mining, cement manufacturing, palm oil, wood products and oil / electrical based power generation which that release waste into Port Klang (Sany et al., 2013). During sampling, there were ships on the sea and a lot of domestic waste in the sediments, which was also the reason for the existence of heavy metals. It known that waste discharges and oil spills from ships can release metal pollutants into the sea and result in the contamination of the land (Rozainah et al., 2014). The shipping activities in the Straits of Malacca result in marine pollution by metals such as Pb, As and Cu (Abdullah et al., 1999). In addition, there is a lack of waste treatment at the port and poor legislation guidelines in the action-taking on foreign and domestic ships that discharge their oily waste into the sea. However, the uptake of heavy metals in the mangrove sediments by plants is also influenced by the sediment-related factors such as pH, redox conditions, chlorine contents, and cation exchanges (Nazli & Hashim, 2010). Defew et al. (2005) showed that the mangroves located within the Punta Mala Bay in Panama received high inputs of heavy metal (Zn, Cd and Pb) from anthropogenic activities, which included untreated domestic sewage dumping, shipping activities, and storm water runoffs. Machado et al. (2002) reported that the metal concentrations (Zn, Pb, Cu, Mn) were significantly different between stations at Guanabara Bay, Brazil. In addition, it was suggested that the distribution of Pb, Zn and Cu tended to be largely affected by anthropogenic activities, while Mn tended to be less affected. Tam and Wong (2000) reported that heavy metal concentrations at different geographical locations were different due to the degree of anthropogenic pollution. Vertacnik et al. (1995) also illustrated that the characteristics of the sediments such as quantities and type of organic matter, mineral contents, cation exchange capacity and grain size can affect the concentrations of heavy metals retained in the sediments.

5.2 Heavy metal concentrations in mangrove fresh tissues and litter

In this study, the heavy metal concentrations varied in fresh tissues in mangrove forests, while Pb, Hg, Cd are enriched in the branch. Meanwhile, As showed similar concentrations in fresh branch and fresh leaves. According to a previous study in China, there were no significant differences in the trace metal concentrations between mangrove species, but the metal concentrations in various tissues of mangroves were different. In general, Zn and Cu were found mostly in the fruits, Hg in the leaves, Pb and Cd in the branch and As in the roots (Qiu et al., 2011).

The comparison of the heavy metal concentrations in tissues between different land use types in this study showed that the concentrations of all the tested elements differed between mangroves. The difference in the concentrations of Cd in fresh branch and litter branch were not significant between mangroves. The same applied for Pb and Cd in fresh leaves. Similarly, in a study done by Machado et al. (2002), it was illustrated that the heavy metal concentrations in *L. racemosa* did not have significant differences between locations.

This study also showed that the accumulation coefficients for the four heavy metals at the three mangrove forests were different, and that all the values were less than 1 (Table 4.10 and Table 4.16). In this study, the results for the mangrove specimens, including fresh tissues and litter tissues were as follows : Hg (0.345) > Pb (0.187) > Cd

(0.043) > As (0.040). During the study on metals in Hainan Island, the results for the mangrove specimens had the following sequence : Hg (0.430) > Cu (0.270) > Cd (0.220) > Zn (0.170) > Pb (0.070) > As (0.020) (Qiu, et al. 2011). At the study by He et al. (2014) reported that the range of the accumulation coefficient for Cr in stems was between 1 and 1.5, thus suggesting the potential usage of the stems as an indicator of Cr pollution in sediments. Hg exhibited the highest values because it is a semi-volatile metal. The accumulation coefficient still can reveal the features of mangrove plants for different heavy metals under different land use types being primary producers, mangrove forests can provide clean food for consumers (Wen - jiao et al., 1997). Low accumulation coefficients have also been reported in *A. marina*, *R. stylosa* and *K. candel* mangroves (Feng Zhong et al., 1996; Zheng et al., 1995; Zheng & Lin, 1995). Low absorption ability of mangroves for heavy metals may relate to the low amount of heavy metals in the sediments. It was demonstrated by Wen-jiao et al. (1997) that the reason was that high concentrations of sulfur in the mangroves can combine with heavy metals to form less soluble complexes.

Although the concentrations of heavy metals varied in different tissues in all three sampled mangroves, it is obvious that the concentrations of heavy metals in litter were higher than those in fresh tissues. Generally, the average concentrations of Pb, Hg, Cd and As in the litter branch were higher than those in the fresh branch by 35.9%, 28.9%, 42.6% and 91.1% respectively. Meanwhile, the aforementioned concentrations in the litter leaves were higher than those in the fresh leaves by 40.9%, 34.9%, 61.1% and 79.6% respectively. Similar results had also been observed in other mangroves. Peng et al. (1997b) reported the levels of Cu, Pb, Zn, Cr, Zn and Mn in the litter leaves and branch were higher than those in the fresh leaves and branch. The former was at least 3 times higher than the latter, with the reason being that the litter leaves and branch can have direct contact with the soil and sea water to adsorb heavy metals. Lin and Ronghua

(1990) also illustrated that the litterfall of a mangrove plant named *K. candel* is able to adsorb mercury. Mangrove litter falls are an important food and energy source for a variety of aquatic organisms (including cultures of oysters, clams, crabs, shrimps, and fishes) and hence, they have an impact on the coastal productivity. The residues with high potential concentrations of heavy metals were unfavorable for the animals that feed on residues on the ground. However, the litter also can be shifted seawards with tidal flow, which will release the accumulated products and recycle the contamination in the mangrove zones. A study by Wen - jiao et al. (1997) showed that the concentrations of Cu, Pb, Zn, Cd, Cr, Ni and Mn in the residues were higher than those in litterfalls of *R. stylosa*. However, there was another study that showed the essential elements such as Cu were of lower concentrations in litter. The lower Cu levels in leaf litter suggested a transport of Cu to other plant organs during senescence, or leaf decomposition, or the leaching of more soluble ions (Siddiqui & Qasim, 1994; Peng et al., 1997a).

5.3 Comparison of heavy metal concentrations in sediments, litter and plant in mangroves

In this study, the concentrations of Pb, Hg, Cd and As in the mangroves were in the order of sediments > litter (branch and leaves) > fresh (branch and leaves). The heavy metal concentrations in different fresh tissues were not significantly correlated with those in the sediments. The same method was also used by Pakzadtoochaei (2013) to test the relationships of Cd, Cu, Ni and Zn in the root, leaf and stem with those in the sediments. It was shown that Cd was not significantly correlated between tissues and sediments. However, Ni was significantly correlated, hence illustrating that the tissues of *A. marina* can be a good bioindicator of this element. The tissues (branch and leaves) of these three mangroves in the present study cannot act as bioindicators of Pb, Hg, Cd and As concentrations.

CHAPTER 6: CONCLUSION

The concentrations of Pb, Hg, Cd and As in the sediments (of depth 0 - 15 cm) were in the order of Pb > As > Cd > Hg in all the sampled mangroves. At the same time, the concentrations of these heavy metals in the sediments correlated with the tidal gradient, although not significantly. In addition, the concentrations of the heavy metals differed between land use types, especially between Pulau Carey and Telok Gong that were significantly different. The Telok Gong mangrove had higher heavy metal concentrations than Pulau Carey. In this study, As probably had adverse effects on organisms, according to the New York Sediment Criteria and Provincial Sediment Quality Guidelines. Comparison of the heavy metal concentrations between mangroves in plant tissues, the results showed that Cd in the fresh branch, fresh leaves, and litter branch did not significantly different between locations. Pb in the fresh leaves also did not significantly different in fresh leaves between locations. In these three mangroves, the tracing heavy metal elements cannot become as the bioindicator at plant branch and leaves because all elements concentrations in the branch and leaves were not significantly correlated with the concentrations in sediments.

According to geoaccumulation index, all the sampled mangroves were considered as uncontaminated to moderately contaminated (Class 1) by these four heavy metals. The accumulation coefficient of the heavy metals of plant branch for heavy metals in sediments was following Hg > Pb > Cd > As, while for leaves it was in the order of Hg > Pb > As > Cd. In addition, all accumulation coefficient values of the tested heavy metals (Pb, Hg, Cd, As) were less than 1, suggesting that the branch and leaves of these three mangroves were not potentially useful as indicators of sediment contamination by these four elements.

Finally, the concentrations of Pb, Hg, Cd and As in the mangroves were in the order of sediments > litter (branch and leaves) > fresh (branch and leaves). Pb, Hg and Cd were enrich in branch, while As shared similar concentration between branch and leaves.

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