UNIVERSITI PUTRA MALAYSIA

HEAVY METAL CONCENTRATIONS IN SURFACE SEDIMENTS AND TISSUES OF Periophthalmodon schlosseri (Pallas, 1770) AND Arius sumatranus (Bennett, 1830) FROM SELECTED SITES IN PENINSULAR MALAYSIA

AHMED JAFARU MALAM

FS 2021 1

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By

AHMED JAFARU MALAM

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

December 2018

DEDICATION

Dedicated to the memory of my late father, Mal Ahmad K. Hashim Bunza who showed me the path to knowledge and my beloved wife Ramlatu Ahmad, may Allah grant the two Jannatul Firdaus. And To my elderly mother, Hauwa'u Ahmad who has been there for me through thick and thin. I pray to Allah to grant her good ending.

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

HEAVY METAL CONCENTRATIONS IN SURFACE SEDIMENTS AND TISSUES OF *Periophthalmodon schlosseri* (Pallas, 1770) AND *Arius sumatranus* (Bennett, 1830) FROM SELECTED SITES IN PENINSULAR MALAYSIA

By

JAFARU MALAM AHMED

December 2018

Chairman : Professor Ahmad bin Ismail, PhD
Faculty : Science

Heavy metals have been studied extensively in sediments and tissues of aquatic biota due to their toxic effects and non biodegradable nature in aquatic environment. Heavy metals concentrations in surface sediments and their effects on giant mudskipper *Periophthalmodon schlosseri* Pallas 1770 and marine catfish *Arius sumatranus* Bennet 1830 from selected sites in Peninsular Malaysia were investigated. The anthropogenic source of heavy metal pollution in the selected sites was examined by geochemical fractionation and relationships between histological structures and reduced glutathione; GSH were determined. Triplicate samples of surface sediment, *P. schlosseri* and *A. sumatranus* were collected from selected sampling sites in Peninsular Malaysia. The mudskipper lives directly on the surface sediment and feeds on the organisms found there while the marine catfish lives just above the surface sediments and feeds on the bottom organisms also. The two species are readily available in the selected sampling sites. The sites include; Bagan Lalang (BL) in Selangor, Kuala Gula (KG) in Perak, Kuala Juru (KJ) in Penang, Sungai Puluh (SP) in Klang and Sungai Tiga (ST) in Johor in 2010, 2012 and 2014 respectively. Surface sediments and fish samples were collected in March and June 2010, February and May 2012 and March and May 2014 from the sampling sites. Geochemical speciation and total concentrations of heavy metals in sediments were extracted using a modified 4-step sequential extraction technique (SET) (Easily, freely or leachable and exchangeable fraction (EFLE), Acid reducible, oxidisable organic and resistant fractions) and aqua-regia procedures (QA/QCT). Histopathological examination of some tissues (liver, muscles and gills) was carried out. Reduced Glutathione concentration was determined in liver, gill, muscle and kidney of *P. schlosseri* and *A. sumatranus* using QuantiChrom Glutathione Assay Kit (Catalog no. DIGT-250 from BioAssay Systems Company, USA). The data obtained was analysed using analysis of variance (ANOVA) at 95% confidence interval, multiple comparison test was carried out using Duncan's Multiple Range Test. Pearson's correlation coefficient (r)

was used to test the relationships between heavy metals and tissues of the fish species and GSH levels. Results obtained shows that the mean total concentration of the metals in surface sediment was significantly ($p<0.05$) highest for Cd at SP 2.21±0.43 and lowest at BL 0.91±0.22, while Cu, Ni, Pb, V and Zn were 42.96±0.45 at KJ and 8.44±0.11 at SP, 34.79±0.75at KJ and 10.79±0.69 at ST, 42.72±0.89 at KJ and 21.58±1.19 at BL, 33.81±5.53 at ST and 20.02±1.43 at BL, 214.82±1.54 at KJ and 70.75±3.72 at BL µg/g dry weight respectively. Geochemical fractions of metals indicates that of all the metals under the present study only Cu has low anthropogenic contribution as its resistant fractions were higher than the non-resistant fractions in all the sites (except in BL) with highest in KJ (67.88%) and lowest in BL (46.25%). Non-resistant fractions from all the metals except Cu and in all the other sampling sites were higher than the resistant fractions (Cd 80.33% at SP, Ni 75.44% at KJ, Pb 65.01% at SP, V 76.89% at KG and Zn 86.54% at BL). The concentrations of Cd from all the sampling sites were above the values of ERL for Cd (1.2 µg/g) except for BL (0.91 µg/g). Concentrations of Cu in all the sampling sites except for KJ (42.96 µg/g) were below the ERL value of Cu (34 µg/g) and below the ERM value of Cu (270 µg/g). Nickel concentrations at both KG (21.63 µg/g) and KJ (34.79 µg/g) were above the ERL value (20.9 µg/g) while all other sites were below the ERL value and below ERM value of Ni (51.6 µg/g). The concentrations of Pb in all the sampling sites were below both ERL (46.7 µg/g) and ERM (218 µg/g) values while Zn concentrations in KJ (214.82 µg/g) was above the ERL value of Zn (150 µg/g) but below ERM value (410 µg/g). The highest mean enrichment factor (EF) values were recorded for Cd (50.78 at SP), Cu (2.72 at KJ), Ni (1.72 at KG), Pb (6.28 at KG), V (1.15 at KG) and Zn (5.38 at KJ). The values obtained for index of geoaccumulation were either above or below the three background concentrations used. Pollution load index (PLI) values obtained for the present study (0.98, 1.39, 2.54, 1.29 and 1.31) for BL, KG, KJ, ST and SP respectively, were higher than all the three background concentrations used. The concentration of metals in the tissues of *P. schlosseri* showed lowest and higest concentrations in gill (BL - 0.28 µg/g) and bone (SP - 2.12 µg/g), muscle (BL - 1.10 µg/g) and liver (ST - 5.54 µg/g), muscle (SP - 0.59 µg/g) and bone (ST - 8.340 µg/g), muscle (BL - 0.92) µg/g and bone (SP - 63.96 µg/g), scale (ST - 0.130 µg/g) and bone (BL - 1.03 µg/g) and muscle (SP - 33.84 µg/g) and skin (SP - 145.54 µg/g) for Cd, Cu, Ni, Pb, V and Zn µg/g dry weight respectively. And for *A. sumatranus* showed lowest and highest concentrations in skin (KG - 0.13 µg/g) and bone (SP - 1.83 µg/g), skin (SP - 0.35 µg/g) and liver (ST - 28.27 µg/g), muscle (KG - 0.19 µg/g) and bone (ST- 9.47 µg/g), skin (ST - 2.14 µg/g) and bone (KG - 57.90 µg/g), mmuscle (SP - 0.09 µg/g) and gill (SP - 1.09 µg/g) and muscle (ST - 43.78 µg/g) and liver (SP - 793.25 µg/g) for Cd, Cu, Ni, Pb, V and Zn µg/g dry weight respectively. For both species the concentrations of metals in the tissues indicate significant differences ($p<0.05$) between the sampling sites except in few cases which shows non-significant differences ($p>0.05$). The bioconcentration factors (BCF) calculated for both species designate that some of the tissues were grouped as macro concentrators. In *P. schlosseri* Cu and V had the highest average values of BCF in gill (3.80) and bone (6.08) respectively while in *A. sumatranus* V and Zn had the highest average BCF values both in liver (6.87 and 10.51) correspondingly. The Pearson's correlation coefficient (r) analysis between metal concentrations in the tissues of both species and geochemical fractions of sediment indicate significant ($p<0.05$) correlations in some fractions signifying that both species are suitable for use as biomonitoring agents for heavy metal pollution in the region. Histopathological examinations of some tissues of both species indicated from mild, moderate to severe forms of alterations of the tissues such as congested vessels, vacuolation of hepatocytes, matting of villi (gill synechiae) and heterophil infiltration. The liver had the highest mean GSH value of 207.72 and gill had the lowest mean GSH values of 110.15 in *P. schlosseri* and *A. sumatranus* respectively. Domination of non-resistant over resistant fraction indicates anthropogenic contribution of the metals in the study areas. Overall results of concentrations of heavy metals, GCF and indices of pollution in the surface sediment signifies that all the sampling sites used in the present study could be regarded as either unpolluted, moderately or strongly polluted. The lowest concentration of heavy metal in this study displays higher PLI values compared to the other three background concentrations used signifying anthropogenic input of heavy metals in the sampling sites.

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

KONSENTRASI LOGAM BERAT DALAM SEDIMEN PERMUKAAN DAN TISU *Periophthalmodon schlosseri* (Pallas, 1770) DAN *Arius sumatranus* (Bennett, 1830) DARI LOKASI TERPILIH DI SEMENANJUNG MALAYSIA

Oleh

JAFARU MALAM AHMED

Disember 2018

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Logam berat telah dikaji secara meluas dalam sedimen dan tisu biota akuatik akibat kesan toksiknya dan sifat tidak terbiodegradasi dalam persekitaran akuatik. Kepekatan logam berat di sedimen permukaan dan kesannya pada ikan belacak raksasa *Periophthalmodon schlosseri* Pallas 1770 dan ikan duri *Arius sumatranus* Bennet 1830 dari lokasi terpilih di Semenanjung Malaysia telah dikaji. Sumber antropogenik pencemaran logam berat di lokasi tersebut diperiksa dengan fraksinasi geokimia dan hubungan antara struktur histologi dan Glutation yang berkurangan; GSH. Tiga replikasi sampel sedimen permukaan, sampel *P. schlosseri* dan *A. sumatranus* telah diambil dari lokasi terpilih di Semenanjung Malaysia. Ikan belacak hidup secara langsung di atas sedimen permukaan atas dan memakan organisma yang terdapat di situ manakala ikan duri hidup di permukaan sedimen dasar serta memakan organisma di situ. Kedua-dua spesies ini mudah didapati di lokasi terpilih yang dinyatakan. Lokasi tersebut merangkumi; Bagan Lalang (BL) di Selangor, Kuala Gula (KG) di Perak, Kuala Juru (KJ) di Pulau Pinang, Sungai Puluh (SP) di Klang dan Sungai Tiga (ST) di Johor, sepanjang tempoh kajian pada tahun 2010, 2012 dan 2014. Sedimen permukaan dan sampel dikumpulkan pada bulan Mac dan Jun 2010, Februari dan Mei 2012 dan Mac dan Mei 2014 dari tapak kajian. Spesiologi geokimia dan logam berat dalam sedimen permukaan diekstrak menggunakan teknik pengekstrakan berurutan 4-langkah (SET) yang diubahsuai (Fraksi mudah, bebas, boleh laras dan boleh ditukar ganti (EFLE), pecahan organik, pecahan organik dan tahan oksida) dan aqua-regia prosedur (QA / QCT). Pemeriksaan histopatologi beberapa tisu (hati, otot dan insang) turut dijalankan. Kepekatan Glutation yang berkurangan ditentukan di dalam hati, insang, otot dan buah pinggang *P. schlosseri* dan *A. sumatranus* menggunakan QuantiChrom Glutation Assay Kit (Katalog no. DIGT-250 daripada BioAssay Systems Company, USA). Data yang diperoleh dianalisis menggunakan analisis varians (ANOVA) pada selang keyakinan 95%, ujian perbandingan berganda dilakukan dengan menggunakan Ujian Pelbagai Duncan. Koefisien Korelasi Pearson

(r) digunakan untuk menguji hubungan antara logam berat dan tisu spesies ikan dan tahap GSH. Keputusan yang diperolehi menunjukkan bahawa purata kepekatan logam dalam sedimen permukaan adalah signifikan ($p < 0.05$) untuk Cd di SP (2.21 ± 0.43) dan paling rendah di BL (0.91 ± 0.22), manakala Cu , Ni, Pb, V dan Zn masing-masing adalah 42.96 ± 0.45 di KJ dan 8.44 ± 0.11 di SP, 34.79 ± 0.75 di KJ dan 10.79 ± 0.69 di ST, 42.72 ± 0.89 di KJ dan 21.58 ± 1.19 di BL, 33.81 ± 5.53 di ST dan 20.02 ± 1.43 di BL, 214.82 ± 1.54 di KJ dan 70.75 ± 3.72 di BL µg/g dry weight. Fraksi geokimia logam berat menunjukkan di antara semua logam yang di kaji, hanya Cu mempunyai sumbangan antropogenik yang rendah kerana pecahan rintangnya lebih tinggi daripada pecahan tidak rintang, di semua lokasi (kecuali di BL) dengan nilai tertinggi di KJ (67.88%) dan terendah di BL (46.25%). Fraksi tidak rintang logam berat yang lain (kecuali Cu) di semua lokasi adalah lebih tinggi daripada pecahan rintang (Cd 80.33% di SP, Ni 75.44% di KJ, Pb 65.01% di SP, V 76.89% di KG dan Zn 86.54 % di BL). Kepekatan Cd di semua lokasi persampelan adalah di atas nilai ERL (1.2 µg/g) kecuali BL (0.91 µg/g). Konsentrasi Cu di semua tapak sampel kecuali KJ (42.96 µg/g) berada di bawah nilai ERL (34 µg/g) dan di bawah nilai ERM (270 µg/g). Kepekatan Ni pada KG (21.63 µg/g) dan KJ (34.79 µg/g) berada di atas nilai ERL (20.9 µg/g) manakala lokasi lain berada di bawah nilai ERL dan di bawah nilai ERM (51.6 µg/g). Kepekatan Pb di semua lokasi adalah di bawah nilai ERL (46.7 µg/g) dan ERM (218 µg/g) manakala kepekatan Zn di KJ (214.82 µg/g) berada di atas nilai ERL (150 µg/g) tetapi di bawah nilai ERM (410 µg/g). Nilai faktor pengayaan (EF) tertinggi dicatatkan untuk Cd (50.78 di SP), Cu (2.72 di KJ), Ni (1.72 di KG), Pb (6.28 di KG), V (1.15 di KG) dan Zn (5.38 di KJ). Nilai-nilai yang diperolehi untuk indeks geo-pengumpulan berada sama ada di atas atau di bawah tiga kepekatan latar belakang yang digunakan. Indeks nilai pencemaran (PLI) yang diperolehi untuk kajian ini masing-masing adalah 0.98, 1.39, 2.54, 1.29 dan 1.31 untuk BL, KG, KJ, ST dan SP. Ia adalah lebih tinggi daripada semua tiga kepekatan latar belakang yang digunakan. Kepekatan logam dalam tisu *P. schlosseri* menunjukkan kepekatan yang paling rendah dan tertinggi di insang (BL - 0.28 µg/g) dan tulang (SP - 2.12 µg/g), otot (BL - 1.10 µg/g) dan hati (ST - 5.54 µg/g), otot (SP - 0.59 µg/g), tulang (SP - 63.96 µg/g), kulit (ST - 0.130 µg/g) dan tulang (BL - 1.03 µg/g) dan otot (SP - 33.84 µg/g) dan kulit (SP - 145.54 µg/g) bagi Cd, Cu, Ni, Pb, V dan Zn. *Arius sumatranus* menunjukkan kepekatan yang paling rendah dan tertinggi dalam kulit (KG - 0.13 µg/g) dan tulang (SP - 1.83 µg/g) hati (ST - 28.27 µg/g), otot (KG - 0.19 µg/g) dan tulang (ST - 9.47 µg/g), kulit (ST - 2.14 µg/g) dan tulang (KG - 57.90 µg/g), insang (SP - 1.09 µg/g), otot (ST - 43.78 µg/g) dan hati (SP - 793.25 µg/g) masing - masing bagi Cd, Cu, Ni, Pb, V dan Zn. Bagi kedua-dua spesies, kepekatan logam dalam tisu menunjukkan perbezaan yang signifikan ($p<0.05$) di antara lokasi persampelan kecuali dalam beberapa kes yang menunjukkan perbezaan tidak signifikan ($p>0.05$). Faktor bio-konsentrasi (BCF) yang dikira untuk kedua-dua spesies ini menunjukkan bahawa beberapa tisu boleh dikategorikan sebagai konsentrator makro. Dalam *P. schlosseri*, Cu dan V mempunyai nilai purata tertinggi BCF di insang (3.80) dan tulang (6.08) manakala bagi *A. sumatranus*, V dan Zn mempunyai nilai BCF purata tertinggi di dalam kedua-dua hati (6.87 dan 10.51). Analisis koefisien korelasi Pearson (r) antara kepekatan logam dalam tisu kedua-dua spesies dan pecahan geokimia sedimen menunjukkan korelasi signifikan ($p<0.05$) dalam beberapa pecahan menandakan bahawa kedua-dua spesis ini sesuai digunakan sebagai agen bio-monitor untuk pencemaran logam berat di rantau ini. Pemeriksaan histopatologi beberapa tisu daripada kedua-dua spesies ini menunjukkan terdapat alterasi bersifat ringan, sederhana dan teruk seperti kongesi pada tisu, vakulasi pada

tisu hepatosit, gabungan vili (sinesi insang) dan infiltrasi heterophil. Hati mempunyai nilai GSH purata tertinggi iaitu 207.72 dan insang mempunyai nilai purata GSH terendah 110.15 pada *P. schlosseri* dan *A. sumatranus*. Dominasi fraksi tidak rintang berbanding fraksi rintang menunjukkan sumbangan antropogenik di lokasi kajian. Keputusan keseluruhan daripada kepekatan logam berat, GCF dan indeks pencemaran di sedimen permukaan menunjukkan bahawa semua lokasi kajian boleh dianggap samada sebagai tidak tercemar, sederhana atau sangat tercemar. Kepekatan logam berat yang paling rendah dalam kajian ini memaparkan nilai PLI yang lebih tinggi berbanding dengan tiga konsentrasi latar belakang lain yang digunakan untuk menandakan input antropogenik logam berat di lokasi kajian.

ACKNOWLEDGEMENTS

In the name of Allah, the Beneficent, the Most Merciful, all thanks and praises are due to Him, the Lord of the worlds who taught man that which he knew not. First and foremost, I begin by expressing my profound gratitude to Almighty Allah for sparing my life to witness this important opportunity of starting and completing this task successfully. His bounties are enormous, for there were times when things were very tough and He gave me the strength, courage and ability to persevere.

I remain grateful to the chairman of my supervisory committee Prof. Dr. Ahmad Ismail for constructively and critically supervising the work from start to finish. I am also very greatful to members of the supervisory committee; Dr Syaizwan Zahmir Zulkifli and Dr Hishamudin Omar for their kind help and care throughout the work. My thanks also goes to the postgraduate coordinator and all academic and non academic staff members of the department. I must thank Faid Rahman for being there for me when it matters. I will like to remember the help and cooperation of friends and colleagues in the laboratory; such as Tijjani Buhari, Musa Ibrahim and a host of others, thank you all.

I am indeed grateful to my family for being part of this study; my wife Fatima Abubakar and my children for their patience throughout the period of this study. I will not forget my brothers Nasir Ahmad, Abbas Ahmad and others who stood for me in my absence. My regards to all my friends and colleagues for encouraging me to persevere and complete the journey. To all of you I remain grateful.

This thesis was submitted to the Senate of the Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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Date: 12 August 2021

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LIST OF ABBREVIATIONS

AAS	Atomic absorption spectrophotometer
Anova	Analysis of variance
BCF	Bio concentration Factor
BDL	Below detection limit
CF	Concentration Factor
cm	Centimeter
CRM	Certified Reference Material
DDW	Deionised Distilled Water
dw	Dry Weight
EDTA	Ethylene diamine tetra acetic acid
EF	Enrichment Factor
EFLE	Easily or freely, leachable and exchangeable
ERL	Effective Range Low
ERM	Effective Range Medium
g	Gram
H	Hour
GHS	Glutathione
HCl	Hydrochloric acid
H_2O_2	Hydrogen peroxide
$HClO_4$	Perchloric acid
HNO_3	Nitric acid
I_{geo}	Index of geoaccumulation
Km^2	Kilometre square
mg/L	Milligram per litre
ml	Millilitre

Mg/Kg	Milligram per kilogram
ND	Not detectable
nm	Nano metre
NH_4CH_3COO	Ammonium acetate
No.	Number
PLI	Pollution Load Index
ppm	Parts per million
SET	Sequential Extraction Technique
SQG	Sediment Quality Guideline
Sg	Sungai
w/w	Wet weight
^0C	Degrees Celsius
µg/g	Microgram per gram
µl	Micro litre
µm	Micrometre
$^0/_0$	Percentage
GPS	Global Positioning System
^{0}	Degree
′	Inch
BDH	Bilisim Destek Hiz

CHAPTER 1

INTRODUCTION

This chapter consists of background of the study which highlighted the background information of the study, statement of the research problem that explains the problems that necessitate conducting this study, scope and limitations of this study and the main and specific objectives of the present study.

1.1 Background of the study

Heavy metals discharge and contamination in aquatic environments especially the coastal waters has become one of the major environmental problems of concern universally. Due to their toxic effects, persistence and bioaccumulation in the aquatic environment, heavy metals are among the major pollutants in the natural environment. The perspective view of the present study has to do with total heavy metals content from sediments, the tissues of the two fish species and their effects on the tissues of the fishes such as liver, gills, muscles and kidney. Ecotoxicology research is important in the study of instant effect of a toxic material on organisms in food chain with the eventual goals of forecasting consequences on wildlife biota, ecosystems and on food resources like fish and other aquatic organisms.

Heavy metals usually find their way into the sediments of aquatic ecosystems through sedimentation, precipitation and adsorption to particulate matter. Metals that are associated with sediments can cause direct effect to bottom feeders like fish and other organisms and can also cause long term effects to organisms at the higher trophic level like humans and birds (Eimers, Evans, & Welbourn, 2001). Continuous release of essential and non-essential heavy metals into the aquatic system can render aquatic organisms vulnerable to abnormally high concentrations of the metals triggering dreadful consequences on the aquatic environment (van Dyk, Pieterse, & van Vuren, 2007).

Naturally there are two sources of heavy metals into the aquatic environment; weathering of rocks and soils and via anthropogenic activities through release of industrial and urban wastes into aquatic systems (Singh, Mohan, Singh, & Malik, 2005). Suspended particulate matter and sediments are the chief sources of heavy metals in the marine environment and act as vital materials in the transportation and storing of possibly harmful metals (Cuong *et al*., 2008).

Extensive studies have been conducted and reported on heavy metal concentrations in sediments and biological samples by many workers (Rahman, Ismail, & Yusof, 2013; Naji, Ismail, & Ismail, 2010; Kamaruzzaman, Ong, Rina, Istana, & Terengganu, 2010; Zulkifli, Mohamat-Yusuff, Arai, Ismail, & Miyazaki, 2010a & 2010b; Ikram, Ismail, Yap, & Azwady, 2010; Ismail, & Safahieh, 2005; Yap, Ismail, & Chui, 2005).

However, information on histological study of fish tissues exposed to heavy metal pollution in this area is not well documented.

Among all biomonitors, fishes are regularly used as they provide numerous benefits in defining the natural characteristics of aquatic environments and in measuring fluctuations to habitats. Such benefits include; i) being continuously in the water, as such allowing the incessant monitoring of contaminants and permitting spatial assimilation of pollutant data ii) they live longer and assimilate variations of contaminants over period and iii) the magnification provided by bioaccumulation increases the precision and minimizes the cost of analysis of trace pollutants (Lamas, Fernández, Aboal, & Carballeira, 2007). Also, due to their position at the top of food chain in the aquatic ecosystem, fishes are good indicators of environmental pollution (Bury, Walker, & Glover, 2003; Türkmen, Türkmen, Tepe, & Akyurt, 2005).

Very few studies however, focused on mudskippers as biomonitoring agents of heavy metals pollution (Buhari, & Ismail, 2016; Ikram et al., 2010). Giant mudskipper *Periophthalmodon schlosseri* and marine catfish *Arius sumatranus* were chosen as biological organisms in this study.

Mudskipper are amphibious fishes that demonstrate an extreme amphibious behaviour, with some species living more outside water than in it and possess different physiological, morphological and behavioural characteristics for amphibious life (Ip, & Chew, 2018; Zulkifli et al., 2012; Gordon, 1998; Graham, 1997). By feeding on small crustaceans and scraping on algae and diatoms on mudflats, mudskippers play a significant impact on benthic ecology. They differ in the type of habitats they colonize, ranging from brackish to normal seawater, swamps and estuaries, mudflats, intertidal habitats and mangrove habitats (Sarhadizadeh, Afkhami, Ehsanpour, & Bastami, 2014).

Marine catfish *A. sumatranus* are important for both eco-toxicological and ecological researches serving as very good bio-indicators of pollution assessment and monitoring in coastal environments. Catfish are bottom dwelling fish living and feeding on or around the bottom sediment which is the sink of heavy metals. They are important in the ecology of benthic organisms because they feed mainly on invertebrates that live on the bottom sediment. The two species offer enough tissues as sample due to their large sizes.

Heavy metal exposure to fish organs such as liver, muscles, kidney, gills etc., can cause some histological alterations. Organs like liver whose function is cleansing the body is important for both the metabolism and the elimination of poisonous materials in the body. For this reason, histological study or examination of exposed samples may therefore bring significant results. Buslé & Anadon (1996) states that fish liver histology could serve as a model for studying the interactions between environmental factors and hepatic structures and functions. Detrimental effect of heavy metal pollution on fish organs histology could, nevertheless, rest on the period of the exposure (chronic or acute) and the concentration level of the particular metal.

Although vanadium has been regarded as an essential element and beneficial to normal cell growth, it becomes toxic when its concentration is increased to higher levels. It is toxic to plants, fresh water and marine organisms. Vanadium is toxic to cells, causing effects on some important organs such as liver, muscles, and kidney at higher concentrations. It causes lipid peroxidation, oxidative damage, changes in reproductive, respiratory and haematological structures (Soares, Martins, Gutiérrez-Merino, & Aureliano (2008).

Reduced glutathione (GSH) is one of the main antioxidant elements involved in the glutathione metabolism that help in managing the oxidative stress triggered by metals and other stressors of the environment (Tripathi, Mehta, Amar, & Gaur, 2006; Pinto, Sigaud- Kutner, Leitao, & Okamoto, 2003). Biomarkers are vital as "early warning signals" prior to harmful effects occur in aquatic organisms. They can also be essential tools used in assessing the sub-lethal levels of toxic substances in the aquatic environment and thus, they have acquired significance in monitoring metal toxicity programs. Tissues of fish, particularly liver and kidney, comprise of certain antioxidant systems to defend them from the oxidative stress produced by xenobiotic like metals (Baysoy et al., 2012; Atli & Canli, 2007; Basah & Rani, 2003).

Due to rapid industrial development Malaysia has recorded thereby resulting in discharge of effluents into aquatic water bodies, it became necessary to measure or evaluate the concentrations of heavy metals in the sediment and different tissues of the two fish species. And also taking into consideration the importance of mudskipper and catfish in intertidal mudflats and bottom part of the water respectively from selected arrears of Peninsular Malaysia and relate them with the histological structures of some tissues. The selected coastal environments are highly productive and vulnerable areas which are directly affected and exposed to this contaminants due to their closeness to pollution sources.

1.2 Statement of the Problem

Over twenty rivers pass through areas of human activities with heavy metals pollution transporting a lot of pollutants into Malacca Straits. As stated by Abdullah et al. (1999), the region consists of fifty four ports and twenty eight industrial domains out of which three are major ones (Penang, Klang and Malacca). These serve as pollution threat and have negative impact on the ecosystem and biota.

Malaysia's coastal environment is endowed with a lot of natural resources, thus its economic and environmental importance cannot be overemphasised. For this reason most of the population is concentrated here with a lot of human activities such as agricultural activities, urban development, aquaculture, fisheries activities, exploration of oil and gas, industrial activities, transportation, tourism activities, communication and many more making the area as hub of economic accomplishments (Abdullah, Tahir, Loong, Hoque, and Sulaiman,1999).

Greater part of Malaysia's human population, economic activities, coastline incomes as well as agricultural activities are situated in the western part of Peninsular Malaysia and as such, most of the researches as far as heavy metals are concerned are concentrated here (Abdullah et al., 1999). Hence the source of heavy metal pollution in this region are through agricultural activities (Ismail, 1993), urban development and agro based and manufacturing industries (DOE, 1998), mining of steel, iron, tin, gold (Hashim, Nayan, Saleh, Mahat, Said, & Shiang, 2018), plastics and electronics industries (Maznah, Yahya, Talib, Merican, & Shuhaida, 2018: Hamid & Sidhu, 1993), shipping activities (Abdullah et al., 1999). All these led to rapid industrial development for Malaysia which consequently could lead to pollution of the environment. Alkarkhi, Ismail, Ahmed, & Easa (2009) have reported incidence of pollution from industries and due to Malaysia's extensive coastlines along the Straits of Malacca and the South China Sea, aquatic environmental pollution could be a very serious threat. Hence the selected study sites are very important as described above and became necessary to carry out this study to understand the status of pollution therein.

1.3 Objectives of the Study

The main objective of the present study was to ascertain the heavy metal concentrations in surface sediment and some tissues of giant mudskipper *Periophthalmodon schlosseri* and marine catfish *Arius sumatranus.* The study aimed at achieving this via the specific objectives listed below:

1. To determine the concentration of heavy metals (Cd, Cu, Ni, Pb, V, and Zn) in surface sediment and some tissues of *P. schlosseri* and *Arius sumatranus.*
2. To examine the anthropogenic source of heavy metal pollution in the selected sites by geochemical fractionation and evaluate the level of heavy metal pollution using recognised Pollution Indices and Standard Quality Guidelines
3. To determine the relationship between heavy metals and histological structures of some tissues of the two fish species.
4. To establish the relationship between *P. schlosseri* and *A. sumatranus* and the concentrations of heavy metals by measuring reduced glutathione (GSH) in the liver, muscle, kidney and gill.

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