HEAVY METALS CONTAMINATION IN SEDIMENTS AND SELECTED COMMERCIALLY IMPORTANT BIVALVES IN MARUDU BAY

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THESIS SUBMITTED IN FULFILLMENT FOR THE DEGREE OF MASTER OF SCIENCE

BORNEO MARINE RESEARCH INSTITUTE
UNIVERSITI MALAYSIA SABAH
2018

UNIVERSITI MALAYSIA SABAH

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ACKNOWLEDGEMENT

I would like to express my deepest gratitude and appreciation to my supervisor, Assoc. Prof. Dr. Julian Ransangan for all his advices, guidance and support in throughout this research work that lead to the completion of this study. I also wish to thank my co-supervisor, Dr. Faihanna Ching Abdullah for her helpful teaching and recommendations in many parts of this research. Moreover, I must thank Dr. Tan Kar Soon, staffs and postgraduate students at the Microbiology and Fish Disease Laboratory, Borneo Marine Research Institute for helping me in field sampling and to the staffs at the Faculty of Science and Natural Resources for their laboratory assistance in heavy metal content determination using the ICP-OES (Inductively Coupled Plasma- Optical Emission Spectrometry).

Last but not least, I would like to thank the Ministry of Education Malaysia for granting me a two year scholarship (MYBRAIN) and also for approving a Niche Research Grant Scheme (NRGS0003) to financially support my research.

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ABSTRACT

Marudu Bay has become an important fishing ground due to its high marine biodiversity. Other than that, Kota Marudu district has been identified as potential area for agriculture apart from a holding potential for eco-tourism activities. However, the growing activities of agriculture (especially oil palm plantation) which involved the usage of chemicals (fertilizer, pesticides etc.) were blamed for the mass mortality of cultured green mussel in Tanjung Batu, Marudu bay. The mass mortality event was severely affected the green mussel aquaculture in the area. Hence, this study was conducted in order to measure the level of heavy metal in commercially important bivalves originating from Marudu bay and to investigate its relationship with the physicochemical variables. Other than that, this study also examine if the level of metals in the bivalves can possibly bring about health risk to consumers. Current study was conducted by collecting green mussel (n=120) and sediment (n=36) samples monthly for 12 months period. The samples were acid digested using hot block digestor in order to determine the heavy metal content. Principal component analysis (PCA) was done to highlight the relationship between variables (environmental variables, heavy metals content in sediment and bivalves). For human health risk assessment, four species of bivalve (green mussel, pacific oyster, marsh clam and Asiatic hard clam) were collected and assessed for potential risk to human health due to heavy metal intoxication. It was found that, the heavy metal content in green mussel affected by season. However, some metal elements in sediment were not significantly different throughout the study period. Through the PCA, it was found that, heavy metals in the green mussel and sediment may have been contributed by anthropogenic sources (such as chemical residues, weathering processes and boating activities) in the bay. Moreover, pacific oyster (MPI=15.03) was the most heavy metal contaminated followed by green mussel (MPI=8.31), marsh clam (MPI=6.86) and Asiatic hard clam (MPI=5.79). The four species of bivalves were found to present health risk of heavy metal intoxication when consumed in excessive amount over a prolonged period.

ABSTRAK

Pencemaran Logam Berat dalam Sedimen dan Bivalvia Komersial Terpilih di Teluk Marudu

Teluk Marudu merupakan kawasan perikanan yang penting disebabkan kekayaan biodiversiti marinnya. Selain itu, daerah Kota Marudu telah dikenalpasti sebagai kawasan berpotensi dalam pertanian selain aktiviti-aktiviti eko-pelancongan. Walaubagaimanapun, aktiviti pertanian yang semakin membangun (terutamanya penanaman kelapa sawit) di mana penggunaan bahan kimia (baja, racun serangga dll.), telah dikecam menyebabkan kematian siput sudu yang dikultur di Tanjung Batu, Teluk Marudu. Kejadian tersebut telah memberi kesan yang teruk kepada akuakultur siput sudu yang ada di kawasan itu. Oleh itu, kajian ini dijalankan untuk mengukur kandungan logam berat di dalam bivalvia komersial yang ada di Teluk Marudu dan untuk menyiasat hubungannya dengan pembolehubah physicokimia. Selain itu, kajian ini juga mengkaji sama ada kandungan logam berat dalam bivalvia tersebut berkemungkinan membawa risiko kesihatan kepada pengguna. Kajian ini dilaksanakan dengan mengambil sampel siput sudu (n=120) dan sedimen (n=36) setiap bulan selama 12 bulan. Sampel tersebut dicerna menggunakan acid dalam 'hot-block digestor' untuk mengetahui kandungan logam berat. Analisis komponen prinsipal (PCA) dijalankan untuk menerangkan lagi hubungan di antara pemboleh ubah (pembolehubah persekitaran, kandungan logam berat dalam sedimen dan bivalvia). Untuk kajian risiko kesihatan manusia, empat spesis bivalvia (siput sudu, tiram, lokan dan dalus) dianalisa untuk mengetahui potensi risiko kesihatan kepada manusia berkaitan dengan keracunan logam berat. Telah diketahui bahawa, kandungan logam berat di dalam siput sudu dipengaruhi oleh kesan musim. Walau bagaimanapun, kandungan beberapa unsur logam di dalam sedimen adalah tidak signifikan sepanjang tempoh kajian. Melalui PCA, kandungan logam berat dalam siput sudu dan sedimen berkemungkinan daripada sumber antropogenik (seperti sisa kimia, proses hakisan dan aktiviti bot) di teluk itu. Lebih daripada itu, tiram (MPI=15.03) didapati paling tercemar dengan logam berat diikuti oleh siput sudu (MPI=8.31), lokan (MPI=6.86) dan dalus (MPI=5.79). Keempat-empat spesis bivalvia itu didapati boleh menyebabkan risiko kesihatan akibat keracunan logam berat kepada manusia sekiranya dimakan pada jumlah yang berlebihan dalam jangkamasa yang panjang.

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°C	-	degree celcius
%	-	percent
Σ	-	sigma (summation notation)
µg	-	microgram
µg/L	-	microgram per litre
A	-	Angstroms (unit for atomic radii)
C or Cf	-	heavy metal content in sample
cm	-	centimeter
g	-	gram
g/cm^3	-	gram per cubic centimeter
mg	-	milligram
ml	-	millilitre
mg/L	-	milligram per litre
mg/kg	-	milligram per kilogram
m/min	-	metre per minute
kg	-	kilogram
N	-	normal (unit for normality)
n	-	number of sample

LIST OF ABBREVIATIONS

ANOVA	-	analysis of variance
As	-	arsenic
Bdl	-	below detection limit
Ca	-	calcium
Cd	-	cadmium
CKDu	-	chronic kidney disease of unknown etiology
CCME	-	Canadian Council of Minister of Environment
Co	-	cobalt
Cr	-	chromium
Cu	-	copper
DES	-	Department of Environmental Services
DNA	-	deoxyribonucleic acid
DO	-	dissolved oxygen
DOE	-	Department of Environment
DOF	-	Department of Fisheries
ED	-	Exposure duration
EF	-	Exposure frequency
EU	-	European Union
FAO	-	Food and Agriculture Organization
Fe	-	ferrum/ iron
Hg	-	mercury
HQ	-	hazard quotient
HRA	-	health risk assessment
IR	-	ingestion rate
ISQGs	-	interim sediment quality guidelines
ITOPF	-	International Tanker Owners Pollution Federation
Kg.	-	*kampung*
Li	-	lithium
Mg	-	magnesium
Mn	-	manganese
Mo	-	molybdenum

MPI	-	metal pollution index
NAS	-	National Academy of Sciences
NEM	-	northeast monsoon
NOAA	-	National Oceanic and Atmospheric Administration
NRCC	-	National Research Council Canada
Pb	-	plumbum/lead
PCA	-	principal component analysis
Ppt	-	part per thousand
RFD	-	oral reference dose
ROS	-	reactive oxygen species
Se	-	selenium
Sg.	-	*Sungai*
Sp.	-	species
SPSS	-	Statistical Package for the Social Sciences
SQGs	-	sediment quality guidelines
SRM	-	standard reference material
SWM	-	southwest monsoon
TA	-	average exposure
Tg.	-	*Tanjung*
THI	-	total hazard index
THQ	-	total hazard quotient
TOC	-	total organic carbon
UMS	-	Universiti Malaysia Sabah
UNEP	-	United Nations Environment Programme
UNESCO	-	United Nations Educational, Scientific and Cultural Organization
USA	-	United State of America
V	-	vanadium
WAB	-	average body weight
WHO	-	World Health Organization
Zn	-	zinc

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CHAPTER 1

INTRODUCTION

1.1 Bivalve Aquaculture

Bivalves are sessile animal that usually inhabit the coastal area. The sessile mode of life is attained either by byssal attachment or by valve fixation to hard substrates (Hrs-Brenko and Legac, 2006). To the coastal community, bivalves serve as low cost protein provider and other important nutrient such as omega-3.

Many species of bivalves such as oysters, cockles, mussels and clams are known to be commercially important in Malaysia. Particularly in Perak, Selangor and Penang, the production areas for cockle are identified due the presence of extensive mudflat area (Kechik, 1995) which is about 4000 to 5000 hectares (Liong *et al.*, 1988). In the aquaculture industry, bivalve production has contributed singnificantly particularly to the brackish aquaculture production. In 2012, the brackish aquaculture production in Malaysia including cockles and mussels has contributed 139,129.51 tonne which valued RM 1,566.78 million (Yusoff, 2015). Meanwhile, the estimated gross production of oysters and clams in Sabah was reported to be continuously increased from the year 2012 to 2016. On the other hand, the production of mussels was, however, fluctuated throughout the same period (Table 1.1). The statistic shown in Table 1.1 proves that the demand for bivalve is increasing either to fulfil the local demand (Hamli *et al.*, 2012) or the export market.

Table 1.1: Estimated gross bivalve production in Sabah from 2012 to 2016.

Bivalves\Year	Tonne				
	2012	**2013**	**2014**	**2015**	**2016**
Mussels (*Siput sudu*)	1.95	1.74	2.48	2.0	2.26
Oysters (*Tiram*)	649.01	668.65	727.42	757.23	767.88
Clam (*Lokan*)	25.0	26.47	32.27	37.51	38.92

Source: DOF Sabah (2012- 2016)

The pressure faced by aquaculture industry in Malaysia is generally caused by the increasing global demand of aquaculture products in countries like China and India, where economic improvement is rapidly going on (Yusoff, 2005). Consequently, this condition leads to severe depletion in the fishery resources. In addition, environmental pollution as well as the impact of climate change may also contribute in adding pressure on the overall fisheries production in Malaysia (Yusoff, 2005). In case of cockle production, a decline has been observed due to fully utilization of mudflat area which hinders further expansion (FAO, 2009).

In order to satisfy the increasing demand for shellfish, aquaculture is seen as a smart alternative to prevent the fishery production from solely depends on wild catch. In case of bivalve production in Sabah, Marudu Bay has been gazetted by the Fisheries Department of Sabah as bivalve aquaculture development zone (Denil *et al.*, 2017b) where green mussel (*Perna viridis*) is prominently cultured. The green mussel culture was first introduced in the bay in the year 2000 using broodstock transplanted from Johor (Tan and Ransangan, 2015; 2016). However, the production of the mussel in Marudu bay began to decline in late 2009 due to unexplained mass mortality (Taib *et al.*, 2016). Besides infectious diseases, mussel mortality can also be caused by severe chemical contamination of the culture site. Contamination does not only cause the mussels to experience mortality but also market rejection.

One of the most leading contaminants in the coastal area or bays is heavy metal. Positive relationship between heavy metal contamination in the habitat and that in bivalves is evidenced from many studies (Gregori *et al.*, 1994; Rzymski *et al.*, 2014).

Marudu Bay is a productive bay and becomes important area for fishing activity. Its surrounding is also extensively developing for human settlement and for commercial scale palm oil plantation. In Malaysia, oil palm plantation activities have been reported to cause contamination of Cd, Pb and As in the soil due to the usage of fertilizers (Atafar *et al.*, 2010). This is supported by another study conducted in Felda Jengka, Pahang, which blamed the usage of fertilizers in the oil palm plantation for the accumulation of Cu and Zn in the soil. Moreover, herbicides such as diuron is prominently used in oil palm plantation to get rid of the broadleaf weeds but is often associated with risk of food contamination (Muhamad *et al.*, 2010).

It is anticipated that the development which are taking place in the bay would promote leaching of chemicals from fertilizers, pesticides and herbicides used in the plantation sites and can become worse during rainy season. Consequently, aquatic organisms especially bivalves which are filter feeders may accumulate and biomagnify those contaminants in their body, and cause heavy metal intoxication to human upon consumption. The initial effects of heavy metal toxicity are often similar to common sicknesses such as fatigue, headaches, allergic reactions and diarrhea (Jaishankar *et al.*, 2014). However, prolonged exposure to heavy metal may cause serious diseases such as organ damages, various kind of cancers and in a worst- case scenario, it can be fatal (Javed and Usmani, 2013; Jaishankar *et al.*, 2014; Tornero and Hanke, 2016). This is because, although heavy metals do not have any biological role, upon its introduction to the human body, they may interfere with various metabolic processes and hinder the proper functioning of cellular components (Jaishankar *et al.*, 2014).

1.2 Problem Statement

Marudu Bay is a natural habitat for many marine organisms. This is owed to the richness of mangrove forest in the bay which serves as ideal nursery and foraging ground. It has been reported that, Marudu Bay sheltered as many as 22 aquatic invertebrate species, 36 fish species, 74 birds species, 4 reptiles species and 4 mammals species (Zakaria and Rajpar, 2015).

Due to the abundance of marine organisms, Marudu Bay has become an important fishing ground especially for the fishers community of Kota Marudu. Other than that, farming activity of bivalves especially green mussel and oyster have been going on in the bay for sometimes. Green mussel aquaculture was first introduced in the bay by outsourcing breeding stock from Johor in the late 1990's (Tan and Ransangan, 2015; Taib *et al.*, 2016). A few years after, the green mussel culture in Marudu Bay became commercially important that attracted investors to invest in the industry. Unfortunately, the production of the green mussel suffered loss when massive mortality occurred in the late 2009. Despite the mass declination of the green mussel production in the bay, other bivalve species such as oysters, marsh clam and Asiatic hard clam, fortunately, were not affected.

Other than holding potential for eco-tourism activities such as mangrove cruising and educational tourism (Hanum *et al.*, 2012), Kota Marudu district has been identified as potential area for agriculture activities. The land area within the main districts of Kudat, Pitas and nearby Kota Marudu, are dominated by agricultural activities such as coconut and oil palm plantations (Liew-Tsonis *et al.*, 2012), and lately, rubber plantation. This leads to vast land clearing to give way to agriculture activity especially oil palm plantation. This is supported by previous study (Hanum *et al.*, 2012) who claimed that one-third of the mangrove area in Marudu bay has been cleared and converted to meet agriculture activities (rubber and oil palm plantation).

Since, these plantations use huge amount of fertilizers, herbicides and pesticides, the fate of the fisheries resources of Marudu Bay is doubted. Rumours are already going on, whereby fishers are blaming the oil palm plantation for the high mortality of culture green mussel in the bay. However, these claims have never been backed by scientific data.

Therefore, the purpose of the study is to measure the level of metals in commercially important bivalves originating from Marudu bay and to investigate its relationship with the physicochemical variables. Other than that, this study also examines if the level of metals in the bivalves can possibly bring about health risk to consumers.

1.3 Significance of Study

Current study determines the possibility of heavy metal contamination in the culture green mussel and their possible sources. It also reviews the relationship between physicochemical parameters and the bioaccumulation of heavy metals in the culture green mussel as well as in sediment. Moreover, this study provides baseline data for the possible health risk associated with heavy metal intoxication due to bivalve (originated from Marudu bay) consumption.

1.4 Objectives of Study

The specific objectives of this study are as follows:

1. To investigate the relationship between heavy metal content in sediment and the culture green mussel with the environmental variables.
2. To compare the levels of heavy metals in culture green mussel with three other bivalve species which are commercially important and extensively harvested from Marudu bay.
3. To assess the possible health risk due to heavy metal intoxication associated with consumption of these bivalves originating from Marudu bay.