UNIVERSITI PUTRA MALAYSIA

HUMAN HEALTH, ECOLOGICAL RISK AND HEAVY METAL POLLUTION ASSESSMENT IN LANGAT RIVER BY TRANSPLANTATION OF Corbicula javanica

WONG KOE WEI

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By

WONG KOE WEI

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

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HUMAN HEALTH, ECOLOGICAL RISK AND HEAVY METAL POLLUTION ASSESSMENT IN LANGAT RIVER BY TRANSPLANTATION OF *Corbicula javanica*

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Chairman: Associate Professor Yap Chee Kong, PhD
Faculty: Science

Heavy metal pollution has become a major concern nowadays because these pollutants are not biodegradable and may be biomagnified and bioaccumulated and could potentially be harmful to biota over certain level. Therefore, assessment of the potential risks of heavy metal pollutions to ecosystem and human health are necessary. The freshwater bivalve clam *Corbicula javanica* (Family: Cyrenidae) is abundant, easily identifiable, sedentary life style and tolerant to the increased bioavailable heavy metals in the environment, besides having large enough for analysis and the potential to reflect bioavailability. Therefore, it is suitable to be a good biomonitor. The first objective of this study was to assess the heavy metal pollution in the Langat River by transplantation of *C. javanica* from upstream pristine site to downstream polluted sites of the river. The freshwater clams *C. javanica* was transplanted from upstream Pangsun (PS) to Semenyih (SM), Dusun Tua (DT) and Kajang (KJ). Heavy metal in *C. javanica* and surface sediments was analysed by using FAAS for Cu, Fe, Ni, Pb and Zn, and ICP-MS for As, Co, Cd, Cr and Mn. Besides, the geochemical speciation of the heavy metals in surface sediments were analysed by using sequential extraction technique (SET). The second objective was to determine the ecological risk assessment of heavy metals in the surface sediments of the Langat River by comparison with sediment quality guidelines (SQGs), and calculations of geoaccumulation index (I_{geo}), enrichment factor (EF), potential risk for individual metal (Er), potential ecological risk index (PERI) and combined pollution index (CPI). Lastly, the third determine the human health risk assessment via the consumption the clam collected from the transplantation study in the Langat River by comparison with food safety guidelines and calculation of estimated daily intake (EDI) and target hazard quotients (THQ).

From the results, the higher concentration of Co, Cr, Fe, Mn, Ni and Zn in the sediment of PS, are associated with the periodic discharge of dam impoundment of Pangsun Dam. Higher As and Cd in SM, are related to the use of phosphate fertilizers and pesticides in the oil palm plantation nearby the particular site. Higher Pb, and Cu were KJ, are associated to the domestic waste emission in urban area. The clams' Total Soft Tissue (TST) has the ability to accumulate Fe, Zn, Cu, Mn, Co, Cr, As and Cd and the shell has the ability to accumulate higher level of Pb and Ni. The correlation analysis revealed that 8 metals (As, Co, Cr, Fe, Mn, Ni, Pb and Zn) in TST while 5 metals (As, Cd, Cr, Fe and Mn) in shell have positively and significantly correlation

with respective metal concentration in sediment, indicating the clams is a good biomonitor of the metal levels. Geo-chemical fractions extraction revealed that As, Cr, Fe, Mn, Ni and Pb in the sediment were dominated by the resistant (R) fraction suggested that it may be less potentially bioavailable and be poorly mobilized. However, care should be taken for As since its concentrations are higher than previous studies. Zn and Cd were found to be abundant in Acid Reducible fraction (AR) in all sites, this indicated that these two metals were highly mobilized with potential environmental consequences. However, the impact might be limited since the concentration of Zn and Cd are not higher than previous studies. The difference in geochemical fraction distribution in Co indicates four sites may have different source. Cu in KJ are abundant in oxidizable-organic (OO) fraction. This indicates that the Cu in KJ were organic in nature and it is easily absorbed. Various geochemical indexes for a single metal pollutant (I_{geo}, EF, C_f, and Er) all agreed that Cd, Co, Cr, Cu, Fe, Mn, Ni and Zn are not likely to cause adverse effect to the river ecosystem, but As and Pb could pose a potential ecological risk to the river ecosystem. When all metals were accounted, all indexes (C_d, CPI and PERI) showed that overall metal concentrations in Langat River are still within safe limit. The values of EDI of *C. javanica* were found to be all lower than oral reference dose (RfD) guidelines for all metals. Furthermore, the calculated THQ and total THQ were found to be less than 1. Therefore, there will be no non-carcinogenic human health risk on both average and high level consumption of total soft tissue of *C. javanica*.

The finding of present study indicated that TST and shell of *C. javanica* are good biomonitors of heavy metal bioavailability. We deduced that the elevated heavy metals of PS, SM and KJ are associated with dam discharge, use of fertilizers or pesticides and domestic waste discharge respectively. Ecological risk assessment indicated that most of the metals are unlikely to cause adverse effects except As and Pb. Human health risk assessment indicated that prolonged consumption of *C. javanica* in this river are safe.

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

PENILAIAN RISIKO KESIHATAN MANUSIA, EKOLOGI DAN PENCEMARAN LOGAM SURIH DI SUNGAI LANGAT DENGAN TRANSPLANTASI *Corbicula javanica*

Oleh

WONG KOE WEI

Julai 2016

Pengerusi: Profesor Madya Yap Chee Kong, PhD
Fakulti: Sains

Pencemaran logam surih menjadi kebimbangan utama sekarang sebab pencemar ini tidak terbiodegradasi, dapat biomaknifikasi dan bioakumulasi serta berpotensi membahayakan unsur biota dalam ekosistem lepas tahap tertentu. Oleh itu, penilaian potensi risiko kepada ekosistem dan juga kesihatan manusia disebabkan pencemaran logam surih adalah diperlukan. Kerang air tawar *Corbicula javanica* (Famili: Cyrenidae) berpotensi digunakan sebagai bio-pemantau kerana populasinya yang banyak, mudah untuk dikenal pasti, cukup besar untuk dianalisis, gaya hidup tidak aktif dan berupaya untuk bertoleransi dengan peningkatan logam berat yang bio-tersedia di alam sekitar, selain mempunyai cukup besar untuk analisis logam dan juga berpotensi untuk mencerminkan bio-kesediaan logam surih. Objektif pertama kajian ini adalah untuk menilai pencemaran logam berat di Sungai Langat melalui pemindahan *C. javanica* dari tapak bersih di hulu (Pangsun, PS) ke hilir sungai yang tercemar [Semenyih (SM), Dusun Tua (DT) dan Kajang (KJ)]. Kaedah FAAS (untuk Cu, Fe, Ni, Pb dan Zn)dan ICP-MS (As, Co, Cd, Cr dan Mn) untuk telah digunakan untuk menganalisis kandungan logam surih di dalam *C. javanica* dan juga sedimen permukaan sungai. Fraksi geokimia logam surih dalam sedimen juga dianalisis dengan *Sequential Extraction Technique* (SET). Objektif kedua kajian ini adalah untuk menjalankan penilaian risiko ekologi logam surih dalam sedimen permukaan Sungai Langat dengan perbandingan dengan panduan kualiti sedimen (SQGs) dan pengiraan *geoaccumulation index* (I_{geo}), *enrichment factor* (EF), *potential risk for individual metal* (Er), *potential ecological risk index* (PERI) dan *combined pollution index* (CPI). Akhirnya, objektif ketiga kajian ini adalah untuk menjalankan penilaian risiko kesihatan manusia melalui pemakanan kerang yang dikumpulkan dari Sungai Langat dalam kajian transplantasi ini. Penilaian risiko kesihatan manusia ini dilakukan dengan perbandingan dengan garis panduan kesihatan makanan dan pengiraan indeks estimated daily intake (EDI) dan target hazard quotients (THQ).

Dari keputusan, sedimen dari PS yang mengandungi Co, Cr, Fe, Mn, Ni dan Zn yang paling tinggi adalah berkaitan dengan pelepasan berkala air takungan empangan Pangsun. As dan Cd didapati tinggi di SM. Ia adalah berkaitan dengan penggunaan baja fosfat dan racun perosak di tapak penanaman kelapa sawit. Manakala, pelepasan sisa domestik menyebabkan peningkatan Pb dan Cu di KJ. Analisis kolerasi menunjukkan kolerasi signifikan ($p<0.05$) dan positif di antara 8 elemen logam di jumlah tisu lembut (TST) (As, Co, Cr, Fe, Mn, Ni, Pb dan Zn) dan 5 elemen logam (As,

Cd, Cr, Fe dan Mn) di cangkerang dengan kandungan logam di sedimen. Ini menunjukkan bahawa kerang ini adalah agen biomonitor untuk pencemaran logam surih yang baik. Pengekstrakan fraksi geokimia menunjukkan dominasi fraksi *Resistant* (R) untuk As, Cr, Fe, Mn, Ni and Pb. Ini menunjukkan logam-logam tersebut adalah sukar dimobilisasi dan kurang bioavailabiliti. Namun, perhatian masih perlu dibagi kepada As memandangkan kandungan As di sedimen lebih tinggai berbanding dengan kajian dahulu. Kandungan Zn dan Cd di semua tapak tertumpu di fraksi *Acid Reducible* (AR). Ini menunjukkan kedua-dua logam ini mempunyai mobiliti yang tinggi dan berpotensi mengakibatkan kesan negatif alam sekitar. Tetapi, kesan mobiliti tinggi logam tersebut mungkin terhad kerana kandungan Zn dan Cd bukanlah lebih tinggi daripada kajian dahulu. Perbezaan agihan kandungan Co antara fraksi geokimia menunjukkan Co di empat tapak mungkin berasal dari sumber yang berlainan. Cu di KJ tertumpu di fraksi *oxidizable organic* (OO), menunjukkan kebanyakan logam ini adalah bersifat organik dan senang diserap. Semua indeks-indeks geokimia untuk satu pencemar (I_{geo}, EF, C_f, dan Er) bersetuju bahawa Co, Cr, Cu, Fe, Mn, Ni and Zn tidak akan membawa kesan buruk kepada ekosistem sungai. Indeks-indeks pencemar tergabung (C_d, CPI and PERI) menunjukkan bahawa kandungan logam surih secara keseluruhan adalah dalam had selamat. Nilai EDI untuk *C. javanica* dijumpai lebih rendah daripada garis panduan *oral reference dose* (RfD) untuk semua logam yang dikaji. Tambahan pula, pengiraan THQ dan jumlah THQ menunjukkan nilai kurang daripada 1. Oleh itu, tiada risiko kesihatan manusia bukan karsinogenik akan dialami oleh pengguna tahap sederhana dan tinggi TST *C. javanica*.

Penemuan kajian ini menunjukkan TST dan cangkerang *C. javanica* adalah bio-pemantau yang baik untuk menunjukkan bioavailabiliti logam surih. Kita menyimpulkan bahawa kandungan logam yang tinggi di PS, SM, DT dan KJ adalah berkaitan dengan pelepasan empangan, penggunaan baja dan racun perosak dan pelepasan sisa domestik. Penilaian risiko ekologi menunjukkan selain daripada As dan Pb, semua logam lain tidak akan menyebabkan kesam buruk kepada ekosistem. Penilaian risiko kesihatan manusia menunjukkan penggunaan kepanjangan *C. javanica* adalah selamat.

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I certify that a Thesis Examination Committee has met on 21 July 2016 to conduct the final examination of Wong Koe Wei on his thesis entitled "Human Health, Ecological Risk and Heavy Metal Pollution Assessment in Langat River by Transplantation of *Corbicula javanica*" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

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Professor
Faculty of Science
Universiti Putra Malaysia
(Chairman)

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Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Internal Examiner)

Mohd. Talib Latif, PhD
Associate Professor
National University of Malaysia
Malaysia
(External Examiner)

ZULKARNAIN ZAINAL, PhD
Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 28 September 2016

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

Yap Chee Kong, PhD
Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Rosimah Nulit, PhD
Senior Lecturer
Faculty of Science
Universiti Putra Malaysia
(Member)

Mohd Suhaimi Hamzah, PhD
Senior Researcher
Waste and Environmental Technology Division
Malaysian Nuclear Agency
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LIST OF ABBREVIATIONS

Zn	Zinc
Cu	Copper
Pb	Lead
Ni	Nickel
Fe	Iron
Mn	Manganese
Co	Cobalt
Cr	Chromium
As	Arsenic
Cd	Cadmium
PS	Pangsun
SM	Semenyih
DT	Dusun Tua
KJ	Kajang
FAAS	Flame Atomic Absorption Spectrometry
ICP-MS	Induced Coupled Plasma-Mass Spectrometry
SET	Sequential Extraction Technique
EFLE	Easily, freely or leachable and exchangeable
PERI	Potential Ecological Risk Index
EDI	Estimated Daily Intakes
CPI	Combined Pollution Index
THQ	Target Hazard Quotient
RfD	Oral reference dose
CI	Condition index
DO	Dissolved oxygen
Cond	Electrical conductivity
ALM	Average level mollusc consumer
HLM	High level mollusc consumer
EF	Enrichment factor
CV	Coefficient of variation

CHAPTER 1

INTRODUCTION

Heavy metals exist naturally in large variations in concentration. Nowadays however, due to rapid development and industrialization, anthropogenic heavy metals introduced to the ecosystem causing heavy metal pollution. There are several sources including the periodic discharge of river dam impoundment, agricultural activities and discharge of domestic wastes are identified as a possible source of anthropogenic heavy metals (Wang *et al.,* 2015; Fuge, 2013). Much of the heavy metals with industrial significance have known to toxic not only to human beings but to other organisms. The bioconcentration of certain pollutants including heavy metals may increase significantly through the food chains by the effect of bioaccumulation and biomagnification. The heavy metals as a pollutant are non-degradable in nature, in contrast of organic pollutants that can be degenerated, producing carbon dioxide and water as by-product (Lim *et al.,* 2013; Khan *et al.,* 2004; Gupta *et al.,* 2001). Therefore, it will be wise that heavy metal pollution be constantly monitored. Rapid development has increases the introduction of heavy metal pollutants into the environment. Along with the increasing of the industrial activity across the globe, development of a way of monitoring the pollution level of the environment with the hope of controlling, reducing and ultimately solving the pollution has been crucial to any nations in the world.

Malaysia underwent mass industrialization since independence. the rapid modernization in Malaysia has caused the degradation of river quality, depletion of river resources, public health risks and loss of river biodiversity due to artificial alteration of river environment. The river ecosystem are blessed with immense biodiversity, biological productivity and high assesibility. It is one of the crucial natural asset to human society. The high economic value of a clean river is relied on their suitability for aquaculture activities, food source, recreation, ecotourism and as a genetic resource (Lim *et al*., 2013). Therefore, in order to maintain the health of such valueable asset, constant monitoring of the river ecosystem health and level of potential pollutions are a crucial part of a broader effort of ecosystem preservation and conservation.

The utilization of aquatic organism to monitor trace metal abundance and bioavailability has been well established (Phillips *et al*., 1988). Various aquatic organisms existing in aquatic environment can be potentially useful as biomonitor of heavy metal pollution, including fishes, molluscs, aquatic plants and algae (Torres *et al*., 2008). The idea of using the molluscs as biomonitor to monitor heavy metal pollution was proposed by Goldberg (1975) as Mussel Watch Program. Its method has since become a common practice in biological monitoring or biomonitoring of the heavy metal pollution (Yap *et al.* 2007). The Goldberg's (1975) study has also established *Perna viridis* as a biomonitor species in various place in the Asia-pacific coastal region (Yap, 2012). But the use of *Perna viridis* are limited to coastal area. Therefore, there is a necessity to established a freshwater based biomonitor in order to implement Goldberg (1975) concept of biomonitoring in inland freshwater context.

The concept of large scale biomonitoring was proposed by Goldberg *et al.* (1978) as 'Mussel Watch' program in order to examine the spatial and temporal trends in chemical pollution in estuarine and coastal environment. This program was used bivalves as sentinel organism that is capable of sensing trends in the concentration of numerous marine pollutants (Goldberg *et al.*, 1978; Zhou *et al.*, 2008). Due to their ability of mussels or other bivalves to accumulate pollutants from their living habitat in quantities that are many times higher than the background levels, they are a popular choice for biomonitoring of aquatic heavy metal pollution (Cheng and Yap, 2015). Molluscs also exhibit greater spatial sensitivity to heavy metal concentration in the environment, therefore it can be considered as a more reliable method to point out the sources of heavy metal in the environment that are bioavailable (Goldberg *et al.*, 1978; Koide *et al.*, 1982; Thomson *et al.*, 1984; Hamed and Emara, 2006). The heavy metal uptake and accumulation into the tissue of the molluscs are in proportional to the degree of environmental pollution signifies their ability as biomonitor of heavy metal pollution (Goldberg *et al.*, 1983; Elder and Mattraw, 1984; Bu-Olayan and Subrahmanyam, 1997; Yap, 2012).

In Malaysia, biomonitoring works and studies of the background levels of heavy metals were mainly focused on marine mussels and intertidal snail that can be widely found in the on the coastal and intertidal area in Peninsular Malaysia (Cheng and Yap, 2015; Yap *et al.*, 2012, 2002). For better monitoring of the biomonitor candidate for freshwater region located within continent should be established. Freshwater clam *Corbicula javanica* were chosen due to its availability at the site of study. Although *C. javanica*'s genus sibling, *Corbicula fluminea* were well studies as a biomonitor in various countries due to its invasive nature, there are lack of information about *C. javanica* in Malaysia, and there is no literature from this region ventured the possibility to use *C. javanica* as a potential biomonitor of heavy metal pollution in Malaysia, nor in other countries within South East Asia. Being a genus sibling of a well-established biomonitor, it has potential to be a potential biomonitor for heavy metal pollution in this region. Since *C. javanica* is not an invasive species like *C. fluminea*, there were some places around study area which *C. javanica* cannot be found, therefore transplantation of the *C. javanica* from sites where this species can be found to other sites, were done to cope with the uneven distribution of this species.

Comparing the heavy metal concentration between river surface sediment and the hard and soft tissue of *C. javanica* are crucial. The potential risk of the heavy metal poses to both ecosystem and human beings in both sediments and edible tissue of *C. javanica* are equally important. Therefore, ecological risk and human health risk assessment were done by calculation of several established indices. Since the main objective of ecotoxicological study are to evaluate the impact of the alteration of environmental chemical and physical properties to ecosystem and human health. The assessment of the risk caused by a pollutant to both ecosystem and human health would be a crucial aspect of ecotoxicology. Ecological risk assessment (ERA) were done in order to assess the ecological impact of heavy metal. Enrichment factors (EF), geo-accumulation index (I_{geo}), potential ecological risk index (PERI) were calculated based on the findings. By these indices, the degree of heavy metal pollution can be determined. Human health risk assessment was done to determine the potential health risks related with trace metal contamination via the consumption of edible parts of the sentinel organism. Human health risk assessment was usually achieved by screening level risk assessment that are usually conducted through consideration of established international dietary guidelines and determination of estimated daily intake (EDI) and target hazard quotient (THQ) (Cheng and Yap, 2015). The estimated daily intake (EDI) and target hazard quotient (THQ) were calculated to assess the potential impact of the heavy metal concentration to human health based on the amount of consumption.

1.1 Objectives of Study

The main objectives of this study were to:

1. To assess the heavy metal pollution in the Langat River Basin by transplantation of *Corbicula javanica* from upstream pristine site to downstream polluted sites of the river.
2. To determine the ecological risk assessment of heavy metals in the surface sediments of the Langat River.
3. To determine the human health risk assessment via the consumption the clam *C. javanica* collected from the transplantation study in the Langat River.

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