UNIVERSITI PUTRA MALAYSIA

HEAVY METALS POLLUTION AND HEALTH RISK IN FISH IN SUNGAI GALAS KELANTAN, MALAYSIA

ZARITH SUFIANI BINTI BAHAROM

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By

ZARITH SUFIANI BINTI BAHAROM

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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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Master of Science

HEAVY METALS POLLUTION AND HEALTH RISK IN FISH IN SUNGAI GALAS, KELANTAN, MALAYSIA

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ZARITH SUFIANI BINTI BAHAROM

August 2017

Chair: Mohd Yusoff Bin Ishak, PhD
Faculty: Environmental Studies

A study on water surface and fish tissue for heavy metals concentrations was carried out at five sampling stations along Sungai Galas, Kelantan. Concentrations of heavy metals namely copper (Cu), zinc (Zn), lead (Pb), manganese (Mn) and nickel (Ni) were determined in the muscle, liver and gills of *Hampala microlepidota* (sebarau), *Barbonymus schwanenfeldii* (lampam sungai), *Mystacoleucus obtusirostris* (sia), *Hemibagrus nemurus* (baung) and *Cyclocheilicthys apogon* (temperas). The samples were collected by using gill nets during the wet (August to January) and dry seasons (February to September). The fish organs and water samples were analysed using Inductively Coupled Plasma- Mass Spectrometry (ICP-MS). The results of this study showed that the level of Zn, Cu and Pb concentrations at Dabong and Kuala Geris were higher in wet season when compared to dry season. There was positive correlation between heavy metal concentrations in fish tissues with heavy metal concentrations in water and physiochemical properties of water (p<0.05). The heavy metal concentrations found in muscles, liver and gills of all studied fish ranged from Cu: 0.30-0.39, Zn: 4.44-8.08, Pb: 0.17-0.51, Ni: 0.26-0.99, and Mn: 0.38-0.58 mg/kg wet weight; Cu: 2.19-6.16, Zn: 13.5-41.6, Pb: 2.06-7.14, Ni: 1.09-4.59, and Mn: 2.71-12.1 mg/kg wet weight, and Cu: 0.45-1.68, Zn: 10.49-40.99, Pb: 1.38-4.53, Ni: 1.25-5.75, and Mn: 8.81-44.11 mg/kg wet weight, respectively. The mean concentrations of Cu, Zn, Pb, Ni and Mn were significantly (p<0.05) different between fish organs and seasons. The liver samples showed highest levels of heavy metals compared to the gills and muscle. The Cu, Zn, Ni and Mn concentrations in liver and gills of *M. obtusirostris* were higher than other species. Muscles of *H. macrolepidota* (mg/kg wet weight) were found to have the highest concentrations of Zn (8.08-13.04) and Mn (0.41-0.58); while muscle of *H. nemurus* was detected having the highest concentration of Ni (0.72-1.67) and Pb (0.27-0.66). There was a positive correlation between Cu and Zn concentration in muscle with increasing weight and length of fish (p<0.05). The Pb concentrations at Gua Musang in muscles of *B. schwanenfeldii*, and *C. apogon* were generally higher than safety thresholds values recommended by Commission Regulation (EC, 2001) while concentrations of Pb at Gua Musang in *H. nemurus* exceeded the limit set by Food and Drug Administration, (USFDA, 1983), Food and Agriculture Organization (FAO, 2003) and EC (2001). Only Ni levels in *C. apogon* at Gua Musang did not exceed the limit stipulated by World Health Organization (WHO, 1985). All investigated metals in surface water (below 10 cm from

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surface) were significantly lower than the limits sets by WHO (2011) and class II National Water Quality Standards of Malaysia (NWQS), except for Pb which exceeded WHO (2011) and Class II NWQS standards, while Mn fell under Class III in NWQS. Total hazard quotient values indicated there was no public health hazard posed by consumption of the investigated fish species concerning all metals, for child and adult because the THQ is < 1.0.

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

PENCEMARAN LOGAM BERAT DAN RISIKO KESIHATAN DALAM IKAN DI SUNGAI GALAS, KELANTAN, MALAYSIA

Oleh

ZARITH SUFIANI BINTI BAHAROM

Ogos 2017

Pengerusi: Mohd Yusoff Bin Ishak, PhD
Fakulti: Pengajian Alam Sekitar

Satu kajian menentukan kepekatan logam berat dalam air dan tisu ikan telah dijalankan di lima stesen persampelan sepanjang Sungai Galas, Kelantan. Kepekatan logam berat iaitu tembaga (Cu), zink (Zn), plumbum (Pb), mangan (Mn) dan nikel (Ni) telah ditentukan dalam otot, hati dan insang ikan spesis *Hampala microlepidota* (sebarau), *Barbonymus schwanenfeldii* (lampam sungai), *Mystacoleucus obtusirostris* (sia), *Hemibagrus nemurus* (baung), dan *Cyclocheilicthys apogon* (temperas). Persampelan ikan dilakukan pada musim kering (Ogos hingga Januari) dan hujan (Februari hingga September) menggunakan pukat. Organ-organ ikan dan sampel air telah dianalisis menggunakan Spektrometer Jisim- Gandingan Aruhan Plasma (ICP-MS). Keputusan kajian ini menunjukkan bahawa tahap kepekatan Zn, Cu dan Pb di Dabong dan Kuala Geris adalah lebih tinggi pada musim basah berbanding musim hujan. Terdapat korelasi positif dalam kepekatan logam berat dalam otot ikan dengan kepekatan logam berat dalam air dan sifat fisiokimia air ($p < 0.05$). Kepekatan logam berat yang terdapat dalam otot, hati dan insang ikan yang dikaji adalah dari Cu: 0.30-0.39, Zn: 4.44-8.08, Pb: 0.17-0.51, Ni: 0.26-0.99 dan Mn: 0.38-0.58 mg/kg berat basah; Cu: 2.19-6.16, Zn: 13.5-41.6, Pb: 2.06-7.14, Ni: 1.09-4.59 dan Mn: 2.71-12.1 mg/kg berat basah dan Cu: 0.45-1.68, Zn: 10.49-40.99, Pb: 1.38-4.53, Ni: 1.25-5.75 dan Mn: 8.81-44.11 mg/kg berat basah, masing-masing. Kepekatan Cu, Zn, Pb, Ni dan Mn antara organ ikan dan musim menunjukkan perbezaan ketara. Tahap kandungan logam berat dalam sampel hati lebih tinggi berbanding insang dan otot. Kepekatan Cu, Zn, Ni dan Mn dalam hati dan insang *M. obtusirostris* lebih tinggi berbanding spesies ikan lain. Otot tisu *H. macrolepidota* mempunyai kepekatan tertinggi (mg/kg berat basah) dalam Zn (8.08-13.04) dan Mn (0.41-0.58), manakala *H. nemurus* dikesan mempunyai kepekatan tertinggi bagi Ni (0.72-1.67) dan Pb (0.27-0.66). Korelasi positif ($p < 0.05$) dikesan dalam kepekatan Cu dan Zn dalam otot ikan terhadap peningkatan dalam berat badan dan panjang ikan. Kepekatan Pb di Gua Musang dalam otot tisu *B. schwanenfeldii* dan *C. apogon* secara amnya lebih tinggi daripada nilai had kritikal yang disarankan oleh *Commission Regulation* (EC, 2001), manakala kepekatan Pb di Gua Musang dalam *H.nemurus* telah melebihi had yang telah ditetapkan oleh *Food and Drug Administration* (USFDA, 1983), *Food and Agriculture Organization* (FAO, 2003) dan EC (2001). Hanya kepekatan Ni dalam *C. apogon* di Gua Musang tidak melebihi had yang ditetapkan oleh *World Health Organization* (WHO, 1985). Hampir semua logam berat yang dikaji di dalam permukaan

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air (bawah 10cm dari permukaan air) didapati jauh lebih rendah daripada had yang ditetapkan oleh WHO (2011) dan kelas II Piawaian Kualiti Air Kebangsaan Malaysia (NWQS). Kepekatan Pb telah melebihi had WHO (2011) dan Kelas II NWQS, manakala Mn telah melebihi had pada Kelas III dalam NWQS. *Hazard Quotient* menunjukkan tidak ada bahaya terhadap kesihatan pemakan spesies ikan yang disiasat bagi semua jenis logam kepada kanak-kanak dan dewasa kerana jumlah $THQ < 1.0$.

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I certify that a Thesis Examination Committee has met on 1 August 2017 to conduct the final examination of Zarith Sufiani Binti Baharom on her thesis entitled "Heavy Metal Pollution and Health Risk in Fish in Sungai Galas, Kelantan, Malaysia" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U. (A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

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Universiti Putra Malaysia
(Chairman)

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

Ahmad Abas Kutty, PhD
Professor
Universiti Kebangsaan Malaysia
Malaysia
(Extermal Examiner)

NOR AINI AB. SHUKOR, PhD
Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 28 September 2017

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Mohd Yusoff Ishak, PhD
Senior Lecturer
Faculty of Environmental Studies
Universiti Putra Malaysia
(Chairman)

Rohasliney Hashim, PhD
Senior Lecturer
Faculty of Environmental Studies
Universiti Putra Malaysia
(Member)

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LIST OF ABBREVIATIONS

APHA	American Public Health Association
ASTDR	Agency For Toxic Substances And Disease Registry
BOD	Biochemical Oxygen Demand
CO_2	Carbon Dioxide
COD	Chemical Oxygen Demand
cm	Centimetre
CRM	Certified Reference Material
E	East
DID	Drainage And Irrigation Department
DO	Dissolved Oxygen
DOE	Department Of Environment
DWAF	Department Of Water Affairs And Forestry
EC	Commission Regulation
ED	Exposure Duration
EF	Exposure Frequency
EPA	Environmental Protection Agency
FAO	Food And Agriculture Organization
FIR	Fish Ingestion Rate
g	Gram
gcm^{-3}	Gram Per Cubic Centimetre
H_2O_2	Hydrogen Peroxide
H+	Hydrogen Ions
HNO_3	Nitric Acid
HQ	Hazard Quotient
IARC	International Agency For Research On Cancer
ICP-MS	Inductively Coupled Plasma-Mass Spectrometry
IQ	Intelligent Quotient
JKOAK	Kelantan Network Of Orang Asli Villages
JUPEM	The Department Of Survey And Mapping Malaysia
JECFA	Joint FAO/WHO Expert Committee On Food Additives
km^2	Square Kilometre
m	Metre
mm	Millimetre
m^3s^{-1}	Cubic Metre Per Second
MFA	Malaysian Food Act
mg	Milligrams
mL	Milli Litres
mg/kg	Milligram Per Kilogram
mg/L	Milligram Per Litre
MOH	Malaysian National Standard For Drinking Water Quality
MPI	Metal Pollution Index
N	North
NH_3-N	Ammoniocal Nitrogen
NIST	National Institute Of Standards And Technology
NWQS	National Water Quality Standards For Malaysia
ppb	Parts-Per-Billion
USEPA	United States Environmental Protection Agency
USFDA	Food And Drug Administration
µm	Micrometre

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µg/g	Microgram Per Gram
µg/kg	Microgram Per Kilogram
ug/L	Microgram Per Litre
µmhos/cm	Micromhos Per Centimetre
µs/cm	Microsiemens Per Centimetres
PPLRNK	*Perbadanan Pembangunan Ladang Rakyat Negeri Kelantan*
RfDs	Oral Reference Doses
SD	Standard Deviation
SL	Standard Length
SS	Suspended Solids
TH	Target Hazard
THQ	Target Hazard Quotient
TL	Total Length
TSS	Total Suspended Solid
WHO	World Health Organization
wt	Weight
WQI	Water Quality Index

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CHAPTER 1

INTRODUCTION

1.1 General

Contamination of heavy metals in aquatic environment especially in developing countries continue to attract a lot of attention from environmental researchers due to its increasing input to water bodies. Its harmful characteristic of persistent, toxic and highly susceptible to bioaccumulation had caused heavy metals to be a serious pollutant problem in natural environment. Some of the horrifying tragedies associated with heavy metals poisoning include Itai-Itai disease and Minimata disease which were resulted from mishandling of industrial wastes.

Presence of heavy metals in aquatic ecosystems is associated with natural processes and anthropogenic activities (Kamaruzzaman et al. 2010). The natural heavy metals occur as a result of geological weathering of rocks and soils while heavy metals from anthropogenic activities comes from sources which include industrial and agriculture activities, soil erosion, disposal of effluents, chemicals runoff, and atmospheric deposition; all of which will then eventually be deposited into sediments in the water bodies (Ochieng et al. 2007; Guo et al. 2015).

In Malaysia, the river environment is an important surface water bodies which became an increasingly sought out subject due to increase intensification of pollutants caused by human activities. A lot of research has been done in relation to heavy metals contamination and the tolls it took on related subjects, particularly the aquatic environment, organisms and human beings. Fishes which are commonly consumed by humans serve as great indicator in the assessment of pollution on trace metals (Makedonski et al. 2017). These fishes that dwell in contaminated rivers will likely to accumulate deadly trace metals through food pyramid and could pose threat to humans later when consumed.

There are several metal and chemical industries as well as agriculture industries operating in area of interest in this study; the Sungai Galas in Kelantan. As the fourth most populated state in Malaysia, the state of Kelantan has the lowest in percentage (0.13%) of scheduled waste managed by the state (DOE, 2014). Most of the people living in the proximate riverbank area are fishermen, whom their boat activities were expected to effectuate pollution including heavy metals contamination in the surrounding area. Accumulation of heavy metals in fish is commonly associated with its presence in environment and it is thus crucial to determine the different concentrations of these metals in different fish species by studying their metabolic capability and bio-magnification of food chains (Wei et al. 2014).

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Humans might get exposed to pollutants from increasingly polluted river and without them realizing it may then cost them negative health effects years to come. Thus study on the accumulation of metals based on fish organ and physiochemical properties is crucial for a better understanding and fate of these metals in environment. This study was conducted to determine heavy metal concentrations of freshwater fish in Sungai Galas, Kelantan by using ICP-MS. The relationship of heavy metals (Cu, Mn, Ni, Pb, and Zn) with water and physiochemical parameters was examined; and identification of relationship between fish species and weight of fish with heavy metal was established. The metal pollution index (MPI) and Target Hazard Index (THQ) was also calculated as it is crucial in measuring level of exposure from the intake of metal by human consumption of edible fish.

1.2 Problem Statement

Water pollution is a serious concern that affects aquatic ecosystem. High level of heavy metal concentrations could bring about devastating effect to the ecological balance and altering the diversity of organisms in the water. These negative impacts of heavy metals toxicity on aquatic organisms could eventually affect the human lives. Different organs of aquatic organism especially fish, has tendency to accumulate heavy metals which in turn, could lead to health risks to humans by entering the metabolism system when consumed.

Sungai Galas is one of the developing regions in Kelantan and had been subjected to negative impacts of anthropogenic activities associated with industrial development and rapid urban expansion. As local population increase along with increase dependence on food source and source of income, Sungai Galas are heavily utilized by the locals for transportation, domestic routines, sand mining processes, agriculture and fishing activities which in turn gives effect to the balance of river ecosystem (Mokhtar et al. 2009). Over time, the discharge from these activities are very likely to have caused substantial effect in building up of concentration of metals in fishes and other aquatic organisms.

Due to the state's geographical location which is adjacent to the South China Sea, Kelantan is one of the states in Malaysia that are highly exposed to the northeast monsoon seasons occurring from November to March. The recurring flooding events had caused intense sedimentation and contributed to large amount of sediment depositions along the river as the flood water receded. Land clearing has been also identified as one of the factors that contributed to the unprecedented high degree of erosion and sedimentation and thus changing the topography along the river (Yahaya et al. 2015). Land clearing on the hill slope upstream and irresponsible mining was also observed near the Sungai Galas.

There had been studies done by researches such as Peck Yen & Rohasliney (2013); Rohasliney et al. (2014); and Ahmad et al. (2009) in describing the impact of sand mining activities with respect to several key indicators of water quality in Sungai Kelantan, and the effect of those activities toward freshwater fish. However to date, there has been no

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study done yet in term of those impacts to the freshwater environment along the tributaries of Sungai Galas. This study thus provides beneficial interpretation and information on bioavailability of particular elements in regard to pollution of Sungai Galas in Kelantan.

1.3 Objective of the study

The objective of this study was to assess level of heavy metal pollution in freshwater environment of Sungai Galas by emphasizing on surface water and fish organs as biomonitor organism. The specific objectives of the study were:

1) To determine the level of heavy metals (Cu, Zn, Pb, Mn and Ni) in surface water and in tissues of five different fish species (*Barbonymus schwanenfeldii, Mystacoleucus obtusirostris, Cyclochelichthys apogon, Hampala macrolepidota* and *Hemibagrus nemurus*) obtained from Sungai Galas.
2) To compare the heavy metal levels of Cu, Zn, Pb, Ni and Mn in liver, gills, and muscles of five different fish species.
3) To investigate whether the metals present in the fishes were within the permissible limits for human consumption based on Target Hazard Quotient (THQ).

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