UNIVERSITI PUTRA MALAYSIA

ASSESSMENT OF HEAVY METAL CONTAMINATION IN WATER AND SEDIMENTS OF BERTAM RIVER IN CAMERON HIGHLANDS AND ITS ASSOCIATION WITH ECOLOGICAL AND HEALTH RISKS

AZLINI RAZALI

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By

AZLINI RAZALI

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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July 2019

Chair : Sharifah Norkhadijah Syed Ismail, PhD
Faculty : Medicine and Health Sciences

Rampant land clearing for agriculture, urbanization, development of hotel and residential areas have rapidly altered the land use setting of Cameron Highlands. Unsustainable development and encroachment of river reserve areas have continuously deteriorated the Bertam River quality. The pollutants from Bertam River also may flow to downstream river network that serves as raw water sources for water treatment plants. Thus, it increases the potential health risk to the population downstream. This study aimed to assess heavy metal contamination in water and sediments of Bertam River in Cameron Highlands and its association with ecological and health risks. The differences in water quality status between Upper and Lower stations as well as seasonal changes were determined. The concentration of aluminium (Al), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), and zinc (Zn) in water and sediment samples were tested via Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The results showed that all studied physicochemical parameters were generally higher at the Lower stations. For example, in the wet season, the turbidity at the Lower stations (191.72 ± 139.78 NTU) was recorded higher than the Upper area (55.77 ± 147.55 NTU). Similarly, total suspended solids (TSS) was higher in the Lower area (158.07 ± 107.71 mg/L) compared to the Upper area (43.98 ± 78.97 mg/L). All physicochemical parameters were within the Malaysian National Drinking Water Quality Standards (NDWQS) except for turbidity, ammoniacal-nitrogen (NH_3-N) and *Escherichia coli* (*E. coli*). The heavy metal distribution in Upper and Lower stations was varied. Elements such as Iron (Fe), Al, Cu, and Zn were the most prominent and were higher during the wet season. The overall mean concentration of Cd, Fe and Pb exceeded the NDWQS. Seasonal changes do influence some of the river water properties such as dissolved oxygen (DO), turbidity and total suspended solids (TSS) where they were significantly higher during the wet season. The contamination factor (CF), geo-accumulation index (I_{geo}) and pollution load index (PLI) showed low ecological risk from a bioavailable fraction of heavy metal in the sediment of Bertam River. As for health risk, the carcinogenic risk of Cd in male adult (3.05×10^{-3}), female adult (2.97×10^{-3}) and children (4.91×10^{-3}) were $>10^{-4}$ which indicates an unacceptable carcinogenic risk. Three main potential pollution sources contributed to river water deterioration in this study were the artificial phosphate fertilizer and agrochemical products, soil erosion and land clearing. While the potential pollution sources in sediment were anthropogenic activities such as agriculture and sand dredging, soil erosion and the adsorption with soil mineral oxides. In conclusion, heavy metal in river and sediment samples were generally low and within acceptable risk to human and ecology except for Cd. Findings from this study have improved our understanding of how the agricultural activities and land use changes could interfere with the ecological niche particularly river networks in this area.

Keywords: Water quality; heavy metal; Cameron Highlands; health risk; ecological risk

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

PENTAKSIRAN PENCEMARAN LOGAM BERAT DI DALAM AIR DAN SEDIMEN SUNGAI BERTAM, CAMERON HIGHLANDS BERSERTA KAITANNYA TERHADAP RISIKO EKOLOGI DAN KESIHATAN

Oleh

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Penerokaan tanah secara berleluasa untuk pertanian, pembandaran, pembangunan hotel dan kawasan kediaman telah mengubah landskap guna tanah di Cameron Highlands. Pembangunan yang tidak mampan dan pencerobohan kawasan rizab sungai terus mengakibatkan kemerosotan kualiti Sungai Bertam. Bahan pencemar dari Sungai Bertam juga boleh mengalir ke rangkaian sungai di hilir yang berfungsi sebagai sumber air mentah untuk loji rawatan air. Oleh itu, ia meningkatkan potensi risiko kesihatan kepada penduduk di kawasan hilir. Matlamat kajian ini adalah untuk mentaksir pencemaran logam berat di dalam air dan sedimen Sungai Bertam, Cameron Highlands beserta kaitannya terhadap risiko ekologi dan kesihatan. Perbezaan status kualiti air antara stesen di bahagian atas dan bawah serta pengaruh perubahan musim telah ditentukan. Kepekatan aluminium (Al), kadmium (Cd), kromium (Cr), kuprum (Cu), ferum (Fe), plumbum (Pb) dan sampel sedimen diuji menerusi Induktif Pasangan Plasma-Spektrometri Jisim (ICP-MS). Hasil kajian menunjukkan bahawa secara umumnya, stesen di bahagian bawah mencatatkan nilai tertinggi untuk semua parameter fizikokimia yang dikaji berbanding stesen di bahagian atas. Sebagai contoh, di musim lembap, nilai kekeruhan di stesen bawah (191.72 ± 139.78 NTU) lebih tinggi daripada stesen atas (55.77 ± 147.55 NTU). Begitu juga dengan nilai pepejal terampai (TSS) di kawasan bawah (158.07 ± 107.71 mg/L) adalah lebih tinggi berbanding kawasan atas (43.98 ± 78.97 mg/L). Semua parameter adalah berada pada paras yang di benarkan berdasarkan Standard Kualiti Air Minum Kebangsaan Malaysia (NDWQS) kecuali bagi parameter kekeruhan, ammonia-nitrogen (NH3-N) dan *Escherichia coli* (*E. coli*). Kepekatan logam berat di stesen atas dan bawah adalah berbeza-beza. Ferum (Fe), Al, Cu, dan Zn didapati paling banyak di dalam air sungai dan paling tinggi ketika musim lembap. Purata kepekatan keseluruhan Cd, Fe dan Pb telah melebihi NDWQS. Perubahan musim didapati mempengaruhi beberapa parameter seperti oksigen terlarut (DO), kekeruhan dan TSS di mana ia lebih tinggi pada musim lembap. Faktor pencemaran (CF), indeks geo-terkumpul (I_{geo}) dan indeks beban pencemaran (PLI) menunjukkan risiko ekologi yang rendah daripada kandungan bio-tersedia logam berat dalam sedimen di Sungai Bertam. Risiko karsinogenik Cd pada lelaki dewasa (3.05 x 10^{-3}), wanita dewasa (2.97 x 10^{-3}) dan kanak-kanak (4.91 x 10^{-3}) adalah >10^{-4} yang menunjukkan risiko karsinogenik yang tidak boleh diterima. Tiga sumber pencemaran utama yang menyumbang kepada kemerosotan air sungai dalam kajian ini ialah baja fosfat tiruan dan produk agrokimia, hakisan tanah dan penerokaan tanah. Manakala potensi sumber pencemaran di dalam sedimen adalah aktiviti antropogen seperti pertanian dan pengorekan pasir, hakisan tanah, dan penyerapan dengan mineral oksida tanah. Logam berat di sungai dan sedimen umumnya rendah dan tidak mendatangkan risiko terhadap manusia dan ekologi kecuali Cd. Dapatan kajian ini telah meningkatkan pemahaman tentang bagaimana aktiviti pertanian dan perubahan guna tanah boleh mengganggu struktur ekologi terutama rangkaian sungai di kawasan ini.

Kata kunci: Kualiti air; logam berat; Cameron Highlands; risiko kesihatan; risiko ekologi

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I certify that a Thesis Examination Committee has met on 19 July 2019 to conduct the final examination of Azlini binti Razali on her thesis entitled "Assessment of Heavy Metal Contamination in Water and Sediments of Bertam River in Cameron Highlands and Its Association with Ecological and Health Risks" 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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LIST OF ABBREVIATIONS

Al	Aluminium
ANZECC	Australian and New Zealand Guidelines for Fresh and Marine Water Quality
APHA	American Public Health Association
BDL	Below detection limit
BOD	Biochemical oxygen demand
CCME	Canadian Council of Ministers of the Environment
Cd	Cadmium
CDI	Chronic daily intake
CF	Contamination factor
COD	Chemical oxygen demand
Cu	Copper
Cr	Chromium
DCCH	District Council Cameron Highlands
DLO	District and Land Office Cameron Highlands
DO	Dissolved oxygen
DOA	Department of Agriculture (Ministry of Agriculture and Agro-based Industry Malaysia, MOA)
DOE	Department of Environment (Ministry of Energy, Technology, Science, Environment & Climate Change Malaysia, MESTECC)
DOS	Department of Statistics
EC	Electric conductivity
EDTA	Ethylenediaminetetraacetic acid
Fe	Iron
GPS	Global Positioning System
HI	Hazard index
HQ	Hazard quotient
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
IEPA	Environmental Protection Agency, Ireland
I_{geo}	Geo-accumulation index
LCR	Lifetime cancer risk
LOI	Loss on ignition
LULC	Land use/land change
MANS	Malaysian Adult Nutrition Survey
Mn	Manganese
NA	Not applicable/ Not available
NAHRIM	National Hydraulic Research Institute of Malaysia
NDWQS	National Drinking Water Quality Standard (Ministry of Health Malaysia, MOH)
NH_3-N	Ammoniacal-nitrogen
NOAA	National Oceanic and Atmospheric Administration of United State
OEHHA	California Office of Environmental Health Hazard Assessment
PAIP	Pengurusan Air Pahang Berhad
Pb	Lead
PEL	Probable effect level

PLI	Pollution load index
RTD	Cameron Highlands Local Development Plan
SPSS	Statistical package for the social sciences
SQG	Sediment quality guideline
TDS	Total dissolved solids
TEL	Threshold effect level
TOM	Total organic matter
TSS	Total suspended solids
UNESCO	United Nations Educational, Scientific and Cultural Organization
USDA	United States Department of Agriculture
USDOE	United States Department of Energy
US EPA	United States Environmental Protection Agency
USGS	United States Geological Survey
WHO	World Health Organization
WQI	Water quality index
WTP	Water treatment plant
Zn	Zinc

CHAPTER 1

INTRODUCTION

1.1 Background

Highland regions act as water catchment and serve as the hydropower generation sources. It also provides water resources for agriculture, industries and domestic use (Roozitalab, Serghini, Keshavarz, Eser, & De-Pauw, 2013). Malaysia exploits fresh water for domestic use mostly from surface water (99%), while another 1% from groundwater. Surface water and groundwater are highly susceptible to heavy metal contamination caused mostly by anthropogenic activities (Ab Razak, Praveena, Aris, & Hashim, 2015). Heavy metal generally not removable even after the treatment at water treatment plant (WTP), thus increase the risk of trace metals contamination through ingestion of water (Maigari, Ekanem, Garba, Harami, & Akan, 2016).

Cameron Highlands' main economies are agriculture and tourism. The cold climate provide the best environment for temperate type of vegetation, flower and tea plantation (Weebers & Idris, 2016). However, rampant and unplanned development for agriculture, urbanization, hotels, and residential areas have create stress on the environment especially in the river system (Gasim et al., 2009; Khalik, Abdullah, Amerudin, & Padli, 2013a; Rasul, Islam, Yahaya, Alam, & Mokhtar, 2015). Unsustainable developing methods, farming on steep slopes, abuse usage of agrochemicals have caused influx of pollutants and high sedimentation rate in water bodies. These scenarios have triggered in deterioration of river water quality and increase the potential health risk to public (Aminuddin, Ghulam, Abdullah, Zulkefli, & Salama, 2005; Barrow, Weng, & Masron, 2009; Weebers & Idris, 2016).

1.2 Problem Statement

The policy of encouraging agriculture and development for mass tourism in Cameron Highlands has led to environmental problems. In 2015, the agriculture area in Cameron Highland has increased for 2.6% (RTD, 2018b). The problem with rampant agricultural activities in this area has introduced anthropogenic heavy metal in the freshwater ecosystems through repeated application of metal-containing agrochemicals (Cai et al., 2012). The use of inorganic phosphatic fertilizers also produced trace amount of heavy metal particularly As, Cd, Cu, Fe, Pb, and Zn (Atafar et al., 2010; Wuana & Okieimen, 2011). Heavy metal contamination in the freshwater ecosystem has attracted widespread attention due to its persistence, accumulation in the food chain and negative effects on the ecological and human health (Chen, Chen, Teng, & Wu, 2016; Ezemonye, Adebayo, Enuneku, Tongo, & Ogbomida, 2019). Heavy metal release in sediments and equilibrate in water column may alter the river water quality and have created a major concern on health (Ezekiel, Hart, & Abowei, 2011). Previous literatures around the globe also have reported a significant contribution of agricultural activities toward degradation of river water quality (Jo, Lee, Park, & Owen, 2010; Naveedullah et al., 2014; Perera, Sundarabarathy, Sivananthawerl, Kodithuwakku, & Edirisinghe, 2016; Yusoff, Jaafar, Toriman, & Kamarudin, 2015).

Farmers in Cameron Highlands also used the untreated chicken manure as fertilizers because of rich nitrogen (NPK) content that helps to speed up and elevate the growth of their crops (Barrow et al., 2009). The untreated and uncomposted chicken manure can lead to manure enrichment runoff and increase the risk of pathogens in streams (Barrow et al., 2009; Eisakhani & Malakahmad, 2009). Past studies also have reported a declining trend of river water quality in the river network of Cameron Highlands that was caused by intensive land clearing and agricultural activities (Al-Nafiey, Jaafar, & Bauk, 2014; Aminu, Matori, & Yusof, 2014; Eisakhani & Malakahmad, 2009; Gasim et al., 2009; Khalik, Abdullah, Padli, & Amerudin, 2013b; Rasul et al., 2015; Tan & Beh, 2016; Zulkipli, 2017). The water quality also greatly influenced by the high precipitation due to sudden influx of runoff and untreated sewage overflow into the water bodies (Eisakhani & Malakahmad, 2009; Gasim et al., 2009).

These facts have become the major concern in Bertam River, one of the main rivers in Cameron Highlands. Apart from serving as water sources for local population, Bertam River also merges with the main river network downstream that utilize the river water for drinking water sources (Pengurusan Air Pahang [PAIP], 2018). The pollutants from rivers in Cameron Highlands have the potential to contaminate the river water that supplies raw water in the water treatment plant (WTPs) for population downstream. Approximately, 5.8 million litres of water per day is utilized by water treatment plants (WTPs) along rivers that originating from Cameron Highlands (Antony & Chantal, 2006).

Bertam River is also a home to Cameron Highlands' hydroelectric power scheme. The hydrological flow of Bertam River also highly influenced by the dam construction and channels modification especially at the Lower Bertam. Thus, it causes pressure on the Lower area due to sediment that absorbed pollutants from flowing water and deposited into the reservoir (Gregory, 2006; Wong et al., 2017).

Therefore, this study was designed to assess the heavy metal contamination in the river water and sediments impacted from agricultural activities in the study area. This study also aiming to determine the association between heavy metal contamination with the ecological and health risks. It extends the existing knowledge about Bertam River's water quality by integrating multi-parameters (i.e. physical, chemical and biological) and multi-compartment system (i.e. water and sediment) to establish a comprehensive monitoring design. By incorporating multi-parameter and multi-compartment system into the study, it will provide a better insight on the pollution issues in Bertam River.

1.3 Study Significant

The continuous trend of river water deterioration in Cameron Highlands has become the main concern and has raised the significant needs for this study to be conducted. Since the pollutants such as metals have tendency to bio-accumulate in water (pose health risk) and in sediment (pose ecological risk), it is important to conduct a study to address this potential risk. Moreover, to our knowledge, there is limited existing data to address the issue relating to health risk poses by metal contamination in river network of Cameron Highlands (Zulkipli, 2017). In addition, human health risk assessment is of great importance for holistic water quality monitoring (Naveedullah et al., 2014).

Furthermore, water quality assessment provides the baseline information on water safety. Continuous monitoring is vital for early detection of pollution influx in water bodies. This study also in line with the Malaysia Government efforts under the 11th Malaysia Plan which allocate RM50 million for river basin management plans. The plans aim to improve water quality, reduce the risk of floods, protect the environment, and ensure there is enough water in any particular basin.

Cameron Highlands serve as one of the main temperate highland location for local and foreign tourists in Malaysia. It is important to preserve the natural service which serve as the main attraction to generate the economy from tourism activities. In the year of 2014 recorded the extreme declining in tourist number visiting Cameron Highlands which was -18.9%. The main reasons were because of natural disasters such as flood, mud flood and landslides within the particular year which possibly triggered by unsustainable and intensive land development (RTD 2018b). Many strategies have been outlined in Cameron Highlands Local Development Plan (RTD 2030) focusing on to produce a sustainable highland tourism activity through sustainable agricultural management.

The outcome from this study provides the current status of water quality in Bertam River and will be helpful to monitor the effectiveness of every efforts and strategies undertaken by the local authorities in addressing the environmental issues particularly toward river network in Cameron Highlands. The results also can be utilize to build an advanced modelling to give a prediction of high, moderate and low-risk areas along the river. The information and data gain will enable local authorities to implement the best solution to tackle the pollution issues specifically in the highland area. Finding from this study can be helpful to produce an action plan to restore the Bertam River by emphasizing on rehabilitation, preservation and enhancement of the surroundings through good agricultural practices and proper development plans. This study also will serve as the preliminary data to provide baseline information to elaborate the potential health risk through water consumption as well as for better understanding regarding water safety.

1.4 Conceptual Framework

Figure 1.1 presents the conceptual framework of this study. Agricultural activities, land clearing, municipal and domestic waste water were identified as point and non-point sources of runoff into the river system of Cameron Highlands. The runoff also carried various type of pollutants such as organochlorine pesticides, heavy metal and macronutrients to name a few. These contaminants tend to bio-accumulate and bio-magnified in the water, sediment and biota due to its persistency nature (Wong et al., 2017). All these contaminants pose a threat toward river water, sediment and living biota. In this study, water and sediment samples were taken as indicators for contamination in Bertam River. For water sample, heavy metal concentration, physicochemical parameters and *E. coli* level were determined. For sediment sample, heavy metal concentration and physicochemical parameters were determined. On the health risk assessment, human exposure could occur via three routes; ingestion, inhalation and dermal. As the concern arise for downstream population that utilized water from downstream rivers that have high risk to be contaminated from rivers in the highlands area, the health risk assessment in this study was focused on ingestion route. Contamination factor (CF), index of geo-accumulation (I_{geo}) and pollution load index (PLI) were calculated to assess the potential ecological risk associated with metal contamination in surface sediment.

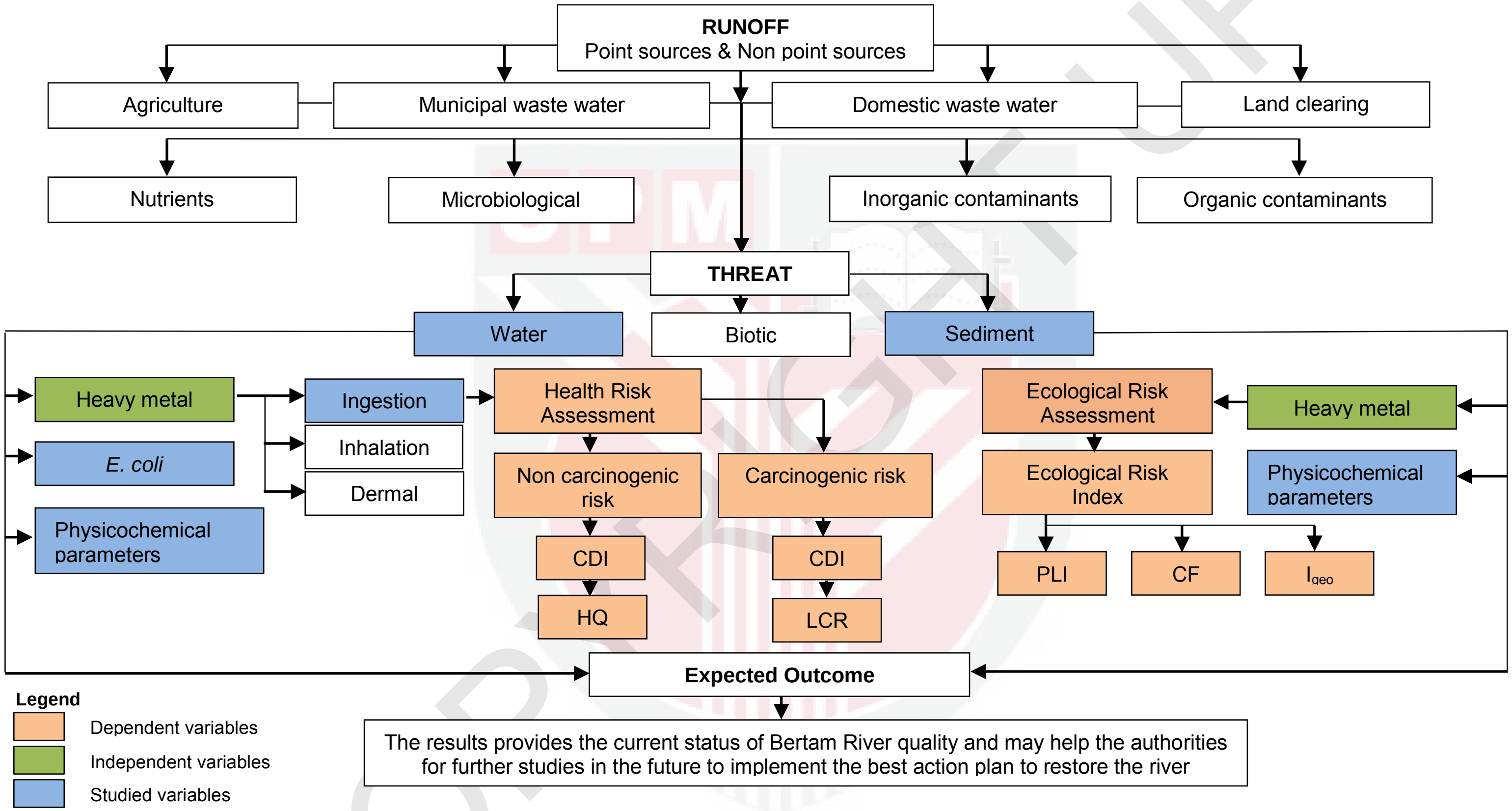

Figure 1.1 Conceptual Framework.

1.5 Research Questions

1) What is the physicochemical, biological properties and heavy metal concentration (Al, Cd, Cr, Cu, Fe, Pb, and Zn) in surface water and sediment of Bertam River and to the standard values?
2) How the physicochemical, biological properties and heavy metal concentration were influenced by the seasonal changes and the correlation between them?
3) How the heavy metal contamination associated with the ecological and health risk?
4) What are the potential sources of these pollution at the study area?

1.6 Objectives

1.6.1 Main Objective

The aim of this study was to assess heavy metal contamination in water and sediments of Bertam River and its association with ecological and health risk in intensive agriculture area, Cameron Highlands.

1.6.2 Specific Objectives

1) To determine and compare the physicochemical properties and heavy metal concentration (Al, Cd, Cr, Cu, Fe, Pb, and Zn) between the Upper and Lower Bertam River and to the National Drinking Water Quality Standard (NDWQS).
2) To compare the physicochemical properties and heavy metal concentration of Bertam River between wet and dry seasons and to the monitoring data by Department of Environment.
3) To determine and compare the physicochemical properties and heavy metal concentration (Al, Cd, Cr, Cu, Fe, Pb, and Zn) in the sediment between Upper and Lower Bertam River and to the background values.
4) To determine the correlation between physicochemical properties with heavy metal concentration of surface water and sediment.
5) To determine the ecological pollution risk from heavy metal contamination in the river sediment (i.e. contamination factor, geo-accumulation index) and the human health risk from heavy metal contamination in the surface water.
6) To identify the potential sources of pollution along Bertam River that contribute to the water and sediment quality.

1.7 Research Hypotheses

1) There is a significant differences of physicochemical properties and heavy metal concentration between the Upper and Lower Bertam River.
2) There is a significant differences of physicochemical properties and heavy metal concentration of Bertam River between wet and dry seasons.
3) There is a significant differences of physicochemical properties and heavy metal concentration in the sediment between the Upper and Lower Bertam River.
4) There is a significant correlation between physicochemical properties with heavy metal concentration of surface water and sediment.
5) There is a significant ecological and human health risk from heavy metal contamination in the river sediment and water.

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LIST OF PUBLICATIONS

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