DEVELOPMENT OF WATER QUALITY INDEX OF EX-MINING PONDS IN MALAYSIA

ISA BABA KOKI

THESIS SUBMITTED IN FULFILMENT OF THE REQUIREMENTS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

DEPARTMENT OF CHEMISTRY
FACULTY OF SCIENCE
UNIVERSITY OF MALAYA
KUALA LUMPUR

2017

UNIVERSITY OF MALAYA

ORIGINAL LITERARY WORK DECLARATION

Name of Candidate: ISA BABA KOKI

Matric No: SHC130115

Name of Degree: DOCTOR OF PHILOSOPHY

Title of Project Paper/Research Report/Dissertation/Thesis ("this Work"):

DEVELOPMENT OF WATER QUALITY INDEX OF EX-MINING PONDS IN MALAYSIA

Field of Study: ANALYTICAL CHEMISTRY AND CHEMOMETRICS

I do solemnly and sincerely declare that:

(1) I am the sole author/writer of this Work;
(2) This Work is original;
(3) Any use of any work in which copyright exists was done by way of fair dealing and for permitted purposes and any excerpt or extract from, or reference to or reproduction of any copyright work has been disclosed expressly and sufficiently and the title of the Work and its authorship have been acknowledged in this Work;
(4) I do not have any actual knowledge nor do I ought reasonably to know that the making of this work constitutes an infringement of any copyright work;
(5) I hereby assign all and every rights in the copyright to this Work to the University of Malaya ("UM"), who henceforth shall be owner of the copyright in this Work and that any reproduction or use in any form or by any means whatsoever is prohibited without the written consent of UM having been first had and obtained;
(6) I am fully aware that if in the course of making this Work I have infringed any copyright whether intentionally or otherwise, I may be subject to legal action or any other action as may be determined by UM.

Candidate's Signature Date: 23rd October, 2017

Subscribed and solemnly declared before,

Witness's Signature Date: 23rd October, 2017

Name: Prof Dr. Sharifuddin Md Zain

Designation: Professor, Department of Chemistry, University of Malaya

DEVELOPMENT OF WATER QUALITY INDEX OF EX-MINING PONDS IN MALAYSIA

ABSTRACT

Assessment of the water quality of ex-mining ponds for its proper utilization is pertinent considering the metal pollutants, and lack of proper environmental measures associated with the mining operations. Water samples from selected lakes in the Central Region of Peninsular Malaysia (Selangor and Negeri Sembilan) and Malacca from the Southern part were analysed in-situ for pH, DO, SS, EC, BOD, AN, and TDS; while metal elements including As, Cd, Pb, Mn, Fe, Na, Mg, and Ca were analysed off-site using inductively coupled plasma mass spectrometry and verified via certified reference materials. The variations in these water quality parameters were explored with chemometric techniques. The results from the principal component analysis, factor analysis and hierarchical cluster analysis suggested that in addition to the degree of contamination, the variations in heavy metal concentrations were mainly attributed to the ex-mining activities, whereas deviations in BOD, TDS, AN, and EC were subjected to the current domestic inputs. The linear discriminant analysis showed that water samples from the ex-mining ponds were highly associated with the concentrations of Mn, Cd, Ca and As; the model constructed with the training set gave a precise prediction with R^2 = 1.000, with no misclassification shown with the validation set. In Klang Valley, ex-mining ponds generally show low pH compared to the other lakes; the As and Cd levels recorded in most of the ex-mining ponds surpassed the limits of the Malaysian Water Quality Standard which make them unfit for domestic uses. A study of man-made lakes in Melaka and Negeri Sembilan revealed basic pH and low metal concentrations (below the reference values) in which As, Mn, and Mg were associated with ex-mining ponds while pH and Na were associated with other lakes. In this case, the high positive loadings of pH, DO, Ca, and TDS on principal component 1 suggests the absence of significant anthropogenic input. Overall, the differences in geology and domestic inputs into the ex-mining ponds significantly affect the water quality and results in variation in the levels of metals and physical-chemical parameters. The associated health risk via ingestion and dermal contacts were also modelled using Monte Carlo simulation and the estimated carcinogenic and non-carcinogenic risks for both adult and children were compared with their benchmarks. Water quality index was then formulated based on the Malaysian Water Quality Standard and results from chemometric analyses were applied in the parameter selection so as to minimize subjectivity; consequently, As, Cd, Pb, pH, DO, BOD and AN were chosen. The index was applied for the evaluation of water quality of the above mentioned lakes where most of the ex-mining ponds in Klang Valley were classified as very polluted, while those in Melaka and Negeri Sembilan were rated as excellent.

Keywords: Water quality, Ex-mining ponds, Lake, Chemometrics, Metals.

INDEKS KUALITI AIR BEKAS KOLAM PENAMBANGAN DI MALAYSIA

ABSTRAK

Penilaian kualiti air kolam bekas lombong untuk kegunaan yang bersesuaian adalah penting memandangkan wujudnya pencemaran logam dan kurangnya langkah-langkah mesra alam berkaitan dengan operasi perlombongan. Sampel air dari tasik terpilih di Wilayah Tengah Semenanjung Malaysia (Selangor dan Negeri Sembilan) serta Melaka dari bahagian selatan dianalisis secara *in-situ* untuk pH, DO, SS, EC, BOD, AN, dan TDS. Analisis unsur logam termasuk As, Cd, Pb, Mn, Fe, Na, Mg, dan Ca dilakukan di makmal dengan menggunakan alat plasma gandingan induktif-spektrometri jisim dan disahkan dengan bahan rujukan yang diperakui. Variasi dalam parameter kualiti air tersebut diteroka menggunakan pelbagai teknik kimometrik. Keputusan daripada analisis komponen utama dan analisis kelompok hierarki menunjukkan bahawa selain daripada tahap pencemaran, variasi dalam kepekatan logam berat juga berlaku disebabkan oleh aktiviti bekas perlombongan, manakala sisihan dalam BOD, TDS, AN, dan EC pula tertakluk kepada input domestik semasa. Analisis diskriminan linear pula menunjukkan bahawa sampel air dari kolam bekas lombong mempunyai korelasi dengan kepekatan Mn, Cd, Ca, dan As; model yang dibina dengan set latihan memberi ramalan tepat dengan R^2 = 1.000, tanpa sebarang kesalahan klasifikasi berdasarkan set pengesahan. Di Lembah Klang, secara umumnya kolam bekas lombong menunjukkan pH rendah berbanding dengan tasik lain; tahap As dan Cd yang terekod di kebanyakan kolam bekas lombong tersebut melepasi had Piawai Kualiti Air Malaysia, yang menjadikannya tidak sesuai untuk kegunaan domestik. Kajian tasik buatan manusia di Melaka dan Negeri Sembilan pula mendedahkan pH bes dan kepekatan logam yang rendah (bawah nilai rujukan) di mana kepekatan As, Mn, dan Mg dapat dikaitkan dengan kolam bekas lombong manakala pH dan Na dikaitkan dengan tasik yang lain.

Dalam kes ini, muatan positif tinggi pH, DO, Ca, dan TDS ke atas komponen utama 1 menggambarkan ketiadaan input antropogenik yang ketara. Secara keseluruhannya, perbezaan dalam geologi dan input domestik ke atas kolam bekas lombong memberi kesan yang ketara terdahap kualiti air dan menyebabkan perubahan dalam tahap logam dan parameter fiziko-kimia. Risiko kesihatan susulan dari pengambilan dan sentuhan kulit dimodelkan dengan simulasi Monte Carlo dan anggaran risiko karsinogenik dan bukan karsinogenik untuk kumpulan dewasa dan kanak-kanak dibanding dengan penanda aras masing-masing. Indeks kualiti air kemudiannya dirumuskan berdasarkan piawaian Kualiti Air Malaysia, dan berdasarkan penemuan di atas, pendekatan kimometrik telah digunakan dalam pemilihan parameter supaya dapat mengurangkan subjektiviti di mana As, Cd, Pb, pH, DO, BOD dan AN telah dipilih. Indeks ini telah digunakan untuk penilaian tasik yang dinyatakan di atas di mana sebahagian besar kualiti air kolam bekas lombong di Lembah Klang telah dikelaskan sebagai sangat tercemar, manakala kualiti air di Melaka dan Negeri Sembilan adalah sangat baik.

Kata kunci: Kualiti air, Kolam penambangan, Tasik, Kimometrik, Logam.

ACKNOWLEDGEMENTS

In the first place, I would like to express sincere appreciation to my supervisors, Prof. Dr. Sharifuddin Md Zain and Dr. Low Kah Hin for their guidance, good suggestions, patience, and full support during the course of this research project. Im very thankful to Assoc. Prof. Dr. Hafizan Juahir and his research team from East coast environmental research institute (ESERI), University Sultan Zainal Abidin, Kuala Terengganu for the sampling tour in the selected sates. Their technical assistance and cooperation in making this project successful is highly appreciated. Im grateful to Mr Shaharudin Zainal of Chemistry Department University Malaya, and Mrs Munira Binti Abdul Zali of Department of Chemistry Malaysia (Jabatan Kimia) for their help in ICP-MS analysis. I would also like to thank all staffs of Faculty of science and Chemistry department for their help. I wish to acknowledge all those who helped me academically, technically, and personally but could not be mentioned here.

I would like to thank Prof. Dr. Sharifuddin Md Zain for his financial support, moral advices, and concern throughout my stay in Malaysia. Furthermore, the financial support of Northwest University Kano, and my brothers in person of Ado Baba Koki, and Muhammad Baba Koki (Mai'ashafa) are well appreciated and acknowledged. The advices of my brother Mahmoud Baba Koki (Alhaji) are also valued and acknowledged.

Last but not least, I deeply appreciate the prayers, concern and encouragements of my late father Alhaji Baba Koki, and mother Hajiya Aisha Baba Koki. May his soul rest in Jannatil Firdaus. I want to warmly thank my wife Aisha Abdullahi Idris for the patience, kind consideration and taking care of my lovely Son Dayyab (Walid) during my stay in Malaysia, I will forever be grateful for that. The advice, concern and prayers of Hassana Abdullahi are fully acknowledged.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF SYMBOLS AND ABBREVIATIONS

AMD	:	Acid Mine Drainage
AN	:	Ammoniacal Nitrogen
ANOVA	:	Analysis of Variance
AT	:	Average Time
ATSDR	:	Agency for Toxic Substances and Disease Registry
BLL	:	Blood Lead Level
BOD	:	Biological Oxygen Demand
BW	:	Body Weight
CCME	:	Canadian Council of Ministers of Environment
CF	:	Conversion Factor
CR	:	Carcinogenic Risk
Cx	:	Concentration of the x_{th} metal
NCR	:	Non-carcinogenic Risk
DL	:	Detection Limit
DO	:	Dissolved Oxygen
DOE	:	Department of Environment
DWQSRW	:	Drinking Water Quality Standard for Raw Water
ED	:	Exposure Duration
EF	:	Exposure Frequency
ET	:	Exposure Time
EXP	:	Exposure
HCA	:	Hierarchical Cluster Analysis
HI	:	Hazard Index
HORAS	:	Hybrid off River Augmentation System

HQ	:	Hazard Quotient
IAR	:	International Agency for Research on Cancer
ICP-MS	:	Inductively Coupled Plasma Mass Spectrophotometer
INWQS	:	Interim National Water Quality Standard
IQ	:	Intelligent Quotient
IR	:	Ingestion Rate
K	:	Proportionality Constant
Kp	:	Dermal Permeability Coefficient
LDA	:	Linear Discriminant Analysis
LRM	:	Linear Regression Model
LUAS	:	Lembaga Urus Air Selengor
MANOVA	:	Multivariate Analysis of Variance
MERC	:	Malaysia Environmental Research Council
MOH	:	Ministry of Health
MSM	:	Multivariate statistical model
MST	:	Monte Carlo Simulation Technique
N.D.	:	Not Detected
NSFWQI	:	National Sanitation Foundation Water Quality Index
PCA	:	Principal Component Analysis
Q_i	:	Quality Rating
QC	:	Quality Control
R^2	:	Coefficient of Determination
RfD	:	Reference Dose
RME	:	Reasonable Maximum Exposure
SA	:	Surface Area
SF	:	Slope Factor

SI	:	Sub index
TSS	:	Total Suspended Solid
UPW	:	Ultra-Pure Water
USEPA	:	United State Environmental Protection Agency
W_i	:	Weightage
WHO	:	World Health Organization
WQI	:	Water Quality Index

LIST OF APPENDICES

CHAPTER 1: INTRODUCTION

Water quality illustrates the state or condition of water incorporating physical, chemical and biological characteristics, with respect to its acceptability for a specific purpose (WHO, 2011). Good quality drinking water has become a scarce commodity due to rapid growth in industrialization and continuous increase in population. The environmental status of lakes depends upon the type of the lake and its exposure to various factors in the environment. Hence, the quality of surface water depends not only on natural environmental processes such as weathering, erosion and precipitation, but also on the influence of anthropogenic activities including urban, agricultural and industrial activities (Papatheodorou et al., 2006).

Industrial activities like mineral and ore processing generate pollutants into the environment. Accumulation of the pollutants in water bodies is a threat to humans, plants and animals (Nazir et al., 2015). The impairment of water quality and pollution of adjacent rivers and lakes with heavy metals as the environmental effect of mining has been reported in many countries among which are Malaysia, Serbia, Spain, and Kenya (Ashraf et al., 2011b; Atanacković et al., 2013; Navarro et al., 2008; Ngure et al., 2014). The absence of environmental protection laws in Malaysia's mineral exploration industry leads to the destruction of natural habitat. Introduction of the mining enactment act in the year 1934 enforces some environmental standards; such as the law that required the tin mining companies to conduct rehabilitation of the mined excavated land (Balamurugan, 1991). But still there are hundreds of abandoned ex-mining ponds in Malaysia with large volume of water which quality and safety is of much concern. The extent of water contamination with respect to water quality determinants can be established using multivariate statistical analysis and parameter responsible for the deterioration of water quality can be identified, and the level of the pollutants compared to Malaysia's interim national water quality standards (INWQS).

1.1 Mining in Malaysia

Mining is the selective removal or extraction of precious minerals from the earth's crust usually from their ores (Norgate & Haque, 2010). Tin mining was mostly practiced in Malaysia by the 19th century, particularly in Selangor which accounted for a large portion of the total country's output. The common ore mined was cassiterite (SnO_2) (Balamurugan, 1991). The tin mining had dominated Malaysia's mineral industry with an output of 61,404 tons in 1980 estimated at hundreds of million dollars, thus the major foreign exchange earner for the country (Ashraf et al., 2015). The booming mining operation contributed positively to the economic development of Malaysia as one of the world's largest tin producer. However, mining activity results in environmental degradation and consequently left abandoned mines characterized by large holes or man-made lakes (Yaacob et al., 2009). Western peninsular Malaysia was known to experience the extensive mining operation (Takaijudin et al., 2012a). The existence of hundreds of abandoned ex-mining ponds, mostly in Selangor is one of the consequences of extensive and uncontrolled mining operation during the 19th century. There are about 442 ex-mining ponds in Bestari Jaya, many are spread in Puchong, and Kelana Jaya in Selangor with 13,667 hectares of mining land (Balamurugan, 1991; DMG, 2003; Yap et al., 2005). Consequently Selangor and Melaka states of the western part of peninsular Malaysia were selected as the case study in this research.

1.1.1 Water Pollution in Ex-mining Ponds

Malaysia has the world's largest reserve of tin and other heavy metal ores which are consequently released to the environment, specifically the water bodies (Murcott, 2012).

The increasing public outcry that mining has left derelict sites, huge post-mining lands appear to be a serious obstacle to sustainable development in many countries of the world which had mining industries. After the commencement of mining operation in Malaysia, the authorities could not focus on the environmental hazards especially heavy metal pollution. The enactment of the mining code of Negeri Sembilan (1895) imposed that slime and sand obtained from hydraulic mining be retained or dumped in old mining holes, and only fine slimes could be discharged into rivers without giving due consideration to the metal pollutants introduced. The Selangor mining act no. 19 (1901) directed that water used for mining must be free from sand, gravel, sludge, dirt, and tailing before being discharged to the environment, yet no attention was given to the heavy metal contents.

Water pollution due to mining operation later received much attention, mainly related to the widespread contamination due to improper mining management (Kemp et al., 2010). The extensive pollution witnessed in the areas with mining and ore extraction industry often appears not to have a contemporary origin, but a consequence of previous activities (Paulo, 2005). Abandoned mining sites cause substantial impairment to water quality worldwide (Fields, 2003). After mining operations had ceased, oxygen-rich groundwater floods abandoned mining areas and can promote oxidation of pyrite (FeS_2) and other metal sulfide minerals, producing acidity and releasing co-occurring trace metals (Younger et al., 2002a). The acid mine drainage (AMD) containing harmful pollutants (heavy metals) formed during and after the mining operation is a major threat to the environment (Akcil & Koldas, 2006). The heavy metals associated with ex-mining water are As, Pb, Cd, Mn, Cu and Zn; other physical-chemical parameters are pH (high or low depending on the geology of the mines), high electrical conductivity (EC), total dissolve solid (TDS), and low dissolved oxygen (DO) (Ashraf et al., 2010; Iavazzo et al., 2012). Arsenic (As) was found to coexist with tin ore in the mining areas of Malaysia (Ismail, 1973). As concentration was reported in high concentration near the mining sites, abandoned mining ponds therefore pose a threat and contribute to high arsenic concentration in drinking water (Murcott, 2012). High As concentrations was detected in a well water near a tin mining area in Malaysia where 3 cases of cutaneous lesions related to arsenical poisoning were discovered (Jidon, 1993).

Most of the ex-mining ponds in Malaysia are used for recreational activities, flood retention or to receive domestic effluent from the surrounding residences (Ghani et al., 2008; Takaijudin et al., 2012a; Yap et al., 2005). The activities around the catchments will also influence variations in water quality of the ponds (Boyd, 2015). The concentration of the metals in ex-mining ponds can increase (due to evaporation and concentration, and flow of industrial wastes or effluents) or decrease (due to dewatering, flow of rain water “flood retention” and domestic effluents). It is therefore of most importance to carefully study the activities around the ex-mining ponds.

Malaysia has adequate rainfall and abundant water resources, but yet facing water quality problems and shortages (Aini et al., 2007). The increasing demand for a sufficient water supply, especially in drought season lead to a search for alternative water resources, and a suggestion was made by the Selangor authority to utilize water from ex-mining ponds for human consumption and domestic purposes through conventional water treatment processes (LUAS, 2008, 2012). This resulted in arguments and media hype on the safety of ex-mining water considering the metal pollutants associated with mining operations (Low et al., 2016), especially considering its potential to retain the metals at elevated levels for centuries or even millennia after the closure of the mine (Younger & Wolkersdorfer, 2004). The ex-mining water was used in the year 2014 in Kuala Lumpur as an additional water source without giving due consideration to the health risks associated with the exposures (Daniel & Kawasaki, 2016), as at high exposure level even low metal concentrations could pose a health threat to humans. And exposure levels of some heavy metals, especially Cd were found to be below what was previously expected (Chen et al., 2014; Järup, 2003).

A new project, "Hybrid off-river augmentation system (HORAS)" has commenced on a small scale in Selangor which drain water from ex-mining ponds into the river, and to the water treatment plant to support the additional raw water supply and to meet up with the increasing demand of 2670 million liters per day (Kusin et al., 2016). Similarly, the government authorities in Melaka identified an ex-mining pond "Tasik Biru Chin Chin" to be utilized and meet up with the daily water supply of 500 million liters by the year 2017 (Azzlan et al., 2016). This became necessary due to the very low water level at the two major sources of water supply in Melaka, the Jus reservoir and Tasik Durian Tunggal. Therefore the need arises to carefully and intensively analyze and confirm the levels of metals in the ex-mining ponds and evaluate the health risks before its utilization for beneficial purposes.

1.2 Water Quality Index (WQI)

The index approach in the assessment of water quality is regarded as a simple and concise means to express water quality, taking into consideration numerous key water quality parameters that are aggregated to represent the overall quality of a water body (Tyagi et al., 2013). WQI is a concept which allows for reporting of water quality by a single numerical index, with respect to the use for which the water is intended. It reduces a sizable amount of data of numerous water quality parameters to only one number in an easy, objective and consistent manner. The index approach permits for significant spatial and temporal trends to be established and incorporate the effects of the different pollutants present. Hence water quality index not only presents a point of reference for water quality, but can also show the effect of the possible changes on water use of the aquatic ecosystem.

WQI creates a standard, and water quality standard plays an extremely important role in water resource management framework. There may be differences in the goal, approaches, legal framework, and methodology, nevertheless, the ultimate aim of the entire water quality standard is to ensure the water resources are being protected in a sustainable manner (MERC, 2012). WQI is a means to convey information on the status of water quality to the consumers and policy makers for the evaluation and management of surface water (Udom et al., 2016). The Malaysia Department of Environment (DOE) applies WQI to assess the water quality status of rivers, but the index can not accommodate lakes and ex-mining ponds. Considering that lakes are few, and most lakes in Malaysia are ex-mining ponds that are mostly polluted with high concentration of toxic heavy metals especially As (Ashraf et al., 2011b; Ashraf et al., 2010; Ashraf et al., 2015; Chang et al., 2008; Orji et al., 2014; Sharip & Zakaria, 2007), there is a need to develop an index that accommodates the heavy metals in order to adequately monitor ex-mining ponds and other existing lakes for effective management and possible utilization.

1.3 Risk Assessment

Risk assessment is a systematic quantitative identification and evaluation of variables that have potential risks associated with a projected activity, involving an understanding of the sources, pathways and the target (Yilmaz, 2011). The information on risk assessment is essential for decision makers to create policies and protect the health of the population (Sobus et al., 2011). The concept of risk assessment is mostly applied in studies related to environmental pollution. The risks in pollution of the environment beside natural sources could be from anthropogenic origin, such as mining (Muhammad et al., 2011). Metal contamination due to mining can affect public health through water, and soil, thus there is a need for strict environmental regulation and risk analysis (Béjaoui et al., 2016; Ezekwe et al., 2012). Ex-mining water pollute underground water and gets into nearby rivers and streams thereby posing a health threat (Ban et al., 2015; Li, 2016).

Heavy metal contaminants, especially As, Pb and Cd are the main focus in risk assessment studies due to their high toxicity to humans and animal; these metals are highly lethal and can cause dysfunction in humans. The risk of As toxicity is of high concern due to its high bioavailability in aqueous phase comparable to solid medium. As is related to diseases such as skin cancer, bladder, and kidney impairment (Palmer et al., 2016). Dermal contact and ingestion of Pb in contaminated water and soil causes respiratory and Dermato-generic complications (Nevárez et al., 2015; Yang et al., 2016a). Exposure to Cd causes cancer, hypertension and kidney dysfunction (Woo et al., 2015). Considering the health complications associated with these metals, it is pertinent to fully analyze the risks associated with exposures in water from ex-mining ponds.

1.3.1 Human Health Risk Assessment Techniques

These are techniques aimed at assessing and estimating the chances or probability and the nature of the adverse health effect in humans that may be exposed to toxic chemical in a polluted environment, now or in the future. It is applied to identify and characterize the type and magnitude of health risks to humans. Therefore, risk assessment techniques are decision making requirements that could provide a fast and vital decision making support on environmental safety (Rebelo et al., 2014). Generally, all human health risk assessment techniques of toxic substances involve hazard identification, exposure assessment, risk estimation, and risk characterization (Figure 1.1).

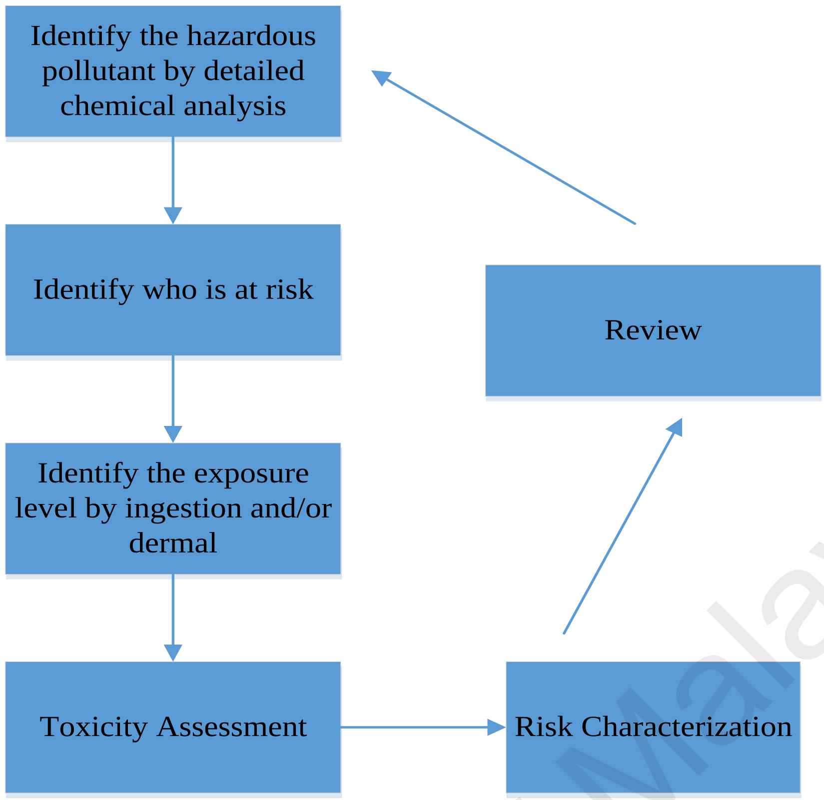

Figure 1.1: The human health risk analysis process

1.3.1.1 Hazard Identification

Hazard identification is one of the important aspects of health risk assessment which involves the determination of possible conditions of human exposure to hazardous substances. It is the first step in a risk assessment procedure which comprises of a qualitative evaluation of the hazards that might exist in the medium under study. Sufficient evidence must be established to classify a substance as the cause of adverse health effects. In this phase of risk assessment, data collection and analysis of a particular chemical substance of concern is carried out based on past researches. The general overview of the toxic metals of concern is highlighted in chapter two of this study.

1.3.1.2 Exposure Assessment

The essence of exposure assessment is to measure and quantify the amount, frequency and duration of exposure to a chemical substance among the population exposed. The quantitative exposure assessment presents an estimate of the rates at which the toxic metals are ingested or absorbed using mathematical equations. The exposure assessment evaluates the source of the toxic metals as well as the pathways and routes of individual exposure (USEPA, 1992).

In light of this, a detailed and a standard chemical analysis of the toxic metals of interest are of utmost priority, so as to ensure data quality and reliability. In this study, a standard experimental procedure is applied with quality control measures including certified reference materials for proper validation.

1.3.1.3 Toxicity Assessment

This is one of the major aspects of risk assessment that is concerned with establishing the relationship between the selected contaminants (toxic metals) and the receptor (humans). It involves the numerical evaluation of toxicity information and detailed characterization of the exposure-response relationship. The data involved in the dose-response analysis is analyzed to identify the toxicity and the risk involved (USEPA, 1987). Therefore, effort must be made to establish a link between the toxic metals ingested or received and human health effect. Health risk assessment is achieved by deriving vital information from toxicological references and profiles such as the Agency for Toxic Substances and Disease Registry (ATSDR) and the United States Environmental Protection Agency (USEPA).

1.3.1.4 Risk Characterization

The main goal of risk characterization is to present an idea on the type and magnitude of the adverse health effect that a particular toxic metal can have in an environment. Risk characterization is the final step in the health risk assessment process which explains the description of the public health risks related to toxic substances. The information on the exposure assessment is compared to the toxicity assessment to conclude on the likelihood that the toxic metals could cause harm to individuals and the population. Generally, site-specific data and assumptions are applied in the formula to compute cancer risks and non-cancer hazards of the toxic substances (USEPA, 1989). The hazard index (HI), hazard quotient (HQ), and carcinogenic risk are employed in this study to explain the potential cancer risk and non-carcinogenic hazards associated with exposure to the toxic metals in ex-mining ponds based on USEPA formulation. The carcinogenic risk is estimated using exposures and slope factor for the specific metals of interest.

1.4 Chemometrics

Analytical chemists apply statistics to solve chemical problems by analyzing the chemical data and gain ideas about the chemical system. It involves the use of multivariate statistical methods to explain the observations and characteristics of complex data. In achieving a meaningful result, the use of specific statistical techniques is highly essential for an objective exploratory assessment. Chemometrics is therefore the best approach for the classification and monitoring of environmental data.

The environmental data is usually characterized by high variability due to various natural and anthropogenic influences, and chemometric methods are thus continuously utilized in identifying and managing the possible sources of pollution (Wu & Kuo, 2012). Treatment of environmental data using multivariate approach characterizes and evaluates the surface water quality. The interpretation of the environmental data has the advantage of visualizing and treating large quantity of raw analytical measurement, and extracting useful information. Principal component analysis (PCA), Factor analysis (FA), Hierarchical cluster analysis (HCA) and Linear discriminant analysis (LDA) are the techniques mostly employed in the literature related to water quality interpretation using multivariate data analysis, classification and modelling (Bouguerne et al., 2016; Iscen et al., 2008; Li et al., 2015; Noori et al., 2010; Shrestha & Kazama, 2007; Singh et al., 2007; Varol et al., 2012; Wallace et al., 2016).

1.5 Research Aims and Objectives

The general aim of this study is to assess and investigate the suitability of water from ex-mining ponds for consumption and recreational activities, by analyzing the presence of toxic heavy metals associated with mining operations. The research focuses on Selangor state that had extensive mining activity from central Malaysia, and Melaka state from southern region of Malaysia was included in this study due to its recent water crises. The central objectives of this research are;

A) To determine the levels and variations of heavy metal pollutants and general water quality parameters in ex-mining ponds, with a view to utilize it as an alternative source of water for consumption and recreational activities.

B) To discriminate ex-mining ponds from lakes that had no mining operation with a view to sort out the targeted toxic heavy metals using chemometric tools.

C) To ascertain the health risks associated with exposures to water from ex-mining ponds by ingestion and dermal contacts.

D) To develop the water quality index of water from ex-mining ponds using the relevant selected parameters.

1.6 Thesis Outline

The general background of the research and related subject areas are provided in the first two chapters which forms the introduction and literature review. Chapter 1 points out the aims and objectives of this research work. Chapter 2 presents the literature review on the development of water quality index, risk assessment on exposure to hazardous metal pollutants, and multivariate statistical techniques.

Chapter 3 discusses the methods and methodology applied in analyzing water quality parameters. The instrumentation, preservation, and analysis of water samples are discussed. It discusses the climate, geology, and activities around the studied sites.

Chapter 4 evaluates the details related to the trend in variation of heavy metal concentrations and physical-chemical parameters in ex-mining ponds and lakes in comparison to INWQS. It discusses levels of pollutants, sources and influence on water quality. Activities around the sample locations are also examined to monitor their influences. The chapter discusses and examines the results of variations and characterization of the studied parameters using PCA, HCA, FA and LDA. It explains the risks associated with exposure to the ex-mining water and lakes by ingestion and dermal routes, and finally the development and application of WQI in the classification of ex-mining and lake waters is discussed.

Chapter 5 presents the overall conclusion of the thesis based on the findings obtained.

CHAPTER 2: LITERATURE REVIEW

This chapter analyzes different WQI models used to assess the surface water quality of lakes and rivers. The WQI models and their applications are examined in section 2.1, and section 2.2 studied health risk assessment of exposure to ex-mining water. Section 2.3 focuses on multivariate statistical techniques and its application in lake water quality assessment in Malaysia.

2.1 Water Quality Index Models

Various models of WQI have been efficiently utilized in pollution evaluation of water resources, and their application depends on the purpose and objectives to which they are intended for. Besides pollution monitoring of water resources, the development of WQI may cover trend analysis and enforcement of standards. The health risk evaluation of exposures to hazardous substance in water is an integral part of water quality monitoring and assessment. The parameters involved in the quantitative evaluation of ingestion and dermal exposures to ex-mining water are key aspects to consider. Multivariate statistical models are essential in monitoring environmental influence on surface water quality. Application of a specific chemometric technique depends on the objectives to be achieved; LDA and PCA are mostly utilized in this study to differentiate the most significant parameters, and to determine the latent variables in ex-mining ponds and lakes respectively.

Horton's approach to the development of WQI back in 1965 was the first attempt to represent overall water quality using the numerical index system with 10 variables (Coliform density, specific conductance, pH, DO, carbon chloroform extract, temperature, alkalinity and chlorides, sewage treatment, and obvious pollution) and ranges from 0-100 (equation 2.1). Though the formulation of WQI models was faced with challenges from parameter selection to subjectivity in assigning weightages to the selected parameters, Horton's technique is easy to compute and highlights the challenge of the right choice of parameters to represent the overall water quality (Horton, 1965). These are the major advantages of Horton's approach in water quality assessment. The weaknesses are that index formulation, quality rating, and weightages are highly subjective based on his judgement and few associates.

$$WQI = \left[\frac{w_1S_1+w_2S_2+w_3S_3+\ldots+w_nS_n}{w_1+w_2+w_3+\ldots+w_n}\right] m_1m_2 \qquad \textbf{(2.1)}$$

where; S = sub index, w = weighting factor (1-4), m_1 = correction factor for temperature (0.5 when temperature < 34 °C, otherwise 1), m_2 = correction of pollution (0.5 or 1).

The British Columbia water quality index (BCWQI) was developed in the year 1995 by the Canadian ministry of environment for the evaluation of water quality. The formulation of this index is similar to the Canadian Council of Ministers of Environment (CCME) where the violation of the measured water quality parameters is determined by comparison with the established limit. The number of repeated sampling and stations determines the accuracy of the BCWQI. Its major limitation is the deviation in the water quality trend from the standard limit due to maximum percent deviation used (Salim et al., 2009), also the index is not appropriate for the spatial and temporal trend analysis. Advantage of BCWQI is the sensitivity of the index value to the parameters incorporated in the calculation and objectives interpretation of the reference condition (Zandbergen & Hall, 1998). The final index value is calculated using equation 2.2;

$$BCWQI = 100 - \left(\sqrt{\frac{F1^2 + F2^2 + F3^2/3^2}{1.453}}\right) \qquad \textbf{(2.2)}$$

where F1 is the percentage of variables above the guideline, F2 is the frequency by which the objectives are not met, and F3 is the range to which the failed tests are above the guideline. 1.453 in the formula is the scaling factor of 0-100.

In the year 1990, Smith's Index was developed which serves as a hybrid based on expert opinion as well as water quality standards. Its major advantage is the index addresses four types of water use including contact and non-contact uses (water supply, fish spawning, general, and regular public bathing). Delphi method was applied in parameter selection, developing sub-indices, and assigning weightages. The final index score was calculated using minimum operator technique as displayed in equation 2.3 below;

$$I_{min} = \sum \min (I_{sub1}, I_{sub2}, \ldots\ldots\ldots\ldots I_{subn}) \quad \textbf{(2.3)}$$

where I_{min} equals the lowest sub index value (D. G. Smith, 1990).

This approach utilizes additional rounds of questionnaire beside the usual Delphi steps. This is aimed to arrive at a greater convergence of opinion (Abbasi & Abbasi, 2012). Smith's approach was considered more prudent and slight deviation from the standard Delphi procedure that discouraged discussion among the panel members on the assumption that such discussion may influence the decision of others. Unlike indices with relative weighting, advantage of smith index is introducing or omitting new determinant at a later stage without affecting the index formulation. Also eclipsing effect of multiple over few determinants is reduced to barest minimum.

The creation of national sanitation foundation water quality index (NSFWQI) based on a nationwide survey of water quality experts and application of the Delphi technique addressed the problem of parameter choice in a more consensual manner. The NSFWQI accommodates 9 parameters (BOD, DO, turbidity, total solids, nitrate, phosphate, temperature, pH, fecal coliform). The multiplicative version of NSFWQI (equation 2.5) was considered a more superior formulation compared to the additive (equation 2.4) which lacks the sensitivity with respect to the effect of a single bad parameter on the WQI (MERC, 2012).

$$\mathrm{WQI} = \sum_{i=1}^{n} w_i q_i \qquad (\mathbf{2.4})$$

$$\mathrm{WQI} = \prod_{i=1}^{n} q_i^{w_i} \qquad (\mathbf{2.5})$$

where; w = weighting factor (0-1), q = water quality rating curves (0-100%), n = number of parameters. The advantages of NSFWQI are providing useful insights into temporal water quality trends, the index value relates to potential water use, and evaluation between areas and identifying changes in the water quality. Its limitation involves losing vital information during data processing, and lack of dealing with uncertainty and subjectivity present in complex environmental issues (Gopaul et al., 2009; Tyagi et al., 2013).

Canadian council of ministers water quality index (CCME-WQI) is a modification of BCWQI, it comprises of three factors each of which is scaled between 0 and 100. The factors are; scope, frequency, amplitude (equation 2.6).

$$\text{CCME-WQI} = 100 - \left(\sqrt{\frac{F1^2 + F2^2 + F3^2}{1.732}}\right) \qquad (\mathbf{2.6})$$

F1 is the scope which assesses the extent of water quality guideline over the period of interest, F2 represent percentage of individual tests that do not meet the objectives, and F3 illustrate the amount by which the failed test values met their objectives. The value of 1.732 is the correction factor of the overall index (CCME, 2001). CCME-WQI is best suited for use in continuous monitoring of water quality. The advantages of CCME-WQI are utilizing many variables, easy to calculate, and tolerance to missing data. The major weakness is the insensitivity of a particular parameter in the process of aggregation where the same importance is given to all variables (Tyagi et al., 2013).

Several indices were developed either on the basis of Hortons or NSFWQI, among which are the stream monitoring, Oregon, and universal WQI (Boyacioglu, 2007; Cude, 2001; Hallock, 2002; Liebman, 1969) as shown in Table 2.1. The indices formulated encompass physical, biological and chemical parameters and different priority was given in terms of quality rating and weightage to suit the desired objectives.

The Malaysian water quality index (DOE-WQI) is also a derivative of NSFWQI developed to monitor river water quality based on 6 parameters only (pH, DO, BOD, COD, SS, and AN) as given by equation 2.7 below;

$$WQI = 0.22*SIDO + 0.19*SIBOD + 0.16*SICOD + 0.15*SIAN + 0.16*SISS + 0.12*SIpH \quad (2.7)$$

where SIDO = sub index DO, SIBOD = sub index BOD, SICOD = sub index COD, SIAN = sub index AN, SISS = sub index SS, SIpH = sub index pH. $0 \leq WQI \leq 100$. The WQI model utilized by Malaysia DOE (equation 2.7) is the only existing and functioning tool to monitor river water quality in Malaysia (Naubi et al., 2016). It exclusively contains physical-chemical parameters hence cannot be used to assess the water quality of ex-mining ponds and lakes that are mostly polluted with heavy metals (Low et al., 2016). In this study, toxic heavy metals associated with mining operation such as As, Cd, and Pb (Acheampong et al., 2013; Low et al., 2016; Martha et al., 2011; Popa et al., 2013; Schaider et al., 2014) are selected in the formulation of the WQI of ex-mining ponds considering geological and other environmental factors. The

Malaysian DOE-WQI was used to assess water quality status of ex-mining ponds in Bestari Jaya (Kusin et al., 2016). However, it has been confirmed that river WQI cannot be used to assess ex-mining ponds and lakes due to its inability to accommodate the toxic metals especially As and Cd (Low et al., 2016).

The weakness of the pioneer indices and its derived models (DOE-WQI inclusive) is its formulation based on the Delphi method with much subjectivity due to difference in opinion of the experts on the perceived parameter importance to water quality (Singh et al., 2007; Tarantola, 2000). Notwithstanding this, the class IIA standard parameter values of Malaysian INWQS were used in this study to assign weightage to the selected parameters; this qualifies the WQI model developed to have a direct evaluation of the status of ex-mining water in Malaysia especially in its consideration for consumption and domestic uses. Furthermore, the index developed in this study utilizes validated primary data obtained from standard experimental procedures, unlike the existing Malaysian DOE-WQI which only exploited secondary data obtained from different water monitoring stations with different equipments and calibrations. It has been established that data from secondary sources may be unreliable (Sullivan et al., 2003); this qualifies the new WQI of ex-mining ponds to be more dependable and specific in addressing this research problems. The indices developed, including the Malaysia DOE-WQI did not recognize and integrate the status of the water for different functions and uses such as drinking water supply, recreation and irrigation. It only classifies the waters based on pollution with no recommendation for its future utilization (Low et al., 2016). In this study, recommendations are made on uses of the ex-mining ponds and lakes for human consumption and/or recreational activities.

Table 2.1: Selected WQI of surface water

Index	Parameters	Scale
Horton's Index	DO, pH, sewage treatment, coliform density, specific conductance, carbon chloroform extract, alkalinity, chlorides, temperature and obvious pollution.	0-100
National Sanitation Foundation Index (NSFWQI)	BOD, DO, total solid, turbidity, nitrates, phosphates, temperature, fecal coliform.	0-100
Oregon WQI	BOD, DO, temp, AN, pH, Total phosphorus, total solid, and fecal coliform,	0-100
Universal WQI	Nitrate, BOD, DO, As, Fluoride, Total phosphorous, Hg, Se, CN, Cd, total Coliforms, and pH.	0-100
Canadian WQI (CCME WQI)	Relevant water quality parameters with corresponding standards.	0-100
DOE-WQI	BOD, COD, DO, SS, AN, and pH.	0-100
General WQI	Total phosphorous, DO, fecal coliform, turbidity, and specific conductivity.	0-3
River Status Index	DO, BOD, AN, SS, turbidity, temp, fecal coliform, pH, Pb, Cu, Cd, Cr and Zn.	1-10
Stream Monitoring WQI	DO, temp, pH, fecal coliform	0-100

2.2 Risk Assessment Models

Several human health risk assessment techniques involve the use of formulation developed by the United States Environmental Protection Agency and the Agency for Toxic Substances, Diseases and Registry (ATSDR, 1993; USEPA, 1992, 1999, 2005). The models utilized in the risk assessment studies (equations 2.8 and 2.9) include the exposures due to ingestion and skin surface (Alkarkhi et al., 2009; Gupta et al., 2015; Igwe et al., 2014; Iqbal & Shah, 2013; Low et al., 2016; Li et al., 2013; Man et al., 2010). The indices uses USEPA default values for volume of water ingested (2.2 L/day), and body weight (70 kg and 15 kg for adult and child respectively).

Consideration was not given to the differences in water consumption and the weight of individuals in a particular region or country of study (WHO, 2009). It has been established that different country has dissimilar average weights and daily water consumption due to differences in dietary, climate, social and economic status (Eveleth & Tanner, 1976; Michaelsen, 2015). In Malaysia, the average weight for an adult and child are 63.69 ± 11.42 kg and 32.89 ± 8.95 kg respectively (Azmi et al., 2009; Gan et al., 2011; Zaini et al., 2005). The skin surface area exposed on the other hand are 5700 and 2800 cm^2/day, respectively for adult and children (Sany et al., 2014). The average volume of water consumed by Malaysian adult and children is 1.7 and 1.2 L/day, respectively (MDG, 2013; Mohamad, 2016), and average lifetime exposure (AT) of Malaysians is 74.7 years (27265 days) in 2016 (DOS, 2016). The exposures due to ingestion and dermal contacts are evaluated using equation 2.8 and 2.9 below.

$$Exp_{Ix} = \frac{C_{water} * IR * EF * ED}{BW * AT} \quad \textbf{(2.8)}$$

$$Exp_{Dx} = \frac{C_{water} * SA * K_p * ET * EF * ED * CF}{BW * AT} \quad \textbf{(2.9)}$$

where; Exp_{Ix} and Exp_{Dx} are exposure doses due to ingestion of water and skin surface (µg/kg/d) respectively, C_{water} is the concentration of metal in water (µg/L), IR water ingestion rate (2.2 L/day) and Child (1.0 L/day), EF exposure frequency (360 days/year), ED exposure duration (30 years), BW adult average body weight (70 kg) and Child (15 kg), AT average time (25550 days), SA exposed skin area (28000 cm^2/day), ET exposure time (0.6 hr/day), CF unit conversion factor (0.001 L/cm^3), and Kp is dermal permeability coefficient for specific metal (cm/h) (Smith, 1994; USEPA, 1989).

Most risk assessment studies in Malaysia also used hypothetical weights or measurements obtained from few selected respondents in the hazard evaluation (Ashraf et al., 2011a; Dzulfakar et al., 2011; Qaiyum et al., 2011). Similar approach was applied in the risk analysis of exposure to ex-mining water (Alshaebi et al., 2009). In this study, the existing models were modified where exposure at different human weights, volume of the water consumed, average exposure time, and skin surface area exposed were altered so as to give a more accurate health risks evaluation. This shows a new trend in the risk analysis and specificity of the modified models in determining the health risk associated with utilizing ex-mining water in Malaysia.

2.3 Statistical Modelling/Chemometric

It is a process of predicting from the observed data sets with a view to extract information on the relationship between variables. The specified mathematical equations used are the statistical models that relate one or more variables; it is therefore a formal presentation of a theory. The choice and application of the appropriate statistical technique are recommended for the hypothesis test, interpretation of data and to arrive at logical assumption. The validity of the data can be evaluated or examined using descriptive statistics, plots and graphs which gives an exploratory idea of the relationship between model variables. There are myriad of modelling and statistical analysis techniques, the choice of which depends on the data type and objectives.

2.3.1 Linear Regression Model (LRM)

Regression analysis is the most widely used statistical technique for investigating and modelling the relationship between variables (Montgomery et al., 2015). Linear regression modelling has been applied in environmental researches and applied sciences to establish correlation among variables. This modelling approach explains the relationship between a dependent (response) variable and independent (explanatory) variable. A dependent variable of interest is modelled as a linear combination of one predictor variable. Linear regression analysis yields an approximate b parameter of the linear equation as given in equation 2.10 below;

$$y = a + bx \tag{2.10}$$

where a is the intercept, y is the predicted score of the dependent variable (response), b represent the slope and the contribution of the independent variable x to the prediction of y.

The relationship between variables of interest should first be studied before a linear model is assumed suitable using the observed data. This does not certainly indicate that one variable influences the other, but there is notable association between the two variables. The validity of the scatter plot can be used to identify the relationship between the variables (Montgomery et al., 2012).

Linear regression analysis has been universally applied in studies related to modelling of water quality, treatment and management including development of rating curves for WQI of surface and underground water (Buytaert et al., 2016; Lermontov et al., 2009).

2.3.2 Multivariate Statistical Models (MSM)

The relationship between multiple parameters can be analyzed using MSM, also known as chemometric or environmetric techniques. It was hypothesized that a desirable response in some researches is often determined or influenced by more than one variable and therefore gives a better and more reliable data evaluation (Varmuza & Filzmoser, 2016). MSM is used for the classification of temporal and spatial data, and characterization of different environmental samples. The most widely utilized MSM in environmental research are the principal component analysis (PCA), factor analysis

(FA), hierarchical cluster analysis (HCA) and linear discriminant analysis (LDA) (Chow et al., 2016; Gasmi et al., 2016; Juahir et al., 2008; Juahir et al., 2010; Juahir et al., 2011; Krishna et al., 2009; Muangthong & Shrestha, 2015; Simeonov et al., 2002; Varol et al., 2012; Zhao et al., 2012).

PCA (also called eigenvector analysis) is mostly utilized as data reduction and modelling technique. It determines the degree or extent to which variables are related. Large data of many variables are unavoidably superfluous and overlap, the use of correlation matrix generally quantifies these anomalies by extracting the eigenvalues and eigenvectors from the square matrix originated by multiplying the data matrix. PCA introduces uncorrelated linear functions of the standardized variables called principal components (Simeonov et al., 2002). In FA, the varimax rotation is applied on the significant principal components to produce new class of variables named varimax factors. The loadings of a variable (positive or negative) determine its importance and contribution to the variation. HCA uses Euclidean distance, which is generally a reliable measure of similarity between the sites. A suitable linkage algorithm (Ward's, single, centroid, or average) is established to link the cluster (group) of sites or objects with similar distances and to isolate those clusters located at large distances (Simeonov et al., 2002). The correct interpretation of the clusters is a crucial task and allows a good explanation of justifications leading to the type of clustering. LDA determines the variables that distinguish one group from another by constructing discriminant function (DF) for each group. LDA establish a DF for each group given various independent variables and dependent variables (Juahir et al., 2010). The forward, backward, and stepwise approaches are used to construct the DFs to evaluate the differences between groups. The stepwise method has been widely applied in the environmental data analysis. Four of the MSM are utilized in this study and comprehensively discussed in chapter 4.

2.3.3 Monte Carlo Simulation Technique (MST)

MST is applied to obtain a forecast in statistical probability distribution where analytical solutions are impossible or difficult to obtain, or when specific regression assumptions are under suspicion of violation. The simulations are repeated a large number of times with the model defined for each repetition by the values of the stochastic inputs to obtain statistics output variables of the system model (Mooney, 1997). The possible value of the input variable X (X_1, X_2,… X_n) are sampled according to their distribution, the value of the output variable Y are then calculated through performance function Y = f (X) at the samples of input variable (Robert, 2004). MST is applied for dependent variables and the three steps generally required are;

A) Sampling on random input variables X

B) Evaluating model output Y

C) Statistical analysis on model output

The purpose of selecting the input variable is to create samples that represent distribution of the input variables, the samples of the input variable will then be used to the simulation experiment. After samples of output variable Y have been obtained, the characteristics of the output variable such as mean, variance, reliability, can be statistically analyzed.

MST was successfully applied in water quality modelling and sensitivity analysis (Beck, 1987; Ma et al., 2000; Van-Griensven et al., 2006; Whitehead & Young, 1979). Most recent studies related to environmental problems used MST in water quality modelling, management and decision making (Han & Zheng, 2016; Kuria & Vogel, 2015; Shojaei et al., 2015; Sun et al., 2015; Zhi et al., 2016). MST was also applied in risk assessment based on water quality model, and risk analysis due to pollution in rivers and lakes (Alam et al., 2015; Geng et al., 2016; Langhans et al., 2016; Lu et al., 2015; Yao et al., 2015). MST was applied in a study on water quality and multiple pathway exposure assessment of human health risk in water (Harris et al., 2017; Jiang et al., 2013; Saha et al., 2017; Yao et al., 2015). Numerous researchers applied MST in studies related to environmental modelling (Alves et al., 2014; Cao et al., 2014; Cao et al., 2015; Chabukdhara & Nema, 2013; Lonati & Zanoni, 2013; Qu et al., 2012).

2.4 Heavy Metals in Ex-mining ponds and Lakes

Most heavy metals occur naturally in the environment as ores, but their concentrations in the environment increase significantly from anthropogenic sources such as industrial and farming activities, thereby altering their geochemical distribution. Among the anthropogenic activity that discharges heavy metal and polluting the environment is mining (Akcil & Koldas, 2006; Ashraf et al., 2011b; Ashraf et al., 2015; Igwe et al., 2014; Razo et al., 2004; Younger & Wolkersdorfer, 2004). The heavy metal pollution associated with mining activity received global attention in many countries that engaged in mineral exploration including Malaysia (Acheampong et al., 2013; Alshaebi et al., 2009; Atanacković et al., 2013; Fields, 2003; Jones et al., 2013; Williams, 2001; Younger et al., 2002b). Several studies also reported the presence of toxic heavy metals in ex-mining water and the surrounding environment (El Khalil et al., 2008; Fadiran et al., 2014; Jidon, 1993; Luís et al., 2011; Navarro et al., 2008; Rybicka, 1996).

Contamination of surface water was reported in lake Dianchi China, with high concentration of Pb with other heavy metals also exceeded their background values, the source of which was traced to industrial pollution and automobiles (Wang et al., 2014). Four great rift lakes in Kenya were polluted with As and Hg to a level that might pose adverse health effect. Other metals like Cd, Pb, Cr, Cu, Zn, Ni, and Mn were found to be of lower concentrations. High levels of As and Hg in the lakes were generally linked to the anthropogenic factors and less of natural factors (Yang et al., 2017a). Similarly, Cd, Pb, Mn and Zn were found to contaminate the surface water of lake Manzala in Egypt due to continuous discharge of untreated water into the lake (Hamed et al., 2013). Elevated concentrations of Cr, Pb, Co, and Mn in Mariut lake were found to be above USEPA limits, which were linked to the industrial activities near the lake (Rostom et al., 2017). Considering that level of metal pollutants in the lakes exceeded the approved standard limits, there is likelihood of toxic effects either through the food chain or by direct utilization.

The toxicity of heavy metals varies, and the main threat of heavy metals to human health is associated with exposures to arsenic, cadmium, lead and mercury. The effects of these metals on human health had been studied extensively with regular review by international health organizations (Järup, 2003). Government agencies, researchers and the public have been raising concern on the metal concentrations in large volumes of ex-mining water, particularly with the increasing global demand of water for domestic and recreation, and forecast for water crisis by the year 2025 (Rosegrant et al., 2002).

2.4.1 Arsenic

Arsenic is commonly found in nature as mineral ore with other elements in soil and rocks. It is discharged into the environment by soil leaching, volcanic activities and weathering of rocks. Mining is considered one of the major sources of arsenic in the environment which is released in substantial quantity from its common sulfide ores such as arsenopyrite (FeAsS) and less commonly in tennantite ($Cu_{12}As_4S_{13}$) and enargite (CuAsS) (Gul et al., 2013). Besides the mining operation, water at low pH or from acid rain could facilitate the dissolution of metals from their ores thereby elevating the arsenic concentration in the surrounding environment (Acheampong et al., 2013;

Yaacob et al., 2009). Arsenic could also be released into ground water by natural oxidation of the sulfide minerals in bedrock aquifers under alkaline and anaerobic conditions (Kim et al., 2000; Williams, 2001). Arsenic is toxic and considered the third most toxic substance after lead and mercury in the United States toxic substance and disease registry. The exposure to its toxicity is mostly by drinking arsenic contaminated water and food (Murcott, 2012).

The most common and abundant arsenic species in aqueous compounds are the inorganic arsenite (As (III)) and arsenate (As (V)), while the organic arsenic compound arsenobetaine ($C_5H_{11}AsO_2$) is found predominantly in fish; both of which may result in high human exposure (Bissen & Frimmel, 2003; Järup, 2003). The concentrations of arsenic in urine, blood, nails and hair have been used as bio indicators of exposure. Acute toxicity of arsenic causes digestive, gastrointestinal and central nervous disorder, cardiovascular disturbances and finally death (Mandal & Suzuki, 2002). In the event of victim survival, haemolysis, bone marrow depression, and melanosis may be observed. Evaluation done by WHO concluded that exposure to arsenics through drinking water causes cancer related to kidney, lungs, and precancerous lesion (WHO, 2001). Ingestion of arsenic to a concentration of 100 µg/L lead to cancer, and skin cancers are associated with 50 – 100 µg/L of arsenic (Järup, 2003). Several research findings also discussed the health effects of exposure to arsenic at different concentration levels (Chen et al., 2016; Kazi et al., 2016; Ramakrishnan, 2015; Rasheed et al., 2016).

Several cases of arsenic poisoning were reported and documented in Malaysia and around the globe as a result of mining and other anthropogenic activities. In Malaysia, a case of 3 patients with cutaneous lesion was reported. The patients developed chronic arsenical poisoning due to ingestion of underground well water close to a tin mining site (Jidon, 1993). Hassan (2005) reported that the arsenic poisoning from drinking water in

Bangladesh was the worst mass poisoning in human history (WHO report). The concentration of arsenic was high in ground water, which affects 61 out of the 64 districts, where 36,477 patients were admitted. The cases of skin cancer and cardiovascular mortality in Chile and Taiwan as a result of ingestion of arsenic-contaminated water received global attention as well. The levels of arsenic ingested was 0.8 mg/L to 1.82 mg/L (ATSDR, 2007). About 6,000 cases of arsenic poisoning as a result of ingestion of arsenic-contaminated beer was reported in England with 71 deaths in the year 1900. The concentration of arsenic in the beer ranged from 2 mg/L – 4 mg/L (Reynolds, 1901). The permissible level of arsenic in drinking water for WHO and drinking water quality standard for raw water (DWQSRW) is 10 µg/L, and the Malaysian INWQS is 50 µg/L (DOE, 2012; MOH, 2004; WHO, 2011).

High arsenic concentrations of 420 µg/L in ex-mining water of Mexican mining districts of Santa Maria de la Paz was found to be 20 and 8 times higher than the natural background concentration and the Mexican drinking water quality standard respectively (Razo et al., 2004). A study conducted in south and southwestern Nigeria revealed arsenic concentrations in the ground water aquifers of the basement rock ranging from 3 to 25 µg/L and in the sedimentary rock from 6 to 40 µg/L (Murcott, 2012). Elevated arsenic concentration was detected in 98 lakes close to the mines in Yellowknife area Canada, which is related to historic mining operation. Some of the lakes recorded 60 times the limit of 10 µg/L set by the federal drinking water guideline (Palmer et al., 2016). The mine tailing of a historic mining site in Ghana elevates the concentrations of arsenic to 73 µg/L in the river Tarkwa and nearby streams (Asante et al., 2007). Therefore, contamination of water with arsenic due to mining and other anthropogenic is a threat to the environment, and needs to be fully studied and monitored to prevent the risk of exposures especially by ingestion.

2.4.2 Cadmium

Cadmium is found in nature in very small quantities, and natural sources in the earth's crust are related to volcanic activities and rock weathering, and as an impurity in the ores of metals such as zinc, lead, and copper. Presence of cadmium in the environment was highlighted in the previous studies, and the source related to metal ores (Bebbington & Williams, 2008; Limei et al., 2008; Naidoo, 2017; Sun et al., 2010). Cadmium is mostly a byproduct of sphalerite (ZnS) found in mineral greenockite (CdS), it is also found as an impurity in phosphate minerals (Cobb, 2007; Donovan et al., 2016). Several findings revealed that anthropogenic sources of cadmium include mining and metallurgical industry, combustion of fossil fuel and phosphate fertilizers (Quraishi et al., 2015; Shi et al., 2016). Weathering of rocks in particular releases cadmium to the aquatic ecosystem contributing to the global cadmium cycle, which is enhanced by acidic emissions (UNEP, 2010). The levels of cadmium in most drinking water supplies are less than 1 µg/L, though levels may vary depending on locality and the acidic influence of the environment. The WHO and DWQSRW permissible levels of cadmium in drinking water is 3 µg/L, and INWQS is 10 µg/L (DOE, 2012; MOH, 2004; WHO, 2011).

Cadmium is categorized as a human carcinogen by the International Agency for Research on Cancer (IARC, 1993). There was strong evidence that links cadmium to lung and prostate cancers (Nawrot et al., 2015; Sorahan & Lancashire, 1997). Also related to cadmium exposure is increased risk of kidney diseases and difficulty in learning for children, and loss of sense of smell (anosmia). Most damage caused by cadmium to the kidneys is irreparable (Ciesielski et al., 2012; Navas-Acien et al., 2009). Cadmium is also confirmed to cause toxicity, skeletal demineralization, impairment of the pulmonary functions, and elevation of blood pressure (Bernard, 2008; Staessen et al., 1999; Tellez-Plaza et al., 2008).

A study in Japan relates cadmium to increased mortality due to heart failure and cerebral infarction (Nishijo et al., 2004). Elevated urinary level of cadmium was noticed in Belgium that leads to the subsequent development of lung cancer. Tubular dysfunction was noticed from the general population due to high levels of urinary cadmium in the range of 1 to 2 µg/g. Similarly, the higher threshold value of 8 to 10 µg/g was observed among the Chinese population (Bernard, 2008). Low bone density was associated with cadmium exposure in a study comprising of 1021 persons in southern Sweden. A significant negative correlation was observed between the concentration of cadmium in the urine and low bone mineral density (Barregard et al., 2016; Järup et al., 1998; Kakei et al., 2016).

In New Zealand, cadmium concentration of 100 mg/Kg were released to the environment from phosphate fertilizers applied during farming, which is absorbed by plants and subsequently consumed by humans (Taylor, 1997). Similarly, drinking water from wells in Sweden recorded a cadmium concentration of 5 µg/L due to the influence of acidic soil. Cadmium concentration of 100 µg/L was also reported in natural water in Peru, and this value is higher than concentrations of < 1 µg/L obtained in 110 stations around the world (WHO/UNEP, 1989). In a similar study in Saudi Arabia, potable water from underground sources recorded a mean cadmium concentration of 1 – 26 µg/L (Fowler et al., 2015; Langard et al., 1986; Mustafa et al., 1988). The influence of mining activity in Bolivia raises the cadmium concentrations in the river of Pilcomayo catchment, an area that experiences extensive tin mining operation (Smolders et al., 2003). In a related finding, the analysis of water samples from Dabaoshan mine and the adjacent Hengshi river in China revealed high cadmium concentrations of 8.05 mg/L (Zhuang et al., 2009). High metal discharges in UK from abandoned mines into the rivers, about 6% of the river catchment in England and Wales have cadmium as the major pollutant of concern (Mayes et al., 2009). This raises the alarm on the safety of the people living along the river due to exposure to cadmium by either ingestion or body contacts.

2.4.3 Lead

Lead is a naturally occurring element but not particularly abundant. Its ores are distributed across the world. Natural lead occurs mostly as galena (PbS), and less commonly as cerussite ($PbCO_3$) or anglesite ($PbSO_4$), the occurrence of lead in these ores qualifies it to support the mining industry (Bao et al., 2016; Kosobrukhov et al., 2004; Zimdahl et al., 1973). The distribution of lead in the environment, especially soil and water is altered by man's activities due to its ability to remain virtually immobile. Lead can be released into the environment by the natural process of weathering and other sources related to human activity such as mining, burning of fossil fuel and industrial discharges (Loh et al., 2016; Marín et al., 2016; Puga et al., 2015). The crushing and panning processes in mining release a substantial concentration of lead into the soil, surface and underground water. A significant concentration of lead in soil from three different land uses with mining have been reported (Martín et al., 2014). High concentration of lead in ex-mining water was reported in some literature which might have originated from the dissolution of the ores, this find its way to the underground water and nearby streams (Bui et al., 2016; Lu et al., 2015; Rybicka, 1996; Zhang et al., 2012). The permissible concentration of lead in Malaysian DWQSRW and WHO are 50 µg/L and 10 µg/L, respectively (MOH, 2004; WHO, 2011).

Lead is poisonous with several toxic effects that are permanent. Lead toxicity, particularly affects children, because their body system absorbs lead faster than the adult with little or no symptoms thereby destroying their brain and nervous system. The result is low intelligent quotient (IQ), slowed growth and anemia. Its effect on pregnant mothers includes reduced growth of the fetus and premature birth. Ingestion of lead causes coma and subsequent death (Arnemo et al., 2016; Hanna-Attisha et al., 2016; Mielke, 2016; Needleman & Bellinger, 1991; Xie et al., 2013).

The lead poisoning and death of over 350 children in Zamfara Nigeria as a result of water and soil pollution from active gold mines attracted global attention. The blood lead level (BLL) of the children < 5 years was analyzed to be > 45 µg/dl higher than the limit of 10 µg/dl set by the U.S. center for disease control and prevention (Bello et al., 2016; Canfield et al., 2003; CDC, 2002; Lo et al., 2012; Thurtle et al., 2014; Wurr & Cooney, 2014). Similar incidence occurred in Kabwe, Zambia where children developed high BLL due to extensive contamination of soil and surface water with lead from historic Pb-Zn mine site (Yabe et al., 2015). In the United States of America, 20% to 40% of the rises in BLL of children are due to exposure to lead through drinking water (Davis et al., 1992).

Attention was raised in the Northwest of England over lead pollution in the Ullswater's lake which supports many consumers that extended down to the Manchester. This places a burden on the regional government to find alternative sources of water supply (Beattie, 1996). An effort was made to assess the potential losses caused by the continuous pollution of Ullswater lake by the Greenside mine (Bardsley, 2000). Glenridding Beck River was polluted with lead from the mines, raising the concentrations above natural background to 0.029 mg/L and 5.85 mg/kg for the surface water and sediment respectively (Mitchell, 2009). Similarly, lead concentrations in about 53% of well water studied near mining and smelting site in Zanjan, Iran were higher than WHO standard limit (Mohammadian et al., 2008). The pollution of surface and underground water with lead due to mining and other anthropogenic sources is therefore of concern due to its mobility and toxic nature (Sankhla et al., 2016; Wongsasuluk et al., 2014).

2.4.4 Manganese

Manganese is very abundant and distributed in the earth's crust in association with ores of iron. The major ores of manganese are pyrolusite (MnO_2), hausmannite (Mn_3O_4) and manganite (MnO(OH)). Manganese is an essential trace element found naturally in lakes, rivers, and drinking water supply (Keen & Zidenberg-Cherr, 1994; Muthaiah et al., 2016; Takeda, 2003). Mining operation elevates the concentrations of manganese and iron beyond the permissible limits, especially in the surface water. Though not a priority pollutant, high manganese concentration in the environment may become a threat to humans and aquatic organisms (Caruso et al., 2012). The health based values of manganese in drinking water are 0.4 mg/L and 0.2 mg/L, respectively for WHO and DWQSRW (MOH, 2004; WHO, 2011).

Manganese is considered among the least toxic elements, however, manganese concentrations of 1 mg/L can be toxic to aquatic organisms, and countries around the world adopted 0.2 mg/L as the safe limit for the protection of aquatic species (Howe et al., 2004). Prolonged exposure to manganese on human is associated with toxicity to the nervous system, resulting in syndrome similar to Parkinsonism. Several studies have associated high manganese concentrations in drinking water with the neurological defects in children (Bouchard et al., 2011; Khan et al., 2012; Kondakis et al., 1989; Oulhote et al., 2014). The exposure to elevated concentrations of manganese at the prenatal and early childhood results in the children's behavioral problems (Rahman et al., 2016). The case of chronic manganese poisoning was also diagnosed in a 52-year old man due to high exposure from the manganese ore crushing site. The autopsy revealed distribution of manganese in the brain, which resulted in a neuropsychiatric problem such as monotonous speech, euphoria, emotional inconsistence and tremor of the eyelids (Yamada et al., 1986).

About three public water supplies were polluted in Georgia due to high manganese concentrations exceeding the water quality guideline of 0.4 mg/L. The river Kvirila recorded high manganese concentrations in the surface water and the sediments above the standard permissible values of 0.1 mg/L and 1500 mg/kg respectively due to pollution from mine water (Caruso et al., 2012). The pollution of the underground water samples above the permissible value was also observed in Greece where many samples analyzed reached 3,700 µg/L (Varnavas, 2016). In a study in Japan, dissolved manganese concentration of 28 mg/L was found in drinking water 70 times above the guideline of 400 µg/L which resulted in 16 cases of poisoning (Frisbie et al., 2012). These findings call for proper analysis and evaluation of manganese in ex-mining water, also in raw and treated waters considering the high natural abundance of manganese.

However, previous findings on exposure to manganese in fish, soil and tailings from the former mining area in Malaysia revealed little, no health threat, or not even considered in the risk analysis (Ashraf et al., 2012; Ashraf et al., 2011a; Low et al., 2015; Yaacob et al., 2009).

2.4.5 Other Metals

Ex-mining water may contain significant concentrations of other metals that are considered essential for a healthy life. The metals include zinc and copper, which exist with other ores or as impurities and gets into the environment in the cause of mining processes. The occurrence and pollution of the metals had been thoroughly discussed in the literature (Chowdhury et al., 2016; Flemming & Trevors, 1989; Glover, 1983; Jahanshahi & Zare, 2015; Salvarredy-Aranguren et al., 2008). In spite of the health importance of these metals, they could pose a threat at high concentrations and longtime exposure. However, the metals are not included in this study due to their low toxicity and are of less concern in several reports of Malaysian ex-mining ponds (Ashraf et al., 2011b; Hamzah et al., 2011; Kusin et al., 2016; Orji et al., 2014).

2.5 Physical-Chemical Parameters in Ex-mining Ponds

Besides the consideration of the metal levels in ex-mining water, another crucial factor worthy of concern is the physico-chemical parameter levels. The concentrations of these parameters have direct influence on the metal levels especially pH. The concentrations of the metals are also affected by seasonal inconsistencies (Cánovas et al., 2008; Caraballo et al., 2016). Other parameters that determine the aesthetic nature of the water, especially the user acceptance based on the physical appearance are also considered. The physico-chemical parameters of interest are pH, biological oxygen demand (BOD), dissolved oxygen (DO), suspended solid (TSS), and ammoniacal nitrogen (AN).

2.5.1 pH

Several research findings recognize pH as one of the most important parameter**s** that determines the quality of water for human consumption and applications (Abbasi & Abbasi, 2012; Boyd, 2015; Kannel et al., 2007; Ravikumar et al., 2013). The beneficial nature of any water source is in the pH range of 6.0-8.5 (Garg et al., 2010). The pH ranges of 5.5-9.0 and 6.0-9.0 are the limits for the Malaysian DWQSRW and INWQS respectively (DOE, 2012; MOH, 2004). In recent times, the standard limits are difficult to maintain due to increase in anthropogenic activities such as mining and mineral exploration. The mining operation generates acidic water as a result of oxidation of the metal ores in the presence of water and oxygen. The acid generated further attack other minerals liberating additional heavy metals into the environment (Low et al., 2016; Ogola et al., 2002; Sáinz et al., 2004). However, when the metal ores are under carbonate rocks basement such as dolomite, limestone, and limonite, the acidity is neutralized and the resulting solution is basic or neutral pH. Other metal ions are also discharged due to weathering and decomposition of the ores as shown in the equations 2.11 - 2.15 (Dold, 2005; Low et al., 2016; Nuttall & Younger, 2002; Paikaray, 2015; Rudall & Jarvis, 2011; Wolkersdorfer, 2008; Woo & Choi, 2001).

$$4FeAsS_{(s)} + 13O_2 + 6H_2O \rightarrow 4Fe^{2+} + 4SO_4^{2-} + 4AsSO_4^{3-} + 12H^+ \quad \textbf{(2.11)}$$

$$2FeS_{2(s)} + 7O_2 + 2H_2O \rightarrow 2Fe^{2+} + 4SO_4^{2-} + 4H^+ \quad \textbf{(2.12)}$$

$$CaCO_{3(s)} + H^+ \rightarrow Ca^{2+} + HCO_3^- \quad \textbf{(2.13)}$$

$$Mg_2SiO_{4(s)} + 4H^+ \rightarrow 2Mg^{2+} + H_4SiO_4 \quad \textbf{(2.14)}$$

$$2NaAlSi_3O_{8(s)}+2H_2CO_3+9H_2O \rightarrow 2Na^++2HCO_3^-+4H_4SiO_4+Al_2Si_2O_5(OH)_4 \quad \textbf{(2.15)}$$

The solubility of heavy metals is suppressed greatly by the alkaline pH of the water; therefore, toxic metals might not be present in high concentrations in mine water from a region with limestone basement (Albu et al., 2012; Jusop et al., 1986). Limestone inhibits the oxidation of sulphide ores subsequently preventing acid production (Tiwary, 2001). At high pH, the water quality in ex-mining ponds is also degraded due to degradation of contaminants (McCullough & Schultze, 2015). Most studies in different regions reported pH of ex-mining water within the acidic range rather than the alkaline resulting in high concentrations of metal discharges (Ashraf et al., 2011b; Banks et al., 1997; Kusin et al., 2016; Madzin et al., 2015; Ogola et al., 2002; Räisänen et al., 2005). Acidic ex-mining water therefore contains high concentrations of toxic metals and metalloids such as arsenic due to variations in the mineral ores (Johnson et al., 2000; Lindsay et al., 2015). This confirms that geology of the areas under study directly influences the pH of ex-mining water and metal concentrations; hence pH is a good choice as one of the parameters in understanding the variations of metal concentrations and for the development of water quality index in this study. It was further emphasized the influence of pH on other water quality parameters, electrical conductivity and salinity (Yaacob et al., 2009). These parameters increase while dissolved oxygen decrease at low pH values. Similarly, pH was included in many existing water quality indices starting from the Hortons pioneer WQI (Boyacioglu, 2007; CCME, 2001; Cude, 2001; Horton, 1965).

2.5.2 Dissolved Oxygen

Oxygen is the most essential parameter in water for consumption and the survival of aquatic organisms. It indicates the levels of oxygen that is dissolved in water from an atmospheric source or waste product of photosynthesis. The level of dissolved oxygen in natural water varies greatly and is a function of temperature, atmospheric pressure, and dissolved substances. The acceptable level of dissolved oxygen that will support aquatic organisms is 4 to 5 mg/L, and below 3 mg/L the level is critical and results in death of many organisms. The permissible limit of dissolved oxygen in Malaysian DWQSRW is 5 - 7 mg/L (MOH, 2004). The concentration of dissolved oxygen is influenced greatly by the organic enrichment of the water; hence the waste assimilative capacity of water is measured in reference to dissolved oxygen level (Srinivas & Nageswara, 2010). The organic matter associated with mining operation are due to a wide variety of anthropogenic activities, resulting in low dissolved oxygen, high biological oxygen demand and chemical oxygen demand (Sfikas et al., 2013; Swer & Singh, 2004). Low dissolved oxygen in ex-mining water was reported in a research findings in Malaysia (Hatar et al., 2013; Takaijudin et al., 2012a; Yap et al., 2007), similar to the results of low dissolved oxygen obtained in other countries due to organic enrichment and silting by sand particles (Mishra et al., 2008; Saviour, 2012; Swer & Singh, 2004; Zakir et al., 2013).

Dissolved oxygen is given the highest priority in the existing Malaysian river WQI due to its perceived importance. In this study, dissolved oxygen is selected in the formulation of the index. Other existing indices, especially for drinking purposes also attached a priority to the dissolved oxygen concentration in the water.

2.5.3 Biological Oxygen Demand

The amount of dissolved oxygen in water consumed by anaerobic bacteria in the decomposition of organic matter is important in rating quality status of the water. The depletion of dissolved oxygen level results in stress on aquatic organisms, mostly due to influx of organic matter. The acceptable levels of biological oxygen demand is 6 mg/L and 3 mg/L for Malaysian DWQSRW and INWQS respectively (DOE, 2012; MOH, 2004). The continuous increase in human activities could raise the biological oxygen demand of water bodies. Large quantity of water discharged to facilitate mining operation contains high quantity of particulate matter, oil and grease which contaminate the surface water (Fardushe et al., 2016; Tiwary, 2001; Tiwary & Dhar, 1994). Many anthropogenic activities in the course of mineral exploration are associated with different types of organic matter pollution. High biological oxygen demand was reported in Malaysian ex-mining water (Takaijudin et al., 2012a), and similar results were also observed in Ghana and India (Acheampong et al., 2013; Mishra et al., 2008).

Biological oxygen demand has the second priority after dissolved oxygen in the existing Malaysian WQI, signifying the level of pollution due to organic matter in the Malaysian rivers. This justifies the consideration of biological oxygen demand, especially in Klang valley with much anthropogenic input into ex-mining ponds.

2.5.4 Ammoniacal Nitrogen

Presence of ammoniacal nitrogen (AN) in water signifies pollution due to leaching of waste product such as sewage. It indicates the presence of ammonia that could be toxic to humans, which consequently makes AN a major parameter in determining water quality of lakes, rivers and reservoirs. The major source of pollution due to ammoniacal nitrogen is domestic waste from residential units, especially in areas with high human habitation or farms. The INWQS permissible level of ammoniacal nitrogen in Malaysia is 0.3 mg/L (DOE, 2012). Many Malaysian ex-mining ponds are either close to farms or residential units, therefore, likely pollution of the ex-mining water by toxic ammonia could be experienced. High concentration of ammoniacal nitrogen beyond INWQS was reported in Kelana Jaya ex-mining pond (Yap et al., 2007). Toxicity of ammonia to humans had been emphasized in many research findings, where consideration had been given to the sources of the pollutants (Romano & Zeng, 2013; Zhou & Boyd, 2015). Ammoniacal nitrogen is considered in the existing Malaysian DOE-WQI, and is also considered in this study.

2.5.5 Total Suspended Solid

Suspended particles that can be trapped on a filter are also considered as a key factor in water quality assessment. It is a visual indicator of water quality which has a direct or indirect relationship with other water quality parameters such as dissolved oxygen and biochemical oxygen demand. The major natural sources of total suspended solid in water are soil erosion and decaying plants and animals and among the main anthropogenic sources is mining operation (Acheampong et al., 2013; Tiwary, 2001). The INWQS permissible limit of total suspended solid in Malaysia is 50 mg/L (DOE, 2012). Ex-mining water in Bestari Jaya was analyzed to contain total suspended solid above INWQS (Kusin et al., 2016). The effects of suspended solid has been fully explained (Bilotta and Brazier, 2008), which include increase in surface water temperature, low dissolved oxygen and limiting or blocking of light rays for the aquatic plants which are the source of food and oxygen in the aquatic ecosystem.

Suspended solid is included in the Malaysian DOE-WQI and its importance or priority is at par with chemical oxygen demand and higher than pH. This could be due to the high suspended solids in Malaysian rivers (Al-Mamun & Zainuddin, 2013; Amneera et al., 2013; Yen & Rohasliney, 2013). In this study, total suspended solid is not included in the WQI considering that very low suspended solid below detection limit is recorded in most of the ex-mining ponds and lakes examined.

CHAPTER 3: MATERIALS AND METHODS

3.1 Instrumentation

The instruments used in this study were calibrated and tested to ensure accurate and reliable results. ICP-MS, YSI multi probe, Modern water meter, and Spectrophotometer (DR 300) were successfully utilized for the quantitative analysis of metals and the selected water quality parameters.

3.1.1 Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

ICP-MS is widely used in the assessment of metals in water and other environmental samples (Bosch et al., 2016; Griboff et al., 2017; Sastre et al., 2002; Yang et al., 2016b). ICP-MS received a wider acceptance with about 8000 ICP-MS instrument installed globally (Thomas, 2008). Its advantages include high detection power, precision, sensitivity, and low volume of sample consumption (Ammann, 2007; Hauser, 2016; Poirier et al., 2016).

The metals determination in water samples from lakes and ex-mining ponds in the Klang Valley and Melaka were performed using ICP-MS 7500ce (Agilent Scientific Technology Ltd., USA) as shown in Figure 3.1. The sample injection system is made up of a nebulizer and temperature controlled spray chamber attached to an auto-sampler. In order to maintain the sensitivity of the instrument, the operating conditions listed in Table 3.1 were tuned on a daily basis with a tuning mixture which consists of 10 µg/L Ce, Co, Tl, Li and Y in 0.5% HCl and 2% HNO_3 (Low et al., 2012).

Table 3.1: Instrumental and operating conditions for ICP-MS 7500ce

Parameters	Conditions
Power (Watt)	1550
Plasma gas flow rate (L/min)	15
Auxiliary gas flow rate (L/min)	0.75
Sample depth (mm)	6-8
Carrier gas flow (L/min)	0.8-1.3
Sampler and skimmer cone	Ni
He or H_2 gas flow (L/min)	3-5

Figure 3.1: Inductively Coupled Plasma Mass Spectrometer (ICP-MS) (Agilent7500ce)

3.1.2 YSI Multiparameter Probe

For the in situ measurement of water quality parameters, the YSI multi probe as shown in Figure 3.2 was used. It consists of several sensors and probes assembled into single equipment. The portable multiprobe provides flexible measurements of many physical and chemical parameters simultaneously such as pH, DO, BOD, TDS, EC and

AN. The use of several optical and electrochemical sensors in YSI multiprobe gives it a wider acceptance for the field surface water assessment. It gives and excellent information by providing an instantaneous data which can be used at intervals to monitor the temporal variability of the surface water quality.

Figure 3.2: YSI Pro Multiparameter Meter (Professional Series, Yellow Springs, USA)

3.1.3 Suspended Solid Meter

For the measurement of solid particles in water samples, suspended solid meter (DR 900 colorimeter, HACH USA) as shown in Figure 3.3 was used. It allows for fast and easy monitoring of surface water quality. It uses sensors that operate in the infrared region which ensures long sensor life and reduces the effect of change in sample color. The sensors uses 'backscatter' that allow higher levels solid measurement.

Figure 3.3: DR 900 Colorimeter (HACH, USA)

3.1.4 Modern Water BOD Meter

Modern portable water meter (Shanghai, s/no 005) was used for in-situ BOD measurement of water samples as shown in Figure 3.4. Its operating principle is based on the excitation and fluorescence of tryptophan and similar compounds in a UV visible wavelength band. These compounds are essential amino acids, which are associated with microbial activity in waste water or sewage, and are found to correlate with bacterial contamination and BOD.

3.1.5 Water Sampler

Water sampler (Figure 3.5) was used for sampling in ex-mining ponds and lakes. It is suitable for surface water sampling due to easy control of the seals and drain valves for sample removal. It retrieves large volume of water samples at the required depth and floating sediment.

Figure 3.4: Modern Water BOD Check (Ansac Tech, Singapore)

Figure 3.5: Water Sampler (Wildco, USA)

3.2 Reagents and standard materials

The reagents used in this study were all analytical grade. All the plastic-wares used were soaked in 15% HNO_3 (v/v), and rinsed twice with ultrapure water (UPW) produced from the PURELAB® UHQ II system (ELGA®, UK). In the preparation of blanks and standard solutions, UPW was also used.

3.2.1 Calibration standard

The multi elemental calibration stock solution of 1000 mg/L for Fe, Na, Ca and Mg and 10 mg/L for As, Cd, Pb, and Mn (Agilent Technologies, Newcastle) was used to prepare the calibration and quality control (QC) solutions by appropriate dilution.

3.2.2 Certified reference materials

The standard reference materials for ICP Certipur® (Merck, Germany), SLR-4 riverine water reference material for trace metals (National Research Council of Canada), and National Institute of Science and Technology standard reference materials for trace elements in water (NIST 1643f, USA) were used to validate and verify the ICP-MS procedure.

3.3 Study area

Tin mining activity occurred predominantly in the western part of Peninsular Malaysia with ex-mining land covering about 113,700 ha (Takaijudin et al., 2012a). The study areas include Klang Valley in Selangor state in central Malaysia, and Melaka state in southern Malaysia. These areas were selected based on their relevance in mining operation, and reports on water scarcity in the periodic water crisis (Azzlan et al., 2016; Kusin et al., 2016). The water crisis in Klang Valley in February to August 2014 was due to the continuous rise in demand for municipal, industrial and domestic uses as a result of rapid population increase. Melaka experienced the worst water crisis in 1991 due to a very low level of water from its major water source, the Durian Tunggal Dam

(TDT) and Jus Dam. At the time of sampling for this study, the water levels in Durian Tunggal and Jus Dams were very low, indicating the urgent need to supplement with other possible sources. In the quest for alternative sources of water in the study areas, safety and suitability must be taken into consideration. This is achieved by detailed physical and chemical analysis and assessments of the relevant parameters based on the established water quality guidelines.

3.3.1 Klang Valley

The Klang Valley (2.6817°N, 101.6613°E) is an area in central Selangor, centered in Kuala Lumpur, and its connecting cities as shown in Figure 3.6. Demarcated between the Mountains of Titiwangsa to the north and east, and to the west by the Strait of Malacca, Klang Valley is the central industrial and commercial region of Malaysia. It has a population of about 7.5 million people (about a quarter of the Malaysian total population). The climate of the Klang Valley is tropical rainforest that is warm with sunshine. There is abundant rainfall, especially between October to March, with an average annual rainfall of 2000 – 3000 mm. The highest temperature in the study area is between 29 °C to 32 °C, with average humidity of 65 % to 70 %, except in June, July and September which are usually considered as dry season with low rainfall (Alatas, 2011; Althuwaynee et al., 2012).

The geology of the Klang Valley varies with different rock types, but mostly dominated by the Kenny hill formation, especially in Puchong area with most of the ex-mining ponds, and there is also deposit of limestone basement predominantly around Kuala Lumpur (Morgan, 1968). Selangor state was one of the largest producers of tin in Malaysia, producing up to 22% of the total Malaysia's tin output. There are abundant ex-mining ponds in Klang Valley due to the intensive mining activity with about 4909.6 hectares of ex-mining land. The ex-mining ponds and lakes are either abandoned, used for recreational activity, as a flood retention or to receive domestic effluents in the residential areas as indicated in Table 3.2 (Takaijudin et al., 2012a).

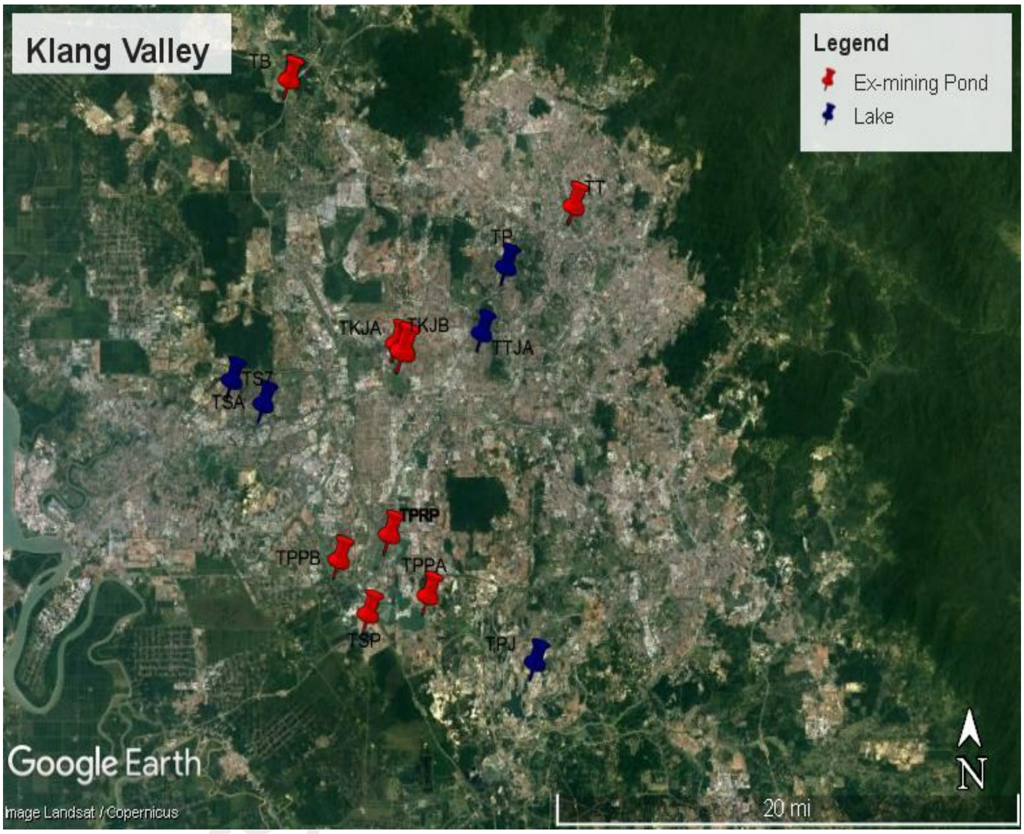

Figure 3.6: Map of the sampling locations in Klang Valley, Malaysia

Table 3.2: Sampling Sites in Klang Valley

Type	Area	Sites	Code	Location	Uses
Ex-mining Ponds	Kelana Jaya	Tasik Kelana Jaya	TKJA	N 03° 05' 35.4" E 101° 35' 53.2"	Recreational, flood retention, domestic effluent.
			TKJB	N 03° 05' 57.1" E 101° 35' 40.9"	
	Gombak	Tasik Biru	TB	N 03° 14' 50.9" E 101° 31' 38.6"	Recreational
	Puchong	Tasik Saujana Putra	TSP	N 02° 56' 50.4" E 101° 34' 37.2"	Recreational
		Tasik Putra Perdana	TPPA	N 02° 57' 27.8" E 101° 36' 52.2"	Recreational
			TPPB	N 02° 57' 46.4" E 101° 36' 21.8"	
		Tasik Prima Perdana	TPRP	N 02° 59' 10.0" E 101° 35' 49.4"	Recreational, water-sports
	Titiwangsa	Tasik Titiwangsa	TT	N 03° 10' 35.3" E 101° 42' 21.3"	Recreational, water-sports
Lakes	Petaling Jaya	Tasik Taman Jaya	TTJA	N 03° 06' 17.8" E 101° 38' 53.0"	Recreational, residential waste
	Kuala Lumpur	Tasik Perdana	TP	N 03° 08' 31.9" E 101° 41' 06.5"	Recreational
	Shah Alam	Tasik Shah Alam	TSA	N 03° 04' 27.0" E 101° 30' 47.3"	Recreational
		Tasik Shah Alam 7	TS7	N 03° 04' 42.4" E 101° 29' 28.7"	Recreational, domestic effluent
	Putrajaya	Tasik Putrajaya	TPJ	N 02° 55' 12.5" E 101° 40' 52.6"	Recreational, water-sports

3.3.2 Melaka

Melaka (2°12′N, 102°15′E) is a state located in the southwestern part of Peninsular Malaysia bordered by Johor to the south, and Negeri Sembilan to the north and west. It is a historic state having a population of 913,210 with agricultural activities as the major land use. Melaka is hot and humid with average temperatures ranging from 26 °C to

30°C and average annual rainfall of 1430 to 2152 mm. It has limited water resources due to its small size and low rainfall compared to other states (Shirazi et al., 2013).

The geology of Melaka is dominated by phyllite, but in the Jasin district up to Johor the geological basement is limestone, schist sand and alluvium (Shirazi et al., 2015). The ex-mining area in Melaka covers an area of 367 hectares, the ninth largest in peninsular Malaysia. Most of the ex-mining land (356 ha) are located in Jasin where TBC1, TBC2 and TBC3 are located (Figure 3.7). TR a large ex-mining pond in Negeri Sembilan state near Melaka was included in this study for comparison with Melaka to ascertain the geological influence on metal levels. The ex-mining ponds are used for recreational and agricultural activities; the lakes are mostly utilized for water supply and agricultural purposes (Table 3.3).

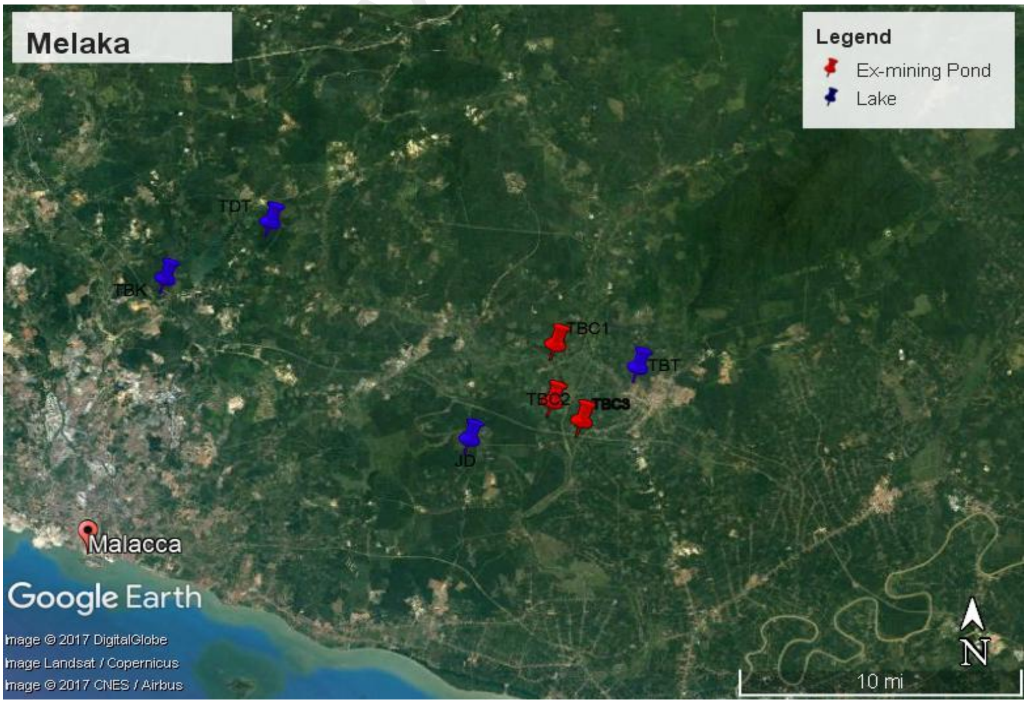

Figure 3.7: Map of the sampling locations in Melaka, Malaysia

Table 3.3: Sampling Sites in Melaka

Type	Area	Sites	Code	Location	Uses
Ex-mining Ponds	Jasin	Tasik Biru Chin Chin	TBC1	N 02°16'13.8" E 102°29'17.0"	Recreational
	Jasin	Tasik Biru Chin Chin	TBC2	N 02°16'15.8" E 102°29'37.9"	Recreational
	Jasin	Tasik Biru Chin Chin	TBC3	N 02°16'27.3" E 102°29'34.6"	Recreational
	Negeri Sembilan	Tasik Rantau	TR	N 02°31'00.3" E 101°57'47.7"	Agriculture
Lakes	Jasin	Jus Dam	JD	N 02°26'38.3" E 102°24'00.2"	Water Supply
	Alor Gajah	Tasik Durian Tunggal	TDT	N 02°21'11.4" E 102°17'55.6"	Water Supply, Windsurfing
	Jasin	Tasik Tebat	TBT	N 02°16'38.0" E 102°29'05.5"	Fishing
	Bukit Katil	Tasik Bukit Katil	TBK	N 02°16'30.94" E 102°17'08.30"	Agriculture

3.4 Water Sampling and Analysis

Water samples were collected from ex-mining ponds and lakes that had no mining activity in the Klang Valley. Sampling was carried out from the 13 sites in the month of September 2015, which is the end of a period considered to have low levels of rainfall. The second phase of the sampling was carried out on 6th October 2016 from 7 ex-mining ponds and lakes in Melaka, and 1 ex-mining pond in Negeri Sembilan. The samples were collected from a 25 cm depth using a Wildco water sampler with nine samples per sites for analysis (MacDougall & Crummett, 1980). Conc. HNO_3 (Merck suprapur®) was used to acidify all the water samples to lower the pH < 2 to prevent metals sedimentation in the container (Sadeghi et al., 2012), then stored in acid washed polyethylene bottles and transported to the laboratory in ice boxes conserved at 4°C for metals analysis. Other parameters such as dissolved oxygen (DO), pH, electrical conductivity (EC), total dissolved solid (TDS), and ammoniacal nitrogen (AN) were measured and recorded in the field using portable YSI Pro multi parameter water quality meter (professional series), and the total suspended solid (TSS) was measured using the DR 900 suspended solid meter. The biological oxygen demand (BOD) was also measured in-situ using a portable water meter to avoid changes in the bacterial concentration with time. The triplicates of each water sample were filtered using 0.45μm PTFE filters before metal analysis using inductively coupled plasma-mass spectrophotometry (ICP-MS) were carried out. The filtered water samples were analysed for As, Cd, Pb, Mn, Fe, Na, Mg, and Ca with ICP-MS 7500ce (Agilent Scientific Technology Ltd., USA). Blank and QC samples were checked after every ten samples to demonstrate the validity of the previous runs. All analyses were carried out in triplicates and the results were expressed as 95% confidence interval of the mean in μg/L.

3.5 Data analysis

Descriptive statistics and one way ANOVA were carried out using MS Excel 2013 to analyze the differences among sampling locations, and the results were reported at $p<0.05$ levels. The non-detected concentrations were reported as half detection limit of the metals (Farnham et al., 2002). Multivariate statistical analyses were carried out using JMP Pro 12. PCA, FA, LDA and HCA analyze the variations in concentrations of the metals and physical chemical parameters among the sampling sites.

Risk assessment of exposure through ingestion and dermal routes was carried out using MS Excel 2013. Estimation of the metal concentrations was carried out using Monte Carlo algorithm by appropriate selection from its particular distribution. In view of the differences in physiological and behavioral differences, this study focuses on estimating the exposure and risks of adult and children separately. The water quality index formulation was also carried out using MS Excel 2013. The results of the multivariate and risk analysis, and WQI were then thoroughly analyzed and discussed.

CHAPTER 4: RESULTS AND DISCUSSION

This chapter explains the variation of some physical and chemical parameters in ex-mining ponds and the lakes in the studied areas. The analysis of health risks and exposure assessments, and water quality index formulation are also discussed.

4.1 ICP-MS Method verification

The coefficients of determination (R^2) for ICP-MS calibration curves were all close to 1.0. The measurement results as shown in Table 4.1 demonstrate a good agreement with the certified values with recovery values ranging between 88% and 107%. This is comparable with the analytical performance reported in a similar study (Assubaie, 2015).

4.2 Analysis of Metal Concentrations and Physical-Chemical Parameters

4.2.1 Klang Valley

The results of the 15 water quality variables are presented, and references made to the Malaysian water quality standards in Table 4.2. The means and standard deviations of the studied parameters are presented in Tables 4.3 and 4.4. Among the parameters, heavy metals (Table 4.3) are of much concern in water researches due to their ability to deteriorate the aquatic ecosystem and bio-accumulate to toxic levels, rendering the water unsuitable for human consumption, irrigation, and recreational activities (Zhang et al., 2009). The concentration of the metals vary significantly (p<0.05) with As, 0.50-118.36 μg/L; Cd, 0.10-13.18 μg/L; Pb, <0.05-13.77 μg/L; Mn, 1.02-440.71 μg/L; Fe, 15-1185 μg/L; Na, 3810-17495 μg/L; Mg, 710-4949 μg/L; and Ca, 12084-44944 μg/L. Such variation could originate from the past mining activity, historical background of the ex-mining ponds and additional variations by the on-going anthropogenic inputs. The ore commonly mined in Selangor was tin from the mineral cassiterite as tin oxide (SnO_2). The metals associated with tin-ore are As, Cd, Pb, and Zn, and are usually released to the environment by dissolution and erosion (Yusof et al., 2001). As, Cd, Pb, Mn, Fe and Ni were found in mining areas and related to the ores and rocks crushed during the mining process (Ashraf et al., 2015). However, most variations in the heavy metal concentrations are usually due to the percentage natural abundance, natural precipitation and aeration and dilution factor due to water from external sources (Ashraf et al., 2010). As is one of the heavy metals of concern and it is a threat to the environment due to its toxicity.

Cd is naturally found in small quantities in water, soil, and air (Suthar et al., 2009), and it is a toxic element not required by plants. Based on the Interim National Water Quality Standards for Malaysia (INWQS), the mean concentrations of both elements in all the samples are below the benchmark level for Class IIA water supplies (which require conventional treatment before use) (DOE, 2012). When referring to the Drinking Water Quality Standard for Raw Water (DWQSRW), As and Cd concentrations in TPPA, TPPB, and TPRP, As concentration in TS7 and Cd concentrations in TPJ and TT are higher than the recommended values of 10 and 3 μg/L respectively (MOH, 2004). As shown in Figure 4.1, high As and Cd are detected in TPPA, TPPB and TPRP located in Puchong, which is an area that had active and extensive mining activities (Balamurugan, 1991). Similarly, a high concentration of As (66 mg/L) had been reported in the Bestari Jaya ex-mining ponds (Ashraf et al., 2011b), and traces of Cd (0.04 mg/L) in the Kelana Jaya ex-mining ponds. These results suggest that relatively high concentrations of As and Cd in the ex-mining ponds are plausibly related to the ex-mining activities (Navarro et al., 2008). Mining operation results in the elevated concentrations of As, Cd and Mn in the surface water above WHO drinking water standard values. The dissolved As and Cd were derived from the ore mineral phase controlled by carbonate minerals and amorphous iron (Woo & Choi, 2001).

Table 4.1: ICP-MS Measurements on Certified Reference Material

Isotope of Analyte		**^{75}As**	**^{111}Cd**	**^{208}Pb**	**^{55}Mn**	**^{56}Fe**	**^{23}Na**	**^{24}Mg**	**^{43}Ca**
Analysis Mode		He	Normal	Normal	He	H_2	Normal	Normal	Normal
Detection Limit (µg/L)		0.1	0.004	0.05	0.03	2	1.27	0.3	1.18
Certipur® Reference Material for ICP	**Certified (µg/L)**	50 ± 5	20 ± 5	25 ± 5	30 ± 5	100± 10	8000±500	15000±500	35000±1000
	Measured (µg/L)	46 ± 1	19 ± 1	27 ± 2	29 ± 1	105 ± 5	8300±900	15500±500	36000±1000
	Recovery (%)	92 ± 1	97 ± 4	106 ± 17	98 ± 8	107 ± 12	102 ± 4	101 ± 1	104 ± 7
SLRS-4	**Certified (µg/L)**	0.68 ± 0.06	0.012±0.002	0.086±0.007	3.37±0.18	103 ± 5	2.4 ± 0.2	1.6 ± 0.1	6.2 ± 0.2
	Measured (µg/L)	0.59±0.011	0.011±0.001	0.081±0.003	3.36 ± 0.03	108 ± 3	2.5 ± 0.2	1.5 ± 0.2	6.7 ± 0.3
	Recovery (%)	88 ± 5	98 ± 9	94 ± 4	99 ± 3	105 ± 6	101 ± 8	97 ± 7	106 ± 5
NIST® Trace Metal	**Certified (µg/L)**	57.42±0.37	5.89 ±0.13	18.48±0.081	37.14±0.58	93.44±0.77	18830±240	7454 ±58	29430±320
	Measured (µg/L)	56.55±1.46	6.04 ±0.60	16.71 ±1.50	39.23±0.30	100.89±1.10	-	6773.5±77.00	27809.07±205.00
	Recovery (%)	98 ± 5	102 ± 9	90 ± 9	105 ± 1	107 ± 2	-	91 ± 11	94 ± 3

Mean ± 95% confidence limit of three replicates

Conversely, TS7 (a non-ex-mining lake) was also found to have a high As concentration. This is possibly related to the urbanization and dense industrialization in Shah Alam area (Abdullah et al., 2011; Khailani & Perera, 2013), where the sources of As include industrial burning of fossil fuel, agrochemical and steel and metal coating industries (Balamurugan, 1991; Hamzah et al., 2011). Similarly, it appears that the source of high Cd concentrations in TPJ could be due to the frequent national and international water sports competitions, transport and other recreational activities.

Pb accounts for 13 mg/kg of the earth's crust (Ashraf et al., 2015). In this study, Pb concentrations found in all cases are below the DWQSRW requirement of 50 μg/L; and are even below the method detection limit (0.05 μg/L) for TB and TSA. Pb concentrations in the range of 19 to 75 μg/L had been reported in the ex-mining ponds of Perak (Orji et al., 2014).

The variations in physical-chemical parameters in ex-mining ponds and lakes are presented in Figure 4.1. Ex-mining ponds can be observed to be acidic or alkaline depending on the mineralogy. The measure of acidity or alkalinity of water samples can be numerically expressed with pH, which at lower values become corrosive, and at high values have bad taste and harmful to the eyes and skin (Srinivas & Nageswara, 2010). The useful nature of any water source is within the pH range of 6.0 and 8.5 (Garg et al., 2010). Table 4.3 reveals that most of the ex-mining ponds are acidic. TKJA, TKJB, TB, and TSP are below the DWQSRW and INWQS Class IIA minimum permissible values, and may not be suitable for domestic uses; whereas TPPB had a pH value of 10.0, which is higher than the maximum limit of 9. This could be attributed to the variation in tin ore type. Two distinct cassiterite were reported in Puchong area of the Klang Valley with different dissolutions of the rock formations which might have contributed to the dissimilarity in the levels of pH among other sites (Teh & KIang, 2002).

Table 4.2: Malaysian Water Quality Standards (DOE, 2012; MOH, 2004)

Standards	As	Cd	Pb	Mn	Fe	Na	Mg	Ca	DO	BOD	TDS	pH	TSS	AN	EC
DWQSRW	10	3	50	200	1000	200000	150000	-	-	6	1500	5.5-9.0	-	-	-
INWQS-Class IIA	50	10	-	100	1000	-	-	-	5-7	3	100	6-9	50	0.3	1000

Metals (µg/L), EC (µs/cm), pH (No unit), other parameters (mg/L)

Table 4.3: Concentrations of metals and water quality parameters in ex-mining ponds Klang Valley

Variable	TKJA	TKJB	TB	TSP	TPPA	TPPB	TPRP	TT
As	7.5 ± 0.2	6.8 ± 0.3	0.53 ± 0.02	0.67 ± 0.03	11.3 ± 0.4	116 ± 2	42.0 ± 0.5	2.13 ± 0.05
Cd	0.03 ± 0.01	0.09 ± 0.01	0.01 ± 0.01	1.14 ± 0.02	12.9 ± 0.2	12.23 ± 0.02	12.19 ± 0.01	12.15 ± 0.01
Pb	0.23 ± 0.03	0.79 ± 0.06	< 0.05	13.5 ± 0.1	4.5 ± 0.3	3.6 ± 0.2	3.7 ± 0.3	3.51 ± 0.07
Mn	124 ± 3	36.4 ± 0.8	18.0 ± 0.6	438 ± 2	125 ± 2	1.4 ± 0.3	3.3 ± 0.1	11.4 ± 0.2
Fe	1166 ± 11	241 ± 1	240 ±2	360 ± 5	460 ± 4	35.8 ± 0.6	15.1 ± 0.1	34 ± 2
Na	8093 ± 69	17247 ± 199	4303 ± 34	3876 ± 43	14516 ± 153	12367 ± 205	10572 ± 68	1484 ± 7
Mg	1554 ± 20	1561 ± 27	718 ± 6	4903 ± 40	2219 ± 106	1853 ± 32	3863 ± 23	5628 ± 25
Ca	23513± 374	28545 ± 326	12500 ± 336	25428 ± 256	21277 ± 161	34246 ± 179	44346 ± 454	27632 ± 256
DO	0.91 ± 0.07	1.35 ± 0.07	4.2 ± 0.1	5.2 ± 0.2	6.1 ± 0.1	0.38 ± 0.03	0.47 ± 0.04	4.11 ± 0.09
BOD	5.9 ± 0.4	9.7 ± 0.1	2.5 ± 0.2	3.4±0.8	8 ± 1	2.8 ± 0.3	2.77 ± 0.07	1.5 ± 0.1
TDS	89 ± 1	125 ± 1	46 ± 1	172 ± 1	132 ± 1	161 ± 1	161 ± 1	113 ± 2
pH	0.93 ± 0.09	1.35 ± 0.06	4.2 ± 0.2	5.2 ± 0.1	6.1 ± 0.2	10.00 ± 0.09	8.23 ± 0.09	8.25 ± 0.05
TSS	4.6 ± 0.5	16 ± 1	< 5	< 5	6.7 ± 0.5	< 5	< 5	< 5
AN	0.16 ± 0.02	0.33 ± 0.01	0.06 ± 0.01	0.09 ± 0.01	0.25 ± 0.03	0.16 ± 0.01	0.19 ± 0.01	0.05 ± 0.01
EC	145 ± 2	203 ± 1	75 ± 1	286 ± 2	217 ± 2	276 ± 1	281 ± 1	189 ± 1

Metals (µg/L), EC (µs/cm), pH (No unit), other parameters (mg/L)

Lakes showing slightly basic pHs within the tolerable range are shown in Table 4.4. This is similar to the findings in the Bukit Merah reservoir and Lake Chini with mean pH values of 7.0 and 6.5 respectively (Akinbile et al., 2013; Shuhaimi-Othman et al., 2007). Basically, during the oxidation of metal ores such as FeS_2, H^+ is released which consequently lowers the pH of the mine water (Singer & Stumm, 1970; Yaacob et al., 2009). Meanwhile, carbonates increase the pH within the alkaline region (Johnson & Hallberg, 2003). In general, low pH is likely to precipitate metals in solution resulting in high concentrations and impairment of the water quality. Therefore, acidic water usually contains high concentration of metals and metalloids such as As due to the variation in the environment and mineral ores (Johnson et al., 2000; Salomons, 1995). These possibly explain the notable discrepancies in metal concentrations and pH values among the sites.

The depletion of DO level is one of the most recurrent effects of pollution in water bodies (Srivastava et al., 2009). The DO values for the studied lakes are mostly lower than 5 mg/L (INWQS Class IIA), except for TSP and TPPA. DO values of 3.32 mg/L, 4.26 mg/L and 4.17 mg/L had been previously reported in the ex-mining ponds of Kelana Jaya. This is a threat to aquatic lives as DO is essential for the metabolism of all aquatic organisms. This might be the reason for dead fishes noticed floating in the Kelana Jaya ex-mining pond (TKJA). The low DO in some of the ex-mining ponds could be associated with the low pH value, and acidic medium is generally found to lower the DO of water samples due to the decomposition of organic matter (Akinbile et al., 2013; Kannel et al., 2007; Shuhaimi-Othman et al., 2007). Presence of organic matter such as carbon based materials comprising of dead plants and animals, kitchen waste and rotten leaves deplete the oxygen level in water bodies due to microbial decomposition.

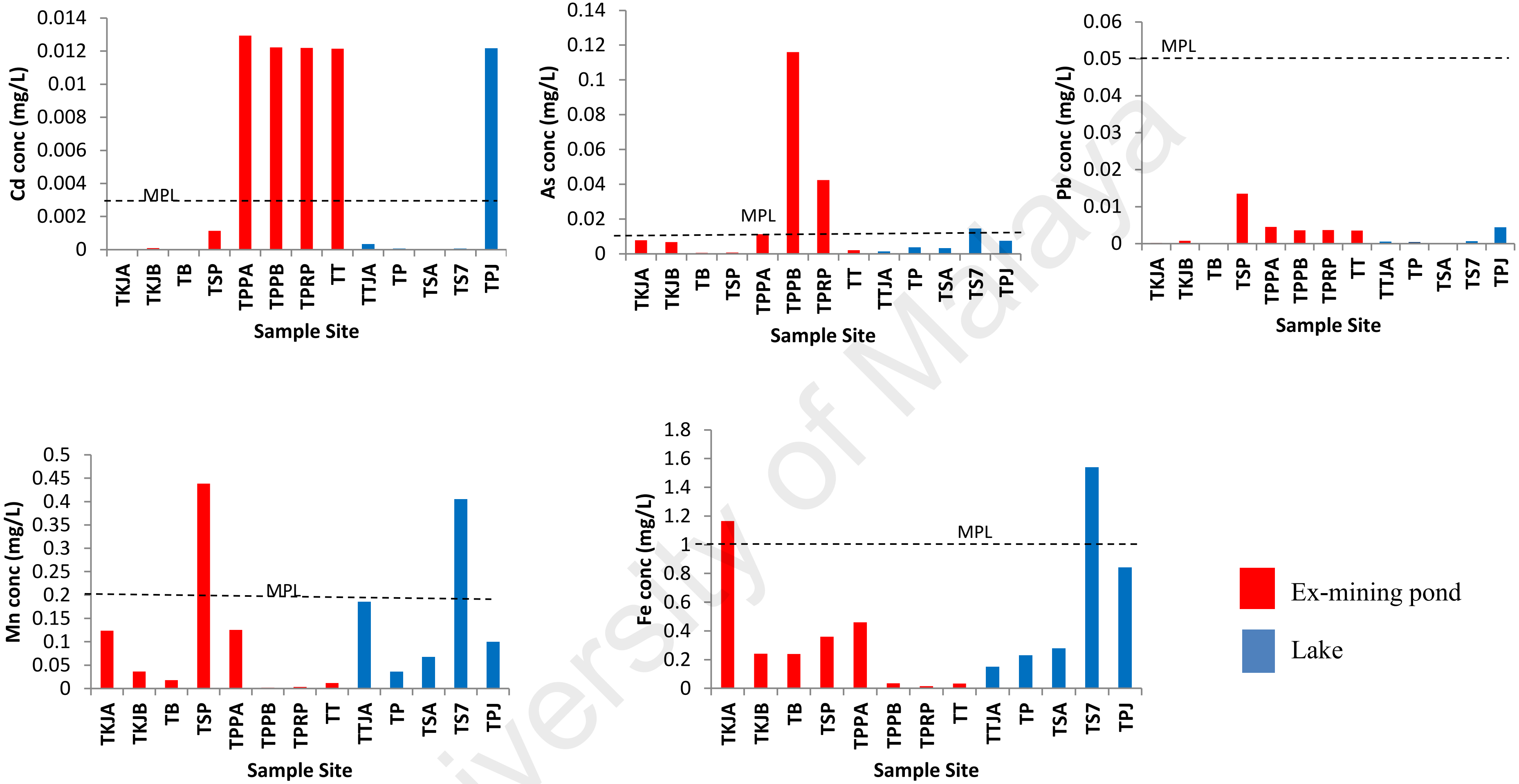

Figure 4.1: Heavy metal levels and physico chemical parameters in ex-mining ponds and lakes in Klang Valley
MPL = Maximum permissible limit, LPL = Lower permissible limit

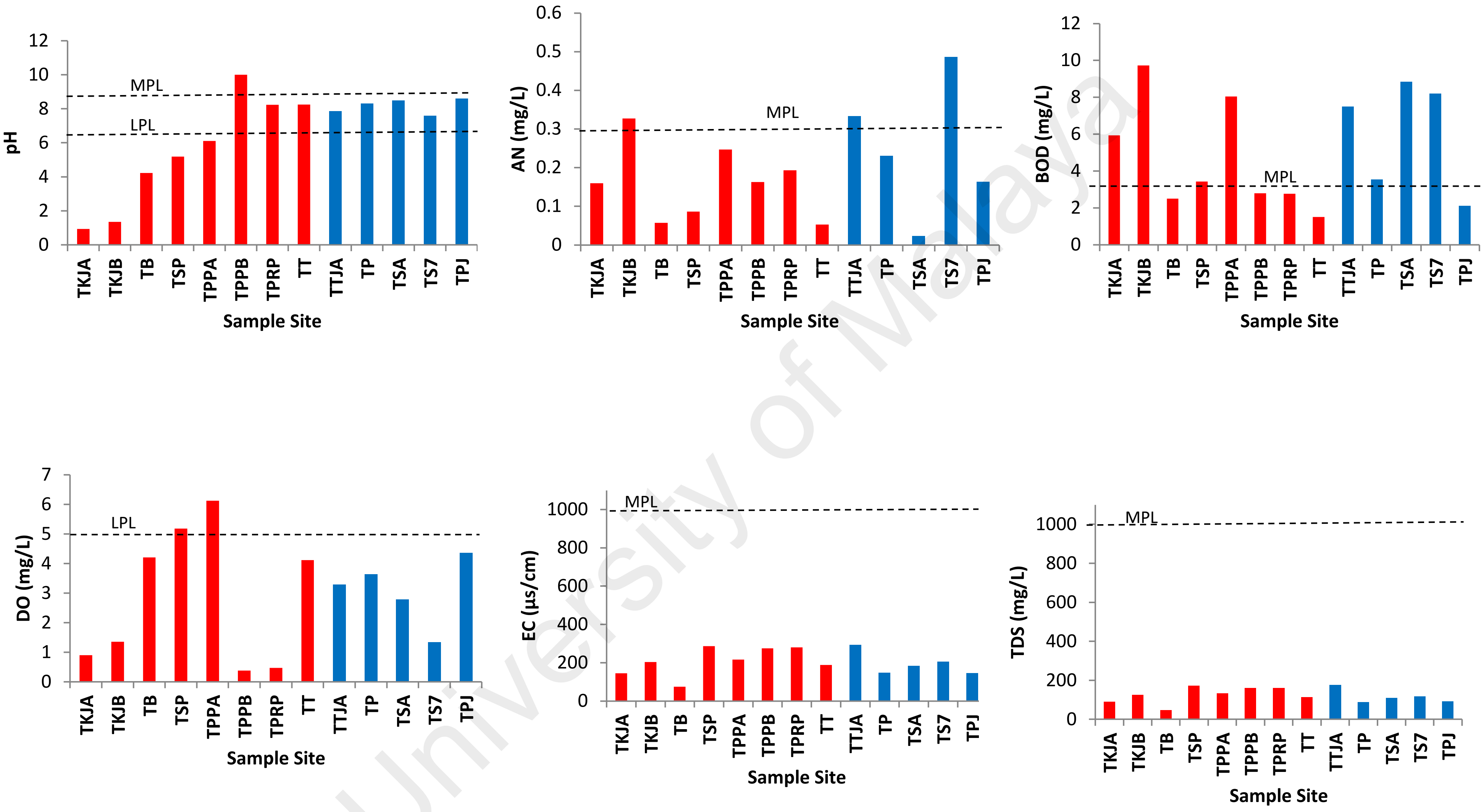

Figure 4.1: Continued

To a certain extent, the waste assimilative capacity of water can be measured with reference to the DO levels (Srinivas & Nageswara, 2010). The low DO is likely attributed to the decaying organic matter by bacteria from domestic wastes, as noticed that some of the lakes served as a reservoir for domestic effluents from residence areas. In this context, BOD is commonly used to assess the contamination of surface water due to domestic and industrial activities. BOD is the amount of oxygen needed by bacteria to decompose organic matter under aerobic conditions. The presence of organic compound in surface water enhances microbial growth which increases the concentration of BOD. In Tables 4.3 and 4.4, BOD values above 6 mg/L (DWQSRW) are observed in TKJA, TKJB, TPPA, TTJA, TSA, and TS7; where Tasik Kelana Jaya and Tasik Shah Alam receive domestic waste from the surrounding residential areas. The effect of domestic waste noticed on the water surface can easily be quantified by the amount of BOD, as most of the lakes directly received residential effluent.

The presence of AN in water bodies is also an indication of pollution; it is the level of toxic ammonia present as a result of leaching or disposal of sewage or manure. The levels of AN show a range of 0.02-0.48 mg/L and 0.05-0.32 mg/L for the ex-mining ponds and the lakes (Tables 4.3 and 4.4). The TKJB, TTJA, and TS7 lakes have AN concentrations greater than the INWQS Class IIA maximum limit of 0.3 mg/L. The high AN levels at these sites coincide with the low DO and high BOD values, indicating that the pollution is due to organic matter as a result of the flow of domestic effluent and recreational activities. In a similar study, high AN concentration was reported in the Kelana Jaya ex-mining pond as a result of high loading of organic matter from the sewage (Yap et al., 2007).

The anthropogenic input in surface water determine the quantity of the insoluble particulate matter mostly related to organic particles, surface runoff, sewage and waste water (Amneera et al., 2013; Chigor et al., 2012; Zhang et al., 2016). The levels of TSS in most of the ex-mining ponds studied in Klang Valley are generally low compared to the lakes, and all values are below the INWQS Class IIA limit of 50 mg/L. The polluted ex-mining TKJB pond records the highest TSS value of 16 ± 1 mg/L, while TSA is 13.7 ± 0.5 mg/L which could be attributed to the domestic sewage and waste water from the surrounding residences. In a similar study for comparison, the TSS in Chini lake and Bukit merah reservoir was found to be 8.72 and 1.75 mg/L respectively (Akinbile et al., 2013; Shuhaimi-Othman et al., 2007).

Table 4.4: Concentrations of metals and water quality parameters in lakes of Klang Valley

Variable	TTJA	TP	TSA	TS7	TPJ
As	1.13 ± 0.06	3.8 ± 0.1	3.37 ± 0.2	14.4 ± 0.4	7.5 ± 0.3
Cd	0.33 ± 0.02	0.06 ± 0.01	0.02 ± 0.01	0.05 ± 0.01	12.17 ± 0.01
Pb	0.52 ± 0.02	0.36 ± 0.01	<0.05	0.66 ± 0.02	4.4 ± 0.2
Mn	186 ± 1	35.7 ± 0.3	68 ± 2	405 ± 3	100 ± 1
Fe	151 ± 4	228 ± 6	279 ± 6	1540 ± 25	841 ± 7
Na	5269 ± 34	3320 ± 37	5540 ± 44	9870 ± 25	21520 ± 276
Mg	1829 ± 16	2234 ± 40	2716 ± 46	3043 ± 30	1650 ± 21
Ca	15280 ± 73	25194 ± 57	43423 ± 184	21256 ± 70	31245 ± 128
DO	3.3 ± 0.2	3.6 ± 0.1	2.8 ± 0.1	1.33 ± 0.05	4.4 ± 0.1
BOD	7.5 ± 0.2	3.54 ± 0.07	8.85 ± 0.08	8.2 ± 0.2	2.10 ± 0.09
TDS	176 ± 2	87 ± 2	109 ± 1	117 ± 2	90 ± 2
pH	7.9 ± 0.2	8.32 ± 0.02	8.49 ± 0.03	7.6 ± 0.1	8.5 ± 0.1
TSS	9.7 ± 0.5	6.7 ± 0.9	13.7 ± 0.5	< 5	< 5
AN	0.33 ± 0.02	0.23 ± 0.02	0.02 ± 0.01	0.49 ± 0.05	0.16 ± 0.01
EC	291 ± 1	147 ± 1	184 ± 1	205 ± 3	146 ± 2

Metals (µg/L), EC (µs/cm), pH (No unit), other parameters (mg/L)

4.2.1.1 Principal Component Analysis

PCA was carried out to show the distribution of parameters in the studied areas, in which their variability in water samples was analyzed. The PCs which are usually important (eigen values > 1) describe the largest variations in the entire data set up to 82% (Table 4.5). The first factorial plane (plot of PC1 against PC2) contained most of the information and therefore primarily considered (Alberto et al., 2001; Atanacković et al., 2013; Zhang et al., 2009). PC1 and PC2 respectively account for 26.6% and 18.3% of the total variation of water quality in Klang Valley.

The PC loadings indicate the directions and strength of the variations of the water quality parameters, whereas the scores summarize the relationships among the samples. The loadings of PC1 contain positively correlated parameters which comprises of As, Cd, Pb, Mg, Ca, TDS, pH and EC as the major contributors (Figure 4.2). This is indicative of heavy metal pollution which is likely related to the previous mining activities. PCA was applied to study metal pollutants in the abandoned mines, in which PC1 shows strong loading of parameters related to dissolution of sulfide minerals during mining (Atanacković et al., 2013). Similarly, PCA was applied in the evaluation of anthropogenic levels and mobility of rare earth metals in ex-tin mining area (Khan et al., 2016).

Besides that, the dominance of Ca over K and Na also suggest that weathering is also one of the main sources of the metals in the water samples (Subramani et al., 2010). By comparing the scores and loadings in Figure 4.2, it can be inferred that the ex-mining ponds TPRP, TPPB, and TSP are highly loaded with hazardous metals associated with mining activities. Again, this renders the water unfit for neither domestic nor recreational activities. PC2 has high positive loadings for Mn, BOD, TDS, AN, EC and high negative loading for DO; which are indicative of pollution as a result of domestic effluent discharges (Mustapha et al., 2013). Hence, the scores on PC2 show that TKJB, TTJA and TS7 are polluted mainly by domestic sewages. Emphases was given on the adverse health effects and environmental risk associated with domestic wastes (Babayemi et al., 2016). Generally, the PCA bi plot separates the PCs into two parts showing deterioration of water quality based on metals (PC1) and organic pollutants (PC2).

PC3 explain 17.50% of the total variation in the data set with positive loading on Mg, DO, Pb, Mn, and Fe which could be due to dissolution of rock minerals, and pollution from anthropogenic source (Baltrusaitis & Sviklas, 2016; Mustapha et al., 2013). The strong positive loading on Fe and Na, and negative loading on DO in PC4, and Cd, Pd, and DO in PC5 could be due to weathering and organic pollution, and the results of past mining operation respectively (Low et al., 2016).

The results of the PCA show a clear variation in levels of metal pollutant among the sites studied which confirm the sources to be anthropogenic, and traces its origin to previous mining operation.

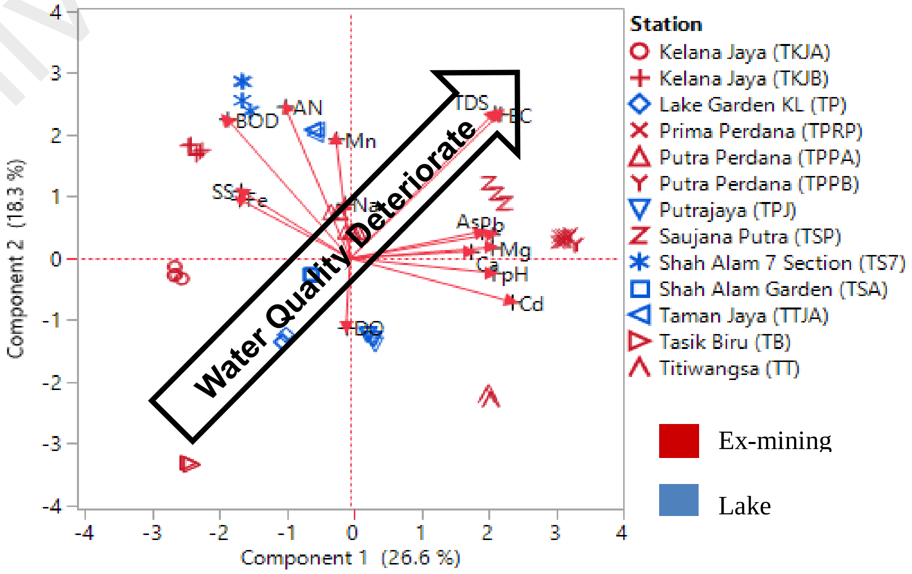

Figure 4.2: PCA bi-plot of metals and physical-chemical parameters in Klang Valley

Table 4.5: Percent explained variance for PCA in Klang Valley

Variable	PC1	PC2	PC3	PC4	PC5
As	0.38929	0.07819	-0.35845	0.19950	-0.18525
Cd	0.35642	-0.12528	-0.15755	0.24196	0.35651
Pb	0.31181	0.07053	0.36546	0.05857	0.27730
Mn	-0.03136	0.34763	0.45211	0.18719	-0.09242
Fe	-0.24585	0.17282	0.13013	0.50125	-0.12062
Na	-0.01319	0.15983	-0.29563	0.38187	0.56948
Mg	0.31175	0.03210	0.31227	-0.15482	-0.15875
Ca	0.26539	0.02141	-0.29655	-0.14963	-0.00556
DO	0.00933	-0.19949	0.40241	-0.07796	0.52490
BOD	-0.27196	0.40544	-0.08551	-0.21621	0.15714
TDS	0.31799	0.42266	0.03630	-0.16443	0.03778
AN	-0.14450	0.44190	-0.06342	0.25175	-0.05172
pH	0.31467	-0.04169	-0.07906	0.05183	-0.13053
SS	-0.24248	0.19755	-0.19401	-0.50183	0.26014
EC	0.33010	0.42025	0.02165	-0.14830	-0.00521
Eigen Value	3.98	2.74	2.62	1.75	1.19
Variance (%)	26.60	18.30	17.50	11.70	7.99
CV (%)	26.60	44.85	62.32	74.00	81.99

CV = Cumulative Variance

4.2.1.2 Factor Analysis

Factor analysis (FA) was applied to provide information on the most meaningful parameters in ex-mining ponds and lakes that describe the whole data set allowing data reduction with least loss of information (Zhou et al., 2007).

PCA generates PCs which are sometimes not easily interpreted; therefore it is desirable to rotate the PCs by varimax rotation. Varimax rotation was applied on PCs with eigen value > 1 considered as significant to obtain new group of variables called varimax factors (VF) (Juahir et al., 2011). In FA, the number of VFs produced due to varimax rotation is equal to the number of variables in accordance with common features including latent, hypothetical, and unobserved variables. The correlation of VF greater than 0.75 are considered strong, 0.75 – 5.0, 0.5 – 0.3, as moderate and weak significant factor respectively (Liu et al., 2003; Vega et al., 1998).

Table 4.6 reveals the selected VFs and corresponding variable loadings. Among the five VFs, 27% of the total variance is contributed by VF1 strongly associated with EC and TDS, and moderately associated with Pb, and Mg which are related to ions due to weathering and dissolution of rocks, and heavy metal pollution which may originate from previous mining operation (Modoi et al., 2014). VF2 explaining 18% of the total variance has strong positive loadings on BOD and SS, associated with organic pollutants and anthropogenic inputs. The ex-mining ponds and lakes in Klang Valley especially TTJA, TKJA, and TKJB are influenced by domestic input from residences which raise the levels of organic matter. Fe and Mn are the significant parameters in VF3 which accounts for 17% of the total variance, which may be due to natural background concentrations. Fe and Mn often occur naturally in strong association in water and sediment of lakes, and released to the environment due to weathering of Fe and Mn bearing minerals (Davison, 1993; Kacmaz & Burns, 2017). VF4 is a factor of pollution due to oxygen consuming substances; it explains about 12% of the total variance and has a strong negative loading on DO which can be explained in consideration to residence discharges into the lakes. VF5 explains 8% of the total variance and has strong loading on Na and weak loadings on As and Cd which may be associated with past mining activity. As and Cd are the major heavy metal pollutants in this study.

The results of the factor analysis generally reveal dominance of heavy metal and organic matter influences on water quality of ex-mining ponds and lakes which agrees with the results of PCA. This justifies that ex-mining water needs to be analyzed carefully on the intended use for human consumption.

Table 4.6: Loadings of parameters on significant VFs for ex-mining ponds and Lakes in Klang Valley

Variable	VF1	VF2	VF3	VF4	VF5
As	0.268993	-0.343671	-0.208155	-0.665130	0.333790
Cd	0.244413	-0.556063	-0.380194	0.000003	0.599397
Pb	0.642828	-0.414877	0.048290	0.502478	0.067237
Mn	0.440694	-0.004093	0.797292	0.278590	-0.201031
Fe	-0.259623	-0.018074	0.862054	-0.051156	0.129207
Na	-0.035187	0.109709	0.126701	-0.090781	0.947282
Mg	0.612959	-0.379833	-0.097813	0.199828	-0.380655
Ca	0.312625	-0.087223	-0.504316	-0.382739	0.176896
DO	-0.012509	-0.099504	-0.065255	-0.931608	0.007360
BOD	0.102741	0.869764	0.286224	0.072011	0.090225
TDS	0.954929	0.085152	-0.019837	-0.151513	0.074291
AN	0.167754	0.348355	0.630328	-0.303180	0.249564
pH	0.328221	-0.442459	-0.270910	-0.227620	0.051959
SS	-0.026113	0.949329	-0.236455	0.010621	0.016951
EC	0.955605	0.051150	-0.018975	-0.202444	0.064142
Eigen Value	3.9	2.7	2.6	1.7	1.1
Variation (%)	27.0	18.4	17.7	11.8	8.07
CV (%)	27.0	45.4	63.1	74.9	82.9

4.2.1.3 Hierarchical Cluster Analysis

The essence of applying HCA is to identify the grouping among the sampling sites based on their similarity, and to determine the specific contaminated sampling locations based on the pollution levels (Simeonov et al., 2002; Sundaray et al., 2011).

Clusters are classified based on the physical analysis of the dendrogram using Ward's method as a linkage rule (Atanacković et al., 2013; Sonğur & Top, 2016). Euclidean distance is chosen as a measure of similarity between the sampling sites. The dendrogram (Figure 4.3) shows clusters based on water quality at each sampling site with respect to the variations in the physical-chemical parameters and metals concentrations.

The thirteen sampling sites are classified into two separate clusters. The clusters show the influence/dominance of the pollution sources on water quality as suggested in PCA. As shown in the dendrogram, Cluster 1 is formed with TJYA, TKJA, TKJB, TP, TB, TSHA and TSH7, while Cluster 2 has TSP, TPPA, TPJ, TT, TPPB and TPRP. Cluster 1 is associated with contaminants (AN, BOD, and DO) possibly from domestic source, while Cluster 2 contains 63% of the ex-mining ponds characterized by high metal concentrations (As, Cd, Pb, Mn and Fe) that could be related to ex-mining activities. This makes the sites in Cluster 2 potential sources of toxic metal pollutants. Similarly, the sub-clusters reveal a similarity in terms of the degree of pollution among the linked sampling sites. Within Cluster 1 sites that show similar patterns of contamination are TP, TB, and TKJA.

Based on their specific locations, sites in Cluster 1 are surrounded by residential, commercial, and industrial areas while sites in Cluster 2 are surrounded by residential and industrial areas. The metal pollution in most of the sites (Puchong) of Cluster 2 could possibly be influenced by past mining operation being a predominant mining area (Ahmad & Jones, 2013; Balamurugan, 1991).

The results of the HCA confirm the influence of heavy metals and domestic wastes in groupings of the sites which could assist in consideration of the water for beneficial uses.

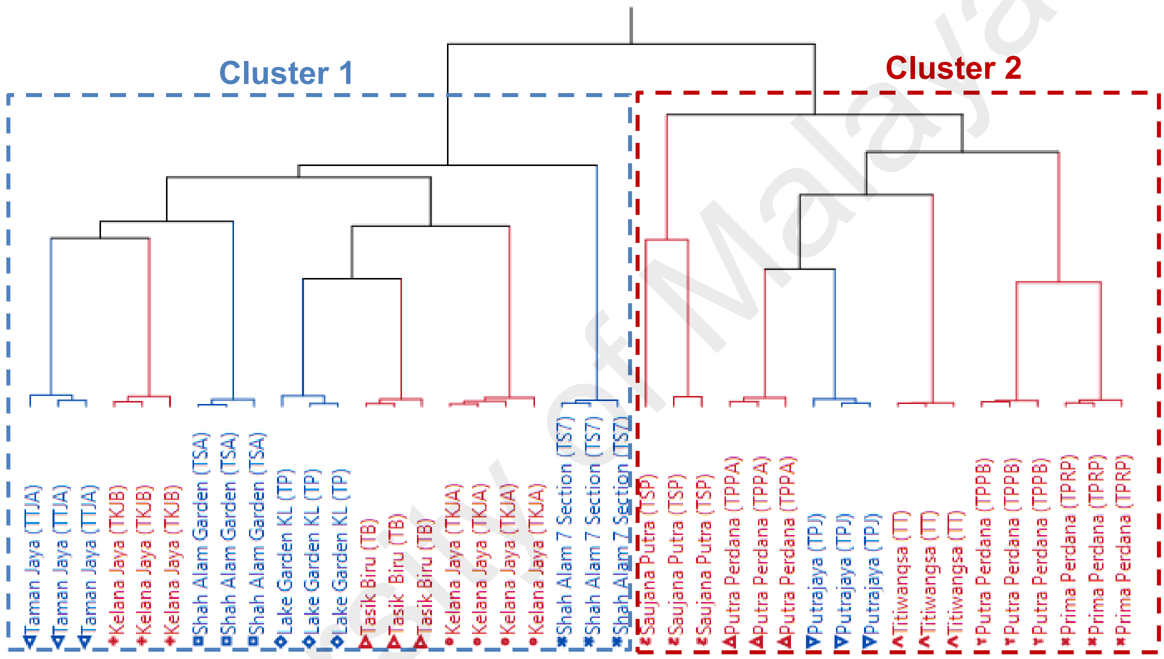

Figure 4.3: Schematic dendrogram on the pollution level of man-made lakes in Lakes in Klang Valley. ■ Ex-mining pond, ■ Lake

4.2.1.1 Linear Discriminant Analysis

LDA was used to predict the parameters which distinguish the ex-mining ponds from other lakes, by identifying the most significant parameters among the predictors (parameters under study). The model constructed with the training set give an accurate prediction (R^2 = 1.000), with no misclassification for the validation set, the overall classification matrices reveal that 100% of the samples are correctly classified.

Furthermore, the multivariate analysis of variance (MANOVA) for Pillai's Trace, Wilk's Lambda, Hotelling Lawley, and Roy's – Max Root give significant results ($F = 385.8740$, $p < 0.0001$).

In Figure 4.4 water samples from the ex-mining ponds are perfectly separated from others in the direction of Canonical 1. In this context, Mn, Cd, Ca, and As are highly associated with the ex-mining ponds, while pH, Fe, Mg, Na and EC are associated with the lakes. Similarly, the larger F – ratio value (much greater than 1) and the corresponding p – value, represent the significant difference ($p < 0.0001$) between the parameters in ex-mining ponds and lakes (Table 4.7). pH has the largest F – ratio of 103.601 ($p < 0.0001$) indicating much differences in acidity between ex-mining ponds and lakes in Klang Valley. Among the toxic heavy metal pollutants, As makes the largest contribution (F – ratio = 31.446, $p = 0.0000090$) in explaining the difference in water quality between ex-mining ponds and lakes, followed by Cd with (F – ratio = 18.714, $p = 0.0000306$). Though Pb has an F – ratio slightly greater than 1, no significant difference is observed (F – ratio = 1.832, $p = 0.1885145$). This is attributed to the low Pb concentrations in the studied sites. Mg (F – ratio = 0.073, $p = 0.7898319$) make the least contribution to the variation in water quality of the studied sites; these results justify the consideration given to the selected metals in this study.

The results presented for Klang Valley lakes/ex-mining ponds reaffirm that mining activities resulted in the release of toxic heavy metals, thereby significantly affecting the useful nature of the surface water quality (Wolkersdorfer, 2008). Hence there is a need to improve the water quality by metal removal and treatment processes for its proper utilization.

4.2.2 Melaka

The results of the metal concentrations and physical-chemical parameters are presented in Table 4.8 and Table 4.9. There are significant variations at (p<0.05) with As, 0.7-11.71 μg/L; Mn, 20.34-170.71 μg/L; Fe, 1.95-1586.05; Na, 2412.96-5562.75 μg/L; Mg, 845.52-2270.16 μg/L; and Ca, 2856.37-9134.99 μg/L. The variations in As, Mn and Fe concentrations could be attributed to the natural background concentration (Kusin et al., 2016; Ng et al., 2003). Mn and Fe, mostly have strong association in natural water (Swistock et al., 2015). The ex-mining ponds studied in Melaka and Negeri sembilan are found to contain low metal concentrations below INWQS and DWQSRW as shown in Figure 4.5, TR ex-mining pond recorded the least As concentration of 2.62 μg/L. Pb and Cd are not detected in both ex-mining ponds and lakes. Melaka is situated on limestone basement and granite peaks (Khoo & Tan, 1983; Schwartz & Askury, 1989), and these influence the geochemical properties of the ex-mining water resulting in high pH and low metal concentrations.

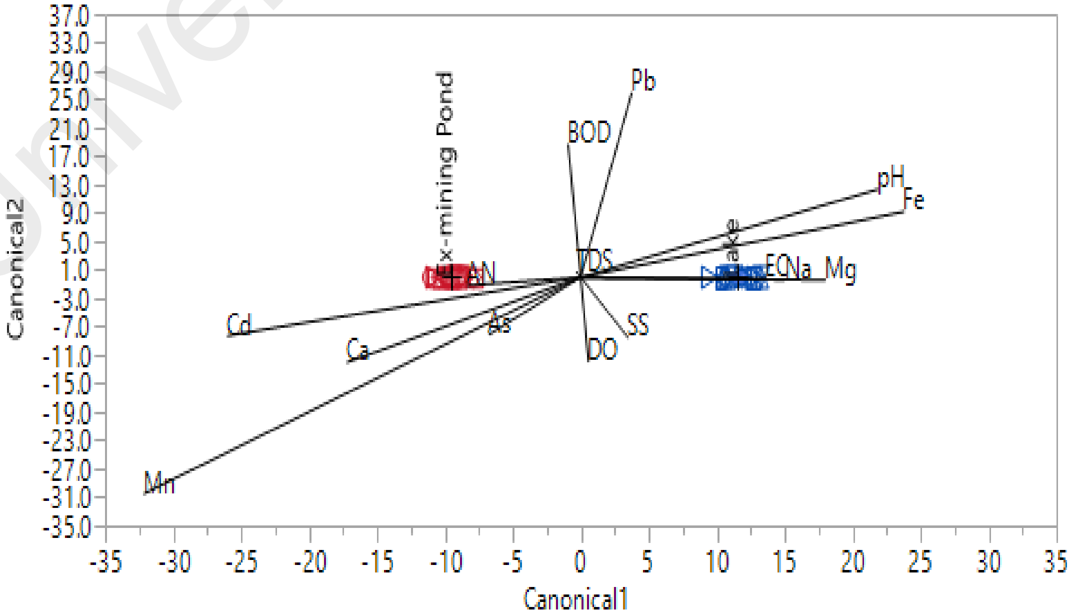

Figure 4.4: Canonical plot for discrimination of water samples from ex-mining ponds and lakes in Klang Valley

Table 4.7: Analysis of discriminating variables in ex-mining ponds and lakes in Klang Valley

Variable	F – Ratio	p - Value
As	31.446	0.0000090
Cd	18.714	0.0000306
Pb	1.832	0.1885145
Mn	8.122	0.0088393
Fe	41.481	0.0000012
Na	12.871	0.0014826
Mg	0.073	0.7898319
Ca	4.808	0.0382683
DO	0.386	0.5403236
BOD	0.476	0.4969912
TDS	1.510	0.2310475
AN	3.893	0.0601130
pH	103.601	0.0000000
SS	0.709	0.4079633
EC	13.549	0.0011749

A high Fe concentration of 1437 μg/L, and slightly high As concentrations above recommended standards of 1000 μg/L and 10 μg/L, respectively are reported in Tasik Tebat, a lake in Jasin with frequent fishing activity. The source of the As could be associated with the pesticides used to preserve the wooden boats, and the natural soil background concentration (Xing & Liu, 2011). A study of the geological analysis of Melaka ex-mining lands, shows the absence of metallic minerals (Osman & Siong, 2012). It is generally observed that ex-mining ponds in Melaka are not influenced by domestic or anthropogenic inputs. Agricultural activities around the studied sites are mostly rubber and palm oil cultivation whereby Melaka has the largest rubber plantation industry in Malaysia, covering over 1200 hectares (Courtenay, 1981; Leong et al., 2007). The variations in metal concentrations could therefore be due to the natural percentage abundance and geological background.

The physical-chemical parameters are also found to be within the INWQS and DWQSRW as shown in Figure 4.5. The pH of all the ex-mining ponds and lakes are within the acceptable range of 6.5 to 8.5, except Tasik Rantau which has a pH value of 10.3 ± 0.06. Tasik Rantau is located in Negeri Sembilan state near Melaka with a more basic rock basement (Ghani et al., 2008). Similarly Tasik Rantau surrounded by greeny vegetation has a DO of 9.16 mg/L, and this is higher than the values recorded in other sites studied in Melaka. DO concentration in Tasik Bukit Kalil and Tasik Tebat are observed to be less than the recommended guideline of 5 mg/L. This could be attributed to the new residential water inlet to Tasik Bukit Katil, and very low water level in Tasik Tebat. Organic matter are discharged from residential houses, and low dissolved oxygen is associated with the bottom water in aquatic ecosystem due to imbalance between oxygen supply from the surface waters and removal of oxygen from bottom water (Breitburg et al., 1997; Seitaj et al., 2017).

Table 4.8: Concentrations of metals and water quality parameters in Melaka ex-mining ponds

Variables	TBC1	TBC2	TBC3	TR
As	9.28±0.15	6.39±0.07	6.79±0.21	2.62±0.06
Cd	<0.3	<0.3	<0.3	<0.3
Pb	<0.2	<0.2	<0.2	<0.2
Mn	23±1	51±1	112±1	132±2
Fe	10±6	70±15	312±54	342±18
Na	2888±2	4078±44	4068±26	3372±9
Mg	1923±13	2238±23	2059±12	1771±17
Ca	6202±13	6127±26	5295±19	9084±40
DO	5.14±0.23	6.11±0.09	4.99±0.19	9.16±0.33
BOD	0.9±0.01	1.15±0.02	1.16±0.02	1.96±0.00
pH	7.24±0.00	7.31±0.01	7.01±0.05	10.28±0.06
TDS	47±0.00	59±0.31	56±0.00	77±0.92
TSS	<5	<5	<5	<5
AN	0.02±0.00	0.07±0.00	0.07±0.00	0.22±0.02
EC	82±1	102±1	97±1	136±2

Metals (µg/L), EC (µs/cm), pH (No unit), other parameters (mg/L)

Table 4.9: Concentrations of metals and water quality parameters in Melaka lakes

Variables	JD	TDT	TTB	TBK
As	0.72±0.02	1.28±0.02	10.48±0.89	4.67±0.64
Cd	<0.3	<0.3	<0.3	<0.3
Pb	<0.2	<0.2	<0.2	<0.2
Mn	128±11	20±0.09	107±16	170±0.49
Fe	194±18	109±13	1437±141	857±44
Na	3666±44	5497±46	2438±19	5072±49
Mg	1197±15	1626±1346	979±11	861±6
Ca	4379±16	5466±106	2875±14	5763±28
DO	4.89±0.01	6.09±0.53	4.56±0.30	3.67±0.12
BOD	1.25±0.03	1.09±0.01	2.12±0.00	1.5±0.01
pH	8.2±1.47	7.18±0.06	7.05±0.01	6.88±0.01
TDS	45±0.00	62±0.00	27±0.00	48±0.31
TSS	<5	<5	<5	<5
AN	0.28±0.04	0.17±0.00	0.03±0.01	0.09±0.01
EC	77±1	107±1	46±1	80±2

Metals (µg/L), EC (µs/cm), pH (No unit), other parameters (mg/L)

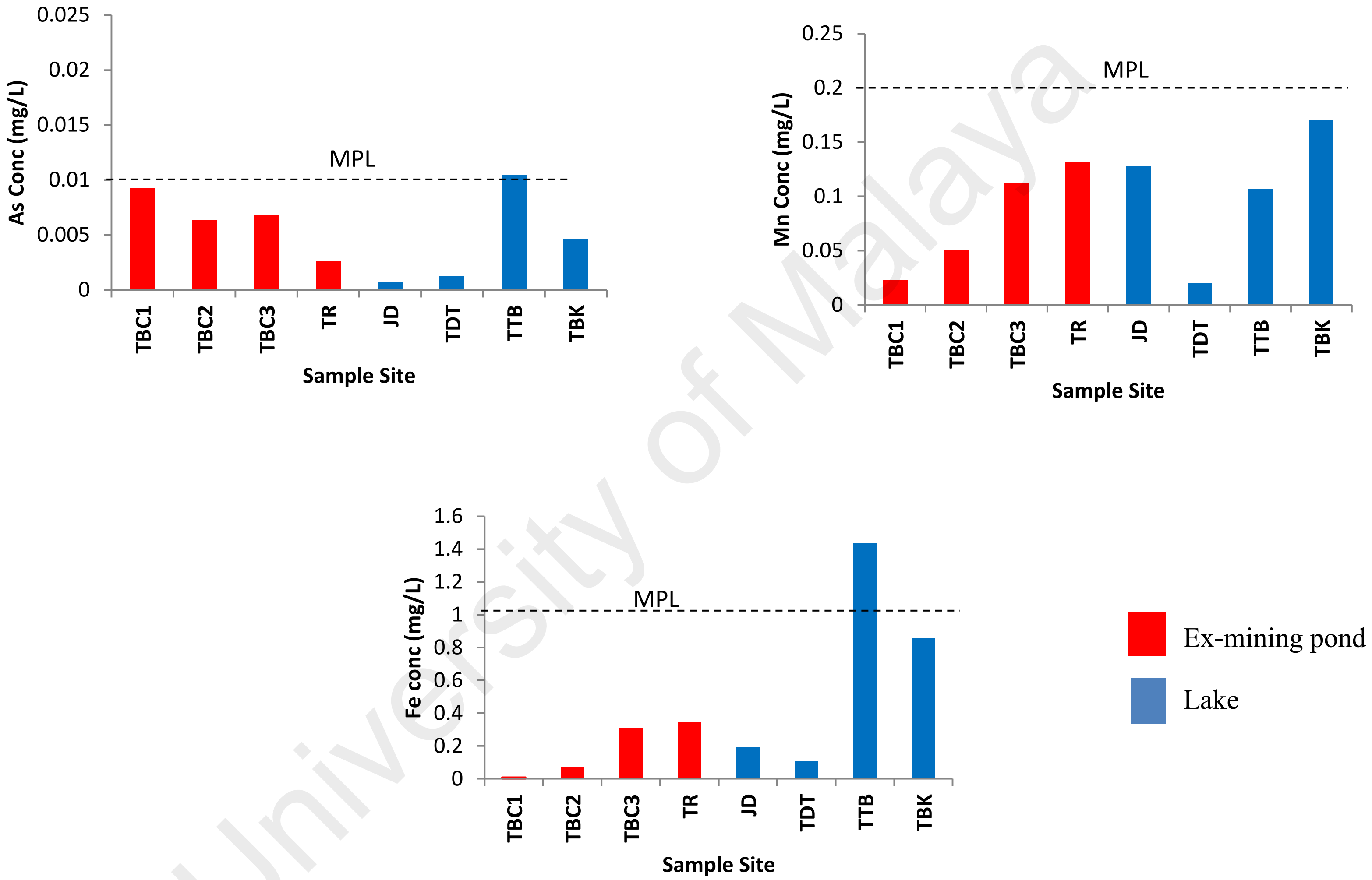

Figure 4.5: Heavy metal levels and physical-chemical parameters in ex-mining ponds and lakes in Melaka

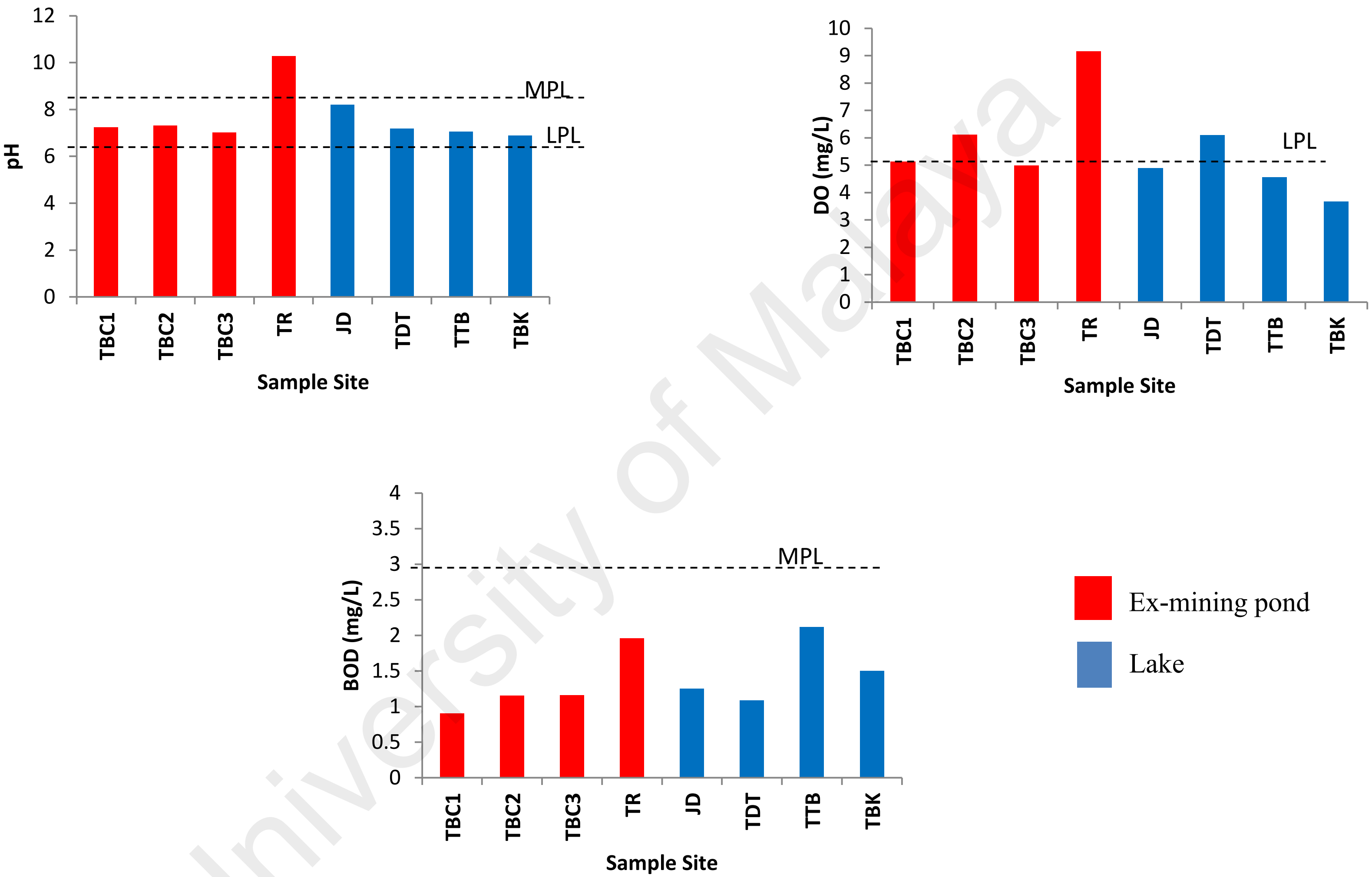

Figure 4.5: Continued

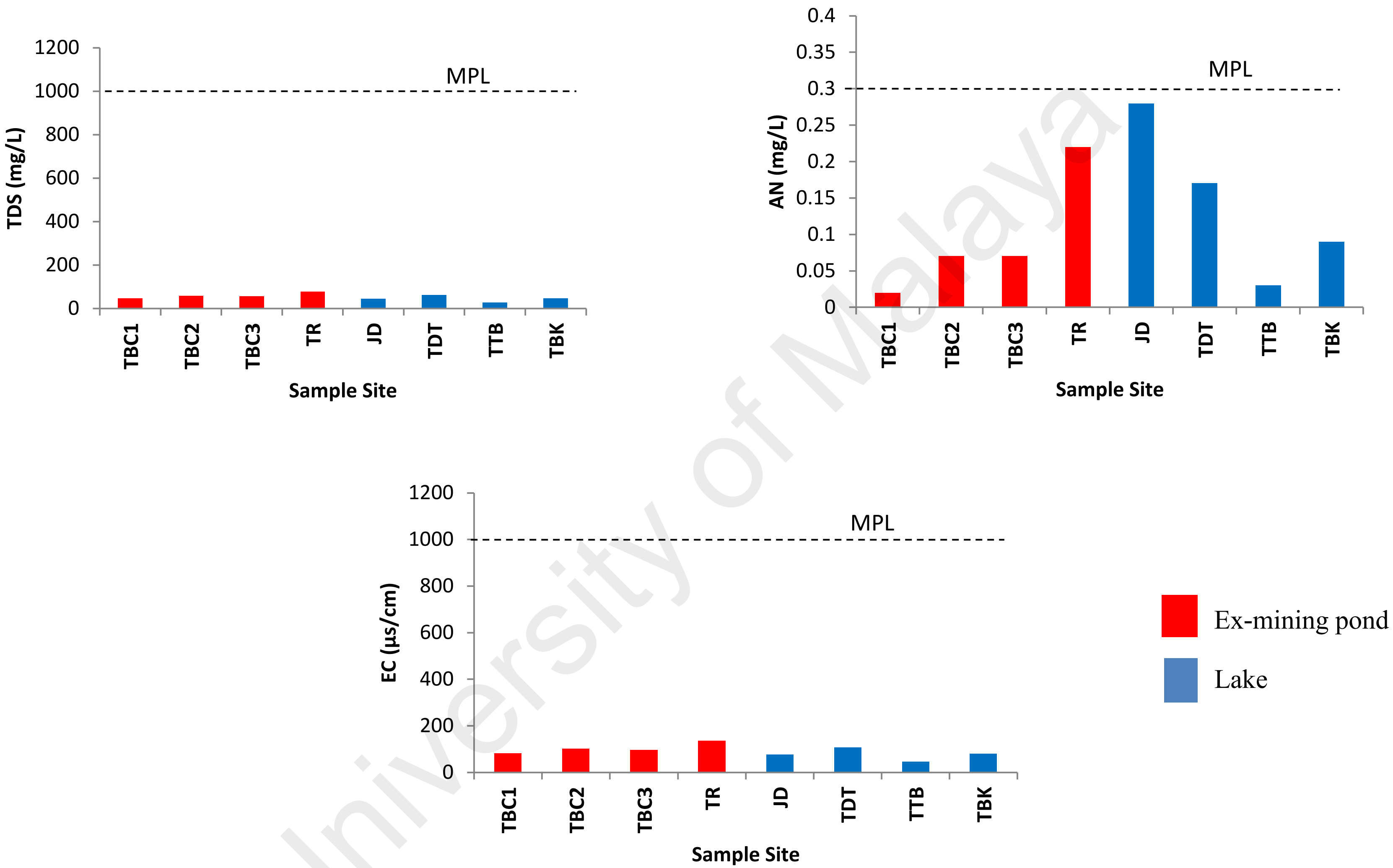

Figure 4.5: Continued

4.2.2.1 Principal Component Analysis

The variations in water quality parameters in Melaka ex-mining ponds and lakes were studied using principal component analysis, which reveals that PC1 and PC2 account for 46.2% and 23.6% of the total variance respectively. The loading of PC1 comprises of EC, DO, TDS and Ca as the major contributors (Figure 4.6). This is indicative of dissolution of rocks due to weathering and leaching (Atanacković et al., 2013), and absence of anthropogenic influence around the ex-mining ponds and lakes studied. PC2 is strongly loaded with Mn, Fe, pH, and BOD which could be attributed to natural background concentrations, geological background and low water level in most of the studied lakes. The land use around the studied sites are forest and agriculture mostly palm oil and rubber plantation (NAHRIM, 2009). In a study on water quality of Malaysian lakes, good water quality index of lakes, and rivers in Melaka were reported as class IIA suitable for human consumptions (Sharip et al., 2014). Table 4.10 gives the significant PCs and parameter loadings in Melaka.

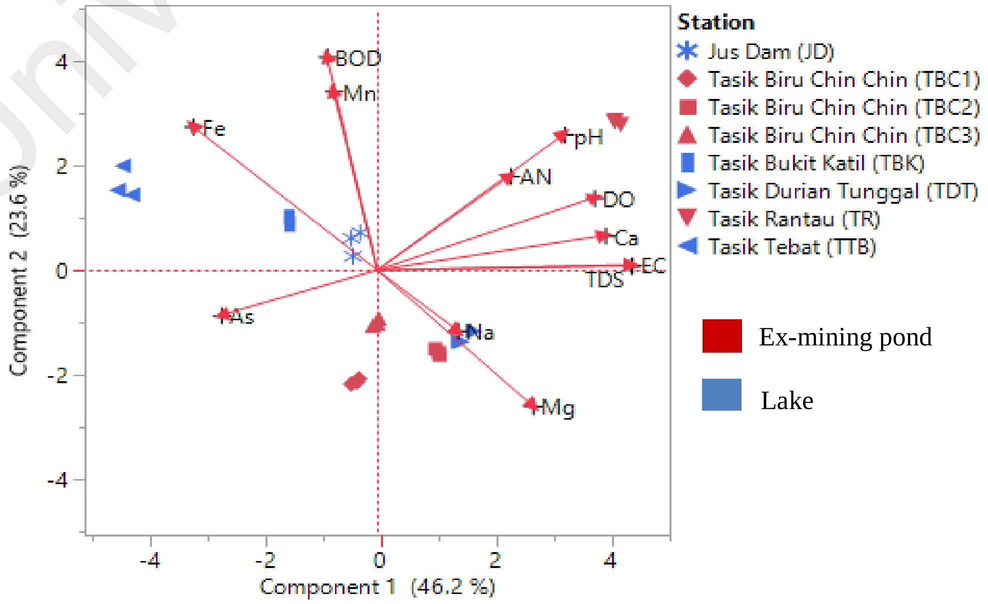

Figure 4.6: PCA bi-plot of metals and physical-chemical parameters in Melaka

Table 4.10: Percent explained variance for PCA in Melaka

Variable	PC1	PC2	PC3
As	-0.25543	-0.11357	0.51491
Mn	-0.07173	0.45331	-0.19535
Fe	-0.30200	0.36399	0.07955
Na	0.13064	-0.15196	-0.50385
Mg	0.25399	-0.34286	0.31430
Ca	0.37279	0.08796	0.15347
pH	0.30552	0.34352	0.25143
DO	0.35433	0.18302	0.24415
BOD	-0.08267	0.54034	0.18344
TDS	0.41444	0.01180	0.00195
AN	0.21724	0.23865	-0.39670
EC	0.41614	0.01372	0.01808
Eigen Value	5.5	2.8	2.1
Variance (%)	46.2	23.6	17.4
CV (%)	46.2	69.8	87.2

4.2.2.2 Factor Analysis

The most important variables in explaining water quality of ex-mining ponds and lakes in Melaka are explained using three VFs (Table 4.11). VF1 explains 35% of the total variance with significant loading on Mg and Fe which are related to weathering and dissolution of rocks minerals (Juahir et al., 2011). The negative loadings on As signifies the absence of toxic heavy metals influence on water quality of Melaka lakes and ex-mining ponds. Ca and pH are the most significant parameters in VF2 which explain 27% of the total variance suggesting high pH and dissolution of carbonate rocks; this justifies low levels of toxic metals in Melaka ex-mining ponds. VF3 explain 13% of the total variance with strong positive loading on TDS and EC, and strong negative loadings on As which indicate presence of inorganic salts and dissolved ions due to weathering, and absence of toxic heavy metals. The land use around Melaka ex-mining ponds is largely limited to agriculture (rubber and palm oil) and forestry.

4.2.2.3 Hierarchical Cluster Analysis

Grouping among the sampling sites and identifying the polluted sampling locations were analyzed using HCA. However, two-way HCA was introduced to study the distribution of the studied parameters due to the very low metal concentrations in the sample locations as shown in Figure 4.7. Three clusters are observed in which all the ex-mining ponds in Jasin and lakes formed cluster 1 with similar patterns of metal distribution among the sub-clusters which could be from the dissolution of rocks (Atanacković et al., 2013; Berger et al., 2000; Caraballo et al., 2016). Tasik Tetab formed cluster 2 which shows the influence of human activity such as fishing. Tasik Rantau an ex-mining pond from the neighboring state of Negeri Sembilan belongs to cluster 3 with significant parameters suggesting weathering and dissolution of rocks, and absence of anthropogenic input. The two-way HCA clearly reveals the absence of significant influence of toxic heavy metals (As, Cd, and Pb) on the water quality of Melaka ex-mining ponds and lakes.

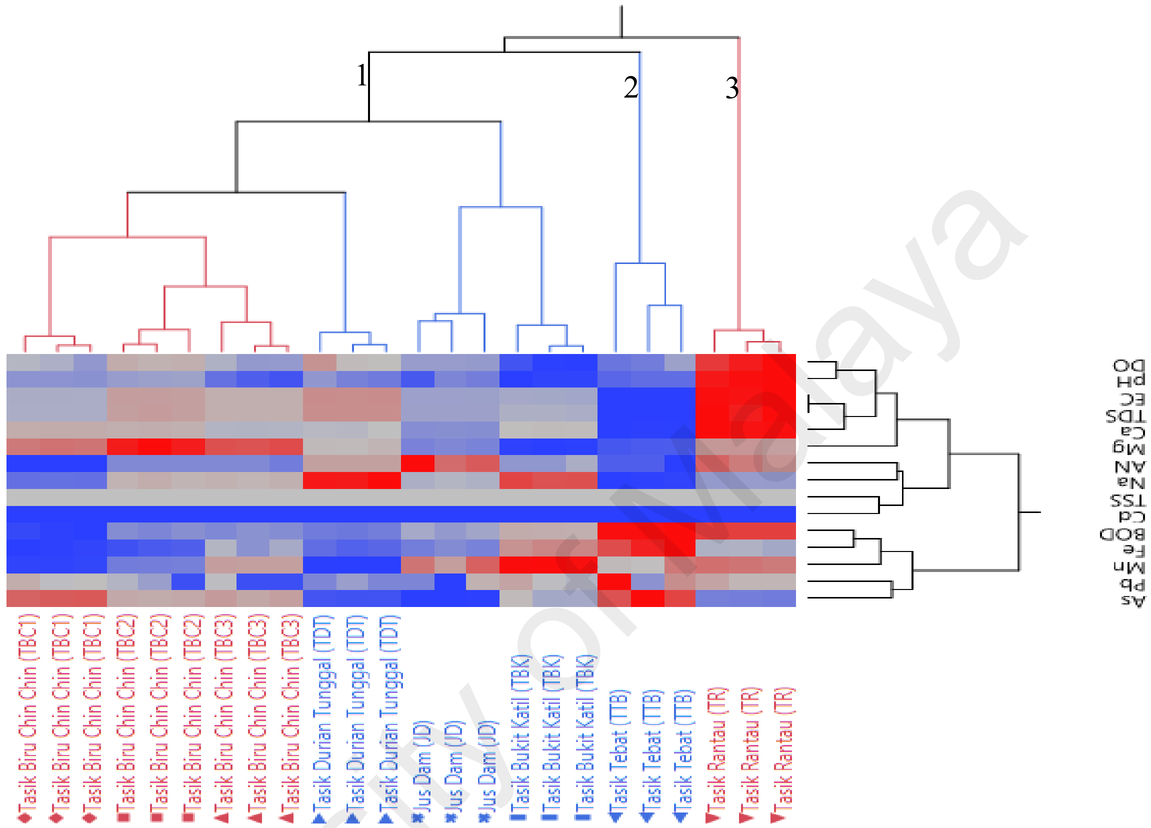

Figure 4.7: Two way HCA of ex-mining ponds and lakes in Melaka

It is clear that Tasik Tebat, a popular fishing lake, shows noticeable As concentration which is possibly related to the preservatives used on the fishing boats (Edwin & Thomas, 2001; Sreeja, 2008). The low metal levels could plausibly be associated with buffering of the ex-mining water due to dissolution of limestone host rocks (Berger et al., 2000).

Table 4.11: Loadings of the parameters on the Varimax-rotated PCs for ex-mining ponds and lakes in Melaka

Variable	VF1	VF2	VF3
As	-0.366750	0.065292	-0.911744
Mn	-0.678215	-0.595996	-0.232758
Fe	0.731216	-0.260477	0.441138
Na	0.544570	0.701043	-0.299781
Mg	0.900682	0.182523	-0.186980
Ca	0.087484	0.928686	-0.063088
pH	0.245810	0.864669	0.169602
DO	0.279822	0.470352	-0.106327
BOD	0.119770	0.290037	0.197399
TDS	0.025409	-0.009227	0.965569
AN	0.199124	0.336411	0.201565
EC	-0.366750	0.065292	0.911744
Eigen Value	4.5	3.5	1.7
Variance (%)	35	27.6	13
CV (%)	35	62.6	75.6

4.2.2.4 Linear Discriminant Analysis

LDA was successfully applied to study the parameters that differentiate the ex-mining ponds from the lakes. The model obtained gave accurate predictions with R^2 = 1.00 and no misclassification observed. As shown in Figure 4.8, there was a clear separation between water samples from ex-mining ponds and lakes. The parameters associated with ex-mining ponds are As, Mn, Ca, Mg, and EC, on the other hand pH, and Na were highly associated with lakes. This demonstrates that weathering and dissolution of rocks are the main sources of metals in the ex-mining ponds. Weathering of host rocks and mine tailing were associated with high metal levels in mines and adjacent rivers (Fanfani et al., 1997; Favas et al., 2011). Similarly, association of Na and pH in the lakes indicate a natural source of the parameters. Furthermore, MANOVA reveals no significant difference between the ex-mining ponds and lakes. F-ratio values (Table 4.12) shows P values > 0.05 except for Mn with the highest contribution (F - Ratio = 9.520, p = 0.0215123), Na (F - Ratio = 3.584, p = 0.1071900), and As (F - Ratio = 2.912, p = 0.1387763). Fe (F - Ratio = 0.05, p = 0.9469950) make the least contribution to the differences in water quality of ex-mining ponds and lakes in Melaka.

This finding suggests no influence of toxic heavy metals in ex-mining ponds of Melaka, and they can be considered for human consumption and domestic purposes.

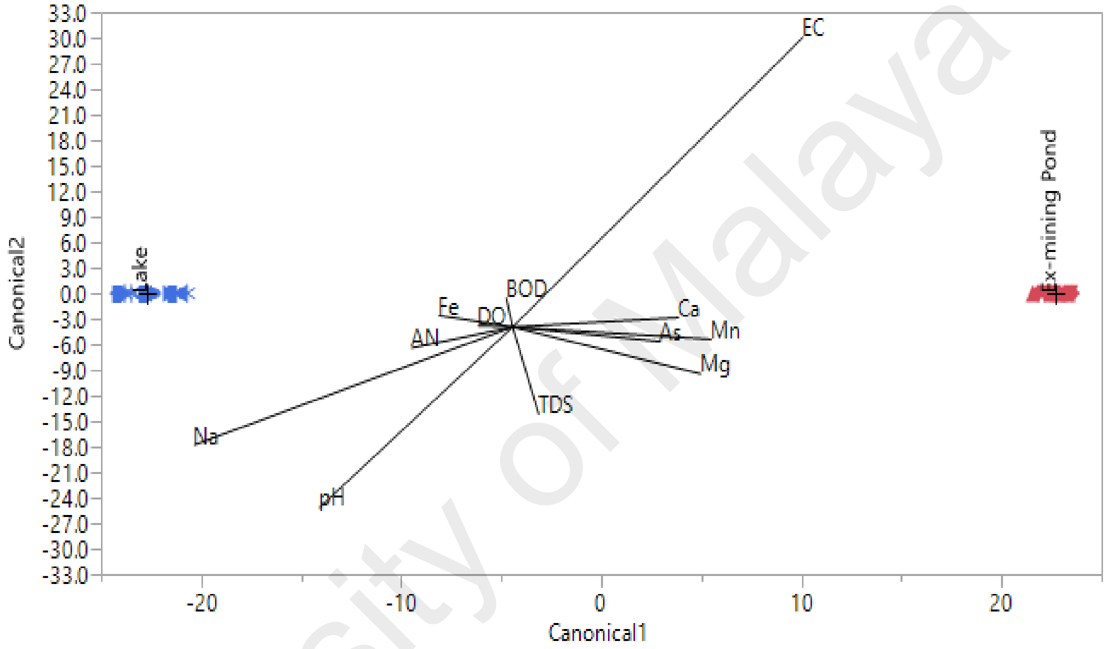

Figure 4.8: Canonical plot for the discrimination of water samples from ex-mining ponds and lakes in Melaka

Table 4.12: Analysis of discriminating variables in ex-mining ponds and lakes in Melaka

Variable	F – Ratio	P - Value
As	2.912	0.1387763
Cd	0.310	0.5979105
Pb	0.209	0.6638567
Mn	9.520	0.0215123
Fe	0.005	0.9469950
Na	3.584	0.1071900
Mg	0.834	0.3962430
Ca	0.556	0.4842161
pH	1.113	0.3320963
DO	0.188	0.6794826
BOD	0.110	0.7517046
TDS	0.882	0.3839634
AN	1.381	0.2844826
EC	0.056	0.8213686

4.3 Preliminary Human Health Risk Assessment

Studies on human health risk assessments relate the risks and exposure to the metals in a medium and the duration of the exposure. Considering that water ingestion goes directly into the human system, a study on toxicity of the metals and proper risk evaluations are of great concern. As, Cd and Pb were selected for the risk analysis, but emphasis was given to As due to its dominance in the ex-mining water (Ashraf et al., 2011b; Ngure et al., 2014; Palmer et al., 2016; Razo et al., 2004). The chemical speciation and mobility of As determined its toxicity (Bowell et al., 2014) and its bioavailability which is higher in aqueous phase and easily gets into the digestive system of humans. Inorganic As species are generally more toxic than the organic form (Hughes, 2002). Commonly found in surface water and oxidizing environment is the inorganic As, while the organic form is predominant in the sediment under reducing conditions (Bowell et al., 2014). Previous findings have established the presence of inorganic As in Malaysian ex-mining ponds (Yusof et al., 2001), hence the quantitative estimation of the risks of exposure to As is of much relevance.

To quantitatively study the risk of exposures to toxic metals in the Klang Valley, HCA was applied to the ex-mining ponds and lakes based on As, Cd and Pb concentrations as shown in Figure 4.9. Two clusters are observed based on the metal levels. Cluster 1 is dominated by lakes and ex-mining ponds with low metal levels; Cluster 2 is dominated with ex-mining ponds of higher metal concentrations due to mining mostly from Puchong area. This classification is made to cater for the much anthropogenic input into the water in Klang Valley. In Melaka, only As concentrations is detected in all the sites studied while Cd and Pb are not detected as shown in Table 4.13, hence are assigned values of half the detection limit ($L/2$), where L is the limit of detection (Beyer et al., 2016; Finkelstein & Verma, 2001; Hornung & Reed, 1990; Larter et al., 2016). The ex-mining pond in Negeri Sembilan (TR) included in this study for comparison has the least As concentration compared to Melaka lakes and ex-mining ponds. Pb and Cd were also not detected, hence not further considered in the risk assessment.

The distribution of the selected metals in the ex-mining ponds of Klang Valley and Melaka are clearly illustrated in a 3D scatter plot (Figure 4.10a). Most ex-mining ponds with high Cd and As are separated from other low metal loadings at the intersection of the three axis. The PCA bi-plot reveals high Cd and As loading in PC1 which contributed about 56% of the total variation and PC2 is loaded with Pb (Figure 4.10b). The selected metals are clearly relevant and have been taken into consideration in the risks of exposures to both adults and children (Saha et al., 2017; Tripathee et al., 2016; Wang et al., 2005).

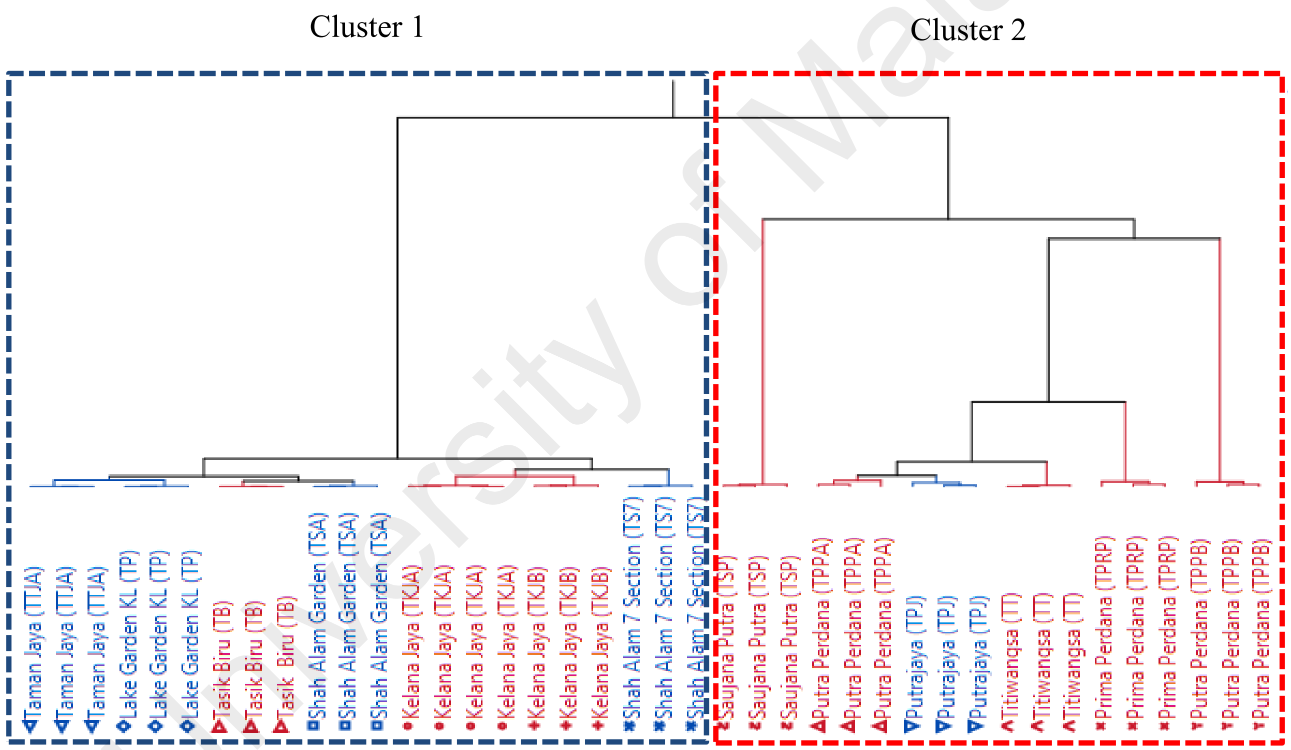

Figure 4.9: Hierarchical cluster analysis on As, Cd and Pb concentrations in ex-mining ponds and lakes in Klang Valley

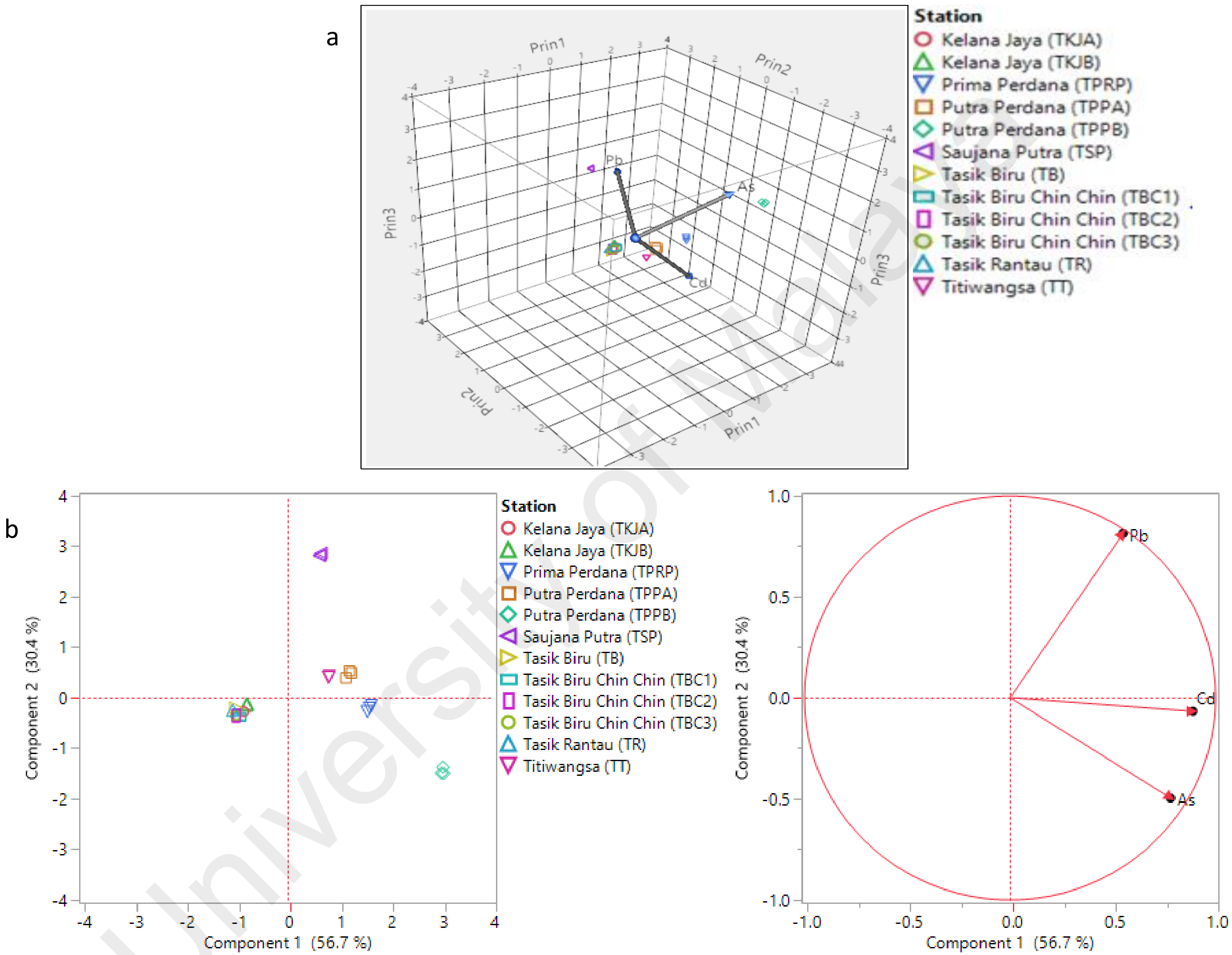

Figure 4.10: 3D scatter plot (a) and PCA bi-plot (b) on the distribution of As, Cd, and Pb in ex-mining ponds of Klang Valley and Melaka

Table 4.13: The distributions of selected heavy metals in the study areas

Normal distribution values for the selected heavy metals in Klang valley*		
Parameter	**Cluster 1**	**Cluster 2**
As	N (7.72, 4.19)	N (58.85, 33.90)
Cd	N (0.19, 0.10)	N (7.13, 3.50)
Pb	N (0.45, 0.24)	N (8.58, 3.01)
The distribution of selected heavy metals in Melaka		
Parameter	**Ex-mining Ponds**	**Lakes**
As	N (5.97, 2.50)	ln N (0.94, 1.05)
Cd	U (0.04, 0.09)	U (0.04, 0.09)
Pb	U (0.04, 0.09)	U (0.04, 0.09)

The concentrations are presented in µg/L

The values that follow normal distribution are written as N (μ_x, σ_x) signifying mean of metal concentrations and standard deviation. ln N (μ_x, σ_x) stands for log-normal distribution with μ_x, and σ_x as arithmetic mean and arithmetic standard deviation, U (a, b) refers to uniform distribution with minimum and maximum value.

4.3.1 Non-Carcinogenic Risk Assessment

Hazard quotient (HQ) is generally used to characterize non-carcinogenic risk. It is pertinent to consider the metal intake and dose of the exposure that could lead to deleterious health effects. The hazard quotient is defined as the chronic daily intake divided by the reference dose. For the estimation of daily exposure and non-carcinogenic hazards of a single chemical substance, average daily exposure ($Exp_{Ix/Dx}$) and hazard quotient (HQ) were used (USEPA, 1989). This is denoted by the equation below;

$$HQ = \frac{Exp_{Ix/Dx}}{RfD_{Ix/Dx}} \quad \textbf{(4.1)}$$

where Exp_{Ix} is the daily exposure or dose through ingestion of water (µg/kg/d), Exp_{Dx} is the exposure or dose through dermal absorption (µg/kg/d), $RfD_{Ix/Dx}$ are the reference dose values of the x^{th} metal (µg/kg/d) that indicate the maximum acceptable oral and dermal exposures. The RfD values of As, Cd and Pb for the ingestion and dermal routes are 0.3, 0.5 and 1.4 µg/kg/d; and 0.123, 0.025, and 0.42 µg/kg/d respectively (USEPA, 2006; WHO, 2006b; Wu et al., 2009). For a value of HQ less than 1, there is low risk and acceptable level of non-carcinogenic toxic effects. There could be a health concern associated with the exposure at HQ value greater than 1 (USEPA, 2011; Zhuang et al., 2014).

In view of the nature of ex-mining water in containing different heavy metal compositions, the evaluation of cumulative health risks became necessary. This is achieved by summing the individual HQ of each metal to give HI (Li et al., 2014) as shown in equation 4.2 below;

$$HI = \sum_{x=1}^{n} HQ_x \quad \textbf{(4.2)}$$

where HI is the hazard index (no unit), and HQ is the hazard quotient. For HI values greater than 1, it indicates that the exposed individuals may face adverse health effects. If the HI value is less than 1, then it is improbable to have harmful effects (Muhammad et al., 2011; Wongsasuluk et al., 2014).

Ingestion of water is the most common exposure route to hazardous metals and chemical substances, but dermal route is also considered in this study, because most Malaysian ex-mining ponds are used for recreational activities (Ghani et al., 2008;

Takaijudin et al., 2012b). These two routes are the usual paths to toxic heavy metals exposure in the environment (Kavcar et al., 2009; Ordóñez et al., 2011; Qu et al., 2012; Yi et al., 2011). The evaluations of the daily exposure by ingestion of the x^{th} metal can be accomplished using equation 4.3 below (Iqbal et al., 2013; USEPA, 1989);

$$Exp_{Ix} = \frac{C_x * IR * EF * ED}{BW * AT} \quad \textbf{(4.3)}$$

where Exp_{Ix} is the daily exposure or dose through ingestion of water (μg/kg/d), C_x is the concentration of the x^{th} metal in ex-mining ponds or lake (μg/L), IR is the average daily ingestion rate of the individual (L/day), EF is the exposure frequency (Days/year), ED is the exposure duration of the individual (Year), BW is the body weight of the individual (Kg), and AT is the period of time in which the dose is averaged (Days).

Monte Carlo simulation was applied to the experimental results to obtain C_x. The BW of the Malaysian adult and child were obtained from a survey on adult body mass index of Malaysians (Azmi et al., 2009) and a study on the nutritional status of Malaysian children (Zaini et al., 2005) respectively. Details of the parameter values and their sources are presented in Table 4.14. In Monte Carlo simulation technique, the aforementioned independent exposure variables were considered as input variables for estimating the probability of the daily exposure doses using equation 4.3. The simulation was performed with 45 thousand iterations using SimulAr in Microsoft Excel 2013. The results of the exposures were treated separately for adult and child, and expressed at 95^{th} and 99^{th} percentile of the simulated distributions which are considered to be of reasonable maximum exposures (RME), because average concentrations and average exposure to ingestions could not give cumulative health risks of toxic metals exposure in water (Kavcar et al., 2009; Sany et al., 2014).

Table 4.14: Parameters for the risk assessment and evaluation

Parameters	Adult	Child	Source
C (µg/L)	Please refer to Table 4.13		This study
ED (Years)	30	7	This study
EF (Days/Year)	365	365	This study
IR (L/day)	1.69	1.2	(MDG, 2013; Mohamad, 2016)
BW (Kg)	N (62.65,18.6)	N (32.57,8.7)	(Azmi et al., 2009; Zaini et al., 2005)
ET (hr/d)	0.6	0.6	(Iqbal & Shah, 2013)
SA (cm^2/day)	5700	2800	(Sany et al., 2014)
AT* (Days)	27265	27265	(DOS, 2016)
AT** (Days)	10950	2555	(Bonn et al., 2011; USEPA, 2005)
CF (L/cm^3)	0.001	0.001	(USEPA, 1989)

AT* = Average time for carcinogenic risk
AT** = Average time for non-carcinogenic risk

N (μ_x, σ_x) denotes a normal distribution with an arithmetic mean of μ_x and arithmetic standard deviation of σ_x.

Individual exposure through the dermal route was evaluated using equation 4.4 by considering the dermal permeability constant values for the selected toxic metals (USEPA, 1997). The dermal permeability constant represents the strength or effectiveness of the penetration of chemical substances through the human skin (Potts & Guy, 1992).

$$Exp_{Dx} = \frac{C_x * SA * K_p * ET * EF * ED * CF}{BW * AT} \quad \textbf{(4.4)}$$

In the equation, Exp_{Dx} is the exposure or dose through dermal absorption (µg/kg/d), SA is the exposed skin surface area of the individual available for contact (cm^2/day), ET is the exposure time for contact with water (hr/day), CF is the unit conversion factor (L/cm^3), Kp is the dermal permeability constant.

The details of the reference values used for the ingestion and dermal exposures are shown in Table 4.15 below.

Table 4.15: Reference dose, slope factor and dermal permeability constant values for the selected metals (USEPA, 1997,; Wang et al., 2015; Wu et al., 2009)

Parameter	RfD_{Ix} (μg/kg/d)	RfD_{Dx} (μg/kg/d)	SF_{Ix} $(μg/kg/d)^{-1}$	SF_{Dx} $(μg/kg/d)^{-1}$	Kp
As	0.3	0.123	0.0015	0.0015	0.001
Cd	0.5	0.005	0.00038	0.00038	0.001
Pb	1.4	0.420	0.0000085	-	0.0001

It is observed that HI values for sites in Klang Valley for both clusters 1 and 2 exceed the reference limit of 1 with the highest HQ_{Ix} contribution from As, while the HQ_{Ix} for Pb and Cd are far below 1 (Table 4.16). Therefore, among the selected metals, As may pose a health hazard and considered to have a potential non-carcinogenic risk to both adults and children. This implies that consumption of water from the lakes by the average adult could result in deleterious health effects (Figure 4.11) due to As especially in cluster 2 which is dominated by ex-mining ponds having extreme HQ_{Ix} values of 11.02 and 12.90 for the 95th and 99th percentile respectively. For children, they receive the highest As exposure due to ingestion of water from the sites in cluster 1 and 2 (Figure 4.13), with the highest HQ_{Ix} value of 14.96 and 15.61 at 95th and 99th percentile respectively in cluster 2. These values are more than 50 times the As RfD (Table 4.17). This suggests that water consumption from the studied sites could result in deleterious health effect due to exposure to As in both adults and children. More attention has been given to children, for the fact that children are more exposed to dangers of heavy metals than adults (Zhuang et al., 2014). Previous findings showed to a greater extent the exposure of children to toxic heavy metals than adults, which consequently affects their brain and nervous systems (Saleh et al., 2017; Tripathi et al., 1999).

The source of high As content in the water may be attributed to the ex-mining activities as previously demonstrated in the principal component analysis (Figure 4.2), and previous related findings in ex-mining water (Low et al., 2016; Razo et al., 2004; Rojas & Vandecasteele, 2007; Yusof et al., 2001). Other sources of As in the lakes beside natural background, especially in Shah Alam include industrialization. As was found to be discharged from agrochemical and metal coating industries, among others (Balamurugan, 1991; Hamzah et al., 2011).

Figure 4.12 shows that exposure due to dermal contacts seems to possess no health threat with HQ_{Dx} less than 1 as shown in (Table 4.16 and Table 4.17). HQ_{Dx} for all the selected metals in the study areas were found to be less than 1, hence dermal contacts in both adult and child categories might not cause any health effects. The values for the ingestion and dermal exposures for all the sites were found in order of As>Pb>Cd except for the dermal exposure in cluster 2 with order As>Cd>Pb.

The exposure assessment to ex-mining ponds and lake water in Melaka are also described in Table 4.16 and 4.17. There is low adult exposure to As, Cd and Pb for both the ingestion and dermal contact (Figure 4.15), the HI_{Ix} and HI_{Dx} at 95th and 99th percentile were found to be below 1 respectively. These values are less than 1 indicating the absence of any detrimental effect and non-carcinogenic health risk to users by ingestion and dermal contacts. However, children are vulnerable to exposure effects of As by ingestion in both ex-mining ponds and lakes (Figure 4.16). The HI_{Ix} of 1.19 and 1.29 for both ex-mining ponds and lakes at 95th and 99th percentile are observed. This indicates there could be health threat from As ingestion with HQ_{Ix} of 1.1 and 1.2 for both ex-mining ponds and lakes. This may pose adverse health hazards and potential non-carcinogenic effect to children. The HQ_{Ix} of Cd and Pb are found to be significantly below 1, indicating very improbable health threat. The dermal exposure by child appears to pose no possible health threat in both ex-mining ponds and lakes.

Among the selected heavy metals in this study, As has the lowest ingestion RfD of 0.3 µg/kg/d (Table 4.15), and its concentrations in lake TTB is 10.48 µg/L which is slightly higher than INWQS value of 10 µg/L. Similarly, ex-mining pond TBC1 has As concentration of 9.28 µg/L slightly below INWQS which could be hazardous considering the RME. This explains why there is a risk associated to children in Melaka in which the WQI gives a good water quality rating based on the metal levels.

Due to numerous factors, children have a greater tendency or potential for detrimental health effects than adults even below the metal permissible limits (CDCP, 2017; Guruge et al., 2017), and may not be able to clear themselves of exposure due to undeveloped body mechanism for detoxification. Child's activities, age, and behavior and biology results in exposure differences from adults living in the same environment (Selevan et al., 2000). For instance, the ratio of skin surface area to body mass of child is 2.7 times greater than adult (Snodgrass, 1992), and water consumption is more than 2 times higher than adult per body weight (USEPA, 1997). Similarly, Zaldivar (1977) reported that young children received a higher daily dose of As per unit body weight than adult under the same arsenic water concentration because of their higher intake per unit body weight.

Table 4.16: Non-carcinogenic Risk Assessment on Adult Exposure by Ingestion/Skin Contact with Water from Ex-mining ponds and lakes

	Klang Valley (Cluster 1)								Klang Valley (Cluster 2)							
	Exp_{Ix} (µg/kg/d)		**HQ_{Ix}**		**Exp_{Dx} (µg/kg/d)**		**HQ_{Dx}**		**Exp_{Ix} (µg/kg/d)**		**HQ_{Ix}**		**Exp_{Dx} (µg/kg/d)**		**HQ_{Dx}**	
	Percentile		Percentile		Percentile		Percentile		Percentile		Percentile		Percentile		Percentile	
	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}
As	3.8×10^{-1}	3.9×10^{-1}	1.2×10^{0}	1.3×10^{0}	7.6×10^{-4}	8.10×10^{-4}	6.3×10^{-3}	6.5×10^{-3}	3.0×10^{1}	3.1×10^{1}	10.1×10^{0}	10.5×10^{0}	6.1×10^{-3}	6.3×10^{-3}	4.9×10^{-2}	5.1×10^{-2}
Cd	9.4×10^{-3}	9.7×10^{-3}	1.8×10^{-2}	1.9×10^{-2}	$1.9\times10\text{-}^{5}$	1.96×10^{-5}	7.6×10^{-4}	7.8×10^{-4}	3.4×10^{-1}	3.5×10^{-1}	6.7×10^{-1}	7.0×10^{-1}	6.8×10^{-4}	7.1×10^{-4}	2.7×10^{-2}	2.8×10^{-2}
Pb	2.2×10^{-2}	2.3×10^{-2}	2.1×10^{-2}	7.5×10^{-2}	4.4×10^{-5}	4.60×10^{-5}	1.0×10^{-5}	1.1×10^{-4}	3.5×10^{-1}	3.7×10^{-1}	2.5×10^{-1}	1.7×10^{0}	2.8×10^{-4}	2.9×10^{-3}	6.0×10^{-4}	7.0×10^{-4}
HI			**1.23**	**1.39**			**0.0071**	**0.0073**			**11.02**	**12.90**			**0.07**	**0.08**

	Melaka Ex-mining Ponds								Melaka Lakes							
	Exp_{Ix} (µg/kg/d)		**HQ_{Ix}**		**Exp_{Dx} (µg/kg/d)**		**HQ_{Dx}**		**Exp_{Ix} (µg/kg/d)**		**HQ_{Ix}**		**Exp_{Dx} (µg/kg/d)**		**HQ_{Dx}**	
	Percentile		Percentile		Percentile		Percentile		Percentile		Percentile		Percentile		Percentile	
	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}
As	2.4×10^{-1}	2.5×10^{-1}	8.0×10^{-1}	8.2×10^{-1}	4.8×10^{-4}	5.0×10^{-4}	3.9×10^{-3}	4.0×10^{-3}	2.6×10^{-1}	2.7×10^{-1}	8.6×10^{-1}	9.2×10^{-1}	5.2×10^{-4}	5.6×10^{-4}	4.2×10^{-3}	5.7×10^{-3}
Cd	2.5×10^{-2}	2.6×10^{-2}	5.1×10^{-2}	5.3×10^{-2}	5.1×10^{-5}	5.4×10^{-5}	2.0×10^{-3}	2.1×10^{-3}	2.5×10^{-2}	2.6×10^{-2}	5.1×10^{-2}	5.3×10^{-2}	5.1×10^{-5}	5.4×10^{-5}	2.0×10^{-3}	2.1×10^{-3}
Pb	2.5×10^{-2}	2.6×10^{-2}	1.8×10^{-2}	1.9×10^{-2}	2.0×10^{-4}	2.1×10^{-4}	4.9×10^{-4}	5.1×10^{-4}	2.5×10^{-2}	2.6×10^{-2}	1.8×10^{-2}	1.9×10^{-2}	5.1×10^{-6}	5.4×10^{-6}	1.2×10^{-5}	1.3×10^{-5}
HI			**0.869**	**0.892**			**0.0064**	**0.0066**			**0.929**	**0.992**			**0.0062**	**0.0078**

Table 4.17: Non-carcinogenic Risk Assessment on Child Exposure by Ingestion/Skin Contact with Water from Ex-mining Ponds and lakes

	Klang Valley (Cluster 1)								Klang Valley (Cluster 2)							
	Exp_{Ix} (μg/kg/d)		HQ_{Ix}		Exp_{Dx} (μg/kg/d)		HQ_{Dx}		Exp_{Ix} (μg/kg/d)		HQ_{Ix}		Exp_{Dx} (μg/kg/d)		HQ_{Dx}	
	Percentile		Percentile		Percentile		Percentile		Percentile		Percentile		Percentile		Percentile	
	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}
As	5.2×10^{-1}	5.4×10^{-1}	1.7×10^{0}	1.8×10^{0}	7.3×10^{-4}	7.6×10^{-4}	5.9×10^{-3}	6.1×10^{-3}	4.1×10^{0}	4.3×10^{0}	13.7×10^{0}	14.3×10^{0}	5.7×10^{-3}	6.0×10^{-3}	4.6×10^{-2}	4.8×10^{-2}
Cd	1.2×10^{-2}	1.3×10^{-2}	2.5×10^{-2}	2.6×10^{-2}	1.8×10^{-5}	1.9×10^{-5}	7.2×10^{-4}	7.4×10^{-4}	4.6×10^{-1}	4.8×10^{-1}	9.2×10^{-1}	9.6×10^{-1}	6.4×10^{-4}	6.7×10^{-4}	2.5×10^{-2}	2.7×10^{-2}
Pb	3.0×10^{-2}	3.1×10^{-2}	2.1×10^{-2}	7.5×10^{-2}	1.6×10^{-5}	1.7×10^{-5}	4.0×10^{-5}	4.2×10^{-5}	4.8×10^{-1}	5.0×10^{-1}	3.4×10^{-1}	3.5×10^{-1}	2.7×10^{-4}	2.8×10^{-4}	6.5×10^{-4}	6.7×10^{-4}
HI			**1.75**	**1.90**			**0.0067**	**0.0069**			**14.96**	**15.61**			**0.072**	**0.076**

	Melaka Ex-mining Ponds								Melaka Lakes							
	Exp_{Ix} (μg/kg/d)		HQ_{Ix}		Exp_{Dx} (μg/kg/d)		HQ_{Dx}		Exp_{Ix} (μg/kg/d)		HQ_{Ix}		Exp_{Dx} (μg/kg/d)		HQ_{Dx}	
	Percentile		Percentile		Percentile		Percentile		Percentile		Percentile		Percentile		Percentile	
	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}	95^{th}	99^{th}
As	3.2×10^{-1}	3.3×10^{-1}	1.1×10^{0}	1.2×10^{0}	4.5×10^{-4}	4.7×10^{-4}	3.7×10^{-3}	3.8×10^{-3}	3.5×10^{-1}	3.8×10^{-1}	1.1×10^{0}	1.2×10^{0}	4.9×10^{-4}	5.3×10^{-4}	4.0×10^{-3}	4.3×10^{-3}
Cd	3.5×10^{-2}	3.6×10^{-2}	7.0×10^{-2}	7.2×10^{-2}	4.9×10^{-5}	5.1×10^{-5}	1.0×10^{-4}	2.0×10^{-4}	3.5×10^{-2}	3.6×10^{-2}	7.0×10^{-2}	7.2×10^{-2}	4.9×10^{-5}	5.1×10^{-5}	1.9×10^{-3}	2.0×10^{-3}
Pb	3.5×10^{-2}	3.6×10^{-2}	2.5×10^{-2}	2.6×10^{-2}	4.9×10^{-6}	5.1×10^{-6}	1.1×10^{-5}	1.2×10^{-5}	3.5×10^{-2}	3.6×10^{-2}	2.5×10^{-2}	2.6×10^{-2}	4.9×10^{-6}	5.1×10^{-6}	1.1×10^{-5}	1.2×10^{-5}
HI			**1.19**	**1.29**			**0.0038**	**0.0040**			**1.19**	**1.29**			**0.0059**	**0.0063**

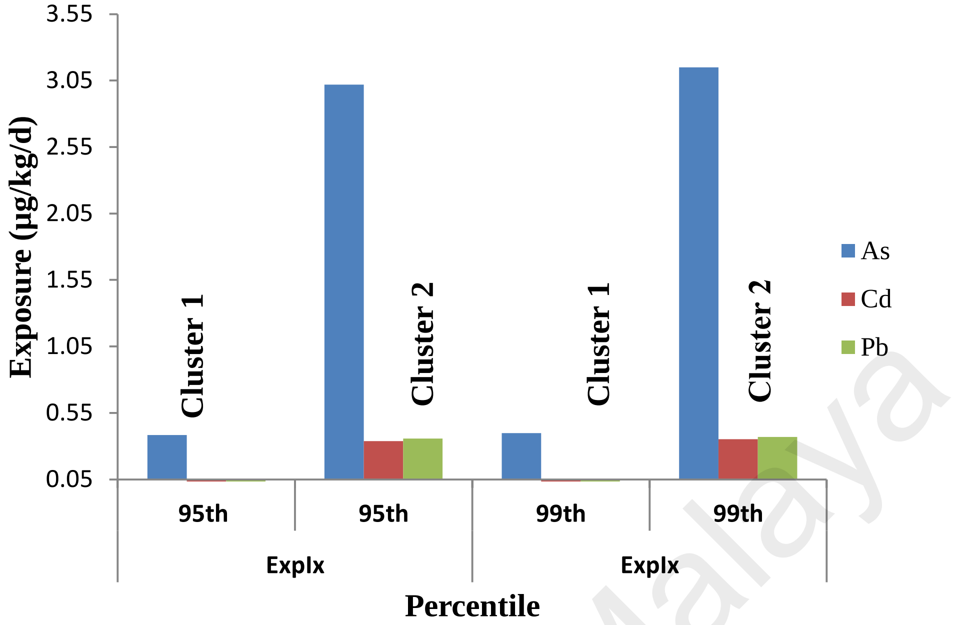

Figure 4.11: Non-carcinogenic risk exposure of adult by ingestion of water from ex-mining ponds and lakes in Klang Valley

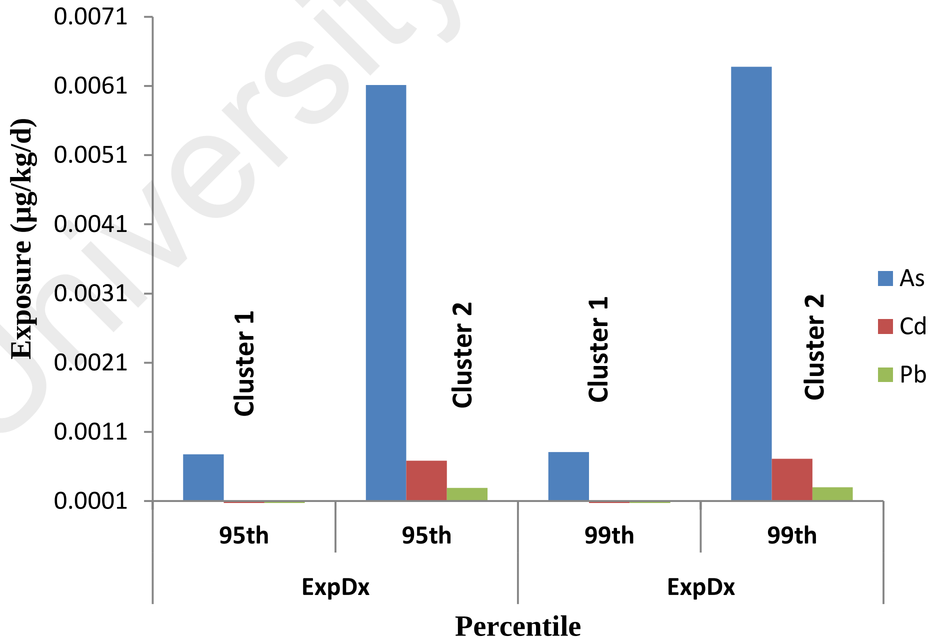

Figure 4.12: Non-carcinogenic risk exposure of adult by dermal contact with water from ex-mining ponds and lakes in Klang Valley

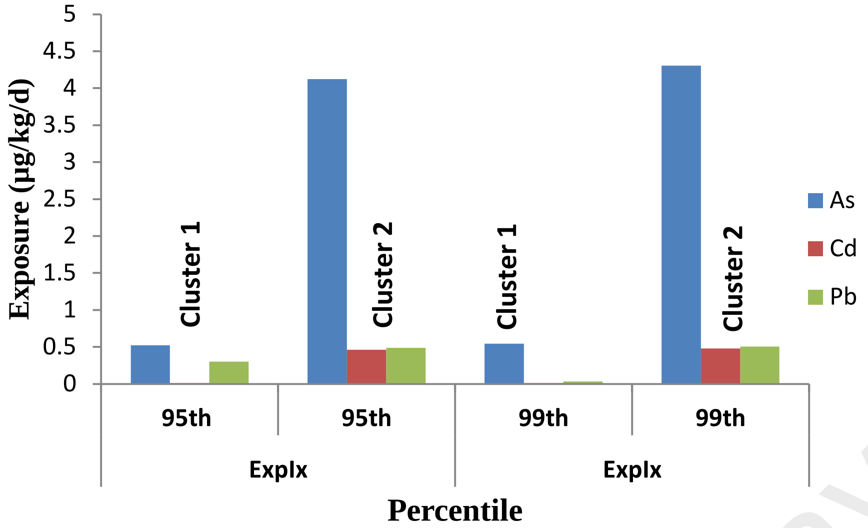

Figure 4.13: Non-carcinogenic risk exposure of child by ingestion of water from ex-mining ponds and lakes in Klang Valley

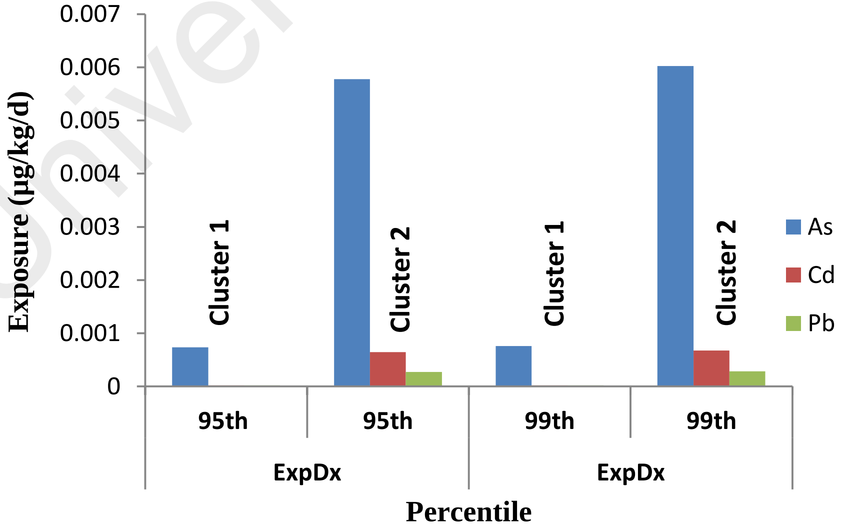

Figure 4.14: Non-carcinogenic risk exposure of child by dermal contact with water from ex-mining ponds and lakes in Klang Valley

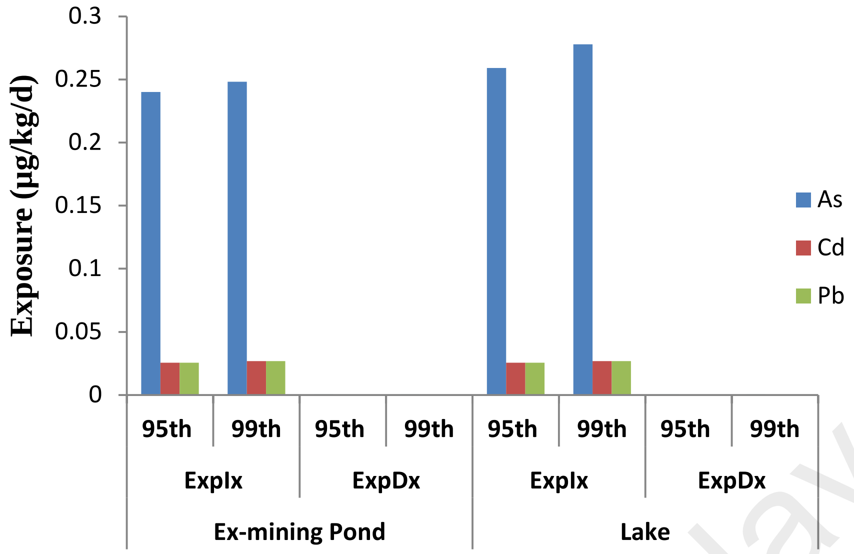

Figure 4.15: Non-carcinogenic risk exposure of adult by ingestion and dermal contact with water from ex-mining ponds and lakes in Melaka

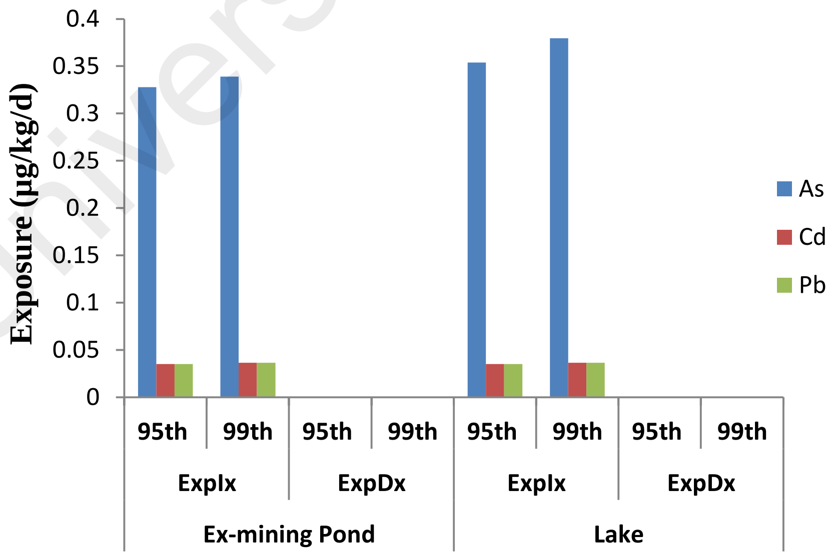

Figure 4.16: Non-carcinogenic risk exposure of child by ingestion and dermal contact with water from ex-mining ponds and lakes in Melaka

4.3.2 Carcinogenic Risk Assessment

Carcinogenic risk assessment estimates the incremental probability of a person to develop cancer over a lifetime due to exposure to a potential carcinogenic substance (USEPA, 1989). In the evaluation of carcinogenic risk, the slope factor (SF) converts the estimated daily intake of the toxic substances for a lifetime exposure directly to the increasing risk of a person to develop cancer. The evaluation of carcinogenic risk is computed using equation 4.5;

$$CR_x = Exp_{Ix/Dx} \times SF \quad \textbf{(4.5)}$$

where CR_x is the tendency of a person to develop cancer over a lifetime upon exposure to the x^{th} metal (No unit), $Exp_{Ix/Dx}$ is the daily exposure by ingestion and dermal routes (µg/kg/d), and SF is the slope factor for the carcinogenic substance (µg/kg/d). The exposure duration is considered as the exposure frequency (EF) in 365 days/year for ED of 30 years and 7 years for adult and child respectively, Average time (AT) of 27265 days is the Malaysian lifetime expectancy (DOS, 2016). Substituting the above parameters in equation 4.5 gives the exposure risk probabilities at 95^{th} and 99^{th} percentile. An acceptable carcinogenic risk value of 10^{-4} to 10^{-6} implies a safe level of developing cancer, while a risk value greater than 10^{-4} is considered unacceptable and could cause cancer upon long time exposure (Wongsasuluk et al., 2014).

The results of the cancer risk assessment due to ingestion for adult exposed to the sites in cluster 2 Klang Valley (Figure 4.17) revealed that only As records carcinogenic risk of 1.82×10^{-3} and 1.89×10^{-3} for 95^{th} and 99^{th} percentile respectively. These values exceed the safe limits for cancer risk implying a probability of having 2 people developing cancer out of 1000 people. As carcinogenic risk in cluster 1, Cd and Pb in both clusters were found to be within the acceptable limit as shown in Table 4.18. The total carcinogenic risk in Klang Valley varies generally in the order As > Cd > Pb. Dermal contact was found to pose no threat of cancer in both clusters as shown in Figure 4.18 and Figure 4.20, the Pb slope factor is not available and therefore its dermal cancer risk was not evaluated. The carcinogenic risks of ingestion of the selected metals by children in cluster 1 and 2 are within the safe limit, and not appear to pose any cancer health effect (Figure 4.19). The total cancer risk of children in Klang Valley is within the acceptable limit, while adults may be of total cancer threat due to exposure to ex-mining ponds and lakes in cluster 2. Comparably, Tables 4.18 and 4.19 shows that As, Cd and Pb carcinogenic risks for the ingestion and dermal contact for Melaka lakes and ex-mining ponds are also found to be within acceptable limits in both adults and children (Figure 4.21 and Figure 4.22). The total cancer threat is also within safe limit for both adult and children.

Shorter exposure duration of children is much related to the low carcinogenic risk compared to adults (Li et al., 2014). Additionally, in a study on the sensitivity of children versus older ages in exposed population, adults received more cumulative cancer risk of arsenic in water over their life time period (Tsuji et al., 2004). In a similar study, high cancer risk for adult compared to child due to exposure to surface as well as ground water was reported in Bangshi River using both probabilistic and deterministic estimations (Saha et al., 2017).

Nonetheless, there could be some existing uncertainties in the risk characterization as emphasized in the USEPA and other relevant documents in the formulation of RfD and cancer SF which might not be specific to Malaysians (USEPA, 2004; Wu et al., 2009). This finding therefore presents a preliminary result, and more accurate analysis should be carried out for RfD and cancer SF which are specific for Malaysians.

Table 4.18: Carcinogenic risk assessment on Adult exposure to ingestion/skin contact with water from ex-mining ponds and lakes

	Klang Valley (Cluster 1)				**Klang Valley (Cluster 2)**			
	Percentile	**CR_{Ix}**	**CR_{Dx}**	**Total CR**	**Percentile**	**CR_{Ix}**	**CR_{Dx}**	**Total CR**
As	95^{th}	2.31×10^{-4}	4.67×10^{-7}	2.31×10^{-4}	95^{th}	1.82×10^{-3}	3.68×10^{-6}	1.82×10^{-3}
	99^{th}	2.40×10^{-4}	4.86×10^{-7}	2.40×10^{-4}	99^{th}	1.89×10^{-3}	3.84×10^{-6}	1.89×10^{-3}
Cd	95^{th}	1.44×10^{-6}	2.91×10^{-9}	1.44×10^{-6}	95^{th}	5.16×10^{-5}	1.04×10^{-7}	5.17×10^{-5}
	99^{th}	1.48×10^{-6}	2.91×10^{-9}	1.48×10^{-6}	99^{th}	5.38×10^{-5}	1.08×10^{-7}	5.39×10^{-5}
Pb	95^{th}	7.55×10^{-8}	NA	NA	95^{th}	1.21×10^{-6}	NA	NA
	99^{th}	7.91×10^{-8}	NA	NA	99^{th}	1.25×10^{-6}	NA	NA
	Melaka Ex-mining Ponds				**Melaka Lakes**			
	Percentile	**CR_{Ix}**	**CR_{Dx}**	**Total CR**	**Percentile**	**CR_{Ix}**	**CR_{Dx}**	**Total CR**
As	95^{th}	1.44×10^{-4}	2.92×10^{-7}	1.44×10^{-4}	95^{th}	1.56×10^{-4}	3.15×10^{-7}	1.56×10^{-4}
	99^{th}	1.49×10^{-4}	3.02×10^{-7}	1.49×10^{-4}	99^{th}	1.67×10^{-4}	3.38×10^{-7}	1.67×10^{-4}
Cd	95^{th}	3.91×10^{-6}	7.91×10^{-9}	3.91×10^{-6}	95^{th}	3.91×10^{-6}	7.91×10^{-9}	3.91×10^{-6}
	99^{th}	4.07×10^{-6}	8.24×10^{-9}	4.07×10^{-6}	99^{th}	4.07×10^{-6}	8.24×10^{-9}	4.07×10^{-6}
Pb	95^{th}	8.74×10^{-8}	NA	NA	95^{th}	8.74×10^{-8}	NA	NA
	99^{th}	9.11×10^{-8}	NA	NA	99^{th}	9.11×10^{-8}	NA	NA

NA =Not Available

Table 4.19: Carcinogenic risk assessment on Child exposure to ingestion/skin contact with water from ex-mining ponds and lakes

	Klang Valley (Cluster 1)				Klang Valley (Cluster 2)			
Parameter	**Percentile**	**CR_{Ix}**	**CR_{Dx}**	**Total CR**	**Percentile**	**CR_{Ix}**	**CR_{Dx}**	**Total CR**
As	95th	7.36×10^{-5}	1.03×10^{-7}	7.37×10^{-5}	95th	5.80×10^{-4}	8.11×10^{-7}	5.80×10^{-4}
	99th	7.65×10^{-5}	1.07×10^{-7}	7.66×10^{-5}	99th	6.04×10^{-4}	8.46×10^{-7}	6.04×10^{-4}
Cd	95th	4.59×10^{-7}	6.42×10^{-10}	4.59×10^{-7}	95th	1.64×10^{-5}	2.29×10^{-8}	1.71×10^{-5}
	99th	4.72×10^{-7}	6.61×10^{-10}	4.72×10^{-7}	99th	1.71×10^{-5}	2.39×10^{-8}	1.71×10^{-5}
Pb	95th	2.40×10^{-8}	NA	2.40×10^{-8}	95th	3.88×10^{-7}	NA	3.88×10^{-7}
	99th	2.52×10^{-8}	NA	2.52×10^{-8}	99th	4.01×10^{-7}	NA	4.01×10^{-7}
	Melaka Ex-mining Ponds				**Melaka Lakes**			
Parameter	**Percentile**	**CR_{Ix}**	**CR_{Dx}**	**Total CR**	**Percentile**	**CR_{Ix}**	**CR_{Dx}**	**Total CR**
As	95th	4.61×10^{-5}	6.45×10^{-8}	4.61×10^{-5}	95th	4.97×10^{-5}	6.96×10^{-8}	4.97×10^{-5}
	99th	4.76×10^{-5}	6.67×10^{-8}	4.76×10^{-5}	99th	5.33×10^{-5}	7.46×10^{-8}	5.33×10^{-5}
Cd	95th	1.24×10^{-6}	1.74×10^{-9}	1.24×10^{-6}	95th	1.24×10^{-6}	1.74×10^{-9}	1.24×10^{-6}
	99th	1.29×10^{-6}	1.81×10^{-9}	1.29×10^{-6}	99th	1.29×10^{-6}	1.81×10^{-9}	1.29×10^{-6}
Pb	95th	1.24×10^{-6}	NA	NA	95th	2.78×10^{-8}	NA	NA
	99th	1.29×10^{-6}	NA	NA	99th	2.90×10^{-8}	NA	NA

NA = Not Available

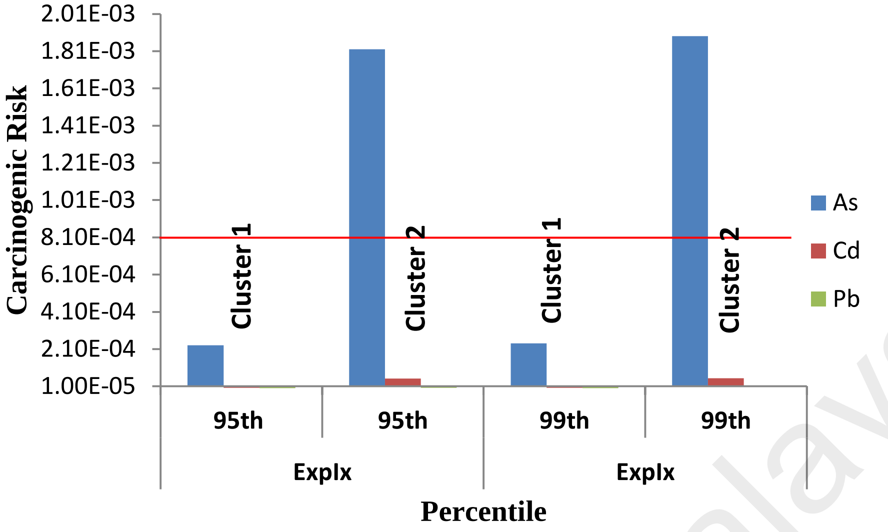

Figure 4.17: Carcinogenic risk exposure of adult by ingestion of water from ex-mining ponds and lakes in Klang Valley

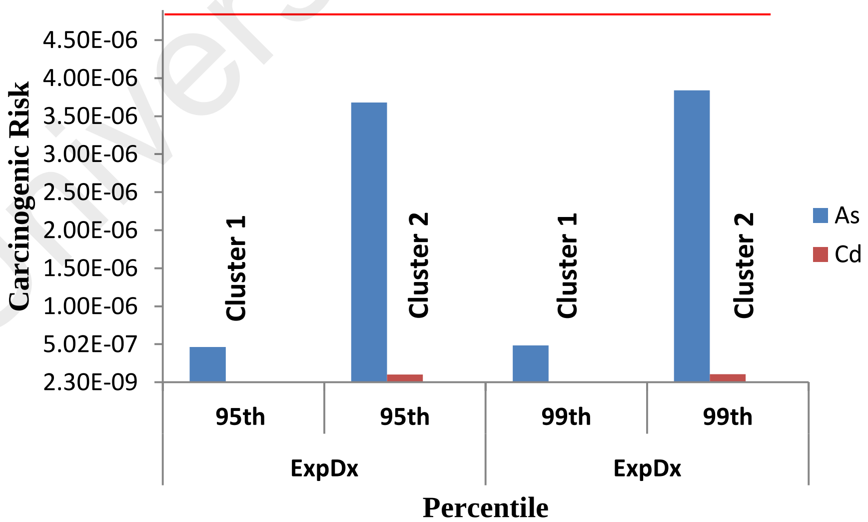

Figure 4.18: Carcinogenic risk exposure of adult by dermal contact with water from ex-mining ponds and lakes in Klang Valley

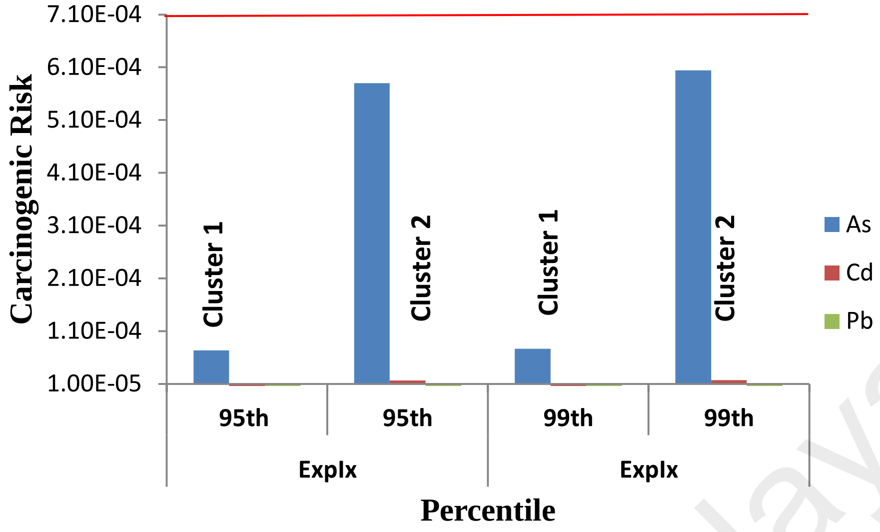

Figure 4.19: Carcinogenic risk exposure of child by ingestion of water from ex-mining ponds and lakes in Klang Valley

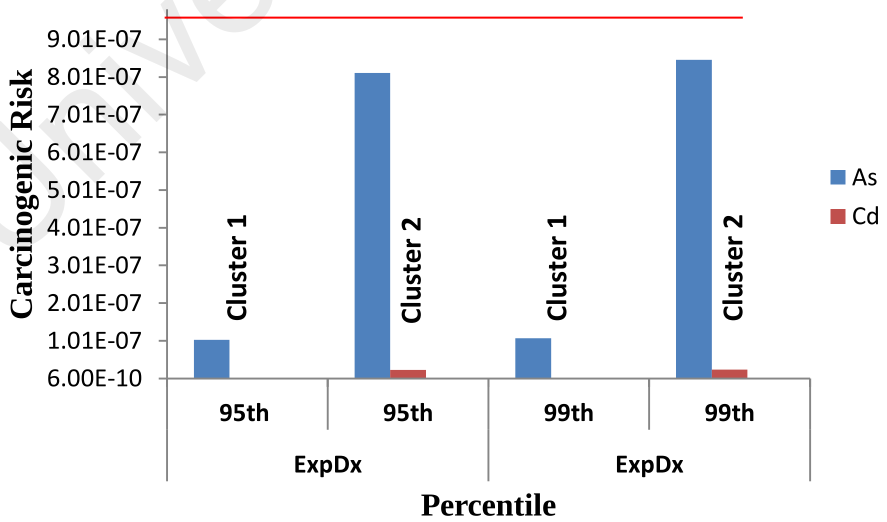

Figure 4.20: Carcinogenic risk exposure of child by dermal contact with water from ex-mining ponds and lakes in Klang Valley

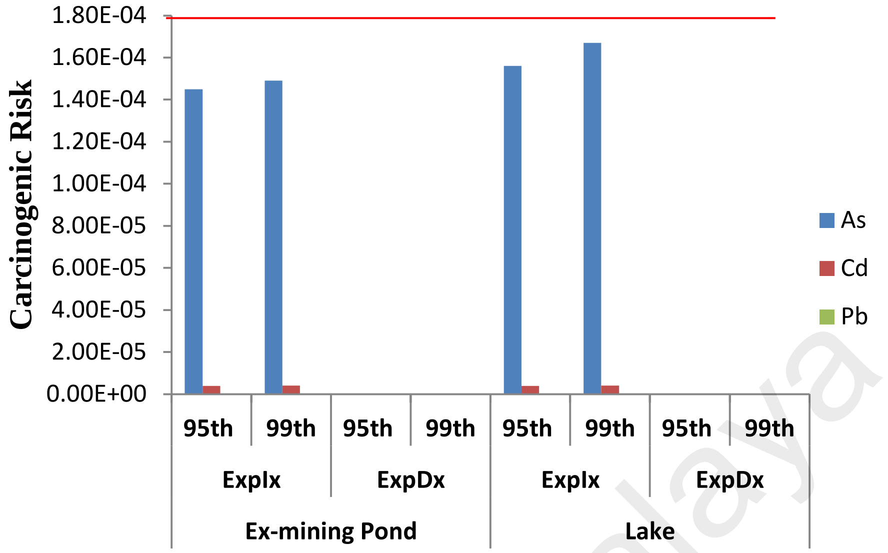

Figure 4.21: Carcinogenic risk exposure of adult by ingestion and dermal contact with water from ex-mining ponds and lakes in Melaka

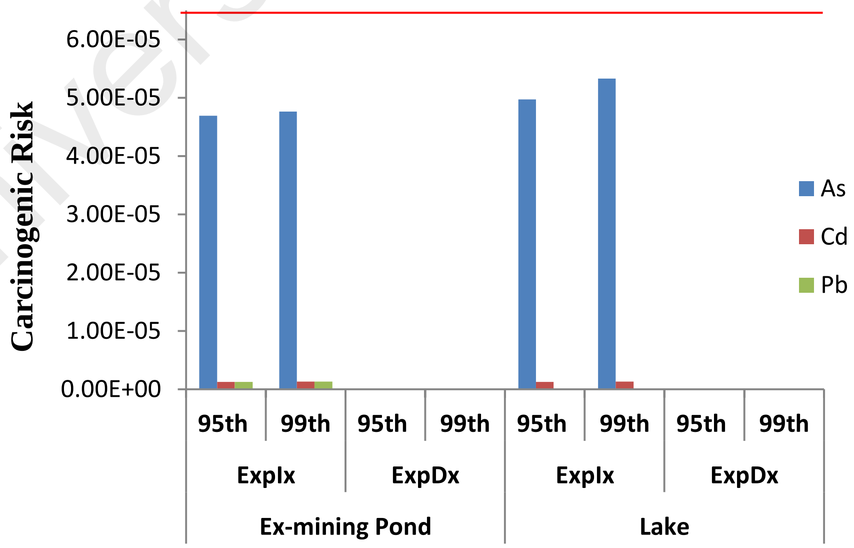

Figure 4.22: Carcinogenic risk exposure of child by ingestion and dermal contact with water from ex-mining ponds and lakes in Melaka

4.4 Water Quality Index

WQI gives a rating of water quality for specific use with respect to the selected relevant parameters. Such index is designed to solve water classification problems considering environmental influences. The chemometric analysis and health risk assessment carried out in this study were aimed at developing a suitable WQI for the accurate assessment of ex-mining ponds and lakes for human consumption. In the development of WQI of ex-mining ponds, toxic pollutants related to mining, and other environmental/anthropogenic pollutants are taken into consideration. Two separate WQI's are developed based on the selected heavy metal pollutants and physical-chemical parameters. The following steps are adopted in the development of the WQI; parameter selection based on comparing results of the extensive literature survey with the experimental values, development of the weightage based on the INWQS values, sub index calculation, and finally aggregating the weightage and sub index to give the overall WQI. The final decision on utilization of the water is taken based on the results of the indexes obtained.

4.4.1 Parameter Selection

For the water intended for human consumption, priority is given to parameters of health concern (WHO, 2006a). The incessant complaints among Malaysian citizens, researchers, and government officials were centered only on the levels of heavy metals in the ex-mining water (Bernama, 2014a, 2014b; Charles, 2014; Eileen, 2014; Heng & Kannan, 2014; Izham, 2014; Khairi, 2014; Nation, 2014). In this regard, toxic metals associated with mining are the main focus on the final decision, because conventional water treatment processes consider and regulate other physical-chemical parameters through a sequential process of rapid mixing, flocculation, sedimentation, filtration, and disinfection (Asami et al., 2016; Ayekoe et al., 2017; Westerhoff et al., 2005). Similarly, pH, BOD, and DO are considered easily purified parameters and skillfully handled in the water treatment plants (Hou et al., 2016). However, toxic metals are not removed during conventional water treatment processes, but using expensive techniques such as forward osmosis, floatation, enhanced ultra-filtration and use of cellulose modified filters (d'Halluin et al., 2017; Deliyanni et al., 2017; Huang et al., 2017; Low et al., 2016; You et al., 2017).

The parameters selected in the development of the WQI of ex-mining ponds were also mostly highlighted in previous studies related to the pollutants in mining areas across the world including Malaysia (Ashraf et al., 2011b; Low et al., 2016; Navarro et al., 2008; Ngure et al., 2014; Ogola et al., 2002). Therefore, past literatures form the justification of this study. The experimental results of this study confirm the presence of these pollutants, especially the toxic heavy metals which are of much priority. Furthermore, PCA and LDA of the water samples from ex-mining ponds and lakes justify the parameter selection. Similarly, the carcinogenic and non-carcinogenic risk evaluations in this study supported the preference of the selected heavy metals. PCA was also employed in the parameter selection processes for the development of WQI in lakes and rivers in different countries (Gao et al., 2016; Hou et al., 2016; Yidana & Yidana, 2010).

The parameters selected for the development of WQI based on metals are As, Cd, and Pb, while physical-chemical parameter WQI include pH, DO, BOD, and AN. These selected parameters will reflect the general water quality for the intended use, thereby reducing the subjectivity in the parameter selection process, mostly done by experts in the development of the pioneer and popular existing indices (Abbasi & Abbasi, 2012; Cude, 2001). Similarly, it was reported that some participants did not respond and choose among the set of the proposed parameters for the Marine WQI development

(MERC, 2012). Principal component analysis was therefore employed to reduce the subjectivity in the parameter selection processes.

4.4.2 Weightage Assignment (Wi)

Assigning weightage to the selected parameters is an important stage in developing WQI as it entails assigning a significance or priority in the form of a numerical value to the individual parameters in accordance with the contribution to the overall water quality (Gupta et al., 2003; Lumb et al., 2011; Prasad et al., 2014). The standard permissible values of the parameters in Malaysian water quality standard INWQS were used to assign the weightages that added up to 1. Table 4.20 and Table 4.21 show the selected parameters and the allocated weightages respectively for metals and physical-chemical parameters. The weightage equation is given by equation 4.6;

$$W = \frac{K}{S} \quad \textbf{(4.6)}$$

where W is the weightage, K is proportionality constant, and S is the parameter standard permissible value (Bhutiani et al., 2016; Jha et al., 2015). Equation 4.6 can be rewritten as follows;

$$K = \frac{1}{\sum_{1}^{n}\frac{1}{S}} \quad \textbf{(4.7)}$$

Substituting the values in equation 4.7 gives K value for the metals and physical-chemical parameters respectively in equations 4.8 and 4.9;

$$K = 0.002205 \quad \textbf{(4.8)}$$

$$K = 0.250984 \quad \textbf{(4.9)}$$

Table 4.20: Weighting factor of the selected heavy metals parameters

Parameter	Standard Values (INWQS, MOH)	Relative Weight	Assigned Weighting Factor
As	0.01	100	0.22
Cd	0.003	333	0.74
Pb	0.05	20	0.04
Total		**453**	**1**

Table 4.21: Weighting factor of the selected physical-chemical parameters

Parameter	Standard Values (INWQS, MOH)	Relative Weight	Assigned Weighting Factor
pH	8.5	0.117	0.03
AN	0.3	3.33	0.84
DO	5	0.2	0.05
BOD	3	0.33	0.08
Total		**3.98**	**1**

From Table 4.20, it can be inferred that toxic heavy metals associated with mining have a relative weight of 453 which is by far greater than 3.98 for the physical-chemical parameters as shown in Table 4.21. The assigned weightages factors of the metal pollutants are 0.74, 0.22, and 0.04 for Cd, As, and Pb respectively. This agrees with levels of the selected metal pollutants in this study (Figure 4.1), and their maximum permissible limits in drinking water. Besides the very high levels of Cd in four of the ex-mining ponds (TPPA, TPPB, TPRP, and TT) and lake TPJ, Cd has the least maximum permissible limit in drinking water of 3 µg/L (MOH, 2004). AN has the highest weightage of 0.83 in the physical-chemical parameter index, this agrees with the levels of domestic sewage discharge into the ex-mining ponds and lakes. AN is toxic to humans, animals, and plants and therefore not desirable in drinking water (Dahlberg et al., 2016; Willingham et al., 2016). This suggests the suitability of the indices in assessing the quality of water from ex-mining ponds that is predominantly loaded with toxic heavy metals (Ashraf et al., 2011b; Atanacković et al., 2013; Ning et al., 2011). The chemometric study also reveals that larger variation in ex-mining ponds is associated with the toxic heavy metals. Similarly, As, Pb, and Fe were reported as the most significant parameters in PC1 due to mining (Bhuiyan et al., 2010). As reported in this study, water samples analyzed from the ex-mining ponds have very low total suspended solid and are not turbid, but with high concentration of dissolved organic matter resulting in a clean and clear physical appearance. This justifies the low relative weight associated with the physical-chemical parameters.

4.4.3 Sub Index/Quality Rating (Qi)

Sub-indices transform different units of the water quality parameters into a common scale (Saeedi et al., 2010). The quality rating of each selected parameter can be achieved either by using mathematical equations or rating curves formulated by opinion of the selected water quality experts. Several researchers pointed the highly subjective nature of the rating curves (Abbasi & Abbasi, 2012; MERC, 2012; Nasirian, 2007). Similarly, Boyacioglu (2007) reported the use of sub-index rating based on the personal opinion of 14 water quality experts in the formulation of the universal water quality index. However, quality rating using observed experimental values (Primary data) was applied to obtain the sub-indices in this study. It is the ratio of the parameter concentration in the water sample to the standard permissible value as given in equation 4.10 and 4.11 below (Vasanthavigar et al., 2010; Wanda et al., 2016; Yidana & Yidana, 2010). This is more acceptable considering that water from the ex-mining ponds is proposed for human consumption.

$$Q_i = \frac{C_i}{S_i} \times 100 \quad \textbf{(4.10)}$$

$$Q_{pH,\, DO} = \frac{C_i - V_i}{S_i - V_i} \times 100 \quad \textbf{(4.11)}$$

In equation 4.10 and 4.11, C_i is the parameter concentration in mg/L, S_i is the standard parameter value in INWQS, V_i is the ideal value which is considered as 7.0 for pH, and 14.6 for DO. The sub-index is further computed using equation 4.12 (Lumb et al., 2011; Vasanthavigar et al., 2010);

$$SI_i = W_i \times Q_i \quad \textbf{(4.12)}$$

The sub-indices of the selected parameters in ex-mining ponds and lakes are shown in Tables 4.23 to 4.26.

4.4.4 Development of Water Quality Index

The WQI is computed by summing up the sub-indices of the individual parameters to obtain the overall index as shown in equation 4.13;

$$WQI = \sum SI_i \quad \textbf{(4.13)}$$

where SI is the sub-index of the individual parameter. The WQI of the ex-mining ponds and lakes with respect to heavy metals (WQI_{HM}) is calculated using equation 4.14 below;

$$WQI_{HM} = 0.74{*}SICd + 0.22{*}SIAs + 0.04{*}SIPb \quad \textbf{(4.14)}$$

where SIAs is the sub index for As, SICd is sub index for Cd, and SIPb is sub index for Pb.

The WQI for the physical-chemical parameters (WQI_{PC}) is calculated using equation 4.15 below;

$$WQI_{PC} = 0.03*SIpH + 0.84*SIAN + 0.08*SIBOD + 0.05*SIDO \qquad \textbf{(4.15)}$$

where SIpH is sub index for pH, SIAN is sub index for AN, SIBOD is sub index for BOD, SIDO is sub index for DO.

The WQI are classified into five categories as shown in Table 4.22. The classification is based on decreasing scale in which the water quality increases with decreasing WQI (Yidana & Yidana, 2010). The results of the classification reveal a significant difference in the WQI_{HM} values among the sites studied in Klang valley ($p < 0.05$). The ex-mining ponds (TPPA, TPPB, TPRP and TT) and lake TPJ are very polluted and are rated poor with WQI_{HM} of 302 – 557 and 315 respectively (Table 4.23). The water is classified as not suitable for human consumption due to high metal loadings. Nevertheless, all other ex-mining ponds and lakes in Klang Valley, having metal pollutants below INWQS limits are rated excellent with WQI_{HM} of 1.4 – 33 as displayed in Figure 4.23. Table 4.24 shows physical-chemical parameter rating for all the ex-mining ponds and lakes as Good to Excellent except TKJB, TTJA, and TS7 with WQI_{PC} values of 120, 118, and 163 respectively. These are sites with high organic matter pollution from residences and other anthropogenic inputs. The dissimilarity in WQI_{PC} among the studied sites also confirms differences in the pollution sources and influence of external or anthropogenic input. Therefore, WQI_{PC} helps to maintain a good water quality of lakes and ex-mining ponds. Conversely, no significant difference in the WQI values of ex-mining ponds and lakes in Melaka. Tables 4.25 and 4.26 show

WQI_{HM} and WQI_{PC} as excellent from 4.0 – 25 and 15 – 89 respectively, this rating qualifies the water for human consumption after proper conventional water treatment processes (Figure 4.25 and Figure 4.26).

The heavy metal pollutants in the ex-mining ponds originated from the previous mining operation (Hogarh et al., 2016; Thienpont et al., 2016), and the influence of anthropogenic pollutants was also considered one of the major threat to water quality in lakes (Liou et al., 2004; Poerschmann et al., 2017). In a similar study, WQI was applied in the assessment of overall water quality for dinking purposes in which Cd, Cu, Pb, and Zn were found to be within the acceptable limit and safe for human consumption (Mohan et al., 1996). However, a seasonal pollution of water with high levels of Cd, Ni, and Pb was reported in river Soan Pakistan (Nazeer et al., 2014). The WQI_{HM} was found to be relatively acceptable for drinking purposes during pre-monsoon season only.

It was observed that As, and Cd are the major metal contaminants in the ex-mining ponds and lakes classified as very polluted, nonetheless, the studied sites with very low metal concentrations are classified excellent. WQI_{HM} is therefore a priority in consideration of the water for human consumption. If the WQI_{HM} fails, the water cannot be used for drinking until it undergoes sufficient metal removal processes. However, the water can be considered for drinking after necessary treatment processes.

Table 4.22: Water Quality classification (Sahu & Sikdar, 2008; Yidana & Yidana, 2010).

Range	Water Quality Rating
< 50	Excellent water
50 – 100	Good water
100 – 200	Poor water
200 – 300	Very poor water
> 300	Water unsuitable for drinking purposes

Note: All the water class can be used for recreational activities

Table 4.23: Sub-indices of the selected heavy metals and WQI_{HM} of ex-mining ponds and lakes in Klang valley

Station	As	Cd	Pb	WQI_{HM}	Status
TPPA	24.9278	316.05	0.396	341.3738	Very Polluted
TPPB	255.78	299.635	0.3168	555.7318	Very Polluted
TPRP	92.61	298.655	0.3256	391.5906	Very Polluted
TSP	1.47735	27.93	1.188	30.59535	Excellent
TT	4.69665	297.675	0.30888	302.6805	Very Polluted
TKJA	16.5375	0.735	0.02024	17.29274	Excellent
TKJB	14.994	2.205	0.06952	17.26852	Excellent
TB	1.16865	0.245	0.0044	1.41805	Excellent
TTJA	2.49165	8.085	0.04576	10.62241	Excellent
TP	8.379	1.47	0.03168	9.88068	Excellent
TSA	7.43085	0.49	0.0044	7.92525	Excellent
TS7	31.752	1.225	0.05808	33.03508	Excellent
TPJ	16.5375	298.165	0.3872	315.0897	Very Polluted

Table 4.24: Sub-indices of the selected physical-chemical parameters and WQI_{PC} of ex-mining ponds and lakes in Klang Valley

Station	DO	BOD	pH	AN	WQI_{PC}	Status
TPPA	6.12318	22.13333	2.081176	69.66667	100.0044	Good
TPPB	0.381444	7.746667	3.411765	44.58667	56.12654	Good
TPRP	0.471786	7.663667	2.807882	52.94667	63.89	Good
TSP	5.21976	9.406667	1.774118	25.08	41.48054	Excellent
TT	4.125618	4.15	2.814706	13.93333	25.02366	Excellent
TKJA	0.913458	16.32333	0.317294	44.58667	62.14075	Good
TKJB	1.35513	26.83667	0.460588	91.96	120.6124	Poor
TB	4.21596	6.916667	1.432941	16.72	29.28557	Excellent
TTJA	3.31254	20.75	2.695294	91.96	118.7178	Poor
TP	3.61368	9.794	2.838588	64.09333	80.3396	Good
TSA	2.81064	24.485	2.896588	5.573333	35.76556	Good
TS7	1.335054	22.68667	2.592941	136.5467	163.1613	Poor
TPJ	0.038482	5.81	2.9	44.58667	53.33515	Good

Table 4.25: Sub-indices of the selected heavy metals and WQI_{HM} of ex-mining ponds and lakes in Melaka

Station	As	Cd	Pb	WQI_{HM}	Status
TBC1	20.4624	2.45	0.0088	22.9212	Excellent
TBC2	14.08995	2.45	0.0088	16.54875	Excellent
TBC3	14.97195	2.45	0.0088	17.43075	Excellent
TR	5.7771	2.45	0.0088	8.2359	Excellent
JD	1.5876	2.45	0.0088	4.0464	Excellent
TDT	2.8224	2.45	0.0088	5.2812	Excellent
TTB	23.1084	2.45	0.0088	25.5672	Excellent
TBK	10.29735	2.45	0.0088	12.75615	Excellent

Table 4.26: Sub-indices of the selected physical-chemical parameters and WQI_{PC} of ex-mining ponds and lakes in Melaka

Station	DO	BOD	pH	AN	WQI_{PC}	Status
TBC1	5.14	2.49	2.470118	5.573333	15.6734	Excellent
TBC2	6.11	3.181667	2.494	19.50667	31.2923	Excellent
TBC3	4.99	3.209333	2.391647	19.50667	30.0976	Excellent
TR	9.16	5.422667	3.507294	25.08	43.1699	Excellent
JD	4.89	3.458333	2.797647	78.02667	89.1726	Excellent
TDT	6.09	3.015667	2.449647	47.37333	58.9286	Excellent
TTB	4.56	5.865333	2.405294	8.36	21.1906	Excellent
TBK	3.67	4.15	2.347294	25.08	35.2472	Excellent

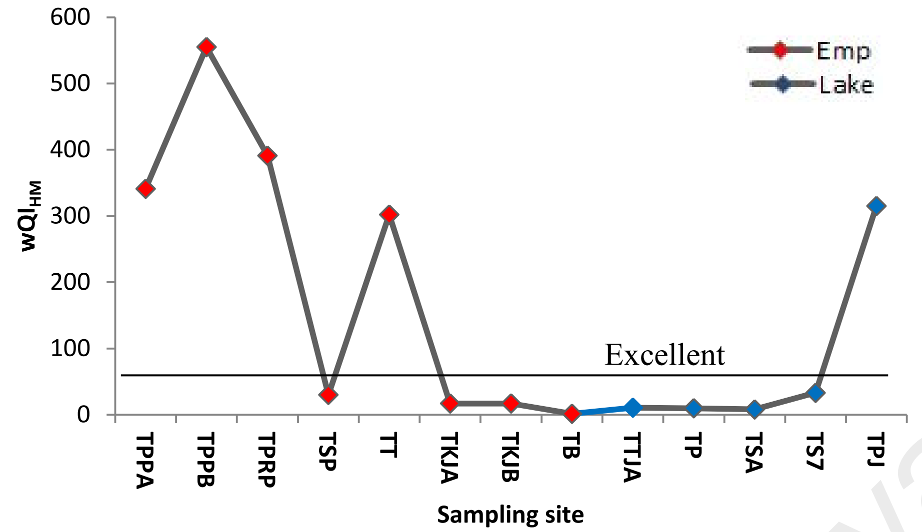

Figure 4.23: Heavy metals WQI_{HM} of ex-mining ponds and lakes in Klang Valley

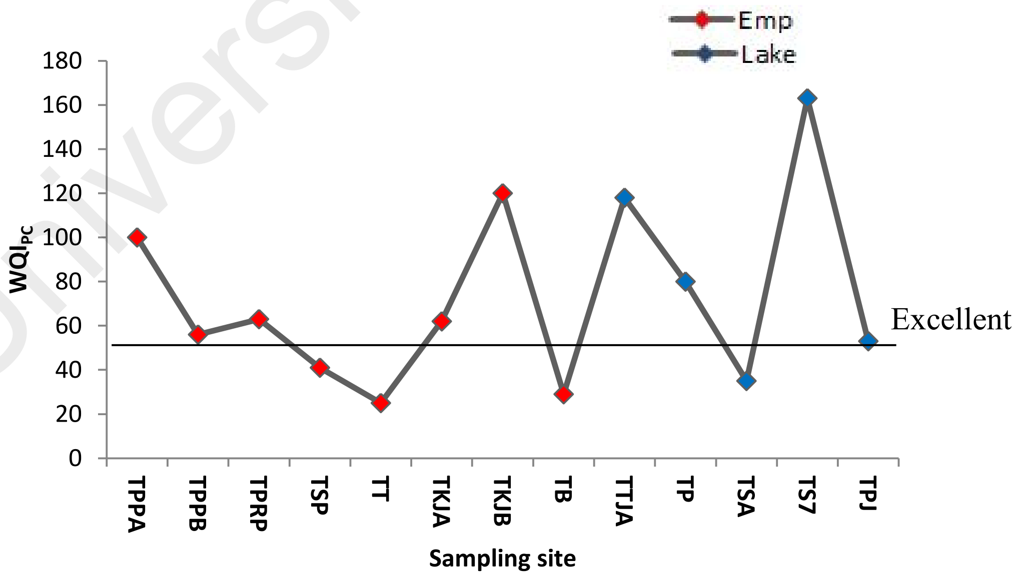

Figure 4.24: Physical-chemical parameter WQI_{PC} of ex-mining ponds and lakes in Klang Valley

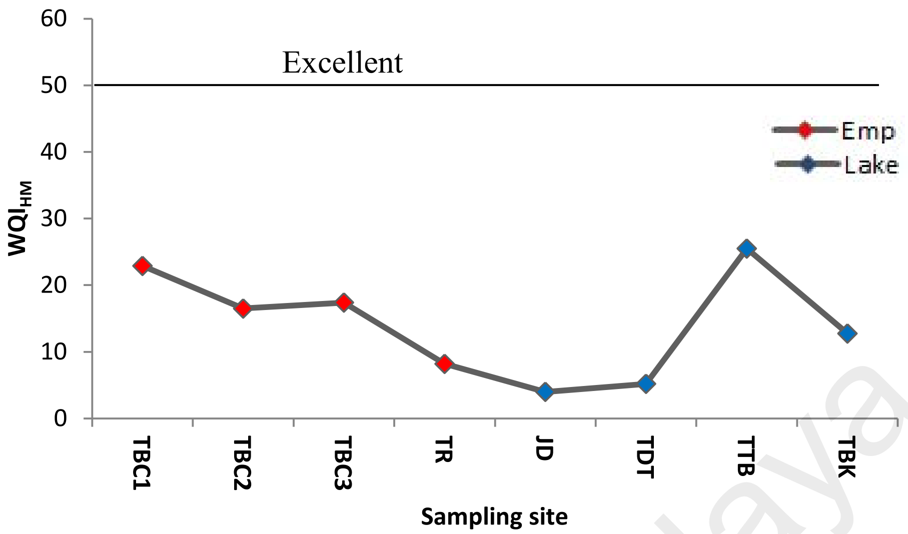

Figure 4.25: Heavy metals WQI_{HM} of ex-mining ponds and lakes in Melaka

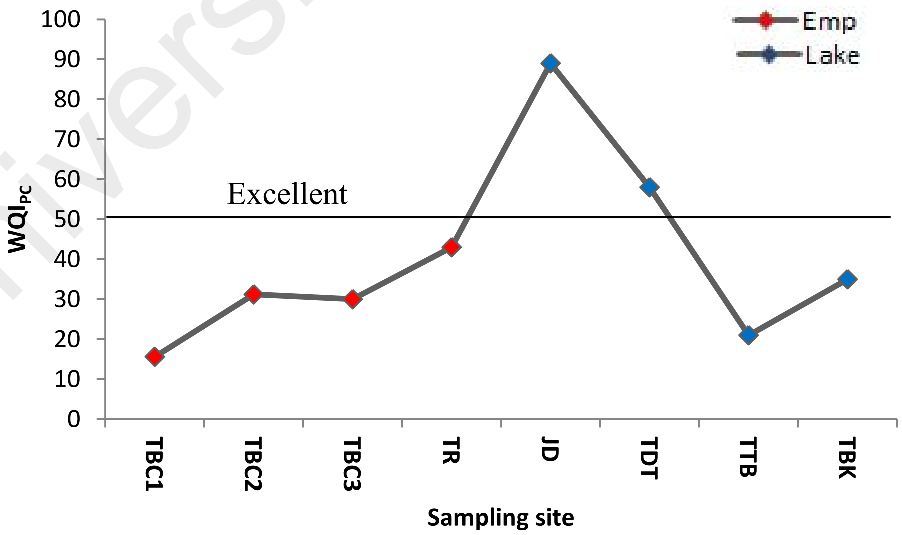

Figure 4.26: Physical-chemical parameter WQI_{PC} of ex-mining ponds and lakes in Melaka

4.4.5 Uncertainty in WQI

Several WQI have been formulated and efforts had been made to address the limitations observed. Recent WQI developed to assess the water quality of reservoirs in China reported a good rating from good to very poor water quality (Hou et al., 2016). However, uncertainty was not considered and suggestion was given to improve the model, thereby enhancing the consistency of water quality assessments (Azarnivand, 2017). All the WQI developed has advantages and limitations, but the limitations can be minimized by reducing the subjectivity (Lumb et al., 2011). Up till now no single WQI has been accepted globally, but some countries have used combined water quality data in their formulation. In this study, uncertainty was reduced to the barest minimum level by adopting established water quality guidelines (INWQS) separately for the heavy metals and physical-chemical parameters, and chemometric approaches. In a similar way, the statistical approaches improve the accuracy and reliability of the index (Salim et al., 2009).

CHAPTER 5: CONCLUSION

Water quality index of ex-mining ponds was developed in this study using Malaysian water quality standard as a reference in assigning weightage to the selected parameters. This approach employs the use of primary data and extensive literature comparisons in the parameter selection. Consideration was also given to the geology and anthropogenic inputs into the water bodies for the general water quality assessment. The chemometric and health risks evaluations were included to support the WQI development.

Significant variations are observed in the levels of heavy metals and physical chemical parameters in ex-mining ponds and lakes. This is attributed to the anthropogenic inputs especially in Klang valley where As and Cd exceeded the reference limit in most ex-mining ponds. High levels of these toxic metals could also be plausibly linked to previous mining activities. The levels of DO and BOD in most of the ex-mining ponds and lakes in Klang Valley points to anthropogenic activities around the studied sites. Melaka ex-mining ponds and lakes records low metal levels below INWQS which can be attributed to geological influence. The indicators of organic pollution are also within the recommended limits.

It is clearly shown in this study that PCA and FA identifies the parameters which contribute most to the variations in ex-mining ponds and lakes, but no general trend was observed due to differences in geological and anthropogenic influence. In the Klang Valley, the results revealed that heavy metals especially As, Cd and Pb record the highest variations in ex-mining ponds, while BOD and AN showed larger variations in most of the lakes. The observed variations in Melaka ex-mining ponds are due to Mg, Ca, EC and DO, while the lakes have Fe, Mn, pH and BOD as the major contributors to the variation. In addition, the results of the linear discriminant analysis (LDA) clearly distinguish ex-mining ponds and lakes in both Klang Valley and Melaka in which As, Mn and Ca are associated to ex-mining ponds, whereas Fe, Na and pH were the common parameters associated to the lakes. The hierarchical cluster analysis (HCA) could not achieve a distinct grouping of ex-mining ponds and lakes especially in Klang Valley due to anthropogenic impact. The land use in most of the ex-mining ponds and lakes significantly influenced the water quality, while two-way HCA for Melaka and Negeri Sembilan confirms low metal concentrations and absence of anthropogenic inputs. The land uses in the areas are predominantly rubber and palm oil plantation.

In the evaluation of health risk on exposure to As, Cd and Pb in water samples from ex-mining ponds and lakes, the non-carcinogenic risk reveals the threat of As only due to ingestion by both adult and children in the studied sites of Klang Valley, and children only in Melaka. However, the exposure by dermal contact is within tolerable limit. It is evident that As is the major pollutant in the ex-mining ponds which originates from the past mining operation. Considering the RME, As carcinogenic risk of ingestion by a Malaysian adult is very alarming in the studied sites of cluster 2 in Klang Valley. The carcinogenic risk level well exceeded the reference limit of 1×10^{-4}. It is therefore recommended to remove As from the ex-mining water to a tolerable level before utilization especially for drinking purposes. There could be non-carcinogenic health effects for child only as a result of exposure to water from ex-mining ponds and lakes in Melaka specifically due to As having HQ > 1, but no carcinogenic threat is seen in both adult and children. Dermal contact also might have no risk of health complication in all the sites studied; hence, the ex-mining ponds in the Klang Valley and Melaka may be used for recreational activities. Generally, the heavy metals of relevance in the carcinogenic and non-carcinogenic health risk associated with the ingestion of water from ex-mining ponds for both adult and child can be prioritized as As > Cd > Pb. This asserts the potential hazard related to the toxic heavy metals in water from ex-mining ponds.

Utilization of multivariate statistical analysis minimizes assumptions due to subjectivity, and improves the accuracy of the WQI. The heavy metal WQI_{HM} gives more weightage to the toxic heavy metals associated with mining operation. This emphasis is important due to the fact heavy metals of concern are not removed in the course of water treatment processes considering that ex-mining water were intended for human consumption. As and Cd are the dominant and most important parameters in the metal WQI_{HM}, hence ex-mining ponds in Puchong (TPPA, TPPB, and TPRP), Titiwangsa (TT) and Putrajaya lake (TPJ) recorded very poor water quality and as such cannot be utilized for drinking purposes but recreational body contact is relatively safe. Other ex-mining ponds and lakes studied in the Klang Valley, Melaka and Negeri Sembilan have heavy metal WQI_{HM} classification ranging from good to excellent, therefore may be utilized after proper treatment processes. Based on the physical-chemical parameter, WQI_{PC}, TKJB, TTJA, and TS7 in Klang Valley are of poor water quality, hence the need to undergo further purification before utilization.

REFERENCES

Abbasi, T., & Abbasi, S. A. (2012). *Water quality indices*. Amsterdam: Elsevier.

Abdullah, M., Tan, C. Y., Ramli, N., Yahaya, A., & Fitri, N. (2011). Modelling of PM10 concentration for industrialized area in Malaysia: A case study in Shah Alam. *Physics Procedia, 22*, 318-324.

Acheampong, M. A., Adiyiah, J., & Ansa, E. D. (2013). Physico-chemical characteristics of a gold mining tailings dam wastewater. *Journal Environmental Science and Engineering, 2*, 469-475.

Ahmad, S., & Jones, D. (2013). Investigating the Mining Heritage Significance for Kinta District, the Industrial Heritage Legacy of Malaysia. *Procedia-Social and Behavioral Sciences, 105*, 445-457.

Aini, M., Fakhrul-Razi, A., Mumtazah, O., & Chen, J. M. (2007). Malaysian households' drinking water practices: A case study. *The International Journal of Sustainable Development & World Ecology, 14*(5), 503-510.

Akcil, A., & Koldas, S. (2006). Acid mine drainage (AMD): causes, treatment and case studies. *Journal of Cleaner Production, 14*(12), 1139-1145.

Akinbile, C. O., Yusoff, M. S., Talib, S. H., Hasan, Z. A., Ismail, W. R., & Sansudin, U. (2013). Qualitative analysis and classification of surface water in Bukit Merah Reservoir in Malaysia. *Water Science and Technology: Water Supply, 13*(4), 1138-1145.

Al-Mamun, A., & Zainuddin, Z. (2013). Sustainable river water quality management in Malaysia. *IIUM Engineering Journal, 14*(1), 29-42.

Alam, L., Mokhtar, M. B., Alam, M. M., Bari, M. A., Kathijotes, N., Ta, G. C., & Ern, L. K. (2015). Seasonal Variation And Preliminary Risk Assessment Of Trace Element Pollution In Surface Water From Langat River, Malaysia. *International Journal of Applied Environmental Sciences, 10*(1), 19-40.

Alatas, S. M. (2011). Governance and freshwater in the greater Kuala Lumpur area/Klang Valley: success or failure? *Akademika, 81*(3), 95-102.

Alberto, W. D., del Pilar, D. M., Valeria, A. M., Fabiana, P. S., Cecilia, H. A., & de los Ángeles, B. M. a. (2001). Pattern Recognition Techniques for the Evaluation of Spatial and Temporal Variations in Water Quality. A Case Study:: Suquía River Basin (Córdoba–Argentina). *Water Research, 35*(12), 2881-2894.

Albu, M., Banks, D., & Nash, H. (2012). *Mineral and thermal groundwater resources.* London :Springer Science & Business Media.

Alkarkhi, A. F., Ahmad, A., & Easa, A. M. (2009). Assessment of surface water quality of selected estuaries of Malaysia: multivariate statistical techniques. *The Environmentalist, 29*(3), 255-262.

Alshaebi, F. Y., Yaacob, W. Z., Samsudin, A. R., & Alsabahi, E. (2009). Risk assessment at abandoned tin mine in Sungai Lembing, Pahang, Malaysia. *Electronic Journal of Geotechnical Engineering, 14*, 1-9.

Althuwaynee, O. F., Pradhan, B., & Lee, S. (2012). Application of an evidential belief function model in landslide susceptibility mapping. *Computers & Geosciences, 44*, 120-135.

Alves, R. I., Sampaio, C. F., Nadal, M., Schuhmacher, M., Domingo, J. L., & Segura-Muñoz, S. I. (2014). Metal concentrations in surface water and sediments from Pardo River, Brazil: human health risks. *Environmental Research, 133*, 149-155.

Ammann, A. A. (2007). Inductively coupled plasma mass spectrometry (ICP MS): a versatile tool. *Journal of Mass Spectrometry, 42*(4), 419-427.

Amneera, W., Najib, N., Yusof, R. M., & Ragunathan, S. (2013). Water quality index of Perlis River Malaysia. *International Journal on Civil and Environmental Engineering, 13*(2), 1-6.

Arnemo, J. M., Andersen, O., Stokke, S., Thomas, V. G., Krone, O., Pain, D. J., & Mateo, R. (2016). Health and Environmental Risks from Lead-based Ammunition: Science Versus Socio-Politics. *EcoHealth*, *13(4)*, 618-622.

Asami, T., Katayama, H., Torrey, J. R., Visvanathan, C., & Furumai, H. (2016). Evaluation of virus removal efficiency of coagulation-sedimentation and rapid sand filtration processes in a drinking water treatment plant in Bangkok, Thailand. *Water Research, 101*, 84-94.

Asante, K. A., Agusa, T., Subramanian, A., Ansa-Asare, O. D., Biney, C. A., & Tanabe, S. (2007). Contamination status of arsenic and other trace elements in drinking water and residents from Tarkwa, a historic mining township in Ghana. *Chemosphere, 66*(8), 1513-1522.

Ashraf, M., Maah, M., & Yusoff, I. (2012). Chemical speciation and potential mobility of heavy metals in the soil of former tin mining catchment. *The Scientific World Journal, 2012*.

Ashraf, M. A., Maah, M., & Yusoff, I. (2011a). Assessment of heavy metals in the fish samples of mined out ponds Bestari Jaya, Peninsular Malaysia. *Proceedings of the Indian National Science Academy, 77*(1), 57-67.

Ashraf, M. A., Maah, M. J., & Yusoff, I. (2011b). Analysis of physio-chemical parameters and distribution of heavy metals in soil and water of ex-mining area of Bestari Jaya, Peninsular Malaysia. *Asian Journal of Chemistry, 23*(8), 3493.

Ashraf, M. A., Maah, M. J., & Yusoff, I. B. (2010). Study of water quality and heavy metals in soil & water of ex-mining area Bestari Jaya, peninsular Malaysia. *International Journal of Basic and Applied Sciences, 10*(3), 7-27.

Ashraf, M. A., Sarfraz, M., Naureen, R., & Gharibreza, M. (2015). Mining and Economic Development: Mining and Environmental Problems *Environmental Impacts of Metallic Elements*. Singapore: Springer.

Assubaie, F. N. (2015). Assessment of the levels of some heavy metals in water in Alahsa Oasis farms, Saudi Arabia, with analysis by atomic absorption spectrophotometry. *Arabian Journal of Chemistry, 8*(2), 240-245.

Atanacković, N., Dragišić, V., Stojković, J., Papić, P., & Živanović, V. (2013). Hydrochemical characteristics of mine waters from abandoned mining sites in Serbia and their impact on surface water quality. *Environmental Science and Pollution Research, 20*(11), 7615-7626.

ATSDR. (1993). *Toxicological profile for Arsenic*, TP-92/02, Agency for toxic substances and disease registry. Atlanta, GA: EPA.

ATSDR. (2007). *Toxicological profile for arsenic.* Agency for Toxic Substances and Disease Registry, Division of Toxicology. Atlanta, GA: EPA.

Ayekoe, C. Y., Robert, D., & Lanciné, D. G. (2017). Combination of coagulation-flocculation and heterogeneous photocatalysis for improving the removal of humic substances in real treated water from Agbô River (Ivory-Coast). *Catalysis Today, 281*, 2-13.

Azarnivand, A. (2017). Comment on “Assessing water quality of five typical reservoirs in lower reaches of Yellow River, China: Using a water quality index method” by Wei Hou, Shaohua Sun, Mingquan Wang, Xiang Li, Nuo Zhang, Xiaodong Xin, Li Sun, Wei Li, and Ruibao Jia (2016)[Ecological Indicators, 61, 309–316]. *Ecological Indicators, 75*, 8-9.

Azmi, M., Junidah, R., Mariam, A. S., Safiah, M., Fatimah, S., Norimah, A., ... Abdul-Manan, W. (2009). Body mass index (BMI) of adults: findings of the Malaysian Adult Nutrition Survey (MANS). *Malaysian Journal of Nutrition, 15*(2), 97-119.

Azzlan, A., Cheong, C. B., & Lau, K. L. (2016). Sustainable water management for water supply efficiency: A case study at Syarikat Air Melaka Berhad. *Journal of Technology Management and Business, 3*(1), 1-21.

Babayemi, J. O., Ogundiran, M. B., & Osibanjo, O. (2016). Overview of Environmental Hazards and Health Effects of Pollution in Developing Countries: A Case Study of Nigeria. *Environmental Quality Management, 26*(1), 51-71.

Balamurugan, G. (1991). Tin mining and sediment supply in Peninsular Malaysia with special reference to the Kelang River basin. *Environmentalist, 11*(4), 281-291.

Baltrusaitis, J., & Sviklas, A. M. (2016). From insoluble minerals to liquid fertilizers: magnesite as a source of magnesium (Mg) nutrient. *ACS Sustainable Chemistry & Engineering, 4*(10), 5404-5408.

Ban, M. L., Guo, S. Q., Yu, R., & Huang, R. Z. (2015). Environmental Health Risk Assessment of Underground Water in Mining Area of Nandan County. *Advanced Materials Research*. (1092), 673-677.

Banks, D., Younger, P. L., Arnesen, R. T., Iversen, E. R., & Banks, S. B. (1997). Mine-water chemistry: the good, the bad and the ugly. *Environmental Geology, 32*(3), 157-174.

Bao, K. S.., Shen, J., Wang, G. P., & Tserenpil, S. (2016). Anthropogenic, detritic and atmospheric soil-derived sources of lead in an alpine poor fen in northeast China. *Journal of Mountain Science*, *13*(2), 255-264.

Bardsley, P. (2000). Project Initiation Document – *Greenside Mines, Glenridding.* Penrith, England: Published by the Environment Agency.

Barregard, L., Sallsten, G., Fagerberg, B., Borné, Y., Persson, M., Hedblad, B., & Engström, G. (2016). Blood cadmium levels and incident cardiovascular events during follow-up in a population-based cohort of Swedish adults: the Malmö Diet and Cancer Study. *Environmental Health Perspectives, 124*(5), 594-600.

Beattie, L. (1996). *A study of the water and sediment chemistry of key Cumbrian lakes*, Ullswater. Penrith, England: Published by the Environment Agency.

Bebbington, A., & Williams, M. (2008). Water and mining conflicts in Peru. *Mountain Research and Development*, *28*(3), 190-195.

Beck, M. B. (1987). Water quality modeling: a review of the analysis of uncertainty. *Water Resources Research*, *23*(8), 1393-1442.

Béjaoui, I., Kolsi-Benzina, N., Sappin-Didier, V., & Munoz, M. (2016). Health Risk Assessment in Calcareous Agricultural Soils Contaminated by Metallic Mining Activity Under Mediterranean Climate. *CLEAN–Soil, Air, Water, 44(10),* 1385-1395.

Bello, O., Naidu, R., Rahman, M. M., Liu, Y., & Dong, Z. (2016). Lead concentration in the blood of the general population living near a lead–zinc mine site, Nigeria: Exposure pathways. *Science of the Total Environment, 542*, 908-914.

Berger, A. C., Bethke, C. M., & Krumhansl, J. L. (2000). A process model of natural attenuation in drainage from a historic mining district. *Applied Geochemistry, 15*(5), 655-666.

Bernama. (10^{th} May, 2014). Stop using water from mining pools, warns MMA *The Malaysian Times*. Retrieved from http://www.themalaysiantimes.com.my/ on 21^{st} April, 2017.

Bernama. (29^{th} April, 2014). Water from former mining pools safe, assures Selangor gov't. *The Malaysia Online*. Retrieved from http://www.themalaysiantimes on 21^{st} April, 2017.

Bernard, A. (2008). Cadmium & its adverse effects on human health. *Indian Journal of Medical Research*, *128*(4), 557-564.

Beyer, W. N., Basta, N. T., Chaney, R. L., Henry, P. F., Mosby, D. E., Rattner, B. A., . . . Weber, J. (2016). Bioaccessibility tests accurately estimate bioavailability of lead to quail. *Environmental Toxicology and Chemistry, 35(9),* 2311-2319.

Bhuiyan, M. A., Parvez, L., Islam, M., Dampare, S. B., & Suzuki, S. (2010). Heavy metal pollution of coal mine-affected agricultural soils in the northern part of Bangladesh. *Journal of Hazardous Materials, 173*(1), 384-392.

Bhutiani, R., Khanna, D., Kulkarni, D. B., & Ruhela, M. (2016). Assessment of Ganga river ecosystem at Haridwar, Uttarakhand, India with reference to water quality indices. *Applied Water Science, 6*(2), 107-113.

Bilotta, G., & Brazier, R. (2008). Understanding the influence of suspended solids on water quality and aquatic biota. *Water Research, 42*(12), 2849-2861.

Bissen, M., & Frimmel, F. H. (2003). Arsenic—a review. Part I: occurrence, toxicity, speciation, mobility. *Acta Hydrochimica et Hydrobiologica, 31*(1), 9-18.

Bonn, B. Y., Shariff, A. A., Mohamed, A. M., & Merican, A. F. (2011). (*Body Mass Index of Malaysian School Children and Adolescents*). Paper presented at the International Conference on Mathematics and Sciences, 12^{th} - 13^{th} Ocober, 2011. Surabaya, Indonesia. p. 1-8.

Bosch, A. C., O'Neill, B., Sigge, G. O., Kerwath, S. E., & Hoffman, L. C. (2016). Heavy metal accumulation and toxicity in smoothhound (Mustelus mustelus) shark from Langebaan Lagoon, South Africa. *Food Chemistry, 190*, 871-878.

Bouchard, M. F., Sauvé, S., Barbeau, B., Legrand, M., Brodeur, M. È., Bouffard, T., . . . Mergler, D. (2011). Intellectual impairment in school-age children exposed to manganese from drinking water. *Environmental Health Perspectives, 119*(1), 138-143.

Bouguerne, A., Boudoukha, A., Benkhaled, A., & Mebarkia, A.-H. (2016). Assessment of surface water quality of Ain Zada dam (Algeria) using multivariate statistical techniques. *International Journal of River Basin Management*, 1-11.

Bowell, R. J., Alpers, C. N., Jamieson, H. E., Nordstrom, D. K., & Majzlan, J. (2014). The Environmental Geochemistry of Arsenic—An Overview—. *Reviews in Mineralogy and Geochemistry, 79*(1), 1-16.

Boyacioglu, H. (2007). Development of a water quality index based on a European classification scheme. *Water Sa, 33*(1), 101-106.

Boyd, C. E. (2015). *Water quality: an introduction*. Boston: Springer.

Breitburg, D. L., Loher, T., Pacey, C. A., & Gerstein, A. (1997). Varying effects of low dissolved oxygen on trophic interactions in an estuarine food web. *Ecological Monographs, 67*(4), 489-507.

Bui, A. T., Nguyen, H. T., Nguyen, M. N., Tran, T. H. T., Vu, T. V., Nguyen, C. H., & Reynolds, H. L. (2016). Accumulation and potential health risks of cadmium, lead and arsenic in vegetables grown near mining sites in Northern Vietnam. *Environmental Monitoring and Assessment, 188*(525), 1-11.

Buytaert, W., Dewulf, A., De Bièvre, B., Clark, J., & Hannah, D. M. (2016). Citizen Science for Water Resources Management: Toward Polycentric Monitoring and Governance? *Journal of Water Resources Planning and Management, 142*(4), 01816002-1-4.

Canfield, R. L., Henderson Jr, C. R., Cory-Slechta, D. A., Cox, C., Jusko, T. A., & Lanphear, B. P. (2003). Intellectual impairment in children with blood lead concentrations below 10 µg per deciliter. *New England Journal of Medicine, 348*(16), 1517-1526.

Cánovas, C., Hubbard, C., Olías, M., Nieto, J., Black, S., & Coleman, M. L. (2008). Hydrochemical variations and contaminant load in the Río Tinto (Spain) during flood events. *Journal of Hydrology, 350*(1), 25-40.

Cao, S., Duan, X., Zhao, X., Ma, J., Dong, T., Huang, N., . . . Wei, F. (2014). Health risks from the exposure of children to As, Se, Pb and other heavy metals near the largest coking plant in China. *Science of the Total Environment, 472*, 1001-1009.

Cao, S., Duan, X., Zhao, X., Wang, B., Ma, J., Fan, D., . . . Jiang, G. (2015). Health risk assessment of various metal (loid) s via multiple exposure pathways on children living near a typical lead-acid battery plant, China. *Environmental Pollution, 200*, 16-23.

Caraballo, M. A., Macías, F., Nieto, J. M., & Ayora, C. (2016). Long term fluctuations of groundwater mine pollution in a sulfide mining district with dry Mediterranean climate: Implications for water resources management and remediation. *Science of the Total Environment, 539*, 427-435.

Caruso, B. S., Mirtskhulava, M., Wireman, M., Schroeder, W., Kornilovich, B., & Griffin, S. (2012). Effects of Manganese Mining on Water Quality in the Caucasus Mountains, Republic of Georgia. *Mine Water and the Environment, 31*(1), 16-28.

CCME. (2001). Canadian council of ministers of environment . *Canadian water quality guidelines for the protection of aquatic life:* CCME water quality index 1.0, user's manual. In: Canadian environmental quality guidelines, 1999. Winnipeg.

CDC. (2002). *Managing Elevated Blood Lead Levels Among Young Children*: Recommendations from the Advisory Committee on Childhood Lead Poisoning Prevention. Atlanta, GA.

CDCP. (2017). *Low Level Lead Exposure Harms Children*: A Renewed Call for Primary Prevention. Centers for Disease Control Prevention. Georgia.

Chabukdhara, M., & Nema, A. K. (2013). Heavy metals assessment in urban soil around industrial clusters in Ghaziabad, India: probabilistic health risk approach. *Ecotoxicology and Environmental Safety, 87*, 57-64.

Chang, C., Ghani, A., Zakaria, N., Yusof, M., Ayub, K., Lai, S., . . . Ainan, A. (2008). (*Rehabilitation of Ex-Mining Pond and Existing Wetland for Integrated Stormwater Management*). Proceedings of the 11th International Conference on Urban Drainage. 31st August - 5th September, 2008. Edinburgh, Scotland. p 1-10.

Charles, S. (12th June, 2014). Make findings on heavy metal In Bestari Jaya, Selangor public or face legal action! Member of Parliament for Klang. *Online news.* Retrieved from http://charlessantiago.org/2014/06/12/ on 8th March 2015.

Chen, L., Liang, R., Tan, T., Ai, X., Hao, T., Chen, X., & Wang, Y. (2016). Recent development in arsenic speciation and toxicity reduction of inorganic arsenic in food. *European Journal of BioMedical Research, 2*(1), 27-31.

Chen, Y., Wu, P., Shao, Y., & Ying, Y. (2014). Health risk assessment of heavy metals in vegetables grown around battery production area. *Scientia Agricola, 71*(2), 126-132.

Chigor, V. N., Umoh, V. J., Okuofu, C. A., Ameh, J. B., Igbinosa, E. O., & Okoh, A. I. (2012). Water quality assessment: surface water sources used for drinking and irrigation in Zaria, Nigeria are a public health hazard. *Environmental Monitoring and Assessment, 184*(5), 3389-3400.

Chow, M., Shiah, F., Lai, C., Kuo, H., Wang, K., Lin, C., . . . Ko, C. (2016). Evaluation of surface water quality using multivariate statistical techniques: a case study of Fei-Tsui Reservoir basin, Taiwan. *Environmental Earth Sciences, 75*(1), 1-15.

Chowdhury, S., Mazumder, M. J., Al-Attas, O., & Husain, T. (2016). Heavy metals in drinking water: Occurrences, implications, and future needs in developing countries. *Science of the Total Environment, 569*, 476-488.

Ciesielski, T., Weuve, J., Bellinger, D. C., Schwartz, J., Lanphear, B., & Wright, R. O. (2012). Cadmium exposure and neurodevelopmental outcomes in US children. *Environmental Health Perspectives, 120*(5), 758.

Cobb, A. (2007). *Cadmium*: New York: Marshall Cavendish/Benchmark.

Courtenay, P. P. (1981). The plantation in Malaysian economic development. *Journal of Southeast Asian Studies, 12*(2), 329-348.

Cude, C. G. (2001). Oregon water quality index a tool for evaluating water quality management effectiveness. *Journal of the American Water Resources Association, 37*(1), 125-137.

d'Halluin, M., Rull-Barrull, J., Bretel, G., Labrugère, C., Le Grognec, E., & Felpin, F.-X. (2017). Chemically modified cellulose filter paper for heavy metal remediation in water. *ACS Sustainable Chemistry & Engineering, 5(2),* 1965-1973.

Dahlberg, D., Ivanovic, J., & Hassel, B. (2016). Toxic levels of ammonia in human brain abscess. *Journal of Neurosurgery, 124*(3), 854-860.

Daniel, R., & Kawasaki, N. (2016). The Distribution of Heavy Metals and Nutrients along Selangor River and its Adjacent Mining Ponds, Malaysia. *International Journal of Advances in Agricultural & Environmental Engineering, 3*(2), 241-244.

Davis, J. M., Elias, R. W., & Grant, L. (1992). Current issues in human lead exposure and regulation of lead. *Neurotoxicology, 14*(2-3), 15-27.

Davison, W. (1993). Iron and manganese in lakes. *Earth-Science Reviews, 34*(2), 119-163.

Deliyanni, E. A., Kyzas, G. Z., & Matis, K. A. (2017). Various flotation techniques for metal ions removal. *Journal of Molecular Liquids, 225*, 260-264.

DMG. (2003). *Malaysia minerals yearbook 2002*. Department of Minerals and Geosciences Kuala Lumpur: Geological Survey Department.

DOE. (2012). *Department of Environment. Malaysia: Environmental Quality Act report,* Ministry of Science, Technology and the Environment, Putrajaya, Malaysia.

Dold, B. (2005). *Basic concepts of environmental geochemistry of sulfide mine-waste management*. XXIV Curso Latinoamericano de Metalogenia UNESCO-SEG Mineralogia, geoquimica y geomicrobiologia para el mentejo ambiental de desechos mineros, Lima, Peru: Intech Publishers.

Donovan, G. H., Jovan, S. E., Gatziolis, D., Burstyn, I., Michael, Y. L., & Monleon, V. J. (2016). Using an epiphytic moss to identify previously unknown sources of atmospheric cadmium pollution. *Science of the Total Environment, 559*, 84-93.

DOS. (2016). *Malaysians life expectency.* Malaysia: Department of Statistics.

Dzulfakar, M. A., Shaharuddin, M. S., Muhaimin, A. A., & Syazwan, A. I. (2011). Risk assessment of aluminum in drinking water between two residential areas. *Water, 3*(3), 882-893.

Edwin, L., & Thomas, S. N. (2001). *Wood preservation for marine application*. India : India Council for Agricultural Research.

Eileen, N. (12th June, 2014). DAP MP gives Selangor 2 weeks to reveal study on hazards in old mining pools. *The Malaysian Insider*. Retrieved from https://news.malaysianreview.com/6357/dap-mp on 16th May, 2015.

El Khalil, H., El Hamiani, O., Bitton, G., Ouazzani, N., & Boularbah, A. (2008). Heavy metal contamination from mining sites in South Morocco: monitoring metal content and toxicity of soil runoff and groundwater. *Environmental Monitoring and Assessment, 136*(1-3), 147-160.

Eveleth, P. B., & Tanner, J. M. (1976). *Worldwide variation in human growth*. CUP Archive. New York: Cambridge University Press.

Ezekwe, I., Odu, N., Chima, G., & Opigo, A. (2012). Assessing regional groundwater quality and its health implications in the Lokpaukwu, Lekwesi and Ishiagu mining areas of southeastern Nigeria using factor analysis. *Environmental Earth Sciences, 67*(4), 971-986.

Fadiran, A., Dlamini, C., & Thwala, J. (2014). Environmental assessment of acid mine drainage pollution on surface water bodies around Ngwenya mine, Swaziland. *Journal of Environmental Protection, 5*(2), 164-173.

Fanfani, L., Zuddas, P., & Chessa, A. (1997). Heavy metals speciation analysis as a tool for studying mine tailings weathering. *Journal of Geochemical Exploration, 58*(2-3), 241-248.

Fardushe, R. S., Hoque, M. M., & Roy, S. (2016). Assessment of soil and water quality of Barapukuria coal mining site, Dinajpur, Bangladesh. *Bangladesh Journal of Scientific Research, 27*(1), 63-73.

Farnham, I. M., Singh, A. K., Stetzenbach, K. J., & Johannesson, K. H. (2002). Treatment of nondetects in multivariate analysis of groundwater geochemistry data. *Chemometrics and Intelligent Laboratory Systems*, *60*(1), 265-281.

Favas, P. J., Pratas, J., Gomes, M. E. P., & Cala, V. (2011). Selective chemical extraction of heavy metals in tailings and soils contaminated by mining activity: environmental implications. *Journal of Geochemical Exploration, 111*(3), 160-171.

Fields, S. (2003). The earth's open wounds: abandoned and orphaned mines. *Environmental Health Perspectives, 111*(3), A154.

Finkelstein, M. M., & Verma, D. K. (2001). Exposure estimation in the presence of nondetectable values: another look. *AIHAJ-American Industrial Hygiene Association*, *62*(2), 195-198.

Flemming, C., & Trevors, J. (1989). Copper toxicity and chemistry in the environment: a review. *Water, Air, and Soil Pollution, 44*(1-2), 143-158.

Fowler, B. A., Nordberg, G. F., & Nordberg, M. (2015). *Handbook on the Toxicology of Metals*: San Diego, USA: Academic Press.

Frisbie, S. H., Mitchell, E. J., Dustin, H., Maynard, D. M., & Sarkar, B. (2012). World Health Organization discontinues its drinking-water guideline for manganese. *Environmental Health Perspectives*, *120*(6), 775-778.

Gan, W. Y., Mohd, N., Zalilah, M., & Hazizi, A. (2011). Differences in eating behaviours, dietary intake and body weight status between male and female Malaysian university students. *Malaysian Journal of Nutrition, 17*(2), 213-228.

Ghani, A., Zakaria, N., & Ainan, A. (2008). (*Sustainable Urban Drainage System (SUDS)–Malaysian Experiences*). Paper presented at the Proceedings of the 11th International Conference on Urban Drainage. 31st August - 5th September, 2008. Edinburgh, Scotland.

Gao, Q., Li, Y., Cheng, Q., Yu, M., Hu, B., Wang, Z., & Yu, Z. (2016). Analysis and assessment of the nutrients, biochemical indexes and heavy metals in the Three Gorges Reservoir, China, from 2008 to 2013. *Water Research, 92*, 262-274.

Garg, R., Rao, R., Uchchariya, D., Shukla, G., & Saksena, D. (2010). Seasonal variations in water quality and major threats to Ramsagar reservoir, India. *African Journal of Environmental Science and Technology, 4*(2), 062-076.

Gasmi, T., Khouni, I., & Ghrabi, A. (2016). Assessment of heavy metals pollution using multivariate statistical analysis methods in Wadi El Bey (Tunisia). *Desalination and Water Treatment*, 1-14.

Geng, M., Qi, H., Liu, X., Gao, B., Yang, Z., Lu, W., & Sun, R. (2016). Occurrence and health risk assessment of selected metals in drinking water from two typical remote areas in China. *Environmental Science and Pollution Research, 23*(9), 8462-8469.

Ghani, A., Azirun, M., Ramli, R., & Hashim, R. (2008). Geochemical contrast between Beroga (Kuala Kelawang pluton) and Semenyih (Kuala Lumpur pluton) granites in Kuala Kelawang area, Negeri Sembilan. *Bulletin of the Geological Society of Malaysia, 54*, 71-47.

Glover, H. G. (1983). Mine water pollution–an overview of problems and control strategies in the United Kingdom. *Water Science and Technology, 15*(2), 59-70.

Gopaul, P. R., Nowbuth, M. D., & Baguant-Moonshiram, Y. (2009). Water quality indexing for predicting variation of water quality over time. *University of Mauritius Research Journal, 15*(1), 186-199.

Griboff, J., Wunderlin, D. A., & Monferran, M. V. (2017). Metals, As and Se determination by inductively coupled plasma-mass spectrometry (ICP-MS) in edible fish collected from three eutrophic reservoirs. Their consumption represents a risk for human health? *Microchemical Journal, 130*, 236-244.

Gul, A., Bulut, G., & Sirkeci, A. A. (2013). Beneficiation of arsenic bearing complex sulphide ore by flotation. *Physical-chemical Problems of Mineral Processing, 49*(1), 203-212.

Gupta, A., Gupta, S., & Patil, R. S. (2003). A comparison of water quality indices for coastal water. *Journal of Environmental Science and Health, Part A, 38*(11), 2711-2725.

Gupta, S. K., Chabukdhara, M., Singh, J., & Bux, F. (2015). Evaluation and Potential Health Hazard of Selected Metals in Water, Sediments, and Fish from the Gomti River. *Human and Ecological Risk Assessment: An International Journal, 21*(1), 227-240.

Guruge, K. S., Goswami, P., Watanabe, I., Abeykoon, S., Prabhasankar, V., Binu, K., . . . Munuswamy, N. (2017). Trace element distribution and risk assessment in South Indian surface waterways. *International Journal of Environmental Science and Technology, 14*(1), 1-18.

Hallock, D. (2002). *A water quality index for ecology's stream monitoring program*: Washington: State Department of Ecology Olympia.

Hamed, Y., Abdelmoneim, T., ElKiki, M., Hassan, M., & Berndtsson, R. (2013). Assessment of heavy metals pollution and microbial contamination in water, sediments and fish of Lake Manzala, Egypt. *Life Science Journal, 10*(1), 86-94.

Hamzah, Z., Saat, A., Wood, A. K., & Bakar, Z. A. (2011). Sedimentation, heavy metals profiles and cluster analysis of a former tin mining lake. *International Journal of Environmental Science and Development, 2*(6), 448-453.

Han, F., & Zheng, Y. (2016). Multiple-response Bayesian calibration of watershed water quality models with significant input and model structure errors. *Advances in Water Resources, 88*, 109-123.

Hanna-Attisha, M., LaChance, J., Sadler, R. C., & Champney Schnepp, A. (2016). Elevated blood lead levels in children associated with the Flint drinking water crisis: a spatial analysis of risk and public health response. *American Journal of Public Health, 106*(2), 283-290.

Harris, M. J., Stinson, J., & Landis, W. G. (2017). A Bayesian Approach to Integrated Ecological and Human Health Risk Assessment for the South River, Virginia Mercury-Contaminated Site. *Risk Analysis, 37(7)*, 1341-1357.

Hassan, M. M. (2005). Arsenic poisoning in Bangladesh: spatial mitigation planning with GIS and public participation. *Health Policy, 74*(3), 247-260.

Hatar, H., Rahim, S. A., Razi, W. M., & Sahrani, F. K. (2013). (*Heavy metals content in acid mine drainage at abandoned and active mining area*). Paper presented at the AIP Conference Proceedings. 3rd-5th April, 2013, Kedah Malaysia. American Institute of Physics, 1571 (1), 641 - 646. doi.org/10.1063/1.4858727.

Hauser, P. C. (2016). *Determination of Alkali Ions in Biological and Environmental Samples.* The Alkali Metal Ions: Their Role for Life. New York: Springer. (p.11-25)

Heng, S. B., & Kannan, H. (19th May, 2014). Health Ministry calls for halt to using mining pool water. *My Medicnews*. Retrieved from http://mymedicnews.com on 13th March, 2015.

Hogarh, J. N., Adu-Gyamfi, E., Nukpezah, D., Akoto, O., & Adu-Kumi, S. (2016). Contamination from mercury and other heavy metals in a mining district in Ghana: discerning recent trends from sediment core analysis. *Environmental Systems Research, 5*(1), 1-9.

Hornung, R. W., & Reed, L. D. (1990). Estimation of average concentration in the presence of nondetectable values. *Applied Occupational and Environmental Hygiene, 5*(1), 46-51.

Horton, R. K. (1965). An index number system for rating water quality. *Journal of Water Pollution Control Federation, 37*(3), 300-306.

Hou, W., Sun, S., Wang, M., Li, X., Zhang, N., Xin, X., . . . Jia, R. (2016). Assessing water quality of five typical reservoirs in lower reaches of Yellow River, China: using a water quality index method. *Ecological Indicators, 61*, 309-316.

Howe, P. D., Malcolm, H., & Dobson, S. (2004). *Manganese and its compounds: environmental aspects*. Concise international chemical assessment document. Geneva: World Health Organization.

Huang, J., Yuan, F., Zeng, G., Li, X., Gu, Y., Shi, L., . . . Shi, Y. (2017). Influence of pH on heavy metal speciation and removal from wastewater using micellar-enhanced ultrafiltration. *Chemosphere, 173*, 199-206.

Hughes, M. F. (2002). Arsenic toxicity and potential mechanisms of action. *Toxicology Letters, 133*(1), 1-16.

IARC. (1993). *Beryllium, Cadmium, Mercury, and Exposures in the Glass Manufacturing Industry* World Health Organization: International Agency for Research on Cancer.

Iavazzo, P., Ducci, D., Adamo, P., Trifuoggi, M., Migliozzi, A., & Boni, M. (2012). Impact of past mining activity on the quality of water and soil in the High Moulouya Valley (Morocco). *Water, Air, & Soil Pollution, 223*(2), 573-589.

Igwe, O., Adepehin, E. J., Iwuanyanwu, C., & Una, C. O. (2014). Risks associated with the mining of Pb–Zn minerals in some parts of the Southern Benue trough, Nigeria. *Environmental Monitoring and Assessment, 186*(6), 3755-3765.

Iqbal, J., & Shah, M. H. (2013). Health risk assessment of metals in surface water from freshwater source lakes, Pakistan. *Human and Ecological Risk Assessment: An International Journal, 19*(6), 1530-1543.

Iqbal, J., Shah, M. H., & Akhter, G. (2013). Characterization, source apportionment and health risk assessment of trace metals in freshwater Rawal Lake, Pakistan. *Journal of Geochemical Exploration, 125*, 94-101.

Iscen, C. F., Emiroglu, Ö., Ilhan, S., Arslan, N., Yilmaz, V., & Ahiska, S. (2008). Application of multivariate statistical techniques in the assessment of surface water quality in Uluabat Lake, Turkey. *Environmental Monitoring and Assessment, 144*(1-3), 269-276.

Ismail, M. N. (1973). (*A study of trace elements in steam sediments from road/river interception in Central Pahang and the potential value of such a method as an aid in reconnaissance prospecting in tropical terrain*). (Master's thesis) University Malaya.

Izham, P. L. (5th August, 2014). Expert urges extra caution on ex-mining pond water in drought. *Malaysia Online News*. Rerived from http://themalaymailonline.com on 10th May, 2015.

Jahanshahi, R., & Zare, M. (2015). Assessment of heavy metals pollution in groundwater of Golgohar iron ore mine area, Iran. *Environmental Earth Sciences, 74*(1), 505-520.

Järup, L. (2003). Hazards of heavy metal contamination. *British Medical Bulletin, 68*(1), 167-182.

Järup, L., Alfvén, T., Persson, B., Toss, G., & Elinder, C. G. (1998). Cadmium may be a risk factor for osteoporosis. *Occupational and Environmental Medicine, 55*(7), 435-439.

Jha, D. K., Devi, M. P., Vidyalakshmi, R., Brindha, B., Vinithkumar, N. V., & Kirubagaran, R. (2015). Water quality assessment using water quality index and geographical information system methods in the coastal waters of Andaman Sea, India. *Marine Pollution Bulletin, 100*(1), 555-561.

Jiang, Y., Nan, Z., & Yang, S. (2013). Risk assessment of water quality using Monte Carlo simulation and artificial neural network method. *Journal of Environmental Management, 122*, 130-136.

Jidon, A. (1993). Skin cancer caused by chronic arsenical poisoning-a report of three cases. *Medical Journal Malaysia, 48*(1), 87.

Johnson, D. B., & Hallberg, K. B. (2003). The microbiology of acidic mine waters. *Research in Microbiology, 154*(7), 466-473.

Johnson, R., Blowes, D., Robertson, W., & Jambor, J. (2000). The hydrogeochemistry of the Nickel Rim mine tailings impoundment, Sudbury, Ontario. *Journal of Contaminant Hydrology, 41*(1), 49-80.

Jones, A., Rogerson, M., Greenway, G., Potter, H., & Mayes, W. (2013). Mine water geochemistry and metal flux in a major historic Pb-Zn-F orefield, the Yorkshire Pennines, UK. *Environmental Science and Pollution Research, 20*(11), 7570-7581.

Juahir, H., Ekhwan, T. M., Zain, S. M., Mokhtar, M., Zaihan, J., & Ijan Khushaida, M. (2008). The use of chemometrics analysis as a cost-effective tool in sustainable utilisation of water resources in the Langat River Catchment. *American-Eurasian Journal of Agricultural & Environmental Sciences, 4*(1), 258-265.

Juahir, H., Zain, S. M., Aris, A. Z., Yusoff, M. K., & Mokhtar, M. B. (2010). Spatial assessment of Langat river water quality using chemometrics. *Journal of Environmental Monitoring, 12*(1), 287-295.

Juahir, H., Zain, S. M., Yusoff, M. K., Hanidza, T. I., Armi, A. S. M., Toriman, M. E., & Mokhtar, M. (2011). Spatial water quality assessment of Langat River Basin (Malaysia) using environmetric techniques. *Environmental Monitoring and Assessment, 173*(1), 625-641.

Jusop, S., Wan, N., Mokhtar, N., & Paramananthan, S. (1986). Morphology, mineralogy and chemistry of an ex-mining land in Ipoh, Perak. *Pertanika, 9*(1), 89-97.

Kacmaz, H., & Burns, P. C. (2017). Uranyl phosphates and associated minerals in the Koprubasi (Manisa) uranium deposit, Turkey. *Ore Geology Reviews, 84*, 102-115.

Kakei, M., Yoshikawa, M., & Mishima, H. (2016). Fluoride exposure may accelerate the osteoporotic change in postmenopausal women: Animal model of fluoride-induced osteoporosis. *Advanced Techniques in Biology & Medicine, 4(1),* 1-4..

Kannel, P. R., Lee, S., Lee, Y. S., Kanel, S. R., & Khan, S. P. (2007). Application of water quality indices and dissolved oxygen as indicators for river water classification and urban impact assessment. *Environmental Monitoring and Assessment, 132*(1-3), 93-110.

Kavcar, P., Sofuoglu, A., & Sofuoglu, S. C. (2009). A health risk assessment for exposure to trace metals via drinking water ingestion pathway. *International Journal of Hygiene and Environmental Health, 212*(2), 216-227.

Kazi, T. G., Brahman, K. D., Afridi, H. I., Arain, M. B., Talpur, F. N., & Akhtar, A. (2016). The effects of arsenic contaminated drinking water of livestock on its total levels in milk samples of different cattle: Risk assessment in children. *Chemosphere, 165,* 427-433.

Keen, C., & Zidenberg-Cherr, S. (1994). Manganese toxicity in humans and experimental animals. *Manganese in health and disease*. Florida: CRC Press. p. 193-205.

Kemp, D., Bond, C. J., Franks, D. M., & Cote, C. (2010). Mining, water and human rights: making the connection. *Journal of Cleaner Production, 18*(15), 1553-1562.

Khailani, D. K., & Perera, R. (2013). Mainstreaming disaster resilience attributes in local development plans for the adaptation to climate change induced flooding: A study based on the local plan of Shah Alam City, Malaysia. *Land Use Policy, 30*(1), 615-627.

Khairi, S. (7th May, 2014). LUAS denies treated water from mining pools 'unsafe'. *ABN News*. Retrieved from https://abnnews.com/malaysia on 19th August 2015.

Khan, A. M., Yusoff, I., Bakar, N. K. A., Bakar, A. F. A., & Alias, Y. (2016). Assessing anthropogenic levels, speciation, and potential mobility of rare earth elements (REEs) in ex-tin mining area. *Environmental Science and Pollution Research, 23*(24), 25039-25055.

Khan, K., Wasserman, G. A., Liu, X., Ahmed, E., Parvez, F., Slavkovich, V., . . . Graziano, J. H. (2012). Manganese exposure from drinking water and children's academic achievement. *Neurotoxicology, 33*(1), 91-97.

Khoo, T., & Tan, B. (1983). (*Geological evolution of peninsular Malaysia*). Paper presented at the Proceedings of the Workshop on Stratigraphic Correlation of Thailand and Malaysia. Geological Society of Thailand, Bangkok/Geological Society of Malaysia, Kuala Lumpur. 8th - 10th September, 1983. 1:253-290.

Kim, M. J., Nriagu, J., & Haack, S. (2000). Carbonate ions and arsenic dissolution by groundwater. *Environmental Science & Technology, 34*(15), 3094-3100.

Kondakis, X. G., Makris, N., Leotsinidis, M., Prinou, M., & Papapetropoulos, T. (1989). Possible health effects of high manganese concentration in drinking water. *Archives of Environmental Health: An International Journal, 44*(3), 175-178.

Kosobrukhov, A., Knyazeva, I., & Mudrik, V. (2004). Plantago major plants responses to increase content of lead in soil: growth and photosynthesis. *Plant Growth Regulation, 42*(2), 145-151.

Krishna, A. K., Satyanarayanan, M., & Govil, P. K. (2009). Assessment of heavy metal pollution in water using multivariate statistical techniques in an industrial area: a case study from Patancheru, Medak District, Andhra Pradesh, India. *Journal of Hazardous Materials, 167*(1), 366-373.

Kuria, F., & Vogel, R. (2015). Uncertainty analysis for water supply reservoir yields. *Journal of hydrology, 529*, 257-264.

Kusin, F., Zahar, M., Muhammad, N., Zin, Z., & Sharif, S. (2016). Assessing the potential use of abandoned mining pools as an alternative resource of raw water supply. *Water Science and Technology: Water Supply, 16*(2), 410-417.

Kusin, F. M., Muhammad, S. N., Zahar, M. S. M., & Madzin, Z. (2016). Integrated River Basin Management: incorporating the use of abandoned mining pool and implication on water quality status. *Desalination and Water Treatment*, 1-11.

Kusin, M., Nurjaliah, M., Mohamad, Z., & Madzin, Z. (2016). Hybrid off-river augmentation system as an alternative raw water resource: the hydrogeochemistry of abandoned mining ponds. *Environmental Earth Sciences, 75*(3), 1-15.

Langard, S., Norseth, T., & Friberg, L. (1986). *Handbook on the Toxicology of Metals*. North Holland: Elsevier. p. 185-210.

Langhans, C., Smith, H. G., Chong, D. M., Nyman, P., Lane, P. N., & Sheridan, G. J. (2016). A model for assessing water quality risk in catchments prone to wildfire. *Journal of Hydrology, 534*, 407-426.

Larter, N., Macdonald, C., Elkin, B., Wang, X., Harms, N., Gamberg, M., & Muir, D. (2016). Cadmium and other elements in tissues from four ungulate species from the Mackenzie Mountain region of the Northwest Territories, Canada. *Ecotoxicology and Environmental Safety, 132*, 9-17.

Leong, K. H., Tan, L. B., & Mustafa, A. M. (2007). Contamination levels of selected organochlorine and organophosphate pesticides in the Selangor River, Malaysia between 2002 and 2003. *Chemosphere, 66*(6), 1153-1159.

Lermontov, A., Yokoyama, L., Lermontov, M., & Machado, M. A. S. (2009). River quality analysis using fuzzy water quality index: Ribeira do Iguape river watershed, Brazil. *Ecological Indicators, 9*(6), 1188-1197.

Li, D., Huang, D., Guo, C., & Guo, X. (2015). Multivariate statistical analysis of temporal–spatial variations in water quality of a constructed wetland purification system in a typical park in Beijing, China. *Environmental Monitoring and Assessment*, *187*(1), 1-14.

Li, F., Huang, J., Zeng, G., Yuan, X., Li, X., Liang, J., . . . Bai, B. (2013). Spatial risk assessment and sources identification of heavy metals in surface sediments from the Dongting Lake, Middle China. *Journal of Geochemical Exploration, 132*, 75-83.

Li, P. (2016). Groundwater quality in Western China: challenges and paths forward for groundwater quality research in Western China. *Exposure and Health, 8*(3), 305-310.

Li, Z., Ma, Z., van der Kuijp, T. J., Yuan, Z., & Huang, L. (2014). A review of soil heavy metal pollution from mines in China: pollution and health risk assessment. *Science of the Total Environment, 468,* 843-853.

Liebman, H. (1969). *Atlas of water quality: methods and practical conditions*. Munich: R. Oldenbourgh.

Limei, Z., Xiaoyong, L., Tongbin, C., Xiulan, Y., Hua, X., Bin, W., & Lixia, W. (2008). Regional assessment of cadmium pollution in agricultural lands and the potential health risk related to intensive mining activities: A case study in Chenzhou City, China. *Journal of Environmental Sciences, 20*(6), 696-703.

Lindsay, M. B., Moncur, M. C., Bain, J. G., Jambor, J. L., Ptacek, C. J., & Blowes, D. W. (2015). Geochemical and mineralogical aspects of sulfide mine tailings. *Applied Geochemistry, 57*, 157-177.

Liou, S. M., Lo, S. L., & Wang, S. H. (2004). A generalized water quality index for Taiwan. *Environmental Monitoring and Assessment*, *96*(1), 35-52.

Liu, C. W., Lin, K. H., & Kuo, Y.-M. (2003). Application of factor analysis in the assessment of groundwater quality in a blackfoot disease area in Taiwan. *Science of the Total Environment*, *313*(1), 77-89.

Lo, Y. C., Dooyema, C. A., Neri, A., Durant, J., Jefferies, T., Medina-Marino, A., . . . Dankoli, R. S. (2012). Childhood lead poisoning associated with gold ore processing: a village-level investigation-Zamfara State, Nigeria, October-November 2010. *Environmental Health Perspectives, 120*(10), 1450.

Loh, N., Loh, H. P., Wang, L. K., & Wang, M. H. S. (2016). Health effects and control of toxic lead in the environment. In Wang L.K., Wang M-H. S., Hung Y-I. & Shammas, M. K. (Eds.), *Natural Resources and Control Processes.* Handbook of Environmental Engineering (p 233-284). Switzerland: Springer International Publishing.

Lonati, G., & Zanoni, F. (2013). Monte-Carlo human health risk assessment of mercury emissions from a MSW gasification plant. *Waste Management*, *33*(2), 347-355.

Low, K. H., Zain, S. M., & Abas, M. R. (2012). Evaluation of microwave-assisted digestion condition for the determination of metals in fish samples by inductively coupled plasma mass spectrometry using experimental designs. *International Journal of Environmental Analytical Chemistry, 92*(10), 1161-1175.

Low, K. H., Zain, S. M., Abas, M. R., Salleh, K. M., & Teo, Y. Y. (2015). Distribution and health risk assessment of trace metals in freshwater tilapia from three different aquaculture sites in Jelebu Region (Malaysia). *Food Chemistry, 177*, 390-396.

Low, K. H., Isa Baba, K., Hafizan, J., Azman, A., Shima, B., Rabia, I., . . . Sharifuddin, M. Z. (2016). Evaluation of water quality variation in lakes, rivers, and ex-mining ponds in Malaysia (review). *Desalination and Water Treatment,57(58),* 28215- 28239.

Lu, L., Cheng, H., Liu, X., Xie, J., Li, Q., & Zhou, T. (2015). Assessment of regional human health risks from lead contamination in Yunnan province, southwestern China. *PloS one, 10*(4), e0119562.

LUAS. (2008). *IRBM Approach in Water Quality Management*—Case Study in Sungai Selangor River Basin. Lembaga Urus Air Selangor. Kuala Lumpur.

LUAS. (2012). *Water Resources Management in Selangor—Issues and Strategies*, Malaysia Water Resources Management Forum 2012. Lembaga Urus Air Selangor. Kuala Lumpur.

Luís, A. T., Teixeira, P., Almeida, S. F. P., Matos, J. X., & da Silva, E. F. (2011). Environmental impact of mining activities in the Lousal area (Portugal): chemical and diatom characterization of metal-contaminated stream sediments and surface water of Corona stream. *Science of the Total Environment, 409*(20), 4312-4325.

Lumb, A., Sharma, T., & Bibeault, J. F. (2011). A review of genesis and evolution of water quality index (WQI) and some future directions. *Water Quality, Exposure and Health, 3*(1), 11-24.

Ma, L., Ascough II, J., Ahuja, L., Shaffer, M., Hanson, J., & Rojas, K. (2000). Root zone water quality model sensitivity analysis using Monte Carlo simulation. *Transactions of the ASAE, 43*(4), 883.

MacDougall, D., & Crummett, W. B. (1980). Guidelines for data acquisition and data quality evaluation in environmental chemistry. *Analytical Chemistry, 52*(14), 2242-2249.

Madzin, Z., Shai-in, M. F., & Kusin, F. M. (2015). Comparing heavy metal mobility in active and abandoned mining sites at Bestari Jaya, Selangor. *Procedia Environmental Sciences, 30*, 232-237.

Man, Y. B., Sun, X. L., Zhao, Y. G., Lopez, B. N., Chung, S. S., Wu, S. C., . . . Wong, M. H. (2010). Health risk assessment of abandoned agricultural soils based on heavy metal contents in Hong Kong, the world's most populated city. *Environment International, 36*(6), 570-576.

Mandal, B. K., & Suzuki, K. T. (2002). Arsenic round the world: a review. *Talanta, 58*(1), 201-235.

Marín, A., Andrades, M., Iñigo, V., & Jiménez-Ballesta, R. (2016) Lead and Cadmium in Soils of La Rioja Vineyards, Spain. *Land Degradation. Development*, 27, 1286–1294. doi: 10.1002/ldr.2471.

Martha B., Luis M., & M., J. (2011). Sorption Of heavy metals from gold mining Waste water using chitosan *Journal of the Taiwan Institute of Chemical Engineers, 42*, 976-988.

Martín, J. A. R., Gutiérrez, C., Escuer, M., García-González, M. T., Campos-Herrera, R., & Águila, N. (2014). Effect of mine tailing on the spatial variability of soil nematodes from lead pollution in La Union (Spain). *Science of the Total Environment*, *473*, 518-529.

Mayes, W., Johnston, D., Potter, H., & Jarvis, A. (2009). A national strategy for identification, prioritisation and management of pollution from abandoned non-coal mine sites in England and Wales. I.: methodology development and initial results. *Science of the Total Environment, 407*(21), 5435-5447.

McCullough, C., & Schultze, M. (2015). (*Riverine Flow-Through of Mine Pit Lakes: Improving both Mine Pit Lake and River Water Quality Values?*) Paper presented at the Proceedings of the 9th International Mine Water Association (IMWA)/International Conference on Acid Rock Drainage (ICARD), 20^{th} - 24^{th} April, 2015. Santiago, Chile.

MDG. (2013). *Malaysian Dietary Guidelines for Childrens and Adolescents*. Kuala Lumpur: Ministry of Health Malaysia.

MERC. (2012). *Study on the establishment of marine water quality index for Malaysia*. Kuala Lumpur: Malaysian Environmental Resources Centre.

Michaelsen, K. F. (2015). *Child Growth.* Germany: Karger Publishers.

Mielke, H. W. (2016). Editorial: Lead Risk Assessment and Health Effects. *International Journal of Environmental Research and Public Health*, *13*(6), 587.

Mishra, V. K., Upadhyay, A. R., Pandey, S. K., & Tripathi, B. (2008). Concentrations of heavy metals and aquatic macrophytes of Govind Ballabh Pant Sagar an anthropogenic lake affected by coal mining effluent. *Environmental Monitoring and Assessment, 141*(1-3), 49-58.

Mitchell, J. W. (2009). An assessment of lead mine pollution using macro-invertebrates at Greenside Mines, Glenridding. *Earth & Environment, 4*, 27-57.

Modoi, O.-C., Roba, C., Török, Z., & Ozunu, A. (2014). Environmental risks due to heavy metal pollution of water resulted from mining wastes in NW Romania. *Environmental Engineering and Management Journal, 13*(9), 2325-2336.

MOH. (2004). *National Standard for Drinking Water Quality*, Ministry of Health, Malaysia. Document D1.

Mohamad, H. A. (2016). *(Key Findings of Recent Food Consumption and Nutrition Surveys in ASEAN - Malaysian Adult Nutrition Survey (MANS) 2014).* Paper presented at the Symposium on Dietary Intake, 26th April, 2016. Singapore.

Mohammadian, M., Nouri, J., Afshari, N., Nassiri, J., & Nourani, M. (2008). Investigation of heavy metals concentrations in the water wells close to Zanjan zinc and lead smelting plant. *Iranian Journal of Health and Environment, 1*(1), 51-56.

Mohan, S. V., Nithila, P., & Reddy, S. J. (1996). Estimation of heavy metals in drinking water and development of heavy metal pollution index. *Journal of Environmental Science & Health Part A, 31*(2), 283-289.

Montgomery, D. C., Peck, E. A., & Vining, G. G. (2012). *Introduction to linear regression analysis*. New Jersey: John Wiley & Sons.

Mooney, C. Z. (1997). *Monte carlo simulation*. California: Sage Publications.

Morgan, R. (1968). An Analysis of Basin Asymmetry in the Klang Basin, Selangor. *Bulletin of the Geological Society of Malaysia* (3-5), 17.

Muangthong, S., & Shrestha, S. (2015). Assessment of surface water quality using multivariate statistical techniques: case study of the Nampong River and Songkhram River, Thailand. *Environmental Monitoring and Assessment, 187*(9), 1-12.

Muhammad, S., Shah, M. T., & Khan, S. (2011). Health risk assessment of heavy metals and their source apportionment in drinking water of Kohistan region, northern Pakistan. *Microchemical Journal, 98*(2), 334-343.

Murcott, S. (2012). Arsenic contamination in the world. *An international sourcebook*. London : IWA Publishing.

Mustafa, H. T., Hassan, H. M., Abo-Melha, A., & Rihan, T. I. (1988). Cadmium and zinc concentrations in the potable water of the Eastern Province of Saudi Arabia. *Bulletin of Environmental Contamination and Toxicology, 40*(3), 462-467.

Mustapha, A., Aris, A. Z., Juahir, H., Ramli, M. F., & Kura, N. U. (2013). River water quality assessment using environmentric techniques: case study of Jakara River Basin. *Environmental Science and Pollution Research, 20*(8), 5630-5644.

Muthaiah, V. P. K., Chen, G.-D., Ding, D., Salvi, R., & Roth, J. A. (2016). Effect of manganese and manganese plus noise on auditory function and cochlear structures. *Neurotoxicology, 55*, 65-73.

NAHRIM. (2009). *Study on the Status of Eutrophication of Lakes in Malaysia*. NAHRIM, Seri Kembangan. Austria : JohnWiley & Sons.

Naidoo, S. (2017). *Water Mining and Development in South Africa Acid Mine Drainage in South Africa*. Switzerland: Springer Nature.

Nasirian, M. (2007). A new water quality index for environmental contamination contributed by mineral processing: a case study of Amang (Tin Tailing) processing activity. *Journal of Applied Sciences, 7*(20), 2977-2987.

Nation. (2014, May, 1st). Geologist: Heavy metal pollution at some mining pools. *The Star Online*. Retrieved from http://www.thestar.com.my/news/nation on 18th July, 2015.

Naubi, I., Zardari, N. H., Shirazi, S. M., Ibrahim, N. F., & Baloo, L. (2016). Effectiveness of water quality index for monitoring Malaysian river water quality. *Polish Journal of Environmental Studies, 25(1)*, 231-239.

Navarro, M., Pérez-Sirvent, C., Martínez-Sánchez, M., Vidal, J., Tovar, P., & Bech, J. (2008). Abandoned mine sites as a source of contamination by heavy metals: a case study in a semi-arid zone. *Journal of Geochemical Exploration, 96*(2), 183-193.

Navas-Acien, A., Tellez-Plaza, M., Guallar, E., Muntner, P., Silbergeld, E., Jaar, B., & Weaver, V. (2009). Blood cadmium and lead and chronic kidney disease in US adults: a joint analysis. *American Journal of Epidemiology, 170*(9), 1156-1164.

Nawrot, T. S., Martens, D. S., Hara, A., Plusquin, M., Vangronsveld, J., Roels, H. A., & Staessen, J. A. (2015). Association of total cancer and lung cancer with environmental exposure to cadmium: the meta-analytical evidence. *Cancer Causes & Control, 26*(9), 1281-1288.

Nazeer, S., Hashmi, M. Z., & Malik, R. N. (2014). Heavy metals distribution, risk assessment and water quality characterization by water quality index of the River Soan, Pakistan. *Ecological Indicators, 43*, 262-270.

Nazir, R., Khan, M., Masab, M., Rehman, H. U., Rauf, N. U., Shahab, S., . . . Rafeeq, M. (2015). Accumulation of heavy metals (Ni, Cu, Cd, Cr, Pb, Zn, Fe) in the soil, water and plants and analysis of physico-chemical parameters of soil and water collected from Tanda Dam kohat. *Journal of Pharmaceutical Sciences and Research, 7*(3), 89-97.

Needleman, H. L., & Bellinger, D. (1991). The health effects of low level exposure to lead. *Annual Review of Public Health, 12*(1), 111-140.

Nevárez, M., Leal, L. O., & Moreno, M. (2015). Estimation of seasonal risk caused by the intake of lead, mercury and cadmium through freshwater fish consumption from urban water reservoirs in arid areas of northern Mexico. *International Journal of Environmental Research and Public Health, 12*(2), 1803-1816.

Ng, J., Wang, J., & Shraim, A. (2003). A global health problem caused by arsenic from natural sources. *Chemosphere, 52*(9), 1353-1359.

Ngure, V., Davies, T., Kinuthia, G., Sitati, N., Shisia, S., & Oyoo-Okoth, E. (2014). Concentration levels of potentially harmful elements from gold mining in Lake Victoria Region, Kenya: Environmental and health implications. *Journal of Geochemical Exploration, 144*, 511-516.

Ning, L., Liyuan, Y., Jirui, D., & Xugui, P. (2011). Heavy metal pollution in surface water of Linglong gold mining area, China. *Procedia Environmental Sciences, 10*, 914-917.

Nishijo, M., Nakagawa, H., Morikawa, Y., Kuriwaki, J.-i., Miura, K., Kido, T., & Nogawa, K. (2004). Mortality in a cadmium polluted area in Japan. *Biometals, 17*(5), 535-538.

Noori, R., Sabahi, M. S., Karbassi, A., Baghvand, A., & Zadeh, H. T. (2010). Multivariate statistical analysis of surface water quality based on correlations and variations in the data set. *Desalination, 260*(1), 129-136.

Norgate, T., & Haque, N. (2010). Energy and greenhouse gas impacts of mining and mineral processing operations. *Journal of Cleaner Production, 18*(3), 266-274.

Nuttall, C., & Younger, P. (2002). Secondary minerals in the abandoned mines of Nenthead, Cumbria as sinks for pollutant metals. *Geological Society, London, Special Publications, 198*(1), 241-250.

Ogola, J. S., Mitullah, W. V., & Omulo, M. A. (2002). Impact of gold mining on the environment and human health: a case study in the Migori gold belt, Kenya. *Environmental Geochemistry and Health, 24*(2), 141-157.

Ordóñez, A., Alvarez, R., Charlesworth, S., De Miguel, E., & Loredo, J. (2011). Risk assessment of soils contaminated by mercury mining, Northern Spain. *Journal of Environmental Monitoring, 13*(1), 128-136.

Orji, K. U., Sapari, N., Yusof, K. W., Asadpour, R., & Olisa, E. (2014). Water Quality Assessment of Ex-mining Lakes in Perak, Malaysia as Alternative Source of Water Supply. *Applied Mechanics & Materials,* 567, 177-182.

Osman, R. M., & Siong, L. C. (2012). *Geospatial Analysis of the Ex-Mining Land of Melaka*. Paper presented at the National Geoscience Conference, 23-34th June, 2012, Kuching, Malaysia.

Oulhote, Y., Mergler, D., Barbeau, B., Bellinger, D. C., Bouffard, T., Brodeur, M. È., . . . Bouchard, M. F. (2014). Neurobehavioral function in school-age children exposed to manganese in drinking water. *Environmental Health Perspective, 122(12)*, 1343 - 1350.

Paikaray, S. (2015). Arsenic geochemistry of acid mine drainage. *Mine water and the environment, 34*(2), 181-196.

Palmer, M., Galloway, J., Jamieson, H., Patterson, R., Falck, H., & Kokelj, S. (2016). The concentration of arsenic in lake waters of the Yellowknife area. *Yellowknife,Canada NT: NWT Open File, 6*.

Papatheodorou, G., Demopoulou, G., & Lambrakis, N. (2006). A long-term study of temporal hydrochemical data in a shallow lake using multivariate statistical techniques. *Ecological Modelling, 193*(3), 759-776.

Paulo, A. (2005). Economical and natural conditions applicable to the development of post-mining areas. *Polish Geological Institute Special Papers, 17*, 49-69.

Poerschmann, J., Koschorreck, M., & Górecki, T. (2017). Organic matter in sediment layers of an acidic mining lake as assessed by lipid analysis. Part II: Neutral lipids. *Science of the Total Environment, 578*, 219-227.

Poirier, L., Nelson, J., Leong, D., Berhane, L., Hajdu, P., & Lopez-Linares, F. (2016). Application of ICP-MS and ICP-OES on the Determination of Nickel, Vanadium, Iron, and Calcium in Petroleum Crude Oils via Direct Dilution. *Energy & Fuels, 30*(5), 3783-3790.

Popa, M., Mihai, C., Dorin, P., & Mirel, G. (2013). Educating Future Engineers for the Prevention of Heavy Metals Contamination of Surface Waters in Mining Areas. The Case of Zlatna, Alba County. *Procedia-Social and Behavioral Sciences, 83*, 408-412.

Potts, R. O., & Guy, R. H. (1992). Predicting skin permeability. *Pharmaceutical Research, 9*(5), 663-669.

Prasad, B., Kumari, P., Bano, S., & Kumari, S. (2014). Ground water quality evaluation near mining area and development of heavy metal pollution index. *Applied Water Science, 4*(1), 11-17.

Puga, A., Abreu, C., Melo, L., & Beesley, L. (2015). Biochar application to a contaminated soil reduces the availability and plant uptake of zinc, lead and cadmium. *Journal of Environmental Management, 159*, 86-93.

Qaiyum, M., Shaharudin, M., Syazwan, A., & Muhaimin, A. (2011). Health risk assessment after exposure to aluminium in drinking water between two different villages. *Journal of Water Resource and Protection, 3*(4), 268-274.

Qu, C., Sun, K., Wang, S., Huang, L., & Bi, J. (2012). Monte carlo simulation-based health risk assessment of heavy metal soil pollution: A case study in the Qixia mining area, China. *Human and Ecological Risk Assessment: An International Journal, 18*(4), 733-750.

Quraishi, S. M., Adams, S. V., Shafer, M., Meliker, J. R., Li, W., Luo, J., . . . Newcomb, P. A. (2015). Urinary cadmium and estimated dietary cadmium in the Women's Health Initiative. *Journal of Exposure Science and Environmental Epidemiology, 26(3)*, 303-308.

Rahman, S. M., Kippler, M., Tofail, F., Bölte, S., Hamadani, J. D., & Vahter, M. (2016). Manganese in drinking water and cognitive abilities and behavior at 10 years of age: a prospective cohort study. *Environmental Health Perspectives. 125 (5)*, 1-8.

Räisänen, M., Heikkinen, P., Pulkkinen, K., Korkka-Niemi, K., & Salonen, V. (2005). Finland–Mine water quality in some abandoned and active Finnish metal sulphide mines. *Mine Water and the Environment, 24,* 7-10.

Ramakrishnan, B. (2015). Inexplicable Arsenic: Human Dimensions of Soil, Water, Food and Microbial Resources. *Journal of Environmental & Analytical Toxicology, 5*(277), 2161-0525.

Rasheed, H., Slack, R., & Kay, P. (2016). Human health risk assessment for arsenic: a critical review. *Critical Reviews in Environmental Science and Technology, 46*(19-20), 1529-1583.

Ravikumar, P., Mehmood, M. A., & Somashekar, R. (2013). Water quality index to determine the surface water quality of Sankey tank and Mallathahalli lake, Bangalore urban district, Karnataka, India. *Applied Water Science, 3*(1), 247-261.

Razo, I., Carrizales, L., Castro, J., Díaz-Barriga, F., & Monroy, M. (2004). Arsenic and heavy metal pollution of soil, water and sediments in a semi-arid climate mining area in Mexico. *Water, Air, and Soil Pollution, 152*(1-4), 129-152.

Rebelo, A., Ferra, I., Gonçalves, I., & Marques, A. M. (2014). A risk assessment model for water resources: releases of dangerous and hazardous substances. *Journal of Environmental Management, 140,* 51-59.

Reynolds, E. S. (1901). An account of the epidemic outbreak of arsenical poisoning occurring in beer drinkers in the North of England and the Midland Counties in 1900. *The Lancet*, *157*(4038), 166-170

Robert, C. P. (2004). *Monte carlo methods*. New York: Wiley. .

Rojas, J. C., & Vandecasteele, C. (2007). Influence of mining activities in the North of Potosi, Bolivia on the water quality of the Chayanta River, and its consequences. *Environmental Monitoring and Assessment, 132*(1), 321-330.

Romano, N., & Zeng, C. (2013). Toxic effects of ammonia, nitrite, and nitrate to decapod crustaceans: a review on factors influencing their toxicity, physiological consequences, and coping mechanisms. *Reviews in Fisheries Science, 21*(1), 1-21.

Rosegrant, M. W., Cai, X., & Cline, S. A. (2002). Global water outlook to 2025. Averting an impending crisis. *New England Journal of Public Policy, 21(2),* 102-125.

Rostom, N. G., Shalaby, A. A., Issa, Y. M., & Afifi, A. A. (2017). Evaluation of Mariut Lake water quality using Hyperspectral Remote Sensing and laboratory works. *The Egyptian Journal of Remote Sensing and Space Science, 20*, S39-S48.

Rudall, S., & Jarvis, A. (2011). Diurnal fluctuation of zinc concentration in metal polluted rivers and its potential impact on water quality and flux estimates. *Water Science and Technology, 65*(1), 164-170.

Rybicka, E. H. (1996). Impact of mining and metallurgical industries on the environment in Poland. *Applied Geochemistry, 11*(1), 3-9.

Sadeghi, S., Harchegani, M. K., & Younesi, H. (2012). Suspended sediment concentration and particle size distribution, and their relationship with heavy metal content. *Journal of Earth System Science, 121*(1), 63-71.

Saeedi, M., Abessi, O., Sharifi, F., & Meraji, H. (2010). Development of groundwater quality index. *Environmental Monitoring and Assessment, 163*(1), 327-335.

Saha, N., Rahman, M. S., Ahmed, M. B., Zhou, J. L., Ngo, H. H., & Guo, W. (2017). Industrial metal pollution in water and probabilistic assessment of human health risk. *Journal of Environmental Management, 185*, 70-78.

Sahu, P., & Sikdar, P. (2008). Hydrochemical framework of the aquifer in and around East Kolkata Wetlands, West Bengal, India. *Environmental Geology, 55*(4), 823-835.

Sáinz, A., Grande, J., & De la Torre, M. (2004). Characterisation of heavy metal discharge into the Ria of Huelva. *Environment International, 30*(4), 557-566.

Saleh, T. A., Adio, S. O., Parthasarathy, P., & Danmaliki, G. I. (2017). Scientific insights into modified and non-modified biomaterials for sorption of heavy metals from water. In Saleh, T. A., (Ed), *Advanced Nanomaterials for Water Engineering, Treatment, and Hydraulics (pp 13-39).*. Pennsylvania, USA: IGI Global.

Salim, N. A. A., Wood, A. K., Yusof, A. M., Hamzah, M. S., Elias, M. S., & Rahman, S. A. (2009). A study of arsenic and chromium contamination in sediments of freshwater bodies. *Fresenius Environmental Bulletin, 18*(9), 1618-1623.

Salomons, W. (1995). Environmental impact of metals derived from mining activities: processes, predictions, prevention. *Journal of Geochemical Exploration, 52*(1), 5-23.

Salvarredy-Aranguren, M. M., Probst, A., Roulet, M., & Isaure, M.-P. (2008). Contamination of surface waters by mining wastes in the Milluni Valley (Cordillera Real, Bolivia): Mineralogical and hydrological influences. *Applied Geochemistry, 23*(5), 1299-1324.

Sankhla, M. S., Kumari, M., Nandan, M., Kumar, R., & Agrawal, P. (2016). Heavy Metals Contamination in Water and their Hazardous Effect on Human Health-A Review. *International Journal of Current Microbiology and Applied Sciences 5*(10), 759-766.

Sany, S. B. T., Hashim, R., Rezayi, M., Salleh, A., Rahman, M. A., Safari, O., & Sasekumar, A. (2014). Human health risk of polycyclic aromatic hydrocarbons from consumption of blood cockle and exposure to contaminated sediments and water along the Klang Strait, Malaysia. *Marine Pollution Bulletin, 84*(1), 268-279.

Sastre, J., Sahuquillo, A., Vidal, M., & Rauret, G. (2002). Determination of Cd, Cu, Pb and Zn in environmental samples: microwave-assisted total digestion versus aqua regia and nitric acid extraction. *Analytica Chimica Acta, 462*(1), 59-72.

Saviour, M. N. (2012). Environmental impact of soil and sand mining: a review. *International Journal of Science, Environment and Technology, 1*(3), 125-134.

Schaider, L. A., Senn, D. B., Estes, E. R., Brabander, D. J., & Shine, J. P. (2014). Sources and fates of heavy metals in a mining-impacted stream: Temporal variability and the role of iron oxides. *Science of the Total Environment, 490*(0), 456-466.

Schwartz, M. O., & Askury, A. (1989). Geologic, geochemical, and fluid inclusion studies of the tin granites from the Bujang Melaka pluton, Kinta Valley, Malaysia. *Economic Geology, 84*(4), 751-779.

Seitaj, D., Sulu-Gambari, F., Burdorf, L. D., Romero-Ramirez, A., Maire, O., Malkin, S. Y., . . . Meysman, F. J. (2017). Sedimentary oxygen dynamics in a seasonally hypoxic basin. *Limnology and Oceanography, 62*(2), 452-473.

Selevan, S. G., Kimmel, C. A., & Mendola, P. (2000). Identifying critical windows of exposure for children's health. *Environmental Health Perspectives, 108*(3), 451-455.

Sfikas, A., Angelidis, P., Samaras, P., Zoras, S., & Evagelopoulos, V. (2013). Utilization of a multi-parameter sensor network for online monitoring of the water quality in the lignite mining area of Kozani, Greece. *Desalination and Water Treatment, 51*(13-15), 2977-2986.

Sharip, Z., & Zakaria, S. (2007). (*Lakes and reservoir in Malaysia: management and research challenges.*) Proceedings of Taal 2007: The 12th World Lake Conference. 28th October - 2nd November, 2007, Rajastan, India. 1349-1355.

Sharip, Z., Zaki, A. T., Shapai, M., Suratman, S., & Shaaban, A. J. (2014). Lakes of Malaysia: Water quality, eutrophication and management. *Lakes & Reservoirs: Research & Management, 19*(2), 130-141.

Shi, Y., Wang, R., Lu, Y., Song, S., Johnson, A. C., Sweetman, A., & Jones, K. (2016). Regional multi-compartment ecological risk assessment: Establishing cadmium pollution risk in the northern Bohai Rim, China. *Environment International, 94*, 283-291.

Shirazi, S., Imran, H., Akib, S., Yusop, Z., & Harun, Z. (2013). Groundwater vulnerability assessment in the Melaka State of Malaysia using DRASTIC and GIS techniques. *Environmental Earth Sciences, 70*(5), 2293-2304.

Shirazi, S. M., Adham, M., Zardari, N. H., Ismail, Z., Imran, H. M., & Mangrio, M. A. (2015). Groundwater quality and hydrogeological characteristics of Malacca state in Malaysia. *Journal of Water and Land Development, 24*(1), 11-19.

Shojaei, M., Nazif, S., & Kerachian, R. (2015). Joint uncertainty analysis in river water quality simulation: a case study of the Karoon River in Iran. *Environmental Earth Sciences, 73*(7), 3819-3831.

Shrestha, S., & Kazama, F. (2007). Assessment of surface water quality using multivariate statistical techniques: A case study of the Fuji river basin, Japan. *Environmental Modelling & Software, 22*(4), 464-475.

Shuhaimi-Othman, M., Lim, E. C., & Mushrifah, I. (2007). Water quality changes in Chini Lake, Pahang, West Malaysia. *Environmental Monitoring and Assessment, 131*(1-3), 279-292.

Simeonov, V., Einax, J., Stanimirova, I., & Kraft, J. (2002). Environmetric modeling and interpretation of river water monitoring data. *Analytical and Bioanalytical Chemistry, 374*(5), 898-905.

Singer, P. C., & Stumm, W. (1970). Acidic mine drainage: the rate-determining step. *Science, 167*(3921), 1121-1123.

Singh, R. K., Murty, H., Gupta, S., & Dikshit, A. (2007). Development of composite sustainability performance index for steel industry. *Ecological Indicators, 7*(3), 565-588.

Smith, D. G. (1990). A better water quality indexing system for rivers and streams. *Water Research, 24*(10), 1237-1244.

Smith, R. L. (1994). Use of Monte Carlo simulation for human exposure assessment at a superfund site. *Risk Analysis, 14*(4), 433-439.

Smolders, A., Lock, R., Van der Velde, G., Hoyos, R. M., & Roelofs, J. (2003). Effects of mining activities on heavy metal concentrations in water, sediment, and macroinvertebrates in different reaches of the Pilcomayo River, South America. *Archives of Environmental Contamination and Toxicology, 44*(3), 0314-0323.

Snodgrass, W. R. (1992). Physiological and biochemical differences between children and adults as determinants of toxic response to environmental pollutants. *Guzlean, et al., Similarities and Differences Between Children and Adults: Implications for Risk Assessment*. Washington DC. : ILSI Press. p 35-42.

Sonğur, C., & Top, M. (2016). Regional clustering of medical imaging technologies. *Computers in Human Behavior, 61*, 333-343.

Sorahan, T., & Lancashire, R. J. (1997). Lung cancer mortality in a cohort of workers employed at a cadmium recovery plant in the United States: an analysis with detailed job histories. *Occupational and Environmental Medicine, 54*(3), 194-201.

Sreeja, A., & Leela, E. (2008). (*Chromated copper arsenate (CCA) treatment for rubber wood preservation and its impact on the aquatic biota*) (Doctoral dissertation, Cochin University of Science and Technology, India.

Srinivas, R. G., & Nageswara, R. G. (2010). Study of groundwater quality in Greater Visakhapatnam City, Andhra Pradesh (India). *Journal of Environmental Science & Engineering, 52*(2), 137-146.

Srivastava, N., Harit, G., & Srivastava, R. (2009). A study of physico-chemical characteristics of lakes around Jaipur, India. *Journal of Environmental Biology, 30*(5), 889.

Staessen, J. A., Roels, H. A., Emelianov, D., Kuznetsova, T., Thijs, L., Vangronsveld, J., & Fagard, R. (1999). Environmental exposure to cadmium, forearm bone density, and risk of fractures: prospective population study. *The Lancet, 353*(9159), 1140-1144.

Subramani, T., Rajmohan, N., & Elango, L. (2010). Groundwater geochemistry and identification of hydrogeochemical processes in a hard rock region, Southern India. *Environmental Monitoring and Assessment, 162*(1-4), 123-137.

Sullivan, C. A., Meigh, J. R., & Giacomello, A. M. (2003, August). The water poverty index: development and application at the community scale. In *Natural Resources Forum 27(3)*, 189-199.

Sun, A. Y., Miranda, R. M., & Xu, X. (2015). Development of multi-metamodels to support surface water quality management and decision making. *Environmental Earth Sciences, 73*(1), 423-434.

Sun, H. F., Li, Y. H., Ji, Y. F., Yang, L. S., Wang, W. Y., & Li, H. R. (2010). Environmental contamination and health hazard of lead and cadmium around Chatian mercury mining deposit in western Hunan Province, China. *Transactions of Nonferrous Metals Society of China, 20*(2), 308-314.

Sundaray, S. K., Nayak, B. B., Lin, S., & Bhatta, D. (2011). Geochemical speciation and risk assessment of heavy metals in the river estuarine sediments—a case study: Mahanadi basin, India. *Journal of Hazardous Materials, 186*(2), 1837-1846.

Suthar, S., Nema, A. K., Chabukdhara, M., & Gupta, S. K. (2009). Assessment of metals in water and sediments of Hindon River, India: Impact of industrial and urban discharges. *Journal of Hazardous Materials, 171*(1), 1088-1095.

Swer, S., & Singh, O. (2004). (*Status of water quality in coal mining areas of Meghalaya, India*). Paper presented at the Proceedings of the National Seminar on Environmental Engineering with Special Emphasis on Mining Environment. 19th - 20th March, 2004. India.

Swistock, B., Sharpe, W., & Robillard, P. (2015). (*Iron and manganese in private water systems*). Pennsylvania, USA. The Pennsylvania State University.

Takaijudin, H., Shahir, A., & Hashim, A. (2012a). The utilization of an abandoned mining pond as a retention pond in dealing with stormwater runoff. *Flood Recovery, Innovation and Response III, 159*, 11127.

Takaijudin, H., Tajuddin, S. A., Kamil, S. A., Hashim, A. M., & Ishak, S. (2012b). (*Monitoring Stormwater Quality of Potential ex-Mining Ponds*). Paper presented at the International Conference on Environmental Science and Technology. 25th - 29th June, 2012. Texas, USA. 30, 35-40.

Takeda, A. (2003). Manganese action in brain function. *Brain Research Reviews, 41*(1), 79-87.

Tarantola S, J. J., Puolamaa M (2000). Global sensitivity analysis: a quality assurance tool in environmental policy modelling. In Saltelli A, Chan K, & Scott M (eds), (*Sensitivity analysis*). New York: Wiley.

Taylor, M. (1997). Accumulation of Cadmium derived from fertilisers in New Zealand soils. *Science of the Total Environment, 208*(1), 123-126.

Teh, G., & KIang, A. K. (2002). EPMA Characterisation and Geochemistry of Cas site rites from the Kuala Lumpur area. *Bulletin of Geological Society Malaysia, 45*, 363-368.

Tellez-Plaza, M., Navas-Acien, A., Crainiceanu, C. M., & Guallar, E. (2008). Cadmium exposure and hypertension in the 1999-2004 National Health and Nutrition Examination Survey (NHANES). *Environmental Health Perspectives*, *116(1)*, 51-56.

Thienpont, J. R., Korosi, J. B., Hargan, K. E., Williams, T., Eickmeyer, D. C., Kimpe, L. E., . . . Blais, J. M. (2016). Multi-trophic level response to extreme metal contamination from gold mining in a subarctic lake. *Proceeding of Royal Society B.* 283 (1836), 1125.

Thomas, R. (2008). (*Practical guide to ICP-MS: a tutorial for beginners*) (Second ed.). Florida: CRC Press, Boca Raton.

Thurtle, N., Greig, J., Cooney, L., Amitai, Y., Ariti, C., Brown, M. J., . . . Akpan, H. (2014). Description of 3,180 courses of chelation with dimercaptosuccinic acid in children≤ 5 y with severe lead poisoning in Zamfara, Northern Nigeria: a retrospective analysis of programme data. *PLoS Medicine, 11*(10), e1001739.

Tiwary, R. (2001). Environmental impact of coal mining on water regime and its management. *Water, Air, and Soil Pollution, 132*(1-2), 185-199.

Tiwary, R., & Dhar, B. (1994). Environmental pollution from coal mining activities in Damodar river basin, India. *Mine Water and the Environment*, *13*, 1-10.

Tripathee, L., Kang, S., Sharma, C. M., Rupakheti, D., Paudyal, R., Huang, J., & Sillanpää, M. (2016). Preliminary health risk assessment of potentially toxic metals in surface water of the Himalayan Rivers, Nepal. *Bulletin of Environmental Contamination and Toxicology*, *97*(6), 855-862.

Tripathi, R., Raghunath, R., Sastry, V., & Krishnamoorthy, T. (1999). Daily intake of heavy metals by infants through milk and milk products. *Science of the Total Environment, 227*(2), 229-235.

Tsuji, J. S., Benson, R., Schoof, R. A., & Hook, G. C. (2004). Health effect levels for risk assessment of childhood exposure to arsenic. *Regulatory Toxicology and Pharmacology, 39*(2), 99-110.

Tyagi, S., Sharma, B., Singh, P., & Dobhal, R. (2013). Water quality assessment in terms of water quality index. *American Journal of Water Resources, 1*(3), 34-38.

Udom, G., Nwankwoala, H., & Daniel, T. (2016). Determination of water quality index of shallow quaternary aquifer systems in ogbia, Bayelsa state, Nigeria. *British Journal of Earth Sciences Research, 4*(1), 23-37.

UNEP. (2010). *Final reviews of scientific information on Cadmium.* United nation environment programme, Chemicals branch DTIE. United nation document.

USEPA. (1987). *The Risk Assessment Guidelines of 1986.* EPA-600/8-87-045. 20460. Washington, DC.: United States Environmental Protection Agency.

USEPA. (1989). *Risk assessment guidance for Superfund*. Human health evaluation manual, [EPA/540/1-89/002]. Washington DC.: United States Environmental Protection Agency.

USEPA. (1992). *Guideline for exposure assessment*, EPA/600/Z-92/001,. Washington, DC: United States Environmental Protection Agency.

USEPA. (1997). *Exposure Factors Handbook.* Washington, DC.: United States Environmental Protection Agency.

USEPA. (1999). *Guidance for performing aggregate exposure and risk assessment.* Washington, DC.: United States Environmental Protection Agency.

USEPA. (2004). *Risk assessment guidance for superfund*: Human health evaluation manual (Part E, Supplemental Guidance for Dermal Risk Assessment). EPA/540/R/99/005. Washington, D.C.: United States Environmental Protection Agency.

USEPA. (2005). *Guideline for carcinogen risk assessments, EPA/630/P-03/001F*. Washington, DC.: United States Environmental Protection Agency.

USEPA. (2006). *Risk-based concentration table.* Washington D.C.: United States Environmental Protection Agency.

USEPA. (2011). *Exposure Factors Handbook.* Washington D.C.: United States Environmental Protection Agency.

Van Griensven, A., Meixner, T., Grunwald, S., Bishop, T., Diluzio, M., & Srinivasan, R. (2006). A global sensitivity analysis tool for the parameters of multi-variable catchment models. *Journal of Hydrology, 324*(1), 10-23.

Varmuza, K., & Filzmoser, P. (2016). *Introduction to multivariate statistical analysis in chemometrics*. New York: CRC press.

Varnavas, S. P. (2016). Manganese in drinking water and human health. A case study from Greece. *International Journal of Global Environmental Issues, 15*(1-2), 112-120.

Varol, M., Gökot, B., Bekleyen, A., & Şen, B. (2012). Water quality assessment and apportionment of pollution sources of Tigris River (Turkey) using multivariate statistical techniques—a case study. *River Research and Applications, 28*(9), 1428-1438.

Vasanthavigar, M., Srinivasamoorthy, K., Vijayaragavan, K., Ganthi, R. R., Chidambaram, S., Anandhan, P., . . . Vasudevan, S. (2010). Application of water quality index for groundwater quality assessment: Thirumanimuttar sub-basin, Tamilnadu, India. *Environmental Monitoring and Assessment, 171*(1-4), 595-609.

Vega, M., Pardo, R., Barrado, E., & Debán, L. (1998). Assessment of seasonal and polluting effects on the quality of river water by exploratory data analysis. *Water Research, 32*(12), 3581-3592.

Wallace, J., Champagne, P., & Hall, G. (2016). Multivariate statistical analysis of water chemistry conditions in three wastewater stabilization ponds with algae blooms and pH fluctuations. *Water Research, 96*, 155-165.

Wanda, E. M., Mamba, B. B., & Msagati, T. A. (2016). Determination of the water quality index ratings of water in the Mpumalanga and North West provinces, South Africa. *Physics and Chemistry of the Earth, Parts A/B/C, 92*, 70-78.

Wang, L., Cui, X., Cheng, H., Chen, F., Wang, J., Zhao, X., . . . Pu, X. (2015). A review of soil cadmium contamination in China including a health risk assessment. *Environmental Science and Pollution Research, 22*(21), 16441-16452.

Wang, X., Sato, T., Xing, B., & Tao, S. (2005). Health risks of heavy metals to the general public in Tianjin, China via consumption of vegetables and fish. *Science of the Total Environment, 350*(1), 28-37.

Wang, Z., Yao, L., Liu, G., & Liu, W. (2014). Heavy metals in water, sediments and submerged macrophytes in ponds around the Dianchi Lake, China. *Ecotoxicology and Environmental Safety, 107*, 200-206.

Westerhoff, P., Yoon, Y., Snyder, S., & Wert, E. (2005). Fate of endocrine-disruptor, pharmaceutical, and personal care product chemicals during simulated drinking water treatment processes. *Environmental Science & Technology, 39*(17), 6649-6663.

Whitehead, P., & Young, P. (1979). Water quality in river systems: Monte Carlo analysis. *Water Resources Research, 15*(2), 451-459.

WHO. (2001). *Arsenic and Arsenic compounds*. Environmental health criteria, Geneva : World Health Organization.

WHO. (2006a). *Establishing National Drinking-Water Standards*. Guidelines for Drinking Water Quality Training Pack. Geneva: World Health Organization.

WHO. (2006b). *Guidelines for drinking water quality*, Incorporating first addendum. Recommendations. Geneva: World Health Organization.

WHO. (2009). *Global health risks:* mortality and burden of disease attributable to selected major risks. Geneva: World Health Organization.

WHO. (2011). *Guidelines for drinking-water quality*: Geneva: World Health Organization.

WHO/UNEP. (1989). *Global fresh water quality*. World Health Organization, United Nations Environment Programme. Oxford, Blackwell Reference.

Williams, M. (2001). Arsenic in Mines Waters: An international study. *Environmental Geology*, *40*, 267-278.

Willingham, W. T., Thurston, R. V., Luedtke, R. J., & Russo, R. C. (2016). Toxicity of Ammonia and Nitrite to Aquatic Macroinvertebrates. *Intermountain Journal of Sciences*, *22*(4), 138-139.

Wolkersdorfer, C. (2008). *Water management at abandoned flooded underground mines*: fundamentals, tracer tests, modelling, water treatment. Berlin: Springer Science & Business Media.

Wongsasuluk, P., Chotpantarat, S., Siriwong, W., & Robson, M. (2014). Heavy metal contamination and human health risk assessment in drinking water from shallow groundwater wells in an agricultural area in Ubon Ratchathani province, Thailand. *Environmental Geochemistry and Health*, *36*(1), 169-182.

Woo, H. D., Chiu, W. A., Jo, S., & Kim, J. (2015). Benchmark Dose for Urinary Cadmium based on a Marker of Renal Dysfunction: A Meta-Analysis. *PloS one*, *10*(5), e0126680.

Woo, N., & Choi, M. (2001). Arsenic and metal contamination of water resources from mining wastes in Korea. *Environmental Geology*, *40*(3), 305-311.

Wu, B., Zhao, D., Jia, H., Zhang, Y., Zhang, X., & Cheng, S. (2009). Preliminary risk assessment of trace metal pollution in surface water from Yangtze River in Nanjing Section, China. *Bulletin of Environmental Contamination and Toxicology*, *82*(4), 405-409.

Wu, E. M. Y., & Kuo, S. L. (2012). Applying a multivariate statistical analysis model to evaluate the water quality of a watershed. *Water Environment Research*, *84*(12), 2075-2085.

Wurr, C., & Cooney, L. (2014). Ethical dilemmas in population-level treatment of lead poisoning in Zamfara State, Nigeria. *Public Health Ethics*, *7*(3), 298-300.

Xie, X., Ding, G., Cui, C., Chen, L., Gao, Y., Zhou, Y., . . . Tian, Y. (2013). The effects of low-level prenatal lead exposure on birth outcomes. *Environmental Pollution, 175*, 30-34.

Xing, W., & Liu, G. (2011). Iron biogeochemistry and its environmental impacts in freshwater lakes. *Fresenius Environmental Bulletin, 20*(6), 1339-1345.

Yaacob, Z., Pauzi, S., & Mutalib, H. (2009). Acid mine drainage and heavy metals contamination at abandoned and active mine sites in Pahang. *Bulletin of the Geological Society of Malaysia, 55*, 15-20.

Yabe, J., Nakayama, S. M., Ikenaka, Y., Yohannes, Y. B., Bortey-Sam, N., Oroszlany, B., . . . Ntapisha, J. (2015). Lead poisoning in children from townships in the vicinity of a lead–zinc mine in Kabwe, Zambia. *Chemosphere*, *119*, 941-947.

Yamada, M., Ohno, S., Okayasu, I., Okeda, R., Hatakeyama, S., Watanabe, H., . . . Tsukagoshi, H. (1986). Chronic manganese poisoning: a neuropathological study with determination of manganese distribution in the brain. *Acta Neuropathologica, 70*(3-4), 273-278.

Yang, Y., Chen, W., Wang, M., & Peng, C. (2016a). Regional accumulation characteristics of cadmium in vegetables: Influencing factors, transfer model and indication of soil threshold content. *Environmental Pollution*, *219*, 1036-1043.

Yang, R., Ning, Y., Zhang, A., Li, Y., & Su, H. (2016b). Analysis of Cu, Co, V and Zn in coastal waters of the East China Sea by inductively coupled plasma mass spectrometry (ICP-MS). *Journal of Ocean University of China*, *15*(5), 825-834.

Yang, Y., Wei, L., Cui, L., Zhang, M., & Wang, J. (2017). Profiles and Risk Assessment of Heavy Metals in Great Rift Lakes, Kenya. *CLEAN–Soil, Air, Water, 45(3).* 1-28.

Yao, H., Qian, X., Yin, H., Gao, H., & Wang, Y. (2015). Regional risk assessment for point source pollution based on a water quality model of the Taipu River, China. *Risk Analysis, 35*(2), 265-277.

Yap, C., Ismail, A., & Chiu, P. (2005). Concentrations of Cd, Cu and Zn in the fish tilapia Oreochromis mossambicus caught from a Kelana Jaya Pond. *Asian Journal of Water, Environment and Pollution, 2*(1), 65-70.

Yap, C., Ismail, A., & Chiu, P. (2007). Water quality and dissolved heavy metal concentrations in surface water collected from Kelana Jaya Lakes. *Asian Journal of Water, Environment and Pollution*, *4*(1), 187-190.

Yen, T. P., & Rohasliney, H. (2013). Status of water quality subject to sand mining in the Kelantan River, Kelantan. *Tropical Life Sciences Research*, *24*(1), 19-34.

Yi, Y., Yang, Z., & Zhang, S. (2011). Ecological risk assessment of heavy metals in sediment and human health risk assessment of heavy metals in fishes in the middle and lower reaches of the Yangtze River basin. *Environmental Pollution, 159*(10), 2575-2585.

Yidana, S. M., & Yidana, A. (2010). Assessing water quality using water quality index and multivariate analysis. *Environmental Earth Sciences, 59*(7), 1461-1473.

Yilmaz, E. (2011). Advances in reducing large volumes of environmentally harmful mine waste rocks and tailings. *Gospodarka Surowcami Mineralnymi, 27*, 89-112.

You, S., Lu, J., Tang, C. Y., & Wang, X. (2017). Rejection of heavy metals in acidic wastewater by a novel thin-film inorganic forward osmosis membrane. *Chemical Engineering Journal, 320*, 532-538.

Younger, P. L., Banwart, S. A., & Hedin, R. S. (2002a). *Mine Water Hydrology*. New York: Springer.

Younger, P. L., Banwart, S. A., & Hedin, R. S. (2002b). *Mine water: hydrology, pollution, remediation.* New York: Springer Science & Business Media.

Younger, P. L., & Wolkersdorfer, C. (2004). Mining impacts on the fresh water environment: technical and managerial guidelines for catchment scale management. *Mine Water and the Environment, 23*, s2-s80.

Yusof, A., Mahat, M., Omar, N., & Wood, A. (2001). Water quality studies in an aquatic environment of disused tin-mining pools and in drinking water. *Ecological Engineering, 16*(3), 405-414.

Zaini, M. A., Lim, C., Low, W., & Harun, F. (2005). Factors affecting nutritional status of Malaysian primary school children. *Asia-Pacific Journal of Public Health, 17*(2), 71-80.

Zakir, H., Islam, M., Arafat, M., & Sharmin, S. (2013). Hydrogeochemistry and quality assessment of waters of an open coal mine area in a developing country: a case study from Barapukuria, Bangladesh. *International Journal of Geoscience Research, 1(1),* 20-44.

Zaldivar, R. (1977). Ecological investigations on arsenic dietary intake and endemic chronic poisoning in man: dose-response curve. *Zentralblatt fur Bakteriologie, Parasitenkunde, Infektionskrankheiten und Hygiene. Erste Abteilung Originale. Reihe B: Hygiene, praventive Medizin, 164*(5-6), 481-484.

Zandbergen, P. A., & Hall, K. J. (1998). Analysis of the British Columbia water quality index for watershed managers: a case study of two small watersheds. *Water Quality Research Journal Canada, 33(4),* 519-549.

Zhang, X., Yang, L., Li, Y., Li, H., Wang, W., & Ye, B. (2012). Impacts of lead/zinc mining and smelting on the environment and human health in China. *Environmental Monitoring and Assessment, 184*(4), 2261-2273.

Zhang, Y., Guo, F., Meng, W., & Wang, X. Q. (2009). Water quality assessment and source identification of Daliao river basin using multivariate statistical methods. *Environmental Monitoring and Assessment, 152*(1-4), 105-121.

Zhang, Y., Shi, K., Zhou, Y., Liu, X., & Qin, B. (2016). Monitoring the river plume induced by heavy rainfall events in large, shallow, Lake Taihu using MODIS 250m imagery. *Remote Sensing of Environment*, *173*, 109-121.

Zhao, Y., Xia, X., Yang, Z., & Wang, F. (2012). Assessment of water quality in Baiyangdian Lake using multivariate statistical techniques. *Procedia Environmental Sciences, 13*, 1213-1226.

Zhi, G., Chen, Y., Liao, Z., Walther, M., & Yuan, X. (2016). Comprehensive assessment of eutrophication status based on Monte Carlo–triangular fuzzy numbers model: site study of Dongting Lake, Mid-South China. *Environmental Earth Sciences, 75*(12), 1-11.

Zhou, F., Guo, H., Liu, Y., & Jiang, Y. (2007). Chemometrics data analysis of marine water quality and source identification in Southern Hong Kong. *Marine Pollution Bulletin*, *54*(6), 745-756.

Zhou, L., & Boyd, C. E. (2015). An assessment of total ammonia nitrogen concentration in Alabama (USA) ictalurid catfish ponds and the possible risk of ammonia toxicity. *Aquaculture, 437*, 263-269.

Zhuang, P., Lu, H., Li, Z., Zou, B., & McBride, M. B. (2014). Multiple exposure and effects assessment of heavy metals in the population near mining area in South China. *PloS one, 9*(4), e94484.

Zhuang, P., McBride, M. B., Xia, H., Li, N., & Li, Z. (2009). Health risk from heavy metals via consumption of food crops in the vicinity of Dabaoshan mine, South China. *Science of the Total Environment, 407*(5), 1551-1561.

Zimdahl, R. L., Arvik, J. H., & Hammond, P. B. (1973). Lead in soils and plants: A literature review. *Critical Reviews in Environmental Science and Technology, 3*(1-4), 213-224.

LIST OF PUBLICATIONS AND PAPERS PRESENTED

A) Publications

1 Koki, I. B., Low, K. H., Juahir, H., Azid, A., & Zain, S. M. (2017). Assessment of water quality of man-made lakes in Klang Valley (Malaysia) using chemometrics: the impact of mining. *Desalination and Water Treatment*, *74*, 125-136.

2. Low, K. H., Isa Baba, K., Hafizan, J., Azman, A., Shima, B., Rabia, I., Hamisu A. M., .Sharifuddin, M. Z. (2016). Evaluation of water quality variation in lakes, rivers, and ex-mining ponds in Malaysia (review). *Desalination and Water Treatment,57(58),* 28215- 28239.

B) Papers Presented

1. I. B. Koki**,** K. H. Low**,** H. Juahir, A. Azid, S. Behkhami, R. Ikram, H. M. Aliyu, S. M. Zain (2016). (*Evaluation of Water Quality Variation in Lakes, Rivers and Ex Mining Ponds in Malaysia*). International Conference on Environmental and Natural Science (ICENS), Kuala Lumpur, Malaysia 20th December 2015.

2. I. B. Koki, S. M. Zain, K. H. Low, H. Juahir, A. Azid. (*Evaluation of lake water quality in Klang Valley (Malaysia) using Multivariate statistical techniques*). The 16th World Lake Conference Bali Indonesia, 7th – 11th November 2016

3. I. B. Koki, S. M. Zain, K. H. Low, H. Juahir, A. Azid. (*Application of Chemometric Techniques in the Evaluation of Water Quality of Ex-mining ponds and lakes in Malaysia*). The 13th Asian Conference on Analytical Sciences (ASIANALYSSIS XIII), Chiang Mai Thailand, 8th – 11th December 2016

4. I. B. Koki, S. M. Zain, K. H. Low, H. Juahir, M. B. Sale. (*A study on health risk assessment of exposure to ex-mining water in Klang Valley Malaysia*). The 6th International Conference for Young Chemist, University Sains Malaysia, Penang, Malaysia, 16th – 18th August 2017