STUDY OF INORGANIC POLLUTANT IN URBAN LAKES AND ASSESSMENT OF *Riccia fluitans* TO REMEDIATE Fe, Mn, Zn, Cu AND Pb TOXICITY THROUGH IN VIVO MODEL SYSTEM

BY

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ABSTRACT

Heavy metals represent a serious form of pollution as they do not easily degrade and have a tendency to bioaccumulate. Malaysian waters suffered with 30 to 100 times more pathogens, heavy metals and poisons from industry and agriculture above than government standards permit. A presence of excessive nutrients input to a water bodies resulting increased biomass, nuisance algal blooms, species fatality due to low or depleted oxygen caused impairment of freshwater and lake aging. A broad number of physico-chemical parameters and biological characteristics render the degree of quality of water resources. This research aimed to study the effectiveness and ability of selected aquatic plants as ecological indicators for monitoring and assessing aquatic environmental conditions. To achieve this ten different locations were selected as site studies which located in the district of Selangor and Kuala Lumpur. All of these sites are urban lakes which have been identified and observed based on the lake surrounding and abundance of aquatic plant life. These sites were chosen based on its location, human and physical characteristics as well as for their tolerance to eutrophic and heavy metal contaminants through various physico-chemical parameters and water quality analysis. In order to assess type of aquatic plant species that can tolerate low, medium and high concentrations of contaminants an in vivo model system with a fully controlled environment in laboratory scale was established. In this model system, metals accumulation from different concentration of heavy metals was investigated using *Riccia fluitans* to assess the effectiveness of selected aquatic species in remediating urban lake contaminants. Analysis of nutrient contaminants from ten different urban lakes and a total of nine different aquatic plant species which are submergent (*Hydrilla verticillata* and *Cabomba caroliniana*), emergent (*Eleocharis ochrostachys* and *Ipomoea aquatic*) and free-floating (*Nymphaea lotus, Nymphaea pubescens* and *Nelumbo* nucifera) and Algae sp. demonstrated highly significant differences ($P< 0.0001$) between aquatic plant species widespread, locations and the nutrients content. Results indicated that *E. ochrostachys* was a good phytoindicator for NO_3^- excess, *H. verticillata* and *I. aquatica* were efficient phytoindicator for PO_4^{3-}, Algae species for Mn, *N. pubescens* and *C. caroliniana* for Fe whereas *H. verticillata* for Zn. Interestingly all measured lakes were found at the stage of hypereutrophic or exceeded the normal level of NWQS for Malaysian rivers which is 0.2 mg/L (Class II and III). Analysis of heavy metals sequestration rate by *R. fluitans* at three different concentrations (1.0, 2.0 and 5.0 mg l^{-1}) and at four different periods of time (week 1 to week 4) revealed that this species was a good agent to sequester Cu>Mn>Zn>Fe>Pb at 60.4%, 20.3%, 5.4%, 1.3% and 1.3%, respectively. The analysis affirmed the accumulation of different metals within the plant and an equally lessen of metals in the water. As a conclusion, *Riccia fluitans* is proven to be a potential biosequester agent for polluted aquatic environment. As a whole, the abundance or loss of certain key species can be used as indicator for eutrophication state and level for urban lakes management and maintenance as well as confirmed the aquatic macrophytes is a promising candidate as a phytoremediation agent.

ملخص البحث

إنَّ إضافة المزيد من المغذيات إلى المسطحات المائية يؤدي إلى ازدياد الكتلة الحيوية، والطحالب المزعجة، وموت الأصناف بسبب انخفاض أو نضوب الأكسجين المذاب مما يؤدي إلى ضعف المياه العذبة والبحيرات. وهناك العديد من العوامل الفيزيائية والكيميائية والخصائص البيولوجية التي تؤثر على درجة الجودة للموارد المائية. وقد تم إجراء عدة دراسات لتحديد قيمة النتريت والنترات والفوسفات في محطات مختلفه لبحيرات لمناطق حضريه في كوالاميور وسلانجور وكذلك لتخمين قابلية *Riccia fluitans* على علاج النبات من التلوث بالمعادن الثقيلة في وسط مائي غير صحي. وكنتيجة لذلك فإنَّ جميع البحيرات مشبعة غذائياً حيث وجد أن تركيز الفوسفات قد تجاوز الحد الاعتيادي للـ NWQS للأنهار في ماليزيا والبالغ 0.2 مغ لكل لتر (للاصناف 2 و 3). حالة التشبع الغذائي للمياه بالاعتماد على (TSI). وعلى رأس كل الأنواع التي تم تحليلها، هناك ثلاث انواع يعتمد عليها كمؤشرات لتحديد التشبع الغذائي وهي: (NO_3^-) *Eleocharis ochrostachys* و *Hydrilla verticillata Ipomoea aquatica* (PO_4^{3-}). وقد ظهر من خلال تحليل الملوثات الغذائية لعشر بحيرات في مناطق حضرية مختلفة أن تسعة منها مطمورة (*Hydrilla verticillata*)، وظاهرة (*Eleocharis ochrostachys* و*Ipomoea aquatica*) وطافية (*Nymphaea lotus*، *Nymphaea pubescens*، *Nelumbo nucifera*) وأنواع طحالب *Cabomba caroliniana* منتشرة وباختلافات عالية وواضحة ($P< 0.0001$) بين الأنواع المائية النباتية واسعة الانتشار ومواقع ومحتوى العناصر الغذائية. لاختبار استعداد *Riccia fluitans*، تم تعريض النبات لثلاث معادن ثقيلة (منغنيز، رصاص وزنك) وبتراكيز مختلفه (1، 2 و 5 مغ لكل لتر) وبفترات زمنيه مختلفه (اسبوع الى اربعة اسابيع). وكنتيجة بين خمسة معادن مستخدمة، أظهرت *Riccia fluitans* نتائج للعزل حسب الترتيب Cu>Mn>Zn>Fe>Pb عند 60.4%، 20.3%، 5.4% و 1.3%. وقد أكدت التحاليل بأن تراكم المعادن المختلفة داخل النبات يقلل بنفس القدر من نسبة المعادن في الماء. وأن التكافؤ بين نوع المعادن الثقيلة والتركيز، والوقت و *Riccia fluitans* قد تحقق. والخلاصة أن *Riccia fluitans* قد اثبتت بأنها عامل عزل للوساط المائية الملوثة. على العموم فإنّ وفرة صنف معين أو تناقصه يمكن ان يستخدم كمؤشر للتشبع ولمستوى ادارة البحيرات وصيانتها، وكذلك يمكن أن يستخدم كعامل لعلاج النباتات.

APPROVAL PAGE

I certify that I have supervised and read this study and that in my opinion, it conforms to acceptable standards of scholarly presentation and is fully adequate, in scope and quality, as a thesis for the degree of Master of Science (Built Environment).

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DECLARATION

I hereby declare that this thesis is the result of my own investigations, except where otherwise stated. I also declare that it has not been previously or concurrently submitted as a whole for any other degrees at IIUM or other institutions.

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^{o}C	degree
=	equal
>	more than
%	percent
±	standard deviation

LIST OF ABBREVIATIONS

%	Percent	NO	Nitric oxide
<	Less than	K	Kalium
Cd	Cadmium	N	Nitrogen
Cr	Cromium	DO	Dissolved oxygen
Cu	Copper	$mg\ l^{-1}$	Miligrams per litre
DW	Dry weight	NO_x	Nitrogen oxide
Fe	Iron	NO_3^-N	Nitrate as nitrogen
Hg	Mercury	P	Phosphorus
m^3	Cubic meter	TSI	Trophic states Index
Mn	Manganese	NH_4	Ammonium
N_2O	Nitrous oxide	$mg\ g^{-1}$	Miligrams per gram
Ni	Nickel	ISC	Impervious surface cover
NO_2^-	Nitrite	g/cm3	Gram per cubic centimetre
NO_3^-	Nitrate	COD	Chemical oxygen demand
Pb	Lead	BOD	Biochemical oxygen demand
PO_4^{3-}	Phosphate	$\mu g\ l^{-1}$	Microgram per liter
UN	United Nations	TDS	Total Dissolved Solid
Zn	Zinc	NH_4^-N	Ammonium as N

CHAPTER 1

INTRODUCTION

1.1 RESEARCH BACKGROUND

Water plays a vital role as it roundly affects human activity and the earth's climate. Societies have benefited tremendously through their freshwater usage. Mankind has caused depletion and degradation of global freshwater ecosystems by extracting and consuming quantities of fresh water from the environment that far exceed the rate for which those supplies are replenished by the natural water cycle. Key examples of human-induced changes include alteration of the natural flow regimes in rivers and waterways, fragmentation and loss of aquatic habitat, species extinction, water pollution, depletion of groundwater aquifers, and ‘‘dead zones’’ (aquatic systems deprived of oxygen) found in many inland and coastal waters. Therefore, tons of effort have been carried out clearly between human and natural system requirements for fresh water purification services.

1.1.1 Global

Numerous researches have shown major factors in the increasing global water usage and global demand for fresh water which are largely driven by rapid population growth and agricultural intensification. Despite these factors, the demand for water is also rising because of urbanization, economic development, and improved living standards. The Millennium Ecosystem Assessment projected that by 2025, 3.5 billion people will be unable to feed themselves due to freshwater scarcity (MEA, 2005). Solomon et al., (2007) briefly explained the relationship between climate change and the freshwater crisis. He concluded that climate changes could cause damage through greater variability and extremity of water related events such as droughts, floods, mudslides, rising sea levels and glacier-melting that overpower water infrastructures built for traditional weather patterns. His research, like many others, highlighted how the water crisis is inseparably linked to climate change. Humans are already appropriating more than half of all accessible surface water runoff, and this may increase to 70% by 2025. The three largest water users in global terms are: agriculture (67%), industry (19%), and both municipal and residential (9%) (United Nations, 2004).

The world is fast approaching an era of severe water shortage. Though Asia is the largest and most populous continent with a population of 60%, it benefits from only 36% of the world’s water resources. Based on the EM-DAT database, from 1980 to 2006, the recorded victims of water-related disasters were estimated around 1.36 million of which Asia accounted for 46%, and the ratio of people affected by water-related disasters in Asia accounted for 90%. Surprisingly, there has been a yearly increase of 1% in overall total freshwater withdrawals since the late 1980s. This is expected to rise to over 44% by 2050 due to growing demand from manufacturing, thermal power generation (mainly from the expansion of coal and gas powered plants), agriculture and domestic use (WWD, 2014). The scarcity and impurity of water resources are enraged by physical, economic, and environmental conditions. Thus, freshwater regions are jeopardized by pollution, poor supervision, and climate change. There is an approximate 263 major international river basin in the world, which covers 231 059 898 km² or 45.3% of the Earth’s land surface (excluding Antarctica) (UNEP, 2002).

Recent evidence has shown that due to massive exploitation of global aquifers, groundwater supplies have decreased by 20% (WWD, 2014). In the 20th century, there has been massive water withdrawal for the agriculture sector and the water usage for this sector has greatly affected crop production. The agriculture sector is a major user of water and has unfortunately caused major water pollution from excess nutrients, pesticides, and other pollutants. At present and more so in the future, agriculture faces enormous challenges due to pressure from urbanisation, industrialisation, and climate change. A water withdrawal for agriculture accounts for some 80% of global withdrawal and the majority of it is used for irrigation purposes (Siebert et al., 2010). Regrettably, the increasing use of fertilizers and pesticides to improve agricultural production has contaminated the available water supply in many regions. In contrast, Europe and North America currently use more water in industry compared to agriculture. Numerous studies have proven that agriculture is one the main sources of diffuse pollution and a leading source of phosphorus compounds and sediments. Two important pollutants associated with diffuse pollution are nitrogen and phosphorus. Excessive nutrients, loaded into water bodies can be harmful to the ecosystem. It will affect the dissolved oxygen and suffocate fish and other aquatic life. Agriculture has been a primary concern since the 1970s and a main cause of the incremental amount of nitrogen, phosphorus, and pesticide runoff into surface and groundwater. The largest source of non-point pollutants to surface and groundwater pollution comes from the growth of aquaculture-based. Among domestic, industrial, and agricultural use, agriculture remains the leading source of phosphorus compounds and sediments based on a comparison made among coastal zones along the Mediterranean. According to Spalding et al., (1993), the most common chemical contaminant in the world's groundwater and aquifers is nitrate. 90% of the world's freshwater resources are from groundwater, which is reducing 2% per year. This situation adds to the water stress. Surprisingly, some 1.5 billion people depend on this as their daily drinking water (UNEP, 2002; WWD, 2014). Based on results from surveys in India and Africa, approximately 50% of wells contain nitrate levels higher than 50 milligrams per litre, and in some cases as high as several hundred milligrams per litre. In the last decade, nitrate concentrations levelled up in most watersheds in the Americas, Europe, Australia, and mainly in Africa and the eastern Mediterranean (FAO, 1996).

Land is always associated with water pollution as these two components are precious natural resources which rely on the sustainability of agriculture and the civilization of mankind. In spite of eutrophication issues, heavy metals are also a major threat to freshwater bodies. Heavy metals could discharge from an assortment of sources, including point and non-point sources. Point sources are from emission, effluents, and solid discharge from industries. Meanwhile, non-point sources come from pesticides, disposal of industrial and municipal wastes in agriculture, and excessive use of fertilizers (Lone et al., 2008). All countries are affected by diffuse pollution, but the area and severity differ. In Western Europe, 1,400,000 sites were affected by heavy metals (McGrath et al., 2001), of which, over 300,000 were contaminated. The estimated total number in Europe could be much larger, as pollution problems occur more regularly in Central and Eastern European countries (Gade, 2000).

Domestic sewage is among the major concerns as it directly affects aquatic ecosystems, particularly near densely populated areas. Approximately 150-250 million m^3 per day of untreated wastewater from urban areas are channelled into open water bodies or absorbed into the subsoil. The affects human health, the rising rate of infant fatality,and widespread environmental degradation (WWDR4, 2012). Water use demands are at a rate double of that for the population rate. It is expected that more than 1700 million people, namely two-thirds of the world population will live with water scarcity. Over 80% of global used water is not collected or treated (Corcoran et al., 2010).

1.1.2 Local

Malaysia is prioritizing water resources and its role in the development of the country. In general, Peninsular Malaysia receives 342 billion m^3 of rainwater whereas the current demand is about 11 billion m^3 annually (Mamun and Zainudin, 2013). This number is expected to increase to 18 billion m^3 in 2050. Despite the richness of water resources in Malaysia, uneven distribution has led to water excess and shortage (Chan, 2006). The water network supply sometimes fails to distribute water to high demand areas, and water turns unusable and polluted when it reaches the urban centres. In other words, Malaysia is facing an issue of declining quality of raw water and water scarcity is not a common phenomenon in Malaysia (Mamun and Zainudin, 2013). Looking at river water quality trends in Malaysia (recorded in DOE Malaysia), rivers in Malaysia are becoming increasingly polluted. As Malaysia is fast becoming an industrial country, many rivers are fast becoming a wastebasket for chemicals, sewage disposal, and pollutants. In Malaysia, the Department of Environment (DOE) is charged with monitoring river water quality using the Water Quality Index (WQI). Over 1000 manual and automatic river water quality monitoring stations in 146 basins are being monitored by the Department of Environment (DOE) with the help from other agencies such as the Department of Irrigation and Drainage (DID) and state agencies. DOE adopts the Interim National Water Quality Standard for Malaysia (INWQS) as a tool to measure and assess the river water quality and classification.

The Water Quality Index (WQI) is used to reflect the water quality status based on the comparison of water quality parameters.

In spite of the effluence from point sources, pollutants from non-point sources (urban and rural runoff) are diversely affecting water quality. Management of these two sources needs to be taken seriously to sustain the value of water resources. This reflects poor performance after the implementation of the EQA which is responsible to control the flow of non-point and point (mainly) sources. Drop-off water quality interrupts the drinking water supply. About 60% from 90 selected lakes and reservoirs were eutrophic and primarily contain nitrates and phosphates resulting from runoff fertilizers and pesticide use in agriculture (NAHRIM, 2005; Sharip et al., 2007). However, untreated or weakly treated sewage disposal is still a prime point pollution of rivers. Point sources are believed to be channelled constantly to a water body and then transmit organic and inorganic particles, nutrients, pesticides and herbicides (Cleophas et al., 2013) which directly affect the aquatic ecosystem.

1.1.3 Factors Affecting Water Quality

Most countries experience water scarcity. The causes of this vary. They can be either natural or human activity. Human activity has roundly impacted on the quality of water resources. These include sedimentation, pollution, climate change, deforestation, landscape changes, and urban growth. Water resources are placed under increasing pressure from human activity such as urbanisation, changes in population, increased living standards, water demanding supply and pollution. All of these are influenced by climate change and changing natural conditions.

Climate change has remarkably influenced the pattern of water availability. As a whole, climate change adversely affects biodiversity, changing the species diversity, affecting their habitats and niches of others, but most significantly on water scarcity (Morrison et al., 2009). This is supported by Chakraborty et al., (2011) and Bates et al., (2008), who pointed out that climate change has greatly influenced the world's freshwater resources, water quality, and water management. Human and ecosystem health is affected the most when there is a change in water temperature and runoff. This has led to critical changes in surface-water quality. In most developing countries, climate change is considered an additional stressor to many present degraded systems, for instance, agriculture, forestry, fisheries, and ecosystems (Morrison et al., 2009). Alterations on water quality and temperature are also greatly affected by urban, industrial, and agricultural use including on aquatic ecosystems. Degradability in water quality influences the cost of water purification for urban water usage. Meanwhile, the incremental level in turbidity, nutrients and pathogen content of surface water sources, influences the amount of precipitation (IPCC, 2007). Surface-water quality is needed by water temperature as it controls the survival of aquatic life, stimulates the sum of dissolved oxygen in the water, and manipulates chemical and biological reactions. Higher surface-water temperature caused by climate change will hasten biological productivity, level up the sum of bacteria and fungi in the water and promote growth of algal (Kundzewicz et al., 2007). The relationship between wastewater and climate change can be categorised into the following three parts (UNEP, 2010):

1. Water volume, availability, and quality changes and directly influences water usage practices.
2. Wastewater management should be a high priority as it is a central part of the urban environment, to food production, industry, human health and the environment as a whole.

3. Wastewater treatment results in the emission of greenhouse gases, particularly carbon dioxide (CO_2) and methane (CH_4) and nitrous oxide (N_2O).

Human activities simultaneously affect the changes in water quality which occurs naturally along the river. Some of them are from industries, agriculture, and human settlements which lead to excessive nutrient concentrations and poisonous pollutants. Rapid pace urbanisation and industrialisation have gained increased public interest, especially among those moving to cities and towns. This led to more physical disturbance of land, insufficient sewage disposal and treatment, increase amounts nutrients (lead to eutrophication), heavy metals washed out into streams and groundwater by mining industries, and increased soil erosion due to the physical disturbance of soil and vegetation.

The world is facing a rapidly increasing set of water quality challenges. It is widely recognised that many countries are suffering from severe water scarcity. The prime reason behind this phenomenon is because of urbanisation and increased water demands to meet human needs. Water quality is affected the most by both human activities and natural factors. Agriculture is the most cited causes in alteration water quality as it diversely channels nutrient and pesticide contamination and levelled up salinity. Nutrient enrichment is a global water quality problem known as eutrophication as it elevates growth of algae and depletes BOD. The body of water becomes stagnant and devoid of aquatic life ecosystem. Meanwhile, climate change is a worldwide additional stressor to most affected countries as it accelerates biological reactions and affects the ecosystem. When water pollution loads into water surface directly going beneath the surface, it is contributing to a high incidence of water-related disease. Lack and poor management of wastewater could add more severe pollution, which influences the health of human population and the environment. The management should not solely focus on urban but also the rural context. The effects of wastewater directly enters water bodies can be controlled and minimised by the enhancement in ecosystem management, these include agriculture, forestry, livestock, wetland and riparian.

1.2 PROBLEM STATEMENT

Technology, human activities, success in economic growth, and industrialisation has caused environmental problems leading to increased levels of land, air, and water pollution. Toxic elements in both soil and water are serious environmental problems that threaten human health and the environment. Malaysian rivers have become a wastebasket for chemicals, sewage disposal, and pollutants. Most of lakes and reservoirs in Malaysian are classified as eutrophic and primarily contaminated by nitrates and phosphates resulting from runoff fertilizers and pesticide use in agriculture. Various mechanisms exist to treat these problems. A method using living aquatic plants to remove metals from water is an alternative cost-effective, efficient, and technologically feasible. The use of plants for remediation of metals offers an attractive alternative because it is solar-driven and can be carried out *in situ*, minimising cost and human exposure.

1.3 ISSUES ON WATER QUALITY

1.3.1 Water Pollution

Asian rivers are the most polluted in the world with three times more bacteria from human waste as the global average, and 20 times more lead than industrialised countries. Based on a report by Executive Summary UN World Water Development, 300-500 million tonnes of heavy metals, solvents, toxic sludge and other wastes accumulate annually in water resources from industries. Because of contamination in drinking water and poor sanitation, more than 2.2 million people die from related diseases. The report stated that about 2 million tonnes of waste are dumped directly into rivers, lakes, and streams. This has caused some 6000 people, mostly children under the age of five to die from diarrheal diseases. Furthermore, every 1 litre of wastewater is able to pollute about 8 liters of fresh water. There is an estimated 12,000km of polluted water worldwide, which is more than the total amount of water contained in the world’s largest river basins. 97% of water is salty and almost 3% is locked away in ice gaps, glaciers, or deep underground. It is estimated that only 0.003% is actually available for use. This is enough to show that our water is facing critical pollution and if there is nothing done to address this problem, there might be no more safe water to consume.

Based on research, more than 63% of the rivers in Malaysia are classified as moderately to highly pollute. The Malaysian rivers receive urban runoff polluted with domestic sewage discharges and livestock excreta, as well as from agricultural uses and wastewater from factories (Sim et al., 2008). Excess nutrients are the main agricultural pollutants in Malaysia. Drainage and irrigation channels, ponds and other waterways are polluted by agricultural runoff from fertilizer rich land, such as vegetable farms, fruits and flower nurseries, golf courses, and animal farms. A study of water quality changes in Chini Lake, Pahang, showed that the constructed downstream can disrupt the structure and the function of the river ecosystem by modifying flow regimes, disrupting sediment transport, altering water quality, and severing their biological continuity (Othman et al., 2007). Chini Lake experienced