MODELLING OF ROAD SURFACE POLLUTION BUILDUP AND WASHOFF USING RAINFALL SIMULATOR

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requirements for the award of the degree of
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Specially dedicated to my father, mother, wife, children, brothers and sisters I don't have enough words to thank you, for your immense support, care, and love.

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ABSTRACT

Water quality management of nonpoint source (NPS) pollution is still being confronted with identification and assessment. The extent of pollution due to NPS in tropics is not yet affirmed, and the relative influences of its associated sources were not yet totally understood. This study explored the significance of road as a NPS unit in tropical region of persistent rainfall, and investigated the possible sources of heavy metals in urban areas. To achieve the objectives of this study, the natural rainfall dynamic of the study area was appraised using the flour pellet method. The information was used as a basis for developing a highly efficient Rainfall Simulator (RS) that was used to investigate pollutant washoff process under different rainfall depth and intensities. A total of 30 buildup samples were collected from five chosen roads of varying characteristics, and fractionated into 10 classes of particle sizes each. For quantitative analysis, 60 samples were analysed for dissolved Zn, Fe, Cd, Pb, Cu, Ni, Mn, Al, and Cr concentrations. A multivariate principal component and factor analyses were used to investigate the likely sources of these heavy metals. Three sources were identified, the indigenous, geogenic and scavenge. The natural raindrop sizes were found to vary from less than 1.2 mm to as big as 7.0 mm with median raindrop diameters (D_{50}) of 2.51 mm and a mean of 2.56 mm. These raindrops have an average rain kinetic energy content of 30 J m^{-2} mm^{-1}. The developed RS can satisfactorily simulate rain intensity similar to natural rainfall, with an average kinetic energy content of 42 J m^{-2} mm^{-1} and a D_{50} between 2.41 and 2.64 mm. An advanced principal component and cluster analysis identified TDS as a surrogate for measuring dissolved metals pollution among eight physicochemical parameters considered, and was therefore used in the modelling of the washoff process. The developed models suggested that the rain intensity plays a more prominent role in the occurrence of first flush, while the rain depth plays a central role in the total washoff event. This research demonstrated that the influence of sediment to retain mass loading did not necessarily translate to higher pollution loading of heavy metals, and the residency of heavy metals in different particle classes cannot be generalised.

ABSTRAK

Pengurusan kualiti air yang melibatkan pencemaran punca bukan titik (NPS) masih dihambat dengan isu pengenalpastian dan penilaian. Tahap pengurusan NPS di rantau tropika masih belum mantap dan faktor-faktor relatif yang mempengaruhi punca pencemaran ini masih belum difahami keseluruhannya. Kajian ini meneroka kepentingan jalan raya sebagai unit NPS di rantau tropika yang menerima hujan sepanjang tahun, dan menyiasat punca sumber logam berat dalam kawasan bandar. Bagi mencapai objektif kajian ini, kedinamikan hujan semula jadi di kawasan kajian telah diteliti menggunakan kaedah pelet tepung. Maklumat ini dijadikan asas untuk membangunkan simulator hujan (RS) yang effisien untuk menyiasat proses basuhan pencemaran bagi taburan hujan yang berbeza kedalaman dan keamatan. Sebanyak 30 sampel penumpukan telah diambil dari lima permukaan jalan raya yang mempunyai ciri-ciri berlainan. Dan diasingkan secara berperingkat kepada 10 kelas saiz partikel. Untuk penentuan kualiti, sebanyak 60 sampel telah dianalisis untuk kandungan Zn, Fe, Cd, Pb, Cu, Ni, Mn, Al, dan Cr. Kaedah komponen utama multivariat dan analisis faktor telah digunakan untuk menentukan punca logam berat. Tiga punca utama telah dikenalpasti, iaitu punca setempat, punca geogenik dan punca luar kawasan. Saiz titisan hujan semula jadi didapati berbeza-beza dengan garispusat kurang dari 1.2 mm hingga sebesar 7.0 mm dengan nilai median (D_{50}) 2.51 mm dan purata 2.56 mm. Titisan hujan ini mengandungi purata tenaga kinetik sebanyak 30 J m^{-2} mm^{-1}. RS yang dibangunkan telah dapat mensimulasi keamatan hujan semulajadi dengan memuaskan dengan purata kandungan tenaga kinetik 42 J m^{-2} mm^{-1} dan D_{50} di antara 2.41 dan 2.64 mm. Hasil analisis lanjutan komponen utama dan kluster bagi lapan parameter fisikokimia mendapati bahawa TDS sesuai dijadikan parameter wakil untuk mengukur tahap pencemaran logam dan boleh digunakan dalam pemodelan proses basuhan. Model yang dibangunkan mencadangkan bahawa keamatan hujan memainkan peranan penting dalam penyahan pertama, manakala kedalaman hujan mempunyai peranan penting dalam menentukan keseluruhan kejadian basuhan. Kajian ini menunjukkan bahawa pengaruh sedimen dalam menjerap beban jisim tidak semestinya menghasilkan beban logam berat yang lebih tinggi dan hayat logam berat yang dijerap oleh sedimen dengan saiz berbeza tidak boleh ditentukan secara umum.

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LIST OF ABBREVIATIONS

ADD	-	Antecedent dry day(s)
ADT	-	Average daily traffic
ARI	-	Average return interval
Bi	-	Bismuth (chemical element, with atomic number 83)
BOD	-	Biological oxygen demand
Co	-	Cobalt(chemical element, with atomic number 27)
COD	-	Chemical oxygen demand
Cr	-	Chromium (chemical element, with atomic number 24)
D	-	Drop diameter
D50	-	Mean drop diameter
DCIA	-	Directly connected impervious area
DO	-	Dissolved oxygen
DOC	-	Dissolved organic carbon
DSD	-	Drop size distribution
EC	-	Electrical conductivity
EMC	-	Event mean concentration
FA	-	Factor analysis
F:C	-	fine to coarse ratio
GIS	-	Geographic information system
HEPA	-	High-Efficiency Particulate Air
I	-	Rain intensity
IC	-	Inorganic carbon
IDF	-	Intensity-Duration-Frequency
KE	-	Kinetic energy
KEmm	-	Kinetic energy (raindepth)
KEtime	-	Kinetic energy (time)
MHCM	-	Malaysian Highway manual
MTD	-	Mean texture depth
NPS	-	Nonpoint source
PAR	-	Predicted to actual ration

PCA	-	Principal components analysis
PCCA	-	Principal components and classification analysis
PSD	-	particle size distribution
PS	-	Point source
R	-	Rain depth
RDS	-	Road dust sediment
RS	-	Rainfall simulator
TDS	-	Total dissolved solids
TC	-	Total carbon
TOC	-	Total organic carbon
TS	-	Total solids
TSS	-	Total suspended solids

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CHAPTER 1

INTRODUCTION

1.1 General

Nonpoint source (NPS) pollution is generally considered to be a diffuse source of pollution not associated with a specific temporal point of entry into the water body (Wyoming, 1999). NPS can further be defined as anything other than pollutants that enter the runoff transport routes at discrete identifiable locations that can usually be measured (Loague and Corwin, 2006). Point source (PS) pollutants are generally more toxic, and can readily be identified, quantified, and controlled. NPS pollution, on the other hand, is typically difficult or impossible to trace to a source (Loague and Corwin, 2005; Novotny, 2003; Stein *et al.*, 2006), quantify (Candela *et al.*, 2009; Ferrier *et al.*, 2005) and control (Albiac *et al.*, 2009; Ferrier *et al.*, 2005) making implementation of effluent limitations almost impossible because they vary with the seasons and the weather (Carpenter *et al.*, 1998). The NPS clean-up is costly and nearly impossible to accomplish (Loague *et al.*, 1998), it also poses a more technical problem because of the extent of their contamination, which increases the amount of data required far beyond that of PS pollutants (Wagenet and Corwin, 1996). Therefore, the understanding of different surfaces' sources and responses to the generation, transportation and transformation of NPS pollutants to receiving stream is crucial to any successful implementation of mitigating measures.

1.2 Research Background

Water quality management in developing countries, including Malaysia, is still being confronted with identification, assessment and control of nonpoint source pollution. Since, the majority of the pollutants are of diffuse origin, an in-depth understanding of the possible pollutant's pathways, and their contribution is of essence for drawing strategy to reduce their impacts on receiving waters (Ferrier *et al.*, 2005).

Tropical regions, like Malaysia, have higher susceptibility to NPS pollution for both rural and urban areas (Al-Mamun, 2005) and data on pollutant buildup and washoff on distinct impervious surfaces in the tropics is extremely lacking and necessary for improved water quality (Rahmat, 2005; Chow, 2011). Generally, researchers have different understanding on the contribution from various sources of pollution to the overall loading of NPS, from severe (Quek, 1993; Si mmons, 2001; Chang *et al.*, 2004) to moderate (Lee, 1982; Pazwash, 1997). This Perhaps is largely because NPS is associated to certain uncontrollable climatic events, specific to geographic and geologic conditions, and may vary significantly in time and space (Zakaria *et al.*, 2002; Novotny, 2003).

Surface runoff accounts for over 50% runoff volume in metropolitan area (Forster, 1996; Jain and Ali, 2001). Roads play a central role in urban hydrology, and their runoff is the most important of all runoffs from municipal area (Murakami *et al.*, 2004); therefore, road runoff is one of the areas where knowledge of science and engineering is inevitably necessary when strategies for optimum solution of an environmental problem have to be developed. So, the contribution of roads' runoff to the pollution of lakes and streams in Malaysia cannot be over emphasised.

1.3 Statement of the Problem

NPS inputs are the major source of water pollution today, and their impacts are profound with urban runoff ranked third most important causative agent of lake deterioration in the U. S. (Carpenter *et al.*, 1998) and second in Agricultural watershed in Malaysia (Eisakhani *et al.*, 2009). Although the threat of NPS pollutants differs throughout the world, they are generally of global importance because NPS pollution problems do not recognise the boundaries between nations, nor are they necessarily isolated by the physical barriers between continents (Corwin and Wagenet, 1996).

Globally, there were many studies on NPS pollution over the years and commensurate government and private sector participation but NPS is still a major source of water quality impairment in rivers, lakes and estuaries, and remain the number one threat to water quality in the US (Gannon, 1996; Griffith, 1999). Over a century, this problem has been recognised and systematically studied in the developed countries with enactment of laws and mitigating measures, and some gains have been made. However, very little has been done to study and curtail the emergence of NPS in the developing countries like Malaysia.

Malaysia has enjoyed remarkable growth over the last few decades, with industrialization, agriculture, and tourism playing leading roles in its success story. However, today, despite its relatively positive environmental record, it faces problems of pollution from diffuse sources due to rapid increase in its population and urbanization (Yusop *et al.*, 2005; Yusop *et al.*, 2007; Zainudin *et al.*, 2009; Chow *et al.*, 2012; Chow *et al.*, 2013). The cutting down of trees to accommodate large industrial factories and human shelters, the clearance of more agricultural land to meets food demand and boost national income were also a leading cause of water pollution in Malaysia (Malakahmad *et al.*, 2008; Eisakhani *et al.*, 2009). Migration of people to industrial and commercial centres further accelerates the transformation of its small towns to municipalities, and existing urban landscape into more densely populated areas. Moreover, not only did the industries depleting oxygen supply and spewing out poisonous gases during their production, the increase of cars for transporting goods and services in and around urban centres were also lending a hand in the pollution (Abdullah, 1995). This trend becomes accelerated with the shift of Malaysia's development strategy, from agro based to manufacturing in the last four decades, in addition to the adoption of effective management systems for the handling of agricultural waste. This development ensures declined concern of agro-based pollution from 67% in 1980 to a steady state of 15% in the 90s (DOE, 1991).

The states in the western peninsular Malaysia were more prone to alarming level of anthropogenic contamination due to intensified urbanisation (Abdullah, 1995), Recently, Nazahiyah *et al.* (2007) studied the suspended solids (SS), biochemical oxygen demand (BOD_5), chemical oxygen demand (COD), nutrients (NO_3-N, NO_2-N, NH_3-N and P) and metal Pb in runoff from a residential catchment of 85% imperviousness in Skudai, Johor. They observed variations of BOD_5, COD, SS, and nutrients, in both the washoff and in the first flush amongst different storms. Similarly, Chow and Yusop (2009) carried out similar study in a different residential catchments. The result showed an elevated levels of BOD_5, COD, TSS, and O&G in the storm runoff samples. They also observed that natural rainfall intensity has influence on the washoff of TSS, and O&G, while the concentrations of BOD_5, and COD were influenced by ADD. Chow *et al.* (2011) found that the site mean concentrations in runoff from commercial catchment in Johor exceeded that reported in the Texas, and

Florida in the US; Saskatoon in Canada; and Shanghai, China; but lower than reported value in ChongJu, Korea. They attributed this to non-effective NPS management implementation. Chow *et al.* (2012) modelled the quantity and quality of runoff from residential, commercial, and industrial areas in Johor using SWMM to establish and compare the nature of pollutants buildup and washoff using local data. They concluded that buildup in Malaysia was small compared with the temperate zones, and its progression was limited due to its frequent rainfall.

There were few conducted studies on NPS pollution in Malaysia (e.g. Abdullah, 1995; Al-Mamun, 2005; Nazahiyah, 2005; Malakahmad *et al.*, 2008; Eisakhani *et al.*, 2009; Zainudin *et al.,* 2009; Chow, 2011). These studies were localised to only urban and agricultural catchment scales. Among them, Al-Mamun (2005), Nazahiyah, (2005) and Chow (2011) seems to be pioneers in conducting a detailed study on urban catchment nonpoint source pollution. Presently, there is need to investigate the role played by various urban surfaces like road in the urban NPS pollution. Road surfaces can significantly contribute to the retention of heavy metals and sediments during dry weather periods as a result of atmospheric deposition, and other local sources. These accumulated pollutants are the major threats to the urban environment due to their conveyance to the surrounding catchments during wet weather washoff process (Yusop *et al.*, 2005). The study of the Malaysian river and estuaries by Zakaria *et al.* (2002) concluded that the receiving water bodies were heavily polluted. They reported concentrations of organic and inorganic compounds from different land uses.

This research hopes to deepen or open a new discovery on the role of roads as a source of NPS pollution in urban setup. It will also investigate the possible primary sources where these pollutants originates. The implication of different rain events on the transportation of these pollutants during the wet-weather washoff process will also be appraised. The outcome from this study would ultimately deepen the understanding of NPS and their likely primary sources in an urban landscape. The result can be of utmost help to managers for a holistic control, by targeting the sources where pollutants originate.

1.4 Research Objectives

This research is aimed to close up the existing data gap on the role of road as an impervious unit in urban landscape and its relative contributions to the overall NPS pollution load in Malaysia. Specifically, this study objectives are to:

1. Determine the influence of natural rain intensity on the raindrop diameter and its kinetics.
2. Develop an artificial rainfall simulator that will replicate the natural rainfall of the study area as closely as possible.
3. Identify the possible primary sources where heavy metals originate using a multivariate analytical technique.
4. Determine a heavy metal pollution surrogate, and investigate the transportation of pollutants during a wet weather washoff process on different road surfaces under different rain event.

1.5 Scope of the Study

Since the extent of diffuse pollution is related to certain uncontrollable climatic events, as well as geographic and geologic conditions, and may differ greatly from place to place and from year to year (Novotny, 2003, Forster, 1998, Egodawatta *et al.*, 2009) this research will undertake the following to achieve the objectives of the study.

1. Appraisal of natural rainfall characteristics of the study area.
2. Development of an artificial rainfall simulator to generate rainfall parameters similar to the natural rainfall of the study area.
3. Laboratory analyses to determine the concentration of heavy metals (Zn, Fe, Cd, Pb, Cu, Ni, Mn, Al and Cr), the carbon matrices [Inorganic carbon(IC), total carbon (TC), and total organic carbon (TOC)] and the associated physicochemical parameters [dissolve oxygen (DO), Electrical conductivity (EC), total dissolve solids (TDS), total suspended solids (TSS), salinity, and pH].
4. Investigate possible sources of heavy metals on roads using a principal component analysis (PCA), and factor analysis.

5. Empirical analysis to establish the washoff relationship of pollutants with different roads type, under different rain intensity and duration.

1.6 Significance of the Study

Pollution profile and their sources differed between nations, and the mitigation approaches used in industrialized countries cannot often be applied directly in the developing countries, and each country is saddled with the responsibility of developing its own based on its prevailing conditions. Therefore, this work will be of great importance to government, individuals and private organisations to better understand the pattern of buildup and washoff on road surfaces under tropical condition. This study would also present a clear understanding of pollutant's origins, and the washoff dependants influencing factors.

1.7 Identification of Research Methodology

Due to unpredictability of runoff quality which is reported to be sensitive to metrological conditions (Novotny, 2003; Vialle *et al.*, 2011) and its complexity as a function of different environmental parameters (He *et al.*, 2000). This work used laboratory simulation to answer questions of the research. The laboratory simulation on one hand, allowed the adjustment of certain influencing parameters which will otherwise be impossible in the field within reasonable time frame and on the other hand its necessity as a pathway for an in depth understanding of patterns and variability that may exist between buildup and washoff processes. So, the methodology are grouped in the following hierarchal order:

1. Literature review.
2. Design and fabrication of rainfall simulator based on literature.
3. Selection of study site and rainfall characteristics for simulation.
4. Collection, testing and characterisation of buildup and washoff samples.

The schematic flow chart of the methodology is presented in Figure 1.1.

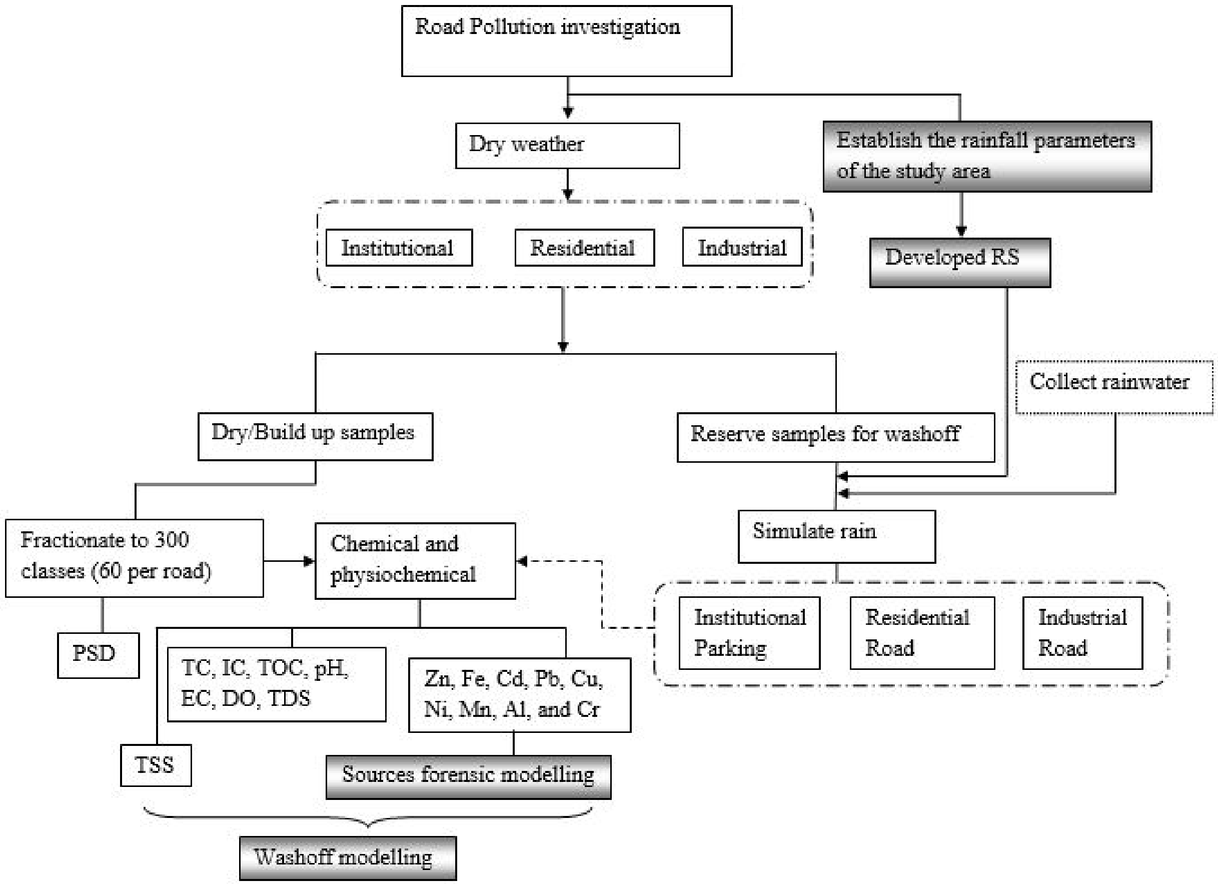

Figure 1.1: The schematic flow chart of the methodology

1.7.1 Literature Review

Extensive literature was carried out to gain a comprehensive knowledge of wet weather urban pollution, characteristics, influential parameters, response on different surfaces and impact. Specifically the aims are to:

1. understand urban hydrological changes and responses due to increase population and imperviousness.
2. understand sources, types, composition and characteristics of pollutants from different urban surfaces.
3. present contemporary knowledge in pollutant buildup and washoff process under different rainfall characteristics (intensity and duration).

1.7.2 Development of a rainfall simulator

The rainfall simulator was designed to ensure it can reproduce rainfall parameters as closely as possible, and was such that it can model the spatial variation of different rainfall intensities and rainfall depth. The component and setup of the simulator were such that the RS was easy to transport, assemble and dismantled. Plots were setup such that it was easy for instant measurement of runoff at any given time. The RS minimised generation of excessive volume of rain within short period of time by incorporating an oscillating boom. This arrangement has ensured spatial and temporal variation of the rainfall.

1.7.3 Selection of study site and sampling

Five potential road surfaces were selected based on their land uses and their exposure to local sources. To differentiate the pollutants originating from atmospheric deposition and that originating from the localised sources, the roads' location were strategically chosen to receive similar atmospheric fall outs. Samples were collected during the dry weather period, and analysed for nine metals (Zn, Fe, Cd, Pb, Cu, Ni, Cr, Mn, and Al). These chosen metals were of particular interest to water runoff pollution researchers (Papiri *et al.*, 2008; Poleto *et al.*, 2009a; Zhao *et al.*, 2011).

The washoff samples were collected using the simulated rain. The simulation took place on three chosen locations. A total of 22 events were simulated for washoff study. Because of the nature of washoff, the flow weighted measure was employed to each subsamples in proportion to the inflow volume and effluent. For quality control, duplicate samples were provided to ensure the repeatability and precision of the obtained result. Care was exercised to ensure there was no foreign substrate introduced into the sampling and throughout the experimental procedures.

Heavy metals can be quantified as total or soluble analytes, the latter is generally of environmental concern due to it's bio-availability (Duncan *et al.*, 2007). Buildup samples were filtered through 0.45μm filter, and petitioned for physicochemical, and heavy metal analyses.

1.8 Organisation of Thesis

Chapter 1 presents, the intents and the motivation to undertake this study. The problem statement was conceptualised and presented. In addition, it gives the framework on how these research objectives will be achieved, and the lead benefits of undertaking the research.

Chapter 2 recapitulates the past and current knowledge in the area of the urban NPS pollution. It gives an overview of the relationship between urban hydrology, and the influence of increasing urban sealed surfaces on the pollution level. It described in detail the importance of particle in vectoring heavy metals in the environment and the importance of their source identification.

Chapter 3 this chapter gives an overview of the peculiarity of the study region in terms of its rainfall dynamics, and how it shaped the sampling protocol implemented in obtaining the data. It described in details the procedure followed in carrying out the experimental work of the study. It specifically described the sample collection, storage, preparation, processing and analyses. It also justified the choice of the sampling equipment, and the calibration of the RS. An overview of principal component and factor analysis was also undertaken in detail.

Chapter 4 discussed the result of the study area's rainfall governing parameters, the modelled rainfall drop diameter, and its distribution. The knowledge of the natural rainfall characteristics of the study area was used as a baseline in the development of a RS. The chapter reported in details, the choice of the RS components that ensured replication of the natural rainfall's characteristics. It also reported extensively on the evaluation and calibration of the RS. The chapter also reported the screening of the data, and further gives an additional information on the suitability of the identified method for further analysis.

Chapter 5 deals with investigating the possible sources of the heavy metals in roads, and the modelling of heavy metals in washoff process. The heavy metal sources investigation was undertaken using the principal component, and the Factor multivariate analyses to identify the possible sources of the heavy metals in a typical tropical urban environment. The particle mass analysis was also undertaken to further investigate the influence of different land uses, and the impact of a longevity factors in urban NPS. This chapter also presented the descriptive modelling of the washoff process under different rainfall depth and duration using a defined surrogate measure of dissolved heavy metals' concentration. The washoff models were established using selected typical rainfall duration of the study area.

Chapter 6 presented the summary of the findings from this study, and highlighted areas that needs to be researched further.

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