MULTIMEDIA ENVIRONMENTAL FATE AND TRANSPORT MODEL OF DICHLORODIPHENYLTRICHLOETHANE AT SAYONG RIVER WATERSHED BASED ON FUGACITY APPROACH

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To my beloved parents, husband, siblings, and supervisors…….

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ABSTRACT

Dichlorodiphenyltrichloroethane (DDT) is one of the most concerning compounds in the group of persistent organic pollutants (POPs) due to its persistence and harmfulness on environment. This thesis details the development of a multimedia environmental fate and transport model to assess the distribution and transfer processes of DDT in air, soil, water and sediment in the Sayong River watershed. Geographical Informational System (GIS) was employed to divide the watershed into up, mid and down-streams. The levels of DDT in the air, soil, water and sediment in the Sayong River watershed were monitored the period of between November 2014 and May 2015. Samples were collected and extracted through Solid Phase Extraction (SPE) and ultrasonication. Extracted samples were analyzed using Gas Chromatography-Mass Spectrometry (GC-MS). For the development of model, the compartments and tranfer processes were setup along with the steady state and chemical equilibrium assumptions. Fugacity concept was used to formulate the distribution mechanism processes. Input parameters, consisting of chemical emission data, environmental properties and physical-chemical properties, were selected as secondary data. Microsoft Excel-Visual Basic Application (VBA) was used to encode the calculation. The total concentrations of DDT were observed to be in the range of 5.25-53.53 µg/g for soil, 0.22-37.88 µg/g for sediment and 0-0.38 µg/g for air. Meanwhile, there was no DDT found in the water samples. In addition, the model predicted reasonably accurate concentrations within an order of magnitude (0.01-0.25) in log unit. The advection outflow in air was determined to be the most important process of DDT in this model with the rate range of 0.12-0.26 mol/h. From the sensitivity analysis, the vapour pressure (Ps) and organic carbon - water partition coefficient (KOC) were concluded to be the most influential parameters where the Sensitivity Coefficient (SC) being higher than 0.5. This model is important as it can provide an efficient and cost effective measure to assess the fate and movement of DDT in the Sayong River watershed.

ABSTRAK

Dichlorodiphenyltrichloroethane (DDT) merupakan salah satu sebatian penting dalam kumpulan pencemar organik berterusan (POPs) kerana ia berbahaya dan sukar terurai terhadap alam sekitar. Tesis ini menerangkan pembangunan model multimedia taburan akhir dan proses pemindahan pencemar untuk menilai taburan dan proses pemindahan DDT dalam udara, tanah, air dan sedimen di kawasan tadahan air Sungai Sayong. Sistem Maklumat Geografi (GIS) telah digunakan untuk membuat pembahagian kawasan tadahan kepada hulu, pertengahan and hilir. Tahap DDT dalam air, udara, tanah dan sedimen telah dipantau dalam tempoh antara November 2014 dan Mei 2015. Sampel telah diambil dan diekstrak melalui pengekstrak fasa pejal dan ultrasonik. Sampel yang diekstrak kemudian dianalisis menggunakan Kromatografi Gas – Spektrometer Jisim (GC-MS). Untuk pembangunan model bahagian dan proses pemindahan telah dirangka. Andaian dibuat untuk keadaan tetap, bahagian bercampur dan keseimbangan kimia. Pendekatan fugasiti digunakan untuk memformulasi taburan dan proses pemindahan DDT dalam udara, air, tanah dan sedimen di kawasan tadahan Sungai Sayong. Parameter masukan terdiri daripada data pelepasan kimia, sifat alam sekitar dan sifat fizikal dan kimia bahan pencemar telah diambil sebagai data sekunder. Aplikasi Visual Basic (VBA) dari MS Excel telah digunakan untuk mengekod pengiraan. Kepekatan DDT dikesan wujud pada julat 5.25-53.53 µg/g untuk tanah, 0.22-37.88 µg/g untuk sedimen and 0-0.38 µg/g untuk udara. Sementara itu, tiada DDT dijumpai dalam sampel air. Kepekatan yang diramal oleh model adalah tepat dalam aturan magnitud (0.01-0.25) dalam unit log. Aliran keluar udara telah diputuskan sebagai proses DDT yang terpenting dalam model ini dengan kadar julat 0.12-0.26 mol/j. Analisis sensitiviti model menunjukkan tekanan wap air (Ps) dan pekali pembahagi karbon organik-air (KOC) telah disimpulkan sebagai parameter yang paling berpengaruh di mana pekali sensitiviti (SC) lebih tinggi daripada 0.5. Model ini sangat penting kerana ia menyediakan satu alat yang efisyen dan menjimatkan kos untuk menilai taburan dan pemindahan DDT di kawasan tadahan air Sungai Sayong.

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LIST OF ABBREVIATION/SYMBOLS

POPs	Persistence Organic Pollutants
OCPs	Organochlorine pesticides
LRT	Long-Range Transport
DDT	Dichlorodiphenyltrichloroethane
MEM	Multimedia Environmental Model
GIS	Geographical Informational System
VBA	Visual Basic Application
SPE	Solid Phase Extraction
GC-MS	Gas chromatography-mass spectrometry
A_{AIR}	Advective in air
R_{AIR}	Degradative reactive in air
R_{SOIL}	Degradative reactive in soil
A_{WATER}	Advective in water
R_{WATER}	Degradative reactive in water
$R_{SEDIMENT}$	Degradative rective in sediment
SC	Sensitivity Coefficient
C_i	Concentration DDT in a compartment
C_R	Residue concentration
D	Transfer rate coefficient
T	Residence time
V	Volume
Z	Fugacity capacity
F	Fugacity
K	Reaction half life
R	Gas constrant
H	Henry's Law constant
T	Ambient temperature

ϕOC	Organic carbon content
Cm	Centimetre
ng/g	Nanogram per gram
ng/L	Nanogram per liter
m^3	Meter cubic
m^2	Meter square
Pa	Pascal
K	Kelvin
K_{OC}	Organic carbon-water partition coefficient
Log K_{OW}	Log octanol-water partition coefficient
D value	Tranfer rate coefficient
Z value	Fugacity capacity

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CHAPTER 1

INTRODUCTION

1.1 Background of Research

Over the past, environmental deterioration and natural resource destruction occurred due to agricultural and industrial development (Sultana *et al.*, 2014). In chemical management and environmental decision-making, it is very important to assess the regional ecological and human health risk of chemicals released into the environment (Liu *et al.*, 2014). For this purpose, sampling and analysis methods are always used by researcher, but unfortunately in real environment, dynamic behavior of pollutants cannot be studied. Moreover, these methods are laborious in work, long time-consuming and expensive (Wang *et al.*, 2012). Thus, a new tool called multimedia environmental modeling (MEM) has been introduced to predict the level distribution of a contaminant in all connected environment (Luo *et al.*, 2007). Therefore, in Decision Support System (DSS) for chemical risk assessment, this model can be applied and is urgently needed for management of persistent organic pollutants (POPs) in Malaysia. The most important thing, this study provide a useful tool for chemical fate and transport assessment, especially at Sayong River watershed.

As an agricultural country, pesticides are widely used in Malaysia. Among large numbers of pesticides, organochlorine pesticides (OCPs) included as POPs cause wide attention from environmental researcher and became an important environmental problem in public (Kim *et al.*, 2015). Several OCPs such as dichlorodiphenyltrichloroethane (DDT), dieldrin and hexachlorocyclohexanes

(HCH) have been withdrawn or banned in many countries for environmental reasons and public health (Mahugija *et al.*, 2014). Because of good effect in controlling insects and relatively low cost, a number of OCPs are still in use in South Asian Countries such as Malaysia even though prohibitions on their uses have been implemented in developing countries (Usman *et al.*, 2014). Due to the ability of OCPs to accumulate mainly in animal tissue and enter the food chain, thus they are considered as toxic substances (Luzardo *et al.*, 2012). OCPs are categorized under long range transport (LRT) compounds, thus they were able to be transported far from its initial point source (Usman *et al.*, 2014). OCPs were found in the various environmental phases such as organisms, water, soil, suspended particulate matter (SPM), atmosphere and sediment due to their LRT characteristic (Yu *et al.*, 2014).

Mathematically, fugacity approach has been widely used to describe the environmental behaviour of organic pollutants in local environment, regional and global environments (Xiangzhen *et al.*, 2014). The concept of fugacity and mass balance principle are suitable to describe the partitioning processes in different environmental compartments and predict the concentration level, distribution and persistence of the chemicals (Liu *et al.*, 2014).

Sayong River is one of the major tributaries at Johor River basin. It is an important source of freshwater supply, not only for Johor State but also for Singapore. This watershed is dominated by natural forest and oil palm plantation (Jabatan Pengairan dan Saliran Johor, 2010). The river 122.7 km long with total watershed area of 480 238 $km^{2.}$. In general, the economic activities at the watershed are oil palm plantation with in the area of 57701 km^2 or 88.49 % of the total watershed area. Downstream area has the largest oil palm plantation with 47.77 % of total oil plantation area followed by midstream 27.03 % and upstream 25.19 %. Besides, other agricultural activities are also conducted at Sayong River watershed such as vegetables farming at 0.22 % of the total watershed area. Other than oil palm plantation and agriculture, the watershed also has urban area at 2.21 % where most of this area is filled with residential, schools, offices and shops. Unexplored land at the watershed is only 5.60 %.

The aim of this study is to provide a multimedia environmental fate and transport model of DDT in air, soil, water and sediment at Sayong River watershed. Sampling activity was carried out from November 2014 until May 2015 to monitor the distribution of DDT in air, soil, water and sediment. Data of DDT collected in this study is very important to validate the developed fugacity model.

1.2 Problem Statement

A global distillation effect cause DDT to be distributed widely in various regions and it can still be detected in different environmental media, even though since 1980s, it had been restricted in several countries including China. The South Asia region including Malaysia is a place where primarily emissions are still taking place and thus it is important to assess the current status of dichlorodiphenyltrichloroethane (DDT) pollution (Usman *et al.*, 2014). DDT was banned in Malaysia on 1999 because of its persistent effect (Jabatan Pertanian Negeri Pahang, 2016).

For in-use and historic use of DDT, both monitoring and modeling method play complementary roles. Both methods were used widely for many chemical fate and transport assessment in multimedia environments. However, monitoring method was more costly, laborious and time consuming to assess DDT in multimedia environments. Moreover, this method cannot be used to determine the transport rate of a chemical in multimedia environment. Therefore, a fast and inexpensive mechanisms need to be used to assess the fate and transport of DDT in multimedia environment. This method is known as multimedia environmental fate and transport model. In Malaysia, there are still lack of work on simulation fate and transport of pesticides in multimedia environments. Thus, this thesis provides a fast and inexpensive method for government agency to trace any pesticides in multimedia environments especially at watershed level.

1.3 Objectives

The aim of this study is to develop a model that can be used as a tool for prediction of chemical fate and transport of dichlorodiphenyltrichloroethane (DDT) in air, soil, water and sediment at Sayong River watershed. Thus, specific objectives to be achieved are as follows:

1. To investigate data on DDT distributions in air, soil, water and sediment at Sayong River Watershed.
2. To develop a multimedia chemical fate and transport model of DDT at Sayong River watershed using fugacity approach.
3. To validate the model via comparison of measured and modeled data using log-difference method.
4. To analyze sensitivity of the model to input parameters using Sensitivity Coefficient (SC) method.

1.4 Scope of Study

Sayong River watershed is one of the major tributaries at Johor River Basin. This watershed needs to be managed properly because Sayong River is an important water source in Johor. Moreover, the main land use activity at the watershed is oil palm plantation. The use of many agricultural chemicals for this plantation might affect the environment and quality of river water. Therefore, Geographical Informational System (GIS) was used the for study area and watershed division in this research.

To provide distribution data of dichlorodiphenyltrichloroethane (DDT) at Sayong River watershed, sampling activities were conducted for air, soil, water and sediment phases at the divided watershed area. Then, the samples extraction processes and analysis were carried out to measure the distribution level of dichlorodiphenyltrichloroethane (DDT) in air, soil, water and sediment at Sayong River watershed.

For the model development, compartments and mechanisms processes treated in the model were setup for each divided watershed area. Only two environmental loss processes were considered in this model which was advection out and degradation processes. The level of the model (fugacity level II) only treats mechanism loss processes. The environmental distribution and mechanism processes were formulated using fugacity concept. Model assumptions such as homogenous, equilibrium and steady state were considered at this formulation stage.

The collection of secondary data such as chemical emission data, environmental properties and physical-chemical properties of dichlorodiphenyltrichloroethane (DDT) were also carried out .The calculations were carried out by using Microsoft Excel – Visual Basic Application (VBA).

1.5 Significance of Study

Significance of study are as follows;

This study is very important since screening processes conducted on OCPs showed that DDT detected in samples and found high in soil and sediment. This study provides a model which can be a tool to assess the fate and transport of a chemical in multimedia environments at watershed level. Therefore, the fate and transport of dichlorodiphenyltrichloroethane (DDT) in air, soil, water and sediment at Sayong River watershed can be determined using this model. Next, the development of this model was very important since it provides a fast and inexpensive mechanism for the determination of dichlorodiphenyltrichloroethane (DDT) fate and transport in multimedia environments. Development of new model in this study is important because of region specific for examples Malaysia’s climate is different with China’s climate.

1.6 Thesis Organisation

This thesis is organized into four chapters. Chapter 1 is the introductory chapter which contains background of study, problem statement, objectives and significance of study. Chapter 2 presents in detail some related literature reviews and similar previous works. Several topics related to this study are review to give an overall picture of the background knowledge required for this work. Chapter 3 deals with the methodology employed in this study. Chapter 4 discusses the monitoring data of DDT at Sayong River watershed and simulated concentration and transport processes of DDT in air, soil, water and sediment at Sayong River watershed. Chapter 5 concludes the research findings and suggests potential future work.

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