UNIVERSITI PUTRA MALAYSIA

DERMAL EXPOSURE ASSESSMENT OF GLYPHOSATE HERBICIDE AMONG PESTICIDE HANDLERS IN SELECTED AGRICULTURE SECTORS IN MALAYSIA

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By

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Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Doctor of Philosophy

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

Malaysian farmers are still relying on pesticides to govern agriculture activities. Occupational exposure to pesticides can adversely affect farmers' health, especially those in developing countries where the methods for evaluating dermal exposure are not reasonably well standardized. Therefore, this study aimed to assess dermal exposure and health symptoms among pesticide handlers in selected agriculture sectors in Malaysia (i.e., paddy, vegetable, cocoa and oil palm). This cross-sectional study was conducted in 2016 in four sampling locations, been divided into three sub-studies which are; 1) Dermal exposure assessment using DREAM, 2) Dermal exposure assessment using whole-body dosimetry (WBD) method and 3) Identification of health symptoms. In the first sub-study, socio-demographic characteristics, information on pesticide application and dermal exposure information were assessed. Then, total potential (tPDE) and actual dermal exposure (tADE) among pesticide handlers were determined using DREAM model, and factors affecting the exposure were analysed using logistic regression analysis. In the second sub-study, the concentration of uranine as pesticide surrogate and total PDE were determined using the WBD method. The correlations between the measurements with tPDE from DREAM (Sub-study 1) were then analyzed by estimating the Spearman correlation coefficient. In the third sub-study, health symptoms of skin problems and pesticide poisoning perceived by respondents after pesticide spraying were identified. Then, the association between DREAM data and perceived health symptoms was determined through logistic regression analysis. Based on DREAM, tPDE estimates between different agricultural sectors were significantly different (χ^2 = 118.093, p < 0.001). The prominent highest exposure was recorded among paddy field sprayers (151.39 ± 22.64 DU), and the lowest exposure was among respondents in oil palm plantation (47.67 ± 18.47 DU). The factors significantly related to high tPDE (≥100 Dermal Unit) were pesticide deposition, sprayer's age, gloves usage and immediate clothes changing after pesticide spraying. The total amount of uranine on Tyvek sections of respondents in the paddy field was 0.281 mg/kg, followed by respondents in oil palm (0.043 mg/kg), vegetable (0.042 mg/kg) and cocoa farming (0.013 mg/kg). The total potential dermal exposure (PDE) was significantly highest among respondents in paddy field with a mean total PDE of 1.56E-03±1.73E-04 (p < 0.001). A significant positive correlation was found between logged total PDE estimates from DREAM and logged total PDE measured through WBD (r_s =0.752, p=0.001). Most of the respondents in the paddy field reported having skin itchiness (53.3%) and itchy wheals (24.4%), while respondents in vegetable farm reported increased salivation (29.4%), diarrhoea (14.7%), urinary frequency (23.5%), cramps (26.5%), muscle weakness (29.4%) and restlessness (17.6%). Total PDE from DREAM was the only variable that significantly contributed to the logistic regression model associated with skin problems and pesticide poisoning symptoms. Our study supports that DREAM able to provide estimates of exposure levels among respondents, gives insight into the distribution and route of dermal exposure, and can be associated with health symptoms. Nevertheless, DREAM estimates on variability in the distribution of dermal exposure on body parts over the measurement data should be further verified for model improvement. (494 words)

Keywords: Dermal exposure; pesticide; DREAM model; whole-body dosimetry; health symptoms

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

PENILAIAN PENDEDAHAN KULIT TERHADAP RACUN GLIFOSAT DI KALANGAN PENGENDALI RACUN PEROSAK DALAM SEKTOR PERTANIAN DI MALAYSIA

Oleh

NURULAIN BINTI MUSTAFA UDIN

Mei 2021

Pengerusi : Sharifah Norkhadijah binti Syed Ismail, PhD
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Petani di Malaysia masih bergantung kepada racun perosak untuk menjalankan aktiviti pertanian. Pendedahan terhadap racun perosak semasa pengendalian boleh menjejaskan kesihatan petani, terutamanya di negara membangun di mana kaedah untuk menilai pendedahan kulit tidak seragamkan dengan baik. Oleh itu, kajian ini bertujuan menilai pendedahan kulit dan gejala kesihatan di kalangan pengendali racun perosak di beberapa sektor pertanian terpilih di Malaysia (iaitu padi, sayur, koko dan kelapa sawit). Kajian keratan rentas ini dijalankan pada tahun 2016 di empat lokasi persampelan, dibahagikan kepada tiga sub-kajian iaitu; 1) Penilaian pendedahan kulit melalui *DREAM*, 2) Penilaian pendedahan kulit melalui kaedah *whole-body dosimetry* (*WBD*) dan 3) Penilaian gejala kesihatan. Dalam sub-kajian pertama, ciri sosio-demografi, maklumat penggunaan racun makhluk perosak, dan maklumat pendedahan pada kulit telah dinilai. Kemudian, jumlah *potential dermal exposure* (*tPDE*) dan *actual dermal exposure* (*tADE*) di kalangan penyembur racun perosak telah ditentukan dengan menggunakan model *DREAM*, dan faktor-faktor yang mempengaruhi kadar pendedahan telah dianalisis melalui analisis regresi logistik. Dalam sub-kajian kedua, kepekatan Uranin sebagai pengganti racun perosak dan jumlah potensi pendedahan kepada kulit telah ditentukan melalui kaedah *WBD*. Kemudian, korelasi di antara kadar pendedahan yang diperolehi dengan pendedahan kulit dari *DREAM* (Sub-kajian 1) ditentukan dengan menganggar pekali korelasi *Spearman*. Melalui sub-kajian ketiga, gejala masalah kulit dan keracunan racun perosak yang dialami oleh responden selepas menyembur racun perosak telah dikenalpasti. Kemudian, hubungkait antara kadar pendedahan yang diperoleh melalui model *DREAM* dan gejala kesihatan yang dialami telah ditentukan memalui analisis regresi logistik. Berdasarkan *DREAM*, anggaran *tPDE* di kalangan penyembur racun perosak di setiap subsektor pertanian adalah berbeza secara signifikan (χ^2 = 118.093, p < 0.001). Pendedahan tertinggi di catatkan di kalangan penyembur sawah padi (151.39 ± 22.64 DU) dan pendedahan terendah adalah di kalangan responden di ladang kelapa sawit (47.67 ± 18.47 DU). Faktor-faktor yang menyumbang kepada nilai *tPDE* yang tinggi (≥100 Unit Dermal) adalah pemendapan (*deposition*) racun perosak, umur pengendali, penggunaan sarung tangan dan penggantian segera pakaian yang digunakan selepas penyemburan. Jumlah kepekatan Uranin yang terdapat pada bahagian-bahagian *Tyvek* pada responden di sawah adalah 0.281 mg/kg, diikuti oleh responden di ladang kelapa sawit (0.043 mg/kg), sayur (0.042 mg/kg) dan koko (0.013 mg/kg). Jumlah potensi pendedahan kulit (*tPDE*) adalah paling tinggi di kalangan responden di sawah dengan jumlah purata *PDE* 1.56E-03±1.73E-04 ($p < 0.001$). Hubungkait positif yang signifikan telah dikenalpasti di antara log *tPDE* yang diperolehi melalui model *DREAM* dengan log tPDE yang diukur melaui *WBD* (r_s =0.752, p=0.001). Responden yang bekerja di sawah padi mengalami gelaja kegatalan kulit (53.3%) dan ruam gatal (24.4%), sementara responden di kebun sayur mengalami gejala peningkatan penghasilan air liur (29.4%), cirit-birit (14.7%), kerap kencing (23.5%), kekejangan (26.5%), lemah otot (29.4%) dan kegelisahan (17.6%). Jumlah *PDE* dari *DREAM* adalah pemboleh ubah yang mempengaruhi model regresi logistik yang meramalkan gejala masalah kulit dan keracunan. Kajian kami menyokong bahawa *DREAM* dapat menganggar tahap pendedahan, taburan dan laluan pendedahan racun perosak, dan boleh dihubungkaitkan dengan gejala kesihatan. Walaubagaimanapun, anggaran *DREAM* terhadap pendedahan pada bahagian badan berbanding data pengukuran perlu disahkan lebih lanjut untuk tujuan penambakbaikan model. (494 patah perkataan)

Kata kunci: Pendedahan kulit; racun perosak; model *DREAM*; *whole body dosimetry*; gejala kesihatan

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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Declaration by Members of Supervisory Committee

This is to confirm that:

- the research conducted and the writing of this thesis was under our supervision;
- supervision responsibilities as stated in the Universiti Putra Malaysia (Graduate Studies) Rules 2003 (Revision 2012-2013) are adhered to.

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LIST OF ABBREVIATIONS

AChE	Acetylcholinesterase
ADE	Actual Dermal Exposure
AOR	Adjusted Odd Ratio
BERNAS	Padiberas Nasional Berhad
BMI	Body Mass Index
BPs	Body parts
BS_{BP}	Body surface factor
C	Concentration
CI	Confidence Interval
DDT	Dichlorodiphenyltrichloroethane
D_{BP}	Deposition on body parts
DOSH	Department of Occupational Safety and Health
DREAM	Dermal Exposure Assessment Method
DU	Dermal Unit
E_{BP}	Emission on body parts
E_{I}	Intrinsic emission (physical and chemical characteristics of liquid pesticide)
ER_{E}, ER_{D}, ER_{T}	Exposure route factors for emission, deposition and transfer
GDP	Gross Domestic Product
Ha	Hectare
$I_{D.BP}$	Amount of pesticide on clothing and uncovered skin from deposition
$I_{E.BP}$	Amount of pesticide on clothing and uncovered skin from emission
$I_{T.BP}$	Contamination level of contact surface

LD_{50}	Lethal dose, 50
MARDI	Malaysian Agricultural Research and Development Institute
MPOA	Malaysian Palm Oil Association
MPOB	Malaysian Palm Oil Board
MPOC	Malaysian Palm Oil Council
MW	Molecular weight
MYR	Malaysian Ringgit
NaOH	Sodium Hydroxide
OC	Organochlorine
O_{BP}	Clothing protection factor on body parts
OMAFRFA	Ontario Ministry of Agriculture, Food and Rural Affairs
OP	Organophosphorus
$P_{D.BP}$	Frequency of deposition on clothing and uncovered skin
PDE	Potential Dermal Exposure
$P_{E.BP}$	Frequency of emission on clothing and uncovered skin
PPE	Personal protective equipment
PS	Physical state
$P_{T.BP}$	Frequency of pesticide contact with surfaces or tools
RF	Relative Fluorescence
SD	Standard Deviation
T_{BP}	Transfer on body parts
tADE	Total Actual Dermal Exposure estimates
tPDE	Total Potential Dermal Exposure estimates
USEPA	United States Environmental Protection Agency
V	Viscosity

WBD	Whole Body Dosimetry
WHO	World Health Organization

CHAPTER 1

INTRODUCTION

1.1 Background of study

The Malaysian agricultural sector can be categorized into three sub-sectors, namely (i) agro-industrial subsector which serves the export market; comprise of oil palm, rubber, cocoa, and timber industries, (ii) food subsector for domestic consumption including rice, fruits and vegetables, livestock and fisheries and (iii) miscellaneous group serve domestic and export market; including pepper, coconuts, sweet potato, cassava and tea (Rahman, 2012). Agriculture sector played an important role in national economic development, contributing 8.9% to the national Gross Domestic Product (GDP) in 2015 (Department of Statistics Malaysia, 2016). The major contributor to the GDP of the agriculture sector was oil palm with 46.9%, followed by other agriculture with 17.7%. Moreover, paddy production in 2015 has increased compared to the previous year, by 473.0 thousand tonnes (16.6%), where the paddy-growing area was increased from 2014 by 7.0% from 679.2 thousand hectares to 730.0 thousand hectares in 2015.

Extensive use of agrochemicals is one of the issues in the agricultural economy where the Malaysian farming sector is still relying on pesticides to sustain the needed yields and ensure adequate profit levels of producers (Jamal & Yaghoob, 2014). Pesticides can be defined as the substance used to prevent, destroy, repel or mitigate any pest which is harmful living things ranging from insects, animals, weeds to microorganisms (USEPA). It can be classified according to target organisms into herbicide, fungicide, insecticide, nematicide, rodenticide and bactericide, or based on the chemical structure of the compound, or type of health hazard involved. The principle of pesticides usage is that it should finally decompose rapidly into harmless compounds after reaching the target organism and having its intended effect (Snelder et al., 2008). However, their inappropriate use has affected the health of the workers who handle such toxic substances.

According to the WHO, about 300,000 people were killed of pesticide poisoning annually in Asian rural areas from 3 million cases occurs worldwide (WHO, 2009). There are three routes of pesticides exposure which are inhalation, oral ingestion and dermal uptake. However, the most common exposure routes are inhalation and dermal (Gao et al., 2014). Incidents due to skin absorption accounted for >90% of pesticides poisoning cases (An et al., 2014). According to a conceptual model of dermal exposure developed by Schneider et al. (1999), transport of contaminant mass from its sources to the skin surface occurred through three main routes, which are emission, deposition and transfer.

Occupational pesticide exposure can adversely affect workers' health and their capacity to perform work, which results in significant economic losses, decreased productivity, medical expenses, and loss of work. The previous studies associated pesticide exposure with diseases including cancers, leukemia and asthma (Kim et al., 2017). However, the risk of health hazards from pesticide exposure depends not only on the toxicity of the ingredients but also the exposure level. Moreover, pesticide exposure assessment is a complicated task as it depends on several factors.

1.2 Problem statement

Malaysian agriculture consumes more than 200000 tons of pesticides annually, comprised more than 50000 tons of active ingredients (Mazlan et al., 2016; Kamaruzaman et al., 2020). The import value was estimated to be US$202 572.48. According to the Malaysian Department of Agriculture, pesticide use is regulated in Malaysia, and the most common pesticides used in agriculture are organophosphates, pyrethroids, glyphosate, and 2,4- D- dimethylammonium (Kamaruzaman et al., 2020). The occupational disease and poisoning cases in the agriculture sector were associated mainly with the use of pesticides, which are widely used to control pests and pest-induced diseases. Farmers are exposed to pesticides while mixing, loading and spraying activities, or while cleaning the tools and equipment, and disposing of the pesticide containers (Baldi et al., 2006; Kapka-Skrzypczak et a., 2011; Lesmes-Fabian et al., 2012; Remoundou et al., 2015).

Meanwhile, a total of 6020 occupational disease and poisoning cases have been reported to the Department of Occupational Safety and Health (DOSH) in the year 2017, with occupational skin disease (69 cases) and occupational poisoning (44 cases) were among reported cases. The agricultural sector recorded 108 cases from a total of occupational disease and poisoning cases received (DOSH, 2017). SOCSO has paid compensation to 2435 employees in the agriculture, forestry and fishing sectors in 2018, involving both temporary and permanent disability (Social Security Organization, 2018).

Dermal is the most common route of pesticide exposure suggested of about 60% of total exposure, and unintended exposure through dermal contact can be extremely hazardous to humans (Acquavella et al., 2004; Ye et al., 2013; Lesmes-Fabian et al., 2013). Farmers are potentially exposed to pesticides through emission (i.e. spills, splashes and immersion of hands or other body parts onto pesticide in the tank), deposition (i.e. direct spray contact and drift during spraying) and transfer (i.e. contact with surfaces such as the floor, worktables, working tools and equipment that have been contaminated with pesticide, or contact with plant that recently been sprayed) (Van-Wendel-De-Joode et al., 2003; Blanco et al. 2008; Lesmes-Fabian et al., 2012; Galea et al., 2015).

Several studies have reported the occurrence of acute health symptoms as an indicator of pesticide poisoning associated with pesticide exposure among agriculture workers, ranging from mild to fatal symptoms including headaches, nausea, vomiting, dermatitis, burns, abdominal pain, insomnia, dizziness, shortness of breath, excessive salivation, blurred vision, mood changes, diarrhea, convulsions, lachrymation and fatigue (Kapka-Skrzypczak et al., 2011; Garcia et al., 2012; Muñoz-quezada et al., 2017). Besides, chronic effects include various diseases such as cancer, leukemia, hormone disruption, hypersensitivity as well as respiratory diseases such as asthma and allergies (Kim et al., 2017). A study by Mazlan et al. (2016) determine the presence of imazapic and estimate the potential dermal exposure among farmers in the paddy area of Tanjung Karang, Selangor Malaysia. This is due to the herbicide that persists in the environment for up to 39 days and may cause several health effects to the farmers.

Previous studies had reported that pesticide exposure was strongly influenced by factors such as crop type, exposure route, types of spraying equipment, usage of Personal Protective Equipment (PPE) and hygiene practices (Hughes et al., 2008; Nuyttens et al., 2009; Cao et al., 2015). For instance, pesticide drift may cause exposure among farmers, since a previous study showed that drift could be measured hundreds of yards to hundreds of feet away from the application site. In order to understand the dermal contamination to farmers from pesticide drift mechanism, a study by How et al. (2015) among farmers in Tanjung Karang, Selangor Malaysia has evaluated the distribution of dermal contamination when the pesticide was sprayed under different flow rates (L/min).

After several fatalities from pesticide exposure, dermal exposure assessment has received more attention. However, methods for evaluating dermal exposure are not reasonably well standardized, where there is a lack of consensus or general agreement regarding a wide range of methods with different underlying assumptions (Schneider et al., 2000; Geer et al., 2004; Lesmes-Fabian et al., 2013). One of the approaches to assess dermal exposure is model estimations, where pesticide exposures are measured or modeled by monitoring and questionnaire-based measurements. Nevertheless, tools available such as EASE, EUROPOEM, PHED, RISKOFDERM, COSHH, STOFENMANAGER, DREAM and the approaches proposed by the USEPA often developed based on industrial sectors in Europe and the USA. Besides, some models are unable to differentiate the pesticide transport processes, unvalidated, and have been criticized on the reliability and reproducibility of the outcomes. Despite the limitations, the application of model estimation becomes important for exposure reduction purposes due to easy-to-use, cost-effective and less time-consuming. In many developing countries like those in Southeast Asia, there are no national systems to monitor pesticide risk on a routine basis (Schreinemachers et al., 2015). Therefore, established exposure databases and models from developed countries and international organizations are often used as substitutes in the evaluation of pesticides (Atabila et al. 2017; Jansen 2017). According to the manual of recommended practice on the assessment of the health risks arising from the use of chemicals hazardous to health at the workplace provided by DOSH Malaysia, the degree of dermal exposure can be estimated by considering two factors, which are the hazardous properties and exposure rating which includes extent and duration of dermal contact. The level of risk can be determined using a risk matrix and categorized into three categories (i.e. Low risk, moderate risk and high risk).

The previous study has found that Dermal Exposure Assessment Method (DREAM) assesses dermal exposure most appropriately in farming systems in Colombia, hence applying this method in agriculture sub-sectors in Malaysia was intended to avoid generalization of exposure level due to dermal exposure that depends on several factors (Lesmes-Fabian et al., 2013; Hughes et al., 2006). For instance, dermal exposure level to pesticides depends greatly on crop height and density (Machera et al., 2002; Hughes et al., 2008; An et al., 2014; Cao et al., 2015). Higher plant density generally results in greater pesticide exposure levels due to increasing direct contact frequency of applicators with the plants. Besides, applicators who sprayed on higher crops might have well-distributed exposure on the body due to the movements of spraying upwards and downwards to cover the plant (Hughes et al., 2008; Zhao et al., 2016).

On the other hand, quantitative dermal exposure measurement such as patch method and whole body sampling involves the quantification of pesticide concentration at the immediate contact interface between workers and pesticides (Ye et al., 2013). USEPA recommends the application of whole-body dosimetry technique for assessing dermal exposure to the pesticide, regardless of the limitations of this method such as expensive to be carried out and the unestablished standard garments that should be used (Fenske et al., 2005).

Owing to this fact, the application of DREAM and the correlation of its estimates with the measurement techniques (i.e. whole-body dosimetry) in determining potential and actual dermal exposure to pesticide among sprayers in Malaysian agriculture subsectors remain a question due to the varieties in the crop high, difference agriculture setting and environmental conditions that may influence the ability of the model to predict the risk. In addition, how this model can be associated with major health symptoms perceived by pesticide handlers is also interesting to be studied as it can be applied to establish causal relationships between the occurrence of a health outcome and the pesticide exposure. This study was conducted through three substudies; sub-study 1) dermal exposure assessment using DREAM; sub-study 2) dermal exposure assessment using whole-body dosimetry method; sub-study 3) identification of health symptoms. The subsectors involved were paddy (*Oryza sativa*), cocoa (*Theobroma cacao)*, vegetables and oil palm (*Elaeis guineensis*).

1.3 Research questions

This research intended to answer three main research questions;

1. How does DREAM determine the potential and actual dermal exposure to pesticide among sprayers in Malaysian agriculture subsectors (i.e. paddy, vegetable, cocoa and oil palm)?
2. What is the correlation between dermal exposure measured by DREAM and whole-body dosimetry method?
3. What are the major health symptoms occurred among pesticide handlers?
4. Is there any association between perceived health symptoms among respondents with the studied variables in DREAM?

This study is classified into three sub-studies; Sub-study 1) Dermal exposure assessment using Dermal Exposure Assessment Method (DREAM), Sub-study 2) Dermal exposure assessment using whole-body dosimetry method, Sub-study 3) Identification of health symptoms.

1.4 Research objectives and hypothesis

i. General objective

The general objective of this study is to assess dermal exposure and health symptoms among pesticide handlers in selected agriculture sectors in Malaysia. In order to achieve this objective, the experiments were divided into 3 sub-studies with specific objectives as follows:

Specific objective	Hypothesis
Sub-study 1: Dermal Exposure Assessment using Dermal Exposure Assessment Method (DREAM)	
1. To determine socio-demographic characteristics of the pesticide handlers in agriculture subsectors (i.e. paddy, vegetable, cocoa and oil palm)	-
2. To compare pesticide application (i.e. types of pesticides, spraying equipment and usage practices), personal protective equipment used and hygiene practices among pesticide handlers	-
3. To compare pesticide dermal exposure, frequency, amount and distribution of exposure through emission, deposition and transfer on different body parts using Dermal Exposure Assessment Method (DREAM)	The pesticide dermal exposure, frequency, amount and distribution of exposure indicate higher value through emission on lower body parts among respondents in all agriculture subsectors
4. To compare the total potential and actual dermal exposure between pesticide handlers in agriculture subsectors (i.e. paddy, vegetable, cocoa and oil palm) using DREAM	The total potential dermal exposure among respondents in each agriculture subsector is higher than total actual dermal exposure
5. To analyze factors affecting dermal exposure levels among pesticide handlers in the study areas.	The dermal exposure levels among pesticide handlers are influenced by socio-demographic, pesticide exposure information, personal protective equipment and hygiene practices
Sub-study 2: Correlation between DREAM and whole body dosimetry Method in Assessing Dermal Exposure	
1. To compare the level of uranine (mg/kg) and total potential dermal exposure on body parts among sprayers in different agriculture subsectors (i.e. paddy, vegetable, cocoa and oil palm) using whole-body dosimetry method	The level of uranine (mg/kg) and total potential dermal exposure were higher on lower body parts among sprayers in vegetable and paddy fields
2. To model the correlations between total potential dermal	Total potential dermal exposure from DREAM (Sub-study 1) shows

exposure from DREAM (Sub-study 1) with measurement data using whole-body dosimetry method	good positive correlations with measurement data using whole-body dosimetry method
Sub-study 3: Identification of Health Symptoms	
1. To compare the skin problem symptoms perceived by respondents after pesticide spraying activity	Respondents in each agriculture subsectors perceive different skin problem symptoms after pesticide spraying activity
2. To compare the pesticide poisoning symptoms perceived by respondents after pesticide spraying activity	Respondents in each agriculture subsectors perceive different poisoning symptoms after pesticide spraying activity
3. To determine the association between the studied variables in DREAM with perceived health symptoms among sprayers.	The occurrence probability of perceived health symptoms among sprayers can be associated with the variables in DREAM.

1.5 Study justification

Dermal exposure assessment is important to provide an insight into the dynamic interaction between pesticides and human skin (Fenske, 2000). Lesmes-Fabian et al. (2013) found that DREAM is the most appropriate model to be applied in developing countries because of the type of determinants included in the model. However, studies on the application of this method in dermal exposure to pesticides are still limited. Meanwhile, patch sampling and whole body sampling are commonly applied methods for quantifying dermal exposure (Fenske, 1993; Soutar et al., 2000). The whole-body sampling technique involves measurement of the amount of contaminant landing on clothing and skin or penetrating clothing layers. It has an advantage over the patch sampling method since it does not rely on uniform distribution of the contaminant over large sections of the body. Lightweight coveralls are commonly used to estimate exposure to body areas covered, while exposure to hands and feet are measured by using gloves and socks respectively. Exposure to different body parts will be determined by sectioning of the suit, and analysis of the relevant parts. This is important to prevent underestimation or over-estimation from extrapolation of proportion from a particular region. Usage of coverall to measure potential dermal exposure (PDE) will help to explain the process of contamination better (Soutar et al., 2000).

PDE is the amount of pesticide landing on the outer layer of work clothing which was measured during pesticide application activities. In this study, Tyvek garments were used over the work clothing together with the cotton gloves. The selection of tracer uranine as pesticide surrogate is due to its low detection level, rapid quantification, solubility in spray mixtures, minimum physical effect on droplet evaporation, distinctive property differentiating it from background or naturally occurring substances, stability, moderate cost, nontoxicity and acceptability under Food and Drug Regulations (Lesmes-Fabian et al., 2012).

1.6 Novelty

Pesticide use in developing countries like those in Southeast Asia was less strictly regulated, and national systems to monitor pesticide risk on a regular basis are unavailable (Schreinemachers et al. 2015). Thus, pesticide use in developing countries may pose greater potential health effects than in developed countries. Besides, exposure assessment methods available were inadequate to generate robust exposure estimates. Therefore, more systematic studies are necessary to address gaps in knowledge and improve exposure estimates for health and risk assessment (Pan and Siriwong, 2010).

Available tools to describe pesticide exposure include direct measurements, biological monitoring and predictive models, which combination of measured exposure data and modelling approaches may provide the most appropriate assessment. Direct measurement and biological monitoring were the most commonly used methods in developing countries to measure dermal exposure to pesticides (Wong & Brown, 2020). Limited studies used interviews and questionnaires as part of exposure assessment to collect information regarding agricultural practices and personal protective measures. Hence, there can be a significant gap in knowledge related to exposure measurements, where established exposure databases and models from developed countries and international organisations are often used as substitutes in the assessment of pesticide exposure (Atabila et al. 2017).

Intended to fill the gaps of limited exposure measurements, our research has applied the DREAM model and direct measurement (i.e. whole-body dosimetry) to determine the range of dermal exposure levels, mainly explaining variability in exposure levels between groups having different working conditions. This research has evaluated the exposure information to provide a better understanding regarding the distribution of pesticides through dermal exposure routes (i.e. emission, deposition and transfer) across the body parts through DREAM. Wong & Brown (2020) has reviewed the pesticide exposure assessment methods that have been applied among rice farmers in developing countries. Based on their study, only three of 22 studies used predictive exposure models to estimate daily pesticide exposure in the rice fields, while other studies used direct exposure measurement methods (e.g. whole body dosimetry) and biological monitoring. Predictive models used include Pesticide Handlers' Exposure Database (PHED) and the Agricultural Handlers' Exposure Database (AHED) (Gammon et al., 2011), Dermal Exposure Assessment Method (DREAM) (Baharuddin et al., 2011), and World Health Organisation predicted exposure assessment model (WHO-PEAM) (Phung et al., 2019).

The outcome of this study ware the model for predicting high potential dermal exposure to pesticide among pesticide handlers in the study areas, which explains the interactions between studied variables and pesticide exposure. This can complement the limited field measurements in a cost-effective way, due to the method that requires mathematical skills and is easy to be carried out in the field (Fabian et al., 2013). Besides, this study provides the correlation between dermal exposure from DREAM and measurement data from whole-body dosimetry method. This study also evaluates the association between the studied variables in DREAM with perceived health symptoms among pesticide handlers. For instance, the outcomes can be integrated to recommend proper and safe pesticide handling practices in order to ensure the safety and health of smallholders and agricultural workers. This is important as a review by Lam et al. (2017) confirmed that the limitations in exposure assessment in Southeast Asia result in significant difficulties in establishing causal relationships between the occurrence of a health outcome and the pesticide exposure.

1.7 Significance and contribution of study

The main objective of this study is to assess the DREAM in determining dermal exposure in Malaysian agriculture subsectors. Under the Occupational Safety and Health Act 1994, employers in the farming sector have an obligation to perform risk management through identifying workplace hazards, assessing the associated risks, and minimize those risks. Besides, in compliance with the Pesticide Management Guidelines of Malaysian Good Agricultural Practice (MyGAP), one of the elements of good agricultural practices includes protecting the safety and health of the workers. The information provided in the first sub-study including the determinants of dermal exposure (e.g. intensity, duration and exposed surface areas) and variability of exposure routes and distribution, is important for proper assignment of dermal exposure risks to the pesticide. The information on types of pesticide used by farmers is useful for regulatory authorities in monitoring the use of prohibited pesticides with immediate phasing out the hazardous pesticides through national policies and enforcing more restricted access to these chemicals. Besides, the model predicting high dermal exposure developed in this study may be useful for agriculture agencies and employers in pesticide risk assessment in the process of developing farm safety management program, integrating four risks management steps (i.e. hazard identification, risk assessment, development of safety measures and review changes).

Sub-study 2 provides information on the correlation between the dermal exposure by semi-quantitative DREAM as an option of easy-to-use, cost-effective and less time-consuming method, with quantitative whole-body dosimetry method. The data may be useful for other research studies in further exploring the validity and application of combined direct measurement methods and predictive models. In line with Sustainable Development Agenda Goal 3 (Target 3.9) on substantially reducing the number of illnesses and deaths from hazardous chemicals**,** sub-study 3 provides identification of perceived health symptoms and association with the dermal exposure variables. The data from this study may be useful for employers and agriculture agency in monitoring purposes, as well as designing an induction training program for people working on the farm, which integrate information on pesticide handling, exposure risk factors and potential health effects to enable workers to perform work without risk of injury and illness.

1.8 Conceptual framework

Figure 1.1 illustrates the conceptual framework of this study. Pesticides are commonly used in farming sectors to kill another form of life in order to maintain higher yields. However, overdependence on pesticides may pose negative effects on human health as well as the environment including water, air, soil and other non-target organisms (Baharuddin et al., 2011; Delhomme et al., 2011; Kilinc et al., 2013; Okoya et al., 2013). Pesticide handlers were selected as they regularly engaged in pesticide mixing, loading and application at different growing stages of a particular crop as well as cleaning the equipment (Baldi et al., 2006; Lesmes-Fabian et al., 2012; Remoundou et al., 2015).

Previous studies had reported that pesticide exposure was strongly dependent on several factors; such as crop type, exposure route, types of spraying equipment, usage of Personal Protective Equipment (PPE) and hygiene practices (Hughes et al., 2008; Nuyttens et al., 2009; Cao et al., 2015). Among all pesticide exposure routes available, dermal exposure was selected as it has been reported as the most significant way that farmers become exposed during pesticide handling. Pesticide exposure has been associated with impacts on human health. For example, some previous studies mentioned by Atreya et al. (2012) reported acute health symptoms through individual surveys as an indicator of pesticide poisoning without blood analysis.

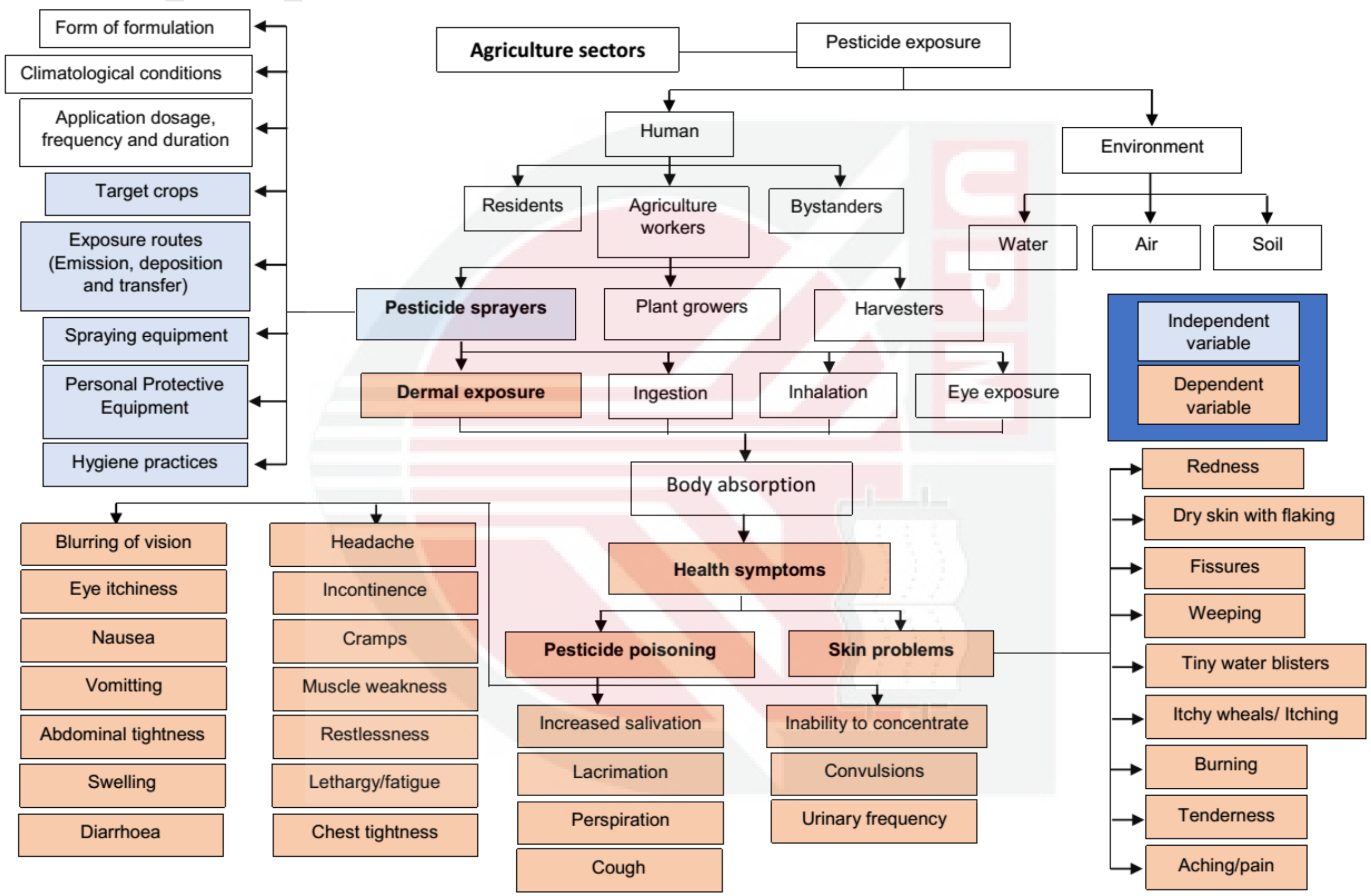

Figure 1.1: Conceptual framework of the study

1.9 Thesis outline

This thesis consists of six chapters purposely to provide the data on the dermal exposure assessment and health symptoms among pesticide handlers in selected agriculture sectors in Malaysia. Each chapter has been organized as follows:

Chapter 1 delivers a brief introduction on the study background, problem statement, research questions, objectives, research hypotheses, novelty and significance of the study. In Chapter 2, a comprehensive literature review related to this study is provided, particularly on pesticide usage in Malaysian agricultural sectors, its regulations and effects of pesticides on human health. Besides, pesticide exposure routes comprised of emission, deposition and transfer, and factors that influenced dermal exposure are detailed out in this chapter. Application of Dermal Exposure Assessment Method (DREAM), whole-body dosimetry (WBD) and fluorescence tracer technique in current research on pesticide exposure assessment is also provided.

Chapter 3 highlights the result of sub-study 1; the dermal exposure assessment to pesticides among sprayers in different agriculture sectors using DREAM. The chapter starts with an introduction section consisting of research questions and the objectives of the sub-study. The description, sample size calculation and inclusion criteria can be found in the methodology section. Besides, this chapter also explains the sample selection and ethical consideration, followed by the description of data collection. The results section begins with a determination of socio-demographic characteristics of the respondents, the pesticide application, personal protective equipment used and hygiene practices among respondents. The subsequent section presents the frequency and amount of dermal exposure and the distribution on different body parts. Besides, the total potential and actual dermal exposure among pesticide handlers are also presented. Then, the relationships between independent variables and dermal exposure levels are included in this chapter. The discussion section presents the explanation and the summary of the results.

Chapter 4 highlights the result of sub-study 2; the dermal exposure assessment among sprayers in different agriculture sectors using whole-body dosimetry method (WBD) with the tracer uranine as pesticide surrogate. The chapter begins with a brief introduction to the exposure assessment, followed by research questions and specific objectives. The methodology section is comprised of the description of the study area and sample size calculation. The sampling section provides information on the field characteristics, applications and environmental conditions of the study area. Then, the chapter details the dermal exposure assessment method including the sampling procedure, quality control, calibration procedure, analytical method and calculations and data interpretation. The results section begins with the determination of the amount of uranine on body parts among respondents followed by the total potential dermal exposure. This chapter also presents the correlations coefficient of the DREAM estimates and measured dermal exposure through WBD. The findings are discussed and concluded in the last section.

Chapter 5 provides the section for identification of health symptoms, sub-study 3, where it begins with a brief introduction on the health symptoms of pesticides to human health, followed by research questions and specific objectives. The method used to determine the common symptoms of skin problems and pesticide poisoning experienced by respondents after pesticide spraying is detailed out in the methodology section. Finally, the results are discussed and concluded. Chapter 6 provides the conclusions from each sub-study. Besides, this chapter also addresses the limitations of this study with several recommendations which might be beneficial for the pesticide management activities and appropriate dermal exposure assessment.

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