



**EFFECTS OF PESTICIDES ON POPULATION
DYNAMICS OF NON-TARGET ORGANISMS IN
AGRICULTURAL SETTINGS: AN OBJECT-ORIENTED
APPROACH**

BY

MR. MWEMA FELIX

**A DISSERTATION SUBMITTED IN PARTIAL FULFILLMENT OF
THE REQUIREMENTS FOR THE DEGREE OF DOCTOR OF
PHILOSOPHY (ENGINEERING AND TECHNOLOGY)
SIRINDHORN INTERNATIONAL INSTITUTE OF TECHNOLOGY
THAMMASAT UNIVERSITY
ACADEMIC YEAR 2019
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DISSERTATION

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ENTITLED

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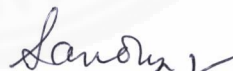
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ABSTRACT

Pesticides are chemicals that are used for pest control. They are grouped according to the type of target pests as herbicides, insecticides, fungicides, algicides, nematicides, molluscicides, etc. It is estimated that herbicides, insecticides, fungicides, and other groups of pesticides accounts for 50%, 30%, 17%, and 3% of the global pesticide use. The use of herbicides for weed control have increased the amount of food produced worldwide but also have been widely associated with the population decline of non-target organisms such as amphibians.

Using an online-based survey and an object-oriented algorithm (PestTox), this study (i) surveyed how aware people in Tanzania, are of pesticides and their associated health effects, (ii) simulated fate and transport of glyphosate and its effects on population dynamics of frogs (*Crinia insignifera*), and (iii) simulated the effect of spray drifts on population dynamics of frogs. Two scenarios were simulated using the width of no spray zone (buffer zone) of thirty meters and zero meters around the agricultural field for year 2012 and 2013.

Results of the survey show that one hundred fifty-seven people responded to the online survey concerning the pesticide awareness. Results reveal that respondents

were aware that pesticides could potentially kill them and damage the environment, yet their pesticide management practices were poor. Results suggest that poor pesticide management practices could be due to negligence. Therefore, there is a need to increase tailor-made motivational and awareness programs on best pesticide management practices.

Results of the simulation modeling of fate and transport of glyphosate as well as modeling of spray drifts show that when using buffer zone width of thirty meters, spray drifts were very little compared to when using buffer zone width of zero meters. In one hundred forty-eight days after the first application, all glyphosate deposited on the crop/leaf surfaces were removed completely, suggesting that glyphosate does not stay longer on the crop/leaf surfaces. If there was no washoff events that transferred glyphosate from the crop/leaf surfaces to the topsoil, all glyphosate in the topsoil could have been removed completely by the competing processes by day forty of the simulation, implying that glyphosate does not stay longer in the topsoil.

When using buffer zone width of thirty meters, glyphosate runoffs were little compared to when using buffer zone width of zero meters. This suggests that buffer zone helps to reduce the runoff of glyphosate. When using buffer zone width of thirty meters, glyphosate runoffs did not have high impact on population dynamics of frogs for year 2012 and 2013 respectively.

When using buffer zone width of zero meters, the first two glyphosate runoff events for year 2013 that occurred earlier than the first glyphosate runoff event for year 2012 caused high mortality rates that killed all tadpoles within eighty-six days of the simulation, while for year 2012, all tadpoles died within one hundred five days after the first application due to high glyphosate contamination in the water body.

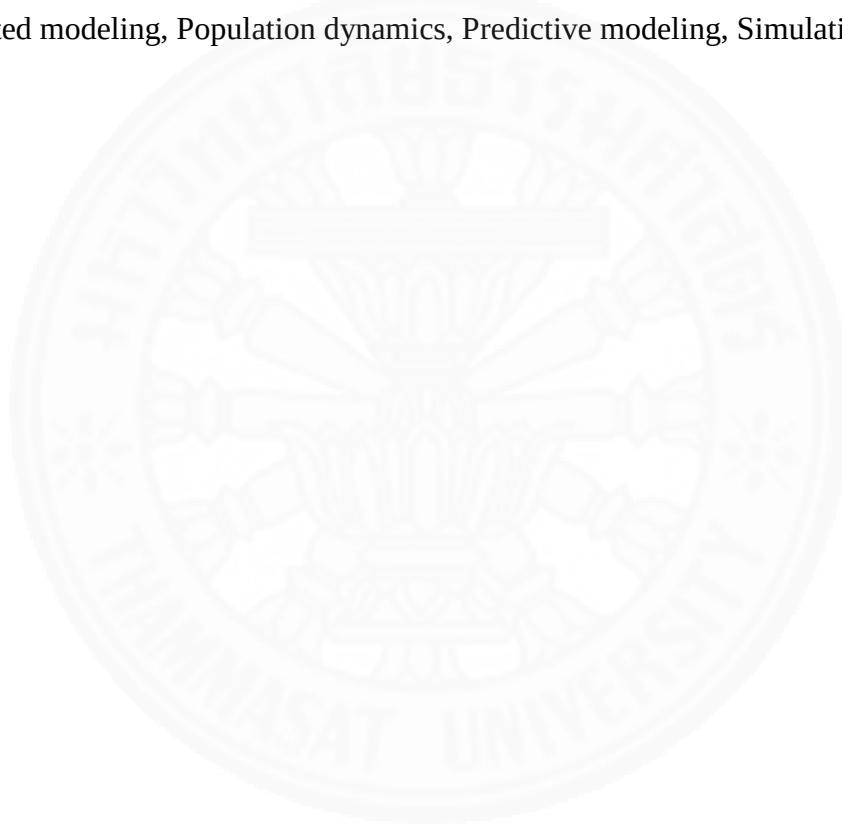
Also, when using buffer zone width of zero meters, results of the simulation modeling of spray drifts show that no eggs were laid and the number of tadpoles, froglets, and adult frogs were very small for both year 2012 and 2013. Results suggest that high glyphosate concentration and its high toxicity could be responsible for the declining population of frogs.

Results suggest that there is a need to switch to either less toxic formulations of herbicides or the use of integrated pest management tactics, or both so that

herbicides may be used only when necessary, specifically, when integrated pest management tactics provide inadequate pest control.

In conclusion, the algorithm presented in this study could be of practical significance to those involved in ecological and environmental modeling as well as within a management framework to evaluate potential effects of using agrochemicals in agricultural settings.

Keywords: Amphibians decline, Ecological modeling, Exposure assessment, Object-oriented modeling, Population dynamics, Predictive modeling, Simulation modeling



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Mr. Mwema Felix

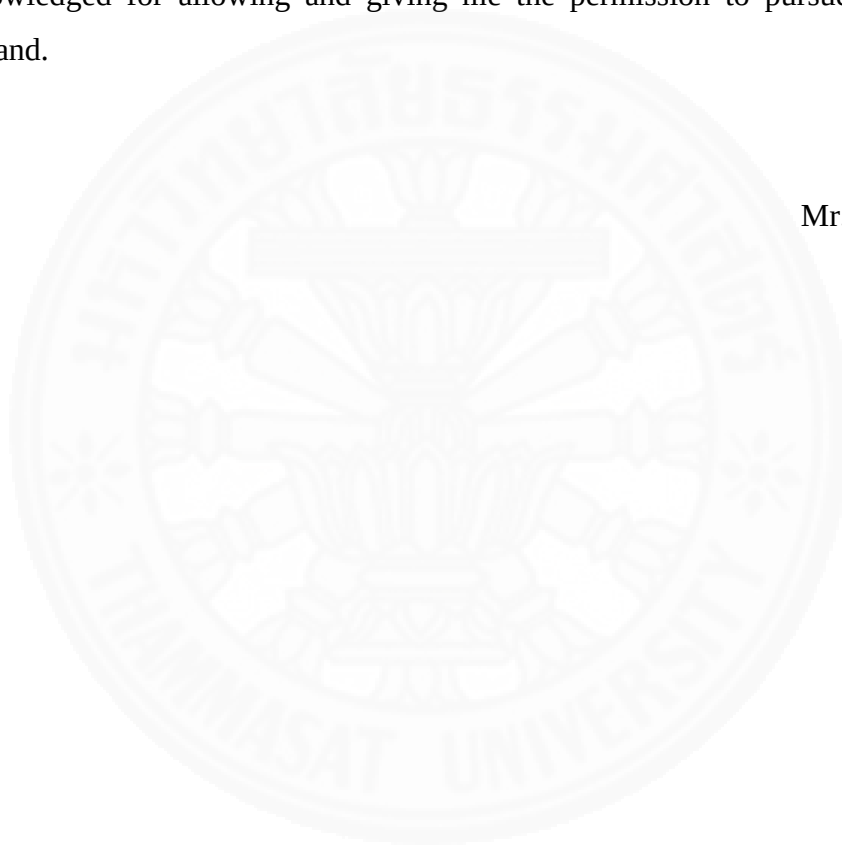


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CHAPTER 1

INTRODUCTION

1.1 Background information

Food is one of the basic needs for human development (Pyszczyński et al., 1997). Food, together with water, and a healthy environment are vital components to human well-being. With the current trend in the increase in global population (Lutz & Samir, 2010; Lutz et al., 2001), it means that more food is needed. To meet the demand, agriculture has to be expanded and developed (Tilman, 1999). Global agricultural area is expected to expand from the current area of 5.1 billion ha to 5.4 billion ha in year 2030 (Wirsén et al., 2010). In the past 40 years, agricultural expansion and development have increased the quantity of food produced and improved the quality of fresh food produced worldwide (De Geronimo et al., 2014).

This expansion has however led to the deforestation (Angelsen, 1999), and the increase in human health and environmental impacts (Ericksen et al., 2009; Tilman, 1999; Tilman et al., 2011) due to the increase in the use of pesticides and non-compliance to good agricultural practices and good pesticides application techniques (Ganpat et al., 2014). Despite the ongoing expansion, still, agriculture has been inefficient in many parts of the world, due to lack of modern agricultural tools and technologies (Laurance et al., 2014; Masters et al., 2013; Sayer & Cassman, 2013).

Agriculture has always been facing the challenges of pests and weeds. Pesticides have been designed to tackle this problem, and they are widely used for controlling pests for the purpose of increasing agricultural produce (Owusu-Boateng & Amuzu, 2013). Pesticides are defined as substances or mixture of substances which are used for repelling, preventing, and destroying pests and weeds (Arias-Estevez et al., 2008). The global pesticide consumption is estimated at 2 million tonnes a year, with 45%, 25%, and 30% of the global pesticides being used in Europe, USA, and the rest of the world combined respectively (Abhilash & Singh, 2009; De et al., 2014). It is estimated that herbicides, insecticides, fungicides, and other groups of pesticides accounts for 50%, 30%, 17%, and 3% of the global pesticide use (Abhilash & Singh, 2009). Of all pests, weeds pose great threats for many crops as potentially could

reduce crop yield by 34%. In year 2001-2003 about 10% of the global crop yield were lost due to weeds, and about 31% of global maize yield were lost due to pests (Oerke, 2006). Maize and other field crops are highly affected by weeds such as *Imperata cylindrical* (L.) (Chikoye et al., 2010) which are commonly found in West Africa (Chikoye et al., 1999) and Southeast Asia (Chikoye et al., 2010). It is with these reasons that farmers use pesticides to reduce loss of yields.

Pesticides contain active ingredients which makes them toxic chemicals thus their presence in the environment pose a threat to non-target living organisms (Mulla et al., 1981; Stephenson, 2001; Stephenson, 2003). Improper handling of pesticides, violation of both safety rules and application procedures (Matyjaszczyk, 2013) could lead to both direct and indirect contamination of pesticides (Mulla et al., 1981) which could potentially damage non-target crops (Al-Khatib et al., 1992; Ding et al., 2011; Hebert & Miller, 2004), non-crop species (Velini et al., 2008), as well as could cause undesired effects to humans (Jeyaratnama, 1990). Children, particularly those whose parents work and live in the vicinity of the agricultural fields (Schwartz et al., 2015) are mostly affected. Also, mishandling of pesticides could cause pesticide contamination in non-target sites through disposal of obsolete pesticides, disposal of empty containers, and either through cleaning or washing of spraying equipments.

Through agricultural runoff, potential contamination could be transported (Takahashi, 2003) to rivers, ponds, lakes and nearby source of waters which are used for human and animal consumption (Adjrah et al., 2013; Menn, 1980) thus causing health risk and problems, and environmental pollution (Son & Rutto, 2002). Chances are pesticides in the environment could be found in consumable food, soils, and groundwater (Abbassy et al., 1999; Kookana et al., 1995). Contamination in fresh waters could kill fish and other aquatic life while contamination in drinking water supplies could kill humans and animals.

Failure to exercise reasonable care i.e. not following both good agricultural practices and best pesticide management practices (Felix & Sharp, 2015; Felix & Sharp, 2016) are some of the reasons for the increase in exposure of pesticides in agriculture. Therefore, in this study, a survey was conducted to assess how aware people in Tanzania, are of pesticides and their associated health effects. This survey was necessary because pesticide management practices have an influence on impact

of pesticides (Neary et al., 1993) to both target pests and non-target organisms. In addition, best management practices of pesticides minimizes undesired problems associated with the use of pesticides (Clemente et al., 1993). Also, an object-oriented model (PestTox) was developed to simulate the fate and transport of pesticides in the environment and the effects of pesticides on population dynamics of non-target organisms in an agricultural setting using frog as case study. Frog was chosen because studies (Miller, 2006; Relyea, 2005b) have demonstrated that frogs could be highly affected by the use of pesticides including herbicides such as glyphosate-based formulations (e.g. Roundup[®]). The present study studied the population dynamics of frogs because although pesticides' mode of actions to target species are known, still, little is known (Gaupp-Berghausen et al., 2015; Kohler & Triebskorn, 2013; Zaller et al., 2014) regarding the undesired side effects on non-target species at the higher levels of biological interactions, particularly at population level (Kohler & Triebskorn, 2013). Simulation was also performed to establish and inform the importance of width of no spray zones (buffer zones) around the agricultural field to reducing exposure to spray drifts to non-target organisms, using object-oriented programming (OOP) (Acock & Reddy, 1997; Sequeira et al., 1997; Silvert, 1993).

The OOP was used because real world modeling can be easily studied, the code created can be reused on other applications, as well as it is reliable and flexible (Acock & Reddy, 1997). In addition, when implementing OOP, parameter values can be easily integrated and scaled, as well as the model can be easily understood and model components can be traced based on the way model objects are interconnected (Acock & Reddy, 1997; Mazzoni et al., 2015). Many models available to date do not allow users the ability to either expand the models e.g. adding new model components, or to change parameters, change of either regression equations or functions. Also in many cases, users have no choice but to contact the developers in case they would like to change data in the databases. These reasons inspired the development of this study.

1.2 Objectives of the study

The study had two main objectives; (i) to assess how aware people in Tanzania are on pesticides and their associated health effects, and (ii) to simulate the

effects of glyphosate on population dynamics of frogs in an agricultural settings using object-oriented programming. Specific objectives were:

- i) To assess how aware people in Tanzania, are of pesticides and their associated health effects,
- ii) To create a model on fate and transport of glyphosate in the environment, and
- iii) To assess the effects of fate and transport of glyphosate on population dynamics of frogs.

1.3 Scope of the study

The study was initially designed and intended for Tanzania as the study area. In the early stages of development it became evident that there was lack of majority of the data that were needed for simulation modeling. The lack of required data would have rendered the intended study. In this case, Thailand was chosen instead, as the study area due to the availability of the required data. Also, there came up the lack of specific toxicity data (48 h LC₅₀) of glyphosate-based formulations for frog species native to Thailand for simulating toxicity of glyphosate on population dynamics of frogs. Therefore, *Crinia insignifera* commonly known as Western Sign-bearing Froglet, a species native to Australia (Moore, 1954), was the selected non-target organisms due the availability of its toxicity data (48 h LC₅₀) for glyphosate (Roundup®) for simulating mortality rates of its four growth development stages. In addition, this species is found in both subtropical or tropical seasonal and permanent wetlands (Aubret, 2004), conditions assumed to be similar to both Thailand and Tanzania.

The study was divided into three main parts. The first part involved the use of questionnaire to assess the awareness of people in Tanzania on pesticides and their effects. Questionnaires were sent online to random people in Tanzania. Questionnaires had a mixture of closed and open-ended questions. Questions were mainly on pesticide practices (e.g. usage, use of protective gears), management (e.g. storage and disposal), and their associated health problems.

The second part involved simulation modeling of fate and transport of glyphosate in the environmental media namely air, crop/leaf, soil, and water. Two

scenarios were modeled using width of no spray zone (buffer zone) of thirty meters and zero meters around the agricultural field (Figure 1.1). Also, effects of glyphosate on population dynamics of frogs were studied.

In the air media, the study assessed the amount of glyphosate that were lost by wind as spray drifts from the agricultural field to the air. Due to the lack of data, the study assumed that spray drifts remained in the air.

In the crop/leaf media, the study assessed the amount of glyphosate deposited on the crop/leaf surfaces at the time of glyphosate application. Also, the study assessed the amount of glyphosate removed from the crop/leaf surfaces by washoff, photo-degradation, uptake, and volatilization processes.

In the soil media, the study assessed the amount of glyphosate reached the topsoil at the time of glyphosate application. Also, the study assessed the amount of glyphosate removed from the topsoil by runoff, biodegradation, volatilization, and macropore flow processes.

In the water media, the study assessed the amount of glyphosate reached the water body via runoff. Also, the study assessed the amount of glyphosate removed from the water body by biodegradation.

The third part involved modeling the effects of spray drifts (of glyphosate) on population dynamics of frogs. Also, two scenarios were modeled using width of no spray zone (buffer zone) of thirty meters and zero meters around the agricultural field. In these scenarios, the study assumed that spray drifts were deposited in the water body located next to the buffer zone.

The fate and transport modeling of glyphosate was conducted using object-oriented algorithm which were written in standard C++ language using Qt Creator (QtCreator, 2016). Computer simulation was implemented using open source software called Universal Simulator. Notepad++ was used as the Extensible Markup Language (XML) editor. The present model (PestTox) was adopted and modified from PestLCI (Birkved & Hauschild, 2006) and PestLCI 2.0 (Dijkman et al., 2012) which is a state of the art inventory model for pesticide emissions in life cycle assessment (LCA). Other data used in the simulation such as glyphosate properties and biochemistry, crop phenology, frog phenology were obtained from the literature (i.e. existing

ecological data). The study assumed that fate and transport distribution of glyphosate in the environment was as per Figure 1.2.

Maize was selected as the study crop because it is one of the main crops grown in Tanzania, Thailand, and many other parts of the world. Herbicide glyphosate was selected because it is widely used for weed control in Tanzania, Thailand, and many other parts of the world. Weather data i.e. daily rainfall, daily maximum temperature, and daily minimum temperature used in the simulation study are for both year 2012 and 2013, and were obtained from Thai Meteorological Department (TMD).

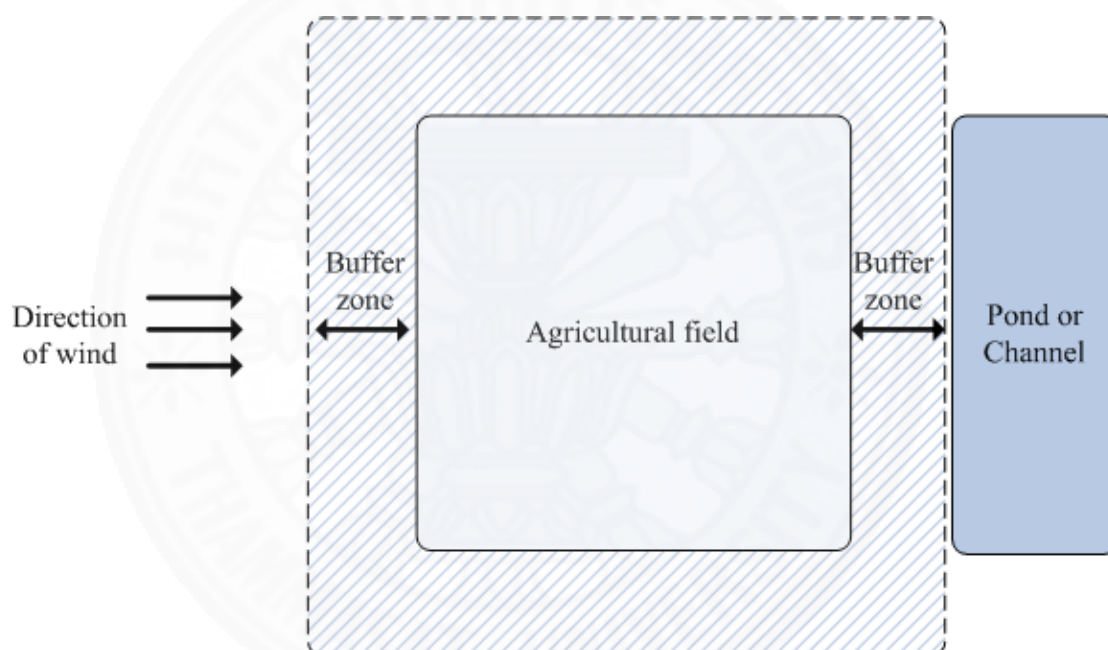


Figure 1.1 Agricultural field surrounded by the buffer zone.

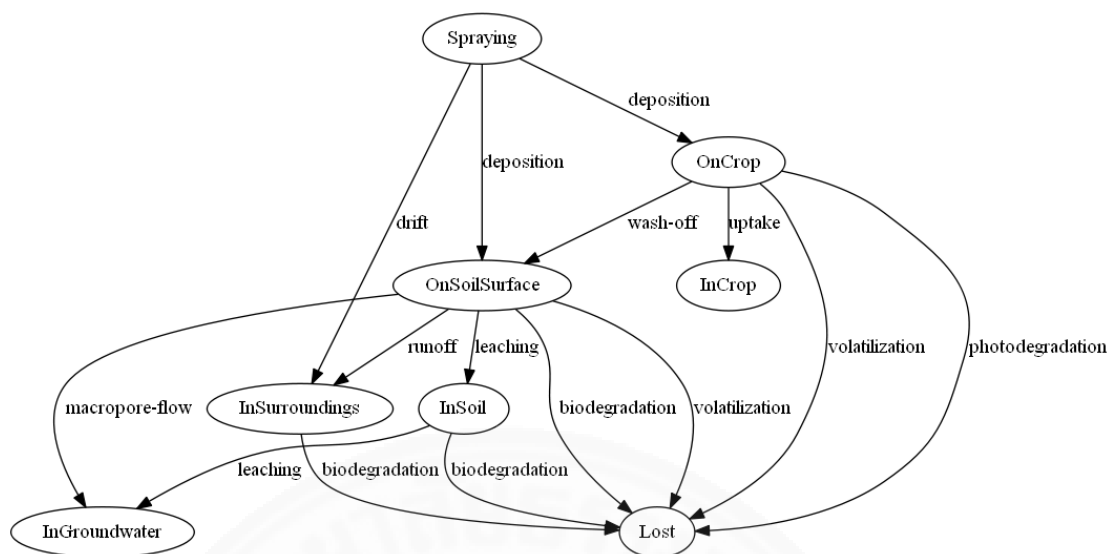


Figure 1.2 Fraction distributions after the application (spraying).

CHAPTER 2

REVIEW OF LITERATURE

2.1 What are pesticides

Agriculture has been facing a challenge of pests. These pests include insects, animals, and weeds that destroy plants, food, etc. These pests are largely attributed by the climate change (Chakraborty et al., 2000). To reduce the problem of pests, pesticides have been in use for the past 5 decades. They are chemical substances that are intended to either control or kill targeted pests. Pesticides are synthetic and natural chemicals that are used for plant protection against pest and vectors of animal and human diseases (Chen et al., 2012). For the first time synthetic pesticides became available and introduced into the market in year 1940s (van der Werf, 1996). They are grouped according to the type of pest such as herbicides, insecticides, fungicides, algicides, nematocides, molluscicides etc. They can also be categorized as organochlorines, organophosphates, carbamates, and pyrethrums (Chen et al., 2012; Lilliehöök, 1996).

According to the World health Organization (WHO), pesticides are classified based on their acute toxicities on rats (LD_{50}) i.e. lethal dose in 50% of the population; both oral and dermal (Table A.1). According to the form of pesticides, liquid pesticides are more dangerous than solid pesticides. According to the International Agency for Research on Cancer (IARC) carcinogenic pesticides are classified as in Table A.2. Stenersen (2004) defined average daily intake (ADI) as the amount of pesticides in mg/kg of body weight that can be ingested daily by humans for a life time without causing noticeable adverse effect. Also, according to Stenersen (2004), ADI is estimated by dividing the no observed adverse effect level (NOEL) by the unsafety factor. Classification of dose intake is presented in Table A.3. The unsafety factor of one hundred or one thousand can be used depending on the pre-set conditions (Stenersen, 2004). According to Stenersen (2004), the unsafety factor of one hundred imply that humans are considered to be ten times more sensitive than the most sensitive test organism. Also, due to differences in toxicodynamics and

toxicokinetics, some humans may be two and five times more sensitive respectively i.e. $10 \times 2 \times 5$ (Stenersen, 2004). Some ADI values are given in Table A.4.

2.2 Why pesticides are toxic

Pesticides contain active ingredients that make them toxic and poisonous. The active ingredients are the substances that when in contact with pests can cause death or retards their activity. The toxicological property of the active substance is determined by its molecular structure, its biochemistry and the physiology of the organism under exposure (Stenersen, 2004). A good example is (i) organophosphate pesticides have shorter half-life in the soil environment but are highly toxic to humans compared to organochlorines, and (ii) acute oral dose LD_{50} for rats for parathion is 6-12 mg/kg and DDT is 100-400 mg/kg (Kearney & Roberts, 1998). The toxicity of pesticides is measured by their endpoints. Fundamental endpoints for non-human organisms are death, reduced reproduction (Mehrpour et al., 2014), reduced growth and behavioral change. Human endpoints include well-being and health, and risk of cancer (Stenersen, 2004).

The burning of pesticide treated biomass oxidizes nitrogen, sulphur, chlorine and phosphorous fragments that are present in pesticides to toxic compounds that when inhaled causes human death (Chen et al., 2012). Inefficient burning results to the yield of Polycyclic aromatic hydrocarbons (PAHs) and semi-volatile organics and un-burnt parental residues. An open-air burning experiment revealed that 0.02 g of Thimet (phorate) was released to the atmosphere while the solid residue was 0.05g from each bag. An empty 22.7 kg bag of Thimet (phorate) granules contain on average 4.8 g of formulated material (one gram active ingredient). Yearly accumulation of such residues would yield large quantities of residues (Kearney & Roberts, 1998). Despite all of the above risks, in most developing countries farmers burn the biomass left on their farms in preparation for new agricultural season. Most of these farmers are not aware that the burning of pesticide-treated biomass releases toxic products which could be dangerous to their health, health of other people, and the ecosystem (Chen et al., 2012).

2.3 How pesticides work

Pesticides work in a different ways. Some pesticides inhibit Adenosine Triphosphate (ATP) synthase enzyme which is responsible for the production of ATP from Adenosine diphosphate (ADP) in the inner mitochondria or chloroplast membrane (Stenersen, 2004). Most insecticides target an enzyme acetylcholinesterase (AChE). This is why insecticides are nerve poisons. The poison blocks the sodium channels in the nerves leading to paralysis. Most insecticides have lethal dose value in 50% of the population in mammals from 1-1,000 mg/kg (Stenersen, 2004). The AChE hydrolyzes acetylcholine, an ester, which is released when a nerve impulse is transmitted between nerve cells, from a nerve cell to a muscle or to an endocrine cell. The poisoning is described by the low level of this enzyme, ideally below the threshold level (Stenersen, 2004). Organochlorines and carbamates inhibit the hydrolysis of AChE causing overstimulation of the cholinergic synapses. Some pesticides are behavior-modifying compounds (Stenersen, 2004).

2.4 Global pesticide use

Pesticides are commonly used in agriculture and urban settings. In agriculture, herbicides are used for weed control to reduce human labor and to increase energy efficiency by reducing cycle of primary tillage or secondary tillage especially when growing corn or soybean (Stephenson, 2001; Stephenson, 2003). Some of the herbicides such as glyphosate are widely used along the highways and in urban environment for weed control. This poses a potential point-source environmental contamination along the roadside and the nearby public land (Huang et al., 2004).

Agriculture accounted for the 87% of the pesticides used primarily herbicides in Europe, USA and Japan whereas the remaining 13% was dominated by insecticides which was widely used in Asia, Latin America, and Africa (Stenersen, 2004). The global and USA pesticide use by the year 1995 is presented in Table A.5. Pesticides could increase global food production by 30-60% of the theoretical potential (Zadoks, 1996). In Shinyanga, Tanzania cotton yields improved from less than 30 kg/ha to more than 700 kg/ha while farmers in Hai District in Kilimanjaro region harvested about 1,870 kg/ha of coffee compared to the farmers who did not use pesticides harvesting about 300 kg/ha (Agenda, 2006).

The rise in pesticide use has been due to the need to raise agricultural and livestock production and for human safety from pests and vector borne diseases (Figure 2.1). Between the year 1943 and 1974 the production of dichlorodiphenyltrichloroethane (DDT) alone was estimated at 2.8 million tonnes and the world consumption of pesticides was estimated at 2.6 million tonnes in year 1995 (Stenersen, 2004). Since year 1955 people in different parts of the world have been using pesticides for agricultural activities (Landau-Ossondo et al., 2009). In addition, the rise in pesticide use could be due to the incentives offered by the governments (Ngowi et al., 2016).

For the past 50 years, pesticides have played major roles in tripling world food production (Stephenson, 2001; Stephenson, 2003). Most of the pesticides used by then were organochlorines such as DDT (Landau-Ossondo et al., 2009) which was considered to be very effective because it had the capacity to destroy variety of pests (Stenersen, 2004).

The intensive use of pesticides for the past 50 years has resulted to high contamination of soils, fresh water and the agricultural food (Landau-Ossondo et al., 2009; Mehta et al., 1990) posing health risks to humans and other living organisms. Pimentel (1995) discovered that about 0.3% of the pesticides applied go into direct contact with the target pests, while 99.7% of the pesticides applied go somewhere else in the environment. Kearney & Roberts (1998) reported that when pesticides are applied at normal agricultural rates approximately <1-4.5 kg a.i/ha the residue in surface soils range from less than 0.5 to 2.5 ppm based on the assumption that the surface of soil (15 cm) weighs approximately 2,200 tonnes/ha furrow slice.

Most pesticides that are used in developing countries are imported from the developed countries (Satapornvanit et al., 2004). They are neither well studied nor tested in developing countries with respect to climate or weather conditions. Therefore, limited information exists on their fate, transport and effects based on local environmental conditions (Racke et al., 1997). The available information is often from temperate regions where it was developed, tested and well studied (Malherbe, 2013). Inherent hazard of the pesticides are the same worldwide. However, the exposure, depending on traditions, climate, and agriculture techniques could differ from country to country.

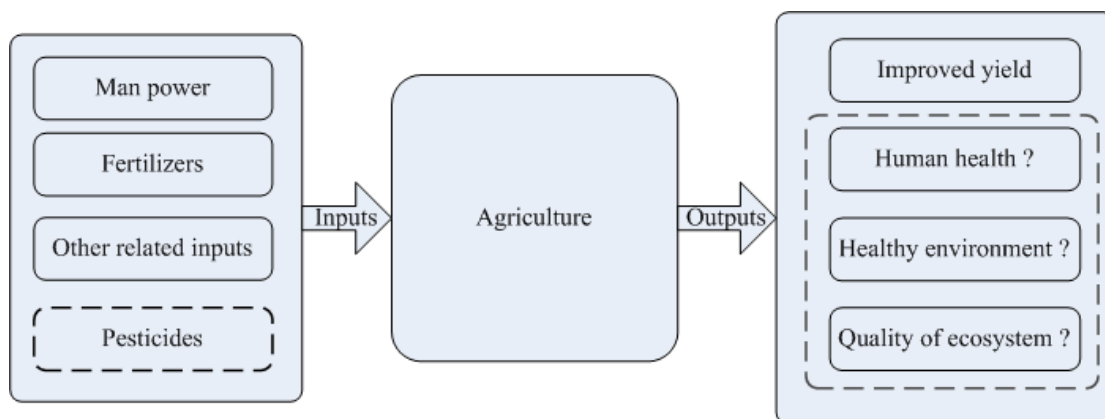


Figure 2.1 Rationale of the study.

2.5 Impacts and problems associated with the use of pesticides

Application of pesticide could cause pesticide residues on food crops and in the environment. Pesticide residues refer to the amounts of pesticides that may remain on/in air, soil, water, food, etc. after they are applied. Margni et al. (2002) demonstrated that food intake is responsible for higher exposure of pesticides to humans by a factor of 10^3 to 10^5 than the exposure from drinking water or through inhalation. Pesticides-related health symptoms include skin problems, dizziness, and headache (Ngowi et al., 2007a; Stenersen, 2004). Pesticide exposure is also associated with prostate and breast cancer (Damalas et al., 2008; Landau-Ossondo et al., 2009), high mortality rates, vomiting and lost consciousness; caused by organophosphorus pesticides, a major problem in China (Dong et al., 2013), hormone disruption through endocrine disrupting mechanisms (Landau-Ossondo et al., 2009; Stenersen, 2004), and problems with reproduction and fetal development. DDT, which is currently used in Malaria control, causes whole-body tremor and pyrethroids cause salivation and choreoathetosis in mammals. DDT and pyrethroids react with the voltage-gated sodium channel. Pyrethroids poisoning prolongs the period the sodium channels are in open position thus sodium leaks out when the channel should be closed. Opening and closing normally occurs in less than a millisecond when an impulse passes (Stenersen, 2004). Natural pyrethroids are derived from pyrethrums, which are extracted from flowers of plant species chrysanthemum (Stenersen, 2004).

The effects of pesticides in the environment depend on their interaction, distribution and both chemical stability and decomposition, and the seasonal

variations (Kapka-Skrzypczak et al., 2013). The effect of climate change on fate and transport of pesticide is however, quite variable and unpredictable (Bloomfield et al., 2006). The environmental risks associated with pesticides are functions of amount of pesticides applied, partitioning coefficient, breakdown (degradation), and fate and transport in the environment, while the risk to human health is associated with the degree of toxicity and the duration of exposure (van der Werf, 1996). Fate and transport of pesticides in the soils depends on type of soils, pesticides' properties, type of crops, and the environmental conditions (Berg et al., 1999). The most widely studied fate of pesticides include adsorption; the binding of pesticides to organic matters and minerals, transfer movement in the environment and degradation (Weijiang et al., 1997). Temperature affects the degradation kinetics of pesticides by affecting the adsorption, solubility and hydrolysis in the soil (Chowdhury et al., 2008; Racke et al., 1997). Kearney & Roberts (1998) reported that in the environment, the rate of pesticide degradation by microorganism is affected by the temperature, water potential and the pH. The effects are specific to type of organisms (e.g. low moisture content and low pH favors fungi than bacteria).

Studies have demonstrated that spray drifts are major source of pesticide exposures in non-target areas around the agricultural fields such as nearby source of waters, bystanders, farm workers, livestock, etc (Bird et al., 2002; Butler Ellis et al., 2010; Gil & Sinfort, 2005; Gil et al., 2007; Gil et al., 2008; Lebeau et al., 2011; Siebers et al., 2003; Wang & Rautmann, 2008). This drift has resulted to over 90% of the crops being sprayed ineffectively (Ngowi et al., 2016). Pimentel & Levitan (1986) suggested that under good spraying conditions, up to 10% of the amount of pesticides applied in the field could be carried away by wind drifts to other non-target sites when using spray booms with nozzles in downward direction near both the ground and the crop. Also, Pimentel & Levitan (1986) reported that at the time of spraying, if there is wind of about 4.44 m/s and if sizes of the spray droplets are 50 μ , about 30-50% of the applied dose could be carried away from the sprayed field by wind drifts. Audsley et al. (1997) reported that 10% of the applied dose could be carried away by wind drifts from the agricultural fields as spray drifts. Eker et al. (2006) and Schrübbers et al. (2014) reported that up to 10% of the applied glyphosate could be moved by wind drifts as spray drifts to non-target plants. Nordby & Skuterud (1974) concluded that

under the ideal spraying conditions and with spraying equipment correctly adjusted, spray drift was 14% of the applied dose, while under the unfavourable conditions and with spraying equipment wrongly adjusted; spray drift was 37% of the applied dose. Roider et al. (2007) reported that herbicide drift could be about 1-10% of the applied dose while Emans et al. (1992) reported that 1-10% of the applied dose could be carried away by wind drifts from the agricultural fields when using boom sprayers.

One of the factors influencing spray drifts is non compliance to directions of usage. Other factors include size of the nozzle, spray pressure, size of the droplets, humidity, air temperature, operating speed, wind speed, application rate, and spray heights (Rautmann et al., 1999; Technologies, 2015). Also, formulation of the pesticide, application methods, choice of the spraying equipment, and the spraying techniques influences spray drifts particles (Berg et al., 1999). Technologies (2015) have demonstrated that small drops less than 200 μm could easily be moved by wind from target areas during application, while Farooq et al. (2001) have demonstrated that droplets are likely to drift at higher pressures. Wang & Rautmann (2008) demonstrated that wind speed, type of nozzle, pressure used during spraying, and the amount of relative humidity, have high influence on spray drifts. The continued increase in spray drifts have raised concerns which has led to the use of portable devices (Zhu et al., 2011), models for predicting, and quantifying the exposure of spray drifts for management of spray drifts as well as for issuing informed decisions. Generally, most of these models employ different approaches, and modeling platforms, thus advances our understanding (Baetens et al., 2007; Craig, 2004; Holterman et al., 1997; Kennedy et al., 2012; Lebeau et al., 2011; Thompson & Ley, 1983).

2.5.1 Thailand

Agriculture is the main activity in most developing countries, Thailand is no exception; with about 42% of the working population depending on it (Kaewboonchoo et al., 2015). Major agricultural crop grown in Thailand include maize, rice, cassava, sugarcane, and rubber (Ekasingh et al., 2004). Majority of the Thai farmers use pesticides inefficiently (Kaewboonchoo et al., 2015; Poapongsakorn et al., 1998). In Thailand, there was no significant difference between the use of less

hazardous and hazardous pesticides by the farmers who follow good agricultural practices standards (Schreinemachers et al., 2012). Although the majority of Thai maize farmers possess high knowledge, positive attitude, and good agricultural practices, they however, bears herbicide poisoning symptoms which is caused by unsafe practices (Kaewboonchoo et al., 2015; Raksanam et al., 2012) such as misuse of protective gears when spraying (Wongwichit et al., 2012b) and the use of banned pesticides such as endosulfan (Plianbangchang et al., 2009).

Between the year 1987 and 1996, about 303,689 metric tons of pesticides were imported. About 6,187 metric tons of glyphosate and 2,665 metric tons of 2,4-D were amongst the top pesticides imported in year 1995 (Thapinta & Hudak, 2000). Glyphosate is a widely used herbicide in the country with about 43% usage (Kaewboonchoo et al., 2015). In year 2010, the use of glyphosate and paraquat accounted for 41% of all pesticides used in the country (Praneetvatakul et al., 2013). From year 2000 to 2010, importation of pesticides (total) increased four-fold from about 30,000 tons to 120,000 tons; with the importation of herbicides alone rising from 20,000 tons to 80,000 tons (Panuwet et al., 2012). Abhilash & Singh (2009) reported that country's pesticide consumptions were 132,509 tonnes of products (49,108 tonnes of active ingredients). From year 2000-2009, the country had experienced an increase of 9.1% of pesticide use per hectare (Schreinemachers & Tipraqsa, 2012). The cost of glyphosate in Thailand ranges from 14-18 US\$ per four liters (Suvittawat, 2014).

Siriwong et al. (2008a) and Siriwong et al. (2008b) reported that there was high risk of cancer at the area around *khlong* seven (Pathumthani, Thailand) due to the contamination of organochlorinated pesticides in fish, edible aquatic invertebrates, and other edible aquatic plants and its plankton communities. In Thailand, pesticide poisoning is highly attributed by the occupational exposure (46%) and suicide (36%) (Thapinta & Hudak, 2000).

2.5.2 Tanzania

The economy of Tanzania is based on rain-fed agriculture with 70% of Tanzanians depending on it (Pauw & Thurlow, 2011), and quite a few depends on irrigated agriculture (Mwakalila, 2006). It is estimated that 90% of the adult

population are farmers (Ngowi, 2002). In year 1993, about 90% of the 26 million people depended on agriculture which accounted for 46% of the GDP (Nkonya et al., 1997). Crops grown in Tanzania include maize, wheat, millet, sorghum, rice, root crops (cassava and potatoes), pulses and oil seeds, and horticulture. Maize is the main staple food crop grown (Paavola, 2008) with the yield estimated at 0.88 tonnes/ha. Although 50% of the harvested land grows maize (Pauw & Thurlow, 2011), still the country depends on importation of cereals such as rice.

The risk of pesticides exposure is huge leading to death and other serious health implications and the environmental pollution (Ngowi et al., 2007a). Some of the victims of pesticides exposure have not been able to identify the cause of their health disorders such as skin diseases due to their poor knowledge on pesticides exposure risks (Ngowi et al., 2007a). Organochlorinated pesticides were detected in Pangani river basin. The water of this river is used for both domestic and agricultural use; therefore, millions are at risk because they heavily depend on it. Although the detected levels, in Tanzania, are far less than the FAO and WHO Maximum Residue Limits (MRL) (FAO & WHO, 2013), the continued use of this contaminated water would certainly result to chronic effect.

The persistence of the organochlorinated pesticides which were banned few decades ago continues to be a country's biggest challenge towards soil remediation (Mahugija, 2013). Residues of lindane, chlorpyrifos, and endosulfan sulphate were found in spring water (in the field) at the concentration of 3.0, 0.27, and 3.35, and 2.8, 0.06, and 3.13 mg/L (Kihampa et al., 2010). Organochlorinated pesticides residues were also detected in fish samples (Mwevura et al., 2002). Organochlorines continues to be pesticides of concerns as they have been detected to other parts of the world including Thailand (Tayaputh, 1996).

About three hundred twenty-nine common names of insecticides, one hundred forty-three herbicides, two hundred twenty-two fungicides, twenty-four acaricides, four rodenticides, one avicide, three nematocides, and seven plant growth regulators were fully registered for general use as of year 2010 for a period of five years (MAFSC, 2008). The consumption of pesticides in Tanzania increased from 0.33 kg/capita in year 1977 to 0.5 kg/capita in year 1988. Pesticide use increased from 2,000 tonnes in mid 1960s to 4,000 tonnes in mid 1970s. The share of pesticide

imports in year 1977 was 47% insecticides, 48% fungicides, and 5% herbicides. Agriculture accounted for the 77%, livestock accounted for 18%, and public health accounted for 5% of the pesticide use (Ak'hbuhaya & Lodenius, 1988). About 14,000 tonnes of pesticides were imported between 1989 and 1992 (Kishimba et al., 2004) and 500 tonnes were imported in year 2000 to 2,500 tonnes in year 2003 (Agenda, 2006).

About 104 pesticides chemical names and 179 trade names were compiled in year 2002 mostly organochlorines, organophosphate, carbamates and pyrethroids. The exact figure on quantity of pesticides used/sold to the agricultural sector is not known, although estimates shows that purchase of pesticides rose five folds between year 1984 and 1986 (Ngowi, 2002). About 736 pesticide poisoning cases (of which 93% were adults) were reported in the country from year 1989-1990 including suicidal cases but the nature of poisoning was not clearly stated with women being the most affected group (Ngowi, 2002). It is also estimated that about 1,000 people die every year from pesticide poisoning (Ak'hbuhaya & Lodenius, 1988). Despite the lack of proper documentation, Ngowi et al. (2007b) estimated the health costs associated with the use of pesticides between 0.018-116 US dollars.

2.5.3 China

The use of pesticides have increased rapidly during the last decades (Zhang & Shan, 2014). Concerns have emerged that this extensive use of pesticides could lead to the deterioration of environmental water quality and human health in the watershed in Yangtze-Huaihe region (Zhang & Shan, 2014). On the other hand, this increase has resulted to the increase in number of poisoning cases. In Zhejiang province, about 20,097 pesticide poisoning cases were recorded with 1,413 deaths. The non-occupational exposure was 79.86%, in which about 14% of the cases were of non-intentional pesticide poisoning (Zhang et al., 2013). Also, Dong et al. (2013) reported that 2 days after exposure to organophosphorus pesticide, a patient developed redness and several blisters on her skin and had burst. The patient tested organophosphorus levels of 332 ng/mL and 490 ng/mL in her blood and urine samples respectively while her cholinesterase level was 251 U/L.

2.5.4 Other countries

It is common practices in developing countries such as India that farmers often take loans to buy agricultural inputs; one of which is pesticides. However, due to poor yields; which lead to less cash returns, some of the farmers fail to pay back their loans. Some of these farmers who fail to pay back their loans, end up committing suicides. From year 1995 to 2010, over 250,000 small farmers committed suicide. Their death could be associated with indebtedness from exploitative markets, unfair trading arrangements, and unscrupulous pesticide suppliers (PAN-UK, 2013).

In Taiwan, from January 1996 to December 2007, about 27,046 patients were hospitalized for Acute Pesticide Poisoning (APP). However, the overall prevalence of the APP per 100,000 patients decreased from 12.43 in year 1996 to 6.87 in year 2007 (Chen et al., 2015).

Pesticide use in Spain include fungicides, herbicides and insecticides which amounted to 100,000 tonnes per year in year 2005 (Vila et al., 2013). A conditional logistic regression analysis found an increase in the risk of cancer at all studied sites, with odds ratios ranging from 1.15 to 3.45 for the population that lived in areas with higher pesticide use. The study suggested that the exposure to pesticides increases a chance for different types of cancer to the general population (Parron et al., 2014).

Landau-Ossondo et al. (2009) reported that organochlorine was the major cause of prostate and breasts cancer in French Caribbean Island, Martinique. In Metropolitan France, based on World standardized rates per 100,000, the prostate cancer incidence was 75.3 and breast cancer incidence was 91.9. In Martinique, the incidence was 152.7 for prostate cancer, and 65.3 for breast cancer. Freire et al. (2013) reported that organochlorines pesticides could affect the thyroid system through gender-specific mechanism that differs from one compound to another.

2.6 Fate and transport of glyphosate

Glyphosate is a post-emergence (Amrhein et al., 1980) non-selective systemic herbicide (Henderson et al., 2010) commonly used in agriculture for weed control (Borggaard & Gimsing, 2008). According to the International Union of Pure and Applied Chemistry (IUPAC), it is named as *N*-(phosphonomethyl)glycine. Its molecular mass is 169.07 g/mol (Henderson et al., 2010; Laitinen et al., 2007). Its

molecular formula is $C_3H_8NO_5P$ and its Chemical Abstracts Service (CAS) registry number is 1071-83-6 (Mensah et al., 2014).

Depending on the type of use, WHO recommends an application rate of 0.3-5.8 kg a.i/ha (Lajmanovich et al., 2003). Its residues are immobile in the soils. It is stable to both chemical and photo decomposition. Its formulation includes monoammonium salt, isopropylamine salt, diammonium salt, sodium salt, potassium salt, trimethylsulfonium/trimesium salt, and an acid. Maize, soybeans, cotton, and canola are plants that are resistant to glyphosate application. It is considered to be non-volatile with vapour pressures between 1.84×10^{-7} mmHg and 6.75×10^{-8} mmHg at 25 °C; hence its exposure to inhalation is less significant. Its solubility in water ranges from 11,500 mg/L (Stenersen, 2004) to 900,000 mg/L (Kearney & Roberts, 1998). Water solubility of its main metabolite; AMPA is 5.8 g/L (Farenhorst et al., 2009).

Its no observed effect level (NOEL) on animals is 750 mg/kg/day and its chronic reference dose 1.75 mg/kg/day. Its sodium salts act as growth regulator and enhances fruit ripening. Its acute oral LD_{50} is greater than 4,320 mg/kg for rats, greater than 10,000 mg/kg for mice, and 3,530 mg/kg for goats. Its acute dermal LD_{50} for rabbits is greater than 2 g/kg. Based on four hours inhalation study on nose only, its acute inhalation LC_{50} for rats is greater than 4.43 mg/L. When ingested, its metabolism is little and its often excreted unchanged through the urine and feces (Henderson et al., 2010). Single oral dose of 10 mg/kg or 1,000 mg/kg of glyphosate was administered to rats, and after seven days the dose was distributed throughout the body, concentrated in the bone. Glyphosate was completely removed from the body of rats 168 h after the administration (Henderson et al., 2010). Also, the toxicity of technical grade glyphosate ranged from slightly toxic to non-toxic to freshwater fish, with a 48 h LC_{50} values from 24 mg/L to 140 mg/L (Henderson et al., 2010).

On target organisms i.e. weed plants, glyphosate is absorbed, translocated and deposited on meristems. About 30-36% of the glyphosate ingested is absorbed. It affects (cause disorder of) the shikimic acid pathway by inhibiting the enzyme 5-enolpyruvylshikimate-3-phosphate (EPSP) synthase which result to deficiency in EPSP production and lead to the reduction in aromatic amino acids which are essential for the production of protein and general plant growth. The endpoint is the

death of the target plants within 4 to 20 days after its initial application. Its toxicity is considered to be low towards non-target organisms. Henderson et al. (2010) reported that study animals which were exposed to formulations of glyphosate herbicides displayed the following symptoms; lethargy, anorexia, hyper salivation, diarrhea, and vomiting. These symptoms lasted for 2 to 24 h following the exposure (Henderson et al., 2010).

Its effects on humans and the environment depend on its concentration duration of exposure, and frequency of exposure. The effects also depend on the health of the person who is exposed to it as well as on many other environmental conditions and factors (Mulla et al., 1981). In humans, peak dose of glyphosate may not be detected 12 h after it was first ingested (Henderson et al., 2010).

Soil particles absorb glyphosate tightly thus any potential residues become immobile despite its high water solubility. Maximum runoff concentration of less than 1.9% of the initial application of 8.96 kg a.e/ha was found in the runoff water 1 day after the heavy rainfall (Giesy et al., 2000). Half-life of glyphosate varies from 3 to 174 days (Stenersen, 2004), 2 to 197 days (Giesy et al., 2000; Henderson et al., 2010), 3 to 130 days (Schuette, 1998), and 47 days (Kearney & Roberts, 1998). Its half-life in the water varies from about few days to 91 days (Henderson et al., 2010), 4.5 days (Focks et al., 2014a), 7 to 10 weeks in natural water (Struger et al., 2008). Generally, it is considered to have a short half-life and therefore it does not stay longer in the environment (Henderson et al., 2010). Its estimated half-life for dissipation from plants is 4 days (Fantke et al., 2014). Its dissipation in the air is limited due to its non-volatile nature (Kearney & Roberts, 1998). It has no significant vapour pressure therefore its loss through vaporization from the treated surfaces is negligible, although its offset movement is possible. The spray drifts from low boom ground application was estimated to be 4% of the application rate at a distance of 1 meter from the field edge, and 0.6% at a distance of five meters. For an aerial application at 25 m and 75 m the spray drifts was estimated to be 10% and 1% of the application rate respectively (Giesy et al., 2000).

Although glyphosate can enter the environment (in plants, soil and water) through runoff or leaching (Borggaard & Gimsing, 2008) from terrestrial application or through drift during normal or aerial applications, microorganisms (Borggaard &

Gimsing, 2008) mainly bacteria and fungi (Giesy et al., 2000) can however, rapidly degrade it to yield aminomethylphosphonic acid (AMPA) (Rueppel et al., 1977) and Polyethoxylated Tallowamine (POEA). It is estimated that bacteria could convert 99.5% of glyphosate to AMPA and also could convert 2% of AMPA to other metabolites (Nourouzi et al., 2011). Glyphosate in the soil (Contardo-Jara et al., 2009) and water (Rueppel et al., 1977) can be degraded by microorganisms. AMPA is degraded at a slower rate than glyphosate itself (Balci et al., 2009) due to its high adsorption to soils, estimated to take 76 to 240 days. The transportability of glyphosate in the soils is largely affected by the soil type, pH, level of phosphate (Sprankle et al., 1975), temperature, and sunlight (Mallat & Barceló, 1998). The half-life of POEA is estimated at 21 to 42 days, and it is readily degraded by microorganisms (Giesy et al., 2000). These two products are degraded further to carbon dioxide gas (EPA, 1993; Henderson et al., 2010).

Studies have shown that first acid dissociation constants of glyphosate ranges from 0.8-1.2 (Goodwin et al., 2003), and 2.32 (Wauchope, 1976). The organic-carbon partitioning coefficient (K_{oc}) ranges between 9-60,000 L/kg, manufacturer suggested a value of 24,000 L/kg (Battaglin et al., 2005), and 20,100 L/kg (Henderson et al., 2010). Glyphosate and AMPA have strong adsorptive characteristics, therefore, do not likely to move below 15 cm in the soil and most likely cannot move to the groundwater (Borggaard & Gimsing, 2008). In a groundwater monitoring study conducted in the USA, glyphosate was detected in 7 out of 247 wells of which further investigations justified that there were errors in the analytical methodology used (Giesy et al., 2000).

Although glyphosate could easily contaminate surface waters since it adsorbs tightly to soil particles in runoff, it is not easily broken down by either water or sunlight (EPA, 1993). However, it dissipates rapidly in standing or flowing water through dilution, dispersion, adsorption to suspended particles and microbial degradation, within few days to two weeks. For standing water, chemical, physical and biological factors determine its dissipation. The half-life of AMPA in the water is similar to the half-life of glyphosate (Giesy et al., 2000). In addition, glyphosate is considered to be environmentally neutral (Borggaard & Gimsing, 2008). It is considered not to be bio-accumulative in animal tissues because of its high water

solubility and a very low log P_{ow} of -3.2 (Contardo-Jara et al., 2009) and it is slightly toxic to fish, birds, and aquatic invertebrates. Although the use of glyphosate has recently increased, its environmental fate is not fully studied (Battaglin et al., 2005) especially in the tropical regions and developing countries. In addition, little is known about its degradation in plants (Laitinen et al., 2007).

2.7 Maize crop and its life cycle

The scientific name of maize is *Zea mays*. Maize crops are wind-pollinated. Its optimum temperature for growth is about 25-27 °C. Maize is generally grown in April and harvested in July or August and takes about 120 to 130 days for maize to be harvested (Edwards, 2009). Threshold temperature for maize development ranges from 5 °C to 10 °C (Brewbaker, 2003; Dijkhuis, 1956; Edwards, 2009). The ideal soil for growing maize is heavy-textured soils, such as clay and clay loam; also, maize also grows well in areas with sandy loam soils (with good drainage) as well as on reddish and black loam soils (Ekasingh et al., 2004). Denmead & Shaw (1960) demonstrated that prior to silking, soil moisture stress reduces grain yield by 25%, at silking, moisture stress reduces grain yield by 50%, and after silking, moisture stress reduces grain yield by 21%. Çakir (2004) demonstrated that vegetative growth is highly affected by the water stress at different growth stages. The growth stages of maize can be categorized into vegetative and reproductive stages. More precisely its growth stages can be divided into 4 main stages namely seedling, vegetative, flowering and fertilization, and grain filling and maturity (Çakir, 2004).

Maize is more tolerant to diseases. Major pest and diseases include weed (Wongwichit et al., 2012a; Wongwichit et al., 2012b), downy mildew, rust, rats, and stem borers (Ekasingh et al., 2004). Weeds such as itchgrass in maize field could be controlled by the application of herbicide nicosulfuron at the rate of 1,500 g a.i/ha (Waramit & Suwanketnikom, 1999; Waramit & Suwanketnikom, 2000). Herbicides such as atrazine, metribuzin, alachlor, and 2,4-D Amine are often used for pre-emergence and post-emergence weed control in maize (MAFSC, 2008). Maize is resistant to glyphosate application. Using glyphosate with an application rate of 2.5 kg a.i/ha could provide weed control by 80% while 3 kg a.i/ha by 90% (Sopit, 2012; Suvittawat, 2014).

Maize was first planted in Thailand in year 1950. North Thailand is the largest maize producer. About 90% of maize is planted in April to July and 10% towards the end of July or August. The average seeding rate of maize is 16.9 kg/ha (Ekasingh et al., 2004). It is grown for both human consumptions and animal feed. In year 1984 to 1985, maize was planted on about 2 million hectares of land. In year 2000, maize was planted on about 1.35 million hectares of land. This decrease could be due to the increase in the demand for land for growing other crops. In year 1984, the country exported about 3-3.7 million tonnes of maize. The country earned about 400 million US\$. Unlike Thailand, maize is the staple cereal crop grown and consumed in Tanzania (Paavola, 2008) with the yield estimated at 0.88 tonnes/ha. It was estimated that maize contributed to 60% of the food calorie intake of the consumers (Kaliba et al., 2000). Per capita maize consumption was estimated at 112.5 kg/year while the national maize consumption was estimated at 3 million tons/year (Kaliba et al., 2000). About 50% of the harvested land in Tanzania grows maize (Pauw & Thurlow, 2011).

2.8 Amphibians

2.8.1 Status

Studies show that population of amphibians have rapidly been declining worldwide than mammals and birds (Stuart et al., 2004; Wyman, 1990) due to diseases such as chytrid fungus, *Batrachochytrium dendrobatidis* (McLeod et al., 2008), loss of habitat, and other unidentified processes which threaten up to 48% of the declining species (Stuart et al., 2004). Other reasons include contamination of chemicals in aquatic and terrestrial habitats; environments required by amphibians. These chemicals include fertilizers, insecticides, herbicides (Davidson, 2004; Figueiredo & de Jesus Rodrigues, 2014; Mann et al., 2009), widely used in agriculture (Dinehart et al., 2009). The continued rate of decline have however resulted to extinction of local species thus resulting to loss of species (Blaustein et al., 1994). It is estimated that one hundred twenty species of amphibians have become extinct during the past decades (Stuart et al., 2004).

The decline of amphibians have an impact on balance of nutrient cycles, change of dynamics of organic matter, energy transfers between riparian habitats and streams is reduced, and change in algal growth in the ecosystem (Whiles et al., 2006).

They also feed on mosquitoes (DuRant & Hopkins, 2008) thus reducing the possibility of disease outbreak such as malaria and rift-valley fever. Therefore, population decline/extinction of amphibians would results to an increase in harmful insect population.

Amphibians such as European Common Brown Frog (*Rana temporaria*) produce secretions (more than 76 different antimicrobial peptides) which are beneficial to human health as their secretions are used in pharmaceutical industries for preventing both pathogenic and antibiotic resistant bacterial strains (Samgina et al., 2012). Despite the fact that nearly less than one-third of the known genera of amphibians have been examined for potential biologically active substances (Daly et al., 2004), extinction of variety of species would impact the advances in human medicine.

Most amphibians require good quality habitat; therefore, any potential disturbances such as toxic chemicals would make them susceptible. Due to their sensitivity, they are good environmental indicators for ecological studies such as disease, loss of habitat, and pollution (Hopkins, 2007). Landscape has greater influence on sex ratio of offspring in frog populations (Lambert et al., 2015). Lambert et al. (2015) found that the sex ratio of green frog (*Lithobates clamitans*/*Rana clamitans*) is dominated by females. Navarro-Martín et al. (2014) found that there was no significant difference in sex ratio between males and females for tadpoles of wood frogs (*Lithobates sylvaticus*) from the theoretical value of 50%:50% after the application of glyphosate-based herbicides. However, sex ratio for bullfrogs (*Rana catesbeiana*) have been reported to be fewer males than females (Howard, 1981).

2.8.2 Frogs in Thailand

There are 130 species of amphibians in Thailand (Khonsue & Thirakhupt, 2001). Frogs are widely used for food. Due to their economic importance and market demand, bullfrogs were introduced (Hero & Roberts, 2004). Frog species found in Thailand include *Microhyla inornata* (Heyer & Berven, 1973), commonly found in other parts of the Southeast Asia such as Vietnam, Lao People's Democratic Republic, China, Indonesia, Cambodia, Malaysia, and Andaman Island (India). Its synonym is *Micryletta inornata*. This species breeds rapidly usually after the heavy rains usually

in small ponds. It is widely found in disturbed places often at the forest edge (van Dijk et al., 2009). This species is hardly found near the agricultural areas and residential areas. Although population trend of this species is considered to be stable, further research on population size, distribution and trends is still needed (van Dijk et al., 2009). Assuming that this species lives in the vicinity of the agricultural land, the main threat is that human (Thais) uses it for food (van Dijk et al., 2009) as well as exposure to runoff of pesticides from the agricultural land to freshwater.

Other species include *Rana nigrovittata* (Khonsue et al., 2000; Matsui et al., 2001) commonly known as sapgreen stream frog, *Fejervarya limnocharis* commonly known as rice frog (Sumida et al., 2007), *Rhacophorus jarujini* known as tree frog (Matsui & Panha, 2006), *Leptolalax fuliginosus*, *Leptolalax melanoleucus*, and *Leptolalax solus* (Matsui, 2006), *Limnonectes kuhlii* known as large-headed frog (McLeod, 2008; McLeod, 2010), *Theloderma licin* also known as the smooth frogs are commonly found in the gardens of the resort areas (McLeod & Norhayati, 2007), *Ansonia inthanon* (Matsui et al., 1998) of which its skin is widely used (van Dijk & Khonsue, 2004), *Ansonia kraensis* (Matsui et al., 2005), *Amolops panhai* (Matsui & Nabhitabhata, 2006), *Ansonia siamensis* (Heang, 1984), *Rana banaorum* and *Rana morafkai* (Bain & Stuart, 2005). Additional list of species is presented by Daly et al. (2004) and Pauwels et al. (2002).

In this study, *Crinia insignifera* was used to simulate population dynamics of non-target species. This species is not native to Thailand. It is native to Australia (Moore, 1954). This species was selected because it is found in subtropical or tropical seasonal and permanent wetlands (Aubret, 2004), conditions similar in Thailand and Tanzania. The study assumed that both pre-metamorphic and post-metamorphic lives of this species would be exposed to glyphosate after its contamination in freshwater. Females of this species lay about 66 to 268 eggs and its tadpoles develop in about 3 to 5 months (Hero & Roberts, 2004; Tyler & Knight, 2011).

Most parts of the country have canals commonly known as *khlong* connected to small ponds, streams, lakes, and rivers; environments needed for the life cycle of amphibians. More often, these canals surround the agricultural fields; therefore, there is a chance of pesticide contamination into these canals. Since water bodies in small ponds, streams, lakes, and rivers have been polluted by various kinds of pesticides

(Thapinta & Hudak, 2000) due to spray drifts, direct contamination, runoff from the sprayed agricultural field, etc., the question is what would happen to species of amphibians in these pesticide contaminated waters and the surroundings? Are there potential impacts to their population dynamics? These were some of the questions that inspired the present study.

2.8.3 Effects of pesticides on frogs

To study the effects of pesticides on amphibians, standard tests on mammals and birds have been used as proxy for terrestrial amphibians life stages while standard tests on fish and other aquatic invertebrates have been used as proxy for aquatic amphibian's life stages (Relyea, 2011). For the first time, 20 years after Roundup® and its formulations entered the commercial market, a study was conducted by Bidwell & Gorrie (1995) on potential impacts of these herbicides to amphibians. Ever since there has been a continuous debate and exchange of scientific arguments concerning the effects of Roundup® on amphibians (Relyea, 2011).

Perkins et al. (2000) conducted an experiment to study the toxicity of glyphosate on the embryonic development of *Xenopus laevis* (The African clawed frog) using Frog Embryo Teratogenesis Assay-*Xenopus* (FETAX). The 96 h LC₅₀ of Roundup® (the IPA salt of glyphosate formulated with a surfactant) was 9.4 mg a.e/L (12.5 mg ai/L), i.e. 1 mg a.i/L equals to 0.75 mg a.e/L (Relyea, 2006). The study found that mortality increased with the increase in exposure concentration. Malformations in control embryos such as uncoiling of the gut, abnormal pigmentation, blistering, edema, and axial twisting were below 5%. Glyphosate had no influence on the ninety-six hours growth of *Xenopus laevis* embryos, see Table A.6 (Perkins et al., 2000).

Mann (2000) conducted a study on 4 Australian frog species; *Crinia insignifera*, *Helcioporus eyrei*, *Limnodynastes dorsalis*, and *Litoria moorei*. The study found that Roundup Biactive (MON 77920) was non-toxic to the tadpoles of the selected species with the 48 h LC₅₀ values of 911 mg/L for *Litoria moorei*, over 1,000 mg/L for *Crinia insignifera*, *Helcioporus eyrei*, and *Limnodynastes dorsalis*. Glyphosate isopropylamine had no mortality effect to all tadpoles of the selected 4 species over 48 h, for concentrations between 503-684 mg/L. The study concluded

that the toxicity of technical grade glyphosate acid at a 48 h LC₅₀ of 81.2-121 mg/L was likely to be caused by the acid intolerance. Also, tadpoles were more sensitive than adult frogs and newly emerged metamorphs (Mann & Bidwell, 1999). Relyea (2011) found that typical application rates of glyphosates can kill up to 30% of the adult frog. Adult and juvenile green frogs (*Lithobates clamitans*) were exposed to glyphosate-based herbicide at a rate of 2.16 kg a.e/ha and 4.27 kg a.e/ha for 14 days in littoral enclosures. The survival rate was found to be 70% (i.e. mortality 30%). The study did not attribute mortality due to *Bd* infection on the toe webbing which was 45% of the species studied (Edge et al., 2011). Abnormalities shown by tadpole often which are not easily quantifiable, include slow swimming ability before death, bent tails, early metamorphosis events, erupted forelimbs and head out of water (Cauble & Wagner, 2005). Other abnormalities include gonadal abnormalities, decreased length of the snout-vent at metamorphosis, and increased time to metamorphosis (Howe et al., 2004). According to Howe et al. (2004), these abnormalities possibly are due to the disruption of the hormone signaling caused by the elevated exposure to formulations containing POEA and glyphosate.

Lajmanovich et al. (2013) found that a recommended application rate of 2.3 kg a.i/ha for glyphosate applied by mixing with water at a rate of 150 L/ha resulted to initial glyphosate concentration of 11,454.6 mg a.i/L, was enough to kill more than 50% of the tadpole. However, Table A.7 shows that there are variations in pesticide tolerance for different species of amphibians (Bridges & Semlitsch, 2000; Howe et al., 2004).

Dinehart et al. (2009) conducted a study on survival of the juvenile New Mexico spadefoots (*Spea multiplicata*) and Great Plains toads (*Bufo cognatus*) from the Southern High Plains (USA); 48 h after the direct exposure to Roundup Weed and Grass Killer Super Concentrate[®], Roundup WeatherMAX[®], and Roundup Weed and Grass Killer Ready-To-Use Plus[®] performed on soil and moist paper towels (Table A.8). Results showed that the survival of the juvenile New Mexico spadefoots (*Spea multiplicata*) and Great Plains toads (*Bufo cognatus*) were significantly reduced by the exposure to Roundup Weed and Grass Killer Ready-To-Use Plus[®] both on soil and moist paper towels. The survival of the Great Plains toad was reduced by the exposure to Roundup Weed and Grass Killer Super Concentrate[®] on paper towels.

However, results showed that New Mexico spadefoot and Great Plains toad survival were not significantly affected by the exposure to both Ignite[®] 280 SL and Roundup WeatherMAX[®] on both soil and moist paper towels. Dinehart et al. (2009) concluded that under field conditions, these herbicides may be not posing potential risks to the species of frogs studied.

Relyea (2005b) conducted a toxicity study of Roundup on amphibians in an outdoor setting to mimic a natural environment by assembling communities of three species of the North American tadpoles. The settings contained different types of soils which could absorb the Roundup applied following direct overspray. Relyea (2005b) found that Roundup killed 96-100% of the larval amphibians 3 weeks after the first exposure. Relyea (2005b) conducted a follow up experiment by exposing three species of juvenile anurans (post-metamorphic); to a direct overspray of Roundup in laboratory settings. Relyea (2005b) found that Roundup killed 68-86% of the juvenile amphibians after 1 day of the exposure. These results suggest that Roundup could cause high mortality rates (i.e. low survival rates) to amphibians that are exposed to it; hence amphibians' population might be on the verge of declining.

Relyea & Jones (2009) conducted a toxicity study of Roundup Original Max[®] to test the lethal concentrations required to kill 50% of the population (i.e. LC₅₀) of 13 selected species after 96 h of exposures for nine species of larval anurans from 3 families; Ranidae (*Rana pipiens*, *Rana clamitans*, *Rana sylvatica*, *Rana catesbeiana*, and *Rana cascadae*), Bufonidae (*Bufo americanus* and *Bufo boreas*), and Hylidae (*Hyla versicolor* and *Pseudacris crucifer*), and 4 species of larval salamanders from 2 families; Ambystomatidae (*Ambystoma gracile*, *Ambystoma maculatum*, and *Ambystoma laterale*), and Salamandridae (*Notophthalmus viridescens*) from eastern and western North America. Relyea & Jones (2009) found that the 96 h LC₅₀ values for the 9 species of larval anurans were between 0.8-2.0 mg a.e/L (1.1-2.7 mg a.i/L) while the 96 h LC₅₀ values for the 4 species of larval salamanders were between 2.7-3.2 mg a.e/L (3.6-4.3 mg a.i/L).

King & Wagner (2010) examined the toxicity of Roundup[®] on 6 amphibian species namely *Ambystoma macrodactylum*, *Ambystoma gracile*, *Anaxyrus* [*Bufo*] *boreas*, *Rana luteiventris*, *Pseudacris regilla*, and *Rana cascadae* by exposing larvae (tadpoles) of all 6 species to 6 different treatments of

Roundup® of 0 (as control), 0.1, 0.5, 1.0, 2.0, and 5.0 mg a.i./L dilutions of regular glyphosate. The study was monitored for 16 days. The 24 h LC₅₀ varied significantly with the concentrations ranging from 0.43 mg a.i./L for *Pseudacris regilla* to 2.66 mg a.i./L for *Anaxyrus boreas*. The study found that Ambystomatid and Bufonid larvae were less sensitive to Roundup® regular compared to Ranid and Pseudacrid. No 24 h mortality occurred for salamander larva. The average time to death varied from 1 day for *Pseudacris regilla* to 8.3 days for *Ambystoma gracile*. There was no significant differences on average time to death for exposure times longer than 24 h, for *Anaxyrus boreas* and salamanders.

The impact of glyphosate on amphibians depend on its formulations with larvae being the most sensitive life stage (Wagner et al., 2013). Edginton et al. (2004) reported that larvae of *Rana clamitans* and *Bufo americanus* were 1.5-3.8 times more sensitive than their embryos, while the larvae of *Xenopus laevis* and *Rana pipiens* were 6.8-8.9 times more sensitive than their embryos.

Frogs breathe and absorbs water through their skin (Campbell, 1999), therefore, potential contamination in their habitats make them vulnerable. It is this characteristic that makes them good bio-indicators for potential environmental contaminants (Campbell, 1999). Review of the literature show that most available data are associated with the mortality of amphibians and population dynamics (Campbell, 1999) of frogs or toads using the LC₅₀ values (Pérez et al., 2011; Relyea, 2011). Mortality occurs mainly due to chronic exposure, and not due to the presence of acute conditions (Cauble & Wagner, 2005). Although glyphosate and its formulations have shorter half-lives, the main concern is that most amphibians die within a day of exposure (Relyea, 2011). Bernal et al. (2009) concluded that sensitivity of amphibian species to Roundup-type formulations of glyphosate was significantly similar for both tropical and temperate species. Table A.9 shows the categories of toxicity for aquatic organisms.

2.9 The need for simulation modeling

Pesticide interacts with the biological environment in a complex way. Our understanding of the pesticide-biological environment interactions could however be enhanced by computer modeling. A mathematical model can be defined as a set of

equations that represents the behavior of a system (Doyle, 1991). Models are scaled down version of real world things and thus they are used by modelers to understand properties of the real world, particularly are very useful in indentifying research gaps and research planning (Doyle, 1991). However, the real world is infinitely complex, so data are collected and simplified models are built using those data.

Models are widely used in ecology studies as fundamental component of sciences (Axelsen et al., 1997; Bolker, 2008; Calvet, 1995; May, 1974; May & Mclean, 2006; Rinella & Sheley, 2005a; Rinella & Sheley, 2005b). Models could be used for prediction purposes, decision support, and for examining consequences associated with the management changes on biodiversity (Doyle, 1991; Freckleton & Stephens, 2009). Models could also be used to (i) understand basic spatial-temporal dynamics of the subject under study e.g. weeds, and (ii) analyze and understand how different systems function (Doyle, 1991). Results of the simulation models help in designing and planning of experiments as well as in development of new ideas (Doyle, 1991; Holst et al., 2007).

Model simulations are widely used in the European Union to support pesticide registration (Dubus et al., 2002), e.g. to demonstrate that leaching concentrations of pesticides to groundwater do not exceed the acceptable levels (Vanderborght et al., 2011). It is therefore, certain that complex relationship between pesticide and the technosphere be studied using modeling packages. However, the complex interaction between the pesticides and the environment increases when mixture of pesticides are used (Swarcewicz et al., 2013).

Predictive environmental modeling with the use of mathematical models to describe the ecotoxicology of pesticide in the environment, their degradation kinetics and risk to living organisms, have been an area of considerable activity over the last decade. This is because there are different kinds of models that have been built to simulate the interactions in different ways. These models differ in their assumptions, strengths, and weaknesses. van Zelm et al. (2014) have summarized models related to life cycle inventory of pesticides.

Mathematical models have become important tools in ecology to understand the complex system interaction and to predict the short and long-term effects of pesticides (Jepson et al., 2014), thus exposing gaps in the scientific body of

knowledge (Ludwig et al., 2001). Without models, qualitative or rough estimates of the ecological system interaction are likely to be made (Freckleton & Stephens, 2009). These model systems can be successfully applied to provide a rational framework for understanding the ecotoxicology of pesticides and can be used to provide information for quantitative risk assessment (Baker, 1995).

Modeling of ecotoxicology of pesticides does not only contribute to an increased knowledge about pesticide toxicology but it is also essential for either development of new pesticides or optimization of the existing pesticides. In addition, computer-based simulation models could be used to replace costly and time consuming field measurements (Dubus et al., 2002; Ozturk et al., 2012) especially when key questions cannot be answered using conventional field trials (Doyle, 1991). With the help of field data, computer models could be used to quantify the exposure to agricultural chemicals (Gustafson, 1995). Models could also be used to overcome the shortcomings of the laboratory and field experimentation procedures (Bera, 2003). Therefore, their designing must be robust in such a way that could perform the intended integrated prediction (Laender et al., 2014).

2.10 Existing pesticide models

There are a handful of available simulation models. These range from pesticide estimation and transformation in head-water catchments (Gassmann et al., 2013) and modelling of pesticide biodegradation in the soil (Loos, 2010). These models are often designed to reflect the conditions of the country they were intended for. Some models, e.g. PestLCI 2.0 (Dijkman et al., 2012) requires that the user first contact the model developer to request the addition of some data in the database so that the model could be used to reflect the user's study conditions (Renaud-Gentié et al., 2014). The ChimERA project have been developing (2013-2016) a model to assess ecological risk assessment (ERA) to simulate exposures and effects of chemicals and their mixtures. The model is anticipated to represent the wide range of environmental conditions in the European Union (EU) surface waters (Laender et al., 2014).

Tabashnik & Croft (1982) simulated the influence of operational and biological factors on the rate of resistance evolution to pesticides under four

conditions common to agricultural arthropod pests and natural enemies. Gil et al. (2008) studied pesticide loss to the air. The study was implemented by using fuzzy inference models. Racke et al. (1997) recommended the application of models to simulate fate of pesticides under tropical conditions, specifically pesticide dissipation and mobility. Tiktak et al. (2012) simulated the movement of pesticides towards drains with a preferential flow version of PEARL. Adriaanse et al. (2013) estimated the degradation rates of prosulfocarb in outdoor stagnant water by using inverse modelling with TOXSWA. The main and important input parameter was the degradation rate.

Pesticide Root Zone Model (PRZM) could be used to simulate non-linear dissipation kinetics for parent pesticide. The model includes volatilization model (Carsel et al., 1984). Even with limited spatial and temporal windows, a good number of pesticides transport models that describes the behavior of pesticides in the soil, and in the environment have been studied (Arias-Estevez et al., 2008). These models include evaluation of the fate of surface-applied pesticides (Reichman et al., 2000). Run-off models include AGNPS (Young, 1987) and CREAMS (Knisel, 1980). Pesticide transport models in soils include SWACRO (Belmans et al., 1983), PRZM-2 (Mullins et al., 1993), MACRO for water movement and solute transport (Jarvis, 1991), LEACHM (Hutson & Wagenet, 1992), and PESTFADE (Clemente et al., 1993); a process-based non-point source model.

Leaching models include Pesticide Leaching Model (PLM) (Hall, 1994; Nicholls & Hall, 1995) and CRACK-NP. PLM assumes that water moves downward in the soil profile in the ‘tipping-bucket’ fashion (Köhne et al., 2009). CRACK-NP is a leaching model that simulates flow of pesticides in clay soils with shrinkage cracks. This model is used only to study those soils in which macropore flow is the dominant mode of water movement (Armstrong et al., 2000). Dubus et al. (2002) recommended the use of PELMO and PEARL models to simulate the complex degradation kinetics of pesticides, and PEARL, PELMO, PRZM to simulate pesticide losses by volatilization.

CHAPTER 3

METHODOLOGY

3.1 Survey

An online comprehensive semi-structured survey was conducted using questionnaire to survey how aware individuals, in Tanzania, are of pesticides and their associated health effects. The questionnaire had a mixture of both closed and open-ended questions. Respondents were asked to respond to the questionnaire. Data collected were analyzed for descriptive statistics using Microsoft Excel spreadsheets. The questionnaire used in the study is attached in Appendix B.

3.2 Model formulation

Model algorithm has been programmed in standard C++ language using Qt Creator (an integrated development environment) and its libraries (QtCreator, 2016). Fast computational speed and wide applicability on many platforms are the main reasons for choosing C++ language while built-in capacity for creating plug-ins was the main reason for choosing Qt libraries (Holst, 2013). Universal Simulator version 2 (Holst, 2016), a collaborative ecological modelling software package, was used to load and run the model. Model components were specified in the XML files (box script). Functionality of the model components were defined in plug-ins libraries i.e. programmed in C++ language (Holst, 2016). The XML syntax was chosen because its a simple and flexible text format which allows an exchange of wide variety of data on different platforms (Holst, 2010). Notepad++ was the text editor used. When running a box script, Universal Simulator performs the following steps (1) construct all boxes, (2) amend all boxes, (3) run root box; initialize, reset, update, cleanup, and debrief (Figure 3.1). Steps (1) and (2) are performed when a box script is loaded while step (3) is performed when a box script is run. During simulation, boxes are processed from top to bottom; children first (Figure 3.2). Output from the Universal Simulator is then exported to R program (RCoreTeam, 2017) via a text file and also it is copied to the clipboard (Holst, 2016).

Universal Simulator is explained by Holst (2016). This simulator has been used by Holst et al. (2013) to predict the increase in mortality of *Inachis io* larvae

caused by Bt-maize pollen in European farmland, Ozturk et al. (2012) to simulate leaf photosynthesis of C_3 plants under fluctuating light and temperatures, and Mweya et al. (2014) to simulate population dynamics of mosquito vectors for rift valley fever virus in a disease epidemic setting.

The algorithm used in this study is an open source, therefore, codes can be reused and extended by the user (Booch, 1986; Kindler & Krivy, 2011). Users are allowed to change or modify the parameter values depending on the need of their studies (Larocque et al., 2014). Software requirements, software availability, and instructions on how to install and run the model and source codes are provided in Appendix C. The simulation environment is presented in Figure 3.3.

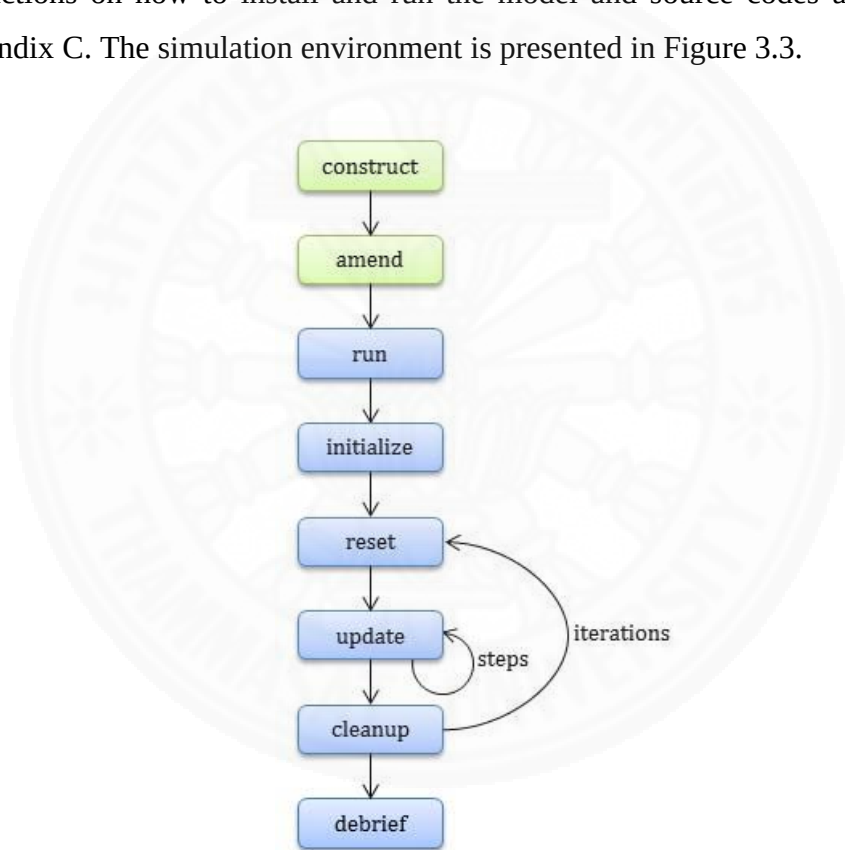


Figure 3.1 Computation steps in Universal Simulator (Holst, 2016).

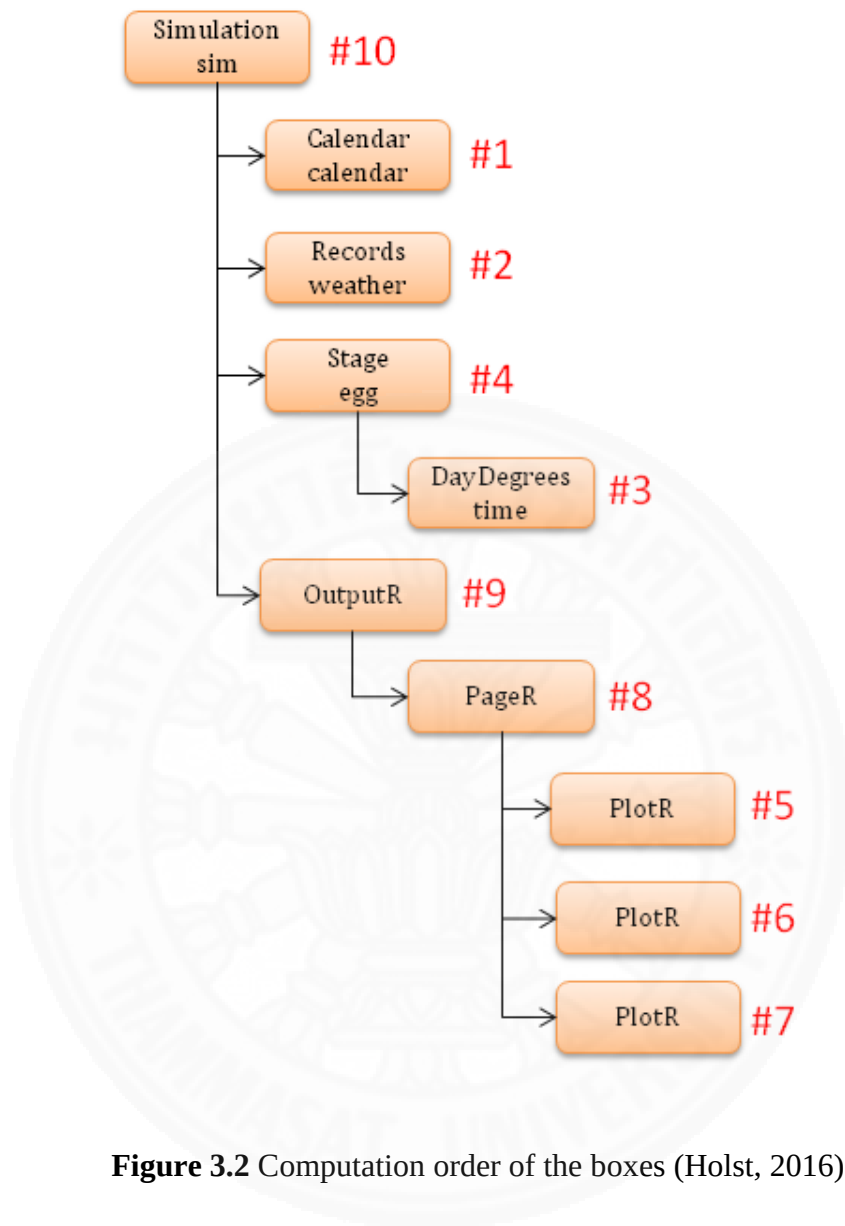


Figure 3.2 Computation order of the boxes (Holst, 2016).

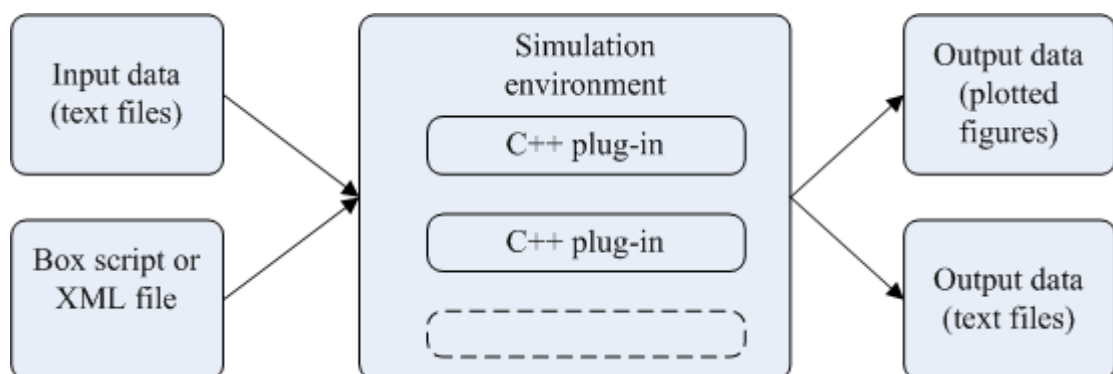


Figure 3.3 Simulation environment, adopted from Holst (2013).

3.3 Model parameterization

3.3.1 Model adoption and modification

The study modified PestLCI models which were initially intended for modelling pesticide inventory in life cycle assessment (Birkved & Hauschild, 2006; Dijkman et al., 2012). By design, PestLCI models estimate the aggregated fractions (amount) of pesticides as emissions to air, surface water, ground water, etc. Then, results from PestLCI models are used as inputs to e.g. USEtox for characterization of toxicity (Xue et al., 2015). Modification was necessary to incorporate daily simulation time steps, crop model, frog population dynamics model, and sensitivity analysis. Therefore, the modified algorithm includes model components (plug-ins) for weather model, crop model, fate and transport model, and frog population dynamics model. All these sub-models (boxes) are connected to each other like objects using inputs and outputs ports (Holst, 2016).

3.3.2 Weather model

The weather model mimics the climate or weather conditions (e.g. rainfall and temperature) of Pathumthani province, Thailand (i.e. study area) for both year 2012 and 2013. These weather data were obtained from the Thai Meteorological Department (TMD, 2014) and were saved in a text file which was linked to the weather model. Additional information is in Table A.10 under weather data.

Weather data were analyzed for statistical differences by one-way analysis of variance (ANOVA) with significant level of the test i.e. alpha value (α) of 0.05, using statistical software Instat Plus (SSC, 2016) to determine variability of rainfall and temperature between year 2012 and 2013. Statistical differences were tested against the null hypothesis (H_0) that there was no significant difference on variability of rainfalls and temperatures for year 2012 and 2013. The study assumed the alternative hypothesis (H_1) that there was significant difference on variability of rainfalls and temperatures for year 2012 and 2013. A value of $p < 0.05$ implies that there was a significant statistical difference while $p > 0.05$ implies that there was no significant statistical difference.

3.3.3 Crop model

Crop model is a stage-structured model that simulates 4 crop development stages namely leaf development, tillering, stem elongation, and senescence. In this study, maize was selected as the study crop. For maize crop, deposition fractions of cereals are used to estimate the amount deposited on the crop/leaf surfaces at each development stage at the time of application. Since the present study modeled glyphosate, applications were done before sowing (on 14th March and 28th March). The study assumes that maize seeds were sown on 4th April for both year 2012 and 2013 under conventional tillage. Degree day values were used to model development (life cycle) of maize crop. Additional information is in Table A.10 under crop data. Also, more details about the crop model are provided in Appendix D, section D.1.

3.3.4 Fate and transport model

Fate and transport model simulates fate and transport behavior of glyphosate in the environment. These multimedia transport phenomena include spray drift, volatilization (on the crop/leaf surface), uptake, photo-degradation, washoff, volatilization (in the topsoil), biodegradation, runoff, and macropore flow (Figure 1.2). They are responsible for the movement of pesticides from one environmental media (point of emission) to other environmental media (van Zelm et al., 2014). A new approach to estimate washoff from the crop/leaf surfaces has been introduced following the approach of Smith & Carsel (1984). The study assumes that mass balance for primary and secondary distribution is as per Figure 1.2.

Leaching processes to groundwater via soil horizon (i.e. soil layers) is not included in this study due to the lack of specific soil characteristics for 10 soil layers from zero to one meter below the topsoil (but leaching via macropores is included). These characteristics include the boundary of start of each soil layer, pH for each layer, percent of clay, silt and sand for each layer, and percent of organic content for each layer. However, Siimes & Kämäri (2003) demonstrated that leaching could be between 0.01-1% of the applied dose. Rasmussen et al. (2015) demonstrated that leaching of glyphosate could be up to 1.7% of the applied dose for a short duration intense rainfall event (1 h) and up to 0.4% when rainfall event was longer than one hour. Geisler et al. (2004) conducted a leaching study up to one meter depth in soils

using a logic tree method to study pesticide leaching in agriculture. The study found that majority of pesticides had a leaching fraction of less than 0.1%. This suggests that majority of pesticides applied remain on the topsoil where it is subjected to volatilization, runoff, and biodegradation. Also, Kladvko et al. (2001) reported that subsurface drainage losses of herbicides could be less than 0.1% of the dose applied while, subsurface drainage losses of mobile insecticides such as carbofuran could be less than 0.5%. In addition, Feng & Thompson (1990) found that more than 90% of the total glyphosate residue is retained in the zero to 15 cm layer of the topsoils. Furthermore, for majority of pesticides, emission to groundwater is higher for sandy soils than for silty and loamy soils (Xue et al., 2015).

Malformation on tadpoles, froglets, and adult frogs were not included due to lack of data. However, Lajmanovich et al. (2003) found that for a 24 h exposure of commercial glyphosate formulations, malformation on tadpoles was low (5%) for a concentration of 3.07 mg/L while malformation was higher (70%) for a concentration of 7.5 mg/L. The percent malformation included both dead and alive tadpoles. In this case, the present study could not model malformation as it was not clear that how many tadpoles died as a result of malformation. Furthermore, percent malformations aggregated malformation of bent curved tails, mouth deformities, and eye abnormalities.

3.3.5 Frog population dynamics model

Frog population dynamics model is a stage-structured model that simulates the growth and development of 4 development stages of frogs namely eggs, tadpoles, froglets, and adults. Female frogs lay about 66 to 268 eggs and its tadpoles develop in about 3 to 5 months (Hero & Roberts, 2004; Tyler & Knight, 2011). The study assumed that the proportion of female frogs in the population were 50% (Navarro-Martín et al., 2014). Also, the study assumed that no eggs are laid if there are less than 2 adult frogs at the time of fertilization. Daily predicted concentrations of glyphosate in the water body are used to simulate mortality rates of eggs, tadpoles, froglets, and adult frogs. Due to the limited availability of data, the study does not account for the mortalities due to natural disasters, over-crowding, and weather conditions i.e. stage-

specific natural (non-pesticide-induced) mortality rates. Also, the study does not account for the migration or harvesting of frogs in the population.

3.3.6 Sensitivity analysis

Sensitivity analysis of the model outputs due to the variations of input parameters in the simulation period is modeled using Latin Hypercube Sampling (LHS) method (Holst, 2016). More details about the frog population dynamics model and the sensitivity analysis are provided in Appendix D, section D.5 and D.6. In addition, Table A.10 has groups of data that can also be applied to other crop types, pesticides, and locations. The data is grouped into general data, field data, crop data, pesticide data, soil data, weather data, and frog data.

3.3.7 Distribution

Assuming that glyphosate distribution was as per Figure 1.2, if spraying was done on day d ; then washoff, runoff, volatilization, photo-degradation, biodegradation, macropore flow, etc. took place, therefore, on day $d+1$; the concentration in each environmental media would be equal to the concentration remained on day d after the removal of fraction or amount washoff, runoff, volatilization, photo-degradation, biodegradation, macropore flow, etc. On day $d+2$; concentration in each environmental media would be the same as the concentration remained on day $d+1$. This allows the estimation of daily mortality rates using the predicted daily concentrations of glyphosate (in the water body) and the LC_{50} values of glyphosate at each development stage of frogs. Equations and assumptions used in the simulation study are described in detail in Appendix D. Data used as model inputs in the simulation study are provided in Table A.10.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Pesticide awareness and management practices in Tanzania

Like in Thailand and other parts of the world, food, water, and a healthy environment are vital components (Gleick, 1998; Lubchenco, 1998; Postel, 2000; Tilman et al., 2009) to human well-being in Sub-Saharan Africa, Tanzania is no exception. With over 70% of the population depending on rain-fed agriculture (Pauw & Thurlow, 2011), Tanzania is among the countries with the lowest income and with its people living in poverty (Adebayo et al., 2013); estimated at less than 2 US\$ per day (Chien & Ravallion, 2001).

Agriculture has been a backbone of the country's economy with the agricultural gross domestic product (GDP) for agricultural sector increased at 4.4% annually from year 1998 to year 2007 (Pauw & Thurlow, 2011). With the land area of about 880,000 km² (Sheya & J.S. Mushi, 2000), the country has had an increase in competition between land and water resources for agricultural production to meet the growing food demand due to the increase in population. Figure 4.1 shows that population has increased from about 32 million people in year 2000 to about 51 million people in year 2015 (World-Bank, 2015). The situation in Tanzania is quite similar to the global situation whereby the world food demand is high (Cranfield et al., 1998) to feed the population of 7 billion people (Lutz & Samir, 2010), which is expected to increase to 8 billion people by the year 2050 (Lutz & Samir, 2010), and to 9 billion people by the year 2070.

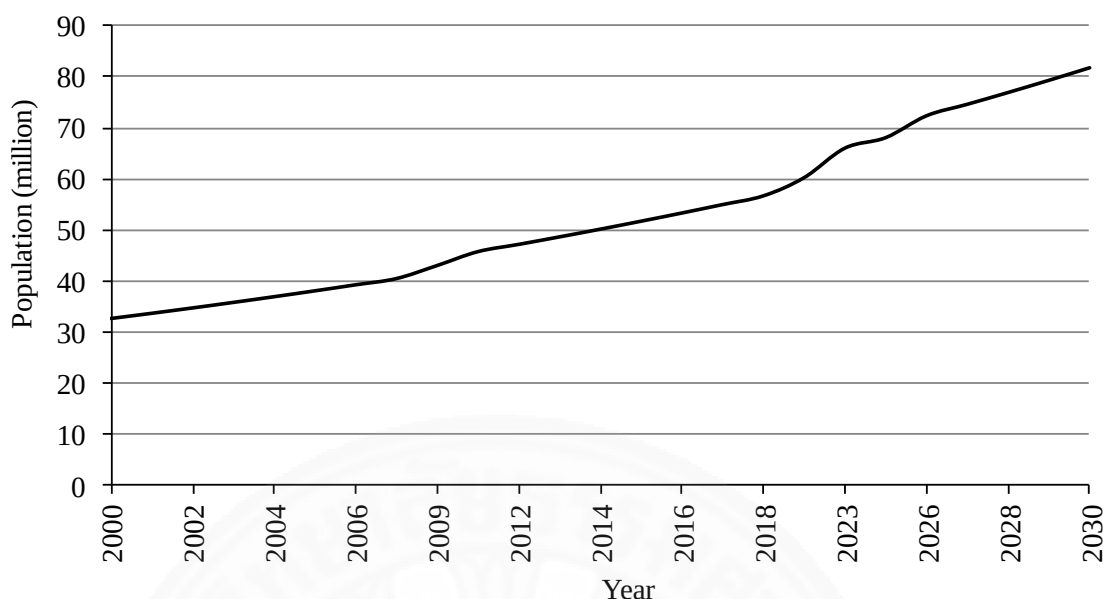


Figure 4.1 Population trend of Tanzania (Felix & Gheewala, 2012).

To meet the food demand to feed this population, there has been a need for the agricultural expansion. During the expansion processes, the main concern is the way agriculture is performed (practiced) as it could lead to undesired environmental as well as human health problems (Tilman et al., 2011) due to non compliance to good agricultural practices (Ganpat et al., 2014). One of the main concerns in agriculture is the use of pesticides. These are toxic chemicals that are intended to kill or repel target pests (Arias-Estevez et al., 2008) to reduce production losses caused by pests (Owusu-Boateng & Amuzu, 2013). Being toxic chemicals, pesticides needs careful attention when using them. Studies have demonstrated that the environmental risk of using pesticides depends on volume of pesticide used, persistence of the pesticide, pesticide mobility in the environment, potential toxicity to non-target organisms, as well as misuse and poor management (Stephenson, 2001; Stephenson, 2003; Wong & Ng, 1984).

Poor pesticide management and practices (Adjrah et al., 2013), misuse, and violation of safety rules (Matyjaszczyk, 2013) pose a potential risk to farmers, non-farmers, consumers, and the environment (Menn, 1980) via contamination on/in food crops (Jeyaratnama, 1990). In addition, poor pesticide management practices have resulted to the increase in pest resistances (Philbert et al., 2014) and number of pesticide poisoning cases. In Tanzania, it is estimated that about 1,000 people die

every year from pesticide poisoning (Ak'hbuhaya & Lodenius, 1988). Pesticide poisoning is a concern not only in Tanzania, but also in many other parts of the world e.g. Thailand. It is estimated that, globally, 220,000 people die each year from pesticides poisoning (Konradsen et al., 2003; Ngowi et al., 2016). Also, it is also estimated that 39,600 Thai farmers are poisoned each year (Lianjumroon, 1999). Thailand has 42% of the working population depending on agriculture (Kaewboonchoo et al., 2015). Majority of the Thai farmers use pesticides inefficiently (Kaewboonchoo et al., 2015; Poapongsakorn et al., 1998). Schreinemachers et al. (2012) demonstrated that in Thailand, there was no significant difference between the use of less hazardous and hazardous pesticides by the farmers who followed good agricultural practices standards. Although the majority of Thai maize farmers had positive attitude, high knowledge, and good agricultural practices, they however, bore herbicide poisoning symptoms which was caused by unsafe practices (Kaewboonchoo et al., 2015; Raksanam et al., 2012) such as not using protective gears when spraying (Wongwichit et al., 2012b) and the use of banned pesticides e.g. endosulfan (Plianbangchang et al., 2009).

To ensure that both Thailand and Tanzania are facing the same problem, a study was conducted to survey the level of awareness and pesticide management practices and their associated health effects in Tanzania. Results of the survey could be of significant importance to those involved in agricultural and pesticide training as well as in policy making.

To accomplish this study, a comprehensive semi-structured survey was prepared and respondents were asked to respond from all over the country. Respondents responded at random as there was no restriction on age, region, education, job, etc. The questionnaire had a mixture of both closed and open-ended questions. Respondents were asked to respond to the questionnaire referring to as back as to ten years. The survey was mainly for assessing pesticide awareness and management practices. This is very important because the country depends on agriculture, and the majority of the population depends on agriculture (Pauw & Thurlow, 2011). If respondents (pesticide users) are not employing best management practices when working with pesticides, there could be consequences such as pesticide contamination in consumable food, feeds, and also in water and the

environment. These contamination due to negligence would kill non-target organisms. Simulation modeling could be used to inform them about the potential consequences of not following the best management practices (i.e. effects of pesticides on non-target organisms). Simulation modeling could also be used to inform pesticide users the effects of pesticides on non-target organisms from practical differences when both following and not following best management practices.

Survey was responded by 157 people. These are the only people who were willing to take part in the survey. With this number of respondents, result was not generalized to the true population of the country. However, results obtained from the survey are very important and cannot simply be ignored because here we are dealing with toxic chemicals (i.e. pesticides). This is because misuse of pesticides by one person could potentially affect the lives of many other people and non-target organisms. Ninety-four respondents (59.87%) reported that agriculture was one of their main activities (AMA) while 63 respondents (40.13%) agriculture was not one of their main activities (ANMA).

Respondents were 121 males and 36 females. Level of education of all respondents combined was university education (142 respondents), advanced diploma (7 respondents), diploma (5 respondents), secondary education (2 respondents), and primary education (1 respondent). Level of education of those agriculture is one of their main activities was university education (86 respondents), advanced diploma (4 respondents), diploma (1 respondent), secondary education (2 respondents), and primary education (1 respondent). Level of education of those agriculture is not one of their main activities was university education (56 respondents), advanced diploma (3 respondents), diploma (4 respondents), and none for both secondary education and primary education.

Respondents by region-wise were from Dar es Salaam (36.31%), Arusha (19.11%), Tanga (7.01%), and Morogoro (5.10%). In fact, these are agricultural dominated regions (Figure 4.2). Respondents from other regions combine were 32.47% of the total respondents. The average age of all respondents combined is 39.31 ± 0.86 years. Maximum age of all respondents combined was 65 years and the minimum was 23 years while the standard deviation of their age was 10.73 years. The average age of those agriculture is one of their main activities is 40.46 ± 1.17 years.

Maximum age of those agriculture is one of their main activities is 64 years and the minimum was 24 years while the standard deviation of their age is 11.33 years. The average age of those agriculture is not one of their main activities is 37.56 ± 1.21 years. Maximum age of those agriculture is not one of their main activities was 65 years and the minimum was 23 years while the standard deviation of their age is 9.57 years.

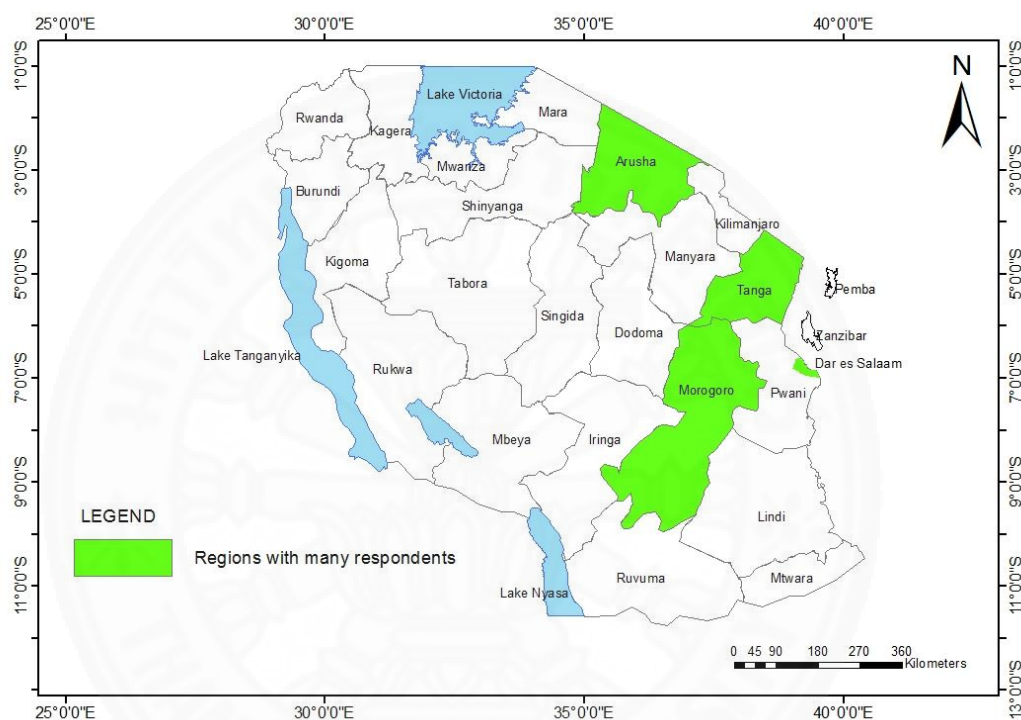


Figure 4.2 Map of Tanzania showing regions (Felix & Sharp, 2016).

The study found that 86.17% of those agriculture is one of their main activities and 79.37% of those agriculture is not one of their main activities, for the first time, had heard about general information related to the usage of pesticide and their associated risks more than 10 years ago (Figure 4.3). This shows that most of the respondents were aware of the existence of pesticides for a quite some time. However, none of those agriculture is one of their main activities and 3.17% of those agriculture is not one of their main activities had heard about general information related to the usage of pesticide and their associated risks within the past 2 years.

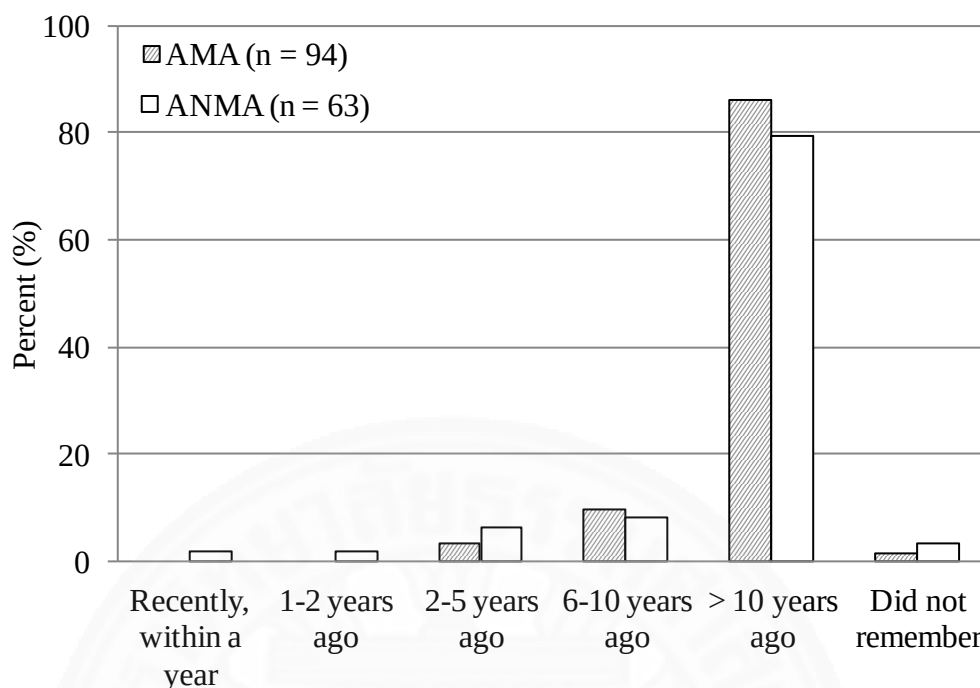


Figure 4.3 Respondents who had heard general information related to the usage of pesticide and their associated risks (for the first time).

The study assessed if respondents were getting general information on regular basis about pesticides through various means such as training, newspapers, televisions, radios, etc. The study found that 56.38% of those agriculture is one of their main activities and 38.10% of those agriculture is not one of their main activities had heard some general information about pesticides recently within a year while 21.28% of those agriculture is one of their main activities and 26.98% of those agriculture is not one of their main activities had heard some general information about pesticides more than 10 years ago (Figure 4.4). This suggests that recently, there has been some sought of awareness programs regarding the use of pesticides in the country.

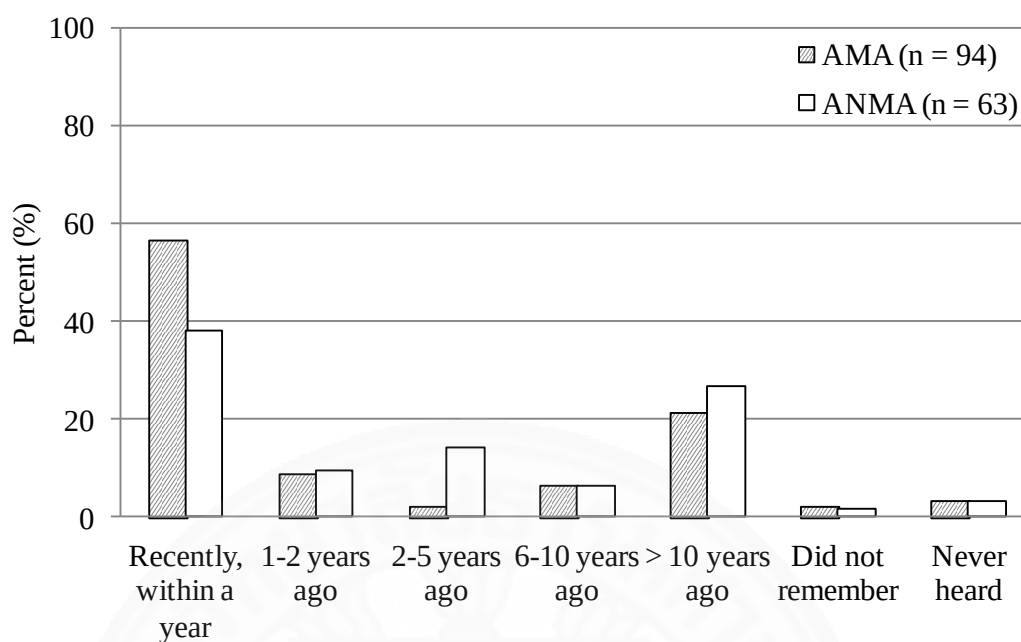


Figure 4.4 Respondents who had heard general information about pesticides (on regular basis) through various means such as training, newspapers, televisions, and radios.

The study assessed the frequency of using pesticides among respondents. The study found that 39.36% of those agriculture is one of their main activities and 9.52% of those agriculture is not one of their main activities used pesticides often, while 52.13% of those agriculture is one of their main activities and 52.38% of those agriculture is not one of their main activities used pesticides few times (occasionally). Also the study found that 7.45% of those agriculture is one of their main activities and 30.16% of those agriculture is not one of their main activities did not use pesticides at all (Figure 4.5).

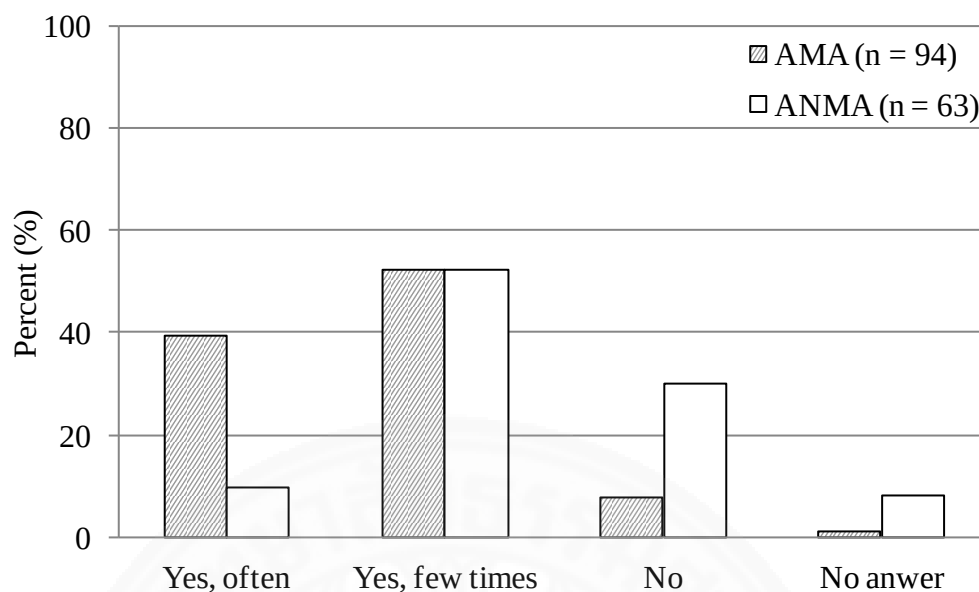


Figure 4.5 Frequency of using pesticides among respondents.

The study assessed whether respondents used to wear protective gears (in the past years) when working with pesticides. The study found that only 15.96% of those agriculture is one of their main activities and 14.29% of those agriculture is not one of their main activities used all required protective gears while working with pesticides. Results reveal that 39.36% of those agriculture is one of their main activities and 25.40% of those agriculture is not one of their main activities did not wear any protective gear when working with pesticides (Figure 4.6); a practice which has negative consequences on their health. This is very dangerous because unsafe practices (Kaewboonchoo et al., 2015; Raksanam et al., 2012) such as not using protective gears when spraying (Wongwichit et al., 2012b) could have serious negative impacts on pesticide users (i.e. the person who is spraying).

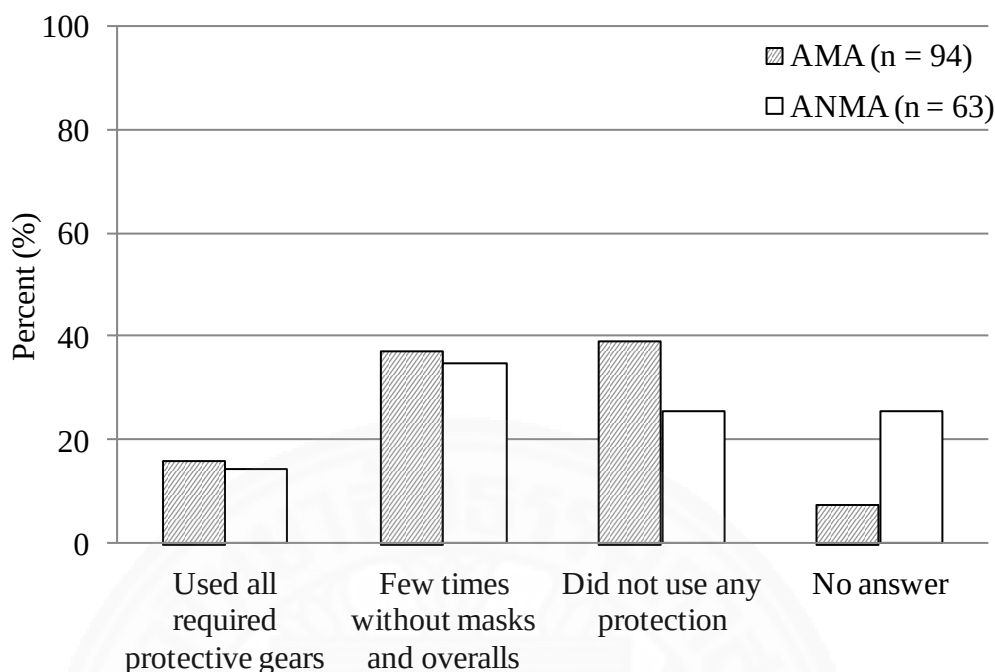


Figure 4.6 Respondents who used protective gears when working with pesticides (in the past).

The study assessed if respondents are presently using protective gears when working with pesticides. Results reveal that 15.96% of those agriculture is one of their main activities and 17.46% of those agriculture is not one of their main activities are currently using all required protective gears, while 34.04% of those agriculture is one of their main activities and 17.46% of those agriculture is not one of their main activities do not use any protection gear when working with pesticides (Figure 4.7). Results further reveal that 84.04% of those agriculture is one of their main activities and 82.54% of those agriculture is not one of their main activities are at risk of being poisoned due to the failure of not following recommended practices when working with pesticides. The none use of protective gears which could be due to either cost or unavailability of protective gears is one of the main concerns that need not be undermined. Results reveal that more of those agriculture is one of their main activities are currently not using protective gears when working with pesticides. Under normal circumstances, it is expected that those agriculture is one of their main activities could be more aware of the importance of using protective gears through meetings conducted by agricultural associations than those agriculture is not one of their main activities who engage in agricultural activities occasionally and more often

are not members of agricultural associations, yet they are not using any protection. However, Figure 4.6 and Figure 4.7 further reveal that there is a reduction by 5.32% (of those agriculture is one of their main activities) and 7.94% (of those agriculture is not one of their main activities) in the number of respondents who did not use any protective gear in the past but are currently using protective gears. This means that more respondents are currently using protective gears when comparing their past and present pesticide management practices.

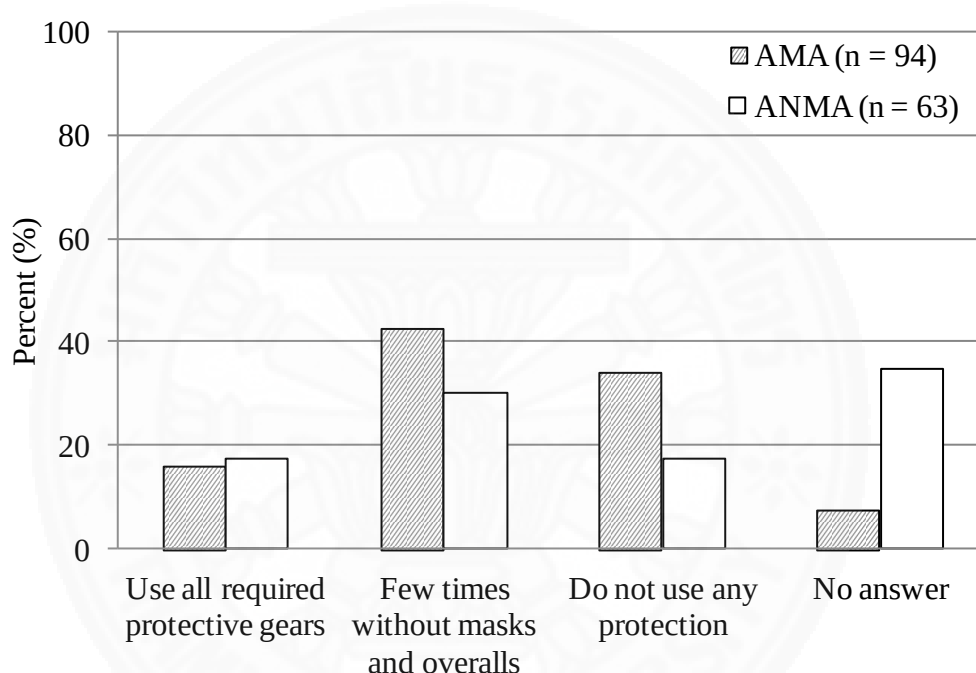


Figure 4.7 Respondents who use protective gears when working with pesticides (presently).

The study found that 63.83% of those agriculture is one of their main activities and 98.41% of those agriculture is not one of their main activities only washed their skins but did not seek medical help, while 36.17% of those agriculture is one of their main activities and 1.59% of those agriculture is not one of their main activities both washed their skins (body) and sought medical help when accidentally contacted pesticides (Figure 4.8). Figure 4.8 further reveal that more of those agriculture is not one of their main activities only washed pesticides from their skins, but more of those agriculture is one of their main activities both washed and sought medical help.

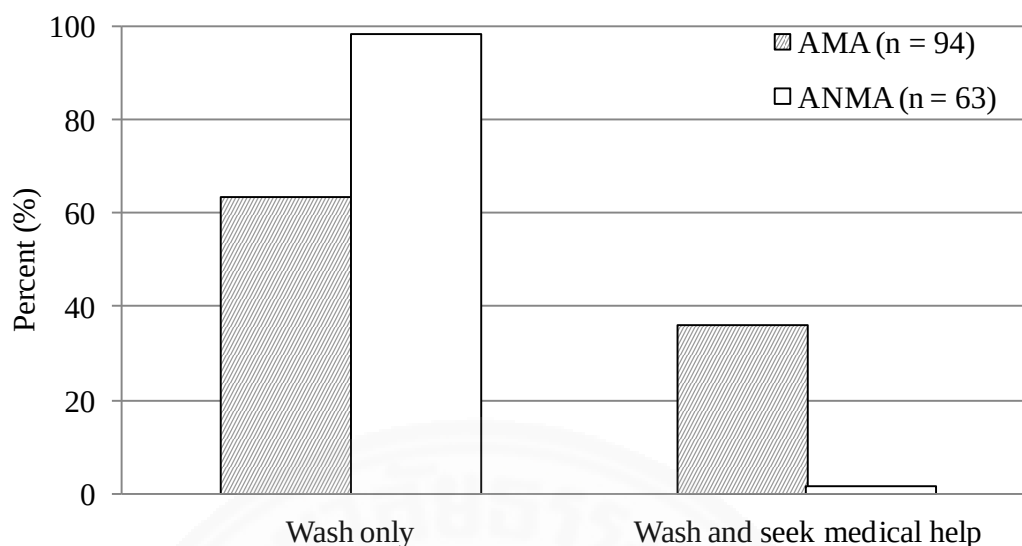


Figure 4.8 Respondents who washed and sought medical help after accidentally contacted pesticides on their skins (body).

The study found that 97.87% of those agriculture is one of their main activities and 42.86% of those agriculture is not one of their main activities only washed their eyes but did not seek medical help, while 2.13% of those agriculture is one of their main activities and 57.14% of those agriculture is not one of their main activities both washed their eyes and sought medical help when accidentally contacted pesticides (Figure 4.9). Figure 4.9 further reveal that more of those agriculture is one of their main activities only washed pesticides from their eyes, but more of those agriculture is not one of their main activities both washed and sought medical help.

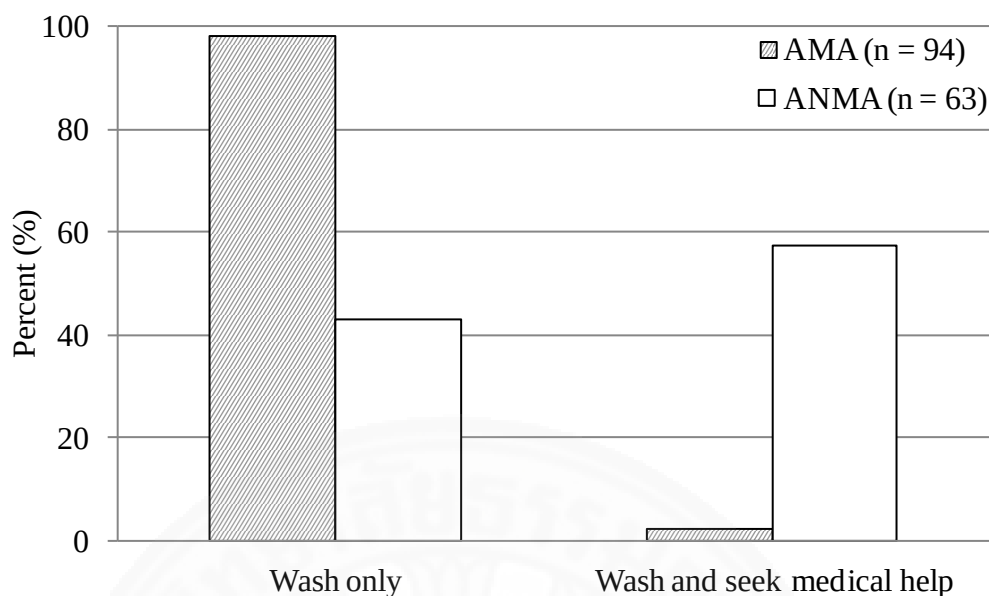


Figure 4.9 Respondents who washed and sought medical help after accidentally contacted pesticides in their eyes.

Although the survey of the present study was carried out online and most of the respondents have university education, the findings in Figure 4.8 and Figure 4.9 agree with Lekei et al. (2014b). Lekei et al. (2014b) interviewed 121 head of households in Arumeru district in Arusha region, northern Tanzania with most of the interviewee having received primary education (basic education). Lekei et al. (2014b) assessed pesticide handling practices such as storage and the use of protective gears, pesticide disposal and calibration of instruments prior to pesticide application. The study found that 93% of the farmers had been poisoned in the past. Also, 79% of the farmers who had been poisoned in the past did not seek proper medical help. The level of education in Lekei et al. (2014b) and this study suggest that level of education might not influence knowledge on pesticide management and practices. Furthermore, Lekei et al. (2014b) suggested that there is a need for improving pesticide labeling and establishment of integrated pest management.

Results of Figure 4.8 and Figure 4.9 also agree with Ngowi et al. (2007a) and Lekei et al. (2014a) that many victims of pesticide poisoning, mostly in rural areas, often remain at home without seeking proper medical help until the poisoning subdue by itself. Lekei et al. (2014a) reported that in most cases, organophosphates (WHO class I and II) are the main poisoning agents with suicide being very significant

(27.8% of all cases of the retrospective study), while accidental cases were 26.4% of all cases. However, Lekei et al. (2014a) found that 41% of all acute pesticide poisoning were unknown.

In the present study, respondents were asked for their opinions as to what they think if it is important for pesticide users to use protective gears when working with pesticides. Only 1.0% of those agriculture is one of their main activities responded that it is very important while 3.17% of those agriculture is not one of their main activities responded that it is very important (Figure 4.10).

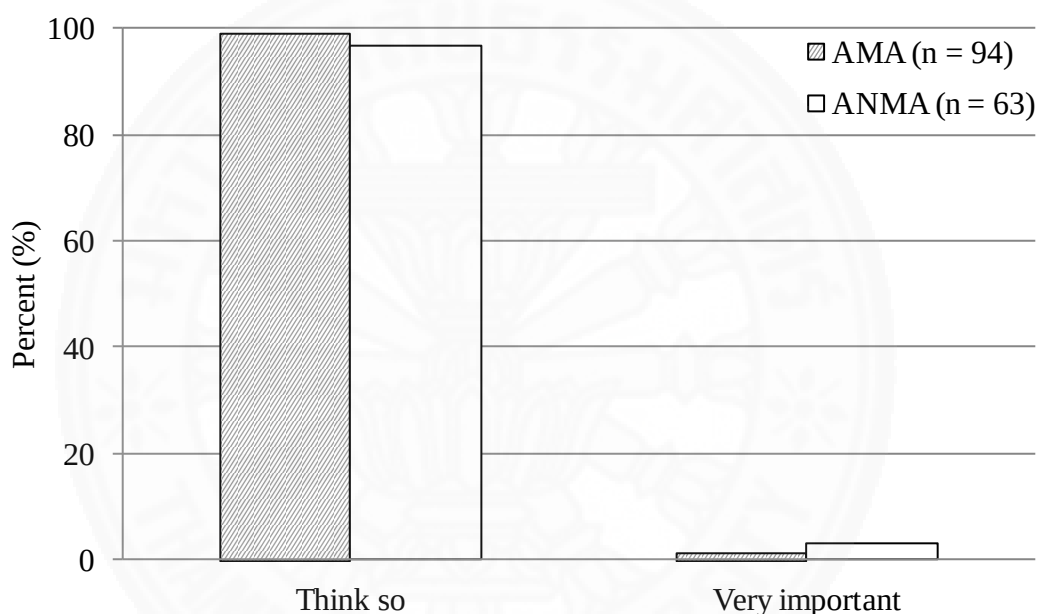


Figure 4.10 Respondent's opinion on importance for pesticide users to use protective gears when working with pesticides.

The study assessed the disposal practices of pesticide empty containers among respondents. Only 10.64% of those agriculture is one of their main activities and 11.11% of those agriculture is not one of their main activities would dispose empty containers in a privately-owned special disposal locations as instructed or else would keep empty containers in secured areas (Figure 4.11). Also, 70.21% of those agriculture is one of their main activities and 66.67% of those agriculture is not one of their main activities would dispose empty containers in the soil either in a landfill or by burying underground. This increases the chances of any remained pesticides in the containers to be found in the soils and the environment. This practice is dangerous

because while in the environment, pesticides could be transported from one point to another (Takahashi, 2003) thus there is a chance to be found in consumable food and groundwater (Abbassy et al., 1999; Kookana et al., 1995).

The study also found that 13.83% of those agriculture is one of their main activities and 11.11% of those agriculture is not one of their main activities would dispose pesticides by burning (Figure 4.11). This is dangerous because the burning of pesticide empty containers or pesticide treated biomass oxidizes nitrogen, sulphur, chlorine and phosphorous fragments to toxic compounds that when inhaled could kill humans (Chen et al., 2012). In addition, inefficient burning could yield polycyclic aromatic hydrocarbons (PAHs) and semi-volatile organics and unburnt parental residues (Kearney & Roberts, 1998). Kearney & Roberts (1998) further demonstrated in an open-air burning experiment that 0.02 g of Thimet (phorate) was released to the atmosphere while the solid residue was 0.05g from each bag. An empty 22.7 kg bag of Thimet (phorate) granules contain on average 4.8 g of formulated material (one gram active ingredient). Therefore, yearly accumulation of such residues would certainly yield large quantities of residues.

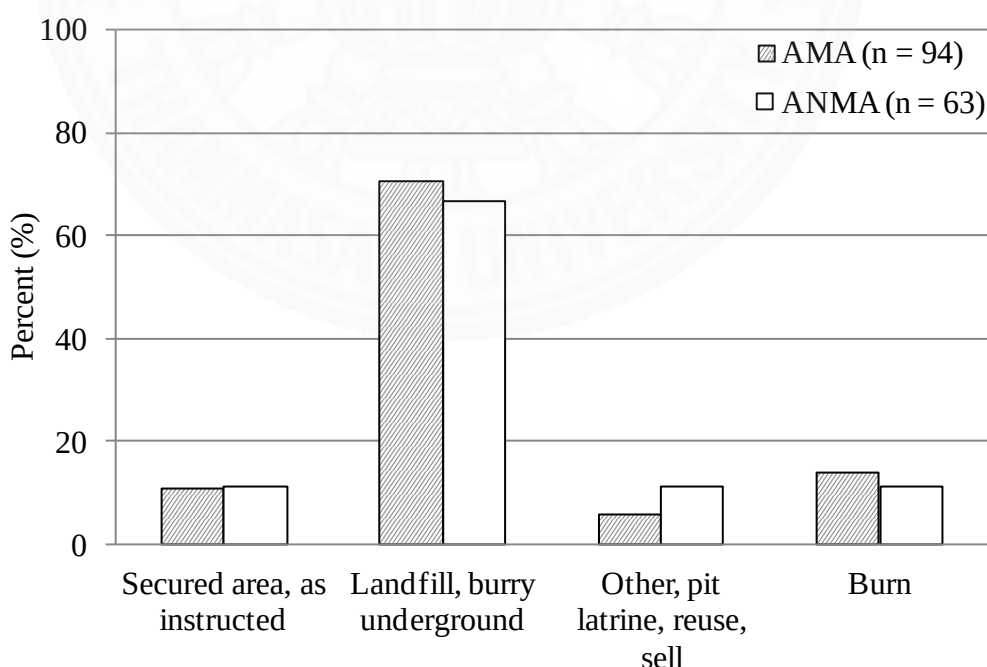


Figure 4.11 Disposal of pesticide empty containers.

The study assessed disposal practices of obsolete pesticides among

respondents. Only 15.96% of those agriculture is one of their main activities and 15.87% of those agriculture is not one of their main activities would dispose obsolete pesticides in a privately-owned special disposal locations as instructed or else would keep obsolete pesticides in secured areas. Also, 81.91% of those agriculture is one of their main activities and 73.02% of those agriculture is not one of their main activities would dispose obsolete pesticides in the soil; either in a landfill or by burying underground (Figure 4.12). This practice is dangerous because it increases the chances of obsolete pesticides to be found in the environment. While in the environment, pesticides are subjected to translocation (Takahashi, 2003) which increases their chance to be found in consumable food and groundwater (Abbassy et al., 1999; Kookana et al., 1995). The study found that less than 2.13% of those agriculture is one of their main activities and less than 11.11% of those agriculture is not one of their main activities did not know where to dispose obsolete pesticides. Those who did not know where to dispose obsolete pesticides could potentially dispose it anywhere. This also increases the possibility of pesticides to be found in non-target sites. However, to the best of my knowledge, the country has no system to collect and properly dispose off empty containers and obsolete pesticides.

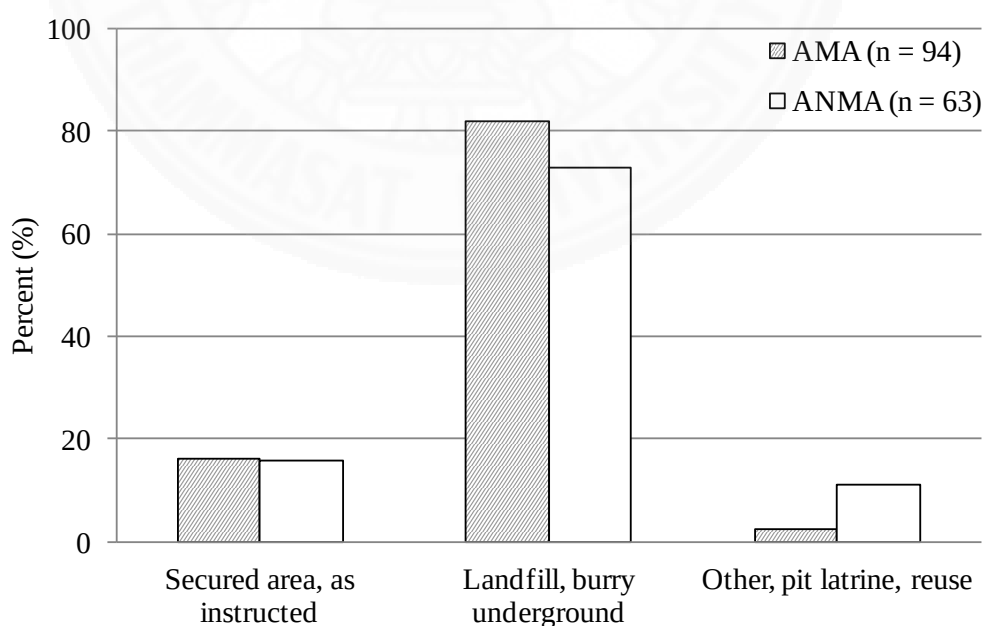


Figure 4.12 Disposal of obsolete pesticides.

The study assessed the storage of pesticide inventories among respondents. Storage of pesticide inventories is one of the important aspects of pesticide management. The study found that 86.17% of those agriculture is one of their main activities and 93.65% of those agriculture is not one of their main activities would store pesticide inventories either in a special designated storage area, keep outside the house, or other places such as garage. Also, 13.83% of those agriculture is one of their main activities and 6.35% of those agriculture is not one of their main activities would store pesticide inventories in other areas such as kitchen, bedroom, and above the cupboard; areas within the reach of children (Figure 4.13). Storage of pesticides in the kitchen, bedroom, and above the cupboard increases the chances of home pesticide exposure to children (Lu et al., 2000). This practice is dangerous because Bailey et al. (2015) found that early childhood exposure to pesticides increased the risk of childhood leukemia. It could be true that those who can keep pesticide inventories in the kitchen, bedroom, and above the cupboard either did not read instructions on the labels that all pesticides must be kept away from the reach of children or they just ignore. Also, keeping pesticide inventories in such areas could contaminate consumables in the houses.

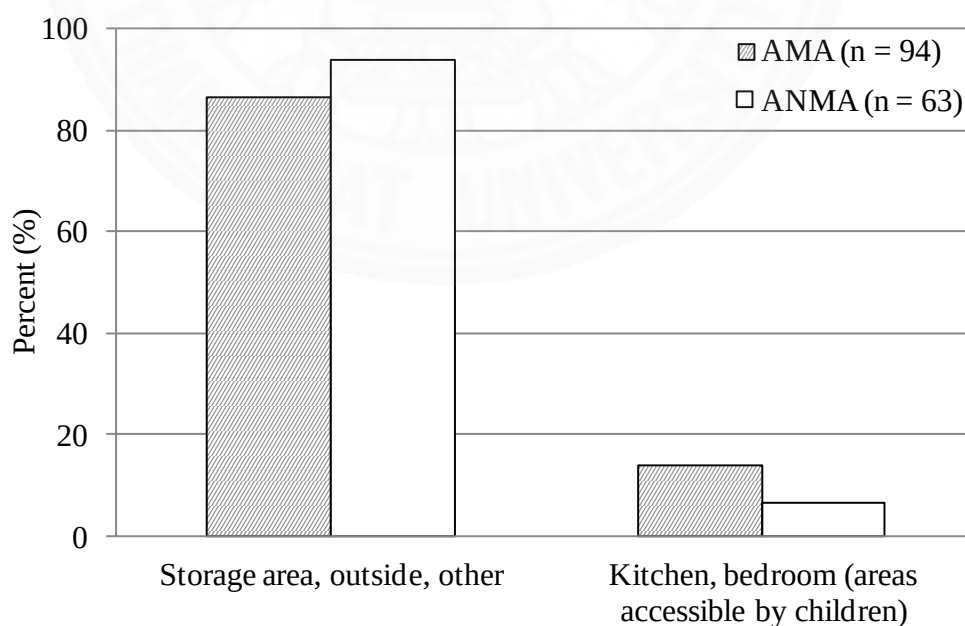


Figure 4.13 Storage of pesticide inventory.

The study assessed whether respondents were able to identify negative health effects associated with the use of pesticides such as death, skin cancer, cancer, loss of vision, etc. The study found that 85.11% of those agriculture is one of their main activities and 74.60% of those agriculture is not one of their main activities were able to indentify negative health effects associated with the use of pesticides, while 14.89% of those agriculture is one of their main activities and 25.40% of those agriculture is not one of their main activities were not able to indentify negative health effects associated with the use of pesticides (Figure 4.14). Although Figure 4.14 shows that most respondents were aware of the ill-effects of pesticides, still, some respondents exercised a potential risk to their health and the environment from the way they (would) handle pesticides. There is no doubt that pesticides solve apparent problems such as hunger by reducing the loss of agricultural produce by eliminating pests and that companies involved make money (profits), still it should be noted that at the end of the spectrum they create undesirable problems and untreatable diseases to farmers, non-farmers, suppliers, consumers, environment, and the ecosystem at large.

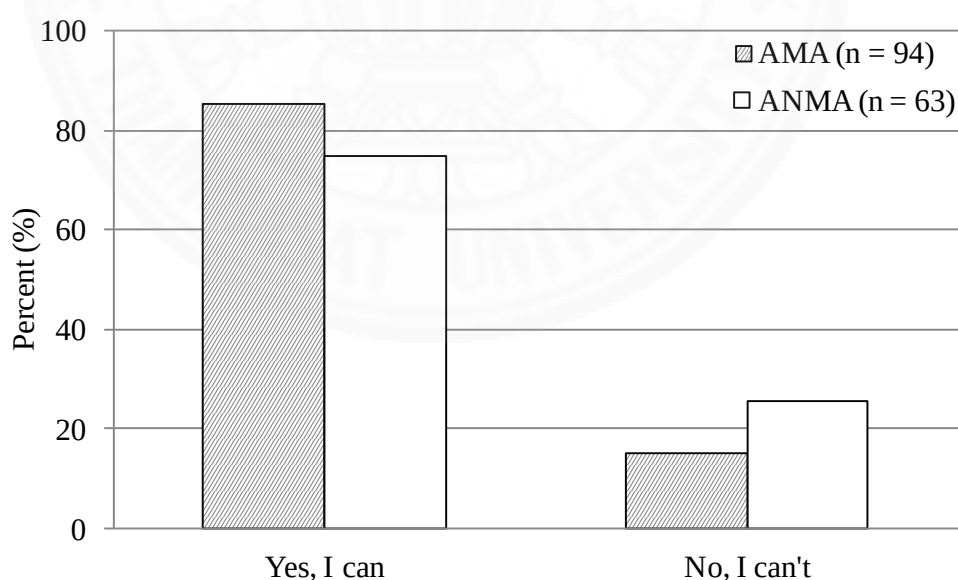


Figure 4.14 Identification of negative health effects associated with the use of pesticides.

The study assessed whether respondents would consume food kept in a

container which was used to keep pesticides. Results reveal that 85.11% of those agriculture is one of their main activities and 93.65% of those agriculture is not one of their main activities would not consume food kept in a container which was used to keep pesticides (Figure 4.15). This implies that majority of the respondents are aware that pesticides are toxic chemicals, that is why they would not eat food kept in containers which was used to keep pesticides.

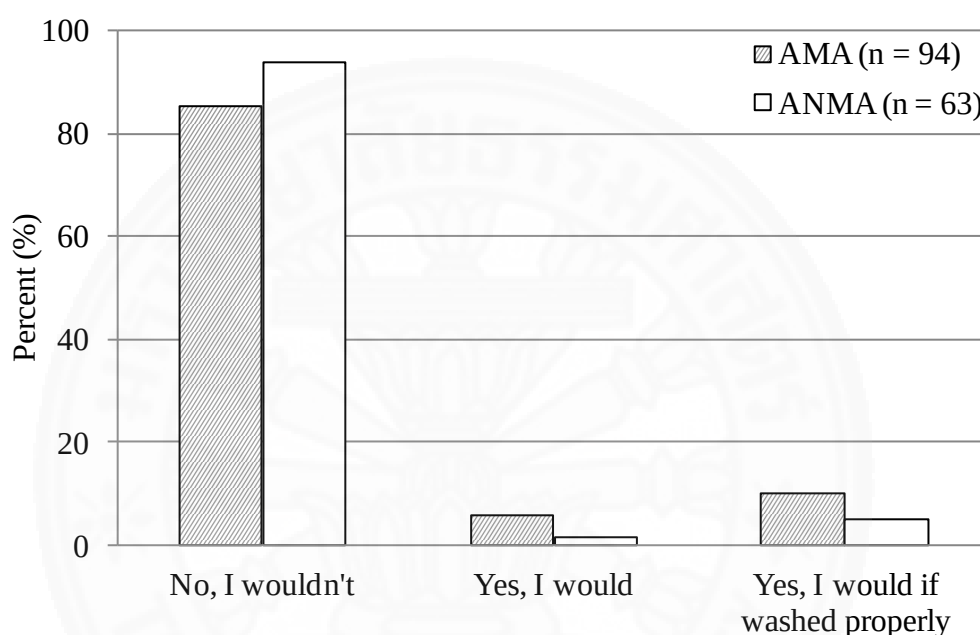


Figure 4.15 Respondents willingness to consume food kept in a container which was used to keep pesticides.

The study also assessed whether respondents would drink water kept in a container which was used to keep pesticides. Results reveal that 84.04% of those agriculture is one of their main activities and 95.24% of those agriculture is not one of their main activities would not drink water kept in a container which was used to keep pesticides (Figure 4.16). This implies that majority of the respondents are aware that pesticides are toxic chemicals, that is why they would not drink water kept in containers which was used to keep pesticides. Figure 4.16 further reveal that 15.96% of those agriculture is one of their main activities and 4.76% of those agriculture is not one of their main activities would drink water kept in a container which was used to keep pesticides. Results suggest that respondents were aware what pesticides could cause harm to them and the environment, yet they handled it poorly. Mishandling

could however be due to negligence.

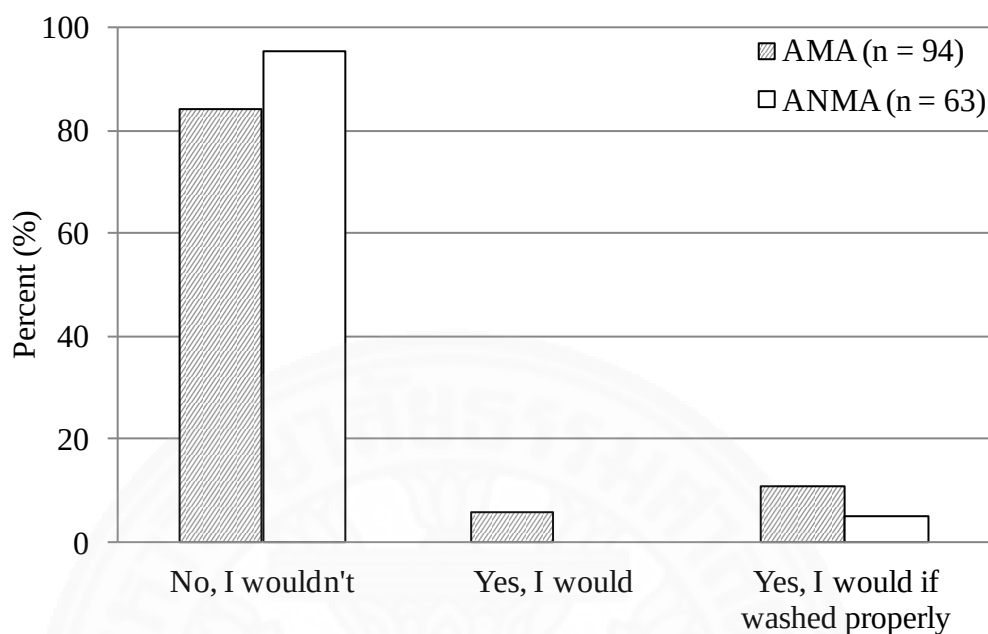


Figure 4.16 Respondents willingness to drink water kept in a container which was used to keep pesticides.

The study found that most respondents were aware that pesticides are toxic chemicals and thus must be handled with care (Figure 4.17). However, few of them responded that pesticides are non-toxic, can be kept anywhere in the house, and anyone (including children) can have access to pesticides (Figure 4.17). This may seem a small number to be ignored but bad pesticide practices from this small number of respondents could cause severe consequence. Therefore, more tailor-made motivation and awareness programs are needed to further reduce this number.

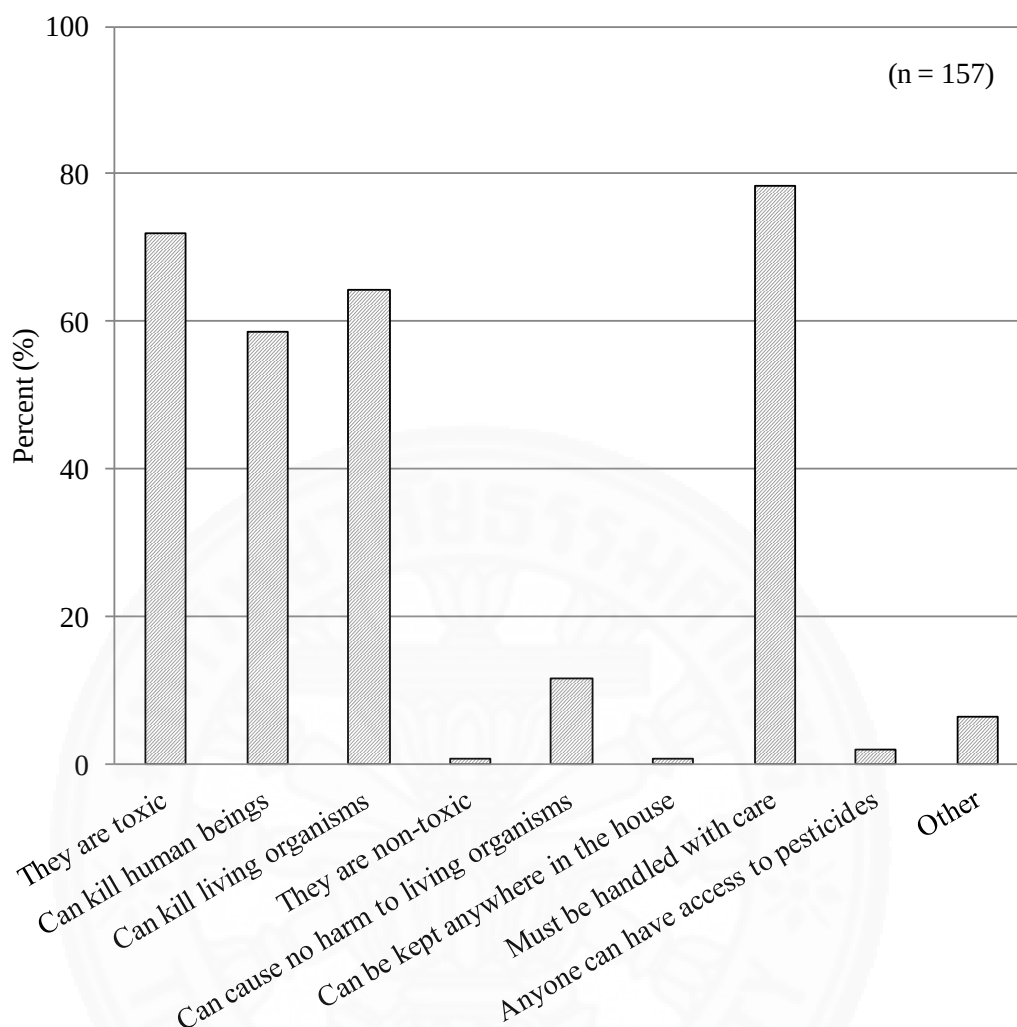


Figure 4.17 Respondents opinion about pesticides.

The study assessed if respondents were worried about fresh fruits, fresh vegetables, and tea leaves possibly being contaminated with pesticides when buying them. The study found that most of the respondents were worried about fresh fruits, fresh vegetables, and tea leaves possibly being contaminated with pesticides when buying them (Figure 4.18). However, the study found that 40.10% of the respondents believe that proper washing with clean water completely remove pesticides from the fruits and vegetables while 59.90% did not believe that proper washing with clean water completely remove pesticides from the fruits and vegetables.

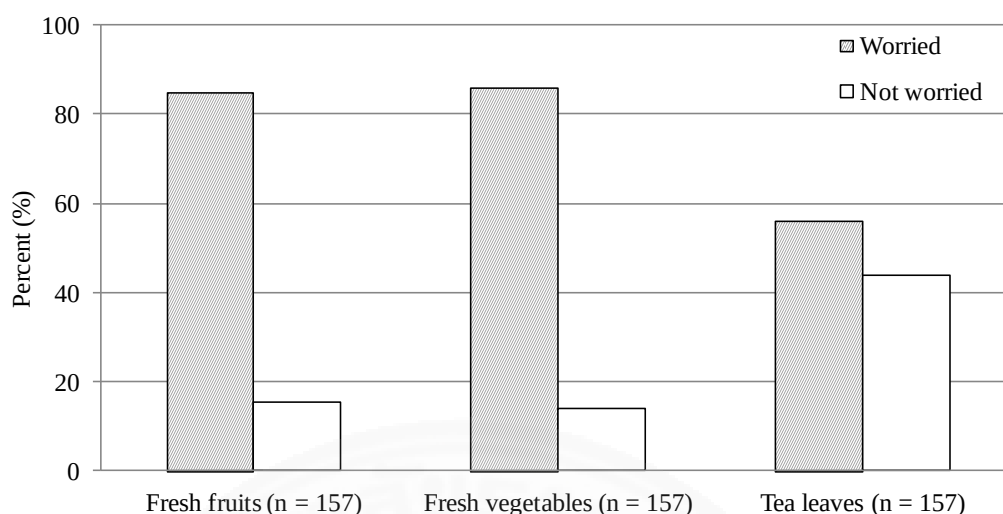


Figure 4.18 Respondents who were worried about fresh fruits, fresh vegetables, and tea leaves possibly being contaminated with pesticides.

The study found that 80.90% of the respondents were able to identify the adverse or negative health effects associated with pesticides while 19.10% were not able. Skin cancer was the most identified negative health effect associated with pesticides followed by death and loss of vision (Figure 4.19). The study found that 43.30% of the respondents have experienced pesticide-related poisoning symptoms such as either severe headaches or skin burns right after a pesticide application, while 5.70% of the respondents were not able to identify such poisoning symptoms as well as 3.80% of the respondents have never been to those areas where pesticide was recently applied (Figure 4.20). The study found that 47.10% of the respondents have experienced pesticide-related poisoning symptoms right after coming in contact with pesticides (Figure 4.21). The most experienced symptoms were headache (59.30%), skin burn (47.70%), dizziness (41.90%), chest discomfort (24.40%), irritation of the nose and throat (54.70%), and eye irritation (57.00%) (Figure 4.22). The study found that some of those who were poisoned by pesticides in the last two years, spent between 0.2 and 4,550 US\$ for medical services and treatments.

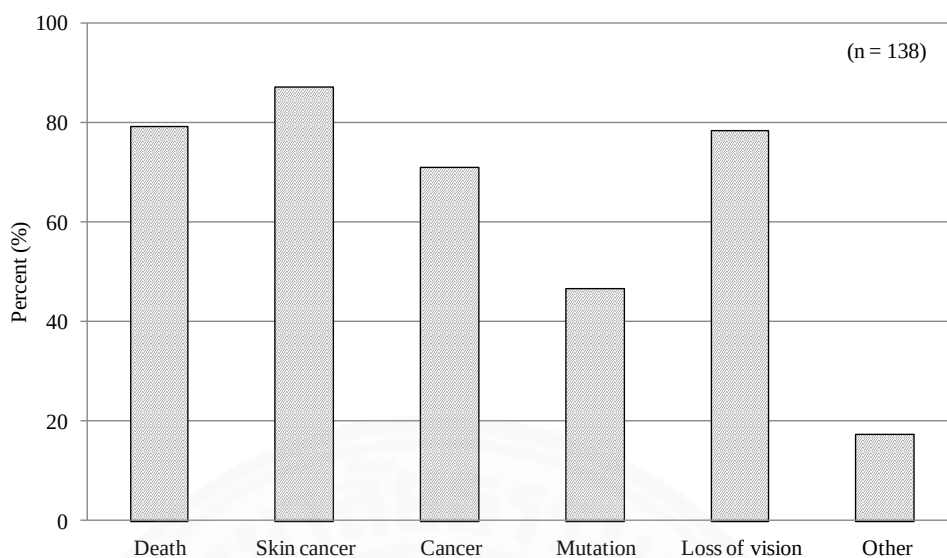


Figure 4.19 Negative health effects identified by the respondents.

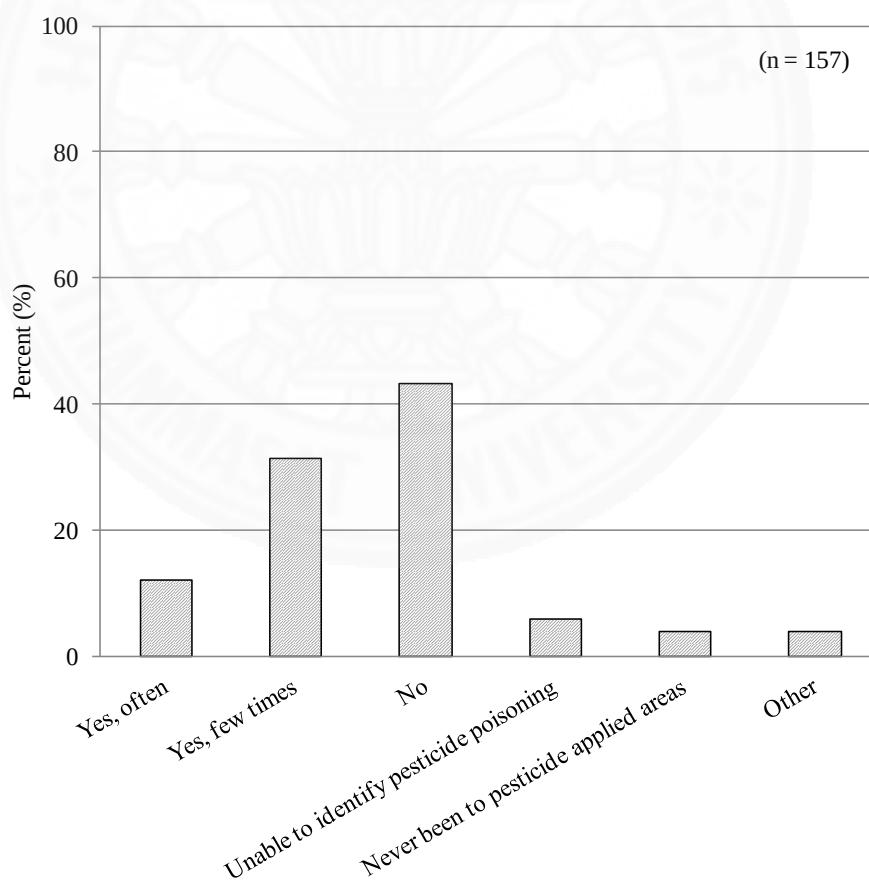


Figure 4.20 Pesticide-related poisoning symptoms experienced by the respondents (either severe headaches or skin burns) right after pesticide application.

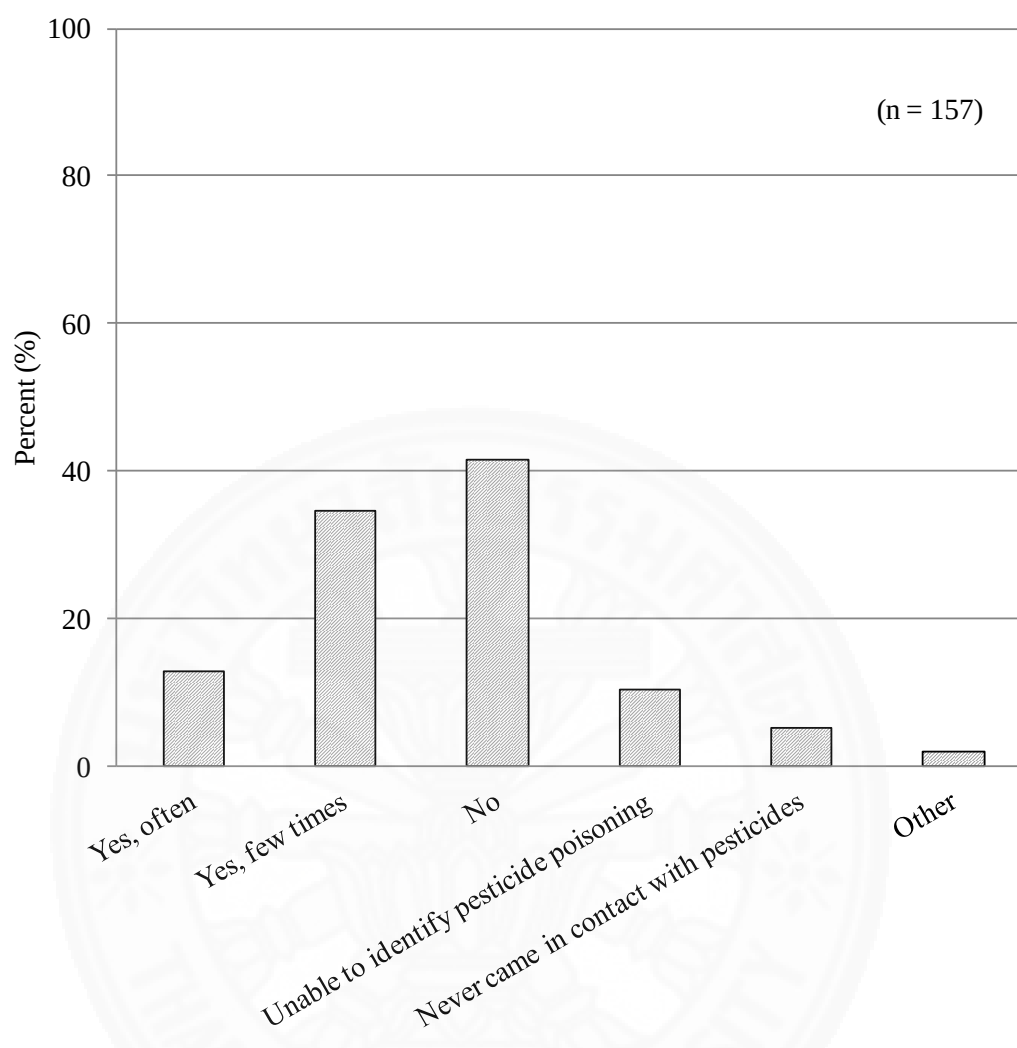


Figure 4.21 Pesticide-related poisoning symptoms experienced by the respondents right after coming in contact with pesticides.

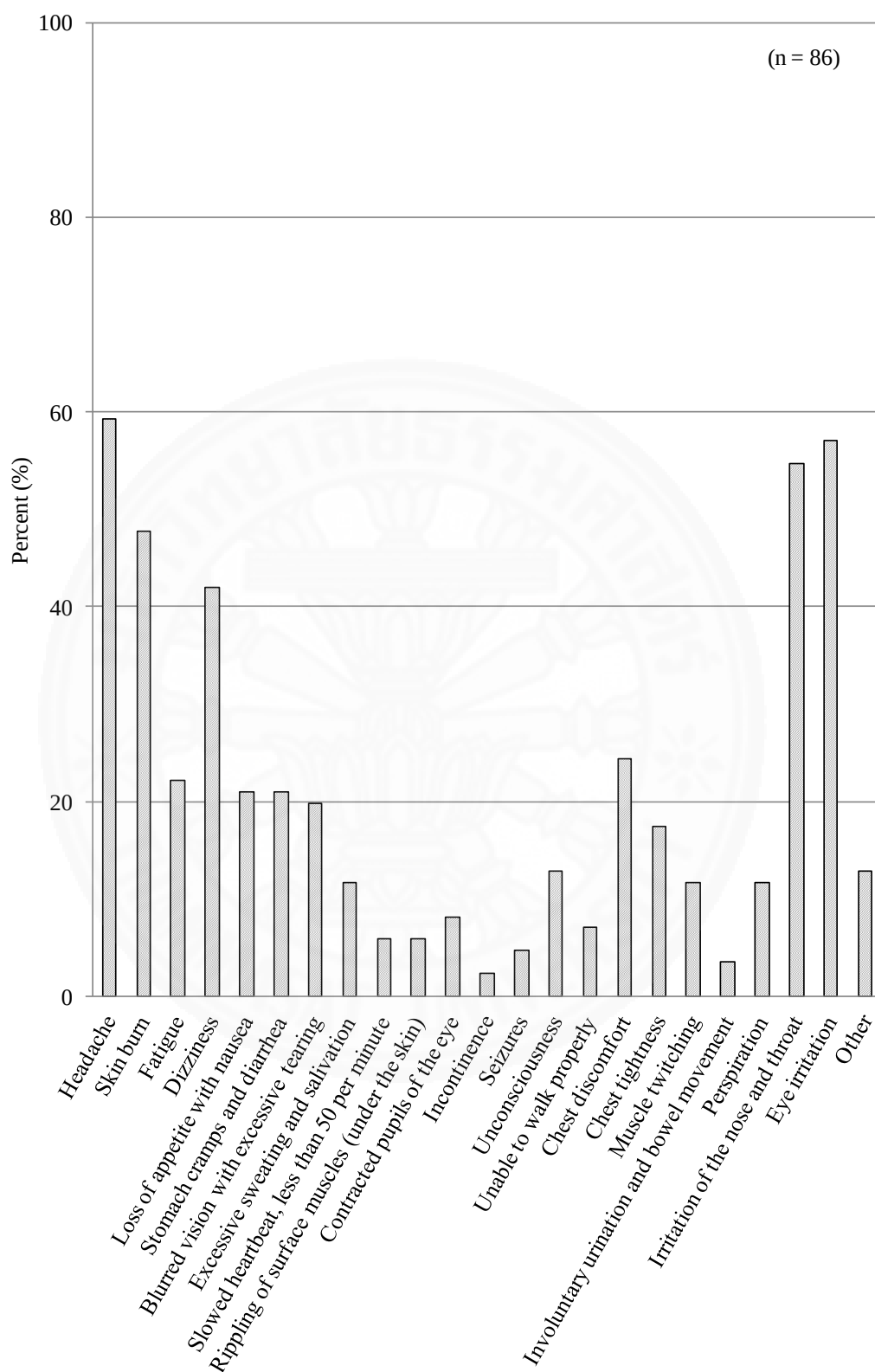


Figure 4.22 Pesticide-related poisoning symptoms experienced by the respondents.

Results reveal that despite that majority of respondents had University education, still, their pesticide management practices was poor which could severely deteriorate the environmental health and quality, affect human health, and have negative impacts on non-target organisms. It could be true that lack of tailor-made motivational programs on proper use of pesticides could have a significant contribution in the results of the present study. These tailor-made motivational programs could be in form of regular training offered to pesticide users from time to time on best pesticide management practices and good agricultural practices. There could also be regular tailor-made motivational programs aired on public television and radio stations. It is expected that this would somehow reduce negligence; consequently it would improve the quality of human health, environmental health and the ecosystem, as these respondents could change from bad to good pesticide practices. Also, results of the simulation modeling of fate and transport of glyphosate and its effects on frogs could be used to motivate pesticide users on the importance of employing best pesticide management practices.

4.2 Modeling

Glyphosate which is a non-selective herbicide was simulated with the application dose of 1.5 kg a.i ha⁻¹ (Waramit & Suwanketnikom, 1999) on a one hundred meters by one hundred meters agricultural field, using a conventional boom sprayer. Two scenarios were modeled of which the study assumed that the agricultural field was surrounded by a width of no spray zone (buffer zone) of thirty meters and zero meters on each side to protect spray drifts from reaching non-target sites (Figure 1.1). The study assumed that one side of the field was located parallel with the direction of wind. The number of passes required to cover the sprayed area exposed to wind drift was eighty passes. Simulation was performed for two hundred fifty days. Weather data used in the simulation were for both year 2012 and 2013.

4.2.1 Weather variability

Results show that when considering the entire duration of the simulation (250 days); there were no statistical significant difference on variability of temperatures ($p > 0.05$) and rainfalls ($p > 0.05$) between year 2012 and 2013. When considering the first 50 days of the simulation; there was no statistical significant difference on

variability of temperatures ($p > 0.05$) and rainfalls ($p > 0.05$) between year 2012 and 2013 ($p < 0.05$). However, distributions of temperatures (Figure 4.23) and rainfalls (Figure 4.24) have shown profound impact on the simulated results of fate and transport processes of glyphosate. Further analysis of the weather data reveals that within the first thirty days after the first application of glyphosate, year 2013 had significant rainfall events than year 2012.

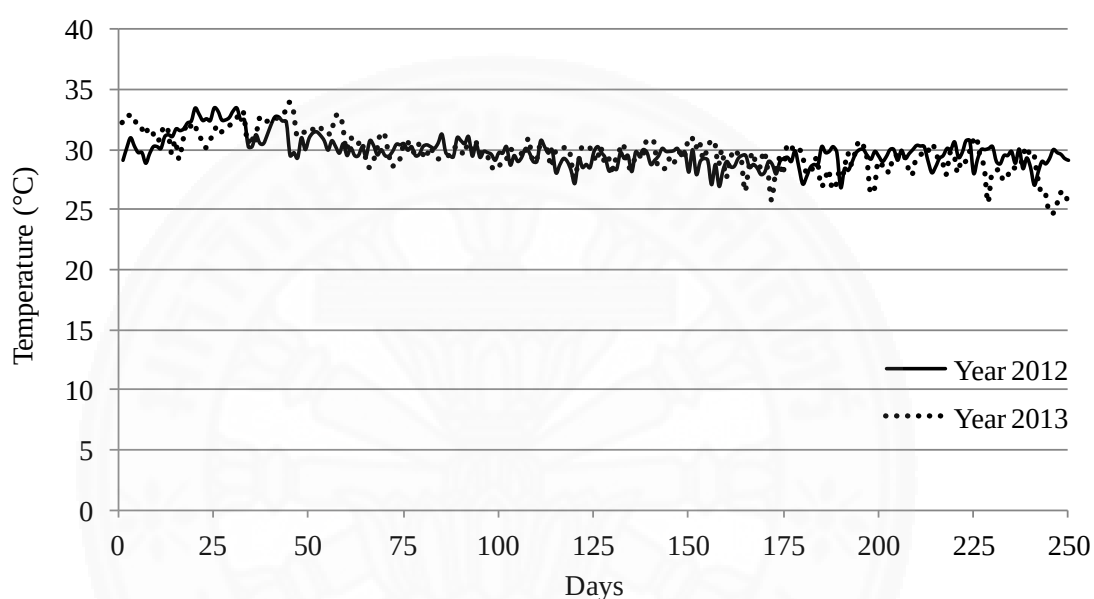


Figure 4.23 Distribution of temperatures

(Day 0 means before simulation starts and day 1, 2, 3, ... 250 are simulation days starting from the first of April).

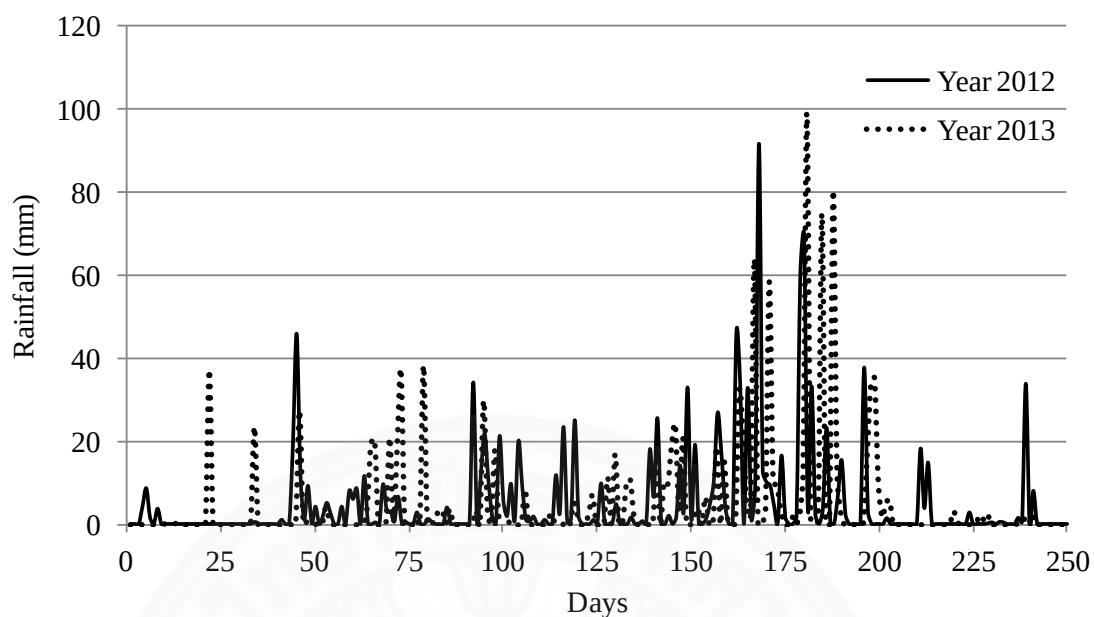


Figure 4.24 Distribution of rainfalls.

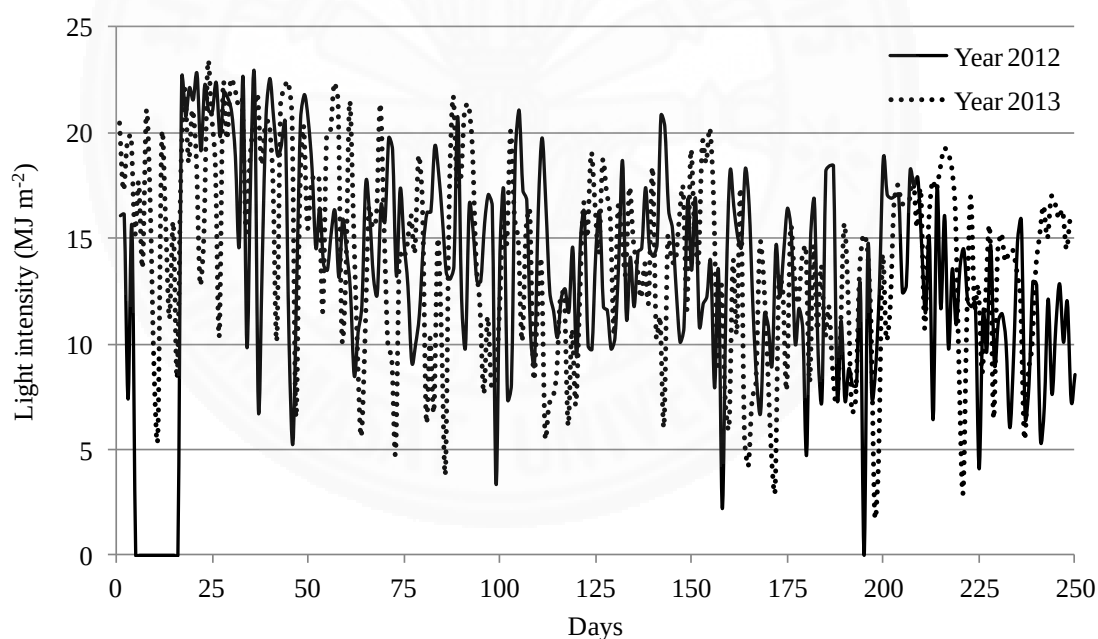


Figure 4.25 Distribution of light intensities.

4.2.2 Primary distribution

The study assumed that after spraying, three main distributions occurs as follows; some amount of glyphosate was carried away by wind from the target site (as spray drifts), some amount of glyphosate was deposited on the crops (as crop/leaf deposition), and some amount of glyphosate was deposited on the soils (as soil

deposition) (van Zelm et al., 2014). Also, the study assumed that the amount of glyphosate applied was equal to the sum of the main distributions. This distribution depends on weather conditions during spraying, application technique used, and the crop stage at the time of spraying (Margni et al., 2002). Due to the lack of data, the study assumed that there were no further processes that removed glyphosate that was drifted to the air through wind drifts (spray drifts). Also, when studying the fate and transport of glyphosate and its impact on population dynamics of frogs, the study assumed that the amount of glyphosate that crossed the agricultural field via wind drift remained in the air.

The study assumed that manual weeding was done before sowing of maize seeds. Manual weeding was followed by two incidents of glyphosate application on day fourteen and twenty-eight after sowing of maize seeds each at $1.5 \text{ kg a.i ha}^{-1}$ (Waramit & Suwanketnikom, 1999). Glyphosate losses due to wind drift was modeled following Rautmann et al. (1999) and Holterman & van de Zande (2003).

Results show that spray drifts for first application (on day fourteen) and second application (on day twenty-eight) at a distance of thirty meters downwind from the edge of the agricultural field was each $6.42 \times 10^{-8} \text{ kg a.i ha}^{-1}$ for both year 2012 and 2013. This was about $4.28 \times 10^{-6}\%$ of the glyphosate applied for each application. Spray drifts for first application (on day fourteen) and second application (on day twenty-eight) at a distance of zero meters downwind from the edge of the agricultural field was each $0.013 \text{ kg a.i ha}^{-1}$ for both year 2012 and 2013. This was about 0.89% of the glyphosate applied for each application for year 2012 and about 0.87% of the glyphosate applied for each application for year 2013 (Table 4.1). Spray drifts when using buffer zone width of thirty meters were very little compared to when no buffer zone width was used (zero meters) mainly because buffer zones around the agricultural fields prevents the spray drifts from crossing the field.

Results of the spray drifts is in agreement with Maybank et al. (1978) who reported that herbicide spray drifts for ground-rig application could be from less than 0.5% to 8% of the dose applied, depending on speed of the wind and nozzle type. It should be noted that there are many other factors that could affect spray drifts, e.g. Nordby & Skuterud (1974) found that increase in boom height from forty to eighty centimeters increased spray drifts from 1% to 3-2% of the dose applied. Also, Nordby

& Skuterud (1974) found that increase in pressure from 2-5 bar to 10 bar increased spray drifts from 1-4% to 2-9%, and increase in wind speed from 1-5 m s⁻¹ to 4-0 m s⁻¹ increased spray drifts from 1-4% to 2-9%.

Maize crop development (Figure 4.26 and Figure 4.27) was modeled by simulating crop stages namely leaf development, tillering, stem elongation, and senescence using degree day values i.e. heat unit accumulations following Dijkhuis (1956). Results show that when using buffer zone width of thirty meters, the amount of glyphosate deposited on crop/leaf surfaces was 50.13% and 70.00% of the glyphosate applied for first and second application respectively for year 2012 and 50.71% and 70.00% of the glyphosate applied for first and second application respectively for year 2013. Also, when using buffer zone width of zero meters, the amount of glyphosate deposited on the crop/leaf surfaces was 49.68% and 69.38% of the glyphosate applied for first and second application respectively for year 2012 and 50.27% and 69.40% of the glyphosate applied for first and second application respectively for year 2013 (Table 4.1).

The present finding is not in agreement with Audsley et al. (1997) that 5% of the applied dose could be deposited on the crops. This difference could be due to the reason that during simulation, the present study used deposition fractions following Linders et al. (2000), with the lowest deposition fraction of 25% during leaf development stage of crops and the highest deposition fraction of 90% during senescence stage of crops (Table A.11). Also, it could be due to the difference in the crop phenology characteristics and type of application method used.

For both scenarios, results show that amount of glyphosate deposited on the crop/leaf surfaces for second application was 1.40 and 1.38 times higher than the first application for both year 2012 and 2013 because at that time of application crops usually have more number of leaves. This means that the deposition fraction is usually higher during the second application than during the first application.

By applying the principle of mass balance (i.e. soil deposition = dose applied – (spray drifts + crop/leaf deposition), when using buffer zone width of thirty meters, the amount of glyphosate deposited in the topsoil were 49.87% and 30.00% of the glyphosate applied for first and second application respectively for year 2012 and 49.29% and 30.00% of the glyphosate applied for first and second application

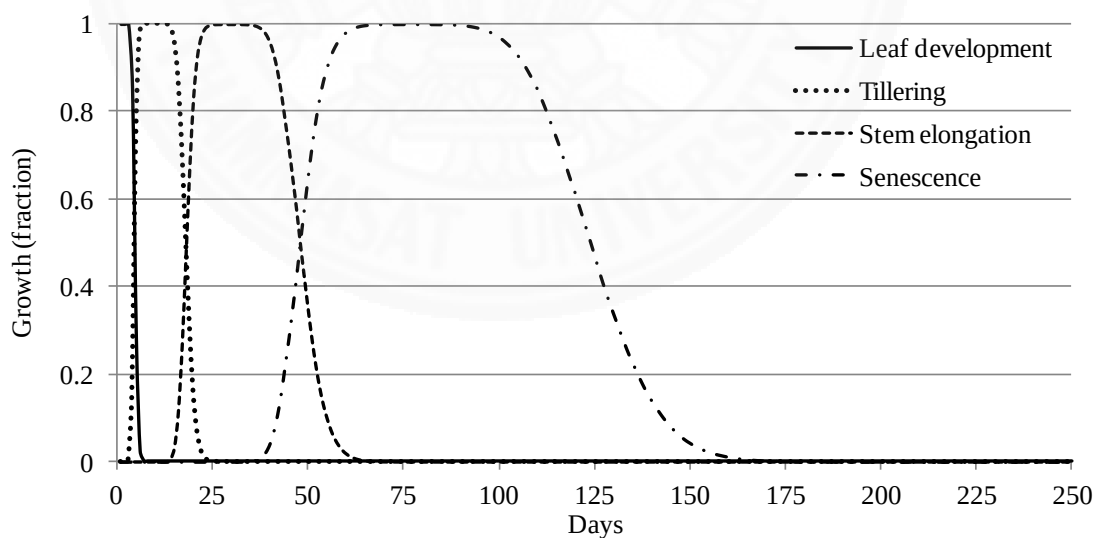
respectively for year 2013. Also, when using buffer zone width of zero meters, the amount of glyphosate deposited in the topsoil were 49.43% and 29.73% of the glyphosate applied for first and second application respectively for year 2012 and 48.87% and 29.73% of the glyphosate applied for first and second application respectively for year 2013 (Table 4.1). The finding is not in agreement with Audsley et al. (1997) that 85% of the applied dose could be deposited on soils after the application. This could possibly be due to the reason that during the simulation, the present study used deposition fractions following Linders et al. (2000). Also, it could be due to other reasons such as difference in weather conditions.

For both scenarios, results show that amount of glyphosate deposited in the topsoil for second application was 0.6 times lower than the first application for both year 2012 and 2013. This was due to the more number of leaves during the second application that caused more glyphosate deposition on the crop/leaf surfaces.

Nevertheless, it appears that different studies have assumed or used different emissions fractions (values) of pesticides after application, e.g. Panichelli et al. (2009) assumed that pesticide inputs are entirely emissions to the soil. This is why Rosenbaum et al. (2015) suggested that there could be inconsistencies in the emissions of pesticide inventories among studies because there is no standard guideline (consensus) as how much on average should be considered as default values for primary distribution of pesticides after the application (spraying) for different conditions and crop types.

Table 4.1 Primary distribution of glyphosate after the application (spraying).

Day of application	Glyphosate applied (kg a.i ha ⁻¹)	Spray drifts (kg a.i ha ⁻¹)	Glyphosate deposited on the crop/leaf surfaces (kg a.i ha ⁻¹)	Glyphosate deposited on the soil (kg a.i ha ⁻¹)	Year
Before sowing	Manual weeding	0	0	0	2012
Before sowing	Manual weeding	0	0	0	2013
With the width of no spray zone (buffer zone) set to thirty meters					
14	1.5	6.42×10^{-8}	0.752	0.748	2012
28	1.5	6.42×10^{-8}	1.050	0.450	2012
14	1.5	6.42×10^{-8}	0.761	0.739	2013
28	1.5	6.42×10^{-8}	1.050	0.450	2013
With the width of no spray zone (buffer zone) set to zero meters					
14	1.5	0.013	0.745	0.741	2012
28	1.5	0.013	1.041	0.446	2012
14	1.5	0.013	0.754	0.733	2013
28	1.5	0.013	1.041	0.446	2013

**Figure 4.26** Maize crop development stages (year 2012).

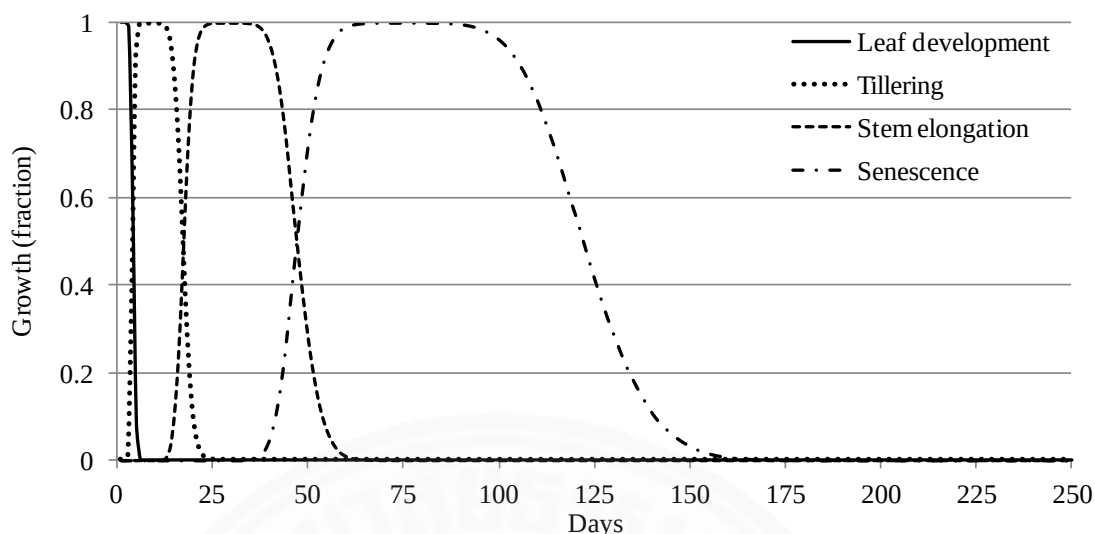


Figure 4.27 Maize crop development stages (year 2013).

4.2.3 Crop/leaf processes

When using buffer zone width of thirty meters, results show that deposition on maize crop/leaf surfaces were $0.752 \text{ kg a.i ha}^{-1}$ and $1.050 \text{ kg a.i ha}^{-1}$ of glyphosate during the first application (day fourteen) and second application (day twenty-eight) respectively for year 2012 and $0.761 \text{ kg a.i ha}^{-1}$ and $1.050 \text{ kg a.i ha}^{-1}$ of glyphosate during the first application and second application respectively for year 2013. Also, when using buffer zone width of zero meters, results show that deposition on maize crop/leaf surfaces were $0.745 \text{ kg a.i ha}^{-1}$ and $1.041 \text{ kg a.i ha}^{-1}$ of glyphosate during the first application and second application respectively for year 2012 and $0.754 \text{ kg a.i ha}^{-1}$ and $1.041 \text{ kg a.i ha}^{-1}$ of glyphosate during the first application and second application respectively for year 2013.

The study assumed that washoff, volatilization, uptake, and photo-degradation were the main competing processes that removed glyphosate from the crop/leaf surfaces (Birkved & Hauschild, 2006; Dijkman et al., 2012). However, transpiration through stomata could be responsible for removing glyphosate from the crop/leaf surfaces (Pilon-Smits, 2005).

4.2.3.1 Washoff

The study assumed that rainfall was responsible for removing glyphosate from the crop/leaf surfaces. The amount of glyphosate washed off from the crop/leaf

surfaces by rainfall was modeled following Smith & Carsel (1984). When using buffer zone width of thirty meters, Figure 4.28 shows that between day fourteen and day fifty of the simulation, year 2012 had highest washoff events on day forty-four (0.20 kg a.i ha⁻¹), forty-five (0.25 kg a.i ha⁻¹), forty-six (0.06 kg a.i ha⁻¹), and forty-eight (0.04 kg a.i ha⁻¹) while year 2013 had highest washoff events on day twenty-two (0.16 kg a.i ha⁻¹), thirty-four (0.24 kg a.i ha⁻¹), and forty-six (0.16 kg a.i ha⁻¹). Results reveal that year 2013 had its first washoff event nineteen days before the first washoff event in year 2012. Also, three washoff events occurred in year 2013 before the first washoff event occurred in year 2012. This had a significant influence in the differences of the results when modeling mortality and survival rates of eggs, tadpoles, and froglets.

When using buffer zone width of zero meters, Figure 4.29 shows that there was no noticeable difference in the amount of glyphosate washed off from the crop/leaf surfaces when comparing with Figure 4.28 mainly because buffer zone width is not one of the factors that affects washoff. However, Figure 4.28 and Figure 4.29 show that the amount of glyphosate washed off from the crop/leaf surfaces is in decreasing trend because washoff is a function of amount of glyphosate present at the time when rainfall events (Figure 4.24) of significant intensities occurred. This is to say that in the early days of simulation, if glyphosate concentration is high, potential washoff amount is expected to be higher at that time and subsequently decreases with time because of the decrease in concentration of glyphosate on crop/leaf surfaces. The amount of glyphosate washed off from the crop/leaf surfaces was added to the topsoil. Results show that total amount of glyphosate removed from the crop/leaf surfaces by washoff was 26.60% and 28.60% of the glyphosate applied (two applications, each at 1.5 kg a.i ha⁻¹) for year 2012 and 2013 respectively when using buffer zone width of thirty meters, and 26.32% and 28.35% for year 2012 and 2013 respectively when using buffer zone width of zero meters.

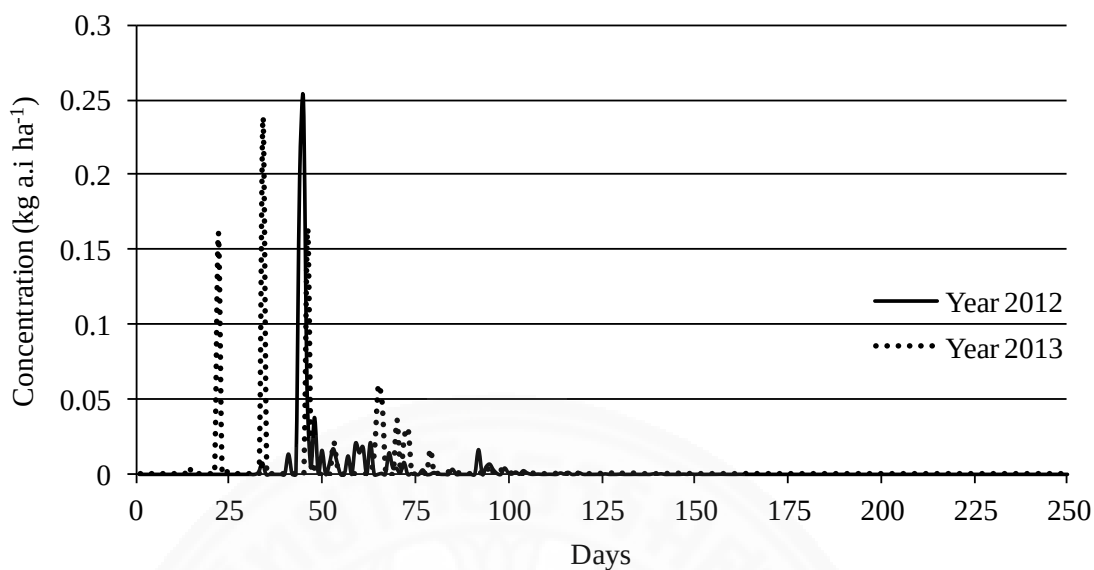


Figure 4.28 Amount of glyphosate removed by washoff from the crop/leaf surfaces (using buffer zone width of thirty meters).

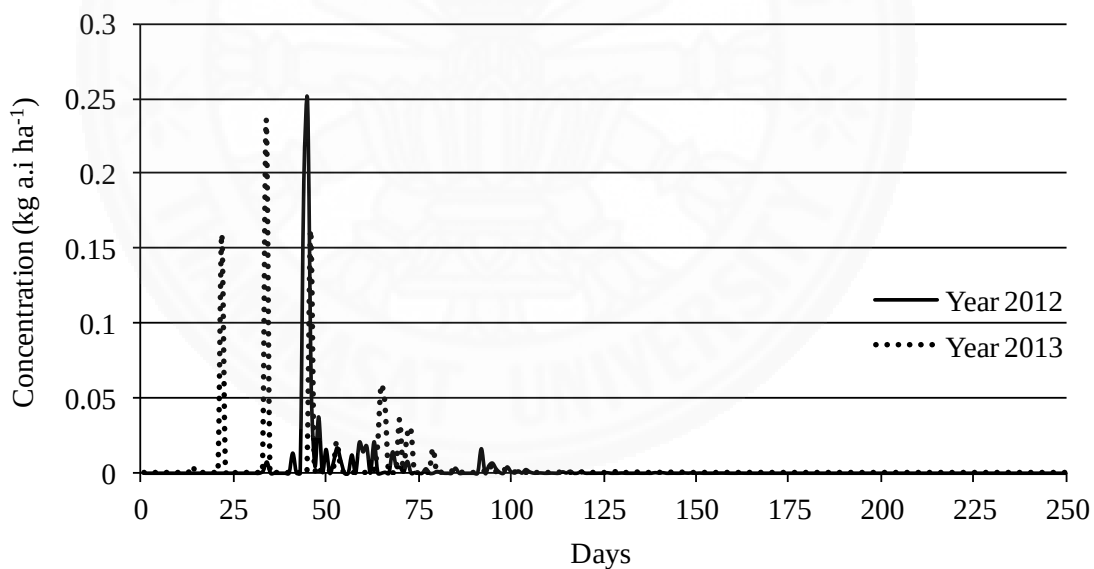


Figure 4.29 Amount of glyphosate removed by washoff from the crop/leaf surfaces (using buffer zone width of zero meters).

4.2.3.2 Volatilization, uptake, and degradation

The rates of glyphosate removal by volatilization from the crop/leaf surfaces were simulated following van Wesenbeeck et al. (2008). Figure 4.30 and Figure 4.31

show that daily volatilization rates were roughly between $8 \times 10^{-4} \text{ kg a.i ha}^{-1} \text{ day}^{-1}$ and $2.5 \times 10^{-3} \text{ kg a.i ha}^{-1} \text{ day}^{-1}$ for the simulated period.

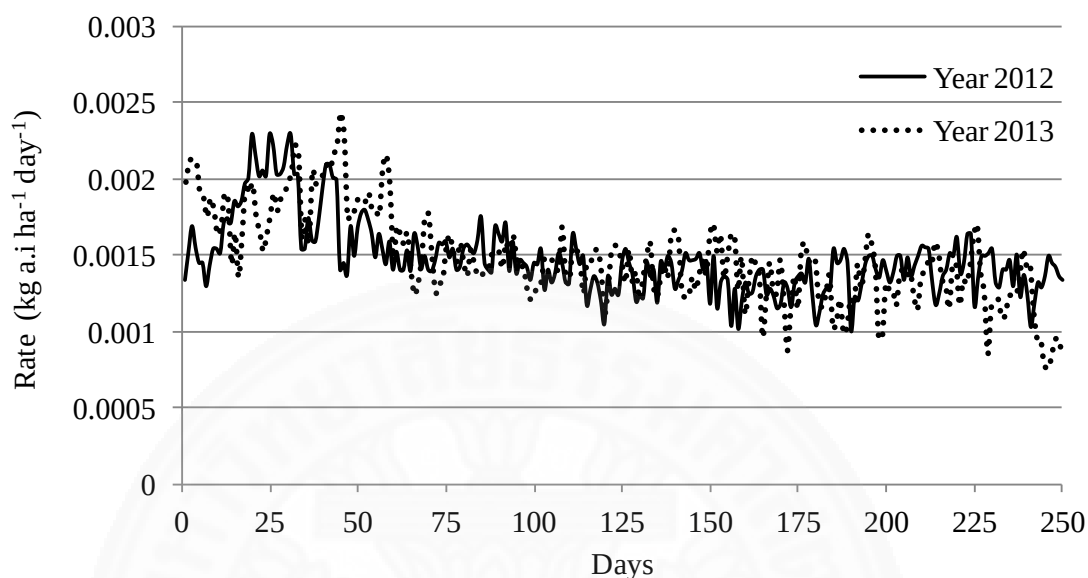


Figure 4.30 Rates of glyphosate removal by volatilization from the crop/leaf surfaces (using buffer zone width of thirty meters).

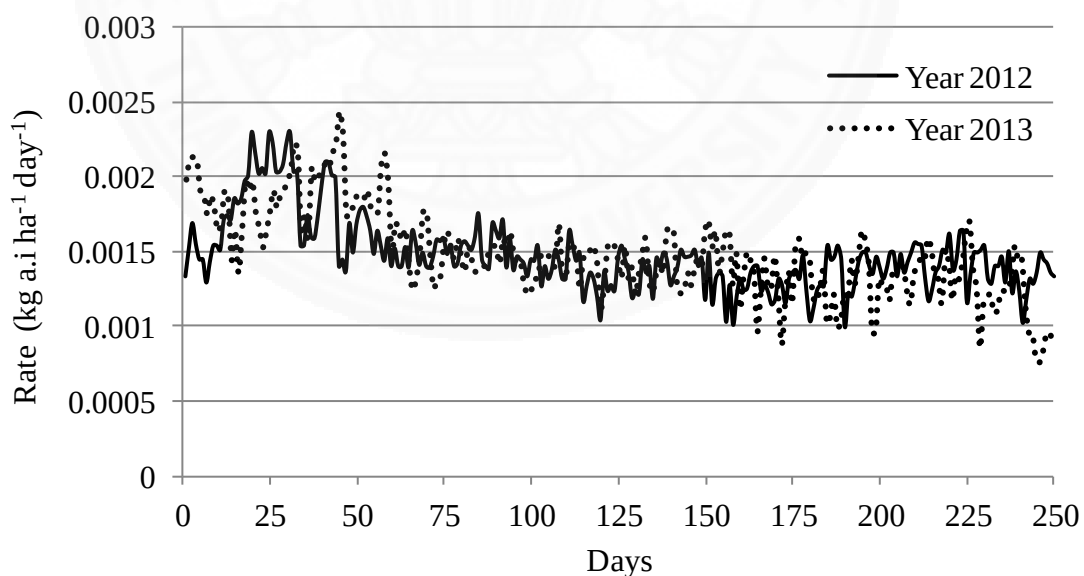


Figure 4.31 Rates of glyphosate removal by volatilization from the crop/leaf surfaces (using buffer zone width of zero meters).

The rate of glyphosate removal by uptake from the crop/leaf surfaces through cuticles was modeled following Baur & Schönherr (1995). Figure 4.32 and Figure

4.33 show that the rates of uptake were roughly between $0.002 \text{ kg a.i ha}^{-1} \text{ day}^{-1}$ and $0.008 \text{ kg a.i ha}^{-1} \text{ day}^{-1}$ for the simulated period. However, the findings are not in agreement with Brecke & Duke (1980) and Green et al. (1992) that the uptake process of glyphosate was slow. Also, the finding is not in agreement with Gottrup et al. (1976) and Wyrill & Burnside (1976) that generally glyphosate is not metabolized by crops/plants. Possible reason to this difference could be in the assumptions made and the approach followed when modeling crop uptake processes.

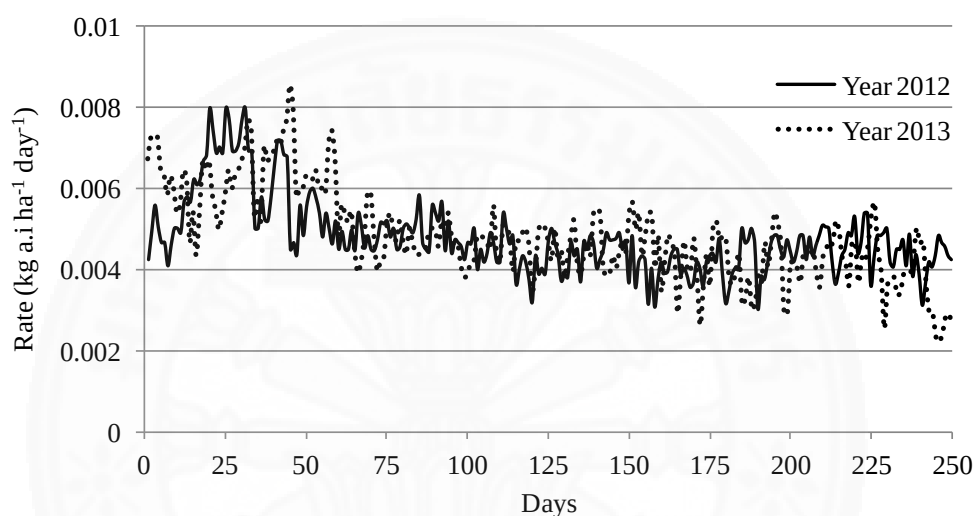


Figure 4.32 Rates of glyphosate removal by uptake from the crop/leaf surfaces (using buffer zone width of thirty meters).

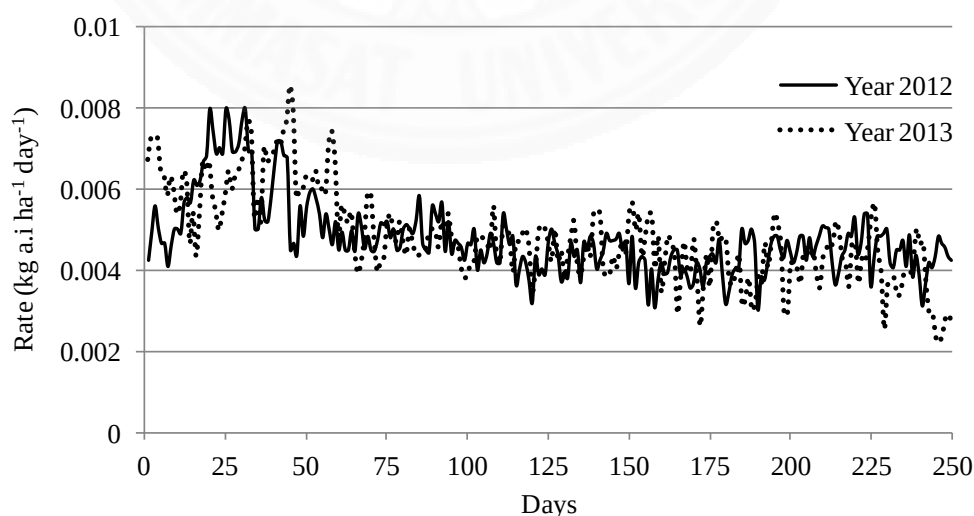


Figure 4.33 Rates of glyphosate removal by uptake from the crop/leaf surfaces (using buffer zone width of zero meters).

The rates of glyphosate removal by degradation from the crop/leaf surfaces were modeled assuming that degradation on crop/leaf surfaces was due to the effect of sunlight (Assalin et al., 2010) and that photo-degradation was the main degradation process following Dijkman et al. (2012). Light intensities were used to simulate concentrations of OH radicals ($\text{OH}\cdot$). Length of the days were influential to the results. The findings (Figure 4.34 and Figure 4.35) are not in agreement with EPA (1993) that that glyphosate was not readily broken down by sunlight. The finding is also not in agreement with Bowmer (1987) who suggested that light had no effect on degradation of glyphosate from crop/leaf surfaces. The difference in the results could be due to the assumptions made and the modeling approach followed (e.g. weather condition is different between tropical and temperate regions). Also, difference in day lengths could be one of the reasons. All these were adjusted in the present study. Results suggest that further research is needed in future studies on parameters that affect photo-degradation.

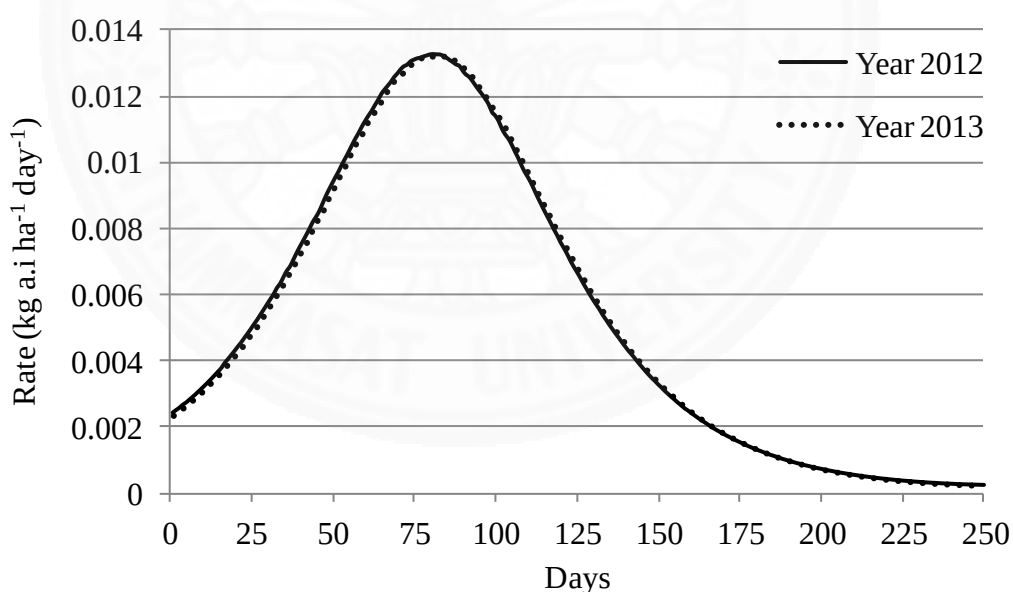


Figure 4.34 Rates of glyphosate removal by photo-degradation from the crop/leaf surfaces (using buffer zone width of thirty meters).

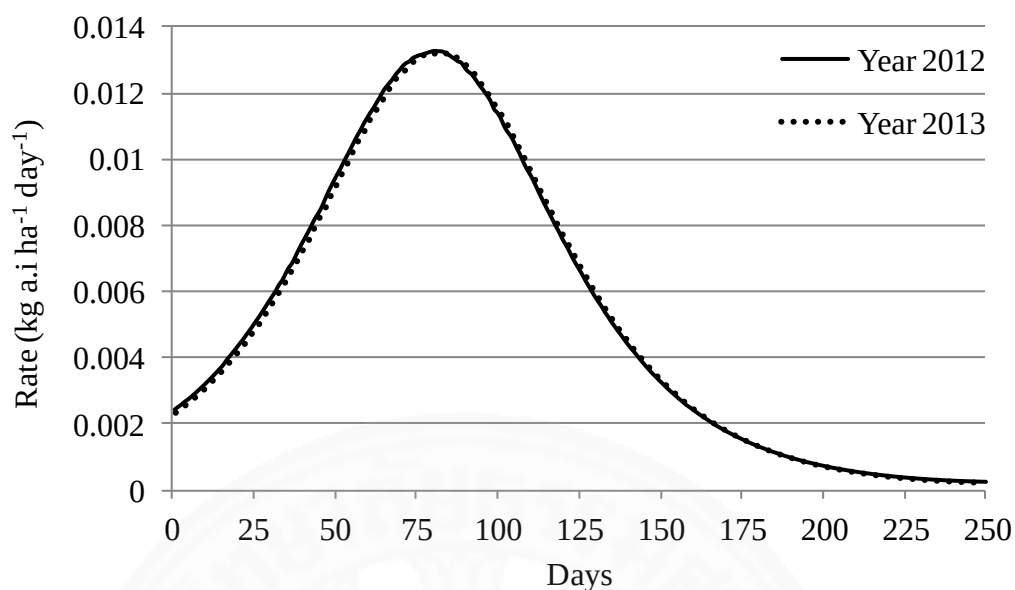


Figure 4.35 Rates of glyphosate removal by photo-degradation from the crop/leaf surfaces (using buffer zone width of zero meters).

When using buffer zone width of thirty meters, Figure 4.36 shows that on day fourteen of the simulation, $0.752 \text{ kg a.i ha}^{-1}$ and $0.761 \text{ kg a.i ha}^{-1}$ of glyphosate were deposited on the crop/leaf surfaces after spraying for year 2012 and 2013 respectively. The study assumed that volatilization, uptake, and photo-degradation processes were the main competing processes that removed glyphosate from the crop/leaf surfaces (Birkved & Hauschild, 2006). On day twenty-seven of the simulation, $0.631 \text{ kg a.i ha}^{-1}$ and $0.404 \text{ kg a.i ha}^{-1}$ of glyphosate were still present on crop/leaf surfaces for year 2012 and 2013 respectively. At that time, glyphosate was more removed from crop/leaf surfaces for year 2013 than 2012. On day twenty-eight, there was a second glyphosate spraying event which added $1.050 \text{ kg a.i ha}^{-1}$ of glyphosate on crop/leaf surfaces for both year 2012 and 2013. The amount of glyphosate deposited on the crop/leaf surfaces on the second application resulted to the increase in the amount of glyphosate on crop/leaf surfaces to $1.672 \text{ kg a.i ha}^{-1}$ and $1.449 \text{ kg a.i ha}^{-1}$ for year 2012 and 2013 respectively. From day twenty-nine to day one hundred, the amount of glyphosate remained on crop/leaf surfaces decreased steadily to $0.018 \text{ kg a.i ha}^{-1}$ and $0.013 \text{ kg a.i ha}^{-1}$ for year 2012 and 2013 respectively. In one hundred forty-eight days after the first application, all glyphosate deposited on the crop/leaf surfaces were removed completely. This suggests that glyphosate does not stay longer on the

crop/leaf surfaces. When using buffer zone width of zero meters (Figure 4.37), there was no noticeable difference in the amount of glyphosate that remained on the crop/leaf surfaces with when buffer zone width of thirty meters was used (Figure 4.36) because buffer zones do not affect processes that occur on the crop/leaf surfaces.

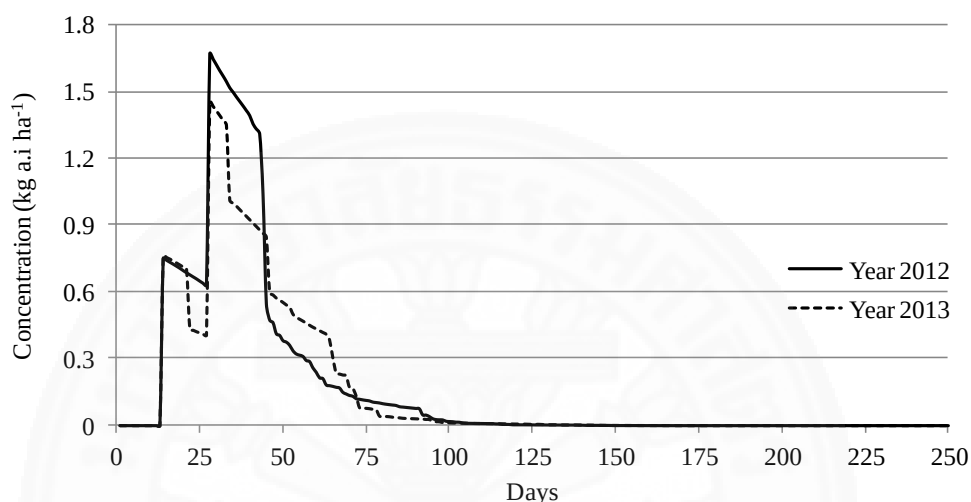


Figure 4.36 Concentration of glyphosate remained on crop/leaf surfaces after washoff, volatilization, uptake, and photo-degradation (using buffer zone width of thirty meters).

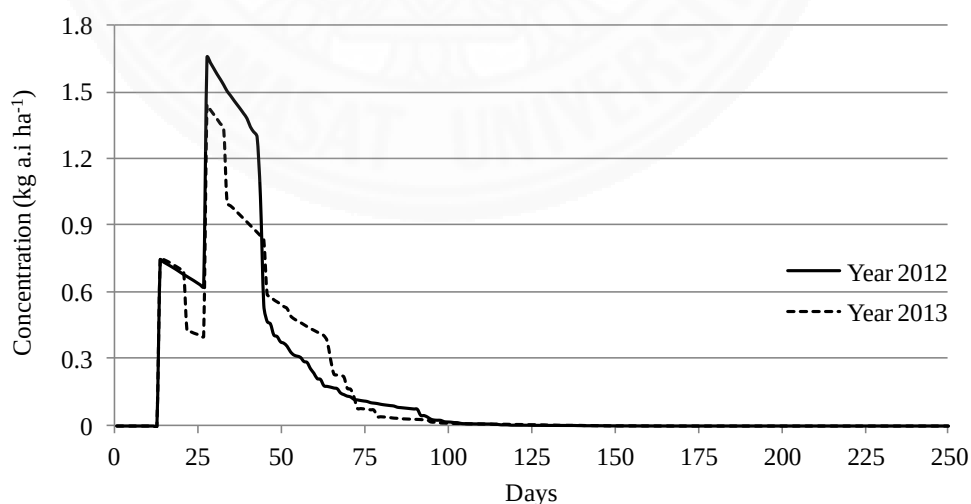


Figure 4.37 Concentration of glyphosate remained on crop/leaf surfaces after washoff, volatilization, uptake, and photo-degradation (using buffer zone width of zero meters).

4.2.4 Soil processes

When using buffer zone width of thirty meters, results show that the amount of glyphosate that was deposited in the topsoil was 0.748 kg a.i ha⁻¹ and 0.450 kg a.i ha⁻¹ of glyphosate during the first application (day fourteen) and second application (day twenty-eight) respectively for year 2012 and 0.739 kg a.i ha⁻¹ and 0.450 kg a.i ha⁻¹ of glyphosate during the first application and second application respectively for year 2013. When using buffer zone width of zero meters, results show that the amount of glyphosate that was deposited in the topsoil was 0.741 kg a.i ha⁻¹ and 0.446 kg a.i ha⁻¹ of glyphosate during the first application and second application respectively for year 2012 and 0.733 kg a.i ha⁻¹ and 0.446 kg a.i ha⁻¹ of glyphosate during the first application and second application respectively for year 2013. The study assumed that volatilization, runoff, macropore flow, and biodegradation were the main competing processes that removed glyphosate from the topsoil (Birkved & Hauschild, 2006; Dijkman et al., 2012).

4.2.4.1 Volatilization

Volatilization from the topsoil was modeled using fugacity approach following Mackay (2001). When using buffer zone width of thirty meters, Figure 4.38 shows the highest removal of glyphosate on day fourteen, twenty-eight, and forty-five. The highest glyphosate removal on day fourteen and day twenty-eight were the results of direct glyphosate contamination in the topsoil right after the application. The highest glyphosate removal on day forty-five was the result of amount of glyphosate washed off from the crop/leaf surfaces on day forty-five which was 0.251 kg a.i ha⁻¹ (Figure 4.28). This amount of glyphosate washed off from the crop/leaf surfaces was added to the topsoil. Also, other amount of glyphosate removed from the topsoil by volatilization in Figure 4.38 was the results of amount of glyphosate that was washed off from the crop/leaf surfaces to the topsoil (Figure 4.28).

Results suggest a gradual decrease in the amount of glyphosate removed from the topsoil by volatilization because there was also a reduction in the amount of glyphosate present in the topsoil due to removal of glyphosate from the topsoil by other competing processes. Results reveal that the amount of glyphosate removed from the topsoil by volatilization was very little, in order of magnitude of 10⁻⁶ to 10⁻¹⁶

for year 2012 and 10^{-6} to 10^{-18} for year 2013. This very little amount of volatilization from the topsoil could be due to the low vapour pressure of glyphosate. The finding is in agreement with Bowmer (1987) who suggested that glyphosate was not removed from the topsoil by volatilization.

When using buffer zone width of zero meters (Figure 4.39), there was no noticeable difference in the results with when using buffer zone width of thirty meters (Figure 4.38) because buffer zones do not influence topsoil volatilization. Results show that total amount of glyphosate removed from the topsoil by volatilization was each 0.0005% of the glyphosate applied (two applications, each at $1.5 \text{ kg a.i ha}^{-1}$) for year 2012 and 2013 (for buffer zone widths of both thirty and zero meters).

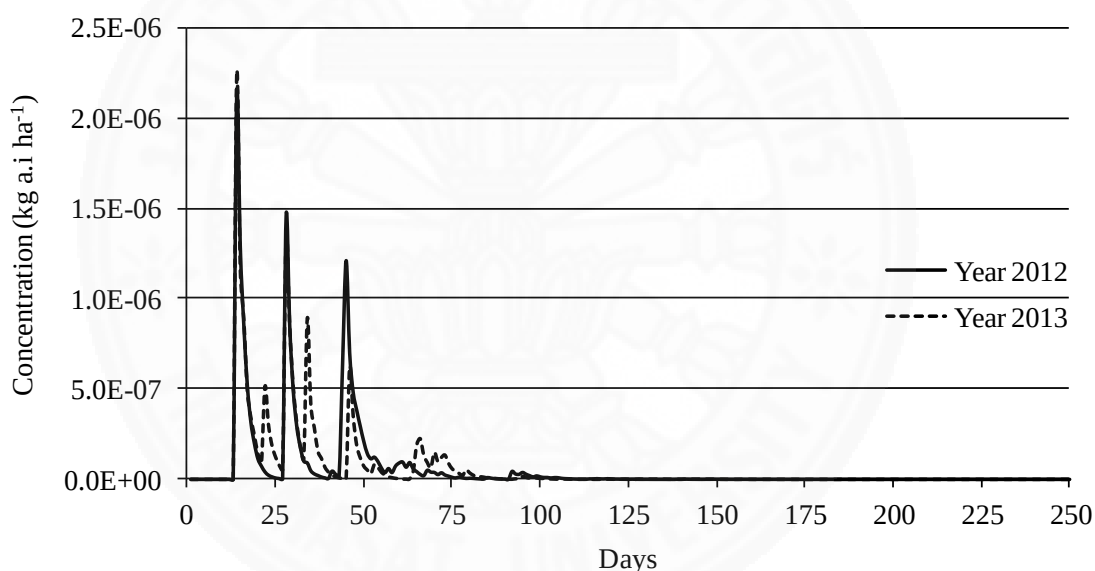


Figure 4.38 Amount of glyphosate removed from the topsoil by volatilization (using buffer zone width of thirty meters).

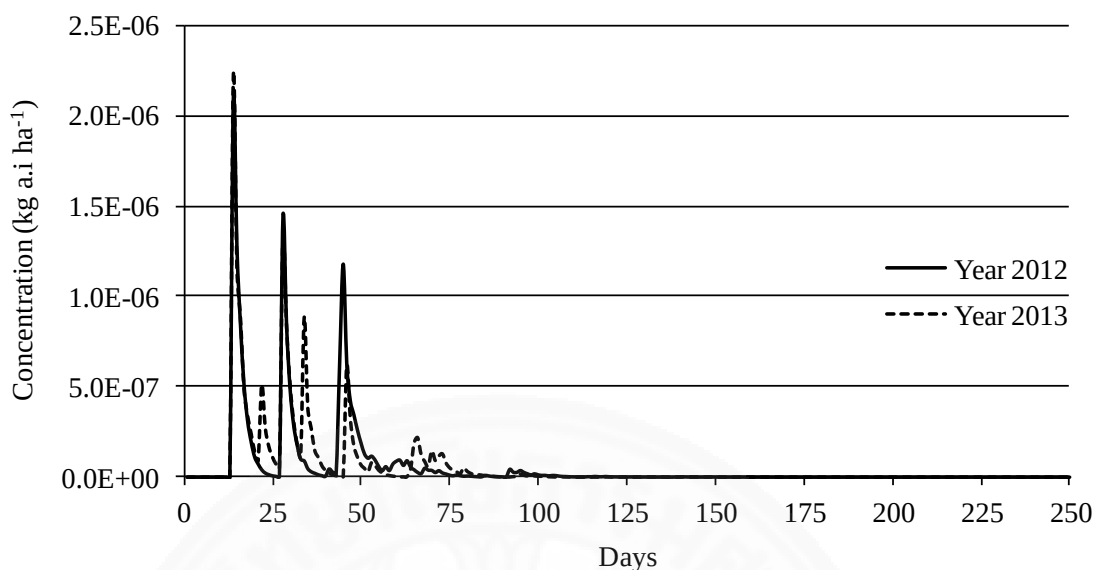


Figure 4.39 Amount of glyphosate removed from the topsoil by volatilization (using buffer zone width of zero meters).

4.2.4.2 Runoff

When using buffer zone width of thirty meters, Figure 4.40 shows that year 2012 had highest amount of glyphosate removal from the topsoil by runoff on day forty-five (3.68×10^{-5} kg a.i ha⁻¹), forty-six (6.94×10^{-5} kg a.i ha⁻¹), forty-seven (1.10×10^{-5} kg a.i ha⁻¹), forty-nine (3.97×10^{-6} kg a.i ha⁻¹), sixty (1.00×10^{-6} kg a.i ha⁻¹), sixty-four (1.31×10^{-6} kg a.i ha⁻¹), ninety-three (1.89×10^{-6} kg a.i ha⁻¹), and ninety-six (1.36×10^{-6} kg a.i ha⁻¹) of the simulation, while year 2013 had highest amount of glyphosate removal from the topsoil by runoff on day twenty-three (2.15×10^{-5} kg a.i ha⁻¹), thirty-five (1.88×10^{-5} kg a.i ha⁻¹), forty-seven (1.41×10^{-5} kg a.i ha⁻¹), sixty-five (1.30×10^{-6} kg a.i ha⁻¹), sixty-six (9.46×10^{-6} kg a.i ha⁻¹), sixty-seven (6.52×10^{-6} kg a.i ha⁻¹), seventy-one (3.68×10^{-6} kg a.i ha⁻¹), seventy-three (5.92×10^{-6} kg a.i ha⁻¹), seventy-four (7.69×10^{-6} kg a.i ha⁻¹), and eighty (2.72×10^{-6} kg a.i ha⁻¹).

Results reveal that year 2013 had two glyphosate runoff events (on day twenty-three and thirty-five) that occurred earlier than the first glyphosate runoff event for year 2012 that occurred on day forty-five. Also, the first runoff event in year 2013 occurred twenty-two days before the first runoff event occurred in year 2012. Results also reveal that the amount of glyphosate removed from the topsoil by runoff

was very little. Total amount of glyphosate removed from the topsoil by runoff was 0.004% and 0.003% of the glyphosate applied (two applications, each at 1.5 kg a.i ha⁻¹) for year 2012 and 2013 respectively. The finding is in agreement with Giesy et al. (2000) who reported that maximum runoff concentration of less than 1.9% of the initial application (8.96 kg a.e ha⁻¹) was found in the runoff water one day after the heavy rainfall. Also, with the exception of wettable-powder herbicides which could lose up to 5% of the dose applied (depending on weather conditions and slope of the field), results is in agreement with Wauchope (1978) that for majority of commercial pesticides, runoff could be less or equal to 0.5% of the dose applied, unless there was a very high rainfall intensity within one to two weeks after the application.

Figure 4.40 shows that after application, the first glyphosate runoff was high and then decreased steadily to the levels it was before the application, the findings similar to Botta et al. (2009). Also, results agrees with Botta et al. (2009) that high rainfall intensity transferred more glyphosate to the water bodies through runoff. Results agrees with Ramwell et al. (2014) that rainfall is responsible for the loss of glyphosate from the topsoil to the surface water, and thus, the first rainfall event after the application would cause more glyphosate runoff to the surface water and subsequently the runoff concentration would decline with time.

Figure 4.40 show that runoff amount was decreasing as the rainfall intensity decreased and vice versa. It was noted that the runoff amount decreased day by day due to the continued decrease in the predicted glyphosate concentration on the soil (i.e. due to the presence of low concentration of glyphosate at the time of runoff event). The decreasing trend in Figure 4.40 is in agreement with Peruzzo et al. (2008).

When using buffer zone width of zero meters, results show that amount of glyphosate removed from the topsoil by runoff were one hundred eighty-one to two hundred sixty-five and one hundred forty-two to two hundred sixty-five times higher than when using buffer zone width of thirty meters for year 2012 and 2013 respectively. Total amount of glyphosate removed from the topsoil by runoff was 1.11% and 0.78% of the glyphosate applied (two applications, each at 1.5 kg a.i ha⁻¹), for year 2012 and 2013 respectively. The finding is in agreement with Giesy et al. (2000) who reported that maximum runoff concentration of less than 1.9% of the initial application (8.96 kg a.e ha⁻¹) was found in the runoff water one day after the

heavy rainfall. The finding is in agreement with Wauchope (1978) that for majority of commercial pesticides, runoff could be less or equal to 0.5% of the dose applied, unless there was a very high rainfall intensity within one to two weeks after the application. In addition, the finding is lower than the finding of Kladviko et al. (2001) who reported that surface runoff for most herbicides was between 2-5% of the dose applied.

Results show that when using buffer zone width of zero meters, high concentration of glyphosate was found in the runoff water which in turn had a profound impact on mortality and survival rates of eggs, tadpoles, and froglets in the water (for both year 2012 and 2013). Results suggest that if vegetative buffer zones are used e.g. of thirty meters wide, could reduce the runoff of glyphosate. This is supported by Dunn et al. (2011) who reported that ten meters buffer zone reduced 52% of the pesticide concentrations (in runoff) while thirty meters buffer zone reduced 78% of the pesticide concentrations (in runoff). Also, Syversen (2005) reported that specific retention (m^{-2}) is higher when using a buffer zone width of five meters. However, Wauchope (1978) suggested that during runoff, erosion control practices would not reduce amount runoff for pesticides with solubilities of 10 ppm (9.98 mg L^{-1}) or higher because runoff is in the water phase (in the simulation, solubility of glyphosate used was $11,500 \text{ mg L}^{-1}$). On the other hand, Watanabe et al. (2007) suggested that organic carbon partition coefficient could be a strong indicator on fate of herbicides in the aquatic environments as compared to the water solubility.

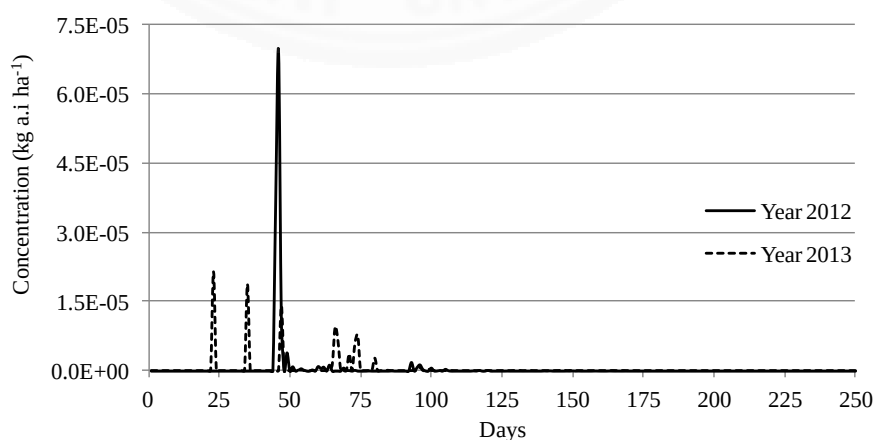


Figure 4.40 Amount of glyphosate removed from the topsoil by runoff (using buffer zone width of thirty meters).

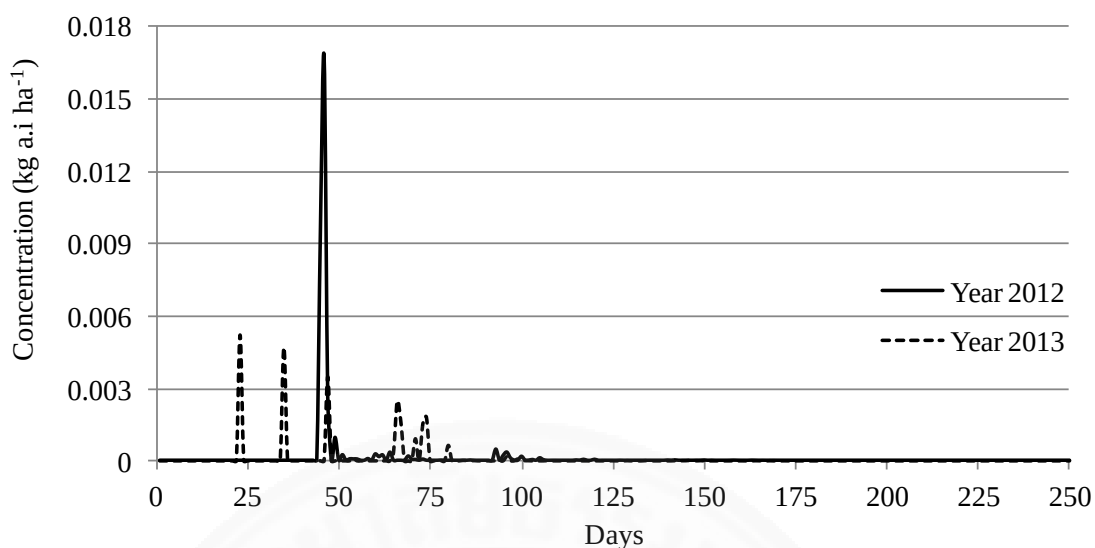


Figure 4.41 Amount of glyphosate removed from the topsoil by runoff (using buffer zone width of zero meters).

The study assumed that rain transferred glyphosate from the agricultural field (topsoil) to the nearby water bodies by runoff (Kladivko et al., 2001; Peruzzo et al., 2008). Figure 4.42 and Figure 4.43 show that year 2012 had the highest concentration of glyphosate contamination in the water than year 2013 because total glyphosate runoffs for year 2012 were 1.43 times higher than year 2013 (for both buffer zone widths of thirty and zero meters). The glyphosate present in the water were degraded by microorganisms, therefore, its concentration decreased gradually with time. When using buffer zone width of zero meters, for both year 2012 and 2013, glyphosate concentration in the water had profound impact on mortality and survival rates of eggs, tadpoles, and froglets.

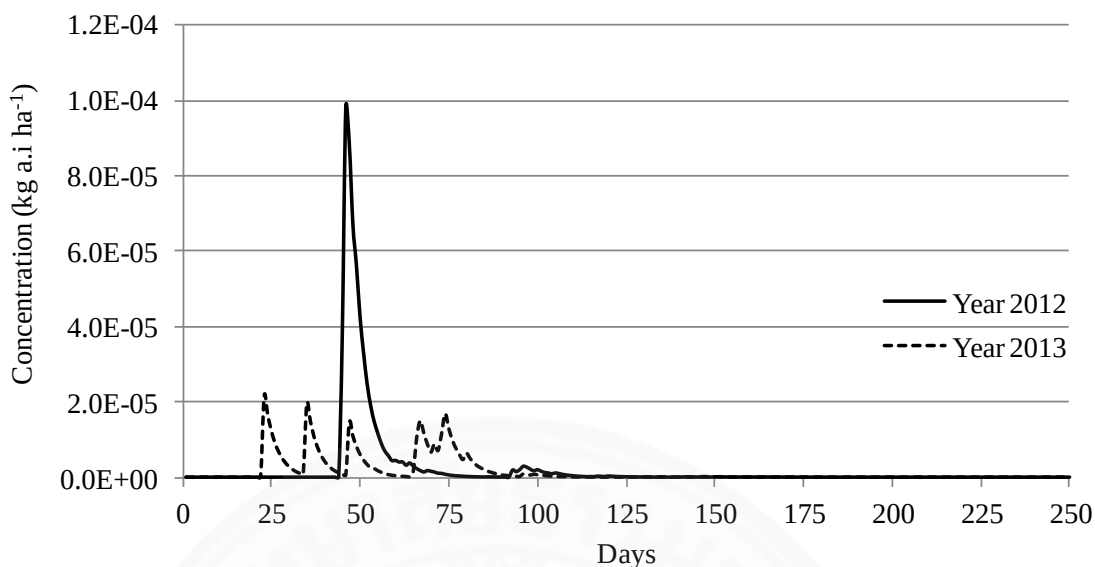


Figure 4.42 Concentration of glyphosate in the water (using buffer zone width of thirty meters).

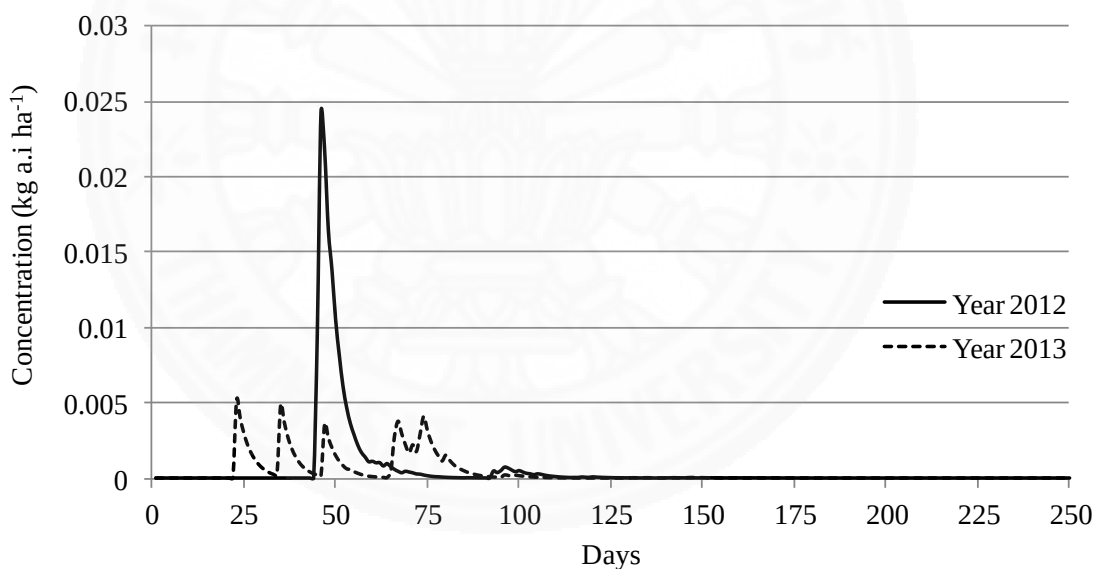


Figure 4.43 Concentration of glyphosate in the water (using buffer zone width of zero meters).

4.2.4.3 Macropore flow

Macropores are cavities that are responsible for transferring (passing) pesticides and other materials from the topsoil to deep soils. The flow (passage) of these materials is referred to as macropore flow. Hall (1993) demonstrated that macropore flow depends on soil composition i.e. percent of clay, silt, and sand in the

soil. In this study, macropore flow was assumed to be through cavities larger than seventy-five micrometer (Dijkman et al., 2012). The study assumed that precipitation (rainfall) water flowed into macropores only if water filled both immobile and slow flowing pores (Dijkman et al., 2012).

When using buffer zone width of thirty meters, Figure 4.44 shows that year 2012 had highest concentration of glyphosate removed by macropores from the topsoil on day forty-five (0.014 kg a.i ha⁻¹), forty-six (0.014 kg a.i ha⁻¹), and forty-seven (0.006 kg a.i ha⁻¹). Year 2013 had highest concentration of glyphosate removed by macropores on day twenty-three (0.005 kg a.i ha⁻¹), thirty-five (0.006 kg a.i ha⁻¹), forty-seven (0.004 kg a.i ha⁻¹), sixty-six (0.004 kg a.i ha⁻¹), sixty-seven (0.002 kg a.i ha⁻¹), seventy-one (0.001 kg a.i ha⁻¹), seventy-three (0.002 kg a.i ha⁻¹), and seventy-four (0.002 kg a.i ha⁻¹).

The study found that total macropore flow were 1.43% and 0.94% of the glyphosate applied (two applications, each at 1.5 kg a.i ha⁻¹) for year 2012 and 2013 respectively. When using buffer zone width of zero meters, total macropore flow were 1.33% and 0.88% of the glyphosate applied for year 2012 and 2013 respectively (Figure 4.45). There is no noticeable difference in the amount of macropore flow between when using buffer zone width of thirty meters and zero meters because buffer zones do not affect macropore flow.

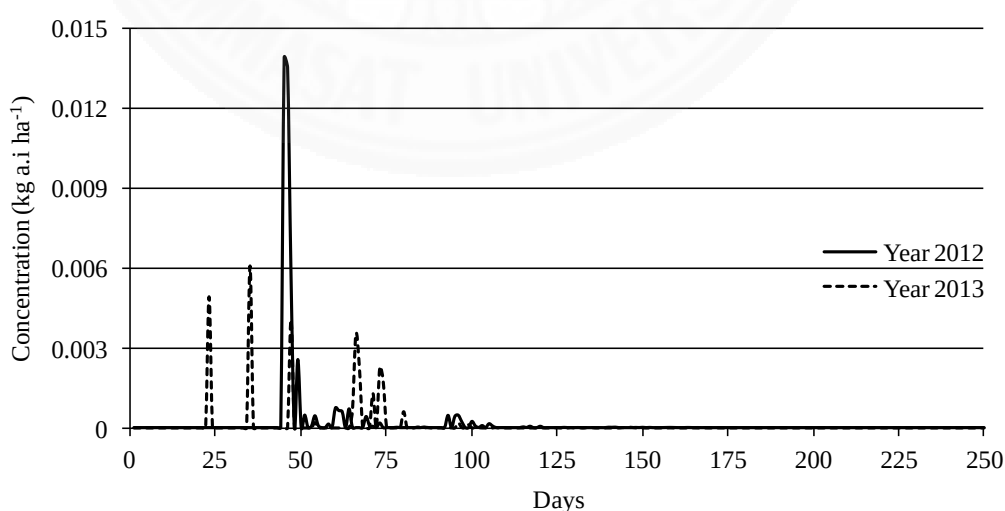


Figure 4.44 Amount of glyphosate removed from the topsoil by macropore flow (using buffer zone width of thirty meters).

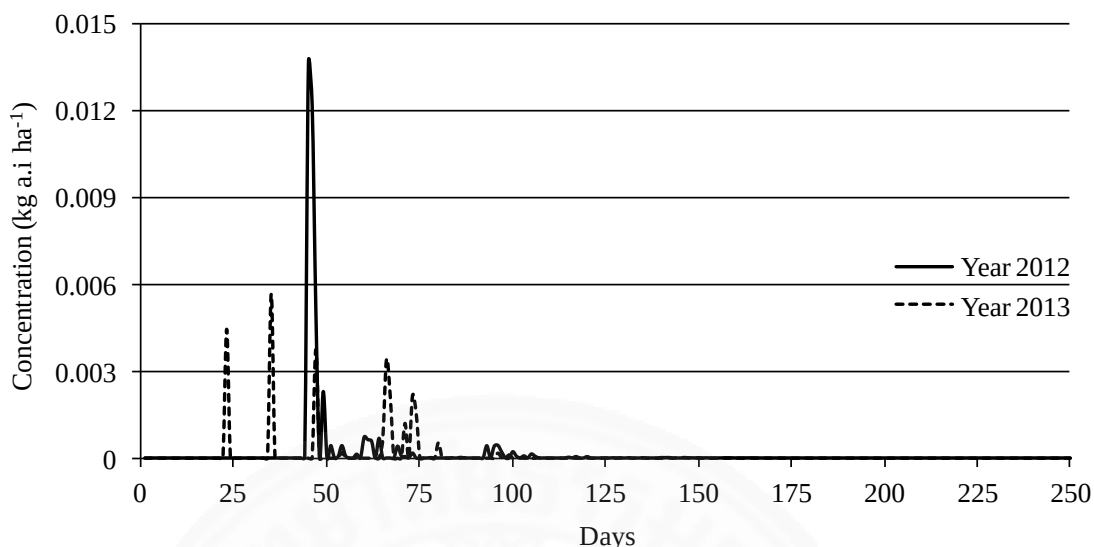


Figure 4.45 Amount of glyphosate removed from the topsoil by macropore flow (using buffer zone width of zero meters).

4.2.4.4 Soil degradation

The study assumed that degradation process in the topsoil was mainly due to the action of microorganisms (Contardo-Jara et al., 2009) following the first-order degradation kinetics. When using buffer zone width of thirty meters, Figure 4.46 shows the highest amount of glyphosate removed from the topsoil by biodegradation on day fourteen ($0.27 \text{ kg a.i ha}^{-1}$) and twenty-eight ($0.18 \text{ kg a.i ha}^{-1}$) of the simulation for year 2012. These two concentrations were high because they were drawn from the initial amount of glyphosate that were directly deposited in the topsoil. The amount of glyphosate removed by biodegradation from the topsoil decreased steadily to almost zero by day forty of the simulation. Glyphosate was also removed from the topsoil on day forty-four ($0.08 \text{ kg a.i ha}^{-1}$), forty-five ($0.12 \text{ kg a.i ha}^{-1}$), and forty-six ($0.10 \text{ kg a.i ha}^{-1}$) mainly due to the glyphosate present in the topsoil as a result of the washoff events that transferred glyphosate from the crop/leaf surfaces to the topsoil on day forty-four ($0.20 \text{ kg a.i ha}^{-1}$), forty-five ($0.25 \text{ kg a.i ha}^{-1}$), and forty-six ($0.06 \text{ kg a.i ha}^{-1}$) (Figure 4.28).

For year 2013, Figure 4.46 shows the highest amount of glyphosate removed from the topsoil by biodegradation on day fourteen ($0.24 \text{ kg a.i ha}^{-1}$) and twenty-eight ($0.18 \text{ kg a.i ha}^{-1}$) of the simulation. These were drawn from the initial amount of

glyphosate that were directly deposited in the topsoil. The amount of glyphosate that were removed from the topsoil decreased steadily to almost zero by day forty of the simulation. Glyphosate was also removed from the topsoil by biodegradation on day twenty-two (0.06 kg a.i ha⁻¹), thirty-four (0.09 kg a.i ha⁻¹), and forty-six (0.07 kg a.i ha⁻¹) due to the presence of glyphosate in the topsoil which were the results of the washoff events that transferred glyphosate from the crop/leaf surfaces to the topsoil on day twenty-two (0.16 kg a.i ha⁻¹), thirty-four (0.24 kg a.i ha⁻¹), and forty-six (0.16 kg a.i ha⁻¹) (Figure 4.28). However, the mobility of glyphosate in the topsoil is affected by soil type, pH, level of phosphate (Sprankle et al., 1975), temperature, and sunlight (Mallat & Barceló, 1998).

When using buffer zone width of thirty meters, total amount of glyphosate removed from the topsoil by biodegradation were 64.56% and 66.12% of the glyphosate applied (two applications, each at 1.5 kg a.i ha⁻¹) for year 2012 and 2013 respectively. Also, when using buffer zone width of zero meters, total amount of glyphosate removed from the topsoil by biodegradation were 62.78% and 64.37% of the glyphosate applied for year 2012 and 2013 respectively. Results show that there is no noticeable difference in the amount of glyphosate removed from the topsoil by biodegradation when using buffer zone width of thirty meters (Figure 4.46) and when using buffer zone width of zero meters (Figure 4.47).

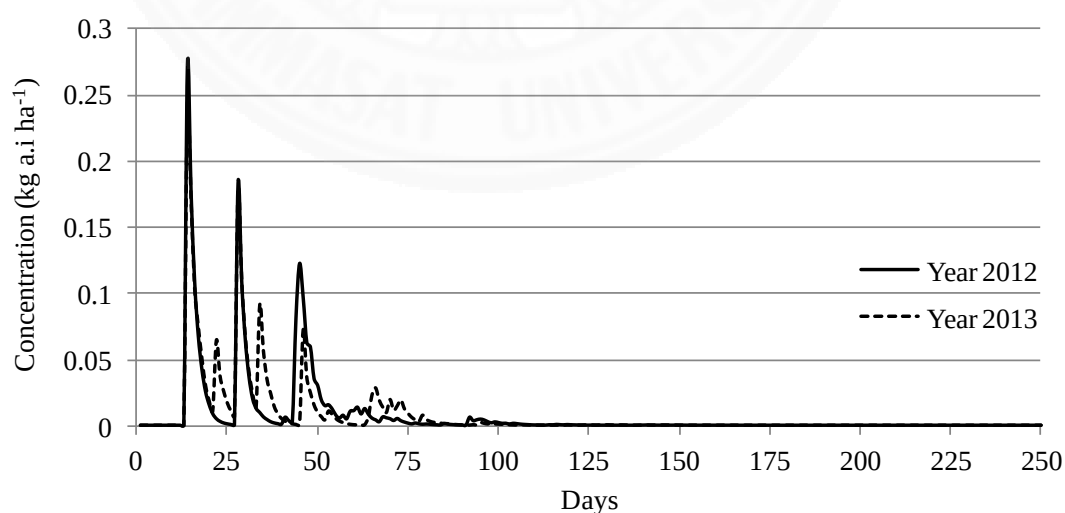


Figure 4.46 Amount of glyphosate removed from the topsoil by biodegradation (using buffer zone width of thirty meters).

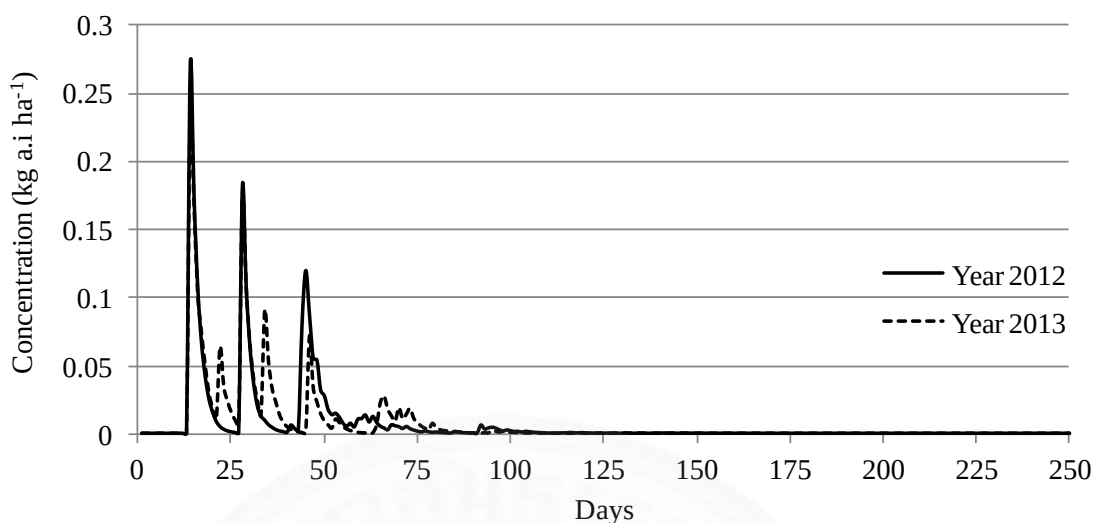


Figure 4.47 Amount of glyphosate removed from the topsoil by biodegradation (using buffer zone width of zero meters).

Amount of glyphosate remained in the topsoil was assessed using mass balance. Results suggest a gradual decrease with time in the amount of glyphosate remained in the topsoil. When using buffer zone width of thirty meters, Figure 4.48 suggests that if there was no washoff events that transferred glyphosate from the crop/leaf surfaces to the topsoil on day forty-four (0.20 kg a.i ha⁻¹), forty-five (0.25 kg a.i ha⁻¹), forty-six (0.06 kg a.i ha⁻¹), and forty-eight (0.04 kg a.i ha⁻¹) for year 2012 and on day twenty-two (0.16 kg a.i ha⁻¹), thirty-four (0.24 kg a.i ha⁻¹), and forty-six (0.16 kg a.i ha⁻¹) for year 2013 (Figure 4.28), all glyphosate in the topsoil could have been removed completely by the competing processes by day forty of the simulation. This result suggests that glyphosate does not stay longer in the topsoil. The finding agrees with Henderson et al. (2010) who reported that glyphosate does not stay longer in the environment. Also, results show that there is no noticeable difference in the amount of glyphosate that remained in the topsoil when using buffer zone width of thirty meters (Figure 4.48) and when using buffer zone width of zero meters (Figure 4.49).

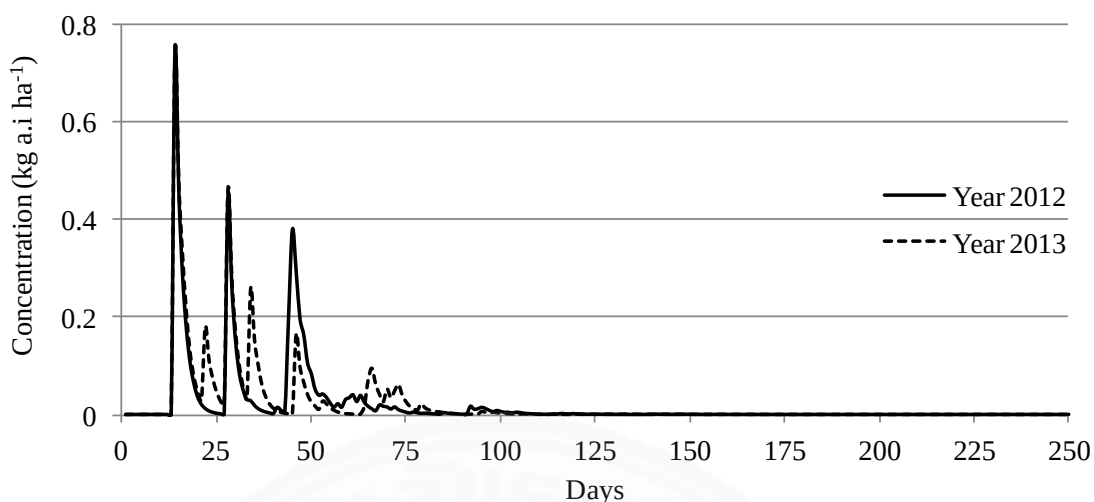


Figure 4.48 Concentration of glyphosate remained in the topsoil (using buffer zone width of thirty meters).

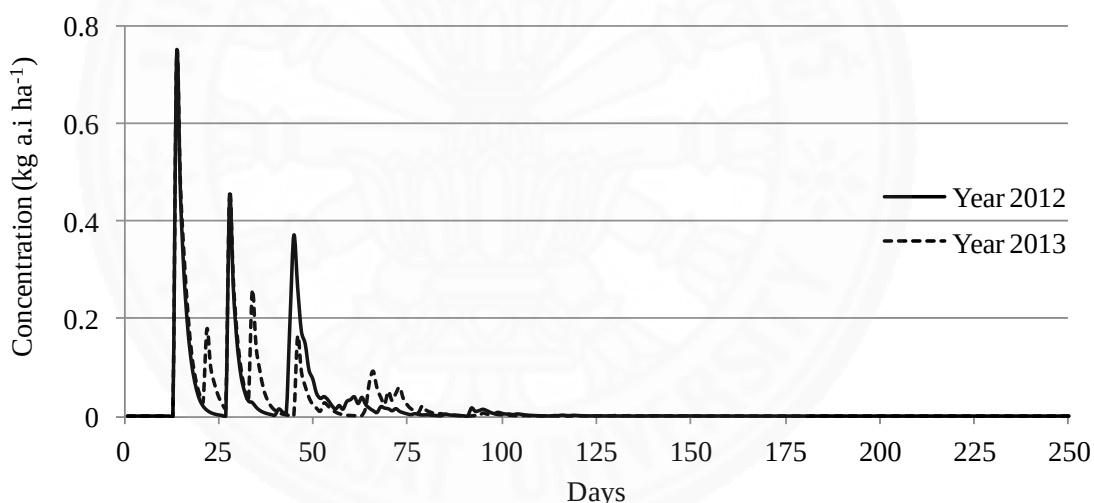


Figure 4.49 Concentration of glyphosate remained in the topsoil (using buffer zone width of zero meters).

4.2.5 Mortality and population dynamics

The study modeled the population dynamics of frogs by studying its four development stages namely eggs, tadpoles, froglets, and adults. The amount of glyphosate that was removed from the topsoil by runoff was assumed to contaminate the water body near the agricultural field. The amount of glyphosate in the water body was used to estimate both glyphosate-induced daily mortality and survival rates of

eggs, tadpoles, and froglets while the amount of glyphosate in the topsoil was used to estimate both glyphosate-induced daily mortality and survival rates of adult frogs. Glyphosate-induced daily mortality and survival rates were modeled following Johnston et al. (2014), Focks et al. (2014a), Galic et al. (2014), and Focks et al. (2014b). Two scenarios were modeled, using runoff of glyphosate from the topsoil as direct contamination in the water body when width of buffer zone was (i) thirty meters and (ii) zero meters. The study started the simulation with 1,000 tadpoles.

When using buffer zone width of thirty meters, amount of glyphosate that was present in the runoff from the topsoil to the water body were very little (Figure 4.40). Glyphosate-induced mortality rates were therefore low, which resulted to high survival rates of eggs, tadpoles, and froglets for both year 2012 (Figure 4.50) and 2013 (Figure 4.51). The survival rates of adult frogs varied with the predicted concentration of glyphosate in the topsoil (Figure 4.52). Figure 4.53 and Figure 4.54 show that the survival rates of eggs, tadpoles, and froglets increased when the concentration of glyphosate in the water body decreased. Figure 4.55 and Figure 4.56 show that many eggs and tadpoles survived the very little amount of glyphosate that was present in the water and grew to froglets and adults frogs.

Based on Table 4.2, LC_{50} values of glyphosate were assumed to be 684 mg a.i L^{-1} for each development stage of frogs to simulate potential population dynamics. This LC_{50} value was 142.5 times less toxic for eggs and tadpoles stages and 9.9 and 10.4 times less toxic for froglets and adults stages respectively. Simulation was repeated for both year 2012 and 2013. Figure 4.57 and Figure 4.58 did not show noticeable difference with Figure 4.55 and Figure 4.56 due to low mortality of frogs that occurred mainly because the concentration of glyphosate in the water body and in the topsoil was very little as well as the assumed LC_{50} values of glyphosate of 684 mg a.i L^{-1} was less toxic.

When using buffer zone width of zero meters, the amount of glyphosate that was present in the runoff was higher than when using buffer zone width of thirty meters. The highest concentration of glyphosate runoff for year 2013 on day twenty-three and thirty-five (Figure 4.41) that occurred earlier than the first runoff event for year 2012 had profound impact on mortality and survival rates of eggs, tadpoles, froglets (Figure 4.59 and Figure 4.60) and adult frogs (Figure 4.61). Figure 4.59 and

Figure 4.60 suggest that between day one and day forty of the simulation, survival rates were higher for year 2012 than 2013. This was due to the variations in the amount of glyphosate transferred by runoff from the topsoil to the water body. From day forty to day two hundred fifty, Figure 4.59 shows that there were variations in the survival rates which varied with the predicted glyphosate concentrations in the topsoil (Figure 4.61) and in the water body (Figure 4.62 and Figure 4.63).

Results further reveal that within the first fifty days of simulation, year 2012 had small amount of washoff and runoff compared to year 2013. Within twenty days after the first application, there was high mortality of eggs, tadpoles, and froglets for year 2013 than it was for year 2012. Results show that for year 2013, all tadpoles died within eighty-six days (Figure 4.64) while for year 2012 all tadpoles died within one hundred five days after the first application due to high glyphosate contamination in the water body (Figure 4.65).

Simulation of the population dynamics using weather data for year 2013 suggests increased death of eggs, tadpoles and froglets (Figure 4.64) mainly because the first rainfall event was recorded eight days after the first application, while for year 2012 the first rainfall event was recorded nineteen days after the first application. The death of eggs, tadpoles, and froglets suggest that glyphosate contamination in the water body could be lethal to the survival of frogs. Results found that population size of amphibians highly depend on survival of eggs, tadpoles and froglets, the finding similar to Swanack et al. (2009). Results suggest that the use of glyphosate in agriculture could therefore be responsible for the population decline of frogs.

Based on Table 4.2, LC_{50} values of glyphosate were assumed to be 684 mg a.i L^{-1} for each development stage of frogs to simulate potential population dynamics. The LC_{50} values used were 142.5 times less toxic for eggs and tadpoles, as well as 9.9 and 10.4 times less toxic for froglets and adults frogs respectively. Simulation was repeated for both year 2012 and 2013. Figure 4.66 and Figure 4.67 show that between day one and day one hundred of the simulation, there were small number of eggs, tadpoles, froglets, and adults in the population. The number increased rapidly between day one hundred and day two hundred fifty of the simulation mainly due to the low concentration of glyphosate in the water body and in the topsoil as well as due to the reduced toxicity of glyphosate (reduced LC_{50} values). Results suggest that toxicity of

glyphosate to eggs, tadpoles, froglets, and adult frogs (LC_{50}) has significant impact on population dynamics of frogs. Also, when using LC_{50} of 684 mg a.i L^{-1} and buffer zone width of thirty meters, population (Figure 4.57 and Figure 4.58) were almost twice as much as when the population were when using LC_{50} of 684 mg a.i L^{-1} and width of buffer zone of zero meters (Figure 4.66 and Figure 4.67). Results suggest that the use of less toxic formulations of glyphosate could help to reduce the impact of glyphosate on population of frogs. Furthermore, the use of buffer zone around the agricultural field helps to reduce the amount of glyphosate from being transferred by runoff from the agricultural field to the non-target sites (e.g. water body).

Table 4.2 LC_{50} values of glyphosate formulations for *Crinia insignifera*

Life stage	Chemical assessed	Parameter assessed	Type of study	Effects concentrations/LC ₅₀ (mg a.i L ⁻¹)	Species found in	Reference
Tadpoles	Glyphosate Isopropylamine	Morality	Single species laboratory test (48 h)	48 h LC ₅₀ = >684	Southwestern Australia	(Mann & Bidwell, 1999)
	Roundup®			48 h LC ₅₀ = 10		
	Touchdown®			48 h LC ₅₀ = 27.3		
	Roundup® Biactive			48 h LC ₅₀ = 1,372		
Metamorph	Roundup®	Morality		48 h LC ₅₀ = 144		(Bidwell & Gorrie, 1995)
Adult	Roundup®			48 h LC ₅₀ = 137		
	Technical-Grade Glyphosate Acid			48 h LC ₅₀ = 111.5		

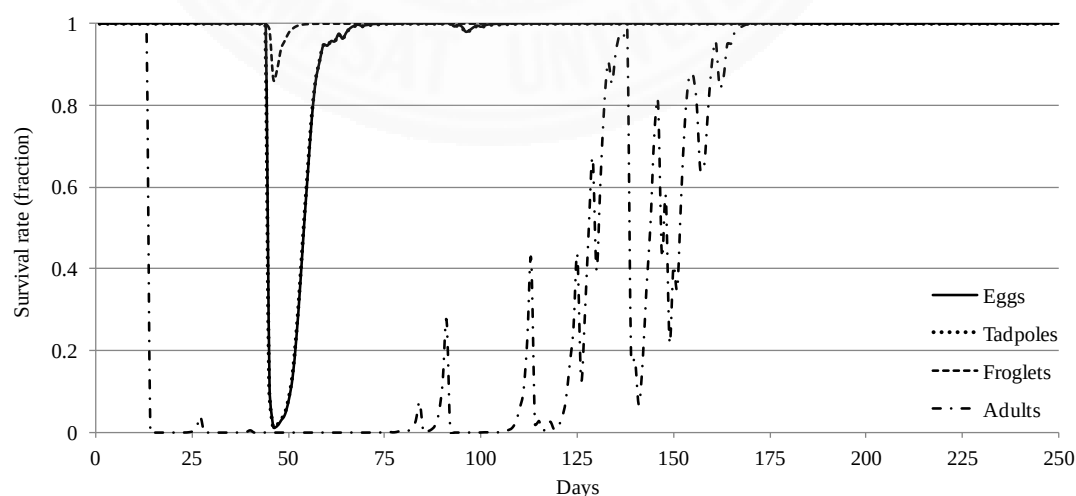


Figure 4.50 Survival rates of frogs (year 2012, using buffer zone width of thirty meters).

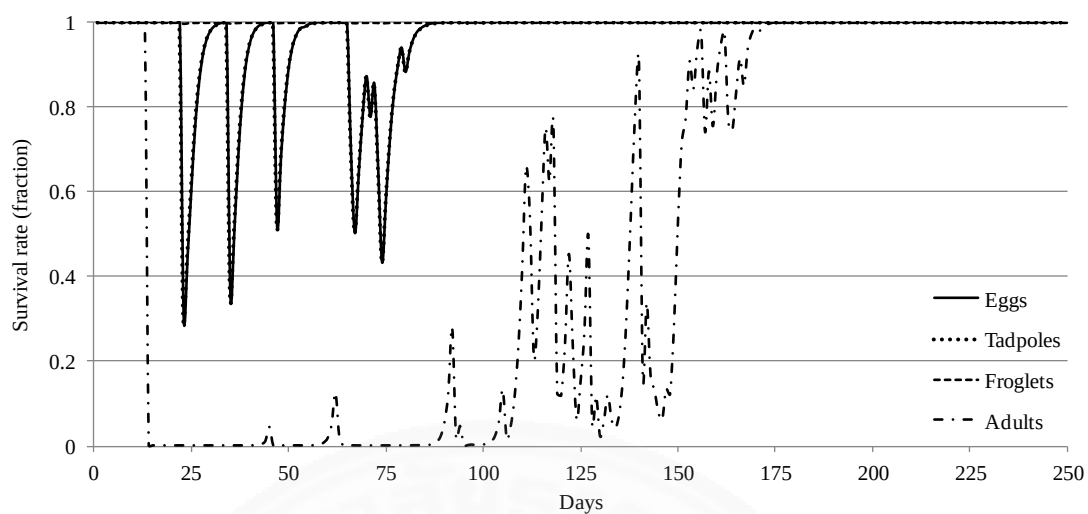


Figure 4.51 Survival rates of frogs (year 2013, using buffer zone width of thirty meters).

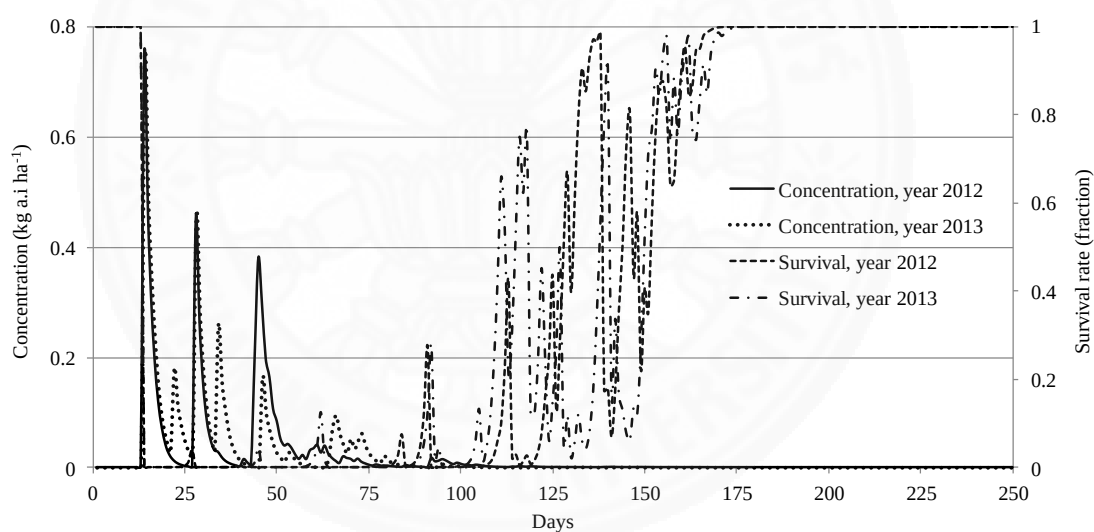


Figure 4.52 Concentration of glyphosate in the topsoil versus survival rates of adult frogs (using buffer zone width of thirty meters).

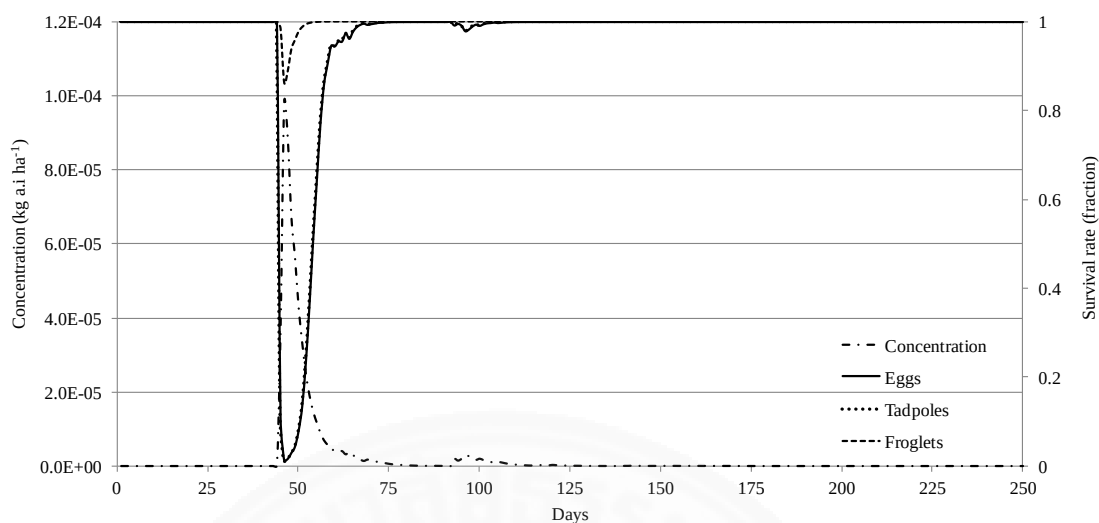


Figure 4.53 Concentration of glyphosate in the water body versus survival rates of eggs, tadpoles, and froglets (year 2012, using buffer zone width of thirty meters).

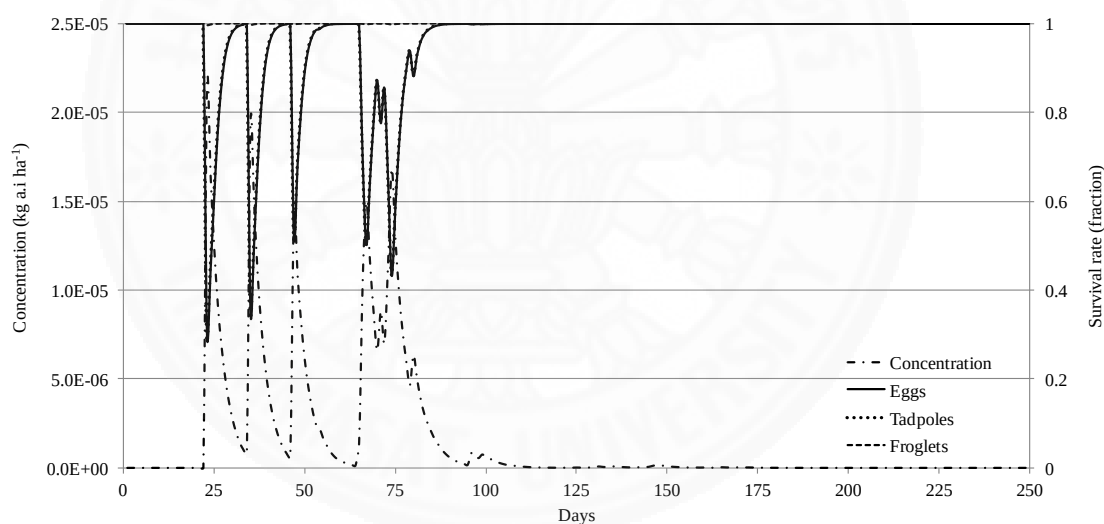


Figure 4.54 Concentration of glyphosate in the water body versus survival rates of eggs, tadpoles, and froglets (year 2013, using buffer zone width of thirty meters).

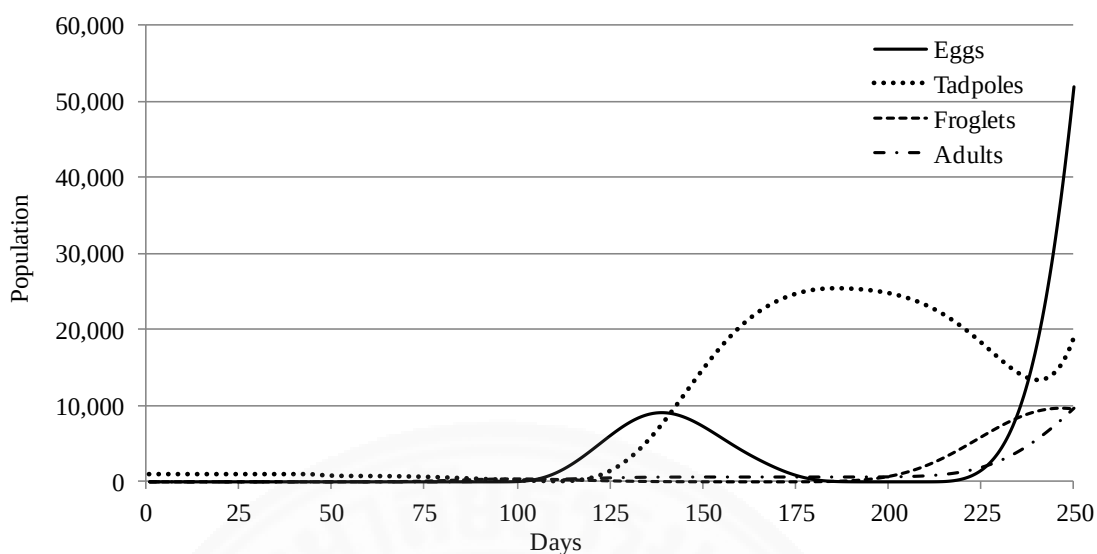


Figure 4.55 Population dynamics of frogs (year 2012, using buffer zone width of thirty meters).

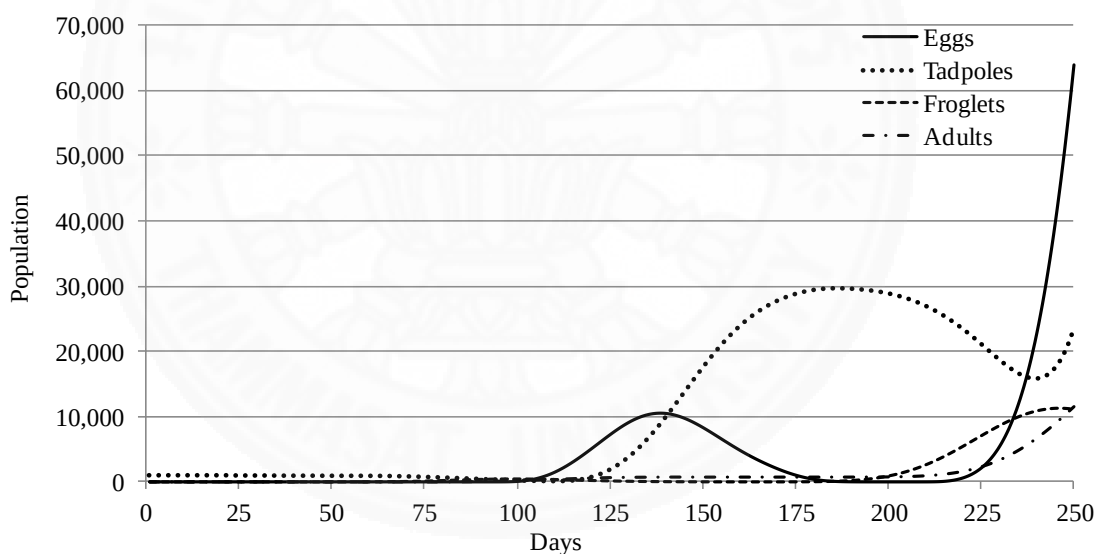


Figure 4.56 Population dynamics of frogs (year 2013, using buffer zone width of thirty meters).

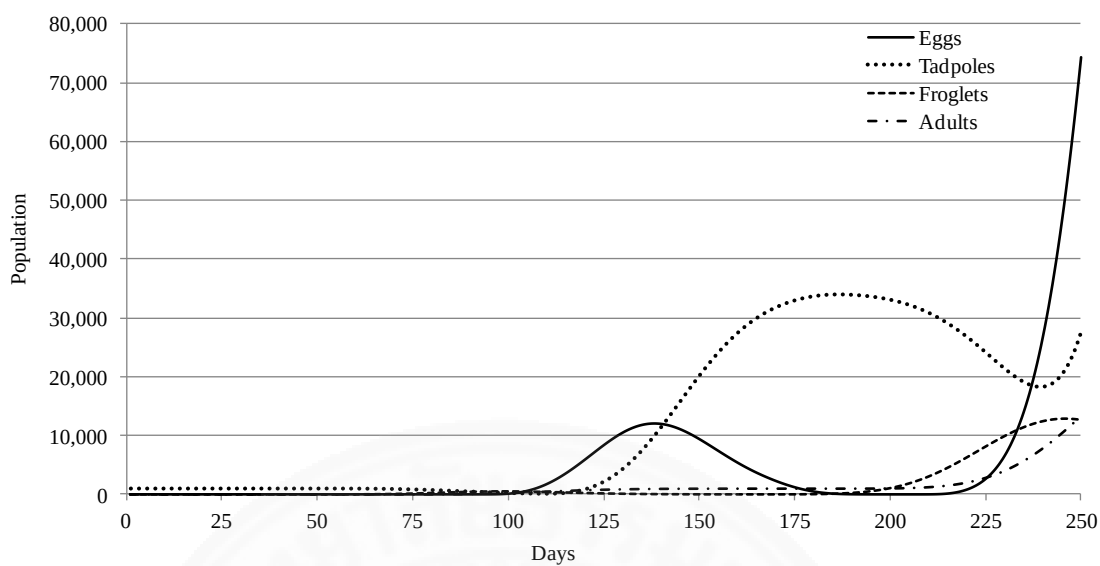


Figure 4.57 Population dynamics of frogs (year 2012, using buffer zone width of thirty meters, LC_{50} of $684 \text{ mg a.i L}^{-1}$).

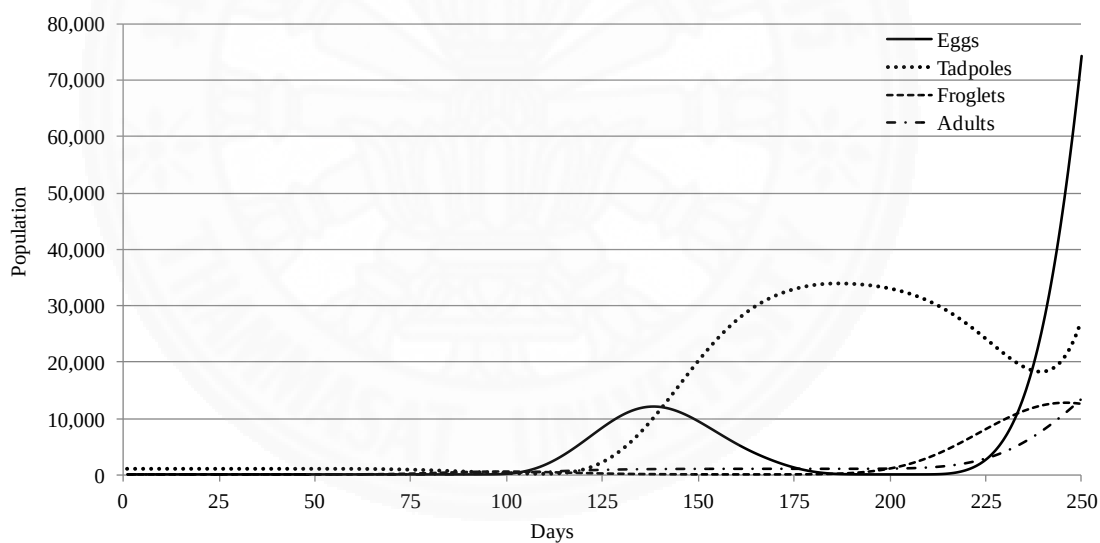


Figure 4.58 Population dynamics of frogs (year 2013, using buffer zone width of thirty meters, LC_{50} of $684 \text{ mg a.i L}^{-1}$).

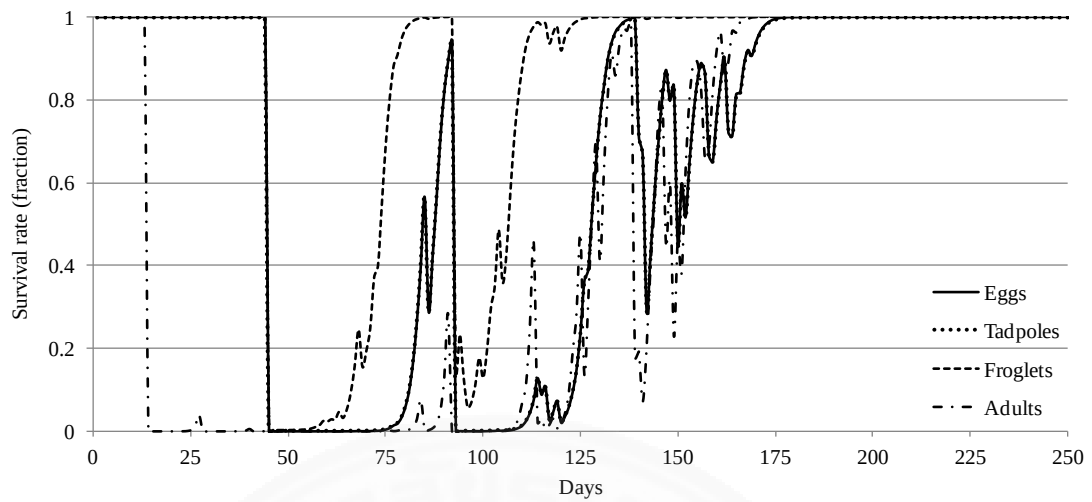


Figure 4.59 Survival rates of frogs (year 2012, using buffer zone width of zero meters).

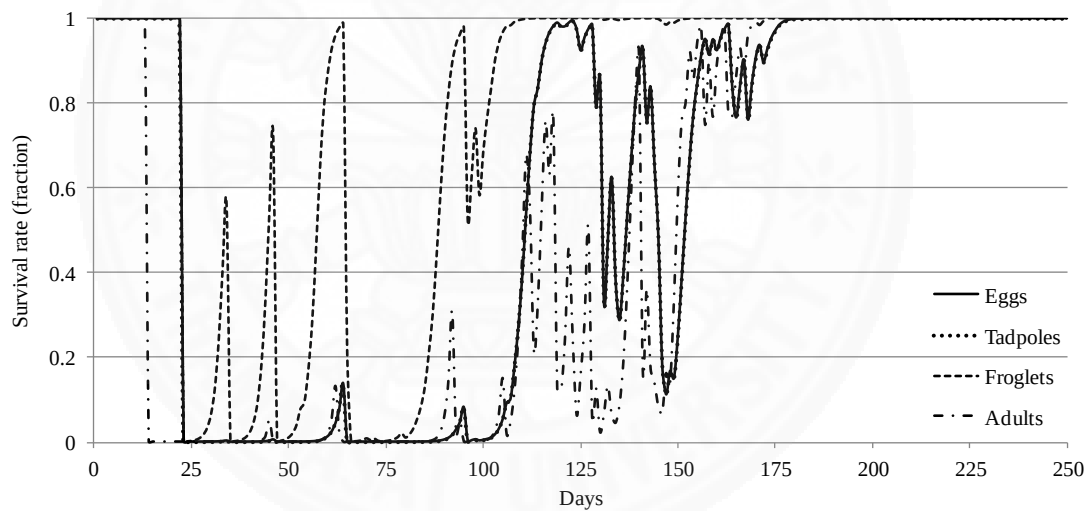


Figure 4.60 Survival rates of frogs (year 2013, using buffer zone width of zero meters).

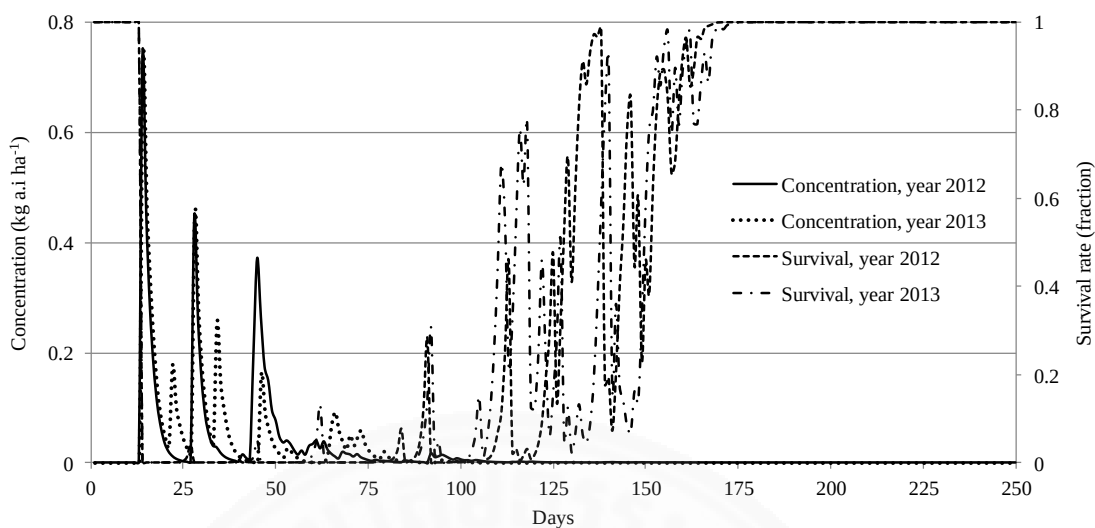


Figure 4.61 Concentration of glyphosate in the topsoil versus survival rates of adult frogs (using buffer zone width of zero meters).

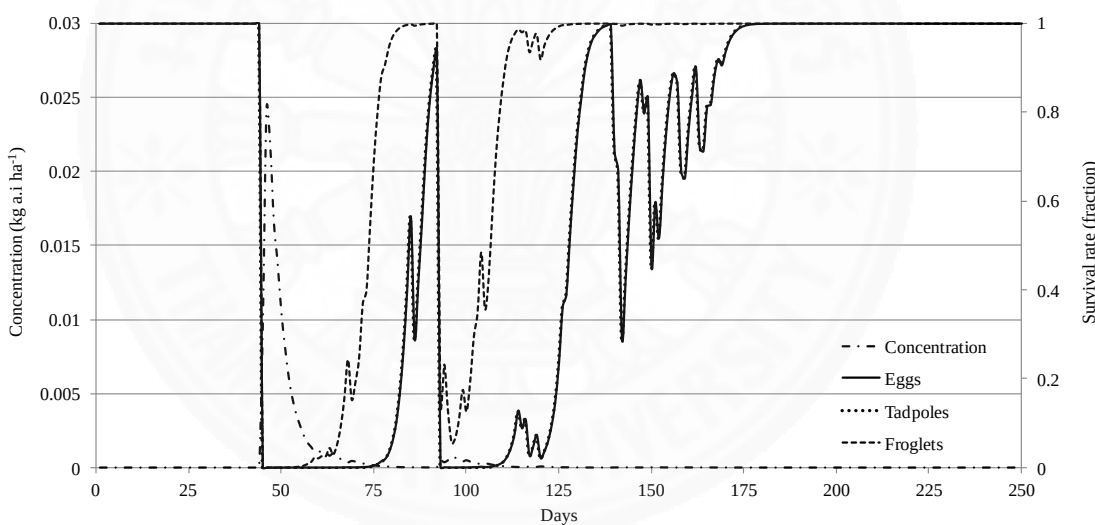


Figure 4.62 Concentration of glyphosate in the water body versus survival rates of eggs, tadpoles, and froglets (year 2012, using buffer zone width of zero meters).

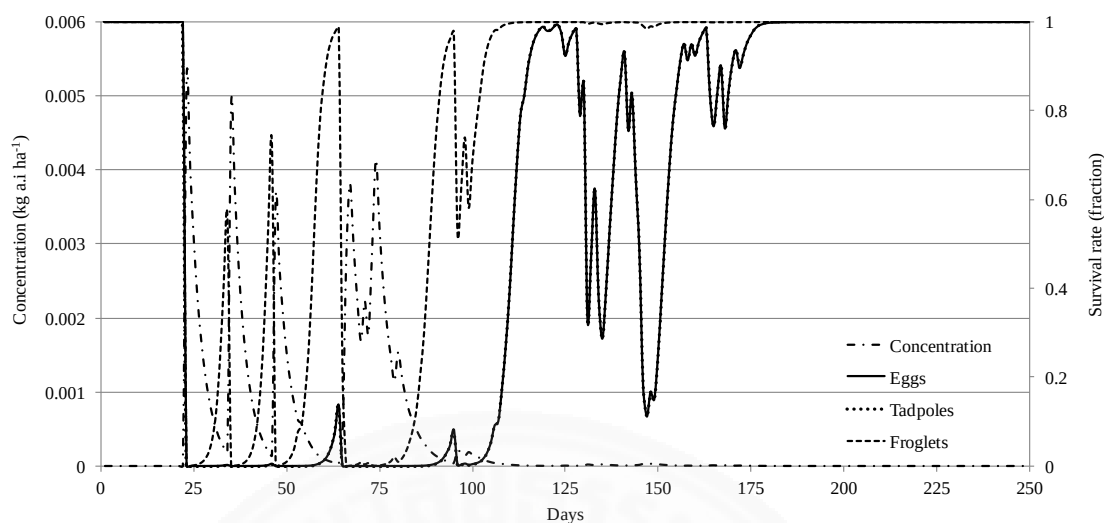


Figure 4.63 Concentration of glyphosate in the water body versus survival rates of eggs, tadpoles, and froglets (year 2013, using buffer zone width of zero meters).

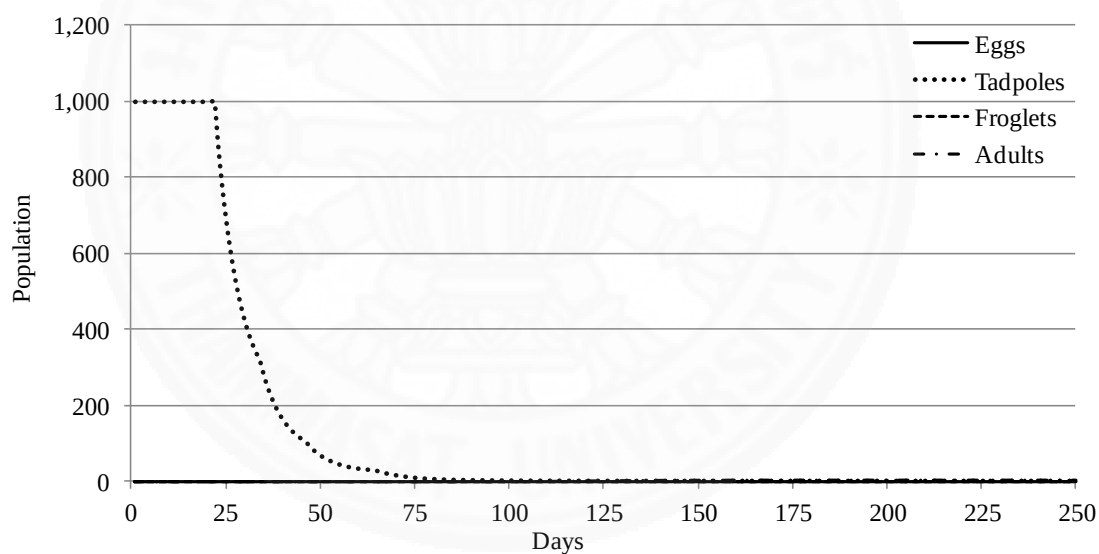


Figure 4.64 Population dynamics of frogs (year 2013, using buffer zone width of zero meters).

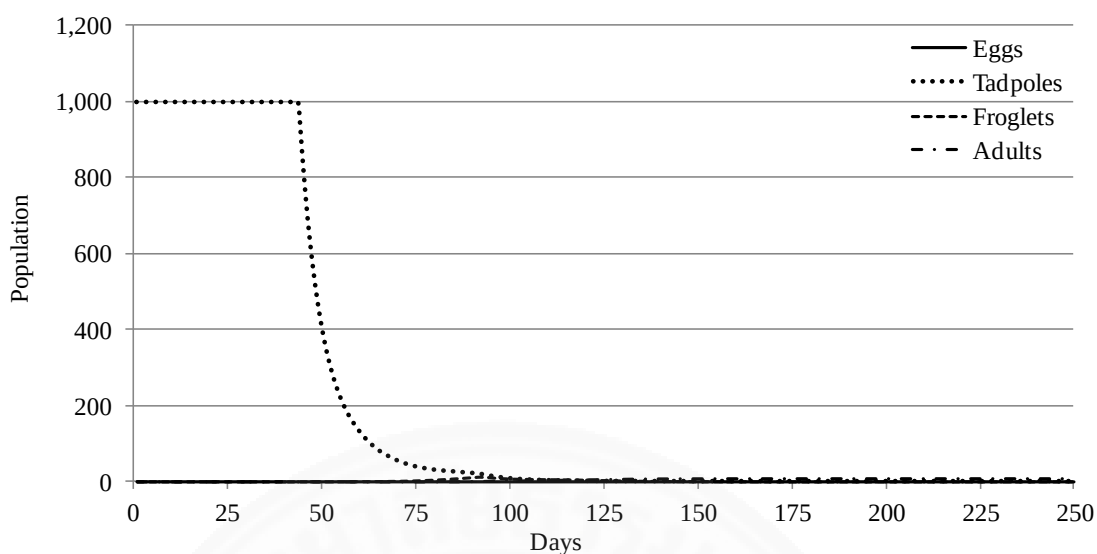


Figure 4.65 Population dynamics of frogs (year 2012, using buffer zone width of zero meters).

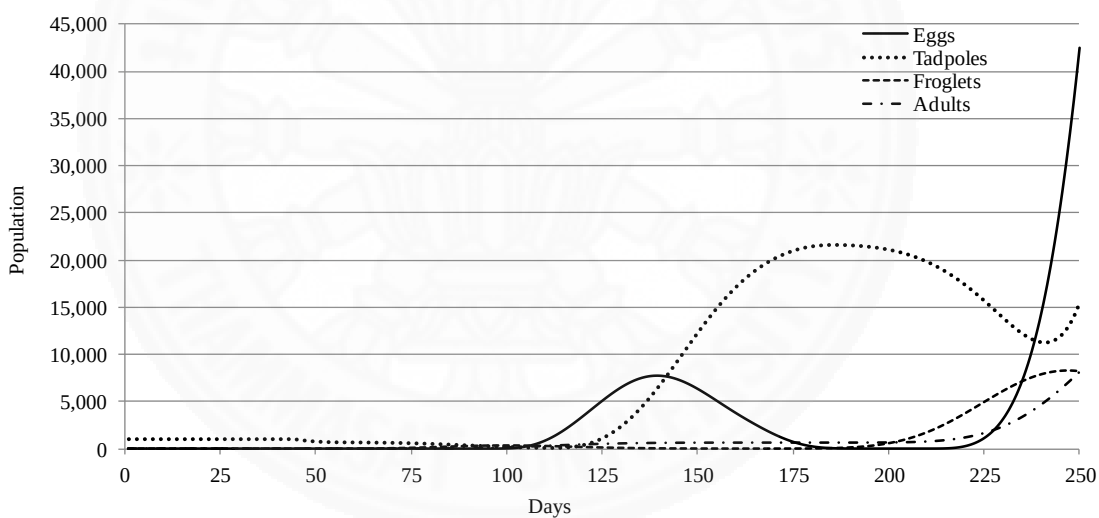


Figure 4.66 Population dynamics of frogs (year 2012, using buffer zone width of zero meters, LC_{50} of $684 \text{ mg a.i L}^{-1}$).

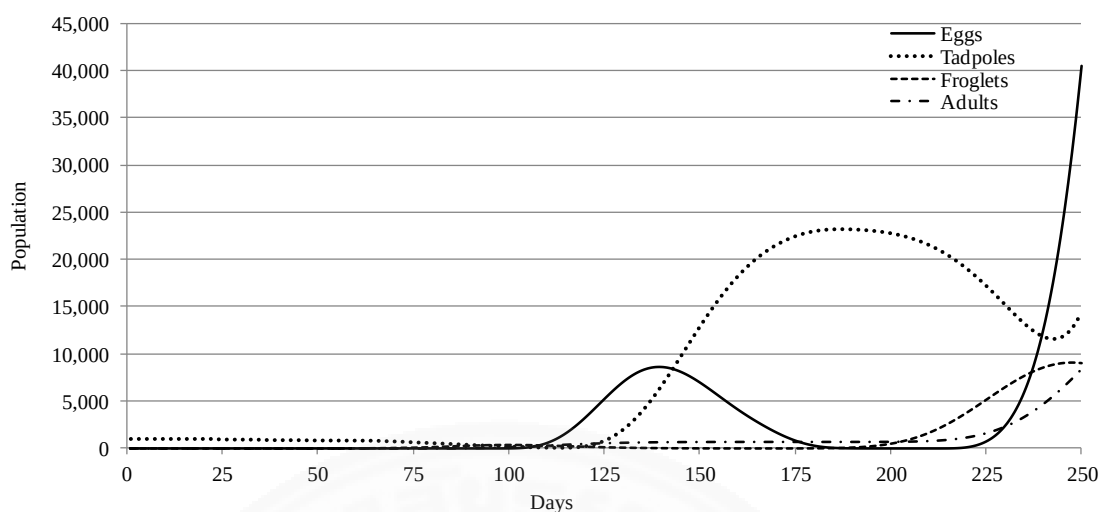


Figure 4.67 Population dynamics of frogs (year 2013, using buffer zone width of zero meters, LC_{50} of $684 \text{ mg a.i L}^{-1}$).

4.3 Effects of spray drifts on frogs

Gilreath et al. (2001) suggested that glyphosate drifts could kill or damage non-target organisms, such as frogs (Miller, 2006). Velini et al. (2008) also suggested that there could be potential effects to non-crop species due to long-term exposure to drifts of glyphosate-based formulation, of which there is still lack of understanding. It is with this reasons that the present study wanted to find out what would happen to non-target organisms, specifically frogs, if pesticide users fail to follow recommended practices when working with pesticides (Felix & Sharp, 2015; Felix & Sharp, 2016).

The study simulated the effect of no spray zone widths (buffer zone widths) around the agricultural field. The reason that this safety zone is ignored could be associated with the perception that the zone is wasted for nothing instead of being used to grow crops. Simulation was therefore performed to study the effect of glyphosate spray drifts on population dynamics of non-target organisms using frog as case study. Climate conditions for year 2012 and 2013 were used in the simulation. Two scenarios were studied using (i) width of no spray zone of zero meters and (ii) width of no spray zone of thirty meters. These two scenarios were chosen based on several studies (Table A.12) which have recommended that the width of no spray zone of five meters to thirty meters around the agricultural field reduces spray drifts from crossing the agricultural field to non-target sites.

When using width of no spray zone of zero meters, the study assumed that there was no buffer zone that prevented spray drifts from the agricultural field to the water body. When using width of no spray zone of thirty meters, the study assumed that agricultural field and the water body were separated by the width of no spray zone of thirty meters. The study assumed that spray drifts (glyphosate) that crossed the agricultural field were deposited in the water body located adjacent to the agricultural field. Furthermore, the study assumed that glyphosate in the water body was degraded by microorganisms (Borggaard & Gimsing, 2008; Rueppel et al., 1977). In addition, the study assumed that eggs, tadpoles, froglets, and adult frogs were exposed to glyphosate in the water body.

Simulation was done for two hundred fifty days. The study assumed that the predicted glyphosate concentration in the water body on the first day of simulation was equal to the amount of spray drifts that crossed the agricultural field after the application. The predicted glyphosate concentration in the water body on the second day of simulation was estimated by taking the predicted concentration that remained on the previous day minus the amount of glyphosate degraded on the second day. The same principle was applied for the subsequent days. Glyphosate-induced daily mortality and survival rates were estimated using predicted daily glyphosate concentrations in the water body and the LC_{50} values of glyphosate for each development stage of frogs following Focks et al. (2014a), Focks et al. (2014b), Galic et al. (2014), and Johnston et al. (2014).

When using width of no spray zone of zero meters, simulations were performed with two glyphosate applications each at $1.5 \text{ kg a.i ha}^{-1}$ on day fourteen and day twenty-eight of the simulation. Results found that spray drifts were each $0.013 \text{ kg a.i ha}^{-1}$ for first application and second application for both year 2012 and 2013. Figure 4.68 shows the predicted concentration of glyphosate with the highest glyphosate concentration on day fourteen and day twenty-eight of the simulation. These highest concentrations were the results of direct contamination of spray drifts from the agricultural field to the water body after the application.

Results show that glyphosate concentrations decreased steadily following first order degradation kinetics as glyphosate was degraded by the action of microorganisms in the water body (Borggaard & Gimsing, 2008; Rueppel et al.,

1977). Figure 4.69 and Figure 4.70 show the survival rates of eggs, tadpoles, froglets, and adult frogs for year 2012 and 2013 respectively. Results show that between day one and day fourteen of the simulation, survival rates were high because there was no glyphosate in the water body that could kill eggs, tadpoles, froglets, and adult frogs. Figure 4.69 and Figure 4.70 show that between day fourteen and day forty of the simulation, survival was very low (due to high glyphosate concentrations in the water body), but between day forty and day fifty-four of the simulation, survival rates increased steadily as there was steady decrease of glyphosate concentration in the water body (Figure 4.68). Figure 4.69 and Figure 4.70 show that from day fifty-four to two hundred fifty, survival rates were very high as there was no glyphosate in the water body that could kill eggs, tadpoles, froglets, and adult frogs. Figure 4.71 and Figure 4.72 show that no eggs were laid and the number of tadpoles, froglets, and adult frogs were very small due to high glyphosate concentration in the water body. Results suggest that high glyphosate concentration in the water body could affect the population dynamics of frogs.

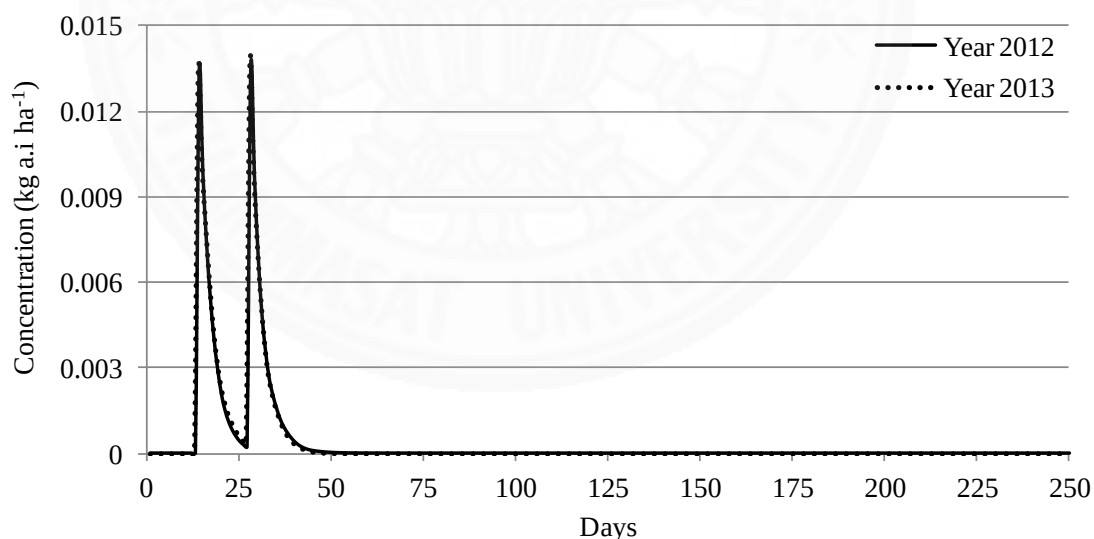


Figure 4.68 Glyphosate concentrations in the water body (using width of no spray zone of zero meters).

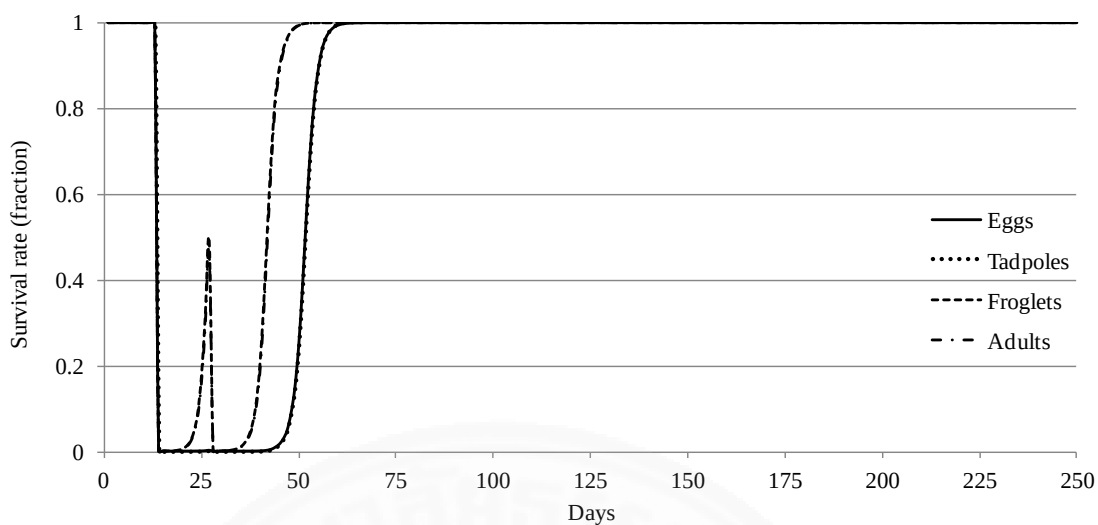


Figure 4.69 Survival rates of frogs in the water body (year 2012, using width of no spray zone of zero meters).

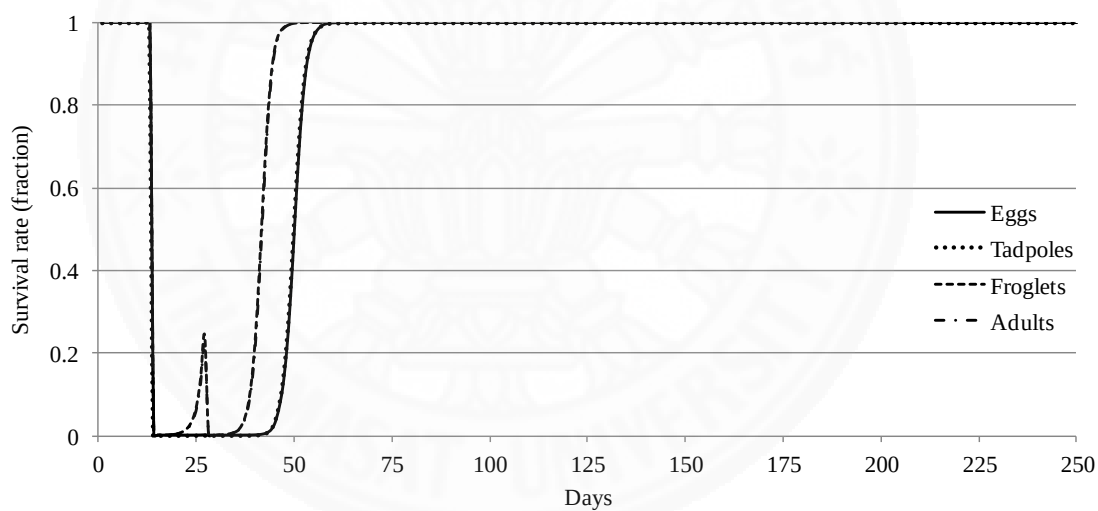


Figure 4.70 Survival rates of frogs in the water body (year 2013, using width of no spray zone of zero meters).

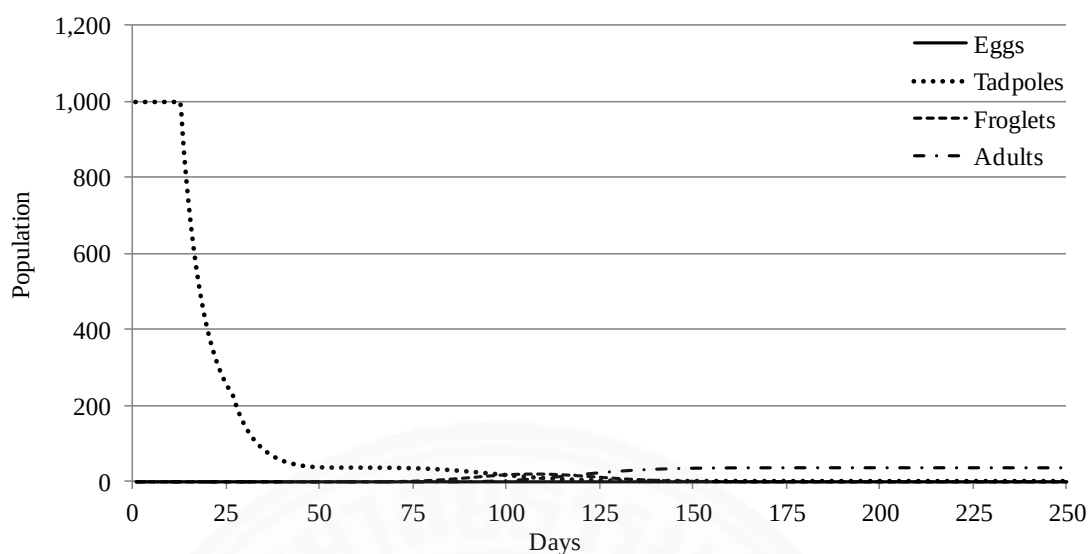


Figure 4.71 Population dynamics of frogs (year 2012, using width of no spray zone of zero meters).

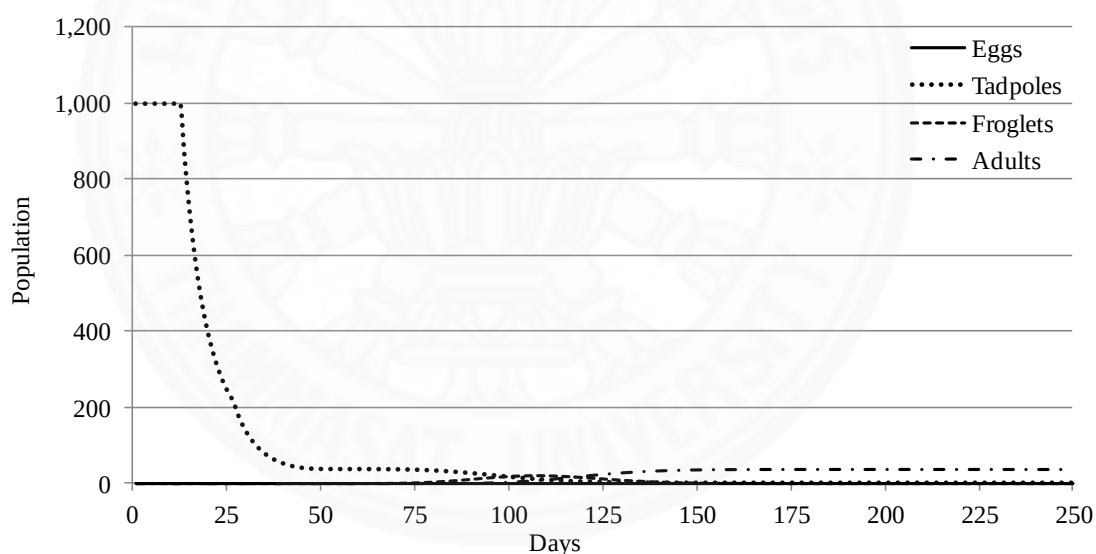


Figure 4.72 Population dynamics of frogs (year 2013, using width of no spray zone of zero meters).

When using width of no spray zone of thirty meters, simulation was performed with two glyphosate applications each at $1.5 \text{ kg a.i ha}^{-1}$ on day fourteen and day twenty-eight of the simulation. Results show that spray drifts were each $6.42 \times 10^{-8} \text{ kg a.i ha}^{-1}$ for the first application (day fourteen) and $6.42 \times 10^{-8} \text{ kg a.i ha}^{-1}$ for the second application (day twenty-eight) for both year 2012 and 2013. Results suggest that when using width of no spray zone of thirty meters, very small amount of glyphosate cross

the agricultural field to the water body. Figure 4.73 shows that the initial glyphosate contamination in the water body was degraded completely between day fourteen and day twenty-eight of the simulation. Then, on day twenty-eight of the simulation, there was another application of glyphosate which resulted to the additional contamination which was later degraded completely by day fifty of the simulation. Figure 4.73 suggests that glyphosate contamination was very little, which in turn, resulted to very high survival rates of eggs, tadpoles, froglets and adult frogs for the entire simulation (Figure 4.74 and Figure 4.75). Results suggest that very little amount of glyphosate contamination in the water body would not kill all eggs, tadpoles, and froglets, and therefore, eggs, tadpoles, and froglets would grow to reach adult frogs (Figure 4.76 and Figure 4.77).

Furthermore, results suggest that width of no spray zone should be taken seriously when working with glyphosate, failure of which could results to the continued decline of frog's population. In this regard, there is a need to employ sustainable agricultural production systems. The main components of the sustainable agriculture are good agricultural practices (GAP) and the integrated pest management (IPM). The integrated pest management employs controlled tactics that are environmentally and ecologically friendly and thus the use of pesticides is only when integrated pest management tactics provide inadequate pest control (Dingha et al., 2013). These controlled tactics include behavioral manipulation methods (Foster & Harris, 1997) and push-pull strategies (Cook et al., 2007).

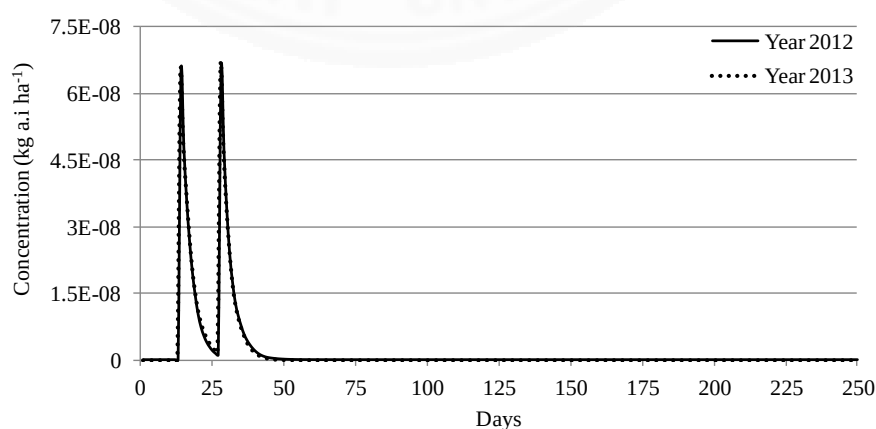


Figure 4.73 Glyphosate concentrations in the water body (using width of no spray zone of thirty meters).

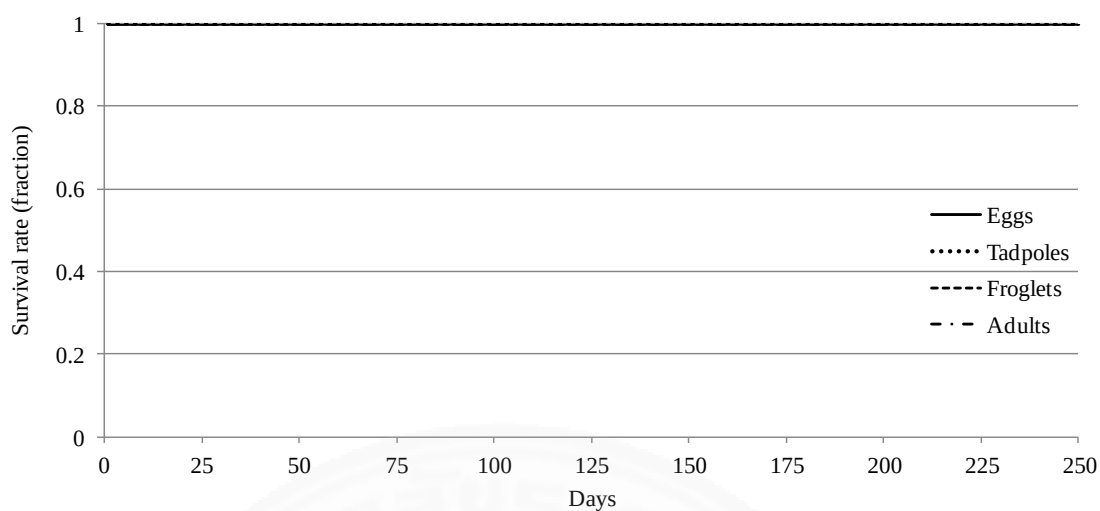


Figure 4.74 Survival rates of frogs in the water body (year 2012, using width of no spray zone of thirty meters).

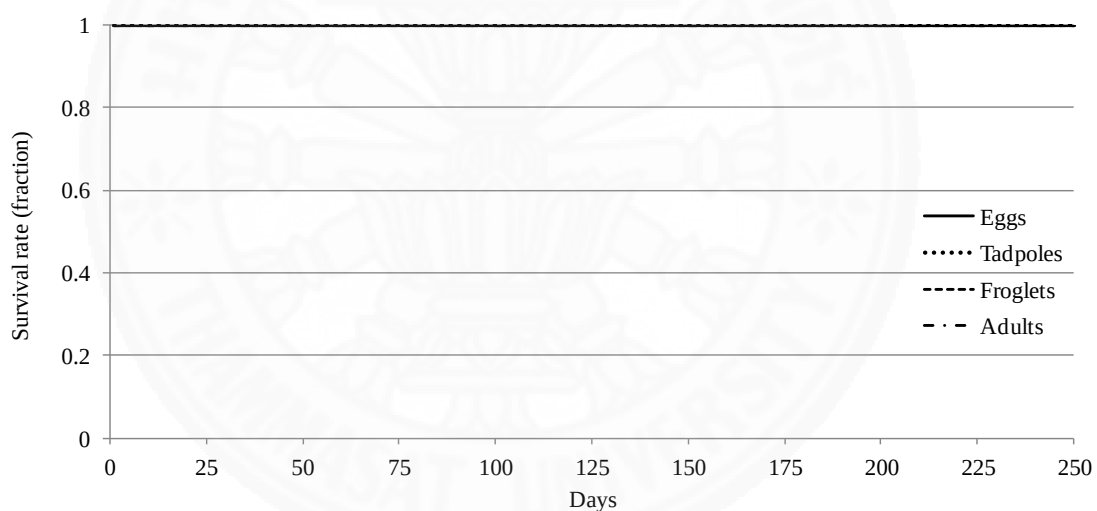


Figure 4.75 Survival rates of frogs in the water body (year 2013, using width of no spray zone of thirty meters).

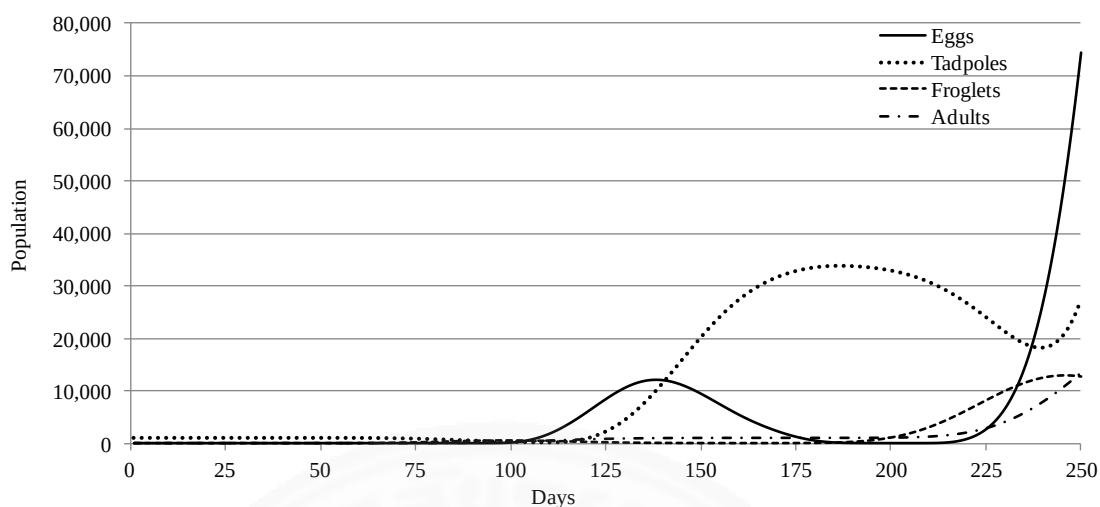


Figure 4.76 Population dynamics of frogs (year 2012, using width of no spray zone of thirty meters).

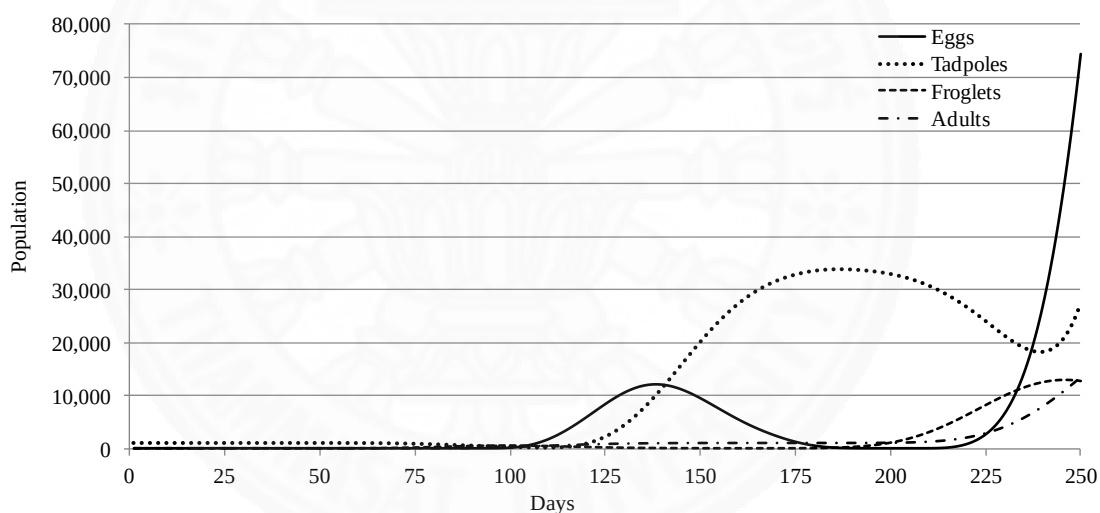


Figure 4.77 Population dynamics of frogs (year 2013, using width of no spray zone of thirty meters).

4.4 Sensitivity analysis

4.4.1 Population versus glyphosate concentration

Sensitivity of population of frogs to changes in the predicted concentrations of glyphosate in the water body and in the topsoil was assessed by varying the concentrations of glyphosate in the water body from zero to 1×10^{-5} kg a.i ha⁻¹ and in the topsoil from zero to 0.8 kg a.i ha⁻¹. These maximum and minimum glyphosate concentrations in the water body and in the topsoil were chosen based on the results

of Figure 4.42 and Figure 4.48 respectively. New uniformly distributed random values of glyphosate concentrations in the water body and in the topsoil were generated before each simulation run. These generated values were used to estimate survival rates at each development stage (i.e. eggs, tadpoles, froglets, and adults). Simulation was repeated for a hundred iterations and maximum number of eggs, tadpoles, froglets, and adults were recorded. Maximum populations attained were plotted against the variations of glyphosate concentrations in the water body and in the topsoil.

Results show the same variation pattern for all life stages. The response to glyphosate concentration in the water is sigmoidal with negligible variation (Figure 4.78 to Figure 4.85) in response caused by the concomitant variation in glyphosate concentration in the topsoil (Figure 4.86 to Figure 4.93). Results show no systematic variation in response to glyphosate concentration in the topsoil. The variation does not show correlation between population of frogs and the concentrations of glyphosate in the topsoil. However, dominant outlying points in Figure 4.78 to Figure 4.85 are not caused by variation in glyphosate concentrations in the water body but rather appear at lower glyphosate concentrations. This suggests that high glyphosate concentrations in the water body could have an impact on population dynamics of frogs.

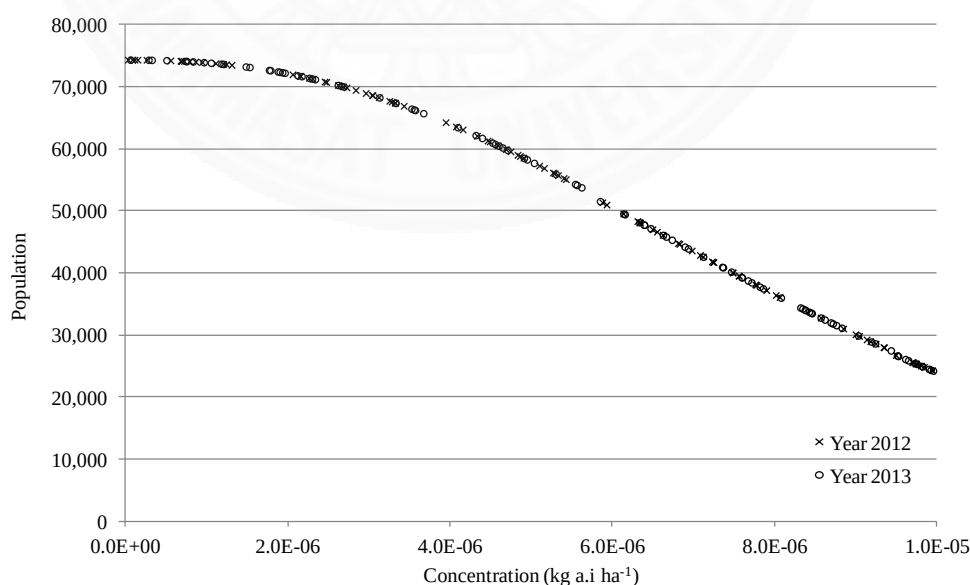


Figure 4.78 Sensitivity of eggs to changes in the glyphosate concentration in the water body (using buffer zone width of thirty meters).

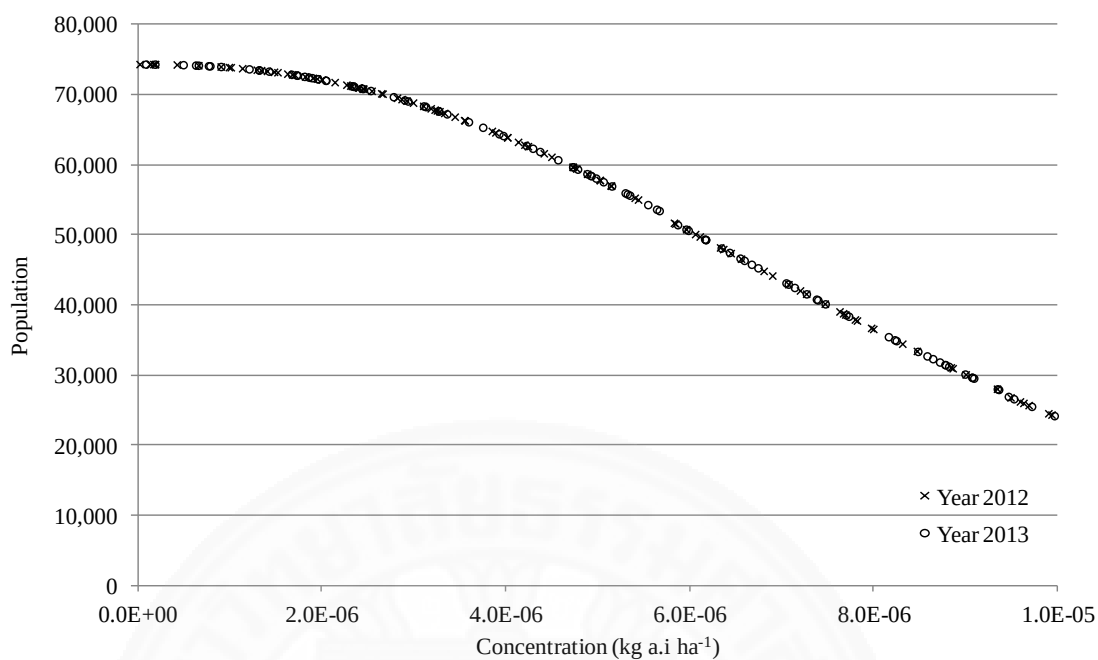


Figure 4.79 Sensitivity of eggs to changes in the glyphosate concentration in the water body (using buffer zone width of zero meters).

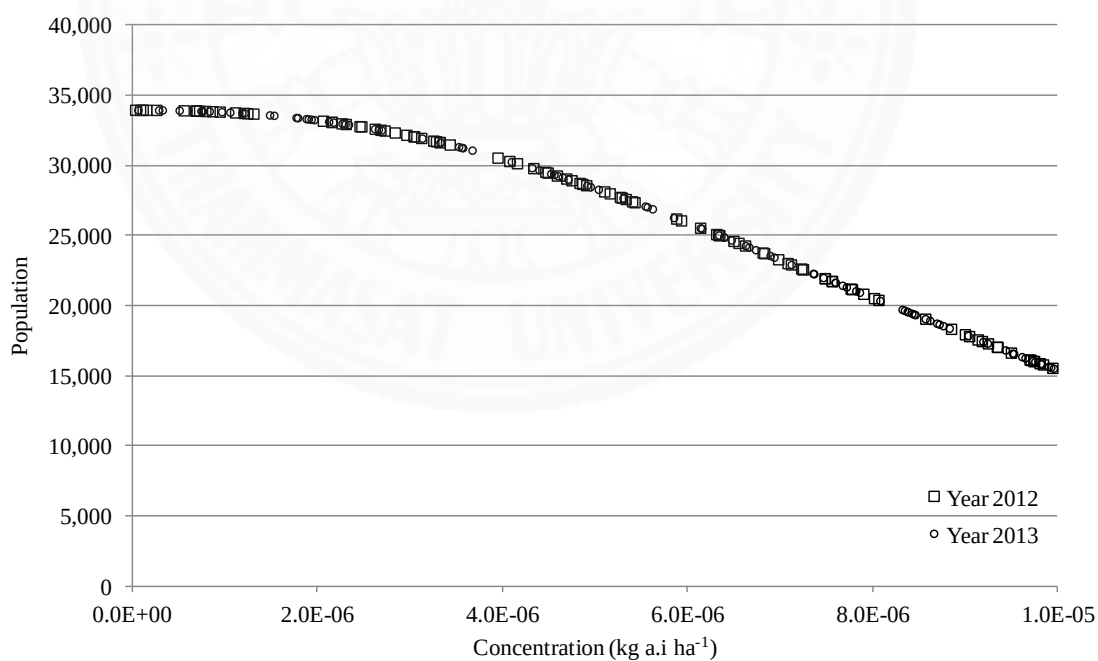


Figure 4.80 Sensitivity of tadpoles to changes in the glyphosate concentration in the water body (using buffer zone width of thirty meters).

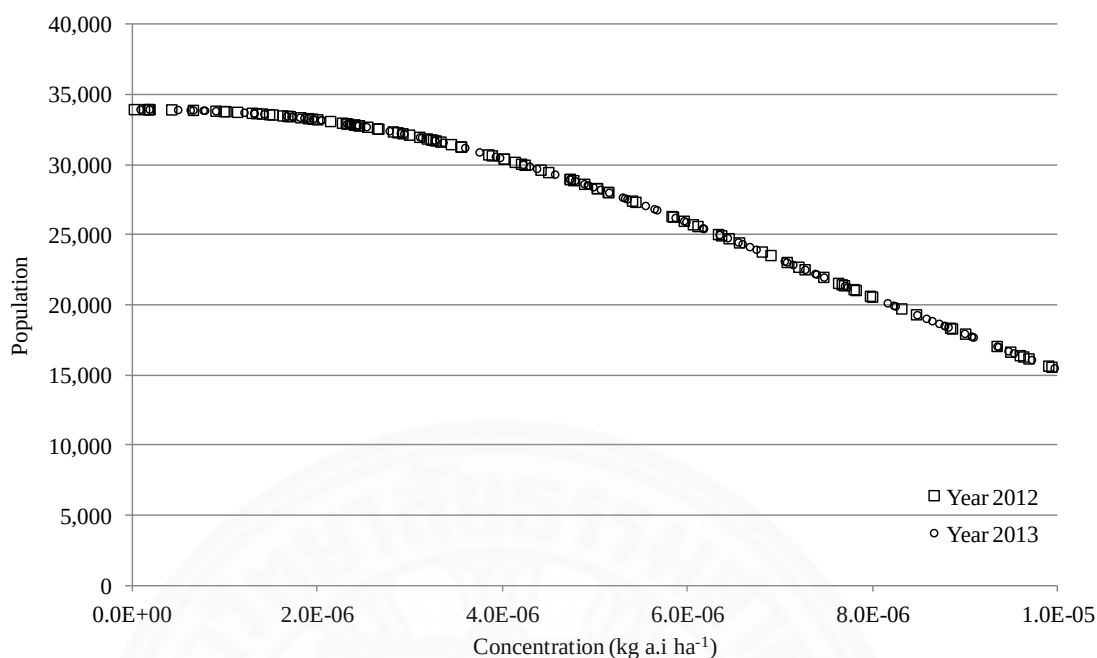


Figure 4.81 Sensitivity of tadpoles to changes in the glyphosate concentration in the water body (using buffer zone width of zero meters).

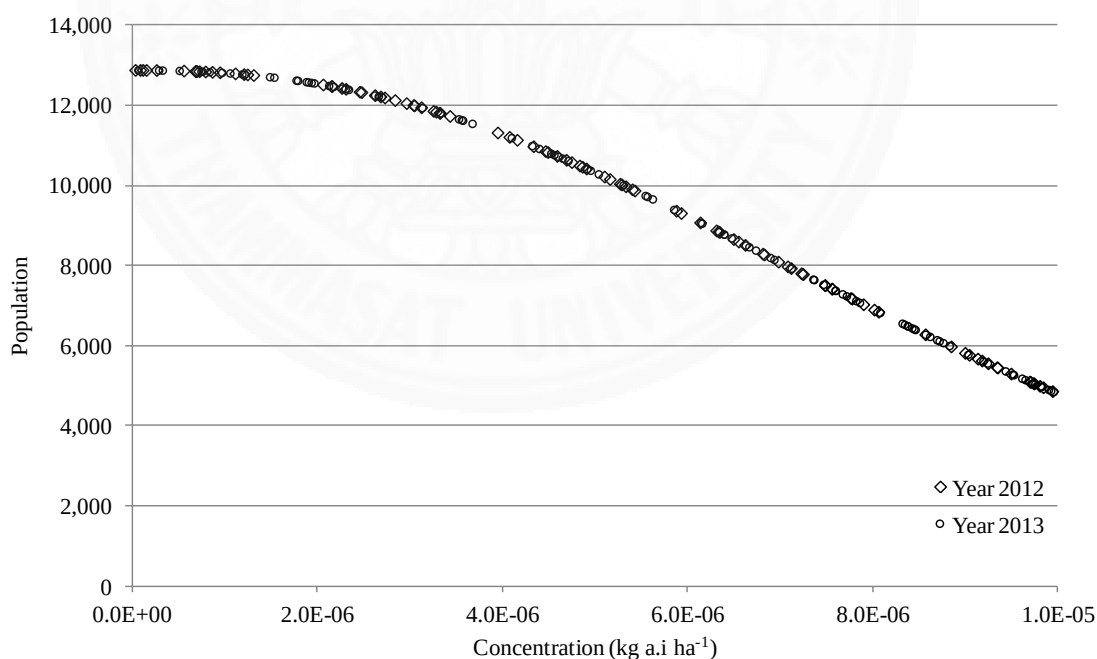


Figure 4.82 Sensitivity of froglets to changes in the glyphosate concentration in the water body (using buffer zone width of thirty meters).

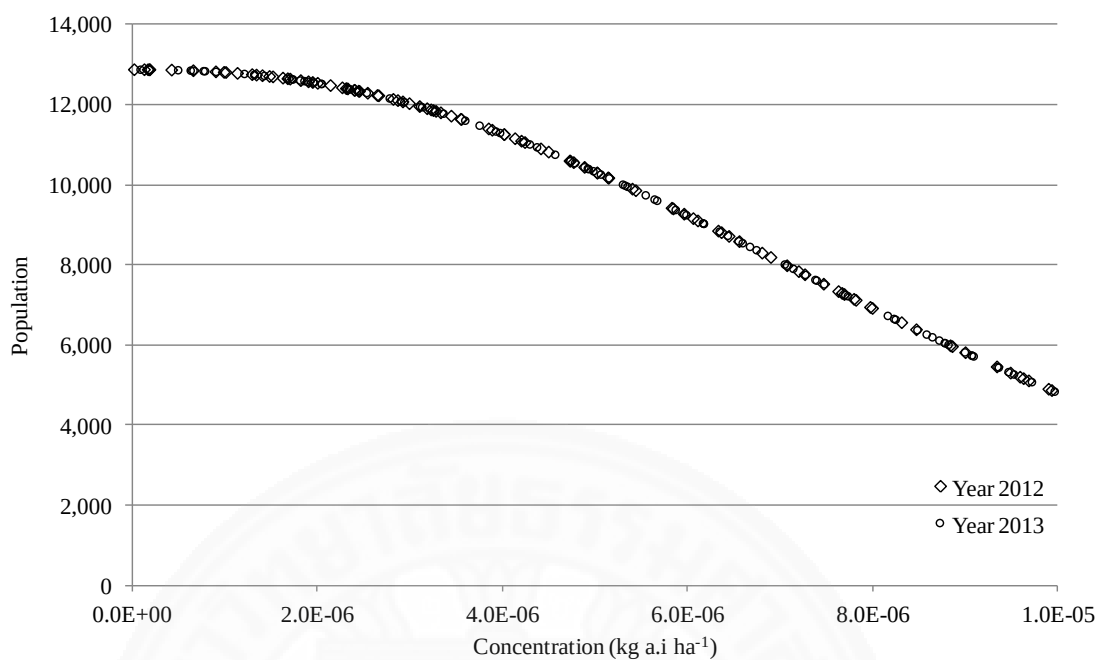


Figure 4.83 Sensitivity of froglets to changes in the glyphosate concentration in the water body (using buffer zone width of zero meters).

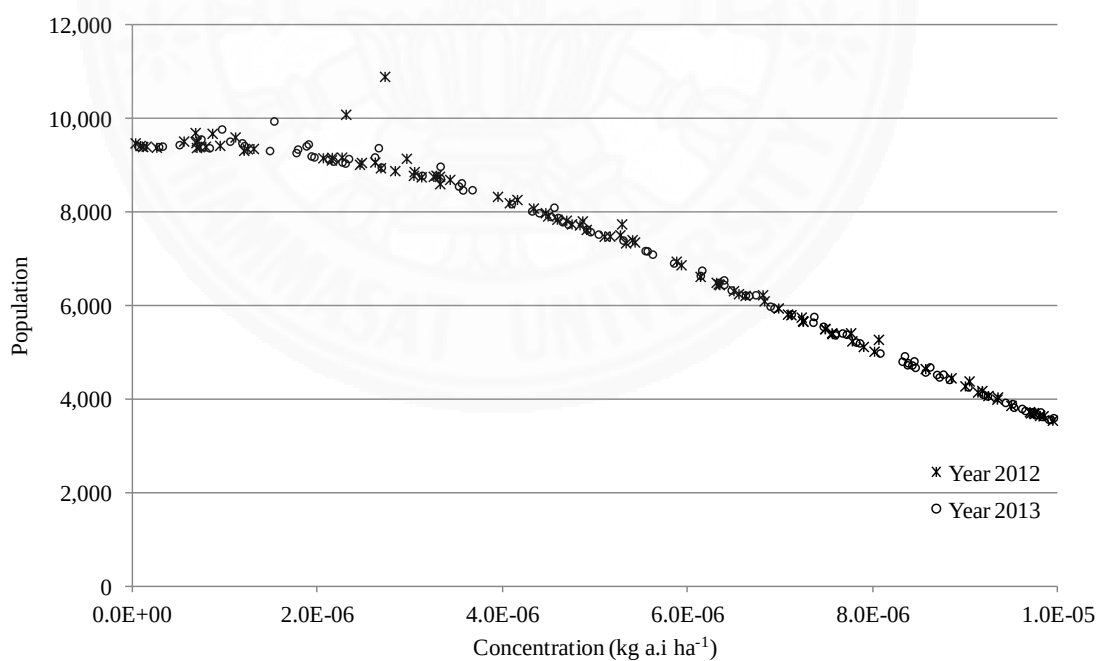


Figure 4.84 Sensitivity of adults to changes in the glyphosate concentration in the water body (using buffer zone width of thirty meters).

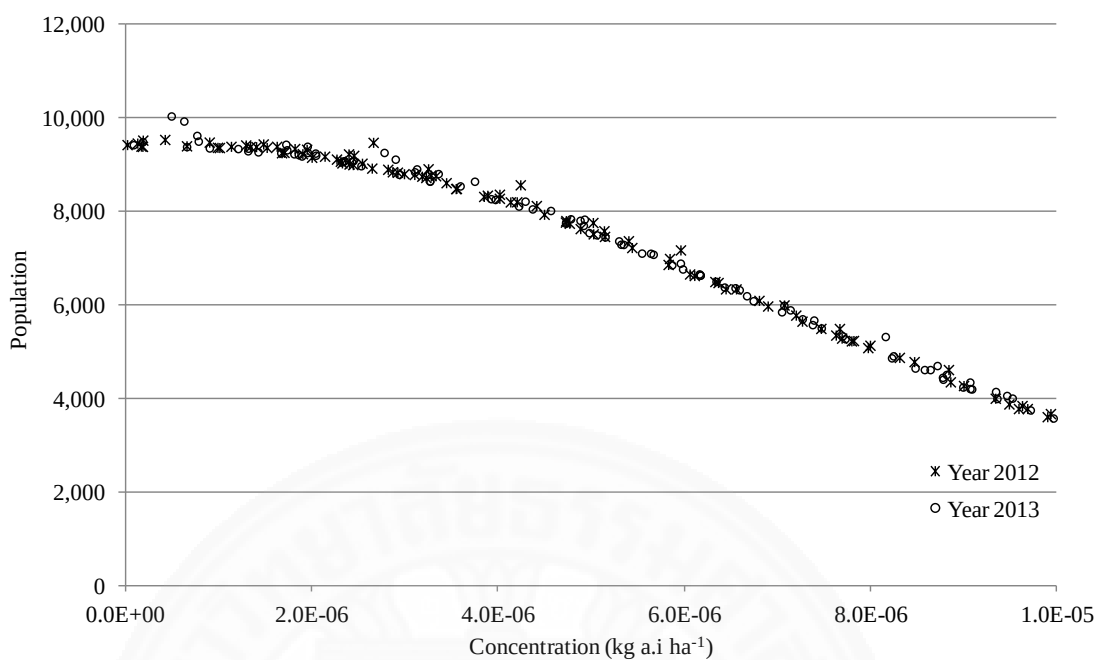


Figure 4.85 Sensitivity of adults to changes in the glyphosate concentration in the water body (using buffer zone width of zero meters).

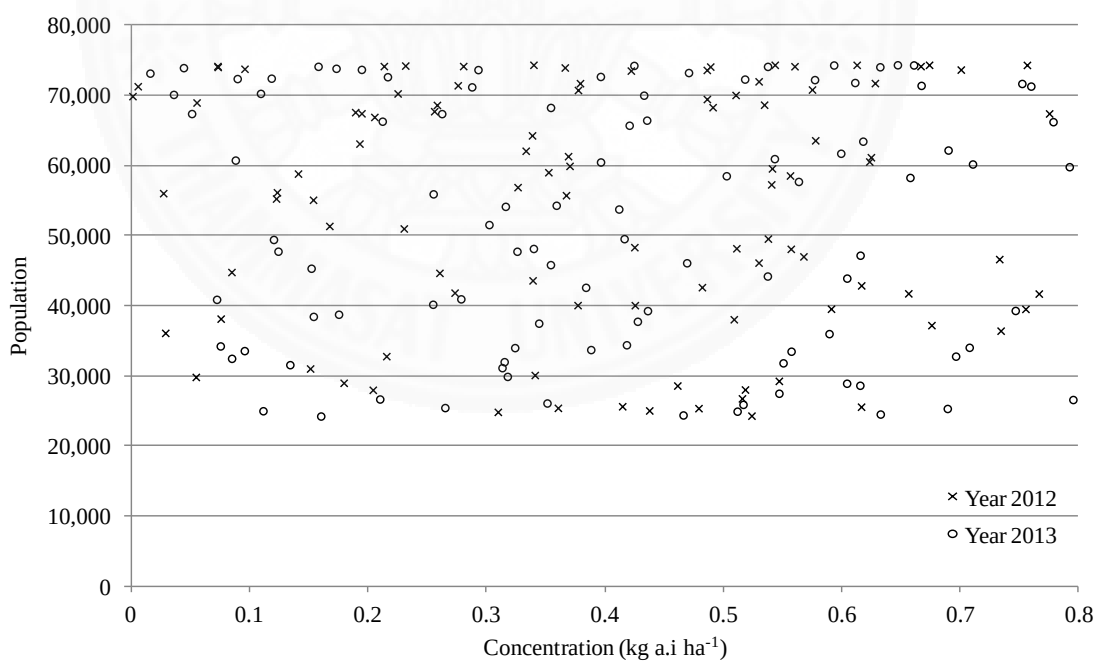


Figure 4.86 Sensitivity of eggs to changes in the glyphosate concentration in the topsoil (using buffer zone width of thirty meters).

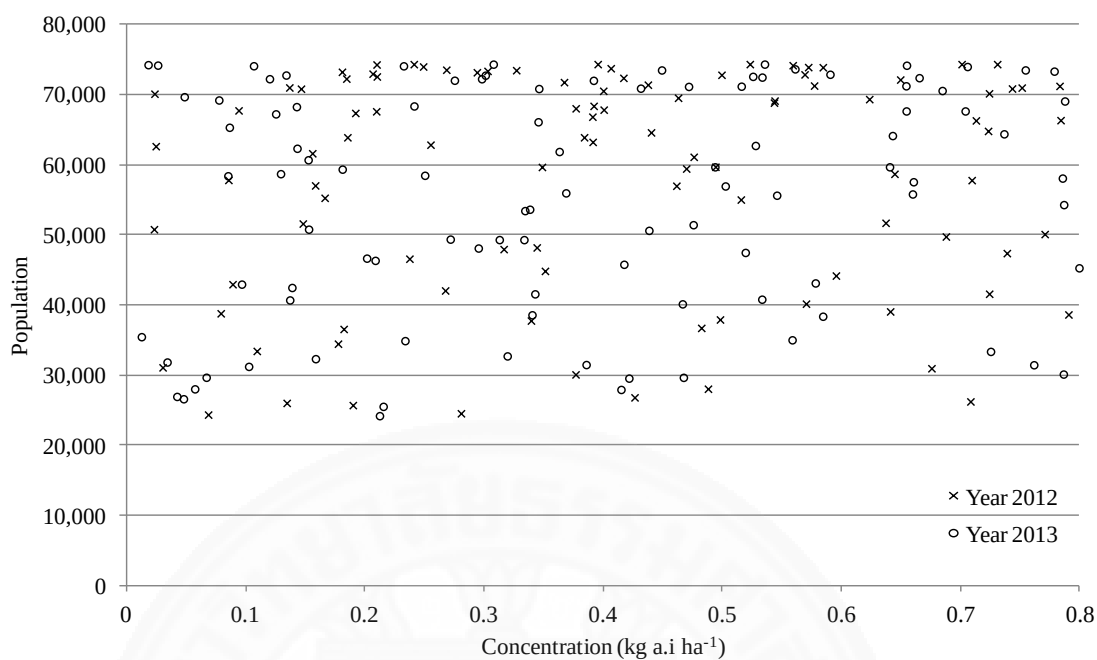


Figure 4.87 Sensitivity of eggs to changes in the glyphosate concentration in the topsoil (using buffer zone width of zero meters).

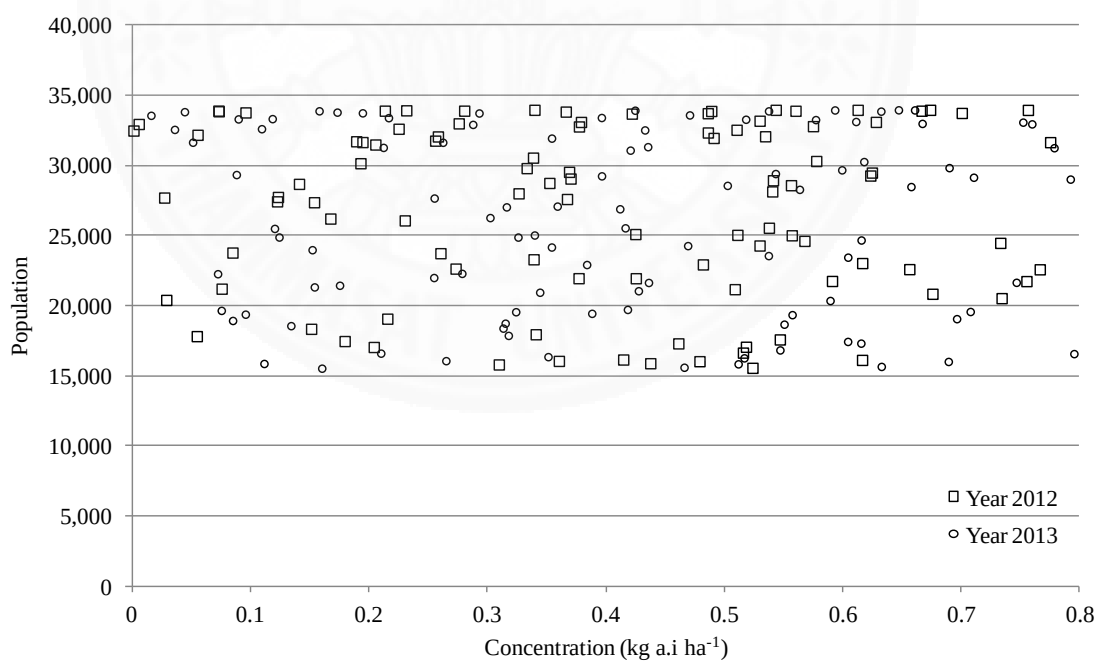


Figure 4.88 Sensitivity of tadpoles to changes in the glyphosate concentration in the topsoil (using buffer zone width of thirty meters).

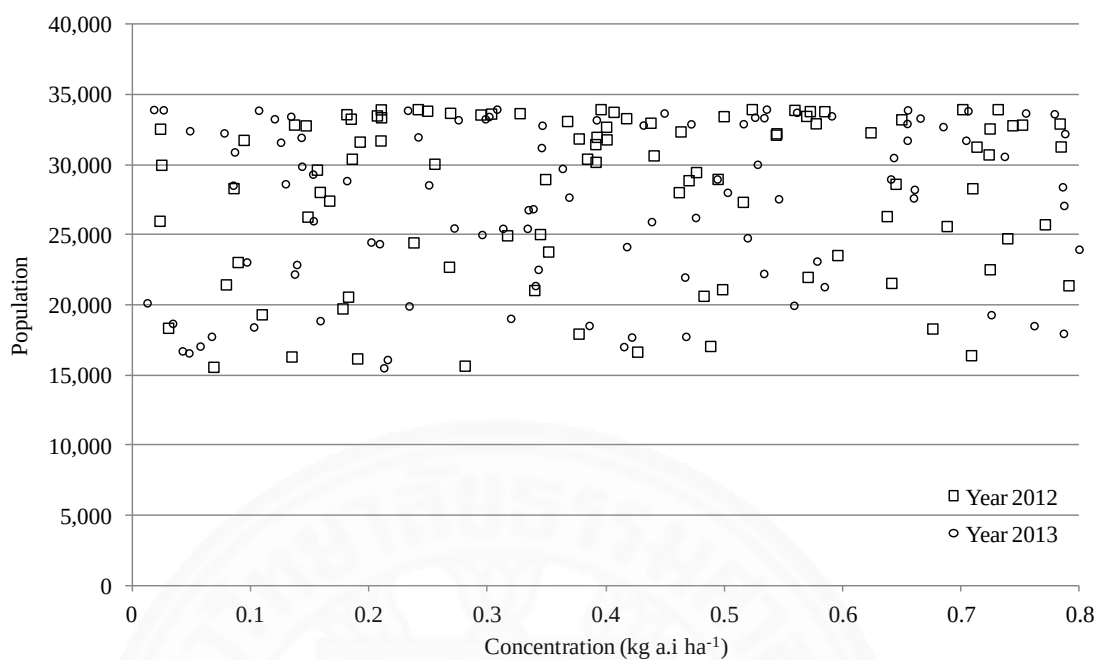


Figure 4.89 Sensitivity of tadpoles to changes in the glyphosate concentration in the topsoil (using buffer zone width of zero meters).

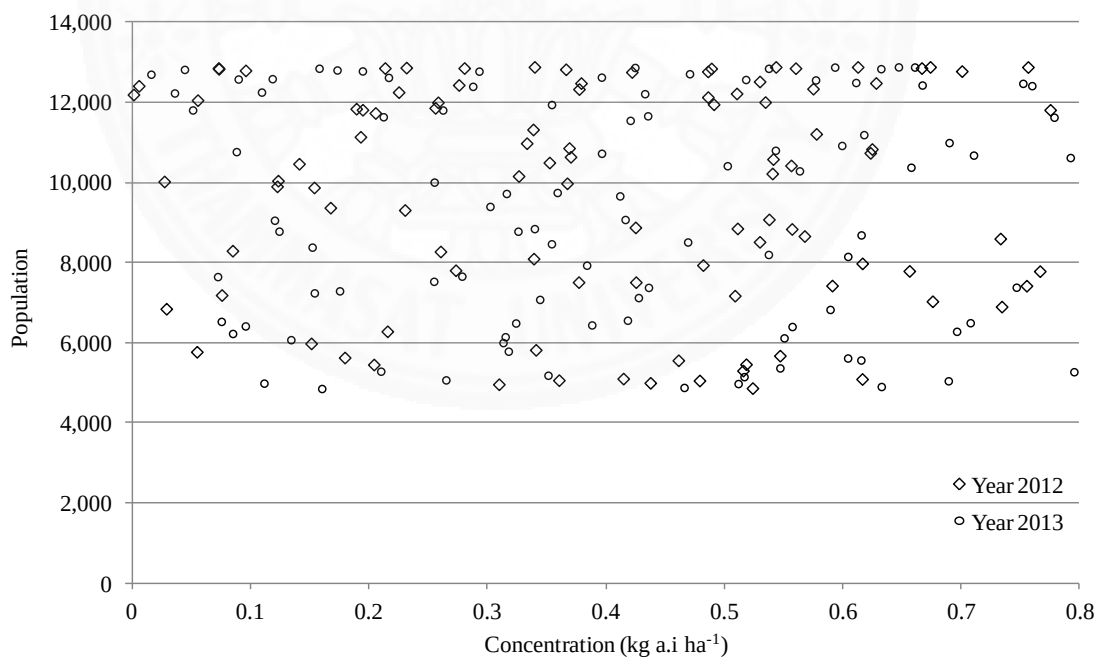


Figure 4.90 Sensitivity of froglets to changes in the glyphosate concentration in the topsoil (using buffer zone width of thirty meters).

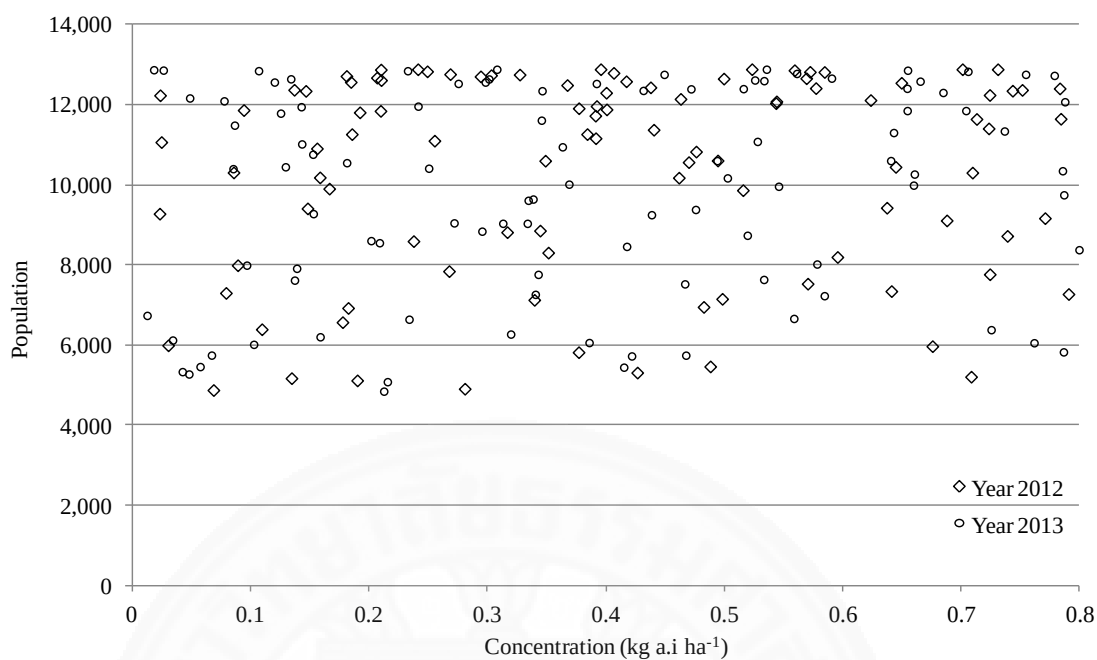


Figure 4.91 Sensitivity of froglets to changes in the glyphosate concentration in the topsoil (using buffer zone width of zero meters).

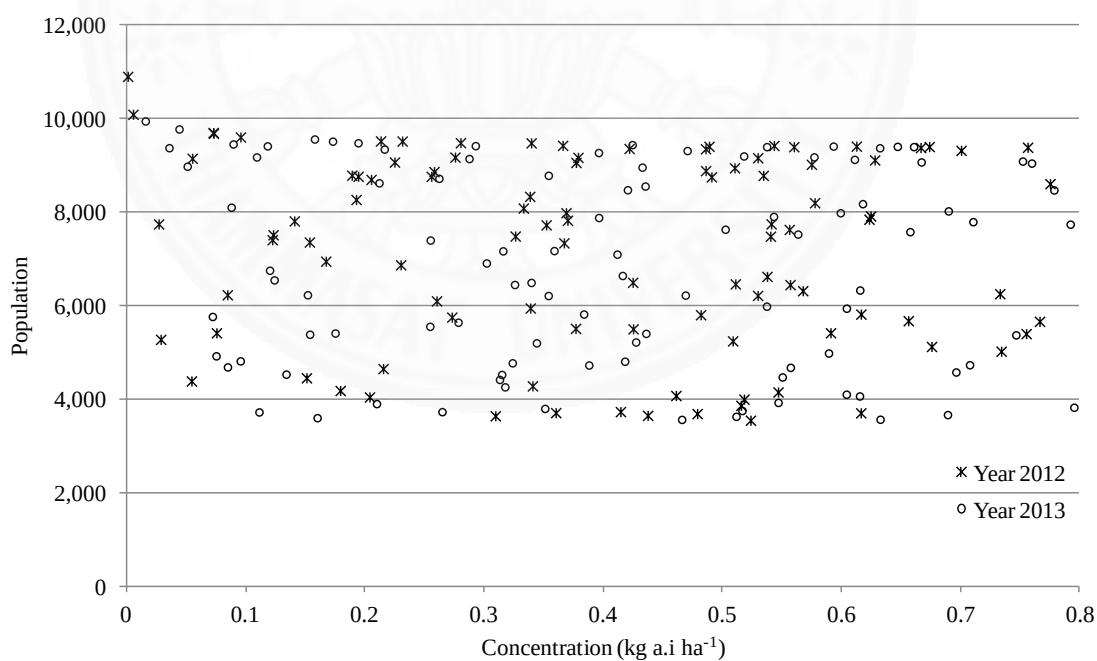


Figure 4.92 Sensitivity of adults to changes in the glyphosate concentration in the topsoil (using buffer zone width of thirty meters).

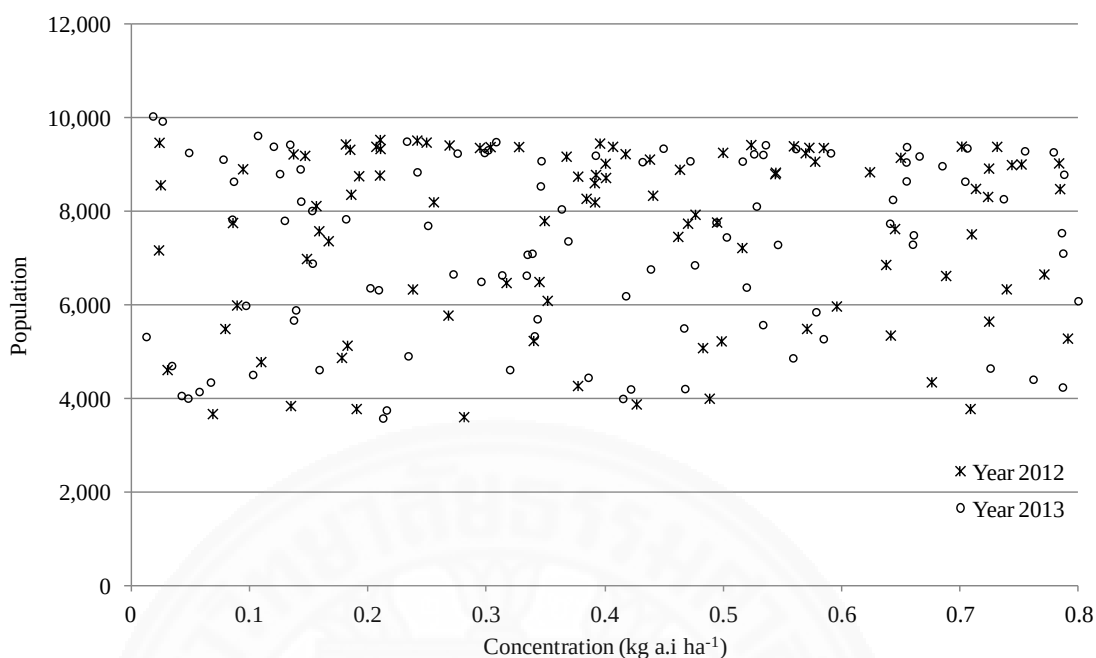


Figure 4.93 Sensitivity of adults to changes in the glyphosate concentration in the topsoil (using buffer zone width of zero meters).

4.4.2 Population versus eggs laid per female

The sensitivity of population of frogs to changes in the number of eggs laid per female was assessed using the minimum number of sixty-six eggs while the maximum was two hundred sixty-eight eggs (Hero & Roberts, 2004; Tyler & Knight, 2011). This sensitivity analysis was performed using buffer zone width of thirty meters only (because when using buffer zone width of zero meters, most tadpoles died and did not grow to adults due to high mortality rates, therefore, no eggs were laid).

Results show that there is a correlation between the population of frogs and the number of eggs laid per female (Figure 4.94 to Figure 4.97). Results show that population increased when the number of eggs laid per female increased. The finding suggests that if this species of frog lay more number of eggs per female, it could help to increase their population.

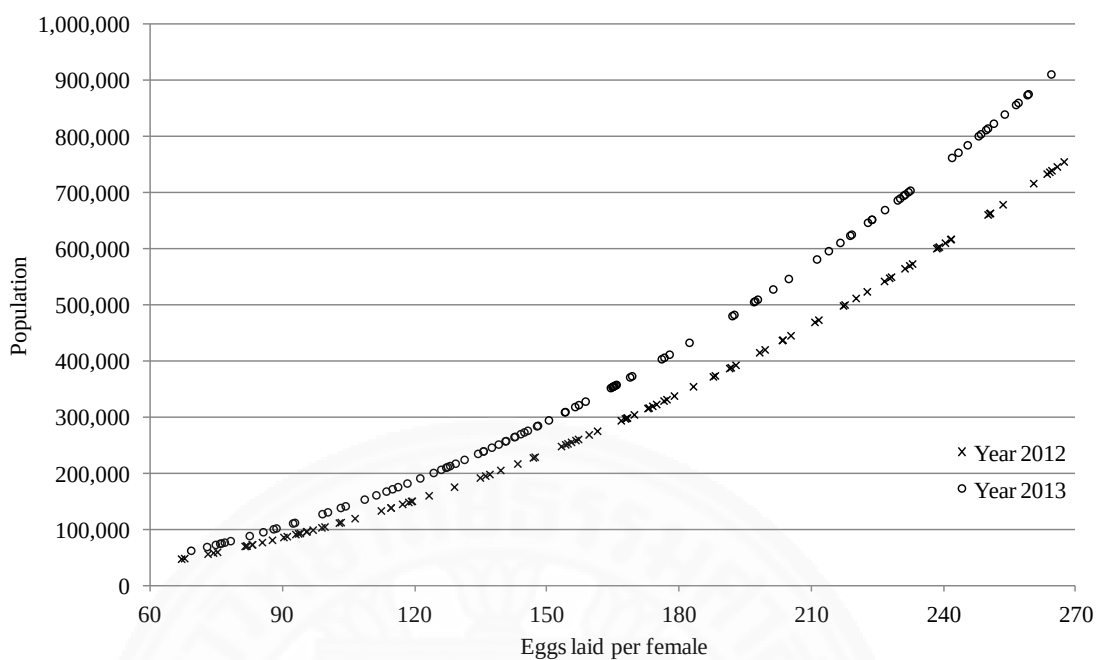


Figure 4.94 Sensitivity of eggs to changes in the number of eggs laid per female (using buffer zone width of thirty meters).

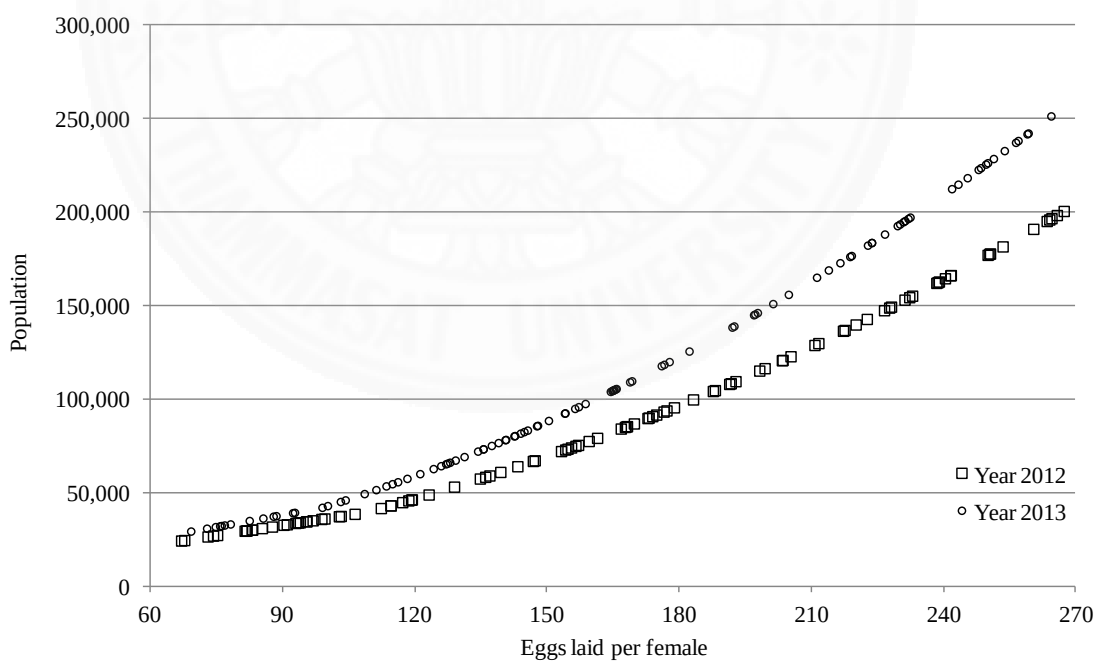


Figure 4.95 Sensitivity of tadpoles to changes in the number of eggs laid per female (using buffer zone width of thirty meters).

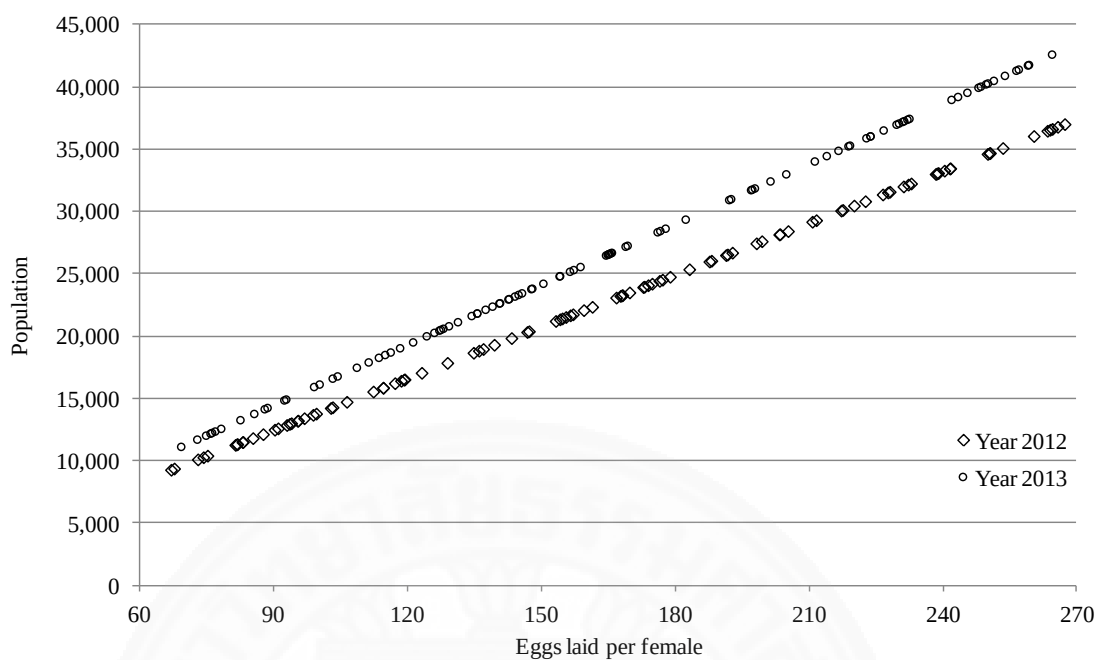


Figure 4.96 Sensitivity of froglets to changes in the number of eggs laid per female (using buffer zone width of thirty meters).

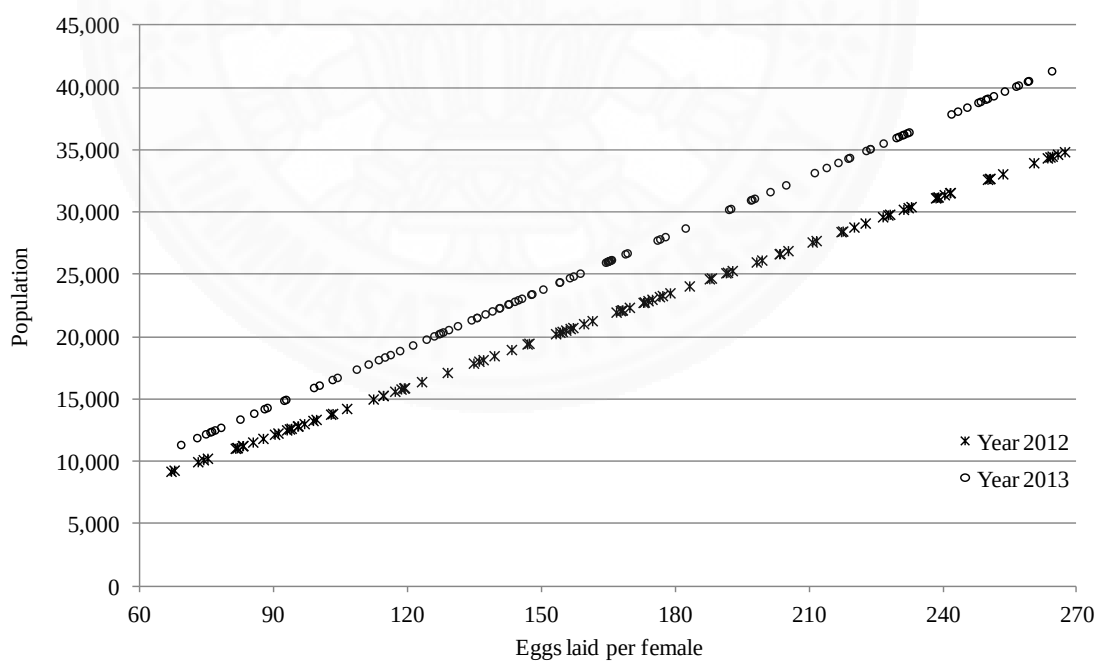


Figure 4.97 Sensitivity of adults to changes in the number of eggs laid per female (using buffer zone width of thirty meters).

4.4.3 Washoff versus rainfall

Sensitivity of washoff from the crop/leaf surfaces to changes in the amount of rainfall was assessed using the minimum amount of rainfall of zero millimeters while the maximum was one hundred millimeters. Results show that there is a correlation between the amount of glyphosate removed from the crop/leaf surfaces (by washoff) and the amount of rainfall (Figure 4.98 to Figure 4.99). Results suggest that the amount of glyphosate removed from the crop/leaf surfaces by washoff is directly proportional to the amount of rainfall. This means that the higher the amount of rainfall, the higher the amount of washoff. The finding suggests that the use of glyphosate should be avoided during rainy seasons.

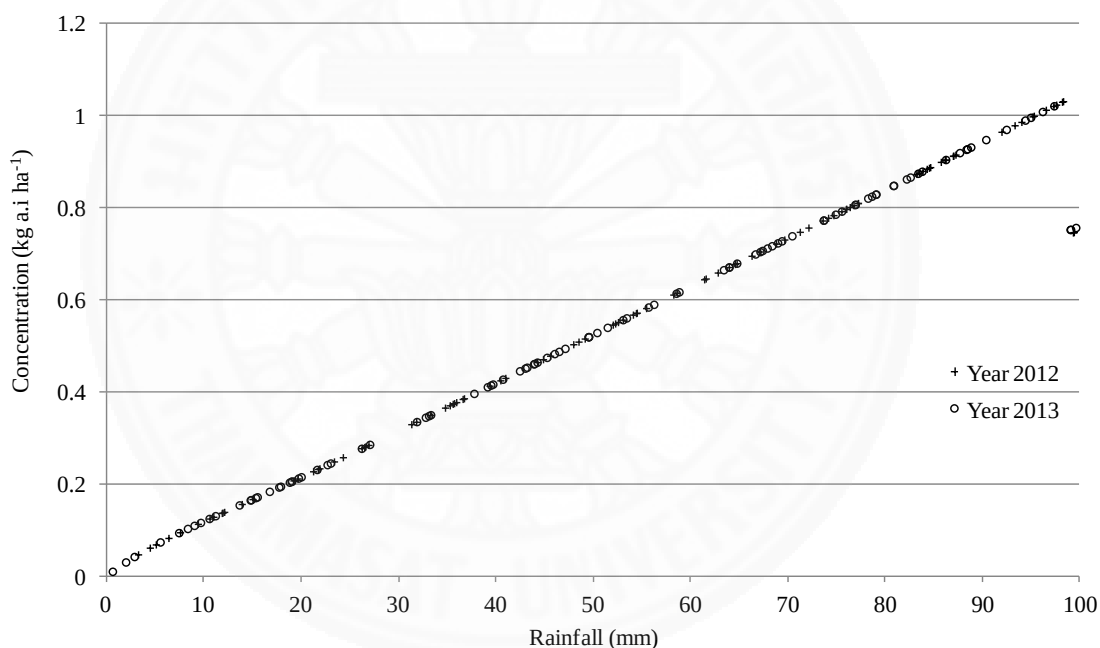


Figure 4.98 Sensitivity of washoff to changes in the amount of rainfall (using buffer zone width of thirty meters).

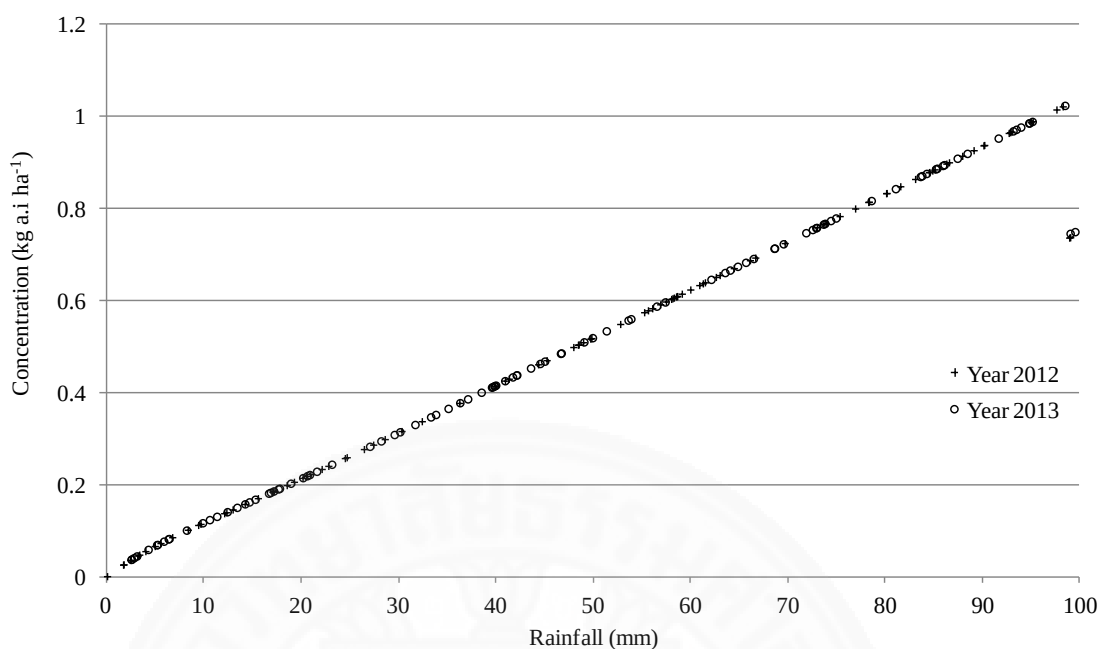


Figure 4.99 Sensitivity of washoff to changes in the amount of rainfall (using buffer zone width of zero meters).

4.4.4 Runoff versus rainfall

Sensitivity of runoff from the topsoil to changes in the amount of rainfall was assessed using the minimum amount of rainfall of zero millimeters while the maximum was one hundred millimeters. Results show that there is a correlation between the amount of glyphosate removed from the topsoil (by runoff) and the amount of rainfall (Figure 4.100 to Figure 4.101). Results suggest that the higher the amount of rainfall, the higher the amount of runoff from the topsoil. The finding also suggests that the use of glyphosate should be avoided during rainy seasons.

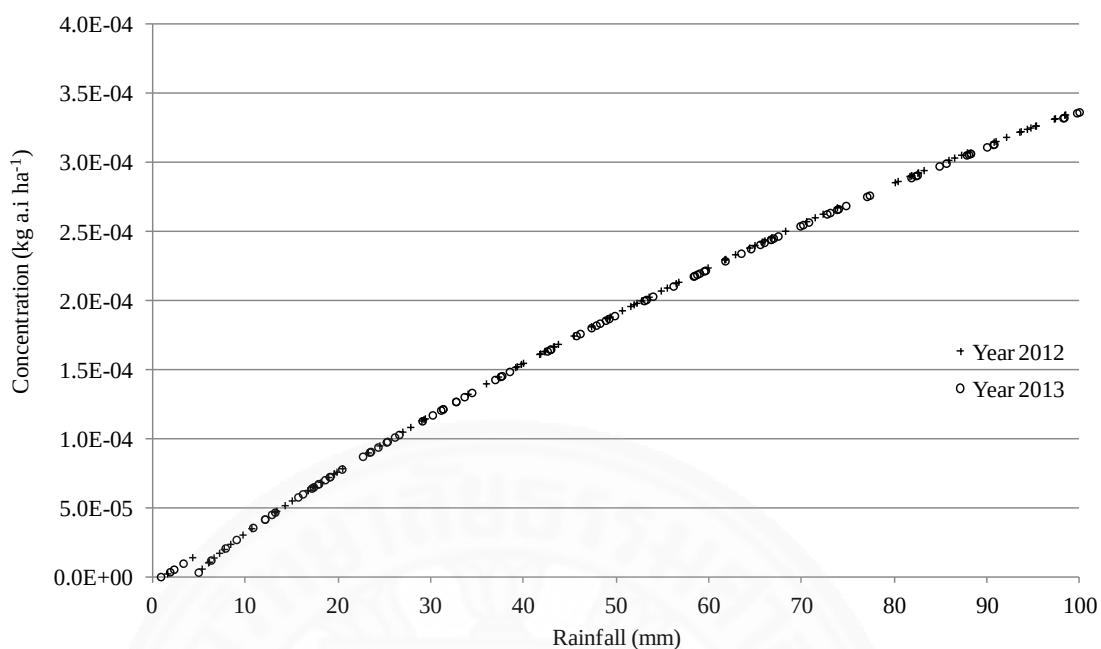


Figure 4.100 Sensitivity of runoff to changes in the amount of rainfall (using buffer zone width of thirty meters).

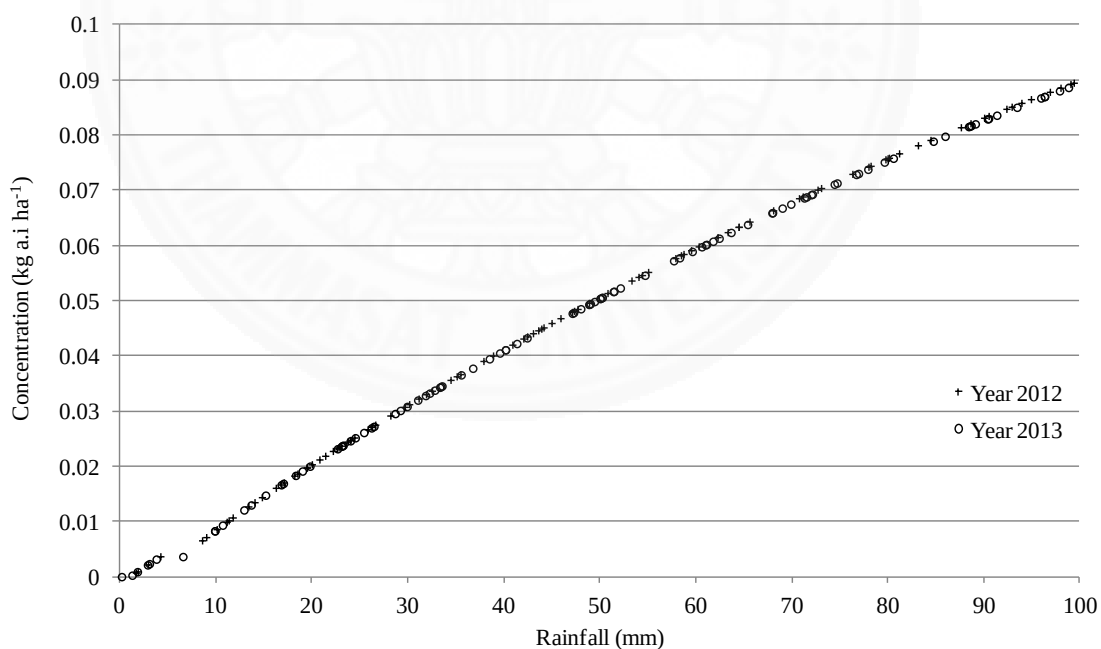


Figure 4.101 Sensitivity of runoff to changes in the amount of rainfall (using buffer zone width of zero meters).

4.4.5 Macropore flow versus rainfall

Sensitivity of macropore flow from the topsoil to changes in the amount of rainfall was assessed using the minimum amount of rainfall of zero millimeters while the maximum was one hundred millimeters. Results show that there is a correlation between the amount of glyphosate removed from the topsoil (by macropore flow) and the amount of rainfall (Figure 4.102 to Figure 4.103). Results show that macropore flow increases exponentially when the amount of rainfall increases. Results also suggest that the higher the amount of rainfall, the higher the amount of runoff from the topsoil.

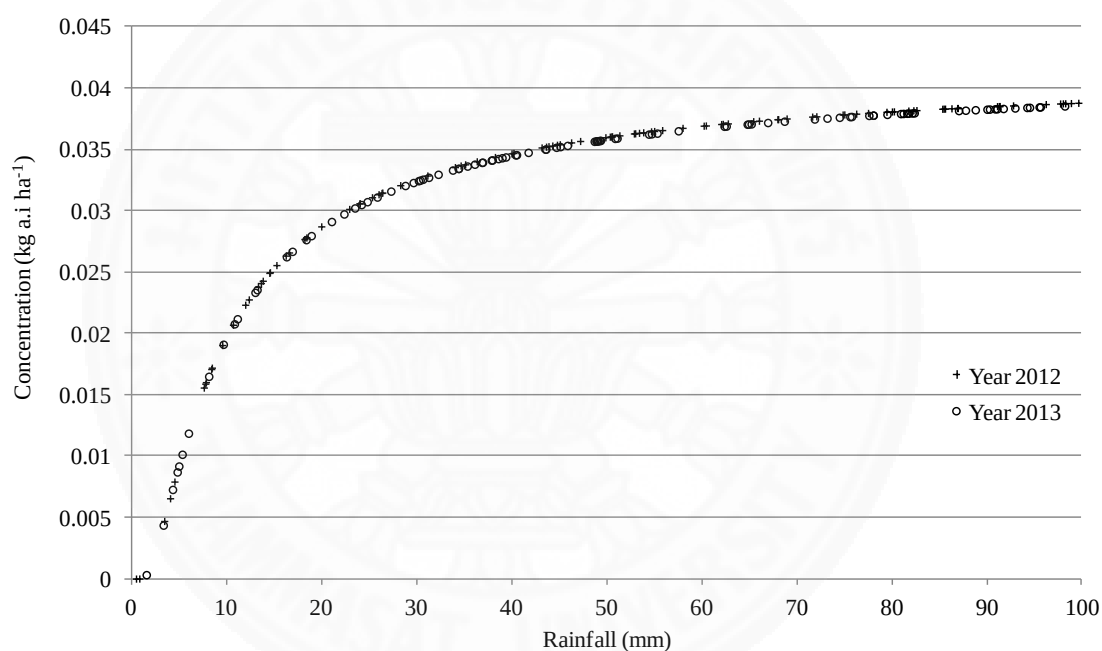


Figure 4.102 Sensitivity of macropore flow to changes in the amount of rainfall (using buffer zone width of thirty meters).

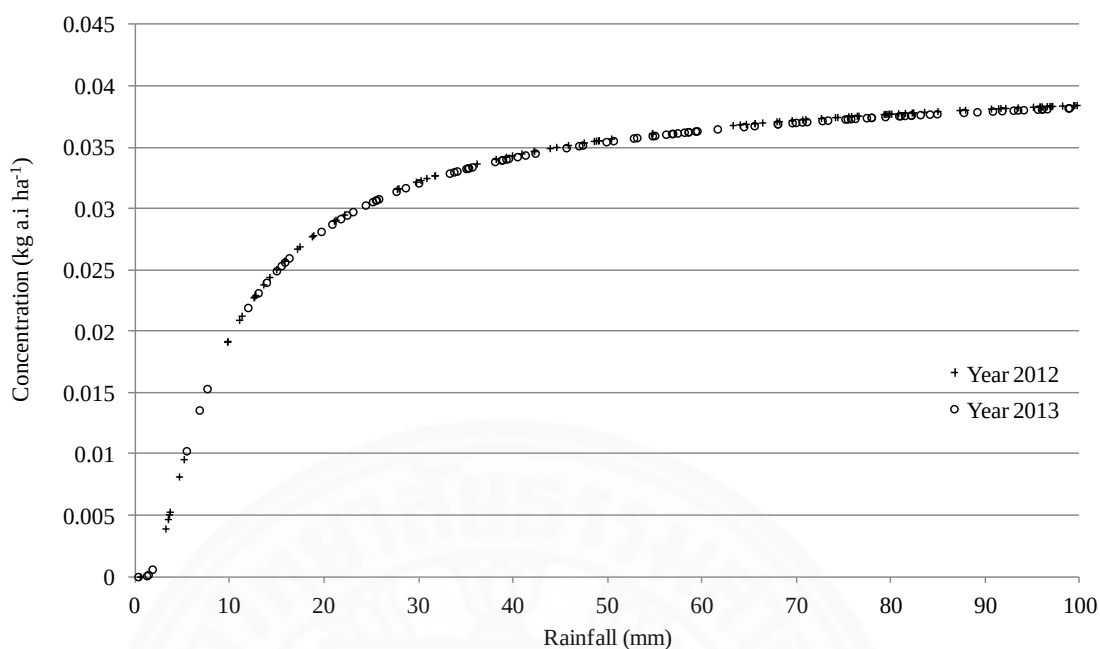


Figure 4.103 Sensitivity of macropore flow to changes in the amount of rainfall (using buffer zone width of zero meters).

4.4.6 Runoff versus buffer zone width

Sensitivity of runoff from the topsoil to changes in the buffer zone width was assessed using the minimum buffer zone width of zero meters while the maximum was one hundred meters. Results show that there is a correlation between the amount of glyphosate removed from the topsoil (by runoff) and the buffer zone width (Figure 4.104 to Figure 4.105). Results show that runoff decreases exponentially when the buffer zone width increases. Results suggest that the higher the buffer zone width, the lower the amount of runoff from the topsoil.

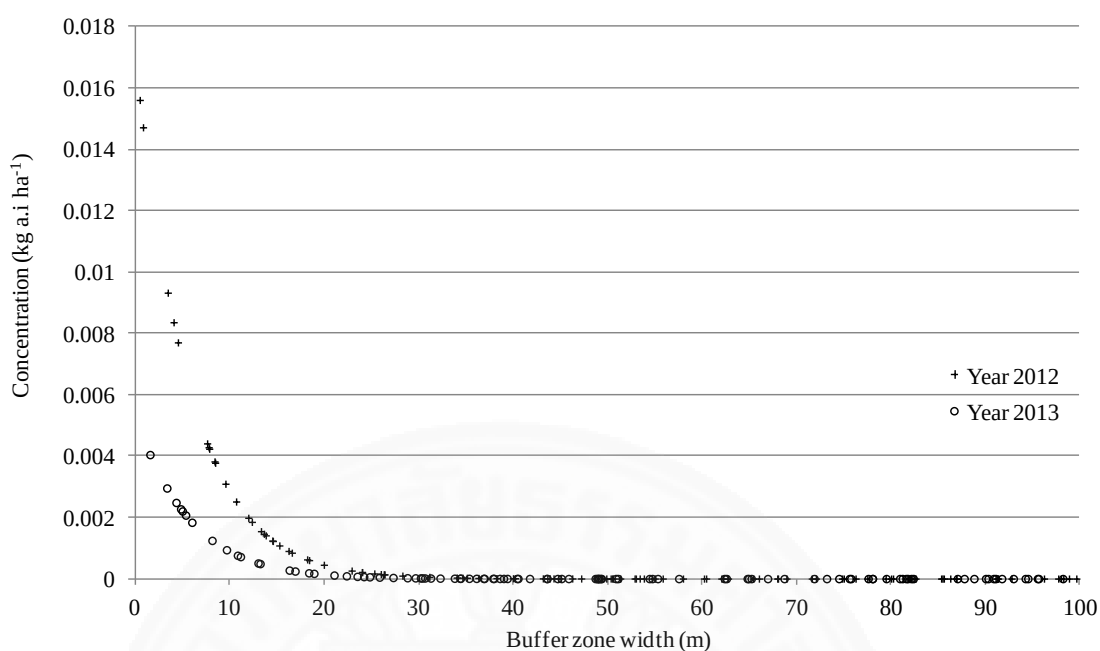


Figure 4.104 Sensitivity of runoff to changes in the buffer zone width (using buffer zone width of thirty meters).

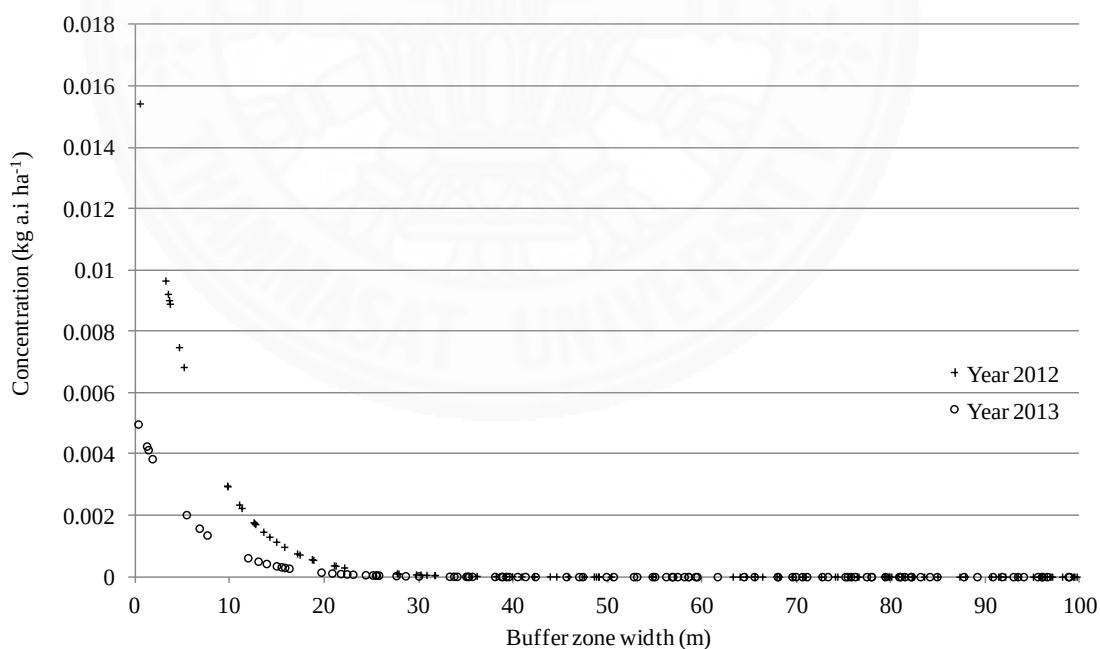


Figure 4.105 Sensitivity of runoff to changes in the buffer zone width (using buffer zone width of zero meters).

4.4.7 Population versus buffer zone (effects of spray drifts)

The sensitivity of population of frogs to changes in the buffer zone width was assessed using the minimum buffer zone width of zero meters while the maximum was thirty meters. Results show that there is a correlation between the population of frogs and the buffer zone width (Figure 4.106 to Figure 4.109). Results show that when buffer zone width increases, population increases exponentially; mainly because spray drifts decreases. However, at seventeen meters buffer zone to thirty meters, there is no noticeable change in the population of frogs. This suggests that a buffer zone of at least seventeen meters should be enforced around the agricultural fields to prevent spray drifts of glyphosate from affecting frogs in the nearby areas when working with cereal crops (e.g. maize).

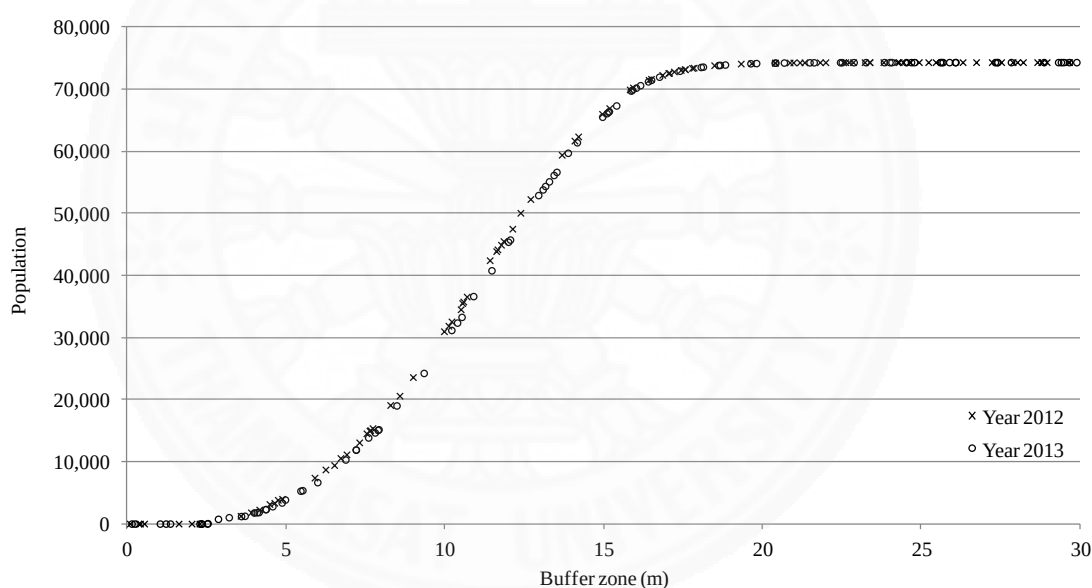


Figure 4.106 Sensitivity of eggs to changes in the buffer zone width.

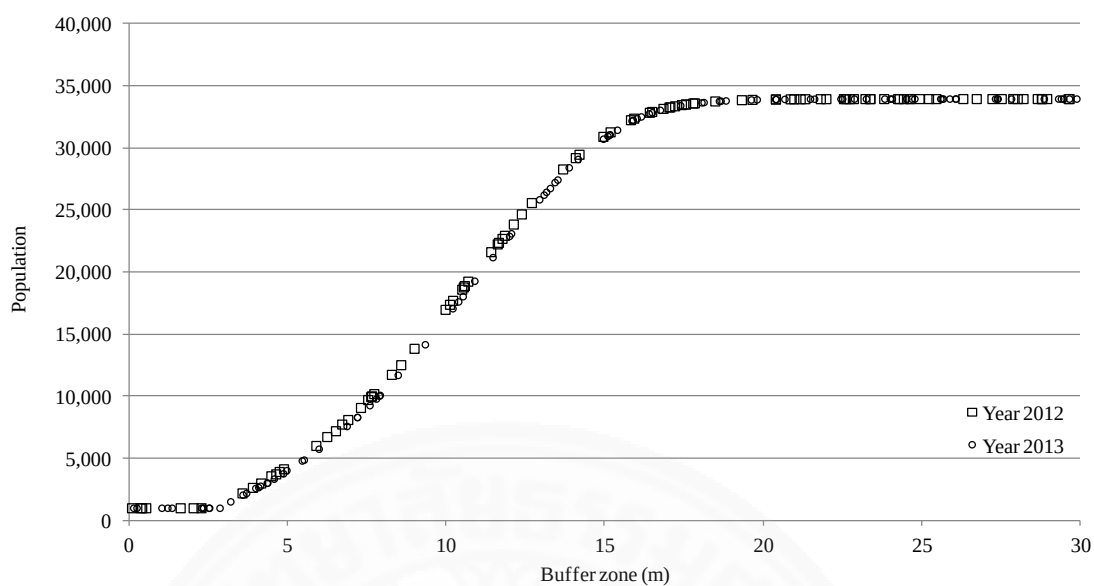


Figure 4.107 Sensitivity of tadpoles to changes in the buffer zone width.

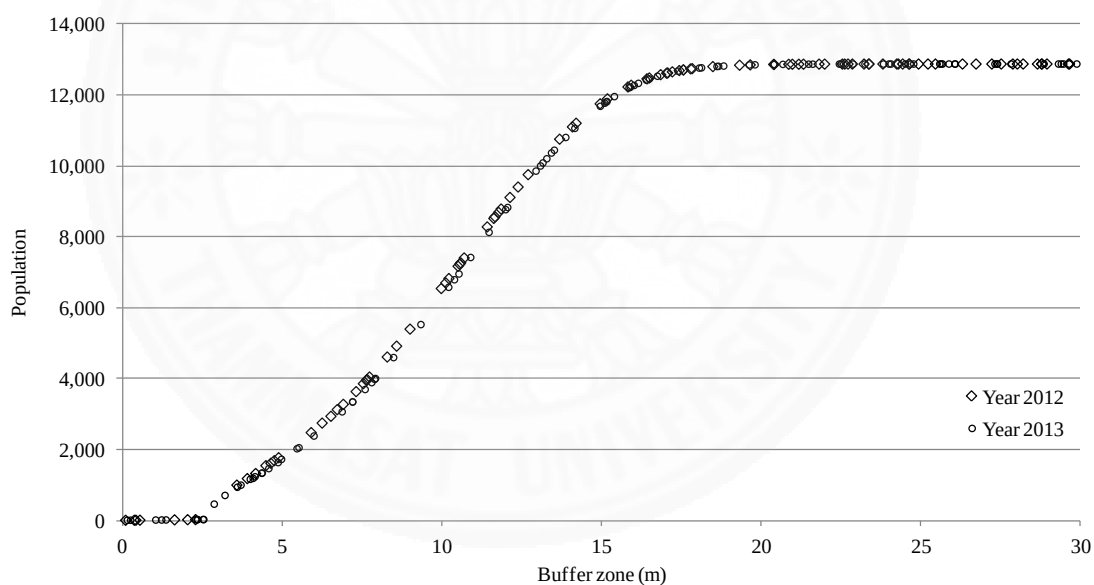


Figure 4.108 Sensitivity of froglets to changes in the buffer zone width.

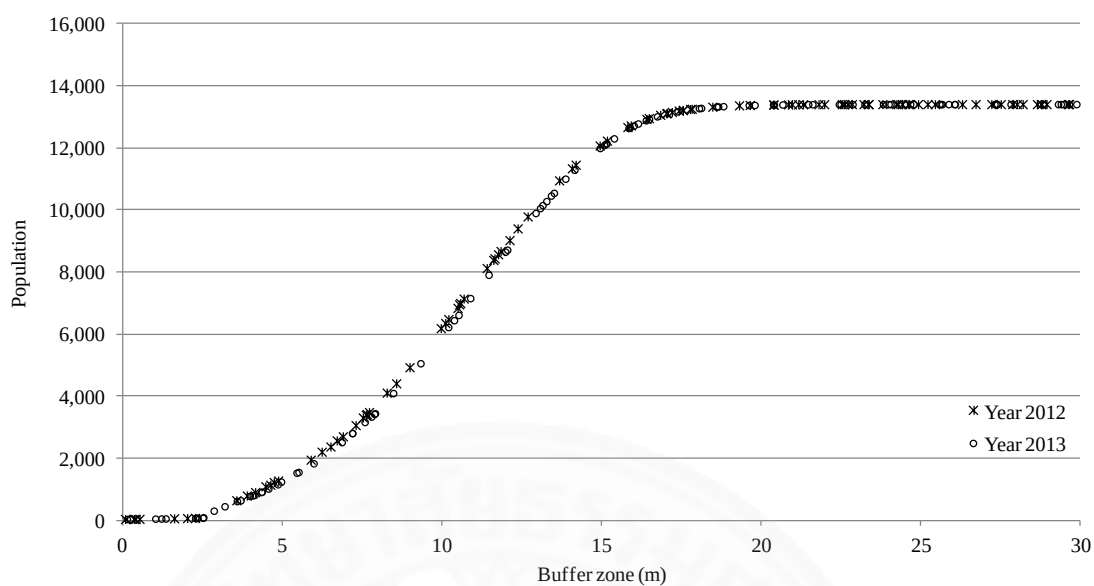


Figure 4.109 Sensitivity of adults to changes in the buffer zone width.

CHAPTER 5

CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

This study surveyed how aware people in Tanzania, are of pesticides and their associated health effects. Results reveal that respondents employ poor pesticide management practices which could severely deteriorate the environmental health and quality, as well as the human health, and the health of other non-target species. Poor pesticide disposal practices and the nonuse of protective gears which could be due to either unavailability or high cost of protective equipment, are also some of the main concerns that need immediate attention. Respondents were aware that pesticides could kill them and damage the environment when mishandled, yet they practiced and still practicing poorly, mainly due to negligence.

The study cites that the lack of tailor-made good agricultural practices guidelines (i.e. Tanzania GAP) and tailor-made motivational programs on proper and safe use of pesticides could have a significant contribution. Good agricultural practices guidelines tailored for Tanzanian conditions could be helpful because users of pesticides will find tailored guidelines on how to use pesticides on a specific crop. For tailor-made motivational programs, Institutions responsible for pesticide registration, control, and management could arrange in field training on proper and safe use of pesticides on regular basis. Institution could identify farmers and the types of pesticides they use, and then arrange in field training in their farms (trainers go to the farmer's farms). Although pesticides have labels and direction of usage, still, pesticide sellers could give more information regarding proper and safe use of pesticides to the persons who go to buy from them. This survey was necessary because pesticide practices influence the amount of pesticides that end up in the environment as well as the exposure pathways.

Weather data used in the simulation were for both year 2012 and 2013. When considering the entire duration of the simulation (250 days); there were no statistical significant difference on variability of temperatures ($p > 0.05$) and rainfalls ($p > 0.05$) between year 2012 and 2013. When considering the first 50 days of the simulation;

there was no statistical significant difference on variability of temperatures ($p > 0.05$) and rainfalls ($p > 0.05$) between year 2012 and 2013. However, distribution of temperatures and rainfalls had profound impact on fate and transport of glyphosate and its effects on population dynamics of frogs.

Open source object-oriented algorithm for modeling fate and transport of glyphosate and its spray drifts and their effects on population dynamics of frogs are presented. Simulation was performed for 250 days. Results of the fate and transport show that in 250 days, all glyphosate which was deposited on the crop/leaf surfaces was removed completely by competing processes from the crop/leaf surfaces. This suggests that glyphosate does not stay longer on the crop/leaf surfaces.

The study found that washoff due to the effect of rainfall was largely responsible for removing glyphosate from the crop/leaf surfaces. Results suggest that if there were no washoff events that transferred glyphosate from the crop/leaf surfaces to the topsoil, all glyphosate in the topsoil could have been removed completely by the competing processes by day 40 of the simulation, meaning that glyphosate does not stay longer in the topsoil. Results show that volatilization from the topsoil were very little. Also, results show that total macropore flow were 1.33% and 0.88% of the glyphosate applied for year 2012 and 2013 respectively, suggesting that the transfer of glyphosate to the subsoil could be low.

Results show that runoff highly depend on the amount of rainfall, amount of glyphosate present before the runoff event occurred, and the width of buffer zone around the agricultural field. When using width of buffer zone of zero meters, glyphosate runoffs were higher compared to when using width of buffer zone of 30 meters. These runoffs were responsible for the contamination of glyphosate in the water body located adjacent to the agricultural field.

When using buffer zone width of thirty meters, little runoffs occurred but did not cause high impact on the population dynamics of frogs for both year 2012 and 2013. When using buffer zone width of zero meters, more runoffs crossed the agricultural field to the water body. Results show that glyphosate runoff events in year 2013 occurred earlier than when it occurred in year 2012. These glyphosate runoff events for year 2013 (when using buffer zone width of zero meters) caused high mortality rates (low survival rates), which killed all tadpoles within 86 days of the

simulation. Results show that for year 2012, all tadpoles died within 105 days after the first application.

Results suggest that high glyphosate concentration and high toxicity of glyphosate in the environment could be responsible for the declining population of frogs. Therefore, the use of glyphosate may be restricted especially in areas close to the water body, as well as the use of low toxic formulations of glyphosate is highly recommended. Also, integrated pest management tactics could be used to reduce the use of herbicides for weed control.

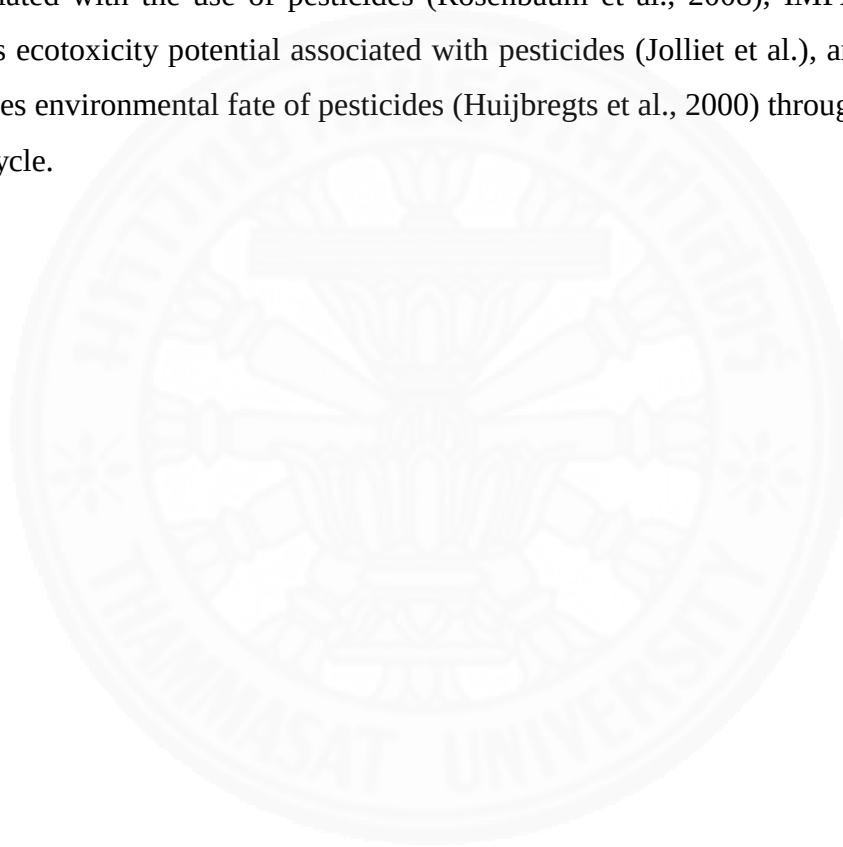
Effect of spray drifts (of glyphosate) on population dynamics of frogs was studied using object-oriented programming. The study found that width of no spray zone around the agricultural field (buffer zone) highly reduces spray drifts of glyphosate from crossing the agricultural field to non-target sites, thus, non-target species around the agricultural field are likely to stay free from exposure of glyphosate. Results show that very small amount of glyphosate contamination in the water body had no impact on population dynamics of frogs, but high glyphosate contamination killed majority of tadpoles, froglets, and adult frogs and there were no eggs laid.

Sensitivity analysis show that (i) there is a correlation between glyphosate concentrations in the water body and the population dynamics of eggs, tadpoles, froglets, and adult frogs, (ii) there is no correlation between glyphosate concentrations in the topsoil and the population dynamics of eggs, tadpoles, froglets, and adult frogs, (iii) there is a correlation between the population of frogs and the number of eggs laid per female frog, (iv) there is a correlation between the amount of glyphosate removed from the crop/leaf surfaces (by washoff) and the amount of rainfall, (v) there is a correlation between the amount of glyphosate removed from the topsoil (by runoff) and the amount of rainfall, (vi) there is a correlation between the amount of glyphosate removed from the topsoil (by macropore flow) and the amount of rainfall, (vii) there is a correlation between the amount of glyphosate removed from the topsoil (by runoff) and the buffer zone width, and (viii) there is a correlation between the population of frogs and the buffer zone width.

The study could be reproduced and the model could be used elsewhere to simulate similar objectives. It is with hope that results of this study could be of

practical significance to pesticide regulators and those involved in environmental modeling and assessments. In addition, the model (algorithm) presented in this study will provide modelers a simulation tool which could be used to conduct research studies and could be of practical significance to those involved in ecological modeling.

Furthermore, results of the model could also be combined with the life cycle assessment models such as USEtox to assess the freshwater ecotoxicity potential associated with the use of pesticides (Rosenbaum et al., 2008), IMPACT 2002+ to assess ecotoxicity potential associated with pesticides (Jolliet et al.), and USES-LCA to assess environmental fate of pesticides (Huijbregts et al., 2000) throughout the entire life cycle.



5.2 Recommendations

The study recommends the following:

5.2.1 Pesticide users (farmers)

- i) To use herbicides or pesticides according to the instructions provided on the labels,
- ii) To use protective gears when working with herbicides or pesticides,
- iii) To seek medical help in case of herbicides or pesticide accidents,
- iv) To use buffer zones of at least thirty meters around their fields (more precisely, according to the recommendations based on crop types and application method),
- v) To not spray during rainy days,
- vi) To use less toxic formulations of herbicides or pesticides,
- vii) To use IPM to reduce the frequency of using herbicides or pesticides,
- viii) To refrain from eating frogs captured around their farms due to the reasons that they could be contaminated with herbicides or pesticides, and
- ix) To regularly test the water around their fields for potential herbicides or pesticide contamination if that water is used for human and animal consumptions.

5.2.2 Policy makers

- i) To enforce regulations on regular training to farmers on good agricultural practices,
- ii) To offer incentives to those who use less toxic formulations of herbicides or pesticides e.g. lowering of costs or giving discounts,
- iii) To enforce regular environmental monitoring programs especially in agricultural regions, and
- iv) To offer incentives to those who use buffer zones around their fields e.g. giving discounts when paying for checking their health (medical checkup).

5.3 Limitations of the Study

Limitations of this study include:

- i) Survey on pesticide awareness was conducted online,
- ii) Number of samples (respondents) from the survey was one hundred fifty-seven which was below three hundred eighty, that led to the results not being generalized to the true population of Tanzania,
- iii) Questionnaire section and simulation modeling section can only be partially linked because data used were not from the same country i.e. data from the same country will obtain better results,
- iv) Lack of climate data from Tanzania, which resulted to the use of climate data from Thailand instead,
- v) The study used secondary data from various sources,
- vi) Lack of toxicity data for the species of frog used for the simulation of population dynamics i.e. species of frog used is not native to both Tanzania and Thailand but from Australia,
- vii) Numerous processes are based on analytical expressions/regressions from various pedoclimatical contexts,
- viii) There could be better modeling regressions and equations other than the ones used for the simulation which could make a difference,
- ix) The study (model) cannot be used to assess herbicide or pesticide leaching in the soil, and
- x) The study (model) cannot be used to assess the effect of herbicide or pesticide on malformation on tadpoles, froglets, and adult frogs.

5.4 Future work

Future work will involve the following:

- i) Addition of leaching processes to the model (from the topsoil to subsoil and groundwater),
- ii) Modeling the effect of glyphosate on malformation of tadpoles, froglets, and adult frogs,

- iii) Migration of the model from UniSim-1.52 (current version) to UniSim2 (under development),
- iv) Addition of more scenarios (geographical locations) e.g. Kamphaeng Phet, Loei, Phichit, etc.



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The seal of Thammasat University is a circular emblem. It features a central five-tiered umbrella (parasol) with a flame-like finial. The umbrella is surrounded by a wreath of rice and cotton stalks. The entire emblem is enclosed within a circular border containing the university's name in Thai script at the top and "THAMMASAT UNIVERSITY" in English at the bottom, separated by small floral motifs.

APPENDICES

APPENDIX A

TABLES

Table A.1 Classification of pesticides according to the WHO (Stenersen, 2004).

Class	Details	LD ₅₀ for the rat (mg/kg of body weight)			
		Oral toxicity		Dermal toxicity	
		Solids	Liquids	Solids	Liquids
Ia	Extremely hazardous	≤5	≤20	≤10	≤40
Ib	Highly hazardous	5-50	20-200	10-100	40-400
II	Moderately hazardous	50-500	200-2,000	100-1,000	400-4,000
III	Slightly hazardous	≥501	≥2,001	≥1,001	≥4,001
Table 5	Unlikely to present acute hazard in normal use	≥2,000	≥3,000		
Table 6	Not classified, believed obsolete				
Table 7	Fumigants not classified under WHO				

Table A.2 Classification of carcinogenicity of pesticides according to the International Agency for Research on Cancer (Stenersen, 2004).

Class	Details
Group 1	Carcinogenic to humans
Group 2A	Probably carcinogenic to humans
Group 2B	Possibly carcinogenic to humans
Group 3	Not classifiable as to carcinogenicity to humans
Group 4	Probably not carcinogenic to humans

Table A.3 Classification of dose intake (Stenersen, 2004).

Dose	Time scale under exposure
Acute	The dose is given during a period shorter than 24 hours
Sub-acute	The dose is given between 24 hours and 1 month
Sub-chronical	The dose is given between 1 month and 3 months
Chronical	The dose is given for more than 3 months

Table A.4 Toxicity class and their average daily intake (Stenersen, 2004).

Group	Pesticides	Rat oral LD ₅₀ (mg/kg)	Toxicity class (WHO)	ADI (mg/kg of body weight)
Organophosphorus insecticides	Parathion	2	Ia	0.004
	Azinphos-methyl	9	Ib	0.005
	Dichlorvos	50	Ib	50
Insecticides acting on Na channels	DDT	115	II	0.02
	Deltamethrin	5,000	II	0.01
Fungicide	Captan	9,000	III	0.1
Herbicide	Glyphosate	5,600	III	0.3

Table A.5 Global and USA pesticide use (Kearney & Roberts, 1998).

Type of pesticides	Global pesticide use (million kg)	Pesticide use in the USA (million kg)
Herbicide	1,002	301
Insecticide	767	153
Fungicide	256	74

Table A.6 Influence of glyphosate (Roundup®) on the 96 h growth of *Xenopus laevis* embryos (Perkins et al., 2000).

Concentration		Mean length (mm) ± SD
mg a.e/L	mg a.i/L	
5	6.67	7.12 ± 0.26
6	8	7.44 ± 0.27
8	10.67	7.0 ± 0.24
10	13.33	6.72 ± 0.24

Table A.7 Effects of glyphosate and its formulation on frogs and toads, adopted from Pérez et al. (2011) and Mann et al. (2003).

Species (frog/toad)/stage	Chemical assessed	Parameter assessed	Type of study	Effects concentrations/LC ₅₀ (mg a.i./L)	Species found in	Reference
Bullfrog (tadpole)	Roundup®	Mortality	Single species laboratory test (SSLT) (48 h)	48 h LC ₅₀ = 1.33 (1 mg/L)	Brazil	(Costa et al., 2008)
Bullfrog (tadpole)	Glyphosate isopropylamine salt	Mortality		48 h LC ₅₀ = 108 mg/L	Southern Ontario, Canada	(Clements et al., 1997)
<i>Lymnodynastes dorsalis</i> (tadpole)	Glyphosate Isopropylamine	Morality	SSLT (48 h)	48 h LC ₅₀ = >587	Southwestern Australia	(Mann & Bidwell, 1999)
	Roundup®			48 h LC ₅₀ = 8.3		
	Touchdown®			48 h LC ₅₀ = 36.2		
	Roundup®Biactive			48 h LC ₅₀ = >1,111		
<i>Heleioporus eyrei</i> (tadpole)	Glyphosate Isopropylamine	Morality		48 h LC ₅₀ = >548		
	Roundup®			48 h LC ₅₀ = 23.9		
	Touchdown®			48 h LC ₅₀ = 50.2		
	Roundup®Biactive			48 h LC ₅₀ = >1,186		
<i>Crinia insignifera</i> (tadpole)	Glyphosate Isopropylamine	Morality		48 h LC ₅₀ = >684		
	Roundup®			48 h LC ₅₀ = 10		
	Touchdown®			48 h LC ₅₀ = 27.3		
	Roundup®Biactive			48 h LC ₅₀ = 1,372		
<i>Litoria moorei</i> (tadpole)	Technical-Grade Glyphosate Acid	Morality		48 h LC ₅₀ = 108.3		
	Glyphosate Isopropylamine			48 h LC ₅₀ = >503		
	Roundup®			48 h LC ₅₀ = 8.1		
	Roundup®Biactive			48 h LC ₅₀ = 911		
<i>Litoria moorei</i> (tadpole)	Technical-Grade Glyphosate Acid	Morality, (lake water was the diluents)	48 h LC ₅₀ = 161.3		(Bidwell & Gorrie, 1995)	
	Roundup®		48 h LC ₅₀ = 32.2			
<i>Crinia insignifera</i> (metamorph)	Roundup®	Morality (aged tapwater was the diluents)	48 h LC ₅₀ = 144			
<i>Crinia insignifera</i> (adult)	Roundup®		48 h LC ₅₀ = 137			
	Technical-Grade Glyphosate Acid		48 h LC ₅₀ = 111.5			
<i>Xenopus laevis</i> (embryonic development)	Rodeo®	Morality	SSLT (96 h)			96 h LC ₅₀ = 7,209
	Roundup®			96 h LC ₅₀ = 12.5		
	POEA			96 h LC ₅₀ = 2.7		
<i>Scinax nasicus</i> (tadpole)	Glyfos®	Morality/Mortality	SSLT (48 h)	48 h LC ₅₀ = 3.62	Paraná, Argentina	(Lajmanovich et al., 2003)
			SSLT (96 h)	96 h LC ₅₀ = 2.64		
<i>Rana pipiens</i> (tadpole)	Vision®	Morality	SSLT (8 days)	1 mg a.i./L evoked 100% mortality	Western Vermont, USA	(Chen et al., 2004)
<i>Rana pipiens</i> (tadpole)	Roundup®	Mortality and growth	SSLT (16 days)	No observed significant effects	USA	(Relyea, 2004)
<i>Rana clamitans</i> (tadpole)				2 mg a.i./L reduced survival and growth		
<i>Rana catesbeiana</i> (tadpole)				2 mg a.i./L reduced survival and growth		
<i>Bufo americanus</i> (tadpole)				2 mg a.i./L reduced growth		

Species (frog/toad)/stage	Chemical assessed	Parameter assessed	Type of study	Effects concentrations/LC ₅₀ (mg a.i./L)	Species found in	Reference
<i>Hyla versicolor</i> (tadpole)				No observed significant effects		
<i>Rana sylvatica</i> (tadpole)	Roundup®	Mortality and biomass	Mesocosm studies (15 days)	3.8 mg a.i./L evoked 100% mortality		(Relyea, 2005a)
<i>Rana pipiens</i> (tadpole)				3.8 mg a.i./L evoked 98% mortality		
<i>Bufo americanus</i> (tadpole)				3.8 mg a.i./L evoked 98% mortality		
<i>Hyla versicolor</i> (tadpole)				3.8 mg a.i./L evoked 100% mortality		
<i>Pseudacris crucifer</i> (tadpole)				No observed significant effects		
<i>Rana cascadae</i> (tadpole)	Roundup®	Mortality and metamorphosis	SSLT (43 days)	1.94 mg a.i./L evoked 51.4% mortality, and earlier metamorphosis times	Washington, USA	(Caulble & Wagner, 2005)
				0.95 mg a.i./L evoked 8.6% mortality		
<i>Rinella arenarum</i> (tadpole)	Glyphosate	Mortality	SSLT (48 h)	48 h LC ₅₀ = 13.20	Argentina	(Lajmanovich et al., 2013)
				NOEC at 10 mg a.i./L		
				LOEC at 20 mg a.i./L		
<i>Rinella arenarum</i> (tadpole)	Glyphosate	Mortality	SSLT (48 h)	48 h LC ₅₀ = 3.23	Argentina	(Lajmanovich et al., 2011)
<i>Rana clamitans</i> and <i>Rana pipiens</i> (tadpole)	Vision®	Mortality, avoidance response, and growth	SSLT (96 h)	96 h LC ₅₀ = 3.6-15.3	Northern Ontario, Canada	(Wojtaszek et al., 2004)
				Substantial mortality and incidences of abnormal avoidance response occurred at > 1.9 mg a.i./L		
<i>Xenopus laevis</i> (embryo)	Vision®	Mortality and the prevalence of malformations	SSLT (96 h)	96 h LC ₅₀ = 20.8, at pH 6	Guelph, Canada	(Edgington et al., 2004)
				96 h LC ₅₀ = 10.53, at pH 7.5		
<i>Xenopus laevis</i> (larvae)				96 h LC ₅₀ = 2.8, at pH 6		
				96 h LC ₅₀ = 1.17, at pH 7.5		
<i>Bufo americanus</i> (embryo)				96 h LC ₅₀ = 6.4, at pH 6		
				96 h LC ₅₀ = 8.53, at pH 7.5		
<i>Bufo americanus</i> (larvae)				96 h LC ₅₀ = 3.86, at pH 6		
				96 h LC ₅₀ = 2.27, at pH 7.5		
<i>Rana clamitans</i> (embryo)				96 h LC ₅₀ = 7.06, at pH 6		
				96 h LC ₅₀ = 5.46, at pH 7.5		
<i>Rana clamitans</i> (larvae)				96 h LC ₅₀ = 4.66, at pH 6		
				96 h LC ₅₀ = 1.86, at pH 7.5		
<i>Rana pipiens</i>				96 h LC ₅₀ =		

Species (frog/toad)/stage	Chemical assessed	Parameter assessed	Type of study	Effects concentrations/LC ₅₀ (mg a.i./L)	Species found in	Reference
(embryo)				20.13, at pH 6		
				96 h LC ₅₀ = 10, at pH 7.5		
<i>Rana pipiens</i> (larvae)				96 h LC ₅₀ = 2.4, at pH 6		
				96 h LC ₅₀ = 1.46, at pH 7.5		
<i>Rana pipiens</i> (tadpole, Gosner stage 25)	Roundup Original®	Mortality and growth	SSLT (96 h)	96 h LC ₅₀ = 3.87	Peterborough, Canada	(Howe et al., 2004)
<i>Rana pipiens</i> (tadpole, Gosner stage 20)				96 h LC ₅₀ = 8.67		
<i>Rana sylvatica</i> (tadpole, Gosner stage 25)				96 h LC ₅₀ = 6.8		
<i>Rana sylvatica</i> (tadpole, Gosner stage 20)				96 h LC ₅₀ = >10.67		
<i>Bufo americanus</i> (tadpole, Gosner stage 25)				96 h LC ₅₀ = <5.33		
<i>Bufo americanus</i> (tadpole, Gosner stage 20)				96 h LC ₅₀ = 10.67		
<i>Rana clamitans</i> (tadpole, Gosner stage 25)				96 h LC ₅₀ = 2.67		
<i>Rana clamitans</i> (tadpole, Gosner stage 20)				96 h LC ₅₀ = 9.47		
<i>Rana clamitans</i> (tadpole, Gosner stage 20)	Glyphosate technical			96 h LC ₅₀ = >23.87		
<i>Rana clamitans</i> (tadpole, Gosner stage 20)	POEA			96 h LC ₅₀ = 2.93		
<i>Rana clamitans</i> (tadpole, Gosner stage 20)	Roundup Biactive®			96 h LC ₅₀ = >23.87		
<i>Rana clamitans</i> (tadpole, Gosner stage 20)	Touchdown®			96 h LC ₅₀ = >23.87		
<i>Rana clamitans</i> (tadpole, Gosner stage 20)	Glyfos BIO®			96 h LC ₅₀ = >23.87		
<i>Rana clamitans</i> (tadpole, Gosner stage 20)	Glyfos AU®			96 h LC ₅₀ = 11.87		
<i>Rana clamitans</i> (tadpole, Gosner stage 20)	Roundup Transorb®			96 h LC ₅₀ = 2.93		
<i>Hyla versicolor</i> (tree frog tadpole), <i>Bufo americanus</i> (toad tadpole), <i>Rana pipiens</i> (leopard frog tadpole)	Roundup®	Mortality, survival	SSLT (20 days)	3.8 mg a.i./L evoked 96-100% of larval amphibians	Pennsylvania, USA	(Relyea, 2005b)
			SSLT (24 h)	3.8 mg a.i./L evoked 68-86% of juvenile		

Species (frog/toad)/stage	Chemical assessed	Parameter assessed	Type of study	Effects concentrations/LC ₅₀ (mg a.i./L)	Species found in	Reference
				amphibians		
<i>Dendropophus microcephalus</i> (tadpole)	Formulated Glyphosate and Cosmo-Flux	Mortality	SSLT (96 h)	96 h LC ₅₀ = 1.6	Colombia	(Bernal et al., 2009)
<i>Rhinella typhonius</i> (tadpole)				96 h LC ₅₀ = 2		
<i>Scinax ruber</i> (tadpole)				96 h LC ₅₀ = 2.18		
<i>Hypsiboas crepitans</i> (tadpole)				96 h LC ₅₀ = 2.75		
<i>Rhinella granulosa</i> (tadpole)				96 h LC ₅₀ = 3.13		
<i>Centrolene prosoblepon</i> (tadpole)				96 h LC ₅₀ = 3.22		
<i>Rhinella marina</i> (tadpole)				96 h LC ₅₀ = 3.64		
<i>Engystomops pustulosus</i> (tadpole)				96 h LC ₅₀ = 3.72		
<i>Bufo melanostictus</i> Asian common toad (tadpole)	Glyphosate	Mortality	SSLT (48 h)	48 h LC ₅₀ = 49.44	Asia	(Jayawardena et al., 2011)

Table A.8 The survival rates of juvenile *Spea multiplicata* and *Bufo cognatus* 48 h after exposure to herbicides (Dinehart et al., 2009).

Plastic tubs lined with	48 h direct exposure survival (%)				
	Control (aged well water)	IG	WM	WGKC	WGKP
<i>Spea multiplicata</i> (New Mexico spadefoot)					
Soil	100	100	100	100	0
Paper towels	71.1	74.1	59.6	97.2	0
<i>Bufo cognatus</i> (Great Plains toad)					
Soil	97.5	100	100	100	22.5
Paper towels	97.5	75	87.5	47.5	0

Ignite® 280 SL (IG); 0.21 mL glufosinate/m²,

RoundupWeatherMAX® (WM); 0.16 mL glyphosate/m²,

Roundup Weed and Grass Killer Super Concentrate® (WGKC); 1.33 mL glyphosate/m²,

Roundup Weed and Grass Killer Ready-To-Use Plus® (WGKP); 1.33 mL glyphosate/m²,

“Animals were exposed in plastic tubs lined with soil (shaded bars) and paper towels (open bars)” (Dinehart et al., 2009).

Table A.9 Categories of toxicity for aquatic organisms (EPA, 2013; Pérez et al., 2011).

LC ₅₀ (mg a.i./L)	Toxicity category
<0.1	Very highly toxic
>0.1 - 1	Highly toxic
>1 - 10	Moderately toxic
>10 - 100	Slightly toxic
>100	Practically nontoxic

Table A.10 Data used to simulate results.

Parameter	Values	Description	Reference
General data			
steps	250 days	Maximum simulation steps	User defined
initialDateTime	13/03/2012, 13/03/2013	First day of the simulation	User defined
timestep	1 day	Time step during simulation	User defined
timeunit	day	Time unit	User defined
iterations	30 iterations	Number of iterations for sensitivity analysis	User defined
Field data			
area	10,000 m ²	Area of the whole field	User defined
wbz	0 m, 30 m	Width of the buffer zone	User defined
x	0 m, 30 m	Distance off field edge	User defined
latitude	14.02° N	Latitude of the study area (Pathumthani Province, Thailand)	(Hiranvarodom, 2003; Pongswat et al., 2004)
S	0.18	Slope fraction of the field	(Phewnil et al., 2010)
T _f	1	Tillage factor	(Dijkman et al., 2012)
Crop data			
sowingDate	04/04	Day of sowing (04/04/2012 and 04/04/2013)	User defined
seeds	1	Fraction of seeds sown	User defined
T ₀	10 °C	Threshold temperature for maize development. Varies from 5°C (Dijkhuis, 1956) to 10 °C (Brewbaker, 2003; Edwards, 2009)	(Brewbaker, 2003; Edwards, 2009)
f _i	leafDevelopment 0.25, tillering 0.50, stemElongation 0.70,	Crop deposition fractions for cereals	(Linders et al., 2000)

Parameter	Values	Description	Reference
	and senescence 0.90		
duration	leafDevelopment 87, tillering 282, stemElongation 657, and senescence 1,518	Degree day values for maize development (leaf development, tillering, stem elongation, and senescence)	(Edwards, 2009)
leafType	Waxy	Cuticle characteristics similar to that of citrus leaves (Table A.13)	(Dijkman et al., 2012)
a ₁	18.1851%	Regression constants	(van de Zande et al., 2015)
a ₂	0.5553%		
b ₁	1.0701 m ⁻¹		
b ₂	0.0871 m ⁻¹		
Pesticide data			
applicationDate	14/03, 28/03	Day of application; 14/03 (app1), 28/03 (app2)	User defined
concentration	480 g a.e/L	Concentration of the active ingredient, based on application dose of 1,500 g a.i/ha (Waramit & Suwanketnikom, 1999)	(Waramit & Suwanketnikom, 1999)
rate	3.125 L/ha	Application rate, based on application dose of 1,500 g a.i/ha (Waramit & Suwanketnikom, 1999)	(Waramit & Suwanketnikom, 1999)
VP	2.45x10 ⁻⁵ Pa	Vapour pressure	(Stenersen, 2004)
T _{refVP}	25°C	Reference temperature at which vapour pressure was measured	(Stenersen, 2004)
MW	169.07 g/mol	Molar mass of the pesticide	(Henderson et al., 2010; Laitinen et al., 2007)
MV	96.61 cm ³ /mol	Molecular volume of the	User defined

Parameter	Values	Description	Reference
		pesticide (density 1.75 g/cm ³ (Nandula, 2010).	
T _{ref}	25°C	Reference temperature at which the half-life time of pesticide was determined in the soil	User defined
Q ₁₀	2.58	Increase in biodegradation rate per 10 °C	(EFSA, 2007)
k _{OH}	79x10 ⁻¹² cm ³ /molecules/h	Overall OH• oxidation rate constant	(Dijkman et al., 2012)
pK _a	0.8	First acid dissociation constant of glyphosate	(de Ridder et al., 2010; Goodwin et al., 2003)
K _{oc}	20,100 L/kg	Organic carbon-water partitioning coefficient	(Henderson et al., 2010)
P _{sol}	11.5 g/L	Solubility of the pesticide at 25°C	(Stenersen, 2004)
DT ₅₀	3 days	Soil half-life (parent material); varies from 3-174 days (Stenersen, 2004), 2-197 days (Giesy et al., 2000; Henderson et al., 2010), 3-130 days (Schuette, 1998) and 47 days (Kearney & Roberts, 1998)	(Giesy et al., 2000; Henderson et al., 2010; Schuette, 1998; Stenersen, 2004)
DT ₅₀	4.5 days	Half-life in water (parent material); varies from few days to 91 days (Henderson et al., 2010), 4.5 days (Focks et al., 2014a), 7-10 weeks in natural water (Struger et al., 2008).	(Focks et al., 2014a)

Parameter	Values	Description	Reference
Weather data			
T_{air}	Read from the weather file	Average air temperature (°C)	(TMD, 2014)
P	Read from the weather file	Daily rainfall (mm)	(TMD, 2014)
$t_{p,\text{event}}$	3 h	Number of hours precipitation occurs on a rainy day	(Hung et al., 2009)
Soil data			
f_{oc}	0.2	Fraction of organic carbon in the topsoil	(Wu et al., 1998)
ρ_b	1.365 kg/L	Soil bulk density (average, Thailand)	(Simmons et al., 2005)
f_{sand}	0.1043	Fraction of sand in the soil	(Waramit & Suwanketnikom, 1999)
f_{silt}	0.3114	Fraction of silt in the soil	(Waramit & Suwanketnikom, 1999)
f_{clay}	0.5849	Fraction of clay in the soil	(Waramit & Suwanketnikom, 1999)
f_{om}	0.1	Fraction of organic matter in the topsoil	(Bot & Benites, 2005)
pH	5	pH of the soil; can range between 4-7 (Lesturgez et al., 2006; Phewnil et al., 2010; Waramit & Suwanketnikom, 1999)	(Waramit & Suwanketnikom, 1999)
f_a	0.25	Fraction of air in the soil	User defined
f_w	0.25	Fraction of water in the soil	User defined
f_s	0.5	Fraction of soil solids in the soil	User defined
Frog data			

Parameter	Values	Description	Reference
initial	1,000 tadpoles	Initial number of tadpoles	User defined
duration	14 days	Longevity of eggs	User defined
duration	90 days	Longevity of tadpoles. It takes about three to five months (Hero & Roberts, 2004; Tyler & Knight, 2011)	(Hero & Roberts, 2004; Tyler & Knight, 2011)
duration	21 days	Longevity of froglets	User defined
duration	365 days	Longevity of adults	User defined
duration	21 days	Days after entering adult stage that eggs will be laid	User defined
eggsPerFemale	70 eggs	Number of eggs laid per female, varies between 66-268 eggs	(Hero & Roberts, 2004; Tyler & Knight, 2011)
LC ₅₀	4.8 mg a.i/L	Egg stage (48 h), assumed based on LC ₅₀ for tadpole stage	User defined
LC ₅₀	4.8 mg a.i/L	Tadpole stage (48 h), converted from 3.6 mg a.e/L	(Mann & Bidwell, 1999)
LC ₅₀	69 mg a.i/L	Froglet stage (48 h), converted from 51.8 mg a.e/L	(Mann & Bidwell, 1999)
LC ₅₀	65.9 mg a.i/L	Adult stage (48 h), converted from 49.4 mg a.e/L	(Mann & Bidwell, 1999)
sexRatio	0.5	Proportion of female frogs in the population	(Navarro-Martín et al., 2014)
slope	2.341 volume/amount	Sigmoidal function	(Focks et al., 2014b)

Table A.11 Crop deposition fractions (Linders et al., 2000).

Crop	Development stage	Deposition fractions (<i>f</i>)
Cereals I	Leaf development	0.25
Cereals II	Tillering	0.50
Cereals III	Stem elongation	0.70
Cereals IV	Senescence	0.90

Table A.12 Recommended width of no spray zones from the edge of the agricultural field to non-target sites, partly adopted from Ucar & Hall (2001).

Pesticide	Application type	Application rate	Recommended width of no spray zone	Reference
Insecticide (Triazophos)	Conventional hydraulic sprayers	25 g a.i/ha	12 m	(Davis et al., 1993)
Insecticide (Permethrin)	Ground application	<35 g/ha	20 m	(Payne et al., 1988)
Insecticide (Cypermethrin)	Conventional hydraulic sprayers	25 g a.i/ha	16-24 m	(Davis et al., 1993)
Herbicide (Glyphosate)	Aerial	3.4 kg a.i/ha	25-30 m	(Payne et al., 1990)
Herbicide (Glyphosate)	Sprayer with 6 m boom (Tractor mounted)	2.2 kg a.i/ha	15-20 m	(Marrs et al., 1993)
Herbicides	Tractor mounted sprayers		6-10 m (perennials), 20 m (seedling)	(Marrs et al., 1992)
Pesticides	Spraying in Orchard		45.8 m	(Felsot, 1997)
Pesticides			10-30 m (water habitat creation)	(Burn, 2003)
Pesticides			3 - 30 m (riparian buffer strip)	(Sieber et al., 2010)
Formulated pesticides and fluorescent tracer	24 m boom sprayer	200 L/ha	6 m	(Cilgi, 1993)
Formulated pesticides and fluorescent tracer	Ground	200 L/ha	6 m	(Longley et al., 1997)
Herbicides	Simulated wetland		10 m	(Brown et al., 2004)

Table A.13 Species-dependent constant (Dijkman et al., 2012).

Species (leaf type)	a	b
Pear (NonWaxy)	-4.93	5.30×10^{-3}
Citrus (Waxy)	-5.31	5.84×10^{-3}

Table A.14 Concentration of OH radicals at 0.5 km height ($\times 10^5$ molecules/cm³) (Bahm & Khalil, 2004).

Latitude (° S or N)/Month (day of the year)	Dec (-16)	Mar (74)	Jun (166)	Sep (258)	Dec (349)	Mar (439)
-85	5.1	0	0	0	5.1	0
-75	5.1	0.3	0	0.2	5.1	0.3
-65	3.8	1	0	0.9	3.8	1
-55	5.8	2.5	0.3	2.8	5.8	2.5
-45	9.6	5.6	1.4	6.2	9.6	5.6
-35	16.6	10.9	4.4	11.5	16.6	10.9
-25	23.6	15.7	9.1	19.6	23.6	15.7
-15	22.7	15.1	12.2	25.8	22.7	15.1
-5	21.4	18.1	17.4	28.8	21.4	18.1
5	17.2	21.6	20.9	25.5	17.2	21.6
15	15.1	22.5	25.6	23.8	15.1	22.5
25	12.6	24.2	33.6	26.1	12.6	24.2
35	4.7	15.9	38.2	22.1	4.7	15.9
45	0.7	6.7	30	11.7	0.7	6.7
55	0.1	2.1	20.3	4.8	0.1	2.1
65	0	0.6	15.5	1.6	0	0.6
75	0	0.1	8.3	0.6	0	0.1
85	0	0	5.9	0.1	0	0

Note that, on latitude column, - means S (south).

Table A.15 Tillage factor (T_f) (Dijkman et al., 2012).

Tillage type	Tillage factor
Conventional tillage	1
Reduced tillage	3.5
No tillage	7.5

APPENDIX B

QUESTIONNAIRE ON AWARENESS OF PESTICIDES AND THEIR ASSOCIATED HEALTH EFFECTS IN TANZANIA

Pesticides are chemicals that are applied on crops and plants to control pests. Pests are insects, animals, and weeds that destroy plants, food etc. This survey seeks to understand how aware people in Tanzania, are of pesticides and their associated health effects. It's a comprehensive survey that includes everyone (i.e. farmers and non-farmers). This survey will take about 15-20 minutes of your precious time to complete. We greatly value and appreciate your time. All data is collected on the basis of anonymity i.e. no personal data such as name, and date of birth shall be collected. This survey can be filled in by anyone living in Tanzania (resident, working, studying, etc.) or Tanzanians who may be currently (temporarily) living abroad but their country of residence is Tanzania. This survey is accepting responses until 31st December 2015. Please note that * means required question that cannot be left unanswered.

1. When was the first time you heard about pesticides?*

(Choose only one that best describes you)

- ☐ More than 10 years ago
- ☐ 6-10 years ago
- ☐ 2-5 years ago
- ☐ 1-2 years ago
- ☐ Recently, within a year
- ☐ Never heard
- ☐ I don't remember

2. When was the first time you heard about health effects associated with pesticides?*

(Choose only one that best describes you)

- ☐ More than 10 years ago

- ☐ 6-10 years ago
- ☐ 2-5 years ago
- ☐ 1-2 years ago
- ☐ Recently, within a year
- ☐ Never heard
- ☐ I don't remember

3. When was the last time you heard about health effects associated with the use of pesticides?*

(Choose only one that best describes you)

- ☐ More than 10 years ago
- ☐ 6-10 years ago
- ☐ 2-5 years ago
- ☐ 1-2 years ago
- ☐ Recently, within a year
- ☐ Never heard
- ☐ I don't remember

4. When was the last time you heard anything in general about pesticides?*

(Choose only one that best describes you)

- ☐ More than 10 years ago
- ☐ 6-10 years ago
- ☐ 2-5 years ago
- ☐ 1-2 years ago
- ☐ Recently, within a year
- ☐ Never heard
- ☐ I don't remember

5. Is agriculture one of your activities?*

(Choose only one that best describes you)

- ☐ Yes
- ☐ No

6. If agriculture is one of your activities, have you ever used pesticides?
(If agriculture is not one of your activities, skip this question. Choose only one that best describes you)
- ☐ Yes, often
 - ☐ Yes, few times
 - ☐ No
7. If agriculture is not one of your activities, have you ever used pesticides?
(If agriculture is one of your activities, skip this question. Choose only one that best describes you)
- ☐ Yes, often
 - ☐ Yes, few times
 - ☐ No
8. If you ever used pesticides, did you use protective gears?
(Mark all that apply. If other, please specify)
- ☐ Yes, all required protective gears
 - ☐ Yes, few times without masks and overalls
 - ☐ No, I didn't use any protection
 - ☐ Never used goggles
 - ☐ Never used gloves
 - ☐ Other
9. When working with pesticides, do you use protective gears?*
- (Mark all that apply. If other, please specify)
- ☐ Yes, all required protective gears
 - ☐ Yes, few times without masks and overalls
 - ☐ No, I don't use any protection
 - ☐ I don't use goggles
 - ☐ I don't use gloves
 - ☐ My work doesn't involve the use of pesticides
 - ☐ Other

10. What is your opinion about pesticides?*

(Mark all that apply. If other, please specify)

- ☐ They are toxic
- ☐ Can kill human beings
- ☐ Can kill living organisms
- ☐ They are non-toxic
- ☐ Can cause no harm to living organisms
- ☐ Can be kept anywhere in the house
- ☐ Must be handled with care
- ☐ Anyone can have access to pesticides including children
- ☐ Other

11. In the last 1-5 years, has any pesticide come in contact with your skin?*

(Choose only one that best describes you)

- ☐ Yes, often
- ☐ Yes, few times
- ☐ No
- ☐ Possible, I don't remember

12. In the last 6-10 years, has any pesticide come in contact with your skin?*

(Choose only one that best describes you)

- ☐ Yes, often
- ☐ Yes, few times
- ☐ No
- ☐ Possible, I don't remember

13. In the last 1-5 years, has any pesticide come in contact with your eyes?*

(Choose only one that best describes you)

- ☐ Yes, often
- ☐ Yes, few times
- ☐ No
- ☐ Possible, I don't remember

14. In the last 6-10 years, has any pesticide come in contact with your eyes?*

(Choose only one that best describes you)

- ☐ Yes, often
- ☐ Yes, few times
- ☐ No
- ☐ Possible, I don't remember

15. If a pesticide comes in contact with your skin (body), what would you do?*

(Mark all that apply. If other, please specify)

- ☐ Rinse with clean water
- ☐ Take antidote; a chemical substance that stops or controls the effect of a poison
- ☐ Nothing
- ☐ Get medical help
- ☐ Take a bath (shower)
- ☐ Other

16. If a pesticide comes in contact with your eyes, what would you do?*

(Mark all that apply. If other, please specify)

- ☐ Rinse with clean water
- ☐ Take antidote; a chemical substance that stops or controls the effect of a poison
- ☐ Nothing
- ☐ Get medical help
- ☐ Take a bath (shower)
- ☐ Other

17. In your opinion, do you think it is important for farmers to use protection when working with pesticides?

(Choose only one that best describes you. If other, please specify)

- ☐ Yes, I think so
- ☐ No, I don't think so
- ☐ Other

18. In your opinion, what would you suggest that the farmers do to protect themselves when working with pesticides for their safety and the safety of their consumers?*(Briefly, specify. May have more than one answer)
19. What can farmers use to protect themselves and the consumers against the negative effects of pesticides when treating crops?(Briefly, specify. May have more than one answer)
20. What can farmers do to protect themselves and the consumers against the negative effects of pesticides when treating crops?(Briefly, specify. May have more than one answer)
21. When buying fresh fruits in a market, are you worried about them being possibly contaminated with pesticides?*(Choose only one that best describes you)
- ☐ Yes, I'm worried
- ☐ No, I'm not worried
22. When buying fresh vegetables in a market, are you worried about them being possibly contaminated with pesticides?*(Choose only one that best describes you)
- ☐ Yes, I'm worried
- ☐ No, I'm not worried
23. When buying tea leaves, are you worried about them being possibly contaminated with pesticides?*(Choose only one that best describes you)
- ☐ Yes, I'm worried
- ☐ No, I'm not worried
24. Do you believe that proper washing with clean water completely remove pesticides from fruits and vegetables?*(Choose only one that best describes you)
- ☐ Yes, it does

☐ No, it doesn't

25. What is your attitude towards the use of pesticides in agriculture?*

(Briefly, specify)

26. Where would you keep your pesticide inventory?*

(Mark all that apply. If other, please specify)

- ☐ Store
- ☐ Kitchen (in sealed container)
- ☐ Bed room
- ☐ Above the cupboard
- ☐ Garage
- ☐ Outside
- ☐ Other

27. Can you identify the adverse or negative health effects associated with pesticides?*

(Choose only one that best describes you)

- ☐ Yes, I can
- ☐ No, I can't

28. If you can identify the adverse or negative health effects associated with pesticides; please mention few examples.

(Mark all that apply. If other, please specify)

- ☐ Death
- ☐ Skin cancer
- ☐ Cancer
- ☐ Mutation
- ☐ Loss of vision
- ☐ Other

29. Have you ever experienced pesticide-related poisoning symptoms such as either severe headaches or skin burns right after a pesticide application?*

(Choose only one that best describes you)

- ☐ Yes, often
- ☐ No
- ☐ Yes, few times
- ☐ I don't know how to identify pesticide poisoning
- ☐ I've never been to those areas right after pesticide application
- ☐ Other

30. Have you ever experienced pesticide-related poisoning symptoms right after coming in contact with pesticides?*

(Mark all that apply. If other, please specify)

- ☐ Yes, often
- ☐ No
- ☐ Yes, few times
- ☐ I don't know how to identify pesticide poisoning
- ☐ Never came in contact with pesticides
- ☐ Other

31. If yes, what were the symptoms that you experienced?

(Mark all that apply. If other, please specify)

- ☐ Headache
- ☐ Skin burn
- ☐ Fatigue
- ☐ Dizziness
- ☐ Loss of appetite with nausea
- ☐ Stomach cramps and diarrhea
- ☐ Blurred vision with excessive tearing
- ☐ Excessive sweating and salivation
- ☐ Slowed heartbeat, less than 50 per minute
- ☐ Rippling of surface muscles (under the skin)
- ☐ Contracted pupils of the eye
- ☐ Incontinence
- ☐ Seizures
- ☐ Unconsciousness

- ☐ Unable to walk properly
- ☐ Chest discomfort
- ☐ Chest tightness
- ☐ Muscle twitching
- ☐ Involuntary urination and bowel movement
- ☐ Perspiration
- ☐ Irritation of the nose and throat
- ☐ Eye irritation
- ☐ Other

32. If you have ever been poisoned by pesticides, could you please estimate how much money you spent on the associated medical treatment (TZS) in the last 2 years?
(e.g. 5,000 TZS)

33. How would you dispose empty containers of pesticides?*

(Mark all that apply. If other, please specify)

- ☐ In a landfill
- ☐ Burn
- ☐ Keep in a store
- ☐ Keep in a garage
- ☐ Bury underground
- ☐ Other

34. Where would you dispose obsolete (expired) pesticides?*

(Mark all that apply. If other, please specify)

- ☐ Dump in the soil (discharge to the soil)
- ☐ Keep in a store
- ☐ Keep in a garage
- ☐ Bury underground
- ☐ Other

35. Would you consume food kept in a container which used to keep pesticides?*

(Choose only one that best describes you)

- ☐ Yes, I would
- ☐ No, I wouldn't
- ☐ Yes, I would if the container is washed properly
- ☐ Other

36. Would you drink water kept in a container which used to keep pesticides?*

(Choose only one that best describes you)

- ☐ Yes, I would
- ☐ No, I wouldn't
- ☐ Yes, I would if the container is washed properly
- ☐ Other

37. What advice do you have for pesticide users?

(Briefly, specify)

38. What advice do you have for pesticide manufacturers?

(Briefly, specify)

39. We would like to know your age (years).*

(e.g. 18)

40. We would like to know your gender.*

- ☐ Male
- ☐ Female

41. We would like to know your highest level of education.*

(Choose only one that best describes you)

- ☐ Primary education
- ☐ Secondary education (Ordinary level)
- ☐ Secondary education (Advanced level)
- ☐ Diploma
- ☐ Advanced diploma
- ☐ University degree

42. We would like to know your profession.*

(e.g. Teacher)

43. We would like to know your location (region, in Tanzania).*

(e.g. Tanga)



APPENDIX C

HOW TO USE THE SOURCE CODE

These are instructions to guide you on installation and use of PestTox plug-ins source code. You need the following to run the model:

- i) The end-user version of Universal Simulator 2 installed on your computer, available for free on <http://www.ecolmod.org>
- ii) The developer version of Universal Simulator 2 together with its source code installed on your computer, available for free on <http://www.ecolmod.org> or <https://github.com/NielsHolst/UniSim2>
- iii) Boost libraries, available for free on <http://www.boost.org>
- iv) The Qt development environment (MinGW), available for free on <https://www.qt.io> and
- v) Text editor Notepad++, available for free on <http://notepad-plus-plus.org>
- vi) R for statistical computing, available for free on <https://www.r-project.org/>

After the above installation, download the Universal Simulator explained book available for free on <http://www.ecolmod.org> and follow the instructions on how to install and build the source code of Universal Simulator 2 (developer version).

After successful installation of the Universal Simulator 2 source code:

- i) Locate PestTox plug-ins found in PestTox folder in the Universal Simulator 2 (developer version), and
- ii) Build the Universal Simulator 2 (refer to Universal Simulator 2 explained book).

APPENDIX D

EQUATIONS USED IN SIMULATION MODELING

D.1 Sowing of seeds

Crop model is a stage-structured model that mimics the phenology of the crop (Holst, 2016). The model keeps track of the growth stages of a crop using degree day values i.e. heat unit accumulations (Dijkhuis, 1956). These degree day values are needed for the crop development throughout the life stages of the crop. Simulated development stages are leaf development, tillering, stem elongation, and senescence. The *outflow* of one stage (a variable) is used as the *inflow* of another stage (a parameter) whereas degree day values are used as *duration* (a parameter) for a particular stage. Also, threshold temperature (T_0) is used as a parameter of the crop model.

D.2 Application (spraying)

Spraying of pesticides depend on favorable conditions for application. This includes allowable wind speed, optimal moisture content, and no rainfall. During the days of spraying, the dose applied is modeled as

$$Dose_{appl} = concentration \times rate \quad (D.1)$$

where $Dose_{appl}$ is the dose applied (g a.i/ha), *concentration* is the concentration of the active ingredient (g a.i/L), and *rate* is the application rate (L/ha). The dose applied is added to the model otherwise during the days of no spray, the dose applied is set to zero.

D.3 Primary distribution

Three main distributions are assumed after the spraying of pesticides, namely drift to air, crop/leaf deposition, and soil deposition (van Zelm et al., 2014). The study assumes the mass balance that the sum of spray drifts, crop deposition, and soil deposition is equal to the dose applied.

D.3.1 Spray drift

The model simulates the fraction of the applied dose which is most likely to be drifted off the field edge. This fraction is then multiplied by the dose applied to get the amount of the dose applied that would most likely be drifted off field.

The fraction of the applied dose carried away by wind drift off field edge (f_d) at a distance x is modeled following van de Zande et al. (2015) as

$$f_d = \frac{a_1}{100} \exp\left(\frac{-x}{b_1}\right) + \frac{a_2}{100} \exp\left(\frac{-x}{b_2}\right) \quad (\text{D.2})$$

where f_d is the fraction of the applied dose carried away by wind drift off field edge, x is the distance off field edge, and a_1 , a_2 , b_1 , and b_2 are regression constants (wind drift coefficients).

The amount of pesticide applied (g a.i/ha) that is carried away by wind drift off field edge ($Dose_{drift}$) is modeled as

$$Dose_{drift} = Dose_{appl} \times f_d \quad (\text{D.3})$$

D.3.2 Crop/leaf deposition

The model makes use of the crop/leaf deposition fractions (Table A.11). These deposition fractions are multiplied by the dose applied to estimate the amount of pesticide deposited on the crop/leaf surfaces. These deposition fractions vary with the growth stage of the crop.

The dose reaching the crop/leaf surfaces through crop deposition is modeled as

$$Dose_{rl} = (Dose_{appl} - Dose_{drift}) \times f_i \quad (\text{D.4})$$

where $Dose_{rl}$ is the amount of applied pesticides that is deposited on the crop/leaf surfaces (g a.i/ha), $Dose_{appl}$ is the amount of pesticide applied (g a.i/ha), $Dose_{drift}$ is the amount of applied pesticide that has drifted off field edge (g a.i/ha), and f_i is the crop/leaf deposition fraction.

D.3.3 Soil deposition

To simulate the dose reaching the topsoil through soil deposition, the model employs mass balances. Therefore, sum of dose deposited on the crop/leaf surfaces and spray drifts is deducted from the dose applied. Hence, the amount of pesticide deposited on the soil ($Dose_{rs}$) (g a.i/ha) is modeled as

$$Dose_{rs} = Dose_{appl} - (Dose_{rl} + Dose_{drift}) \quad (D.5)$$

D.4 Secondary distribution

D.4.1 Crop/leaf processes

The study assumes that volatilization, uptake, photo-degradation, and washoff processes are the main competing processes that remove pesticides from the crop/leaf surfaces (Birkved & Hauschild, 2006; Dijkman et al., 2012).

D.4.1.1 Volatilization

Volatilization is one of the processes that influence the dissipation of pesticides on the crop/leaf surfaces. The evaporation rate at a reference temperature (ER_{Tref}) is modeled following van Wesenbeeck et al. (2008) as

$$ER_{Tref} = \exp(12.2 + 0.933 \ln(VP)) \times 10^{-6} \times 10^4 \quad (D.6)$$

where ER_{Tref} is the evaporation rate at a reference temperature (g/ha/h) and VP is the vapour pressure at a reference temperature (Pa).

Assuming a first order evaporation kinetics of the pesticide, and a constant activation energy over a small temperature range (Schwarzenbach et al., 2003), the rate constant of leaf vaporization (k_{lv}) (g a.i/ha/day) at actual temperature is modeled using a temperature corrected evaporation rate at a reference temperature (i.e. rate constant of leaf vaporization) as

$$k_{lv} = \frac{24 \times ER_{Tref} \exp\left(\frac{1000 E_a}{R} \left(\frac{1}{T_{ref} + 273.15} - \frac{1}{T_{air} + 273.15} \right)\right)}{Dose_{present}} \quad (D.7)$$

where E_a is the activation energy (kJ/mol), which is by default set to 95 kJ/mol (Leistra & Wolters, 2004), R is gas constant 8.314 J/mol/K, T_{air} and T_{ref} are average air temperature and reference temperature at which the vapour pressure was measured (°C), and $Dose_{present}$ is the current concentration of the pesticide on crop/leaf surfaces (g a.i/ha).

D.4.1.2 Uptake

The first-order rate constant (k_{lu}) of the uptake process through cuticles (g a.i/ha/day) is modeled following Baur & Schönherr (1995) as

$$\log(k_{lu}) = a - bMV - \frac{1000E_D}{2.3R(T_{air} + 273.15)} \quad (D.8)$$

The first-order rate constant (k_{lu}) of the uptake process is modeled following Birkved & Hauschild (2006) as

$$\log(k_{lu}) = a - bV - \frac{1000E_D}{2.3R(T_{air} + 273.15)} \quad (D.9)$$

$$k_{lu} = 3600 \times 24 \left(10^{\left(a - bV - \frac{1000E_D}{2.3R(T_{air} + 273.15)} \right)} \right) \quad (D.10)$$

where k_{lu} is the first-order rate constant of the uptake process (g a.i/ha/day), a and b are default values plant species-depended constants (Table A.13), V is the characteristic volume (cm³/mol), MV is the molecular volume (cm³/mol), E_D is the activation energy of diffusion (kJ/mol), T_{air} is average air temperature (°C), and R is gas constant, 8.314 J/mol/K.

Baur & Schönherr (1995) suggested that $E_D \propto e^{MV}$. Therefore, E_D is modeled following Birkved & Hauschild (2006). According to Dijkman et al. (2012), cuticle composition and thickness of citrus and pear leaves are different. Citrus leaves have thicker cuticles and are waxier. Thus, for crops with cuticle characteristics similar to that of citrus leaves (Waxy), E_D is modeled as

$$E_D = 78.875 \exp\left(3.470 \times \frac{MV}{1000}\right) \quad (D.11)$$

For crops with cuticle characteristics similar to that of pear leaves (NonWaxy), E_D is modeled as

$$E_D = 63.6 \exp\left(2.874 \times \frac{MV}{1000}\right) \quad (D.12)$$

Characteristic volume (cm^3/mol) of pesticides is modeled following Birkved & Hauschild (2006) and Dijkman et al. (2012) as

$$V = 2.7717 + 0.9445MV \quad (D.13)$$

where V is the characteristic volume (cm^3/mol), and MV is the molecular volume (cm^3/mol).

D.4.1.3 Degradation

The study assumes that crop/leaf degradation is due to the effect of sunlight (Assalin et al., 2010). Therefore, photo-degradation is the main degradation process modeled. The concentration of OH radicals $[\text{OH}\bullet]$ is interpolated using latitude and day of the year (Table A.14) following Bahm & Khalil (2004). The first-order crop/leaf photo-degradation constant (k_{ld}) (g a.i/ha/day) is modeled as

$$k_{ld} = k_{OH} \times [\text{OH}\bullet] \quad (D.14)$$

where k_{ld} is the first-order photo-degradation rate constant (g a.i/ha/day), k_{OH} is the overall OH• oxidation rate constant ($\text{cm}^3/\text{molecules/h}$), and $[\text{OH}\bullet]$ is the latitude-specific concentration of OH radicals ($\text{molecules}/\text{cm}^3$). Note that, this approach is different from that of PestLCI 2.0 (i.e. light intensity is used to estimate $[\text{OH}\bullet]$).

Day length (d_l) is modeled following Forsythe et al. (1995) as

$$\theta = 0.2163108 + 2 \tan^{-1}\left[0.9671396 \tan\left[0.00860(J - 186)\right]\right] \quad (D.15)$$

$$\phi = \sin^{-1}\left[0.39795 \cos\theta\right] \quad (D.16)$$

$$d_l = 24 - \frac{24}{\pi} \cos^{-1} \left[\frac{\sin \frac{\rho\pi}{180} + \sin \frac{L\pi}{180} \sin \phi}{\cos \frac{L\pi}{180} \cos \phi} \right] \quad (\text{D.17})$$

where k_{ld} is the first-order photo-degradation rate constant (g a.i/ha/day), k_{OH} is the overall OH• oxidation rate constant (cm³/molecules/h), $d_l/24$ is the correction factor for k_{ld} , d_l is the day length (h), θ is the revolution angle (radian) from the day of the year (J), J is the day number of the application day in the year, ϕ is the angular distance (radian) at solar noon between the sun and the equator from the earth orbit revolution angle (sun's declination angle), ρ is the day length coefficient (0.8333 °), π is pi, and L is the latitude (°).

The overall rate constant (k_{ov}) for the crop/leaf processes (g a.i/ha/day) is modeled as

$$k_{ov} = k_{lv} + k_{lu} + k_{ld} \quad (\text{D.18})$$

Leaf dose removed by photo-degradation ($Dose_{ldd}$) (g a.i/ha) is modeled as

$$Dose_{ldd} = Dose_{rl} \times \exp(-k_{ld}) \quad (\text{D.19})$$

Leaf dose removed by volatilization ($Dose_{ldv}$) (g a.i/ha) is modeled as

$$Dose_{ldv} = Dose_{rl} \times \exp(-k_{lv}) \quad (\text{D.20})$$

Leaf dose removed by uptake ($Dose_{ldu}$) (g a.i/ha) is modeled as

$$Dose_{ldu} = Dose_{rl} \times \exp(-k_{lu}) \quad (\text{D.21})$$

D.4.1.4 Washoff

Crop/leaf dose removed by washoff ($Dose_{ldw}$) is modeled following Smith & Carsel (1984) as

$$Dose_{ldw} = Dose_{rl} \times woc \times P \quad (\text{D.22})$$

where $Dose_{ldw}$ is the amount of pesticide washed off by rainfall (g a.i/ha), P is precipitation (mm), and woc is the washoff coefficient per mm of precipitation. The

woc is constant, by default set to 0.01 for any given pesticide regardless of its compound class (Smith & Carsel, 1984). This approach was not used in PestLCI 2.0. Instead, in PestLCI 2.0, dose left on crop/leaf surfaces in the beginning of the next rainfall event was modeled using first-order degradation equation in which time between rainfall events was used as one of the parameters. Other parameters are the overall rate constant for the leaf processes and the dose reaching crop/leaf surfaces.

Sum of dose removed from crop/leaf surfaces by volatilization, uptake, photo-degradation, and washoff processes ($Dose_{ldr}$) (g a.i/ha) is modeled as

$$Dose_{ldr} = Dose_{lde} + Dose_{ldv} + Dose_{ldu} + Dose_{ldw} \quad (D.23)$$

D.4.2 Soil processes

The study assumes that volatilization, biodegradation, runoff, and macropore flow are the main competing processes that remove pesticides from the topsoil (Birkved & Hauschild, 2006; Dijkman et al., 2012).

D.4.2.1 Volatilization

Volatilization is modeled using fugacity approach following Mackay (2001). In this approach, relative tendencies of molecules to flee from the soil are quantified (Schwarzenbach et al., 2003). The study assumes that the topsoil layer is the upper one centimeter section of the soil following Birkved & Hauschild (2006) and Dijkman et al. (2012).

The pH-dependent organic carbon-water partitioning coefficient ($k_{oc,pH}$) is modeled as

$$k_{oc,pH} = \frac{k_{oc}(10^{-pH})}{10^{-pH} + 10^{-pK_a}} \quad (D.24)$$

where $k_{oc,pH}$ is the organic carbon-water partitioning coefficient (L/kg), pH is the pH of the soil, and pK_a is the first dissociation constant of the pesticide.

Soil porosity (S_p) is modeled as

$$S_p = f_a + f_w \quad (D.25)$$

where f_a is the fraction of air in the soil and f_w is the fraction of water in the soil.

Following Dijkman et al. (2012) the study assumes that the vapour pressure at the ambient temperature is determined by assuming that the vapour pressure of the active ingredient increases exponentially with increasing temperature following a tenfold increase per 10 °C.

The vapour pressure ($VP_{T,adap}$) at actual temperature is modeled as

$$VP_{T,adap} = VP \exp\left(\frac{1}{10}(T_{air} - T_{refVP})\right) \quad (D.26)$$

where $VP_{T,adap}$ is the vapour pressure at actual temperature (Pa), VP is the vapour pressure of the pesticide at reference temperature (Pa), T_{air} is the average air temperature (°C), and T_{refVP} is the reference temperature at which vapour pressure was measured (°C).

The Z-value of the pesticide in the water phase (Z_w) is modeled as

$$Z_w = \frac{1000 \times P_{sol}}{VP_{T,adap} \times MW} \quad (D.27)$$

where Z_w is the Z-value of the pesticide in water phase (mol/m³/Pa), P_{sol} is the solubility of the pesticide (g/L), $VP_{T,adap}$ is the vapour pressure at actual temperature (Pa), and MW is the molar mass of the pesticide (g/mol).

The Z-value of the pesticide in the solid matter phase (Z_s) is modeled as

$$Z_s = \frac{k_{oc,pH} f_{oc} \rho_b Z_w}{1 - S_p} \quad (D.28)$$

where Z_s is the Z-value of the pesticide in solid phase (mol/m³/Pa), $k_{oc,pH}$ is the pH-dependent organic carbon-water partitioning coefficient (L/kg), f_{oc} is the fraction of organic carbon in the topsoil, ρ_b is the bulk density of the soil (kg/L), S_p is the soil porosity, and Z_w is the Z-value for the pesticide in water phase (mol/m³/Pa).

The Z-value of the pesticide in the air phase (Z_a) is modeled as

$$Z_a = \frac{1}{R(T_{air} + 273.15)} \quad (D.29)$$

where Z_a is the Z-value of the pesticide in the air phase (mol/m³/Pa), R is the gas constant (J/mol/K), and T_{air} is the average air temperature (°C).

Volume of solids (V_s) in the soil volume susceptible for volatilization is modeled as

$$V_s = P_d \times f_s \times f_{area} \quad (D.30)$$

where V_s is the volume of solids in the soil volume susceptible for volatilization (m^3), P_d is the soil penetration depth (m) which is by default set to 0.01 m (i.e. 1 cm), f_s is the fraction of solids in the soil, and f_{area} is the field area (m^2).

Volume of water (V_w) in the soil volume susceptible for volatilization is modeled as

$$V_w = P_d \times f_w \times f_{area} \quad (D.31)$$

where V_w is the volume of water in the soil volume susceptible for volatilization (m^3), P_d is the soil penetration depth (m), f_w is the fraction of water in the soil, and f_{area} is the field area (m^2).

Volume of air (V_a) in the soil volume susceptible for volatilization is modeled as

$$V_a = P_d \times f_a \times f_{area} \quad (D.32)$$

where V_a is the volume of air in the soil volume susceptible for volatilization (m^3), P_d is the soil penetration depth (m), f_a is the fraction of air in the soil, and f_{area} is the field area (m^2).

The VZ value (mol/Pa) for all three phase i.e. solid matter, air and water phases is modeled as

$$VZ = V_s Z_s + V_w Z_w + V_a Z_a \quad (D.33)$$

The water viscosity (η) is modeled by using a regressed equation ($R^2 = 0.9999$) following Dijkman et al. (2012) as

$$\eta = \left(\frac{1.3374 + 0.0017(T_{soil})}{1 + 0.0185(T_{soil})} \right)^2 \quad (D.34)$$

where η is the water viscosity at soil temperature (centipoise), and T_{soil} is the daily soil temperature ($^{\circ}C$).

The molecular diffusivity (B_w) of the pesticide in water is modeled as

$$B_w = \frac{1.326 \times 10^{-4}}{10^4 \times \eta^{1.14} \times MV^{0.589}} \quad (D.35)$$

where B_w is the molecular diffusivity of the pesticide in water (m^2/s), η is the viscosity (centipoise), and MV is the molar volume of the pesticide (cm^3/mol).

The effective diffusivity (B_{ew}) of the pesticide in water is modeled as

$$B_{ew} = \frac{B_w (f_w)^{10/3}}{(f_a + f_w)^2} \quad (D.36)$$

where B_{ew} is the effective diffusivity of the pesticide in water (m^2/s), B_w is the molecular diffusivity of the pesticide in water (m^2/s), f_w is the fraction of water in the soil, and f_a is the fraction of air in the soil.

The diffusion path length (D_{pl}) is modeled as

$$D_{pl} = \frac{P_d}{2} \quad (D.37)$$

where D_{pl} is the diffusion path length (m), and P_d is the depth of layer or penetration depth (m) from which pesticide volatilizes.

The D-value for pesticide in water (D_w) is modeled as

$$D_w = 3600 \times 24 \times \frac{B_{ew} f_{area} Z_w}{D_{pl}} \quad (D.38)$$

where D_w is the D-value for pesticide in water ($\text{mol}/\text{m}^3/\text{day}$), B_{ew} is the effective diffusivity of the pesticide in water (m^2/s), f_{area} is the field area (m^2), Z_w is the Z-value of the pesticide in the water phase ($\text{mol}/\text{m}^3/\text{Pa}$), and D_{pl} is the diffusion path length (m).

The molecular diffusivity of pesticide in air (B_a) for the diffusion of organic molecules in air is modeled following Fuller et al. (1966) and Schwarzenbach et al. (2003) as

$$B_a = \left(\frac{1}{10^4} \right) \left(\frac{1}{10^3} \right) \left(\frac{(T_{air} + 273.15)^{1.75} \left(\frac{1}{MW_{air}} + \frac{1}{MW} \right)^{0.5}}{P_{air} \left((MV_{air})^{1/3} + (MV)^{1/3} \right)^2} \right) \quad (D.39)$$

where B_a is the molecular diffusivity of the pesticide in air (m^2/s), T_{air} is the daily average air temperature ($^{\circ}C$), MW_{air} is the molar weight of the air (g/mol) which is by default set to 28.97 g/mol (Khélifa et al., 2007; Lugg, 1968), MW is the molar mass of the pesticide (g/mol), P_{air} is the gas phase pressure of the air (atm) which is by default set to 1 atm , MV_{air} is the volume of one mol of air (i.e. molar volume air) (cm^3/mol) which is by default set to 20.1 cm^3/mol (Fuller et al., 1966; Lugg, 1968), and MV is the molecular volume of the pesticide (cm^3/mol).

The effective diffusivity of the pesticide in air (B_{ea}) is modeled as

$$B_{ea} = \frac{B_a (f_a)^{10/3}}{(f_a + f_w)^2} \quad (D.40)$$

where B_{ea} is the effective diffusivity of the pesticide in air (m^2/s), B_a is the molecular diffusivity of the pesticide in air (m^2/s), f_a is the fraction of air in the soil, and f_w is the fraction of water in the soil.

The D-value for pesticide in air (D_a) is modeled as

$$D_a = 3600 \times 24 \times \frac{B_{ea} \times f_{area} \times Z_a}{D_{pl}} \quad (D.41)$$

where D_a is the D-value for pesticide in air ($mol/m^3/day$), B_{ea} is the effective diffusivity of the pesticide in air (m^2/s), f_{area} is the field area (m^2), Z_a is the Z-value of the pesticide in the air phase ($mol/m^3/Pa$), and D_{pl} is the diffusion path length (m).

The transport velocity through the boundary layer (k_e) is modeled as

$$k_e = \frac{B_a}{BL_{air}} \quad (D.42)$$

where k_e is the transport velocity through the boundary layer (m/s), B_a is the molecular diffusivity of the pesticide in air (m^2/s), and BL_{air} is the thickness of the air boundary layer for the evaporation which is by default set to 0.00475 m (Dijkman et al., 2012).

The D-value of the pesticide diffusion through the air boundary layer (D_{bl}) is modeled as

$$D_{bl} = 3600 \times 24 \times k_e \times f_{area} \times Z_a \quad (D.43)$$

where D_{bl} is the D-value of the pesticide diffusion through the air boundary layer ($\text{mol/m}^3/\text{day}$), k_e is the transport velocity through the boundary layer (m/s), f_{area} is the field area (m^2), and Z_a is the Z-value of the pesticide in the air phase ($\text{mol/m}^3/\text{Pa}$).

The total D-value (D_v) of the volatilization process is modeled as

$$D_v = \frac{1}{\left(\frac{1}{D_{bl}} + \frac{1}{D_a + D_w} \right)} \quad (D.44)$$

where D_v is the total D-value of the volatilization process ($\text{mol/m}^3/\text{day}$), D_{bl} is the D-value of the pesticide diffusion through the air boundary layer ($\text{mol/m}^3/\text{day}$), D_a is the D-value of the pesticide in air ($\text{mol/m}^3/\text{day}$), and D_w is the D-value of the pesticide in water ($\text{mol/m}^3/\text{day}$).

The rate constant of pesticide evaporation (k_{sv}) from the soil is modeled as

$$k_{sv} = \frac{D_v}{VZ} \quad (D.45)$$

where k_{sv} is rate constant of pesticide evaporation from the soil (day^{-1}), D_v is the total D-value of the volatilization process ($\text{mol/m}^3/\text{day}$), and VZ is the total VZ value (mol/Pa) of all three phase i.e. solid, air, and water phases.

Dose of pesticide in soil solids ($Dose_{rss}$) is modeled as

$$Dose_{rss} = \left(\frac{V_s Z_s}{VZ} \right) (Dose_{rs} + Dose_{ldw}) \quad (D.46)$$

Fraction of pesticide in soil solids (f_{rss}) is modeled as

$$f_{rss} = \frac{V_s Z_s}{VZ} \quad (D.47)$$

where $Dose_{rss}$ is the dose of pesticide in soil solids (g a.i./ha), f_{rss} is the fraction of pesticide in soil solids, V_s is the volume of solids in the soil volume susceptible for

volatilization (m^3), Z_s is the Z-value of the pesticide in solid phase ($mol/m^3/Pa$), $Dose_{rs}$ is the dose of the applied pesticide reaching the soil ($g\ a.i./ha$), and VZ is the total VZ value of all three phase i.e. solid matter, air, and water phases (mol/Pa).

Dose of pesticide in soil water ($Dose_{rsw}$) is modeled as

$$Dose_{rsw} = \left(\frac{V_w Z_w}{VZ} \right) (Dose_{rs} + Dose_{ldw}) \quad (D.48)$$

Fraction of pesticide in soil water (f_{rsw}) is modeled as

$$f_{rsw} = \frac{V_w Z_w}{VZ} \quad (D.49)$$

where $Dose_{rsw}$ is the dose of pesticide in soil water i.e. dose of pesticides reaching soil in the soil water ($g\ a.i./ha$), f_{rsw} is the fraction of pesticide in soil water i.e. fraction of pesticides reaching soil in the soil water, V_w is the volume of water in the soil volume susceptible for volatilization (m^3), Z_w is the Z-value of the pesticide in water phase ($mol/m^3/Pa$), $Dose_{rs}$ is the dose of the applied pesticide reaching the soil ($g\ a.i./ha$), and VZ is the total VZ value of all three phase i.e. solid matter, air, and water phases (mol/Pa).

Dose of pesticide in soil air ($Dose_{rsa}$) is modeled as

$$Dose_{rsa} = \left(\frac{V_a Z_a}{VZ} \right) (Dose_{rs} + Dose_{ldw}) \quad (D.50)$$

Fraction of pesticide in soil air (f_{rsa}) is modeled as

$$f_{rsa} = \frac{V_a Z_a}{VZ} \quad (D.51)$$

where $Dose_{rsa}$ is the dose of pesticide in soil air i.e. dose of pesticides reaching soil in the soil air ($g\ a.i./ha$), f_{rsa} is the fraction of pesticide in soil air i.e. fraction of pesticides reaching soil in the soil air, V_a is the volume of air in the soil volume susceptible for volatilization (m^3), Z_a is the Z-value of the pesticide in air phase ($mol/m^3/Pa$), $Dose_{rs}$ is the dose of the applied pesticide reaching the soil ($g\ a.i./ha$), and VZ is the total VZ value (mol/Pa) of all three phase i.e. solid, air, and water phases.

Dose of pesticide vaporized ($Dose_{sv}$) (g a.i/ha) is modeled as

$$Dose_{sv} = (Dose_{rs} + Dose_{ldw})(f_{rsa} + f_{rsw})\exp(-k_{sv}) \quad (D.52)$$

D.4.2.2 Degradation

Pesticide degradation in the topsoil is mostly via microbial degradation, chemical degradation, and photo-degradation (Büyüksönmez et al., 1999). In this study, first-order degradation kinetics (Beulke & Brown, 2001) is assumed. Therefore, the dissipation dynamics of the pesticides in the topsoil is modeled following Huan et al. (2013) as

$$\frac{\partial C}{\partial t} = -kt \quad (D.53)$$

Its exponential form is expressed as

$$C_t = C_0 \times \exp(-kt) \quad (D.54)$$

where C is the concentration (g a.i/ha), t is the time (days), k is the biodegradation rate (day^{-1}), C_t is the concentration at time t (g a.i/ha), and C_0 is the concentration at time zero (g a.i/ha).

The first-order rate constant for soil biodegradation (k) (day^{-1}) is modeled using the half-life (days) or days to 50% dissipation (DT_{50}) following Mueller et al. (2010) as

$$k = \frac{0.693}{DT_{50}} \quad (D.55)$$

Temperature and soil moisture influence soil biodegradation. Therefore, the rate constant of biodegradation in the topsoil (k_{sd}) (day^{-1}) is corrected using temperature and soil moisture following Costantino et al. (1998).

Soil temperature (T_{soil}) is modeled following Donatelli et al. (2014) as

$$T_{soil} = \left(\frac{d_l}{24} \times T_{\max} \right) + \left(1 - \frac{d_l}{24} \right) \times T_{\min} \quad (D.56)$$

where T_{soil} is the soil temperature ($^{\circ}\text{C}$), $T_{\max}$ is the daily maximum temperature ($^{\circ}\text{C}$), $T_{\min}$ is the daily minimum temperature ($^{\circ}\text{C}$), and d_l is the day length (h).

The temperature correction factor ($f_{sd,T}$) is modeled following FOCUS (2006) as

$$f_{sd,T} = \begin{cases} Q_{10}^{(T_{soil} - T_{ref})/10} & \text{for } T_{soil} > 0^\circ\text{C} \\ 0 & \text{otherwise} \end{cases} \quad (\text{D.57})$$

where $f_{sd,T}$ is the temperature correction factor, T_{soil} is the soil temperature ($^\circ\text{C}$), T_{ref} is the reference temperature at which the half-life time of pesticide was determined in the soil ($^\circ\text{C}$), and Q_{10} is the increase in biodegradation rate per 10 $^\circ\text{C}$ (Q_{10} is used to model the temperature correction factor for soil biodegradation rate assuming that the soil biodegradation rate increases with a fixed factor per 10 $^\circ\text{C}$) (Boesten et al., 1996; Dijkman et al., 2012). The value of Q_{10} is by default set to 2.58 (EFSA, 2007). Note that, an error has been corrected from PestLCI 2.0.

The volumetric soil moisture content is modeled as

$$V_{smc} = \frac{f_w}{S_p} \quad (\text{D.58})$$

where f_w is the fraction of water in the soil and S_p is the soil porosity.

The moisture correction factor is modeled following FOCUS (2006) as

$$f_{sm} = \begin{cases} \left(\frac{V_{smc}}{V_{rsmc}} \right)^{r_f} & \text{for } V_{smc} < V_{rsmc} \\ 1 & \text{otherwise} \end{cases} \quad (\text{D.59})$$

where f_{sm} is the moisture correction factor, V_{smc} is the volumetric soil moisture content, V_{rsmc} is the reference soil moisture content; by default set to 0.5 (Dijkman et al., 2012), and r_f is the exponential response factor for the influence of moisture content on biodegradation rate. The value of r_f is by default set to 0.7 (FOCUS, 2006).

The rate constant for biodegradation (k_{sd}) of pesticide in the topsoil is modeled as

$$k_{sd} = k \times f_{sd,T} \times f_{sm} \quad (\text{D.60})$$

where k_{sd} is the rate constant for biodegradation of pesticide in the topsoil (day^{-1}), k is the rate constant of biodegradation in soil (day^{-1}), $f_{sd,T}$ is the temperature correction factor, and f_{sm} is the moisture correction factor.

Dose of pesticide degraded ($Dose_{sd}$) (g a.i/ha) is modeled as

$$Dose_{sd} = f_{rss} (Dose_{rs} + Dose_{ldw}) \exp(-k_{sd}) \quad (D.61)$$

where f_{rss} is the fraction of pesticide in the soil solids.

D.4.2.3 Runoff

The amount of pesticides removed from the topsoil by runoff is modeled following Berenzen et al. (2005). The study assumes that runoff is caused by rainfall (Reus et al., 1999). The runoff amount, Q (mm) is modeled following Lutz (1984) and Maniak (1992). Additional information is given by Berenzen et al. (2005) and Dijkman et al. (2012).

For loamy soils (fraction of sand <0.5), Q is modeled as

$$Q = \begin{cases} -0.0041626 P + 0.0040395 P^2 - 9.0361 \times 10^{-6} P^3 & \text{for } Q < 0 \\ -0.061108 - 0.0041626 P + 0.0040395 P^2 - 9.0361 \times 10^{-6} P^3 & \text{otherwise} \end{cases} \quad (D.62)$$

For sandy soils (fraction of sand >0.5), Q is modeled as

$$Q = \begin{cases} -0.011377 P + 0.0026284 P^2 - 5.8564 \times 10^{-6} P^3 & \text{for } Q < 0 \\ -0.016427 - 0.011377 P + 0.0026284 P^2 - 5.8564 \times 10^{-6} P^3 & \text{otherwise} \end{cases} \quad (D.63)$$

where Q is the runoff amount (mm) and P is the daily rainfall (mm).

The fraction of rain runoff (f_{rr}) is modeled as

$$f_{rr} = \frac{Q}{P} \quad (D.64)$$

Soil runoff is corrected using slope of the field and buffer zones following Beinat & Berg (1996).

Slope correction factor is modeled as

$$F_{slope} = \begin{cases} 0.02153S + 0.001423S^2 & \text{for } S < 0.2 \\ 1 & \text{for } S > 0.2 \\ 0.02153 + 0.0014426 & S = 0 \end{cases} \quad (D.65)$$

Buffer zone correction factor is modeled as

$$F_{buffer} = 0.83^{wbz} \quad (D.66)$$

where S is the slope of the field (fraction) and wbz is the width of the buffer zone (m). The study assumes that if the buffer zone is covered with vegetation, resistance is created against the runoff. If buffer zone is not covered with vegetation, the buffer zone width is set to zero (Berenzen et al., 2005).

The overall correction factor (F) is modeled as

$$F = F_{slope} \times F_{buffer} \quad (D.67)$$

The ratio of dissolved and sorbed pesticide (K_d) is modeled as

$$K_d = k_{oc,pH} \times f_{om} \quad (D.68)$$

where $k_{oc,pH}$ is the pH-dependent organic carbon-water partitioning coefficient (L/kg) and f_{om} is the fraction of organic matter in the topsoil.

The amount of pesticide that runoff to surface water ($Dose_{sr}$) is modeled following Berenzen et al. (2005) as

$$Dose_{sr} = \frac{Q}{P} F \left(\frac{1}{1 + K_d} \right) (Dose_{rs} + Dose_{ldw}) \quad (D.69)$$

where $Dose_{sr}$ is the amount of pesticide that runoff to surface water (g a.i/ha), Q is the runoff amount (mm), P is the daily rainfall (mm), F is the overall correction factor, K_d is the ratio of dissolved and sorbed pesticide, $Dose_{rs}$ is the amount of pesticide deposited in the topsoil (g a.i/ha), and $Dose_{ldw}$ is the amount of pesticide on the crop/leaf surfaces (g a.i/ha) that is washed off during rainfall event (this amount is assumed to be washed-off to the soil surface, and it is available for runoff).

D.4.2.4 Macropore flow

The mobile pore fraction ($f_{pore, mobile}$) depends on soil composition (Hall, 1993), and it is modeled as

$$f_{pore, mobile} = 0.72 f_{sand} + 0.35 f_{silt} + 0.14 f_{clay} \quad (D.70)$$

where f_{sand} , f_{silt} , and f_{clay} are fractions of sand, silt, and clay in the soil, f_a is the fraction of air in the soil, and f_w is the fraction of water in the soil.

The immobile pore fraction ($f_{pore,immobile}$) is modeled as

$$f_{pore,immobile} = (f_a + f_w) - f_{pore,mobile} \quad (D.71)$$

The study assumes that precipitation water flows into macropores only if water fills both immobile and slow flowing pores. Flow rate of 0.35 m/day is assumed for slow mobile water.

The mobile pore volume ($V_{pore,mobile}$) (m^3/m^3) in the topsoil i.e. volume of pores in the topsoil in which water is mobile is modeled as

$$V_{pore,mobile} = f_{pore,mobile}(f_a + f_w) \quad (D.72)$$

The immobile pore volume in the topsoil ($V_{pore,immobile}$) (m^3/m^3) i.e. volume of pores in the topsoil in which water is not mobile is modeled as

$$V_{pore,immobile} = (f_a + f_w) - V_{pore,mobile} \quad (D.73)$$

The volume of water that can be taken up by the topsoil in one hour i.e. a one hour storage capacity (C) (m/h) is modeled as

$$C = 0.01 \times V_{pore,immobile} + (1 - f_{mp}) \times \left(\frac{f_{rate}}{24} \right) \times V_{pore,mobile} \quad (D.74)$$

where f_{mp} is the fraction of macropore i.e. pore fractions which acts as macropores. This fraction is by default set to 0.3, and f_{rate} is the speed of the slow-flowing mobile water which is by default set to 0.35 m/day following Hall (1993).

The volume of water that is used to fill the water pore volume in the soil i.e. used capacity (m/h) is modeled as

$$C_{used} = \left(\frac{f_w}{f_a + f_w} \right) C \quad (D.75)$$

The volume of water that is free in the soil i.e. free capacity (m/h) is modeled as

$$C_{free} = C - C_{used} \quad (D.76)$$

Free capacity ($P_{capacity}$) re-calculated as the amount of rain that can be taken up in one hour (mm/h) is modeled as

$$P_{capacity} = 1000 \times C_{free} \quad (D.77)$$

Rainfall required before macropore flow starts ($P_{req,h}$) (mm/h) is modeled as

$$P_{req,h} = \frac{P_{capacity}}{1 - f_{rr}} \quad (D.78)$$

where f_{rr} is the fraction of rain runoff.

Rainfall required before macropore flow starts ($P_{req,day}$) (mm/day) is modeled as

$$P_{req,day} = 24 \times P_{req,h} \quad (D.79)$$

The integrated distribution of precipitation on a rainy day (P_i), giving the frequency distribution, based on an exponential distribution of precipitation following Eagleson (1972) is modeled as

$$P_i = \exp\left(\frac{t_{p,event}}{24 \times P} \times P_{req,day}\right) \quad (D.80)$$

where $t_{p,event}$ is the number of hours precipitation occurs on a rainy day (h) and P is the daily rainfall (mm).

The amount of the dose applied that goes into macropores (D_{mp}) (g a.i/ha) is modeled as

$$D_{mp} = P_i (Dose_{rs} + Dose_{ldw}) f_{rs} \left(\frac{T_f}{7.5} \right) \quad (D.81)$$

where T_f is the tillage factor (Table A.15).

D.5 Population dynamics

Population dynamics model is a stage-structured model that implements a distributed delay process (Manetsch, 1976; Vansickle, 1977). The implementation is described in detail by Holst (2016). Both *duration* and k are the parameters that

determine the average and variance of stage duration with k being the number of age classes in the stage (Holst, 2016). A small value of k (e.g. 30) is generally preferred to speed up the computational speed without having much change in the results (Gutierrez et al., 1988). The model time determines the time scale used by the stage model. The *outflow* (a variable) from the stage in latest time step is used as the *inflow* (a parameter) into the following stage in the next time step. The *initial* parameter is used as *inflow* into the chosen stage at time zero. A stage model *eggsToBeLaid* allows the modeling of the second generation (population dynamics) with *growthFactor* (a parameter) corresponding to the life time fecundity of an adult (Holst, 2016). Parameters used to determine the number of eggs to be laid include *sexRatio*, *eggsPerFemale*, and current *population*. The model also estimates pesticide-induced daily mortality rates (*mortalityRate*). This means that for a given day, if the mortality rate is 60% and the number of tadpoles is n , then the number of dead tadpoles on that day would be $0.6n$ (Holst, 2016). Pesticide-induced daily mortality rates are calculated using predicted daily exposure concentrations by applying the dose response equation following Johnston et al. (2014), Focks et al. (2014a), Galic et al. (2014), and Focks et al. (2014b) as

$$mortalityRate = \frac{1}{1 + \exp(-slope \times (\ln C_t - \ln LC_{50}))} \quad (D.82)$$

where C_t is the daily predicted pesticide concentration in the water body (mg a.i/L) at time t , LC_{50} is the 48 h lethal concentration that kills 50% of the frogs' population (mg a.i/L), and *slope* is a sigmoidal function (volume/amount) that scale the specific toxicity of the simulated pesticide (Galic et al., 2014).

D.6 Sensitivity analysis

The sensitivity analysis of the model outputs due to the variations of input parameters in the simulation period is modeled using Latin Hypercube Sampling (LHS) method (Holst, 2016). Repeated simulation is achieved using the number of iterations (e.g. 30). Random numbers can be drawn from either of the four statistical distributions; normal, uniform, lognormal, and loguniform. A new random number is generated before each simulation run. Example, for a uniform distribution, for a

uniform distribution, the model generates random numbers uniformly within the chosen interval (e.g. between 1 and 10). Lastly, the degree that the inputs determine the outputs is analyzed.



BIOGRAPHY

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