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Modeling of the residue transport of lambda cyhalothrin, cypermethrin, malathion and endosulfan in three different environmental compartments in the Philippines

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ABSTRACT

This study aims to determine the environmental transport and fate of the residue of four Philippines priority chemicals; i.e., lambda cyhalothrin (L-cyhalothrin), cypermethrin, endosulfan and malathion, in three different environmental compartments (air, water and soil). In the Philippines, pesticide application is the most common method of controlling pests and weeds in rice and vegetable farming. This practice aided the agricultural industry to minimize losses and increase yield. However, indiscriminate use of pesticides resulted to adverse effects to public health and environment. Studies showed that 95% of the applied pesticides went to non-target species. Data from previous studies in Pagsanjan Laguna, Philippines were used as input data. Dispersion, Gaussian plume, and regression equations were employed to simulate the behavior of L-cyhalothrin, cypermethrin, endosulfan and malathion in air, water and soil. Substance decay was calculated using first order reaction. This study showed how L-cyhalothrin, cypermethrin, endosulfan, and malathion behaved in the environment after release from nozzle spray, and its possible duration of stay in the environment. It will also show a tool in determining the percolation depth through soil by endosulfan. This tool can be utilized in determining the depth of contaminated soil during remediation strategic planning and project implementation of similar environmental condition.

1. Introduction

In the Philippines, pesticides play a vital role in the increase and sustainability of food production. Pesticide manifests greater productivity that alleviates crop production problems such as pest infestation and poor product yield. Total pesticides usage in the Philippines increased from 3.74 kt in 1977 to 10.8 kt in 1991 [1] in Ref. [2]. This is an increase pesticide usage of 0.5 kt per year during 14 yr span. Considering the country's increase in population growth with constant rate of pesticide usage, the probable amount of pesticide usage in the Philippines may reach to 22.8 kt by 2015.

Pesticide transport and fate are significantly affected by climate and the environmental condition of the Philippines. It has two pronounced seasons; i.e., the wet and dry. Rice farmers around Pagsanjan-Lumban catchment (Fig. 1) applied pesticides three times per season. Vegetable growers in Lucban and Laguna area applied pyrethroid based insecticide such as L-cyhalothrin and cypermethrin five times throughout the cropping season. Other insecticides such as malathion and endosulfan were applied two to four times within the cropping season [2]. Depending on the types

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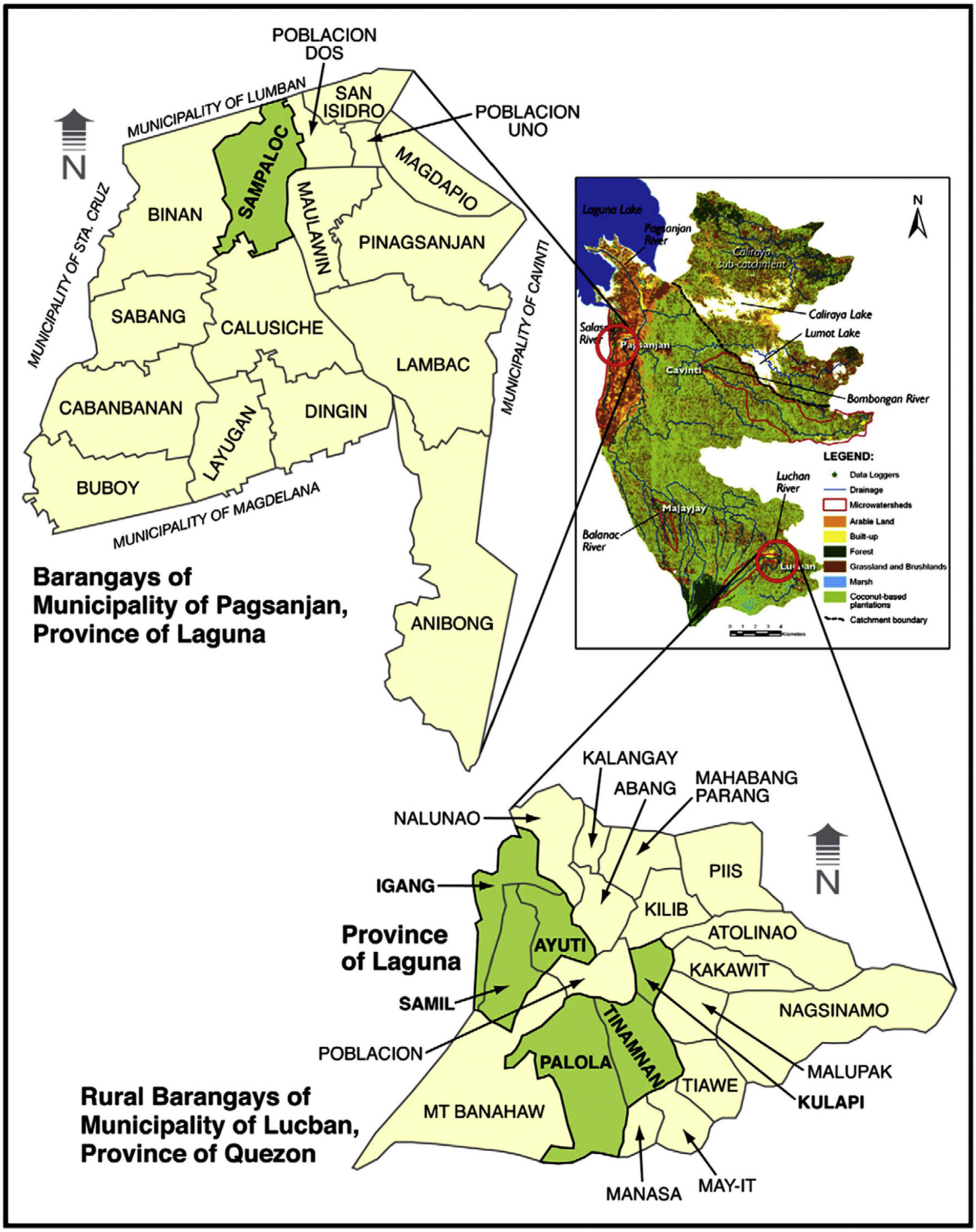

Fig. 1. Map of the Pagsanjan-Lumban catchment [2].

of vegetables, the cropping period ranges from two to six months. Farmers in the Philippines commonly applied pesticides to crops by spraying an aqueous emulsion and/or suspension using backpack knapsack sprayer. Backpack knapsack sprayer is a compressed air sprayer with a harness that allows it to be carried on the operator's back. Another type of backpack sprayer has a hand-operated hydraulic pump that forces liquid pesticide through a hose and one or more nozzles. Both types of sprayer have nozzle.

Cypermethrin [$C_{22}H_{19}O_3NCl_2$] has low solubility in water (K_w = 0.009 mg L^{-1} at 20 °C) and has higher affinity to soil and sediment particles (Koc = 6.1 × 10^4 mL g^{-1}). The field dissipation, aerobic, and anaerobic half-life are 4–12, 6–20, and < 14 d, respectively [3]. Cypermethrin is immobile in soil, however, its degradates (i.e., phenoxybenzoic acid and dichlorovinyl acid) are mobile in soil. Endosulfan in river system is stable at pH 5 with increasing disappearance at pH 7 and 9 by chemical hydrolysis [4]. L-cyhalothrin has low water solubility, not volatile, stable in water at pH < 8 with half-life greater than 3 wk under natural irradiation [5]. Malathion is non-persistent, degrades quickly in soil (3 d) compared with water (11 d). However, when malathion is absorbed by soil, degradation becomes faster [6] which tends to become non-mobile in the environment and does not exist in large scale. However, there are studies showing transport of malathion over far distances.

During release of pesticides in the environment, studies showed that 95–98% goes to non-target areas [7] and 30% is lost due to airborne drift [8]. Processes like adsorption, transfer, breakdown and degradation happen during its release in the environment. Transfer in air transpires during spray drift and volatilization while transfer in water happens during runoff and leaching. In soil, pesticides tend to be adsorbed onto soil, absorbed by plants and microorganisms or transferred from one area to another during crop removal. There are two main routes by which pesticides enter the soil: (1) spray drift to soil during foliage treatment plus wash-off from treated foliage, and (2) release from granulates applied directly to the soil [7]. Soil is the main medium for pesticide deposition [9] hence soil types at the concerned areas were assessed. Paningbatan et al. [10] studied the movement and accumulation of endosulfan used by rice farmers in Laguna.

Pesticides can migrate from treated fields to air, nearby land areas and water bodies. Information on the actual input of pesticides into the environment is crucial for proper risk assessment and the rational design of risk reduction measure. Concern about the environmental and health impact of indiscriminate use of pesticide [11] has prompted research into the environmental fate and transport of these agricultural chemicals [7]. Results of modeling studies on environmental fate and transport are helpful in making remediation plan and strategies. Toxicity to non-target species and environmental safety of these chemicals are attracting global attention. Under laboratory conditions, pyrethroid insecticides like L-cyhalothrin and cypermethrin in Pagsanjan-Lumban catchment was found to be highly toxic to freshwater shrimp and *Tilapia fingerling* [12].

Malathion degrades into malaoxon which is 10-fold more toxic based on 96-h LC_{50} for frog embryos [13]. Studies show that prolong human exposure to malathion adversely affects the respiratory, gastrointestinal, and neurological systems [14]. Endosulfan on the other hand was issued by US Environmental Protection Agency with a product cancellation and phase out order in July 2010 [15] due to the carcinogenic effects, congenital physical disorders, mental disabilities and death in farm workers and communities across the globe [16]. Further, endosulfan has been included by the Swedish Chemicals Agency in the Stockholm Convention on Persistent Organic Pollutants in May 2011 [17]. Therefore, a study aimed to determine the fate and transport of L-cyhalothrin, cypermethrin, endosulfan and malathion, is important thus the objective of this study. Results of this modeling study helps in understanding the transport and fate of L-cyhalothrin, malathion, and endosulfan. This will significantly contribute in decision making for the development of guidance document and values, policy development, remediation planning and strategies for project implementation.

2. Materials and methods

Primary method used in this study was on based regression statistical modeling and prediction until 2020 using secondary data. This method was also used by Frykman and Deutsch [18]. Leibniz Institute for Prevention Research and Epidemiology in Bremen, Germany in their epidemiological study in Germany [19] as well as Robert Koch Institute in Berlin, Germany in disease control prevention research [20].

Seven groups (composed of twenty) farmers surveyed in Lucban grew rice, tomatoes, stringbeans, bittergourd, raddish, cabbage, coconut, chayote, celery, spinach and coriander while farmers in Pagsanjan grew mango and rice. Almost all growers in Lucban and Pagsanjan-Lumban, Laguna, regardless of crop grown, used pyrethroid-based insecticides L-cyhalothrin and cypermethrin. Other pesticides used were malathion and endosulfan.

2.1. Assessment of secondary data

Fig. 1 illustrates the catchment indicating the location of the concerned areas. According to the watershed information portal of the Philippines, the topography of the area is moderately sloping to rolling with about 18% slope. Average rainfall in Lucban and Pagsanjan is 2565 mm with average temperature and wind speed of 27 °C and 2.06 m s^{-1}, respectively. The exposure to typhoons of the area is 16% [18] and Philippines mean annual exposure to typhoon is 22% which means the Lumban-Pagsanjan catchment experienced an average typhoon exposure of 3–4 times in a year. Cruz et al. [21] provide biophysical characteristics of Pagsanjan-Lumban catchment which were useful to the modeling activity. The farming details and usage of L-cyhalothrin are illustrated in Table 1.

Empirical data [10,22] were used for the simulation and prediction of L-cyhalothrin, cypermethrin, malathion, and endosulfan in air, water and soil. This study considered that pesticide application was carried out five times throughout the cropping season. Potential load of cypermethrin is shown in Table 2. Data shown in Table 3 were used to determine the behavior, extract mathematical model, and illustrate the effect of time on the concentration of endosulfan at different soil depths.

2.2. Modeling of fate and transport of four pesticides

This section illustrates how modeling was carried out to understand the behavior of the four chemicals in the environment.

2.2.1. Pesticide residue in air

The Gaussian plume dispersion model was used to describe the transport and the dispersion of the drops after their ejection from the nozzles (Fig. 2). Equations (1) and (2) were used to predict the concentration and drop deposition rate of lambda cyhalothrin at certain distances [23]:

$$C(x,y,z,H_s) = \frac{Q_m}{2\pi\sigma_y\sigma_z U} exp\left\{\left(-\frac{y^2}{2\sigma_y^2}\right) - \left(\frac{\left(z-\left(H_s-\frac{v_p x}{U}\right)\right)^2}{2\sigma_z^2}\right)\right\} \quad (1)$$

$$q_m = v_p C(x,y,0:H_s) = \frac{v_p Q_m}{2\pi\sigma_y\sigma_z U}\exp\left\{\left(-\frac{y^2}{2\sigma_y^2}\right) - \left(\frac{\left(H_s-\frac{v_p x}{U}\right)^2}{2\pi\sigma_z^2}\right)\right\} \quad (2)$$

where *C(x,y,z; H)* denotes for particles concentration at a receptor (g m^{-3}); *x, y, z* is horizontal, transversal and vertical distances (m), respectively; H_s is effective height of nozzle with respect to the ground (m); Q_m is the rate of mass release from the nozzle (g s^{-1}); σ_y, σ_z, is horizontal and vertical Gaussian dispersion parameters (m); U is mean wind speed according to the x-axis (m s^{-1}); v_p is gravity settling velocity (m s^{-1}); and q_m is drop deposition rate (g m^{-2} s^{-1}). Seven groups of farmers among the 38 who used L-cyhalothrin were chosen in this study.

2.2.2. Pesticide residue in water

ANOVA (Analysis of Variance) for regression was used in the determination of coefficient (r^2) of analytical curves

Table 1
Lambda-cyhalothrin usage of rice farmers in Barangay Samil, Lucban, Quezon and Barangay Sampaloc, Pagsanjan, Laguna.

Growers	Crop	Size of farm (ha)	Terrain	Season of planting	Months of planting	Frequency of application	Spray loading (TL)[a]
Barangay Samil							
2	Rice	0.5	Terrace	Wet, dry	December	2	5
3	Rice	0.8	Terrace	Dry	November	2	4
4	Rice	1.5	Terrace	Dry	December	2	4
5	Rice	1.0	Terrace	Dry	November	2	4
6	Rice	0.5	Terrace	Dry	December	1	3
7	Rice	1.0	Terrace	Dry	December	2	4
9	Rice	1.5	Terrace	Dry	December	1	4
Barangay Sampaloc							
1	Mango	5	Plain			2	20/tree
2	Rice	2	Plain	Dry	December–January	1	20
3	Rice	2	Plain	Dry	December–January	1	20
4	Rice	2	Plain	Dry	December–January	1	20
5	Rice	2	Plain	Dry	December–January	1	16–20
6	Rice	6	Plain	Dry	December–January	2	48–60
7	Rice	6	Plain	Wet	June–July	4	60
9	Rice	1	Plain	Wet	June–July	1	10

[a] 1 TL = 16 L; Application rate to all areas is 250 g.a.i ha^{-1}.
Source: [2].

Table 2
Approximation of Cypermethrin potential load being applied in the regions around Pagsanjan, Laguna.

Chemical	Average application rate (g.a.i ha^{-1})	Approximate area grown in region (ha)	Number of applications in season	Approximate load applied (kg)	Approximate annual amount potentially moving off-site (kg)
Cypermethrin	50	500	2	100	0.3

Source: [2].

Table 3
Endosulfan concentration (μg L^{-1}) at different soil depth.

Soil depth (cm)	Days after spraying			
	6	31	59	73
0	0.037	0.000	0.038	0.000
25	2.467	0.000	0.483	0.000
50	0.039	0.060	2.620	0.000
125	0.104	0.035	0.127	0.153
175	0.000	0.053	0.044	0.009

Source: [10].

showing the extent of malathion pesticide concentration which allows the quantification of the pesticide by the method of external standardization. Results were then extrapolated and determined the concentration at different distances from point source.

2.2.3. Pesticide residue in soil

Endosulfan concentration at different soil depth (0–175 cm) after days of spraying was calculated using Microsoft Excel regression. Equations for 3rd degree polynomial, projection of endosulfan concentration at different soil depth were used.

2.2.4. Pesticide degradation

Pesticide dissipation rate [24] was used to determine the microbial decay rates of the four priority chemicals up to year 2020 from the initial application.

$$C_t = C_0 e^{-kt} \quad (3)$$

where k is dissipation rate of the pesticide, t is time, C_t is amount of pesticide at time t after application and C_0 is the initial amount of pesticide after application.

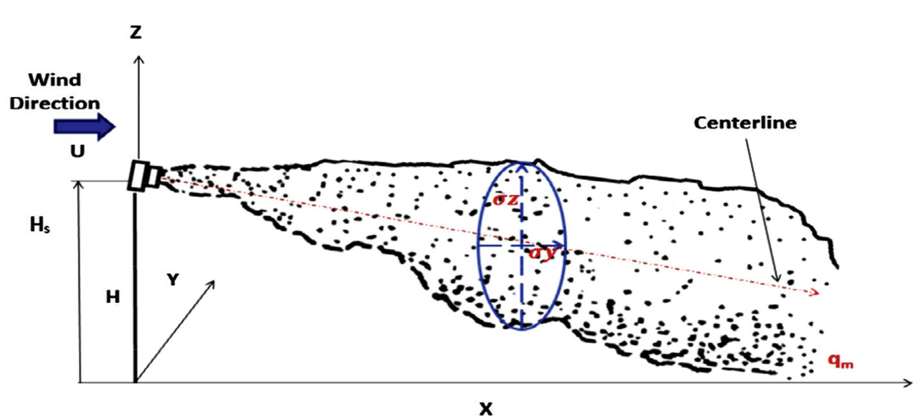

Fig. 2. Schematic diagram of pesticide drift during spraying using backpack sprayer.

2.3. Some factors for the transport of pesticides

It was considered that pesticide application was carried out by all rice farmers in Barangay Samil from 6 up to 9 am when the sun was not very bright. The model considered settling particles remained at the ground surface upon striking [23]. Other variables such as the rate of mass release from the nozzle was calculated. Pesticides application considered was 45 d after planting. Some additional considerations were the meteorological data; i.e., average wind speed and direction, constant pressure that produces a droplet size equivalent to 240 μm for a hollow cone type nozzle which is the most suitable for pesticide application [24], wind speed was 2.06 m s^{-1}. Other factors considered were: (1) advection was by the East Southeast (ESE) wind; (2) dispersion occurred in the crosswind and in the vertical direction, (3) the mean trajectory of the drops, and (4) the slope being conditioned by the gravity settling velocity.

3. Results and discussion

Results of the modeling of the residue transport are discussed in the subsequent sub-items.

3.1. Fate and transport of pesticides in air

The highest concentration of L-cyhalothrin in air and drops deposition rate occurs at a 2 m distance from the initial application as shown in Fig. 3. The results showed that the relative drift deposition in soil and concentration in air of L-cyhalothrin decreased exponentially with the increase in distance from the point of application. These results are comparable with the results of the studies carried out by Garcia-Santos et al. [8] and Lebeau et al. [23].

Among the 7 selected groups of farmers (Fig. 3), Group 4 recorded the highest L-cyhalothrin concentration of 2.83 μg m^{-3} at 2 m from the point of application. The lowest was from Group 2 which recorded 0.76 μg m^{-3}. Both Group 2 and 4 recorded 99% reduction of L-cyhalothrin concentration at 20 m-distance. These results are attributed to the effect of meteorological factor such as wind speed of 2.06 m s^{-1} blowing from ESE. It was recorded that L-cyhalothrin in air could travel up to 20 m with concentration of 9.63E-05 μg m^{-3}, the lowest concentration recorded among the 7 groups of farmers.

3.2. Fate and transport of pesticide in water

Pesticide residue contamination in rivers, lakes, and Pagsanjan-Lumban catchment was perceived to come from crop and rice plantation at higher elevation of Lucban and Pagsanjan as shown in Fig. 4. This was associated with the precipitation and runoff events together with the 18% topography slope of the area. Concentrations of these chemicals in water bodies are influenced by its distance of application point, time of application, climatic condition, soil type, and topography. Similar result was also discussed in Simon and Agulto [25].

3.2.1. L-cyhalothrin

The initial distance of 50 up to 250 m [25] was considered in this study. The resultant mathematical models (Fig. 5) was used to predict the distance could L-cyhalothrin traveled from 2008 to 2020. Fig. 5b illustrates that the concentration of L-cyhalothrin approached to zero at 200 m. This is attributed to the high K_{oc} (partition coefficient) value (180,000) of L-cyhalothrin [26] that makes its strong affinity to soil and sediments. Hence, concentration prediction showed a shorter distance compared to malathion (Fig. 6) which has lower K_{oc} value.

3.2.2. Malathion

Fig. 6 illustrates that Malathion concentration used in the area approached to zero at a distance of about 560 m from the application area. This is attributed to malathion's low soil partition coefficient (K_{oc}) value of 1800 [26] which increases the possibility of its movement from soil to water bodies. Malathion easily dissolves in water [12] and rainfall aided its movement to water bodies. This condition predominates in soluble phase from the site of application to surface water [20]. Hence, resulted distance traveled by malathion is longer than L-cyhalothrin.

3.2.3. Cypermethrin

The elevation (Fig. 4) of Lucban, Quezon and Pagsanjan, Laguna is 454 and 12 m, respectively. The average rainfall in Lucban and Pagsanjan is 101 inches (256.54 cm) with wind speed of 27 °C and 2.06 m s^{-1}, respectively. The flow of cypermethrin from higher to lower elevation was attributed to the 16% exposure of the area to typhoons. This information is a good direction for future study on understanding the behavior of cypermethrin in soil and sediment particles during runoff event. Runoff through Laguna Lake is possible based on the Laguna Lake watershed and the identified elevation of Lucban and Pagsanjan.

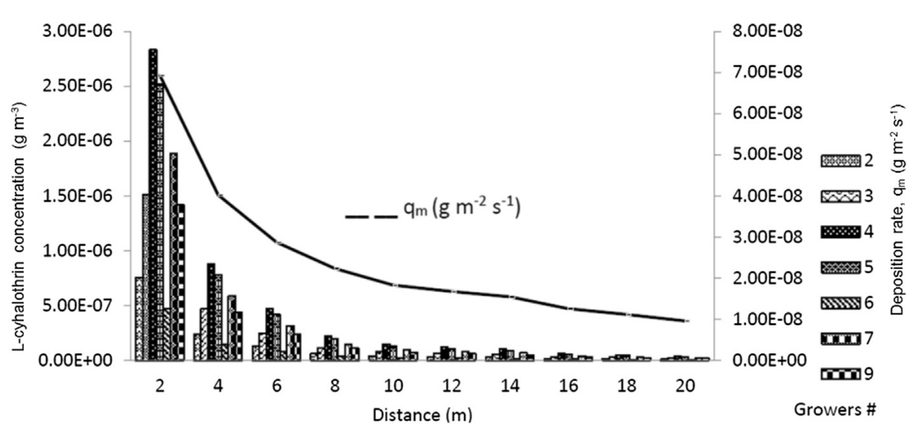

Fig. 3. Graph of L-cyhalothrin concentration with distance using Gaussian dispersion model.

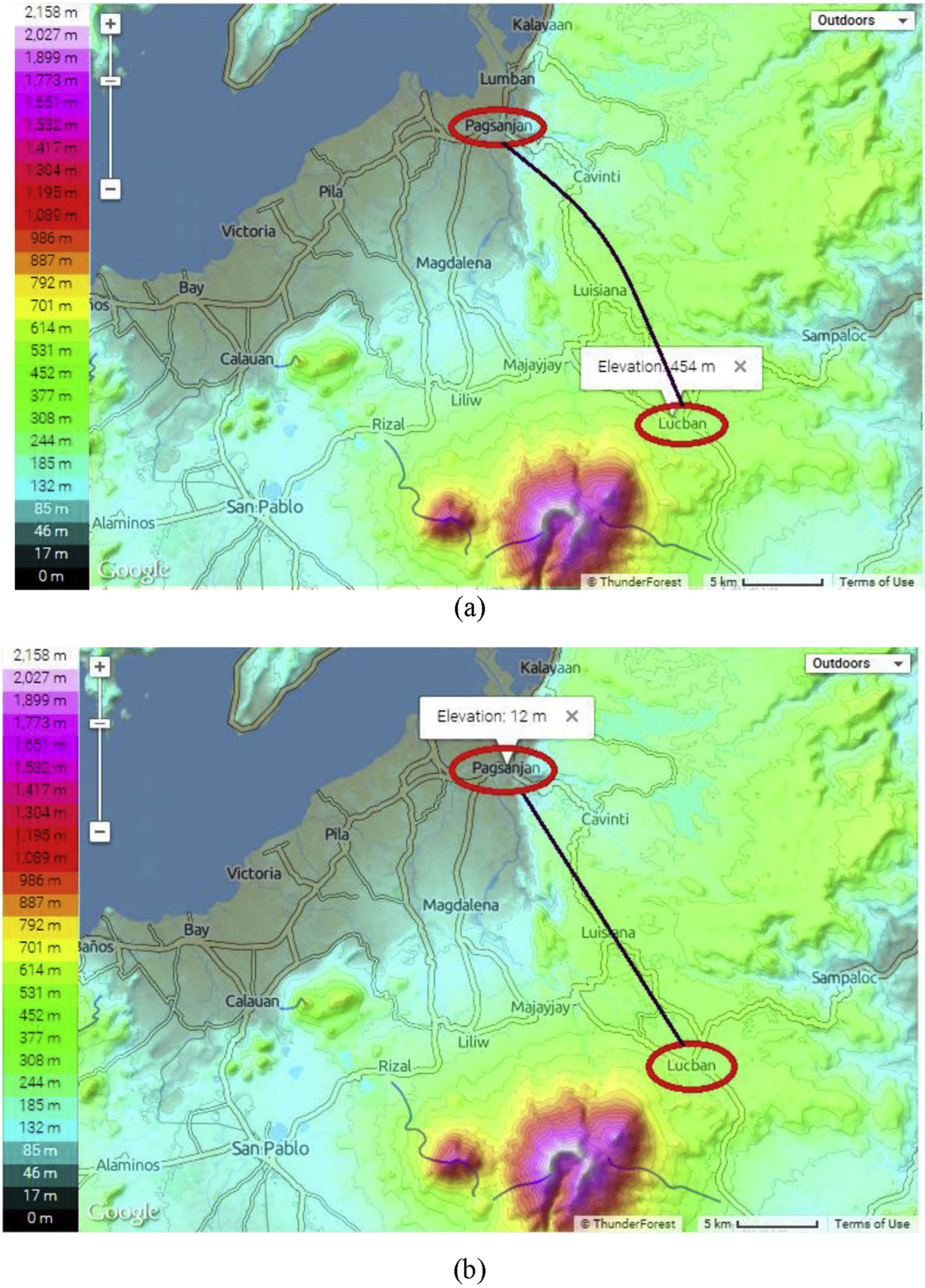

Fig. 4. Location and elevation of (a) Lucban and (b) Pagsanjan.

3.3. Fate and transport of pesticide in soil

Major pathway of pesticide dissipation in soil is caused by microbial activities like catabolism and metabolism [21]. Some chemicals are easily broken down while others are very persistent and took longer time to degrade. One of these persistent organic pollutant is endosulfan which occurs as α- and β-isomers in temperate/tropical soil with endosulfan sulfate as its major metabolite [27,28].

Soil properties and other environmental conditions play a major role in the movement and degradation of these two isomers.

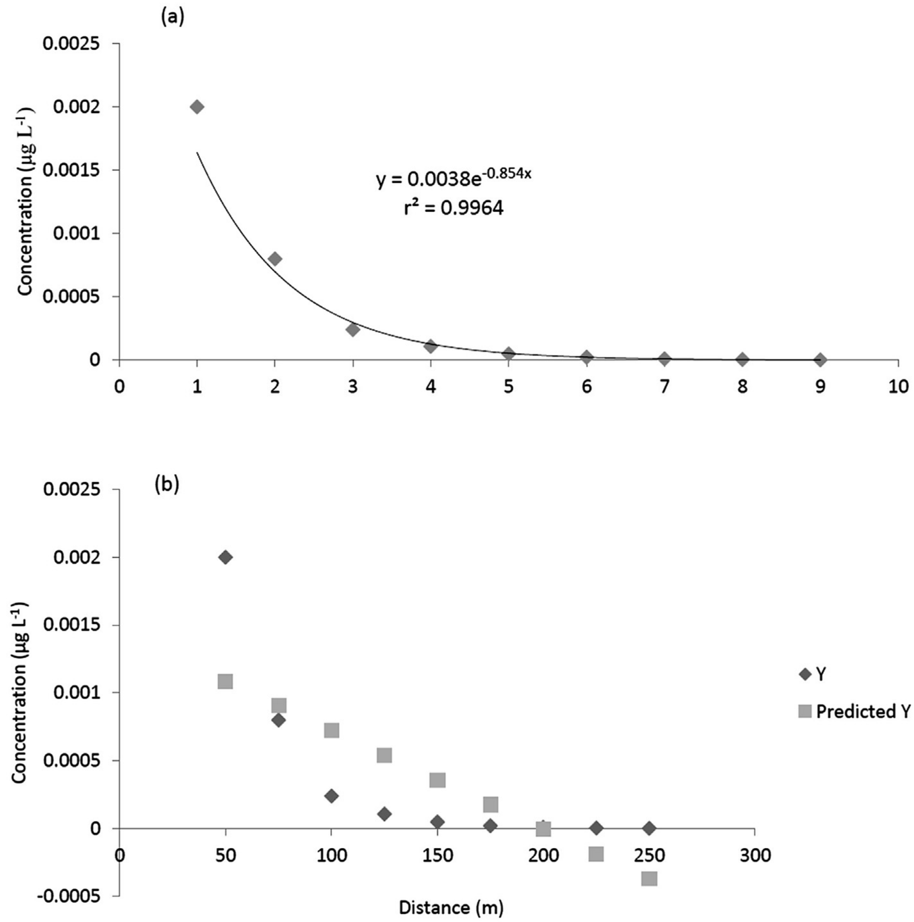

Fig. 5. L-cyhalothrin concentration (μg L^{-1}) (a) with extracted equation and predicted 2020 concentration at different distance (b) using the extracted equation.

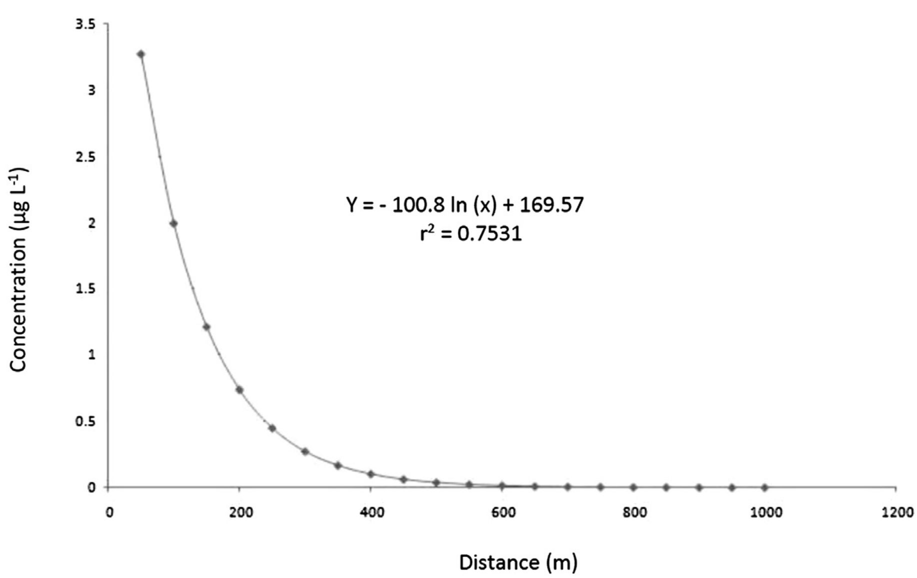

Fig. 6. Concentration of malathion (μg L^{-1}) with distance in Pagsanjan-Lumban catchment.

Ultisols such as those found in Laguna [29], are type of soils that are usually moist or moist for 90 consecutive days with subsurface horizon of clay accumulation and is suitable for planting [30]. Soil type at Pagsanjan-Lumban catchment particularly at the outlet of the watershed specifically at the northwest section of Lumban and Pagsanjan is “Marikina soils [31]”. It is a light brown soil and good internal–external drainage which considered a highly productive soil for vegetable crops. Downward movement of the residues followed the chemical behavior of the movement of solutes in the soil. The residues at a particular soil depth increased, reached a peak value and then declined over time. This pattern can be expected with chemicals which can dissolve in the soil water, which moves by mass flow and diffusion, but which are degraded, delayed or absorbed in the soil [21].

The endosulfan concentration profile in soil (Fig. 7) follows a polynomial pattern of 3rd degree and recurring shown in Fig. 8. At a depth range of 125 cm from ground surface, endosulfan concentration showed a decreasing trend and recurred during the 25th day on average from the date of application (i.e., spraying). This behavior can be attributed to low water solubility which caused its persistence in soil and water bodies [22,27] although runoff took place. The soil types in the area are cambisols, luvisols, fluvisols, gleysols, and vertisols [32]. The study recorded that at a depth of 175 cm from ground surface, the concentration shows an increasing trend at initial days of application and started to decrease after 40 d of application. In this study, the rate of recurrence of endosulfan, shown as Fig. 8a–e specifically within 125 and 175 cm soil deep from ground surface was about 5 and 1 cm d^{-1}, respectively. This illustrates that shorter period of recurrence happened in shallow depth (0–175 cm) of soil. This also shows that recurrence of endosulfan in soil decreased with depth. A significant amount of endosulfan moved through the soil during the first week (Fig. 8a). Small amounts of endosulfan would be detected up to 175 cm below ground. This indicates that endosulfan has the potential to percolate and contaminate the soil with probability to contaminate shallow aquifer if the soil is saturated [21,22] and possess property for drainage. The above results point to the direction that endosulfan concentration on soil needs to be examined further beyond 175 cm below ground due to the concentration of endosulfan at this level (0.06 g). This is significant as roots of some plants reach to about 1 m depth which possibly takes-up endosulfan concentration of concern. The above concentration becomes a concern as the EPA chronic dietary exposure of endosulfan for children and adults is 0.0006 and 0.006 mg kg^{-1} d^{-1}, respectively. Fig. 8a–e also show that endosulfan concentration is of concern up to 175 cm below soil surface and until 55 d.

3.4. Pesticides degradation

Pesticide degradation increases with temperature and sunlight intensity [33]. Calculation of chemical degradation of L-cyhalothrin in the environment based on its half-life of 9 d [34] showed that with the initial concentration of 4.71E04 g m^{-3} in 2007, reduced to 2.06E-101 g m^{-3} in 2015; that is 8.63 years later. L-cyhalothrin is

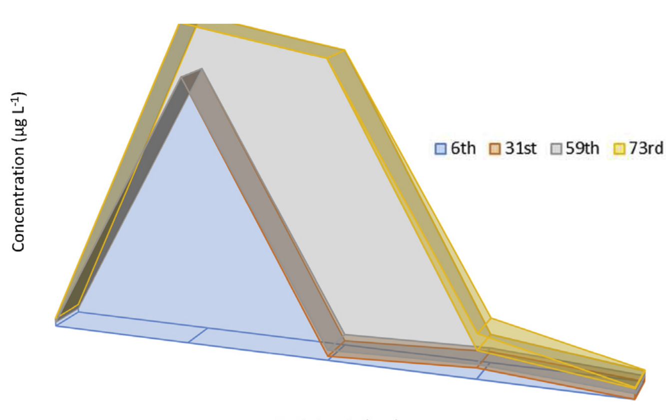

Fig. 7. Endosulfan concentration (μg L^{-1}) profile in soil depth (cm).

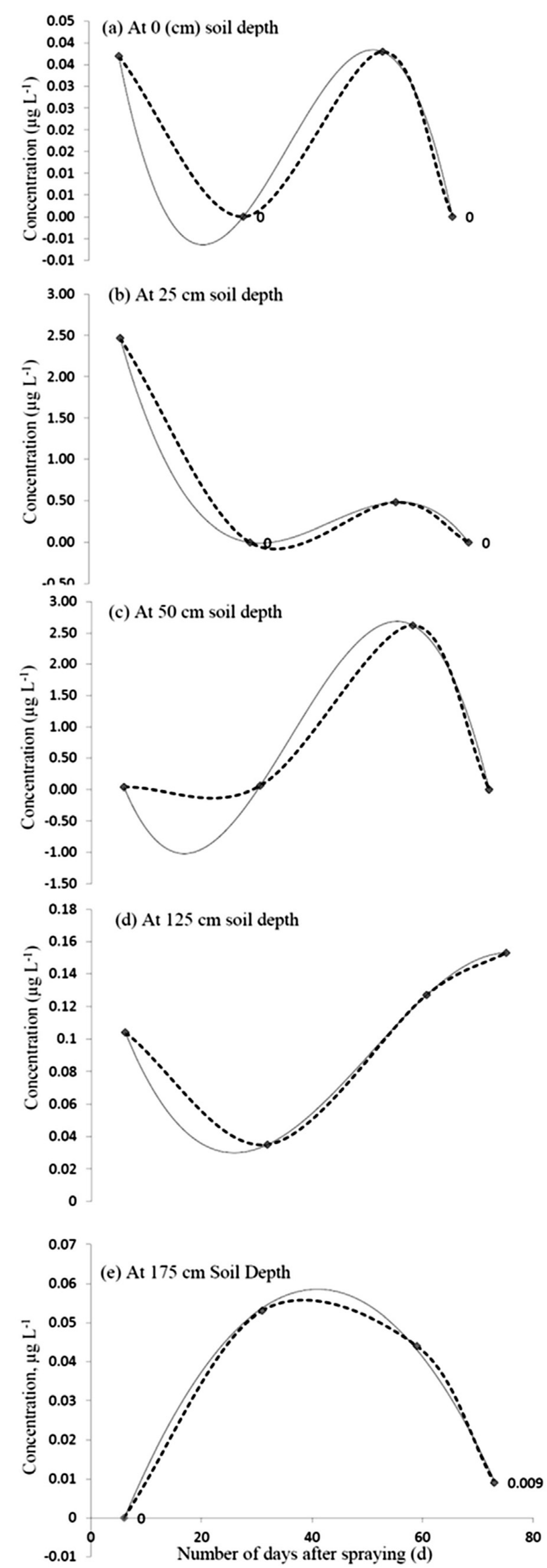

Fig. 8. Concentration of endosulfan after days of spraying at different soil depth (a–e) (Dot and solid lines are experimental and simulated data, respectively).

moderately persistent in soil with a half-life of 30 d [35] and with regular application, e.g., several applications in a year, possible accumulation is anticipated. This becomes a concern to some animals, and living organisms; hence this concern shall be given attention.

Pesticide degradation in water is affected by pH and temperature [12]. Malathion in this study does not accumulate through time due to its half-life of 1.65 at pH 8.16 in water and 0.8–1.4 d in sediments under aerobic conditions [12]. Its concentration degrades rapidly thus, may not be hazardous to the environment and public health.

Endosulfan isomers degradation does occur in temperate/tropical soil and aquatic systems [35]. Whereas, Cypermethrin degrades rapidly in soil under aerobic conditions with a half-life of 4 d (incubated soil)-4 wk in sandy soil depending on its concentration [36–38].

4. Conclusions

Modeling of the four pesticides (L-cyhalothrin, cypermethrin, endosulfan, and malathion) using Gaussian dispersion equation; ANOVA and linear regression resulted to information and mathematical models. Results of modeling showed exponential decrease in concentration with respect to time and distance as it travels in air, soil, and water. The residual amount of L-cyhalothrin in air traveled up to 20 m with concentration of 5E5 $\mu g\ m^{-3}$ and the highest concentration of drops deposition occurred at 2.0 m horizontal distance from point of application. Lowest concentration was observed at 1200 m from the point of application. It was recorded the endosulfan recurrence in the Lucban area within 125 and 175 cm deep from ground surface. The rate of recurrence was about 5 and 1 cm d^{-1}, respectively. Further, the model $y = -100.8 \ln(x) + 169.57$ predicts the transport of malathion in surface water. Another, cypernethrin contaminated the water catchment area due to its high affinity to soil/sediments which were carried by runoff from 454 m elevation. It has been noted that information and models produced by secondary data assessment, from credible sources, aided in understanding the transport and fate of L-cyhalothrin, cypermethrin, malathion, and endosulfan in three different environmental media. These models can be utilized to contribute in the decision making at similar environmental condition for the development of guidance document and values, policy development, remediation planning and strategies for project implementation. It also can be used for future research direction.

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