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Behaviour Study of Four Insecticides using different Mathematical Modeling

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Abstract: The adsorption, distribution coefficient, Langmuir and Freundlich models and the mathematical evaluation of pesticides have become more interesting from an environmental point of view. The outcomes revealed that indoxacarb, imidacloprid, and lambda-cyhalothrin are subjected to the Pseudo-first order reaction (PFO). The rate of degradation was reached, into 0.01, 0.07, and 0.04 a minute respectively. While chlorantraniliprole reached 0.00002 a minute. This indicates that these insecticides are decreasing in their concentrations depending only on the time. Hence, the time required to decrease 50% of each insecticide (DT_{50}) was various periods. The DT_{50} for indoxacarb, imidacloprid, and lambda-cyhalothrin, and chlorantraniliprole 3.2, 1.9, 10.1, and 2.3 days respectively. The distribution Coefficient Kd as well scored (5.25, 1.30, 0.562, and 0.639) mL g^{-1} respectively. This indicated that indoxacarb, imidacloprid has a mobility behaviour, while the lambda-cyhalothrin and chlorantraniliprole are less mobile in the soil. In terms of Freundlich model (aF), chlorantraniliprole 2.82 and lambda-cyhalothrin 2.75 are more fit than indoxacarb 0.013 L g^{-1} and imidacloprid 0.249 L g^{-1} subjected to the Langmuir model (KL).

Keywords: Adsorption, Batch study, Chlorantraniliprole, Imidacloprid, Indoxacarb, Lambda-cyhalothrin.

Introduction

The accumulation of persistent poisonous substances, chemicals, salts, radioactive materials, or disease-causing agents in the soils that have a negative impact on plant and animal health is referred to as soil pollution (Okrent, 1999). The Persistent Organic Pollutants (POPs) have long half-lives in the sediment, soil, air, and biota. These substances could be transferred into fresh and marine water even with the low concentrations (Ashraf, 2017). All living organisms, especially people, are frequently harmed and poisoned by the wide variety of soil contaminants (Ashraf *et al*., 2014). This pollution is mostly due to frequently used pesticides in agriculture or the public sector (Li *et al.,* 2008). The pesticides can enter the soil directly by the intentional application or indirectly through wet and dry deposition, wastewater, sewage sludge, air, or other means (Fenoll *et al.,* 2011). The main factor influencing a pesticide's ecotoxicological, environmental mobility, and the rate of degradation is soil adsorption. Since it controls the release rate and potential mobility of pesticides in the soil, the desorption process of pesticides is also a crucial process. Since the amount of adsorbed pesticide and its rate of desorption determine its adverse effects on the next crop, the adsorption/desorption behavior is particularly a big problem for pesticides (Liu *et al.,* 2010). Hence, in this field, Al-Farttoosy (2021) pointed out that the sorption of pesticides in the soil loses their biological effectiveness. Because of an interaction of the adsorbent particles with the absorption site on the surface of the adsorbent (Rasool *et al.,* 2022). Moreover, they mentioned two types of adsorption. Firstly chemisorptions in, which chemical bonds are formed between the adsorbent and adsorbate, and secondly, physical adsorption in, which interaction results from weak van der Waals forces (Rasool *et al.,* 2022).

Indoxacarb ((S)-methyl 7-chloro-2,5-dihydro-2-{[(methoxycarbonyl)[4-(trifluoromethoxy)phenyl]amino]carbonyl}in deno[1,2-e] (Sachdeva *et al*., 2013; Tse & Eslick, 2014) oxadiazine-4a(3*H*)-carboxylate is extremely effective against the lepidopteran insects (Patra *et al.,* 2022). Since the early 1990s, imidacloprid, a neonicotinoid insecticide, has been approved for numerous agricultural and residential purposes. Nicotinic acetylcholine receptors are the mechanism of action (nAChR) (Duke & Powles*,* 2008). Alpha-Cypermethrin, a pyrethroid insecticide with high activity, is efficient against a variety of pests found in farming and animal husbandry (Kocaman & Topaktas, 2009). An insecticide that belongs to the novel class of anthranilic diamides is chlorantraniliprole. It works by releasing intracellular Ca^{2+} stores through the ryanodine receptor, it affects Lepidoptera. This causes Ca^{2+} depletion, the halt of eating, lethargy, muscle paralysis, and ultimately insect death (Lahm *et al*., 2005).

In our study, we highlighted four common insecticides which used to control *Tuta absoluta*. Where those insecticides are implicated to tomato's field contamination by their residues. Because of little information on the environmental behaviour of these insecticides, the aim of this study is firstly to predict the potential pollution of the groundwater and secondly to understand the kinetic fate of these insecticides under Basra soil condition using the mathematical models.

Materials & Methods

The environmental behaviour of four different insecticides including; indoxacarb, imidacloprid, lambda-cyhalothrin, and chlorantraniliprole, has been studied (Table 1). The study's defined protocol has been adhered to (Al-Farttoosy, 2021). To assess the adsorption kinetics reaction, 50 mL of each trade insecticide formulations indoxacarb, imidacloprid, lambda-cyhalothrin, and chlorantraniliprole were added to three 250 mL flasks each holding 100 g of soil. Three more flasks were treated with distilled water as a control. All flasks were shaken at 150 rpm for 24 hours in an incubator to bring them to balance. One day after the equilibrium was reached, 1 mL of supernatant was added to 1 mL of eppendorf. Running the centrifuges at 3500 rpm for 30 minutes. The 022 filter was then applied to the aliquot. The quantities of indoxacarb, imidacloprid, lambda-cyhalothrin, and chlorantraniliprole were then calculated from the filtered aliquot using spectrophotometry at 310 nm, 270 nm, 266 nm, and 260 nm respectively.

Statistical analysis and calculations

To evaluate the data using different models, the linear model and kinetics models were utilized. All data were calculated as a mean standard deviation (SD) (the pseudo-first, the pseudo-second- order kinetic models, Langmuir, Freundlich models, and Thomas model). The study was carried out using Graph Pad Prism 8.0.1 (244), a 2D graphing and statistics program, developed by Inc. in San Diego, California 92108.

The pseudo-first (PFO) and pseudo-second order (PSO) kinetic were performed using the following equations of Revellame *et al.* (2020).

The PFO was determined using:

$$In\ (qe - q_t) = -K_1 + In\ qe,$$

Where: - k1 = is the equilibrium rate constant of the PFO sorption (ppm), t = the time (min), qe = is the initial concentration of the studied insecticide concentrations (ppm) and qt = the amount adsorbed of the studied insecticide concentrations ppm) at time t. Thus, the K1 was calculated from the plot of (qe-qt) versus t.

While the PSO was carried out by following equation:

$$\frac{t}{qe} = \left(\frac{1}{q_e}\right)t + \left(\frac{1}{K_2\ q_e^2}\right)$$

Where: qe, qt, t = above described. K2= equilibrium rate constant of the PSO sorption (min^1). According to this equation, the PSO rate reaction of the studied insecticide concentrations (K_{2nd}) and equilibrium sorption (qe) will be generated via the plotting of $t\ qt$ versus a time (t).

To calculate Freundlich model, the following equation $qe = aFC_e^{bf}$ has been used.

Where qe: insecticide's concentrations adsorbed per soil, bF, aF are Freundlich isotherm constants, and Ce mol L^{-1} is the equilibrium Insecticides concentrations in the solution. The plotting of Ln (qe) versus Ln (Ce) will provide the linear form of the Freundlich constant model.

To estimate Langmuir model, the equation has been followed:

$$qe = \frac{q_{m\ +K_L C_e}}{1 + q_L C_e}$$

Where: qe: the studied insecticide concentrations adsorbed on the soil, KL: Langmuir isotherm constant, Ce is the studied insecticide concentrations at equilibrium, aL: Langmuir isotherm constant. The value of KL obtained by applying the linear form of the Langmuir through the plotting of the Ce/Cs versus Ce will generate a straight line (Ho *et al.,* 2002).

The desorption % was measured by applying the following equation:

$$\%Desorption = \frac{amount\ of\ adsorbed\ insecticide}{total\ amount\ of\ adsorbed\ insecticides} \times 100$$

In order to determine the Kd, the below equation was used (OECD, 2000).

$$Kd = \frac{q_e}{C_e}$$

Where the Kd is the partition coefficient of the studied concentrations sorption constant, qe = the studied insecticide concentrations in the soil (mg g^{-1}), and Ce is the studied insecticide concentrations (mg L^{-1}) during the equilibrium. While the qe is estimated by applying the below equation (Bezzina *et al.*, 2018).

$$qe = \frac{(Ci - Ce)Vaq}{m}$$

Where Ci = is the studied insecticide concentrations used (mg L^{-1}), Ce = previously described, Vaq is volume analysed (mL) and m= the mass of soil used (g).

Results & Discussion

The Pseudo-First and Second Order Kinetic Models

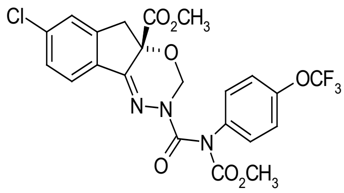
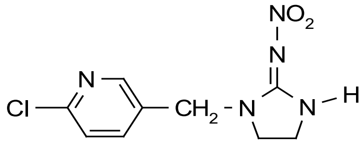
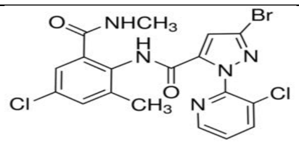
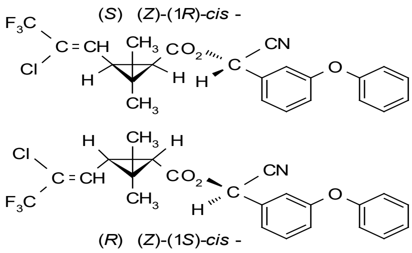
The information for pseudo-first-order and pseudo-second-order kinetics was presented in (Table 2). The slope of k1 and the intercept of ln qe were determined by the pseudo-first order linear plot of ln (qe - qt) vs t in (Fig. 1). It showed the graph for indoxacarb, imidacloprid, chlorantraniliprole, and lambda-cyhalothrin adsorption the values for k1, and R^2. The linear plot of t/qt versus t in fig. (2) gave the slope of 1/qe and intercept of 1/ (K2 qe2), and the correlation coefficient, R^2, which was acquired from the graph for the adsorption of these pesticides.

These results revealed the kinetic evaluation of the behaviour of four insecticides in the soil: indoxocarb, imidacloprid, chlorantraniliprole, and lambda-cyhalothrin. It appears from the results that two insecticides underwent the PFO are imidacloprid and lambda-cyhalothrin. In contrast, indoxocarb and chlorantraniliprole undergoes the PSO. In general, we can see all the insecticides breakdown rate in the soil was sluggish. The slow degradation rate was recorded in all insecticides that underwent the PSO compared to the PFO. Chlorantraniliprole and lambda-cyhalothrin were the lowest degradation, scoring 0.00002 and 0.0012 ppm minute^{-1} for both insecticide respectively than the indoxacarb and imidacloprid that recorded 0.002 and 0.001 ppm minute^{-1} respectively. However the values for K1 and K2 are different. The K2 values were quite small in comparison with the K1 in the PFO. In the PFO, indoxacarb had the highest rate of degradation, reaching 0.01 ppm minute^{-1}, followed by lambda-cyhalothrin at a rate of 0.04 ppm minute^{-1}. While the lowest breakdown were scored in chlorantraniliprole with the rate was 0.009 ppm minute^{-1} respectively.

As a result, the time required to break down half of the insecticide concentrations was varied. The imidacloprid was recorded as DT_{50}, followed by lambda-cyhalothrin for two days. As for the DT_{50} for chlorantraniliprole, it was 3.9 days, followed by indoxacarb 3.2 days after application. On the contrary, the DT_{50} of these insecticides under the PSO was the longer than the PFO. The longest time required to degrade chlorantraniliprole was 10 days compared to the indoxacarb, imidacloprid and lambda-cyhalothrin, which reached 7.8, 6.1 and 6.0 days respectively after the application.

In consequence, the importance of this procedure is to determine the behaviour kinetic adsorption of various insecticides involved: indoxacarb, imidacloprid, chlorantraniliprole, and lambda-cyhalothrin. Because understanding these features, enables us full knowledge on these studied insecticide demeanour when they applied and reached the soil. In the line with this study, numerous studies that applied to study the kinetic of various pesticide have revealed that the kinetic does not depend on the concentration only, for instance glyphosate residues were decreasing relied on the time (Al-Farttoosy & AlSadoon, 2022), as well as the fungicide carbendazim residues, which dropped into lower concentrations (Hameed & Al-Farttoosy, 2022). While the kinetic of diazinon behaviour in the soil did not depend only on time; but also another factor like humidity of the soil contributes in the kinetic reaction of the organic pollutants in the soil (Al-Farttoosy, 2021). In conclusion, these findings assisted us to understand that the soil contamination by these insecticides do not remain for a long time due to the decreasing of their residues during a specific time.

Table (1): Common information about the studied insecticides*

active ingredient name and chemical group	Trade name	Chemical structure	Company
Indoxacarb (Oxadiazine)	Avunt 15% EC		FMC
Imidacloprid (Neonicotinoid)	Modesta 35% SC		AgroScience LTD
Chlorantraniliprole (Anthranilic diamide)	Coragen 20% SC		FMC
Lambda-cyhalothrin (Pyrethroid)	Karate (with zeon technology)1 0%CS		Syngenta

* Tomlin C (2003) the pesticide manual, 13th ed. British Crop Protection Council, UK.

Table (2): The rate of four insecticides degradation and half-lives

Insecticide name	Degradation rate (ppm min^{-1}) Correlation coefficient R^2		Half-lives DT50 day^{-1}	
	PFO	PSO	PFO	PSO
Indoxacarb	0.01 0.743	0.002 0.734	3.2	7.8
Imidacloprid	0.07 0.83	0.001 0.81	1.9	6.1
Chlorantraniliprole	0.009 0.9173	0.00002 0.9179	3.9	10.1
Lambda-cyhalothrin	0.04 0.90	0.0012 0.89	2.3	6.0

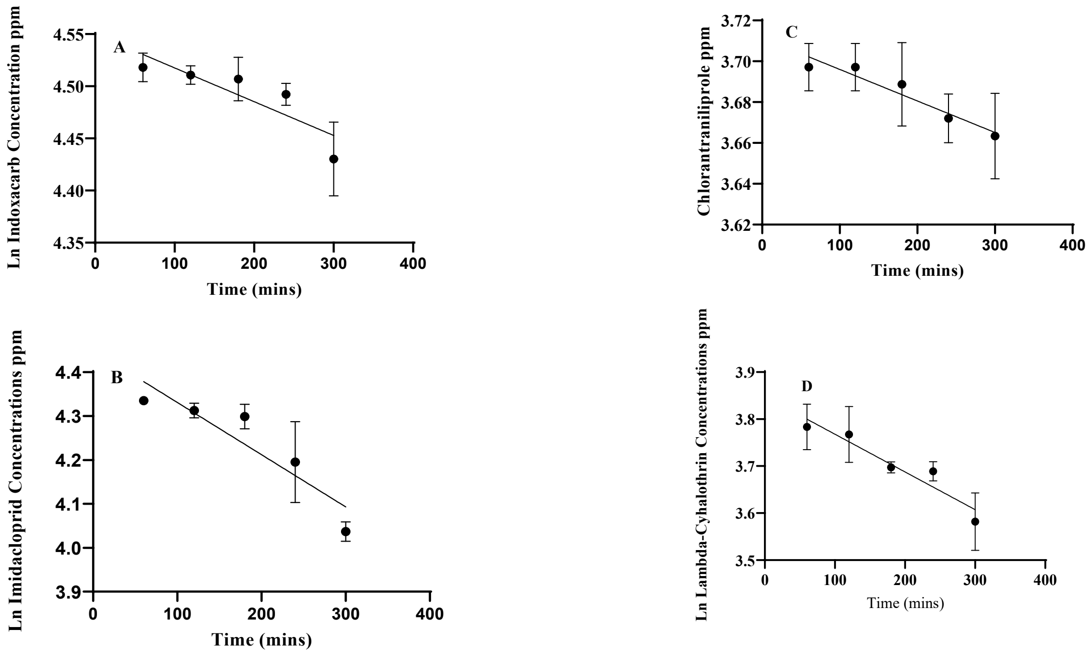


Fig. (1): The pseudo-First order kinetics based on the real data: A: Indoxocarb, B: Imidacloprid, C: Chlorantraniliprole, D: Lambda-cyhalothrin

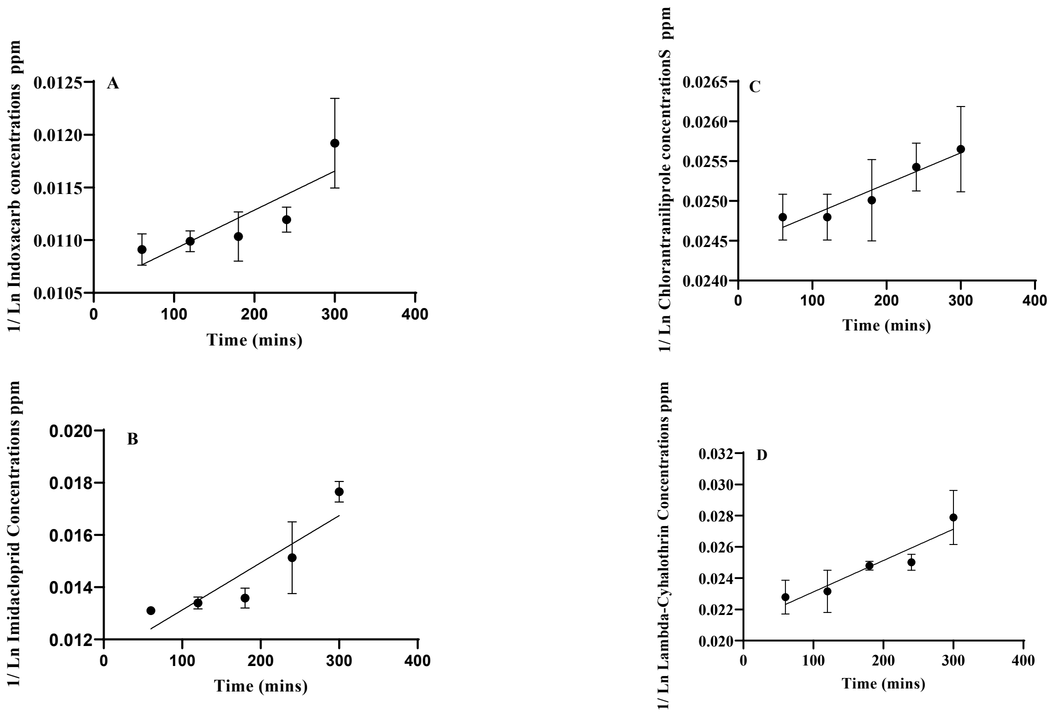


Fig. (2): The pseudo-Second order kinetics based on the real data: A: Indoxocarb, B: Imidacloprid, C: Chlorantraniliprole, D: Lambda-cyhalothrin

The distribution Coefficient

The adsorption coefficient (Kd) calculates the ratio of chemical material to water adsorbed onto soil. Freundlich solid-water distribution coefficients are another name for it (Kf) (ChemSafety, 2016). It illustrates the ability of chemical distribution between the soil particles and the water solution. Understanding the mobility of chemicals in the ecosystem and how they are distributed among soil, sediment, and water requires calculating the Kd coefficient (OECD, 2000). Accordingly, the outcomes in the table (3) confirmed that indoxacarb and imidacloprid have high capability for distribution between the soil solution and the soil particles. The Kd value has recorded (5.25 and 1.30) mL g^{-1} respectively. Whilst both Chlorantraniliprole and lambda-cyhalothrin have scored (0.639 and 0.562) $ml.g^{-1}$ which mean they implicated moderate mobility in the soil. The significance of the Kd resulted from not only due to its ability to describe the insecticides behaviour kinetic capacity in the soil, but also because those insecticides are non-ionic chemicals (Thorstensen *et al*., 2001). In this direction, Hu *et al*. (2019), sulfamethoxazole (SMX), which had a range of (1.13-2.41) mL g^{-1} in the soil, had lower the Kd values than sulfadiazine (SDZ), which varied from (1.54 to 3.41) mL g^{-1}. This is showing that compared to SMX, SDZ had a greater affinity for soil adsorption. Therefore Kd is a key parameter to determine the pollutants behaviour in the environment. It plays a critical role in predicting the possibility that dissolved contaminants would bind to soil (Weber *et al*., 2004).

Freundlich and Langmuir Models

The R^2, indoxacarb, and imidacloprid behaviour in soil were compared, and it was found that the Langmuir model described our results better than the Freundlich model (Tables 4 and 5). As long as the chemical absorbent and adsorbent system's equilibrium is described using the Langmuir adsorption model (Fig. 4). In light of this, the Langmuir model predicts that indoxacarb and imidacloprid have both been adsorbed onto a single molecule (Al-Farttoosy, 2020). Salman & Hameed (2010) revealed that the fitting data for the Langmuir, means that the adsorption has occurred on the monolayer of the soil. Hence, they demonstrated that the monolayer adsorption capacities of granular activated carbon, Filtersorb 300 (GAC F300) for 2, 4-D and carbofuran due to homogeneous distribution of active sites. Moreover, as shown in (Tables 4 and 5), both chlorantraniliprole and lambda-cyhalothrin experienced the Freundlich model (Fig. 3). This suggests that both insecticides are adsorbents since they create a monomolecular layer on the heterogeneous adsorbent surface. The chemical components in the soil are addressed in two different scenarios. According to a first scenario (Shariff, 2011), the chemical molecule is initially quickly removed from the solution before being adsorbed on soil sites. A chemical first passes through the sorption sites of clay colloids, organic matter, and soil organic matter colloid complexes, according to Chaudhary & Prasad (1999).

The possibility of the chemical substance being rapidly absorbed by the soil as a result of the abundance of vacant sites is the ultimate scenario. The amount of chemical (s) that travelled through the column decreases whenever these gaps are filled.

Table (3): The distribution Coefficient Kd

Insecticide name	Distribution Coefficient Kd mL g^{-1}
Indoxacarb	5.25
Imidacloprid	1.30
Chlorantraniliprole	0.639
Lambda-cyhalothrin	0.562

Table (4): Data of Four insecticides isotherm constant of Freundlich model for the adsorption by linear regression

Freundlich model			
secticides	Freundlich isotherm	Freundlich constant	R^2
Indoxacarb	13.3	0.136	0.9928
Imidacloprid	4.56	0.492	0.9874
Chlorantraniliprole	2.82	1.51	1
Lambda-cyhalothrin	2.75	1.49	0.9972

* R^2 is the coefficient of determination.

Table (5): Data of Four insecticides isotherm constant of Langmuir model for the adsorption by linear regression

Langmuir model			
Insecticides	Langmuir KL L. g^{-1}	Langmuir aL L. $mmol^{-1}$	R^2
Indoxacarb	0.013	76.63	0.9997
Imidacloprid	0.249	43.2	0.9946
Chlorantraniliprole	2.3	15.75	0.9999
Lambda-cyhalothrin	2.26	15.9	0.9960

* R^2 is the coefficient of determination.

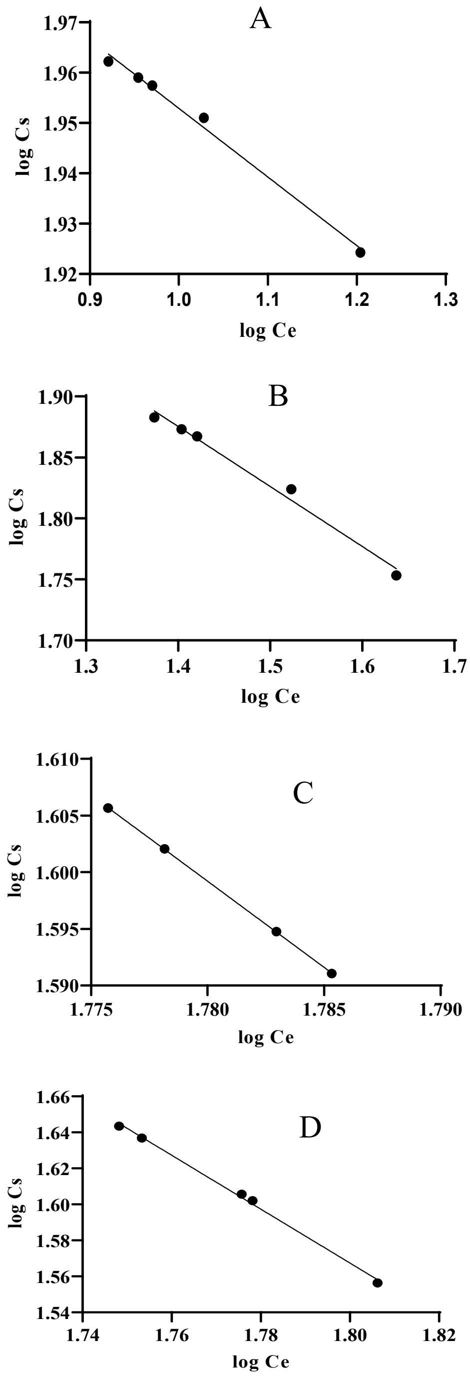


Fig. (3): The Freundlich model: A: Indoxocarb, B: Imidacloprid, C: Chlorantraniliprole, D: Lambda-cyhalothrin

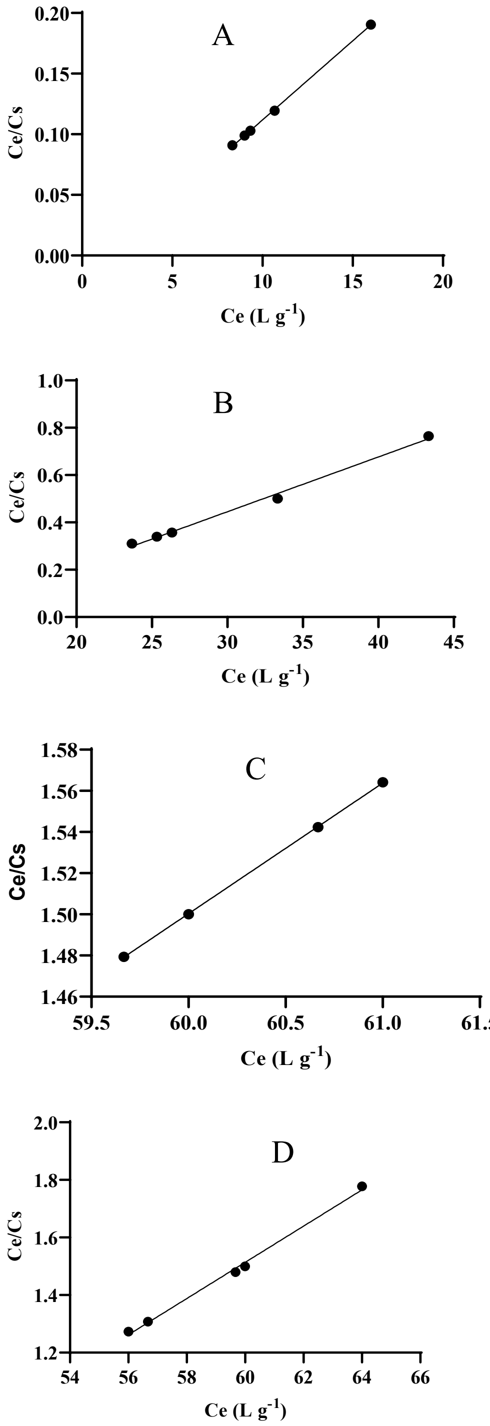


Fig. (4): The Langmuir model: A: Indoxocarb, B: Imidacloprid, C: Chlorantraniliprole, D: Lambda-cyhalothrin

Desorption process percentage

Fig. (5) showed the insecticide desorption results of the soil. It reveals that both indoxacarb and imidacloprid suffered in desorption for less than an hour. This result can be confirmed by table (3) which demonstrated that those insecticides have the ability to move with a high score of Kd. In addition, both indoxacarb and imidacloprid involved groundwater pollution. Furthermore the application of those insecticides caused a decreasing their biological activity to the targeted pests after a one hour. However, we can see that both chlorantraniliprole and lambda-cyhalothrin started to release their residues to the soil after two hours and a half. This confirmed that the biological effect will be dropped after the application and being active after two hours. Meantime those insecticides are not engaged to cause soil contamination due to their ability to adsorb on the soil particles.

Based on the findings of pesticide desorption to the soil, which were shown in fig. (5).

It demonstrates that indoxacarb and imidacloprid both experienced desorption in less than an hour. This suggests that the use of these pesticides, perhaps leads to a decrease in their biological effectiveness due to their movement or absorption of soil particles. Meanwhile, the use of such insecticides may have reduced their biological activity against the pests that were being targeted.

On the other hand, we can see that after two and a half hours, both lambda-cyhalothrin and chlorantraniliprole began to discharge their residues into the soil. This provided confirmation that the biological effects will diminish after application and resume two hours later. Due to their capacity to bind to soil particles, such insecticides are not currently a reason for the soil contamination. The varying data of each insecticide desorption show that the desorption depends on two factors. The first is the time where the desorption increased over time. Hence, we can see the desorption% ranged from 7.63-17.85% for indoxacarb, 9.17-26.47% for the imidacloprid, indicating that the desorption of these insecticides is difficult. On the contrary, chlorantraniliprole and lambda-cyhalothrin were ranged from 148.7-156.4% and 136.3-169.4% respectively. In this field, Kaur *et al.* (2016) found that the desorption of Pretilachlor herbicide is a complex. Moreover, its concentrations decreased as the adsorption increases. The second factor is likely to be the chemical properties of each insecticide as long as the studied insecticides belong to various chemical groups.

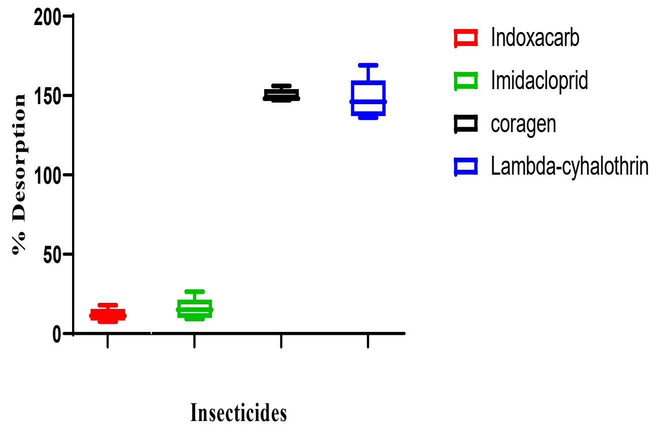


Fig. (5): Desorption of four insecticides in the soil

Conclusion

This study used a variety of mathematical modelling techniques to examine how four insecticides, indoxacarb, imidacloprid, lambda-cyhalothrin, and chlorantraniliprole behave in the soil. The results of this investigation show that indoxacarb, imidacloprid, and lambda-cyhalothrin are subjected to the PFO. While chlorantraniliprole undergoes the PSO.

The DT_{50} of each insecticides were determined. It is noted that DT_{50} depends on the degradation rate of each insecticide (K constant) The Kd values made a significant indicator to insecticide mobility. The values illustrated that indoxacarb and imidacloprid are likely to be mobile. Otherwise, chlorantraniliprole and lambda-cyhalothrin tend to be less moved during the first hour of application, then being gradually free in the soil. Two of the environmental behaviour important conclusions from this research are that chlorantraniliprole and lambda-cyhalothrin are more fit to the Freundlich model. In contrast, indoxacarb and imidacloprid underwent the Langmuir model.

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Contributions of authors

J.A.I.: The preparing, conducting of experiments, modelling computations, and gathering the investigated soil.

A.H.A.: Idea, checking the whole paper up in terms of scientific discussion, and its conclusions, and revise the manuscript.

A.A.M.: Read and collecting the references relevant to the paper.

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Conflicts of interest

There is no conflict of interest among the authors for publishing this manuscript.

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دراسة سلوكية لأربعة مبيدات باستخدام نمذجة رياضية مختلفة

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المستخلص: أصبح الامتزاز ومعامل التوزيع ونماذج لانكموير وفريندلش والتقييم الرياضي للمبيدات أكثر اثارة للاهتمام من الناحية البيئية. تم تقييم أربعة مبيداتٍ حشريةٍ اندوكسارب و ايميداكلوبرايد و لامبدا–سيهالوثرين و كلورانترانيليبرول رياضياً. كشفت النتائج بان اندوكسارب و ايميداكلوبرايد و لامبدا–سيهالوثرين خضعَ لتفاعل الدرجة الاولى (PFO) بلغَ معدل التحللُ 0.01 و 0.07 و 0.04 دقيقة$^{-1}$ على التوالي. بينما مبيد كلورانترانيليبرول بلغَ 0.00002 دقيقة$^{-1}$. هذا يشيرُ الى ان تناقصَ تراكيز المبيدات يعتمدُ على الوقتِ فقط، وبالتالي فأن الوقتَ اللازمُ لتناقصَ 50% من كل مبيدٍ حشري (DT50) مختلفة. بلغت فترة نصف العمر للاندوكساكارب و ايميدا كلوبرايد و لامبدا–سيهالوثرين كلورانتانيليبرول 3.2 و 1.9 و 10.1 و 2.3 يوم$^{-1}$ على التوالي. سجلَ معاملُ التوزيع Kd ايضا (5.25 و 1.30 و 0.562 و 0.639) مل غم$^{-1}$ على التوالي. تشير هذه النتائجُ الى ان المبيدين اندوكساكارب واميداكلوبرايد لهما سلوكاً حركياً، في حين ان المبيد لامبدا–سيهالوثرين و الكلورانترانيليبرول كانا اقلَ حركةٍ في التربة. كما اظهرت النتائجُ ان مبيد الكلورانترانيليبرول اكثر ملائمة لنموذج فرندلش (aF) 2.82 وكذلك مبيد اللامبدا–سيهالوثرين (aF)2.75 مقارنةً بمبيد الاندوكساكارب و اميداكلوبرايد اللذان كانا ملائمين لنموذج لانكموير (KL) وبلغا 0.013 و 0.249 لتر.غم$^{-1}$ على التوالي.

كلمات المفتاحية: اندوكساكارب، اميداكلوبرايد، لامبدا-سيهالوثرين، كلورانتانيليبرول، ادمصاص، نموذج لانكموير، نموذج فرندليش.