

Persistence and Mobility of Imidacloprid and Abamectin Residues in Green House Soil

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Abstract: Two pesticides were selected, imidacloprid (Konfidor®) and abamectin (Vertimec®) which are widely used in controlling insects and pests in greenhouses in Palestine. Imidacloprid and Abamectin adsorption onto greenhouse soil surfaces was studied by batch experiments. Experiments were conducted in a set of 100 mL capped conical flasks. In each pesticide adsorption experiment, an aliquot (50 mL) of (10-50 mg/L) solutions was used. Pesticides concentration was analyzed at the end of each experiment. The results indicate that: (1) the degradation of Imidacloprid and Abamectin soils was fitted to the second-order reaction kinetics model and showed good performance for all treatments, (2) the GUS values obtained for Imidacloprid and Abamectin were ranging between 1.95 and 3.3 and 1.68 to 3.31, respectively which is rated moderate to slightly high leachable/transportable to groundwater [understanding], (3) the distribution coefficient for both tested pesticides exhibit increasing adsorption on soil surface with increasing concentration in solution, (4) the observed persistence, half-life for Imidacloprid and Abamectin was 61 and 41 days, respectively, and in good agreement with reported in literature values, (5) mobility rate constants (Kd of 2 to 11 and Koc of 142 to 817) obtained for both Imidacloprid and Abamectin were higher than those reported in literature revealing that the tested soil is higher in leaching capacity, (6) the risk of particle-bound pesticide transport through soil to groundwater was rated slightly high to high for both pesticides, and (7) mobility and persistence results of Imidacloprid and Abamectin on soil obtained in this study were highly influenced by soil composition of high silt and low organic matter content leading to lower sorption rates and higher leaching to groundwater.

Keywords: Pesticides, persistence, mobility, environmental hazards, imidacloprid, abamectin, soil, green houses.

1. INTRODUCTION AND BACKGROUND

Pesticides, a chemical or biological agent, are substances or mixture of substances intended for preventing, destroying, repelling or mitigating any pest [1]. Pesticides are made for crop protection from damaging influences such as weeds, plants pathogens, insects, worms, birds, and others.

All pesticides contain active ingredients and inert ingredients. The active ingredients are the substances that perform the desired effect of the pesticide. Inert ingredients are mixed with the active ingredients added for several different purposes including increasing the effectiveness of the active ingredients, making the pesticide easier to use or apply, or allowing several active ingredients to combine into a solution. The inert ingredients can make up as much as 99% of the final product. Just because these inert ingredients don't specifically target the pest doesn't mean that they aren't equally toxic as the active ingredients.

When pesticides are applied in greenhouses they diffuse into and/or attach to air, soil, water, plants, and humans and eventually become mobile. It is important

to know, among others, where pesticides will move in soil, where it will collect, how fast this will occur, and how long it will stay in the soil.

Several factors affect how a pesticide will move in the soil once it is introduced including: how much pesticide is applied, how and where pesticide is applied, when pesticide is applied, the nature of the pesticide, type and structure of soil, irrigation method, frequency, and intensity. There are two main routes by which pesticides enter the soil: spray drift to soil during foliage treatment plus wash-off from treated foliage [2] and release from granulates applied directly to the soil [3].

There are many different kinds of pesticides including biopesticides, insecticides, herbicides and disinfectants, antiseptics, sterilizers and sanitizers. other animals. Pesticides are categorized into four main substituent chemicals: herbicides, fungicides, insecticides and bactericides [4,5].

Pesticide residue refers to the pesticides that may remain on or in the environment after they are applied [6]. The mobility and fate of Pesticides residues in the environment including air, agricultural soil, agricultural products, and irrigation water has been recognized as one of the emerging issues in environmental pollution [7-16].

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Although there are benefits from the use of pesticides, some drawbacks might occur such as potential toxicity to the environment where pesticides are applied. Some pesticides can cause nerve or liver damage, birth defects and cancer. Many of these chemical residues, especially derivatives of chlorinated pesticides, exhibit bioaccumulation which could build up to harmful levels in the body as well as in the environment [17].

Health impact to pesticide exposures can produce two distinct types of adverse health effects – acute (short-term) effects and chronic (long-term) effects. The severity of adverse health effects caused by pesticides are determined by several factors related to the pesticide type and application or to human's genetic vulnerability, age, health conditions, length of exposure, and others [18].

The greatest concern regarding human exposure to pesticides is their presence in water [19]. In 1999, the U.S. Geological Survey found widespread contamination of U.S. water resources; in particular, more than 95% of samples collected from streams, and almost 50% of samples collected from wells, contained at least one pesticide [20].

In Mediterranean countries, most protected cultivation growers use soil – often associated with soil pests, salinity problems and excessive application of pesticides (nematocides, fungicides, insecticides and herbicides). Residues can be a danger to human health (for both consumers and producers) and often lead to environmental pollution [21].

The use of pesticides in the Middle East including Palestine is not only an issue of uncontrolled use, but it is also a problem pertaining to the handling, misuse and disposal of unwanted pesticides. This is exacerbated by undeveloped national laws and regulations in regards to potential fate and residuals impacts of pesticides on groundwater, food safety and public health. Extensive use of pesticides with residual contents exceeding the maximum residue limits on produce, urged many European countries to ban certain agricultural exports from several Arab countries, [22].

Lack of reliable data and knowledge gap on quantities utilized and impacts induced to humans and the surrounding environment alerted scientific community and public, [23].

According to a field survey conducted in 2012, imidacloprid (Konfidor) and abamectin (Vertimec) are

the two pesticides that mostly marketed and widely used in the Palestinian agriculture [24].

In Palestine most farms are small in size, family owned and operated, and grow vegetables, orchards, and dry land crops. Such family farms are labor intensive and limited in technical as well as in investment capacity. Although agriculture in Palestine remains the dominant economic sector, agricultural extension services are poor and Palestinian authority control of pesticides and their application is also poor. The average Percent of farmers Using pesticides in Palestine is 38.5% while this use in the Gaza Strip reach 71% [25]. Over 13% of the total pesticide use in the West Bank is in green houses [26].

Most agricultural wells in Palestine are shallow wells with depths between 80 to 120 meters. Such groundwater are highly susceptible to pollution from uncontrolled agricultural practices including excessive pesticide use. Due to the competition over land and other natural resources of Historic Palestine between Israelis and Palestinians and to fulfill increasing food demands, the use of greenhouses and intensive agriculture is increasing [27].

An understanding of the persistence and mobility of pesticides in Palestinian agricultural soil is essential for the government as well as the public to know and be aware of potential pollution risks to groundwater in order to take actions and measures and to care about the impacts induced.

The Kinetics of Abamectin and Imidacloprid adsorption on soil in greenhouse in Palestine was reported separately [28]. In this paper emphasis is given to understanding and assessing the mobility and persistence behavior of the residues of Imidacloprid and Abamectin in greenhouse soil in Palestine.

2. MATERIAL AND METHODS

2.1. Experimental Program

The research experimental work basically depends on determining of residues of Imidacloprid and Abamectin versus time in soil. Samples of soil and leachate were analyzed by UV spectrophotometer and high performance liquid chromatography (HPLC). The room temperature recorded ranged between 18 - 25 °C. All glassware used were cleaned and dried before measurement and each measurement of this study was the average of three readings to ensure that consist values were obtained. Standard readings were

Table 1: Wavelength and Retention Time for Imidacloprid and Abamectin Using HPLC

Pesticide Name	Wavelength (nm)	Retention Time (min)
Imidacloprid	270	6.5
Abamectin	210	4.49

obtained for imidacloprid and abamectin and were plotted against absorbance readings in order to calculate the concentrations of these compounds using calibration curves.

2.2. Instrumentation

The detection of wavelengths for pesticides residues compound were confirmed using high performance liquid chromatography (HPLC - SHIADZU CORPORATION), with Lichoro CART, C18 Column (150 x 4.6mm, 20 μ m) Detector FLUROCENCES ARRAY. Absorbance readings of imidacloprid and abamactine were detected using UV-VIS SHIMADZU, Model No: UV-1601 double beam spectrophotometer wavelength range from 190- 1100 nm, accuracy \pm 0.004. The wave lengths were 270 nm and 210 nm for imidacloprid and abamactin respectively (see Table 1 below).

2.3. HPLC Scanning of Imidacloprid

For detection of imidacloprid using UV at 270 nm, chemicals and reagents used were Acetonitrile solution, Triethylamine, distilled water and Imidacloprid. In this experiment two solutions were prepared: Mobile phase solution: solution prepared from 1 ml of triethylamine in 1600 ml of distilled water then add 400 ml acetonitrile, mix good and adjust the pH to 5.9 \pm 0.1. The standard solution: was prepared by dissolving an accurately weighed quantity of imidacloprid in diluents to obtain solution having a known concentration of about 0.0292mg/ ml. Procedure: inject equal volume (20 μ L) of standard solution into HPLC to take retention time, and then inject equal volume of two sample solutions into HPLC with cleaning by mobile phase after each sample.

2.4. HPLC Scanning of Abamectin

For detection of abamectin use UV at 210 nm, chemicals and reagents used were methanol, distilled water and Abamectin. Two solution of mobile phase and standard solutions were prepared. The mobile phase solution was prepared from methanol and distilled water(85:15 v/v) and the standard solution was prepared from 18 mg of abamactin dissolved in 100 ml

methanol. Procedure: inject equal volume (20 μ L) of standard solution into HPLC to take retention time and then inject equal volume of the two samples solutions into same device, with cleaning by mobile phase after each sample. The retention time and wavelength for each pesticides is given in Table 1.

2.5. Calibration Curves for Imidacloprid and Abamectin

2.5.1. Calibration Curve for Imidacloprid

A standard calibration curves for imidacloprid and abamectin were performed by preparing diluted solution to get the concentration of 1ppm, 10ppm, 20ppm, 30ppm and 50ppm, using a control concentration of zero ppm distilled water. A 2.857 ml of imidacloprid was placed in 1 liter volumetric flask and filled with distilled water to the mark, the concentration become 1000 ppm (stock solution). A 5 ml of this stock solution to 100 ml volumetric flask and filled to the mark using distilled water, the new concentration become 50 ppm. Then take 3 ml from the stock solution to 100 ml volumetric flask and filled with distilled water to the mark, the concentration became 30 ppm. After this 2 ml from the stock solution was taken and transferred to the 100 ml volumetric flask and filled with distilled water to the mark, the concentration became 20ppm. After this 1 ml from the stock solution was taken and transferred to the 100 ml volumetric flask and filled with distilled water to the mark, the concentration became 10ppm. After this 0.1 ml of the stock solution was taken and transferred to the 100 ml volumetric flask and filled with distilled water to the mark, the concentration became 1ppm. Absorbance readings were recorded at 266nm for imidacloprid using UV-1601 SHIMADZU Spectrophotometer.

2.5.2. Calibration Curve for Abamectin

The calibration curve for abamectin, 55.55 ml of abamectin was placed in 1 liter volumetric flask and filled with distilled water to the mark, the concentration became 1000ppm (stock solution). A 5 ml of this stock solution to 100 ml volumetric flask and filled to the mark using distilled water, the new concentration became 50ppm. Then 3 ml from the stock solution was taken and transferred to 100 ml volumetric flask and filled

with distilled water to the mark, the concentration became 30ppm. After this 2 ml from the stock solution was taken and transferred to the 100 ml volumetric flask and filled with distilled water to the mark, the concentration became 20ppm. After this 1 ml from the stock solution was taken and transferred to the 100 ml volumetric flask and filled with distilled water to the mark, the concentration became 10ppm. After this 0.1 ml from the stock solution was taken and transferred to the 100 ml volumetric flask and filled with distilled water to the mark, the concentration became 1ppm. Absorbance readings were recorded at 244 nm for abamectin using UV-1601 SHIMADZU Spectrophotometer.

2.6. Soil Sampling and Analysis

Five kilograms samples of red soil were collected from the greenhouse used for growing vegetables. One kilogram of soil sample was weighted accurately, sieved in 2.0 mm sieve, and dried at 105 °C. Samples of red soil were collected from the greenhouse and analyzed in the lab in order to evaluate the soil texture, moisture, pH value, and specific gravity. All soil analysis were carried in the laboratory according to standard methods for soil sampling and method of analysis [29]. All standard solutions used in the experiment were analytical reagent grade or of extra pure quality unless otherwise indicated. All glassware and plastic containers were soaked overnight in 10% nitric acid and rinsed with double distilled water before they were used for analysis.

3. RESULTS AND DISCUSSION

3.1. Soil Analysis Results

Soil analysis were conducted in the laboratory prior to the start of sorption experiments [30]. Table 2 lists soil analysis results obtained including soil specific gravity, pH, texture, organic content, and moisture. Knowing that sorption of pesticides to soil generally increases with soil organic matter content [31], it was noticed that the silt percentage of this soil is slightly larger than the clay and organic carbon and organic matter in soil are less than the percentage that increase pesticide adsorption which reduced the adsorption of pesticides (see Table 2). Accordingly and to this soil composition, it is expected that adsorption on soil will be less and leachability of pesticide to groundwater will be higher.

Table 2: Soil Specific Gravity, pH, Texture, Organic Content, and Moisture.

Soil properties	Result
Specific gravity	2.34
pH	7.18
Clay	43%
Silt	57%
Moisture content	13.12%
Organic carbon	1.38%
Organic matter	2.37%

3.2. Pesticide Retention in Soil

The retention of pesticides in soils is mainly due to the adsorption of pesticide on soil surface. However,

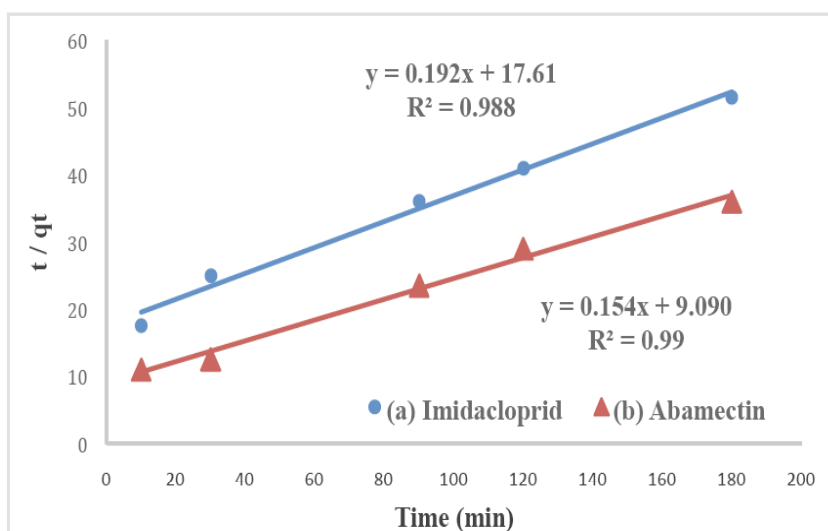


Figure 1: Kinetics of pesticides removal according to the pseudo-second-order model by soil at (initial conc: 15 mg/L, initial pH: 4, temperature: 25°C and solid/liquid ratio 1.0 g/50 mL).

Source: Jodeh *et al.* Khalaf.

the adsorption on soil is highly influenced by soil pH, type, texture, moisture, and organic matter content, and ambient temperature and sunlight hours. This adsorption could be reversible or irreversible depending on various environmental conditions [32-34].

The degradation of Imidacloprid and Abamectin soils was fitted to the second-order reaction kinetics model and showed good performance for all treatments (see Figure 1 and Table 3), with r^2 values ranging from 0.988 for Imidacloprid to 0.994 for Abamectin.

Table 3: Pseudo-Second-Order Kinetic Model Parameters for Imidacloprid and Abamectin Adsorption onto Soil at 25 °C.

Adsorbate	Parameters		
	K ₂ (g/mg min)	q _e (calc)	R ²
Abamectin	0.0026	6.45	0.99
Imidacloprid	0.0022	5.18	0.98

Source: Jodeh *et al.* Khalaf.

3.3. Pesticide Mobility

The mobility of Imidacloprid and Abamectin was determined based on its leaching potential. The GUS index (groundwater ubiquity score; [35]) is used to estimate the potential of a pesticide to contaminate ground-water by leaching.

The GUS is based on two active ingredient properties: the sorption in soil or adsorption coefficient (Koc) and pesticide persistence or soil half-life (TD50). These properties are used in the following equation:

$$\text{GUS} = \log(\text{TD50}) * (4 - \log(\text{Koc}))$$

The distribution coefficient will vary depending on the ratio of soil to water and the chemical properties of both soil and water. For this reason, a different number, sorption coefficient (Koc) is used to compare the relative sorption of pesticide. A low distribution coefficient indicates that more of the pesticide is in solution, a higher value indicates that the pesticide is more strongly sorbed to soil. The distribution coefficient, Kd was estimated based on the ratio of the amount of pesticide adsorbed on soil and left in solution at equilibrium:

$$\text{Kd} = \text{CAe} / \text{Ce}$$

Where,

CAe (mg/L) is the pesticides adsorbed on solid surface at equilibrium and

Ce (mg/L) is the pesticides equilibrium concentration in solution.

The adsorption coefficient, Koc is estimated using the organic content of soil and the distribution coefficient as follows:

$$\text{Koc} = (\text{Kd} * 100) / \text{C}_{\text{org}}$$

where,

C_{org} is the percentage of organic carbon content in soil.

Kd is the distribution coefficient

The results obtained from the sorption laboratory soil experiments along with estimates of Kd, Koc, TD50, and GUS are given in Tables 4 and 5.

GUS is used to rate the pesticides for their potential mobility towards groundwater and the higher the Koc values the more strongly the pesticide is adsorbed to the soil surface and consequently the less mobile to groundwater it is. The GUS values obtained for Imidacloprid and Abamectin were ranging between 1.95 and 3.3 and 1.68 to 3.31, respectively (see Tables 4 and 5) which is rated moderate to slightly high leachable/transportable to groundwater [36].

Comparison of the distribution coefficient, Kd for the Imidacloprid and Abamectin in tested soils reveals that both ranged from 1 to 11 and consequently both exhibit increasing adsorption on soil with increasing concentration of pesticide added.

The observed half-life, TD50, for Imidacloprid of 61 days in the soils were similar to those previously reported in literature varying from 48 to 190 days for imidacloprid [36]. However, the observed half-life for Abamectin of 41 days, was relatively higher than those reported (8 hours to 47 days [37]). This is because Abamectin is rapidly degraded in soils.

Mobility rate constants obtained (see Tables 4 and 5) for both Imidacloprid and Abamectin were highly variable indicating the influence of equilibrium conditions on the results.

3.4. Pesticide Persistence in Soil

Persistence of pesticide in soil is often expressed by pesticide half life in soil, TD 50, which is the length of time required for one-half of the original quantity to break down. This is the length of time required for one-half of the original quantity to break down pesticides

Table 4: Imidacloprid Concentrations Mobility and Persistence Constants

qe	ce	Kd	Koc	TD50	GUS
94	48.1	1.954	141.61	60.94	3.30
190	46.2	4.112	298.01	60.94	2.72
290	44.2	6.561	475.44	60.94	2.36
368	42.7	8.618	624.51	60.94	2.15
460	40.8	11.274	816.99	60.94	1.94

qe = Amount of adsorbate per unit mass of adsorbent at equilibrium.

Table 5: Abamectin Concentrations Mobility and Persistence Constants

qe	ce	Kd	Koc	TD50	GUS
44.5	49.1	0.906	65.67	41.41	3.53
96.5	48.1	2.006	145.37	41.41	2.97
234	45.32	5.163	374.15	41.41	2.31
281	44.4	6.328	458.61	41.41	2.16
441.5	41.2	10.716	776.52	41.41	1.79

qe = Amount of adsorbate per unit mass of adsorbent at equilibrium.

can be divided into three categories based on half-lives: non persistent pesticides with a typical soil half-life of less than 30 days, moderately persistent pesticides with a typical soil half-life of 30 to 100 days, or persistent pesticides with a typical soil half-life of more than 100 days [38].

The potential for or risk of particle-bound pesticide transport was rated high for pesticides with half-life of greater than or equal 40 days and/or for Koc greater than or equal 1000 [39]. The data of this research indicated that Imidacloprid with TD50 of 61 days and Koc up 817 has high potential for leaching and transport while Abamectin with TD50 of 41 days and Koc up 777 has moderate to slightly high leaching and transport potential. Accordingly and in similarity with published consequences, care should be given to their monitoring in the soil and groundwater for better control of their present and future transport, sorption, and leaching to soils [40-42].

The obtained values for Kd (2 to 11 - see Tables 4 and 5) and Koc (142 to 817 - see Table) for Imidacloprid were higher than those reported in literature (Kd 1-4 and Koc 132-310 [43]) revealing that the tested soil is higher in leaching capacity. This result is confirmed with GUS values obtained (see Tables 4 and 5). Similar results were found in Koc comparison for Abamectin between published of 4 [37] to those obtained from these experiments of 1-11 (see Tables 4 and 5). In addition, the Kocs for both

Imidacloprid and Abamectin are having similar variability as that of the Kd values for the same soils.

The purification rate for bioaccumulation of pesticides in groundwater [44-46], was estimated at 1-3 days based on the values of pesticide persistence obtained for Imidacloprid and Abamectin: TD50 of 61 and 41 days (see Tables 4 and 5). This high value indicates the need for close monitoring of the system including the application of both pesticides, soil, and groundwater.

It is worth mentioning that while the use of Imidacloprid and Abamectin pesticides in Palestine is traced to tens of years, the qualitative and quantitative monitoring and reporting of their impacts on soil and movement to groundwater was not done neither with time and/or with space. The results of this research is an important call to government as well as to all involved for immediate action in this direction.

Mobility and persistence results of Imidacloprid and Abamectin on soil obtained in this study were highly influenced by soil composition of high silt and low organic matter content leading to lower sorption rates and higher leaching to groundwater.

CONCLUSIONS

Based on the results obtained in this study the following concluding points were reached:

- Although Abamectin and Imidacloprid as insecticides and since tens of years are widely used in greenhouses in Palestine, the qualitative and quantitative monitoring and reporting of their impacts on soil and movement to groundwater was not done neither with time nor with space. Government as well as to all involved need to work immediately on their control.
- Observed half-life for Imidacloprid in the soils were in agreement with those previously reported in literature while those for Abamectin are slightly higher.
- Abamectin showed rapid degradation in soil.
- Abamectin and imidacloprid has slightly high to high potential for leaching and transport to groundwater and consequently care should be given to their monitoring in the soil and groundwater for better control of their present and future impacts.

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