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Influence of Fertilizer Sources on Pesticides Fate in Tomato Fruits and Soil with Special References to Efficiency of Some Herbicides

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Abstract : The objective of this investigation aimed to study the efficiency of some herbicides on weed infestation of tomato plants; chlorpyriphos – methyl and acetamiprid residues in tomato fruits were determined also. In addition, the role of fertilizer types on tested pesticides dissipation in soil was estimated. Results illustrated that all weeded control treatments decreased the number and dry weight of weeds comparing to the unweeded one. Pendimethalin and metribuzin were more efficient than other treatments on decreasing the number and dry weight of total weeds. Fluazifop- P-butyl was the best option to attain acceptable grassy weeds. Results showed also, the amount of residues varies for each insecticide to another, the initial deposits depending on the rate of use, so it's ranged from 30.9 to 38.0 ppm in tomato treated with chlorpyriphos – methyl while it's ranged from 9.8 to 11.7 ppm in the case of acetamiprid. The first five days were, however, the most critical period at which most of the residues (> 65.0 %) were dissipated in tomato fruits. The soil organic matter or soil rich in humus content are more chemically reactive with pesticides than nonhumified soil. Afterwords, tested pesticides was faster disappearance in organic and compost fertilized soils (total amount of detected residues were 0.81 and 1.07 ppm, respectively) than other treatments.
Keywords: Pesticides, Fertilizers, Degradation, Tomato.

Introduction

Tomato (*Lycopersicon esculentum,* Mill) is one of the most important vegetable crops grown in Egypt. It occupies the first place among vegetable crops with regard to cultivated area, production and value. Thus, a great attention should be paid to raise its productivity per unit area. This can be achieved by planting the high yielder cultivars and improving its agricultural practices. Pest control treatments are the limiting factors for tomato production (Shalaby *et al*[1]). Weed control play an active role in raising the yield of tomato since weeds cause great losses in yield. As hand labor became scarce and costly, herbicides replaced it as a cheap an easy method for weed control in tomato fields. In general, application of herbicides depends not only on its efficiency in controlling weeds but also on its effect on tomato plants. Hand hoeing treatment suppressed the growth of tomato weeds as compared with unweeded treatment (Ahmed *et al*[2]). Pendimethalin and Metribuzin have applied pre-emergence treatments. They provided excellent annual grasses and broad-leaved weeds control, respectively (Chnnappagoudar *et al*[3,] Shivalingappa et al[4,] Shil and Nath[5]). While, Fluazifop- P-butyl came in the first order for controlling total grasses weeds in faba bean (El-Metwally *et al*[6]). Fertilizers have a very profound effect on crop growth and yield especially nitrogen which is an essential element for plant growth and maintenance since it is considered a key nutrient in crop production (Abo Arab *et al* [7]). A variety of microorganisms (bacteria and fungi) have been used in soil inoculations intended to improve the supply of nutrients to crop plants, to stimulate plant growth, to control or inhibit the activity of plant pathogens and to improve soil structure. Other more recent objectives for the introduction of microorganisms into the soil are the mineralization of organic pollutants (bioremediation of polluted soils) (van Veen *et al* [8]). In the same respect, some microorganisms living in soil are known to be detoxification agents of pesticides, although pesticides may have a degree of persistence despite the same microorganisms. That may due to the difference in the physicochemical properties of soils and also the environmental factors such as pH, moisture content and temperature as well [(9)]. The rate of pesticide degradation in soil does not depend on organic contents of soil. Although, adsorption of pesticides increases with soil organic matter content and that possibly resulting in reduce in an availability of pesticides for degradation of soil water. This often offset by an increase in microbial biomass, which increases the rate of degradation (Nicholls [9] , Abdel-Rahman [10]). So, the objective of the present study aimed to investigate the following aspects:

1. Effect of fertilizer types on chlorpyriphos – methyl and acetamiprid residues in tomato fruits.
2. Potential of fertilizer types on remediation soil contaminated by pesticides.
3. The efficiency of some herbicides against narrow and broad-leaved weeds in tomato plants.

Materials and Methods

Field trails:

The field experiment was carried out in El-Mahmodia village, Dekernis district, Dakahlyia Governorate, Egypt. The experimental area was planted with tomato *Lycopersicon esculentum* (Mill.) variety "Super Strain B" after seeded in a greenhouse and then transferred to the field during summer cultivation season of 2015, under normal field and agricultural practices. The experiments were laid out in Randomized Complete Blocks Design with four replicates for each treatment. Each plot had five rows with 25 plants row^{-1}. The 15 plants of the medium row were used for data collection. The plant space was 0.5 x 1.0 m. The experiment area was divided into five main treatments, these are:

1. Without fertilizer.
2. Bio-fertilizer: A mixture of four microbial species in equal portions (*Bacillus megatherium*, *Azotobacter* sp., *Azospirillium* sp. and *Pseudomonas* sp.), it was applied to tomato seedlings dipping (250 g litter^{-1} water during 5 min.) just before transplanting (Chatterjee and Bandyopadhyay [12]).
3. Organic fertilizer: Cattle dung, the rate of use is 20 m^3 / fed.
4. Compost fertilizer: Compost El- Wadi, it produced by Delta Bio-Tec Co.; the rate of used was 5 ton / fed.
5. Chemical fertilizer (NPK): The rate of used was 400, 200 and 200 kg / fed. of mineral nitrogen, phosphorus, and potassium fertilizers, respectively.

The efficiency of some herbicides against narrow and broad – leaved weeds in tomato plants:

Pendimethalin is used a selective herbicide to control annual grasses and broad-leaved weeds in tomato as a pre-emergence or pre-planting soil incorporation treatment. While, metribuzin is used a selective herbicide to control annual grasses and broad-leaved weeds in tomato after 10 days from transplanting. Fluazifop- P-butyl is used a selective herbicide to control annual grasses weeds in tomato as a post- emergence. Also, common, trade and chemical names of each herbicide are shown in Table (1). Weeds were hand pulled from one square meter of each experimental unit at 60 days after sowing, then identified and classified into grasses and broad-leaved weeds. After air drying for 8 days and oven drying at 105° C for 24 hours, dry weight of both weed groups, as well as total dry weight, was recorded.

Table (1): Trade, Common and chemical names; rate and time of application of herbicides and insecticides used.

Trade Name	Common name	Chemical name	Rate of application	Time of application
Stomp 400 SC	Pendimethalin	*N*-(1-ethylpropyl)-3,4-dimethyl-2,6-dinitrobenzenamine	1.7 L/fed	Before cultivating irrigation
Sencor 70% WP	Metribuzin	4-amino-6-(1,1-dimethylethyl)-3-(methylthio)-1,2,4-triazin-5(4*H*)-one	0.3 kg/fed	After 10 days from transplanting
Fusalide Super E.C. 12.5 %	Fluazifop- P-butyl	butyl (*R*)-2-[4-[[5-(trifluoromethyl)-2-pyridinyl]oxy]phenoxy]propanoate	1L/fed	After 30 days from sowing
Reldan	Chlorpyriphos - methyl	*O,O*-dimethyl *O*-(3,5,6-trichloro-2-pyridinyl) phosphorothioate	1L/ fed	At the fruiting stage
Mospilan	Acetamiprid	(*E*)-*N*-[(6-chloro-3-pyridinyl)methyl]-N -cyano-N-methylethanimidamide	25 g / fed	

Effect of fertilizer types on chlorpyriphos–methyl and acetamiprid fate in tomato fruits:

Each main treatment was divided into two sub treatment; both sub-treatments were treated by chlorpyriphos-methyl and acetamiprid as the foliar spray on April 10, 2015, at the fruiting stage, once at the recommended rates (Table 1) using a knapsack sprayer provided with one nozzle delivering 200 liters water/ feddan. This was proved to be sufficient to give good coverage on the treated plants. Representative samples of tomato fruits were collected from the treated plots after 1 h (initial deposits) and 1, 3, 5, 7, 10, 15 and 21 days after spraying. Clean polyethylene bags were used for preserving the collected samples. The samples were stored at -20 ºC in the deep freezer until analysis.

Effect of fertilizer types on some herbicides and insecticides fate in soil:

Soil samples (500 g) were collected in different treatments at the end of the experiment; samples were put in chemically clean bags and stored at -20 ^{o}C in a deep freezer until analysis.

Determination of Pesticide residues:

1-Tomato fruits:

Extraction from plant samples by using QuEChERS method, this method is known as the quick, easy, cheap, effective, rugged and safe (QuEChERS) method for pesticide residues in vegetables and herbs. The procedure involved the extraction of a 15 g sample with 15 ml acetonitrile, followed by a liquid–liquid partitioning step performed by adding 6 g anhydrous MgSO4 plus 1.5 g NaCl. After centrifugation, the extract was decanted into a tube containing 300 mg primary secondary amine (PSA) sorbent plus 1.8 g anhydrous MgSO4, which constituted a cleanup procedure called dispersive solid-phase extraction (dispersive-SPE). After a second shaking and centrifugation step, the acetonitrile extract was transferred to autosampler vials for concurrent analysis by gas chromatography/mass [13].

2- Soil:

The adopted method of Krause *et al*. [14] was followed by partitioning by chloroform. 50 grams soil was shaken mechanically with 100 ml of acetone – water (3/1 v/v) for one hour in 250 ml glass stopper bottle. The extract was carefully decanted and filtered through a clean pad of cotton – 75 ml from the filtrate was concentrated by using a rotary evaporator in a water bath at 40 ^{o}C to remove acetone and then extracted twice with 100 ml chloroform. The combined chloroform was dried over anhydrous sodium sulfate and then evaporated near dryness at 40 ^{o}C using a rotary evaporator. Then, the residues of tested insecticides were determined by GC-Mass.

Determination of Pesticide Residues:

The SHIMADZU GC/MS (GC-17A), equipped with fluorescence detector was used for the chromatographic separation, the oven was programmed as follows: initial temperature 40 °C, 1.5 min, 25 °C/min to 150 °C, 0.0 min, 5 °C/min to 200 °C, 7.5 min, 25 °C/min to 290 °C with a final hold time of 12 min and a constant column flow rate of 1 mL/min. The detection of the pesticides was performed using the GC-ion trap MS with optional M*sn* mode. This scanning mode offered enhanced selectivity over either full scan or selected ion monitoring (SIM). The GC-ion trap MS was operated in the M*sn* mode and performed tandem MS functions by injecting ions into the ion trap and destabilizing matrix ions, isolating only the pesticide ions.

Results and Discussion

Efficiency of some herbicides against narrow and broad – leaved weeds in tomato plants:

Weed flora presented in the experimental area included common purslane (*Portulaca oleracea* L.) nalta jute (*Sida alba*, L.) and cocklebur (*Xanthium brasilicum*, Vellozo) as broadleaf weeds and barnyard grass (*Echinochloa colonum*, L.) as grass. All weed control treatments reduced the number and dry weight of broadleaf, grassy and total weeds than a weedy check (Table 2). Pendimethalin was more efficient than other treatments on decreasing the dry weight (92.7, 92.0 and 93.6% reduction) of grassy, broad and total weeds as compared with unweeded treatments. The insignificant difference was recorded between pendimethalin and metribuzin treatments on the number and dry weight of total weeds (Table 2). The data also indicated that two hand hoeing came in the second order after pendimethalin and metribuzin. Fluazifop- P-butyl was more effective than other treatments against grassy weeds. Fluazifop- P-butyl at 1 L /fed reduced the biomass of grassy weeds by 95.4% compared with a unweeded check. Similar results were reported by Ahmed *et al* [2], El-Metwally and Shalaby [6], Chnnappagoudar *et al* [3], Shivalingappa *et al* [4] and Shil and Nath [5].

Table (2): Number, dry weight and reduction % of tomato weeds (g/m^2) after 60 days from sowing transplanting as affected by weed control treatments during 2014/2015 season.

Weed control Treatments	Narrow-leaved			Broad-leaved			Total weeds		
	No.	Weight	% R	No.	Weigh t	% R	No.	Weigh t	% R
Unweeded	24.9	67.5	-	25.8	80.3	-	50.7	147.8	-
Hand weeding	5.0	13.5	80.0	5.3	16.5	79.5	10.3	30.0	79.7
Fluazifop- P-butyl	1.6	3.1	95.4	18.7	43.0	46.5	20.3	47.9	69.6
Pendimethalin	2.2	6.7	90.1	1.3	4.0	95.0	3.5	10.7	92.8
Metribuzin	2.9	4.9	92.7	1.9	6.4	92.0	4.8	9.5	93.6
LSD 5 %	0.5	1.1	….	1.2	1.5	….	2.3	3.4	….

% R = Percent of reduction

Table (3): The correlation between Chlorpyriphos - methyl dissipation in tomato fruits and fertilizer sources

Treatments	Without fertilizer		Bio-fertilizer		Chemical fertilizer		Organic manure		Compost	
Days	ppm	% loss	Ppm	% loss	Ppm	% loss	Ppm	% loss	ppm	% loss
*Initial deposits	34.7	…..	38.0	……	36.9	……	30.9	…….	34.1	……
1st	27.2	21.6	30.1	20.8	29.1	21.1	26.5	14.2	28.4	16.7
3rd	18.0	48.1	20.2	46.8	18.6	49.6	17.0	45.0	19.8	41.9
5th	11.9	65.7	12.6	66.8	11.4	69.1	10.6	67.6	11.9	65.1
7th	6.8	80.4	7.2	81.0	7.0	81.0	6.2	79.9	6.3	81.5
10th	3.4	90.2	3.7	90.2	3.3	91.0	2.9	90.6	3.2	90.6
15th	1.8	94.8	1.6	95.8	1.0	97.3	1.3	95.8	1.1	96.8
21st	0.36	98.9	0.52	98.6	0.28	99.2	0.5	98.3	0.44	98.7
RL_{50} days	4.7		5.43		3.77		5.05		4.72	

*= One hour after application RL_{50} = Residual half-lives

Effect of fertilizer type on chlorpyriphos – methyl and acetamiprid fate in tomato fruits:

Results in Table 3 show the concentration of the initial deposits of chlorpyriphos – methyl was (residue level after 1 h of application) 34.7, 38.0, 36.9, 30.9 and 34.1 ppm on tomato fruits of unfertilized, bio-fertilized, chemical, organic and compost fertilizer treatments, respectively. These amounts decreased to 27.2, 30.1, 29.1, 26.5 and 28.4 ppm; indicating the rate of loss 21.6, 20.8, 21.1, 14.2 and 16.7 %, respectively within 24 hrs after application. The results also revealed that the residues decreased to different degrees with the time elapsed after spraying. Thus, chlorpyriphos – methyl reached 0.36, 0.52, 0.28, 0.5 and 0.44 ppm after 21 days and the loss rate amounted to 98.9, 98.6, 99.2, 98.3 and 98.7%, respectively. In this respect, data obtained by Abbassy *et al*[15] indicated that the initial deposits of chlorpyriphos – methyl in tomato fruits was 4.05 mg kg $^{-1}$ and decreased gradually with time elapsed to reached 1.739 mg kg $^{-1}$ after 21 days post spraying.

Table (4): The correlation between acetamiprid dissipation in tomato fruits and fertilizer sources

Treatments Days	Without fertilizer		Bio-fertilizer		Chemical fertilizer		Organic fertilizer		Compost	
	ppm	% loss	ppm	% loss	ppm	% loss	Ppm	% loss	ppm	% loss
*Initial deposits	11.3	…..	10.8	……	10.4	……	11.7	…….	9.8	……
1st	8.2	27.4	7.9	26.9	8.1	22.1	9.0	23.1	7.5	23.5
3rd	5.7	49.6	5.5	49.1	5.0	51.9	6.1	47.9	4.9	50.0
5th	3.4	69.9	3.1	71.3	2.9	72.1	3.0	74.4	2.8	71.4
7th	1.6	85.8	2.0	81.5	1.8	82.7	2.1	82.1	1.7	82.7
10th	0.79	93.0	0.6	94.4	0.8	92.3	0.73	93.8	0.64	93.5
15th	0.34	97.0	0.4	96.3	ND	>99.99	0.2	98.3	0.3	96.9
21st	0.12	98.9	ND	>99.99	ND	>99.99	ND	>99.99	0.13	98.7
RL_{50} days	2.0		2.7		3.2		2.2		1.9	

***= One hour after application** **RL_{50} = Residual half-lives** **ND = none detected**

The obtained data in Table 4 cleared that the initial deposit of acetamiprid on tomato fruits was 11.3, 10.8, 10.4, 11.7 and 9.8 ppm in following treatments: unfertilized, bio, chemical, organic and compost fertilizer, respectively. These amounts dropped to 8.2, 7.9, 8.1, 9.0 and 7.5 ppm indicating the rate of loss 27.4, 26.9, 22.1, 23.1 and 23.5 %, respectively within 24 hrs for spraying. The loss of residues was increased as time elapsed, after the 10th day, more than 90 % of acetamiprid residues were disappeared in all treatments. In the same respect, after 15 days no residues were detected in the case of chemical fertilizer and at 21 days no residues were found in both bio and organic fertilizer treatments; while, negligible amounts were detected in unfertilized (0.12 ppm) and compost (0.13ppm) after 21 days. According to the maximum residue limits (MRL_s) of chlorpyriphos – methyl (1.0 mg kg^{-1}) and acetamiprid (0.2 mg kg^{-1}) in tomato fruits, presented in Anonymous [16], treated tomato fruits can be picked up after 21 days (Preharvast intervals, PHI), except in chemical and organic fertilizer in case of acetamiprid after 15 days. When the preharvest times between treated and harvest is not respected by the farmers, the risk of having higher pesticide levels is not controlled. In this case, the higher levels of pesticides can involve considerable economic losses if the MRL's established by FAO/WHO are surpassed (Shalaby et al. [1], Radwan *et al*. [17]). Our results disagree with those obtained by Abbassy *et al.* [15], who reported that the residue level of chlorpyrifos-methyl in tomato fruits after 21 days of application (1.739 mg kg G 1) was more than its maximum residue limit (MRL). The amount of residues recorded during the experimental period varies for each insecticide to another. These levels depended on the rate of use, the initial deposits, the rate of exposure of the fruits to the environmental factors and the reaction between the treated surface and the chemical applied (Solliman *et al.* [18]). Also, Stevens *et al* [19] demonstrated that uptake of pesticides on plant surface is affected by the chemical structure, formulation as well as the rate of used insecticide, the nature of the recipient surface, the used spraying equipment and the climatic conditions, especially the ambient temperature during pesticide application.

As a general trend, the first five days were, however, the most critical period at which most of the residues (> 65.0 %) were dissipated. Acetamiprid was degraded faster than chlorpyriphos – methyl, the calculated half – lives ($t_{1/2}$) values of chlorpyriphos – methyl residues were 4.7, 5.43, 3.77, 5.05 and 4.72 days in unfertilizer, bio, chemical, organic and compost treatments, respectively. The corresponding amounts of acetamiprid half-lives were 2.0, 2.7, 3.2, 2.2 and 1.9 days (Figure 1). On the contrary, Abbassy *et al.* [15] reported that the half-life time of chlorpyriphos-methyl in tomato fruits was 11.99 days.

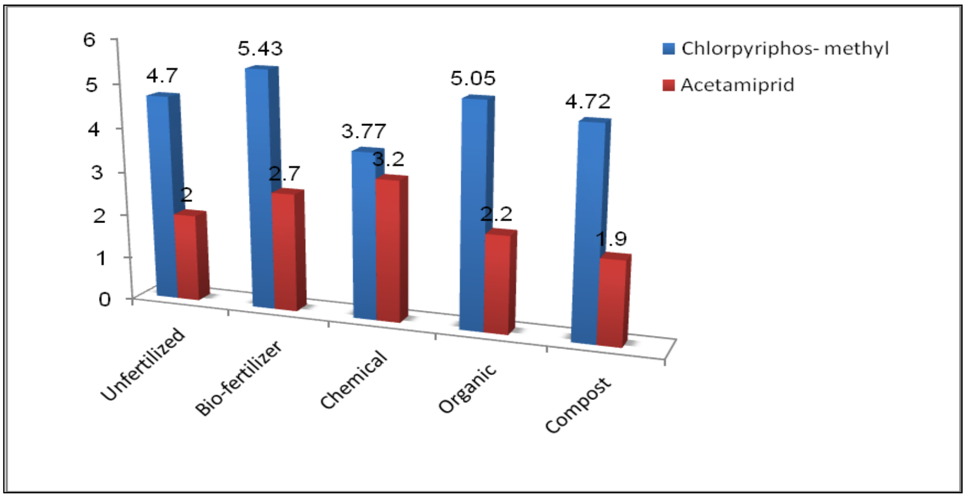

Fig. 1: Residual half –lives periods (days) of chlorpyriphos - methyl and acetamiprid insecticides in tomato fruits

Table (5): Pesticide residues (ppm) in soil after harvested tomato plants

Treatments Pesticide	Unfertilized	Bio-fertilizer	Organic manure	Compost	Chemical fertilizer	Total (ppm)
Metribuzin	0.81	0.73	0.4	0.62	0.78	3.34
Pendimethalin	0.22	0.24	ND	ND	0.18	0.64
Fluazifop-P-butyl	0.74	0.83	0.38	0.43	0.92	3.3
Chlorpyriphos-methyl	0.31	0.28	0.03	0.02	0.18	0.82
Acetamiprid	0.06	0.1	ND	ND	0.02	0.18
Total (ppm)	2.14	2.18	0.81	1.07	2.08	8.28

ND= none detected

Potential of fertilizer types on remediation soil contaminated by pesticides:

Data in Table 5 indicated that the highest amount of metribuzin residues was detected in unfertilized soil followed by chemical and bio-fertilizer (0.81, 0.78 and 0.73 ppm, respectively). The lowest concentration of this herbicide was noticed in organic (0.4 ppm) and compost (0.62 ppm). No pendimethalin residues were found in soils fertilized with organic manure and compost, but its residues 0.22, 0.24 and 0.16 ppm in unfertilized, biofertilized and chemical fertilized soils, respectively.

Fluazifop-P-butyl herbicide was applied within thirty days of a seedling to control post-emergence narrow-leaves weeds, its residues were detected in all treatments. The highest amount of this compound was 0.92 followed by 0.83 and 0.74 ppm in chemical, biofertilizer, and unfertilizer treatments, respectively. The lowest amount was noticed in organic and compost fertilizer (0.38 and 0.42 ppm). The organophosphate chlorpyriphos – methyl insecticide was used as a foliar application against insect pests attack tomato plants and fruits. Its residues in soil ranged from 0.02 (compost fertilizer) to 0.31 ppm (unfertilized soil). The neonicotinoid insecticide acetamiprid was used against sucking pests; its rate of used was 25 g / fed. So, negligible amounts were detected in unfertilized, bio and chemical fertilizer treatments (0.06, 0.1 and 0.02 ppm, respectively), while no residues were found in the case of organic and compost fertilizer. In this context, soil organic matter or soil rich in humus content are more chemically reactive with pesticides than nonhumified soil [(20,21)]. Afterwords, tested pesticides was faster disappearance in organic and compost fertilized soils (total amount of detected residues were 0.81 and 1.07 ppm, respectively) than other treatments.

In conclusion, the current study provides residue data which may be useful for assessing the amount of chlorpyriphos – methyl and acetamiprid residues in tomato fruits under Egyptian field conditions and suggests the need for implementation of these safety intervals before harvesting and marketing crops. In addition, soil organic matter or soil rich in humus content are more chemically reactive with pesticides than nonhumified soil. Thereby, the dissipation of tested pesticides in organic and compost fertilized soil was faster than in other treatments. Also, metribuzin and fluazifop-P-butyl had the highest amount (3.34 and 3.3 ppm), while acetamiprid was the lowest value (0.18 ppm).

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