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Potential of Fertilizer Types on Remediation of Pesticide Residues in Broad Bean and Soil

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Abstract: The present investigation was planned to evaluate the influence of fertilizer types on degradation of some pesticides used against broad bean pests. All tested pesticides were detected in unfertilized soil and chemical fertilizer treatments. Chlorpyrifos have the highest amount; it was detected in all analyzed samples (4.628 ppm), followed by fluazifop-p-butyl (2.976 ppm) and carbofuran (2.385ppm), while, the lowest value was noticed in indoxacarb samples (0.05 ppm) and glyphosate (0.073 ppm). The dissipation of tested pesticides was faster in organic, compost and biofertilizers treatments than other treatments. Also, despite the fact that tested pesticides dissipated more slowly in soil than in plant material, therefore, the detected amounts of these chemicals were higher in soil than those founded in both broad bean seeds and straw. On the other hand, all detected residues in seeds and been straw were below the maximum residue limits (MRL).
Keywords: fertilizer types – pesticides remediation.

Introduction

Faba bean (*Vicia faba*, L.) is one of the most popular vegetable crops in Egypt. It is an important source of dietary protein and is used in many popular dishes which are consumed by a majority of the population. Unfortunately, this crop is liable to attack by several pests, right from the early stage of growth through the late development to the post-harvest stage [1, 2, 3]. 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 [4]. 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 soil are the mineralization of organic pollutants (bioremediation of polluted soils) [5]. 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 of the same microorganisms. That may due to the difference in the physico-chemical properties of soils and also the environmental factors such as pH, moisture content and temperature as well [6]. 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 availability of pesticides for degradation of soil water. This often offset by an increase in microbial biomass, which increase the rate of degradation [7]. So, this study was carried out to determine the potential of fertilizer types on remediation of some pesticides used for controlling broad bean pests.

Materials and Methods

Field experiment:

A field experiment was conducted during the successive growing season of 2013 – 2014 at El-Mahmodah village, Dekernes district, Ad-Daqahliah governorate, Egypt. The experimental area was divided into plots of 1/100 feddan (feddan equal 4200 m^2) each arranged in randomized blocks design with three replicates for each treatment. The faba bean variety Giza 843 was cultivated at 19 November 2013 in both sides of each raw. The normal agricultural practices were achieved. The experiment area was divided into five treatments which included:

1. Without fertilizer.
2. Bio-fertilizer: A mixture of four microbial species in equal portions (*Bacillus megatherium*, *Azotobacter* sp., *Azospirillium* sp. and *Pseudomonus* sp.) it mixed with seeds before cultivated (half hour).
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 100, 150 and 48 kg / fed. of mineral nitrogen, phosphorus and potassium fertilizers, respectively.

Pesticides used: This experiment was carried out to determine the residues of 7 compounds (two herbicides and insecticides) belonging to different groups of pesticides. These pesticides were:

1. Fluazifop- P-butyl E.C. 12.5 % (Fusalide Super), this herbicide was applied at four weeks of sown to control post emergence narrow – leaves weeds and the rate of used was 2 L / fed.
2. Glyphosate EC 48 % (Raundup), this herbicide was used to control broomrape (*Orabanche crenata*, Forsk), the rate of used was75 ml/fed. three times (15 days between each others) the 1st was applied at 55 days of sown.
3. Carbofuran G 10 % (Furadan), the rate of used was 3 kg / fed. before sowing.
4. Chlorpyrifos EC 48 % (Tafaban), was used to control some insects (aphids, leafminor) its rate of use was 1 L / fed.
5. Emamectin benzoate SG 5% (Procliam blus), was used to control some insects (aphids, leafminor) its rate of use was 60 gm / fed.
6. Indoxacarb EC 15 % (Avaunt) was used to control some insects (aphids, leafminor) its rate of use was 50 ml / fed.
7. Acetamiprid SP 20% (Mospilan) was used to control some insects (aphids, leafminor) its rate of use was 50 gm / fed.

These pesticides were used a foliar application by using knapsack sprayer equipped with one nozzle boam, except carbofuran was used a soil treatment.

Sample collections:

250 g of soil, dry seeds and bean straw were collected in different treatments after bean plants harvested; samples were put in chemically clean bags and stored at -20 ^{o}C in deepfreezer until analysis. Soil samples were divided into two portions, the 1st was analyzed for soil properties, while the 2nd was analyzed for pesticide residues.

Soil analysis:

Particle size distribution, soil organic matter, EC, pH and TDS were determined according to Black *et al.* [8].

Determination of Pesticide residues:

1- Broad bean dry seeds:

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 [9].

2- Soil:

The adopted method of Krause *et al.*, [10], was followed through 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 filtrate was concentrated by using a rotary evaporator on a water bath at 40 ^{o}C to remove acetone and then extracted twice by 100 ml chloroform. The combined chloroform was dried through 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.

3- Broad bean straw:

The extraction procedure of bean straw was carried out according to Gong *et al*., [11] method with slight modifications. Fifty gm of bean straw were shaken mechanically (one hour) with 100 ml acetone, the extract was filtered through Buchner funnel containing filter paper. This step was repeated twice with 50 ml acetone; the filtrate was collected and transferred to separately funnel (250 ml), then 100 ml dichloromethane were added and surged for 2 min. The dichloromethane phase was collected after partitioning and transferred to a funnel with anhydrous sodium sulfate; the extract was concentrated to near dryness on a rotary evaporator. The extract passing through a florisil (17 g) column, which had been activated at 130 ºC for 12 h. and partially deactivated with 0.5 % water [12]. The resultant elute was evaporated on a rotary and transferred to flask and made up to a volume 5 ml with acetonitrle for GC / MS determination.

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 GCion trap MS with optional MS*n* 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 MS*n* mode and performed tandem MS functions by injecting ions into the ion trap and destabilizing matrix ions, isolating only the pesticide ions. Statistical significance of data was assessed by Duncan and Tukey test at P0.05 [13].

Results and Discussion

Effect of fertilizer sources on soil properties:

Application of different types of fertilizer had a significant effect on particle size distribution (Table 1). Significant decrease was noticed in sand and silt percent in fertilized soil compared with unfertilized (48.17 and 32.61 %), except in sand percent of chemical fertilized treatment (50.12 %); while, significant increase was happened in clay content in organic and compost treatments. Also, organic and compost fertilizer caused a significant decrease on soil pH and both treatments had significant increase in organic matter. A significant increase in EC (S.P) and TDS concentrations were noticed all fertilizer sources compared with unfertilized soil. These results were coincide with those obtained by Mohammadi *et al.*, [14], who reported that the average of organic matter content in the soil increased as a result of organic fertilizer applications. Also, Simon and Czako [15] reported that the additions of organic matter from various sources differ in the effects on soil organic matter and biological activity.

Table (1): Mechanical and physical characteristics of the tested soil

Soil samples	Particle size distribution				Chemical properties			Organic matter
	Sand	Silt	Clay	Textural class	Soil pH	EC (S.P)	TDS Ppm	
Without fertilizer	48.17b	32.61a	19.22c	Loam	8.0 a	8.37 d	697 d	1.86 b
Biofertilizer	47.93b	32.87a	19.20c	Loam	8.0 a	8.61 d	706 d	1.89 b
Organic fertilizer	43.12d	22.0b	34.88a	Clay loam	7.7 b	9.00 c	761 c	2.12 a
Compost	44.64c	23.83b	31.53b	Clay loam	7.7 b	10.38b	879 b	2.09 a
Chemical fertilizer	50.12a	30.71a	19.17c	loam	7.9 a	11.3 a	898 a	1.90 b
LSD 5 %	0.787	2.87	2.65		0.14	0.26	9.9	0.121

Table (2): Pesticide residues (ppm) in soil after harvested broad bean plants

Treatments Pesticide	Unfertilized	Bio-fertilizer	Organic manure	Compost	Chemical fertilizer	Total
Fluazifop-P-butyl	1.13	0.67	0.14	0.17	0.85	2.96
Glyphosate	0.02	0.013	ND	ND	0.04	0.073
Carbofuran	0.8	0.45	0.17	0.19	0.73	2.34
Chlorpyrifos	1.13	1.05	0.38	0.43	1.31	4.3
Emamectin benzoate	0.12	0.03	ND	ND	0.02	0.17
Indoxacarb	0.04	ND	ND	ND	0.01	0.05
Acetamiprid	0.26	0.01	ND	0.01	0.15	0.43
Total (ppm)	3.5	2.223	0.69	0.8	3.11	10.323

ND = none detected

Effect of fertilizer types on pesticides dissipation in soil:

Data in Table 2 indicated that the highest amount of fluazifop- P-butyl residues was noticed in unfertilized soil (1.13 ppm) followed by in chemical fertilized (0.85 ppm) and bio-fertilized soil (0.67 ppm). While, the lowest values were observed in organic manure (0.14 ppm) then compost fertilized soil (0.17 ppm). No glyphosate residues were detected in soils fertilized by organic manure and compost, but its residues were 0.02, 0.13 and 0.04 ppm in unfertilized, bio-fertilized and chemical fertilized soil, respectively. The amount of carbofuran could be descendingly as follow: unfertilized, chemical, bio-fertilized, compost and organic manure fertilized soil (0.8, 0.73, 0.45, 0.19 and 0.17 ppm, respectively). At the same respect, the highest amount of chlorpyrifos was detected in chemical fertilized soil, while, the lowest was observed in organic manure fertilized soil, it's ranged from 0.38 to 1.31 ppm. Emamectin benzoate residues were detected in unfertilized, bio and chemical fertilized soils (0.12, 0.03 and 0.02 ppm), but no residues were found in soils fertilized by organic manure and compost. The same trend was observed in case of indoxacarb, its amounts were 0.04 and 0.01 ppm in unfertilized and chemical fertilized soils, while, no residues were detected in other treatments. Also, acetamiprid residues ranged from 0.01 to 0.26 ppm, but no residues were found in organic manure fertilized soil. More importantly, tested pesticides more degradation in soil fertilized by organic manure followed by compost, but bio-fertilizer had a slight role in pesticides disappearance in soil. No effect of chemical fertilizer on pesticides amounts was noticed compared with those in unfertilized treatment. Similarly, Shalaby and Abdallah [16] found that adding compost and manure to carbofuran and ethoprophos- treated soil reduced the half life values from 8.86 and 8.04 days to 5.66 or 4.83 and 4.57 or 6.3 days, respectively. Similarly factors which influence root uptake and degradation of pesticides in soil are physicochemical behavior, application method and amount, physicochemical and biochemical reactions in the soil, climatic factors and plant development [17]. Microbial degradation is the breakdown of pesticides by microorganisms such as fungi, bacteria, and other soil microorganisms. Soil organic matter, texture, and site characteristics such as moisture, temperature, aeration, and pH-all affect microbial degradation [18]. Microbial activity usually is greatest in warm, moist, well aerated soils with a neutral pH. Microbial breakdown tends to increase when: temperatures are warm soil pH is favourable, soil moisture and oxygen are adequate, soil fertility is good. Microbial degradation occurs at a higher rate in the surface soil horizons, particularly in areas with high organic matter. Usually, the rate decreases with depth in the soil, where conditions such as moisture, temperature, and aeration are less favorable for microbial activity [19, 20]. Also, soil rich on humus content are more chemically reactive with pesticides than nonhumified soil [21].

Table (3): Pesticide residues (ppm) in dry broad bean seeds

Treatments Pesticide	Unfertilizer	Bio-fertilizer	Organic manure	Compost	Chemical fertilizer	Total	MRL Mg / kg
Fluazifop-P-butyl	ND	ND	ND	ND	0.01	0.01	0.2
Glyphosate	ND	ND	ND	ND	ND	ND	2.0
Carbofuran	0.01	0.003	ND	ND	0.001	0.014	unknown
Chlorpyrifos	0.002	0.001	ND	ND	0.003	0.006	0.01
Emamectin benzoate	ND	ND	ND	ND	ND	ND	0.3
Indoxacarb	ND	ND	ND	ND	ND	ND	0.5
Acetamiprid	0.011	ND	ND	ND	0.003	0.014	0.3
Total	0.023	0.004	ND	ND	0.017	0.044	

MRL= maximum residue limit according to Anonymous, 2013 ND= none detected

Table (4): Pesticide residues (ppm) in dry broad bean straw

Treatments Pesticide	Unfertilizer	Bio-fertilizer	Organic manure	Compost	Chemical fertilizer	Total	MRL Mg / kg
Fluazifop-P-butyl	0.004	ND	ND	ND	0.002	0.006	0.2
Glyphosate	ND	ND	ND	ND	ND	ND	2.0
Carbofuran	0.013	0.011	ND	ND	0.007	0.031	unknown
Chlorpyrifos	0.11	0.06	0.012	0.01	0.13	0.322	0.01
Emamectin benzoate	0.014	0.003	ND	ND	ND	0.017	0.3
Indoxacarb	ND	ND	ND	ND	ND	ND	0.5
Acetamiprid	0.011	ND	ND	ND	0.003	0.014	0.3
Total	0.152	0.074	0.012	0.01	0.142	0.39	

MRL= maximum residue limit according to Anonymous, 2013 ND= none detected

2- Effect of fertilizer types on pesticides fate in dry broad bean seeds:

Table 3 shows the amount of tested pesticides in broad bean dry seeds sown in soils fertilized by different types of fertilizers. Obtained data revealed that no residues of glyphosate, emamectin benzoate and indoxacarb were detected in all treatments, while fluazifop- P-butyl residues was found in seeds of chemical fertilized soil (0.01 ppm). Also, carbofuran and chlorpyrifos residues were detected in seeds of unfertilized, bio and chemical fertilized soils (0.01, 0.003 and 0.001 ppm for carbofuran; 0.002, 0.001 and 0.003 ppm for chlorpyrifos), but no residues of both insecticides were found in seeds of organic manure and compost fertilized soil. Acetamiprid residues in dry seeds were detected in case of unfertilized soil (0.011 ppm) and chemical fertilized soil (0.003 ppm), but no residues of this insecticide were found in other treatments. Concerning health hazards, all detected residues in dry seeds were below the maximum residue limits (MRL) according to Anonymous [22].

3- Effect of fertilizer types on pesticides residues in broad bean straw:

Generally, no residues of all tested pesticides were detected in bean straw in organic manure and compost fertilized soil (Table 4), with exception in chlorpyrifos, its amounts were 0.012 and 0.01 ppm, respectively. Also, no glyphosate and indoxacarb residues were found in all treatments. fluazifop- P-butyl residues were 0.004 and 0.002 ppm in been straw of unfertilized and chemical fertilized soil, but no residues of this herbicide detected in other treatments. On the contrary, chlorpyrifos residues were detected in all treatments, its amounts ranged from 0.01 to 0.13 ppm, while emamectin benzoate were found in been straw of unfertilized (0.014 ppm) and bio-fertilized treatments (0.003 ppm) only. Acetamiprid was appeared in straw of unfertilized and chemical fertilized soil (0.011 and 0.003 ppm, respectively). These results are agreement with those found by Führ [17], who reported that factors which influence leaf uptake and metabolism of pesticides are physicochemical behavior, formulation, droplet size and application technique, precipitation or rainfall and relative humidity, temperature, sunlight, plant species and pyhsiological differences, e.g. stomata, upper/lower leaf surface, hairs, waxes, and time of application during the vegetative period. Also**,** our results were agreement with those obtained by Jones and Norris [23]; Shalaby and Abdallah [16]; Shalaby and Abdou [24], they reported that when an insecticide reaches the soil, its fate is depended on a host of conditions including soil type, pH, organic content, mineral ion content, moisture content, the nature of the soil colloids, the flow of liquid and air through the soil, the amount of cultivation and plant growth present, and the exposure to environmental parameters, such as wind, sunlight, rain, temperature, humidity, etc. In Egypt, straw of bean is used as animal feed, fortunately all detected residues in bean straw were below the maximum residue limits (MRL) according to Anonymous [22].

Generally, the dissipation of tested pesticides in treatments fertilized by organic, compost and bio-fertilizer were faster than in both of chemical and unfertilizer treatment (Fig.1). Also, despite the fact that tested pesticides dissipated more slowly in soil than in plant material, therefore, the detected amounts of these chemicals were higher in soil than those founded in both broad bean seeds and straw. Chlorpyrifos have the highest amount; it was detected in all analyzed samples (4.628 ppm), followed by fluazifop-p-butyl (2.976 ppm) and carbofuran (2.385ppm), while, the lowest value was noticed in indoxacarb samples (0.05 ppm) and glyphosate (0.073 ppm)(Fig.2).

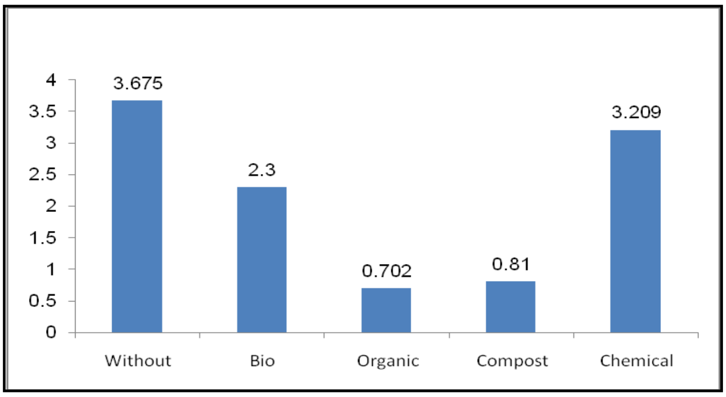

Fig. 1: Total amounts of pesticide residues were detected in different treatments

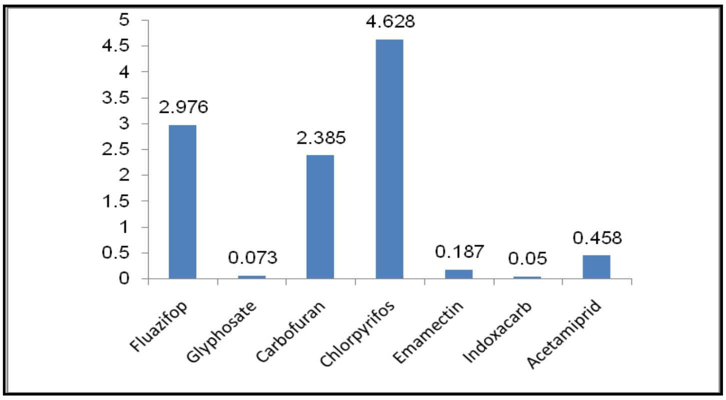

Fig. 2: Total amounts of pesticide residues were detected in all samples

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