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Effects of Insecticide Residues on Some Quality Attributes in Tomato Fruits and Determination their residues

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Abstract : Pesticides are known to interfere with the biochemical processes of plants, lowering their food quality. So this study was designed to evaluate persistence of some insecticide residues in fresh tomato fruits and investigate its effects on fruits quality. The results showed that, the preharvest interval (PHI) for tomato fruits treated by chlorpyrifos, carbosulfan and acetamiprid was more than 15 days, while it could be safely used at 15 days after spraying in case of betacyfluthrin. However, chlorpyrifos appeared to have relatively longer persistence with $t_{1/2}$ 6.86 days than other compounds. Obtained data revealed also there is fluctuation in all quality parameters activities during the experiment periods, however, the peak of these activity was happened after 3 or 5 days for application. Also, all insecticides caused significant increase in average of fruits quality parameters values.
Keywords : Tomatoes, Pesticide residues, Fruits quality

Introduction:

Rapid growth in horticultural production has been accompanied by heavy use of pesticides and by heightened concern over health effects associated with pesticide use and abuse. Potential food safety risks from pesticide residues are a significant issue for importers of fresh vegetables and fruits and market-risk factor for exporters who may have shipments detained or rejected if residues exceed allowable limits[1,2] Pesticides are known to interfere with the biochemical processes of plants, lowering their food quality[3]. The environmental and health problems and the risk involved in the use of chemicals, especially pesticides, in agriculture are very high [4], which not only leads to the chemical build up of pesticide residues in crops but also disrupts the biochemical parameters of plants. The total phenolics, total ortho-dihydroxy phenols and ascorbic acid which act as non enzymatic antioxidants in tomatoes and other crops are reported to vary because of the inability of the plants to uptake the essential micronutrients due to a biotic stress caused by pesticides [3]. So, the present work was undertaken with a view to study the behavior of four insecticides represented to various pesticide groups in fresh tomato fruits and investigates the effects of its residues on some biochemical quality parameters in tomatoes at harvest.

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. In this experiment we use four insecticides represented to various pesticide groups (Table 1), to study the effect of their residues on some parameters in treated tomato fruits. For chemical analysis, tomato fruits were harvested at firm red maturity stage after 1 hr, 1, 3, 5 and 7 days after application; while the for residue analysis, samples were collected at 1 hr, 1, 3, 5, 7, 10 and 15 days.

Table (1): Trade, common names, chemical groups and rate of used insecticides.

Trade name	**Common name**	**Chemical group**	**Recommendation rate***
Dursban	Chlorpyrifos 48 % EC	Organophosphorous	1 L / Feddan
Marshal	Carbosulfan 25 % WP	Carbamate	200 g / Feddan
Betacyfluthrin	Betacyfluthrin 10 % EC	Pyrithroid	250 ml / Feddan
Mospilan	Acetamiprid 20 % SP	Neonicotinoid	50 g / Feddan

***According to Anonymous [5].**

Determination of Pesticide residues:

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 of homogenized tomato fruits 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 [6]. The half- life ($t_{0.5}$) values were calculated using Moye's *et al*., equation [7].

Chemical analysis:

For the extraction of carotenoids (lycopine and carotene), the method suggested by Lin and Chen [8] was used with some modifications. While, ascorbic acid is a hydrophilic constituent of tomatoes, and for the extraction of ascorbic acid in tomato samples, the method proposed by Giovanelli *et al.* [9] was used with some modifications. Fresh, dry matter and total protein were determined according to the methods of Gabal *et al*., [10]. Total soluble solids (T.S.S.), total protein, lycopine, vitamin "A" as carotene and vitamin "C" as L-Ascorbic acid were determined according to A.O.A.C [11]. Macronutrients (N, P and K) and micronutrients (Fe, Mn, Zn and Cu) as well as cobalt of maize grains were determined according to Cottenie *et al*., [12].

Statistical significance of data was assessed by Duncan test at $P_{0.05}$ [13].

Results and Discussion

1. Insecticide residues:

Data in Table 2 revealed that the initial deposit amounts of chlorpyrifos, carbosulfan, betacyfluthrin and acetamiprid were 32.3, 11.63, 16.7 and 10.87 ppm (one hr after application) depending on the rate of use. These amounts dropped to 26.2, 8.23, 11.43 and 7.8 ppm indicating the rate of loss 18.9, 29.2, 31.6 and 28.2 % , respectively within 24 hrs after spraying. Generally, the loss of residues was increased with the time elapsed, after 10 days, more than 90 % of initial deposits of all insecticides were disappeared. After 15 days these percent reached 95.2, 96.8 and 97.4 % in chlorpyrifos, carbosulfan and acetamiprid treatments, while no residues were detected in fruits treated by betacyfluthrin. Concerning health hazards, the maximum residue limits (MRLs) for chlorpyrifos, carbosulfan, betacyfluthrin and acetamiprid in and on tomato fruits established by Codex Alimentarius Committee for pesticide residues [14] were 0.1, 0.1, 0.2 and 0.2 mg / kg. Accordingly, the preharvest interval (PHI = safety period) for tomato fruits treated by chlorpyrifos, carbosulfan and acetamiprid was more than 15 days, while it could be safely used at 15 days after spraying in case of betacyfluthrin insecticide. Also, the residues half-lives ($t_{1/2}$) of chlorpyrifos, carbosulfan, betacyfluthrin and acetamiprid were 6.86, 2.89, 2.35 and 2.32 days (Fig. 1) in tomato fruits, respectively. This large variation in half – life has been attributed to variation in factors such as pH, temperature, moisture content, organic carbon content, and pesticide formulation [15,16].

2. Effects of some insecticides on some biochemical parameters in tomato fruits:

Since our knowledge about the effect of tested insecticides on the quality attributes in tomatoes is quite meager. It is of interest to investigate the effect of these compounds on fresh, dry weight, T.S.S, total protein percent, lycopine, vitamin "A" as carotene, vitamin "C" as ascorbic acid, macronutrients and micronutrients (Tables 3-7).

2.1. Effects on fresh and dry weight:

Data in Table 3 indicated that all tested insecticides caused increase in fresh weight comparison with untreated plants. High fresh weight was noticed in acetamiprid treated fruits (the average was 118.4 g), followed by betacyfluthrin and chlorpyrifos (108.6 and 107.6 g). While dry weight of fruits was lower than it in untreated plants except in case of acetamiprid insecticide. Low dry weight was happened in carbosulfan treated plants (4.82 g) followed by betacyfluthrin and chlorpyrifos (5.14 and 5.56 g). Also, our results revealed that there is a significant increase in the average of fresh weight in all pesticides treated tomatoes compared with untreated except in case of carbosulfan; on the contrary, there is significant decrease in dry weight in all treatments except in case of acetamiprid compared with control. In the same trend data obtained by Radwan *et al*., [1] indicated that pirimiphos-methyl caused significant increase in dry matter of green pepper, while profenofos induced significant decrease. Also, who reported that no significant changes in dry mater of eggplant treated by both insecticides. Increasing dry weight of tomato fruits indicate positive effect due to several induced effects in hormonal synthesis and metabolic activity while reduce some enzymes such as peroxidase activity in plants and hence increasing the catabolism rather than anabolism [17] which reflected on tomato fruits weight and improve its quality.

2.2. Effects on total soluble solids (TSS) and total protein:

Obtained data (Table 4) revealed that all insecticides caused increase in TSS percent of tomato fruits comparison with untreated plants (the average was 2.5 %). High percent was noticed in chlorpyrifos treated fruits (5.02 %) followed by acetamiprid then betacyfluthrin and carbosulfan (the average was 3.82, 3.53 and 2.88 %, respectively). The same trend was observed in total protein, all compounds gave increase in protein percent, the high amount was noticed in acetamiprid treated fruits followed by betacyfluthrin and carbosulfan (the average was 0.54, 0.486 and 0.41 %, respectively).

2.3. Effects on Lycopine, Carotene and Ascorbic acid concentrations:

Data in Table 5 showed that all tested insecticides caused increase in lycopine concentration in treated plants comparison with control except in case of chlorpyrifos. The same trend was observed in carotene, acetamiprid caused high increase (the average was 12.82 µg / g) while betacyfluthrin and chlorpyrifos caused decrease in carotene amount (8.61 and 8.87 µg / g). On the other hand, a negligible change of ascorbic acid amount was happened in treated plants compared with untreated plants, acetamiprid had the highest amount (the average was 13.49 µg / 100 g of fresh weight). These results agree with obtained by Chauhan *et al*., [3] who reported that pesticides are known to interfere with the biochemical processes of plants, lowering their food quality. The amounts of total soluble solids, total protein, vitamin "A" as corotenoids, vitamin "C" as ascorbic acid and lycopine in tomato fruits were significantly increased compared with those obtained by control. In this concern, Nadia Gad and Nagwa Hassan[(18)] revealed that cobalt and chicken manure caused significantly increased in all chemical constituents such as total protein, total soluble solids, total soluble sugars, lycopine and vitamin "C" compared with control. Increasing all chemical constituents improve the quality of tomato fruits. Vitamin "A" carotenoids is an important antioxidant and is essential to human growth, normal physiological functions, health of the skin as well as mucus membranes. Moreover, vitamin "C" is an antioxidant and is necessary to several metabolic processes[19].

2.4. Effects on macronutrients concentration:

Data in Table 6 showed a positive response of macronutrients percent to different insecticides. Our results revealed that all compounds caused significant increase in nitrogen percent in treated tomatoes compared with control. The highest concentration was happened in acetamiprid followed by chlorpyrifos (3.14 and 2.85 %, respectively), while the lowest increase was observed in carbosulfan (1.82 %). In this context, similar effects was noticed in phosphorus content, chlorpyrifos and betacyfluthrin caused high concentration (0.662 and 0.609 %) followed by acetamiprid (0.565 %), but no significant changes between carbosulfan and untreated tomatoes was noticed (0.395 and 0.407 %, respectively). The data revealed also all tested chemicals caused significant increase in potassium concentration. Chlorpyrifos caused high increase (the average was 9.2 %) while carbosulfan gave the lowest increase (6.5 %).

2.5. Effects on micronutrients concentration:

Presented data in Table 7(a & b) showed the effect of different insecticides on micronutrients (Fe, Cu, Mg and Zn) composition of tomato fruits. Data revealed that all used insecticides caused significantly increased the average amounts of these elements compared with untreated plants. Chlorpyrifos caused high increase in manganese content (34.14ppm) followed by acetamiprid (31.18ppm) while carbosulfan caused the lowest increase (25.5 ppm). The same trend was noticed with zinc amounts, chlorpyrifos gave high amount (the average was 25.14 ppm) followed by other insecticides, acetamiprid, carbosulfan and betacyfluthrin (there is no significant changes between each other). High increase of iron amount was observed in acetamiprid treated plants (105.8 ppm) followed by chlorpyrifos and betacyfluthrin (104.6 and 104.4 ppm). Also, chlorpyrifos caused high increase in copper content (20.86 ppm) while the lowest amount was noticed in carbosulfan insecticide (14.1 ppm).

Generally, our results revealed that there is a fluctuation in all quality parameters activities during the experiment periods, however, the peak of these activity was happened after 3 or 5 days for application. Also, all insecticides caused significant increase in average of tomatoes quality parameters values.

In conclusion, this study provides residue data which may be used to propose MRL′s for these insecticides on tomatoes under Egyptian field conditions and suggests the need for implementation of these safety intervals before harvesting and marketing such crop fruit, and the possible effect on fruit quality that should be considered to avoid problems which may arise as a result of side effects of insecticides.

Table (2): Residue of some insecticides in and on tomato fruits

Insecticides / Periods	Chlorpyrifos		Carbosulfan		Betacyfluthrin		Acetamiprid	
	ppm	% loss	ppm	% loss	ppm	% loss	ppm	% loss
Initial deposits*	32.3	…..	11.63	…..	16.7	…..	10.87	…..
1st day	26.2	18.9	8.23	29.2	11.43	31.6	7.8	28.2
3rd days	17.6	45.5	6.1	47.5	7.91	52.6	5.6	48.5
5th days	10.7	66.9	3.82	67.2	3.12	81.3	2.8	74.2
7th days	5.94	81.6	2.04	82.5	1.8	89.2	1.34	87.7
10th days	3.14	90.3	0.93	92.0	0.4	97.6	0.67	93.8
15th days	1.56	95.2	0.37	96.8	ND	> 99.9	0.28	97.4
MRL*	0.1		0.1		0.2		0.2	
$T_{1/2}$ days	6.86		2.89		2.35		2.32	

*one hour after application ND = none detected *MRL = maximum residue limits (Anonymous, 2013) $t_{1/2}$ = residual half-lives

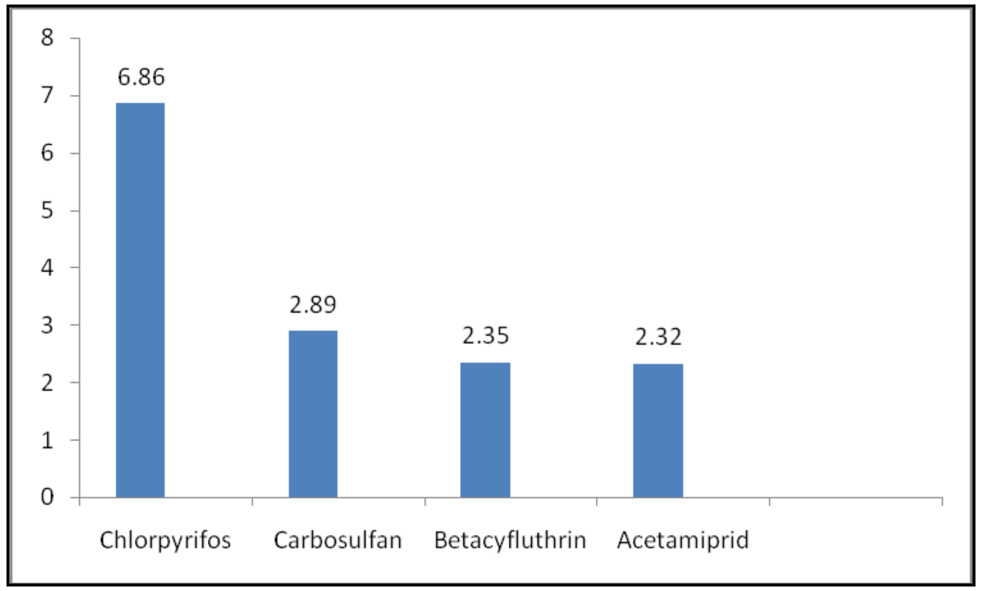
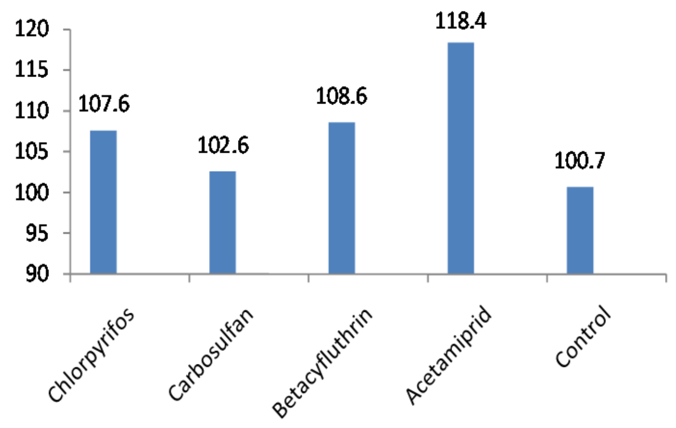


Fig. 1: Residual half –lives periods (days) of tested insecticides in tomato fruits

Table (3): Effect of pesticide residues on fresh and dry weight of treated tomato fruits

Parameters / Treatments	Fresh (g)						Dry (g)					
	1 hr	1 day	3 days	5 days	7 days	Average	1 hr	1 day	3 days	5 days	7 days	Average
Chlorpyrifos	79.6	96.7	114.7	134.3	112.9	107.6 b	4.4	6.8	4.8	5.4	6.4	5.56 b
Carbosulfan	72.0	74.3	154.4	106.6	105.9	102.6 c	4.2	3.7	6.6	5.3	4.3	4.82 d
Betacyfluthrin	84.4	79.8	133.6	141.7	103.3	108.6 b	4.2	3.5	7.0	5.9	5.1	5.14 c
Acetamiprid	101.8	169.5	109.6	103.5	107.7	118.4 a	5.2	9.7	5.4	4.7	4.8	5.96 a
Control	71.9	85.1	132.1	104.8	109.6	100.7 c	4.2	3.9	8.7	7.6	5.1	5.9 a
LSD 5 %	2.74						0.13					



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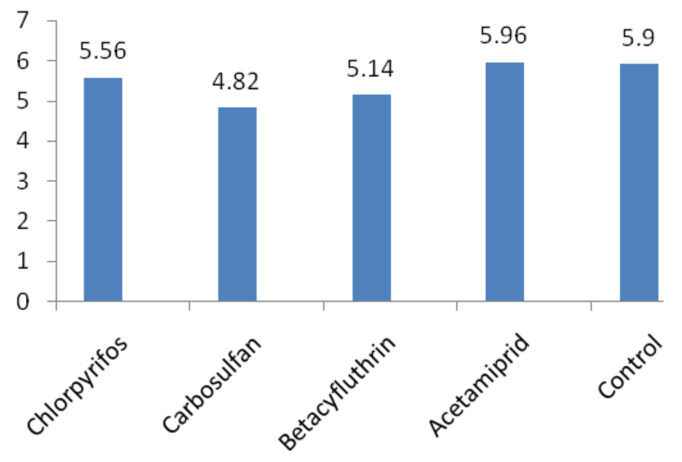


(B)

Figure 2): Effect of pesticide residues on the average of fresh (A) and dry weight (B) of treated tomato fruits

Table (4): Effect of pesticide residues on total soluble solids (TSS) and total protein (%) of treated tomato fruits

Parameters / Insecticide	Total soluble solids (TSS) %						Total Protein (%)					
	1 hr	1 day	3 days	5 days	7 days	Average	1 hr	1 day	3 days	5 days	7 days	Average
Chlorpyrifos	**4.53**	**6.77**	**4.34**	**6.45**	**3.02**	**5.02 a**	**0.21**	**0.7**	**0.52**	**0.16**	**0.23**	**0.36 b**
Carbosulfan	**3.56**	**2.21**	**2.83**	**2.83**	**2.98**	**2.88 d**	**0.49**	**0.29**	**0.41**	**0.39**	**0.46**	**0.41 b**
Betacyfluthrin	**4.23**	**3.89**	**4.36**	**1.77**	**3.42**	**3.53 c**	**0.59**	**0.52**	**0.65**	**0.18**	**0.49**	**0.49 a**
Acetamiprid	**2.91**	**4.65**	**4.2**	**3.01**	**4.32**	**3.82 b**	**0.33**	**0.76**	**0.67**	**0.24**	**0.7**	**0.54 a**
Control	**2.69**	**2.31**	**2.96**	**1.69**	**2.87**	**2.5 e**	**0.39**	**0.31**	**0.34**	**0.21**	**0.47**	**0.34 b**
LSD 5 %	**0.124**						**0.07**					

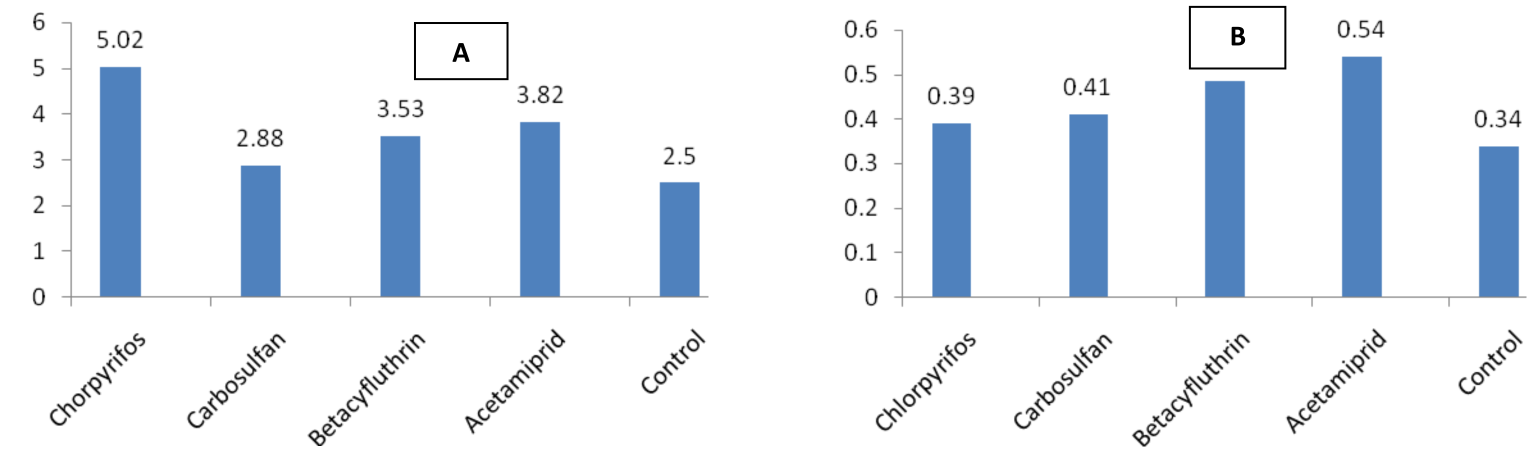


Figure 3): Effect of pesticide residues on the average of total soluble solids (A) and total protein (B) of treated tomato fruits

Table (5): Effect of pesticide residues on lycopine, carotene and ascorbic acid concentrations in treated tomato fruits

Parameters	Lycopine (µg / g FM)						Carotene (µg / 100g fresh tissue)						Ascorbic acid (mg / 100g fresh tissue)					
Treatments	1 hr	1 day	3 days	5 days	7 days	Average	1 hr	1 day	3 days	5 days	7 days	Average	1 hr	1 day	3 days	5 days	7 days	Average
Chlorpyrifos	6.69	11.43	4.83	5.04	4.64	6.53 e	8.45	19.02	8.78	4.86	3.23	8.87 d	11.71	14.66	12.53	10.82	13.0	12.54 e
Carbosulfan	11.51	8.7	8.09	5.5	8.55	8.47 c	10.19	12.93	11.12	6.4	14.14	10.96 b	13.46	10.92	13.01	12.89	13.19	12.69 d
Betacyfluthrin	8.78	10.79	12.56	4.19	19.43	11.15 a	9.79	15.88	7.55	2.0	7.84	8.61 e	12.84	11.79	14.39	14.02	14.69	13.55a
Acetamiprid	6.68	3.54	6.71	21.49	11.88	10.06 b	13.07	2.56	9.95	22.12	16.38	12.82 a	13.19	13.97	14.51	11.91	13.87	13.49 b
Control	10.67	4.64	6.86	5.14	9.10	7.28 d	12.36	4.77	8.3	6.82	20.37	10.52 c	13.8	11.3	14.9	10.0	14.4	12.88 c
LSD 5 %	0.17						0.102						0.044					

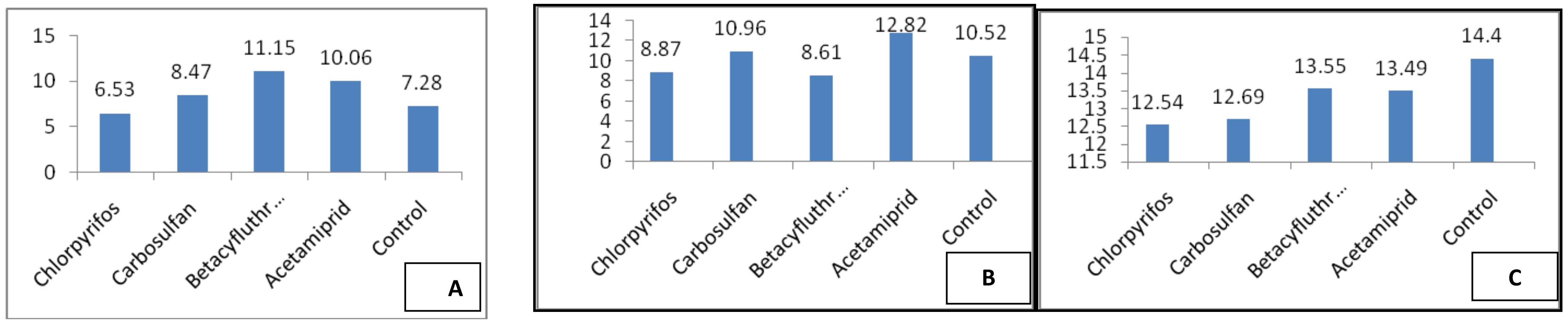


Fig.4. Effect of pesticide residues on the average of lycopine (A), carotene (B) and ascorbic acid (C) concentrations

Table (6): Effect of insecticide residues on macronutrients concentration in treated tomato fruit

Parameters	N (%)						P (%)						K (%)					
Insecticides	1 hr	1 day	3 days	5 days	7 days	Average	1 hr	1 day	3 days	5 days	7 days	Average	1 hr	1 day	3 days	5 days	7 days	Average
Chlorpyrifos	3.38	3.62	2.36	3.78	1.12	2.85 b	0.652	0.683	0.661	0.711	0.602	0.662 a	10.4	11.18	8.32	12.48	3.64	9.2 a
Carbosulfan	2.31	1.18	1.81	1.77	2.06	1.82 d	0.492	0.343	0.381	0.369	0.392	0.395 c	7.8	4.68	6.5	6.24	7.28	6.5 d
Betacyfluthrin	2.59	2.34	3.36	2.29	1.59	2.43 c	0.643	0.629	0.650	0.578	0.543	0.609ab	9.36	8.32	10.4	2.86	7.8	7.75 c
Acetamiprid	3.47	2.59	3.41	2.72	3.5	3.14 a	0.654	0.544	0.672	0.646	0.566	0.565 b	5.2	12.22	10.66	3.9	11.18	8.63 b
Control	1.72	1.23	1.62	1.16	2.28	1.6 e	0.398	0.356	0.375	0.339	0.569	0.407	6.24	4.94	5.46	3.38	7.56	5.52 e
LSD 5 %	0.036						0.062						0.139					

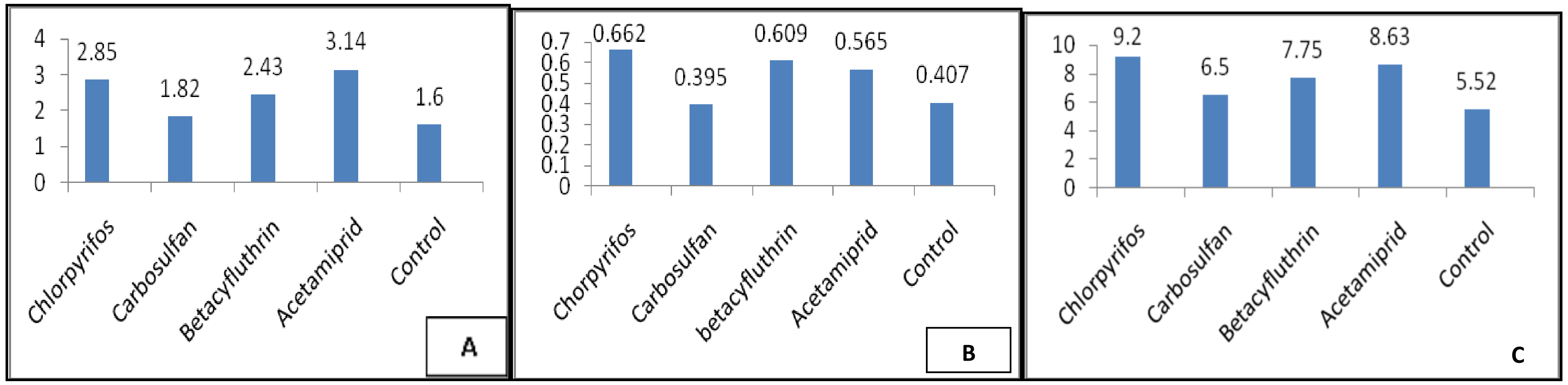
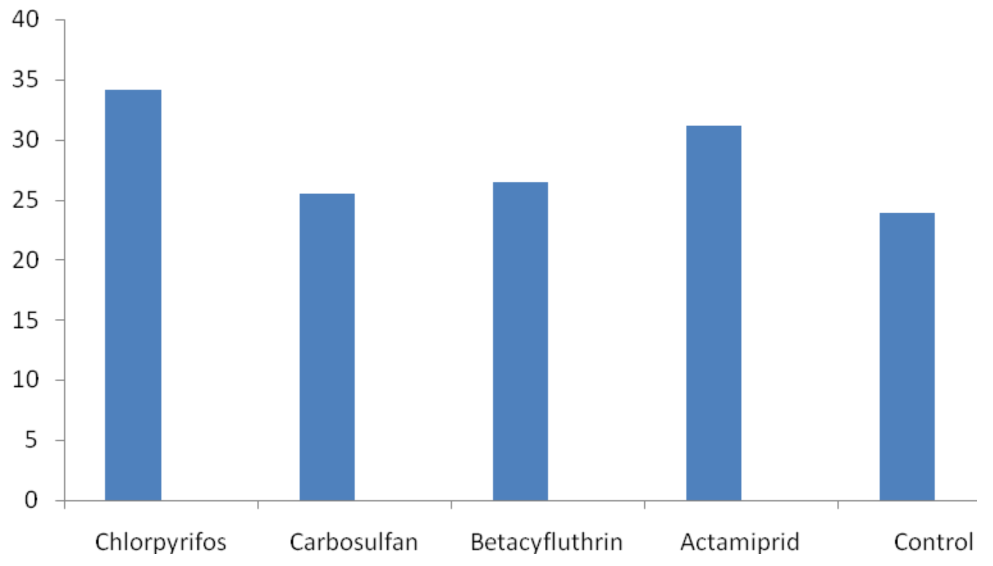


Fig.5. Effect of pesticide residues on the average of nitrogen (A), phosphorus (B) and potassium (C) concentrations

Table (7a): Effect of pesticide residues on micronutrients (ppm) concentrations in treated tomato fruits

Treatments	Mn (ppm)						Zn (ppm)					
	1 hr	1 day	3 days	5 days	7 days	Average	1 hr	1 day	3 days	5 days	7 days	Average
Chlorpyrifos	33.3	34.5	34.9	36.5	31.5	34.14 a	24.5	23.2	26.1	28.0	23.9	25.14 a
Carbosulfan	28.3	22.9	25.4	24.2	26.7	25.5 d	26.0	19.1	23.5	21.9	24.4	23.0 b
Betacyfluthrin	27.5	27.3	27.8	25.8	24.4	26.56 c	23.5	23.5	23.2	20.8	21.2	22.44 b
Acetamiprid	32.3	30.0	33.4	32.2	28.0	31.18 b	24.0	22.6	25.2	23.4	20.5	23.14 b
Control	26.5	22.7	23.0	21.8	25.4	23.88 e	23.3	19.0	20.5	18.2	22.0	20.6 c
LSD 5 %	0.842						1.13					



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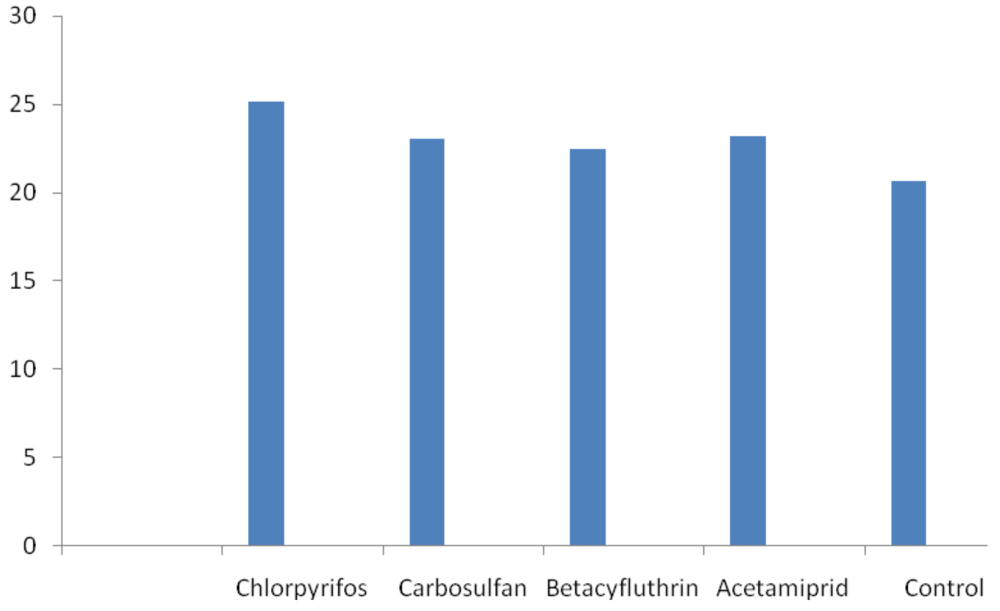

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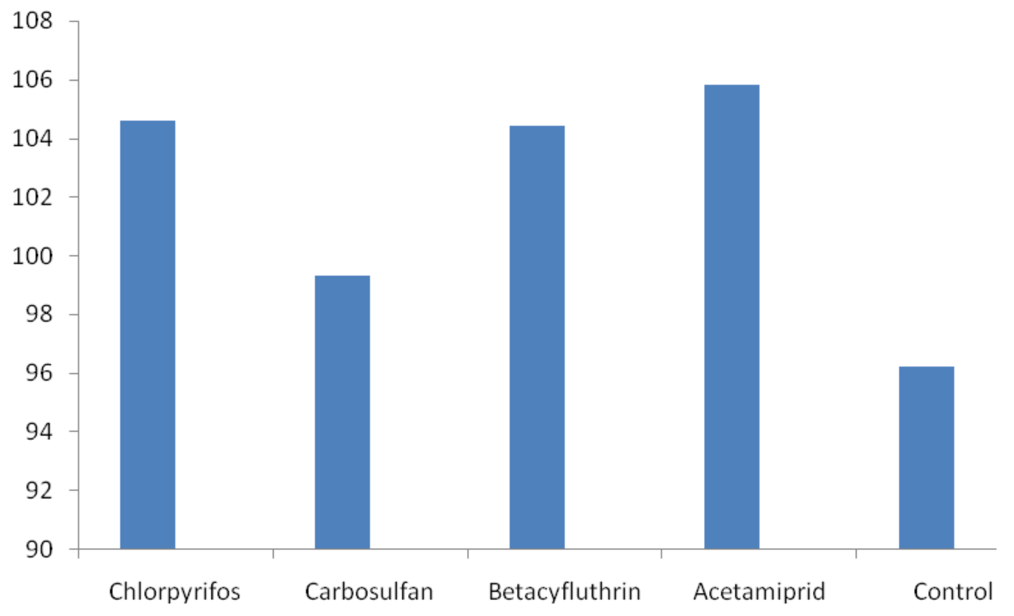
Fig. 6: Effect of pesticide residues on the average of Manganese (A), Zinc (B) concentrations

Table (7b): Effect of pesticide residues on micronutrients (ppm) concentrations in treated tomato fruits

Treatments	Fe (ppm)						Cu (ppm)					
	1 hr	1 day	3 days	5 days	7 days	Average	1 hr	1 day	3 days	5 days	7 days	Average
Chlorpyrifos	105.0	106.0	103.0	108.0	101.0	104.6 b	19.2	21.1	21.0	23.4	19.6	20.9 a
Carbosulfan	102.0	98.7	100.0	97.8	97.8	99.3 c	17.3	14.9	12.8	12.0	13.3	14.1 c
Betacyfluthrin	104.0	101.0	105.0	107.0	105.0	104.4 b	16.6	16.0	17.5	15.9	14.7	16.1 b
Acetamiprid	107.0	105.0	108.0	103.0	106.0	105.8 a	16.8	16.1	15.4	15.0	16.0	15.9 b
Control	98.3	96.8	97.5	93.6	95.0	96.2 d	13.5	12.9	13.0	11.8	14.4	13.1 d
LSD 5 %	0.816						0.889					



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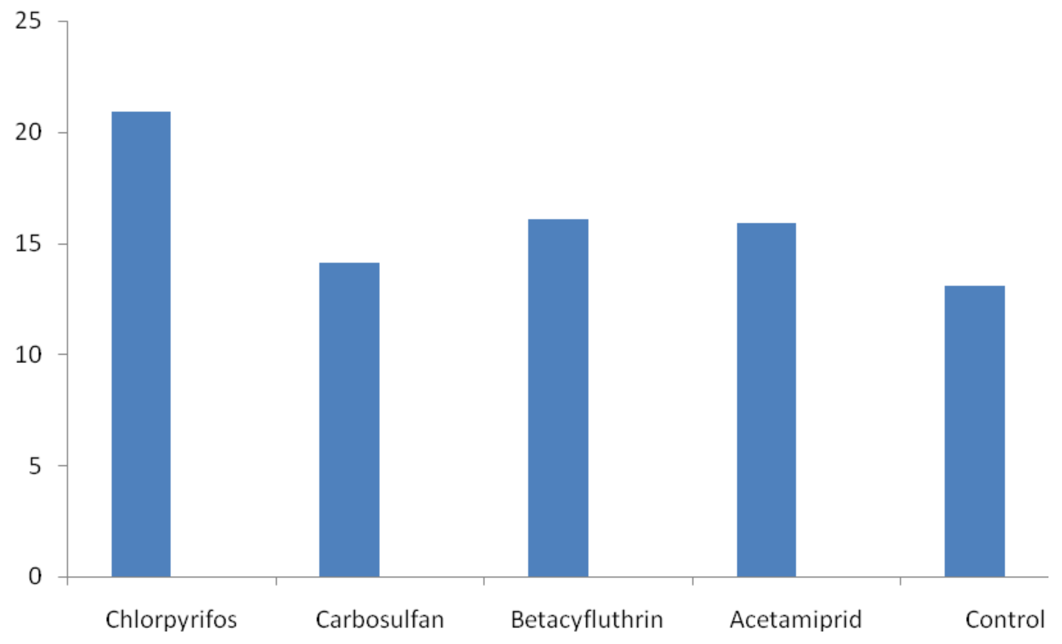

(B)

Fig. 7: Effect of pesticide residues on the average of Iron (A), Copper (B) concentrations

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