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EFFECT OF PREPARATION AND PROCESSING ON SOME PESTICIDE, FUNGICIDE AND HEAVY METALS RESIDUES IN TOMATOES

A.A. El-Bedawey[1], A. E. El-beltagy[1], E. M. El-Kholie[2] and M. B. Azzazy[1]

[1] Dep. of Food Science and Technology, Faculty of Agriculture, Minufiya University
[2] Dep. of Nutrition and Food Science, Faculty of Home Economics, Minufiya University

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ABSTRACT: *The aim of this research was to study the effect of preparation and processing on the percentage loss of some pesticide and fungicide residues as well as some heavy metal (iron, zinc, manganese, lead and copper) residues in tomato.*

The effect of preparation and processing on the pesticides, fungicide and heavy metal residues were evaluated. Generally, all preparation and processing of tomato fruits remarkably increased the percentage of loss of all pesticide and fungicide residues some of processing treatments lead to a complete loss in some pesticides such as:

Different processing of tomato with spintor, pride and vertimic.

Paste processing with reldan and diazinon.

Ketchup processing with reldan and cascade. It is clear to notice that washing reduced the percentage loss of pesticides more than 50%, while the heat reduced it to 100% with some pesticides and 70% with the other pesticides.

On the other hand, the different treatments by using fertilizers lead to increase the values of different heavy metals especially iron, zinc and manganese than those in control samples.

Key words: *Pesticide, Fungicide, Heavy metal and Tomato products.*

INTRODUCTION

Pesticides are used for pest control in large quantities all-over the world and particularly in developing countries. Although pesticides chemicals provide numbers of benefits in terms of increased production quality, but pesticides residues in the environment are of concern everywhere (Sannino *et al.*, 1999).

It is well known that pesticide residues and their metabolites can find their way into the food chain from several sources, the main reason for application of pesticides on crops is to control various pest species. However, the direct and the indirect pollution of food with pesticides that persist in the environment is the another source of contamination (Cabras *et al.*, 1998).

Deposite of all debates concerning pesticides to the country, the essentiality of pest-control chemicals to adequate food production, manufacture, marketing and storage, especially in developing countries is out of question. However, without continuous surveillance and intelligent control, some of those, that persist in our foods, could conceivable endanger the public health (Sannino *et al.,* 1999).

Tap water washing of tomatoes after one hour from the application removed 31.69% of chloropyrifos-methyl, 64.91% of diazinon and 26.41% of phenthoate, while The corresponding value after peeling were 95.07, 76.02 and 94.32%, respectively (Hegazy *et al.,* 2004).

Hanuman-Tharju and Awasthi (2003) reported that fungicide residues, decreased upon washing and cooking of contaminated tomato fruits collected after 5 and 10 days of spray application. The rate of loss of the residues was almost the same from both application rates (0.2 and 0.4%) upon washing and washing followed by cooking, while the intensity of dislodging the residues was more due to washing followed by cooking.

The aim of this research is to study the effect of preparation and processing of tomatoes on the percentage losses of pesticide, fungicide and heavy metal residues in tomatoes fruits and its processed products.

MATERIALS AND METHODS

1. Materials:

1.1. Tomato seeds:

Tomato seeds (Nada, from Petoseed Company, U.S.A.) were obtained from Samtrade Company located in El-Maadi, Cairo, Egypt.

1.2. Tested pesticides:

Cascade, pride and kuik were obtained from Samtrade Company but Reldan, Diazinon, Match, Actara, Spintor and Vertimic were obtained from Agriculture Local Market in Ismaelia region and its characteristics are shown in Table (A) according to The Pesticide Manual (2006) and all of these pesticides were used according to the Recommended Dose by the Egyptian Ministry of Agriculture.

1.3. Tested fungicides:

Teldor, Milor.-Co., Previure-N. and Idroram were obtained from Samtrade Company and its characteristics are shown in Table (B) according to The Pesticide Manual (2006) and used according to the Recommended Dose by the Egyptian Ministry of Agriculture.

Table (A). Characteristics of tested pesticides used in this investigation.

Chemical names	Active ingredients	Common names	$L.D_{50}$ (mg/kg)	Purity (%)
o,o-dimethyl o-(3, 4, 6-trichloro-2-pyridinyl) phosphorothioate	Chloropyrifos-methyl	Reldan	> 3000	97
o,o-diethyl o-[6-methyl-2- (1-methylethyl)-4-pyrimidinyl] phosphorthioate	Diazinon	Diazinon	1250	≥ 95
N-[[[2, 5 dichloro-4-(1, 1, 2, 3, 3, 3-hexa fluoropropoxy) phenyl] amino]carbonyl]-2, 6 difluorobenzamide	Lufenuron	Match	> 2000	≥ 98
N-[[[4-[2-chloro-4-(trifluoromethyl) phenoxy]-2-fluorophenyl] amino] carbonyl]-2, 6 difluorobenzamide	Flufenoxuron	Cascade	> 3000	≥ 95
3-[(2-chloro-5-thiazolyl) methyl] tetrahydro-5-methyl-N-nitro-4 H-1, 3, 5 oxadiazin-4-imine	Thiamothoxam	Actara	1563	98
2-[(6 deoxy-2, 3, 4-tri-o-methyl-α-L-mannopyranosyl) oxy]-13- [[5-(dimethyl amino) tetrahydro-6-methyl-2H-pyran-2-yl]oxy]-9-ethyl	Spinosad	Spintor	3783 male rats > 5000 female rats	> 90
4-[[4-1, 1-dimethyl-ethyl) phenyl] ethoxy] quinazoline	Fenazaquin	Pride	134 male-r 138 female-r	98
methyl N-[[(methyl amino) carbonyl] oxy] ethonimidothioate	Mithomyl	Kuik	34 male-r 30 female-r	≥ 98
5-o-demethyl avermectin A_{1a} (i) mixture with 5-o-demethyl-25-de (1-methyl-propyl)-25-(1-methyl-ethyl) avermectin A_{1a} (ii)	Abamectin	Vetimic	10	99

$L.D_{50}$: The dose which kill 50% from experimental animals
Ref. The Pesticide Manual (2006). Twelfth Edition, Editor CDS tomlin. British Crop Protectino Council.

Table (B). Characteristics of tested fungicides used in this investigation.

Chemical names	Active ingredi-ents	Common names	L.D$_{50}$ (mg/kg)	Purity (%)
N-(2, 3-dichloro-4-hydroxyphenyl)-1-methylcyclohexane-carboxamide	Fenhexamid	Teldor	> 5000	> 95
Methyl N-(2, 6 dimethyl-phenyl)-N-(methoxy-acetyl)-DL-alaninate	Metalaxyl	Milor-co	633	98
Propyl [3-dimethyl-amino propyl] carbamate hydro-chloride	Propamocarb hydrochloride	Previcure-N	2000 – 2900	99
Copper hydroxide sulfate	Copper sulfate	Idroram	100	98

Ref. The Pesticide Manual (2006). Twelfth Edition, Editor CDS tomlin. British Crop Protectino Council.

1.4. Tested fertilizers:

Librel BMX Fe, Zn, Mn were obtained from Agriculture Local Market in Ismaelia region, and used according to the Recommended Dose by Ministry of Agriculture and Manufactured Company.

2. Methods:

2.1. Preparation and processing of tomato samples:

The tomato samples were harvested after one day of spraying and divided into groups for processing techniques as following:

a. Raw tomato: the tomato samples were taken directly without any treatments to determine the pesticide and heavy metal residues after one day and after one week from spraying.

b. Washing: the tomato samples were washed by running tap water for 3 minutes.

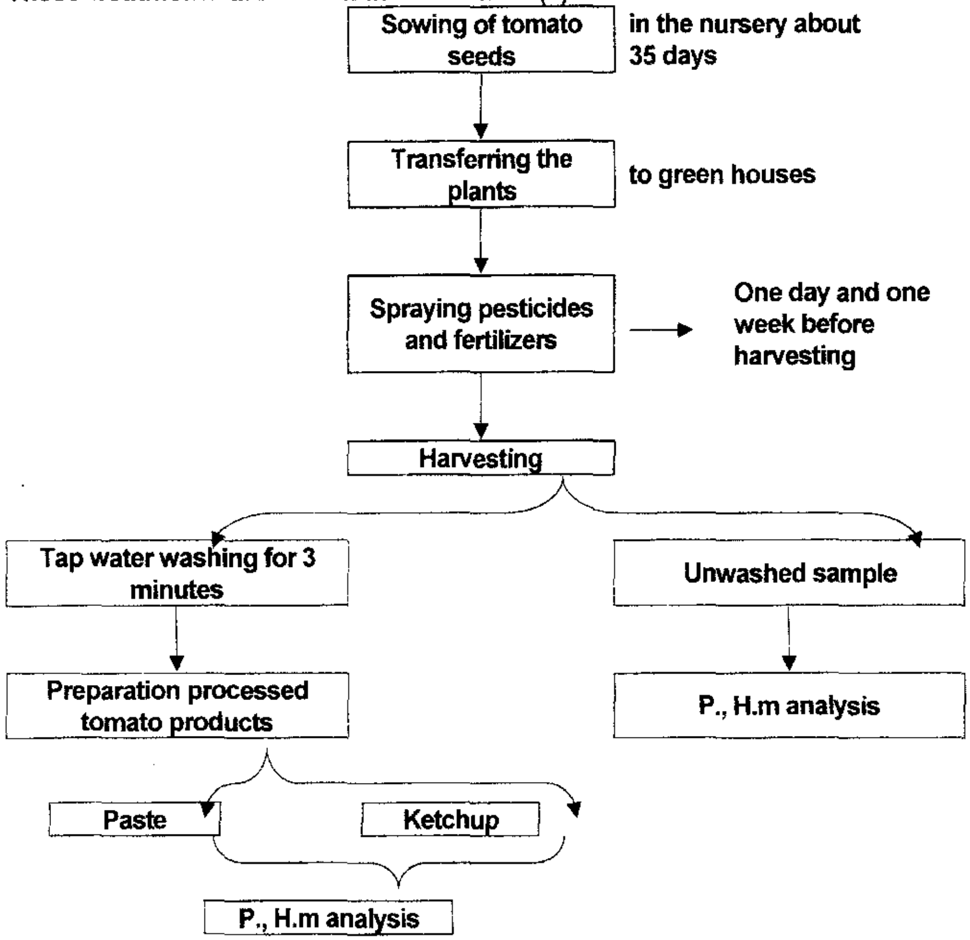
c. Paste processing: the tomato samples were processed to tomato paste by the following manner:

   1. Tomatoes samples were washed in running tap water for 3 minutes to remove any soil grits. Then crushed by using electric blender (Al Araby M × 900), and the seeds and husks were removed by sieving from the juice.
   2. Tomato juice previously extracted was heated to remove water and increase the T.S.S. to 33%. The concentration process was done by direct heat with stirring. The paste was cooled to room temperature, packed and used for analysis.

d. Ketchup processing: the same previous method in paste preparing tomato was followed but the concentration was 25% and a mixture of spiceses was added during boiling.

e. Control sample: the sample was taken directly without any treatments and without any spraying pesticides or fertilizers to determine the pesticide and heavy metal residues.

These treatments are shown in Flow Sheet (1).



Flow sheet (1). Tomato preparation and processing for pesticides and heavy metals analysis.

P., H.m: Pesticides and heavy metals analysis.

2.2. Pesticide and fungicide extraction methods:

The analytical method of the A.O.A.C (2005) was applied.

2.3. Determination of organophosphorus, actara and fungicides residues by using gas chromatography (GC):

The tested organophosphorus pesticides were determined using a Hewlett Packard 5980 series II gas chromatograph Fitted with both flame photometric detector (FPD) with 550 nm phosphorus filter and a nitrogen phosphorus detector (NPD) attached to a DB 1701 column (30 m × 530 um × 1.0 um film), with the split made with the split temperature ratio 250°C using auto injector. Helium was used as the carrier gas at a pressure of 10 psi. The detector temperature was maintained at 250°C and the oven temperature programed from an initial temperature was 100°C (hold for 0.5 mm) to 250°C at 10°C / min. and hold for 10 min. The pesticide was detected with both flame photometric detector (FPD) and nitrogen phosphorus detector (NPD) at 330°C according to the method described by (A.O.A.C, 2005).

2.4. Determination of tested another pesticide residues by using high performance liquid chromatography (HPLC):

A HPLC system consisting of a Perkin Elmer (Norwalk, CT, USA) model 410 LC pump, an ISS 200 injector with a 150 ml sample 100 p and a 5 um particle size, 250 mm × 4.6 mm C18 column, (Pickering Laboratories, Inc., Mountain View, CA, USA), attached to fluorescence LC detector 240, Perkin Elmer). Samples were run using the mobile phase 75% water : 25% methanol for 5 min). The system was attached pickering Laboratories Postcolumn deirvatization reactor. The analytes, after elution from the column were hydrolysed by NaOH (Hydrolysis Reagent, Pickering Laboratories) and then coupled with OPA reagent (consisting of 100 mg of 0-phthaladehyde and 2 g of thioflor, N.N.-dimethyl-2-mercaptopropionic acid, mixed in 950 ml of phthaladehyde dilutent), (Pickering Laboratories, Inc., Mountain View, CA, USA). The fluorescence diervatization of carbamate residues were detected using detector LC 240 (Perkin Elmer, Norwalk, CT) and operated at an excitation wavelength of 250 nm, and a emission wave length of 450 nm. 10 ml of extracted sample was injected. Peaks were recorded and quantified using Turbochrom 4.0 Soft ware (PE Nelson) according to Columns Method for Pesticide Analysis (1993).

The recovery percentage of these pesticides is shown in Table (C). The sensitivity and recovery for the applied methods are carried out using samples that spiked with 250 ppm of the tested pesticides. All the obtained results had been corrected according to the recovery percentage of the individual analyzed pesticide sample.

2.5. Determination of heavy metals:

Some heavy metals were determined in tomato samples using the atomic absorption spectrophotometric technique. The different minerals elements were determined by the method of Nation and Robinson (1971).

Tomatoes samples were digested with 3 : 2 : 1 v/v of nitricacid : perchloric: sulphuric acid digestate were used to determine some heavy metal using the atomic.

Table (C). Recovery percentage of tested pesticides and fungicides.

Tested pesticides and fungicides	% of recovery
Reldan	92.20
Diazinon	95.31
Match	90.42
Cascade	91.22
Actara	97.10
Spintor	82.39
Pride	92.10
Kuik	85.02
Vertimic	91.80
Teldor	94.37
Milor Co.	91.50
Previcure N.	94.05
Idroram	92.34

RESULTS AND DISCUSSION

1. Pesticide residues in raw and processed tomatoes:

1.1. Organophosphorus residues in tomatoes:

Data in Table (1) shows the effect of preparation and processing on the percentage loss of organophosphorus residues in Raw and processed tomatoes. The concentration of tested organophosphorus insecticide residues in unwashed tomatoes samples were 5.37 and 3.97 ppm for reldan and diazinon, respectively.

It is clear to notice that all tested tomatoes samples had higher organophosphorus residues (reldan) than that of diazinon and it refers to the velocity of breaking down from pesticide to the others in the same group. On the other hand, washing process leads to remarkable reduction in tested organophosphorus residues by different rates. The highest percentage loss after washing process was recorded with reldan (67.99%), while the lower was recorded with diazinon (51.28%). This finding is in agreement with the results obtained by Kamil (1993) and Zidan *et al.* (1997).

Processing tomato paste caused an increase in the percentage loss of all tested organophosphorus residues by different rates. Reldan and diazinon were completely eliminated due to processing tomatoes into paste. On the other hand, ketchup processes recorded a complete loss (100%) of reldan organophosphorus from tomato samples, while the lowest loss recorded with diazinon were 79.35%. In case of tomatoes samples after one week from spraying, the maximum percentage loss was recorded with diazinon (47.46%), while only 44.53% of reldan were losted. That's may be refer to the effect of heat in breaking the organophosphorus pesticides.

Table (1). Effect of preparation and processing on the percentage loss of organophosphorus residues in raw and processed tomatoes.

Treatments	Residues (ppm)		Percentage loss (%)	
	Reldan	Diazinon	Reldan	Diazinon
Control sample	N.D	N.D	N.D	N.D
Raw tomatoes (after one day of spraying) "unwashed"	5.378	3.976	0.0	0.0
After one week from spraying	2.983	1.678	44.533	57.796
Washed	1.721	1.937	67.999	51.282
Ketchup	N.D	0.821	100.000	79.351
Paste	N.D	N.D	100.000	100.000

Control sample: without spraying.
N.D.: Not detected.

1.2. Benzoyl urea residues in tomatoes:

The effect of preparation and processing on the percentage loss of benzoyl urea residues in tomatoes are shown in Table (2). From the obtained results it could be observed that the concentration of tested benzoyl urea residues in unwashed tomatoes samples were 3.891 and 2.781 ppm for Match and cascade, respectively. On the other hand, washing process remarkably reduced the benzoyl urea residues by 60.90 and 46.78% for match and cascade benzoyl urea, respectively.

It is worth to mention that processing of tomatoes into ketchup caused an increase in the percentage loss of all tested benzoyl urea residues by different rates. The highest percentage loss was recorded with cascade (100%). On the other hand, processing of tomato into paste recorded a complete loss (100%) of cascade while the percentage loss of match, being 47.03%. In case of tomato samples after one week of spraying, a complete loss was recorded with cascade (100%), while a lower percentage of loss recorded with match (60.92%). This may be due to that the post harvest interval (P.H.I) of cascade is short compared to that of Match.

Table (2). Effect of preparation and processing on the percentage loss of benzoyl urea residues in tomatoes.

Treatments	Residues (ppm)		Percentage loss (%)	
	Match	Cascade	Match	Cascade
Control sample	N.D	N.D	N.D	N.D
Raw tomatoes (after one day of spraying) "unwashed"	3.891	2.781	0.0	0.0
After one week from spraying	2.531	N.D	34.952	100.000
Washed	1.521	1.480	60.909	46.781
Ketchup	1.871	N.D	51.914	100.000
Paste	1.983	N.D	49.036	100.000

Control sample: without spraying.
N.D.: Not detected.

1.3. Miscellaneous pesticide residues in tomatoes:

Data given in Table (3) show the effect of preparation and processing on the percentage loss of the other different pesticide residues in tomatoes. The obtained results indicated that the concentration of actara, spintor, pride, kuik and vertimic residues in raw tomatoes samples were 8.391, ND, ND, 4.283 and ND ppm, respectively.

Table (3). Effect of preparation and processing on the percentage loss of miscellaneous pesticide residues in tomatoes.

Treatments	Residues (ppm)					Percentage loss (%)				
	Actara	Spintor	Pride	Kuik	Vertimic	Actara	Spintor	Pride	Kuik	Vertimic
Control sample	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D
Raw tomatoes (after one day of spraying) "unwashed"	8.391	N.D	N.D	4.283	N.D	0.0	N.D	N.D	0.0	N.D
After one week from spraying	6.001	N.D	N.D	0.984	N.D	28.482	N.D	N.D	77.025	N.D
Washed	8.212	N.D	N.D	1.831	N.D	2.133	N.D	N.D	57.249	N.D
Ketchup	4.200	N.D	N.D	0.652	N.D	49.946	N.D	N.D	84.777	N.D
Paste	3.801	N.D	N.D	0.421	N.D	54.701	N.D	N.D	90.170	N.D

Control sample: without spraying.
N.D.: Not detected.

On the other hand, washing process caused a clear reduction in the another tested pesticide residues. A complete percentage loss (100%) after washing process was recorded with spintor, pride and vertimic pesticide residues. While the lowest percentages of loss were recorded with actara pesticide residues (2.13%). This finding are in agreement with the results obtained by Abd El-Daim and Zidan (1996).

On the other hand, processing of tomatoes into ketchup recorded a complete loss (100%) of spintor, pride and vertimic, while the lowest loss in ketchup was recorded with actara pesticide (49.94%).

In case of tomato samples after one week of spraying, a complete loss was recorded with spintor, pride, vertimic and kuik (100%), mean while 28.48% loss was recorded for actara.

It's worth to mention that spintor, pride and vertimic are classified as Bioinsecticides because it's degredation time is very rabid and short. Therefore, it were not detected in the analysis.

2. Fungicide residues in tomatoes:

The effect of preparation and processing on the percentage loss of fungicide residues in tomatoes is shown in Table (4). The concentration of tested fungicide residues in raw tomato samples were 1.91, 2.87, 2.10 and ND ppm for teldor, previcure-N, milor-Co and idroram, respectively. On the other hand, washing process markedly reduced the tested fungicide residues by different rates. The maximum percentage loss after washing process was recorded with idroram (100%). While the lowest percentage of loss was recorded with previcure-N (2.45%). This finding are in agreement with the results obtained by Chavarri and Herrera (2005).

It is worth to mention that processing of tomatoes into paste increased the percentage loss of all tested pesticide residues by different rates. The highest percentage loss was recorded with idroram, milor-coand teldor (100%).

On the other hand, processing of tomatoes into ketchup caused a complete loss (100%) for teldor and idroram fungicide, while, the lowest loss was recorded with milor-co fungicide (90.48%) and that's may be refer to the interference between same fungicide and some heavy metals because idroram fungicide contain copper in its structure for this point, it was not detected in the analysis. In case of tomato samples after one week of spraying, a complete percentage loss was recorded with idroram fungicide (100%), while the lowest recorded with previcur N. fungicide (84.30%).

Table (4). Effect of preparation and processing on the percentage loss of fungicide residues in tomatoes.

Treatments	Residues (ppm)				Percentage loss (%)			
	Teldor	Previcure-N	Milor-Co.	Idroram	Teldor	Previcure-N	Milor-Co.	Idroram
Control sample	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D
Raw tomatoes (after one day of spraying) "unwashed"	1.918	2.879	2.103	N.D	0.0	0.0	0.0	N.D
After one week from spraying	0.208	0.452	0.784	N.D	89.155	84.300	62.719	N.D
Washed	0.783	2.531	1.985	N.D	59.176	12.087	5.611	N.D
Ketchup	N.D	1.352	0.200	N.D	100.000	53.039	90.489	N.D
Paste	N.D	1.121	N.D	N.D	100.000	61.062	100.000	N.D

Control sample: without spraying.
N.D.: Not detected.

3. Heavy metals residue in raw and processed tomatoes

3.1. Effect of different treatments on heavy metals residue in tomato (ppm) after one day from spraying fertilizers:

Data presented in Fig (1) show the effect of different treatments on heavy metals residue in tomato (ppm) after one day from spraying fertilizers.

The highest heavy metals residue were recorded with all fertilizers treatments than those of control samples. Values were 1241.90, 332.51, 78.3, 6.03 and 194.96 ppm for iron, zinc, manganese, lead and copper, respectively, while the lowest values were recorded with control samples. Values were 257.88, 223.95, 67.02, 0.44 and 104.52 ppm for iron, zinc, manganese, lead and copper, respectively. The obtained results refers to the structure of sprayed fertilizers which contain Fe, Zn, Mn and that explain the higher values of it than those in control "without spraying".

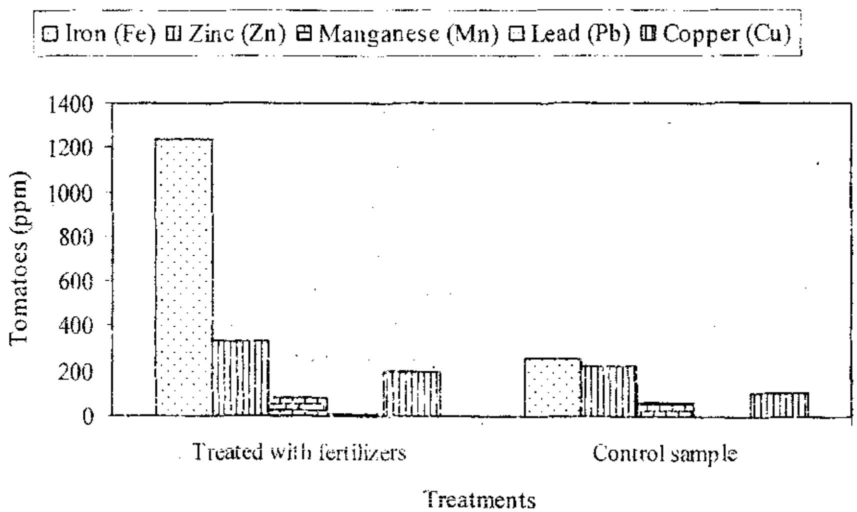


Fig. (1). Effect of treatments on heavy metals residue in tomatoes (ppm) after one day from spraying fertilizers.

3.2. Effect of different treatments on heavy metals residue in tomato (ppm) without spraying fertilizers:

Data in Table (5) shows the effect of different treatments on heavy metals residue in tomato (ppm) without spraying fertilizers.

It is obvious to notice that the highest iron as ppm was recorded with the raw tomatoes (925.92 ppm) while, the lowest was recorded with the washed sample (202.83 ppm). In case of zinc, manganese and copper, the obtained data indicated that the highest values of zinc, manganese and copper were recorded with the raw samples (299.41, 127.27 and 423.05 ppm, respectively). While the lowest values of zinc were noticed in ketchup which recorded 100.04 ppm. So, the lowest values of manganese were noticed in paste (26.28 ppm), and the lowest values of copper were in ketchup sample (19.29 ppm). On the other hand, the lead not detected in paste sample while the highest values were recorded in washed sample (2.43 ppm).

Table (5). Effect of treatments on heavy metals residue in tomatoes (ppm) without spraying fertilizers.

Treatments / Metals	Iron (Fe)	Zinc (Zn)	Manganese (Mn)	Lead (Pb)	Copper (Cu)
Control sample	257.88	223.950	67.020	0.440	104.520
Unwashed "Row tomatoes"	925.920	299.410	127.270	2.430	423.050
After one week from spray	240.000	230.010	48.025	2.080	84.091
Washed	202.830	250.960	72.670	3.840	61.490
Ketchup	588.250	100.040	37.710	2.620	19.290
Paste	384.250	100.600	26.280	0.000	45.355
Egyptian standard	–	–	–	0.300	5.000

Control sample: without spraying.

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تأثير عمليات الإعداد والتصنيع على بقايا بعض المبيدات والمعادن الثقيلة فى الطماطم

أبو الفتح عبد القادر البديوى[1] ، علاء الدين السيد البلتاجى[1] ،
عماد محمد عبد الحليم الخولى[2] ، ممدوح بسيونى عزازى[1]
[1] قسم علوم وتكنولوجيا الأغذية – كلية الزراعة – جامعة المنوفية
[2] قسم التغذية وعلوم الأطعمة – كلية الاقتصاد المنزلى – جامعة المنوفية

الملخص العربى :

يهدف هذا البحث إلى دراسة تأثير عمليات الإعداد والتصنيع على نسبة الفقد والتخلص من بقايا بعض المبيدات الحشرية والفطرية على الطماطم ، وعلى الجانب الآخر دراسة تأثير المعاملات المختلفة على بقايا بعض المعادن الثقيلة مثل الحديد والزنك والمنجنيز والرصاص والنحاس على الطماطم . ولقد أوضحت النتائج المتحصل عليها أن كل معاملات الإعداد والتصنيع لمنتجات الطماطم مثل الكاتشب والصلصة أدت وبشكلٍ ملحوظ على زيادة النسبة المئوية للفقد فى بقايا كل أنواع المبيدات المستخدمة فى هذا البحث سواء الحشرية أو الفطرية. فى حين أن بعض المعاملات التصنيعية أدت إلى فقد كامل لكل بقايا المبيدات الحشرية والفطرية على سبيل المثال :

١- كل المعاملات التصنيعية أدت إلى فقد كامل لمبيدات اسبنتور وبرايد وڤيريتمك .
٢- صناعة الصلصة أدت إلى فقد كامل لمبيد الريلدان والديازينون .
٣- صناعة الكاتشب أدت إلى فقد كامل لمبيد الريلدان والكاسكيد .

ومن الواضح أيضاً أن عملية الغسيل أدت إلى فقد أكثر من ٥٠% من معظم المبيدات الحشرية فى حين أن الحرارة العالية أدت إلى فقد ١٠٠% من بعض المبيدات وبنسبة تصل إلى ٧٠% فى البعض الآخر .

وعلى الجانب الآخر فإن المعاملات المختلفة باستخدام الأسمدة أدت إلى زيادة قيم المعادن الثقيلة خاصةً (الحديد والزنك والمنجنيز) عن مثيلاتها فى العينة الكنترول .