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Behavior of pesticides in tomatoes during commercial and home preparation

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Abstract

Monitoring of pesticide residues in Egyptian tomatoes and its products was studied. The average contents of HCB, lindane, dieldrin, heptachlor epoxide and DDT derivatives were detected at levels 0.009, 0.003, 0.006, 0.008 and 0.083 mg/kg, respectively. On the other hand, the levels of dimethoate, profenofos and pirimiphos-methyl were 0.461, 0.206 and 0.114 mg/kg, respectively. In ketchup and paste samples, most organochlorine and organophosphorus pesticide residues were not detectable. The distribution patterns of pesticide residues within the cuticular and subcuticular tissues in tomatoes were also studied. The skin samples were found to contain the highest levels of HCB, lindane, dieldrin and DDT derivatives. The investigation also indicated that washing with water and/or detergent solution were necessary to decrease the intake of pesticide residues. Freezing, as well as juicing and peeling, were necessary to remove pesticide residues in the skin. Cooking of tomatoes (including processing tomato to paste) helped to eliminate most pesticide residues from contaminated tomatoes.

1. Introduciton

Vegetables and fruits are susceptible to insect and disease attacks, so pesticides are widely used. Therefore, residues of pesticide could effect the ultimate consumers especially when freshly consumed. Many studies were carried out on pesticide residues in vegetables and fruits. It was reported that the main residues were HCB, lindane, heptachlor and its epoxide, DDT and its derivatives, as well as malathion, pirimiphos-methyl, profenofos and dimethoate (Abou-Arab, Soliman, Amra, & Naguib, 1995; Abou-El-Ghar, 1961; El-Lakwah, Hamed, Darwish, & Shams El-Din, 1995).

Tomato fruits are considered to be an important crop in Egypt. The average yield per feddan is relatively low under the Egyptian conditions and this may be due to many factors, including insect infestation (El-Nabarawy, Abou-Donia, & Amra, 1992).

The objectives of this study were to study: (1) the levels of pesticide residues in Egyptian tomatoes and its products; (2) distribution pattern of pesticide residues within the cuticular and subcuticular tissues of tomato fruits, and, (3) effect of washing procedures well as processing of tomatoes, on pesticide residues.

2. Materials and Methods

2.1. Samples

Fifty tomato fruit samples and 25 samples of each ketchup and paste were collected randomly from different regions in Great Cairo Governorate (Cairo, Giza and Kalubia). To study the distribution pattern of pesticide residues within the different tissues of the contaminated tomatoes, 25 samples each of skin, pulp, seeds, and juice were collected. Washing by acetic acid and sodium chloride at levels of 2, 4, 6, 8 and 10%, as well as tap water, freezing at −10°C (for 1, 3, 6, and 12 days), juicing, peeling, and home canning at 100°C for 30 min, were applied to tomato fruit samples polluted by HCB, lindane, p.p-DDT, dimethoate, profenofos and pirimiphos-methyl at levels about 1.0 mg/kg.

2.2. Standards

Pesticide standards of hexachlorobenzene (HCB), lindane, aldrin, dieldrin, heptachlor, heptachlor epoxide; chlordane, Endrin, 1,1,1-tri-chloro-2,2 bis (p-chlorophenyl) ethane (p.p- DDT); 1-(o-chlorophenyl)-1-(p-

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chlorophenyl)-2,2,2-tri-chloroethane (o. p-DDT), 1, 1-di-chloro-2,2-bis (p-chlorophenyl) ethylene (p. p-DDE), 1-(o-chlorophenyl)-1-(p-chlorophenyl)-2,2-dichloro-ethylene (o.p-DDE), 1,1-di-chloro -2,2 bis (p-chloro-phenyl) ethane (p.p-DDD), 1-(o-chlorophenyl)-1-(p-chloro-phenyl)-2,2-dichloroethane (o.p-DDD), malathion, parathion, parathion-methyl, dimethoate, profenofos, pirimiphos-methyl, and diazinon were purchased from Chem Service, Inc. (West Chester, PA).

2.3. Extraction and determination

Pesticide residues were extracted from different samples according to the methods of AOAC (1995) and the Pesticides Analytical Manual (1991). Aliquots of 1–2 µl of extract were injected into a Hewlett Packard gas chromatograph Model 5890 equipped with Ni^{63} electron capture detector (ECD), flame ionization detector (FID), and integrator 3392, fitted with HP-101 capillary column (Cross linked methyl silicon Gum), 30 m × 0.25 mm × 0.25 µm film thickness. The column oven temperature was programmed from 80 to 160°C at a rate of 3°C/min, then increased to 220°C at a rate of 5°C/min, and held for 20 min. Injection and detector temperature were 220 and 300°C, respectively.

3. Results and discussion

3.1. Monitoring of pesticide residues in tomatoes and its products

The content of pesticide residues in tomato samples and its products (ketchup and Paste) are summarized in Table 1. The average and maximum values found are given, as well as the percent of positive findings. Results indicated that organochlorine (HCB, lindane, aldrin, dieldrin, heptachlor and its epoxide, chlordane, endrin as well as DDT and its derivatives) and organophosphorus (malathion, parathion, parathion-methyl, dimethoate, profenofos, pirimiphos-methyl as well as diazinon) pesticides were present in tomatoes and its products. Organochlorine compounds are highly persistent insecticides, and have been used in Egypt for more than 20 years. Most of them have been banned, yet their residues still appear as pollutants in food as well as in the environment. Organophosphorus compounds are widely used in agriculture and animal production for the control of various insects. These compounds have higher acute toxicities than other chlorinated insecticides and they have the advantage of being more rapidly degraded in the environment.

Table 1
The contents (mg/kg) of chlorinated and phosphorus pesticide residues in tomato and its products collected from Great Cairo Governorates

Compound	Tomatoes				Ketchup				Paste			
	Mean ± SD	Range	Positive sample		Mean ± SD	Range	Positive sample		Mean ± SD	Range	Positive sample	
			No.	%			No.	%			No.	%
Organochlorines												
HCB	0.009 ± 0.004	0.0-0.28	12	24	–	–	–	–	0.008 ± 0.002	0.0–0.016	4	16
Lindane	0.003 ± 0.001	0.0–0.008	16	32	–	–	–	–	0.011 ± 0.008	0.0–0.014	2	8
Aldrin	–	–	–	–	–	–	–	–	–	–	–	–
Dieldrin	0.006 ± 0.003	0.0–0.009	6	12	–	–	–	–	–	–	–	–
Heptachlor	–	–	–	–	–	–	–	–	–	–	–	–
Hep.–epoxide	0.008 ± 0.003	0.0–0.18	6	12	–	–	–	–	–	–	–	–
Chlordane	–	–	–	–	–	–	–	–	–	–	–	–
Endrin	–	–	–	–	–	-	–	–	–	–	–	–
p.p–DDT	–	–	–	–	–	–	–	–	–	–	–	–
o.p–DDT	–	–	–	–	–	–	–	–	0.061 ± 0.041	0.0–0.088	4	16
p.p–DDE	–	–	–	–	–	–	–	–	–	–	–	–
o.p–DDE	0.006 ± 0.002	0.0–0.11	8	16	0.018 ± 0.006	0.0–0.026	12	48	0.034 ± 0.022	0.0–0.066	6	24
p.p–DDD	0.011 ± 0.008	0.0–0.26	12	24	0.026 ± 0.012	0.0–0.038	11	44	–	–	–	–
o.p–DDD	0.66 ± 0.014	0.0–0.98	9	18	0.048 ± 0.036	0.0–0.052	16	64	0.042 ± 0.020	0.0–0.050	4	16
Organophosphorus												
Malathion	–	–	–	–	–	–	–	–	–	–	–	–
Parathion	–	–	–	–	–	–	–	–	–	–	–	–
Para.–methyl	–	–	–	–	–	–	–	–	–	–	–	–
Dimethoate	0.461 ± 0.364	0.0–0.686	15	30	–	–	–	–	–	–	–	–
Profenofos	0.206 ± 0.116	0.0–0.308	18	36	–	–	–	–	–	–	–	–
Pirimiphos–methyl	0.114 ± 0.110	0.0–0.311	22	44	–	–	–	–		–	–	–
Diazinon	–	–	–	–	–	–	–	–	–	–	–	–

[a] – non detectable.

In tomatoes, the average contents of HCB, lindane, dieldrin, heptachlor epoxide and DDT derivatives were relatively low, i.e. 0.009, 0.003, 0.006, 0.008 and 0.083 mg/kg, respectively. However, aldrin, chlordane and endrin were not detectable. On the other hand, the levels of dimethoate (0.461 mg/kg), profenofos (0.206), and pirimiphos methyl (0.114) in tomatoes were higher than the organochlorine compounds, but were lower than the Maximum Residue Limits (MRLs) of FAO/WHO (1993). The findings reveal that the predominant pesticide residues in tomatoes were pirimiphos methyl (44%), followed by profenofos (36%), lindane (32%), dimethoate (30%), o-p-DDD and HCB, which recorded 24% for both. p.p-DDD, p.p-DDE, heptachlor epoxide and dieldrin were found in few samples, recording frequency distributions of 18, 16, 12 and 12%, respectively. This accords with reports by Abou-El-Ghar (1961), El-Lakwah et al. (1995) and Abou-Arab et al. (1995). They showed that most of tomato samples collected from Egypt contained lindane, DDT, aldrin, dieldrin, pirimiphos - methyl, parathion, and malathion at levels below the acceptable residue limits of FAO. Similar results were obtained by Kwon et al. (1984) and Goedick, Riebel, and Beitz (1978).

In various samples of ketchup, most of the organochlorine and organophosphorus pesticides under investigation were non-detectable with the exception of DDT derivatives, which detected at relatively low levels (Table 1). Data proved also that the predominant pesticide residues in ketchup samples occurred in 48% of samples for p.p-DDE, 44% for o.p-DDD, and 64% for p.p-DDD. In contrast, no residues of organophosphorus compounds were detected in any samples.

Paste samples in Table 1, had levels of DDT derivatives higher than those of HCB and lindane, but neither the average nor the maximum values exceeded the MRLs of FAO/WHO (1993). The positive samples were 16, 8; 16, 24 and 16% of the samples contaminated by HCB, lindane, p.p-DDT, p.p-DDE and o.p-DDD, respectively. There were no residues of organophosphorus compounds detected in any of the collected samples.

It is clear that most samples of tomato contained different types of pesticides at levels below the MRLs. Pollution by HCB was found to be comparatively minimal in the tested samples, which is probably due to the relatively small dose used. Besides, HCB is a by-product of various chlorination processes and combustion of industrial products, which might also be a means of entry into the environment (Nair & Pillai, 1989). Aldrin and dieldrin were expected appear in some samples, due to their long use in Egypt for agriculture. Aldrin is converted to dieldrin by an epoxidation process in biological systems (Rumsey & Bond, 1974) and therefor dieldrin is expected to be found in a relatively higher levels than aldrin. The reason for contamination by heptachlor epoxide despite the complete bay might be the transformation of chlordane (El-Mekkawi, 1994). There are decreased levels and low frequencies of DDT derivatives in the current study; probably this insecticide is still being applied despite the Ministry's recommendation. Continuous contamination with DDT might be due to its presence in low concentrations in the irrigation water coming mainly from the River Nile (El-Makkawi, 1994), which passes through other developing countries at, which organochlorine pesticides are still in use.

3.2. Distribution pattern of pesticide residues within the cuticular and subcuticular tissues of the contaminated tomatoes

The distribution patterns of pesticide residues within the cuticular and subcuticular tissues of the contaminated tomatoes are presented in Table 2. The skin samples were found to have the highest levels of organochlorine pesticides, i.e. HCB (0.016), lindane (0.011), dieldrin (0.008) as well as DDT derivatives (0.074 mg/kg). However, the other organochlorine compounds under investigation were not detectable. On the other hand, the organophosphorus pesticides were also detected. The highest mean value was dimethoate followed by profenofos and pirimiphos-methyl, which had 0.588, 0.211 and 0.148 mg/kg, respectively. These levels of pesticides are the extracuticular residue, which adheres to the waxy layer (this forms the true surface residue) and the cuticular residue, which is the residue embeded or dissolved in the waxy-layer.

It is known that some of the pesticides penetrate from the surface into the deep layers of plant tissues. Usually the pesticide is dissolved in the top wax-like layers and then moves to the inside. Since tomato fruits are covered with a waxy layer and have a very thin skin, it is thought that the pesticides might easily penetrate the fruits and accumulate inside the pulp. Table 2 shows that HCB, lindane, DDT derivatives, dimethoate and profenofos had penetrated to the pulp, which recorded 0.003, 0.006, 0.011, 0.084 and 0.066 mg/kg, respectively.

On the other hand Table 2 indicates that all tomato seeds are found free of organochlorine compounds. However, two organophosphorus pesticides (dimethoate and profenofos were detected with the same concentrations (0.009 mg/kg for both)

Juicy tomato fruits with a thin skin, might be less subject to pesticide penetration (Gunther & Blinn, 1955). This statement is mainly based on the fact that penetration depends on the stability of the insecticidal material in the lipoid-like or waxy-like layer which cover the cuticle. The more thick and oily the skin, the more likely is the penetration. The results obtained proved that juice samples under investigation did not contain any type of organochlorine pesticides. However, trace residues of organophosphorus pesticides, i.e. dimethoate (0.048) and pirimiphos-methyl (0.098 mg/kg) were recorded (Table 2).

Table 2
The distribution pattern of pesticide residues within the cuticular and subcuticular tissues of the contaminated tomatoes

Compound	The levels of pesticide residues (mg/kg ± SD)			
	Skin	Pulp	Seeds	Juice
Organochlorines				
HCB	0.016 ± 0.008	0.003 ± 0.001	–	–
Lindane	0.011 ± 0.009	0.006 ± 0.003	–	–
Aldrin	–	–	–	–
Dieldrin	0.008 ± 0.003	–	–	–
Heptachlor	–	–	–	–
Hep.-epoxide	–	–	–	–
Chlordane	–	–	–	–
Endrin	–	–	–	–
p.p-DDT	–	–	–	–
o.p-DDT	–	–	–	–
p.p-DDE	–	–	–	–
o.p-DDE	0.008–0.004	–	–	–
p.p-DDD	0.018 ± 0.009	0.003 ± 0.001	–	–
o.p-DDD	0.048 ± 0.034	0.008 ± 0.006	–	–
Organophosphorus				
Malathion	–	–	–	–
Parathion	–	–	–	–
Parathion-methyl	–		–	–
Dimethoate	0.588 ± 0.286	0.084 ± 0.022	0.009 ± 0.003	0.048 ± 0.036
Profenofos	0.211 ± 0.146	0.066 ± 0.044	0.009 ± 0.003	0.098 ± 0.048
Pirimiphos-methyl	0.148 ± 0.120	–	–	–
Diazinon	–	–	–	–

[a] – non detectable.

Residue data for the waste solids resulting from the peeling and juicing operations, confirm the presence of residue in the skins and pulp residue from the juice extractor which do not form part of the final product (Powell, Stevens, & McCully, 1970).

3.3. Effect of washing procedures and processing of tomatoes on pesticide residues

Data concerning the effect of commercial processing on the residues of studied pesticides in tomatoes are presented in Tables 3 and 4. Examination of the obtained results indicates the important role of washing procedures, freezing, juicing, peeling, as well as pasting in removal of pesticide residues from the contaminated tomatoes.

Tables 3 and 4 indicate the efficient role of the washing process (tap water as well as different levels of both sodium chloride and acetic acid solutions) in the elimination of organochlorine (HCB, lindane, and DDT) and organophosphorus (dimethoate, profenofos, and pirimiphos-methyl) pesticides from contaminated tomatoes. Data proved that washing with different levels of acetic acid solution gave 51.3, 47.0, 33.7, 91.5, 86.0 and 93.7% loss in HCB, lindane, p.p-DDT, dimethoate, profenofos and pirimiphos-methyl, respectively. Sodium chloride washing came next in importance to washing by acetic acid solutions, giving 42.9, 46.1, 27.2, 90.8, 82.4 and 91.4% loss in the same pesticides, respectively (at 10% NaCl). The trends of the data indicated that the loss of different pesticides under investigation depends on the levels of acetic acid and NaCl solutions (2, 4, 6, 8, 10%). On the other hand, washing by tap water proved the least effective, showing 9.62, 15.3, 9.17, 18.8, 22.7 and 16.2% loss of HCB, lindane, p.p-DDT, dimethoate, profenofos, and pirimiphos-methyl, respectively. These results agree with those obtained by Fahey, Could, and Nelson (1969); Powell et al. (1970), Mesallaw and Moharran (1980), Rao, Subbiah, and Murthy (1984), Chirila and Florll (1985), Barrow, Esack, and Kalloo (1987), Gangwar and Kumar (1988), Sarode and Adule (1989), El-Nabarawy et al. (1992), Ismail, Ali, and Habiba (1993) and Abou-Arab et al. (1995). They reported that partial removal was effected by the washing operation.

In general, it is clear that washing with water and/or detergent to remove most of the surface residues is adviseable before using tomatoes.

Contaminated tomato fruits stored at −10°C showed no significant decreases in pesticides after one day (Tables 3 and 4). After three days of freezing, HCB, lindane, p.p-DDT, dimethoate, profenofos and pirimiphos-methyl decreased by 4.91, 6.32, 4.07, 13.0, 11.5 and 9.35%, respectively. The results obtained indicated that pesticide residues in tomatoes were reduced by increasing the time of freezing. Significant decreases in

Table 3
The role of washing process, freezing, juicing, peeling and home-canning in decreasing organochlorine pesticides from contaminated tomato fruits

Treatments	HCB		Lindane		p.p-DDT	
	Mean (mg/kg)	Reduction(%)	Mean (mg/kg)	Reduction(%)	Mean (mg/kg)	Reduction(%)
Contaminated samples washing	1.06	–	1.14	–	1.08	–
2% acetic acid solution	0.938	11.5	0.948	18.6	0.956	11.5
4% acetic acid solution	0.894	15.7	0.905	20.6	0.911	15.7
6% acetic acid solution	0.806	24.0	0.811	28.9	0.866	19.8
8% acetic acid solution	0.684	35.5	0.711	37.6	0.801	25.8
10% acetic acid solution	0.516	51.3	0.604	47.0	0.716	33.7
2% NaCl solution	0.948	10.6	0.968	17.7	0.964	10.7
4% NaCl solution	0.905	14.6	0.914	19.8	0.948	12.2
6% NaCl solution	0.806	24.2	0.811	28.2	0.896	17.0
8% NaCl solution	0.704	33.6	0.719	36.9	0.848	21.5
10% NaCl solution	0.605	42.9	0.614	46.1	0.786	27.2
Tap water	0.958	9.62	0.966	15.3	0.981	9.17
Freezing at $-10^{\circ}C$*:*						
for one day	1.04	1.98	1.10	3.68	1.06	1.76
for three days	1.008	4.91	1.07	6.32	1.04	4.07
for 6 days	1.004	5.28	1.06	7.02	1.018	5.74
for 12 days	0.948	10.6	0.954	16.3	0.94	13.0
Juicing	0.244	77.0	0.311	72.7	0.242	77.6
Peeling	0.206	80.6	0.201	82.4	0.203	81.2
Paste (home-canning)	0.615	42.0	0.622	45.4	0.748	30.7

Table 4
The role of washing process, freezing, juicing, peeling, and home-canning in decreasing organophosphorus pesticides from contaminated tomato fruits

Treatments	Dimethoate		Profenofos		Pirimiphos-methyl	
	Mean (mg/kg)	Reduction(%)	Mean (mg/kg)	Reduction(%)	Mean (mg/kg)	Reduction(%)
Contaminated samples washing	1.11	–	1.182	–	1.091	–
2% acetic acid solution	0.884	20.4	0.904	23.4	0.702	35.7
4% acetic acid solution	0.664	40.2	0.708	40.1	0.646	40.8
6% acetic acid solution	0.316	71.5	0.464	60.7	0.308	71.8
8% acetic acid solution	0.116	89.6	0.214	81.9	0.108	90.1
10% acetic acid solution	0.094	91.5	0.166	86.8	0.069	93.7
2% NaCl solution	0.714	35.7	0.864	27.0	0.703	35.6
4% NaCl solution	0.696	37.3	0.709	40.0	0.664	39.1
6% NaCl solution	0.501	54.9	0.614	48.1	0.491	55
8% NaCl solution	0.311	72.0	0.412	65.1	0.306	72.0
10% NaCl solution	0.102	90.8	0.208	82.4	0.094	91.4
Tap water	0.901	18.8	0.914	22.7	0.914	16.2
Freezing at $-10^{\circ}C$						
for one day	1.08	2.61	1.16	1.52	1.06	2.47
for three days	0.966	13.0	1.05	11.5	0.989	9.35
for 6 days	0.794	28.5	0.868	26.6	0.805	26.2
for 12 days	0.748	32.6	0.849	28.2	0.749	31.4
Juicing	0.294	73.5	0.308	73.9	0.288	73.6
Peeling	0.167	85.0	0.198	83.3	0.118	89.2
Paste (home-canning)	0.301	72.9	0.218	81.6	0.318	71.0

the contents were recorded after 6 and 12 days. The reduction of residues were 5.28, 7.02, 5.74, 28.5, 26.6 and 26.2% after 6 days and 10.6, 16.3, 13.0, 32.6, 28.2 and 31.4% loss after 12 days with HCB, lindine, p.p-DDT, dimethoate, profenofos and pirimiphos-methyl, respectively. These results *coincide* with those recorded by Farrow et al. (1968), who showed no significant decreases in DDT of contaminated tomato when stored at 55°F. However, an apparent malathion decrease of 30% was noted after 7 days' storage.

It can be concluded that freezing reduced pesticide residues.

The juicing operation data in Tables 3 and 4 clearly indicate the great role of the skins in adsorbing residues of pesticide under investigation. The reduction of pesticide residues ranged from 72.7 to 77.6% on juicing. Similar results obtained by Farrow et al. (1968), Powell et al. (1970) and Sarode and Adule (1989). They reported that tomato juicing removed most of pesticides, but trace amounts of DDT and malathion were found. Data clearly indicate that the peeling process had a significant effect on the elimination of pesticide residues from tomato fruits. In general, a decrease in the concentrations of pesticides under investigation was observed. The efficient role of the peeling process in elimination of pesticide residues was more clear; loss range from 80.6 to 89.2%. These results confirm the presence of residues in the skin Tables 3 and 4. They accord with those obtained by Powell et al. (1970) and Chirila and Florll (1985), who reported that pesticide residues were removed by peeling.

Home-canning at 100°C for 30 min diminished the pesticide residues in tomatoes. Data presented in Tables 3 and 4, show a significant decrease in the levels of pesticides under study. Results clearly indicate that organophosphorus pesticides were decreased more than the organochlorine compounds. The levels of reduction were 71.0 to 81.6%; however, the levels of reduction of organochlorine pesticides ranged only from 30.7 to 45.4%. This result may be due to the high stabilities of organochlorine pesticides to heat treatments. These findings clearly indicate that heat treatments had a significant effect on the decomposition of pesticides. Farrow et al. (1968), Fahey et al. (1969), Sarode and Adule (1989), El-Nabarawy et al. (1992) and Ismail et al. (1993), also reported that home-canning removed most of the organophosphorus pesticide residues. However, organochlorine compounds were not completely destroyed by paste processing. So, cooking of tomatoes helped to eliminate pesticide residues.

From the above results, it is clear that the advantages of the application of pesticides in agriculture in producing better crops must be weighed against the possible health hazard arising from the toxic pesticide residues in food. Pesticides should be applied correctly according to good agricultural practice, using only the required amounts. Washing with water and/or detergent solution for kitchen use were necessary to decrease the intake of pesticide residues. Freezing as well as juicing and peeling are necessary to remove the pesticide residues in the skins. Cooking of tomatoes (such as processing tomato to paste) helps to eliminate most of the pesticide residues.

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