

# Monitoring of Pesticide Residues in Egyptian Fruits and Vegetables During 1996

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Samples of the most common fruits and vegetables were collected from 8 local markets in 6 governorates. These 1579 samples were analyzed for residues of 53 pesticides, which included organophosphorus and organonitrogen compounds and some synthetic pyrethroids. Samples were also analyzed for residues of organochlorine pesticides, although they had been prohibited from use several years ago. Only 510 of the 1579 samples were analyzed for dithiocarbamate pesticide residues, which were determined as CS<sub>2</sub>. Overall, 76.1% of the total analyzed samples had no detectable residues, 23.9% contained detectable residues, and 2.59% contained residues that exceeded maximum residue limits. For individual crops, contaminated samples ranged from 0 to 96% of the number of samples analyzed. However, the highest violative percentage for samples of individual crops was 12.5. Chlorpyrifos, carbaryl, dimethoate, bromopropylate, and profenofos were the violative pesticides determined in fruit and vegetable samples. The results of the current study demonstrated that no restricted or banned pesticides such as DDT, HCH, and their isomers were found in any of the samples analyzed. Dithiocarbamate residues were detected in 9.4% of the 510 samples analyzed, with a violative percentage of 0.39, representing one grape sample and one peach sample.

Food contamination monitoring is an essential component of ensuring the safety of the food supply and managing health and environmental resources. It provides information on the levels and sources of contamination in foods, the amounts of contaminants ingested by humans, and contamination levels. A series of measures of good agricultural practice including optimum dosage, number of applications, and maximum intervals between application and harvest can be used to keep residue levels as low as possible. Implementation of these measures will ensure that pesticides are applied as safely as possible.

A monitoring program is an essential tool to ensure that the safety criteria are met, and it is also the key to evaluating the extent of risk through calculation of dietary intake and, consequently, to assessing the risk of different chemicals. Previous studies (1–6) have reported the levels of pesticide residues in Egyptian food and the environment.

This study reports results of the ongoing monitoring program for fruits and vegetables in order to assess the level of pesticide contamination in Egypt during 1996; such work provides useful information for the decision-makers who determine the agricultural and environmental policies of the country.

## Experimental

### Apparatus

(a) *Gas chromatographs*.—(1) Hewlett-Packard Model 5890 equipped with a double electron capture detector with 2 capillary columns, an injector at 225°C, and a detector at 300°C. Operating conditions: nitrogen carrier gas, 2.5 mL/min; carrier and makeup gas, 75–90 mL/min; and column head pressure, 82 kPa. (2) Hewlett-Packard Model 5890 equipped with a double nitrogen–phosphorus detector, an injector at 225°C, and a detector at 280°C. Operating conditions: hydrogen, 3.5 ± 0.1 mL/min; air, 100–200 mL/min; and nitrogen carrier gas, 25 mL/min.

(b) *UV spectrophotometer*.—Double-beam Unicam SP 1800 (UK).

(c) *Chromatography columns*.—Used with either gas chromatograph. (1) PAS-5 tested Ultra 2 Silicon, 25 m × 0.32 mm, and 0.52 μm film thickness. (2) PAS-1701 tested 1701 Silicon, 25 m × 0.32 mm, 0.25 μm film thickness. Temperature programs for both: initial temperature, 90°C for 2 min; increase at 20°C/min to 150°C; increase at 6°C/min to 270°C; hold for 15 min.

### Reagents

(a) *Solvents*.—Acetone, dichloromethane, *n*-hexane, and petroleum ether (Pestiscan chromatography grade or similar quality); ethanol, 95–96%; diethanolamine, 98%; HCl; toluene; and carbon disulfide.

(b) *Chemicals*.—Anhydrous sodium sulfate (Riedel-de-Haen, Germany); NaCl; NaOH; copper(II) acetate monohydrate, 98%; and tin(II) chloride dihydrate.

(c) *Pesticide reference standards*.—(1) *Organophosphorus and nitrogen-containing compounds*.—Atrazine, bendiocarb,

**Table 1. Results of analyses of 1579 fruit and vegetable samples collected from the Egyptian local markets during 1996**

Commodity	Total No. of samples analyzed	Pesticides found	No. of samples contaminated with each pesticide	Mean, mg/kg	Contaminated samples of each commodity		MRL, mg/kg <sup>a</sup>	Violative samples		Total violative samples for commodity	
					No.	%		No.	%	No.	%
Leafy vegetables											
Cabbage	7	Chlorpyrifos	1	0.57 <sup>b</sup>	2	28.6	0.05	1	14.3	2	28.6
		Profenofos	2	5.0			1	1	14.3		
Grape leaf	24	Dimethoate	1	8.6 <sup>b</sup>	2	8.3	1 EU	1	4.17	1	4.2
		Dithiocarbamates (5) <sup>c</sup>	1	0.70 <sup>b</sup>			1 F	—	—		
		Omethoate	1	0.78 <sup>b</sup>			0.2 EU	1	4.17		
		Pirimiphos-methyl	1	0.02 <sup>b</sup>			0.5 F	—	—		
Lettuce	38	Chlorothalonil	1	0.05 <sup>b</sup>	3	7.9	1 F	—	—	—	—
		Dimethoate	2	0.94			2	—	—		
		Dithiocarbamates (35) <sup>c</sup>	1	0.26 <sup>b</sup>			5	—	—		
Melokhia	35	Dimethoate	1	0.04 <sup>b</sup>	2	5.7	1 EU	—	—	—	—
		Malathion	1	0.02 <sup>b</sup>			0.5 EU	—	—		
		Profenofos	1	0.02 <sup>b</sup>			0.5 F	—	—		
Dry melokhia	18	Carbaryl	1	1.4 <sup>b</sup>	4	22.2	10	—	—	—	—
		Malathion	1	0.22 <sup>b</sup>			3 EU	—	—		
		Pirimicarb	1	0.15 <sup>b</sup>			0.5 F	—	—		
		Profenofos	1	0.20 <sup>b</sup>			0.5 F	—	—	4	12.5
Spinach	32	Bromopropylate	1	0.17 <sup>b</sup>	4	12.5	1	—	—		
		Chlorpyrifos-methyl	1	0.13 <sup>b</sup>			0.1 F	1	3.13		
		Dimethoate	3	2.12			1	2	6.25		
		Omethoate	2	0.44			0.1	2	6.25		
Watercress	44	Dimethoate	3	0.42	3	6.8	1	—	—	—	
Total for leafy vegetables					20	10.10				7	3.53
Vegetables											
Artichoke	30	—	—	—	—	—	—	—	—	—	—
Broad bean	9	Dicofol	1	0.11 <sup>b</sup>	1	11.1	5	—	—	—	—
Cauliflower	5	—	—	—	—	—	—	—	—	—	—
Cantaloupe	32	Bromopropylate	1	0.11 <sup>b</sup>	10	31.3	0.5	—	—	—	—
		Dicofol	8	0.12			5	—	—		

Table 1. (continued)

Commodity	Total No. of samples analyzed	Pesticides found	No. of samples contaminated with each pesticide	Contaminated samples of each commodity		MRL, mg/kg <sup>a</sup>	Violative samples		Total violative samples for commodity		
				Mean, mg/kg	No.		%	No.	%	No.	%
Cucumber	94	Dimethoate	2	0.07		1 EU	—	—	—	—	
		Dithiocarbamates (22) <sup>c</sup>	1	0.21 <sup>b</sup>		1	—	—	—	—	
		Tetradifon	1	0.08 <sup>b</sup>		0.5 F	—	—	—	—	
		Bromopropylate	7	0.12	29	30.9	0.5	—	—	2	2.12
		Chlorothalonil	3	0.19		5	—	—	—	—	
		Dicofol	13 <sup>e</sup>	0.22		0.5	2	2.12	—	—	
		Dimethoate	5	0.31		1 EU	—	—	—	—	
		Dithiocarbamates (67) <sup>c</sup>	7	0.22		0.5	—	—	—	—	
		Fenitrothion	1	0.03 <sup>b</sup>		0.05	—	—	—	—	
		Fenvalerate	1	0.14 <sup>b</sup>		0.2	—	—	—	—	
		Iprodione	1	0.16 <sup>b</sup>		2	—	—	—	—	
		Metaxyl	2	0.18		0.5	—	—	—	—	
		Pirimiphos-methyl	2	0.07		1	—	—	—	—	
Eggplant	64	Procymidone	5	0.31		2	—	—	—	—	
		Tetradifon	4	0.16		5	—	—	—	—	
		Bromopropylate	1	0.07 <sup>b</sup>	5	7.8	1	—	—	—	
		Dimethoate	1	0.40 <sup>b</sup>		1 EU	—	—	—	—	
		Dithiocarbamates (58) <sup>c</sup>	1	0.26 <sup>b</sup>		1 F	—	—	—	—	
		Omethoate	1	0.20 <sup>b</sup>		0.2 EU	—	—	—	—	
		Profenofos	2	0.05		0.5 F	—	—	—	—	
		Chlorpyrifos	1	0.84 <sup>b</sup>	41	25.5	0.2	1	0.62	4	2.48
		Chlorothalonil	2	0.07		5	—	—	—	—	
		Dicofol	20 <sup>e</sup>	0.19		2	—	—	—	—	
		Dimethoate	13 <sup>e</sup>	0.14		2	—	—	—	—	
		Dithiocarbamates (30) <sup>c</sup>	2	0.23		0.5	—	—	—	—	
		Omethoate	2	0.16		0.2	1	0.62	—	—	
Green beans	161	Pirimiphos-methyl	1	0.03 <sup>b</sup>		0.5	—	—	—	—	
		Profenofos	2	0.54		0.1	2	1.24	—	—	
		Tetradifon	2	0.30		0.5 F	1	—	—	—	

Table 1. (continued)

Commodity	Total No. of samples analyzed	Pesticides found	No. of samples contaminated with each pesticide	Contaminated samples of each commodity		MRL, mg/kg <sup>a</sup>	Violative samples		Total violative samples for commodity		
				Mean, mg/kg	No.		%	No.	%	No.	%
Green peas	71	Chlorpyrifos	1	0.02 <sup>b</sup>	9	0.1 F	—	—	2	2.8	
		Chlorothalonil	1	0.28 <sup>b</sup>	—	—	1 F	—	—	—	
		Dimethoate	5	0.16	—	—	0.5	2	2.82	—	
		Dithiocarbamates (24) <sup>c</sup>	1	0.27 <sup>b</sup>	—	—	1 EU	—	—	—	
		Omethoate	1	0.42 <sup>b</sup>	—	—	0.1	1	1.4	—	
		Tetradifon	1	0.05 <sup>b</sup>	—	—	0.5 F	—	—	—	
		Triadimefon	1	0.08 <sup>b</sup>	—	—	0.1	—	—	—	
		Dicofol	1	0.05 <sup>b</sup>	2	8.3	5	—	—	—	
		Profenofos	1	0.08 <sup>b</sup>	—	—	0.5 F	—	—	—	
		Profenofos	2	0.25	3	6.9	0.2	1	2.3	1	2.3
Okra	24	Vinclozolin	1	0.10 <sup>b</sup>	—	1	—	—	—	—	
		Bromopropylate	3	0.13	27	29.7	1	—	—	5	5.5
		Chlorpyrifos	1	0.06 <sup>b</sup>	—	—	0.5	—	—	—	
		Chlorpyrifos-methyl	1	0.68 <sup>b</sup>	—	—	0.5	1	1.1	—	
		Chlorothalonil	1	0.92 <sup>b</sup>	—	—	1 F	—	—	—	
		Dicofol	7	0.32	—	—	1	—	—	—	
		Dimethoate	7	0.36	—	—	1	1	1.1	—	
		Dithiocarbamates (62) <sup>c</sup>	10 <sup>e</sup>	0.39	—	—	1	—	—	—	
		Fenitrothion	1	0.24 <sup>b</sup>	—	—	0.1	1	1.1	—	
		Iprodione	1	0.43 <sup>b</sup>	—	—	5	—	—	—	
Squash	56	Pirimiphos-methyl	7	0.39	—	1	2	2.2	—	—	
		Procymidone	2	0.10	—	—	5	—	—	—	
		Profenofos	2	1.90	—	—	0.5	1	1.1	—	
		Tetradifon	4	0.16	—	—	0.5 F	—	—	—	
		Dimethoate	2	0.19	3	5.4	1 EU	—	—	—	
		Vinclozolin	1	0.30 <sup>b</sup>	—	—	2 F	—	—	—	
		Bromopropylate	6	0.17	37	43	1	—	—	3	3.49
		Chlorpyrifos	1	0.10 <sup>b</sup>	—	—	0.5	—	—	—	
		Chlorpyrifos-methyl	4	0.10	—	—	0.5	—	—	—	
		Chlorothalonil	6	0.35	—	—	5	—	—	—	
Tomato	86	Chlorpyrifos	1	0.10	—	—	—	—	—	—	
		Chlorpyrifos-methyl	4	0.10	—	—	0.5	—	—	—	
		Chlorothalonil	6	0.35	—	—	5	—	—	—	

Table 1. (continued)

Commodity	Total No. of samples analyzed	Pesticides found	No. of samples contaminated with each pesticide	Contaminated samples of each commodity		MRL, mg/kg <sup>a</sup>	Violative samples		Total violative samples for commodity	
				Mean, mg/kg	No.		%	No.	%	No.
Total for vegetables	766	Cypermethrin	8	0.28	1	0.5	1	1.2	17	2.22
		Dicofol	12 <sup>e</sup>	0.58	1	1	1.2			
		Dithiocarbamates (62) <sup>c</sup>	3	0.38	—	—	—			
		Iprodione	1	0.06 <sup>b</sup>	—	—	—			
		Profenofos	6	0.31	—	—	—			
		Tetradifon	2	2.10	1	1.2	—			
Root vegetables										
Carrot	42	Chlorpyrifos	1	0.12 <sup>b</sup>	2	0.5	—	—	—	—
		Pirimiphos-methyl	1	0.08 <sup>b</sup>	—	—	—			
Sweet potato	42	Dicofol	1	0.05 <sup>b</sup>	1	2.4	—	—	—	—
Taro	44	—	—	—	—	— <sup>d</sup>	—	—	—	—
Total for root vegetables	128	—	—	—	3	2.34	—	—	—	—
Fruits										
Apple	49	Bromopropylate	6	0.27	18	36.7	2	—	2	4.1
		Chlorpyrifos	2	0.12	—	—	—			
		Cypermethrin	1	0.14 <sup>b</sup>	—	—	—			
		Dicofol	7	0.44	—	—	—			
		Dimethoate	4	1.10	2	4.1	—			
		Dithiocarbamates (41) <sup>c</sup>	6	0.20	3	—	—			
Apricot	5	Malathion	1	0.05 <sup>b</sup>	—	—	—	—	—	—
		Phosalone	1	0.07 <sup>b</sup>	—	—	—			
		Tetradifon	2	0.12	—	—	—			
		Cypermethrin	1	0.57 <sup>b</sup>	3	60	1			
		Dicofol	1	0.60 <sup>b</sup>	—	—	—			
		Dimethoate	2	0.85	—	—	—			
Profenofos	2	0.12	—	—	—					

Table 1. (continued)

Commodity	Total No. of samples analyzed	Pesticides found	No. of samples contaminated with each pesticide	Contaminated samples of each commodity		MRL, mg/kg <sup>a</sup>	Violative samples		Total violative samples for commodity	
				Mean, mg/kg	No.		%	No.	%	No.
Banana	36	Chlorothalonil	1	0.03 <sup>b</sup>	1	0.2	—	—	—	—
Dates	54	Carbaryl	1	5.00 <sup>b</sup>	10	3 EU	1	1.85	5	9.3
		Cypermethrin	4	0.37		0.5	4	7.4		
		Dicofol	1	0.28 <sup>b</sup>		5	—	—		
		Malathion	1	0.07 <sup>b</sup>		0.5 EU	—	—		
		Procymidone	1	0.48 <sup>b</sup>		2 F	—	—		
Fig	11	Dicofol	3	0.16	3	5	—	—	—	—
Grape	47	Bromopropylate	2	0.62	23	5	—	—	5	10.6
		Cypermethrin	3	0.46		0.5	2	4.3		
		Dicofol	6	0.09		5	—	—		
		Dimethoate	4	0.19		1	—	—		
		Dithiocarbamates (25) <sup>c</sup>	4	0.49		1	1	2.1		
		Iprodione	1	0.85 <sup>b</sup>		10	—	—		
		Malathion	1	0.32 <sup>b</sup>		8	—	—		
		Metalaxyl	1	0.20 <sup>b</sup>		1	—	—		
		Procymidone	3	0.49		5	—	—		
		Profenofos	3	0.43		0.5 F	2	4.3		
		Prothiofos	3	0.65		1 F	—	—		
Guava	72	Cypermethrin	1	0.12 <sup>b</sup>	21	— <sup>d</sup>	—	—	—	—
		Dicofol	6	0.25		5	—	—		
		Dimethoate	6	0.12		1 EU	—	—		
		Fenitrothion	1	0.04 <sup>b</sup>		0.5 F	—	—		
		Malathion	9	0.07		0.5 EU	—	—		
		Tetradifon	1	0.08 <sup>b</sup>		0.5 F	—	—		
Lemon lime	20	Chlorpyrifos-methyl	1	0.08 <sup>b</sup>	7	0.5	—	—	—	—
		Malathion	3	0.12		4	—	—		
		Profenofos	3	0.43		1	—	—		
Mango	18	Dicofol	1	0.07 <sup>b</sup>	2	5	—	—	—	—
		Dimethoate	1	0.10 <sup>b</sup>		1 EU	—	—		

Table 1. (continued)

Commodity	Total No. of samples analyzed	Pesticides found	No. of samples contaminated with each pesticide	Contaminated samples of each commodity			MRL, mg/kg <sup>a</sup>	Violative samples		Total violative samples for commodity			
				Mean, mg/kg	No.	%		No.	%	No.	%		
Orange	64	Bromopropylate	1	0.15 <sup>b</sup>	26	40.6	5	—	—	—	—		
		Cypermethrin	1	0.05 <sup>b</sup>	—	—	2	—	—	—	—		
		Dicofol	3	0.16	—	—	5	—	—	—	—		
		Dimethoate	12 <sup>e</sup>	0.24	—	—	2	—	—	—	—		
		Malathion	3	0.11	—	—	4	—	—	—	—		
		Bromopropylate	1	0.10 <sup>b</sup>	27	96.4	1 F	—	—	—	2	7.1	
		Chlorothalonil	2	0.25	—	—	25	—	—	—	—	—	
		Dicofol	4	0.78	—	—	5	—	—	—	—	—	
		Dimethoate	11 <sup>e</sup>	0.74	—	—	2	—	—	—	2	7.1	
		Dithiocarbamates (16) <sup>c</sup>	5	0.56	—	—	1	—	—	—	1	6.3	
Pear	3	Omethoate	1	0.44 <sup>b</sup>	—	—	2	—	—	—	—		
		Procymidone	2	0.07	—	—	10	—	—	—	—		
		Prothiofos	1	0.39 <sup>b</sup>	—	—	0.5 F	—	—	—	—		
		Tetradifon	1	0.35 <sup>b</sup>	—	—	0.5 F	—	—	—	—		
		Dicofol	1	0.10 <sup>b</sup>	2	67	5	—	—	—	—		
		Dithiocarbamates (3) <sup>c</sup>	1	0.20 <sup>b</sup>	—	—	3	—	—	—	—		
		Dimethoate	2	0.21	—	—	1	—	—	—	—		
		Bromopropylate	2	0.22	4	28.6	2	—	—	—	—		
		Dicofol	1	0.25 <sup>b</sup>	—	—	1	—	—	—	—		
		Dimethoate	1	0.09 <sup>b</sup>	—	—	0.5	—	—	—	—		
Pomegranate	28	Dimethoate	8	0.13	13	46.4	1 EU	—	—	—	1	3.6	
		Dicofol	6	0.15	—	—	5	—	—	—	—	—	
		Malathion	3	0.03	—	—	0.5 EU	—	—	—	—	—	
		Omethoate	1	2.80 <sup>b</sup>	—	—	0.5 EU	—	—	—	—	—	
		Bromopropylate	1	0.89 <sup>b</sup>	28	73.4	2	—	—	—	—	2	5.26
		Carbaryl	1	0.60 <sup>b</sup>	—	—	7	—	—	—	—	—	
		Chlorothalonil	2	1.50	—	—	1 F	—	—	—	—	—	
		Cypermethrin	2	0.35	—	—	0.5	—	—	—	—	—	
		Dicofol	21 <sup>e</sup>	0.82	—	—	5	—	—	—	—	—	
		Dimethoate	2	0.53	—	—	1	—	—	—	—	—	
Strawberry	38	Bromopropylate	1	0.89 <sup>b</sup>	28	73.4	2	—	—	—	—		
		Carbaryl	1	0.60 <sup>b</sup>	—	—	7	—	—	—	—		
		Chlorothalonil	2	1.50	—	—	1 F	—	—	—	—		
		Cypermethrin	2	0.35	—	—	0.5	—	—	—	—		
		Dicofol	21 <sup>e</sup>	0.82	—	—	5	—	—	—	—		
		Dimethoate	2	0.53	—	—	1	—	—	—	—		
		Bromopropylate	1	0.89 <sup>b</sup>	28	73.4	2	—	—	—	—		
		Carbaryl	1	0.60 <sup>b</sup>	—	—	7	—	—	—	—		
		Chlorothalonil	2	1.50	—	—	1 F	—	—	—	—		
		Cypermethrin	2	0.35	—	—	0.5	—	—	—	—		

Table 1. (continued)

Commodity	Total No. of samples analyzed	Pesticides found	No. of samples contaminated with each pesticide	Contaminated samples of each commodity		Violative samples		Total violative samples for commodity		
				Mean, mg/kg	No.	%	MRL, mg/kg <sup>a</sup>	No.	%	No.
	487	Dithiocarbamates(22) <sup>c</sup>	5	0.67	188	38.6	3	—	17	3.49
		Iprodione	3	0.41			10	—		
		Pirimiphos-methyl	3	0.09			1	—		
		Procymidone	6	0.24			5	—		
		Tetradifon	6	0.08			0.5 F	—		
		Vinclozolin	1	0.87 <sup>b</sup>			10	—		
Total for fruits	487				188	38.6			17	3.49
Total No.	1579				378	23.9			41	2.59

<sup>a</sup> MRL issued by the Codex Committee of Pesticides Residues unless indicated as EU (European MRL) or F (Finnish MRL).

<sup>b</sup> This pesticide was present in only one sample.

<sup>c</sup> No. of samples analyzed for dithiocarbamates.

<sup>d</sup> No MRL available.

<sup>e</sup> For commodities containing a pesticide in  $\geq 10$  samples, the following 90th percentile values were calculated: cucumber, dicofol = 0.62 mg/kg; green beans, dicofol = 0.38 mg/kg; dimethoate = 0.24 mg/kg; pepper, dithiocarbamates = 0.68 mg/kg; tomato, dicofol = 0.65 mg/kg; orange, dimethoate = 0.5 mg/kg; peach, dimethoate = 1.27 mg/kg; and strawberry, dicofol = 1.29 mg/kg.

carbaryl, carbosulfan, chlorpyrifos, chlorpyrifos-methyl, cyanophos, diazinon, dimethoate, fenitrothion, fenthion, malathion, metalaxyl, omethoate, parathion-ethyl, phosalone, pirimicarb, pirimiphos-ethyl, pirimiphos-methyl, profenofos, prothiofos, pyrazophos, tolclofos-methyl, and triazophos. (2) *Organochlorine compounds and pyrethroids.*—Bromopropylate, carbosulfan, chlorothalonil, cyfluthrin, cypermethrin, *p,p'*-DDD, *p,p'*-DDE, *o,p'*-DDT, *p,p'*-DDT, deltamethrin, dichlofluanid, dicofol, dieldrin, endrin, fenvalerate, alpha-HCH, beta-HCH, gamma-HCH (lindane), delta-HCH, heptachlor epoxide, hexachlorobenzene, iprodione, permethrin, procymidone, propiconazole, tetradifon, triadimefon, triadimenol, trifluralin, and vinclozolin.

All reference materials were certified standards provided by Dr. Ehrenstorfer GmbH. (Germany) and the Food and Agriculture Organization of the United Nations (Rome, Italy) and were prepared in *n*-hexane-acetone. The reference standard for dithiocarbamate compounds was sodium diethyldithiocarbamate, >95%.

### Sampling

A total of 1579 samples were collected from 8 Egyptian local markets located in 6 governorates (Cairo, Giza, Qalyubia, Beni Suef, Minufiya, and Ismailia) throughout 1996. The number of samples analyzed for each commodity is shown in Table 1. For residue analysis, 2 kg of each commodity was prepared according to Codex guidelines (7).

### Residues

The generally recommended method of sampling was used to obtain a representative part of the material to be analyzed (7).

Samples were analyzed immediately upon their arrival at the laboratory, or they were stored at 0–5°C for  $\leq 4$  days before analysis. However, the test samples for dithiocarbamate determination were analyzed immediately after cutting to preclude decomposition of the chemical.

Samples were analyzed for 53 pesticides, which included organophosphorus, organonitrogen, and organochlorine compounds, and certain pyrethroids. A separate specified method of analysis was followed for determination of dithiocarbamates as a group of pesticides; 510 samples were analyzed for dithiocarbamates.

### Extraction and Cleanup

*Multiresidues method.*—In analyses according to the method described by Luke et al. (8, 9), residues are extracted from nonfatty foods by blending with acetone or water-acetone. The pesticides are transferred from the aqueous filtrate into the organic phase by shaking with petroleum ether and dichloromethane; after drying, the organic phase is concentrated just to dryness and then dissolved in hexane-acetone for determination by gas chromatography (GC; 6).

The method allows determination of the 53 pesticide residues listed in Table 2, which also shows the commodities, spiking levels, average recoveries, and coefficients of variation (CVs).



*Dithiocarbamates.*—Residues of dithiocarbamates should be expressed as CS<sub>2</sub> (carbon disulfide) for comparison with the Codex maximum residue levels established on a CS<sub>2</sub> basis. The method selected for analysis is based on the carbon disulfide evaluation procedure (10, 11). The evolved CS<sub>2</sub> is distilled, purified, and collected in an ethanol solution of copper(II) acetate and diethanolamine, in which a yellow complex is formed; the absorbance of the reaction product is measured spectrophotometrically at 435 nm.

### GC Determination

Detection and confirmation of the presence of pesticide residues in food samples depend on the use of chromatography columns of different polarities. An internal standard technique is used for quantitation. Aldrin for organochlorine compounds and pyrethroids with electron capture detection (ECD) and ditalimfos for organophosphorus and nitrogen-containing compounds with nitrogen–phosphorus detection (NPD) were used as internal standards (6).

### Quality Assurance Procedure

All analytical methods and instruments were fully validated as part of a laboratory quality assurance system and were audited and accredited by the Centre for Metrology and Accreditation, Finnish Accreditation Service, Helsinki, Finland (12). This quality system is described in ref. 13.

The criteria of the Codex Committee for quality assurance were followed to determine the performance of the multiresidue method. Recovery, accuracy, limit of determination, and CV were calculated for test compounds determined in different commodities. The recoveries of several test compounds showed that the method could be applied to ca 52 different pesticides efficiently. The average recoveries and CVs of the test compounds ranged from 72 to 118% and from one to 20%, respectively, at the spiking levels shown in Table 2. The reproducibility, expressed as the relative standard deviation (RSD), was <20%. The limit of determination in fruit and vegetable samples ranged between 0.01 and 0.1 mg/kg. The measurement uncertainty including random and systematic error (95% confidence level) were less than ±40%.

The spiked sample analyzed with each set of samples contained 7 indicators representing different types of pesticides: gamma-HCH (lindane), vinclozolin, procymidone, fenvalerate, pirimicarb, dimethoate, and malathion. The blank sample was fortified with the pesticide mixture and analyzed as a normal sample (6).

Recoveries of ethylenebisdithiocarbamates (EBDCs), at various levels of fortification, i.e., 0.1, 1, and 10 mg/kg, from cucumber, tomato, and eggplant were previously studied; they ranged from 80 to 110% (14). The RSD was <20%, and the limit of determination was 0.2 mg/kg (14).

## Results and Discussion

A total of 1579 samples of different types of fruits and vegetables were examined for 53 pesticide residues. Only 510 samples were analyzed for dithiocarbamates.

Twenty-three vegetable crops were analyzed: cabbage, grape leaf, lettuce, Melokhia, dry Melokhia, spinach, watercress, artichoke, broad bean, cauliflower, cantaloupe, cucumber, eggplant, green beans, green peas, okra, onion, pepper, squash, tomato, carrot, sweet potato, and taro. The survey also included 15 types of fruit: apple, apricot, banana, dates, fig, grape, guava, lemon lime, mango, orange, peach, pear, plum, pomegranate, and strawberry.

All samples were examined for residues of 53 pesticides listed in Table 2, and dithiocarbamates were determined as CS<sub>2</sub> in 510 samples of grape leaf, lettuce, cantaloupe, cucumber, eggplant, green beans, green peas, pepper, tomato, apple, grape, peach, and strawberry.

Overall, 76.1% of the samples had no detectable pesticide residues, 23.9% contained detectable residues, and 2.59% contained residues that exceeded maximum residue limits (MRLs). Artichoke, cauliflower, and taro samples were free from pesticide residues. The residues detected, mean levels found, contamination ranges, and the numbers of violative samples are shown in Table 1. The MRLs of Codex Alimentarius were used for comparison when those limits were available. In the absence of Codex MRLs, European and Finnish limits were used. Of the 53 pesticides listed in Table 2, 24 were detected in the analyzed samples. The frequency percentages of the pesticide residues found are shown in Figure 1.

Chlorpyrifos, carbaryl, dimethoate, bromopropylate, and profenofos were the violative pesticides found in the fruit and vegetable samples analyzed. In comparison, chlorothalonil, dicofol, and omethoate were the violative pesticide residues found in the analysis of samples collected in 1995 (6).

The findings of detectable residues were as follows: 91 pepper samples, 5.76% of all samples, contained residues of 13 pesticides, 94 cucumber samples (5.95%) contained residues of 12 pesticides, 38 strawberry samples (2.4%) contained residues of 12 pesticides, 47 grape samples (2.97%) contained residues of 11 pesticides. Other commodities contaminated with residues of ≤10 pesticides are listed in Table 1. Multiple residues are expected on fruits and vegetables because various classes of pesticides must be alternated to prevent resistance from developing in pests.

The rates of sample contamination with various pesticides ranged from 0 to 96.4% in individual fruits and vegetables. The highest violative rate for such contamination was 12.5%; however, the violative rate for cabbage samples was excluded because of the low number of samples analyzed (7 samples, Table 1).

For fruit samples, 38.6% had detectable residues, with 3.49% exceeding the MRLs; in comparison, for vegetable samples, the corresponding values were 21.8 and 2.22%, respectively; and for leafy vegetable samples, they were 10.10 and 3.53%, respectively. These differences would be expected because pesticides are applied directly to the edible commodity, and fruit is often treated close to the time of harvest to ensure that wholesome produce reaches the consumer. In addition, processing treatments such as washing, peeling, canning, or cooking that most foods receive before consumption are very important factors leading to a decrease in the levels of

**Table 2. Recoveries of target pesticides from fortified commodities**

Compound	Pesticide added, mg/kg	Commodity	No. of samples analyzed	Avg. recovery, %	CV, % <sup>a</sup>
Organophosphorus and organonitrogen compounds					
Atrazine	0.2	Apple	2	78	4.6
	0.3	Apple	12	94	2.6
Bendiocarb	0.1	Apple	2	92	6.2
		Pepper	6	87	1
Carbaryl	0.5	Apple	2	92	7.8
		Pepper	6	109	1
Carbosulfan	0.2	Pepper	1	90	—
	0.3	Apple	12	93	3.6
Chlorpyrifos	0.02	Pepper	5	80	12
	0.04	Apple	2	97	11.2
Chlorpyrifos-methyl	0.02	Pepper	5	100	11
Cyanophos	0.05	Pepper	6	81	3
		Apple	2	72	3.9
Diazinon	0.02	Pepper	5	90	15
	0.04	Apple	2	76	2.3
Dimethoate	0.06	Pepper	5	105	9
Fenitrothion	0.02	Pepper	5	80	11
Fenthion	0.05	Pepper	6	94	2
		Apple	2	90	4.8
Malathion	0.06	Cucumber	4	108	1
		Pepper	5	75	4
Metalaxyl	0.5	Pepper	1	110	—
Omethoate	0.05	Pepper	1	83	—
Parathion-ethyl	0.3	Apple	12	93	3.1
Phosalone	0.04	Pepper	2	102	—
	0.3	Apple	12	94	1.9
Pirimicarb	0.06	Pepper	5	82	4
		Cucumber	4	90	1
Pirimiphos-ethyl	0.02	Pepper	2	83	—
	0.3	Apple	12	93	1.4
Pirimiphos-methyl	0.02	Pepper	4	91	8
		Apple	2	82	2.2
Profenofos	0.02	Apple	2	93	11.5
Prothiofos	0.02	Pepper	4	82	20
		Apple	2	83	4.3
Pyrazophos	0.02	Pepper	4	105	17
		Apple	15	90	2.8
Tolclofos-methyl	0.02	Pepper	4	82	2
		Apple	2	82	3
	0.3	Apple	12	94	3.3
Triazophos	0.02	Apple	2	90	6
Organochlorine and pyrethroid compounds					
Bromopropylate	0.05	Pepper	3	97	8
Chlorothalonil	0.03	Pepper	3	91	6

Table 2. (continued)

Compound	Pesticide added, mg/kg	Commodity	No. of samples analyzed	Avg. recovery, %	CV, % <sup>a</sup>
Cyfluthrin	0.1	Pepper	1	114	—
	1.0	Apple	12	95	4.5
Cypermethrin	0.1	Pepper	6	117	2
		Apple	3	112	6.4
<i>p,p'</i> -DDD	0.02	Pepper	6	113	5
	0.05	Apple	12	104	2.1
<i>p,p'</i> -DDE	0.02	Apple	3	112	2.6
<i>o,p'</i> -DDT	0.02	Pepper	3	95	5
	0.05	Apple	12	95	4
<i>p,p'</i> -DDT	0.02	Pepper	6	76	5
		Apple	3	112	2.6
Deltamethrin	0.2	Pepper	6	101	4
Dichlofluanid	0.05	Pepper	9	107	9
		Apple	2	109	1.3
Dicofol	0.02	Pepper	2	118	8
Dieldrin	0.01	Pepper	3	99	8.3
	0.05	Apple	12	94	2.9
Endrin	0.06	Orange	1	82	—
	0.05	Apple	12	97	1.6
Fenvalerate	0.02	Pepper	5	94	11
		Cucumber	4	107	5
alpha-HCH	0.01	Pepper	6	94	3
		Apple	3	10	8.7
beta-HCH	0.01	Pepper	3	110	13
gamma-HCH (lindane)	0.01	Cucumber	4	85	4
		Pepper	5	83	13
delta-HCH	0.01	Pepper	3	100	14
Heptachlor epoxide	0.01	Pepper	6	94	8
		Apple	15	108	2.4
Hexachlorobenzene	0.01	Pepper	3	111	18
Iprodione	0.5	Pepper	6	111	4
		Apple	3	110	2.8
Permethrin	1.0	Pepper	6	101	9
		Apple	15	103	3.8
Procymidone	0.06	Pepper	5	77	9
		Cucumber	4	104	3
Propiconazole	0.05	Pepper	3	100	6
Tetradifon	0.03	Pepper	2	90	11
Triadimefon	0.05	Pepper	6	109	5
		Apple	3	98	3.9
Triadimenol	0.1	Apple	1	76	—
Trifluralin	0.01	Pepper	3	100	19
	0.1	Apple	12	90	3.3
Vinclozolin	0.01	Pepper	5	97	15
		Cucumber	4	99	9

<sup>a</sup> CV = coefficient of variation.

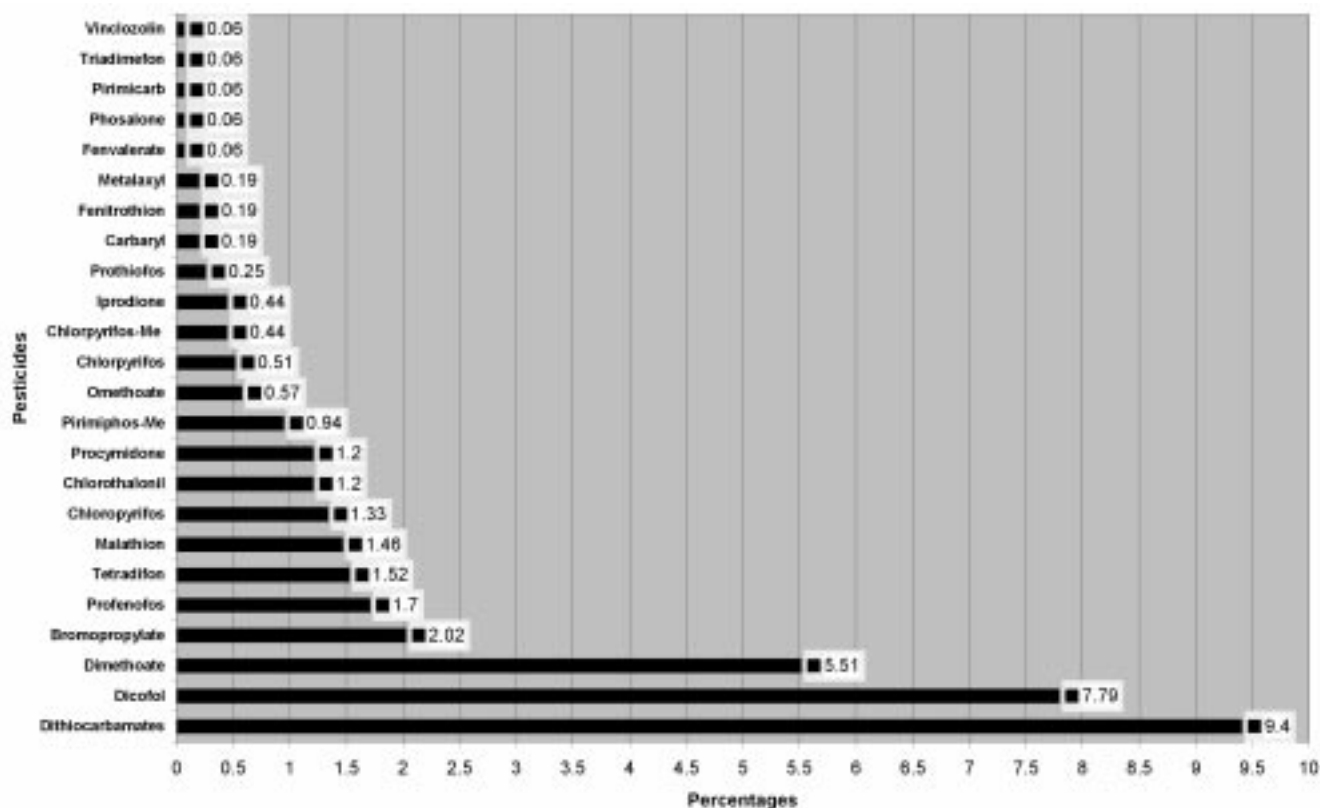


Figure 1. Frequency percentages of residues found over all concentration ranges in fruit and vegetable samples collected from Egyptian local markets during 1996.

any residues left on crops at harvest. The data in Table 1 also show that root vegetable samples had the lowest contamination rate, with none exceeding the MRLs.

The results of the current study, in which no residues of restricted or banned pesticides such as DDT, HCH, and their metabolites were detected in any analyzed samples, were comparable with the results of the previous monitoring study in 1995 (14). However, the pesticides found in the previous study were very persistent in soil.

In comparison, the rates of contamination in the 1996 monitoring program of the U.S. Food and Drug Administration (15) were 47.4 and 65.3%, for fruit samples and vegetable samples, respectively; the corresponding violative rates were 1.3 and 1.1%, respectively. The current study had slightly lower contamination rates for fruit and vegetable samples, i.e., 38.6 and 21.8%, respectively, and higher violative rates for fruit and vegetable samples, i.e., 3.49 and 2.22%, respectively.

Also, a study conducted in Belgium from 1991 to 1993 (16) showed contamination and violative rates that were higher than those in the current study.

Table 1 shows that dithiocarbamate residues were found in 48 of 510 samples analyzed for dithiocarbamates, resulting in a contamination rate of 9.4% and a violative rate of 0.39%. Analyses of 11 squash, 3 carrot, 4 apricot, 9 fig, 2 mango, and 9 plum samples did not show any detectable residues of dithiocarbamates. Of the samples analyzed, only 2 contained dithiocarbamates that exceeded the MRLs: one grape sample

and one peach sample, each containing 1.2 mg/kg. These results suggest successful implementation of good agricultural practices.

The concentrations of dithiocarbamate residues found in the current study were lower than those found in the previous monitoring studies conducted during 1993 (14) and 1995 (6).

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