The Connecticut Agricultural Experiment Station, New Haven

Pesticide Residues in Produce Sold in Connecticut 1999

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Pesticides play a vital role by controlling insects, vermin, and disease in the agricultural, industrial, home/garden, and public health sectors. Historically pesticides have been regulated by both the United States Environmental Protection Agency (EPA) and the Food and Drug Administration (FDA) under the Federal Insecticide, Rodenticide, and Fungicide Act (FIFRA) and the Federal Food, Drug, and Cosmetic Act (FFDCA). The definition of ‘pesticide chemical’ under FFDCA was narrower than the definition of ‘pesticide’ under FIFRA. The 1996 Food Quality Protection Act (FQPA) amended both FIFRA and FFDCA. FQPA specifically amended the definition of ‘pesticide chemical’ in FFDCA to mean “…any substance that is a pesticide under FIFRA, including all active and inert ingredients of such pesticide…”(Federal Register 1998). Now, under both FIFRA and FFDCA, a ‘pesticide’ is defined as “…any substance or mixture of substances intended for preventing, destroying, repelling, or mitigating any insects, rodents, nematodes, fungi or weeds or any other forms of life declared to be pests, and any substance or mixture of substances intended for use as a plant regulator, defoliant or desiccant…” (FIFRA 1947, 1959). The term ‘pesticide’ includes the categories of insecticides, herbicides, rodenticides, fungicides, nematicides, and acaracides as well as disinfectants, fumigants, wood preservatives, and plant growth regulators.

There are 860 active ingredients (a.i.) registered as pesticides which are contained in 20,700 registered pesticide products regulated by EPA under FIFRA and FFDCA. In 1997, a typical year and the last year for which figures are available, 4.5 billion pounds of chemicals were used as pesticides (based on a.i.) in the United States. Because pesticides appear to be so prevalent in society, it is important to put their use into perspective. Conventional pesticides, those developed and produced primarily for use on crops, comprise some 21% of the total. Other pesticide chemicals, such as sulfur and petroleum distillates, produced for other uses but also used as pesticides, compose another 6% of the total. Taken together these two categories comprise 27% (1.23 billion pounds of a.i.) of the annual sum of pesticides used in the United States as seen in Table 1. Pesticides used in the production of food and fiber comprise about 77% (0.94 billion pounds of a.i.) of this quantity. They account for about 20% of the total pesticide usage per annum in the United States. Nearly three quarters of the pesticides fall into the categories of wood preservatives, specialty biocides, and chlorine and hypochlorites. These are all regulated under FIFRA and FFDCA. Larger quantities of pesticides are used annually to disinfect drinking and wastewater than are used in agriculture (Table 1). Annually, per capita, it is estimated that 4.6 pounds of conventional and other pesticide chemicals and 17 pounds of all pesticides are used in the United States at a cost of $44 per individual. The average American household spends about $20 for conventional pesticides applied by the homeowner. This figure does not include expenditures for pesticides applied to homes and gardens by hired contractors (Aspelin and Grube 1999).

Within the United States, the use of pesticides on crops falls under the jurisdiction of the EPA. All pesticides used to protect commodities in the United States must be registered with the EPA. The EPA sets allowable tolerances for pesticides for each commodity. A tolerance is defined as the maximum quantity of a pesticide residue allowable on a raw agricultural commodity. Tolerances impact food safety by limiting the concentration of a pesticide residue allowed on a commodity, and by limiting the type of commodity on which it is allowed. Tolerances are the only tool the EPA has under the law to control the quantity of pesticides on the food we consume. The enforcement of these food tolerances falls under the jurisdiction of the FDA, and in this state, the Connecticut Department of Consumer Protection (DCP). To be able to enforce the EPA mandated tolerances, these agencies must know the quantity and the type of pesticide residue present in foodstuffs offered for sale.

The Department of Analytical Chemistry at The Connecticut Agricultural Experiment Station has established a program in conjunction with DCP to examine fruits and vegetables sold in the state for pesticide residues. This market basket survey concentrates on fresh produce grown in this state, but also includes fresh produce from other states and foreign countries and some processed food. The primary goal is to determine if the amounts and types of pesticides found on fruits and vegetables are in accordance with the tolerances set by EPA. Violations of the law occur when pesticides are not used in accordance with label registration and are applied in excessive amounts, or when pesticides are applied to crops on which they are not allowed.

METHODS

Samples of produce grown in Connecticut, other states, and foreign countries are collected at various Connecticut producers, retailers, and wholesale outlets by inspectors from the DCP. The samples collected are brought to our laboratory in New Haven for pesticide residue testing. These market basket samples are collected without prior knowledge of any pesticide application.

Commodities are tested for pesticides using a multi-residue method developed in our laboratories (Pylypiw 1993). In most cases, each sample is prepared in its natural state as received, unwashed and unpeeled. The sample is chopped and a portion is placed in a blender. Organic solvents are added and the mixture is blended to extract the pesticides from the sample. Interfering coextracted compounds, such as organic acids, are removed from the solvent extract with water. A small amount of the extract is then injected into various gas chromatographic instruments to determine how much, if any, pesticides are present. Our method is capable of determining pesticides with recoveries ranging from 81% to 114%, and has an average detection limit of 10 parts per billion.

RESULTS AND DISCUSSION

In 1999 a total of 195 samples representing a wide variety of fresh and processed produce were tested. Of these 143 (73%) were fresh produce and 52 (27%) were processed foods. Pesticide residues were found in 70 samples or 49% of the fresh produce samples and 10 samples or 19% of the processed foods, see Tables 2 and 3. The value of 49% in fresh produce is somewhat higher than the corresponding value of 39% for fresh produce found in 1998. The value of 10% for processed foods was identical to that found in 1998. The apparent above normal value for residues found in fresh produce is likely the result of sampling a narrower range of commodities in 1999 that tend to be pesticide intensive.

Of the samples analyzed in 1999, eight samples, or 4.1%, were found to contain pesticide residues that were violative. This percentage is higher than any other year since the inception of the Connecticut market basket survey, and may be due to the fact that fewer commodity types were analyzed. Seven of the eight violations were found on fresh produce, and six of the eight were on produce grown within the state. Specifically, chlorothalonil was found on two pepper samples, permethrin was found on a sample of plums, and dicloran (DCNA) was found on two samples of strawberries. A sample of Canadian cucumbers was found to contain residues of iprodione, and a sample of processed Belgian peas was found to contain vinclozolin. These are all violations because none of these pesticides is allowed on the corresponding commodities. Violations are immediately reported to DCP, which has the responsibility for enforcing pesticide tolerances in Connecticut. In the case of a violation from produce grown outside of Connecticut, DCP notifies the FDA. The Belgian peas were recalled voluntarily first within the state and then nationally by the distributor as a direct result of this testing program.

In 1999, seven samples (3.6%) of fresh produce were found to contain residues of chlordane or DDE, a soil metabolite of DDT. This number of persistent organohalogen pesticides (POPs) is identical to the average number of 3.6% found annually on food crops in our survey since 1990. The use of POPs on food crops was banned in the U.S. in 1978. Residues of these pesticides continue to persist in the environment, and their uptake and accumulation by crops such as squash, cucumbers and carrots have been well documented (Pylypiw et al. 1991, Pylypiw et al. 1997). In 1999 DDE or chlordane was found in fresh samples of snap beans, cucumbers, eggplant, lettuce, potatoes, peppers, and squash. The FDA has set action levels (allowable amounts) for these residues in produce (Duggan 1998). No sample that contained DDE or chlordane was above the FDA action level.

A total of 119 pesticide residues was found on 80 samples of processed and fresh produce in 1999. The most commonly found pesticides were the fungicides, captan and iprodione, and the insecticide endosulfan. Captan was found 30 times in 1999 over a wide variety of fresh produce; no processed produce contained captan. Captan accounted for 25% of the residues found during the past year, and has accounted for approximately 12% of our findings since 1990. It is interesting to note that captan is more prevalent on fresh produce and has only been found once on processed produce during the past ten years. Iprodione and endosulfan were found 17 and 16 times respectively on both fresh and processed produce. Endosulfan is the most commonly found pesticide residue on fresh and processed produce, accounting for nearly 28% of the residues found since 1990. Iprodione is the fourth most commonly found pesticide residue through the 10-year period of the survey, accounting for some 7% of all residues (Krol et al. 1999).

RINSING PRODUCE REDUCES PESTICIDE RESIDUE LEVELS

Over the past 10 years more residues have consistently been found on raw produce than on the corresponding processed commodities. This led to the hypothesis that certain types of processing or household preparation may reduce pesticide residues. Suspecting that rinsing may play some role in reducing residue levels, we initiated a study to examine the effects that rinsing produce under tap water has upon pesticide residue levels. Several crops commonly grown in Connecticut were planted at Lockwood Farm and treated by us with eleven pesticides: the insecticides chlorpyrifos, diazinon, endosulfan, malathion, methoxychlor, bifenthrin, permethrin and the fungicides captan, chlorothalonil, iprodione and vinclozolin. Residues of DDE, a metabolite of DDT, were also studied as they were present in the soil. Following application, the pesticides were allowed to weather naturally. The crops were harvested at marketable size; the produce was brought to the laboratory, and split into two subsamples. One of these subsamples was analyzed for residues using our standard procedure (Pylypiw 1993), and the second subsample was rinsed with tap water prior to standard analysis.

Statistical analysis of 98 pairs of data showed that rinsing removed residues for nine of the twelve pesticides studied. Residues of vinclozolin, bifenthrin and chlorpyrifos were not reduced. In general, the rinsability of a pesticide is not correlated with its water solubility (Krol et al. 2000).

Captan is the most common fungicide found as part of our market basket survey. When found, its residues average 0.98 ppm. Captan is used widely both pre- and post- harvest on numerous crops and is allowed at relatively high concentrations, up to 100 ppm on several commodities. Captan is described as a probable carcinogen (Extension Toxicology Network 2000), and therefore, it was particularly interesting that captan is drastically reduced by routine rinsing under tap water.

The acceptable daily intakes (ADI) are listed in Table 5 as a means of comparing the relative toxicity of the pesticides examined (Extension Toxicology Network 2000). An ADI represents a 100-fold safety factor to the no-observed-adverse-effect level (NOAEL) as determined in animal studies (Fong et al. 1999). This number is useful when comparing acute toxicity only. The ADI of captan, a compound with strong evidence of being a carcinogen, is higher than the ADI of diazinon, a non-carcinogen, because diazinon is an acute toxin. For captan, the ADI is 0.1 mg/kg of body weight per day, meaning that a person weighing 50 kg (110 lbs) could consume 5 mg of captan per day with no observable effect.

As an example, the tolerance of captan on strawberries is 25 mg/kg. At the maximum tolerance level, a 50 kg (110 lbs) person would have to eat 200 g of strawberries to equal the ADI. A child of 20 kg (44 lbs) would only have to eat 80 g, or about four medium sized strawberries. This study has shown that captan is reduced by about 77% during routine rinsing with tap water. As a result of rinsing produce prior to ingestion the quantity of strawberries ingested could be increased to 910 g for a person of 50 kg without exceeding the ADI. At the average of 0.98 ppm found on strawberries in our market basket survey, a 50 kg person would need to consume about 5 kg (11 lbs) of strawberries in a day, and a 20 kg (44 lbs) child would need to consume 2 kg (4.4 lbs) of strawberries in a single day to equal the ADI. Nevertheless, reduction in dietary exposure to pesticides, in this case as the result of rinsing of the produce, is highly desirable.

CONCLUSIONS

A summary of our market basket survey findings over the past 10 years and the results of the past 10 years of the FDA residue-monitoring program are presented in Figure 1. The pie charts show that 34% of the samples in our market basket survey contain pesticide residues, and that 36% of the samples in the FDA residue program contain pesticide residues. In 1999, 41% of the produce tested in our market basket survey were found to contain pesticide residues. This number is slightly above the average value of 34% over the past ten years of our survey. Our findings continue to show that residues of pesticides on fruits and vegetables in Connecticut are generally well within tolerances set by the EPA. Work conducted in our laboratories indicate that rinsing fresh produce under tap water helps reduce the levels of nine of the twelve pesticides studied, and that rinsibility of pesticide residues is not correlated to their water solubility.

ACKNOWLEDGEMENTS

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Table 1. Breakdown of Pesticide Usage in the United States for the Year 1997.

Type	Billions of Pounds ai	Billions of Pounds ai	%
Conventional pesticides		0.97	21
Other pesticide chemicals (sulfur, petroleum, etc.)		0.26	6
Subtotal		*1.23**	*27**
Wood preservatives		0.66	14
Specialty biocides		0.27	6
Swim pools, spas, industrial water treatment	0.186		
Industrial / Household disinfectants	0.035		
Other (paints / coatings / adhesives / polymers / plastics / etc.)	0.051		
Chlorine / Hypochlorites		2.46	53
Disinfection of potable and waste water	1.476		
Disinfectants and pool disinfectants	0.983		
Subtotal	*2.73*	*3.39*	*73*
Total		4.63	100

***** 77% (0.95 billion lbs of a.i.) of this is used in the production of food and other agricultural commodities. The remainder is used by private homeowners and/or other pest control (Courtesy of Aspelin & Grube, 1999).

Table 2. Summary of pesticides found in fresh fruits and vegetables sold in Connecticut, 1999.

Commodity	Pesticide	Samples with residues	No. of times detected	Residue range (ppm)	EPA tolerance (ppm)
Apples (29 samples)		17			
	Captan		10	0.01-3.3	25
	Chlorpyrifos		7	0.003-0.07	1.5
	Dicofol		3	0.05-.21	5
	Endosulfan		1	0.01	2.0
Asparagus (2 samples)		0			
Beans, Snap (2 samples)		1			
	Chlorothalonil		1	0.17	5
	DDE		1	0.003	0.2(a)
	Dicofol		1	0.18	5
Blueberries (5 samples)		1			
	Captan		1	0.18	25
Broccoli (3 samples)		1			
	Iprodione		1	0.05	25
Carrots (3 samples)		0			
Cucumbers (5 samples)		4			
	Chlorothalonil		1	0.03	5
	DDE		1	0.05	0.1(a)
	Endosulfan		1	0.01	2.0
	Iprodione		1	0.1	0(b)
Eggplant (2 samples)		2			
	Chlorothalonil		1	0.02	0(b)
	DDE		1	0.01	0.1(a)
Grapefruit (2 samples)		0			
Grapes (10 samples)		7			
	Captan		3	0.3-20	50
	Chlorpyrifos		1	0.004	0.5
	Iprodione		6	0.003	60.0
Lettuce (5 samples)		1			
	DDE		1	0.003	0.5(a)
Nectarines (1 sample)		1			
	Permethrin		1	0.02	0.5
Peaches (5 samples)		4			
	Captan		3	0.5-1.5	50
	Iprodione		2	0.85-0.5	20.0

Table 2. Summary of pesticides found in fresh fruits and vegetables sold in Connecticut, 1999 (contnued).

Commodity	Pesticide	Samples with residues	No. of times detected	Residue range (ppm)	EPA tolerance (ppm)
Pears (5 samples)		1			
	Endosulfan		1	0.06	2.0
Peppers, Bell (8 samples)		4			
	Chlorothalonil		2	0.03-0.21	0(b)
	Chlorpyrifos		1	0.06	1.0
	DDE		1	0.003	0.1(a)
	Endosulfan		2	0.01-0.01	2.0
	Permethrin		1	0.21	1.0
Plums (1 sample)		1			
	Permethrin		1	0.04	0(b)
Potatoes (4 samples)		3			
	CIPC		2	0.5-0.02	50
	DDE		1	0.003	1
Squash, Summer (4 samples)		3			
	Endosulfan		3	0.01-1.4	2.0
Squash, Winter (3 samples)		2			
	Chlordane		1	0.02	0.1
	Chlorothalonil		1	1.7	5
	DDE		1	0.004	0.1
	Dicofol		1	0.55	5
Strawberries (22 samples)		16			
	Captan		13	0.01-6.8	25
	DCNA		2	0.01-0.24	0(b)
	DCPA		1	0.04	2
	Endosulfan		6	0.01-0.07	2.0
	Iprodione		8	0.86-0.04	15
	Vinclozolin		6	0.07-1.1	10
Tomatoes (7 samples)		1			
	Chlorothalonil		1	0.14	5
	Endosulfan		1	0.02	2.0
	Permethrin		1	0.09	2
Miscellaneous (1 sample of each)		0			

Cabbage, Cantaloupe, Celery, Corn, Endive, Kiwifruit, Melon, Onions, Peas, Pumpkins, Radishes, Raspberries, Spinach, Tangelo, Watermelon

(a) Allowed as per FDA Action Level
(b) Residue not allowed on this commodity

Table 3. Summary of pesticides found in processed fruits and vegetables sold in Connecticut, 1999.

Commodity	Pesticide	Samples Analyzed	Samples with residues	No. of times detected	Residue range (ppm)
Juices					
Apple Cider/Juice		15	0		
Fruits & Vegetables, canned					
Beans, snap		4	0		
Mushrooms		1	1		
	Chlorpyrifos			1	0.05
Peas		2	1		
	Vinclozolin			1	0.02(a)
Peaches		2	0		
Spinach		2	2		
	Permethrin			2	1.3-0.5
Miscellaneous	(1 sample of each)		0		
Apples, Asparagus, Clementines, Cucumbers, Pears, Pineapples, Water Chestnuts					
Fruits & Vegetables, packaged fresh					
Lettuce/Lettuce Mix		4	1		
	Endosulfan			1	0.1
	Permethrin			1	0.33
Miscellaneous	(1 sample of each)		0		
Celery, Chard, Onions, Spinach, Alfalfa Sprouts					
Fruits & Vegetables, frozen					
Strawberries		2	2		
	Iprodione			2	0.05-0.2
Spinach		1	1		
	Permethrin			1	4.7
Baby Food					
Apples		2	0		
Bananas		2	0		
Peaches		2	2		
	Chlorpyrifos			1	0.01
	Permethrin			2	0.06-0.15
Pears		1	0		

(a) Violative Sample, no tolerance

Table 4. Ten year summary of all market-basket samples tested, including organic and processed food samples.

Year	Total Samples Tested	Samples With No Residues	Samples With Residues Within EPA Tolerances	Samples With Residues Over EPA Tolerances	Samples With Residues With No EPA Tolerances
1990	418	186	230	0	2
1991	285	190	94	0	1
1992	273	179	89	1	4
1993	441	305	128	3	5
1994	545	414	125	1	5
1995	444	307	129	0	8
1996	327	188	134	1 (a)	4
1997	412	266	144	0	2
1998	180	115	63	0	2
1999	195	115	72	0	8
Total	3520	2265	1208	6	41

(a) Over FDA action level.

Table 5. Rinsability and ADI values for pesticides studied.

Pesticides	Reduced With Rinsing	Acceptable Daily Intake (ADI) mg/kg/day	Water Solubility (mg/L @ 20°C)
Insecticides			
Endosulfan	Yes	0.006	0.32
Permethrin	Yes	0.05	0.2
Diazinon	Yes	0.002	40
DDE	Yes	0.02 (DDT)	<1
Chlorpyrifos	No	0.01	2
Methoxychlor	Yes	0.1	0.1
Malathion	Yes	0.02	130
Bifenthrin	No	0.015	0.1
Fungicides			
Captan	Yes	0.1	3.3
Vinclozolin	No	0.07	3.4
Iprodione	Yes	0.2	13
Chlorothalonil	Yes	0.03	0.6

(Extension Toxicology Network, 2000)

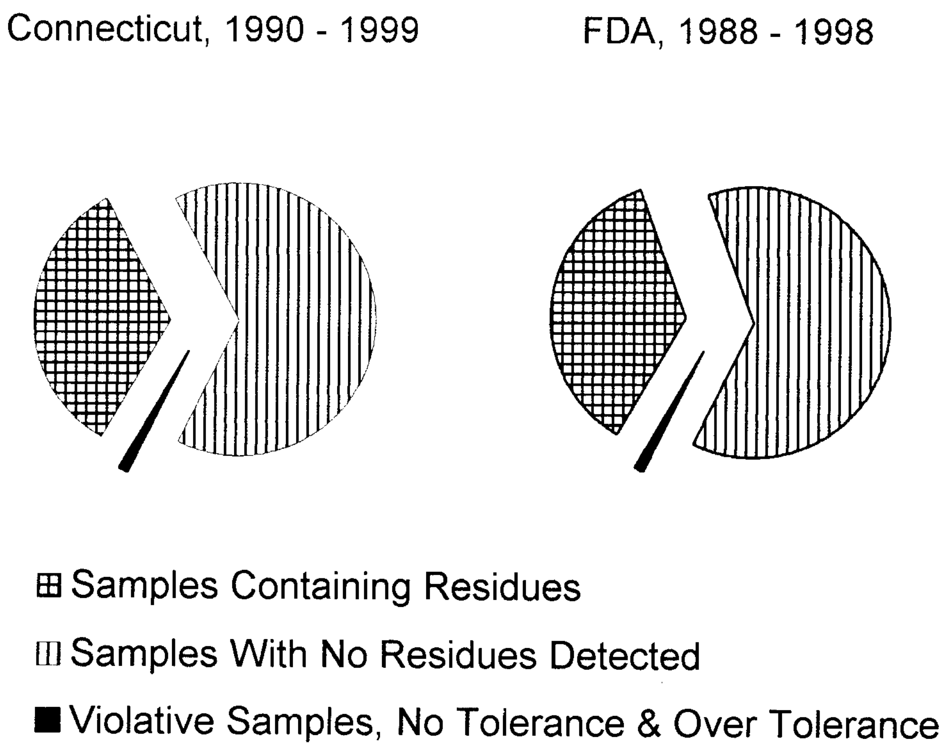

Figure 1. Summary of Connecticut data from 1990 – 1999, and of FDA data from 1988 – 1998.