Reduction of Pesticide Residues on Produce by Rinsing

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In 1997 this laboratory initiated a research program with the objective of examining the effect that rinsing of produce with tap water would have on pesticide residues. Samples were obtained from local markets and/or grown at our experimental farm. Because approximately 35% of produce from retail sources contains pesticide residues, growing and treating produce at an experimental farm had the advantage that all such samples contain pesticide residues. Pesticides were applied under normal field conditions to a variety of food crops and the vegetation was allowed to undergo natural weathering prior to harvest. The resulting samples contained field-incurred or "field-fortified" residues. This experimental design was employed to mimic as closely as possible real world samples. Crops were treated, harvested, and divided into equal subsamples. One subsample was processed unwashed, whereas the other was rinsed under tap water. The extraction and analysis method used was a multi-residue method developed in our laboratory. Twelve pesticides were included in this study: the fungicides captan, chlorothalonil, iprodione, and vinclozolin; and the insecticides endosulfan, permethrin, methoxychlor, malathion, diazinon, chlorpyrifos, bifenthrin, and DDE (a soil metabolite of DDT). Statistical analysis of the data using the Wilcoxon signed-rank test showed that rinsing removed residues for nine of the twelve pesticides studied. Residues of vinclozolin, bifenthrin, and chlorpyrifos were not reduced. The rinsability of a pesticide is not correlated with its water solubility.

Keywords: *Field-incurred residues; pesticide water solubility*

INTRODUCTION

The use of pesticides in commercial agriculture has led to an increase in farm productivity so that a relatively small number of farmers can produce a wide variety and abundance of agricultural commodities at a reasonably low cost. The disadvantage of pesticide use is that residues may remain on agricultural commodities where they contribute to the total dietary intake of pesticides. Many of these compounds are known carcinogens and/or toxins, and therefore, it is desirable to reduce these residues.

The U.S. Environmental Protection Agency (EPA) has established tolerances (Code of Federal Regulations, 1999), or allowable residue levels, for all pesticides, and the U.S. Food and Drug Administration (FDA) is responsible for enforcing these tolerances. In its market-basket survey of all produce sold in the United States, the FDA monitors the nation's food supply for pesticide residues and publishes its results yearly (Food and Drug Administration, 1998). The Department of Analytical Chemistry at the Connecticut Agricultural Experiment Station (CAES) also conducts a market basket survey in conjunction with the Connecticut Department of Consumer Protection (DCP) to examine fruits and vegetables sold in this state for pesticide residues. This pesticide residue monitoring program is a continuing effort within the state and summary reports are published annually by the Experiment Station (Krol et al., 1999). The results of our program from 1990 through 1999 compare well with results obtained by the FDA for those years. On average, 37% of the FDA samples tested contained residues (Food and Drug Administration, 1998), and 1% are violative. The Connecticut results (Krol et al., 1999) showed that 35% of the samples tested contained residues, and 1% are violative.

Although it has been assumed for many years that rinsing fruits and vegetables prior to consumption reduces the amount of residues, this anecdotal approach needs laboratory confirmation. There are several studies that have examined the effect of washing produce to remove pesticide residues as a step in commercial crop processing (Mergnat et al., 1995; Cabras et al., 1997; Cabras et al., 1998a and b; Abou-Arab, 1999). These studies are of little practical use to the consumer who wants to know what effect household preparation has upon reducing pesticide residue levels. There are a handful of studies that have examined the effect of washing as part of larger home processing studies (Khaire, 1983; Burchat et al., 1988; Celik et al., 1995; Schattenberg et al., 1996; Ramesh and Balasubramanian, 1999), and a single study of washing alone on a single pesticide on a single crop (Leyva et al., 1998).

In 1996 Schattenberg examined the effects of common household preparation, and reported that pesticide residue levels were reduced by washing, peeling, and/or cooking (Schattenberg et al., 1996). In Schattenberg's study samples obtained in the marketplace were analyzed for residues and those found to contain residues were subjected to a secondary process at a later date. The results suggested that residues decreased with all of the treatments; however in most cases, the sample sizes were too small to apply statistical analysis. In

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Table 1. Summary of Samples Containing Pesticide Residues

commodity, number of samples	endo-sulfan	per-methrin	diazinon	DDE	chlor-pyrifos	methoxy-chlor	mala-thion	bi-fenthrin	captan	vin-clozolin	iprodione	chloro-thalonil	total
strawberries, 36	21	3	3		2	2	4	2	21	20	7		85
lettuce, 23	16	12	9	9	5	5	2	1	5			3	67
spinach, 18	8	10	6	4		3	1	2	5			3	42
peas, 4	3	4	2			2		2				1	14
raspberries, 3	3								3	3	3		12
beet tops, 3	3	3	1	3	1							1	12
beets, 2	3	1	1	3	2							1	11
tomatoes, 2	1	2			2								5
peaches, 2											2		2
green beans, 1	1	1		1									3
cucumbers, 1	1			1									2
apples, 1					1								1
asparagus, 1		1											1
nectarines, 1											1		1
total samples, 98	60	37	22	21	13	12	7	7	34	23	13	9	258

addition, sample storage may lead to some degradation of residues. Chlorothalonil has been shown to degrade on produce during storage in a refrigerator (Pylypiw et al., 1997). Schattenberg's study did not distinguish between the individual effects of washing with and without a surfactant, peeling, or cooking.

In 1997 we initiated a research program with the objective of examining the effect that the simple household technique of rinsing with tap water would have on reducing pesticide residues in produce. A wide variety of crops that contained pesticide residues were examined (Table 1). Some samples were collected at local farms and grocery stores as part of a market basket study and others were grown at our experimental farm. At the experimental farm, a series of pesticides were applied simultaneously to achieve levels approximating those found in our market basket survey. This approach allowed the rinsability profile of several pesticides to be studied on a single sample.

Twelve pesticides were studied: the insecticides chlorpyrifos, diazinon, endosulfan, malathion, methoxychlor, bifenthrin, and permethrin; the fungicides captan, chlorothalonil, iprodione, and vinclozolin. Residues of DDE, a metabolite of DDT, were also studied. Although many persistent organohalogen pesticides (POPs), such as DDT, were banned for use on food crops between 1972 and 1978 in the United States, they have remained in the environment where they continue to be incorporated into plant biomass (Pylypiw et al., 1991). Following application, the pesticides were allowed to weather naturally. The crops were harvested at marketable size, and 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. The data were analyzed using a one-tailed Wilcoxon signed-rank test for paired data that are not normally distributed.

MATERIALS AND METHODS

Apparatus. *Gas chromatograph (GC).* Model 5890 (Hewlett-Packard Co., Avondale, PA), equipped with the following detectors: (1) ^{63}Ni electron capture detection (ECD) system; (2) flame photometric detection (FPD) system operating in P mode; (3) electrolytic conductivity detector (ELCD) operating in halogen mode or a halogen specific detector (XSD). One GC was configured with dual detectors (for ELCD or XSD and FPD), and another was configured using a single detector (for ECD). General operating conditions were as follows: initial temperature 175 °C; no initial hold time; ramp rate 1 °C/min; final temperature 250 °C; final hold time, 10 min; total run time, 85 min; carrier gas, He; makeup gas for ECD, 5% CH_4/Ar, with flow rate of 20 mL/min; makeup gas for XSD, air, with flow rate of 30 mL/min. Gas purifier (all gases except air), OMI-1 (Supelco Inc.) indicating purifier. Injector, HP-19251A; temperature 225 °C; operated in the splitless mode; purge off time, 0.50 min. Auto injector, HP-7673; 2−4 μL injection volume. Detector conditions: ECD, 325 °C; FPD, 265 °C, flame mixture, hydrogen-air; ELCD, model 4420; reactor temperature, 900 °C; vent time 3.5 min; XSD, model 5360 (both from OI Analytical Corp., College Station, TX); reactor temperature 1000 °C.

Chromatographic column. Capillary, 30 m × 0.32 mm, 0.25 μm film, SPB-5 (cat. no. 2-4048, Supelco, Inc., Bellefonte, PA).

Data Collection. All GC data were collected on a Hewlett-Packard Vectra 486 data station running HP 3365 series II Chemstation version 3.34 software.

Glassware. Separatory funnel, 500 mL, with Teflon stopcock and glass stopper; filtering funnel, 150 × 150 mm; glass wool, 40-mL screw-cap vial.

Food cutter. Model 84145 (Hobart Corp., Troy, OH).

Blender. Explosion-resistant (Waring), cat. no. 14-509-53; blender containers, 1 qt, cat. no. 14-509-11A (Fisher Scientific).

Reagents. *Solvents.* Petroleum ether (30−60 °C) cat. no. 9265-03; 2-propanol, cat. no. 9334-03; toluene, cat. no. 9336-03; and 2,2,4-trimethylpentane, cat. no. 9355-03 were purchased from J. T. Baker Chemical Inc., Phillipsburg, NJ.

Distilled Water. Water was used directly from the still (Barnstead/Thermolyne, model A1013-B) without further purification.

Saturated sodium sulfate. Approximately 250 g of anhydrous granular sodium sulfate (cat. no. 3375-05, resi-analyzed grade, Baker) was added to 800 mL of distilled water, and the mixture was warmed on a steam bath until the sodium sulfate crystals dissolved. The solution was cooled overnight at room temperature to allow the excess sodium sulfate crystals to precipitate.

Pesticide Standards. All pesticide standards were obtained from the U. S. Environmental Protection Agency, National Pesticide Standard Repository, Fort Mead, MD. All standards were prepared from the compound as received. An approximately 10−15-mg sample of each pesticide was weighed accurately on an analytical balance and dissolved in 10 mL of toluene, and the concentration was calculated. This was the individual pesticide stock standard. The stock standard was diluted with 2,2,4-trimethylpenatane to give a 10 or 100 μg/mL intermediate standard, from which individual and mixed standard solutions were prepared. Individual standards were prepared at 0.2 and 1 μg/mL; mixed standard solutions were prepared with individual analyte concentrations ranging from 0.1 to 3 μg/mL.

Sample Preparation. *Produce Samples.* Over the course of three years, various crops were seeded directly in the field, or they were seeded in a greenhouse and transplanted outdoors as appropriate. Pesticides were applied in accordance with

label instructions prior to maturity and harvest. In addition, other samples were collected at local farms and grocery stores.

Pesticide Formulations. Ambush (permethrin) was obtained from Zeneca Ag Products, 1800 Concord Pike, Wilmington, DE 19850-5458; Brigade (bifenthrin) and Thiodan (endosulfan) were obtained from FMC Corporation, Agricultural Products Group, Philadelphia, PA 19103; all other pesticides were obtained as over-the-counter formulations.

Pesticide Applications. All applications were made using a one-gallon hand-held home and garden sprayer. Pesticides of interest were mixed and diluted in 0.5 or 1 gallon of water as required. Pesticides were mixed in the tank to concentrations between 300 μg/mL and 2000 μg/mL. Applications were made to field crops such that residue levels on the crops would approximate those levels observed throughout the course of our market basket survey.

Harvest. Crops were harvested, placed into brown paper bags, labeled with identification, and immediately brought into the laboratory for processing.

Sub-Sampling and Rinsing. The samples were equally divided into two subsamples by hand and assigned unique identifiers. The sample to be rinsed was placed in a plastic colander and rinsed under cold tap water for 15−30 s, with gentle rotation by hand. We believe that this method accurately mimics actual household food preparation. Both subsamples were processed concurrently to avoid problems with degradation of residues during storage.

Sample Extraction. A multi-residue procedure developed in our laboratories and published elsewhere was used for sample extraction (Pylypiw, 1993). Briefly, samples of produce were homogenized in a food cutter or blender. A 50-g portion was extracted with 50 mL of 2-propanol and 100 mL of petroleum ether using an explosion-proof blender. The resulting extract was filtered into a 500-mL separatory funnel and back-washed with 150 mL of distilled water three times. Five mL of a solution of saturated sodium sulfate was added to the first and last wash to minimize emulsions and maximize extraction efficiency. The petroleum ether layer was collected in a 40-mL screw-cap vial and dried over sodium sulfate. The extract was analyzed by GC with ECD, FPD, XSD, and/or ELCD detection. The number of samples and pesticides found on each are presented in Table 1.

Statistical Analysis. A one-tailed Wilcoxon signed-rank test was used to judge the statistical significance of pesticide rinsability. Analysis of data was performed using the Jandel SigmaStat Data package, version 2.0.

RESULTS AND DISCUSSION

The field-incurred or “field-fortified" method used in the present study differs from laboratory fortification in that it mimics pesticide application under normal, real-world, agricultural conditions. Field-fortification allows the pesticides to interact intimately with biologically active plant matrixes. The extractability profile of the pesticide residues may be affected by absorption or translocation in the living plant tissue or by weathering on the plant in the field. Laboratory fortification of pesticides, on the other hand, may not accurately represent the rinsability profile of pesticide residues. Over the three-year period of this study, 98 paired samples were analyzed, each containing between one and five pesticide residues, as summarized in Table 1. Overall, 258 different pesticide residues were detected on the fourteen crops.

Normal parametric analysis is not suitable in this study for two reasons; the data are paired, and the resulting data are not normally distributed. The data are paired because of the assumption that the concentration of pesticide residue was the same in each paired subsample prior to rinsing. The formal null hypothesis of the test is that the population of differences in pesticide concentration between unrinsed and rinsed is symmetrical about zero (Ott, 1984). This violates the assumption of independent samples required by a t-test, and because the data are not normally distributed, the Wilcoxon signed-rank test was used. A one-tailed probability was used because rinsing cannot result in an increase of pesticide residue levels.

Table 2. Results of Wilcoxon Signed-rank Test for Significance of Rinsing in Reducing Pesticide Residues Across All Commodities

pesticides	number of pairs	significant ($P < 0.05$)	P−Value (one-tailed)	water solubility (mg/L @ 20 °C)
		insecticides		
endosulfan	60	yes	0.0025	0.32
permethrin	37	yes	<0.001	0.2
diazinon	22	yes	0.035	40
DDE	21	yes	0.001	<1
chlorpyrifos	13	no	0.32	2
methoxychlor	12	yes	0.0025	0.1
malathion	7	yes	0.008	130
bifenthrin	7	no	0.29	0.1
		fungicides		
captan	34	yes	<0.001	3.3
vinclozolin	23	no	0.095	3.4
iprodione	13	yes	0.04	13
chlorothalonil	9	yes	0.002	0.6

The second issue is choosing a nonparametric alternative to the paired t-test. This is necessary because of the large number of very small residue levels that skew the distributions toward zero. This was intentional in the experimental design as a way to mimic the actual values of pesticide residues that are common on produce. In three instances (DDE, malathion, and methoxychlor) the difference data were normally distributed according to SigmaStat, but the paired t-test result was identical to the result for the Wilcoxon signed-rank, so only those latter results are presented.

To compute the Wilcoxon signed-rank, the absolute values of the nonzero differences between unrinsed and rinsed pairs are ranked from lowest to highest, and then a sign is given the rank based on the original sign of the difference. The negative and positive ranks are then summed separately, and the lower of the values ignoring the sign is compared to a table value. In the case where all data points are positive, such as captan, the test statistic is zero, and if there are enough data points, the test will always be significant. Statistical analysis using a one-tailed Wilcoxon signed-rank test gave p values less than the null hypothesis value of 0.05, indicating that there is a significant difference between the rinsed and unrinsed samples, for nine of the 12 pesticides examined (see Table 2). Significant reductions in residue levels for the pesticides endosulfan, permethrin, diazinon, DDE, methoxychlor, malathion, captan, iprodione, and chlorothalonil were obtained through the simple act of rinsing the produce under tap water. Conversely, no significant reduction in pesticide residues was observed for chlorpyrifos, bifenthrin, or vinclozolin upon rinsing under tap water.

The paired data are presented in scatter plots for each pesticide studied (Figures 1 and 2). Each point represents a paired sample. The reference line represents the null hypothesis that rinsing does not change the concentration of pesticide residues. Points below the line represent sample pairs in which rinsing decreased pesticide residue concentration, and points at or above

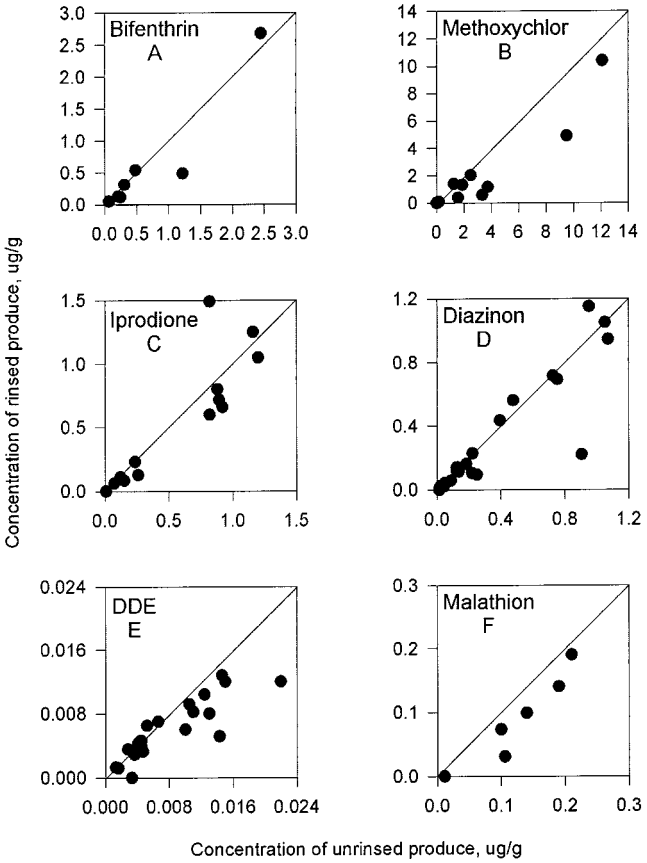

Figure 1. Scatter plots with linear axes. Bifenthrin did not decrease with rinsing, all others did.

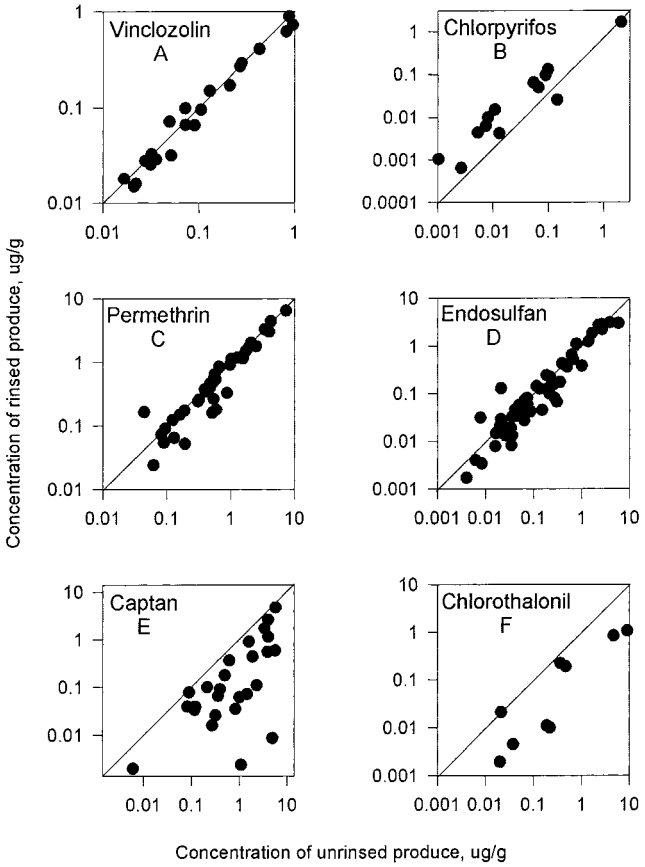

Figure 2. Scatter plots with common log axes. Vinclozolin and chlorpyrifos did not decrease with rinsing, all others did.

the line represent a sample in which rinsing did not decrease pesticide residue and/or experimental error. The regression line is not represented because it is not meaningful in a paired data analysis such as this, where the value for the unrinsed sample, *x*, is not independent from the value of the rinsed sample, *y*. Note that in some plots a logarithmic axis is used to better depict the data.

Several comments regarding pesticide water solubility and the data presented here are merited. With the exception of malathion, the water solubility of most of the pesticides examined is relatively low, as can be seen in Table 2. The water solubilities of the fungicides captan and vinclozolin are very similar, 3.3 and 3.4 mg/L, respectively. Although captan is dramatically reduced with rinsing, vinclozolin is not reduced with rinsing. Methoxychlor and bifenthrin have the same water solubility of 0.1 mg/L, yet methoxychlor is removed with rinsing, and bifenthrin is not. Last, chlorpyrifos is more water soluble than both endosulfan and permethrin, and yet it is not reduced with rinsing.

The results from the current study are consistent with an earlier study that showed residues of six pesticides on olives decreased after washing with no correlation to water solubility of the pesticides (Cabras et al., 1997). The pesticide in that study with the highest water solubility, dimethoate, decreased by 15% on the first harvested sample, while the other five pesticides decreased between 29% and 39% on the first harvested samples. Olives from subsequent harvests contained smaller residue levels, and the reduction from washing was smaller. The authors noted that dimethoate was the only systemic pesticide applied, and considering the treatment was made in a field trial on metabolically active fruit, this may account for the smaller decrease of dimethoate residues. Similarly, in another study (of plums), two pesticides, iprodione and phosalone, were shown to decrease with a five minute wash in water, wheras two other pesticides, bitertanol and procymidone, did not show a decrease (Cabras et al., 1998). A twenty-minute wash did not change the results. Both studies conclude that the water solubility is not an important factor in removal of pesticide residues from food crops. The present study also found that water solubility is not an important factor, and that the much shorter rinsing time of 30 seconds reduces many pesticide residues.

None of the pesticides examined in this study is considered a systemic pesticide (Extension Toxicology Network). Systemic is defined in the *Farm Chemicals Handbook* (Meister, 1999) as a "Pesticide that is translocated to other parts of the plant or animal than those to which originally applied." It is interesting to note that both captan and diazinon are specifically called nonsystemic, yet are described as being translocated in plants (Extension Toxicology Network). It is possible that pesticide residues are incorporated into plant tissue proportional to the time they remain on biologically active crops in the field. This may even be true of pesticides that are not specifically labeled as systemic. This underscores the requirement that studies which examine pesticide residue reduction on produce due to processing methods be conducted on field-fortified crops. One study (Ramesh and Balasubramanian, 1999) presented in the literature appears to use laboratory-fortified samples and reports a 65-95% reduction in pesticide residues. Given the present data and reports in the literature, these results should be reexamined.

The removal of surficial residue with routine rinsing is consistent with the results for DDE, the soil metabolite of DDT. DDE is known to translocate through the roots of certain plants (Pylypiw et al., 1991), and is systemic according to definition. However, in 14 of the 21 samples examined, levels of DDE in the rinsed samples were lower than in the unrinsed samples, a statistically significant effect. Possibly the unrinsed samples contained traces of soil. Therefore, the observed reduction of DDE residue with rinsing may be due to soil removal.

Additional comments on the data are merited. In our market basket survey, the most common pesticide residue found is the insecticide endosulfan. EPA tolerances for at least 38 fresh produce commodities are 2.0 ppm, including tree fruits such as apples and cherries, and leafy greens such as spinach and collards (Code of Federal Regulations, 1999). Endosulfan is a restricted-use pesticide that is considered highly toxic via the oral route, but carcinogenic (Extension Toxicology Network). The reduction in endosulfan residue (Figure 2D) was not as dramatic as that of other pesticides, but is statistically significant.

Permethrin, a synthetic pyrethroid insecticide, has a relatively high tolerance of 20 ppm on both lettuce and spinach. It has been found in our market basket survey on fresh, canned, and frozen spinach as well as other commodities at a relatively high average concentration of 0.56 μg/g. The statistics show that permethrin is removed partially by rinsing, and the scatter plot suggests the decrease is proportional to the concentration for this pesticide. Since permethrin is common on leafy greens and seems to survive commercial processing, it is notable that rinsing alone decreases residues.

Captan is the most common fungicide found in our market basket survey, and it is found in relatively high concentrations, on 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), and therefore, it was particularly interesting that captan is drastically reduced by routine rinsing under tap water, as shown in the scatter plot (Figure 2e.). Schattenberg et al. (1996) also found that captan levels are reduced on strawberries and grapes by washing with dilute surfactant, and statistical analysis of the data presented found it to be a significant effect.

CONCLUSIONS

Data presented in this study show that a short rinse in tap water reduces pesticide residues on many types of produce. This study confirms that the water solubility of pesticides does not play a significant role in the observed decrease. The majority of pesticide residue appears to reside on the surface of produce where it is removed by the mechanical action of rinsing. In addition, the data presented here indicate that studies using produce containing laboratory-fortified pesticide residues will likely generate data that do not reflect real world conditions.

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