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Dermal Exposure to Carbaryl by Strawberry Harvesters

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Dermal exposure to carbaryl by 18 strawberry harvesters was measured in the morning and afternoon on three consecutive days. Pesticide exposure was estimated from cotton gauze patches and light cotton gloves worn by the workers throughout the workday or shorter time periods. Carbaryl concentrations were determined on patches, gloves, and strawberry plants by reverse-phase high-performance liquid chromatography (HPLC). The study has revealed that during the early morning period, when there was a considerable dew deposited on leaves, carbaryl exposure was usually higher than in the afternoon. Classification of pickers by age and body weight showed that younger and/or lighter subjects exhibited lower dermal carbaryl exposure, expressed as dose rate (mg/h). The ratios of dermal exposure to dislodgeable foliar residues were within the same order of magnitude as those obtained from other pesticide–crop experiments found in the literature.

Studies on the assessment of pesticide exposure (captan and benomyl) by strawberry harvesters have been reported by Popendorf et al. (1982), Everhart and Holt (1982), Zweig et al. (1983), and, most recently, Winterlin et al. (1984). In an effort to examine possible differences in pesticide exposure by different age groups of harvesters, this study reported here was designed to examine the exposure to the insecticide carbaryl (1-naphthyl *N*-methylcarbamate) by a group of strawberry harvesters, ranging in ages from 12 to 40. These results were to be compared with those from

Table I. Physical Characteristics and Productivity of Strawberry Harvesters

ID	sex	age	wt, kg	ht, cm	surface area, m^2	daily prod. crates/h
6	F	40	69.5	168	1.77	1.04
7	F	12	42.3	163	1.52	0.68
8	F	18	63.6	175	1.78	0.91
9	F	29	56.7	164	1.58	0.99
10	F	13	43.1	149	1.35	0.78
11	F	32	61.3	175	1.72	1.00
12	F	15	59.0	160	1.62	1.00
13	F	16	50.4	173	1.56	0.72
14	M	13	63.6	178	1.76	0.97
15	M	13	70.4	168	1.78	0.57
17	F	12	45.4	157	1.42	0.74
18	F	12	49.9	163	1.52	0.60
20	M	14	56.7	180	1.70	1.04
21	F	14	54.5	165	1.59	0.60
22	F	37	49.9	152	1.46	0.89
23	F	16	49.9	165	1.53	0.96
25	M	12	45.4	160	1.43	0.70
26	M	15	63.6	183	1.84	0.72

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an exposure study of apple thinners (Maitlen et al., 1982). Ratios of dermal exposure to dislodgeable foliar residues obtained in this study will be compared with ratios obtained in exposure studies involving other crops and pesticides. If these ratios fall within a narrow range of an order of magnitude, it might be possible to apply these ratios to a first estimation of fieldworker dermal exposure by applying dislodgeable residue values without the involvement of human subjects.

EXPERIMENTAL SECTION

Description of Field Studies. A privately owned 15-acre strawberry farm near Corvallis was selected as the site of the field studies. The strawberries were of the Benton variety, and the time chosen was the first week of the 1982 harvest. The plot had been previously treated with the following pesticides: side delivery air blast sprayer at 20 gal/acre; Ronilan (vinclozolin), 50% WP, 1.0 lb of active ingredient (a.i.)/acre, three separate applications on 5, 15, and 22, 1982; Thiodan (endosulfan), 50% WP, 1.0 lb/acre, May 15, 1982; Sevin (carbaryl), 2 lb of a.i./acre, June 7, 1982; final spray solutions contained 0.5 gal of Ag-98 spreader–sticker/20 gal. Three consecutive study dates, June 22, 23, and 24, 1982, corresponded to the 15th, 16th, and 17th day postapplication with respect to carbaryl.

Eighteen volunteer workers participated in the three-day study. The physical characteristics, sex, age, height, and estimated body surface (Sendroy and Cecchini, 1954), are shown in Table I. At the beginning of day 1 and day 3 (0600 to 0800), the workers were provided with dermal monitors consisting of light cotton gloves for hands and cotton patches for forearms and lower legs. Body patches were fastened with surgical tape underneath the outer garments normally worn by the field workers, consisting of long trousers and long-sleeve shirts. As the temperature rose durng the workday, some of the workers removed their outer shirts or rolled up their sleeves, but no attempt was made to modify this behavior. On day 2, as will be explained later, gloves were put on subjects' hands about 2 h after the workers had initially entered the field.

A detailed description of patch monitors may be found in Zweig et al. (1983). Patch monitors were worn throughout the workday (up to 8 h), while morning gloves were removed after one or two crates had been harvested and brought to the weighing station (less than 2 h). After the lunch break, another set of gloves was issued to each subject. Accurate times were kept for the length of times the monitors were worn by each subject. Gloves were removed from subjects' hands by peeling them inside out in order to minimize contamination. Gloves and patches, after removal from subjects' bodies, were individually stored in plastic zip-closure bags over dry ice, transported to the laboratory, and placed in the deep-freeze until they could be extracted and analyzed.

Leaf disks were sampled with a leaf punch equipped with a 3 cm diameter circular stainless steel die, as described by Smith and Little (1954). The device functions like a pair of spring handle pliers that punches disks out of leaves and pushes them into 4-oz widemouthed glass jar that is directly attached to the punch and at the same time serves as a storage container for the leaf disks. Each action of the punch activates a mechanical counter to keep track of the number of leaf disks already collected.

The strawberry plants were sampled diagonally across the study plot every three or four rows at points that were randomly distributed in the area traversed by the field workers. The outer and inner canopy leaves of the strawberry plants from either side of the row and center were sampled until 48 leaf disks were collected. If the edge of the field was reached before the full complement of leaf disks had been collected, additional samples were taken from a new diagonal across the plot. Replicate 48-disk samples were collected for each sampling date.

Meteorological data for each day of the study was taken by us and verified by information from NOAA. The weather data were as follows:

	day 1		day 2		day 3	
	a.m.	p.m.	a.m.	p.m.	a.m.	p.m.
temperature, °C	12–21	26	16–22	27	17–26	32
wind speed, km/h	3.07		3.73		5.76	
precipitation, mm	0		0		0	
humidity, % RH	65–100	50	70	60	90	50

Sample Extraction and Analysis. Gloves and patches were thawed to room temperature and placed into a 500-mL widemouthed LP-plastic bottle fitted with a screw cap and extracted with 100 and 50 mL of acetonitrile, respectively, by shaking on a reciprocal shaker for 2 h. Ten-milliliter aliquots were filtered through a 0.22-μm Millipore filter, and the clear filtrate was directly analyzed by HPLC as, described below.

Dislodgeable foliar pesticide residues and dust were isolated from leaf punches according to methods developed by Gunther et al. (1973, 1974), Iwata (1977), and Popendorf and Leffingwell (1977). The sample is surface-extracted 3 times with 100 mL of a 60-ppb aqueous solution of dioctyl sodium sulfosuccinate (Surten), and the residue is transferred into dichloromethane. The combined organic extracts are filtered and evaporated in vacuo to dryness, and the residue is taken up in 10.0–25.0 mL of acetonitrile. Aliquots of this solution are directly analyzed for carbaryl.

Leaf dust is originally washed off the surfactant solution and remains at the interface between solvent and water in the separatory funnel. This is quantitatively transferred after the last solvent extraction to a preweighed glass filter. After the drying of the filters at 110 °C overnight, they are reweighed, and the amount of foliar dust, thus isolated, is calculated by difference in weights. Results are expressed as $\mu g/cm^2$ based on the area of a single-sided leaf disk surface (7.07 cm^2) or "ppm" of leaf dust.

Analytical Methods. Reference standard of carbaryl was obtained from the EPA Reference Standard Repository Laboratory at Research Triangle Park, NC 27711. Technical 1-naphthol was recrystallized several times from hot ethanol to remove colored impurities and until a single HPLC peak was observed (see below).

Carbaryl and 1-naphthol were analyzed by reverse-phase HPLC using a Waters 6000A solvent delivery system;, WISP automatic sample processor, Waters data module and automatic integrator, and a Model 4530 variable-wavelength detector. The column was a Waters μBondapak C_{18} reverse-phase 25 cm × 2 mm i.d. column. Carbaryl and 1-naphthol were resolved under the following experimental conditions: mobile phase, acetonitrile–water (40:60); flow rate, 2 mL/min; wavelength, 230 nm. Under these conditions, carbaryl had a retention time of 4.6 min and 1-naphthol 5.1 min. Sensitivity as limited by instrumental noise, and automatic integration was 2 ng for each compound. The injection volumes throughout these studies for samples and standards were 25–100 μL depending on the concentration of pesticide. Glove extracts usually had to be diluted 1:10 with acetonitrile, and 25 μL-aliquots were analyzed, while 100 μL-aliquots of the straight patch extract were sometimes necessary to give a detector response. When running unknowns, each sixth injection was followed by two replicated standard solutions, covering the linear range of response tested (10–250 ng).

Table II. Dermal Exposure to Carbaryl by Eighteen Strawberry Harvesters Monitored on Three Consecutive Days

anatomical region	monit. period[a]	exposure, mg/h[b]			
		day 1	day 2	day 3	mean
hands	a.m.	3.01 (1.70)	1.23 (0.62)	1.47 (0.82)	1.90 (1.38)
hands	p.m.	1.42 (0.80)	1.12 (0.73)	0.72 (0.38)	1.09 (0.71)
left hand	a.m. + p.m.	1.09 (0.59)	0.69 (0.36)	0.51 (0.25)	0.76 (0.48)
right hand	a.m. + p.m.	0.90 (0.39)	0.51 (0.28)	0.51 (0.28)	0.64 (0.37)
hands	a.m. + p.m.	1.99 (0.92)	1.20 (0.54)	1.02 (0.47)	1.40 (0.78)
forearms	all day	0.66 (0.41)	0.41 (0.27)	0.43 (0.30)	0.50 (0.35)
lower legs	all day	0.07 (0.23)	0.07 (0.13)	n.d.[c]	0.05 (0.16)
hands + arms	a.m. + p.m., all day	2.65 (1.14)	1.55 (0.61)	1.45 (0.69)	1.89 (1.00)

[a] Monit. period refers to time of day when exposure was measured. [b] Exposures are means of measurements from 18 subjects. [c] Nondetectable; numbers in parentheses are standard deviations.

Recovery studies for carbaryl and 1-naphthol were performed as follows: Known amounts of carbaryl (69–173 μg) and 1-naphthol (76–152 μg) were added to control gloves and cotton patches, which were then extracted as described above. To 100 mL of Surten solution was added 828 μg of carbaryl; this was processed as described above. All samples were analyzed by HPLC, and recoveries were found to range from 92.6% to 106.7% for gloves and patches and 84.9–97.3% for detergent solutions.

Estimation of Dermal Exposure. A detailed description for the estimation of dermal exposure of pesticides on hands and lower arms of strawberry harvesters can be found in Zweig et al. (1983). Extrapolation of pesticide concentration from patches and gloves to specific anatomical regions is made by the estimation technique using the 50-percentile man (Popendorf and Leffingwell, 1982) and a nomograph relating body weight and height to body surface (Sendroy and Cecchini, 1954). No extrapolations were needed for gloves, because they cover the subject's entire hand surface. Values are expressed in dose rates, mg/h.

Statistical Analysis. Statistical analyses were performed using SAS, a statistical computer software program developed by Statistical Analysis System, Inc.

RESULTS AND DISCUSSION

Dermal Exposure. Exposure data were assembled from the results of chemical analyses of gloves and patches worn by the subjects in this study. Exposure data for a given glove or patch is expressed as exposure rate (mg h^{-1} $person^{-1}$). Exposure rates are calculated by dividing the carbaryl concentration found on an individual's monitor by the length of time (in hours) that the monitor was worn by that individual during a given observation period. There were six observation periods for gloves, namely, mornings and afternoons on days 1, 2, and 3. For body patches there were three observation periods, the entire work period on the three days of the study. All of these data were used in the statistical analyses described below. A summary of these data is presented in Table II. Raw data for each individual worker for the dermal exposure to carbaryl are available in the supplementary material (see paragraph at end of paper regarding supplementary material).

Popendorf et al. (1982), when studying captan exposure by strawberry harvesters, demonstrated that the principal areas of exposure occurred on the hands, forearms, and lower legs. For this reason, the very same anatomical regions were monitored in the present study. Furthermore, since 1-naphthol was not found in any of the field samples, all results will be presented only for carbaryl exposure.

As shown in Table II, carbaryl exposure to lower ankles amounted only to 0.07 mg/h for days 1 and 2 and nondetectable for day 3 or no more than 4% of total body exposure. Only 1 subject out of 18 exhibited considerably higher ankle exposure than the rest of the group (1.04 and 0.79 mg/h for days 1 and 2, respectively). This unusually high ankle exposure appears to be an aberrant case and may have been due to high saturation of subject's pants legs from heavy morning dew and a concomitant contamination of the lower leg patch, worn underneath the outer garment.

We consider, therefore, dermal exposure to carbaryl by strawberry harvesters to be mainly on hands and forearms and, to a much lesser degree, on the lower legs. This finding is in contrast to observations on captan exposure by Winterlin et al. (1984), who reported that thigh and shin exposure amounted to more than half of total body exposure. The difference in observations may be due to the respective experimental protocols in the two studies. As already mentioned, our protocol directs that the body patches be worn underneath the work clothes so that the monitors would measure exposure to the skin rather than "outer" exposure as described by the other researchers. Another difference between these two studies is that we prescribe light cotton gloves to serve as hand monitors, while Winterlin et al. (1984) used latex gloves. This may account for other differences in our observations as will be discussed below.

Table II also shows that gloves worn on day 1 had a higher carbaryl concentration than gloves worn the other 2 days. Dose rates for hands on day 1 and day 3 were also significantly higher in the morning than afternoon ($T = 3.5$; df = 34; $p < 0.01$). No difference between a.m. and p.m. hand exposure was found on day 2. It was on day 2 that gloves were issued to the workers after most of the dew had dried off the strawberry leaves. We observed especially on day 1 that early morning dew had caused the glove monitors to become quickly saturated with mositure mixed with fruit juice, as exhibited by the red color of the gloves. One may speculate that wet gloves exhibit a different behavior toward absorption of dislodgeable foliar residues and may no longer possess a linear absorptive capacity. Contrary to this speculation, however, it was found that the highest concentration of carbaryl occurred on day 1 when the morning humidity had reached the saturation point, and one could clearly observe a large amount of dew on the leaf surface. One possible explanation for this finding might be that dislodgeable pesticide residues are more efficiently transferred from the leaf to glove surface in a homogeneous, aqueous phase. This phenomenon may be unique for crops like strawberries with delicate texture, easily damaged during harvesting, especially by inexperienced pickers. Popendorf and Leffingwell (1982) theorize that dermal exposure in tree crops may be the result of indirect transfer of dry dislodgeable foliar residues present on dust to the body of harvesters.

Table III. Dermal Hand Exposure According to Age and Body Weight of Strawberry Harvesters

type of exposure	variable	N	exposure, mg/h	Kruskall-Wallis X^2	p
right hand, means, 3 days	youths (≤14)	21	0.54	4.94	0.026
	adults (≥15)	33	0.74		
left + right hands, p.m., means, 3 days	<50 kg	21	0.80	5.87	0.015
	>50 kg	33	1.27		
left + right hands, p.m., day 1	<50 kg	7	0.88	$T^a \cong -3.10$	
	>50 kg	11	1.76	df ≅ 14.4	
				$p = 0.0076$	

[a] Unequal variances.

Table IV. Dislodgeable Foliar Residues of Carbaryl on Strawberry Leaves[a]

sample i.d.	days post-harvest	dislodgeable residues µg/cm²	dislodgeable residues ppm (dust) × 10³
1.1	1	7.74	50.46
1.2	1	8.41	65.09
3.1	3	3.36	57.10
3.2	3	2.98	56.53
3.3	3	1.35	16.45
7.7	7	1.32	22.55
14.1	14	0.56	8.30
14.2	14	0.51	12.43
15.1	15	0.77	16.06
15.2	15	0.45	6.53
16.1	16	0.41	9.42
16.2	16	0.69	14.57
17.1	17	0.32	2.92
17.2	17	0.15	1.67

[a] Pesticide treatment: 2 lbs of carbaryl/acre (Sevin 4F, flowable) on June 7, 1982.

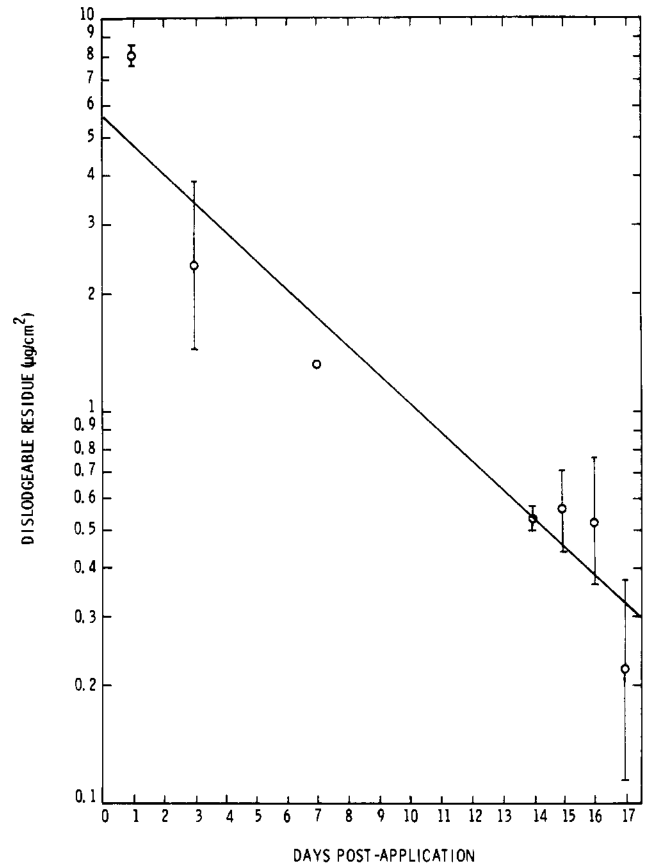

Figure 1. Carbaryl dislodgeable foliar residue decline curve; strawberry leaves; duplicate samples were taken, as explained in the text, on specific days starting with the first day after pesticide application; dislodgeable residues, expressed in µg/cm², are plotted on a log scale; experimental data are found in Table IV of the text; $y = 0.755 - 0.0732x$; $r = -0.925$.

Comparison of Dermal Exposure by Age and Body Weight. U.S. Government regulations forbid 10–11-year-old children to work in fields that have been sprayed with pesticides suspected to be carcinogens (*Fed. Regist.*, 1979). Therefore, one of the goals of this study was to determine if the age of strawberry harvesters affected dermal exposure to pesticides. Since the group of strawberry harvesters in this study did not contain any 10–11-year-olds (see Table I), the group was arbitrarily divided into two other groups, "youths" (≤14 year of age) and "adults" (≥15 years of age). As is seen in Table III, a marginally significant difference in dermal exposure for the right hand was observed, namely, that the right-hand exposure of the youths group was lower than the corresponding exposure by adults. No other significant differences between these two groups were found.

When other variables were substituted for age, e.g., body weight, height, and body surface, the only difference was seen when the group was divided according to weight; pickers weighing less than 50 kg (presumably younger subjects) had lower afternoon hand exposure than those who weighed more than 50 kg. No differences were found for height (< 165 cm and >165 cm) and body surface (< 1.50 m² and >1.50 m²).

Productivity, Age, and Exposure. Productivity, expressed as average crates harvested per hour, of each worker may be seen in Table II. The productivity of the workers in this study appears to be much lower than was found in an earlier study carried out in California (Zweig et al., 1983). The larger size of the crates used in Oregon compared to the ones in California (Zweig et al., 1983) might partially account for the difference in productivity. Another plausible explanation might be the makeup of the two harvester groups; the one in the earlier study in California was composed of 10 highly motivated and experienced pickers, whereas the group in this study were mostly school children who were less experienced, especially since the study commenced during the first week of the Oregon strawberry harvest.

It can be shown that morning, afternoon, or daily productivity was not different for the 3 days of the study (Kruskal-Wallis $X^2 = 0.63–4.18$; df = 2; $p > 0.1$). When examining youths and adults, however, it was found that daily productivity of youths was lower than that of the adults (0.72 vs. 0.89 crates/h; $F = 11.56$; df = 1,48; $p = 0.0014$). This is in agreement with the finding that daily productivity was positively correlated with age ($N = 51$; $r = 0.458$; $p = 0.0007$).

No significant correlation was found for productivity and dermal exposure on any individual day or all 3 days together. This finding is in agreement with the results obtained from a similar study with captan but is contrary to the finding that benomyl dermal exposure did correlate with productivity (Zweig et al., 1983). Further studies are

Table V. Ratios of Dermal Exposure to Dislodgeable Foliar Residues

pesticle	crop	activity	$R \times 10^{-3}$	lit.
carbaryl (day 1)	strawberries	harvesting	4.34	this study
carbaryl (day 2)	strawberries	harvesting	2.82	this study
carbaryl (day 3)	strawberries	harvesting	6.17	this study
captan	strawberries	harvesting	8.57	Zweig et al. (1983)
captan	strawberries	harvesting	2.90	Popendorf et al. (1982)
captan	strawberries	harvesting	8.00	Popendorf et al. (1982)
captan	strawberries	harvesting	2.62	Popendorf et al. (1982)
captan	strawberries	harvesting	5.97	Popendorf et al. (1982)
captan	strawberries	harvesting	4.73	Popendorf et al. (1982)
benomyl	strawberries	harvesting	7.19	Zweig et al. (1983)
benomyl	apples	thinning	0.5	Maitlen et al. (1982)
chloro-benzilate	citrus	harvesting	5.33[a]	Nigg et al. (1984)
OP com-pounds	citrus	harvesting	5.1[b]	Popendorf and Leffingwell (1982)
OP com-pounds	peaches	harvesting	1.9[c]	Popendorf and Leffingwell (1982)

[a] Corrected for one side of the leaf surface. [b] Geometric mean from 14 observations. [c] Geometric mean from nine observations.

needed to verify if such a correlation between dermal exposure and the number of fruit picked does exist.

Dislodgeable Foliar Residues. Table IV is a summary of results of analyses of dislodgeable foliar residue samples taken at various times after the last application of carbaryl up to and including the 3 study days. The plot of log concentration vs. time, as seen in Figure 1, resulted in a straight line, demonstrating that the decline of foliar dislodgeable residues follows first-order kinetics. The half-life of carbaryl on strawberry leaves is 4.1 days, as calculated from these data.

The ratio for dermal exposure rate and dislodgeable foliar residue can be calculated for each day of the study (days 1, 2, and 3 of the study are identical with days 15, 16, and 17 of the decline curve). Table V lists these ratios as well as ratios taken or calculated from the literature. These ratios are reasonably similar, given the range of chemicals and crops studied in which the dose distribution and presumably the exposure mechanism might be quite different. The low value obtained by Maitlen et al. (1982) for apple thinners exposed to carbaryl may be due to the fact that the only data available to calculate the ratio was total and not dislodgeable foliar residues, which would lead to a smaller number.

It might be possible to calculate the dermal exposure of fieldworkers, as a first approximation, by multiplying the dislodgeable foliar pesticide concentration by this empirical factor (approximately 5×10^3). Further studies should demonstrate if this simplified technique might be feasible for practical purposes in establishing reentry intervals for fieldworkers exposed to different pesticides during the harvesting operation of row and tree crops.

Registry No. Carbaryl, 63-25-2.

Supplementary Material Available: Raw data for each individual worker for the dermal exposure to carbaryl (16 pages). Ordering information is given on any current masthead page.

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