

DERMAL AND URINARY MONITORING OF PEACH AND APPLE HARVESTERS EXPOSED TO
ORGANOPHOSPHATE RESIDUES IN SUTTER, STANISLAUS AND MADERA COUNTIES,
1989 and 1990

by

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HS-1577 September 1, 1993

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INTRODUCTION

Estimates of occupational exposure to pesticide residues are a critical component of relative risk assessment. The California Department of Pesticide Regulation (DPR), Worker Health and Safety Branch (WH&S), conducts pesticide exposure studies as a means to evaluate the effectiveness of reentry intervals, to develop exposure assessments for incorporation in risk assessments and to develop worker monitoring strategies. This paper summarizes two years' exposure of peach and apple harvesters to azinphos-methyl and phosmet as measured by dermal monitoring and excretion of urinary dimethyl phosphate metabolites. The study was conducted to characterize tree fruit harvester exposure to organophosphate insecticides and to investigate urinary metabolite excretion as an exposure modeling strategy. Blood cholinesterase monitoring was also conducted.

Harvester exposures have historically been difficult to measure since dermal exposure assessment is technically complex. Foremost among these difficulties are uncertainties about the kinetics and estimation of absorbed residues. The traditional method of using gauze patch dosimeters gives data that is a poor predictor of exposure in humans (Franklin, 1984). Biological monitoring provides an indirect and complementary means of addressing exposure. Franklin and co-workers reported a strong linear correlation between urinary alkyl phosphate levels and both dermal doses of azinphos-methyl in rats (Franklin et al., 1986) and amount of pesticide sprayed by orchard applicators (Franklin et al., 1981). In this study, dislodgeable foliar residue data ($\mu\text{g}/\text{cm}^2$) are reported as an environmental indicator of worker exposure. WH&S has previously investigated residue degradation for azinphos-methyl and phosmet and mixer/loader/applicator exposure to azinphos-methyl. (Formoli and Fong, 1993; Maddy et al., 1977).

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MATERIALS AND METHODS

Location and Crop Characteristics

Tree fruit pickers at five sites in California's Central Valley were monitored in 1989 and 1990. Table I presents a study outline and summary of cultural practices for each site. Peach harvesters were monitored in Sutter County in 1989 and 1990 and in Stanislaus County in 1989. The peaches at both locations were picked for processing. Trees at the Sutter County site ranged from six to twenty years old and those in Stanislaus County were about twenty years old. The younger trees had considerably fuller and denser foliage than the older trees and much of the fruit was obscured within the foliage. The older trees had a very open canopy.

Apple harvesters were monitored in Madera County in 1989 and 1990. The apples were Granny Smith and were picked for fresh market. The trees were supported on cross-wires to form a hedgerow that was flat and narrow. The fruit stood out prominently from this hedgerow.

Treatments

Workers at all sites were exposed to azinphos-methyl (Guthion[®] 50WP) residues. Peaches were treated once each season with azinphos-methyl while apples were treated five times each season. All acreage at the Stanislaus and Madera County sites was treated, while only 40% - 50% of the orchards in Sutter County were treated.

Work in phosmet-treated fields (Imidan[®] 30WP) occurred on all study days in Stanislaus County (1989) and on the first study day in Madera County in 1990 (day 25 post-application). The peaches in Stanislaus County were treated once with phosmet and the apples in Madera County, 1990, were treated twice. Approximately 10% of the orchards at these two sites were treated.

Worker Characteristics

The crews were male and consisted primarily of pickers, but some members performed other tasks including sorting, fruit hauling, supervising or irrigating. All crews spoke Spanish as their primary language. The typical work attire consisted of a long-sleeved buttoned shirt worn over a short-sleeved T-shirt, long pants, tennis shoes, socks and a baseball cap.

Task Characteristics

All harvesters used ladders to reach the fruit. Workdays at all sites were 8 hours except for apple harvesters in Madera County, 1990, who worked 10 hours each day. Work histories for the study period were obtained from the crew boss at each site.

Peach harvesting at the Sutter and Stanislaus County sites took place from mid-July to early September, spanning about six weeks. Harvesters in Stanislaus County were exposed to organophosphate residues daily and in Sutter County, for about 40% - 50% of the season, as not all acreage was treated. Peach harvesters reached into the tree to pick the fruit and a worker was sometimes immersed in the foliage from head to knee.

On the first study day in Sutter County in 1989 (day 31 post-application, Table I), the crew performed thinning and propping tasks. Thinning required the workers to remove excess fruit to allow the remaining fruit to achieve greater size. The work is similar to harvesting and involved using either the hands or a pole with a hook at one end to pull off the fruit. Some of the work was performed from a ladder. The workers had somewhat less contact with the foliage than when harvesting. Propping required leaning 1" x 6" x 14' boards against a tension wire strung near the tree top to support limbs heavy with fruit. The work involved little contact with the tree foliage. Thinning, propping and pruning tasks were performed by all peach harvester crews intermittently throughout the season. However, the majority of thinning was done from April - June, prior to field treatment.

All apple harvesters were required to wear nylon knit gloves while picking to maintain the fruits' "bloom" or natural waxy, powdery coating. Apple harvesters contacted the treated foliage primarily with the hands and arms. They did not have extensive full-body contact with the foliage because the apples were easily accessible in the hedgerowed trees. Additionally, the foliage was sparser than in the peach orchards. Since the tree branches were trained along wires, no propping was necessary. Harvesters did not thin fruit during the harvest season.

Table I. Summary of Cultural Practices and Study Outline

County/ Crop	Study Date	Pesticide, Application Rate ^{/a}	Task	Post- Application Day	Dermal Exposure n Workers	Urine Collection n Workers	Blood Draws n Workers
Sutter Peach	7/20/89	Guthion ^{/b}	NE ^{/c}				19
	7/26/89	50WP,	Prop	31*	6	6	
	7/26/89	1.5	Thin	31*	4	10	
	7/27/89		Pick	32*	10	16	
	7/28/89		Pick	33*	10	16	19
	9/4/89		Pick	70			13
Sutter Peach	8/16/90	Guthion 50WP,	Pick	52*	11	25	17
	8/17/90	1.5	Pick	53*	11	24	
	8/18/90		NE	54*		24	
	8/19/90		NE	55*		24	
	8/20/90		NE	56*		7	
	8/23/90		Pick	59			17
	9/4/90		Pick	71			11
Stanislaus Peach	8/21/89	Guthion 50WP,	Pick				8
	8/22/89	0.75 and	Pick	60/34*	9	9	
	8/23/89	Imidan ^{/d} , 30WP,	Pick	61/35*	9	9	
	8/24/89	3	Pick	62/36*	9	9	8
Madera Apple	9/19/89	Guthion 50WP,	Pick	42*	10	10	10
	9/20/89	5 apps. at	Pick	43*	10	10	
	9/21/89	1.5-2	Pick	44*	10	10	10
Madera Apple	9/11/90	Guthion 50WP, 5 apps. at 2 & Imidan 30WP, 1 app. at 3	Pick	41/25*	8	9	13
	9/12/90	Guthion 50WP,	Pick	23*	8	10	
	9/13/90	5 apps. at 2	Pick	24*	8	10	
	9/25/90		Pick	36			7

Only dermal and urinary exposure monitoring days have exposure information available

* DFR samples taken

/a lb. active ingredient (a.i.) per acre

/b azinphos-methyl

/c No organophosphate exposure on this work day

/d phosmet

Exposure Monitoring

After receiving study approval from the University of California, San Francisco, Human Subjects Review Committee, an interpreter explained the procedures and solicited the workers' voluntary cooperation. Dislodgeable foliar residue (DFR) sampling, dermal exposure monitoring and urinary dimethyl phosphate and blood cholinesterase monitoring were conducted at each site. Table I gives the number of total

participants for each part of the study. Dermal exposure samples, dimethyl phosphates, creatinine and DFR were analyzed by California Department of Food and Agriculture (CDFA) Chemistry Laboratory Services, Sacramento. Blood samples were analyzed for erythrocyte and plasma acetylcholinesterase using the Ellman method (1961). For all sites, analyses were conducted by Roche Biomedical Laboratories. For the Sutter County sites (1989 and 1990), duplicate samples were analyzed by University of California, Davis.

Dislodgeable Foliar Residues (DFR)

The orchards were sampled each study day for DFR using the methods of Gunther et al. (1973), according to the schedule detailed in Table I. Samples were taken from 10 trees in each of 3-5 locations within each orchard, depending on the size of the orchard, at a height of 5 - 6 feet. Sample jars were sealed with aluminum foil, capped and kept on ice for shipment to the laboratory. In addition to the DFR samples taken on study days, six orchards in Sutter County were sampled several times in the first week post-application, then weekly for 7-11 weeks, to allow characterization of azinphos-methyl decay and estimation of half-life (Fig. 1).

Dermal Monitoring

Dermal exposure was measured by clothing dosimeters that were worn by each worker for the entire workday. The sole exception was Madera County apple harvesters, who did not wear clothing dosimetry on day 1 in 1989 (day 42 post-application). Workers were provided with new, 100% cotton, long-sleeved, white, knit shirt (Health Knit[®]) each monitoring day. They wore the shirts next to the skin under a regular cotton work shirt. The shirts covered the hip region and were tucked into the workers' trousers. In 1989, workers also wore a pair of knee-length, 80% cotton/20% orlon athletic socks. At the end of the monitoring period, clothing dosimeters were stored in separate one-gallon Ziploc[®] bags.

Hand residue samples were obtained from the peach harvesters by wipes followed by a wash. Each worker wiped his hands with two pre-moistened disposable wipes (Chubs[®]) which were combined for analysis. Workers then washed their hands for one minute in 500 mL of 1% sodium dioctyl sulfosuccinate contained in a one-gallon plastic bag. The apple harvesters wore nylon knit harvester gloves as standard clothing. Their gloves were collected and placed in a one-gallon Ziploc[®] bag. Their hand residue sample consisted of two sequential wipes of ungloved hands. Face and neck residues were obtained by wiping these regions with two pre-moistened disposable wipes that were combined for analysis. Wipes were stored in four-ounce glass jars and hand wash solution in 0.5-liter Nalgene[®] bottles. All dermal exposure samples were frozen until extraction.

Urine Monitoring

Each worker was provided with three one-liter polyethylene urine collection bottles each day. Workers were instructed to collect all urine for the 24-hour period. Daily volumes were recorded and a 100-mL aliquot was stored in a 250-mL polyethylene bottle for shipment to the laboratory.

Cholinesterase Monitoring

Blood draws, taken by licensed phlebotomists at each field site from both exposed workers and unexposed worker controls, provided an exposure index (Table I). For Stanislaus and Madera Counties, there were two blood draws, one at the beginning of the study and one at the end. In Sutter County, there were three draws each year, one at the beginning of the study, one at the end and the third draw two to six weeks post-study, at the end of the harvest season. Initial draws provided a study baseline. In Sutter County, 1989, the first blood draws were taken prior to organophosphate residue exposure. For all other sites, initial blood draws were taken early in the workers' seasonal exposure period. The end-of-study draws provided a short-term exposure index; post-study draws in Sutter County provided mid- and late-season exposure indices. The analyses of duplicate samples from Sutter County by the clinical and University laboratories allowed for independent examination of laboratory artifact, which often confounds cholinesterase results.

Sample Analysis

Leaf discs were shaken three times with 50 mL sodium dioctyl sulfosuccinate solution. This aqueous solution was extracted three times using 50 mL ethyl acetate which was then dried by the addition of sodium sulfate (Gunther et al., 1973). After volume reduction the samples were analyzed by gas liquid chromatography. Socks, shirts, hand and face/neck wipe extracts were analyzed similarly. Prior to their distribution to study subjects, all shirts had undergone two hot water wash cycles to remove potentially interfering fabric and finish additives. Hand washes were extracted using ethyl acetate, which was dried with anhydrous sodium sulfate and diluted as necessary for analysis. Azinphos-methyl and phosmet were co-analyzed on a Hewlett-Packard 5880A chromatograph equipped with a phosphorus detector. The chromatographic conditions were: column, 10m x 0.53 mm HP 50% phenyl methyl silicone; carrier gas (He), 20 mL/min; H₂, 4 mL/min; air, 90 mL/min; injector and detector temperature, 250 °C; oven temperature, 240 °C isothermal. Using these conditions, the retention times were 6.00 and 4.40 minutes for azinphos-methyl and phosmet, respectively, and 4.89 and 3.63 minutes for azinphos-methyl oxon and phosmet oxon, respectively. Minimum detectable levels for azinphos-methyl and phosmet in µg/sample were 5, 2, 1, 1, 1 and 0.25, for the shirts, socks, wipes, hand washes, gloves and dislodgeable foliar residues, respectively. The corresponding minimum detectable levels for the oxons were 10, 5, 1, 2, 2 and 0.5.

Urine samples were analyzed for dimethyl phosphate metabolites and creatinine. Dimethyl phosphates [dimethylphosphate (DMP), dimethylthiophosphate (DMTP), dimethyldithiophosphate (DMDTP)] were determined (Weisskopf and Seiber, 1989) using a Varian 6000 gas chromatograph equipped with a flame photometric detector in phosphorus mode. The chromatographic conditions were: column, 30 x 0.53 mm DB 1 (J&W Scientific); gas flows were: carrier (He): 4.5 mL/min, H₂: 150 mL/min, air #1 80 mL/min, air #2 170 mL/min and make-up He 30 mL/min. The injector temperature was 310 °C and column temperature was programmed 130 °C - 210 °C. Under these conditions the retention times for DMP and DMTP were 2.32 and 4.07 minutes, respectively. The minimum detectable level was 0.05 ng per injection which translated to 25 ppb in urine for all dimethyl phosphates. Analyses for creatinine were conducted on a Technicon[®] AutoAnalyzer II, using clinical method number 11 (Technicon Instruments Corp., 1972).

Data Analysis

DFR results for the parent and the oxon for each pesticide were summed for each study day and daily means reported. For the Sutter County orchards, linear regression was performed on the log₁₀ of residues vs. day post-application. The regressions were tested for significance at the 0.05% level. A half-life was calculated for each orchard using the rate constant from the regression equation (Snedecor and Cochran, 1973).

Individual daily dermal exposure (DE) was calculated by summing the contribution of the thion and oxon of each pesticide for each dermal medium. While crew bosses participated in the study, their exposure was limited and analyses were restricted to workers performing harvest tasks. Daily group means for each site, by pesticide, were calculated from the individual data and reported. DE for Madera County, 1989, day 1, was estimated as the mean of DE for days 2 and 3. Gloves were analyzed for the Madera County site, but only hand wipe residues collected after glove removal were included in calculations of DE. Glove residues are presented separately in Table VI.

Daily excretion of dimethyl phosphates was used to indirectly estimate absorbed dose. Analyses were restricted to harvest workers with daily urine volumes greater than 100 mL. Absorbed dose (mg urinary pesticide equivalents) was calculated by multiplying urinary metabolites (mg/L) by sample volume (L) and the ratio of the molecular weight of the pesticide to the molecular weight of each metabolite. Daily group means for percent pesticide absorbed [urinary pesticide equivalents/(dermal exposure + urinary pesticide equivalents) x 100] were calculated from the sum of the individual ratios. For four of the five sites, urine collections were concurrent with dermal exposure monitoring. For Sutter County, 1989, urine collections continued through day 5 following dermal exposure monitoring on days 1 and 2. Excretion of metabolites over the five days reflects exposure on day 1 and 2. Linear regression was performed on daily urinary

pesticide equivalents (UPE) vs. DFR and cumulative UPE vs. cumulative DE. Creatinine analyses gave a relative index of the degree of compliance with urine collections, however, results were not adjusted. Statistical significance was defined as $p < 0.01$ (Snedecor and Cochran 1973).

The results of cholinesterase monitoring for workers and control individuals were compared by paired t-tests to determine whether a significant ($p < 0.05$) drop in worker plasma or RBC cholinesterase levels occurred following exposure to organophosphate residues. When three blood draws were conducted, cholinesterase levels were first compared by ANOVA.

RESULTS

Sample Fortification

Recovery efficiencies for technical azinphos-methyl and the oxon were determined from dermal media fortified in the field. Fortification recoveries of azinphos-methyl were $80 \pm 11\%$, $97 \pm 7\%$, $90 \pm 9\%$, $102 \pm 15\%$ and $53 \pm 34\%$ for socks, shirts, wipes, gloves and hand washes, respectively. The corresponding fortification recoveries for the oxon were $69 \pm 14\%$, $89 \pm 31\%$, $81 \pm 9\%$, $103 \pm 19\%$ and $90 \pm 15\%$. Blank samples had no detectable azinphos-methyl or oxon. Mean recovery efficiencies for field fortifications of unexposed study staff urine samples with the dimethyl phosphates DMP and DMTP were $80 \pm 18\%$. Blank samples had no detectable DMP or DMPT. Results were not adjusted for recoveries.

Dislodgeable Foliar Residues (DFR)

Study means for azinphos-methyl, phosmet and their respective oxons are given in Table II and daily means in Table III. Azinphos-methyl DFR were similar for the Sutter and Madera County sites in 1989 and 1990 (0.46 - $0.63 \mu\text{g}/\text{cm}^2$) and about 20 times greater than levels found at the Stanislaus County site ($0.026 \mu\text{g}/\text{cm}^2$). Six orchards in Sutter County were monitored to characterize azinphos-methyl decay. The estimated half-lives ranged from 18.5 - 43 days. The regression lines for the \log_{10} residues vs. days post-application are plotted in Figure 1 ($r^2 = 0.50$ - 0.73 , $n = 6$). The regression lines were not coincident; both the slopes and intercepts differed significantly ($p > 0.05$). Phosmet residues in Stanislaus County, 1989, were about twice those in Madera County, 1990 ($2.48 \mu\text{g}/\text{cm}^2$ vs. $1.35 \mu\text{g}/\text{cm}^2$). In Madera County, the phosmet residues were about twice those for azinphos-methyl ($1.31 \mu\text{g}/\text{cm}^2$ vs. $0.56 \mu\text{g}/\text{cm}^2$), while in Stanislaus County, phosmet was present at nearly 100 times the azinphos-methyl residues ($2.48 \mu\text{g}/\text{cm}^2$ vs. $0.026 \mu\text{g}/\text{cm}^2$). The oxon for the respective pesticides was detected at about half the study sites and averaged 2.4% of the thion residue.

Table II. Dislodgeable Foliar Residues (DFR) During Harvester Monitoring Studies, Means \pm SD, $\mu\text{g}/\text{cm}^2$

County, Year	Crop	Azinphos-methyl	Azinphos-methyl oxon	Phosmet	Phosmet oxon
Sutter, 1989	Peach	0.59 ± 0.23	0.011 ± 0.004	NA	NA
Sutter, 1990	Peach	0.46 ± 0.18	0.008 ± 0.002	NA	NA
Stanislaus, 1989	Peach	0.026 ± 0.053	ND	2.48 ± 0.52	ND
Madera, 1989	Apple	0.63 ± 0.09	ND	NA	NA
Madera, 1990	Apple	0.56 ± 0.4	0.015 ± 0.007	1.31 ± 0.04	0.045 ± 0.009

NA Not Applied

ND Not Detected, below minimum detection level

Table III. Mean Daily Harvester Exposure Levels to Azinphos-methyl and Phosmet Residues at Five Sites, 1989-90

County, Pesticide	Study Day	DFR ($\mu\text{g}/\text{cm}^2$)	mg DE ^{/a} Mean	DE SD	DE (N) ^{/b}	mg UPE ^{/c} Mean	UPE SD	UPE (N) ^{/d}
Sutter, 1989								
AZ ^{/e}	1 ^{/f}	0.50	0.70	0.65	6	0.56	0.38	6
	1 ^{/g}	0.49	13.00	2.82	4	1.92	0.92	4
	2	0.66	15.62	3.78	10	3.00	1.25	16
	3	0.62	15.47	4.97	10	3.74	1.02	16
Sutter, 1990								
AZ	1	0.36	12.02	3.04	11	1.06	0.64	24
	2	0.61	14.04	4.06	11	1.86	1.02	22
	3	NE ^{/h}				0.52	0.40	21
	4	NE				0.30	0.36	13
	5	E ^{/i}				0.18	0.10	6
Stanislaus, 1989								
AZ	1	0.009	0.44	0.29	8	14.17	6.03	7
	2	0.011	1.25	1.36	9	9.27	5.06	9
	3	0.07	4.30	3.97	8	16.03	5.89	8
PM ^{/j}	1	2.5	28.17	8.09	8	/k	/k	/k
	2	2.5	31.57	7.71	9	/k	/k	/k
	3	2.5	39.27	6.00	8	/k	/k	/k
Madera, 1989								
AZ	1	0.59	1.84/l	NS ^{/m}	NS	0.89	0.81	9
	2	0.70	2.02	0.90	9	0.71	0.44	9
	3	0.58	1.66	0.81	9	1.32	0.64	9
Madera, 1990								
AZ	1	0.32	1.51	0.86	8	10.15	6.70	9
	2	0.55	6.52	3.14	7	5.86	1.49	8
	3	0.79	6.46	3.21	8	8.80	3.66	9
PM	1	1.40	4.00	3.20	8	/k	/k	/k

/a Dermal Exposure = sum of hand, shirt, sock and face/neck dosimetry

/b Analyses restricted to harvest workers

/c Urinary Pesticide Equivalents = μg metabolites(MW parent/MW metabolite)

/d Analyses restricted to harvest workers providing 24-hour urine volumes of > 100 mL

/e Azinphos-methyl

/f Workers performed propping tasks

/g Workers performed thinning tasks

/h No organophosphate exposure

/i Exposed to AZ residues

/j Phosmet

/k UPE (dimethyl phosphate metabolites) from tandem exposure to azinphos-methyl and phosmet

/l Not sampled; mean of Day 2 and 3 exposure

/m Not sampled

Dermal Exposure (DE)

The mean daily DE for each pesticide at each site is given in Table III. The average daily DE to azinphos-methyl ranged from 1.7 ± 0.8 mg for apple harvesters in Madera County, 1989, to 15.6 ± 3.8 mg for peach harvesters in Sutter County, 1989. In Sutter County, 1989, DE measurements while thinning peaches (~13 mg/day) were about 85% of those measured for harvesting, while propping (~0.7 mg/day) averaged less than 5% of the DE for harvesting. Daily DE to phosmet ranged from about 4 mg in Madera, 1990, to nearly 40 mg in Stanislaus County, 1989. The coefficient of variation for the dermal data was about 50%.

Harvester exposure to either pesticide present at similar DFR levels resulted in lower dermal exposures for apple harvesters compared to peach harvesters. Apple harvesters exposed to azinphos-methyl residues in Madera County, 1989-90, (DE = 1.5 - 6.5 mg) received 2-10 times less exposure than did peach harvesters in Sutter County, 1989-90 (DE = 12 - 15.6 mg). Similarly, for phosmet, while the DFR in Madera, 1990, was one-half that in Stanislaus County ($1.31 \mu\text{g}/\text{cm}^2$ vs. $2.48 \mu\text{g}/\text{cm}^2$), mean DE for apple harvesters (~4.0 mg) was about 1/8 that of peach harvesters (~33 mg).

Exposure potential can also be examined by comparing dermal transfer factors specific to task, site and residue level. Nigg et al. (1984), Pependorf et al. (1979) and Zweig et al. (1984, 1985) were among the first to use an empirically-derived dermal transfer factor to describe the rate of residue transfer to a worker performing a particular work task. Transfer factors (TF) are expressed as units of hourly exposure ($\mu\text{g DE}/\text{hr}$) per unit of DFR ($\mu\text{g}/\text{cm}^2$), giving units of cm^2/hr . Zweig et al. (1984) suggested a TF of 5,000 cm^2/hr for harvesting fruit crops. TF for the present study (calculated using DE and DFR values from Table III) are given in Table IV and range from 175 - 9332 cm^2/hr . TF for apple harvesters are an order of magnitude less than for peach harvesters. Exposure potential for peach propping is about 5% of that for thinning and harvesting. The TF for AZ exposure of peach harvesters in Stanislaus County, 1989, is likely skewed by the extremely low DFR values (0.009 - 0.07 $\mu\text{g}/\text{cm}^2$). The plot of hourly dermal exposure on DFR for all sites and tasks is given in Figure 2. The slope of this regression line (1485 cm^2/hr) is the composite transfer factor for this study. The magnitude of the regression coefficient ($r^2 = 0.71$) is strongly influenced by the Stanislaus County phosmet residues.

Table IV. Transfer Factors (TF) (cm^2/hr) for Tree Fruit Harvesters at Five Sites, 1989-90

County/Year	Crop	Task	Pesticide	n Days	TF
Sutter, 1989	Peach	Propping	AZ ^{/a}	1	175
		Thinning	AZ	1	3316
		Harvest	AZ	3	3038
Sutter, 1990	Peach	Harvest	AZ	3	3526
Stanislaus, 1989	Peach	Harvest	AZ	3	9332
		Harvest	PM ^{/b}	3	1651
Madera, 1989	Apple	Harvest	AZ	2	359
Madera, 1990	Apple	Harvest	AZ	3	468
		Harvest	PM	1	286

/a Azinphos-methyl

/b Phosmet

Regional Exposure Distribution

Shirts: Exposure to the shirt region (torso + arms) was by far the largest component of dermal exposure (Table V). For all sites combined, the shirt contributed $77 \pm 12\%$ to DE. The contribution of the shirt to DE is most consistent by site, even when the two pesticides are present at disparate DFR (Stanislaus, 1989) or when monitoring differs by year (Sutter and Madera Counties).

Table V. Regional Exposure Distribution (Percent)

County, Pesticide	Shirt	Hand Total	Handwipe	Handwash	Face/Neck	Socks
Sutter, 1989 ^{/a}						
AZ ^{/b}	66	31	21	10	2	1
Sutter, 1990 ^{/a}						
AZ	57	42	26	16	1	NS ^{/c}
Stanislaus, 1989 ^{/a}						
AZ	75	24	17	7	<1	1
PM ^{/d}	74	23	16	7	2	1
Madera, 1989 ^{/e}						
AZ	88	6	6	NS	4	2
Madera, 1990 ^{/e}						
AZ	91	7	7	NS	2	NS
PM	86	10	10	NS	4	NS
Mean ± SD	77 ± 12	30±8.8 ^{/a} 7.7±2.1 ^{/e}	20± 4.5 ^{/a} 7.7± 2.1 ^{/e}	10 ± 4.2	2.4 ± 1.5	1.3 ± 0.6

/a Ungloved workers, hand residues = wipe + wash

/b Azinphos-methyl

/c Not Sampled

/d Phosmet

/e Gloved workers, hand residues = wipe after glove removal

Hands: Exposure to the hands accounted for 30 ± 8.8% of DE in Sutter and Stanislaus Counties (wipes followed by wash) and 7.7 ± 2.1% in Madera County (residues under gloves, wipe only). For the peach harvesters the hand wipes captured about two-thirds of hand residues while the hand wash represented about one-third of total hand exposure. Daily glove residues for the Madera County site averaged 7.8 mg in 1989 and 16.9 mg in 1990 (Table VI).

Table VI. Apple Harvester Glove Residues, Madera County, 1989 and 1990 (mg)

Year	Day 1	Day 2	Day 3	Mean	Cumulative
1989	6.8	9.0	7.7	7.8	23.5
1990	16.9	17.3	16.4	16.9	50.6
Mean				12.4	

Other: For all sites, face/neck residues comprised no more than 4% of DE. Knee-length socks were employed in 1989 to evaluate the exposure to the lower leg. As this contribution to DE never exceeded 2%, the use of socks was discontinued in 1990.

Biological Monitoring

Urine Monitoring

Mean daily dimethyl phosphates, converted to µg UPE (urinary pesticide equivalents) are presented in Table III. The results reflect the combined contribution of exposure to both azinphos-methyl and phosmet, when residues of both pesticides were present in an orchard. For the study cohort, the mean daily volume (n = 300) and creatinine excreted (n = 280) were 919 ± 406 mL and 1017 ± 518 mg,

respectively. The correlation between volume and creatinine for analyses ≥ 100 mL urine (277 paired samples) was significant at $p < 0.001$ ($r = 0.45$). The coefficient of variation for UPE was about 60%.

Figure 3 presents the relationship between DE and UPE vs. total DFR for the five study sites for harvest days with concurrent dermal and urinary monitoring. The correlations are significant at $p < 0.01$. DE increases faster than UPE with increasing DFR, since UPE is damped by absorption. Table VII presents the average percentage of cumulative DE absorbed for each study site. While there is a broad range of DE and UPE among the five sites, mean percentage DE absorbed was lower for the peach harvesters (17.1 - 27.3%) and about one-half that of the apple harvesters (34.5 - 57.7%). The appearance of absorption exceeding exposure for the Madera County, 1990, apple harvesters indicates that a portion of the glove residues were absorbed. Figure 4 presents cumulative UPE vs. cumulative DE for the five study sites. While the regression was only significant at $p < 0.10$, the slope (0.32) nonetheless closely approximates the mean absorption value for the study (31.2%). Cumulative DE calculated including glove residues, and the corresponding percent absorption, are given in parentheses in Table VII.

Table VII. Cumulative Dermal Exposure (DE), Urinary Pesticide Equivalents (UPE) and Percent Absorption^a

Site	Year	mg DE	N	mg UPE	N	Percent Absorption
Sutter	1989	44.8	30	9.2	42	17.1
Sutter ^b	1990	16.0	22	3.9	96	19.6
Stanislaus	1989	105.0	25	39.5	24	27.3
Madera	1989	5.5 ^c (56.1) ^d	18	2.9	27	34.5 (4.9) ^e
Madera	1990	18.5 (42.0) ^d	23	25.2	27	57.7 (37.5) ^e
Mean						31.2 (21.3)^e

/a Percent absorption = cumulative UPE/(cumulative DE + cumulative UPE) x 100

/b DE for day 1 and 2, UPE for days 1 - 5

/c Estimated DE for day 1 is mean of day 2 and 3 DE

/d Cumulative DE including glove residues

/e Percent Absorption including glove residues in cumulative DE

Cholinesterase Monitoring

No clinical symptoms of organophosphate poisoning were reported by any of the workers or observed by study staff. Means for plasma and RBC cholinesterase for harvesters and control subjects are given in Table VIII and reflect clinical laboratory values. There was no significant difference in plasma or RBC means for workers except for Sutter County, 1989, where a significant decline in worker RBC means between the first and third draws was found (bold type). Clinical laboratory analyses showed a marginally significant decrease in RBC cholinesterase levels for both worker and control groups for the second draw ($p = 0.039$) and a return to at or near baseline for both groups at the third draw. However, the University analyses of duplicate samples demonstrated that at the third draw control RBC levels returned to baseline while worker RBC values did not. This confirmed that the decrease on the second draw for both the control (13%) and exposed groups (20%) reflected laboratory artifact in the clinical laboratory analyses, while the decline in worker RBC levels for the third draw (15% below baseline, $p < 0.001$) reflected response to exposure to OP residues.

Table VIII. Plasma and RBC Cholinesterase Means for Harvesters (H) and Control Subjects (C), 1989-90 in international enzyme units/liter, (U/L) from clinical laboratory data

Site	Year	Group	Plasma			RBC		
			First	Second	Third	First	Second	Third
Sutter	1989	H	2475	2575	2320	8441	6812	7157
		C	2536	2488	2207	8772	7614	8784
Sutter	1990	H	2568	2572	2581	6017	6060	6114
		C	2532	2500	2493	7089	7074	7478
Stanislaus	1989	H	2518	2374	NS ^{/a}	7035	7070	NS
		C	2539	2605	NS	9457	8898	NS
Madera	1989	H	2709	2597	NS	7755	8247	NS
		C	2652	2622	NS	7734	8340	NS
Madera	1990	H	2230	2483	NS	9102	9565	NS
		C	2615	2652	NS	9070	9729	NS

/a Not Sampled

DISCUSSION

Dislodgeable Foliar Residue (DFR)

The estimated half-lives for azinphos-methyl in this study (18.5 - 43 days) are consistent with previous DPR half-life data for stone fruit. In six studies of peaches, nectarines and plums, investigators found a mean half-life of 23 ± 14 days (n=18) (Maddy et al., 1982; Maddy et al., 1984; Maddy et al., 1986; Schneider et al., 1990; Spencer et al., 1988; Spencer et al., 1989). Application rates were similar, ranging from 0.7-1.5 lb. active ingredient/acre. There was a significant correlation ($p < 0.01$) between both daily dermal exposure (DE) and urinary pesticide equivalents (UPE) vs. total DFR (combined AZ and PM residues, where both were present) (Figure 3, n = 12 harvester dermal monitoring days). This lends support to the concept of using DFR to estimate harvester exposures. More research is needed to characterize the relationship in other work tasks and pesticides. More than 90% of the DFR were either below $0.80 \mu\text{g}/\text{cm}^2$ or greater than $2.40 \mu\text{g}/\text{cm}^2$. Exploring the relationship between exposure and DFR at intermediate DFR values may strengthen the existing correlation.

Dermal Exposure (DE)

In evaluating dermal exposure of agricultural workers to pesticides or pesticide residues, investigators have traditionally used gauze or cloth pads mounted on the skin, outer clothing or underside of outer clothing. Residues collected on the pads are extrapolated to the corresponding anatomical region and summed for an estimate of DE (inner pads) or potential DE (outer pads) (EPA, 1987; Durham and Wolfe, 1962). This method implicitly assumes a homogeneous distribution of the residues across all areas of the monitored region. Recently, DPR has used clothing dosimeters to monitor worker DE. While the precise amount available for dermal absorption is uncertain, residues on shirts and socks worn under outer clothing provide an exposure index that responds to the variation in degree of foliage contact by body regions, and the spatial influence inherent with patch dosimetry is minimized. Since this method involves less set-up time than is required for the attachment of pads, task interruption is reduced and a larger number of workers can be monitored. Potential disadvantages of using garments to assess DE have been cited and include the difficulty in changing clothes after each exposure period, contamination of the garment by residues on the face and hands while removing it and pre-extraction of interfering fabric or finish additives (EPA, 1987). Procedures followed in conducting this study have minimized these difficulties. Garments were pre-washed with two hot water wash cycles, which helped to reduce interferences to the MDL for azinphos-methyl (AZ) and phosmet (PM) ($5 \mu\text{g}$ for the shirts or less than

0.1% of the mean shirt residues found). New garments were distributed to the workers at the end of each workday so they could arrive at the work site wearing the dosimetry clothing the next morning. The monitoring period consisted of the entire workday, so subsequent changes were not needed. The workers provided hand residue samples followed by face and neck residue samples prior to removing the shirts and socks. Thus, contamination of the shirts and socks from these sources was minimized. This left the head region as a potential source of residue contamination. All workers wore a baseball cap while working, which likely intercepted some of the impinging residue. The wearing of caps by harvesters appears to be a standard work practice that may mitigate exposure.

The head and thigh regions were not monitored in this study. Caps or hoods and long underwear have been included in previous DPR monitoring strategies, but were excluded here because their use in high temperatures (85-110 °F) posed a risk of heat stress for the harvesters. Peach harvesters were observed to frequently contact the foliage with the thigh region, while apple harvesters were not. Both groups of harvesters had minimal lower leg contact with the foliage and similar regional exposure distributions to this area (1-2%, 1989 data, Table V). It was speculated that the thigh would contribute a similar amount to apple harvesters and a somewhat greater amount to peach harvesters. Previous investigators have attributed 10% of DE to the leg and hip regions (Spear et al., 1977). In this study, hip exposure was monitored by the dosimetry shirt.

The mean DE for harvesting peaches in this study was 23.2 mg (n = 7 harvester dermal monitoring days).

In a previous Branch study (Schneider et al., 1990), dermal exposure of nectarine harvesters exposed to AZ residues in Fresno County was monitored using a single long-sleeved shirt. The single shirt layer allowed the measurement of surface residues potentially available for absorption. The Fresno County harvesters received a mean potential DE of 17.2 mg. DE would be less than this amount, as only a portion of the residues would penetrate through a cloth layer. Using estimates for clothing penetration ranging from 10-45% (Popendorf et al., 1979; Thongsinthusak and Krieger, 1989), DE is calculated to be 1.7-7.7 mg. Lower DE estimates in the Fresno study compared to the present study are related to the lower mean AZ DFR of 0.31 $\mu\text{g}/\text{cm}^2$, or about one-fourth the mean DFR (AZ + PM DFR=1.41 $\mu\text{g}/\text{cm}^2$) measured for peach orchards in this study. Similarly, in the present study, in Stanislaus County, 1989, the lower level of AZ DE (2.0 mg) compared to PM DE (33 mg) is related to the much lower AZ residue at this site (mean DFR= 0.03 $\mu\text{g}/\text{cm}^2$) relative to PM residues (mean DFR=2.5 $\mu\text{g}/\text{cm}^2$).

Apple harvesters received about one-fourth the AZ DE of peach harvesters in Sutter County (3.5 mg vs. 14.3 mg) although AZ DFR for the Madera County site were similar to AZ DFR in Sutter County (0.59 $\mu\text{g}/\text{cm}^2$ vs. 0.56 $\mu\text{g}/\text{cm}^2$). There appears to be a lower exposure potential for harvesting crops grown as a hedgerow and this method of cultivation may be a useful engineering control to reduce contact with treated foliage. Previous studies have not investigated the relationship between differing agronomic practices and exposure potential for tree fruit harvesters. In recent work conducted by WH&S, investigators found that California grape girdlers contacted more than twice the amount of foliage as did California table grape harvesters; thus, exposure potential is greater for the girdlers (Dong et al., 1992)

Regional exposure distribution (%) is most consistent by site (Table V). This trend is noticeable even when two pesticides are present at varying DFR (Stanislaus County, 1989; Madera County, 1990; Table III) or when monitoring differs by year (Sutter and Madera Counties). Site-specific characteristics such as canopy, weather, irrigation method and cultivation practices, while difficult to quantify, appear to be influential in this phenomenon.

Hands DPR has been investigating the use of hand wipes to assess hand exposure. Hand wipes have many advantages compared to hand washes as they are light, convenient to use, can be purchased at retail outlets, require less sample storage space, reduce analyte hydrolysis while in storage and reduce the time and solvent volume involved in extraction procedures. However, for peach harvesters in this study, hand wipes appear to remove only two-thirds of total residues. In another investigation, the second of two sequential hand washes removed $22 \pm 8.7\%$ of the total residues of phthalate ester (n =

11, Kazen et al., 1974) which is comparable to DPR's hand wash recoveries after hand wipes. Nigg et al. (1990) found similar values for three sequential 95% ethanol hand rinses which removed 78%, 21% and 7%, respectively, of ethion hand residues. Factors that may influence the efficiency of residue removal by hand wash or hand wipe include their use for bare-handed vs. gloved harvesters and the sequence of wipe or wash.

Glove residues were not included in calculating DE as investigators have suggested that gloves are more absorptive than skin. Davis et al. (1983) found AZ residues on cotton and nylon gloves worn by apple thinners gave mean hand exposures 4-5 times greater than did hand rinses. Fenske et al. (1989) found a 1.5- to 2.5-fold greater hand exposure rate for peach harvesters when using glove monitoring compared to hand washes in captan-treated orchards. In studies of peach harvesters, Pependorf et al. (1974, 1979), using gloves backed by a pad, found that the gloved hand accounted for 68%-80% of DE for citrus and peach harvesters in phosalone, parathion and AZ-treated fields. Similarly, in the present study, if glove data for apple harvesters were included (Madera County, mean glove residues = 7.8 and 16.9 mg, for 1989 and 1990, respectively; Table VI) estimates of hand exposure would represent 73 - 81% of mean daily DE (Table III). The retention of considerably greater pesticide residues by gloves may lead to over-estimates of total exposure. It has also been suggested that gloves exhibit loading early in the exposure period and residues are therefore dependent on both sampling interval and production rate, while hand accumulation rates are constant with respect to time (Fenske et al., 1989). However, these previous studies did not include biological monitoring to determine the possible differences in absorbed dose for gloved and ungloved hands.

Pependorf et al. (1974, 1979) found that glove penetration averaged 6-8% for phosalone and parathion. In the current study, glove penetration averaged about 1 - 4.5% (estimates of hand residues from Tables III and V and overall mean glove residues of 12.4 mg from Table VI). In comparing the use of gloves vs. hand residues beneath gloves to assess hand exposure of tree fruit harvesters, it appears each contributes consistent proportions to DE, with gloves giving much greater estimates than hand wipe/hand wash data. DPR will continue investigations in this area.

Urine Monitoring Urine monitoring can provide a more sensitive indicator of exposure than either cholinesterase monitoring or passive dosimetry (Franklin, 1984; Drevenkar et al., 1991). It allows the use of familiar analyses (e.g., dimethyl phosphates) to compare both similar and dissimilar work tasks. In this study, while there was a greater than 10-fold difference in cumulative DE among the five study sites, percent absorption varied by less than three-fold (Table VII). Figure 3 demonstrates that exposure estimates from DE would over-estimate exposure compared to UPE. Urine monitoring thus provided a more precise measure of exposure (absorbed dose) than did passive dosimetry. A previous Branch study found substantially reduced exposure estimates for strawberry harvesters in captan-treated fields when urinary monitoring results were compared to dermal monitoring results (Maddy et al., 1989).

Measurement of urinary dimethyl phosphates showed a higher percentage of DE absorbed by the gloved apple harvesters (Table VII, 34.5 - 57.7%) than by the ungloved peach harvesters (17.1 - 27.3%). Occlusion of the hand may have increased the absorption of those residues penetrating the glove and environmental factors may have increased the availability of residues at this site. Estimating hand exposure using only those residues reaching the hand beneath the glove may not account for all the residues available for dermal absorption. Further research is needed to quantify the relationship between glove residues and hand exposure.

Cholinesterase Monitoring Meaningful conclusions from cholinesterase monitoring are problematic for harvester populations. The reentry interval (California Department of Food and Agriculture, 1991) protects the harvester from acute exposure to residues, and classic OP poisoning symptoms are rarely observed. Routine cholinesterase testing is not required for harvesters, so individual baselines are not usually available. The coefficient of variation for the normal range is 10 - 25%, which makes interpretation of marginally low values difficult (Duncan et al., 1986). If the work force is migrant, cholinesterase levels may be depressed or recovering while little or no information is available on the

type or degree of prior exposure. Drawing several samples per individual would allow characterization of these variables, but traditionally, worker acceptance for intensive monitoring has been poor. Additionally, a large, unexposed control group is often unavailable for comparison with field worker values. Thus, cholinesterase monitoring, while a valuable diagnostic tool in cases of organophosphate poisonings among pesticide handlers, has less clinical and practical utility for field workers. The availability of two independent laboratories for analyses of duplicate samples in Sutter County eliminated laboratory artifact as a confounding issue. The 15% depression in RBC levels seen for harvesters at this site in 1989 is not biologically significant (Table VIII). However, as cholinesterase depression was observed only for this harvester group while urinary dimethyl phosphates were detected for all harvester groups, cholinesterase monitoring is a much less sensitive indicator of exposure than urinary metabolite monitoring.

Crop-, task- and pesticide-specific exposure monitoring studies permit a more accurate estimate of worker exposure than do estimates based on surrogate data. For example, tree fruit harvesters receive greater exposure to pesticide residues than do harvesters of row crops, since a larger portion of their body is in contact with the treated foliage. Over-estimates of exposure for harvesters of row crops could result if surrogate data for tree fruit harvesters were used. Pesticide-specific study results can also be implemented to estimate the worker exposure potential of newly developed pesticides with similar chemical properties. DPR will continue to conduct studies to characterize different work tasks and pesticide exposure scenarios so that regulatory decisions to mitigate exposure, such as reentry intervals, will be exposure-based.

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