

Behavior of Parathion Residues in the Florida "Valencia" Orange Agroecosystem

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Parathion and paraoxon residues were measured in air, on foliage, fruit, and cover crop, and on the soil surface in a "Valencia" orange grove ecosystem. On the basis of residues, the greatest potential for worker exposure to parathion was on leaf and soil surfaces. For paraoxon, the greatest potential worker exposure was on the leaf surface. Negligible potential worker exposure was indicated in ambient air or on the fruit surface. Seventy-six percent of the parathion residue behavior on fruit and leaves could be explained by solar radiation, rainfall, and temperature, whereas only 55% of this variation was explained by time alone. Seventy-eight percent of the soil surface parathion residue behavior could be explained by solar radiation, rainfall, and temperature and only 42% with time alone. Solar radiation was the most highly correlated environmental variable with parathion disappearance. Rainfall was the most highly correlated environmental variable with paraoxon disappearance.

Parathion (*O,O*-diethyl *O-p*-nitrophenyl phosphorothioate) and its environmental conversion product, paraoxon (*O,O*-diethyl *O-p*-nitrophenyl phosphite), have been linked to fieldworker organophosphate intoxications, and parathion residue dissipation has been studied in various other geographical regions (Gunther et al., 1977). Preliminary studies of parathion residues in or on Florida citrus foliage and fruit have also been reported (Nigg et al., 1978; Thompson and Brooks, 1976).

Fieldworker illnesses due to parathion exposure have not been reported in Florida and the potential for worker exposure has not been determined. The purposes of this study were to determine the residue behavior of parathion on "Valencia" orange leaves and fruit, in grove air, on the native cover crop, and on the soil surface and to relate these data to possible worker exposure and to Florida weather conditions.

EXPERIMENTAL DESIGN

Field Plot Treatments. A mature "Valencia" grove in excellent horticultural condition owned by the University of Florida was used for these experiments. This grove is typical of the approximately 162000 ha of "Valencia" orange groves in Florida. Parathion emulsifiable concentrate (EC) and wettable powder (WP) were applied in separate treatments at 4.48 kg of active ingredient (AI)/ha in 285 dkl of water/ha (750 gal/acre) with an air-blast sprayer. Each treatment was replicated four times in a random block design including four no-spray treatments with two buffer rows between each 48-tree replicate. Three individual experiments were applied to the same plot. Experiment I was conducted from Jan 1–Mar 1, 1977; experiment II from July 12–Aug 2, 1977; and experiment III from Oct 1–Dec 14, 1977.

FIELD SAMPLING

Orange fruit and leaf samples were collected as described by Gunther et al. (1973), Westlake et al. (1973), and Iwata et al. (1977) except that eight fruit per plot were collected. Random cover crop samples were obtained by carefully clipping leafy portions of the cover crop into glass leaf-punch bottles. Plots were sampled on days 1, 3, 5, 7, and then at weekly intervals until residue levels were below 0.005 $\mu\text{g}/\text{cm}^2$ for two consecutive sampling days.

Soil samples were collected by vacuum by the method of Spencer et al. (1975, 1977) except that a 40-mesh screen

was used so that at least 10 g of soil could be obtained. Five 20 \times 20 cm areas were vacuumed per sample. A dripline and a middle (between trees) sample was obtained from each plot. These samples were not random samples in that areas previously sampled were marked with stakes so that no resampling occurred.

Mini-personnel samplers (Andersen 2000, Inc.) were used to obtain dislodged samples by attaching the sampler to a pole and sweeping the foliage for 5 min.

Ambient air was sampled for parathion by using Smith-Greenburg impingers as described by Sherma and Shafik (1975). Air flow was monitored at the beginning and end of each 4-h sample with a Gilmont Model No. F1400 flowmeter (Roger Gilmont, Instr., Great Neck, NY). Two-hundred milliliters of ethylene glycol was used per impinger. A prefilter was not used so ambient air samples represent an estimate of the vapor phase and airborne particulate parathion. Air samples were taken from 10 a.m.–2 p.m. each sampling day. All other sampling was begun at 9:30 a.m. and ended at approximately 11:00 a.m. each sampling day.

LABORATORY PROCESSING

Leaf and cover crop samples were processed as described by Gunther et al. (1974) as modified by Nigg et al. (1977). The Sur-Ten solution was 2 mL of a 1:50 dilution of Sur-Ten per 1.0 L of water. Fruit were clipped in the field into 2.5-gal pickle jars. Fruit were rolled twice in the same pickle jar at 70 rpm for 5 min in 200 mL of Sur-Ten solution to obtain dislodgable fruit residues. Two 40-mL methylene chloride extractions were used to recover parathion and paraoxon from fruit washes.

Dust levels on leaf and fruit surfaces were determined by filtering leaf and fruit washes through preweighed Whatman No. 1 filter paper after extraction with methylene chloride. Filters were dried at room temperature and reweighed, and the dust level was calculated in $\mu\text{g}/\text{cm}^2$ of surface (Westlake et al., 1973; Pependorf et al., 1975).

Soil samples were mixed by hand shaking, and a 10-g subsample was weighed into a 100-mL beaker. Two milliliters of water was used to wet the 10 g of soil; the beaker was covered with aluminum foil and placed at 4 °C. After 16–18 h, 40 mL of acetone was added to the beaker and the soil was sonicated for 30 s at 15000 kHz. The beaker was immediately recovered with aluminum foil and the sediment allowed to settle for 5 min. A 20-mL aliquot was taken and evaporated in a 50-mL volumetric flask under N_2 at 50 °C almost to dryness. After cooling for approximately 5 min, 5 mL of benzene was added, followed by 40 mL of 2% sodium sulfate. The volumetric flask was capped and shaken vigorously, the layers were allowed to

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Table I. Simple Correlation Matrix for WP Parathion Residues and Environmental Variables

leaves	fruit	soil-mid.	soil-drip	time ^a	HDD1 ^b	HDD2 ^c	CR ^d	CLW ^e	CS ^f	CAT ^g
1.000	0.905 ^h	0.847	0.577	-0.625	-0.636	-0.579	-0.847	-0.637	-0.770	-0.646
	1.000	0.850	0.615	-0.774	-0.690	-0.536	-0.741	-0.742	-0.851	-0.761
		1.000	0.649	-0.666	-0.742	-0.704	-0.748	-0.742	-0.728	-0.696
			1.000	-0.642	-0.625	-0.526	-0.566	-0.665	-0.654	-0.618
				1.000	0.883	0.653	0.660	0.940	0.946	0.981
					1.000	0.931	0.755	0.985	0.806	0.945
						1.000	0.736	0.861	0.586	0.766
							1.000	0.719	0.721	0.715
								1.000	0.863	0.977
									1.000	0.923
										1.000

^a Time in days. ^b HDD1 temperature limits 0 °C, 37.8 °C. ^c HDD2 temperature limits 10 °C, 37.8 °C. ^d Cumulative rainfall (inches). ^e Cumulative leaf wetness (hours). ^f Cumulative solar incidence in cal cm⁻². ^g Cumulative average temperature. ^h When $R > 0.496$, $P < 0.01$.

Table II. Simple Correlations for EC Parathion Residues and Environmental Variables

	time ^a	HDD1 ^b	HDD2 ^c	CR ^d	CLW ^e	CS ^f	CAT ^g
leaves	-0.604 ^h	-0.601	-0.537	-0.789	-0.604	-0.725	-0.616
fruit	-0.760	-0.652	-0.484	-0.694	-0.714	-0.837	-0.748
soil, med.	-0.636	-0.777	-0.784	-0.811	-0.758	-0.688	-0.321
soil, drip	-0.280	-0.374	-0.390	-0.386	-0.360	-0.289	-0.321

^a Time in days. ^b HDD1 temperature limits 0 °C, 37.8 °C. ^c HDD2 temperature limits 10 °C, 37.8 °C. ^d Cumulative rainfall (inches). ^e Cumulative leaf wetness (hours). ^f Cumulative solar incidence in cal cm⁻². ^g Cumulative average temperature. ^h When $R > 0.496$, $P < 0.01$.

separate, and the benzene layer was brought into the neck of the flask with distilled water. The benzene layer was removed by pipet to a brown bottle over a few grams of Na₂SO₄.

Mini-sampler impinger plates were extracted by rinsing each plate three times in 10 mL of benzene, evaporating the benzene to 10 mL, and transferring the benzene to a brown bottle over Na₂SO₄. The glass fiber filters were inserted into brown bottles, and 10 mL of benzene was added to the bottle.

Ambient air samples were processed by the method of Sherma and Shafik (1975). Processed samples were stored for less than 14 days at -20 °C prior to GLC analysis.

Recoveries by these methods were: fortified Sur-Ten washes; parathion 99 ± 3%, paraoxon 94 ± 4%, fortified soil, parathion 97 ± 2%, paraoxon 92 ± 2%; fortified ethylene glycol, parathion 100 ± 3%, paraoxon 98 ± 2%.

Analysis. Samples were analyzed by GLC with a flame photometric (FPD) phosphorus detector on a Tracor 550 GC. The 0.8-m silylated glass column was packed with 5% SP2100 on 100–120 mesh GCQ. Operating conditions were: injection 210 °C, column 190 °C, detector 170 °C, N₂ 60 mL/min. Selected samples were also chromatographed on a 5% OV-210, 100–120 mesh GCQ column at 170 °C.

After GLC analysis, samples of each type were combined and subjected to LC, TLC, column chromatography, and GLC-mass spectral analyses. Paraoxon was confirmed in each combined type sample by its GLC retention time on a 1.5-m column packed with 3% SP2100 on 100–120 mesh Supelcoport at 200 °C, 30 mL/min helium flow and by comparison of the mass spectra of peaks eluting at approximately 5 min with that of a known sample of paraoxon. Details have been previously described (Nigg et al., 1977, 1978).

Calculations and Statistics. Both sides of the leaf punch were used for calculating residue concentration in μg/cm². For fruit areas, the diameter and length measurements of each fruit were used for an area calculation of an oblate, prolate, or regular sphere on a Wang 720C computer (Buslig, 1978). Other statistical methods, computer programs, the heating-degree day transformation, and the first-order weather-time model have been

previously described (Nigg et al., 1977).

The percent total residue calculation used 240 × 10⁶ cm² of leaf surface area (Gunther et al., 1973; Turrell, 1961; Wilson, 1978), 20.4 × 10⁶ cm² of soil surface, and 9850 m³ of air per 48-tree plot. Total fruit surface per plot was calculated using actual fruit production times the average fruit surface area. These values were: 9.3 × 10⁶ cm² of fruit surface (experiment 1); 5.9 × 10⁶ cm² of fruit surface (experiment 2); and 8.6 × 10⁶ cm² of fruit surface (experiment 3). These values are based on a 48, 4.9 m tall tree plot spaced 7.6 × 7.6 m. The percentage of the total residue in each ecosystem component was obtained by calculating the residue in each component and dividing by the total. Since values did not differ between experiments, the average percent total residue was used in Figure 4. It was assumed that the concentration of parathion in air was homogeneous. The average of WP and EC dripline and middle residue determinations was used as the soil residue value. Diffusion kinetics and mass transfer processes were ignored for this calculation.

Environmental Monitoring. Rainfall, temperature, and leaf wetness methods have been reported (Nigg et al., 1977). For total solar incidence, a Matrix MK 1-G solarimeter (Matrix, Inc., Mesa, AZ) was used. This solarimeter was calibrated by triangulation on a clear day assuming 0.8 transmissivity according to the equation $I(\text{cal cm}^{-2} \text{min}^{-1}) = (2 \text{ cal cm}^{-2} \text{min}^{-1})0.8^{\text{csc } \theta} \sin \theta$, where θ is the solar elevation angle (Schultze, 1976). Determinations were obtained from 11:30 a.m.–12:45 p.m. EST at 15-min intervals for this calibration.

RESULTS AND DISCUSSION

Fruit and Leaf Residue. The weather model for parathion disappearance on "Valencia" orange leaves and fruit is presented in Figure 1. It should be noted in Tables I and II that the data trends for fruit and leaves and EC and WP formulations are not statistically different. The model of parathion disappearance in Figure 1 utilized both EC and WP parathion residue averages of leaves and fruit residues. Time is not used in this model and cumulative rainfall (inches), cumulative solar incidence (cal/cm²), cumulative leaf wetness (hours), and cumulative average temperature (°C days) account for 75.7% of the variation

Table III. Simple Correlations for WP Paraoxon Residues and Environmental Variables

	time ^a	HDD1 ^b	HDD2 ^c	CR ^d	CLW ^e	CS ^f	CAT ^g
leaves	-0.504 ^h	-0.659	-0.691	-0.708	-0.642	-0.546	-0.584
fruit	-0.406	-0.595	-0.665	-0.803	-0.556	-0.435	-0.485
soil, mid.	-0.276	-0.479	-0.570	-0.530	-0.435	-0.280	-0.376
soil, drip	-0.485	-0.565	-0.548	-0.716	-0.549	-0.519	-0.534

^a Time in days. ^b HDD1 temperature limits 0 °C, 37.8 °C. ^c HDD2 temperature limits 10 °C, 37.8 °C. ^d Cumulative rainfall (inches). ^e Cumulative leaf wetness (hours). ^f Cumulative solar incidence in cal cm⁻². ^g Cumulative average temperature. ^h When $R > 0.496$, $P < 0.01$.

Table IV. Simple Correlations for EC Paraoxon Residues and Environmental Variables

	time ^a	HDD1 ^b	HDD2 ^c	CR ^d	CLW ^e	CS ^f	CAT ^g
leaves	-0.529 ^h	-0.585	-0.554	-0.772	-0.581	-0.559	-0.560
fruit	-0.157	-0.176	-0.200	-0.574	-0.178	-0.342	-0.186
soil, mid.	-0.449	-0.557	-0.573	-0.671	-0.553	-0.461	-0.508
soil, drip	-0.442	-0.433	-0.389	-0.637	-0.444	-0.448	-0.435

^a Time in days. ^b HDD1 temperature limits 0 °C, 37.8 °C. ^c HDD2 temperature limits 10 °C, 37.8 °C. ^d Cumulative rainfall (inches). ^e Cumulative leaf wetness (hours). ^f Cumulative solar incidence in cal cm⁻². ^g Cumulative average temperature. ^h When $R > 0.496$, $P < 0.01$.

observed in the residue data. Time alone accounts for only 55% of the variation observed. Cumulative average temperature and heating degree days correlated equally well with parathion residues (Table I). For this reason, cumulative average temperature was used in place of heating-degree-days since they are approximately proportional and the calculation of cumulative average temperature does not require a computer.

The weather model presented in Figure 1 was the best overall model. No improvement in variation explained was obtained by including time instead of one of the environmental variables. In the overall fruit and leaf model, cumulative solar incidence appeared to be the most important environmental variable with an $R = 0.828$. The addition of rainfall yielded a multiple R of 0.851, cumulative average temperature a multiple R of 0.861, and cumulative leaf wetness a multiple R of 0.870.

On the other hand, the paraoxon-environmental variable correlations in Tables III and IV indicate the importance of cumulative rainfall. This is similar to previous weather correlations of paraoxon residues in Florida citrus (Nigg et al., 1978). This result also supports the supposition that differences in the levels of paraoxon residues between Florida and California citrus are due to wet conditions in Florida and dry conditions in California (Gunther et al., 1977; Nigg et al., 1978). However, the reasons for paraoxon residue differences between geographical regions are blurred because of simultaneous paraoxon production from parathion and disappearance behavior of paraoxon itself. The environmental production and persistence of paraoxon apparently depends on dusty conditions, ozone, and dry weather (Pependorf and Leffingwell, 1978; Spear et al., 1975, 1978). The determination in a mathematical sense of the dependence of paraoxon production on plant surfaces on environmental factors is an important consideration in prediction of safe worker reentry times (Spear, 1978). Once this dependence is known, accurate correlations of paraoxon disappearance and environmental factors can be made from field data as presented here.

The use of the Andersen mini-sampler for assessing airborne particulate matter dislodged during picking operations has been successful when worn by pickers (Westlake et al., 1973). No parathion, paraoxon, or dust was obtained by the sweeping techniques described here. It appears that it is necessary for a picker to wear the minisampler for at least 1 h for a measurable sample.

Dust levels on leaf and fruit surfaces as determined gravimetrically did not vary significantly over the course of an experiment or between experiments. Dust levels

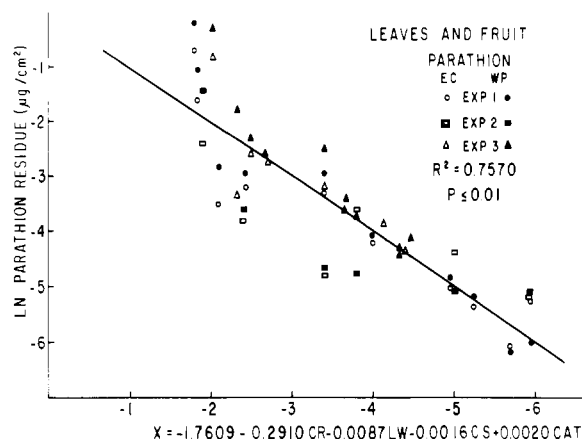


Figure 1. Weather model of emulsifiable concentrate (EC) and wettable powder (WP) parathion disappearance on "Valencia" orange leaves and fruit. Residue values are the average of fruit and leaf residues in $\mu\text{g}/\text{cm}^2$. Each point is the average of eight replicates (four fruit, four leaf). CS = cumulative solar incidence, cal/cm² (total h), LW = cumulative leaf wetness (h), CR = cumulative rainfall (inches), CAT = cumulative average temperature ($^{\circ}\text{C}$ days > 0).

averaged 105 $\mu\text{g}/\text{cm}^2$ (range 20–170 $\mu\text{g}/\text{cm}^2$) on leaf surfaces and 70 $\mu\text{g}/\text{cm}^2$ (range 44–96 $\mu\text{g}/\text{cm}^2$) on fruit surfaces. These dust levels are ca. one-third of those reported by Pependorf et al. (1975) and these data indicate an additional difference in Florida and California conditions which explains the lack of reports of acute picker exposure in Florida citrus. Differences between Florida and California regarding actual amounts of particulate matter dislodged during picking operations await elucidation.

Cover Crop Residues. Cover crop residues were obtained during experiment I (Jan–Mar 1977) after having allowed the native weed and grasses unrestricted growth from Aug 1976–Jan 1977. The disappearance of the residue from the cover crop was more rapid than from any other sample. Cover crop residues of WP parathion were about 12.0 ppm on day 1, 1.3 ppm on day 3, and 0.2 ppm on day 7. Residues of EC parathion on the cover crop were about 4.0 ppm on day 1, 0.70 ppm on day 3, and 0.15 ppm on day 7. No residues of either EC or WP parathion were detected after day 7 postapplication. The half-life of parathion on the cover crop was about 1 day compared to 1.3 days for air and 2.0 days for fruit, leaves, and soil surface. We attribute this observation to increased hours of foliage wetness as dew condenses from the ground up

Table V. Multiple Linear Regression^a of Parathion and Paraoxon Residues and Environmental Variables

	R ² × 100	Ln initial concn	coefficients			
			A ₁	A ₂	A ₃	A ₄
EC parathion						
leaves	72.7	-1.9764	-0.7912	-0.0065	0.0024	-0.0070
fruit	72.6	-2.2522	-0.1282	-0.0046	0.0010	-0.0041
soil, mid.	79.1	2.3005	-0.6386	-0.0045	0.0046	-0.0305
soil, dripline	18.3	1.5237	-0.4953		0.0020	-0.0175
EC paraoxon						
leaves	60.0	-4.9967	-1.1123			
fruit	55.1	-5.6513	-1.0834	-0.0041	0.0020	0.0004
soil, mid.	48.3	-1.9429	-1.6476		0.0033	-0.0265
soil, dripline	41.0	-3.5277	-1.7148		0.0004	
WP parathion						
leaves	85.3	-1.1538	-0.9763	-0.0093	0.0041	-0.01483
fruit	78.7	-1.6067	-0.2304	-0.0057	0.0019	-0.0091
soil, mid.	77.5	2.6433	-0.4415	-0.0075	0.0057	-0.0349
soil, dripline	61.6	2.7685	-0.0310	-0.0133	0.0095	-0.0591
WP paraoxon						
leaves	57.8	-4.9641	-0.6760	-0.0017	0.0030	-0.0227
fruit	74.3	-4.0270	-1.4043	0.0012	0.0026	-0.0181
soil, mid.	36.3	-1.8490	-1.8246	0.0091		-0.0170
soil, dripline	51.2	-1.8935	-1.9019			

^a Residue = Ln initial concn + A₁ (cumulative rain, in.) + A₂ (cumulative solar, h) + A₃ (cumulative average temperature) + A₄ (cumulative leaf wet, h.)

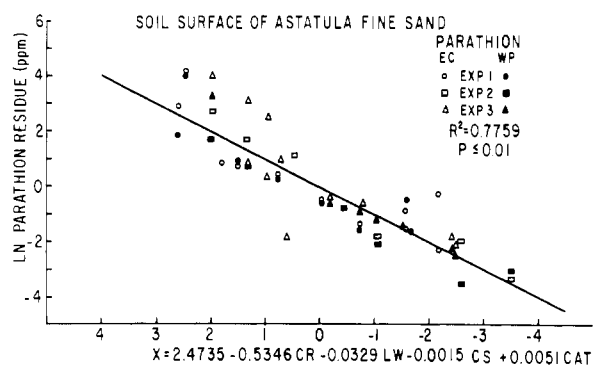


Figure 2. Weather model of emulsifiable concentrate (EC) and wettable powder (WP) parathion disappearance on the soil surface of Astatula fine sand between trees. Each point is the average of four replicates. CS = cumulative solar incidence, cal/cm² (total h), LW = cumulative leaf wetness (h), CR = cumulative rainfall (inches), CAT = cumulative average temperature (°C days > 0).

and disappears from the top down. The cover crop was observed to remain wet long after the tree foliage was dry.

The speculation that fruit pickers and grove workers might be exposed to residues on the cover crop is not supported by our residue data nor by normal grove practice. Florida citrus groves are regularly disked and mowed before picking operations at the demand of pickers to reduce inconvenience and the possibility of rattlesnake bite.

Soil Surface Residues. Parathion disappearance from the soil surface between trees, although slightly slower, is essentially the same as for leaves and fruit. There was no significant difference in the rate of decay between EC and WP formulations on the soil surface and these data are combined in Figure 2. The overall weather model and simple correlations indicate that cumulative rainfall may be important to disappearance of parathion on the soil surface between trees in Florida (Tables I, II, and V). The weather model accounts for about 78% of the soil surface parathion residue data between trees, whereas time alone explains only 42% of the data variation.

Paraoxon residue behavior on the soil surface between trees showed the strongest correlation with cumulative rainfall (-0.530 for WP and -0.671 for EC), suggesting the importance of hydrolysis for the disappearance of para-

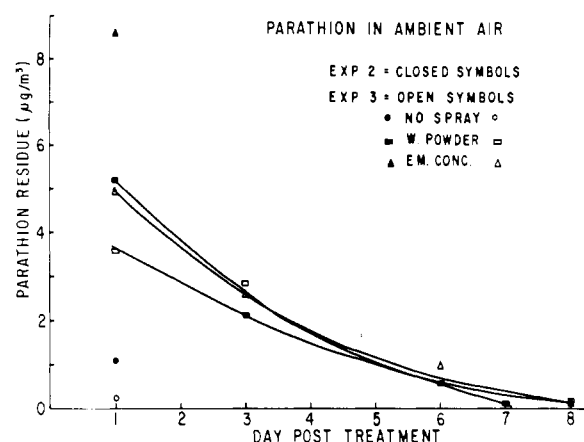


Figure 3. Parathion in ambient air in µg/m³.

oxon. Compared to leaf surface paraoxon correlations, however, both the simple and multiple correlations for paraoxon behavior on the soil surface were poor (Tables III, IV, and V).

Soil surface residue data for parathion disappearance at the dripline of the tree did not fit first-order kinetics for EC parathion, but were approximately first-order for WP parathion (Table V). The overall model for WP parathion at the tree dripline does not show the dependence on rainfall that was observed with the soil residue between trees. Differences in the solar and temperature processes at the dripline and nonuniform foliage channeling of residue runoff at application and during rainstorms might account for this observation.

Residues in Ambient Air. The ambient air parathion data in Figure 3 show levels of about 5.0, 2.5, 0.5, and 0.1 µg/m³ of parathion 1, 3, 6 and 8 days posttreatment in experiments II and III. There was no difference between WP and EC formulations nor between experiments for parathion levels in air. The very low air levels of parathion in the no-spray plots indicate that the 0.75-acre plot size used in these experiments may be adequate for determination of pesticides in air. No paraoxon was detected in any air sample.

Total Residues. It is readily apparent in Figure 4 that the leaf surface residue represents the greatest potential for worker exposure. For parathion, the soil surface

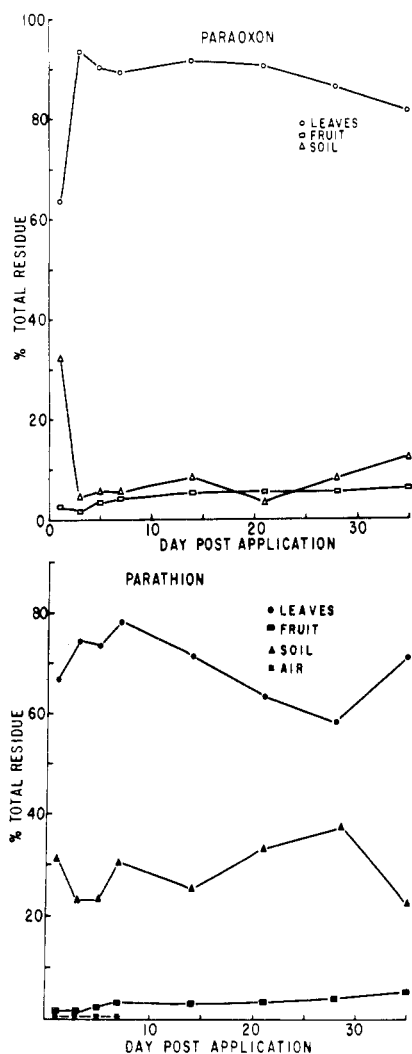


Figure 4. Percentage of parathion and paraoxon residues in air, on "Valencia" orange leaves, on "Valencia" orange fruit, and on the soil surface.

contained half as much compared to the leaf surface of the total plot residue hazardous to workers, while fruit and ambient air contained negligible percentages of residue available to workers. The leaf surface generally contained 90%, whereas the soil and fruit contained about 10% each of the paraoxon residues. These results are in substantial agreement with Westlake et al. (1973), Gunther et al. (1973), and Leffingwell et al. (1975). Based on these results, leaf surface and soil surface residue determinations may be sufficient to assess residue levels of pesticidal chemicals for the protection of farmworkers. Respiratory exposure from ambient air and exposure to the hands in picking operations would appear to be not significant, agreeing with Gunther et al. (1973) and Wolfe et al. (1975). These type of data may become useful when correlations with actual worker exposure are obtained.

If accurately constructed, models of pesticide behavior should have worldwide application. While the slope coefficient multiplying the weather variable in the multiple regression equation must remain constant, the variable itself may vary greatly from time to time or region to region. This can produce the effect of altering or even completely reversing the relative "importance" of different weather variables in different regions or at different times.

Monitoring studies of residue dissipation on plant parts and on the soil surface are currently required for the registration of pesticides. Methods for estimating worker

reentry times both mathematically (Serat, 1973, 1974, 1978; Serat et al., 1975) and with laboratory animals (Guthrie et al., 1974) have been suggested. The weather model of parathion disappearance presented here indicates that the estimation of residues on plant parts, which is critical for the mathematical approach, can be done accurately. This accuracy, however, can probably be improved by investigation of the importance of individual processes to pesticide disappearance from plant parts, i.e., hydrolysis, volatilization, photolysis, etc.

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