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Pesticide Exposure to a Greenhouse Drencher

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The pesticide exposure of greenhouse applicators is a current regulatory interest of the U.S. Environmental Protection Agency (U.S. EPA). The U.S. EPA is specifically faced with the task of (1) assessing the pesticide exposure of greenhouse applicators and (2) for pesticide label requirements, suggesting protective clothing which is both effective and comfortable. This study is a first step toward providing the data necessary for these evaluations.

The questions addressed were the following: (1) What was the potential for dermal exposure to the greenhouse pesticide applicator, i.e., at what rate did pesticide accumulate on the body (excluding hands) of the applicator, unprotected by clothing of any kind? We term this "estimated total body accumulation rate" (ETBAR) and measure it in µg/hr. Also, did the ETBAR depend upon the rate of pesticide leaving the spray nozzle or the kind of pesticide applied? (2) How was the ETBAR distributed over the anatomy of the applicator and upon what factors did this distribution depend? (3) What was the accumulation rate of pesticide on the hands of the applicator? Was there a relationship between worker hand preference and exposure to the right and left hands? Did hand exposure depend upon the pesticide effluent rate or the compound applied? (4) What was the atmospheric contamination from the pesticide in the breathing zone of the worker as he applied the compound? Did it depend upon the compound used or its effluent rate? (5) What was the penetration of pesticide through the various types of protective clothing worn by the applicator? Did penetration depend upon the compound used? (6) How did samples of the spray mixture, taken pre- and post-application, compare in pesticide concentration with that presumed to exist in the tank based on the mixture recipe, the pesticide label, and an assumption of thorough mixing?

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MATERIALS AND METHODS

The study was conducted during the period July-October, 1985, at a commercial greenhouse facility near Parrish, Florida, devoted primarily to growing chrysanthemums and African violets. The chemicals applied, sometimes in combination, were Mavrik™, 22.3% EC fluvalinate [(RS)-α-cyano-3-phenoxybenzyl(R)-2-[2-chloro-4-(trifluoromethyl) anilino]-3-methyl-butanoate]; Dursban™, 50% WP chlorpyrifos [O,O-diethyl O-(3,5,6-trichloro-2-pyridyl)-phosphorothioate]; Truban™, 30% WP ethazol (5-ethoxy-3-trichloromethyl-1,2,4-thiadiazole); and Daconil™, 75% WP chlorothalonil (tetrachloroisophthalonitrile). The primary subject for this study was male, 26 years of age, 183 cm in height, weighed 79 kg, and was right-handed. His total body surface area was estimated to be 2.02 m^2 (Gehan and George 1970). A single exposure measurement was also taken from a 30-yr-old female drencher of 176 cm height and 63 kg weight (1.76 m^2 estimated body surface area) who was left-handed. The subjects were instructed to change no aspect of their normal application routine and to wear only their usual protective clothing. The subjects sprayed with a single-nozzle handgun equipped with an adjustable nozzle set to provide a low pressure, coarse spray and intended to completely "drench" the plant and surrounding soil. Applications were made in an open-sided greenhouse with a translucent plastic roof. However, on September 26, 1985, the primary subject applied in an enclosed, but ventilated, structure as did the female subject on July 23, 1985, her only application.

The primary subject wore a rubber apron with a bib. However, he wore the bib down on all exposure days but July 31, 1985, and replaced the apron with a Tyvek™ coverall and hood for the first exposure of October 16, 1985. The female subject wore a rubber apron with the bib up. Aprons extended from the chest (bib up) or waist (bib down) to mid-calf. Both subjects wore rubber boots, extending to mid-calf. Additionally, the female subject wore rubber gloves, extending to mid-forearm, and a respirator.

Sampling, extraction, and analytical methods have been described elsewhere (Stamper et al., in press). Outside exposure pads were placed on the subjects at the back and chest, and on both shoulders, forearms, thighs, and shins. These pads were outside all clothing and entirely exposed to pesticide. If an apron or boot covered any of the above sites, the pads were placed on the outside of the apron or boot at the same level as described above. Left and right pairs of pads (shoulders, forearms, thighs, and shins) were combined for extraction and analysis. Timed exposure periods were at the convenience of the subject, lasting an average of 40 min.

The analytical results for pesticide compound, uncorrected for recovery, divided by the pad area (one or two pads) and exposure time, give the pad fluxes appearing in Table 1. The ETBAR was calculated from the outside pad fluxes by multiplying each flux by the anatomical area represented and summing these accumulation rates. If any of the individual accumulation rates could not be estimated because of a lost or missing sample, no ETBAR was calculated. Handwash accumulation rates were not included in the ETBAR since they were dermal exposures and measured by a different method. Also appearing in Table 1 are spray rates, the quotient of the amount of compound sprayed (calculated from the volume of mixture sprayed and the presumed concentration of compound in the mixture) and the exposure time. The atmospheric pesticide contamination near the breathing zone of the subject is also given and is based upon a 3 L/min intake of air by the personal air sampler (Dupont P-4000) worn by the subject. Tank mixture samples, taken pre- and post-application from the handgun itself, are presented. Inside pads were placed immediately inside protective gear (if worn) just beneath, but not overlapping, the outside pad at the chest, both forearms, both thighs, and both shins. While outside pads were without exception always exterior, inside pads were protected by various garments: either by a rubber apron or a Tyvek™ coverall at the chest and thighs, by the coverall or gloves at the forearms, and by boots or boots plus coverall at the shins. Ambient air temperature and relative humidity were taken at the application site pre- and post-application. Means of these values appear in Table 1.

Mean (± S.E.) recoveries from fortified media in preliminary laboratory experiments were: fluvalinate, pad 89 ± 9%, handwash 76 ± 5%, air sampler plug 67 ± 2%; chlorpyrifos, pad 91 ± 1%, handwash 94 ± 4%, air sampler plug 77 ± 2%; ethazol, pad 74 ± 6%, handwash 98 ± 12%, air sampler plug 50 ± 4%; and chlorothalonil, pads 94 ± 1%, handwash 53 ± 2%, air sampler plug 68 ± 4%. In addition, loss studies were conducted on the exposure pads and the extracts therefrom. No significant loss of compound(s) could be validated by any of these studies, except that the ethazol extract showed a 16% loss at 91 days of storage. Field blanks were blank and field fortifications were within recovery ranges. All analyses were done by gas chromatography using Ni^{63} electron capture detection.

RESULTS AND DISCUSSION

Each monitoring period was an experiment unto itself since variations existed in compounds, compound spray rates, exposure times, and subjects. Therefore, no exposure constituted a true replication of any other exposure because of these confounding variables which were not under experimental control. In order to draw any general conclusions, however, some grouping of the data into classes was required. In practice, there was little variation in presumed effluent rate (kg a.i./hr) from the applicator's spray nozzle when he applied a given compound (Table 2). The phrase "presumed effluent rate" suggests some ambiguity and derives from our observation that thorough mixing of the compound was not usually accomplished in the spray tank. While t-tests ($p < 0.05$) showed no statistical difference between pre- and post-spray samples, the data indicated that for fluvalinate and chlorpyrifos, only about one-half of the calculated concentration was actually leaving the spray nozzle (50 ± 7% and 47 ± 9%, respectively). This circumstance, however, did not exist for ethazol and chlorothalonil (109 ± 10% and 117 ± 21%, respectively).

It was evident (Table 2) that the differences in mean ETBAR, total handwash (left plus right), and air samples which existed among compounds were in part explainable on the basis of mean presumed spray rate differences. Consequently, the individual exposure

Table 1. Individual exposure values

Compound[1]	8/22/85 F	9/9 A[2] F	9/9 B F	9/27 F	10/16 A F	10/16 B F	10/16 C F	7/23[3] Cs	7/31 Cs	9/9 A Cs	9/9 B Cs
Exposure period (hr)	0.350	0.850	1.233	0.417	0.550	0.717	0.500	0.167	0.967	0.850	1.233
Spray rate (kg a.i./hr)	0.023	0.046	0.032	0.038	0.055	0.042	0.036	0.213	0.235	0.334	0.230
Flux ($\mu g/cm^2hr$)											
Outside pads											
Back	0.080	ND[4]	ND	ND	ND	ND	ND	0.002	0.006	0.003	0.008
Chest	0.079	ND	ND	ND	ND	ND	ND	0.007	0.149	0.005	0.015
Shoulders	0.049	ND	ND	ND	ND	ND	ND	0.023	0.059	0.004	0.009
Forearms	0.070	ND	ND	ND	ND	ND	ND	0.010	1.660	0.011	0.016
Thighs	0.241	0.001	0.022	0.029	ND	ND	ND	2.593	4.251	0.021	1.355
Shins	4.266	0.061	0.046	0.039	ND	0.039	0.286	0.380	2.467	1.541	5.265
Inside pads											
Chest	—	—	—	—	ND	—	—	0.009	0.008	—	—
Forearms	—	—	—	—	ND	—	—	0.016	—	—	—
Thighs	ND	0.003	0.007	0.016	ND	ND	0.018	0.020	0.001	0.012	0.016
Shins	ND	ND	ND	ND	ND	ND	ND	0.003	0.002	0.006	0.010
Accumulation rate ($\mu g/hr$)											
Left handwash	391.3	81.7	95.0	8.1	47.7	3.4	15.9	82.1	363.0	209.8	132.8
Right handwash	433.0	75.6	62.3	13.4	19.2	12.3	62.4	34.2	474.5	184.2	154.2
Est. total body[5]	18808.5	251.0	266.9	263.5	ND	154.0	1143.5	9306.4	31016.3	6293.6	26178.1
Air sampler ($\mu g/L$)	ND	ND	ND	0.0117	ND	0.0015	ND	ND	0.0041	0.0029	0.0080
Tank mixture (ppm a.i.)											
As mixed	84	84	84	84	80	80	80	468	600	600	600
Pre-spray	52	63	64	71	22	11	14	119	295	310	723
Post-spray	54	66	75	24	13	26	32	183	354	243	756
Mean air temperature (°C)	29	31	27	30	28	25	25	30	28	31	27
Mean relative humidity (%)	78	73	83	70	81	87	90	76	81	73	83

[1]F: fluvalinate, Cs: chlorpyrifos, E: ethazol, Cl: chlorothalonil [2]Letters denote spraying at different times on the same day
[3]Female drencher (sprayed on 7/23/85 only) [4]None detected [5]Excluding hands

Table 1. (Cont'd)

Compound	9/26/85 A Cs	10/10 Cs	10/16 A Cs	10/16 B Cs	10/16 C Cs	9/9 A E	9/9 B E	8/22 Cl	9/26 A Cl	9/26 B Cl
Exposure period (hr)	0.300	0.833	0.550	0.717	0.500	0.850	1.233	0.350	0.300	0.567
Spray rate (kg a.i./hr)	0.378	0.272	0.412	0.316	0.272	0.150	0.103	0.243	0.567	0.300
Flux ($\mu g/cm^2 hr$)										
Outside pads										
Back	—	0.005	0.007	0.062	0.011	0.005	0.007	0.134	—	0.036
Chest	0.023	0.005	0.004	0.011	0.016	0.011	0.033	0.379	0.019	0.045
Shoulders	0.011	0.012	0.003	0.004	0.012	0.013	0.007	0.456	0.194	0.069
Forearms	0.091	0.013	0.007	0.006	0.018	0.010	0.020	1.604	0.149	0.287
Thighs	0.204	6.995	0.009	0.011	0.033	0.018	0.096	3.239	0.575	7.140
Shins	0.605	0.242	0.010	0.603	12.802	0.131	1.306	25.630	1.186	13.248
Inside pads										
Chest	—	—	0.012	—	—					
Forearms	—	—	0.005	—	—					
Thighs	0.014	0.024	0.014	0.030	0.054	0.015	0.020	0.004	0.017	0.073
Shins	0.004	0.001	0.003	0.001	0.003	0.005	0.003	0.004	0.001	0.004
Accumulation rate ($\mu g/hr$)										
Left handwash	6938.8	397.1	347.1	262.1	350.6	2.8	1.0	1125.0	1403.8	1251.1
Right handwash	5902.6	574.9	442.5	212.2	1057.5	5.9	1.1	1020.0	3629.5	1001.6
Est. total body	—	27010.1	142.9	2760.1	51419.9	692.5	5792.4	121354.9	—	80604.7
Air sampler ($\mu g/L$)	0.0037	0.0122	0.0057	0.0158	0.0111	0.0343	0.1369	0.0334	ND	0.0065
Tank mixture (ppm a.i.)										
As mixed	600	600	600	600	600	135	135	899	899	899
Pre-spray	430	10	163	157	170	109	176	1980	925	946
Post-spray	404	68	77	170	211	155	147	680	865	912
Mean air temperature (°C)	28	26	28	25	25	31	27	29	28	26
Mean relative humidity (%)	92	89	81	87	90	73	83	78	92	—

Table 2. Mean[1] values for pesticide spray rate, drencher accumulation rates, and normalized values

Compound	Spray rate (kg a.i./hr)	ETBAR[2] (μg/hr)	Left handwash[3] (μg/hr)	Right handwash[3] (μg/hr)	Air sampler[4] (μg/L)
Fluvalinate	0.039 ± 0.004 (7)	3,000 ± 2,600 (7)	92 ± 52 (7)	97 ± 57 (7)	0.002 ± 0.002 (7)
Chlorpyrifos[5]	0.306 ± 0.023 (8)	20,700 ± 7,000 (7)	1,100 ± 800 (8)	1,100 ± 700 (8)	0.008 ± 0.002 (8)
Ethazol	0.127 ± 0.024 (2)	3,200 ± 2,500 (2)	2 ± 1 (2)	4 ± 2 (2)	0.086 ± 0.051 (2)
Chlorothalonil	0.370 ± 0.100 (3)	101,000 ± 20,400 (2)	1,300 ± 100 (3)	1,900 ± 900 (3)	0.013 ± 0.010 (3)

Accumulation rates, *normalized* for spray rate (mg deposited/kg sprayed)

	NETBAR[2]	Normalized total handwash[3]	Normalized air sampler[4]
Fluvalinate	125 ± 116 (7)	7 ± 5 (7)	0.0088 ± 0.0078 (7)
Chlorpyrifos[5]	80 ± 27 (7)	7 ± 4 (8)	0.0050 ± 0.0011 (8)
Ethazol	28 ± 28 (2)	0.04 ± 0.02 (2)	0.1402 ± 0.0990 (2)
Chlorothalonil	384 ± 115 (2)	8 ± 0 (3)	0.0095 ± 0.0077 (3)

[1]± standard error, with number of "replications" in parentheses [2]Does not include handwash [3]Subject wore no gloves [4]Applications made in an open-sided structure, except for three applications made in an enclosed structure [5]Data for female drencher, not included in the above: Spray rate = 0.213, ETBAR = 9,300, Left handwash = 82, Right handwash = 34, Air sampler = none detected, NETBAR = 44, Normalized total handwash = 1, Normalized air sampler = none detected, with all units the same as in the table above (subject wore gloves and sprayed in an enclosed structure)

Table 3. Mean[1] transmittance to inside pads

	Fluvalinate	Chlorpyrifos	Ethazol	Chlorothalonil
Chest, covered by				
coverall only	0[2] ± — (1)	fn.4		
apron only		0.054 ± — (1)[4]		
Forearms, covered by				
coverall only	0 ± — (1)	fn.4		
gloves only		fn.4		
Thighs, covered by				
coverall only	0 ± — (1)	fn.4		
apron only	0.218 ± 0.134 (4)[3]	0.111 ± 0.093 (6)[3]	0.521 ± 0.313 (2)	0.014 ± 0.009 (3)
Shins, covered by				
boots only	0 ± 0 (6)	0.003 ± 0.001 (8)	0.020 ± 0.018 (2)	0 ± 0 (3)
coverall plus boots	0 ± — (1)	fn.4		

[1] ± standard error, with number of "replications" in parentheses [2] Indicates < 0.001 [3] Two transmittance measurements absent (see text) [4] One transmittance measurement absent (see text)

parameters of Table 1 were normalized for (divided by) spray rate and the mean values recalculated. What resulted was a measure of the applicator's mean exposure in mg-deposited-per-kg-sprayed, the time units having cancelled out. Air sampler values had first to be translated to mg/hr via the 3 L/min factor. It should be noted that the air sampler data underestimate the respiratory exposure of an unmasked greenhouse applicator by a factor of about six. A study by Adamis et al. (1985) found that greenhouse workers inhaled air at a rate of approximately 1 m^3/hr (17 L/min). Left and right mean handwash data showed no significant differences ($p < 0.05$) throughout and were summed to give "normalized total handwash".

To determine whether significant differences existed among compounds for the various normalized statistical parameters in Table 2, a Duncan's Multiple Range Test ($p < 0.05$) was applied. The primary subject was significantly more exposed to normalized ETBAR (NETBAR) contamination from chlorothalonil than from ethazol or chlorpyrifos. Neither normalized handwashes nor normalized air samples could be differentiated for compound. We cannot explain the increased NETBAR risk from chlorothalonil, but it calls into question the generic data base approach currently under way at the U.S. EPA.

The distribution of the ETBAR to various body regions of the subjects (excluding hands) was calculated. Normalization for spray rate was not done here, under the assumption that spray rate should not affect the percent of compound deposited on a given body area. For each body area, two-factor analyses of variance ($p < 0.05$) showed no statistical difference in percent of ETBAR among either subjects or compounds. Mean ± S.E. percent allocations of the ETBAR from 17 replications were: head-neck (1 ± 0%), front torso (2 ± 1%), back torso (2 ± 1%), arms (2 ± 1%), upper legs (20 ± 6%), lower legs (74 ± 5%). It is to be noted that, although the upper and lower legs together comprised only 38% of the body's surface area, this region accounted for 94 ± 3% of the ETBAR (hands excluded).

The female drencher (footnoted in Table 2) received a lower NETBAR, normalized total handwash, and normalized air sampler deposit than the mean normalized chlorpyrifos values of the primary subject. Her ETBAR distribution was similar to that of the primary subject.

Penetration of pesticide through protective clothing was assessed by computing the ratio of inside pad flux to that of the corresponding outside pad. This value we call "transmittance", a term borrowed from optics. Real penetration is now frequently measured in a laboratory setting with controlled "applications" to a sample of fabric. Transmittance, as we employ the term, is to be contrasted with this process. In the field, inside pads can suffer contamination by other routes than directly through protective clothing. For example, samplers observed that when the applicator extended his arm, the sleeve of the coverall had a tendency to ride up, exposing the inside forearm pad. Furthermore, the outside and inside pads occupied adjacent rather than overlapping positions, so they were not necessarily subjected to equal amounts of pesticide. We include all such events in "transmittance", with the latter term interpreted very broadly. Mean transmittance is presented in Table 3. Significant differences among compounds in Table 3 were assessed statistically through a Duncan's Multiple Range Test ($p < 0.05$).

None could be confirmed. Table 3, however, is absent ten separate transmittance determinations. They are: for fluvalinate, 9/9/85 A and 10/16/85 C thighs; and for chlorpyrifos, 7/23/85 chest and forearms, 10/16/85 A all body locations, 10/16/85 B and C thighs. Each of these transmittances was near or greater than one in magnitude and each derived from very low exposures to both the outside and inside pads (Table 1). Their inclusion into Table 3 would have skewed the mean values reported there toward larger but unrepresentative values.

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