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Monitoring Human Exposure During Pesticide Application in the Forest

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The extent to which exposure to pesticides may be hazardous to applicators depends upon exposure levels and the toxicity of the compounds. The phenoxy herbicides have been used for nearly 40 years, and no injury to workers properly using these herbicides has been clearly established.

In spite of their record of producing no detectable harm to humans, the phenoxy herbicides 2,4-dichlorophenoxy acetic acid (2,4-D) and 2,4,5-trichlorophenoxy acetic acid (2,4,5-T) have acquired a less than desirable reputation. This reputation has been the result of their association with low levels of impurities. They have commonly been used as a mixture, which contains trace amounts of highly toxic 2,3,7,8-tetrachlorodibenzo-*p*-dioxin, a minor product in the manufacturing of 2,4,5-T. In early production of 2,4,5-T a low level of dioxin was retained. Today's manufacturing process produces 2,4,5-T with no more than 0.1 ppm of the 2,3,7,8 tetrachlorodibenzo-*p*-dioxin. This association with toxic dioxin and confusion of the public and the media regarding these issues have led to public distrust in the safety of using phenoxys and to the need to establish clearly the extent of human exposure to these compounds as well as the resulting effects of this exposure.

The phenoxys have become a major tool in silviculture. They have allowed the forest industries to eliminate more economically the competing vegetation which impedes the rapid growth and harvest of conifer forests.

Until recently little data had been gathered on human exposure to these compounds. To evaluate their safety, the exposure received and dose absorbed must be considered in relation to their toxicity. Since restrictions were placed on the use of 2,4,5-T by the EPA in 1978, several exposure studies have been conducted with 2,4,5-T and also with 2,4-D and other compounds used in forest operations. Recent interest in evaluating human

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exposure to pesticides in forestry has paralleled these interests in agronomic and horticultural crop production. However, numerous studies in these other areas preceded most of the forest work. Since the 1970's several studies have specifically dealt with forest applications of phenoxys and other pesticides. Since different pesticides as well as different experimental designs and methods have been used in the evaluations, it is not always simple to make accurate comparisons among studies. However, some comparisons can be made and each study has contributed to the studies of today. The review of studies on 2,4,5-T by Leng et al. (1980) suggests the kinds of differences as well as the comparisons that can be made among exposure studies.

Tarrant and Allard (1972) tested forestry workers in the early 1970's to determine dose absorbed from the application of cacodylic acid by analyzing the urine for arsenic. Analysis of urine has come to be regarded as the best method for determining dosage for the phenoxys. Exposure studies on 2,4-D and 2,4,5-T in recent years have included those by Sauerhoff et al. (1977). Kolmodin-Hedman et al. (1979) in Sweden compared plasma and urine levels for 2,4-D and 2,4,5-T in a four-man forestry study. They speculated that uptake was both by inhalation and dermal exposure and that elimination through urine was rapid. They concluded that the highest levels of phenoxy acids were found in the urine. Draper and Street (1982) monitored two groups performing ground applications of a 1:1 mixture of the dimethylamine salt of 2,4-D and dicamba. One group sprayed once for 5.5 hours; the other continued daily spraying. First void urine samples were used each morning of the experiment. Ethanol hand rinses removed considerable 2,4-D and dicamba from the hands. The effect of rinsing in this way on the amount absorbed and later excreted is not known. Removing the herbicide should mean that the internal dose would be reduced since dermal exposure to the hands would be reduced. However, the possibility exists that the herbicide dissolved in the solvent may more readily penetrate the skin, increasing the internal dose. Maximum urinary excretion occurred after 48 hours. Higher concentrations of 2,4-D were excreted than dicamba or its isomer. They also concluded that respiratory exposure was minor compared to dermal exposure.

Our evaluations using 2,4,5-T and 2,4-D have been conducted over the past 5 years in forests in Arkansas, Oregon and Washington. Objectives were to measure external exposure and internal doseage as determined by the total amount of the herbicide excreted in the urine and also to develop the best possible techniques for assessing exposure and dose absorbed. Exposure levels were related to job responsibilities and to protective techniques designed to limit exposure.

The data collected from exposure studies can be used with toxicological data to assess the safety of applying the pesticides in forest operations.

Experimental Procedures

Field crews that normally apply pesticides were monitored during their routine working day with as little interruption as possible to their customary work procedures or habits. When human error or mechanical irregularities occurred, the study was continued, and the irregularity was incorporated into the analysis of data. In this way we could monitor exposure that would include unexpected difficulties and spontaneous or habitual human reactions under actual "real-life" conditions.

Measurements were made of the concentration of pesticide in the breathing zone of the workers, on patches attached to the workers' clothing, and in the urine of crew members. In the 2,4-D tests, comparisons were made between amounts found under normal spray operations and amounts found when techniques for limiting exposure were used including special instructions and the use of protective clothing consisting of hat, boots, gloves, and Tyvek coveralls.

The dermal exposure patches were made of 9-ply gauze (2,4,5-T study) or denim (2,4-D study) and were attached with safety pins to workers' clothing by research team members wearing clean gloves. Following the spray activities, the patches were placed in individual specimen bottles and transported to the laboratory for analysis. In the 2,4,5-T study, all six patches from each individual were pooled before analyses were made; in the 2,4-D studies the patches were kept separate and analyzed individually. Using a photograph of the worker in his spray attire and the amounts of pesticide found on the patches, we estimated total dermal exposure for each worker (Durham and Wolfe, 1962).

Pesticide vapors and airborne particles in a worker's breathing zone were pulled through a trapping medium by a battery-powered air pump attached to his belt. Cassettes containing the trapping medium were removed at the conclusion of the test and transported to the laboratory where they were analyzed. Collection of the total urine voided began 1 (2,4,5-T) or 2 (2,4-D) days prior to each spray operation and continued for at least 4 days afterward. Samples were collected in 12-hour intervals. These specimens were kept in a cool location and transported to a central storage facility at 2-day intervals. To ensure integrity throughout the analytical determination, blind-fortified specimens containing known levels of the pesticide were intermingled with the actual field specimens.

2,4,5-T Study

Twenty-one crew members participated in a 2,4,5-T forestry study which was repeated after an interval of one or two weeks.

Exposure was measured for four crews in Arkansas using three methods of pesticide application: backpack, tractor-drawn mist blower, and helicopter (two crews). Comparisons of exposure levels were made between crews and within crews in relation to work duties.

The backpack team was composed of seven crew members, including a mixer-supervisor and six applicators. The tractor-mounted mist blower operation included a supervisor, two tractor drivers, and a mixer. Each of the helicopter crews had a pilot, a mixer, a supervisor, and two flagmen. All workers, except two backpack applicators, were men. Prior to this spray program, each worker filled out a form which provided personal information regarding the worker's vital statistics and history of any previous involvements with 2,4,5-T use. Workers indicated that they had not worked with the test compound for two weeks prior to the study. The typical attire for members of the spray crews included long trousers, shirt (long or short sleeves), and cloth sneakers, leather shoes, or field boots. Most crew members did not wear gloves or other protective clothing, but all wore hats except four members of the backpack crew.

The air pump worn by each worker in the 2,4,5-T study contained an Amberlite XAD-II resin. Air from the breathing zone was drawn across the resin at an approximate rate of 0.1 to 0.15 litres/min. The resin was retrieved after each operation and analyzed for 2,4,5-T.

Patches to collect measurements of dermal exposure were attached to clothing on the chest, back, thighs and forearms of each individual. To calculate total dermal exposure, the concentration of 2,4,5-T detected on the patch area was multiplied by the total skin area exposed (Lavy, 1978).

2,4-D Study

The 2,4-D study was similar to the 2,4,5-T study which included analyses of air, patches and total urine. In this study three helicopter crews were monitored during their routine forest spray operations in Washington and Oregon. An additional objective in this study was to compare exposure from the routine operation (T_1) with that received when workers wore protective clothing and followed added precautions designed to limit exposure (T_2). The T_1 and T_2 operations were conducted with the same individuals in each crew with a 1-week interval between spray operations. Each crew included a pilot, a batchman, a mechanic, a supervisor, and two observers. The observers were located from 67 to 168 m away from the spray operator. Their role was to represent persons who might be in the area, but who were not directly associated with the spray operation. Protective clothing worn in the T_2 operation included Tyvek coveralls, clean hats, rubber gloves, rubber boots, and goggles.

The collection and handling techniques for monitoring 2,4-D exposure levels in the breathing zone, on patches to measure dermal exposure, and in urine were similar to the procedures carried out in the 2,4,5-T tests. One difference was in the type and location of patches used. Denim strips were attached to workers' clothing near bare skin areas. A 2.5 by 40-cm strip was attached to the workers' collar, a 2.5 by 48-cm strip to the hatband, and two 2.5 by 15-cm strips around the wrists to the cuff.

Results and Discussion

Determining Acceptable Parameters for Field Measurement

Patches attached to clothing commonly have been used to obtain predictions of the amount of dermal pesticide exposure a field worker using pesticides would receive. The ease of patch construction, simplicity of attaching to clothing, and the fact that conceivably an exposure study could be completed during one application day make the use of patches highly attractive. By analyzing the amount of spray material deposited on the patches and evaluating the area of bare skin exposed for each worker via photographs, one can theoretically obtain a good estimate of the amount of pesticide contacting the exposed dermal area of the worker.

Pharmacokinetic studies with 2,4-D in rats (Sauerhoff et al., 1977) have shown that orally ingested or intravenously administered 2,4-D is excreted primarily in the urine by a first order process with a half-life of approximately 2 hours. Thus, the rapid and efficient urinary excretion of 2,4-D appears to be essentially independent of the route of administration. Further studies (Wolfe et al., 1972) have shown that the propylene glycol butyl ether esters of 2,4-D applied to the skin of rats are absorbed through the skin at a first order rate with a half-life of about 20 hours, and are then rapidly excreted as 2,4-D acid in the urine. In human volunteers (Gehring et al., 1973) given an oral dose of 5 mg 2,4-D per kg body weight, virtually the entire dose (greater than 95%) was excreted in the urine as 2,4-D and 2,4-D conjugates by a first order process with an average half-life of approximately 11 hours.

Since analysis of urine is an acceptable means of assessing the absorbed dose, it appears to be a relatively simple matter of collecting a urine sample at a pre-specified time and analyzing it for the pesticide. From our studies (Lavy, 1978) when consecutive 12-hour samples were collected, diurnal fluctuations in pesticide excretion were common among the different crewmembers.

When a specimen is collected at one specific time of day, one person may be excreting at his maximum concentration and another person at his minimum concentration. For example, one day a person excreted 6300 ml of urine while one of his colleagues employed in a similar duty excreted 606 ml. Assuming both had absorbed the same amount of pesticide, we would expect similar amounts to be excreted. If only a partial urine sample was collected and analyzed, a tenfold error would be made due to dilution. Consequently, all of the urine excreted daily must be collected and the volume recorded before an aliquot is taken for analysis.

Of primary concern in exposure studies is the amount of compound actually entering the body via ingestion, inhalation, or dermal absorption. In order to evaluate the effectiveness of patches in predicting the absorbed dose, during two studies we attached patches to the clothing at strategic locations in addition to collecting total urine samples. As an example of the fluctuation in 2,4,5-T exposure from one patch to another, Table 1 provides information derived from individual patch analyses from four mist blower crewmembers.

Table I. Micrograms 2,4,5-T detected on 100 cm^2 gauze patches of individual mist blower crewmembers.

	Chest	Back	Left arm	Right arm	Left thigh	Right thigh	Total on patches
	(μg)						
Driver I	31	102	44	57	17	101	352
Driver II	58	111	156	66	58	77	526
Mixer	74	8	2	449	108	876	1517
Supervisor	27	38	52	47	64	130	358

Although EPA estimates that 10% of the pesticide contacting dermal surfaces will be absorbed, this value will probably vary depending on compound, carrier type, formulation, the amount of moisture on the skin, which area of the body is contacted, and several other factors. In addition to analyzing for the amount of 2,4,5-T on the patches, the size of the crewmember and the amount of bare skin exposed must also be known and appropriate calculations made. Results obtained from correlating exposure information (patch vs. urine) for 57 forest workers indicate that the values were not highly correlated (Lavy, 1978; Lavy, 1980).

Table II lists the potential exposure via inhalation and dermal absorption and the amount of 2,4,5-T excreted for four of the more highly exposed 2,4,5-T crewmembers.

Table II. Levels of 2,4,5-T detected in air, patch, and urine samples for four of the more highly exposed forestry crewmembers.

Duty	Exposure	Potential exposure: Air (resin)	Potential exposure: Skin (patch)	Actual excretion (urine)
		(mg/kg)		
Backpack sprayer	1	0.00058	0.711	0.069
	2	0.00089	0.807	0.074
Mist blower driver	1	0.00019	0.179	0.042
	2	0.00040	2.987	0.032
Helicopter pilot	1	nd	nd	0.031
	2	nd	nd	0.039
Helicopter mixer	1	nd	0.085	0.071
	2	nd	nd	0.138

Exposure and Work Duty

No significant difference in exposure level occurred between work crews. Data indicate that backpack and mist blower crews received more exposure; however, this exposure was not significantly different from that of the aerial crew (Table III). Each spray operation had one mixer whose exposure level was relatively high. If he had not been included in the calculations there would have been statistical differences in the means.

Table III. Mean exposures of 2,4,5-T received as determined by urine analysis: classified by spray operation and duty of crewmember. (Modified from Lavy et al., 1980.)

Spray operation	Mean[a] (µg/kg)	Duty		Mean (µg/kg)
Backpack (7)[b]	55 a	Mixer	(4)	62 a
Mist blower (4)	44 a	Backpack sprayer	(6)	47 a
Aerial (10)	22 a	Mist blower driver	(2)	35 ab
		Helicopter pilot	(2)	22 ab
		Supervisor	(4)	11 b
		Helicopter flag-man	(4)	1 b

[a]Means within a group followed by the same letter are not different at the 0.05 significance level as determined by Duncan's multiple range test.
[b]Number of workers in the group.

Differences did, however, occur in relation to work duties within crews (Table III). Totals per exposure ranged from a high of 0.096 mg/kg (mixer) to a low of 0.001 mg/kg (flagmen). With one exception the mixer in each of the four crews showed higher exposure levels than any of his fellow crew members. These three mixers also had higher 2,4,5-T excretion values on day 0 than others in their crew probably because they mixed the 2,4,5-T the day before the actual spray occurred. Optimum preexposure data would have required the mixers to begin urine collection at least 1 day earlier. The fact that the one exceptional mixer endorsed cautious work habits and wore gloves may account for the comparatively low level of 2,4,5-T measured in his urine.

Categorized by work duties, mixers (those handling concentrate) received the highest internal dose of 2,4,5-T, followed in order by backpack sprayers, mist blower drivers, helicopter pilots, supervisors, and flagmen for the helicopter operation. One helicopter pilot excreted considerably less 2,4,5-T in his urine than did the other pilot. This difference appeared to be related to the fact that the second pilot routinely checked and unplugged nozzles at each fill-up time. In addition, he helped change the spray boom on the helicopter before and after each spray period.

2,4-D Study

Although none of the 2,4,5-T crewmembers received doses approaching health endangering levels, some of the crewmembers received considerably more exposure than others. The study using 2,4-D was designed to give us additional data on exposure under routine operations (T_1) and to see if the use of protective clothing and special precautions (T_2) could be employed to decrease exposure.

Even in the T_1 study, levels were so low that there was hardly a possibility of noting significantly reduced exposure in the T_2 test where protective measures were taken. In spite of the low levels of exposure, there was still a relationship between exposure and workers duties as had been evident in the 2,4,5-T study.

Less than 30% of the 524 urine samples analyzed contained levels of 2,4-D above the 0.04-ppm detection limit. Table IV reveals that most of the positive samples were from the crew members most closely involved with the actual spraying (batchman-loaders, pilots, and mechanics). Except for one pilot who had assisted in cleaning spray nozzles, batchman-loaders and mechanics showed the highest levels of 2,4-D in the urine, while observers received the lowest levels. Urine samples from observers standing near the heliport rarely contained any 2,4-D and then in only negligible amounts approaching the limit of detection. The only supervisor excreting 2,4-D was probably exposed when the automatic transfer system for moving the concentrate from the barrels to the mix truck failed and he helped manually transfer the chemical with buckets during the T_1 application. Similar exposure did not occur during T_2, and no 2,4-D was detected in his urine in T_2 (Table IV).

Nash et al. (1982) studying the exposure of ground applicators to 2,4-D found maximum mean one-day 2,4-D urinary excretion of 0.002, 0.003, and 0.004 mg/kg body weight, respectively, for applicators, mixer/loaders, and mixer/loader/applicators from a one-time exposure. When aerial application was used they found from 0.006 mg/kg body weight for pilots to 0.02 mg/kg body weight for mixer/loaders. The Nash study was conducted with applicators of 2,4-D in wheat fields. They found levels similar to those exposure levels found in forest operations in Arkansas, Washington, and Oregon (Lavy et al. 1980, Lavy et al. 1982). Newton and Norris (1981) pursued additional studies on dose absorbed by applying known quantities of 2,4,5-T to human skin. They found 2,4,5-T excretion rates which were similar to those we found in the field studies.

Table IV. Comparisons of total dose 2,4-D received by workers during normal operations (T_1) and "protective clothing" operations (T_2). (Modified from Lavy et al., 1982.)

Worker duty	Dose mg/kg[a] ± SD	
	T_1	T_2
Pilots (3)[b]	0.0198 ± 0.310	0.00854 ± 0.01316
Mechanics (3)	0.00545 ± 0.00712	0.00301 ± 0.00269
Batchmen (3)	0.0196 ± 0.0018	0.0140 ± 0.0117
Supervisors (3)	0.00231 ± 0.00400	0.000013 ± 0.00022
Observers (6)	0.00049 ± 0.00059	0.00009 ± 0.00023
Total dose	0.00802	0.00429

[a]Values include 2,4-D excreted on the spray day plus 5 days following.
[b]Number in parenthesis represents the number of workers in the group.

This study found that some crew members involved in the aerial application of 2,4-D for forestry purposes absorbed low levels of 2,4-D, but the doses as indicated by urine analyses were several orders of magnitude below the 24 mg/kg no-observable-effect-level determined in toxicology studies. These results are in agreement with those of Nash et al. (1982). The doses were comparable to those found in an earlier test involving aerial application of 2,4,5-T but were substantially lower than those found for ground application of that herbicide (Lavy et al., 1980).

The absorbed dose measured in this study, as shown by the urine analyses, were too low and the replications too limited to allow accurate statistical comparisons for each worker duty. However, the total dose absorbed by workers in T_1 was nearly double that of workers wearing the protective clothing in T_2.

If one assumes a no-observable-effect-level of 24 mg/kg of body weight, as determined from toxicology tests with laboratory animals, then safety factors for the categories of workers involved in this test are substantial (Hall, 1980). They ranged from 1212 for the pilots and batchmen in T_1 to 266,667 for the observers in T_2.

The literature contains reports of many exposure studies. To conduct a good exposure study requires considerable forethought, an in-depth literature search, detailed protocol development, and extensive planning. Even then loopholes may exist. Findings from our studies reveal shortcoming in the following areas:

1. Inadequate pre-exposure information. Although workers fill out questionaires indicating that they have not used phenoxy herbicides during the previous two weeks, sometimes these workers come into the study with positive background levels of phenoxy in their urine.

2. Lack of ensuring that there is no post-application exposure. Excretion curves for several workers in our studies indicate that occasionally some avenue of re-exposure occurs after the actual spray day.

The source of the exposure either before or after the actual spray date appears to be related to some contact with the phenoxys of which the crewmember was not aware. Possible avenues of re-exposure include wearing phenoxy contaminated clothing on days other than the planned spray day, i.e., gloves, boots, pants, shirts, or chaps. Workers may also have received some exposure from their phenoxy application equipment. This may occur if a worker has a spray operation scheduled and wants to clean or check his equipment in advance. Another potential source of exposure is the vehicles in which the workers ride. Often workers, pesticide concentrate, empty containers, and equipment are hauled in the same pick-up truck or van.

Due to our awareness that extraneous exposure can occur, we have taken measures to limit these types of pre-exposure in our most recent studies. The data we have collected supply adequate evidence that extraneous means of exposure are common. If it occurs in these phenoxy studies, it is likely that it occurs for workers applying more toxic pesticides.

Including this extraneous exposure, the degree of safety that we calculated for forest workers using phenoxy herbicides was such that even the most highly exposed crewmembers received exposure which was several orders of magnitude below the no-observable-effect-level. Decreases in the level of exposure with the use of protective measures, however, may be of real consequence to workers applying more toxic materials.

We suggest that exposure to any pesticide may be decreased with the following precautions:

1. Wear clean clothing
2. Wash or shower soon after application
3. Launder clothes properly

4. Do not use tobacco while working
5. Wear gloves impervious to chemicals
6. Cover bare skin areas
7. Know factors contributing to exposure

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Literature Cited

Draper, W. H. and Street, J. C. 1982. J. Environ. Sci. Health B17(4)321-339.

Durham, W. F. and Wolfe, H. R. 1962. Bull. WHO 26: 75-91.

Gehring, P. J.; Kramer, C. G.; Schwetz, B. A.; Rose, J. Q.; and Rowe, V. K. 1973. Toxicol. Appl. Pharmacol. 26: 352.

Hall, J. F. 1980. Preface letter in Lavy, T. L., Ed. Project Completion Report submitted to National Forest Products Association; Washington, D.C., 1980. pp. 1-6.

Kolmodin-Hedman, B.; Erne, K.; Hakansson, M.; and Engquist, A. 1979. Arbete och halsa, Vetenskaplig Skrift Serie No. 17.

Lavy, T. L. 1978. Measurement of 2,4,5-T exposure workers. Project Completion Report submitted to National Forest Products Association; Washington, D.C. 1978.

Lavy, T. L. 1980. Determination of 2,4-D exposure received by forestry applicators. Project Completion Report submitted to National Forest Products Association; Washington, D.C. 1980.

Lavy, T. L.; Shepard, J. S.; and Mattice, J. D. 1980. J. Agric. Food Chem. 28: 626-630.

Lavy, T. L.; Walstad, J. D.; Flynn, R. R.; and Mattice, J. D. 1982. J. Agric. Food Chem. 30: 375-381.

Leng, M. L.; Ramsey, J. C.; Braun, W. H. and Lavy, T. L. 1980. Chapter II in Pesticide Residues and Exposure, American Chemical Society Symposium Series 182.

Nash, R. G.; Kearney, P. C.; Maitlen, S. C.; Soll, C. R.; and Fertig, S. N. 1982. Chapter 10 in Pesticide Residues and Exposure, American Chemical Society Symposium Series 182.

Newton, M. and Norris, L. A. 1981. Fund. Appl. Toxicol. 1: 339-346.

Sauerhoff, M. W.; Brun, W. H.; Blau, G. E.; and Gehring, P. J. 1977. Toxicology 8: 3-11.

Tarrant, R. F. and Allard, J. 1972. Arch. Environ. Health 24(4): 277-280.

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