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### Case Studies: Forest Worker Exposure to Garlon™ 4 Herbicide

Dawn Tharr Column Editor

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## Case Studies

### Forest Worker Exposure to Garlon™ 4 Herbicide

*Dawn Tharr, Column Editor*

**Reported by P. Middendorf, C. Timchalk, B. Kropscott, and D. Rick**

#### Background

Garlon™ 4, a triclopyr herbicide, is used by the U.S. Forest Service for forest vegetation management. Specifically, Garlon 4 is used for directed foliar application, a process by which applicators walk through an area spraying the foliage of undesirable plant species. The Forest Service contacted the Georgia Tech Research Institute to prepare a worker exposure study protocol and conduct field studies to evaluate worker exposures to the herbicide. The objective of the study was to determine mixer/applicator exposures and doses to Garlon 4 herbicide during directed foliar application when using backpack sprayers to apply the herbicide. Additional objectives included the identification of potential routes of exposure as well as work practices and procedures that contribute to the overall exposures.

#### Introduction

The butoxyethyl ester of triclopyr (3,5,6-trichloro-2-pyridinyloxyacetic acid) is the active ingredient of Garlon 4. Triclopyr has a low acute toxicity via oral or percutaneous exposure routes.<sup>(1)</sup> In one study, only 1.65 percent of the dermal dose applied to human volunteers was absorbed through the skin in 8 hours and the absorption was slow with a half-life ( $t_{1/2}$ ) of approximately 16 hours. Once absorbed, triclopyr was rapidly eliminated, primarily in the urine with a  $t_{1/2}$  of 5 hours.<sup>(2)</sup> The results of the study indicate that triclopyr has a low potential to accumulate in humans or to be absorbed through the skin in acutely toxic amounts. In addition, triclopyr is not carcinogenic in rats or mice in lifetime feeding studies.<sup>(3)</sup> No signs of overt toxicity, gross or

microscopic pathological changes were observed in dogs fed triclopyr diets for 1 year. However, renal phenol-sulfonphthalein excretion was decreased in the 25 and 50 mg/kg body weight/day groups due to competitive inhibition of the renal excretory system by triclopyr.<sup>(4)</sup> Based on these data, the no-observed-effect level (NOEL) for triclopyr was set at 25 mg/kg/day, which is equivalent to 175 mg/day for a 70-kg person.

In this study, forest worker exposures were monitored during the mixing and directed foliar application of Garlon 4 herbicide. Opportunity for exposure among pesticide applicators may occur during handling of the herbicide concentrate and during preparation and application of the spray mixture. As a result of the relatively low vapor pressure of Garlon 4, the potential for exposure via inhalation of vapors is low; however, this does not eliminate the potential for exposure from aerosols. Exposure via the dermal route may occur following accumulation of the herbicide on the skin surface either directly or after penetration

through protective clothing. Oral exposure could occur from handling food, beverages, or tobacco products with contaminated hands, as well as from touching the mouth with contaminated hands.

During directed foliar application, herbicides are sprayed from a pressurized backpack through a hand-held spray gun system. During this study, application methods were to follow label instructions; the backpack sprayers, spray guns, nozzles, and mixing equipment used in the study were those currently used by the U.S. Forest Service for forest applications. Volunteers were to be trained and to wear personal protective equipment in accordance with U.S. Forest Service Guidelines.

Garlon 4 herbicide for the study was supplied by Dow Elanco in 2.5-gallon containers which are normally used by Forest Service workers. Three methods were used to dilute the herbicide to a 3 percent solution of Garlon 4 herbicide (concentrate) in water. These methods and the application equipment utilized by site are summarized in Table I.

**TABLE I. Environmental, Site, and Application Conditions During Directed Foliar Application of Garlon™ 4 Herbicide to Forest Sites**

	Site 1	Site 2	Site 3	Site 4
Weather	Clear, dry 81–93°F Low wind	Clear, dry 71–93°F Low wind	Clear, dry 78–89°F Low wind	Overcast, dry 72–89°F Low wind
Vegetation	Low height (2–3 ft) Low density	Medium to tall height (4–8 ft) Medium density	Medium to very tall height (4–12 ft) High density	Medium to tall height (4–6 ft) Medium density
Mixing	Mixed concentrate in 50 gallon drum	Mixed concentrate into backpack	Mixed concentrate in 5 gallon jugs	Mixed concentrate into 50 gallon drum
Adjuvant	None	None	2.5 ounces Cide-Kick per 3 gallons mix	16 ounces Cide-Kick per 50 gallons
Application Equipment	Wand applicator tip #2503	Model 30 gunjet tip #2503	Model 30 gunjet tip #2503	Model 30 gunjet Wand applicator tip #2503

The Forest Service supplied and required all volunteers to wear tightly woven, prewashed, long-sleeved shirts and long pants. All volunteers also wore leather boots and a hard hat. Gloves were available for use at each site during applications; their use was required when handling the concentrate. The clothing met Forest Service Guidelines.

## Methods

Four survey sites were selected. The choice of forest sites, application procedures, and work practices were determined by the local Forest Service representatives. The environmental, site, and application conditions are summarized in Table I.

A crew of Forest Service employees or contractors who apply herbicides in the forest as part of their job requirements was formed at each location. The crews participated in an open meeting to discuss the purpose of the study, the conduct of the study, and how the results were to be used. As an incentive to collect their urine, each volunteer was given \$100 after the study.

Two types of exposure assessments were made during the mixing and application of Garlon 4 herbicide.

- Exposure monitoring of 1) air concentrations in the breathing zones and 2) body surface deposition were used to estimate inhalation and dermal exposures, respectively, of volunteers.
- Biological monitoring of triclopyr in the urine was conducted to estimate the actual amount of herbicide excreted by the volunteers.

Three types of samples were taken in the field to estimate exposures: patch samples to estimate skin deposition, air samples to estimate inhalation exposure, and hand wash samples to estimate skin exposure on the hands.

Patches were 10 cm by 10 cm squares made of griegge fabric supplied by Thomaston Mills, Inc. Griegge fabric was used instead of the typical gauze material because the forest vegetation was likely to rip the gauze. Patches

were backed with glassine paper and attached with safety pins to 11 different locations on the clothing. In addition to the outside patches, patches were attached beneath the outer clothing to estimate clothing penetration. Two of the volunteers from each of Sites 1, 2, and 3 wore three patches underneath their clothing, while at Site 4 all volunteers wore five patches underneath their clothing.

Air samples were taken with a sampling train of a constant flow pump attached to a 150-mg Chromosorb 102 tube, preceded by a 37-mm 0.8  $\mu$  mixed cellulose ester fiber filter. The filters were placed in the breathing zones of the volunteers. Air samples were taken for the duration of the mixing and application procedure. During the application period, hand rinse samples were taken before volunteers consumed food or handled urine containers; this was usually just before lunch. Hand rinse samples were also taken at the end of the application period before leaving the site.

Volunteers were instructed to collect all urine voids for 5 days: the day before application, the day of application, and for three full days after application. The voided urine for each volunteer was pooled into two samples for each 24-hour period.

Each of the patch and air samples was extracted in high-performance liquid chromatography-grade toluene. An aliquot from each extract was analyzed by gas chromatography with either a flame ionization detector or an electron capture detector.

The hand wash samples were analyzed for both the butoxyethyl ester (BEE) of triclopyr and the triclopyr acid (ACID) using two different preparative and analytical procedures. To prepare the samples for BEE analysis, an aliquot of the detergent solution was extracted with a solution of ethyl acetate/isopropanol and then analyzed by gas chromatography. For analysis of the hand wash solutions for the ACID, aliquots were analyzed by reverse-phase high performance liquid chromatography with UV detector.

Urinary triclopyr was measured by

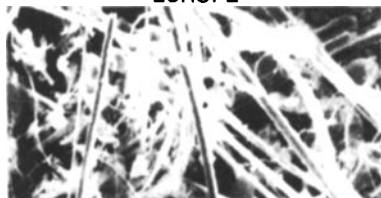
gas chromatography/mass spectrometry. An aliquot of urine was mixed with 6 N HCl and heated. To serve as an internal standard, 3,5,6-trichloro-2-pyridinyloxypropionic acid in ethyl acetate was added. Samples were methylated with diazomethane. Aliquots of the concentrated ethyl acetate extract were analyzed by gas chromatography with a mass selective detector. Each urine sample was also analyzed for creatinine using a modification of the JAFFE reaction.<sup>(9)</sup>

Two methods were used to calculate the amount of triclopyr absorbed by each volunteer. In the first method, the amount of triclopyr absorbed was calculated by dividing the total amount of triclopyr excreted in the urine by a correction factor of 0.789 obtained from human pharmacokinetic studies.<sup>(9)</sup> For volunteers who had three or more urine specimens containing triclopyr above detection limits, a second estimate was obtained by using a pharmacokinetic model. The amounts of triclopyr found in the urine were fit to the one-compartment pharmacokinetic model to describe the pharmacokinetics of dermally administered triclopyr in human volunteers. In general, the larger of the two estimates of absorbed triclopyr was used to represent each volunteer's dose.

Estimates of dose were obtained for the inhalation and dermal routes of exposure. The results of the breathing zone air samples were multiplied by an assumed breathing rate of 20 L/min, and the amount of triclopyr in that volume of air was assumed to enter the blood at the lungs. To estimate the dermal dose, patches were used to represent deposition of triclopyr on various areas of the body. The amounts of triclopyr measured on the patches were scaled by surface area factors. The amounts of triclopyr on clothed areas were multiplied by a clothing penetration factor, assumed to be 10 percent, to determine the amount of triclopyr on the skin. The estimate of the total dermal exposure was the sum of the dermal exposure and hand rinse exposure multiplied by the skin absorption factor of 1.65 percent.

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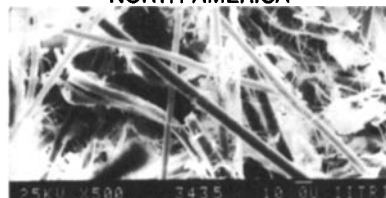
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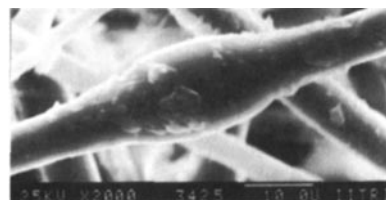
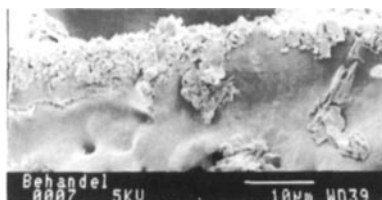
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### Results

At each worksite, the entire day was spent preparing for application, applying herbicide, or cleaning up after the application; therefore, the measured exposures and doses are based on a full work shift. Several work practices and procedures potentially affecting exposure were tracked for each volunteer including: personal hygiene; training and experience; use of tobacco products; application equipment; equipment malfunction; personal pro-

TECTIVE equipment; the amount of herbicide applied; and site conditions.

The mean dose for each treatment site was calculated as a geometric mean because exposure data are usually log-normally distributed. One of the 22 volunteers had triclopyr in his prespray urine as well as a large increase in the amount of triclopyr in his urine on the third day after spraying, suggesting he had been exposed to triclopyr shortly before the study began and during the elimination phase of the study. The triclopyr in his

urine was not successfully modeled to eliminate the two additional exposures, and his results were excluded from the analysis. The results of the biomonitoring dose are summarized by site in Table II. The overall geometric mean of the biomonitoring dose of triclopyr from all four sites was 106 µg, a factor of 158 times less than the NOEL observed in animal studies. Six of the 21 volunteers had doses greater than 1 percent of the NOEL. Of the six, only one did not have an attributable, controllable reason for the dose. The esti-

TABLE II. Application Rates and Doses of Triclopyr

Site	Application Rate (lbs a.i./acre)	Number of Volunteers	Estimated % of Dose From Dermal Route	Estimated % of Dose From Inhalation	Biomonitored Dose of Triclopyr (µg) Range	Geometric Mean	Geometric Mean Dose as % of NOEL
1	10.5	5	85.8	14.8	207 - 1459	591	0.34
2	13.2	5	80.2	19.8	374 - 2106	947	0.54
3	7.2	5	90.9	9.1	900 - 7690	2889	1.65
4	8.3	6	87.5	12.5	267 - 3737	955	0.55
Overall		21	86.2	13.8		1106	0.63

mates of dose by the dermal route consistently dominated the total dose estimate. The average estimated dose from the dermal route was 86.2 percent of the total estimated dose.

## Discussion

An analysis of factors at the various sites indicated that site conditions such as the height and density of vegetation, work practices (e.g., the use of gloves), mixing procedures, and equipment maintenance as well as experience and adherence to established procedures affected the doses.

## Weather

Weather conditions at all four sites were very similar and did not allow for an analysis of the effects of weather on the observed doses of triclopyr. However, because the temperatures were near the upper limit of the recommended application conditions, inhalation exposures for each site should be at the upper range of expected inhalation exposures for the site and application conditions.

## Site Conditions

The geometric mean of triclopyr doses at each site increased as the vegetation height and density increased. The shortest, sparsest vegetation was found at Site 1 where the geometric mean of triclopyr doses was 591  $\mu\text{g}$ . The conditions at Sites 2 and 4 were similar and had taller vegetation and denser underbrush than Site 1. At these sites, the geometric means of biomonitored doses of triclopyr were 962  $\mu\text{g}$  (Site 2) and 955  $\mu\text{g}$  (Site 4). The doses were largest at Site 3 which had the tallest vegetation and densest underbrush.

The pattern of deposition of triclopyr onto patches (data not presented) was consistent with the differences in vegetation. As the height of vegetation and density of underbrush increased, the amount of deposition on all patches increased. This is expected because taller vegetation requires the volunteer to spray higher to reach the growing tips and leads to greater

overspray and possible drift. Denser vegetation increases the chances that the volunteers will brush against already treated vegetation. Both of these factors lead to greater dermal exposure. The highest estimated percent of dose from the dermal route was observed at Site 3.

Spraying taller vegetation can also lead to greater inhalation exposure, which is seen in the air sampling results where inhalation exposures generally increased with the height of vegetation. An exception is that the volunteers at Site 2, where the vegetation height was similar to Site 4, had the highest overall inhalation exposure. This may be explained, however, by the amount of material applied. The amount of active ingredient applied per person at Site 2 was approximately twice the amount applied at either Site 3 or Site 4. Other factors affecting inhalation and dermal exposures are overspray and drift. Drift should have been minimal because the wind was not strong on any of the four days of application. The position within the group relative to where other volunteers are spraying may lead to variations in exposure. Volunteers downwind or near to and in the spray direction of another volunteer could have been exposed to larger amounts of triclopyr.

## Gloves

The volunteers wore different types of gloves at each of the sites, and the differences in the ability of triclopyr to penetrate the various materials and thicknesses may partially explain the observed differences in hand exposures between sites. The hand exposures at Site 3 were the highest, and the three volunteers who did not wear gloves had the highest hand exposure at this site. The volunteer who had the greatest hand exposure worked on his gunjet several times and also had a leak in a hose connection that had to be fixed. Two of the three volunteers also handled the concentrate, which may have increased hand exposures. All of the volunteers at Site 2, where the second largest amounts of triclopyr in hand rinses for all sites were found,

wore latex gloves, but each volunteer also handled the concentrate. The average hand exposure at this site was much higher than at Site 1 or Site 4 where the concentrate was handled only once by one or two volunteers. At Site 1, the volunteers all wore new leather gloves while applying. Their hand exposures were the least for all sites in the survey. It would be expected that the leather could become saturated with the herbicide mixture and expose the hands. However, the gloves may have acted to absorb small amounts of the mixture and retained it, thus minimizing exposures. Reuse of the same gloves may lead to increased hand exposures because of the contamination. Only at higher amounts would the material penetrate to the skin during one application. Alternatively, the low hand exposures may be the result of the use of wands rather than gunjets at this site. This is discussed in the section on Equipment.

## Training and Experience

Training and/or experience in applying was a potential contributing factor to doses of triclopyr. Only two volunteers did not have previous experience or did not apply on a regular basis. These two individuals had the highest doses observed in the study. Factors related to inexperience which may have contributed to their large doses were handling the concentrated formulation without gloves and not using gloves while spraying. In addition, one of these two volunteers had equipment problems; there was a leak in the piston pump and his gunjet stopped up twice, which may have been related to lack of training with the equipment and how to fix the equipment when problems arose.

## Equipment

Equipment failure generally led to larger exposures. The piston pumps generate a higher internal pressure than diaphragm pumps, which may explain the greater number of leaks with piston pump backpacks. However, the diaphragm pumps were generally newer, so the age of the piston pump

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backpacks may have contributed to the increased failure rate. The geometric mean of the doses of triclopyr for volunteers who used piston pump backpacks, which tended to leak, was 24 times larger than for those who used diaphragm pump backpacks (see Table III).

The gunjets jammed for several volunteers, while the wands did not. When the gunjets jammed, volunteers usually handled the gunjets with bare hands to fix them. The wands also allow the volunteer to spray the same vegetation with the herbicide release point at a greater distance from the body, which may account for some of the lower exposures at Site 1 compared to Site 2. The geometric mean of doses for volunteers who used wands was approximately one-half that for those who used gunjets. This result may be highly related to other site factors, however, since all Site 1 volunteers used the wands, while all Site 2 and Site

**TABLE III. Comparison of Factors Across Sites Associated with Exposure of Triclopyr During Backpack Directed Foliar Application of Garlon™4 Herbicide to Forest Sites**

Source	Number	Geometric Mean of Dose (μg)
Backpack		
Diaphragm	18	0.982
Piston	3	2.38
Handle concentrate		
Yes	10	1.19
No	11	1.05
Use tobacco		
Yes	11	1.01
No	10	1.24
Regularly apply		
Yes	19	0.898
No	2	8.62
Applicator		
Wand	8	0.655
Gunjet	13	1.55
Gloves		
Yes	18	0.884
No	3	4.48
Leaks		
Yes	6	1.42
No	15	1.01

3 volunteers used gunjets. Only at Site 4 were both used, and no clear association of dose level with applicator type was observed.

### Mixing

The biomonitored doses of triclopyr were lowest in volunteers at Sites 1 and 4 where the herbicide concentrate was handled by a limited number of volunteers and only when mixing a large drum. The biomonitored doses at Site 2 were very similar to Site 4, while Site 3 was much higher; this suggests that the mixing method may affect dose, but the specific procedures and personal protective equipment used may affect the dose more. In any case, the method that provides the lowest potential of increased dose should be chosen. The geometric mean of the doses of volunteers who handled the concentrate was 13 percent larger than those who did not handle the concentrate (Table III).

## Adjuvant

Adjuvant was used on Sites 3 and 4, and the dermal doses might be expected to be greater for these sites because the adjuvant reduced surface tension in the droplets, allowing them to spread out and increase the surface area of the skin covered. However, additional adjuvant does not appear to be a major factor in causing additional doses because of the small difference observed between biomonitoring doses at Site 4, where adjuvant was used, and Sites 1 and 2, where adjuvant was not used. The higher doses at site 3 are likely to be attributed to other factors. When the mixer at Site 3 is removed from the Site 3 mean of dermal doses, the differences between Sites 2 and 3 are eliminated.

## Use of Tobacco

The use of tobacco products during application was expected to increase the potential for ingestion of triclopyr and would tend to increase the volunteer's dose. However, the geometric mean of the doses of triclopyr was actually less for those who used tobacco than for those who did not, as was the estimated oral component of dose. Good industrial hygiene practice, however, dictates that food and beverages should not be ingested, and other materials that are put in the mouth such as tobacco products and chewing gum should not be used while applying herbicide so that doses from this route are minimized.

## Summary

The factors potentially contributing to dose are many and interrelated. In this study, the various factors that may contribute to dose cannot be rigorously analyzed by standard statistical techniques because the factors were not able to be standardized across all sites. However, several factors appear to emerge as influencing exposure and doses to triclopyr:

- height and density of vegetation
- training and experience

- use of gloves
- mixing procedures
- equipment maintenance

In directed foliar applications for forest pest management, the applicators' doses of triclopyr can be minimized as long as a number of work practices are controlled. Biomonitoring doses and exposures were all below the NOEL; however, doses could be reduced even further if several modifications are implemented by the Forest Service, as described below.

1. The sites should be chosen so that the average height of the vegetation does not exceed 6 feet. This recommendation is consistent with the Garlon 4 herbicide product label. Generally, the sites treated sooner after replanting will have shorter vegetation, and the underbrush will be less dense resulting in lower doses. The optimal time for site treatment should be determined using minimization of potential exposures as one of the primary criteria.
2. All workers who apply pesticides and crew leaders should be thoroughly and formally trained in the goals of herbicide application and procedures for mixing and application which include handling procedures to minimize the potential for exposures. Crew leaders should have additional training and evaluation to ensure they are competent to lead an application crew. Following safety and health procedures should be a part of the annual personnel evaluation process.
3. An operations and maintenance plan for the spray equipment should be developed and implemented. Equipment should be maintained in excellent operating condition. Only low pressure (diaphragm pumps) should be used. The seals should be inspected and replaced periodically to minimize the possibility of seal breakage and leaks. Wands should be used whenever possible rather than gunjets.
4. Personal protective equipment

should be worn during the mixing and application of Garlon. Nitrile gloves or other gloves with similar resistance to triclopyr penetration should be worn when mixing, applying, and handling gunjets or wands. An apron and chemical goggles or faceshield should be worn by the mixer while handling the concentrate to minimize skin and eye contact.

5. Personal hygiene should be stressed for workers who apply pesticides. In the event that a backpack leaks, or another accident occurs which results in a substantial contamination of the work clothing, soap and water should be available to wash the affected areas thoroughly. A change of clothing should be available for each member in the crew.

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