RESIDUES OF FORESTRY HERBICIDES IN PLANTS OF INTEREST TO NATIVE AMERICANS IN CALIFORNIA NATIONAL FORESTS

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STATE OF CALIFORNIA
California Environmental Protection Agency
Department of Pesticide Regulation
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Executive Summary

Purpose

The purpose of this study was to determine dissipation and off-site movement of four forestry herbicide products containing glyphosate, triclopyr, or hexazinone on native plant materials. This information will be used to determine if California Indians gathering and processing plant materials from inside and outside herbicide treatment areas are at risk from pesticide exposure.

Background

The United States Forest Service is responsible for conifer reforestation in units of land within the National Forests that have burned due to forest fires or have been heavily logged. Glyphosate, triclopyr, and hexazinone are commonly applied to barren sites as pre-plant herbicides to eliminate unwanted plants prior to conifer planting, and also as post-plant herbicides to control re-emerging weeds in young established conifer plantations. Typically, glyphosate is applied as the liquid Accord®, triclopyr as the liquid Garlon® 4, and hexazinone as the liquid Velpar® L and also as the granular Pronone® 10G. With the exception of granular hexazinone, which can by applied by ground or air (helicopter), all other herbicide products are ground applied by spray crews carrying backpack sprayers to treat targeted plant pests.

National Forests and other public lands are used by many California Indians as sites for gathering native plant materials for food, medicine, ceremonies, or basketry purposes. Herbicide exposure may occur to those gathering plants within or adjacent to treatment areas; therefore, California Indians are concerned with possible health risks associated with the traditional gathering, processing, and consuming of plant materials. Forestry herbicide registrants have not been required to address these unique exposure scenarios when registering herbicide products within the state. The United States Forest Service and the California Department of Pesticide Regulation recognize that plant-herbicide residue information is lacking for tribal cultural activities involving treated vegetation. Consequently, this forestry herbicide study was conducted to gather residue information on selected plants of importance to California Indians.

Study Methods

During 1997 to 2001, ground applications of glyphosate, triclopyr, and liquid hexazinone, and aerial applications of granular hexazinone were evaluated for resulting herbicide residue levels in bracken fern roots (rhizomes), buckbrush shoots, golden fleece foliage, and manzanita berries growing in herbicide application areas of the Eldorado, Sierra, and Stanislaus National Forests located in California.

Herbicide dissipation rates were determined for each plant-part and herbicide-product combination. Plant parts were collected at seven intervals: within 1-3 days (application-day sample), and 3-5, 7-9, 11-13, 19-21, 27-29, and 35-37 weeks following herbicide application for each herbicide product. Additional samples were collected beyond 35-37 weeks if herbicide residues persisted. Off-site herbicide movement for each herbicide product was also documented following application by analyzing plant material growing at specific intervals up to 100 feet (ft) down slope from the treatment edge.

Results

One to three days following application, the mean application-day herbicide concentration ranges in treated plants were 0.5 to 241 ppm for glyphosate, 0.2 to 81 ppm for liquid hexazinone, 0.1 to 19 ppm for triclopyr, and not detected to 0.3 ppm for granular hexazinone treated plants. Maximum residue levels for foliar-applied herbicides were generally observed on either application day or at week 4, the second sampling interval. Due to direct deposition of spray materials on shoots, foliage, and berries, and also possible rapid foliar uptake and transport to roots, plants receiving foliar herbicide treatments resulted in higher application-day residue levels than those plants treated by the granular herbicide applied by air. High glyphosate spray-day residue levels were attributed to application method in conjunction with a higher application rate than used for the other herbicides. Overall, plants treated with glyphosate, triclopyr, and either formulation of hexazinone showed a general concentration decline in roots, shoots, foliage, and berries during the study period.

Maximum residue levels for aerially applied granular hexazinone were reached at week 8 post-application after the compound was washed into the soil, absorbed by roots, and translocated within the plant. In contrast, maximum residue levels for liquid hexazinone treated plants were typically observed in application-day samples. However, at week 28 post-application, plants treated with either the granular or liquid hexazinone formulation showed similar residue levels in sampled roots, shoots, and foliage.

The longest observed dissipation period for an herbicide residue level to reach the non-detectable level commencing from the herbicide's maximum concentration in a plant medium was 130 weeks for shoots of liquid hexazinone-treated buckbrush. Herbicide half-life averages in plant media ranged from to 1 to 19 weeks. In general, buckbrush and bracken fern plants treated with liquid hexazinone showed the longest half-lives of 18 and 19 weeks, in buckbruch shoots and bracken fern roots, respectively, compared to plants treated with the other three herbicides products.

Residues of glyphosate, triclopyr, and granular hexazinone were detected in off-site plant materials sampled 1 to 3 days following application. Application-day samples showed the presence of triclopyr up to 50-100 ft down slope from the treatment sites. Glyphosate residues were also detected off-site in application-day samples collected at the 5-15 ft distance from the treatment edge. Glyphosate is suspected to have drifted distances equivalent to that of triclopyr at sites where both chemicals were co-applied and co-monitored, but glyphosate was probably undetected due to its higher MDL.

Residues from liquid hexazinone application were observed in off-site plant samples collected 12 weeks following treatment. It is possible that herbicide residues were transported off-site with runoff water/snowmelt and subsequently taken up by roots and translocated to sampled plant parts. Granular hexazinone was also detected up to 50-70 ft off-site on application day. It is possible that herbicide dust from aerial application may have drifted off-site from the treatment area. Overall, since residues were only detected in 8% of 240 off-site samples collected, off-site movement from drift and runoff was low.

Conclusions

Due to varied environmental conditions, different plant growth stages, and time of herbicide applications, results were highly variable. In general, low residue levels were detected in the roots, shoots, foliage, and berries of plants treated with granular hexazinone and also in roots of bracken fern treated with glyphosate, triclopyr, or liquid hexazinone. Although levels were low, residues persisted in many of the sampled media, with glyphosate remaining detectable in bracken fern roots at 67 weeks post-application, the last sampling period for that plant-herbicide combination.

Also, gatherers sampling shoots, foliage, and berries in glyphosate, triclopyr, or liquid hexazinone treatment areas may be exposed to herbicide. The highest residue levels were generally observed on application day or 4 weeks following application (second sampling interval) with residues remaining detectable in plant materials for several weeks thereafter. Consequently, herbicide residue data should be used for exposure assessment to determine if gatherers and basketweavers are exposed to hazardous levels of the four forestry herbicides.

As herbicide residues were found to move off-site to non-treatment areas, plant gatherers and basketweavers may want to select plants beyond the 100 ft down slope from treated areas for up to 12 weeks following treatment.

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Disclaimer

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INTRODUCTION

The United States Department of Agriculture Forest Service (US Forest Service) is responsible for reforestation within each of the National Forests. In general, units of land with fire-killed trees or land that has been heavily logged for timber sale do not meet the requirements of the National Forest Management Act. That act prescribes that the National Forests be maintained with the proper tree species, density, and growth rate (US Forest Service, 1994). Natural reforestation is a very slow process occurring over many decades. To quickly achieve maximum benefits equivalent to that of pre-fire or pre-logging status, human intervention is needed to reforest the land so that it may provide valuable renewable resources such as timber, watershed, wildlife, fish and wilderness (US Forest Service, 1994).

Most reforestation activities involve the use of herbicides applied at specific times to eliminate shrubs, grasses and other unwanted vegetation that would interfere with conifer growth (McWhorter and Gebhardt, 1987). These plants are undesirable because they compete for sunlight, nutrients, and soil moisture that are needed for young conifers to survive (US Forest Service, 1994). Reforestation herbicides such as glyphosate, triclopyr, and hexazinone are very effective in vegetation control. They have been used to prepare units of land prior to conifer planting ("site preparation"), and/or to eliminate re-invading plants in units with young established conifers ("conifer release"). In conifer release treatment units, plants growing within a five-foot (ft) radius around the conifer tree are treated with herbicide, while in site preparation units plant pests are treated in the entire unit.

Glyphosate, as Accord®, is a liquid formulated herbicide. It has non-selective, non-residual, post-emergent activity and may be applied to foliage at any growth stage. It is effective at controlling perennial, deep-rooted grasses, woody brush, and broadleaf weeds. Triclopyr, as Garlon® 4, is an emusifiable concentrate formulation that may be applied directly to foliage or soil where it is moved to the roots by rainfall to control woody brush and perennial broadleaf weeds. It provides little control on grasses (Byrd et al., 1974). Two formulations of hexazinone, Velpar® L and Pronone® 10G, are a liquid and granular formulation, respectively. The liquid formulation can act as a foliar contact or systemic herbicide and the granular formulation as a soil residual herbicide. It is effective for controlling broadleaf trees and bushes, as well as annual and perennial weeds (Gana, 1997). Hexazinone can be tolerated by many conifers without sustaining damage (Ashton and Craft, 1981).

Native Americans gather plant materials for food, medicine, ceremonies or basketry in or near National Forests and other public lands (Goode, 1992). Herbicide exposure may occur when they collect and handle plants from herbicide treated units. In California, the US Forest Service and some Native American Indians (California Indians) have begun a cooperative program to identify and protect sensitive gathering sites from herbicide application. This eliminates or reduces herbicide exposure for persons involved in the program. However, California Indians not participating in the cooperative program may be unintentionally exposed. Some of those not participating are, of course, unaware the program exists. There are others who choose not to disclose their gathering locations to the US Forest Service as these locations are considered sacred sites passed down in the family, clan or teacher (Goode, 1992). Consequently, those California Indians would like to know what health risks are associated with gathering, processing, and consuming plant materials from inside and adjacent to herbicide treatment areas.

Staff of the Department of Pesticide Regulation (DPR) monitored herbicide residues of glyphosate, triclopyr, and hexazinone in various plant species, determined herbicide dissipation rates in those species, and estimated the potential for herbicide residues to move off-site following application. This report summarizes data collected over a four-year period (1997-2001) from three California National Forests.

MATERIAL and METHODS

US Forest Service Herbicide Program Treatments

The US Forest Service applied four herbicide products: glyphosate applied as Accord®, triclopyr applied as Garlon® 4, and hexazinone applied as Velpar® L and as Pronone® 10G. Herbicide applications occurred during 1997-2000 in the Eldorado, Sierra, and Stanislaus National Forest (Figure 1). Applications were made by commercial pesticide applicators under contract with the US Forest Service.

Figure 1. National Forest land herbicide treatment monitoring sites

With the exception of aerially applied granular hexazinone (Pronone® 10G), all other herbicide applications were made by ground crews (Table 1). Some applications of granular hexazinone were also applied by ground crews, but were not monitored in this study due to their limited use.

Table 1. Forestry herbicide active ingredient, formulation, application method, and location monitored

Herbicide Active Ingredient	Product Name	Formulation	Application Method	National Forest
Glyphosate	Accord®	Liquid	Ground	Eldorado, Sierra, and Stanislaus
Triclopyr	Garlon® 4	Liquid	Ground	Eldorado and Stanislaus
Hexazinone	Velpar® L	Liquid	Ground	Sierra and Stanislaus
Hexazinone	Pronone® 10G	Granular	Air	Stanislaus

Glyphosate (Accord®) and triclopyr (Garlon® 4) were directly applied to foliage with hand pressurized backpack sprayers to treat individual or clusters of unwanted plants. Tank mixes contained a color dye marker to stain vegetation indicating to spray crews which plants were treated. Also included were a surfactant, water as the carrier, and either glyphosate or triclopyr singly or in combination. By reducing surface tension, surfactants assist in spray solution movement into plant tissue (Greene and Bukovac, 1974). Accord® and Garlon® 4 both contain 4 pounds of active ingredient per gallon of product and were applied at the rates of 0.4 to 4.8 and 0.3 to 1.8 pounds active ingredient per treated acre, respectively. Application rates for all herbicides were obtained from US Forest Service staff.

Liquid hexazinone (Velpar® L) was used either undiluted or diluted with water, and like glyphosate and triclopyr, it was also mixed with a color dye marker. Concentrated hexazinone with no dilution was basally applied to the soil underneath the plant pest by spot gun application. By directing the gun to the soil, each draw of the gun's trigger delivered a pre-determined volume of herbicide material. The volume of material applied was dependent upon plant height. Following a rain event, the basally applied hexazinone is released into the soil and available for uptake by the plant root system.

In contrast, diluted hexazinone was applied by hand pressurized backpack sprayer to plant foliage so that foliar absorption would occur. Velpar® L contains 2 pounds of hexazinone per gallon of product and was applied at a rate of 1.5 to 3 pounds active ingredient per treated acre for diluted and undiluted hexazinone.

Pronone® 10G is composed of irregularly shaped granules coated with 10% hexazinone by weight (Feng *et al.*, 1988). To reduce dust formation, the granules contain an additional exterior coating devoid of the active ingredient. Pronone® 10G granules were dispersed by helicopter using a hopper attachment. This method of application facilitated dispersal over large acreages and steep terrain at an application rate of 3 pounds active ingredient per treated acre.

DPR Dissipation Study

This dissipation study was conducted to address the potential exposure of California Indian plant gatherers and basketmakers to herbicide residues in plant materials collected in treated areas.

Following consultation with California Indians, four plant species of interest were selected for herbicide monitoring in the Eldorado, Sierra, and Stanislaus National Forests (Table 2). These readily available plants in the treatment areas represent one root-type plant (bracken fern rhizomes), one brush-type plant (buckbrush shoots), one foliage-type plant (golden fleece foliage), and one food-type plant (manzanita berries).

Table 2. Four plants and their parts used by California Indians

Common Name	Scientific Name	Sampled Plant Part	California Indian Use
Bracken Fern	*Pteridium aquilinum var. pubescens*	Roots	Basketry
Buckbrush	*Ceanothus cuneatus*	Shoots	Basketry
Golden Fleece	*Ericameria aborescens*	Foliage	Medicine
Manzanita	*Arctostaphylos spp.*	Berries	Food

The four plant parts were monitored for the four forestry herbicide products applied: glyphosate, triclopyr, and liquid hexazinone by ground and granular hexazinone by air (Table 1). Herbicide applications in selected monitoring sites occurred during 1997 to 2000.

Sixty-four monitoring sites (four sampling sites or replications for each plant part and herbicide product combination) were selected in the three National Forests as described in the study protocol (DPR, 1997). Sites were accessible, had the appropriate plants growing in the herbicide treatment area, and had an adequate supply of plant materials available for a minimum of seven sampling periods for chemical analyses. Selected manzanita berry sites had flowers or berries present at the time of herbicide application. Site locations are presented in Appendix A.

Anywhere from six to 20 buckbrush, golden fleece or manzanita plants at each monitoring site were tagged with flagging tape to identify them for sampling. Bracken fern was generally pervasive in an area, so a 10 to 20 ft diameter circle was marked off with flagging stakes so that root samples would be collected within this enclosed region. Plant samples were collected at seven intervals: within 1-3 days following herbicide application (application-day sample), and within 3-5, 7-9, 11-13, 19-21, 27-29, and 35-37 weeks following herbicide application.

Sixteen sites (one sampling site for each plant-herbicide combination) were monitored with 11 sites extended beyond the 35-37 week study period due to continued herbicide detections at several sites originally treated with herbicide in 1997 and 1998. Monitoring was later discontinued at these high residue sites due to lack of further herbicide detections, inaccessible roads due to snow or heavy rains or inadequate supply of plant material for further chemical analysis.

Table 3. Occurrence of extended monitoring

Chemical	Number of additional sampling periods beyond 35-37 week post-application period for 3 plant media		
	Bracken Fern Root	Buckbrush Shoot	Golden Fleece Foliage
Glyphosate	2	1	1
Triclopyr	1	2	1
Hexazinone – Velpar® L	-	3	2
Hexazinone – Pronone® 10G	2	3	2

Additional samples were collected only at the monitoring site that had the highest residue concentration for each plant-herbicide combination reported at 35-37 week sample period.
Additional manzanita berry samples were not collected beyond the 35-37 week sample period due to insufficient berry supply..

DPR Off-Site Movement Study

The second portion of this study addressed the potential for off-site movement of the four herbicide products from the application area due to drift at the time of application or due to other influences following application. This information would help in determining an acceptable distance to gather plant materials from treated areas.

From 1997 to 1999 DPR staff monitored native plant parts offsite in the three National Forests for the four forestry herbicide products applied: glyphosate, triclopyr, and liquid hexazinone by ground and granular hexazinone by air. Twenty-four off-site monitoring locations (6 sites per herbicide product) were selected for the study (DPR, 1997). Off-site herbicide residues were monitored in bracken fern roots, buckbrush shoots, and deerbrush shoots (*Ceanothus integerrimus*).

Off-site monitoring locations were selected prior to herbicide application. Monitoring sites were adjacent to the outside treatment area, easily accessible, had the targeted plants, and had an adequate supply of plant material available for chemical analyses for three sampling periods during the 12-week monitoring period. Off-site monitoring plants up to 100 ft down slope from the treatment area were marked at four distance ranges (5-15, 20-40, 50-70, and 80-100 ft) using flagging tape.

Plant samples were collected at each distance to determine herbicide movement from the edge of the application area. Samples were collected at four periods: prior to herbicide application (background sample), 1-3 days post-application (application day sample), and 3-5 and 11-13 weeks post-herbicide application. Background samples were randomly collected from the entire transect distance of 5-100 ft to determine if residues were present prior to treatment. Background plant samples were collected in 1998 and 1999, but not in 1997.

Dissipation and Off-Site Sampling Methods and Equipment Cleaning Procedures

Whenever possible, roots, shoots, foliage, and berries samples were collected from tagged plants with green vegetation, showing no signs of herbicide effects (e.g. brown discolored vegetation). If green vegetation was not available for sampling, plants were assumed to be dead and random plant samples was collected (over all or several of these tagged plants).

Clean disposable latex gloves were worn to collect approximately 0.2 pounds of plant material that were placed into 1-quart glass sample jars. Gloves were changed with each sample collected to prevent herbicide cross-contamination.

Clean hand-held pruning shears were used to cut buckbrush shoots, golden fleece foliage, and manzanita berries directly into the sample jar. Bracken fern roots and deerbrush shoots required additional handling. A clean shovel or hand trowel was used to expose bracken fern roots that were shaken to remove soil before being placed in the sample jar. Leaves from deerbrush shoots were removed with hand-held pruning shears and discarded in the field and then the barren shoots were placed into the sample jar. All sample jar openings were lined with an aluminum foil sheet and were then tightly sealed with a lid. Samples were placed on dry ice after collection and remained frozen until chemically extracted in the laboratory.

Sampling equipment was cleaned in the field using a deep plastic container lined with a disposable plastic bag used to hold the cleaning solution of de-ionized water and Liqui-Nox® soap. Disposable cleaning sponges and scouring pads were used to vigorously clean the sampling equipment to remove any herbicide or plant residue. Once washed, equipment was rinsed with de-ionized water, then alcohol, and then allowed to air dry before being placed in a clean plastic storage bag.

Chemical Analyses

The analyzing laboratory was the California Department of Food and Agriculture's Center for Analytical Chemistry in Sacramento, California. Laboratory personnel developed chemical analytical methods to determine residues of glyphosate, triclopyr, and hexazinone in several plant media (Appendix B). Appendix B also contains method validation results (percent herbicide recovery) for the three active ingredients. The minimum detection limit (MDL) for each plant-active ingredient combination is presented in Table 4. Levels ranged from 0.03 to 0.1 ppm and are the lowest amounts of a chemical that are detectable in each plant part using the analytical method.

Control charts were developed from method validation studies for ongoing quality control purposes. Lower and upper warning limits (mean $\pm$ 2 standard deviations) and control limits (mean $\pm$ 3 standard deviations) were created to determine if results were within an acceptable range. Field samples were extracted along with plant parts fortified with known herbicide addition(s) and also with unfortified plant parts containing no herbicide residue. If recoveries were outside the warning or control limits for the fortified samples or if residue was detected in unfortified samples, chemical analyses were redone. Ongoing quality control data and storage stability data are presented in Appendix B.

Table 4. Minimum detection limit (ppm) for three forestry herbicide active ingredients in various plant media

Chemical Active Ingredient	Plant Media	Minimum Detection Limit
Glyphosate (Accord®)	Bracken Fern Roots	0.1
	Buckbrush Shoots	0.1
	Golden Fleece Foliage	0.1
	Manzanita Berries	0.1
Triclopyr (Garlon® 4)	Bracken Fern Roots	0.03
	Buckbrush Shoots	0.05
	Golden Fleece Foliage	0.07
	Manzanita Berries	0.03
Hexazinone (Velpar® L/Pronone® 10G)	Bracken Fern Roots	0.05
	Buckbrush Shoots	0.1
	Golden Fleece Foliage	0.1
	Manzanita Berries	0.05

Statistical Analyses

The mean, standard deviation, and standard error of the mean were calculated for each plant part/herbicide product combination. When herbicides were not detectable in a medium, one-half the MDL was used in the calculations. Half-lives were determined using an exponential decay equation. Further statistical information and field data are provided in Appendix C.

RESULTS and DISCUSSION

Glyphosate, triclopyr, and hexazinone data are presented on a fresh weight basis. When herbicide residue levels were below the MDL in a medium, one-half the value of the MDL was used in statistical analyses. Also, data collected beyond the 35-37 week sample period for each plant part were taken from the site with the highest residue level for each plant-herbicide product combination and, therefore, may not be representative of a population.

Quality Control

Ongoing quality control information was obtained for the three forestry herbicide active ingredients in bracken fern roots, buckbrush shoots, golden fleece foliage, and manzanita berries. Average recoveries ranged from 69 to 95% (Table 5) for all plant-active ingredient combinations. Of the 634 dissipation and off-site monitoring samples collected, 48 individual samples (7%) were outside the set warning limits established for each plant-active ingredient combination. The quality control data fell outside the established warning limits more often for bracken fern roots than for the other three plant media with regard to all three active ingredients. Occurrences were below and above the lower and upper warning limits, respectfully, about an equal number of times with the majority of these occurrences noted during 1997 analyses. The data presented in this study were not adjusted based upon the laboratory quality control results.

Table 5. Average analytical method recoveries for three forestry herbicide active ingredients

Plant Media	Percent Recovery		
	Glyphosate	Triclopyr	Hexazinone
Bracken Fern Roots	79.1	75.2	90.5
Buckbrush Shoots	93.6	85.0	90.2
Deerbrush Shoots	94.8	87.8	89.2
Goldenfleece Foliage	93.9	68.8	92.9
Manzanita Berries	94.3	92.8	89.5

General

Monitoring sites were located inside or adjacent to treatment areas that ranged from 2 to 270 acres in size and were at elevations ranging from approximately 2,500 to 6,000 ft. Other differences between sites included month of herbicide application, climatic conditions, and stage of plant maturity.

Dissipation Monitoring

Dissipation data was collected from 53 monitoring sites located in the three National Forests. Overall, the four herbicide products showed a general decline in pesticide concentration in treated plant material during the course of the study (Table 6).

Glyphosate mean application-day residue levels ranged from 0.5 to 241 ppm for bracken fern roots, buckbrush shoots, golden fleece foliage, and manzanita berries sampled 1 to 3 days following application. Glyphosate levels were approximately 2 to 900 times greater than mean application-day residue levels for the three remaining herbicide products on the same media. High levels are attributed to direct deposition of glyphosate on plant materials using a generally higher application rate than used for triclopyr or liquid hexazinone, the other two foliar applied herbicides (Figures 2 and 3).

Foliar applied glyphosate was quickly translocated to roots of bracken fern plants where it was detected at a mean concentration of 0.5 ppm 1 to 3 days following treatment. At week 67 post-application, the last sampling date for bracken fern roots, glyphosate residue remained detectable at 0.4 ppm.

Mean glyphosate residue levels in buckbrush shoots, golden fleece foliage, and manzanita berries remained above 75 ppm during the first 20 weeks following herbicide application. By week 60, residues in buckbrush shoots declined to less than 1 ppm and residues in golden fleece foliage were undetectable. Mean glyphosate residue levels in manzanita berries were 31 ppm at week 36, the last sampling date that berries were available for sampling.

Table 6. Mean herbicide residue levels reported in sampled plant media.

Chemical	Weeks After Treatment	Mean Residue Level Detected in Sampled Plant Part (ppm)			
		Bracken Fern Roots	Buckbrush Shoots	Golden Fleece Foliage	Manzanita Berries
Glyphosate	0	0.5	241	173	74.4
	4	1.4	230	265	123
	8	0.3	165	157	150
	12	0.3	164	77	136
	20	0.2	100	203	150
	28	0.2	16.0	24.8	80.3
	33	0.4 (1)*	-	-	-
	36	ND (<0.05)	1.8	7.4	30.8 (1)
	41	0.7 (1)		-	-
	60	-	0.3 (1)	ND (<0.05)(1)	-
	67	0.4 (1)	-	-	-
Triclopyr	0	0.1	18.8	5.9	3.1
	4	0.1	13.6	1.7	1.9
	8	0.06	5.0	0.8	2.8
	12	0.07	14.0	0.5	1.9
	20	0.1	3.1	1.0	3.0
	28	ND (<0.015)	1.2	0.6	3.0
	32	ND (<0.015) (1)	-	-	
	36	ND (<0.015)	0.5	0.6	2.6 (1)
	41	ND (<0.015) (1)	-	-	-
	60	-	0.7 (1)	ND (<0.035) (1)	-
	80	-	0.6 (1)	-	-
Hexazinone Velpar® L	0	0.2	26.0	81.2	-
	4	0.3	2.1	11.6	-
	8	0.1	0.9	2.4	0.08
	12	0.2	0.8	3.1	0.2
	20	0.08	1.7	5.3	0.05
	28	0.09	0.1	ND (<0.05) (1)	ND (<0.025)
	36	0.07	0.2	0.7	0.1
	60	-	-	0.2 (1)	-
	62	-	0.2 (1)	-	-
	80	-	0.4 (1)	ND (<0.05) (1)	-
	130	-	ND (<0.05) (1)	-	-
Hexazinone Pronone® 10G	0	ND (<0.025)	0.3	0.2	-
	4	0.2	ND (<0.05)	0.2	-
	8	0.3	0.9	0.6	0.06
	12	0.2	0.6	0.5	ND (<0.025)
	20	0.2	0.5	0.09	ND (<0.025)
	28	0.2	0.3	ND (<0.05)	ND (<0.025)
	36	0.09	0.4	0.4	ND (<0.025)
	60	0.06 (1)	0.2 (1)	-	-
	62	-	-	0.2 (1)	-
	80	ND (<0.025) (1)	0.4 (1)	ND (<0.05) (1)	-
	130	-	ND (<0.05) (1)	-	-

ND is not detected. Value shown is one-half the MDL.
*(1) indicates that only one sample was collected.
- indicates that no sample was collected.

Triclopyr residues were detected in all four application-day media sampled with mean levels ranging from 0.1 to 19 ppm. As with glyphosate, triclopyr was detected in bracken fern root samples collected on application day at a mean concentration of 0.1 ppm, indicating that this systemic compound was quickly translocated from treated foliage to the underground root system. Bovey *et al.* (1983) also reported that triclopyr was rapidly translocated to the root system of the honey mesquite plant (*Prosopis juliflora* var. *glandulosa*) within 4 hours following triclopyr application to a single leaf. Residues were not detectable in bracken fern roots at 28-41 weeks after treatment.

Triclopyr residues in buckbrush shoots remained detectable at a concentration of 0.6 ppm at week 80 but were not detectable in goldenfleece foliage sampled at week 60, the last sampling dates for both media. No other samples were collected between weeks 36-60 for golden fleece foliage. In manzanita berries, triclopyr was detected at a mean residue level of 2.6 ppm at week 36, the last sampling date for that medium.

Mean application-day hexazinone residue ranges for liquid and granular hexazinone applications, respectively, were 0.2 to 81 ppm and not-detected to 0.3 ppm in sampled bracken fern roots, buckbrush shoots, and golden fleece foliage. Manzanita berries were not available for sampling on application day because they were unformed.

Buckbrush shoots and golden fleece foliage sampled 1 to 3 days following treatment with granular hexazinone showed the presence of residues in sampled plant parts. It is possible that application-day detections may have resulted from hexazinone dust residue settling on above ground sampled plant parts. Also, it is possible that granular material was washed into the soil and actual uptake occurred following aerial application, although hexazinone application-day detections were not reported in bracken fern roots. Feng *et al.* (1988) reported that any amount of moisture from rainfall, dew or condensation was enough to result in significant release of hexazinone from coated granules making it available for absorbtion by the plant root system (Ghassemi *et al.*, 1989).

Granular-applied maximum mean hexazinone residue levels in sampled roots, shoots, and foliage were observed at week 8 post-application, when they were approximately three or four times greater than application-day residue levels. It is suspected that time was needed for the granules to be washed into the soil and taken up by the roots and translocated to the sampled above ground plant parts. By week 28, however, granular and liquid hexazinone treated plants showed similar residue levels in sampled plant parts with residues ranging from not-detected to 0.3 ppm in roots, shoots, and foliage. By weeks 80 and 130 post application, hexazinone residues were no longer detectable in golden fleece foliage and buckbrush shoots from either granular or liquid hexazinone applications. In bracken fern roots, residues remained detectable at a mean concentration of 0.07 ppm at week 36, the last sampling date for liquid hexazinone in root medium. Residues in bracken fern roots were not detected at week 80, the last sampling date for granular hexazinone in root media.

When manzanita berries were available for sampling starting at week 8 post-application, hexazinone residues were detected in berries in only 1 of 5 sampling periods for granular applied material while in comparison, detections were reported in 4 of 5 sampling periods for liquid applied hexazinone. Low granular hexazinone detections may indicate low uptake, low translocation, or rapid degradation with the plant (Sidhu and Feng, 1993) compared to liquid hexazinone treated plants. Sidhu and Feng (1993) reported that hazelnut (*Corylus cornuta* Marsh.) was less sensitive to hexazinone and absorbed and translocated less hexazinone to the foliage than other plant species tested. Baron and Monaco (1986) also reported that plant cuttings of rabbiteye blueberry (*Vaccinium Ashei* Reade) and highbush blueberry (*Vaccinium corymbosum* L.) absorbed less hexazinone from hexazinone root applications than did hollow goldenrod (*Solidago fistulosa* Miller) plant cuttings, resulting in less hexazinone residue translocated to the leaves of both species of blueberry plants. Baron and Monaco (1986) suggested that the low translocation of hexazinone from the root to the shoot may be due to the restricted translocation from the site of uptake (roots) to the site of action (chloroplasts).

Also examined at each monitoring location was the dissipation time in weeks observed for an herbicide to reach the non-detectable residue level, starting from the herbicide's maximum concentration in a plant part (Table 7). In general, large time variations existed between the number of weeks to reach the non-detectable level for each plant-herbicide product combination with elapsed time, ranging from 4 to 130 weeks. Herbicide residues appeared persistent in

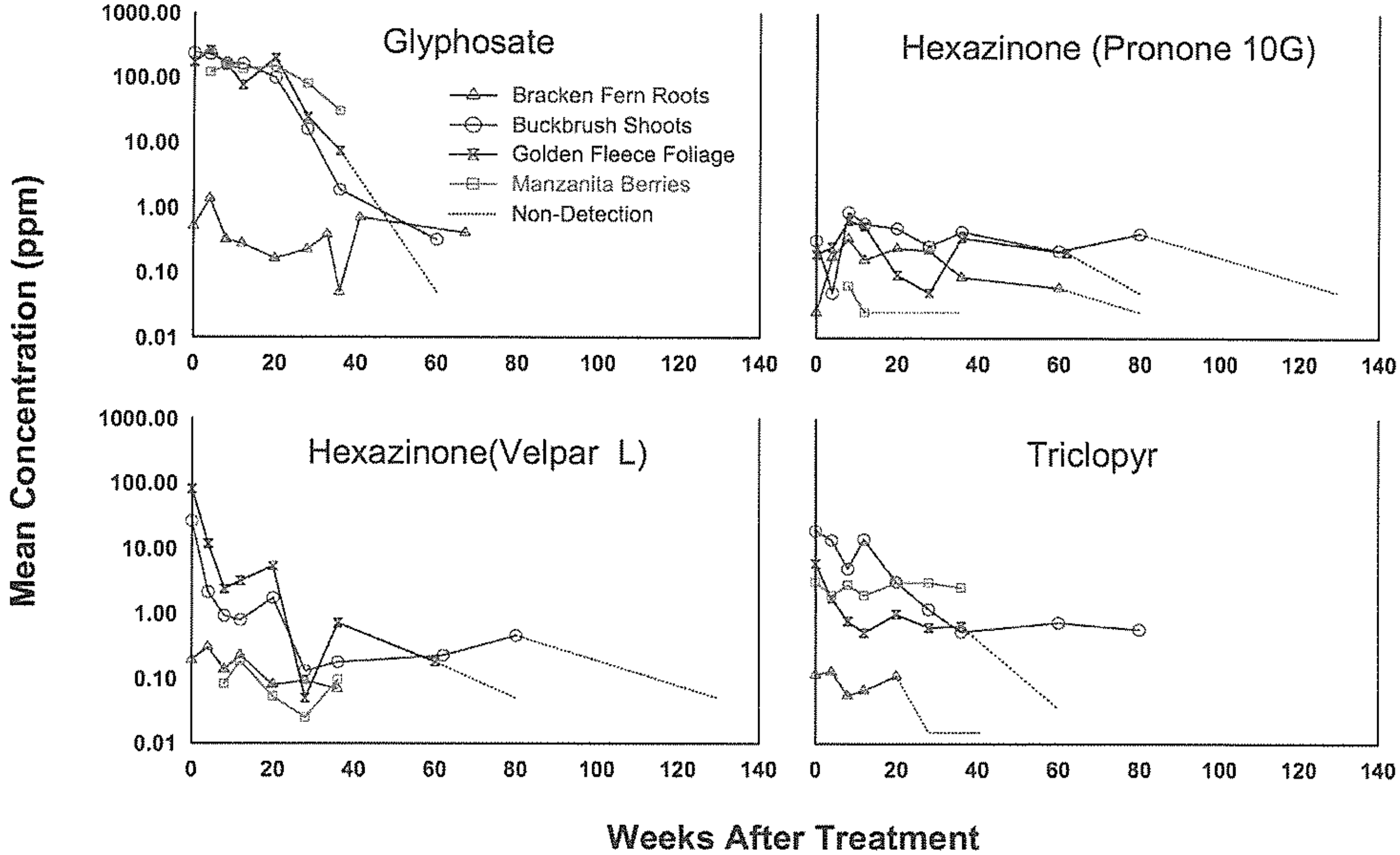

Figure 2. Dissipation of herbicide in plants, Eldorado, Sierra and Stanislaus National Forests, Calif., 1997-2001.

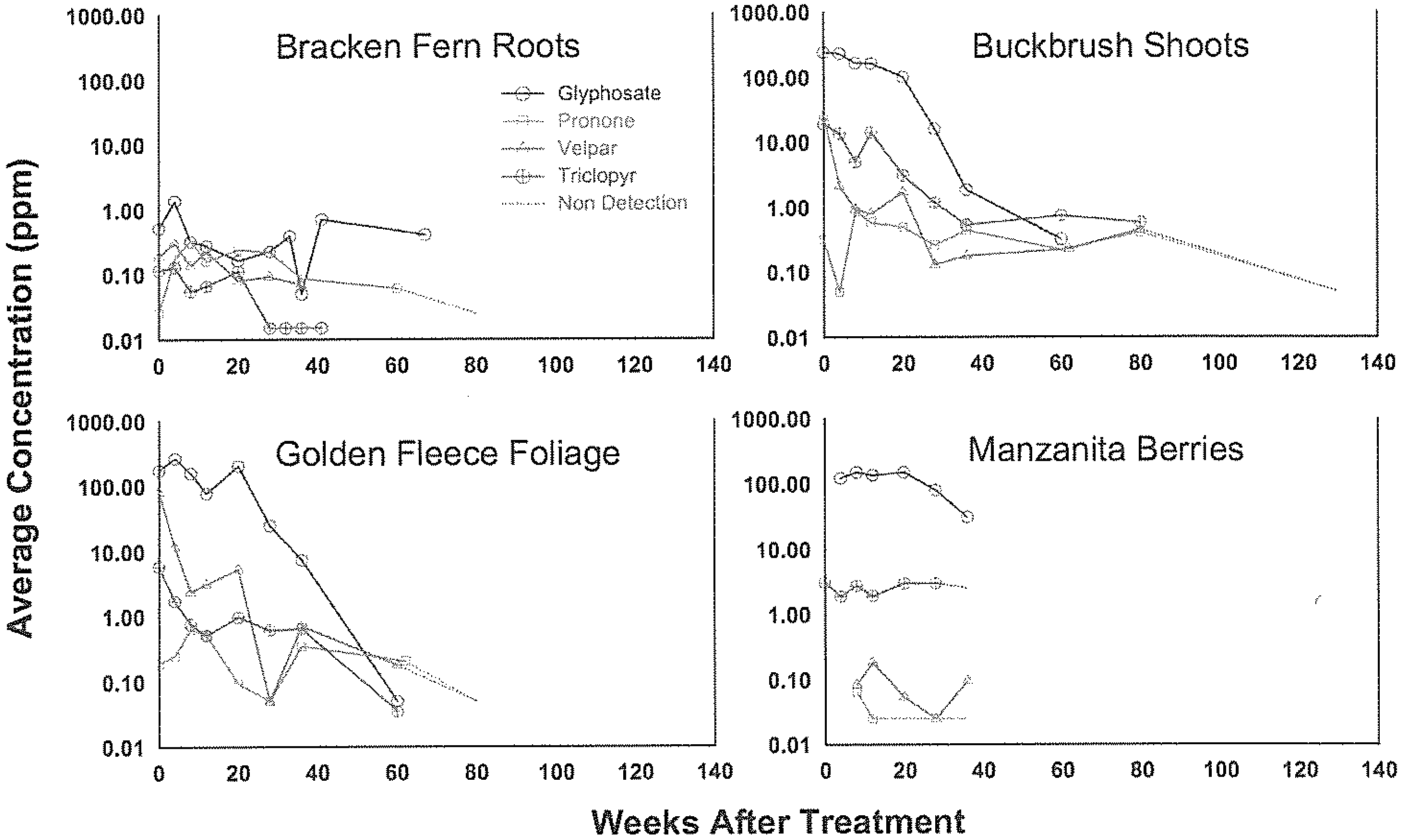

Figure 3. Plant materials on the dissipation of herbicides from Eldorado, Lassen, Sierra and Stanislaus in California, 1997-2001

buckbrush shoots treated with liquid hexazinone, as residues were detectable in shoots sampled at week 80, but residues were not detectable the following sampling period at 130 weeks post-application. This was the longest observed dissipation period for an herbicide residue level to reach the non-detectable level commencing from the herbicide's maximum concentration in a plant medium. Hexazinone dissipated from granular treated bracken fern roots within an average of 29 weeks after the maximum residue level was reached while it dissipated from liquid treated bracken fern roots within an average of 4 weeks after reaching the maximum residue level.

Table 7. The number of weeks observed from the maximum herbicide concentration to the non-detectable level

Chemical	Site	Plant Part Sampled: Bracken Fern Roots	Buckbrush Shoots	Golden Fleece Foliage	Manzanita Berries
Glyphosate	1	8	na*	na	na
	2	na	na	60	na
	3	na	na	na	na
	4	4	na	24	na
	Average	6	na	42	na
Triclopyr	1	8	na	na	na
	2	24	na	na	na
	3	8	na	56	-
	4	4	-**	-	-
	Average	11	na	56	na
Hexazinone Velpar® L	1	4	na	20	8
	2	na	na	20	4
	3	-	130	na	-
	4	-	-	-	-
	Average	4	130	20	6
Hexazinone Pronone® 10G	1	4	4	12	8
	2	na	4	16	8
	3	24	4	20	8
	4	60	4	12	-
	Average	29	4	15	8

*na denotes herbicide residue level was at or above the minimum detection limit.
**A dashed line indicates that a monitoring site was not selected.

Golden fleece plants treated with liquid hexazinone showed similar dissipation rates in foliage to those plants treated with granular hexazinone. Likewise, manzanita plants treated with liquid hexazinone showed similar dissipation rates to those treated with granular hexazinone. Additional data was not available to determine dissipation rates at about 40% of the monitoring locations because residue levels remained detectable at the last sampling period or as with manzanita berries, the fruit supply was depleted at week 36 post-application.

The means and standard errors for each plant-herbicide product combination are presented in Figures 4 to 7. Bracken fern roots were the only medium observed to have small standard error at each sampling period for all four herbicide products, with residue data showing low variability about the sample mean. Greater variability (large standard error) was observed with buckbrush shoots, golden fleece foliage, and manzanita berry residue data, making herbicide residue level inferences about a population less certain. Large variability was also observed in many instances where only two samples were collected on a sampling date. In situations where only one sample was collected, a single point is shown in the figures.

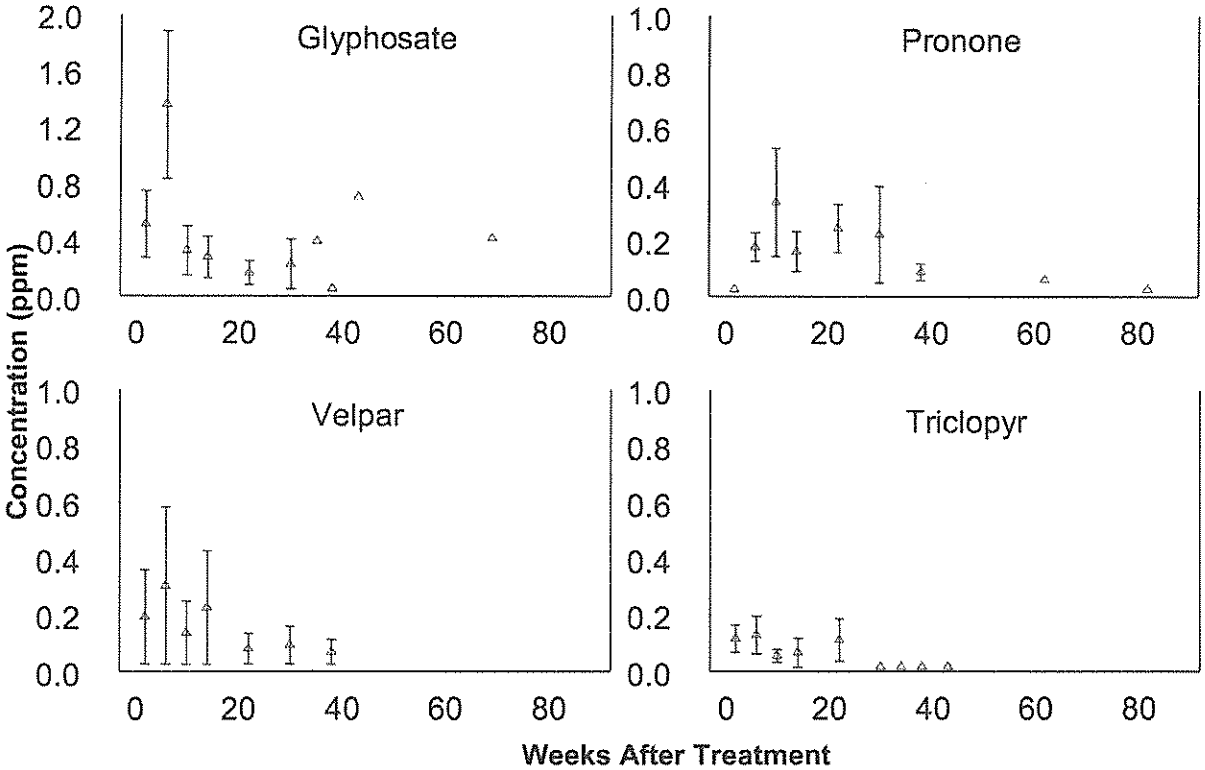

Figure 4. Herbicide residue distribution of sample means (mean ± standard error) for four forestry herbicide products in bracken fern roots

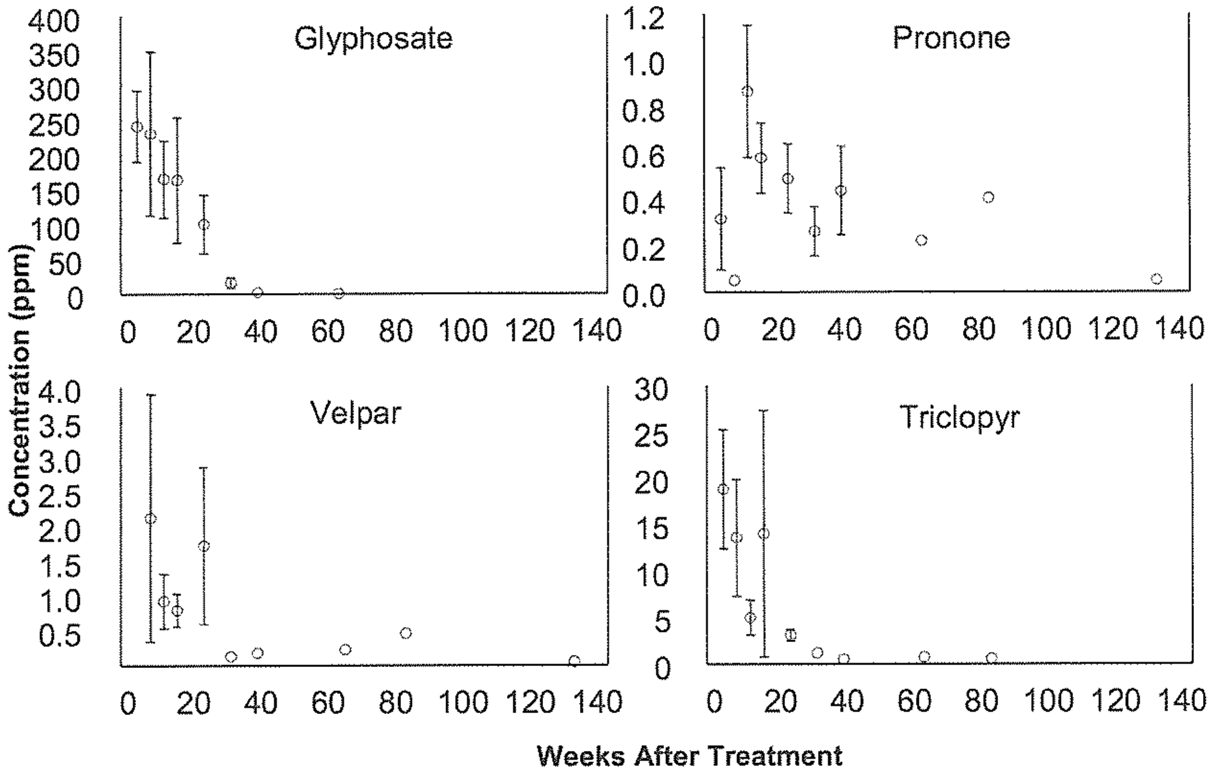

Figure 5. Herbicide residue distribution of sample means (mean ± standard error) for four forestry herbicide products in buckbrush shoots

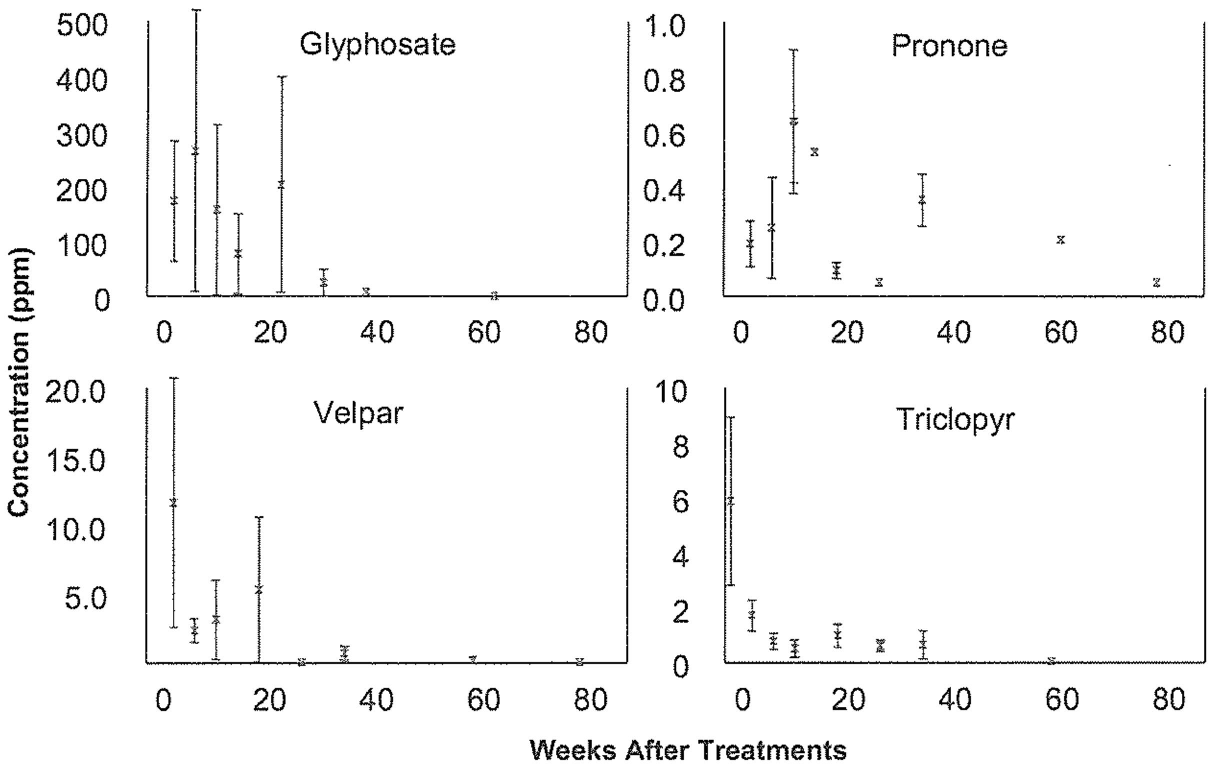

Figure 6. Herbicide residue distribution of sample means (mean ± standard error) for four forestry herbicide products in goldenfleece foliage

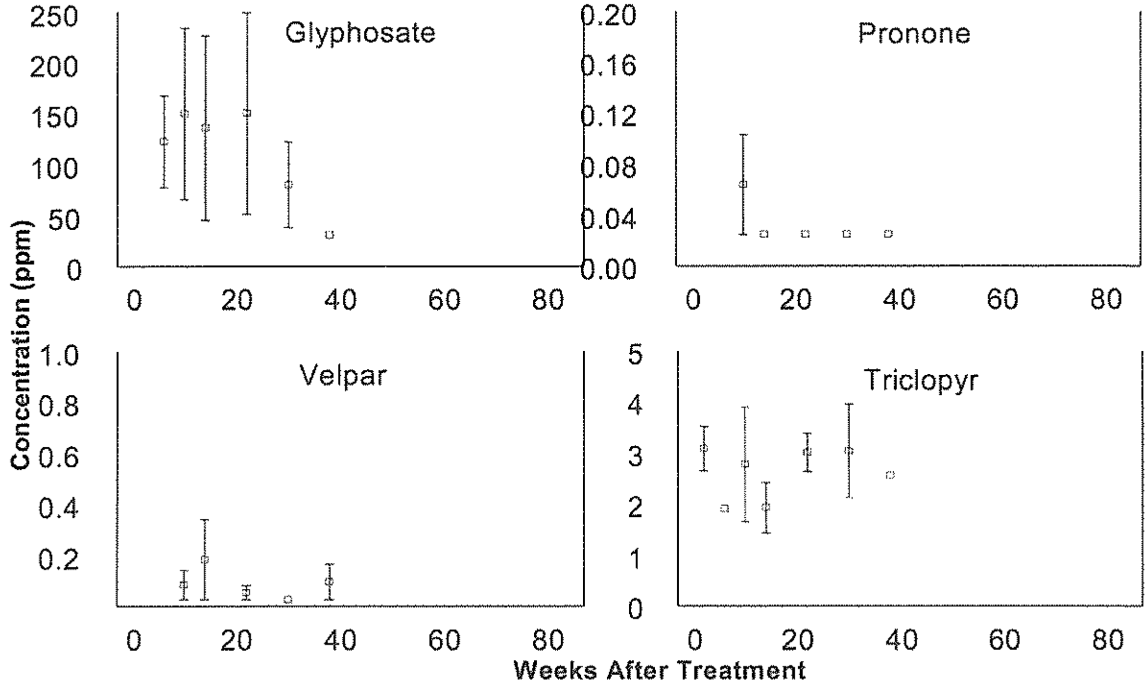

Figure 7. Herbicide residue distribution of sample means (mean ± standard error) for four forestry herbicide products in manzanita berries

Chemical half-life values in plant parts should be used with reservation due to the limited number of site specific data sets contributing to the half-life calculations at the 90% significance levels. Overall, average half-lives ranged from 1 to 19 weeks (Table 8) for the various plant part-herbicide product combinations. Half-lives were greater for bracken fern roots than they were for plant parts sampled above ground level. Half-lives were longest for liquid hexazinone treated plants (1 to 19 weeks), followed by glyphosate (8 to 12 weeks), and then triclopyr (2 to 6 weeks). Liquid hexazinone half-lives obtained in this study were longer than those observed by Michael (1990) who reported Velpar® L half-lives of less than 9 weeks in terminal shoots of dogwood (*cornus florida* L.), blueberry (*Vaccinium* sps.), and bracken fern (*pteridium aquilinum* L.).

Table 8. Average half-life of four forestry herbicides in plant parts used by California Indians

	Average Half-Life for Plant Media Sampled (weeks)			
Herbicide	Bracken Fern Roots	Buckbrush Shoots	Golden Fleece Foliage	Manzanita Berries
Glyphosate	11.5 (1)*	9.8 (3)	8.2 (2)	na**
Triclopyr	6.1 (2)	2.4 (3)	5.1 (3)	na
Hexazinone-Velpar® L	18.5 (1)	17.6 (2)	0.6 (2)	na
Hexazinone-Pronone® 10G	na	na	na	1.7 (1)

*The number in parentheses indicates the sample size used for the calculation of the mean.
**na denotes that no meaningful regression could be obtained and, therefore, no average half-life was calculated.

Off-Site Monitoring

Off-site monitoring was conducted at 20 locations. Herbicide residue levels in bracken fern roots, buckbrush shoots, and deerbrush shoots ranged from not detected to 2.7 ppm for the four herbicide products (Tables 9-12).

Glyphosate residues ranging in concentration from 0.1 to 2.7 ppm were detected in plant media sampled at three monitoring locations 1 to 3 days following application. Detections were at the 5-15 ft transect distance. At week 4 post-application, residue levels at two of the three sites remained detectable at 0.1 ppm and were undetectable at the third location. By week12, however, residues were not detectable at either of the two sites at the 5-15 ft distance. Contamination of sampling equipment may have been the cause of a glyphosate detection at the

20-40 ft distance at one of these sites at week 12 post-application. Glyphosate contamination may also have occurred resulting in the week 12 detection at the 20-40 ft distance at another unrelated monitoring location.

Triclopyr residues were also found off-site at two locations. Residues were detected up to 100 ft from the treatment area in sampled application-day media. The highest residue level of 1.6 ppm was observed at the 5-15 ft distance from the spray area with residue levels declining further away from treatment site. Residues were not detected at either of these two sites at weeks 4 or 12 post-application.

Monitoring for glyphosate was also conducted at these two locations (Stanislaus/Mi-Wok and Eldorado/Placerville) where triclopyr was detected, as glyphosate was co-applied in the tank mixes. Triclopyr residues were observed to have moved further off-site compared with glyphosate residues. However, it is suspected that both chemicals drift equivalent distances, but glyphosate remained undetected due to the chemical's higher MDL of 0.1 ppm in comparison to 0.03 ppm for triclopyr.

Liquid hexazinone was also detected off-site following application. At one monitoring location, hexazinone was detected at 0.7 ppm 12 weeks post-application and it is thought that rainfall and/or snowmelt was responsible for the off-site herbicide movement. The transect at this monitoring location was parallel rather than perpendicular to the treatment site. DPR staff observed prominent gullies leading from the treatment area to a section of the sampling transect at week 12 post-application; the washed out gullies were not present prior to week 12 sampling and also there were no prior hexazinone detections reported at this site. As hexazinone has a high potential to move off-site due to its high water solubility and low adsorption to soil (Worthing, 1979), it is thought that herbicide residues were transported with water to the 50-70 ft segment of the transect.

The application-day hexazinone detection at the 80-100 ft distance is thought to have been a result of sampling equipment contamination as the residue level was low at the MDL, and this detection could not be explained due to physical features in the surrounding sampling area.

Table 9. Off-site monitoring for glyphosate residues

National Forest/District	Plant Sampled (Application Year)	Weeks after Application	Concentration (ppm) Distance from Edge of Treatment Area			
			5-15 ft	20-40 ft	50-70 ft	80-100 ft
Sierra/Pineridge	Buckbrush Shoots (1997)	0	0.1	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	0.1	ND	ND
Eldorado/Pacific	Deerbrush Shoots (1997)	0	ND	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	0.1-1	ND	ND
Eldorado/Placerville	Deerbrush Shoots (1998)	0	2.7	ND	ND	ND
		4	0.1	ND	ND	ND
		12	ND	ND	ND	ND
Stanislaus/Mi-Wok	Deerbrush Shoots (1998)	0	0.2	ND	ND	ND
		4	0.1	ND	ND	ND
		12	ND	ND	ND	ND
Sierra/Kings River	Buckbrush Shoots (1999)	0	ND	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND
Stanislaus/Groveland	Buckbrush Shoots (1999)	0	ND	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND

ND is not detected.
Glyphosate minimum detection limit is 0.1 ppm for buckbrush and deerbrush shoots.

Table 10. Off-site monitoring for triclopyr residues

National Forest/District	Plant Sampled (Application Year)	Weeks after Application	Concentration (ppm) Distance from Edge of Treatment Area			
			5-15 ft	20-40 ft	50-70 ft	80-100 ft
Eldorado/Pacific	Deerbrush Shoots (1997)	0	ND	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND
Eldorado/Placerville	Deerbrush Shoots (1998)	0	1.6	0.07	0.06	0.03
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND
Stanislaus/Mi-Wok	Deerbrush Shoots (1998)	0	0.03-0.3	0.03-0.3	0.03-0.3	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND

ND is not detected.
Triclopyr minimum detection limit is 0.03 ppm for deerbrush shoots.

Table 11. Off-site monitoring for liquid hexazinone (Velpar® L) residues

National Forest/District	Plant Sampled (Application Year)	Weeks after Application	Concentration (ppm) Distance from Edge of Treatment Area			
			5-15 ft	20-40 ft	50-70 ft	80-100 ft
Stanislaus/Groveland	Deerbrush Shoots (1997)	0	ND	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND
Stanislaus/Mi-Wok	Buckbrush Shoots (1997)	0	ND	ND	ND	0.1
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND
Sierra/Pineridge	Buckbrush Shoots (1998)	0	ND	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	ND	0.7	ND
Stanislaus/Groveland	Deerbrush Shoots (1998)	0	ND	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND
Stanislaus/Groveland	Deerbrush Shoots (1998)	0	ND	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND

ND is not detected.
Hexazinone minimum detection limit is 0.1 ppm for buckbrush shoots and deerbrush shoots.

Table12. Off-site monitoring for granular hexazinone (Pronone® 10G) residues

National Forest/District	Plant Sampled (Application Year)	Weeks after Application	Concentration (ppm) Distance from Edge of Treatment Area			
			5-15 ft	20-40 ft	50-70 ft	80-100 ft
Stanislaus/Groveland	Bracken Fern Roots (1997)	0	ND	ND	ND	ND
		4	ND	ND	ND	Sample Lost
		12	ND	ND	ND	ND
Stanislaus/Mi-Wok	Bracken Fern Roots (1997)	0	ND	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND
Stanislaus/Mi-Wok	Deerbrush Shoots (1997)	0	0.1-1	ND	0.1-1	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND
Stanislaus/Mi-Wok	Deerbrush Shoots (1998)	0	ND	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND
Stanislaus/Mi-Wok	Deerbrush Shoots (1998)	0	ND	ND	ND	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND
Stanislaus/Mi-Wok	Buckbrush Shoots (1999)	0	ND	ND	0.1	ND
		4	ND	ND	ND	ND
		12	ND	ND	ND	ND

ND is not detected.
Hexazinone minimum detection limit is 0.05 ppm for bracken fern roots, and 0.1 ppm for buckbrush shoots and deerbrush shoots.

Hexazinone residues from granular aerial hexazinone applications were also observed off-site. Detections at two monitoring locations were found up to the 50-70 ft distance and are thought to have been the result of dust residue deposition on plant material.

Residues of glyphosate, triclopyr, and hexazinone were detected outside the treatment area immediately following application and up to 12 weeks following application, indicating that drift from sprays or granular material is a possibility with all herbicides and that off-site movement with rainfall/snowmelt may also occur. Of the 240 off-site samples collected, only 19 (7.9%) were positive for herbicide residues and approximately 33% of the detections were at or close to the MDL. Herbicide residues were not detected in any background plant samples collected at the off-site monitoring locations prior to treatment.

Additional Herbicide Residue Monitoring

California Indians also expressed concern about herbicide residue levels in oak acorns used for food and redbud (*Cercis occidentalis*) shoots used for basketry materials. Due to the difficulty in locating redbud plants growing in herbicide treatment areas, Sierra National Forest personnel stationed at the Kings River District basally applied hexazinone in 1997 to the soil underneath redbud shrubs using the spot gun treatment. Shoot samples collected at day 0, and at 4, 8, and 12 weeks following application showed no hexazinone residues in plant materials analyzed. The MDL for redbud is 0.05 ppm.

Each fall, DPR staff surveyed sites treated with herbicides and sampled available acorns that had fallen underneath the oak tree canopy. In 1997, acorns were collected 28 weeks post-application in two areas of the Stanislaus National Forest/Mi-Wok District where a liquid and granular hexazinone application had occurred, respectively. Hexazinone residues were not detected in these acorn samples.

Acorns were also collected at 36 week post-application from a 1998 liquid hexazinone treamtment area in the Sierra Forest/Pineridge District; hexazinone residues were not detected. In 1998, acorn samples from the Stanislaus Forest/Groveland District sampled 36 weeks after glyphosate application contained no glyphosate residues. The MDL for hexazinone and glyphosate in acorn is 0.1 ppm.

SUMMARY

Foliar-applied systemic herbicides resulted in higher application-day residue levels in sampled plant parts in comparison to granular applied material. Highest application-day residue levels were observed with glyphosate, followed by liquid hexazinone, triclopyr, and then granular hexazinone treated plants.

Granular hexazinone requires moisture to be released into the soil so that it could be taken up by the plant root system. This formulation had the lowest herbicide residue levels in all plant materials sampled with maximum concentrations reached at week 8 post-application. However, by week 28 post-application, similar residue levels were observed in roots, shoots, and foliage of plants treated with either granular or liquid hexazinone.

Overall, herbicide residues dissipated most slowly from the maximum residue concentration to the non-detectable level in buckbrush shoots compared with other plant parts sampled. Estimated herbicide half-lives were variable, ranging from 1 to 19 weeks. In decreasing order, half-lives were longest for liquid hexazinone, glyphosate, triclopyr, and then granular hexazinone treated plant materials. Because residue data were variable, a controlled study may be useful to either verify or refine results obtained in this field study.

Glyphosate, triclopyr, and hexazinone were detected off-site following application. Triclopyr residues were detected up to 50-100 ft from the spray area in regions where it was co-applied with glyphosate. It is assumed that glyphosate also traveled distances equivalent to that of triclopyr, but remained undetected due to its higher MDL. Hexazinone is also suspected to have been transported off site in rain runoff/snowmelt from a liquid hexazinone treatment site and also transported off-site in dust residue from a granular hexazinone treatment site during aerial application.

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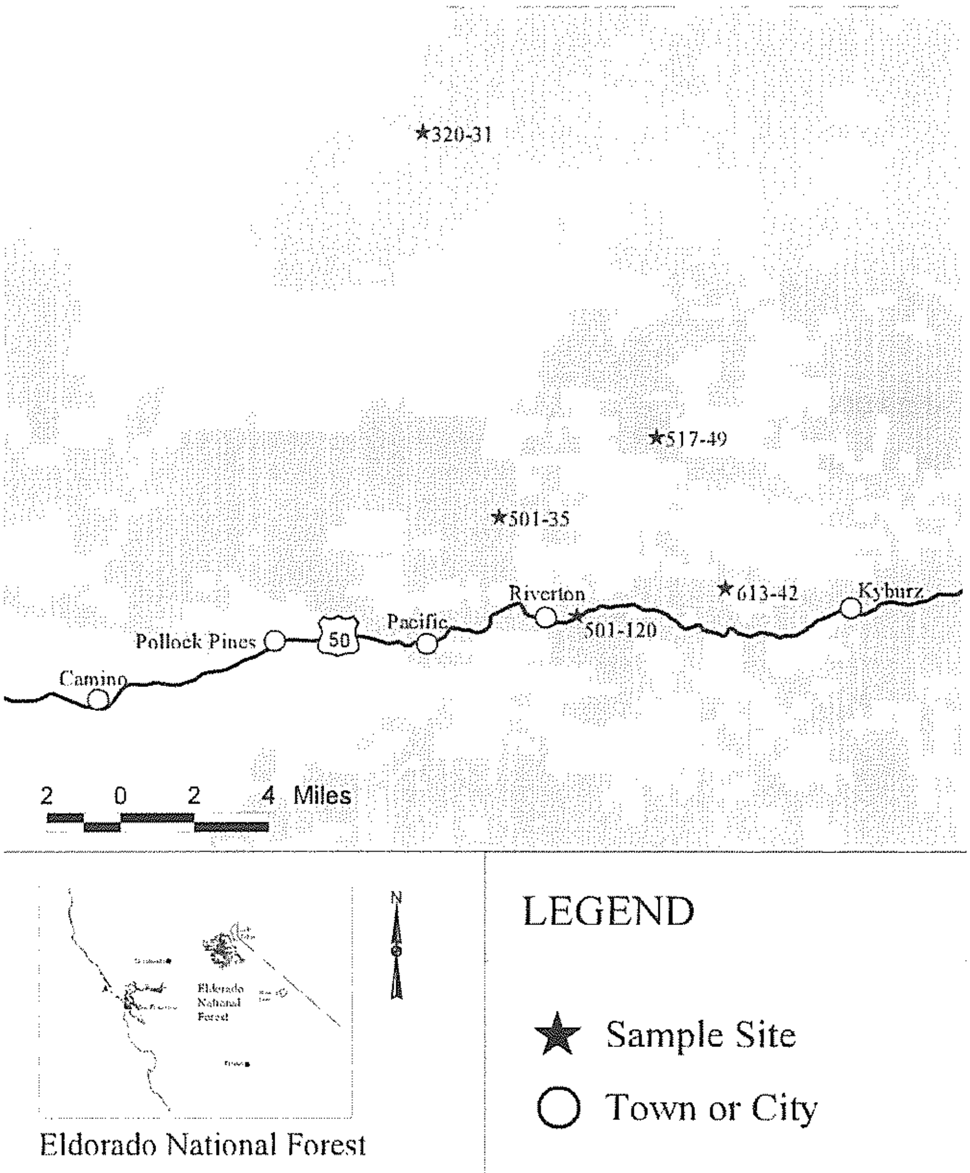
APPENDIX A

Site Locations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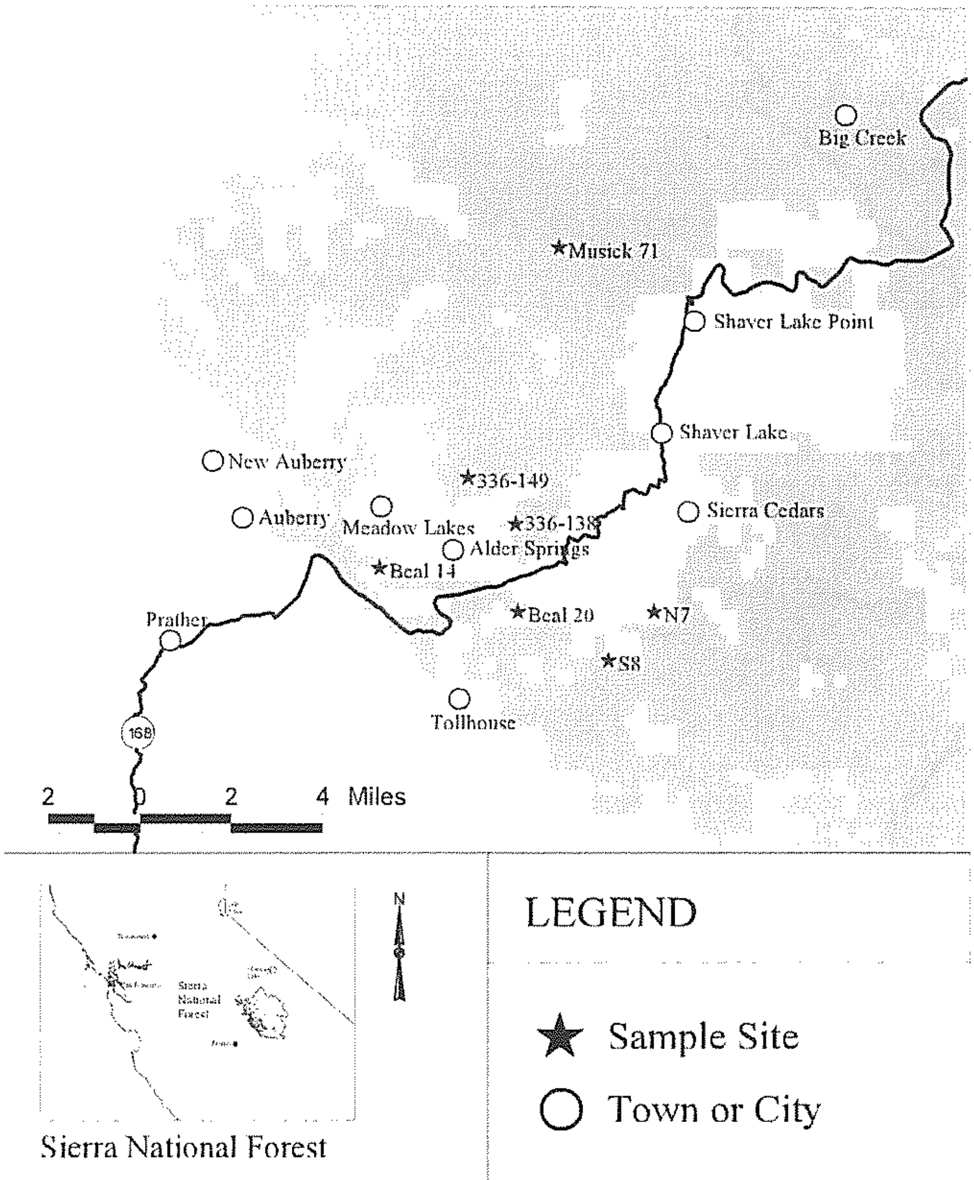
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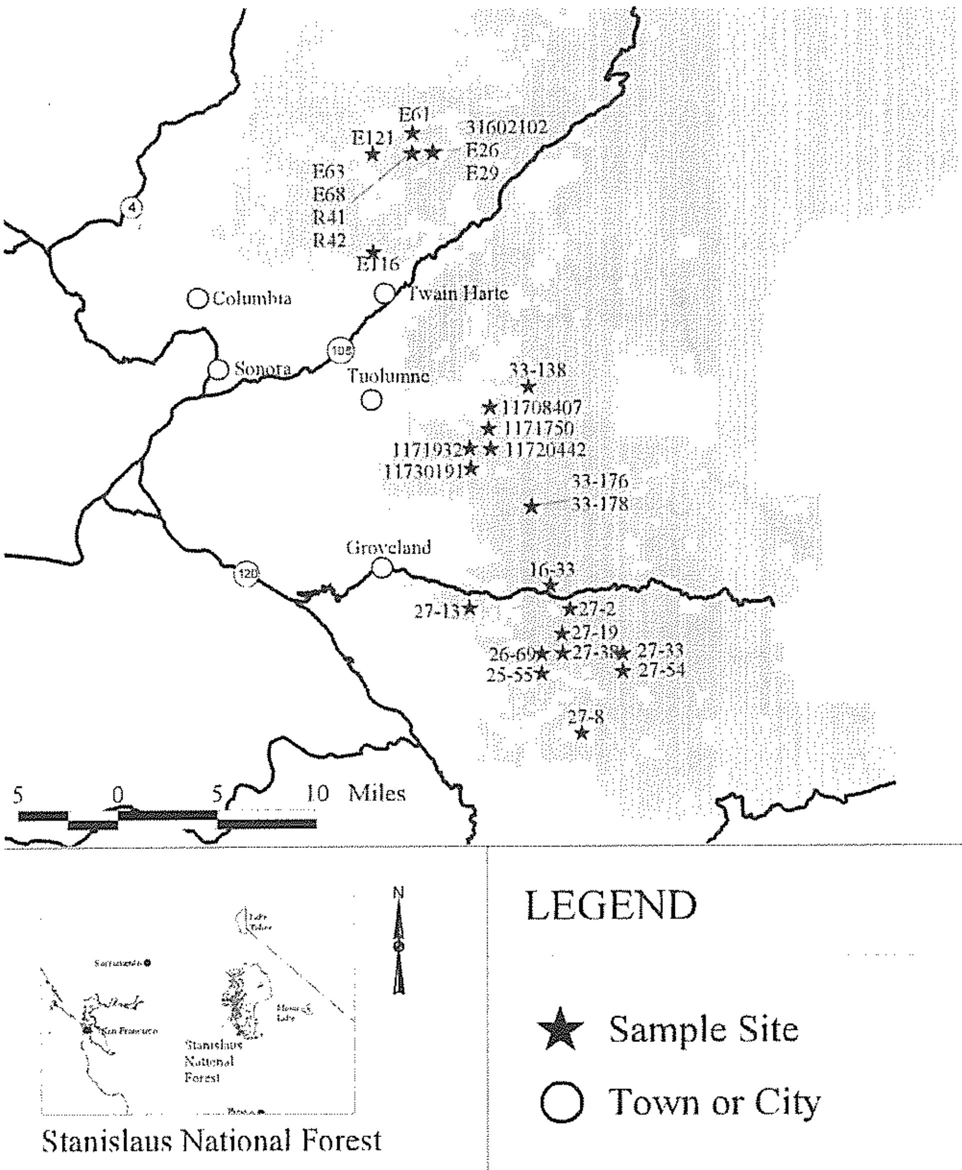
Town or City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Groveland
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27-19
26-69
27-38
27-33
25-55
27-54
27-8
5 0 5 10 Miles
N
Stanislaus National Forest
LEGEND
Sample Site
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