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HERBICIDE DISSIPATION STUDIES IN SOUTHERN FOREST ECOSYSTEMS

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Abstract– Results of research on the movement of hexazinone, imazapyr, picloram, and sulfometuron in first-order watersheds in the southern United States are presented. Herbicides contaminate surface waters to varying degrees, depending on application rate, method of application, product formulation, and site-specific characteristics. Highest concentrations are observed in streams in ephemeral pulses during the first three storm events after application. Streamside management zones greatly reduce the amount of herbicide entering streams from forestry applications. Soil persistence of herbicides is highly variable and a function of many site characteristics. Plant residues have been monitored and found to dissipate rapidly, with half-lives <40 d.

Keywords – Hexazinone Imazapyr Picloram Sulfometuron methyl Fate

INTRODUCTION

Concern for pesticide use permeates nearly every aspect of our daily lives. The occasional abuse or *misuse* of farm chemicals has been widely repotted. During 1988, community, county, and state watch groups were formed in some states to forge public awareness of widespread low-level food contamination (most within Environmental Protection Agency [EPA] food tolerance levels) from agricultural use of pesticides. Public attention thus focused on pesticides in agriculture has spilled over into use of pesticides in forestry. But, whereas forestry-related uses involve one or two applications over a span of 30 to 80 years, agricultural applications are to each crop in its season every year. The low incidence of forestry use (Tables I and 2), relative to agricultural practice, and the benefits (e.g., improved water quality as a result of decreased erosion compared with mechanized site preparation methods, improved wildlife habitat from edge effect and resprouting vegetation, improved game and nongame food availability) are reasons for the popularity of herbicides in forestry. Although risks do exist, risk/benefit should always be considered holistically, with due regard to risks associated with the alternatives.

Most environmental fate and impact concerns associated with the use of forest herbicides are related to off-site movement during and after application. The environmental fate and ecosystem impacts of forest herbicides are governed by movement and transformation in the atmosphere, aboveground vegetation, soil surface, soil rooting zone, unsaturated zone below the rooting depth, and ground water. Herbicides and their breakdown products are transported within ecosystems mainly through the water cycle, but drift, volatilization, photodecomposition, and other forms of degradation also affect movement directly or indirectly. Precipitation and evaporation are the principal driving forces in the processes of runoff, leaching, and plant uptake, and they have been discussed in great detail for forest watersheds [1–3].

The interaction of water cycle driving forces and other processes that affect herbicide persistence with

Table 1. Annual herbicide treatment of National Forest land in the United States [12]

	U.S.	South. U.S.
Total ha (millions)	***34.2***	***4.8***
Treated ha (millions)	***0.047***	***0.007'***
Percentage treated	0.14	0.15

'Sixty-two percent treated for timber management, remainder for rights-of-way, wildlife management, and noxious weed control.

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Table 2. Herbicides used on more than 200 ha of National Forest lands in the United States in 1988 [12][a]

Herbicide	Applied a.i. (kg)	Treated (ha)
Hexazinone	26,981	12,171
Triclopyr	17,889	11,099
2,4-D	9,507	9,332
Picloram	2,157	7,969
Glyphosate	5,519	4,585
Dicamba	1,256	1,873
Sulfometuron methyl	313	1,597
Tebuthiuron	694	806
Imazapyr	190	300
Fosamine	1,223	258
Ammonium sulfamate	908	213
Metsulfuron	Not available in 1988	

[a] Of 47,124 ha treated nationwide (0.14% of National Forest land), 15% was in the southern United States. Some was treated with herbicide mixtures.

individual herbicide characteristics, climatic factors, soil-water properties, and indigenous organisms makes the behavior of any given herbicide on a specific site difficult to predict. Yet the ability to predict site-specific behavior is the key to hazard assessment, as required by the National Environmental Policy Act of 1969 and its amendments. Furthermore, a lack of knowledge of the site-specific behavior of herbicides hinders development of mitigating management practices.

This paper summarizes the findings of several studies that monitored the environmental fate of forestry herbicides in the southern United States (Table 3*), but it focuses on the highlights of three large research projects: a comparison of off-site movement of a herbicide used for single-stem injection (picloram, sites 14-19) and a. herbicide used for "spot treatment" (hexazinone, sites Z-7); an environmental fatestudy on imazapyr that compared off-site movement from two similar sites, but with and without stream protection via an untreated buffer strip streamside management zone (SMZ); and a study to compare the dissipation of sulfometuron methyl in two formulations on adjacent plots and from two very dissimilar sites, all with streamside protection (sites 22-23).

MATERIALS AND METHODS

This research was conducted on a variety of sites with specific goals; therefore, application methods, product formulations, and applied rates vary according to study-specific requirements (Table 3). All sites were selected, from the many available, for uniformity of watershed conditions. Pretreatment samples were taken, control (untreated) watersheds were maintained, and all necessary precautions were taken to preclude site contamination from inappropriate sources (e.g., washing of equipment, spills). Herbicides used in these studies were formulated in various ways (Table 3).

Single-stem injection is the least intensive (ix., it maximizes effectiveness of the applied herbicide and therefore minimizes the amount used) method of herbicide application in control of wood competition in silviculture. It is also the least hazardous to nontarget organisms (except the applicator) but is the most labor intensive. An alternative method that is less labor intensive is spot treatment. In spot treatment, soil-active herbicides are dispersed either via a pump-action-operated gun that projects a metered amount of liquid to aiming points on the ground or, when hand applied, via pellets thrown at desired locations. Both injection and spot treatment, very popular methods of competition control on National Forest and small nonindustrial, private forest, landowner lands, are thought to be similar in terms of environmental safety. To test this hypothesis a large research project was established to compare stream contamination for the two methods (sites 2-7 and 14-19). Hexazinone, a soil-active herbicide, was applied as the liquid formulation (L) containing 0.24 kg active ingredient (a.i.) per liter (sites 2-7). Applied in spot fashion, 2 ml of hexazinone solution was squirted directly onto the soil at spacings appropriate to the density of competition on each site. The product used in sites 14 through 19 was a liquid composed of 0.48 kg 2,4-D acid equivalent (ax.) plus 0.06 kg picloram a.e. per liter. Individual stems were treated with this formulation, which was applied by stem injection either basally using a tubular injector with a 5-cm blade or by hypo-hatchet at waist height. Spacing of injections on individual trees varied slightly but generally was one injection for each 5 cm of diameter at breast height. Injectors were typically set to deliver 2 ml of solution to each cut surface. The number and size of stems to be controlled on a site, therefore, dictated the actual application rate.

Hexazinone was applied as the liquid formulation (L) containing 0.24 kg a.i. per liter or as a pellet (P) formulated with 10% a.i. on a clay substrate approximately 2 cm' in volume to other sites listed in Table 3. Formulated as the liquid, hexazinone was broadcast via tractor-mounted boom sprayer (BCG) or by spot application directly to the soil (SPOT). Pellets were applied by hand in the same

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*Site numbers in text refer to those in Tables 3 and 4.

Table 3. Maximum observed herbicide residues in stream flow from herbicide fate research sites in the southern United States

			Application				
Herbicide[a]	Site	Location	Form[b]	Method[c]	Rate[d]	Source	Surface water[e]
Hexazinone	I	Arkansas	L	SPOT	2.0	[13]	I4
Hexazinone	2	Alabama	L	SPOT	2.3	M[f]	8
Hexazinone	3	Alabama	L			M	24
Hexazinone	4	Alabama	L	SPOT SPOT	2.9.9	M	37
Hexazinone	5	Alabama	L	SPOT	2.9	M	23
Hexazinone	6	Georgia	L	SPOT	1.6	M	6
Hexazinone	7	Georgia	L	SPOT	1.6	M	9
Hexazinone	8	Florida	L	BCG	1.7	M,N[f]	1.3
Hexazinone	9	Alabama	P	BCA	0.8	[14]	2,400
Hexazinone	10	Georgia	P	SPOT	1.7	[15]	442
Hexazinone	II	Tennessee	P	BCA	I.7	[16]	ND
Imazapyr	12	Alabama	L	BCA	2.2	M[g]	680
Imazapyr	13	Alabama	L			M	130
Picloram	14	Georgia	L	BCA INJ	2.0.3	M	ND
Picloram	15	Georgia	L			M	ND
Picloram	16	Georgia	L	INJ	0.8.3	M	6
Picloram	17	Kentucky	L	INJ	1.3	M	21
Picloram	18	Kentucky	L	INJ	0.3	M	10
Picloram	19	Tennessee	L	INJ	0.6	M	4
Picloram	20	Alabama	P	BCA	5.6	[17]	241, 77[h]
Picloram	21	N. Carolina	P	BCG	5.0	[18]	10
Sulfometuron methyl	22	Mississippi	P	BCA	0.4	M,N	23
Sulfometuron methyl	22	Mississippi	DG	BCA	0.4	M,N	44
Sulfometuron methyl	23	Florida	P	BCG	0.4	M,N	5
Sulfometuron methyl	23	Florida	DG	BCG	0.4	[19]	7

[a] Hexazinone – E.I. du Pont de Nemours Inc., Wilmington, DE. Imazapyr – American Cyanamid Co., Wayne, NJ. Picloram – DowElanco Inc., Indianapolis, IN. Sulfometuron methyl -E.I. du Pont de Nemours Inc., Wilmington, DE.

[b] Formulations were liquid (L), dispersible granules mixed in water for spraying (DC), and pellet/granular (P).

[c] Methods of application were stem injection (INJ), soil spot application in a grid network (SPOT), broadcast aerially (BCA), and broadcast by ground equipment or by hand (BCG).

[d] Active ingredient (a.i.) applied in kilograms per hectare.

[e] Expressed as micrograms per liter. ND = not detected. Detection limits of analytical method are 1 μg/L, except for site 9 (not available) and site I I (10 μg/L).

[f] Unpublished data: M-J.L. Michael; N- J.L. Michael and D.G. Neary.

[g] Herbicide fell directly into stream during application.

[h] Postapplication residues. Maximum observed during application was 442.

spot fashion as the liquid or were broadcast aerially by seeders attached to the undercarriage of helicopters (BCA).

Picloram in noninjection studies on sites 20 and 21 was applied as extruded clay pellets containing 10% a.e. by weight. Pellets were either applied by hand (BC) or aerially broadcast from a helicopter (BCA).

Sulfometuron methyl was applied as an experimental pellet formulation and as the dispersible granule in water in a study on separate watersheds in Mississippi (site 22) and in Florida (site 23). The Mississippi watersheds were aerially treated, whereas the application in Florida used ground equipment.

Sampling for water was accomplished in several ways in these studies. Most often used was some form of time-sequenced sampling in which water was collected either manually or by electronic sampling devices at predetermined times or intervals. A technique often used to reduce analytical costs was compositing of time-sequenced samples. Two additional sampling strategies are based on stream stage or flow volume. Flow-proportional sampling used in conjunction with compositing was accomplished with a Coshocton wheel attached to a 30-cm H-flume (site 20, Table 3). The Coshocton wheel is a device that takes a continuous sample of 0.5% of flow. When 0.5% of all flow is collected for a day and subsampled, the subsample represents a flow-averaged sample for the day. Another flow-related sampling strategy was flow-initiated sampling. Discrete samples were taken by electronic samplers at predetermined flow volumes or stream stages. Necessary flow volumes were estimated from weather forecasts of impending storm size, existing soil moisture conditions at the time of the sampling, and *a priori* knowledge of the hydrologic response of the watershed under study.

Herbicide persistence was determined in some studies for soil (sites 1-8, 12-13, 20, and 22-23) and plants (sites 8, 12-13, and 22-23). Dissipation curves were developed from simple linear regressions using logarithm of concentration as the dependent variable and days after treatment as the independent variable. Half-life, the time required for dissipation of half of the applied material, was determined from the regression lines.

RESULTS AND DISCUSSION

Water

Residues of herbicides in surface water from injection-treated sites ranged from not detectable (ND) to 21 µg/L, for soil spot-applied from 6 to 442 µg/L, for broadcast by ground equipment from 1 to 10 µg/L, and for broadcast aerially from ND to 2,400 µg/L (Table 3). The very high (442 µg/L) residues from spot treatment in site 10 are the result of placing pellets directly in ephemeral drainage channels. The very low (ND) levels in site 11 relative to other aerially broadcast treatments are probably due to the small portion (10-18%) of the study watersheds that were treated and the low sampling frequency. In similar studies approximately 75 to 90% of each watershed was treated.

One study was established on similar sites (2-7 and 14-19) in several states to obtain some information on the variability of watershed responses to herbicide treatment and to compare off-site movement for the two herbicides. The stem injection method resulted in the lowest frequency and magnitude of herbicide residues in these 12 sites, but there was also a correlation with rate. Simple linear regression indicated 70% of the variation in maximum observed stream residues was accounted for by applied rate for both methods of application.

One study (sites 22-23) compared formulations on two soil types: a clay in Mississippi and a deep sand in Florida. A dispersible granule was applied as a spray, and a pellet formulation was applied at the same rate in each study. In both studies a higher frequency of residue-positive samples was detected for the pellet formulation, leading to some speculation that the principal mode of transport to the stream was via short-distance, overland flow. Samples with nondetectable residues were frequently interspersed among samples with quantifiable residues, indicating movement of sulfometuron methyl through the environment in more or less discrete pulses. The pulses were highly correlated with storm events. This pulsed movement of sulfometuron methyl was probably a reflection of low K_D (soil sorption coefficient), which would minimize advective dispersion both in and on soil and in organic matter. Introduction of sulfometuron methyl to the stream by base flow was negligible.

The single study with imazapyr (sites 12 and 13) compared two similar watersheds, but an SMZ was maintained on one and not on the other. Recommendations for use of SMZs arise from best management practices (BMPs) promulgated by state and industry environmental concern groups. Previously the best evidence for effectiveness of SMZs came from agricultural research on "filter strips," in which grass strips are used to slow runoff so suspended solids carrying adsorbed pesticide can settle out before reaching streams and other bodies of water [4]. Maximum residues detected on the site without an SMZ were more than five times higher, and the frequency of residue-positive samples was much higher, than those on the buffered site. Short-distance, overland transport probably accounts for the differences in levels of stream contamination in this study. Flow sequence samples taken during the first storm (30 mm) after application indicated movement of imazapyr occurred as a discrete parcel or wave front with a maximum observed concentration of 130 µg/L. Imazapyr residues were not observed in the stream until approximately the time of peak discharge. After the storm event, as runoff flow decreased, imazapyr concentration rapidly decreased and within 9 h dropped to trace or nondetectable levels. A similar pattern of stream contamination was observed for the eighth precipitation event (>10 mm) following application, but imazapyr concentrations were greatly reduced (maximum observed was 6 µg/L) and decreased to nondetectable or trace levels within 1.5 h.

The relative level of surface water contamination is dependent on distance of the sampled water from the treated site. On site 20, surface water samples taken from a stream within the treated site contained a maximum observed postapplication residue concentration of 241 µg/L, whereas samples taken from the same stream 0.4 km below the treated area contained a maximum concentration of 77 µg/L. Presumably, dilution resulting from inputs in the lower reaches of the study watershed explains the observed differences.

Herbicide residues have been detected in ground water only from broadcast treatments and only in about half the studies that monitored for them, but not in confined aquifers. Most of the detected residues were from lysimeters placed 1 m in depth-sampling surficial water tables. Picloram residues in ground water on site 20 (1,830 μg/L) were from a lysimeter installed 1 m deep in an ephemeral stream bed that did not develop surface flow during the course of the study. Picloram was also detected on another site in a lysimeter sampling soil solution at a depth of 1 m (site 21, 350 μg/L), and spring flow from this site contained trace levels of picloram. Hexazinone residues were detected in seven samples from 23 6-m sampling wells in very deep sand on site 9 (maximum value was 69 μg/L). All wells on this site were sampled during the 14 months immediately following application. Two springs arising from the site were sampled during this period, but hexazinone was not detected in any of these samples.

Soil

Residues and persistence of hexazinone, imazapyr, picloram, and sulfometuron methyl have been monitored in forest soils in the South (Table 3). Half-lives reported for these sites (Table 4) are highly variable due to the influence of site-specific characteristics (i.e., application rate, precipitation, soil, temperature). Whereas rates of first-order chemical kinetics are not concentration dependent, field observations of half-life are dependent on initial concentration (application rate) due to the influence of nonchemical routes of herbicide disappearance (e.g., storm runoff, leaching). Soil composition is particularly important in determining herbicide persistence. Clay and organic matter adsorb herbicide molecules to varying degrees, and this interaction results in advective dispersion and retarded movement for water-soluble herbicides. Organic matter content is probably the most important soil characteristic affecting the soil sorption coefficient (K_D) [5] and greatly affects microbial degradation through its influence on microbial population density. In these studies (except site I, not given), organic matter content was very low (<2%).

Table 4. Herbicide persistence in forest soils and plants following application for vegetation management

Herbicide	Rate (kg/ha)	Half-life” (days)	
		Soil	Plants[b]
Hexazinone	1.6–2.9	11–180	4–15 (5)
Imazapyr	2.2	34-65	15-37 (C)
Picloram	5.0	131	
Sulfometuron methyl	0.4	5-33	4–11 (9)

[a] All values are from J.L. Michael, and J.L. Michael and D.G. Neary (unpublished data), indicated in Table 3 except the hexazinone half-life of I I d, which was approximated from published data [13], and picloram calculated from published data [17]. See text for method of calculation.

[b] Numbers of considered species are in parentheses; C = composited vegetation.

Microbial degradation produces metabolites that may have herbicidal properties. Microbial degradation of hexazinone results in five major metabolites [6], but only one of the metabolites retains any of hexazinone's ability to inhibit photosynthesis, and that is at reduced levels [7].

The fate of imazapyr was studied in Piedmont and Upper Coastal Plain soils of Alabama following aerial application of 2.24 kg a.e. per hectare. Imazapyr had a half-life of 34 to 65 d (Table 4). It was detected below 30 cm in only 1% of the studied samples. Movement below 30 cm was probably due to mass transfer in solution flow through large macropores (rotted nut stump holes and root channels).

Sulfometuron methyl was studied on a clay loam soil in Mississippi and a sandy soil in Florida following application of 0.42 kg a.i. per hectare. Soil half-life ranged from 5 to 33 d (Table 4), and although slightly more mobile than imazapyr, sulfometuron methyl was not detected below 30 cm at either site. Sulfometuron methyl is rapidly hydrolyzed under acidic conditions to form saccharin and methyl-2-aminosulfonylbenzoate at a rate governed by soil pH and moisture [S-IO]. Other significant literature has been reviewed [1 I] and will not be covered in this paper.

Plants

Studies conducted in the southern United States to determine herbicide residue levels in plant tissue are limited in number and scope, but in general half-lives do not exceed 37 d (Table 4) and residue levels are species- and application-rate dependent. Plant tissue residues are usually highest immediately after application and decline very rapidly for the first few days following application, even in the absence of precipitation, indicating possible mechanical loss from foliar surfaces. Rapid translocation to underground plant parts may also contribute to the observed short half-life. Maximum observed plant tissue concentrations in field studies ranged from 15 to 130 mg/kg (based on plant tissue fresh weight) for each kilogram a.i. per hectare.

CONCLUSIONS

Herbicides that are broadcast applied to forest sites contaminate surface waters to varying degrees. There is a direct correlation between the applied rate and maximum observed surface water concentrations. In general, surface waters are more likely to be contaminated by aerial applications and least likely to be contaminated by stem injection. Soil spot treatments result in only slightly higher levels of contamination than those observed with stem injection. However, improper application directly into drainage channels results in higher contamination, whereas maintenance of an SMZ beside drainage channels greatly reduces the degree of contamination. Product formulation may also affect the degree of contamination, with pelleted formulations contributing more to streams than the same herbicide applied as a liquid. The data base on groundwater contamination from forest herbicide use in the southern United States is very limited. Few studies have focused on the nonpoint-source aspects of forest herbicide fate and transport. Some information for typical forest operations is available from these studies. There are few data available on ground water, but contamination has been observed to a slight degree in surficial water tables on a very localized basis. All of these data are from unconfined water tables near the soil surface, and there have been no observations of aquifer contamination from use of forest herbicides.

It is difficult to equate the levels of stream contamination observed in these and other studies with toxicological significance because toxicological testing methods are based on static exposures and do not reflect the pulsed movement of herbicides in streams from treated sites. Nor are there adequate data available on the impacts of low-level, ephemeral herbicide concentrations on aquatic ecosystems.

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