WATER RESOURCES BULLETIN
VOL. 20, NO. 6 AMERICAN WATER RESOURCES ASSOCIATION DECEMBER 1984

SIMULATION OF HERBICIDE CONCENTRATIONS IN STORMFLOW FROM FORESTED WATERSHEDS[1]

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ABSTRACT: The breakpoint rainfall hydrology and pesticide options of the field scale model CREAMS (Chemicals, Runoff, and Erosion from Agricultural Management Systems) were used to predict average concentrations of hexazinone [3 cyclohexyl-6-(dimethyl-amino)-1-methyl-1,3,5-triazine-2,4(1H,3H)-dione] in stormflow from four forested watersheds in the upper Piedmont region of Georgia. Predicted concentrations were compared with measured concentrations recorded over a 13-month period. CREAMS accurately predicted hexazinone concentrations in the initial stormflow events which also contained the highest concentrations. The model underestimated the hexazinone concentrations in stormflow two months and greater following pesticide application. In a companion study, the daily rainfall option of the CREAMS model was used to evaluate the relative risk associated with the maximum expected concentration of hexazinone, bromacil (5-bromo-3 sec-butyl-6 methyuracil), picloram (4-amino-3,5,6 trichloropicolinic acid), dicamba (3,6-dichloro-0-anisic acid), and triclopyr {[(3,5,6-trichloro-2-pyridinyl)oxy]acetic acid} in stormflow from small forested watersheds. The model predicted the following order of potential residue appearance in stormflow: bromacil > triclopyr > hexazinone > picloram > dicamba. Subsurface movement of residues via interflow and deep leaching losses are not simulated by the version of CREAMS used in these studies.
(KEY TERMS: CREAMS; field scale model; herbicide residue; stormflow; forests.)

INTRODUCTION

Development of new herbicides for agriculture and forestry uses has focused on producing compounds which are not only effective herbicides but also environmentally safe. Safety concerns not only human exposure hazards, but also the interactions between biodegradability, movement in the environment, and nontarget impacts as they affect productivity of local and distant ecosystems. Because of the wide range of new herbicides being developed and marketed, land managers have a need to evaluate the off-site movement potential of alternative herbicides. The availability of a usable model for evaluating pesticide movement in agricultural and forest ecosystems has been the key to developing and implementing best management practices to limit nonpoint source pollution from organic chemicals to acceptable levels.

Over the past 15 years there has been a fairly intense effort to model pesticide loss from agricultural areas (Bailey, *et al.*, 1974; Bruce, *et al.*, 1975; Crawford and Donigan, 1973; Haith, 1980; Knisel, 1980; Wauchope and Leonard, 1980). All of the models function on an edge-of-field basis, and most deal only with surface movement of pesticides. Most models have applications to herbicides used in forests, but none have been specifically developed for the edaphic and hydrologic conditions of forest ecosystems. In most reported field studies, maximum herbicide concentration and total amounts lost in runoff under forested conditions were less than those reported for agricultural land (Newton and Norgren, 1977; Norris, 1981; Wauchope, 1978).

A pesticide simulation model developed recently for agricultural use is CREAMS (Chemicals, Runoff, and Erosion from Agricultural Management Systems) (Knisel, 1980). The model is a physically based, daily simulation model that estimates stormflow, sediment transport, nutrient, and pesticide movement from field-sized areas. The pesticide submodel provides procedures to evaluate the effects of alternative chemical selection on potential pesticide residue losses in stormflow. The model's benefit to land management decision-making processes lies in its ability to provide relative comparisons among alternatives rather than to provide absolute pesticide concentration predictions. Simulation of pesticide movement by CREAMS provides total mass and storm-mean concentrations on a field-edge basis and does not provide information on downstream routing or subsurface fate (Leonard and Nowlin, 1980).

Hexazinone, symmetrical triazine herbicide, has become widely used in southern forests because it is effective in controlling a broad spectrum of annual and perennial weeds at herbicide rates tolerated by many conifers (Gonzalez, 1980; Hamilton, 1979; Michael, 1980; Neary, *et al.*, 1981). It is manufactured under the trade name "Velpar® Gridball® Brushkiller" by E. I. DuPont de Nemours & Company, Inc., as 10 percent active ingredient (a.i.) granular and pellet formulations. (Use of trade and corporation names does not constitute

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endorsement by USDA but is provided as a reference.) The selective action, ease of application, and low toxicity of the compound indicate its considerable potential as a cost-effective silvicultural tool. Off-site movement of hexazinone in stormflow from small forested watersheds (<1 ha in size) was evaluated by field studies and reported in considerable detail by Neary, *et al.* (1983). Loss of hexazinone in stormflow over a 13-month period from four treated watersheds averaged 0.53 percent of that applied. Site-specific models of hexazinone residue behavior have been produced by Wauchope and Leonard (1980) using a power-curve function and a general concentration estimation equation.

Several interesting questions regarding simulation potential arose during the course of the hexazinone study reported by Neary, *et al.* (1983). The first had to do with how accurate a nonsite specific model like CREAMS could be. Field testing of CREAMS with atrazine and toxaphene under agricultural use showed that the model accurately reproduced field runoff data and estimated chemical residue losses within reasonable limits (15-30 percent) (Lorber and Mulkey, 1982; Steenhuis, 1979). Given that CREAMS is not designed to make absolute predictions and was designed for agricultural situations, how accurate could it predict hexazinone concentrations in stormflow from small forested watersheds? Secondly, if the CREAMS predictions of hexazinone concentrations were adequate, how would alternative herbicides rate in terms of runoff potential on the hexazinone study site? This paper addresses both these questions and draws conclusions regarding the use of CREAMS as a management tool for assessing the impact of forest vegetation control decisions on water quality.

METHODS

The Hexazinone Field Study

The hexazinone study site is located in the upper Piedmont of the southeastern United States on the Chattahoochee National Forest of north Georgia within the drainage of Moonshine Creek, a tributary of the Broad River. A complete description of the study site is given by Neary, *et al.* (1983). Briefly, the site consists of five sub-watersheds with well-defined ephemeral channels (Figure 1). The channels are indicative of erosion induced by past agricultural practices. The watersheds are wide and bowl-shaped in their upper reaches, incised at their midpoints, and broad-bottomed in their lower reaches. The channel area in the lower reaches contain fine-textured, alluvial deposits which occasionally reach 2 m in depth. Soils on the ridges and slopes are sandy loam typic Hapludults of the Cecil series with an A horizon 0 to 10 cm thick overlying a massive B2t horizon. The A horizon increases in thickness on the slope toe.

The 60- to 80-year-old mixed pine-hardwood overstory contained scattered shortleaf pine (*Pinus echinata* Mill.), chestnut oak (*Quercus prinus* L.), white oak (*Q. alba* L.), black oak (*Q. velutina* Lam.), dogwood (*Cornus florida* L.), sourwood (*Oxydendrum arboreum* (L.) DC.), hickory (*Carya glabra* (Mill.) Sweet), and various other species (Neary, *et al.*, 1981). The sparse understory consisted mainly of the same overstory hardwood species and various herbaceous plants.

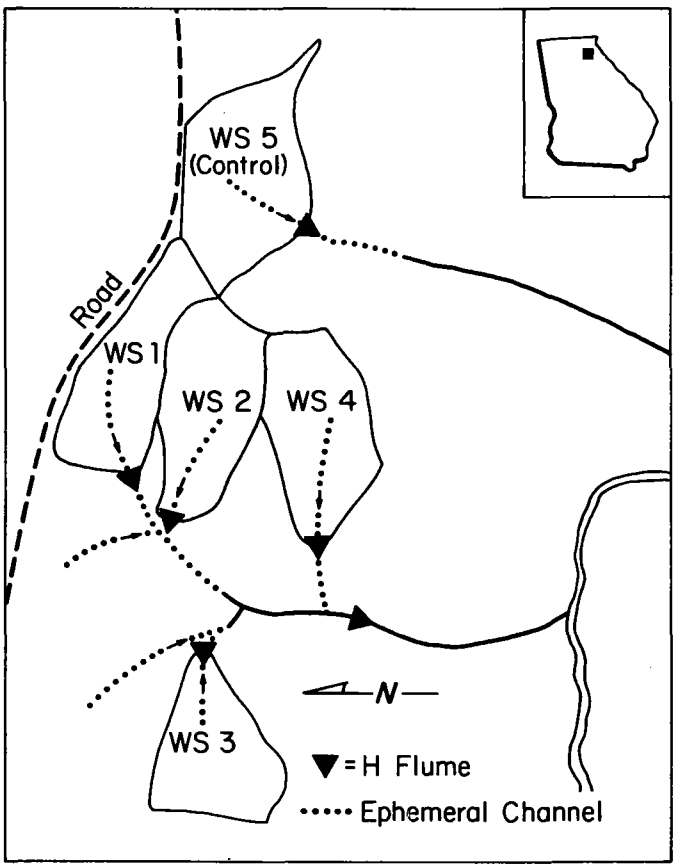

Figure 1. Watershed Locations and Stream Gauging Stations for the Hexazinone Study Area (WS 1 through WS 4 received 1.68 kg/ha a.i. hexazinone; WS 5 served as the control).

Five watersheds (WS 1 through WS 5), 0.85 to 1.09 ha in size (Figure 1), were instrumented with 30-cm H-flumes, Coshocton wheel flow-proportional samplers, analog water level recorders, and sample collection systems. All flow occurred as stormflow; i.e., at no time during the study period did baseflow occur. Standard storage and recording weighing-bucket rain gauges were installed in a clearing on a ridge to the north of WS 5.

Watersheds WS 1 through WS 4 plus some surrounding areas were treated with 1.68 kg a.i./ha of hexazinone (10 percent a.i. pellets) on April 23, 1979. Pellets containing herbicide were placed, by hand, at a spacing of 1.8 x 1.2 m to achieve the desired application rate. Because an aerial application was to be simulated, pellets were placed at the grid spacing regardless of channel location.

Water samples were collected from 26 stormflow events from April 26, 1979, to May 27, 1980, and the hexazinone

residue levels were determined as the trifluoroacetate derivative using nitrogen-selective gas chromatography (Holt, 1981).

CREAMS Computer Simulation

The CREAMS model is composed of a series of submodels linked together to yield an integrated estimate of stormflow, percolation, erosion, and dissolved and absorbed plant nutrients and pesticides. The model is physically based, does not require calibration for use, is somewhat easily understood, requires few and readily available input parameters, and represents the physical system relatively accurately (Knisel, 1980). As a field scale model, CREAMS defines a field unit as a management unit with a single land use, relatively homogeneous soils, and spatially uniform rainfall. The hydrology submodel drives the remaining submodels; i.e., the hydrologic processes provide the transport medium for sediment and chemicals.

The hydrologic section processes precipitation inputs in one of two ways to generate predicted stormflows. If daily precipitation totals are input, stormflow is predicted using the SCS runoff curve number method. If time-depth precipitation data (termed breakpoint data by CREAMS) are used, an infiltration curve based on the Green-Ampt infiltration equation is generated. Each stormflow generation procedure is discussed in detail by Knisel (1980).

Both the precipitation input options were used in this study. Breakpoint type rainfall data, i.e., hourly rainfall, measured during the course of the hexazinone application study were used to generate predicted stormflows. The simulated stormflow data were then used to drive the pesticide model to simulate hexazinone concentrations in stormflow for comparison with the field measured values. Since there was a negligible average of 400 g of sediment loss per watershed over the 13-month sampling period (Neary, *et al.*, 1983), the erosion portion of the CREAMS model was by-passed and the stormflow results of the hydrologic model segment were passed directly to the chemical segment of the model. The model algorithms are described in detail in Knisel (1980).

In a companion study, the daily rainfall submodel was utilized to evaluate the risk to water quality by the application of five different herbicides to the forested watersheds used in the hexazinone study. For this portion of the study, 50 years of daily rainfall data were used.

The average physical parameters for the site required by both the breakpoint and daily rainfall hydrology submodels are presented in Table 1. Values were taken from published sources (Holtan, *et al.*, 1968; Neary, *et al.*, 1983) or calculated according to instructions in the CREAMS manual (Knisel, 1980). Required input data to the hydrology submodel other than rainfall are mean monthly temperature and radiation, watershed physical parameters, and soil hydraulic properties.

The pesticide submodel of CREAMS can account for multiple applications of the same herbicide applied to soil or foliage. The submodel allows specification of different decay rates for chemical residues placed on foliage or in the soil. Movement of pesticide below the surface one cm of soil is estimated for highly soluble compounds, but vertical and horizontal movement at greater depths over time are not simulated by the submodel. Concentrations of herbicide in solution and adsorbed onto sediments, as well as the mass transported by each process, are calculated. Pesticide residues remaining in the surface one cm of the soil residue and total pesticide loss are totaled after each storm (Leonard and Nowlin, 1980).

TABLE 1. Mean Hydrologic and Soil Physical Characteristics for the CREAMS Hydrology Submodel.

Parameter	Value	Units
Rooting depth	91	cm
Saturated conductivity	1.27	cm/hr
Field capacity (as fraction of saturation)	0.70	*
Initial soil-water storage fraction	0.75	*
Soil porosity	0.41	cm^3/cm^3
Immobile soil water content	0.17	cm/cm
Upper limit of plant available soil water storage	0.24	cm/cm
Runoff curve number	55	*
Channel slope	0.20	ft/ft
Watershed length-width ratio	0.056	*

*Non-dimensional.

The most significant input parameters into the CREAMS pesticide submodel are listed for each of the five herbicides evaluated (Table 2). Application rate and date determine the amount of herbicide exposed to storm runoff events, as this is a function of loading and rainfall frequency patterns. Solubility is an indication of a herbicide's tendency to be leached from the soil surface by infiltrating rainfall. For pesticides with a water solubility greater than one ppm, CREAMS assumes that the availability of the chemical for vertical transport with infiltrating water is described by a pesticide distribution coefficient, K_d. This coefficient is defined as the ratio of pesticide concentration in the soil to the concentration in solution at equilibrium. The main assumptions regarding K_d are that it is independent of pesticide concentration and that adsorption/desorption processes in the soil are rapid, reversible processes. Values for K_d vary with soil type, being primarily related to percent organic content or calculated specific surface of the soil (Pionke and DeAngelis, 1980). Another important pesticide parameter used in CREAMS is the decay constant, K_s. Pesticide dissipation from the surface one cm of soil is described by a simple exponential function which combines photodegradation, volatilization, microbial breakdown, and other processes. The decay constant is then used in a first-order rate expression to compute surface concentrations over time.

To evaluate runoff potential and to assess the potential risk of maximum concentrations appearing in stormflow for five different herbicides (hexazinone, bromacil, triclopyr, picloram, and dicamba), CREAMS simulations were run for each herbicide using daily rainfall records for Cornelia, Georgia (Climatological Data, National Oceanographic and Atmospheric Administration, U.S. Department of Commerce)

and the mean watershed characteristics. To generate sufficient frequency of stormflow events the hydrology and pesticide submodels were run for 50 years (1931-1980) with the specified herbicide applications repeated on the same date (May 1) each year. In this way a probability of occurrence can be associated with the concentration of herbicide in stormflow following application.

The simulation results were tabulated and the probability of occurrence for various stormflow concentrations were calculated using standard hydrologic techniques. This approach was used in an earlier study (Nutter, *et al.*, in press) and suggested by Wauchope (1978) to evaluate potential impacts of pesticide runoff to downstream aquatic systems. The maximum annual simulated concentration for each herbicide (i.e., the annual series) was selected and the probability of occurrence for each event calculated using the Weibull plotting position formula (Viessman, *et al.*, 1977).

The herbicide rates listed in Table 2 represent assumed loadings on the forest floor irrespective of application system or formulation. Generally, herbicides applied in liquid formulations (available for all five herbicides) have lower simulated losses than solid formulations. This results from an assumption in CREAMS that 30 percent of spray application is lost due to drift and volatilization, 60 percent is deposited on foliage, and 10 percent reaches the ground. Herbicide on foliage has a shorter half-life than herbicide in the soil and may become available to stormflow by being washed from foliage by rainfall. Since data were not available to confirm the drift-foliage-ground distributions for Piedmont forests, loadings in the CREAMS simulation were assumed to be entirely to the forest floor. Use of this assumption results in prediction of the maximum possible stormflow concentrations.

RESULTS AND DISCUSSION

Hydrology Simulation

The results of the breakpoint hydrologic simulation indicate that CREAMS generally underpredicts stormflow on an event-by-event basis (Figure 2). However, total stormflow measured over the length of the 13-month simulation (10.9 cm) exceeded the predicted value (6.2 cm) by 43 percent. The total rainfall for the period was 2740 mm. Lorber and Mulkey (1982) found that the CREAMS stormflow prediction was 85.5 percent of observed whereas in this case predicted stormflow was only 56.9 percent of observed. Greatest disparities were on March 21, 1980 (Figure 2), when 109 mm of rain generated 1.35 cm of stormflow but was predicted to generate 0.25 cm of stormflow, and on September 18, 1980 (Figure 2), when 93 mm of rain generated 0.03 cm of stormflow but was predicted as 1.24 cm of stormflow. Closest agreement was the very large storm on November 1, 1979, when 94 mm of rain generated 1.24 cm of stormflow, predicted as 1.35 cm.

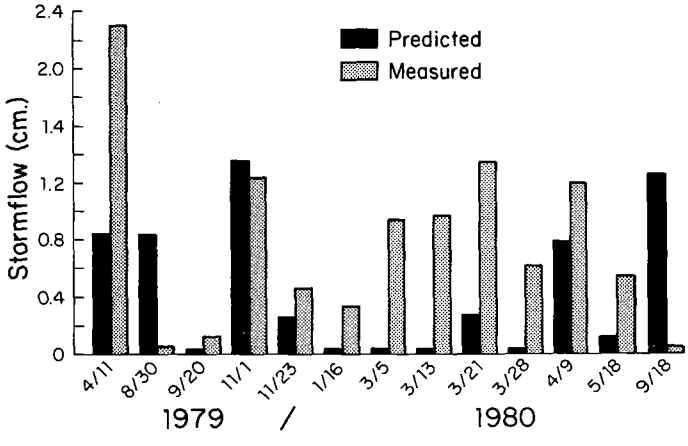

Figure 2. Mean Actual and Predicted Stormflow Volumes for Selected Stormflow Events, 1979 and 1980.

The CREAMS breakpoint hydrology model appears to be weakest in predicting stormflow from long duration, uniform intensity frontal storms. This is illustrated by a predicted cumulative stormflow of 0.03 cm for May 1980 when there was a measured cumulative stormflow of 3.81 cm from a total monthly rainfall of 432 mm. In general, predicted and actual stormflow agreed more closely for short, intense storms. The model is also insensitive to low-volume stormflow events; several (less than 0.13 cm stormflow) occurred which were not

TABLE 2. Significant Chemical and Physical Characteristics of Five Common Forest Herbicides Used as Inputs to the Pesticide Submodel of CREAMS.

Herbicide Characteristic	Unit	Picloram	Hexazinone	Dicamba	Bromacil	Triclopyr
Rate*	kg a.i./ha	0.28	1.68	0.84	6.72	1.40
Application Date	Julian	121	121	121	121	121
Water Solubility	ppm	400,000	33,000	720,000	815	410
K_d		0.7	3.0	0.077	10.0	10.0
K_s		0.0028	0.0100	0.0267	0.0046	0.0150

*Herbicide rates chosen to approximate best forest management practices for the upper Piedmont. Loadings are in kg a.i./ha reaching the forest floor irrespective of application system or formulation. Methods of estimating K_d and K_s are described in the text.

predicted by the model. CREAMS did not predict any stormflow events until 28 days after herbicide application. The first stormflow event actually occurred three days after the application.

Percolation volume and soil water content are also predicted by CREAMS, but these values were not considered in this comparison since there is no accounting of pesticide movement in subsurface water flow except indirectly as the percolate leaves the surface layer.

Hexazinone Runoff Simulation

Outputs from the breakpoint rainfall hydrology submodel were used to drive the pesticide submodel of CREAMS. The results for the simulation of the hexazinone application and its comparison with the field measurements are shown in Figure 3. As expected, the stormflow events with the highest hexazinone concentrations were those closest to the application date. The predicted concentrations decreased over time until 150 days after application, when only small amounts of hexazinone (<0.01 ppb) were predicted to be present in stormflow. The predicted and measured concentration were similar for the first 75 days. However, between 75 and 270 days the measured herbicide concentrations remained elevated at 10 to 20 ppb. In the next stormflow event after 270 days (at 320 days), the herbicide concentration was not detectable.

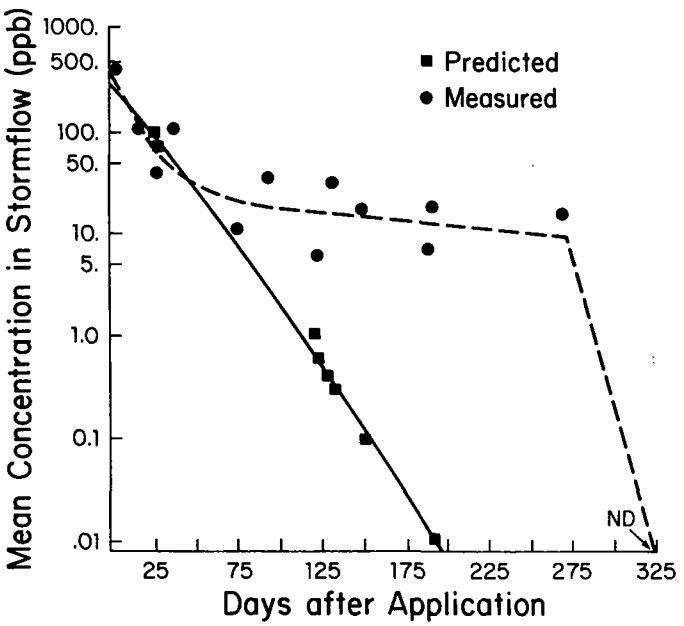

Figure 3. Mean Predicted and Actual Hexazinone Concentrations in Stormflow as a Function of Time After Application.

Predicted and actual hexazinone concentrations in the stormflow are in close agreement for events occurring less than 75 days after application. For storms occurring more than 75 days after application, the model consistently underpredicts hexazinone concentrations in stormflow. The inability to predict hexazinone concentrations after 50 to 75 days may be due to a change in the source of the hexazinone to stormflow. Figure 4 shows the movement of a hexazinone pulse downslope as measured in soil samples taken on the ridge, midslope, and base of slope. The addition of hexazinone to the ephemeral channel region from up slope could account for the hexazinone concentration only slowly decreasing from day 90 to day 275, and also account for the variability in the concentrations. Since the CREAMS model does not account for movement of herbicide within the soil, it would not predict the continual supply of herbicide to the channel region.

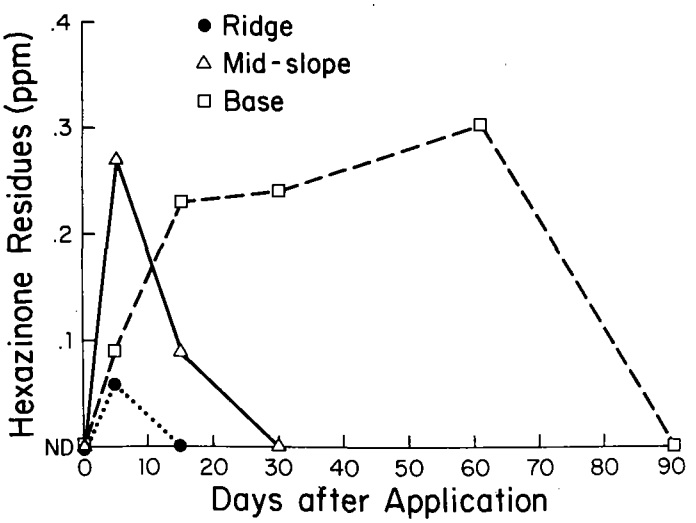

Figure 4. Hexazinone Residues (hexazinone plus metabolite A and B) in the 0-10 cm Soil Depth (after Neary, *et al.*, 1983).

The source area for stormflow is the ephemeral stream channel and the immediate surrounding area. This is the only portion of the watershed where overland flow is likely to occur. Since CREAMS is based on the overland flow origins of stormflow, the ephemeral stream channel is most likely the source of the hexazinone residues in stormflow. Neary, *et al.* (1983), estimated that although 1.5 percent of the hexazinone applied to the watersheds fell into the ephemeral stream channels, 0.53 percent of the applied herbicide was transported off-site in the stormflow during the 397-day period after application. The total loss predicted by CREAMS was 0.09 percent of the applied herbicide, one-fifth of that actually measured. The model apparently predicts the decline of availability of hexazinone in the source area, but cannot include the addition from the delayed, up-slope subsurface flow.

Another possible explanation for the higher measured concentrations after 75 days shown in Figure 3 is hexazinone recycling (Neary, *et al.*, 1983). Hexazinone is absorbed by plant roots and translocated to the site of herbicidal action in the foliage. When the leaves die they begin returning hexazi- to the forest floor via the mechanisms of throughfall and litter

fall. Neary, *et al.* (1983), found the foliage in hexazinone-affected trees contained over 6 mg hexazinone/kg of leaf material. Peak leaf fall of 240 kg/ha/week produced a return of 1.4 g hexazinone/ha/week to the forest floor. Although litter fall may be a possible pathway for herbicide to become part of stormflow, it is expected that it is minor and accounts for only a small fraction.

Long-Term Herbicide Simulations

CREAMS simulations run for a 50-year period of record using the daily rainfall hydrology submodel show that bromacil, hexazinone, and triclopyr have the greatest potential of the five herbicides for movement in stormflow from the forested Piedmont watersheds (Figure 5). Bromacil concentrations declined the least as the recurrence interval decreased. This results from bromacil's tendency to bind to clay and organic matter colloids (high partitioning ratio), and hence its long residence time in the soil surface (Table 2). The concentration of bromacil simulated in stormflow can be expected to be greater than 1000 ppb at least once every 8 years (12 percent probability of occurrence). Hexazinone concentrations can be expected to exceed 100 ppb at least once every 10 years (10 percent probability of occurrence). In contrast, simulated concentrations of picloram and dicamba ($K_d < 1$) never exceeded 30 ppb (Figure 5). Thus, bromacil and triclopyr, with high K_d values and moderate-to-low solubilities, and hexazinone with a high solubility and moderate K_d value produced greatest concentrations in runoff.

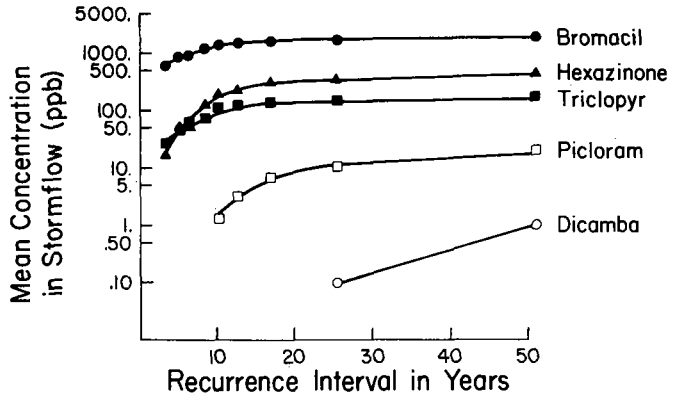

Figure 5. Reoccurrence Intervals of Maximum Annual Concentration for Various Pelleted Herbicide Applications on May 1 Each Year as Predicted From 50 Years of Stormflow and Herbicide Concentration Simulations by CREAMS.

The drop in predicted losses at $K_d < 1$ (picloram and dicamba) can be attributed to reduction in surface concentrations of pesticide due to leaching prior to stormflow. Since the partitioning ratio reflects the tendency to absorb to clay and organic matter colloids, a low K_d means a pesticide has only a weak tendency to bind to colloids. Both herbicides have negative charges and they have only a weak tendency to bind to colloids in the acid soil and are readily leached out of the surface soil horizon. Before CREAMS calculates the pesticide available for extraction to stormflow, the amount of pesticide available is reduced by the vertical movement of pesticide out of the surface zone in the percolate. The amount removed in this way is mainly a function of K_d. Thus, herbicides with low partitioning ratios, such as picloram ($K_d = 0.7$) and dicamba ($K_d = 0.077$), will not be available in the surface zone for extraction to stormflow.

One of the limitations of CREAMS simulation of herbicide movement is that it assumes that water which infiltrates is immediately unavailable for stormflow. Dicamba and picloram are readily leached out of the surface zone and may be immediately unavailable, but it is likely that they will be picked up by interflow and return to the soil surface lower in the watershed. This subsurface movement of picloram was observed by Neary, *et al.* (1979), when picloram was applied to deep-soil, ridge-top areas of mountain watersheds in western North Carolina. At no time during the sampling was there overland flow to ephemeral stream channels. Yet water samples taken from the perennial stream below the application site showed picloram at very low levels for a short duration. Models which simulate subsurface movement of pesticides are needed to predict the impact of highly soluble chemicals such as picloram and dicamba.

All of the herbicides tested are of low toxicity to fish and wildlife. There were no simulated stormflow events which had a mean herbicide concentration close to levels that would be lethal to fish or wildlife. The closest was bromacil with a predicted high concentration of 1.8 ppm (Figure 5). Since bromacil has a 48-hour LC_{50} of 71 ppm for bluegill sunfish [(*Lepomis macrochirus* (Rafinesque)] (Mullison, *et al.*, 1979), the concentration must be increased by more than 35 times over that predicted before the LC_{50} is reached.

In conclusion, comparisons of predicted hexazinone concentrations in stormflow from forested watersheds with field data show that CREAMS, an agricultural runoff model, can be used to predict with reasonable success the herbicide concentrations in stormflow occurring shortly after application. From the standpoint of environmental impact, maximum stormflow residue concentrations occur shortly after application and CREAMS estimated those concentrations within ± 15 percent. This study confirms the conclusions of Lorber and Mulkey (1982) that the current version of CREAMS does not account for subsurface movement and thus tends to underpredict concentrations which may be influenced by subsurface interflow.

CREAMS is useful in evaluating alternative forest herbicides for their potential to appear in stormflow. The model predicts the following order of potential for appearance in stormflow: bromacil > triclopyr > hexazinone > picloram > dicamba. Highly soluble herbicides (picloram and dicamba) with low partitioning ratios (K_d) were not readily lost to stormflow. These compounds move down through the soil profile with infiltrating rainfall and are not available at the soil surface. Subsurface flow models and/or later versions of CREAMS currently under development will likely be more reliable in predicting the movement of picloram and dicamba.

ACKNOWLEDGMENTS

The authors wish to thank the E. I. duPont Nemours & Company, Inc., and the Chattahoochee National Forest, USDA Forest Service, for their assistance in conducting the field research.

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