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WASHINGTON, D.C. 20460

SEP 25 1981

February 23, 1981

OFFICE OF
PESTICIDES AND TOXIC SUBSTANCES

MEMORANDUM

SUBJECT: Trifluralin Protocol

FROM: Chief, Environmental Fate Branch, HED (IS-769)

TO: Director, Special Pesticide Review Division (TS-791)

In response to your note dated February 9, 1981, appropriate testing to satisfy EFB concerns for a field monitoring study on trifluralin are listed below:

1. Under use conditions, apply maximum rates of trifluralin to a corn, sorghum or barely growing area with a slope of about 5-10° into a pond. Testing in two different geographical sites is required, with at least one of the sites having been treated with trifluralin the previous year. (The treated area is to be at least 60% of the drainage basin).
2. Sample pond water and treated soil immediately before application, immediately after application and, until the first rainfall of 0.25 inches, weekly thereafter. After rainfall of 0.25 inches or more has accumulated, sample treated soil, water, suspended sediment, aquatic organisms and bottom sediment according to the following schedule: one day, one week, two weeks, one month and monthly thereafter for a minimum of six months. If patterns of residue buildup and decline are not determinable from these samples, monthly sampling is to be continued.
3. Analysis is to be for parent compound, degradation products and total residues. The aquatic organisms to be sampled and analyzed are to be determined by EEB.
4. After the experimental sites have been chosen and before testing is initiated, the registrant is to contact EPA to specify testing details as demanded by the uniqueness of the testing sites.
5. Daily monitor the temperature, cloud cover, pan evaporation, percent runoff per rainfall event and provide a profile of the soil in the treated areas.

Attached is an article addressing trifluralin runoff without addressing resultant concentrations in receiving ponds.

David J. Severn

Attachment

cc: Director, HED
C. Bushong, EEB
W. Garner, EFB

Handwritten notes and signatures:
Rec'd
HED
1/24/81

Herbicide Runoff from Upland Piedmont Watersheds—Data and Implications for Modeling Pesticide Transport¹

R. A. LEONARD, G. W. LANGDALE, AND W. G. FLEMING²

ABSTRACT

Runoff and persistence of selected herbicides were studied on four small Piedmont watersheds in Georgia during four growing seasons. This is part of a study designed to provide data for developing and testing mathematical models for agricultural chemical transport. Seasonal runoff losses were determined relative to watershed management, herbicide type and persistence, mode of application, and time of runoff in relation to application timing. Seasonal losses were usually <2% of the application, unless large runoff volumes were generated shortly after application. Average storm herbicide concentrations in runoff were correlated with herbicide concentrations at the 0- to 1-cm depth increment of the watershed soils at the time of runoff. Paraquat concentrations in runoff (predominantly sediment associated) were well correlated and positive with the product of soil herbicide concentration and sediment in runoff. Equations describing soil-based herbicide transfer to runoff were power functions with exponents near unity with the form: $Y = a x^b$. Simple relationships such as those developed in this study along with hydrology and erosion/sediment models may be useful in predicting pesticide runoff potential when assessing relative impacts of management decisions. The next step would be to describe key management practices in terms of these coefficients and exponents.

Additional Index Words: agricultural chemicals, atrazine, diphenamid, cyanazine, propazine, paraquat, trifluralin, water quality.

Pesticides are often cited as hazards to water quality, as a result of direct runoff from treated farmland (5). Several recent reviews (4, 15, 21) summarized available pesticide runoff data, and several models have been developed to describe and predict pesticide runoff (1, 2, 6, 7, 8, 11) to better assess the problem and to examine management alternatives so as to limit potential pesticide pollution. This study has provided data in most of the efforts cited above. The specific objectives of this report were to summarize these data and present simplified relationships between concentrations of pesticide in soil and measured concentrations in runoff.

MATERIALS AND METHODS

Four single-field drainage areas instrumented for this study were watersheds designated P1, P2, P3, and P4. These areas are described in Table 1. Their instrumentation and general study procedures are described in detail elsewhere (18). No soil and water conservation structures were present on watersheds P1 and P2. Watersheds P3 and P4 were portions of a parallel-terraced area with grassed outlet channels serving to collect runoff.

Crops planted and herbicides used in the study are shown in Table 2. When possible we selected commonly used herbicides and applied

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them as recommended for crop production. However, compromises had to be made between this need and the need to use compounds that could be easily and accurately measured in the laboratory, provide a predictable mode of transport, and persist sufficiently during the growing season to allow several runoff measurements. We also wanted a compound that was "fixed" or strongly adsorbed to soil and sediment for purposes of tracing sediment transport and better define detachment and transport from the soil surface. Paraquat was selected as this tracer, but its application as a post-plant spray to the soil surface was not for its herbicidal value or to represent normal farming operations. Trifluralin and diphenamid are not normally applied together as in this study; however, this combination produced no deleterious effects on crop growth.

The watersheds were conventionally tilled each year before planting except for P1 in 1975. In 1975, this watershed was no-till planted in small grain residue. Crops on the watersheds were not cultivated after planting to eliminate this source of herbicide redistribution at the soil surface, except for P3 and P4 in 1975 which required one cultivation because of severe weed infestation. In addition to the crops shown in Table 2, a small-grain cover crop was planted each fall on P3 and P4. Watershed soils were characterized according to their particle size distribution, specific surface area, and carbon content. Herbicide residues in watershed soils were determined at various depths and times after application. Runoff samples were analyzed for sediment concentration and composition, herbicide attached to sediment, and herbicide dissolved in water. Details of these analyses have been previously published (18). Rainfall, runoff, and sediment yield records were maintained continuously throughout the study period (1 July 1972 to 1 Oct. 1975). Because of the persistence of the compounds used, herbicide analyses were seasonal, usually during periods not exceeding 90 days after application. Runoff sampling devices employed provided 5 to 15 individual samples for each major runoff event, each sample representing a segment of the storm. Total storm loads were computed by integration of concentrations and discharge. Average storm concentration in water plus sediment, as used in relationships developed for this report, were defined as the total herbicide in both sediment and water for a storm divided by total storm discharge volume.

RESULTS AND DISCUSSION

Rainfall, runoff, and sediment yield from the experimental watersheds during 90-day periods after pesticide application each year are summarized in Table 3. During the study, seasonal rainfall and sediment yield were highly variable, with yields depending on rainfall amounts, intensity, and proximity of intense rainstorms to planting date when the soil was freshly tilled and unprotected by crop canopy. Most of the total annual soil loss occurred during these periods because of intense

Table 1—Characteristics of experimental watersheds.

Watershed	Area ha	Average slope	Maximum slope	Predominant soils†
		%		
P1	2.7	3	8	Cecil sandy loam Pacolet gravelly sandy loam
P2	1.3	4	10	Cecil sandy loam
P3	1.3	3	3	Cecil sandy loam
P4	1.4	3	4	Cecil sandy loam

† Typic Hapludults, clayey, kaolinitic, thermic family.

Table 2—Crops and herbicides used during study.

Watershed	Years	Summer crop	Herbicide applied
P1, P3	1972, 1973, 1974	Soybeans (<i>Glycine max</i> [L.] Merr.)	Diphenamid (<i>N,N</i> -dimethyl-2,2-diphenylacetamide)
			Paraquat (1,1'-dimethyl-4,4'-bipyridinium ion)
			Trifluralin (α,α,α -trifluoro-2,6-dinitro- <i>N,N</i> -dipropyl- <i>p</i> -toluidine)
P2, P4	1975	Grain Sorghum (<i>Sorghum vulgare</i> Pers.)	Paraquat
			Propazine [2-chloro-4,6-bis(isopropylamino)- <i>s</i> -triazine]
P2, P4	1973, 1974	Corn (<i>Zea mays</i> L.)	Atrazine [2-chloro-4-(ethylamino)-6-(isopropylamino)- <i>s</i> -triazine]
			Paraquat
P2, P4	1975	Corn	atrazine
			Cyanazine (2-[[4-chloro-6-(ethylamino)- <i>s</i> -triazine-2-yl]amino]-2-methylpropionitrile)
P3	1975	Soybeans	Diphenamid, paraquat, trifluralin

storms and freshly tilled soil (14). Watersheds P3 and P4, which were managed with conservation practices, yielded considerably less sediment than P1 and P2. However, we could not attribute all this difference in sediment yield to the presence of terraces and waterways, since all four watersheds differed somewhat in slopes, soils, and rainfall distribution (Table 1). Sediment yield was lowest on P1 in 1975, when management was changed to no-till planting. However, paraquat loss in terms of concentration on sediment was highest among all treatments, but this system greatly reduced the significance of soil erosion as a transport process (14).

Herbicide runoff, measured during the study, is summarized in Table 4. Except for paraquat, total seasonal herbicide runoff losses were usually less than 2% of that applied. The large amount of diphenamid (7.2%) in runoff from watershed P1 in 1973 was caused by an intense thunderstorm about 4 hours after herbicide application. About 95% of the total seasonal loss occurred in this one storm. Loss of propazine from P1 in 1975 was relatively high, even though this watershed was in no-till management. This high value (6.7%) occurred 9 days after herbicide was applied when the watershed received a 10-year frequency storm and 3.8 cm of the total 4.7 cm rainfall occurred during the initial 20 min of the 1-hour-long storm.

Paraquat losses during the first year on each watershed were also higher than losses of the other herbicides, since paraquat does not degrade rapidly and strong adsorption prevented leaching; it was immobilized near the soil surface in the active soil erosion zone throughout the season. Since paraquat was used in this study as a tracer only, the runoff losses should not be extrapolated to reflect expected losses under normal application. Trifluralin losses reflected the effects of incorporating the herbicide and reducing the concentration at the soil surface that is susceptible to runoff. Results of this study agreed with those of Wauchope (21) and others (4, 15) that pesticide runoff potential is determined primarily by placement and persistence of the material in the runoff zone in relation to rainfall occurrence.

Table 3—Rainfall, runoff, and sediment yield totals for a 90-day period after herbicide application.

Watershed	Year	Seasonal rainfall cm	Runoff events	Runoff cm	Seasonal sediment yield kg/ha
P1	1972	14.9	6	2.8	3,630
	1973	19.5	9	5.3	9,900
	1974	40.6	10	17.5	11,200
P2	1975	25.7	3	2.6	17
	1973	35.9	11	13.3	11,200
	1974	38.8	7*	9.6	1,750
P3	1975	27.2	7	7.9	5,100
	1972	17.8	10	5.3	1,970
	1973	20.3	5	5.7	1,540
P4	1974	35.4	6	4.6	1,560
	1975	24.4	3	1.0	250
	1973	31.9	9	11.1	3,750
	1974	33.6	4	5.0	410
	1975	27.2	3	2.4	620

Except for paraquat, herbicides in this study were transported primarily in solution rather than adsorbed to sediment. However, concentrations for most pesticides, except for diphenamid in some cases, were highest in the sediment phase by 2 to 15 times that in the water phase. Table 4 shows calculated percentage of the total seasonal losses found in solution at the time of laboratory separation of water and sediment. In other studies (17, 23), sediment transport of trifluralin appeared more important. We do not know whether this apparent discrepancy reflects differences in soils and study conditions or differences in technique. We had other problems interpreting trifluralin data of this study, which we will discuss later.

Mode of transport is an important consideration when designing management strategies to limit movement of chemicals from cropland (19). Paraquat losses were much lower from P3 and P4 as compared with P1 and P2, and in proportion to sediment yields. Losses of other herbicides were, however, more related to runoff volume.

The potential for herbicide runoff was strongly affected by application timing in relation to a runoff-producing rainstorm. Figure 1 illustrates the relationship between herbicide concentrations in runoff and time after herbicide application in 1973. Concentration in runoff generally decreased exponentially with time. The decrease in runoff concentrations for herbicides, other than paraquat, primarily reflected the decrease in herbicide present with time in the runoff-active zone at the soil surface. The decrease in paraquat also reflects the decreased sediment production with time as the surface soil stabilized and became better protected by plant canopy.

Figure 2 shows the herbicide concentrations at the 0- to 1-cm-depth increment of the watershed soils of Fig. 1. Vertical bars show the magnitude of the standard deviation from mean of several different samples analyzed from each watershed on a given date. We arbitrarily chose the 0- to 1-cm-depth increment to represent the depth affected by the runoff process, since this effective depth varies with rainfall, soil, and ratio of rill to interrill erosion. The ratio of rill to interrill erosion is thought to affect chemical transport (2, 11). For interrill erosion, this effective depth may be very shal-

Table 4—Summary of herbicide measured in runoff during 90-day period following application.

Watershed	Date of application	Application rate kg/ha	Days to first runoff event	Total % in runoff	Concentration ranges, averages for individual storms		% in solution phase
					In solution µg/liter	In sediment mg/kg	
<u>Atrazine</u>							
P2	11 May 1973	3.36	8	1.9	2 - 200	0.3 - 3.2	86
	29 Apr. 1974	3.81	6	0.2	0 - 1,900	0 - 4.1	88
	21 May 1975	1.54	10	0.7	0.2 - 100	0 - 1.5	83
P4	11 May 1973	3.36	12	0.8	1 - 160	0.06 - 0.6	95
	29 Apr. 1974	4.03	24	0.2	0.2 - 330	0 - 0.6	99
	14 May 1975	1.45	17	0.3	5 - 35	0.2 - 0.6	95
<u>Cyanazine</u>							
P2	21 May 1975	1.61	10	1.0	0 - 150	0 - 2.3	87
P4	14 May 1975	1.35	17	0.07	2 - 12	0.1 - 0.2	58
<u>Diphenamid</u>							
P1	1 July 1972	3.36	27	0.9	2 - 180	0 - 2	92
	13 June 1973	3.36	<1	7.2	0 - 1,600	0 - 0.7	98
	30 May 1974	3.52	22	0.3	0 - 210	0 - 0.9	94
P3	30 June 1972	3.36	2	1.7	2 - 2,070	0 - 1.7	96
	15 June 1973	3.36	23	0.6	0.3 - 61	0 - 0.6	96
	30 May 1974	3.16	28	0.1	0 - 21	0 - 0.6	92
	28 May 1975	2.31	14	0.3	40 - 73	0.7 - 0.8	96
<u>Paraquat</u>							
P1	1 July 1972	15.3	27	4.8	0 - 1	23 - 230	0
	13 June 1973	1.53	<1	†	0	21 - 62	0
	30 May 1974	2.12	22	†	0	25 - 79	0
	2 June 1975	1.66	9	†	0	130 - 980	0
P2	11 May 1973	1.53	8	10.9	0	4.5 - 90	0
	29 Apr. 1974	2.45	6	†	0	0.5 - 35	0
	21 May 1975	1.93	10	†	0	20 - 72	0
P3	30 June 1972	15.3	2	3.4	0 - 580	110 - 810	0
	15 June 1973	1.53	23	†	0	4.5 - 84	0
	30 May 1974	1.94	28	†	0	10 - 28	0
	28 May 1975	1.84	14	†	0	61 - 70	0
P4	11 May 1973	1.53	12	4.0	0	12 - 34	0
	29 Apr. 1974	1.93	24	†	0	40 - 50	0
	14 May 1975	1.75	17	†	0	9 - 29	0
<u>Propazine</u>							
P1	2 June 1975	1.66	9	6.7	16 - 400	0 - 0.9	>99
<u>Trifluralin</u>							
P1	1 July 1972	1.12	27	0.1	0 - 6	0 - 0.1	89
	13 June 1973	1.12	<1	0.3	0 - 13	0.01 - 0.06	91
P3	30 June 1972	1.12	2	0.2	0 - 21	0 - 0.28	89
	15 June 1973	1.12	23	0.2	0 - 8	0 - 0.09	95

† Percent paraquat in runoff not computed because of residue from previous year present at application.

low and approximate the depth of splash and mixing by raindrop impact, whereas soil extracted from rills may be several centimeters deep. However, the 0- to 1-cm increment reasonably approximates the active zone or at least serves as a useful reference for the purpose of this paper. The nonlinearity of the curves in Fig. 2, particularly those for atrazine and diphenamid, shows that a simple exponential equation would not adequately describe persistence in the 0- to 1-cm increment throughout the season. The atrazine curve showed a distinct break in persistence associated with the first rainfall event at 12 days after application. The curve is displaced as a result of some downward movement out of this zone (Fig. 3). For the remainder of the season, atrazine is more persistent than before rainfall. Herbicides surface-applied to freshly tilled soils are subjected to environmental extremes (3) that can enhance chemical degradation and volatilization. After rainfall with surface

mixing caused by raindrop splash and the dissolved herbicide infiltrating soil with water, biological decay processes can then dominate. Two separate decay equations have been used in model development using these data (7). Beginning with the first runoff event, herbicide concentrations in runoff seemed to decrease in a more nearly uniform exponential manner with time (Fig. 1) than did that measured in the 0- to 1-cm surface increment. This may have been caused by less persistence in the immediate soil surface subjected to runoff than that measured even in the 0- to 1-cm zone. Wauchope (21) cited several references which indicated that bulk soil persistence values overestimated apparent surface persistence. In our study, trifluralin decreased in runoff more rapidly (Fig. 1) than it apparently did in soil (Fig. 2). Harper et al. (12) and White et al. (22) studied trifluralin volatilization from this same watershed and, based on their studies, volatilization would be expected

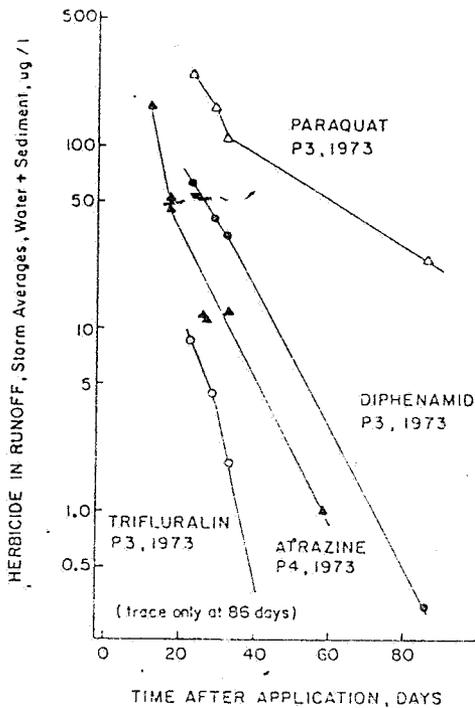


Fig. 1—Herbicide runoff concentrations in relation to application time.

to reduce the immediate surface concentration significantly.

We constructed graphs similar to those in Fig. 2 for each herbicide/watershed/year and the concentrations in the 0- to 1-cm soil zone were estimated by interpolating values for times corresponding to runoff events. For paraquat, volumetric concentrations in runoff correlated with the product of estimated soil concentration in the 0- to 1-cm zone and concentration of sediment in runoff, as shown in Fig. 4. Runoff concentrations used were storm averages, rather than individual sample concentrations. The actual plot is the log transform of the

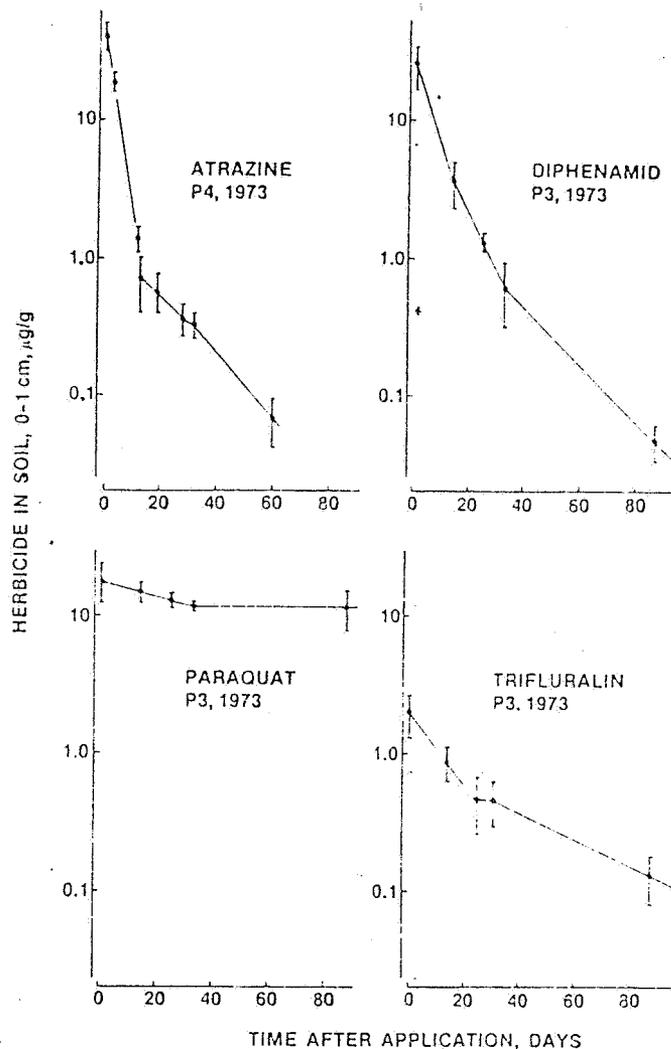


Fig. 2—Persistence of herbicides in 0- to 1-cm zone of watershed soils.

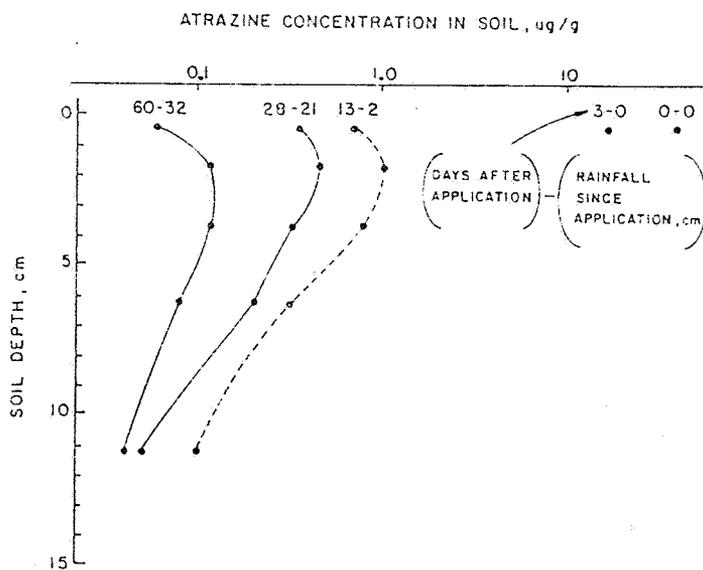


Fig. 3—Atrazine concentration profiles in soil as related to time after application and rainfall. Watershed P4, 1973.

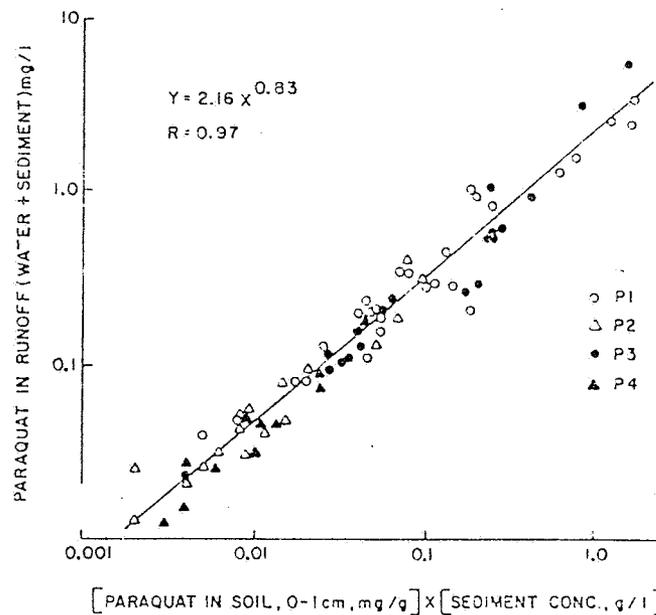


Fig. 4—Relationship between paraquat in runoff and paraquat in soil and concentrations of sediment in runoff.

Enrichment of silt, clay, organic matter, and specific surface in sediment from watersheds.

Watershed	Storm dates	Ratio of sediment composition to soil composition			
		Clay	Silt	Organic matter	Specific surface
P1	28 July 1972	2.6	3.9	2.7	2.7
	10 Aug. 1972	2.8	3.6	2.7	2.5
	13 June 1973	2.4	4.3	2.6	3.4
	30 July 1973	2.8	3.3	2.2	2.9
P2	28 May 1973	2.5	3.7	2.1	3.4
	16 June 1973	2.5	3.7	--	3.0
	21 June 1973	2.1	4.0	--	3.3
P3	31 July 1972	2.3	3.3	2.6	2.0
	4 Sept. 1972	2.4	3.2	2.4	2.0
	8 July 1973	1.9	3.4	2.3	2.8
P4	28 May 1973	1.6	4.1	1.9	3.0
	8 July 1973	1.4	4.4	--	2.7

power function found to best fit the data. Paraquat data for all significant runoff events representing all watersheds and years were fitted to this power function. The correlation was not improved by individual grouping of watersheds or years. The coefficient of about 2 can be identified as an enrichment factor due to preferential removal of fines in runoff. Sediment from selected runoff events on each watershed was characterized as to its particle-size distribution, organic matter content, and specific surface, as measured by N_2 adsorption. We made the same measurements on the watershed soils to allow calculation of enrichment ratios for those fractions important in transport of adsorbed chemicals. These ratios as shown in Table 5 ranged from 2 to 4. Since the soil concentrations in the regression equation were based on an arbitrarily defined runoff zone depth, the correspondence between the two independent measurements of enrichment was remarkably close. With an exponent of 1, the equation in Fig. 4 would reduce to a linear function. However, the fitted exponent value is 0.83. Holt et al. (13) and Duffy et al. (9), using a similar relationship between total phosphorus in runoff and total sediment, found exponents <1 , which they interpret to indicate a changing sediment composition with sediment concentration. That is, as the amount of sediment transported increases with event intensity, the percent fines decreases. This may explain why we found an exponent <1 in this study for a sediment-transported material, like paraquat. Also, since paraquat was immobile and remained in the immediate soil surface, an increasing percentage of rill erosion as compared with interrill erosion with increasing total sediment would remove increasing amounts of soil that would contain less paraquat.

Figure 5 shows the relationships between herbicide concentrations in runoff and that in surface soils (0 to 1 cm) for all water-transported herbicides, except trifluralin. Like paraquat, a power function plotted in linear form gave the best fit. Grouping of herbicides, watersheds, and years did not improve the correlation. Correlation values for regression equations in Fig. 4 and 5 were highly significant and similar in magnitude. Considerable uncertainty is present in the estimation of surface herbicide concentrations (Fig. 2). Therefore, we

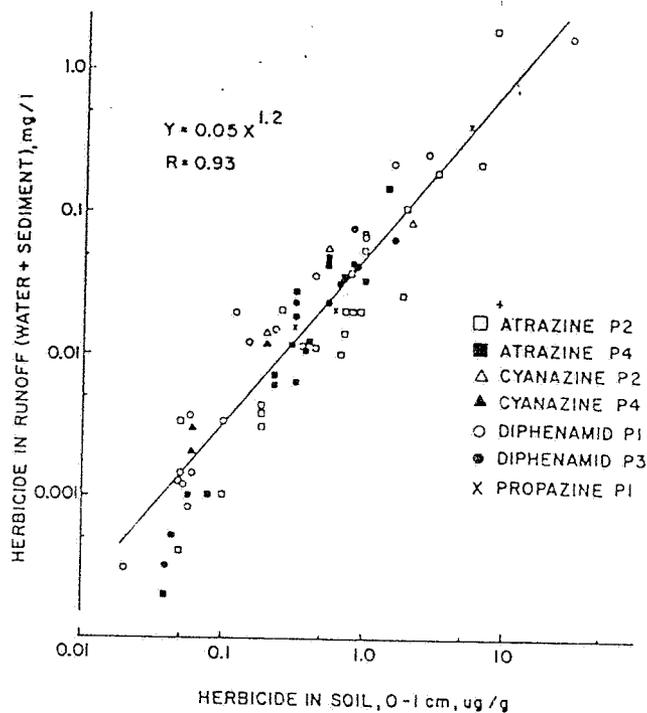


Fig. 5—Relationship between concentrations of water-transported herbicides in runoff and concentrations in the soil surface.

could not separate errors associated with partitioning the herbicide present in runoff from potential errors from estimating the surface concentrations at the time of storm event. No method was available to estimate the effect of downward movement of dissolved herbicides during rainstorms. Except for a limited number of events near the application date, herbicide concentrations of individual samples taken within storm events did not decrease with time after runoff started, as would be expected when surface concentrations are rapidly changing. Within-event variation was mostly random with occasional two- to threefold differences between samples. These differences may reflect spatially nonuniform runoff volumes and source concentrations of herbicide within the watershed. Therefore, the simple correlation in Fig. 5 is good, considering all sources of variation. The coefficient of 0.05 may be considered an "extraction coefficient" reflecting the approximate herbicide distribution between runoff and the soil surface zone. The nonlinearity of the relationship, as indicated by an exponent significantly >1 at 98% confidence limits, may reflect changing partitioning with time of herbicide contact with the soil. That is, extraction efficiency may have been higher soon after application when surface concentrations were also high. This apparent nonlinearity may also reflect an inadequacy in using the 0- to 1-cm zone as a reference throughout the growing season. As time progressed, the actual surface concentration exposed to runoff may have been overestimated.

We did not include the trifluralin runoff date in Fig. 5, since this relationship would overpredict actual observed concentrations. We do not know whether this indicated that trifluralin was distributed to runoff differently, or whether volatilization removed trifluralin from

the immediate soil surface in a manner not reflected by sampling the 0- to 1-cm soil depth increment. Conceivably, some loss of trifluralin during sample processing could have occurred. Only in 1973 were both trifluralin soil surface concentrations and runoff concentrations measured throughout the entire season. This limited data base precluded a separate statistical analysis.

The propazine data was even more limited with only three runoff events measured. However, these data plotted well within the relationship in Fig. 5, even though the P1 watershed in 1975 was in no-till management.

IMPLICATIONS FOR MODELING PESTICIDE RUNOFF

Ideally models contain mathematical structures that represent concepts based on our knowledge of system behavior. As research tools, they may be extremely complex and useful in revealing gaps in knowledge and mathematical representation by how well they simulate measured values. Models that are useful for real-world predictions in terms of relative effects of management options and probabilities of a certain event occurrence may require compromises so that the model is practical in terms of operating cost, simple for a user to understand, and easily applied to situations with limited input data. Many of the present models applied to pesticide runoff are complex and require a large amount of input data. Although these models perform adequately for their intended purpose, there is still a need for simple, user-oriented models. No model to date has adequately predicted solution-transported materials in runoff. This study has shown that simple relationships may be developed between source and runoff concentrations for both solution- and sediment-transported pesticides. Wauchope (21) concluded that pesticides can be classified as to their major transport mode based on solubility. This classification may be improved using methods of Pionke (16), whereby adsorption and soil and sediment properties and sediment/water ratios can also be considered. We are not suggesting that the exact relationships described in this study be extrapolated to different watersheds and pesticides without additional analysis. However, many studies have been performed on pesticide runoff. Perhaps much of this published data could be composited to provide estimates of critical parameters, like extraction coefficients, decay constants, and enrichment factors applicable to a wider range of conditions. This empirical approach to partitioning pesticides could be incorporated into simple surface hydrology and erosion models like that suggested by Frere (10) to provide estimated pesticide concentrations in runoff. Evaluation of the anticipated confidence ranges of this method would, however, be required. Model outputs cannot be more accurate than model inputs. One critical area of uncertainty is the source term of concentration and distribution of pesticide on the watershed or field at the time of runoff. This study and others (20) have shown that even on research watersheds considerable uncertainty and difficulties are encountered in sampling and measuring pesticides in field soils. With this perspective and the known uncer-

tainties associated with quantifying the source term for a farmer's field, a simple, perhaps empirical, approach to distributing pesticide to runoff may be as reliable presently as other more sophisticated approaches, and allow model applications to problems where little input data are available.

ACKNOWLEDGMENT

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