Organic Compounds in the Environment

Fluometuron and Pendimethalin Runoff from Strip and Conventionally Tilled Cotton
in the Southern Atlantic Coastal Plain

Thomas L. Potter,* Clint C. Truman, David D. Bosch, and Craig Bednarz

ABSTRACT

In the Atlantic Coastal Plain region of southern Georgia (USA), cotton (Gossypium hirsutum L.) acreage increased threefold in the past decade. To more effectively protect water quality in the region, best management practices are needed that reduce pesticide runoff from fields in cotton production. This study compared runoff of two herbicides, fluometuron [N,N-dimethyl-N-(3-trifluoromethylphenyl)-urea] and pendimethalin [N-(1-ethylpropyl)-3,4-dimethyl-2,6-dinitro-benzenamine], from plots in strip-tillage (ST) and conventional-tillage (CT) management near Tifton, GA. Rainfall simulations were conducted one day after preemergence herbicide applications to 0.0006-ha plots and runoff from 0.15-ha plots due to natural rainfall following preemergence pendimethalin and fluometuron and postemergence fluometuron use was monitored. Pendimethalin runoff was greater under CT than ST due to strong pendimethalin soil sorption and higher erosion and runoff under CT. The highest losses, 1.3% of applied in CT and 0.22% of applied in ST, were observed during rainfall simulations conducted 1 DAT. Fluometuron runoff from natural rainfall was substantially lower from ST than from CT plots but the trend was reversed in rainfall simulations. In all studies, fluometuron runoff was also relatively low (<1% of applied), and on plots under natural rainfall, desmethylfluometuron (DMF) represented about 50% of total fluometuron runoff. Fluometuron’s relatively low runoff rate appeared linked to its rapid leaching, and high DMF detection rates in runoff support DMF inclusion in fluometuron risk assessments. Results showed that ST has the potential to reduce runoff of both herbicides, but fluometuron leaching may be a ground water quality concern.

Surveys conducted in the 1990s revealed that pesticide residues are widely distributed in streams and rivers throughout the continental United States (USGS, 1999). In watersheds in southeastern states where cotton cultivation is widespread, two frequently detected compounds were the herbicide fluometuron and its degradation, DMF (Coupe et al., 1998; Thurman et al., 2000). Residues of pendimethalin, often applied in tank mixtures with fluometuron, were detected in stream samples collected in the Mississippi delta (Zimmerman et al., 2000). Cotton is intensively produced in the region (USDA, 2003a). In 2002, estimated U.S. cotton acreage treated with fluometuron was 10% and pendimethalin 16% (USDA, 2003a).

Comparison of reported levels of these compounds in aquatic environments with human health guidelines and harm to aquatic life thresholds does not indicate large human and/or ecological risks (USEPA, 1997). However, available monitoring data do not provide the basis for comprehensive exposure assessments for the wide range of conditions under which cotton is grown. Pendimethalin is highly toxic to fish and certain invertebrates; thus, negative effects may occur should runoff or drift contribute residues to surface waters (USEPA, 1997). As with many pesticides, there are uncertainties about the concentrations of fluometuron and pendimethalin that cause negative effects. Thus, more widespread implementation of best management practices (BMPs) that reduce surface water loading of these herbicides is needed.

Reduced or no-tillage management can be a highly effective best management practice for pesticide runoff control (Fawcett et al., 1994). In the Tennessee Valley region of Alabama, pendimethalin runoff was consistently higher from conventionally tilled (CT) cotton than from reduced till with cover crop and no-till plots. Edge-of-field losses were also uniformly low (<0.3% of applied) from all tillage treatments (Yoo et al., 1989).

Reduced fluometuron runoff under conservation tillage was indicated in a laboratory investigation (Reddy et al., 1994). Results of a field study were mixed (Baughman et al., 2001). Simulated rainfall was applied to 0.009-ha plots on Brooksville silt clay (fine, smectitic, thermic Aquic Hapluderts) soil in the Black Belt region of eastern Mississippi 1 to 2 days after treatment (DAT) and runoff was collected following natural rainfall events. Conventional-tillage plots yielded more fluometuron runoff in the first year and fluometuron runoff from no-tillage plots was higher in the second year. In both years, fluometuron runoff was uniformly high (3.2–9.9% of applied) regardless of tillage treatment. On lighter-textured soils and with lower rainfall intensity, fluometuron runoff was <1% of applied (Wiese et al., 1980; Reddy et al., 1994).

Differences in pendimethalin and fluometuron runoff and responses to tillage are anticipated given fluometuron’s much greater water solubility and lower soil organic carbon–water partition coefficient ($K_{oc}$). Fluometuron’s water solubility is 110 mg L$^{-1}$ and its $K_{oc}$ is 100 mL g$^{-1}$. Corresponding pendimethalin values are 0.275 mg L$^{-1}$.

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Abbreviations: CT, conventional tillage; DAT, days after treatment; DMF, desmethylfluometuron; MDL, method detection limit; ST, strip tillage; TFA, trifluoromethylaniline.
and 5000 mL g⁻¹ (Hornsby et al., 1995). Because larger $K_{oc}$ indicates greater soil sorption, conservation tillage practices that lower erosion rates will probably contribute to substantial reductions in pendimethalin runoff. Effects on fluometuron are less certain. Fluometuron sorption to soil is relatively weak (Gaston and Locke, 1995; Willan et al., 1997) and high leaching rates have been observed. Essington et al. (1995) reported that 5 to 53% of fluometuron applied to surface soil was moved through the soil profile by natural rainfall at a field site in western Tennessee. Their study did not demonstrate any trends when no-till and conventional-tillage treatments were compared.

Available data indicate that pendimethalin runoff will be reduced under conservation tillage and reductions in fluometuron runoff are likely. Whether these observations can be generalized is unknown. To our knowledge, published investigations that evaluated tillage effects on runoff of these herbicides in cotton were limited to a single laboratory (Reddy et al., 1994) and two field investigations (Yoo et al., 1989; Baughman et al., 2001). In addition, no investigations were identified in which both compounds were evaluated simultaneously and/or where factors such as soil type, rainfall intensity, and timing with respect to application and tillage were controlled so that runoff of the two compounds could be quantitatively compared.

Our study was motivated by a lack of data to make quantitative assessments of fluometuron and pendimethalin runoff as a function of tillage for Atlantic Coastal Plain conditions. In the southernmost portion of the region, land in cotton production nearly tripled in the past decade (Georgia Agricultural Statistic Service, 2003). The rapid increase in cotton acreage has resulted in increases in agrochemical use and a need to rigorously evaluate land management practices that have potential to reduce and/or eliminate runoff. The current study evaluated pendimethalin and fluometuron runoff from cotton on Tifton loamy sand (fine-loamy, kaolinitic, thermic Plinthic Kandiudults) soil under CT and ST management. The Tifton soil series is one of the most important agriculturally (USDA, 2003b) and ST is a widely used conservation practice in the region (Brown et al., 2003).

MATERIALS AND METHODS

Site Description

Studies were conducted in Tift County, Georgia (31°26′N, 83°35′W), on a 1.8-ha parcel divided into six 0.15-ha and one 0.4-ha plots (Fig. 1). Three 0.15-ha plots were in CT and three in ST. The 0.4-ha plot at the top of the slope where rainfall simulations were conducted was divided equally between ST and CT. The soil is Tifton loamy sand with 3 to 4% slope (Bosch et al., 2000). The sand, silt, clay, and organic carbon content of surface soil samples (0–15 cm) collected before planting the first cotton crop was 85.6 ± 32 g kg⁻¹ sand, 32 ± 2 g kg⁻¹ clay, and 5.1 ± 0.5 g kg⁻¹ organic C, and the median pH was 6.5. Each 0.15-ha plot was surrounded by a 0.6-m-high earthen berm, which directed surface runoff to metal H-flumes at downspout plot corners. Pressure transducers (Druck, New Fairfield, CT) in the flume throats were calibrated to record runoff hydrographs. H-flume dimensions, the minimum depth required for a transducer signal, and data recording intervals indicated that the minimum detectable volume per runoff event was 2.7 L. Rainfall was monitored using a tipping bucket rain gauge.

Runoff Sample Collection

Flow-proportional composite runoff samples from each plot were collected during each event directly into 9-L glass jars using IsoC (Lincoln, NE) autosamplers. The samplers were programmed to withdraw 50 mL for every 520 L that passed the flume. A minimum of 100 mL was necessary for pesticide analysis, thus 1040 L runoff was required for an analyzable sample. This was equivalent to 0.7 mm of runoff. The upper bound on the sample collection system (events that would overfill the bottle) was 63 mm of runoff. In 2000, CT plots yielded samples for 45% and ST, 46% of runoff events. The combined flow of sampled events equaled 94 and 93% of the total runoff volume for all runoff events for CT and ST plots, respectively. In 2001, CT plots yielded samples during 44% of events and ST, 15%. The combined flow of these samples was CT, 84%, and ST, 44% of total runoff. No runoff events exceeded the total volume of the sample collection jars.

Crop and Soil Management

In 2000 and 2001, cotton was planted in rows 91 cm on center in the first week of May and defoliated and picked in mid-September. About 4 wk before planting, the rye (Secale cereale L.) cover crop (planted after harvest of the previous cotton crop) on all plots was killed with glyphosate (1.1 kg ha⁻¹). One week later poultry manure was broadcast-applied at 4.5 Mg ha⁻¹. Conventional-tillage plots were then tilled and bedded. On ST plots, tillage was restricted to 15-cm-wide strips tilled into the killed cover crop mulch just before planting. Cotton was planted into these strips. Fertility and pesticide management practices for both tillage treatments were identical and followed local grower guidelines (Brown et al., 2003). Strip-tillage lint yields averaged 550 kg ha⁻¹ in 2000 and 1450 kg ha⁻¹ in 2001. Corresponding CT yields were 620 and 1380 kg ha⁻¹. The herbicides, Prowl 3.3 EC (BASF, Research Triangle Park, NC) and Cotoran 4L (Griffin LLC, Valdosta, GA), were tank-mixed and applied with a tractor-mounted boom sprayer 1 h after planting, with the exception of the ST area in the plot where rainfall simulations were conducted. Herbicides were applied there 24 h after planting. This allowed staging of rainfall simulations so that the time between herbicide application and rainfall was the same on CT and ST plots. Application rates of the two active ingredients, fluometuron (Cotoran 4L) and pendimethalin (Prowl 3.3 EC), were measured by analysis of filter paper (7-cm diameter) spray targets ($n = 5$ per plot) that had been staked to the soil surface before spray application. In 2000 the fluometuron rate was 0.08 ± 0.16 kg ha⁻¹ and 1.46 ± 0.38 kg ha⁻¹ in 2001. Pendimethalin rates were 0.69 ± 0.13 kg ha⁻¹ and 0.95 ± 0.25 kg ha⁻¹. The fluometuron degrade, DMF, was detected on spray targets. Amounts translated to application rates of 0.01 ± 0.005 kg ha⁻¹ in 2000 and 0.03 ± 0.01 kg ha⁻¹ in 2001. Desmethylfluometuron was presumably present in the Cotoran 4L; however, no measurements were made to confirm this. Six weeks after planting in 2001, a 1.1 kg ha⁻¹ postemergence (directed) fluometuron application was made. The application rate was not measured.

Rainfall Simulation

In both years, four simulator plots were established within the 0.4-ha plot. There were two plots each in ST and CT...
portions of the plot in each year. Approximate simulator plot locations are marked with a “P” on Fig. 1. Plots were defined with aluminum frames, 2 × 3 m, with the 2-m side centered over two cotton rows. Frames were pushed 5 cm into the soil. Runoff was collected from an aluminum trough installed at the downslope end of each plot. Simulated rainfall was applied with an oscillating nozzle rainfall simulator (Foster et al., 1982) at 50 mm h⁻¹ for 1 h beginning 24 h after herbicide application. The simulator’s 80100 Veejet nozzles (Spraying Systems, Wheaton, IL) produce drops with a median diameter of about 2.3 mm. Water was obtained from a nearby well drilled to a depth of 166 m into the Floridian aquifer. The aquifer, which extends over much of the region, is widely used to support irrigated crop production. Simulated rainfall rates were measured at the sides and upslope end of each plot. Simulated storm size and intensity with respect to herbicide application represented a 4-yr return interval for the region (United States Department of Commerce, 1958). Simulated storm duration on each plot was 60 min. At the beginning of each 5-min interval during simulations, runoff was collected directly into 1-L wide-mouth glass bottles and sealed with Teflon-lined screw caps. These samples were transferred to a laboratory refrigerator within 2 h after collection and reserved for herbicide residue analysis. The remaining runoff during each 5-min interval was collected directly into preweighed 1-L Nalgene bottles (Nalge Nunc International, Rochester, NY). Water volume in each bottle was determined gravimetrically. After acidification (to pH < 2 with 12 M HCL) to flocculate sediments, bottles were allowed to stand at room temperature overnight. The clear supernatant was decanted and bottles were dried overnight at 105°C and reweighed. Total sediment was determined by subtracting bottle tare weights. Antecedent soil water content was determined on samples collected just before simulations in sprayed crop areas adjacent to the plots at three depth intervals of 0 to 2, 2 to 8, and 8 to 15 cm. In both years and tillages soil water content was relatively uniform in the 2- to
Soil and Water Sample Preparation for Herbicide Analysis

Within 48 h of collection, water samples were vacuum-filtered (GFF filters, 0.7-μm nominal pore size; Whatman, Maidstone, UK). Filter and solids were weighed and sediment mass was determined by subtracting filter dry weight and dividing by 0.85. This factor was based on replicated moisture measurements on filtered sediments prepared in a separate laboratory study by suspending Tifton surface soil in distilled-deionized water and filtering with the same apparatus used in this study. Filtered sediment from runoff samples collected during rainfall simulations was extracted by shaking for 1 h with 50 mL of methanol followed by GFF filtration. Runoff sample filtrate was solid-phase-extracted using 6-mL Oasis HLB cartridges (Waters, Milford, MA). They were preconditioned with methanol and distilled-deionized water and eluted with 3 mL of methanol followed by 3 mL of methylene chloride. The combined extract was adjusted to 50 mL with methanol. Soil samples were thawed and sieved (10 mesh stainless steel), and 50-g subsamples were sequentially extracted with methanol (3 × 50 mL). Soil water content was determined separately on subsamples by drying overnight at 105°C. Combined extracts of each sediment, water, and soil sample were reduced to 1, 5, and 10 mL, respectively, by evaporative concentration under a N2 gas stream. Extracts were syringe-filtered using 0.45-μm PTFE membranes and stored at −10°C.

Data Analysis

Microsoft Excel 2000 (Microsoft, 2000) was used for all calculations. Unpaired t test statistics were evaluated at P ≤ 0.05. Fluometuron and pendimethalin runoff estimates in rainfall simulations were obtained by multiplying the average dissolved or sediment-bound concentration for each 5-min sample by the corresponding runoff volume or sediment mass. Average concentrations were derived by linear interpolation between adjacent data points on response curves (chemographs). In computing pendimethalin runoff due to natural rainfall on 0.15-ha plots in both 2000 and 2001, data from all samples collected between the date of application and calendar year end were used. The same approach was used for fluometuron in 2000. Fluometuron runoff associated with the 2001 preemergence application was evaluated using results from all runoff samples collected between the preemergence and postemergence applications.

Extract Analysis

Extracts were fortified (5 μg mL⁻¹) with 2-chlorolepine (internal standard), and analyzed by high-performance liquid chromatography (HPLC) with photodiode array detection using a Hewlett-Packard (Palo Alto, CA) Model 1050 HPLC system. The HPLC was fitted with a 4.6-mm-diameter × 15-cm-length Beckman Ultrasphere ODS column (Alltech, Deerfield, IL). Initial conditions of the acetonitrile (B)–water (A) gradient elution were 60% A to 40% B. This was changed linearly to 50% A to 50% B in 5 min and then to 25% A to 75% B 1 min later. Solvent composition was isocratic for 6 min followed by a decrease to initial conditions in 1 min. The flow rate during elutions was 1 mL min⁻¹. Target compounds in each analysis were pendimethalin, fluometuron, and the fluometuron degradates, DMF and trifluoromethylaniline. Peak assignments were confirmed by high performance liquid chromatography (HPLC)–mass spectrometry (MS) using a ThermoQuest LCQ system (ThermoQuest-Finnegan, San Jose, CA). The soil and sediment method detection limit (MDL) based on instrument response to the lowest calibration standard for all compounds was 0.01 μg g⁻¹. The MDLs for water samples were 0.1 to 1.0 μg L⁻¹. The MDL range corresponded to the range in the volume of samples collected and analyzed (0.1–1.0 L).

Chemicals and Supplies

Analytical standards were purchased from Chem Service (West Chester, PA). Sigma-Aldrich (St. Louis, MO) provided the 2-chlorolepine. Other laboratory chemicals and all supplies were obtained from Fisher Scientific (Hampton, NH). Formulated herbicides were purchased locally.

Quality Control

A methanol solution containing each compound at 100 μg mL⁻¹ was used to prepare matrix spikes. On spray targets, 0.1 mL was applied drop wise to filter paper. The same approach was used to spike filtered sediment prepared by suspending Tifton surface soil (collected from plots before herbicide application) in distilled-deionized water and GFF filtering. Water spikes were prepared by adding 50 μL of spiking solution to well water used for rainfall simulations. Replicate (three) subsamples of 0- to 2-cm soil composites were analyzed. One subsample from each set was fortified with 0.5 mL of spiking solution. Recoveries from spray targets averaged 92 to 110% (n = 6), from sediments 88 to 98% (n = 6), from water and soil 94 to 110% (n = 8). The exception was trifluoromethylaniline recovery from soil (49 ± 15%, n = 8). Fluometuron (0.27–8 μg L⁻¹) was detected in all (four) aqueous field blanks prepared for 2000 rainfall simulations. All other analytes in these samples and all analytes in 2001 field blanks were below the MDL. The fluometuron source in the 2000 blanks was not identified. Based on runoff volumes, the percent fluometuron detected in runoff attributable to fluometuron detected in the 2000 blanks was <2% of the runoff total. This is less than measurement error in several other parameters used in calculations and given its magnitude, no “blank” adjustments were made in runoff data. Field “duplicates” prepared by splitting filtrate from samples collected in the 15-min interval during each simulation into two equal volume aliquots gave repeatability indices (difference between paired results divided by their average) averaging 3.2 to 6.6% (n = 8). The corresponding value obtained from analysis of soil duplicates was 5.3 to 72% (n = 8).
the preemergence and postemergence applications. The ST and CT data sets were treated separately, event-based volume weighted concentrations obtained in rainfall simulations were included, and results < MDL were excluded. The $r^2$ for the regressions of In(concentration) and DAT were ST, 0.782, and CT, 0.792. Adjustments accounted for <5% of CT and 24 to 45% of ST runoff estimates.

A feature of all natural rainfall runoff data sets was that they were highly censored (missing values and/or results < MDL). Although not often identified, we expect high censoring rates are a general feature of data obtained from large plot runoff studies due to sampling constraints and measurements < MDL. In our case, there were many small runoff events (55 to 85% of CT and ST totals), whose volume was less than the minimum required for sample collection. In addition, detection frequencies in samples collected, fluometuron 75 to 93%, DMF 63 to 97%, and pendimethalin 0 to 43%, were highly variable. Combining flow and analytical results, DMF, fluometuron, and pendimethalin censoring rates were 41 to 88, 43 to 88, and 74 to 100% respectively, and highest for ST. Because data sets with severe censoring (>50%) and variable MDLs are problematic with regard to hypothesis testing (Helsel and Hirsch, 2002), we make no statistically based inferences from these data. Estimates of each chemical’s runoff maxima and minima expressed as percent of applied are reported. These estimates were computed by inserting either zero (minimum) or the MDL (maximum) for all <MDL results. In addition, fluometuron concentration estimates in the low volume samples that did not yield samples were calculated using linear regression equations relating DAT and In(concentration). The 1 DAT volume-weighted concentrations from rainfall simulations were included in regression analyses. Values < MDL were not included. The same approach was used for 2001 CT pendimethalin estimates. Regression equation $r^2$ values were 0.679 to 0.827 for fluometuron and 0.834 for pendimethalin. Values for DMF and sediment concentration were assigned by inserting the sample average for other ST or CT plots collected on the same date or if no samples were collected on all plots within the treatment block on a given date, by linear interpolation using data from samples collected immediately before and after. Nearly all pendimethalin ST results and most 2000 CT values were <MDL. In this case, values for small volume samples were assigned by interpolation if results in prior and succeeding samples were >MDL or the MDL, if samples collected before and after were <MDL. Other notations regarding data handling include: (i) trifluoro-methylamine was not detected in any runoff samples, thus computations to determine percent loss as a function of fluometuron application rate were not performed; (ii) sediment herbicide concentrations in runoff samples due to natural rainfall on 0.15-ha plots were estimated using dissolved concentration results, sediment concentrations, and average soil–water partition coefficient ($k_d$) determined using rainfall simulation sample data; and (iii) the postemergence fluometuron application was set equal to its target application rate (1.1 kg ha$^{-1}$).

**RESULTS AND DISCUSSION**

**Rainfall, Runoff, and Erosion**

Compared with long-term regional averages there was a rainfall deficit of 3% in 2000 and 30% in 2001 (Table 1). Drought conditions prevailed in the region during the study (University of Georgia, 2003). In spite of much lower rainfall, runoff, erosion, and lint, yields were greater in 2001 (Table 1). This was due to a twofold increase in the irrigation rate in 2001 and because much of the 2001 rainfall occurred early in the growing season. During May to June 2000, rainfall was 70% less than the long-term regional average for these months while it was 5% greater for the same period in 2001.

In both years total runoff was about 20% of rainfall plus irrigation from CT and 5% from ST plots. Differences between the tillage treatments were significant. Total runoff from CT plots during rainfall events that yielded water quality samples was 83 to 95% of total runoff. The flow associated with samples collected from ST plots in 2001 was considerably less, 44%. In total, CT plots yielded nearly three times more samples than ST plots. These results reflect sample collection challenges in large plot runoff studies, where tillage treatments tend to reduce both the frequency of runoff events and their total volume.

The CT plot erosion rate was also higher than the ST rate; however, no inferences are made regarding erosion differences. This is because the sediment concentration associated with small runoff events that did not yield water quality samples was estimated. The uncertainty is unknown and in the case of the ST plots may be relatively high.

The same overall trends were observed in rainfall simulations. Significantly lower total runoff and runoff peak rates from ST plots were observed (Table 2). While CT plots yielded more sediment than ST plots; the difference was not significant because loss from CT plots was highly variable (75% RSD compared with 17% for ST plots).

Lower runoff and erosion rates have often observed with reduced and/or no-tillage soil management (Seta et al., 1993; Bradford and Huang, 1994; Fawcett et al., 1994; Rhoton et al., 2002). This has been related to increases in soil organic C, which promote increased aggregate stability and/or reductions in surface rainfall energy due to interception by desiccated crop residue. Both promote increased infiltration and lower erosion rates. The relatively modest increases in soil organic C reported for Coastal Plain soils after long periods in

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**Table 1. Rainfall and irrigation and average ± one standard deviation of runoff and erosion from 0.15-ha conventional-tillage (CT) and strip-tillage (ST) plots under natural rainfall from the planting date to the end of the calendar year in 2000 and 2001.**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>CT</th>
<th>ST</th>
<th>CT</th>
<th>ST</th>
</tr>
</thead>
<tbody>
<tr>
<td>Runoff, % rainfall + irrigation</td>
<td>17.9 ± 7.2*</td>
<td>4.8 ± 1.4*</td>
<td>24.0 ± 7.4*</td>
<td>4.6 ± 4.1*</td>
</tr>
<tr>
<td>Sediment, Mg ha$^{-1}$</td>
<td>3.1 ± 0.9</td>
<td>0.8 ± 0.2</td>
<td>6.3 ± 2.0</td>
<td>1.4 ± 1.2</td>
</tr>
<tr>
<td>Rainfall, mm</td>
<td>729</td>
<td>729</td>
<td>526</td>
<td>526</td>
</tr>
<tr>
<td>Irrigation, cm†</td>
<td>10.3</td>
<td>10.3</td>
<td>20.3</td>
<td>20.3</td>
</tr>
</tbody>
</table>

* CT values are significantly greater than corresponding ST values at the 0.05 probability level.
† All irrigation was applied in May to August.
Total pendimethalin, % 1.3*  
Sediment pendimethalin, % 0.73  
Dissolved pendimethalin, % 0.61*  
Dissolved desmethylfluometuron % 0.01  
Total fluometuron, % 0.28  
Sediment fluometuron, % 0.03  
Total DMF, % 0.01  
Dissolved desmethylfluometuron (DMF)*, % 0.01  

Subsequent transport through the soil matrix (Peterson compared with CT. Pendimethalin runoff loss, 0 to 0.2% of the rainfall simulations (Fig. 2). Nearly all pendimethalin detected in runoff collected during simulations bound to sediment. This value was nearly equal for both tillage treatments. Higher CT runoff volume and sediment loss was apparently offset by stronger pendimethalin binding to ST sediment. The sediment–water partition coefficient ($k_d$), defined as the ratio between the sediment and dissolved concentrations, averaged 181 ± 9.4 mL g$^{-1}$ for all CT samples and 262 ± 128 mL g$^{-1}$ for ST samples. The higher ST $k_d$ was probably due to enrichment of ST sediment with organic C. This has been reported in other studies (Schieber and McGregor, 1979; Kingery et al., 2002; Potter et al., 2003).

Another indication of strong pendimethalin soil binding was its distribution in soil collected before and after rainfall simulations (Fig. 2). Nearly all pendimethalin was retained in the top 2 cm. Levels in 2- to 8- and 8- to 15-cm soil samples were uniformly close to the MDL. Data indicate low and approximately equal amounts of pendimethalin leached from CT (5.1%) and ST (3.6%) plots. A recently published study has indicated that downward movement of pendimethalin in soil was facilitated by sorption on colloids and their subsequent transport through the soil matrix (Peterson et al., 2002). This mechanism may have been responsible for the pendimethalin movement that our study suggests.

Reduced tillage (Novak et al., 1996) indicate that reduction in the surface energy of rainfall due to interception by crop residue is probably the most significant factor in reducing runoff and erosion in the region.

### Pendimethalin Runoff

The highest loss (percent of applied) was from CT plots during rainfall simulations conducted 1 DAT (Table 2). Conventional-tillage pendimethalin runoff under simulated rainfall was also greater than the CT fluometuron average. Pendimethalin behavior is explained by its strong binding to sediment and that CT plots yielded more runoff and sediment when compared with ST plots.

In total, about 55% (sediment-bound/total) of the pendimethalin detected in runoff collected during simulations was bound to sediment. This value was nearly equal for both tillage treatments. Higher CT runoff volume and sediment loss was apparently offset by stronger pendimethalin binding to ST sediment. The sediment–water partition coefficient ($k_d$), defined as the ratio between the sediment and dissolved concentrations, averaged 181 ± 9.4 mL g$^{-1}$ for all CT samples and 262 ± 128 mL g$^{-1}$ for ST samples. The higher ST $k_d$ was probably due to enrichment of ST sediment with organic C. This has been reported in other studies (Schieber and McGregor, 1979; Kingery et al., 2002; Potter et al., 2003).

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### Table 2. Average ± one standard deviation of runoff, sediment yield, and percent of applied pendimethalin, fluometuron, and desmethylfluometuron (DMF) detected in runoff following application of 50 mm of simulated rainfall in 1 h to 0.0006-ha conventional-tillage (CT) and strip-tillage (ST) plots at 1 day after treatment (DAT).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>CT</th>
<th>ST</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sediment, g m$^{-2}$</td>
<td>$98 ± 74$</td>
<td>$34 ± 5.9$</td>
</tr>
<tr>
<td>Total runoff, mm</td>
<td>$12^a ± 1.3$</td>
<td>$6.5 ± 1.2$</td>
</tr>
<tr>
<td>Maximum runoff rate, mm h$^{-1}$</td>
<td>$26^a ± 2.3$</td>
<td>$9.5 ± 2.7$</td>
</tr>
<tr>
<td>Dissolved fluometuron, %</td>
<td>$0.25 ± 0.06$</td>
<td>$0.69 ± 0.08$</td>
</tr>
<tr>
<td>Sediment fluometuron, %</td>
<td>$0.03 ± 0.003$</td>
<td>$0.01 ± 0.001$</td>
</tr>
<tr>
<td>Total fluometuron, %</td>
<td>$0.28 ± 0.09$</td>
<td>$0.70 ± 0.086$</td>
</tr>
<tr>
<td>Dissolved desmethylfluometuron (DMF)*, %</td>
<td>$0.01 ± 0.01$</td>
<td>$0.01 ± 0.001$</td>
</tr>
<tr>
<td>Sediment DMF, %</td>
<td>&lt;0.01</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Total DMF, %</td>
<td>$0.01 ± 0.01$</td>
<td>$0.01 ± 0.001$</td>
</tr>
<tr>
<td>Dissolved pendimethalin, %</td>
<td>$0.61^a ± 0.14$</td>
<td>$0.10 ± 0.04$</td>
</tr>
<tr>
<td>Sediment pendimethalin, %</td>
<td>$0.73 ± 0.02$</td>
<td>$0.12 ± 0.03$</td>
</tr>
<tr>
<td>Total pendimethalin, %</td>
<td>$1.3^a ± 0.60$</td>
<td>$0.22 ± 0.07$</td>
</tr>
</tbody>
</table>

---

* Indicates that CT average is significantly greater than corresponding ST average at the 0.05 probability level.

† Fluometuron and DMF runoff are expressed as a percent of total fluometuron and DMF applied.

Natural rainfall on 0.15-ha plots produced greater total CT pendimethalin runoff than ST when both maximum and minimum runoff estimates were compared (Table 3). However, inferences regarding trends in differences were limited by the fact that both CT and ST plots were highly censored. Pendimethalin concentration was >MDL in only 1 of 34 runoff samples collected and analyzed from ST plots during the two years of the study. Pendimethalin concentration was >MDL in 30 of the 95 samples from CT plots. The higher detection frequency contributed to a lower censoring rate and much closer agreement between maximum and minimum CT runoff estimates.

In 2001, pendimethalin runoff from CT plots was about twice that observed in 2000 (Table 3). This is probably due to differences in the number and timing of runoff events with respect to application. The first runoff event was 72 DAT in 2000. On CT plots in 2001, 10 runoff samples were collected between 11 and 70 DAT. Pendimethalin concentration was >MDL in 90% of these samples. Typically there is an inverse exponential relationship between the magnitude of runoff loss of pesticides and DAT (Wauchope, 1978; Southwick et al., 1993). This was the trend observed on CT plots in our study (Fig. 3).

Pendimethalin runoff from natural rainfall on our plots was similar to runoff observed in a study conducted on Decatur silt loam (fine, kaolinite, thermic Rhodic Paleudults) soil in the Tennessee Valley region of Alabama (Yoo et al., 1989). Runoff from cotton plots in both studies was monitored for two years and reduced tillage treatments in all cases yielded less pendimethalin compared with CT. Pendimethalin runoff loss, 0 to 0.2% (our study) and <0.3% (Yoo et al., 1989), was also uniformly low for all tillage treatments in both investiga-
Table 3. Average ± one standard deviation of runoff, erosion, and percent of applied pendimethalin, fluometuron, and desmethylfluometuron (DMF) detected in runoff from natural rainfall on 0.15-ha conventional-tillage (CT) and strip-tillage (ST) plots during 2000 and 2001.

<table>
<thead>
<tr>
<th></th>
<th>2000</th>
<th>2001</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>CT plots</td>
<td>ST plots</td>
</tr>
<tr>
<td>% of applied</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dissolved fluometuron, preemergence</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum†‡</td>
<td>0.20 ± 0.12</td>
<td>0.09 ± 0.02</td>
</tr>
<tr>
<td>Minimum†‡</td>
<td>0.20 ± 0.12</td>
<td>0.07 ± 0.02</td>
</tr>
<tr>
<td>Total§ fluometuron, preemergence</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum</td>
<td>0.21 ± 0.12</td>
<td>0.09 ± 0.02</td>
</tr>
<tr>
<td>Minimum</td>
<td>0.21 ± 0.12</td>
<td>0.09 ± 0.02</td>
</tr>
<tr>
<td>Dissolved fluometuron, postemergence</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Minimum</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Total fluometuron, postemergence</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Minimum</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Dissolved DMF</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum</td>
<td>0.17 ± 0.09</td>
<td>0.07 ± 0.02</td>
</tr>
<tr>
<td>Minimum</td>
<td>0.17 ± 0.09</td>
<td>0.06 ± 0.02</td>
</tr>
<tr>
<td>Total DMF</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum</td>
<td>0.18 ± 0.10</td>
<td>0.07 ± 0.02</td>
</tr>
<tr>
<td>Minimum</td>
<td>0.18 ± 0.09</td>
<td>0.06 ± 0.02</td>
</tr>
<tr>
<td>Dissolved pendimethalin</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum</td>
<td>0.10 ± 0.10</td>
<td>0.02 ± 0.01</td>
</tr>
<tr>
<td>Minimum</td>
<td>0.06 ± 0.08</td>
<td>0</td>
</tr>
<tr>
<td>Total pendimethalin</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum</td>
<td>0.13 ± 0.12</td>
<td>0.02 ± 0.01</td>
</tr>
<tr>
<td>Minimum</td>
<td>0.08 ± 0.10</td>
<td>0</td>
</tr>
</tbody>
</table>

† Fluometuron and DMF runoff are expressed as a percent of total fluometuron and DMF applied.
‡ Maximum = censored values < MDL = MDL; minimum = censored values < MDL = 0.
§ Total = dissolved plus computed sediment concentration.

Fluometuron Runoff

Fluometuron runoff losses during rainfall simulations were 0.3 to 0.7% of applied with ST plots yielding the greater amount (Table 2). Because fluometuron losses among ST plots were highly variable, average losses were not significantly different from CT results. Much of the ST variability was due to one ST plot during 2001 simulations. Removing this value from data analysis yielded average losses that were nearly identical (ST = 0.27% and CT = 0.28%).

The relatively low soil organic carbon–water partition coefficient (Koc) and much higher water solubility of fluometuron when compared with pendimethalin were apparent in fluometuron’s distribution between sediment-bound and dissolved forms in simulated rainfall runoff samples (Table 2). On ST plots, >98%, and on CT, >89% of the detected fluometuron was dissolved. Fluometuron sediment–water partition coefficients (Kd) computed using dissolved and sediment concentrations averaged 15 mL g⁻¹ in both ST and CT plot runoff samples. This was about 10 to 20 times lower than pendimethalin Kd values.

Desmethylfluometuron in runoff in these studies accounted for 0.01% of the fluometuron applied and 1 to 2% of the total fluometuron. The range in the mass of DMF relative to fluometuron in the rainfall simulation samples corresponded to the mass detected on spray targets. Thus, it appears that there was little fluometuron degradation in the 24 h between herbicide application and simulations. N-demethylation to DMF is the first step in degradation of fluometuron and other phenylurea herbicides (Berger, 1999).

Unlike rainfall simulations, runoff results from 0.15-ha plots due to natural rainfall indicated that fluometuron losses were much greater from CT when compared with ST plots (Table 3), and fluometuron detection frequencies were much higher than pendimethalin in all runoff samples. Fluometuron was >MDL in 83 of 95 CT and 23 of 34 ST runoff samples analyzed, resulting in a very narrow range in maximum and minimum runoff estimates. Like pendimethalin, a large faction of the fluometuron runoff occurred during events close to times of application (Fig. 4).

Overall, fluometuron losses were primarily in the dissolved form (>95%), and runoff rates were within ranges previously reported, <1% of applied (Reddy et al., 1994; Wiese et al., 1980). Rainfall simulations demonstrated that this can be anticipated even under a severe runoff scenario (i.e., an intense storm 1 DAT). The published study reporting higher fluometuron runoff rate (3.3–9.9% of applied) was conducted in a region where soils are unusually runoff prone (Baughman et al., 2001).

Low fluometuron runoff observed in our study was...
probably due to relatively rapid leaching. Results from soil samples collected before and after rainfall simulations revealed that about 50% of the applied fluometuron leached below 2 cm on ST plots and 70% on CT plots (Fig. 5). This is consistent with reports of low fluometuron sorption (Gaston and Locke, 1995; Willan et al., 1997) and one study that reported leaching of 5 to 53% of the applied fluometuron (Essington et al., 1995).

Greater fluometuron detection in the subsoil of CT plots was surprising because the infiltration rate in ST was more than twice that of CT. A possible explanation is that more fluometuron leached below the lowest sampling point (15 cm) on ST plots and was therefore not detected. Depth concentration profiles do not support this (Fig. 5). Fluometuron concentration was greater in all samples collected at 2 to 8 than at 8 to 15 cm after applying 50 mm of simulated rainfall in samples collected on all plots.

An alternate explanation is that interception of the broadcast herbicide spray by desiccated cover crop residue on ST plots may have retarded leaching. Dried plant residues can bind fluometuron strongly (Gaston et al., 2001). Measurements from post-application CT and ST soil samples indicated ST plot residue intercepted on average 49 to 54% of pendimethalin and fluometuron applied in the 2000 simulations and 79 to 82% in 2001. Sorption by desiccated cover crop residue on ST plots may explain why ST plots yielded more fluometuron runoff than CT during rainfall simulations.

Natural rainfall runoff estimates showed that fluometuron losses were about three times greater following the postemergence application on CT plots in 2001 (Table 3). On ST plots runoff following both preemergence and postemergence runoff applications was very low (about 0.02% of fluometuron applied) although high uncertainty in runoff estimates (CV = 100%) was indicated.

Higher CT postemergence versus preemergence fluometuron runoff were linked to runoff events at 4 and 11 DAT after the postemergence application. The first event following the corresponding preemergence application was at 11 DAT. A likely contributing factor to the relatively high runoff rate after fluometuron was applied postemergence was that all plots received 25 mm of irrigation 3 d after the herbicide treatment. This presumably contributed to high soil antecedent water content and increased runoff from CT plots during rain events that followed. Although ST plots were irrigated at the same time, rainfall received the following day did not generate runoff.

The scenario that contributed to relatively high fluometuron runoff from CT plots after postemergence treatment (irrigation followed by rainfall within a few days) may be becoming more common in the region because irrigated cotton production is increasing (Brown et al., 2003). Rainfall although usually abundant (≥1200 mm yr⁻¹), it is not uniformly distributed. To maintain yields, growers must apply supplemental water to meet crop needs during dry periods. This may contribute to a higher runoff risk with postemergence fluometuron applications because they are made when the crop is actively growing and irrigation is likely. It will likely contribute to high soil antecedent water content and potentially high runoff rates should the irrigation be followed by natural rainfall. Rainfall records compiled for the Little River watershed in south-central Georgia indicate a high probability for this scenario (Bosch et al., 1999).

Notably, our data indicated that ST implementation may substantially reduce fluometuron runoff risk under these circumstances.
that irrigation may play in enhancing runoff risks following postemergence fluometuron application. The highest fluometuron runoff rate that was measured during the study was linked to runoff events that followed a postemergence fluometuron application and irrigation.

Further work on degradates of both herbicides also appears warranted. Our study showed that DMF may account for up to 50% of total fluometuron residues in runoff. Thus, DMF should be included in water quality monitoring programs whenever fluometuron is targeted. Formation rates and runoff potential of pendimethalin degradates were not targeted and are a data gap in pendimethalin environmental fate assessments.

Finally, our studies provided insights and raised questions regarding pesticide runoff study design and interpretation. Our ST plots under natural rainfall provided data sets that were highly censored when both flow and herbicide concentration measurements were considered. Thus, we were not able to make statistically based inferences using these data. To some degree this problem was overcome through use of field-based rainfall simulations, which demonstrated that ST significantly reduced pendimethalin runoff when compared with CT. However, rainfall simulations did not indicate differences in fluometuron runoff risk between ST and CT. This was in contrast to plots under natural rainfall where fluometuron runoff from CT plots was substantially greater than from ST plots. We conclude that to effectively evaluate pesticide runoff potential a combination of rainfall simulation and natural rainfall runoff studies may be required.

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