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Tillage management to mitigate herbicide loss in runoff under simulated rainfall conditions

Martin A. Locke [a,*], Robert M. Zablotowicz [b], Krishna N. Reddy [b], R. Wade Steinriede [a]

[a] USDA-Agricultural Research Service, National Sedimentation Laboratory, Water Quality Ecology Research Unit, P.O. Box 1157, Oxford, MS 38655, USA

[b] USDA-Agricultural Research Service, Southern Weed Science Research Unit, P.O. Box 350, Stoneville, MS 38776, USA

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Abstract

Conservation tillage mitigates soil loss in cropland because plant residues help protect the soil, but effects on pesticide movement in surface runoff are not as straightforward. Effects of soil disturbance on surface runoff loss of chlorimuron and alachlor were evaluated utilizing runoff trays. Soil in the trays was either disturbed (tilled) and kept bare or was not tilled, and existing decomposed plant residue was left on the surface. Rainfall (25 mm, 20 min) was simulated 1 d after alachlor (2.8 kg ha^{-1}) or chlorimuron (54 g ha^{-1}) application, and runoff was collected. Runoff fractions were analyzed for herbicide and sediment. Total alachlor loss from bare plots was greater than that in no-tillage plots (4.5% vs. 2.3%, respectively). More than one-third of total alachlor lost from bare plots occurred in the first l of runoff, while no-tillage plots had less runoff volume with a more even distribution of alachlor concentration in the runoff during the rainfall simulation and subsequent runoff period. In contrast, more chlorimuron was lost from no-tillage plots than bare plots (12% vs. 1.5%) even though total runoff volume was lower in the no-tillage plots (10.6 mm vs. 13.6 mm). This was attributed to dense coverage with partially decomposed plant residue in no-tillage plots (1652 kg ha^{-1}) that intercepted chlorimuron. It was likely that chlorimuron, a polar compound, was more easily washed off surface plant residues and transported in runoff.
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1. Introduction

A variety of conservation measures are used to impede the loss of soil and other potential pollutants in surface runoff. Some of these measures include reduced tillage and cover crops on agricultural fields and vegetated buffer zones at the edges of fields. Several studies have described the effects of these conservation measures on physical processes such as sediment and runoff, with the preponderance of evidence indicating that increasing plant residues on the soil surface reduces those losses (e.g., McGregor, 1978; McGregor and Mutchler, 1983). The effects of conservation management on other potential runoff contaminants such as pesticides are mixed (Fawcett et al., 1994; Locke et al., 2005), likely due to the diversity of agrochemicals and a variety of potential biological, chemical, and physical interactions among plant residues and contaminants. Some studies measured lower pesticide runoff concentrations and total loss of pesticide in runoff due to reducing tillage (e.g., Glenn and Angle, 1987). In a field experiment, Baker and Johnson (1979) reported that average annual alachlor loss over a 3-year period under natural rainfall conditions from reduced tillage corn systems ranged from 28% to 48% that of conventional tillage. Other studies demonstrated that pesticide concentrations in runoff from reduced tillage systems are sometimes equal to or higher than that of conventional systems, but overall reduction in runoff from the conservation systems resulted in a net reduction of pesticide loss (e.g., Sauer and Daniel, 1987).

* Corresponding author. Tel.: +1 662 232 2908; fax: +1 662 232 2988.
E-mail address: martin.locke@usda.gov (M.A. Locke).

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Within agricultural fields where conservation tillage or cover crop management is practiced, the soil surface is often characterized by an accumulation of plant residues or increased organic C levels (Locke and Bryson, 1997). The physical barrier imposed by plant residues can intercept pesticides applied directly to soils. These plant residues may also chemically react with pesticides. Once trapped on or within plant residues, further movement of the herbicides into the soil from plant residue (live or decomposed) is influenced by several factors, including plant uptake, affinity of the pesticide for the plant material (Reddy and Locke, 1996), herbicide formulation or adjuvant (Reddy and Locke, 1996), quantity of plant residue (Baker et al., 1982), quantity and intensity of rainfall (Willis et al., 1986), and length of time after pesticide application before rainfall occurs.

Chlorimuron and alachlor are herbicides applied to the soil surface for preemergence control of weeds and thus may be deposited on existing plant residues during application, especially in conservation systems. It is therefore of interest to determine how they may be influenced by plant residue management. The research described in this paper involved two separate studies using simulated rainfall to quantify on a small scale the impact that residue management practices such as conservation tillage may have on surface water quality. Specific objectives of these studies were to evaluate the effect of tillage on: (a) chlorimuron and (b) alachlor loss in runoff.

2. Materials and methods

Small erosion trays and simulated rainfall were used to assess effects of lack of tillage, with concomitant surface plant residue accumulation on chlorimuron and alachlor loss in runoff. Separate evaluations were conducted for each herbicide, but many of the same conditions were used for both experiments.

Although field conditions are more realistic, simulated rainfall and small-plot studies provide a means to control experimental conditions, thus limiting variables that might obscure treatment effects. The experimental apparatus consisted of eight fiberglass runoff trays (2.73 m^2 area: 2.24 m length × 1.22 m width × 0.25 m deep) supported on concrete block pedestals (30 cm high) positioned at a 1.2% slope and filled with Bosket sandy loam soil (fine-loamy, mixed, active, thermic Mollic Hapludalf) to a depth of 0.23 m. Initial characteristics of the soil included 43% sand, 48% silt, 9% clay, 0.88% organic C, and pH 5.5. The trays were located outside and were therefore subject to local weather conditions (average annual rainfall 114–152 cm, average temperature 18 °C).

2.1. Chlorimuron experiment

In the chlorimuron study, four trays were randomly selected as conventional tillage and four trays were selected as no-tillage. Weeds were manually removed in the conventional tillage trays, and the soil surface was disturbed periodically to simulate tillage. Soil in the no-tillage trays was not disturbed, and plant residue accumulation was from summer annual weeds, primarily smooth pigweed (*Amaranthus hybridus* L.), sicklepod [*Senna obtusifolia* (L.) Irwin & Barneby], (Reddy and Locke, 1996) and large crabgrass [*Digitaria sanguinalis* (L.) Scop.]. The weeds died during the summer due to drought over a period of three months prior to herbicide application and rainfall simulation in late summer (average temperature 29 °C). The no-tillage plots had uniformly covered plant residues of 1652 kg ha^{-1} (standard error, SE = 158). Conventional tillage soil was disturbed ("tilled") for the last time 24 h prior to herbicide application, and simulated rainfall (20 mm) was applied to moisten soil in all trays. Immediately before herbicide application, soil was sampled to a depth of 5 cm for gravimetric moisture determination. Three glass petri dishes were placed diagonally along each runoff bed to collect herbicide deposited during application. Chlorimuron (as Classic, 25% a.i., dry flowable formulation, DuPont Agricultural Products, Wilmington, DE) was applied at 54 g a.i. ha^{-1} using a tractor-mounted sprayer (nozzle tips 8004E [Spraying Systems Co., Wheaton, IL], pressure 193 kPa, 6.4 km h^{-1} speed, spray volume 187 l ha^{-1}). After herbicide application, petri plates were removed, rinsed twice with 4 ml methanol and analyzed by HPLC (Waters Model 510 HPLC, Millipore Inc., Milford, MA). HPLC conditions included 1 ml min^{-1} mobile phase flow rate, with a gradient starting at 50%:50% water (pH 2 acetic acid) to acetonitrile with change to 30% water and 70% acetonitrile over 8 min. The retention time for chlorimuron was 5.8 min with UV detection at 230 nm. The HPLC column used was an Alltima (250 mm × 4.6 mm, C18, 5 μm polymer) (Alltech Assoc. Inc., Deerfield, IL).

Simulated rainfall was applied to each tray for 20 min (75 mm h^{-1} rainfall intensity for a total of 25 mm; Vee-Jet, oscillating nozzle) 1 d after herbicide treatment. Runoff was collected in 1-l fractions, weighed, and then immediately placed in refrigerated storage (4 °C). Within 1–4 h of collection, the 1-l fractions were thoroughly shaken, and subsamples were removed and frozen until analysis. Chlorimuron in previously frozen runoff subsamples was analyzed using enzyme-linked immunosorbent assay (ELISA) techniques (EnviroGard plate kit P00-21 chlorimuron, Millipore, Marlborough, MA), using the manufacturer's protocol. Samples were serially diluted as required to conform to the range of quantification for the assay. Water used in runoff studies was fortified with 1–5 ng l^{-1} chlorimuron, resulting in >98% recovery, and none was detected in non-fortified water using this ELISA assay. Limit of detection for the ELISA assay was 0.03 ng l^{-1}.

2.2. Alachlor experiment

In the alachlor study, three trays were randomly selected for conventional tillage, and three trays were selected for no-tillage. Soil in the no-tillage trays was not disturbed, and plant residue consisting of a mixture of Italian ryegrass (*Lolium multiflorum* Lam.) and crimson clover (*Trifolium incarnatum* L.) was established in early spring. The ryegrass–clover mixture died out during summer months, to be replaced by weeds during the summer, resulting in a mixture of residue from several plant species (72% coverage, SE = 0.05). Plant residue decomposition occurred over a period of four to six months prior to herbicide application and rainfall simulation the following winter (average temperature 12 °C). Soil in conventional tillage trays was disturbed periodically, with the last disturbance occurring just before moisture was applied to all trays (24 h prior to herbicide application as described previously). Immediately before herbicide application, soil was sampled to a depth of 5 cm for gravimetric moisture determination.

Alachlor (Lasso EC, Monsanto, St. Louis, MO) was applied at 2.8 kg a.i. ha^{-1} using a tractor-mounted sprayer, simulated rainfall was applied to each tray for 20 min one d after alachlor application, and runoff samples were collected in 1-l fractions as described previously.

ELISA was used to quantify alachlor in runoff water (Millipore EnviroGard plate kit P00-04 alachlor), using the manufacturer's protocol. Samples were serially diluted as required to conform to the range of quantification for the assay. Water used in runoff studies was fortified with 1–5 ng l^{-1} alachlor resulting in >98% recovery, and none was detected in non-fortified water using this ELISA assay. Limits of detection for the assay were 0.05 ng l^{-1}.

2.3. Sediment

To determine sediment in runoff for both chlorimuron and alachlor studies, approximately 100 ml of well-shaken runoff sample was weighed into a beaker. The subsample was oven-dried, and remaining sediment was weighed.

2.4. Herbicide Sorption

Surface (0–5 cm) soil was collected from tilled and untilled Bosket sandy loam soil in a nearby study, and chlorimuron or alachlor sorption was evaluated in batch experiments. Soil samples were air-dried and ground with mortar and pestle and sieved to 2 mm. Ten grams of soil was weighed into 25-ml Corex tubes (Corning, Inc., Corning, NY), and 10 ml of either 2 μg ml^{-1} ^{14}C alachlor or chlorimuron were added. Samples were shaken for 24 h, centrifuged (8000*g*), and supernatant counted for radioactivity using liquid scintillation (Ecolume scintillation cocktail, ICN, Costa Mesa, CA) counting (Tri-Carb 2500 TR, Packard Instrument Co., Downers Grove, IL). Radioactivity in the ^{14}C-labeled alachlor solution was 2163 Bq ml^{-1} (specific activity of stock was 3699 kBq mg^{-1}, 97% purity), and radioactivity in the ^{14}C-labeled chlorimuron solution was 374 Bq ml^{-1} (specific activity of stock was 2160 kBq mg^{-1}, 99% purity). The difference between initial radioactivity in treatment solutions and that of supernatant was considered to be sorbed, and the K_d, or coefficient for the partition of herbicide between soil and solution, was calculated from these values.

2.5. Statistics

Total runoff parameters (alachlor, chlorimuron, and sediment) and sorption K_d's were analyzed as a completely randomized design using SAS General Linear Model procedures (SAS GLM, v. 9.1, Cary, North Carolina). The chlorimuron runoff study, alachlor runoff study, and sorption study had four, three, and eight replications, respectively, of each tillage treatment.

3. Results and discussion

3.1. Sediment and Total Runoff

Total runoff from no-tillage plots in both studies was less than that of the conventional tillage plots (Tables 1 and 2). These results probably resulted from a combination of vegetation coverage and initial soil moisture conditions. Vegetative residue on the surface of no-tillage soils likely slowed water movement, allowing greater infiltration, but higher soil moisture also would reduce infiltration, particularly as soil moisture content approaches saturation. Vegetative cover in the no-tillage plots for the chlorimuron study was denser and uniformly distributed over the entire area (1652 kg ha^{-1}), while only 72% of the area in the no-tillage alachlor plots was covered by a more random distribution of vegetation. The vegetative cover probably inhibited evaporation during the 24-h interim between initially wetting the soil and rainfall simulation resulting in more moisture in no-tillage soils than in respective conventional tillage soils (Tables 1 and 2). Total runoff from both conventional tillage and no-tillage was greater in the chlorimuron study (Table 1) than that from respective tillage treatments in the alachlor study (Table 2). Also, differences

Table 1
Effect of tillage and vegetative cover on soil moisture, total runoff, total sediment loss, and percentage of chlorimuron lost in runoff

	Gravimetric soil moisture (g g^{-1})	Total runoff (mm)	Total sediment loss (kg ha^{-1})	Chlorimuron loss (%)
No-tillage	37[a] (3)[b]	10.6 (1.4)	236 (28)	12 (5.2)
Conventional tillage	28 (1)	13.6 (0.5)	773 (136)	1.5 (0.5)
t-Test *P* value	0.04	0.09	0.01	0.09

[a] Mean of four replicates.
[b] Numbers in parenthesis are standard error.

Table 2
Effect of tillage and vegetative cover on soil moisture, total runoff, and total sediment and percentage of alachlor lost in runoff

	Gravimetric soil moisture (g g^{-1})	Total runoff (mm)	Total sediment loss (kg ha^{-1})	Alachlor loss (%)
No-tillage	21[a] (1)[b]	5.0 (1.7)	35 (15)	2.3 (1.0)
Conventional tillage	16 (1)	10.6 (0.1)	181 (21)	4.5 (0.7)
t-Test *P* value	0.006	0.10	0.03	0.05

[a] Mean of three replicates.
[b] Numbers in parenthesis are standard error.

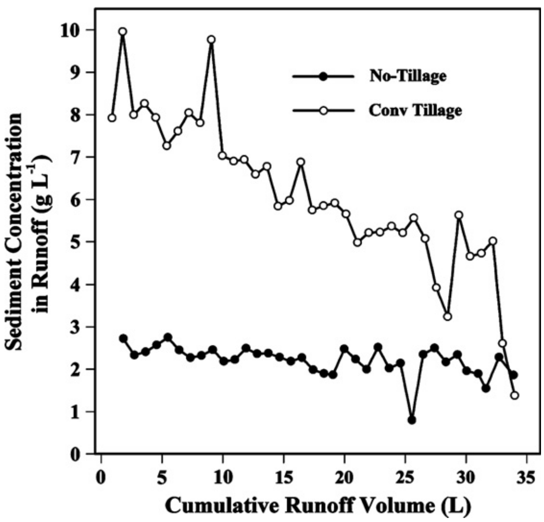

Fig. 1. Effect of tillage on sediment concentration in runoff collected from a rainfall simulation experiment. These data are from the chlorimuron experiment, and each value is the average of four replications.

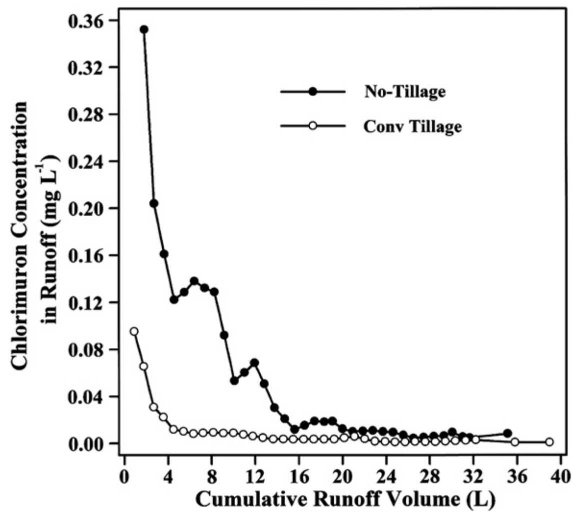

Fig. 2. Effect of tillage on chlorimuron concentration in runoff collected from a rainfall simulation experiment. Each value is the average of four replications.

in runoff between tillage treatments in the chlorimuron study were not as pronounced even though trends were the same. The higher initial soil moistures in the chlorimuron treatment (Table 1) probably inhibited infiltration, although vegetative cover tempered the effect in the no-tillage plots.

While sediment loss in the no-tillage plots remained relatively constant throughout the rainfall event for the chlorimuron study, initial sediment loss was higher in the conventional tillage plots, and gradually declined (Fig. 1), resulting in an overall greater loss of sediment for conventional tillage (Table 1). Sediment loss from the no-tillage plots in the alachlor study was less than one-fourth that of the conventional tillage plots (Table 2). Results from both studies are consistent with numerous studies which have demonstrated the benefit of reduced tillage and plant residues in mitigating sediment loss in runoff (Locke and Bryson, 1997).

3.2. Chlorimuron

Approximately eightfold more chlorimuron was removed in the dissolved phase of runoff from no-tillage plots than from the bare conventional tillage plots, even though runoff volume was lower in the former (Table 1). The highest chlorimuron concentrations for both conventional tillage and no-tillage treatments were in the first 5–10 l of runoff, after which there was a gradual decline in concentration (Fig. 2). This is not an unexpected pattern for sulfonylurea and other herbicides, especially when the rainfall event occurs soon after herbicide application (Wauchope et al., 1990; Reddy and Locke, 1996; Afunyi et al., 1997).

Although more chlorimuron was lost in runoff from no-tillage plots in the current study (Table 1), other reports of the effects of tillage and plant residues on loss of sulfonylurea herbicides in runoff are inconsistent (Wauchope et al., 1990; Afunyi et al., 1997). Higher chlorimuron loss in the dissolved phase of runoff from these no-tillage plots may have been due to a combination of several factors, including chemical characteristics of chlorimuron, varying chlorimuron affinity and retention by soil and surface plant residues, antecedent soil moisture, short interim between herbicide application and rainfall, and herbicide formulation.

The water solubility of chlorimuron (pK_a 4.2) multiplies 40-fold as pH increases from 5 to 7 (11 mg l^{-1} at pH 5; 1200 mg l^{-1} at pH 7, Vencill, 2002). Increases in pH also reduce chlorimuron sorption ($K_{ow} = 320$ at pH 5; $K_{ow} = 2.3$ at pH 7). The soil pH of 5.5 in the current study would have favored chlorimuron sorption, given sufficient residence time in soil. Chlorimuron sorption was higher in no-tillage soil (no-tillage $K_d = 2.06$, SE = 0.04; tilled

$K_d = 0.83$, SE = 0.02, *t*-test *P* value 0.001), but no evaluation was made as to the affinity of the surface plant residues for chlorimuron. In another study (Reddy et al., 1995), chlorimuron sorption to plant residues was higher than that of soil. The soils and residue in that study were treated with chlorimuron dissolved in an aqueous solution.

In this study, chlorimuron was applied as a water dispersible granule formulation in a water carrier. The formulation itself may have more easily washed off the plant residues thereby being removed in the runoff. However, findings relative to effects of formulation are mixed. For example, Wauchope et al. (1990) did not find consistent formulation effects on runoff loss of another sulfonylurea herbicide, sulfometuron-methyl, when it was applied as either a granular or emulsifiable concentrate formulation. Briggs et al. (2002) observed formulation (granular vs. sprayable) differences in isoxaben runoff, but not for trifluralin. Presumably, given sufficient contact time and agitation with plant tissue during the batch sorption experiment (Reddy et al., 1995), sorption to the tissue surface and within the tissue matrix resulted in greater sorption than with soil. In the no-tillage plots, surface plant residue coverage was dense (1652 kg ha^{-1}) and likely intercepted much of the chlorimuron. Sorbents differ as to how strongly a herbicide is retained. The pH of the water applied during the rainfall simulation was 8.4, and this may have promoted chlorimuron desorption from both soil and residue. However, this effect might have been buffered somewhat in the soil (pH 5.5), leading to weaker retention of chlorimuron by the plant residues.

Given the pH of the water in the simulated rainfall and the relatively short duration between chlorimuron application and rainfall simulation, it is likely that the chlorimuron was vulnerable to removal from surface plant residues in the no-tillage plots. Other studies have shown similar results (Reddy and Locke, 1996; Matocha et al., 2006), except that in those studies, washoff was from live plant tissues. Chlorimuron was applied directly to bare soil in conventional tillage plots, and it might be expected that the soil would retain more of the chlorimuron by sorption as compared to residues. In both tillage treatments, the balance of the applied chlorimuron, 88% and 98.5% for no-tillage and conventional tillage, respectively, either remained in the soil or was lost by volatilization or in runoff associated with sediment. Volatilization can be excluded as a significant mechanism of loss for chlorimuron, (vapor pressure 5×10^{-10} at Pa 25 °C, Vencill, 2002). Although the fraction of chlorimuron associated with the sediment phase was not quantified in this study, other reports indicate that sulfonylurea loss in the sediment phase of runoff was negligible (Wauchope et al., 1990; Afunyi et al., 1997). The chlorimuron remaining in soil in the present study would be subject to degradation, sorption, and leaching. Degradation was not likely significant, given the short time lapse. The soil was moistened in all plots just prior to chlorimuron application, and contact with moist soil for 24 h before rainfall simulation may have promoted chlorimuron dissolution and sorption to soil. As indicated earlier, chlorimuron sorption was slightly higher in the no-tillage soil, but either soil could have retained herbicide through this mechanism. Even at the soil moisture levels in both no-tillage and conventional tillage soils prior to rainfall simulation (Table 1) there was opportunity for water infiltration for both tillage treatments, with commensurate leaching of dissolved chlorimuron. A combination of lower moisture content (more water infiltration) and lower chlorimuron sorption in the conventional tillage soils may have contributed to more chlorimuron leaching, thus less chlorimuron available for loss in runoff.

3.3. Alachlor

In the alachlor runoff study, the highest alachlor concentrations in the dissolved runoff phase from bare conventional tillage plots occurred in the first few samplings, after which they gradually declined (Fig. 3). When the initial runoff samples were compared, alachlor concentration from the conventional tillage plots was more than double that from the no-tillage plots (Fig. 3) even though the difference in runoff volume was not of that magnitude. The no-tillage plots had less total runoff volume (Table 2), but the concentration of dissolved herbicide in the runoff was fairly constant during the course of the rainfall event, beginning with the initial runoff sample (Fig. 3). This indicates that alachlor retention by no-tillage soils or plant residues was likely greater. Higher soil sorption for alachlor in no-tillage supports this argument (no-tillage $K_d = 2.72$, SE = 0.03; tilled $K_d = 1.81$, SE = 0.02, *t*-test *P* value 0.001). As well, retention of alachlor by plant residues was likely stronger than was the case with chlorimuron and thus may have inhibited transfer to the dissolved runoff fraction. After the initial runoff samples, alachlor concentrations in both tillage treatments were similar (Fig. 3).

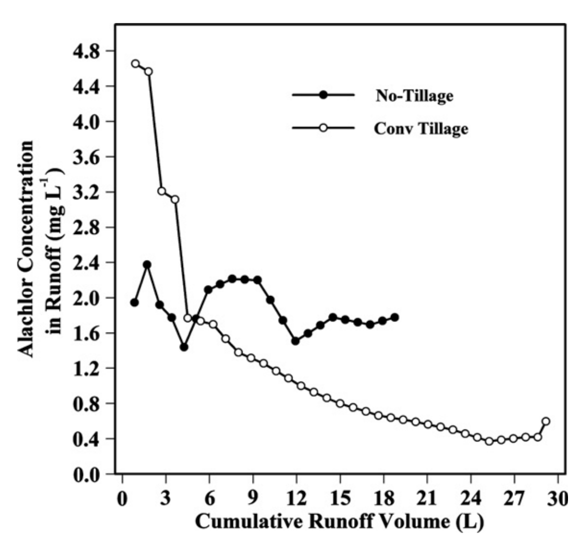

Fig. 3. Effect of tillage on alachlor concentration in runoff collected from a rainfall simulation experiment. Each value is the average of three replications.

The summary outcome of these runoff patterns was that less total herbicide was lost from the no-tillage treatment (Table 2). The range in percentage loss of alachlor and the trends observed compared reasonably with other published values from studies involving simulated rainfall (Baker et al., 1978; Ma et al., 2004). The simulated rainfall in the present study was intense and of short duration. If the rainfall intensity had been less or the duration even shorter, however, the difference in alachlor loss between the conventional tillage and no-tillage treatments may have been even greater. Some researchers have observed greater total annual losses of alachlor in no-tillage systems (0.28% of applied vs. 0.12% for disked systems) (Shipitalo and Owens, 2006). Timing and intensity of rainfall, however, were the most important factors influencing losses in that study as 61% and 72%, respectively, of the alachlor lost in runoff from no-tillage and disked watersheds over a 9-year period resulted from a few intense runoff events less than 40 d after herbicide application.

4. Summary and conclusions

These simulated rainfall and runoff studies demonstrated why evaluating the effects of conservation tillage system on pesticide fate in the environment is complex, and point to the need for more basic research. Results were mixed with regard to effects of tillage management on herbide loss in runoff, and some of those differences might be attributed to the differences in the chemical characteristics and reactions of chlorimuron and alachlor. The susceptibility of a herbicide to loss in runoff appears to be partially dependent upon its solubility. In the present study, alachlor was less water soluble ($242\ \mathrm{mg\ l^{-1}}$) and more nonpolar ($794 K_{ow}$) than chlorimuron (pH range 5–7: water solubility 11–1200 $\mathrm{mg\ l^{-1}}$ and K_{ow} 320–2.3), and had a greater sorption potential in the soil used in these studies, regardless of tillage. It follows that as herbicide sorption increases, loss in runoff associated with sediment may be more important. Herbicide loss in sediment was not measured in the present study, but other researchers who have evaluated alachlor and chlorimuron runoff loss in both dissolved and sediment phases concluded that a majority of the loss was in the dissolved phase (Baker et al., 1982; Sauer and Daniel, 1987; Felsot et al., 1990; Afunyi et al., 1997; Hansen et al., 2001). Since rainfall simulation in this study occurred soon after herbicide application, a significant fraction of either herbicide might be easily desorbed from soil. However, because of a greater capacity for alachlor sorption to soil, more alachlor likely would be associated with sediment in runoff. A reduction in sediment in the no-tillage soil would mitigate potential loss by this mechanism.

Total alachlor loss was greater in conventional tillage plots (4.5% vs. 2.3% in no-tillage), primarily as a function of total runoff loss, with more than one-third of total alachlor loss from conventional tillage plots occurring in the first runoff fractions. Alachlor has more nonpolar characteristics and would tend to be more strongly retained by soil organic components, thus reflected in a more even distribution of alachlor concentration in the dissolved phase of runoff during rainfall simulation. In contrast, chorimuron has pH-dependent polar characteristics, and 12% of applied chlorimuron was lost in runoff from no-tillage plots as compared to 1.5% for the tilled plots even though total runoff volume was lower in the no-tillage plots. This was attributed to dense residue coverage in no-tillage plots ($1652\ \mathrm{kg\ ha^{-1}}$) that intercepted chlorimuron. Because of its polar characteristics, chlorimuron likely was removed from surface plant residues by rainfall and lost in runoff with little interaction in soil, thus, chlorimuron washoff from the residue likely contributed the most to the difference between no-tillage and conventional tillage in runoff loss of chlorimuron in the dissolved phase.

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