Runoff Losses of Norflurazon: Effect of Runoff Timing

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Norflurazon was applied (2.24 kg/ha) in 1988 and 1989 to plots that contained subsurface drains 1 m deep. After soybeans were planted, runoff losses of the herbicide were measured each season for about 300 days. The 1988–1989 season was atypical in that rainfall was insufficient to produce runoff until 171 days after application. From day 171 until day 302, 61 mm of runoff removed 1.5 g/ha (0.07% of application) of herbicide from the plots. In 1989–1990, runoff was measured for 301 days after application. In this period 50.7 g/ha norflurazon (2.3% of the amount applied) was lost from the plots in runoff. The highest concentration in runoff in the first study was 6.6 μg/L on day 171. The highest concentration, 16.5 μg/L, for the second season was also measured during the season's first runoff event, on day 22. The studies provided good examples of the direct relationship between time after application of runoff-producing rainfall and the magnitude of pesticide loss in runoff.

INTRODUCTION

Runoff from agricultural lands is a major nonpoint source of pesticide contamination of surface waters of the United States (Duttweiler and Nicholson, 1983). The greatest volume of pesticides used in American agriculture is accounted for by herbicide application in crop production (Leonard, 1990). Herbicide presence in surface water bodies of the United States has been documented. Some of these studies reported on the following: Chesapeake Bay and tributaries (Kemp et al., 1982; Glotfelty et al., 1984); lower Mississippi River and tributaries (Pereira and Rostad, 1990; Pereira et al., 1992); river basins of the midwestern United States (Thurman et al., 1991). Herbicides in surface waters may damage submerged aquatic vegetation (Forney and Davis, 1981; Kemp et al., 1982; Glotfelty et al., 1984); however, this danger may be greater with metabolites than with parent compounds (Kemp et al., 1982).

Norflurazon [4-chloro-5-(methylamino)-2-[3-(trifluoromethyl)phenyl]-3(2*H*)-pyridazinone] is a major soil-applied herbicide labeled for grass and broadleaf weed control in several crops including cotton and soybean (WSSA, 1989). In Louisiana about 80 000 kg of norflurazon is applied to about 240 000 ha of cotton (Gianessi and Puffer, 1991). As in the United States in general, the chemical is used only occasionally on soybeans in Louisiana. We report here a study of runoff losses of this herbicide from subsurface-drained land planted to soybeans in southern Louisiana.

MATERIALS AND METHODS

Field work was carried out on three replicate plots at the Louisiana Agricultural Experiment Station's Ben Hur Farm, which is about 6 km south of Baton Rouge in East Baton Rouge Parish. The plots, each about 4.0 ha, were on Commerce clay loam (aeric fluvaquent; fine-silty, mixed, nonacid, thermic; pH 5.1; organic matter 1.42%) composed of 36% sand, 31% silt, and 33% clay. Each plot contained three sursurface drains (10-cm-diameter corrugated, perforated polyethylene tubes) 1 m deep, spaced 30 m apart and extending the length of the plot. Each plot was graded to 0.1% slope and bordered to direct runoff through an H-flume. Each flume was equipped with an FW-1 water stage recorder for flow measurement and an automatic sampler for runoff collection. The samplers were set to take 250-mL samples every 3 h during flow events.

Runoff samples (water plus sediment) were stored at 5 °C until extraction. Extraction of the herbicide was accomplished by adding 2 g of sodium chloride to a 250-mL aliquot of the sample and stirring (magnetic) with 100 mL of benzene for 1 h. This procedure removed norflurazon in 71% yield. The runoff results reported in this paper are adjusted for extraction efficiency. Soil samples were collected and worked up as reported in Southwick et al. (1993). The extracts were analyzed by gas chromatography (electron capture detection, 0.53 mm i.d. × 15 m DB-210 megabore column) with the following conditions: injector at 240 °C, column at 195 °C, detector at 350 °C, helium carrier gas at 10 cm^3/min. These conditions provided a 16.9-min retention time for norflurazon.

On June 1, 1988, and on April 26, 1989, Zorial Rapid 80 (Sandoz Crop Protection Corp.) was applied by ground rig at 2.24 kg/ha norflurazon and incorporated to 10 cm. After application, the plots were planted to soybean [*Glycine max* (L.) Merr.]. Previous application of norflurazon to these plots had not been made, since before 1988 the land had been planted to corn.

RESULTS AND DISCUSSION

In the following paragraphs, "1988" refers to the 1988–1989 season (June 1, 1988–March 30, 1989) and "1989" refers to the 1989–1990 season (April 26, 1989–February 21, 1990). Rainfall and runoff for the two study seasons are presented in Figure 1. These graphs are superimposed onto graphs of norflurazon in the top 15 cm of soil, data which were reported by Southwick et al. (1993). In the 1988 season (Figure 1A) rainfall accumulated about half as fast as in the 1989 study (Figure 1B). In 1988 149 mm of rain had fallen by day 50, whereas in 1989, at 49 days after application, 326 mm of cumulative rain was recorded. At 303 days posttreatment in the 1988 study 1000 mm of rain had fallen; 302 days after application in 1989, 2084 mm of rainfall had occurred. This difference in rainfall in the two studies translated into a great difference in runoff volumes for the two seasons. In the 1988 study runoff did not occur until day 171 and amounted to only 61 mm at the end of the investigation (day 302). In 1989 runoff began at the first rainfall event (day 22) and amounted to 825 mm at 301 days.

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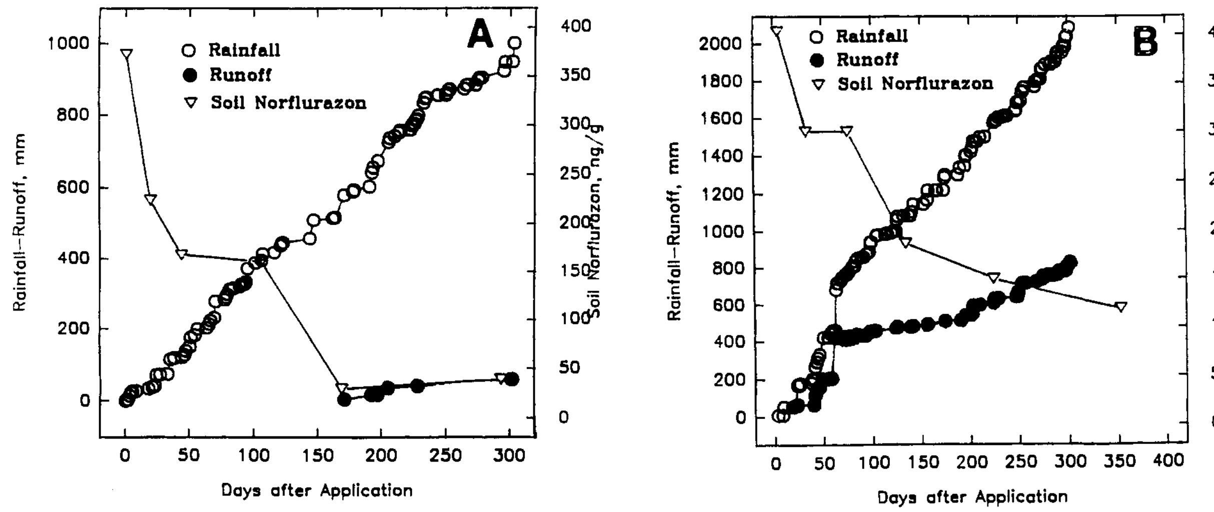

Figure 1. Cumulative rainfall and runoff and norflurazon in top 15 cm of soil: (A) 1988; (B) 1989.

The difference in rainfall for the two seasons (greater for 1989) seems to be inconsistent with the difference in norflurazon soil persistance (DT_{50} was 40 days for 1988 and 120 days for 1989). Increased soil moisture may lead both to greater soil microbial activity (Alexander, 1977) and to higher volatilization rates (Glotfelty and Schomburg, 1989; Taylor and Spencer, 1990). Although runoff did not occur until day 171 in the 1988 season, rainfall routinely fell throughout the season, and at least for microbial activity soil moisture may not have been limiting. Soil temperature differences in the two seasons would have had the opposite effect if any. The first 61 days of the 1988 season (June 1–July 31) were characterized by an average maximum soil temperature of 33 °C, 3 °C higher than the corresponding soil temperature in 1989 (April 26–June 25). During this period, the average minimum soil temperature was 25 °C for 1988, 2 °C higher than for 1989. These data were estimated from two reporting stations (Clinton and Hammond, LA), each about 60 km from the study site (National Climatic Data Center, 1988, 1989). The maximum soil temperatures were in the range (30–40 °C) where differences have little effect on microbial decomposition of organic matter (Alexander, 1977). Small temperature differences can have significant effects on vapor pressure and, therefore, the volatilization rate of chemicals. For example, a temperature increase from 25 to 30 °C more than doubled the vapor pressure of methyl parathion (Glotfelty and Schomburg, 1989). Therefore, the soil temperature difference seems to have been in the right direction, if not magnitude, to have caused greater volatilization loss of norflurazon in the 1988 season. The soil persistance of norflurazon measured by us is similar to reported values. Schroeder and Banks (1986) measured half-lives of about 20–35 days for the herbicide in Georgia soils at 20–30 °C in a greenhouse study. The *Herbicide Handbook* (WSSA, 1989) lists a soil half-life of 45–180 days for the chemical in southeastern soils.

Figure 2 illustrates the concentration pattern of norflurazon in runoff. In each season the first runoff event contained the herbicide in its highest concentration. In the 1988 study (seven runoff events) the first event (day 171) contained norflurazon at 6.59 μg/L. A steady decrease in concentration occurred over the remainder of the study. In the 1989 season (46 events) the herbicide was present at 16.5 μg/L on day 22 (first runoff occurrence). Additional concentration peaks occurred at days 43 (13.1 μg/L) and 58 (11.2 μg/L). After day 100, concentrations of the herbicide in runoff remained below 3.0 μg/L and averaged 1.42 ± 0.65 μg/L (n = 32).

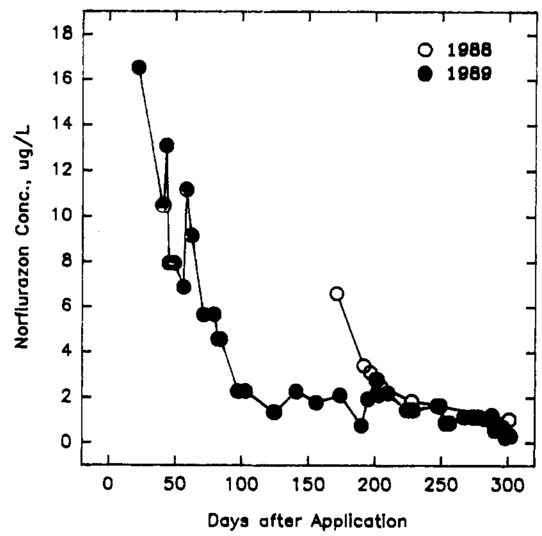

Figure 2. Concentration of norflurazon in runoff.

Cumulative loss of norflurazon in runoff is illustrated in parts A (1988) and B (1989) of Figure 3. Figure 3B also shows individual runoff events. The total loss in the 1988 study amounted to 1.48 g/ha, 0.066% of the application (2.24 kg/ha). This loss was spread out about evenly from day 171 to the end of the study. The total runoff loss of norflurazon in 1989 was 50.7 g/ha, 2.26% of the application. This loss occurred over 280 days (22–301 days after application), but by day 63, 88% of the loss in runoff had occurred. These first 63 days of the season were roughly divided into three time periods: the first event on day 22 amounted to 10.6 g/ha (21% of the total); days 41–58 produced 13.9 g/ha (27% of total) of runoff losses; and on day 62, 19.7 g/ha (39%) of runoff loss occurred. This event on day 62 resulted from 250 mm of rainfall and 216 mm of runoff, the major rainfall–runoff event of the season (Figure 1B). In this single storm event, 14% of the rain fell and 25% of the runoff occurred.

Because subsurface drains increase rainfall infiltration and thereby reduce runoff, norflurazon losses from plots without subsurface drains would have been higher than reported here. In our atrazine–metolachlor study (Southwick et al., 1990), herbicide runoff losses from an area without subsurface drains were 2.3 times the runoff from plots with drains (the same plots used for the present norflurazon study). This difference was due mainly to a greater runoff water volume from the plots without

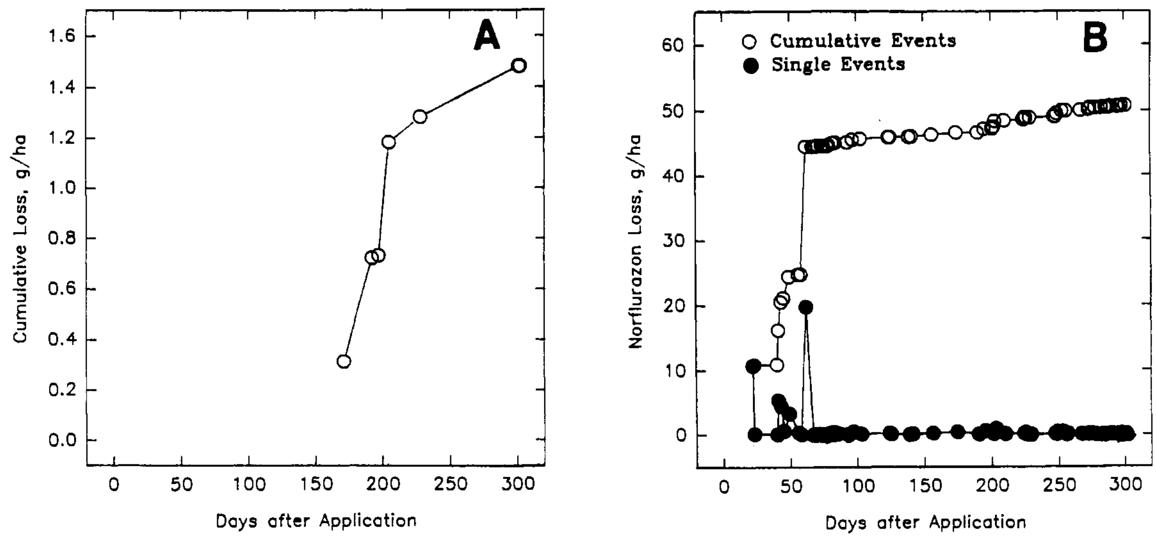

Figure 3. Cumulative loss of norflurazon in runoff: (A) 1988; (B) 1989.

Table I. Comparison of Runoff Studies

herbicide (year)	first runoff event, day	runoff frequency, days	time to 88% of total loss, days	season duration, days	seasonal runoff, mm	seasonal loss, g/ha (% of appl)
norflurazon (1989)	22	6.9	62	125	477	50.7 (2.26)
atrazine (1987)	12	6.5	63	130	254	22.8 (1.40)
metolachlor (1987)	12	6.5	55	130	254	23.1 (1.07)

subsurface drains. Had the split preplant incorporated–preemergence application method been used, more chemical would have been present at the soil surface and higher runoff losses than we report here would have been expected.

In our work we did not distinguish between adsorbed and dissolved norflurazon in runoff. In a review of runoff losses of pesticides from agricultural lands, Wauchope (1978) concluded that for a pesticide with a water solubility greater than 3 mg/L, over 50% of the loss in runoff is likely to be in the dissolved phase. Wauchope et al. (1992) report a water solubility for norflurazon of 28 mg/L and a soil adsorption coefficient (K_{oc}) of 700 mL/g. The sediment load in the runoff events for the first 62 days of the 1989 study averaged 2100 ± 370 mg/L (n = 7). Using K_d = 7 (K_{oc} = 700, soil organic carbon = 0.01), one can estimate from Figure 9-8 of Leonard (1990) that about 98% of the norflurazon load in these runoff events was transported in the water phase.

Our runoff data are a good illustration of the close relationship between time elapsed between application and runoff-producing rainfall with respect to pesticide losses in runoff. The long delay in runoff events in our 1988 study compared to the 1989 work is reflected in the difference in runoff losses for the seasons: the second study produced 34.3 times the losses of the first investigation. Because of dissipation processes, highest pesticide concentrations and most of the losses in runoff tend to occur early in an application season (Leonard, 1990). Bovey et al. (1974) for the triethylamine salts of 2,4,5-T and picloram, Triplett et al. (1978) and Glotfelty et al. (1984) for atrazine and simazine, and Edwards et al. (1980) for glyphosate have observed these trends in runoff. Our work with atrazine and metolachlor (Southwick et al., 1990) also produced examples of the influence of early runoff events on seasonal losses of herbicides. Table I compares the runoff data from the 1989 norflurazon study with this earlier work with atrazine and metolachlor on plots with subsurface drains. In both studies, runoff began early. The time to 88% of total seasonal loss (the present norflurazon study is considered only to day 125 for comparison purposes) agrees within 8 days. Even though the norflurazon work was continued for another 176 days, during which time another 348 mm of runoff occurred, only 4.8 g/ha (0.21% of application) additional norflurazon loss was measured. In the norflurazon study, runoff (millimeters) in 125 days was 1.9 times that observed in the atrazine–metolachlor work in 130 days. This greater runoff volume produced runoff losses (percent of application) of norflurazon that were 1.6 times those for atrazine and 2.1 times those for metolachlor.

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LITERATURE CITED

Alexander, M. *Introduction to Soil Microbiology*, 2nd ed.; Wiley: New York, 1977.

Bovey, R. W.; Burnett, E.; Richardson, C.; Merkle, M. G.; Baur, J. R.; Knisel, W. K. Occurrence of 2,4,5-T and picloram in surface runoff water in the Blacklands of Texas. *J. Environ. Qual.* **1974**, *3*, 61–64.

Duttweiler, D. W.; Nicholson, H. P. Environmental problems and issues of agricultural nonpoint source pollution. In *Agricultural Management and Water Quality*; Schaller, F. W., Bailey, G. W., Eds.; Iowa State University Press: Ames, 1983; pp 3–16.

Edwards, W. M.; Triplett, C. G., Jr.; Kramer, R. M. A watershed study of glyphosate transport in runoff. *J. Environ. Qual.* **1980**, *9*, 661–665.

Forney, D. R.; Davis, D. E. Effects of low concentrations of herbicides on submerged aquatic plants. *Weed Sci.* **1981**, *29*, 677–685.

Gianessi, L. P.; Puffer, C. A. *The use of herbicides in U.S. crop production: use coefficients listed by active ingredient by state*; Resources for the Future: Washington, DC, 1991; p 41.

Glotfelty, D. E.; Schomburg, C. J. Volatilization of pesticides from soil. In *Reactions and Movement of Organic Chemicals in Soils*; SSSA Special Publication 22; Sawhney, B. L., Brown, K., Eds.; Soil Science Society of America: Madison, WI, 1989; pp 181–207.

Glotfelty, D. E.; Taylor, A. W.; Isensee, A. R.; Jersey, J.; Glenn, S. Atrazine and simazine movement to Wye River Estuary. *J. Environ. Qual.* **1984**, *13*, 115–121.

Kemp, W. M.; Means, J. C.; Jones, T. W.; Stevenson, J. C. *Herbicides in Chesapeake Bay and their effect on submerged aquatic vegetation. Chesapeake Bay program technical studies: A synthesis*; U.S. Environmental Protection Agency: Annapolis, MD, 1982; pp 502–566 (National Technical Information Service: Springfield, VA; No. PB84-111202).

Leonard, R. A. Movement of pesticides into surface waters. In *Pesticides in the Soil Environment: Processes, Impacts, and Modeling*: Cheng, H. H., Ed.; SSSA Book Series 2; Soil Science Society of America: Madison, WI, 1990; pp 303–349.

National Climatic Data Center. *Climatological Data Annual Summary, Louisiana*, 1988.

National Climatic Data Center. *Climatological Data Annual Summary, Louisiana*, 1989.

Pereira, W. E.; Rostad, C. E. Occurrence, distributions, and transport of herbicides and their degradation products in the lower Mississippi River and its tributaries. *Environ. Sci. Technol.* **1990**, *24*, 1400–1406.

Pereira, W. E.; Rostad, C. E.; Leiker, T. J. Synthetic organic agrochemicals in the lower Mississippi River and its major tributaries: Distribution, transport and fate. *J. Contam. Hydrol.* **1992**, *9*, 175–188.

Schroeder, J.; Banks, P. A. Persistence and activity of norflurazon and fluridone in five Georgia soils under controlled conditions. *Weed Sci.* **1986**, *34*, 599–606.

Southwick, L. M.; Willis, G. H.; Bengtson, R. L.; Lormand, T. J. Effect of subsurface drainage on runoff losses of atrazine and metolachlor in southern Louisiana. *Bull. Environ. Contam. Toxicol.* **1990**, *45*, 113–119.

Southwick, L. M.; Willis, G. H.; Bengtson, R. L. Leaching losses of norflurazon through Mississippi River alluvial soil. *Bull. Environ. Contam. Toxicol.* **1993**, *50*, 441–448.

Taylor, A. W.; Spencer, W. F. Volatilization and vapor transport processes. In *Pesticides in the Soil Environment: Processes, Impacts, and Modeling*; Cheng, H. H., Ed.; SSSA Book Series 2; Soil Science Society of America: Madison, WI, 1990; pp 213–269.

Thurman, E. M.; Goolsby, D. A.; Meyer, M. T.; Kolpin, D. W. Herbicides in surface waters of the midwestern United States: The effect of spring flush. *Environ. Sci. Technol.* **1991**, *25*, 1794–1796.

Triplett, G. B., Jr.; Conner, B. J.; Edwards, W. M. Transport of atrazine and simazine in runoff from conventional and no-tillage corn. *J. Environ. Qual.* **1978**, *7*, 77–84.

Wauchope, R. D. The pesticide content of surface water draining from agricultural fields—a review. *J. Environ. Qual.* **1978**, *7*, 559–572.

Wauchope, R. D.; Buttler, T. M.; Hornsby, A. G.; Augustijn-Beckers, P. W. M.; Burt, J. P. The SCS/ARS/CES pesticide properties data base for environmental decision-making. *Rev. Environ. Contam. Toxicol.* **1992**, *123*, 1–164.

WSSA. *Herbicide Handbook*; Weed Science Society of America: Champaign, IL, 1989.

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