Influence of Polymers on the Mobility, Loss, and Bioactivity of ¹⁴C from ¹⁴C-Labeled Atrazine, Metolachlor, and Primisulfuron[†]

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Neither soil mobility nor relative losses of ¹⁴C from ¹⁴C-labeled atrazine, metolachlor, or primisulfuron were influenced by tank-mix applications of ASE-108, HA-16, or E-1242 polymer in a 92-day field lysimeter study. In a 3-day soil column laboratory study, however, CGA-A polymer reduced losses of [¹⁴C]atrazine by 7% and addition of E-CELL, CGA-A, CGA-F, or CGA-G polymer increased [¹⁴C]-atrazine mobility by 14–23%. Additions of ASE-108, CGA-A, or CGA-F polymer reduced [¹⁴C]metolachlor losses by 15–23%. G-110, E-CELL, or CGA-C addition reduced [¹⁴C]metolachlor mobility by 10%, and addition of ASE-108, CGA-A, or CGA-F polymer reduced [¹⁴C]metolachlor mobility by 16–21%. Additon of ASE-108 or CGA-B polymer to [¹⁴C]primisulfuron reduced mobility by 8–14%. None of the polymers affected [¹⁴C]primisulfuron losses by more than 7%. Tank-mix additions of G-110 polymer reduced the preemergence bioactivity of atrazine and metolachlor by 20–50% in field studies, but addition of ASE-108, CARBO, or E-CELL polymer had little or no effect. E-CELL polymer additions reduced the postemergence bioactivity of primisulfuron slightly, but the other polymers had little or no effect.

INTRODUCTION

Because nearly 50% of Americans depend on groundwater daily (Severn and Ballard, 1990), groundwater contamination has been a major issue to the public. Approximately 46 pesticides have been found in the groundwater of 26 states (Williams et al., 1988). The majority of the compounds found were preemergence herbicides with moderate to high water solubility and moderate to long half-lives.

Herbicide movement through soil is dependent upon many factors including the chemical and physical properties of the herbicide, the properties of the soil, and climatic conditions (Helling and Gish, 1986; Weber and Miller, 1989).

Atrazine [6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine] and metolachlor [2-chloro-N-(2-ethyl-6-methylphenyl)-N-(2-methoxy-1-methylethyl)acetamide] are two major preemergence crop herbicides that have been found in groundwater of many states (Cohen et al., 1985; Williams et al., 1988). Atrazine is a symmetrical triazine herbicide used to control broadleaf weeds in corn and some other crops (Herbicide Handbook of the Weed Science Society of America, 1989). It has weakly basic properties with a pK_A of 1.68 (Weber, 1970), a low water solubility of 33 mg/L, and a low volatility with a vapor pressure of 3.0×10^{-7} mmHg at 20 °C (Herbicide Handbook of the Weed Science Society of America, 1989). Triazine herbicide sorption in soil has been attributed to binding to organic matter (Harris and Sheets, 1965; Weber et al., 1969) and expanding-type clay minerals (Weber et al., 1965; Weber, 1966) and to be pH dependent (Talbert and Fletchall, 1965; McGlamery and Slife, 1966; Weber, 1966; Weber et al., 1969).

Metolachlor is an acetamide herbicide used to control annual grasses and several broadleaf weeds in corn, cotton, soybeans, and some other crops (Herbicide Handbook of the Weed Science Society of America, 1989). Metolachlor is nonionic, has a high water solubility (530 mg/L at 20 °C), and is moderately volatile (vapor pressure of 1.3×10^{-5} mmHg at 20 °C) (Herbicide Handbook of the Weed Science Society of America, 1989). Major soil constituents reported to inactivate metolachlor included organic matter and expanding-type clay (Ballard and Santelmann, 1973; Kozak et al., 1983; Peter and Weber, 1985).

Primisulfuron [2-[[[[4,6-bis(difluoromethoxy)-2-pyrimidinyl]amino]carbonyl]amino]sulfonyl]benzoic acid] is a recently developed sulfonylurea postemergence herbicide used to control several grassy weeds including johnsongrass and many broadleaf weeds in corn (Ciba-Geigy, 1990). Primisulfuron has weakly acidic properties (pK_A = 5.1), a low water solubility of 70 mg/L (pH 7) at 20 °C, and a low volatility (vapor pressure of $< 7.5 \times 10^{-12}$ mmHg at 20 °C). The mobility of sulfonylurea herbicides in the soil has been correlated with organic matter content and pH (Harvey et al., 1985; Mersie and Foy, 1985, 1986; Goetz et al., 1989;). Soils with high contents of organic matter and low pH tended to reduce the bioactivity of sulfonylurea herbicides.

The mobility of nutrients and pesticides in soils has been shown to be dependent on the properties of the chemicals and the soils using soil leaching columns and lysimeters (Edwards and Glass, 1971; Cassel et al., 1974; Weber and Whitacre, 1982; White et al., 1986; Bowman, 1988). By definition, a lysimeter is a device for measuring water gains (irrigation, precipitation) and losses (evapotranspiration) by a mass of soil under controlled conditions (*Glossary of Soil Science Terms*, 1987). Water and solute movements in disturbed and undisturbed soil lysimeters have also been reported to be different due to the differences in macro- and micropore distributions (Cassel et al., 1974; McMahon and Thomas, 1974).

One method of reducing groundwater contamination by herbicides would be to keep them in the root zone, where they would be taken up by the weeds and available for degradation for a longer period of time. Keeping higher amounts of the chemicals in the zone of germinating weeds might also allow farmers to apply herbicides at lower rates,

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[†] Financial support was provided by Water Resources Research Institute and U.S. Geological Survey (Grant 89-0496), Ciba-Geigy Corp., and Proctor and Gamble Cellulose Co.

thus reducing the amounts of herbicides introduced into the environment and reducing economic costs. Solid porous polymers have been used as controlled-release agents for insecticide vapors and fertilizer nutrients (Baker, 1987), drugs (Rosoff, 1989), and pesticides (Cardarelli, 1976; Lewis and Cowsar, 1977; Scher, 1984, 1988). Several solid porous polymers have been evaluated to determine their ability to decrease the downward movement of herbicides in soils (Cardarelli, 1976). According to Hilton (1987), atrazine movement on soil thin-layer plates was reduced by the addition of a water-soluble ASE-108 polymer obtained from the Rohm and Haas Co. (Philadelphia, PA). Weber et al. (1988) reported that the ASE-108 polymer reduced downward movement of atrazine, metolachlor, and triasulfuron [2-(2-chloroethoxy)-N-[[4methoxy-6-methyl-1,3,5-triazin-2-yl)amino]carbonyl]benzenesulfonamide] in soil leaching columns when applied at a rate of 86 kg of solid/ha. Approximately 90% of each of the three herbicides was recovered in the 0-5-cm section of the leaching columns when ASE-108 was mixed with herbicide prior to application, compared with only 40%when the herbicides were applied alone.

Pesticide bioactivity in soils has been reported to be regulated by both the properties of the specific pesticide and the soil (Harris and Sheets, 1965; Ballard and Santelmann, 1973; Weber and Weed, 1974; Peter and Weber, 1985). Bioactivity is normally reduced when pesticides become bound to soil colloids or diffuse into inaccessible sites. Solid porous polymers used as controlled-release membranes for pesticides and drugs act to reduce chemical concentration at any given moment, thereby regulating chemical losses and utilization by organisms (Cardarelli, 1976; Rosoff, 1989).

The objectives of this experiment were to investigate the effects of three water-soluble acrylic polymers (ASE-108, HA-16, E-1242) on the mobility of atrazine, metolachlor, and primisulfuron in undisturbed soil cores using field soil column lysimeters, to investigate the effects of eight water-soluble acrylic polymers [ASE-108, G-110, CARBOSET-H (CARBO), CGA-A, CGA-B, CGA-C, CGA-F, CGA-G], as well as expanded cellulose (E-CELL), on the mobility of the three herbicides using laboratory soil leaching columns, and to determine the effects of four of the polymers (ASE-108, G-110, CARBO, E-CELL) on the bioactivity of the three herbicides in the field.

MATERIALS AND METHODS

Lysimeter Study. The field lysimeter experiment was conducted on a Dothan loamy sand (fine loamy, siliceous, thermic, *Plinthic Kandiudult*) in 1989 at the Central Crops Research Station, Clayton, NC. Soil properties including percent organic matter (chromic acid oxidation method) (Walkley and Black, 1934), pH (1:1 soil:water), soil texture (hydrometer method; Gee and Bauder, 1986), and cation-exchange capacity (CEC) (1.0 N ammonium acetate replacement method; Thomas, 1982) were determined by A&L Agricultural Laboratories, Omaha, NE. Percent soil humic matter was determined by the North Carolina Department of Agriculture, Raleigh, NC (Mehlich, 1984).

Lysimeters consisted of 0.129 cm (16 gauge) thick steel columns (20.3 cm i.d. and 95 cm in length), which were driven 90 cm into the soil using a tractor-mounted post driver.

Atrazine, as a mixture of formulated AAtrex 4L and ¹⁴C-ringlabeled atrazine (specific activity = 19.5 μ Ci/mg, 99.5%) and each polymer, was applied uniformly in a cross-hatch pattern in 10 mL of water at the rate of 1.12 kg of ai/ha to the soil surface in the lysimeter using a pipet. Total radioactivity for each atrazine-treated column was 15.4 μ Ci. Metolachlor, as a mixture of Dual 8E and ¹⁴C-ring-labeled metolachlor (specific activity = 19.3 μ Ci/mg, 99.7%) and each polymer, was applied in the same manner as for atrazine at the rate of 2.24 kg of ai/ha. Total radioactivity applied was $15.1 \,\mu$ Ci per lysimeter. Primisulfuron, as ¹⁴C-ring-labeled primisulfuron (specific activity = $56.2 \,\mu$ Ci/mg, 99.5%) and each polymer, was applied in the same manner as for the other herbicides at the rate of 0.08 kg of ai/ha. Total radioactivity applied was 14.7 μ Ci per lysimeter. In addition to the herbicide/polymer mixtures, each herbicide was applied without polymer for comparison.

Three Rohm and Haas Co. water-soluble acrylic polymers (ASE-108, HA-16, E-1242) were mixed with the respective herbicides and applied to the soil surface in the lysimeter at rates of 0.64, 1.63, and 1.80 kg of solid/ha, respectively. The amounts of polymer added would be equal to 2% of a water application volume of 168.4 L/ha, which is commonly used by farmers to apply herbicides. The density of all three polymers was 1.1 g/cm³. The polymers were selected to represent anionic, nonionic, and cationic charged materials, respectively. ACRYSOL ASE-108 (ASE-108), described as a 20% solid suspension copolymer emulsion stabilizer, is used to stabilize liquid laundry and machine-dishwashing detergents and to formulate flowable pesticide products. It is shipped in an anionic acid form (pH 3.0) and must be neutralized to pH 8 to initiate polymerization and obtain high viscosity. RHOPLEX HA-16 (HA-16) is a nonionic 45% solid suspension, pH 3.0, self-cross-linking acrylic polymer used in a wide variety of textile applications. EMULSION E-1242 (E-1242) is a cationic 50% solid suspension, pH 4.5, self-crosslinking acrylic emulsion used in a wide variety of textile applications. The advantage of E-1242 is that it polymerizes at room temperature after drying.

After the herbicide or herbicide/polymer application, the soil was allowed to dry overnight (24 h), and water was added weekly, as necessary to maintain the 10-year average weekly precipitation. Lysimeters were removed from the field and sectioned horizontally at the end of 92 days. Soil samples from each 7.6-cm section were placed in plastic bags, mixed thorougly, and stored in the freezer at -20 °C until analyzed. ¹⁴C radioactivity for each soil section was determined by using four 1-g subsamples which were combusted in a biological oxidizer (Model OX-300, R. J. Harvey Instrument Corp., Hillsdale, NJ), and the ${}^{14}CO_2$ was trapped in 15 mL of Harvey ¹⁴C scintillation cocktail. ¹⁴C radioactivity was assayed using a liquid scintillation analyzer (Packard Model 2000 CA, Packard Instrument Co., Downers Grove, IL) and converted to total ¹⁴C activity recovered for each section. Soil moisture content for each section was determined by drying samples in the oven at 105 °C for 24 h. Reproducibility among subsamples was $\pm 3\%$

Modified R_f values were calculated for each column to allow for comparison to be made among herbicide and polymer treatments using (Weber, 1991)

$$R_{f} = \sum_{i=1}^{n} (D_{i}F_{i})/(\text{MD} \times \text{TF})$$
(1)

where R_i is the index of herbicide mobility, n is the number of sections of the entire soil column, D is the mean depth of section i (total distance from soil surface to the midpoint of section i in cm), F is the fraction of herbicide applied in section i, MD is the maximum mean depth (total distance from soil surface to the midpoint of the last section in cm), and TF = 1.0 (total fraction of herbicide applied in the entire soil column).

A small R_f value indicates that the herbicide is nonmobile and travels a short distance in the soil column; a large R_f value indicates that the herbicide is very mobile and travels a long distance. [For instance, the smallest R_f value possible for this lysimeter would be $(3.8 \text{ cm} \times 1.0) + (88.0 \text{ cm} \times 1.0) = 0.04$, which indicates that all applied herbicide was found in the first section (0-7.6 cm,mean = 3.8 cm) of the lysimeter. A large R_f value of 1.0 from $(88.0 \text{ cm} \times 1.0) + (88.0 \text{ cm} \times 1.0)$ indicates that all of the herbicide was found at the bottom (83.6-90.0 cm, mean = 88.0 cm) section of the lysimeter.] The experiment was arranged as a randomized complete block with two replications. Total percent recovery data and R_f values were subjected to analysis of variance at the 5% confidence level.

Leaching Column Study. The A horizon (0-15-cm depth) of Dothan loamy sand was air-dried and passed through a 2-mm sieve mesh before being packed into the 5.1 cm i.d. by 20 cm long cellulose tubing soil leaching columns. One end of each column was covered with a vinyl cap with a centered 1 cm diameter hole. Three layers of cheesecloth followed by 68 g of quartz sand were put in the bottom of each column to prevent soil loss. Dothan soil was added in 70-g increments to each column. Each increment (2 cm) of soil was evenly distributed in the column by tapping the side of the column until a 12-cm soil core with a bulk density of 1.71 g/cm³ was obtained.

All polymer solutions were prepared with $0.01 M (NH_4)_2 HPO_4$, pH 8, buffer solution to meet the polymerization requirement of ASE-108.

Atrazine, as a mixture of formulated AAtrex 4L and ¹⁴C-ringlabeled atrazine (specific activity = $53.5 \ \mu$ Ci/mg, 99.5%) and each polymer, was applied uniformly to the soil surface of each column in a cross-hatch pattern in 5 mL of water at the rate of 1.12 kg of ai/ha.

Metolachlor, as a mixture of formulated DUAL 8E and ¹⁴Cring-labeled metolachlor (specific activity = $71.5 \,\mu$ Ci/mg, 99.7%) and each polymer, was applied in the same manner as for atrazine at the rate of 2.24 kg of ai/ha.

Primisulfuron, as a mixture of formulated BEACON and ¹⁴Cring-labeled primisulfuron (specific activity = $52.4 \ \mu$ Ci/mg, 99.5%) and each polymer, was applied as for the other herbicides at the rate of 0.04 kg of ai/ha. Each column received 0.4 μ Ci of each herbicide.

Polymers used in this experiment were ASE-108, G-110, CARBO, E-CELL, CGA-A, CGA-B CGA-C, CGA-F, and CGA-G. All polymers were mixed with the respective herbicide and applied at a rate of 1.12 kg of solid/ha. In addition to the herbicide/polymer mixtures, herbicides were applied without polymer for comparison.

ASE-108 and G-110 are products of Rohm and Haas Co. G-110, has a density of 1.1 g/cm³. It is an anionic, pH 9.5, ammonium polyacrylate solution that is used in the textile industry for thickening synthetic and natural lattices. CARBOSET-H (CAR-BO), a low molecular weight (density of 1.1 g/cm³), anionic, 40% solid suspension, pH 7.0, acrylic resin which produces a film adhesive to all kinds of materials after drying, is manufactured by BF Goodrich Co. (Cleveland, OH). Expanded cellulose (E-CELL) is a product of the Procter and Gamble Cellulose Co. (Memphis, TN) and is made by dispersing natural fibers in water. The nonionic, 4% solid suspension, pH 7.0, cellulose has a myriad of voids among fibers; however, all fibers collapse upon drying due to the formation of hydrogen bonds, and redispersion is impossible. CGA-A, CGA-B, CGA-C, CGA-F, and CGA-G, with percent solid suspensions of 25, 19, 25, 19, and 17, respectively, were synthesized specifically for this project by Ciba-Geigy Corp. (Greensboro, NC). CGA-A, CGA-B, and CGA-F were described as 50/50, 10/90, and 20/80 butyl esters of acrylic acid, respectively. The polymers CGA-C and CGA-G were described as 50/50 and 20/80 ethyl esters of acrylic acid. Lipophilicity of the CGA polymers was in the order $A \gg C > F \gg G = B$.

After herbicide/polymer applications, the columns were allowed to air-dry for 48 h. One container-capacity volume [approximately 1 field-capacity (FC) pore volume of distilled-deionized water (72 mL, equivalent to 3.3 cm)] was added to each column to distribute water and the applied chemicals chromatographically throughout the soil profile. Twelve hours later each soil column was cut into six 2-cm sections, and each section was placed into a plastic bag and mixed thoroughly. ¹⁴C activity and moisture content of each section were determined as described above. R_f values were calculated using eq 1.

The smallest R_f value possible in this experiment was $(DF) \div (MD \times TF) = (1.0 \text{ cm} \times 1.0) \div (11.0 \text{ cm} \times 1.0) = 0.09$, which indicates that all of the applied herbicide was found in the first section (0-2 cm, mean depth = 1.0 cm). The maximum R_f value of 1.0 would indicate that all of the herbicide was found at the bottom section of the column $(11.0 \text{ cm} \times 1.0) \div (11.0 \text{ cm} \times 1.0)$.

The experiment was arranged as a randomized complete block design with two replications. Total percent recovery data and R_f values were subjected to analysis of variance at the 5% confidence level, using the Statistical Analyses System (User's Guide: Statistics, 1985). All ¹⁴C wastes were disposed of by the North Carolina State University Life Safety Services following proper procedures (Manual For Chemical Waste Management, 1991).

Herbicide/Polymer Bioactivity Study. Corn was planted on a Rains sandy loam soil (fine-loamy, siliceous, thermic Typic Palequult) at the Upper Coastal Plains Research Station, Rocky Mount, NY, on May 14, 1990. Soil property analyses, including soil texture (13, 34, and 54% clay, silt, and sand, respectively), pH (5.2), organic matter content (1.9%), and cation-exchange capacity (CEC = 5.3) were determined on A horizon (0-15-cm depth) soil samples as described previously. Four corn row test plots $(3.6 \times 6.1 \text{ m})$, with two weed species planted on either side of the two center corn rows, were arranged as a randomized complete block design (three replicates). The weeds included barnyardgrass (BG) [Echinochloa crus-galli (L.) Beauv.], prickly sida (PS) (Sida spinosa L.), green foxtail (GF) [Setaria glauca (L.) Beauv.], and velvetleaf (VL) (Abutilon theophrasti Medik.). Dense, uniform weed populations were obtained with few indigenous weeds present.

Atrazine and metolachlor/polymer tank-mix treatments were applied preemergence, and primisulfuron/polymer tank-mix treatments were applied postemergence when corn was at the 6-leaf stage (ca. 15 days after the preemergence treatments). Commercial atrazine (AAtrex Nine-0) and metolachlor (DUAL 8E) and experimental primisulfuron (BEACON) were applied at rates of 0, 0.67, 0.90, and 1.12 kg/ha; 0, 0.45, 0.67, and 0.90 kg/ha; and 0, 0.009, 0.011, and 0.013 kg/ha, respectively, alone and in tank-mix combination with polymers ASE-108, G-110, CARBO, and E-CELL, at rates of 0.06, 0.08, and 0.11 kg/ha; 1.12, 2.24, and 3.36 kg/ha; 1.12, 5.60, and 11.21 kg/ha; and 1.12, 2.24, and 3.36 kg/ha, respectively. ASE-108 solutions were adjusted to pH 8.0 with 0.01 M (NH₄)₂HPO₄ and NH₄OH. All herbicide/polymer treatments were applied with a CO₂ backpack sprayer with hollow cone "whirl chamber" nozzles (Monarch 49×49) at a pressure of 236 kPa.

Weeds were rated visually using a scale of 0% (no injury) to 100% (complete termination) 30 days after application. Injury ratings were subjected to analysis of variance at the 5% confidence level using the Statistical Analysis System (User's Guide: Statistics, 1985).

RESULTS AND DISCUSSION

Soil Properties. The soil clay content increased with depth from 6% in the surface to 29% at 90 cm in the lysimeter study. Soil texture changed from loamy sand at the soil surface (0-45.6 cm) to sandy loam (45.6-60.8 cm)cm) and sandy clay loam (60.8–90 cm) in the subsoil. Soil pH was 6.1 in the surface and decreased with depth to 4.4 at 90 cm. Soil organic matter and humic matter contents also decreased with depth from 1.1 and 0.4%, respectively, in the surface, to 0.4 and 0.1%, respectively, at 90 cm. Soil moisture content inside the lysimeter was determined at the time of column removal, 3 months after treatment. Water content in the lysimeter ranged from a low of 5.4%in the surface soil to a high of 16.5% in the subsoil at the bottom of the lysimeter. CEC in the soil profile was relatively uniform throughout the profile, ranging from 1.0 to 2.1 mequiv/100 g of soil due to the higher organic matter content in the surface soil and higher clay content in the subsoil

Lysimeter Study. Mean temperature for the period of the study was 27.1 °C, and precipitation and irrigation input for July, August, and September 1989 was 18.5, 11.1, and 15.5 cm/month, respectively, which was 47% above the 10-year average for the area.

Total ¹⁴C recoveries in the ¹⁴C-labeled atrazine, metolachlor, and primisulfuron treatment without polymer addition (control) after 92 days in the field were 40.6, 54.8, and 40.5%, respectively (Table I). On the basis of R_f values from treatments with herbicide alone (control), ¹⁴C from [¹⁴C]metolachlor ($R_f = 0.30$) was the most mobile material in this experiment due to metolachlor's nonionic nature and higher water solubility compared to the other two compounds. The major extractable metabolite of metolachlor is an oxalic acid derivative of metolachlor [*N*-(2-

Table I. Effect of Water-Soluble Acrylic Polymers on the ¹⁴C Distribution and Recovery from ¹⁴C-Labeled Herbicides over a 92-Day Period

		total reco	very, %		nc	rmalized recov	ery,ª %	
mean depth, cm	control	ASE-108	HA-16	E-1242	control	ASE-108	HA-16	E-1242
				Atrazine				
3.8	23.6	22.0	16.3	25.1	58.0	58.6	42.3	60.8
11.4	10.3	7.0	13.5	11.0	25.5	18.8	34.9	26.7
19.0	4 4	5.2	4.8	31	10.8	13.8	12.4	74
26.8	14	18	14	0.1	35	4.9	37	2.2
20.0	0.3	0.6	0.9	0.0	0.7	1.5	0.1	0.9
40 1	0.0	0.0	0.9	0.3	0.7	1.0	2.0	0.8
42.1	0.1	0.3	0.4	0.2	0.3	0.0	1.0	0.0
49.1	0.1	0.2	0.3	0.1	0.2	0.0	0.9	0.2
07.4	0.1	0.1	0.3	0.1	0.3	0.3	0.8	0.3
65.0	0.1	0.1	0.6	0.1	0.3	0.2	1.4	0.3
72.7	0.1	0.1	0.1	0.1	0.1	0.2	0.2	0.3
80.3	0.1	0.1	0.1	0.1	0.2	0.2	0.2	0.2
88.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.1
total	40.6	37.5	38.5	41.3	100.0	100.0	100.0	100.0
$LSD_{0.05} =$	8.9				$R_f = 0.10$	0.11	0.14	0.10
					$LSD_{0.05} = 0.05$			
0.0	00.0	17.4	15.0	Metolachior	00.4	04.4	00.0	00.0
3.8	20.0	17.4	15.9	10.0	30.4	34.4	30.8	29.2
11.4	8.7	8.2	7.3	7.1	15.8	16.3	14.0	13.4
19.0	4.9	4.3	5.7	3.8	8.9	8.6	11.0	7.3
26.8	3.4	3.7	3.6	3.0	6.3	7.2	7.0	5.6
34.4	2.5	2.9	2.4	2.2	4.5	5.8	4.7	4.1
42.1	2.0	2.6	2.7	2.1	3.7	5.1	5.2	3. 9
49.7	2.0	2.5	2.6	2.1	3.6	4.9	5.1	4.0
57.4	2.0	2.0	2. 9	3.6	3.7	3.9	5.5	6.9
65.0	3.4	2.7	2.7	4.4	6.1	5.3	5.2	8.3
72.7	2.8	2.1	2.7	4.4	5.1	4.3	5.2	8.3
80.3	1.9	1.4	2.2	3.4	3.5	2.8	4.2	6.5
88.0	1.2	0.7	1.1	1.4	2.2	1.3	2.0	2.6
total	54.8	50.4	51.7	53.0	100.0	100.0	100.0	100.0
$LSD_{0.05} =$	10.0				$R_f = 0.30$	0.29	0.32	0.37
					$LSD_{0.05} = 0.11$			
				Datastaslessa				
9.0	14.0	15 4	10.9		94 5	. 04 9	94.6	05.4
114	14.0	10.4	19.0	14.4	04.0	24.0	0.46	20.4
11.4	14.0	22.4	20.3	20.0	00.0 10.9	30.2	30.3	40.7
19.0	4.4	11.2	6.8	9.3	10.8	18.0	12.3	16.4
26.8	2.8	7.0	2.8	1.9	6.9	11.3	5.0	3.3
34.4	1.1	1.2	0.6	1.1	2.7	1.8	1.1	1.9
42.1	0.8	0.5	1.0	0.8	2.0	0.8	1.7	1.4
49.7	0.4	1.2	0.6	0.6	1.0	1.9	1.0	1.1
57.4	0.8	0.8	1.1	0.6	2.0	1.4	1.9	1.0
65.0	0.5	0.6	1.3	0.5	1.4	1.0	2.3	1.0
72.7	0.4	0.8	1.0	0.2	1.1	1.4	1.7	0.3
80.3	0.6	0.6	0.8	0.5	1.5	0.9	1.5	0.9
88.0	0.3	0.2	0.3	0.3	0.8	0.4	0.6	0.6
total	40.5	61.9	55.8	56.7	100.0	100.0	100.0	100.0
$LSD_{0.05} =$	21.7				$R_{f} = 0.18$	0.19	0.18	0.17
					$LSD_{0.05} = 0.07$			

^a Normalized to 100% recovered, assuming equivalent losses from all depths, so as to compare herbicide mobility and effects of polymers.

methoxy-1-methylethyl)-2-ethyl-6-methyloxalic acid anilide] (Lebaron et al., 1988). As an acid, it was probably repelled by soil clay. ¹⁴C from [¹⁴C]primisulfuron (R_f = 0.18) was less mobile than ¹⁴C from [¹⁴C]metolachlor but more mobile than ¹⁴C from [¹⁴C] atrazine ($R_f = 0.10$). Beyer et al. (1988) reported that the major metabolite from the hydrolysis of sulfonylurea herbicides, such as primisulfuron, is a sulfonamide derivative, which is less mobile than the parent in the soil profile due to its weakly basic properties. In addition, Mersie and Foy (1985, 1986) reported that moderately high soil sorption and low phytotoxicity of chlorsulfuron [2-chloro-N-[[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)amino]carbonyl]benzenesulfonamide], a similar sulfonylurea herbicide, were observed in an acid soil. Atrazine has weakly basic properties and was reported to be sorbed in high amounts on acidic soil surfaces due to its subsequent sorption to exchangeable

sites by Coulombic forces (Weber, 1969, 1970). It was, therefore, the least mobile herbicide in this study over the 92-day period.

Approximately 40% of the total ¹⁴C applied was recovered in the [¹⁴C]atrazine treatment regardless of the addition of polymers, indicating none of the polymers affected the loss of the herbicide (Table I). Most of the ¹⁴C activity (approximately 20% of applied) was retained in the upper 7.6-cm (mean = 3.8 cm) sections of the columns. For a polymer to be effective in either reducing herbicide movement or reducing herbicide loss, the amounts recovered in the 3.8-cm section of herbicide/ polymer treatments must be higher than those from control soils (no polymer). Of the polymers, HA-16 influenced ¹⁴C from [¹⁴C]atrazine mobility the most, increasing it slightly; however, the differences were not significantly different [16.3% recovered (3.8-cm zone) vs 23.6% for the control and an R_f value of 0.14 with HA-16 vs 0.10 for the control].

Calculated R_f values for the various [¹⁴C]atrazine/ polymer treatments showed that none of the polymers affected the [¹⁴C]atrazine leaching pattern, and thus atrazine mobility, to a significant degree (Table I).

Since the organic matter content was very low throughout the soil column, [¹⁴C]metolachlor activity found throughout the lysimeters was not surprising (Table I). Metolachlor was a much higher water solubility than atrazine, and sorption is not pH dependent as is the case for atrazine. Extraction and thin-layer separation of parent herbicides and metabolites in similar lysimeter studies suggest that the ¹⁴C was not parent herbicide (Keller, 1992).

None of the polymers was effective in influencing the loss or mobility of [14C]metolachlor (Table I). Approximately 50% of the total 14C applied was recovered regardless of polymer treatment. Of that recovered, approximately one-third was found in the upper 7.6-cm (mean = 3.8 cm) section regardless of the polymer added or not added. The highest calculated R_f value was obtained in the E-1242 treatment due to the higher ¹⁴C activity recovered in the 61–90-cm depth; however, the value was not statistically different from the control.

Total ¹⁴C recovered in the [¹⁴C]primisulfuron control treatment was 40%, and although lower than where each of the polymers was added, the differences were not significant (Table I). Approximately 30% of the recovered ¹⁴C was located in the upper 7.6-cm (mean depth = 3.8cm) zone regardless of whether polymer was added or not. Calculated R_f values for polymer vs no polymer treatments were not significantly different, suggesting that the added polymers did not affect [14C]primisulfuron mobility in the soil. Since differences in herbicide mobility and losses attributable to polymers were minor, the herbicide molecules appeared to be weakly bound and easily desorbed by infiltrating water. There was no relationship between the properties of the polymers (Table I) and the properties of the herbicides as expressed in the amounts of herbicides recovered or their calculated R_f values.

Leaching Column Study. Since the herbicide/polymer applications (48 h on dry soil) and leaching processes (12 h) were completed in approximately 60 h total in this study, it is likely that all detected ¹⁴C was parent compound, so loss and mobility are of parent compounds only, in contrast with the 92-day lysimeter study in which metabolites were involved. In addition, only the Dothan A horizon (0–15-cm depth) soil was used in the leaching study, while the entire 0–90-cm surface and subsoil were involved in the lysimeter study.

¹⁴C-labeled herbicide distributions, as affected by polymer additions, are shown in Table II. Total recoveries of atrazine, metolachlor, and primisulfuron alone (control) were 81.3, 74.6, and 74.5%, respectively. Calculated R_f values for atrazine, metolachlor, and primisulfuron alone (control) were 0.22, 0.19, and 0.50, respectively; thus, the relative mobility in descending order was primisulfuron \gg atrazine \geq metolachlor. Primisulfuron was present primarily in the parent anion form and was probably repelled by the negatively charged soil clay, as it was carried chromatographically through the soil over the 12-h period; therefore, primisulfuron was the most mobile herbicide in this study. The pH of the Dothan A horizon soil was approximately 6, so parent atrazine was present primarily in the molecular form. Therefore, both parent atrazine and metolachlor were nonionic in this study and had similar mobilities $(R_f = 0.22 \text{ and } 0.19, \text{ respectively})$. They were much less mobile than primisulfuron, which had an R_f value more than twice that of the two herbicides.

Total ¹⁴C recovery data showed that the total amount of atrazine recovered increased significantly from 81.3% without polymer to 89.5% with the addition of CGA-A polymer (Table II). It appears that this polymer reduced atrazine losses by approximately 7%. The eight other polymers had no effect. According to calculated \overline{R}_f values, the addition of E-CELL ($R_f = 0.25$), CGA-A ($R_f = 0.27$), CGA-F ($R_f = 0.25$), and CGA-G ($R_f = 0.26$) increased atrazine mobility significantly over that of atrazine applied alone (control) ($R_f = 0.22$), but the other four polymers (G-110, CARBO, CGA-A, CGA-C) had no effect. Higher herbicide mobility by polymer additions suggests that the chemicals were more easily released from the polymers than from the Dothan soil itself or that the herbicide and polymers moved together more readily than the herbicide moved alone.

Two-thirds of the polymers increased the total recovery of metolachlor, while the others had no effect (Table II). Total recovery of metolachlor was significantly higher than the metolachlor control alone (74.6%) when polymer ASE-108 (97.9%), CGA-F (93.4%), CGA-A (89.9%), G-110 (79.5), CGA-G (79.4%), or CARBO (78.9%) was added. Apparently the polymers bound metolachlor more effectively against losses than did the Dothan A horizon soil. The other three polymers (E-CELL, CGA-B, CGA-C) had no effect.

On the basis of calculated R_f values, polymer ASE-108 ($R_f = 0.15$), G-110 ($R_f = 0.17$), E-CELL ($R_f = 0.17$), CGA-A ($R_f = 0.16$), CGA-C ($R_f = 0.17$), and CGA-F ($R_f = 0.16$) additions reduced metolachlor movement as compared to metolachlor applied alone ($R_f = 0.19$) (Table II). The quantity of metolachlor retained in the 0–2-cm soil zone (mean depth = 1.0 cm) was also higher and ranged from 47.8 to 74.1% for columns where these polymers were added compared with the control column (43.9%). The polymers were apparently more effective in binding metolachlor against desorption by water than was the Dothan soil. The other three polymers (CARBO, CGA-B, CGA-G) had no effect.

Total recovery of primisulfuron was not increased by polymer additions (74.5 \pm 5.0%), but two polymers (G-110, CGA-C) did significantly reduce primisulfuron recovery by 5.7 and 6.9%, respectively (Table II). This suggests that primisulfuron was lost from these two polymers more readily than it was from the Dothan soil; i.e., the polymers were less sorptive for the herbicide than the soil was.

The addition of ASE-108 and CGA-B resulted in significantly smaller R_f values for primisulfuron, $R_f = 0.43$ and 0.46, respectively, than the primisulfuron control, R_f = 0.50, indicating that the two polymers retained primisulfuron against leaching more effectively than did the soil (Table II). A significantly greater amount of primisulfuron was retained in the 1.0- and 3.0-cm zones when ASE-108 was added (33%) than when no polymer was added (24%), also suggesting that the polymer retarded movement of the herbicide. Four of the polymers (CAR-BO, E-CELL, CGA-F, CGA-G) increased R_f values, $R_f =$ 0.56, 0.54, 0.55, respectively, over primisulfuron alone, R_f = 0.50, suggesting that these polymers retained the herbicide against desorption by water less effectively than did the soil or that they moved through the soil along with herbicide. None of these four polymers affected the quantity of primisulfuron that was retained in the 1.0- or 3.0-cm zones of the soil, however. The other three polymers (G-110, CGA-A, CGA-C) had no effect.

Table II. Columns	Effect	of Wa	ter-Sol	uble Acı	rylic Pol	ymers and E-C	ELL 01	the M	obility of	Soil-Ap	plied ¹⁴ C-Lat	eled H	lerbicid	es in L	lborator	y Leaching				
		tota	l recover	y, %		DOT	malized	ecovery."	%			total re	covery, 9			norm	alized re	covery," 9	2	
depth d	control	ASE- 108	93	CARBO	CELL	control	ASE- 108	-91 110	CARBO	E- CELL	CGA A	- CG	- CGA	- CGA-	-GGA- GGA-	CGA-	BGA-	-vo CGV	-GA-	-Vg
	38.6	43.0	37.0	413	37.4	47.5	51.2	45.7	46.9	Atrazin 42.3	e 38.1	38	3 37.4	36.1	33.8	42.6	46.3	46.6	42.9	41.0
30	28.5	30.4	27.2	29.3	27.3	35.1	35.4	32.8	33.3	30.9	8	8	1 25.0	27.9	26.2	29.3	34.6	31.2	33.1	30.6
5.0	11.8	9.7	15.1	14.2	19.5	14.6	11.3	18.2	16.2	22.7	19.	3 11.	5 14.0	15.9	18.1	21.6	14.9	17.5	18.9	21.9
0.7	2.0	1.6	2.5	2.8	3.8	2.5	1.9	3.1	3.2	4.3	ŭ	4	3.4	3.8	4.7	6.1	3.7	4.2	4.6	5.7
9.0	0.3	0.2	0.2	0.4	0.4	0.3	0.2	0.2	. 0.5	0.4	Ö	30.0	3.0.5	0.4	0.6	0.4	9.4	0.6	0.5	0.7
11.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ö	0 0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
total	81.3	86.7	82.9	88.0	88.4	100.0	100.0	100.0	100.0	100.0	ŝ	5 77.	2 80.2	84.2	82.4	100.0	100.0	100.0	100.0	100.0
LSD _{uot} =	7.30					$R_f = 0.22$	0.21	0.24	0.23	0.25	LSD _{0.06} = 7.3	0				$R_{l} = 0.27$	0.23	0.24	0.25	0.26
					-	LSD _{0.06} = 0.03										LSDag = 0.03				
										Metolach	dor									
1.0	43.9	74.1	50.5	45.9	47.8	58.9	76.7	63.6	58.1	61.2	61.	7 44.	1 47.2	62.6	44.8	68.7	58.0	62.6	67.0	56.4
3.0	22.8	19.2	24.1	25.4	2.6.2	30.6	19.5	30.4	32.2	32.2	22	0 22 0	21.5	24.6	26.6	24.5	33.2	29.0	26.4	33.5
6.0	6.8	3.5	4.2	6.4	4.4	9.1	3.6	5.3	8.1	5.6	4	9 5.	7 5.5	4.8	6.5	5.4	7.5	7.2	5.1	8.2
7.0	0.8	0.7	0.5	0.9	0.6	1.1	0.7	0.7	1.2	0.8	Ö	0 6	8 0.7	0.9	1:1	1.0	1.0	1.0	1.0	* 1
9.0	0.2	0.1	0.1	0.3	0.1	0.3	0.1	0.1	0.4	0.2	Ö		202	0.4	0.2	6.4	0.3	0.2	7 00	0.3
11.0	0.0	0.3	0.0	0.1	0.0	0.0	0.3	0.0	0.1	0.0	Ö	0	0.0	0.1	0.1	0.1	0.0	0.0	0.2	0.2
total	746	9.7.9	79.5	78.9	78.2	100.0	100.0	100.0	100.0	100.0	8	9 76.	0 75.4	93.4	79.4	100.0	100.0	100.0	100.0	100.0
LSD _{uos} =	420	2				$R_{f} = 0.19$	0.15	0.17	0.19	0.17	LSD _{0.06} = 4.2	0				$R_f = 0.16$ $1.5 \text{ Ber} = 0.01$	0.18	0.17	0.16	0.19
										Primisulf	non									
1.0	17.8	22.1	17.6	16.9	16.7	23.8	28.3	26.6	23.8	22.8	17.	2 19.	8 16.3	15.6	15.7	23.1	27.0	24.1	20.9	20.6
3.0	6.3	10.9	6.1	4.8	5.4	8.4	13.9	8.8	6.2	7.4	ים. ניים	ର ଜ	2	5.7	* 'L	1.1 1.1	11.6	8 i 9 i	7.6	9.8
5.0	13.6	16.2	12.7	7.9	9.6	18.2	20.7	17.6	11.2	13.4		6 I3.		10.0	12.2	10.4	18.4	19.7	13.6	0.61 16.0
7.0	18.9	16.4	16.2	15.5	17.5	26.4	21.0	23.6	21.9	23.9	8 <u>9</u> 73	6 I7.	1 15.1	18.5	1.71	24.9	23.2	72.4	8.17	9727
9.0	12.7	8.9	11.7	13.9	15.6	17.1	11.4	17.4	18.6	21.4	ŧ.	ה י ה י		101	14.0	20.02	191	A-01	0.12	7.61
11.0	6.3	3.7	6.1	12.3	8.1	1.7	4.7	7.4	17.4	1.11	Ċ.	ۍ ج		1.v	9.0	8.9	9.1	10.3	7.21	R.11
total	74.5	78.2	68.8	71.0	73.1	100.0	100.0	100.0	100.0	100.0	74.	6 73.	4 67.6	74.5	76.0	100.0	100.0	100.0	100.0	100.0
LSD _{uss} =	6.0				·	$R_f = 0.50$ LSD _{0.06} = 0.03	0.43	0.49	0.56	0.54	LSD ₀₀₆ = 5.	•				$R_{f} = 0.52$ LSD _{0.08} = 0.03	0.46	0.52	0.55	0.54
a Norma	alized to	100%	recover	ed, assun	aing equi	valent losses fro	m all de	spths, so	as to con	pare her	bicide mobilit	y and	effecta o	f polyme	ig.					

Herbicide/Polymer Bioactivity Study. The mean temperature for the period of the study (May 14-June 14, 1990) was 23 °C (normal) and total precipitation was 10 cm (normal). Weed and crop growth was excellent, and herbicide injury to the weeds from atrazine and metolachlor was above normal, while that from primisulfuron was generally below normal.

Herbicide/polymer interactions were observed throughout the experiment; however, there were no obvious trends (data not shown). ASE-108 and E-CELL polymers had little or no effect on the bioactivity of any of the herbicides. G-110 reduced the bioactivity of atrazine and metolachlor by 20–50% but had no effect on primisulfuron. CARBO reduced the bioactivity of atrazine by 18–20% but had no effect on metolachlor or primisulfuron.

CONCLUSIONS

Pesticides that are applied to plants or soils may be lost through photodecomposition, volatilization to the atmosphere, or leaching through the soil to groundwater or be carried off the soil in runoff water. If polymers could be added to pesticides to reduce their losses or movement and not detrimentally affect bioactivity, it would improve their efficacy and/or environmental safety. In a 92-day field lysimeter study on a Dothan soil, three differently charged water-soluble polymers [ASE-108 (anionic), HA-16 (nonionic), E-1242 (cationic)] added to three ionically different ¹⁴C-labeled herbicides [atrazine (weak base), metolachlor (nonionizable), primisulfuron (weak acid)] had no effect on the soil mobility or losses of ¹⁴C from the ¹⁴C-labeled herbicides. Apparently, the charge properties of the herbicides and polymers made little difference.

In a 3-day soil column laboratory study, nine nonionic or anionic polymers (ASE-108, G-110, CARBO, E-CELL, CGA-A, CGA-B, CGA-C, CGA-F, CGA-G) added to the three ¹⁴C-labeled herbicides had differing effects on soil mobility and/or losses of the chemicals. The most lipophilic of the CGA polymers, CGA-A, reduced the loss of metolachlor by 15% and reduced soil mobility by 15%but increased the soil mobility of atrazine by 23% and had no effect on primisulfuron. ASE-108 polymer reduced the loss and mobility of metolachlor by 22% and little or no effect (<10%) on atrazine or primisulfuron. CGA-F polymer also reduced the loss of metolachlor by 19% and soil mobility by 16% but had no effect on loss of the other herbicides and increased mobility of atrazine and primisulfuron by 12%. CGA-G and E-CELL polymers, two of the most hydrophilic polymers, increased the soil mobility of atrazine by 14-18% and that of primisulfuron by 8% but had little effect on metolachlor. The two polymers had no effect on losses of any of the three herbicides. CGA-B, CGA-C, G-110, and CARBO polymers had little or no effect on the loss or mobility of the three herbicides.

The observation that the ASE-108 polymer reduced the loss and soil mobility of [¹⁴C]metolachlor in the 3-day laboratory study but did not influence the loss or mobility of ¹⁴C from [¹⁴C]metolachlor in the 92-day field study suggests that the polymer may influence the loss and mobility of parent metolachlor but has no influence on degradation products of metolachlor. This is highly likely since the polymer has an influence on herbicide behavior only for the short time the two are in the immediate vicinity of one another. The polymer exerts little or no influence on the herbicide once the two are separated. The observation that the most lipophilic of the CGA polymers, CGA-A, had the most influence on reducing the loss and mobility of metolachlor and that the more hydrophilic polymers, E-CELL, CGA-B, and CGA-G, had no influence on metolachlor loss or mobility suggests that if more polymers are to be examined, they should probably be of the more lipophilic type. The herbicide/polymer field bioactivity study showed that the polymers had little or no effect on the bioactivity of the three herbicides. G-110 was the only polymer that consistently reduced the bioactivity of atrazine and metolachlor, but it had no effect on primisulfuron.

ACKNOWLEDGMENT

We acknowledge L. R. Swain, W. Baker, H. Marshall, G. Clark, C. R. Bogle, and J. U. Gonese for technical assistance. Products were provided by Ciba-Geigy Corp., Procter and Gamble Cellulose Co., B. F. Goodrich Co., and Rohm and Haas Co.

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Received for review June 24, 1992. Revised manuscript received November 16, 1992. Accepted March 22, 1993. Mention of products in this publication does not represent an endorsement of the authors or the North Carolina Agricultural Research Service or criticism of similar ones not mentioned.