Polymeric Microcapsules of the Herbicides Atrazine and Metribuzin: Preparation and Evaluation of Controlled-Release Properties

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Atrazine and metribuzin were microencapsulated within 5 different polymers by the solvent evaporation process using two different emulsifiers (for a total of 20 formulations). Herbicide efficacy studies on Florida beggarweed, smallflower morningglory, tall morningglory, and Palmer amaranth were conducted in the greenhouse. Nine of the 10 atrazine formulations were at least as effective as a commercial dry flowable formulation, and 5 exhibited superior herbicidal activity and controlled-release properties for over 32 weeks after treatment. The polymers demonstrating the most efficacy were cellulose acetate butyrate and ethylcellulose. Five of the 10 metribuzin formulations had activities comparable to or slightly less than the commercial formulation and exhibited controlled-release properties; the other 5 had moderate to low herbicidal activity.

INTRODUCTION

Recently, concern over the pesticide contamination of groundwater has mounted. Selected pesticides have been detected at extremely low levels in groundwater in isolated locations across the United States. In 1986, the U.S. Environmental Protection Agency disclosed that at least 17 pesticides used in agriculture has been found in groundwater in 23 states (Cohen et al., 1986). According to a 1988 interim report, 74 different pesticides have been detected in the groundwater of 38 states from all sources. Contamination attributable to normal agricultural use has been confirmed for 46 different pesticides detected in 26 states (Williams et al., 1988).

Research must be conducted to reduce the potential for groundwater contamination and improve public perception of agrichemicals in the environment (Schweizer, 1988). Microencapsulation is one method for obtaining this goal (Bahadir and Pfister, 1990; Seaman, 1990). Microencapsulated pesticides should be safer to handle, exhibit controlled-release properties (thus possibly reducing the total amount of pesticide used), and have reduced potential for leaching in the soil profile while maintaining effective biological activity.

The herbicides atrazine, metribuzin, and simazine have been frequently implicated in groundwater contamination (Cohen et al., 1986; Williams et al., 1988). Previously, we reported the preparation of β-cyclodextrin complexes of atrazine, metribuzin, and simazine and the evaluation of their efficacy as herbicides under greenhouse conditions (Dailey et al., 1990). The microencapsulation of atrazine by an interfacial polymerization process has been reported (Beestman and Deming, 1983). In addition, formulations of atrazine encapsulated within a starch matrix have exhibited promising controlled-release properties (Trimnell and Shasha, 1990; Carr et al., 1991). We are investigating a number of methods for the microencapsulation of pesticides. Among these is the solvent evaporation process which yields microcapsules of pesticide within a polymer matrix. The herbicides atrazine and metribuzin have been microencapsulated within cellulose acetate butyrate, ethylcellulose of two different viscosities, and low and medium molecular weight poly(methyl methacrylate) by the solvent evaporation process using two different emulsifiers. The preparation of microcapsules of atrazine or metribuzin using these polymers has not been reported previously. In this paper, we will describe the preparation of the polymeric microcapsules and the evaluation of their effectiveness in controlling weeds in the greenhouse, particularly with regard to controlled-release properties. The chief objectives of our research are to develop pesticide formulations that will maintain or increase efficacy on target organisms and that will not adversely impact on the environment, particularly groundwater.

MATERIALS AND METHODS

Chemicals and Reagents. Technical atrazine [mp 175–177 °C; lit. mp 176 °C (Hartley and Kidd, 1987)] was provided by Ciba-Geigy, Greensboro, NC. Technical Sencor (metribuzin), supplied by Miles, Inc., Kansas City, MO, was recrystallized from hexanes/chloroform, affording material of mp 128–130 °C or higher (lit. mp 125.5–126.5 °C) (Hartley and Kidd, 1987). Samples of the 88% hydrolyzed polyvinyl alcohols Airvol 205 (low viscosity) and Airvol 523 (medium viscosity) were provided by Air Products and Chemicals, Inc., Allentown, PA. Stock 0.5% solutions of Airvol 205 and 523 were prepared by adding the polyvinyl alcohol to the vortex of cold stirred water in a steady stream followed by heating at 85 °C for about 30 min. The following polymers were purchased from Aldrich Chemical Co., Inc.: cellulose acetate butyrate, butyryl content 17%, T_m 235 °C (CAB); ethylcellulose, ethoxyl content 48%, viscosity (5% solution in 80/20 toluene/ethanol) 22 cP (EC22); ethylcellulose, ethoxyl content 48%,

viscosity 100 cP (EC100); poly(methyl methacrylate), low molecular weight (PMML); and poly(methyl methacrylate), medium molecular weight (PMMM).

Preparation of Polymeric Microcapsules. In a typical microcapsule preparation, a solution of 2.50 g of herbicide (atrazine or metribuzin) and 10.0 g of polymer in 200 mL of dichloromethane was added slowly to the vortex of 1000 mL of a 0.25% Airvol 205 or 523 solution, stirred at 350 rpm. A Lightnin variable-speed high-torque mixer equipped with a 5.0-cm-diameter six-bladed turbine impeller was used for all stirring. When cellulose acetate butyrate and the ethylcellulose polymers were used, the dichloromethane was heated to 40 °C to effect complete dissolution. About five drops of 1-octanol was added to the stirred emulsion to reduce foaming. Stirring at 350 rpm was continued for 20–24 h, at which time evaporation of the organic solvent was complete. After the stirring was halted, the microcapsules were allowed to settle. The supernatant liquid (including floating solids) was decanted, 1000 mL of distilled water was added, and the mixture was stirred for 1–2 h. After settling, the microcapsules were filtered, allowed to air-dry, and finally dried in a vacuum desiccator until a constant weight was obtained.

In subsequent discussions, a polymeric microcapsule formulation will be referred to in abbreviated form, such as CAB-205, indicating the use of the polymer cellulose acetate butyrate and the emulsifier Airvol 205.

The herbicidal content of all the polymeric microcapsules prepared was determined by elemental analysis. On the basis of the amounts of materials used, each of the polymeric microcapsule formulations should contain 20% active ingredient. Determination of the herbicidal content of the CAB, EC22, and EC100-523 atrazine formulations was based upon nitrogen and chlorine microanalyses. The atrazine content of the PMML, PMMM, and EC100-205 formulations was determined from nitrogen microanalyses only. High values for the chlorine content of the PMML-523 and PMMM-523 formulations suggested the presence of residual dichloromethane. Herbicidal content of metribuzin formulations was calculated on the basis of nitrogen and sulfur microanalyses.

Greenhouse Studies. A 10-10-10 fertilizer was thoroughly mixed at the rate of 1000 kg/ha into an air-dried Tifton loamy sand top soil (fine-loamy, siliceous, thermic, Plinthic Paleudults) with 83% sand, 10% silt, 7% clay, and 1.0% organic matter and placed in 20 × 35 × 9 cm deep galvanized steel flats. The soil was then uniformly moistened by sprinkler from the top and allowed to equilibrate for 24 h. Either soybean for metribuzin or corn for atrazine and the selected weed species were then planted in rows 3 cm apart (15–20 seeds per row) in each flat. The flats were again lightly moistened with overhead sprinklers and the herbicides applied preemergence to crops and weeds. The commercial herbicide formulations were applied with an enclosed chamber sprayer using a Tee Jet 80067 flat fan spray tip, operating at 160 kPa, which delivered a volume of 187 L/ha at 0.45 m/s. Spray height was 46 cm. The controlled-release formulations were weighed for each individual flat, placed in a small paper envelope, and spread evenly over the soil surface by hand. The treated flats were placed in a greenhouse with day length maintained at approximately 14 h by natural or supplemental fluorescent lighting and the temperature ranging from 20 to 34 °C. The experimental design was a randomized complete block with four replications.

In greenhouse studies on the atrazine formulations CAB-523, EC22-523, EC222-205, EC100-523, and EC100-205, initial planting and herbicide treatment were done on May 2, 1990 (week 0). Herbicides were applied at 1.7 and 3.4 kg of ai/ha. Weed control was measured by comparison of the count of emerged plants in treated flats to that of emerged plants in the untreated check 20 days after each planting. The flats were then air-dried, the tops of dead plants carefully removed without disturbing the soil, and the same crops and weeds replanted 8, 16, 23, and 32 weeks after initial treatment to determine herbicide persistence or release.

In subsequent greenhouse studies involving the atrazine formulations CAB-205, PMML-523, PMML-205, PMMM-523, and PMMM-205, the initial planting date (and date of herbicide treatment) was October 30, 1990, and the weeds were replanted 6, 12, 16, and 24 weeks after initial treatment. In greenhouse studies involving all 10 metribuzin polymeric formulations, the initial planting date (and date of herbicide treatment) was October 30, 1990, and the weeds were replanted 6, 12, 16, 20, and 24 weeks after initial treatment as described above. The formulations were applied at the rates of 0.42 and 0.56 kg/ha, on the basis of pure metribuzin. Percent control of the weeds was determined 20 days after planting. In all of the above experiments, commercial herbicide formulations (atrazine 90 DF and metribuzin 75 DF) were used for comparison.

As anticipated, the lower rate of atrazine (1.7 kg/ha) and metribuzin (0.42 kg/ha) resulted in lower herbicidal activity and reduced length of activity. The controlled-release properties of each formulation were similar to the high rate applied. Therefore, data reported herein are for the higher application rates only.

The weeds smallflower morningglory [*Jacquemontia tamnifolia* (L.) Griseb.], tall morningglory [*Ipomoea purpurea* (L.) Roth], Florida beggarweed [*Desmodium tortuosum* (Sw.) DC], and Palmer amaranth (*Amaranthus palmeri* S. Wats) were chosen for their extensive occurrence in the southeastern United States and for their different levels of resistance to the herbicides used in these experiments (Weed Survey, 1989).

Statistical Analysis. Weed seeds were planted five or six successive times during each greenhouse study. The Julian date was determined for each planting in each study. Counting each day of the year produces Julian date values (i.e., December 31 = 365). Counting is continued into the next year if necessary. For each study, a mean Julian date was calculated. The first Julian date value was subtracted from each of the Julian date values including the mean Julian date. The mean was then subtracted from each date value thus obtained, and these new values were used in the regression analyses. The mean Julian date value is depicted in the figures as a vertical dotted line near the center of the graph. All of the data for each weed species at each of the counting times were analyzed using regression analysis techniques [PROC GLM (SAS, 1989)]. Intercept, slope, and curvature values were obtained from the regression analysis. Replication effects were also included in the regression model. If the number of weeds emerged exceeded the number known to be planted, the higher value was used to calculate the percentage of emerged weeds; otherwise, the number of seeds planted was used. Each of the regression components and their standard error (SE) were compared with each of the other treatments using the unequal n-unequal variance t-test (Steel and Torrie, 1960). The significance level was chosen to be $P = 0.05$. The t-test results were used to construct the multiple comparison letter arrangements appearing with a group of treatments. A line with an intercept and slope only is significantly different from a line with an intercept, slope, and curvature. For a line containing the curvature components, the slope value given is only for the intercept point.

RESULTS AND DISCUSSION

Overall, 9 of the 10 atrazine formulations were at least as efficacious as a commercial dry flowable formulation and 5 exhibited superior herbicidal activity and controlled-release properties for over 32 weeks after treatment. Formulations using ethylcellulose (22 cP) were more effective than those using ethylcellulose (100 cP). Formulations using low molecular weight poly(methyl methacrylate) were more effective than those using the medium molecular weight material. Formulations utilizing the polymers cellulose acetate butyrate and ethylcellulose were the most efficacious.

Five of the 10 metribuzin formulations exhibited controlled-release properties but were not quite as efficacious as the commercial formulation. Best results were obtained with cellulose acetate butyrate and ethylcellulose (100 cP).

The emulsifier used did not have any significant effect on the herbicidal activity of the polymeric formulation.

In the preparation of polymeric microcapsules of atrazine, some difficulties with agglomeration were encountered, especially with the poly(methyl methacrylate) polymers. With these polymers, the microcapsules formed

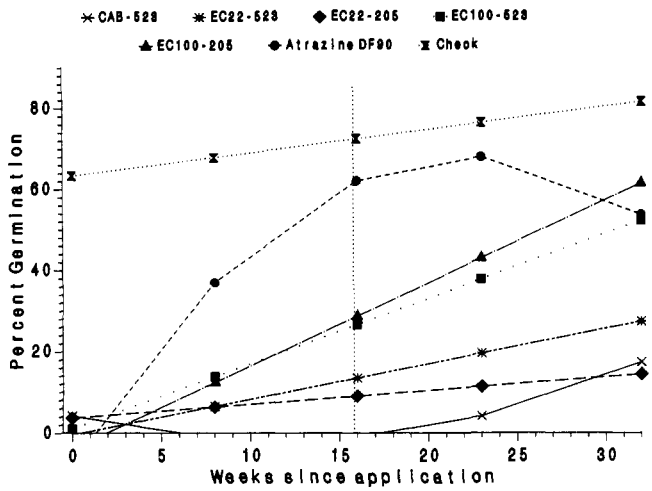

Figure 1. Effect of CAB and EC formulations of atrazine at 3.4 kg of ai/ha on Palmer amaranth germination for 32 weeks after application.

Table I. Statistical Summary of CAB and EC Formulations of Atrazine at 3.4 kg/ha on Palmer Amaranth Germination

formulation	inter-cept	SE	slope	SE	curva-ture	SE
CAB-523	−1.07	4.829 c	0.395	0.188 c	0.046	0.019 a
EC22-523	13.33	13.271 bc	0.872	0.594 abc		na
EC22-205	9.00	5.983 bc	0.337	0.268 c		na
EC100-523	26.33	10.940 b	1.603	0.489 ab		na
EC100-205	28.33	13.074 b	2.043	0.585 a		na
atrazine	61.68	16.696 a	1.970	0.648 a	−0.152	0.066 b
check	72.33	15.501 a	0.561	0.693 bc		na

teardrop-shaped aggregates during the latter stages of the solvent evaporation process. The isolated materials were pulverized for use in the greenhouse studies. In one instance (PMMM-205), the desired discrete microcapsules were isolated, but these results were not reproducible. Discrete microcapsules were isolated using cellulose acetate butyrate and the ethylcellulose polymers, but some agglomeration occurred (manifested by floating solids). This agglomeration was minimal with cellulose acetate butyrate and ethyl cellulose (100 cP), but with ethylcellulose (22 cP) the microcapsules isolated represented only 66% of the theoretical amount using Airvol 523 as emulsifier and only 24% using Airvol 205. In general, less agglomeration occurred using Airvol 523 as emulsifier. No agglomeration occurred in the preparation of polymeric microcapsules of metribuzin.

Microcapsules prepared using cellulose acetate and ethylcellulose varied in size from about 140 to 575 μm in diameter (typically, 350–400 μm); those prepared using poly(methyl methacrylate) ranged in size from about 50 to 100 μm in diameter.

All of the polymeric formulations were analyzed for herbicide content. Nine of the 10 atrazine formulations contained approximately 20% atrazine (the theoretical amount); formulation PMMM-205 contained only 11.4%. The herbicide content of the metribuzin formulations varied from 9.9% (EC100-523) to 14.8% (CAB-205). These low percentages may be attributable to the higher water solubility of metribuzin (1.2 g/L at 20 °C).

The polymeric atrazine formulations CAB-523, EC22-523, and EC22-205 significantly reduced Palmer amaranth germination as compared to EC100-523, EC100-205, atrazine 90 DF, and the untreated check (Figure 1 and Table I). All polymeric formulations exhibited some controlled-release properties as compared to atrazine 90 DF.

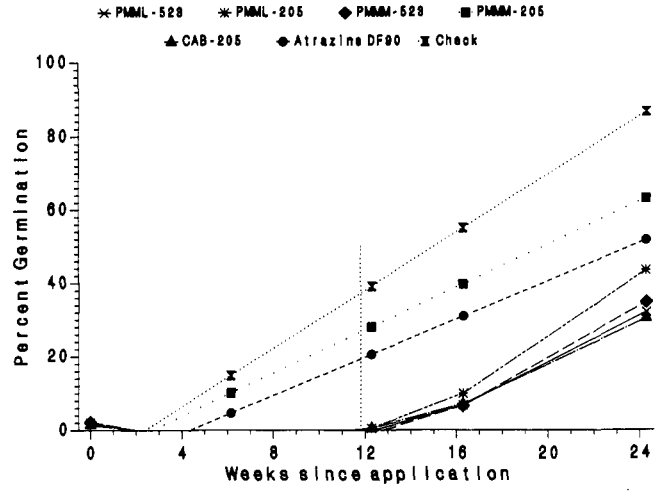

Figure 2. Effect of PMML, PMMM, and CAB formulations of atrazine at 3.4 kg of ai/ha on Palmer amaranth germination for 24 weeks after application.

Table II. Statistical Summary of PMML, PMMM, and CAB Formulations of Atrazine at 3.4 kg/ha on Palmer Amaranth Germination

formulation	inter-cept	SE	slope	SE	curva-ture	SE
PMML-523	−0.84	6.39 c	1.169	0.339 c	0.118	0.044 a
PMML-205	−0.33	9.39 c	1.609	0.497 bc	0.153	0.064 a
PMMM-523	−1.67	7.89 c	1.257	0.413 c	0.134	0.053 a
PMMM-205	26.67	11.10 ab	2.926	0.666 ab		na
CAB-205	−0.09	6.27 c	1.123	0.332 c	0.106	0.043 a
atrazine	9.67	11.67 bc	1.315	0.700 bc		na
check	37.33	11.41 a	3.966	0.684 a		na

PMML-523, PMMM-523, PMMM-205, CAB-205, and atrazine 90 DF were more effective than PMMM-205 in reducing Palmer amaranth germination (Figure 2). Results for the PMMM-205 formulation were highly variable, as indicated by the large standard error (Table II).

The effectiveness of CAB-523, EC22-523, EC22-205, EC100-523, EC100-205, and atrazine 90 DF for reducing smallflower morningglory germination is summarized in Figure 3 and Table III. Smallflower morningglory (Figure 3) was more tolerant to atrazine than Palmer amaranth (Figure 1). EC100-205 was significantly less active on smallflower morningglory than the other controlled-release formulations but not atrazine 90 DF. The CAB-523 formulation of atrazine reduced both smallflower morn-

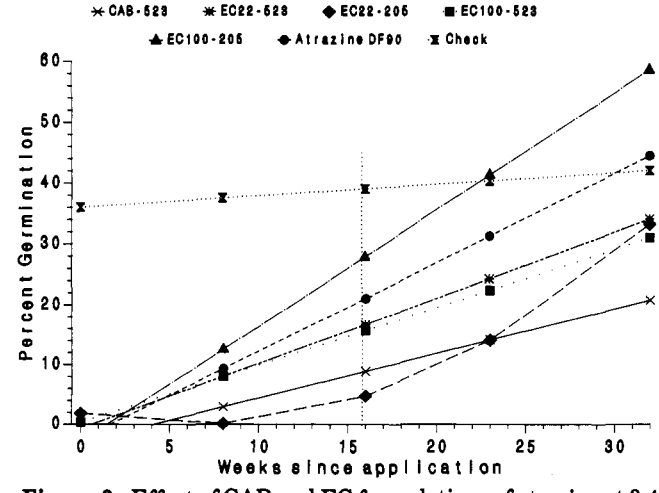

Figure 3. Effect of CAB and EC formulations of atrazine at 3.4 kg of ai/ha on smallflower morningglory germination for 32 weeks after application.

Table III. Statistical Summary of CAB and EC Formulations of Atrazine at 3.4 kg/ha on Smallflower Morningglory Germination

formulation	inter-cept	SE	slope	SE	curva-ture	SE
CAB-523	8.75	6.849 cd	0.746	0.306 cd		na
EC22-523	16.50	8.381 bcd	1.087	0.375 bc		na
EC22-205	4.56	5.810 d	0.961	0.226 c	0.050	0.023 a
EC100-523	15.50	5.035 cd	0.959	0.225 c		na
EC100-205	27.50	5.392 ab	1.919	0.241 a		na
atrazine	20.75	5.775 bc	1.466	0.258 ab		na
check	39.00	6.482 a	0.188	0.290 d		na

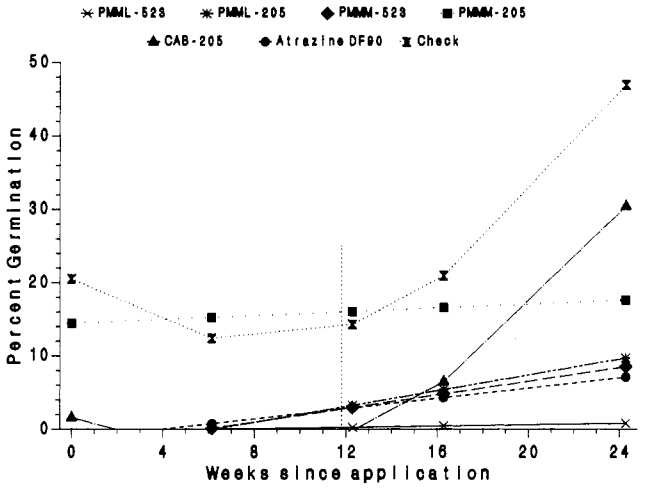

Figure 4. Effect of PMML, PMMM, and CAB formulations of atrazine at 3.4 kg of ai/ha on smallflower morningglory germination for 24 weeks after application.

Table IV. Statistical Summary of PMML, PMMM, and CAB Formulations of Atrazine at 3.4 kg/ha on Smallflower Morningglory Germination

formulation	inter-cept	SE	slope	SE	curva-ture	SE
PMML-523	0.25	0.48 a	0.045	0.029 b		na
PMML-205	3.00	4.67 a	0.538	0.280 a		na
PMMM-523	2.75	3.36 a	0.465	0.202 a		na
PMMM-205	16.00	10.85 a	0.132	0.651 ab		na
CAB-205	−0.74	6.34 a	1.163	0.336 a	0.111	0.043 a
atrazine	2.75	2.39 a	0.350	0.144 a		na
check	13.79	10.05 a	0.999	0.533 a	0.133	0.069 a

ingglory and Palmer amaranth germination more consistently than the other formulations.

The results of PMML, PMMM, and CAB-205 controlled-release formulations of atrazine on smallflower morningglory are shown in Figure 4 and summarized in Table IV. The PMML-523 formulation resulted in consistently low germination of smallflower morningglory as compared to the other herbicide formulations (Figure 4). PMMM-205 showed low initial activity on smallflower morningglory at the time of application but maintained its level of activity for the duration of the experiment.

All polymeric formulations of atrazine possessed some controlled-release properties (Figures 1–4). The PMMM-205 atrazine formulation generally resulted in the least herbicidal activity. The CAB-523 formulation generally was the most effective formulation. The other atrazine formulations in this experiment produced some variable results, depending on weed species, but were generally equally or more effective than the commercial 90 DF formulation for preventing germination of smallflower morningglory and Palmer amaranth at 24–32 weeks after application. Field corn was not injured by any of the herbicide treatments.

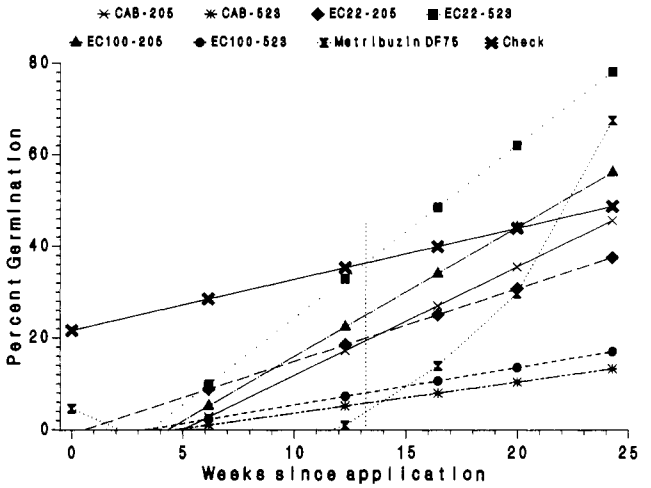

Figure 5. Effect of CAB, EC22, and EC100 formulations of metribuzin at 0.56 kg of ai/ha on Palmer amaranth germination for 24 weeks after application.

Table V. Statistical Summary of CAB and EC Formulations of Metribuzin at 0.56 kg/ha on Palmer Amaranth Germination

formulation	inter-cept	SE	slope	SE	curva-ture	SE
CAB-205	19.44	10.827 abc	2.365	0.659 abc		na
CAB-523	5.83	8.064 bc	0.678	0.491 d		na
EC22-205	20.00	10.333 abc	1.585	0.629 bcd		na
EC22-523	36.39	11.272 a	3.771	0.686 a		na
EC100-205	25.00	10.744 ab	2.814	0.654 ab		na
EC100-523	8.06	4.065 bc	0.814	0.248 d		
metribuzin	0.60	9.249 c	3.027	0.513 a	0.251	0.068 a
check	36.39	11.573 a	1.116	0.705 cd		na

In the greenhouse, the polymeric metribuzin formulations CAB-205, CAB-523, EC22-205, EC22-523, EC100-205, and EC100-523 showed activities comparable to or slightly lower than the commercial 75 DF formulation on Palmer amaranth (Figure 5 and Table V). There were extremely large experimental errors as indicated by calculated standard errors in Table V. The metribuzin formulations CAB-523 and EC100-523 had some controlled-release properties as compared to the other formulations (Figure 5).

The PMML and PMMM formulations of metribuzin on Palmer amaranth are depicted in Figure 6 and summarized in Table VI. Again, a large experimental error

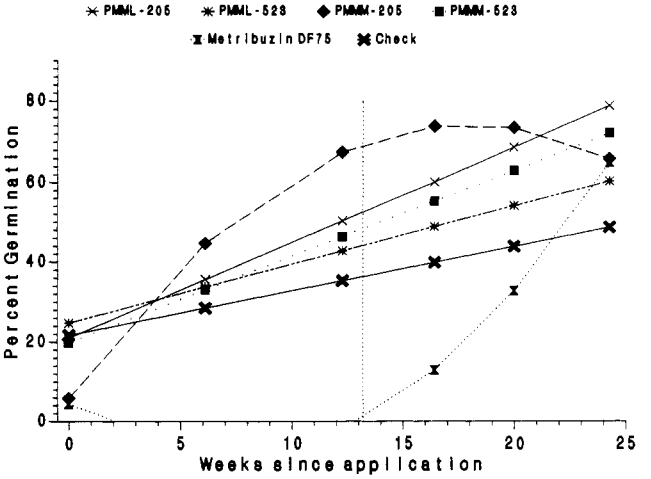

Figure 6. Effect of PMML and PMMM formulations of metribuzin at 0.56 kg of ai/ha on Palmer amaranth germination for 24 weeks after application.

Table VI. Statistical Summary of PMML and PMMM Formulations of Metribuzin at 0.56 kg/ha on Palmer Amaranth Germination

formulation	inter-cept	SE	slope	SE	curva-ture	SE
PMML-205	52.50	10.843 ab	2.390	0.660 ab		na
PMML-523	44.17	12.725 b	1.472	0.775 b		na
PMMM-205	69.56	10.961 a	2.033	0.608 ab	−0.212	0.081 b
PMMM-523	48.33	9.815 b	2.161	0.598 ab		na
metribuzin	0.60	9.249 c	3.027	0.513 a	0.251	0.068 a
check	36.39	11.573 b	1.116	0.705 b		na

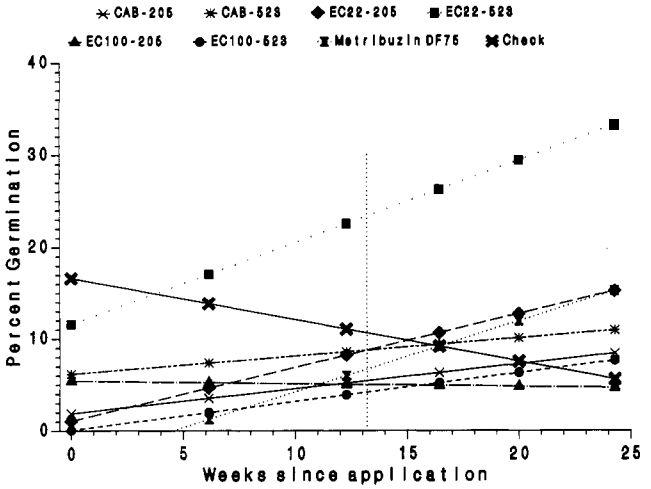

Figure 7. Effect of CAB, EC22, and EC100 formulations of metribuzin at 0.56 kg of ai/ha on smallflower morningglory germination for 24 weeks after application.

Table VII. Statistical Summary of CAB and EC Formulations of Metribuzin at 0.56 kg/ha on Smallflower Morningglory Germination

formulation	inter-cept	SE	slope	SE	curva-ture	SE
CAB-205	5.42	3.834 b	0.268	0.233 ab		na
CAB-523	8.75	4.440 b	0.195	0.270 ab		na
EC22-205	8.75	5.993 b	0.582	0.365 ab		na
EC22-523	23.33	7.675 a	0.891	0.467 a		na
EC100-205	5.00	2.871 b	−0.032	0.175 bc		na
EC100-523	4.17	2.441 b	0.311	0.147 ab		na
metribuzin	6.67	4.803 b	0.771	0.293 a		na
check	10.63	5.863 ab	−0.455	0.357 c		na

occurred, but there was little or no biological activity of metribuzin on Palmer amaranth from these formulations.

The activity of CAB, EC, PMMM, and PMML polymeric formulations of metribuzin on smallflower morningglory are shown in Figures 7 and 8 and statistically summarized in Tables VII and VIII. A large experimental variability resulted in little difference between herbicide formulations. Smallflower morningglory is moderately tolerant to metribuzin, which influenced the biological response. CAB-205, CAB-523, EC22-205, EC100-205, and EC100-523 did exhibit some early controlled-release properties as compared to the 75 DF formulation (Figure 7). The PMML and PMMM formulations exhibited no herbicidal activity on smallflower morningglory (Figure 8). Soybeans were not injured by metribuzin in any of the greenhouse studies.

Over the length of these studies, the germination and emergence of tall morningglory and Florida beggarweed were highly variable, resulting in inconsistent data. Although certain trends on herbicidal activity from the controlled-release formulations of atrazine and metribuzin were observed, data on tall morningglory and Florida beggarweed control are not included.

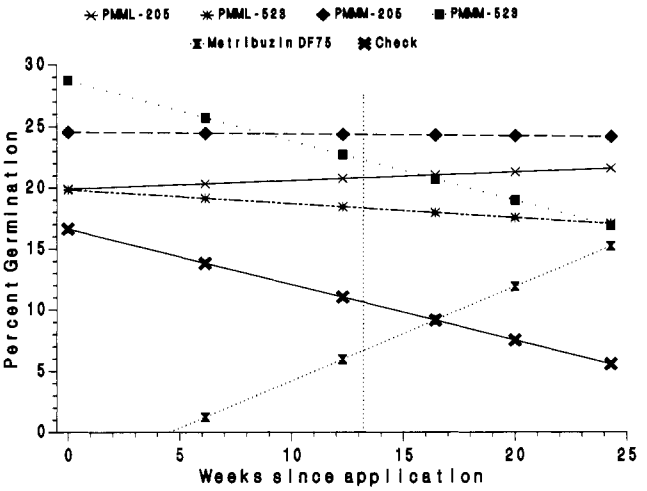

Figure 8. Effect of PMML and PMMM formulations of metribuzin at 0.56 kg of ai/ha on smallflower morningglory germination for 24 weeks after application.

Table VIII. Statistical Summary of PMML and PMMM Formulations of Metribuzin at 0.56 kg/ha on Smallflower Morningglory Germination

formulation	inter-cept	SE	slope	SE	curva-ture	SE
PMML-205	20.83	9.993 ab	0.070	0.609 ab		na
PMML-523	18.33	10.438 ab	−0.114	0.636 ab		na
PMMM-205	24.38	10.721 a	−0.014	0.653 ab		na
PMMM-523	22.29	11.577 ab	−0.487	0.705 b		na
metribuzin	6.67	4.803 b	0.771	0.293 a		na
check	10.63	5.863 ab	−0.455	0.357 b		na

In conclusion, we have reported for the first time the preparation of microcapsules of atrazine and metribuzin with the polymers cellulose acetate butyrate, ethylcellulose, and poly(methyl methacrylate). Microencapsulation of atrazine and metribuzin within cellulose acetate butyrate and ethylcellulose matrices has yielded formulations exhibiting excellent controlled-release properties which might reduce the potential for groundwater contamination.

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LITERATURE CITED

Bahadir, M.; Pfister, G. Controlled Release Formulations of Pesticides. In *Controlled Release, Biochemical Effects of Pesticides, Inhibition of Plant Pathogenic Fungi*; Haug, G., Hoffmann, H., Bowers, W. S., Ebing, W., Martin, D., Wegler, R., Eds.; Springer-Verlag: Berlin, 1990.

Beestman, G. W.; Deming, J. M. Encapsulation by Interfacial Polycondensation. U.S. Pat. 4,417,916, Nov 29, 1983.

Carr, M. E.; Wing, R. E.; Doane, W. M. Encapsulation of Atrazine Within a Starch Matrix by Extrusion Processing. *Cereal Chem.* 1991, *68*, 262–266.

Cohen, S. Z.; Eiden, C.; Lorber, M. N. Monitoring Ground Water for Pesticides. In *Evaluation of Pesticides in Ground Water*; Garner, W. Y., Honeycutt, R. C., Nigg, H. N., Eds.; ACS Symposium Series 315; American Chemical Society: Washington, DC, 1986; pp 170–196.

Dailey, O. D., Jr.; Dowler, C. C.; Glaze, N. C. Evaluation of Cyclodextrin Complexes of Pesticides for Use in Minimization

of Groundwater Contamination. In *Pesticide Formulations and Application Systems: 10th Volume, ASTM STP 1078*; Bode, L. E., Hazen, J. L., Chasin, D. G., Eds.; American Society for Testing and Materials: Philadelphia, 1990.

Hartley, D., Kidd, H., Eds. *The Agrochemicals Handbook*, 2nd ed.; The Royal Society of Chemistry: Nottingham, England, 1987.

SAS. *SAS/STAT User's Guide*, version 6, 4th ed.; SAS Institute: Cary, NC, 1989; Vol. 2, 846 pp.

Schweizer, E. E. New Technological Developments to Reduce Groundwater Contamination by Herbicides. *Weed Technol.* **1988**, *2*, 223–227.

Seaman, D. Trends in the Formulation of Pesticides—An Overview. *Pestic. Sci.* **1990**, *29*, 437–449.

Steel, R. G. D.; Torrie, J. H. *Principles and Procedures of Statistics*; McGraw-Hill: New York, 1960.

Trimnell, D.; Shasha, B. S. Controlled Release Formulations of Atrazine in Starch for Potential Reduction of Groundwater Pollution. *J. Controlled Release* **1990**, *12*, 251–256.

Weed Survey. *Proc. South. Weed Sci. Soc.* **1989**, *42*, 408–420.

Williams, W. M.; Holden, P. W.; Parsons, D. W.; Lorber, M. N. *Pesticides in Ground Water Data Base: 1988 Interim Report*; U.S. EPA, Office of Pesticide Programs, Environmental Fate and Effects Division: Washington DC, 1988.

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