Deposition and Retention of Chlorothalonil Applied to Peanut Foliage: Effects of Application Methods, Fungicide Formulations and Oil Additives[1]

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ABSTRACT

Deposition and retention of chlorothalonil (CTL) on peanut (*Arachis hypogaea* L.) foliage was evaluated by surface-stripping leaf discs with toluene and determining the CTL concentration via gas chromatography. CTL was applied at 1.25 kg/ha via ground sprays, a center pivot-mounted underslung boom, or chemigation in 0.12, 1.7, or 17.8 kL of water/ha, respectively. Ground sprays resulted in the highest concentrations of CTL on peanut leaves followed by the underslung boom and chemigation, respectively. Residue levels were lower with the higher volume applications but were more uniformly distributed throughout the plant canopy. Deposition of chemigated CTL applied as Bravo 500(R), Bravo 720(R), or Bravo 720(R) plus either an emulsifiable vegetable oil (SoyOil 937(R) or a nonemulsifiable petroleum oil (11N Sunspray oil(R)) was also evaluated. Results of residue sampling at 0, 5, 9 and 14 days after treatment indicated that addition of either oil to Bravo 720(R) resulted in the highest initial deposition of fungicide followed by Bravo 500(R) and Bravo 720(R) alone, respectively. However, the half-life of CTL applied as Bravo 720(R) plus either oil was reduced as compared to the half-life for either Bravo 720(R) alone or Bravo 500(R). By day 14, concentrations of CTL had decreased by more than 93% in all treatments. The mean half-life of CTL for all treatments was 3.8, 4.8 and 4.8 days in the top, middle and bottom canopy layers, respectively.

Key Words: Chemigation, chlorothalonil, fungicide residues, peanut leafspot

Chlorothalonil (tetrachloroisophthalonitrile) is a broad spectrum protectant fungicide used to control foliar diseases of numerous crops. In the southeastern United States, peanut (*Arachis hypogaea* L.) growers use an average of 5-8 applications per year to control foliar diseases such as late leafspot (*Cercosporidium personatum* (Berk. & Curt.) Deighton), early leafspot (*Cercospora arachidicola* Hori), rust (*Puccinia arachidis* Speg.) and other potentially damaging diseases. These applications are made by fixed-wing aircraft, various types of tractor-propelled boom sprayers, or via injection into a sprinkler irrigation system (16). Spray volumes with these systems range from 0.03 to more than 125 kL of water/ha. A treatment interval of 10-14 days is recommended depending on weather conditions and disease pressure.

Chlorothalonil has proven to be a versatile fungicide for peanut disease management in that it can be applied with many delivery systems and still provide good control. However, applications utilizing high volumes of water applied by center pivot irrigation systems (chemigation) have given less consistent control of peanut leafspot (1, 3). This was particularly evident in a 1987 study utilizing a new formulation of chlorothalonil, Bravo 720(R) (3). Although in 1987 there was an unusually severe leafspot epidemic, the question still arose as to the efficacy of Bravo 720(R) compared to Bravo 500(R) when applied via chemigation. Different formulations of chlorothalonil have shown various levels of activity against foliar peanut diseases when applied conventionally (2, 15).

An alternative pesticide delivery system consisting of a separate spray boom mounted beneath a center pivot system offers many of the advantages of chemigation but delivers only about 10% of the water volume. It has been used to apply chlorothalonil to peanuts and potatoes, and in both crops has resulted in disease control superior to that of chemigated applications (3, 19). One such system is the Pivot Agrichemical Spray System (PASS) which is being developed by Garvey Irrigation Consultants, Lenox, GA 31637.

Determinations of chlorothalonil deposition on peanut leaves have been conducted. Littrell (11) used residue analysis to compare chlorothalonil deposition from controlled droplet applicators and conventional boom sprayers, concluding that it was a useful tool for evaluating application methods. He later reported that these conventional application methods produced significantly higher chlorothalonil residues than did chemigated applications but did not give actual concentrations (10). Other research has indicated that application of chlorothalonil to potato foliage via con-

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ventional boom sprayers also results in higher residues than does chemigation (12).

Various methods have been used to increase the efficacy of pesticides applied via chemigation. One approach that has shown promise involves the use of pesticides formulated in oil, preferably without or with minimum emulsifiers (20). Alternatively, oil can be added to commercially formulated pesticides containing emulsifiers prior to injection in the irrigation water. The addition of a nonemulsifiable petroleum oil to chemigated tebuconazole, an experimental ergosterol biosynthesis inhibitor, resulted in better control of foliar peanut diseases than when applied in the irrigation water without the oil (4).

The objectives of this research were to evaluate i) initial and subsequent concentrations of chlorothalonil on peanut foliage from applications via conventional boom sprayers, chemigation and a Pivot Agrichemical Spray System, ii) the relative deposition from two chlorothalonil formulations, Bravo 500(R) and Bravo 720(R), when applied via chemigation, and iii) the effects of oil additives to Bravo 720(R) on foliar residues when applying it via chemigation. A preliminary report has been published (5).

Materials and Methods

Sampling Procedures. All peanut leaf tissue was collected directly from plants in the field to avoid loss of residue from abrasion or handling. Leaves were hand picked at random and a mechanical punching device used to remove a 12-mm-dia. disc from each one. Leaf discs were automatically deposited in a 20-mL scintillation vial attached to the punch. The top, middle and bottom third of the plant canopy were sampled separately with leaves coming from both mainstems and lateral limbs. A total of 10 discs were collected per plot per canopy layer. Immediately after sampling, vials were taken to the laboratory and 10 mL of toluene added to each. They were then stored in the dark at room temperature until shipment to Ricerca, Inc., Painesville, OH for analysis.

Samples were analyzed as described previously (18) with an automated sampling system utilizing a Varian 3700 gas chromatograph equipped with dual Ni electron capture detectors, electrometers, and Varian Model 8000 autosamplers. Since chlorothalonil is not systemic, residues were expressed as planimetric mass density (μg/cm^2) of leaf surface area based on the concentration of chlorothalonil in the toluene solvent.

Experiment One - Application Method. Plots in Experiment one consisted of a single two-row bed of Florunner peanuts (7.6 x 1.8 m in 1987 and 6.1 x 1.8 m in 1988). Plots were separated by two border rows and 2.1 m fallow alleys. A completely randomized design with four replications was used. The test was located in one quadrant (0.15 ha) of a single-tower center pivot irrigation system and repeated in 1988 in an adjacent quadrant. Planting date was May 18 both years and standard management recommendations of the Georgia Cooperative Extension Service were followed (8).

The fungicide applied was chlorothalonil (Bravo 720(R)) at 1255 g a.i./ha on a 14-day schedule initiated in the fifth week after planting (seven total applications). Treatments were: i) nonsprayed control, ii) chemigation, iii) pivot-mounted underslung boom, and iv) conventional boom sprayer. Fungicides applied by chemigation or underslung boom were diluted 1:3.7 (v:v, fungicide: water) prior to injection. The underslung boom applied 1.7 kL water/ha via FloodJet(R) nozzles (Tk2 to Tk24) and the chemigated treatments were applied in 17.8 kL water/ha via E53 WhirlJet(R) nozzles (Spraying Systems Co., Wheaton, IL). Plots not being treated during chemigation were covered with either plastic sheets or elevated fiberglass shelters. Ground sprays were applied with a CO_2-pressurized backpack sprayer with three disc-core (D2-13) nozzles per row delivering 124.4 L/ha at 345 kPA.

In 1987, leaf discs for residue analysis were collected just before and after the fourth and seventh fungicide applications. The foliage was allowed to dry before the posttreatment samples were collected. In 1988, leaf discs were collected only before and after the fifth application.

Experiment Two - Formulation Evaluation. Fungicides were applied with an irrigation simulator equipped with E53 WhirlJet(R) nozzles and designed to reproduce conditions present during chemigation with a center pivot irrigation system. This apparatus has been described previously (17). Florunner peanuts were planted as described above and plots were 2.5 beds wide (5 rows) by 10.7 m long in a randomized complete block design with four replications. Residue samples were collected only from the middle row of each plot from each of three canopy layers.

The following treatments were used: i) Bravo 720(R), ii) Bravo 720(R) plus an emulsifiable vegetable oil (SoyOil 937(R), Coastal Chemical Co.), iii) Bravo 720(R) plus a nonemulsifiable petroleum oil (11N Sunspray oil(R), Suntech Inc.), and iv) Bravo 500(R). All treatments contained an equivalent amount of chlorothalonil (1.26 kg a.i./ha) and applications were made in 20.3 kL water/ha. Prior to injection, Bravo 720(R) was mixed thoroughly with water, SoyOil 937(R) or 11N Sunspray oil(R) at a ratio of 1:1.8 (v/v, fungicide:diluent). The Bravo 500(R) was diluted with water to achieve a total volume equivalent to the Bravo 720(R) mixes. Plots were uniformly sprayed with copper hydroxide (2.58 kg/ha) applied by conventional ground sprayer prior to initiation of the experiment. Chemigation treatments were applied after the peanut plants had developed a full canopy (August 3). Foliar samples for residue analysis were collected as described previously before and after application of treatments as well as 5, 9 and 14 days after treatment.

Experimental data were tested by analysis of variance (ANOVA) and means were separated using Waller-Duncan k-ratio t-test ($P \leq 0.05$). Regression analysis was used to evaluate the decline in foliar residue concentrations over time in the second experiment (14).

Results

Experiment One - Application Method. The three trials were considered to be a random effect and the combined data set analyzed accordingly. The pretreatment and posttreatment residue levels were analyzed separately.

The ANOVA for pretreatment levels of chlorothalonil indicated that treatment was the only significant ($P \leq 0.01$) factor and therefore data were combined for all canopy layers. Results indicated that at the time of first sampling, i.e., two weeks after the previous application, only conventional ground-sprayed plots had foliar residue levels significantly ($P \leq 0.05$) greater than nontreated plots. Actual concentrations were found to be 2.44, 0.35, 0.08 and 0.06 μg/cm^2 for leaves from plots treated via conventional boom sprayers, pivot-mounted underslung boom, chemigation and nontreated plots, respectively.

The ANOVA for posttreatment levels of chlorothalonil indicated that treatment, canopy layer, and the treatment by canopy layer interaction were all significant ($P \leq 0.01$). Therefore, treatment means were compared within canopy layers. In all cases, the planimetric density of chlorothalonil on foliage was highest in the ground sprayed plots followed by the PASS applications and chemigation, respectively (Table 1). This difference was significant in all canopy layers

Table 1. Planimetric density (μg/cm^2) of chlorothalonil on peanut foliage immediately after treatment with three delivery systems.

Delivery system and volume of water applied (kL/ha)[a]	Canopy Layer		
	Top	Middle	Bottom
1. Ground Spray (0.12)	9.63 A[b]	7.51 A	2.67 A
2. Underslung Boom (1.7)	2.56 B	1.70 B	1.45 AB
3. Chemigation (17.8)	0.35 C	0.49 B	0.53 B
4. Nontreated	0.08 C	0.04 B	0.03 B

[a] The treatment applied in all cases was chlorothalonil (1.26 kg/ha) supplied as Bravo 720(R).

[b] Numbers in a column with the same letter are not significantly different according to Waller-Duncan k-ratio t-test ($P \leq 0.05$).

except the bottom where residues in ground sprayed plots were statistically equivalent to the PASS applications. Chlorothalonil concentrations in plots treated by the PASS system were higher than those in chemigated plots only in the top canopy layer, and residues in chemigated plots were never significantly ($P \leq 0.05$) higher than the nontreated plants which apparently received low levels of spray drift (Table 1).

To analyze the quantity of chlorothalonil actually deposited with a single application, the pretreatment residues were subtracted from the posttreatment residues found for each plot. Obviously all concentrations are lower than the actual posttreatment residue levels, but the same trends are apparent in that the observed residue density was indirectly related to the concentration of chlorothalonil in the applied spray suspension (Table 2). In the bottom canopy layer no significant differences in residues were found among treatments.

Table 2. Planimetric density (µg/cm²) of chlorothalonil actually deposited in one application to peanut foliage via three delivery systems (i.e. posttreatment residues - pretreatment residues).

Delivery system and volume of water applied (kL/ha)[a]	Canopy Layer Top	Middle	Bottom
1. Ground Spray (0.12)	6.74 A[b]	4.43 A	1.33 A
2. Underslung Boom (1.7)	2.28 B	1.14 B	1.22 A
3. Chemigation (17.8)	0.27 C	0.43 B	0.44 A
4. Nontreated	0.01 C	-0.02 B	-0.02 A

[a] The treatment applied in all cases was chlorothalonil (1.26 kg/ha) supplied as Bravo 720(R).

[b] Numbers in a column with the same letter are not significantly different according to Waller-Duncan k-ratio t-test ($P \leq 0.05$).

Experiment Two - Formulation Evaluation. The use of copper hydroxide for the early season sprays served to protect the foliage from leafspot while developing a full canopy of leaves without chlorothalonil residues. This was verified by a pretreatment sample indicating that no foliar chlorothalonil residues were present.

Analysis of residues immediately following treatment showed a significant ($P \leq 0.01$) treatment effect. Bravo 720(R) plus either SoyOil 937(R) or 11N Sunspray oil (R) resulted in higher levels of deposition than either Bravo 720(R) alone or Bravo 500(R). There was also a significant treatment by canopy layer interaction and analysis by layers demonstrated that deposition among treatments was different only in the top layer. Mean residue levels were 0.50, 1.22, 1.46 and 0.61 µg/cm² for Bravo 720(R), Bravo 720(R) + SoyOil 937(R), Bravo 720(R) + 11N Sunspray oil (R), and Bravo 500(R), respectively (LSD = 0.38, $P \leq 0.05$).

By day 5, significant differences were detected for both treatment and canopy layer. Mean residue levels were 0.44, 0.29, 0.25 and 0.38 µg/cm² for Bravo 720(R), Bravo 720(R) + SoyOil 937(R), Bravo 720(R) + 11N Sunspray oil (R), and Bravo 500(R), respectively (LSD = 0.15, $P \leq 0.05$). Mean residue levels among canopy layers were 0.22, 0.39 and 0.42 µg/cm² for the top, middle and bottom, respectively.

By day 9, treatment was the only significant ($P < 0.01$) effect with means of 0.10, 0.16, 0.04 and 0.29 µg/cm² for Bravo 720(R), Bravo 720(R) + SoyOil 937(R), Bravo 720(R) + 11N Sunspray oil (R) and Bravo 500(R), respectively (LSD = 0.11, $P \leq 0.05$). By day 14, there were no significant differences for any of the variables and chlorothalonil residues in all treatments decreased to nearly zero (all were ≤ 0.08 µg/cm²).

The common logarithms of the percentage of chlorothalonil initially deposited which remained on peanut foliage over time fit a linear model for each treatment with correlation coefficients of 0.91-0.99 (Table 3). Half-lives tended to be shorter in the top canopy layer for all treatments. The addition of either oil to Bravo 720(R) apparently decreased the persistence of chlorothalonil on the foliage as indicated by reduced half-lives in all three canopy layers (Table 3). The persistence of chlorothalonil from all formulations in each canopy layer is shown in Fig. 1.

Table 3. Persistence of chlorothalonil applied via irrigation simulator to peanut foliage in three canopy layers (common log of percent chlorothalonil remaining as a function of X = time in days).

Treatment and Canopy Layer	Linear Regression	Correlation Coefficient	Half-life (Days)
Bravo 720(R)			
Top	Y=-0.101X + 2.138	0.97	4.35
Middle	Y=-0.085X + 2.174	0.92	5.59
Bottom	Y=-0.091X + 2.233	0.92	5.86
Bravo 720(R) + SoyOil 937(R)			
Top	Y=-0.124X + 2.078	0.99	3.07
Middle	Y=-0.110X + 2.147	0.97	4.09
Bottom	Y=-0.105X + 2.198	0.91	4.76
Bravo 720(R) + 11N Sunspray oil(R)			
Top	Y=-0.145X + 2.063	0.98	2.52
Middle	Y=-0.122X + 2.174	0.98	3.89
Bottom	Y=-0.134X + 2.155	0.91	3.40
Bravo 500(R)			
Top	Y=-0.061X + 2.016	0.92	5.19
Middle	Y=-0.077X + 2.140	0.91	5.72
Bottom	Y=-0.106X + 2.229	0.92	5.02

The maximum air temperature during the course of this study was 35.6 C and the minimum was 20.6 C. The mean daily maximum temperature was 33.8 and the mean daily minimum was 21.7 C. The only rainfall was 2.54 and 1.78 cm on days 12 and 13 after treatment, respectively.

Discussion

In the application methods test, ground sprays resulted in the highest concentration of deposited chlorothalonil, but differences between methods were less in the middle and bottom canopy layers. This indicates that although total deposition is higher with conventional ground sprays than with the other delivery methods, penetration of the canopy is reduced due to the relatively small spray volume (0.12 versus 1.7 or 17.8 kL water/ha). Previous studies demonstrated this same effect with conventional boom sprayers and found that controlled droplet applicators applying only 9.4 L of spray/ha provided even less canopy penetration (11).

Although chemigation resulted in less deposition of chlorothalonil than the other methods, the differences were not as great as might be anticipated on the basis of spray volume alone. For example, the amount of water used to chemigate was 148 times the amount used for ground sprays. Therefore, assuming that most of the water for chemigation runs off of the plants, it might be theorized that there should be 148 times more chlorothalonil deposited with the ground spray than by chemigation. In this experiment, however, the actual differences were only 27.5, 15.3 and 5.0 fold for the upper, middle and lower canopy layers, respectively. The same trend is also seen with the PASS application where the volume difference of application water was 14.2 fold that of ground sprays and the actual differences in deposition were 7.3, 3.5 and 2.7 fold the upper, middle and lower canopy layers, respectively. Apparently there is a selective partitioning of chlorothalonil to the leaf surface when it is applied in higher volumes of water.

Results of experiment two indicated that chlorothalonil deposition on peanut foliage did vary with formulation and was influenced by the addition of oil prior to application. Both the nonemulsifiable (11N Sunspray(R)) and the emulsifiable (SoyOil 937(R)) oil increased the initial deposition of chlorothalonil, particularly in the upper canopy. However, those residues apparently had less affinity for the peanut foliage and decreased at a higher rate than residues resulting from application with either commercial formulation alone as evidenced by their respective half-lives (Table 3). This may be due to detrimental interactions between the oils and the formulation adjuvants which could enhance weathering of chlorothalonil residues. It may also be postulated that co-application of oil with chlorothalonil results in enhanced formation of conjugates or complexes which reduce the availability of chlorothalonil *per se* to the stripping solvent (toluene) used in this study. Regardless of the reason, the apparent benefit of either oil on initial deposition may well be negated by the reduced half-life of the fungicide. Efficacy data are needed to fully answer this question, although research by Cu et al. (7) demonstrated that SoyOil 937(R) can increase the efficacy of Bravo 720(R) applied by ground sprayer.

The shorter half-life of residues in the upper canopy may be due to continued plant growth resulting in preferential dilution of chlorothalonil mass by substrate foliage mass. Heat was also important since temperatures > 18 C have been shown to decrease chlorothalonil residues (6), and air temperatures in our study ranged from 20.6 to 35.6 C. Although not documented, temperatures in the lower canopy

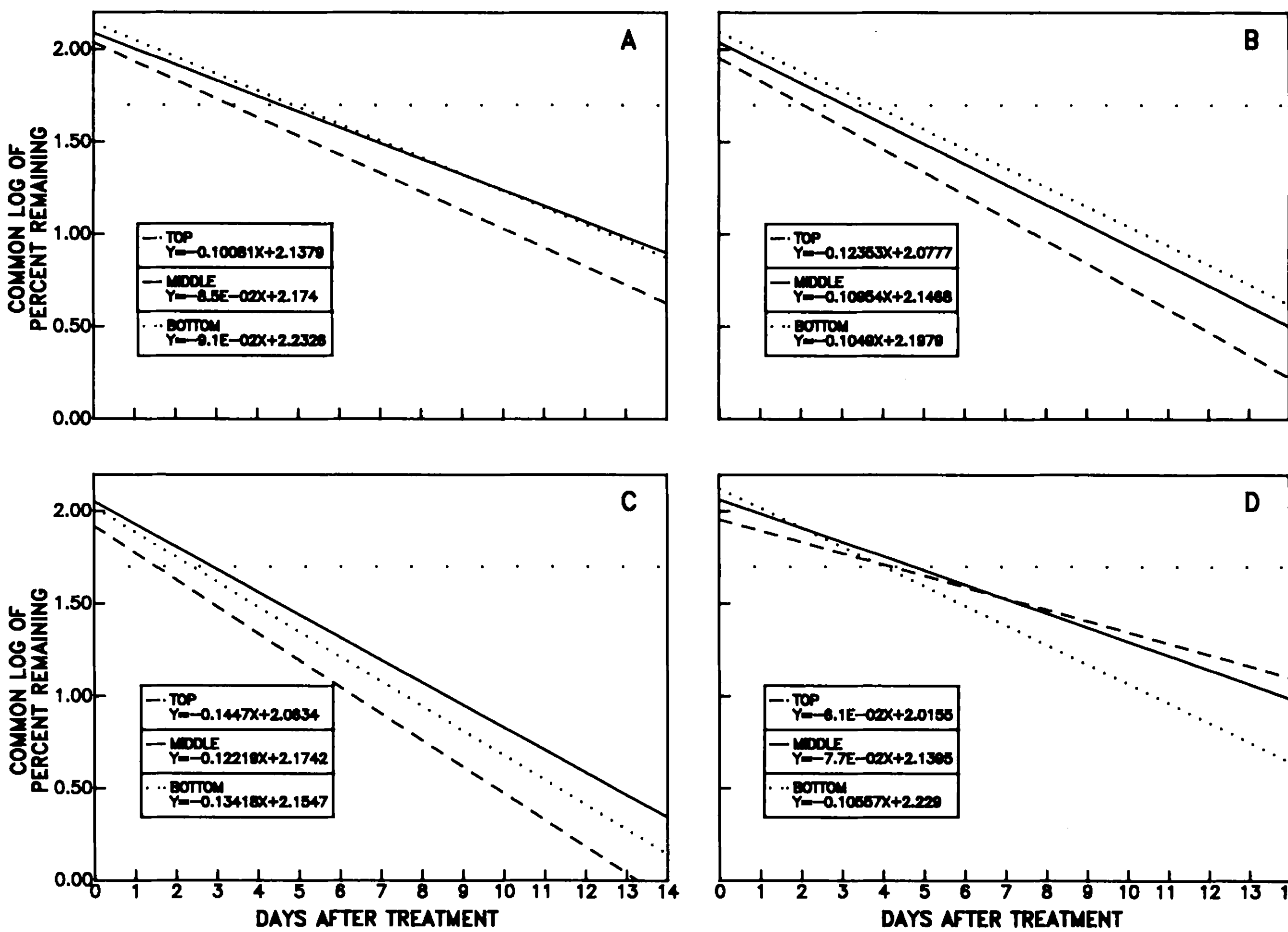

Fig. 1. Retention in three different canopy layers of chlorothalonil applied to peanut foliage as A. Bravo 720(R), B. Bravo 720(R) plus SoyOil 937(R), C. Bravo 720(R) plus 11N Sunspray oil (R), or D. Bravo 500(R) (. . . . indicates 50% remaining, i.e. log = 1.69897).

were probably lower than those at the top of the plant.

Shorter half-lives in the upper canopy may also be attributable to redistribution of chlorothalonil from the upper to the lower canopy levels. Although no rain fell until 12 days after application, heavy dews are often present in south Georgia peanut fields and may have provided enough water to redistribute chlorothalonil. A previous investigation found the half-life of chlorothalonil on potato foliage to be 3.6 and 21.3 days in the top and bottom canopy layers, respectively (6). Other work has shown the half-life of chlorothalonil to range from 1.2 to 2.6 days on passion fruit leaves (9), and Neely (13) showed fungistatic levels of chlorothalonil to persist on various hosts for an average of 2.8 weeks.

The initial density of chlorothalonil residues from chemigation applications of Bravo 720(R) tended to be lower than similar applications of Bravo 500(R). This indicates that the 720 g/L formulation may be less well suited for chemigation applications than the 500 g/L formulation. This does not occur with conventional boom sprayer applications. In fact, comparisons of deposition and retention of chlorothalonil for 13 days after applying the two formulations with ground sprays (124.4 L/ha) consistently showed higher residue levels from Bravo 720(R) than from Bravo 500(R), although these differences were not significant ($P \leq 0.05$).

The exact density of chlorothalonil needed on peanut foliage to prevent infection by *Cercosporidium* spores is not known. It would no doubt vary depending upon genotype, growth stage, environmental conditions, uniformity of distribution of fungicide on the leaf surface, etc. Several disease ratings were taken for both years of experiment one and demonstrated that applications via chemigation were adequate in years of light disease severity but could fail totally when the environment is conducive for rapid disease development (3). PASS applications were much more effective but were less consistent than the ground sprays which provided good disease control both years. Extrapolating from these data, it would seem that approximately 1-2 μg of chlorothalonil per cm^2 of leaf area (if deposited uniformly) is required to ensure protection of Florunner peanut foliage.

Calculations relating efficacy and residue levels fail to account for secondary effects of the fungicide on nontarget diseases or even on inoculum of *C. personatum*. Initial inoculum for peanut leafspot comes from plant debris in the soil. Chlorothalonil is toxic at very low concentrations to *C. personatum* conidia and chemigated applications could serve to delay disease development. Other studies are needed to verify this. Backman (1) suggested that chemigated treatments may also give better coverage of fungicide on the lower surface of the leaf due to the high volume of water used. This greater uniformity of deposition could compensate somewhat for the lower areawise density of residues deposited by chemigation as compared to ground sprays. The data presented here help to explain some of the disease control results observed in the field and provide clues for improvement of chemigation techniques through manipulation of water volumes and fungicide formulations.

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