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Removal of Alachlor Residues from Contaminated Clothing Fabrics*

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The effect of fabric weight, pesticide type, and three laundering variables on removal of alachlor and fonofos from 100% cotton fabrics was reported in our previous paper (Kim et al. 1982). Since the laundering procedures used in the work did not result in complete removal of pesticide residues, there was a need to explore additional fabric and fabric-care variables, on the basis that pesticide is a particular type of soil. The objectives of this research were to determine the effects of fiber content, treatment with prewash materials, detergent type, wash-water temperature, and drying method on removal of alachlor.

Soiling is a complex phenomenon involving the interrelationship between the properties of the fiber, fabric structure, and soil composition. According to Easter (1983), soil content (pesticide type) played an important role in controlling fabric/soil interaction: an aqueous suspension of captan particles was more difficult to remove from 100% cotton fabrics, whereas an oil-based formulation of guthion was more difficult to remove from Gore Tex, a predominantly nylon (oleophilic) protective-clothing fabric. Polyester/cotton blended fabrics are often worn by pesticide applicators; however, published reports show contradicting results as to the effect of fiber content on residue retention. Finley and Rogillio (1969) reported that residue retention of methyl parathion and DDT increased with cotton fiber content, whereas Easley et al. (1981) stated that fiber content made no difference in the efficacy of methyl parathion removal.

Pretreatment in laundering has been shown to reduce the pesticide residues (Easley et al. 1981; Kim et al. 1982; Easley et al. 1983) although the most effective pretreatment for removing a certain pesticide has not been determined. Water alone (rinse wash

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without a detergent) removed from 19.3% to 93.1% of alachlor (Kim et al. 1982), indicating the effectiveness of water-rinse treatment. However, an organic solvent having a matching cohesive energy density should dissolve an oil-based emulsifiable-concentrate formulation more effectively. Alcohol and perchloroethylene, which have been used commercially as spot-cleaning or drycleaning solvents, might be effective as a pretreatment for an oil-based emulsifiable-concentrate formulation.

The chemical interaction between the pesticide and the detergent may play a key role in the effective removal of chemical soils. Previous research on detergent effectiveness has been inconclusive. Easley et al. (1982 a) were not able to confirm the hypothesis that increased alkalinity of the laundering solution resulted in a greater removal of methyl parathion, but there seemed to be an interaction between the phosphate detergent and the wash-water temperature. Later the same researchers reported that heavy-duty liquid detergent without phosphate was more effective than the phosphate detergent in removal of 2,4-D ester and 2,4-D amine (Easley et al. 1983). The detergent mentioned most often by Iowa pesticide-applicator families for use in laundering pesticide-soiled clothing is another phosphate detergent, according to an unpublished survey report.

Another laundering variable studied by researchers with varying results is wash-water temperature. Lillie et al. (1981) observed a tendency for increased residue removal of a number of pesticides (except for chlordane) from 100% cotton fabrics with increased temperature, although the data were not statistically significant. Easley et al. (1982 a) reported no significant difference between hot (60^{o}C) and warm (49^{o}C) washes, whereas cold (30^{o}C) wash produced significantly lower removal of methyl parathion. Later, they noted similar results with 2,4-D ester, but with 2,4-D amine they did not observe significant differences between the hot and cold washes, indicating that the effect of wash-water temperature was dependent on the pesticide type (Easley et al. 1983). They attributed the difference to the difference in solubility of the pesticide. While noting an overall increase in the removal of captan and guthion with an increase in temperature, Easter (1983) reported interactions between the wash-water temperature and the pesticide type, fiber content, and fabric structure. Kim et al. (1982) reported a significant effect of hot wash only for 100% cotton heavy fabrics, but not for light fabrics, with fonofos and alachlor.

With respect to the drying method for contaminated and washed fabrics, there is no published report on how drying temperature and air suction affect pesticide-residue removal. In a controlled study of other laundering and fabric variables, researchers have used air drying at a room temperature. Since some pesticides are degraded or volatilized by heat, theoretically drying at a higher temperature as in a dryer might aid in their removal from fabric structures. However, cross-contamination may result among the fabric specimens during tumbling, unless each specimen is tumbled in a separate chamber. Drying thermoplastic fibers at a high temperature may also be a problem.

MATERIALS AND METHODS

Fabric variables are described in Table 1. Fabrics F1, F2, and F3 were purchased from the TESTFABRICS INC. F4 was provided by the Greenwood Mills and bleached by the SRRC, USDA.

Table 1. Description of fabric variables

Fabric Symbol	Fiber Content	Fabric Weight	Finish and Weave
F1	100% Cotton	125 g/sq m	Bleached, mercerized broadcloth
F2	100% Cotton	250 g/sq m	Bleached, mercerized twill
F3	65/35% P/C	105 g/sq m	Bleached broadcloth
F4	65/35% P/C	250 g/sq m	Bleached twill

Alachlor (2-chloro-2',6'-diethyl-N-(methoxymethyl)acetanilide), a corn and soybean herbicide, was used in an emulsifiable-concentrate formulation (four pounds per gallon) at its undiluted initial concentration. Monsanto Company provided the chemical. The emulsifiable-concentrate formulation has been found most difficult to remove by laundering (Easley et al. 1981). Also, a linear relationship was reported between the initial concentration of the pesticide and the amount of residue retention (Easley et al. 1982 b).

Fabric type at four levels (100% cotton and 65/35% polyester/cotton, each in light and heavy weight), pretreatment type at three levels (alcohol, perchloroethylene, water), detergent type at three levels (AATCC, Tide, heavy-duty liquid), wash-water temperature at two levels (hot, warm), and drying method at two levels (air, dryer) were the independent variables. The dependent variable was the residue amount remaining in the washed and dried specimen. There were 18 cells of variable combinations of pretreatment, detergent type, and wash-water temperature (3x3x2). The sequence of laundering of these 18 cells was selected randomly; in each cell eight specimens, four for each fabric type for air drying and four for each fabric type for dryer drying, were washed in separate metal canisters in the same Launder-ometer wash cycle. After one complete laundering of all the variable cells (18x8=144 specimens), the second and the third replications were repeated, in the same manner as the first replication. The total number of fabric specimens, therefore, was 432 (144x3). The contamination process was the same as in our previous work (Kim et al. 1982).

The contaminated specimens were allowed to dry in the air at room temperature for two minutes and then put in the Launder-ometer canisters for washing. The three pretreatments were 7.5 ml denatured ethyl alcohol with 142.5 ml distilled water, 7.5 ml perchloroethylene with 142.5 ml distilled water, or 150 ml distilled water.

These were placed in the separate canister with 20 steel balls. The pretreatment cycle lasted for two minutes, at a warm temperature (49°C). This solution was decanted before laundering with detergent.

For the laundering cycle, 0.3 g AATCC Standard Detergent Type 124 (12.4% phosphate content), 0.6 g Tide (6.1% phosphate content), or 0.3 ml Wisk (heavy-duty liquid detergent with no phosphate) was put into each canister and distilled water was added to bring the total volume in each canister to 150 ml. The wash cycle lasted for nine minutes, at a warm (49°C) or hot (60°C) temperature. Two rinse cycles followed for three and two minutes, each with 150 ml distilled water at a warm temperature (49°C).

For air drying, washed specimens were placed horizontally on dowels wrapped with aluminum foil in a fume hood at room temperature and allowed to dry for 24 hours. For dryer drying, four specimens representing each fabric type were securely pinned to a metal-mesh rack tucked horizontally inside the tumbling chamber of a regular electric dryer (Speed Queen). The tumbling motor belt was disconnected so that the drying cycle worked without the tumbling motion for 30 minutes at the normal temperature setting (97°C).

After drying, each fabric specimen was placed in a 250 ml Erlenmeyer flask with a stopper and 100 ml solvent (ethyl acetate) was added. After shaking for an hour using a mechanical shaker, the first aliquot was decanted to a medicine bottle with a screw cap. Another 100 ml ethyl acetate was added to the Erlenmeyer flask containing the fabric specimen and shaken again for an hour. The second aliquot was also decanted to the medicine bottle, bringing the total volume of the extract solution to 200 ml.

The extract solution was evaporated using a rotary glass evaporator down to approximately 2 to 6 ml, decanted to a 10 ml cylinder, and the volume adjusted to 10 ml. After mixing, 2 ml of the extract concentrate was transferred from the cylinder to a 2 ml vial with a Teflon-lined screw cap and stored in a freezer until the gas chromatograph analysis.

Pesticide residue analyses were performed by using a Hewlett Packard 5710 gas-liquid chromatograph equipped with a nitrogen/phosphorus flame-ionization detector and a Hewlett Packard 3380A integrator. A glass column (1.83 m x 2 mm i.d.) packed with 3% OV-17 on 100/200 mesh Gas Chrom Q utilizing nitrogen as the carrier gas at 30 ml/min was used. The detector flow rate for hydrogen was 3 ml/min and for air was 50 ml/min. The oven temperature was 210°C, and the injector and detector were maintained at 300°C.

RESULTS AND DISCUSSION

We used the GC analysis data in parts per million (Table 2) for analysis of variance statistical procedures to determine the effects of the fabric, laundering, and drying variables on pesticide-residue removal from the contaminated fabric specimens.

Table 2. Residue amount after laundering (ppm)[a]

Wash Temp.	Detergent Pretrt.	Light Fabrics: 100% Cotton (F1) Air Drying	Light Fabrics: 100% Cotton (F1) Dryer Drying	Light Fabrics: 65/35% P/C (F3) Air Drying	Light Fabrics: 65/35% P/C (F3) Dryer Drying	Heavy Fabrics: 100% Cotton (F2) Air Drying	Heavy Fabrics: 100% Cotton (F2) Dryer Drying	Heavy Fabrics: 65/35% P/C (F4) Air Drying	Heavy Fabrics: 65/35% P/C (F4) Dryer Drying
Hot Wash	AATCC								
	Alcohol	37.3	49.3	240.3	24.7	4629.0	2953.7	8634.0	12044.0
	Perc	194.0	191.0	152.0	36.7	3578.7	2046.7	461.0	496.0
	Water	183.0	97.0	389.7	112.7	5334.7	4239.0	11414.0	10963.3
	Tide								
	Alcohol	189.3	30.0	149.7	811.0	4991.7	2786.0	7999.3	6724.0
	Perc	108.3	36.0	27.3	18.3	3699.7	3383.0	592.7	367.7
	Water	170.0	11.7	209.3	105.3	4267.3	2802.0	7766.7	8112.7
	Heavy Duty								
	Alcohol	234.0	100.7	383.0	429.3	4795.7	5060.0	10826.0	8948.0
	Perc	266.7	175.0	225.7	192.3	4542.0	3153.0	760.3	314.7
	Water	174.7	108.3	595.3	262.3	3493.7	4408.3	8319.0	8165.7
Warm Wash	AATCC								
	Alcohol	88.7	103.7	420.3	86.3	4304.0	3637.0	9650.7	10066.0
	Perc	143.3	218.0	51.7	33.7	5113.7	4061.7	620.0	475.3
	Water	318.0	381.3	420.0	136.7	5711.3	4898.7	9897.3	9287.0
	Tide								
	Alcohol	179.3	58.7	203.3	76.0	7808.3	2718.0	10516.0	7789.3
	Perc	209.3	127.3	24.0	30.0	5559.0	3790.0	612.7	501.7
	Water	202.0	123.0	606.0	215.3	6062.7	4269.3	9938.0	12290.0
	Heavy Duty								
	Alcohol	214.0	204.7	808.0	724.3	5937.7	2179.3	9787.3	7534.3
	Perc	274.0	99.3	176.0	92.0	5137.7	2983.7	759.7	294.3
	Water	253.3	111.7	937.7	372.3	6126.7	4811.3	15096.7	7557.3

[a] Mean of three replications

Fabric weight had a definite influence on the amount of pesticide residue, the light fabrics (F1, F3) having much smaller residue retention than the heavy fabrics (F2, F4) as shown in Table 2. This reinforces our earlier findings on alachlor (Kim et al. 1982). The difference in residue retention between the two fabric weights was large and significant (a significance level of 0.01 or beyond is implied when a result is discussed herein as being significant, except for Duncan's multiple-range tests where a significance level of 0.05 is used). Therefore, the data were divided into two groups, light and heavy; this way we could more logically summarize the effects of other variables on residue retention. Although pesticides are more easily removed from light fabrics, they are not to be recommended because other studies have shown that more pesticide penetrated in thinner 100% cotton chambray fabrics (Orlando et al. 1981). The ease of penetration and absorption characteristics of the fabrics need further study.

Residue retention differed significantly by fiber content in the heavy fabrics, but not in the light fabrics. Duncan tests showed that the 65/35% polyester/cotton fabric (F4) retained significantly more residues than the 100% cotton fabric (F2), indicating the soil formulation (oil-based)/fiber property (oleophilic) interrelationship suggested by Easter (1983). Therefore, in the heavy fabrics 100% cotton may offer greater protection from oil-based pesticide formulations. Inconsistent conclusions on the effect of fiber content which have been reported in the literature (Finley and Rogillio 1969; Easley et al. 1981; Lillie et al. 1981; Easley et al. 1982 a; Easter 1983) may have resulted from pooling the data across different weight groups or from differences in blend ratio and the pesticide chemical itself.

Residue retention differed significantly by pretreatment type in both the light and heavy fabrics. Duncan tests showed that perc pretreatment removed a significantly larger amount of alachlor residues than the alcohol or water pretreatments. The difference in residue retention between the alcohol and the water pretreatments was not significant, although the alcohol pretreatment lowered residue retention more than the water pretreatment did. Because of the unique laboratory pretreatment technique used, the alcohol dissolved in the water, thus getting diluted in concentration, whereas the water-insoluble perc retained its initial concentration. This may explain partly the ineffectiveness of the alcohol pretreatment. The average level of residue retention by the perc pretreatment was approximately half of that by the alcohol, indicating the effectiveness of perc in removing oil-based soils.

In the light fabrics, the effect of detergent type was significant: Tide and AATCC detergents containing medium to high amount of phosphate removed significantly more alachlor residues than the heavy-duty liquid detergent without phosphate did. Between Tide and AATCC, the difference was not significant, although Tide removed slightly more residues than AATCC detergent did. The amount Tide removed was nearly twice that removed by the heavy-duty liquid detergent. In the heavy fabrics, residue retention did not differ significantly by detergent type. However, Kim et al. (1982) reported that laundering with detergent was more effective in residue removal than laundering without detergent, for both the light and heavy fabrics.

The detergents performed more effectively in a hot wash than in a warm wash (in a range of 9% to 29% more soil removal). This result is generally in line with findings by Easley et al. (1982 a); however, there is a disagreement in the rank of detergent types for effectiveness at hot wash. No significant interaction effect was noted between the detergent type and the wash-water temperature.

Although hot wash removed more residues than warm wash did in both the light and heavy fabrics, the difference was not significant. In the heavy fabrics, the effect was significant only at 0.05 significance level. This result further augments our earlier findings (Kim et al. 1982) that hot wash was more effective for the heavy fabrics, whereas for the light fabrics wash-water temperature was not a significant factor. No interaction between the wash-water temperature and the fiber content was noted.

In both the light and heavy fabrics, residue retention was significantly lower by dryer drying than by air drying. We suspect either heat (alachlor decomposes at 105°C) or air suction, or both involved in a dryer drying contributed to the reduction of residues. Further research is recommended to clarify the issue. For this research, tumbling action in the dryer was not allowed in order to prevent the probable cross-contamination of the fabric specimens by direct contact with each other or with dryer walls. Periodic checks revealed no contamination of the dryer interior drum after specimen drying.

The only significant interaction was noted between the fiber content and the pretreatment type in both weights of the fabrics: perc was more effective in removing residues from the polyester/cotton fabrics than from the cotton fabrics, whereas alcohol or water was more effective for the cotton fabrics than for the polyester/cotton fabrics. Due to the increased oleophilic nature, the polyester/cotton fabrics seemed to retain the oil-based emulsifiable-concentrate formulation of alachlor more than the hydrophilic cotton fabrics did. Easter (1983) suggested the same theory on fiber/soil interaction. Whereas the cotton fabrics absorbed water or water/alcohol solution readily in the detergency action, perc seemed to penetrate into the oleophilic polyester/cotton fabric readily and release the oil-based soil.

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