Pesticide Removal From Clothing By Laundering

C.N. Nelson, J.R. Fleeker, and C. Janecek

Surveys of farm operators who apply pesticides have shown that the operators usually wear ordinary work clothing when preparing, transferring, or applying these chemicals (Johnson and Byers, 1979; Waldron and Park, 1981; Rucker, et al., 1987). Although ordinary clothing protects the skin from immediate pesticide contact, it can facilitate pesticide absorption by the body if contaminated clothing is worn rather than removed. Wearing contaminated clothing can cause chronic exposure or illness (Gunn and Stevens, 1976; USDA and USEPA, 1975).

Pesticides must be removed from contaminated clothing before that clothing can be safely worn again. However, the completeness of removal may depend on pesticide chemical class and formulation and laundry conditions.

A five-year research project of the North Central Region Agricultural Experiment Stations and the Agricultural Research Council of Alberta examined methods for removing several different pesticides from contaminated clothing. The North Dakota Agricultural Experiment Station conducted studies with paraquat, carbaryl and chlorothalonil. These pesticides were chosen because of their differing levels of toxicity and solubility in water (Table 2).

The objectives of these studies were to examine various factors that would affect the removal of paraquat, carbaryl, and chlorothalonil in laundering, specifically pesticide solubility and formulation, water temperature, water volume, number of extractions (prerinses or washes), and the use of detergent.

METHODS

Several pesticides and formulations were examined in the laundering experiments (Table 2). Pesticides containing radioactive carbon (C-14) were used for measurement of pesticide remaining on the fabric after various laundering conditions. Each C-14 labeled pesticide (< 0.1 mg) was added to the formulation to be studied and the formulation then dissolved or suspended in distilled water to give 0.6 g/100 mL of active ingredient. Similar solutions were prepared with analytical standards of chlorothalonil and carbaryl in place of the formulated product, but acetone replaced water as the solvent.

The following fabrics were used: blue jean fabric (475 g/m^2, indigo dyed cotton denim); lightweight work pant fabric (250 g/m^2, undyed cotton twill); light shirt fabric (105 g/m^2, 65 percent Dacron polyester/35 percent cotton broadcloth); sock fabric (407 g/m^2, undyed 85 percent cotton/15 percent nylon knit). Sizing and manufacturer applied fabric softeners were removed as per American Association of Textile Chemists and Colorists (AATCC) method. The outer 10 percent of the fabric specimen was removed as indicated for test specimen preparation by ASTM.

Each 4x16 cm fabric specimen was contaminated in a 4x4 cm area at one end with 50 microliters (ul) of solution containing 300 to 330 micrograms (ug) of pesticide. Specimens were dried at 24 h at 22C and 65 percent relative humidity prior to the laundering procedure.

The laundering procedures used 150 ml of moderately hard (8 gr./gal.) municipal water or distilled water with each specimen. This simulated a 2-pound load of clothing in a 12-gallon washer, which is consistent with the generally accepted recommended to wash only a few pesticide-contaminated articles at a time. Several detergents were used in the tests: AATCC detergent 124 (12.4 percent P); nonphosphate Tide (0.5 percent P); phosphate Tide (6.1 percent P); and nonphosphate, heavy duty liquid Era.

After laundering, the specimens were squeezed to remove excess water and air-dried with the originally-contaminated end hanging down. The pesticide remaining on the specimen was determined by either of two methods. In the first method, 2 by 4 cm sections of undyed specimens were placed in 1 ml water and 10 ml scintillation solvent and radioactivity measured in a liquid scintillation counter (Gordon, 1968). The second method was used for the indigo dyed denim; 200 mg pieces of the specimen were oxidized to carbon dioxide in a Harvey biological oxidizer and the carbon dioxide was collected in a scintillation-trapping solvent and assayed for radioactivity. Corrections were made for counting and oxidation efficiency.

RESULTS AND DISCUSSION

The pesticides used in this study have a wide range of solubility in water (Table 1). Paraquat is an organic salt and very soluble in water, carbaryl is of intermediate solubility, and chlorothalonil is almost insoluble in water. Analytical standards of carbaryl and chlorothalonil as well as formulations were used to determine the effects of inactive ingredients in pesticide formulations on the extractibility of the pesticides from clothing fabrics. Analytical standards are not commercially available for use as pesticides.

Distilled water was less effective than hard water alone or with sodium chloride in removing paraquat from fabric (Table 3). Cotton fibers develop a weak anionic surface potential in water which may bind the cationic paraquat molecule to the cotton fiber. Salt such as sodium chloride in

Nelson was assistant professor and Janecek is emeritus professor, Department of Apparel, Textiles, and Interior Design; Fleeker is professor, Department of Biochemistry.

the water may reduce the interaction of paraquat with the cotton fiber and increase removal of the pesticide. Hard water contains low levels of salts, which probably accounts for its being more effective than distilled water. This phenomenon may occur with other cationic pesticides such as difenzoquat and diquat. Little difference was noted in the removal of chlorothalonil and carbaryl from undyed cotton twill with either distilled or municipal water, probably because these pesticides are not ionized (Table 4). Table 3 also shows the effect of fabric weight on pesticide removal. The heavier fabrics, denim and undyed cotton twill, retained more pesticide than the lightweight broadcloth regardless of type of water used for removal.

One extraction with distilled water removed most of the formulated pesticides from the undyed cotton twill (Table 5). In general, more soluble pesticides were removed to a greater extent. This trend was especially apparent after additional extractions, which reduced the contamination to less than 5 percent of the original. An exception was pure chlorothalonil. Most sprayable commercial pesticide formulations contain surfactants. Surfactants in the clorothalonil flowable formulation may account for the ease of removal from fabric over that of the analytical standard.

The residual pesticide after three extractions may be trapped in cotton fiber lumens or other portions of the fabric that are not readily accessible to water. Increasing extraction (laundering) time or soaking the fabric before laundering would be expected to further reduce residual pesticide in the fabric (Table 4).

Table 4 shows that higher water temperature, longer extraction time, and greater extraction volume increased the removal of carbaryl and chlorothalonil. The addition of isopropyl alcohol increased removal of the unformulated analytical standards but was inconsistent in the effect on formulated pesticide removal.

Detergent increased the efficiency of pesticide removal slightly over water alone (Table 6). However, the residue of the chlorothalonil analytical standard was considerably higher than for chlorothalonil in the flowable formulation. Carbaryl analytical standard was efficiently removed regardless of the presence of detergent. This may reflect the greater solubility of carbaryl as compared to pure chlorothalonil.

The results with paraquat are not presented but were similar with respect to enhanced removal of pesticide when detergent was used. Paraquat removal by four different detergents was also compared. There was little difference in removal efficiency by AATCC detergent 124, nonphosphate Tide, phosphate Tide, or the nonphosphate heavy duty liquid Era (Olsen, Janecek, and Fleeker, 1986).

The contamination of specimens in a 4 by 4 cm area at one end permitted the determination of pesticide redeposition on the remaining uncontaminated areas during laundering. The pesticide was redeposited from the treated to untreated areas of this fabric during extraction (Table 5). However, after three extractions the level of contamination was low, less than 1 percent of the original. Redeposition of soiling agents during laundering is a general phenomenon which has been observed for materials other than pesticides (Gordon, 1968).

Table 1. Properties of the pesticides studied.

	Paraquat	Carbaryl	Chlorothalonil
Use	dessicant	insecticide	fungicide
Acres treated in North Dakota[a]	59,000 (1984)	78,400 (1984)	4,200 (1978)
Toxicity	Class II (moderately hazardous)	Class III (slightly hazardous)	Class IV (relatively hazardous)
Water Solubility[b] (mg/L)	>50,000	120	0.6

[a]McMullen et al., 1985.

[b]Worthing and Walker, 1979.

Table 3. Effect of salt and water source on removal of paraquat from fabric.

	Paraquat remaining (percent)[a]		
Treatment	Undyed polyester cotton broadcloth	Undyed cotton twill	Indigo dyed cotton denim
Distilled water	12.7 + 1.7	29.1 + 2.3	37.8 + 3.8
Municipal water	3.6 + 0.8	7.4 + 2.2	22.3 + 5.3
Distilled + 0.5% NaCl	2.7 + 0.3	4.4 + 1.7	9.6 + 1.9
Municipal water + 0.5% NaCl	2.8 + 0.3	4.4 + 0.9	8.8 + 2.3

[a]The data represent the mean and standard deviation from three replications. The specimens were extracted 2 min. at 49 C.

Table 2. Compounds and formulations examined in the laundering tests.

Compound	Formulation type	Designation in text	Brand	EPA reg. No.
Paraquat	Soluble concentrate	SC	Ortho	239-2186-AA
Carbaryl	Soluble concentrate	SC	Liquid Sevin	239-2356-AA
Carbaryl	Flowable	F	Sevin XLR	264-222
Carbaryl	Wettable powder	WP	Sevin	1016-43
Carbaryl	Analytical standard	P	——	None
Chlorothalonil	Flowable	F	Bravo 500	50534-8
Chlorothalonil	Analytical standard	P	——	None

Table 4. The effect of water temperature, laundering time, and water volume on pesticide removal from undyed cotton twill.

	Pesticide Remaining (percent)[a]					
Extraction Method	Carbaryl (SC)[d]	Carbaryl (F)[d]	Carbaryl (WP)[d]	Carbaryl (P)[d]	Chloro-thalonil (F)[d]	Chloro thalonil (P)[d]
2 min; 24° C; 150 mL[b]	5.0 + 0.1	37.4 + 2.0	6.8 + 1.6	27.9 + 1.1	21.1 + 2.1	53.8 + 2.0
2 min; 49° C; 150 mL[b]	4.0 + 0.2	29.1 + 4.6	5.0 + 0.2	4.7 + 0.9	12.1 + 1.0	48.1 + 1.5
5 min; 49° C; 150 mL[b]	3.6 + 0.2	11.5 + 2.5	3.8 + 0.3	3.8 + 0.2	9.4 + 1.0	42.9 + 1.6
5 min; 49° C; 300 mL[b]	2.6 + 0.1	12.9 + 1.9	2.6 + 0.1	3.2 + 0.7	8.7 + 0.3	42.8 + 1.1
5 min; 49° C; 150 mL[b] 10% alcohol[c]	3.0 + 0.5	6.8 + 0.9	3.4 + 0.2	2.4 + 0.3	3.5 + 0.6	38.7 + 1.2
5 min; 49° C; 150 mL municipal water	3.7 + 0.1	12.0 + 2.0	3.9 + 0.1	3.7 + 0.1	11.9 + 1.3	45.4 + 0.8

[a]The date represent the mean and standard deviation from three replicates.

[b]The extraction done with distilled water.

[c]The extraction done with 10% isopropyl alcohol (rubbing alcohol).

[d]Formulations used were soluble concentrate (SC), flowable powder (F), wettable powder (WP), and the unformulated analytical standard (P).

Table 5. Effect of three extractions with distilled water on removal of pesticide from contaminated undyed cotton twill.

Pesticide and formulation[a]	One extraction	Pesticide remaining[b] Two extraction	Three extraction
Paraquat (SC)	2.4 + 0.2 (0.7 + 0.1)	0.5 + 0.1 (0.1 + 0.1)	0.4 + 0.1 (<0.1)
Chlorothalonil (F)	9.4 + 0.9 (1.3 + 0.2)	3.5 + 0.6 (0.4 + 0.2)	3.7 + 0.2 (0.2 + 0.1)
Chlorothalonil (P)	42.6 + 1.2 (1.4 + 0.7)	38.7 + 1.2 (0.6 + 0.3)	37.9 + 1.6 (0.5 + 0.1)
Carbaryl (WP)	3.8 + 0.3 (2.6 + 0.3)	0.9 + 0.1 (0.3 + 0.1)	1.0 + 0.1 (<0.1)
Carbaryl (SC)	3.6 + 0.2 (2.4 + 0.3)	1.3 + 0.1 (0.2 + 0.1)	1.5 + 0.2 (0.1 + 0.1)
Carbaryl (F)	11.5 + 2.5 (2.5 + 0.4)	3.3 + 0.2 (0.4 + 0.1)	3.0 + 0.3 (0.1 + 0.1)
Carbaryl (P)	3.8 + 0.2 (1.4 + 0.2)	1.6 + 0.1 (0.2 + 0.1)	1.0 + 0.1 (0.1 + 0.1)

[a]Formulations used were soluble concentrate (SC), flowable powder (F), wettable powder (WP), and the unformulated analytical standard (P).

[b]The values represent the mean and standard deviation from three replicates. The upper value represents the total pesticide remaining on the entire swatch and the value in the parenthesis represents the residue found on the originally uncontaminated portion of the swatch. The water used to extract paraquat contained 0.5% NaCl.

CONCLUSIONS AND IMPLICATIONS

In general, greater pesticide removal resulted from increased number of extractions and a greater volume of water. Higher water temperature removed more pesticide. The use of detergent also increased pesticide removal.

Additives such as isopropyl alcohol did not significantly improve removal of pesticide formulations; however, it did increase the removal of the analytical standards of carbaryl and chlorothalonil. A pesticide-specific additive such as common salt was useful in paraquat removal but not in carbaryl or chlorothalonil removal.

Several recommendations can be made from the results of these studies. First, predictions of pesticide removal based on solubility and formulation are problematic. Pesticides include a wide variety of chemicals, not all of which are readily removed from clothing fabrics. Formulations of 2,4-D esters (Easley, Laughlin, Gold, and Tupy, 1983) and chlorpyrifos (Keashall, Laughlin, and Gold, 1986) are resistant to removal by laundering, yet pesticides of similar structure are readily removed. One pesticide formulation may be extracted from a fabric more efficiently than another although both contain the same pesticide.

Second, presoak pesticide contaminated clothing first in a separate container of warm water before laundering to reduce possible contamination of laundry equipment. This also removes a considerable amount of the pesticide. Wash only a few items per load and follow the prerinse with a wash and one or two rinse cycles, all in hot water. Use the maximum wash and rinse periods to allow for maximum dissolution of the pesticide residues.

Contaminated clothing should be laundered separately from other family clothing. Even though minute quantities of pesticides were found to transfer to uncontaminated areas of fabric, the transferred pesticide was distributed over the entire uncontaminated area.

Finally, although it was shown that pesticides were more easily and completely removed from light-weight fabrics, heavier weight fabrics should be worn during pesticide application to prevent penetration to the skin of the wearer.

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Table 6. Effect of AATCC detergent 124 on the efficacy of pesticide removal from four fabric types.

Detergent added	Pesticide remaining[a]					
	Carbaryl			Chlorothalonil		
	WP[b]	F[b]	SC[b]	P[b]	F[b]	P[b]
	Undyed polyester/cotton broadcloth					
No	0.7 + 0.1	2.9 + 0.2	1.1 + 0.1	1.2 + 0.2	4.1 + 0.5	40.7 + 1.4
Yes	0.7 + 0.1	2.7 + 0.2	0.9 + 0.1	1.1 + 0.1	3.8 + 0.2	36.6 + 1.2
	Undyed cotton twill					
No	1.0 + 0.1	3.0 + 0.3	1.5 + 0.2	1.0 + 0.1	3.7 + 0.2	37.9 + 1.6
Yes	0.5 + 0.1	2.5 + 0.2	0.9 + 0.1	0.6 + 0.1	3.1 + 0.3	35.6 + 0.6
	Undyed cotton/nylon knit					
No	3.2 + 0.3	4.4 + 0.2	3.8 + 0.3	2.0 + 0.2	5.5 + 0.5	29.9 + 1.1
Yes	4.4 + 1.2	3.6 + 0.4	4.1 + 0.3	2.6 + 0.4	3.6 + 1.1	31.5 + 1.9
	Indigo dyed denim					
No	2.0 + 0.2	8.9 + 1.8	2.4 + 0.5	3.1 + 0.3	9.7 + 1.0	57.7 + 0.8
Yes	1.1 + 0.2	7.6 + 0.8	2.0 + 0.1	2.2 + 0.1	4.6 + 1.2	48.8 + 2.4

[a]The values represent the mean and standard deviation from three replicates.

[b]Formulations used were soluble concentrate (SC), flowable powder (F), wettable powder (WP), and the unformulated analytical standard (P).

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