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**Determination of pesticide levels as the result of cross-contamination
during laundering**

Milikin, Corinth, Ph.D.

The University of North Carolina at Greensboro, 1989

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DETERMINATION OF PESTICIDE LEVELS AS THE RESULT
OF CROSS-CONTAMINATION DURING LAUNDERING

by

Corinth Milikin

A Dissertation Submitted to
the Faculty of the Graduate School at
The University of North Carolina at Greensboro
in Partial Fulfillment
of the Requirements for the Degree
Doctor of Philosophy

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Approved by

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APPROVAL PAGE

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This study investigated the pesticide cross-contamination that occurs during laundering. The effect of pesticide, water temperature, and fabric combination were examined. Three pesticides--atrazine, Diazinon, and metolachlor--were chosen along with two water temperatures 27°C and 60°C, and in conjunction with eight fabric combinations.

The four fabrics tested were two top weight fabrics and two bottom weight fabrics common in pesticide worker clothing. Field strength (1.25% a.i.) pesticides were used to contaminate the fabrics. Pesticide soiled fabric samples were individually laundered along with the same weight fabric that had not been exposed to pesticides in an Atlas Launder-Ometer in a method to represent one home laundry cycle. Samples were individually extracted and analyzed using gas chromatographic techniques. Residues extracted from fabrics range from 0.0 ug to 350.2 ug with a mean level of 60.0 ug.

An analysis of variance procedure was used to test for the main effects (temperature, pesticide, and fabric combination) and interactions of the main effects. All main effects were significant at the .0001 level. Further examination of all possible pair-wise comparisons of treatment means was carried out with a Duncan's multiple

range post hoc procedure with $p < .05$ as indication of significance.

The difference in pesticide cross-contamination between each of the pesticides was significant. Diazinon residue levels were the highest and atrazine the lowest. The pesticide cross-contamination was significantly greater in hot water than in cold. There were significant differences among the fabric combinations in the level of cross-contamination. The greatest levels of pesticide cross-contamination were in 100% cotton denim and knit fabrics.

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CHAPTER I
INTRODUCTION

Pesticides, which include herbicides, insecticides, and fungicides, are used in 90 percent of all United States households. Five pounds of pesticide are used each year for each man, woman, and child in the United States (Ware, 1978). Pesticides are used for a variety of purposes, but primarily to control agricultural pests, weeds, and diseases. Pesticides are selected because they have specific biological effects, usually they are toxic to some organism. These chemicals may not have an acute toxic effect on people, but often, with adequate dosage through time, adverse effects may occur.

It is assumed that a balance exists between risks and benefits of pesticide use. However, frequently, there is no perceived risk on the part of the pesticide user. The agricultural worker is exposed to these toxic chemicals while performing many operations.

The mixing and loading of pesticide chemicals results in a high exposure situation and is the frequent occasion of high concentration pesticide spills. The application of the pesticides with the use of a spray mechanism may result in clothing that is completely soaked with pesticide as the

result of wind drift of the solution. After application, the worker again comes into direct contact with the pesticide solutions with the unloading and cleaning of the agricultural equipment. And finally, the field worker is exposed to the chemicals when re-entering the treated fields.

Repeated exposure to small amounts of some pesticides, as well as acute exposure, can cause sudden severe illness. With some pesticides, dermal contact alone can cause death. However, exposure to pesticides may not result in outward symptoms or obvious signs of poisoning. Symptoms of human pesticide poisoning can include nausea, restlessness, lethargy, loss of libido, impotence, abdominal pains, insomnia, rashes, headache, dizziness, psychological disorders, weight loss and hemorrhaging (Bureau of National Affairs, 1987). Many of these symptoms are associated with other illnesses and therefore may not be perceived as an indication of pesticide poisoning.

The degree of protection offered by clothing depends on the type and amount of clothing worn, the ability of the textile substrate to prevent pesticide transfer to the skin, the interaction of environmental and climatological factors, and the type of work performed. A protective clothing standard was adopted by the Environmental Protection Agency (EPA) on May 10, 1974. This standard defined protective clothing as "at least a hat or other suitable head covering,

a long-sleeved shirt and long-legged trousers or coverall type garment (all of closely woven fabric covering the body, including arms and legs), shoes, and socks" (EPA, 1974).

Pesticides are used in both oil based and water based media. Barrier finishes such as fluorocarbons impart oleophobic and hydrophobic properties, thus limiting wetting of the fabric by the pesticide formula. In cases where garments have been wetted with highly toxic or concentrated pesticides, the United States Department of Agriculture recommends disposal of the contaminated garments rather than laundering. Oil based formulas are especially difficult to remove from synthetics such as nylon or polyester and highly concentrated pesticides are difficult to remove to any safe level.

Even with the availability of specially designed protective clothing, one must recognize that other clothing will continue to be used and become soiled with pesticide. Although disposable protective clothing is available, many consumers will choose not to use these items for various reasons including lack of perceived danger of the pesticides. Even when protective clothing is used, garments worn underneath may become soiled with pesticide (Orlando, Branson, Ayers, & Leavitt, 1981).

The design of nonwoven disposable garments, while providing improved protection from dermal exposure, does allow some penetration of the pesticide solution through

structural seams and zipper closures. While the garments have good strength to thickness ratios, the garments can be damaged and torn during the climbing, reaching, and bending activity required for agricultural work and thus allows additional exposure to pesticides.

Garments worn by pesticide workers should be laundered after each wearing. However, several problems are associated with laundering of pesticide soiled clothing. First, it is difficult to determine if all contaminants have been removed without the use of sophisticated laboratory equipment. Second, all laundering equipment must be decontaminated after each load of pesticide soiled clothing. In addition, it has been determined that cross-contamination can occur when clothing that is free of pesticides is laundered with pesticide soiled clothing or in the same equipment used to launder pesticide soiled clothing (Braun, Frank, & Ritcey, 1989; Easley, Laughlin, Gold & Tupy, 1983; Finley et al., 1979; Laughlin, Easley, Gold, & Tupy, 1981).

Other studies suggest that families of exposed workers may develop health problems after secondary exposure through laundering of work clothing at home (Bellin, 1981). Laughlin et al. (1981) found methyl parathion residues on laundered contaminated denim fabrics to be toxic to German cockroaches.

Laundering studies of pesticide removal have varied in results with the most successful removing 50 to 95 percent of the pesticide. Laundering at high wash temperatures was recommended although they did not give significantly improved results at the .05 level.

Several researchers have studied laundering of pesticide soiled worker clothing. However, only limited work in the literature has addressed the degree of pesticide cross-contamination that occurs during laundering.

Purpose of the Study

Since agricultural workers use a variety of pesticides, there existed the need to investigate common pesticides for the tendency of laundry procedures to redeposit the pesticide. The purpose of this research was to determine the effects of pesticide, water temperature, and fabric combination (pairings of pesticide spiked and unspiked fabrics) on the level of cross-contamination that occurs during laundering of pesticide soiled fabrics with unsoiled fabrics under carefully controlled test conditions.

Laboratory results will give an indication of the cross-contamination that may occur in household laundry.

Therefore the objectives of this study were:

1. To develop laboratory test procedures that will assess the degree of cross-contamination that occurs when pesticide spiked fabrics are laundered with pesticide free fabrics with a commercially available home laundry detergent.

2. To measure the cross-contamination that occurs during laundering with three different pesticide.
3. To measure the cross-contamination that occurs during laundering with hot and cold wash water temperatures.
4. To measure the cross-contamination that occurs during laundering with four different test fabrics in eight combinations of fabrics.

Based on these objectives, the following hypotheses were formulated:

1. There will be a difference among the pesticides in the level of cross-contamination that occurs when a pesticide free fabric is laundered under laboratory test conditions with a fabric that is spiked with pesticide.
2. There will be a difference between the water temperatures in the level of cross-contamination that occurs when a pesticide free fabric is laundered under laboratory test conditions with a fabric that is spiked with pesticide.
3. There will be a difference in the level of cross-contamination that occurs when a pesticide free fabric is laundered under laboratory test conditions with an identical or a different fabric of the same weight class (top or bottom) that is spiked with pesticide.

Limitations

The following were limitations to this study.

1. A limited number of pesticides were studied.
2. The fluorocarbon finish was applied by aerosol spray and was therefore limited in uniformity of application.
3. Only one pesticide concentration was tested.
4. The fabric combinations paired only those fabrics in the same weight classification (top or bottom weight).

CHAPTER II

REVIEW OF LITERATURE

Pesticide is the name given to chemical substances used to kill, repel, or control pests and include insecticides, fungicides, and herbicides. About 48,000 different pesticide products are registered for use in the United States, but these products contain only about 600 groups of active ingredients. Of the pesticides produced, 70% are agricultural products and more than half of these are herbicides (Bureau of National Affairs, 1987). Because pesticides are designed to be toxic to certain organisms, it is important that individuals are aware of potential exposure situations.

Pesticide Absorption

The specific amount of pesticide that will have a toxic effect on humans must be estimated from animal studies. Toxicity is defined by the LD₅₀, expressed as milligrams (mg) of toxicant per kilogram (kg) of body weight, the dose that kills 50% of the test animals to which it is administered under experimental conditions. The LD₅₀ is measured in terms of oral, dermal, and respiratory toxicity. The size of the dose is the most important single item in determining the safety of a given chemical, and actual

statistics of human poisonings correlate reasonably well with toxicity ratings (Ware, 1978).

There are three routes by which pesticides may enter the body: dermal, respiratory, and oral. Research indicates that more pesticide enters the body through dermal contact than through inhalation or ingestion. Therefore it is important to protect the skin as much as possible when exposed to pesticide chemicals. The reports of exposure studies by Wolfe, Armstrong, Staiff, and Comer (1972) indicate that 97% of the pesticide a body is exposed to during most situations is deposited on the skin.

Different parts of the body absorb various pesticides at varying rates. Some areas are much more likely to absorb the pesticides exposed to them. Figure 1 shows the rate of body absorption of pesticides (Branson & Henry, 1982).

Report procedures for pesticide related illness have been developed in several states. In South Carolina from 1979 to 1982, there were an average of 78 hospitalizations per year for pesticide poisoning (Schuman, Caldwell, Whitlock, & Brittain, 1986). California, with its large agricultural industry, reported a total, for 1983, of 1270 illnesses and injuries associated with occupational exposure to pesticides. Of that, 871 cases were agriculturally related (Worker Health and Safety Unit, 1984). In 1982, the North Carolina Department of Human Resources (NCDHR) reported 39 cases of agricultural pesticide related

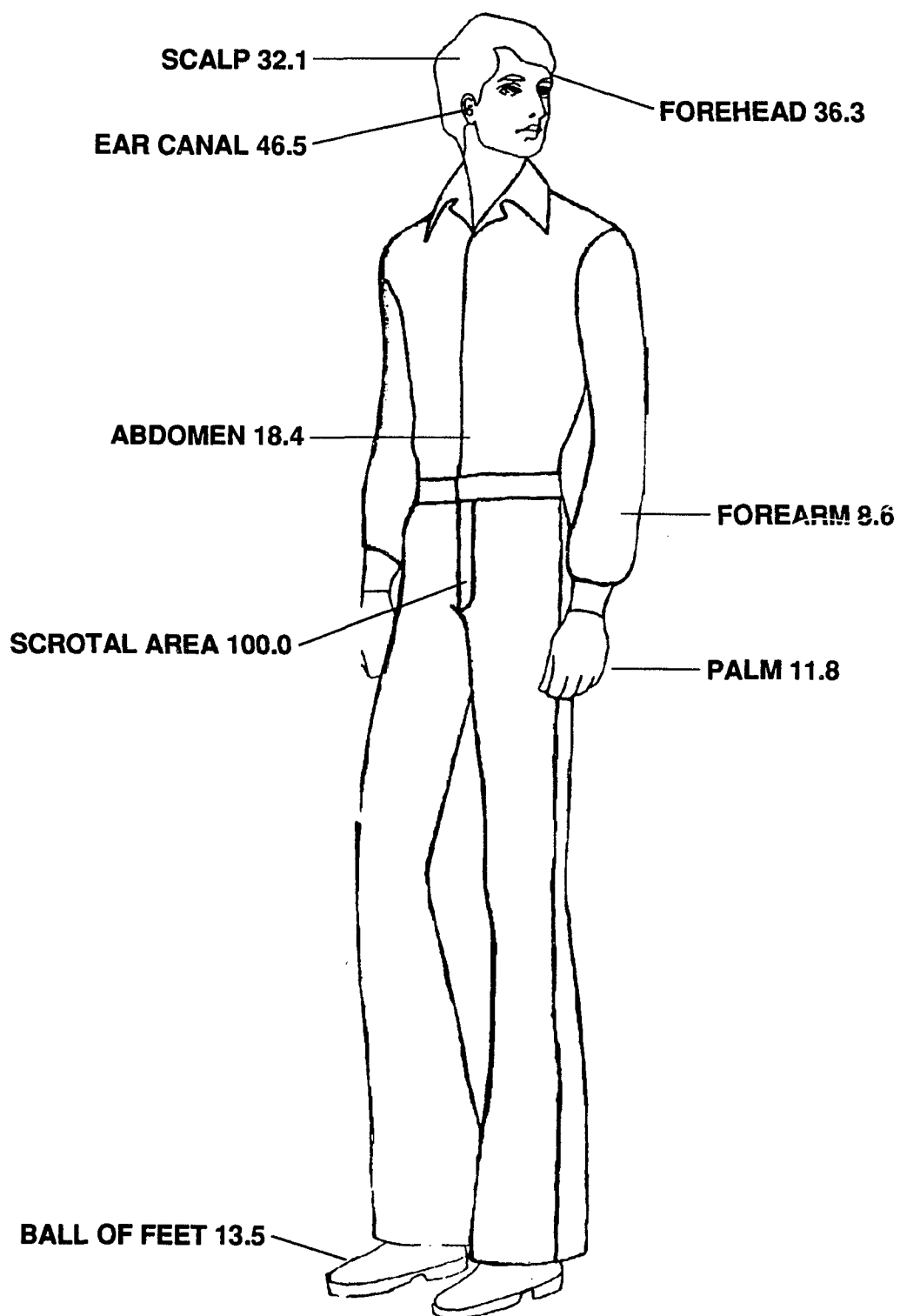


Figure 1. Relative Absorption of Parathion as a Function of Body Location (Branson & Henry, 1982)

illnesses (NCDHR, 1983). Experience in North Carolina has shown that approximately 50% of the injuries resulting from the use of pesticides have occurred because applicators ignored or were unaware of the need to use protective clothing and devices during pesticide mixing and application operations (N.C. Agricultural Chemicals Manual, 1984). Evidence that clothing can be contaminated by pesticide is well documented (Finley, Bellon, Graves, & Koonce, 1977; Finley et al., 1979; Finley & Rogillio, 1969). Exposure studies have determined that pesticide workers are usually subjected to relatively small fractions of an acute dose as they apply the compounds (Wolfe et al., 1972). Personal work habits were identified as a significant factor in determining pesticide absorption levels. Individuals who were obviously wearing contaminated clothes showed greater levels of exposure than other workers. Use of protective clothing or a change of clothing daily appears to be significant in limiting worker exposure (Lavy, Mattice, & Flynn, 1983).

Nonwoven, lightweight, disposable garments have been proposed to eliminate most of the problems associated with dermal pesticide exposure. The garments are sufficiently low in cost to allow disposal after use, therefore eliminating the need for laundering. The garments are lightweight and resist penetration of oils and liquids and some allow water vapor transmission.

Clothing Worn by Pesticide Workers

Little information exists on the extent to which clothing and equipment recommendations for pesticide workers are followed. However, the literature and field observations suggest that agricultural workers often do not dress appropriately to protect themselves from splashes, spills, and spray drifts (Keeble, Norton, & Drake, 1987). DeJonge, Vredevoogd, and Henry (1983-84) stated that currently available protective clothing was not chosen by most farmers, with the exception of the highly pesticide-exposed fruit orchard workers. It was only when desire for protection outranked desire for comfort that protective garments were chosen (DeJonge et al., 1983-84).

Another survey suggested that fruit growers and workers in Virginia and nearby areas often do not wear items of recommended clothing and personal equipment and therefore may not adequately protect themselves when handling pesticides (Keeble et al., 1987). The majority of the sample would normally wear work shirts and pants for mixing and spraying parathion and Captan.

Research conducted in Louisiana indicated the typical work garment of licensed agricultural consultants to consist of a short-sleeved, woven sports shirt and full length pants of denim or khaki work-weight fabric. Short sleeved T-shirts were also frequently worn (Cloud, Hranitzky, Day, & Keith, 1983). Similar clothing was worn by respondents to a

California survey when applying pesticides (Rucker, McGee, & Chordas, 1986). The typical pesticide user in a Canadian survey wore leather shoes or boots, jeans, coveralls, long sleeved shirt, jacket, and a fabric cap (Rigakis, Martin-Scott, Crown, Kerr, & Eggertson, 1987).

Laundry Methods for Removal of Pesticides

The widespread use of everyday work clothing in pesticide application has made research in decontamination techniques necessary. Researchers have reported on a number of laundering variables which have been shown to lower pesticide residues in fabric for a limited number of pesticides. Effective removal techniques have been found for specific pesticides. Variables investigated include frequency of laundering, pre-rinsing, laundry additives, detergent type, water temperature, multiple launderings, and pesticide.

Home laundering greatly reduces the amount of pesticide residues in contaminated clothing (Finley et al., 1974). Because concentrated pesticides are more difficult to remove than field strength pesticides, the first step in determining optimum laundering procedures for insecticide contaminated clothing is to ascertain the level of contamination expected from field exposure (Finley, Graves, Summers, Schilling, & Morris, 1977).

Even after recommended laundering procedures, residues of field strength methyl parathion have been found to be

toxic to German cockroaches (Laughlin et al., 1981). In addition, after laundering residues of full strength (54% a.i.) methyl parathion in fabrics were at levels associated with the death of an adult male (Southwick, Mecham, Cannon, & Gortatowski, 1974).

The wearer may be unaware of residues remaining in the clothing, contaminated garments provide a medium for exposing the skin to the pesticide, making dermal absorption possible. Among the procedures frequently recommended for effective pesticide removal is the daily laundering of clothing exposed to pesticides (Branson & Henry, 1982; Easley, Laughlin, & Gold, 1984; Iowa Cooperative Extension Service [Iowa CES], 1987).

Goodman, Laughlin, and Gold (1988) found methyl parathion contamination was consistently reduced to an acceptable level when items were laundered daily. In contrast, without daily laundering, the methyl parathion contamination increased across a period of five days such that the eventual washing process was not as effective in removing residues. Given the methyl parathion levels in the fabric and in the waste wash and rinse waters, the recommendation to launder protective clothing daily is supported (Goodman et al., 1988).

Pre-rinsing contaminated clothing before washing has also been indicated as an effective method to remove pesticide particles from fabrics (Easley et al., 1984; Iowa

CES, 1987). Of the laundry procedures investigated by Easley, Laughlin, Gold, and Tupy (1981), more complete removal of methyl parathion was found for a pre-rinse followed by a wash and two rinses. Pre-rinsing and/or multiple washing appear to be more effective in methyl parathion residue removal than household ammonia as a laundry additive (Easley, Laughlin, Gold, & Hill, 1982).

However, no significant difference was found between pre-rinsed samples and non-pre-rinsed samples for the removal of 2,4-D ester (Easley et al., 1983). Pre-soak treatment failed to improve removal of field strength or full strength pyrazophos residue over a second wash cycle (Braun et al., 1989).

Laundry additives such as bleach have received limited recommendation for pesticide removal (Finley, Graves et al., 1977). Much of the research has found no significant pesticide residue removal attributable to laundry additives such as ammonia or bleach (Easley et al., 1984; Easley, Laughlin, Gold & Hill, 1982; Easley et al., 1981).

For removal of methyl parathion residues, bleach was found to be slightly more effective than ammonia as a laundry additive (Easley, Laughlin, Gold, & Schmidt, 1982). Diazinon removal may be improved slightly with the addition of bleach, but the difference is not significant when compared with the use of detergent alone (Lillie, Hampson, Nishioka, & Hamilton, 1982).

Keaschall, Laughlin, and Gold (1986) recommended the use of a laundry pretreatment to assist in residue removal of organophosphates, organochlorines, and carbamates. In their study of redeposition of pesticide soil during laundering, Laughlin and Gold (1989b) observed generalized redeposition of pesticide soil to other areas of the fabric. Based on these findings, recommendations were made to pretreat the site of soiling before laundering. Starch and fabric softener were not effective laundry auxiliaries in lowering pesticide residues. Laundering specimens with starch or with fabric softener prior to contamination did not affect after laundering residue levels (Laughlin, Lamplot, & Gold, 1988).

Laundering in hot water was widely recommended for increased pesticide removal (Easley et al., 1984; Finley, Graves et al., 1977; Iowa CES, 1987). Easley et al. (1983) found higher water temperatures were more effective in removing 2,4-D ester formulation from fabrics. Water temperature was not a factor in the removal of 2,4-D amine which was effectively removed from the fabric regardless of the water temperatures (Easley et al., 1983).

Several other studies have reported a tendency for increased residue removal with increased temperature but the difference was often not significant for all pesticides. A trend toward increased residue removal with increased wash water temperature was found for Diazinon, propoxur,

chlordane, bromacil, and malathion (Lillie, Livingston, & Hamilton, 1981).

While an insignificant difference was found in washing temperatures for the removal of methyl parathion, removal was slightly greater at the higher temperature (Laughlin, Easley, Gold, & Hill, 1986; Laughlin & Gold, 1989b). An increase in water temperature resulted in an increase in pesticide removal for Guthion and Captan (Easter, 1983). No significant difference was found in the removal of methyl parathion between 60°C and 49°C water washing, however 30°C water laundering produced significantly lower removal (Easley, Laughlin, Gold, & Schmidt, 1982).

A variety of laundry detergents have been tested and recommended for effective pesticide removal including a heavy duty liquid (Easley et al., 1984), a heavy duty carbonate (Finley, Graves, et al., 1977), and heavy duty phosphate (Iowa CES, 1987). However most research has failed to determine a significant difference in residue removal attributable to detergent type.

Repeated work with methyl parathion, found no significant difference in detergent type for residue removal (Easley, Laughlin, Gold, & Schmidt, 1982; Laughlin et al., 1986; Laughlin & Gold, 1989b). Although detergents could not be statistically separated, heavy duty liquid detergents appeared to provide higher levels of methyl parathion

removal in water temperatures of 49°C and 60°C (Easley, Laughlin, Gold, & Schmidt, 1982).

Obendorf and Klemash (1982) reported that powdered detergents containing carbonate and zeolite builders clean fiber surfaces of oily soil better than an unbuilt liquid laundry detergent. Laundering with this detergent removed more malathion than methyl parathion from the surfaces of polyester fibers (Obendorf & Solbrig, 1986).

Multiple washings and/or an extra rinse is recommended to be certain all pesticide residues are removed (Branson and Henry, 1982; Easley et al., 1984; Finley, Graves et al., 1977; Iowa CES, 1987). Repeated washing was significantly better than single washing in the removal of 2,4-D ester and 2,4-D amine from contaminated denim fabric (Easley et al., 1983).

Fabrics contaminated with field strength concentrations of methyl parathion require a minimum of three launderings before biological activity reaches a harmless level. (Easley, Laughlin, Gold, & Hill, 1982). While clothing worn while using slightly toxic pesticides may be effectively laundered in one to three machine washings (Easley et al., 1984).

All clothing worn during pesticide use should be kept separate from other family laundry (Branson and Henry, 1982; Braun et al., 1989; Easley et al., 1984; Finley, Graves et al., 1977; Finley et al., 1974; Iowa CES, 1987). If several

garments have become contaminated, it was suggested that only one or two garments should be laundered in a single load. Garments contaminated by the same pesticide(s) should be washed together. Overcrowding clothes in the washing machine should be avoided. A full water level allows the water to thoroughly flush the fabric (Easley et al., 1984; Rigakis et al., 1987).

Decontamination of laundry equipment should be done following the laundering of pesticide soiled clothing. The recommended procedure is to rinse the empty washing machine using hot water and the same detergent, machine settings, and cycles used for laundering the contaminated clothing (Easley et al., 1984; Finley, Graves et al., 1977; Iowa CES, 1987).

To avoid contamination of the dryer, line drying is recommended for clothing that was exposed to pesticide and laundered (Easley et al., 1984; Iowa CES, 1987). In addition, post-laundering exposure of apparel to moving air for several days for field strength contamination and longer times for full strength contamination of methyl parathion was found to further reduce residue levels and was therefore recommended (Laughlin & Gold, 1989a). However, simulated sunlight, heat, and humidity exposure treatment did not influence malathion pesticide residue found in either laundered or unlaundered Gore-Tex (Branson & Rajadhyaksha, 1988).

Fabric Characteristics Affecting the
Removal of Pesticides

The effect of fiber content on the removal of pesticide residues has not been completely established. The all cotton or cotton/polyester fiber content of the contaminated fabric was not found to be a contributing factor in the ease of methyl parathion removal in two studies (Easley, Laughlin, Gold, & Schmidt, 1982; Easley et al., 1981).

Finley and Rogillio (1969) found increased cotton fiber content in fabric resulted in increased retention of DDT in fabrics through the laundering process. Residue retention increased with cotton fiber content of a fabric in increased absorption and retention of methyl parathion and DDT (Finley & Rogillio, 1969). Lillie et al. (1981) found a significantly greater amount of the pesticides chlordane, Diazinon, carbaryl and prometon ($p=.01$) penetrated the 100% polyester fabric than the 100% cotton fabric.

Recommendation was made for a fluorocarbon finish for fabrics worn during mixing, handling, or application of pesticides (Keaschall et al., 1986; Laughlin et al., 1986; Laughlin & Gold, 1989a). While the fluorocarbon finish was found to inhibit the absorption of many pesticides, chlorpyrifos was not more completely removed from the fluorocarbon specimens than from the unfinished specimens (Laughlin et al., 1988).

Keaschall et al. (1986) reported that the amount of pesticide absorbed by fluorocarbon finished fabrics was

related to the number of launderings to which the specimen had been subjected prior to contamination with pesticide. The commercially applied finish was not affected by five machine launderings. The renewable finish was effective in reducing absorption through four launderings. The authors recommended that the finish be applied after four wash cycles to achieve continued protection (Keaschall et al., 1986).

Goodman et al. (1988) found the fluorocarbon finish to be effective in lowering the amount of methyl parathion absorbed into the fabric only through two launderings. The recommendation was that renewable fluorocarbon finishes be reapplied after every second laundering.

Gore-Tex, a multi-layer fabric, was not found to be an effective barrier to the full-strength malathion emulsifiable concentrate used by Branson and Rajadhyaksha (1988) to simulate a spill situation. Kim, Stone, and Sizer (1982), suggested that the thickness and weight of a heavier fabric may allow deeper penetration of the pesticide into the fibers as well as into the fabric structure by a wicking process, making the chemical more difficult to remove.

Other Factors Affecting the Removal of Pesticides

Early research by Finley, Graves et al. (1977), suggested that laundering procedures suitable for removing methyl parathion residues were expected to be adequate for other organophosphates such as EPN, Guthion, and malathion.

However, differences in removal were found to occur among insecticides and the best laundry procedures may not be best for all pesticides (Lillie et al., 1982).

Easley et al. (1981) demonstrated success in removal of methyl parathion from fabrics with laundering. Mean percentages removed were higher for encapsulated and wettable powder formulations, with ranges of 93% to 99% methyl parathion removed. Emulsifiable concentrate (EC) methyl parathion removal was lower, ranging from 80% to 88%, indicating that EC formulation apparently was more difficult to remove (Easley et al., 1981).

Although removal is related to pesticide class, which has practical implications to the consumer, there is no predominant trend for differences in laundry removal based on class. In a study of three insecticide classifications, organochlorine residues were the most difficult to remove, followed by organophosphates, and carbamates; residues remaining after laundering for each class were 5.56%, 3.49%, and 0.10% for organochlorine, organophosphates, and carbamates, respectively (Keaschall et al., 1986). Inaccuracies can occur if one laundry treatment is recommended as advantageous for all pesticides or pesticide classes.

The level of active ingredient was a significant variable when tested in laundering research. The doubling of concentrations generally caused decreasing rates of

removal. The level of methyl parathion concentration is inversely related to the amount of residue removed through laundry (Easley, Laughlin, Gold, & Hill, 1982). Finley reported washing to be less effective as the amount of residue in garments increased. Furthermore, a second wash did not remove as much methyl parathion residues (on a % basis) as the initial washing (Finley et al., 1979).

A 1.25% concentrate of methyl parathion was more completely removed during laundering than full strength (54% a.i.) contamination. After the third cycle, the amount of methyl parathion removed was consistently in excess of 99%. However, following ten multiple launderings, residues of an undiluted methyl parathion contaminant can be readily detected in fabrics (Easley, Laughlin, Gold, & Hill, 1982). It is recommended therefore, that clothing contaminated with high pesticide concentrations be disposed of by burning or burial, as the fabric remains unsafe to the wearer (Easley, Laughlin, Gold, & Hill, 1982).

Laundry Methods used for Pesticide Worker Clothing

Surveys of pesticide workers reveal that frequently laundering procedures used do not follow recommended guidelines. Clothing worn during pesticide use was changed daily by only 52% of users (Rigakis et al., 1987). A portion of one sample indicated that it was safe to wear the same clothes day after day without washing them (Rucker et al., 1986).

One survey indicated that 43% of the sample stored pesticide soiled clothing with the family laundry at least sometimes (Rucker et al., 1986). In another survey, such clothing was stored with other clothing prior to laundry by 38% of respondents (Rigakis et al., 1987).

The same survey revealed that 41% washed a variety of clothing or other work clothing together with the pesticide soiled clothes (Rigakis et al., 1987). And 36% of another sample washed the clothing with the family laundry at least sometimes (Rucker et al., 1986).

Twenty-nine percent of the respondents to a Canadian survey laundered contaminated garments with other family wash. Granular or powdered detergent, bleach, and fabric softener were the most commonly used laundry products when cleaning contaminated garments (Cloud et al., 1983).

Transfer of Pesticides

Researchers have given limited attention to the transfer of pesticide soil to uncontaminated fabric. Laundering of pesticide soiled clothing separate from the family wash and the decontamination of the equipment was recommended to prevent cross-contamination in one of the earliest pesticide laundering studies (Finley et al., 1974).

In the Southwick et al. (1974) report of a case in which a man died of a repeated exposure to parathion, other clothing items also contained contamination. The levels of a pooled sample from three pair of pants and two

undergarments were 377 parts per million (ppm) and 324 ppm, respectively, compared to 2,552 ppm in the overalls identified as being a contributing cause of death. Contamination levels in the transfer items were about 14% of that in the overalls. Southwick et al. (1974) stated, "The contamination may have come from the association in laundering with the sprayed clothes."

Easley et al. (1983) and Laughlin et al. (1981) examined the transfer of pesticides from contaminated to clean fabrics during the laundering process. Although the amounts of pesticides transferred were small, recommendations were made for dedicated laundering and cleanup of laundering equipment.

Laughlin et al. (1981) found that the pesticide soil transferred from laundering equipment to clean fabric laundered in the equipment immediately following washing of methyl parathion soiled fabrics was <.01% of initial contamination of pesticide soiled fabric. There was a significant correlation ($r=0.63$) between the amount of methyl parathion on the contaminated denim fabric before laundering and the amount on the transfer fabric after laundering in contaminated equipment.

Easley et al. (1983) documented transfer of 2,4-D ester and 2,4-D amine pesticide soil from contaminated fabric to clean fabric during washing with the amount of transfer .02 to .2% of residues in the initial soiled fabric. The amount

transferred to other fabrics during concomitant laundering was dependent upon pesticide formulation (Easley et al., 1983).

Laundry variables were examined for their effect on pesticide transfer. Higher water temperatures were more effective in removing the ester formulation from the fabric, but also resulted in increased transfer of the pesticide to the untreated fabric (Easley et al., 1983).

The amount of 2,4-D ester or 2,4-D amine in the transfer fabric was not significantly different when a comparison was made between the heavy duty liquid detergent and the AATCC standard Detergent 124. This indicated that the amount of 2,4-D transferred was similar regardless of detergent type (Easley et al., 1983).

Repeated washing resulted in less 2,4-D ester or 2,4-D amine being absorbed by the transfer fabric (Easley et al., 1983). For the transfer fabrics, pre-rinse plus laundry resulted in significantly less 2,4-D ester transferred to concomitantly laundered fabric than those that were not pre-rinsed. However, the pre-rinsing step did not include the transfer fabric (Easley et al., 1983).

Braun et al. (1989) laundered untreated swatches along with spill simulated fabrics. These were found to pick up an average contamination of 85 ug pyrazophos which represented 3.8% of the amount found on the unwashed fabric.

Finley et al. (1979) indicated that clean test fabrics of the same type laundered with the C₁₄ methyl parathion spiked fabrics were just as radioactive as the spiked fabrics. Finley et al. (1974) found laundering of contaminated fabrics with uncontaminated fabrics resulted in a transference of insecticide residue with the amount of pesticide transferred in the range of 0-2% of residues in the initial soiled fabric.

Transfer of pesticide residue was also found to occur during dry cleaning. Chlorpyrifos residue transferred from the contaminated specimens to uncontaminated fabric specimens when the specimens were refurbished simultaneously. Residue transferred in this study were minute, ranging from .002 to .006 ug/cm² (Ringenberg, Laughlin, & Gold, 1988).

CHAPTER III

METHODOLOGY

This research investigated the pesticide cross-contamination that occurs during laundering. The effect of pesticide, water temperature, and fabric combination were examined.

Fabrics

The four fabrics tested were two top weight fabrics--100% cotton knit and a 65/35% cotton/polyester blend chambray--and two bottom weight fabrics--100% cotton denim and a 65/35% polyester/cotton blend twill fabric treated with a fluorocarbon finish. A survey of work-wear catalogs showed these fabrics to be common in men's work clothing and therefore of the type worn by agricultural workers when applying pesticides.

All fabrics were initially stripped of any residual finishes from the fabrication process by washing them one complete cycle, using the washing procedure outlined in American Association of Textile Chemists and Colorists (AATCC) Test Method 135-1987: Dimensional Changes in Automatic Home Laundering of Woven and Knit Fabrics (AATCC, 1988). The fabrics were cut into a sample size of 4 x 4

inches. A 3 x 3 inch specimen was marked at the center of each sample.

Treatment of Fabrics with Pesticides

Pesticide dilutions were prepared from concentrate formulation at 1.25% active ingredient (a.i.), a common concentration for agricultural application. Fabrics of each type were placed on paraffin film (Parafilm) and 1 ml of the pesticide solution was spiked onto the specimen using an Eppendorf digital pipette. The pesticide spiked specimens were allowed to air dry.

Three pesticides were used to contaminate the fabrics. The pesticides selected for use in this study and listed in Table 1 are atrazine, Diazinon, and metolachlor. The three pesticides are among those used in North Carolina, represent

Table 1

Common and Brand Names of Pesticides their Chemical Class, Usage, and Toxicity

Common name	Brand name	Chemical class	Usage ^a	Toxicity
Atrazine	Aatrex 4L	Triazine	Herb	Slight
Diazinon	Diazinon AG500	Organophosphate	Insect	Moderate
Metolachlor	Dual 8E	Chloroacetamide	Herb	Slight to moderate

^a Herb = Herbicide, Insect = Insecticide.

different chemical types, and vary in toxicity (CIBA-GEIGY Corporation, 1985a, 1985b, 1989).

Laundering Procedures

An Atlas Launder-Ometer equipped with stainless steel containers (3 x 5 in) was used to simulate home laundering for the wash cycle. Teflon liners were used in the lids to prevent retention of pesticides by the rubber gaskets. Individual Launder-Ometer canisters allowed for isolated laundering of fabric combinations and steel balls added to each canister simulated laundry agitation. Two water temperatures were examined for pesticide cross-contamination.

Pesticide soiled fabric samples were individually laundered along with fabrics that had not been exposed to pesticides in 500 ml containers. The fabrics were tested in combinations that may be expected to occur when more than one fabric type is laundered in a single washer load. Each fabric was laundered in combination with the same fabric and with the other fabric of the same weight classification (top weight or bottom weight). There were five replications. The eight combinations were:

Pesticide Soiled

Denim
Denim
Chambray
Chambray
Knit
Knit
Twill
Twill

Pesticide Free

Denim
Twill
Chambray
Knit
Knit
Chambray
Twill
Denim

The fabrics were laundered in an Atlas Launder-Ometer using a procedure modified from AATCC test method 61-1986, Colorfastness to Laundering, Home, and Commercial: Accelerated (AATCC, 1988) to simulate one home laundry cycle. The laundry cycle included a preheating cycle, in which 200 ml of the detergent solution and 10 steel balls were placed in each canister. The Launder-Ometer was run for 20 minutes to preheat them to the appropriate temperature before the fabric was added.

Contaminated specimens were Launder-Ometer laundered in cold water, with a wash temperature of $27^{\circ}\text{C} \pm 3\text{C}$ ($80 \pm 5\text{F}$) or in hot, with a wash temperature of $60^{\circ}\text{C} \pm 3\text{C}$ ($140 \pm 5\text{F}$). These wash temperatures are designations II and V respectively as established in Standardization of Home Laundry Test Conditions from the AATCC Technical Manual (1988). The use of the Atlas Launder-Ometer enabled the researcher to control the water temperature to the narrow range defined above. The wash cycle was limited to nine minutes to represent one home laundry cycle.

The detergent used was a 0.5% solution of a heavy duty carbonate detergent that is recommended for laundering pesticide soiled clothing. This detergent and water temperature combination had been shown to affect pesticide removal under laboratory conditions (Finley et al., 1977).

After laundering, the water was decanted and the samples were rinsed three times at 27°C . Each sample was

lightly squeezed to remove water. Specimens were air dried. Jars, liners, gaskets, and steel balls were decontaminated with acetone after each use. After drying, the marked 3 x 3 inch specimen was cut from each sample for extraction.

Preliminary Extraction Analysis

A preliminary extraction analysis of fabrics was done to test the efficiency of the procedure for residue recovery from the samples. Six extraction methods were tested with each pesticide to determine the method and solvent with the best extraction efficiency for the individual pesticides.

Samples measuring 3 x 3 inches were individually extracted in 40 ml of solvent in a 250 ml amber bottle with a teflon lined cap. Bottles were placed in an Eberbach shaker for 30 minutes. The sample was removed and placed in a second bottle with 40 ml of solvent for a second shaking. At the end of the 60 minute extraction time, the fabric specimen was removed and discarded and the two extracts combined. Each pesticide was tested for extractability using the mechanical shaker with HPLC (High Pressure Liquid Chromatography) grade acetone as the solvent. Each pesticide was also tested using the mechanical shaker with Optima grade chloroform as the solvent and with HPLC grade methylene chloride as the solvent. A third extraction did not exhibit additional detectable pesticide with the solvent used. This was consistent with the findings of Easter, Leonas, and DeJonge (1983).

Each pesticide was also extracted using a soxhlet system with HPLC grade acetone as the solvent. Specimens were placed into a soxhlet unit and individually extracted with 60 ml of solvent with a reflux time of 45 minutes. Each pesticide was also extracted using the soxhlet system with Optima grade chloroform as the solvent and with HPLC grade methylene chloride as the solvent.

As a control, samples were cut from each of the four fabrics and extracted. No detectable pesticide residues or other extractable components that could interfere with analysis were found.

Extraction of Residues

Acetone was chosen as the solvent for the extraction of atrazine in combination with the mechanical shaker. The mean recovery of atrazine from spiked samples was 89%.

It was necessary to try an additional extraction method for the extraction of metolachlor in order to receive an acceptable extraction efficiency. The samples tested with metolachlor were extracted with Optima grade chloroform for the first shaking and then with HPLC grade ethyl acetate for the second shaking using the Eberbach shaker. The mean recovery of metolachlor from spiked samples was 75%.

HPLC grade methylene chloride was chosen as the solvent for Diazinon in combination with the soxhlet extraction system. The mean recovery of Diazinon from spiked samples was 85%.

A nitrogen evaporator was used to provide a concentration acceptable for gas chromatograph analysis. Extracts from the fabrics that had not been spiked with pesticide were reduced to 2 ml.

Analysis of Residues

Analysis of the extracts were made using a Tracor 540 gas chromatograph (GC) with a hydrogen flame detector. Injections of 4 ul were made from each sample using a 10 ul Hamilton syringe. Standard solutions of pesticides were injected before and after each series of tests. With a column temperature of 200°C, the retention time for atrazine was 4.8 minutes and the retention time for Diazinon was 5.1 minutes. At a column temperature of 230°C, the retention time for metolachlor was 5.5 minutes. GC data was output to a Spectra-Physics 4290 integrator for determination of peak areas and retention times.

Statistical Analysis of Data

Data for the study were examined with ANOVA with a 2 x 3 x 8 factorial arrangement of treatments (2 water temperatures x 3 pesticides x 8 fabric combinations) in a completely randomized design. Five replications per treatment combination were used.

CHAPTER IV

RESULTS

This study investigated the pesticide cross-contamination that occurs during laundering. The effect of pesticide, water temperature, and fabric combination were examined. Three pesticides--atrazine, Diazinon, and metolachlor--were chosen along with two water temperatures, 27°C and 60°C, and in conjunction with eight fabric combinations. There were five replications.

The results are reported in seven sections: effect of pesticide; effect of water temperature, effect of fabric combination; interactions of pesticide and water temperature; interactions of pesticide and fabric combination; interactions of water temperature and fabric combination; and finally interactions of pesticide, water temperature, and fabric combination.

An analysis of variance procedure (ANOVA) was used to test for the main effects (temperature, pesticide, and fabric combination) and interactions of the main effects. Further examination of all possible pairwise comparisons of treatment means was carried out with a Duncan's multiple range post hoc procedure with $p < .05$ as indication of significance. Averages of the micrograms (ug) of pesticide

deposited on the unspiked fabric swatches laundered with spiked fabric swatches at two water temperatures, with eight fabric combinations, and using three pesticides are shown in Table 2. Residues extracted from fabrics range from 0.0 to 350.2 ug with a mean level of 60.0 ug.

Table 2

Degree of Pesticide Cross-Contamination after Laundering

Variable	Mean ug	% Contamination
Pesticide		
Atrazine	1.3	0.01
Diazinon	161.2	1.17
Metolachlor	17.4	0.13
Water temperature		
Cold	54.2	0.39
Hot	65.8	0.48
Fabric combination ^a		
Chambray-chambray	30.5	0.22
Chambray-knit	78.2	0.57
Denim-denim	85.6	0.62
Denim-twill	23.6	0.17
Knit-chambray	30.4	0.22
Knit-knit	79.6	0.58
Twill-denim	122.5	0.89
Twill-twill	29.4	0.21

^aFabric combinations indicate spiked-unspiked fabric.

Effect of Pesticide

A statistical analysis of the data indicated a significant difference in cross-contamination among pesticides [$F(2, 192) = 1459.30, p < .0001$]. Duncan's multiple range test ($p < .05$) was used to determine the

location of the between pesticide variation. There were significant differences between each of the pesticides (Table 3). The Diazinon had the greatest level of cross-contamination and the atrazine the least.

Table 3

Duncan's Multiple Range Comparison of the Degree of Pesticide Cross-Contamination that Occurred with each Pesticide

Group	Pesticide	n	Mean ug
A	Diazinon	80	161.2
B	Metolachlor	80	17.4
C	Atrazine	80	1.3

Note. Means with the same group letter are not significantly different at $p < .05$.

Effect of Water Temperature

A statistical analysis of the data indicated a significant difference in cross-contamination between the water temperatures at the .0001 level of significance [$F(1, 192) = 18.85$]. More pesticide was deposited on the unspiked fabric when laundered in hot water with a pesticide spiked fabric as shown in Table 4.

Table 4

Duncan's Multiple Range Comparison of the Degree of Pesticide Cross-Contamination that Occurred with Two Water Temperatures (Temp)

Group	Temp	n	Mean ug
A	Hot	120	65.8
B	Cold	120	54.2

Note. Means with the same group letter are not significantly different at $p < .05$.

Effect of Fabric Combination

The ANOVA indicated a significant difference in cross-contamination among the eight fabric combinations [$F(2, 192) = 93.70, p < .0001$]. Duncan's multiple range test ($p < .05$) was used to determine the location of the between fabric combination variation (Table 5).

Pesticide cross-contamination levels were similar for the unspiked fabric whether it was laundered with a spiked fabric of the same fabric type or of a different fabric type from the unspiked fabric. For example, the mean cross-contamination was 79.6 for unspiked knit fabric laundered with spiked knit fabric and 78.2 for unspiked knit fabric laundered with spiked chambray fabric. The only unspiked fabric that did not fit this pattern was denim. The combination of spiked twill fabric laundered with unspiked

denim fabric resulted in the highest level of cross-contamination of any fabric combination. The level of cross-contamination was significantly different from all other fabric combinations.

Table 5

Duncan's Multiple Range Comparison of the Degree of Pesticide Cross-Contamination that Occurred with each of the Fabric Combinations

Group	Fabric combination ^a	n	Mean ug
A	Twill-denim	30	122.5
B	Denim-denim	30	85.6
B	Knit-knit	30	79.6
B	Chambray-knit	30	78.2
C	Chambray-chambray	30	30.5
C	Knit-chambray	30	30.4
C	Twill-twill	30	29.4
C	Denim-twill	30	23.6

Note. Means with the same group letter are not significantly different at $p < .05$.

^aFabric combinations indicate spiked-unspiked fabric.

The three fabric combinations of spiked denim laundered with unspiked denim, spiked knit laundered with unspiked knit, and spiked chambray laundered with unspiked knit were not significantly different from each other, but had a significantly greater level of cross-contamination than the four remaining fabric combinations. Those four fabric

combinations--chambray-chambray, knit-chambray, twill-twill, and denim-twill--did not differ significantly from each other in the level of cross-contamination that occurred when an unspiked fabric was laundered with a pesticide spiked fabric.

Interactions of Pesticide and Water Temperature

Table 6 shows the relation between the two variables pesticide and water temperature. This interaction is significant at the .0001 level [$F(2, 192) = 14.88$]. Duncan's multiple range test ($p < .05$) was used to determine the location of the variation between the interaction combinations.

For each water temperature, the order of the contamination was the same, Diazinon had the greatest level and atrazine had the least. There was a significantly higher level of cross-contamination in hot water for Diazinon than for all other combinations. In addition, the level of cross-contamination was higher for Diazinon in cold water than atrazine or metolachlor in either water temperature. Atrazine showed no cross-contamination in hot water and a small amount in cold water. There was not a significant difference among the means of metolachlor and atrazine in either water temperature.

Table 6

Duncan's Multiple Range Comparison of the Interaction of Pesticide and Water Temperature (Temp) and the Degree of Pesticide Cross-Contamination that Occurred

Group	Pesticide	Temp	Mean ug
A	Diazinon	Hot	176.9
B	Diazinon	Cold	145.4
C	Metolachlor	Hot	20.3
C	Metolachlor	Cold	14.5
C	Atrazine	Cold	2.7
C	Atrazine	Hot	0.0

Note. Means with the same group letter are not significantly different at $p < .05$. $n = 40$ for each pesticide and temperature combination.

Interactions of Pesticide and Fabric Combination

The interaction between pesticide and fabric combination shown in Table 7 is significant [$F(2, 192) = 54.41, p < .0001$]. Duncan's multiple range test ($p < .05$) was used to determine the location of the variation between the interaction combinations.

Pesticide cross-contamination levels were not significantly different for the unspiked fabric whether it was laundered with a spiked fabric of the same fabric type or of a different fabric type from the unspiked fabric. For example, the mean Diazinon cross-contamination was 238.7 for

Table 7

Duncan's Multiple Range Comparison of the Interaction of Pesticide and Fabric Combination and the Degree of Pesticide Cross-Contamination that Occurred

Group	Pesticide	Fabric combination ^a	Mean ug
A	Diazinon	Twill-denim	293.4
B	Diazinon	Knit-knit	238.7
B	Diazinon	Chambray-knit	234.5
C	Diazinon	Denim-denim	183.8
D	Diazinon	Chambray-chambray	91.5
D	Diazinon	Knit-chambray	91.1
DE	Diazinon	Twill-twill	88.3
DEF	Metolachlor	Twill-denim	74.2
EF	Diazinon	Denim-twill	68.0
F	Metolachlor	Denim-denim	62.3
G	Atrazine	Denim-denim	10.8
G	Metolachlor	Denim-twill	2.8
G	Atrazine	Chambray-chambray	0.0
G	Atrazine	Chambray-knit	0.0
G	Atrazine	Denim-twill	0.0
G	Atrazine	Knit-chambray	0.0
G	Atrazine	Knit-knit	0.0
G	Atrazine	Twill-denim	0.0
G	Atrazine	Twill-twill	0.0
G	Metolachlor	Chambray-chambray	0.0
G	Metolachlor	Chambray-knit	0.0
G	Metolachlor	Knit-chambray	0.0
G	Metolachlor	Knit-knit	0.0
G	Metolachlor	Twill-twill	0.0

Note. Means with the same group letter are not significantly different at $p < .05$. Samples with more than one group letter indicate overlapping of groups. $n = 10$ for each pesticide and fabrics combination.

^aFabric combinations indicate spiked-unspiked fabric.

unspiked knit fabric laundered with spiked knit fabric and 234.5 for unspiked knit fabric laundered with spiked chambray fabric. The only unspiked fabric that did not fit this pattern was denim laundered with fabrics spiked with Diazinon. When unspiked denim was laundered with Diazinon spiked twill, the cross-contamination was significantly greater than all other interaction combinations.

All fabric combinations showed pesticide cross-contamination when Diazinon was the pesticide used. Also each fabric combination had greater cross-contamination with Diazinon than with the other two pesticides.

Unspiked denim fabrics laundered with a metolachlor spiked twill or denim fabric showed the greatest level of metolachlor cross-contamination. These levels were significantly higher than all atrazine combinations and the six other metolachlor combinations.

The low level of metolachlor cross-contamination when unspiked twill fabric was laundered with spiked denim fabric and the low level of atrazine cross-contamination when unspiked denim fabric was laundered with spiked denim was not significantly different from those interaction combinations that had no measurable pesticide cross-contamination. Regardless of the fabric combination, chambray and knit fabrics were not cross-contaminated with metolachlor.

Interactions of Water Temperature and
Fabric Combination

Table 8 shows the relation between the two variables water temperature and fabric combination. This interaction was not significant at the .05 level [$F(7, 192) = 1.96$, $p=.0630$]. Because the interaction approached significance, Duncan's multiple range test ($p<.05$) was performed. A significant difference in means was found among the interaction combinations.

For each fabric combination, more pesticide was deposited on the unspiked fabric when laundered in hot water with a pesticide spiked fabric than in cold water. Pesticide cross-contamination levels were not significantly different for the unspiked fabrics whether it was laundered at the same temperature with a spiked fabric of the same type or of a different fabric type from the unspiked fabric. The only unspiked fabric that did not fit this pattern was unspiked denim laundered with spiked fabrics in either hot or cold water.

The pesticide cross-contamination of unspiked denim fabrics laundered with spiked twill fabrics in hot water was significantly higher than all other interaction combinations. The pesticide cross-contamination of unspiked denim fabrics laundered with spiked twill fabrics in cold water was significantly higher than unspiked denim laundered with spiked denim fabric in cold water.

Table 8

Duncan's Multiple Range Comparison of the Interaction of
Water Temperature (Temp) and Fabric Combination and the
Degree of Pesticide Cross-Contamination that Occurred

Group	Temp	Fabric combination ^a	Mean ug
A	Hot	Twill-denim	136.3
B	Cold	Twill-denim	108.7
BC	Hot	Denim-denim	93.9
CD	Hot	Chambray-knit	90.7
CDE	Hot	Knit-knit	84.8
DEF	Cold	Denim-denim	77.4
EF	Cold	Knit-knit	74.3
F	Cold	Chambray-knit	65.7
G	Hot	Chambray-chambray	34.6
G	Hot	Twill-twill	31.0
G	Hot	Knit-chambray	30.6
G	Cold	Knit-chambray	30.2
G	Cold	Twill-twill	27.9
G	Cold	Chambray-chambray	26.4
G	Hot	Denim-twill	24.3
G	Cold	Denim-twill	23.0

Note. Means with the same group letter are not significantly different at $p < .05$. Samples with more than one group letter indicate overlapping of groups. $n = 15$ for each temperature and fabrics combination.

^aFabric combinations indicate spiked-unspiked fabric.

Regardless of water temperature, there was no significant difference in the pesticide cross-contamination levels for unspiked chambray and twill fabrics whether laundered with a spiked fabric of the same fabric type or of

a different fabric type from the unspiked fabric. The denim and knit fabrics had significantly higher levels of pesticide cross-contamination than the chambray and twill fabrics regardless of water temperature and fabric combination.

Interactions of Pesticide, Water Temperature,
and Fabric Combination

The relationship among the three variables--pesticide, water temperature, and fabric combination--is shown in Table 9. The interaction is statistically significant [$F(14, 192) = 1.91, p=.0278$]. Duncan's multiple range test ($p<.05$) was used to determine the location of the variation between the interaction combinations.

Diazinon cross-contamination was greater in hot water than in cold for each fabric combination. Eight of the Diazinon interaction combinations were significantly greater than the forty other combinations. Unspiked denim fabric laundered with Diazinon spiked twill fabric laundered in hot water received significantly more cross-contamination (313.4 ug) than any other combination.

Regardless of water temperature and fabric combination, all mean Diazinon cross-contaminations were significantly greater than the other pesticides with the exception of unspiked denim laundered with fabrics spiked with metolachlor. Metolachlor cross-contamination was found primarily in the unspiked denim fabrics. There was a higher

level of cross-contamination of the denim fabrics in hot water (95.5 ug, 67.3 ug) than in cold (57.4 ug, 52.8 ug). These means were not significantly different from the mean Diazinon cross-contamination of chambray and twill fabrics. Chambray and twill fabrics had lower levels of diazinon

Table 9

Duncan's Multiple Range Comparisons of the Interaction of Pesticide (Pest), Water Temperature (Temp), and Fabric Combination and the Degree of Pesticide Cross-Contamination that Occurred

Group	Pest ^a	Temp	Fabric combination ^b	Mean ug
A	Dzn	Hot	Twill-denim	313.4
B	Dzn	Cold	Twill-denim	273.4
B	Dzn	Hot	Chambray-knit	272.0
B	Dzn	Hot	Knit-knit	254.4
C	Dzn	Cold	Knit-knit	223.0
C	Dzn	Hot	Denim-denim	214.3
C	Dzn	Cold	Chambray-knit	197.0
D	Dzn	Cold	Denim-denim	153.4
E	Dzn	Hot	Chambray-chambray	103.8
EF	Met	Hot	Twill-denim	95.5
EFG	Dzn	Hot	Twill-twill	92.9
EFG	Dzn	Hot	Knit-chambray	91.7
EFG	Dzn	Cold	Knit-chambray	90.6
EFGH	Dzn	Cold	Twill-twill	83.7
EFGHI	Dzn	Cold	Chambray-chambray	79.2
FGHI	Dzn	Hot	Denim-twill	72.8
FGHI	Met	Hot	Denim-denim	67.3
GHI	Dzn	Cold	Denim-twill	63.2
HI	Met	Cold	Denim-denim	57.4
I	Met	Cold	Twill-denim	52.8

Table 9 (cont.)

Group	Pest ^a	Temp	Fabric combination ^b	Mean ug
J	Atz	Cold	Denim-denim	21.5
J	Met	Cold	Denim-twill	5.6
J	Atz	Cold	Chambray-chambray	0.0
J	Atz	Cold	Chambray-knit	0.0
J	Atz	Cold	Denim-twill	0.0
J	Atz	Cold	Knit-chambray	0.0
J	Atz	Cold	Knit-knit	0.0
J	Atz	Cold	Twill-denim	0.0
J	Atz	Cold	Twill-twill	0.0
J	Atz	Hot	Chambray-chambray	0.0
J	Atz	Hot	Chambray-knit	0.0
J	Atz	Hot	Denim-denim	0.0
J	Atz	Hot	Denim-twill	0.0
J	Atz	Hot	Knit-chambray	0.0
J	Atz	Hot	Knit-knit	0.0
J	Atz	Hot	Twill-denim	0.0
J	Atz	Hot	Twill-twill	0.0
J	Met	Cold	Chambray-chambray	0.0
J	Met	Cold	Chambray-knit	0.0
J	Met	Cold	Knit-chambray	0.0
J	Met	Cold	Knit-knit	0.0
J	Met	Cold	Twill-twill	0.0
J	Met	Hot	Chambray-chambray	0.0
J	Met	Hot	Chambray-knit	0.0
J	Met	Hot	Denim-twill	0.0
J	Met	Hot	Knit-chambray	0.0
J	Met	Hot	Knit-knit	0.0
J	Met	Hot	Twill-twill	0.0

Note. Means with the same group letter are not significantly different at $p < .05$. Samples with more than one group letter indicate overlapping of groups. $n = 5$ in every combination.

^aAtz = Atrazine, Dzn = Diazinon, Met = Metolachlor. ^bFabric combinations indicate spiked-unspiked fabric.

cross-contamination than denim and knit fabrics regardless of water temperature.

The only atrazine cross-contamination (21.5 ug) was found when unspiked denim fabric was laundered with spiked denim in cold water. The only unspiked twill fabric that was cross-contaminated with metolachlor (5.6 ug) was laundered with spiked denim fabric in cold water. These low levels of pesticide cross-contamination were not significantly different from those interaction combinations that had no measurable pesticide cross-contamination. Regardless of the fabric combination and water temperature, chambray and knit fabrics were not cross-contaminated with metolachlor.

CHAPTER V
SUMMARY AND CONCLUSIONS

The purpose of this research was to determine the effects of pesticide, water temperature, and fabric combination on the level of cross-contamination that occurs during laundering of pesticide soiled fabrics with unsoiled fabrics under carefully controlled test conditions.

The objectives of the study were the following:

1. To develop laboratory test procedures that will assess the degree of cross-contamination that occurs when pesticide spiked fabrics are laundered with pesticide free fabrics.
2. To measure the cross-contamination that occurs during laundering with three different pesticides.
3. To measure the cross-contamination that occurs during laundering with two water temperatures.
4. To measure the cross-contamination that occurs during laundering with four different test fabrics in eight combinations of fabrics.

Three pesticides, atrazine, Diazinon, and metolachlor were selected for this study because they are among those used in North Carolina, represent different chemical types, and vary in toxicity. In addition, there was little in the literature regarding the laundering of these specific pesticides.

Two water temperatures 27°C and 60°C were selected for this study since they are representative of those chosen by consumers and previous laundering studies have shown water temperature to be a significant factor in pesticide residue removal.

Four fabrics--100% cotton knit, 65/35% cotton/polyester chambray, 100% cotton denim, and a 65/35% polyester/cotton blend khaki twill fabric treated with a fluorocarbon finish--were tested in eight combinations. These fabrics were chosen because they were representative of those available in work-wear clothing for agricultural workers according to the literature.

Eight fabric combinations were tested. Pesticide soiled fabric samples were individually laundered along with fabrics that had not been exposed to pesticides in an Atlas Launder-Ometer. Each fabric was tested in combination with itself and in combination with the other fabric of the same weight classification (top or bottom weight).

The testing of the fabrics for pesticide cross-contamination resulted in the rejection of the following hypotheses.

Hypothesis 1. There will be a difference among the pesticides in the level of cross-contamination that occurs when a pesticide free fabric is laundered under laboratory test conditions with a fabric that is spiked with pesticide.

The ANOVA results indicated that there were significant ($p < .0001$) differences among the pesticides in the level of pesticide cross-contamination of the unspiked fabrics. The differences between each of the pesticides, atrazine, Diazinon, and metolachlor, were significant ($p < .05$). On this basis, Hypothesis I was supported.

Regardless of water temperature and fabric combination, all mean Diazinon cross-contaminations were significantly greater than the other pesticides with the exception of unspiked denim laundered with fabrics spiked with metolachlor. Metolachlor cross-contamination was found primarily in the unspiked denim fabrics. The only atrazine cross-contamination was found when unspiked denim fabric was laundered with spiked denim in cold water.

Hypothesis 2. There will be a difference between the water temperatures in the level of cross-contamination that occurs when a pesticide free fabric is laundered under laboratory test conditions with a fabric that is spiked with pesticide.

The pesticide cross-contamination was significantly greater in hot water ($p < .0001$) than in cold water. Therefore, Hypothesis II was supported.

Diazinon and metolachlor cross-contamination was greater in hot water than in cold. Atrazine had a very low level of cross-contamination of denim fabrics in cold water and did not cross-contaminate in hot water.

Hypothesis 3. There will be a difference in the level of cross-contamination that occurs when a pesticide free fabric is laundered under laboratory test conditions with an identical or a different fabric of the same weight class (top or bottom) that is spiked with pesticide.

The ANOVA results indicated that there were significant ($p < .0001$) differences among the fabric combinations in the level of cross-contamination that occurred on the unspiked fabric samples. Specifically the combination of spiked twill and unspiked denim had significantly greater ($p < .05$) cross-contamination than all other fabric combinations. The combinations spiked and unspiked denim, spiked and unspiked knit, and spiked chambray with unspiked knit while not significantly different from each other had a significantly ($p < .05$) greater level of cross-contamination than the remaining four combinations. Those combinations of spiked and unspiked chambray, spiked and unspiked twill, spiked knit with unspiked chambray, and spiked denim with unspiked twill were not significantly different from each other in the level of cross-contamination that occurred. On this basis, Hypothesis III was supported.

With the exception of unspiked denim, the level of cross-contamination in unspiked fabrics was not significantly different whether the unspiked fabric was laundered with an identical spiked fabric or with a spiked fabric of the same weight class.

Denim fabrics readily accepted cross-contamination of pesticide residues. When unspiked denim were laundered with spiked twill there was a higher level of cross-contamination than when laundered with spiked denim fabrics. Fabrics with a fluorocarbon finish have been shown to resist pesticide residue, therefore there was more residue available in the wash solution for the denim fabric to absorb.

Knit fabrics also readily accepted cross-contamination of pesticide residues. The level of cross-contamination was not significantly affected by the type of spiked fabric.

General Conclusions

The analysis of data resulted in the following conclusions.

1. Those laundering conditions that have been shown to be more effective for removal of pesticide residue provide an opportunity for more pesticide to cross-contaminate fabrics laundered in the same load. Specifically, the hot water temperature allowed more cross-contamination.
2. Cross-contamination of pesticide residue is dependent on pesticide with individual pesticides exhibiting very different patterns of residue transfer.
3. The fabric combination of pesticide soiled fabric and unsoiled fabric influences the level of cross-contamination that occurs. Fabrics that allow for greater removal of pesticide residue provide an opportunity for more pesticide to be transferred to other fabrics. Certain fabrics accept more readily the transfer of pesticide residue.

The results of this study support the recommendation that pesticide soiled clothing be laundered separately from uncontaminated clothing.

Recommendations for Further Study

1. Identify the fabric characteristics that effect the ability to accept pesticide. Fiber content, fabric finish, and physical properties including weight and thickness should be included.
2. Test for increase in pesticide cross-contamination residue over multiple launderings with pesticide soiled fabrics.
3. Test for pesticide cross-contamination with additional pesticides, fabric combinations, and detergent formulations and concentrations.
4. Determine the relationship between pesticide removed from contaminated fabric and the amount of pesticide cross-contamination.

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APPENDIX A.

DATA

SAMPLE CODING:

First letter: Water temperature used for laundering,
C = Cold, H = Hot

Second letter: Pesticide, A = Atrazine, D = Diazinon,
M = Metolachlor

Third letter: Fabric that was spiked with pesticide,
C = Chambray, D = Denim, K = Knit, T = Twill

Fourth letter: Fabric that was laundered with the pesticide
spiked fabric and originally contained no pesticide, the
code is the same as for the third letter.

Fifth letter: Replication, A, B, C, D, or E

Sample	AREA	PPM	ug
CDCC A	359	45.8	91.6
CDCC B	292	37.2	74.5
CDCC C	330	42.1	84.2
CDCC D	266	33.9	67.9
CDCC E	304	38.8	77.6
CDCK A	806	102.9	205.8
CDCK B	782	99.8	199.7
CDCK C	790	100.8	201.7
CDCK D	849	108.4	216.8
CDCK E	629	80.3	160.6
CDDD A	727	92.8	185.6
CDDD B	553	70.6	141.2
CDDD C	533	68.0	136.1
CDDD D	572	73.0	146.1
CDDD E	618	78.9	157.8
CDDT A	214	27.3	54.6
CDDT B	213	27.2	54.4
CDDT C	204	26.0	52.1
CDDT D	299	38.1	76.3
CDDT E	308	39.3	78.6
CDKC A	341	43.5	87.1
CDKC B	393	50.1	100.3
CDKC C	416	53.1	106.2
CDKC D	365	46.6	93.2
CDKC E	258	32.9	65.9
CDKK A	788	100.6	201.2
CDKK B	826	105.4	210.9
CDKK C	894	114.1	228.3
CDKK D	953	121.7	243.4
CDKK E	905	115.5	231.1
CDTD A	886	113.1	226.3
CDTD B	1101	140.6	281.2
CDTD C	1035	132.1	264.3

CDTD	D	1081	138.0	276.1
CDTD	E	1249	159.5	319.0
CDTT	A	224	28.6	57.2
CDTT	B	270	34.4	68.9
CDTT	C	346	44.1	88.3
CDTT	D	311	39.7	79.4
CDTT	E	488	62.3	124.6
HDCC	A	707	90.2	180.5
HDCC	B	277	35.3	70.7
HDCC	C	238	30.3	60.7
HDCC	D	330	42.1	84.2
HDCC	E	480	61.3	122.6
HDCK	A	1146	146.3	292.7
HDCK	B	1244	158.8	317.7
HDCK	C	717	91.5	183.1
HDCK	D	1018	130.0	260.0
HDCK	E	1199	153.1	306.2
HDDD	A	839	107.1	214.3
HDDD	B	832	106.2	212.5
HDDD	C	638	81.4	162.9
HDDD	D	849	108.4	216.8
HDDD	E	1037	132.4	264.8
HDDT	A	375	47.8	95.7
HDDT	B	266	33.9	67.9
HDDT	C	186	23.7	47.5
HDDT	D	231	29.5	59.0
HDDT	E	367	46.8	93.7
HDKC	A	274	34.9	69.9
HDKC	B	425	54.2	108.5
HDKC	C	260	33.2	66.4
HDKC	D	476	60.7	121.5
HDKC	E	360	45.9	91.9
HDKK	A	1009	128.8	257.7
HDKK	B	1345	171.7	343.5
HDKK	C	1064	135.8	271.7
HDKK	D	789	100.7	201.5
HDKK	E	774	98.8	197.7
HDTD	A	1371	175.0	350.1
HDTD	B	1182	150.9	301.9
HDTD	C	1308	167.0	334.1
HDTD	D	1269	162.0	324.1
HDTD	E	1005	128.3	256.7
HDTT	A	493	62.9	125.9
HDTT	B	266	33.9	67.9
HDTT	C	391	49.9	99.8
HDTT	D	338	43.1	86.3
HDTT	E	331	42.2	84.5
CACC	A	0	0.0	0.0
CACC	B	0	0.0	0.0
CACC	C	0	0.0	0.0
CACC	D	0	0.0	0.0
CACC	E	0	0.0	0.0

CACK A	0	0.0	0.0
CACK B	0	0.0	0.0
CACK C	0	0.0	0.0
CACK D	0	0.0	0.0
CACK E	0	0.0	0.0
CADD A	349	26.1	52.3
CADD B	0	0.0	0.0
CADD C	368	27.5	55.1
CADD D	0	0.0	0.0
CADD E	0	0.0	0.0
CADT A	0	0.0	0.0
CADT B	0	0.0	0.0
CADT C	0	0.0	0.0
CADT D	0	0.0	0.0
CADT E	0	0.0	0.0
CAKC A	0	0.0	0.0
CAKC B	0	0.0	0.0
CAKC C	0	0.0	0.0
CAKC D	0	0.0	0.0
CAKC E	0	0.0	0.0
CAKK A	0	0.0	0.0
CAKK B	0	0.0	0.0
CAKK C	0	0.0	0.0
CAKK D	0	0.0	0.0
CAKK E	0	0.0	0.0
CATD A	0	0.0	0.0
CATD B	0	0.0	0.0
CATD C	0	0.0	0.0
CATD D	0	0.0	0.0
CATD E	0	0.0	0.0
CATT A	0	0.0	0.0
CATT B	0	0.0	0.0
CATT C	0	0.0	0.0
CATT D	0	0.0	0.0
CATT E	0	0.0	0.0
HACC A	0	0.0	0.0
HACC B	0	0.0	0.0
HACC C	0	0.0	0.0
HACC D	0	0.0	0.0
HACC E	0	0.0	0.0
HACK A	0	0.0	0.0
HACK B	0	0.0	0.0
HACK C	0	0.0	0.0
HACK D	0	0.0	0.0
HACK E	0	0.0	0.0
HADD A	0	0.0	0.0
HADD B	0	0.0	0.0
HADD C	0	0.0	0.0
HADD D	0	0.0	0.0
HADD E	0	0.0	0.0
HADT A	0	0.0	0.0
HADT B	0	0.0	0.0

HADT C	0	0.0	0.0
HADT D	0	0.0	0.0
HADT E	0	0.0	0.0
HAKC A	0	0.0	0.0
HAKC B	0	0.0	0.0
HAKC C	0	0.0	0.0
HAKC D	0	0.0	0.0
HAKC E	0	0.0	0.0
HAKK A	0	0.0	0.0
HAKK B	0	0.0	0.0
HAKK C	0	0.0	0.0
HAKK D	0	0.0	0.0
HAKK E	0	0.0	0.0
HATD A	0	0.0	0.0
HATD B	0	0.0	0.0
HATD C	0	0.0	0.0
HATD D	0	0.0	0.0
HATD E	0	0.0	0.0
HATT A	0	0.0	0.0
HATT B	0	0.0	0.0
HATT C	0	0.0	0.0
HATT D	0	0.0	0.0
HATT E	0	0.0	0.0
CMCC A	0	0.0	0.0
CMCC B	0	0.0	0.0
CMCC C	0	0.0	0.0
CMCC D	0	0.0	0.0
CMCC E	0	0.0	0.0
CMCK A	0	0.0	0.0
CMCK B	0	0.0	0.0
CMCK C	0	0.0	0.0
CMCK D	0	0.0	0.0
CMCK E	0	0.0	0.0
CMDD A	462	16.7	33.4
CMDD B	1711	62.0	24.0
CMDD C	527	19.1	38.2
CMDD D	573	20.7	41.5
CMDD E	686	24.8	49.7
CMDT A	0	0.0	0.0
CMDT B	0	0.0	0.0
CMDT C	0	0.0	0.0
CMDT D	389	14.0	28.1
CMDT E	0	0.0	0.0
CMKC A	0	0.0	0.0
CMKC B	0	0.0	0.0
CMKC C	0	0.0	0.0
CMKC D	0	0.0	0.0
CMKC E	0	0.0	0.0
CMKK A	0	0.0	0.0
CMKK B	0	0.0	0.0
CMKK C	0	0.0	0.0
CMKK D	0	0.0	0.0

CMKK E	0	0.0	0.0
CMTD A	783	28.3	56.7
CMTD B	613	22.2	44.4
CMTD C	767	27.7	55.6
CMTD D	773	28.0	56.0
CMTD E	706	25.5	51.1
CMTT A	0	0.0	0.0
CMTT B	0	0.0	0.0
CMTT C	0	0.0	0.0
CMTT D	0	0.0	0.0
CMTT E	0	0.0	0.0
HMCC A	0	0.0	0.0
HMCC B	0	0.0	0.0
HMCC C	0	0.0	0.0
HMCC D	0	0.0	0.0
HMCC E	0	0.0	0.0
HMCK A	0	0.0	0.0
HMCK B	0	0.0	0.0
HMCK C	0	0.0	0.0
HMCK D	0	0.0	0.0
HMCK E	0	0.0	0.0
HMDD A	1037	37.5	75.1
HMDD B	901	32.6	65.3
HMDD C	895	32.4	64.8
HMDD D	910	32.9	65.9
HMDD E	898	32.5	65.0
HMDT A	0	0.0	0.0
HMDT B	0	0.0	0.0
HMDT C	0	0.0	0.0
HMDT D	0	0.0	0.0
HMDT E	0	0.0	0.0
HMKC A	0	0.0	0.0
HMKC B	0	0.0	0.0
HMKC C	0	0.0	0.0
HMKC D	0	0.0	0.0
HMKC E	0	0.0	0.0
HMKK A	0	0.0	0.0
HMKK B	0	0.0	0.0
HMKK C	0	0.0	0.0
HMKK D	0	0.0	0.0
HMKK E	0	0.0	0.0
HMTD A	971	35.1	70.3
HMTD B	2254	81.6	163.3
HMTD C	1174	42.5	85.1
HMTD D	1004	36.3	72.7
HMTD E	1184	42.9	85.8
HMTT A	0	0.0	0.0
HMTT B	0	0.0	0.0
HMTT C	0	0.0	0.0
HMTT D	0	0.0	0.0
HMTT E	0	0.0	0.0