

# Comparing the Removal of Pesticide Residue from Clothing with Different Washing and Drying Methods.

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## Abstract

This study investigated numerous factors influencing the removal of carbaryl or permethrin from various types of clothing. These factors included application rate (1X or 9X), washing machine type (full-fill agitator or high efficiency), clothing type (blue jeans, work shirt, T-shirt, or cotton/polyester blend T-shirt), and drying method (electric dryer or clothesline). Additionally, this study examined transference to baby Onesies® during laundering and assessed the role of Ultraviolet-B (UV-B) exposure in reducing residues for articles dried on clotheslines. Contamination inside washing and drying machines and pesticide levels in wastewater were also examined. The results indicated that both washing machine types were effective at removing carbaryl and permethrin from the clothing. Among the different fabric types, blue jeans consistently retained more residues than other clothing types used in the study. Transference of pesticide to the Onesies® occurred with all pesticides at both rates, indicating pesticide-contaminated clothing should be laundered separately from all other laundry, including other work clothes or family clothes. Based on the findings of this study, we provide safety recommendations for applicators and laundering guidelines for effectively decontaminating clothing.

**KEYWORDS:** pesticide removal; clothing decontamination; pesticide transference; pesticides in wastewater

## 1. Introduction

In the United States, the pesticide product label lists the required personal protective equipment (PPE) for pesticide handlers or applicators. The EPA Label Review Manual states, *“All end-use occupational use products (WPS or non-WPS) need to have the minimum baseline label-required work clothes for handlers consisting of long-sleeved*

*shirt, long pants, socks and shoes. Technically these work clothes items are not considered PPE, but they can be required on labels (see 40 CFR 170.240 (b))*" (EPA Label Review Manual, 2016). According to the federal Worker Protection Standard (WPS), the responsibility for cleaning pesticide-contaminated clothing is based on whether the clothing is classified as PPE or work clothing (Fults, 2017). A 2012 survey of 1,868 EPA-registered pesticide product labels indicated that approximately 85% (1,583 labels) required long-sleeved shirt and long pants, not PPE such as cloth or chemical-resistant coveralls (Shaw & Harned, 2013). Therefore, in many cases, the pesticide handler or applicator is responsible for cleaning their work clothing.

Numerous publications address the appropriate methods to decontaminate or launder pesticide-contaminated clothing, but these publications were not based on research examining the use of modern, high-efficiency washing machines and dryers. Methods to remove pesticides from clothing were investigated thoroughly in the 1980s and 1990s, typically researched by analyzing pesticides applied to swatches of clothing and the remaining residues after using lab bench washing machines (Laughlin & Gold, 1988; Laughlin, 1993). However, today's washing machines have been designed to be more efficient by using less water and lower water temperatures during both the wash and rinse cycles. Additionally, phosphates, additives used to increase effectiveness of removing soil, oils, and grease, have been removed from the laundry detergents. These changes were significant enough to require additional research on decontaminating pesticide-contaminated clothing.

The research reported here investigated pesticide contamination and removal from various types of clothing that were washed and dried in commonly used household appliances. We investigated the efficacy of a front-loading, high-efficiency (HE) washer with no agitator and a traditional full-fill washer with an agitator to remove pesticide residues from contaminated clothing. We compared these two washing methods (hereafter referred to as HE and agitator) and provide recommendations based on these comparisons. We also compared two drying methods for washed clothing using an electric dryer or an outdoor clothesline. A third drying method exposed washed, undried clothing to UV-B to assess if sunlight was the contributing factor for reducing pesticide residues when a reduction was observed for clothes dried on a clothesline. DeSantis et al., (2012) found that alachlor, atrazine, and glyphosate degraded in water when exposed to ultraviolet bands between 200 and 500 nanometers (nm). UV-B radiation is the portion of the electromagnetic spectrum from 290 to 320 nm, and the photons in this band are capable of breaking molecular bonds (Gao, 2020). The decision to measure only UV-B's contribution to photodegradation of residues was at the recommendation of the UV-B Monitoring and Research Program (personal communication; <https://uvb.nrel.colostate.edu/UVB/index.jsf>).

Articles of clothing made from various materials were used in the study to evaluate how material may affect clothing contamination and decontamination. Several material characteristics influence whether chemicals will be absorbed into clothing, including, but not limited to, fiber type, weave, and morphology (Licina et al., 2019). We examined four commonly used clothing materials in this study.

This study included pesticides from three chemical classes with different formulations. Water solubility of formulation and volume of water used during the washing cycles also affect removal of chemicals. Easley et al., (1983) found that the insoluble ester formulation of 2,4-D was not as effectively removed from contaminated clothing as the more water-soluble amine formulation and that 1% to 2% of the initial contamination by 2,4-D ester transferred to other clothing.

We also investigated several other dynamics of pesticide removal in clothing using household washing machines. An uncontaminated baby Onesies® was included when washing the contaminated clothing to assess possible transference of pesticide in the washing machines. Recognizing that not all pesticides may be removed at a similar efficiency, this study used three different classes of pesticides applied at two different rates. As the aforementioned research indicated that higher washing temperatures [60°C (140°F)] were more effective at removing residues, temperatures in the washers were also monitored. To determine whether residues remain in the machines after washing or drying, swabs of the washing and drying machines were analyzed for potential residues. A discussion is included on the impact to the environment of pesticide residues released through the machine rinsates.

## **2. Materials and Methods**

### **2.1 Pesticides and Pesticide Application**

This study included three pesticides from different chemical classes: carbaryl (insecticide), permethrin (insecticide), and 2,4-D ester (herbicide), each applied at a 1X rate (highest label rate) and a 9X rate (nine times the highest label rate). The 9X rate was applied by spraying the clothing with the 1X solution nine times. Application for the 9X rate in this manner was designed to simulate one or more of the following scenarios: an applicator making multiple applications per day, an applicator wearing the same clothing for applications multiple days without washing, or a “spill” of the spray solution on an applicator’s clothing. The pesticide formulations chosen include a variety of pesticide chemistries labeled for a diversity of target sites.

### **2.2 Pesticide Materials**

The carbaryl formulation chosen was Sevin® XLR Plus (Bayer CropScience, EPA Reg. No. 264-333), which contains 44.1% (by weight) of carbaryl (1-naphthyl N-methylcarbamate) or 4 pounds carbaryl per gallon. This formulation is labeled for control of numerous arthropods and multiple use sites.

The permethrin formulation chosen was Perm-UP® 3.2 EC Insecticide (United Phosphorous, Inc., EPA Reg. No. 70506-9), which contains 36.8% permethrin, or 3.2 pounds of permethrin per gallon as an emulsifiable concentrate (EC). It also contains petroleum distillates and is a restricted-use pesticide (RUP) due to toxicity to fish and aquatic organisms. This formulation is labeled to control a variety of arthropods in multiple use sites.

The 2,4-D product used was an ester formulation, Shredder™ 2,4-D LV4 (Winfield™, EPA Reg. No. 1381-102), which contains 2-ethylhexyl ester of 2,4-dichlorophenoxyacetic acid (66.2%) or 3.8 pounds of 2,4-dichlorophenoxyacetic acid per gallon. It also contains petroleum distillates. An ester formulation was chosen as esters have low solubility and, theoretically, would be more difficult to remove from contaminated clothing (Monaco et al., 2002). This formulation is labeled for controlling many annual, biennial, and perennial weeds and brush in multiple use sites.

Water used for the mixing spray solutions was obtained from a spigot located at the mixing/loading pad and analyzed by American Agricultural Laboratory, Inc. (McCook, NE). The water analysis indicated that the pH was 7.0 with total hardness (Ca+Mg) as CaCO<sub>3</sub> of 1.99 grains/gallon, classified as “soft” water. Based on this analysis, it was determined that no adjuvants, including water conditioners, would be added to any spray mix.

### 2.3 Method of Application

Sevin® brand XLR Plus was applied at a 1X rate (two quarts per acre) by mixing 14.2 ounces formulated product in two gallons of water. Perm-UP® 3.2 EC Insecticide was applied at a 1X rate (eight ounces per acre) by mixing 1.78 ounces formulated product in two gallons of water. Shredder™ 2,4-D LV4 was applied at a 1X rate (three pints per acre) by mixing eight ounces formulated product in 1.5 gallons of water. The same rate of each formulation was used for the 9X applications. In order to achieve the 9X rate, the clothing was sprayed with the 1X rate consecutively nine times without a drying period between applications.

The pesticides were applied to the clothing using a Solo® backpack sprayer (Model 417-18L) to simulate “real-world” application by a pesticide applicator or handler. This backpack sprayer used a battery-operated pump to provide consistent spray pressure. This model was equipped with an 18L (equivalent to 4.5 gallons) translucent tank and 12V lead gel battery, utilizing a two-stage electric pump. The two-stage pump and control switch are capable of providing low- or high-pressure applications through modulation of the pump speed. The pressure delivered during the trial applications was 22 psi. The original sprayer wand was cut from this unit, and the spray hose was modified to include a female liquid/air quick disconnect coupler to accommodate a wet boom. The spray wand was replaced with a 7.5-foot, six-nozzle, wet boom with 18-inch nozzle spacing (R&D Sprayers). The boom was modified to include a liquid pressure gauge located just before the head of the “T” and a male end quick disconnect coupler on the hose end. The selected nozzle tips were TeeJet® 80015VS with 50-mesh screens. An Innoquest SpotOn® Model SC-1 sprayer calibrator was used to determine spray output of each nozzle. Sprayer output was determined to be nine gallons per acre at 22 psi and three mph.

Each replicate consisted of four sets of the following clothing exposed to the pesticide: Red Kap® men’s 100% cotton long-sleeved work shirt (size large), Red Kap® men’s 100% cotton denim work jeans (size 32x34), Gildan DryBlend® 50/50 (5.6 oz.) long-sleeved T-shirt (size large), and Gildan® heavy cotton (5.3 oz.) long-sleeved T-shirt (size large). A Gerber® 100% cotton long-sleeved Onesies® (size 18-months) was not



**Figure 1.** Magnification of clothing types used in study.

<b>Table 1. Details concerning clothing types</b>			
<b>Clothing Type</b>	<b>4-Inch X 4-inch Test Sample Wt (g)</b>	<b>Fabric Wt (oz/ yd)</b>	<b>Threads Per Inch (TPI)</b>
<b>100% Cotton jeans</b>	4.758	13.5	99
<b>100% Cotton long-sleeved work shirt</b>	2.591	7.4	176
<b>100% Cotton long-sleeved T-shirt</b>	2.054	5.8	40
<b>50/50 Cotton/Poly long-sleeved T-shirt</b>	2.203	6.3	40
<b>100% Cotton Onesies®</b>	1.694	4.8	40

sprayed but included in the wash to investigate potential residue transference to non-contaminated clothing. The cotton jeans and the work shirt were cross-woven fabrics consisting of two separate threads, whereas the fabric for the T-shirts and the Onesies® was woven from a single continuous thread (knitted) (Figure 1). Details concerning clothing type weight, fabric weight, and threads per inch can be found in Table 1.





**Figure 2.** Arrangement of two replicates of clothing for pesticide application.

Each article of clothing in the replicate and a 4-inch x 4-inch fabric swatch from each clothing type was placed in the spray area. The clothing was positioned on turf, front side down so as to avoid buttons and zippers when cutting sample swatches (Figure 2). Each pesticide was applied to the back side of the clothing using the backpack sprayer while walking between the rows of clothing on the ground.

Fabric swatches were placed among the clothing to assess the concentration of active ingredient deposited onto the clothing prior to washing. Clothing and swatches were allowed to dry prior to placing in plastic bags for transport to the laundry area. While there were four replicates of each pesticide and rate, only two replicates were sprayed on each date due to time limitations of laundering. In most cases, one replicate of the exposed clothing was washed the same date as sprayed while the second replicate from the spray date was held overnight before laundering. The spray date with corresponding weather conditions and wash date are listed in Table A1 of the Appendix.

## 2.4 Decontamination of Clothing

After clothes were sprayed, they were decontaminated using either an agitator or HE washing machine. Clothing was then dried in an electric dryer or hung on a clothesline. A swatch sample from the washed, undried clothing was also exposed to UV-B to determine if any observed pesticide residue reductions in clothing dried on a clothesline were due to sunlight exposure or if other factors should be considered. The materials

and methods used are described below and presented graphically in the Appendix (Figure A1).

## 2.5 Washing/Drying Materials

Water used for washing was analyzed by American Analytical Laboratory, Inc. (McCook, NE) after plumbing was installed and prior to using the machines. Water pH was 7.1 with a total hardness (Ca+Mg as CaCO<sub>3</sub>) of 1.84 grains/gallon, categorized as “soft.” A Navien NPE condensing tankless natural gas water heater (Model NPE-240A) was installed and plumbed three feet from the HE washer and five-feet from the agitator washer. The temperature setting was 49°C (120°F), as the manufacturer recommends NOT exceeding this temperature.

A Sotera Systems (Model P0550) meter was used to measure flow rate (3-26 gallons per minute capacity) into the washers – 1” NPT inlet/outlet. A “Save A Drop” (P3 P0550) water meter measured water removal from the washer (0.1-gallon accuracy) ¾” inlet/outlet. The “Save A Drop” water meter proved not accurate enough to measure water rinsate leaving the HE machines as expelled volumes were too low.

The HE machine (GE Model GFWS1700H1WW) used for the study was a 4.3 cu. ft. capacity stainless steel drum front load washer that included load-sensing adaptive fill (measures weight of load and depth in basket), a 1300 rpm spin speed, and an internal water heater (49°C - 66°C (120°-150°F)). Front-load HE washers work by filling the bottom of the inner tub with a small volume of water and using the rotation of the tub and gravity to move the clothes through the water. They spin and rotate the load in both clockwise and counterclockwise motions at high and low speeds so that garments rub against each other for cleaning. In many front-loading HE washers, the centrifugal spin of the tub lifts clothes above the water level and then drops them back into the water. The side paddles on the inside drum aid in lifting the clothes, moving them in and out of the water. This provides the mechanical action (scrubbing) needed to remove soil from fabric ([www.thespruce.com/how-does-front-load-washer-work-2145864](http://www.thespruce.com/how-does-front-load-washer-work-2145864)).

The full-fill agitator machine (GE Model GTWN280DWW) used in the study was a 3.8 cu. ft. capacity stainless steel drum washer with a 630 rpm spin speed and dual action agitator. The function of the agitator is to move the clothing in a way that pushes water through the fabric, removing the dirt. As the dual-action agitator is moved forward, the fins on it lightly grab the clothing forward and outward. In addition, there are secondary fins at the top of the agitator that force the floating (or top clothing) down to the bottom fins. The top to bottom agitation continues for the length of the wash cycle, forcing water and detergent through the fabrics to loosen soil (<https://www.thespruce.com/how-top-load-washing-machine-works-2145865>).

Clothes were dried using a seven cu.ft. electric dryer (Maytag Model MEDC215EW) on high temperature setting for 40 minutes. iButton® Thermochrons F5 (Digi-Key Electronics) were placed into IC Button Capsules to measure temperatures in the washers and dryer.

Persil ProClean™ 2 in 1 was chosen as the laundry detergent for the study as *Consumer Reports* (2016) rated it as the top laundry detergent that can be used in both

HE and agitator machines. Persil ProClean™ 2 in 1 was rated the top brand due to its excellent rating for cold-water cleanability and a very good rating at removing grass, blood, and dust sebum (which simulates body oil and “ring around the collar”).

## 2.6 Decontamination Methods

Decontamination of clothing either occurred the same day as spraying or the following day. Clothes were left in the plastic bags at room temperature if processing was scheduled for the following day after spraying. There were a few exceptions listed in the Appendix (Table A1) where clothing could not be processed (as described) the same day or day after spraying. In those few cases, sprayed clothing was placed in an Amana Deepfreeze® (Model AFC2207BW) at -18°C (0°F). For all decontamination/sampling procedures described below, residue and water samples were refrigerated at 2°C (36°F) until extracted (extraction method is described below).

Two complete sets of pesticide-contaminated clothing from an individual replicate were placed in either the HE machine or the agitator machine. Two sets were used to ensure a sufficient amount of clothing in the washers and adequate statistical power. Two Gerber Onesies® 100% cotton long sleeve bodysuits (size 18-months) were added to each washer to determine if transference of pesticide residues from pesticide contaminated clothing occurs in the washer. An iButton® ThermoChron in a capsule was then placed in each washer prior to starting the wash cycle to measure temperatures during the wash cycle.



**Figure 3.** Modified sediment filter for waste water sampling.



We followed the most current guidelines for laundering pesticide-contaminated work clothes (Thostenson et al., 2016). The agitator washer was set for a "whites, heavy soil, super load size, hot water" wash load. The HE washer was set for a "whites, pre-wash, heavy soil, hot water, high spin, extra rinse". A 100 milliliter (mL) graduated cylinder was used to measure the recommended amount, 95 mL, of laundry detergent for each wash load. The HE machine received an additional 30 mL for the prewash cycle. Wastewater was collected from each machine using a modified sediment filter with screen removed and placed inline from the drain hose for each of the following stages: prerinse (HE only), wash, rinse and extra rinse cycles (Figure 3). During each drain cycle, 500 mL of water was collected after flushing the sediment filter twice while draining. The water samples were refrigerated at 2°C (36°F) until analyzed for pesticide residues.

After the clothing decontamination wash cycle was completed, the washer drum was swabbed using a sterile gauze pad which was placed in a plastic bag and submitted for residue analysis. Continuing to follow the most recent clothing decontamination recommendations (Thostenson et al., 2016), the washer was cleaned by running the complete cycle used for clothing decontamination, including the same volume of laundry detergent (95-mL) with the addition of ½ cup of Clorox® bleach. The swab process was repeated after the washing machine clean cycle (repeated cycle, no clothes).

One set of clothing was removed from each washer type and split longitudinally with one-half of the clothing sampled by cutting a 4-inch x 4-inch square from the back and



**Figure 4.** Measuring UV-B exposure level during line drying.



**Figure 5.** Simulated UV-B exposure indoors.

placed in a plastic bag for analysis to assess the concentration of residues remaining after washing. An additional 4-inch x 4-inch square was removed to expose to UV-B indoors. The remaining half of the clothing was hung on a clothesline outdoors. All clothing samples were refrigerated at 2°C (36°F) until submitted for analysis of pesticide residues.

We implemented an experimental protocol to test if UV-B radiation alone, or other factors such as wind or humidity, play a role in the breakdown of pesticide residues from laundered clothing. We dried clothing at the same level of UV-B exposure under two

conditions: outdoor on the clothesline and indoor in a dark room. At the time clothes were hung on the clothesline, we measured and recorded the UV-B radiation using a Solarmeter® (Model 6.0) Digital UV meter for UV-B (mW/cm<sup>2</sup>). We then set a UV-B light to the same exposure, which was used to dry the other 4-inch x 4-inch clothing sample in a completely dark room for 40 minutes (Figures 4 and 5). All samples from both indoor and outdoor UV-B exposures were placed in a plastic bag and submitted for residue extraction.

The remaining (second) set of clothes was placed in the electric dryer on high for 40 minutes, and then a 4-inch x 4-inch sample was removed from the back of the clothing and placed into a plastic bag for analysis. The dryer was also swabbed in the same manner as the washers after drying was complete, and the gauze swab submitted for analysis.

## 2.7 Pesticide Quantitation Method by Triple Quadrupole UPLC-MS/MS

The exposed fabric swatches were submitted for pesticide residue analysis to the Central Instrumentation Facility Department of Chemistry at Colorado State University. Pesticides from clothing and swab samples were extracted with 150 mL reagent grade methanol in 250 mL Erlenmeyer flasks for 30 min on an orbital shaker. An approximately ten mL subsample of the extractant was transferred to a glass 20 mL scintillation vial for analysis.

Based on some preliminary testing of different extraction methods, 2,4-D laundry water samples were prepared for analysis using a solid phase extraction (SPE) cartridge (Waters Oasis MAX 6cc/500mg). SPE cartridges were prepared by rinsing with methanol and then equilibrating with water. Water samples were first filtered (Whatman 3), basified to pH 12 with 20% ammonium hydroxide, and 100 mL of rinsate was added to the SPE cartridge where a vacuum was applied. The SPE cartridge was washed with 5% ammonium hydroxide and dried. Next, isopropyl alcohol was added to elute 2,4-D.

For carbaryl and permethrin, a QuEChERS method was used for preparing the water samples for analysis. For carbaryl, the European EN 15662 method was used, and for permethrin, the AOAC 2007.01 method was used. Each method had the same initial step with the addition of 20 mL of laundry water and ten mL of acetonitrile added to prepared 50 mL tubes, vortexed for one minute. Next, prepacked salts containing 4 g MgSO<sub>4</sub>, 1 g NaCl, 1 g trisodium citrate dihydrate, 0.5 g disodium hydrogencitrate sesquihydrate (Restek) were added and vortexed for an additional minute. Samples were then centrifuged for six minutes at 4700 rpm, and 1 mL of the supernatant was transferred to dispersive SPE (dSPE) tubes. For carbaryl, the dSPE was 150 mg MgSO<sub>4</sub>, 25 mg PSA, 2.5 mg graphitic black carbon (GCB), and for permethrin, the dSPE was 150 mg MgSO<sub>4</sub>, 50 mg PSA, 50 mg C18. Once transferred to the dSPE tubes, the samples were vortexed and centrifuged for ten minutes at 3000 rpm. For carbaryl, 360 µL was transferred to an LC/MS vial and spiked with <sup>13</sup>C carbaryl internal standard. For permethrin, 100 µL was transferred to a limited volume insert and spiked with the <sup>13</sup>C permethrin internal standard.

Stock solutions (1 mg/mL) of standard carbaryl, 2,4-D, and permethrin were prepared in methanol. Solutions for nine-point calibration curve were prepared in methanol from

stock solutions using isotopically labeled compounds as internal standards. QC samples and blanks were run every eight to ten injections, and all LC/MS injections were introduced in duplicate.

A Waters H-class Acquity UPLC systems in-line with a Waters triple quadrupole mass spectrometer (TQD) equipped with an electrospray ionization (ESI) source was used for separation and detection of carbaryl (positive ion mode) and 2,4-D (negative ion mode). Source conditions on the TQD were as follows: electrospray ionization in positive ion mode, capillary voltage 2.4 kV, cone voltage 40 V, source temperature 150°C (302°F), desolvation temperature 200°C (392°F), desolvation gas flow 550 L/hr, cone gas flow 1 L/hr, extractor 3 V and RF lens 2.5 V. A Waters Acquity BEH UPLC column (50 x 2.1 mm 1.7 µm particle size) was used with gradient separation. Flow rate was 0.4 mL/min.

For carbaryl, mobile phase A was water with 0.1% formic acid; mobile phase B was acetonitrile. The gradient started at 20% B, held for 0.1 minute, ramped to 100% B over 2.5 minutes, held at 100% B for 0.5 minute, returned to 20% B over 0.1 minute, and equilibrated at 50% B for 0.9 minute for total UPLC run time of four minutes.

For 2,4-D, mobile phase A was water with ten mM ammonium acetate; mobile phase B was acetonitrile. The gradient started at 20% B, held for 0.1 minute, ramped to 100% B

<b>Table 2. Transitions used for quantitation as confirmatory qualifiers along with dwell times, cone voltage and collision voltages.</b>						
<b>Pesticide</b>	<b>Platform</b>	<b>Ionization Source</b>	<b>Parent Ion (m/z)</b>	<b>Cone Voltage (V)</b>	<b>Quantifier Ion (Collision Energy)</b>	<b>Qualifier Ion (Collision Energy)</b>
2,4-D	LC/MS-MS	ESI -	219	10	161 (14)	125 (24)
<sup>13</sup> C-2,4-D	LC/MS-MS	ESI -	225	10	167 (14)	131 (24)
carbaryl	LC-MS/MS	ESI +	202	20	145 (8 V)	127 (26 V)
<sup>13</sup> C <sub>6</sub> -carbaryl	LC-MS/MS	ESI +	208	24	151 (12 V)	133 (24 V)
Cis-permethrin	GC-MS/MS	EI	183	-	168 (10V)	165
Trans-permethrin	GC-MS/MS	EI	183	-	168 (10V)	165
<sup>13</sup> C <sub>6</sub> -permethrin	GC-MS/MS	EI	189	-	174 (10V)	-

over 2.5 minutes, held at 100% B for 0.5 minute, returned to 20% B over 0.1 minute, and equilibrated at 50% B for 0.9 minute for total UPLC run time of four minutes.

For permethrin, samples were injected in splitless mode on the gas chromatograph (GC) at an inlet temperature of 250°C (482°F). The GC oven temperature started at 70°C with a one minute hold, then increased to 300°C (572°F) at 10°C (50°F)/minute, with a final hold of ten minutes. The GC column was a Phenomenex ZB-5HT-MS Inferno (30m x 0.25mm x 0.25µm).

Transitions used for quantitation as confirmatory qualifiers along with dwell times, cone voltage and collision voltages are listed in Table 2.

Quantitation data were processed using the response ratio for target analytes to their isotopically labeled internal standards. Injections performed in duplicate were checked for a coefficient of variation <20%.

### 3. Statistical Analysis

Linear mixed regression models were used to analyze mean pesticide concentrations in clothing as a function of different spray rates, clothing types, washing, and drying methods. We used these models to estimate the effects of these factors and to test for differences in mean pesticide concentrations. Unless otherwise noted, carbaryl concentrations were log transformed while permethrin concentrations were square root transformed for these analyses. These transformations were used to satisfy modeling assumptions. Each model included a random effect for the day of spray application. For analyses where the same article of clothing was physically split, we also included a random effect for article of clothing. Pairwise comparisons used Tukey adjustments. Due to difficulties encountered in the laboratory analysis of 2,4-D samples arising from the technical material used, only limited statistical analysis results were reported for this study. While limited observations from this portion of the study were included here, an improved experiment with the 2,4-D ester applications and analysis was currently being conducted.

## 4. Results and Discussion

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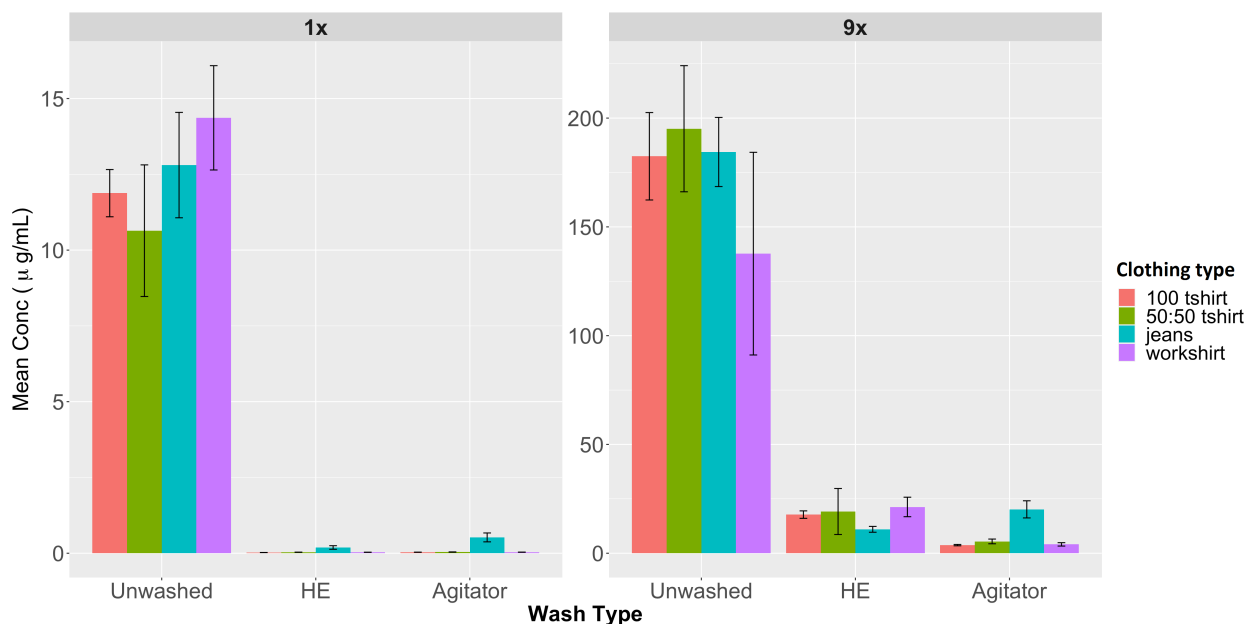
### 4.1 Effects of Spray Rates, Clothing Type, and Washing Method on Pesticide Residues

#### 4.1.1 Carbaryl Results

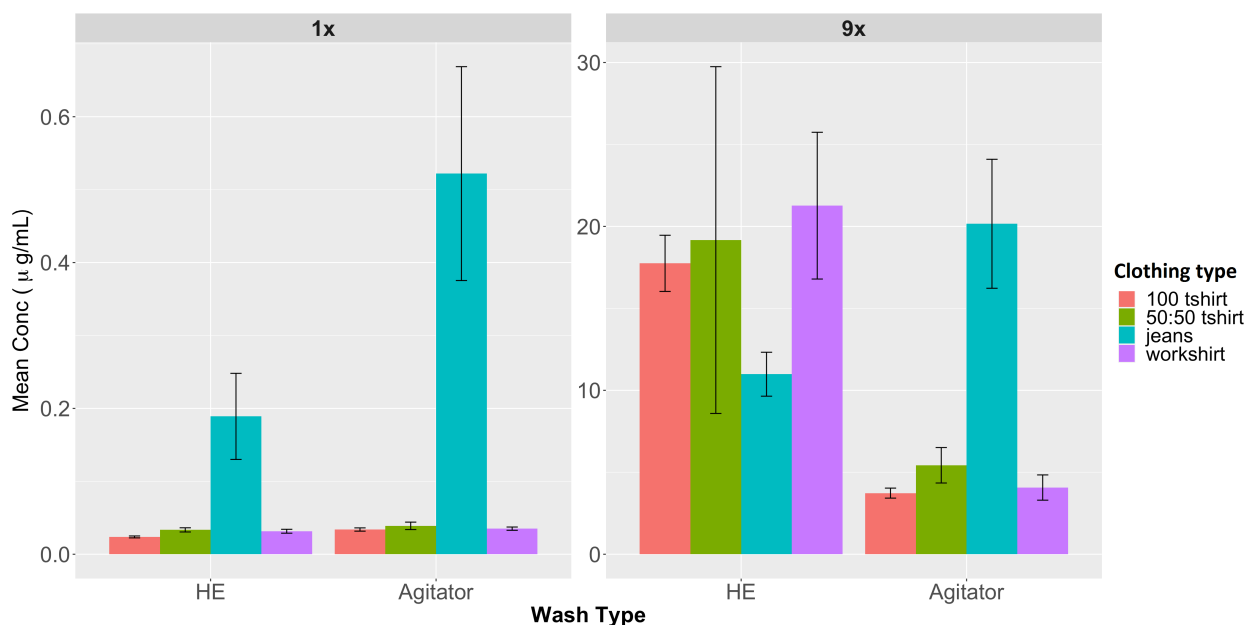
The sample means and standard errors for each combination of spray rate, wash type, and clothing type are shown in Figure 6. We found evidence of a three-way interaction among these three factors ( $p$ -value = 0.016). We provide only the most pertinent comparisons for our research hypothesis rather than an exhaustive discussion of all interactions and pairwise comparisons.

The carbaryl concentrations in unwashed clothing demonstrated that the two application rates (1X and 9X) were effective in inducing different levels of contamination. The mean concentrations in unwashed articles of clothing were 12.4 µg/mL (sd = 3.3,  $n$  = 16) and





**Figure 6.** Mean carbaryl concentrations by spray rate, wash type, and clothing type. Note the differing y-axis for the two figures. The error bars display +/- 1 standard error (sd/sqrt(n)).



**Figure 7:** Comparison of mean carbaryl concentrations for clothing washed in HE and agitator washing machines. Note the change in scale on the y-axis. The error bars display +/- 1 standard error (sd/sqrt(n)).

174.9  $\mu\text{g/mL}$  (sd = 59.7, n = 16) for the 1X and 9X rates, respectively. We did not find differences in contamination among the clothing types. Among unwashed clothing, there was not enough evidence to conclude that there were differences in the mean carbaryl concentrations between clothing types for each of the spray rates ( $p > 0.65$ ).

Across clothing types, there was evidence that both washing methods were effective at reducing carbaryl concentrations for both spray rates ( $p < 0.001$ ). At the 1X application rate, the mean concentrations were 0.16  $\mu\text{g/mL}$  (sd = 25, n = 16) for the agitator and 0.07 (sd = 0.09, n = 16) for the HE machine. These means were just 1.3% and 0.6% of the average concentrations found in the unwashed clothing at the same application rate. Similarly, at the 9X rate, the average concentrations were 8.35 (sd = 8, n = 16) and 17.29 (sd = 11.19, n = 16) for the agitator and HE washer, respectively. These means were 4.8% and 9.9% of the average seen for unwashed clothing at the 9X rate.

The differences between the HE and agitator machines depended on the spray rate and clothing types and indicate that neither machine was superior for reducing pesticide concentrations (see Figure 7). We first compared residue concentrations from the washing machine types averaged across clothing types at each spray rate. The mean carbaryl concentrations across all articles of clothing sprayed at the 1X rate were greater for the agitator than for the HE machine ( $p\text{-value} = 0.007$ ). However, at the 9X rate, articles of clothing washed in the agitator had a lower mean concentration than those in the HE machine, with jeans being an exception ( $p < 0.001$ ). This may have been due to the larger volume of water used during the wash and rinse cycles of the agitator machine (see related discussion in Section 4.5).

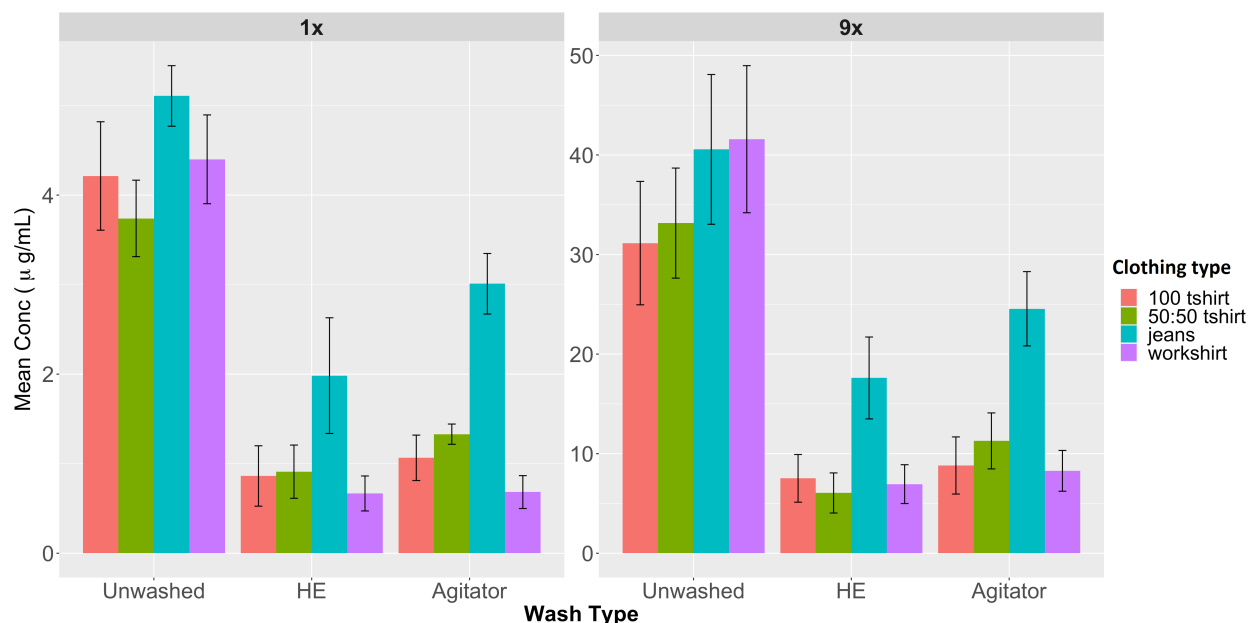
Comparisons of carbaryl concentrations among clothing types indicated varying concentrations with few clear patterns. However, for each given spray rate and washer type, the jeans had the highest mean concentrations except for the HE machine at the 9X rate. Thus, there is some evidence to suggest that carbaryl is more difficult to remove from 100% cotton jeans than the other clothing types. Kim et al. (1982) found that the thickness and weight of the heavier fabrics may allow deeper penetration of the pesticide into the fibers as well as into the fabric structure by a wicking process that made the chemical more difficult to remove.

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#### 4.1.2 Permethrin Results

The sample means and standard errors for permethrin concentrations for each combination of spray rate, wash type, and clothing type are shown in Figure 8. For the permethrin analysis, we did not find evidence of a three-way interaction among these three factors ( $p = 0.74$ ). However, there was evidence of two-way interactions between wash type and spray rate ( $p < 0.001$ ) and clothing type and spray rate ( $p = 0.05$ ). There was also evidence of differences in means for all main effects ( $p < 0.03$ ). Here, we provided only the most pertinent comparisons for our research hypothesis.

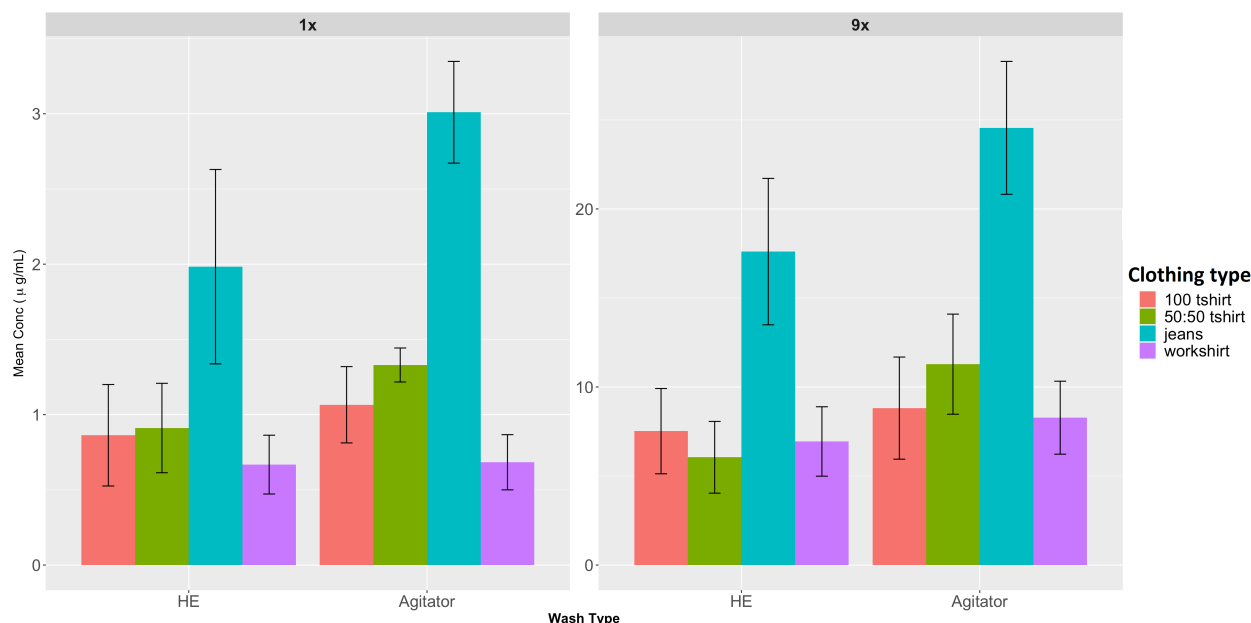
Like the carbaryl experiment, the permethrin concentrations in unwashed clothing demonstrated that the two application rates were effective in inducing different levels of contamination. The mean concentrations in unwashed articles of clothing were 4.4  $\mu\text{g/mL}$  (sd = 1.0, n = 16) and 36.6  $\mu\text{g/mL}$  (sd = 12.9, n = 16) for the 1X and 9X rates,



**Figure 8:** Mean permethrin concentrations by spray rate, wash type, and clothing type. Note the change in scale for the y-axis for the two figures. The error bars display +/- 1 standard error (sd/sqrt(n)).

respectively. Among unwashed clothing, there was not enough evidence to conclude differences in the mean permethrin concentrations between clothing types for each of the spray rates ( $p > 0.43$ ).

We found evidence that both washing machines were effective at reducing permethrin residue concentrations. Across clothing types, there was evidence that washing was effective at reducing permethrin concentrations for both spray rates ( $p < 0.001$ ). At the 1X application rate, the mean concentrations were 1.52 µg/mL (sd = 1.01,  $n = 16$ ) for the agitator washer and 1.11 µg/mL (sd = 0.90,  $n = 16$ ) for the HE washer. These means were 35% and 25%, respectively, of the mean concentrations found in the unwashed clothing at the same application rate. Similarly, at the 9X rate, the mean concentrations were 13.23 (sd = 8.6,  $n = 16$ ) and 9.53 (sd = 6.92,  $n = 16$ ) for the agitator and HE washer, respectively. These means were approximately 36% and 26%, respectively, of the means found for unwashed clothing at the 9X rate. We noted that these percentages are much greater than those observed for the carbaryl analysis, suggesting permethrin is more difficult to remove from clothing than carbaryl. This could be partially due to the emulsifiable concentrate (EC) formulation chosen for the study. Laughlin et al. (1985) reported that an EC of methyl parathion was more difficult to remove than encapsulated and wettable powder (WP) formulations. In addition, permethrin is used to treat clothing to repel insects. The manufacturer of Insect Shield® apparel claims the permethrin in the apparel will remain in the clothing for up to 70 washes (Insect Shield, 2019).



**Figure 9:** Comparison of mean permethrin concentrations for clothing washed in HE and agitator washing machines. Note the change in scale on the y-axis. The error bars display +/- 1 standard error (sd/sqrt(n)).

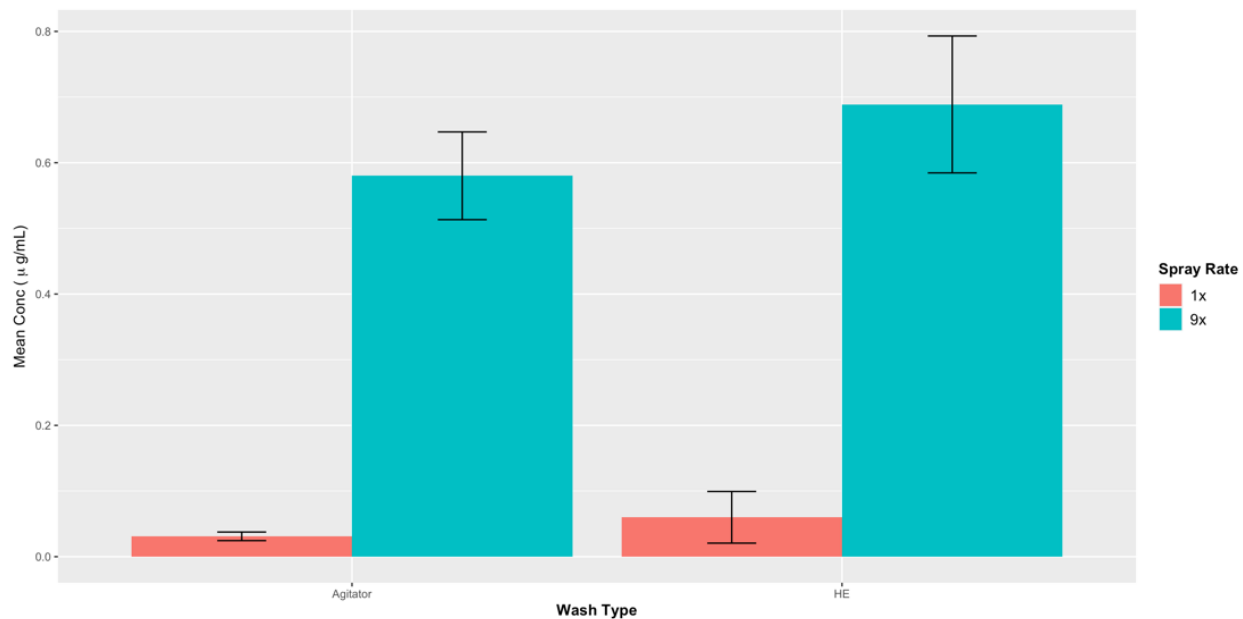
The differences in mean permethrin residues between the two washing machine types depended on the spray rate and clothing type. Figure 9 displays the mean permethrin concentrations for both washing machines at each clothing type and spray rate combination. Averaged across clothing types, the HE washer had smaller permethrin concentrations than the agitator. This difference was small (0.4 µg/mL) for clothing sprayed at the 1X rate ( $p = 0.60$ ) but relatively large (3.7 µg/mL) at the 9X rate ( $p = 0.054$ ). Like the carbaryl results, we observed that the jeans tended to hold a higher concentration of permethrin after washing relative to other types of clothing. Thus, there is some evidence to suggest that permethrin is more difficult to remove from 100% cotton jeans relative to the other clothing types.

## 4.2 Transference

### 4.2.1 Carbaryl

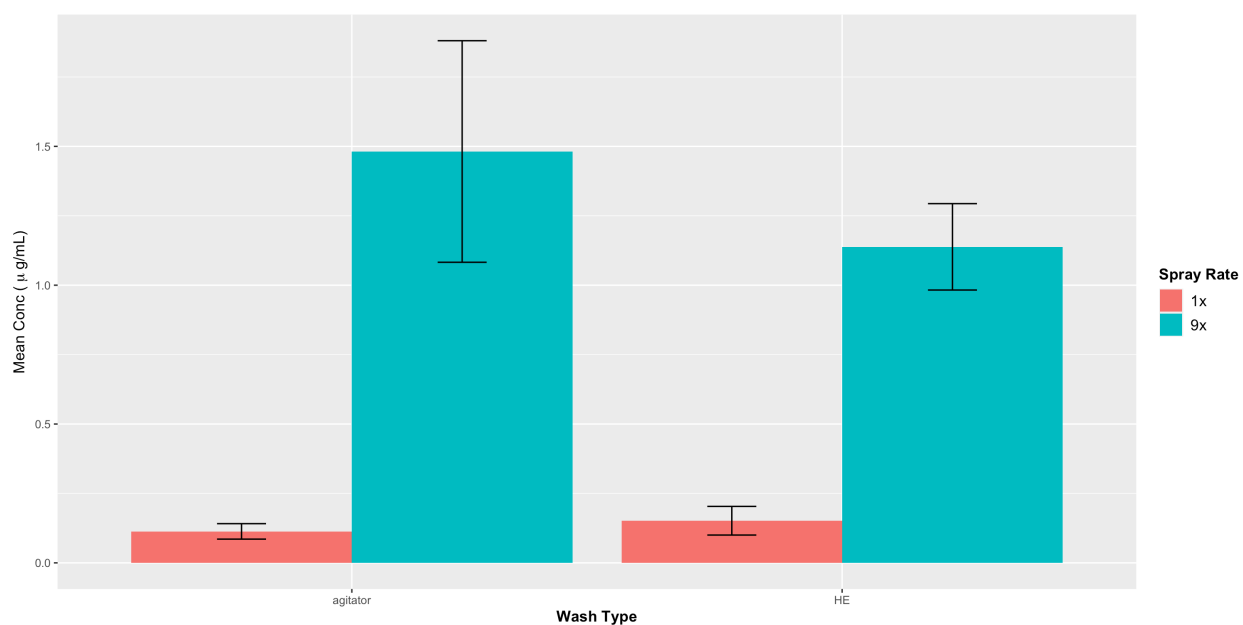
There was transference of pesticide to the Onesies® added to the washing machine at both spray rates and for both washer types. While both washer types transferred carbaryl to the Onesies® during the wash cycle, there was not enough evidence to conclude that the contamination differed by washer type ( $p = 0.59$ ). However, the contamination did differ by spray rate ( $p < 0.001$ ). Onesies® washed with 9X sprayed clothing had much greater contamination than 1X sprayed clothing. Figure 10 displays the mean carbaryl concentrations for the Onesies® for both spray rates and wash types.





**Figure 10:** Transference of carbaryl to Onesies®. The error bars display +/- 1 standard error (sd/sqrt(n)).

Very low levels of carbaryl were transferred to the Onesies® in both washing machines at the 1X rate (0.03 µg/mL for the agitator, 0.06 µg/mL for the HE). The levels of transference detected at the 9X rate (0.58 µg/mL for the agitator, 0.69 µg/mL for the HE) were much larger; mean concentrations were more than ten times greater than those at the 1X rate.



**Figure 11:** Transference of permethrin to Onesies®. The error bars display +/- 1 standard error (sd/sqrt(n)).

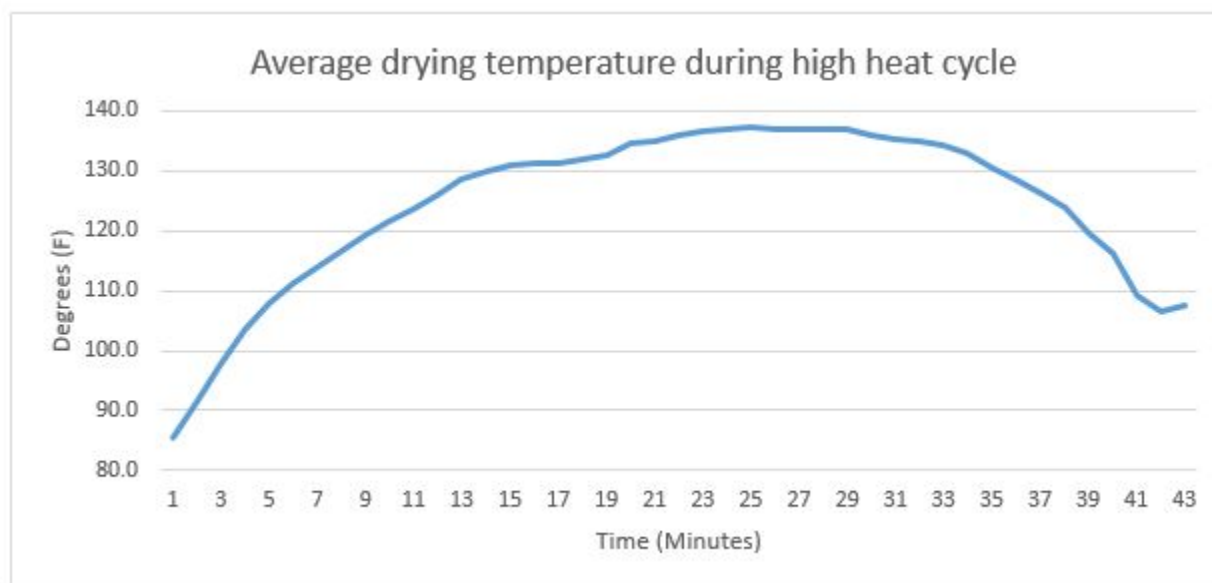
### 4.2.2 Permethrin

Like the carbaryl results, there is evidence of permethrin transference to Onesies® added to the washing machine with contaminated clothing. Permethrin transferred to the Onesies® in both washing machines at the 9X rate (1.48 µg/mL for the agitator, 1.14 µg/mL for the HE) (Figure 11). The levels of transference recorded at the 9X rate are more than 18-times greater than recorded at the 1X rate for the agitator and seven-times greater for the HE machines. There was not enough evidence to conclude that the contamination differed by washer type ( $p = 0.61$ ). However, the mean contamination for Onesies® washed with 9X sprayed clothing is substantially greater than 1X sprayed clothing ( $p < 0.001$ ).

## 4.3 Effect of Drying Method on Pesticide Residues

### 4.3.1 Drying Temperatures

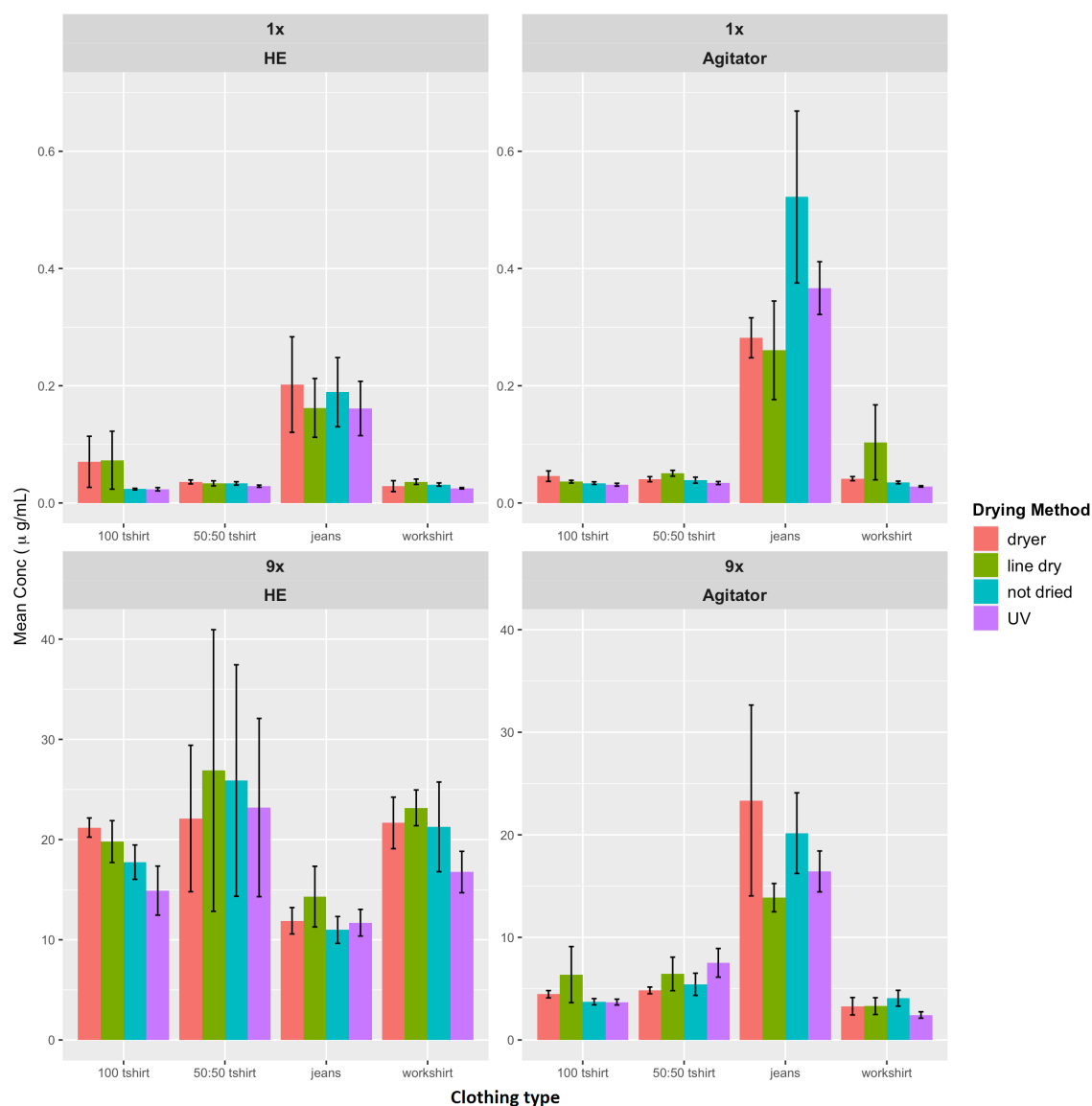
Most research to date has involved laboratory air-drying of laundered specimens. However, Kim et al. (1986) reported significantly lower residue of alachlor in laundered specimens that were dryer-dried, either due to elevated temperature (97°C or 207°F) or airflow. In this study, we compared residues in clothing dried in an electric dryer or dried



**Figure 12.** Average drying temperature as recorded during all drying cycles.

on a clothesline. The average peak drying temperature in the dryer used in this study was 59°C (138°F) approximately 25 minutes into the 40-minute cycle (Figure 12).

Some pesticides are volatilized by heat or degraded by ultraviolet light. Branson and Rajadhyaksha (1988) hypothesized that exposure to simulated sunlight, heat, and humidity might be an effective means of decontaminating fabrics since pesticides break down in the natural environment. A literature review yielded no published information



**Figure 13.** Comparison mean carbaryl concentrations for three drying methods. The error bars display  $\pm 1$  standard error (sd/sqrt(n)).

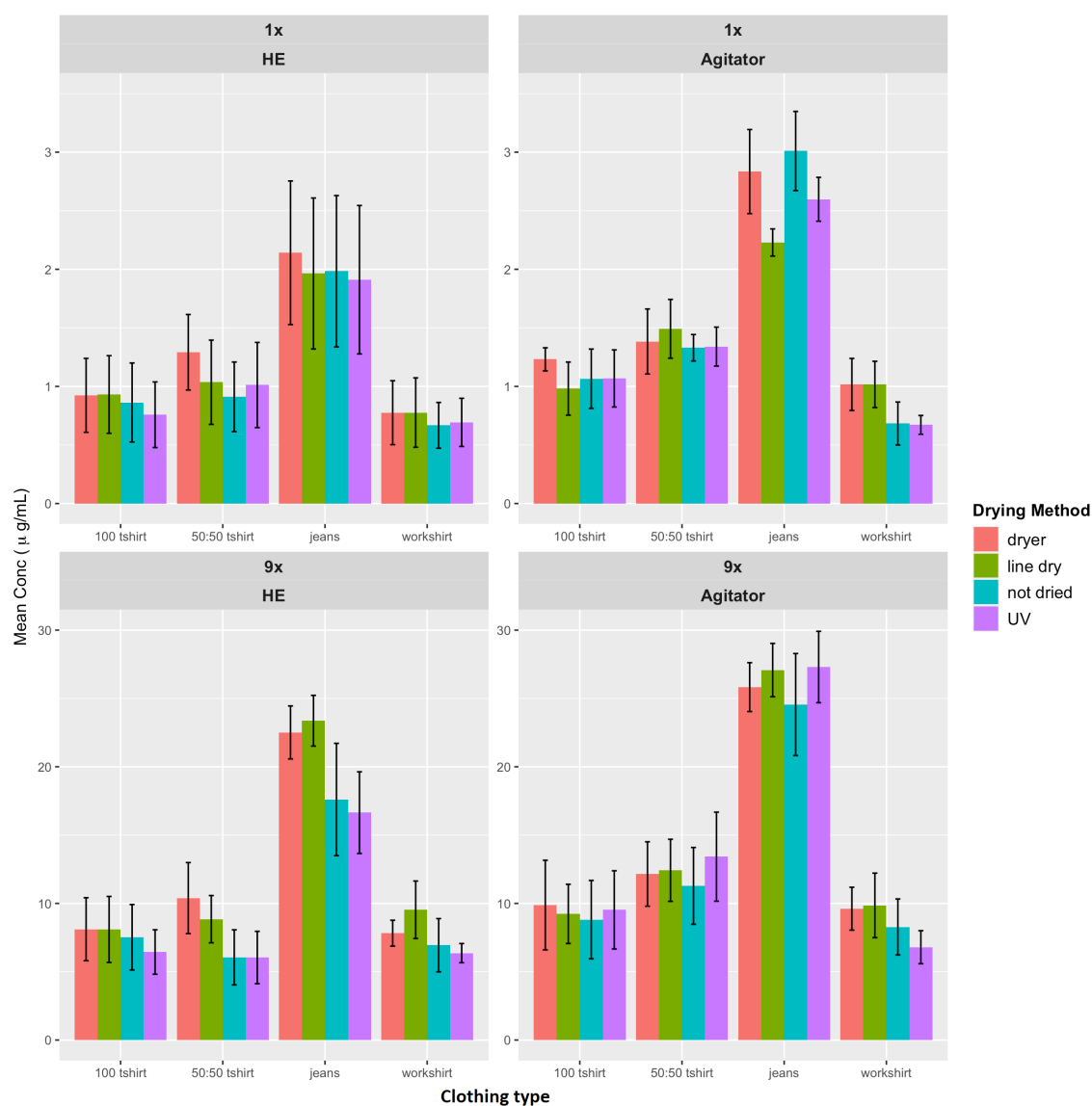
concerning UV or sunlight degradation of pesticide residues in clothing; therefore, we also examined whether UV-B might be a factor in reducing residues in the clothing. The following results compare mean concentrations of pesticide in clothing washed but not dried, to clothing washed and dried in an electric dryer, line-dried, or exposed to UV-B.

#### 4.3.2 Carbaryl

There were no meaningful differences in mean carbaryl concentrations as a function of drying method. Here, we report the main effect of drying method in each of three models used to assess differences in drying methods. Across the spray rates, washing types, and clothing types (Figure 13), there was little evidence to suggest that the mean carbaryl concentration for clothes that were dried in the electric dryer was different than

for clothes that were washed but not dried ( $p = 0.84$ ). Similarly, the difference in the observed mean concentrations when comparing clothesline drying to the electric dryer was near zero ( $p = 0.98$ ). Finally, when comparing the three post washing methods (non-dried, UV-B exposure, and clothesline-dried), there was not enough evidence to conclude that the mean carbaryl concentrations among the three methods were different from one another ( $p = 0.11$ ). Based on these data, UV-B does not appear to further degrade carbaryl residues at either rate. Any of the drying methods investigated in this study can be used after washing carbaryl-contaminated clothing.

### 4.3.3 Permethrin



**Figure 14:** Mean permethrin concentrations for three drying methods. The error bars display  $\pm 1$  standard error (sd/sqrt(n)).



The drying method had an effect on mean pesticide concentrations for the permethrin experiment (Figure 14). We report the main effect of drying method (that is, the effect averaged across spray rate, wash type, and clothing type) in each of three models used to assess differences in drying methods. For the first two models, we used the square root of permethrin concentration as the response variable to satisfy modeling assumptions while we used the log of the concentration for the same reason in the third model.

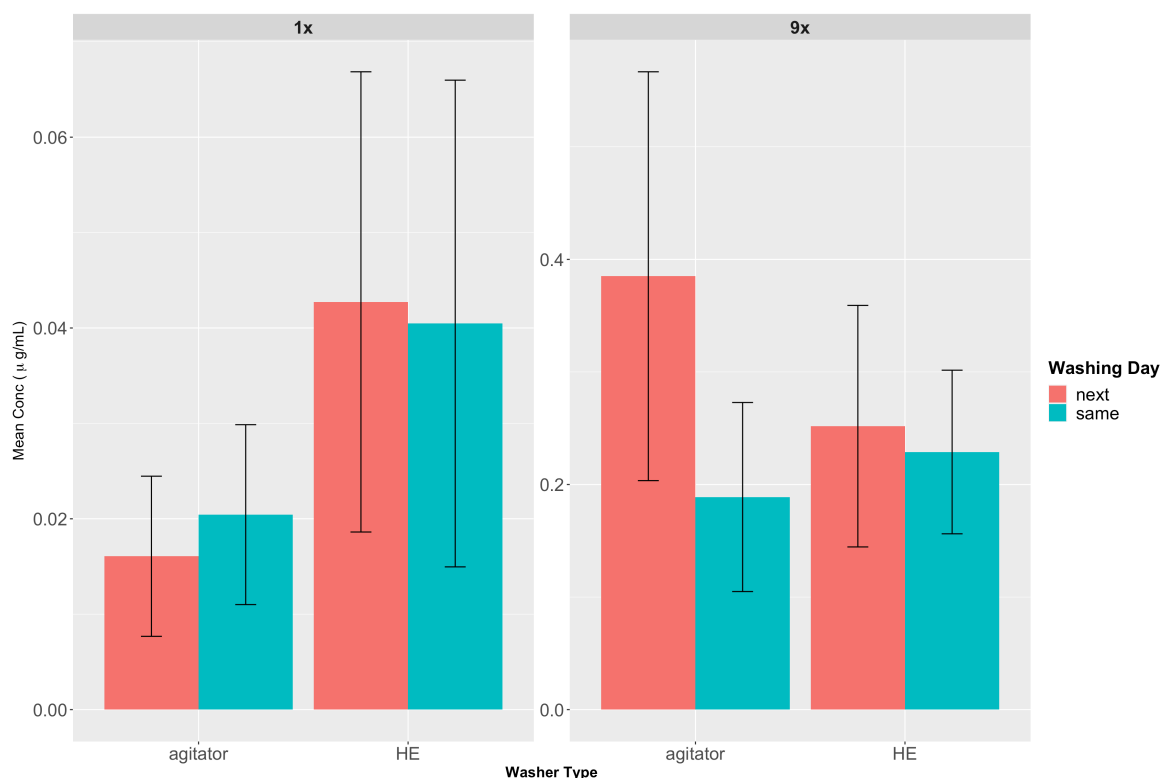
When comparing clothing that was washed and not dried to those dried in an electric dryer, we found similar mean permethrin concentrations (6.3 µg/mL and 7.4 µg/mL, respectively, and there was not enough evidence to conclude these concentrations were different ( $p=0.11$ ). The mean permethrin concentrations were the same for both clothesline dried clothes (mean 7.4 µg/mL) and clothes dried in the electric dryer (given above). Thus, there was no evidence of a difference between these methods.

A comparison of the three post washing methods (non-dried, UV-B exposure, and clothesline-dried) found that mean concentrations for clothesline-dried are greater than for UV-B exposed clothes ( $p=0.045$ ) and non-dried clothes ( $p=0.027$ ). The observed mean permethrin concentrations for these three categories were 7.4 µg/mL for clothesline-dried clothes, 6.4 µg/mL for UV-exposed clothes, and 6.3 µg/mL for non-dried clothes. This suggests that UV-B exposure may be a factor in reducing permethrin residues in clothing.

#### **4.4 Timing of Washing**

Although the quality of 2,4-D data was limited due to challenges with the laboratory analysis, the data were analyzed to investigate a possible effect of delayed washing on pesticide concentrations. On each application date, two replicates were treated. One replicate was laundered the same day as it was sprayed, while the second replicate was stored overnight in a plastic bag for processing the second day. Kim et al.(1982) reported that immediate washing resulted in lower residues of alachlor and fonofos than found in clothing washed after a 24-hour delay.

There was some evidence to suggest that clothing washed on the same day on which it was contaminated has lower mean pesticide concentrations than the clothing washed a day after contamination. Figure 15 displays the mean 2,4-D concentrations for washed clothing as a function of washer type, wash timing, and application rate. For both washers at the 9X rate, a delay in washing was associated with a greater mean concentration. At the 1X rate, the mean concentration was higher for delay-washed clothing compared to same-day washed clothing in the HE machine, but the mean concentration was lower for delay-washed clothing in the agitator machine. An ANOVA analysis did not find evidence of an effect associated with washing timing ( $p=0.69$ ). As previously mentioned, confidence in these data is low, and further experimentation is needed to assess absorption of 2,4-D ester to different clothing types.



**Figure 15.** Mean 2,4-D concentrations as a function of washer type, wash timing, and spray rate. The error bars display  $\pm 1$  standard error ( $sd/\sqrt{n}$ ).

#### 4.5 Washing Temperatures and Water Volumes

Based on research conducted by Easley et al.(1982), one of the most important factors in successfully decontaminating pesticide-exposed clothing is the water temperature used during the wash and rinse cycles. This research investigated laundry detergent and water temperature as factors in pesticide decontamination of work-weight fabrics by laundering in three water temperatures. There was lower methyl parathion removal at 30°C (86°F) than at 49°C (120°F) and 60°C (140°F). Easter (1983) investigated removal of the pesticide active ingredients azinphosmethyl and captan from 100 % cotton denim, chambray, GORE-TEX®, and Tyvek®. An increase in wash water temperature resulted in increased pesticide removal; however, the increase in temperature was more beneficial

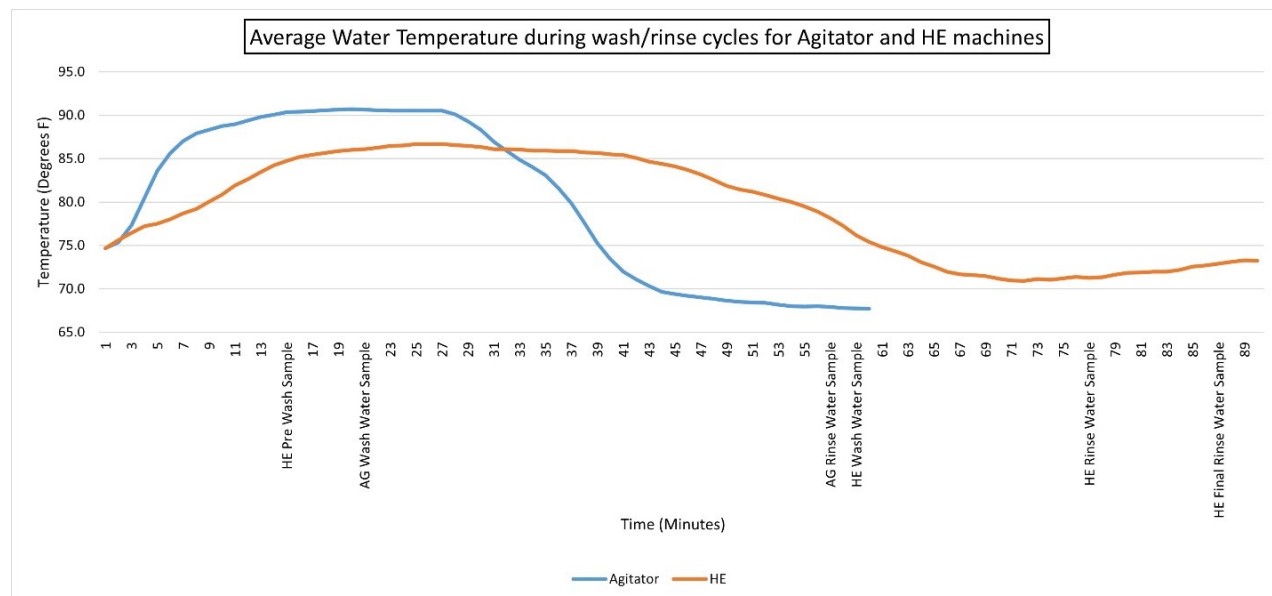
**Table 3. Total gallons of water used during the wash cycle for the agitator and high efficiency washer.**

Machine Type	Cold Water	Hot Water	Total Gallons Used
Agitator	25.1	9.0	34.1
High Efficiency (HE)	11.9	2.3	14.2

for denim than for the others. Easter and Dejonge (1985) recommended wash temperatures of at least 49°C (120°F) for Tyvek® and GORE-TEX® and 60°C (140°F) for all-cotton fabrics. Lillie et al. (1982) studied removal of diazinon, chlordane, chlorpyrifos, bromacil and azinphosmethyl at 30°C (86°F), 43°C (109°F), and 60°C (140°F). They noted a trend for increased pesticide residue removal with increased temperature when detergent or detergent and bleach were used. When laundry products were not used, pesticide removal did not increase as water temperature increased. Laughlin and Gold (1991) concluded that the hotter the water, 30°C (86°F) vs. 60°C (140°F), the lower the residues.

In March 2015, the U.S. Department of Energy revised their standards and federal guidelines for hot water energy consumption for all clothes washers to save energy and conserve resources. Due to these standards, it is nearly impossible to achieve temperatures greater than 32.2°C (90°F). To comply with these standards, the “hot” water temperature selection on washing machines is a mixture of hot and cold water from the household plumbing (GE Appliances). Table 3 shows the average volume of cold and hot water used in this study during the wash/rinse cycles for both the agitator and HE machines.

Typically, washing machines will first dispense cold water at the beginning of every wash cycle regardless of the water temperature selection. This was observed when water temperatures were measured during the machine cycles using an iButton® Thermochron, which was placed in a capsule and added to the washers (Figure 16). Even though the on-demand water heater was set at 48.9°C (120°F) and was within five feet of both machines, the average water temperature did not exceed 32.5°C (90.5°F) for the agitator machine and 30°C (86°F) for the HE machine. Although the HE machine has a built-in water heater, the water did not get as hot as the agitator but may have kept the water temperature more consistent during the wash cycle.

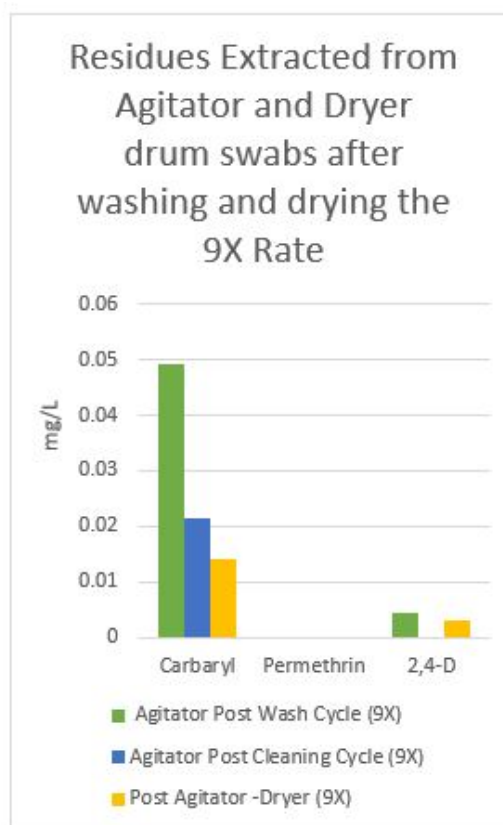
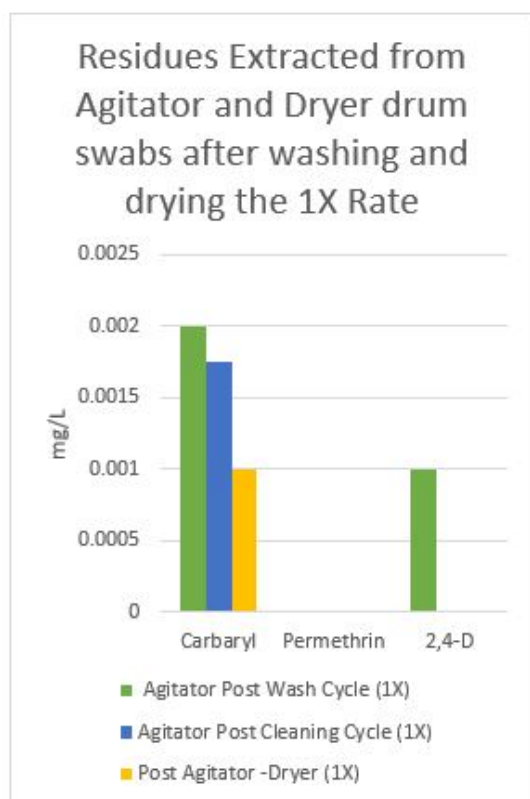


**Figure 16.** Average temperatures for Agitator and HE washing machines.

**Table 4. Gallons of water used during stages of wash cycle for agitator and high efficiency washer.**

Machine type	Prewash	Wash	Rinse	Extra Rinse	Total Gallons
Agitator		17.1	17.0		34.1
High Efficiency (HE)	2.8	2.8	5.4	3.2	14.2

Jacobsen (1999) investigated the differences between agitator and tumbler [sic] washers. (Jacobsen referred to HE washers as tumbler washers.) He described agitator washers as using 17-23 gallons of water per cycle with a total of 34-46 gallons for the complete cycle. The agitator washers fully submerge clothing during the cycles in order to “swish” as part of the cleaning process. By comparison, tumbler washers use 4-8 gallons per cycle for a total of 15-32 gallons for a full cycle. Tumbler washers partially fill the drum and use a tumbling motion to move clothes in/out of the water. The total volume of water used and volume of water per cycle are shown in Table 4. Jacobsen calculated that the ratio of wash solution to clothes changes from about 20:1 for the



**Figures 17 & 18.** Residues extracted from agitator washer and dryer drum swabs at 1X rate (left) and 9X rate (right).



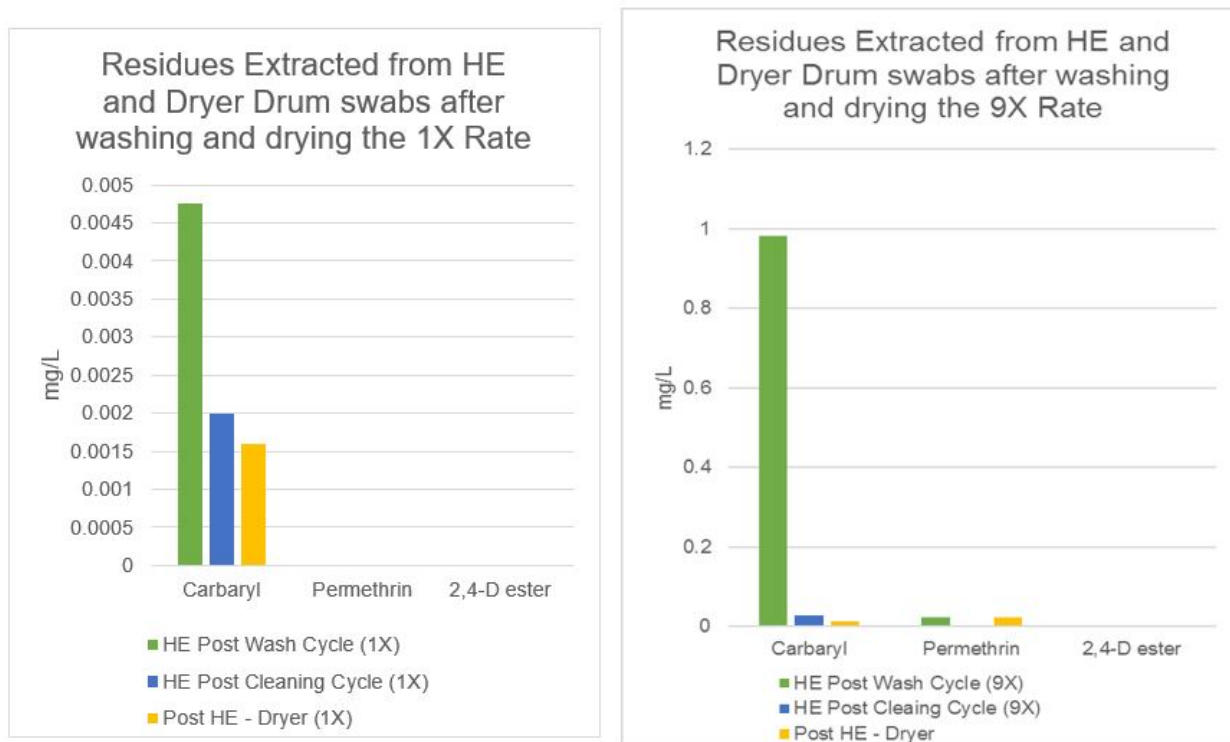
agitator washer to about 7:1 for the tumbler. The increase or decrease in the proportion of wash solution to clothing may account for some of the pesticide residue results previously reported, especially if the pesticide is water soluble.

## 4.6 Machine Swab Samples

One concern when decontaminating pesticide-exposed clothing using home equipment is whether any residues remain in the machines after washing and drying. As mentioned in the washing and drying sections above, swabs were used to wipe the machines after washing, after the cleaning cycle, and after drying. These swabs were analyzed to examine the potential for washer and dryer contamination from cleaning pesticide-contaminated clothing.

### 4.6.1 Agitator Results

The concentrations of residues extracted from swab samples after washing in the agitator washer, agitator washer after a cleaning cycle and dryer are presented in Figures 17 (1X rate) and 18 (9X rate). Carbaryl was detected in swab samples taken after the wash cycle at both the 1X and 9X rates. There was a reduction in residues in swabs taken after the cleaning cycle at both the 1X and 9X rates. Carbaryl was also detected, albeit in relatively very low levels, from the dryer swabs at both the 1X and 9X rates. Permethrin was not detected in any swab samples taken from the machines at either the 1X or 9X rate. At the 1X rate, 2,4-D ester was detected in the washer swabs



**Figures 19 & 20.** Residues extracted from agitator washer and dryer drum swabs at 1X rate (left) and 9X rate (right).

but not in swabs taken after running a cleaning cycle in the washer or in the dryer. At the 9X rate, 2,4-D ester was detected in swabs taken after the wash cycle but not after the cleaning cycle. However, 2,4-D ester was detected in swabs taken from the dryer after clothes were dried and is due to transfer of residues to the machine from the clothing as the residues were not present prior to drying the clothes.

#### **4.6.2 HE Results**

The concentration of residues extracted from swab samples after washing in the HE washer and running a cleaning cycle, and dryer are presented in Figures 19 (1X rate) and 20 (9X rate). At both the 1X and 9X rate, the concentration of carbaryl residues were higher in swabs taken after the wash cycle as compared to the swabs taken after the cleaning cycle. Carbaryl was also detected, albeit in relatively very low levels, from the dryer swabs at both the 1X and 9X rates. Permethrin was not detected in any swab samples taken from the machines at 1X rate but was detected from swabs after washing and drying at the 9X rate. Permethrin was not detected in the swabs post cleaning of the HE washer. Presence of 2,4-D ester was not detected in any of the swabs at the 1X or 9X rate.

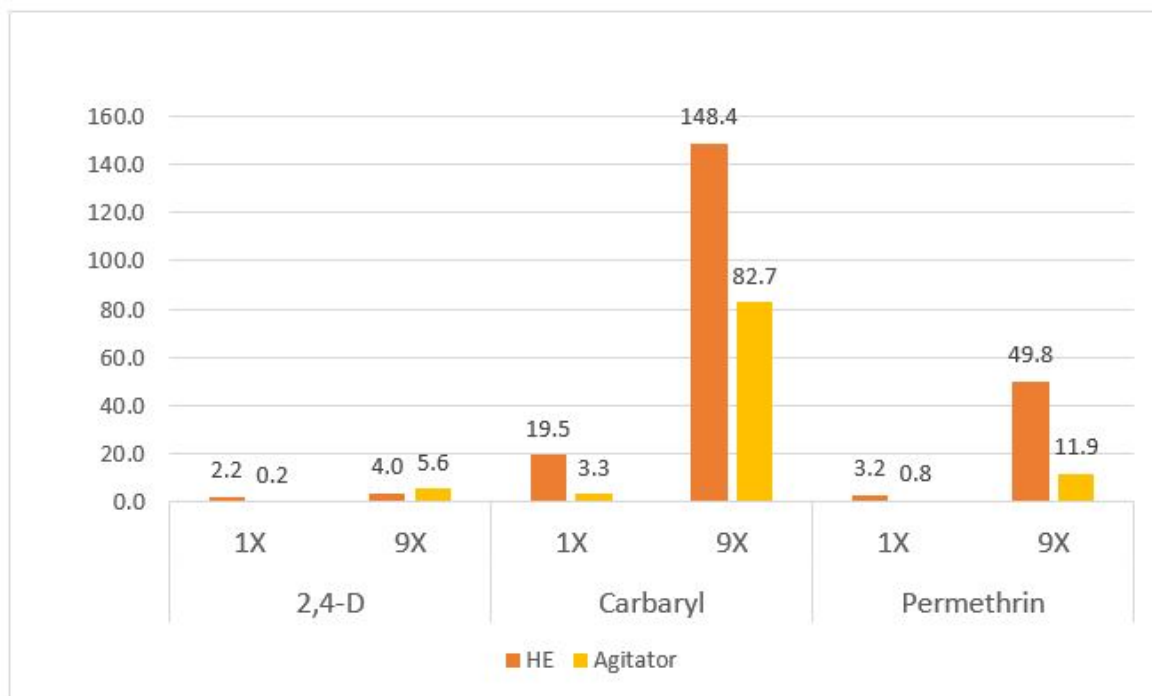
#### **4.6.3 Summary of Machine Swabs**

These results demonstrate the value of performing the cleaning cycle after washing pesticide-contaminated clothing. Due to the observation that there were more residues in the HE machine swabs after cleaning, it may be necessary to run an additional cleaning cycle in these types of machines. It is also a reminder that some residues that remain in clothing can be transferred to the dryer during the drying process and the dryer may require some additional cleaning by wiping the inner surface with a damp disposable towel.

#### **4.7 Discussion of Rinsate Samples and Environmental Fate**

The rinsate samples collected during the wash and rinse cycles reflect the volume of pesticides released into the environment per load. The mean concentration of each pesticide for an entire load was calculated by multiplying the concentration of a sampled wash cycle by the volume of discharged water to obtain a mass of pesticide per wash cycle. This mass was then divided by the total volume for the wash load to obtain a mean concentration across all cycles. This mean was then multiplied by the total wash volume to obtain a total pesticide mass load for the laundry load.

Mean concentration per load of laundry and total load of pesticide are provided in Figures 21 and 22. The pesticide with the highest concentration and load in the rinsate is carbaryl. This result would be expected since this pesticide was applied at a rate (two qt/acre) that far exceeded the other products. Additionally, a higher percentage of carbaryl was also removed from the clothing (Figure 7), which could also explain the higher concentration in the rinsate. The next highest pesticide in concentration and load was permethrin, but it was applied at a rate (eight oz/acre) lower than 2,4-D ester (three pt/acre). It was higher at the 9x rate but similar at the 1X rate to 2,4-D ester. For five of the six pesticide and spray rate combinations, the HE washing machine had a higher concentration of pesticide in the rinsate than the agitator type machine. However, when

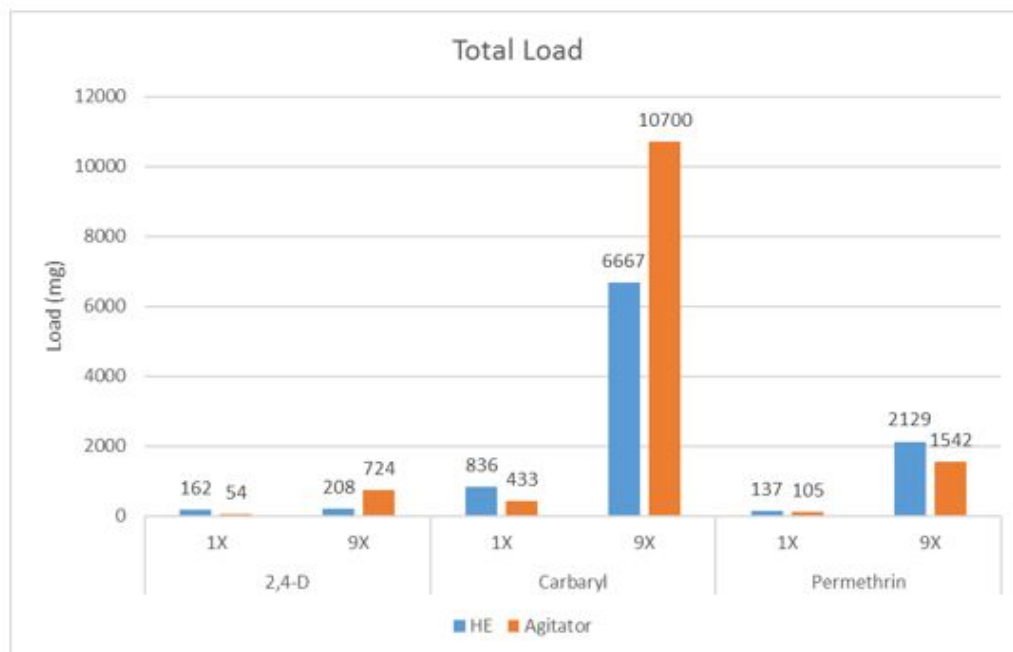


**Figure 21.** Mean concentration of pesticides detected in rinsate water.

total loads are calculated, the volume of pesticide released into the environment was similar or lower for HE due to the lower volume of water released.

The fate of the pesticide loads in the environment would depend on where the laundry of the applicator clothing takes place. In a municipality with a public wastewater treatment plant (WWTP), the pesticide residues could quickly be diluted by other contributions to the waste stream. However, they would contribute to other pesticides entering the system from other sources, especially if the situation were a combined storm and sewer waste stream. Removal and/or degradation of the pesticides would depend upon the type of WWTP employed at the plant.

In a rural environment where individual sanitary disposal systems (ISDS) are more common, pesticide residues in the rinsate would also be diluted by other water and wastes entering the system, but much less than a public system. In this situation, the pesticides could adhere to organic solids in the septic tank and/or remain in solution and be transported to the soil adsorption (leach) field, where the compounds could be adsorbed onto soil colloids and/or be subject to degradation by soil microbes. In an ISDS, the potential fate of moving through the leach field into groundwater should also be considered. The probability of this transport would depend upon the mass of the pesticide (load), the pesticide properties, how well the system is functioning, and the site characteristics. Pesticide properties that affect movement include their solubility, soil absorptivity, and half-life (Hornsby et al., 1996). Based upon these properties, carbaryl, permethrin, and 2,4-D ester are rated to have low, very low, and intermediate leaching potentials, respectively. However, for other pesticides with high leaching properties,



**Figure 22.** Total load of pesticides in rinse water.

frequent laundry washing of pesticide-laden clothing could be a potential source of contamination, especially given the high concentrations and loads shown in this study.

Finally, another scenario for environmental fate of pesticides in laundry water is when rinsate does not get treated in an ISDS or WWTP but is released into the environment as untreated greywater. Greywater is water from bathroom sinks, showers, tubs, and washing machines that has not come into contact with feces, either from the toilet or from washing diapers. In some states, this water is allowed to be used for irrigation, mostly in landscape plantings. Given the concentrations and loads of pesticides found in this study, an area receiving repeated exposure to laundry rinsate could have plant phytotoxicity or harm to beneficial insects. While the volume of pesticide entering the environment from laundry rinsate is low compared to all other sources, the fact that it enters through a wastewater supply could be significant in certain situations.

## 5. Recommendations

Based on the results of this study, both types of washing machines were effective at removing permethrin and carbaryl. While neither machine outperformed the other in this study, it should be noted that the agitator machine used here lacked options for a prewash and extra rinse while the HE washer included prewash and extra-rinse cycles. It is possible that if a consumer could supplement the agitator wash cycle with a prewash and extra rinse, the agitator machine may be more effective due to the extra volume of water used during the decontamination process.

Specific recommendations for which washing machine to use cannot be made based on chemical class. Keaschall et al. (1986) suggested that water solubility of the active ingredient may be a more reliable indicator of the relative ease of removal than

chemical class. However, the solubility of the entire formulation will impact the ease of removal of the pesticide as some are petroleum-based versus water-based. Laughlin et al. (1991) found the EC formulation of cypermethrin was more difficult to remove than the WP formulation. Sevin XLR (carbaryl) is miscible (mixes easily with water). Perm-Up (permethrin) is an EC and contains petroleum distillates, making it difficult to remove with just water, and Shredder 2,4-D ester is oil-based, becoming suspended in water.

The results of this study also did not indicate that one of the drying methods tested reduces pesticide residues much more than others. For carbaryl, either electric dryer or clothesline drying can be used as there was no effect of reducing residues based on drying method. However, permethrin residue levels slightly increased when dried in the electric dryer or on the clothesline, but neither increased residue more than the other. The study results indicate that both electric dryer and clothesline drying reduced residues of 2,4-D ester in all clothing types. Consumers should choose the method most convenient for them.

Whether UV-B was a factor in reducing residues when clothing is dried on clotheslines depended on the pesticide. For carbaryl, drying clothing in the electric dryer or on clotheslines did not further reduce residues, and exposure to UV-B alone did not degrade carbaryl residues. Drying permethrin-exposed clothing in the electric dryer and on the clothesline increased residue concentrations. However, when the clothing was exposed to UV-B alone, residues of permethrin were reduced. Therefore, UV-B can further reduce some pesticide residues after washing, but it appears to be dependent on active ingredient.

The type of clothing material used for personal protection during pesticide applications is important. In this study, permethrin and carbaryl were difficult to remove from the 100% cotton blue jeans. Based on this observation, 100% cotton blue jeans would not be a recommended clothing type for pesticide applications. If choosing to wear them, applicators should consider an additional protective layer in the form of waterproof pants (rain pants) or chemical-resistant chaps. Other options include applying starch to form a repellent surface; however, starch must be reapplied after each washing. Obendorf et al. (1991) and Csiszar et al. (1998) demonstrated that by using methyl parathion, starch can act as a pesticide trap on the fabric surface to decrease pesticide transfer and to enhance pesticide removal. Conversely, Laughlin et al. (1991) found that repellent-finished fabrics did not absorb all the pyrethroid pesticide solution, but unfinished fabric tended to have lower amounts of after-laundrying residues. Newer materials are constantly becoming available, and a viable option is ripstop fabric, Teflon-coated pant available online (<https://www.511tactical.com/stryke-pant.html>).

Heavier-weight fabrics tended to retain more residues than lighter-weight fabrics in this study. The thickness and weight of the 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 (Kim, et. al. 1982). Kim et al. (1986) found that due to their increased oleophilic nature, the polyester/cotton fabrics retained the oil-based EC formulation of alachlor more than the hydrophilic cotton fabrics. Easter (1983) suggested the same theory on fiber/soil interaction. The heavy-duty cotton work shirts used in this study may be appropriate for personal protection during applications, as



they are shown to absorb more of the pesticides, which may create a barrier and prevent dermal exposure. Lighter, non-woven materials used in this study may allow pesticides to reach the applicator's skin, but that was not investigated in this study.

Preliminary observations with 2,4-D ester in this study suggest delayed washing of pesticide-contaminated clothing may result in more pesticides being retained by the clothing. Recommendations should include washing pesticide-contaminated clothing at the end of each workday and not wearing the same clothing for multiple days before washing. Additional recommendations include discarding any clothing that is heavily contaminated by pesticide concentrates or multiple exposures to diluted sprays. The concentration of residues detected post-washing from clothing exposed to the 9X labeled rate were 50 times higher with carbaryl and nine times higher with permethrin, as compared to the labeled rate.

Recommendations should also emphasize that pesticide-contaminated clothing should be washed separately from all other clothing (including other work clothing), but especially the clothing from other family members. Transference of pesticide residues was observed with all the pesticides used in this study, regardless of washing machine used. The results of this study support previous research conducted by Braun et al. (1990) and Easley et al. (1983), which demonstrated that synthetic pyrethroids and 2,4-D esters can transfer from contaminated clothing to uncontaminated clothing during laundering. This led to the conclusion that pesticide-contaminated clothing should be laundered separately from the rest of the family wash (Easley et al., 1984).

Finally, this study confirmed that adding a "cleaning cycle" in the washer after decontaminating pesticide-exposed clothing, before washing other clothing, can reduce any pesticide residues that may remain in the washer.

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## Appendix

**Table A1.** Laundry study spray dates

Chemical	Rate	Rep	Sprayed	Weather	Washed
2,4-D Ester	1X	1	8/23/2016	78-80°F, RH 21-17%, Wind 2-8 mph, 294-30sixdegrees (WNW-NW)	8/23/2016
		2			8/23/2016
		3	9/1/2016	67-70°F, RH 71-63%, Wind 3-6 mph, 145-152 degrees (SE-SSE)	9/1/2016
		4			9/2/2016
	9X	1	9/12/2016	63-65°F, RH 44-40%, Wind 6 mph, 140-89 degrees (SSE-E)	9/12/2016
		2			9/13/2016
		3	9/15/2016	65-66°F, RH 49%, Wind 3-4 mph, 169-173 degrees (S)	9/15/2016
		4			9/16/2016
Carbaryl	1X	1	10/6/2016	45-46°F, RH 89-84%, Wind 2-3 mph, 71-898 degrees (ENE-E)	10/10/2016*
		2			10/11/2016*
		3	10/13/2016	60-65°F, RH 41-33%, Wind 3-4 mph, 147-158 degrees (SSE)	10/13/2016
		4			10/17/2016*
	9X	1	5/17/2017	60-63°F, RH 49-41%, Wind 7-9 mph, 159-153 degrees (SSE)	5/17/2017
		2			5/18/2017
		3	5/24/2017	56-59°F, RH 8-9%, Wind 5-sevenmph, 162-148 degrees (SSE)	5/24/2017
		4			5/24/2017
Permethrin	1X	1	6/21/2017	82-85°F, RH 10-7%, Wind 4-10 mph, 322-306 degrees (NW)	6/21/2017
		2			6/22/2017
		3	6/28/2017	68-75°F, RH 45-32%, Wind 7-9 mph, 163-174 degrees (SSE-S)	6/28/2017
		4			6/29/2017
	9X	1	7/5/2017	67-69°F, RH 33-26%, Wind 2-5 mph, 323-11 degrees (NW-N)	7/5/2017
		2			7/6/2017
		3	7/12/17	67F-70°F, RH 71-62%, Wind 5-9 mph, 11 degrees (N)	7/12/2017
		4			7/13/2017

\*Clothing was stored in a Amana® freezer at 0°F after spraying until washed. Clothing was allowed to "thaw" overnight prior to washing.

**Figure A1.** Washing process (replicated 4X for each treatment).

