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Longitudinal, Seasonal, and Occupational Trends of Multiple Pesticides in House Dust

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Background: Children are especially vulnerable to pesticide exposure and can suffer lasting health effects. Because children of farmworkers are exposed to a variety of pesticides throughout development, it is important to explore temporal patterns of coexposures.

Objectives: The objectives of this study were to characterize the pesticide co-exposures, determine how they change over time, and assess differences between farmworker and nonfarmworker households.

Methods: Dust collected from 40 farmworker and 35 nonfarmworker households in the Yakima Valley of the State of Washington in 2005 and then again in 2011 was analyzed for 99 pesticides. Eighty-seven pesticides representing over 28 classes were detected. Pesticides were grouped into classes using U.S. EPA pesticide chemical classifications, and trends in concentrations were analyzed at the class level.

Results: Levels of organophosphates, pyridazinones, and phenols significantly decreased between 2005 and 2011 in both farmworker and nonfarmworker households. Levels of anilides, 2,6-dinitroanilines, chlorophenols, triclosan, and guanidines significantly increased in both farmworker and nonfarmworker households in 2011 vs. 2005. Among farmworkers alone, there were significantly lower levels of *N*-methyl carbamates and neonicotinoids in 2011.

Conclusions: We observed significant reductions in the concentrations of many pesticides over time in both farmworker and nonfarmworker households. Although nonfarmworker households generally had lower concentrations of pesticides, it is important to note that in comparison with NHANES participants, nonfarmworkers and their families still had significantly higher concentrations of urinary pesticide metabolites. This finding highlights the importance of detailed longitudinal exposure monitoring to capture changes in agricultural and residential pesticide use over time. This foundation provides an avenue to track longitudinal pesticide exposures in an intervention or regulatory context. https://doi.org/10.1289/EHP3644

Introduction

Agriculture in the State of Washington

Agriculture is an important industry in the State of Washington. The food and agriculture industry as a whole makes up approximately 13% of the state's economy and employs around 160,000 people (Washington State Department of Agriculture 2015). Washington State's apples and pears (pome fruits), as of 2013, accounted for 57.0% and 49.5% of total U.S. production, respectively (USDA-NASS 2015).

Chemical Inputs in Agriculture

Pome fruits are susceptible to a number of pests and diseases, so growers often turn to the use of synthetic pesticides to maintain agricultural productivity (Slattery et al. 2011). We used the Quick Stats function of the U.S. Department of Agirgulture's National Agriculture Statistics Service (USDA-NASS) Database (USDA-NASS 2011) to identify commonly used pesticides in the State of Washington's apple production, including organophosphate (OP), neonicotinoid, or carbamate insecticides; azole, or anilide fungicides; and herbicides like 2,4-dichlorophenoxyacetic acid (2,4-D), and pendimethalin. Many of these pesticides have known or suspected adverse health effects. For example, the U.S. Environmental Protection Agency (U.S. EPA) Endocrine Disruptor Screening Program (EDSP) recently assessed the potential endocrine disruption effects of 52 pesticides, 23 of which are used in Washington State apple production (U.S. EPA 2017; USDA-NASS 2011). Of those 23 pesticides—both used in apples and assessed in the EDSP—6 were identified as showing potential interaction with thyroid, androgen, and/or estrogen pathways (U.S. EPA 2017). Additionally, several pesticides used in apple production have been identified as being neurotoxic in human and/or animal systems, including several historically high-use pesticides (i.e., greater than 100,000 pounds applied annually in Washington State apple production), such as the organophosphates (OPs), with reported long-term chronic effects, including neurodevelopmental delays and attention and hyperactivity deficits (Bouchard et al. 2011; Engel et al. 2011; Eskenazi et al. 2007; Furlong et al. 2014; González-Alzaga et al. 2014, 2015; Marks et al. 2010; Rauh et al. 2011). In addition to OPs, *N*-methyl carbamates, dithiocarbamate, and azole fungicides have the potential to cause neurotoxicity in humans (U.S. EPA 2005b, 2006a, 2006c, 2006d, 2008a, 2012b; USDA-NASS 2011). Farmworkers responsible for growing and maintaining crops that have been sprayed with these pesticides are at a much greater risk than nonfarmworking individuals are of being exposed and potentially experiencing adverse health effects.

Occupational Take-Home Pathway

Occupational exposure to pesticides and other agricultural chemicals affects both farmworkers and their families. Previous studies with pome fruit farmworkers in the Yakima Valley of Washington State have provided evidence that agricultural workers carry OP pesticide residues home with them. OPs have been detected in both the house and vehicle dust of agricultural workers to a much higher degree than among nonfarmworkers, and children of agricultural workers have higher levels of OP metabolites in their urine than

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children of nonagricultural households have (Coronado et al. 2006; Curl et al. 2002; Thompson et al. 2014; Tamaro et al. 2018). This take-home exposure pathway is of particular concern because a child's brain continues to develop long after birth, and exposure to neurotoxicants like OPs at a young age can have lasting behavioral and neurological effects (Grandjean and Landrigan 2014; NRC 1993).

Residential Pesticide Exposure

In addition to the exposure to various agricultural pesticides that may be brought home via the agricultural take-home pathway, children can also be exposed to pesticides through residential (household) pesticide use. Many common household pesticides include 2,4-D, glyphosate, carbaryl, malathion, and several pyrethroids, which are all used frequently in agriculture as well (Atwood and Paisley-Jones 2017; Grube et al. 2011; Guha et al. 2013). Thus, the agricultural take-home pathway coupled with residential pesticide exposure may put children living with farmworkers at a higher risk of developing adverse health outcomes, such as neurotoxic effects, from pesticide exposure.

Pesticide Coexposures

With the multiple pesticides children of farmworkers may be exposed to, it becomes critical to evaluate the impact that these coexposures have on their health. For example, OP and *N*-methyl carbamate pesticides both act by inhibiting acetylcholinesterase (Futako 1990); thus, coexposures can lead to even greater accumulation of acetylcholine and subsequent neurotoxic effects. A trio of child health studies observed significant neurodevelopmental delays associated with prenatal and early childhood exposure to OPs (Bouchard et al. 2011; Engel et al. 2011). Other pesticide groups, such as the neonicotinoids have been associated with impaired neurodevelopment in animal models (Crosby et al. 2015), and combined exposure to OPs and pyrethroids have been associated with increased toxicity relative to exposure to a single pesticide (Iyyadurai et al. 2014).

In the present study, we used household dust to characterize pesticide coexposures in a longitudinal agricultural children's cohort. By looking across agricultural seasons in farmworker and nonfarmworker households, we can use pesticide levels detected in household dust to demonstrate how the exposures change over time. This method is especially relevant during the study period, 2005–2011, as the phase-out of the most common agricultural insecticide in Washington State apple production, azinphos-methyl, was implemented during this time period (Goldberger et al. 2011; U.S. EPA 2012a). By examining pesticide coexposures, we can identify which pesticides may be replacing azinphos-methyl and the implications for cumulative health impacts. Furthermore, we hypothesize that we can discern differences in pesticide coexposures between farmworker households, which should be subject to the occupational take-home pathway, and nonfarmworker households.

Methods

Cohort Description and Sample Collection

The University of Washington Center for Child Environmental Health Risks Research (CHC) Cohort is located in the lower Yakima Valley, one of Washington State's major agricultural regions. In 2005, a cohort of 200 households was recruited as previously described in Thompson et al. 2014. The cohort was predominantly Hispanic (>95%) and split approximately evenly between farmworker (FW) and nonfarmworker (NFW) families, each with a referent child age 2–6 y. FWs were defined as individuals who worked with pome fruit crops (apples and pears), as the initial study focus was on orchard crops that received OP applications. House dust was collected from a sample of 75 households (40 FW and 35 NFW), analyzed in both 2005 and 2011, and our findings are included in the analyses in this paper. Demographic characteristics of the households in both sampling years are shown in Table S1. Marital status and housing type were similar between sampling years; however, income was higher in 2011 than in 2005 for both FW and NFW. In both years, most study participants lived in single-family homes. If participants changed residence between the study years, we sampled at their new homes in 2011, as this study focused on the participants and their occupations.

Household dust samples were collected from each residence during April–July of 2005 and June–August of 2011. Although the collection months differed between the two studies, collection periods were selected so the agricultural activities matched between years, both representing the "thinning season," when buds and small fruit are removed from the trees to promote the growth of larger fruit. Previous studies have indicated that OP exposure, both in household dust and urine, is highest during the thinning season (Smith et al. 2017; Thompson et al. 2014; Tamaro et al. 2018). In 2005, household dust samples were collected using a Nilfisk GS-80 vacuum cleaner unit as previously described (Smith et al. 2017). Briefly, dust was collected from areas identified as frequent child play areas. Dust collection was standardized using $0.5 \times 0.5 m^2$ templates, and the floor surface (i.e., plush carpet, thin carpet, hard floors) was accounted for by the number of templates collected. The hose, nozzle, and lower container of the vacuum were cleaned between each use, and a new polyliner and vacuum bag were used for each sample collection. Dust samples from 2011 were collected in the same manner as 2005; however, a Metropolitan VM-500 High-Powered hand-led vacuum was used instead of the Nilfisk. One Metropolitan vacuum was used per household, and the nozzle and stainless steel sections were washed thoroughly before use. We compared the amount of dust collected by the Nilfisk with the amount collected by the Metropolitan vacuum (Table S2) and the detection rates by year (Table 1) to determine that, although the Metropolitan vacuum collected less dust than Nilfisk vacuum, both machines were able to collect sufficient dust for pesticide analysis, based on the level of detection. All participants provided informed consent, and all sample collection procedures were reviewed and approved by the Fred Hutchinson Cancer Research Center's Institutional Review Board (File IR 5946). Samples were analyzed under the University of Washington Institutional Review Board (Files 40794 and 40570).

Sample Preparation

All dust samples were transferred to 150-μm metal sieves (VWR) and sieved for 10 min. Dust passing through the sieve was weighed and partitioned into two 1-g aliquots, and the remaining was saved in a separate container. If less than 2 g of dust were sieved, the second aliquot contained whatever dust remained after the first 1-g aliquot. All dust samples were stored at −10°C until sample analysis. Of the initial cohort of 200 households, dust samples were collected and analyzed from 75 households for both 2005 and 2011.

Pesticide Selection and Analysis

We identified 305 candidate pesticides used in Washington State (USDA-NASS 2003, 2011). Fifty-five of the initial 305 candidates were screened out because of analytical difficulties, including low stability in dust, unavailable LC-MS calibrates, or incompatibility with available LC-MS technology. From the remaining 250 compounds, 145 were selected for further analysis though prioritization

Table 1. Classification, uses, and detection rates for 47 pesticides by class with over 5% of samples above the LOD for either 2005 or 2011.

Pesticide	Registered Agricultural Use	Registered Residential[a] Use	Percent of Households <LOD (By Class)		LOD[b] per Sample (ng/sample)	
			2005	2011	2005	2011
Insecticides						
Organophosphate			4%	2%		
Azinphosmethyl	X (phased out)				2, 8, 10	2, 4, 5
Chlorpyrifos	X				2, 4, 8	2, 4
Coumaphos	X				1, 4	0.5, 1, 4
Diazinon	X	Uses Canceled			0.4, 2, 4	0.2, 0.4, 2
Dichlorvos	X	X			2, 5, 10, 20	2, 5
Malathion	X				0.4, 4, 20	0.2, 0.4, 4
Methamidophos	X				1, 2, 20	1, 2, 4
Phosmet	X	X			0.4, 4, 20	0.2, 0.4, 4
Tetrachlorvinphos	X	X			10, 20	4, 5
N-Methyl Carbamate			16%	7%		
Carbaryl	X	X			1, 2	0.5, 2
Methomyl	X				0.4, 2, 20	0.2, 2
Propoxur		X			0.4, 4	0.2, 0.4
Neonicotinoid			44%	40%		
Acetamiprid	X	X			0.4, 4, 20	0.2, 4
Clothianidin	X				0.4, 2, 20	0.2, 2
Imidacloprid	X	X			1, 4, 20	0.2, 0.5, 2, 4
Pyrethroid			28%	13%		
Cyphenothrin		X			20	10, 20, 200
Imiprothrin		X			10, 20	4, 5
Permethrin	X	X			10, 40	10, 40
S-Bioallethrin		X			2, 20	2, 4, 10
Sumithrin		X			20, 40	20
Tetramethrin		X			2, 4	0.4, 2
Insect Growth Regulator[c]			91%	58%		
Hexythiazox	X	Proposed			4, 10	4, 5
Pyriproxyfen	X	X			0.4, 10	0.4, 5
Urea/Insect Growth Regulator[cd]			13%	4%		
Novaluron	X				2, 4	2, 4
Macrocyclic Lactone[c]			79%	65%		
Spinosyn A	X	X			2, 10	2, 5
Spinosyn D	X	X			2, 10	2, 5
Synergist			27%	33%		
Piperonyl Butoxide	X				4, 10	4, 5, 20
Organosulphite[c]			70%	98%		
Propargite	X				4, 40	2, 20, 40
Fungicides						
Azole			27%	12%		
Myclobutanil	X				0.4, 10	0.4, 2, 5
Propiconazole	X	X			0.4, 2, 8	0.2, 2
Tebuconazole	X	X			0.4, 2, 4	0.2, 2
Azole (Benzimidazole)						
Thiophanatemethyl	X				40	20
Azole (Imidazole)						
Triflumizole	X				0.4, 2, 4	0.2, 0.4, 2
Strobin[c]			91%	49%		
Azoxystrobin	X	X			0.4, 4	0.4, 2
Trifloxystrobin	X				2, 10	2, 5
Anilide			52%	11%		
Boscalid	X				2, 20	4, 10
Quinonline[c]			81%	94%		
Quinoxyfen	X				10	5
Herbicides						
Urea			13%	4%		
Diuron	X	X			2, 4	0.2, 2
Chlorophenoxy acid or ester			35%	16%		
2,4-D	X				20	2, 10
MCPA	X				2, 4	2

[a]Residential use is defined as indoor household use in this analysis.
[b]Multiple LODs are due to analysis was done in batches, and each batch had own LOD.
[c]Macrocyclic lactones, insect growth regulators, quinolines and strobins were all below the LOD for over 75% of households in 2005, and quinolines and organosulphites were below the LOD for over 75% of households in 2011. Thus, these pesticides were excluded from longitudinal analyses.
[d]Novaluron is chemically a benzophenyl urea compound and classified as an insect growth regulator. For the purposes of this analysis, novaluron was grouped with the urea herbicide, diuron based on their shared urea chemical classification.
[e]DGH is registered as a microbiocide for indoor residential use. However, DGH and dodine are both salts of the same chemical, dissociate similarly, are considered bioequivalents, and are toxicologically the same (U.S. EPA 2005a). Dodine is a guanidine fungicide registered for agricultural uses.

Table 1. (*Continued.*)

Pesticide	Registered Agricultural Use	Registered Residential[a] Use	Percent of Households <LOD (By Class) 2005	Percent of Households <LOD (By Class) 2011	LOD[b] per Sample (ng/sample) 2005	LOD[b] per Sample (ng/sample) 2011
MCPP					2, 4	2
2,6-Dinitroaniline			43%	4%		
Pendimethalin	X				0.4, 20	0.2, 0.4
Pyridazinone			73%	58%		
Norflurazon	X				2, 10	2, 5
Pyridaben	X				0.4, 4	0.4, 2
Microbiocides						
Chlorphenol			33%	7%		
Triclosan		X			4, 20	4, 10
Guanidine			46%	9%		
DGH[e]	DODINE	DGH			10, 20	20
Phenol			38%	70%		
Na o-phenylphenate	X	X	38%	70%	4, 20	2, 20

based on their toxicity, agricultural use, and home use in Washington State. Of those 145 compounds, 99 were analyzed based on availability of LC-MS standards.

Dust samples were analyzed for 99 pesticides. Acetone (10 mL) was added to the sample, and samples were sonicated for 1 min at 20 kHz using a horn-type cell disrupter and then centrifuged for 5 min at 3,000 rpm. The supernatant (8.0 ml) was transferred to a 50-mL turbo-vap tube and evaporated to less than 1 mL at 45°C. Samples were vortexed, washed with 3–4 mL of cyclohexane, and evaporated to less than 1 mL at 45°C. Sample volume was increased to 1 mL by adding cylcohexane, then 2.5 mL of 20% dichloromethane in cyclcohexane was added. Samples were centrifuged at 3,000 rpm for 10 min. Gel Permeation Chromatography (GCP) cleanup of dust extract was completed using High Performance Liquid Chromatography (HPLC) and the Waters Sample Manager programmed to inject 3.5 mL of sample with a run time of 55 min. Collection time was 21–55 min. Fractions were evaporated to a volume less than 1 mL by using the sensor endpoint with 14 psi nitrogen at 60°C. A solvent exchange was conducted by rinsing the sides of the Turbovap flask with 4–5 mL Trimethyl phosphate (TMP) and evaporating the sample again to the sensor endpoint, leaving a final volume of slightly less than 0.5 mL. Samples were resuspended to the 0.5-mL mark using TMP. All samples were filtered with a 0.45-µm syringe filter before transferring into a 2-mL GC vial. Vials were stored at −15°C until analysis.

Analysis of the sample extract was performed by HPLC-tandem mass spectrometry (HPLC-MS/MS) using stable isotope-dilution quantification. Briefly, an Agilent 6410 HPLC-MS/MS was operated in positive electrospray ionization (ESI+) and multiple reaction mode (MRM) with nitrogen collision gas (Gas Temp: 350C; Gas Flow: 9 L/minute; Nebulizer: 40psi; Capillary Voltage: 4000V). A subset of compounds (2,4-D; 2,4-DP; MCPA; MCPP; 2-phenyl phenate; triclosan) were quantified by negative electrospray ionization (ESI−).

The HPLC system was equipped with a Gemini (Phenomenex) C18 reverse-phase column (3 micron, 150 × 2.0 mm), with a Gemini 4 × 2.0 mm guard column. All solvents (HPLC-grade) and deionized water (Barnstead Nanopure II, 18 MΩ) used were monitored for background and included in procedural blanks. Additional details on the instrument parameters can be found in Armstrong et al. 2014.

Eighty-seven pesticides representing over 28 classes were detected above the level of quantification in house dust. Seven pesticides were detected but were all below level of quantification, and five had insufficient response to detect level. Analytical QC metrics are shown in Table S3.

Characterization of House Dust Pesticide Profiles

Individual pesticides were grouped into their chemical classes using the U.S. EPA chemical classification system. Any pesticides that were in a class by themselves (e.g., triclosan, boscalid) were listed with the pesticide name in parenthesis [e.g., anilide (boscalid)]. The breadth of pesticides and classes detected are described in the results section and listed in Tables S4 and S5. Longitudinal analysis was completed using pesticides with greater than 5% detection in household dust (47 pesticides; see Table 1). For pesticide classes with levels below the LOD for over 75% of households in only one of the two study years, additional longitudinal analyses could not be completed ($n = 5$; see footnote for Table 1). For the remaining pesticide classes, individual pesticides' levels below the LOD were given a value of one-half LOD as a conservative estimate of the pesticide concentrations. This approach is supported by our previous work (Thompson et al. 2014).

Statistical Analysis

Heat-map generation. A heat map was generated using Microsoft Excel™ to show the overall pesticide profile. By using the pesticide levels detected in household dust, we compared the concentrations of all pesticides in each class detected from individual households in 2005 and 2011. For visualization purposes, a tricolor (red, yellow, green) scoring system was devised to relate each pesticide class to the 2005 levels of OPs and reported as 75th, 50th, or 25th percentile. The 2005 levels of OPs were chosen as the comparison standard because they were the most highly used pesticide class and were reflective of pesticide use in pome fruit production. This selection allowed us to easily visualize changes over time.

FW vs. NFW comparisons. The difference between the levels of pesticides detected in household dust between FW and NFW households was assessed using the heat-map color scheme, described above, to determine the fraction of households with high and low pesticide concentrations. To do this, we generated two categories of pesticide concentrations: one for classes greater than or equal to the 75th percentile of OPs detected in 2005, and one for classes below the 75th percentile of OPs detected in 2005. This binary distinction enabled us to use a nonparametric proportions test to compare the "redness" of FW vs. NFW, where redness was used to indicate households with pesticide levels greater than the 75th percentile of OPs (households colored red in the pesticide heat map). Using a nonparametric proportions test allowed for the determination of whether FWs or NFWs had a higher proportions of households with the highest levels of pesticides detected in dust for each pesticide class, which

provided an indication of how the pesticide-exposure profiles differed between FW and NFW households.

Comparisons across time. For each household, it was determined whether the levels of each pesticide class increased, decreased, or stayed the same between 2005 and 2011 (e.g., $[OP]_{2011} - [OP]_{2005}$). Then, using a nonparametric proportions test, we compared the proportion of FW vs. NFW households that had decreased pesticide levels between 2005 and 2011. This comparison allowed for the determination of how pesticide exposure has changed over time and whether those changes are different between FW and NFW households.

Additionally, a mixed effects model was used to examine the broader trend in pesticide levels between these two assessment periods. The mixed effects model used for this analysis was:

$$Log\,(Y) = a + b(t) + c(occ) + d(t \times occ) + R(\sigma_b) + E(\sigma_w),$$

where Y is the sum of all pesticide levels within a given class (in nmol), a is the 2005 FW average pesticide level, b is the coefficient for the time adjustment (2011 relative to 2005), t is time, c is coefficient for the occupation adjustment (NFW relative to FW), occ is occupation, d is the coefficient for the interaction adjustment for time and occupation, R is the random effect to account for between-household variability (measured by σ_b), and E is the within-household variability (measured by σ_w). This analysis allowed us to determine whether time had an effect on pesticide levels and whether those changes in time were affected by occupation. In addition, we looked at the effect of time within each occupation separately, which allowed us to see whether pesticide levels changed with time for each occupation independently.

Results

Dust Analysis

Eighty-seven of the 99 pesticides were analyzed and detected in the dust and are listed in Tables S4 and S5. Of these 87 pesticides, 47 were detected above the LOD in over 5% of samples from either 2005 or 2011. Table 1 details the full list of 47 pesticides that were analyzed, their registered uses, and the limit of detection for each. Figure 1 shows the relative concentrations of the 47 pesticides by class with over 5% of samples above the LOD for either 2005 or 2011. The red coloring in Figure 1 indicates pesticide levels that were greater than the 75th percentile of OPs in 2005 (4.63 nmol per g dust), which was the pesticide class with the highest geometric mean household dust concentration (1.17 nmol per g dust) for that year.

Pesticide Coexposure Analysis

Insecticides. Of the 87 pesticides detected in household dust, 28 pesticides in 9 different pesticide classes were classified as insecticides. The majority of the OPs, *N*-methyl carbamates, neonicotinoids, insect growth regulators, urea growth regulators, macrocyclic lactones, synergists, and organosulfites were registered for agricultural uses during the study. All of the pyrethroids detected in

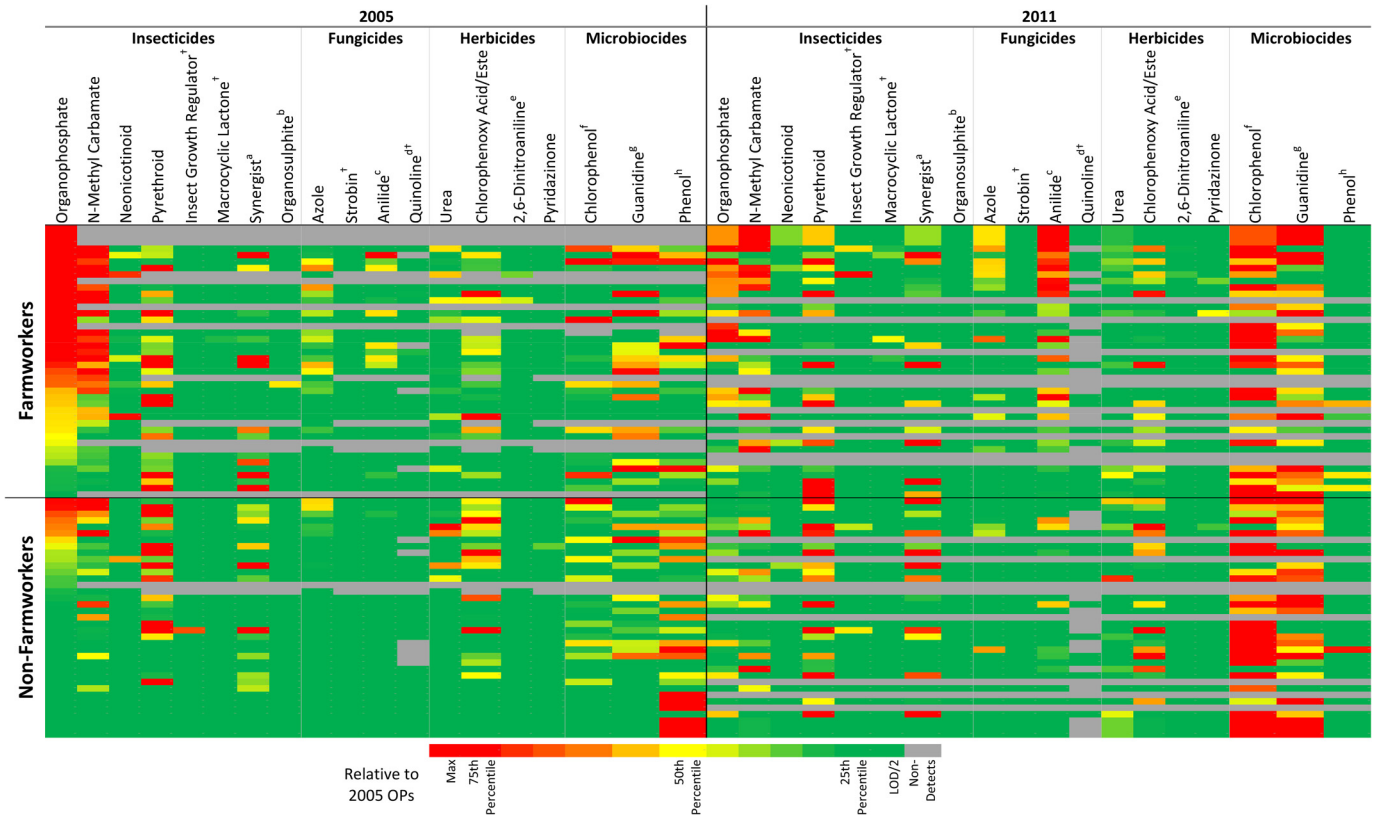

Figure 1. Heat Map for Pesticides Detected in Household Dust. The 47 pesticides with a detection rate greater than 5% in either 2005 or 2011 were grouped into 19 different pesticide classes. The figure shows the relative concentrations of pesticides, by class, over the two sampling periods (2005, left, and 2011, right). The coloring is relative to the 25th, 50th, and 75th percentiles of organophosphates in 2005, with red representing higher pesticide concentrations. Pesticide classes marked by a single dagger (†) denote classes that had over 75% of households with pesticide levels below the level of detection (LOD). These pesticides were not included in additional analyses. Red coloring defines pesticide concentrations in the 75th percentile of 2005 organophosphate pesticides (OPs) (4.63 nmol/g), yellow represents the 50th percentile of 2005 OPs (1.13 nmol/g), and green represents the 25th percentile (0.32 nmol/g). [a]Piperonyl Butoxide, [b]Propargite, [c]Bosaclid, [d]Quinoxyfen, [e]Pendimethalin, [f]Triclosan, [g]DGH, [h]Propargite.

household dust, with the exception of permethrin, are registered for residential household use only (i.e., no registered agricultural uses; Table 1).

Organophosphate insecticides in household dust. In 2005, 48% of FW households had OP levels higher than the 75th percentile (4.63 nmol/g dust; Table 2). This finding was significantly higher than that of the NFW households, of which none had OP levels greater than the 75th percentile (Table 2, $p < 0.001$). Although fewer FW households had pesticide levels above 4.63 nmol/g dust in 2011 (13%), there was still a significantly greater proportion of FW households than NFW households above this level (Table 2, $p = 0.05$). OP concentrations in household dust were significantly lower in 2011 than in 2005 (Table 3, $p = 0.003$) for both occupational groups combined. This decrease in OP concentrations, however, was largely driven by FW households. Among FWs alone, OP levels were significantly lower in 2011 (Table 3, $p < 0.001$), whereas among NFWs, the decrease in OP levels that occurred between 2005 and 2011 was not significant ($p = 0.55$). The proportion of individual FW households that showed a decrease in OP pesticide levels between 2005 and 2011 was 83%, which is significantly greater than the 56% of NFW households that showed decreased OP levels (Table 2, $p = 0.02$).

***N*-methyl carbamates in household dust.** In 2005, 32% of FW households had *N*-methyl carbamate levels that were higher than the 75th percentile for OPs (4.63 nmol/g dust), which was significantly greater than the 9% of NFW households (Table 2, $p = 0.02$). In 2011, however, no statistically significant difference was found in the number of FW and NFW households with pesticides above this level (Table 2, $p = 0.10$). The concentration of *N*-methyl carbamates in the household dust was significantly lower in 2011 than in 2005 among FWs (Table 3, $p = 0.02$), but not among NFWs ($p = 0.40$). However, there was no significant difference between FWs and NFWs in the proportion of individual households that showed a decrease in *N*-methyl carbamate levels between 2005 and 2011 (Table 2, $p = 0.30$).

Neonicotinoid insecticides in household dust. In both 2005 and 2011, there were no households with neonicotinoid levels higher than the 75th percentile for OPs (4.63 nmol/g dust; Figure 1) in either occupational group (Table 2). The concentration of neonicotinoids in household dust was significantly lower in 2011 than in 2005 among FWs (Table 3, $p = 0.03$), but not among NFWs ($p = 0.40$). Additionally, 71% of FW households had decreased neonicotinoid levels in dust between 2005 and 2011, which is significantly greater than the 37% of NFW households with decreased levels over the same time period (Table 2, $p = 0.02$).

Pyrethroid insecticides in household dust. In 2005, we found no significant difference in the proportion of households higher than the 75th percentile of OPs between FWs and NFWs for the pyrethroids (Table 2, p = 0.83). Likewise, in 2011, the proportion of FW households with pesticide levels above 4.63 nmol/g dust was not significantly different from the proportion of NFW households above this level (Table 2, p = 0.59). The concentration of pyrethroids was not significantly different between 2005 and 2011, regardless of occupation (Table 3). However, 77% of FW households had lower pyrethroid levels in 2011, which is significantly greater than the 48% of NFW households that had decreased pyrethroid levels (Table 2, $p = 0.04$).

Other insecticides detected in household dust. In 2005, concentrations of macrocyclic lactones and insect growth regulators were below the LOD for over 75% of households; in 2011, the levels of the organosulfite insecticide propargite was below the LOD for over 75% of households (Table 1). As a result, these insecticides were excluded from further longitudinal analyses and will not be discussed in detail. Additionally, the levels of

Table 2. Summary of proportional statistical tests.

	Proportion Red, 2005[a]				Proportion Red, 2011[a]				Proportion Decrease ('05-'11)			
	FW	NFW	Difference (95% CI)	*p*-value	FW	NFW	Difference (95% CI)	*p*-value	FW	NFW	Difference (95% CI)	*p*-value
Insecticides												
Organophosphate	0.48	0.00	0.48 (0.32, 0.63)	<0.001***	0.13	0.00	0.13 (0.01, 0.25)	0.05*	0.83	0.56	0.28 (0.05, 0.51)	0.02*
N-Methyl Carbamate	0.32	0.09	0.23 (0.05, 0.42)	0.02*	0.23	0.07	0.16 (−0.02, 0.34)	0.10	0.63	0.48	0.14 (−0.13, 0.41)	0.30
Neonicotinoid	0.00	0.00	—	—	0.00	0.00	—	—	0.71	0.37	0.34 (0.08, 0.60)	0.02*
Pyrethroid	0.27	0.24	0.03 (−0.19, 0.24)	0.83	0.20	0.26	−0.06 (−0.28, 0.16)	0.59	0.77	0.48	0.29 (0.03, 0.55)	0.04*
Insect Growth Regulator	0.00	0.00	—	—	0.03	0.00	0.03 (−0.03, 0.10)	0.34	NA	NA	NA	NA
Macrocyclic Lactone	0.00	0.00	—	—	0.00	0.00	—	—	NA	NA	NA	NA
Synergist (Piperonyl Butoxide)	0.17	0.06	0.11 (−0.05, 0.26)	0.18	0.13	0.15	−0.02 (−0.20, 0.17)	0.87	0.59	0.52	0.07 (−0.21, 0.35)	0.61
Organosulphite (Propargite)	0.00	0.00	—	—	0.00	0.00	—	—	NA	NA	NA	NA
Fungicides												
Azole	0.00	0.00	—	—	0.00	0.00	—	—	0.58	0.59	−0.01 (−0.28, 0.26)	0.95
Strobin	0.00	0.00	—	—	0.00	0.00	—	—	NA	NA	NA	NA
Anilide (Boscalid)	0.03	0.00	0.03 (−0.03, 0.10)	0.29	0.17	0.00	0.17 (0.03, 0.30)	0.03*	0.27	0.15	0.13 (−0.10, 0.35)	0.28
Herbicides												
Urea	0.00	0.03	−0.03 (−0.09, 0.03)	0.31	0.00	0.00	—	—	0.50	0.44	0.06 (−0.22, 0.33)	0.69
Chlorophenoxy Acid/Ester	0.07	0.09	−0.02 (−0.16, 0.11)	0.75	0.00	0.11	−0.11 (−0.23, 0.01)	0.06	0.62	0.48	0.14 (−0.14, 0.41)	0.34
2,6-Dinitroaniline (Pendimethalin)	0.00	0.00	—	—	0.00	0.00	—	—	0.17	0.33	−0.17 (−0.40, 0.07)	0.17
Pyridazinone	0.00	0.00	—	—	0.00	0.00	—	—	0.68	0.78	−0.10 (−0.35, 0.15)	0.45
Microbiocides												
Chlorophenol (Triclosan)	0.07	0.03	0.04 (−0.07, 0.15)	0.48	0.43	0.59	−0.16 (−0.42, 0.10)	0.23	0.14	0.07	0.07 (−0.11, 0.25)	0.44
Guanidine (DGH)	0.13	0.03	0.10 (−0.03, 0.24)	0.13	0.23	0.26	−0.03 (−0.25, 0.20)	0.82	0.36	0.11	0.25 (0.02, 0.49)	0.04*
Phenol (Na o-Phenylphenate)	0.07	0.15	−0.08 (−0.24, 0.07)	0.31	0.00	0.00	—	—	0.71	0.74	−0.03 (−0.28, 0.23)	0.84

Note: For each pesticide class, we compared the proportion of farmworker (FW) households that had pesticide concentrations greater than 4.63nmol/g dust to the proportion of non-farmworker (NFW) households above this level. This table provides the proportion of FW and NFW households with pesticides above this level, the difference between those two proportions, and the *p*-value associated with those differences based on a nonparametric proportions test. Additionally, for each pesticide class, we compared the proportion of FW households that had a decrease in pesticide concentrations between 2005 and 2011 with the proportion of NFW households that decreased in pesticide concentrations. NA, Pesticide excluded from longitudinal analysis due to >75% of households have pesticide levels <LOD for one of the study years. *$p < 0.05$; ***$p < 0.001$.
[a]Proportion Red refers to the proportion of households with pesticide levels above 4.63 nmol/g dust.

Table 3. Mixed effects analysis models and results (P-values).

Pesticides	Null vs. Time	Null vs. Time Fixed within Occupation	
		FW[a]	NFW[a]
Insecticides			
Organophosphate	0.003**	<0.001***	0.55
N-Methyl Carbamate	0.374	0.018*	0.402
Neonicotinoid	0.317	0.030*	0.402
Pyrethroid	0.836	0.189	0.385
Insect Growth Regulator	NA	NA	NA
Macrocyclic Lactone	NA	NA	NA
Synergist (Piperonyl Butoxide)	0.728	0.502	0.263
Organosulphite (Propargite)	NA	NA	NA
Fungicides			
Azole	0.373	0.78	0.30
Strobin	NA	NA	NA
Anilide (Boscalid)	<0.001***	<0.001***	0.001***
Herbicides			
Urea	0.158	0.674	0.163
Chlorophenoxy Acid/Ester	0.610	0.187	0.672
2,6-Dinitroaniline (Pendimethalin)	<0.001***	0.001***	0.010**
Pyridazinone	<0.001***	0.029*	0.003*
Microbiocides			
Chlorophenol (Triclosan)	<0.001***	<0.001***	<0.001***
Guanidine (DGH)	<0.001***	0.017*	<0.001***
Phenol (Na o-Phenylphenate)	<0.001***	0.015*	<0.001***

Note: Table 3 shows the models used for the mixed effects analysis of the cohort- and occupation-level changes in pesticide levels detected in household dust between 2005 and 2011. The models were compared as listed above, and the resultant *p*-values are reported. Variables included: FW (farmworkers), NFW (non-farmworkers), pest (pesticide), tm (time), house (household participating in the study). NA indicates pesticide excluded from longitudinal analysis due to >75 percent of households have pesticide levels less than LOD for one of the study years. *$p < 0.05$; **$p < 0.01$, ***$p < 0.001$.

[a]*P*-values represent the comparison of log(pest) ~ 1 + (1|house) and log(pest) ~ tm + (1|house) using analysis of variance (ANOVA).

synergist (Piperonyl Butoxide) detected in household dust were not statistically significantly different between 2005 and 2011 or between FW and NFW households and will not be discussed in detail.

Fungicides. Of the 87 pesticides detected in household dust, 9 were classified as fungicides. Half of these fungicides fell under the broad classification of azoles, and the remaining fungicides fell under the classes of strobins, anilides, and quinolines. All of the fungicides detected in household dust were registered for agricultural uses, and three of those were also registered for indoor residential uses (Table 1).

Anilide Fungicides (Boscalid) in Household Dust. In 2005, 3% of FW households showed boscalid levels greater than the 75th percentile of OPs (4.63 nmol/g dust, Figure 1), which is not significantly different from levels found in NFW households, none of which were greater than the 75th percentile of OPs (Table 2, $p = 0.29$). In 2011, 17% of FW households were above this level, which is significantly greater than the proportion of NFW households above this level, of which there were none (Table 2, $p = 0.03$). The levels of boscalid detected in household dust were significantly higher in 2011 than in 2005 for FWs (Table 3, $p < 0.001$), NFWs ($p = 0.001$), and for both occupational groups combined ($p < 0.001$). The proportion of FW households that had decreased boscalid levels between 2005 and 2011 was 27%, which was not significantly different from the 15% of NFW households that showed a decrease over the same period (Table 2, $p = 0.28$).

Other Fungicides Detected in Household Dust. In 2005, strobin levels detected in household dust were below the LOD for more than 75% of households (Table 1). As a result, strobins were excluded from longitudinal analyses and will not be discussed further. Additionally, there were no statistically significant differences between study years or FW and NFW households in the levels of azole fungicides detected in household dust; thus, the azoles will not be discussed in further detail (Tables 2 and 3).

Herbicides. Of the 87 pesticides detected in household dust, 7 were herbicides falling under the classes of urea, chlorophenoxy acid or ester, 2,6-dinitroaniline, and pyridazinone (Table 1). Five of these pesticides are registered for agricultural and other outdoor uses only (2,4-D, MCPA, Pendimethalin, Norflurazon, Pyridaben). Diuron is registered for use in aquariums and paints. MCPP is only registered for use on lawns and turf (U.S. EPA 2003, 2007).

2,6-Dinitroaniline herbicides (pendimethalin) in household dust. In both 2005 and 2011, there were no households with pendimethalin levels higher than the 75th percentile for OPs (4.63 nmol/g dust) in either occupational group (Table 2). The levels of pendimethalin in household dust were significantly greater in 2011 than in 2005 for FWs (Table 3, $p < 0.001$), NFWs ($p = 0.01$), and for both occupations combined ($p < 0.001$. Additionally, the proportion of FW households that had lower pendimethalin levels in 2011 than 2005 was 17%, which was not significantly different from the 33% of NFW households (Table 2, $p = 0.17$).

Pyridazinone herbicides in household dust. In both 2005 and 2011, there were no households with pyridazinone levels higher than the 75th percentile for OPs (4.63 nmol/g dust) in either occupational group (Table 2). The levels of pyridazinones were significantly lower in 2011 than in 2005 for FWs (Table 3, $p = 0.03$), NFWs ($p = 0.003$), and for both occupations combined ($p < 0.001$). Additionally, the proportion of FW households that had lower pyridazinone levels in 2011 than in 2005 was 68%, which was not significantly different from the 78% of NFW households (Table 2, $p = 0.45$).

Other herbicides detected in household dust. There were no statistically significant differences between study years or FW and NFW households for the ureas or the chlorophenoxy acids/esters detected in household dust (Tables 2 and 3).

Microbiocides. Three of the 87 pesticides detected in household dust were microbiocides (Table 1), although all three may have some fungicidal properties as well (U.S. EPA 2006b, 2008b, and 2005a). Triclosan (TCS), dodecylguanidine hydrochloride (DGH, a compound chemically similar to Dodine) and sodium ortho-phenylphenate have residential uses (U.S. EPA 2008b, 2005a, and 2006b). Dodine and sodium ortho-phenylphenate have a registered agricultural use (U.S. EPA 2005a and 2006b).

Chlorophenol microbiocides (triclosan) in household dust. In 2005, no significant difference was found between the proportion of FW and NFW households that had TCS levels greater than the

75th percentile for OPs (4.63 nmol/g dust (Table 2, $p = 0.48$). This finding was also the case in 2011 (Table 2, $p = 0.23$). The levels of TCS were significantly higher in 2011 than in 2005 for FWs, NFWs, and for both occupations combined (Table 3, $p < 0.001$). Additionally, only 14% of FW households had lower TCS levels in 2011 in comparison with levels in 2005, which was not significantly different from the 7% of NFW households (Table 2, $p = 0.44$).

Guanidine microbiocides (DGH) in household dust. In 2005, 13% of FW households showed DGH levels greater than the 75th percentile for OPs (4.63 nmol/g dust), which is not significantly different from the 3% of NFW households (Table 2, $p = 0.13$). In 2011, there was likewise no statistically significant difference in the proportion of households with pesticides above this level between FWs and NFWs (Table 2, $p = 0.82$). The levels of DGH were significantly higher in 2011 than in 2005 for FWs (Table 3, $p = 0.02$), NFWs ($p < 0.001$), and across both occupation groups combined ($p < 0.001$). Although DGH levels increased across the cohort in 2011, 36% of FW households showed a decrease in DGH levels between 2005 and 2011, which is significantly greater than the 11% of NFW households that showed decreased DGH levels (Table 2, $p = 0.04$).

Phenol microbiocides (Sodium Ortho-Phenylphenate (Na OPP)) in household dust. In 2005, no significant difference was found between the proportion of FW households that had Na OPP levels greater than the 75th percentile for OPs (4.63 nmol/g dust) and the proportion of NFW households with pesticides above this level (Table 2, $p = 0.31$). In 2011, there were no households with pesticide levels above 4.63 nmol/g dust, regardless of occupation. The levels of Na OPP were significantly lower in 2011 than in 2005 for FWs, NFWs, and both occupations combined (Table 3, $p = 0.02$, $p < 0.001$, and $p < 0.001$, respectively).

Change in Pesticide Usage between 2005 and 2011

Usage statistics of key agricultural pesticide classes for pome fruit were collected from the USDA-NASS chemical use statistics database (USDA-NASS 2011). Figure 2 illustrates the change in the amounts of the six main agricultural pesticide classes used in 2005 and 2011 in the State of Washington. Although the use of OPs, *N*-methyl carbamates, neonicotinoids, and pyridazinone declined from 2005 to 2011, the amount of anilides and 2, 6-dinitroanilines used in Washington increased in the same time period.

Discussion

Using Household Dust for Assessing Pesticide Exposure Profiles

In this study, we examined more than 80 pesticides across different classes in FW and NFW homes in 2005 and 2011. Although the concept of coexposures is of great interest for risk assessment and regulatory purposes, implementation is still challenging, especially for children's health which is affected by cumulative coexposures across the lifespan (NRC 2009; Patel 2017). Understanding pesticide coexposures in this way provides us with a glimpse into the pesticide exposome, defined in Shaffer et al. 2017.

Declines in Pesticide Concentrations in Dust in FW and NFW Households

In 2005, FWs had a significantly greater proportion of households with pesticide levels greater than 4.63 nmol/g dust than did NFWs for the organophosphates and the *N*-methyl carbamates, and in 2011, in comparison with NFWs, FWs had a greater proportion of households above this level for the organophosphates and boscalid (Table 2). NFW households did not have a significantly greater proportion of households above this level than did FWs for any pesticide class in either study year. This finding is consistent with our previous findings that support an occupational take-home pathway for OPs (Coronado et al. 2006; Curl et al. 2002; Thompson et al. 2014). Additionally, our previous studies have shown significant correlations between OP levels in household dust and urinary OP metabolites (Tamaro et al. 2018). The reduction in exposure among FWs might be related to educational outreach efforts in the region. However, it should be noted that, in this cohort, both FW and NFW adults and children showed higher urinary OP metabolite levels than did similar demographic groups in the corresponding NHANES years (Thompson et al. 2014). Thus, although NFW households did tend to have lower OP exposure than FW households did, those exposures were still greater than the exposure found for the average NHANES participant.

Most of the pesticides that were analyzed are used in agriculture, particularly in pome fruit production. Thus, it would be expected that FW households would have a higher proportion of households with higher levels of these pesticides detected in their dust than NFW households would have. Between 2005 and 2011, FW households overall experienced a significant decrease in concentrations of OPs, *N*-methyl carbamates, neonicotinoids, pyridazinones, and phenols (Na OPP) (Table 3). NFW households' levels decreased only in the latter two pesticide classes. A greater proportion of FW households showed a decrease in OPs, neonicotinoids, pyrethroids, and guanidines (DGH) than NFW households showed (Table 2). Taken together, this finding suggests that although FWs are more likely to have high (i.e., greater than the 75th percentile of OPs) pesticide levels in their household dust, they are also more likely to have decreased pesticide levels over time.

Trends in Agricultural Pesticide Usage and Household Dust of Farmworkers

In FW households, we observed a significant decrease in agricultural pesticide levels detected in dust between 2005 and 2011 for OPs, *N*-methyl carbamates, neonicotinoids, and pyridazinones, and an increase in pendimethalin and boscalid. We used the Quick Stats function of the USDA-NASS database (USDA-NASS 2011) to identify pesticide classes in Washington state used in apple production. We found that all of these pesticide classes are used in apple production in Washington state, and the changes in the use of these six pesticide classes align with the trends observed in household dust (Figure 2). Although the decline in OPs—both in household dust and in statewide use statistics—was expected over this time period due to the phase-out of azinphos-methyl, the concurrent decline of *N*-methyl carbamates and neonicotinoids (which are some of the recommended OP substitutes) is somewhat surprising (Doerr et al. 2012; Washington State University Extension 2008, 2011). We hypothesize that this decline may be due to the toxicity of neonicotinoids to bee populations and the cost of using pyrethroids. Nevertheless, the concurrence between pesticide levels in household dust in FW households and the statewide use of these pesticides suggests that FW households are directly affected by changes in agricultural pesticide usage. The concurrence consistent with our previous findings regarding the importance of the occupational take-home pathway (Coronado et al. 2006; Curl et al. 2002; Thompson et al. 2014). These findings further demonstrate the utility of using household dust as a way to characterize pesticide exposure profiles. The connection between agricultural pesticide usage and FW pesticide exposure suggests that household dust can be used to evaluate the effectiveness of pesticide

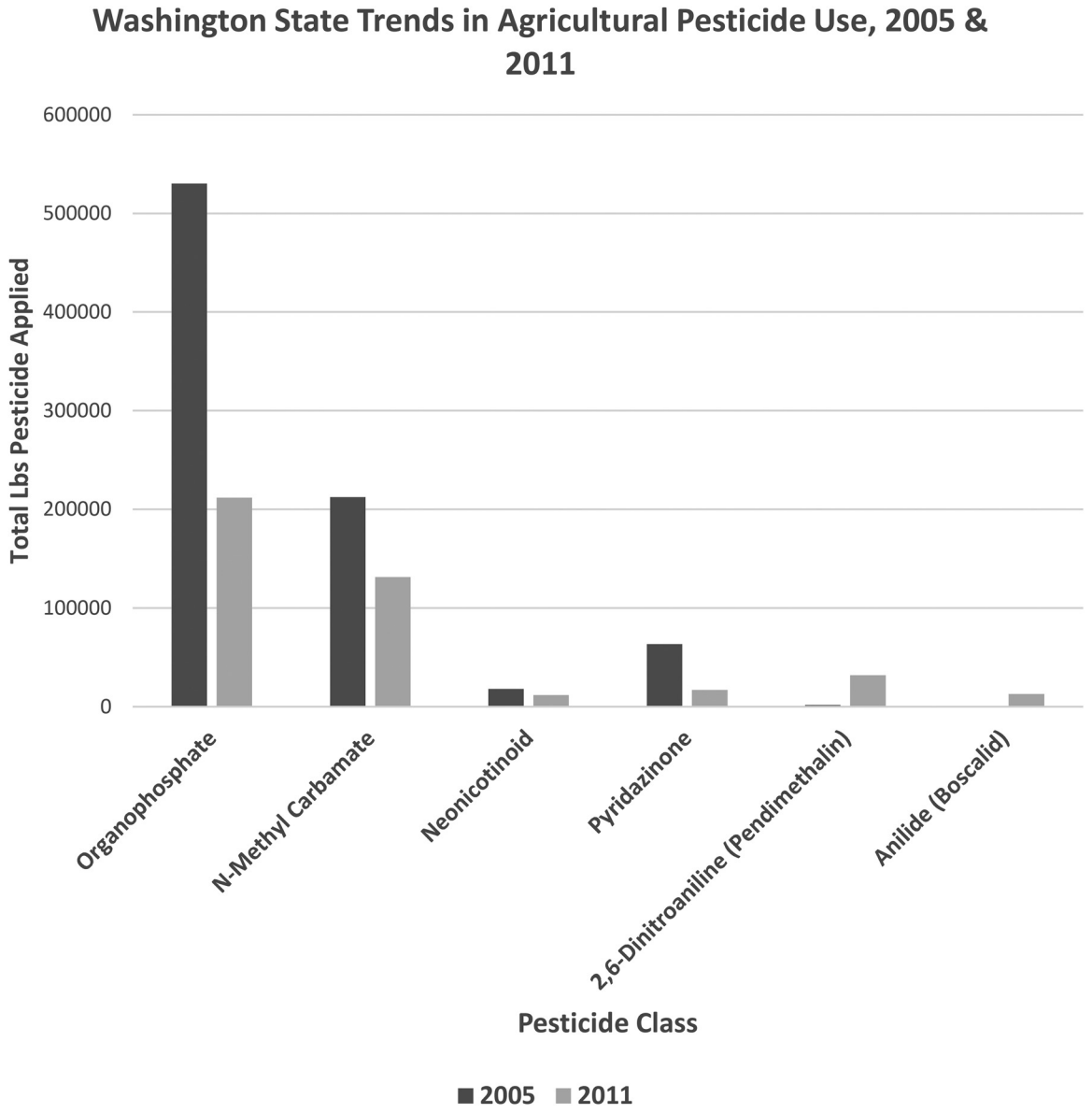

Figure 2. Changes in pesticide use between 2005 and 2011 for select agricultural pesticides for pome fruit. Pesticide use statistics (USDA-NASS 2011) for pome fruit in total pounds applied for six agricultural pesticides in 2005 (dark gray) and 2011 (light gray).

regulations and link pesticide use with the potential for direct population exposures.

The Complex Role of Residential Pesticide Use in Pesticide Exposure

Because this cohort is located within an agricultural region, the majority of the pesticides analyzed have agricultural uses (Table 1). However, several agricultural pesticides, such as permethrin, malathion, and carbaryl, are also used in indoor residential settings (Table 1; Atwood and Paisley-Jones 2017; Grube et al. 2011; Guha et al. 2013), which makes separating agricultural vs. residential sources difficult for these dual-use pesticides. Furthermore, the use of residential pesticides is not documented as agricultural pesticide usage is, and so sales estimates of active ingredients are the best available approximation (Atwood and Paisley-Jones 2017; Grube et al. 2011). Additionally, some pesticides registered for use by the U.S. EPA, such as the antimicrobial triclosan (TCS), are incorporated into consumer products (U.S. EPA 2008b), which makes tracking usage even more difficult. Thus, without detailed household data, fully linking residential pesticide use to household dust concentrations is challenging.

In our cohort, we were able to address these questions more directly through the collection of household level data across time by comparing exposures in 2005 with exposures six years later in 2011. We observed interesting trends in residential pesticide use, particularly among the microbiocides. TCS is a phenol compound and is registered for use as a material preservative in a number of consumer products and has no direct uses in food (U.S. EPA 2008b). Na OPP is likewise used as a materials preservative but is also registered for use in carpet, bathroom, and other cleaners (U.S. EPA 2006b). TCS levels in household dust increased significantly between 2005 and 2011 across the entire cohort and within each occupational group. This finding is particularly notable given that urinary TCS in the U.S. population (according to the NHANES) decreased from a geometric mean concentration of 18.5 μg/L to 11.8 μg/L. Among Mexican Americans, a group which more closely represents the ethnicity of our cohort, the decrease is even

larger: from 26.7 μg/L in 2005/06 to 12.6 μg/L in 2011/12 (CDC 2017). Although household dust and urine are not equivalent media, the fact that we observe the trend opposite to that found in NHANES is notable. One interpretation of these data is that the presence of TCS in those households increased as well. It is possible that the integration of TCS in consumer products (e.g., toys, toothbrushes, textiles, garbage bags, etc.) increased over this period, and the degradation of these products led to a higher TCS signal in household dust than found in urine, which could explain the discrepancy between the change observed in NHANES and the change observed in the present study. Indeed, this interpretation is consistent with reported increases in U.S. imports of TCS and increasing antimicrobial product markets over this period (Han et al. 2016), particularly with the increased promotion of antimicrobial products due to flu outbreaks such as the H1N1 swine flu in 2009 (U.S. EPA 2009).

As with observations of increases in TCS, we observed a significant increase in DGH levels among both FW and NFW households (Table 3). It is worth noting that the chemical salt DGH, dodine, is an agricultural fungicide registered for use in pome fruit production. However, its use in Washington State pome fruit production is negligible (i.e., no available USDA chemical use data), and the use of dodine in agriculture declined between 2005 and 2011 (Baker and Stone 2015; Thelin and Stone 2013; USDA-NASS 2011). Additionally, DGH has registered uses that are similar to those of TCS, such as its uses in papers that come into contact with food, paint, and diapers (U.S. EPA 2005a), which is supported by the significant increase in household dust levels observed in our cohort for both of these microbiocides. Thus, although dodine may be present, we most likely detected r DGH in household dust.

In contrast to the trend toward increasing levels of TCS and DGH in household dust, Na OPP, the third microbiocide detected, was lower in 2011 than in 2005. The increases of TCS and DGH in our cohort are particularly notable because although we observe a decrease in many agricultural pesticides in household dust, including the only microbiocide we examined that has registered agricultural uses, we also observe a concurrent increase in pesticides with potential uses in consumer products. The reasons for these opposing trends are uncertain. Although agricultural pesticide use has declined overall, pesticide exposures still occur through other (i.e., residential) routes, some of which may be increasing in magnitude. From a regulatory perspective, it is important to evaluate these multiple and varied exposures collectively to fully capture the spectrum of pesticide exposure. Although our exposure analysis alone does not provide information about the health impact of these multiple exposures on children's health, the complexity of the changes in pesticide exposures highlights a need for longitudinal life-stage monitoring. Also, because we found these exposures to be dynamic, we stress the continuing need to analyze multiple coexposures and time points.

The strengths of this study include the assessment of multiple pesticide coexposures with shared mechanisms of toxicity. Understanding how pesticide coexposures shift over time is important to predicting and preventing potential health effects in children living in an agricultural region. Although we were able to characterize over 80 pesticides, we were unable to include some pesticides due to lack of available standards or analytical procedures. Additionally, some pesticides were excluded due to a high proportion of samples occurring below the LOD. Longitudinal analyses are also complicated by changes in LOD and price per analysis. We optimized our funding so we could characterize the breadth of pesticide exposure needed to identify trends over time. However, this method meant that the LOD increased for some analytes over time (Table 1). We managed this LOD increase over time by examining our data by quartiles for evaluation. Another limitation of this study is the inability to fully relate our observations with pesticide use statistics. In California, the state requires detailed reporting of pesticide use (California Department of Pesticide Regulation, 2018). However, Washington state data were available only by crop and year; thus, understanding how these observations relate to seasonal use rates was not possible (USDA-NASS 2018). Additionally, it has been reported that bifenthrin (Washington State Department of Health, 2013) was the major pyrethroid used to replace the OPs. We were unable to include bifenthrin because laboratory methods were not available during our analysis period and use statistics had not yet been reported. Further developing this link would increase the regulatory implications of this study.

Conclusion

This study identified longitudinal pesticide exposure profiles in household dust in a children's agricultural cohort. We demonstrate a clear link between reported agricultural chemical use and the levels of pesticides detected in the household dust of FWs, which supports the utility of using household dust as an indication of the pesticide coexposures. In 2005, FWs showed higher concentrations of organophosphates and *N*-methyl carbamates, two major agricultural pesticides used in apple crop production, in their household dust than did NFWs. In 2011, FWs had higher levels of organophosphates and the anilide fungicide boscalid than NFW households did, further demonstrating the link between agricultural pesticide use and the levels detected in the dust of FW households. Additionally, this work highlights the importance of detailed longitudinal exposure monitoring, especially for the purposes of capturing the changes in both agricultural and residential pesticide use, which can have different trends and public data availability. The observed increase in DGH and TCS suggests that the presence of microbiocide-containing consumer products increased over the study period. This hypothesis is difficult to verify, however, because residential pesticide use is not as well monitored as agricultural pesticide use.

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