Investigating Seasonal and Occupational Trends of Five Organophosphate Pesticides in House Dust in the Lower Yakima Valley, WA

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ABSTRACT

Organophosphate pesticides (OPs) are a class of insecticides and pesticides that have become widely used. Adult exposure routes and health effects of OPs have been researched in detail. However, data characterizing chronic low-dose childhood exposures to pesticide sources, such as those that may occur in indirect outdoor and indoor environmental pathways, is limited. This is especially important as childhood age-specific behaviors may predispose young children to chronic and/or increased episodic pesticide exposures. This research characterizes home environment dust as a potential source for indoor pesticide exposure in agricultural communities in the Lower Yakima Valley, located in Eastern Washington. Over the last 13 years, the University of Washington Center for Child Environmental Health Risks Research (CHC) has followed a community-based participatory research (CBPR) strategy in this area to assess and reduce pesticide exposure among children of Hispanic Farmworkers. This research uses dust samples collected in the CHC pesticide research project to identify the episodic nature of OP exposures. The CHC study collected house and vehicle dust samples, urine, saliva, and blood samples from participants across three agricultural seasons throughout the year with known OP application schemes and farmworker work tasks. House dust levels of 5 OPs in this study were found to vary throughout the year based on occupation with farmworkers consistently presenting higher concentrations of OPs. The largest seasonal differences observed occurred from the Harvest-to-Non-Spray and Thinning-to-Non-Spray seasons for Azinphos methyl (AZ), Phosmet (PH), Malathion (ML), Diazinon (DZ), and Chlorpyrifos (CP). House dust levels of OPs were influenced by householder work status in pome and orchard fruit crops, agricultural season, use classification of

pesticides, and potential for contact with sprayed fruit and orchard trees dictated by work status. As levels of AZ, PH, ML, DZ and CP in house dust were episodic depending on season, they present episodic exposure risk to populations residing indoors, especially children.

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Chapter 1: Background

Background

Pesticides are widely used across the United States in agriculture. Organophosphate pesticides (OPs) are a class of non-specific systemic insecticides and pesticides that became widely used after organochlorine pesticides were banned in the late 1970's. OPs are used in a variety of settings as they are relatively inexpensive, less persistent in the environment, and less susceptible to pest resistance. Although major uses are in crop and livestock agriculture, OPs are also used in civil pest management programs and public health pest eradication campaigns for vector-borne diseases. Humans may be exposed to OPs through food, drinking water, in and around places of work, residence, and school (USEPA 2002).

The general structure of all OPs is similar, consisting of a central phosphate moiety which is dialkyl substituted, commonly with dimethyl or diethyl alkyl groups, and an organic group which is specific to each pesticide (Mileson, Chambers et al. 1998). Organophosphates are typically highly lipid-soluble, facilitating absorption through the skin, in oral mucous membranes, in gastrointestinal systems, and via respiratory routes (Kamanyire and Karalliedde 2004). Metabolism of organophosphate pesticides occurs via hydrolysis and detoxification of the OP; forming a pesticide-specific metabolite and dialkylthionate metabolites. In certain instances enzymatic conversion and activation to an oxon-intermediate form of the OP occurs, presenting increased toxicity when compared to the parent compound for a variety of OPs including chlorpyrifos, parathion, and diazinon among others. After conversion to an oxon-form, enzymatic or spontaneous hydrolysis occurs to form a dialkylphosphate (DAPs) metabolite and pesticide-specific moiety. Ultimately glucuronidation or sulfonation of the compounds occurs followed by excretion in the urine (Wessels, Barr et al. 2003).

The United States Environmental Protection Agency (EPA) currently assesses organophosphate pesticides as a class of compounds whose exposure is considered with dose additivity, under the rationale that OPs share a common mechanism of action (USEPA 2006). This includes the binding, phosphorylation, and inhibition of acetylcholinesterase (AChE), a critical enzyme of the neurotransmitter acetylcholine. Upon AChE inhibition, acetylcholine accumulates in muscarinic sites, nicotinic sites, and within the central nervous system; causing cholinergic toxicity as continuous stimulation of cholinergic receptors throughout the central and peripheral nervous systems ensues. OP exposures are commonly assessed by measuring parent compound in the blood or by measuring organophosphate metabolites in urine; these metabolites can be non-specific DAPs or OP-specific. Measurement of inhibition of plasma, red blood cell, and brain cholinesterase activity are utilized as common mechanistic endpoints for diagnosing and assessing OP toxicity.

Adult exposure routes and health effects of organophosphate pesticides have been researched in detail. This examination of acute and chronic exposures to organophosphate pesticides has been in part facilitated by occupational uses and exposures in agriculture (Magnotti, Dowling et al. 1988; Simcox, Camp et al. 1999). As the majority of OP applications are performed for agricultural purposes, agricultural workers experience higher exposures than the general population (Lee, Burnett et al. 2002; Muniz, McCauley et al. 2008; Atherton, Williams et al. 2009). Common symptoms of OP exposure include headache, nausea, dizziness, and hypersecretion (which includes: sweating, salivation, and lacrimation). Muscle twitching, weakness, tremors, incoordination, vomiting, abdominal cramps, and diarrhea can all also signal OP poisoning. Adults may also experience altered motor activity and abnormal neurobehavioral effects up to several weeks after exposure. Some suffer the consequences of OP-induced delayed polyneuropathy, a well characterized syndrome that occurs after axonal death resulting from OP inhibition of a neural enzyme called neuropathy target esterase (Keifer and Mahurin 1997; Salvi, Lara et al. 2003; Rothlein, Rohlman et al. 2006). The syndrome involves sensory abnormalities, muscle cramps and weakness, and potentially paralysis, primarily in the legs; for some individuals this syndrome is irreversible. In an effort to reduce pesticide exposures and subsequent health effects, restricted reentry periods and the use of personal protective equipment have been mandated in state-wide and federal regulatory efforts (Fenske, Curl et al. 2003; Vitali, Protano et al. 2009).

Although multiple biomonitoring studies have suggested that children can be more widely exposed to pesticides including OPs, research efforts investigating childhood acute and chronic low level exposures to OPs are limited (Aprea, Strambi et al. 2000; Fenske, Lu et al. 2000; Adgate, Barr et al. 2001; Bradman, Barr et al. 2003; Barr, Allen et al. 2005), as are studies that distinguished between preformed residuals of the OPs from OP parent components (Zhang, Driver et al. 2008; Engel, Wetmur et al. 2011). Extrapolation from studies in adults is also difficult as children are believed to face OP exposures at levels much lower than those encountered occupationally, exposures that occur intermittently, and exposures that occur at variable intensity (Wessels, Barr et al. 2003).

Thus far, increased risk for development of neurologic impairments, some cancers, birth defects and abnormal reflexes have been reported to be associated with childhood pesticide exposures (Guillette, Meza et al. 1998; Blain 2001; Kirkhorn and Schenker 2002; Rohlman, Arcury et al. 2005; Mills and Yang 2007). Other studies include several longitudinal birth cohorts across the United States with intent to research pre- and post-natal exposures to pesticides, including OPs, where preliminary results display a pattern of early cognitive and behavioral deficits in children related to OP exposure in both agricultural and urban communities utilizing different biomarkers of exposure (Bouchard, Chevrier et al. 2011; Engel, Wetmur et al. 2011; Rauh, Arunajadai et al. 2011).

OP exposures in children living in agricultural areas or with farmworker families have been the topic of several studies (Simcox, Fenske et al. 1995; Loewenherz, Fenske et al. 1997; Lu, Fenske et al. 2000; O'Rourke, Lizardi et al. 2000; Koch, Hardt et al. 2001; McCauley, Beltran et al. 2001; Quandt, Arcury et al. 2004). Research thus far has suggested that farmworker children are particularly vulnerable to pesticide exposure as they can experience increased exposures via multiple pathways including pesticide drift from nearby sprayed fields and the take-home pathway, where occupational pesticides are tracked from work-to-home via parents’ vehicles and clothing (Fenske 1997; Lu, Fenske et al. 2000; Curl, Fenske et al. 2002; Thompson, Coronado et al. 2003).

While the body of documented childhood health effects grows, data do lack in characterizing chronic low-dose childhood exposures to pesticide sources such as those that may occur in indirect outdoor and indoor environmental pathways. This is especially disconcerting as childhood age-specific behaviors may predispose young children to chronic and/or increased episodic pesticide exposures. These behaviors include: increased proximity and activity on surfaces covered in dust (potential for dermal uptake), exploration of the environment with their mouth's (non-dietary and hand-to-mouth ingestion), dietary patterns allowing for greater pesticide exposure (consumption of treated fruits, vegetables, juice), and the childhood condition Pica (increased geophagy) (Faustman, Silbernagel et al. 2000; Fenske, Lu et al. 2002; Lambert, Lasarev et al. 2005; Rao, Quandt et al. 2006). Further complicating these unique behavior considerations are important considerations and risks associated with childhood developmental and toxicodynamic processes. Specifically, children have greater surface area-to-body weight ratios, higher circulatory flow rates (distribution faster throughout the body), immature metabolism and elimination systems (altering exposure time and efficacy in clearance) and dynamic liver detoxifying protein profiles throughout development (Faustman, Silbernagel et al. 2000; Fenske, Lu et al. 2002; Lambert, Lasarev et al. 2005; Rao, Quandt et al. 2006).

Recently, research efforts have begun focusing on characterizing indoor exposures of children to environmental chemicals, including pesticides which may persist in indoor areas that are considered childhood microenvironments (Lewis, Fortmann et al. 1994). These areas have a lack of degradative forces such as light, temperature, water, and microorganisms. Researchers have used house dust as an environmental biomarker for indoor pesticide contamination, indoor pesticide loading, and as a potential vehicle for childhood pesticide exposure (Lu, Fenske et al. 2000; Fenske, Lu et al. 2002; McCauley, Michaels et al. 2003; Shalat, Donnelly et al. 2003; Coronado, Thompson et al. 2004; Curwin, Hein et al. 2005).

As dust is a ubiquitous environmental material, it serves as an important route of toxicant exposure especially as it has the potential to act as a sink for semi-volatile organic compounds and particle-bound matter (Butte and Heinzow 2002; Layton and Beamer 2009). Compounds banned long ago are at detectable levels in house dust, providing a potential significant source of exposure for the general public and especially children (Butte and Heinzow 2002). As flecks and bits of dust can behave differently and present different levels of health risk, the route of exposure, size of the particulate, and composition of the particulate are critical characteristics to be identified (Lioy, Yiin et al. 1998). Compounds adsorbed to house dust may enter the human body by: inhalation of suspended and re-suspended particles, through non-dietary ingestion of dust, through ingestion of particles adhering to food surfaces in the homes, and on the skin by absorption through the skin (Butte and Heinzow 2002). Numerous research evidence exists identifying mechanisms employed in public health regimes that can reduce dust loading, exposure, and subsequent onset of health sequelae (Lioy, Yiin et al. 1998; Galke, Clark et al. 2001; Thompson, Coronado et al. 2003).

Objective

The purpose of this research is to characterize home environment dust as a potential source for indoor pesticide exposure in agricultural communities by utilizing the unique community-based participatory research (CBPR) strategy and complimentary environmental sampling and health effect assessments developed by the University of Washington Center for Child Environmental Health Risks Research (CHC). More specifically, this research aims to make use of dust samples collected as part of the CHC pesticide research project to inform of the episodic nature of OP exposures that occur on a local scale in the predominantly Hispanic agricultural Yakima Valley in Eastern Washington.

Setting

The state of Washington value of apples in 2005 totaled $1.23 billion dollars alongside an estimated $142.8 million dollars in revenue from pear production (USDA 2006). Washington is second only to California in diversity of crops grown, with 230 different varieties represented. United States Department of Agriculture statistics indicate that WA ranks first in the US for production of apples, at 60% of the country's total and also leads US crop production for a variety of other fruit crops including but not limited to: raspberries (92%), sweet cherries (50%), concord grapes (43%), pears (48%), and all hops (80%)(USDA 2011). Use of insecticides reported by the CropLife Foundation revealed that in 2002, WA ranked 3rd in the country in total use of insecticides annually at approximately 11,222,396 lbs. active ingredient applied (Gianessi and Reigner 2006).

The Lower Yakima Valley, located in Eastern Washington, is a rich agricultural region comprised of many small agricultural communities. It is estimated that approximately 50,000 individuals work in agriculture in the region (Coronado, Holte et al. 2011). According to the 2010 Census data, the Lower Yakima Valley is predominantly Hispanic (65% of the population, see Appendix), and the Valley, along with Adams and Franklin counties, has one of the greatest representations of Hispanics across Washington State. In line with the demographic distribution in the Lower Yakima Valley, most of the agricultural work is done by farmworkers who are of Hispanic origin (Coronado, Holte et al. 2011).

Pesticides used throughout the Yakima Valley include OP insecticides such as Azinphos methyl (AZ), Phosmet (PH), Diazinon (DZ), Malathion (ML), and Chlorpyrifos (CP) among others (Coronado, Holte et al. 2011). Seasonal variation in pesticide use is evident, with heaviest applications of Restricted Use Pesticides such as AZ in the Thinning season, reduced applications in the Harvest season, and no applications in the dormant Non-Spray season. Although general residential uses of OP insecticides have declined in recent years after several respective phase-outs of CP, DZ and AZ, their uses are still permitted for defined agricultural purposes in the Valley (USEPA 2002; USEPA 2004; USEPA 2006).

Codling moth (*Cydia pomonella*) is the main agricultural pest in the Yakima Valley and has the greatest potential for causing damage of any apple pest (Caprile and Vossen 2011). Population levels of the moth are influenced by orchard conditions, local air and soil temperatures, wind and humidity (Coronado, Thompson et al. 2004). The moth has anywhere from two to four generations each season and as such, pesticide use each year is determined by infiltration levels of the pest. Organophosphate pesticides with documented activity against codling moth include Azinphos methyl (Caprile and Vossen 2011).

Data for Washington from the Agricultural Chemical Use Database show that in 2005, greater than 196,000 lbs of AZ and greater than 87,000 lbs of PH were used on apple crops alone (NASS 2005). Other organophosphate pesticides with use on apple crops in 2005 include CP (187,000 lbs) and DZ (13,000 lbs). Other major WA agricultural fruit uses of organophosphate pesticides, such as those in pears and sweet cherries, that were considered in this study can be found in Table 1.

Chapter 2: Introduction

Over the last 13 years, the University of Washington Center for Child Environmental Health Risk Research (CHC) has followed a community-based participatory research (CBPR) strategy in the Lower Yakima Valley, WA to assess and reduce pesticide exposure among children of Hispanic Farmworkers (Thompson, Coronado et al. 2001; Thompson, Coronado et al. 2003; Thompson, Griffith et al. 2013). This cohort is part of a larger longitudinal study lead by Dr. Beti Thompson within the Cancer Prevention Program at the Fred Hutchinson Cancer Research Center. One of the aims of the CHC study is to identify the pathways to which children may be exposed to pesticides. To this end, CHC collected numerous biological and environmental samples for subsequent analysis and characterization of exposure pathways. A schematic of this population and recruitment description can be found in Thompson (2003)(Thompson, Coronado et al. 2003).

Community interest expressed by CHC participants and the Community Advisory Board (CAB) formed as part of the CBPR strategy in 1999 and 2002 led to the examination agricultural contaminants such as organophosphate pesticides (OPs) in dust from vehicles and houses for selected communities and labor camps across the Yakima Valley (Curl, Fenske et al. 2002; Coronado, Thompson et al. 2004). The first wave of research efforts focused on using dialkylphosphate (DAP) urinary metabolites to monitor pesticide exposure in adult-child farmworker and non-farmworker pairs (Thompson, Coronado et al. 2001; Curl, Fenske et al. 2002). Biomonitoring data were collected with extensive concomitant household surveys that investigated crop, type of work, the use of personal protective equipment at work, workplace exposures to pesticides, and personal hygiene practices during and after work. Results from these studies determined that farmworkers who worked within pome fruit crops (e.g. apples and pears) had higher exposures to select OPs than workers who did not work in these crops (Coronado, Vigoren et al. 2006; Coronado, Vigoren et al. 2009; Thompson, Griffith et al. 2013).

In line with these findings and input from our CAB, in 2005-2006 the CHC began a cross-sectional follow up study aiming to compare household dust differences of pesticide profiles between farmworker households with an adult who worked in pome fruit crops, and non-farmworker households with an adult who worked in other agricultural, non-orchard settings within the Yakima Valley. An occupationally identified adult and referent child aged 2–6 years were enrolled in this study and represented a cohort of 99 FW and 95 NFW households recruited from our larger cohort (Thompson, Coronado et al. 2003; Locke, Coronado et al. 2009). The term farmworker (FW) hereafter describes enrolled agricultural workers classified by self-reported work status in orchard fruit crops, with a majority of orchard work in pome fruits. The term non-farmworker (NFW) hereafter describes individuals who had reported no work activity in an orchard fruit crop. The worker classification statuses in the study population were obtained during survey procedures occurring in each of the Thinning, Harvest, and Non-Spray seasons.

The CHC study collected house and vehicle dust samples, urine, saliva, and blood samples from participants across three agricultural seasons throughout the year, Thinning, Harvest, and Non-Spray, with known OP application schemes and farmworker work tasks. This study hypothesizes that four factors influenced levels of OP pesticide concentrations in house dust and utilized a seasonal sampling campaign to investigate. The factors postulated to be major determinants of house dust pesticide burden included work status as defined by activity in pome fruit orchards, the use classification of OP pesticides (Restricted vs General) used in fruit crops, the seasonal time of agricultural work activities, and the potential for pesticide contact via sprayed fruit and fruit trees.

Chapter 3: Materials and Methods

Sample Collection

Trained field staff from the Yakima Valley collected household dust samples during agriculturally seasonal periods of the year, spanning May 2005–Feb 2006. Collection was completed with a Nilfisk vacuum cleaner unit (GS-80; Nilfisk of America, Malvern, PA). A, fresh polyliner vacuum bag, cleaned hose and wand were used for each household.

In each household, adult participants identified the area where the child played most frequently and dust was collected from there. The area vacuumed depended on the floor surface type. A standardized collection scheme employed the use of (0.5m x 0.5m) square templates. Generally, plush carpet had 4 templates vacuumed, thin/flat carpet had 6 templates vacuumed, and hard/smooth floors had 8 templates vacuumed. Field staff recorded areas of sample collection, housing type, and number of templates vacuumed in each collection event.

After collection, samples were stored at -10°C in the field laboratory prior to transport to the University of Washington. Upon receipt, samples were again stored at -10°C until analysis occurred in the University of Washington Environmental Health Laboratory.

Dust Analysis

Sample preparation and analytic methods can be found described elsewhere (Curl, Fenske et al. 2002; Anastassiades, Lehotay et al. 2003; Alder, Greulich et al. 2006).

Briefly, samples were transferred from vacuum cleaner bags to 150-µm sieves (VWR, West Chester, PA) and sieved for 10 minutes in a sieve shaker (Model RX-24; WS Tyler Inc, Mentor, OH). To complete analysis, at least 0.2 µg of fine sieved dust was needed.

The first chemical analysis (2007) determined concentration levels of six organophosphate pesticides in house dust of FW and NFW households during the Thinning season (May–July 2005). Analysis was completed via an acetonitrile extraction with liquid-liquid partitioning and quantitative and confirmatory analysis with liquid chromatography tandem mass spectrometry using electrospray ionization. Samples not analyzed were labeled and stored at -10°C at the University of Washington Environmental Health Laboratory until further analysis. The preliminary Thinning season results on AZ and PH from this portion of the study can be found elsewhere (Coronado, Holte et al. 2011).

The second analysis (2011), determined concentration levels of an expanded set of 11 OPs in all remaining house dust samples across the three seasons (Thinning, Harvest, and Non-Spray). House dust samples were analyzed following the technique described above, and also included analysis for 18 other insecticides, herbicides, and fungicides (see Appendix). A reanalysis of a fraction of the original samples provided a comparative basis for subsequent analysis.

Statistical Analysis

All statistical analyses were performed in Microsoft Excel® (2010) and the statistical package STATA-12.0 (StataCorp. 2011. College Station, TX).

To deal with values below the limit of detection (LOD), the lowest measurable value that can be distinguished from background, the following procedure was used. Values falling below the LOD for any analysis were substituted with the value of the LOD, data were then log-transformed and the normal distribution quantiles were calculated based upon the ranks of the data. Data falling below the LOD were then excluded prior to using the least squares regression function LINEST to fit the log-transformed data to the normal distribution quantiles in Microsoft Excel to calculate the geometric mean (GM) and geometric standard deviation (GSD) of each pesticide by season and occupation. Quantile-quantile plots (not shown) illustrate that concentration of pesticide residues in house dust were normally distributed after log-transformation. Concentrations for the house dust samples are presented in units of nanograms of pesticide per gram of dust (ng/g). Significant differences between seasons and between FW and NFW were calculated with the non-parametric Wilcoxon test (Armitage, Berry et al. 2008).

This thesis emphasizes and reports on AZ, PH, ML, DZ and CP as they were the most frequently detected OPs in house dust across the three agricultural seasons. Pesticide-specific LOD can be found in the Appendix. Summary statistics including detection frequency (DF), GM, GSD, and 95% confidence interval concentrations for select OP pesticides are reported.

Chapter 4: Results

Sample Collection

A total of 320 samples of house dust were collected and analyzed for agricultural contaminants including 11 OPs and 18 other insecticides, herbicides, and fungicides. Samples were collected across known agricultural seasons in the Lower Yakima Valley from May 2005–Feb 2006. The agricultural seasons of collection (with house dust sample total) were the Thinning season from May–July (n=117), the Harvest season from Sep–Oct (n=117), and the Non-Spray season from Dec–Feb (n=86).

FW households were classified by the adult participant's self-reported work activity in an orchard fruit crop, primarily in pome fruit crop, during each agricultural season. NFW were individuals reporting no work activity in an orchard fruit crop each season. An expanded description of demographic characteristics of participants in this study can be found elsewhere (Thompson, Griffith et al. 2013). Briefly, FW households comprised 44, 48, and 47 percent of house dust samples collected in each of the Thinning, Harvest, and Non-Spray seasons, respectively.

Pesticide Use and Agricultural Crop Seasons

Table 1 illustrates agricultural fruit crop uses and classification information for OP pesticides detected in house dust in the Lower Yakima Valley, WA and includes information on registered pesticide uses during 2005–2006.

As shown, Restricted Use Pesticides in this study with permitted uses only in plant and livestock agriculture included AZ, DZ and CP. General Use Pesticides with agricultural as well as residential and civil uses include PH and ML. The heaviest reported uses of OPs in orchard fruit crop in 2005–2006 were uses of AZ, CP and PH respectively. ML and DZ had least use in lbs of application in orchard fruit crops.

Table 1. Washington's Agricultural Fruit Chemical Usage: Agricultural Crop Use and Classification Information of 5 Commonly Detected OP Pesticides in House dust in the Lower Yakima Valley, WA

Pesticide Common Name	Trade Name[a]	CAS. Registration No.	WA's Agricultural Fruit Chemical Usage 2005[b]	Common Application by approximate quantity of use[c,1,2]	Other Uses[3,4]	Chronic Oral RfD (mg/kg/B.Wt/day)	EPA Cancer Class (2006)
Azinphos methyl	Azinphos-M 2 EC, Azinphos-M 50 WP, Azinphosmethyl 50W, Guthion 2L, Guthion 35% WP, Guthion 3F, Guthion Solupak 50%, Sniper 50W	86-50-0	231.9	Apple(196.4), Pear(18.7), Cherry(16.8)		0.00149	Not likely to be carcinogenic to humans
Phosmet	Imidan 12.5%, Imidan 50-WSB, Imidan 70 WSB (WP)	732-11-6	106.2	Apple(87.1), Pear(14.9), Grape(4.2)	^, #	0.02	Suggestive evidence of carcinogenicity, but not sufficient to assess human carcinogenic potential
Malathion	Agway Fruit Tree Spray, Atrapa 5E, Cythion ULV (9.33lbs), Fyfanon, Fyfanon ULV 9.9lbs (96.5%)	121-75-5	18.4	Apple(4), Sweet Cherry(8.8), Raspberry(5.6)	^, #	0.07	Suggestive evidence of carcinogenicity, but not sufficient to assess human carcinogenic potential
Diazinon	Diazinon 4 Spray, Diazinon 40WP, Diazinon 4E, Diazinon 50W, Diazinon AG500 (4E), Diazinon AG600 WBC, Spectracide 25	333-41-5	26.4	Apple(13.3), Pear(0.1), Sweet Cherry(5.6), Raspberry(7.3)	^	0.0002	Not likely to be carcinogenic to humans
Chlorpyrifos	Chlorpyrifos 4E AG, Dursban 1% Granules, Govern 4E, Lorsban 15G, Lorsban 4E, Lorsban 4E (USE-1069), Lorsban 50W, Lorsban 75WG, Nufos 4E, Warhawk, Whirlwind, Yuma 4E	2921-88-2	226.4	Apple(186.7), Pear(13.2), Sweet Cherry(26.5)	^, *	0.003	Group-E evidence of non-carcinogenicity for humans

[a] Chemical usage data for selected Washington fruit crops (NASS, 2005). [b] Summed Annual select applications in Washington State from common application by approx. quantity of use (1,000s lbs) (NASS, 2005). [c] A sample of the most common WA fruit crop uses of pesticide, uses are listed in (1,000s lbs) of use (NASS, 2005; USEPA 2006; NASS, 2007). [1] Washington's Agricultural Fruit Chemical Usage, (NASS, 2005). [2] Washington Department of Agriculture State Use Summary, 2004. [3] Agriculturla Chemical Usage Diary Cattle and Dairy Facilities Summary (USDA, 2006), (NASS, 2007). [*] Livestock Dairy Agriculture use in Washington. [^] Livestock Dairy Agriculture use in US. [4] Environmentla Protection Agency Registration Eligibility Documents: Malathion RED (USEPA, 2006) & Phosmet RED (USEPA, 2006). [#] EPA RED Permitted Residential Uses.

Fruit Planting and Harvest Intervals

Figure 1 illustrates fruit crop planting and harvest intervals for the state of Washington (2005) along with time of CHC sample collection during each agricultural season and corresponding work tasks for FW in each season (NASS 2005).

Generally, pome fruit crops illustrated in Figure 1 have longer and later seasons than non-pome fruit crops with sweet cherries and raspberries having the shortest seasons, while apples and winter pears have the longest seasons. Pome fruit crops also have the more diverse and numerous OP usage when compared to non-pome fruit crops on a local (WA use) and national scale (National Ranking) (NASS 2005).

Work tasks during the Thinning season include farmworker removal of small buds and shoots of tree limbs by hand with a Restricted Entry Interval (REI) for worker protection during this activity and season ranging from 7–19 days depending on crop sprayed, pesticide applied (AZ, PH, ML, CP or DZ), and rate of application. Azinphos methyl, for example, has REI's ranging from 7 days for blueberries, to 14 days for apples and pears, to 19 days for sweet and tart cherries at a 50% wettable powder formulation (USEPA 2006). Similarly, Pre Harvest Intervals (PHI), during the Harvest season when workers harvest fruit by hand can last up to 21 days again depending on crop, pesticide, and formulation. Azinphos methyl PHI's range from 14 days at rates less than 1 lb active ingredient per acre applied to 21 days at rates greater than or equal to 1 lb active ingredient per acre applied (USEPA 2006).

Figure 1. Fruit Planting and Harvest Interval with Agricultural Seasons and Known Work tasks for Study Participants Throughout the Year (2005)

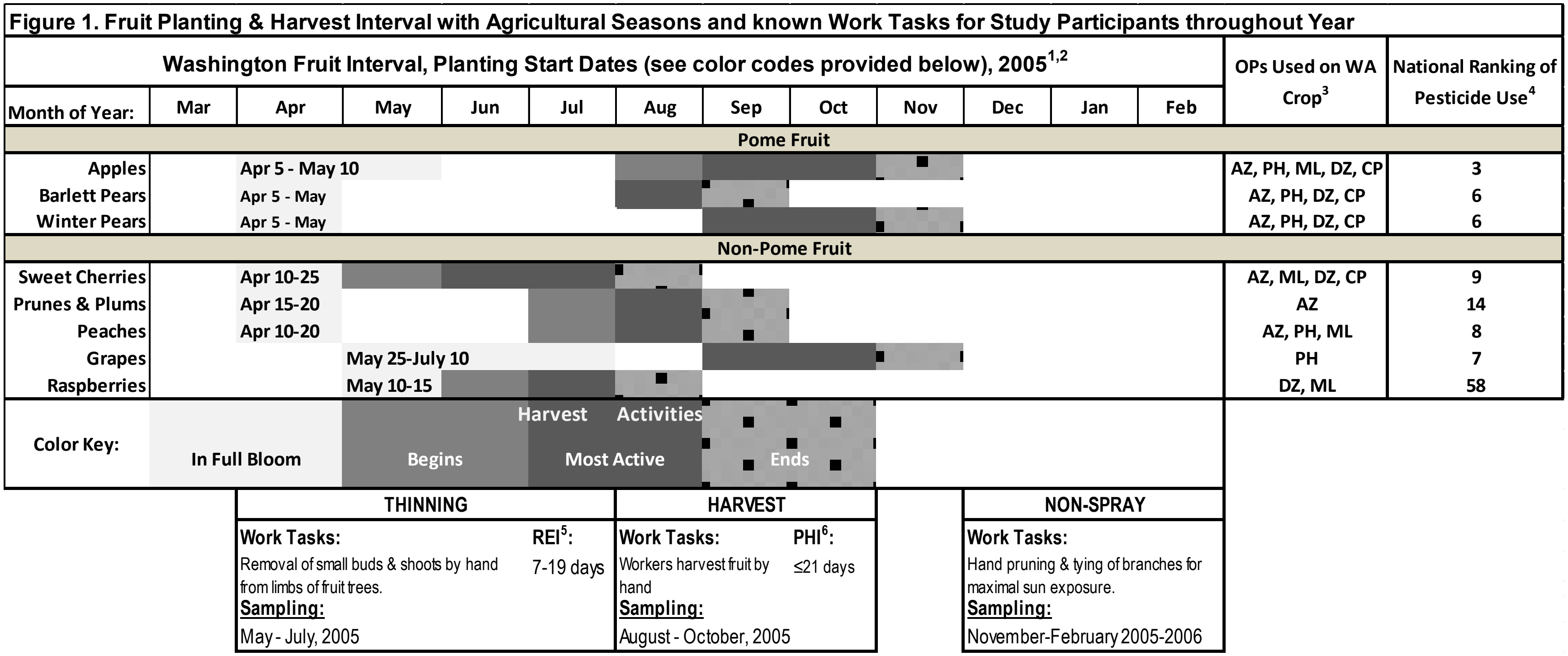

Figure 1. Fruit Planting & Harvest Interval with Agricultural Seasons and known Work Tasks for Study Participants throughout Year

Washington Fruit Interval, Planting Start Dates (see color codes provided below), 2005[1,2]

Month of Year:	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb	OPs Used on WA Crop[3]	National Ranking of Pesticide Use[4]
Pome Fruit														
Apples		░ Apr 5 - May 10	░			▒	▓	▓	▨				AZ, PH, ML, DZ, CP	3
Barlett Pears		░ Apr 5 - May				▓	▨						AZ, PH, DZ, CP	6
Winter Pears		░ Apr 5 - May					▓	▓	▨				AZ, PH, DZ, CP	6
Non-Pome Fruit														
Sweet Cherries		░ Apr 10-25	▒	▓	▓	▨							AZ, ML, DZ, CP	9
Prunes & Plums		░ Apr 15-20			▒	▓	▨						AZ	14
Peaches		░ Apr 10-20			▒	▓	▨						AZ, PH, ML	8
Grapes			░ May 25-July 10	░	░		▓	▓	▨				PH	7
Raspberries			░ May 10-15	▒	▓	▨							DZ, ML	58

Color Key:
- ░ In Full Bloom
- Harvest Activities: ▒ Begins, ▓ Most Active, ▨ Ends

THINNING	HARVEST	NON-SPRAY
Work Tasks: Removal of small buds & shoots by hand from limbs of fruit trees. **REI[5]:** 7-19 days **Sampling:** May - July, 2005	**Work Tasks:** Workers harvest fruit by hand **PHI[6]:** ≤21 days **Sampling:** August - October, 2005	**Work Tasks:** Hand pruning & tying of branches for maximal sun exposure. **Sampling:** November-February 2005-2006

[1] Adapted from "Usual Planting Harvesting Dates, Washington", Wasghington Agriculturla Statistics Service, 2005. [2] Several crops represented have multiple varieties with different planting seasons. [3] See Table 1 for OP specific usage on crops in 2005 (1000's lbs). [4] National Ranking of Pesticide Use by Crops in lbs Active Ingredient/Year in "Pesticide Use in U.S. Crop Production: 2002" CropLife Foundation, 2006. [5] Restricted Entry Interval (REI) time lapse between OP application and Thinning Field work (pesticide-specific). [6] Pre-Harvest Interval (PHI) time lapse between OP application and Hand Harvest of fruit for sale (pesticide-specific).

Organophosphates in House dust

In this study, we characterized the occupational (FW vs NFW) seasonal profiles of household dust for 11 OPs in the Lower Yakima Valley, Washington state, during the 2005–2006 year. Organophosphate pesticides were measured in 320 house dust samples collected across the Thinning (n=117), Harvest (n=117), and Non-Spray seasons (n=86). Figure 2 illustrates geometric mean (GM) concentrations of AZ, PH, ML, CP, and DZ in house dust of FW (circles) and NFW (triangles) across the three seasons.

Concentrations of OP pesticides in house dust were elevated for FW across all seasons. The exception to this is DZ, where seasonal differences were observed, but significant differences between occupations were not. Highest concentrations of OPs in house dust were seen during the Thinning and Harvest seasons and this correlates with known time of OP usage in pome fruit crops and to a larger extent, uses in orchard fruit crops.

Overall, AZ was seen at the highest levels in house dust for both FW and NFW. This was anticipated as the heaviest applications of any OP during 2005 occurred with the use of AZ on the pome fruit crops of apples (196,400 lbs) and pears (18,700 lbs) (NASS 2005). PH had the second highest concentration profile of both occupational groups across all seasons. While this result was not indicative of reported use in lbs per year as illustrated in Table 1, it is reflective of the dynamic and heavy uses of PH on a variety of pome and orchard fruit crops in the Yakima Valley throughout the year, where the highest proportions of active ingredient (a.i) per pound per acre applied are reported for PH are often times in excess of 5.0 lbs a.i./pound/acre (Armitage, Berry et al. 2008). Seasonal trends, increases and decreases, for AZ, PH, CP, and DZ were mirrored across seasons in both occupational groups. However, trends in ML differed by season between FW and NFW, with the FW group showing a downward seasonal trend.

Figure 2. OP Pesticides Detected in House Dust of FW and NFW in Yakima Valley Across Three Seasons: Thinning, Harvest, Non-Spray (see Figure 1 for months) illustrated by geometric mean (ng/g) ± 95% confidence limit. Details for statistical analysis are given in Table 2.

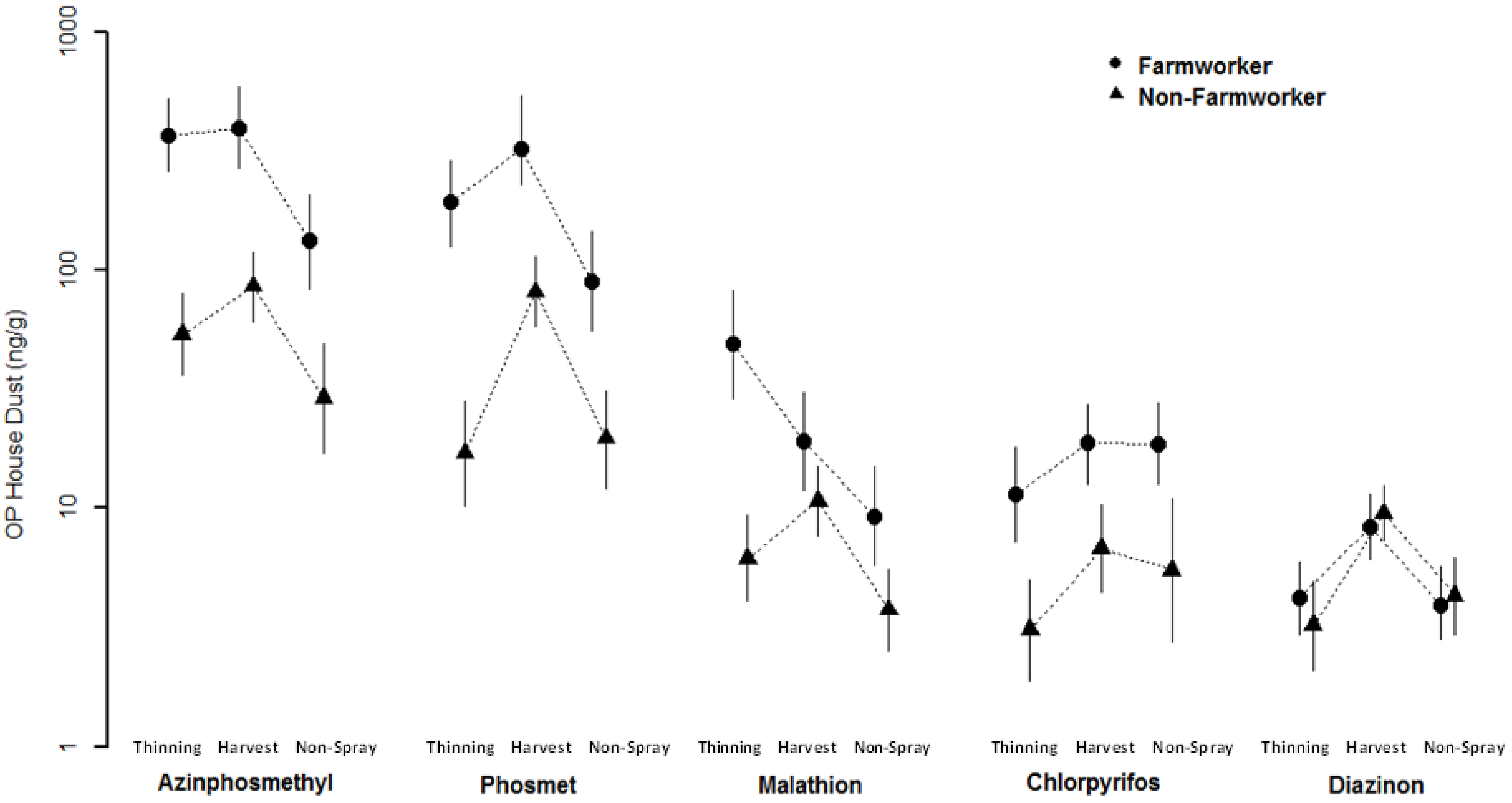

Table 2 provides specific concentrations for the 5 OPs most commonly detected in house dust of FW and NFW participating in this study across all agricultural seasons. Comparisons of GM concentration by occupation in each season for the 5 OPs are noted in the table through the use of asterisks.

Looking across the table from Thinning to Non-Spray, it is evident that AZ and PH had significantly higher levels of pesticide residue in house dust in each season when comparing FW vs NFW, ($p<0.001$, each season). CP also had significantly higher levels of house dust throughout the year when comparing FW and NFW, ($p<0.05$, each season). ML exhibited elevated levels in FW house dust relative to NFW during the Thinning and Non-Spray seasons ($p<0.001$), but the difference was not statistically significant during the Harvest season. And again as illustrated in Figure 2, DZ was the most similar between occupations throughout the year.

Details on occupational and seasonal profiles of each of the 5 OPs (as well as statistics) can be found in Table 2.

Table 2. Concentration (Geometric Mean) of Five Organophosphate Pesticides Most Commonly Detected in House dust in Lower Yakima Valley, WA

		Thinning					Harvest					Non-Spray				
Pesticide:	**Occ[1].**	**Sample(n)**	**DF(%)**	**GM(ng/g)**	**95%CI**	**GSD**	**Sample(n)**	**DF(%)**	**GM(ng/g)**	**95%CI**	**GSD**	**Sample(n)**	**DF(%)**	**GM(ng/g)**	**95%CI**	**GSD**
Azinphos methyl:	FW	51	100	368**	(261.5, 517.1)	3.5	56	98	394**	(268.5, 578.0)	4.3	40	95	133**	(87.9, 200.4)	4.2
	NFW	66	76	52	(35.3, 76.6)	5	61	95	85.5	(62.0, 117.8)	3.6	46	74	29.1	(17.5, 48.5)	5.9
Phosmet:	FW	51	100	192**	(129, 286)	4.3	56	100	342**	(216, 542)	5.8	40	100	89.7**	(57.0, 141.3)	4.3
	NFW	66	83	16.7	(10.3, 27.2)	7.5	61	100	80.7	(58.2, 111.9)	3.7	46	89	19.6	(12.4, 31.1)	4.9
Malathion:	FW	51	100	48.8**	(29.4, 80.8)	6.3	56	100	19.1	(12.1, 30.3)	5.8	40	95	9.3**	(5.8, 14.7)	4.5
	NFW	66	85	6.2	(4.2, 9.3)	5.2	61	100	10.7	(7.7, 14.9)	3.7	46	87	3.7	(2.5, 5.5)	3.8
Diazinon:	FW	51	100	4.2	(3.0, 5.9)	3.5	56	100	8.4	(6.2, 11.4)	3.2	40	95	3.9	(2.8, 5.5)	3.0
	NFW	66	86	3	(2.1, 4.2)	4.1	61	98	9.5	(7.3, 12.2)	2.8	46	96	4.3	(3.0, 6.2)	3.5
Chlorpyrifos:	FW	51	55	12.3*	(8.0, 18.9)	4.8	56	59	18.7*	(12.8, 27.4)	4.3	40	75	18.6*	(12.7, 27.2)	3.4
	NFW	66	30	3	(1.9, 4.8)	7.2	61	43	6.8	(4.5, 10.2)	5.1	46	41	5.5	(2.8, 10.7)	10.2

[1] Occupation: Farmworker (FW) or Non-Farmworker (NFW). DF: Detection Frequency greater Limit of Detection. GM: Geomean (ng/g). GSD: Geometric standard deviation. Significance indicated for between Occupation comparisons: *(p<0.05). **(p<0.001).

Azinphos methyl

As evident from Figure 2, the highest concentrations of azinphos methyl (AZ) in FW house dust were observed during the Thinning and Harvest seasons when agricultural use of AZ occurs to chemically thin fruit to enhance fruit production and to preserve fruit integrity during and after hand harvest. Peak levels were seen during the Harvest season (GM 394 ng/g house dust; GSD 4.3). A significant reduction in FW house dust AZ occurred between the Harvest-to-Non-Spray seasons ($p<0.001$) and from the Thinning-to-Non-Spray seasons ($p<0.05$).

Significantly lower concentrations of AZ were detected in NFW house dust across all seasons. These concentrations followed seasonal profiles like FW house dust, with peak detection during the Harvest season (GM 85.5 ng/g house dust; GSD 3.6). NFW AZ house dust concentrations had greatest reductions in concentration of AZ from Harvest-to-Non-Spray ($p<0.001$).

Table 2 shows specific concentrations and percent detection by occupation of AZ in house dust across the Thinning, Harvest, and Non-Spray seasons. Detection frequency of AZ in house dust was higher for FW across all seasons, illustrative of an occupational signal of this Restricted Use Pesticide in house dust.

FW house dust in the Thinning season had a significantly higher concentration of AZ (7-fold greater) when compared to NFW house dust ($p<0.001$), with GM concentrations of 368 ng/g house dust and 52 ng/g house dust, respectively. During the Harvest season FW house dust sustained a higher level of AZ in than that observed during the Thinning season. NFW also had slight increases in AZ house dust from the Thinning to Harvest seasons, although this increase was not significant. Overall, FW

Harvest season AZ residue in house dust (GM 394 ng/g) was 4.6-fold higher than NFW Harvest season AZ levels in house dust (85.5 ng/g), ($p<0.001$). AZ concentrations were lowest in the Non-Spray season for either occupation, reflective of restrictions on dormant season applications. Both FW and NFW AZ in house dust decreased by approximately 66% during the Non-Spray season, from peak concentrations observed for both occupational groups seen in the Harvest season, with GM concentrations of 133 ng/g and 29.1 ng/g house dust respectively. Although both groups saw great reduction in AZ concentration in house dust, FW again had significantly higher levels of AZ, 4.6-fold greater ($p<0.001$).

Phosmet

Figure 2 shows that phosmet (PH) had the second highest concentration profile across all seasons for FW and NFW. The highest levels of PH in house dust were seen in the Harvest season for both FW and NFW similar to the profiles seen in AZ for both occupational constructs. As stated previously, these dust concentrations are reflective of PH reported use in 2005 relative to other reported OPs. Significant changes were seen in FW household PH dust concentrations between the Thinning-to-Harvest (increase) ($p<0.05$), from the Harvest-to-Non-Spray (decrease) ($p<0.001$), and from the Thinning-to-Non-Spray (decrease) ($p<0.001$). Although NFW PH house dust profiles in Figure 2 are much lower than those of FW, the data show that across seasons trends were similar in both groups, reinforcing the hypothesis that pesticide levels in house dust are influenced by agricultural seasons throughout the year, regardless of occupation. Significant changes in seasonal concentrations for NFW were apparent from Thinning-to-

Harvest (increase) ($p<0.001$) and from Harvest-to-Non-Spray (decrease) ($p<0.001$). Concentrations of PH in NFW house dust were very similar in the Thinning and Non-Spray seasons.

Table 2 provides specific concentrations of PH in house dust of FW and NFW across the Thinning, Harvest, and Non-Spray seasons. The table shows that the peak FW concentration observed occurred during the Harvest season (GM 342 ng/g house dust) before the tremendous decline in PH in house dust from Harvest-to-Non-Spray, where concentrations reduced 3.8-fold (GM 89.7 ng/g house dust).

Levels of PH exceeded the limit of detection in the Thinning season for FW in 100% of samples and for NFW in 83% of samples. Although PH is not a Restricted Use Pesticide, its major uses occur in agriculture, potentially owing to the difference in detection during the Thinning season. Thinning season FW house dust had GM concentrations 11.5-fold greater than NFW house dust, specifically 192 ng/g FW house dust vs 16.7 ng/g NFW house dust, ($p<0.001$). Harvest season FW PH was again elevated, 4.2-fold greater when compared to NFW Harvest season PH in house dust, ($p<0.001$). The concentration of PH in the Non-Spray season house dust was dramatically reduced relative to the Harvest season house dust for both occupations with a FW GM concentration of 89.7 ng/g house dust and NFW GM concentration of 19.6 ng/g house dust. Overall, FW PH in house dust during the Non-Spray season was approximately 4.6-fold higher than NFW Non-Spray house dust ($p<0.001$).

Malathion

FW malathion (ML) house dust trends in Figure 2 appear somewhat different when examined against seasonal trends for the other OPs and the NFW ML house dust trends. FW house dust ML peaked during the Thinning season, and steadily declined through the remainder of the year, with the largest difference in FW ML concentration profile occurring from Thinning-to-Non-Spray ($p<0.001$) and from Thinning-to-Harvest ($p<0.01$), reflecting 81% and 61% declines in ML respectively. NFW house dust ML profiles were similar to trends observed for other OPs including AZ, PH and DZ with highest detection of ML in house dust during the Harvest season. The greatest reduction in NFW house dust occurred from the Harvest-to-Non-Spray season, where concentrations dropped by approximately 65% ($p<0.001$).

Concentrations provided in Table 2 indicate peak FW ML house dust concentrations at 48.4 ng/g house dust during the Thinning season and these levels of ML in house dust were 7.9-fold greater than those observed in NFW house dust ($p<0.001$). During the Non-Spray season, FW levels of ML in house dust were 2.5-fold greater than NFW ML levels in house dust, ($p<0.001$). Peak NFW house dust concentrations were observed at 10.7 ng/g house dust during the Harvest season. Differences in detection frequency were most apparent during the Thinning and Non-Spray seasons when GM concentrations of ML detected in house dust differentiated by occupation, suggestive of occupational signals in use during these seasons.

Diazinon

Diazinon (DZ) had the lowest detection of any of the OPs throughout the year as shown in Figure 2. Also evident in this figure are the mirrored seasonal profiles of DZ throughout the year for both occupational constructs. The seasonal variation observed was similar to that of the other OPs with peak detection occurring during the Harvest season, with FW GM concentration 8.4 ng/g house dust and NFW GM concentration 9.5 ng/g house dust respectively. Significant increases occurred for both occupational constructs from Thinning-to-Harvest season, approximately 2-fold ($p<0.001$), followed by significant decreases from Harvest-to-Non-Spray, again approximately 2-fold ($p<0.001$). DZ levels in house dust were lowest during the Thinning and Non-Spray seasons and were not significantly different between occupational groups ($p>0.95$) and seasons ($p=0.3$), suggesting related use of this pesticide during these two seasons.

As a Restricted Use Pesticide, differences in detection frequency between FW and NFW households were anticipated but not observed. As shown in Table 2, the largest difference in detection frequency between FW and NFW house dust occurred during the Thinning season where DZ was detected in 100% of FW house dust samples and 86% of NFW house dust samples. Detection frequency for NFW house dust DZ increased from Thinning-to-Harvest season (86% to 98%) and then decreased slightly from Harvest-to-Non-Spray (98% to 96%), suggesting a potential increased use of DZ during the Harvest season.

Chlorpyrifos

Seasonal and occupational trends of chlorphyrifos (CP) in house dust are illustrated in Figure 2 with FW consistently having elevated levels of CP in house dust across the year. The highest and most similar levels of CP in FW house dust were detected in the Harvest and Non-Spray seasons ($p=0.9$). Although increases of CP in FW house dust occurred from Thinning-to-Harvest and Thinning-to-Non-Spray, these increases were not significant. NFW CP house dust mirrored the trends seen with FW CP house dust, with highest and most similar levels of CP detected in house dust during the Harvest and Non-Spray seasons. Again although increases were observed from Thinning-to-Harvest and Thinning-to-Non-Spray for NFW, none of these increases were significant.

Specific concentrations and detection frequency for CP in house dust each season for FW and NFW are presented in Table 2. Immediately apparent from the table are the stark differences in detection frequency throughout the year by occupation for CP, as these differences in detection frequency are greatest for CP when compared to any other OP in this study. As CP saw earliest phase-outs of any OP included in this study, the differences in detection frequency of this Restricted Use Pesticide are anticipated. Specifically, NFW households' percent detection ranged from 30, 43 and 41% respectively across the Thinning, Harvest and Non-Spray Seasons. Detection frequencies in FW households were slightly higher and ranged from 55, 59 and 75% for Thinning, Harvest and Non-Spray Seasons, respectively. Depending on season, difference in detection frequency varied from 16 to 34%, with FW consistently having higher detection of CP in house dust.

The lowest concentration of CP was observeded in the Thinning season for both occupational groups, with FW having 4-fold greater levels of CP in house dust than NFW at 12.3 ng/g house dust ($p<0.05$). Highest concentrations for FW and NFW occurred in the Harvest season with FW GM concentration of 18.7 ng/g and NFW GM concentration of 6.8 ng/g; a 2.8-fold difference by occupation ($p<0.05$). During the Non-Spray season house dust CP of FW was again elevated at 18.6 ng/g house dust while NFW house dust has CP at concentrations 5.5 ng/g house dust, reflective of an occupational difference of 3.4-fold ($p<0.05$).

The greatest paired difference of CP in house dust as evaluated by fold-difference in GM concentration and percent-difference in detection frequency occurred during the Non-Spray season, where FW house dust had CP levels 3.4-fold greater than NFW with concomitant detection of CP in FW house dust at 34% greater than in NFW households. This pattern points to the greatest occupational and seasonal difference of CP to occur during the Non-Spray season which is in line with permitted applications of CP outlined in registration eligibility documents for this OP.

Chapter 5: Discussion

In response to questions expressed by the Community Advisory Board after research conducted in 1999 and 2002, this project sought to better characterize occupational (FW vs NFW), and seasonal (Thinning, Harvest, and Non-Spray) profiles of agricultural contaminants in the Lower Yakima Valley, Washington. Specifically, the research sought to test the hypothesis that indoor concentration profiles of agricultural pesticides in house dust vary according to worker status, pesticide use classification, agricultural season of year, and potential for sprayed crop contact during each season. Results for AZ, PH, ML and CP validated this hypothesis.

Looking at Figure 2 and Table 2, it is evident that of all OPs examined in this study AZ had the strongest occupational and seasonal signal in house dust and these observations are supportive of our previous studies conducted through the CHC in this agricultural cohort and exemplify the hypothesis examined here. AZ concentrations in FW house dust were consistently elevated when compared to AZ profiles in NFW house dust, regardless of season. As AZ is a Restricted Use Pesticide with time-limited crop specific uses; this trend indicated an occupational and use classification signal in house dust. The house dust pesticide contaminant level profiles for AZ also most closely reflected the active portions of the pome and orchard fruit seasons during the transition from Thinning-to-Harvest seasons, where highest concentrations of AZ in house dust were observed with known AZ applications (2–3 times per yr). AZ in house dust also reflected the most dormant portions of the pome and orchard fruit seasons during the Non-Spray season, when lowest levels of AZ were detected in a period of no applications permitted (USEPA 2006).

Knowledge about dominant pest lifecycles such as that of codling moth in the Yakima Valley (which has up to 4 generations spanning the agricultural year), identifies that applications of AZ to pome and orchard fruit crops can occur pre-Thinning, during the Thinning season, and also up-to pre-Harvest season to treat the pest. Self-reported practices of apple growers in the Yakima Valley, Apple and Pear Pesticide Use Survey (1989-2000), indicate that 69% of survey participants (n=98) responded to treating for codling moth on an average 2.8 applications per year (Washington State University Extension 2003). Growers also reported average and range of last applications for each agricultural chemical class during 2000, including the insecticide class encompassing AZ. On average, last applications occurred on July 10th, but could have occurred over a time period ranging from the June 27th to August 13th of that year. The trends in Thinning and Harvest season AZ FW house dust observed herein reflect this dynamic and are consistent with staggered AZ pome fruit crop management. As AZ has been found as a surface residue up to 14–119 days after application with only slight to moderate absorption in plants such as apples (Magill and Everett 1966; Drager 1987; Krolski 1988; Gronber, Pitcher et al. 1998). Its use in codling moth management potentiates the opportunity for AZ worker fruit contact. FW can then track AZ home during two active seasons, Thinning and Harvest, the peak AZ detection seasons seen in our study.

Urinary dimethyl metabolite analysis for this cohort (2005-2006) has been previously completed, with peaks also occurring during the Thinning and Harvest seasons for AZ (Thompson, Griffith et al. 2013). These urinary metabolite profiles trend in a similar manner with house dust profiles for AZ (data not shown). In addition, parent OP levels for AZ and CP align with our findings (Thompson, Griffith et al. 2013).

Previous research in this cohort has documented adult urine DMP and DMTP concentrations significantly correlated with vehicle and house dust AZ concentrations. Child urine DMP and DMTP concentrations were also significantly correlated with house dust AZ concentrations (Coronado, Vigoren et al. 2006). Integration of the urine and house dust biomarkers in this second wave of research establishes house dust as an environmental biomarker that is influenced and reflective of multiple factors including worker status, agricultural season, pesticide use classification, and contact with sprayed fruit and orchard crops. Our house dust AZ results support this hypothesis. We are continuing to show additional factors such as pesticide drift from spraying OPs which could affect dust and urine levels, and diet which might affect urine levels. As we are able to account for additional factors we will be able to better understand the connection between OPs in dust and exposure of household members to OPs.

Results from this study also further contribute to a growing body of research evidence in the Lower Yakima Valley confirming the potential for FW and NFW adult and child to be exposed to toxic Restricted Use Pesticides such as AZ and CP. As the EPA continues to carry out total phase-outs of these products, information such as concentration of AZ and CP in house dust during years of use (such as in this study) will become increasingly valuable in assessing the environmental persistence and degradation of these products indoors after use has been banned. More specifically, the 7 phased-out and 8 time-limited uses of AZ expired on September 30, 2012 (USEPA 2006). As no Non-OP alternative pesticides provide adequate protection against pests such as codling month (most resulting in secondary outbreaks), economic projections for the ban of AZ uses in apples have estimated losses in excess of $50 million dollars (USEPA 2006).

Although Phosmet has reduced residual activity in apples when compared to AZ for pests such as codling moth, opportunity exists for increased use of this OP to supplement the AZ-banned apple industry. Future UW CHC house dust samples should be watched for reflection of this AZ use-ban phenomenon in this predominantly agricultural area.

Households participating in this study had mostly Low Plush Carpet or Tile flooring and most collection occurred in the living room area (data not shown), which was the area parent-identified place of where child spent most of his/her time playing. The influence of floor type on pesticide loading and concentration in house dust has been established previously in this cohort (Coronado, Griffith et al. 2010). Assessment of floor type would be important for future studies as carpeting (with underlying polyurethane foam padding) has the ability to act as a sink for pesticide-laden particulate matter (dust) with opportunity for transfer, resuspension, and replenishment of pesticides in carpet fibers during cleaning events (Camann 1994; D.R., Blanchard et al. 2000).

While efforts to characterize household cleaning practices via the use of household surveys were undertaken, these data have not yet been fully investigated for influence on house dust concentration profiles. As mentioned previously, published research have demonstrated that public health prevention regimes can reduce dust loading and exposure via altered household hygiene practices (e.g sweeping and vacuuming) (Lioy, Yiin et al. 1998; Galke, Clark et al. 2001; Thomson, Petticrew et al. 2003). Other research has focused on household hygienic characteristics to investigate include participant access to cleaning supplies (e.g. broom or vacuum) and prevalence of traditional cultural gender roles in participating households as these have been documented to influence divergent household sanitation practices between men and women (Rao, Quandt et al. 2006).

Starting in 1999, a community-based intervention study was conducted to reduce the occupational take-home exposure pathway. Initial results suggested minimal impacts (Thompson, Coronado et al. 2008) however subsequent detailed analysis showed significant reductions within households (Vigoren EM, Griffith WC et al. 2006). These results support further intervention research with a focus on household dust as a reservoir.

Overall, the intensive sampling campaign of biomarker of exposure (house dust) and biomarker of effect (OP blood and urinary analysis) in participating households have the ability to generate results with potential application in observational studies such as the National Children's Study, a multiagency longitudinal cohort study, examining the association of pesticide exposure with adverse health effects in children (Branum, Collman et al. 2003). Our study has focused on farmworkers who are not applicators, however these finding are also relevant for applicators who already receive training but could benefits from additional emphasis from our findings. Results may also aid in occupational health and safety protection for migrant agricultural communities and families to decrease pesticide exposures.

Chapter 6: Conclusion

The longitudinal sampling scheme of the Center for Child Environmental Health Risks Research (CHC) has provided multiple advantages for evaluating and characterizing potential exposure pathways in agricultural communities and as it has collected home environment dust across household, seasons and years, with detailed detection of multiple organophosphate pesticides. This research was completed in partnership with Dr. Beti Thompson as a continuation of the community-based participatory research (CBPR) campaign originally undertaken by the CHC in the Lower Yakima Valley, Washington. Retention success throughout the campaign led to the 200 households explored in this study (Thompson, Coronado et al. 2003; Thompson, Griffith et al. 2013).

House dust levels of 5 OP pesticides in this study were found to vary throughout the year based on occupation with FW consistently presenting higher concentrations of OPs, including consistently higher levels of Restricted Use Pesticides. Specifically, pesticides with the strongest differences by occupation throughout the year were AZ ($p<0.001$), PH ($p<0.001$) and CP ($p<0.05$). These results were anticipated as all three OPs had the heaviest reported use during 2005 on pome and orchard fruit crops. Overall AZ was detected in highest concentration throughout the year followed closely by PH.

The exception to this trend was DZ, which had the lowest concentrations detected and exhibited the least difference by occupation in this study throughout the year and presented nearly identical concentration profiles between occupational groups (Harvest season, $p=0.9$). Minimal information exists on DZ use in pome and orchard fruit crops and to a larger extent agriculture use in the Yakima Valley. Other information does however exist illustrating that DZ has registered use as an insecticide in the livestock industry for a variety of species including: cattle (dairy, non-dairy), swine, and sheep (USEPA 2004) as pointed out in Table 1. While the CHC has surveyed self-report of occupational status of participants in this study (one adult per household), these questions have focused on eliciting information surrounding orchard crops, and more specifically pome fruit crops, as increased risk for pesticide exposure has been identified in this crop occupation in previous studies within this cohort.

The Yakima Valley ranks 1st by county in the state of Washington for “milk and other dairy products from cows” and “sheep, goats, and their products”(NASS 2007). Furthermore, the county is ranked 2nd for commodity sales of “cattle and calves” and 3rd for “hogs and pigs”(NASS 2007). While non-farmworkers reported no occupational activity in orchard and pome fruit crops during this study, they did not report occupational activity of adults in participating households in another agricultural industry such as livestock that could explain the trend seen in DZ house dust here. As such our ability to detect an occupational trend for DZ may have been minimized.

Seasonality, in line with pome and orchard fruit crops, was observed and these associations were strongest for AZ, PH and CP, with peaks and troughs in house dust levels closely reflecting published use summaries and reports on pesticide applications to pome and orchard fruit crops. ML FW house dust exhibited a unique seasonal trend unlike those observed for AZ, PH, DZ and CP FW house dust, and ML NFW house dust, where highest concentrations for those OPs were reached during the Harvest season. ML in FW households peaked during the Thinning season and generally declined throughout the remainder of the year. As ML is a General Use Pesticide with permitted agricultural, residential, civil, and public health program applications, further investigation of residential uses is needed to explain the trends observed of ML in house dust here.

The largest seasonal differences observed in this study occurred from the Harvest-to-Non-Spray and Thinning-to-Non-Spray seasons for AZ, PH, ML, DZ and CP. Almost all pesticides were detected in the lowest amounts during the Non-Spray season, truly reflective of this season as an “off” season in the Yakima Valley. The exception to this was CP, which had lowest detections in the Thinning season. CP has a unique application scheme in pome and orchard fruit crops allowing for one dormant “off” season application in the Non-Spray season occurring directly to plants/crops where FW contact occurs via work tasks (e.g. pruning) specified in Figure 1. Latter in the year up-to 2 more applications are permitted, however these applications are required to occur at the trunk and base of orchard fruit crops. Based on these regulatory stipulations, the results observed for CP in house dust were consistent with our hypotheses (USEPA 2002).

Overall, house dust levels of pesticides were variable throughout the year and were influenced by householder work status in pome and orchard fruit crops, agricultural season, use classification of pesticides, and potential for contact with sprayed fruit and orchard trees dictated by work status. The Yakima Valley is a cornucopia of agricultural activities and most associations and trends of OPs in house dust observed were explainable by Federal and State-level reported use summaries and crop schedules when work activities were considered.

As levels of AZ, PH, ML, DZ and CP in house dust were episodic depending on season, they present episodic exposure risk to populations residing indoors, especially children. This is of particular concern as dynamic developmental and behavioral life stages may predispose children to greater exposure to house dust laden with pesticides. A frequent question for investigators with longitudinal cohorts, is the required frequency of sampling and in this study significant differences in pesticide levels across seasons suggests the need for repeated samples in order to understand factors affecting within person and across household variability.

Significant differences were seen across occupational constructs based on the use classification of pesticides, illustrating a para-occupational take-home pathway and potential increased pesticide exposure and risk for FW families in the Yakima Valley. As these pesticides are classified as Restricted Use based on risk of use and potential for toxicological effect, special attention should be given to them. Further investigation incorporating these episodic and Restricted Use house dust concentration profiles is warranted in indoor residential pesticide exposure assessment.

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Appendix

Table 1. Limit of Detection of 11 Organophosphate Pesticides and 18 Other Pesticides

OPs	Limit of Detection (ng/g)*	Other Pesticides	Limit of Detection (ng/g)*
Methamidophos	1	Oxamyl	1
Dimethoate	1	Methomyl	0.4
Dichlorvos	10	Thiamethoxam	0.4
Phosmet	0.4	Imidacloprid	1
Malathion	0.4	Acetamiprid	0.4
Diazinon	0.4	Aldicarb	100
Coumaphos	1	Carbofuran	1
Phorate	4	Pirimicarb	0.4
Chlorpyrifos	4	Carbaryl	1
Azinphos-Methyl	10	Diuron	0.4
Methyl Parathion	40	Linuron	0.4
		Tebuconazole	0.4
		S-Metolachlor	0.4
		Propiconazole	0.4
		Triflumizole	0.4
		Pendimethalin	0.4
		Clothianidin	0.4
		Propoxur	0.4

* Limited of detection assumes 1g of dust was provided for analysis of pesticide concentration in house dust.

Table 2. Hispanic Population in the Lower Yakima Valley

	Mabton CCD*	Sunnyside CCD	Toppenish-Wapato CCD	Northeast Benton CCD	Total	%
Hispanic	2,925	36,431	17,374	5,661	62,391	65%
Non Hispanic	515	15,234	9,776	7,961	33,486	35%
Grand Total	3,440	51,665	27,150	13,622	95,877	100%

* CCD - Census County Division

http://www.census.gov/2010census/popmap/ipmtext.php?fl=53
http://www.census.gov/2010census/popmap/index.php