10-19-2016

A Literature Review of Wipe Sampling Methods for Pesticides in Published Exposure Measurement Studies in the United States

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A Literature Review of Wipe Sampling Methods for Pesticides in Published Exposure Measurement Studies in the United States

by

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A thesis submitted in partial fulfillment
of the requirements for the degree of
Master of Science in Public Health
Department of Environmental and Occupational Health
College of Public Health
University of South Florida

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Date of Approval:
October 16, 2016

Keywords: Pesticide, Surface Wipe Sampling

Dedication

This thesis is dedicated to my wife, Novella Low. It has been a marvelous journey together through our unique educational paths. *Grazie per avermi incoraggiato. Ti amerò per sempre.*

Acknowledgments

I would first like to thank my wife, Novella, for supporting me through all of my challenges and always motivating me when times were tough. I would also like to thank the Navy for providing all of their support and funding for my graduate education at USF. I would like to give a distinct recognition to my advisor, Dr. Steven Mlynarek, for devoting his time and knowledge throughout my two years in the program. I must also thank Dr. Giffe Johnson for all of his time helping me with this thesis. Most importantly, I would like to thank the Industrial Hygiene Faculty Dr. Thomas Bernard, Dr. Yehia Hammad, Dr. René Salazar, and Dr. John Smythe for providing me with an excellent education and preparing me for a professional career as an industrial hygienist.

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List of Abbreviations and Acronyms

AAPCC	American Association of Poison Control Centers
AHHS	American Healthy Homes Survey
AIHce	American Industrial Hygiene Conference and Expo
CBPR	Community-Based Participatory Research
C18	Carbon-18
DI	Deionized
EPA	Environmental Protection Agency
EL	Edwards and Lioy
HUD	Department of Housing and Urban Development
HPHI	Healthy Public Housing Initiative
HVS3	High Volume Small Surface Sampler
IPA	Isopropyl Alcohol
IRB	Institutional Review Board
LWW	Lioy, Wainman, and Weisel
MNCPES	Minnesota Children's Pesticide Exposure Study

NCS	National Children's Study
NIOSH	National Institute for Occupational Safety and Health
OP	Organophosphate
OSHA	Occupational Safety and Health Administration
PCBs	Polychlorinated Biphenyls
PBDEs	Polybrominated Diphenyl Ethers
PTFE	Polytetrafluoroethylene
PUF	Polyurethane Foam
QA	Quality Assurance
QC	Quality Control
SDRs	Surface Dislodgeable Residues
SOP	Standard Operating Procedure
SRSs	Surrogate Standard Recoveries
TPs	Transformation Products
U.S.	United States
USF	University of South Florida
USDA	Unites States Department of Agriculture
2, 4-D	2, 4-Dichlorophenoxyacetic Acid

Abstract

Pesticides in the United States are frequently used to control pests in many settings from residential homes to agricultural crops. Most pesticides, when used in accordance with their manufacturer's label are relatively safe, and will naturally degrade once exposed to the environment, however, these natural degradative processes can be hindered when introduced indoors. Furthermore, it has been shown that pesticides can easily bond to surface dislodgeable residues (SDRs) commonly known as dust. There are various methods that can be used to characterize the presence and exposure of pesticides indoors. Wipe sampling is one of the important methods commonly used to measure pesticides on surfaces due to its simple and inexpensive nature, however, several methods exist for wipe sampling and each method has varying steps involving different wiping material, pre-treatment of wipes, wetting solvent, surface type, collection pattern, and storage.

The purpose of this literature review is to summarize concisely the methods from eighteen recent studies that used surface wipes to sample for pesticides from indoor environments. This report details the methods applied to perform the literature review, provide general wipe sampling information from government agencies, discuss other related surface sampling methods, provide a brief summary of wipe sampling methods applied in each study, and compare the methods applied to provide considerations for those seeking to use surface wipes for sampling pesticides.

Overall, it would seem that there are more variations than similarities between wipe sampling methods from the literature reviewed. Similarities included the use of isopropyl alcohol (IPA) as the wetting solvent and how wipe samples were stored after collection. The differences in wiping materials, pre-treatment of wipes, surface types, and collection patterns still demonstrate the need for a standardized method. Until a standardized method is established, poor comparisons of study results will continue and knowledge gaps will remain.

Introduction

Approximately 5 billion pounds of pesticides was estimated to have been used in the United States (U.S.) in 2006 and 2007 (EPA, 2011). The Environmental Protection Agency (EPA) estimates that the volume of pesticides used in the U.S. has remained relatively stable over the years and attributes this to the development of more potent pesticides (Harbison, 2015). Each year there are approximately 900 pesticides used, each with different dilutions and ingredients that are configured in more than 20,000 formulations (Schenker, 2007). Most of these pesticides are relatively safe when used in accordance with their manufacturer's label. Typically, when pesticides are applied outside they are subjected to various environmental degradation processes such as sunlight, moisture, and soil, however, these processes are reduced indoors and pesticides may persist and accumulate over time. Organic compounds, like pesticides, have been shown to bind easily to dust particles and can be found in surface dislodgeable residues (SDRs), also known as house dust or indoor dust (Weschler & Nazaroff, 2008; Cettier et al., 2015).

There are various methods to collect chemical residues and SDRs however, surface wipes represent an important, inexpensive, and simple method for the estimation of pesticide deposition on surfaces and in SDRs. Currently, a standardized method for the collection of pesticides from surfaces does not exist. Certain government agencies have published methods to collect surface contaminants such as volatile organic compounds and lead, but not pesticides specifically. Many studies have used surface wipes to evaluate pesticide exposure, although their methods typically are altered versions for other environmental contaminants or they do not clearly state within their study the source of their method. This variability in the collection of data makes it difficult for professionals in the same community to efficiently and effectively compare data.

The purpose of this literature review was to compile studies that have used surface wipes to evaluate pesticide residue or SDRs on surfaces to compare their methods. By evaluating the various methods of eighteen relevant pesticide exposure studies, the expectation is to reveal any underlying commonality between methods that can aid in the future standardization of a surface wipe sampling for pesticides.

The specific objectives of this literature review were:

1. Provide general wipe sampling information from various government agencies.
2. Concisely summarize the methods for surface sampling of pesticides to identify common methodical steps.
3. Detail some considerations to take when choosing a method and materials for sampling surfaces for pesticides.

Literature Search Methods

The literature review consisted of finding the most current methods used for surface wipe sampling of pesticides. This entailed searching major government agency websites, peer-reviewed journals, and various certified analytical laboratories. The initial search began by meeting with representatives of certified laboratories at the 2016 American Industrial Hygiene Conference and Expo (AIHce) in Baltimore, Maryland. The information provided by these laboratories aided in the extensive internet search. These searches used PubMed and the University of South Florida's (USF) online library services search system. In addition, the following government agency websites were used: Occupational Safety and Health Administration (OSHA), National Institute for Occupational Safety and Health (NIOSH), Unites States Department of Agriculture (USDA), and Environmental Protection Agency (EPA). These government agencies were selected because they were entities most likely to have documented and standardized methods for surface wipe sampling of pesticides.

To keep the scope of the literature review focused on recent methods applied to collecting surface wipe samples for pesticides, generalized keywords and statements were used for the database search included the following phrases: wipe OR surface sampling OR sampling OR pesticide OR pesticide AND sampling OR pesticide AND wipe AND sampling. Furthermore, results were limited to date of publication from the years 2000 to 2015 and studies completed in the United States.

General Wipe Sampling Information

Methods by Government Agency

Currently, there are no government agencies that have published a standard specifically for surface wipe sampling of pesticides however, most government agencies do have a general guideline for the wipe sampling of surfaces. The following section will outline the general wipe sampling techniques used by different government agencies. This is not meant to provide an exhaustive detail of all government agencies' wipe sampling methods but a brief overview of some wipe sampling methods by the previously stated government agencies.

For the analysis of occupational exposures, surface wipe sampling is typically performed on workplace surfaces that have been suspected of contamination with hazardous liquids, particles, or dried residues that may not be visible to the naked eye (OSHA, 2012). OSHA has published evaluation guidelines for analyzing surface samples in order to provide scientists and researchers a uniform method for analysis. The guidelines provide details on how to perform a wipe sample. A 100 cm^2 area that is smooth and non-porous is recommended for ideal surface samples. There are various wetting agents suggested for wetting wipes such as deionized water (DI) water for metals, isopropyl alcohol (IPA) for non-volatile organics, or other various compounds depending upon the compound to be sampled (OSHA, n.d.). The recommended surface wipe materials by OSHA include the DURX 670 (10 cm×10 cm polyester and cellulose), Pro-Wipe 880 (20 cm × 25 cm polypropylene), Dissolving (Ghost) Wipes (12 cm ×12 cm cross-linked polyvinyl alcohol) and AlphaWipes (23 cm × 23 cm polyester). OSHA has also published a chapter in their technical manual that details information on surface sampling which is similar to their evaluation guidelines (OSHA, 2012).

NIOSH has prescribed methods for evaluating pesticides through air sampling that is detailed in NIOSH Method 5600 and NIOSH Method 5601, however, there are no current NIOSH methods designed to sample surfaces for pesticides with wipes. While there are no current NIOSH methods for surface wipe sampling of pesticides, NIOSH Method 9100 details surface wipe sampling for lead that some laboratories have altered to sample for pesticides (NIOSH, 1994). This method requires the use of a 2" x 2" sterile gauze pad (Curity, Johnson & Johnson, or equivalent) to be wetted with 1 to 2 ml of distilled water and then used to wipe within a 10 cm x 10 cm template. Currently, there is a laboratory that has a draft method for surface sampling of pesticides under review by NIOSH (ALS, 2016). NIOSH draft method 9204 is similar to NIOSH method 9100 however, this method is under review and cannot be publicly circulated.

A standard operating procedure (SOP) published by the USDA details the collection of wipe samples for residue analysis (USDA, 2012). This SOP, No. EM – 24, specifically outlines the procedure for the collection of wipe samples for organic chemical residues from surfaces. It recommends a 3" x 3" sterile gauze pad to be moistened with IPA and then to wipe an area of approximately 100 cm^2. The wipe pattern performed begins by wiping the outside edge of the 100 cm^2 area and progressing inward making concentric squares of decreasing size. The wipe is then folded and placed back into the sampling bag it came in and then placed in a cooler.

The EPA has conducted many exposure studies for organic compounds and pesticides. A method for wipe sampling polychlorinated biphenyls (PCBs) on hard, smooth, non-porous surfaces by the EPA uses Whatman filter paper or gauze pads wetted with isooctane or hexane to wipe a 100 cm^2 area (Boomer et al., 1985; Kelso et al., 1986; Billets, 2007). The EPA has also published an extensive literature review on wipe sampling methods for chemical warfare agents and toxic industrial chemicals (Billets, 2007). This document is the only report found that has compiled literature to identify the state-of-the-art sampling methods for surface wipes. The EPA's review concludes that there are gaps within the methodology for wipe sampling, there is no overwhelming consensus on how to take wipe samples, and these gaps can only be filled through research and method validation (Billets, 2007).

Other Surface Sampling Methods

Surface sampling methods have multiple variations and can be used on many different surface types. This section of the report will discuss related sampling techniques that involve alternative methods to sampling smooth surfaces and other surfaces such as carpets.

To sample SDRs or dust from carpets, the preferred method typically involves a vacuum. Most studies utilize the High Volume Small Surface Sampler (HVS3) that is basically a vacuum modified to capture dust particles into an attached sampling container (Billets, 2007; Bradmen et al., 2007; Curwin et al., 2005; Lu et al., 2000). Commercially available vacuums also have been modified to collect samples from carpets such as the Hoover Wind Tunnel and the 9A Eureka Mighty-mite (Julien et al., 2008; Obendorf et al., 2006). In all studies that used vacuum samplers, the samples were taken by vacuuming a specified area with overlapping path lengths that captured dust particles into a container.

A polyurethane foam (PUF) roller was used in a study by Nishioka et al. (2001) to collect dust samples from floors suspected of herbicide contamination from 2, 4-dichlorophenoxyacetic acid (2, 4-D). The PUF roller is an aluminum wheel that has a PUF sleeve placed over it and then is rolled back and forth over a selected 100 cm path (Billets, 2007). These PUF sleeves were pre-cleaned before use with a water and a buffer solution (Nishioka et al., 2001).

In the Minnesota Children's Pesticide Exposure Study (MNCPES), a comparison of two different surface sampling methods was evaluated to determine estimates of pesticide exposure (Lioy et al., 2001). The pesticides malathion, atrazine, diazinon, and chlorpyrifos were assessed using the Edwards and Lioy (EL) sampler and the Lioy, Wainman, and Weisel (LWW) surface wipe sampler (Billets, 2007; Lioy et al., 2001). The EL sampler uses carbon-18 (C18) filters and is a press sampler that collects dust from carpets or hard surfaces. In the Lioy et al. (2001) study, the EL sampler collected dust from carpet and flat living room surfaces of 150 cm^2. The LWW sampler uses C18 filters impregnated with Teflon and wetted with IPA. The filters are then placed on the LWW pressure plate and slid over approximately a 100 cm^2 area three times. The EL sampler was determined to provide the best representation of SDRs available for a single hand pressed onto a surface, one time, while the LWW sampler only measured total collectible pesticides on a surface (Lioy et al., 2001).

In the EPA's literature review by Billets (2007), direct sensing techniques are mentioned. The portable photoionization monitor was one of the instruments, in particular, that can detect organic compounds such as pesticides. Although, they did not research any more instruments or go into great detail, due to the limitations associated with direct sensing equipment. These limitations include higher limits of detection, analyte selectivity, and only qualitative results (Billets, 2007). Due to these limitations with direct sensing equipment, the EPA stated that direct sensing instruments should be used with wipe sampling or replaced with wipe sampling methods altogether.

Results of Literature Review

Controlled Laboratory Studies

There were five articles reviewed that used surface wipes in a controlled laboratory or a controlled environment setting. Four of these articles examined surface wipe sampling techniques to determine their efficacy to collect pesticide residue from hard surfaces (Bernard et al., 2008; Cettier et al., 2015; Deziel et al., 2011; Mohan & Weisel, 2010). One of the articles used surface wipes to study the movement and formation of pesticide degradates from indoor applications of pesticides (Starr et al., 2014). The following section will concisely summarize the methods applied for surface sampling.

A comparison study by Bernard et al. (2008) evaluated the efficiencies of a modified press sampler against solvent moistened gauze pads to collect pesticide residues from different surface types. Four surfaces were tested: ceramic tile, vinyl tile, hardwood flooring, and low pile carpet. The three hard surfaces were grouped together under smooth, hard, non-porous surfaces due to their low differences in mean transfer efficiencies. For this study a spray chamber was fabricated and calibrated to administer a uniform spray onto surfaces at 100 ng/cm^2 and 1000 ng/cm^2. The press sampler was constructed out of Teflon with a sampling block that held two disks with a diameter of 8.5 cm. This study used three disk types for the modified press sampler a C18, PUF, and a 100% cotton disk. More detailed information on the C18 and PUF material is referenced in the general wipe methods section under miscellaneous notes on other surface sampling methods. Wipe sampling consisted of four non-sterile cotton 10 cm x 10 cm, 12-ply gauze pads to wipe a predefined area of 8.5 cm x 8.5 cm. The first gauze pad was laced with 10 ml of IPA, then the area was wiped in a single horizontal direction from left-to-right, and then repeated with a fresh portion of the same pad. A second wipe not laced with IPA was then wiped in the same single horizontal direction as previously described until the area was dry and both horizontal wipes were combined and stored. The third and fourth wipes were applied in the same manner except the single wiping direction was vertical from top-to-bottom and the vertical wipes were combined for storage. The results of this comparison study found that the IPA moistened gauze pads were significantly more efficient than the press sampler for hard surfaces and carpet. Also, the moistened gauze pads had the ability to remove every type of pesticide applied in this study while the press sampler did not. The following Table I represents the mean transfer efficiency range % from the surfaces sprayed.

Table I – Mean Transfer Efficiency Range % From Sprayed Surfaces

	Hard Surfaces 100 ng/cm^2	**Hard Surfaces 1000 ng/cm^2**	**Carpet 100 ng/cm^2**	**Carpet 1000 ng/cm^2**
C18	3-55%	3-35%	<1-12%	<1-3%
Cotton	2-27%	2-9%	<1-13%	<1-3%
PUF	1-30%	2-10%	<1-18%	<1-4%
Wipe	84-97%	92-100%	31-39%	29-38%

Note. Adapted from "Sampling household surfaces for pesticide residues: Comparison between a Press Sampler and solvent-moistened wipes," by Bernard, C. E., Berry, M. R., Wymer, L. J., & Melnyk, L. J, 2008, *Science of the Total Environment,* 389, 514-521. Copyright 2008 by Elsevier B.V.

Cellulose wipes were evaluated for their efficiency and repeatability in the determination of pesticide and PCBs on three surface types in a study by Cettier et al. (2015). This study performed three experiments: 1) assess direct spiking to three sample surfaces, 2) assess the capacity of synthetic dust to be collected by cellulose wipes, and 3) assess spiked synthetic dust collected from the tile. The cellulose wipes 11 cm x 21 cm were pre-cleaned with 99.9% purity dichloromethane, dried, and then stored in a glass jar until sampling. Before each sample was taken each pre-cleaned cellulose wipe were soaked in 10 ml of IPA in a beaker. Three sample surfaces were used - tile, laminate, and hardwood, each with a sample area of 40 cm x 40 cm. All samples were taken with two wipes using a Z-shape pattern while making small circular motions. The first wipe was from left-to-right and then top-to-bottom followed by a second wipe with movement from right-to-left and then bottom-to-top. Both wipes were then placed into a sterile Pyrex Erlenmeyer flask and stored at 4 ^{o}C. The synthetic dust used in this experiment was ASHRAE 52/76 that was comprised of 23 % black carbon, 72 % mineral dust, and 5 % cotton linters. From the results of their experiments, it was found that cellulose wipes had a good overall collection efficiency for a significant amount of pesticides and PCBs when adsorbed onto synthetic dust. Hard and smooth surfaces were identified as the ideal sample surface for this method. Table II displays the mean % collection efficiency for cellulose wipes.

Table II – Overall % Mean Collection Efficiency for Cellulose Wipes for Pesticides and PCBs

	Tile	**Laminate**	**Hardwood**	**Tile with Dust**
% Mean Collection Efficiency	38	40	34	72

Note. Adapted from "Efficiency of wipe sampling on hard surfaces for pesticides and PCB residues in dust," by Cettier, J., Bayle, M., Béranger, R., Billoir, E., Nuckols, J. R., Combourieu, B., & Fervers, B, 2015, *Science of the Total Environment*, 505, 11-21. Copyright 2015 by Elsevier B.V.

The National Children's Study (NCS) was a multiyear prospective study that examined multiple hazards in a child's environment from birth to the age of 21. One of the hazards examined was concerned with the long-term exposure to pesticides. In an effort to provide the NCS with the best practical sampling method, the study by Dezial et al. (2011) compared wipe sampling materials and wetting agents for the collection of 27 pesticides from hard surfaces. Two wiping materials were chosen a 9" x 9" tightly woven cotton wipe known as a Twillwipe and a 6” x 6” packaged pre-wetted polyvinyl alcohol wipe known as a Ghost Wipe. The Twillwipes were cut into 4.5” x 4.5” sections and pre-cleaned with superclean water, IPA, and hexane to extract any contaminants. Before sampling, the dried pre-cleaned Twillwipes were wetted with either 2 ml of 99.8% purity IPA or 2 ml of DI water. Ghost Wipes were pre-wetted with 4 ml of DI water by the manufacturer and did not require any pre-treatment. Stainless steel tool wrap was chosen as the sampling surface and an area of 1 ft^2 (929 cm^2) was sampled. For all wipe samples collected, an overlapping S-shape pattern was used first from left-to-right, the wipe was folded in half and the same area was wiped in an up-and-down motion, then folded in half again to wipe the perimeter of the sampling area, and finally stored in a 60 ml amber glass jar with a Polytetrafluoroethylene (PTFE) lined lid at -20 ^{o}C. A total of 27 pesticides in low and high concentrations were used to spike the surface. The study concluded that the Twillwipe wetted with IPA was generally recommended due to its collection efficiency, accuracy, and precision. Although, it was noted that this method requires multiple preparation steps and can present challenges for large-scale studies. The following Table III displays the high and low spike mean % collection efficiency of wipes for pesticides.

Table III – High and Low % Mean Collection Efficiency of Wipes for Pesticides

	High Spike % Mean Collection Efficiency	**Low Spike % Mean Collection Efficiency**
Twillwipe with IPA	69.3	55.6
Twillwipe with DI Water	10.3	6.9
Ghost Wipe	31.1	22.5

Note. Adapted From “Comparison of wipe materials and wetting agents for pesticide residue collection from hard surfaces,” by Deziel, N. C., Viet, S. M., Rogers, J. W., Camann, D. E., Marker, D. A., Heikkinen, M. S. A., … Dellarco, M, 2011 *Science of the Total Environment*, 409(20), 4442–4448. Copyright 2011 by Elsevier B.V.

Aircraft disinsection is the process of eliminating insects from international flights through the application of insecticides. The application of insecticides inside commercial aircraft is routinely required by some countries and airlines. This raises a concern for dermal absorption of insecticides by the crew and passengers of these aircraft. A study by Mohan & Weisel (2010), used an economy row of three airline seats in a controlled environment to evaluate the efficiencies of multiple wipe sampling techniques to characterize the pyrethroid, permethrin. By spraying a uniform amount of permethrin on the desired area researchers could determine the amount recovered by each wipe sampling method deployed. An LWW sampler, previously described under the general wipe sampling information section, was used with either a polyethylene drain disc filter or a cotton cloth. IPA and water were the wetting agents for the polyethylene disc and water was the only wetting agent used for the cotton cloth. On hard and soft surfaces, the LWW was wiped across a 100 cm^2 area wetted with IPA or water. Whatman circle filter paper measuring 9 cm in diameter was placed on hard surfaces, sprayed with 0.7 ml of water, rotated by 90^o, and then transferred to a container. This procedure was repeated with a second Whatman filter and any residual liquid was wiped with a third dry Whatman circle filter. For soft surfaces the Whatman filter was wetted with water and an area was pressed or blotted with the filter. All samples from this study were kept refrigerated until analysis. From this study, it was concluded that the LWW sampler and the Whatman filters can be used to estimate dermal exposure to individuals on airline seats for extended periods. Table IV displays the % recovery of permethrin from aircraft surfaces.

Table IV - % Recovery of Permethrin from Hard and Soft Aircraft Surfaces

	LWW with IPA	LWW with Water	Cotton LWW with Water	Whatman Circle Filter with Water
Hard Surfaces % Recovery	89.7	45.6	74.1	89.5
Rug % Recovery	N/A	14.0	20.0	70.00
Seat % Recovery	N/A	14.0	N/A	40.1

Note. Adapted from "Sampling scheme for pyrethroids on multiple surfaces on commercial aircrafts," by Mohan, K. R., & Weisel, C. P, 2010, *Journal of Exposure Science and Environmental Epidemiology*, 20(4), 320-325. Copyright 2010 by Nature Publishing Group
N/A = Not Available

Pesticide degradates have been used as biomarkers in human urine to assess the estimated exposure or dose to the parent pesticide. Starr et al. (2014) performed a study that aimed to characterize the formation and movement of pesticides and their degradation products from their application in a controlled test house. This study was done over a 5-week period in a test house that was a single-story, unoccupied, and unfurnished. Four pesticides cypermethrin, permethrin, propoxur, and fipronil were mixed, applied according to the individual product labels, and delivered into fabricated cracks and crevices. The cracks and crevices were fabricated as wooden slats (0.5" x 0.75" x 48") with a 48" crack made lengthwise down the slats and then attached to the living room wall just above the floor. Sampling media consisted of two 4" x 4" woven cotton wipes that were pre-cleaned with IPA and hexane. Samples were taken from the living room and adjacent den floors using a distinct sampling area of three by four square rows comprised of twelve 929 cm^2 templates. Prior to sampling, each wipe was laced with 6 ml of IPA and then the areas were wiped, sequentially, using an overlapping pattern until the area was completely wiped. Both wipes that were used for each sampling area were combined and stored in an amber glass jar at -20 ^oC. The analysis of the wipe samples combined the data from the living room and den. Spiked clean wipes were prepared with known amounts of analyte to establish accuracy and precision of the sampling process. Recovery from clean spiked wipes ranged from 89 ± 9% to 105 ± 9%. In this study, they found that pyrethroids and fipronil had some indoor movement but the amount was small and not practical for describing indoor movement. However, propoxur was readily transported from the application site to flooring in both rooms. In the 5-week period, no significant formation of the degradation products from the applied parent pesticide was evident. A summary of the surface wipe sampling methods applied in these studies are at the end of this chapter in Table V.

Agricultural Worker Housing Exposure Studies

There are a variety of occupational settings where humans are exposed to pesticides, these include agriculture, pest control, florists, and hazardous waste treatment facilities. However, in recent years there has been a concern for take-home exposures to the family members of those who work in agriculture. Multiple studies in recent years have relied on a variety of techniques, including wipe samples, to characterize the presence of pesticides and their degradation products that may have been tracked in from these workers. The methods for wipe sampling from five published articles related to agricultural pesticide exposure were reviewed and the following is a concise summary of the surface sampling methods.

Migrant farmworkers are exposed to a broad range of different pesticides. These workers have the greatest potential for carrying home unwanted pesticides and contaminating their homes. In a study by Acury et al. (2014), they described the presence of organophosphate (OP) and pyrethroid pesticides in migrant farmworker camps in North Carolina. Wipe samples were taken from two uncarpeted areas in the entry way of the bedrooms and one from the common area. For each sample a 50 cm x 50 cm area was wiped with two sterile 4” x 4” dressing gauze, each laced with 2 ml of pesticide grade IPA. All gauze pads were then placed into an amber glass bottle and placed into a cooler. Field blanks and matrix spikes were made on selected days of sampling, field blanks consisted of clean gauze pads placed in jars, and matrix spikes were similar, except known amounts of pesticides were added. Analysis of field blanks spiked with internal standards contained trace levels (<1 ng) of bifenthrin, allethrin, tefluthrin, chlorpyrifos, and 19 ng of resmethrin. Acury et al. (2014) corrected their results from the blank levels and matrix spikes. Their findings indicated that the levels of OP pesticides were not associated with normal camp characteristics, while the levels of pyrethroids were. This information shows that these migrant farmworkers continue to be exposed to pesticides in their homes.

A study to test field methods was performed by Bradman et al. (2007) to describe pesticide exposures to 20 farmworkers' children in Salinas Valley, California. Methods were tested for collecting the following: house dust, indoor and outdoor air, SDRs from surfaces and toys, residues on clothing, food, and urine samples. A teething ring (area: 99.2 cm^2) and a ball (area: 283.5 cm^2) were provided to the children 1.5 days before sampling. Wipe samples were taken from hard surfaces of the kitchen or dining area near the carpet boundary. An area of 30 cm x 30 cm and the provided toys were wiped thoroughly with a 10 cm x 10 cm Johnson & Johnson SOF-WICK rayon dressing sponges wetted with reagent-grade IPA. Field matrix spikes were fortified with all analytes, recoveries for pyrethroids in all matrix spikes averaged 87%, while recoveries for OPs averaged 76%. An EL press sampler with C18 disks was used but it did not detect any pesticides. Bradman et al. (2007) found measurable levels of organochlorine (OC), OP, and pyrethroid pesticides in house dust, indoor and outdoor air, surface samples, clothing, and food. However, the house dust, surface wipes, and clothing samples found the largest variety and amount of pesticides compared to other media. Bradman et al. (2007), stated that these media may be the best for characterizing what type of pesticides are present in a home.

In order to study and compare agricultural pesticide contamination in homes, Curwin et al. (2007) evaluated 25 farm households and 25 non-farm households in Iowa. For homes to be eligible for the study, each home had to have at least one child 8 years old or younger, farm households had to be located on farmland, and non-farm households had to be located on land not used for farming. Methods for sample collection in this study included questionnaires, air samples, dust samples, and wipe samples. Surface wipe samples were collected from the kitchen counter, top of the washing machine, steering wheel and driver seat of the primary vehicle, and various hard floors in the homes. The wipe sampling areas were 1 ft x 1 ft and two 4" x 4" Johnson & Johnson SOF-WICK sponges were used. The first sponge was wetted with 10 ml of 100% IPA and wiped the designated area with four adjacent overlapping wipes in one direction, folding the sponge after each wipe to reveal a clean surface for each pass. The second sponge was implemented in the same manner but in a perpendicular direction to the first sponge. Both sponges were placed in an amber jar and covered with a Teflon-lined cap. The steering wheel was wiped with one sponge wrapped around the steering wheel and half the wheel was wiped. A second sponge was wrapped around the steering wheel and wiped the second half. The sponges were prepared and stored in a similar fashion as previously stated. PUF sponges wetted with 6 ml of IPA were also used to sample for glyphosate and 2, 4-D. The PUF sponges followed the same procedures as the Johnson & Johnson SOF-WICK sponges. The percent recoveries for the surface wipes ranged from 90-103%. Curwin et al. (2007) found that farm homes had higher average pesticide levels than non-farm homes. The herbicides atrazine and metolachlor were suspected of being brought into farm homes by the farmer's shoes and clothing since these herbicides were not applied in or around the homes.

In central Washington State, children in an agricultural community were evaluated for exposures to OP pesticides azinphos-methyl and phosmet in a study by Lu et al. (2000). The children's exposures were assessed through urine, hand wipe, house dust, and surface wipe samples. Each child's household was categorized according to their parent's occupation (agricultural or nonagricultural) and by proximity to pesticide treated orchards. Surface wipe samples were taken from vehicle steering wheels used to travel to work, work boots, and non-carpeted flooring where children played. Sampling procedures consisted of two sterile 4" x 4" all-cotton gauze pads wetted with 1-2 ml of 100% IPA. Non-carpeted floor areas were measured with a 50 cm x 50 cm template, wiped in a sequence of three vertical and horizontal strokes, and the wipes were stored in an ice chest. Work boots were sampled in a similar fashion, but only one 4" x 4" all-cotton gauze pad was used and wiped an area of 5 cm x 5 cm. After wiping the boot surface in three vertical strokes, the wipe was folded to expose a clean area and then wiped the area with three horizontal strokes. The steering wheel sample was prepared in a similar fashion but a template was not used, instead one wipe was used to make a continuous stroke across the top half of the steering wheel. Extraction recoveries from the gauze pads for azinphos-methyl was 117 ± 39% and phosmet was 101 ± 12%. No adjustments were made for the surface wipe results. The results from this study found that pesticide applicator family households had the highest median concentrations of both OP pesticides followed by agricultural workers and then nonagricultural workers. Homes with closer proximity to pesticide-treated farmland also had higher exposure levels than those of the reference families.

As stated earlier, a growing concern for take-home exposure of pesticides to farmworker families has risen, especially for those with young children. A study by Quandt et al. (2004) evaluated 41 farmworker homes in North Carolina and Virginia for eight agricultural pesticides reported to be used locally and thirteen pesticides commonly found in U.S. homes. All homes in this study had at least one child < 7 years of age. Data collection methods were comprised of a questionnaire, wipe samples, and observations of the family and neighbors. Wipe samples were taken from uncarpeted floors, toys and children's hands according to protocols described in Geno et al. (1996) and Harding et al. (1993). The floor samples were taken from two to four 18" square sections from the living areas of each home, two or three suitable toys identified as most frequently handled were wiped, and children's hands were wiped with two sponges. Field blanks were collected at eleven of the homes that were comprised of two sponges wetted with 15 ml of IPA. Before field blanks were sealed with Teflon tape and placed into a cooler, an additional 50 ml of IPA was added. Floor samples were found to be repeatable from the 34 pairs of collocated duplicate floor samples with detectable loadings. From these pairs 59% had differences < 20% and 94% had differences < 50%. Matrix spikes fortified with all analytes had a mean recovery efficiency ranging from 77 ± 11% to 137 ± 42%. Degradation of the spiking solution and extraction efficiency was suspected, a new spiking solution was used for the final batch samples, and had a recovery efficiency of 62%. Results from this study found that the floors of farmworker homes may be a reservoir for pesticide residue. A summary of the surface wipe sampling methods applied in these studies are at the end of this chapter in Table VI.

Residential and Urban Housing Exposure Studies

Pesticides are commonly used inside and outside the household to control different types of insects. It was estimated that approximately 74% of households in the U.S. in 2007 used pesticides in and around their homes (EPA, 2011). Unlike residential homes, urban dwellings are often comprised of multiple units and the usage of pesticides can sometimes be excessive due to pest infestations or over usage by multiple tenants. In 2014, there were 82,459 cases of pesticide poisonings reported in the U.S. to the American Association of Poison Control Centers (AAPCC) and of those cases 34,196 involved children under 5 years old (Mowry, 2015). The following section reviews the wipe sampling methods applied in eight studies. Seven of the studies evaluated pesticide exposure in residential or urban housing. An additional study was included in this section that surveyed child care centers for pesticides.

From January 2009 to September 2010 a study by Boyle et al. (2015), evaluated the feasibility and informative value of environmental sample collection methodology in the pilot phase of the NCS. The NCS was discussed earlier in the controlled laboratory studies section in the study by Deziel et al. (2011). The environmental collection methods evaluated during the pilot phase of the NCS were air, dust, wipe, and water sampling. Wipe sampling methodology involved the use of Ghost Wipes, pre-packaged polyvinyl alcohol wipes wetted with water by the manufacturer. Samples were collected from a 1 ft^2 area on hard floor surfaces in the most commonly used room or kitchen. Results from this study found that their wipe sampling methodology was very feasible, however, Boyle et al. (2015) believes that wipe samples could be replaced with dust bulk collection with vacuums that may yield higher detection frequencies.

When using surface wipes to determine exposure estimates from pesticides the cost associated with the analysis of a large number of samples is of great concern. Deciding to analyze a single compound or a particular class such as pyrethroids can affect the level of its detection and specificity during the extraction process. In an effort to analyze a broad suite of chemicals, a study by Clifton et al. (2013) developed a new method for the analysis of wipe samples for selected OPs, pyrethroids, pyrethroid transformation products (TPs), bisphenol A, and polybrominated diphenyl ethers (PBDEs). The wipe sampling procedure consisted of wiping kitchen flooring areas of 48” x 48” and window areas of 40” x 40” with two 4” x 4” cotton Twillwipes wetted with 6 ml of pesticide-grade IPA. Prior to use, the cotton Twillwipes were pre-cleaned with IPA and hexane. Each wipe was stored in a pre-cleaned 60 ml amber jar and placed into a cooler. Quality assurance (QA) and quality control (QC) samples were comprised of field blanks, field controls, field duplicates, storage spikes, storage blanks, methods spikes, and recovery spikes. Method detection limits in this study were calculated based on a 929 cm^2 area for comparability to other studies. Method spike recoveries ranged from 63 ± 15% to 106 ± 24% for OPs and pyrethroids except for malathion (9.5 ± 4.4%) and *l*-cyhalothrin (40 ± 13%). The pyrethroid transformation products ranged between 32 ± 13% to 71 ± 21%. The results of the new analysis method by Clifton et al. (2013), concluded that their method was successful in the ability to accurately measure most of the defined chemicals at trace levels, which were comparable to what was reported in similar studies. Pyrethroid TPs produced inconsistent results in this study that Clifton et al. (2013) attributes to chemical interferences.

The Healthy Public Housing Initiative (HPHI) is a longitudinal intervention study of pediatric asthmatics who live in urban apartment dwelling that is between the ages of 4 and 17. Julien et al. (2008), investigated the level and distribution of pyrethroid and OP pesticide loadings for dust and wipe samples from 42 urban dwelling in Boston, Massachusetts. During each visit, they performed environmental measurements via floor wipes and vacuum samples in conjunction with a quality of life inspection related to asthma. The floor wipes were taken from vinyl floor surfaces in the kitchen adjacent to the stove, in the living room adjacent to the sofa, and hallway adjacent to the linen closet. Wipe sampling involved a 3 in^2 Johnson & Johnson sterile gauze wetted with 5 ml 99% IPA that wiped a 929 cm^2 area. Each sample was stored in an amber glass jar and placed in a cooler. QA and QC samples were comprised of field matrix blanks and matrix spikes. The average matrix spike recovery was 98% with a range of 86 ± 9%

for cypermethrin isomers to 112 ± 13% for chlorpyrifos. Surrogate standard recoveries (SRSs) displayed a good method performance with recoveries of 78 ± 15% for fenchlorphos and 91 ± 25% for C_6-*trans*permethrin. Results found a concerning level of cyfluthrin that is the active ingredient in the insecticide product known as Tempo. The investigators in Julien et al. (2008), discovered that the tenants were applying the concentrated form of Tempo without mixing it with water as per labeling instructions. Compared results of dust and wipe sampling concluded that there were positive correlations between vacuum dust and floor wipe samples.

A community-based participatory research (CBPR) project by Lu et al. (2013), enrolled 20 families with young children living in low-income urban housing in order to assess residential exposures to pesticides. The goal of this research was to encourage low-income housing communities to participate in an integrated pest management program in order to improve their quality of life, health, and household conditions. The environmental exposure assessments were composed of surface wipe and indoor air samples. Surface wipe samples were collected from the living room floor, children's bedroom floor, and kitchen countertops. All floor areas were covered in vinyl tile and were areas most likely to be encountered by children. Two 3" x 3" sterile cotton gauze pads were wetted with about 2 ml of IPA and wiped an area of 30 cm x 30 cm with three sequential vertical and horizontal strokes. Both sterile cotton gauze pads were then placed into a jar and put in a cooler until analysis. QC samples consisted of blank samples prepared by adding an internal standard to new gauze pads. Blank control samples contained trace levels, 1 ng per sample, of bifenthrin, allethrin, tefluthrin, chlorpyrifos, and 19 ng of resmethrin. QA samples were prepared by spiking gauze pads at two different levels 50 ng and 500 ng along with the internal standard to determine recovery efficiencies. For OP recovery efficiencies the 50 ng spikes ranged from 112-117% and the 500 ng spikes ranged from 80-

138%. For pyrethroid recovery efficiencies, the 50 ng spikes ranged from 110-136% and the 500 ng spikes ranged from 60-124%. The results from surface wipe samples found six pyrethroids and five OP pesticides at quantifiable levels.

The distribution of pesticide residues was assessed within rural farm homes, rural non-farm homes, and urban houses from central New York State in a study by Obendorf et al. (2006). Surface wipe samples were taken from smooth floors, tables, shelves, and windowsills. Smooth floor sample areas measured approximately 0.392 m^2 and were wiped with Whatman filter paper wetted with a 70/30 (by volume) mixture of methanol and DI water. The floor areas were wiped with four strokes left and right, then back and forth, and sealed in a glass container and put on ice. Flat surface samples, such as tables, shelves, and windowsills were wiped in the same manner as the smooth floors except the area was measured and wiped in only two directions. This study extends the work done in Lemley et al. (2002). From their results, Obendorf et al. (2006) concluded that pesticide residue distribution varies with seasons, is more prevalent in carpets, and redistributes within the household by airborne routes.

The EPA and the U.S. Department of Housing and Urban Development (HUD) collaborated on a national survey of homes to identify environmental hazards in U.S. residences. This study is known as the American Healthy Homes Survey (AHHS) and its goal was to evaluate lead, allergens, and insecticides in a nationally representative sample of homes. Stout et al. (2009) presents the data related to insecticide loadings from surface wipe samples collected in the AHHS. Samples were randomly selected from the 1131 homes of which a subset of 500 homes was selected for surface wipe sampling. The sampling procedure consisted of two Excilon brand 10 cm x 10 cm surgical sponges composed of rayon microfibers wetted with 6 ml of analytical grade IPA each. Excilon sponges were pre-cleaned with dichloromethane followed by hexane and then dried before sampling. Floor surface areas were taken from two locations within the kitchen or other areas with a 929 cm^2 aluminum template. One sponge per location was wiped in an "S" or "Z" pattern from side-to-side folded, then wiped top-to-bottom, and both sponges were stored in a jar on ice. QC was comprised of field blank media, blind blank media, blind spiked media, laboratory solvent blanks and spikes, and laboratory media blanks and spikes. The QC results had elevated background concentrations of select insecticides that Stout et al. (2009) accredits to accidental use of field blanks to sample surfaces. Field blanks with >100 ng of any analyte were omitted from the report. Results from the AHHS suggest that insecticides used in homes can be found in measurable amounts long after application. Further evidence of this is supported by the measured amounts of insecticides known to have been removed from the market.

With the growing concern of pesticide exposure to children, a joint project was undertaken by the EPA, HUD, and U.S. Consumer Product Safety Commission. The First National Environmental Health Survey of Child Care Centers set out to characterize the environmental hazards of young children by measuring lead, allergens, and pesticides in a random nationally representative sample of child care centers. The data concerning pesticide results are presented in a study by Tulve et al. (2006). Surface wipe sampling was collected from a floor area where children spent a significant amount of time and a desk or tabletop that the children used. Both floor and surface samples were taken from an area of 929 cm^2 and a sterile non-woven gauze dressing sponge (100%, 4" x 4", 6-ply, Johnson & Johnson SOF-WICK) was wetted with 10 ml of high purity IPA. The gauze wiped the designated area using an "S" shape from left-to-right and then flipped inside-out and wiped right-to-left. A second gauze sponge wiped the same area in a similar manner except from top-to-bottom, bottom-to-top, and one final wipe around the perimeter. Both dressings were placed in a certified pre-cleaned glass jar. This study developed and validated a multi-residue analysis method for their particular target analytes. The limits of detection for this method can be seen in Table VII. Field controls were performed as matrix spikes and the average recoveries were >80% with a range from 81-137% and the average recovery for diazinon was 65%. The results of the survey demonstrate the potential for children's exposure to pesticides in child care centers, but cannot quantitatively estimate exposures due to ancillary data needed for exposure algorithms.

Tulve et al. (2008), performed a pilot study as a component of a much larger joint study by the EPA, CDC, and Duval County Health Department in Florida. The pilot study evaluated pesticide exposures to nine young children in their homes via air, dust, food, and urine samples. Dust samples were collected with surface wipes from the main play area of the home and where the guardian reported past pesticide applications. Wipe samples were collected from an area of 929 cm^2 with a sterile non-woven gauze dressing sponge (100%, 4" x 4", 6-ply, Johnson & Johnson SOF-WICK) wetted with 10 ml of IPA. Wipe sampling procedures followed previous methods described in the study by Tulve et al. (2006). Field blanks measured were low and all wipe samples were corrected for field blank levels. Matrix spikes were created for eleven target pesticides and recoveries averaged from 74% to 131%. Wipe sample results suggested that multiple different pyrethroid products were used in these sample areas with pyrethroid concentrations higher than OPs. This also suggested that pyrethroid pesticides are more likely to stick to surfaces and dust particles. The following tables are a summary of the surface wipe sampling methods applied in all of the studies. Table VI is the summary of surface wipe sampling methods from the urban and residential housing exposure measurement studies.

Tables

Table V - Summary of Surface Wipe Sample Methods from Controlled Exposure Measurement Studies

Reference	Wipe Material	Pre-Treatment of Wipes	Wetting Solvent	Surface Type	Wipe Pattern	Storage	Analytical Method	Pesticide Class
(Bernard et al., 2008)	Cotton gauze pads 10 cm × 10 cm	N/A	10 ml of IPA	Collection area 8.5 cm x 8.5 cm (ceramic tile, hard wood, vinyl tile) and low pile carpet	Hard surfaces: Horizontal from left-to-right and vertical from top-to-bottom	N/A	Gas Chromatograph/Mass Spectrometer	Organophosphate, Pyrethroid, Phenylpyrazole
(Cettier et al., 2015)	11 cm x 21 cm cellulose wipes	DCM	10 mL of IPA	Collection area 40 cm x 40 cm (tile, laminate, and hardwood)	Small circular motion in a Z-shape; stored in sterile Pyrex Erlenmeyer flask at 4ºC	Sterile Pyrex Erlenmeyer flask at 4ºC	Gas Chromatograph/Mass Spectrometer and High Performance Liquid Chromatography/Mass Spectrophotometer	Organochlorine, Triazole, Organophosphate, Pyrethroid, Carbamate, Chloroacetamid, Chloroacetamide, Morpholine, Straobilurin, Anylinopyrimidine, Benzoylurea, Cyanoacetamide, Oxime, Dinitroanilinine, Dinitrophenol, Neonicotinoid, Phenol, Phenylpiridinamine, Phenylpyrazole, Phenylpyrrole, Phenylurea, Phtalimide, Piperonyl

Table V - Summary of Surface Wipe Sample Methods from Controlled Exposure Measurement Studies (cont.)

Reference	Wipe Material	Pre-Treatment of Wipes	Wetting Solvent	Surface Type	Wipe Pattern	Storage	Analytical Method	Pesticide Class
(Deziel et al., 2011)	9" x 9" Twillwipe sectioned into 4.5" x 4.5" and Pre-packaged Ghost wipes 6" x 6"	Twillwipes cleaned with superclean water, IPA, and hexane	2 mL of IPA or DI water	Collection area 1ft^2 (stainless steel)	Overlapping S-shape	Amber glass jar at -20 ºC	Gas Chromatograph/Mass Spectrometer	Organochlorine, Pyrethroid, PhenylPyrazole, Piperonyl
(Mohan & Weisel, 2010)	Circular Whatman filter	N/A	0.7 mL water	Collection area 100 cm^2 (hard surfaces, rug, seat cushion)	Sprayed with water, rotated 90º	Stored in refrigerator.	Gas Chromatograph/Mass Spectrometer	Pyrethroid
(Starr et al., 2014)	Woven cotton wipes 4" x4"	IPA and hexane	6 mL IPA	Collection area 929 cm^2 uncarpeted Floor	Overlapping pattern	Amber Jars stored at -20ºC	Gas Chromatograph/Mass Spectrometer	Phenylpyrazole, Carbamate, Pyrethroid, Organophosphate

N/A = Not Available
DCM = Dichloromethane

Table VI - Summary of Surface Wipe Sample Methods from Agriculture Housing Exposure Measurement Studies

Reference	Wipe Material	Pre-Treatment of Wipes	Wetting Solvent	Surface Type	Wipe Pattern	Storage	Analytical Method	Pesticide Class
(Arcury et al., 2014)	Sterile 4” x 4” dressing gauze	N/A	2 ml of pesticide grade IPA	Collection area 50 cm x 50 cm uncarpeted Floor	Unspecified wipe pattern	Sterile amber glass bottles in a cooler	Gas Chromatograph/Mass Spectrometer	Organophosphate and Pyrethroid
(Bradman et al., 2007)	10 cm x 10 cm Johnson and Johnson SOF-WICK rayon dressing sponges	N/A	10 mL reagent-grade IPA	Collection area 30 cm x 30 cm uncarpeted floor	Unspecified wipe pattern	Stored on ice	Gas Chromatograph/Mass Spectrometer	Organophosphate, Pyrethroid, Organochlorine, Herbacide, Fungacide
(Curwin et al., 2005)	Two 4" x 4" Johnson and Johnson SOF-WICK sponges	N/A	10 mL of IPA	Collection area 1 ft x 1 ft (uncarpeted floor, steering wheel, and washing machine)	Four adjacent overlapping wipes perpendicular to each other	Stored in amber glass jar	Gas Chromatograph/Mass Spectrometer	Organophosphate, Triazine, Chloroacetanilide, Herbacide

Table VI – Summary of Surface Wipe Sample Methods from Agriculture Housing Exposure Measurement Studies (cont.)

Reference	Wipe Material	Pre-Treatment of Wipes	Wetting Solvent	Surface Type	Wipe Pattern	Storage	Analytical Method	Pesticide Class
(Lu et al., 2000)	Sterile 4" x 4" all-cotton gauze pads	N/A	1-2 mL 100% IPA	Collection area 50 cm x 50 cm (uncarpeted Floor, work boots, steering wheel)	3 vertical and horizontal strokes	Stored in cooler	Gas Chromatograph/Mass Spectrometer	Organophosphate
(Quandt et al., 2004)	Sponges	N/A	N/A	Collected from two to four 18" square sections (0.42-0.84 m^2) for uncarpeted floors	Unspecified wipe pattern	N/A	Gas Chromatograph/Mass Spectrometer	Organophosphate, Pyrethroid, Organochlorine, Carbamate, Herbacide

N/A = Not Available

Table VII - Summary of Surface Wipe Sample Methods from Urban and Residential Housing Exposure Measurement Studies

Reference	**Wipe Material**	**Pre-Treatment of Wipes**	**Wetting Solvent**	**Surface Type**	**Wipe Pattern**	**Storage**	**Analytical Method**	**Pesticide Class**
(Boyle et al., 2015)	Ghost Wipes pre-packaged polyvinyl alcohol wipes	N/A	Pre-wetted with water by manufacturer	collection area 1-ft^2 uncarpeted floor	Unspecified for pattern	N/A	Gas Chromatograph/Mass Spectrometer	Organophosphate, Pyrethroid, Phenylpyrazole, Organochlorine, Insecticide synergist
(Clifton et al., 2013)	4" x 4" cotton Twillwipes	IPA and hexane	6 mL of IPA	Collection area 48" x 48" kitchen and 40" x 40" main living area uncarpeted floor and windows	Unspecified pattern	Stored in pre-cleaned amber jars in cooler	Gas Chromatograph/Mass Spectrometer	Organophosphate and Pyrethroid
(Julien et al., 2008)	58 cm2 Johnson & Johnson sterile gauze	N/A	5 mL of IPA	Collection area 929 cm2 Vinyl floor	Unspecified pattern	Stored in amber jars in cooler	Gas Chromatograph/Mass Spectrometer	Organophosphate and Pyrethroid
(Lu et al., 2013)	3" x 3" sterile cotton gauze pads	N/A	~2 mL IPA	Collection area 30 cm x 30 cm uncarpeted floor and kitchen counter	3 vertical and horizontal strokes	Stored in cooler	Gas Chromatograph/Mass Spectrometer	Organophosphate and Pyrethroid

Table VII – Summary of Surface Wipe Sample Methods from Urban and Residential Housing Exposure Measurement Studies (cont.)

Reference	Wipe Material	Pre-Treatment of Wipes	Wetting Solvent	Surface Type	Wipe Pattern	Storage	Analytical Method	Pesticide Class
(Obendorf et al., 2006)	Whatman filter paper	N/A	70/30 (by volume) methanol and DI water	Collection area 0.372 m2 uncarpeted floor, tables, shelves and windowsills	Floor: 4 strokes left-to-right and back-and-forth; Flat surfaces: wiped in two directions	Stored in glass container in cooler	Gas Chromatograph/Mass Spectrometer	Organophosphate, Pyrethroid, Triazine, Chloroacetanilide, Carbamate, Aryloxyalkanoic Acid, Benzoic Acid, Cloroacetanilide, Pyridinecarboxylic Acid, Dintroaniline
(Stout et al., 2009)	10 cm x 10 cm Excilon rayon surgical sponges	DCM and hexane	6 mL of IPA	Collection area 929 cm2 uncarpeted Floor	Side-to-side S or Z pattern	Stored in jar in cooler	Gas Chromatograph/Mass Spectrometer	Organophosphate, Pyrethroid, Organochlorine, Phenylpyrazole, Insecticide synergist
(Tulve et al., 2006)	4" x 4" Johnson and Johnson SOF-WICK rayon dressing sponges	N/A	10 mL of IPA	Collection area 929 cm2 uncarpeted floor, desk, tabletop	S-shape pattern wiping left-to-right and right-to-left	Stored in clean amber jar	Gas Chromatograph/Mass Spectrometer	Organophosphate, Pyrethroid, Phenylpyrazole, Insecticide synergist
(Tulve et al., 2008)	Johnson & Johnson SOF-WICK rayon dressing sponges	N/A	10 mL of IPA	Collection area 929 cm2 uncarpeted floor	Unspecified for pattern	N/A	Gas Chromatograph/Mass Spectrometer	Pyrethroid, Pyrethrin, Pipernyl, Organophosphate, Phenylpyrazole

N/A = Not Available
DCM = Dichloromethane

Discussion

Comparison of Wipe Sampling Methods

A total of eighteen published articles were reviewed that used surface wipes to characterize pesticides in the environment or in a controlled laboratory setting. Similarities and differences in wipe sampling methods for all of the articles regarding wipe materials, pre-treatment of wipes, wetting solvents, wipe surfaces, collection of samples, and storage of wipes are discussed below. In addition, an attached file containing an Excel Spreadsheet with supplemental material has a more detailed summary of the methods from each study including the limits of detection and/or quantification for each pesticide.

In eleven out of the eighteen studies, medical gauze was chosen as the wiping material (Acury et al., 2014; Bernard et al., 2008; Bradman et al., 2007; Clifton et al., 2013; Curwin et al 2005; Julien et al., 2008; Lu et al., 2000; Lu et al., 2013; Stout et al., 2009; Tulve et al., 2006; Tulve et al., 2008). Most medical gauze varies in size, can be woven or non-woven, and be made from cotton, rayon, polyester, or a combination of these fibers. While none of the articles reviewed stated whether their medical gauze was woven or non-woven, three of the articles stated the use of cotton gauze (Bernard et al., 2008; Lu et al., 2000; Lu et al., 2013), four articles stated the use of rayon gauze (Bradman et al., 2007; Stout et al., 2009; Tulve et al., 2006; Tulve et al., 2008), and three articles did not state what the wipe material was composed of (Acury et al., 2014; Curwin et al 2005; Julien et al., 2008). Wipe material sizes in all studies varied from 4” x 4” to 11 cm x 21 cm. The studies that did not use medical gauze used one of the following wiping materials: cellulose wipes, cotton Twillwipes, Whatman filter paper, or pre-packaged Ghost Wipes.

Surface wipe materials may cause matrix interferences during the extraction process. The cleanliness of wipes varies by brand and therefore it would be essential to pre-clean wipes to ensure purity. Only five studies reviewed stated the pre-treatment of their wiping materials. The compounds used to perform their pre-treatment of wipe materials were dichloromethane, superclean water, IPA, hexane, or a combination of these compounds (Cettier et al., 2015; Clifton et al., 2013; Deziel et al., 2011; Starr et al., 2014; Stout et al., 2009). Four out of five of these studies used the Soxhlet extractor for the pre-cleaning procedure and dried their wipes via a vacuum oven.

Many solvents have been used and recommended by various groups and studies. Choosing the appropriate solvent will determine what target pesticides will solubilize onto the sampling material. Most of the studies reviewed used reagent-grade IPA or DI water with the exception of two articles. One article by Obendorf et al. (2006) used a mixture of methanol and DI water and the article by Quandt et al. (2004) did not clearly state what the wetting solvent was. Published articles note that IPA yields a higher collection efficiency for a broader range of pesticides compared to water (Bernard et al., 2011; Billets, 2007; Deziel et al., 2011). IPA in most studies was chosen due to its low toxicity, familiarity, ability to not damage most surface types, and ability to solubilize organic substances. The amount of solvent used to wet wiping material varied from 1 ml to 10 ml in all studies. Deziel et al. (2011), was the only study to determine the maximum amount of solvent that their cotton Twillwipes could absorb without leaving any droplets or liquid behind after wiping a surface. The issue with a heavily wetted IPA wipe is that it is thought to extract chemical residues from within the sampling surface and not just the top of the surface (Billets, 2007).

The collection of pesticide residue from surfaces can vary with the surfaces' characteristics. Most of the studies reviewed that sampled households typically stated that uncarpeted surfaces were sampled, but did not specify what the surface was composed of or provide any descriptive characteristics. A study by Bernard et al. (2008) compared a press sampler and solvent moistened wipes on household surfaces in a controlled laboratory environment. The household surfaces sampled were ceramic tile, vinyl tile, hardwood, and low-pile carpet. Researchers in this study grouped the hard surfaces together due to similar recovery values that only had differences of <22-31% relative standard deviation. However, Cettier et al. (2015) found that roughness and porosity of surfaces, such as hardwood and laminate flooring, reduced collection efficiencies in their study of cellulose wipes. As for carpeting, most agree that an HVS3 or a modified vacuum cleaner is the preferred and superior method for collecting SDRs from carpet (Billets, 2007; Bernard et al., 2008; Julien et al., 2008).

Wipe sampling methods have a huge variability when it comes to collecting samples from surfaces. Almost all studies had variability when it came to the size of the area to be sampled, how to wipe the designated area, how many areas, how many wipes, and where to sample within a dwelling. The sizes of areas sampled from the reviewed studies ranged from 8.5 cm x 8.5 cm to 48" x 48", however, seven studies used a sampling area of 929 cm^2 or 1 ft^2 (Boyle et al., 2015; Curwin et al., 2005; Deziel et al., 2011; Starr et al., 2014; Stout et al., 2009; Tulve et al., 2006; Tulve et al., 2008). Wipe patterns varied with some methods calling for vertical and horizontal strokes, "S" or "Z" wipe patterns, and others simply stated to wipe the area. Most studies also specified that after each wipe the material should be folded to expose a fresh surface for each wipe. In total, four studies used the “S” or “Z” pattern, five performed vertical and horizontal strokes, one sprayed the wipe material with water and rotated it 90^{o}, and one used an overlapping pattern. The remaining seven were categorized as an unspecified collection pattern. The number of areas to wipe and how many wipes to use for each area varied greatly from study-to-study. Most studies would take one sample from areas of high traffic, where children mostly played, or areas suspected of contamination. The number of wipes used per sample area ranged from one wipe to four wipes. Combining wipes from one sample area were common but some studies would combine all wipes taken from the entire household. In almost all studies, samples were stored in a chilled environment. Storage containers for wipes were typically in a glass amber jar or similar and sealed with a Teflon cap or Teflon tape.

Considerations on the use of Wipe Sampling

In order to provide aid to decision-makers in the selection of a method, the following section will outline some considerations based on the following areas concerning the surface wipe method: wipe material, pre-treatment of material, solvent selection, surface types, collection pattern, and storage of wipe material. These considerations are based on the literature reviewed and do not represent all of the knowledge concerning surface wipe methods for pesticide residue.

A majority of the studies used medical gauze because it is easily attainable and affordable. There are some drawbacks that make standardization difficult with store bought medical gauze as a wipe material. These commercially available products are for the general consumers and are susceptible to changes such as thickness, size, texture, composition, and chemical additives. Discontinuation of commercial brands such as the Johnson & Johnson SOF-WICK sponges can also make standardization and comparative studies difficult to undertake.

Less commonly used in the reviewed studies, cotton Twillwipes are widely available and have been standardized by the textile industry (Deziel et al., 2011). Utilizing materials that have been tested and standardized such as the cotton Twillwipes or similar, could reduce variability between studies. The collection efficiencies of cotton Twillwipes were evaluated in a comparison study by Deziel et al. (2011) and the Twillwipes wetted with IPA generally were recommended due to its superior collection efficiency, precision, accuracy, availability, and versatility.

In addition to the selection of wipe material, the pre-treatment of wipe material should be implemented to ensure the absence of any chemicals or contamination so to avoid any interferences during analysis. One may refer to any of the five reviewed studies mentioned in the previous section where pre-treatment of wiping materials was performed (Cettier et al., 2015; Clifton et al., 2013; Deziel et l., 2011; Starr et al., 2014; Stout et al., 2009). In three out of the five studies that used pre-treatment techniques, IPA and hexane were used while the other two studies used DCM and/or hexane.

While there are other solvents available for the collection of pesticide residues, IPA was the most widely used in the reviewed studies. In the controlled laboratory studies, IPA proved to have superior collection efficiencies for a multitude of pesticides across different wiping materials and surfaces (Bernard et al., 2008; Cettier et al., 2015; Deziel et al., 2011; Mohan & Weisel., 2010). When determining the amount of solvent used to wet wiping material one may refer to the study by Deziel et al. (2011), where the amount of solvent that could be completely absorbed by the wiping material was determined. The main issue with the amount of solvent used to wet the wipe is over-wetting that can result in residual solvent left behind on the sampling surface. An alternative to this is to consider using one wetted wipe followed by a dry wipe to recover any residual solvent similar to the method applied in Bernard et al. (2008).

Concerning what surface type to sample, inferences can be made from the collection efficiencies seen in Tables I - IV for the surfaces used in controlled laboratory studies. From these studies' collection efficiencies, it would seem that smooth, hard, and non-porous surfaces are the ideal surface type for the collection of pesticides (Bernard et al., 2008; Cettier et al., 2015; Deziel et al., 2011; Mohan & Weisel, 2010).

There is no overwhelming evidence that states what the optimum sample size area is for wiping a surface. Government agencies such as the EPA, OSHA, and USDA use a 100 cm^2 area to sample for various chemical residues. However, the collection area for seven of the reviewed studies used a 929 cm^2 or 1 ft^2 sampling area with only one study by Mohan & Weisel (2010) using a 100 cm^2 sampling area, similar to government agencies. Sampling the designated area also had huge variability within the studies, while seven of the studies did not specify a wiping pattern the other eleven were almost equally divided between two wipe patterns. These patterns were the "S" or "Z" wipe pattern or the vertical and horizontal strokes.

One area that all studies shared similarity was the storage of retrieved samples. Almost all studies stated storing their field samples in a chilled environment such as a cooler with ice or ice packs. This was done until the samples could be stored properly in a refrigeration unit or freezer. Storage containers consisted either of amber glass jars or certified pre-cleaned glass containers sealed with Teflon tape or a Teflon Cap.

Limitations

Limitations should be considered regarding this literature review of wipe sampling methods for pesticides. Only the databases stated earlier were accessed and only studies in the U.S. conducted between 2000 and 2015 were included. While the qualitative methods of the studies were analyzed, the quantitative data cited were mostly limited to the collection or transfer efficiencies from the controlled laboratory studies.

Future Research

Without a standardized method in place, future studies should consider the state-of-the-art for wipe sampling methods of pesticides. It would also benefit future studies to adopt the most commonly performed and validated method. By doing so, more extensive comparisons of quantitative data can be made to begin furthering our knowledge of characterizing pesticide contamination in our environment.

Conclusion

From this literature review, it is clear that there are more variations between wipe sampling methods of pesticide residue than there are similarities. These variations express the need for a standardized method of sampling surfaces for pesticide residues and SDRs. Although, the similarities between studies such as the use of IPA as a wetting solvent and the storage of field samples placed into glass jars on ice are steps closer to validating a method. While it does not seem that there will be an approved method disseminated anytime soon, professionals seeking to evaluate pesticide residues on surfaces should strive towards carrying out commonly performed or tested methods from controlled studies. Furthermore, without standardization data collected from surface wipes may only indicate whether or not a particular pesticide is present in the environment. Until a standardized method is established, the data collected from surface wipes for establishing human exposure to pesticides will be difficult to substantiate.

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