TODDLERS' INHALATION EXPOSURE TO PERMETHRIN IN HOUSE DUST

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ABSTRACT OF THE DISSERTATION

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The overall objective of this study is to better characterize a toddler's inhalation exposure to permethrin in house dust by conducting the following three studies: 1) measuring permethrin concentrations in a toddler's breathing zone via three different sampling approaches: mobile, stationary and settled dust on vinyl floor and carpeted floor in a simulated indoor environment; 2) identifying the particle size distribution in resuspended dust and settled dust; 3) performing Monte-Carlo simulation to probabilistically estimate toddlers' inhalation exposures to permethrin via the three sampling approaches considering toddlers' time and activity pattern.

The mean permethrin airborne concentrations in the stationary and mobile samples were 0.065 $\mu g/m^3$ and 0.14 $\mu g/m^3$ for the vinyl floor with 1 g/m^2 dust loading, and 0.034 $\mu g/m^3$ and 0.061 $\mu g/m^3$ for the carpeted floor with 10 g/m^2 dust loading, respectively. Permethrin concentrations in the settled dust samples were approximately one-fourth of that measured in the stationary and mobile samples in the carpeted floor experiments. Thus, the use of stationary samples and settled dust samples may underestimate a

toddler's personal inhalation exposure to permethrin in residential houses by approximately a factor of 2 and 4, respectively.

Particle mass concentrations measured in mobile samples were significantly higher than that measured in stationary samples. Thus, using stationary sampling would underestimate toddlers' inhalation exposure to particles and potentially the contaminants attached onto the particles. Particle size distributions in mobile and stationary samples were not statistically significantly different from each other. However, settled dust samples have a significantly higher percentage of large particles (5-10 μm) and lower percentage of small particles (1-2.5 μm). Smaller particles have a larger surface area per volume, potentially resulting in more toxic semi-volatile chemicals attached per mass. Therefore, using settled dust as an indicator of young children's exposure would underestimate their exposure to toxic chemicals.

Toddlers' inhalation exposure to permethrin in the simulated residential environment and the impact of toddlers' activities on the estimation of toddlers' inhalation exposure to permethrin were evaluated using Monte Carlo simulation and sensitivity analysis. Comparing three different modeling approaches (mobile, stationary and settled dust), toddlers' inhalation exposure to permethrin was impacted by their indoor activities. If the mobile sample best represents a toddler's exposure, using settled dust to estimate toddlers' daily inhalation intake might overestimate this value, while using stationary samples might underestimate toddlers' daily inhalation intake compared to using mobile samples.

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List of Abbreviations

$Intake_{mobile}$	Short term inhalation intake of permethrin for a toddler via mobile sampling (µg/kg/day)
$Intake_{settled\ dust}$	Short term inhalation intake of permethrin for a toddler via settled dust (µg/kg/day)
$Intake_{stationary}$	Short term inhalation intake of permethrin for a toddler via stationary sampling (µg/kg/day)
ANOVA	Analysis of Variance
BW	Toddler's body weight (kg)
C	Airborne particle mass concentration (g/m^3)
$C_{airborne}$	Dust airborne concentration ($\mu g/m^3$)
C_{floor}	Dust surface loading ($\mu g/m^2$)
C_{mobile}	Pyrethroid air concentration in mobile samples ($\mu g/m^3$)
$C_{outdoor}$	Pyrethroid air concentration outdoors ($\mu g/m^3$)
$C_{settled\ dust}$	Pyrethroid concentration in settled dust (µg/g)
$C_{stationary}$	Pyrethroid air concentration in stationary samples ($\mu g/m^3$)
$f_{on\ floor}$	Fraction of time toddler spends on the floor, bounded between 0 and 1
GC-ECD	Gas Chromatography-Electron Capture Detector
GM	Geometric Mean
H	Sampling height (m)
$InhR_{off\ floor}$	Toddler's inhalation rate for off floor activities (m^3/min)
$InhR_{on\ floor}$	Toddler's inhalation rate for on floor activities (m^3/min)

$InhR_{outdoor}$	Toddler's inhalation rate outdoors (m^3/min)
k	Correction factor for permethrin concentration in settled dust
K_{ow}	Octanol-water partition coefficient
K_{oa}	Octanol-air partition coefficient
L	Surface loading of the dust (g/m^2)
LCD	Liquid-Crystal Display
NOAEL	No Observed Adverse Effect Level
OPC	Optical Particle Counter
OPFRs	Organophosphate Flame Retardants
PAHs	Polycyclic Aromatic Hydrocarbons
PIPER	Pre-toddler Inhalable Environmental Robot
PM	Particulate Matter
PM_{10}	Particulate matter 10 micrometers or less in diameter
R	Resuspension flux (g/m^2/hr)
r	Resuspension rate (/hr)
SD	Standard deviation
SVOCs	Semi-volatile Organic Compounds
t	Time (hr)
t_{indoor}	Toddler's time spent indoors (min/day)
Tukey's HSD Test	Tukey's Honest Significant Difference Test
ε	Amount of dust that children inhale per day (μg/day)

Chapter 1

Introduction

1.1. Background

Pesticides are commonly used worldwide to control insects and pests in agricultural and residential settings. Nearly 6 billion pounds of pesticides were applied worldwide annually in both 2011 and 2012, while in the United States alone, the usage of pesticides was over 1.1 billion pounds (1, 2). Pyrethroids, a group of synthetic insecticides, are one of the most frequently used pesticides (2-5). They are widely used in agriculture and residential houses for pest control. Their usage has increased extensively in recent years. According to U.S. EPA Permethrin Facts (6), approximately 2 million pounds of permethrin are applied annually to agricultural, and residential sites and public health uses. Studies in Northern California involving 259 residential households showed that 77% of pesticides used are pyrethroids (7). Consistent with its growing application, there have been increasing concerns about adverse human health effects to pyrethroids.

Several pyrethroids are known to cause seizures and paresthesias by affecting humans' central nervous system (8-11). It is also suggested that they have a suppressive effect on the immune system and may cause lymph node and spleen damage (3, 12). Permethrin, one of the most widely used pyrethroids, is suspected of being an endocrine-disrupting chemical (13, 14) and, along with fenvalerate, has been classified as a potential carcinogen at high exposure levels (6). In addition, pyrethroids have also

been linked to respiratory diseases such as hyper-sensitization. They have strong excitatory action in the vertebrate skin and upper respiratory tract (15, 16).

Children, with developing physiological and behavioral characteristics, are often more vulnerable to pyrethroids exposure than adults. Physiologically, they are at an early-stage of physical development. Pharmacokinetics (including absorption, distribution, metabolism and elimination of a chemical) differ between children and adults. Children's developing organs are particularly susceptible to toxic insult since cell division occurs at an increased rate in children and some functional excretion systems are immature (17). In addition, compared with adults, children have much greater metabolic rates and activity levels, which may lead to greater breathing rate and consumption rate of food and water on a per-body-weight basis (18). Physically, children's behavior and the way they interact with the environment also make them more vulnerable to environmental contaminants. Their activities are closer to the ground and thus they face a more contaminated, dustier environment. In addition, for infants and toddlers, their mouthing behaviors can increase their exposure to environmental agents (19, 20).

With the increasing application of pyrethroids in residential houses and the increasing concern of pyrethroids' adverse health effects on children, several studies have been conducted to investigate the multiple exposure pathways and routes of children's exposure to pyrethroids. Studies have shown that residential pesticide use represents an important risk factor for children's exposure to pyrethroid insecticides (3, 21). Though inhalation is estimated to contribute <1% of the total pyrethroid exposure for children

(22), it could impact a child's respiratory system (23). The focus of this thesis is on children's inhalation exposure to pyrethroids in house dust. As semi-volatile compounds, with a high partition coefficient between solid phase and gas phase (log K_{oa} around 10), pyrethroids applied in the indoor environment mostly partition into house dust (24). Thus house dust is a major reservoir for pyrethroids in residences. Human activities such as walking and cleaning are the primary activities leading to particle resuspension to the indoor air (25). Children's movement indoors can result in dust resuspension leading to elevated indoor particle air concentration, thereby increasing children's potential inhalation exposure to pyrethroids from resuspended dust.

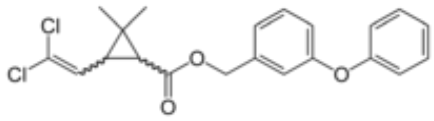
Toddlers are the age group with the greatest potential exposure to pyrethroids considering both their developmental stage and activity patterns. However, the size and weight of the sampling equipment make it very difficult to collect personal air samples from this age group. Several studies have estimated toddlers' inhalation exposures using toxicant concentrations in surface dust samples assumed to be available for resuspension (26-28), but those estimations might not adequately accounts for dust resuspension by a moving toddler. Thus, a toddler's personal inhalation exposure to pyrethroids remains unknown. Therefore, a better understanding and characterization of toddlers' inhalation exposure to pyrethroids in homes is needed to characterize their multi-route multimedia exposure to pyrethroids.

1.2. Permethrin in homes

1.2.1. Characteristics

Permethrin is one of the most commonly found and used pyrethroids in homes (29). Physical and chemical properties of permethrin are presented in Table 1.1.

Table 1.1. Physical and chemical properties of permethrin

Name	Permethrin
Chemical name	(3-Phenoxyphenyl)methyl (±)-cis,trans-3-(2,2-dichloroethenyl)-2,2-dimethylcyclopropanecarboxylate
Trade name	Elimite, Nix, Lyclear, Acticin
Chemical structure	
Molecular weight	391.3 g/mol
Chemical formula	$C_{21}H_{20}Cl_2O_3$
CAS number	52645-53-1
log K_{ow}	7.43
Vapor pressure	2.48 µPa at 20°C (cis), 1.49 µPa at 20°C (trans)

1.2.2. Pesticides application frequency/ pattern in homes

The National Home and Garden Pesticides Survey of 2,078 households found that for homes that treated the primary living area, the most common frequency of treatment was 13 to 52 applications per year (30). Bass et al. (31) conducted a pesticide use survey in 107 households and found that 32% of the pesticides were used at least once a week, while 28% of households used it once a month.

Pesticide application methods varied depending on the target pest (s), target site (s), properties of the pesticides, etc. In residential households, broadcast application, spot treatment and crack and crevice (perimeter) applications were often used. Broadcast application refers to uniformly spraying the pesticides to an entire area or field. Spot treatment refers to applying the pesticides to a small, distinct area. Crack and crevice application refers to placing small amounts of pesticide into cracks and crevices (30).

1.2.3. Indoor distribution, source and concentration of permethrin

When pesticides are sprayed indoors, there is a tendency for pesticides to get redistributed from an initial location to all indoor surfaces. Weschler et al. (32) proposed a model that included two source terms and three sink terms to determine pyrethroid levels in the indoor air. The two source terms were the escape rate of permethrin molecules from the source and the desorption rate from room surfaces. The three sink terms were the return rate of permethrin molecules from air to the source, the sorptive uptake rate onto other indoor surfaces, and the removal rate by means of ventilation. Permethrin is semi-volatile with a large partition coefficient between solid phase and gas

phase (log K_{oa} around 10.6) (EPISuite™, https://www.epa.gov/tsca-screening-tools/epi-suitetm-estimation-program-interface). Thus, permethrin would potentially stay on indoor surfaces, with house dust being a major reservoir for pyrethroids in homes (33). Permethrin is commonly detected in dust and indoor air samples in residential houses, though its concentration is highly variable (Tables 1.2 & 1.3). Floor dust is a major source of exposure for infants and toddlers, via inhalation of resuspended particles, dermal absorption and non-dietary ingestion. Toddlers are at an early stage of walking and their activities indoors can resuspend dust, resulting in inhalation exposure to the resuspended particles and to the permethrin adsorbed onto the resuspended particles.

Table 1.2. Permethrin concentrations measured in indoor dust samples (ng/g)

Compounds	N	Range (ng/g)	Mean (ng/g)	Reference
Cis-permethrin	120	16.6-79,600	2,320	Morgan et al. (34)
Trans-permethrin	118	16.5-78,800	2,340	
Cis-permethrin	N/A	26-30,600	2,740	Starr et al. (35)
Trans-permethrin	N/A	24-30,400	2,700	
Cis-permethrin	N/A	ND-1,410	N/A	Trunnelle et al. (36)
Trans-permethrin	N/A	ND-1,737	N/A	
Permethrin	35	130-13,100	N/A	Julien et al. (37)
Cis-permethrin	N/A	ND-240,000	4,340	Colt et al. (38, 39)
Trans-permethrin	N/A	ND-328,000	7,500	

Permethrin	N/A	ND-187,000	140	Becker et al. (40)
Cis-permethrin	11	ND-864	182	Hwang et al. (41)
Trans permethrin	11	ND-2,164	403	
Cis-permethrin	20	13-2,900	150 (median)	Bradman et al. (33)
Trans-permethrin	20	22-5,800	230 (median)	
Cis-permethrin	119	<300-61,900	2,680	Rudel et al. (42)
Trans-permethrin	119	<400-98,000	5,030	
Permethrin	1215	<20-267,000	3,170	Seifert et al. (43)
Cis-permethrin	181	16-168,000	N/A	Harnly et al. (44)
Trans-permethrin	177	146-265,000	N/A	

Table 1.3. Permethrin concentrations measured in indoor air (ng/m^3)

Compounds	N	Range	Mean	Reference
Cis-permethrin	125	ND-5.4	N/A	Morgan et al. (34)
Trans-permethrin	125	ND-6.8	N/A	
Permethrin	N/A	ND-3.03	N/A	Lu et al. (45)
Cis-permethrin	9	ND-92	N/A	Tulve et al. (21)
Trans-permethrin	9	ND-130	N/A	
Cis-permethrin	102	<0.4-125	N/A	Whyatt et al. (46)

Trans permethrin	102	<0.1-164	N/A	
Cis-permethrin	20	ND-8.2	N/A	Bradman et al. (33)
Trans permethrin	20	ND-11	N/A	

1.2.4. Toxicity and health effects

Common symptoms associated with pyrethroid exposures includes adverse respiratory (e.g., cough or upper respiratory irritation), neurological (e.g., headache or dizziness), gastrointestinal (e.g., nausea and vomiting), ocular (e.g., irritation) and/ or dermal (e.g., paresthesia) outcomes (47, 48). In a study of 4,974 cases of acute pyrethrin/pyrethroid-related illnesses occurring between 2000-2008, the most common symptom (48%) was adverse respiratory effects (49). Several studies of workers spraying pyrethroids on crops or in occupational settings reported cutaneous paraesthesia and respiratory sensations (50, 51). Respiratory irritation was also reported in laboratory animals acutely exposed to pyrethroids (52, 53) and for repeated 90 day pyrethroid exposures (54). Less is known about the long-term health effects of repeated exposure to low levels of pyrethroids (48). Besides the toxicity and potential health effects discussed above, permethrin is suspected of being an endocrine disrupting chemical (13, 55, 56) and has been classified as a potential carcinogen at higher exposure levels (57).

Children may be more sensitive than adults to pyrethroids. Studies have shown that the developing lungs of children are especially vulnerable to inhalation exposure of particles and contaminants attached to those particles (58, 59). Although no health reports on

children's potential pesticide exposures were located in the literature, it is reasonable to assume that children would exhibit symptoms and disease endpoints similar to those found in adults.

1.3. Dust resuspension and loadings on the floor

People spend approximately 90% of their time indoors and very young children might spend even more time indoors, nearly 99% (60). Therefore, determining their inhalation exposure to airborne particles indoors is critical to estimate risk from pesticide exposures. Dust resuspension is one of the major sources of indoor particles and is reported to increase the risk for inhalable particulate matter exposure (61). Typical indoor activities, such as cleaning and walking, can cause significant dust resuspension (62). Hu et al. (63) concluded that human activity can generate particle resuspension indoors by influencing mechanical vibration, aerodynamic and electrostatic forces. Thatcher et al. (26) found that even normal activities, such as walking and sitting, increase indoor concentrations of particles greater than 1 μm (29). Ferro et al. (64) reported that most of the resuspended particle mass from human activities indoors was larger than 5 μm and the amount of resuspension was affected by the number of persons performing the activity, the vigor of the activity, the type of activity and the type of flooring.

The extent of dust resuspension from different flooring materials can differ for similar human activities or disturbances. Thus the ratio of the resuspended dust air concentration to the dust loadings on the floor can differ across floor types. The term "dust resuspension factor" is used here to describe the ratio between airborne particle

concentration and surface particle loading (65). The equation for the dust resuspension factor is:

Equation 1.1. $RF(m^{-1}) = \frac{C_{airborne}}{C_{floor}}$

Where, $C_{airborne}$ is the dust airborne concentration ($\mu g/m^3$)

C_{floor} is the dust surface loading ($\mu g/m^2$)

In addition, the average dust loading also varies across different flooring materials present in residences, with carpeted floor usually having the highest dust loading (26). Table 1.4 presents dust loading levels reported in the literature.

Table 1.4. Dust loadings in residential houses published in previous studies

Flooring material	N	Range (g/m^2)	Reference
Smooth floor	488	0-13.86 (Mean: 0.311)	Johnson et al. (55)
Smooth floor	444	0.05-7 (GM: 0.42)	Adgate et al. (66)
Carpeted floor	376	0.3-99 (GM: 7.8)	Adgate et al. (66)
Carpeted floor	11	0.32-14.4 (Median: 1.3)	Roberts et al. (67)
Carpeted floor	73	1-136 (Median:16)	Wang et al. (68)

Thus a toddler's inhalation exposure to pyrethroids in residential houses will vary with different flooring materials. Two flooring materials were tested in our study: vinyl

flooring and carpeting. A dust loading of 1 g/m^2 and 10 g/m^2 were selected for vinyl floor and carpeted floor, respectively, to be representative of real world conditions.

1.4. Toddlers' inhalation exposure estimation

1.4.1. Toddlers' characteristics

Toddlers, one to three years old, are at an early stage of physiological and behavioral development. They breathe at a higher frequency than adults and have a greater dose of contaminants per surface area due to smaller lung sizes (69). A study by Bennett et al. (70) comparing the nasal deposition efficiency of fine particles in children (6-10 yr) versus young adults suggested that children have less efficient nasal filtering for larger particles and that children's lungs may potentially be exposed to higher concentrations of inhaled particles. Tracheobronchial and pulmonary deposition fractions of particles per unit volume of air or per unit area of the lung surface were reported to be greatest for infants (71). Xu et al. (72) conducted theoretical calculations for the deposition of inhaled aerosol particles in the respiratory tract of humans from birth to adulthood and found that children have higher total deposition in the respiratory tract and deposition in the head region for all particle sizes. Similar conclusions can be found in another study by Musante et al.(73). Therefore, when young children are exposed to contaminants or toxic laden particles, they have increased exposure and risks compared to adults.

Young children's behaviors and the ways they interact with their environment affect the magnitude of their exposure to contaminants (74, 75). Toddlers are at an early stage of mastering walking skills and are beginning to become involved in more vigorous

activities. They move in a manner different than adults and have a breathing zone height closer to the floor (around 80 cm) (76), therefore they encounter a dustier environment. In addition, their hand to mouth behaviors increase their contact with contaminants, resulting in an elevated exposure (77, 78).

1.4.2. Young children' exposure to pyrethroids at homes

Young children can be exposed to pyrethroids via multiply routes and exposure pathways, e.g. dietary and non-dietary ingestion, dermal contact. Several studies have been conducted to examine children's pyrethroid exposure at home via multiple exposure routes (22, 33, 34, 79). Morgan et al. (34) estimated the permethrin exposure of 57 children aged 2-5 years and reported that the primary exposure route was dietary ingestion of solid foods, followed by non-dietary ingestion of dust. Tulve et al. (22) estimated that dermal contact was the primary route of exposure, followed by dietary ingestion in a study evaluating cumulative pyrethroids exposure of children aged 4-6 years in homes with frequent pesticides use. Zartarian et al. (79) applied the SHEDS-Multimedia Model to estimate U.S. population permethrin exposures for 3-5 year old children and found that for children in households where residential applications of permethrin occurred, the non-dietary exposure route was most important while dietary exposure dominated when all households were included. Though inhalation exposure was evaluated in some of the studies mentioned above (22, 33), the measured pyrethroids air concentrations were negligible.

However, even low inhalation exposures may be important since animal studies indicated potential hazard effects via inhalation of pyrethroids (80-84).Toxicity and occupational studies (11, 84-86) also reported potential respiratory irritation and other symptoms following inhalation exposures to permethrin. Thus, it is critical to have an accurate evaluation of the inhalation route of pyrethroid exposure to estimate the associated risk of respiratory diseases.

1.4.3. Estimation of toddlers' inhalation exposure to pyrethroids

Several observational exposure measurement studies have assessed young children's inhalation exposure to pyrethroids in homes based on two surrogates for exposure concentration, stationary indoor air sampling and settled dust sampling.

Bradman et al. (33) collected 24-hr integrated stationary indoor air samples to evaluate 5-27 months old children's inhalation exposure to pyrethroids. Tulve et al. (22) collected 24-hr stationary integrated indoor air sample to evaluate 4-6 year old children's inhalation exposure to pyrethroids. Kawahara et al. (87) measured 24-hr stationary airborne organophosphorus pesticide concentrations in an agricultural community in the suburbs of Tokyo, Japan to assess the inhalation exposure of children 1-6 years old. Daily inhalation exposure estimated using the stationary monitoring data and time-activity questionnaire ranged from 0 to 35 ng/kg/day for trichlorfon, from 0 to 26 ng/kg/day for dichlorvos, and from 0 to 44 ng/kg/day for fenitrothion. For the above discussion, stationary sampling data were used to estimate young children's inhalation exposures.

They did not fully consider resuspended dust and young children's indoor activities, therefore, these studies could be underestimating the children's inhalation exposures.

A limited number of publications have reported the amount of household dust a child would potentially inhale per day. Hawley et al. (88) reported an indoor dust inhalation exposure of 0.15 mg/day for warm weather and 0.34 mg/day the rest of the year for 2-3 year old children. Oomen et al. (89) estimated children's daily inhaled dust to be 0.8 mg/day by assuming a constant suspended particle air concentration of 100 $\mu g/m^3$, and the volume of inhaled air being 7.6 m^3 for a child. Since directly measuring young children's personal exposures is not feasible, some researchers have used settled dust samples as a surrogate to estimate a child's inhalation exposure. However, particle size distributions in resuspended dust are skewed to a smaller particle size distribution than settled dust and the concentrations of toxic chemicals typically increases with decreasing particle sizes (90, 91). As a result, using settled dust samples will likely underestimate young children's inhalation exposures.

1.4.4. Robotic simulation of toddlers' movement

Toddlers' personal exposure is usually estimated by stationary sampling due to ethical concerns and the difficulty of attaching samplers directly to a toddler. Several studies by Shalat et al. (92-98) applied a robotic surrogate, the Pre-toddler Inhalable Environmental Robot (PIPER), to simulate toddlers' movement indoors while collecting air samples. PIPER is an autonomous robot used to improve estimation of a toddler's personal exposure. In our study, we utilized a commercially available robot, the ReCon 6.0

Programmable Rover from SmartLab Toys (https://smartlabtoys.com/products/recon-6-0-programmable-rover, Bellevue, WA, USA), as a surrogate to simulate toddlers' indoor floor activities. ReCon Rover is capable of mechanically resuspending dust to simulate a young child's generation of airborne house dust s during walking or running. It also provides a platform to collect air samples at a toddler's breathing zone height.

No side by side sampling was performed directly between a toddler and our robot due to ethical concerns and the difficulty of placing samplers on a toddler. Currently we could not locate any study in the literature reporting particle concentrations resulting from dust resuspension by toddlers to evaluate young children's exposure to particles. However, Rosati et al. (99) reported PM_{10} concentrations in a range of 9-518 $\mu g/m^3$ for walking-induced dust resuspension in carpeted floor experiments. The particle concentrations measured in our study were within that range.

We did comparison tests between the ReCon Rover and PIPER. The details of this comparison can be found in Appendix I. The particle concentrations measured by the ReCon Rover were approximately three times higher than that measured by PIPER, which indicated that the ReCon Rover was capable of simulating a dust cloud effect and generating dust resuspension similar to a toddler's movement.

1.5. Overview of study

The overall objective of this study was to characterize a toddler's short term inhalation exposure to permethrin in house dust. To achieve this goal, three sub-studies were conducted: 1) permethrin concentrations were measured via three different sampling

approaches: mobile, stationary and settled dust on vinyl and carpeted floors in an indoor office setting before and after permethrin application; 2) particle size distributions of resuspended dust and settled dust were measured and compared; 3) Monte-Carlo simulations were performed to probabilistically estimate toddlers' inhalation exposures to permethrin via the three sampling approaches mentioned above.

The hypotheses of this study were:

Hypothesis 1: Stationary and settled dust samples underestimate toddlers' personal inhalation exposure to permethrin in house dust compared to mobile samples.

Hypothesis 2: Estimation of toddlers' inhalation exposure to permethrin will be underestimated when toddlers' indoor activities are not included in exposure models.

Hypothesis 1 is evaluated in Chapters 2 and 3 using the follow specific aims. Specific aim 1 was to simulate a residential room by installing vinyl or carpeted flooring in an empty office. A typical household dust loading was added and perimeter spray of permethrin was conducted to mimic a residential application scenario. Specific aim 2 was to measure permethrin concentrations at a toddler's breathing zone height using mobile sampling and stationary sampling as well as in settled dust samples. Paired t-tests with an accepted statistical significance of $p<0.05$ were used to determine if statistically different permethrin air concentrations existed between mobile and stationary samples. One way ANOVA tests and post hoc Tukey's HSD tests were used to evaluate differences in permethrin dust concentrations among mobile, stationary and settled dust samples. Specific aim 3 was to measure particle size distributions for the mobile and stationary

samples using a handheld optical particle counter. Microscopy analysis along with ImageJ processing was used to determine the particle size distribution in settled dust. ANOVA tests were performed to identify significant differences in particle size distributions among the different sampling approaches (mobile, stationary and settled dust sampling).

Hypothesis 2 is evaluated in Chapter 4 using specific aim 4. Specific aim 4 was to perform a probabilistic exposure assessment using a Monte Carlo approach to estimate toddlers' inhalation exposure to permethrin in house dust based on measured environmental data and empirical distributions of toddlers' physical characteristics from the literature. Toddlers' indoor activity patterns were taken into account when generating parameter distributions and calculating exposure intakes.

Chapter 2

Pyrethroid Levels in Toddlers' Breathing Zones Following an Indoor Pesticide Spray

2.1. Introduction

The widespread use of pyrethroids in residential houses (49, 100) and the increasing concern about pyrethroids' adverse health effects on children (29, 101) have led to several studies investigating multiple exposure pathways and routes of children's exposure to pyrethroids (3, 21, 33, 34, 37, 102-104). Residential pesticide use represents an important source of children's exposure to pyrethroid insecticides (3, 21). Pyrethroids applied in indoor environments redistribute from their initial locations to all indoor surfaces (105), especially to house dust because they are semi-volatile organic compounds with high partition coefficients between the solid and gas phases (log K_{oa} around 10) (EPISuite^{TM}, https://www.epa.gov/tsca-screening-tools/epi-suitetm-estimation-program-interface). Thus, house dust is a major reservoir for pyrethroids in homes (33).

Children are more vulnerable to pyrethroid exposure than adults due to their developing physiology and their vigorous indoor activities. Physiologically, children are at an early stage of physical development, and pharmacokinetics (including absorption, distribution, metabolism, and elimination of a chemical) differ between children and adults. Children's developing organs are particularly susceptible to toxic insult because cell

division occurs at a higher rate compared to adults, and some functional excretion systems are still immature (106). Also compared with adults, children have greater metabolic rates and activity levels, which leads to a greater breathing rate on a per-body-weight basis (18). Physically, children's behaviors and their interactions with their environment can increase their exposure to environmental contaminants, particularly indoors where they spend approximately 90% of their time (60). Their vigorous activities such as playing on the floor can resuspend dust, leading to an increased indoor particle concentration in their breathing zone. Children's higher breathing rates per body weight then lead to potentially increased inhalation exposures, especially since their breathing zone is closer to the floor, where resuspended particle concentrations are higher when compared to the breathing zone height of adults (107). In addition to pyrethroid exposure due to inhalation of resuspended dust, infants' and toddlers' mouthing behaviors also result in ingestion exposure to dust contaminated with pyrethroids (74, 108). Though inhalation is estimated to contribute <1% of the total pyrethroid exposure to children (22), evidence in the literature that pyrethroid inhalation exposure could adversely impact a child's respiratory system (23).

Toddlers, who are one to three years old and at the early stages of walking, potentially are in the most vulnerable children's age group when it comes to pyrethroid exposure. However, collecting personal air samples for this age group is difficult due to the size and weight of the sampling equipment and toddlers' propensity for putting objects in their mouths. Characterization of children's inhalation exposure typically relies on stationary air sampling or settled dust measurements (26-28, 109). Stationary air sampling is likely

to underestimate children's inhalation exposure because it underestimates concentrations of airborne dust caused by children's indoor activities. The underestimation of particulate matter exposure is likely greater for young children because the commonly used height for stationary samplers of 110 cm (110) is higher than a toddler's average breathing zone height (94). Previous studies have used the concentration of semi-volatile organic compounds in the settled dust to estimate concentrations of these contaminants in the air (111-113). However, the particle size distribution in the resuspended dust and settled dust might be different which potentially results in different contaminant concentrations. This difference is especially important because the smaller size fraction of resuspended dust is expected to be enriched in semi-volatile organic compounds compared to settled dust due to a greater surface area to volume ratio in smaller particles (90, 91).

Given the uncertainties in estimating children's exposures to dust in general and to pyrethroids in particular, the overall objectives of this study are to compare different approaches for collecting airborne and surface-borne dust when estimating pyrethroid concentrations in toddlers' breathing zones and to provide a better understanding and characterization of toddlers' inhalation exposures to pyrethroids in homes. We used a robotic surrogate to simulate toddlers' indoor activities and mounted a personal sampler on the robotic surrogate to collect mobile air samples that represented children's exposures. Stationary air samples and settled dust samples were collected concurrently. Pyrethroid concentrations as well as particle mass concentrations were analyzed and compared among mobile air, stationary air and settled dust samples.

2.2. Methods

2.2.1. Description of experimental room

The study was conducted in an empty office in the Robert Wood Johnson Medical School in Piscataway, NJ. The room dimensions were 3.6 m (length) x 3.3 m (width) x 3.5 m (height), yielding a floor area of 11.9 m^2 and a volume of 41.6 m^3.

2.2.2. Surrogate toddler-robotic ReCon Rover

A commercially available robot, ReCon 6.0 Programmable Rover from SmartLab Toys (https://smartlabtoys.com/products/recon-6-0-programmable-rover, Bellevue, WA, USA) (Figure 2.1), was used to simulate a toddler's indoor movements and carry sampling equipment. It is powered by three C-type batteries and can be programmed through an LCD screen and 10-button keypad to control its movements in a sequence of directions: forward, backward, 45 and 90 degree turns. The rover weighs 1.5 kg and has dimensions of 23 × 20 × 28 cm. When equipped with sampling devices (a PM_{10} sampler, a sampling pump, and an optical particle counter), it weighs around 3 kg. We programmed the ReCon Rover to go in a spiral pattern in the room to cover the entire room area at its own fixed speed of 8 cm/s. Since the robot was not able to avoid obstacles, the researcher stayed in the experimental room to restart and redirect the robot when it encountered an obstacle.

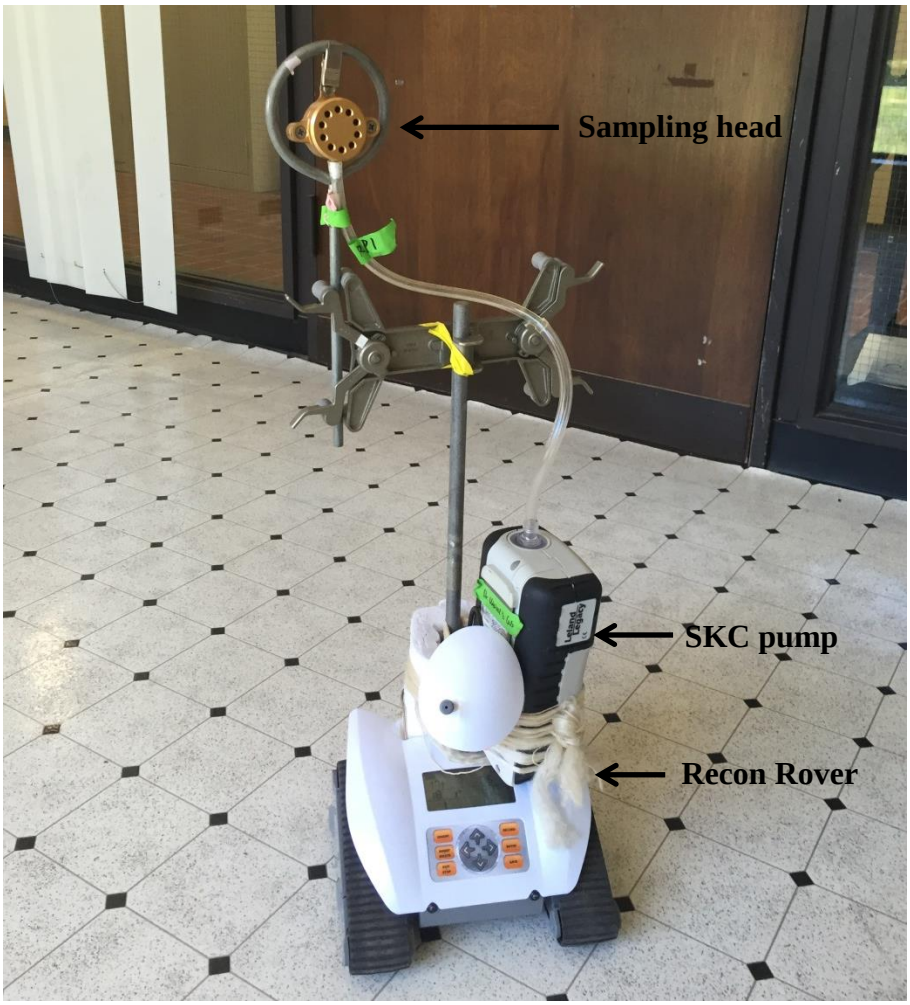


Figure 2.1. ReCon Rover and samplers.

2.2.3. Flooring types

Two flooring types, a vinyl floor (Traffic MASTER black and white decorative paver vinyl sheet, Model #U9430.271K509G144) and a medium pile carpet (Hot Shot II - Color Tuscan Texture carpet, Model # H2004-402-1200-AB) were used in this study. After installation, the carpet was vacuumed using a Scorpion Quick Flip handheld vacuum cleaner (Model 08220, Dirt Devil, Glenwillow, OH, USA) until no large fibers

were visible in a dust collector after vacuuming an area of 60 cm × 60 cm (approximately 5 min collection time).

Between tests on the vinyl floor, a broom was used to remove the bulk of the floor dust, and a mop wetted with an ethanol and water mixture (volume ratio of 1:2) was used to mop the floor 3 times. A towel wetted with an ethanol and water mixture was used to clean the walls and other interior surfaces. After cleaning, the room was left for 1-2 days to equilibrate. For each carpet test, the old carpet was removed from the room, and a new piece of carpet was used.

2.2.4. Simulated dust loading

House dust was obtained from vacuum cleaners from 16 residential houses in New Jersey, USA. The dust was initially sieved through a 125 μm mesh to remove very large particles and fibers as they would not contribute to the resuspended dust particles, which are primarily <10 μm (61). A 200 mg portion of dust from each house was analyzed for pyrethroid levels as described later in the sample analysis section. Dust samples with pyrethroid levels below the detection limit of 10 ng/g were selected for background dust: these samples were homogenized and stored at -4 °C in a freezer for subsequent use. The average dust loading and the extent of dust resuspension varies with flooring types, with carpets reported to have higher dust loadings than hard floor surfaces (26). A vinyl floor dust loading of 1 g/m^2 and a carpet dust loading of 10 g/m^2 were selected to be within ranges reported in U.S. homes (27, 28) and to provide sufficient resuspended dust to be measured. The total amount of applied dust (g) was calculated as the floor area (m^2)

multiplied by the target dust loading (g/m^2), and was 12 g for the vinyl floor and 120 g for the carpeted floor.

The dust was loaded onto the floor using a dust dispenser: a 25 mL Pyrex midget impinger (Corning Inc., Corning, NY, USA) with an inlet connected to an air pump (positive pressure) providing an air flow of 25 L/min through the impinger. The dust was put at the bottom of the impinger, and the air was forced through the inlet nozzle, which resulted in the dust being suspended within the impinger and released through the outlet nozzle.

The room was divided into 4 sections for the vinyl floor and 8 sections for the carpeted floor. The dust was sprayed using the impinger while slowly walking across each section to deposit the dust evenly across the entire room. The impinger was periodically tapped with a stainless steel laboratory spatula to prevent the dust from clogging the nozzle. After the dust was sprayed, the room was left undisturbed overnight to allow the dust to settle.

After spraying the carpet, the settled dust was embedded into the carpet to be representative of in-use carpets (114). Briefly, a carpet roller (Model 10-935, Roberts Consolidated Industries, Inc. Boca Raton, FL, USA) was used as a dust embedment tool. The roller was pulled in both directions with the handle held at an angle of 30-45 degrees to the floor. The dust embedment tool was rolled over the entire carpeted area for exactly 30 strokes (a movement in one direction covering the length of the flooring is one "stroke"). The applicator walked at a uniform speed of approximately 0.5 m/s. After the

dust embedment had been completed, the room was left for at least one day to allow any resuspended dust to settle and to allow the temperature and relative humidity in the room to equilibrate.

In order to evaluate the floor dust loading distribution, sixteen pieces of weighing paper were placed on the vinyl floor prior to spraying the dust and left overnight while the dust settled. The weighing papers were collected, and the weight of the dust was measured using a Mettler Toledo AT261 balance (Mettler-Toledo GmbH, Laboratory & Weighing Technologies, Greifensee, Switzerland). The dust loading on each section was calculated and compared with the expected value (data presented in the supplemental material).

2.2.5. Indoor pyrethroid application

A 0.5% emulsion permethrin spray solution was prepared by diluting a permethrin concentrate purchased from Control Solutions, Inc. (Lot#: 20184, Pasadena, TX, USA). A commercially available polyethylene sprayer (Model 2121 from Chapin International, Inc., Batavia, NY, USA) was used to spray pyrethroids in the room. Permethrin was sprayed along the baseboards of the room after the background PM_{10} and settled dust samples were collected. The sprayer nozzle was held approximately 30 cm from the floor, and the applicator moved at a walking speed of 0.5 m/s. The spray resulted in a slightly visible film on the floor. The permethrin spray protocol was intended to simulate residential baseboard spraying practices for pest control.

2.2.6. Sample collection

Airborne samples (mobile samples and stationary samples, Figure 2.2) and settled dust samples were collected prior to the permethrin application (background samples), then at one day and three days after the permethrin application. Airborne samples were collected on a 37 mm Teflon filter with 2.0 μm pore size from Pall Life Sciences (Port Washington, NY, USA) installed in a Personal Environmental Monitor (PEM) (10-μm cut-point at 10 L/min, SKC Inc., Eighty Four, PA, USA). The PEM samplers were connected to pumps with Tygon tubing and placed at a height of 80 cm. This height represents a toddler's breathing zone following WHO Child Growth Standards, which reports average heights for toddlers of 1 to 3 years of age in a range of 74 cm to 95 cm (76). We used a Model 400 Micro-Environmental Monitor (MEM™) (MSP Corporation, Shoreview, MN, USA) and an SKC Leland Legacy pump to provide 10 L/min flow rate for the stationary and mobile samplers, respectively. The pump flow rates were checked before and after sampling using a DryCal® DC-Lite primary flow meter (MesaLabs, Butler, NJ, USA). One stationary sample and one mobile sample were collected on each sampling day. Sampling duration was 4 hours for the vinyl floor experiments and 8 hours for the carpeted floor experiments. The sampling time was extended for the carpeted floor to improve the detection limit of permethrin.

In addition to airborne samples, settled dust samples were collected. For the vinyl floor experiments, settled dust samples were collected using pre-weighed PreSep, Drain Disc. 50 mm × 55 mm wipe filters (GE Water & Technologies, Feasterville-Trevose, PA, USA). The surface was wiped with 3 successive filters for each sample using water as a

filter wetting solvent. Each day, one 23 cm × 23 cm center sample and one 23 × 23 cm corner sample from one side of the room were collected. For the carpeted floor experiments, settled dust samples were collected by mounting a DustChek™ (EM lab P&K, Marlton, NJ, USA) to the hose of a Scorpion Quick Flip Handheld Vacuum (Model 8220, Dirt Devil, Glenwillow, OH, USA). DustChek™ is a plastic dust collector that is attached to the vacuum cleaner hose to collect dust samples. Each day, one 5-minute center sample was collected from a 60 cm × 60 cm area, and one 6-minute composite corner sample was collected from three sides of the room, each with an area of 45 cm × 45 cm.

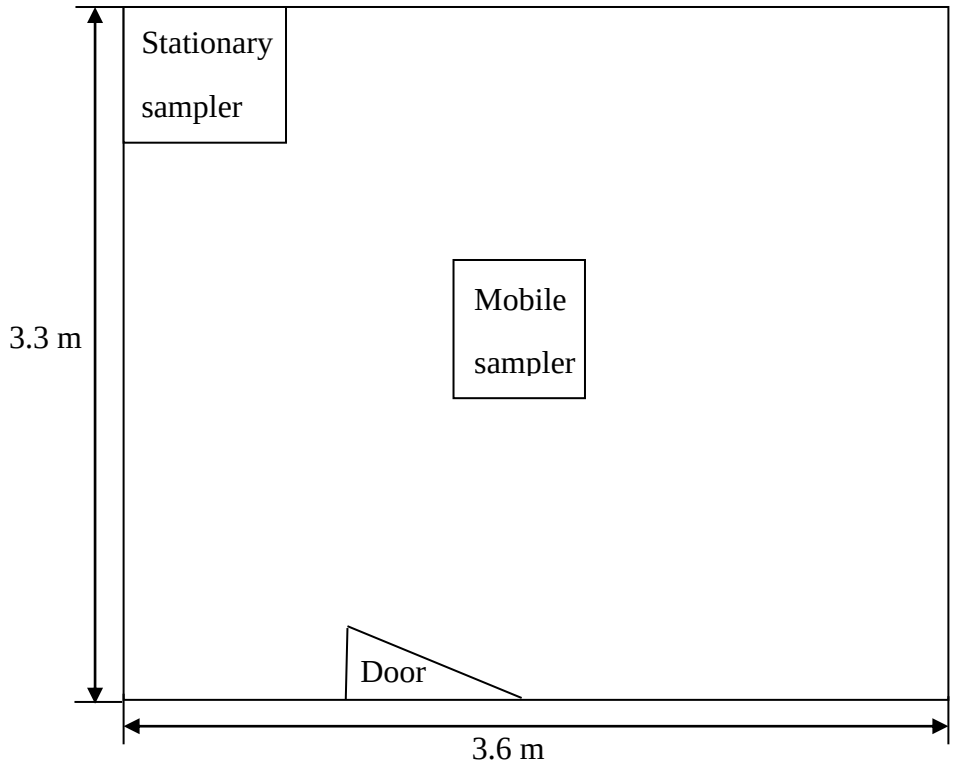


Figure 2.2. Sampling location

2.2.7. Sample preparation and analysis

All 37mm Teflon filters were equilibrated for a minimum of 72 hours prior to weighing in a weighing room with a narrow range of temperature (22-24°C) and relative humidity (30-40%). Filters were weighed using a Mettler Toledo MT5 Microbalance (Columbus, OH, USA) and placed in individual containers until they were loaded into the samplers. After sample collection, the filters were returned to the same weighing room to equilibrate at the same temperature and humidity for a minimum of 72 hours prior to reweighing with the same microbalance (97). PM_{10} concentrations were calculated as the amount of collected particles divided by the volume of sampled air.

After the filters were weighed, pyrethroids from filter samples were extracted using 6 mL of a 1:1 hexane: acetone solution while being sonicated for 10 min. The extract was then centrifuged at 4,000 rpm for 5 minutes and the supernatant transferred to a clean test tube. The volume was reduced using a Meyer N-Evap analytical nitrogen evaporator (Organomation, Berlin, MA, USA) to approximately 0.3-0.5 mL. The extract was then transferred to glass inserts that fit into GC vials and evaporated to dryness. Lastly, the extract was reconstituted in 100 μL of hexane.

A 1 μL aliquot of the sample extract was analyzed by Gas Chromatography-Electron Capture Detector (HP 5890 Series II equipped with an HP 7673 auto-sampler and a DB1701 column (30 m × 0.25 mm × 0.25 μm)). The injector was operated at a temperature of 250 °C, in the splitless injection mode. Helium was used as the carrier gas at a constant flow of 1.4 mL/min. The initial column temperature of 140 °C was held for 2 minutes, then raised to 230°C at a rate of 10 °C per minute, held for 2 minutes, and

finally raised to 250 °C at a rate of 8 °C per minute, where it was held for 10 minutes. The Electron Capture Detector was maintained at 300 °C.

2.2.8. Statistical analysis

In total, three sets of vinyl floor experiments and five sets of carpeted floor experiments were conducted. Comparison of permethrin airborne PM_{10} concentrations ($\mu g/m^3$) measured in stationary to the mobile samples was performed using a two-tailed paired t-test with statistical difference of $p<0.05$. Comparison of permethrin dust concentrations ($\mu g/g$) in mobile, stationary and settled dust samples was performed using a one-way ANOVA test, and a post hoc Tukey's HSD test applied to identify which pair (s) of samples had statistical differences of $p<0.05$.

2.3. Results

2.3.1. PM_{10} concentrations in air samples

Nine pairs of mobile and stationary airborne samples were collected in the vinyl floor experiments, while fifteen pairs of mobile and stationary airborne samples were collected in the carpeted floor experiments. The mean and standard deviation of the PM_{10} concentrations in the stationary and mobile samples measured for the vinyl floor and carpeted floor experiments are presented in Table 2.1. The overall observed means of the PM_{10} concentrations collected from stationary samples were 17.1 $\mu g/m^3$ and 27.0 $\mu g/m^3$ for the vinyl floor and carpeted floor experiments, respectively. The means of the PM_{10} concentrations measured in mobile samples were 38.4 $\mu g/m^3$ and 40.4 $\mu g/m^3$ for the vinyl floor and carpeted floor experiments, respectively.

Table 2.1. PM_{10} concentration in stationary and mobile airborne samples ($\mu g/m^3$)

		Mean ± standard deviation	Paired t-test
Vinyl floor	Stationary	17.1±7.1	N=9, p-value=0.001
	Mobile	38.4±10.4	
Carpeted floor	Stationary	27.0±10.4	N=15, p-value=0.001
	Mobile	40.4±19.7	

Paired t-test results between PM_{10} concentrations in mobile and stationary samples showed that mobile PM_{10} concentrations were statistically significantly higher than stationary PM_{10} concentrations for both vinyl floor and carpeted floor experiments with $p<0.001$. This finding supports our hypothesis that stationary sampling underestimates toddlers' inhalation exposure to resuspended particles.

2.3.2. Permethrin dust concentrations in airborne and settled dust samples (μg/g)

Permethrin dust concentrations in mobile, stationary and settled dust samples measured in the vinyl floor and carpeted floor experiments are presented in Figure 2.3. For the carpeted floor, permethrin dust concentrations (mean ± standard deviation) were 2,080±1,030 μg/g and 1,550±700 μg/g for the mobile and stationary samples, respectively. The permethrin dust concentration in the settled dust was 400±240 μg/g, which was approximately 20% and 25% of that measured in mobile and stationary samples, respectively. The permethrin concentration in the settled dust was statistically different from the stationary and mobile samples based on a post-hoc Tukey HSD test

with $p<0.05$. For the vinyl floor, the highest permethrin dust concentrations were found in settled dust samples with a mean ± standard deviation of 6,430±5,630 μg/g, while permethrin dust concentrations in mobile and stationary samples were 4,210±1,630 μg/g and 4,750±2,930 μg/g, respectively. None of the permethrin concentrations were statistically different based on the ANOVA test.

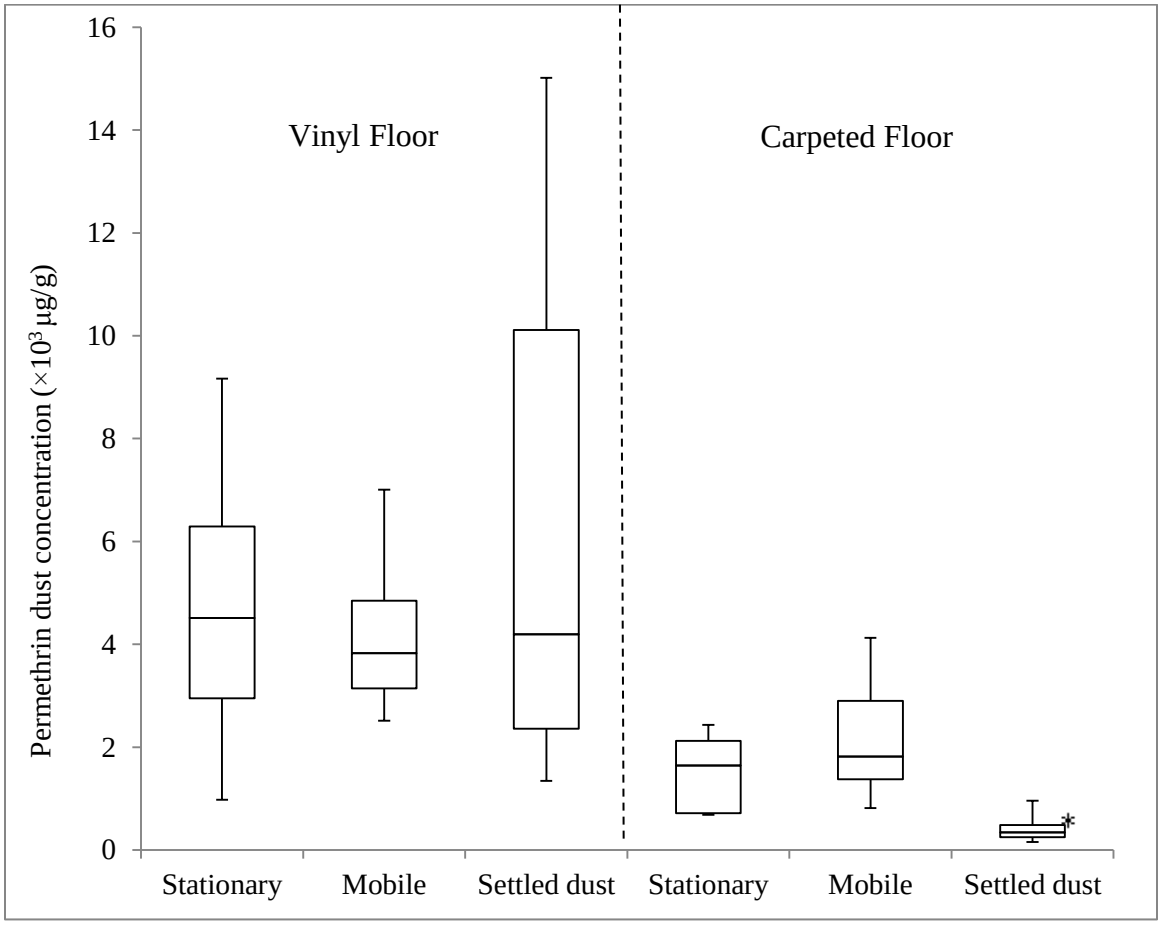


Figure 2.3. Box plot of permethrin dust concentrations (μg/g) in stationary, mobile and settled dust samples for the vinyl and carpeted floor experiments

*: Statistically different at $p<0.05$.

2.3.4. Airborne permethrin concentrations (μg/m^3)

Airborne permethrin concentrations in stationary samples and mobile samples measured on the vinyl floor and carpeted floor are presented in Table 2.2. The mean permethrin airborne concentrations measured in the vinyl floor experiments were 65±26 ng/m^3 and 143±51 ng/m^3 for stationary and mobile samples, respectively. The mean permethrin airborne concentrations in the carpeted floor experiments were lower than that for the vinyl floor measurements, being 34±20 ng/m^3 and 61±30 ng/m^3 for stationary and mobile samples, respectively. Paired t-tests between permethrin airborne concentrations in mobile and stationary samples showed that mobile permethrin airborne concentrations were significantly higher than stationary permethrin concentrations in both vinyl floor and carpeted floor experiments at a significance level of 0.05, suggesting that stationary measurements of airborne pyrethroid concentrations potentially underestimate toddlers' inhalation exposures to pyrethroids by approximately a factor of two.

Table 2.2. Airborne permethrin concentration (ng/m^3)

		Mean ± Standard deviation	Paired t-test
Vinyl floor	Stationary	65±26	N=6, p-value=0.029
	Mobile	143±51	
Carpeted floor	Stationary	34±20	N=10, p-value=0.002
	Mobile	61±30	

2.4. Discussion

2.4.1. Comparison of particle mass concentrations in airborne samples

There is limited PM exposure data for children in the age group of 1 to 3 years. Shalat et al. (97) applied a robotic personal sampling platform (PIPER: Pre-Toddler Inhalable Particulate Environmental Robot) to measure inhalable PM concentrations in a toddler's breathing zone in residential homes with carpeted and bare floors. PIPER was programmed to change both the speed of movement and the vertical sampling height to simulate a toddler's movement according to his/her age and gender. Based on measurements in 55 homes, the authors reported arithmetic means of 30.7 $\mu g/m^3$ and 34.6 $\mu g/m^3$ for stationary and mobile inhalable particle (<100 μm) concentrations measured in homes with bare floors, while arithmetic means of 41.5 $\mu g/m^3$, and 95.6 $\mu g/m^3$ were reported for stationary and mobile inhalable PM concentrations in homes with carpeted floors, respectively. Sagona et al. (95) measured inhalable PM concentrations in 2-year-old children's breathing zones via a lightweight personal sampler in residential homes to evaluate personal PM exposures. They reported an average inhalable PM concentration of 331 $\mu g/m^3$. The PM concentrations measured in our study were lower than those presented in the Shalat and Sagona studies. The major reason for our lower observed PM values is we measured PM_{10} ($d < 10$ μm) and not inhalable particles ($d<100$ μm). An additional contributor could be the differences in the dust loadings of the floors (115).

The particle mass concentrations measured in mobile samples were similar for both vinyl and carpeted flooring experiments, with mean concentrations of 38.4 $\mu g/m^3$ and 40.4

μg/m^3, respectively. However, the particle mass concentrations measured in stationary samples in the carpeted flooring experiments (27.0 μg/m^3) were higher than those measured in the vinyl flooring experiments (17.1 μg/m^3). Differences in air concentrations might have resulted from differences in dust loadings in two flooring experiments. However, we do not know why only the stationary but not the mobile samples had different air concentrations for PM_{10}.

2.4.2. Comparison of permethrin dust concentrations

A significant difference was found between the airborne and settled dust permethrin concentrations for the carpeted floor experiments, but not for the vinyl floor experiments, which indicates there might be two different release scenarios occurring depending on flooring type. For the carpeted floor experiments, the difference in particle size distributions between settled dust and resuspended dust might cause the measured differences in permethrin dust concentrations on the settled dust and resuspended dust. Lewis et al. (90) measured pesticide concentrations in different size fractions of residential house dust and reported that the pesticide dust concentrations increased with decreasing particle sizes. Cao et al. (91) reviewed several studies on the distribution of toxic chemicals according to particle size in settled dust and concluded that concentrations of toxic chemicals increased with decreasing particle sizes. In our study, the greater surface area to mass ratio of PM_{10} fraction of the resuspended dust compared to the sieved settled dust particles with a diameter <125 μm could be expected to result in a greater permethrin concentration in the resuspended dust since it is comprised of smaller particles. While the permethrin concentrations in stationary and mobile samples

were statistically higher than the permethrin concentrations in the settled dust for carpet experiments, no difference was found for the vinyl floor experiments. One possible explanation for the permethrin levels being the same in the settled dust, stationary and mobile samples for the vinyl floor experiments is that the wipe samples used to collect the settled dust also collect permethrin adhering to the flooring material (permethrin that would not be resuspended) and not just dust particles that would be resuspended. To evaluate this possibility, one set of experiments was performed on the vinyl floor with one wipe sample and one vacuum sample (no beater bar used) collected to evaluate the difference in permethrin dust concentrations between the wipe sample and the vacuum sample. Comparable permethrin concentrations were found in the wipe sample and the vacuum sample, which indicated that the wipe sample was collecting predominately dust and not pesticide residue from the vinyl sheet (no statistical test was used). Future investigations are needed to evaluate why the expected differences were not observed among the stationary, mobile and settled dust samples for the vinyl floor experiments.

Average permethrin dust concentrations (µg/g) were compared using unpaired t-tests between the vinyl floor and carpeted floor experiments. Significant differences were found between permethrin concentrations measured on vinyl and carpeted floors across the three different sampling methods, with concentrations from vinyl floor experiments being higher. T-test p-values for mobile, stationary and settled dust comparisons on vinyl and carpeted floors were 0.0061, 0.0046 and 0.0038, respectively. There are several possible explanations for the differences. In our experiments, the carpet had a ten-fold higher dust loading (10 g/m^2) compared to the vinyl floor (1 g/m^2). Since the same

amount of pesticide was applied to both floor types, the permethrin dust concentration is expected to be higher for the vinyl floor experiments. On average, a 15-fold higher permethrin concentration (μg/g) was found in vinyl floor dust samples compared to the carpeted floor samples. A second difference is that the carpet has a larger surface area to adsorb permethrin than the vinyl floor, which may have resulted in a portion of the permethrin being absorbed by carpet fiber rather than the dust. This would further reduce the permethrin concentrations in the resuspended dust and settled dust from the carpet. The vinyl floor has a flat surface and therefore lower adsorption capacity, which may have resulted in higher permethrin concentrations in the dust.

2.4.3. Comparison of permethrin airborne concentrations

Airborne pyrethroid concentrations measured in our study were consistent with airborne concentrations measured in homes following a pesticide application. Berger-Preiß et al. (116) applied three different formulations of pyrethroids including permethrin, in a model house with carpeted floor to simulate an indoor pest control. They found that the permethrin concentration in resuspended particles was approximately 40 $\mu g/m^3$ immediately after application. The measured stationary permethrin concentrations are comparable to those in our study, which were 65 ± 26 $\mu g/m^3$ and 37 ±19 $\mu g/m^3$ (mean ± standard deviation) for the vinyl and carpeted floor stationary samples, respectively. Further, in our study, paired t-tests showed that there were statistically significant differences between stationary and mobile permethrin airborne concentrations for both vinyl and carpeted floors, with stationary permethrin airborne concentrations being lower.

The results indicated that stationary samples underestimated airborne permethrin concentrations compared to mobile samples.

Some limitations of our study are worth noting. The robot used in our study was not programmed to closely match toddlers' indoor activities as it moved at a constant speed in a pre-set pattern rather than using different activity profiles that a toddler may engage in. This could have affected the amount of dust resuspended and the simulated breathing zone height. In addition, our study was performed in a simulated home environment instead of an actual residential house.

Our study results indicate that stationary samples and settled dust samples likely underestimate young children's exposures to SVOCs associated with resuspended dust.

2.5. Conclusions

The airborne permethrin concentrations collected in the mobile samples were twice as high as those measured in the stationary samples for both vinyl and carpeted floor experiments, and permethrin dust concentrations in the resuspended dust collected by the mobile robot samples were four-fold higher than those in the settled dust samples for the carpeted floor experiments. The results indicate that using stationary and settled dust samples may underestimate toddlers' inhalation exposures to pyrethroids.

Chapter 3

Characterization of Particles in Settled Dust and Resuspended Dust in Toddlers' Breathing Zones

3.1. Introduction

Toddlers are at an early stage of walking. Their indoor activities potentially resuspend floor dust increasing the particulate matter concentration in the air. Since their breathing zone is closer to the floor than older children and adults, they would inhale more particulate matter. Indoor dust is a main reservoir for many toxic chemicals, particularly semi-volatile organic compounds (SVOCs) (32, 117, 118). When resuspended particles are inhaled, toddlers are exposed to the toxic chemicals which may affect their developing respiratory systems. Accurate determination of a toddler's inhalation exposure to indoor dust and the associated SVOCs is needed to assess the risk of potential adverse health effects.

Toddlers' inhalation exposures to particles have typically relied on stationary sampling or collection of settled dust samples, since it is difficult to collect toddlers' personal exposure samples due to the weight of sampling devices and ethical concerns (87, 119, 120). However, these sample types may result in inaccurate exposure estimates because particle size distributions and particle mass concentrations vary among the different sources of dust, PM concentrations in a toddler's breathing zone and stationary samples collected from the corner of a room. A series of studies have been completed using a

robotic surrogate, the Pre-toddler Inhalable Particulate Environmental Robotic (PIPER), to simulate a toddler's movement indoors and measure PM and bioaerosols concentrations in a toddler's breathing zone. These studies found significantly elevated levels of PM and bioaerosols in samples mounted on PIPER compared to those collected from a stationary location, indicative of particle resuspension generated when a toddler moves (92, 94, 95, 97, 98). Among these studies, particle resuspension resulting from toddlers' indoor movements and the differences in particle size and mass concentrations between mobile and stationary samples were evaluated (94, 95). However, a comparison of particle sizes between resuspended dust and settled dust has not been characterized. As one of the sources for particles, settled dust has been suggested to be used for exposure estimation (28). Thus, investigation of particle size distributions in settled dust and the relationship between particle size distributions of settled dust and resuspended dust are critical. Currently, no relevant studies were found in the literature. To fill this knowledge gap, we conducted controlled studies in an empty office and used a robot to mimic a toddler's indoor movements in order to resuspend dust. Mobile air, stationary air and settled dust samples were collected and their particle size distributions were measured. This manuscript reports the particle mass, number concentrations and particle size distributions in settled and resuspended dust in order to better characterize toddlers' potential inhalation exposure to household dust.

3.2. Methods

An empty office with a floor area of 11.9 m^2 was used. The floor was fully covered with vinyl flooring (Traffic Master Black and White Decorative Paver, Shaw Industries, Inc.,

Dalton, GA, USA) or medium pile carpet (Hot Shot II - Color Tuscan Texture, Engineered floors LLC, Dalton, GA, USA). The same piece of vinyl flooring was used for all vinyl floor experiments. It was thoroughly cleaned between experiments. A new piece of carpet was installed for each carpeted floor experiment. Details of the experimental setting have been previously described (121).

Dust for experiments was obtained from 16 residential houses in New Jersey. The dust was sieved through a 125 μm mesh to remove larger particles and fibers. A target floor dust loading of 1 g/m^2 and 10 g/m^2 was selected for vinyl floor and carpeted floor, respectively, based on reported ranges in U.S. homes (27, 28). Dust was sprayed using a 25 mL midget impinger (Pyrex) while slowly walking across the room in a "z" pattern to provide a layer of evenly deposited dust at the target loadings. To spray the dust, the upper inlet of the impinger was connected to a pump that provided an airflow of 26 L/min, which was sufficient to re-suspend the dust from the bottom of the impinger and then sprayed out the side arm. After the dust was sprayed, the room was left undisturbed overnight to allow the dust to settle completely. For the carpeted floor, settled dust was embedded into the carpet following ASTM method F608-13. Briefly, a 35-pound floor roller (Roberts Model 10-935) was dragged over the entire floor for 30 strokes (a movement in one direction is one "stroke") with the handle held at an angle of 30-45° to the floor and at a walking speed of around 0.55 m/s. The dust was left undisturbed prior to running any experiments. The experiments conducted on the vinyl and carpeted floors were repeated three and five times, respectively.

3.2.1. Particle mass concentration measurements

Stationary and mobile PM_{10} air samples were collected at 80 cm above the floor, an average toddlers' breathing zone height. Stationary PM_{10} samples were collected in one corner of the room using 37 mm Teflon filters, pore size 2.0 µm (Pall Corporation, Port Washington, NY, USA) using a Personal Environmental Monitor (PEM) sampling head (SKC Inc., Eighty Four, Pennsylvania, USA 10-µm cut-point at 10 L/min) with a Model 400 Micro-Environmental Monitor (MEM™) (MSP Corporation, Shoreview, MN, USA) operated at a flow rate of 10 L/min.

Mobile PM_{10} samples were collected using the same type of PM_{10} sampler with an SKC Leland Legacy pump (Eighty Four, Pennsylvania, USA) operated at a flow rate of 10 L/min. The pump and PM_{10} sampler were mounted on a robot, Recon Rover, with the sampler head attached at a height of 80 cm. The robot, ReCon 6.0 Programmable Rover from SmartLab Toys (https://smartlabtoys.com/products/recon-6-0-programmable-rover, Bellevue, WA, USA), was used to simulate toddlers' indoor movements while carrying the mobile air samplers. It was programmed to go in a spiral pattern in the room at a speed of 8 cm/s (fixed setting speed of the robot). During the experiments, the researcher stayed in the experimental room to restart the robot if it became stuck. The stationary and mobile samples were collected for four hours or eight hours each day for the vinyl floor and carpet experiments, respectively. The pump flow rate was verified before and after sampling using a DryCal® DC-Lite primary flow meter (MesaLabs, Butler, NJ, USA). All filters were equilibrated for a minimum of 72 hours prior to weighing in a weighing room that was maintained at a constant temperature (22-24°C) and relative humidity (30-

40%). Filters were weighed using a Mettler Toledo MT5 Microbalance (Columbus, OH, USA) and placed in individual containers until loaded into the samplers. After sample collection, filters were returned to the same weighing room to equilibrate at the same temperature and humidity for a minimum of 72 hours prior to reweighing with the same microbalance (97).

3.2.2. Particle size distribution measurements

An Aerotrak handheld optical particle counter (OPC) 9306-V2 was used to measure particle size distributions of the mobile and stationary samples. It measures the number of particles per cubic meter in six size channels (0.3-0.5 μm, 0.5-1.0 μm, 1.0-2.5 μm, 2.5-5.0 μm, 5.0-10.0 μm and >10 μm) as one-minute averages in each channel and operates at a sampling flow rate of 2.83 L/min.

The sampling timeframe is presented in Figure 3.1. Before starting the robot, background particle number concentrations were measured by placing the OPC in the middle of the room with the inlet at a height of 80 cm (a toddlers' breathing zone height (76). The OPC was then turned on and run for 30 minutes with a 6-s delay between measurements, resulting in 26 one-minute particle number concentration averages.

The OPC was placed on the ReCon Rover with the sampling inlet, a PEM without the impact ring and filter installed connected to the OPC using Tygon tubing at 80 cm to collect particle size distribution information for the mobile sample. The OPC was turned on 30 minutes after the ReCon began moving in the room. A 30 minute period was chosen to provide sufficient time for the particle number concentration to reach a steady

state concentration (94). Thatcher et al. (26) found that indoor particle concentrations associated with dust resuspension did not increase substantially between samples taken at 5 and 30 minutes after a home is occupied. In our experiment, the particle size distribution was measured for 60 minutes using the mobile platform with a 6-s delay between measurements, resulting in 52 one-minute particle number concentration averages. Following the mobile sample measurement, the OPC was placed at a stationary sampling location in one corner of the room with the inlet at a height of 80 cm with the robot running in the room. The OPC was run for 60 minutes with a 6-s delay between measurements, resulting in 52 one-minute particle number concentration averages.

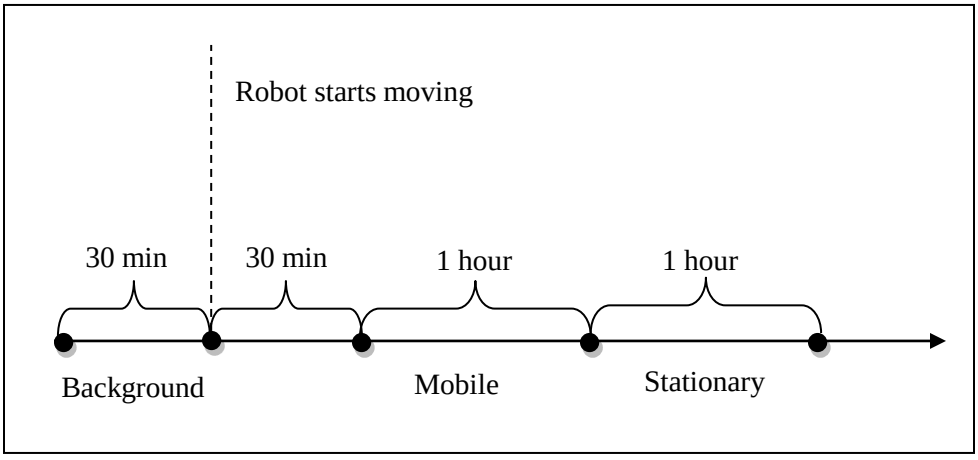


Figure 3.1. Sampling timeframe for particle size distribution measurement

Settled dust samples were collected overnight on six 25mm Teflon filters, pore size 0.5 μm (Pall Corporation, Port Washington, NY, USA) by placing them on the floor (3 in the middle and 3 in one corner of the room) prior to dust spraying in order to determine the particle size distribution of the sprayed dust. The filters were analyzed by microscopy to

identify the particle size distribution in settled dust samples. Five replicated tests were done resulting in the collection of 30 settled dust samples. The particle size distribution collected on each filter was analyzed using a Zeiss Imager.A1 microscope (Carl Zeiss AG, Oberkochen, Germany) set at a 40x magnification. The microscope was set on dark field to avoid filter interference. Transmitted light mode was used and the light setting was 90% of the total available light. At least 60 images, corresponding to 60 fields on each filter, were taken with subsequent analysis using ImageJ, an image-processing software developed by the National Institute of Health (https://imagej.nih.gov/ij/).The settled dust distribution was only determined for the vinyl floor experiments because the higher dust loading used for the carpeted floor experiments resulted in dust coagulation and too high of an accumulation on the filter paper to be counted accurately using the microscope technique.

3.2.3. Resuspension rate estimates

When settled dust is disturbed by human activities, the extent of resuspension depends on floor types. The term "resuspension rate" is used to describe the fraction of dust particles removed from the surface per unit time (122, 123). The equation for calculating resuspension rate is as follows:

Equation 3.1. $r = \frac{R}{0.2 \times L}$

Where r is the resuspension rate, hr^{-1};

R is the resuspension flux, $(g/m^2)/hr$;

L is the surface loading of the dust, g/m^2.

To calculate the resuspension flux R, we used the following equation:

Equation 3.2. $R = \frac{C*H}{t}$

Where C is the airborne particle mass concentration, in our case PM_{10}, in g/m^3;

H is the sampling height, 0.8 m;

t is the time, hr.

Two approaches were considered for the dust surface loading, L: 1) the concentration of dust loaded onto the surface, and 2) the concentration of dust on the surface available for resuspension. For the first approach, we calculated L as the amount of sprayed dust divided by the room area. For the second approach, we used the amount of dust collected in the wipe and vacuum dust samples divided by the sampling area. In both cases, a factor of 0.2 was applied to the measured mass. 0.2 was estimated based on weight measurements of size-separated dust fractions for house dust in previous literature (90).

3.2.4. Data analysis

The OPC was connected to Tygon tubing and a Personal Environmental Monitor (PEM) without the impactor ring and filter installed at the toddlers' breathing zone height (80cm). It is possible that the sampling efficiency of particle number concentration might be altered when the air passed through the OPC sampling line. The PEM is designed to operate at 10 L/min for PM_{10} while the OPC provides a flow rate of 2.83 L/min. This changes the particle sampling efficiency. To assess the potential change, the sampling efficiency was calculated for the OPC sampling line in our study using Aerocalc (TSI,

Inc. Shoreview, MN, USA. http://www.tsi.com/SiteSearch.aspx?q=Aerocalc&page=1&count=15&folderId=588&orderBy=prodOrder). Details can be found in Appendix M. We found no change in the sampling efficiency for particles smaller than 5 µm (approximately 100%). The sampling efficiency for 5 µm particles was around 90% while for 10 µm particles it was between 60% to 80%. To correct for possible particle losses, a correction factor was calculated as the reciprocal of the sampling efficiency. Average correction factors of 1.1 and 1.3 were applied to the stationary and mobile measured particle number concentrations, respectively, for particles ranging in size from 5-10 µm. Since background particle number concentrations were measured following the same method as the stationary measurements, we applied the correction factor of 1.1 for particles ranging in size 5-10 µm. All particle number concentration calculations presented in the dataset were corrected. Mean particle number concentrations were calculated for each particle size bin for a 30-min background sample, 60-min mobile sample and 60-min stationary sample collected on the carpet and vinyl floors. Normality tests on the particle number concentrations for each particle size bin showed that they were normally distributed.

To distinguish between background particles and resuspended particles and to better characterize resuspension, background particle number concentrations were subtracted from mobile and stationary particle number concentrations. A value of zero was used for the number of resuspended particles in a size bin when the background value exceeded the counts measured when the robot was moving. Background subtracted mobile and stationary particle number concentrations are plotted in Figure 3.2. Particles smaller than

1 μm were not elevated compared to the background samples, indicating that little if any dust was resuspended in that size range.

PM_{10} mass concentrations for mobile and stationary samples were estimated for each particle size range as the product of the assumed volume of the particles and an assumed particle density of 1.65 g/cm^3 (124). The background particles were not subtracted when estimating the PM_{10} mass concentrations since the filter collected both the background particles and resuspended particles. The particle volume within each size channel was based on the assumption that the particles were spheres with a diameter equal to the arithmetic mean of the size range. The masses in channels 1-5 (particle size<10 μm) were added to estimate the PM_{10} mass concentrations. Correlations between filter measured PM_{10} mass concentration and estimated PM_{10} mass concentration were evaluated using Spearman correlation tests.

For the carpeted floor, 15 complete datasets of background, mobile and stationary particle number concentrations were evaluated. For the vinyl floor, 3 sets of experiments were conducted with 9 datasets of mobile and stationary particle number concentration measurements. However, since only one set of experiments included measurements of the background particle number concentrations, only 3 complete datasets of background, mobile and stationary particle number concentrations were evaluated and compared with the settled dust particle size distributions. In addition, the OPC measured particle sizes ranged from 0.3 to 10 μm while the microscopy analysis had a lower limit of particles >1 μm. Therefore, three particle size channels for the OPC were selected: 1-2.5 μm, 2.5-5 μm, and 5-10 μm for the comparison. A particle fraction for a specific size range was

calculated as the percentage of particle number concentrations in that specific particle size range divided by the particle number concentrations summed over three particle size bins: 1-2.5 µm, 2.5-5 µm, and 5-10 µm. For the carpeted floor experiments, ANOVA tests and Tukey's HSD tests were performed to evaluate if significant differences in particle size distributions existed among the different sampling approaches. For the vinyl floor experiments, unpaired t-tests between each pair of measurements (mobile, stationary and settled dust) were performed since the sample size (N=3) was too small to perform ANOVA tests.

3.3. Results

3.3.1. Particle number concentrations in mobile and stationary samples

Correction factor applied and background subtracted mobile and stationary particle number concentrations measured in the carpeted floor experiments are plotted in Figure 3.2. Paired t-tests between mobile and stationary particle number concentrations showed that for particle size ranges 2.5-5 µm and 5-10 µm, the mobile particle number concentrations were statistically significantly higher than stationary particle number concentrations. No statistically significant differences were found for particles in the size range of 1-2.5 µm. This is consistent with toddlers' movements being the source of the larger size (>2.5 µm) resuspended dust particles. For particle number concentrations measured in vinyl floor experiments, paired t-tests showed no statistically significant difference for particles across all size ranges, but the N was small.

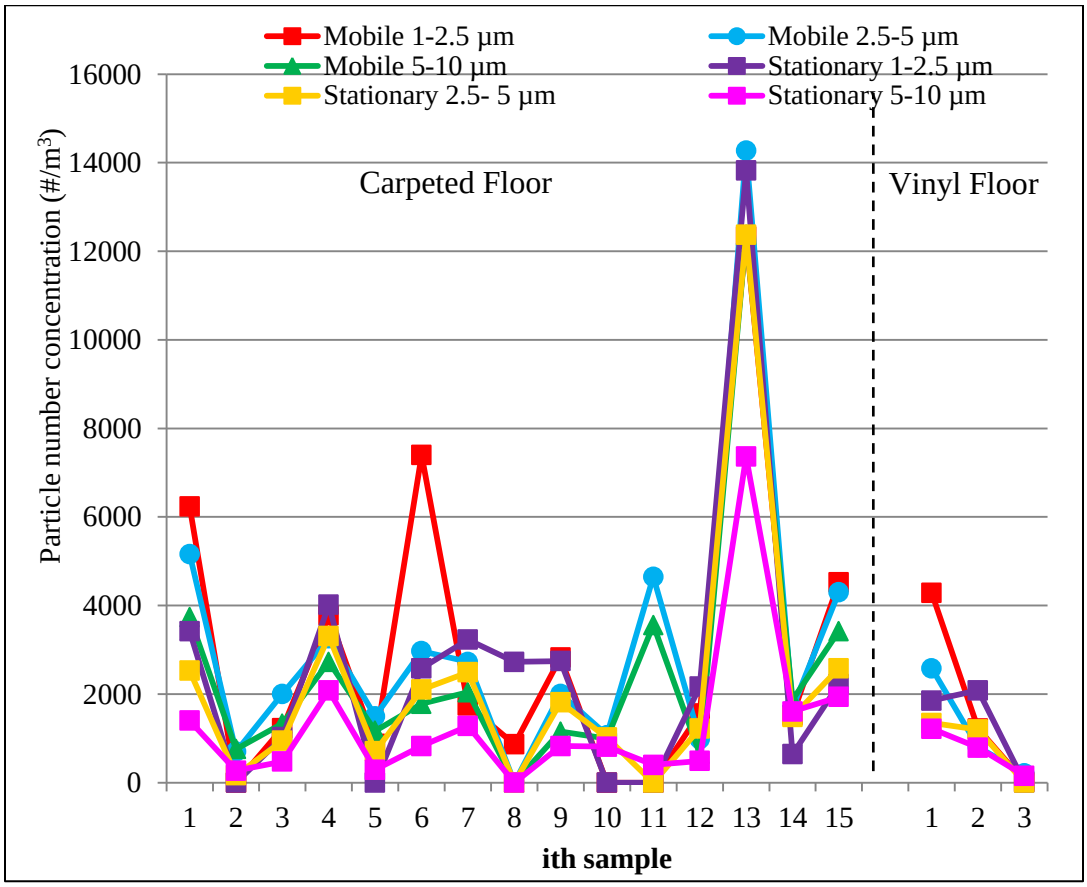


Figure 3.2. Resuspended dust particle number concentrations (#/m^3) in vinyl and carpeted floor experiments.

3.3.2. Particle mass concentrations (µg/m^3)

Measured and estimated particle mass concentrations (mean ± standard deviation) in stationary and mobile samples from the vinyl and carpeted floor experiments are presented in Table 3.1. The measured and estimated particle masses in the mobile samples exceeded the stationary samples for both the vinyl and carpeted floor surfaces.

Paired t-tests for the stationary and mobile air samples showed statistically significant differences between the measured mobile and stationary pairs for both the vinyl and carpeted floors and in estimated mobile and stationary pairs for the carpeted floor ($p<0.05$), but not for the vinyl floor experiments ($p>0.05$).

Table 3.1. Measured and estimated particle mass concentrations in stationary and mobile samples ($\mu g/m^3$)

			Mean ± standard deviation	Paired t-test p-value
Vinyl floor (N=9)	Measured	Stationary	17.1±7.1	0.004
		Mobile	38.4±10.4	
	Estimated	Stationary	6.6±7.1	0.179
		Mobile	7.6±5.9	
Carpeted floor (N=15)	Measured	Stationary	27.0±10.4	0.001
		Mobile	40.4±19.7	
	Estimated	Stationary	7.84±6.81	0.004
		Mobile	12.1±10.8	

The Spearman correlation coefficients between the estimated and measured PM_{10} mass concentrations for the carpet stationary, carpet mobile, vinyl stationary and vinyl mobile samples were all positive (Table 3.2). However, only the correlation between the OPC estimated PM_{10} mass concentrations and filter measured PM_{10} mass concentrations for carpet stationary samples was statistically significant ($p<0.05$).

Table 3.2. Spearman correlation coefficients: measured particle mass concentrations vs. estimated particle mass concentrations based on OPC data

	Carpet		Vinyl	
	Stationary	Mobile	Stationary	Mobile
	Measured vs. Estimated	Measured vs. Estimated	Measured vs. Estimated	Measured vs. Estimated
P-value	0.0164	0.5075	0.5165	0.1250
rho	0.6071	0.1857	0.25	-0.55

3.3.3. Resuspension rate of PM_{10} from vinyl and carpeted floors

Estimated resuspension rates of PM_{10} from the vinyl and carpeted floors are presented in Table 3.3. Resuspension rates of PM_{10} for total dust loading concentrations were $(8.7\pm2.0) \times 10^{-3}$ hr^{-1} from the vinyl floor and $(9.7\pm4.7) \times 10^{-4}$ hr^{-1} from the carpeted floor. The estimated resuspension rates of PM_{10} for the concentration of dust on the surface available for resuspension were $(1.6\pm0.7) \times 10^{-2}$ hr^{-1} and $(2.4\pm1.8) \times 10^{-2}$ hr^{-1} for the vinyl and carpeted floors, respectively.

Table 3.3. Estimated resuspension rate for vinyl and carpeted floors (hr^{-1})

	Resuspension rate (hr^{-1})	
	Loaded dust	Available dust
Vinyl floor	$(8.7\pm2.0) \times 10^{-3}$	$(1.6\pm0.7) \times 10^{-2}$
Carpeted floor	$(9.7\pm4.7) \times 10^{-4}$	$(2.4\pm1.8) \times 10^{-2}$

3.3.4. Comparison of particle size distribution in stationary, mobile and settled dust samples

Particle fractions for different particle size bins (1-2.5 µm, 2.5-5 µm and 5-10 µm) for settled dust, stationary and mobile samples from the vinyl and carpeted floor experiments are presented in Table 3.4. For the vinyl floor experiments, unpaired t-tests between each pair of measurements (mobile, stationary and settled dust) showed that mobile and stationary samples have statistically significantly higher percentages of smaller particles (1-2.5 µm) than the settled dust samples. Mobile and stationary samples have statistically significantly lower percentages of particles (2.5-5 µm) than the settled dust samples. No significant difference was found between mobile and stationary samples for particles size bins 1-2.5 µm and 2.5-5 µm. No significant difference was found between mobile, stationary and settled dust measurements for particles in the range of 5-10 µm.

For the carpeted floor experiments, ANOVA test and post hoc Tukey's test results showed that mobile and stationary samples have statistically significantly higher percentage of smaller particles (1-2.5 µm) compared with settled dust samples. For the carpeted floor, the settled dust samples have a higher percentage of larger particles (5-10 µm). There were no statistically significant differences between the mobile and stationary particle number distributions for either floor type.

Table 3.4. Percentage of particle number concentrations in difference particle size bins for mobile, stationary and settled dust samples in the vinyl and carpeted floor experiments. (Mean ± standard deviation)

Vinyl floor	1-2.5 µm	2.5-5 µm	5-10 µm
Mobile	31.2±24.9	28.3±1.4	40.5±25.6
Stationary	31.0±27.2	20.0±17.3	49.0±44.4
Settled dust	11.4±6.4	39.2±6.9	48.9±10.0
Unpaired t-test p-value	M vs.St:0.9884 M vs. Se:0.0007 St vs. Se: 0.0013	M vs.St:0.4298 M vs. Se:0.0121 St vs. Se: 0.0003	M vs. St:0.7918 M vs. Se:0.2525 St vs. Se: 0.9886
Carpeted floor	1-2.5 µm	2.5-5 µm	5-10 µm
Mobile	34.2±25.7	36.4±13.2	29.4±13.2
Stationary	34.8±27.6	35.1±17.4	30.1±24.5
Settled dust	11.4±6.4	39.2±6.9	48.9±10.0
ANOVA test P-value	<0.001	0.5151	<0.001
Tukey's HSD test P-value	M vs St:0.90 M vs Se: 0.001 St vs Se: 0.001		M vs St:0.90 M vs Se: 0.001 St vs Se: 0.001

Note: M—Mobile sample, St-Stationary sample, Se-Settled dust

3.4. Discussion

3.4.1. Particle number concentration in stationary and mobile samples

Higher particle number concentrations were found in most mobile samples (>90%) compare to stationary samples for three particle size bins (1-2.5 μm, 2.5-5 μm, 5-10 μm) for both vinyl and carpeted floor experiments, indicating that particles larger than 1 μm can be resuspended effectively. This is consistent with several studies evaluating dust resuspension. Sagona et al. (94) reported that there was significantly greater resuspension of particles larger than 2.5 μm in samples collected by PIPER compared to stationary measurements in 71% of carpeted homes. Other studies found that children playing indoors resuspend particles >1 μm (125) and particles 5-25 μm can be most readily resuspended indoors while particles smaller than 1 μm were not likely to be resuspended (26). The higher particle concentrations in mobile samples compared to stationary samples implies that using particle number concentrations measured using stationary samples will underestimate young children's exposure to particles associated with dust resuspension.

3.4.2. Measured and estimated particle mass concentration (μg/m^3)

Several studies by Shalat et al. attempted to estimate toddlers' breathing zone PM concentrations using a robotic approach. Shalat et al. (96) examined 13 paired mobile and stationary samples collected from 7 residences using PIPER. They found means of 98.6 μg/m^3 and 49.8 μg/m^3 for mobile and stationary airborne inhalable particle concentrations, respectively. In a second study using PIPER in 55 homes in central New Jersey (97),

mean PM_{100} levels were 30.7±13.5 μg/m^3, 41.5±27.2 μg/m^3, 34.6±15.8 μg/m^3, and 95.6±85.5 μg/m^3 for stationary bare floor, stationary carpet, mobile bare floor and mobile carpet samples, respectively. Their measured PM air concentrations were higher than what we observed. Our lower observed PM values likely reflect our measuring PM_{10} ($d < 10$ μm) rather than inhalable particles ($d<100$ μm). The stationary particle mass concentrations were statistically significantly lower than that measured using PIPER on carpeted and bare floors (96, 97).

Sagona et al. (94) compared the measured inhalable PM mass concentrations with estimated PM_{10} mass concentrations for stationary and mobile measurements in 65 homes with bare and carpeted floors, and found a statistically significant positive correlation between the measured and estimated particle mass concentrations. While we found the same trend, most of our comparisons were not statistically significant. There are several possible reasons for the weaker association we observed. First, the measured particle mass concentrations were sampled for a much longer time (8 hours for carpeted floor measurements and 4 hours for vinyl floor measurements) than the OPC measurements (1 hour for mobile and 1 hour for stationary particle number concentrations). Our mismatched sampling timeframes could contribute to the variations in measured and estimated particle mass concentrations. Second, while the Personal Environmental Monitor used for measuring PM_{10} mass concentrations had a cut-off size at 10 μm, some particles larger than 10 μm may have been collected, while the estimated particle mass concentrations were calculated solely on particles smaller than 10 μm. This might lead to an underestimation of particle mass concentrations. Future parallel particle number

concentration measurements for mobile and stationary samples are recommended. Paralleled OPC and filter measurements are suggested as well. Our sampling line for the OPC resulted in the need to correct for aspiration efficiency, which could increase the uncertainty in the particle number counts. An improved OPC sampling line is needed.

3.4.3. Particle resuspension rate

The estimated resuspension rate was higher for the vinyl floor than the carpeted floor under scenario one, i.e. when the loaded dust concentration was used in the estimation, even though the loaded dust concentration for the carpeted floor was approximately 10-fold higher than that for the vinyl floor. On the other hand, when the resuspension-available dust concentration was applied in the estimation, we found that the carpeted floor had a higher estimated resuspension rate. These data suggest that flooring plays an important role in resuspension rates. The data also indicate that only a fraction of the dust loading on the carpeted floor is resuspended. In fact, going from scenario one to scenario two, the estimated resuspension rate for the vinyl floor increased by 1.8-fold, while it increased by approximately 25-fold for the carpeted floor. Some limitations are worth noting. The dust spraying on the vinyl floor was not perfectly uniform. We found a lower dust loading measured by wipe samples than the actual amount of dust added per area, which might contribute to the overestimation of the particle resuspension rate for the vinyl floor. For the vacuum dust samples collected on the carpeted floor, some of the dust may have adsorbed onto the dust collector, sampling hose, etc. Thus, the estimated resuspension-available dust loading would be lower than the actual amounts, which resulted in overestimating the resuspension rate for the carpeted floor. Another potential

reason for the estimated amount of resuspension-available dust being lower than the amount available is as the dust was removed from the carpet by vacuuming, the amount remaining that could be resuspended declined.

Resuspension rates measured in our studies were consistent with those presented in the literature. Qian et al. (126) reported an average estimated PM_{10} resuspension rate of (1.4 ± 0.6) × 10^{-4} hr^{-1} in 14 resuspension experiments in a single family residence with a person walking on a carpeted floor. Qian et al. (122) estimated resuspension rates ranging from 10^{-5}-10^{-2} hr^{-1} across several particle size ranges: 0.8-1 µm, 1-2 µm, 2-5 µm, and 5-10 µm, with higher resuspension rates associated with larger particles.

3.4.4. Particle size distribution in stationary, mobile and settled dust samples

No studies were identified in the literature that compared particle size distribution for indoor resuspended dust and settled dust or particle size distributions measured by optical particle counter and by microscopy analysis. Several studies reported particle size distribution of indoor particles using particle number concentrations or particle mass concentrations (Table 3.5). To compare our measured data with available literature, we applied methods discussed above to convert between particle number concentrations and particle mass concentrations and further calculated percentage of particles size bins. Consistent with the literature, we found that particles < 10 µm occupied more than 90% of total resuspended particles.

Table 3.5. Particle size distribution in resuspended particles and settled dust in previous studies

Indoor media	Particle size range (Percentage of particles)	Reference
Resuspended particle	1-2.5 µm (45.9%) 2.5-5 µm (34.2%) 5-10 µm (10.4%) 10-25 µm (9.4%)	Montoya et al. (127)
Resuspended particle	1-2 µm (40.5 %) 2-5 µm (47.9%) 5-10 µm (8.8%) 10-20 µm (2.8 %)	Mohammed et al. (128)
Resuspended particle	1-5 µm (92.3%) 5-10 µm (7.2%) >10 µm (0.5%)	Thatcher et al. (26)
Settled dust	<44 µm (18%) 44-149 µm (58%) 149-177 µm (4.5%) 177-246 µm (2.7%) 246-392 µm (6.1%) 392-833 µm (11%)	Hee et al. (129)
Settled dust (pre sieved 2 mm)	<30 µm (0.3-24%) 30-63 µm (6-35%)	Seifert et al (43)

Generally toxic semi-volatile chemicals in dust increase as the particle size decreases since smaller particles have a larger surface area to volume ratio enabling smaller particles to adsorb more semi-volatile contaminants (91, 130). Lewis et al. (90) measured

pesticides and polycyclic aromatic hydrocarbon (PAHs) concentrations in seven size fractions ranging from <4 to 500 μm from residential house dust and found that the concentrations of nearly all of the target analytes increased with decreasing particle size. Yang et al. (131) analyzed the concentration of 10 organophosphate flame retardants (OPFRs) in the suspended particles with different diameters and found that the detected OPFRs mainly located on particles <2.5 μm. A larger percentage (around 60%) of smaller particles (1-2.5 μm) were detected in the resuspended particles and were significantly higher than the settled dust in our study, which is expected to result in higher concentrations of contaminants being present in resuspended particles than settled dust. Thus, using settled dust would underestimate human's inhalation exposure to semi-volatile contaminants in the house dust.

3.5. Conclusions

This study evaluated the differences in particle number and mass concentrations measured by mobile, stationary and settled dust samples to assess toddlers' inhalation exposure to particles and the potentially to contaminants attached to those particles. Particle mass concentrations measured in mobile samples were significantly higher than those measured in stationary samples. As a result, using stationary sampling would underestimate toddlers' inhalation exposures to particles and contaminants attached to those particles. Particle size distributions in mobile and stationary samples were not statistically significantly different from each other. However, settled dust samples had significantly higher percentages of large particles (5-10 μm) and lower percentage of

small particles (1-2.5 μm). Therefore, using settled dust as an indicator of young children's exposures would underestimate their exposure to toxic chemicals.

Chapter 4

A Probabilistic Assessment of Toddlers' Inhalation Exposure to Permethrin in House Dust

4.1. Introduction

Early exposures to contaminants might have significant impact on children's health (132). Toddlers, 1 to 3 years old, spend more than 90% of their time at home (60) and their on-floor activities generate elevated particle concentrations in their breathing zones due to dust resuspension (94, 96). The "personal dust cloud" created around them leads to an increased inhalation exposure to particles as well as to the contaminants the particles contain, especially SVOCs with a high partition coefficient between solid phase and gas phase. Toddlers are at an early stage of physiological development and their lung systems are more vulnerable to exposures to irritants and sensitizing agents than older children and adults (74, 75). Adams et al. (133) studied 10,061 cases of pesticide-related investigations in the UK from 2004-2007 and of the 2364 suspected exposure events, 1162 involved children with 60.5% of those children being less than 2 years old. The most common scenario for acute pesticide exposure occurred shortly after application (28.7%). A study by Spann et al.(134) assessed acute hazards to 7,434 children younger than 6 years old and found that children 2 years old and younger were the predominant age group exposed (75%) to different contaminants.

Pyrethroids comprise a group of pesticides frequently detected and used indoors (29). Exposure to pyrethroids can cause a series of acute symptoms: dyspnea, coughing and bronchospasm, nausea and vomiting, as well as dermal effects (45, 48). Ujvary (135) reported that pyrethroids can cause local paresthesia and allergies via inhalation or direct dermal exposure. A study by Wang et al.(136) on organophosphate and pyrethroid pesticides exposures of 406 children aged 3-6 years old from Nanjing, China indicated that organophosphates and pyrethroids may have a significant association with children's memory and verbal comprehension. Studies have shown positive associations between indoor pyrethroid pesticides exposures and childhood leukemia (137-140). In addition, the toxicity of pyrethroids is age dependent, with younger age groups being more vulnerable (141, 142). Shafer et al. (141) reported that the magnitude of age-related toxicity of pyrethroids was much larger than for other pesticide classes. Cantalamessa (142) reported a 6-fold acute lethality of permethrin in PND8 rats compared with adult rats.

Residential use of pyrethroids is the major source of young children's pyrethroid exposure. Acute inhalation exposure is a major concern after pesticide applications inside residential settings. Several animal studies have identified toxic effects of pyrethroids on brain function, hormonal abnormalities, liver and kidney damage/dysfunction, decreased pulmonary function (80, 82, 143-148). U.S EPA (149) reported a no observed adverse effect level (NOAEL) of 0.042 mg/L for short-term inhalation exposure of permethrin based on clinical signs (tremors and hypersensitivity). Thus, the level of concern due to

inhalation exposure to permethrin is equal to 0.42 µg/L for children <6 years old with an uncertainty factor of 100 applied.

There is a knowledge gap in toddlers' inhalation exposures to pyrethroids indoors, due to the difficulty of collecting personal air samples from this age group. Therefore, most studies used stationary sampling or settled dust samples to estimate young children's inhalation exposures to pyrethroids or other contaminants (26-28). However, these approaches underestimate exposure since they do not consider the impact of toddlers' behaviors and activities on exposure levels (94, 121). In this study, Monte Carlo simulation was performed to probabilistically assess toddlers' inhalation exposures to pyrethroids in house dust taking toddlers' floor activities into consideration. The overall objective was to estimate the distribution of toddlers' inhalation exposures to pyrethroids in the residential environment and how toddlers' activities impact those exposure estimates.

4.2. Methods

4.2.1. Monte Carlo simulation

Monte Carlo simulation was selected to probabilistically estimate the distribution of toddlers' inhalation exposures to pyrethroids in residential houses. It selects a random value from the given distribution of parameters to estimate outcomes. The result of the model is recorded and the process is repeated hundreds or thousands of times, using different randomly-selected values from the distribution. In this way, a large number of results are obtained from the model at the completion of the simulation. The simulation

provides a distribution of outcomes, which in this study, is the distribution of toddlers' inhalation exposures to pyrethroids. Monte Carlo simulations were run 10,000 times using Crystal Ball 11.1.2.4 (Oracle Corporation, CA, USA).

4.2.2. Exposure equations

Toddlers' (age one to three years old) short term average daily inhaled dose of permethrin is computed using the following equations:

Equation 4.1.

$$Intake_{mobile} = \frac{C_{stationary} \times \left(1 - f_{onfloor}\right) \times t_{indoor} \times InhR_{off\ floor}}{1440 \times BW} + \frac{C_{mobile} \times f_{on\ floor} \times t_{indoor} \times InhR_{on\ floor}}{1440 \times BW} + \frac{C_{outdoor} \times (1440 - t_{indoor}) \times InhR_{outdoor}}{1440 \times BW}$$

Equation 4.2.

$$Intake_{stationary} = \frac{C_{stationary} \times (1 - f_{onfloor}) \times t_{indoor} \times InhR_{off\ floor}}{1440 \times BW} + \frac{C_{stationary} \times f_{on\ floor} \times t_{indoor} \times InhR_{on\ floor}}{1440 \times BW} + \frac{C_{outdoor} \times (1440 - t_{indoor}) \times InhR_{outdoor}}{1440 \times BW}$$

Equation 4.3.

$$Intake_{settled\ dust} = \frac{k \times C_{settled\ dust} \times \varepsilon}{10^3 \times BW}$$

Where

- $Intake_{mobile}$=short term inhalation intake of permethrin for a toddler via mobile sampling (μg/kg/day)
- $Intake_{stationary}$= short term inhalation intake of permethrin for a toddler via stationary sampling (μg/kg/day)
- $Intake_{settled\ dust}$= short term inhalation intake of permethrin for a toddler via settled dust (μg/kg/day)
- $C_{stationary}$= pyrethroid air concentration in stationary samples (μg/m^3)
- C_{mobile}= pyrethroid air concentration in mobile samples (μg/m^3)
- $C_{settled\ dust}$= pyrethroid concentration in settled dust (μg/g)
- $C_{outdoor}$= pyrethroid air concentration outdoors (μg/m^3)
- $f_{onfloor}$= fraction of time toddler spends on the floor, bounded between 0 and 1
- t_{indoor}= toddler's time spent indoors (min/day)
- $InhR_{on\ floor}$ = toddler's inhalation rate for on floor activities (m^3/min)
- $InhR_{off\ floor}$ = toddler's inhalation rate for off floor activities (m^3/min)
- $InhR_{outdoor}$ = toddler's inhalation rate outdoors (m^3/min)
- ε = amount of dust a toddler inhales per day (mg/day), 2 mg/day
- *k* = correction factor for permethrin concentration in settled dust

- BW = toddler's body weight (kg)

For the mobile approach, the daily inhalation intake is calculated for three microenvironments: on floor, off floor and outdoors. The on-floor part of the equation calculates a toddler's exposure to pyrethroids when playing on the floor indoors. The off-floor part quantifies the exposure when a toddler's activities were not on the floor indoors, such as resting in a bed or sitting at a table. The outdoors part calculates a toddler's exposure in the outdoor environment, though for the scenarios used the outdoor air concentration was assumed to be zero. The mobile measurements were used to estimate exposure while a toddler was involved in on-floor activities. The stationary measurements were used to estimate exposure when not on the floor. Since we are only considering a toddler's indoor inhalation exposure to permethrin, we assume that $C_{outdoor}$ is zero. Though this study focused on a toddler's indoor inhalation intake, the outdoor components were included to account for a full 24 hour day. Inhalation rates vary with a toddler's activity levels. When toddlers' are involved in on-floor activities, they potentially move faster and have a higher inhalation rate compared to off-floor activities. For the stationary approach, pyrethroids measured with a stationary sampler were used to calculate the exposure for all indoor activities. For the settled dust approach, the permethrin intake was calculated by multiplying a toddler's daily inhaled dose of dust by the permethrin concentration in dust. A correction factor, *k*, was applied in the equation to quantify the inhalation of permethrin from settled dust to airborne particles. *k* was

equal to the average ratio of stationary permethrin concentration (µg/g) to settled dust permethrin concentration (µg/g).

Several factors not evaluated here potentially impact a toddler's daily inhalation intake of permethrin, such as air exchange rate, floor cleaning, etc. However, to reduce model complexity, they were not considered in the current study.

4.2.3. Distribution development for model parameters

Model inputs were categorized into four groups: i) measured concentrations; ii) toddlers' physical characteristics; iii) time and activity-patterns; and iv) other. The distributions developed for this study were based on the experimental measurements discussed in Chapters 2 and 3 and data from the U.S.EPA's Exposure Factors Handbook 2011 Edition (20). A list of parameters is given in Table 4.1.

Table 4.1.Summary of input distributions

		Unit	Type of Distribution	Source
Measured concentrations				
Carpeted floor	Stationary permethrin air concentration	µg/m^3	Lognormal	Experimental measurement
	Mobile permethrin air concentration	µg/m^3	Lognormal	
	Settled dust permethrin concentration	µg/g	Lognormal	
Vinyl floor	Stationary permethrin air concentration	µg/m^3	Lognormal	
	Mobile permethrin air concentration	µg/m^3	Lognormal	

Settled dust permethrin concentration	μg/g	Lognormal	
Physical characteristics			
Inhalation rate for 1-2 years old (Sedentary/Passive-Light intensity)	m^3/min	Lognormal	U.S.EPA (20)
Inhalation rate for 2-3 years old (Sedentary/Passive-Light intensity)	m^3/min	Lognormal	
Body weight for 1-2 years old	kg	Lognormal	
Body weight for 2-3 years old	kg	Lognormal	
Time and activity			
Exposure time(Time spent at home) for 1-2 years old	min/day	Minimum extreme	U.S.EPA (20)
Exposure time(Time spent at home) for 2-3 years old	min/day	Minimum extreme	
Floor activity (on floor time) ratio for 1-2 years old	%	Weibull	Personal communication
Floor activity (on floor time) ratio for 2-3 years old	%	Weibull	
Other inputs			
Toddlers' daily inhaled dust	mg	Constant	Oomen et al. (89)
Correction factor for settled dust permethrin concentrations	unitless	Lognormal	Calculation

4.2.3.1. Measured concentrations

4.2.3.1.1. Permethrin concentrations

Permethrin concentrations were measured in a series of experiments that used a robot to simulate a toddler's activity (121). The sample size for experimentally measured

concentrations was 10 for the carpeted floor and 6 for the vinyl floor and the means were within approximately three standard deviations of zero. It is inappropriate to use a normal distribution since negative values are not physically meaningful. Rather a lognormal distribution is used, which is consistent with the concentrations of many pollutants in the environment (85, 150). Parameters used to describe the permethrin concentration distribution are the means and standard deviations. In addition, a boundary was used with a minimum of 0 and a maximum of 110% of the measured values.

Table 4.2. Summary of values used to fit the lognormal distributions for permethrin concentrations

	Parameter	Mean	SD	Min	Max
Carpeted floor	Stationary permethrin air concentration ($\mu g/m^3$)	0.034	0.020	0	0.085
	Mobile permethrin air concentration ($\mu g/m^3$)	0.061	0.030	0	0.116
	Settled dust permethrin concentration ($\mu g/g$)	403	237	0	1,050
Vinyl floor	Stationary permethrin air concentration ($\mu g/m^3$)	0.065	0.026	0	0.099
	Mobile permethrin air concentration ($\mu g/m^3$)	0.143	0.051	0	0.223
	Settled dust permethrin	6,429	5,628	0	16,519

concentration (µg/g)

4.2.3.2. Toddlers' characteristics

4.2.3.2.1. Inhalation rate

Previous studies indicated that a lognormal distribution should be used in modeling human's inhalation rate (81, 151). Data to generate lognormal distributions for inhalation rate were acquired from the U.S. EPA's Exposure Factors Handbook 2011 Edition (20). Short-term inhalation rate is determined largely by activity level, thus the selection of toddlers' short term inhalation rates would correspond to the toddlers' activities (walking, sleeping, sedentary, etc.). The inhalation rates for sedentary and passive activities, including sleeping activities, were selected for stationary exposure calculations, while inhalation rates for light intensity were selected for mobile exposure calculations. We define the lower boundary of our distribution as the minimum of the 5th percentile of male and female inhalation rates, while the upper boundary is defined as the maximum of male and female inhalation rates in the dataset.

Table 4.3. Summary of input values for inhalation rates (m^3/min)

Age Group	Performing Activities	Mean	95%	Min	Max
1-2 years old	Sedentary and Passive activities	4.7E-3	6.5E-3	3.2E-03	9.9E-03
	Light intensity	1.2E-2	1.6E-2	8.6E-03	2.1E-02
2-3 years old	Sedentary and Passive activities	4.8E-3	6.5E-3	3.2E-03	9.4E-03
	Light intensity	1.2E-2	1.6E-2	8.5E-03	2.4E-02

4.2.3.2.2. Body weight

Studies indicated that the distribution of body weight was skewed to the right and was not normally distributed (79, 152). A lognormal distribution was selected for body weight using data from the U.S. EPA's Exposure Factors Handbook 2011 Edition (20). We used the mean and 90th percentile to fit lognormal distributions for the two age groups (1-2 and 2-3 year olds) with the upper boundary set at the 95th percentile and the lower boundary set at the 5th percentile.

Table 4.4. Summary of input values for body weight (kg)

Age Group	Sample size	Mean	90th percentile	Min (5th percentile)	Max (95th percentile)
1-2 years old	1176	11.4	13.4	8.9	14
2-3 years old	1144	13.8	16.3	10.9	17.1

4.2.3.3. Time and activity data

In our study, we focused on toddlers' indoor time. Data were abstracted from the U.S. EPA's Exposure Factor Handbook 2011 Edition (20). The distribution for time spent indoors is extremely positively skewed and the time spent in a residence was highly variable (60). Since the data are right skewed, we selected a minimum extreme distribution with the 10th percentile and 90th percentile as input parameters, a lower boundary of 0 minutes and an upper boundary of 1440 minutes (Table 4.5).

Table 4.5. Summary of input values for time spent indoors (min)

Age group	N	Mean	Min	10th percentile	90th percentile	Max
1 to 2 years old	118	1047	0	705	1440	1440

2 to 3 years old	118	971	0	727	1232	1440

The mobile approach requires the toddlers' contact time with the floor to select the permethrin concentrations. It is critical to generate a proper distribution. Research data from a study investigating toddlers' floor activities were used to fit distributions (personal communication with Dr. Kathy Black). Briefly, in that study, children 36 months of age and younger from a small border community south of Laredo, Texas were recruited and videotaped in their home for 4 hours using a handheld camcorder. The video recordings were then analyzed for floor contact patterns. In total, thirty-three on floor activity ratios (the time toddlers spending on the floor divided by the entire recording time) for 1-2 years old and twenty on floor activity ratios for 2-3 years old were acquired. Distributions were fitted in Crystal Ball and the best fit for both age groups were Weibull distributions. The fitted parameters are presented in Table 4.6.

Table 4.6. Fitted parameters for toddlers' on-floor activity ratios (%)

Weibull distribution	Location	Scale	Shape
1-2 years old	32.7	105.3	7.2
2-3 years old	13.5	59.5	4.9

4.2.3.4. Other inputs

4.2.3.4.1. Toddlers' daily inhaled dust

There are limited publications reporting the amount of dust a child potentially inhales per day. Oomen et al. 2008 (89) estimated that children inhale 2 mg dust per day based on the assumption of a constant suspended dust particulate concentration of 100 $\mu g/m^3$, and a

volume of inhaled air of 7.6 m^3 for a child. The daily inhaled dust value (2 mg) was used to further compute the distribution of permethrin that a toddler might be exposed to via settled dust. To account for differences in the permethrin concentration in settled dust and resuspended dust due to differences in size distributions, a correction factor was applied to the permethrin concentration measured in settled dust when estimating toddlers' daily inhalation intake of permethrin. The correction factor used was calculated as the average ratio of permethrin concentrations measured in stationary samples to that measured in settled dust samples (Appendix B and C). A lognormal distribution for the correction factor is presented in Table 4.7.

Table 4.7. Calculated parameters for correction factor

	Mean	Standard deviation	Min	Max	50th percentile
Vinyl floor	1.3	1.4	0.4	4.1	0.6
Carpeted floor	4.2	1.6	2.3	7.2	4.1

4.2.4. Sensitivity analysis

The average daily inhalation intake of permethrin was selected as a metric to test the sensitivity of the outputs to the inputs and parameters. Rank correlation coefficients between every parameter and every equation were computed. The larger the absolute value of the correlation coefficient, the greater the sensitivity of the calculated dose to that specific parameter.

4.3. Results and Discussions

4.3.1. Distribution of toddlers' short term daily inhalation intake of permethrin

Cumulative probability plots of the estimated toddlers' short term inhalation intake of permethrin indoors are presented in Figures 4.1-4.4. Results for the corrected and uncorrected (k=1) settled dust approach are presented. If the mobile sample best represents a toddler's personal exposure to permethrin, then exposure estimates based on the settled dust samples (corrected and uncorrected) overestimates a toddler' inhalation exposure to permethrin while estimates from the stationary samples underestimates this value. The estimated daily inhalation intake for the vinyl floor was higher than the carpeted floor by factor of 2 to 10 when other conditions were held constant. This suggests that toddlers might have a higher inhalation exposure to permethrin in homes with vinyl floor for the same permethrin loading. The partitioning of permethrin sprayed on the floor between the deposited dust and the flooring material might contribute to the differences calculated. Shin et al. (153) indicated that there is a higher exposure concentration on vinyl surfaces since the resuspension available fraction of compounds for the vinyl surface is 100% while for the carpet floor it is only 1.5%. For the experimental data used for this study, the permethrin loading was the same, but the dust loading on the carpeted was ten times that on the vinyl floor. Thus, the permethrin settled dust concentration per gram of dust was higher for the vinyl floor (Appendix B).

The distributions of toddlers' inhalation exposure of permethrin generated in our study were similar to that reported in the literature. Li et al. (154) estimated the potential

exposure of pyrethroids through inhalation exposure during the application of various mosquito repellents indoors and reported that the toddlers' inhaled dose of pyrethroids ranged from 0.004±0.001 to 2.38±0.86 µg /kg/day during the application of various mosquito repellents in indoor environment. Schleier et al. (155) reported a total acute exposure ranging from 0.03 to 0.3 µg/kg/day for toddlers and infants exposed to aerosol applications of insecticides for managing mosquitoes considering ingestion, dermal and inhalation exposures. Furthermore, they found that the mean inhalation exposure contributed about 60% to the total exposure to adults but only contributed 8% to the total exposure to toddlers and infants. The authors stated that 60% of the exposure to the infants and toddlers were from non-dietary exposure of pyrethroids in dust from hand-to-mouth activities.

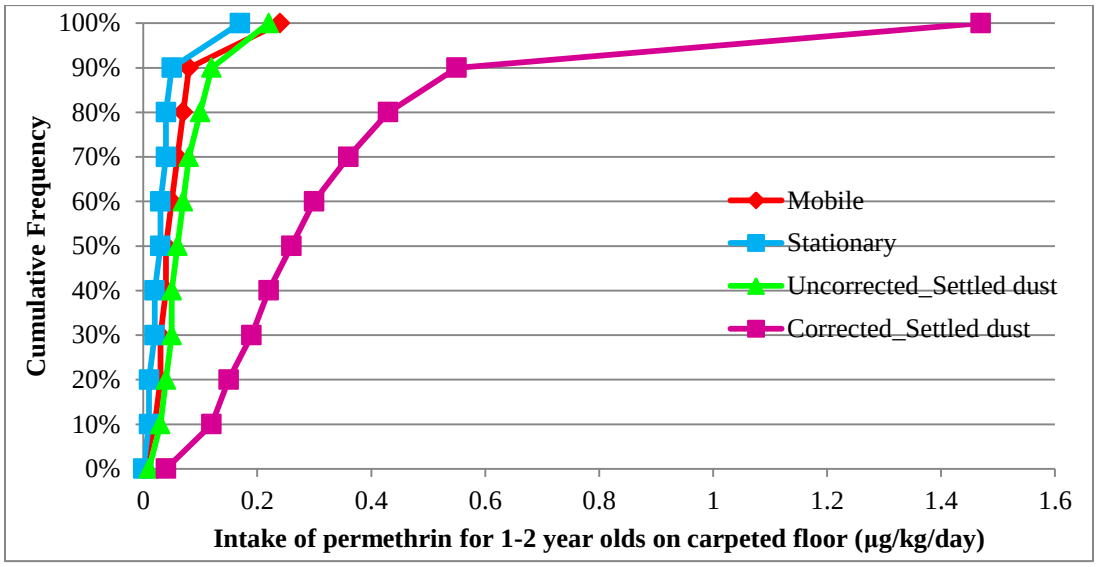


Figure 4.1. Short term inhalation exposure to permethrin for 1-2 years old on carpeted floor

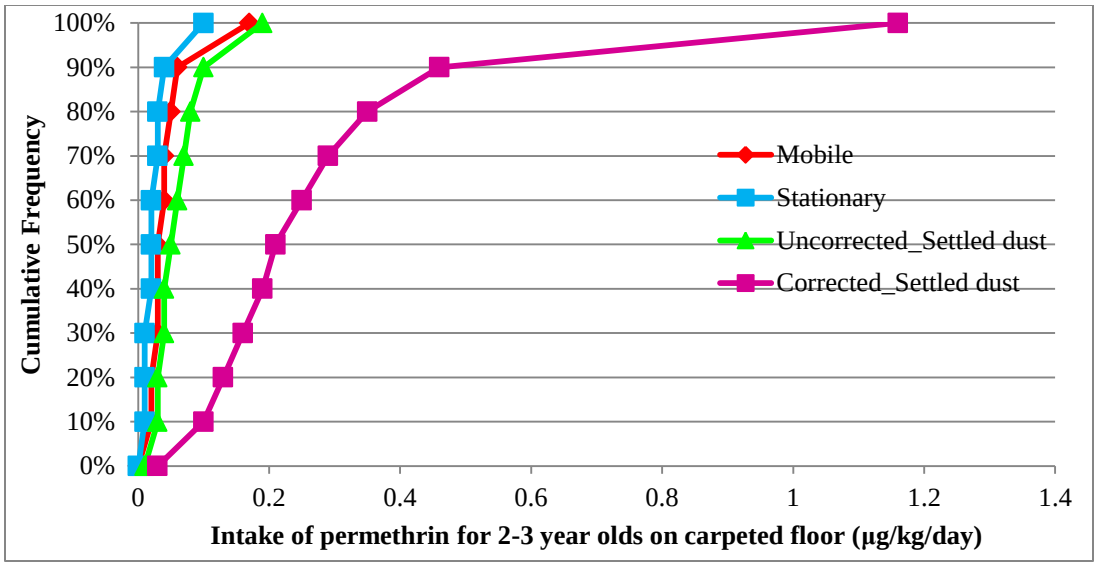


Figure 4.2. Short term inhalation exposure to permethrin for 2-3 years old on carpeted floor

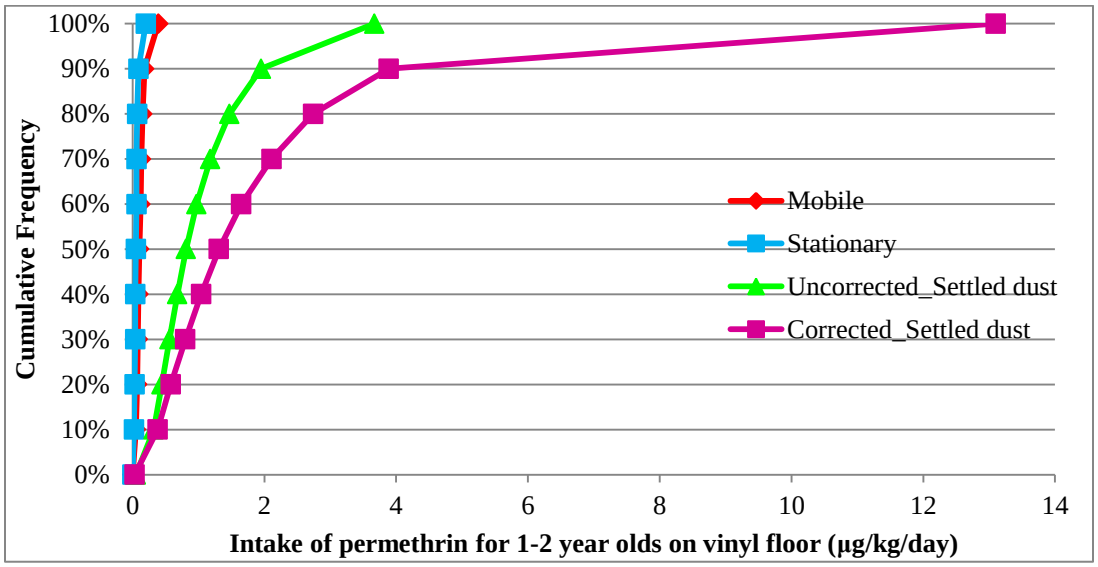


Figure 4.3. Short term inhalation exposure to permethrin for 1-2 years old on vinyl floor

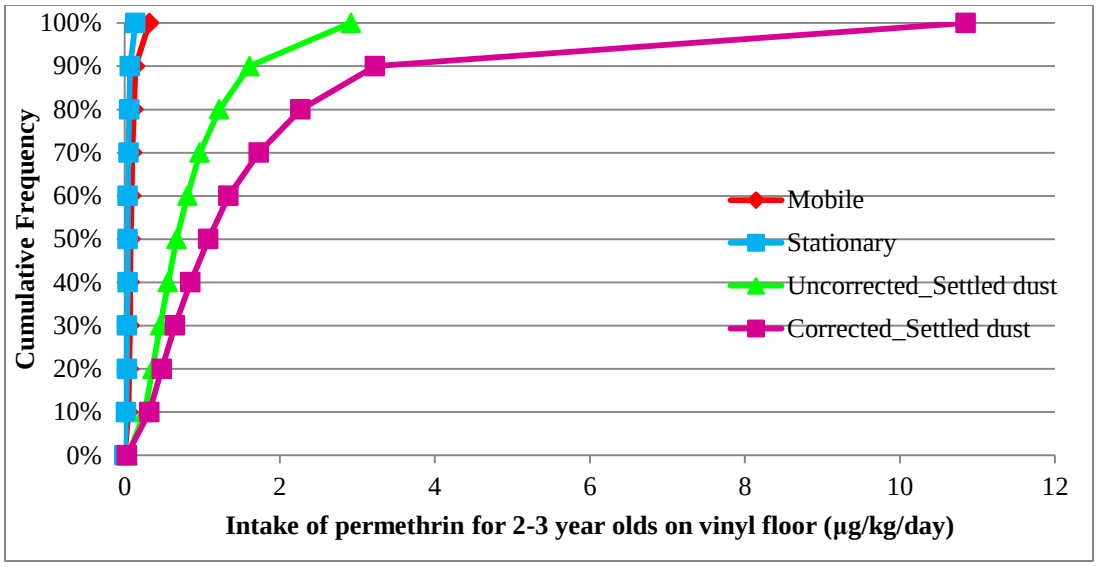


Figure 4.4. Short term inhalation exposure to permethrin for 2-3 years old on vinyl floor

Table 4.8. Statistics for estimated toddlers' short term inhalation intake of permethrin (µg/kg/day)

Floor material	Age group	Sampling method	Range	Mean	10% Percentile	50% Percentile	90% Percentile
Carpeted floor	1-2 years old	Mobile	0-0.24	0.05	0.02	0.04	0.08
		Stationary	0-0.17	0.03	0.01	0.03	0.05
		Uncorrected settled dust	0-0.22	0.07	0.03	0.06	0.12
		Corrected settled dust	0-1.47	0.30	0.12	0.26	0.55
	2-3 years old	Mobile	0-0.17	0.04	0.02	0.03	0.06
		Stationary	0-0.10	0.02	0.01	0.02	0.04
		Uncorrected settled dust	0-0.19	0.06	0.03	0.05	0.10
		Corrected	0-1.16	0.25	0.10	0.21	0.46

		settled dust					
Vinyl floor	1-2 years old	Mobile	0-0.39	0.11	0.05	0.10	0.18
		Stationary	0-0.20	0.05	0.02	0.05	0.09
		Uncorrected settled dust	0-3.67	0.98	0.32	0.81	1.95
		Corrected settled dust	0-13.10	1.79	0.38	1.31	3.89
	2-3 years old	Mobile	0-0.32	0.09	0.05	0.08	0.14
		Stationary	0-0.14	0.04	0.02	0.04	0.07
		Uncorrected settled dust	0-2.92	0.81	0.26	0.67	1.61
		Corrected settled dust	0-10.85	1.48	0.32	1.08	3.23

4.3.2. Sensitivity analysis

Sensitivity analysis was performed in Crystal Ball and the results are shown in Table 4.9 (using "Carpet_1-2 years old" as an example). Other simulations follow a similar ranking pattern. The sensitivity analysis ranks assumptions from most to least important. For the mobile samples, the measured mobile permethrin air concentration ranks as the most important (0.67), followed by time and activity profile (time spent at home) (0.48), floor activity (0.30), inhalation rates for light intensity (0.24), bodyweight (-0.19), stationary permethrin air concentration (0.10) and inhalation rates when sedentary or passive (0.05). The body weight coefficient is negative, since the inhaled dose of permethrin was calculated on a per body weight basis (Equation 4.1). For the stationary sampling approach, the impact of parameters are similar to those identified for the mobile sampling approach, except that the measured stationary permethrin air concentrations have a larger

impact on the estimated inhalation intake (0.80). For the uncorrected settled dust and corrected settled dust approach, permethrin concentrations measured in settled dust have the largest impact on the estimates (0.86 for corrected and 0.98 for uncorrected, respectively). Since the permethrin concentration is the source of the exposure, it is expected to have the greatest impact the outcome. Similarly, Wason et al. (156) found that the measured wipe/dust concentrations impacted the total absorbed dose output the most as evident from the correlation coefficients they reported ranging between 65%-87%.

Table 4.9. Rank correlations for 1-2 years old' inhalation intake of permethrin on carpeted floor

Assumptions	Rank Correlation
Mobile	
Mobile permethrin air concentration	0.67
Exposure time(time spent at home) for 1-2 years old	0.48
Floor activity (on floor time) ratio for 1-2 years old	0.30
Inhalation rate for 1-2 years old (light intensity)	0.24
Body weight for 1-2 years old	-0.19
Stationary permethrin air concentration	0.10
Inhalation rate for 1-2 years old (sedentary and passive)	0.05
Stationary	

Stationary permethrin air concentration	0.80
Exposure time(time spent at home) for 1-2 years old	0.38
Inhalation rate for 1-2 years old (light intensity)	0.21
Floor activity (on floor time) ratio for 1-2 years old	0.19
Body weight for 1-2 years old	-0.17
Inhalation rate for 1-2 years old (sedentary and passive)	0.04
Corrected_Settled dust	
Settled dust permethrin concentration	0.86
*k*_ratio of stationary to settled dust_Carpeted floor	0.45
Body weight for 1-2 years old	-0.17
Uncorrected_Settled dust	
Settled dust permethrin concentration	0.98
Body weight for 1-2 years old	-0.20

4.4. Conclusions

For the vinyl and carpeted floor experimental conditions that the model was based on, using settled dust permethrin concentrations to estimate toddlers' daily inhalation intake might overestimate the intake value, while using stationary permethrin concentrations might underestimate toddlers' daily inhalation intake compared to using mobile samples.

Chapter 5

Future Research Recommendations

1. We performed simulated studies in an empty office rather than actual houses. Field sampling of permethrin levels with toddlers' activity patterns considered are needed to further validate our results and the modeling outcomes
2. Time and activity patterns are important factors that impact young children's exposure levels. A better understanding of children's daily activity patterns can be helpful to improve the exposure estimations.
3. The robot used in our study was not programmed to closely match toddlers' indoor activities. This could have affected the amount of dust resuspended and the simulated breathing zone height. Similar studies should be performed using advanced robotic surrogates.
4. Young children's long-term or intermediate exposure to pyrethroids might impact their neurodevelopment and other health outcomes. A probabilistic assessment of long-term exposure should examine multiple health outcomes.

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Appendix A: PM_{10} concentrations ($\mu g/m^3$)

Table A1. PM_{10} concentrations ($\mu g/m^3$) in mobile and stationary samples measured in the carpeted floor and vinyl floor experiments.

Carpeted Floor			Vinyl Floor		
Sample	Stationary	Mobile	Sample	Stationary	Mobile
1	36.1	32.5	1	16.8	40.5
2	21.6	28.0	2	18.5	27.7
3	19.6	22.3	3	13.2	29.5
4	38.2	70.3	4	9.4	42.2
5	32.4	65.0	5	23.9	42.2
6	19.8	28.2	6	17.3	20.1
7	29.8	56.2	7	31.7	50.7
8	23.2	33.3	8	13.0	49.1
9	19.0	34.6	9	9.8	43.8
10	36.5	75.2			
11	24.5	32.9			
12	12.1	14.3			
13	52.5	62.8			
14	18.5	25.2			
15	22.0	25.7			
Mean	27.1	40.4	Mean	17.1	38.4
SD	10.4	19.7	SD	7.1	10.4

Appendix B: Permethrin dust concentrations (µg/g)

Table B1. Permethrin dust concentrations (µg/g) in mobile, stationary and settled dust samples.

Vinyl Floor	Mobile	Stationary	Settled dust
	7007	979	2344
	5082	5487	1341
	3018	2754	6000
	3520	3531	2389
	4138	6556	15018
	2516	9169	11482
Mean	4213	4746	6429
SD	1634	2933	5628
Carpeted Floor	Mobile	Stationary	Settled dust
	816	726	234
	865	686	155
	1609	2373	955
	1592	2436	339
	2405	1262	252
	1545	1965	666
	2023	1325	348
	2949	684	299
	2887	2021	422
	4125	2039	357
Mean	2082	1552	403
SD	1028	700	237

Appendix C: Permethrin air concentrations (µg/m^3)

Table C1. Permethrin air concentrations (µg/m^3) measured in mobile and stationary samples for the vinyl floor and carpeted floor experiments.

	Carpeted Floor		Vinyl Floor	
	Stationary	Mobile	Stationary	Mobile
	0.016	0.023	0.018	0.194
	0.013	0.019	0.072	0.150
	0.077	0.105	0.066	0.127
	0.048	0.045	0.061	0.071
	0.029	0.080	0.086	0.203
	0.037	0.053	0.090	0.110
	0.032	0.067		
	0.008	0.042		
	0.037	0.073		
	0.045	0.106		
Mean	0.034	0.061	0.065	0.143
SD	0.020	0.030	0.026	0.051

Appendix D: Particle number concentrations in background, mobile and stationary samples (#/m^3) (Correction Factor applied)

Table D1. Particle number concentrations in background, mobile and stationary samples (#/m^3)

Carpeted Floor Experiments					
Bin size (µm)	0.3-0.5	0.5-1	1-2.5	2.5-5	5-10
Background	61767	5468	1538	690	430
	143029	17447	6448	1849	610
	434262	48080	7142	1743	563
	737508	84352	9851	1949	505
	598716	69915	6842	1908	922
	1633577	146724	6737	974	372
	917741	87870	4579	830	317
	1054819	88380	7113	3921	2432
	836172	60551	2024	257	94
	1556066	179953	16707	1999	453
	695861	457071	113746	8367	1194
	178521	18410	1221	315	155

	807611	50541	3340	1912	1171
	115476	11604	2438	1423	610
	261101	17303	2328	1370	572
Mean	668815	89578	12803	1967	693
Standard deviation	492759	113275	28211	1983	577
Mobile	39763	9645	7777	5854	4168
	126262	13609	5831	2547	1368
	265045	34547	8368	3748	1896
	668182	87762	13631	5209	3234
	356514	43312	7716	3409	2093
	1872780	184677	14137	3942	2154
	626973	57399	6324	3557	2358
	860238	84549	7981	2967	1664
	654080	51246	4848	2263	1247
	1216382	141156	14723	3073	1446
	518147	394590	107496	13017	4752
	165657	20664	2781	1302	810
	674884	51079	15677	16184	13547
	210764	17194	3964	3110	2478

	235062	18969	6851	5674	3994
Mean	566049	80693	15207	5057	3147
Standard deviation	482112	99954	25852	4109	3100
Stationary	38870	8033	4959	3220	1832
	136532	14753	5041	2015	881
	173886	26765	8167	2687	1040
	659182	89864	13868	5257	2586
	296285	35093	6027	2616	1212
	1254081	114314	9321	3077	1200
	822517	79648	7808	3323	1596
	972713	107984	9840	2522	1050
	485822	38833	4771	2070	922
	1054428	119798	12317	3017	1266
	294410	199950	43866	5400	1596
	151567	19380	3390	1538	646
	628752	52904	17164	14283	8533
	284565	21670	3080	2909	2216
	225092	19028	4553	3953	2507
Mean	498580	63201	10278	3859	1939

Standard deviation	380211	54319	10143	3078	1918
Vinyl Floor Experiments					
Bin size (µm)	0.3-0.5	0.5-1	1-2.5	2.5-5	5-10
Background	71376	5500	1065	316	178
	231467	12727	932	139	52
	666463	27464	2712	803	287
Mean	323102	15230	1570	419	172
Standard deviation	307944	11194	992	344	117
Mobile	406948	21912	5349	2898	2699
	79603	8188	2152	939	709
	258697	20591	2731	1013	820
Mean	248416	16897	3411	1617	1409
Standard deviation	163914	7571	1704	1110	1118
Stationary	242489	14281	2918	1663	1396
	98253	10013	3016	1337	843
	228439	16424	1716	668	443
Mean	189727	13573	2550	1222	894
Standard deviation	79530	3264	724	508	479

Appendix E: Resuspended particle number concentrations in mobile and stationary samples (#/m^3)

Table E1. Resuspended particle number concentrations in mobile and stationary samples (#/m^3)

Carpeted floor experiments								
Sample #	Bin size (μm)	1-2.5	2.5-5	5-10		1-2.5	2.5-5	5-10
1	Mobile	6240	5164	3738	Stationary	3421	2530	1402
2	Mobile	0	699	758	Stationary	0	166	271
3	Mobile	1226	2006	1333	Stationary	1026	945	477
4	Mobile	3780	3260	2729	Stationary	4016	3308	2081
5	Mobile	875	1500	1171	Stationary	0	708	291
6	Mobile	7400	2968	1782	Stationary	2584	2103	828
7	Mobile	1745	2727	2041	Stationary	3229	2493	1280
8	Mobile	868	0	0	Stationary	2727	0	0
9	Mobile	2824	2006	1153	Stationary	2747	1813	828
10	Mobile	0	1074	993	Stationary	0	1018	814
11	Mobile	0	4650	3559	Stationary	0	0	402
12	Mobile	1560	987	655	Stationary	2168	1223	491
13	Mobile	12337	14271	12375	Stationary	13824	12370	7361

14	Mobile	1526	1687	1869	Stationary	642	1486	1606
15	Mobile	4523	4304	3422	Stationary	2225	2582	1935
	Mean	2994	3153	2505		2574	2183	1338
	SD	3433	3422	2953		3410	2996	1781
Bin size (µm)		1-2.5	2.5-5	5-10				
Paired t test p-value (M vs. S)		0.3730	0.0123	0.0052				
Vinyl floor experiments								
Sample #	Bin size (µm)	1-2.5	2.5-5	5-10		1-2.5	2.5-5	5-10
1	Mobile	4285	2581	2521	Stationary	1853	1347	1218
2	Mobile	1220	801	657	Stationary	2084	1198	790
3	Mobile	18	211	533	Stationary	0	0	156
	Mean	1841	1198	1237		1313	848	722
	SD	2200	1234	1114		1143	738	534
Bin size (µm)		1-2.5	2.5-5	5-10				
Paired t test p-value (M vs. S)		0.6452	0.5393	0.3447				

Appendix F: Percentages (%) of particle number concentrations in different particle size bins for mobile, stationary and settled dust samples (Correction factor and background subtraction applied for mobile and stationary samples)

Table F1. Percentages (%) of particle number concentrations in different particle size bins for mobile, stationary and settled dust samples

	Settled dust			Carpet _Mobile			Carpet_Stationary		
Bin size (µm)	1-2.5	2.5-5	5-10	1-2.5	2.5-5	5-10	1-2.5	2.5-5	5-10
1	23.3	34.9	41.8	41.2	34.1	24.7	46.5	34.4	19.1
2	8.9	31.6	59.5	0.0	48.0	52.0	0.0	38.0	62.0
3	0.0	21.3	78.7	26.9	43.9	29.2	41.9	38.6	19.5
4	5.3	33.3	61.3	38.7	33.4	27.9	42.7	35.2	22.1
5	4.1	49.0	46.9	24.7	42.3	33.0	0.0	70.9	29.1
6	0.0	43.1	56.9	60.9	24.4	14.7	46.9	38.1	15.0
7	4.5	35.8	59.7	26.8	41.9	31.3	46.1	35.6	18.3
8	15.0	43.5	41.5	100.0	0.0	0.0	100.0	0.0	0.0
9	2.0	32.7	48.3	47.2	33.5	19.3	51.0	33.7	15.4
10	11.3	24.5	64.2	0.0	51.9	48.1	0.0	55.6	44.4
11	16.7	38.1	45.2	0.0	56.6	43.4	0.0	0.0	100.0
12	5.0	35.3	59.7	48.7	30.8	20.5	55.9	31.5	12.6
13	9.3	47.5	43.2	31.6	36.6	31.7	41.2	36.9	21.9
14	20.7	42.3	36.9	30.0	33.2	36.8	17.2	39.8	43.0
15	21.0	41.2	37.8	36.9	35.1	27.9	33.0	38.3	28.7
16	12.4	39.1	48.5	Vinyl_Mobile			Vinyl_Stationary		

17	13.4	41.6	45.0	1-2.5	2.5-5	5-10	1-2.5	2.5-5	5-10
18	10.4	51.9	37.7	45.6	27.5	26.9	41.9	30.5	27.6
19	16.0	37.8	46.2	45.6	29.9	24.5	51.2	29.4	19.4
20	20.5	45.6	33.9	2.4	27.6	70.0	0.0	0.0	100.0
21	9.8	50.9	39.3						
22	23.5	41.5	35.0						
23	10.4	32.4	57.1						
24	13.3	42.2	44.5						
25	8.9	37.6	53.5						
26	7.8	46.4	45.8						
27	13.9	39.9	46.2						
28	13.3	36.3	50.4						
29	8.3	34.0	57.6						
30	8.5	39.5	51.9						

Appendix G: Dust loading simulation tests

To evaluate dust loading simulation, we performed four sets of dust loading simulation tests on vinyl floor. Dust loading area was 3.3 m* 3.6 m=11.9 m^2 and the expected dust loading was 1 g/m^2. Prior to spraying the dust in the room, 16 pre-weighed weighing papers were placed on the floor (Figure G.1).

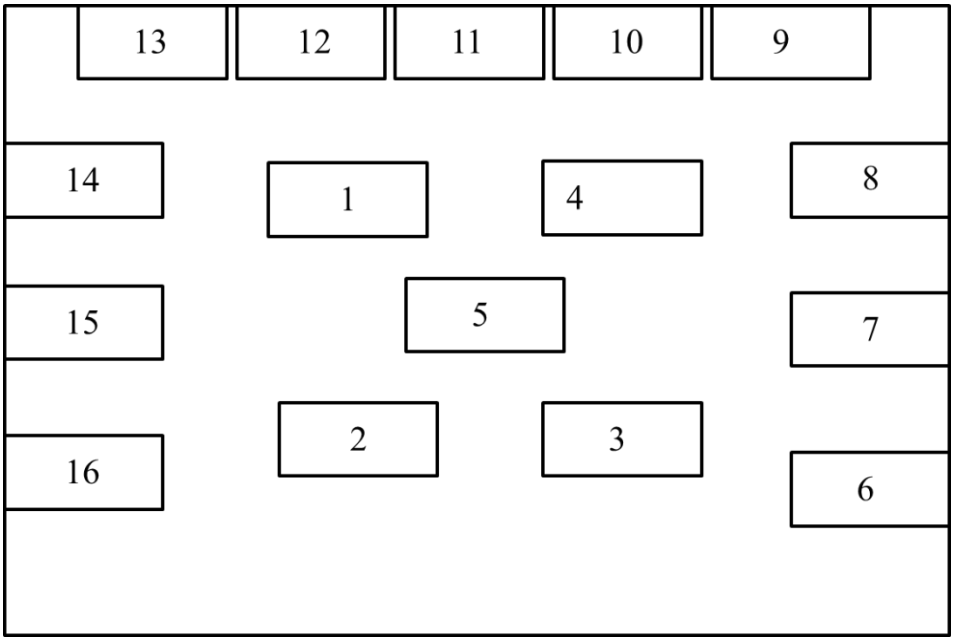

Figure G.1. Location of weighing papers

Area of weighing paper=7.5 cm*7.5 cm=0.005625 m^2 ($\pm 2.25*10^{-6}$ m^2)

Wait overnight for dust deposition and on the next day, all the weighing papers were collected and weighed.

Calculated dust loadings are presented in Figure G.2 as mean ± standard deviation. Four dust loading experiments on the vinyl floor were conducted, and the average vinyl floor dust loading was 1.2±0.5 g/m^2

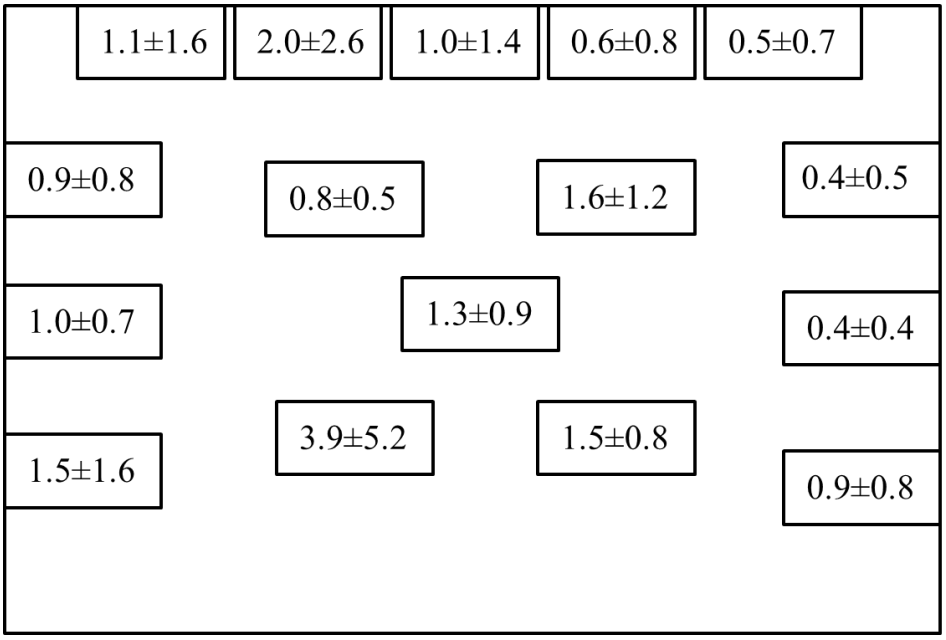

Figure G.2. Distribution of dust loadings (g/m^2) (mean ± standard deviation)

Appendix H: Method development for dust sample extraction

Experiments identifying a simple, efficient method for dust sample extraction compatible with lab instrumentation were performed. Three methods most extensively evaluated were

i). Dust (sieved)--liquid-liquid extraction --GC/MS

ii). Dust (sieved) -- liquid-liquid extraction ---Glass wool filtered--GC/MS

iii). Dust (sieved) -- liquid-liquid extraction --SPE--GC/MS

Recoveries of pyrethroids via the three methods were shown in Table H.1-H.3:

Table H.1. Recovery of pyrethroids in dust samples under method i

Compound	Pyrethroids spiked dust	Matrix blank	Solvent blank
Dichloran	1.20	1.14	0.00
Tefluthrin	0.68	0.00	0.00
Tetrachlorvinphos	1.05	0.06	0.06
L-Cyhalothrin	1.12	0.00	0.04
Permethrin	1.49	0.50	0.05
Cyfluthrin	1.81	0.00	0.00
Fenvalerate	1.59	0.00	0.00
Deltamethrin	0.97	0.00	0.00

Table H.2. Recovery of pyrethroids in dust samples under method ii

Compound	Pyrethroids spiked dust	Matrix blank	Solvent blank
Dichloran	0.64	0.42	0.00
Tefluthrin	0.39	0.00	0.00
Tetrachlorvinphos	0.97	0.33	0.06
L-Cyhalothrin	0.58	0.00	0.00
Permethrin	0.74	0.25	0.08
Cyfluthrin	0.76	0.00	0.00
Fenvalerate	0.67	0.00	0.00
Deltamethrin	0.52	0.00	0.00

Table H.3. Recovery of pyrethroids in dust samples under method iii

Compound	Pyrethroids spiked dust	Matrix blank	Solvent blank
Dichloran	0.78	0.00	0.00
Tefluthrin	0.85	0.00	0.00
Tetrachlorvinphos	2.56	0.12	0.06
L-Cyhalothrin	1.07	0.00	0.00
Permethrin	1.44	0.38	0.04
Cyfluthrin	1.55	0.00	0.00
Fenvalerate	1.24	0.00	0.00

Deltamethrin	0.90	0.00	0.00

Based on the recovery of each method and simplicity of sample preparation, the following sample preparation method was selected for use:

(1) Extract using 6 mL of a mixture of 1:1 hexane: acetone.

(2) Sonicate for 10 minutes and then centrifuge at 4,000 rpm for 5 minutes.

(3) Transfer the supernatant to a clean test tube and reduce the volume to approximately 0.3-0.5 mL.

(4) Transfer the extracts to a glass insert and evaporate to dryness.

(5) Reconstitute the sample in 100 μL of hexane.

(6) Analyze by GC-ECD.

Appendix I: Comparison of particle mass concentration measured by ReCon Rover and PIPER

Comparison experiments were performed on the vinyl floor using ReCon Rover and the Pre-toddler Inhalable Particulate Environmental Robotic sampler (PIPER). Dust loading was 1 g/m^2.

For PIPER experiments, airborne samples were collected on a 25 mm Teflon filter with 2.0 μm pore size from Pall Life Sciences (Port Washington, NY, USA) installed in a Button Aerosol Sampler. The Button Aerosol Sampler (SKC Inc., Eighty Four, PA, USA) was connected to a Leland Legacy pump (SKC Inc., Eighty Four, PA, USA) providing a flow rate of 10 L/min. Sampling duration was 4 hours. In total, three samples were collected.

For ReCon Rover experiments, airborne samples were collected on two types of samplers. One was collected by Button Aerosol Sampler following the same method used in PIPER experiments and the other was collected on a 37 mm Teflon filter with 2.0 μm pore size from Pall Life Sciences (Port Washington, NY, USA) installed in a Personal Environmental Monitor (PEM) (10-μm cut-point at 10 L/min, SKC Inc., Eighty Four, PA, USA). Each sampler was connected to an SKC Leland Legacy pump providing a flow rate of 10 L/min. They were attached to the ReCon Rover and placed at a height of 80 cm. Sampling duration was 4 hours and six samples were collected (three ReCon-PEM samples and three ReCon-Button Aerosol Sampler samples).

The mean and standard deviation of measured particle mass concentrations are presented in Table I1. The ReCon Rover generated a higher particle mass concentration than PIPER. This confirms that the ReCon Rover resuspends dust when it moves across a dusty floor. The difference in measured particle mass concentrations between the two robots might result from the design of the two robots, the way of contacting the floor, sampling height, etc. ReCon Rover has its limitations in simulating a toddler's activities. However, its movement generates a dust cloud that can be collected using air samples mounted at a toddler's breathing zone height.

Table I1. Comparison of particle mass concentrations measured using ReCon and PIPER

Robot_Sampler (N=3)	Mean ± standard deviation ($\mu g/m^3$)
ReCon_PEM	32.5±6.9
ReCon_Button Aerosol Sampler	54.1±15.7
PIPER_Button Aerosol Sampler	15.3±4.4

Appendix J: Measurement of air exchange rates

Air exchange rates were measured in each set of experiments following a tracer gas decay method. A CO_2 tank was put in one corner of the room. External CO_2 concentrations were measured before and after the indoor measurements. The initial background CO_2 concentration in the room was recorded. A Telaire 7001 CO_2 monitor and a Hobo temp/RH logger—UX100-003 were used. CO_2 was released in the room and the researcher would quickly walk outside the room after initiating the release. The equation for calculating air exchange per unit time is given as following:

Equation J.1. $N = \frac{\left[In\left(C_{int}^{t_0} - C_{ext}\right) - In\left(C_{int}^{t_1} - C_{ext}\right)\right]}{t^1 - t^0}$

Where

N=number of air changes

$C_{int}^{t_0}$=internal concentration of CO_2 in enclosure at start

C_{ext}=external concentration of CO_2 in room

$C_{int}^{t_1}$=internal concentration of CO_2 in enclosure at end

t^0=time at start (days)

t^1=time at end (days)

In=natural logarithm.

Table J1.Air exchange rates (/hour) for vinyl and carpeted floor experiments.

Test #	Carpeted floor_Air exchange rate (/hour)	Vinyl floor_Air exchange rate (/hour)
1	1.00	2.3
2	1.09	1.8
3	1.73	2.4
4	0.85	
5	0.79	

Appendix K: Permethrin migration experiments

To evaluate permethrin concentration changes over time after permethrin application indoors, one set of experiments was performed on the vinyl flooring with wipe samples collected one day, three days and five days post-permethirn spray. Dust loading was $1g/m^2$. Experiment protocol was as follows:

- Background sample collection before spraying the dust: two wipe samples, one from the corner and one from the middle of the room: BLK11, BLK12
- Background sample collection after spraying the dust: two wipe samples, one from the corner and one from the middle of the room: BLK21,BLK22
- Perimeter spray of permethrin in the room
- Day1—24hr post spray---wipe sample collection: A11, A21, A31, A41, B11,B21,B31,B41, C11,C12
- Day3---72hr post spray---wipe sample collection: A12,A22,A32,A42,B12,B22,B32,B42,C21,C22
- Day5---120hr post spray---wipe sample collection: A13,A23,A33,A43,B13,B23,B33,B43,C31,C32

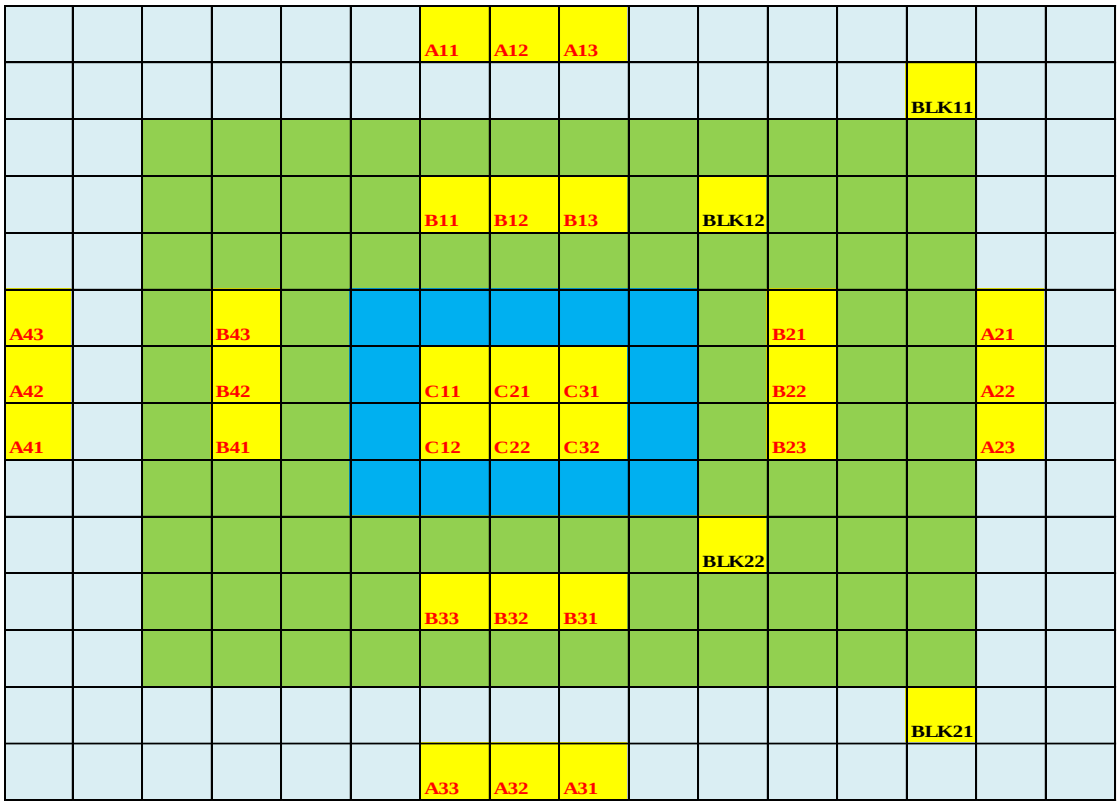

Figure K1. Sampling locations

Permethrin loadings on the floor and permethrin concentrations in dust are presented in Table K1. We calculated the ratio of permethrin concentration in dust measured on day 3 to the permethrin concentration measured on day 1 and the ratio of day 5 to day 1 for each sampling zone area. The average ratio of day 3 to day 1 and day 5 to day 1 is 0.80 and 1.09, respectively. This indicates that permethrin concentrations within 5 days post-application were comparable in the simulated room environment.

Table K1. Permethrin loadings on the floor and permethrin concentrations in dust

Sample ID	Permethrin loading on the floor ($\mu g/m^2$)	Permethrin concentration in dust ($\mu g/g$)
control1	0.00	0.00
control2	0.00	0.00
BLK11	2.70	129.83
BLK12	3.81	403.29
BLK21	2.17	18.55
BLK22	1.32	4.73
A11	1274.69	3920.40
A21	562.46	438.20
A31	202.64	864.50
A41	6309.10	11051.37
B11	241.16	631.57
B21	53.14	76.18
B31	18.18	54.33
B41	241.49	206.38
C11	29.35	69.31
C12	50.26	135.66
A12	1653.52	5085.52
A22	180.82	171.43
A32	96.34	553.97
A42	3247.51	3100.96
B12	39.41	106.36
B22	46.94	91.62
B32	43.53	151.50
B42	34.36	43.48
C21	27.63	63.56

C22	4.76	15.56
A13	1973.43	4442.32
A23	286.58	170.92
A33	551.11	3470.67
A43	5450.27	2431.02
B13	119.90	205.27
B23	78.38	139.60
B33	20.67	31.51
B43	69.56	418.15
C31	11.77	28.69
C32	0.00	0.00

Appendix L: Temperature and relative humidity

Tempetature and relative humidity were continously monitored with a Hobo Temp/RH data logger—UX100-003 (Onset Company, Bourne, MA, USA). The average temperature and relative humidity for each set of experiments are shown in Table L1.

Table L1. Temperature and relative humidity measurements in study.

	Temperature (F)	Relative Humidity (%)
Vinyl Floor Test #	Mean± SD	Mean± SD
1	76.1±2.9	46.2±6.9
2	78.1±2.3	62.4±8.0
3	75.1±1.8	58.6±5.9
Carpeted Floor Test #	Mean± SD	Mean± SD
1	71.4±3.4	34.7±6.5
2	68.9±2.6	17.2±3.9
3	72.1±2.5	19.9±2.7
4	69.8±2.4	24.7±8.5
5	73.8±3.2	32.3±5.9

Appendix M: Calculation of sampling efficiency

Sampling efficiency for the particle size distribution sampling line was calculated using Aerocalc (TSI, Inc. Shoreview, MN, USA. http://www.tsi.com/SiteSearch.aspx?q=Aerocalc&page=1&count=15&folderId=588&orderBy=prodOrder). The selected equation for each part is presented in Table M1.

Table M1. Selected equations for sampling efficiency calculation in Aerocalc.

Parameters	Reference and notes to selected equations
Sampling Efficency	BluntSampler
Inlet efficiency	
Aspiration efficiency	Aspiration efficiency of a blunt sampler worn on the body. (B&W 8-31 to 8-34; Tsai, Vincent and Mark. Ann. Occup. Hyg. 40(1) 93-113, 1996
Gravitational transmission efficiency	Gravitational losses in an inlet (B&W 8-23, 8-24; W&B 6-23, 6-24)
Inertial transmission efficiency	Inertial losses in a sharp-edged inlet (B&W 8-25 to 8-29; W&B 6-25 to 6-29)
Transport efficiency	
Gravitational settling	Gravitational settling is 1 since vertical tubing was used
Diffusional deposition	Diffusion losses in a tube--fraction passing through tube (B&W 56 to 8-60, 19-19 to 19-23; W&B 6-42, 6-43, 6-44, 19-19 to 19-23)
Turbulent inertial deposition	Not considered since by calculation, the air flow in the tubing was laminar flow with Reynolds number

	less than 2000
Inertial deposition at a bend	Not considered----no bend in the sampling system
Inertial deposition at flow constrictions	Inertial deposition at flow constrictions is 1 since the constriction is always from tubing with a smaller diameter to a tubing with a larger diameter, no loss was expected
Electrostatic deposition	Electrostatic deposition is 1----Tygon and metal tubing were used in the sampling line
Thermophoretic deposition	Thermophoretic deposition is 1. No temperature gradient is expected to be present in a short sampling line
Diffusiophoretic deposition	Diffusiopheretic deposition is 1 or loss is 0----no concentration gradient is expected to be present in a short sampling line
Sampling efficiency=Inlet efficiency*Transport efficiency	

For mobile and stationary sampling, different free-stream velocities were used in the calculations. For mobile sampling, we considered the robotic's movement with a 0.1 m/s, while for stationary sampling, it was fixed in its location with zero speed applied in the calculation.

Table M2. Sampling efficiency calculation for mobile sampling

Particle diameter (μm)	10.0	5.0	2.5	1.0	0.5	0.3
Inlet efficiency						

Aspiration efficiency	0.6	0.9	1.0	1.0	1.0	1.0
Gravitational transmission efficiency	1.0	1.0	1.0	1.0	1.0	1.0
Inertial transmission efficiency	1.0	1.0	1.0	1.0	1.0	1.0
Transport efficiency						
Gravitational settling	1.0	1.0	1.0	1.0	1.0	1.0
Diffusional deposition	1.0	1.0	1.0	1.0	1.0	1.0
Turbulent inertial deposition	N/D	N/D	N/D	N/D	N/D	N/D
Inertial deposition at a bend	N/D	N/D	N/D	N/D	N/D	N/D
Inertial deposition at flow constrictions	1.0	1.0	1.0	1.0	1.0	1.0
Electrostatic deposition	1.0	1.0	1.0	1.0	1.0	1.0
Thermophoretic deposition	1.0	1.0	1.0	1.0	1.0	1.0
Diffusiophoretic deposition	1.0	1.0	1.0	1.0	1.0	1.0
Sampling efficiency	0.6	0.9	1.0	1.0	1.0	1.0
Correction Factor	1.5	1.1	1.0	1.0	1.0	1.0
Average correction factor for 5-10 µm	1.3					

Table M3. Sampling efficiency calculation for stationary sampling

Particle diameter (µm)	10.0	5.0	2.5	1.0	0.5	0.3
Inlet efficiency						
Aspiration efficiency	0.8	1.0	1.0	1.0	1.0	1.0
Gravitational transmission efficiency	1.0	1.0	1.0	1.0	1.0	1.0
Inertial transmission efficiency	1.0	1.0	1.0	1.0	1.0	1.0

Transport efficiency						
Gravitational settling	1.0	1.0	1.0	1.0	1.0	1.0
Diffusional deposition	1.0	1.0	1.0	1.0	1.0	1.0
Turbulent inertial deposition	N/D	N/D	N/D	N/D	N/D	N/D
Inertial deposition at a bend	N/D	N/D	N/D	N/D	N/D	N/D
Inertial deposition at flow constrictions	1.0	1.0	1.0	1.0	1.0	1.0
Electrostatic deposition	1.0	1.0	1.0	1.0	1.0	1.0
Thermophoretic deposition	1.0	1.0	1.0	1.0	1.0	1.0
Diffusiophoretic deposition	1.0	1.0	1.0	1.0	1.0	1.0
Sampling efficiency	0.8	1.0	1.0	1.0	1.0	1.0
Correction Factor	1.2	1.0	1.0	1.0	1.0	1.0
Average correction factor for 5-10 µm	1.1					