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Studies on Particle Resuspension, Infant Exposure,  
and the Sleep Microenvironment

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Studies on Particle Resuspension, Infant Exposure,  
and the Sleep Microenvironment

by

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Dissertation

Presented to the Faculty of the Graduate School of

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## Dedication

To my wife, Nusrat Jung, for her love.

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# Studies on Particle Resuspension, Infant Exposure, and the Sleep Microenvironment

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Understanding the transport of particulate and gaseous indoor air pollutants from source to exposure is paramount to improve our understanding of the complexities of the built environments in which we spend the majority of our time. This dissertation offers new insights on particle resuspension from indoor surfaces, infant exposure to organic contaminants released from crib mattresses, and the dynamics of pollutant transport and human exposure while sleeping.

Particle resuspension is the physical process by which settled particles detach from a surface and become airborne through application of various aerodynamic and mechanical removal forces. Resuspension is an important indoor source of coarse mode particles ( $> 1 \mu\text{m}$  in diameter) and can be a source mechanism for biological matter and organic contaminants that accumulate in house dust. Settled dust deposits on indoor surfaces can vary considerably in their structure and mass loading, yet little is known as to how these parameters affect resuspension. Through wind tunnel experiments, this research demonstrates that the deposit structure (monolayer or multilayer) can have a

significant impact on the number of particles that aerodynamically resuspend. Furthermore, this dissertation presents the first full-scale experimental chamber study to show that human body movements in bed can resuspend settled mattress dust particles. An indoor aerosol model was utilized to provide a mechanistic understanding of the impact of movement intensity, surface vibrations, bedroom ventilation rate, and dust loading on the resuspension flux and intake fraction of resuspended particles.

Infants spend most of their time sleeping and are likely to be exposed to elevated concentrations of chemicals released from their crib mattresses. Through a combination of chamber experiments and solvent extractions, this research shows that infant crib mattresses can emit a variety of volatile organic compounds (VOCs) and contain numerous chemical additives, including phthalate and alternative plasticizers, flame retardants, and unreacted isocyanates. Additionally, this study discovered that infants are exposed to approximately twice the concentrations of VOCs in their breathing zones as compared to the bulk bedroom air, due to their close proximity to the source.

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## List of Publications

This dissertation consists of an executive summary, followed by six peer-reviewed journals publications: two literature reviews and four original research papers. Throughout the executive summary, the papers are referenced to their corresponding appendix notation, A-F.

A. Boor, B.E., Siegel, J.A., and Novoselac, A. (2013). Monolayer and Multilayer Particle Deposits on Hard Surfaces: Literature Review and Implications for Particle Resuspension in the Indoor Environment. *Aerosol Science & Technology*, 47(8):831-847.

B. Boor, B.E., Laverge, J., Spilak, M.P., Novoselac, A., and Xu, Y. Human Exposure to Indoor Air Pollutants in Sleep Microenvironments: A Literature Review. In Preparation for Submission to *Atmospheric Environment*.

C. Boor, B.E., Siegel, J.A., and Novoselac, A. (2013). Wind Tunnel Study on Aerodynamic Particle Resuspension from Monolayer and Multilayer Deposits on Linoleum Flooring and Galvanized Sheet Metal. *Aerosol Science & Technology*, 47(8):848-857.

D. Boor, B.E., Spilak, M.P., Corsi, R.L., and Novoselac, A. (2015). Characterizing Particle Resuspension from Mattresses: Chamber Study. *Indoor Air*, 25(4):441-456.

E. Boor, B.E., Järnström, H., Novoselac, A., and Xu, Y. (2014). Infant Exposure to Emissions of Volatile Organic Compounds from Crib Mattresses. *Environmental Science & Technology*, 48(6):3541-3549.

F. Boor, B.E., Liang, Y., Crain, N.E., Järnström, H., Novoselac, A., and Xu, Y. (2015). Identification of Phthalate and Alternative Plasticizers, Flame Retardants, and Unreacted Isocyanates in Infant Crib Mattress Covers and Foam. *Environmental Science & Technology Letters*, 2(4):89-94.

## Chapter 1: Introduction

### *Overview of the Executive Summary*

The executive summary is composed of six chapters, with the aim of providing a condensed overview of the research included within the six peer-reviewed journals publications of this dissertation. Chapter 1 introduces the objectives of the dissertation research; Chapter 2 provides a brief literature review of particle resuspension in indoor environments, dust deposits on indoor surfaces, and the dynamics of human exposure and pollutant transport in the sleep microenvironment; Chapter 3 describes the experimental methodologies employed in this research; Chapter 4 highlights new insights on indoor particle resuspension; Chapter 5 presents key findings on chemical emissions from crib mattresses and infant exposures to volatile organic compounds while sleeping; and Chapter 6 discusses the contributions of this research to the fields of indoor air, aerosol, and exposure sciences, as well as the practical implications of this work.

### *Research Objectives*

This dissertation aims to fulfill the following three research objectives, each of which represents a core theme of the dissertation (illustration 1.1):

*Objective 1:* To provide a better understanding of how particle deposits on indoor surfaces vary in structure and mass loading; the impact of particle deposit structure (monolayer vs. multilayer) and dust load on aerodynamic-induced particle resuspension from ventilation ducts and flooring materials; how body

movements in bed resuspend mattress dust particles; and the underlying physical mechanisms of particle detachment and transport in the sleep microenvironment.

*Objective 2:* To evaluate new and used crib mattresses as a source of volatile organic compounds (VOCs), phthalate and alternative plasticizers, flame retardants, and unreacted isocyanates and establish the source-proximity effect of the infant sleep microenvironment and its role in elevating infant exposures to chemical contaminants which volatilize from mattresses and bedding materials.

*Objective 3:* To establish the important role of human exposures to indoor air pollutants while sleeping and to analyze the transport dynamics of mattress-released particulate and gaseous pollutants around the human body.

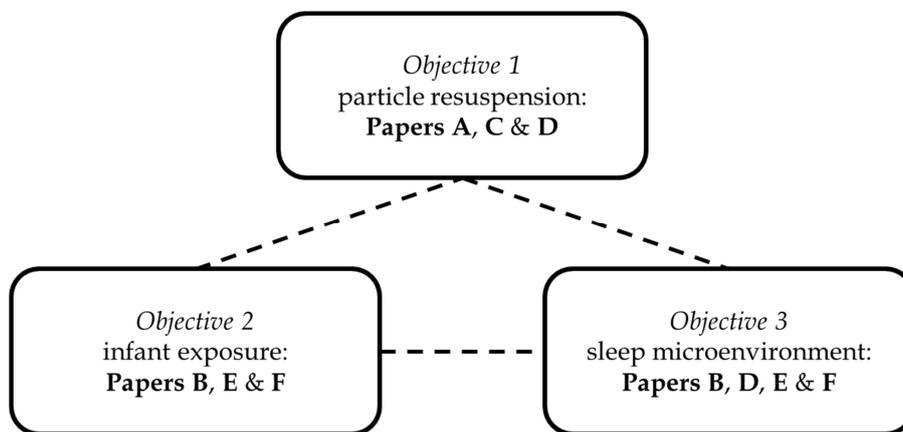


Illustration 1.1: Organization of the Papers A-F within the three themes of this dissertation: particle resuspension, infant exposure, and the sleep microenvironment.

## Chapter 2: Background

This chapter provides a summary of selected sections of the two literature reviews included in this dissertation, Papers A and B.

### PARTICLE DEPOSITS ON INDOOR SURFACES

Particle deposits in the indoor environment can be very complex and indoor dust loads can vary across several orders of magnitude. Table A.1 in Paper A provides a summary of dust loads on hard indoor surfaces (ventilation ducts and hard flooring) from selected field studies ( $n = 29$ ), along with wind tunnel ( $n = 29$ ) and full-scale ( $n = 11$ ) resuspension studies.

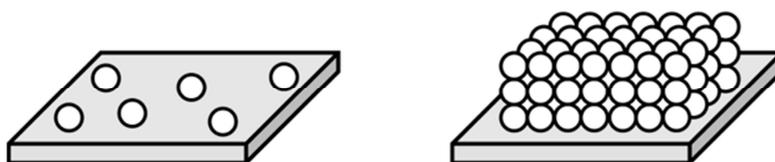


Illustration 2.1: Monolayer (left) and multilayer (right) particle deposits.

On the surfaces of ventilation ducts, dust loads can range from less than  $1 \text{ g/m}^2$  to loads in excess of  $100 \text{ g/m}^2$  (Nielson et al. 1990, Laatikainen et al. 1991, Pasanen et al. 1992, EPA 1996, Fortmann et al. 1997, Möriz et al. 2001, Kolari et al. 2005, and Lavoie et al. 2011). On hard flooring, such as vinyl, linoleum, and hardwood, dust loading is typically in the range of  $0.1$  to  $1 \text{ g/m}^2$ , although lighter and heavier dust loads are commonly reported (e.g. Adgate et al. 1995, Rao et al. 2005, Johnson et al. 2009, Hoh et al. 2012). The wide range of dust loads suggests that there exist different types of particle deposits on indoor surfaces, including both monolayer and multilayer deposits (Illustration 2.1). A

monolayer deposit is one in which particles are sparsely deposited on a surface and there is minimal to no contact between them. A multilayer deposit is a porous structure of particles deposited on top of one another, forming multiple layers.

The diversity of dust loads and particle deposits may have important implications for indoor particle resuspension. There are fundamental differences in the resuspension process between monolayer and multilayer deposits, as outlined in Papers A and C, and summarized in Chapter 4. Learning how dust deposits on indoor surfaces vary in structure provides a useful basis to understand how this parameter influences particle resuspension.

#### *Deposit Classification: From Sparse Monolayers to Thick Multilayers*

Paper A presents a simplified, approximate method to classify a given dust load as either a monolayer, multilayer, or intermediate particle deposit. The scaling analysis (Equation 2.1) estimates the height of the particle deposit ( $\delta$ ), which is a function of the dust load ( $m_0$ , g/m<sup>2</sup>), particle density ( $\rho$ , kg/m<sup>3</sup>) and deposit porosity ( $\varepsilon$ ). The porosity is linked to the deposition mechanism, e.g. dense deposits formed by inertial impaction can be considered “cake-like” deposits, whereas those formed by gravitational settling result in “fluffy” deposits (as illustrated in Figure 1 of Friess and Yadigaroglu 2002).

$$\delta \sim \frac{6m_0}{\pi\rho(1-\varepsilon)} \quad (2.1)$$

The deposit height can be compared with reference mass median diameters ( $D$ ) to approximate the type of particle deposit. The classification criteria is as follows: (i.) if  $\delta \leq D$ : the deposit is a monolayer; (ii.) if  $D < \delta < 2D$ : the deposit is an intermediate between a monolayer and multilayer; (iii.) if  $\delta \geq 2D$ : the deposit is a multilayer. Paper A provides an overview of the assumptions made in this analysis and the associated limitations. The dust loads reported in field studies may be biased by the dust collection (e.g. vacuuming, sieving) and analysis technique (e.g. gravimetric) used (Qian et al. 2014).

#### *Summary of Dust Loadings*

Table A.1 of Paper A presents the results of the particle deposit classification analysis and Table 2.1 provides a summary of how often each deposit type was identified. For ventilation ducts, dust loading is highly variable, and dust loads greater than 10 g/m<sup>2</sup> are common. Studies by Nielsen et al. (1990), Laatikainen et al. (1991), Pasanen et al. (1992), Pasanen et al. (1995), EPA (1996), Ito et al. (1996), Fortmann et al. (1997), Küchen (1998), Kolari et al. (2005), Lavoie et al. (2011), and Zuraimi et al. (2012) have reported heavy dust loads representative of multilayer deposits, whereas studies by Auger (1994), Kalliokoski et al. (1995), Fransson et al. (1995), and Holopainen et al. (2002) have reported lighter dust loads suggestive of monolayer deposits. Dust loading in ventilation ducts can be influenced by many factors, such as characteristics of the building and its ventilation system, indoor particle sources, in-duct filtration, deposition mechanisms, and frequency of duct cleaning. Therefore, based on the field studies as presented in Table A.1, it would be expected that dust loading would vary considerably from any one building to the next, suggesting that deposits ranging from sparse monolayers to heavy multilayers are likely to exist.

Table 2.1: Number of studies for each category with dust loads classified as:

	monolayer	intermediate	multilayer
field study: ventilation duct	16	15	12
field study: hard flooring	12	9	7
resuspension study: wind tunnel	20	2	8
resuspension study: full-scale	4	4	9

On hard flooring, such as linoleum, wood, and vinyl, lighter dust loads representative of monolayer deposits are frequently observed. Dust loads reported by Thatcher and Layton (1995), Franke et al. (1997), Salares et al. (2009), and Raja et al. (2010) were classified as monolayer deposits. Field studies by Adgate et al. (1995), Rich et al. (1999), Rao et al. (2005), Lewis et al. (2006), Johnson et al. (2009), Layton and Beamer (2009), and Hoh et al. (2011) reported ranges of dust loads that likely include both monolayer and multilayer deposits. The range of dust loads can be attributed to the frequency of floor cleaning or the number of particle sources indoors, among other factors.

Although we see the existence of both monolayer and multilayer deposits indoors, the bulk of experimental wind tunnel studies have primarily focused on the former in order to develop a more fundamental understanding of the resuspension process and particle-to-deposition surface interactions and adhesion. Only a few experimental wind tunnel studies have explored resuspension from multilayer deposits, including those by Fromentin (1989), Matsusaka and Masuda (1996), Loosmore and Hunt (2000), Adhiwidjaja et al.

(2000), Chiou and Tsai (2001), Huang et al. (2005), Gomes et al. (2007), Nitschke and Schmidt (2010). The findings of the monolayer and multilayer wind tunnel studies are described in detail in Paper A to highlight some of the unique attributes of resuspension from both types of deposits. For full-scale resuspension studies, e.g. studies not conducted in small-scale wind tunnels, dust loads were found to be more representative of those found in the indoor environment.

#### THE SLEEP MICROENVIRONMENT

Papers B, D, E, and F focus on human exposures to mattress-released particulate and gaseous pollutants and the dynamics of pollutant transport in the sleep microenvironment. Adults sleep for approximately 8 hours per day, which corresponds to one third of their lifetime, making sleep microenvironments particularly important in contributing to both their acute and chronic exposures to indoor air pollutants. The sleep microenvironment can be defined as the space encompassing a mattress, pillow, bedding materials, bed frame and the volume of air above these items that includes an individual's breathing zone (BZ) and buoyant thermal plume.

The sleep microenvironment can be home to a diversity of chemical contaminants and biological material that have been shown to impact human health. Mite and animal allergens, bacteria, fungi, and semi-volatile organic compounds (SVOCs) can accumulate in settled dust on mattresses, pillows, and bed sheets. Table B1 in Paper B provides a comprehensive overview of the

chemical and biological composition of mattress dust. Paper D will demonstrate that human body movements in bed can resuspend settled dust particles, thereby serving as a source mechanism for the various pollutants found in mattress dust. The materials used to manufacture mattresses and bedding products, such as polyurethane foam and vinyl mattress covers, are possible sources of a myriad of chemical contaminants, including volatile organic compounds (VOCs), plasticizers, flame retardants, and unreacted (free) isocyanates (NCO). Papers E and F will provide new experimental data on the emissions of selected organic contaminants, and their material-phase concentrations, from infant crib mattress covers and foam.

#### *Exposure Considerations for Infants and Adults*

Humans spend a considerable amount of time sleeping. The U.S. Environmental Protection Agency (EPA) provides data on the duration of time spent (hours/day) in a sleep or nap activity in the Exposure Factors Handbook (EFH) (EPA 2009). To investigate the dependence of sleep duration on age, EFH's data is plotted in Figure 2.1. During the first few months to years of life, infants (<1 year of age) and toddlers (1-3 years of age) spend a considerable amount of time sleeping, with an average of 13.3 hours/day in the first year of life, 12.6 hours/day in the second year, and 12.1 hours/day in the third year. In addition, other researchers (Diez et al. 2000, Iglowstein et al. 2003) have also reported durations of sleep greater than 14 hours/day during infancy. The data suggests that sleep microenvironments play a critical role in characterizing exposures of very young children to indoor air pollutants.

Using the data from the EFH and the National Human Activity Pattern Survey (NHAPS) Study conducted by Klepeis et al. (2001) it was found that adults spend about 34% of their day in the sleep microenvironment, which equates to about 50% of the time they spend in a residence, and 39% of the time they spend indoors. Thus, the sleep microenvironment becomes the predominant indoor space where American adults spend most of their time.

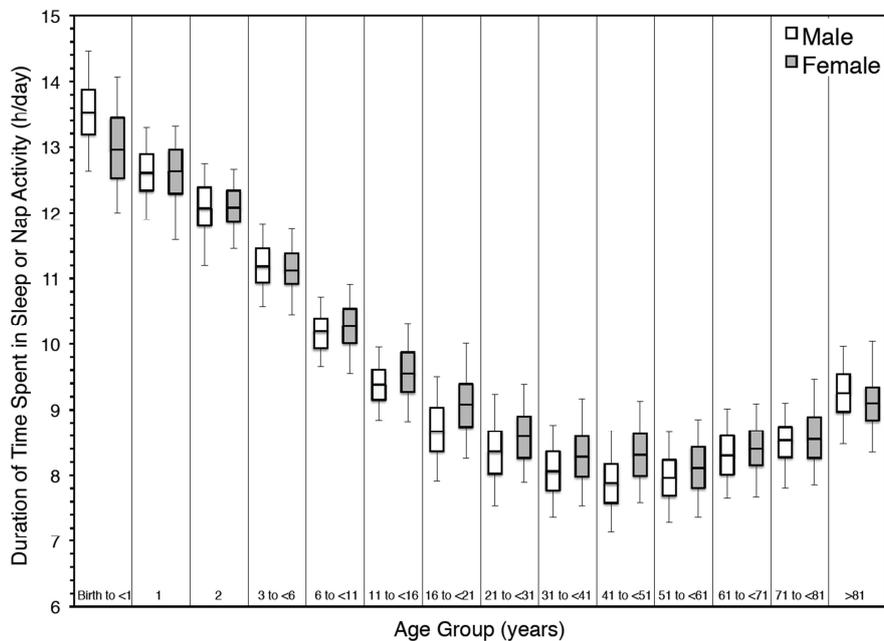


Figure 2.1: Duration of time (hours/day) spent in sleep or nap activity (EPA ID = 14500), categorized by age group and gender (Adapted from U.S. EPA EFH data, Sleep or Nap Activity, EPA Activity ID = 14500, U.S. EPA 2009). Box plots represent interquartile range and whiskers represent the 5<sup>th</sup> and 95<sup>th</sup> percentiles.

Exposure to mattress-released pollutants can occur via inhalation and dermal pathways, as discussed in Paper B. Infants can receive much greater doses of a chemical additive released from their mattress compared to adults since they inhale approximately an order of magnitude more air per body mass than adults and have skin surface area to body mass ratios three times greater than adults. Special considerations for assessing infant exposures while sleeping are presented in Paper B.

#### *Pollutant Transport Dynamics*

Along with the extended exposure period in the sleep microenvironment, the source-proximity effect, in which pollutant concentrations near a source are greater than those in the bulk air of a room, may lead to elevated inhalation and dermal exposures to pollutants originating in mattresses, bedding materials, and settled mattress dust. There are several key factors that likely influence the source-proximity effect (illustration 2.2), including the spatial proximity of the BZ to the source (influenced by sleep position, e.g. prone or supine), incomplete mixing of bedroom air, concentrations gradients near an actively emitting source, the personal cloud due to human movement-induced particle resuspension, the buoyant human thermal plume, heat transfer from the human body to the source, which may elevate the emissions of gaseous pollutants, and direct dermal contact with the source (e.g. mattress cover).

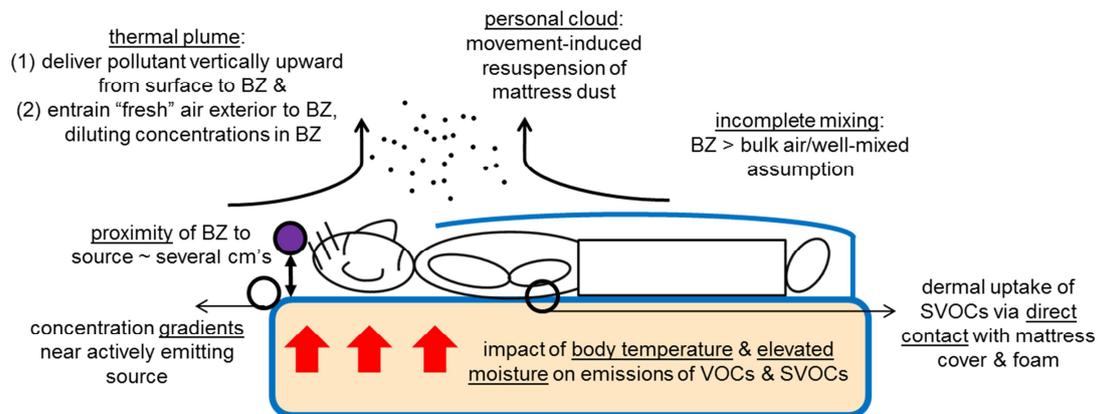


Illustration 2.2: The source-proximity effect of the sleep microenvironment.

The spatial proximity of a person’s BZ to their mattress and bedding can lead to elevated concentrations in their BZ relative to the bulk bedroom air. Laverge et al. (2013), using a breathing thermal manikin positioned on a twin-size mattress with a pillow, found BZ concentrations of a tracer gas released from the mattress to be significantly greater than those measured in the bulk air and found BZ concentrations to be influenced by sleep position, with elevated exposures for the prone position (on the stomach) compared to the supine (on the back). The spatial proximity effect is due in part to incomplete mixing of the released pollutant with the bulk air, which leads to the development of concentration gradients in the space. Incomplete mixing may be aided by the low ventilation rates that have been reported for bedrooms and the reduced run time of ventilation systems at night. Bekö et al. (2010) measured the ventilation rates in the naturally ventilated bedrooms of 500 children in Denmark and reported a mean ventilation rate of  $0.46 \text{ h}^{-1}$ , with rates as low as  $0.1 \text{ h}^{-1}$ , suggesting that children’s bedrooms are poorly ventilated. The personal cloud effect, whereby people can be envisioned as being surrounded by a cloud of

particles that they stir-up from indoor surfaces, is another factor that may lead to elevated exposures in the sleep microenvironment. This effect forms the basis of Paper D and Spilak et al. (2014) on human body movement-induced mattress dust resuspension.

The buoyant human thermal plume may also be responsible for elevated BZ concentrations for pollutant sources in close proximity of the human body. Rim and Novoselac (2009) and (2010) demonstrated that the thermal plume is an effective mechanism for transporting pollutants, released from various locations in close vicinity to the human body, vertically upward, toward to BZ. Thus, the thermal plume that develops around a sleeping person may aid in transporting pollutants released from the mattress surface, upward and toward their BZ. However, the thermal plume may also serve the secondary role of entraining “fresh” air exterior to the sleep microenvironment, thereby diluting concentrations of mattress-released pollutants in the BZ, as suggested by Laverge et al. (2013).

Another unique attribute of the sleep microenvironment that may play an important role in enhancing the source-proximity effect is the warming of nearby objects via heat exchange with the human body. As discussed in Papers B, E, and F, emissions of VOCs and SVOCs from mattresses may increase due to localized elevations in mattress surface temperature (in the range of 30 to 36°C). Additionally, contact transfer and air-to-skin uptake of SVOCs may occur due to the intimate and prolonged contact periods between human skin and their

mattresses (e.g. covers and foam) and bedding materials (e.g. surface-sorbed SVOCs on bedding fibers).

## Chapter 3: Methods

This chapter provides a summary of the methods employed in the original research presented in Papers C, D, E, and F of this dissertation.

### WIND TUNNEL AND CHAMBER MEASUREMENTS

A range of wind tunnels and chambers, varying in size and function, were used in the experimental work of Papers C, D, and E, as outlined below. Such chambers provide a highly controlled means by which to study, systemically, the impact of parameters related to the source, environmental conditions, ventilation system, and the pollutant itself on the release of the pollutant from a surface or material, such as the resuspension fraction and rate of deposited particles and the emission rate of organic contaminants. Findings from chamber measurements can be used to validate indoor air and aerosol models and are useful in interpreting results from field investigations. As discussed in Papers C, D, and E, chamber studies have inherent limitations since they cannot capture the many complexities of real indoor environments and their results should be viewed through the context of the conditions in which the experiments were conducted.

#### *Wind Tunnel Resuspension Study*

An experimental methodology was developed in Paper C to study aerodynamic resuspension from monolayer and multilayer particle deposits on linoleum flooring and galvanized sheet metal. To generate both types of deposits, two different seeding methods were developed. Monolayer deposits were created by depositing neutralized 3 and 10  $\mu\text{m}$  spherical polystyrene

fluorescent particles in a 50 L seeding chamber, as shown in Figure 3.1 (i.). Multilayer deposits were created by first depositing a surface layer of 10  $\mu\text{m}$  fluorescent particles, followed by a varying dust load-deposit of polydisperse 1 to 20  $\mu\text{m}$  Arizona Test Dust (ATD) and a top, canopy layer of 3  $\mu\text{m}$  fluorescent particles. Seeded samples were conditioned at the studied relative humidity for 24 hours, after which they were exposed to controlled airflow in a wind tunnel for a period of 100 seconds (Figure 3.1 (ii.)). Additional details on the particle generation and wind tunnel methods can be found in Paper C and Boor et al. (2011), the latter of which presents the design of a high velocity wind tunnel with a turbulent wall jet.

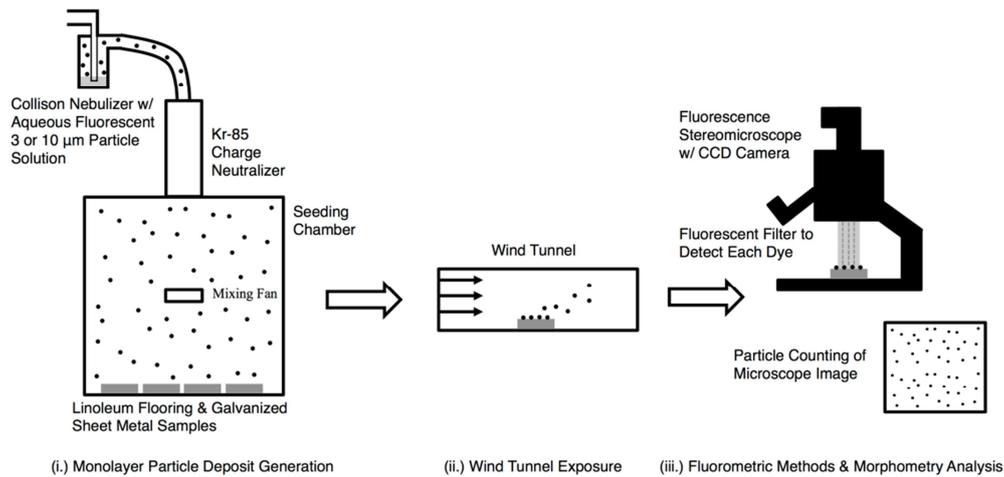


Figure 3.1: Experimental methodology for monolayer resuspension: (i.) particle deposit generation, (ii.) wind tunnel exposure, and (iii.) microscopy imaging and morphometry analysis.

The resuspension metric used in this study is an absolute resuspension fraction,  $\Phi$ .  $\Phi$  is defined as the change in seeding density before and after the seeded sample is exposed to a given flow condition in the wind tunnel, divided

by the initial seeding density. It varies between 0, in which there is no detectable resuspension, and 1, for maximum resuspension.

$$\Phi = \frac{\sigma_i - \sigma_f}{\sigma_i} \quad (3.1)$$

The initial,  $\sigma_i$ , and final,  $\sigma_f$ , seeding densities are expressed as the number of particles per unit area, particles/mm<sup>2</sup>. To determine the absolute resuspension fraction,  $\Phi$ , for the monolayer particle deposits and the surface and canopy layers of the multilayer deposit, a fluorescence stereomicroscope equipped with a charge-coupled device (CCD) camera was used (Figure 3.1 (iii.)). Initial and final seeding densities were determined by counting individual particles with a morphometry program. In addition to studying how resuspension varied between the two types of deposits, Paper C evaluated the impact of air velocity, particle size, relative humidity, the type of indoor surface, dust loading, and layer location on  $\Phi$ .

#### *Mattress Dust Resuspension Study*

Paper D extends the experimental resuspension work of Paper C to a full-scale investigation of human movement-induced resuspension of settled mattress dust particles. Resuspension measurements were conducted in a 14.75 m<sup>3</sup> stainless steel chamber supplied with HEPA-filtered air (Figure 3.2). Ten human volunteers were instructed to perform a prescribed routine of five movements on an artificially seeded twin-size coil mattress.

Similar to the indoor material samples from Paper C, the test mattress (wrapped in two layers of 225-thread count bed sheets) was artificially seeded with a deposit of polydisperse 1 to 20  $\mu\text{m}$  ATD in a 2.8  $\text{m}^3$  seeding chamber, as shown in Figure 3.2. Mattress dust loads collected in field studies are typically in the range of 0.1 to  $> 1.0 \text{ g/m}^2$ , as described in Paper B. To represent this range in mattress dust loading, two dust loads were examined: 0.1 and 1.0  $\text{g/m}^2$ . Particles with diameters in the range of 1 to 20  $\mu\text{m}$  are representative of mite and animal allergen particles, fungal spores and fragments, human- and animal-associated bacteria, and skin flakes, as discussed in Paper B. Actual mattress dust particles may vary considerably in their shape, surface features and roughness, hygroscopicity, and composition, properties that can affect their adhesion to mattress and bedding fibers (Paper A). The use of a standardized test dust in Paper D cannot capture the impact of these parameters on resuspension.

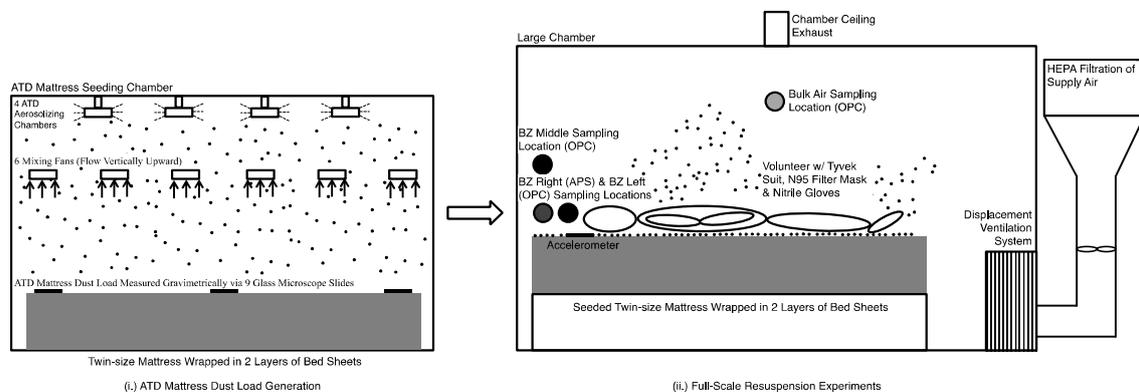


Figure 3.2: Mattress seeding process and large chamber setup for full-scale resuspension experiments.

The seeded mattress was then placed in the full-scale chamber, where a combination of three optical particle counters (OPCs) and one aerodynamic

particle sizer (APS) were utilized to measure particle number concentrations in both the bulk chamber air and the volunteer's breathing zone (BZ) (Figure 3.2). The particle concentrations for all sampling instruments were divided into five particle size fractions: 1-2, 2-3, 3-5, 5-10, and 10-20  $\mu\text{m}$ , corresponding to the size distribution (and size fractions) of the ATD.

Each of the 10 volunteers were instructed to perform a routine of five movements on the mattress, designed to represent common movements of varying intensities that an occupant may perform while in bed (see Figure D2 of Paper D and discussion on movements during sleep periods, e.g. NREM, REM, in Paper B). Upon entering the chamber, the volunteer performed the following routine: M1, sit on mattress; M2, lay in the supine; M3, full 360° rotation to supine position; M4, lay in prone position; and M5, lay in supine position. The entire movement set lasted 12.5 minutes. All volunteers wore a Tyvek clean suit outfit, with booties and a hood, filter mask, and Nitrile gloves. The outfit was selected to protect the volunteer from particle exposure and to prevent the volunteer from acting as a source of particles. In total, twenty-six experiments were conducted (with 10 volunteers), each with two-repeated movement routine sets, for a total of fifty-two sets.

The resuspension rate ( $RR$ ,  $\text{h}^{-1}$ ) metric was used to mechanistically evaluate the impact of various parameters (e.g. dust load, bedroom ventilation rate) on movement-induced resuspension of mattress dust particles. The resuspension rate is defined as the fraction of surface species removed per unit time (Slinn 1978). It is the ratio of the resuspension flux of particles from the

mattress surface ( $\#/m^2h$ ) to the concentration of deposited particles on the mattress surface ( $\#/m^2$ ). It can be determined by applying a two-compartment mass balance for a given size fraction,  $i$ , to the full-scale chamber, as follows:

$$V \frac{dC_i(t)}{dt} = RR_i(t) L_i(t) A - a V C_i(t) - k_i V C_i(t) \quad (3.2)$$

$$A \frac{dL_i(t)}{dt} = k_i V C_i(t) - RR_i(t) L_i(t) A \quad (3.3)$$

where  $V$  is the chamber volume ( $m^3$ ),  $C(t)$  is the particle number concentration in the bulk chamber air ( $\#$  particles/ $m^3$ ),  $A$  is the surface area of the resuspension source (e.g. mattress) ( $m^2$ ),  $k$  is the deposition rate ( $h^{-1}$ ), and  $L(t)$  is the surface concentration of deposited particles ( $\#$  particles/ $m^2$ ). Here, the bulk chamber air is assumed to be uniform and well represented by the OPC positioned at the fixed bulk air monitoring location. Derivation of the expression for  $RR$  and limitations of the two-compartment mass balance are provided in Paper D.

### *Infant Crib Mattress Chamber Study*

In addition to studying the resuspension and transport of mattress dust, this dissertation examined the volatilization of organic contaminants from mattresses. Paper E integrates both small-scale and full-scale chamber measurements to study the emissions of volatile organic compounds (VOCs) from crib mattresses and an infant's exposure to these emissions while sleeping. A collection nine new crib mattresses, manufactured in 2011, and eleven used crib mattresses, manufactured in various years between 1993 and 2009, were analyzed in this study. The mattresses were composed of a thick layer of polyurethane or polyester foam padding (inner-springs were also used) encased

within a thin, waterproof plastic cover. The cover and foam layers of each mattress were analyzed for semi-VOC (SVOC) content in Paper F.

Small-scale emission chambers were used to measure the area-specific emission rate (*SER*) of VOCs from the crib mattress samples at temperatures of 23 and 36°C (infant skin temperature). The small-scale chamber included a field and laboratory emission cell (FLEC) mounted to a 2.5 L cylindrical stainless steel emission chamber. Throughout an emissions experiment, air samples were periodically collected at the chamber exhaust using sorbent tubes (see Figure E1). Several samples were tested without the mattress cover layer to provide insight into the role of the mattress cover in acting as a barrier to the transport of VOCs originating from the foam layer.

Along with the small-scale *SER* experiments, full-scale chamber experiments (similar to the full-scale resuspension measurements in Paper D) were conducted in a 4.5 m<sup>3</sup> emission chamber with a full-size crib mattress and a heated infant thermal manikin (Figure 3.3). The aim of these experiments was to simulate a semi-realistic exposure scenario and experimentally determine the BZ concentration for VOCs emitting from a crib mattress to explore the impact of the source-proximity effect, as outlined in Paper B. The simplified infant thermal manikin was constructed using a hollow galvanized steel cylinder, with heating elements placed uniformly inside. As shown in Figure 3.3, air samples were simultaneously collected at three locations: the interior of the mattress foam, the BZ on the manikin, and the chamber exhaust, representing the bulk room air.

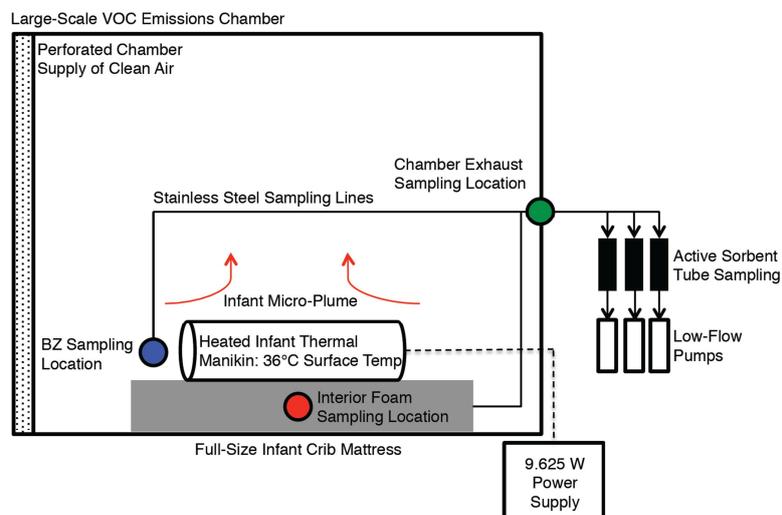


Figure 3.3: Experimental setup for large-scale VOC emission measurements.

### *Analytical Methods*

Various analytical methods were employed to identify and quantify target organic contaminants in both the material-phase (Paper F) and gas-phase (Paper E). The sorbent tubes collected in the small- and large-scale VOC emission measurements of Paper E were thermally desorbed and analyzed with a gas chromatograph (GC) connected to a mass spectrometer detector (MSD, for identification via comparison with a mass spectral library) and a flame ionization detector (FID, for quantification). VOC concentrations were calculated as toluene equivalents in accordance with ISO 16000-6. In Paper F, all crib mattress cover and foam samples were first screened for unreacted isocyanates (NCO) and possible phthalate content via non-destructive photoacoustic (PAS) Fourier transform-infrared (FT-IR) spectroscopy (identification only). The samples were then extracted via ultrasonication in hexane and analyzed by GC-MSD for target plasticizers and flame retardants, including: bis(2-ethylhexyl) phthalate (DEHP),

bis(2-ethylhexyl) isophthalate (iso-DEHP), diisononyl phthalate (DINP), diisononyl 1,2-cyclohexanedicarboxylic acid (DINCH), bis(2-ethylhexyl) adipate (DEHA), triphenyl phosphate (TPP), tris(1,3-dichloro-2-propyl)phosphate (TDCPP), and pentabromodiphenyl ether (pentaBDE), among others. Additional technical details on the analysis of VOCs and SVOCs can be found in Papers E and F and their associated supporting information sections.

#### EXPOSURE CALCULATIONS

In Papers D and E, human exposure to mattress-released pollutants was characterized with two exposure metrics: intake fraction and inhalation intake dose. Paper B provides an additional discussion on exposure characterization for sleep periods.

##### *Intake Fraction*

The intake fraction ( $iF$ ) for a pollutant is defined as the total mass inhaled per unit mass released from a source and can be expressed on a part per million basis (ppm) (Bennett et al. 2002, Marshall and Nazaroff 2007). In the context of the mattress dust resuspension study, it is defined as the ratio of the number of particles inhaled to the number of particles that resuspend. Paper D provides a detailed description of how  $iF$  is determined for the particular experimental sequence of this study. An occupant is exposed to resuspended particles during both the resuspension period (e.g. movements on the mattress) and the following decay period, during which the fate of the resuspended particles is resolved (e.g. via ventilation, deposition, and inhalation). The number of released particles is the product of the resuspension rate ( $RR$ ), surface concentration ( $L$ ), and source

area ( $A$ ). A simplified expression for the time-averaged intake fraction for a given size fraction,  $i$ , derived from BZ particle number concentrations is presented below:

$$\overline{iF}_i = \frac{Q_B}{A} \frac{\left( \overline{C_{i,BZ,Resuspension}} + \overline{C_{i,BZ,Decay}} \right)}{\overline{RR}_i \times \overline{L}_i} \quad (3.4)$$

where  $C_{BZ,Resuspension}$  is the time-averaged BZ concentration during the resuspension period,  $C_{BZ,Decay}$  is the time-averaged BZ concentration during the decay period, and  $Q_B$  is the breathing rate based on data provided by the U.S. EPA EFH (mean of 0.299 m<sup>3</sup>/h for adults 21 to > 81 in age for sleep or nap activity, U.S. EPA 2009).

#### *Inhalation Intake Dose*

In Paper E, the TVOC concentrations as measured in the BZ of the infant thermal manikin were used to estimate an infant's inhalation exposure to mattress-released VOCs. The daily inhalation intake dose can be estimated as the product the BZ TVOC concentration and the volume of air inhaled/kg-day during a sleep period (Figure ES2, based on data provided by the U.S. EPA EFH, U.S. EPA 2009). The volume of air inhaled/kg-day is estimated by taking the product of the mean normalized volumetric breathing rate during a sleep or nap activity (as defined in the U.S. EPA EFH) and the mean duration of time spent in a sleep or nap activity. As discussed in Papers B, E, and F, infants inhale about 250 to 300 L/kg-day, nearly an order of magnitude more air per body mass than adults, which emphasizes the importance of early-life exposures to chemical stressors.

## Chapter 4: Particle Resuspension: Results and Discussion

This chapter discusses results from the experimental resuspension work of Papers C and D, supported with findings from the review of resuspension literature in Paper A.

### AERODYNAMIC PARTICLE RESUSPENSION FROM INDOOR SURFACES

The findings of the wind tunnel resuspension study of Paper C demonstrate that the structure of a particle deposit strongly influences the number of particles that aerodynamically resuspend from an indoor surface due to inherent differences in the resuspension process between monolayer and multilayer deposits.

#### *Monolayer Deposits*

Resuspension of 3 and 10  $\mu\text{m}$  particles from monolayer deposits on linoleum flooring and galvanized sheet metal was found to be generally low for air velocities in the range of 25 to 75 m/s and negligible at velocities below 25 m/s (Table 4.1). This suggests that very high velocities, unrealistic of what would be found in the indoor environment (e.g. airflow in ventilation ducts or airflow induced by footfalls), would be required to aerodynamically-resuspend meaningful quantities of 3 to 10  $\mu\text{m}$  particles from monolayer deposits. Monolayer resuspension may be enhanced when deposited particles experience a combination of aerodynamic and mechanical (surface vibrations, abrasion) removal forces and during periods of highly impulsive airflow, all of which are

likely associated with human movement-induced resuspension (e.g. walking or movements in bed, as discussed in Paper D).

Table 4.1: Absolute resuspension fractions ( $\Phi$ ) for monolayer and multilayer deposits on galvanized sheet metal at velocities from 2.5 to 25 m/s.

velocity (m/s)	monolayer: 3 $\mu\text{m}$ , mean $\pm$ stdev for 30% & 75% RH	multilayer: 3 $\mu\text{m}$ canopy layer, mean $\pm$ stdev for all dust loads
2.5	~ 0	0.05 $\pm$ 0.02
5	~ 0	0.06 $\pm$ 0.01
7.5	~ 0	0.55 $\pm$ 0.32
10	~ 0	0.65 $\pm$ 0.30
25	0.03 $\pm$ 0.01	0.74 $\pm$ 0.07

Resuspension was found to increase with increasing particle size, decrease with increasing relative humidity, and was greater for linoleum flooring compared to galvanized sheet metal. The size-dependence of particle resuspension has been established in early wind tunnel studies (e.g. Corn and Stein 1965, Wu et al. 1992, Nicholson 1993, Braaten 1994). As the particle size increases, so does the ratio of removal forces to adhesion forces (Hinds 1999). The role of relative humidity can be explained by its impact on the adhesion force between the particle and deposition surface (e.g. Corn and Stein 1965, Hinds 1999, Paajanen et al. 2006, Cleaver and Looi 2007, You and Wan 2012). The observed impact of the surface material may be due in part to the greater average surface roughness of linoleum (13.13  $\mu\text{m}$ ) compared to sheet metal (4.31  $\mu\text{m}$ ). Additional parameters have been shown to influence resuspension from

monolayer deposits (see Paper A), including the composition, shape, and surface roughness of the particles; residence time of the particle on a deposition surface; and the acceleration and turbulence of the airflow. For example, particles with irregular surface features, such as pollen or fungal spores, will tend to resuspend at lower air velocities compared to smooth, spherical particles (e.g. Nitschke and Schmidt 2009), such as those used in Paper C.

### *Multilayer Deposits*

Aerodynamically-induced resuspension from multilayer deposits occurred at significantly lower air velocities compared to the monolayer deposits (Table 4.1). For example, absolute resuspension fractions at an air velocity of 5 m/s for the canopy layer of multilayer deposits were similar to those found for monolayer deposits at 50 m/s. Figure 4.1 shows that resuspension from the 3  $\mu\text{m}$  canopy layer of multilayer deposits increased as the dust load increased. Resuspension from the 3  $\mu\text{m}$  canopy layer increased with increasing velocity. The impact of velocity is strongly coupled with the level of dust loading, with the two heaviest dust loads showing a significant increase in resuspension between 5 and 7.5 m/s, and the two lightest dust loads exhibiting a steady increase in resuspension with increasing velocity from 2.5 to 12.5 m/s. The impact of the surface material (linoleum flooring vs. galvanized) was small for multilayer deposits. Resuspension was significantly greater for the 3  $\mu\text{m}$  canopy layer of the multilayer deposit compared to the 10  $\mu\text{m}$  surface layer. For many cases, no detectable resuspension was observed for the surface layer.

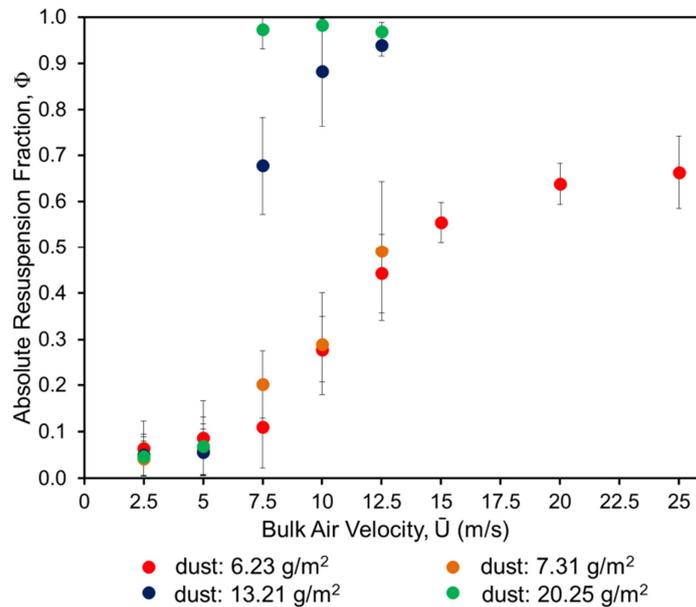


Figure 4.1: Absolute resuspension fractions,  $\Phi$ , for 3  $\mu\text{m}$  canopy layer of multilayer deposit on linoleum flooring.

The findings of this investigation can be explained by considering the fundamental differences in the resuspension process between monolayer and multilayer deposits, as discussed in detail in Paper A. Several unique attributes of multilayer resuspension are highlighted in illustration 4.1: saltation, aggregate resuspension and de-aggregation, deposit porosity, and particle-to-particle adhesion. Saltation may play a role in resuspending smaller particles in multilayer deposits. Large particles or aggregates,  $\sim 100 \mu\text{m}$  in diameter, can be lifted away from a deposit by aerodynamic stresses. These particles are too large to remain airborne, so they return to the deposit and begin to hop along the surface and impact settled particles in a process known as saltation. Thus, a large saltating particle or aggregate can be responsible for the resuspension of smaller particles.

Previous wind tunnels studies have shown that resuspension from multilayer deposits can occur in the form of large particle aggregates, which may de-aggregate due to stresses imparted by turbulent eddies (Gac et al. 2008). Loosely bound and “fluffy” multilayer deposits that are formed by gravitational settling of particles onto indoor surfaces likely results in enhanced resuspension compared to more compact and “cake-like” deposits that are formed by deposition mechanisms such as inertial impaction (Friess and Yadigaroglu 2002). Multilayer resuspension is also influenced by particle-to-particle adhesion forces. Lazaridis and Drossinos (1998) demonstrated that the adhesion force between two spherical particles is less than that between a spherical particle and a flat surface. This reduced particle-to-particle adhesion may explain why the particles from the canopy layer of the multilayer deposit detached at lower air velocities compared to the particles distributed along the surface of the monolayer deposits.

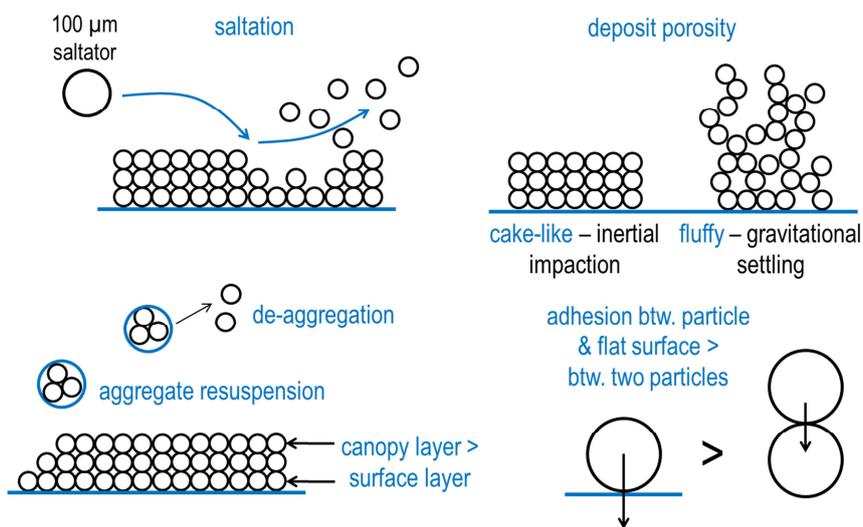


Illustration 4.1: Multilayer resuspension dynamics.

### *Particle Deposit Height and Viscous Sublayer Thickness*

The differences in aerodynamic resuspension between monolayer and multilayer deposits can be further explained by considering the relationship between the absolute resuspension fraction,  $\Phi$ , and the ratio of particle deposit height,  $\delta$  (Equation 2.1) to the thickness of the viscous sublayer,  $y_{VSL}$ , as shown in Figure 4.2. An empirical logistic relationship between  $\Phi$  and  $\delta/y_{VSL}$  is presented in Paper C.

Observing the logistic trend presented in Figure 4.2,  $\Phi$  grows exponentially to a  $\delta/y_{VSL}$  of unity, after which, its growth rate slows and it begins to asymptotically approach 1. For small values of  $\delta/y_{VSL}$ , minimal resuspension was found to occur. When the particle deposit is completely immersed in the viscous sublayer ( $y_{VSL} \gg \delta$ ), as is the case for monolayer deposits at low velocities, particles will not experience the enhanced removal associated with frequent collisions with turbulent eddies, and resuspension only occurs due to the periodic penetration of turbulent bursts from the fully turbulent sublayer (e.g. Cleaver and Yates 1973). As  $\delta$  begins to approach  $y_{VSL}$ , a sharp increase in  $\Phi$  is observed. As  $\delta/y_{VSL}$  approaches unity, where we observe an inflection point in  $\Phi$ 's dependence on  $\delta/y_{VSL}$ , the edge of  $y_{VSL}$  is at roughly the same height as the particle deposit. This was only achieved for the multilayer deposits, as the velocities studied for the monolayer deposits (25 to 75 m/s) were not large enough to reduce  $y_{VSL}$  to a height equivalent to that of the particles (3 and 10  $\mu\text{m}$ ).

Beyond a  $\delta/y_{VSL}$  of unity, where the height of the deposit begins to surpass  $y_{VSL}$  and enter the fully turbulent sublayer, very high levels of resuspension are achieved and  $\Phi$  begins to asymptotically approach 1. We can expect that at increasingly larger values of  $\delta/y_{VSL}$ , due to an increase in bulk air velocity and/or an increase in the dust load,  $\Phi$  will remain near 1 and the canopy layer will be completely resuspended. When increasing fractions of the multilayer particle deposit enter the fully turbulent sublayer, they likely experience the enhanced removal forces associated with turbulent eddies.

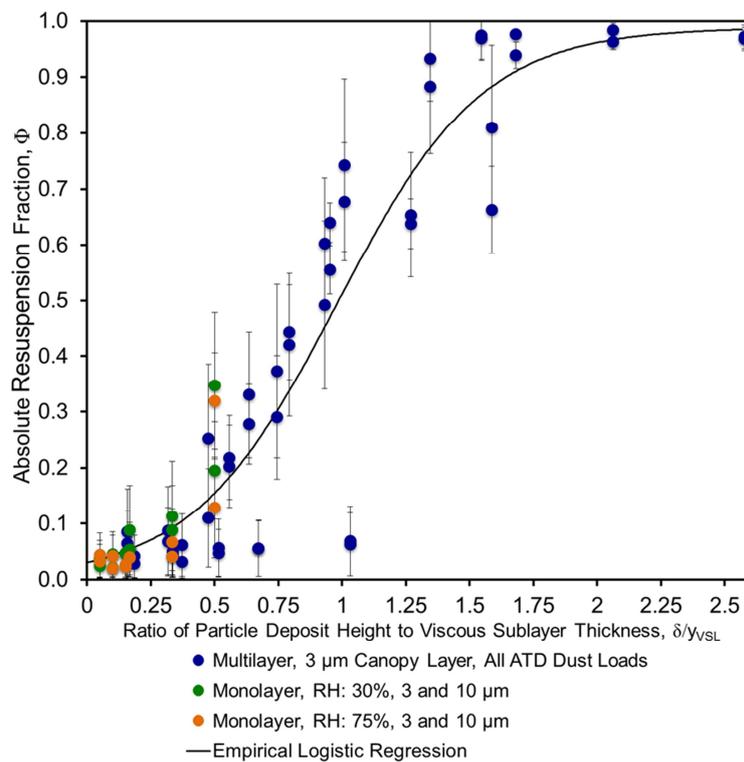


Figure 4.2: Relationship between the absolute resuspension fraction,  $\Phi$ , of both monolayer deposits and the canopy layer of multilayer deposits with the ratio of  $\delta$  to  $y_{VSL}$ .

The concept of  $\delta/y_{VSL}$  can help explain the role of the level of dust loading on resuspension from multilayer deposits. For velocities between 2.5 and 12.5 m/s,  $\Phi$  was found to increase with dust loading from 6.23 to 20.25 g/m<sup>2</sup>. As dust load increases,  $\delta$  is expected to increase, and therefore, the ratio of  $\delta/y_{VSL}$  for a given air velocity. The ratio of  $\delta/y_{VSL}$  can also increase due to an increase in the air velocity (decreasing  $y_{VSL}$ ). As previously discussed, this study found  $\Phi$  to be strongly dependent on the air velocity and increase with increasing velocity. Fromentin (1989), Huang et al. (2005), and Matsusaka and Masuda (1996) observed similar trends in their respective wind tunnel studies.

#### MATTRESS DUST RESUSPENSION

Although wind tunnel studies, such as the one conducted in Paper C, provide basic knowledge on resuspension, they cannot capture the complexities of human-induced resuspension. As such, the findings presented in Paper D provide a more complete picture of the dynamics of human-induced resuspension. Human body movements in bed were found to resuspend mattress dust particles in the range of 1 to 20  $\mu\text{m}$ . Significant elevations in airborne concentrations of resuspended particles were observed for each individual movement, M1-M5, as shown in a typical concentration time series in Figure 4.3. These short-term concentration peaks were approximately one to two orders of magnitude greater than the background concentration. Dust loading had a significant impact on particle number concentration. Concentrations during the movement routine on the mattress seeded with 1 g/m<sup>2</sup> of dust were typically an order of magnitude greater than those with 0.1 g/m<sup>2</sup>.

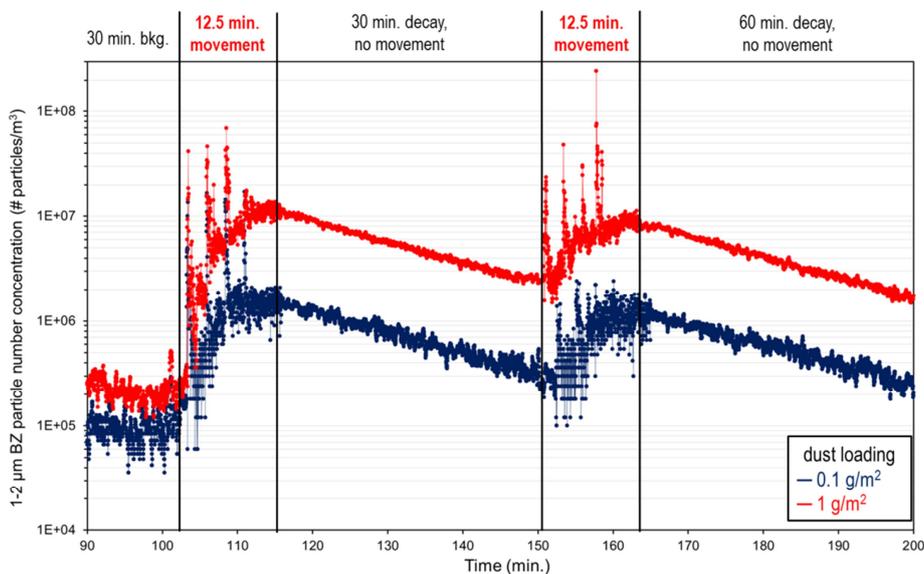


Figure 4.3: Example particle number concentration profile during a resuspension experiment: 1 to 2  $\mu\text{m}$  particles at dust loads of 0.1 and 1.0  $\text{g}/\text{m}^2$ .

#### *Resuspension Rate: Influencing Factors*

The average resuspension rates among the 10 volunteers (over entire movement routine, M1-M5) for selected data are shown in Figure 4.4. The *RRs* were found to range over four orders of magnitude from  $10^{-3}$  to  $10^1 \text{ h}^{-1}$ . *RR* increases with increasing particle size, by approximately 3 orders of magnitude, from 1 to 2  $\mu\text{m}$  to 10 to 20  $\mu\text{m}$ . The size-dependency of resuspension is in agreement with the results of Paper C and is expected, given the basic mechanisms of particle detachment from surfaces, as discussed earlier in this chapter and in Paper A.

Dust loading appears to have a negligible impact on *RR*, despite the significant differences observed in airborne particle number concentrations. For

dust loads of 0.1 and 1.0 g/m<sup>2</sup>, particles are very likely deposited as a monolayer along the fiber, with minimal particle-to-particle contact. Thus, the impact of the type of particle deposit is much less pronounced than in the wind tunnel study of Paper C. The size-resolved *RRs* estimated here are similar in magnitude to those reported by walking-induced resuspension studies (e.g. Qian and Ferro 2008). This suggests that movement-induced resuspension in bed is an important source of coarse-mode dust particles, comparable to particles released from flooring via footfalls. Interestingly, body mass had a minimal impact on *RR* (Figure DS2). Intuitively, resuspension would be expected to increase with body mass, however, it is possible that beyond a certain threshold weight, the removal forces induced by body movements in bed is much more dependent on movement intensity and technique, rather than their body mass alone.

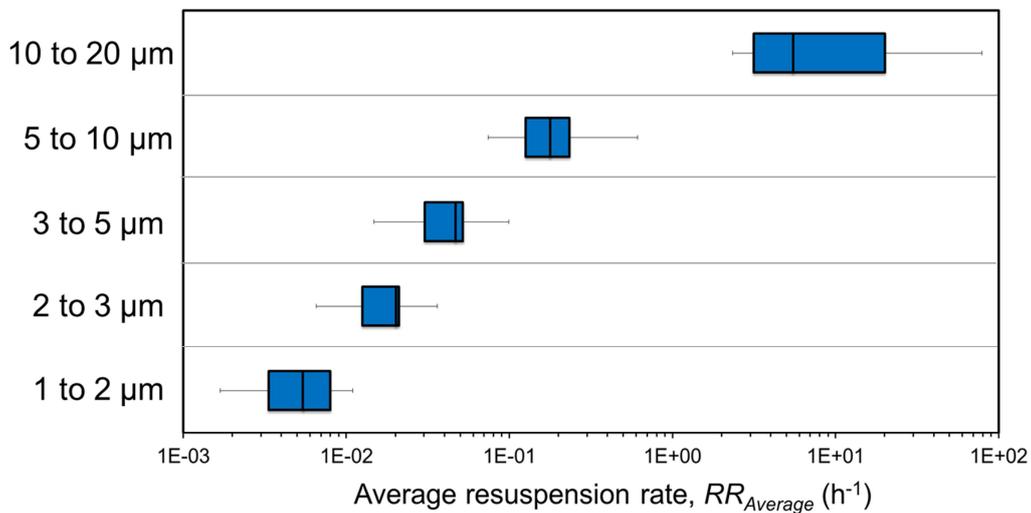


Figure 4.4: Average resuspension rates (*RR*) among 10 volunteers over entire movement routine of set 1 (M1-M5), for 0.1 g/m<sup>2</sup> dust load at a ventilation rate of 2.9 h<sup>-1</sup>. Box plots represent interquartile range and whiskers represent the 5th and 95th percentiles.

### *Movement Intensity and Resuspension Mechanisms*

Body movements of greater intensity (e.g. M3, 360° rotation of the torso) increased particle resuspension compared to less intense movements (e.g. M1, sitting on the mattress), as shown in Figure 4.5. This can be explained by considering the surface vibrations and impulsive airflow induced by each movement. Each of the five movements, M1-M5, was associated with a peak in mattress surface vibration, generally between 0.1 and 1 *g* (9.81 m/s<sup>2</sup>) in magnitude. To put these values in perspective, Gomes et al. (2007) reported walking-induced peak floor vibrations of generally  $\leq 0.1$  *g*. Movements of greater intensity, such as a full body rotation (M3), generate greater surface vibrations, and in turn, increase the magnitude of the associated removal forces, such as the wall vibration force (Theerachaisupakij et al. 2002) and lift-off drag force (Gomes et al. 2007), thereby enhancing particle resuspension.

Each of the five movements, M1-M5, was associated with a peak in air velocity ~ 1 cm above the mattress surface. The air velocities increased suddenly, suggesting impulsive and highly accelerated airflow, similar to airflows generated by footfalls or descending objects (e.g. Khalifa and Elhadidi 2007). The impulsive nature of the flow may be considerably more important in inducing resuspension from the bed sheets than the maximum velocities achieved, as discussed in Paper A. Mechanical abrasion caused by direct contact between the volunteer and the deposited particles, as well as sections of the bed sheets rubbing against each other, likely contributed to particle detachment.

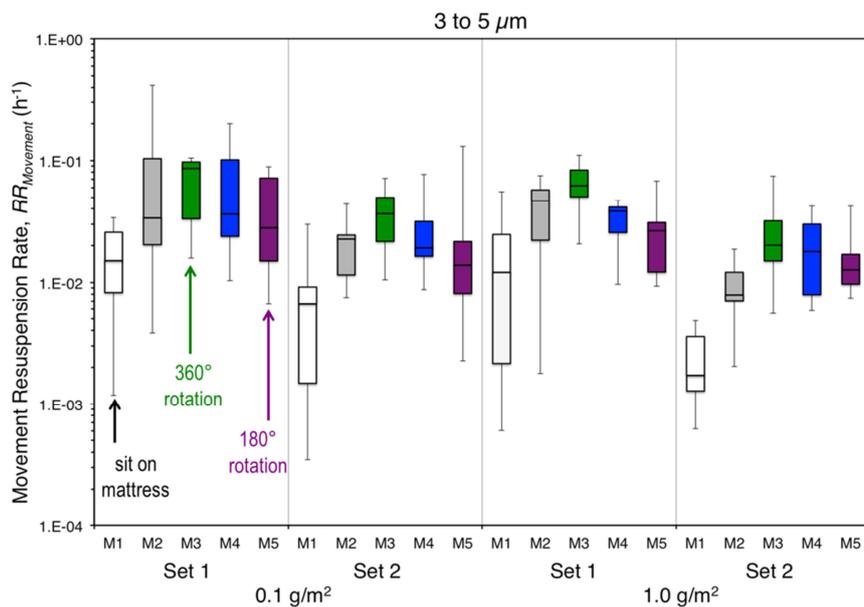


Figure 4.5: Average resuspension rates ( $RR$ ) among 10 volunteers for each individual movement (M1, M2, M3, M4, M5) for 3 to 5  $\mu\text{m}$  particles. Box plots represent interquartile range, whiskers represent the 5th and 95th percentiles.

#### *Intake Fraction and Inhalation Exposure to Resuspended Particles*

Time-averaged intake fractions ( $iF$ ) were generally in the range of  $10^2$  to  $10^4$  inhaled particles per million resuspended (Figure 4.6). For particles in the range of 1 to 10  $\mu\text{m}$ ,  $iFs$  are generally on the order of  $10^3$  to  $10^4$  ppm, and for particles in the range of 10 to 20  $\mu\text{m}$ ,  $iFs$  are generally on the order of  $10^2$  ppm. Although the  $RR$  increases with increasing particle size, the opposite is observed for  $iF$ . Thus, even though larger particles more easily resuspend, a greater fraction of smaller particles that resuspend will be inhaled. This is due to a combination of lower deposition rates for smaller particles (listed in Figure 4.6) and greater breathing zone concentrations for smaller particles, the latter of which is due in part to the size distribution of the ATD used to generate the

artificial mattress dust deposits (mass median diameter of 4.5  $\mu\text{m}$ ). The smaller particles have a greater likelihood of being inhaled, rather than be removed via gravitational settling to the mattress surface, compared to the larger particles (Marshall and Nazaroff 2007, Nazaroff 2008).  $RR$  was not strongly influenced by the chamber ventilation rate, however,  $iF$  increased with decreasing ventilation rate from 7.4 to 0.9  $\text{h}^{-1}$  for particles in the 1 to 10  $\mu\text{m}$  range. Increasing the ventilation rate increases the fraction of particles that are removed from the chamber air rather than inhaled, thus decreasing  $iF$ .

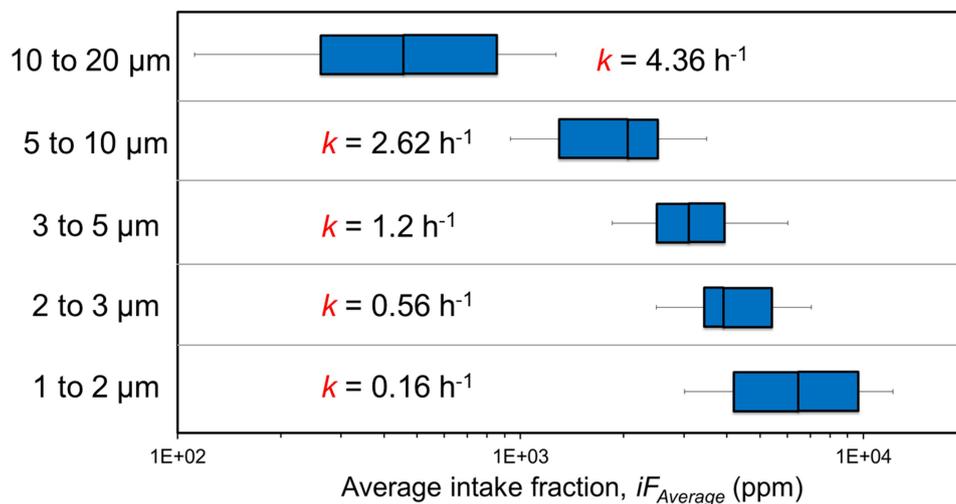


Figure 4.6: Average intake fractions ( $iF$ ) among 10 volunteers over entire movement routine of set 1 (M1-M5), for 0.1  $\text{g}/\text{m}^2$  dust load at a ventilation rate of 2.9  $\text{h}^{-1}$ . Box plots represent interquartile range and whiskers represent the 5th and 95th percentiles. Deposition rates ( $k$ ) for each size fraction are listed.

The non-negligible fractions of resuspended particles that are inhaled are due in part to the important role of the source-proximity effect on exposure to pollutants originating in the sleep microenvironment, as discussed in Paper B. The ratios of the spatial BZ average concentrations to the bulk air concentrations

were in the range of 1.07 to 1.94. These results are similar to those reported in Paper E for VOCs. Lastly, as shown in Figure 4.3, elevated concentrations after the completion of a movement routine suggests that people can be exposed to resuspended particles as they lay still in bed, following a resuspension event.

## Chapter 5: Infant Exposure to Chemical Emissions from Crib Mattresses: Results and Discussion

This chapter discusses results from the experimental work of Papers E and F on characterizing crib mattresses as a source of organic contaminants in the sleep microenvironment and infant exposure to selected compounds while sleeping.

### VOLATILE ORGANIC COMPOUNDS

#### *VOC Area-Specific Emission Rates*

All new and used crib mattresses were found to emit VOCs. Across all twenty samples, TVOC area-specific emission rates (*SERs*) ranged from 3 to 385  $\mu\text{g}/\text{m}^2\text{h}$  (mean of 56  $\mu\text{g}/\text{m}^2\text{h}$ ) at 23°C and from 8 to 697  $\mu\text{g}/\text{m}^2\text{h}$  (mean of 139  $\mu\text{g}/\text{m}^2\text{h}$ ) at 36°C (Figure 5.1) TVOC *SERs* in this range are similar to emissions from other consumer products and building materials that may be found in an infant bedroom or nursery, such as PVC and laminate flooring, wall and floor coverings, and plastic toys (see references in Paper E).

VOC emissions were influenced by temperature, age of the mattress, and presence of the cover layer. As shown in Figure 5.1, the TVOC *SER* increased with temperature and was about double at 36°C compared to 23°C. A VOC's vapor pressure tends to increase with temperature. At 36°C, a VOC molecule has a greater affinity for the gas phase, thereby enhancing its partitioning from the material-phase to the chamber air. The temperature dependence is particularly relevant to the infant sleep microenvironment (as introduced in Paper B),

because heat released from a sleeping infant will warm surrounding objects, including their mattress. Thus, VOC emissions may increase due to localized elevations in mattress surface temperature.

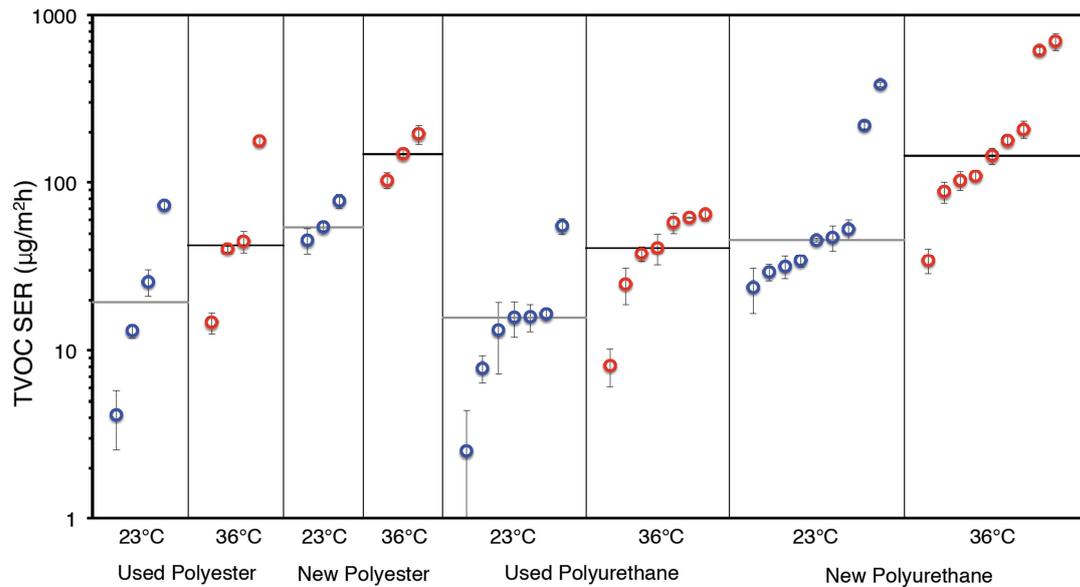


Figure 5.1: TVOC *SERs* categorized by temperature, usage, and foam material.

New crib mattresses were found to release greater quantities of VOCs compared to used mattresses (Figure 5.1). This is expected, as VOC emissions tend to decay as the material-phase concentration in the source depletes over time. The crib mattress cover layer was found to act as a diffusive barrier to the transport of VOCs originating in the foam layer, with the TVOC *SER* increasing as the cover layer was removed for two mattress samples. Although not investigated in this chamber study, elevated relative humidity in the sleep microenvironment will likely increase VOC emissions (see Haghghat et al. 1998).

### *VOC Speciation*

The most abundant VOCs were identified and their individual VOC *SERs* are presented in Table 5.1. *SERs* varied widely among different compounds and crib mattress samples; they ranged from < 1 to 62  $\mu\text{g}/\text{m}^2\text{h}$  at 23°C and from 2 to 257  $\mu\text{g}/\text{m}^2\text{h}$  at 36°C. The impact of foam material is evident in regard to the specific VOCs released from the crib mattress samples. Polyurethane foam showed a greater diversity of VOCs, many of which were not identified in polyester foam (or identified much less frequently), including phenol, 2-ethyl-hexanoic acid, d-limonene, and linalool. Phenol is used as an intermediate in the production of phenolic resins that may be used in the production of polyurethane foam; 2-ethyl-hexanoic acid is used as a catalyst in the production of polyurethane foam; d-limonene is commonly used as a fragrance, as well as a solvent and wetting agent in the manufacture of resins; and linalool is used as a fragrance. Several VOCs were detected more frequently among used samples, including nonanal (6 used, 1 new), isopropyl myristate (5 used, 0 new), and palmitic acid (2 used, 0 new). Nonanal, which was detected in 6 of the 11 used samples, is an alkyl aldehyde that is released by human skin and also found in building materials, while isopropyl myristate and palmitic acid are fatty acid esters that can be found in personal care products, such as lotions and ointments. Because polyurethane foam has a high sorption capacity and can adsorb airborne VOCs over time, the compounds detected among the used samples may represent adsorbed VOCs that originated elsewhere in a residence rather than VOCs used in the production of the mattress.

Table 5.1: VOC *SERs* of the most abundant and commonly identified VOCs among new and used infant crib mattresses.

Compound <sup>1</sup>	New/ Used	Number of Samples		VOC <i>SER</i> at 23°C (µg/m <sup>2</sup> h)	VOC <i>SER</i> at 36°C (µg/m <sup>2</sup> h)
		PUF	Poly.		
Phenol	New	5	1	< 1 – 62	3 – 257
	Used	1	1		
Isooctanol	New	1	1	< 1 – 6	4 – 7
	Used	1	--		
Neodecanoic acid	New	--	3	3 – 22	9 – 40
	Used	--	1		
Hexanoic acid, 2-ethyl-	New	5	--	< 1 – 55	5 – 213
	Used	1	1		
1-Heptanol, 3-methyl	New	2	2	7 – 21	7 – 22
	Used	--	--		
D-Limonene	New	2	--	4 – 11	9 – 18
	Used	--	--		
2,6-Bis(1,1-dimethylethyl)-4-(1-oxopropyl)phenol	New	2	--	4 – 14	12 – 61
	Used	--	--		
(S)-3-Ethyl-4-methylpentanol	New	1	1	2 – 6	3 – 9
	Used	1	--		
Linalool	New	2	--	3 – 41	10 – 44
	Used	--	--		
Nonanal	New	1	--	< 1 – 5	2 – 10
	Used	2	4		
Decanal	New	1	--	< 1 – 5	2 – 10
	Used	1	1		
Isopropyl Myristate	New	--	--	< 1 – 3	3 – 11
	Used	3	2		
Palmitic acid	New	--	--	2 – 10	12 – 43
	Used	1	1		
2-Ethylhexanol	New	1	--	3 – 6	7 – 8
	Used	1	1		

<sup>1</sup>: All compounds identified with at least 75% match with NIST mass spectral library.

### *Source-Proximity Effect of the Infant Sleep Microenvironment*

The full-scale chamber measurements of Paper E demonstrate that infants can be exposed to elevated VOC concentrations in their BZ compared to levels in

the bulk bedroom air. As shown in illustration 5.1, TVOC concentrations sampled at the BZ were significantly greater than those of bulk air sampled at the chamber exhaust. BZ concentrations ranged from 14.2 to 33.2  $\mu\text{g}/\text{m}^3$  (mean of 26.2  $\mu\text{g}/\text{m}^3$ ), the exhaust concentrations ranged from 8.0-18.4  $\mu\text{g}/\text{m}^3$  (mean of 13.5  $\mu\text{g}/\text{m}^3$ ), and the ratio of the BZ concentration to the exhaust concentration ranged from 1.7 to 2.4 (mean of 2), depending on the particular experimental case (defined in Paper E). These results suggest that there is a source-proximity effect associated with exposure to VOCs released from crib mattresses (also identified for resuspended mattress dust particles in Paper D), which can be explained by considering the unique attributes of exposures in the sleep microenvironment, as outlined in Chapter 2 and Paper B. TVOC concentrations sampled in the interior foam the crib mattress (range of 137.9-168.4  $\mu\text{g}/\text{m}^3$ , mean of 154.4  $\mu\text{g}/\text{m}^3$ ) were found to be nearly an order of magnitude greater than those in the BZ region or bulk room air (illustration 5.1). The highly concentrated pore air may be released as the foam is compressed and decompressed in a cyclic manner due to infant body movements.

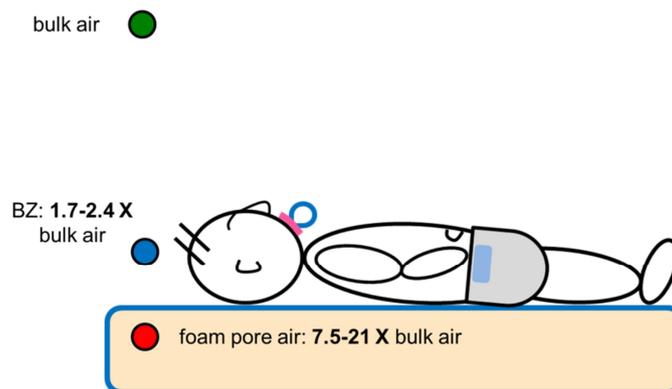


Illustration 5.1: The source-proximity effect of the infant sleep microenvironment.

The BZ TVOC concentrations were used in a simple inhalation exposure analysis to estimate sleeping inhalation doses for infants. An infant's daily sleeping inhalation dose for VOCs originating in a crib mattress was found to range from 6.4 to 8  $\mu\text{g}/\text{kg}\text{-day}$ , depending on age (newborn to 2 years). For comparison, inhalation doses on this order of magnitude are greater than those reported by Masuck et al. (2011) for infant exposure to VOC emissions from toy fragrances (range of 2.2 to 220  $\text{ng}/\text{kg}\text{-day}$ ) and on the same order of magnitude as those reported by Kim et al. (2007) for toluene (4.5  $\mu\text{g}/\text{kg BW}/\text{day}$ ) and benzene (1  $\mu\text{g}/\text{kg BW}/\text{day}$ ) in living room air.

#### CHEMICAL ADDITIVES IN CRIB MATTRESS COVERS AND FOAM

Paper F extends the work of Paper E and evaluates crib mattress covers and foam (same 20 mattress samples) as a source of phthalate and alternative plasticizers, flame retardants, and unreacted (free) isocyanates (NCO). Material-phase concentrations of target compounds are presented in the following subsections as mass of the compound per mass of material,  $\text{mg}/\text{g}$ , or % by weight of material.

##### *Phthalates and Alternatives Plasticizers*

17 of the 20 crib mattress covers contained at least one identifiable plasticizer at concentrations greater than 1  $\text{mg}/\text{g}$  (0.1% by weight) and the most common plasticizer detected was DINP, found in 40% of all samples. The mean total plasticizer content among new and used covers was 135.5  $\text{mg}/\text{g}$  and 151.1  $\text{mg}/\text{g}$ , respectively. 60% of the samples contained more than 90  $\text{mg}/\text{g}$  of

plasticizers and 20% contained over 200 mg/g of plasticizers. The plasticizer concentrations in crib mattress covers are comparable to those reported for a vinyl pillow protector (140 mg/g of DEHP) (Dodson et al. 2012), a nursing pillow cover (144 mg/g of DINP) (Danish Ministry of the Environment 2008), and a children's sofa (210 mg/g of DEHP) (Kawakami et al. 2011).

DINCH, marketed under the name Hexamoll® (BASF 2014), was the most frequently detected plasticizer in new covers (44.4% of samples) and DINP was the most frequently detected plasticizer in used covers (63.6% of samples). The two phthalate plasticizers, DEHP and DINP, were detected in nine of the 20 covers. Five samples contained both phthalates, with DINP typically occurring at greater concentrations relative to DEHP. DEHA was primarily detected in crib mattresses made in 2008 and earlier, whereas two of the three samples containing iso-DEHP, marketed under the name "Flexol Plasticizer 380" (Morose and Becker 2013), were made in 2011.

Plasticizer concentrations in crib mattress polyurethane or polyester foam were typically in the range of 0.1 to 10 mg/g, although several foam samples contained higher concentrations, such as sample 13 with 63.6 mg/g of DINP. There were 13 occurrences of paired-detection of a specific plasticizer in both the cover and foam layers and higher plasticizer concentrations were associated with older mattresses. This suggests that, over time, the foam behaved as a sorptive reservoir for gas-phase plasticizers released from the overlying cover layer or from products found elsewhere in a residence. The gas-phase plasticizers may

also sorb to bedding and blanket fibers, a process that likely influences their transport to the infant BZ.

Although concentrations of a specific plasticizer in crib mattress covers did not show strong associations with the age of the mattress, trends in the occurrence of plasticizers were observed in regard to the U.S. Consumer Product Safety Improvement Act of 2008 (CPSIA), as shown in Figure 5.2. The CPSIA limits the concentrations of DEHP, DBP, and BBP in children’s toys or child-care articles to below 0.1% by weight (1 mg/g) (U.S. CPSC 2008). Furthermore, a recent report by the Chronic Hazard Advisory Panel (CHAP) to the U.S. Consumer Product Safety Commission (CPSC) recommended that DINP be permanently included in this list (CHAP 2014). However, the inclusion of a crib mattress in the definition of a child-care article is not clear (e.g. NRDC 2013).

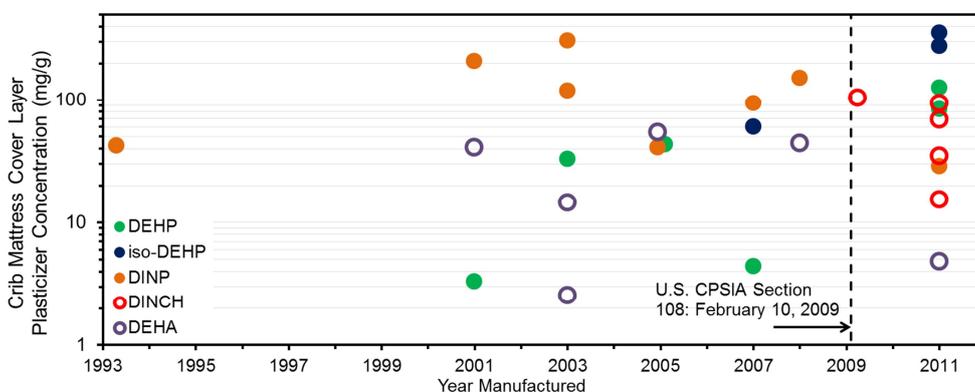


Figure 5.2: Plasticizer concentrations in the mattress cover layers versus the year crib mattress samples were manufactured. Date on which the U.S. Consumer Product Safety Improvement Act (CPSIA) Section 108 was enacted in the U.S (February 10, 2009) denoted by dashed line.

Since the CPSIA went into effect, there has been more frequent use of phthalate alternatives, including DINCH and iso-DEHP, which were identified

in seven of the 10 cover samples manufactured from 2009 to 2011. The use of DINCH as a phthalate replacement in crib mattresses follows the trend observed in other soft PVC products (ECHA 2006), and isophthalates (iso-DEHP) are being used in the same manner (Morose and Becker 2013). However, DEHP concentrations in two of the crib mattress covers manufactured after the CPSIA went into effect (sample 3: 83.1 mg/g, sample 20: 125.7 mg/g) exceeded the 1 mg/g limit of the CPSIA for child-care articles. Considering all 20 crib mattresses, nine would not have met the current CPSIA limits or CHAP recommendations as they contained either DEHP or DINP at concentrations greater than 1 mg/g.

#### *Flame Retardants and Unreacted (Free) Isocyanates*

Polybrominated diphenyl ether (PBDE) congeners associated with the pentaBDE mixture were identified in three crib mattress foam samples, two of which were manufactured before the 2004 pentaBDE phase-out. Triphenyl phosphate (TPP) was identified in the three samples containing pentaBDE congeners, as well as several additional foam samples and two crib mattress covers, where it may have been used as a plasticizer. The co-occurrence of the two flame retardants is likely the result of both being used in combination during the manufacture of the foam, or adsorption of gas-phase TPP or pentaBDEs in a residence over time. In total, eight of the 20 crib mattress foam layers contained pentaBDE congeners, TPP, or tris(1,3-dichloro-2-propyl)phosphate (TDCPP), all of which were manufactured with polyurethane foam. All crib mattresses with a label indicating that it met one of the following three flammability standards: Technical Bulletin (TB) 116, TB 117, or FF 4-72, contained either pentaBDEs or

TPP, or both. NCO, as identified through PAS-FT-IR analysis, was detected in all 13 polyurethane foam samples, likely at greater levels in new mattresses relative to used.

### *Exposure Implications*

The identified plasticizers and flame retardants may partition to and accumulate on mattress dust particles and bedding fibers. Infants may be exposed to the particle-bound SVOCs through body movement-induced resuspension of mattress dust (Paper D) and may be exposed to the surface-sorbed SVOCs via direct contact between their skin and bedding fibers. Given the slow and nearly constant emission rate of plasticizers from soft PVC materials, it is likely that crib mattresses are a constant plasticizer source in the infant sleep microenvironment, which is important, considering the long lifetime of a crib mattress (~10 years) and mattress re-use in families with multiple children. This is in contrast to VOC emissions that tend to decay over time. Thus, even though older mattresses emit less VOCs than newer mattresses (Paper E), they may contain high material-phase concentrations of DEHP and pentaBDEs (Paper F).

Infant exposure to flame retardants originating in the crib mattress foam should be compared to an infant's dermal exposure while wearing pajamas and sleepwear treated with flame retardants (Blum et al. 1978). Additionally, the inhalation and dermal exposures of pregnant women while sleeping on a flame retardant-treated mattress requires further investigation.

## Chapter 6: Conclusions

This dissertation fulfilled three research objectives (Chapter 1) related to particle resuspension, infant exposure, and the sleep microenvironment. In summary, this research contributed to the fields of indoor air quality (IAQ), aerosol science, and exposure science by (practical implications of this work are also discussed):

- 1) Demonstrating that dust loads on indoor surfaces can range over several orders of magnitude, suggesting the existence of both monolayer and multilayer deposits, which may have important implications for particle resuspension in buildings. Controlling the accumulation of dust on flooring through regular cleaning and on ventilation ducts via duct cleaning services can be a practical means to reduce the mass of house dust that is aerosolized. Concerning the latter, the IAQ benefits of duct cleaning remain unclear (see Paper A and review by Zuraimi 2010).
- 2) Discovering that particle resuspension can occur at considerably lower air velocities for the canopy layer of multilayer deposits compared to monolayer deposits. Thus, particles, including allergens, fungi, and bacteria, contained within multilayer deposits will be more easily released into the air upon a physical disturbance.
- 3) Showing, for the first time, that human body movements in bed can resuspend mattress dust particles and that the resuspension process is

influenced by mattress surface vibrations, impulsive airflow across the bed sheets, and intensity of the body movement.

- 4) Establishing that human body movement-induced resuspension can be a source mechanism for the broad spectrum of biological matter (e.g. allergens, fungi, and bacteria) and chemical contaminants (e.g. SVOCs) that accumulate in mattress dust and that people can inhale on the order of 100 to 10,000 particles per million that resuspend. Source control (e.g. vacuuming of mattresses and washing of bed sheets), use of portable filtration units in close proximity to the bed, and improved bedroom ventilation may provide a means to reduce sleep exposures to resuspended mattress dust particles.
  
- 5) Discovering that crib mattresses can emit a multitude of chemical contaminants into the air around a sleeping infant and that the emission rate of VOCs is influenced by the body temperature of an infant, age of the mattress, and presence of the mattress cover. Extended "air-out" periods for crib mattresses prior to mattress use, either at the manufacturing plant or in a well-ventilated room in a residence, are recommended to reduce infant VOC exposure as VOC emissions tend to decay with time as the concentration in the material-phase is slowly reduced. When possible, parents should research crib mattresses that are certified by independent laboratories as having low VOC emission rates.
  
- 6) Identifying various chemical additives in crib mattress covers and foam, including compounds that have known endocrine disrupting properties in

humans, e.g. DEHP, DINP, and pentaBDE congeners, and trends in the use of plasticizers associated with the U.S. CPSIA of 2008. These results provide valuable information on the prevalence of plasticizers and flame retardants in crib mattresses, which can be used for policy decisions in the development of a new and modernized version of the Toxic Substances Control Act. The use of a used crib mattress (for lower VOC emissions) must be considered carefully, as older mattresses can contain high material-phase concentrations of DEHP and pentaBDE congeners.

- 7) Introducing the source-proximity effect of the sleep microenvironment and the underlying mechanisms contributing to elevated human exposures to mattress-released pollutants. Research related to “sleep IAQ” is growing, but insufficient to elucidate the impact of sleep exposures to indoor air pollutants on human health.
- 8) Presenting experimental wind tunnel and chamber methods and providing comprehensive literature reviews that offer a foundation for future research on indoor particle resuspension and pollutant transport in the sleep microenvironment.

## Appendix A

## Paper A. Monolayer and Multilayer Particle Deposits on Hard Surfaces: Literature Review and Implications for Particle Resuspension in the Indoor Environment

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### ABSTRACT

Particle deposits on indoor surfaces can be as complex and diverse as the indoor environments in which they exist. Dust loading can range over several orders of magnitude, suggesting the existence of different types of particle deposits. These deposits can be broadly classified as either a monolayer, in which particles are sparsely deposited on a surface, or a multilayer, in which particles are deposited on top of one another and there is particle-to-particle adhesion and interaction. Particles within these diverse structures of settled indoor dust can become airborne through a process known as resuspension, which can occur due to airflow in ventilation ducts or human activity indoors. The dust loading and deposit structure on an indoor surface may have important implications for resuspension in the indoor environment. This literature review provides a summary of dust loads found on indoor surfaces in field studies and classifies each dust load as either a monolayer or multilayer particle deposit. The paper highlights the unique attributes associated with resuspension from both types of particle deposits by summarizing key findings of the experimental resuspension literature. The fundamental differences in the resuspension process between monolayer and multilayer deposits suggests that resuspension may vary considerably among the broad spectrum of dust loads found on indoor surfaces.

## INTRODUCTION

Resuspension has been identified as an important secondary source of particles in the indoor environment. Resuspension from indoor particle deposits can occur due to airflow in ventilation ducts (e.g. Krauter and Biermann 2007, Wang et al. 2012) and human activities indoors (e.g. Thatcher and Layton 1995, Ferro et al. 2004, Qian and Ferro 2008, Tian et al. 2011, Shaughnessy and Vu 2012). Additionally, resuspension can be an exposure pathway to the multitude of pollutants that are commonly found in settled indoor dust, such as: allergens (e.g. O'Meara and Tovey 2000), lead, pesticides, phthalates, and flame retardants (e.g. Roberts et al. 2009).

Particle deposits in the indoor environment can be very complex and indoor dust loads can vary across several orders of magnitude. On the surfaces of ventilation ducts, dust loads can range from less than 1 g/m<sup>2</sup> to loads in excess of 100 g/m<sup>2</sup> (Nielson et al. 1990, Laatikainen et al. 1991, Pasanen et al. 1992, EPA 1996, Fortmann et al. 1997, Möritz et al. 2001, Kolari et al. 2005, and Lavoie et al. 2011). On hard flooring, such as vinyl, linoleum, and hardwood, dust loading is typically in the range of 0.1 to 1 g/m<sup>2</sup>, although lighter and heavier dust loads are commonly reported (e.g. Adgate et al. 1995, Rao et al. 2005, Johnson et al. 2009, Hoh et al. 2012). The wide range of dust loads suggests that there can exist different types of particle deposits on indoor surfaces, including both monolayer and multilayer deposits (Tovey and Ferro 2012). A monolayer deposit is one in which particles are sparsely deposited on a surface and there is minimal to no contact between them. A multilayer deposit is a porous structure of particles deposited on top of one another, forming multiple layers.

The diversity of dust loads and particle deposits may have important implications for particle resuspension from indoor surfaces. The resuspension literature has highlighted unique characteristics associated with resuspension from monolayer and multilayer particle deposits. For monolayer deposits, we are primarily interested in particle-to-surface attractive forces. Monolayer resuspension is strongly influenced by particle size and air velocity (Corn and Stein 1965, Wu et al. 1992, Nicholson 1993, Braaten 1994, Ibrahim et al. 2003, Jiang et al. 2008, Mukai et al. 2009, Goldasteh et al. 2012b), surface material and roughness (Wu et al. 1992, Nicholson 1993, Gomes et al. 2007, Jiang et al. 2008, Mukai et al. 2009, Goldasteh et al. 2012b, Kassab et al. 2013); particle composition (Wu et al. 1992, Braaten 1994, Ibrahim et al. 2003, Goldasteh et al. 2012a); characteristics of the airflow, such as acceleration (Wu et al. 1992, Nicholson 1993, Ibrahim et al. 2003), turbulence (Ibrahim et al. 2004, Mukai et al. 2009), exposure time (Ibrahim et al. 2003); and relative humidity and residence time (Ibrahim et al. 2004).

As we transition from a sparse monolayer to a complex multilayer deposit, additional parameters begin to influence resuspension, most notably particle-to-particle adhesion (Lazaridis and Drossinos 1998); layer location (Lazaridis and Drossinos 1998, Friess and Yadigaroglu 2001); aggregate formation and de-aggregation (Matsusaka and Masuda 1996, Kurkela et al. 2006, Gac et al. 2008, Gotoh et al. 2011); possible saltation effects (Bagnold 1941, Shao et al. 1993, Kok et al. 2012); dust loading (Fromentin 1989, Nitschke and Schmidt 2010); and the deposit's structure and porosity (Friess and Yadigaroglu 2002).

The primary aim of this literature review is to demonstrate the important role of the type of particle deposit on resuspension from indoor surfaces. The

paper begins with a comprehensive overview of dust loads reported in field studies in the literature and presents a simple scaling analysis to classify each dust load as either a monolayer or multilayer deposit. The paper then transitions to a discussion of the unique attributes of resuspension from both types of deposits based on findings in the experimental resuspension literature. The paper concludes with a discussion about the implications of different dust loads and particle deposits on resuspension from ventilation ducts and hard flooring.

#### INDOOR PARTICLE DEPOSIT CHARACTERIZATION

Numerous field studies have measured dust loads on a variety of surfaces in indoor environments. These studies are often aimed at investigating particle deposition, transport dynamics, and identifying pollutants in deposited dust. Table A1 presents a summary of dust loads from selected field studies (n = 29). In addition to field studies, Table A1 also presents dust loads from both wind tunnel (n = 29) and full-scale (n = 11) resuspension studies.

##### *Particle Deposit Classification*

For this literature review, a simplified, approximate method was developed to describe dust loads on hard, flat surfaces, including ventilation ducts and hard flooring (complex surfaces such as carpet are not considered here), for the field studies presented in Table A1. This scaling method applies the particle deposit structural analysis presented in Friess and Yadigaroglu (2002). They proposed a quantity called the layer number,  $\lambda$ , to distinguish between monolayer and multilayer deposits.  $\lambda$  represents the average number of

particles intersected on a line perpendicular to the wall, and is based on several physical parameters of the deposit:

$$\lambda = \frac{6 m_0}{\pi \rho D} \quad (\text{A.1})$$

where  $m_0$  is the dust load ( $\text{g}/\text{m}^2$ ),  $\rho$  is the particle density ( $\text{kg}/\text{m}^3$ ), and  $D$  is the mass median diameter of deposited particles ( $\mu\text{m}$ ).

Particle deposits are porous, and can also be defined by their porosities,  $\varepsilon$ , which represents the fraction of a deposit not occupied by particles. The structure, and therefore the porosity, of a deposit are determined by the deposition mechanism. Dense deposits formed by inertial impaction can be considered “cake-like” deposits, whereas those formed by gravitational settling result in “fluffy” deposits (as illustrated in Figure 1 of Friess and Yadigaroglu 2002). By combining  $\lambda$  and  $\varepsilon$ , this investigation proposes an approximation for the height,  $\delta$ , of the particle deposit:

$$\delta \sim \frac{\lambda}{(1-\varepsilon)} D \sim \frac{6 m_0}{\pi \rho (1-\varepsilon)} \quad (\text{A.2})$$

The height of the deposit increases with porosity, suggesting fluffy deposits formed by gravitational settling will be taller than those formed by impaction.  $\delta$  can be used as a basis to classify a particle deposit given its dust load.

To determine an approximate  $\delta$  for a dust load in the literature, the following assumptions were made when applying Equation A.2: homogenous porosity within the deposit; the particles are of unit particle density,  $1000 \text{ kg/m}^3$ ; deposits in ventilation ducts are formed by a combination of gravitational settling and inertial impaction due to convective airflow and turbulence; and deposits on indoor flooring are formed primarily by gravitational settling (for coarse particles in both cases). Based on the analysis presented in Friess and Yadigaroglu (2002), deposits in ventilation ducts are assumed to have porosities of approximately 0.50, and those on flooring are assumed to be approximately 0.75.

There is relatively little information on the size distribution of settled dust in different indoor environments. Furthermore, the distribution of a reported dust load can be influenced by the dust collection and sieving methods employed in a field study. Thus, it is difficult to estimate the mass median diameter for each dust load in Table A1. However, several studies offer some insight into the polydisperse size distribution, in the form of mass fractions and mass median diameter of settled indoor dust. Que Hee et al. (1985) found 18% of particles in dust to be  $< 44 \mu\text{m}$ , 58% in the fraction  $44\text{-}149 \mu\text{m}$ , and 24%  $> 149 \mu\text{m}$ . Seifert (1998) found a wide range of 0.3-24% for the fraction  $< 30 \mu\text{m}$  and a range of 6-35% for the fraction  $30\text{-}63 \mu\text{m}$ . Edwards et al. (1998) analyzed settled dust on deposition plates near the floor and found 99% of the particles to be  $< 50 \mu\text{m}$ . Lewis et al. (1999) found the mass median diameter of settled dust to be  $\sim 100 \mu\text{m}$ , with 25% of the mass less than  $25 \mu\text{m}$ , Rodes et al. (2001) found the mass median diameter to be  $\sim 60 \mu\text{m}$ , Wei et al. (2009) found the mass median diameter

to be  $\sim 75 \mu\text{m}$ , and Southey et al. (2011) found the mass median diameter to be  $\sim 90 \mu\text{m}$ . However, as discussed in Lewis et al. (1999) and Rodes et al. (2001), size distributions likely shift towards larger particles due to the agglomeration of smaller particles during the dust collection process and loss of particles  $< 10 \mu\text{m}$  in diameter due to adherence to vacuum collection bags. Given the variability in size distributions among these studies, and the potential for particle agglomeration during dust collection, two reference diameters were selected for the particle deposit classification: 10 and 100  $\mu\text{m}$ .

After determining  $\delta$  for a given dust load, it can be compared with the reference diameters to approximate the type of particle deposit. The classification criteria is as follows:

- if  $\delta \leq D$ : the deposit is a monolayer,
- if  $D < \delta < 2D$ : the deposit is an intermediate between a monolayer and multilayer,
- if  $\delta \geq 2D$ : the deposit is a multilayer.

There is inherent uncertainty in the assumptions used to estimate  $\delta$  and to describe each dust load in Table A1; however, the assumptions provide a good basis for classifying particle deposits given the limited data available on the structure and size distribution of deposits in different indoor environments.  $\delta$ , and thus the deposit classification, could change if  $\rho$ ,  $\varepsilon$ , and/or  $D$  were different from the values listed in the assumptions.

Table A1: Dust loads and particle deposit classification in selected field, full-scale laboratory, and wind tunnel studies.

Study	Description	Dust Load, $m_0$ (g/m <sup>2</sup> )	Particle Deposit Classification for Given Range of Dust		Surface(s)
			Loads <sup>2</sup>		
			Reference $D = 10 \mu\text{m}$	Reference $D = 100 \mu\text{m}$	
<b>Field Studies: Ventilation Ducts &amp; Hard Flooring</b>					
Nielsen et al. (1990)	Measured dust loading in office and school ventilation systems	Mean: 6.8, range: 1.1-50.9	Mono., Inter. & Multi. <sup>3</sup>	Mono. & Inter. <sup>3</sup>	Ventilation duct
Laatikainen et al. (1991)	Measured dust loading in office, school, and residential ventilation systems	Mean: 18.2, range: 3.6-140.8	Inter. & Multi. <sup>3</sup>	Mono., Inter. & Multi. <sup>3</sup>	Ventilation duct
Pasanen et al. (1992)	Examined the composition and location of settled dust in ventilation systems of public buildings	Mean: 10.6, range: 1.2-58.3	Mono., Inter. & Multi. <sup>3</sup>	Mono., Inter. & Multi. <sup>3</sup>	Ventilation duct
Auger (1994)	Measured dust loading in residential ventilation systems	Mean: 0.2, range: < detection limit (DL)-2.7	Mono. & Inter. <sup>3</sup>	Monolayer <sup>3</sup>	Ventilation duct
Kalliokoski et al. (1995)	Explored the impact of cleaning residential ventilation systems	Mean: 1.2, range: 0.2-3.9	Mono. & Inter. <sup>3</sup>	Monolayer <sup>3</sup>	Ventilation duct
Fransson et al. (1995)	Measured dust loading in supply ducts	Mean: 2.6, range: 1.9-3.0	Mono. & Inter. <sup>3</sup>	Monolayer <sup>3</sup>	Ventilation duct
Pasanen et al. (1995)	Measured dust loading in office ventilation systems	Mean: 13.2, range: 1.2-158	Mono., Inter. & Multi. <sup>3</sup>	Mono., Inter. & Multi. <sup>3</sup>	Ventilation duct
Adgate et al. (1995)	Measured lead in house dust in 216 homes	Bare floor mean: 0.42, range: 0.05-7	Mono., Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	Flooring: bare floor
Thatcher and Layton (1995)	Characterization of indoor particle dynamics in a residence	Tracked area of linoleum: 0.58	Monolayer <sup>3</sup>	Monolayer <sup>3</sup>	Flooring: linoleum

EPA (1996)	Measured lead concentrations in settled dust	Ventilation duct mean: 3.03, range: 0.054-1,385 Uncarpeted floor mean: 1.94, range: 0.005-155	Duct: Mono., Inter. & Multi. <sup>3</sup> Flooring: Duct: Mono., Inter. & Multi. <sup>3</sup>	Duct: Mono., Inter. & Multi. <sup>3</sup> Flooring: Duct: Mono., Inter. & Multi. <sup>3</sup>	Ventilation duct and uncarpeted flooring
Ito et al. (1996)	Studied particle deposition in ventilation systems	Mean: 7.5	Mono., Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	Ventilation duct
Fortmann et al. (1997)	Measured dust loading in residential ventilation systems	Mean: 6.4, range: 1.5-26	Mono., Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	Ventilation duct
Franke et al. (1997)	Assessed the effects of surface cleaning on indoor air quality	Routine and improved housekeeping mean: 0.08 Loading rates at 0.3 m above floor (per day):	Monolayer <sup>3</sup>	Monolayer <sup>3</sup>	Flooring: vinyl
Edwards et al. (1998)	Measured seasonal differences in dust loading rates in a home	Summer mean: 0.0042, winter mean: 0.0028 Assumed loading for 1 month: Summer mean: 0.126, winter mean: 0.084 Collection method 1 mean: 18.8, range: 4.0-131	Monolayer <sup>3</sup>	Monolayer <sup>3</sup>	Deposition plate: glass
Küchen (1998)	Measured dust loading in ventilation systems of public buildings	Collection method 2 mean: 7.0, range: 0.2-82 Collection method 3 mean: 1.9, range: <DL-21	Mono., Inter. & Multi. <sup>3</sup>	Mono., Inter. & Multi. <sup>3</sup>	Ventilation duct
Rich et al. (1999)	Measured dust and lead loading in residences	Floor (pre-clean) mean: 6.45 Floor (post-clean) mean: 4.64 Collection method 1 mean: 13.8, range: 3.1-52.7	Mono., Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	Flooring: linoleum, wood, stone, and tile
Möriz et al. (2001)	Measured dust loading using different collection methods	Collection method 2 mean: 8.6, range: 1.1-50.1 Collection method 3 mean: 5.6, range: 0.6-22.6	Mono., Inter. & Multi. <sup>3</sup>	Mono., Inter. & Multi. <sup>3</sup>	Ventilation duct

Holopainen et al. (2002)	Measured dust loading in recently installed ventilation systems	Collection method 4 mean: 6.9, range: 0.8-36.5 Collection method 5 mean: 2.1, range: 0.3-10.9 Cleanliness category 1 mean: 0.9, range: 0.4-2.9 Cleanliness category 2 mean: 2.3, range: 1.2-4.9	Mono. & Inter. <sup>3</sup>	Monolayer <sup>3</sup>	Ventilation duct
Kolari et al. (2005)	Measured dust loading in office ventilation systems	Before duct cleaning mean: 8.8, range: ~2-19	Mono., Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	Ventilation duct
Rao et al. (2005)	Measured dust loading and bioaerosols in hospitals	Mean: 1.0, range: 0.2-8.9	Mono., Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	Flooring: vinyl
Lewis et al. (2006)	Measured dust and lead loading on indoor surfaces	Vinyl mean: 4 Wood mean: 3	Mono., Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	Flooring: vinyl and wood
Johnson et al. (2009)	Measured dust loading in 488 homes	Mean: 0.325, range: 0.04-13.860	Mono., Inter. & Multi. <sup>3</sup>	Mono. & Inter. <sup>3</sup>	Flooring: smooth/hard
Layton and Beamer (2009)	Modeled transport of contaminated soil and airborne particles	Mean: 0.28, range: 0.04-6 (values referenced from NHEXAS study, Midwest residences)	Mono., Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	Flooring: flat, variety
Salares et al. (2009)	Studied the impact of cleaning on dust loading and levels of pollutants in dust	Small dust fraction (< 150 µm) range across cleaning intervals: 0.055-0.326	Monolayer <sup>3</sup>	Monolayer <sup>3</sup>	Flooring: hard
Khoder et al. (2010)	Measured dust loading rate in non-smoking and smoking residences	Loading rates (per week): Mean: 1.45, range: 0.49-3.77 Assumed loading for 1 month: Mean: 6.3, range: 2.1-16.4 Living room mean: 0.952, range: ~0.4-1.6	Mono., Inter. & Multi. <sup>3</sup>	Mono. & Inter. <sup>3</sup>	Deposition plate: glass
Raja et al. (2010)	Measured the resuspension of allergens from settled dust	Bedroom mean: 0.418, range: ~0.1-1.9 School classroom mean: 0.245, range: ~0.1-1.5	Mono. & Inter. <sup>3</sup>	Monolayer <sup>3</sup>	Flooring: not specified

Lavoie et al. (2011)	Evaluated a criteria for the initiation of duct cleaning in nonindustrial ventilation systems	Collection method 1 mean: 2.51, range: 0.012-28.14 Collection method 2 mean: 4.3, range: 0.014-33.38 Collection method 3 mean: 1.18, range: 0.002-15.58	Mono., Inter. & Multi. <sup>3</sup>	Mono. & Inter. <sup>3</sup>	Ventilation duct
Zuraimi et al. (2012)	Developed a protocol to assess duct cleaning impact in office buildings	Case study B1 pre-duct cleaning: 0.28 Case study B1 post-duct cleaning: 6.6	Mono., Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	Ventilation duct
Hoh et al. (2012)	Analyzed pollutants in household dust of smokers and non-smokers	Hard flooring median in homes of smokers and non-smokers: 1.07, range: ~0.1-10	Mono., Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	Flooring: wood, tile, and linoleum
<b>Wind Tunnel Studies: Various Hard Surfaces</b>					
Corn and Stein (1965)	Fundamental resuspension study	Mean seeding density <sup>1</sup> : ~ 0.14 particles/mm <sup>2</sup> 5.3 µm glass beads: 2.7 x 10 <sup>-5</sup> 10.6 µm glass beads: 2.1 x 10 <sup>-4</sup>	Monolayer <sup>4</sup>		Deposition plate: steel and glass
Fairchild and Tillery (1982)	Impact of saltating particles on resuspension	Not reported, monolayer mentioned explicitly Mean seeding density <sup>1</sup> : ~ 1000 particles/mm <sup>2</sup>	Monolayer <sup>4</sup>		Deposition plate: steel
Wen and Kasper (1989)	Resuspension model and experimental data for validation	0.412 µm latex particles: 4 x 10 <sup>-5</sup> 0.509 µm latex particles: 7 x 10 <sup>-5</sup> 1.019 µm latex particles: 6 x 10 <sup>-4</sup>	Monolayer <sup>4</sup>		Deposition plate: stainless steel
Fromentin (1989)	Time-dependence of multilayer resuspension	Range: 100-1000	Multilayer <sup>4</sup>		Deposition plate: stainless steel
Braaten et al. (1990)	Fundamental resuspension study and model validation	Not reported, monolayer mentioned explicitly Seeding density <sup>1</sup> range: 800-1200	Monolayer <sup>4</sup>		Deposition plate: glass
John et al. (1991)	Resuspension by impacting particles	8.6 µm ammonium fluorescein particles: 0.36-0.54 Note: authors mention 8-16% of particles	Monolayer <sup>4</sup>		Deposition plate: polyvinyl fluoride

		were touching one another			
Wu et al. (1992)	Examined particle bounceoff and resuspension mechanisms	50 to 100 particles/microscope field of view, monolayer mentioned explicitly	Monolayer <sup>4</sup>		Deposition plate: glass, plexiglas, and white oak leaves
Taheri and Bragg (1992)	Resuspension by turbulent flow	Not reported, monolayer deposit implied	Monolayer <sup>5</sup>		Deposition plate: glass
Nicholson (1993)	Resuspension from concrete and grass surfaces	Not reported, monolayer mentioned explicitly Mean seeding density <sup>1</sup> : ~ 5 particles/mm <sup>2</sup> 28 µm lycopodium spores: 0.07	Monolayer <sup>4</sup>		Deposition plate: concrete and grass
Braaten (1994)	Resuspension characteristics of particles 18 to 34 µm in diameter	34 µm timothy pollen: 0.11 30 µm microballoons: 0.08 20 µm glass spheres: 0.05 18 µm nickel spheres: 0.14	Monolayer <sup>4</sup>		Deposition plate: glass
Otani et al. (1995)	Resuspension by impinging jets	Not reported, monolayer deposit implied	Monolayer <sup>5</sup>		Deposition plate: glass and silicon
Matsusaka and Masuda (1996)	Aggregate resuspension from a fine powder layer	Not reported, multilayer mentioned explicitly	Multilayer <sup>4</sup>		Dust bed
Smedley et al. (1999)	Resuspension by impinging jets	Seeding density <sup>1</sup> : 300 particles/mm <sup>2</sup> 8.3 µm polystyrene particles: 0.10	Monolayer <sup>4</sup>		Deposition plate: glass
Loosmore and Hunt (2000)	Resuspension from a dust bed without saltation	Not reported, multilayer deposit implied	Multilayer <sup>6</sup>		Dust bed: acrylic
Adhiwidjaja et al. (2000)	Simultaneous deposition and resuspension of particles	Loading when deposition and resuspension are at an equilibrium state: 4.7 µm alumina powder: 22 5.6 µm alumina powder: 18	Multilayer <sup>3</sup>	Inter. <sup>3</sup>	Deposition plate: brass, copper, aluminum, and stainless steel
Reeks and Hall (2001)	Fundamental particle adhesion and resuspension study	Not reported, monolayer mentioned explicitly	Monolayer <sup>4</sup>		Deposition plate: stainless steel

Chiou and Tsai (2001)	Resuspension of road dust	Not reported, multilayer deposit implied	Multilayer <sup>6</sup>	Dust bed: aluminum
Ziskind et al. (2002)	Resuspension by pulsed air jets	Seeding density <sup>1</sup> range: ~ 2800-4500 particles/mm <sup>2</sup> 2-5 µm alumina silicate particles: ~ 0.3-0.5 Mean seeding density <sup>1</sup> : ~ 0.5 particles/mm <sup>2</sup> 30 µm lycopodium spores: 0.01 52 µm glass particles: 0.09 64 µm stainless steel spheres: 0.55 72 µm glass particles: 0.24 76 µm stainless steel spheres: 0.92 90 µm glass particles: 0.46 111 µm glass particles: 0.86 18-29.1 µm: 1095 10-18 µm: 210 5.6-10 µm: 35 3.2 to 5.6 µm: 27 1.8-3.2 µm: 25	Monolayer <sup>4</sup>	Deposition plate: glass and silicon
Ibrahim et al. (2003); Ibrahim et al. (2004); Ibrahim and Dunn (2006); Ibrahim et al. (2008)	Impact of airflow and particle deposition characteristics on resuspension		Monolayer <sup>4</sup>	Deposition plate: glass
Huang et al. (2005)	Reduction in road dust resuspension using a porous fence		Multilayer <sup>3</sup>	Deposition plate: aluminum cell
Miguel et al. (2005)	Deposition and resuspension from indoor surfaces	Not reported, monolayer deposit implied	Monolayer <sup>5</sup>	Flooring: metal plates coated with paper
Gomes et al. (2007)	Resuspension of allergens from a variety of indoor surfaces	Quartz particles on linoleum: 0.5 and 6.2 Roach dust particles on linoleum: 6.2	Monolayer <sup>7</sup> & Multilayer <sup>7</sup> Multilayer <sup>7</sup>	Flooring: linoleum
Jiang et al. (2008)	Impact of surface roughness on resuspension	Not reported, monolayer mentioned explicitly	Monolayer <sup>4</sup>	Deposition plate: stainless steel
Nitschke and Schmidt (2010)	Development of an experimental methodology to create reproducible particle layers	Range: 6.5-14	Multilayer <sup>3</sup>	Mono. & Inter. <sup>3</sup> Deposition plate: PMMA and steel

Goldasteh et al. (2012b)	Experimental modeling study of resuspension from flooring	Not reported, monolayer mentioned explicitly	Monolayer <sup>4</sup>		Flooring: hardwood and vinyl Deposition plate: hardwood, ceramic and glass
Kassab et al. (2013)	Experimental study of particle motion during resuspension	26.41 $\mu\text{m}$ glass particles: 0.10 36.24 $\mu\text{m}$ glass particles: 0.22 45.31 $\mu\text{m}$ glass particles: 0.24	Monolayer <sup>4</sup>		
<b>Full-Scale Laboratory Studies: Ventilation Ducts &amp; Hard Flooring</b>					
Karlsson et al. (1999)	Human-induced resuspension study	Seeding density <sup>2</sup> ~ 100 particles/ $\text{mm}^2$ 12 $\mu\text{m}$ <i>Bacillus Subtilis</i> spore clusters: 0.13	Monolayer <sup>4</sup>		Flooring: vinyl
Foarde and Menetrez (2002)	Impact of dust loading on antifungal sealants	Moderately soiled: 10 Heavily soiled: 100 Alumina particles, mean: 30	Multilayer <sup>3</sup>	Mono., Inter. & Multi. <sup>3</sup>	Ventilation duct
Hu et al. (2008)	Electrostatic detachment of particles from indoor surfaces	<i>Bacillus Thuringiensis</i> spores, mean: 10 Note: authors mention formation of particle aggregates	Inter. & Multi. <sup>4</sup>		Flooring: rubber and vinyl
Qian and Ferro (2008)	Walking-induced resuspension study	Mean: 20	Multilayer <sup>3</sup>	Inter. <sup>3</sup>	Flooring: vinyl
Kubota et al. (2009)	Walking-induced resuspension study	Mean: 56	Multilayer <sup>3</sup>	Multilayer <sup>3</sup>	Deposition plate: plexiglas Flooring: wood and vinyl
Tian et al. (2011)	Walking-induced resuspension study	House dust: 2 and 8	Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	
Gotoh et al. (2011)	Resuspension induced by an ascending flat object	Not reported, multilayer mentioned explicitly Note: authors mention formation of 2 and 8 mm thick powder layers of 26 $\mu\text{m}$ silica particles	Multilayer <sup>4</sup>		Deposition plate: flat disk
Shaughnessy and Vu (2012)	Walking-induced resuspension study	Test dust from occupied school classrooms: Mean: 18	Multilayer <sup>3</sup>	Inter. <sup>3</sup>	Flooring: vinyl
Wang et al. (2012)	Short-term resuspension in full-scale ventilation ducts	Polydisperse 0.25-10 $\mu\text{m}$ calcium salt particles, range: 18.34-21.06	Multilayer <sup>3,4</sup>		Ventilation duct

Hubbard et al. (2012)	Resuspension due to mechanical impulses	Note: authors mention formation of multiple layers Not reported, monolayer mentioned explicitly Note: authors mention surface coverage of particles was 5-10%.	Monolayer <sup>4</sup>		Deposition plate: titanium dioxide and silicon dioxide wafers
Kubota and Higuchi (2013)	Walking-induced resuspension study	Mean: 56	Multilayer <sup>3</sup>	Multilayer <sup>3</sup>	Deposition plate: smooth and hard surface

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Additional Studies: Ventilation Ducts

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Waring and Siegel (2008)	Modeled dust loading rates on various surfaces in ventilation systems	Residential supply duct median: 0.0051 g/month, range: ~0.0001-0.1 g/month Commercial supply duct median: 1.00 g/month, range: ~0.001-10 g/month Commercial return duct median: 0.262 g/month, range: ~0.001-1 g/month Note: authors did not normalize dust loading on duct surfaces by area Limit values for after & prior to duct cleaning:	N/A		Ventilation duct
Zhou et al. (2011) (and references therein)	Modeling of resuspension in ventilations systems before and after duct cleaning. Provides a list of dust load limit values in ventilation ducts in different countries.	China: after: 1, prior: 20 Japan: after: 1, prior: -- U.S.: after: 0.075, prior: -- Finland: after: 1-2.5, prior: -- Great Britain: after: 0.1, prior: 1-6	Mono., Inter. & Multi. <sup>3</sup>	Monolayer <sup>3</sup>	Ventilation duct

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<sup>1</sup>: Seeding density is defined as the number of particles deposited in a given area. The dust loading is calculated as the product of the seeding density and particle mass.

<sup>2</sup>: If a mass median diameter is mentioned explicitly in a reference, it was used in the deposit classification, as opposed to the reference diameters of 10 and 100 µm.

- 3: Approximate classification of deposit based upon particle classification method presented in our investigation. Dust loads on surfaces such as deposition plates are assumed to have porosities of 0.75, due to gravitational settling mechanisms (same as hard flooring surfaces).
- 4: Type of deposit mentioned explicitly in study.
- 5: Monolayer deposit implied in article due to the use of microscope particle counting methods to determine an absolute resuspension fraction.
- 6: Multilayer deposit implied in article due to the presence of a thick dust bed or powder layer.
- 7: Approximate classification of deposit based upon mass median diameter and density of particles used in study.

### *Monolayer and Multilayer Deposits: Field Studies*

The results of the classification analysis are presented in Table A1 for each field study. If a range of dust loads is reported, the possible range of particle deposits is presented. It is evident that monolayer, intermediate, and multilayer deposits can exist on the surfaces of ventilation ducts and hard flooring. As expected, the presence of an intermediate or multilayer deposit is more likely for a reference  $D$  of 10  $\mu\text{m}$  compared to 100  $\mu\text{m}$ . Regardless of the size distribution of a reported dust load, it is very likely we may see significant particle-to-particle contact in the heavier dust loads ( $> 5$  or  $10 \text{ g/m}^2$ ), in which the resuspension mechanisms would be more closely aligned with that of multilayer deposits, as compared to monolayer deposits, in which there is no particle-to-particle contact.

### *Ventilation Ducts*

As shown in Table A1, dust loading on the surfaces of ventilation ducts is highly variable, and dust loads greater than  $10 \text{ g/m}^2$  are common. Studies by Nielsen et al. (1990), Laatikainen et al. (1991), Pasanen et al. (1992), Pasanen et al. (1995), EPA (1996), Ito et al. (1996), Fortmann et al. (1997), Küchen (1998), Kolari et al. (2005), Lavoie et al. (2011), and Zuraimi et al (2012) have reported heavy dust loads representative of multilayer deposits, whereas studies by Auger (1994), Kalliokoski et al. (1995), Fransson et al. (1995), and Holopainen et al. (2002) have reported lighter dust loads suggestive of monolayer deposits. Dust loading in ventilation ducts can be influenced by many factors, such as characteristics of the building and its ventilation system, indoor particle sources, in-duct filtration, deposition mechanisms, and frequency of duct cleaning.

Therefore, based on the field studies as presented in Table A1, it would be expected that dust loading would vary considerably from any one building to the next, suggesting that deposits ranging from sparse monolayers to heavy multilayers are likely to exist.

### *Hard Flooring*

On hard flooring, such as linoleum, wood, and vinyl, lighter dust loads representative of monolayer deposits are frequently observed. Dust loads reported by Thatcher and Layton (1995), Franke et al. (1997), Salares et al. (2009), and Raja et al. (2010) were classified as monolayer deposits. Field studies by Adgate et al. (1995), Rich et al. (1999), Rao et al. (2005), Lewis et al. (2006), Johnson et al. (2009), Layton and Beamer (2009), and Hoh et al. (2011) reported ranges of dust loads that likely include both monolayer and multilayer deposits. The range of dust loads can be attributed to the frequency of floor cleaning or the number of particle sources indoors, among other factors.

For both reference particle diameters of 10 and 100  $\mu\text{m}$ , the classification analysis identified a greater prevalence of multilayer deposits in ventilation ducts than on hard flooring. However, the potential for both types of deposits on ventilation duct surfaces and hard flooring emphasizes the need to consider the role of the deposit characteristics when studying particle resuspension and transport in the indoor environment.

### *Monolayer and Multilayer Deposits: Wind Tunnel and Full-Scale Laboratory Studies*

Table A1 also presents the results of the classification analysis for selected wind tunnel and full-scale resuspension studies. Wind tunnel studies commonly report a seeding density. To convert to a dust load in g/m<sup>2</sup>, the seeding density (particles/m<sup>2</sup>) was multiplied by the particle's mass (g/particle). For some resuspension studies reported in Table A1, the type of deposit was mentioned explicitly in the article.

#### *Wind Tunnel Studies*

Although we see the existence of both monolayer and multilayer deposits indoors, the bulk of experimental wind tunnel studies have primarily focused on the former in order to develop a more fundamental understanding of the resuspension process and particle-to-deposition surface interactions and adhesion. Monolayer deposits were examined in studies on the aerodynamic resuspension of particles by Corn and Stein (1965), Wen and Kasper (1989), Braaten et al. (1990), John et al. (1991), Wu et al. (1992), Taheri and Bragg (1992), Nicholson (1993), Braaten (1994), Otani et al. (1995), Smedley et al. (1999), Reeks and Hall (2001), Ziskind et al. (2002), Ibrahim et al. (2003), Ibrahim et al. (2004), Miguel et al. (2005), Ibrahim and Dunn (2006), Ibrahim et al. (2008), Jiang et al. (2008), Goldasteh et al. (2012b), and Kassab et al. (2013). For these studies, dust loads are on the order of 10<sup>-5</sup> to 1 g/m<sup>2</sup>.

Only a few experimental wind tunnel studies have explored resuspension from multilayer deposits, including those by Fromentin (1989), Matsusaka and

Masuda (1996), Loosmore and Hunt (2000), Adhiwidjaja et al. (2000), Chiou and Tsai (2001), Huang et al. (2005), Gomes et al. (2007), Nitschke and Schmidt (2010). As shown in Table A1, only a few of these studies evaluated the impact of dust loading on resuspension, and none have systematically studied resuspension from both monolayer and multilayer deposits on indoor surfaces.

#### *Full-Scale Laboratory Studies*

Dust loads for full-scale resuspension studies, e.g. studies not conducted in small-scale wind tunnels, are also reported in Table A1. These studies are primarily aimed at exploring human-induced resuspension due to walking, resuspension in full-scale ventilation systems, or resuspension due to application of electrostatic or mechanical forces. Dust loads were found to be more representative of those found in the indoor environment. For example, Foarde and Menetrez (2002), Hu et al. (2008), Qian and Ferro (2008), Kubota et al. (2009), Shaughnessy and Vu (2012), Wang et al. (2012), and Kubota and Higuchi (2013) studied dust loads in excess of 10 g/m<sup>2</sup>, which were classified as multilayer deposits.

In summary, Table A1 provides a comprehensive overview of dust loads and particle deposits on the surfaces of ventilation ducts and hard flooring, along with the types of deposits examined in various resuspension studies. Collectively, these resuspension studies highlight important fundamental differences in the resuspension process between monolayer and multilayer deposits. It is important to understand these distinctions when studying particle

resuspension and transport in the indoor environment, where a wide range of deposit structures can be found.

## MONOLAYER DEPOSITS

The majority of experimental wind tunnel resuspension studies have focused on monolayer deposits. These studies, including many of those presented in Table A1, offer valuable insight into the key variables influencing aerodynamic resuspension from monolayer deposits, such as particle size and air velocity; surface material and roughness; shape and composition of the deposited particles; characteristics of the airflow, such as exposure time, acceleration, and turbulence; and relative humidity and residence time. These variables are likely to influence resuspension from the monolayer deposits that were identified in the indoor environmental field studies of Table A1.

### *Particle Size and Air Velocity*

As previously discussed, settled indoor dust can have wide particle size distribution, with particles  $< 10 \mu\text{m}$  to  $> 100 \mu\text{m}$  in diameter. Additionally, air velocities over indoor surfaces may vary considerably, from air bursts associated with human movement to airflow in ventilation systems. Wind tunnel resuspension studies, such as those by Corn and Stein (1965), Wu et al. (1992), Nicholson (1993), Braaten (1994), Ibrahim et al. (2003, 2004, 2008), Jiang et al. (2008), Mukai et al. (2009), among others, have demonstrated the important role of particle size and air velocity on resuspension from monolayer deposits.

Generally, the amount of particles that resuspend from a surface increases with increasing particle size and air velocity.

Of particular interest in the indoor environment is the resuspension of particles 2.5 to 10  $\mu\text{m}$  in diameter (coarse particles). Many monolayer studies have examined particles  $> 10 \mu\text{m}$  in diameter, e.g. Braaten (1994), Ibrahim et al. (2003, 2004, 2008), which more easily resuspend compared to their smaller counterparts, and only a few studies, e.g. Corn and Stein (1965), Jiang et al. (2008), Goldasteh et al. (2012b), have examined particles near 10  $\mu\text{m}$  in diameter. The results of the later demonstrate that very high velocities, unrealistic of what would be found in the indoor environment, are required to resuspend significant quantities of coarse particles. Corn and Stein (1965) did not observe resuspension for 10.6  $\mu\text{m}$  glass particles at 30, 60, and 90 m/s, and it was not until a velocity of 117 m/s that resuspension was reported, and Jiang et al. (2008) found that velocities greater than 50 m/s were necessary to resuspend 11  $\mu\text{m}$  poly(methyl methacrylate) (PMMA) particles. Additionally, a recent wind tunnel investigation by Goldasteh et al. (2012b) observed minimal resuspension for 1 to 10  $\mu\text{m}$  dust particles from linoleum flooring for velocities below 18 m/s (resuspension fraction remained below 0.10). It is apparent that phenomenally high velocities, often in excess of 25 m/s, are required to resuspend significant fractions of coarse particles from monolayer deposits due solely to the application of aerodynamic removal forces, e.g. lift and drag, via convective airflow in wind tunnels.

### *Surface Material and Roughness*

Given the diversity of surfaces that can be found indoors, from metal ventilation ducts to vinyl flooring, it is important to understand the impact of the deposition surface when studying resuspension from monolayer deposits. Various characteristics of the deposition surface material have been found to influence resuspension, such as the Hamaker constant between the particle and surface, surface electrostatic charge, and surface roughness. Many monolayer wind tunnel resuspension experiments have been conducted using glass deposition plates (see Table A1) and there are limited studies that have systematically investigated resuspension from different surfaces.

Wu et al. (1992) found that resuspension of lycopodium spores (30  $\mu\text{m}$ ) was significantly greater from glass compared to plexiglass due to the enhanced electrostatic adhesion of the plexiglass. Mukai et al. (2009) investigated resuspension of potassium chloride particles (1 to 20  $\mu\text{m}$ ) from two flat indoor surfaces, galvanized sheet metal and linoleum, and reported greater relative resuspension fractions for linoleum compared to sheet metal. Goldasteh et al. (2012b) found greater resuspension of dust particles to occur from vinyl flooring compared to hardwood flooring due to the greater contact area, and thus adhesion force, between the particles with hardwood. Jiang et al. (2008) investigated resuspension from stainless steel of varying surface roughness and found resuspension to increase with increasing submicron-scale surface roughness (from 0.01 to 0.3  $\mu\text{m}$ ), although micron-scale surface roughness (0.3 to 1.64  $\mu\text{m}$ ) had minimal impact. Lastly, Kassab et al. (2013) found surface material and roughness to influence particle motion and trajectories during the

resuspension process, with more rapid liftoff and minimal rolling/bouncing motion for surfaces with greater roughness, e.g. hardwood compared to glass.

### *Particle Composition and Shape*

In the indoor environment, there can exist a broad spectrum of particles, which vary in composition, surface features, shape, and density. The impact of particle composition has also been explored in monolayer wind tunnel resuspension studies. Wu et al. (1992) explored resuspension of uranine particles, polystyrene/divinylbenzene particles, lycopodium spores, and two types of pollen. Low resuspension was found for uranine and polystyrene/divinylbenzene particles. The wind tunnel experiments were performed at relative humidities in the range of 58 to 78%, and polystyrene particles have been found to plastically deform at relative humidities above 65%, resulting in enhanced adhesion and reduced resuspension (Cleaver and Looi 2007). Lycopodium spores were found to resuspend in significant fractions at very low velocities (4 to 8 m/s). Ibrahim et al. (2003) reported a similar trend, with threshold velocities for 30  $\mu\text{m}$  lycopodium spores roughly half of those as found for 32  $\mu\text{m}$  glass microspheres. Lycopodium spores are spheres with small bars along their surface, which significantly reduce their contact area and adhesion force with a surface (Nitschke and Schmidt 2009). Braaten (1994) also found the threshold velocity of 28  $\mu\text{m}$  lycopodium spores (8.73 m/s) to be slightly less than that for 34  $\mu\text{m}$  timothy pollen (12.57 m/s) and 30  $\mu\text{m}$  glass microballoons (9.72 m/s). Ibrahim et al. (2003) found resuspension of stainless steel microspheres to be greater than that of glass microspheres due to the

reduced adhesion between stainless steel and the glass deposition surface. Lastly, Goldasteh et al. (2012a) highlighted the impact of particle surface roughness and irregularity on resuspension in their monolayer modeling study.

#### *Airflow Characteristics: Exposure Time, Acceleration, and Turbulence*

Monolayer wind tunnel studies have highlighted the important role of various characteristics of the airflow on resuspension. The time a deposit is exposed to a controlled airflow in a wind tunnel is particularly important. Wu et al. (1992) confirmed the findings of Hall and Reed (1989) and found that two distinct temporal regimes exist during the resuspension process: a short period of less than one minute with very high resuspension, and an extended period of minimal resuspension. Nicholson (1993) investigated resuspension for exposure times of 10 to 3600 seconds, and found that almost half of the resuspended particles were removed in the first 10 seconds, with the resuspension rate decreasing by three orders of magnitude over 3600 seconds of exposure.

The acceleration of the airflow during the initial period of exposure is likely responsible for the enhanced resuspension. Ibrahim et al. (2003) also found that two distinct temporal regimes exist, a period of high resuspension during the acceleration of the airflow to the steady-state velocity, and a period of low resuspension during steady-state airflow. As discussed in Tadmor and Zur (1981), an additional aerodynamic removal force, known as the Basset force, can arise as the airflow is accelerated. Ibrahim et al. (2003) found that the

resuspension rate during the acceleration period ( $4.6 \text{ s}^{-1}$ ) is roughly six hundred times greater than during the steady-state period ( $0.0075 \text{ s}^{-1}$ ).

Mukai et al. (2009) examined the role of turbulence and found threshold velocities to decrease with increasing turbulence intensity of the airflow. The increased penetration of turbulent bursts into the viscous sublayer is likely responsible for enhanced particle resuspension at higher levels of turbulence (Cleaver and Yates 1973). Turbulent airflow is often associated with flow across joints in ventilation systems and air bursts generated by human movements such as walking, and is therefore an important variable to consider when studying particle resuspension.

#### *Relative Humidity and Residence Time*

Relative humidity has been found to influence the adhesion force between a particle and a deposition surface (e.g. Corn and Stein 1965, Hinds 1999, Paajanen et al. 2006, Cleaver and Looi 2007, You and Wan 2012). Thus, it is an important variable to consider when studying resuspension from monolayer deposits in the indoor environment. As demonstrated in Ibrahim et al. (2004), increasing both the relative humidity and the residence time over which a particle is deposited on a surface increased the velocity required to resuspend a particle. At 30% relative humidity, and a very short residence time (several hours), the threshold velocity for stainless steel microspheres (64 to 76  $\mu\text{m}$ ) was 4.2 m/s. For the same residence time, but at 61% relative humidity, the threshold velocity increased to 10.7 m/s. By increasing the residence time to 24 hours, the

threshold velocities were found to increase considerably. This suggests that the impact of relative humidity on the adhesion force, and therefore resuspension, is a time-dependent process. Some of wind tunnel studies presented in Table A1 exposed samples to the test flow conditions a short time after the particles were deposited (residence of several minutes to hours) (e.g. Nicholson 1993, Ibrahim et al. 2003). Based on the findings of Ibrahim et al. (2004), it is likely that the resuspension rates and fractions presented in these studies would decrease for longer residence times.

#### MULTILAYER DEPOSITS

As shown in Table A1, the majority of fundamental wind tunnel resuspension studies have focused on monolayer deposits. As such, the bulk of our knowledge on the resuspension process is derived from these studies, including the impact of the numerous parameters that were addressed in the preceding section. However, multilayer experimental and modeling studies have elucidated a few unique attributes of resuspension from multilayer deposits, including: the impact of the layer location, particle-to-particle adhesion, resuspension in the form of particle aggregates, impact of saltation, the relevance of the dust loading on a deposition surface, varying deposit porosity resulting from different deposition mechanisms, and time dependency of the resuspension flux.

### *Layer Location and Particle-to-Particle Adhesion*

An important variable for resuspension from multilayer deposits is the layer location. A multilayer modeling study by Lazaridis and Drossinos (1998) considered a two-layer deposit of spherical particles and found particles from the canopy layer to resuspend at lower velocities compared to particles along the surface layer. Additionally, a model proposed by Friess and Yadigaroglu (2001) found the resuspension flux at a given exposure time to increase with the layer number. For example, for 1 second of exposure, resuspension from the 100th layer was found to be approximately two orders of magnitude greater than resuspension from the surface layer.

The enhanced resuspension associated with the outermost layers, relative to the surface layer, may be explained by considering the varying magnitudes of adhesion forces within a deposit. Lazaridis and Drossinos (1998) demonstrated that the adhesion force between two spherical particles is less than that between a spherical particle and a flat surface. Similarly, they found that the interaction potential between two 10  $\mu\text{m}$  aluminum oxide particles to be approximately one-half of that between a particle and a flat, stainless steel deposition surface. Additionally, Zhu et al. (2012) highlighted the importance of considering the reduced adhesion forces between particles in multilayer deposits. They modified their adhesion force equation to account for the reduced Van der Waals forces between particles, compared to that between particles and a flat deposition surface. By accounting for particle-to-particle adhesion in their revised model, they found the velocity required to induce resuspension to decrease. For

multilayer deposits of many layers, the reduced particle-to-particle adhesion forces may result in greater resuspension relative to monolayer deposits, where there is only deposition surface adhesion. Lastly, given the elevated resuspension along the outermost layers relative to the surface layer, it would be expected that the characteristics of the underlying deposition surface might have less of an impact on resuspension from multilayer deposits compared to monolayer deposits, where only the surface layer of particles interact with the airflow.

#### *Aggregate Resuspension*

One unique characteristic of multilayer deposits is resuspension in the form of larger particle aggregates. Matsusaka and Masuda (1996) studied the resuspension of particle aggregates from multilayer deposits. They deposited a multilayer of 3  $\mu\text{m}$  fly ash particles and found resuspension to typically occur in small aggregates, with diameters ranging from 10 to 30  $\mu\text{m}$ . Additionally, a multilayer study by Gotoh et al. (2011) on the resuspension induced by an ascending circular plate found silica particles to resuspend in aggregates of similar size, regardless of their initial size. These experimental studies confirm the modeling study of Friess and Yadigaroglu (2002), who found the resuspended aggregates to be larger than the deposited particles and discussed that the tendency for particles to resuspend in aggregates is likely due to the lower aerodynamic removal forces that are necessary to resuspend a larger aggregate compared to smaller, individual particles.

Due to the large size of the resuspended aggregates, it may be expected that the particle aggregate will simply deposit back to the deposition surface from which it detached. However, as discussed in Gac et al. (2008), resuspended particle aggregates are often broken apart due to stresses imparted to the aggregate by turbulent eddies. By increasing turbulence in their wind tunnel, Gac et al. (2008) noticed a decrease in the size of the resuspended particles, suggesting enhanced de-aggregation of the resuspended aggregates due to higher levels of turbulence. Kurkela et al. (2006) also found particle de-aggregation to increase with increasing Reynolds number of the airflow. Therefore, once broken up, the smaller particles are more likely to be carried away with the airflow, rather than settle back to the surface. The de-aggregation process is an important consideration when studying particle resuspension and transport from multilayer deposits on indoor surfaces.

### *Saltation*

For multilayer deposits on indoor surfaces, which can contain particles on the order of 100  $\mu\text{m}$  in diameter (as discussed in the classification analysis section), saltation may play a role in resuspending smaller particles. Large particles or aggregates,  $\sim 100 \mu\text{m}$  in diameter, can be lifted away from a deposit by aerodynamic stresses. These particles are too large to remain airborne, so they return to the deposit and begin to hop along the surface and impact settled particles in a process known as saltation. Thus, a large saltating particle or aggregate can be responsible for the resuspension of smaller particles. Shao et al. (1993) demonstrated that impacts by saltating particles, as opposed to direct

aerodynamic resuspension, are the primary mechanism of resuspension for smaller particles from outdoor sand and dust. Additionally, Fairchild and Tillery (1982) found the resuspension flux of  $< 10 \mu\text{m}$  aluminum spheres to increase by factors of 1.33 and 2.3 when 100 and 200  $\mu\text{m}$  saltating particles, respectively, were injected in the upstream airflow. Lastly, resuspension may occur due to the fragmenting of saltating particle aggregates, that is, the breaking apart of the aggregate into smaller fragments as the aggregate impacts the surface (Kok et al. 2012).

#### *Dust Loading and Air Velocity*

Given the wide range of dust loads found in the field studies of Table A1, it is important to consider the impact of dust loading on resuspension from multilayer deposits. Gomes et al. (2007) studied resuspension from dust loads of 0.5 and 2.5  $\text{g}/\text{m}^2$  and observed more particles to resuspend at the higher dust loading. A wind tunnel study by Nitschke and Schmidt (2010) found resuspension to generally increase with dust loading. Between an exposure time of 3 and 8 seconds, the resuspension fraction increased as the dust load increased from 6.5 to 14  $\text{g}/\text{m}^2$  for both steel and PMMA deposition surfaces. Additionally, as with monolayer deposits, resuspension from multilayer deposits has been found to increase with increasing air velocity. Fromentin (1989) found a similar trend for heavy multilayer deposits of 100 to 1000  $\text{g}/\text{m}^2$  and observed a significant increase in the resuspension flux by increasing the bulk air velocity from 8.5 to 20  $\text{m}/\text{s}$ . Huang et al. (2005) and Matsusaka and Masuda (1996) observed similar trends in their respective wind tunnel studies.

### *Deposit Porosity*

Particles may form very complex structures in multilayer deposits (Friess and Yadigaroglu 2002, Henry et al. 2012). One parameter relating to the deposit structure that can influence resuspension is the porosity. As discussed in the deposit classification analysis, porosity is an important variable in approximating the height of a deposit, and is linked to the deposition mechanism. Based on the work of Friess and Yadigaroglu (2002), porosity might have a significant impact on resuspension. The authors found the resuspension fraction for a given exposure time to be nearly an order of magnitude greater for a multilayer deposit with a porosity of 0.76 (at 30 minutes, resuspension fraction of ~ 0.97) compared to one with a porosity of 0.45 (at 30 minutes, resuspension fraction of ~ 0.03). The “fluffy” nature of more porous deposits formed by gravitational settling likely results in enhanced resuspension compared to the compact nature of “cake-like” deposits formed by mechanisms such as inertial impaction. In denser deposits, particles are in contact with more particles, thereby increasing the total adhesion force acting on a particle.

### *Time-Dependence*

As with monolayer deposits, resuspension from multilayer deposits is a time-dependent phenomenon. Fromentin (1989) explored the time dependency of multilayer resuspension, finding that resuspension decreases with time at a rate of approximately  $1/\text{time}$ . The decay in resuspension from multilayer deposits may be explained by considering the enhanced resuspension along the

outermost layers relative to the surface layer. Studies by Chiou and Tsai (2001), Mortazavi (2005), and Wang et al. (2012) have observed that loosely adhered particles along the outermost layers would resuspend initially, sometimes in the form of a large puff of dust, leaving behind the strongly adhered particles along the surface. This phenomenon was also observed in the STORM experiments analyzed by Friess and Yadigaroglu (2002). The authors discussed that a large amount of “loose, fragile material” will resuspend initially, leaving behind a more “robust” particle deposit, from which resuspension does not occur at such a high rate. Lastly, Loosmore and Hunt (2000) found the resuspension from a multilayer deposit to approach a long-term, steady-state flux after some peak initial period.

#### IMPLICATIONS FOR RESUSPENSION IN THE INDOOR ENVIRONMENT

##### *Dust Loading on Indoor Surfaces*

A wide range of dust loads representing both monolayer and multilayer deposits can be found on indoor surfaces (Table A1). The resuspension literature has offered valuable insight into the fundamental differences associated with resuspension from both types of particle deposits. As such, it is expected that the source strength of resuspension, in the form of a resuspension fraction or rate, may vary considerably across the diverse dust loads found in the indoor environment. Given the enhanced resuspension that may occur from multilayer deposits due to the impact of particle-to-particle adhesion, aggregate resuspension, and possible saltation effects, we may expect to see a greater number of particles resuspend from heavier dust loads in excess of 5 or 10 g/m<sup>2</sup>,

as compared to very light loadings representing sparse monolayers. Based on the findings of several multilayer resuspension studies, we would also expect resuspension to increase with increasing dust load as more and more particles accumulate on an indoor surface. Additionally, for a given multilayer dust load, resuspension may vary due to differences in deposit structure and particle size distribution. Lastly, there may be considerable variability among monolayer deposits indoors. These deposits can contain a variety of particles of different sizes and composition and can exist on a variety of different surfaces at different environmental conditions. As discussed in the preceding sections, all of these parameters have been shown to influence resuspension from monolayer deposits.

### *Ventilation Ducts*

Resuspension from the surfaces of ventilation ducts is primarily associated with aerodynamic removal forces (e.g. Krauter and Biermann 2007, Wang et al. 2012). For most residential and commercial building applications, velocities in ventilation ducts are generally below 10 m/s. Wang et al. 2012 reported resuspension in a full-scale ventilation duct from multilayer dust loads in the range of 18 to 21 g/m<sup>2</sup> at velocities in the range of 3.8 to 8.8 m/s and Krauter and Biermann 2007 detected significant resuspension of spores (dust load not reported as a mass basis) in a full-scale ventilation system operating at a flow rate of 2.83 m<sup>3</sup>/min. In both studies, resuspension rates were found to reach peak values during transient operation of the ventilation system, e.g. initial period of airflow acceleration or periodic pulsations, and then decayed with

time. This further demonstrates the strong time-dependence of resuspension from both monolayer (Wu et al. 1992, Nicholson 1993) and multilayer (Fromentin 1989, Loosmore and Hunt 2000) deposits. Additionally, turbulent flow and complex flow regimes may develop over duct surfaces, such as irregularly shaped flex-duct, and in duct bends, which may play an important factor in particle resuspension (e.g. Mukai et al. 2009).

As shown in Table A1, dust loading on ventilation ducts is highly variable, and loading rates can range from ~0.0001 to 10 g/month (e.g. Waring and Siegel 2008). Given the possibility for greater resuspension from multilayer deposits compared to monolayer deposits, it may be desirable to prevent the accumulation of heavy dust loads in ventilation systems. Dust loading can be significantly reduced in ventilation ducts through proper duct cleaning techniques (Kolari et al. 2005, Zuraimi 2010), however, the process of duct cleaning itself may cause elevations in particle concentrations due to resuspension of the settled dust (e.g. Auger 1994, Zuraimi 2010) and dust loading may actually increase after ducts are cleaned (Zuraimi et al. 2012).

Zhou et al. (2011) reported limit values for dust loads in ducts in different countries, which are reference values for the maximum acceptable dust loading permitted on the duct surface (see Table A1). Many of these countries have limit values near 1 g/m<sup>2</sup>, which, based on the preceding particle classification analysis, would ensure the existence of a monolayer deposit. With annual dust loading rates in the range of less than 1 to as high as 5 g/m<sup>2</sup>year (Zuraimi 2010), ducts should be cleaned at least once a year to prevent the formation of

multilayer deposits. Zhou et al. (2011) considered the exposure implications for resuspension in ventilation ducts with varying dust loads. Cleaning a duct to reduce the dust load from 20 g/m<sup>2</sup> (multilayer) to 0.075 g/m<sup>2</sup> (monolayer) was found to significantly reduce particle inhalation exposure in a room by a factor of 267. This can be explained by the enhanced resuspension associated with multilayer deposits.

### *Hard Flooring*

Resuspension from hard flooring has been primarily associated with human walking. Full-scale walking-induced resuspension studies have reported resuspension from both monolayer (Karlsson et al. 1999, Tian et al. 2011) and multilayer deposits (Qian and Ferro 2008, Kubota et al. 2009, Tian et al. 2011, Shaughnessy and Vu 2012, Kubota and Higuchi 2012) on hard flooring. Gomes et al. (2007) found aerodynamic removal forces associated with airflow disturbances generated by human walking to be the primary mechanism for particle resuspension from flooring, although surface vibrations, mechanical abrasion, and electrostatic forces can contribute to resuspension (Gomes et al. 2007, Hu et al. 2008, Qian and Ferro 2008, Hubbard et al. 2012). Several studies investigated the airflow generated by foot motions. Kubota et al. (2009) and Kubota and Higuchi (2013) reported jet velocities of approximately 2-3 m/s associated with the downward foot motion, Gomes et al. (2007) reported peak air velocities of 1.5 to 2 m/s associated with walking-related airflow near the floor, and a modeling study by Zhang et al. (2008) found a maximum radial velocity of 18.3 m/s beneath the foot. Additionally, the airflows are likely very impulsive

with high acceleration (Khalifa and Elhadidi 2007), an important factor affecting resuspension (Ibrahim et al. 2003).

As shown in the field studies of Table A1, light dust loads, and thus monolayer deposits, are found to be more common on hard flooring compared to ventilation ducts. However, in cases where an occupant does not frequently clean their flooring, multilayer deposits may form, as may be the case for some of the higher dust loads reported in Table A1, including values reported by EPA (1996), Rich et al. (1999), Rao et al. (2005), Lewis et al. (2006), Johnson et al. (2009), and Hoh et al. (2012). As discussed in Franke et al. (1997), routine housekeeping and floor cleaning can prevent the accumulation of particles on hard flooring, maintaining dust loads below levels of 0.08 g/m<sup>2</sup>. Rich et al. (1999) also found floor cleaning to reduce dust loading. Given the elevated resuspension that may occur from multilayer deposits, it may be desirable to prevent the formation of heavy multilayer deposits on flooring.

Along with hard flooring, carpet is a common indoor flooring material, although it was not considered in this review and deposit classification analysis as the focus was on flat indoor surfaces. Higher dust loads are often reported for carpet compared to hard flooring, e.g. Chuang et al. (1995) (7.43-8.48 g/m<sup>2</sup>), Adgate et al. (1995) (0.3-99 g/m<sup>2</sup>), and Roberts et al. (2004) (0.7-21.1 g/m<sup>2</sup>, surface dust). However, higher dust loads may not necessarily lead to the formation of multilayer deposits, as the total surface area is much greater for carpet and particles may be distributed across the entire surface area of a fiber (e.g. Rosati et al. 2008). Qian and Ferro (2008) compared walking-induced resuspension

between carpet and hard flooring (vinyl) and found resuspension to be greater for carpet. Additionally, Mukai et al. (2009) observed greater levels of resuspension for carpet when compared to linoleum flooring.

## CONCLUSION

This literature review provided a comprehensive summary of dust loads on the surfaces of ventilation ducts and hard flooring reported in indoor environmental field studies and classified each dust load as either a monolayer or multilayer deposit. Dust loads on indoor surfaces were found to range over several orders of magnitude, representing both monolayer and multilayer deposits. Key findings from the experimental resuspension literature were summarized to highlight important differences in the resuspension mechanisms associated with both types of particle deposits. Resuspension from monolayer deposits can be influenced by numerous variables of relevance to the indoor environment, including characteristics of the deposited particles and deposition surface, airflow dynamics, and environmental conditions. The literature suggests that resuspension from multilayer deposits can be considerably different, and possibly be enhanced, compared to monolayer deposits. This is due to the effects of parameters unique to multilayer deposits, such as particle-to-particle adhesion forces, aggregate resuspension, saltating particles, deposit structure and porosity, and dust loading. Therefore, the type of particle deposit may have important implications for the resuspension and transport of particles from indoor surfaces, where a diversity of dust loads and particle deposits can be found. Future research efforts should aim at better characterizing the structure

and size distribution of settled indoor dust, along with developing a more comprehensive understanding of resuspension from real indoor particle deposits, with consideration for multilayer deposits on indoor surfaces, for which there is limited experimental data in the literature.

#### AUTHOR CONTRIBUTIONS

B.E.B. wrote the paper. J.A.S. and A.N. provided guidance for the structure of the literature review, advised in the interpretation of the literature, and provided detailed comments on draft manuscripts.

## Appendix B

## Paper B. Human Exposure to Indoor Air Pollutants in Sleep Microenvironments: A Literature Review

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In Preparation for Submission to *Atmospheric Environment*

### ABSTRACT

We spend approximately one-third of our lives sleeping, yet little is known as to how human exposure to indoor air pollutants in the sleep microenvironment impacts human health. This paper provides a comprehensive literature review of the current state-of-knowledge pertaining to human inhalation and dermal exposures while sleeping. An analysis of the duration of sleep exposure periods is provided, demonstrating that the sleep microenvironment is the predominant indoor space where humanity spends most of its time. Mattress dust is found to contain a diverse spectrum of biological matter and chemical contaminants and their concentrations in dust can span orders of magnitude from one bed to another. These dust particles can become airborne through human body movement-induced particle resuspension. Mattress foam and covers, pillows, and bed frames can emit a variety of volatile and semi-volatile organic compounds, including phthalate plasticizers and organophosphate flame retardants, and emission rates can increase due to localized elevations in surface temperature and moisture near the bed. This literature review demonstrates that human exposures to mattress-released pollutants can be amplified due to the source-proximity effect inherent to the sleep microenvironment, where the human body and breathing zone are in close and intimate contact with potential pollutant sources for prolonged periods.

Given the findings of this review, human exposures to indoor air pollutants in the sleep microenvironment should receive more attention and future research is needed to fully understand how sleep exposures affect human health and sleep quality, and how sleep exposures compare to exposures in other indoor spaces.

## INTRODUCTION

Many particulate and gaseous pollutants that are potentially health hazardous originate from indoor sources and concentrations are often much higher indoors than outdoors (Rudel and Perovich 2009). Since people spend about 90% of their time indoors (Klepeis et al. 2001), human exposures in the indoor environment are often substantially greater than exposures occurring outdoors. Adults sleep for approximately 8 hours per day, which corresponds to one-third of their lifetime, making sleep microenvironments particularly important in contributing to both their acute and chronic exposures to indoor pollutants. Mattresses, pillows, bedding materials, and bed frames are possible pollutant sources unique to sleep microenvironments. People are likely to be exposed to elevated concentrations of various chemical contaminants (e.g. volatile organic compounds and flame retardants) and biological material (e.g. bacteria, fungi, and mite allergens) since they are in close and intimate contact with these items. Early-life exposures to mattress-released pollutants are a particular concern, given the extended sleep periods of infants (12-14 hours per day) and their low body weights. (Boor et al. 2014).

The sleep microenvironment can be defined as the space encompassing a mattress, pillow, bedding materials, bed frame and the volume of air above these items that includes an individual's breathing zone (BZ) and buoyant thermal plume. The seemingly innocuous sleep microenvironment can be home to a diversity of pollutants that have been shown to impact human health. Mite and animal allergens, human- and animal-associated bacteria, fungi, and semi-volatile organic compounds (SVOCs) can accumulate in settled dust on mattresses, pillows, and bed sheets. The materials used to manufacture mattresses and bedding products, such as polyurethane foam and vinyl mattress covers, are possible sources of a myriad of chemical contaminants, including volatile organic compounds (VOCs), plasticizers, flame retardants, and unreacted (free) isocyanates (NCO).

Measuring indoor air pollutant concentrations at a reference location in the bulk air of a room may not be sufficient to characterize indoor exposures (e.g. Ferro et al. 2004). This discrepancy, which is known as the source-proximity effect, is mainly due to the non-uniform distribution of pollutants that arises due to source location, the buoyant human thermal plume, occupant movements, and the overall airflow pattern within a space (e.g. Rim and Novoselac 2009). Investigations into the source-proximity effect were motivated by early studies (e.g. U.S. EPA TEAM & PTEAM, Hartwell et al. 1992), which demonstrated that personal exposure, or BZ, concentrations of various particulate and gaseous pollutants are typically greater than those measured at stationary monitoring locations in the bulk indoor air by factors from 1 to > 2 (e.g. Rodes et al. 1991, Hartwell et al. 1992, Clayton et al. 1993, Edwards et al. 2001, Rodes et al. 2001).

The source-proximity effect is an important characteristic of the sleep microenvironment, where the human body and BZ are in close proximity to potential pollutant sources for extended periods of time. Therefore, people may be exposed to elevated concentrations of pollutants via inhalation and dermal pathways as they sleep. Research on the underlying physical processes governing the source-proximity effect of the sleep microenvironment is limited.

Human exposure in sleep microenvironments is important, but has not been extensively researched with respect to exposures in other types of indoor environments, such as classrooms, kitchens, and occupational workplaces. Therefore, it offers challenging research opportunities to advance our understanding of the pollutants commonly found in sleep microenvironments, the mechanisms by which pollutants are transported around the human body to an individual's BZ, pollutant concentrations and exposure levels that individuals experience while sleeping, and the total amount of pollutants that are inhaled or absorbed via dermal exposure. This knowledge is important to investigate subsequent health effects and to develop strategies to promote healthy bedrooms. As a key step in the advancement of this knowledge, we provide a comprehensive literature review of the current understanding of human exposure in sleep microenvironments, including: exposure characteristics, a summary of the biological and chemical composition of mattress dust, an overview of chemical emissions from mattresses and bedding materials, and the potential for elevated exposures due to the source-proximity effect.

## EXPOSURE CHARACTERISTICS

### *Exposure Pathways*

Inhalation exposure can be defined as the contact between an agent (pollutant) and a target (person) (Zartarian et al. 1997). The contact between a pollutant and a person occurs at an exposure point, which we can define as a person's BZ. The inhalation exposure concentration can be defined as the mass of a pollutant (e.g. # of particles or  $\mu\text{g}$  of a VOC) in a person's BZ divided by the BZ volume. The volume of the BZ is variable, has been shown to be dependent on the transport dynamics of a specific pollutant (e.g. Lidén and Waher 2010), and is likely influenced by physical obstructions, such as a pillow for a person sleeping in the prone position (Laverge et al. 2015). However, it is generally defined as a hemisphere with a radius of 30 to 50 cm, with its center of origin at the mouth or the nose (Jensen and O'Brien 1993, Rodes 2011). An expression for the time-integrated inhalation exposure,  $E_{i,j}$ , of a specific pollutant,  $i$ , in a particular microenvironment,  $j$ , is:

$$E_{i,j} = \int_{t_0}^t C_{BZ,i,j}(t) dt \quad (\text{B.1})$$

where  $C_{BZ,i,j}(t)$  is the BZ concentration at time  $t$ , although time-averaged BZ concentrations are commonly reported. Two key variables thus define inhalation exposures: the amount of pollutant that enters the BZ and the length of time over which continuous contact occurs between the pollutant and the BZ.

In addition to inhalation exposure assessment, it is necessary to evaluate transdermal uptake of chemical pollutants (e.g. SVOCs) in the sleep microenvironment, given the extended periods of contact between human skin and vinyl mattress covers, pillow protectors, and assorted bedding materials (bed sheets, blankets, duvet covers). Dermal exposures while sleeping can occur via both direct contact transfer with these items and air-to-skin uptake (Weschler and Nazaroff 2012, 2013), the latter of which is linked to concentrations of gas-phase pollutants released from the mattress, which are much greater near the mattress surface compared to the bulk bedroom air. Dermal exposure via direct contact transfer ( $E_{Dermal\ Contact}$ ) can be calculated using Equation B.2:

$$E_{Dermal\ Contact} = C_s \cdot SA \cdot TE \cdot AF \quad (B.2)$$

where  $C_s$  is the surface concentration of a pollutant,  $SA$  is the contact area,  $TE$  is the efficiency of contact transfer, which represents the fraction of the surface pollutant that is transferred to the skin during a contact event, and  $AF$  is the fractional absorption factor. Although contact transfer has been well studied, numerical values of  $TE$  are difficult to measure accurately and thus remain poorly characterized. The value of  $TE$  can be significantly influenced by the chemical compound, physical nature of the surfaces, exposure period, contact pressure and motion, concentration on the surface, and temperature and humidity (e.g. Cohen Hubal et al. 2008).  $AF$  is typically an empirical quantity that is assumed to be a fixed value for a specific chemical regardless of exposure conditions. The value of  $AF$  is commonly determined from in vitro or in vivo dermal absorption studies. Although the  $AF$  approach has been widely adopted,

Frasch et al. (2014) discussed its potential limitations and concluded that loading, evaporation or sublimation of VOCs, and experimental duration, may have strong impacts on the fraction absorbed by skin.

Dermal exposure due to direct air-to-skin transport has received attention recently (Weschler and Nazaroff 2008, 2012, 2013, Xu et al. 2010). The transdermal uptake of a gas-phase pollutant ( $E_{Dermal\ Gas}$ ) can be estimated using Equation B.3:

$$E_{Dermal\ Gas} = C_{gas} \cdot k_{p-g} \cdot BSA \quad (B.3)$$

where  $C_{gas}$  is the gas-phase concentration of a pollutant in proximity to human body,  $k_{p-g}$  is the transdermal permeability coefficient, and  $BSA$  is the body surface area. Weschler and Nazaroff (2012 and 2013) demonstrated that the abundance of SVOCs on hand wipe samples can be predicted reasonably well from gas-phase concentrations this expression. They developed equations to estimate  $k_{p-g}$  based on the knowledge of physical and chemical properties of indoor pollutants, such as molecular weight ( $MW$ ), octanol-water partition coefficient ( $K_{ow}$ ), and Henry's constant, and extended their analysis of transdermal penetration to approximately eighty VOC and SVOC compounds.

### *Exposure Period*

The exposure period can be defined as the time of continuous contact between a pollutant and a person (Zartarian et al. 1997). It is the magnitude of

this period that makes the sleep microenvironment particularly important in contributing to human inhalation and dermal exposures to various pollutants originating in mattresses, pillows, and bedding.

Humans spend a considerable amount of time sleeping. The U.S. Environmental Protection Agency (EPA) provides data on the duration of time spent (hours/day) in a sleep or nap activity in the Exposure Factors Handbook (EFH) (EPA 2009), which is categorized by both age group and gender. To investigate the dependence of sleep duration on age, we plotted the EFH's data, as shown in Figure B1. During the first few months to years of life, infants (<1 year of age) and toddlers (1-3 years of age) spend a considerable amount of time sleeping, with an average of 13.3 hours/day in the first year of life, 12.6 hours/day in the second year, and 12.1 hours/day in the third year. In addition, other researchers (Diez et al. 2000, Iglowstein et al. 2003) have also reported durations of sleep greater than 14 hours/day during infancy. The data suggests that sleep microenvironments may play a critical role in characterizing exposures of very young children to indoor air pollutants. As expected, sleep duration continues to decline with age through much of adulthood, with an average of 8.2 hours/day for the U.S. mean age group of 37 years (U.S. Census Bureau 2009). An inflection is observed around 50 years of age, after which, sleep duration begins to steadily rise with age to an average of 9.2 hours/day for people older than 81 years of age.

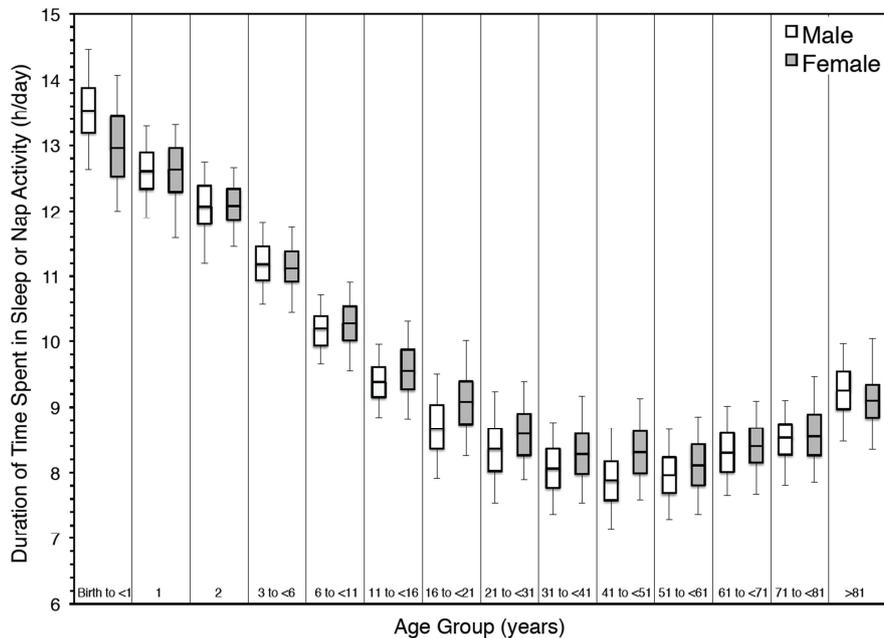


Figure B1: Duration of time (hours/day) spent in sleep or nap activity (EPA 2009, data ID = 14500). Box plots represent interquartile range and whiskers represent the 5th and 95th percentiles.

For adults, it is also constructive to evaluate how the time spent in the sleep microenvironment compares to other microenvironments in which people commonly spend their time. The National Human Activity Pattern Survey (NHAPS) Study conducted by Klepeis et al. (2001) surveyed over 9,000 adults (> 18 years of age) in the U.S. on their activity patterns. Respondents were asked to keep 24-hour diaries documenting their activities and the location where the activity occurred. Klepeis et al. (2001) reduced the survey data to six general locations: residence, office-factory, bar-restaurant, other indoor locations, enclosed vehicle, and outdoors, and reported the mean percentage of time spent in each location, finding that Americans spent nearly 87% of their time indoors.

The NHAPS data can be used along with the EFH data set to estimate the average percentage of time an adult in the U.S. spends in sleep microenvironments. We assumed that the time spent in the sleep or nap activity (mean of 8.2 hours for adult of mean age in the U.S.), as defined by the U.S. EPA, is spent entirely in a person's bedroom, on their mattress or similar bedding, and in their personal residence. As shown in Figure B2, the modified activity pattern data shows us that adults spend about 34% of their day in the sleep microenvironment, which equates to about 50% of the time they spend in a residence, and 39% of the time they spend indoors. Assuming the "in a residence, other activity" location category can be partitioned among several different microenvironments, such as living areas, kitchen, office, and bathroom, the sleep microenvironment becomes the predominant indoor space where American adults spend most of their time. Furthermore, the percentage of time spent in sleep microenvironments is likely an underestimate, given that the U.S. EPA EFH data is limited to time spent sleeping or napping, and does not include other activities common to this indoor space (e.g. reading a book or watching television in bed).

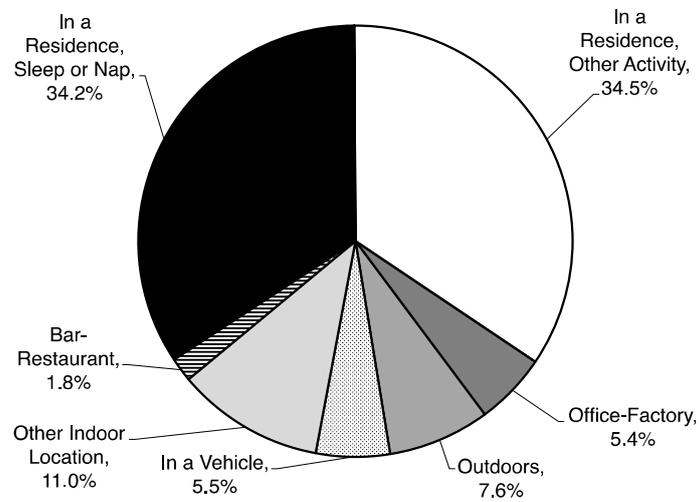


Figure B2: Fraction of time adults spent sleeping or napping in a residence (U.S. EPA 2009, Klepeis et al. 2001).

#### *Exposure of Infants and Toddlers: The Impact of Body Weight*

Because of their low body weight, infants and toddlers inhale considerably more air per kg of body weight as they sleep compared to adolescents and adults. The volume of air inhaled/kg-day can be estimated with the U.S. EPA EFH data set by taking the product of the mean normalized volumetric breathing rate in the sleep or nap activity (L/h-kg) and the mean duration of time spent in the sleep or nap activity (h/day). The normalized inhaled air volumes,  $V^*_{Sleep}$ , are categorized by age group and gender and presented in Figure B3. It is apparent that infants and toddlers inhale nearly an order of magnitude more air per body mass than adults. During the first and second years of life, they inhale approximately 300 L/kg-day, which decreases to about 244 L/kg-day during the third year of life.  $V^*_{Sleep}$  continues to decrease through childhood, adolescence, and adulthood. Between 21 and < 31 years of

age, adults breath about 30 L/kg-day. There appears to be no significant differences in  $V_{Sleep}^*$  between males and females.

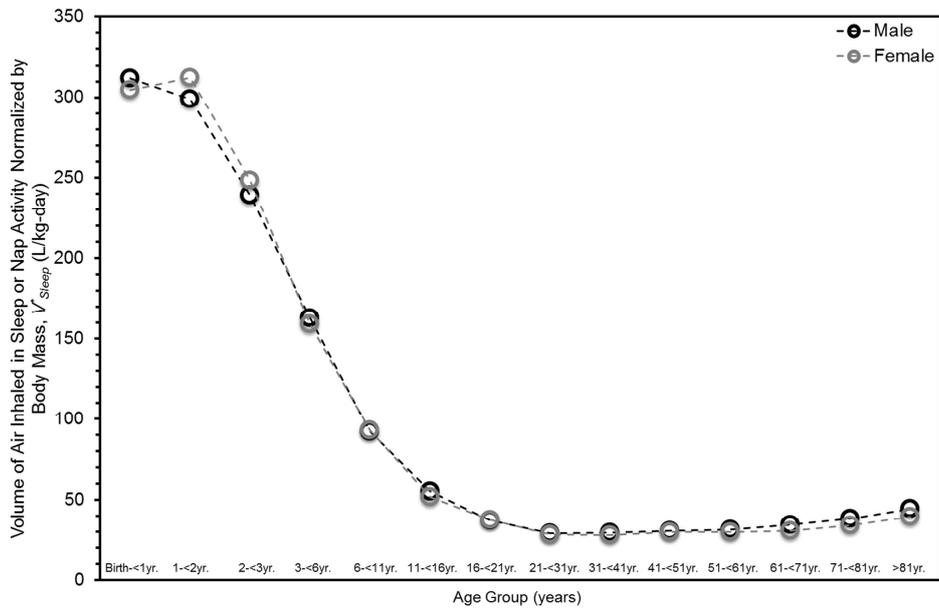


Figure B3: Volume of air inhaled during sleep or nap activity per day, normalized by body mass for each age group and gender (calculated using U.S. EPA EFH data set, U.S. EPA 2009).

The analysis above provides a crucial key to understanding how the dose of an inhaled pollutant is considerably different between very young children and adults. The daily inhalation intake dose,  $D_{i,j}$ , (mass of pollutant inhaled per body weight per day) of pollutant  $i$  in microenvironment  $j$  can be calculated as:

$$D_{i,j} = \frac{\int_{t_0}^t Q_{B,j}(t) \times C_{BZ,i,j}(t) dt}{BW} \quad (B.4)$$

where  $Q_{B,j}(t)$  is the volumetric breathing rate (L/h) and  $BW$  is body weight (kg). This expression can be simplified for the sleep microenvironment and by assuming time-averaged BZ concentrations and breathing rates. Additionally, the normalized inhaled air volumes during a sleep period,  $V^*_{Sleep,i}$ , can be substituted into the equation to obtain:

$$\overline{D}_{i,Sleep} \cong \frac{\overline{Q}_{B,Sleep} \times \overline{t}_{Sleep} \times \overline{C}_{BZ,i,Sleep}}{BW} \cong V^*_{Sleep,i} \times \overline{C}_{BZ,i,Sleep} \quad (B.5)$$

Thus, to estimate the daily sleeping dose, one can simply take the product of  $V^*_{Sleep,i}$ , as calculated from the U.S. EPA EFH data set, and the average BZ concentration of a pollutant originating in the sleep microenvironment. Therefore, if a very young child (an infant or toddler) and an adult are exposed to the same BZ concentration of a pollutant released from a mattress, the child-normalized dose will be an order of magnitude greater than that of the adult. Similarly, to compare an infant's dose of dermally absorbed pollutants to that of an adult, we can use the skin surface area to body mass ratio ( $BSA/BW$ ), as presented in Figure B4. Infants have  $BSA/BW$  three times greater than adults, emphasizing the importance of early-life dermal exposures.

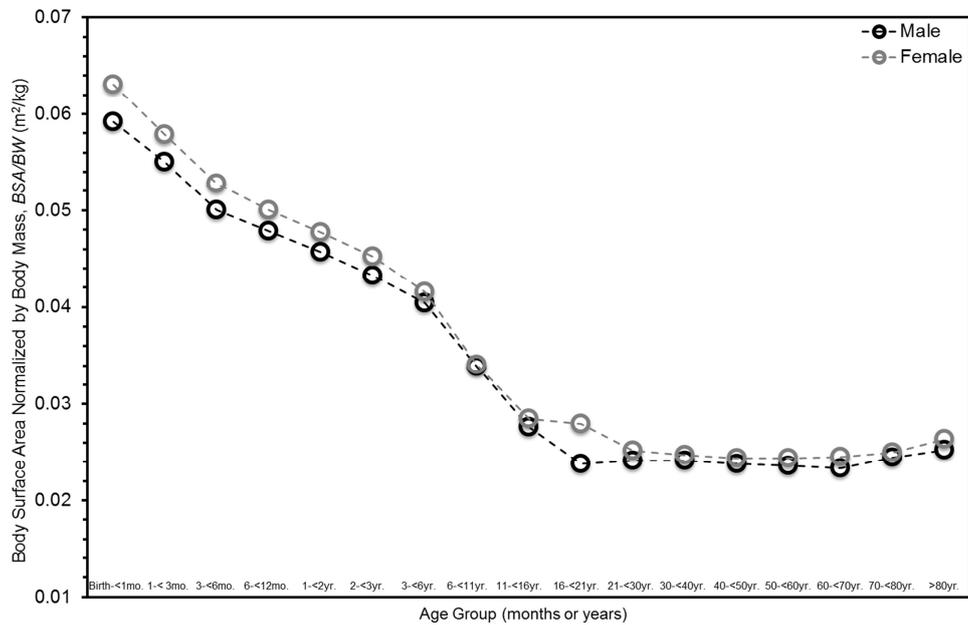


Figure B4: Infant dermal exposure dose considerations in the sleep microenvironment: body surface area normalized by body mass ( $BSA/BW$ ,  $m^2/kg$ ) for each age group and gender (calculated using U.S. EPA EFH data set, U.S. EPA 2011).

#### BIOLOGICAL AND CHEMICAL COMPOSITION OF MATTRESS DUST

Mattresses, pillows, and bedding materials serve as an accumulation zone for a diverse spectrum of pollutants, many of which are of biological origin. Table B1 provides an overview of pollutants identified in mattress dust in selected field studies ( $n = 43$ ) and their corresponding dust concentrations. Many of these studies are aimed at identifying associations between levels of allergens, fungi, and bacteria in mattress dust with allergies, asthma, wheeze, and other respiratory problems in infants, children, and adults. Mattress dust is commonly sampled as a surrogate for human exposure in large cohort studies. However, air sampling, particularly near the BZ, may provide valuable insight in

characterizing sleep exposures (e.g. Sakaguchi et al. 1992, Mahmic et al. 1998, Gore et al. 2002, Rabinovitch et al. 2005), as dust levels are not always correlated with concentrations in the air.

### *House Dust Mite and Animal Allergens*

Biological matter in mattress dust is comprised of a wide range of organisms and their associated allergens. Among house dust mite (HDM) allergens, Der p 1, Der f 1, Der p2, and Blo t 5, have been identified in many regions of the world. The high moisture levels and plentiful supply of skin flakes makes the sleep microenvironment an ideal environment for HDMs to flourish. For example, in Brazil, da Silva et al. (2005) detected 400-1,000 HDM bodies (eggs, larvae, nymphs, and adults) per gram of mattress dust. Additionally, cat (Fel d 1), dog (Can f 1), cockroach (Bla g 2), and mouse/rat urinary allergens have also been detected, along with allergens originating outdoors (Ph1 p 5, timothy pollen). Concentrations of these allergens can range over several orders of magnitude, from  $< 1$  to  $> 10^3$   $\mu\text{g/g}$ , as shown in Table B1 (levels are also reported as  $\mu\text{g/m}^2$  mattress area sampled).

The type of mattress and bedding materials has been shown to influence levels of HDM allergens. Van den Bemt et al. (2006) reported higher levels on inner spring mattresses, compared to polyester and latex, and waterbeds. In Thailand, Visitsunthorn et al. (2010) found varying levels Der p 1 and Der f 1 in different mattresses, with kapok showing highest concentrations, followed (in decreasing order) by synthetic, coconut, and polyurethane sponge. Der p 1 levels

also tend to increase with mattress age (Mihirshahi et al. 2002 and van den Bemt et al. 2006). In addition, the frequent use of wool, synthetic, and sheepskin bedding materials was found to be associated with higher levels of Der p 1 allergens (Garrett et al. 1998 and Mihirshahi et al. 2002); and pillow dust can also be a source of Der p 1, Der f 1, and Blo t 5 allergens (Wu et al. 2009). Finally, differences in HDM allergen levels have also been observed between the mattresses of mothers and their infants (Chen et al. 2007), between rural and urban residences (El Sharif et al. 2004), before and after home renovation (Hirsh et al. 2000), as well as variations among the seasons (Su et al. 2001).

Additional studies have reported elevated levels of airborne HDM allergens during sleep periods, suggesting that deposited HDM particles are released into the air via human-induced resuspension (Spilak et al. 2014, Boor et al. 2015a). Gore et al. (2002) measured airborne Der p 1 allergen exposure via nasal air sampling and estimated inhalation exposures of 6 ng/night and 2 µg/year during sleep periods. Sakaguchi et al. (1992) reported airborne Der p 1/Der f 1 and Der 2 concentrations of 223 and 87.1 pg/m<sup>3</sup>, respectively, in the air directly above a mattress. These levels were found to be a factor of ten greater than those measured in the living room air of the same residence, which may suggest significant resuspension of settled HDM particles from mattresses.

### *Fungi*

A wide variety of fungal genera and species are commonly identified in mattress dust, including *Penicillium* spp., *Cladosporium* spp., *Aspergillus* spp.,

*Aspergillus fumigatus* spp., *Alternaria* spp., *Eurotium* spp., *Epicoccum*, and yeasts, among many others. Along with dust, many fungal species have been found within mattress and pillow foam, with early studies by Conant et al. (1936) and Cooke and Foter (1958) recognizing the impact of fungal contamination of pillows on human health. Concentrations of fungal species are reported as CFU (colony forming units) per g of mattress dust, and are typically in the range of  $10^3$  to  $10^4$  CFU/g. Mattresses and pillows in the sleep microenvironment are an ideal fungal culture medium, given the high moisture levels caused by human sweating (~ 100 liters of sweat per year) and elevated temperatures (~ > 30°C) (Woodcock et al. 2006). Fungi may consume HDM fecal matter and human skin flakes in mattress dust, providing them with necessary nutrients; and subsequently, HDMs may consume fungi, leading to a “miniature ecosystem” within mattresses and pillows (Kort et al. 1997; Woodcock et al. 2006). Studies also report levels of (1-3)- $\beta$ -Glucan, a component of the cell wall of many fungi (also found in some bacteria) and a common microbial marker used to assess fungal exposure. Typical concentrations are in the range of 10 to  $10^3$   $\mu$ g/g of mattress dust.

Similar fungal species have been detected in mattress dust throughout the world, including the United Kingdom (Woodcock et al. 2005), Northern California, U.S. (Hicks et al. 2005), Germany (Hirsh et al. 2000, Pitten et al. 2001, Jovanovic et al. 2004, Vogel et al. 2008), and Australia (Begum et al. 2012). Giovannangelo et al. (2007), Tischer et al. (2011), and Casas et al. (2013) found small differences in (1-3)- $\beta$ -Glucan levels in mattress dust in large cohort studies across several European countries. Additionally, the fungal composition of

mattress dust can vary with bedding material, as observed by Pitten et al. (2001) for dust on cotton and synthetic mattress encasings. Higher concentrations of *Aspergillus fumigatus* and (1-3)- $\beta$ -Glucans have been reported in synthetic compared to feather pillows (Woodcock et al. 2005, Heyes et al. 2009).

### *Bacteria*

The sleep microenvironment is also home to an array of bacterial phyla and genera, many of which are associated with human origins (skin, oral, intestinal/fecal, and genital) and specifically, the shedding of human skin (e.g. Täubel et al. 2009, Hospodsky et al. 2012). Bacteria identified in mattress dust include: *Staphylococcus*, *Lactobacillus*, *Streptococcus* sp., *Lactococcus*, *Bacillus* sp., *Listeria* spp., *Zoogloea* sp., *Duganella* sp., *Aurantimonas ureolytica* sp., *Serratia marcescens*, *Pseudomonas fluorescens*, *Corynebacterium tuberculostearicum*, *Gardnerella vaginalis*, *Lactobacillus curvatus*, *Moraxella* sp., *Staphylococcus sciuri* sp., *Jeotgalicoccus* sp., *Macrococcus brunensis*, *Aerobacter ureolytica*, *P. fluorescens*, *Triatoma infestans*, and *Neisseria meningitidis*, among a host of others. In addition to identifying specific bacteria, endotoxin concentrations are also commonly reported in mattress dust. Endotoxin is the biologically active lipopolysaccharide (LPS) of gram-negative bacteria and is a common marker to assess bacterial exposure (although it only represents defined subpopulations of bacteria and does not differentiate between human and non-human sources) (Täubel et al. 2009). Endotoxin levels are reported as endotoxin units (EU) per gram of mattress dust, and concentrations are typically in the range of  $10^3$  to  $10^5$  EU/g (levels are also reported as EU/m<sup>2</sup> mattress area sampled).

Recent studies applying next generation DNA sequencing have offered new insights about the bacterial communities in mattress dust. Täubel et al. (2009) and Ege et al. (2012) provide examples of the different bacterial phyla and genera identified in both Finland and Germany, respectively. In addition, greater levels of bacterial endotoxin loading have been found in infant mattresses, compared to those of their mothers (Gehring et al. 2004, Chen et al. 2007). Furthermore, Doyen et al. (2011) found endotoxin levels to increase in infants' bedrooms over the first 6 months of life. As with fungal (1-3)- $\beta$ -Glucans, small variability in endotoxin levels have been found among large cohort studies in Europe (Tischer et al. 2011, Casas et al. 2013).

#### *Plasticizers and Flame Retardants*

Due to their low volatility and high molecular weight, semi-volatile organic compounds (SVOCs) tend to accumulate in house dust (e.g. Hwang et al. 2008, Weschler and Nazaroff 2008). Although many field studies have measured the concentrations of various SVOCs in settled dust on flooring, only a few have explicitly reported levels in mattress dust. Hsu et al. (2012) detected several phthalate plasticizers in mattress dust in Taiwan, including DEHP, BBzP, and DBP (nomenclature listed in Table B1). Concentrations are on the order of 10 to 10<sup>2</sup>  $\mu\text{g/g}$  of mattress dust, with higher levels reported in the presence of visible mold on the mattress. In New Zealand, Ali et al. (2012) identified numerous flame retardants in mattress dust, including polybrominated diphenyl ethers (e.g. BDE 47, BDE 99, BDE 183, BDE 197, and BDE 209), organophosphates (e.g.

TEP, TCEP, TCPP, and TDCPP, among others), HBCDs, and novel flame retardants (BTBPE, TBB, and TBH, among others). Concentrations for the various flame retardants ranged over several orders of magnitude, from  $< 1$  to  $10^3$  ng/g mattress dust. Plasticizers and flame retardants may be found as chemical additives in mattresses, especially infant crib mattresses (Boor et al. 2015b), and have been shown to have endocrine disrupting properties. It is likely that some of these compounds partitioned directly into settled mattress dust as they migrated from the underlying mattress foam and encasing, whereas others may have originated from common indoor sources, such as vinyl flooring, foam furniture, and electronic casings, and then deposited (after partitioning to airborne particles) on the mattress and bedding.

#### *Particle Deposits on Mattresses and Bedding Materials*

In addition to the pollutants identified in mattress dust, the mass of dust that accumulates on mattresses, pillows, and bedding is an important parameter that may affect exposure and surface-to-air transport of the settled particles. Field studies that reported the dust loads were listed in Table B1, typically in the range of 0.1 to 1 g/m<sup>2</sup>. Jovanovic et al. (2004) reported median loads of 0.393 g/m<sup>2</sup> for children with allergic history, and 0.403 g/m<sup>2</sup> for a control group in Southwest Germany. In Ramallah, Palestine, El Sharif et al. (2004) reported loadings (mean±SD) of 0.29±0.20 g/m<sup>2</sup> for city residences, 0.59±0.31 g/m<sup>2</sup> for village residences, and 0.62±0.36 g/m<sup>2</sup> for camp residences, demonstrating that dust loading tends to increase from urban to more rural environments. Gehring et al. (2005) found higher mean dust loads on the mattresses of mothers (0.7

g/m<sup>2</sup>) compared to their infants (0.2 g/m<sup>2</sup>) in residences in Munich and Leipzig, Germany. A follow-up study by Chen et al. (2007) reported similar loadings (mean and range) of 0.74 (0.45-1.17) g/m<sup>2</sup> on mothers' mattresses and 0.12 (0.12-0.29) g/m<sup>2</sup> on infants' mattresses. A cross-country study by Giovannangelo et al. (2007) reported loadings of (median and range) 0.25 (0.01-1) g/m<sup>2</sup> in Germany, 0.25 (0.01-1) in the Netherlands, and 0.15 (0.02-0.9) g/m<sup>2</sup> in Sweden, finding loading to be generally the same across the three countries. In Saskatchewan, Canada, Rennie et al. (2008) found dust loadings of (mean±SD) 0.37±0.26 g/m<sup>2</sup>. Another cross-country study by Tischer et al. (2011) reported dust loadings (median and interquartile range) of 0.26 (0.14-0.47) g/m<sup>2</sup> in Germany and 0.25 (0.15-0.37) g/m<sup>2</sup> in Netherland. Lastly, in Taiwan, Wu et al. (2012) found dust loading (mean and 95% of confidence interval) to decrease with daily vacuuming of the mattress, from 1.5 (1.12-2) g/m<sup>2</sup> initially, to 0.27 (0.18-0.41) g/m<sup>2</sup> after 8 weeks of vacuuming. Caution should be taken when comparing the results of dust loading reported among these studies, because dust collection and sieving methods employed by a particular field and study locations may significantly influence the results (Boor et al. 2013a).

There have been no published studies that have explicitly measured the size distribution of settled mattress dust, although it likely ranges from < 1 µm to > 100 µm, given the diverse spectrum of biological and inorganic particles that can exist: allergen particles, fungal spores, bacteria, skin flakes, bedding fiber fragments, dust mite bodies (~ 200-300 µm), and particles originating elsewhere in the bedroom that have deposited on the mattress. Mite and animal allergens, such as Der p 1, Der f 1, Fel d 1, Bal g 2, Mus m 1 (mouse), and Rat n 1 (rat), are

primarily carried on particles ~ 5-10  $\mu\text{m}$  in size, although their size distribution can range from < 1 to > 20  $\mu\text{m}$  (Platts-Mills et al. 1986, Custovic et al. 1999, O'Meara and Tovey 2000, Chang and Gershwin 2004). Reponen et al. (2001) measured the size of various fungal species, including *Penicillium brevicompactum*, *Penicillium melinii*, *Cladosporium cladosporioides*, *Septomyces albus*, and *Thermoactinomyces vulgaris*, with various techniques including Scanning Electron Microscope (SEM), optical microscope, aerodynamic particle sizer (APS), and found that the spores are typically in the range of 1 to 4  $\mu\text{m}$ . Smaller fungal fragments less than 1  $\mu\text{m}$ , are also common (Reponen et al. 2007). Górný et al. (1999) reported the size distribution of various bacterial genera, including *Micrococcus/Kocuria*, *Staphylococcus*, *Bacillus*, *Pseudomonadaceae*, *Aeromonas*, and *Nocardia*, with aerodynamic diameters within the range 1 to > 7  $\mu\text{m}$ . Meklin et al. (2002) reported similar size ranges of 1 to > 7  $\mu\text{m}$  for viable airborne bacteria. As discussed in Weschler et al. (2011), human body sheds about  $5 \times 10^8$  skin cells per day, a significant fraction of which will accumulate in mattress dust during extended sleep periods, with the average skin flake about  $40 \times 30 \times 2 \mu\text{m}$  in size.

#### CHEMICAL EMISSIONS FROM MATTRESSES, PILLOWS, AND BED FRAMES

Although there is a robust body of literature on the identification and quantification of pollutants in mattress dust, there is comparatively less research on characterizing emissions of chemical pollutants from mattresses, pillows, and bed frames. Several recent studies have found that infant crib mattresses are a likely source of a variety of chemical additives, including VOCs, plasticizers, flame retardants, and unreacted isocyanates (Boor et al. 2014, 2015b, Liang and

Xu 2014b), which can enter the infant sleep microenvironment through volatilization as they partition from the material-phase to the gas-phase. Additionally, bed frames may release formaldehyde, due to its presence in the resin of composite wood products used to construct the bed structure.

### *Volatile Organic Compounds*

The composition of a typical crib mattress includes a thick layer of polyurethane or polyester foam padding (inner-springs are also used) encased within a thin, waterproof plastic cover to protect the mattress foam and to provide an easy-to-clean surface. Studies by Anderson and Anderson (1999) and (2000) and Boor et al. (2014) have shown that infant crib mattresses and mattress covers are a source of a variety of VOCs. VOCs identified in the Anderson and Anderson (1999) and (2000) studies include: styrene, trimethylbenzene, dichlorobenzene, isopropylbenzene, limonene, phenol, m-xylene, nitrobenzene, and toluene, among others. Boor et al. (2014) identified a different collection of compounds from a collection of 20 new and used crib mattresses, including: phenol, isooctanol, neodecanoic acid, d-limonene, linalool, nonanal, decanal, palmitic acid, and isopropyl myristate. Associations between VOC exposure and poor respiratory health have been observed among infants. Early life exposures to VOCs may affect the developing immune system and increase the risk of allergic disease, asthma, and pulmonary infections in young children, even at low concentrations (e.g. Diez et al. 2000, Rumchev et al. 2004, Franklin 2007). Additionally, Strachan and Carey (1995) discussed how VOCs released from pillows in close proximity to the BZ might increase mucosal permeability to

inhaled allergens, which the authors believe may explain the association between the use of foam pillows and childhood asthma.

### *Flame Retardants*

Crib mattresses manufactured with polyurethane foam are highly flammable and flame retardants are added so that they meet various flammability standards. To be effective, flame retardants are often present in the finished product at percent to tens-of-percent levels by weight (Stapleton et al. 2009, 2011). Concerns over flame retardant additives in children's products arose in the 1970s, when researchers demonstrated that children who wore sleepwear impregnated with the flame retardant tris(2,3-dibromopropyl)phosphate (tris-BP) were dermally exposed to the compound (Blum et al. 1978). Tris-BP was found to be a mutagen and eventually removed from commercial use (Blum et al. 1978). Throughout the next few decades, polybrominated diphenyl ether (PBDE) flame retardants, specifically pentaBDE congeners, were commonly added to crib mattress foam.

PBDEs are persistent and bioaccumulative compounds (Rudel and Perovich 2009). Infant exposure from indoor sources, such as a crib mattress, can occur through multiple pathways, including inhalation, dermal uptake, and ingestion of settled dust (Harrad et al. 2004). Infants are disproportionately exposed to PBDE flame retardants and have substantially higher serum PBDE concentrations than adults (Toms et al. 2009). Their serum concentrations have been correlated with concentrations in bedroom dust (Watkins et al. 2012).

Additionally, Toms et al. (2009) suggested that infant-specific sources, such as crib mattresses containing PBDEs, might play a factor in their elevated exposures and Heffernan et al. (2011) commented that little is known about the potential role of crib mattresses in contributing to an infant's cumulative body burden of PBDE flame retardants.

There is a growing body of evidence which suggests that exposure to PBDEs can cause a variety of deleterious health effects in infants. Rudel and Perovich (2009) highlighted studies that have demonstrated that PBDEs can interfere with thyroid hormone homeostasis, and consequently, delay neurological development in humans and rodents. Additionally, PBDE exposure has been found to impair a child's motor, cognitive, and behavioral performance and decrease fecundability (Roze et al. 2009, Herbstman et al. 2010). Concern over the negative health impacts of pentaBDE flame retardants led industry to end production in the U.S. in 2004. Although no longer found in new crib mattresses, these persistent compounds are still present in many older mattresses and can continue to volatilize and enter the indoor environment. This is especially important considering the long lifetime of a crib mattress (~10 years) and their common reuse in families with multiple children. Flame retardants currently added to crib mattresses as pentaBDE replacements include organophosphates and the Firemaster® 550 mixture. Unfortunately, these chemicals also present a health risk for infants.

Organophosphate flame retardants added to crib mattresses include tris(1,3-dichloro-2-propyl)phosphate (TDCPP), tris(2-chloroethyl)phosphate

(TCEP), and tris(1-chloropropyl)phosphate (TCPP) (Stapleton et al. 2011, Boor et al. 2015). Dishaw et al. (2011) demonstrated that organophosphate flame retardants might affect neurodevelopment with similar or greater potency compared to known and suspected neurotoxicants, such as chlorpyrifos pesticides. Meeker and Stapleton (2010) found elevated levels of these compounds in house dust to be associated with altered hormone levels and decreased sperm quality in males. Of even greater concern is that the most frequently detected organophosphate flame retardant, TDCPP, has been designated as carcinogenic by the State of California under Proposition 65, as it has been found to be associated with increases in kidney, liver, and testicular tumors in rats (Freudenthal and Henrich 2000, Faust and August 2011). Lastly, a recent study by Patisaul et al. (2012) found the flame retardant mixture Firemaster® 550 to be an endocrine disruptor and cause advanced female puberty, weight gain, and cardiovascular health effects in rats. Despite these concerning developments, organophosphate flame retardants and Firemaster® 550 are still added to crib mattresses today (Stapleton et al. 2011).

### *Plasticizers*

Most crib mattresses have a plastic cover for waterproofing and antibacterial purposes, and phthalates are extensively used as plasticizers to enhance the softness and flexibility of the cover. Because they are not chemically bound to the polymer matrix, they slowly volatilize from the material and migrate into surrounding environments (Xu et al. 2006, Clausen et al. 2012, Xu et al. 2012, Liang and Xu 2014a, 2014b). Phthalate plasticizers identified in crib

mattress covers include bis(2-ethylhexyl) phthalate (DEHP), bis(2-ethylhexyl) isophthalate (iso-DEHP), diisononyl phthalate (DINP) (Boor et al. 2015b).

As with flame retardants, infant exposure to phthalates can occur through multiple pathways and infants typically receive higher doses compared to adults, making them more susceptible to the adverse health effects of phthalates (Heudorf et al. 2007). Phthalates have endocrine disrupting properties (Bergman et al. 2013) and have been shown to retard male development (Adibi et al. 2003, Swan et al. 2005), alter semen quality (Hauser et al. 2006), cause irreversible changes to the male reproductive tract (Matsumoto et al. 2008), and increase the risk of allergic disease and asthma (Bornehag et al. 2004, Kolarik et al. 2008, Larsson et al. 2010).

Recently, the Consumer Product Safety Improvement Act was enacted in the U.S., restricting the use of certain phthalates (i.e., di-n-butyl-phthalate (DnBP), butyl-benzyl-phthalate (BBP), and DEHP) in toys and child care articles (66). However, there is some debate as to whether or not crib mattresses are included in the definition of “child care articles” (U.S. CPSC 2008, NRDC 2009). In addition, the ban on DINP and di-iso-decyl-phthalate (DIDP) only applies to children’s toys that can be placed in the mouth. Alternative plasticizers that have a chemical structure similar to phthalates, such as di(2-ethylhexyl) adipate (DEHA) and diisononyl cyclohexane-1,2-dicarboxylate (DINCH), have emerged very recently. Boor et al. (2015b) identified both DEHA and DINCH in new crib mattress covers. DEHA may induce mild developmental toxicity (Dalgaard et al. 2003) and has been shown to decrease anogenital distance and retention of

nipples in rats (Jarfelt et al. 2005). Studies suggest that DINCH may not have endocrine disrupting properties or reproductive toxicity, although it has been associated with thyroid hyperplasia and renal toxicity (EFSA 2006).

### *Unreacted Isocyanates*

An excess of toluene diisocyanate (TDI, the predominate diisocyanate used in polyurethane foam production, Vangronsveld et al. 2013a) is typically added during the manufacture of polyurethane foam, above which is necessary for it to react with hydroxyl groups of polyols and water (Hugo et al. 2000). As such, unreacted isocyanate may be present in the final polyurethane foam product (Hugo et al. 2000, Krone et al. 2003, CA OEHHA 2010). Krone et al. (2003) identified NCO in polyurethane foam-containing consumer products, including a mattress and sofa padding. The authors detected unreacted isocyanates in numerous foam samples, including a mattress pad (2,4-TDI concentration of 20.3 µg/g of foam), carpet pad (2,6-TDI of 2.5 µg/g and 2,4-TDI of 3.6 µg/g), and a 30 year old sofa pad (2,6-TDI of 8.6 µg/g). NCO was also detected in polyurethane-foam containing crib mattresses via PAS-FT-IR analysis in Boor et al. (2015b). The foam in crib mattresses is somewhat protected from excess moisture released from the infant body, which can react with the isocyanates (Bello et al. 2006). Isocyanates may play an important role in the development of childhood asthma (Krone and Klinger 2005 and references therein) and exposure has been associated with allergic sensitization, asthma, rhinitis, and contact dermatitis (Bello et al. 2007, Verschoor and Verschoor 2014).

## *Formaldehyde*

Formaldehyde may be found in resins used in composite wood products of bed structures. In a test of seven new wooden cribs, Madsen and Gibson (2008) found three to emit formaldehyde at a high rate. Recently, formaldehyde has been classified as a human carcinogen, and inhalation exposure may cause sinus and nasal cancers, leukemia, inflammation of lung tissue, and irritation of the upper airways. In addition, epidemiological studies in young children (6 months to 3 years of age) have shown a significant association between formaldehyde exposure and the risk of childhood asthma. In a review of 21 epidemiological studies, formaldehyde and the presence of formaldehyde-emitting particleboard were identified as two of the most important risk factors for childhood respiratory and allergic effects. Children's exposure to formaldehyde has been positively associated with lower-respiratory and asthma-related symptoms, allergy, eczema, and altered immune system responses. Asthma-related and lower respiratory symptoms have also been associated with new furniture and particle board in the homes of infants, including apartments redecorated during "nesting" periods and following infant birth.

## THE SOURCE-PROXIMITY EFFECT OF THE SLEEP MICROENVIRONMENT

In the sleep microenvironment, there are several key factors that contribute to source-proximity effect (Figure B5), including the spatial proximity of the BZ to the source, incomplete mixing of room air, concentrations gradients near an actively emitting source, the personal cloud due to human body movement-induced particle resuspension, the buoyant human thermal plume,

heat transfer from the human body to the source, which may elevate the emissions of gaseous pollutants, and direct dermal contact with the source.

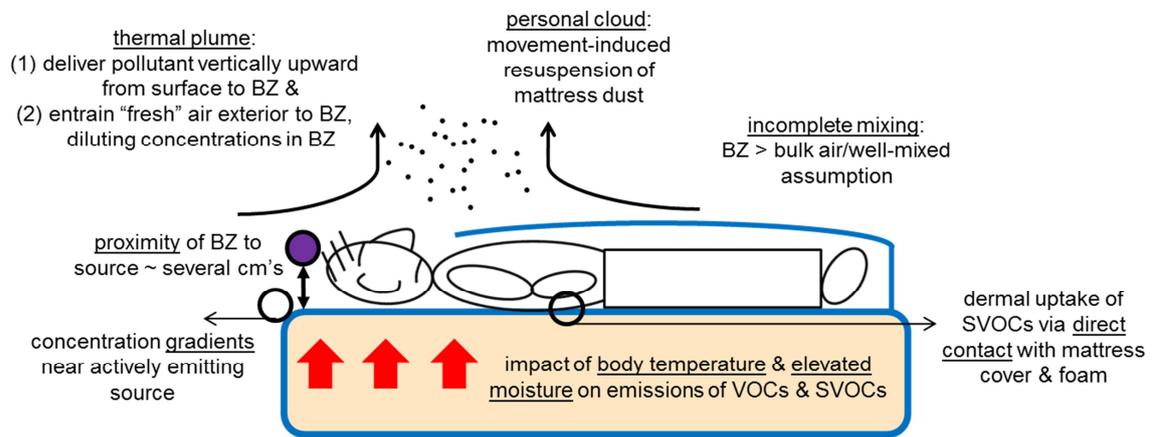


Figure B5: The source-proximity effect of the sleep microenvironment.

### *Spatial Proximity and Concentration Gradients near Actively Emitting Sources*

The spatial proximity of a person’s BZ to their mattress and bedding can lead to elevated concentrations in their BZ relative to the bulk bedroom air. The spatial proximity effect has been evaluated in numerous studies, which typically release inert tracer gases (CO, CO<sub>2</sub> or sulfur hexafluoride (SF<sub>6</sub>)) from point or area sources and use sampling arrays to measure the uniformity of the dispersion of the tracer gas in a space (e.g. Kulmala et al. 1996, McBride et al. 1999, Acevedo-Bolton et al. 2012). Collectively, these studies have shown that tracer gas concentrations near actively emitting sources are greater than those at locations further from the source, typically by a factor greater than 2.

A recent study by Laverge et al. (2013) highlighted the important role of the spatial proximity effect on inhalation exposure to pollutants originating in the sleep microenvironment. A breathing thermal manikin was positioned on a

twin-size mattress with a pillow, from which SF<sub>6</sub> was uniformly released to simulate the emission of gaseous pollutants. The relative intake fraction, defined as the ratio of the BZ concentration to the well-mixed chamber concentration, was reported for different sleeping positions and bedding arrangements. BZ SF<sub>6</sub> concentrations were found to be significantly greater than those measured in the bulk air, with mean relative intake fractions for the mattress source of 1.24 for the supine position, 1.39 for the lateral position, and 1.73 for the prone position. Higher mean relative intake fractions were reported for the pillow source with 2.16 for the supine position and between 3.39 and 4.02 for the lateral position. Additionally, covering the entire manikin's body and head with a blanket increased the relative intake fraction to 32.7. Boor et al. (2014) reported similar findings for VOCs emitted from a crib mattress, with BZ TVOC concentrations greater than those in the bulk chamber air by factors in the range of 1.8 to 2.4. In regard to resuspended mattress dust particles, Boor et al. (2015a) also found BZ concentrations to be greater than bulk air concentrations by factors of 1.07-1.94, depending on particle size. The collective findings of these studies demonstrate that a person will receive elevated exposures to particulate and gaseous pollutants that are released from their mattresses, beyond what would be estimated from bulk bedroom air measurements.

#### *Incomplete Mixing of Bedroom Air*

The spatial proximity effect is due in part to incomplete mixing of the released pollutant with the bulk air, which leads to the development of concentration gradients in the space, as suggested in some of the aforementioned

studies. Additional studies by Bowes et al. (1993), Drescher et al. (1995), Furtaw et al. (1996), Mage and Ott (1996), Yang et al. (1998), Wang and Chen (2007), and Rim and Novoselac (2010), among others, have found that incomplete mixing conditions are associated with non-uniform pollutant concentration distributions in indoor environments.

Incomplete mixing may be aided by the low ventilation rates that have been reported for bedrooms and the reduced run time of ventilation systems at night. Bekö et al. (2010) measured the ventilation rates in the naturally ventilated bedrooms of 500 children in Denmark and reported a mean ventilation rate of  $0.46 \text{ h}^{-1}$ , with rates as low as  $0.1 \text{ h}^{-1}$ , suggesting that children's bedrooms are poorly ventilated. In addition, Stephens et al. (2011) found that the fractional operation time of residential ventilation systems is the lowest during the late evening/early morning hours (10PM to 7AM). As the ventilation system run time is reduced, a residence relying on forced mechanical ventilation to deliver fresh (or filtered) air to the bedroom during sleep periods may see poor mixing conditions in the space (e.g. more stagnant air due to reduced ventilation and less human activity and movement to enhance mixing) and inadequate dilution of pollutants released within the sleep microenvironment, leading to the accumulation of pollutants in the air. Furthermore, it is expected that for an actively emitting diffusion-controlled source, such as a mattress, concentrations in very close proximity (several cm) will be greater than those in the bulk air, as a chemical compound partitions from the source to the air immediately above it. Thus, it cannot be assumed that a pollutant released from a mattress or bedding will be well mixed with the bulk bedroom air, and BZ concentrations should be

measured to accurately estimate inhalation exposure to pollutants originating in this microenvironment.

#### *Personal Cloud Effect and Human-Induced Resuspension of Mattress Dust*

The personal cloud effect, whereby people can be envisioned as being surrounded by a cloud of particles, is another factor that may lead to elevated exposures in the sleep microenvironment. The personal cloud effect was used to explain the consistently higher personal exposures to particles compared to stationary monitoring locations (Wallace 1996). Rabinovitch et al. (2005) provides one example in which personal (approximate BZ) bacterial endotoxin concentrations measured with personal monitors are about 3 times greater than those measured at stationary indoor locations, suggesting that endotoxin exposures are better correlated with sources in close proximity to the person and their BZ than those in the bulk indoor air. Human-induced particle resuspension from indoor surfaces, including mattresses, is one of the physical processes contributing to the personal cloud effect, leading to elevations in BZ concentrations beyond those in the bulk air. In the sleep microenvironment, the personal cloud effect is likely to be enhanced due to the close proximity of a person's BZ to the surfaces from which particle resuspension is occurring, an important consideration for larger particles ( $> 10 \mu\text{m}$ ) that have high settling velocities (on the order of  $\sim 10^{-3} \text{ m/s}$ ), and likely short trajectories upward from a surface. Investigations by Spilak et al. (2014) and Boor et al. (2015a) have shown that human body movements in bed, such as rolling from the prone to

supine position, can resuspend significant quantities of settled particles on mattresses, pillows, bed sheets, and blankets.

### *Body Movements during Sleep Periods*

Mattress dust resuspension is driven by human body movements in bed. Human activity in bed can occur during periods of wakefulness, e.g. as a person enters a bed and prepares themselves for sleep, and throughout their extended sleep state. Although the absence of voluntary motor behavior is a characteristic of the sleep state, movements are commonly reported, ranging from about ten significant body posture shifts (e.g. Aaronson et al. 1982), to several hundred smaller body movements (e.g. Azumi et al. 1977). The cause of body movements during sleep periods is largely unknown, although several theories suggest that movements occur to relieve muscular discomfort during extended periods of immobility (Howat 1972, Giganti et al. 2008). Monitoring sleep body movements is an integral part of the fields of sleep science and physiology, and studies are often conducted to elevate sleep quality.

Sleep is typically divided into several sleep stages: stage W (Wakefulness), NREM stage 1 (non-rapid eye movement), NREM stage 2, NREM stage 3 or SWS (Slow Wave Sleep, e.g. "deep sleep"), and REM (rapid eye movement) (e.g. Silber et al. 2007). It is important to understand the dependency of body movements on sleep stages, as body movements are responsible for inducing the necessary removal forces (aerodynamic, mechanical) to resuspend mattress dust particles. Shimohira et al. (1998) found a strong dependency of body movement intensity

on sleep stage among children 4 to 12 years of age. Body movements were classified into four groups, GM-1: axial rotation of the body, GM-2: movement of limbs and trunk (torso) muscles without rotation, GM-3: movement of two or more limbs but not trunk muscles, and GM-4: movement of only one limb. The number of body movements per hour was reported for each sleep stage (Figure 1 in Shimohira et al. 1998). The authors found that movements are much more frequent during both NREM stage 1 and REM sleep, with a decrease during SWS (deep sleep). Additionally, the most commonly reported movement was GM-1, in which the child rotates some degree, with up to 13 movements/hour during NREM 1 sleep.

Wilde-Frenz and Schulz (1983) observed a similar trend among adults, 21 to 36 years of age, where body movement frequency was the greatest during stage W, followed by NREM 1, then REM, then NREM 2, and SWS. Body movement frequency ranged from  $< 0.1$  to  $0.8$  body movements per sleep minute (bm/min), depending on the sleep stage. Among 11 subjects, the total number of body movements ranged from 105 to 182 for the entire sleep period. Giganti et al. (2008) monitored the sleep quality of adults in two age groups: 61-75 (group 1) and 76-98 (group 2), and reported a weaker dependency of body movement on sleep stage than that reported among younger subjects. Body movements were greatest during NREM 1 sleep, with movement frequency remaining nearly constant during NREM 2, SWS, and REM sleep. Body movements were found to decrease with age, with an average of  $0.25$  bm/min for age group 1 and  $0.09$  bm/min for age group 2, equating to an average of 86 body movement during the entire sleep period for group 1 (mean sleep time of 342 min.) and 27

movements for group 2 (mean sleep time of 297 min.). Although fewer movements were observed for group 2, the authors found a greater probability of awakening after a movement, compared to group 1.

The findings of these studies suggest that body movement frequency is a function of both sleep stage and age. Thus, the potential for human-induced particle resuspension would vary throughout a sleep period, and may be greater during periods of more frequent activity (e.g. NREM 1 and REM stages) than during deep sleep stages (SWS). Boor et al. (2015a) demonstrated that more intense body movements in bed, such as a full 360° rotation of the torso, are associated with higher surface vibrations and emissions of resuspended particles compared to less intense movements (e.g. sitting on the mattress). Studies have suggested that asthmatic children and adults have more disturbed sleep patterns (Janson et al. 1996, Stores et al. 1997), and Zanobetti et al. (2010) found sleep-disordered breathing, defined as the recurrent episodic disruption of normal breathing during sleep, and decreases in sleep efficiency to be associated with increases in short-term variation in the airborne concentration of 10 µm particles. People with asthma and other breathing ailments may have a greater frequency of body movements during their disrupted sleep period, which may put them at greater risk for elevated exposure to resuspended mattress dust particles.

### *Human Thermal Plume*

In the sleep microenvironment, the airflow pattern is a result of two different flows and their interaction: the buoyant human thermal plume and

transient flows generated by breathing (Homma and Yakiyama 1988, Zukowska et al. 2008). While the thermal plume depends on the metabolic rate of a sleeping person and the sleep stage, transient flow is mainly dependent on whether the sleeping person inhales/exhales via nose or mouth. The buoyant human thermal plume may also be responsible for elevated BZ concentrations for pollutant sources in close proximity of the human body. Rim and Novoselac (2009) and (2010) demonstrated that the thermal plume is an effective mechanism for transporting pollutants, released from various locations in close vicinity to the human body, vertically upward, toward to BZ. Thus, the thermal plume that develops around a sleeping person may aid in transporting pollutants released from the mattress surface, upward and toward their BZ. However, the thermal plume may also serve the secondary role of entraining “fresh” air exterior to the sleep microenvironment, thereby diluting concentrations of mattress pollutants in the BZ, as suggested by Laverge et al. (2013). The thermal plume also interacts with the shape of the breathing zone, especially the inhalation zone, reducing it to a shallow area of only a few centimeters wide stretching down around the cheeks of a person resting in supine position (Laverge et al. 2015).

#### *Impact of Body Temperature and Sweating on Chemical Emissions from Mattresses*

Another unique attribute of the sleep microenvironment that may play an important role in enhancing the source-proximity effect is the warming of nearby objects via heat exchange with the human body. Along with warming the surrounding air via convective heat exchange, leading to the development of the thermal plume, our bodies will also transfer heat to surfaces that are in direct

contact via conduction and those in close proximity via radiation. Lu et al. (1999) monitored the surface temperature of a mattress around sleeping adult male subjects with a matrix of 16 thermistors. Maximum surface temperatures were found to range from 31 to 36°C beneath and around the lower limbs and between 34 and 36°C beneath and around the waist. Thus, it is to be expected that the sleeping human body will increase the surface temperature of their mattress, pillow, and bedding materials. The emissions of gaseous pollutants, such as VOCs and SVOCs, from building materials has been shown to increase with temperature (e.g. Haghghat et al. 1998, Lin et al. 2009, Clausen et al. 2012), with recent studies by Boor et al. (2014) and Liang and Xu (2014b) observing this effect in crib mattresses and mattress covers. Thus, the close proximity of the human body to potential pollutant sources may enhance volatilization, and subsequently, further increase BZ concentrations. Elevated moisture levels may also have a strong impact on emissions of VOCs and SVOCs from mattresses. Throughout the sleep period, the sleeping thermal environment is a permanently humid environment due to the production of large quantities of moisture through sweating and the large buffering capacity provided by mattresses and bedding materials.

#### *Direct Dermal Contact with Mattresses and Bedding Materials*

Transdermal uptake of pollutants, such as SVOCs, via contact transfer and air-to-skin transport, is another contributing factor to the source-proximity effect. During sleep periods, a person's body, and exposed skin, is in intimate contact with their pajamas, mattress, mattress cover, and bedding materials. As outlined

in a review by Weschler and Nazaroff (2012), dermal exposure is an important pathway for certain SVOCs, including plasticizers that are added to mattress covers (Boor et al. 2015b). If the gas-phase concentrations of these pollutants are greater near the source, dermal uptake via air-to-skin transport may be enhanced. Laverge et al. (2013) demonstrated that concentrations of mattress-released gaseous pollutants ( $\text{SF}_6$ ) under the bedding (where most of the body is exposed) can be up to 30 times higher than those in the bulk air of the room. Additionally, during the day, SVOCs found in mattresses, as well as in the bulk bedroom air, may partition to bedding fibers. Weschler and Nazaroff (2012) hypothesize that temperature elevations during sleep periods may promote desorption of the sorbed SVOCs, thereby offering an additional route of transdermal uptake.

#### AUTHOR CONTRIBUTIONS

B.E.B. wrote the paper, with writing contributions from J.L., M.P.S., and Y.X. Y.X. and A.N. provided guidance for the structure of the literature review, advised in the interpretation of the literature, and provided detailed comments on draft manuscripts.

Table B1: HDM/animal/plant allergens, fungi, fungal (1-3)- $\beta$ -Glucan, bacteria, bacterial endotoxin, plasticizers, and flame retardants detected in mattress dust in selected field studies.

Study	Description	Location of Field Study	Pollutant	Concentration	Units	Dust Load, $m_0$ (g/m <sup>2</sup> )
Garrett et al. (1998)	Evaluated the impact of bedding characteristics on HDM concentrations	Latrobe Valley, Victoria, Australia	Der p 1	Wool bedding: Always: 30.6 (24.8-37.8) Sometimes: 23.6 (20.0-27.7) Never: 11.2 (8.6-14.6) Type of mattress: Inner-spring: 25.0 (21.9-28.6) Foam: 13.0 (8.9-19.1)	$\mu$ g/g mattress dust (Mean (95% CI))	Not reported
Fahlbusch et al. (2000)	Quantified indoor grass pollen allergens in house dust	Hamburg, Erfurt, Zerbst, Bitterfeld & Hettstedt, Germany	Phl p 5	<0.03 (<0.03-0.120)	$\mu$ g/g mattress dust (Median (25 <sup>th</sup> -75 <sup>th</sup> percentiles))	Not reported
Stubner et al. (2000)	Examined the impact of home remediation interventions on lowering allergen levels in low-income households	Birmingham, Alabama, USA	Various allergens (e.g. Der p 1, Der f 1, Fel d 1)	Home 1, Mattress: 15 (before intervention), <2 (after intervention) Home 2, Mattress: 20 (before intervention), 4 (after intervention) Home 3, Mattress: 235 (before intervention), 16 (after intervention) Home 4, Mattress: 840 (before intervention), -- (after intervention) Home 5, Mattress 1: 630 (before intervention), 240 (after intervention) Home 5, Mattress 2: 565 (before intervention), -- (after intervention) Home 6, Mattress: 5 (before intervention), -- (after intervention)	$\mu$ g/mg mattress dust	Not reported
Hirsh et al. (2000)	Measured HDM and mold spore concentrations before and after installation of windows and central heating systems	Dresden, Germany	Der f 1 <i>Cladosporium</i> spp. <i>Penicillium</i> spp. <i>Alternaria</i> spp. <i>Aspergillus</i> spp. <i>Aspergillus fumigatus</i> spp.	Before renovation: 1.56 (0.89-2.74) After renovation: 2.40 (1.33-4.36) Before renovation: 130 (50-370) After renovation: 60 (20-160) Before renovation: 1500 (740-3,020) After renovation: 1,250 (590-2,660) Before renovation: 10 (0-20) After renovation: 10 (10-30) Before renovation: 590 (270-1,320) After renovation: 820 (400-1,710) Before renovation: 10 (0-10) After renovation: 10 (10-20)	$\mu$ g/g mattress dust (Mean (95% CI))  CFU/g mattress dust (Mean (95% CI))	Not reported
Douwes et al. (2000)	Investigated the relationship between endotoxin, $\beta$ -Glucans, and	Amsterdam, Netherlands	Endotoxin	Non-symptomatics: 1,820 $\pm$ 3.6 (level), 4,772 $\pm$ 2.8 (load) Symptomatics: 2,082 $\pm$ 4.3 (level), 5,696 $\pm$ 3.6 (load) Asthmatics: 1,202 $\pm$ 3.3 (level), 3,983 $\pm$ 1.8 (load) Cough: 3,402 $\pm$ 4.7 (level), 7,670 $\pm$ 4.3 (load)	Level: EU/m <sup>2</sup> mattress area sampled Load: EU/g mattress dust (Mean $\pm$ SD)	Not reported

	HDM allergens in mattress dust with peak expiratory flow in children		(1-3)- $\beta$ -D-Glucan  Der p 1	Non-symptomatics: 276 $\pm$ 2.4 (level), 718 $\pm$ 1.8 (load) Symptomatics: 293 $\pm$ 2.3 (level), 792 $\pm$ 2.1 (load) Asthmatics: 283 $\pm$ 1.9 (level), 903 $\pm$ 1.8 (load) Cough: 303 $\pm$ 2.6 (level), 683 $\pm$ 2.3 (load)  Non-symptomatics: 537 $\pm$ 6.1 (level), 1,440 $\pm$ 6.1 (load) Symptomatics: 432 $\pm$ 7.5 (level), 1,147 $\pm$ 6.4 (load) Asthmatics: 318 $\pm$ 9.0 (level), 960 $\pm$ 7.8 (load) Cough: 646 $\pm$ 5.9 (level), 1,481 $\pm$ 5.9 (load)	Level: $\mu$ g/m <sup>2</sup> mattress area sampled Load: $\mu$ g/g mattress dust (Mean $\pm$ SD) Level: ng/m <sup>2</sup> mattress area sampled Load: ng/g mattress dust (Mean $\pm$ SD)	
Su et al. (2001)	Measured seasonal concentrations of HDM allergens and endotoxin	Southern Taiwan	Der p 1 Der p 2 Endotoxin	Spring: 3.78, Summer: 5.61, Fall: 9.11, Winter: 10.15 Spring: 1.41, Summer: 1.49, Fall: 1.96, Winter: 3.32 Spring: 45.18, Summer: 17.96, Fall: 50.25, Winter: 18.84	$\mu$ g/g mattress dust (Mean) ng/mg mattress dust (Mean)	Not reported
Pitten et al. (2001)	Investigated the impact of mattress encasings on fungal growth	Germany		Fungal genera and species identified in dust on mattresses with cotton encasings: <i>Alternaria</i> spp., <i>Aspergillus</i> spp., <i>Candida</i> spp., <i>Cladosporium cladosporioides</i> , <i>Mycelia sterilia</i> , <i>Penicillium</i> spp., <i>Rhodotorula</i> spp., <i>Scopulariopsis brevicaulis</i> , <i>Scopulariopsis</i> spp., <i>Sirodesmium-like fungus</i> , <i>Trichoderma</i> spp., <i>Ulocladium</i> spp. Fungal genera and species identified in dust on mattresses with synthetic encasings: <i>Aspergillus</i> spp., <i>Cladosporium cladosporioides</i> , <i>Mycelia sterilia</i> , <i>Penicillium</i> spp., <i>Scopulariopsis brevicaulis</i> , <i>Scopulariopsis</i> spp.		Not reported
Oppermann et al. (2001)	Measured levels of HDM allergens and fungi in children's mattresses	East & West Germany	Der f 1 Der p 1 Common fungal species identified: <i>Penicillium</i> spp., <i>Eurotium</i> spp., <i>Aspergillus</i> spp., <i>Alternaria</i> spp., <i>Epicoccum</i> spp., <i>Cladosporium</i> spp.	1.36 1.14  Total fungi: 26,500 (1,400-300,000)	$\mu$ g/g mattress dust (Mean)  CFU/g mattress dust (Mean (Range))	Not reported
Mihrshahi et al. (2002)	Measured HDM allergens in residential homes and investigated the impact of mattress and bedding characteristics on concentrations	Sydney, Australia	Der p 1	Type of mattress: Foam: 21.39 (13.77-33.25) Inner spring: 14.22 (12.58-16.07) Mattress Age: $\leq$ 2 years: 10.31 (8.03-13.24) >2 years: 15.47 (13.47-17.67) Wool blankets: Yes: 27.18 (21.06-35.08) No: 12.88 (11.38-14.57) Synthetic blankets: Yes: 22.18 (17.51-28.11) No: 13.21 (11.64-14.99) Sheepskin: Yes: 22.95 (14.37-36.66)	$\mu$ g/g mattress dust (Mean (95% CI))	Not reported

				No: 13.84 (12.31-15.56)		
Jovanovic et al. (2004)	Measured indoor fungi levels in homes of children with and without a history of allergies	Southwest Germany	Common fungal species identified: <i>Aspergillus</i> spp., <i>Penicillium</i> spp., <i>Alternaria</i> spp., <i>Cladosporium</i> spp.	Total viable mold spores: 16,250/38,238 (0-2,500,000)	CFU/g mattress dust (Median/Mean (Range))	Children with allergic history: 0.393 Controls: 0.403 (Median)
Gehring et al. (2004)	Measured endotoxin concentrations in the mattress dust of mothers and their infants	Munich & Leipzig, Germany	Endotoxin	Mothers' mattress: Munich: 2,000 (level), 3,000 (load) Leipzig: 2,200 (level), 3,100 (load) Infants' mattress: Munich: 900 (level), 4,900 (load) Leipzig: 1,000 (level), 6,600 (load)	Level: EU/m <sup>2</sup> mattress area sampled Load: EU/g mattress dust (-Mean)	Infants' mattress: ~0.2 Mothers' mattress: ~0.7
El Sharif et al. (2004)	Measured concentrations of HDM allergens, pet allergens, and endotoxin in mattress dust	Ramallah, Palestine	Der p 1 Der f 1 Can f 1 Fel d 1 Endotoxin	City residences: 958 (load), 4,290 (level) Village residences: 1,941 (load), 3,776 (level) Camp residences: 3,100 (load), 5,656 (level) City residences: 48 (load), 206 (level) Village residences: 33 (load), 64 (level) Camp residences: 24 (load), 42 (level) City residences: 100 (load), 451 (level) Village residences: 99 (load), 192 (level) Camp residences: 164 (load), 298 (level) City residences: 91 (load), 408 (level) Village residences: 210 (load), 408 (level) Camp residences: 151 (load), 275 (level)  City residences: -- (load), 18,738 (level) Village residences: 19,214 (load), 25,700 (level) Camp residences: 23,972 (load), 43,696 (level)	Load: ng/g mattress dust Level: ng/m <sup>2</sup> mattress area sampled (Mean)  Load: EU/g mattress dust Level: EU/m <sup>2</sup> mattress area sampled (Mean)	City residences: 0.29±0.20 Village residences: 0.59±0.31 Camp residences: 0.62±0.36
Instanes et al. (2005)	Measured concentrations of allergens and endotoxin in mattress dust in day-care centers	Oslo, Norway	Fel d 1 Can f 1 Endotoxin	1.5 (load), 0.17 (level)  10.0 (load), 1.7 (level)  5.0 (load), 0.9 (level)	Load: µg/g mattress dust Level: µg/m <sup>2</sup> mattress area sampled (Median)  Load: ng/g mattress dust Level: ng/m <sup>2</sup> mattress area sampled (Median)	Not reported
da Silva et al. (2005)	Investigated mite fauna in mattress dust	Londrina, Brazil	HDM bodies: total mites HDM bodies: Pyroglyphidae mites	Crib: 404.5±183.3 Bed: 1,075.8±198.8  Crib: 289.9±136.7 Bed: 875.0±183.6	Mite bodies/g mattress dust (Mean±95% CI) Note: bodies represent all life stages: eggs,	Not reported

			Der p 1	Crib: 5.8±2.7 Bed: 17.5±3.7	larvae, nymphs, and adults. µg/g mattress dust (Mean±95% CI, estimate)	
Hicks et al. (2005)	Analyzed fungal flora and concentrations in settled dust	Northern California, USA	Culturable fungi <i>Cladosporium</i> <i>Penicillium</i> spp. Yeasts <i>Aureobasidium</i> <i>Aspergillus niger</i> Nonsporulating fungi <i>Alternaria</i> <i>Epicoccum</i>	62,300 (37,300-104,000) 22,000 (ND-180,000) 4,000 (ND-120,000) 8,800 (NS-200,000) 4,300 (ND-68,000) 400 (ND-56,000) 800 (ND-1,400) 1,800 (ND-12,000) 800 (ND-12,000)	CFU/g bedspread and furniture dust (Mean (95% CI))  CFU/g bedspread and furniture dust (Median (Range))	Not reported
van den Bemt et al. (2006)	Evaluated the influence of mattress characteristics on HDM concentrations	Netherlands	Der p 1	Type of mattress: Latex: 1.191 (0.744-1.908) Waterbed: 0.433 (0.073-2.559) Polyester: 1.199 (0.592-2.431) Inner spring: 1.418 (0.594-3.387) Upper layer material: Cotton: 0.801 (0.343-1.873) Synthetic: 2.606 (1.527-4.448) Cotton/synthetic: 1.382 (0.885-2.158) Other: 0.447 (0.111-1.792) Mattress age: ≤5 years: 0.986 (0.517-1.883) >5 years: 1.190 (0.731-1.939) Relative humidity in bedroom: ≤50%: 0.796 (0.444-1.426) >50%: 1.975 (1.312-2.974)	log µg/g mattress dust (Mean (95% CI))	Not reported
Woodcock et al. (2006)	Identified fungal flora in used (18 months to >20 years) synthetic and feather pillows	United Kingdom	<i>Penicillium</i> spp. <i>Aspergillus vitus</i> <i>Rhodotorula mucilaginosa</i> <i>Aspergillus glaucus</i> <i>Scopulariopsis brevicaulis</i> <i>Aspergillus fumigatus</i>	15,100 27,800 69,400 41,700 13,900 Synthetic pillows: 2,745 Feather pillows: 1,863	CFU/g pillow dust Mean  CFU/g pillow material (not dust)	Not reported

			<i>Aureobasidium pullulans</i>	Synthetic pillows: 1,926 Feather pillows: 5,110	Mean	
Chen et al. (2007)	Explored associations between socioeconomic status and the concentrations of indoor bio-contaminants	Munich & Leipzig, Germany	Der p 1 Der f 1 Fel d 1 Endotoxin	Mothers' Mattress: 97.7 (5-608.3) Infants' Mattress: 5.0 (5.0-642.9) Mothers' Mattress: 634.1 (125.4-3,601.8) Infants' Mattress: 525.1 (5.0-2,324.7) Mothers' Mattress: 180.2 (68.6-820.5) Infants' Mattress: 326.0 (84.7-1,322.5) Mothers' Mattress: 3,008 (1,045.5-7,913) Infants' Mattress: 5,866 (2,336-14,669)	ng/g mattress dust (Median (25 <sup>th</sup> -75 <sup>th</sup> percentiles)) ng/g mattress dust (Median (25 <sup>th</sup> -75 <sup>th</sup> percentiles)) ng/g mattress dust (Median (25 <sup>th</sup> -75 <sup>th</sup> percentiles)) EU/g mattress dust (Median (25 <sup>th</sup> -75 <sup>th</sup> percentiles))	Mothers' Mattress: 0.7385 (0.453-1.166) Infants' Mattress: 0.188 (0.120-0.289)
Krop et al. (2007)	Assessed the presence of occupational laboratory animal allergens in mattress dust of workers	Netherlands	Rat urinary allergen Mouse urinary allergen Mouse urinary protein	Laboratory animal workers: 39.3 (19.8-78.0) Non-laboratory animal workers (controls): 7.6 (4.7-12.2) Laboratory animal workers: 29.5 (11.7-74.6) Non-laboratory animal workers (controls): 8.8 (4.6-16.8) Laboratory animal workers: 30.9 (12.8-74.8) Non-laboratory animal workers (controls): 5.6 (2.0-16.0)	ng/g mattress dust (Median (Range))	Not reported
Giovannangelo et al. (2007)	Measured levels of $\beta$ -Glucans and fungal extracellular polysaccharide in mattress dust of children	Germany, Netherlands & Sweden	(1-3)- $\beta$ -Glucan Fungal Extracellular Polysaccharide (EPS)	Germany: 450 (150-1,050) (level), 1,050 (900-2,050) (load) Netherlands: 400 (150-1,000) (level), 1,050 (750-3,000) (load) Sweden: 350 (100-1,000) (level), 225 (1,050-4,000) (load) Germany: 10,000 (2000-50,000) (level), 40,000 (10,500-100,000) (load) Netherlands: 9,000 (1,050-45,000) (level), 40,000 (10,500-100,000) (load) Sweden: 2,050 (400-10,500) (level), 15,000 (4,000-50,000) (load)	Level: $\mu$ g/m <sup>2</sup> mattress area sampled Load: $\mu$ g/g mattress dust (-Median (95% CI)) Level: EPSU/m <sup>2</sup> mattress area sampled Load: EPSU/g mattress dust (-Median (95% CI))	Germany: ~ 0.25 (0.01-1) Netherlands: ~ 0.25 (0.01-1) Sweden: ~ 0.15 (0.02-0.9) (-Median (Range))
Rennie et al. (2008)	Investigated associations between endotoxin levels and asthma and wheeze in children	Saskatchewan, Canada	Endotoxin	2,069.79 (4,650.02) (level), 7.99 (9.85) (load)	Level: EU/m <sup>2</sup> mattress area sampled Load: EU/mg mattress dust (Median (IQR))	0.365 $\pm$ 0.26 (Mean $\pm$ SD)
Nam et al. (2008)	Measured HDM allergen concentrations in house dust	Cheonan, Korea	Der f 1 Der p 1	4.0 (0.01-112.1) (level), 10.2 (0.01-230.9) (load) 0.02 (0.01-8.8) (level), 0.14 (0.01-30.0) (load)	Level: $\mu$ g/m <sup>2</sup> mattress area sampled Load: $\mu$ g/g mattress dust (-Median (Range))	Not reported
Korthals et al. (2008)	Investigated the occurrence of <i>Listeria</i> spp. bacteria in mattress dust of	Bavaria, Germany	<i>Listeria</i> spp. <i>L. monocytogenes</i> <i>L. innocua</i>	Detected in 8% of mattress dust samples (Culture methods) Detected in 3% of mattress dust samples (Culture methods) Detected in 8% of mattress dust samples (Real-time PCR methods) Detected in 8% of mattress dust samples (Culture methods)		Not reported

	farm children					
Vogel et al. (2008)	Identified microbial flora on the mattresses of farm children	Bavaria, Germany	<i>Bacillus</i> spp. Coliform bacteria <i>Lactobacillus</i> spp. <i>Enterococcus</i> spp. <i>Clostridium</i> spp. <i>Aspergillus</i> spp. <i>Mucor</i> spp. <i>Cladosporium</i> spp. <i>Penicillium</i> spp. <i>Wallemia</i> spp.	5.25 (0.87) 5.67 (0.94) 3.00 (3.30) 3.15 (4.47) 0.00 (3.00) 4.00 (1.10) 3.75 (1.00) 0.00 (3.74) 3.00 (4.00) 3.48 (4.00)	log. CFU (Median (IQR))	Not reported
Soleimani and Rafinejad (2008)	Analyzed HDM contamination in hotels and inns	Bandar-Abbas, Iran	HDM species: <i>D. pteronyssinus</i> (identified in 84.5% of samples), <i>D. farinae</i> (identified in 13% of samples), <i>C. malaccensis</i> (identified in 2.5% of samples)			Not reported
Heyes et al. (2009)	Measured (1-3)- $\beta$ -Glucan levels in pillows, duvets, and mattresses	Wellington, New Zealand	(1-3)- $\beta$ -Glucan	Mattress: 15.7 (10.4-23.9) (level), 76.6 (61.4-94.0) (load) Duvets: 8.8 (5.4-14.1) (level), 132.1 (68.9-207.9) (load) Pillows: 3.5 (2.5-4.8) (level, $\mu\text{g}$ pillow), 132.1 (68.9-207.9) (load) Synthetic Pillows: 3.8 (1.4-11.0) (level, $\mu\text{g}$ /pillow), 105.4 (76.9-144.6) (load) Feather Pillows: 2.0 (0.8-5.1) (level, $\mu\text{g}$ /pillow), 76.6 (36.7-159.9) (load)	Level: $\mu\text{g}/\text{m}^2$ mattress/pillow/duvet area sampled Load: $\mu\text{g}/\text{g}$ mattress/pillow/duvet dust (Mean (95% CI))	Not reported
Wu et al. (2009)	Measured allergen and microbial bio-contaminant concentrations in homes of asthmatic children	Central Taiwan	Der p 1 Der f 1 Blo t 5 Total HDM allergen Endotoxin $\beta$ -Glucan	Pillow: 1.33 (0.91-1.94) Bamboo side of mattress: 0.38 (0.25-0.58) Conventional side of mattress: 0.77 (0.51-1.17) Pillow: 0.22 (0.14-0.33) Bamboo side of mattress: 0.24 (0.14-0.44) Conventional side of mattress: 0.55 (0.30-1.01) Pillow: 0.014 (0.011-0.017) Bamboo side of mattress: 0.024 (0.017-0.033) Conventional side of mattress: 0.165 (0.115-0.237) Pillow: 2.72 (1.99-3.73) Bamboo side of mattress: 1.35 (0.95-1.91) Conventional side of mattress: 3.50 (2.65-4.62) Bamboo side of mattress: 163.7 (82.6-244.8) Conventional side of mattress: 358.2 (211.7-504.8) Bamboo side of mattress: 27.9 (22.4-34.8) Conventional side of mattress: 25.5 (21.9-29.8)	$\mu\text{g}/\text{g}$ mattress/pillow dust (Mean (95% CI))  EU/mg mattress dust (Mean (95% CI)) $\mu\text{g}/\text{g}$ mattress dust (Mean (95% CI))	Not reported
Chen et al. (2009)	Investigated associations between infant exposure to	Munich & Leipzig, Germany	Endotoxin	Mothers' mattress: 2,071 (595-6,919) (level), 3,008 (1,046-7,913) (load) Children's mattress: 1,015 (330-3,022) (level), 5,866 (2,336-14,669) (load)	Level: EU/m <sup>2</sup> mattress area sampled Load: EU/g mattress dust	Not reported

	endotoxins and development of eczema				(Median (IQR))	
Täubel et al. (2009)	Explored the origins of bacteria in house dust	Finland	Bacterial phyla and genera  Endotoxin  Lipopolysaccharide (LPS)  Muramic acid	Gram-positive bacterial groups (phylum:genera) identified in mattress dust: Actinobacteria: <i>Corynebacterium</i> , <i>Propionibacterium</i> , <i>Micrococcus</i> Firmicutes: <i>Staphylococcus</i> , <i>Lactobacillus</i> , <i>Streptococcus</i> , <i>Lactococcus</i> Deinococcus-Thermus: <i>Deinococcus</i> Gram-negative bacterial phyla identified in mattress dust: Proteobacteria: <i>Paracoccus</i> , <i>Aggregatibacter</i> Bacteroidetes: <i>Prevotella</i> , <i>Bacteroides</i>  1.4±3.7 (load), 194.1±5.8 (level)  0.031±1.379 (load), 4.396±1.944 (level)  15.5±1.7 (load), 2191.2±3.3 (level)	Load: EU/mg mattress dust Level: EU/m <sup>2</sup> mattress area sampled (Mean±SD) Load: nmol/mg mattress dust Level: nmol/m <sup>2</sup> mattress area sampled (Mean±SD) Load: ng/mg mattress dust Level: ng/m <sup>2</sup> mattress area sampled (Mean±SD)	Not reported
Eberlein et al. (2009)	Measured endotoxin and allergen levels in hospitals and adjacent homes	Bavaria, Germany	Endotoxin Der p 1 Der f 1 Fel d 1	Hospitals: 321,720 (230,500-518,700) Adjacent Homes: 243,980 (156,520-399,020) Hospitals: 0 (0-208) Adjacent Homes: 163 (0-391) Hospitals: 409 (152-839) Adjacent Homes: 791 (320-2,074) Hospitals: 663 (117-2,426) Adjacent Homes: 1227 (261-2,824)	EU/g mattress dust (Median (IQR))  ng/g mattress dust (Median (IQR))	Not reported
Visitsunthorn et al. (2010)	Measured HDM allergen concentrations in different types of mattresses	Bangkok, Thailand	Der p 1 + Der f 1	Coconut mattress: 0.96 (0.39-1.45) (month 1), 16.60 (0-33.7) (month 12) Kapok mattress: 1.44 (1.06-1.83) (month 1), 31.30 (17.40-42.80) (month 12) Sponge (polyurethane foam) mattress: 0.96 (0.23-1.75) (month 1), 12.90 (1.00-17.90) (month 12) Synthetic mattress: 1.30 (0.80-1.89) (month 1), 25.30 (12.10-32.50) (month 12)	µg/g mattress dust (Median (IQR))	Not reported
Doyen et al. (2011)	Measured endotoxin concentrations in mattresses at time of birth of newborn and at 6 months of life	Belgium	Endotoxin	At birth, mattresses in newborn's bedroom: 17.6 (0.4-346.7) (Site A), 20.4 (0.8-26.3) (Site B) 6 months of life, mattresses in newborn's bedroom: 79.6 (3.8-518.8) (Site A), 101.8 (6.5-634.4) (Site B)	EU/mg mattress dust (Median (Range))	Not reported

Leung et al. (2011)	Measured allergen levels in settled dust in residences of asthmatic children	Hong Kong	Fel d 1 Bla g 2 Blo t 5	0.06 (0.03-0.10) 24.8 (14.2-47.7) 0.26 (0.14-0.79)	µg/g mattress dust (Median (IQR)) ng/g mattress dust (Median (IQR)) µg/g mattress dust (Median (IQR))	Not reported
Tischer et al. (2011)	Investigated exposure of children to endotoxins, β-Glucans, and EPS mold components in house dust	Germany & Netherlands	Endotoxin (1-3)-β-D-Glucan Extracellular polysaccharides (EPS)	German LISA & GINI Studies: 3,053 (1,521-6,015) (level), 12,222 (7,379-21,337) (load) Dutch PIAMA Study: 2,356 (1,461-4,208) (level), 10,608 (6,550-17,366) (load) German LISA & GINI Studies: 421 (238-865) (level), 1,859 (1,277-2,396) (load) Dutch PIAMA Study: 380 (199-625) (level), 1,662 (1,135-2,205) (load) German LISA & GINI Studies: 1,008 (4,458-25,904) (level), 40,792 (24,235-65,371) (load) Dutch PIAMA Study: 8,257 (3,890-17,310) (level), 34,696 (20,364-58,156) (load)	Level: EU/m <sup>2</sup> mattress area sampled Load: EU/g mattress dust (Median (IQR)) Level: µg/m <sup>2</sup> mattress area sampled Load: µg/g mattress dust (Median (IQR)) Level: EPSU/m <sup>2</sup> mattress area sampled Load: EPSU/g mattress dust (Median (IQR))	German LISA & GINI Studies: 0.257 (0.139-0.471) Dutch PIAMA Study: 0.247 (0.148-0.366) (Median (IQR))
Hsu et al. (2012)	Explored associations between indoor phthalate concentrations and mold growth	Tainan, Taiwan	DEHP BBzP DBP	w/o visible mold: 656.6 (459.4-980.9) w/ visible mold: 933.3 (580.0-1,404.9) w/o visible mold: 2.1 (1.0-6.4) w/ visible mold: 3.7 (1.0-6.1) w/o visible mold: 16.6 (10.4-22.8) w/ visible mold: 20.2 (12.4-40.9)	µg/g mattress dust (Median (25 <sup>th</sup> -75 <sup>th</sup> percentiles))	Not reported
Ege et al. (2012)	Identified environmental bacteria in dust on children's mattresses	Bavaria, Germany		Bacterium identified which are related to a physician's diagnosis of asthma: <i>Corynebacterium mycetoides</i> , <i>Zoogloea</i> sp., <i>Duganella</i> sp., <i>Aurantimonas ureolytica</i> sp., <i>Serratia marcescens</i> , <i>S. nematodiphila</i> , <i>Serratia</i> sp., <i>Pseudomonas fluorescens</i> , <i>Corynebacterium tuberculostearicum</i> , <i>Gardnerella vaginalis</i> , <i>Lactobacillus curvatus</i> , <i>Lactobacillus sakei</i> , <i>Streptococcus</i> sp., <i>Moraxella</i> sp., <i>Staphylococcus sciuri</i> sp., <i>Jeotgalicoccus</i> sp., <i>Salinicoccus</i> sp., <i>Macrococcus brunensis</i> , <i>Bacillus</i> sp. Bacterium identified which are related to hay fever: <i>Aerobacter ureolytica</i> , <i>P. fluorescens</i> , <i>Corynebacterium mucifaciens</i> , <i>C. freiburgense</i> , <i>C. variabile</i> , <i>C. sp. Triatoma infestans</i> , <i>Neisseria meningitidis</i> , <i>N. mucosa</i> , <i>N. subflava</i> Bacterium identified which are related to atopic sensitization: <i>Enterobacter cloacae</i> , <i>E. aerogenes</i> , <i>E. cancerogenus</i> , <i>E. ludwigii</i> , <i>Pantoea</i> sp., <i>Kluyvera cryocrescens</i> , <i>Erwinia persicina</i> , <i>Aurantimonas altamirensis</i> ; <i>A. ureolytica</i> , <i>Mesorhizobium</i> sp., <i>Lactobacillus iners</i> , <i>Acinetobacter Iwoffii</i>		Not reported
Barnig et al. (2012)	Measured endotoxin concentrations in the dust of rural and urban residences	Strasbourg & Haut-Doubs, France	Endotoxin	Type of farm: Non-modern: 3,700±4,900 Modern: 1,200±1,600 Barn position: Adjacent to house: 3,400±4,700 Separated to house: 2,200±3,600 Storage hay mode: Round bales: 3,200±4,700 In bulk: 1,100±900 Other: 2,700±3,600	ng/g mattress dust (Mean)	Not reported

Begum et al. (2012)	Evaluated the prevalence of fungi in used pillows	Perth, Australia	Fungi: <i>Alternaria</i> sp., <i>Aureobasidium</i> sp., <i>Cladosporium</i> sp., <i>Epicoccum</i> sp., <i>Monilia</i> sp., <i>Penicillium</i> sp., Yeast, <i>Rhizopus</i> sp.	Total fungal counts: 241 (22-563) (Yeast most prevalent)	CFU/pillow Mean (Range)	0.06 to 0.36 g pillow dust/pillow
Lawson et al. (2012)	Measured endotoxin concentrations and loading in mattress dust	Humboldt, Saskatchewan, Canada	Endotoxin	Children with asthma or wheeze (cases): 19.6 (15.6-24.8) Children without asthma or wheeze (controls): 21.1 (15.6-24.8) Children with asthma or wheeze (cases): 240.5 (212.1-370.9) Children without asthma or wheeze (controls): 376.2 (324.4-436.2)	EU/mg mattress dust (Mean (95% CI))  EU/m <sup>2</sup> mattress area sampled (Mean (95% CI))	Not reported
Ali et al. (2012)	Identified and quantified flame retardants in floor and mattress dust	Wellington, Wairarapa, Christchurch, & North Canterbury, New Zealand	OPFRs HBCDs NBFRs BFRs	TEP: 10 (<10-235), TnBP: 65 (20-1,920), TCEP: 38 (<20-475), TCPP: 250 (133-1,920), TBEP: 1,545 (635-3,610), TPhP: 240 (20-35,190), TDCPP: 103 (20-6,465), TCP: 157 (<50-2,155) $\alpha$ -HBCD: 41 (3-345), $\beta$ -HBCD: 9 (<2-122), $\gamma$ -HBCD: 80 (18-1,260) BTBPE: 1 (<2-37), TBB: 3 (<2-40), TBPH: 1 (<2-50), DBDPE: 9 (<5-220) BDE 47: 35 (2-290), BDE 99: 40 (8-540), BDE 183: 6 (<2-20), BDE 197: 4 (<2-18), BDE 209: 735 (106-21,960)	ng/g mattress dust (Median (Range))	Not reported
Wu et al. (2012)	Assessed the impact of daily vacuuming of mattresses to reduce levels of HDM allergens, bacterial endotoxin and fungal $\beta$ -Glucans	Taiwan	Der p 1 Der f 1 Endotoxin $\beta$ -Glucan	Week 0: 0.68 (0.32-1.40), Week 8: 0.46 (0.23-0.92) Week 0: 1.18 (0.60-2.30), Week 8: 0.69 (0.41-1.14) Week 0: 91 (64-130), Week 8: 97 (75-125) Week 0: 63.2 (46.1-86.6), Week 8: 75.1 (54.1-104.0)	$\mu$ g/g mattress dust (Mean (95% CI)) EU/mg mattress dust (Mean (95% CI)) $\mu$ g/g mattress dust (Mean (95% CI))	1.5 (1.12-2) at Week 0, to 0.27 (0.18-0.41) at Week 8 (Mean (95% CI))
Casas et al. (2013)	Analyzed the results of the European HITEA project (Health Effects of Indoor Pollutants: Integrating microbial, toxicological, and epidemiological approaches) with 4 European birth cohorts	Finland, Germany, Netherlands, & Spain	Endotoxin Fungal Extracellular Polysaccharide (EPS) (1-3)- $\beta$ -Glucan	German LISA Study: 23.0 (21.1-25.1) Dutch PIAMA Study: 22.0 (19.8-24.5) Spanish INMA Study: 3.2 (2.7-3.9) Finnish LUKAS2 Study: 17.6 (15.8-19.5) German LISA Study: 47.5 (43.7-51.6) Dutch PIAMA Study: 30.7 (27.8-33.8) Spanish INMA Study: 116.7 (107.2-127.0) Finnish LUKAS2 Study: 39.6 (34.1-46.0) German LISA Study: 0.9 (1.8-2.0) Dutch PIAMA Study: 1.7 (1.5-1.8) Spanish INMA Study: -- Finnish LUKAS2 Study: 2.4 (2.2-2.7)	EU/mg mattress dust (Mean (95% CI))  EPSU/mg mattress dust (Mean (95% CI))  $\mu$ g/mg mattress dust (Mean (95% CI))	Not reported
Callesen et al. (2014)	Evaluated	Odense,	Der p 1	Healthy Controls: 110.0	ng/g mattress dust	Not reported

associations between selected allergens and asthma, rhinoconjunctivitis, and atopic dermatitis in preschool children	Denmark	Der f 1 Der p 2 Fel d 1 Can f 1	<p>Asthma: 132.2, s-IgE -: 145.4, s-IgE +: 132.0  Rhinocconjunctivitis: 119.4, s-IgE -: 164.9, s-IgE +: 104.8  Atopic dermatitis: 131.8, s-IgE -: 176.0, s-IgE +: 151.0  Healthy Controls: 977.2</p> <p>Asthma: 988.4, s-IgE -: 471.8, s-IgE +: 1,726  Rhinocconjunctivitis: 1,251, s-IgE -: 173.7, s-IgE +: 988.4  Atopic dermatitis: 751.9, s-IgE -: 672.4, s-IgE +: 1,211  Healthy Controls: 121.5</p> <p>Asthma: 119.5, s-IgE -: 69.2, s-IgE +: 135.5  Rhinocconjunctivitis: 153.4, s-IgE -: 158.1, s-IgE +: 130.4  Atopic dermatitis: 119.5, s-IgE -: 88.3, s-IgE +: 130.0  Healthy Controls: 114.5</p> <p>Asthma: 76.7, s-IgE -: 210.7, s-IgE +: 42.1  Rhinocconjunctivitis: 85.6, s-IgE -: 61.6, s-IgE +: 99.6  Atopic dermatitis: 150.4, s-IgE -: 170.9, s-IgE +: 116.6  Healthy Controls: 81.6</p> <p>Asthma: 71.1, s-IgE -: 77.7, s-IgE +: 71.8  Rhinocconjunctivitis: 54.2, s-IgE -: 28.5, s-IgE +: 66.4  Atopic dermatitis: 53.0, s-IgE -: 53.3, s-IgE +: 57.2</p>	(Median)
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CFU: Colony Forming Units; EU: Endotoxin Units; EPSU: Fungal Extracellular Polysaccharide (EPS) Units; PCR: Polymerase Chain Reaction; IgE: Immunoglobulin E antibody; CI: Confidence Interval; IQR: Interquartile Range; ND: Not Detected; DEHP: Bis(2-ethylhexyl) phthalate; BBzP: Benzyl butyl phthalate; DBP: Dibutyl phthalate; OPFRs: Organophosphate flame retardants: TEP (tri-ethyl-phosphate), TnBP (tri-*n*-butyl-phosphate), TCEP (tris-(2-chloroethyl)-phosphate), TCP (tri-cresyl-phosphate), TBEP (tri-(2-butoxyethyl)-phosphate), TCPP (tris-(1-chloro-2-propyl) phosphate), TDCPP (tris-(2,3-dichloropropyl)-phosphate), TPhP (tri-phenyl-phosphate); BFRs: Brominated flame retardants; HBCDs: Hexabromocyclododecanes:  $\alpha$ -HBCD,  $\beta$ -HBCD,  $\gamma$ -HBCD; NBFRs: Novel Brominated flame retardants: BTBPE (1,2-bis(2,4,6-tribromophenoxy)ethane), DBDPE (decabromodiphenyl ethane), TBB (2-ethylhexyl-2,3,4,5-tetrabromobenzoate), TBPH (bis(2-ethylhexyl)-3,4,5,6-tetrabromophthalate); Der p 1: Dermatophagoides pteronyssinus Group 1 (European HDM); Der p 2: Dermatophagoides pteronyssinus Group 2 (European HDM); Der f 1: Dermatophagoides farinae Group 1 (American HDM); Can f 1: Canis familiaris Group 1 (Dog allergen); Fel d 1: Felis domesticus Group 1 (Cat allergen); Phl p 5: Phleum pratense Group 5 (Common timothy pollen); Bla g 2: Aspartic protease Group 2 (Cockroach allergen); Blo t 5: Blomia tropicalis Group 5 (Blomia tropicalis HDM); LISA Study: Lifestyle Related Factors on the Immune System and the Development of Allergies in Childhood (Germany); GINI Study: German Infant Nutritional Intervention (Germany); PIAMA Study: Prevention and Incidence of Asthma and Mite Allergy (Netherlands); INMA Study: Infancia y Medio Ambiente (Environment and Childhood) (Spain); LUKAS2 Study: Lapsuuden kasvuympäristö ja allergiat 2 (Childhood environment and allergies) (Finland)



## Appendix C

## Paper C. Wind Tunnel Study on Aerodynamic Particle Resuspension from Monolayer and Multilayer Deposits on Linoleum Flooring and Galvanized Sheet Metal

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### ABSTRACT

Resuspension is an important source of indoor particles and the amount of dust loading is an important factor in resuspension emission rates. Field studies have shown that light to heavy dust loads can be found in the indoor environment, on both the surfaces of flooring and ventilation ducts. These diverse particle deposits can be broadly classified as either a monolayer, in which particles are sparsely deposited on a surface, or a multilayer, in which particles are deposited on top of one another and there is particle-to-particle adhesion and interaction. This experimental wind tunnel study explores the role of the type of particle deposit on aerodynamic resuspension from linoleum flooring and galvanized sheet metal. Resuspension fractions are reported for both monolayer and multilayer deposits exposed to a wide range of air velocities. The type of particle deposit is found to strongly influence resuspension. In general, the results show that resuspension from multilayer deposits can occur at significantly lower velocities compared to monolayer deposits. For example, resuspension fractions at an air velocity of 5 m/s for the canopy layer of multilayer deposits were similar to those found for monolayer deposits at 50 m/s. Additionally, for multilayer deposits, resuspension fractions for the canopy layer increased with increasing dust load and negligible resuspension occurred along the surface layer. It was found that the relationship between the particle deposit height and the viscous sublayer thickness of the airflow can help explain

the differences in resuspension that were observed between the two types of deposits. The impact of the type of particle deposit on resuspension may have important implications for resuspension in the indoor environment, where a diversity of deposits can be found.

## INTRODUCTION

Resuspension is an important secondary source of particles in the indoor environment and has been found to be associated with convective airflow in ventilation ducts (e.g. Krauter and Biermann 2007, Wang et al. 2012) and human activities indoors (e.g. Thatcher and Layton 1995, Ferro et al. 2004, Qian and Ferro 2008, Tian et al. 2011, Shaughnessy and Vu 2012). Particle resuspension induced by aerodynamic removal forces can be influenced by numerous variables. Previous experimental wind tunnel studies have demonstrated the important role of a variety of parameters on resuspension, including: particle size and air velocity (Corn and Stein 1965, Wu et al. 1992, Nicholson 1993, Braaten 1994, Ibrahim et al. 2003, Jiang et al. 2008, Mukai et al. 2009, Goldasteh et al. 2012b), surface material and roughness (Wu et al. 1992, Nicholson 1993, Gomes et al. 2007, Jiang et al. 2008, Mukai et al. 2009, Goldasteh et al. 2012b, Kassab et al. 2013); particle composition (Wu et al. 1992, Braaten 1994, Ibrahim et al. 2003, Goldasteh et al. 2012a); characteristics of the airflow, such as acceleration (Wu et al. 1992, Nicholson 1993, Ibrahim et al. 2003), turbulence (Ibrahim et al. 2004, Mukai et al. 2009), exposure time (Ibrahim et al. 2003); and relative humidity and residence time (Ibrahim et al. 2004). Particle resuspension may also be strongly dependent on whether the deposit is a monolayer or a multilayer (Boor et al. 2013). A monolayer deposit is one in which particles are sparsely deposited on a

surface, with negligible particle-to-particle contact. A multilayer deposit is a porous structure of particles deposited on top of one another, forming multiple layers. In these complex structures, the particle resuspension process is heavily affected by particle-to-particle adhesion and interaction between the larger particles aggregates and the airflow.

The type of particle deposit is of particular relevance to resuspension in the indoor environment, where we see a diversity of dust loads and deposit structures. Boor et al. (2013a) summarized field studies that reported dust loads on indoor surfaces. On hard flooring, dust loads were typically in the range of less than 0.1 to 1 g/m<sup>2</sup>, with lighter and heavier dust loads frequently reported in the literature (Table 1 in Boor et al. (2013a)). In ventilation ducts, dust loads were found to range over several orders of magnitude, from less than 0.1 to greater than 100 g/m<sup>2</sup>. The authors presented a simple scaling analysis that demonstrated that this wide range of dust loading represents both monolayer and multilayer deposits. The authors also summarized key findings from the experimental resuspension literature that highlighted important differences in the resuspension process between monolayer and multilayer deposits.

Only a few modeling and experimental wind tunnel studies have explored multilayer resuspension, including those by Fromentin (1989), Matsusaka and Masuda (1996), Lazaridis and Drossinos (1998), Chiou and Tsai (2001), Friess and Yadigaroglu (2001), Friess and Yadigaroglu (2002), Gac et al. (2008), and Nitschke and Schmidt (2010), among others. Collectively, these studies have identified

several unique characteristics associated with resuspension from multilayer deposits, including the following:

- Particles from the canopy layer of a multilayer deposit resuspend at lower velocities relative to particles in layers closer to the surface (Lazaridis and Drossinos 1998, Friess and Yadigaroglu 2001).
- There are reduced adhesion forces between spherical particles compared to that between a particle and a flat deposition surface (Lazaridis and Drossinos 1998).
- Resuspension often occurs in the form of larger particle aggregates, which, when airborne, can subsequently break apart due to forces imparted by turbulent bursts (Matsusaka and Masuda 1996, Kurkela et al. 2006, Gac et al. 2008, Gotoh et al. 2011).
- Enhanced resuspension may occur due to possible saltation effects (Bagnold 1941, Fairchild and Tillery 1982, Shao et al. 1993, Kok et al. 2012).
- The deposit structure and porosity is dependent on the deposition mechanism, e.g. gravitational settling may produce a “fluffy” deposit, compared to a “cake-like” deposit originating from inertial impaction (Friess and Yadigaroglu 2002).

Since both monolayer and multilayer deposits may be found on indoor surfaces, and there are fundamental differences in the resuspension process associated with both types of particle deposits, we expect the type of particle deposit to play an important role in the fraction of particles that resuspend from

a surface. The primary aim of this investigation is to develop a better understanding of the impact of the type of particle deposit on resuspension. An experimental methodology is developed to generate monolayer and multilayer deposits on linoleum flooring and galvanized sheet metal and to expose the deposits to a range of air velocities in a wind tunnel. The impact of the type of particle deposit is quantified by directly comparing resuspension fractions of fluorescent tracer particles from both deposits at different air velocities.

#### METHODOLOGY

An experimental methodology was developed to study aerodynamic resuspension from monolayer and multilayer particle deposits. For monolayer deposits, several variables were investigated: air velocity, particle size, relative humidity, and the type of indoor surface. Independent variables investigated for the multilayer deposits included: dust loading, air velocity, type of indoor surface, and the layer location (canopy compared to surface, Figure C1). Two particle diameters were studied to explore the impact of particle size: 3 and 10  $\mu\text{m}$ . Two flat indoor surfaces were examined for both deposits: linoleum, a common flooring material, and galvanized sheet metal, which is typically used to manufacture ventilation ducts. Two different seeding methods were developed to generate the monolayer and multilayer deposits. The deposits were then exposed to a range of air velocities in a wind tunnel. A fluorescence stereomicroscope was used to detect the deposited particles on the sample surface and a morphometry program was developed to count the number of

particles. Figure C2 outlines the experimental sequence for both monolayer (Figure C2 (a.)) and multilayer deposits (Figure C2 (b.)).

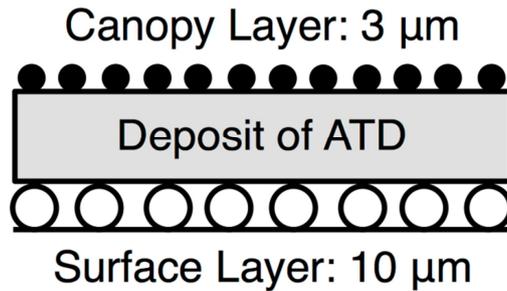


Figure C1: Multilayer formation: canopy layer of 3 μm fluorescent particles, varying dust load deposit of ATD, and surface layer of 10 μm fluorescent particles.

The resuspension metric used in our investigation is an absolute resuspension fraction,  $\Phi$ .  $\Phi$  is defined as the change in seeding density before and after the seeded sample is exposed to a given flow condition in the wind tunnel, divided by the initial seeding density. It varies between 0, in which there is no detectable resuspension, and 1, for maximum resuspension.

$$\Phi = \frac{\sigma_i - \sigma_f}{\sigma_i} \quad (\text{C.1})$$

The initial,  $\sigma_i$ , and final,  $\sigma_f$ , seeding densities are expressed as the number of particles per unit area, particles/mm<sup>2</sup>. The absolute resuspension fractions presented in our investigation are reported for a 100 second wind tunnel exposure time for both types of deposits.

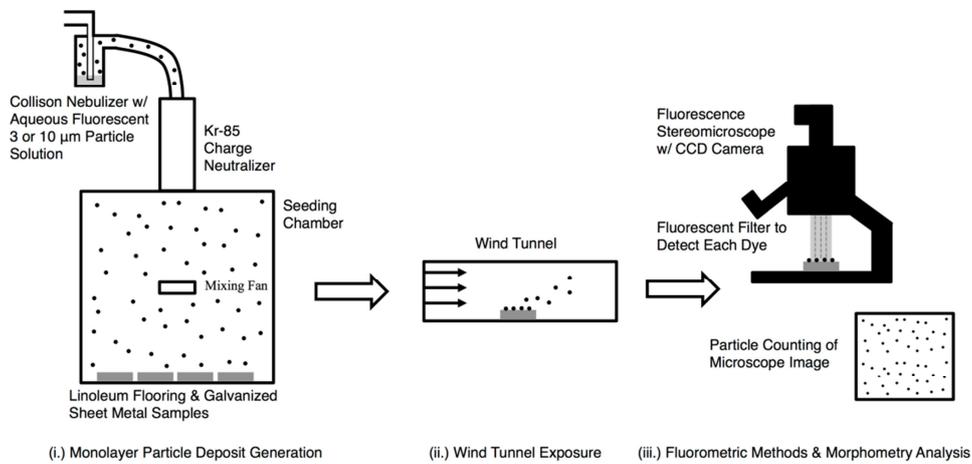


Figure C2 (a).

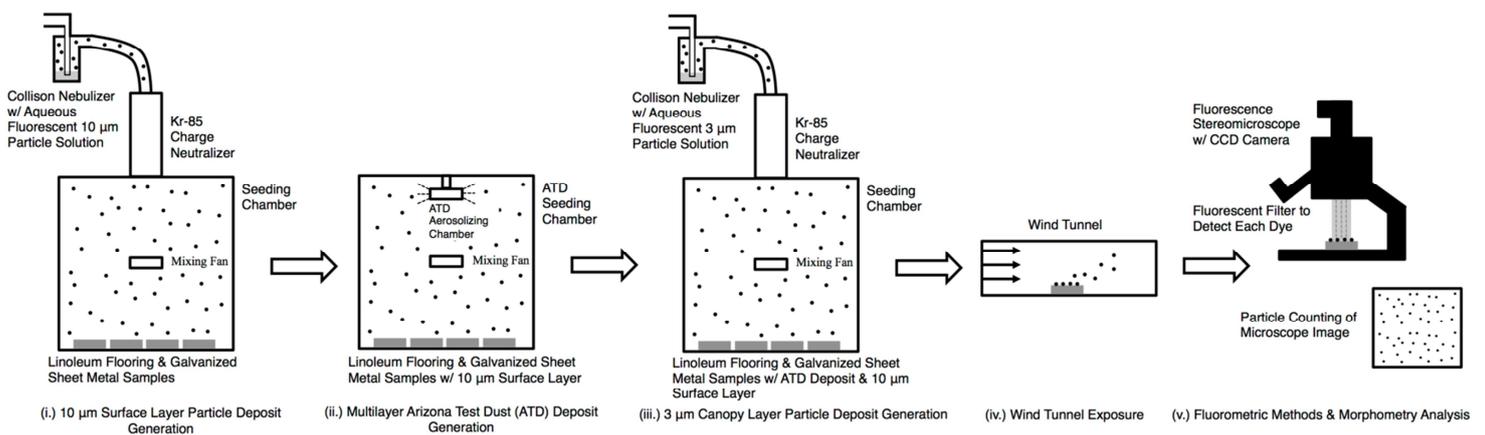


Figure C2 (b).

Figure C2: Experimental sequence for (a.) monolayer resuspension, (b.) multilayer resuspension.

### Generation of Monolayer and Multilayer Particle Deposits

To generate the monolayer and multilayer deposits, two different seeding methods were employed. An aqueous solution of internally dyed, spherical

polystyrene fluorescent particles (Thermo Scientific Inc., density of 1060 kg/m<sup>3</sup>) was used to generate a sparse monolayer deposit. To easily distinguish between particle sizes and to forgo actual measurement of individual particles during microscopy analysis, a specific fluorescent dye was used to represent each particle size: red dye for 3 μm and green dye for 10 μm. The actual size of the supplied particles was verified via air sampling with an aerodynamic particle sizer (TSI Inc., Model 3321). The fluorescent tracer particles were selected for their ease of generation; known, monodisperse size distribution; and detection via available instrumentation (e.g. Leica MZ16FA fluorescence stereomicroscope).

To generate the fluorescent particles for the monolayer deposit, the highly concentrated aqueous solution was first diluted with isopropyl alcohol (99% isopropyl alcohol, 1% deionized water). The diluted solution was then placed in a three-jet Collison Nebulizer (BGI Inc., Model CN24). Filtered, pressurized air supplied by the laboratory's compressed air system was directed into the Collison Nebulizer at 127 kPa. Isopropyl alcohol droplets were subsequently generated, carrying the fluorescent particles with the effluent air stream. Because a residual electrostatic charge can accumulate on the particles within the glass jar of the nebulizer, the particle stream was passed through a Kr-85 Aerosol Charge Neutralizer (TSI Inc., Model 3012) to ensure all particles had a Boltzmann charge distribution.

The particle stream was then directed into a 50 L square box that served as a seeding chamber, with the samples positioned at the bottom (Figure C2 (a.)). A

small mixing fan ensured the chamber particle concentration remained well mixed, which was subsequently verified by assessing the seeding density uniformity among the samples (seeding density coefficient of variance among the samples was generally below 5%). Each sample was 4.5 by 4.5 cm in size and thoroughly cleaned before seeding with 99% isopropyl alcohol to minimize surface contamination and residual electrostatic charges. A steady-state particle concentration was reached in the seeding chamber after an injection period of 15 minutes (the nebulizer discharge produces an air exchange rate of approximately  $6 \text{ h}^{-1}$ ), after which the particles were deposited via gravitational settling for approximately 6 hours. Initial seeding densities for the 3 and 10  $\mu\text{m}$  particles were (mean $\pm$ SD)  $67\pm 19$  and  $0.73\pm 0.16$  particles/ $\text{mm}^2$ , respectively. Multiplying by the particle mass, the corresponding monolayer dust loads for the 3 and 10  $\mu\text{m}$  particles were (mean $\pm$ SD)  $9.5\pm 2.7 \times 10^{-4}$  and  $3.8\pm 0.84 \times 10^{-4}$  g/ $\text{m}^2$ , respectively. As shown in Boor et al. (2013a), similar levels are reported for numerous monolayer wind tunnel studies.

The seeded samples were then placed in a 30 L conditioning chamber for 24 hours prior to wind tunnel exposure, where the relative humidity was controlled and recorded with a HOBO data logger (HOBOWare Pro, Onset Computer Co., Model U12-012). Two relative humidities were investigated: 35 and 70% (recorded values of  $35\pm 5\%$  and  $71\pm 3\%$ ). The humidities were selected to represent both a dry and moist indoor environment.

To generate the multilayer deposit, two seeding chambers and three seeding stages were required (Figures C1 and C2 (b.)). First, a surface layer of 10

$\mu\text{m}$  fluorescent particles was deposited in a sparse monolayer employing the aforementioned seeding method (initial seeding density of (mean $\pm$ SD)  $1.2\pm 0.11$  particles/ $\text{mm}^2$ ). The seeded samples were then placed in a second seeding chamber, where they were seeded with a multilayer deposit of polydisperse (1 to  $20 \mu\text{m}$ ) ISO 12103-1 A1 Ultrafine Arizona Test Dust (ATD) (Powder Technology Inc.). ATD was chosen over latex and silica microspheres and potassium chloride particles because it is both inexpensive and easily distributed along the sample surfaces in large quantities. An aerosolizing chamber was developed in which ATD was contained and an impinging jet of filtered air aerosolized the powder, which was then evenly dispersed through small inlets into the well-mixed seeding chamber. The ATD loading was measured gravimetrically with an analytical balance (Mettler-Toledo International Inc., model AB135-5). Four ATD dust loads were examined (mean $\pm$ SD):  $6.23\pm 1.10$ ,  $7.31\pm 1.00$ ,  $13.21\pm 3.33$ , and  $20.25\pm 1.91$   $\text{g}/\text{m}^2$ . The dust load of  $6.23 \text{ g}/\text{m}^2$  was selected as the minimum dust load such that none of the  $3 \mu\text{m}$  particles along the canopy layer would penetrate through the porous ATD to the surface. Trial experiments at dust loads below  $5 \text{ g}/\text{m}^2$  were unsuccessful, as a pure canopy layer could not be achieved due to this penetration.

Ultrafine ATD has a mass median diameter of  $4.5 \mu\text{m}$  and a bulk density of  $500 \text{ kg}/\text{m}^3$ . Applying the simplified particle deposit scaling analysis presented in Boor et al. (2013a), and assuming a porosity of 0.75 corresponding to gravitational settling in the seeding chamber, all four ATD dust loads were verified as multilayer deposits. The deposit height was estimated to range from approximately  $100 \mu\text{m}$  for a seeding density of  $6.23 \text{ g}/\text{m}^2$  to  $300 \mu\text{m}$  for a seeding

density of 20.25 g/m<sup>2</sup> (Table CS2). The four dust loads are representative of levels found in the indoor environmental field studies summarized by Boor et al. (2013a).

Lastly, the samples were seeded with a monolayer of 3 μm polystyrene fluorescent particles on the canopy of the existing multilayer deposit (initial seeding density of (mean±SD) 66±20 particles/mm<sup>2</sup>). The canopy layer was used to assess the impact of the multilayer deposit and particle-to-particle contact on the absolute resuspension fraction when compared to the monolayer experiments. The surface and canopy layers were distinguished by the different fluorescent dyes used for the 3 and 10 μm particles. It is important to note that the absolute resuspension fractions are only reported for these two layers and do not represent the total fraction of particles removed from the entire ATD deposit. All multilayer deposits were conditioned at a relative humidity of (mean±SD) 58±3% for 24 hours prior to wind tunnel exposure (value based on laboratory ventilation conditions).

#### *Wind Tunnel Exposure*

Two wind tunnels were used to investigate resuspension from monolayer and multilayer particle deposits. Preliminary experiments for monolayer deposits found no detectable resuspension to occur for 3 and 10 μm particles at velocities below 25 m/s ( $\Phi$  was ~0 between 5 and 20 m/s). To initiate aerodynamic resuspension of 3 and 10 μm particles, velocities greater than 25 m/s were required (also based on findings by Corn and Stein 1965 and Jiang et

al. 2008 for particles near 10  $\mu\text{m}$  in diameter, see Figure CS1 in the Supplemental Information (SI) section). To achieve air velocities of 25 m/s and greater, a high velocity wind tunnel with a turbulent wall jet was designed and built. Computational fluid dynamics was used in the design of the high velocity wind tunnel for the monolayer deposits. The wind tunnel was 20 cm in length, 5 cm wide, and 1.25 cm tall and was constructed with custom laser cut 0.635 cm thick acrylic sheets. In order to generate high velocities above the sample surface, a wall jet was created via a 1 mm by 5 cm rectangular nozzle. The wall jet was found to produce a very uniform discharge over the sample surface and exhibited the characteristic profile for turbulent plane wall jets (Rajaratnam 1976). Additional details on the wind tunnel design can be found in Boor et al. (2011). For the multilayer deposits, where resuspension occurred at much lower air velocities ( $< 25$  m/s), a small-scale wind tunnel was used (Omega Engineering Inc., Model WT-4401-S-110V). Additional information about this wind tunnel and the associated flow characteristics, including velocity and turbulence profiles, can be found in Mukai et al. (2009).

Air velocity measurements were recorded with a one-dimensional constant-temperature hot-wire anemometer (MiniCTA probe 55P16, DanTec Dynamics). The anemometer took velocity measurements at a frequency of 1 kHz, which was necessary to capture the turbulent fluctuations of the high velocity wall jet. For the monolayer deposits, three velocities,  $\bar{U}$ , were studied (mean $\pm$ SD): 25 (actually 24.1 $\pm$ 6.2), 50 (50.0 $\pm$ 11.7), and 75 (73.9 $\pm$ 17.7) m/s (velocity measurements taken at the approximate midpoint of the wall jet,  $\sim$ 1 mm above the surface). The acceleration of the flow was regulated by an automatically

controlled needle valve and was approximately  $2 \text{ m/s}^2$  for each of the three velocities. The turbulence intensities remained roughly the same for each velocity, and were approximately 26%, 23%, and 24% at 25, 50, and 75 m/s, respectively. For the multilayer particle deposits, the velocities studied were (nominally): 2.5, 5, 7.5, 10, 12.5, 15, 20, and 25 m/s; the acceleration was approximately  $5 \text{ m/s}^2$ ; and the average near-surface ( $\sim 1 \text{ mm}$ ) turbulence intensities were approximately 10%.

The exposure time for both the monolayer and multilayer deposits was 100 seconds. The exposure time can be divided into two temporal regimes: a period of flow acceleration, which was typically less than 30 seconds, depending on the final, steady-state velocity, and a period of steady-state flow. A few pilot experiments at low velocities found no significant change in particle resuspension between an exposure time of 10 seconds and 100 seconds, suggesting the majority of resuspension occurs during the acceleration period.



Figure C3 (a.)

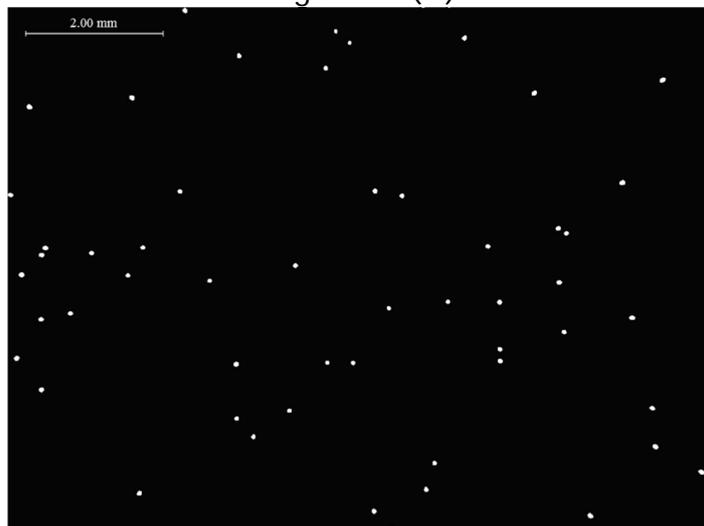


Figure C3 (b.)

Figure C3: Fluorescence stereomicroscope images. (a.) 3  $\mu\text{m}$  particles on galvanized sheet metal, image size: 1.55 x 1.16 mm, (b.) 10  $\mu\text{m}$  particles on galvanized sheet metal, image size: 10.2 x 7.66 mm.

#### *Fluorometric Methods and Morphometry Analysis*

To determine the absolute resuspension fraction,  $\Phi$ , for the monolayer particle deposits and the surface and canopy layers of the multilayer deposit, a fluorescence stereomicroscope (Leica MZ16FA, Leica Microsystems GmbH) equipped with a charge-coupled device (CCD) camera was used. The microscope and camera, along with morphometry analysis, were used to determine the seeding density,  $\sigma$ . The red 3  $\mu\text{m}$  and green 10  $\mu\text{m}$  fluorescent particles were each detected using a different fluorescent filter and microscope and camera settings, as outlined in Table CS1. The microscope was equipped

with a MultiStep bi-directional scan feature through a motorized X/Y stage control that automatically scans a specified area and compiles the individual images into a larger, mosaic image. This allows a greater fraction of the sample area to be further analyzed with morphometry software. The camera settings were modified to obtain images with very high resolution and good contrast between the particle and background surface. As shown in Figure C3, a sparse monolayer deposit was verified by ensuring no particle-to-particle contact existed (Note: the images in Figure C3 are only portions of the full, MultiStep images generated by the microscope for each sample).

To count the number of particles within the area of the MultiStep image, and to determine the seeding density, a morphometric program was developed in the programming software MATLAB (MathWorks Inc.). The grayscale image produced by the microscope camera software (Figure C3) was converted to a binary image of fully saturated white and black objects through a process known as thresholding. This helps isolate individual white objects, which represent individual particles. A threshold value of 5, on the standard grayscale of 0 to 255, was found to sufficiently isolate particles and remove any background noise produced by the inherent fluorescence of the linoleum flooring, galvanized sheet metal, or ATD in the case of the multilayer deposits. A histogram displaying the area distribution (in pixels<sup>2</sup>) was also generated to determine the impact of any outlying objects on the seeding density. Additionally, all microscope images were manually inspected to ensure the white objects were isolated, circular in shape, and fully saturated. The accuracy and precision of the morphometry program was verified by comparing the seeding density with values derived

from manual particle counting, as well as results from another morphometry program, MetaMorph (Molecular Devices, LLC). Lastly, no significant differences in seeding densities were observed between the leading and trailing edges of the sample, suggesting minimal re-deposition of particles to the sample surface (although resuspended particles may have re-deposited further downstream of the sample within the wind tunnel, but this was not evaluated in this investigation).

#### *Uncertainty and Quality Control*

For the monolayer experiments, 8 duplicate tests were performed for each indoor surface at each combination of air velocity and relative humidity (to determine  $\sigma_f$ ). 12 samples from each seeding batch were used to determine  $\sigma_i$ . For the multilayer experiments, 6 duplicate tests for each indoor surface and dust load were performed for a given air velocity (to determine  $\sigma_f$ ). 6 samples from each seeding batch were selected to determine  $\sigma_i$  for the 3  $\mu\text{m}$  canopy layer and 10  $\mu\text{m}$  surface layer, and 6 samples were selected to gravimetrically determine the dust load. Gross counts were used for both  $\sigma_i$  and  $\sigma_f$ , and particles were not tracked individually. To determine the uncertainty in  $\Phi$ , the error in measuring both  $\sigma_i$  and  $\sigma_f$  was propagated. A bias error in the seeding density, based on the microscopy and morphometry analysis, of 5% (based on repeat analyses at different microscope and morphometric settings) was combined with the precision error of the sample population for a given set of conditions. In all results figures, the error bars represent the propagated uncertainty in  $\Phi$ . Samples (approximately 10%) were excluded if the seeding density was 3

standard deviations from the mean, if there were noticeable deformations in the multilayer ATD dust load during the seeding process, or if the sample was improperly handled during wind tunnel exposure or microscopy analysis. Additionally, many trial experiments (data not reported here) were conducted for both the monolayer and multilayer (at approximately 20 g/m<sup>2</sup>) deposits when developing the experimental methodology to ensure adequate repeatability of the seeding methods and wind tunnel exposure.

## RESULTS

Results for both the monolayer and multilayer deposits are shown in Figure 4. Each point on the plot represents the mean of one set of samples at the same conditions and the error bars represent the calculated error in  $\Phi$ .

### *Monolayer Deposits*

Resuspension of 3 and 10  $\mu\text{m}$  particles from monolayer deposits is generally low across all three velocities studied, with  $\Phi$  (mean $\pm$ calculated error) ranging from 0.020 $\pm$ 0.018 to 0.048 $\pm$ 0.043 for 3  $\mu\text{m}$  particles and from 0.039 $\pm$ 0.035 to 0.348 $\pm$ 0.131 for 10  $\mu\text{m}$  particles (range across both surfaces and relative humidities), as shown in Figure C4 (a.) and (b.). As previously discussed,  $\Phi$  was  $\sim$ 0 between 5 and 20 m/s. In general,  $\Phi$  increases as velocity increases (beyond 25 m/s). At both 25 and 50 m/s,  $\Phi$  generally does not exceed 0.10. At 75 m/s,  $\Phi$  is found to increase to levels beyond 0.10 for 10  $\mu\text{m}$  particles, although no significant increase is observed for 3  $\mu\text{m}$  particles. The size-dependence of resuspension is also observed in Figure C4 (a.) and (b.).  $\Phi$  is generally greater for

10  $\mu\text{m}$  particles compared to 3  $\mu\text{m}$  particles at the same conditions.  $\Phi$  is typically greater at 30% relative humidity compared to 75% relative humidity. On average, for 3  $\mu\text{m}$  particles,  $\Phi$  is 1.4 times greater at a relative humidity of 30% when compared to 75%. For 10  $\mu\text{m}$  particles, a moderately stronger dependence on relative humidity is observed, with  $\Phi$  increasing by a factor of 1.7 due to a reduction in relative humidity from 75% to 30%. The type of indoor surface also influenced resuspension from monolayer deposits. On average, for all velocities and at both relative humidities,  $\Phi$  is 1.2 times greater for 3  $\mu\text{m}$  particles on linoleum flooring when compared to galvanized sheet metal and 1.6 times greater for 10  $\mu\text{m}$  particles.

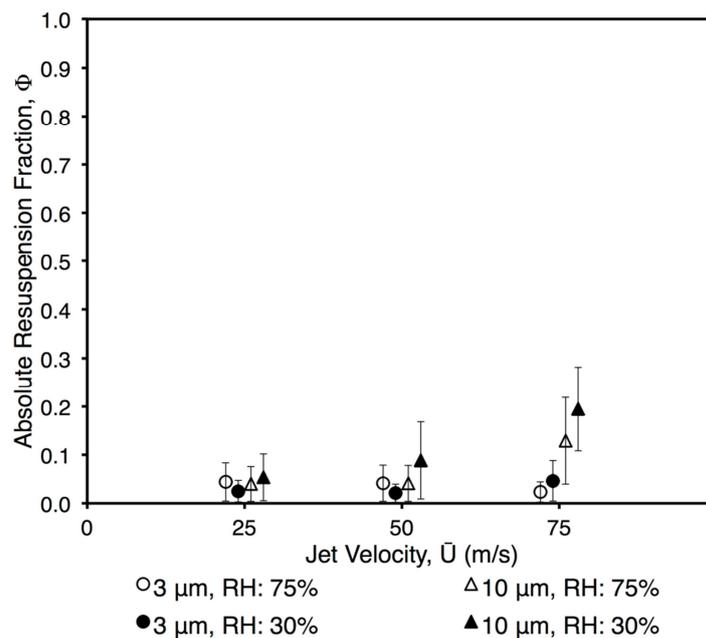


Figure C4 (a.)

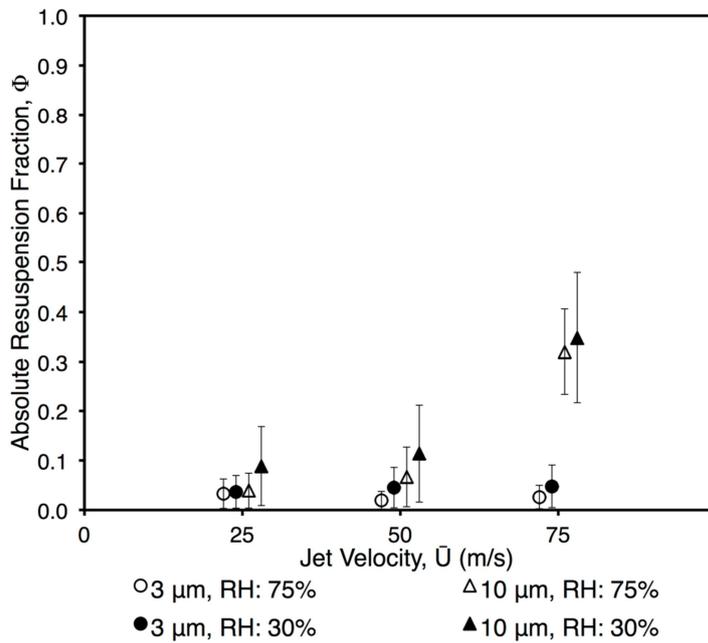


Figure C4 (b.)

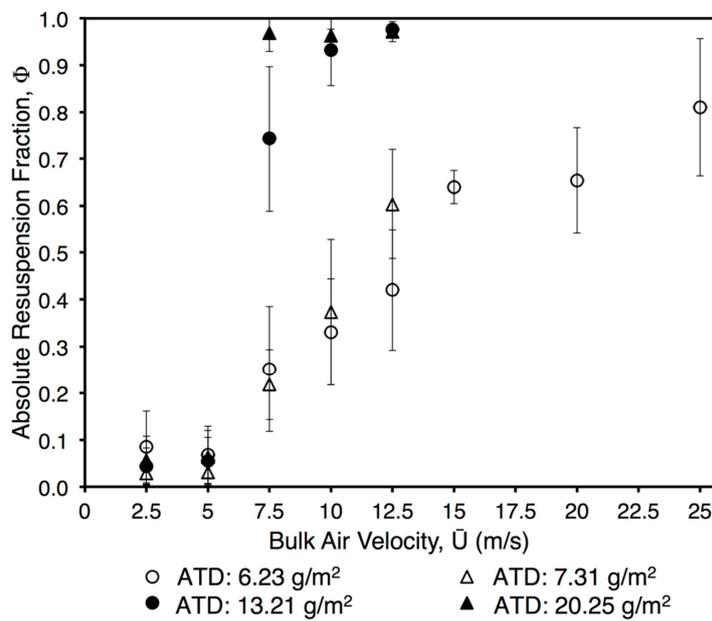


Figure C4 (c.)

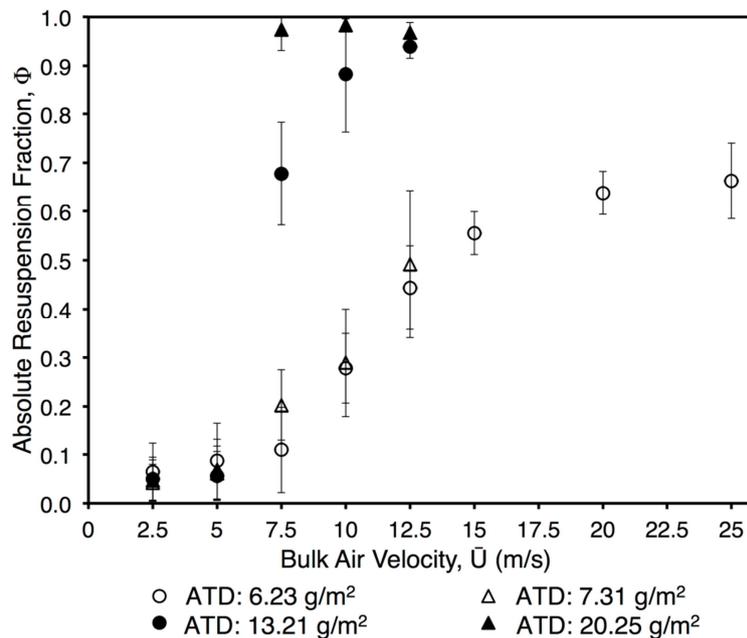


Figure C4 (d.)

Figure C4: Absolute resuspension fractions,  $\Phi$ , for: (a.) monolayer deposit on galvanized sheet metal, (b.) monolayer deposit on linoleum flooring, (c.) 3  $\mu\text{m}$  canopy layer of multilayer deposit on galvanized sheet metal, and (d.) 3  $\mu\text{m}$  canopy layer of multilayer deposit on linoleum flooring.

### *Multilayer Deposits*

Resuspension from multilayer deposits is found to be considerably different than resuspension from monolayer deposits. Numerous variables, including the dust loading, air velocity, the type of indoor surface, and the layer are all found to influence resuspension to varying extents. The results in Figure C4 (c.) and (d.) show that resuspension from the 3  $\mu\text{m}$  canopy layer of multilayer deposits increases as the dust load increases. As with the monolayer deposits,

resuspension from the 3  $\mu\text{m}$  canopy layer generally increases with an increase in velocity. The impact of velocity is strongly coupled with the level of dust loading, with the two heaviest dust loads showing a significant increase in resuspension between 5 and 7.5 m/s, and the two lightest dust loads exhibiting a steady increase in resuspension with increasing velocity from 2.5 to 12.5 m/s. The impact of the surface appears to be small for resuspension from the canopy layer.  $\Phi$  does not vary significantly between galvanized sheet metal and linoleum flooring for all four ATD dust loads. This makes sense given that the primary resuspension layer is the dust layer and not the surface material.

Resuspension is significantly greater for the 3  $\mu\text{m}$  canopy layer of the multilayer deposit compared to the 10  $\mu\text{m}$  surface layer (surface layer results not reported in Figure C4).  $\Phi$  for the surface layer is typically near 0 for all cases studied, with the exception of the lightest loading of 6.23 g/m<sup>2</sup>. For many cases, the 10  $\mu\text{m}$  particles along the surface layer are completely covered by ATD and could not be detected via fluorescence microscopy. Thus,  $\Phi$  could not be accurately determined for many cases and is assumed to be 0 when covered by layers of ATD. For the 6.23 g/m<sup>2</sup> dust load,  $\Phi$  is computed for velocities above 5 m/s. The surface layer  $\Phi$  are: 0.043 $\pm$ 0.002, 0.029 $\pm$ 0.017, 0.051 $\pm$ 0.008, 0.033 $\pm$ 0.002, 0.010 $\pm$ 0.014, and 0.084 $\pm$ 0.048 for 7.5, 10, 12.5, 15, 20, and 25 m/s, respectively (average between both surfaces).

Resuspension occurs at significantly lower velocities for the multilayer deposits compared to the monolayer deposits. For ATD dust loads of 13.21 g/m<sup>2</sup> and 20.25 g/m<sup>2</sup>,  $\Phi$  exceeds 0.883 $\pm$ 0.120 for the canopy layer of 3  $\mu\text{m}$  particles at

10 m/s, whereas for the monolayer deposits, the highest achieved resuspension was for 10  $\mu\text{m}$  particles at 75 m/s ( $\Phi$  of  $0.348 \pm 0.131$  for linoleum flooring at 30% relative humidity). Resuspension from multilayer deposits achieves similar levels at 5 m/s as those observed for monolayer deposits at 50 m/s. The average canopy layer  $\Phi$  across all ATD dust loads at 5 m/s is  $0.061 \pm 0.015$  and the average  $\Phi$  for a monolayer of 3  $\mu\text{m}$  particles is  $0.032 \pm 0.012$  at 50 m/s, and for a monolayer of 10  $\mu\text{m}$  particles, is  $0.078 \pm 0.027$ . Thus, there is an apparent order of magnitude difference in the velocities required to resuspend similar levels of particles from the canopy layer of multilayer deposits and particles from monolayer deposits.

## DISCUSSION

Resuspension was found to be considerably greater for multilayer deposits compared to monolayer deposits. The findings of this investigation can be explained by considering the fundamental differences in the resuspension process between the two types of deposits. A detailed discussion of the unique attributes of resuspension from monolayer and multilayer deposits can be found in Boor et al. (2013a), who summarize findings of the resuspension literature that address the key variables that impact resuspension from both types of deposits. The enhanced resuspension from multilayer deposits compared to monolayer deposits is likely due to reduced particle-to-particle adhesion forces, resuspension in the form of larger aggregates, and possible saltation effects (Bagnold 1941, Fairchild and Tillery 1982, Shao et al. 1993, Matsusaka and Masuda 1996, Lazaridis and Drossinos 1998, Friess and Yadigaroglu 2001, Kurkela et al. 2006, Gac et al. 2008, Gotoh et al. 2011, Kok et al. 2012). Boor et al.

(2013a) also contains a discussion on how the deposit structure and dust load may have important implications for particle resuspension and transport in the indoor environment.

### *Impact of Particle Deposit Height and Viscous Sublayer Thickness on Resuspension*

Resuspension from both monolayer deposits and the canopy layer of multilayer deposits appears to be dependent on the relationship between the particle deposit height,  $\delta$  (as defined in Boor et al. 2013a) and the thickness of the viscous sublayer,  $y_{VSL}$ .  $\delta$  for each of the four ATD dust loads was approximated using the scaling analysis presented in Boor et al. (2013a), along with the known physical properties of ATD, and assuming the deposit has a porosity of 0.75 due to gravitational settling in the seeding chamber. For the monolayer deposits,  $\delta$  is simply equal to the diameter of deposited particles, either 3 or 10  $\mu\text{m}$ . Additionally,  $y_{VSL}$  ( $\mu\text{m}$ ) was approximated for each air velocity studied by applying the scaling relationship presented in Bejan (2004):

$$y_{VSL} \sim C \frac{\nu}{\bar{U}} \quad (\text{C.2})$$

where  $\bar{U}$  is the free stream velocity (m/s),  $\nu$  is the kinematic viscosity of air ( $1.5 \times 10^{-5} \text{ m}^2/\text{s}$  at  $20^\circ\text{C}$ ), and  $C$  is a unit-specific, dimensionless constant ( $\sim 10^8$  for the units above).  $\delta$  for each ATD dust load and monolayer deposit, and  $y_{VSL}$  for all velocities studied, are reported in Tables CS2 and CS3 in the SI section. Although the values for both  $\delta$  and  $y_{VSL}$  are based on simplified approximations, they still provide a good starting point for the following analysis.

Using Equation C.2, the ratio of  $\delta$  to  $y_{VSL}$  was determined for all monolayer and multilayer cases. Figure C5 shows the dependence of  $\Phi$ , for both the monolayer deposits and canopy layer of the multilayer deposits, on the ratio of  $\delta$  to  $y_{VSL}$ . An approximate logistic relationship was observed between  $\Phi$  and  $\delta/y_{VSL}$ . Similar logistic trends can be observed between  $\Phi$  and the bulk air velocity in the monolayer wind tunnel studies of Ibrahim et al. (2003), Ibrahim and Dunn (2006), and Jiang et al. (2008), and between  $\Phi$  and the volumetric airflow rate in the multilayer investigation of Gac et al. (2008). Fitting a logistic curve to all data points in Figure C5, the following empirical relationship between  $\Phi$  and  $\delta/y_{VSL}$  was determined:

$$\Phi\left(\frac{\delta}{y_{VSL}}\right) = \frac{K}{1 + e^{-r\left(\frac{\delta}{y_{VSL}} - \left(\frac{\delta}{y_{VSL}}\right)_0\right)}} \quad (C.3)$$

where  $K$ ,  $r$ , and  $(\delta/y_{VSL})_0$  are parameters determined through a least squares solution in MATLAB (MathWorks Inc.).  $K$  was found to be 0.99,  $r$  to be 3.51, and  $(\delta/y_{VSL})_0$  to be 0.98. The correlation coefficient between the empirical relationship and measured data was found to be 0.938 (along with a root mean square error of 0.12), suggesting a strong, positive correlation between  $\Phi$  and  $\delta/y_{VSL}$ . The purpose of the empirical relationship, presented by Equation C.3, is to help demonstrate that, for the data set presented in our investigation, there is an approximate logistic relationship between resuspension and the ratio of  $\delta$  to  $y_{VSL}$ .

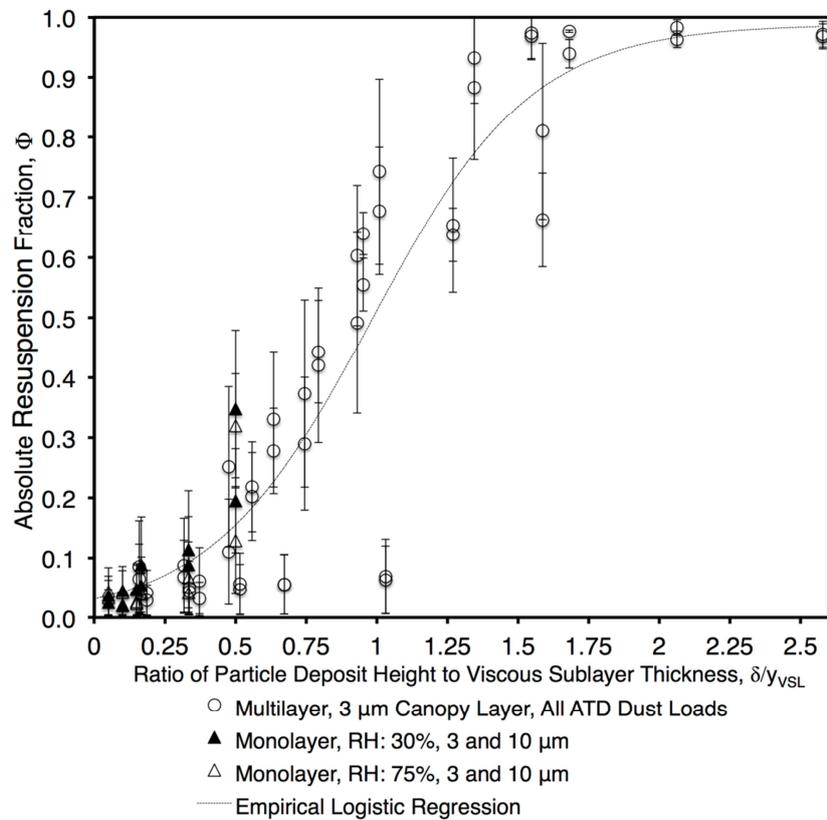


Figure C5: Relationship between the absolute resuspension fraction,  $\Phi$ , of both monolayer deposits and the canopy layer of multilayer deposits with the ratio of  $\delta$  to  $y_{VSL}$ . Additionally, the empirical relationship derived for  $\Phi(\delta/y_{VSL})$  is presented (Equation C.3).

Observing the logistic trend presented in Figure C5,  $\Phi$  grows exponentially to a  $\delta/y_{VSL}$  of unity, after which, its growth rate slows and it begins to asymptotically approach 1. For small values of  $\delta/y_{VSL}$ , minimal resuspension was found to occur. When the particle deposit is completely immersed in the viscous sublayer ( $y_{VSL} \gg \delta$ ), as is the case for monolayer deposits at low velocities, particles will not experience the enhanced removal associated with

frequent collisions with turbulent eddies, and resuspension only occurs due to the periodic penetration of turbulent bursts from the fully turbulent sublayer (Cleaver and Yates 1973, Braaten et al. 1990, Jurcik and Wang 1991). As  $\delta$  begins to approach  $y_{VSL}$ , a sharp increase in  $\Phi$  is observed. As  $\delta/y_{VSL}$  approaches unity (Figure C5), where we observe an inflection point in  $\Phi$ 's dependence on  $\delta/y_{VSL}$ , the edge of  $y_{VSL}$  is at roughly the same height as the particle deposit.  $(\delta/y_{VSL})_0$  is the actual location of the inflection point, 0.98, where  $\Phi \sim 0.5$ ). This was only achieved for the multilayer deposits, as the velocities studied for the monolayer deposits (25 to 75 m/s) were not large enough to reduce  $y_{VSL}$  to a height equivalent to that of the particles (3 and 10  $\mu\text{m}$ ).

Beyond a  $\delta/y_{VSL}$  of unity, where the height of the deposit begins to surpass  $y_{VSL}$  and enter the fully turbulent sublayer, very high levels of resuspension are achieved and  $\Phi$  begins to asymptotically approach 1. We can expect that at increasingly larger values of  $\delta/y_{VSL}$ , due to an increase in bulk air velocity and/or an increase in the dust load,  $\Phi$  will remain near 1 and the canopy layer will be completely resuspended. When increasing fractions of the multilayer particle deposit enter the fully turbulent sublayer, they likely experience the enhanced removal forces associated with turbulent eddies.

The concept of  $\delta/y_{VSL}$  can help explain the role of the level of dust loading on resuspension from multilayer deposits. For velocities between 2.5 and 12.5 m/s,  $\Phi$  was found to increase with dust loading from 6.23 to 20.25 g/m<sup>2</sup>. As dust load increases,  $\delta$  is expected to increase, and therefore, the ratio of  $\delta/y_{VSL}$  for a given air velocity. Gomes et al. (2007) studied resuspension from dust loads of

0.5 and 2.5 g/m<sup>2</sup> and observed more particles to resuspend at the higher dust loading. A wind tunnel study by Nitschke and Schmidt (2010) found resuspension to generally increase with dust loading. Between an exposure time of 3 and 8 seconds,  $\Phi$  increased as the dust load increased from 6.5 to 14 g/m<sup>2</sup> for both steel and PMMA surfaces. The findings of both Gomes et al. (2007) and Nitschke and Schmidt (2010) agree favorably with our investigation and help demonstrate the dependence of resuspension on the dust load, and therefore,  $\delta$ .

The ratio of  $\delta/y_{VSL}$  can also increase due to an increase in the air velocity (decreasing  $y_{VSL}$ ). Our investigation found  $\Phi$  to be strongly dependent on the air velocity and increase with increasing velocity. Fromentin (1989) found a similar trend for heavy multilayer deposits of 100 to 1000 g/m<sup>2</sup> and observed a significant increase in the resuspension flux by increasing the bulk air velocity from 8.5 to 20 m/s. Huang et al. (2005) and Matsusaka and Masuda (1996) observed similar trends in their respective wind tunnel studies.

### *Limitations of the Present Investigation*

The focus of this study was to make direct comparisons of resuspension fractions between monolayer and multilayer deposits on indoor surfaces exposed to a range of air velocities in a wind tunnel, and not necessarily simulate realistic conditions under which resuspension occurs in the indoor environment. This investigation highlighted the important role of the type of particle deposit on aerodynamic resuspension from indoor surfaces, although it is important to discuss several limitations of this research. The wind tunnel resuspension

experiments focused solely on aerodynamic-induced resuspension, whereby the airflow was accelerated at a nearly constant rate to some steady state velocity. For flow in ventilation ducts, it would be expected that resuspension occurs primarily due to aerodynamic removal forces, although vibrational forces may be present. Particle deposits may be exposed to periods of high acceleration when a ventilation system cycles on and off, depending on the nature of the fan speed control. In addition, the turbulence of the airflow may vary considerably over different ventilation duct elements, such as duct bends and irregularly shaped flex-duct. For resuspension from flooring, airflow associated with the downward foot motion is likely very impulsive with high acceleration (Khalifa and Elhadidi 2007). Resuspension due to human activity may also generate additional removal forces, such as mechanical forces, due to surface vibrations, and electrostatic forces associated with the walking process (Gomes et al. 2007, Hu et al. 2008, Qian and Ferro 2008). The impact of airflow acceleration, turbulence levels, and additional removal mechanisms were not considered in this investigation, although these variables may have significant impact on resuspension under real conditions in the indoor environment.

The spherical fluorescent particles used in this investigation do not necessarily represent particles found in the indoor environment, which may vary considerably in their shapes, surface characteristics, and material compositions. However, they provide a basis to compare resuspension between two types of particle deposits. As such, the resuspension fractions provided in this investigation are used solely to compare monolayer and multilayer deposits, and do not necessarily represent resuspension fractions of actual indoor particles

from indoor surfaces. It would be expected that resuspension may increase for irregularly shaped particles, such as spores, which have reduced contact area, and likely reduced adhesion, with deposition surfaces relative to spherical particles (e.g. Wu et al. 1992, Goldasteh et al. 2012a). Additionally, indoor particle deposits have a wide range of polydisperse size distributions (see Boor et al. 2013a), which may also influence resuspension.

#### SUMMARY

An experimental methodology was developed to determine aerodynamic resuspension from both monolayer and multilayer deposits on indoor surfaces. Resuspension was found to be strongly dependent on the type of particle deposit, with significantly greater levels of resuspension observed from multilayer deposits compared to monolayer deposits. Resuspension fractions at an air velocity of 5 m/s for the canopy layer of multilayer deposits were similar to those found for monolayer deposits at 50 m/s. Additionally, for monolayer deposits, resuspension fractions increased with increasing particle size and air velocity, and for multilayer deposits, resuspension fractions for the canopy layer increased with increasing dust load and air velocity. Relative humidity, the type of indoor surface, and layer location were also found to influence resuspension. Through scaling analysis, a relationship was found between the particle deposit height and viscous sublayer thickness that can help explain why elevated resuspension was observed from multilayer deposits compared to monolayer deposits. Other unique attributes of multilayer resuspension, including reduced particle-to-particle adhesion, resuspension in the form of larger aggregates,

saltation effects, and deposition structure and porosity can help explain the enhanced resuspension that was observed. Future work should consider the impact of other removal mechanisms and airflow and environmental parameters on resuspension from multilayer deposits.

#### AUTHOR CONTRIBUTIONS

B.E.B., J.A.S., and A.N. defined the overall scope of the study and developed the experimental methods. B.E.B. conducted the experiments and analyzed the data. B.E.B. wrote the paper. J.A.S. and A.N. provided guidance in the execution of the experiments, advised in the interpretation of the results, and provided detailed comments on draft manuscripts.

#### SUPPORTING INFORMATION

Table CS1: Fluorescence stereomicroscope, camera, and morphometry settings

Particle Diameter ( $\mu\text{m}$ )	Dye/Filter	Excitation Wavelength (nm)	Emission Wavelength (nm)	Zoom Drive	Focus Drive	Exposure (s)	Gamma	Gain	Image Area ( $\text{mm}^2$ )	Image Threshold
3	Red/TXR	542	612	72.5	88-90	0.18-1.2	10	10	1.8	25
10	Green/GFP3	468	508	10-11	90-91	1.2-1.5	10	10	55-80	25

Table CS2: Approximation of the particle deposit height,  $\delta$ , and viscous sublayer thickness,  $y_{VSL}$ , for multilayer deposit experiments of varying dust load,  $m_0$

$m_0$ ( $\text{g}/\text{m}^2$ )	$\delta^*$ ( $\mu\text{m}$ )	$\bar{U}$ (m/s)	$y_{VSL}$ ( $\mu\text{m}$ )
6.23	95	2.5	600
		5	300
7.31	112	7.5	200

13.21	202	10	150
		12.5	120
		15	100
20.25	309	20	75
		25	60

+: Determined using Equation 2 of Boor et al. (2013). ATD mass median diameter of 4.5  $\mu\text{m}$  and bulk density of 500  $\text{kg}/\text{m}^3$ . Assumed porosity of 0.75 due to gravitational settling in the seeding chamber.

Table CS3: Approximation of the particle deposit height and viscous sublayer thickness for monolayer deposit experiments

$D$ ( $\mu\text{m}$ )	$\delta^+$ ( $\mu\text{m}$ )	$\bar{U}$ (m/s)	$y_{VSL}$ ( $\mu\text{m}$ )
3	3	25	60
		50	30
10	10	75	20

+: For monolayer deposits,  $\delta$  is equal to the diameter of the deposited particles,  $D$

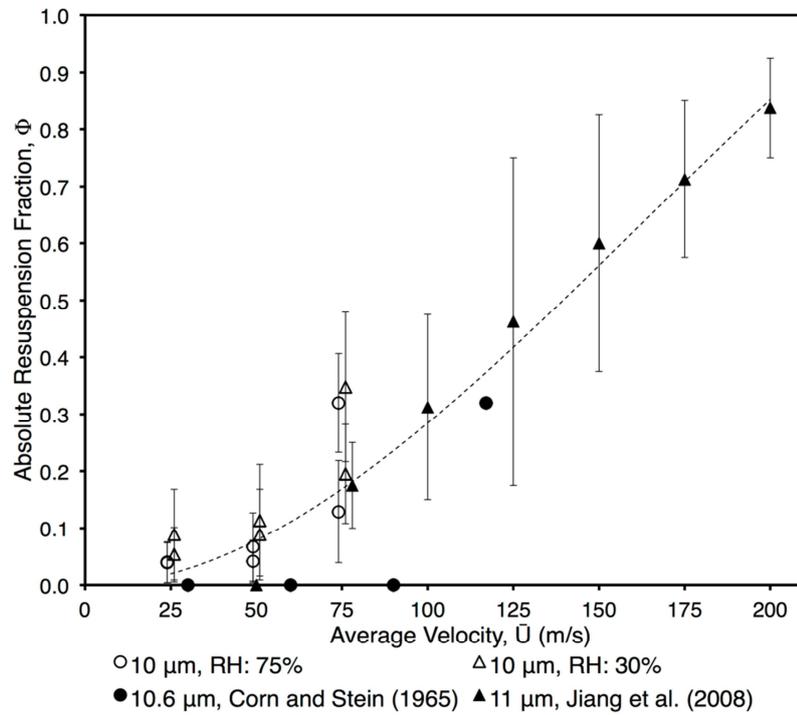


Figure CS1: Comparison of monolayer deposit results for 10  $\mu\text{m}$  particles from both galvanized sheet metal and linoleum at 30% and 75% RH with monolayer deposit results from Corn and Stein (1965) (10.6  $\mu\text{m}$  glass particles at 35% RH) and Jiang et al. (2008) (11  $\mu\text{m}$  particles at 35-50% RH; approximate mean, and upper and lower bounds across range of surface roughness). Note: dashed line is added for visual representation of trend among data points.

## Appendix D

## Paper D. Characterizing Particle Resuspension from Mattresses: Chamber Study

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### ABSTRACT

People spend approximately one-third of their lives sleeping, where they can be exposed to a myriad of particle-bound biological agents and chemical pollutants that originate within mattresses and bedding, including allergens, fungal spores, bacteria, and particle-phase semi-volatile organic compounds. Full-scale particle resuspension experiments were conducted in an environmental chamber, where volunteers performed a prescribed movement routine on an artificially seeded mattress. Human movements in bed, such as rolling from the prone to supine position, were found to resuspend settled particles, leading to elevations in airborne particle concentrations. Resuspension rates were estimated for the size fractions of 1 to 2  $\mu\text{m}$ , 2 to 3  $\mu\text{m}$ , 3 to 5  $\mu\text{m}$ , 5 to 10  $\mu\text{m}$ , and 10 to 20  $\mu\text{m}$ , and were in the range of  $10^{-3}$  to  $10^1 \text{ h}^{-1}$ . Particle size had the most significant impact on the resuspension rate, whereas dust loading, volunteer body mass, and ventilation rate had a much smaller impact. Resuspension increased with the intensity of a movement, as characterized by surface vibrations, and decreased with repeated movement routines. Inhalation exposure was characterized with the intake fraction metric. Intake fractions increased as the particle size and ventilation rate decreased and ranged from  $10^2$  to  $10^4$  inhaled particles per million resuspended, demonstrating that a significant fraction of released particles can be inhaled by sleeping occupants.

## PRACTICAL IMPLICATIONS

Full-scale chamber experiments with human volunteers demonstrate that body movements in bed can resuspend settled particles from mattresses, leading to elevated airborne particle concentrations in both the breathing zone and bulk air of the chamber. Numerous variables influence resuspension, including particle size and intensity of a specific body movement. The results suggest that human-induced resuspension in the sleep microenvironment may play an important role in contributing to our inhalation exposure to mattress dust pollutants.

## NOMENCLATURE

$a$ : Chamber ventilation rate ( $\text{h}^{-1}$ )

$A_M$ : Seeded mattress (top surface) surface area ( $\text{m}^2$ )

APS: Aerodynamic particle sizer

ATD: Arizona Test Dust

BZ: Breathing zone

$C_{Average}$ : Average particle number concentration at a given sampling location

$C_{i,Bulk Air}(t)$ : Particle number concentration in the bulk chamber air as measured at the bulk air OPC sampling location (# particles/ $\text{m}^3$ )

$C_{i,Bulk Air, Clean}(t)$ : Particle number concentration in the bulk chamber air as measured at the bulk air OPC sampling location during the clean set (# particles/ $\text{m}^3$ )

$C_{i, BZ}(t)$ : Spatially-averaged BZ particle number concentration, average of BZ right, BZ left, and BZ middle OPC sampling locations (# particles/ $\text{m}^3$ )

$C_{i, BZ, clean}(t)$ : Spatially-averaged BZ particle number concentration, average of BZ right, BZ left, and BZ middle OPC sampling locations, during the clean set (# particles/ $\text{m}^3$ )

$\overline{C_{iBZSet}}$  and  $\overline{C_{iBZDecay}}$ : Time-averaged & spatially-averaged BZ particle number concentration, average of BZ right, BZ left, and BZ middle OPC sampling locations, during the movement set and decay period, respectively (# particles/ $\text{m}^3$ )

$iF_i$ : Intake fraction (part per million, ppm)

$\overline{iF_i}$  or  $iF_{Average}$ : Time-averaged intake fraction (part per million, ppm)  
 $k_i$ : Deposition rate ( $h^{-1}$ )  
 $L_{i,0}$ : Initial mattress dust loading (# particles/ $m^2$ )  
 $L_i(t)$ : Continuous mattress dust loading during movement set (# particles/ $m^2$ )  
 $\overline{L_i}$  : Time-averaged dust loading during movement set ( $h^{-1}$ )  
 OPC: Optical particle counter  
 $Q_B$ : Volumetric breathing rate ( $m^3/h$ )  
 RR: Resuspension rate ( $h^{-1}$ )  
 $RR_i(t)$ : Resuspension rate during movement set ( $h^{-1}$ )  
 $\overline{RR_i}$  or  $RR_{Average}$ : Time-averaged resuspension rate during movement set ( $h^{-1}$ )  
 $RR_{Movement}$ : Time-averaged resuspension rate for each individual movement ( $h^{-1}$ )  
 $\Delta t$ : Time-step (sampling interval of particle sampling instrument) (s)  
 $\Delta t_{Set}$  or  $t_{Set}$  and  $t_{Decay}$  : Duration of movement set and decay period, respectively (min)  
 $V_C$ : Chamber volume ( $m^3$ )

## INTRODUCTION

The sleep microenvironment is an important, yet understudied, indoor space. It can be defined as the space encompassing a mattress, pillows, bedding, and volume of air above these items that includes both an individual's breathing zone (BZ) and thermal plume. There are several defining attributes of this microenvironment that make it unique from both an indoor air quality and exposure/health perspective, including: significant exposure period, roughly equating to one-third of our lives; diversity of pollutants and pollutant sources; and potential for elevated exposures due to the source-proximity effect.

Humans spend a considerable amount of time sleeping. According to the U.S. Environmental Protection Agency's (EPA) Exposure Factors Handbook (EFH) data set (EPA Activity ID = 14500, sleep or nap activity), the average sleep

duration for the U.S. mean age group of 37 years (U.S. Census Bureau 2009) is 8.2 hours/day, with longer sleep periods for infants, children, and the elderly. Applying the U.S. EPA EFH data set to the National Human Activity Pattern Survey (NHAPS) study conducted by Klepeis et al. (2001), adults spend about 34% of their day in the sleep microenvironment, which equates to about 50% of the time they spend in a residence, and 39% of the time they spend indoors. The magnitude of the sleep exposure period makes the sleep microenvironment particularly important in contributing to both our acute and chronic exposures to various pollutants originating in mattresses and bedding.

The seemingly innocuous sleep microenvironment can be home to a diversity of pollutants that have shown to adversely affect the health of the human body. Mattresses are possible sources of a myriad of chemical species, such as volatile organic compounds (VOCs), plasticizers, flame retardants, and unreacted isocyanates (Stapleton et al. 2011, Boor et al. 2014). Furthermore, mattresses, pillows, and bedding serve as an accumulation zone for a diverse spectrum of particulate pollutants, many of which are of biological origin.

Biological matter in mattress dust consists of a wide range of organisms and their associated allergens. House dust mite allergens (Der p 1, Der f 1, Der p2, and Blo t 5), along with cat (Fel d 1), dog (Can f 1), and cockroach (Bla g 2) allergens have been detected in mattress dust, with concentrations ranging over several orders of magnitude, from  $< 1$  to  $> 10^3$   $\mu\text{g/g}$  of mattress dust (Su et al. 2001, Mahrshahi et al. 2002, Instanes et al. 2005, Wu et al. 2009, Leung et al. 2011).

A variety of fungal genera and species are commonly found in mattress dust, including *Penicillium* spp., *Cladosporium* spp., *Aspergillus fumigatus* spp., *Alternaria* spp., *Eurotium* spp., *Epicoccum*, among many others (Jovanovic et al. 2004, Hicks et al. 2005, Woodcock et al. 2006, Vogel et al. 2008, Begum et al. 2012). Mattresses are an ideal fungal culture medium, given the high moisture levels (humans produce ~ 100 L of sweat in bed per year) and elevated surface temperatures around a sleeping human (~ > 30°C) (Woodcock et al. 2006). Concentrations are typically in the range of 10<sup>3</sup> to 10<sup>4</sup> colony forming units/g of mattress dust.

The sleep microenvironment is also home to an array of bacterial phyla and genera, many of which are associated with human origin (skin, oral, intestinal/fecal, and genital) and specifically, the shedding of human skin (e.g. Täubel et al. 2009, Hospodsky et al. 2012). Bacteria identified in mattress dust include: *Staphylococcus*, *Lactobacillus*, *Streptococcus* sp., *Lactococcus*, *Bacillus* sp., *Listeria* spp., *Zoogloea* sp., *Corynebacterium tuberculostearicum*, *Moraxella* sp., and *Staphylococcus sciuri* sp., among a host of others (Korthals et al. 2008, Täubel et al. 2009, Ege et al. 2012). Bacterial endotoxin (the biologically active lipopolysaccharide (LPS) of gram-negative bacteria) levels are typically in the range of 10<sup>3</sup> to 10<sup>5</sup> endotoxin units/g of mattress dust. Mattress dust may also contain significant quantities of skin cells, as the human body sheds about 5 x 10<sup>8</sup> skin cells per day (Weschler et al. 2011).

In addition to particles of biological origin, semi-VOCs (SVOCs) originating in the mattress foam or elsewhere in a residence, may also

accumulate in mattress dust due to their low volatility and high molecular weight. Phthalate plasticizers have been detected at levels of 10 to  $10^2$   $\mu\text{g/g}$  of mattress dust (Hsu et al. 2012), and brominated and organophosphate flame retardants have been detected at levels of  $< 1$  to  $10^3$   $\text{ng/g}$  mattress dust (Ali et al. 2012).

Despite extensive research to characterize particulate pollutants in mattress dust, there is a paucity of research on how these pollutants are removed from mattress dust deposits and transported within the vicinity of the human body in bed. The physical process of settled particles detaching from a surface and becoming airborne through application of various aerodynamic and mechanical removal forces is referred to as resuspension. Previous field and laboratory experimental research has shown that human activities, such as walking, can induce resuspension of settled dust from indoor surfaces (Thatcher and Layton 1995, Karlsson 1999, Ferro et al. 2004a, Ferro et al. 2004b, Qian and Ferro 2008, Qian et al. 2008, Tian et al. 2014, You and Wan 2014). Resuspension can be influenced by numerous factors, such as characteristics of the particle deposit and deposition surface, airflow dynamics, environmental conditions, and intensity of the human activity (see reviews by Boor et al. 2013a and Qian et al. 2014). It is likely that human movements in bed can resuspend mattress dust particles, thereby serving as a source mechanism for the various biological and organic particle-phase pollutants.

Human activities in bed can occur during periods of wakefulness and throughout the extended sleep state. Although the absence of voluntary motor

behavior is a characteristic of the sleep state, movements are commonly reported, ranging from about ten significant body posture shifts (e.g. Aaronson et al. 1982), such as rolling from the supine to prone position, to several hundred smaller body movements (e.g. Azumi et al. 1977). Body movement frequencies can range from  $< 0.1$  to 1 body movements per sleep minute, depending on sleep stage and age, among other factors (Wilde-Frenz and Schulz 1983, Shimohira et al. 1998, Giganti et al. 2008).

Elevated inhalation exposures to resuspended particle-phase mattress dust pollutants can occur due to the source-proximity effect, in which pollutant concentrations near a source are greater than those in the bulk air of a room. The source-proximity effect may be influenced by a number of factors, such as the spatial proximity of a sleeping person's BZ to the source, incomplete mixing of bedroom air, concentration gradients near an actively emitting source, the personal cloud due to human-induced particle resuspension, and the buoyant human thermal plume (Mage and Ott 1996, Wallace 1996, McBride et al. 1999, Rim and Novoselac 2009 and 2010, Laverge et al. 2013). Laverge et al. (2013) simulated the release of gaseous pollutants from an adult mattress with an inert tracer gas and found the BZ concentrations for an adult thermal manikin to be significantly greater than those measured in the bulk air, typically by a factor of 1.1 to  $> 2$ .

The primary aim of this investigation is to fill knowledge gaps in the literature, particularly as related to human-induced particle resuspension from mattresses and bedding. The impact of particle size, mattress dust load,

volunteer body mass, ventilation rate, repeated movement sets, and movement intensity on human-induced resuspension from mattresses were quantified.

## MATERIALS AND METHODS

### *Experimental Design*

Full-scale experiments were conducted in an environmental chamber with ten human volunteers. A detailed experimental matrix is presented in Table DS1 of the Supporting Information (SI) section. Mattress dust loads are typically in the range of 0.1 to > 1.0 g/m<sup>2</sup>, as reported in field measurements by Gehring et al. (2004), Chen et al. (2007), Giovannangelo et al. (2007), Rennie et al. (2008), Tischer et al. (2011), and Wu et al. (2012). To represent this range in mattress dust loading, two dust loads were examined: 0.1 and 1.0 g/m<sup>2</sup>. For all experiments, volunteers were instructed to perform a prescribed movement routine on an artificially seeded twin-size coil mattress. Airborne particle number concentrations were measured in both the bulk chamber air and volunteer's BZ. Twenty-six experiments were conducted (with 10 volunteers), each with two-repeated movement routine sets, for a total of fifty-two sets. Although in this investigation only a wrapped mattress was studied, a complementary study by Spilak et al. (2014) examined the impact of pillows, blankets, and bedding arrangements on resuspension.

### *Particle Deposit Generation*

The test mattress was artificially seeded in a 2.8 m<sup>3</sup> seeding chamber. The seeding chamber was constructed with styrofoam panels mounted to a wood

frame and was internally lined with grounded aluminum foil to reduce deposition to the chamber walls. The mattress was first wrapped in two layers of 225-thread count bed sheets (60% cotton, 40% polyester) in order to reduce contamination of the mattress, which was re-used for each experiment. Before seeding, the bed sheets were washed in a standard wash cycle with detergent and then air-dried for at least 48 hours. The wrapped mattress was positioned at the bottom of the seeding chamber, where it was seeded with a deposit of polydisperse (1 to 20  $\mu\text{m}$ ) ISO 12103-1 A1 Ultrafine Arizona Test Dust (ATD) (Powder Technology Inc.) (Figure D1). ATD has been used in full-scale resuspension experiments and was selected for its known size distribution and ease of generation. ATD also offers insight into the resuspension of actual mattress dust particles that are of similar size, including: mite and animal allergen particles (< 1 to > 20  $\mu\text{m}$ , e.g. O'Meara and Tovey 2000, Chang and Gershwin 2004), fungal spores (1 to 4  $\mu\text{m}$ , Reponen et al. 2001), fungal fragments (< 1  $\mu\text{m}$ , Reponen et al. 2007), bacteria (1 to > 7  $\mu\text{m}$ , Górný et al. 1999, Meklin et al. 2002), and skin flakes (40 x 30 x 2  $\mu\text{m}$ ).

The ATD was contained within four aerosolizing canisters positioned at the top of the seeding chamber and was aerosolized with pressurized air (described in further detail in Boor et al. 2011 and 2013b). Six small mixing fans were used to improve mixing conditions and the uniformity of particle deposition to the mattress. The mattress remained in the seeding chamber for a 24 h conditioning period to ensure deposition of the entire size distribution of the ATD. Relative humidity was recorded with a HOBO data logger (Model U12-012, HOBOware Pro, Onset Computer Co.) and remained in the range of 52-59%

across all experiments. Deposited particles were collected on nine microscope slides (25 x 75 mm) distributed across the mattress surface. The ATD loading on the microscope slides was then measured gravimetrically with an analytical balance (Model AB135-5, Mettler-Toledo International Inc.). The resulting dust loads for each experiment for are presented in Table DS1 and were typically within 10-20% of the nominal value.

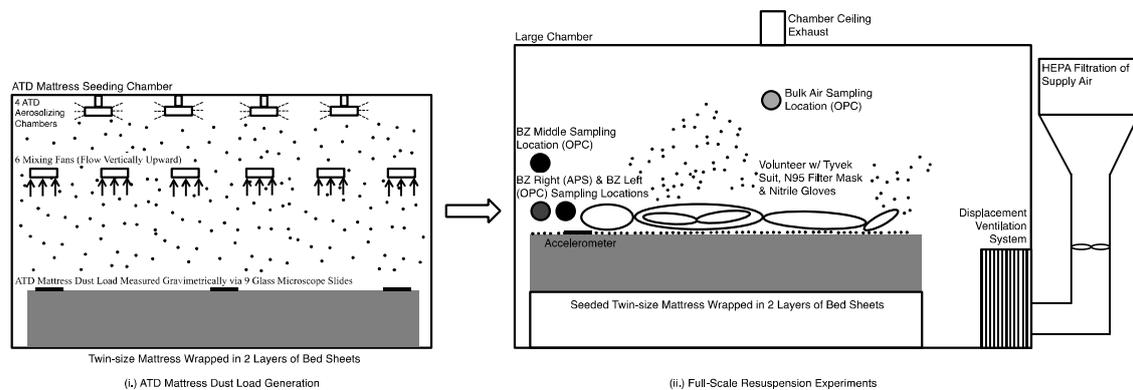


Figure D1: Mattress seeding process and large chamber setup for full-scale resuspension experiments.

### Chamber Configuration and Instrumentation

Resuspension experiments were performed in a 14.75 m<sup>3</sup> stainless steel chamber. Influent air was filtered with an in-duct HEPA filter and supplied via a displacement ventilation diffuser. The chamber remained positively pressurized. The test mattress was placed above a box spring and steel frame, located approximately at the center of the chamber. A three-axis linear accelerometer (10 Hz sampling frequency, Model LIS302DL, STMicroelectronics) was mounted to the wrapped mattress surface, near the volunteer's head region, to monitor

surface vibrations. Air velocities above the bedding surface were also recorded in several additional experiments (measured with 5 Hz omnidirectional anemometers, Model HT-400, Sensor Electronics).

Four particle sampling instruments were positioned throughout the chamber, as shown in Figure D1. One optical particle counter (OPC, 0.05 Hz sampling frequency over which concentration is averaged, AeroTrak Model 8220, TSI Inc.) (“bulk air”) was positioned at 0.83 m above the mattress surface, at the approximate mid-point between the mattress surface and the chamber exhaust. This position was used to approximate the particle concentration in the bulk chamber air (Figure D1). To measure the BZ particle concentration, two OPCs and one aerodynamic particle sizer (APS, 1 Hz, Model 3321, TSI Inc.) were used, as shown in Figure D1. The APS (“BZ right”) and one OPC (“BZ left”, 0.1 Hz, AeroTrak Model 9306, TSI Inc.) represent the approximate height of the BZ of an occupant lying in the prone position or on their side (2.5 cm above the mattress surface). Another OPC (“BZ middle”, 0.1 Hz, AeroTrak Model 9306, TSI Inc.) was positioned 25 cm above the mattress surface to represent the approximate BZ height for an occupant lying in the supine position. To address the variability in the particle size classification by the three OPCs, the instruments were corrected against the APS in co-location measurements using aerosolized ATD and correction factors were applied on a size-resolved basis. The particle concentrations for all sampling instruments were divided into five particle size fractions: 1-2, 2-3, 3-5, 5-10, and 10-20  $\mu\text{m}$ , corresponding to the size distribution (and size fractions) of the ATD.

### *Volunteer Movement Routine and Experimental Sequence*

Each volunteer was instructed to perform a routine of five movements on the mattress, designed to represent common movements of varying intensities that an occupant may perform while in bed. Upon entering the chamber, the volunteer performed the following routine (Figure D2): M1, sit on mattress (then hold still in position for 2.5 min.); M2, lay in the supine position (then hold still in position for 2.5 min.); M3, full 360° rotation to supine position (then hold still in position for 2.5 min.); M4, lay in prone position (then hold still in position for 2.5 min.); and M5, lay in supine position (then hold still in position for 2.5 min.). The entire movement set lasted 12.5 minutes. All volunteers wore a Tyvek clean suit outfit, with booties and a hood (Model TY122 S, DuPont™), filter mask (OSHA & NIOSH N95 rating, Model 8210, 3M™), and Nitrile gloves. The outfit was selected to protect the volunteer from particle exposure and to prevent the volunteer from acting as a source of particles.

The movement routine was repeated three times for each resuspension experiment, as outlined in the experimental sequence, Figure D3. First, the volunteer performed the routine on an unseeded mattress, referred to as the “clean set,” to evaluate background particle concentrations. The seeded wrapped mattress was then carefully placed in the chamber. Background particle concentrations were allowed to decay for a period of 30 minutes. The volunteer then performed two sets of the movement routine, “set 1” and “set 2,” which are separated by a 30 minute decay period. Set 2 offers insight into how resuspension changes in time after numerous movements on the seeded mattress. Set 2 was followed by an hour-long decay period.

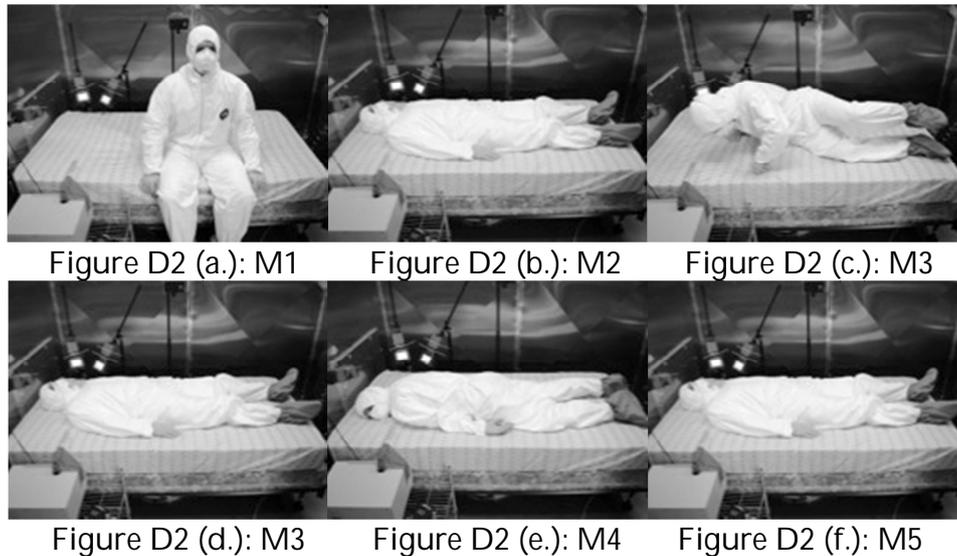


Figure D2: Volunteer movement routine, (a.) movement 1 (M1): sit on mattress, (b.) movement 2 (M2): lay in supine position, (c.-d.) movement 3 (M3): 360° rotation to supine position, (e.) movement 4 (M4): lay in prone position, and (f.) movement 5 (M5): lay in supine position.

Figure 3 illustrates the characteristic particle number concentration profile throughout the entire experimental sequence (Note: particle number concentration is presented on logarithmic scale and is a schematic, not actual data). Short-term concentrations peaks were observed at the commencement of each movement, M1-M5, during the clean set (suggesting the resuspension of residual particles and zeolite particles (originating in the detergent) on the bedding), set 1, and set 2. Throughout each movement set, a gradual elevation in the particle concentration was observed. The baseline particle concentration continued to increase until the cessation of the movement set, where it was then followed a decay period. The particle concentration profile and experimental sequence can be interpreted as what might occur as an occupant gets into bed

(M1), re-positions themselves as they attempt to fall asleep (M2-M5), and then lays still as they enter their sleep cycle (decay periods).

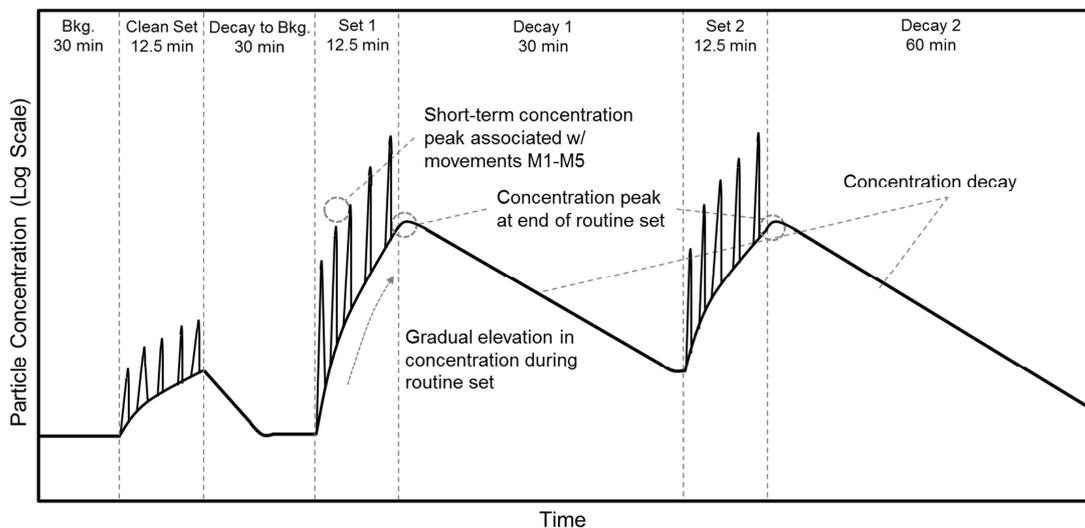


Figure D3: Characteristic particle number concentration profile during the experimental sequence (Note: figure is for illustrative purposes and does not present actual data).

### Resuspension Rate Estimate

Resuspension can be quantified through the resuspension rate ( $RR$ ,  $h^{-1}$ ) metric, defined as the fraction of surface species removed per unit time (Slinn 1978):

$$RR = \frac{\text{resuspension flux}}{\text{surface concentration}} \quad (D.1)$$

$RR$  has been widely used by others for indoor particles (Thatcher and Layton 1995, Gomes et al. 2007, Qian and Ferro 2008, Oberoi et al. 2010, Raja et al. 2010, and Shaughnessy and Vu 2012).

For this investigation,  $RR$  is determined by applying a two-compartment mass balance (e.g. Schneider et al. 1999, Qian and Ferro 2008) to model the particle number concentration for each size fraction,  $i$ :

$$V_C \frac{dC_{i,BulkAir}(t)}{dt} = RR_i(t)L_i(t)A_M - aV_C C_{i,BulkAir}(t) - k_i V_C C_{i,BulkAir}(t) \quad (D.2)$$

$$A_M \frac{dL_i(t)}{dt} = k_i V_C C_{i,BulkAir}(t) - RR_i(t)L_i(t)A_M \quad (D.3)$$

The following expression can then be derived from Equations D.2 and D.3:

$$A_M \frac{dL_i(t)}{dt} = -V_C \left[ \frac{dC_{i,BulkAir}(t)}{dt} + aC_{i,BulkAir}(t) \right] \quad (D.4)$$

The continuous mattress dust loading,  $L_i(t)$ , can be estimated by integrating Equation D.4 from time  $t = 0$  to  $t$ :

$$L_i(t) = L_{i,0} - \frac{V_C}{A_M} \left[ (C_{i,BulkAir}(t) - C_{i,BulkAir,Clean}(t)) + a \int_{t_0}^t (C_{i,BulkAir}(t) - C_{i,BulkAir,Clean}(t)) dt \right] \quad (D.5)$$

where  $L_{i,0}$  is the initial mattress dust loading. The initial dust loading for each size fraction was estimated using the size distribution provided by the ATD manufacturer. The dust loading at the end of set 1 was used as the initial dust loading for set 2. The bulk air concentration during the clean set is subtracted from the bulk air concentration during sets 1 and 2 at the same time  $t$  during the routine. The concentration during the clean set, which remained nearly one to two orders of magnitude lower than that during Sets 1 and 2, includes

contributions from non-ATD resuspension (e.g. bedding fibers and zeolite particles in the detergent), infiltration, and filter by-pass. As such, this correction ensures the bulk air concentration represents resuspended ATD particles as best possible. The integral in Equation D.5 was estimated using Simpson's rule.

Knowing  $L_i(t)$  and  $C_{i, Bulk Air}(t)$ ,  $RR_i(t)$  can be determined using a numerical forward difference approximation with a time-step of  $\Delta t = 20$  seconds (sampling interval of bulk air OPC):

$$RR_i(t + \Delta t) = \frac{V_C}{A_M L_i(t)} \left[ \frac{C_{i, Bulk Air}(t + \Delta t) - C_{i, Bulk Air}(t)}{\Delta t} + (a + k_i)(C_{i, Bulk Air}(t) - C_{i, Bulk Air, Clean}(t)) \right] \quad (D.6)$$

As with the  $L_i(t)$ , the bulk air concentration during the clean set is subtracted from the bulk air concentration during sets 1 and 2. The time-averaged dust loading and resuspension rate was then determined for the entire 12.5 minute routine ( $\Delta t_{Set} = 12.5$  min.) for sets 1 and 2, along with the time-averaged resuspension rate for the duration of each individual movement, M1-M5.

In the above analysis, ATD resuspension is assumed to be the only particle source (after correcting for the clean set concentration). It is assumed that there is negligible track-in of ambient particles. The chamber ventilation rate,  $a$ , was measured via CO<sub>2</sub> decay, and values are listed in Table DS1 for each experiment. The deposition rate was determined by calculating the loss rate ( $a + k_i$ ) from the final 1 hour decay period. The average deposition rates for each size fraction were: 1-2  $\mu\text{m}$ :  $4.4 \times 10^{-5}$ , 2-3  $\mu\text{m}$ :  $1.6 \times 10^{-4}$ , 3-5  $\mu\text{m}$ :  $3.3 \times 10^{-4}$ , 5-10  $\mu\text{m}$ :  $7.3 \times 10^{-4}$ , and 10-20  $\mu\text{m}$ :  $1.2 \times 10^{-3} \text{ s}^{-1}$ . These values are consistent with those reported in

the literature (Byrne et al. 1995, Thatcher et al. 2002, Lai et al. 2002). It is assumed that the deposition rates remain the same for the decay period, with no volunteer present, and for the movement sets, with a volunteer present.

### *Exposure Estimate*

In the sleep microenvironment, an individual is positioned in very close proximity to the source (settled dust on mattress) with their BZ only centimeters above the mattress surface. Figure DS1 shows the average particle concentrations at each of the sampling locations, bulk air, BZ right, BZ left, BZ middle, along with the spatial BZ average (mean of three BZ sampling locations). The BZ concentrations offer a starting point to estimate an occupant's exposure to resuspended particles.

Occupant exposure was characterized with the intake fraction,  $iF$ , metric. The  $iF$  for a pollutant is defined as the total mass inhaled per unit mass released from a source and can be expressed on a part per million basis (ppm) (Bennett et al. 2002, Marshall and Nazaroff 2007). Here,  $iF$  is defined as the ratio of the number of particles inhaled to the number of particles which resuspend.

For our particular experimental sequence, the exposure period to resuspended particles is greater than the emission period. Thus, the total number of particles inhaled was determined by integrating the product of the occupant's breathing rate with the concentration of resuspended particles in their BZ (spatial average) during the 12.5 minute movement set (corrected for the clean set concentration) and the following decay period (the full 30 minutes for the first decay period and the first 30 minutes of the second decay period, for

consistency). The total number of particles released was determined by integrating the product of the resuspension rate, continuous dust loading, and seeded surface area. The size-resolved  $iF$  can be presented as:

$$iF_i(t) = \frac{\int_{t_0=0}^{t_{Set}=12.5 \text{ min}} Q_B(t) (C_{i,BZ}(t) - C_{i,BZ,Clean}(t)) dt + \int_{t_{Set}=12.5 \text{ min}}^{t_{Set}+t_{Decay}=42.5 \text{ min}} Q_B(t) C_{i,BZ}(t) dt}{A_M \int_{t_0=0}^{t_{Set}=12.5 \text{ min}} RR_i(t) L_i(t) dt} \quad (D.7)$$

where  $Q_B(t)$  is the occupant's volumetric breathing rate ( $\text{m}^3/\text{h}$ ). We assume a temporally constant breathing rate,  $Q_B$ , based on data provided by the U.S. EPA EFH (mean of  $0.299 \text{ m}^3/\text{h}$  for adults 21 to > 81 in age for sleep or nap activity, U.S. EPA 2009). We then apply a simplified expression to compute the average  $iF$  for the entire experimental sequence using time-averaged quantities. As defined below, this is the ratio of the average number of particles inhaled during both the movement set and decay periods to the average number of particles that resuspend during the movement set:

$$\overline{iF}_i = \frac{Q_B}{A_M} \frac{(\overline{C_{i,BZ,Set}} + \overline{C_{i,BZ,Decay}})}{\overline{RR}_i \times \overline{L}_i} \quad (D.8)$$

### Statistical Analysis

Statistical analysis was performed to evaluate the impact of various parameters (excluding the chamber ventilation rate due to a small sample size) on the resuspension rate and intake fraction using Wilcoxon non-parametric, two-related samples tests with statistical software (SPSS® version 20.0.0 2011,

IBM®).  $p$ -values equal to or below 0.05 were considered as statistically significant.

## RESULTS AND DISCUSSION

The following sections present the results from this investigation, including: particle number concentrations, impact of various parameters on the resuspension rate and intake fraction, a discussion of resuspension mechanisms, and an overview of exposure to resuspended particles.

### *Particle Number Concentration Profile*

Volunteer body movements on a seeded twin-size mattress were found to resuspend deposited mattress dust (ATD) particles in the range of 1 to 20  $\mu\text{m}$ . Significant elevations in airborne concentrations of resuspended particles were observed for each individual movement, M1-M5, as shown in a typical concentration profile in Figure D4 for volunteer RSV04. These short-term concentration peaks were approximately one to two orders of magnitude greater than the background concentration, for both movement sets and dust loads. The BZ and bulk air concentrations gradually increased over the entire 12.5 minute movement routine. At the cessation of the movement routine, particles concentrations were approximately an order of magnitude greater than background levels. The elevations in airborne particle concentrations follow a similar trend of what has been observed in full-scale walking resuspension studies by others (Karlsson 1999, Ferro et al. 2004a, Ferro et al. 2004b, Qian and Ferro 2008, Shaughnessy and Vu 2012).

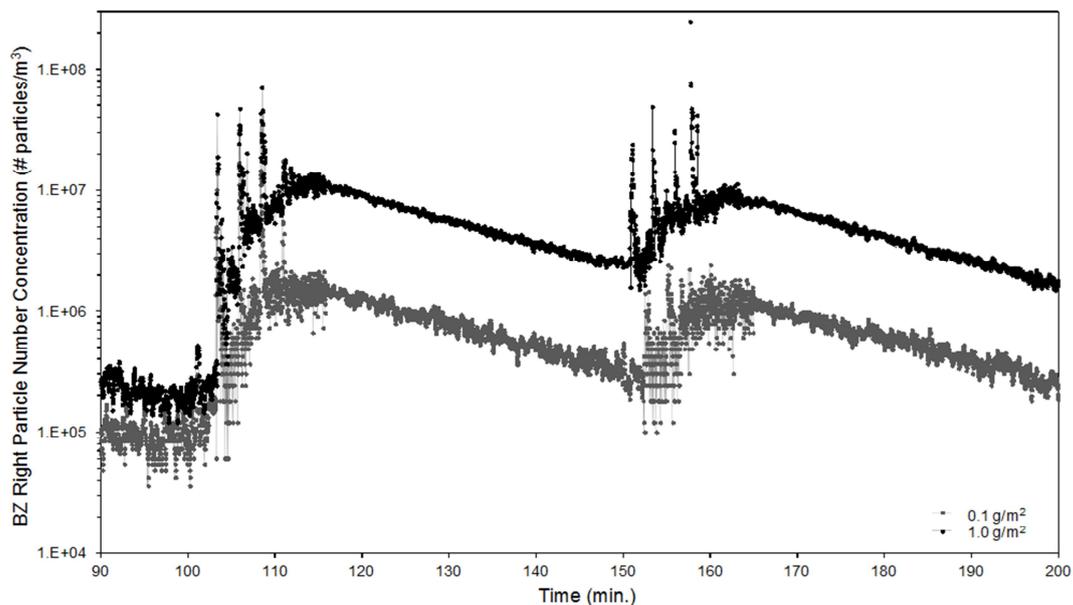


Figure D4: Example particle number concentration profile during a resuspension experiment: 1 to 2  $\mu\text{m}$ , both 0.1 and 1.0  $\text{g}/\text{m}^2$  dust loads, sampled at BZ right with APS for volunteer RSV04. Note: figure shows 1 sec. sampling average during sets 1 and 2, and 10 sec. sampling average during background and decay periods.

Dust loading had a significant impact on particle number concentration. Concentrations during the movement routine on the mattress seeded with 1  $\text{g}/\text{m}^2$  of ATD were typically an order of magnitude greater than those with 0.1  $\text{g}/\text{m}^2$ . In general, airborne concentrations reached approximately  $10^6$  particles/ $\text{m}^3$  for 0.1  $\text{g}/\text{m}^2$  and  $10^7$  particles/ $\text{m}^3$  for 1.0  $\text{g}/\text{m}^2$  at the cessation of a movement routine (Figure D4). In general, particle concentrations during the second movement set were only slightly lower than those during the first set.

Particle concentrations decayed linearly (on a logarithmic scale) during both decay stages due to removal via deposition and ventilation. The decay stages represent what an occupant might be exposed to after a resuspension event and during periods of inactivity while sleeping.

#### *Resuspension Rate: Particle Size, Movement Set, and Dust Load*

Figure D5 includes a summary of the average resuspension rates (time-averaged over 12.5 minute movement routine) among the 10 volunteers. The *RRs* were found to range over four orders of magnitude from  $10^{-3}$  to  $10^1$   $h^{-1}$ . *RR* increases with increasing particle size, by approximately 3 orders of magnitude from 1 to 2  $\mu m$  to 10 to 20  $\mu m$ . Differences in *RRs* between size fractions were found to be statistically significant (Table DS2). The size-dependence of *RR* is expected, given the basic mechanisms of particle detachment from surfaces and the findings of previous particle resuspension studies. For particles in the 1 to 20  $\mu m$  size range, resuspension tends to increase with increasing particle size as the ratio of removal forces to adhesion forces increases (Hinds 1999). The impact of particle size is particularly relevant to the sleep microenvironment, given that particle-phase mattress dust pollutants are associated with different size particles.

*RRs* tend to decrease from movement set 1 to set 2, and the differences were found to be statistically significant for most particle sizes and both dust loads (Table DS4). As discussed in Qian and Ferro (2008), the decay in *RR* may be due to a reduction in the number of particles available for resuspension

during periods of human activity, likely due to the distribution of adhesion forces for any given particle size. The slight reduction in  $RR$  may also be explained by the migration of particles deeper into the bed sheet fiber matrix due to pressure applied by the volunteer positioned above, as well as variability in movement technique and style between both movement sets for each volunteer. It is expected that prolonged periods of activity in bed may further reduce the  $RR$  throughout a sleep period.

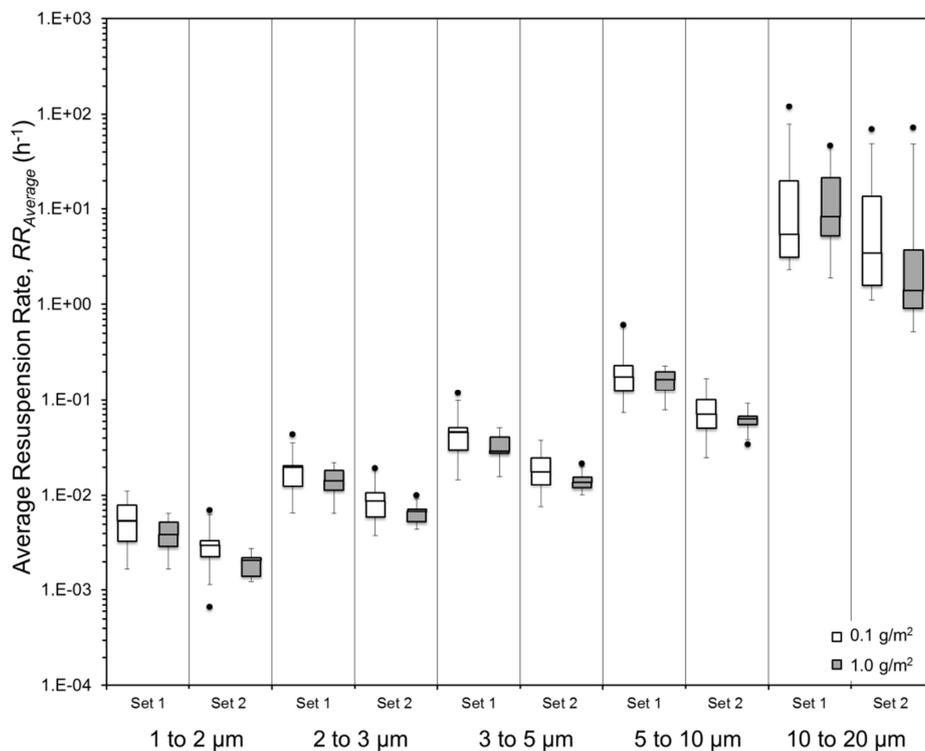


Figure D5: Average resuspension rates ( $RR$ ) among 10 volunteers over entire movement routine (M1-M5), for both  $0.1 \text{ g}/\text{m}^2$  and  $1.0 \text{ g}/\text{m}^2$  dust loads. Box plots represent interquartile range, whiskers represent the 5th and 95th percentiles, and dots represent outliers.

Dust loading appears to have a negligible impact on *RR*, despite the significant differences observed in particle concentrations. As shown in Table DS3, differences in *RR* between 0.1 and 1.0 g/m<sup>2</sup> were not found to be statistically significant for all particle sizes. Dust loading is an important parameter affecting resuspension, although the impact is much more pronounced when comparing monolayer and multilayer particle deposits (Boor et al. 2013a). For dust loads of 0.1 and 1.0 g/m<sup>2</sup>, particles are very likely deposited as a monolayer along the fiber, with minimal particle-to-particle contact. Thus, the impact of the type of particle deposit is much less pronounced, although resuspension can be influenced by seeding density (# of particles per deposition area) (Ibrahim et al. 2004). Mattress dust loads are typically in the 0.1 to 1.0 g/m<sup>2</sup> range, although lighter and heavier dust loads are likely, depending on how often bed sheets and the mattress are cleaned and the general cleanliness in a bedroom. The *RRs* reported in this investigation may not necessarily represent those for sparser or heavier deposits.

The size-resolved *RRs* estimated for human movement in bed are similar in magnitude to those reported by walking-induced resuspension studies by Qian and Ferro (2008) and Shaughnessy and Vu (2012). It is difficult to make direct comparisons in *RR* between studies due to differences in experimental methods, ventilation systems, particle sampling techniques, *RR* model assumptions, and movement frequency. It can be concluded, however, that the similar ranges in *RRs* suggest that particle resuspension associated with human body movements in bed is an important source of mattress dust particles, comparable to particles released from flooring via footfalls.

*Resuspension Rate: Volunteer Body Mass and BMI, Ventilation Rate, and Movement Type*

The impact of additional factors on the *RR* were also investigated (a detailed discussion can be found in the SI section, along with associated figures). Volunteer body mass and body mass index (BMI) had a minimal impact on the *RR*. Intuitively, resuspension would be expected to increase with body mass, however, it is possible that beyond a certain threshold weight, the removal forces induced by body movements in bed is much more dependent on movement intensity and technique, rather than their body mass alone. The chamber ventilation rate was also found to have a minimal impact on the *RR*. However, since the particle loss rate ( $a + k$ ) increases with the ventilation rate, improving ventilation may reduce the period over which an occupant is exposed to resuspended particles after cessation of body movements. Lastly, body movements of greater intensity (e.g. M3, 360° rotation of the torso) increased particle resuspension compared to less intense movements (e.g. M1, sitting on the mattress), as shown in Figure D6 for 3 to 5  $\mu\text{m}$  particles. A summary of movement-specific *RRs* can be found in the SI section.

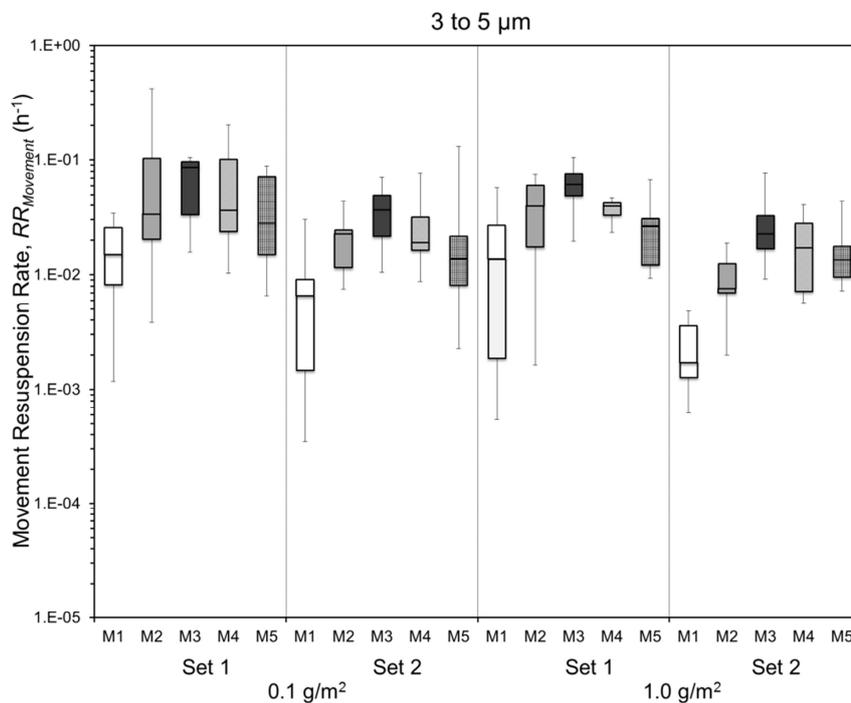


Figure D6: Average resuspension rates ( $RR$ ) among 10 volunteers for each individual movement (M1, M2, M3, M4, M5), 3 to 5  $\mu\text{m}$  (remaining size fractions can be found in Figure DS4). Box plots represent interquartile range, whiskers represent the 5th and 95th percentiles, and shading represents specific movements (M1-M5), as denoted on the x-axis.

### *Discussion on Resuspension Mechanisms: Mattress Surface Vibrations, Air Bursts, and Other Factors*

The impact each of the five movements, M1-M5, was associated with a peak in mattress surface vibration, as shown in Figure DS5 (a.) and (b.). Peak surface vibrations were generally between 0.1 and 1  $g$  ( $9.81 \text{ m/s}^2$ ) in magnitude. To put these values in perspective, Gomes et al. (2007) reported walking-induced peak floor vibrations of generally  $\leq 0.1 \text{ g}$ .

The *RR* for a specific movement was found to increase with increasing magnitude of surface vibrations. As previously discussed, *RR* was found to be the greatest for M3: full 360° rotation (Figure D6). The surface vibrations induced by M3 were also the largest of the five movements. Movements of greater intensity, such as a full body rotation, can generate greater surface vibrations, and in turn, increase the magnitude of the associated removal forces, such as the wall vibration force (Theerachaisupakij et al. 2002) and lift-off drag force (Gomes et al. 2007), thereby enhancing particle resuspension. Additionally, M1 was generally associated with the lowest surface vibration magnitudes, and in turn, the lowest *RRs*.

In addition to surface vibrations, each of the five movements, M1-M5, was associated with a peak in air velocity ~ 1 cm above the mattress surface as measured in a trial set of experiments (Figure DS5 (c.)). The airflow regime induced by human movements in bed is complex. During movements, pockets of air beneath the loosely bound bed sheet (visible ripples) were observed. Thus, there is likely a combination of airflow through the porous bedding fabric matrix (“air burst”), as well across the bedding surface. This complex airflow pattern may aid in increasing the aerodynamic removal forces acting on the deposited particles.

The air velocities increased suddenly, suggesting impulsive and highly accelerated airflow, similar to airflows generated by footfalls or descending objects (e.g. Khalifa and Elhadidi 2007, Kubota et al. 2009, Choi et al. 2012). The impulsive nature of the flow may be considerably more important in inducing resuspension from the bed sheets than the maximum velocities achieved. Ibrahim et al. (2003) found *RRs* to be two orders of magnitude greater during

periods of acceleration compared to steady-state velocity conditions in their wind tunnel study. This is in part due to the additional aerodynamic removal force, known as the Basset force, which can arise as the flow is accelerated (Tadmor and Zur 1981). The turbulent airflow may also induce vibrations of the bedding fibers. Additionally, Fletcher et al. (2008) found turbulent and impulsive pulsed air jets to resuspend significant fractions of 1 to 45  $\mu\text{m}$  particles from muslin cloth. Impulsive “air bursts” may be an important mechanism in detaching settled mattress dust particles.

Mechanical abrasion caused by direct contact between the volunteer and the deposited particles, as well as sections of the bed sheets rubbing against each other, may generate additional removal forces. During the movement routine, it is likely that some fraction of the deposited ATD particles were transferred to the volunteer’s clean suit outfit. This contact transfer could have dual effect of reducing the number of particles available for resuspension from the bed sheet ( $L_{i,0}$ ) and causing secondary resuspension of the transferred particles from the Tyvek clean suit. In regard to the latter, studies have demonstrated that particles can be efficiently resuspended from clothing worn by occupants (Bloor and Dinsdale 1962, Bohne and Cohen 1985, Cohen and Positano 1986, You et al. 2013, McDonagh et al. 2014). Resuspension from the clean suit likely occurred to some extent during the experiments, although it is not considered in the  $RR$  and  $iF$  analysis.

### *Intake Fraction*

Time-averaged intake fractions ( $iF$ ) were generally in the range of  $10^2$  to  $10^4$  inhaled particles per million resuspended, as shown in Figure D7 (a.).

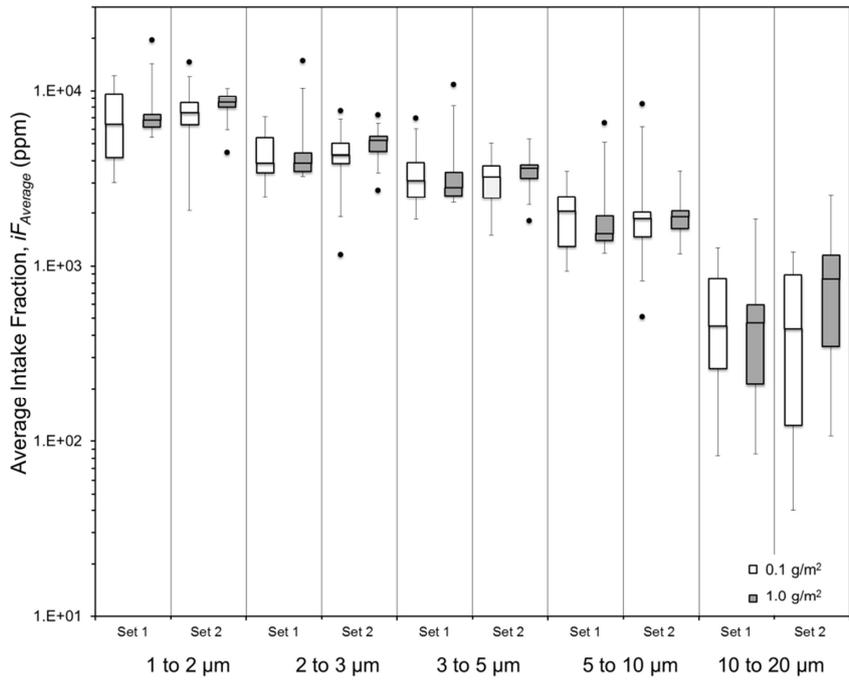


Figure D7 (a.)

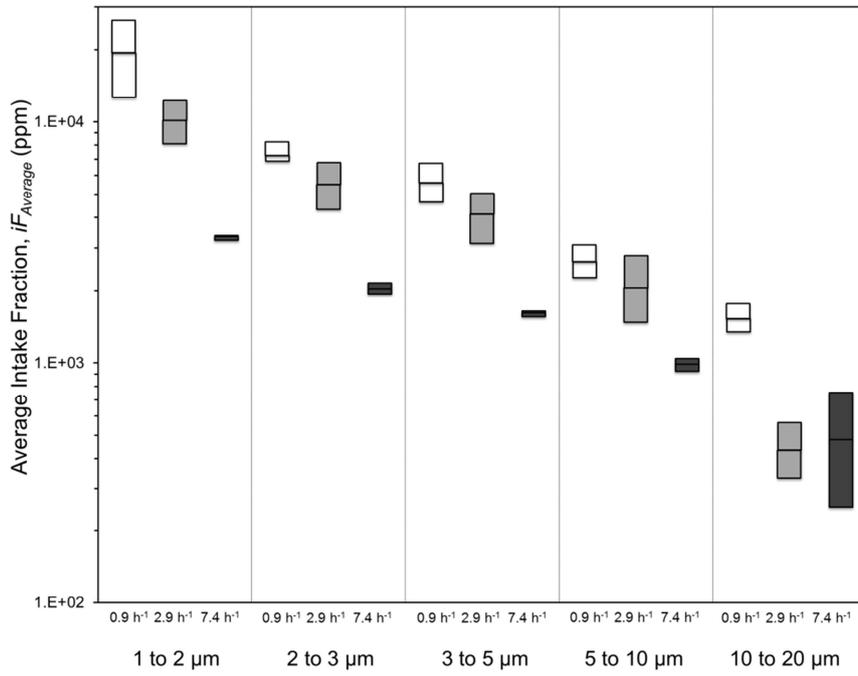


Figure D7 (b.)

Figure D7: (a.) average intake fraction ( $iF$ ) among 10 volunteers over entire movement sequence (M1-M5 and decay), for both 0.1 g/m<sup>2</sup> and 1.0 g/m<sup>2</sup> dust loads. Box plots represent interquartile range, whiskers represent the 5th and 95th percentiles, and dots represent outliers, (b.) impact of chamber ventilation rate on average intake fraction ( $iF$ ) over entire movement sequence (M1-M5 and decay) for two volunteers (RSV01 & RSV02). Box plots represent interquartile range and shading represents ventilation rate, as denoted on the x-axis.

For particles in the range of 1 to 10  $\mu\text{m}$ ,  $iF$ s are generally on the order of  $10^3$  to  $10^4$  ppm, and for particles in the range of 10 to 20  $\mu\text{m}$ ,  $iF$ s are generally on the order of  $10^2$  ppm. Although the  $RR$  increases with increasing particle size, the opposite is observed for  $iF$ . Differences in  $iF$  among the different size fractions were found to be statistically significant (Table DS2). Thus, even though larger particles more easily resuspend, a greater fraction of smaller particles that resuspend will be inhaled. This is due to a combination of lower deposition rates for smaller particles and greater breathing zone concentrations for smaller particles, the latter of which is due in part to the size distribution of the ATD used to generate the artificial mattress dust deposits (mass median diameter of 4.5  $\mu\text{m}$ ). The smaller particles have a greater likelihood of being inhaled, rather than be removed via gravitational settling to the mattress surface, compared to the larger particles (Marshall and Nazaroff 2007, Nazaroff 2008).

$iF$  was found to be slightly greater for a dust load of 1.0 g/m<sup>2</sup> compared to 0.1 g/m<sup>2</sup>, although a statistically significant difference was not observed.  $iF$  remained nearly the same from movement set 1 to set 2.  $RR$  was not strongly influenced by the chamber ventilation rate, however,  $iF$  increased with decreasing ventilation rate from 7.4 to 0.9 h<sup>-1</sup> for particles in the 1 to 10  $\mu\text{m}$  range, as shown in Figure D7 (b.). Increasing the ventilation rate increases the fraction

of particles that are removed from the chamber air rather than inhaled, thus decreasing  $iF$ . Low ventilation rates have been reported for naturally ventilated children's bedrooms by Bekö et al. (2010), with a mean ventilation rate of  $0.46 \text{ h}^{-1}$ , and rates as low as  $0.1 \text{ h}^{-1}$ . Lower ventilation rates may lead to elevated exposures and increase the  $iF$  beyond values reported here, while proper ventilation strategies and effective use of in-duct and portable particle filtration technologies may reduce exposure and  $iF$  below values reported here.

$iFs$  on the order of  $10^2$  and  $10^4$  ppm are similar in magnitude to those reported for indoor particle sources (e.g. Lai et al. 2000, Marshall and Nazaroff 2007). Lai et al. (2000) reported  $iFs$  in the range of  $10^3$  to  $10^5$  ppm for nonreactive pollutants in a single zone residence with 1-5 occupants based on a modeling analysis. Size-resolved  $iFs$  for a single, well-mixed building were found to be approximately  $7.5 \times 10^3$  for  $1 \mu\text{m}$  particles at a ventilation rate of  $0.2 \text{ h}^{-1}$ ,  $10^3$  for  $1 \mu\text{m}$  particles at a ventilation rate of  $2.0 \text{ h}^{-1}$ , and  $10^2$  for  $10 \mu\text{m}$  particles at both ventilation rates. The impact of particle size and ventilation rate is similar to the trend observed in this investigation for resuspended mattress dust particles.

#### *Implications for Inhalation Exposure*

The non-negligible fractions of resuspended particles that are inhaled are in part due to the important role of the source-proximity effect on exposure to pollutants originating in the sleep microenvironment. The ratios of the spatial BZ average concentrations to the bulk air concentrations were (mean $\pm$ SD among all tests): 1-2  $\mu\text{m}$ :  $1.94\pm 0.52$ , 2-3  $\mu\text{m}$ :  $1.26\pm 0.30$ , 3-5  $\mu\text{m}$ :  $1.07\pm 0.23$ , 5-10  $\mu\text{m}$ :  $1.63\pm 0.38$ , and 10-20  $\mu\text{m}$ :  $1.10\pm 0.34$ . Similar findings have been reported for the distribution of  $\text{SF}_6$  tracer gas released from a test mattress by Laverge et al. (2013)

and for VOCs emitted from a crib mattress by Boor et al. (2014). Additionally, for a person to be exposed to particles  $\geq 10 \mu\text{m}$ , which have high settling velocities (on the order of  $\sim 10^{-3} \text{ m/s}$ ), close proximity of the BZ to the particle deposit is critical, as is the case in the sleep microenvironment.

### *Study Limitations*

The *RR* and *iF* values reported in this investigation should be viewed as best estimates that are specific to the experimental setup and the set of prescribed movements that the volunteers undertook. There is uncertainty in extrapolating these results to actual bedroom environments, where there are numerous complexities that cannot be captured in a laboratory setting. There is also wide variability in the type of ventilation systems installed in a residence, mixing conditions in a room, characteristics of actual mattress dust particles on an occupant's bed (surface features, size distribution, composition, and shape, etc.), moisture levels near bedding surfaces, bedding arrangements and materials, and occupant movement patterns in bed, all of which are parameters that may impact resuspension and exposure.

The resuspension rate analysis is based upon a two-compartment model (bulk chamber air and surface loading). A two-compartment model was selected over a three-compartment model (BZ, bulk chamber air, and surface loading) due to the complexity of defining the inner-zonal air exchange between the BZ and bulk chamber air compartments and lack of experimental data on the inner-zonal air exchange rate (see Earnest and Corsi 2013). A three-compartment model would improve the accuracy of the *RR* estimate, however, the two-compartment model provides a reasonable estimate of *RR*. Additionally, the

particle concentration in the bulk chamber air was assumed to be uniform and well represented by the OPC positioned at the fixed bulk air monitoring location. Resuspension mixing tests were completed with five OPCs positioned throughout the bulk chamber air to evaluate this assumption. The percent difference in resuspended particle concentrations measured at each location with the reference location, bulk air, was found to be below 30% for all size fractions, and below 20% for the total particle concentration, sum of 1 to 20  $\mu\text{m}$ , similar to the mixing conditions reported in Qian and Ferro (2008).

#### CONCLUSIONS

Full-scale chamber experiments were conducted to investigate human-induced particle resuspension from mattresses. Significant elevations in airborne particle concentrations were observed as human volunteers performed prescribed routines of five movements on an artificially seeded mattress wrapped in two layers of bed sheets. Resuspension rates were estimated for ATD particles in the 1 to 20  $\mu\text{m}$  size range. Resuspension rates increased with increasing particle size, decreased with repeated movement sets, and were in the range of  $10^{-3}$  to  $10^1 \text{ h}^{-1}$ . Resuspension generally did not show a strong association with (for the ranges examined) dust loading, volunteer body mass and body mass index, and chamber ventilation rate. Movements of greater intensity, such as a full rotation of the body, were found to be associated with higher resuspension rates than lower intensity movements, such as sitting on the mattress. The possible mechanisms of resuspension likely include a combination of surface vibrations of the bedding fibers, aerodynamic removal forces associated with impulsive "air bursts" through the porous bedding, mechanical abrasion, and contact transfer.

Intake fractions, as estimated from breathing zone particle number concentrations, were in the range of  $10^2$  to  $10^4$  inhaled particles per million resuspended, demonstrating that a significant fraction of resuspended particles may be inhaled by a sleeping occupant. Additionally, elevated concentrations after the completion of a movement routine suggests that people may be exposed to resuspended particles as they lay still in bed following a resuspension event. The results suggest that resuspension is an important source mechanism for particle-phase pollutants originating in mattress dust.

#### AUTHOR CONTRIBUTIONS

B.E.B., M.P.S., R.L.C., and A.N. defined the overall scope of the study and developed the experimental methods. B.E.B. and M.P.S. conducted the experiments and analyzed the data. B.E.B. wrote the paper. M.P.S. provided detailed comments on draft manuscripts. R.L.C. and A.N. provided guidance in the execution of the experiments, advised in the interpretation of the results, and provided detailed comments on draft manuscripts.

#### SUPPORTING INFORMATION

*Detailed Discussion on the Impact of Volunteer Body Mass, Ventilation Rate, and Movement Type*

##### *Volunteer Body Mass*

Body mass appears to have a minimal impact on *RR*, as shown in Figure DS2 (a.) and (b.) (impact of body mass index (BMI) shown in Figure DS3). For a dust load of  $0.1 \text{ g/m}^2$  and all particle sizes, differences in *RR* between a group of

the lightest five volunteers and a group of the heaviest five volunteers was not found to be statistically significant (Tables DS5 and DS6). However, for several size bins at 1.0 g/m<sup>2</sup>, statistically significant differences were determined between the two body mass groups, with a slight increase in *RR* for the lighter group.

Intuitively, resuspension would be expected to increase with body mass (or BMI), however, it is possible that beyond a certain threshold weight, the removal forces induced by body movements in bed is much more dependent on the intensity of the volunteer's movements, the surface vibrations they induce, and their particular movement technique, rather than their body mass alone. For example, a person with a body mass of 55 kg moving in a very abrupt, intense manner may have a similar, or even more pronounced, impact on particle resuspension as a 120 kg individual who moves very gently and cautiously in bed. Qian and Ferro (2008) reported a similar finding, in that resuspension tended to be greater for faster and more intense walking styles compared to less active styles, regardless of the volunteer's body mass.

#### *Chamber Ventilation Rate*

The impact of the chamber ventilation rate (0.9, 2.9, and 7.4 h<sup>-1</sup>) on airborne particle concentrations is shown in Figure DS6 (a). Similar concentration profiles were observed during the movement routine among the three ventilation rates, with the highest concentrations observed for the lowest ventilation rate of 0.9 h<sup>-1</sup>. As expected, ventilation rate appears to have the most significant impact on particle concentrations during the two decay periods. The particle loss rate ( $a + k$ ) increases with the ventilation rate, thus, improving

ventilation may reduce the period over which an occupant is exposed to resuspended particles after cessation of body movements. This is especially important for accumulation-mode particles that have low deposition rates.

*RR* was not strongly influenced by the chamber ventilation rate (Figure DS6 (b.)), with *RR*s at 0.9 h<sup>-1</sup> similar to those at 2.9 and 7.4 h<sup>-1</sup>. It is likely that ventilation rates across this range have minimal impact on affecting the mechanisms of resuspension at the surface of the bedding fibers. The ventilation rate, however, may impact mixing conditions, airflow distribution within the chamber, the volunteer's thermal plume, and the spatial uniformity of the particle concentrations in the bulk chamber air (e.g. Rim and Novoselac 2009). The simplified two-compartment *RR* model cannot capture these complex effects. Thus, caution is advised in making conclusions about the impact of ventilation rate on resuspension.

#### *Movement-Specific Resuspension Rates*

Figure DS4 (a.-e.) show the *RR*s time-averaged over each 2.5 min. movement, M1, M2, M3, M4, and M5. Table DS7 lists the *P*-values associated with comparisons between different movements. Movement-specific *RR*s were generally found to be statistically significantly different when comparing M3, 360° rotation of the torso, with M1 (M3 > M1), M2 (M3 > M2), M4 (M3 > M4), and M5 (M3 > M5) (some cases, depending on particle size, movement set, or dust load, were not statistically significant). Similarly, *RR*s for M1, sitting on the mattress, were found to be statistically significantly lower from the other movements for numerous cases. M2, laying in supine position, and M5, 180° rotation of torso from prone to supine position, were not found to be statistically

significantly different, whereas differences between M4 with M2 ( $M4 > M2$ ) and M5 ( $M4 > M5$ ) were found to be statistically significant for numerous cases. These findings make sense, given that M3 represents the greatest degree of rotation of the torso, M1 is the least intense movement, and M2 and M5 represent similar intensities.

Figure DS4 (a.-e.) can be viewed as a time-series of *RR*, from M1 in movement set 1 to M5 in movement set 2. It can be observed that *RR* increases from M1 to M3 in movement set 1, with a slight decrease from M3 to M4, and an additional decrease to M5. Movement set 2 follows a nearly identical trend. The movement routines examined in this investigation were not intended to represent actual sequences of movements sleeping occupants may perform. Furthermore, the order of the specific movements may be a factor. If the 360° rotation of the torso was performed first, rather than third (M3) in the sequence, *RRs* associated with M3 may be greater, given the general decay in resuspension with time, as previously discussed. This decay may partially explain why  $M4 > M5$ , even though both movements were very similar.



Figures

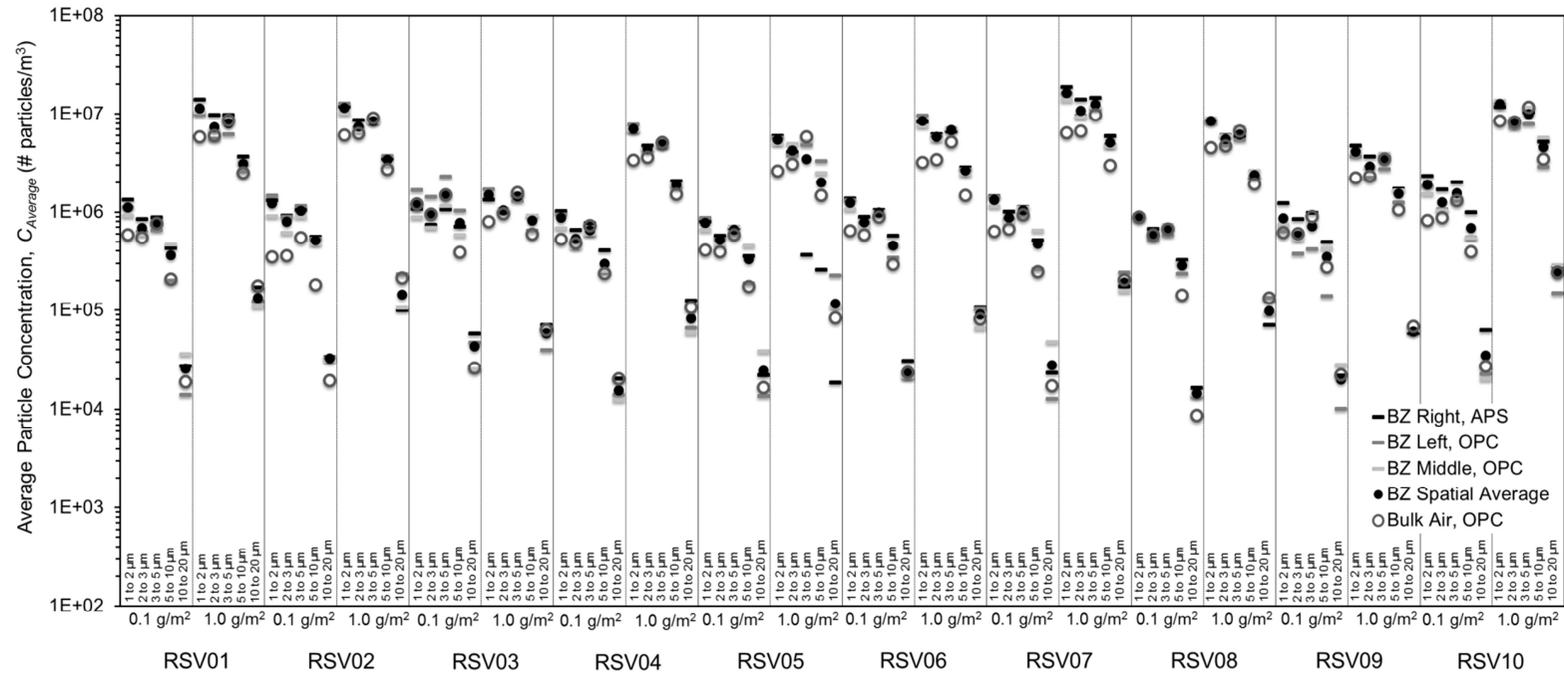


Figure DS1: Spatial distribution of average particle concentration over entire movement routine (M1-M5, Set 1) among all sampling locations: Bulk Air, BZ Right, BZ Left, BZ Middle, and spatial BZ average for each experiment, highlighting the source-proximity effect.

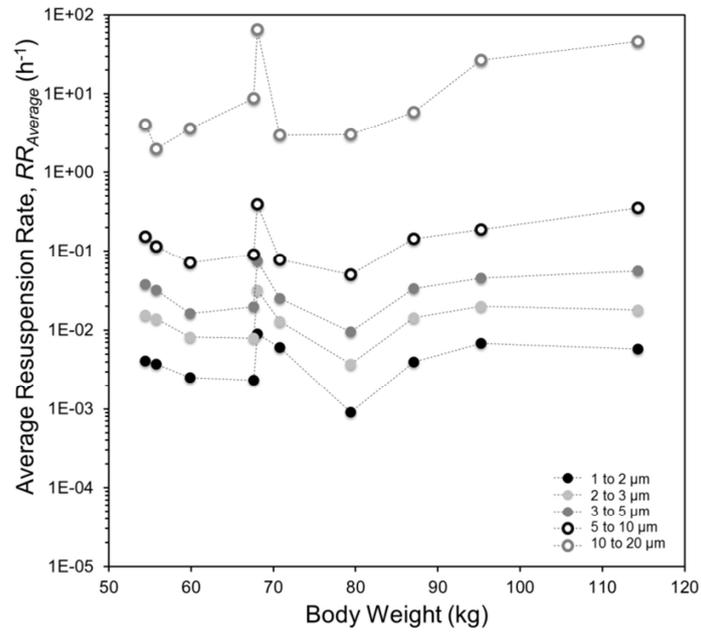


Figure DS2 (a.) 0.1 g/m<sup>2</sup>

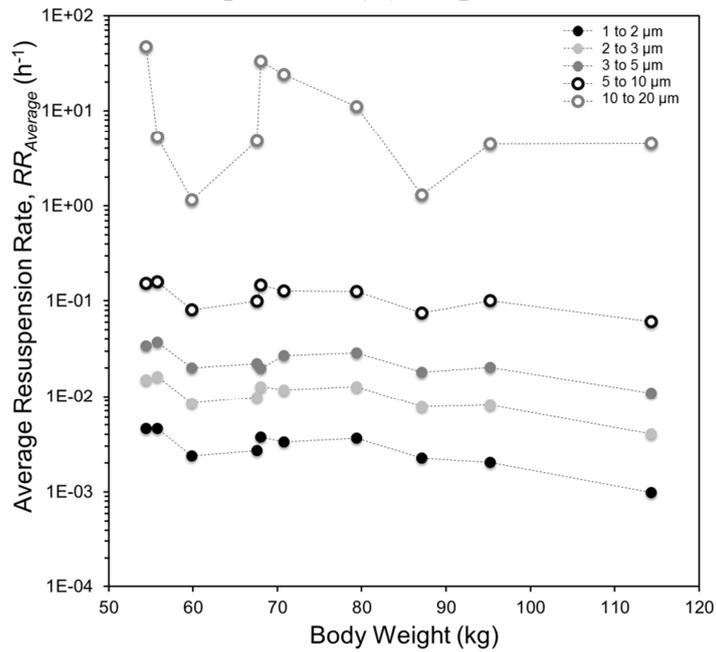


Figure DS2 (b.) 1.0 g/m<sup>2</sup>

Figure DS2: Average resuspension rates ( $RR$ ) over entire movement routine (M1-M5) as a function of volunteer body weight, (a.) 0.1 g/m<sup>2</sup> and (b.) 1.0 g/m<sup>2</sup> dust loads.

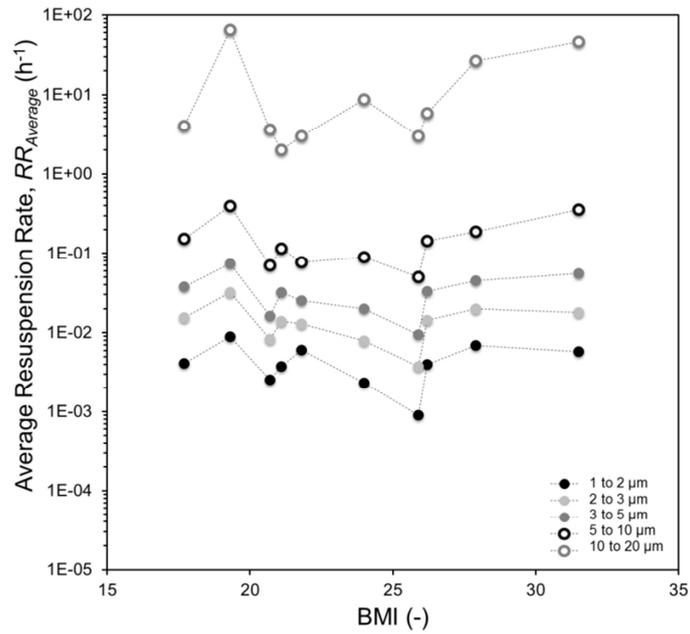


Figure DS3 (a.) 0.1 g/m<sup>2</sup>

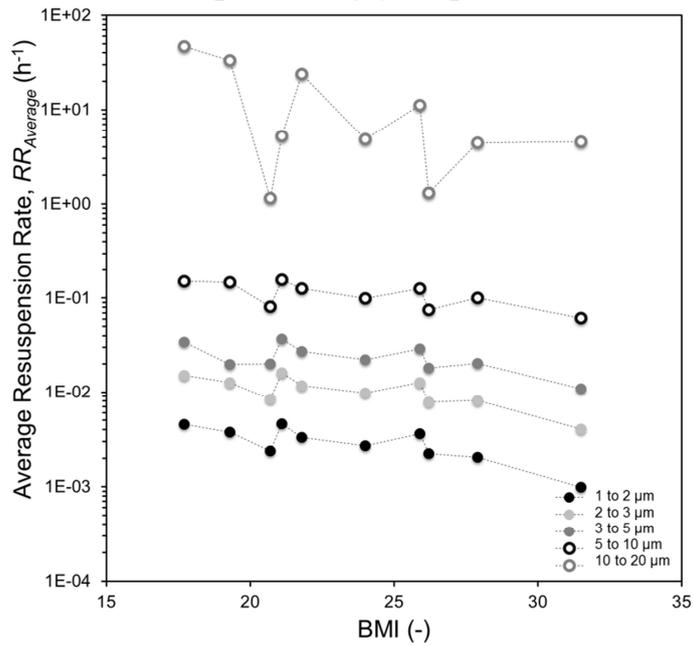


Figure DS3 (b.) 1.0 g/m<sup>2</sup>

Figure DS3: Average resuspension rates ( $RR$ ) over entire movement routine (M1-M5) as a function of volunteer body mass index (BMI), (a.) 0.1 g/m<sup>2</sup> and (b.) 1.0 g/m<sup>2</sup> dust loads.

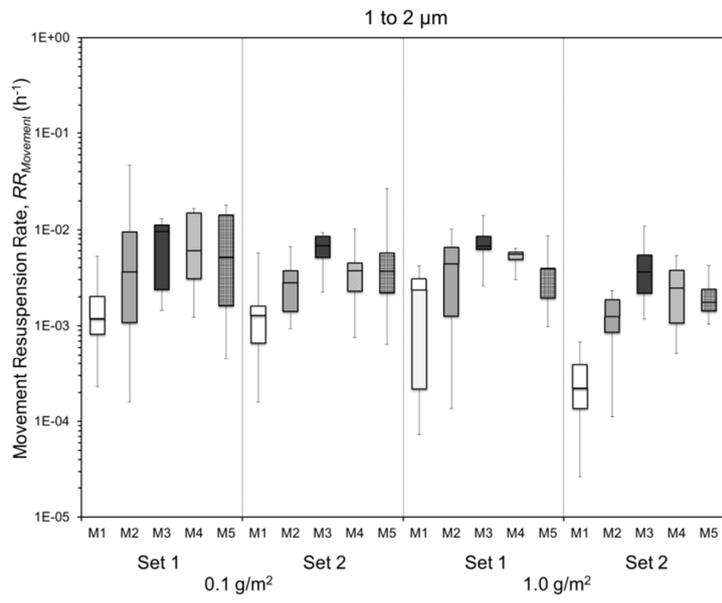


Figure DS4 (a.)

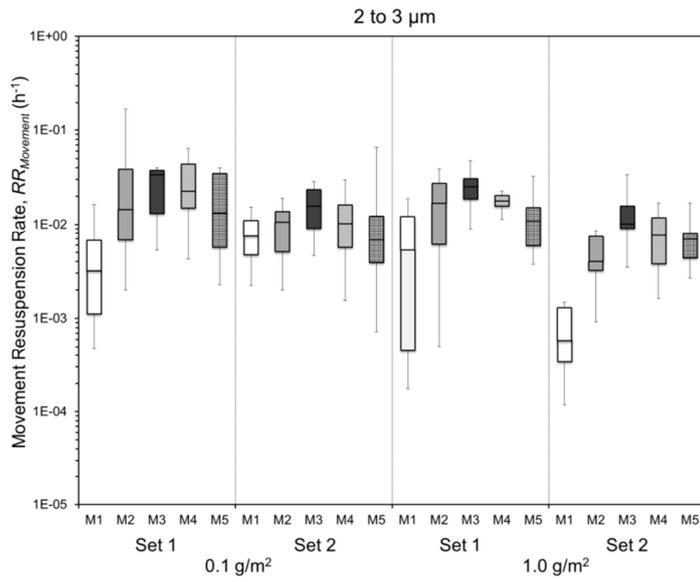


Figure DS4 (b.)

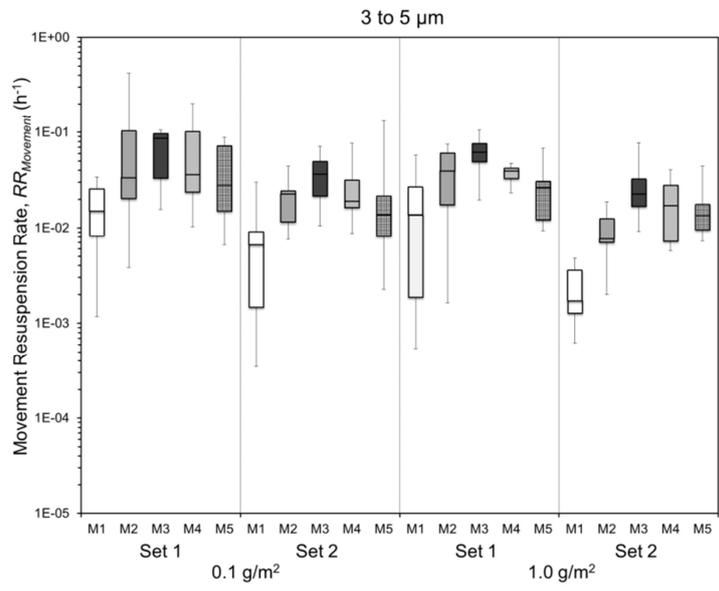


Figure DS4 (c.)

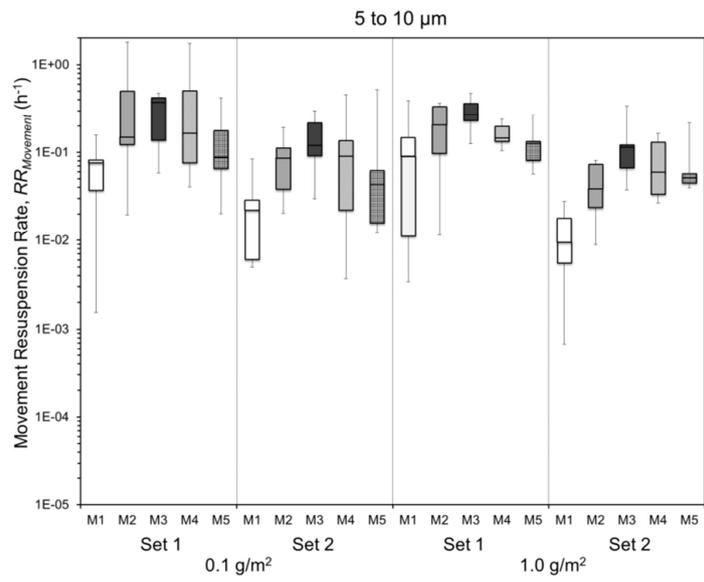


Figure DS4 (d.)

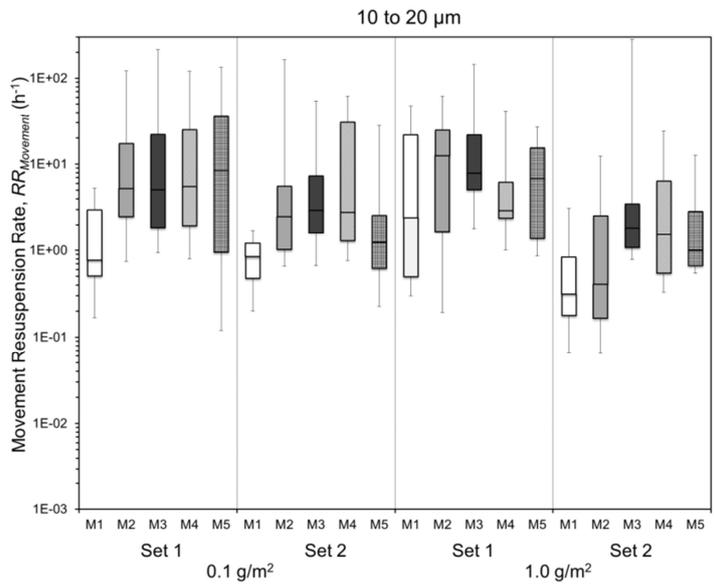


Figure DS4 (e.)

Figure DS4: Average resuspension rates ( $RR$ ) among 10 volunteers for each individual movement (M1, M2, M3, M4, M5), (a.): 1 to 2  $\mu\text{m}$ , (b.) 2 to 3  $\mu\text{m}$ , (c.) 3 to 5  $\mu\text{m}$ , (d.) 5 to 10  $\mu\text{m}$ , and (e.) 10 to 20  $\mu\text{m}$ . Box plots represent interquartile range, whiskers represent the 5th and 95th percentiles, and shading represents specific movements (M1-M5), as denoted on the x-axis.

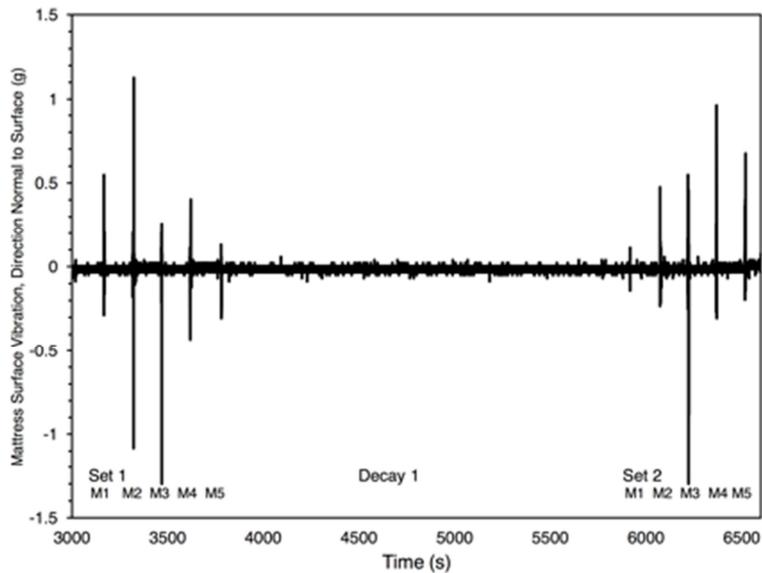


Figure DS5 (a.)

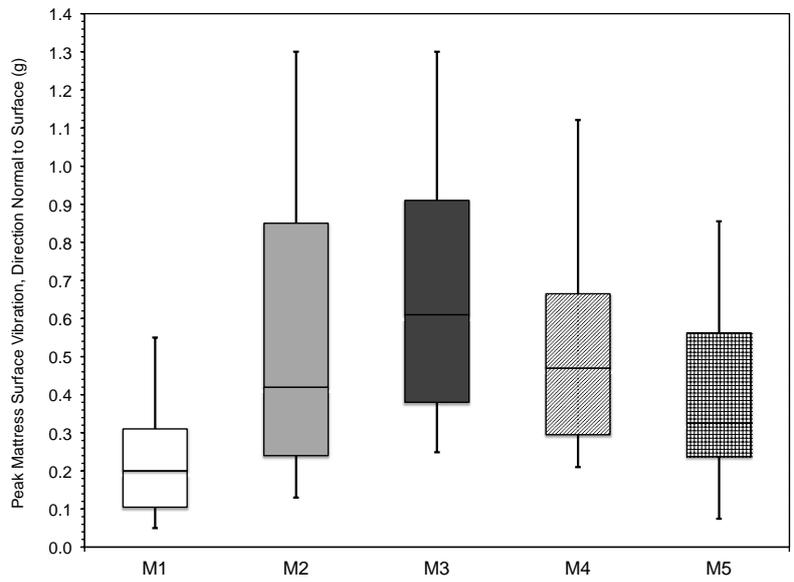


Figure DS5 (b.)

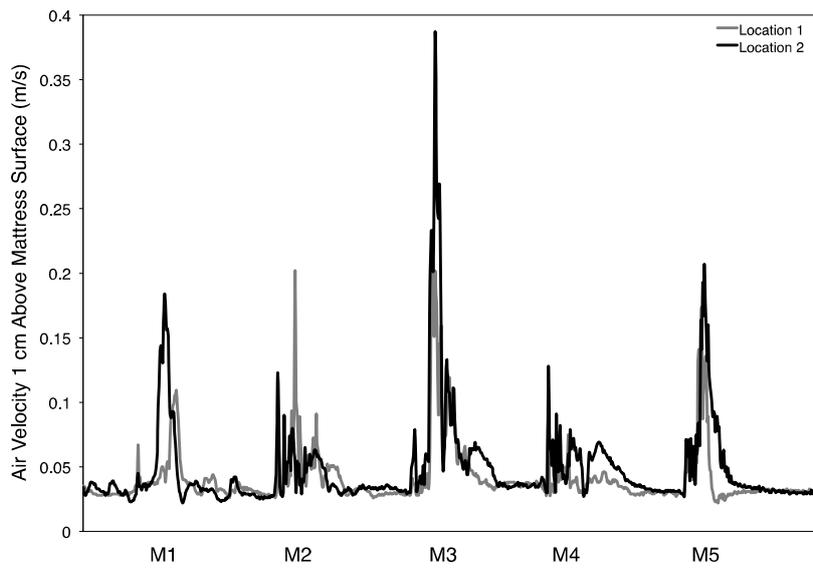


Figure DS5 (c.)

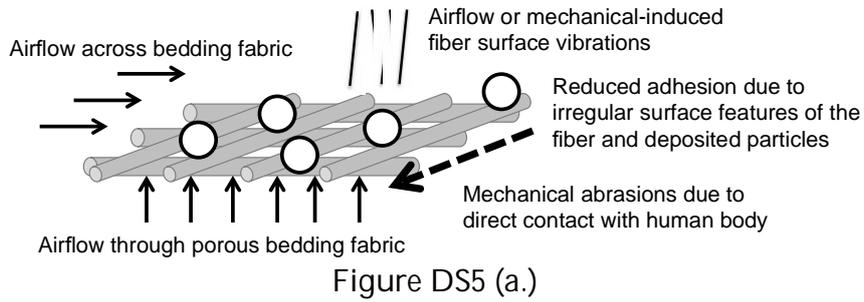


Figure DS5: (a.) Example profile of mattress surface vibrations during a resuspension experiment, (b.) the distribution of peak mattress surface vibrations (direction normal to surface, magnitude) across all resuspension experiments for each movement, (c.) example of air velocity 2.5 cm above mattress surface, and (d.) possible mechanisms that may be responsible for particle resuspension from the mattress and bedding fabric surfaces. Box plots represent interquartile range and whiskers represent the 5th and 95th percentiles.

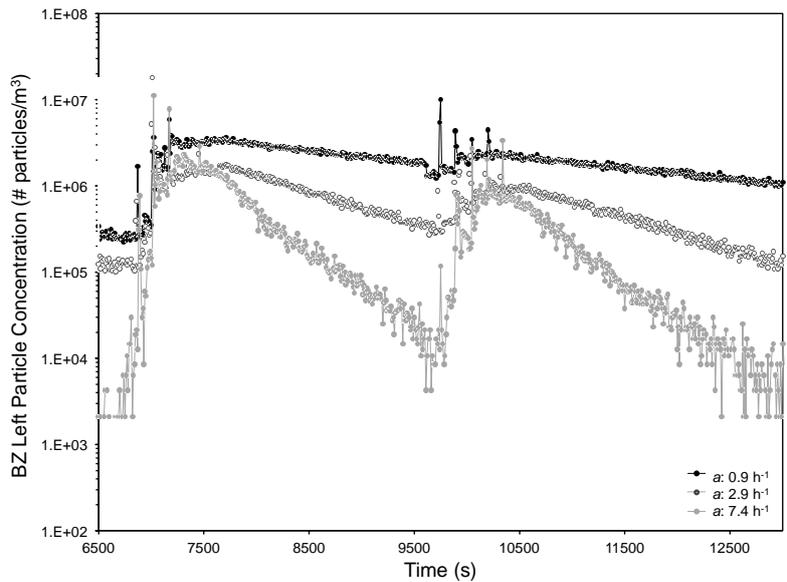


Figure DS6 (a.)

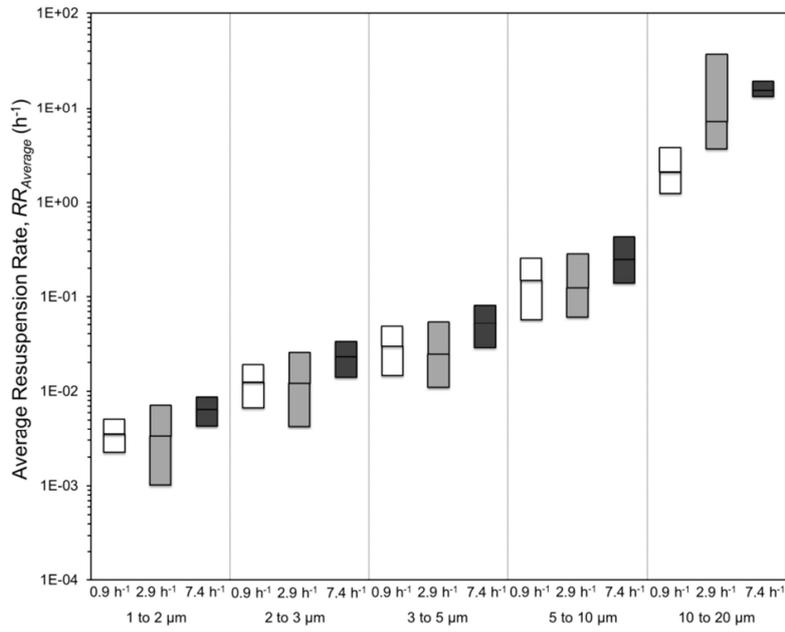


Figure DS6 (b.)

Figure DS6: (a.) impact of ventilation rate on particle concentration during movement routine and decay periods, (b.) impact of ventilation rate on average resuspension rate ( $RR$ ) over entire movement routine (M1-M5) for two volunteers (RSV01 & RSV02). Box plots represent interquartile range and shading represents ventilation rate, as denoted on the x-axis.

Tables

Table DS1: Experimental matrix.

Volunteer ID	Weight (kg)	Height (cm)	BMI <sup>1</sup>	Dust Load, $L_0$ (g/m <sup>2</sup> )	
				Nominal <sup>2</sup>	Actual <sup>3</sup>
RSV01	68	188	19.2	0.1 ( $a = 0.9 \text{ h}^{-1}$ )	0.14±0.06
				0.1 ( $a = 2.9 \text{ h}^{-1}$ )	0.08±0.03
				0.1 ( $a = 7.4 \text{ h}^{-1}$ )	0.06±0.02
				0.5	0.56±0.17
				1	1.12±0.17
RSV02	79	175	25.8	0.1 ( $a = 0.9 \text{ h}^{-1}$ )	0.14±0.06
				0.1 ( $a = 2.9 \text{ h}^{-1}$ )	0.20±0.09
				0.1 ( $a = 7.4 \text{ h}^{-1}$ )	0.09±0.05
				0.5	0.58±0.13
				1	1.12±0.22
RSV03	114	191	31.2	0.1	0.10±0.08
				1	0.98±0.17
RSV04	68	168	24.1	0.1	0.12±0.06
				1	0.85±0.10
RSV05	95	185	27.8	0.1	0.10±0.04
				1	0.90±0.09
RSV06	60	170	20.8	0.1	0.10±0.04
				1	0.91±0.05
RSV07	56	163	21.1	0.1	0.10±0.05
				1	1.19±0.18
RSV08	71	180	21.9	0.1	0.11±0.04
				1	1.07±0.12
RSV09	87	182	26.3	0.1	0.08±0.04
				1	1.01±0.14
RSV10	54	175	17.6	0.1	0.11±0.04
				1	1.26±0.17

<sup>1</sup>: Calculated with The National Heart, Lung, and Blood Institute's (NHLBI) Body Mass Index (BMI) calculator.

<sup>2</sup>: All experiments were performed at a chamber ventilation rate,  $a$ , of  $2.9 \text{ h}^{-1}$ , unless otherwise specified.

<sup>3</sup>: Mean of the nine microscope slides  $\pm$  standard deviation (SD)

Table DS2: Impact of particle size on average resuspension rate (*RR*) and intake fraction (*iF*), *P*-values from Wilcoxon non-parametric, two-related samples tests<sup>1</sup>.

		Movement Set 1				Movement Set 2			
		1 to 2 $\mu\text{m} < 2$ to 3 $\mu\text{m}$	2 to 3 $\mu\text{m} < 3$ to 5 $\mu\text{m}$	3 to 5 $\mu\text{m} < 5$ to 10 $\mu\text{m}$	5 to 10 $\mu\text{m} <$ 10 to 20 $\mu\text{m}$	1 to 2 $\mu\text{m} < 2$ to 3 $\mu\text{m}$	2 to 3 $\mu\text{m} < 3$ to 5 $\mu\text{m}$	3 to 5 $\mu\text{m} < 5$ to 10 $\mu\text{m}$	5 to 10 $\mu\text{m} <$ 10 to 20 $\mu\text{m}$
Dust Load: 0.1 g/m <sup>2</sup>	<i>RR</i>	0.013	0.007	0.005	0.005	0.005	0.005	0.005	0.005
	<i>iF</i>	0.005*	0.005*	0.005*	0.005*	0.005*	0.013*	0.005*	0.005*
Dust Load: 1.0 g/m <sup>2</sup>	<i>RR</i>	0.005	0.059	0.005	0.005	0.005	0.005	0.005	0.005
	<i>iF</i>	0.005*	0.005*	0.074*	0.005*	0.005*	0.009*	0.005*	0.005*

<sup>1</sup>: The non-parametric differences were considered significant for  $P \leq 0.05$  (bold).

\*: *P*-values represent negative trend between variables (e.g. *RR* for smaller size fraction is greater than that for larger size fraction).

Table DS3: Impact of dust load on average resuspension rate (*RR*) and intake fraction (*iF*), *P*-values from Wilcoxon non-parametric, two-related samples tests<sup>1</sup>.

		Movement Set 1					Movement Set 2				
		1 to 2 $\mu\text{m}$	2 to 3 $\mu\text{m}$	3 to 5 $\mu\text{m}$	5 to 10 $\mu\text{m}$	10 to 20 $\mu\text{m}$	1 to 2 $\mu\text{m}$	2 to 3 $\mu\text{m}$	3 to 5 $\mu\text{m}$	5 to 10 $\mu\text{m}$	10 to 20 $\mu\text{m}$
Dust Load: 0.1 g/m <sup>2</sup> < 1.0 g/m <sup>2</sup>	<i>RR</i>	0.241	0.445	0.333*	0.646	0.959	0.059	0.169	0.203	0.285	0.285
	<i>iF</i>	0.959	0.333*	0.646*	0.575*	0.575*	0- 333	0.646	0.508	0.959	0.169

<sup>1</sup>: The non-parametric differences were considered significant for  $P \leq 0.05$  (bold).

\*: *P*-values represent negative trend between variables.

Table DS4: Impact of movement set on average resuspension rate (*RR*) and intake fraction (*iF*), *P*-values from Wilcoxon non-parametric, two-related samples tests<sup>1</sup>.

	Dust Load: 0.1 g/m <sup>2</sup>					Dust Load: 1.0 g/m <sup>2</sup>					
		1 to 2 µm	2 to 3 µm	3 to 5 µm	5 to 10 µm	10 to 20 µm	1 to 2 µm	2 to 3 µm	3 to 5 µm	5 to 10 µm	10 to 20 µm
Movement Set 1	<i>RR</i>	0.013*	0.005*	0.005*	0.005*	0.386*	0.007*	0.007*	0.007*	0.007*	0.074*
Movement Set 2	<i>iF</i>	0.646	0.959	0.799	0.959	0.878*	0.139	0.114	0.169	0.333	0.169

<sup>1</sup>: The non-parametric differences were considered significant for  $P \leq 0.05$  (bold).

\*: *P*-values represent negative trend between variables.

Table DS5: Impact of volunteer body mass on average resuspension rate (*RR*), *P*-values from Wilcoxon non-parametric, two-related samples tests<sup>1</sup>.

	Dust Load: 0.1 g/m <sup>2</sup>					Dust Load: 1.0 g/m <sup>2</sup>					
		1 to 2 µm	2 to 3 µm	3 to 5 µm	5 to 10 µm	10 to 20 µm	1 to 2 µm	2 to 3 µm	3 to 5 µm	5 to 10 µm	10 to 20 µm
Body Mass 1-5 < Body Mass 6-10 <sup>#</sup>	<i>RR</i>	0.893	0.686*	0.893*	0.893*	0.893	0.043*	0.043*	0.043*	0.080*	0.345*

<sup>1</sup>: The non-parametric differences were considered significant for  $P \leq 0.05$  (bold).

<sup>#</sup>: Divided 10 volunteers into two groups: lightest 5 and heaviest 5, average between both movement sets.

\*: *P*-values represent negative trend between variables.

Table DS6: Impact of volunteer body mass index on average resuspension rate (RR), *P*-values from Wilcoxon non-parametric, two-related samples tests<sup>1</sup>.

	Dust Load: 0.1 g/m <sup>2</sup>					Dust Load: 1.0 g/m <sup>2</sup>					
		1 to 2 µm	2 to 3 µm	3 to 5 µm	5 to 10 µm	10 to 20 µm	1 to 2 µm	2 to 3 µm	3 to 5 µm	5 to 10 µm	10 to 20 µm
BMI 1-5 < BMI 6- 10 <sup>#</sup>	<i>RR</i>	0.686*	0.686	0.893	0.686	0.500	0.043*	0.069*	0.138*	0.043*	0.080*

<sup>1</sup>: The non-parametric differences were considered significant for  $P \leq 0.05$  (bold).

<sup>#</sup>: Divided 10 volunteers into two groups: lowest 5 BMI and greatest 5 BMI, average between both movement sets.

\*: *P*-values represent negative trend between variables.

Table DS7: Impact of each individual movement (M1-M5) on average resuspension rate (RR), *P*-values from Wilcoxon non-parametric, two-related samples tests<sup>1</sup>.

Movement Set 1		Dust Load: 0.1 g/m <sup>2</sup>					Dust Load: 1.0 g/m <sup>2</sup>				
		1 to 2 µm	2 to 3 µm	3 to 5 µm	5 to 10 µm	10 to 20 µm	1 to 2 µm	2 to 3 µm	3 to 5 µm	5 to 10 µm	10 to 20 µm
M1 < M2	R R	0.017	0.012	0.018	0.018	0.012	0.063	0.093	0.237	0.398	0.237
M2 < M3	R R	0.028	0.043	0.063	0.091	0.889 *	0.066	0.139	0.110	0.139	0.515
M3 < M4	R R	0.735	0.735	0.889	0.866	0.917	0.038 *	0.139 *	0.038 *	0.028 *	0.051 *
M4 < M5	R R	0.612 *	0.050 *	0.011 *	0.028 *	0.753 *	0.161 *	0.208 *	0.207 *	0.263 *	0.327
M1 < M3	R R	0.028	0.018	0.043	0.028	0.012	0.012	0.008	0.012	0.017	0.107
M1 < M4	R R	0.063	0.091	0.176	0.463	0.116	0.063	0.069	0.310	0.735	0.866
M1 < M5	R R	0.028	0.128	0.345	1*	0.237	0.176	0.327	0.499	0.612	0.237
M2 < M4	R R	1	1	0.859	0.889	0.735 *	0.779	0.263 *	0.123 *	0.069 *	0.093 *
M2 < M5	R R	0.575 *	0.327 *	0.674	0.314 *	0.889	0.889	0.779	0.674	0.263 *	0.123 *
M3 < M5	R R	0.866 *	0.237 *	0.069 *	0.093 *	1	0.021 *	0.021 *	0.021 *	0.015	0.314 *
Movement Set 2		Dust Load: 0.1 g/m <sup>2</sup>					Dust Load: 1.0 g/m <sup>2</sup>				
		1 to 2 µm	2 to 3 µm	3 to 5 µm	5 to 10 µm	10 to 20 µm	1 to 2 µm	2 to 3 µm	3 to 5 µm	5 to 10 µm	10 to 20 µm
M1 < M2	R R	0.028	0.715	0.012	0.017	0.051	0.021	0.011	0.017	0.038	0.116
M2 < M3	R R	0.051	0.074	0.022	0.037	0.767 *	0.007	0.005	0.009	0.022	0.110
M3 < M4	R R	0.237 *	0.441 *	0.484 *	0.889	0.600	0.445 *	0.575	0.646 *	0.878	0.767 *
M4 < M5	R R	0.735 *	1*	0.735 *	0.612	0.109 *	0.674 *	0.779 *	0.674 *	0.674 *	0.263
M1 < M3	R R	0.028	0.068	0.012	0.012	0.017	0.008	0.008	0.012	0.008	0.018
M1 < M4	R R	0.068	0.593	0.028	0.173	0.046	0.008	0.008	0.012	0.008	0.018

M1 < M5	R	0.109	<b>0.109</b> *	0.116	0.173	0.225	0.018	0.018	0.028	0.018	0.028
M2 < M4	R	0.093	<b>0.374</b> *	0.263	<b>0.575</b> *	0.735	0.028	0.037	0.047	0.022	0.069
M2 < M5	R	0.499	1	0.779	0.401	0.600	0.484	0.575	0.401	0.779	0.214
M3 < M5	R	0.116	<b>0.327</b> *	<b>0.208</b> *	<b>0.161</b> *	0.600	<b>0.161</b> *	<b>0.208</b> *	<b>0.327</b> *	<b>0.263</b> *	<b>0.260</b> *

<sup>1</sup>: The non-parametric differences were considered significant for  $P \leq 0.05$  (bold).

\*:  $P$ -values represent negative trend between variables.

## Appendix E

## Paper E. Infant Exposure to Emissions of Volatile Organic Compounds from Crib Mattresses

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Published in *Environmental Science & Technology*, 48(6): 3541-3549

### ABSTRACT

Infants spend most of their time sleeping and are likely to be exposed to elevated concentrations of chemicals released from their crib mattresses. Small-scale chamber experiments were conducted to determine the area-specific emission rates (*SERs*) of volatile organic compounds (VOCs) in a collection of twenty new and used crib mattresses. All mattress samples were found to emit VOCs and the mean values of total VOC (TVOC) *SERs* were 56  $\mu\text{g}/\text{m}^2\text{h}$  at 23°C and 139  $\mu\text{g}/\text{m}^2\text{h}$  at 36°C. TVOC *SERs* were greater for new mattresses compared to used ones and were influenced by the type of foam material and the presence of mattress cover layer. A variety of VOCs were identified, with polyurethane foam releasing a greater diversity of VOCs compared to polyester foam. Large-scale chamber experiments were conducted with an infant thermal manikin. TVOC concentrations sampled in the breathing zone and interior pore air of the crib mattress foam were found to be greater than the bulk room air by factors in the range of 1.8 to 2.4 and 7.5 to 21, respectively. The results suggest that crib mattresses are an important source of VOCs and infant exposure to VOCs are possibly elevated in their sleep microenvironments.

## INTRODUCTION

Infants spend a considerable amount of time sleeping, typically ranging from 12 to 13 hours/day in their first three years of life, significantly more than adults (average of 8.2 hours/day) (Figure ES1, Table and Figure numbers preceded by an "S" are in the Supporting Information). Sleep durations greater than 14 hours/day during the first year of life are also commonly reported (Diez et al. 2000, Iglowstein et al. 2003). The length of the sleep period makes infant sleep microenvironments particularly important in contributing to both their acute and chronic exposures to various gaseous indoor air pollutants, particularly those originating in the crib mattresses on which they sleep.

The composition of a typical crib mattress includes a thick layer of polyurethane or polyester foam padding (inner-springs are also used) encased within a thin, waterproof plastic cover to protect the mattress foam and to provide an easy-to-clean surface. The use of petroleum-derived polyurethane and plastic materials, along with various adhesives and additives used in the manufacturing and assembly processes, suggests that crib mattresses are a likely source of volatile organic compounds (VOCs) (Rothe et al. 2001, Zhao et al. 2004). Anderson and Anderson (1999, 2000) identified a variety of VOCs released from one crib mattress sample and one waterproof crib mattress cover sample, many of which were solvents or chemicals likely used in the manufacture of the crib mattress. The emissions were found to cause sensory irritation, pulmonary irritation, and decreases in mid-expiratory airflow velocity in mice. Furthermore, VOC emissions from consumer and building products have been shown to increase with temperature (Haghighat et al. 1998, Lin et al. 2009,

Masuck et al. 2011, VanderWal et al. 1997), suggesting that VOC emissions from crib mattresses may increase due to localized elevations in mattress surface temperature caused by heat transfer from a sleeping infant (Wheldon 1982, Vuillerme et al. 1998, Elabbassi et al. 2001).

Infants' inhaled dose of VOCs originating in their crib mattresses may be augmented due to the significant amount of air they inhale per body mass and the impact of the source-proximity effect. The volume of air inhaled/kg-day can be estimated by taking the product of the mean normalized volumetric breathing rate during a sleep or nap activity (U.S. EPA 2009) and the mean duration of time spent in a sleep or nap activity (Figure ES2). Infants inhale about 250 to 300 L/kg-day, nearly an order of magnitude more air per body mass than adults. Additionally, the source-proximity effect, in which pollutant concentrations near a source are greater than those in the bulk room air, may also lead to elevated infant inhalation exposure to VOCs. There are several key factors that likely influence the source-proximity effect, including the spatial proximity of an infant's breathing zone (BZ) to the crib mattress, incomplete mixing of bedroom air, concentrations gradients near an actively emitting crib mattress, and the buoyant thermal plume around an infant (Mage and Ott 1996, Furtaw et al. 1996, McBride et al. 1999, Rim and Novoselac 2009 and 2010, Acevedo-Bolton et al. 2012, Laverge et al. 2013). Laverge et al. (2013) simulated the release of gaseous pollutants from an adult mattress with an inert tracer gas and found the BZ concentrations for an adult thermal manikin to be significantly greater than those measured in the bulk air, typically by a factor of 1.1 to > 2, depending on sleep position and bedding arrangement.

Associations between VOC exposure and poor respiratory health effect have been observed among infants. Early life exposures to VOCs may impact the developing immune system and increase the risk of allergic disease in young children, even at low concentrations (Franklin 2007 and references therein). Diez et al. (2000) found that the presence of VOCs (i.e., styrene, benzene) in a bedroom increased the risk of pulmonary infections in six-week old infants. Among a cohort of children 6 months to 3 years of age, Rumchev et al. (2004) found numerous VOCs (i.e., benzene, toluene, ethylbenzene, m-xylene) to be a significant risk factor for asthma, with every 10  $\mu\text{g}/\text{m}^3$  increase in toluene and benzene increasing the risk of having asthma by a factor of 2 and 3, respectively. Additionally, studies have shown that the primary exposure route of VOCs in infants is through inhalation of indoor air, 25 to 135 fold higher than through ingestion of a mother's breast milk (Kim et al. 2007). Strachan and Carey (1995) surmised that VOCs released from pillows in close proximity to the BZ might increase mucosal permeability to inhaled allergens, which the authors believe may explain the association between the use of foam pillows and childhood asthma. Furthermore, Abraham et al. (2005) applied a pharmacokinetic model to compare internal exposures to a common VOC, styrene, between infants and adults. The arterial blood concentration in newborns and 1 year old infants was found to be approximately 1.2 to > 3 times higher than that of an adult exposed to the same airborne concentration. The authors attributed these findings to the relatively high alveolar ventilation rate and immature metabolism in infants.

Therefore, there is a need to understand the role of crib mattresses as a source of VOCs in infant sleep microenvironments, given the potential for elevated infant exposure to VOCs that may volatilize from this source and the health implications of early life exposure to VOCs. The objectives of this paper are to: measure the area-specific emission rate (*SER*) of VOCs from crib mattresses in small-scale chambers; determine the parameters that may have an influence on *SERs*; identify common VOCs emitted from crib mattresses; and measure the VOC BZ concentration in a large-scale chamber with an infant thermal manikin to explore the source-proximity effect.

## MATERIALS AND METHODS

### *Material Selection*

Twenty crib mattresses, of varying usage, quality, and material composition, were selected for this study (Table ES1). Nine new crib mattresses made by different manufacturers were purchased from an online retail store. Eleven used crib mattresses were obtained through donations in Austin, Texas and Helsinki, Finland. The used crib mattresses were manufactured in various years between 1993 and 2009, were actively used for different durations, and were stored in different indoor spaces, including bedrooms, closets, and attics. The foam layers of the mattresses were manufactured out of polyurethane foam (PUF), polyester foam (Poly.), or polyurethane foam with a fraction of soy-derived foam.

### *SER Experiments in Small-Scale Chambers*

The crib mattress samples were cut to the appropriate size (14.25 Ø × 7.5 cm) to fit within the small-scale chamber (Figure E1). The edges of the samples were sealed with aluminum foil and low-VOC aluminum tape to minimize edge effects. Following sample preparation, the samples were conditioned in the laboratory at 23°C and 50% relative humidity (RH) for at least one month prior to the VOC *SER* experiments. All new samples were tested approximately six months after they were manufactured, thereby avoiding the peak emission period and better representing long-term emissions.

Small-scale emission chambers (Figure E1) were used to measure the *SER* of VOCs from the crib mattress samples. The small-scale chamber included a field and laboratory emission cell (FLEC) (0.035 L) mounted to a 2.5 L cylindrical stainless steel emission chamber. The small-scale chamber was supplied with clean, purified air at 23±1°C and 50±2% RH. Table ES2 provides an overview of the operational parameters. The area specific airflow rate ( $q$ , m/h) for the VOC *SER* experiments was 1.52 m/h, and was selected to be equivalent to the large-scale chamber experiments. Translating to a standard-size 14.4 m<sup>3</sup> bedroom, this area specific airflow rate equates to a bedroom air exchange rate of 0.16 h<sup>-1</sup>, representative of ventilation levels in actual children's bedrooms (Bekö et al. 2010). Prior to the emissions test, all components of the small-scale emissions chamber were thoroughly cleaned with methanol and placed in an oven at 70°C overnight for approximately 12 hours.

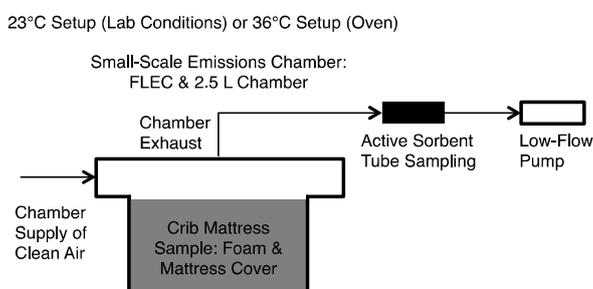


Figure E1 (a.)

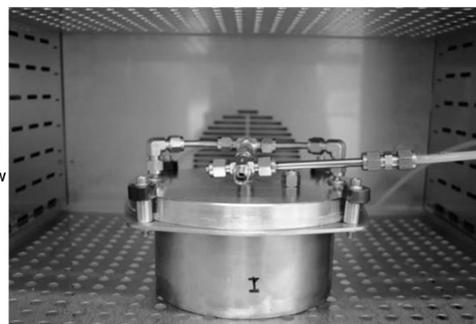


Figure E1 (b.)

Figure E1: (a.) Schematic and (b.) photo of the experimental setup for small-scale chamber VOC *SER* experiments.

Active air sampling was performed at the exhaust of the small-scale emission chambers using sorbent tubes packed with Tenax® TA and a low-flow pump (Gilliam Low Flow Sampler, Model LFS-113DC, Sensidyne LP). Backup tubes were connected to each primary sampling tube to check for breakthrough. Prior to a test, an air sample was taken from the empty chamber to ensure the background total VOC (TVOC) concentration was below  $5 \mu\text{g}/\text{m}^3$ . The crib mattress was then placed in the chamber for approximately 20 hours before sampling, to allow it to reach thermal equilibrium with the chamber (at  $36^\circ\text{C}$ ) and to avoid short-term peak emissions. The experiments were carried out at a temperature of  $23^\circ\text{C}$  and  $36^\circ\text{C}$  and, in all cases, steady-state VOC concentrations were achieved during air sampling (Figure ES3). Additionally, three crib mattresses (samples 2, 8, and 20) were tested without the mattress cover layer. These experiments provided insight into the role of the mattress cover in acting as a barrier to the transport of VOCs originating from the foam layer and the

contribution of the cover to the total VOC SER from the entire crib mattress. In total, 46 VOC SER experiments were performed.

### *Large-Scale Chamber Experiments*

Full-scale chamber experiments were conducted in a 4.5 m<sup>3</sup> emission chamber with a new full-size crib mattress (sample 6, polyurethane foam) and an infant thermal manikin (Figure E2, Table ES2). The aim of these experiments was to simulate a semi-realistic exposure scenario and experimentally determine the BZ concentration for VOCs emitting from a crib mattress to explore the impact of the source-proximity effect.

A simplified infant thermal manikin was constructed using a hollow galvanized steel cylinder (oval cross section, 50×15×12 cm), with heating elements placed uniformly inside. The cylinder was wrapped with aluminium foil and aluminium tape and the edges were sealed. The manikin was heated for approximately four weeks to bake out any VOCs originating within the cylinder materials. Background TVOC concentrations in the empty full-scale chamber with the heated thermal manikin were found to be below 1 µg/m<sup>3</sup>. To simulate the convective airflow around a sleeping infant, the surface temperature of the manikin was set to 35.6±0.7°C, the approximate skin surface temperature of a sleeping infant (Wheldon 1982, Vuillerme et al. 1988, Elabassi et al. 2002), via a power input of 9.625 W for an approximate heat dissipation per unit manikin surface area of 40 W/m<sup>2</sup> (Varivolt Metric Power Supply). The uniformity of the

heat flux was verified by taking four surface temperature measurements with thermistors (44000 Series, Omega Engineering).

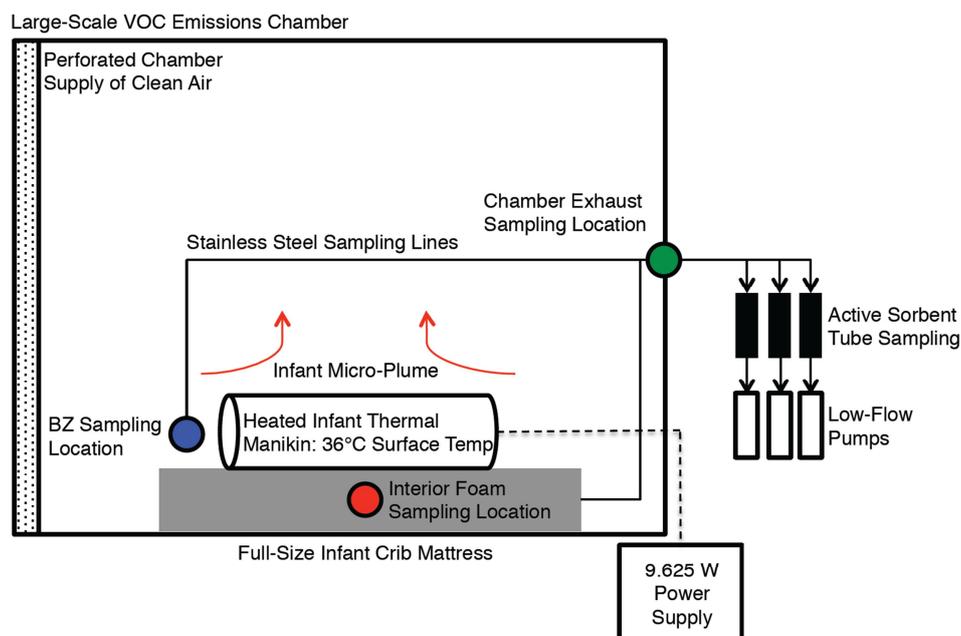


Figure E2: Schematic of the experimental setup for large-scale VOC infant manikin experiments.

The large-scale emissions chamber was supplied with clean, purified air at  $23\pm 1^\circ\text{C}$  and  $50\pm 2\%$  RH. Experiments were first conducted to evaluate the “sleeping infant micro-plume” generated by the thermal manikin (measured with omnidirectional anemometers, Model HT-400, Sensor). Three cases were considered: manikin heating on with mattress cover, manikin heating on without mattress cover, and manikin heating off without mattress cover. The crib mattress (sample 6) was conditioned at laboratory conditions ( $23^\circ\text{C}$  and 50% RH)

for one-month prior the full-scale emissions test. Upon placing the crib mattress and infant thermal manikin in the chamber, conditions were allowed to equilibrate for at least 24 h prior to sampling. VOC concentrations were sampled simultaneously at three locations with stainless steel tubing and fittings: interior of the crib mattress foam, at the centre of the foam block; BZ, 2.5 cm above the mattress surface, 5 cm from the top edge of the manikin, and 3 cm from the side edge of the manikin; and the chamber exhaust, representing the bulk room air. For each of the three cases, four to six samples were taken at each of the three locations, over a period of two to three days. Steady-state VOC concentrations were achieved for all cases.

#### *Analytical Methods*

The sorbent tubes were thermally desorbed at 260°C for 6 min (cold trap temperature of -30°C) using a thermal desorber (TurboMatrix ATD, PerkinElmer, Inc.) and analyzed with a gas chromatograph (GC) connected to a mass spectrometer detector (MSD) and flame ionization detector (FID) (Clarus 500, PerkinElmer, Inc.). The gas chromatograph was equipped with a double-capillary column (column: 50 m, 0.2 mm ID, film thickness: 0.5 µm) (HP-PONA, Agilent Technologies, Inc.) and the sample was split 1:1 between the two columns, which were connected to the MSD and FID detectors. The temperature program of the GC oven was as follows: 40°C (5 min hold), ramp 1 (6°C/min) to 280°C (5 min hold), ramp 2 (45°C/min) to 40°C (1 min hold), with a total run time of 51 min. MSD in SCAN mode was used to identify single VOCs and individual peaks were compared with a mass spectral library (NIST08).

Identification is based on a minimum of a 75% match, and therefore, should be regarded as a qualitative best estimate since reference standards were not used. The FID response was used for quantification. VOC concentrations ( $C_{VOC}$ ,  $\mu\text{g}/\text{m}^3$ ) were calculated as toluene equivalents (ISO 16000-6) with a bias error of  $\pm 10\%$ . TVOC concentrations were calculated as the total integrated FID signal between hexane and hexadecane. An external toluene standard (100 ng of toluene) was used for calibration (one per ten sorbent tubes analyzed). The VOC *SER* was calculated as:

$$SER_{VOC}(\mu\text{g}/\text{m}^2\text{h}) = \left[ \overline{C_{VOC}}(\mu\text{g}/\text{m}^3) - C_{VOC,Bkg.}(\mu\text{g}/\text{m}^3) \right] \times q(\text{m}/\text{h}) \quad (\text{E.1})$$

where  $\overline{C_{VOC}}$  ( $\mu\text{g}/\text{m}^3$ ) is taken as the average concentration over the 10 h sampling period (based on 3 to 5 samples) and  $C_{VOC,Bkg.}$  ( $\mu\text{g}/\text{m}^3$ ) is the background concentration for an empty chamber. To determine the uncertainty in the VOC *SER*, the error in measuring both  $C_{VOC}$  and  $q$  was propagated. For  $C_{VOC}$ , the precision error (standard deviation) based on the 3 to 5 repeated concentration measurements taken at steady-state chamber conditions was combined with an instrument-calibration bias error of  $\pm 10\%$  in  $C_{VOC}$ . The instrument error for the bubble flow meter was  $\pm 1\%$ .

## RESULTS

### *Small-Scale Chamber Experiments and TVOC SER*

The results of the small-scale chamber experiments are presented in Figure E3, with the TVOC *SERs* categorized by temperature, usage, and foam

material. The TVOC *SERs* are plotted on a logarithmic axis to improve visualization of the data and the error bars represent the calculated error in the *SER*. The detailed results with measurement uncertainties are also listed in Table ES3. All new and used crib mattresses were found to emit VOCs. Across all twenty samples, TVOC *SERs* ranged from 3 to 385  $\mu\text{g}/\text{m}^2\text{h}$  (mean of 56  $\mu\text{g}/\text{m}^2\text{h}$ ) at 23°C and from 8 to 697  $\mu\text{g}/\text{m}^2\text{h}$  (mean of 139  $\mu\text{g}/\text{m}^2\text{h}$ ) at 36°C. TVOC *SERs* in this range are similar to emissions from other consumer products and building materials (Lundgren et al. 1999, Wiglusz et al. 2002, Salthammer et al. 2003, Järnström et al. 2007, Masuck et al. 2011, An et al. 2011) that may be found in an infant bedroom or nursery, such as soft polyurethane foam (< 10  $\mu\text{g}/\text{m}^2\text{h}$  at 23°C and 45%RH), PVC flooring (108  $\mu\text{g}/\text{m}^2\text{h}$ ), laminate flooring (154 to 249  $\mu\text{g}/\text{m}^2\text{h}$ , depending on floor temperature), parquet floor covering (80  $\mu\text{g}/\text{m}^2\text{h}$ ), wall coverings (51  $\mu\text{g}/\text{m}^2\text{h}$ ), and plastic toy materials (4.1 to 1,080  $\mu\text{g}/\text{m}^2\text{h}$  at 23°C and 50%RH).

VOC emissions increased with temperature, as shown in Figures E3 and ES4. The TVOC *SER* at 36°C is about double that at 23°C (Figure ES4). The temperature dependence of the VOC emissions follows a similar trend as previous emission studies (van der Wal et al. 1997, Haghghat and De Bellis 1998, Lin et al. 2009, Masuck et al. 2011, An et al. 2011). A VOC's vapor pressure tends to increase with temperature. At 36°C, a VOC molecule has a greater affinity for the gas phase, thereby enhancing its partitioning from the solid phase (mattress cover or foam) to the chamber air. The temperature dependence is particularly relevant to the infant sleep microenvironment, because heat released from a sleeping infant will warm surrounding objects, including their mattress. Thus,

VOC emissions may increase due to elevations of localized mattress surface temperature.

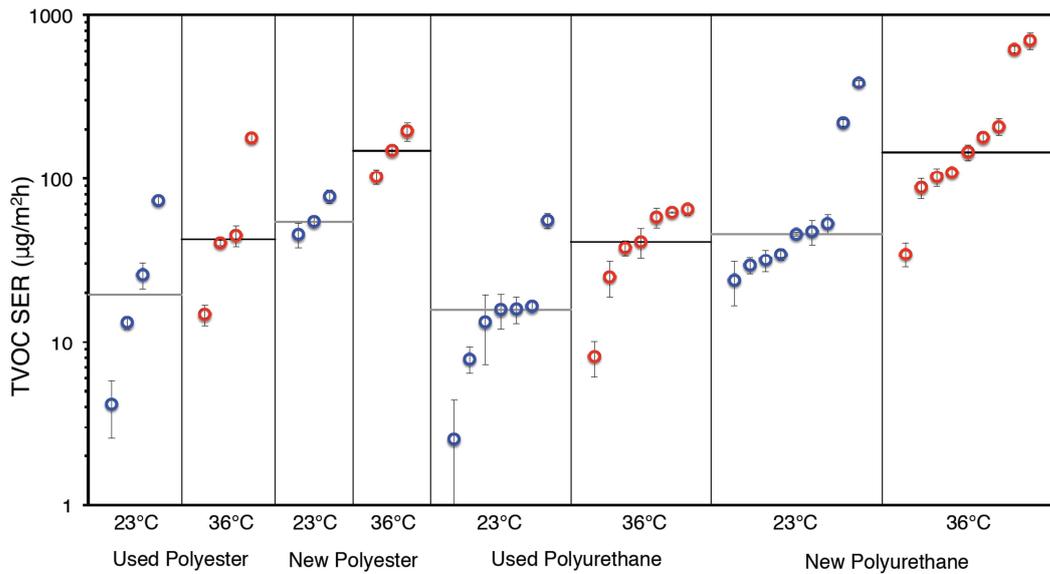


Figure E3: TVOC *SER* categorized by temperature, usage, and foam material (plotted on logarithmic axis).

TVOC *SERs* were greater for the nine new crib mattress samples (mean of 87.1 µg/m²h at 23°C, mean of 218.8 µg/m²h at 36°C) compared to the eleven used samples (mean of 22.1 µg/m²h at 23°C, mean of 52.1 µg/m²h at 36°C) by factors of 3.9 and 4.2 at 23°C and 36°C, respectively. VOC emissions tend to decay as the material-phase concentration in the source depletes over time. Lundgren et al. (1999) showed that TVOC *SERs* of PVC flooring can decrease about 60% from 4 to 26 weeks after manufacture. As previously discussed, the

new samples were tested approximately six months after they were manufactured, thereby avoiding the peak emission period and better representing long-term emissions. Lower TVOC *SERs* among the used samples suggests the re-use of an older crib mattress or an extended “air out” period of a new mattress may be desirable as a preventive approach to reduce infant VOC exposures. However, crib mattress re-use must be considered carefully, because older crib mattresses may contain toxic substances (i.e., flame retardants) that have been banned for a period of time.

Mattress samples 2, 8, and 20 were also tested without the mattress cover layer. For samples 2 and 20, the TVOC *SER* increased as the mattress cover was removed (Table ES3). This suggests that the crib mattress cover may act as a barrier to VOCs originating in the foam layer. Its effectiveness depends on the diffusion coefficients of the mattress cover for VOCs, which are influenced by both porosity and tortuosity of the material, as well temperature (Xiong et al. 2008). In general, the lower the diffusion coefficient, the more effective the layer will be in preventing transport of VOCs from the underlying foam layer (Yuan et al. 2007). Despite the findings for samples 2 and 20, the TVOC *SER* decreased for sample 8 when the cover layer was removed (Table ES3). Thus, for some crib mattresses, the mattress cover layer may be a more significant source of VOCs than the foam layer. Lastly, the interior surface of the mattress cover may also act as a sink to VOCs that volatilize from the underlying foam layer. Both the adsorption of VOCs to the cover, and desorption from the cover, is a temperature-dependent process (Bodalal et al. 2001, Zhang et al. 2002a, 2002b, Deng et al. 2009, 2011). Thus, the dynamic VOC sorption process will be

influenced by the repeated heating and cooling of the mattress cover during occupied and unoccupied periods on the crib mattress, respectively.

The foam material, polyester or polyurethane, was also found to influence TVOC *SERs*. TVOC *SERs* from new samples with polyurethane foam (mean of 70.0  $\mu\text{g}/\text{m}^2\text{h}$  at 23°C, mean of 197.7  $\mu\text{g}/\text{m}^2\text{h}$  at 36 °C) were slightly greater than those with polyester foam (mean of 59.1  $\mu\text{g}/\text{m}^2\text{h}$  at 23°C, mean of 148.6  $\mu\text{g}/\text{m}^2\text{h}$  at 36°C). The three highest TVOC *SERs* were found for sample 2 (polyurethane with and without cover layer) and sample 20 (polyurethane without cover layer). No association was observed between TVOC *SER* and the quality (i.e., retail cost in USD) of the new crib mattresses.

Additional factors, such as relative humidity, chamber design, and sink effects may also influence VOC emissions from materials to varying extents; however, their impact on crib mattress emissions were not investigated here. Relative humidity is particularly relevant to the infant sleep microenvironment due to elevated moisture levels that may occur as the infant breathes and perspires. Emission studies have shown that VOC *SERs* tend to increase with increasing relative humidity (Haghighat and De Bellis 1998, Lin et al. 2009). Afshari et al. (2003) demonstrated that chamber size and design does not have a strong impact on VOC *SERs*, with similar *SERs* determined for a FLEC, CLIMPAQ (Chamber for Laboratory Investigation of Materials Pollution and Air Quality), and a 1 m<sup>3</sup> emission chamber. However, inter-laboratory studies have shown significant variation in VOC emission rates, suggesting lab-specific chamber setup, sampling techniques, and analytical methods may influence

emission results (Howard-Reed et al. 2011). Therefore, the *SERs* reported in this investigation might be specific to the experimental methods in which they are determined. Lastly, sink effects may influence *SERs* for crib mattresses, especially if bedding is used to wrap the crib mattress. VOCs released from the underlying crib mattress may adsorb to the bedding over time, thereby impacting airborne VOC concentrations.

### *VOC Speciation*

The most abundant VOCs were identified and their individual VOC *SERs* are reported in Table E1. Table ES5 provides a complete list of VOCs identified (some compounds having boiling points > 240°C, can be classified as semi-VOCs (SVOCs)). *SERs* varied widely among different compounds and crib mattress samples; they ranged from < 1 to 62 µg/m<sup>2</sup>h at 23°C and from 2 to 257 µg/m<sup>2</sup>h at 36°C. Nearly all VOC *SERs* increased with temperature from 23°C to 36°C. Some VOCs exhibited a greater temperature dependence, including phenol, 2-ethyl-hexanoic acid, palmitic acid, and 2,6-bis(1,1-dimethylethyl)-4-(1-oxopropyl)phenol, whereas other VOCs were less sensitive to the temperature increase, including 3-methyl-1-heptanol and isooctanol. The temperature dependence of *SER* for a particular VOC likely depends on a combination of factors, including the VOC's boiling point, material-air partition coefficient, material-phase diffusion coefficient, and the way the VOC is incorporated into the solid matrix of the material.

Table E1: VOC SERs of the most abundant and commonly identified VOCs among new and used infant crib mattresses in small-scale chamber experiments.

Compound <sup>1</sup>	New/ Used	Number of Samples		VOC SER at 23°C (µg/m <sup>2</sup> h)	VOC SER at 36°C (µg/m <sup>2</sup> h)
		PUF	Poly.		
Phenol	New	5	1	< 1 – 62	3 – 257
	Used	1	1		
Isooctanol	New	1	1	< 1 – 6	4 – 7
	Used	1	--		
Neodecanoic acid	New	--	3	3 – 22	9 – 40
	Used	--	1		
Hexanoic acid, 2-ethyl-	New	5	--	< 1 – 55	5 – 213
	Used	1	1		
1-Heptanol, 3-methyl	New	2	2	7 – 21	7 – 22
	Used	--	--		
D-Limonene	New	2	--	4 – 11	9 – 18
	Used	--	--		
2,6-Bis(1,1-dimethylethyl)-4-(1-oxopropyl)phenol	New	2	--	4 – 14	12 – 61
	Used	--	--		
(S)-3-Ethyl-4-methylpentanol	New	1	1	2 – 6	3 – 9
	Used	1	--		
Linalool	New	2	--	3 – 41	10 – 44
	Used	--	--		
Nonanal	New	1	--	< 1 – 5	2 – 10
	Used	2	4		
Decanal	New	1	--	< 1 – 5	2 – 10
	Used	1	1		
Isopropyl Myristate	New	--	--	< 1 – 3	3 – 11
	Used	3	2		
Palmitic acid	New	--	--	2 – 10	12 – 43
	Used	1	1		
2-Ethylhexanol	New	1	--	3 – 6	7 – 8
	Used	1	1		

<sup>1</sup>: All compounds identified with at least 75% match with NIST mass spectral library.

The impact of foam material is evident in regard to the specific VOCs released from the crib mattress samples. Polyurethane foam showed a greater diversity of VOCs, many of which were not identified in polyester foam (or identified much less frequently), including phenol, 2-ethyl-hexanoic acid, d-

limonene, and linalool. Phenol is used as an intermediate in the production of phenolic resins (U.S. EPA 2002) that may be used in the production of polyurethane foam; 2-ethyl-hexanoic acid is used as a catalyst in the production of polyurethane foam (EHC 2011); d-limonene is commonly used as a fragrance, as well as a solvent and wetting agent in the manufacture of resins (Lewis 2001); and linalool is used as a fragrance (Lewis 2007). Neodecanoic acid, a carboxylic acid that is used as a polymerization initiator, was only associated with polyester foam crib mattress samples. Other compounds detected include isooctanol and 2-ethylhexanol, which are used in the production of plasticizers, as an intermediate for resin solvents, emulsifiers, and antifoaming agents (Lewis 2001). Similar compounds and TVOC *SERs* were detected for the polyurethane foam samples manufactured with a fraction of soy-derived foam (samples 7 and 8) as with the normal polyurethane foam samples; however, dimethylformamide, a solvent used in production of polyurethane products (CA OEHHA 2000), was only detected in the soy-based sample 8 (Table ES5).

Numerous VOCs were more commonly detected among new crib mattress samples, including phenol (6 new, 2 used), 2-ethyl-hexanoic acid (5 new, 2 used), 3-methyl-1-heptanol (4 new, 1 used), d-limonene (2 new, 0 used), neodecanoic acid (3 new, 1 used), and linalool (2 new, 0 used). D-limonene, a terpene, and linalool, a terpene alcohol, were only detected among new mattress samples, suggesting that the used samples were nearly depleted of these compounds, which are highly reactive with ozone (Nazaroff and Weschler 2004). In addition, several VOCs were detected more frequently among used samples, including nonanal (6 used, 1 new), isopropyl myristate (5 used, 0 new), and

palmitic acid (2 used, 0 new). Nonanal, which was detected in 6 of the 11 used samples, is an alkyl aldehyde that is released by human skin (Gallagher et al. 2008) and also found in building materials (Järnström et al. 2006), while isopropyl myristate and palmitic acid are fatty acid esters that can be found in personal care products, such as lotions and ointments (Fulton et al. 1982, Cardoso et al. 2006). Because polyurethane foam has a high sorption capacity and can adsorb airborne VOCs over time (Zhao et al. 2004), the compounds detected among the used samples may represent adsorbed VOCs that originated elsewhere in a residence rather than VOCs used in the production of the mattress.

#### *Large-Scale Chamber Experiments*

As shown in Figure E4 and Table ES4, TVOC concentrations sampled at the BZ were significantly greater than those of bulk air sampled at the chamber exhaust. BZ concentrations ranged from 14.2 to 33.2  $\mu\text{g}/\text{m}^3$  (mean of 26.2  $\mu\text{g}/\text{m}^3$ ), the exhaust concentrations ranged from 8.0-18.4  $\mu\text{g}/\text{m}^3$  (mean of 13.5  $\mu\text{g}/\text{m}^3$ ), and the ratio of the BZ concentration to the exhaust concentration ranged from 1.7 to 2.4 (mean of 2), depending on the particular case. These results suggest that there is a source-proximity effect associated with exposure to VOCs released from crib mattresses. It can be explained by the development of VOC concentration gradients around the emission source (crib mattress), the close proximity of the BZ sampling location (2.5 cm above the mattress surface), and the incomplete mixing of chamber air. Indeed, these ratios (i.e., concentration ratios of BZ to bulk air) follow the same trend as those reported by

Laverge et al. (2013) for a tracer gas and Boor et al. (2015a) for particles resuspended from mattresses (range of 1.07 to 1.94). However, caution should also be taken when extrapolating the experimental results to actual infant bedroom environments, because different air exchange rates and airflow distributions may significantly influence the results.

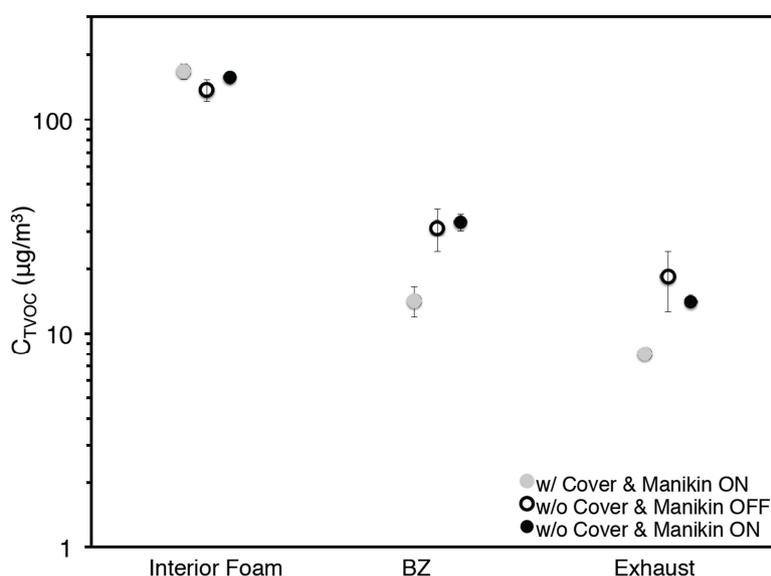


Figure E4: Spatial distribution of TVOC concentrations in large-scale emissions experiments with infant thermal manikin. Error bars represent the calculated error in  $C_{TVOC}$ .

Switching on the heat output of the infant manikin resulted in a small increase in TVOC BZ concentrations (difference within instrument error), although the ratio of the BZ concentration to the exhaust increased from 1.7 to 2.4. Because the manikin may only raise the surface temperature for a small fraction of crib mattress, the increase in emissions is not as pronounced as when the entire mattress sample in the small-scale chamber is thermally conditioned at

36°C. In addition, TVOC BZ concentrations may also be influenced by the micro-plume generated by the heated infant manikin. Sharp velocity increases above the upper surface of the manikin was observed as the buoyant plume develops (Figure ES5). Thermal plumes may serve two competing roles: effectively transporting pollutants, released from various locations in close vicinity to the human body, vertically upward, toward to BZ (Rim and Novoselac 2009, 2010) and entraining “fresh” air exterior to the BZ region, thereby diluting concentrations of gaseous pollutants and improving mixing conditions (Laverge et al. 2013). Therefore, as an infant sleeps, heat dissipation from their bodies may elevate the crib mattress surface temperature, thereby increasing VOC emissions, and generate a thermal plume that aids both in delivering VOCs upward to the BZ and diluting BZ concentrations by entraining bulk room air.

TVOC concentrations sampled in the interior foam at the center of the crib mattress (range of 137.9-168.4  $\mu\text{g}/\text{m}^3$ , mean of 154.4  $\mu\text{g}/\text{m}^3$ ) were found to be nearly an order of magnitude greater than those in the BZ region or bulk room air. The very high pore concentration may have important implications for infant exposure. As the porous and sponge-like mattress foam is compressed and decompressed in a cyclic manner due to infant body movements, the pore air may be released in short-term bursts, thereby elevating short-term BZ concentrations. The impact of the “mattress pumping effect” requires further investigation and emphasizes the dynamic interaction between source and receptor in the sleep microenvironment. Furthermore, removing the crib mattress cover increased the BZ and bulk air TVOC concentrations, while slightly reducing the interior foam concentration (Table ES4). Thus, depletion of

VOCs in the pore air led to an elevation in airborne TVOC concentrations. Given these findings, as well as those for the VOC *SER* experiments, for particular products where the foam is a stronger VOC source relative to the cover, the mattress cover may play an important role in acting as a barrier to the transport of VOCs originating within the foam layer and more saturated pore air. Although not investigated here, adult mattresses typically do not include a plastic mattress cover layer, rather a porous fabric mattress encasing. Thus, VOCs originating in polyurethane foam may be more easily released into the adult sleep microenvironment.

#### *Estimation of Infant Inhalation Exposure*

The BZ TVOC concentrations can be applied in a simple inhalation exposure analysis to estimate sleeping inhalation doses for infants. The daily inhalation intake dose (Zartarian et al. 1997) can be estimated as the product the BZ TVOC concentration and the volume of air inhaled/kg-day during a sleep period (Figure ES2). Assuming BZ TVOC concentrations of 26.2  $\mu\text{g}/\text{m}^3$  from the large-scale emission chamber experiments (mean of three cases considered), the daily sleeping inhalation dose for VOCs originating in a crib mattress for infants would be approximately 8  $\mu\text{g}/\text{kg}\text{-day}$  from birth to 1 year of age and 6.4  $\mu\text{g}/\text{kg}\text{-day}$  for 2 years of age. Thus, infants may receive doses on the order of 1  $\mu\text{g}/\text{kg}\text{-day}$  of the sum of all VOCs that may be released from their crib mattresses. For comparison, inhalation doses on this order of magnitude are greater than those reported by Masuck et al. (2011) for infant exposure to VOC emissions from toy fragrances (range of 2.2 to 220  $\text{ng}/\text{kg}\text{-day}$ ) and on the same order of magnitude

as those reported by Kim et al. (2007) for toluene (4.5 µg/kg BW/day) and benzene (1 µg/kg BW/day) in living room air. Furthermore, if an infant and adult are exposed to the same BZ concentration of a VOC released from a mattress, the infant normalized dose will be an order of magnitude greater than that of the adult (Figure ES2), emphasizing the seriousness of early life exposures to gaseous indoor air pollutants. Lastly, the infant thermal plume, along with the airflow patterns within an infant's crib and bedroom, will impact BZ concentrations, and thus, the inhalation intake dose. More research is needed to fully characterize airflow regimes in infant sleep microenvironments and how they impact infant exposure to gaseous and particulate pollutants originating in an infant's crib mattress and bedding.

#### DISCUSSION: BEYOND VOCs

In this study, small-scale chamber experiments were conducted to measure the emissions of VOCs from crib mattresses and to identify the most abundant compounds. Large-scale chamber experiments were then performed with a heated infant thermal manikin to explore the impact of the source-proximity effect on infant exposure to VOCs originating in their crib mattresses. The results suggest that crib mattresses are an important source of VOCs and infant exposure to VOCs are possibly elevated in sleep microenvironments. Therefore, efforts should be made to mitigate infant exposure to VOCs originating in their crib mattress through careful selection of low-emitting materials used in the manufacture of the mattress, including various foams, plastics, solvents, and adhesives.

In addition to VOCs, crib mattresses may contain semi-volatile organic compounds (SVOCs), including plasticizers, flame retardants, and unreacted isocyanates. Most crib mattresses have a vinyl cover for waterproofing and antibacterial purposes, and phthalates are extensively used as plasticizers to enhance the softness and flexibility of the cover. Because they are not chemically bound to the polymer matrix, they slowly volatilize from the material and migrate into surrounding environments (Xu and Little 2006, Xu et al. 2012). In 2009, the Consumer Product Safety Improvement Act was enacted in the U.S., placing restrictions on phthalates (i.e., bis(2-ethylhexyl) phthalate (DEHP), dibutyl phthalate (DBP), and benzyl butyl phthalate (BBP)) in toys and child care articles (U.S. CPSC 2008). However, there is some debate as to whether or not crib mattresses are included in the definition of “child care articles” (U.S. CPSC 2009, NRDC 2009). Furthermore, the ban on diisononyl phthalate (DINP) and diisodecyl phthalate (DIDP) only applies to children’s toys that can be placed in a child’s mouth. Alternative plasticizers, such as diisononyl cyclohexane-1,2-dicarboxylate (DINCH), have emerged very recently, but toxicological information is limited. As a result, various phthalate plasticizers and their alternatives were found in crib mattress covers (Boor et al. 2015b).

Crib mattresses manufactured with polyurethane foam are highly flammable and require flame retardant additives (commonly 1 to 15% by weight) to meet various flammability standards (Stapleton et al. 2009, 2011). For several decades, polybrominated diphenyl ether (PBDE) flame retardants, specifically pentaBDEs, were commonly added to crib mattress foam until industry ended

production in the U.S. in 2004. Flame retardants currently used in crib mattresses include various organophosphates and industry mixtures (Stapleton et al. 2011). There is limited published data available regarding the emissions of these compounds (Kemmler et al. 2003). Additionally, unreacted isocyanates may be present in crib mattresses if excess levels of toluene diisocyanate are used in the production of the polyurethane foam (Krone et al. 2003, CA OEHHA 2010).

Therefore, a clear understanding of infant exposure to these emerging SVOC contaminants emitted from crib mattresses, which may disproportionately affect infants and children and lead to lifelong illnesses and disabilities, is urgently needed. The experimental data and the methodology developed in this study will help to achieve this goal.

#### AUTHOR CONTRIBUTIONS

B.E.B., H.J., A.N., and Y.X. defined the overall scope of the study and developed the experimental plan. B.E.B. conducted the experiments and analyzed the data. B.E.B. wrote the paper. Y.X., A.N., and H.J. provided guidance in the execution of the experiments, advised in the interpretation of the results, and provided detailed comments on draft manuscripts.

SUPPORTING INFORMATION

Figures

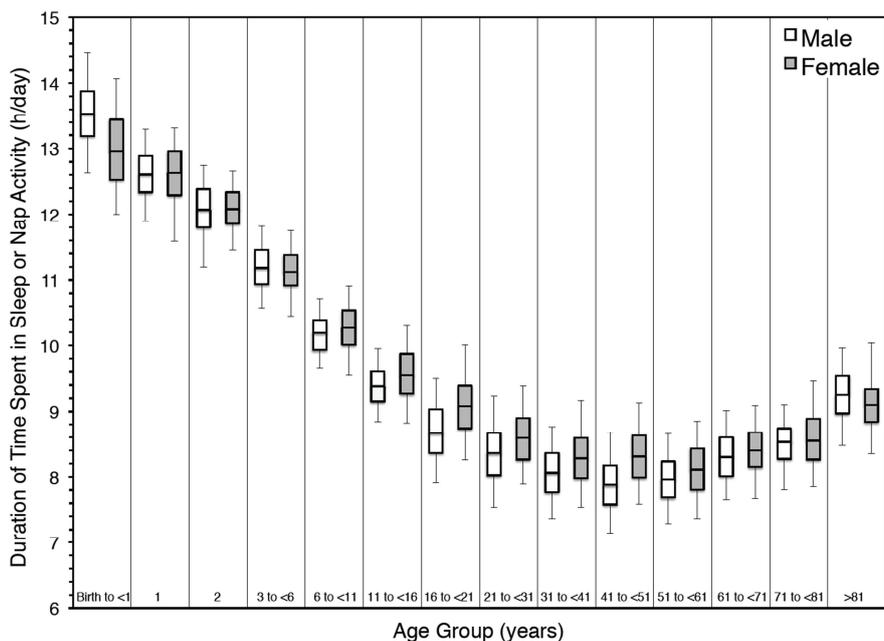


Figure ES1: Duration of time (hours/day) spent in sleep or nap activity (EPA ID = 14500), categorized by age group and gender (Adapted from U.S. Environmental Protection Agency (EPA) Exposure Factors Handbook (EFH) data, Sleep or Nap Activity, EPA Activity ID = 14500, U.S. EPA 2009). Box plots represent interquartile range and whiskers represent the 5<sup>th</sup> and 95<sup>th</sup> percentiles.

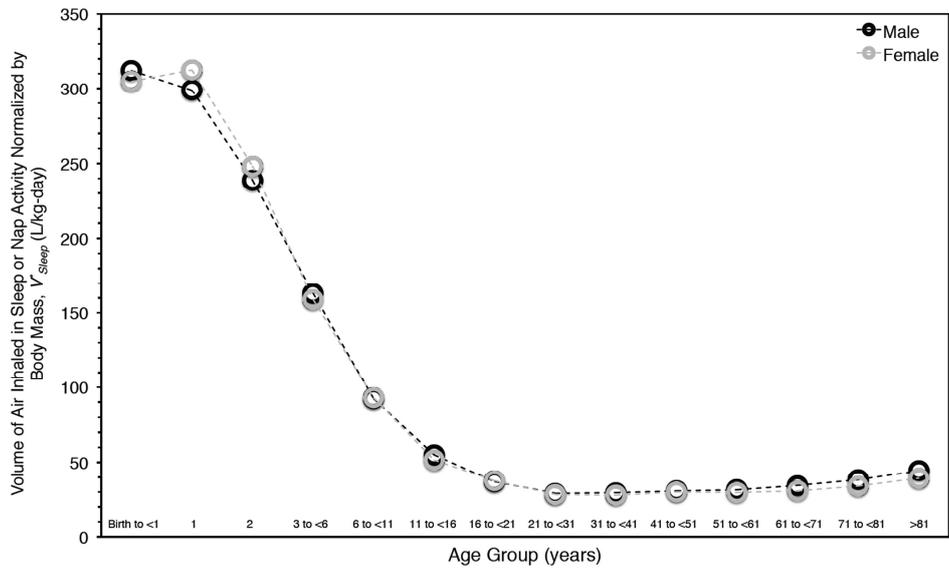


Figure ES2: Volume of air inhaled during sleep or nap activity (EPA ID = 14500) per day, normalized by body mass for each age group and gender (calculated using U.S. EPA EFH data set, U.S. EPA 2009).

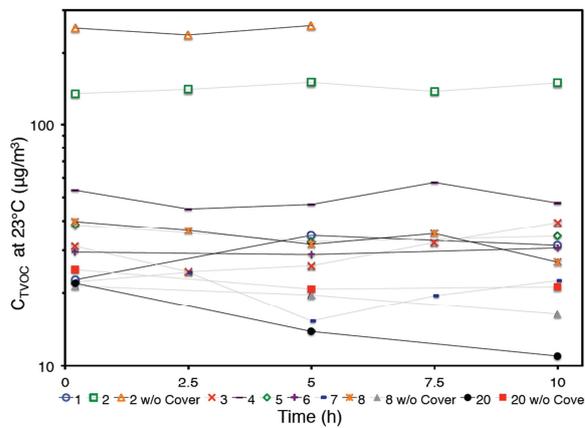


Figure ES3 (a.)

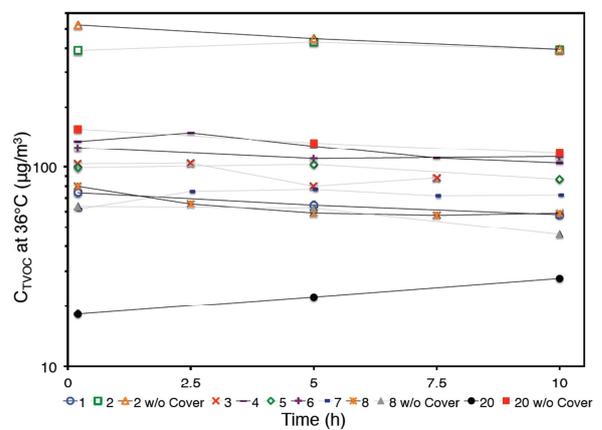


Figure ES3 (b.)

Figure ES3: Small-scale emissions chamber concentration profiles for new samples (used samples not shown) at 23°C and (b.) at 36°C (plotted on logarithmic axis).

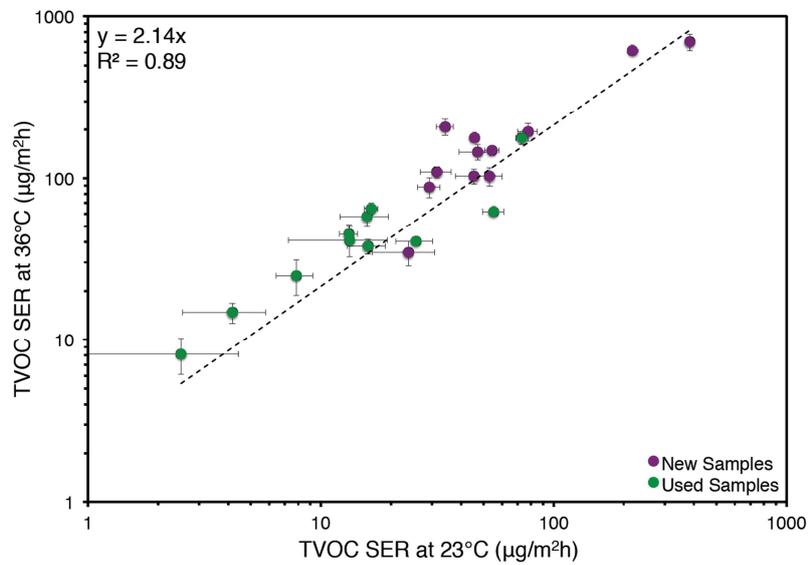


Figure ES4: Relationship between TVOC *SER* (both new and used samples) at 23°C and 36°C (plotted on logarithmic axis). Error bars represent the calculated error in the *SER*.

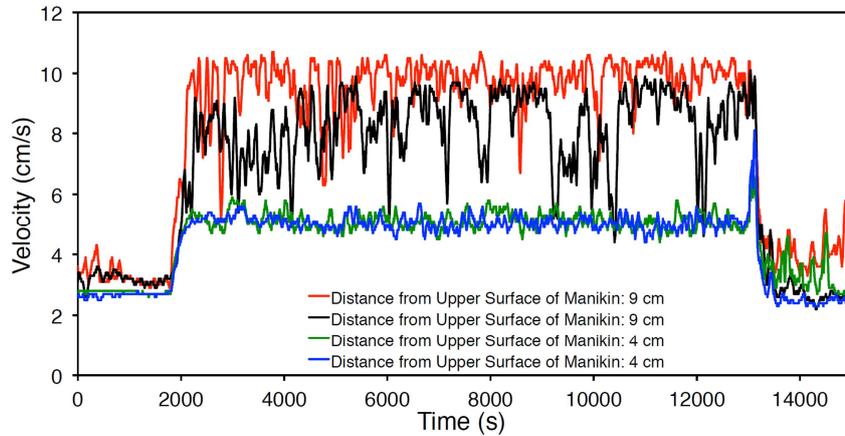


Figure ES5: Infant micro-plume: example of air velocity at two heights above heated infant thermal manikin.

Tables

Table ES1: New and used crib mattress samples.

New Samples					Used Samples				
Sample ID	Country of Manufacture	PUF/Poly	Year Manufactured	Retail Cost	Sample ID	Country of Manufacture	PUF/Poly	Year Manufactured	Usage
1	USA	Poly.	2011	53USD	9	USA	Poly.	2009	1 yr.
2	USA	PUF	2011	43USD	10	USA	PUF	2005	2 yr.
3	USA	PUF	2011	39USD	11	USA	Poly.	2008	2 yr.
4	USA	Poly.	2011	35USD	12	USA	Poly.	2005	2 yr.
5	USA	Poly.	2011	89USD	13	USA	PUF	2001	2 yr.
6	USA	PUF	2011	70USD	14	USA	PUF	2003	3.5 yr.
7	USA	PUF w/ Soy	2011	120USD	15	USA	PUF	1993	10 yr.
8	USA	PUF w/ Soy	2011	148USD	16	USA	PUF	2007	4 yr.
20	USA	PUF	2011	22USD	17	USA	Poly.	2007	3 yr.
					18	USA	PUF	2003	< 1 yr.
					19	Finland	PUF	2000	Unk.

Table ES2: Small-scale and large-scale emission chamber operational and sampling parameters.

Volume, V	Chamber Volumetric Flow Rate, Q (m <sup>3</sup> /h)*	Chamber Air Exchange Rate, λ (h <sup>-1</sup> )	Exposed Sample Area, A (m <sup>2</sup> )	Loading Factor, L (m <sup>2</sup> /m <sup>3</sup> )	Area Specific Airflow Rate, q (m/h)	Air Sampling Flow Rate, Q <sub>s</sub> (mL/min)*	Air Sampling Duration, t <sub>s</sub> (min)
<i>Small-Scale Chamber</i>							
0.035 (L)	0.024	695	0.016	457	1.52	100	25
<i>Large-Scale Chamber</i>							
4.5 (m <sup>3</sup> )	2.25	0.50	1.49	0.33	1.52	100	50

\*Volumetric flow rates were measured with a bubble flow meter (mini-BUCK calibrator, A.P. Buck, Inc.).

Table ES3: TVOC *SERs* for new and used infant crib mattresses in small-scale chamber experiments (mean±calculated error).

New Crib Mattress Samples			Used Crib Mattress Samples		
Sample ID	TVOC <i>SER</i> ( $\mu\text{g}/\text{m}^2\text{h}$ )		Sample ID	TVOC <i>SER</i> ( $\mu\text{g}/\text{m}^2\text{h}$ )	
	23°C	36°C		23°C	36°C
1	45.5±7.8	102.7±10.6	9	73.2±4.4	177.0±14.2
2	218.4±9.5	617.1±27.1	10	15.8±3.7	57.7±8.0
2 w/o Mattress Cover	384.6±14.4	697.2±80.3	11	4.2±1.6	14.7±2.1
3	47.3±8.1	145.2±15.9	12	25.6±4.6	40.4±1.5
4	77.6±7.4	194.4±24.9	13	16.5±1.1	64.9±5.5
5	54.4±3.7	148.7±10.7	14	13.3±6.0	41.0±8.6
6 w/o Mattress Cover	45.7±1.2	178.1±9.9	15	7.8±1.4	24.8±6.1
7	31.5±4.8	109.1±8.2	16	55.2±5.9	62.0±0.3
8	53.1±6.8	102.5±13.4	17	13.2±1.2	44.8±6.4
8 w/o Mattress Cover	29.3±3.3	87.9±12.5	18	2.5±1.9	8.1±2.0
20	23.8±7.1	34.4±5.8	19	15.9±2.9	37.6±4.0
20 w/o Mattress Cover	34.2±2.9	208.2±24.4			

Table ES4: Large-scale emissions chamber concentration results (mean±calculated error), sample 6.

Case	$C_{\text{TVOC}}$ ( $\mu\text{g}/\text{m}^3$ )			Ratio of Interior Foam to Exhaust	Ratio of BZ to Exhaust
	Interior Foam	BZ	Exhaust		
Manikin Heating On With Mattress Cover	168.4±13.7	14.2±2.3	8.0±0.4	21.0	1.8
Manikin Heating Off Without Mattress Cover	137.9±15.6	31.2±7.1	18.4±5.8	7.5	1.7
Manikin Heating On Without Mattress Cover	157.0±8.9	33.2±2.9	14.1±0.7	11.2	2.4

Table ES5: VOC *SERs* for new and used infant crib mattresses in small-scale chamber experiments  
(mean±calculated error).

Compound <sup>1</sup>	MSD Retention Time (min)	CAS	MW <sup>2</sup> (g/mol)	BP <sup>2</sup> (°C)	Sample ID	New/Used	PUF/Poly.	VOC <i>SER</i> at 23°C (µg/m <sup>2</sup> h)	VOC <i>SER</i> at 36°C (µg/m <sup>2</sup> h)
Phenol	17.65	108-95-2	94.1112	182	2	New	PUF	34.2±1.8	151±5.4
					2 w/o Cover	New	PUF	62.2±3.1	257±18.6
					3	New	PUF	1.0±0.5	4.6±0.8
					4	New	Poly.	1.7±0.7	3.4±2.6
					6 w/o Cover	New	PUF	1.3±0.3	6.7±0.8
					7	New	PUF	2.9±0.7	20.2±1.8
					20	New	PUF	< 1	11±2.8
					20 w/o Cover	New	PUF	10.0±0.7	74.9±5.7
					9	Used	Poly.	4.9±0.7	8.5±1.0
19	Used	PUF	< 1	7.1±0.1					
Isooctanol	17.93	26952-21-6	130.2279	180	1	New	Poly.	6.3±0.4	6.6±2.2
					2 w/o Cover	New	PUF	< 1	4.9±2.2
					19	Used	PUF	2.8±1.1	4.3±0.8
Neodecanoic acid	26	26896-20-8	172.2646	262	1	New	Poly.	3.4±0.1	9.2±0.4
					4	New	Poly.	21.9±0.9	33.4±2.9
					5	New	Poly.	18.4±0.5	39.8±1.7
					9	Used	Poly.	4.0±0.2	9.9±0.7
Hexanoic acid, 2-ethyl-	21.97	149-57-5	144.2114	227	2	New	PUF	54.8±2.7	213.4±11.2
					2 w/o Cover	New	PUF	45.5±7.1	118.9±54.6
					6 w/o Cover	New	PUF	< 1	6.3±0.4
					7	New	PUF	2.2±0.5	8.2±1.0
					8	New	PUF	6.3±0.8	32.5±1.1
					20	New	PUF	1.9±0.4	12.0±1.2
					20 w/o Cover	New	PUF	14.2±0.8	72.6±4.9

					9	Used	Poly.	3.8±0.4	7.1±0.5
					13	Used	PUF	1.2±0.1	4.9±0.3
1-Heptanol, 3-methyl-	19.22	1070-32-2	130.2279	186	1	New	Poly.	6.7±1.7	12.2±1.5
					2 w/o Cover	New	PUF	21.1±10.9	7.3±2.1
					5	New	Poly.	8.4±0.5	22±2.8
					8	New	PUF	8.0±2.0	8.1±3.1
D-Limonene	19.31	5989-27-5	136.2340	177	3	New	PUF	10.6±0.9	18±2.8
					7	New	PUF	3.6±0.2	9.2±0.8
Propanenitrile, 2,2'-azobis[2-methyl-	17.91	78-67-1	164.2077	236	6 w/o Cover	New	PUF	10.9±1.9	18.1±2.9
2-Propanol, 1,3-dichloro-	14.26	96-23-1	128.985	175	7	New	PUF	2.3±0.3	11.6±2.0
(S)-3-Ethyl-4-methylpentanol	19.36	--	130.2279	--	1	New	Poly.	6.1±0.8	8.7±1.3
					20	New	PUF	1.5±0.6	2.8±0.6
					19	Used	PUF	1.5±0.1	3.1±0.8
Linalyl acetate	25.59	115-95-7	196.2860	220	3	New	PUF	6.7±0.5	21±3.3
					7	New	PUF	2.4±0.1	6.7±0.5
Cyclopropane, pentyl-	19.28	2511-91-3	112.2126	128	1	New	Poly.	6.4±4.4	21.8±2.5
Linalool	21.23	78-70-6	154.2493	199	3	New	PUF	2.8±0.7	10.1±1.4
					2 w/o Cover	New	PUF	40.6±1.0	33.5±3.9
Benzenemethanol, α,α-dimethyl-	20.65	617-94-7	136.1910	202	6 w/o Cover	New	PUF	1.3±0.1	4±0.2
1-Hexanol, 2-ethyl-	19.1	104-76-7	130.2279	183	6 w/o Cover	New	PUF	2.5±0.4	7.7±1.7
					12	Used	Poly.	2.8±0.8	7.4±0.2
					16	Used	PUF	6.4±0.8	8.3±0.5
2,6-Bis(1,1-dimethylethyl)-4-(1-oxopropyl)phenol	34.31	14035-34-8	262.3871	--	6 w/o Cover	New	PUF	14.4±0.1	61.2±0.9
					8	New	PUF	3.8±0.1	11.9±0.3
					8 w/o Cover	New	PUF	9.0±0.3	32.5±3.6
2-(2-Dimethylaminoethyl)isothiourea	21.26	86114-63-8	147.2418	213	6 w/o Cover	New	PUF	1.3±0.1	14.5±0.5
Dimethylformamide	10.15	68-12-2	73.0938	153	8	New	PUF	18.6±1.8	32.8±2.6
Oxalic acid, bis(2-ethylhexyl) ester	31.64	13675-20-2	314.4601	375	20	New	PUF	2.8±0.1	6.4±0.5

					20 w/o Cover	New	PUF	3.8±0.3	10.5±1.2
1-Decene, 3,4-dimethyl-	31.61	50871-03-9	168.3190	--	2 w/o Cover	New	PUF	31.2±0.9	23.8±1.6
Ethanol, 2-(2-butoxyethoxy)-	23.67	112-34-5	162.2267	231	2 w/o Cover	New	PUF	2.4±0.8	10.1±0.5
Phenol, 2-(1-methylethyl)-	23.93	88-69-7	136.1910	214	2 w/o Cover	New	PUF	5.2±0.6	11.5±1.1
Hexanal	11.04	66-25-1	100.1589	129	9	Used	Poly.	< 1	5.9±1.3
Pentanoic acid	17.82	109-52-4	102.1317	186	9	Used	Poly.	< 1	9.5±2.6
Nonanal	21.15	124-19-6	142.2386	191	6 w/o Cover	New	PUF	< 1	2.3±0.1
					9	Used	Poly.	4.5±0.2	9.6±1.9
					10	Used	PUF	2.0±0.2	3.2±1.2
					11	Used	Poly.	1.2±0.4	2.2±0.2
					12	Used	Poly.	4.3±0.1	7.0±0.3
					17	Used	Poly.	2.0±0.4	3.6±0.5
					18	Used	PUF	1.1±0.4	3.4±0.1
Decanal	23.97	112-31-2	156.2652	208	6 w/o Cover	New	PUF	< 1	1.6±0.1
					9	Used	Poly.	4.7±0.5	10.1±0.9
					19	Used	PUF	1.3±0.2	2.3±0.3
Benzoic acid, 2-ethylhexyl ester	35.94	5444-75-7	250.3334	313	9	Used	Poly.	2.3±0.7	12.2±0.7
Isopropyl Myristate	37.84	110-27-0	270.4507	320	9	Used	Poly.	1.6±0.7	6±0.8
					13	Used	PUF	< 1	5.4±0.3
					15	Used	PUF	< 1	3.3±0.3
					17	Used	Poly.	3.0±0.1	10.6±0.4
					18	Used	PUF	1.5±0.3	5.7±1.0
Palmitic acid	39.69	112-39-0	270.4507	352	9	Used	Poly.	9.7±1.3	42.9±1.4
					10	Used	PUF	2.0±0.2	11.9±1.8
Propanoic acid, 2-methyl-, 1-(1,1-dimethylethyl)-2-methyl-1,3-propanediyl ester	33.59	74381-40-1	286.4118	340	10	Used	PUF	1.9±0.1	10.3±2.4
					11	Used	Poly.	1.8±1.3	4.1±0.1
					13	Used	PUF	1.1±0.2	5.1±1
Homosalate	39.4	118-56-9	262.3441	341	11	Used	Poly.	1.3±1.1	3.3±0.1

					17	Used	Poly.	1.9±0.1	8.7±0.3
Octane, 1,1'-oxybis-	34.8	629-82-3	242.4406	287	13	Used	PUF	7.8±1.6	24.2±0.8
					15	Used	PUF	2.5±0.1	7.1±0.4
Isopropyl Palmitate	41.35	142-91-6	298.5038	341	17	Used	Poly.	8.3±0.4	38.7±2.1

<sup>1</sup>: All compounds identified with at least 75% match with NIST mass spectral library. SVOCs, with BPs > 240°C, are also presented, however, compounds outside of the hexane to hexadecane range were not included in the TVOC analysis.

<sup>2</sup>: From NIST Chemistry WebBook.

## Appendix F

Paper F. Identification of Phthalate and Alternative Plasticizers,  
Flame Retardants, and Unreacted Isocyanates in Infant Crib  
Mattress Covers and Foam

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ABSTRACT

Infants spend most of their time sleeping, where they are in intimate contact with their crib mattresses. In this study, we analyzed the cover and foam layers of 20 new and used crib mattresses for selected chemical additives. 17 of the 20 crib mattress covers contained at least one identifiable plasticizer, with concentrations ranging from 1% to greater than 35% by weight. Sixty percent of the covers contained a total plasticizer content of over 9% by weight. Nine of the 20 covers contained either bis(2-ethylhexyl) phthalate (DEHP) or diisononyl phthalate (DINP). In contrast, phthalate alternatives, including diisononyl 1,2-cyclohexanedicarboxylic acid (DINCH) and bis(2-ethylhexyl) isophthalate (iso-DEHP), were the most frequently identified plasticizers in crib mattresses manufactured after the U.S. Consumer Product Safety Improvement Act (CPSIA) went into effect. Flame retardants, including pentabromodiphenyl ether (pentaBDE) congeners and triphenyl phosphate (TPP), and unreacted isocyanates (NCO) were also identified in crib mattresses with polyurethane foam.

## INTRODUCTION

Infants spend a considerable amount of their time sleeping, typically 12 to 13 h/day, making the sleep microenvironment particularly important in contributing to both their acute and chronic exposures to indoor air pollutants, particularly those originating in crib mattresses on which they sleep. The infant sleep microenvironment can be defined as the space encompassing a crib mattress, bedding, and crib and volume of air above these items that includes both an infant's breathing zone (BZ) and thermal plume. Crib mattresses are typically assembled in multiple layers, with a thick layer of polyurethane or polyester foam (inner-springs are also used) encased within a thin, waterproof, and soft polyvinyl chloride (PVC) cover to protect the mattress foam and to provide an easy-to-clean surface. Different classes of chemical additives can be used in each material layer, which may migrate from the mattress to the air around a sleeping infant and accumulate in settled mattress dust and on bedding fibers.

The soft and flexible nature of crib mattress covers suggests the use of plasticizers (Rossi 2002, Needham et al. 2005); however, the occurrence of such additives in this particular baby product, as well as their concentrations, has not been previously reported in the literature. To meet various flammability standards, brominated and organophosphate flame retardants are often added to the foam padding at percent levels, ranging from 0.1% to 10% by weight (Stapleton et al. 2011). Crib mattresses have also been shown to be an important indoor source of volatile organic compounds (VOCs), with emission rates comparable to flooring materials, wallcoverings, and plastic toys (Boor et al. 2014). In addition, studies have suggested that unreacted isocyanates may

remain in polyurethane foam-based consumer products if excess levels of toluene diisocyanate (TDI) are used in the foam's production (Krone et al. 2003, CA OEHHA 2010). Health effects of the various chemical additives found in crib mattresses are discussed in the Supporting Information (SI).

An infant's exposure via inhalation and dermal pathways to these compounds may be augmented due to several factors. Firstly, infants are in very close contact with their crib mattress and can receive elevated exposures due to the source-proximity effect intrinsic to the sleep microenvironment. Recent laboratory studies have shown that BZ concentrations of gases (e.g., VOCs) and particles (e.g., resuspended mattress dust) released from mattresses are greater than those in the bulk bedroom air by factors typically in the range of 1.5 to 2.5 (Laverge et al. 2013, Boor et al. 2014, 2015, Spilak et al. 2014). Secondly, heat transfer from a sleeping infant (skin temperature ~36°C) (Wheldon 1982, Elabbassi et al. 2001) to their crib mattress can increase the emission rates of VOCs and semi-VOCs (SVOCs) by factors ranging from two to ten, depending on the particular compound (Boor et al. 2014, Liang and Xu 2014a). Lastly, infants can receive much greater doses of a chemical additive released from their mattress compared to adults since they inhale approximately an order of magnitude more air per body mass than adults and have skin surface area to body mass ratios three times greater than adults (Figure FS1, table and figure numbers preceded by an "S" are in the SI. The latter is especially important when considering an infant's exposure to plasticizers in the crib mattress cover via contact transfer between the skin and the cover and air-to-skin uptake (Weschler and Nazaroff 2012, 2013).

In previous papers (Boor et al. 2014, Liang and Xu 2014b), we studied the emission dynamics of VOCs from crib mattresses and examined the influence of temperature on emissions of phthalates from PVC products including crib mattress covers. In the present paper, we focused on levels and trends associated with the usage of various chemicals in crib mattresses over the past decade. The main objective was to investigate the occurrence of phthalate and alternative plasticizers in new and used crib mattresses and to determine their concentrations. Our secondary objective was to identify selected flame retardants, unreacted isocyanates (NCO), and several lower volatility compounds in crib mattress foam. The new data gathered from this study will help to inform policy decisions in the U.S., educate parents and general public, and advance research on infant exposures to pollutants in sleep microenvironments, all of which are urgently needed.

## MATERIALS AND METHODS

### *Sample Collection and Extraction*

A collection of 20 new and used crib mattresses were analyzed in this study. Nine new crib mattresses, made by different manufacturers, were purchased from an online retail store in 2011. 11 used crib mattresses, manufactured between 1993 and 2009, were obtained through donations in Austin, Texas (10 mattresses) and Helsinki, Finland (one mattress). The samples are described in further detail in Boor et al. (2014). For each crib mattress, a sample of both the cover layer and the underlying foam layer were examined.

### *Sample Analysis by PAS-FT-IR and Mass Spectrometry*

Crib mattress cover and foam samples were first screened for possible phthalate content via non-destructive photoacoustic (PAS) Fourier transform-infrared (FT-IR) spectroscopy, as described in the SI. This screening-level analysis suggested that phthalates, or compounds with similar chemical structure, were likely present in many of the mattress covers (Figure FS3 (a.)). Foam samples were also screened for the presence of NCO (Figure FS3 (b.)).

Following PAS-FT-IR screening analysis, all crib mattress cover and foam samples were extracted via ultrasonication in hexane and analyzed by gas chromatography mass spectrometry (GC/MS) following our previously published methods for phthalate and alternative plasticizers with minor modifications (Liang and Xu 2014a, 2014b). All samples were analyzed for eight plasticizers: bis(2-ethylhexyl) phthalate (DEHP), bis(2-ethylhexyl) isophthalate (iso-DEHP), diisononyl phthalate (DINP), diisononyl 1,2-cyclohexanedicarboxylic acid (DINCH), bis(2-ethylhexyl) adipate (DEHA), dimethyl phthalate (DMP), di-n-butyl phthalate (DnBP), and butyl benzyl phthalate (BBP), however, the later three were not detected in any samples. Samples were also analyzed for selected flame retardant additives, including triphenyl phosphate (TPP), tris(1,3-dichloro-2-propyl)phosphate (TDCPP), and pentabromodiphenyl ether (pentaBDE) congeners, along with several additional compounds, such as phenol. Detailed information about the extraction and analytical methods can be found in the SI.

## RESULTS AND DISCUSSION

### *Identification and Quantification of Plasticizers in Crib Mattress Covers and Foam*

Table F1 provides an overview of phthalate and alternative plasticizers detected in the new and used crib mattress covers and foam. 17 of the 20 crib mattress covers contained at least one identifiable plasticizer at concentrations greater than 1 mg/g (0.1% by weight) and the most common plasticizer detected was DINP, found in 40% of all samples. The mean total plasticizer content among new and used covers was 135.5 mg/g and 151.1 mg/g, respectively. 60% of the samples contained more than 90 mg/g of plasticizers and 20% contained over 200 mg/g of plasticizers (samples 5, 7, 13, and 14). The plasticizer content is dependent on several factors, including the amount added during the initial manufacture; the homogeneity of its distribution throughout the product; the extraction and analytical methods employed in this study; and volatilization of the plasticizer from the crib mattress over time, which, for SVOCs, is typically only a small fraction of the initially added mass (Xu et al. 2006, Liang and Xu 2014b). The plasticizer concentrations in crib mattress covers are comparable to those reported for a vinyl pillow protector (140 mg/g of DEHP) (Dodson et al. 2012), a nursing pillow cover (144 mg/g of DINP) (Danish Ministry of the Environment 2008), and a children's sofa (210 mg/g of DEHP) (210 mg/g of DEHP). Plasticizers were not identified in three mattress covers: samples 8, 10, and 19. Sample 19 had a fabric cover (the only non-soft PVC cover tested) and samples 8 and 10 may have contained a plasticizer not included among the eight target compounds.

DINCH, marketed under the name Hexamoll® (BASF, 2009), was the most frequently detected plasticizer in new covers (44.4% of samples) and DINP was the most frequently detected plasticizer in used covers (63.6% of samples). The two phthalate plasticizers, DEHP and DINP, were detected in nine of the 20 covers. Samples 3, 12, 13, 17, and 18 contained both phthalates, with DINP typically occurring at greater concentrations relative to DEHP. A similar observation of the co-occurrence of DEHP and DINP was also made in soft PVC flooring products (Liang and Xu 2014b). The highest detected concentration of DEHP was found in the new crib mattress (sample 20, 125.7 mg/g) with the lowest retail cost (22USD). However, in general, no trends were observed between plasticizer concentrations and crib mattress quality. Two of the new crib mattresses (samples 7 and 8) were marketed as “eco or green mattresses” and manufactured with a fraction of soy-derived foam. Sample 8 did not contain target plasticizers beyond 1 mg/g and sample 7 contained 276.5 mg/g of iso-DEHP. DEHA was primarily detected in crib mattresses made in 2008 and earlier, whereas two of the three samples containing iso-DEHP, marketed under the name “Flexol Plasticizer 380” (NIST 2011, Morose and Becker 2013), were made in 2011. DEHA was always detected in the presence of another plasticizer and may be used as part of a plasticizer mixture in covers.

Plasticizer concentrations in crib mattress polyurethane or polyester foam were typically in the range of 0.1 to 10 mg/g, although several foam samples contained higher concentrations, such as sample 13 with 63.6 mg/g of DINP. These concentrations are comparable to levels of flame retardants in crib mattress foam (Stapleton et al. 2011). Dibutyl phthalate (DBP) was also detected in two foam samples (Table FS1). The identified plasticizers may have originated

during the manufacturing process, as they can be used as dispersion media for catalysts, dyes, fillers, and stabilizers in urethane foam feedstocks (Cooper et al. 1979). There were 13 occurrences of paired-detection of a specific plasticizer in both the cover and foam layers and higher plasticizer concentrations were associated with older mattresses (Figure F1). This suggests that, over time, the foam behaved as a sorptive reservoir for gas-phase plasticizers released from the overlying cover layer or from products found elsewhere in a residence, although greater concentrations of plasticizers may have been used during the initial manufacture of the foam in older mattresses.

#### *Trends Associated with the 2008 U.S. CPSIA*

Although concentrations of a specific plasticizer in crib mattress covers did not show strong associations with the age of the mattress, trends in the occurrence of plasticizers were observed in regard to the U.S. Consumer Product Safety Improvement Act of 2008 (CPSIA), as shown in Figure F1. The CPSIA limits the concentrations of DEHP, DBP, and BBP in children's toys or child-care articles to below 0.1% by weight (1 mg/g) (U.S. CPSC 2008). Furthermore, a recent report by the Chronic Hazard Advisory Panel (CHAP) to the U.S. Consumer Product Safety Commission (CPSC) recommended that DINP be permanently included in this list (CHAP 2014). However, the inclusion of a crib mattress in the definition of a child-care article is not clear (NRDC 2009, U.S. CPSC 2009, Winnebeck 2011).

Since the CPSIA went into effect, there has been more frequent use of phthalate alternatives, including DINCH and iso-DEHP, which were identified in seven of the 10 cover samples manufactured from 2009 to 2011. The use of DINCH as a phthalate replacement in crib mattresses follows the trend observed

in other soft PVC products (ECHA 2013), and isophthalates (iso-DEHP) are being used in the same manner (Morose and Becker 2013). However, DEHP concentrations in two of the crib mattress covers manufactured after the CPSIA went into effect (sample 3: 83.1 mg/g, sample 20: 125.7 mg/g) exceeded the 1 mg/g limit of the CPSIA for child-care articles. Considering all 20 crib mattresses, nine would not have met the current CPSIA limits or CHAP recommendations as they contained either DEHP or DINP at concentrations greater than 1 mg/g.

Table F1: Description of Crib Mattress Samples Analyzed in This Study and a Summary of the Plasticizers Detected in These Samples at Concentrations  $\geq 1.0$  mg/g Cover or  $\geq 0.1$  mg/g Foam

Crib Mattress Samples <sup>1</sup>			Plasticizers (mg/g)											
New Samples (n = 9)			DEHP		iso-DEHP		DINP		DINCH		DEHA		$\Sigma$ Plasticizers <sup>4</sup>	
ID <sup>2</sup>	Year Manufactured	Retail Cost	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam
1	2011	53USD							93.7				93.7	
2	2011	43USD							68.8	0.1	4.8		73.6	0.1
3	2011	39USD	83.1	0.8				28.5					111.6	0.8
4	2011	35USD		1.4					34.9				34.9	1.4
5	2011	89USD			352.5								352.5	
6	2011	70USD							15.4				15.4	
7	2011	120USD			276.5								276.5	
8	2011	148USD		0.5										0.5
20	2011	22USD	125.7	4.2									125.7	4.2
Detection Frequency			22.2%	44.4%	22.2%	n.d.	11.1%	n.d.	44.4%	11.1%	11.1%	n.d.	88.9 %	55.5 %
Mean Concentration (mg/g)			104.4	1.7	314.5	n.d.	28.5	n.d.	53.2	0.1	4.8	n.d.	135.5	1.4
Used Samples (n = 11)														
ID	Year Manufactured	Usage												
9	2009	1 yr.							103.1	0.1		0.2	103.1	0.3
10	2005	2 yr.												
11	2008	2 yr.					149.8				44.1		193.9	
12	2005	2 yr.	43.2	0.9			40.9				53.9		138.0	0.9
13	2001	2 yr.	3.3				207.1	63.6			40.7		251.1	63.6
14	2003	3.5 yr.					306.2	0.6			14.4		320.6	0.6
15	1993	10 yr.		7.9			42.0	16.5					42.0	24.4
16	2007	4 yr.		8.0	60.0	14.4						0.1	60.0	22.5
17	2007	3 yr.	4.4	0.4			93.0						97.4	0.4
18	2003	< 1 yr.	32.9	5.3			118.5	8.5			2.5	0.4	153.9	14.2
19	2000	Unk.		10.6										10.6
Detection Frequency			36.4%	54.5%	9.1%	9.1%	63.6%	36.4%	9.1%	9.1%	45.5%	27.3%	81.8 %	81.8 %
Mean Concentration (mg/g)			21.0	5.5	60.0	14.4	136.8	22.3	103.1	0.1	31.1	0.2	151.1	15.3

<sup>1</sup>: All samples, excluding 19, manufactured in the U.S. Sample 19 was manufactured and used in Finland and had a fabric cover. Samples 2, 3, 6-8, 20, 10, 13-16, 18, and 19 were manufactured with polyurethane foam; samples 7 and 8 were manufactured with a fraction of soy-derived foam; and samples 1, 4, 5, 9, 11, 12, and 17 were manufactured with polyester foam (as listed by manufacturer).

<sup>2</sup>: Sample ID consistent with crib mattress samples analyzed in Boor et al. (2014).

<sup>3</sup>: Blank cell: plasticizer not detected (n.d.) at concentrations  $\geq 1.0$  mg/g Cover or  $\geq 0.1$  mg/g Foam.

Nomenclature: DEHP: Bis(2-ethylhexyl) phthalate; iso-DEHP: Bis(2-ethylhexyl) isophthalate; DINP: Diisononyl phthalate; DINCH: Diisononyl 1,2-cyclohexanedicarboxylic acid; DEHA: Bis(2-ethylhexyl) adipate

<sup>4</sup>: Total plasticizer content in each crib mattress cover or foam layer

not detected (n.d.)  $\geq 0.1$  to 1 mg/g (for foam only)  $> 1$  to 10 mg/g  $> 10$  to 100 mg/g  $> 100$  mg/g

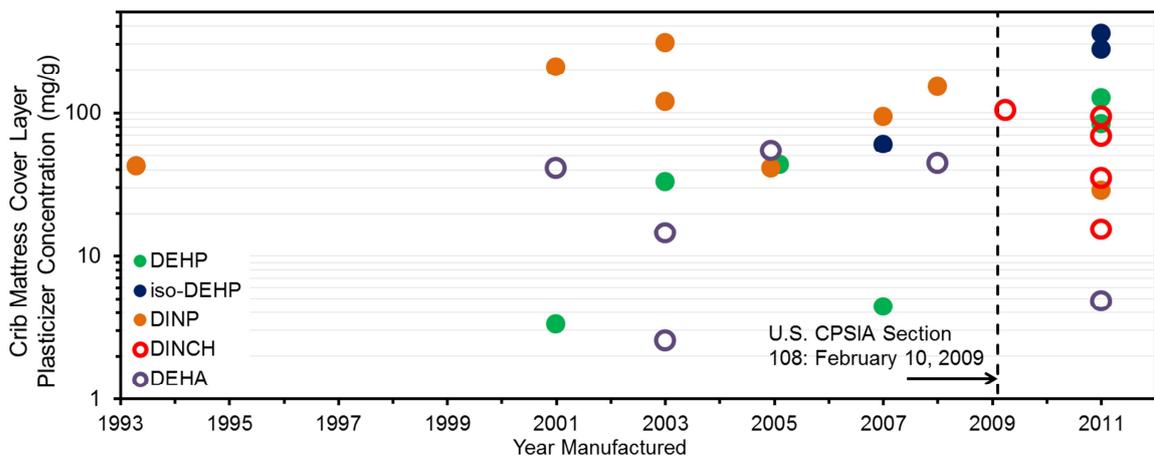


Figure F1 (a.)

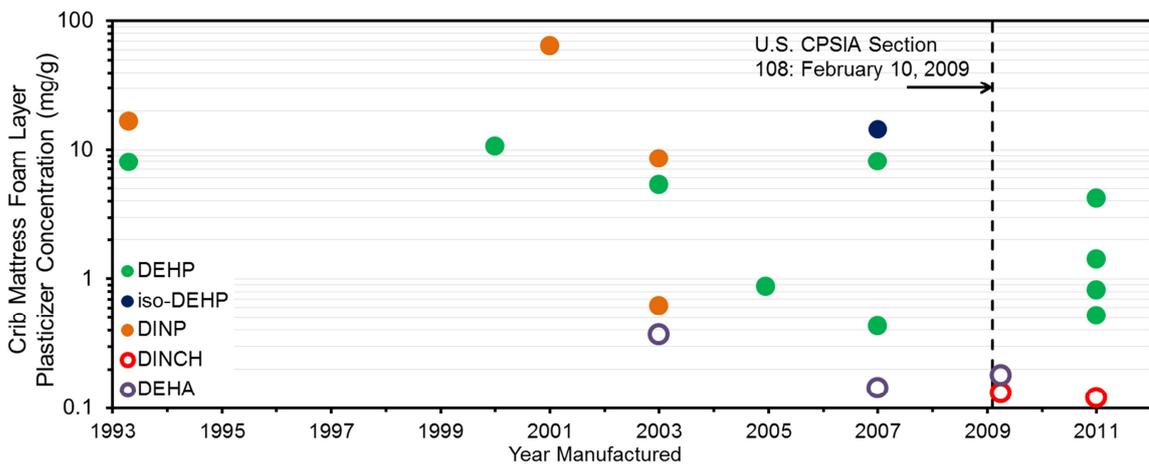


Figure F1 (b.)

Figure F1: Plasticizer concentrations in cover (a.) and foam (b.) layers versus the year crib mattress samples were manufactured. Date on which the U.S. Consumer Product Safety Improvement Act (CPSIA) Section 108 was enacted in the U.S (February 10, 2009) denoted by dashed line.

#### *Additional Compounds Detected in Crib Mattresses*

Polybrominated diphenyl ether (PBDE) congeners associated with the pentaBDE mixture were identified in three crib mattress foam samples: sample

10 (2.2 mg/g), sample 15 (5.5 mg/g), and sample 18 (14.7 mg/g) (Table FS1). Samples 15 and 18 were manufactured before the 2004 pentaBDE phase-out (Tullo 2003), whereas sample 10 was manufactured in 2005. TPP was identified in the three samples containing pentaBDE congeners, as well as several additional foam samples and two crib mattress covers, where it may have been used as a plasticizer (UK Environment Agency 2009, Schossler et al. 2011). The co-occurrence of the two flame retardants is likely the result of both being used in combination during the manufacture of the foam (Stapleton et al. 2011) or adsorption of gas-phase TPP or pentaBDEs in a residence over time. In total, eight of the 20 crib mattress foam layers contained pentaBDE congeners, TPP, or TDCPP (in sample 7 with partial soy-derived foam), all of which were manufactured with polyurethane foam. All crib mattresses with a label indicating that it met one of the following three flammability standards: Technical Bulletin (TB) 116, TB 117, or FF 4-72, contained either pentaBDEs or TPP, or both. NCO, as identified through PAS-FT-IR analysis, was detected in all 13 polyurethane foam samples (Table FS1), likely at greater levels in new mattresses relative to used (Figure FS4). Further discussion on NCO can be found in the SI. Additionally, phenol, 1-decene, 1-nonanol, and palmitic acid were detected in crib mattresses, the latter two of which were likely adsorbed over time by the mattress, as discussed in Boor et al. (2014).

#### *Implications for SVOC Transport and Infant Exposure in the Sleep Microenvironment*

The findings from this study, along with Stapleton et al. (2011) and Boor et al. (2014), collectively demonstrate that infant crib mattresses can contain SVOCs, including plasticizers and flame retardants, VOCs, and unreacted isocyanates. Given the slow and nearly constant emission rate of plasticizers from soft PVC

materials (Xu et al. 2006, Xu et al. 2012), it is likely that crib mattresses are a constant plasticizer source in the infant sleep microenvironment, which is important, considering the long lifetime of a crib mattress (~10 years), mattress re-use in families with multiple children, and purchasing of used mattresses through second-hand venues. This is in contrast to VOC emissions that tend to decay over time.

It is expected that the chemical additives identified in this study will volatilize from the crib mattress, however, experimental data on the emissions of SVOCs from mattresses, pillows, and bedding are limited (Järnström et al. 2009, Rauert et al. 2014). Kemmlein et al. (2003) reported an emission rate of 0.012  $\mu\text{g}/\text{m}^2\text{h}$  for tris (2-chloro-isopropyl)phosphate (TCPP) from a mattress and our parallel study evaluated the emissions of DINCH (sample 9) and DEHA (sample 11) from the cover layers (Liang and Xu 2014b). Crib mattress covers act as a diffusive barrier to the transport of VOCs originating in the underlying foam (Boor et al. 2014), and may do the same for flame retardants as they migrate from the foam to ambient air. This may partially explain the paired detection of TPP in the cover and foam layers of sample 15.

The volatilized gas-phase plasticizers and flame retardants will tend to partition to and accumulate in surface dust particles (e.g., Hwang et al. 2008, Weschler and Nazaroff 2008). As infants move and crawl in their cribs, they will likely resuspend settled mattress dust (Shalat et al. 2011, Spilak et al. 2014, Boor et al. 2015) and thus be exposed to particle-phase plasticizers and flame retardants. Oral exposure may also occur via hand-to-mouth transfer of mattress dust during sleep periods (Stapleton et al. 2008). The gas-phase SVOCs may also adsorb to bed sheets, where they may subsequently desorb during sleep periods,

as the temperature of the bedding fibers increases (Weschler and Nazaroff 2012). In addition, dermal uptake of plasticizers and flame retardants in infants may occur as they sleep via direct contact transfer between the skin and crib mattress cover or surface-sorbed SVOCs on bed sheets, and air-to-skin transport. The latter is linked to concentrations of gas-phase pollutants released from the mattress, which are much greater near the mattress surface compared to the bulk bedroom air (Laverge et al. 2013, Boor et al. 2014).

#### AUTHOR CONTRIBUTIONS

B.E.B., H.J., A.N., and Y.X. defined the overall scope of the study and developed the experimental plan. B.E.B., Y.L., and N.E.C. conducted the experiments and analyzed the data. B.E.B. wrote the paper. Y.X. and A.N. provided guidance in the execution of the experiments, advised in the interpretation of the results, and provided detailed comments on draft manuscripts.

#### SUPPORTING INFORMATION

##### *Health Effects of Target Compounds Identified in this Study*

Phthalates have endocrine disrupting properties (Bergman et al. 2013) and have been shown to retard male development (Adibi et al. 2003, Swan et al. 2005), alter semen quality (Hauser et al. 2006), cause irreversible changes to the male reproductive tract (Matsumoto et al. 2008), and increase the risk of allergic disease and asthma (Bornehag et al. 2004, Kolarik et al. 2008, Larsson et al. 2010). Presently, toxicological data on phthalate replacements, including DEHA, DINCH, and iso-DEHP, are limited. DEHA may induce mild developmental

toxicity (Dalgaard et al. 2003) and has been shown to decrease anogenital distance and retention of nipples in rats (Jarfelt et al. 2005). Studies suggest that DINCH may not have endocrine disrupting properties or reproductive toxicity, although it has been associated with thyroid hyperplasia and renal toxicity (EFSA 2006). Polybrominated diphenyl ethers (PBDEs), including PentaBDE congeners, can interfere with thyroid hormone homeostasis, and consequently, delay neurological development in humans and rodents (Rudel and Perovich 2009). PBDE exposure has been found to impair a child's motor, cognitive, and behavioral performance and decrease fecundability (Roze et al. 2009, Herbstman et al. 2010). Adverse health effects associated with TPP include neurotoxicity (Andresen et al. 2004), cardiotoxicity (McGee et al. 2013), immunotoxicity (Saboori et al. 1991), and contact dermatitis (Camarasa and Serra-Baldrich 1992). TDCPP has been found to be associated with increases in kidney, liver, and testicular tumors in rats (Faust and August 2011). Isocyanates may play an important role in the development of childhood asthma (Krone and Klinger 2005 and references therein) and exposure has been associated with allergic sensitization, asthma, rhinitis, and contact dermatitis (Bello et al. 2007, Verschoor and Verschoor 2014).

### *PAS-FT-IR Analysis*

All twenty crib mattresses were screened for phthalates in the cover layer (prior to extractions and GC/MS analysis) and unreacted isocyanates in the foam layer via Photoacoustic (PAS) Fourier transform-infrared (FT-IR) spectroscopy (FTS 6000, Bio-Rad Laboratories, Inc. equipped with a MTEC 300 photoacoustic detector, as described in Wahlström et al. 2012). PAS-FT-IR is a non-destructive

method that requires no pre-treatment of the samples, and thus, is suitable for screening-level analysis (McClelland et al. 1992). Cover and foam samples 10 mm in diameter were taken from near the middle of a horizontal surface of the crib mattress. Additionally, a liquid standard solution of DEHP (CAS 117-81-7) and hard polyvinyl chloride (PVC) were analyzed to produce instrument-specific spectra for qualitative comparisons. Samples were analyzed at a modulation frequency of 2.5 kHz with helium as the detector gas atmosphere.

Sample spectra were generated for all cover and foam samples and example spectra are shown in Figure FS3. For the detection of phthalates in crib mattress covers, all sample spectra were compared with the reference DEHP spectrum. The sample spectra were manually inspected for peaks representing the structure of DEHP, including C-H stretching (2959, 2933, 2876  $\text{cm}^{-1}$ ), C=O stretching of O=C-O (1726  $\text{cm}^{-1}$ ), C-O stretching of O=C-O (1285  $\text{cm}^{-1}$ ), C-O stretching of O-CH<sub>2</sub> (1124, 1077  $\text{cm}^{-1}$ ), and out of plane bending for aromatics (745  $\text{cm}^{-1}$ ). Based on this analysis, phthalates, or plasticizers with a similar structure to that of DEHP, were determined to be likely present in nine of the eleven used cover samples (samples 10-18) and four of the nine new cover samples (samples 3, 5, 7, 20). Based on the GC/MS analysis, fifteen of the twenty predictions were correct, in that a phthalate or alternative plasticizer was detected at a concentration > 10 mg/g. The utility of PAS-FT-IR for screening of plasticizers requires more research, including generation of instrument-specific reference spectra for the numerous plasticizers in use in baby products.

Unreacted isocyanates (NCO) were identified in all crib mattress polyurethane foam samples via a peak at wavenumber of 2274  $\text{cm}^{-1}$  (Figure FS3 (b.) and Table FS1). Numerous FT-IR studies of polyurethane foam have shown that NCO groups (functional group  $-\text{N}=\text{C}=\text{O}$ ) are characterized by strong asymmetric stretching vibration around 2270-2280  $\text{cm}^{-1}$ , a region where there is minimal interference from other functional groups (e.g., Bhattacharjee and Engineer 1996, Luda et al. 2004, Hatchett et al. 2005, Delucchi 2008).

#### *Chemicals, Solvent Extraction, and GC-MS Analysis*

Standard solutions were used for chemical calibration and identification. Bis(2-ethylhexyl) phthalate (DEHP) was purchased from Absolute Standards, Inc. (Hamden, CT); bis(2-ethylhexyl) isophthalate (iso-DEHP) was purchased from SPEX CertiPrep (Metuchen, NJ); and diisononyl phthalate (DINP, specifically DINP-2, CAS 28553-12-0) and bis(2-ethylhexyl) adipate (DEHA) were purchased from Accustandard, Inc. (New Haven, CT). Pure (anhydrous, >99%) diisononyl 1,2-cyclohexanedicarboxylic acid (DINCH) was purchased from BOC Sciences (Shirley, NY), from which calibration standards were made. Bis(2-ethylhexyl) phthalate-3,4,5,6-d<sub>4</sub> (D<sub>4</sub>-DEHP) was used as an internal standard and purchased from Accustandard, Inc. (New Haven, CT). Hexane was used as the solvent in the extractions and purchased from Sigma-Aldrich Co. LLC (anhydrous, >99%) (St. Louis, MO).

Crib mattress cover samples, typically less than 1 mm in thickness, were taken from near the middle of a horizontal surface of the crib mattress, cut into ~ 1 cm by 1 cm pieces, and accurately weighed (~ 1 g). The "upper" horizontal

surface, upon which an infant sleeps, was selected if known from the manufacturer label or packaging. Please note that the crib mattress cover described here is a structural part of the mattress, and not a secondary mattress protector, e.g., used a barrier for house dust mite allergens. Crib mattress foam samples were taken from the foam layer located immediately below the cover sample and accurately weighed (~ 1 g). The samples were ultrasonically extracted (3800 Ultrasonic Cleaner, Branson Ultrasonic Corp., Danbury, CT) for 30 min with hexane, repeated four times. The sonicator was filled with clean water and several ice bags were used to maintain a relatively low temperature and avoid chemical volatilization. The extracts were reduced to a volume of 5 to 10 mL via rotatory evaporation (RV 10, IKA®-Werke GmbH & Co. KG, Staufen, Germany) and filtered with preassembled filtration devices (Autovial™ syringeless filters, PTFE filter, Whatman®, GE Healthcare Bio-Sciences, Pittsburgh, PA). The extract was further reduced to approximately 1 mL via high purity N<sub>2</sub> evaporation (Reacti-Vap™ Evaporators, Thermo Scientific Inc., Waltham, MA), transferred to a 2 mL amber vial, and stored in a refrigerator at 4°C prior to GC-MS analysis.

1 µL of each sample extract was injected into a sorbent tube packed with Tenax TA, desorbed with a thermal desorber (TD, TurboMatrix ATD, PerkinElmer, Inc., Waltham, MA), and analyzed with a gas chromatograph (GC, 7890A, Agilent Technologies, Santa Clara, CA) mass spectrometer (MS, 5975, Agilent Technologies, Santa Clara, CA) (henceforth referred to as the primary GC-MS). 5 µL of D<sub>4</sub>-DEHP (2 µg/mL) in methanol was injected in each sorbent tube as an internal standard. Additional details can be found in Liang and Xu

(2014). All sorbent tubes were analyzed in two successive desorptions to ensure complete desorption of both the sorbent tube and the TD-GC-MS system. The second desorption showed concentrations below the detection limit in all cases. A calibration standard was regularly run prior to each GC injection, and the variance was always below 5% for all injections.

All analyses were performed in full scan mode and the extracted ions used for identification and quantification of the five target plasticizers can be found in Liang and Xu (2014b). DINP and DINCH are mixtures of numerous isomers (e.g., Schossler et al. 2011, Nagorka et al. 2011) and elute across many peaks for a period of approximately 1 min (Figure FS2), with the response derived from the total area between the first and last detected isomer. All samples were also screened for three additional phthalate plasticizers: dimethyl phthalate (DMP), di-n-butyl phthalate (DnBP), and butyl benzyl phthalate (BBP), however, none were detected at levels > 0.1 mg/g in any of the cover or foam samples. Dibutyl phthalate (DBP) was detected in two foam samples and identified through comparison with a mass spectral database (NIST08).

For the plasticizer concentrations listed in Table F1, a threshold value of 1 mg/g was selected for covers because plasticizers are typically added to soft PVC products at percent levels. Only two crib mattress cover samples contained identifiable plasticizers below the 1 mg/g threshold value. For foam samples, a threshold value of 0.1 mg/g was used because the occurrence of plasticizers is most likely associated with surface adsorption of gas-phase plasticizers

originating in the overlying cover or in a residence, although plasticizers may be used in the foam itself.

Three samples containing PentaBDE congeners, as identified with the primary GC-MS through comparison with a mass spectral database (NIST08), were also analyzed with GC- $\mu$ ECD (micro-electron capture detector) (7890A, Agilent Technologies, Inc., Santa Clara, CA) with a 30 m column (0.25 mm ID, film thickness: 0.1  $\mu$ m) (DB-5ht, Agilent Technologies, Santa Clara, CA) following our previously published methods (Xu et al. 2014). Additional flame retardants, such 2,2-bis(chloromethyl)propane-1,3-diyl-tetrakis(2-chloroethyl)bis(phosphate) (V6), were not screened for, as they require additional analytical techniques to identify (Stapleton et al. 2011, 2012).

#### *Discussion on Unreacted Isocyanates*

An excess of toluene diisocyanate (TDI, the predominate diisocyanate used in polyurethane foam production, Vangronsveld et al. 2013a) is typically added during the manufacture of polyurethane foam, above which is necessary for it to react with hydroxyl groups of polyols and water (Hugo et al. 2000). As such, unreacted isocyanate may be present in the final polyurethane foam product (Hugo et al. 2000, Krone et al. 2003, CA OEHHA 2010). Krone et al. (2003) identified NCO in polyurethane foam-containing consumer products, including a mattress and sofa padding. The foam in crib mattresses is somewhat protected from excess moisture released from the infant body, which can react with the isocyanates (Bello et al. 2006). The detection of NCO in crib mattresses by PAS-FT-IR analysis necessitates that future research measure concentrations

of NCO in crib mattress foam, determine to the extent to which NCO can migrate to air, and if NCO is present in mattress dust.

Figures

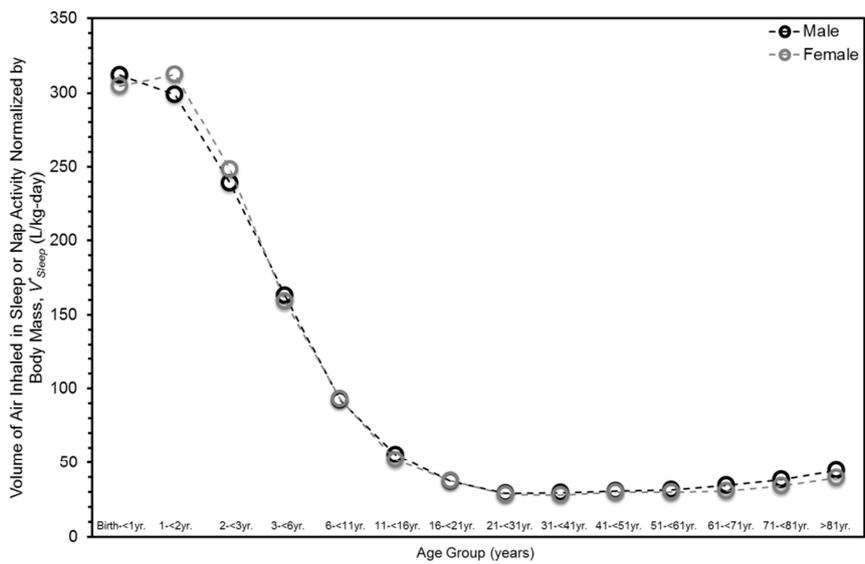


Figure FS1 (a.)

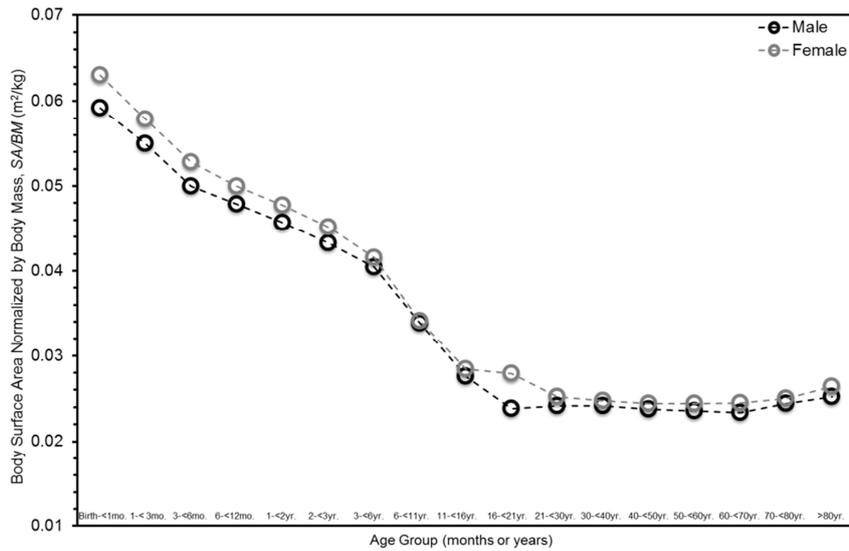


Figure FS1 (b.)

Figure FS1: Infant inhalation and dermal exposure dose considerations in the sleep microenvironment: (a.) volume of air inhaled during sleep or nap activity (EPA ID = 14500) per day, normalized by body mass ( $V_{Sleep}^*$ , L/kg-day) for each age group and gender (calculated using U.S. EPA EFH data set, U.S. EPA 2011), and (b.) body surface area normalized by body mass (SA/BM, m<sup>2</sup>/kg) for each age group and gender (calculated using U.S. EPA EFH data set, U.S. EPA 2011).

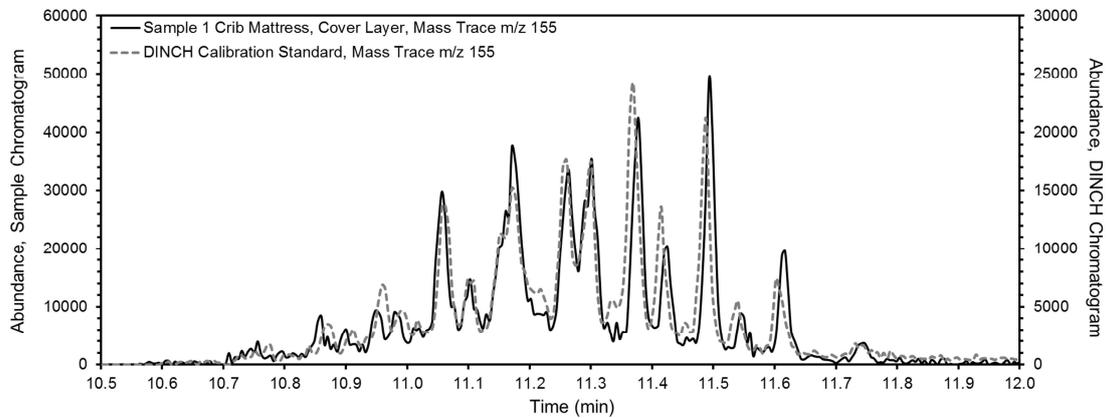


Figure FS2 (a.)

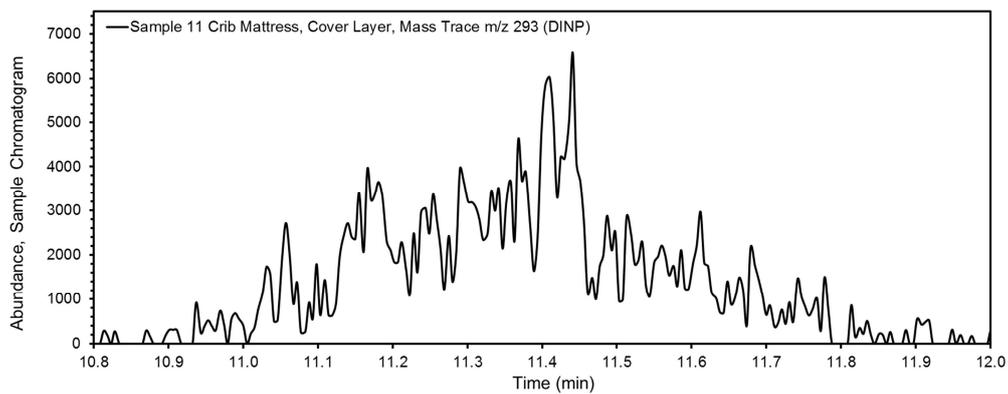


Figure FS2 (b.)

Figure FS2: Example GC-MS chromatograms for: (a.) sample 1 crib mattress, cover layer containing DINCH ( $m/z$  155) and DINCH calibration standard ( $m/z$  155), and (b.) sample 11 crib mattress, cover layer containing DINP ( $m/z$  293).

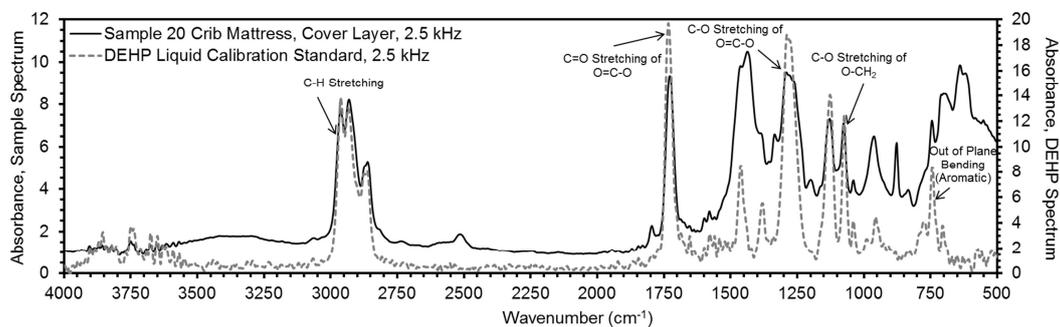


Figure FS3 (a.)

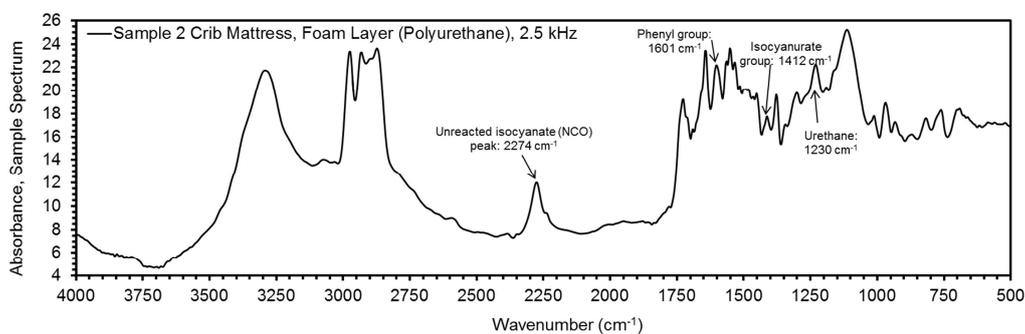


Figure FS3 (b.)

Figure FS3: Example PAS-FT-IR spectra (all at a 2.5 kHz modulation frequency): (a.) sample 20 crib mattress, cover layer containing DEHP and DEHP calibration standard and (b.) sample 2 crib mattress, foam layer with unreacted isocyanate group.

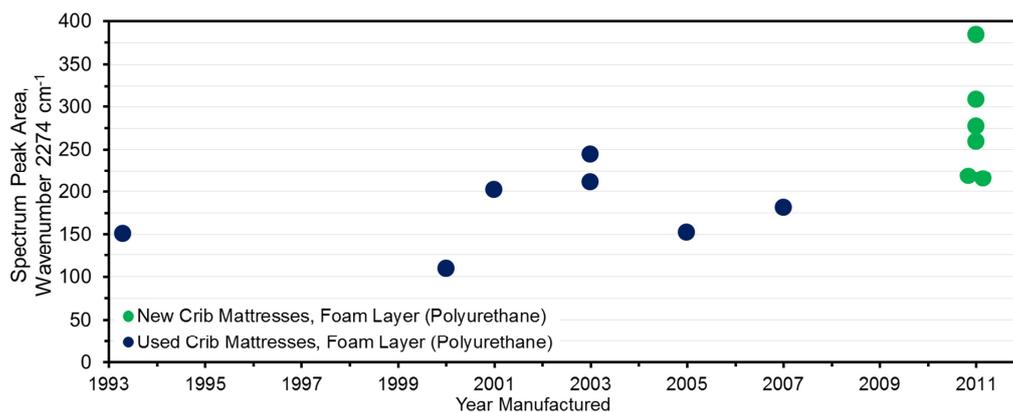


Figure FS4: Unreacted isocyanate peak area (wavenumber 2274 cm<sup>-1</sup>) versus the year crib mattress sample was manufactured (for polyurethane foam samples only).

Table FS1: Additional Chemical Additives Detected in the Crib Mattress Cover and Foam Layers.

Crib Mattress Samples <sup>1</sup>			Additional Chemical Additives <sup>4</sup>																	
New Samples (n = 9)			DBP		TPP <sup>5</sup>		TDCPP		PentaBDE (mg/g)		Phenol <sup>5</sup>		1-Decene <sup>5</sup>		1-Nonanol <sup>5</sup>		Palmitic Acid <sup>5</sup>		NCO <sup>6</sup>	
ID <sup>2</sup>	Year Manuf.	Retail Cost	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam
1	2011	53USD																		
2	2011	43USD				X							X							X
3	2011	39USD				X							X							X
4	2011	35USD												X						
5	2011	89USD								X										
6	2011	70USD													X					X
7	2011	120USD						X		X							X			X
8	2011	148USD																		X
20	2011	22USD		X		X												X		X
Detection Frequency			n.d.	11.1%	n.d.	33.3%	n.d.	11.1%	n.d.	n.d.	22.2%	n.d.	n.d.	22.2%	11.1%	11.1%	11.1%	11.1%	n.d.	66.7%
Used Samples (n = 11)																				
ID	Year Manuf.	Usage	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam	Cover	Foam
9	2009	1 yr.									X									
10	2005	2 yr.				X			X, 2.2		X			X						X
11	2008	2 yr.			X								X							
12	2005	2 yr.								X										
13	2001	2 yr.				X							X							X
14	2003	3.5 yr.									X			X			X			X
15	1993	10 yr.		X	X	X			X, 5.5											X
16	2007	4 yr.								X			X			X				X
17	2007	3 yr.											X							

18	2003	< 1 yr.				X				X, 14.7	X			X	X					X
19	2000	Unk.										X								X
Detection Frequency			n.d.	9.1%	18.2%	36.4%	n.d.	n.d.	n.d.	27.3%	27.3%	18.2%	45.5%	18.2%	27.3%	n.d.	9.1%	9.1%	n.d.	63.6%

1: All samples, excluding 19, manufactured in the U.S. Sample 19 manufactured and used in Finland. Samples 2, 3, 6-8, 20, 10, 13-16, 18, and 19 were manufactured with polyurethane foam; samples 7 and 8 were manufactured with a fraction of soy-derived foam; and samples 1, 4, 5, 9, 11, 12, and 17 were manufactured with polyester foam (as listed by manufacturer).

2: Sample ID consistent with crib mattress samples analyzed in Boor et al. (2014).

3: Blank cell: chemical additive not detected (n.d.).

4: All compounds (aside from NCO) identified with at least 85% match with NIST mass spectral library; quantification not performed (aside from PentaBDE).

5: Detected via analysis with a secondary GC-MS, as described in the SI section.

6: Detected via PAS-FT-IR, as described in the SI section.

Nomenclature: DBP: Dibutyl phthalate; TPP: Triphenyl phosphate; TDCPP: Tris(1,3-dichloro-2-propyl)phosphate; PentaBDE: Pentabromodiphenyl ether congener mixture (congeners detected include: BDE-47, BDE-85, BDE-99, BDE-100, BDE-153, and BDE-154; quantified via GC- $\mu$ ECD analysis with calibration standards, a detailed discussion on the methods can be found Xu et al. 2014); NCO: unreacted isocyanate

not detected (n.d.) X: detected

## References

- Aaronson, S. T., Rashed, S., Biber, M. P., and Hobson, J. A. (1982). Brain state and body position. *Arch. Gen. Psychiat.* 39:330-335.
- Abraham, K., Mielke, H., Huisinga, W., and Gundert-Remy, U. (2005). Elevated internal exposure of children in simulated acute inhalation of volatile organic compounds: effects of concentration and duration. *Arch. Toxicol.* 79:63-73.
- Acevedo-Bolton, V., Cheng, K.-C., Jiang, R.-T., Ott, W. R., Klepeis, N. E., and Hildemann, L. M. (2012). Measurement of the proximity effect for indoor air pollutant sources in two homes. *J. Environ. Monit.* 14:94-104.
- Adgate, J. L., Weisel, C., Wang, Y., Rhoads, G. G., and Liroy, P. J. (1995). Lead in house dust: relationships between exposure metrics. *Environ. Res.* 70:134-147.
- Adhiwidjaja, I., Matsusaka, S., Tanaka, H., and Masuda, H. (2000). Simultaneous phenomenon of particle deposition and reentrainment: effects of surface roughness on deposition layer of striped pattern. *Aerosol Sci. Technol.* 33:323-333.
- Adibi, J. J., Perera, F. P., Jedrychowski, W., Camann, D. E., Barr, D., Jacek, R., and Whyatt, R. M. (2003). Prenatal exposures to phthalates among women in New York City and Krakow, Poland. *Environ. Health Persp.* 111:1719-1722.
- Afshari, A., Lundfren, B., and Ekberg, L. E. (2003). Comparison of three small chamber test methods for the measurement of VOC emission rates from paint. *Indoor Air.* 13:156-165.
- Adolph, K. E., Vereijken, B., and Denny, M. A. (1998). Learning to crawl. *Child Dev.* 69:1299-1312.
- Ahmadi, G., and Guo, S. (2007). Bumpy particle adhesion and removal in turbulent flows including electrostatic and capillary forces. *J. Adhesion.* 83:289-311.
- Ali, N., Dirtu, A. C., Van den Eede, N., Goosey, E., Harrad, S., Neels, H., 't Mannetje, A., Coakley, J., Douwes, J., and Covaci, A. (2012). Occurrence of alternative flame retardants in indoor dust from New Zealand: indoor sources and human exposure assessment. *Chemosphere.* 88:1276-1282.

- Alshittawi, M., and Awbi, H. (2011). The effect of human movement on the resuspension of airborne particles, in Proceedings of the 12th International Conference on Indoor Air and Climate, Austin, TX, paper ID: 728.
- An, J. Y., Kim, S., and Kim, H. J. (2011). Formaldehyde and TVOC emission behavior of laminate flooring by structure of laminate flooring and heating condition. *Journal of Hazardous Materials*. 187(1-3):44-51.
- Andresen, J. A., Grundmann, A., and Bester, K. (2004). Organophosphorus flame retardants and plasticisers in surface waters. *Sci. Total Environ.* 332:155–166.
- Anderson, R.C., and Anderson, J.H. (1999). Respiratory toxicity of mattress emissions in mice. *Arch. Environ. Health*. 54:202-209.
- Anderson, R.C., and Anderson, J.H. (2000). Respiratory toxicity in mice exposed to mattress covers. *Arch. Environ. Health*. 55:38-43.
- Auger, M. (1994). Efficiency of residential duct cleaning, Project No. 93-2-001, in Canada Mortgage and Housing Corporation, Ottawa, Canada.
- Azumi, K., Shirikawa, S., and Takahashi, S. (1977). A proposal of a new classification for body movements during sleep: variations of body movements through a week. *Sleep Res.* 6:44.
- Bagnold, R. A. (1941). *The Physics of Blown Sand and Desert Dunes*. Methuen & Co. Ltd., New York.
- Barnig, C., Reboux, G., Roussel, S., Casset, A., Sohy, C., Dalphin, J.-C., and Blay, F.de. (2012). Indoor dust and air concentrations of endotoxin in urban and rural environments. *Lett. Appl. Microbiol.* 56:161-167.
- BASF. Physical-chemical properties of diisononyl cyclohexane-1,2-dicarboxylate (DINCH).[http://www.plasticizers.basf.com/portal/load/fid247503/Hex\\_DINCH\\_e\\_09\\_13.pdf](http://www.plasticizers.basf.com/portal/load/fid247503/Hex_DINCH_e_09_13.pdf) (Accessed 12/20/2014).
- Begum, M., Aya, A., Cheong, C., Neumeister-Kemp, H., White, K., and Kemp, P. (2012). Prevalence of fungi in used pillows, in Proceedings of Healthy Buildings 2012, Brisbane, Australia, paper ID: 5H.4.
- Bejan, A. (2004). *Convection heat transfer*. John Wiley & Sons, Inc., New Jersey.
- Bekö, G., Lund, T., Nors, F., Toftum, J., and Clausen, G. (2010). Ventilation rates in the bedrooms of 500 Danish children. *Build. Environ.* 45:2289-2295.

- Bello, D., Herrick, C. A., Smith, T. J., Woskie, S. R., Streicher, R. P., Cullen, M. R., Liu, Y., and Redlich, C. A. (2007). Skin exposure to isocyanates: reason for concern. *Environ. Health Persp.* 115:328-335.
- Bennett, D. H., McKone, T. E., Evans, J. S., Nazaroff, W. W., Margni, M. D., Jolliet, O., and Smith, K. R. (2002). Defining intake fraction. *Environ. Sci. Technol.* 36:A206–A211.
- Bergman, Å., Heindel, J. J., Jobling, S., Kidd, K. A., Zoeller, R. T., Jobling, S. K., eds. *State of the Science of Endocrine Disrupting Chemicals 2012*. Geneva: United Nations Environment Programme and World Health Organization. Available: [http://unep.org/pdf/9789241505031\\_eng.pdf](http://unep.org/pdf/9789241505031_eng.pdf) (Accessed 02/07/2015).
- Bhattacharjee, D. and Engineer, R. (1996). An improved technique for the determination of isocyanurate and isocyanate conversion by photoacoustic FTIR. *J. Cell. Plast.* 32:260-273.
- Bloor, W. A., and Dinsdale, A. (1962). Protective clothing as a factor in the dust hazard of potters. *Brit. J. Industr. Med.* 19:229-235.
- Blum, A., Gold, M. D., Ames, B. N., Kenyon, C., Jones, F. R., Hett, E. A., Dougherty, R. C., Horning E. C., Dzidic, I., Carroll, D. I., Stillwell, R. N., and Thenol, J. P. (1978). Children absorb Tris-BP flame retardant from sleepwear: urine contains the mutagenic metabolite, 2,3-dibromopropanol. *Science.* 201:1020-1023.
- Bodalal, A., Zhang, J. S., Plett, E., and and Zhu, J. (2001). Correlations between the internal diffusion and equilibrium partition coefficients of volatile organic compounds (VOCs) in building materials and the VOC properties. *ASHRAE Trans.* 107(1):789-800.
- Bohne, J. E., and Cohen, B. S. (1985). Aerosol resuspension from fabric: implications for personal monitoring in the beryllium industry. *Am. Ind. Hyg. Assoc. J.* 46:73-79.
- Boor, B. E., Siegel, J. A., and Novoselac, A. (2011). Development of an experimental methodology to determine monolayer and multilayer particle resuspension from indoor surfaces. *ASHRAE Trans.* 117(1):434-441.
- Boor, B. E., Siegel, J. A., and Novoselac, A. (2013a). Monolayer and multilayer particle deposits on hard surfaces: literature review and implications for particle resuspension in the indoor environment. *Aerosol Sci. Technol.* 47(8):831-847.

- Boor, B. E., Siegel, J. A., and Novoselac, A. (2013b). Wind tunnel study on aerodynamic particle resuspension from monolayer and multilayer deposits on linoleum flooring and galvanized sheet metal. *Aerosol Sci. Technol.* 47(8):848-857.
- Boor, B. E., Järnström, H., Novoselac, A., and Xu, Y. (2014). Infant exposure to emissions of volatile organic compounds from crib mattresses. *Environ. Sci. Technol.* 48(6):3541-3549.
- Boor, B. E., Spilak, M. P., Corsi, R. L., and Novoselac, A. (2015a). Characterizing particle resuspension from mattresses: chamber study. *Indoor Air.* 25(4):441-456.
- Boor, B. E., Liang, Y., Crain, N. E., Järnström, H., Novoselac, A., and Xu, Y. (2015b). Identification of phthalate and alternative plasticizers, flame retardants, and unreacted isocyanates in infant crib mattress covers and foam. *Environ. Sci. Technol. Letters.* 2(4):89-94
- Bornehag, C.-G., Sundell, J., Weschler, C. J., Sigsgaard, T., Lundgren, B., Hasselgren, M., and Hagerhed-Engman, L. (2004). The association between asthma and allergic symptoms in children and phthalates in house dust: a nested case-control study. *Environ. Health Persp.* 112:1393-1397.
- Bowes, S. M., Mason, E. G, Corn, M. (1993). Confined space ventilation: tracer gas analysis of mixing characteristics. *AIHA J.* 54:639-646.
- Braaten, D. A. (1994). Wind tunnel experiments of large particle reentrainment-deposition and development of large particle scaling parameters. *Aerosol Sci. Technol.* 21:157-169.
- Braaten, D. A., Paw U, K. T., and Shaw, R. H. (1990). Particle resuspension in a turbulent boundary layer – observed and modeled. *J. Aerosol Sci.* 21:613-628.
- Brouwer, D. (2010). Exposure to manufactured nanoparticles in different workplaces. *Toxicology.* 269:120-127.
- Byrne, M. A., Goddard, A. J. H., Lange, C., and Roed, J. (1995). Stable tracer aerosol deposition measurements in a test chamber. *J. Aerosol Sci.* 26:645-653.
- Cadogan, D. F., and Howick, C. J. (1996). Plasticizers. *Kirk-Othmer Encyclopedia of Chemical Technology.* John Wiley & Sons, Inc., New York.
- California Office of Environmental Health Hazard Assessment (CA OEHHA). (2010). Proposed revised reference exposure levels for toluene diisocyanate and methylene diphenyl diisocyanate. CA OEHHA, Sacramento, CA.

- California Office of Environmental Health Hazard Assessment (CA OEHHA) Chronic toxicity summary N,N-Dimethylformamide. [http://oehha.ca.gov/air/chronic\\_rels/pdf/68122.pdf](http://oehha.ca.gov/air/chronic_rels/pdf/68122.pdf) (12/10/2013),
- Callesen, M., Bekö, G., Weschler, C. J., Sigsgaard, T., Jensen, T. K., Clausen, G., Toftum, J., Norberg, L. A., and Høst, A. (In Press). Associations between selected allergens, phthalates, nicotine, PAHs and bedroom ventilation and clinically confirmed asthma, rhinoconjunctivitis and atopic dermatitis in preschool children. *Indoor Air*. doi: 10.1111/ina.12060.
- Camann, D. E., Zuniga, M., Yau, A., Heilbrun, L., Walker, T., and Miller, C. (2011). Persistence of pesticides in residential indoor air and chair seat foam, in Proceedings of the Indoor Air 2011 International Conference, Austin, Texas, paper ID A57.
- Camarasa, J. G. and Serra-Baldrich, E. (1992). Allergic contact dermatitis from triphenyl phosphate. *Contact Derm.* 26:264-265.
- Cardoso, V. M., Solano, A. G. R., Prado, M. A. F., and Nunan, E. D. A. (2006). Investigation of fatty acid esters to replace isopropyl myristate in the sterility test for ophthalmic ointments. *Journal of Pharmaceutical and Biomedical Analysis.* 42(5):630-634.
- Casas, L., Tischer, C., Wouters, I. M., Valkonen, M., Gehring, U., Doekes, G., Torrent, M., Pekkanen, J., Garcia-Esteban, R., Hyvärinen, A., Heinrich, J., and Sunyer, J. (2013). Endotoxin, extracellular polysaccharides, and  $\beta(1-3)$ -glucan concentrations in dust and their determinants in four European birth cohorts: results from the HITEA project. *Indoor Air.* 23:208-216.
- Chang, C., and Gershwin, M. E. (2004). Indoor air quality and human health. *Clin. Rev. Allerg. Immu.* 27:219-239.
- Chen, C.-M., Mielck, A., Fahlbusch, B., Bischof, W., Herbarth, O., Borte, M., Wichmann, H.-E., and Heinrich, J. (2007). Social factors, allergen, endotoxin, and dust mass in mattress. *Indoor Air.* 17:384-393. *Clin. Exp. Dermatol.* 35:238-244.
- Chen, C.-M., Sausenthaler, S., Bischof, W., Herbarth, O., Borte, M., Behrendt, H., Krämer, U., Williams, H. C., Wichmann, H.-E., and Heinrich, J. (2009). Perinatal exposure to endotoxin and the development of eczema during the first 6 years of life. *Clin. Exp. Dermatol.* 35:238-244.

- Chiou, S. F., and Tsai, C. J. (2001). Measurement of emission factor of road dust in a wind tunnel. *Powder Technol.* 118:10-15.
- Choi, J.-I., and Edwards, J. R. (2012). Large-eddy simulation of human-induced contaminant transport in room compartments. *Indoor Air.* 22:77-87.
- Chronic Hazard Advisory Panel (CHAP). (2014). Report to the U.S. Consumer Product Safety Commission on phthalates and phthalate alternatives. Bethesda, Maryland.
- Chuang, J. C., Callahan, P. J., Menton, R. G., Gordon, S. M., Lewis, R. G., and Wilson, N. K. (1995). Monitoring methods for polycyclic aromatic hydrocarbons and their distribution in house dust and track-in soil. *Environ. Sci. Technol.* 29:494-500.
- Clausen, P. A., Liu, Z., Kofoed-Sorensen, V., Little, J., and Wolkoff, P. (2012). Influence of temperature on the emission of Di-(2-ethylhexyl)phthalate (DEHP) from PVC flooring in the emission cell FLEC. *Environ. Sci. Technol.* 46(2):909-915.
- Clayton C. A., Wallace L. A., Ozkaynak H., and Spengler J. D. (1993). Particle total exposure assessment methodology (PTEAM) study: distributions of aerosol and elemental concentrations in personal, indoor, and outdoor air samples in a southern California community. *J. Expos. Anal. Environ. Epidemiol.* 6:227-250.
- Cleaver, J. S., and Looi, L. (2007). AFM study of adhesion between polystyrene particles: the influence of relative humidity and applied load. *Powder Technol.* 174:34-37.
- Cleaver, J. W., and Yates, B. (1973). Mechanism of detachment of colloidal particles from a flat substrate in a turbulent flow. *J. Colloid Interface Sci.* 44:464-474.
- Cleet, C. (2015). Letter to the editor. *Bull. Environ. Contam. Toxicol.* 74:1-3.
- Cohen, B. S., and Positano, R. (1986). Resuspension of dust from work clothing as a source of inhalation exposure. *Am. Ind. Hyg. Assoc. J.* 47:255-258.
- Cohen Hubal, E.A., Nishioka, M. G., Ivancic, W. A., Morara, M., and Egeghy, P. P. (2008). Comparing surface residue transfer efficiencies to hands using polar and nonpolar fluorescent tracers. *Environ. Sci. Technol.* 42(3):934-939
- Colloff, M. J. (2009). *Dust mites*. Springer, Dordrecht, Netherlands.
- Cooke, W. B., and Foter, M. J. (1958). Fungi in used bedding materials. *Appl. Microbiol.* 6:169-173.

- Cooper, J. A., Morgan, A. W., and Wang, D. S. T. (1979). Butyl benzyl phthalate in rigid polyurethane foam. *J. Cell. Plast.* 15:249-257.
- Conant, N. F., Wagner, H. C., and Rackemann, F. M. (1935). Fungi found in pillows, mattresses and furniture. *Allergy.* 7:234-237.
- Corn, M., and Stein, F. (1965). Re-entrainment of particles from a plane surface. *Amer. Ind. Hyg. Assoc. J.* 26:325-336.
- Custovic, A., Simpson, B., Simpson, A., Hallam, C., Craven, M., and Woodcock, A. (1999). Relationship between mite, cat, and dog allergens in reservoir dust and ambient air. *Allergy.* 54:612-616.
- Dalgaard, M., Hass, U., Vinggaard, A. M., Jarfelt, K., Lam, H.R., Sørensen, I. K., Sommer, H. M., and Ladefoged, O. (2003). Di(2-ethylhexyl) adipate (DEHA) induced developmental toxicity but not antiandrogenic effects in pre- and postnatally exposed Wistar rats. *Reprod. Toxicol.* 7(2):163-70.
- Danish Ministry of the Environment, Environmental Protection Agency. (2008). Survey, emission and health assessment of chemical substances in baby products. In *Survey of Chemical Substances in Consumer Products*, No. 90, Denmark.
- da Silva, D. R., Binotti, R. S., da Silva, C. M., de Oliveira, C. H., Condino-Neto, A., and de Capitani, E. M. (2005). Mites in dust samples from mattress surfaces from single beds or cribs in the south Brazilian city of Londrina. *Pediatr. Allergy Immunol.* 16:132-136.
- Delucchi, M. (2008). Knowing the time. *European Coatings J.* 3:1-9.
- Deng, Q., Yang, X., and Zhang, J. S. (2009). study on a new correlation between diffusion coefficient and temperature in porous building materials. *Atmos. Environ.* 43:2080-2083.
- Deng, Q., Yang, X., and Zhang, J. S. (2011). Key factor analysis of VOC sorption and its impact on indoor concentrations: the role of ventilation. *Build. Environ.* 47:182-187.
- Diez, U., Kroebner, T., Rehwagen, M., Richter, M., Wetzig, H., Schulz, R., Borte, M., Metzner, G., Krumbiegel, P., and Herbarth, O. (2000). Effects of indoor painting and smoking on airway symptoms in atopy risk children in the first year of life results of the LARS-study. Leipzig Allergy High-Risk Children Study. *Int. J. Hyg. Environ. Health.* 203:23-28.

- Dishaw, L. V., Powers, C. M., Ryde, I. T., Roberts, S. C., Seidler, F. J., Slotkin, T. A., and Stapleton, H. M. (2011). Is the PentaBDE replacement, tris (1,3-dichloro-2-propyl) phosphate (TDCPP), a developmental neurotoxicant? *Studies in PC12 cells. Toxicol. Appl. Pharm.* 256:281-290.
- Dodson, R. E., Nishioka, M., Standley, L. J., Perovich, L. J., Green Brody, J., and Rudel, R. A. (2012). Endocrine disruptors and asthma-associated chemicals in consumer products. *Environ. Health Persp.* 120(7):935-943.
- Douwes, J., Zuidhof, A., Doekes, G., van der Zee, S. C., Wouters, I., Boezen, M. H., and Brunekreef, B. (2000). (1-3)- $\beta$ -D-Glucan and endotoxin in house dust and peak flow variability in children. *Am. J. Respir. Crit. Care Med.* 162:1348-1354.
- Douwes, J., Thorne, P., Pearce, N., and Heederik, D. (2003). Bioaerosol health effects and exposure assessment: progress and prospects. *Ann. Occup. Hyg.* 47:187-200.
- Doyen, V., Johansson, A.-B., Hanssens, L., Dehennin, N., Dinh, D. H. P., Casimir, G., and Michel, O. (2011). Relationship between the presence of newborn and the house dust endotoxin. *Sci. Total Environ.* 409:5313-5317.
- Drescher, A. C., Lobascio, C., Gadgil, A. J., and Nazaroff, W. W. (1995). Mixing of a point-source indoor pollutant by forced convection. *Indoor Air.* 5:204-214.
- Earnest, C. M., and Corsi, R. L. (2013). Inhalation exposure to cleaning products: application of a two-zone model. *J. Occup. Environ. Hyg.* 10:328-335.
- Eberlein, B., Gulyas, A. F., Schultz, K., Lecheler, J., Flögel, S., Wolfmeyer, C., Thiessen, K., Jakob, T., Schuster, T., Hollweck, R., Ring, J., and Behrendt, H. (2009). Domestic allergens and endotoxin in three hospitals offering in-patient rehabilitation for allergic diseases in the alpine mountain climate of Bavaria – The AURA study. *Int. J. Hyg. Environ. Health.* 212:21-26.
- Edwards, R. D., Yurkow, E. J., and Liroy, P. J. (1998). Seasonal deposition of housedusts onto household surfaces. *Sci. Total Environ.* 224:69-80.
- Edwards, R. D., Jurvelin, J., Saarela, K., and Jantunen, M. (2001). VOC concentrations measured in personal samples and residential indoor, outdoor and workplace microenvironments in EXPOLIS-Helsinki, Finland. *Atmos. Environ.* 35:4531-4543.
- Eisner, A. D., Rosati, J., and Wiener, R. (2010). Experimental and theoretical investigation of particle-laden airflow under a prosthetic mechanical foot in motion. *Build. Environ.* 45:878-886.

- Elabbassi, E. B., Bach, V., Makki, M., Delanaud, S., Telliez, F., Leke, A., and Libert, J. P. (2001). Assessment of dry heat exchanges in newborns: influence of body position and clothing in SIDS. *J. Appl. Physiol.* 91(1):51-56.
- El Sharif, N., Douwes, J., Hoet, P. H. M., Doekes, G., and Nemert, B. (2004). Concentrations of domestic mite and pet allergens and endotoxin in Palestine. *Allergy.* 59:623-631.
- Elabbassi, E. B., Bach, V., Makki, M., Delanaud, S., Telliez, F., Leke, A., and Libert, J. P. (2001). Assessment of dry heat exchanges in newborns: influence of body position and clothing in SIDS. *J. Appl. Physiol.* 91:51-56.
- Ege, M. J., Mayer, M., Normand, A.-C., Genuneit, J., Cookson, W. O. C. M., Braun-Fahrländer, Heederik, D., Piarroux, R., and von Mutius, E. (2011). Exposure to environmental microorganisms and childhood asthma. *N. Engl. J. Med.* 364:701-709.
- Ege, M. J., Mayer, M., Schwaiger, K., Mattes, J., Pershagen, G., van Hage, M., Scheynius, A., Bauer, J., and von Mutius, E. (2012). Environmental bacteria and childhood asthma. *Allergy.* 67:1565-1571.
- Environment Canada Health Canada (EHC) Screening Assessment for the Challenge: Hexanoic acid, 2-ethyl-. <http://www.ec.gc.ca/ese-ees/default.asp?lang=En&n=1D5253CB-1> (12/10/2013).
- European Chemicals Agency (ECHA). Evaluation of New Scientific Evidence Concerning DINP and DIDP in Relation to Entry 52 of Annex XVII to Reach Regulation (EC) No 1907/2006 (Final Review Report). <http://echa.europa.eu/documents/10162/31b4067e-de40-4044-93e8-9c9ff1960715> (Accessed 12/28/2014).
- European Food Safety Authority (EFSA). (2006). Opinion of the Scientific Panel on food additives, flavourings, processing aids and materials in contact with food (AFC) on a request related to a 12th list of substances for food contact materials. *EFSA J.* 395-401:1-21.
- Ewaldsson, B., Fogelmark, B., Feinstein, R., Ewaldsson, L., and Rylander, R. (2002). Microbial cell wall product contamination of bedding may induce pulmonary inflammation in rats. *Lab. Anim.* 36:282-290.

- Fahlbusch, B., Hornung, D., Heinrich, J., Dahse, H.-M., and Jäger, L. (2000). Quantification of group 5 grass pollen allergens in house dust. *Clin. Exp. Allergy*. 30:1645-1652.
- Fairchild, C. I., and Tillery, M. J. (1982). Wind tunnel measurements of the resuspension of ideal particles. *Atmos. Environ.* 16:229-238.
- Faust, J. B., and August, L. M. (2011). Evidence on the carcinogenicity of tris(1,3-dichloro-2-propyl)phosphate. Office of Environmental Health Hazard Assessment, California Environmental Protection Agency, California.
- Ferro, A. R., Kopperud, R. J., and Hildemann, L. M. (2004a). Source strengths for indoor human activities that resuspend particulate matter. *Environ. Sci. Technol.* 38:1759-1764.
- Ferro, A. R., Kopperud, R. J., and Hildemann, L. M. (2004b). Elevated personal exposure to particulate matter from human activities in a residence. *J. Expo. Anal. Env. Epid.* 14:S34-S40.
- Finnish Society of Indoor Air Quality and Climate (FiSIAQ). (2001). Classification of indoor climate 2000, target values, design guidance, and product requirements. FiSIAQ, Helsinki, Finland.
- Fletcher, R., Briggs, N., Ferguson, E., and Gillen, G. (2008). Measurements of air jet removal efficiencies of spherical particles from cloth and planar surfaces. *Aerosol Sci. Technol.* 42:1052-1061.
- Foarde, K. K., and Menetrez, M. Y. (2002). Evaluating the potential efficacy of three antifungal sealants of duct liner and galvanized steel as used in HVAC systems. *J. Ind. Micro. Biotechnol.* 29:38-43.
- Fortmann, R., Gentry, G., Foarde, K. K., VanOsdell, D. W., and Kulp, R. N. (1997). Results of a pilot field study to evaluate the effectiveness of cleaning residential heating and air conditioning systems and the impact on indoor air quality and system performance. U.S. Environmental Protection Agency (EPA), Washington, D.C.
- Franke, D. L., Cole, E. C., Leese, K. E., Foarde, K. K., and Berry, M. A. (1997). Cleaning for improved indoor air quality: an initial assessment of effectiveness. *Indoor Air*. 7:41-54.
- Franklin, P. J. (2007). Indoor air quality and respiratory health of children. *Paediatr. Respir. Rev.* 8:281-286.

- Fransson, J. I., Ruud, S. H., and Rosell, L. (1995). *Rena Ventilationskanaler*, SP-Rapport 1995:38, Sveriges Provnings- och Forskningsinstitut (SP), Energiteknik/Kemisk Analys, Borås, Sweden (in Swedish).
- Frasch, H. F., Dotson, G. S., Bunge, A. L., Chen, C. P., Cherrie, J. W., Kasting, G. B., Kissel, J. C., Sahmel, J., Semple, S., and Wilkinson, S. (2014). Analysis of finite dose dermal absorption data: Implications for dermal exposure assessment. *J. Expo. Anal. Environ. Epid.* 24(1):65-73.
- Freudenthal, R., and Henrich, R. T. (2000). Chronic toxicity and carcinogenic potential of tris (1,3-dichloro- 2-propyl) phosphate in Sprague-Dawley rats. *Int. J. Toxicol.* 19:119-125.
- Friess, H., and Yadigaroglu, G. (2001). A generic model for the resuspension of multilayer aerosol deposits by turbulent flow. *Nucl. Sci. Engr.* 138:161-176.
- Friess, H., and Yadigaroglu, G. (2002). Modelling of the resuspension of particle clusters from multilayer aerosol deposits with variable porosity. *J. Aerosol Sci.* 33:883-906.
- Fromentin, A. (1989). Particle resuspension from a multilayer deposit by turbulent flow. Doctoral Dissertation, Swiss Federal Institute of Technology, Zurich, Switzerland.
- Fulton, J. E. and Pay, S. R. (1984). comedogenicity of current therapeutic products, cosmetics, and ingredients in the rabbit ear. *Journal of the American Academy of Dermatology.* 10(1):96-105.
- Furtaw, E. J., Pandian, M. D., Nelson, D. R., and Behar, J. V. (1996). Modeling indoor air concentrations near emission sources in imperfectly mixed rooms. *J. Air Waste Manag. Assoc.* 46:861-868.
- Gac, J., Sosnowski, T., and Gradon, L. (2008). Turbulent flow energy for aerosolization of powder particles. *J. Aerosol Sci.* 39:113-126.
- Gallagher, M., Wysocki, J., Leyden, J. J., Spielman, A. I., Sun, X., and Preti, G. (2008). Analyses of volatile organic compounds from human skin. *British Journal of Dermatology* 159(4):780-791.
- Garrett, M. H., Hooper, B. M., and Hooper, M. A. (1998). Indoor environmental factors associated with house-dust-mite allergen (Der p 1) levels in south-eastern Australian houses. *Allergy.* 53:1060-1065.

- Gehring, U., Bischof, W., Borte, M., Herbath, O., Wichmann, H.-E., and Heinrich, J. (2004). Levels and predictors of endotoxin in mattress dust samples from East and West German homes. *Indoor Air*. 14:284-292.
- Giganti, F., Ficca, G., Gori, S., and Salzarulo, P. (2008). Body movements during night sleep and their relationship with sleep stages are further modified in very old subjects. *Brain Res. Bull.* 75:66-69.
- Giovannangelo, M. E., Gehring, U., Nordling, E., Oldenwening, M., van Rijswijk, K., de Wind, S., Hoek, G., Heinrich, J., Bellander, T., and Brunekreef, B. (2007). Levels and determinants of  $\beta$  (1-3)-glucans and fungal extracellular polysaccharides in house dust of (pre-)school children in three European countries. *Environ. Int.* 33:9-16.
- Goldasteh, I., Ahmadi, G., and Ferro, A. (2012a). A model for removal of compact, rough, irregularly shaped particles from surfaces in turbulent flows. *J. Adhesion*. 88:766-786.
- Goldasteh, I., Ahmadi, G., and Ferro, A. (2012b). Wind tunnel study and numerical simulation of dust particle resuspension from indoor surfaces in turbulent flows. *J. Adhes. Sci. Technol.* 27:1563-1578.
- Gomes, C., Freihaut, J., and Bahnfleth, W. P. (2007). Resuspension of allergen-containing particles under mechanical and aerodynamic disturbances from human walking. *Atmos. Environ.* 41:5257-5270.
- Gore, R. B., Hadi, E. A., Craven, M., Smillie, F. I., O'Meara, T. J., Tovey, E., Woodcock, A., and Custovic, A. (2002). Personal exposure to house dust mite allergen in bed: nasal air sampling and reservoir allergen levels. *Clin. Exp. Allergy*. 32:856-859.
- Gore, R. B., Boyle, R. J., Hanna, H., Custovic, A., Gore, C., Svensson, P., and Warner, J. O. (2010). Personal allergen exposures are increased by changes in sleep position and improved by temperature-controlled laminar airflow. *Thorax*. 65:A87-A88.
- Gotoh, K., Matsuda, S., Yoshida, M., Oshitani, J., and Ogura, I. (2011). Experimental investigation of particle resuspension from a powder layer induced by an ascending flat object. *Kagaku Kogaku Ronbun*. 37:317-322 (in Japanese).
- Górny, R. L., Dutkiewicz, J., and Krysińska-Traczyk, E. (1999). Size distribution of bacterial and fungal bioaerosols in indoor air. *Ann. Agric. Environ. Med.* 6:105-113.

- Haghighat, F., and De Bellis, L. D. (1998). Material emission rates: literature review, and the impact of indoor air temperature and relative humidity. *Build. Environ.* 33:261-277.
- Hall, D., and Reed, J. (1989). The time dependence of the resuspension of particles. *J. Aerosol Sci.* 20:839-842.
- Harrad, S., Wijesekera, R., Hunter, S., Halliwell, C., and Baker, R. (2004). Preliminary assessment of U.K. human dietary and inhalation exposure to polybrominated diphenyl ethers. *Environ. Sci. Technol.* 38:2345-2350.
- Hartwell, T. D., Perritt, R. L., Pellizzari, E. D., and Michael, L. C. (1992). Results from the 1987 Total Exposure Assessment Methodology (TEAM) Study in Southern California. *Atmos. Environ.* 26A:1519-1527.
- Hatchett, D. W., Kinyanjui, J. M., and Sapochak, L. (2007). FTIR analysis of chemical gradients in thermally processed molded polyurethane foam. *J. Cell. Plast.* 43:183-196.
- Hauser, R., Meeker, J. D., Duty, S., Silva, M. J., and Calafat, A. M. (2006). Altered semen quality in relation to urinary concentrations of phthalate monoester and oxidative metabolites. *Epidemiology.* 17:682-691.
- Healy J. W. (1971). Surface contamination decision levels. LA-4458-MS, UC 41, Los Alamos Scientific Laboratory.
- Heffernan, A. L., Sly, P., Hearn, L., Holling, N., and Mueller, J. F. (2011). The bedroom: an exposure source for polybrominated flame retardants (PBDEs) in infants?, in *Proceedings of Dioxin 2011, Brussels, Belgium.*
- Heissenhuber, A., Heinrich, J., Fahlbusch, B., Borte, M., Wichmann, H.-E., and Bolte, G. (2003). Health impacts of second-hand exposure to cat allergen Fel d 1 in infants. *Allergy.* 58:154-157.
- Henry, C., Minier, J.-P., and Lefèvre, G. (2012). Towards a description of particulate fouling: from single particle deposition to clogging. *Adv. Colloid Interfac.* 185-186:34-76.
- Herbstman, J. B., Sjödin, A., Kurzton, M., Lederman, S. A., Jones, R. S., Rauh, V., Needham, L. L., Tang, D., Niedzwiecki, M., Wang, R. Y., and Perera, F. (2010). Prenatal exposure to PBDEs and neurodevelopment. *Environ. Health Persp.* 118:712-719.

- Heudorf, U., Mersch-Sundermann, V., and Angerer, J. (2007). Phthalates: toxicology and exposure. *Int. J. Hyg. Environ. Health.* 210:623-634.
- Heyes, J., Siebers, R., Biol, F. I., Parkes, A., Pierse, N., and Crane, J. (2009).  $\beta$ -(1,3)-Glucan levels and its determinants in New Zealand bedrooms. *J. Asthma.* 46:64-66.
- Hicks, J. B., Lu, E. T., De Guzman, R., and Weingart, M. (2005). Fungal types and concentrations from settled dust in normal residences. *J. Occup. Environ. Hyg.* 2:481-492.
- Hinds, W. C. (1999). *Aerosol technology: properties, behavior, and measurement of airborne particles.* John Wiley & Sons, Inc., New York.
- Hirsch, T., Hering, M., Bürkner, K., Hirsch, D., Leupold, W., Kerkmann, M. L., Kuhlisch, E., and Jatzwauk, L. (2000). House-dust-mite allergen concentrations (Der f 1) and mold spores in apartment bedrooms before and after installation of insulated windows and central heating systems. *Allergy.* 55:79-83.
- Hoh, E., Hunt, R. N., Quintana, P. J. E., Zakarian, J. M., Chatfield, D. A., Wittry, B. C., Rodriguez, E., and Matt, G. E. (2012). Environmental tobacco smoke as a source of polycyclic aromatic hydrocarbons in settled household dust. *Environ. Sci. Technol.* 46:4174-4183.
- Holopainen, R., Tuomainen, M., Asikainen, V., Pasanen, P., Säteri, J., and Seppänen, O. (2002). The effect of cleanliness control during installation work on the amount of accumulated dust in ducts of new HVAC installations. *Indoor Air.* 12:191-197.
- Hospodsky, D., Qian, J., Nazaroff, W. W., Yamamoto, N., Bibby, K., Rismani-Yazdi, H., and Peccia, J. (2012). Human occupancy as a source of indoor airborne bacteria. *PLoS One.* 7:e34867.
- Howard-Reed, C., Liu, Z., Benning, J., Cox, S., Samarov, D. Leber, D., Hodgson, A. T., Mason, S., Won, D. Y., and Little, J. C. (2011). Diffusion-controlled reference material for volatile organic compound emissions testing: Pilot inter-laboratory study. *Build. Environ.* 46(7):1504-1511.
- Howat, M. G. (1972). Postulates for partitioning variance. I., in *Body Movement Among Its Origins*, *J. Theor. Biol.* 37:125-137.
- Hsu, N.-Y., Chen, C.-Y., Lee, C.-C., and Su, H.-J. (2012). Relationship between indoor phthalate concentrations and dampness or visible mold, in *Proceedings of Healthy Buildings 2012, Brisbane, Australia*, paper ID: 7B.4.

- Hu, B., Freihaut, J. D., Bahnfleth, W. P., and Thran, B. (2008). Measurements and factorial analysis of micron-sized particle adhesion force to indoor flooring materials by electrostatic detachment method. *Aerosol Sci. Technol.* 42:513-520.
- Huang, C. H., Lee, C. I., and Tsai, C. J. (2005). Reduction of particle reentrainment using porous fence in front of dust samples. *ASCE J. Environ. Engr.* 131:1644-1648.
- Hubbard, J. A., Brockmann, J. E., Rivera, D., and Moore, D. G. (2012). Experimental study of impulse resuspension with laser Doppler vibrometry. *Aerosol Sci. Technol.* 46:1303-1312.
- Hugo, J. M., Spence, M. W., and Lickly, T. D. (2000). The determination of the ability of polyurethane foam to release toluene diisocyanate into air. *Appl. Occup. Environ. Hyg.* 15(6):512-519.
- Hwang, H.-M., Park, E.-K., Young, T. M., and Hammock, B. D. (2008). Occurrence of endocrine-disrupting chemicals in indoor dust. *Sci. Total Environ.* 404:26-35.
- Ibrahim, A. H., and Dunn, P. F. (2006). Effects of temporal flow acceleration on the detachment of microparticles from surfaces. *J. Aerosol Sci.* 37:1258-1266.
- Ibrahim, A. H., Dunn, P. F., and Brach, R. M. (2003). Microparticle detachment from surfaces exposed to turbulent air flow: controlled experiments and modeling. *J. Aerosol Sci.* 34:765-782.
- Ibrahim, A. H., Dunn, P. F., and Brach, R. M. (2004). Microparticle detachment from surfaces exposed to turbulent air flow: effects of flow and particle deposition characteristics. *J. Aerosol Sci.* 35:805-821.
- Ibrahim, A. H., Dunn, P. F., and Oazi, M.F. (2008). Experiments and validation of a model for microparticle detachment from a surface by turbulent air flow. *J. Aerosol Sci.* 39:645-656.
- Iglowstein, I., Jenni, O.G., Molinari, L., and Largo, R. H. (2003). Sleep duration from infancy to adolescence: reference values and generational trends. *Pediatrics.* 111:302-307.
- Instanes, C., Hetland, G., Berntsen, S., Løvik, M., and Nafstad, P. (2005). Allergens and endotoxin in settled dust from day-care centers and schools in Oslo, Norway. *Indoor Air.* 15:356-362.

- International Organization for Standardization (ISO). (2006). ISO 16000-10:2006, Indoor air - part 10: determination of the emission of volatile organic compounds from building products and furnishing - emission test cell method. ISO, Switzerland.
- International Organization for Standardization (ISO). (2011). ISO 16000-6:2011, Indoor air - part 6: determination of volatile organic compounds in indoor and test chamber air by active sampling on Tenax TA sorbent, thermal desorption and gas chromatography using MS or MS-FID. ISO, Switzerland.
- Ito, H., Yoshizawa, S., Kumagai, K., Shizawa, K., Kimura, K.-I., Ishikawa, K., and Iwata, T. (1996). Dust deposit evaluation of air conditioning duct, in Proceedings of the 7th International Conference on Indoor Air and Climate, Nagoya, Japan, pp. 965-970.
- Janson, C., De Backer, W., Gislason, T., Plaschke, P., Björnsson, E., Hetta, J., Kristbjarnarson, H., Vermeire, P., and Boman, G. (1996). Increased prevalence of sleep disturbances and daytime sleepiness in subjects with bronchial asthma: a population study of young adults in three European countries. *Eur. Respir. J.* 9:2132-2138.
- Jarfelt, K., Dalgaard, M., Hass, U., Borsh, J., Jacobsen, H., and Ladefoged, O. (2005). Antiandrogenic effects in male rats perinatally exposed to a mixture of di(2-ethylhexyl) phthalate and di(2-ethylhexyl) adipate. *Reprod. Toxicol.* 19:505-515.
- Järnström, H., Saarela, K., Kalliokoski, P., and Pasanen, A. L. (2007). Reference values for structure emissions measured on site in new residential buildings in Finland. *Atmos. Environ.* 41(11):2290-2302.
- Järnström, H., Dols, S., Howard Reed, C., and Persily, A. (2011). Volatile organic compound concentrations and estimation of optimal source strength to achieve a high level acceptance indoors using multi-zone simulation, in Proceedings of the 12th International Conference on Indoor Air and Climate, Austin, TX, paper ID: 990.
- Järnström, H., Vares, S., and Airaksinen, M. (2009). Semivolatile organic compounds and flame retardants: occurrence in indoor environments and risk assessment for indoor exposure. VTT Research Notes 2486.
- Jensen, P. A., and O'Brien, D. (1993). Industrial hygiene, in *Aerosol measurement: principles, techniques, and applications*, Willeke, K., and Baron, P.A., Ed. New York, NY: Van Nostrand Reinhold, pp. 537-559.

- Jiang, Y., Matsusaka, S., Masuda, H., and Qian, Y. (2008). Characterizing the effect of substrate surface roughness on particle-wall interaction with the airflow method. *Powder Technol.* 186:199-205.
- John, W., Fritter, D. N., and Winklmayr, W. (1991). Resuspension induced by impacting particles. *J. Aerosol Sci.* 22:723-736.
- Johnson, D. L., Hunt, A., Griffith, D. A., Hager, J. M., Brooks, J., StellaLevinsohn, H., Lanciki, A., Lucci, R., Prokhorova, D., and Blount, S. L. (2009). Geographic patterns of non-carpeted floor dust loading in Syracuse, New York (USA) homes. *Environ. Geochem. Health.* 31:353-363.
- Jovanovic, S., Felder-Kennel, A., Gabrio, T., Kouros, B., Link, B., Maisner, V., Piechotowski, I., Schick, K. H., Schrimpf, M., Weidner, U., Zöllner, I., and Schwenk, M. (2004). Indoor fungi levels in homes of children with and without allergy history. *Int. J. Hyg. Environ. Health.* 207:369-378.
- Jurcik, B., and Wang, H. C. (1991). The modelling of particle resuspension in turbulent flow. *J. Aerosol Sci.* 22:S149-S152.
- Kalliokoski, P., Kujanpää, L., Pasanen, A.-L., and Pasanen, P. (1995). Cleaning of ventilation systems and its effect on air exchange rates in single family houses, in *Proceedings of Healthy Buildings 1995, Milan, Italy*, pp. 1525-1529.
- Kamens, R., Lee, C., Wiener, R., and Leith, D. (1991). A study to characterize indoor particle in three non-smoking homes. *Atmos. Environ.* 25A:939-948.
- Karlsson, E., Berglund, T., Strömqvist, M., Nordstrand, M., and Fängmark, I. (1999). The effect of resuspension caused by human activities on the indoor concentration of biological aerosols. *J. Aerosol Sci.* 30:S737-738.
- Kassab, A. S., Ugaz, V. M., King, M. D., and Hassan, Y. A. (2013). High resolution study of micrometer particle detachment on different surfaces. *Aerosol Sci. Technol.* 47:351-360.
- Kawakami, T., Isama, K., and Matsuoka, A. (2011). Analysis of phthalic acid diesters, monoester, and other plasticizers in polyvinyl chlorid household products in Japan. *J. Environ. Sci. Heal. A.* 46:855-864.
- Kemmlin, S., Hahn, O., and Jann, O. (2003). Emissions of organophosphate and brominated flame retardants from selected consumer products and building materials. *Atmos. Environ.* 37(39-40):5485-5493.

- Khalifa, H. E., and Elhadidi, B. (2007). Particle levitation due to a uniformly descending flat object. *Aerosol Sci. Technol.* 41:33-42.
- Khoder, M. I., Hassan, S. K., and El-Abssawy, A. A. (2010). An evaluation of loading rate of dust, Pb, Cd, and Ni and metals mass concentration in the settled surface dust in domestic houses and factors affecting them. *Indoor Built Environ.* 19:391-399.
- Kim, S. R., Halden, R. U., and Buckley, T. J. (2007). Volatile organic compounds in human milk: methods and measurements. *Environ. Sci. Technol.* 41:1662-1667.
- Klepeis, N.E., Nelson, W.C., Ott, W.R., Robinson, J.P., Tsang, A.M., Switzer, P., Behar, J.V., Hern, S.C, and Engelmann, W.H. (2001). The National Human Activity Pattern Survey (NHAPS): a resource for assessing exposure to environmental pollutants. *J. Expo. Anal. Environ. Epidemiol.* 11:231-252.
- Kok, J. F., Parteli, E. J. R., Michaels, T. I., and Karam, D. B. (2012). The physics of wind-blown sand and dust. *Rep. Prog. Phys.* 75:1-72.
- Kolari, S., Heikkilä-Kallio, U., Luoma, M., Pasanen, P., Korhonen, P., Nykyri, E., and Reijula, K. (2005). The effect of duct cleaning on perceived work environment and symptoms of office employees in non-problem buildings. *Build. Environ.* 40:1665-1671.
- Kolarik, B., Naydenov, K., Larsson, M., Bornehag, C.-G., and Sundell, J. (2008). The association between phthalates in dust and allergic diseases among Bulgarian children. *Environ. Health Persp.* 116:98-103.
- Korpi, M., Vinha, J., and Kurnitski, J. (2004). Airtightness – measurements in 100 Finnish timber-framed house, in Annex 41 Working Meeting, Glasgow, Scotland.
- Kort, H. S. M., Schober, G., Koren, L. G. H., and Scharringa, J. (1997). Mould-devouring mites differ in guanine excretion from dust-eating Acari, a possible error source in mite allergen exposure studies. *Clin. Exp. Allergy.* 27:921-925.
- Korthals, M., Ege, M., Lick, S., von Mutius, E., and Bauer, J. (2008). Occurrence of *Listeria* spp. in mattress dust of farm children in Bavaria. *Environ. Res.* 107:299-394.
- Kniest, F. M. (1991). Settled house-dust and its aerosols, their importance for allergic patients. *J. Aerosol Sci.* 22:S827-S830.
- Krauter, P., and Biermann, A. (2007). Reaerosolization of fluidized spores in ventilation systems. *Appl. Environ. Microbiol.* 73:2165-2172.

- Krone, C. A., Ely, J. T. A., Klinger, T., and Rando, R. J. (2003). Isocyanates in flexible polyurethane foams. *Bull. Environ. Contam. Toxicol.* 70:328-335.
- Krone, C. A., and Klinger, T. D. (2005). Isocyanates, polyurethane and childhood asthma. *Pediatr. Allergy Immunol.* 16:368-379.
- Krop, E. J. M., Doekes, G., Stone, M. J., Aalberse, R. C., van der Zee, J. S. (2007). Spreading of occupational allergens: laboratory animal allergens on hair-covering caps and in mattress dust of laboratory animal workers. *Occup. Environ. Med.* 64:267-272.
- Kubota, Y., Hall, J. W., and Higuchi, H. (2009). An experimental investigation of the flowfield and dust resuspension due to idealized human walking. *ASME J. Fluids Engr.* 131:081104.
- Kubota, Y., and Higuchi, H. (2013). Aerodynamic particle resuspension due to human foot and model foot motions. *Aerosol Sci. Technol.* 47:208-217.
- Kulmala, I., Säämänen, A., and Enbom, S. (1996). The effect of contaminant source location on worker exposure in the near-wake region. *Ann. Occup. Hyg.* 40:511-523.
- Kurkela, J. A., Brown, D. P., Raula, J., and Kauppinen, E. I. (2006). New apparatus for studying powder deagglomeration. *Powder Technol.* 14:164-171.
- Kurnitski, J., and Seppänen, O.A. (2008). Trends and drivers in the Finnish ventilation and AC market. *AIVC VIP.* 20:1-9.
- Küchen, V. (1998). Konzentration an staub und mikroorganismen in lüftungskanälen von raumluftechnischen anlagen: felduntersuchungen unter einatz unterschiedlicher staubmeßverfahren. Diploma Thesis, Technische Universität Berlin, Berlin, Germany (in German).
- Laatikainen, T., Pasanen, P., Korkonen, L., Nevalainen, A., and Ruuskanen, J. (1991). Methods for evaluating dust accumulation in ventilation ducts, in *Proceedings of Healthy Buildings 1991*, Atlanta, GA.
- Lai, A. C. K., Thatcher, T. L., and Nazaroff, W. W. (2000). Inhalation transfer factors for air pollution health risk assessment. *J. Air & Waste Manage. Assoc.* 50:1688-1699.
- Lai, A. C. K., Byrne, M. A., and Goddard, A. J. H. (2002). Experimental studies of the effect of rough surfaces and air speed on aerosol deposition in a test chamber. *Aerosol Sci. Technol.* 36:973-982.

- Larsen, R. I. (1958). The adhesion and removal of particles attached to air filter surfaces. *Am. Ind. Hyg. Assoc. J.* 19:265-270.
- Larsson, M., Hagerhed-Engman, L., Kolarik, B., James, P., Lundin, F., Janson, S., Sundell, J., and Bornehag, C.-G. (2010). PVC – as flooring material – and its association with incident asthma in a Swedish child cohort study. *Indoor Air.* 20:494-501.
- Lau, S., Illi, S., Sommerfeld, C., Niggemann, B., Bergmann, R., von Mutius, E., and Wahn, U. (2000). Early exposure to house-dust mite and cat allergens and development of childhood asthma: a cohort study. *Lancet.* 356:1392-1397.
- Laverge, J., Novoselac, A., Corsi, R. L., and Janssens, A. (2013). Experimental assessment of exposure to gaseous pollutants from mattresses and pillows while asleep. *Build. Environ.* 59:203-210.
- Laverge, J., Spilak, M. P., and Novoselac, A. (2015). Experimental assessment of the inhalation zone of standing, sitting and sleeping persons. *Build. Environ.* 82:258-266.
- Lavoie, J., Marchand, G., Cloutier, Y., and Lavoué, J. (2011). Validation of the criteria for initiating the cleaning of heating, ventilation, and air-conditioning (HVAC) ductwork under real conditions. *J. Occup. Environ. Hyg.* 8:467-472.
- Lawson, J. A., Dosman, J. A., Rennie, D. C., Beach, J. R., Newman, S. C., Crowe, T., and Senthilselvan, A. (2012). Endotoxin as a determinant of asthma and wheeze among rural dwelling children and adolescents: A case-control study. *BMC Pulm. Med.* 12:56.
- Layton, D. W., and Beamer, P. I. (2009). Migration of contaminated soil and airborne particulates to indoor dust. *Environ. Sci. Technol.* 43:8199-8205.
- Lazaridis, M., and Drossinos, Y. (1998). Multilayer resuspension of small identical particles by turbulent flow. *Aerosol Sci. Technol.* 28:548-560.
- Lefcoe, N. M., and Inoulet, I. I. (1975). Particulates in domestic premises II. Ambient levels and indoor-outdoor relationships. *Arch. Envir. Health.* 30:565-570.
- Leung, T. F., Wong, Y. S., Chan, I. H. S., Yung, E., Sy, H. Y., Lam, C. W. K., and Wong, G. W. K. (2011). Domestic exposure to aeroallergens in Hong Kong families with asthmatic children. *Pediatr. Pulmonol.* 46:632-639.

- Lewis, R. D., Condoor, S., Batek, J., Hean Ong, K., Backer, D., Sterling, D., Siria, J., Chen, J. J., and Ashley, P. (2006). Removal of lead contaminated dusts from hard surfaces. *Environ. Sci. Technol.* 40:590-594.
- Lewis, R. G., Fortune, C. R., Willis, R. D., Camann, D. E., and Antley, J. T. (1999). Distribution of pesticides and polycyclic aromatic hydrocarbons in house dust as a function of particle size. *Environ. Health Perspect.* 107:721-726.
- Lewis, R. J. (2001). *Hawley's Condensed Chemical Dictionary 14th Edition*. John Wiley & Sons: New York.
- Lewis, R. J. (2007). *Hawley's Condensed Chemical Dictionary 15th Edition*. John Wiley & Sons: New York.
- Lewis, R. J. (1993). *Hawley's Condensed Chemical Dictionary 12th Edition*. John Wiley & Sons: New York.
- Liang, Y. and Xu, Y. (2014a). Improved method for measuring and characterizing phthalate emissions from building materials and its application to exposure assessment. *Environ. Sci. Technol.* 48(8):4475-4484.
- Liang, Y. and Xu, Y. (2014b). Emission of phthalates and phthalate alternatives from vinyl flooring and crib mattress covers: the influence of temperature. *Environ. Sci. Technol.* 48(24):14228-14237.
- Lidén, G., and Waher, J. (2010). Experimental investigation of the concept of a 'breathing zone' using a mannequin exposed to a point source of inertial/sedimenting particles emitted with momentum. *Ann. Occup. Hyg.* 54:100-116.
- Lin, C.-C., Yu, K.-P., Zhao, P., Lee, G. W.-M. (2009). Evaluation of impact factors on VOC emissions and concentrations from wooden flooring based on chamber tests. *Build. Environ.* 44:525-533.
- Loosmore, G. A., and Hunt, J. R. (2000). Dust resuspension without saltation. *J. Geophys. Res.* 105:20,663-20,671.
- Lu, L., Tamura, T., and Togawa, T. (1999). Detection of body movements during sleep by monitoring of bed temperature. *Physiol. Meas.* 20:137-148.
- Luda, M. P., Bracco, P., Costa, L., and Levchik, S. V. (2004). Discolouration in fire retardant flexible polyurethane foams. Part I. Characterisation. *Polym. Degrad. Stabil.* 83:215-220.

- Lundgren, B., Jonsson, B., and Ek-Olausson, B. (1999). Materials emission of chemicals - PVC flooring materials. *Indoor Air*. 9(3):202-208.
- Madsen, T. and Gibson, R. (2008). Toxic baby furniture the latest case for making products safe from the start. Environment California Research and Policy Center.
- Mage, D. T., and Ott, W. R. (1996). Accounting for nonuniform mixing and human exposure in indoor environments, in *Characterizing Sources of Indoor Air Pollution and Related Sink Effects*, ASTM STP 1287, Bruce A. Tichenor, Ed., American Society for Testing and Materials, pp. 263-278.
- Mahmic, A., Tovey, E., Molloy, C. A., and Young, L. (1998). House dust mite allergen exposure in infancy. *Clin. Exp. Allergy*. 28:1487-1492.
- Marks, G. B. (1998). House dust mite exposure as a risk factor for asthma: benefits of avoidance. *Allergy*. 53:108-114.
- Marshall, J. D. and Nazaroff, W. W. (2007) Intake fraction, in *Exposure Analysis*, Ott, W. R., Steinemann, A., and Wallace, L. (Eds.), CRC Press, Boca Raton, Florida.
- Masuck, I., Hutzler, C., Jann, O., and Luch, A. (2011). Inhalation exposure of children to fragrances present in scented toys. *Indoor Air*. 21:501-511.
- Matsumoto, M., Hirata-Koizumi, M., and Ema, M. (2008). Potential adverse effects of phthalic acid esters on human health: a review of recent studies on reproduction. *Regul. Toxicol. Pharm.* 50:37-49.
- Matsusaka, S., and Masuda, H. (1996). Particle reentrainment from a fine powder layer in a turbulent air flow. *Aerosol Sci. Technol.* 24:69-84.
- McBride, S., Ferro, A. R., Ott, W. R., Switzer, P., and Hildemann, L. M. (1999). Investigations of the proximity effects for pollutants in the indoor environment. *J. Expo. Anal. Environ. Epidemiol.* 9:602-621.
- McClelland, J. F., Jones, R. W., Luo, S., and Seaverson, L. M. (1992). A practical guide to FTIR photoacoustic spectroscopy, in *Practical Sampling Techniques for Infrared Analysis*. CRC Press.
- McDonagh, A. and Byrne, M. A. (2014) The influence of human physical activity and contaminated clothing on particle resuspension, *J. Environ. Radioactiv*, 127, 119-126.

- McGee, S. P., Konstantinov, A., Stapleton, H. M., and Volz, D. C. (2013). Aryl phosphate esters within a major PentaBDE replacement product induce cardiotoxicity in developing zebrafish embryos: potential role of the aryl hydrocarbon receptor. *Toxicol. Sci.* 133:144-156
- Melkin, T., Reponen, T., Toivola, M., Koponen, V., Husman, T., Hyvärinen, A., and Nevalainen, A. (2002). Size distributions of airborne microbes in moisture-damaged and reference school buildings of two construction types. *Atmos. Environ.* 36:6031-6039.
- Meeker, J. D., and Stapleton, H. M. (2010). House dust concentrations of organophosphate flame retardants in relation to hormone levels and semen quality parameters. *Environ. Health Persp.* 118:318-323.
- Michel, O., Nagy, A.-M., Schroeven, M., Duchateau, J., Néve, J., Fondu, P., and Sergysels, R. (1997). Dose-response relationship to inhaled endotoxin in normal subjects. *Am. J. Respir. Crit. Care Med.* 156:1157-1164.
- Miguel, A. F., Aydin, M., and Reis, A. H. (2005). Indoor deposition and forced re-suspension of respirable particles. *Indoor Built Environ.* 14:391-396.
- Mihrshahi, S., Marks, G., Vanlaar, C., Tovey, E., and Peat, J. (2002). Predictors of high house dust mite allergen concentrations in residential homes in Sydney. *Allergy.* 57:137-142.
- Ministry of the Environment, Housing and Building Department. (2012). D2 National building code of Finland, indoor climate and ventilation of buildings, regulation and guidelines 2012. Ministry of the Environment, Espoo, Finland.
- Montaya, L. D., and Hildemann, L. M. (2001). Evolution of the mass distribution of resuspended cat allergen (Fel d 1) indoors following a disturbance. *Atmos. Environ.* 35:859-866.
- Montaya, L. D., and Hildemann, L. M. (2005). Size distributions and height variations of airborne particulate matter and cat allergen indoors immediately following dust-disturbing activities. *J. Aerosol Sci.* 36:735-749.
- Morose, G. and Becker, M. (2013). A collaborative industry and university alternative assessment of plasticizers for wire and cable. In *Chemical Alternatives Assessments: Issues in Environmental Science and Technology*, The Royal Society of Chemistry.

- Mortazavi, R. (2005). Reentrainment of submicron solid particles. Doctoral Dissertation, Virginia Commonwealth University, Richmond, VA.
- Möritz, M., Peters, H., and Rüdén, H. (2001). Methods to determine the dust loads of air ducts in HVAC systems. VDI-Berichte. 1603:153-161 (in German).
- Mukai, C., Siegel, J. A., and Novoselac, A. (2009). Impact of airflow characteristics on particle resuspension from indoor surfaces. *Aerosol Sci. Technol.* 43:1-11.
- Nagorka, R., Conrad, A., Scheller, C., Süßenbach, B., and Moriske, H.-J. (2011). Diisononyl 1,2-cyclohexanedicarboxylic acid (DINCH) and Di(2-ethylhexyl) terephthalate (DEHT) in indoor dust samples: concentration and analytical problems. *Int. J. Hyg. Environ. Health.* 214:26-35.
- Nam, H.-S., Siebers, R., Lee, S.-H., Park, J.-S., Kim, Y.-B., Choi, Y.-J., Lee, S.-H., and Crane, J. (2008). House dust mite allergens in domestic homes in Cheonan, Korea. *Korean J. Parasitol.* 46:187-189.
- National Institute of Standards and Technology (NIST). NIST Chemistry WebBook, 1,3-Benzenedicarboxylic acid, bis(2-ethylhexyl) ester. <http://webbook.nist.gov/cgi/cbook.cgi?Name=Bis%282-ethylhexyl%29+isophthalate&Units=SI> (Accessed 08/10/2014).
- National Resources Defense Council (NRDC). Letter to U.S. CPSC. Re: Notice of Availability of Draft Guidance Regarding Which Children's Products are Subject to the Requirements of CPSIA Section 108; Request for Comments and Information. [http://www.nrdc.org/health/files/hea\\_09032501a.pdf](http://www.nrdc.org/health/files/hea_09032501a.pdf) (Accessed 12/10/2013).
- Nazaroff, W. W. and Weschler, C. J. (2004). Cleaning products and air fresheners: exposure to primary and secondary air pollutants. *Atmos. Environ.* 38(18):2841-2865.
- Nazaroff, W. W. (2008). Inhalation intake fraction of pollutants from episodic indoor emissions. *Build. Environ.* 43:269-277.
- Needham, L. L., Barr, D. B., and Calafat, A. M. (2005). Characterizing children's exposures: beyond NHANES. *NeuroToxicology.* 26:547-553.
- Nicholson, K. W. (1988). Review article: a review of particle resuspension. *Atmos. Environ.* 22:2639-2651.

- Nicholson, K. (1993). Wind tunnel experiments on the resuspension of particulate material. *Atmos. Environ. Part A. Gen. Topics.* 27:181-188.
- Nielsen, J. B., Valbjorn, O., Gravesen, S., and Molhave, L. (1990). Stov i ventilationsanlaeg Horsholm. Statens Byggeforskningsinstitut, SBI-Rapport 206 (in Danish).
- Nirlo, E. L., Crain, N. E., Corsi, R. L., and Siegel, J. A. (2014). Volatile organic compounds in fourteen U.S. retail stores. *Indoor Air.* 24(5):484-494.
- Nitschke, D., and Schmidt, E. (2009). A new approach to model the re-entrainment of settled particles based on film theory of fluid mass transfer processes. *Part. Part. Syst. Char.* 26:58-68.
- Nitschke, D., and Schmidt, E. (2010). Experimental study and modelling of the resuspension of settled particles. *Chem. Ingen. Technik.* 82:2119-2127 (in German).
- Nordtest. (1995). Building materials: emission of volatile compounds - field and laboratory emission cell (FLEC) (NT BUILD 438). Nordtest, Espoo, Finland.
- Oberoi, R. C., Choi, J.-I., Edwards, J. R., Rosati, J. A., Thornburg, J., and Rodes, C. E. (2010). Human-induced particle re-suspension in a room. *Aerosol Sci. Technol.* 44:216-229.
- O'Meara, T., and Tovey, E. (2000). Monitoring personal allergen exposure. *Clin. Rev. Allergy Immunol.* 18:341-395.
- Oppermann, H., Doering, C., Sobottka, A., Krämer, U., and Thriene, B. (2001). Comparison of East and West German households with regard to exposure to house dust mites and mould fungi. *Gesundh. Wes.* 63:85-89 (in German).
- Otani, Y., Namiki, N., and Emi, H. (1995). Removal of fine particles from smooth flat surfaces by consecutive pulse air jets. *Aerosol Sci. Technol.* 23:665-673.
- Paajanen, M., Katainen, J., Pakarinen, O. H., Foster, A. S., and Lahtinen, J. (2006). Experimental humidity dependency of small particle adhesion on silica and titania. *J. Colloid Interface Sci.* 304:518-523.
- Pasanen, P., Nevalainen, A., Ruuskanen, J., and Kalliokoski, P. (1992). The composition and location of dust settled in supply air ducts, in Proceedings of the 13th Air Infiltration and Ventilation Centre (AIVC) Conference on Ventilation for Energy Efficiency and Optimum Indoor Air Quality, Nice, France, pp. 481-488.

- Pasanen, P., Pasanen, A.-L., and Kalliokoski, P. (1995). Hygienic aspects of processing oil residues in ventilation ducts. *Indoor Air*. 5:62-68.
- Patisaul, H. B., Roberts, S. C., Mabrey, N., McCaffrey, K. A., Gear, R. B., Braun, J., Belcher, S. M., and Stapleton, H. M. (2012). Accumulation and endocrine disrupting effects of the flame retardant mixture Firemaster® 550 in rats: an exploratory assessment. *J. Biochem. Mol. Toxic.* 27:124-126.
- Peat, J. K., Tovey, E., Toelle, B. G., Haby, M. M., Gray, E. J., Mahmic, A., and Woolcock, A. J. (1996). House dust mite allergens. A major risk factor for childhood asthma in Australia. *Am. J. Respir. Crit. Care Med.* 153:141-146.
- Pitten, F.-A., Scholler, M., Krüger, U., Effendy, I., and Kramer, A. (2001). Filamentous fungi and yeasts on mattresses covered with different encasings. *Eur. J. Dermatol.* 11:534-537.
- Platts-Mills, T., Heymann, P., Longbottom, J., and Wilkins, S. (1986). Airborne allergens associated with asthma: particle sizes carrying dust mite and rat allergens measured with a cascade impactor. *J. Allergy Clin. Immunol.* 77:850-857.
- Qian, J., and Ferro, A. R. (2008). Resuspension of dust particles in a chamber and associated environmental factors. *Aerosol Sci. Technol.* 42:566-578.
- Qian, J., Ferro, A. R., and Fowler, K. R. (2008). Estimating the resuspension rate and residence time of indoor particles. *J. Air & Waste Manage. Assoc.* 58:502-516.
- Qian, J., Peccia, J., and Ferro, A. R. (2014) Walking-induced particle resuspension in indoor environments, *Atmos. Environ.*, 89, 464-481.
- Que Hee, S. S., Peace, B., Clark, C. S., Boyle, J. R., Bornschein, R. L., and Hammond, P. B. (1985). Evolution of efficient methods to sample lead sources, such as house dust and hand dust, in the homes of children. *Environ. Res.* 38:77-95.
- Rabinovitch, N., Liu, A. H., Zhang, L., Rodes, C. E., Foarde, K., Dutton, S. J., Murphy, J. R., and Gelfand, E. W. (2005). Importance of the personal endotoxin cloud in school-age children with asthma. *J. Allergy Clin. Immunol.* 116:1053-1057.
- Raja, S., Xu, Y., Ferro, A. R., Jaques, P. A., and Hopke, P. K. (2010). Resuspension of indoor aeroallergens and relationship to lung inflammation in asthmatic children. *Environ. Inter.* 36:8-14.

- Rao, C. Y., Cox-Ganser, J. M., Chew, G. L., Doekes, G., and White, S. (2005). Use of surrogate markers of biological agents in air and settled dust samples to evaluate a water-damaged hospital. *Indoor Air*. 15:89-97.
- Raunemaa, T., Kulmala, M., Saari, J., Olin, M. and Kulamala, M. H. (1989). Indoor air aerosol model: transport indoors and deposition of fine and coarse particles. *Aerosol Sci. Technol.* 11:11-25.
- Rauert, C., Lazarov, B., Harrad, S., Covaci, A., and Stranger, M. (2014). A review of chamber experiments for determining specific emission rates and investigating migration pathways of flame retardants. *Atmos. Environ.* 82:44-55.
- Reeks, M., and Hall, D. (2001). Kinetic models for particle resuspension in turbulent flows: theory and measurement. *J. Aerosol Sci.* 32:1-31.
- Rennie, D. C., Lawson, J. A., Kirychuk, S. P., Paterson, C., Willson, P. J., Senthilselvan, A., and Cockcroft, D. W. (2008). Assessment of endotoxin levels in the home and current asthma and wheeze in school-age children. *Indoor Air*. 18:447-453.
- Reponen, T., Grinshpun, S. A., Conwell, K. L., Wiest, J., and Anderson, M. (2001). Aerodynamic versus physical size of spores: measurement and implication for respiratory deposition. *Grana*. 40:119-125.
- Reponen, T., Seo, S.-C., Grimsley, F., Lee, T., Crawford, C., and Grinshpun, S. A. (2007). Fungal fragments in moldy houses: a field study in homes in New Orleans and Southern Ohio. *Atmos. Environ.* 41:8140-8149.
- Rich, D. Q., Yiin, L-M., Rhoads, G. G., Glueck, D. H., Weisel, C., and Liroy, P. J. (1999). A field comparison of two methods for sampling lead in household dust. *J. Expo. Anal. Environ. Epid.* 2:106-112.
- Rim, D., and Novoselac, A. (2009). Transport of particulate and gaseous pollutants in the vicinity of a human body. *Build. Environ.* 44:1840-1849.
- Rim, D., and Novoselac, A. (2010). Occupational exposure to hazardous airborne pollutants: effects of source characteristics. *J. Occup. Environ. Hyg.* 7:683-692.
- Roberts, J. W., Glass, G., and Mickelson, L. (2004). A pilot study of the measurement and control of deep dust, surface dust, and lead in 10 old carpets using the 3-spot test while vacuuming. *Arch. Environ. Contam. Toxicol.* 48:16-23.

- Roberts, J. W., Wallace, L. A., Camann, D. E., Dickey, P., Gilbert, S. G., Lewis, R. G., and Takaro, T. K. (2009). Monitoring and reducing exposure of infants to pollutants in house dust. *Rev. Environ. Contam. Toxicol.* 201:1-39.
- Rodes, C. E., Kamens, R. M., and Wiener, R. W. (1991). The significance and characteristics of the personal activity cloud on exposure assessment measurements for indoor air. *Indoor Air.* 2:123-145.
- Rodes, C. E., Lawless, P. A., Evans, G. F., Sheldon, L. S., Williams, R. W., Vette, A. F., Creason, J. P., and Walsh, D. (2001a). The relationships between personal PM exposures for elderly populations and indoor and outdoor concentrations for three retirement center scenarios. *J. Expo. Anal. Environ. Epid.* 11:103-115.
- Rodes, C. E., Newsome, J. R., Vanderpool, R. W., Antley, J. T., and Lewis, R. G. (2001b). Experimental methodologies and preliminary transfer factor data for estimation of dermal exposures to particles. *J. Expo. Anal. Environ. Epid.* 11:123-139.
- Rodes, C. E. (2011). Indoor aerosol exposure assessment, in *Aerosol measurement: principles, techniques, and applications*, Kulkarni, P., Baron, P. A., and Willeke, K., Ed. New York, NY: John Wiley & Son, Inc., pp. 615-634.
- Rosati, J. A., Thornburg, J., and Rodes, C. (2008). Resuspension of particulate matter from carpet due to human activity. *Aerosol Sci. Technol.* 42:472-482.
- Rossi, M. Neonatal exposure to DEHP (di-2-ethylhexyl phthalate) and opportunities for prevention. In *Health Care Without Harm*, Washington, D.C., 2002.
- Rothe, J., Cordelair, H., and Wehman, C. (2001). New catalysts for low VOC in flexible slabstock foam. *J. Cell. Plast.* 37:207-220.
- Roze, E., Meijer, L., Bakker, A., Van Braeckel, K. N. J. A., Sauer, P. J. J., and Bos, A. F. (2009). Prenatal exposure to organohalogenes, including brominated flame retardants, influences motor, cognitive, and behavioral performance at school age. *Environ. Health Persp.* 117:1953-1958.
- Rudel, R. A., and Perovich, L. J. (2009). Endocrine disrupting compounds in indoor and outdoor air. *Atmos. Environ.* 43:170-181.
- Rumchev, K., Spickett, J., Bulsara, M., Phillips, M., and Stick, S. (2004). Association of domestic exposure to volatile organic compounds with asthma in young children. *Thorax.* 59:746-751.

- Saboori, A. M., Lang, D. M., and Newcombe, D. S. (1991). Structural requirements for the inhibition of human monocyte carboxylesterase by organophosphorus compounds. *Chem. Biol. Interact.* 80:327-338.
- Sakaguchi, M., Inouye, S., Yasueda, H., and Shida, T. (1992). Concentration of airborne mite allergens (Der I and Der II) during sleep. *Allergy.* 47:55-57.
- Salares, V. R., Hinde, C. A., and Miller, J. D. (2009). Analysis of settled dust in homes and fungal glucan in air particulate collected during HEPA vacuuming. *Indoor Built Environ.* 18:485-491.
- Salthammer, T., Fuhrmann, F., and Uhde, E. (2003). Flame retardants in the indoor environment - Part II: release of VOCs (triethylphosphate and halogenated degradation products) from polyurethane. *Indoor Air.* 13(1):49-52.
- Schneider, T., Kildesø, J., and Breum, N. O. (1999). A two compartment model for determining the contribution of sources, surface deposition and resuspension to air and surface dust concentration levels in occupied rooms. *Build. Environ.* 34:583-595.
- Schossler, P., Schripp, T., Salthammer, T., and Bahadir, M. (2011). Beyond phthalates: gas phase concentrations and modeled gas/particle distribution of modern plasticizers. *Sci. Total Environ.* 409:4031-4038.
- Sehmel, G. A. (1980). Particle resuspension: a review. *Environ. Int.* 4:107-127.
- Seifert, B. (1998). Die untersuchung von hausstaub im hinblick auf expositionsabschätzungen. *Bundesgesundheitsblatt.* 41:383-391 (in German).
- Shalat, S. L., Liroy, P. J., Schmeelck, K., and Mainelis, G. (2007). Improving estimation of indoor exposure to inhalable particles for children in the first year of life. *J. Air & Waste Manage. Assoc.* 57:934-939.
- Shalat, S. L., Stambler, A. A., Wang, Z., Mainelis, G., Emoekpere, O. H., Hernandez, M., Liroy, P. J., and Black, K. (2011). Development and in-home testing of the pretoddler inhalable particulate environmental robotic (PIPER Mk IV) sampler. *Environ. Sci. Technol.* 45:2945-2950.
- Shao, Y., Raupach, M. R., and Findlater, P. A. (1993). Effect of saltation bombardment on the entrainment of dust by wind. *J. Geophys. Res.* 98:12,719-12,726.

- Shaughnessy, R., and Vu, H. (2012). Particle loadings and resuspension related to floor coverings in a chamber and in occupied school environments. *Atmos. Environ.* 55:515-524.
- Shimohira, M., Shiiki, T., Sugimoto, J., Ohsawa, Y., Fukumizu, M., Hasegawa, T., Iwakawa, Y., Nomura, Y., and Segawa, M. (1998). Video analysis of gross body movements during sleep. *Psychiatry Clin. Neurosci.* 52:176-177.
- Silber, M. H., Ancoli-Israel, S., Bonnet, M. H., Chokroverty, S., Grigg-Damberger, M. M., Hirshkowitz, M., Kapen, S., Keenan, S. A., Kryger, M. H., Penzel, T., Pressman, M. R., and Iber, C. (2007). The visual scoring of sleep in adults. *J. Clin. Sleep Med.* 3:121-131.
- Slinn, W. G. N. (1978). Parameterizations for resuspension and for wet and dry deposition of particles and gases for use in radiation dose calculations. *Nucl. Safety* 19:205-219.
- Smedley, G. T., Phares, D. J., and Flagan, R. C. (1999). Entrainment of fine particles from surfaces by gas jets impinging at normal incidence. *Exp. Fluids.* 26:324-334.
- Soleimani, M., and Rafinejad, J. (2008). House dust mite contamination in hotels and inns in Bandar Abbas, South of Iran. *Iran. J. Environ. Health Sci. Eng.* 5:207-210.
- Southey, A., Fox, M., Yeomans, T., and Mitchell, B. (2011). A comparison of the characteristics of ISO fine test dust versus real house dust, in Proceedings of the 12th International Conference on Indoor Air and Climate, Austin, TX, paper ID: 868.
- Spilak, M. P., Boor, B. E., Novoselac, A., and Corsi, R. L. (2014). Impact of bedding arrangements, pillows, and blankets on particle resuspension in the sleep microenvironment. *Build. Environ.* 81:60-68.
- Sporik, R., Holgate, S. T., Platts-Mills, T. A. E., and Cogswell, J. J. (1990). Exposure to house-dust mite allergen (Der p 1) and the development of asthma in childhood. *New Engl. J. Med.* 323:502-507.
- Stapleton, H., Kelly, S., Allen, J., McClean, M., and Webster, T. (2008). Measurement of polybrominated diphenyl ethers on hand wipes: Estimating exposure from hand-to-mouth contact. *Environ. Sci. Technol.* 42(9):3329-3334.
- Stapleton, H. M., Klosterhaus, S., Eagle, S., Fuh, J., Meeker, J. D., Blum, A., and Webster, T. F. (2009). Detection of organophosphate flame retardants in furniture foam and U.S. house dust. *Environ. Sci. Technol.* 43:7490-7495.

- Stapleton, H. M., Klosterhaus, S., Keller, A., Lee Ferguson, P., van Bergen, S., Cooper, E., Webster, T. F., and Blum, A. (2011). Identification of flame retardants in polyurethane foam collected from baby products. *Environ. Sci. Technol.* 45:5323-5331.
- Stephens, B., Siegel, J. A., and Novoselac, A. (2011). Operational characteristics of residential and light-commercial air-conditioning systems in a hot and humid climate zone. *Build. Environ.* 46:1972-1983.
- Stapleton, H. M., Sharma, S., Getzinger, G., Lee Ferguson, P., Gabriel, M., Webster, T. F., and Blum, A. (2012). Novel and high volume use flame retardants in US couches reflective of the 2005 PentaBDE phase out. *Environ. Sci. Technol.* 46(24):13432-13439.
- Stores, G., Ellis, A. J., Wiggs, L., Crawford, C., and Thomson, A. (1998). Sleep and psychological disturbance in nocturnal asthma. *Arch. Dis. Child.* 78:413-419.
- Strachan, D. P., and Carey, I. M. (1995). Home environment and severe asthma in adolescence: a population based case-control study. *BMJ.* 311:1053-1056.
- Stubner, A. H., Dillon, K. H., and Kohler, C. L. (2000). Home remediation for respiratory health: a feasibility study. *Fam. Community Health.* 22:1-15.
- Su, H.-J., Wu, P.-C., Chen, H.-L., Lee, F.-C., and Lin, L.-L. (2001). Exposure assessment of indoor allergens, endotoxin, and airborne fungi for homes in southern Taiwan. *Environ. Res. Sec. A.* 85:135-144.
- Sundell, J., Levin, H., Nazaroff, W. W., Cain, W. S., Fisk, W. J., Grimsrud, D. T., Gyntelberg, F., Li, Y., Persily, A. K., Pickering, A. C., Samet, J. M., Spengler, J. D., Taylor, S. T., and Weschler, C. J. (2011). Ventilation rates and health: multidisciplinary review of the scientific literature. *Indoor Air.* 21:191-204.
- Swan, S. H., Main, K. M., Liu, F., Stewart, S. L., Kruse, R. L., Calafat, A. M., Mao, C. S., Redmon, J. B., Ternand, C. L., Sullivan, S., and Teague, J. L. (2005). Decrease in anogenital distance among male infants with prenatal phthalate exposure. *Environ. Health Persp.* 113:1056-1061.
- Swanson, M., Agarwal, M., and Reed, C. (1985). An immunochemical approach to indoor aeroallergen quantitation with a new volumetric air sampler: studies with mite, roach, cat, mouse and guinea pig antigens. *J. Allergy Clin. Immunol.* 76:724-729.

- Swanson, M., Campbell, A., Klauck, M., and Reed, C. (1989). Correlations between levels of mite and cat allergens in settled and airborne dust. *J. Allergy Clin. Immunol.* 83:776-783.
- Tadmor, J., and Zur, I. (1981). Resuspension of particles from a horizontal surface. *Atmos. Environ.* 15:141-149.
- Taheri, M., and Bragg, G. M. (1992). A study of particle resuspension in a turbulent flow using a Preston tube. *Aerosol Sci. Technol.* 16:15-20.
- Täubel, M., Rintala, H., Pitkäranta, M., Paulin, L., Laitinen, S., Pekkanen, J., Hyvärinen, A., and Nevalainen, A. (2009). The occupant as a source of house dust bacteria. *J. Allergy Clin. Immunol.* 124:834-840.
- Theerachaisupakij, W., Matsusaka, S., Kataoka, M., and Masuda, H. (2002). Effects of wall vibration on particle deposition and reentrainment in aerosol flow. *Advanced Powder Technol.* 13:287-300.
- Tian, Y., Sul, K., Howard Reed, C., Leber, D., and Ferro, A. R. (2011). A comparative study of estimating particle resuspension rate using a consistent test mechanism, in Proceedings of the 12th International Conference on Indoor Air and Climate, Austin, TX, paper ID: 584.
- Tian, Y., Sul, K., Qian, J., Modal, S. and Ferro, A. R. (2014). A comparative study of walking-induced dust resuspension using a consistent test mechanism. *Indoor Air.* 24: 592–603.
- Tischer, C., Gehring, U., Chen, C. M., Kerkhof, M., Koppelman, G., Sausenthaler, S., Herbarth, O., Schaaf, B., Lehmann, I., Krämer, U., Berdel, D., von Berg, A., Bauer, C. P., Koletzko, S., Wichmann, H. E., Brunekreef, B., and Heinrich, J. (2011). Respiratory health in children, and indoor exposure to (1,3)- $\beta$ -D-glucan, EPS mould components and endotoxin. *Eur. Respir. J.* 37:1050-1059.
- Thatcher, T. L., Lai, A. C. K., Jackson, R. M., Sextro, R. G., and Nazaroff, W. W. (2002). Effects of Room Furnishings and Air Speed on Particle Deposition Rates Indoors. *Atmos. Environ.* 36:1811-1819.
- Thatcher, T. L., and Layton, D. W. (1995). Deposition, resuspension, and penetration of particles within a residence. *Atmos. Environ.* 29:1487-1497.
- Toms, L-M. L., Hearn, L., Kennedy, K., Harden, F., Bartkow, M., Temme, C., and Mueller, J. F. (2009). Concentrations of polybrominated diphenyl ethers (PBDEs)

- in matched samples of human milk, dust, and indoor air. *Environ. Int.* 35:864-869.
- Tovey, E., Chapman, M., and Platts-Mills, T. (1981). Mite faeces are a major source of house dust mite allergens. *Nature.* 289:592-593.
- Tovey, E., and Ferro, A. R. (2012). Time for new methods for avoidance of house dust mite and other allergens. *Curr. Allergy Asthma Rep.* 12:465-477.
- Tullo, A. (2003). Great Lakes to phase out flame retardants. *Chem. Eng. News.* 81(45):13-13.
- UK Environment Agency. (2009). Environmental risk evaluation report: Triphenyl phosphate (CAS no. 115-86-6). In Bristol, United Kingdom.
- U.S. Consumer Production Safety Commission (CPSC). (2008). Consumer Product Safety Improvement Act (CPSIA) of 2008. U.S. CPSC, Bethesda, Maryland.
- U.S. Consumer Production Safety Commission (CPSC). (2010). Review of exposure and toxicity data for phthalate substitutes. U.S. CPSC, Bethesda, Maryland.
- U.S. Consumer Production Safety Commission (CPSC). Notice of availability of draft guidance regarding which children's products are subject to the requirements of CPSIA section 108; request for comments and information. <http://www.cpsc.gov//PageFiles/97758/draftphthalatesguidance.pdf> (Accessed 12/10/2013).
- U.S. Environmental Protection Agency (EPA). (1996). Comprehensive abatement performance study, volume 1: summary report, EPA 230-R-94-013a. U.S. EPA, Washington, D.C.
- U.S. Environmental Protection Agency (EPA). (2009). Exposure factors handbook, chapter 6, inhalation rates. U.S. EPA, Washington, D.C.
- U.S. Environmental Protection Agency (EPA). Toxicological review of phenol. <http://www.epa.gov/iris/toxreviews/0088tr.pdf> (12/10/2013),
- van den Bemt, L., de Vries, M. P., van Knapen, L., Jansen, M., Goossens, M., Muris, J. W. M., and van Schayck, C. P. (2006). Influence of mattress characteristics on house dust mite allergen concentration. *Clin. Exp. Allergy.* 36:233-237.

- van Der Wal, J. F., Hoogeveen, A. K., and Wouda, P. (1997). The influence of temperature on the emission of volatile organic compounds from pvc flooring, carpet, and paint. *Indoor Air*. 7:215-221.
- Vangronsveld, E., Berckmans, S., and Spence, M. (2013). Toluene diisocyanate emission to air and migration to a surface from a flexible polyurethane foam. *Ann. Occup. Hyg.* 57(5):650-661.
- Vartiainen, E., Petäys, T., Haahtela, T., Jousilahti, P., and Pekkanen, J. (2002). Allergic diseases, skin prick test responses, and IgE levels in North Karelia, Finland, and the Republic of Karelia, Russia. *J. Allergy Clin. Immunol.* 109:643-648.
- Verschoor, L. and Verschoor, A. H. (2014). Nonoccupational and occupational exposure to isocyanates. *Curr. Opin. Pulm. Med.* 20:199-204
- Visitsunthorn, N., Chirdjirapong, V., Pootong, V., Jirapongsananuruk, O., Pacharn, P., Weeravejsukit, S., Mahakittikun, V., and Vichyanond, P. (2010). The accumulation of dust mite allergens on mattresses made of different kinds of materials. *Asian Pac. J. Allergy Immunol.* 28:155-161.
- Vogel, K., Blümer, N., Korthals, M., Mittelstädt, J., Garn, H., Ege, M., von Mutius, E., Gattermann, S., Bufe, A., Goldmann, T., Schwaiger, K., Renz, H., Brandau, S., Bauer, J., Heine, H., and Holst, O. (2008). Animal shed *Bacillus licheniformis* spores possess allergy-protective as well as inflammatory properties. *J. Allergy Clin. Immunol.* 122:307-312.
- Vuillerme, V., Fohr, J. P., Saulnier, J. P., Oriot, D., Saulnier, J. B., and Blay, D. (1998). Convective heat transfer around an infant head. *Ann. N Y Acad. Sci.* 858:310-317.
- Wahlström, R., Rovio, S., and Suurnäkki, A. (2012). Partial enzymatic hydrolysis of microcrystalline cellulose in ionic liquids by *Trichoderma reesei* endoglucanases. *RSC Advances*. 2:4472-4480.
- Wallace, L. A. (1996). Indoor particles: a review. *J. Air Waste Manag. Assoc.* 46:98-126.
- Wang, L., and Chen, Q. 2007. Evaluation of some assumptions used in multizone airflow network models. *Build. Environ.* 43:1671-1677.
- Wang, S., Zhao, B., Zhou, B., and Tan, Z. (2012). An experimental study on short-time particle resuspension from inner surfaces of straight ventilation ducts. *Build. Environ.* 53:119-127.

- Wang, Z., Shalat, S. L., Black, K., Liroy, P. J., Stambler, A. A., Emoekpere, O. H., Hernandez, M., Han, T., Ramagopal, M., and Mainelis, G. (2012). Use of a robotic sampling platform to assess young children's exposure to indoor bioaerosols. *Indoor Air*. 22:159-169.
- Waring, M. S., and Siegel, J. A. (2008). Particle loading rates for HVAC filters, heat exchangers, and ducts. *Indoor Air*. 18:209-224.
- Watkins, D. J., Mclean, M. D., Fraser, A. J., Weinberg, J., Stapleton, H. M., Sjödin, A., and Webster, T. F. (2012). Impact of dust from multiple microenvironments and diet on PentaBDE body burden. *Environ. Sci. Technol.* 46:1192-1200.
- Weber, D., Fülöp, G., and Hummel, D. O. (1991). Pyrolysis-gas chromatography/Fourier-transform infrared spectrometry of poly(ester urethane) elastomers. *Makromol. Chem., Macromol. Symp.* 52:151-160.
- Wei, H., Turyk, M., Cali, S., Dorevitch, S., Erdal, S., and Li, A. (2009). Particle size fractionation and human exposure to polybrominated diphenyl ethers in indoor dust from Chicago. *J. Environ. Sci. Health, Part A*. 44:1353-1361.
- Welle, F., Wolz, G., and Franz, R. (2005). Migration of plasticizers from PVC tubes into enteral feeding solutions. *Pharma Int.* 3:17-21 (in German).
- Wen, H., and Kasper, G. (1989). On the kinetics of particle reentrainment from surfaces. *J. Aerosol Sci.* 20:483-498.
- Weschler, C. J., and Nazaroff, W. W. (2008). Semivolatile organic compounds in indoor environment. *Atmos. Environ.* 42:9018-9040.
- Weschler, C. J., Langer, S., Fischer, A., Bekö, G., Toftum, J., and Clausen, G. (2011). Squalene and cholesterol in dust from Danish homes and daycare centers. *Environ. Sci. Technol.* 45:3872-3879.
- Weschler, C. J., and Nazaroff, W. W. (2012). SVOC exposure indoors: fresh look at dermal pathways. *Indoor Air*. 22:356-377.
- Weschler, C. J. and Nazaroff, W. W. (2013). Dermal uptake of organic vapors commonly found in indoor air. *Environ. Sci. Technol.* 48(2):1230-1237.
- Wheldon, A. E. (1982). Energy balance in the newborn baby: use of a manikin to estimate radiant and convective heat loss. *Phys. Med. Biol.* 27:285-296.

- Wiglusz, R., Sitko, E., Nickel, G., Jarnuszkiewicz, I., and Igielska, B. (2002). The effect of temperature on the emission of formaldehyde and volatile organic compounds (VOCs) from laminate flooring - case study. *Build. Environ.* 37(1):41-44.
- Wilde-Frenz, J., and Schulz, H. (1983). Rate and distribution of body movements during sleep in humans. *Percept. Mot. Skills.* 56:275-283.
- Williams, L. K., Ownby, D. R., Maliarik, M. J., and Johnson, C. C. (2005). The role of endotoxin and its receptors in allergic disease. *Ann. Allerg. Asthma Im.* 94:323-332.
- Winnebeck, K. H. (2011). An abbreviated alternatives assessment process for product designers: a children's furniture manufacturing case study. *J. Clean. Prod.* 19:464-476.
- Wolkoff, P. (1998). Impact of air velocity, temperature, humidity, and air on long-term VOC emissions from building products. *Atmos. Environ.* 32:2659-2668.
- Woodcock, A. A., Steel, N., Moore, C. B., Howard, S. J., Custovic, A., and Denning, D. W. (2006). Fungal contamination of bedding. *Allergy.* 61:140-142.
- Wu, F. F.-S., Siebers, R., Biol, F. I., Chang, C.-F., Hsieh, S.-W., Wu, M.-W., Chen, C.-Y., Pierse, N., and Crane, J. (2009). Indoor allergens and microbial bio-contaminants in homes of asthmatic children in central Taiwan. *J. Asthma.* 46:745-749.
- Wu, F. F.-S., Wu, M.-W., Pierse, N., Crane, J., and Siebers, R. (2012). Daily vacuuming of mattresses significantly reduces house dust mite allergens, bacterial endotoxin, and fungal  $\beta$ -glucan. *J. Asthma.* 49:139-143.
- Wu, Y.-L., Davidson, C. I., and Russell, A. G. (1992). Controlled wind tunnel experiments for particle bounceoff and resuspension. *Aerosol Sci. Technol.* 17:245-262.
- Xiong, J. Y., Zhang, Y. P., Wang, X. K., and Chang, D. W. (2008). Macro-meso two-scale model for predicting the VOC diffusion coefficients and emission characteristics of porous building materials. *Atmos. Environ.* 42(21):5278-5290.
- Xu, Y. and Little, J. C. (2006). Predicting emissions of SVOCs from polymeric materials and their interaction with airborne particles. *Environ. Sci. Technol.* 40(2):456-461.
- Xu, Y., Cohen Hubal, E. A., and Little, J.C. (2010). Predicting residential exposure to phthalate plasticizer emitted from vinyl flooring: sensitivity, uncertainty, and implications for biomonitoring. *Environ. Health Persp.* 118:253-258.

- Xu, Y., Liu, Z., Park, J., Clausen, P. A., Benning, J. L., and Little, J. C. (2012). Measuring and predicting the emission rate of phthalate plasticizer from vinyl flooring in a specially-designed chamber. *Environ. Sci. Technol.* 46(22):12534-12541.
- Xu, Y., Liang, Y., Urquidi, J. R., and Siegel, J. A. (2014). Phthalates and polybrominated diphenyl ethers in retail stores. *Atmos. Environ.* 87:53-64.
- Yang, X., Chen, Q., and Zhang, J. S. (1998). Impact of early stage incomplete mixing on estimating VOC emissions in small test chambers. *Indoor Air.* 8:180-189.
- You, R., Cui, W., Chen, C., and Zhao, B. (2012). Measuring the short-term emission rates of particles in the "personal cloud" with different clothes and activity intensities in a sealed chamber. *Aerosol Air Qual. Res.* 13:911-921.
- You, S., and Wan, M. P. (2012). A new turbulent-burst based resuspension model with humidity effect, in *Proceedings of Healthy Buildings 2012, Brisbane, Australia*, paper ID: 10F.4.
- You, S. and Wan, M. P. (2014) Experimental investigation and modelling of human-walking-induced particle resuspension, *Indoor Built Environ.*, doi:10.1177/1420326X14526424.
- Yuan, H., Little, J. C., Marand, E., and Liu, Z. (2007). Using fugacity to predict volatile emissions from layered materials with a clay/polymer diffusion barrier. *Atmos. Environ.* 41(40):9300-9308.
- Zanobetti, A., Redline, S., Schwartz, J., Rosen, D., Patel, S., O'Connor, G. T., Lebowitz, M., Coull, B. A., and Gold, D. R. (2010). Associations of PM10 with sleep and sleep-disordered breathing in adults from seven U.S. urban areas. *Am. J. Respir. Crit. Care Med.* 182:819-825.
- Zartarian, V. G., Ott, W. R., and Duan, N. (1997). A quantitative definition of exposure and related concepts. *J. Expo. Anal. Env. Epid.* 7:411-437.
- Zhang, J., Zhang, J. S., Chen, Q., and Yang, X. (2002a) A critical review on VOC sorption models. *ASHRAE Trans.* 108(1):162-174.
- Zhang, J., Zhang, J. S., and Q. Chen. (2002b). Effects of environmental conditions on the VOC sorption by building materials – Part I: experimental results. *ASHRAE Trans.* 108(2):273-282.

- Zhang, J., Zhang, J. S., and Q. Chen. (2003). Effects of environmental conditions on the VOC sorption by building materials– Part II: model evaluations. *ASHRAE Trans.* 109(1):167-178.
- Zhang, X., Ahmadi, G., Qian, J., and Ferro, A. (2008). Particle detachment, resuspension and transport due to human walking in indoor environments. *J. Adhes. Sci. Technol.* 22:591-621.
- Zhao, D., Little, J. C., and Cox, S. S. (2004). Characterizing polyurethane foam as a sink for or source of volatile organic compounds in indoor air. *J. Environ. Eng-ASCE.* 130:983-989.
- Zhou, B., Zhao, B., and Tan, Z. (2011). How particle resuspension from inner surfaces of ventilation ducts affects indoor air quality – a modeling analysis. *Aerosol Sci. Technol.* 45:996-1009.
- Zhu, Y., Zhao, B., Zhou, B., and Tan, Z. (2012). A particle resuspension model in ventilation ducts. *Aerosol Sci. Technol.* 46:222-235.
- Ziskind, G., Yarin, L. P., Peles, S., and Gutfinger, C. (2002). Experimental investigation of particle removal from surfaces by pulsed air jets. *Aerosol Sci. Technol.* 36:652-659.
- Zock, J. P., Brunekreef, B., Hazebroek-Kampschreur, A. A. J. M., and Roosjen, C. W. (1994). House dust mite allergen in bedroom floor dust and respiratory health of children with asthmatic symptoms. *Eur. Respir. J.* 7:1254-1259.
- Zuraimi, M. S. (2010). Is ventilation duct cleaning useful? A review of the scientific evidence. *Indoor Air.* 20:445-457.
- Zuraimi, M. S., Magee, R., and Nilsson, G. (2012). Development and application of a protocol to evaluate impact of duct cleaning on IAQ of office buildings. *Build. Environ.* 56:86-94.

## VITA

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