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Indoor inhalation intake fractions of fine particulate matter: Review of influencing factors

Natasha Hodas[a,b], Miranda Loh[c], Hyeong-Moo Shin[d], Dingsheng Li[e], Deborah Bennett[d], Thomas E. McKone[d,g], Olivier Jolliet[e], Charles J. Weschler[h,i], Matti Jantunen[j], Paul Lioy[h], Peter Fantke[k,*]

[a] Division of Chemical Engineering, California Institute of Technology, Pasadena, CA, USA
[b]Department of Environmental Science and Management, Portland State University, Portland, OR, USA
[c] Institute of Occupational Medicine, Edinburgh, United Kingdom
[d] Department of Public Health Sciences, University of California, Davis, CA, USA
[e] Department of Environmental Health Sciences, University of Michigan, Ann Arbor, MI, USA
[f] School of Public Health, University of California, Berkeley, CA 94720, USA
[g] Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA
[h]Environmental and Occupational Health Sciences Institute, Rutgers University, Piscataway, NJ, USA
[i] International Centre for Indoor Environment and Energy, Technical University of Denmark, Kgs. Lyngby, Denmark
[j]Department of Environmental Health, National Institute for Health and Welfare, Helsinki, Finland
[k] Department of Management Engineering, Technical University of Denmark, Kgs. Lyngby, Denmark

*Corresponding author:
Peter Fantke
Quantitative Sustainability Assessment Division, Department of Management Engineering
Technical University of Denmark, Produktionstorvet 424
2800 Kgs. Lyngby
Denmark
Tel.: +45 45254452
Fax: +45 45933435
E-mail: pefan@dtu.dk

Abstract

Exposure to fine particulate matter ($PM_{2.5}$) is a major contributor to the global human disease burden. The indoor environment is of particular importance when considering the health effects associated with $PM_{2.5}$ exposures because people spend the majority of their time indoors and $PM_{2.5}$ exposures per unit mass emitted indoors are two to three orders of magnitude larger than exposures to outdoor emissions. Variability in indoor $PM_{2.5}$ intake fraction ($iF_{\mathrm{in,total}}$), which is defined as the integrated cumulative intake of $PM_{2.5}$ per unit of emission, is driven by a combination of building-specific, human-specific, and pollutant-specific factors. Due to a limited availability of data characterizing these factors, however, indoor emissions and intake of $PM_{2.5}$ are not commonly considered when evaluating the environmental performance of product life cycles. With the aim of addressing this barrier, a literature review was conducted and data characterizing factors influencing $iF_{\mathrm{in,total}}$ were compiled. In addition to providing data for the calculation of $iF_{\mathrm{in,total}}$ in various indoor environments and for a range geographic regions, this paper discusses remaining limitations to the incorporation of $PM_{2.5}$-derived health impacts into life cycle assessments and makes recommendations regarding future research.

Practical Implications

This paper reviews and summarizes the factors that influence indoor inhalation intake fraction of fine particulate matter, with a focus on primary particle emissions indoors. It provides valuable data for the calculation of indoor inhalation intake fraction for a range of indoor environments and contributes to the effort to incorporate $PM_{2.5}$-derived health impacts into life cycle assessment.

Key words: fine particulate matter ($PM_{2.5}$), human exposure, indoor air, intake fraction, life cycle impact assessment (LCIA), ventilation

Introduction

Human exposure to fine particulate matter ($PM_{2.5}$) is a major contributor to disease burden on a global scale (WHO, 2002, 2013). The indoor environment is a particularly important venue for exposure to $PM_{2.5}$ because people spend the majority of

their time indoors (Klepeis et al., 2001; Phillips and Moya, 2014 and references therein). Further, due to the lesser degree of dilution, chemical transformation, and dispersion, as well as the higher density of occupants indoors, exposures per unit mass of $PM_{2.5}$ emitted indoors are two to three orders of magnitude larger than exposures to emissions to the outdoor environment (Smith, 1988; Lai et al., 2000; Klepeis and Nazaroff, 2006; Ilacqua et al., 2007; Nazaroff, 2008). In order to fully assess the impacts associated with all emission sources of $PM_{2.5}$ and to evaluate the life cycle environmental performance of products and systems (e.g., energy and transport systems, food products and production systems, and consumer products), there is a need for the incorporation of $PM_{2.5}$ exposures and the associated health effects into Life Cycle Impact Assessments (LCIA), with a specific need for the consideration of the impacts related to indoor exposures to $PM_{2.5}$ emitted or formed indoors.

Due to current limitations in data availability and modeling tools that systematically combine indoor and outdoor intakes from indoor and outdoor sources, as well as challenges in consistently linking indoor and outdoor intakes to exposure-response, indoor sources and related intake of $PM_{2.5}$ are currently not considered in product-related assessments (Humbert et al., 2015). To integrate indoor sources into such assessment frameworks, there is a need for (1) the identification of factors contributing substantially to variability in $PM_{2.5}$ exposure and an examination of the value of accounting for this variability when assessing $PM_{2.5}$ health impacts, (2) the aggregation and evaluation of modeling tools and data available for assessing human exposure to $PM_{2.5}$, and (3) a thorough assessment of the availability of exposure-response functions (ERFs) and the appropriateness of ERF shape (e.g., linear, non-linear, presence of a threshold) for a variety of health outcomes (Fantke et al., 2015). With the aim of addressing these barriers and the lack of a standardized methodology to estimate exposures and health effects, the United Nations Environment Programme (UNEP)-Society for Environmental Toxicology and Chemistry (SETAC) Life Cycle Initiative formed a task force to provide guidance for the assessment of $PM_{2.5}$ exposures and associated health effects (Jolliet et al., 2014; Fantke et al., 2015). Under the framework of this task force and with input from an international team of experts, this paper constitutes a first step toward incorporating indoor $PM_{2.5}$ exposures into LCIA by characterizing the

factors that drive variability in the inhalation intake fraction of $PM_{2.5}$ derived from indoor sources.

Inhalation intake fraction (*iF*), which is defined as the ratio of mass of a pollutant inhaled by an exposed human population to the total mass associated with a given source (Bennett et al., 2002), provides a well-suited metric by which to consider $PM_{2.5}$ impacts in the context of LCIA. As an exposure metric, *iF* integrates components that are key to such assessments: (1) it describes source-receptor relationships in a manner that allows for direct comparisons across emission sources and (2) it can readily be related to potential toxicity in terms of specific health outcomes when exposure-response relationships are known (Bennett et al., 2002; Ilacqua et al., 2007; Nazaroff, 2008; Fantke et al., 2015). Table 1 illustrates the contributions of $PM_{2.5}$ derived from indoor sources (S_{in}) and outdoor sources (S_{out}) to indoor intake, outdoor intake, total intake, and the intake fraction of $PM_{2.5}$. As is described in detail below, this paper reviews the major factors influencing the inhalation intake fraction of $PM_{2.5}$ derived from indoor sources (Table 1, Equation 1). Examples of common indoor sources of $PM_{2.5}$ include cooking, household and office appliances, smoking, cleaning, candles, and heating appliances or stoves. Additional efforts are currently underway within the UNEP-SETAC LCIA framework to characterize the other aspects of $PM_{2.5}$ intake and intake fraction shown in Table 1.

Indoor inhalation intake fraction ($iF_{\mathrm{in,total}}$) describes the total inhalation intake of $PM_{2.5}$ (in kg) per unit mass emitted indoors (in kg). Two components contribute to $iF_{\mathrm{in,total}}$ (Table 1, Equation 1): (1) the fraction of $PM_{2.5}$ emitted or formed indoors that is taken in via inhalation indoors ($iF_{\mathrm{in}\rightarrow\mathrm{in}}$) and (2) the fraction of $PM_{2.5}$ emitted or formed indoors that is transported outdoors and taken in via inhalation outdoors ($iF_{\mathrm{in}\rightarrow\mathrm{out}}$). However, because $PM_{2.5}$ of indoor origin experiences a greater degree of dispersion and dilution following transport outdoors and outdoor population density is lower than indoors, $iF_{\mathrm{in}\rightarrow\mathrm{out}}$ is typically three orders of magnitude smaller than $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ (Smith, 1988; Lai et al., 2000; Klepeis and Nazaroff, 2006; Ilacqua et al., 2007; Nazaroff, 2008; Humbert et al., 2011). Thus, in calculations of $iF_{\mathrm{in,total}}$, $iF_{\mathrm{in}\rightarrow\mathrm{out}}$ can be considered negligible compared to $iF_{\mathrm{in}\rightarrow\mathrm{in}}$. As a result, this paper focuses on characterizing the major factors contributing to variability in $iF_{\mathrm{in}\rightarrow\mathrm{in}}$, as this term dominates $iF_{\mathrm{in,total}}$. While not

the main focus, we also note the importance of interactions between pollutants of outdoor and indoor origin and the influence of outdoor $PM_{2.5}$ sources on cumulative indoor intake (Table1, Equation 2) and briefly discuss the current state of knowledge regarding these aspects.

Nazaroff (2008) divided the factors influencing variability in $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ for primary particles into three categories: (1) factors related to building characteristics (e.g., ventilation, airflow, and mixing rates), (2) factors related to occupant characteristics and behaviors (e.g., inhalation rates and occupancy/activity patterns), and (3) pollutant dynamics (e.g., first order removal processes and sorptive interactions). That study noted the need for a "richly constituted tool kit to effectively comprehend the system of the human health risk associated with products and processes in indoor environments." Humbert et al. (2011) provided an initial set of parameters characterizing two archetypal indoor environments (residences within the United States [U.S.] and mechanically ventilated offices). Herein, we expand on that effort by developing an inventory of parameters (i.e., a "tool kit") to (1) address each of the factors influencing $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ discussed by Nazaroff (2008) and (2) allow for the characterization of multiple archetypal indoor environments (e.g., residences, offices, schools, etc.), covering a broad range of geographic scales.

Methods

For each category of factors influencing $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ (building, occupant, and pollutant factors), sub-groups with expertise in that specific field were created within an indoor-air task force. Literature searches conducted by each sub-group were obtained from Web of Science, Google Scholar, and/or SCOPUS with search terms representing sources of variability related to the above-described categories (e.g., "air exchange rate measurements," "building ventilation," "commercial building ventilation rates," "inhalation rates," "indoor particle deposition," "indoor particle emission rates," etc.). When available, review papers were preferentially selected to be included in this review due to its multidimensional focus. Collected references were then reviewed and compiled to provide an inventory of data-sources (e.g., peer-reviewed scientific articles and reports) and data regarding each factor influencing $iF_{\mathrm{in}\rightarrow\mathrm{in}}$. We included key papers (i.e.,

those with the most sound experimental/modeling practices, those that provide the greatest breadth of data, and those that allow for consideration of a range of exposure scenarios) in the present review and provide data from those papers in the supporting information (SI). In general, the data compiled include summary statistics (i.e., mean, standard deviation, geometric mean, geometric standard deviation, percentiles, minimum, and maximum values) from individual studies conducted under a variety of experimental conditions and for a range of geographic locations. Where possible, data are categorized by country/geographic region and specific conditions in order to allow for the selection of data most relevant to an exposure-scenario of interest. Each factor contributing to variability in $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ is discussed in an individual section below.

Building Factors

Building-specific factors influencing $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ include building volume and ventilation (Table 1, Equations 1 and 2). Building ventilation is a key parameter in estimating $iF_{\mathrm{in}\rightarrow\mathrm{in}}$, as it drives the transport, dispersion, and dilution of $PM_{2.5}$ emitted indoors. Indoor ventilation is driven by three processes: (1) leakage through cracks in the building shell and walls (infiltration/exfiltration), (2) airflow through open windows and doors (natural ventilation), and (3) mechanical ventilation (i.e., flow driven by fans; Chan et al., 2005; US EPA, 2011). Infiltration/exfiltration and natural ventilation are driven by pressure gradients that exist across the building envelope due to indoor-outdoor temperature differences and wind (US EPA, 2011). Mechanical ventilation systems range between exhaust- or supply-only systems (e.g., bathroom and kitchen exhaust fans/hoods), balanced supply and exhaust systems, localized unitary/single-zone systems, and central/integrated systems (Sippola and Nazaroff, 2002; Brelih and Seppänen, 2011; Litiu, 2012). Building ventilation is typically quantified as whole-building/whole-zone air exchange rates (AERs) [h^{-1}] or, as is common for non-residential/commercial buildings, volumetric flow rate normalized by building occupancy, volume, or floor area [L s^{-1} $person^{-1}$, L s^{-1} m^{-3}, L s^{-1} m^{-2}] (Persily, 2015). In the following paragraphs, we review the body of literature focused on characterizing these building properties and processes in a range of building archetypes.

Residential Buildings

Residential ventilation rates have been most heavily studied in Europe (Hänninen et al., 2011; Dimitroulopoulou, 2012 and references therein; Asikainen et al., 2013; Orru et al., 2014) and North America (Figure 1a) (Clark et al., 2010; Persily et al., 2010; US EPA, 2011 and references therein; Chen et al., 2012; MacNeil et al., 2012, 2014; El Orch et al., 2014; Bari et al., 2014; Breen et al., 2014; Persily, 2015). While more limited in their number and scope, some studies have also been carried out in New Zealand (McNeil et al., 2012), Asia (Baek et al. 1997; Williams and Eunice, 2013; Huang et al., 2014; Park et al., 2014; Li and Li, 2015; Shi et al., 2015), Africa, and South America (Williams and Eunice, 2013 and references therein) (Figure 1a). In addition to those studying the housing stock in broad geographic regions, some studies have focused on homes with specific characteristics (e.g., new homes, energy-efficient homes, low-income/public housing; Zota et al., 2005; US EPA, 2011). A limited number of studies have characterized ventilation in homes in developing countries (Williams and Eunice, 2013, L'Orange et al., 2015, and references therein) (Figure 1a). The use of solid fuels for cooking and heating, particularly in developing countries, is a leading indoor air quality issue on a global scale, with approximately 4.3 million premature deaths annually attributed to related pollutant exposures (www.WHO.int/indoorair/en). As a result, such measurements for homes in developing countries are very important to the effort to incorporate the impacts of indoor $PM_{2.5}$ exposures into LCIA.

The above-described body of work illustrates that there is spatial variability in residential ventilation with climate, building construction characteristics, home age, heating, ventilation, and air conditioning (HVAC) system configurations, ventilation standards and regulations, and residence type (i.e., detached, single family homes, apartments) (Figure 2a). Temporal heterogeneity in ventilation rates results from variability in meteorological conditions and human behaviors such as window opening and mechanical ventilation system usage. The compilation of data characterizing homes over a broad range of geographic scales, housing types, seasons, and meteorological conditions is needed because the prevalence of different ventilation systems varies strongly across these factors. For example, AERs in 100% of both apartments and detached homes in Bulgaria are driven by infiltration and natural ventilation. On the other

hand, 48% of detached homes in Finland have mechanical ventilation systems. This proportion increases to 72% when considering apartments (Litiu, 2012). To aid in the selection of representative ventilation parameters when calculating $iF_{\text{in}\rightarrow\text{in}}$, the ventilation rates and air exchange rate data provided here are categorized by country, home type, season, and ventilation system where the available data allow for this (Figure 1a and SI). Studies characterizing window-opening behavior and/or mechanical ventilation system usage and runtime (e.g., Iwashita and Akasaka, 1997; Chao, 2001; Wallace et al., 2002; Johnson and Long, 2005; US EPA, 2011; Fabi et al., 2012; Marr et al., 2012; Breen et al., 2014; El Orch et al., 2014; Gorenzenski et al., 2014; Levie et al., 2014; Persily, 2015; Stephens, 2015) provide needed information for accounting for temporal and spatial variability in ventilation conditions.

Figure 2a summarizes available residential air exchange rate data, with detailed data provided in the SI. For all residential AER measurements combined, we observed a median value of 0.50 h^{-1} (95% confidence interval [CI] = 0.08, 8.2 h^{-1}) (Figure 2a), which is slightly higher than the recommended median value of 0.45 h^{-1} for homes in the U.S. provided in the Environmental Protection Agency Exposure Factors Handbook (US EPA EFH) (US EPA, 2011). This difference can likely be attributed, at least in part, to our inclusion of a small number measurements from high AER homes in developing countries, as well as differences in home characteristics and ventilation systems across nations. While treated as a single distribution above for the purpose of comparison against the recommended value in the US EPA EFH, residential AERs are likely best characterized by a bimodal distribution. This is evidenced by differences in the median AER values for homes in developed and developing countries: median (95% CI) = 0.48 (0.08 2.26) h^{-1} and 14.1 (2.0, 61.0) h^{-1}, respectively.

Many of the studies described above in which air exchange and ventilation are measured also provide data regarding the volume/floor area of the homes studied (Figure 1f). It is important to note that homes included are not necessarily statistically representative of the housing stock and this influences estimates of both home volume and ventilation. Population-level data describing home characteristics can also typically be gathered from census and housing survey databases (e.g., the American Census, American Housing Survey, Eurostat, and Census India). Recommended values for

various housing and building characteristics are also available in reports summarizing exposure factors in several countries (US EPA, 2011; Phillips and Moya, 2014 and references therein). Available measurements of residential volumes illustrate their high variability, both within and across nations, with values ranging from 15 – 1446 m^3 (median [95% CI] = 247 [41, 971] m^3) (see SI). The median residential volume for the studies considered in this work is lower than the recommended value provided in the US EPA EFH (492 m^3) (US EPA, 2011), likely illustrating differences in residential volumes across regions of the world.

Non-Residential Buildings

Ventilation measurements have been conducted in a range of non-residential buildings, including retail stores (US EPA, 2011; Zaatari et al., 2014 and references therein; Dutton et al., 2015), schools, kindergartens, and daycare centers (Coley and Beisteiner, 2002; Wargocki et al., 2002; Emmerich and Crum, 2006; Mi et al., 2006; Li et al., 2007; Guo et al., 2008; Santamouris et al., 2008; Brehlih and Seppänen, 2011; Sundell et al., 2011; Aelenei et al., 2013; Canha et al., 2013) offices (Persily and Gorfain, 2004; Dimitroupoulou and Bartzis, 2013), fitness facilities (Zaatari et al., 2014), jails (Seppänen et al., 1999; Li et al., 2007), and healthcare facilities, hospitals, and nursing homes (Wargocki et al., 2002, Li et al., 2007 and references therein). Summary statistics of more than 700 measurements from 17 studies, for example, have been compiled for retail facilities, bars/restaurants, healthcare facilities, fitness facilities, offices, and schools (Zaatari et al., 2014). As is true for residential ventilation rates, measurements in non-residential buildings are more heavily focused in North America and Europe, with a smaller number of studies also conducted in Asia (Figure 1a). Non-residential AERs are summarized in Figure 2a, with more detailed information (e.g., categorized by building type) provided in the SI. We observed a median AER for non-residential buildings of 1.5 h^{-1} (95% CI = 0.29, 9.1 h^{-1}).

The above-described studies again demonstrate geographic variability in ventilation-system characteristics and the prevalence of mechanically and naturally ventilated buildings, as well as temporal variability in ventilation with meteorological conditions, window opening, and HVAC-system operation. For example, 100% of

schools and kindergartens are naturally ventilated in Italy, while only 5% and 28% of kindergartens and schools are naturally ventilated in Finland (Litiu, 2012). Sippola and Nazaroff (2002) note that single-zone HVAC systems are common in smaller commercial buildings with floor areas on the order of 150 m^2, while central systems dominate in larger buildings (>1000 m^2) such as malls, university buildings, theaters, and retail centers.

A small number of studies discuss window-opening and HVAC-system-use behavior in commercial/non-residential buildings (e.g., Fabi et al., 2012; Roetzel et al., 2010; Ramos and Stephens, 2014; D'Oca and Hong, 2014; Li et al., 2015; Stephens, 2015). Two recent studies (Bennett et al., 2012; Chan et al., 2014) conducted detailed measurements of AERs and ventilation rates in thirty seven commercial buildings and nineteen retail stores, respectively, and provided summary statistics for various building types (e.g., grocery stores, hardware stores, restaurants, healthcare facilities, and public assembly spaces) and for varying ventilation conditions (e.g., with doors open/closed, with and without mechanical ventilation systems in use).

As was true for the residential ventilation studies, many of the above-described studies provide information regarding the characteristics of the buildings studied, including building volume and/or floor area; however, again, these values are typically not statistically representative of the full range of non-residential building stock. The Building Assessment Survey and Evaluation (BASE) Study provides measurements of building and occupied-space size for 100 randomly selected large office buildings in the U.S. (Persily and Gorfain, 2004). US EPA (2011) is also a valuable resource for summary statistics of volume data for buildings with a wide range of uses and sizes (e.g., warehouses, shopping malls, schools, and healthcare facilities). As a result of the range of building uses, commercial building volumes display a large degree of variability, ranging from 408 to 849,505 m^3 (median [95% CI] = 3,398 [461, 192,554] m^3) (see SI).

Inter- and Intra-Zonal Airflows and Mixing

Inter-zonal and intra-zonal airflow and local-scale mixing (i.e., convective and advective mixing on intra-zonal scales) can be of importance in both residential and non-residential indoor environments, specifically when considering differences in exposures

and $iF_{\text{in}\rightarrow\text{in}}$ for building occupants with varying proximities to sources of interest (Drescher et al., 1995; Nazaroff, 2008). Measurements of inter-zonal and intra-zonal flows are limited. In addition, these flows vary within and across buildings and depend on multiple factors including door opening, ventilation conditions, home layout, and temperature gradients (Klepeis, 2004; McGrath et al., 2014). Thus, selecting a representative value or sampling from a distribution of measured values when calculating $iF_{\text{in}\rightarrow\text{in}}$ is not straightforward. As a result, such flows typically must be modeled for an exposure scenario of interest.

Commonly used models for the estimation of inter-zonal flows include COMIS (Feustel, 1998) and CONTAM (Walton and Dols, 2010). AER and inter-zonal flows predicted with CONTAM and/or COMIS have been evaluated against measurements conducted in more than ten countries and for a variety of building types (Emmerich, 2001 and references therein; Haas et al., 2002; Emmerich et al., 2004). Details regarding the required inputs and use of these models are available in their respective users' manuals (Feustel, 1998; Walton and Dols, 2010).

Computational fluid dynamics (CFD) has been used to explicitly model airflow and turbulence on smaller, within-room scales (e.g. Gadgil et al., 2003; Zhang and Chen, 2007; Zhao et al., 2007, 2008). Pragmatically, multi-zone and zonal modeling methods can be combined by nesting an intra-zonal model within an inter-zonal model (Stewart and Ren, 2003, 2006; Wang and Chen, 2007), so that a specific room of interest (e.g. the room with a $PM_{2.5}$ source) can be divided into several small zones, while other rooms within the same home/building are treated as larger, well-mixed zones.

Alternatively, Bennett and Furtaw (2004) provide an estimate of a room-to-room air exchange rate distribution (mean = 3 h^{-1}, coefficient of variation = 0.30) based on measurements conducted under varying ventilation conditions within a single house. Du et al., (2012) characterized overall and season-specific inter-zonal airflows between living areas and bedrooms in 126 homes in Detroit, MI as the percentage of room-specific air exchange attributable to air entering from another zone. Along the same lines, Hellweg et al. (2009) suggest ranges of values for within-zone mixing factors (0.1 to 1.0) and inter-zonal air exchange rates (3 to 30 m^3/min). These are examples of midway approaches between the typical single, well-mixed compartment assumption and more

complex approaches based on CFD. Understanding the influence of smaller-scale flows on $iF_{\text{in}\rightarrow\text{in}}$ is an important area of future research, with a rate coefficient representing the airflow between zones (including the near-person zone and the rest of an indoor environment) being a resulting metric of interest for use in LCIA.

Human Exposure Factors

Inhalation Rate

Inhalation intake fraction is directly related to the inhalation rate (*IR*) of the subjects or population of interest (Table 1, Equation 1). Inhalation rates vary within and across individuals with multiple factors including age, sex, body weight, and fitness and activity levels (Figure 2b) (US EPA, 2011). Studies quantifying *IR* are largely based on relationships between oxygen uptake and consumption, metabolism, and energy expenditure (US EPA, 2011). Using various methods to quantify energy expenditure and oxygen consumption, multiple studies have measured *IR* for broad, representative populations (e.g., US EPA, 2011 and references therein; Richardson and Stantec, 2013; Jang et al., 2014a), while others have focused on specific populations of interest (US EPA, 2011 and references therein). Recommended values of *IR* for the general population categorized by age, gender, and activity level are available for the U.S. (US EPA, 2011), Canada (Richardson and Stantec, 2013), and Korea (Figure 1b) (Jang et al., 2014a). As is discussed below, materials are available to allow for the estimation of *IR* for populations for which such measurements have not been conducted. Specific populations of interest for which *IR* studies have been conducted include children, adults and children with asthma, and pregnant and lactating adult and adolescent women (US EPA, 2011). Such studies allow for the consideration of $iF_{\text{in}\rightarrow\text{in}}$ for susceptible populations or during specific periods of susceptibility.

Inhalation rates are commonly reported as long-term (m^3 day^{-1}), or short-term (m^3 min^{-1}) rates. The latter allow for distinguishing differences in *IR* arising from different levels of activity. When assessing chronic exposures, long-term *IR*s can be utilized to characterize $iF_{\text{in}\rightarrow\text{in}}$; however, short-term *IR*s are needed when considering acute exposures or exposures associated with a particular activity (i.e., where the emission is represented by a pulse rather than a continuous term). Short-term *IR*s are generally

categorized by age, sex, and intensity of activity (e.g., resting/napping, sedentary, and light, moderate, and high intensity; Adams, 1993; US EPA, 2011). Some studies are as specific as to provide activity-level-specific, short-term *IR*s for activities conducted in the indoor environment (US EPA, 2011).

In order to use short-term *IR*s in estimates of $iF_{\mathrm{in}\rightarrow\mathrm{in}}$, information regarding the fraction of time spent at various activity levels is needed. As is discussed in more detail below, time-activity patterns have been documented for populations from a wide range of geographic regions (e.g., Klepies et al., 2001; Statistics Canada, 2011; Jang et al., 2014b; ExpoFacts [http://expofacts.jrc.ec.europa.eu/]; Australian Centre for Human Health Risk Assessment, 2012) (Figure 1b). US EPA (2011) also provides age-specific estimates of time spent at various levels of activity intensity. The populations for which short-term *IR*s have been quantified are limited (US EPA, 2011; Jang et al., 2014b). Time-activity datasets can be combined with available short-term *IR* to predict *IR* distributions for populations for which such measurements are not available; however, it must be acknowledged that there is greater uncertainty in these values. Sensitivity analyses may be valuable for evaluating the influence of this uncertainty in $iF_{\mathrm{in}\rightarrow\mathrm{in}}$. Several exposure factor reports detail population demographics and physiological conditions, which can then be used to generate population-specific long- and short-term *IR* distributions from available measurements (Phillips and Moya, 2014 and references therein). Figure 2b summarizes the results of key *IR* studies, with detailed data provided in the SI. Overall, average *IR*s for children, adults, and all age groups for the data gathered here are slightly higher than that provided in the US EPA EFH (0.97, 1.20, and 1.09 $m^3\ h^{-1}$ versus 0.81, 1.04, and 0.92 $m^3\ h^{-1}$). Median values (and 95% CI) of the data provided herein for *IR*s for children, adults, and all age groups are 0.55 (0.17, 3.40), 0.70 (0.26, 4.47), and 0.66 (0.22, 4.23) $m^3\ h^{-1}$, respectively.

Time-Activity Patterns

In addition to serving as a predictor of activity intensity and *IR*, time-activity data provide valuable information regarding the time spent indoors and in various indoor locations. For a given subject, the cumulative intake of $PM_{2.5}$ is a function of the time spent by that subject in various microenvironments (e.g., indoor locations) and the $PM_{2.5}$ concentration profiles he or she is exposed to in each of those microenvironments. Thus,

411 the characterization of activity patterns is crucial to estimating $iF_{\text{in}\rightarrow\text{in}}$. Studies
412 characterizing time-activity patterns generally utilize diaries in which a representative
413 sample of individuals from the general population record their activities over a 24 or 48
414 hour period. The Center for Time Use Study at the University of Oxford provides a
415 database of time-activity diary studies for approximately 100 countries in Africa, Asia,
416 Australia, Europe, North America, and South America (Fisher and Tucker, 2013). Data
417 from multiple nations are harmonized to allow for comparison across countries. In
418 addition to references and links for the studies, where available, this database provides
419 important information such as temporal scale of the study, sampling and data-collection
420 methodology, sample size, and response rates. Some studies provide broader information
421 that is useful for long-term exposure studies (e.g., total time spent indoors and time spent
422 in the residence; Figures 1c and 2c), while others provide more detailed data, including
423 time spent in various types of indoor environments (e.g., home, school, retail stores, etc.),
424 time spent in different rooms within a residence, and time spent engaged in activities of
425 relevance to specific $PM_{2.5}$ emissions sources (e.g., cleaning, cooking; Schweizer et al.,
426 2007; Zhao et al., 2009; US EPA, 2011; Jang et al., 2014b; Matz et al., 2014). Such
427 studies have demonstrated that time-activity patterns vary with age, gender, location of
428 residence (e.g., urban versus rural), and various demographic and socioeconomic factors.
429 Time-activity data are generally categorized by these factors and, thus, activity patterns
430 can be estimated for a population of interest when demographic information is known.
431 For the U.S., the Consolidated Human Activity Database (CHAD;
432 http://www.epa.gov/heasd/chad.html) brings together data from various studies, resulting
433 in several thousand daily diaries that can be used in exposure simulation studies. The
434 advantage of CHAD over other time-use databases is that it is developed specifically for
435 exposure studies and certain parameters, such as time spent in indoor microenvironments,
436 can be more easily distinguished. The Stochastic Human Exposure and Dose Simulation
437 (SHEDS) Model (Burke et al., 2001), for example, simulates a population representative
438 of the study populations, as well as their activity patterns, by sampling from input
439 demographic data and CHAD.

Occupancy

Also key to determining $iF_{\text{in}\rightarrow\text{in}}$ is knowledge regarding the total number of people occupying a space influenced by indoor $PM_{2.5}$ emissions (Nazaroff, 2008). Higher occupancy means a larger number of people in proximity to indoor sources and, thus, a higher population $iF_{\text{in}\rightarrow\text{in}}$. Several studies provide information regarding household size and composition, which can be utilized to estimate residential occupancy in calculations of $iF_{\text{in}\rightarrow\text{in}}$ (Figure 1f). The U.S. Census Bureau (USCB), for example, provides information regarding the number and percentage of homes with household sizes ranging from one person to seven or more people, as well as demographic data describing households of varying sizes (USCB, 2010; Vespa et al., 2013). Similar information is available for the European Union (EU) and individual EU nations from Eurostat (2014). Bongaarts et al. (2001) presented household size and composition for the developing countries based on surveys conducted in forty-three nations in the 1990s, but notes that household-size dynamics can change with increased urbanization and industrialization, trending toward smaller household sizes (i.e., trending toward the nuclear family). That study provided data regarding household size and the demographic characteristics of home occupants for four regions: Asia, Latin America, Near East/North Africa, and Sub-Saharan Africa (see SI). Drivers of within- and between-nation/region variability are discussed and include level of development (e.g., gross national product) and residence in urban versus rural areas. The United Nations Demographic Yearbook is a valuable reference for identifying and locating household occupancy and characteristic data collected through national censuses (United Nations, 2013). For non-residential buildings, US EPA (2011) provides distributions of employee numbers for commercial buildings with a wide range of uses (SI).

Pollutant-Specific Factors

Concentrations of $PM_{2.5}$ and related intake in a given indoor environment or zone within an indoor environment depend on source emissions rates (S_{in}), as well as the removal mechanisms acting on the particles (k_{in}) (Table 1, Equation 2). Such removal mechanisms include the ventilation and transport processes discussed above, particle

deposition, filtration in HVAC-system filters and air cleaners, and, in some cases, chemical transformations/phase changes (Nazaroff, 2004). AERs and ventilation rates can be estimated using the data discussed above. In the following paragraphs, we discuss the data and tools available to take into account other factors influencing indoor $PM_{2.5}$ concentrations and $iF_{\mathrm{in}\rightarrow\mathrm{in}}$, with a primary focus on $PM_{2.5}$ emitted directly from indoor sources.

Indoor $PM_{2.5}$ Emissions

Multiple studies have characterized total $PM_{2.5}$ emissions from common indoor sources and activities such as cooking, cleaning, smoking, use of various home and office appliances, candles, incense, and insect repellent coils (Figure 1e) (e.g., Jetter et al., 2002; Liu et al., 2003; Lung and Hu, 2003; Guo et al., 2004; He et al., 2004; Lee and Wang, 2004; Afshari et al., 2005; Olson and Burke, 2006; He et al., 2007; Evans et al., 2008; See and Balasubramanian, 2011;Torkmahalleh et al., 2012). Substantial variability in $PM_{2.5}$ emission rates has been observed within and across sources (Figures 2e – g). For example, cooking activities can lead to emission rates as high as 467 mg min^{-1} (Olson and Burke, 2006), while emissions from printers were reported to be 2.8×10^{-4} mg min^{-1} (He et al., 2007). He et al. (2004) observed a median emission rate of 2.7 mg min^{-1} for frying food, while Olson and Burke (2006) reported a value of 6 mg min^{-1}. Emission rates for cooking activities vary with the cooking method (e.g., frying, grilling, baking), with the type of food or oils used in the cooking process (He et al., 2004; Olson and Burke, 2006; Torkmahalleh et al., 2012), and with stove type and the source of fuel (e.g., biomass, coal, gas, electric) (SI) (Jetter and Kariher, 2009; Jetter et al., 2012). The importance of a given source in terms of its contribution to $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ varies with a variety of factors including the indoor environment under consideration, occupant activities, and time of day or season. For example, in office environments, appliances (e.g., printers, copy machines) may contribute substantially to indoor $PM_{2.5}$ concentrations, while cooking, a major source in residential environments, is unlikely to be of importance. On the other hand, cleaning products are likely to be significant sources of $PM_{2.5}$ in both office and residential environments.

The influence of specific $PM_{2.5}$ sources on $iF_{in \rightarrow in}$ also varies geographically. Solid fuel combustion, for example, is a particularly important source of indoor $PM_{2.5}$ emissions in the developing world. As noted above, the effects of indoor exposures to solid fuel combustion emissions are a major global environmental health concern (www.who.int/indoorair/en). As a result, controlled laboratory studies and field measurements have been undertaken to characterize $PM_{2.5}$ emissions from various cook stoves and fuel sources (Habib et al., 2008; Edwards et al., 2014 and references therein). It is important to note, however, that there is evidence that emissions rates measured in a laboratory setting differ from those in the field (Edwards et al., 2014) and future efforts are more focused on characterizing emissions in actual household settings. In addition to emissions, data regarding the percentage of households using solid fuels and geographic differences in fuel and stove use are available for estimating $iF_{in \rightarrow in}$ associated with solid fuel use (Rehfuess et al., 2006; Bonjour et al., 2013; www.who.int/indoorair/health_impacts/he_database/en; see SI).

As is discussed in more detail below, particle loss rates vary with particle size and, thus, information regarding the size distributions of particles emitted from specific sources is useful for calculating $iF_{in \rightarrow in}$. Recent work has provided particle size distributions and/or size-resolved emissions rates for a range of common indoor activities or sources including cooking (Li and Hopke, 1993; Abt et al., 2000; Long et al., 2000; Wallace et al., 2004; Hussein et al., 2006; Ogueli et al. 2006; Wallace, 2006), cleaning (Kleeman et al., 1999; Abt et al., 2000; Long et al., 2000; Ogueli et al. 2006; Gehin et al., 2008), candles, incense, and aroma lamps (Li and Hopke, 1993; Kleeman et al., 1999; Hussein et al., 2006; Wallace, 2006; Gehin et al., 2008), smoking (Li and Hopke, 1993; Nazaroff, 2004; Hussein et al., 2006;), cook-stove use in developing countries and residential wood combustion (Kleeman et al., 1999; Hays et al., 2003; Armendriz-Arnez et al., 2010; Shen et al., 2011), fuel-combustion lamps and appliances (Wallace, 2006; Apple et al., 2010), personal care products/appliances (e.g., hairspray, blow dryer) (Hussein et al., 2006), and printers (Gehin et al., 2008; Wang et al., 2012; Stephens et al., 2013).

Particle Losses: Deposition

Particle deposition describes all particle losses driven by Brownian diffusion, gravitational settling, interception, and impaction. Brownian diffusion dominates particle losses for particles with diameters smaller than about 0.1 μm (ultrafine particles [UFP]), while for larger particles, interception, impaction, and gravitational settling are the dominant loss processes (Finlayson-Pitts and Pitts, 2000). As a result, deposition loss rate coefficients (k_{dep} [h^{-1}]) vary with particle size (Ozkaynak et al., 1997; Long et al., 2001; Riley et al., 2002; Nazaroff, 2004; Hering et al., 2007). Multiple studies have measured particle-size resolved values of k_{dep} or indoor particle decay rates (i.e., the sum of all loss mechanisms) (e.g., Thatcher and Layton, 1995; Ozkaynak et al., 1997; Abt et al., 2000; Long et al., 2001; Howard-Reed et al., 2003; Thatcher et al., 2003; Ferro et al., 2004; He et al., 2005; Sarnat et al., 2006; Meng et al., 2007; Stephens and Siegel, 2013). These studies have been conducted under a range of sampling and building ventilation conditions. In addition to their particle size dependence, k_{dep} values vary with airflow conditions and indoor environment surface-to-volume ratios driven by the presence of furnishings and carpets (Lai, 2002; Thatcher et al., 2002; Howard-Reed et al., 2003; Nazaroff, 2004). For example, Thatcher et al. (2002) demonstrated that k_{dep} could vary by as much as a factor of 2.6 across different surface-to-volume (i.e., room-furnishing) scenarios and by as much as a factor of 2.4 with different values of airflow speed. Zhang et al. (2014) brings attention to the fact that variability in k_{dep} to surfaces with varying orientations (e.g., horizontal versus vertical surfaces) can influence indoor $PM_{2.5}$ concentrations and $iF_{\mathrm{in}\rightarrow\mathrm{in}}$. That study provides vertical- and horizontal-surface deposition rates for particles in two broad $PM_{2.5}$ size classes.

Measurements conducted under various conditions have been combined and fit with a polynomial regression that describes k_{dep} as a function of particle size (Riley et al., 2002; Nazaroff, 2004). This fit does not take into account variability with ventilation conditions, room turbulence, surface-to-volume ratios, or room surface orientations; however, Hodas et al. (2014) found that indoor concentrations of ambient $PM_{2.5}$ modeled using k_{dep} values selected with this regression curve were well-correlated with measured indoor $PM_{2.5}$. El Orch et al. (2014) combined measurement data from multiple studies to

predict particle-size-resolved k_{dep} values, fit a curve describing k_{dep} as a function of particle diameter, and developed a method to account for increased indoor airflow speeds when windows are open. In those circumstances, values of k_{dep} selected from curves describing depositional loss rates as a function of particle size (e.g., using Monte Carlo methods to sample from a particle size distribution) can be multiplied by 1.7 for windows open a large amount and by 1.23 when windows are open a small amount. In addition, a small number of studies have quantified deposition or decay rates for total $PM_{2.5}$ (Figures 1d, 2d) (Ozkaynak et al., 1997; He et al., 2005; Olson and Burke, 2006; Wallace et al., 2013). Such information can be useful in circumstances in which particle size distribution data are not available.

Particle Losses: Filtration

For homes with HVAC systems, particle losses will also be related to HVAC system recirculation rates and filter removal efficiencies. Several studies have measured size-resolved particle filtration efficiencies for various filters commonly found in residential and commercial HVAC systems (Hanley et al., 1994; Stephens et al., 2011; Stephens and Siegel, 2012b, 2013; Azimi et al., 2014). Stephens et al. (2011) also studied recirculation rates in residential and light-commercial HVAC systems. El Orch et al. (2014) extended this type of analysis to provide size-resolved filtration efficiencies for five classifications of filters, as well as estimates of the prevalence of these filter categories in homes. Waring and Siegel (2008) and Stephens and Siegel (2013) considered the influence of not only filtration, but also losses to heat exchangers and ducts within HVAC systems. Similarly, Sippola and Nazaroff (2002) reviewed studies of particle deposition in HVAC system ducts. Such losses are likely to be of particular importance in schools and commercial buildings. Filtration and fractional loss curves generated from such measurements have been used in many studies to estimate particle removal efficiencies as a function of particle size (Riley et al., 2002; Hodas et al., 2012, 2014).

HVAC-system air recirculation rates are also key parameters in characterizing filtration rates. Recommended values for HVAC recirculation rates in residences (El Orch et al., 2014; Stephens et al., 2011; Stephens, 2015) and in non-

592 residential buildings (Sundell et al., 1994; Weschler et al., 1996; Zuraimi et al., 2007 and
593 references therein; Fadeyi et al., 2009) are available from a limited number of studies.
594 Note also that the fraction of air that is recirculated in HVAC systems displays large
595 spatial variability. Zuraimi et al. (2007), for example, state that 90% of air in conditioned
596 office buildings in the U.S. and Singapore is recirculated. In some countries (e.g.,
597 Denmark and Germany), however, all mechanical ventilation systems must be single-pass
598 (i.e., no air is recirculated). Similarly, HVAC system runtimes directly govern whether or
599 not a system is in operation and filtering particles at a given point in time, but like
600 recirculation rates, measurements are limited (Thornburg et al., 2004; Stephens et al.,
601 2011).

602 The prevalence of central air and heating systems is commonly documented in
603 housing and energy surveys. US EPA (2011), for example, provides information
604 regarding the prevalence of central heating and cooling systems in residential and
605 commercial buildings. It is important to note, however, that the prevalence of central and
606 recirculating HVAC systems is highly variable both within and across nations and
607 geographic regions. The importance of collecting data regarding the heating and cooling
608 systems (or lack thereof) present in households on a global scale has recently been
609 highlighted (United Nations, 2008).

610

611 **Particle Resuspension**

612 The resuspension of particles that have deposited on surfaces in indoor
613 environments can also influence indoor $PM_{2.5}$ concentrations and $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ (Ferro et al.,
614 2004; Lioy, 2006, and references therein). While typically considered to be an important
615 determinant of exposures to particles larger than $PM_{2.5}$, Ferro et al. (2004) found that
616 resuspension can result in the equivalent of a $PM_{2.5}$ source strength ranging from 0.03 to
617 0.5 mg min^{-1}. The prevalence and magnitude of resuspension are dependent on the
618 activities of building occupants, specifically cleaning (e.g., dusting, vacuuming) and
619 active movement (e.g., walking, dancing, playing) (Ferro et al., 2004; Lioy, 2006). Thus,
620 the influence of resuspension on $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ is expected to vary temporally and spatially.

621

Transformation: Phase Changes and Indoor Chemistry

Phase changes and chemical transformation can lead to both increases and decreases in indoor $PM_{2.5}$ concentrations. The partitioning of semivolatile organic compounds (SVOCs) between the gas and particle phases, for example, is dependent on indoor air temperature and the availability of particle-phase organic matter for sorption (Pankow, 1994). Thus, the extent to which a given indoor source of SVOCs contributes to $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ will depend on the fraction of emissions from that source found in the particle phase, which, in turn, is dependent on the conditions of the indoor environment (i.e., temperature, organic $PM_{2.5}$ concentrations). Examples of indoor sources of SVOCs that display this behavior include environmental tobacco smoke, flame retardants, plasticizers, and pesticides (Liang and Pankow, 1996; Gurunathan et al., 1998; Bennett and Furtaw, 2004; Lioy, 2006; Weschler and Nazaroff, 2008 and references therein). Estimating shifts in partitioning requires knowledge regarding volatility and partitioning coefficients of chemical species commonly found indoors, as well as the development of simplified models to predict SVOC partitioning in indoor air. This is an active area of research (Weschler and Nazaroff, 2008, 2010; Weschler, 2011; Hodas and Turpin, 2014; Liu et al., 2014); however, further work is needed to characterize semi-volatile species of indoor origin before this process can be consistently incorporated into estimates of $iF_{\mathrm{in}\rightarrow\mathrm{in}}$.

The formation of secondary organic aerosols (SOA) from reactions between oxidants and gas-phase compounds emitted indoors can also substantially influence $PM_{2.5}$ concentrations and $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ (Weschler and Shields, 1999; Long et al., 2000; Wainman et al., 2000; Weschler, 2006, 2011; Waring and Siegel, 2010, 2013; Waring et al., 2011; Waring, 2014). Most work in this area has focused on reactions between terpenoids emitted from air fresheners, cleaning products, and scented personal care products and ozone (Nazaroff and Weschler, 2004; Singer et al., 2006; Weschler, 2006; Waring et al., 2011; Weschler, 2011; Waring and Siegel, 2010, 2013). Such studies have demonstrated that indoor SOA formation varies with multiple factors including the chemicals present in indoor air, relative humidity, time of day, season, indoor ventilation conditions and HVAC system use, indoor surface area and surface materials, and geographic location (Waring and Siegel, 2010; Weschler, 2011; Waring and Siegel, 2013; Youseffi and Waring, 2014). Indoor sources of ozone include photocopiers, laser printers, and

electrostatic air cleaners; however, the majority of ozone present indoors is the result of transport from the outdoor environment (Weschler, 2000). SOA generated through reactions between VOCs of indoor origin and ozone of outdoor origin illustrates one mechanism through which interactions between indoor- and outdoor-generated pollutants can influence the intake of $PM_{2.5}$ attributable, at least in part, to indoor sources. This complication of separating outdoor- and indoor-source contributions to the intake of $PM_{2.5}$ in indoor environments is discussed further in the next section.

Influence of outdoor-generated pollutants on cumulative indoor intake of $PM_{2.5}$

The cumulative intake of $PM_{2.5}$ that occurs indoors is influenced by both indoor and outdoor $PM_{2.5}$ sources (Table 1, Equation 2) and depends on (1) primary emissions of $PM_{2.5}$ from indoor sources, (2) the formation of secondary $PM_{2.5}$ from precursors of indoor origin, (3) the transport of outdoor-generated $PM_{2.5}$ into the indoor environment, and (4) interactions between pollutants of indoor and outdoor origin. This latter factor includes SOA formation through reactions of indoor-emitted volatile organic compounds (VOCs) and outdoor-generated oxidants, as well as the partitioning of outdoor-generated gas-phase SVOCs to particulate matter of indoor origin and/or the partitioning of gas-phase SVOCs emitted by indoor sources to outdoor-generated particles that have infiltrated indoors. Prior sections focused on factors (1) and (2). Below, we briefly explore the current state of knowledge regarding interactions between pollutants of outdoor and indoor origin and the influence of outdoor $PM_{2.5}$ sources on cumulative indoor intake.

Outdoor-generated $PM_{2.5}$ (ambient $PM_{2.5}$) that penetrates into and persists in the indoor environment is a major source of indoor $PM_{2.5}$. Multiple studies have quantified the fraction of ambient $PM_{2.5}$ found in indoor air ($f_{\mathrm{out}\rightarrow\mathrm{in}}$) (Chen and Zhao, 2011 and references therein; Diapouli et al., 2013 and references therein). These studies have demonstrated that there is substantial between- and within-home variability in $f_{\mathrm{out}\rightarrow\mathrm{in}}$ (Ozkaynak et al., 1997; Ott et al., 2000; Meng et al., 2005; Weisel et al., 2005; Polidori et al., 2006; Allen et al., 2012; MacNeil et al., 2012; Hänninen et al., 2013; Kearny et al., 2014), illustrating the difficulty in utilizing measured values of $f_{\mathrm{out}\rightarrow\mathrm{in}}$ to estimate contributions of ambient $PM_{2.5}$ to cumulative indoor intake. In addition, most studies are

limited in their geographic and temporal scope and cannot be generalized to a broader population of homes. Two exceptions are the studies conducted by Hänninen et al. (2011) and El Orch et al. (2014). Estimates of $f_{\mathrm{out}\rightarrow\mathrm{in}}$ for homes in ten European countries sampled as part of six studies were aggregated and summary statistics of $f_{\mathrm{out}\rightarrow\mathrm{in}}$ were provided for various climatic regions of Europe (Northern, Central, and Southern Europe) and by season (Hänninen et al. 2011). El Orch et al. (2014) conducted a detailed modeling study in which particle-size-resolved distributions of $f_{\mathrm{out}\rightarrow\mathrm{in}}$ for single-family homes in the U.S. were calculated.

For a given exposure scenario, $f_{\mathrm{out}\rightarrow\mathrm{in}}$ can also be calculated using a mass balance model in which indoor ambient $PM_{2.5}$ concentrations are described as function of AER, the efficiency with which particles penetrate across the building envelope, particle deposition, filtration in HVAC-system filters and air cleaners, and, for semivolatile species, phase changes in indoor air (e.g., Hering et al., 2007; Hodas et al., 2012, 2014). Similarly, these physical and chemical processes also govern the outdoor transport of indoor-generated $PM_{2.5}$ and, thus, $iF_{\mathrm{in}\rightarrow\mathrm{out}}$ and $iF_{\mathrm{in,total}}$ (see Table 1). While the contributions of $iF_{\mathrm{in}\rightarrow\mathrm{out}}$ to $iF_{\mathrm{in,total}}$ are typically negligible compared to that of $iF_{\mathrm{in}\rightarrow\mathrm{in}}$, there is evidence that solid fuel combustion in household cook stoves can contribute substantially to ambient $PM_{2.5}$ concentrations in some regions (e.g., India, China) (Chafe et al., 2014).

The data given above provide inputs to predict AER, deposition, and filtration. Chen and Zhao (2011) provide a detailed review of penetration efficiency measurements and modeling strategies. While the focus of previous work has mostly been on the penetration of ambient $PM_{2.5}$ into the indoor environments, results of these studies can also be used to estimate penetration of indoor-generated particles between separated indoor zones/rooms. Tools are also available to account for evaporative losses of ammonium nitrate (Lunden et al., 2003; Hering et al., 2007), and the development of modeling tools to predict the gas-particle partitioning of SVOCs (of both indoor and outdoor origin) in indoor air is an active area of ongoing research (Weschler and Nazaroff, 2008, 2010; Weschler, 2011; Hodas and Turpin, 2014; Liu et al., 2014).

Because the availability of organic matter for sorption influences the gas-particle partitioning of SVOCs, there is the potential for the indoor formation of particles that are

only present due to interactions between SVOCs of indoor and outdoor origin. For example, gas-phase SVOCs emitted indoors can sorb to indoor particulate matter of outdoor origin that has penetrated into the home (Lioy, 2006; Weschler and Nazaroff, 2008). Similarly, incoming organics from outdoors can shift from the gas phase toward the particle phase as they sorb to particulate organic matter emitted by indoor sources (Naumova et al., 2003; Polidori et al., 2006; Weschler and Nazaroff, 2008; Shi and Zhao, 2012; Hodas and Turpin, 2014). The result is the formation of $PM_{2.5}$ that is in part, but not fully, attributable to indoor sources. Such interactions between pollutants of indoor and outdoor origin highlight the difficulty in fully separating the contributions of indoor and outdoor $PM_{2.5}$ sources to the intake of $PM_{2.5}$.

The formation of SOA from reactions between indoor-generated VOCs and oxidants (e.g., ozone) of outdoor origin is another example of the ways in which outdoor-generated pollutants can influence the intake of $PM_{2.5}$ associated with indoor sources. Contributions of secondary particulate matter derived from well-characterized inorganic systems to outdoor *iF* have previously been accounted for using chemical transport models (e.g., Levy et al., 2003; Greco et al., 2007). The data and modeling tools available to include indoor secondary particulate matter (specifically, SOA) formation in estimates of indoor $PM_{2.5}$ exposures continue to improve. Waring (2014) presented a mechanistic model to calculate time-averaged indoor SOA concentrations formed as a result of the oxidation of reactive organic gases by ozone and the hydroxyl radical. Distributions of model inputs for 66 reactive organic gases relevant to the indoor environment (Weisel et al., 2005; Turpin et al., 2007) are provided in that work. In addition, a linear regression model describing SOA concentrations as a function of AER, indoor concentrations of outdoor-generated ozone and organic aerosols, indoor organic aerosol emission rates, particle and ozone deposition rates, temperature, and emission rates of reactive organic gases described the majority of variability in SOA concentrations calculated using the more complex mechanistic SOA model described above ($R^2 = 0.88$; Waring, 2014). Ji and Zhao (2015) demonstrated that the extent to which indoor SOA formation impacts indoor concentrations of $PM_{2.5}$ varies geographically, with SOA comprising 6 to 30% of indoor $PM_{2.5}$ mass for the U.S. homes included in the Waring (2014) study, but less than 3% of $PM_{2.5}$ mass for homes in Beijing. Accounting for SOA formation indoors is an

active and quickly advancing area of research and is crucial for ensuring that the full impact of specific products, activities, and processes can be taken into account in LCIA.

Discussion

Applications in Life Cycle Impact Assessment

The data provided in this review constitute a first step in addressing key questions and current challenges previously identified for the incorporation of health effects associated with indoor $PM_{2.5}$ emissions into LCIA (Hellweg et al., 2009; Fantke et al., 2015; Humbert et al., 2015). Specifically, this review allows for the characterization of a range of exposure-scenario archetypes, both in terms of indoor setting (e.g., residence, office) and in geographic location, aids in the identification of the major factors influencing $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ and potential spatial and temporal variability in the importance of these key factors, and allows for the assessment of the level of detail and scope needed when developing exposure-scenario archetypes for use in LCIA.

In an ongoing effort, the UNEP-SETAC task force on $PM_{2.5}$ health effects will utilize the data provided in this review to build a quantitative assessment framework for consistently combining and evaluating indoor and outdoor intake fractions from $PM_{2.5}$ sources for application in LCIA. Complementary work is currently focusing on (1) conducting a quantitative assessment of potential variability in $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ (e.g., across exposure scenarios and geographic regions), as well as the sensitivity of calculations of $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ to heterogeneity in the input parameters reviewed here, (2) the evaluation of state-of-the-art modeling tools available to predict indoor and outdoor intake fractions in the context of suitability for use in LCIA, and (3) the consistent incorporation of various shapes of ERFs (Fantke et al., 2015). Together, these efforts will aid in the development of a standardized methodology by which to estimate exposures and will contribute to the effort to include $PM_{2.5}$-related health effects in LCIA.

Key to assessing $PM_{2.5}$-related health effects over the life cycle of products is the ability to evaluate the range of potential human exposure associated with a given particle emissions source. Previous work has illustrated the potential magnitude of spatial and temporal variability in $iF_{\mathrm{in}\rightarrow\mathrm{in}}$. Humbert et al. (2011), for example, estimates that typical values of $iF_{\mathrm{in}\rightarrow\mathrm{in}}$range between approximately10^{-3} and 10^{-2} kg intake at the population

scale per kg emitted indoors. Klepeis and Nazaroff (2006) found that $iF_{\text{in}\rightarrow\text{in}}$ for environmental tobacco smoke varied between 6.6×10^{-4} and 2.6×10^{-3} kg intake per kg emitted within a single simulated home depending on multiple factors including home ventilation conditions and occupant activity patterns. Thus, while a single recommended value meant to characterize a needed modelling parameter is valuable for providing an estimate of the magnitude of $iF_{\text{in}\rightarrow\text{in}}$ (e.g., a single AER value meant to represent typical housing the U.S.), distributions or ranges describing these input parameters are crucial. Such distributions allow for the evaluation of the central tendencies of $iF_{\text{in}\rightarrow\text{in}}$, as well as the extremes, thereby acknowledging the variability in population exposure patterns, housing aspects, and indoor air chemistry. By aggregating the results of multiple studies, the present review provides a broader picture of the range of potential values for a given parameter influencing indoor concentrations of $PM_{2.5}$ and allows for the consideration of a range of archetypal indoor environments. It is important to note that these values vary temporally and spatially with multiple factors, as discussed in the individual sections above, and parameters are not available to describe all exposure scenarios and geographic regions. Thus, understanding the full range of input parameters also allows for the consideration of uncertainty in $iF_{\text{in}\rightarrow\text{in}}$ for $PM_{2.5}$.

Depending on the design of the selected modelling framework, not all of the factors potentially contributing to variability in $iF_{\text{in}\rightarrow\text{in}}$ will necessarily be considered in LCIA. For example, Hellweg et al. (2009) suggested that the representation of the indoor environment as a single, well-mixed compartment provides the most effective way to incorporate indoor $PM_{2.5}$ exposures into LCIA. On the other hand, in regards to assessing exposure to individual VOCs from cleaning products, Earnest and Corsi (2013) propose the use of a two-zone model in which the near-person/near-source region and the rest of the indoor environment are treated as discrete zones. LCIA often follows approaches based on archetypes to account for differences in exposure scenarios or geographic regions. Thus, the parameters that will be of the greatest importance are those that account for geographic variability in more general housing and building characteristics (e.g., volume, whole-building air exchange and ventilation), indoor-environment occupancy, and the prevalence of specific indoor sources (e.g., cooking and heating appliances). Parameters that provide a higher level of detail (e.g., activity-specific

808 breathing rates, local-scale flows), however, will be valuable to higher tier assessments of
809 indoor air quality and epidemiologic studies that aim to characterize indoor $PM_{2.5}$
810 exposures for specific conditions in a well-characterized environment.
811
812 **Remaining Limitations and Recommendations for Future Research**
813 One contributor to limitations in the availability and scope of data like those
814 reviewed here is the fact that the studies carried out to collect the data are expensive and
815 work intensive. As a result, they tend to be carried out in infrequent, intensive campaigns.
816 As noted above, for example, many AER studies are not representative of the full range
817 of housing stock, even for the nations or cities in which they were carried out. Values are
818 more limited or non-existent in some developing countries and are biased towards U.S.
819 and European studies. We suggest that there is a need for studies on AER in developing
820 countries, particularly in rural regions where biomass is used for cooking in homes.
821 Another issue constraining the representativeness of the data is the potential for
822 changes with time. While some values are not expected to vary temporally (i.e., *IR*,
823 although the activity levels driving them may change), others change on timescales faster
824 than the studies characterizing them are carried out. Bongaarts et al. (2001), for example,
825 noted the tendency for household size to converge towards the nuclear family in rapidly
826 industrializing and urbanizing regions. Similarly, there is the potential for changes in
827 human activity patterns with increased access to media, suggesting a need for updated
828 human activity pattern data. Housing construction practices change with advancing
829 technology and materials development, as well as with recent pushes toward energy
830 efficiency. Urban growth (e.g., Seto and Fragikas, 2005; Xiao et al., 2006; Schneider and
831 Woodcock, 2008) may make the lack of data characterizing AERs in apartments and
832 multi-family residences a major issue in both developing and developed countries. New
833 techniques utilizing 3D imaging sensors to evaluate building/room size and leakage
834 characteristics show promise in increasing data availability for leaky buildings (e.g., in
835 developing countries), airtight, energy efficient buildings, and multifamily residences
836 (Gong and Caldas, 2008) and should be a consideration in future work in this area.
837 Finally, while the principles driving pollutant dynamics will not change with time,
838 emission rates, particle size distributions, and particle composition may change with

technology. Cynthia et al. (2007), for example, reported a 35% decrease in $PM_{2.5}$ exposures with the introduction of a higher-efficiency cook stove in an intervention study in rural Mexico. As a result of these ever-changing factors, a continued effort to undertake such studies and to expand their temporal and spatial scope is key to ensuring that the impacts associated with specific products and emission sources can be fully assessed in the context of LCIA.

We also recommend that future efforts focus on a number of key research areas. First, there is a need for a more widespread and detailed characterization of inter- and intra-zonal airflows and the factors that influence them for a range of residence types, commercial buildings, and occupational settings to derive useful information for higher tier assessments of indoor air quality. Such characterizations would be useful in addressing proximity-to-source issues. Of particular importance may be the development of a set of archetypal building layouts that describe a range of building types, so that these highly variable flows can be modelled for a given exposure scenario with tools such as COMIS and CONTAM. For applications in LCIA, a simple two-zone model might be more suitable as more complex approaches might lack data and consistency across indoor and outdoor emission situations. As noted above, there are large geographic differences in the heating and cooling systems present in households and other indoor environments on a global scale. Documenting these differences and the related impacts on indoor particle dynamics is an important area of future work. Finally, there is a need for more research aimed at obtaining a thorough understanding of interactions between indoor- and outdoor-generated pollutants and the formation of SOA in indoor air. Key to this is the development of accurate simplified models that can easily be applied in LCIA. The regression model developed by Waring (2014) to predict indoor SOA formation based on a small number of key parameters provides an example of the type of modeling tools that will advance predictions of $iF_{\mathrm{in}\rightarrow\mathrm{in}}$ for $PM_{2.5}$ in this context.

CONCLUSIONS

The present paper reviews and compiles the results of studies exploring the main factors influencing indoor $PM_{2.5}$ concentrations and associated $iF_{\mathrm{in}\rightarrow\mathrm{in}}$, with an emphasis on primary indoor $PM_{2.5}$ emissions. Specifically, we focus on factors related to building

characteristics, occupant characteristics and behaviors, and pollutant properties and dynamics. The key studies and data sources discussed herein comprise a tool kit of exposure-modelling parameters that can be used to estimate the central tendencies and potential ranges of $iF_{\mathrm{in}\rightarrow\mathrm{in}}$. A follow-up effort will utilize the data provided in the present review to build a framework to consistently integrate indoor and outdoor exposures to $PM_{2.5}$ emitted by indoor and outdoor sources. Combined, the present review and the follow-up work contribute to the effort to consistently include $PM_{2.5}$-derived health effects in LCIA. Continued efforts to characterize the factors influencing indoor $PM_{2.5}$ concentrations will ensure that impacts associated with specific products and emission sources can be fully assessed in LCIA and other comparative human exposure and impact assessment frameworks.

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Emission **[$kg_{emitted}$/h]**	**Indoor intake** **[kg_{intake}/h]**	**Outdoor intake** **[kg_{intake}/h]**	**Total intake** **[kg_{intake}/h]**	**Intake fraction** **[kg_{intake}/h per $kg_{emitted}$/h]**
Indoor ($PM_{2.5}$ or precursor) emissions S_{in}	Indoor intake due to indoor emissions $\overbrace{\left(\frac{IR_{in} \times n_{in}}{V_{in} \times k_{in}}\right)}^{iF_{in \to in}} \times S_{in}$	Outdoor intake due to indoor emissions $\overbrace{\left(iF_{out,total} \times f_{in \to out}\right)}^{iF_{in \to out}} \times S_{in}$	Total intake due to indoor emissions $iF_{in,total} \times S_{in}$	Total intake due to indoor emissions per unit of indoor emissions $iF_{in,total} = \overbrace{\left(\frac{IR_{in} \times n_{in}}{V_{in} \times k_{in}}\right)}^{iF_{in \to in}} + \overbrace{\left(iF_{out,total} \times f_{in \to out}\right)}^{iF_{in \to out}}$ (eq. 1)

Outdoor ($PM_{2.5}$ or precursor) emissions S_{out}	Indoor intake due to outdoor emissions $\overbrace{(iF_{in,total} \times f_{out\rightarrow in})}^{iF_{out\rightarrow in}} \times S_{out}$	Outdoor intake due to outdoor emissions $\overbrace{\left(\frac{IR_{out} \times n_{out}}{V_{out} \times k_{out}}\right)}^{iF_{out\rightarrow out}} \times S_{out}$	Total intake due to outdoor emission $iF_{out,total} \times S_{out}$	Total intake due to outdoor emission per unit of outdoor emissions $iF_{out,total} = \overbrace{\left(\frac{IR_{out} \times n_{out}}{V_{out} \times k_{out}}\right)}^{iF_{out\rightarrow out}} + \overbrace{(iF_{in,total} \times f_{out\rightarrow in})}^{iF_{out\rightarrow in}}$
	Cumulative indoor intake due to indoor and outdoor emissions $\overbrace{\left(\frac{IR_{in} \times n_{in}}{V_{in} \times k_{in}}\right)}^{iF_{in\rightarrow in}} \times S_{in} + \overbrace{(iF_{in,total} \times f_{out\rightarrow in})}^{iF_{out\rightarrow in}} \times S_{out}$ (eq. 2)	Cumulative outdoor intake due to indoor and outdoor emissions $iF_{out\rightarrow out} \times S_{out} + iF_{in\rightarrow out} \times S_{in}$	Cumulative intake due to indoor and outdoor emissions $iF_{in,total} \times S_{in} + iF_{out,total} \times S_{out}$	

Emission [$kg_{emitted}/h$]	**Indoor intake** [kg_{intake}/h]	**Outdoor intake** [kg_{intake}/h]	**Total intake** [kg_{intake}/h]	**Intake fraction** [kg_{intake}/h per $kg_{emitted}/h$]
Indoor ($PM_{2.5}$ or precursor) emissions S_{in}	Indoor intake due to indoor emissions $\overbrace{\left(\frac{IR_{in}\times n_{in}}{V_{in}\times k_{in}}\right)}^{iF_{in\to in}} \times S_{in}$	Outdoor intake due to indoor emissions $\overbrace{\left(iF_{out,total} \times f_{in\to out}\right)}^{iF_{in\to out}} \times S_{in}$	Total intake due to indoor emissions $iF_{in,total} \times S_{in}$	Total intake due to indoor emissions per unit of indoor emissions $iF_{in,total} = \overbrace{\left(\frac{IR_{in}\times n_{in}}{V_{in}\times k_{in}}\right)}^{iF_{in\to in}} + \overbrace{\left(iF_{out,total} \times f_{in\to out}\right)}^{iF_{in\to out}}$ (eq. 1)
Outdoor ($PM_{2.5}$ or precursor) emissions S_{out}	Indoor intake due to outdoor emissions $\overbrace{\left(iF_{in,total} \times f_{out\to in}\right)}^{iF_{out\to in}} \times S_{out}$	Outdoor intake due to outdoor emissions $\overbrace{\left(\frac{IR_{out}\times n_{out}}{V_{out}\times k_{out}}\right)}^{iF_{out\to out}} \times S_{out}$	Total intake due to outdoor emission $iF_{out,total} \times S_{out}$	Total intake due to outdoor emission per unit of outdoor emissions $iF_{out,total} = \frac{\overbrace{\left(\frac{IR_{out}\times n_{out}}{V_{out}\times k_{out}}\right)}^{iF_{out\to out}}}{} + \overbrace{\left(iF_{in,total} \times f_{out\to in}\right)}^{iF_{out\to in}}$
	Cumulative indoor intake due to indoor and outdoor emissions $\overbrace{\left(\frac{IR_{in}\times n_{in}}{V_{in}\times k_{in}}\right)}^{iF_{in\to in}} \times S_{in} + \overbrace{\left(iF_{in,total} \times f_{out\to in}\right)}^{iF_{out\to in}} \times S_{out}$	Cumulative outdoor intake due to indoor and outdoor emissions $iF_{out\to out} \times S_{out} +$	Cumulative intake due to indoor and outdoor emissions	

	(eq. 2)	$iF_{in\rightarrow out} \times S_{in}$	$iF_{in,total} \times S_{in} + iF_{out,total} \times S_{out}$	

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Emission **[$kg_{emitted}$/h]**	**Indoor intake** **[kg_{intake}/h]**	**Outdoor intake** **[kg_{intake}/h]**	**Total intake** **[kg_{intake}/h]**	**Intake fraction** **[kg_{intake}/h per $kg_{emitted}$/h]**
Indoor ($PM_{2.5}$ or precursor) emissions S_{in}	Indoor intake due to indoor emissions $\overbrace{\left(\frac{IR_{in}\times n_{in}}{V_{in}\times k_{in}}\right)}^{iF_{in\rightarrow in}} \times S_{in}$	Outdoor intake due to indoor emissions $\overbrace{\left(iF_{out,total} \times f_{in\rightarrow out}\right)}^{iF_{in\rightarrow out}} \times S_{in}$	Total intake due to indoor emissions $iF_{in,total} \times S_{in}$	Total intake due to indoor emissions per unit of indoor emissions $iF_{in,total} = \overbrace{\left(\frac{IR_{in}\times n_{in}}{V_{in}\times k_{in}}\right)}^{iF_{in\rightarrow in}} + \overbrace{\left(iF_{out,total} \times f_{in\rightarrow out}\right)}^{iF_{in\rightarrow out}}$ (eq. 1)
Outdoor ($PM_{2.5}$ or precursor) emissions S_{out}	Indoor intake due to outdoor emissions $\overbrace{\left(iF_{in,total} \times f_{out\rightarrow in}\right)}^{iF_{out\rightarrow in}} \times S_{out}$	Outdoor intake due to outdoor emissions $\overbrace{\left(\frac{IR_{out}\times n_{out}}{V_{out}\times k_{out}}\right)}^{iF_{out\rightarrow out}} \times S_{out}$	Total intake due to outdoor emission $iF_{out,total} \times S_{out}$	Total intake due to outdoor emission per unit of outdoor emissions $iF_{out,total} = \overbrace{\left(\frac{IR_{out}\times n_{out}}{V_{out}\times k_{out}}\right)}^{iF_{out\rightarrow out}} + \overbrace{\left(iF_{in,total} \times f_{out\rightarrow in}\right)}^{iF_{out\rightarrow in}}$

	Cumulative indoor intake due to indoor and outdoor emissions $\overbrace{\left(\frac{IR_{in}\times n_{in}}{V_{in}\times k_{in}}\right)}^{iF_{in\to in}} \times S_{in} + \overbrace{\left(iF_{in,total} \times f_{out\to in}\right)}^{iF_{out\to in}} \times S_{out}$ (eq. 2)	Cumulative outdoor intake due to indoor and outdoor emissions $iF_{out\to out} \times S_{out} + iF_{in\to out} \times S_{in}$	Cumulative intake due to indoor and outdoor emissions $iF_{in,total} \times S_{in} + iF_{out,total} \times S_{out}$

Table 1. Matrix illustrating the contributions of $PM_{2.5}$ derived from indoor and outdoor sources to indoor intake, outdoor intake, total intake, and intake fraction of $PM_{2.5}$. Aspects discussed in this paper are highlighted in grey and specific areas of focus are in red. Abbreviations: S_{in} or S_{out}, indoor or outdoor $PM_{2.5}$ source emission rate; $iF_{in\to in}$, fraction of $PM_{2.5}$ emitted/formed indoors that is taken in via inhalation indoors; $iF_{in\to out}$, fraction of $PM_{2.5}$ emitted/formed indoors that is transported outdoors and taken in via inhalation outdoors; $iF_{out\to out}$, fraction of $PM_{2.5}$ emitted/formed outdoors that is taken in via inhalation outdoors; $iF_{out\to in}$, fraction of $PM_{2.5}$ emitted/formed outdoors that is transported indoors and taken in via inhalation indoors; IR_{in} or IR_{out}, individual inhalation rate indoors or outdoors [$m^3_{inhaled}/h$]; n_{in} or n_{out}, number of exposed persons in an indoor or outdoor location; V_{in} or V_{out}, volume of indoor or outdoor location [m^3]; k_{in} or k_{out}, total indoor or outdoor particle removal rate attributable to all loss mechanisms (e.g., air exchange, particle deposition) [h^{-1}]; $iF_{in,total}$, total indoor inhalation intake fraction; $iF_{out,total}$, total outdoor inhalation intake fraction; $f_{in\to out}$, fraction of indoor-generated (emitted/formed) $PM_{2.5}$ transported outdoors, $f_{out\to in}$, fraction of outdoor-generated (emitted/formed) $PM_{2.5}$ transported indoors. Note that there is no cumulative intake fraction.

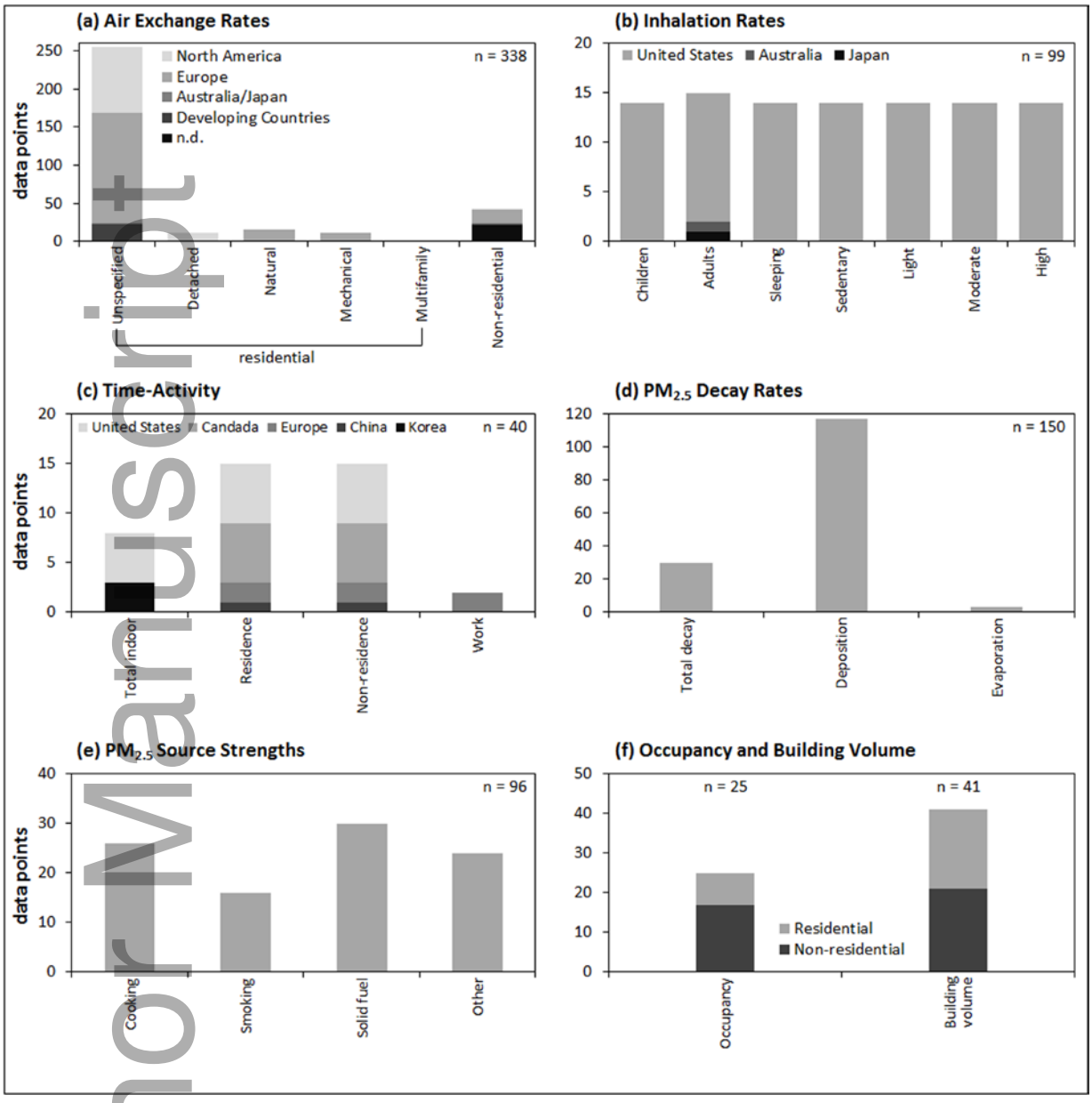


Figure 1. Frequency plot illustrating the number of data points (i.e., measured or modeled value or summary statistic from a distribution of measurements describing the parameter of interest) gathered from the literature for the primary factors influencing indoor inhalation intake fraction of $PM_{2.5}$: (a) air exchange rates, (b) inhalation rates, (c) time-activity factors, (d) particle decay rates, (e) indoor $PM_{2.5}$ source strengths, and (f) occupancy and building volume. (a) Air exchange rates are shown for detached/single-family homes ("Detached"), multifamily homes ("Multifamily"), homes without mechanical ventilation (i.e., infiltration and natural ventilation)

("Non-Mechanical"), mechanically ventilated homes ("Mechanical"), homes in developing countries ("Developing"), residential buildings for which the above-described characteristics have not been specified ("Unspecified"), and non-residential buildings ("Non-residential"). (b) Inhalation rates are for adults, children, and by activity level (sleeping, sedentary, light, moderate, and high). (c) Time-activity factors include total hours spent indoors ("Total Indoors"), in the residence ("Residence"), in other indoor locations ("Non-residence"), and at work ("Work") per day. (d) Particle decay rates are for all particle loss mechanisms combined ("Total Decay") and for losses driven only by deposition. (e) Indoor $PM_{2.5}$ emission source strengths include cooking, smoking, solid fuel combustion, and other indoor sources. (f) Occupancy and building volume data are categorized by residential and non-residential indoor environments. Where possible, data are categorized by country/geographic region (Not determined ("n.d.") means that geographic region is unspecified). Studies included here have primarily been conducted in North America and Europe (a,b,c). In addition, there are disparities in the types of indoor environments studied in previous work, with the majority of studies focusing on residential environments and a smaller number of studies considering industrial and commercial buildings.

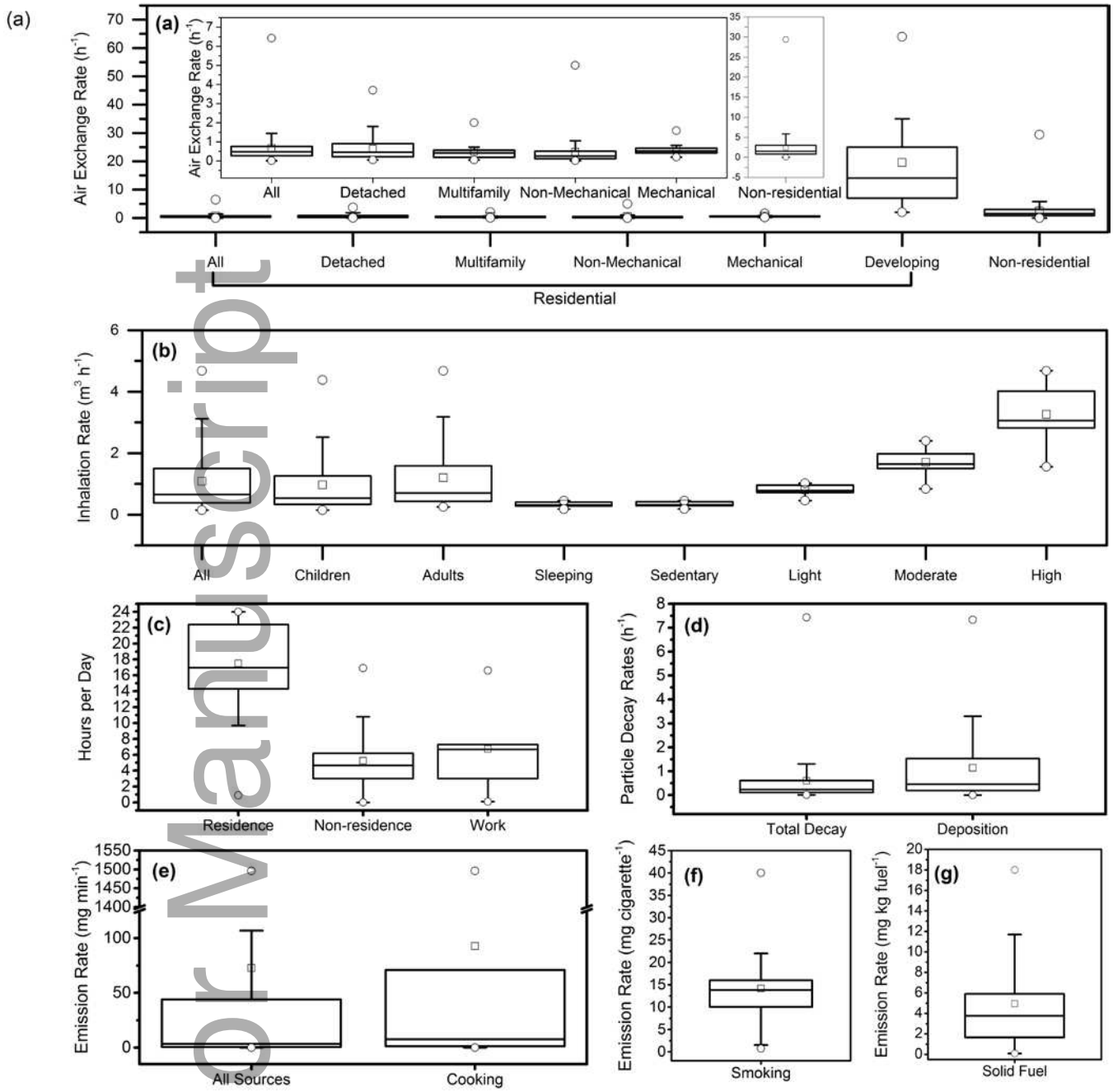
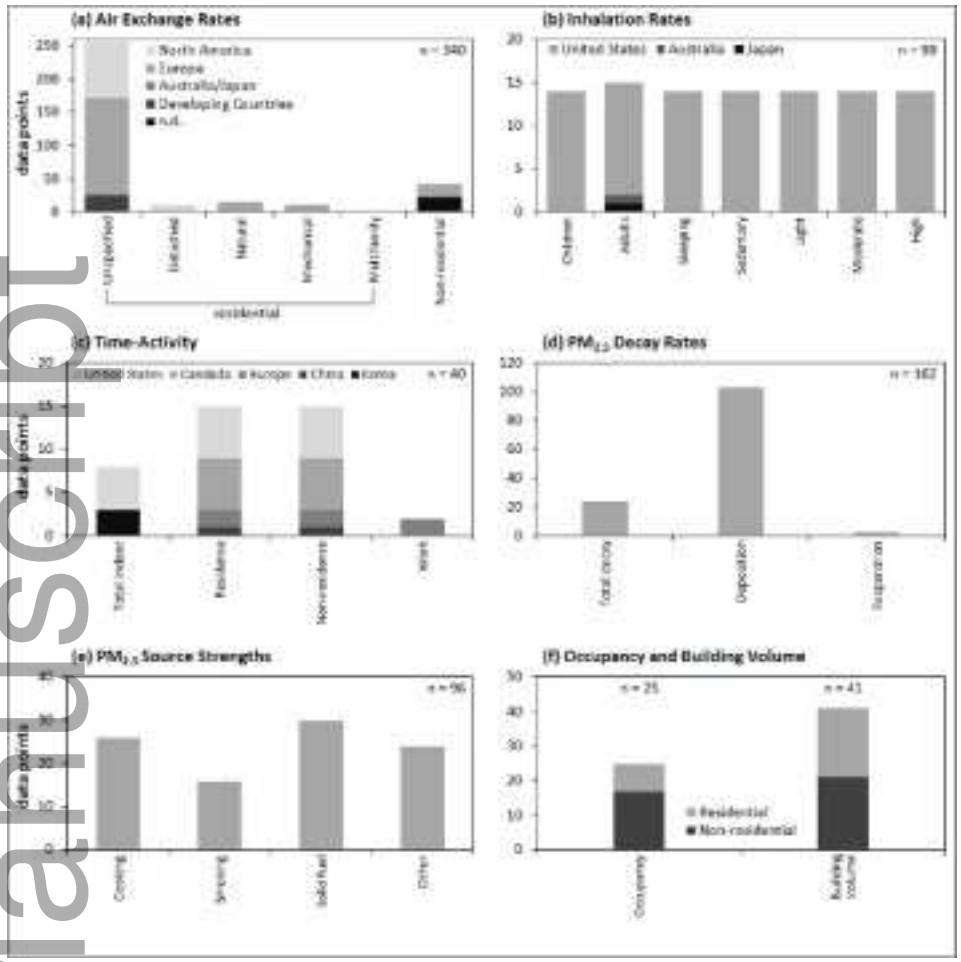


Figure 2. Summary of measured or modeled values describing the parameter of interest for (a) building air exchange rates, (b) inhalation rates, (c) time activity factors, (d) particle decay rates, and (e) – (g) indoor $PM_{2.5}$ source strengths reported in the literature. For all plots, the boxes indicate the 25th percentile, median, and 75th percentile. Minimum and maximum values are indicated with circles and mean values are indicated with squares. (a) Air exchange rates shown are for all homes combined (excluding homes in developing nations) ("All") and separately for detached/single-family homes ("Detached"), multifamily homes ("Multifamily"), homes without

mechanical ventilation (i.e., infiltration and natural ventilation) ("Non-Mechanical"), mechanically ventilated homes ("Mechanical"), homes in developing countries ("Developing"), and non-residential buildings ("Non-residential"). (b) Inhalation rates are for all measurements combined ("All"), and separately for adults (> 21 years), children (≤ 21 years), and activity level (sleeping, sedentary, light, moderate, and high). (c) Time-activity factors include hours per day spent in the residence ("Residence"), in other indoor locations ("Non-residence"), and at work ("Work"). (d) Particle decay rates are given for all particle loss mechanisms combined ("Total Decay") and for losses driven only by deposition. (e) Source emissions are given for common indoor $PM_{2.5}$ sources including cooking, cleaning, smoking, and various appliances combined, excluding the combustion of solid fuels ("All Sources"). (e), (f), and (g) Source emissions are also illustrated for cooking, smoking, and solid fuel combustion separately. The total number of observations for each parameter is shown in Figure 1 and all underlying data are provided in the SI.



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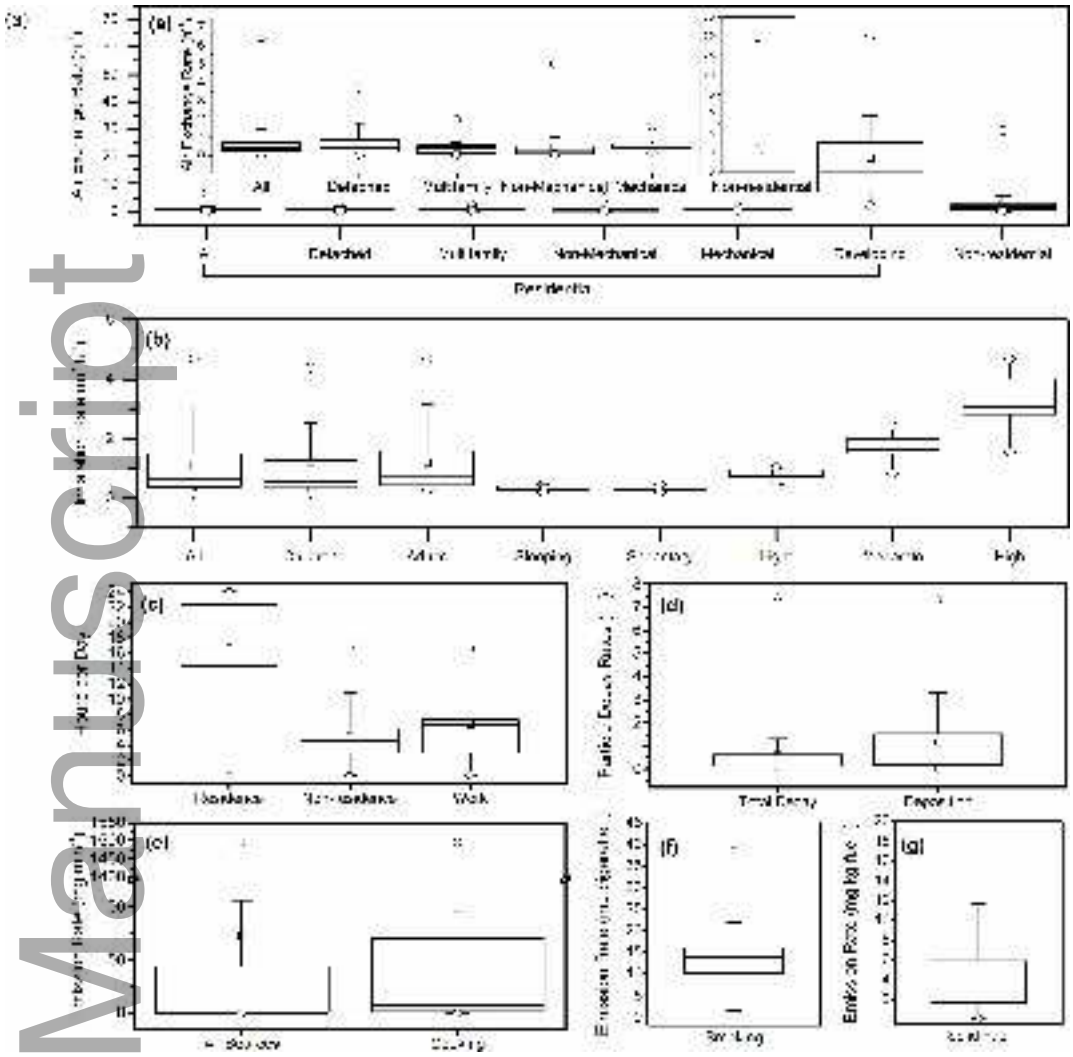

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