INDOOR PARTICLE DYNAMICS

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Abstract

Airborne particulate matter is a diverse pollutant class whose excessive presence in indoor air contributes to an array of adverse health and material-damage effects. Particles are classified according to their diameter into three size modes: ultrafine ($\leq 0.1~\mu m$), accumulation (0.1-2 μm), and coarse ($\geq 2~\mu m$). These modes have largely distinct sources and composition, and they exhibit different dynamic behaviors. The concept of mass conservation or material balance provides a foundation for quantitative and mechanistically linking important outcome variables, such as concentrations and exposures, to the influencing input parameters. The factors governing indoor particle concentrations include direct emissions from indoor sources, ventilation supply from outdoor air, filtration, deposition onto indoor surfaces, and removal from indoor air by means of ventilation. In some circumstances, transport and transformation processes within indoor environments may also play an important role in influencing particle concentrations and consequences. Such processes include mixing, interzonal transport, resuspension, coagulation, and phase change.

Keywords: Deposition; Emissions; Filtration; Material balance; Penetration; Ultrafine

Introduction

Airborne particulate matter (PM) can cause many deleterious effects. Of concern in indoor environments are adverse health outcomes and material damage. The significance of PM as a determinant of indoor air quality is strongly related to airborne particle concentration, size distribution, and chemical or biological composition. In turn, these attributes depend on a suite of other factors, broadly classified as sources, removal mechanisms, and transformation processes. In studying indoor particle dynamics, we seek to elucidate the causal dependencies of indoor particle concentrations and fates on the processes that influence them. In this paper, I first provide an overall framework for thinking about indoor particle dynamics, and then present a synopsis of what is known about the key phenomena.

Importance of Particle Size

The particle size range extends from molecular clusters (diameter, $d_p \sim$ few nm) to larger than 10 μ m in diameter. This is a large range! The proportional difference in diameter between a 3 nm particle and a 10 μ m particle is 3000 ×. Since mass scales with the cube of particle diameter, the 10- μ m particle has a mass that is roughly 30 billion (3 × 10¹⁰) times as large as that of a 3 nm particle. To suggest an intuitive sense of this scale, I would simply note that Microsoft's founder, Bill Gates, the richest individual on the planet, has an estimated net worth (March 2004) of roughly US\$30 billion (http://www.quxxuum.org/~evan/bgnw.html).

Key attributes of particles — sources, composition, dynamic behavior, fates, and effects — are all strongly related to particle size. For the purposes of our discussion, it will be useful to follow the convention of subdividing the particle size spectrum into three modes: ultrafine,

accumulation, and coarse (see Table 1). An alternative classification scheme uses two modes: fine ($< 2 \mu m$) and coarse ($> 2 \mu m$).

Table 1. Attributes of particle size modes.

mode	diameter	sample indoor source	example composition
ultrafine	≤ 0.1 µm	gas cooking	soot
accumulation	0.1 - 2 μm	tobacco smoke	organic liquids
coarse	> 2 μm	cleaning	crustal solids

Material-Balance Principle

The significance of various processes that affect the concentrations and fates of indoor PM can best be understood through the use of a material-balance model. Fundamentally, such a model is based on the concept that mass is conserved. Through a quantitative accounting of all the processes that generate or remove particles from a spatial domain, the model yields equations that mechanistically link outcome variables with important input parameters. A variety of material-balance models have been developed and applied to study indoor PM (Lum and Graedel, 1973; Alzona et al., 1979; Nazaroff and Cass, 1989; Koutrakis et al., 1992; Schneider et al., 1999). These models differ in their treatment of the indoor domain, in their representation of particle characteristics, and in the dynamic processes simulated. Because the indoor particle system is intrinsically complex, even though all deterministic models are based on a single fundamental principle — mass conservation — no single model is well suited for addressing all issues of concern.

As an illustration of the material-balance approach, Fig 1 schematically depicts some of the processes that can affect indoor particle levels. Outdoor air, which contains some particle attribute at a concentration C_o (µg m⁻³), enters the building through three flow pathways: mechanical supply, at flow rate Q_S (m³ h⁻¹); natural ventilation, at flow rate Q_N (m³ h⁻¹); and leakage (infiltration), at flow rate Q_L (m³ h⁻¹). Particles in these airflow streams penetrate into the building to different degrees. Here, the mechanical supply flow is shown passing through a filter with single-pass removal efficiency, η_s (—). It is assumed that there is no attenuation of particles through the natural ventilation flow path, because such airflow passes through relatively large openings. However, only a fraction of the particles, P (—), in the leakage flow path are assumed to penetrate. On a mass basis, these entering airflows must be balanced with airflows exiting the building. When the indoor and outdoor temperatures are similar, then the volume flow rates are also balanced, as suggested by Figure 1. An additional flow path is considered that passes indoor air through a particle-control filter with flow rate Q_F $(m^3 h^{-1})$ and single-pass removal efficiency η_F (—). Within the building there may be an emission source operating at rate E (µg h^{-1}). Particles may be lost from indoor air by deposition onto room surfaces. This process is represented by a first-order loss-rate coefficient β (h⁻¹). The interior volume is V (m³). Let us assume for a moment that these are the only processes that affect the indoor particle levels. If we further assume, as is commonly done in indoor particle dynamics models, that the indoor particle attribute is uniform throughout the interior space, then the conservation of mass can be translated into this governing equation for the indoor concentration of the particle attribute, C_i (µg m⁻³):

$$\frac{d(C_iV)}{dt} = E + C_o \left[Q_s \left(1 - \eta_S \right) + Q_N + Q_L P \right] - C_i \left[Q_F \eta_F + \beta V + \left(Q_S + Q_N + Q_L \right) \right] \tag{1}$$

The term on the left-hand side accounts for the net rate of accumulation of the indoor mass of the particle attribute. The terms on the right-hand side respectively account for indoor emissions, supply from outdoor air via the various ventilation paths, and removal from indoor air, respectively by recirculating filtration, deposition, and ventilation.

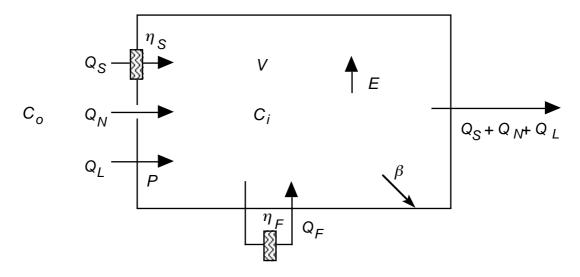


Figure 1. Schematic representation of indoor particle dynamic processes.

Some of the terms in equation (1) vary strongly with particle size: E, η_S , P, η_F , and β . Consequently, equation (1) does not directly represent well the entire airborne particle mass. On the other hand, a reasonable estimate of indoor particle behavior may be obtained by subdividing particle mass into three modes, as presented in Table 1, then writing and solving parallel versions of equation (1) for each mode. The most accurate treatment is obtained by using a particle size distribution function, then applying an equation analogous to (1) to each distinct particle size separately (Riley et al., 2002).

Equation (1) may be extended to include processes in addition to those represented in Fig 1, such as coagulation and phase-change phenomena. The general approach embodied in equation (1) can also be extended to circumstances in which the indoor environment is not represented as a single, well-mixed zone. If the indoor environment can be represented as multiple, well-mixed compartments, then a system of equations of the same general form as equation (1) can be written. Each equation describes the conservation of one particle attribute in a single zone. To the right-hand-side of equation (1) would be added terms to account for supply and loss of the particle attribute by interzonal flow (Nazaroff and Cass, 1989; Miller and Nazaroff, 2001). When airflow conditions are such that uniform mixing within a zone is not reasonably assumed, then a computational fluid dynamics approach may be employed to simulate the spatial variability of particle attributes within the zone (Shimada et al., 1996). To date, such models have tended to treat the particles as static entities, focusing on the details of transport, rather than other aspects of particle dynamic behavior.

Under the idealization that the terms on the right-hand side of equation (1) are time-invariant, then the following steady-state equation describes the relationship between the indoor particle attribute and the key controlling variables.

$$C_i = \frac{E + C_o \left[Q_s \left(1 - \eta_S \right) + Q_N + Q_L P \right]}{Q_F \eta_F + \beta V + \left(Q_S + Q_N + Q_L \right)} \tag{2}$$

The numerator represents the sum of the source terms (units: $\mu g h^{-1}$), accounting for direct indoor emissions plus the sum of the supply rates from outdoor air. The denominator represents the sum of the volumetric loss rates (units: $m^3 h^{-1}$), combining in this case the effects of filtration, deposition, and ventilation.

In general, steady-state conditions do not apply to indoor particle attributes. Indoor emission sources are typically episodic. Outdoor particle levels and ventilation rates commonly vary with time. Under some restrictions about the duration of the averaging period (long compared to the air-exchange time scale) and the independence of the variables, equation (2) may be applied to *estimate* the time-averaged indoor particle attribute as a function of the time-averaged values of the controlling variables (Nazaroff and Klepeis, 2004).

The sections that follow contain discussions of various dynamic processes that affect indoor particle concentrations and fates. First, the terms that appear on the right-hand side of equation (1) are considered. Then, additional dynamic processes that are not incorporated into equation (1), but which are important in some circumstances, are presented.

Indoor Emission Sources (E)

Important sources of indoor particle emissions include combustion processes, cooking, and cleaning activities. Other sources may be important under some circumstances. Table 2 lists some of the studies that have characterized indoor particle sources, focusing on those that have included size-resolved emissions information.

Ultrafine particles are generated by means of gas-to-particle conversion processes, often associated with high temperature phenomena such as combustion or electrical arcing. Depending on the source, the ultrafine mode may be rich in solid carbon (soot) or metals. Substantial ultrafine particle emissions have been reported for cooking activities (Long et al., 2000), especially using gas-fired appliances (Dennekamp et al., 2001). Combustion processes that generate visible plumes, such as cigarette smoking and incense burning, yield particles that are concentrated in the accumulation mode. Cigarette smoking, in particular, has been reported in many field studies to be a major source of indoor fine particulate matter (Wallace, 1996; Nazaroff and Klepeis, 2004). Coarse particles tend to be produced by mechanical means. Such particles may contain common soil elements, such as aluminum and silicon. Particles of biological origin also tend to be coarse, including infectious agents (Nicas et al., 2004), and most bioallergens (Institute of Medicine, 2000).

The relative importance of direct indoor emissions versus intrusion of outdoor particles is reflected in the relative size of the two terms that appear in the numerator of equation (2). If E is much larger than $C_o[Q_S(1-\eta_S)+Q_N+Q_LP]$, then indoor emissions dominate, and vice versa. Based on real-time, size-resolved monitoring data collected in nine nonsmoking Boston-area

homes, Long et al. (2000) found that "indoor particle [emission] events tend to be brief, intermittent, and highly variable."

Table 2. Selected studies of size-resolved indoor particle emission sources

reference	sources studied	
Cheng et al., 1995	incense smoke	
Dennekamp et al., 2001	gas and electric cooking	
Fan and Zhang, 2001	kerosene lamp, oil lamps, candles	
Fine et al., 1999	Candles	
Klepeis et al., 2003	cigarette and cigar smoke	
Li et al., 1993	Cooking	
Li and Hopke, 1993	cigarettes, incense, methane & propane flames, candles	
Long et al., 2000	cooking, cleaning, general indoor activities	
Rodes et al., 1990	ultrasonic humidifiers	
Venkataraman and Rao, 2001	biomass cookstoves	

Particles in Outdoor Air (C_o)

Substantial data are available on ambient particle concentrations, resolved both by size and by chemical composition. Figure 2 displays archetypal size-distribution functions for urban and rural environments. Assuming that the particle density is 1 g cm⁻³, the corresponding mass concentrations are as follows: urban — ultrafine (3.2 μ g m⁻³), accumulation (38 μ g m⁻³), coarse [2-10 μ m] (19 μ g m⁻³); rural — ultrafine (0.2 μ g m⁻³), accumulation (6.2 μ g m⁻³), coarse [2-10 μ m] (8.7 μ g m⁻³).

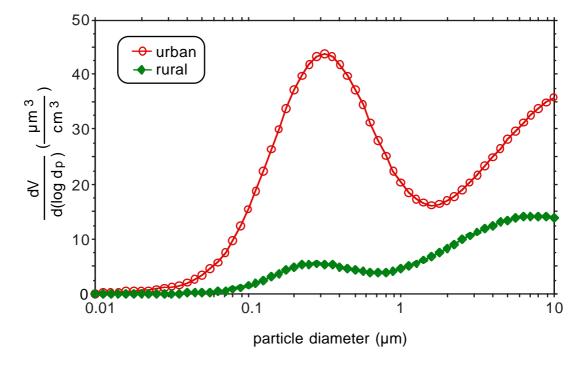
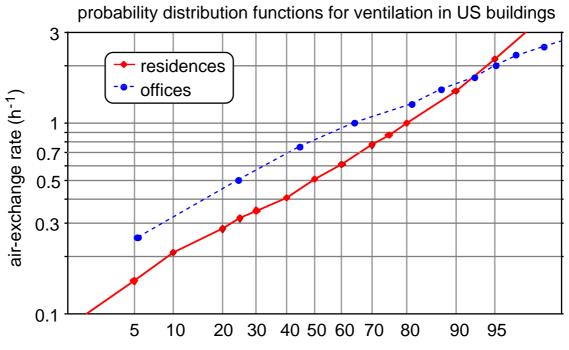


Figure 2. Volume-weighted particle size distribution function for archetypal conditions in ambient urban and rural air. (Source: Seinfeld and Pandis, 1998, Table 7.3)

Ventilation $(Q_S + Q_N + Q_L)$

The process of exchanging indoor air with outdoor air is referred to as *ventilation*. The three main modes are mechanical ventilation (supply flow rate denoted Q_S in Figure 1), natural ventilation (Q_N), and infiltration or leakage flow (Q_L). In the United States, large commercial buildings typically have mechanical ventilation systems, added to which some leakage flow may occur. Small commercial buildings and most residences are ventilated by a combination of natural ventilation through windows and other design openings, plus infiltration. The sum of the flow rates into a building, divided by the interior volume served, is referred to as the *air-exchange rate*. Data on air-exchange rates in US buildings is presented in Fig 3. The data in both cases conform reasonably well to lognormal distributions, with GM = 0.5 h⁻¹ and GSD = 2.1 in residences (N = 2844); and GM = 0.7 h⁻¹ and GSD = 1.8 in the small number of office buildings measured (N = 14).



percentage of building stock with lower air-exchange rate than indicated

Figure 3. Probability distribution functions of air-exchange rates $[(Q_S+Q_N+Q_L)/V]$ measured in US buildings (Sources: Murray and Burmaster, 1995; Persily, 1989).

Filtration (η_S, η_F)

Mechanical ventilation systems are commonly equipped with fibrous filters. Configurations include filtering one or both of the supply and recirculated airflows, although not necessarily using the same filter. In residential settings, filters may be incorporated into central, forced-air thermal conditioning systems. Alternatively, stand-alone recirculating fan-filter units may be deployed to reduce airborne particle concentrations.

Filter performance is characterized in terms of the single-pass removal efficiency (η_S or η_F), which is the fraction of particles captured as air passes through the filter. As depicted in Fig 4, filter efficiency can vary strongly with particle size. As air flows through the filter, the smallest particles are captured on the fibers owing to Brownian motion that causes net

particle motion relative to fluid streamlines. Adhesion results from the nonspecific van der Waals forces, and is sufficient to retain particles on the filter fibers. Coarse particles are captured by the mechanisms of interception and impaction. Interception occurs when a fluid streamline passes within one particle radius of a fiber, causing a particle-fiber collision. Impaction occurs when a particle's inertia causes it to drift toward the fiber and collide with it, even as the air changes direction to flow around the fiber. Particles in the vicinity of 0.3 µm diameter are the most difficult to capture by means of fibrous filtration. They are too large to diffuse, and yet too small for either impaction or interception to cause much removal.

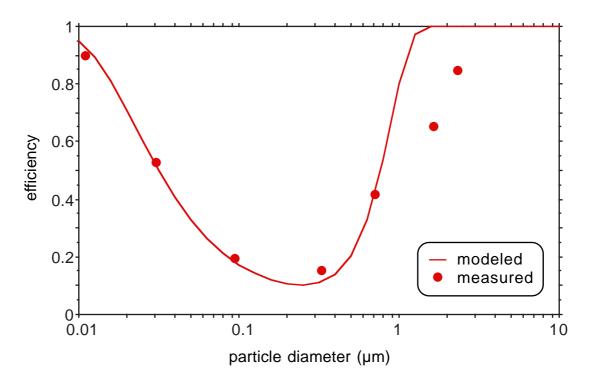


Figure 4. Single-pass filtration efficiency of a clean ventilation filter rated as 65% ASHRAE dust spot average efficiency. Measured data are from Hanley et al., 1994 (Figure 2). Modeled results are based on Hinds, 1982 (Chapter 9). Model parameters: face velocity = 1.3 m/s; solidity = 0.12; fiber diameter = 29 µm; filter thickness = 2.0 cm.

Penetration (P)

Some ventilation air enters buildings through small cracks or gaps in the building envelope. The particles suspended in this air may not make it through the entry pathway, but rather may be deposited onto adjacent surfaces. The parameter *P* denotes the fractional penetration of particles from outdoors to indoors associated with infiltrating airflow. Substantial research during the past half decade has attempted to improve our understanding of particle penetration through building envelopes. This work has employed four approaches: observational studies of indoor-outdoor particle relationships (Long et al., 2001); manipulation experiments in buildings (Thatcher et al., 2003); laboratory measurements of penetration through simulated leakage paths (Mosley et al., 2001; Liu and Nazaroff, 2003); and mathematical modeling of penetration through idealized leakage paths (Liu and Nazaroff, 2001). In sum, this work has illuminated the results expected and the underlying causes, but has not answered every important question about particle penetration. The field experiments are hindered by substantial challenges in the interpretation of inherently complex data. The

laboratory experiments and modeling is constrained to study systems that are more idealized than the reality. Figure 5, based on model calculations, illustrates the expected characteristics for penetration through a thin air-leakage crack. Ultrafine particles deposit on crack walls owing to their rapid Brownian motion. The penetration of coarse particles is limited owing to their tendency to settle onto upward facing surfaces. Particles in the accumulation mode are expected to penetrate most effectively. These results should be seen as analogous to those presented in Figure 4, where we recognize that penetration = 1 - efficiency.

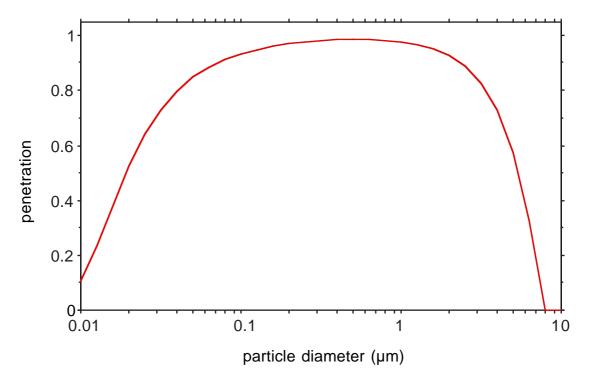


Figure 5. Modeled penetration (*P*) versus particle diameter through a crack in the building envelope (Liu and Nazaroff, 2001). The crack is straight with dimensions 1 mm high by 10 cm long. Airflow is induced by a 4 Pa pressure drop. Particle density is 1.0 g/cm³.

Deposition (β)

Many mechanisms may contribute to particle deposition onto indoor surfaces. Advection and turbulent diffusion are generally strong enough to rapidly transport particles from core air to boundary layers, typically of the order of a centimeter or less from a surface. Transport through the boundary layer controls deposition. For ultrafine particles, Brownian diffusion is an important mechanism. Gravitational settling, possibly augmented by inertial impaction, is important for coarse particles. As with filtration and penetration, accumulation mode particles deposit least effectively.

Figure 6 presents a synthesis of experimental and modeling results, which aims for a single-best estimate of size-dependent particle deposition loss rate for typical indoor environments. Not reflected is the large variability observed among different experiments in deposition rates for particles of any given size (Lai, 2002). Among the factors that can increase particle deposition are more intense airflow and increased indoor surface area (Thatcher et al., 2002).

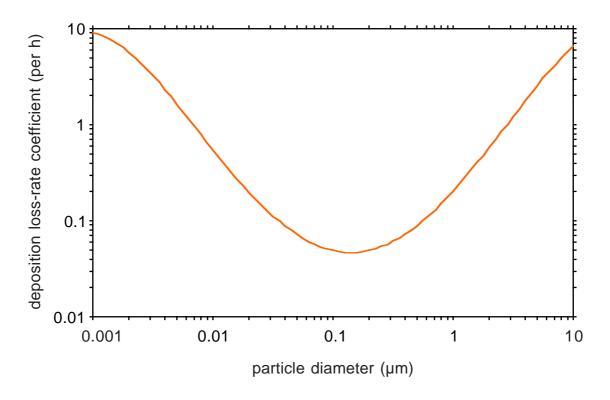


Figure 6. Deposition rate coefficient (β) versus particle diameter. The curve represents a synthesis of model and measurement results, as described by Riley et al. (2002).

Combining Effects

The previous sections of this paper have described a variety of processes and phenomena that can influence indoor particle levels and fates. To illustrate how the various processes operate together, we now consider a specific example. A single-family residence is located in an environment whose outdoor particle concentration is described by the "urban" size-distribution function presented in Fig 2. The building volume is $V = 300 \text{ m}^3$. The respective ventilation flow rates are assumed to be constant at $Q_N = Q_L = 90 \text{ m}^3 \text{ h}^{-1}$, and $Q_S = Q_F = 0$. The particle penetration fraction, P, is assumed to match that presented in Fig 5; likewise, the deposition loss-rate coefficient, β , is the same as presented in Fig 6. As to indoor emissions, only one source is considered: cigarette smoking. It is assumed that 10 cigarettes per day are smoked in the home, each emitting 12 mg of particulate matter whose size distribution is lognormal with a mass median diameter of 0.2 μ m and a geometric standard deviation of 2.1 (Klepeis et al., 2003; Nazaroff and Klepeis, 2004). Particle density is taken as 1.0 g cm⁻³.

Figure 7 displays the predicted time-averaged indoor particle size distribution, evaluated according to equation (2). The upper trace represents the total indoor particle level. The lower trace represents the contribution of particles originating outdoors, whereas the difference between the two curves reflects the contribution of environmental tobacco smoke. The character of a size-distribution plot such as this is that the area under the curve between any two particle size limits is proportional to the volume concentration (and, correspondingly, the mass concentration) between those limits. Integrating these size distributions yields the following cumulative results. The total indoor concentration of ultrafine particles is 7.1 μ g m⁻³, with 38% originating outdoors and 62% caused by cigarette smoking. For the accumulation mode, the total indoor mass concentration is 53 μ g m⁻³, with

38% originating outdoors. For coarse particles, $3.3~\mu g~m^{-3}$ is the total indoor concentration, all of which originates outdoors.

The same conditions were simulated in a second trial, except that the outdoor particle size distribution was changed to the "rural" case depicted in Fig 2. In this case, the total indoor concentration of ultrafine particles was $4.6 \, \mu g \, m^{-3}$ (only 5% were of outdoor origin); the accumulation mode concentration was $25 \, \mu g \, m^{-3}$ (19% of outdoor origin); and the coarse mode concentration was $1.7 \, \mu g \, m^{-3}$ (entirely of outdoor origin).

These exercises are intended to be illustrative only. In homes in which smoking occurs, cigarette combustion is observed to be a strong source of indoor particles (Wallace, 1996; Nazaroff and Klepeis, 2004). Outdoor air, if unfiltered, is also likely to be a consistently strong source of indoor particulate matter (Özkaynak et al., 1996; Riley et al., 2002). However, other sources that directly emit particles into indoor air may also be important, especially so in nonsmoking homes. In their intensive study of nine Boston-area residences, Long et al. (2000) reported that "the impacts of indoor activities are especially pronounced in the ultrafine and coarse modes."

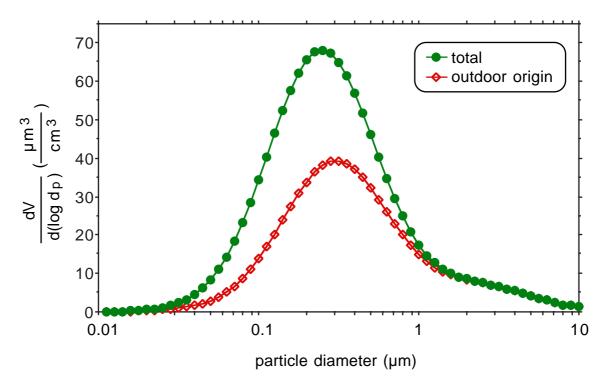


Figure 7. Modeled indoor particle size distribution for a residential environment, according to Equation (2). See text for parameter values.

As another point of comparison, Burke et al. (2001) simulated inhalation exposure to fine particulate matter ($PM_{2.5}$) for the population of Philadelphia, Pennsylvania, USA in a manner that permitted source attribution. They determined the population-mean exposure concentration to be 30 μ g m⁻³. Of this, 70% occurred in residences, 25% occurred in nonresidential buildings, and only 5% occurred outdoors. For the exposure that occurred indoors, 37% was attributable to particles of ambient origin, whereas 63% originated from direct indoor emissions.

Additional Dynamic Processes

The processes and phenomena discussed in the previous sections of this paper have been substantially studied. While not yet complete, our knowledge is sufficient to provide an analytical and quantitative estimate of the effects of these processes on indoor particle levels and fates. Other phenomena may also play a role, in some cases a large role, in affecting indoor particle concentrations and fates. However, these processes have not yet been studied carefully enough to fully incorporate their effects into quantitative assessments. Instead, in the following subsections, key aspects of these phenomena are briefly summarized.

Mixing

Indoor emissions of particulate matter tend to be episodic and localized. Consequently, the effects of direct emissions on inhalation exposure or material damage depends — to an extent — on indoor-air mixing processes. This fact provides both a technical challenge and a technological opportunity. The challenge emerges from the need to consider imperfect mixing in describing the relationships among emissions, concentrations, and exposure. The opportunity arises in exploiting incomplete mixing in exposure-control methods.

In the context of the present discussion, "mixing" refers to the transport and dispersion of a species within a single room. (Transport between rooms is considered in the next subsection.) Mixing is induced by bulk advective flow combined with turbulent fluctuations about the mean flow. Indoor airflow may be induced by mechanical means, such as air discharge from ventilation registers, flow through an open window, or movement of people within a room. Buoyancy owing to temperature differences among indoor-air parcels can induce natural convection flow that contributes to mixing. For example, air will flow upward (downward) along an exterior wall that is warmer (cooler) than the core room air. However, temperature differences can also impede mixing when warmer air is above cooler air.

Experiments have been conducted using tracer gases to investigate mixing in a low-ventilation bedroom-size test chamber. The tracer release occurred over a short time period at a single point in the room. Time-dependent tracer concentrations were measured at an array of sampling points. The mixing time was evaluated as the period after release needed for the spatial variability in concentration to drop to less than 10%. Under natural-convection flow conditions, with significant thermal energy input near the floor (~ 500 W), mixing time scales were roughly 10 min (Baughman et al., 1994). With airflow induced by fans, mixing times were in the range 3-20 min, scaling with the inverse cube root of the rate of power supply into the airflow (Drescher et al., 1995). These latter experiments have been successfully simulated using computational fluid dynamics (Gadgil et al., 2003).

Interzonal Transport

Airflow between rooms can strongly influence indoor air pollutant concentrations and fates. Within the broader field of indoor air quality, this topic has received too little attention. Factors governing the rates of interzonal flow are understood in principle. The driving forces are pressure differences caused by winds, temperature differences, and fan operation. The leakage between rooms limits the flow that is induced by these pressure differences. However, empirical data are insufficient to estimate typical flows or their variability. Specifically with respect to indoor particles, a few studies have explored concentration variability among rooms and the factors that influence them. For example, Ju and Spengler

(1983) measured 24-h average respirable particle concentrations in multiple rooms of four homes and found only small between-room differences. On the other hand, in a laboratory study, Miller and Nazaroff (2001) found that closing a door between two rooms reduced the rate of airflow between them from $\sim 60 \text{ m}^3 \text{ h}^{-1}$ to $\sim 1 \text{ m}^3 \text{ h}^{-1}$. With an open doorway, tobacco smoke particles released in one room became rapidly mixed throughout both. But with the door closed, the nonsmoking room had much lower particle levels than the smoking room even four hours after the cigarette was smoked.

Resuspension

Particles previously deposited onto surfaces may become airborne through resuspension. This process may be a key part of an important exposure pathway for allergens (Institute of Medicine, 2000) and also for semivolatile species such as phthalate esters (Clausen et al., 2003). Ordinary indoor activities — including walking and housekeeping — can cause particle resuspension. Such activities may also generate new particles through abrasive wear of surfaces. Only a few studies have investigated particle resuspension. Thatcher and Layton (1995) observed increases in indoor concentrations of supermicron particles during episodes of human activities in a house. Ferro et al. (2004) quantified effective mass emission rates of PM_{2.5} and PM₅ in one house associated with prescribed household activities: folding blankets, folding clothes, dry dusting, making a bed, dancing, vacuuming, walking, and sitting on upholstered furniture. Kildeso et al. (1999) developed an experimental technique for quantifying dust resuspension potential from carpets.

Coagulation

When particles collide with one another they tend to adhere. This process, known as coagulation, does not directly cause a change in particle mass concentration. However, it does shift particle size distributions. Coagulation is a second-order phenomenon, meaning that the rate of the process depends on the square of the particle concentration. Most other processes affecting indoor particles are first order. The significance is that coagulation can be important when particle concentrations are high, but relatively unimportant when particle concentrations are low. The most important mechanism causing coagulation indoors is Brownian motion, the random particle migration caused by collisions with surrounding gas molecules. Brownian motion is progressively more effective as a transport mechanism for progressively smaller particles. Commonly, the highest number concentration of particles indoors is found in the ultrafine mode, especially in the presence of a fresh source of emissions. In combination, these circumstances cause coagulation to be of primary importance in affecting ultrafine particles during periods when their concentrations are unusually high. These particles can collide with one another, which produces only a moderate shift in size, or they can collide with larger particles. The latter situation would generally result in a transfer of suspended particle mass from the ultrafine mode into the accumulation mode. The influence of coagulation on indoor particle levels has been explored for environmental tobacco smoke (Nazaroff and Cass, 1989). The effects were relatively small for the case of a single cigarette smoked in a low-ventilation room. Coagulation is likely to be important in affecting the evolution of ultrafine particles emitted from unvented natural gas combustion (Dennekamp et al., 2003).

Phase Change

Indoor particulate matter can be altered owing to phase-change processes, in which chemical species undergo a change of state from gas phase to condensed phase, or vice versa.

Published evidence documents at least four classes of phase-change processes affecting indoor particles. Dua and Hopke (1996) documented growth associated with water uptake or release when indoor particles were exposed to changing humidity conditions. Pankow et al. (1994), Naumova et al. (2003), and Weschler (2003) have reported on investigations of the phase-partitioning of semivolatile organic compounds between the gas-phase and sorbed to indoor airborne particles. Several papers have been published during the past half-decade documenting the production of secondary organic particulate matter as a result of indoor reactions between ozone and terpenes (Weschler and Shields, 1999; Wainman et al., 2000; Sarwar et al., 2003; Fan et al., 2003). Most recently, Lunden et al. (2003) reported that, under certain circumstances, ammonium nitrate particles from outdoor air dissociated into their gaseous constituents — nitric acid and ammonia — when the particles were brought into an indoor environment, causing a significant net loss of airborne particle mass. These reports demonstrate circumstances in which indoor particle dynamics cannot be well characterized without some understanding of phase-change phenomena.

Conclusion

Airborne particles are important components of indoor environments. They should not be thought of as a single species, but rather as a major class, characterized by attributes that can vary widely, such as size and composition. A suite of processes controls the particle-related characteristics of indoor air. In many respects, our present level of understanding of these processes is not well developed. However, with attention, rapid progress is possible, as has been demonstrated during the past decade. Continued advances hold the promise of helping us to create and maintain more healthful indoor environments.

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