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Using portable particle sizing instrumentation to rapidly measure the penetration of fine and ultrafine particles in unoccupied residences

Abstract Much of human exposure to particulate matter of outdoor origin occurs inside buildings, particularly in residences. The particle penetration factor through leaks in a building's exterior enclosure assembly is a key parameter that governs the infiltration of outdoor particles. However, experimental data for size-resolved particle penetration factors in real buildings, as well as penetration factors for fine particles less than 2.5 μm ($PM_{2.5}$) and ultrafine particles less than 100 nm (UFPs), remain limited, in part because of previous limitations in instrumentation and experimental methods. Here, we report on the development and application of a modified test method that utilizes portable particle sizing instrumentation to measure size-resolved infiltration factors and envelope penetration factors for 0.01–2.5 μm particles, which are then used to estimate penetration factors for integral measures of UFPs and $PM_{2.5}$. Eleven replicate measurements were made in an unoccupied apartment unit in Chicago, IL to evaluate the accuracy and repeatability of the test procedure and solution methods. Mean estimates of size-resolved penetration factors ranged from 0.41 $\pm$ 0.14 to 0.73 $\pm$ 0.05 across the range of measured particle sizes, while mean estimates of penetration factors for integral measures of UFPs and $PM_{2.5}$ were 0.67 $\pm$ 0.05 and 0.73 $\pm$ 0.05, respectively. Average relative uncertainties for all particle sizes/classes were less than 20%.

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Key words: PM2.5; Indoor aerosols; Infiltration factor; Penetration factor; Building envelope; SMPS.

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Practical Implications

This work provides a novel test method for measuring the penetration of outdoor fine and ultrafine particulate matter into indoor residential environments. The method is designed to minimize the duration of testing without sacrificing accuracy and to provide a practical solution for further application in field measurements in a greater number and variety of buildings.

Introduction

Epidemiology studies have consistently demonstrated associations between increased adverse health effects and elevated outdoor particulate matter, including mass concentrations of fine particulate matter ($PM_{2.5}$: particles less than 2.5 μm) (Brook et al., 2010; Miller et al., 2007; Pope and Dockery, 2006; Pope et al., 2002, 2009) and number concentrations of ultrafine particles (UFPs: particles less than 100 nm) (von Klot et al., 2002; Osunsanya et al., 2001; Penttinen et al., 2001; Stölzel et al., 2007; Weichenthal et al., 2007). Because outdoor particles can infiltrate into buildings (Chen and Zhao, 2011) where people spend most of their time (Klepeis et al., 2001), the majority of human exposure to outdoor $PM_{2.5}$ and UFPs actually occurs indoors, particularly in residences (Allen et al., 2004; Bhangar et al., 2011; Ji and Zhao, 2015; Kearney et al., 2011; Meng et al., 2005, 2009). Additionally, some studies have also suggested that particles of outdoor origin may be more toxic than indoor-generated particles (Ebelt et al., 2005; Koenig et al., 2005). Therefore, to improve our understanding of human exposure to airborne particulate matter, including $PM_{2.5}$ and UFPs, it is crucial to better understand how outdoor particles infiltrate into residential indoor environments.

Under infiltration conditions with doors and windows closed and in the absence of indoor emission

sources, three main parameters describe the fraction of ambient particulate matter that infiltrates and persists indoors (i.e., the infiltration factor, F_{inf}). These include the following: (i) the air exchange rate (λ or AER, h^{-1}); (ii) the particle deposition loss rate constant (k, h^{-1}), which characterizes particle removal due to deposition to interior surfaces, removal by any particle control systems, and any other removal mechanisms; and (iii) the penetration factor, which is defined as the fraction of particles in the infiltrating air that passes through the boundary of the building enclosure (P, dimensionless). An expression for time-averaged infiltration factors for a given class or size of particles in a building relying on infiltration alone is shown in Equation 1.

$$F_{inf} = \frac{P\lambda}{\lambda + k}. \quad (1)$$

Measurements of F_{inf} are relatively straightforward to make as long as indoor and outdoor particle concentrations and size distributions are relatively stable: one must only measure indoor and outdoor particle concentrations in the absence of indoor sources. Previous studies have shown that time-averaged values of F_{inf} commonly vary between residential buildings from less than 0.1 to nearly 1.0, depending on the size/class of particles (e.g., F_{inf} for UFPs is typically lower than for $PM_{2.5}$), window-opening behavior, and other underlying building characteristics such as envelope airtightness (Allen et al., 2012; Bennett and Koutrakis, 2006; Chen and Zhao, 2011; Kearney et al., 2011, 2014; Long and Sarnat, 2004; MacNeill et al., 2012, 2014; Rim et al., 2010; Stephens, 2015; Wallace and Williams, 2005; Zhu et al., 2005).

Measurements of the three underlying parameters that govern infiltration factors are more challenging to perform. Air exchange rates (λ) are the simplest to measure using tracer gas techniques (ASTM, 2006) and have been measured in thousands of homes (Murray and Burmaster, 1995). Deposition loss rate constants (k) are relatively straightforward to measure as well using a particle elevation and decay procedure alongside air exchange rate measurements, and several studies have reported measurements of deposition loss rate constants across a wide range of particle sizes/classes and building operational conditions, including with and without central HVAC filtration (He et al., 2005; Howard-Reed et al., 2003; Lai, 2002; Lee et al., 2014; Thatcher, 2002; Thatcher and Layton, 1995; Wallace et al., 2004, 2013; Zhao and Wu, 2007).

Measurements of envelope penetration factors are generally the most challenging to perform given that two parameters (both P and k) must be estimated from one mass balance equation applied to periods in which the space has no indoor particle sources. Air exchange rates must also be measured simultaneously. However, accurate measurements of penetration factors through building enclosures are crucial. For one, most residential buildings in the United States do not have dedicated mechanical ventilation systems but rely on a combination of infiltration and window opening for ventilation air (Chan et al., 2005). Moreover, window-opening frequencies in U.S. residences are not well known, but they are often less than 20–30% of the time, depending on outdoor weather conditions and occupant preferences (Chen et al., 2012; El Orch et al., 2014; Johnson and Long, 2005; Price and Sherman, 2006). Thus, for many times of the year in many locations, particles of outdoor origin enter residential indoor environments only after penetrating through leaks in the building enclosure assembly. Failing to account for variability in penetration factors accurately can lead to inaccurate estimates of infiltration factors and, ultimately, exposure errors in epidemiology studies (Baxter et al., 2010, 2013; Breen et al., 2015; Chen et al., 2012; Hodas et al., 2012, 2013).

The envelope penetration process is influenced by several factors, including the geometry of leaks, pressure differences across the envelope, air velocities through leaks, and particle size (Liu and Nazaroff, 2001; Nazaroff, 2004). To date, specific measurements of particle penetration factors have been made in fewer than approximately 50 homes worldwide (Chao et al., 2003; Long et al., 2001; Mosley et al., 2001; Rim et al., 2010, 2013; Stephens and Siegel, 2012, 2013; Thatcher and Layton, 1995; Thatcher et al., 2003; Vette et al., 2001; Zhu et al., 2005). Each of these studies has varied considerably in their measurement approach, test duration, resulting uncertainty in parameter estimates, and the sizes and classes of particles that were measured. For example, some studies characterized size-resolved penetration factors only for particles larger than 0.3 μm (Chao et al., 2003; Thatcher et al., 2003), while others characterized only ultrafine or submicron particles, either with or without size resolution (Rim et al., 2010; Stephens and Siegel, 2012; Zhu et al., 2005). We are not aware of any studies that have experimentally characterized $PM_{2.5}$ penetration factors, although several studies have estimated P for $PM_{2.5}$ from time-integrated gravimetric measurements using a variety of statistical techniques (Breen et al., 2015; Meng et al., 2005; Williams et al., 2003).

Further, some of the previously applied specific penetration test methods have also required long test durations that make them impractical to perform in the field and thus limit their applications. For example, some have relied on overnight measurements to estimate penetration factors during periods when occupants were assumed to be inactive (Long et al., 2001; Vette et al., 2001). Others have relied on measurement durations as long as two or more days while the build-

ing was unoccupied (Rim et al., 2010). Thus, there remains a need to integrate these prior methods into a technique that can be used to (i) estimate size-resolved penetration factors for a wider range of particle sizes simultaneously (i.e., from the nanometer to micrometer range), (ii) generate estimates for penetration factors for integral measures of both UFPs and $PM_{2.5}$ (particularly for informing regulations and epidemiology studies), and (iii) decrease the test duration to allow for more practical widespread application in a greater number and variety of homes.

Therefore, in this work, we have developed a method for rapidly measuring size-resolved particle penetration factors for fine and ultrafine particles in residences using portable particle sizing instrumentation. The method should be performed when the building is unoccupied. The resulting size-resolved data are also used to calculate penetration factors for integral measures of UFPs by summing across the sub-100 nm size bins and $PM_{2.5}$ by making assumptions for particle shape and density and summing estimated masses across all sub-2.5 μm size bins. Here, we describe the development of the test method and its repeated application in an unoccupied test apartment in Chicago, IL. Eleven replicate measurements were made over a period of 6 months under a variety of indoor and outdoor environmental conditions. We use the resulting data to: (i) investigate the accuracy and repeatability of the method for solving for both penetration factors and deposition loss rate constants of size-resolved particles and integral measures of UFPs and $PM_{2.5}$; (ii) explore potential influences of indoor and outdoor environmental factors on both penetration factors and deposition loss rate constants; and (iii) conduct side-by-side comparisons of $PM_{2.5}$ infiltration factors measured using fine PM instrumentation and those estimated using the size-resolved instrumentation.

Methods and materials

Test apartment description

Measurements were conducted from March through August 2015 in *studioE* (the Suite for Testing Urban Dwellings and their Indoor and Outdoor Environments), an unoccupied, sparsely furnished apartment unit on the third floor of a nine-story residence hall on the main campus of Illinois Institute of Technology in Chicago, IL. The apartment unit is described in more detail in the SI and in Zhao and Stephens (2015). Briefly, the unit has a floor area of ~60 m^2 and a volume of ~150 m^3. Approximately half of the perimeter walls are exterior walls; the ceiling, floor, and other half of perimeter walls are all adjacent to other interior spaces (i.e., other apartment units and the hallway). All windows and the only perimeter door were kept closed during the measurements. All internal doors were kept opened, and several oscillating fans were operated to enhance mixing.

Measurements of indoor and outdoor particle concentrations

The method utilizes a combination of a TSI NanoScan Scanning Mobility Particle Sizer (SMPS; TSI NanoScan Model 3910, Shoreview, MN, USA) and TSI Optical Particle Sizer (OPS; TSI Model 3330, Shoreview, MN, USA) to measure particle concentrations from ~10 nm to ~2.5 μm in mobility and optical diameter, respectively. We rely on size-resolved measurements because (i) the penetration of both fine or ultrafine particles is governed by the underlying particle size distribution (Chen and Zhao, 2011; Liu and Nazaroff, 2001; Nazaroff, 2004), and (ii) most optical fine PM instrumentation is not sufficiently accurate at low concentrations to estimate both penetration factors and deposition loss rate constants with low uncertainty from time-resolved indoor and outdoor data, at least when applied in locations with low or moderate outdoor particle concentrations such as most of the United States (Wallace et al., 2010). The SMPS utilizes an isopropanol-based condensation particle counter (CPC) and a radial differential mobility analyzer for size resolution across 13 bins of mobility diameter, nominally from 10 to 420 nm (Tritscher et al., 2013). The NanoScan was operated in full scan mode during all tests. The OPS yields particle concentrations in up to 16 bins of optical diameter from 0.3 to 10 μm. The measuring system also included a DustTrak DRX aerosol monitor (TSI DRX Model 8534, Shoreview, MN, USA), which simultaneously measures size-segregated mass fraction concentrations corresponding to PM_1, $PM_{2.5}$, and PM_{10} size fractions using optical methods.

The three portable instruments were all connected to the same sampling system using TSI conductive silicon tubing and placed in a medium-sized audio rack case on rollers located in the middle of the living room and adjacent to an exterior window. Two rigid stainless steel sampling lines approximately 1.5 m in length and 0.6 cm in diameter were used for both indoor and outdoor sampling to minimize particle deposition losses and also to keep losses approximately equal for both indoor and outdoor sampling lines. Outdoor samples were drawn through a small (~0.6 cm) penetration in a clear acrylic window in the living room with the outdoor sampling line extending approximately 1 m out from the exterior window surface. In field tests in a real home, we recommend inserting the outdoor sampling line through a window opening that is just large enough to accommodate the outer diameter of sampling line, which would then be taped and sealed.

The aerosol sampling system was connected to a Swagelok Model SS-43GXS4-42DCX electrically actuated three-way ball valve to alternately monitor indoor and outdoor air. The valve was controlled by a three-channel electronic timer (Sestos B3S-2R-24) set to alternately measure indoor and outdoor particle concentrations at 8-min intervals, alternating between 4-min indoors and 4-min outdoors. The switching valve requires ~5 s to switch between indoors and outdoors, so a two-way normally closed solenoid pinch valve (Cole-Parmer Model 075P2NC-24-01, Vernon Hills, IL, USA) was connected to the sampling system to automatically open for 5 s at 4-min intervals to protect from choking the instrument flows during the switching period. During each test, all of the aerosol monitors logged data at 1-min intervals. Transition points between indoor and outdoor sampling periods were identified visually and discarded prior to data analysis. The entire sampling system was well sealed, and the total transport loss was consistently measured to be approximately 15%.

Penetration test method procedure

The modified test procedure is based largely on shorter-term measurement techniques previously reported in the literature (Chao et al., 2003; Stephens and Siegel, 2012; Thatcher et al., 2003). It involves the following steps to yield estimates of both penetration factors (P) and deposition loss rate coefficients (k) from one mass balance equation in the unoccupied space. First, outdoor particles were introduced by opening windows and operating a blower door fan and frame installed in a doorway to induce cross-ventilation flow. This procedure served to elevate indoor particle concentrations near their outdoor concentrations and also replace indoor aerosols with the same aerosol composition that exists outdoors. The latter is essential for achieving reasonable estimates of deposition loss rate constants for the outdoor-infiltrated aerosol. The use of artificial means to elevate indoor particle concentrations (e.g., burning incense or operating gas burners) will result in decay rate estimates that may differ from those for ambient particles due to different particle composition, shape, and/or density. Periodic measurements with a handheld TSI CPC Model 3007 confirmed that indoor submicron particle concentrations indeed reached to 90% or more of the simultaneous outdoor submicron particle concentrations during the elevation period.

Next, a tracer gas (CO_2) was injected to measure the air exchange rate and the unit was left unoccupied to measure the subsequent decay of both indoor particles and CO_2 concentrations with all exterior windows and doors closed. An automatic CO_2 injection system was activated immediately after the windows and doors were closed, as described in the SI. Indoor particle measurements continued for 40–60 min during an indoor-only measurement period prior to engaging any indoor/outdoor switching. A two-channel timer (Sestos B2S-2R-220) was used to keep the switching valve for the entire sampling system powered off to maintain indoor measurements for this first hour-long period, which served to yield consecutive data points for estimating the first-order decay rate constant from the initial decay period.

Finally, after the initial decay period, indoor and outdoor particle concentrations were alternately measured during normal infiltration conditions over a period of approximately 2–3 h. To initiate this sequence, the two-channel timer automatically turned on the power for the particle sampling switching system (which consisted of the switching valve, the two-way normally closed solenoid pinch valve, and the three-channel electronic timers for the valve). The aerosol instruments then alternately measured indoor and outdoor particle concentrations at 8-min intervals under normal infiltration conditions over a period of at least 2 h, and typically less than 4 h. These alternating measurements allowed for tracking the change in indoor particle concentrations due to the infiltration of outdoor particles alone (in addition to any infiltration from adjacent apartment spaces, which we believe was minimal as described in the SI).

Data analysis and parameter estimates

We first used the resulting data to solve for size-resolved infiltration factors (F_{inf}), penetration factors (P), and deposition loss rate constants (k) for as many size bins as possible, as described in the next section. We then estimated F_{inf}, P, and k for integral measures of UFPs and fine particulate matter mass ($PM_{2.5}$). The integral measures of indoor and outdoor UFP number concentrations were calculated at each time step by summing the resulting concentrations from the first eight bins of the SMPS (i.e., 10–101 nm). Similarly, the integral measures of indoor and outdoor $PM_{2.5}$ mass concentrations were estimated at each time step by calculating the mass concentration in each particle size bin smaller than 2.5 μm from the combination of the SMPS and OPS, assuming spherical particle shape and two different density conditions, and then summing these estimated size-resolved masses across all particle size bins less than 2.5 μm. Similar approaches have been used with an SMPS and OPS system to estimate outdoor $PM_{2.5}$ concentrations (Buonanno et al., 2009; Shen et al., 2002). For the first density condition, we assumed that all particles had a constant unit density of 1 g/cm^3, both for simplicity and to reflect previous assumptions used in the literature (Azimi et al., 2014; El Orch et al., 2014; Riley et al., 2002; Waring and Siegel, 2008; Zaatari et al., 2014). For the second density condition, we assumed that density varied with particle diameter according to the average of that reported during two

experimental campaigns in two European cities (Neusüss et al., 2002): 1.3 g/cm^3 for Dp < 140 nm; 1.4 g/cm^3 for 140 nm < Dp <420 nm; 1.5 g/cm^3 for 420 nm < Dp < 1.2 μm; and 1.6 g/cm^3 for 1.2 μm < Dp < 3.5 μm. Although not necessarily directly applicable to particle densities in Chicago, this approach served to evaluate the sensitivity of these methods to varying assumptions for particle density.

Parameter estimates. We used a dynamic mass balance approach to model the time-varying indoor particle concentration for all size bins, as well as the integral measures of UFPs and $PM_{2.5}$, in the well-mixed environment in the absence of indoor sources, as shown in Equation 2.

$$\frac{dC_{in}}{dt} = P\lambda C_{out} - (\lambda + k)C_{in}. \quad (2)$$

We should note that applying Equation 2 to integral measures of UFPs and $PM_{2.5}$ could lead to biases in estimates of P and k for those measures because it fails to explicitly account for shifts in the underlying size dependencies of P and k. However, practical limitations prevented us from making reasonable estimates of both P and k for many of the larger particle size bins above 1 μm (i.e., low number concentrations and large amounts of scatter prevented parameter estimates). Therefore, we rely mostly on integral measures of UFPs and $PM_{2.5}$ herein, in addition to the sub-1-μm size-resolved bins. We explore the likely magnitude of the impact of this procedure on parameter estimates for integral UFPs in the SI (although the same analysis could not be completed with integral or size-resolved $PM_{2.5}$ data).

For each test, the AER was estimated by regressing the natural logarithm of the tracer gas concentrations versus time (ASTM, 2006), as shown in Equation 3.

$$-\ln\frac{Y_{in,t} - Y_{out}}{Y_{in,t=0} - Y_{out}} = \lambda t \quad (3)$$

where $Y_{in,t}$ and $Y_{in,t=0}$ are the indoor CO_2 concentrations (ppm) measured at time t and $t = 0$, respectively. Y_{out} is the average outdoor CO_2 concentration (ppm) during the test, and λ is the average air exchange rate (h^{-1}). The tracer decay period typically lasted the same duration as the penetration measurements (~3–4 h). Only, data that clearly fit the log-linear exponential decay function in Equation 3 (with $R^2 > 0.99$) were used to estimate AERs. Data from periods of any drastic changes due to changing meteorological conditions were discarded.

To solve for the two unknown parameters in Equation 2 (P and k) using the resulting time-varying data, we explored different methods of solution that have been reported in previous studies (Chao et al., 2003; Rim et al., 2010; Stephens and Siegel, 2012). We also informed our solution methods based on a recent study of ozone penetration factor measurements that were able to achieve solutions for P and k with high fitness and low uncertainty (Zhao and Stephens, 2015). The deposition loss rate constant (k) was first estimated using a log-linear regression solution to the initial indoor decay portion of the data, as shown in Equation 4. Similar approaches have been applied in previous studies (e.g., Chao et al., 2003).

$$-\ln\frac{C_{in,t} - C_{bg}}{C_{in,t=0} - C_{bg}} = (\lambda + k)t \quad (4)$$

where $C_{in,t}$ and $C_{in,t=0}$ are the indoor particle concentrations at time t and $t = 0$, respectively. C_{bg} is the background indoor particle concentration (i.e., the steady-state indoor particle concentration), which was either measured or estimated using one of multiple procedures described in the SI.

Next, we solved for P for each particle size bin (including integral measures of UFPs and $PM_{2.5}$) using a discretized solution to Equation 2 (shown in Equation 5), with prior knowledge of λ from Equation 3 and k from Equation 4.

$$C_{in,t} = P\lambda C_{out,t}\Delta t + (1 - (\lambda + k)\Delta t)C_{in,t-1} \quad (5)$$

where $C_{in,t}$ and $C_{out,t}$ are the indoor and outdoor particle concentrations at time t, respectively, and Δt is the time step (i.e., 8 min). In this one-parameter discretized solution method, P was estimated using a nonlinear least-squares regression combined with the earlier estimates of λ and k. This solution method estimates k separately and takes into account varying outdoor particle concentrations during the test period, typically yielding relatively low uncertainties and high fitness for estimates of both P and k. All parameter estimates were performed using a statistical software package, Stata Version 13 (StataCorp SE, College Station, TX, USA).

Estimation of uncertainty. We used approaches to estimate the uncertainties associated with our estimates of P and k that were similar to our recent work estimating ozone penetration factors (Zhao and Stephens, 2015). The uncertainty in each AER estimate was first calculated using the standard errors of the regression coefficients from Equation 3, and the average accuracy of the CO_2 monitors (±20 ppm) added in quadrature. The relative uncertainty in the total loss rate constant (λ+k) was calculated by combining the relative standard deviation of the average steady-state indoor concentration, infiltration factor, or the standard error from the predicted background concentration, combined with the relative standard error

of the regression coefficient from Equation 4 added in quadrature. The uncertainty in our estimate of k was then calculated by combining the uncertainty of the total loss rate constant and the AER uncertainties. Finally, the propagated uncertainty in P was estimated by error propagation with a combination of relative uncertainties of total loss rate constants, AER, and the standard error of regression coefficients for P.

Results and discussion

An example of resulting indoor and outdoor integral UFP concentration data from one typical elevation and decay experiment conducted on March 25, 2015 is shown in Figure 1a. The first portion of indoor decay data was used to solve for a UFP deposition loss rate constant (k) for this experiment, as shown in Figure 1b. The AER (±propagated uncertainty) during this test was 1.16 ± 0.05 1/h, and the estimated value for k was 0.89 ± 0.09 1/h.

Size-resolved infiltration factors

Figure 2 shows the size-resolved steady-state indoor and outdoor particle infiltration factors measured by both the SMPS and OPS across 18 size bins from 0.01 to 2.5 μm mobility and optical diameter, respectively, during each of the 11 replicate tests performed in the test apartment unit under infiltration conditions. The average value for F_{inf} across all 11 tests is also shown by the thick black line. Air exchange rates measured during each test period, which ranged from 0.49 to 1.16 1/h, are also listed in the legend. Size-resolved infiltration factors were calculated by averaging the ratios of indoor and outdoor concentrations after 3/(λ+k) h, which we considered to be at approximately steady-state conditions.

The minimum and maximum average infiltration factors (±s.d.) were 0.15 ± 0.06 with a geometric mean particle diameter of 0.015 μm and 0.55 ± 0.09 with a geometric mean particle diameter of 0.866 μm, respectively. Infiltration factors for UFPs generally increased with air exchange rate, but the infiltration factors for fine particles were mostly influenced by deposition loss rate constants and/or envelope penetration factors. The overall shape of the size-resolved infiltration factor curve is generally consistent with other experimental (Long et al., 2001; Rim et al., 2010; Zhu et al., 2005) and modeling studies (El Orch et al., 2014; Liu and Nazaroff, 2001; Nazaroff, 2004). We should note that there is a discontinuity between ~0.1 and ~0.3 μm that unfortunately stems from known issues with the TSI NanoScan inaccurately reporting above ~0.1 μm when concentrations of those particle sizes are low relative to the total number concentration (Yamada et al., 2015). According to the manufacturer, the issues arise due to the method of fitting distributions, which is required because of the use of a unipolar charger. Ignoring this discontinuity results in a slight deviation from the traditional upside-down U-shaped curve for infiltration factors, which may lead to slight underestimates of F_{inf}, P, and k for particles between 0.1 and 0.3 μm. We should also note that we were able to estimate F_{inf} for the 115 nm size bins using the available data from the NanoScan in only five of the 11 replicate tests due to this same issue appearing intermittently for that bin.

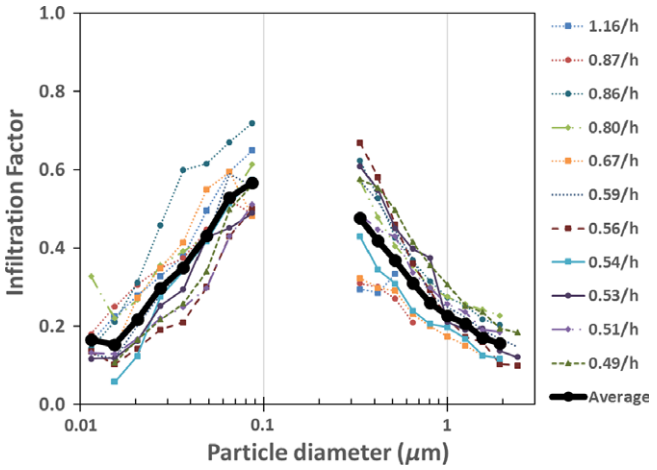

Fig. 2 Size-resolved particle infiltration factors with particle diameters ranging from 0.01 μm to 2.5 μm in mobility and optical diameter, respectively, across 11 replicate tests with various air exchange rates in the apartment unit

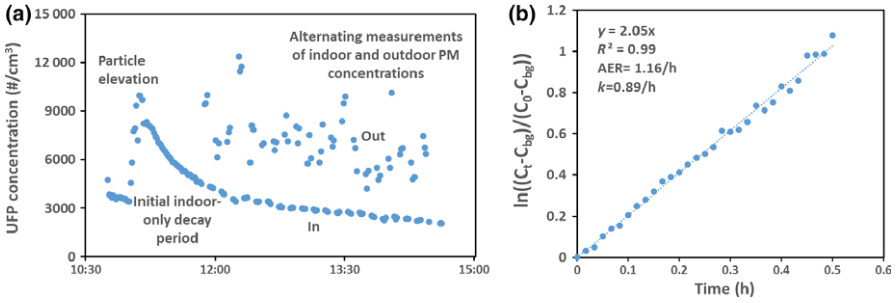

Fig. 1 Example of data from a particle elevation and decay measurement: (a) time-series integral ultrafine particle (UFP) data from one test, and (b) solving for the integral UFP deposition loss rate constant using Equation 4

Integral UFP and $PM_{2.5}$ infiltration factors

Results for estimates of infiltration factors for integral measures of UFPs and $PM_{2.5}$ based on the underlying size-resolved infiltration factors for the 11 replicate tests are shown in Figure 3, along with $PM_{2.5}$ infiltration factors measured using the TSI DustTrak during the same periods. The mean (±s.d.) infiltration factor for UFPs, $PM_{2.5}$ estimated assuming constant unit density, $PM_{2.5}$ estimated assuming varying density, and $PM_{2.5}$ measured with the DustTrak averaged over all 11 replicate tests were 0.39 ± 0.05, 0.49 ± 0.09, 0.49 ± 0.08, and 0.43 ± 0.18, respectively. Values of F_{inf} for UFPs were relatively tightly distributed. Values for F_{inf} for $PM_{2.5}$ estimated using the two different density assumptions were not significantly different (also see Figure S1a in Supporting Information), suggesting that the $PM_{2.5}$ infiltration factor estimates using the SMPS + OPS system were not sensitive to assumptions for particle density. The mean and median $PM_{2.5}$ infiltration factors measured using the DustTrak were slightly lower than those estimated using the SMPS + OPS combination with either assumption for density, but they were also more widely distributed and sometimes did not give reasonable values due to low indoor concentration. For example, results from tests conducted on May 18 and June 3 were discarded because the DustTrak read zero indoors although the SMPS + OPS system yielded nonzero estimates of $PM_{2.5}$ mass concentrations. Comparisons between the SMPS + OPS system and the DustTrak measurements are further explored in the SI.

Size-resolved penetration factors and deposition loss rate constants

Figure 4 shows estimates of size-resolved penetration factors (Figure 4a), and deposition loss rate constants (Figure 4b) made using the resulting data for the first 13 particle size bins from 0.01 to 1 μm in mobility and optical diameter, respectively. Only, estimates of P and k for sub-1-μm particles are shown because the number concentrations measured by the OPS in all size bins above 1 μm were too low (and with too much scatter) to yield meaningful estimates of k for those bins (i.e., R^2 was consistently less than 0.5 using Equation 4). However, the steady-state I/O concentrations in these bins were high enough to calculate F_{inf}, as was shown in Figure 2. We should also note that estimates of P and k for integral $PM_{2.5}$ mass should not be greatly influenced by this failure to estimate size-resolved P and k for particle sizes greater than 1 μm because the estimated mass in these size bins was still added to the integral $PM_{2.5}$ mass concentrations.

Size-resolved envelope penetration factors generally increased with particle size for UFPs and decreased with particle size for particles larger than 0.3 μm. This is consistent with particle penetration theory, because larger particles are more readily removed in cracks by gravitational settling and smaller particles are more readily removed by Brownian diffusion (Liu and Nazaroff, 2001, 2003). The overall minimum and maximum bounds of the average (± s.d.) penetration factors across the 11 replicate tests were 0.41 ± 0.14 and 0.73 ± 0.05 at geometric mean particle diameters of 15 and 87 nm, respectively (Figure 4a). These values were also reasonably consistent with the few prior measurements of size-resolved penetration factors of which we are aware (Chen and Zhao, 2011). Similarly, size-resolved deposition loss rate constants demonstrated a characteristic U-shaped curve, generally decreasing with increasing particle size for UFPs and increasing with increasing particle size above 0.3 μm. Mean (±s.d.) estimates of deposition loss rate constants in this apartment unit with sparse furnishings and no central HVAC systems operating ranged from a maximum of 1.58 ± 0.34 1/h for 11 nm particles to a minimum of 0.11 ± 0.07 1/h for 86 nm particles (Figure 4b).

Estimates of the absolute and relative uncertainty in these size-resolved parameter estimates are shown in Figure S2 in Supporting Information. The mean estimate of the absolute uncertainty in P across all size bins and test replicates was approximately 0.10 (Figure S2a in Supporting Information), or approximately 17% on a relative basis (Figure S2c in Supporting Information). Estimates of absolute uncertainty in P were reasonably similar across all particle sizes, while relative uncertainty varied due to varying P values. These uncertainty estimates are comparable to those in Rim et al. (2010) and substantially lower than those observed in most other previous measurements of size-resolved penetration factors (e.g., figure 6 in Chen

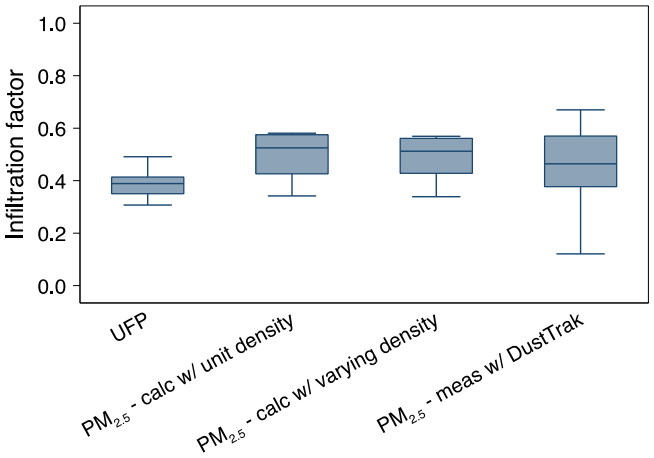

Fig. 3 Infiltration factors estimated for integral measures of ultrafine particles (UFPs), mass concentrations of particle less than 2.5 μm ($PM_{2.5}$) assuming unit density, $PM_{2.5}$ assuming varying density made using the size-resolved instrumentation system, as well as $PM_{2.5}$ measured with the TSI DustTrak (Shoreview, MN, USA). Boxes represent 25th, 50th, and 75th percentiles. Whiskers represent upper and lower adjacent values (1.5 times the differences between 25th and 75th percentiles)

and Zhao, 2011). Estimates of uncertainty in k were somewhat higher, with a mean estimate of absolute uncertainty of 0.20 (Figure S2b in Supporting Information), or approximately 44% on a relative basis (Figure S2d in Supporting Information), largely because of high uncertainties in the larger particle size bins in which the number concentrations were too low to yield clean decay curves. Although our estimates of P have inherent uncertainty in k built-in, we relied on the uncertainty in the total loss rate (λ+k) to estimate uncertainty in P, which had much lower relative standard errors.

Integral UFP and $PM_{2.5}$ penetration factors and deposition loss rate constants

Figure 5 shows distributions of the estimates of (i) penetration factors and (ii) deposition loss rate constants for integral measures of UFPs and $PM_{2.5}$ across the 11 replicate tests in the apartment unit, along with distributions of their estimated uncertainties. Because estimates of $PM_{2.5}$ infiltration factors made using either assumption for particle density were closely correlated (Figure S1 in Supporting Information), here we use only those estimates made assuming constant unity density, primarily for simplicity. The mean (±s.d.) estimates of P for integral measures of UFPs and $PM_{2.5}$ were 0.67 ± 0.05 and 0.73 ± 0.05, respectively, ranging from 0.59 to 0.78 for UFPs and from 0.65 to 0.79 for $PM_{2.5}$ (Figure 5a). Estimates of k were much more widely distributed, with a mean (±s.d.) estimate of 0.68 ± 0.27 1/h for UFPs and 0.45 ± 0.30 1/h for $PM_{2.5}$. The mean (±s.d.) estimates of the relative uncertainty in P for $PM_{2.5}$ and UFPs across the 11 replicate tests were 12 ± 5% and 11 ± 4%, respectively. The mean (±s.d.) estimates of the relative uncertainty in k for $PM_{2.5}$ and UFPs across the 11 replicate tests were 12 ± 7% and 13 ± 6%, respectively.

For comparison of values in the literature, estimates of integral UFP penetration factors were on the upper

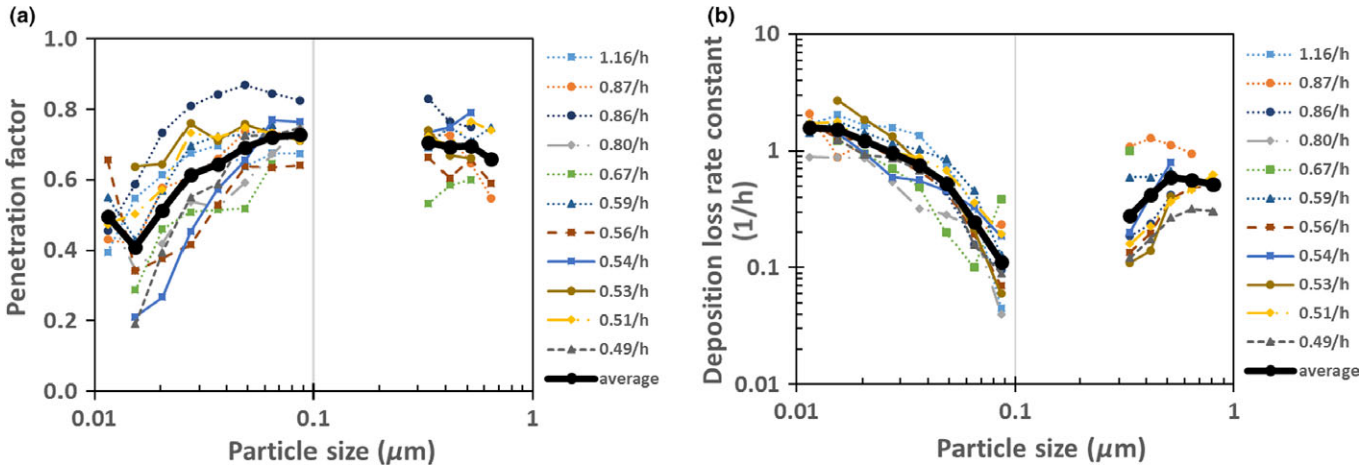

Fig. 4 Size-resolved (a) penetration factors, and (b) deposition loss rate constants estimated across 11 replicate tests with varying air exchange rates for particles 0.01–1 μm in mobility and optical diameter, respectively

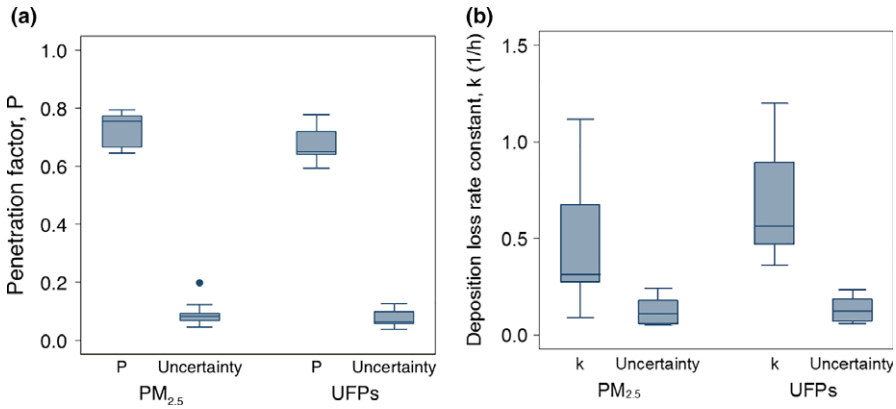

Fig. 5 Estimates of (a) penetration factors (P) and (b) deposition loss rate constants (k) for integral measures of ultrafine particles (UFPs) and mass concentrations of particles less than 2.5 μm ($PM_{2.5}$) with associated uncertainties made across 11 replicate tests in the apartment unit

end of those measured using non-size-resolved instrumentation in a previous study of 18 homes in Texas with widely varying vintages (Stephens and Siegel, 2012). Estimates of $PM_{2.5}$ penetration factors were similar to the average estimates of P from gravimetric $PM_{2.5}$ measurements in 40 homes in North Carolina (Williams et al., 2003). Our measured deposition loss rate constants for both UFPs and $PM_{2.5}$ were somewhat lower than in these same previous studies and others, likely because we conducted experiments in a sparsely furnished apartment with a low surface-area-to-volume ratio.

We should note that we also estimated P for total UFPs using the underlying size-resolved estimates of P from each of the eight UFP size bins using Equation S1 in Supporting Information, in which we weighted the size-resolved estimate of P in each bin by the average contribution of that bin to the total outdoor UFP concentration. In theory, this method would avoid any bias in parameter estimates introduced by the integral UFP measures. We then compared these results to estimates of P using the simpler integral measure of UFPs (i.e., from Figure 5). While the full procedure and comparison is shown in the SI (and Figure S3 in Supporting Information), the simpler integral method gave slightly lower estimates for P for total UFPs than the weighted-size-resolved method (~7% on a relative basis). However, the weighted-size-resolved estimation procedure yielded much higher uncertainties for each replicate test than the integral UFP estimation procedure (Figure S3b in Supporting Information), such that results from the integral UFP measures were always within the uncertainty of the weighted-size-resolved measures. Therefore, we consider it most appropriate to utilize the integral UFP measures to estimate P and k from the size-resolved data collected using this test method. We assume the same holds true for the integral $PM_{2.5}$ measures as well, although we were not able to make the same comparison between integral and size-resolved $PM_{2.5}$ parameter estimates because the large amount of scatter in the concentration data for size bins greater than 1 μm prevented reasonable estimates of k and thus prevented estimates of P (i.e., Figure 4).

Influence of indoor and outdoor environmental factors

Results of the estimates of P and k with uncertainties for integral UFP and $PM_{2.5}$ measures from all 11 replicate penetration tests are shown in Table S1 in Supporting Information, along with a number of coincident indoor and outdoor environmental conditions that could plausibly influence penetration factors or deposition loss rate constants, including data for indoor and outdoor temperature and relative humidity and wind speed and direction. Spearman's rank correlations between P and all indoor and outdoor environmental conditions reported in Table S1 revealed no significant correlations ($P > 0.01$), suggesting that the test procedure and solution methods proposed herein for estimating the penetration factor for both UFPs and $PM_{2.5}$ were not greatly affected by the observed ranges of environmental conditions. However, estimates of deposition loss rate coefficients (k) for both UFPs and $PM_{2.5}$ were significantly (and negatively) correlated with the geometric mean diameter of the average outdoor particle size distribution, with Spearman's rank correlation coefficients of -0.74 ($P = 0.010$) and -0.85 ($P = 0.008$) for UFPs and $PM_{2.5}$, respectively (see Figure S4 in Supporting Information). This is intuitive, as outdoor particle size distributions that infiltrate indoors with a larger geometric mean diameter (i.e., closer to 0.1 μm than 0.01 μm) would be expected to have a lower deposition loss rate coefficient according to deposition theory.

Applications and limitations

Here, we have developed and applied a novel test method for measuring the penetration of outdoor fine and ultrafine particles into indoor residential environments. The method is designed to minimize the duration of testing without sacrificing accuracy and to provide a practical solution for further application in field measurements in a greater number and variety of buildings. We were able to achieve measurements of both P and k for many size-resolved particle size bins and integral measures of UFPs and $PM_{2.5}$ within a relatively short test duration of approximately 4 h, which provides a practical approach for future field experiments. However, we should also note that there are some limitations with this method. First, estimates of both P and k rely on accurate measurements of the air exchange rate. Failure to achieve accurate AER measurements during the test period can occur due to changing weather conditions, mixing issues, or having additional sources of the tracer gas (in our case CO_2) present. Second, the initial process of introducing outdoor particles can be challenging when outdoor particle concentrations are low, leading to losses of data in some size bins. Third, using integral UFP and $PM_{2.5}$ measures can introduce a (likely small) bias in the results compared to using size-resolved parameter estimates to reconstruct UFP and $PM_{2.5}$ parameter estimates, but practical limitations necessitate this approach. Finally, the method does not explicitly consider evaporative losses that may occur for some aerosol constituents, nor can it distinguish between different aerosol compositions using the instrumentation. Future work should further test the application of the method in the field and also work to integrate real-time composition measurements (e.g., with an aerosol mass spectrometer).

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

Figure S1. Regression analysis between (a) PM2.5 infiltration factors calculated using two different density assumptions and (b) PM2.5 infiltration factors calculated assuming constant unit density and measured using a TSI DustTrak.

Figure S2. Size-resolved (a) absolute and (c) relative uncertainties for penetration factors, and (b) absolute and (d) relative uncertainties for deposition loss rate constants estimated across 11 replicate tests with varying AERs for particles 0.01 μm to 1 μm in mobility and optical diameter, respectively, using data from both the SMPS and OPS.

Figure S3. (a) Regression analysis for UFP penetration factors between integral UFP and weighted-size-resolved UFP measures (b) penetration factors and uncertainties estimated using both UFP aggregation measures across 11 replicate tests.

Figure S4. Regression analysis between geometric mean of outdoor particle size and (a) deposition loss rate constants (k) and (b) penetration factors (P) for UFP and PM2.5.

Table S1. Summary of estimates of P and k for integral measures of UFPs and PM2.5 with uncertainties and indoor and outdoor environmental conditions.

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