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A Concentration Rebound Method For Measuring Particle Penetration And Deposition In The Indoor Environment

Tracy L. Thatcher*, Melissa M. Lunden, Kenneth L. Revzan, Richard G. Sextro,
and Nancy J. Brown

Lawrence Berkeley National Laboratory
Environmental Energy Technologies Division
1 Cyclotron Rd
Berkeley, CA 94720

* corresponding author, phone (510) 486-5215

FAX (510) 486-6658

e-mail tlthatcher@lbl.gov

mailstop 90R3058

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ABSTRACT

Continuous, size resolved particle measurements were performed in two houses in order to determine size-dependent particle penetration and deposition in the indoor environment. The experiments consisted of three parts: 1) measurement of the particle loss rate following artificial elevation of indoor particle concentrations, 2) rapid reduction in particle concentration through induced ventilation by pressurization of the houses with HEPA-filtered air, and 3) measurement of the particle concentration rebound after house pressurization stopped. During the particle concentration decay period, when indoor concentrations are very high, losses due to deposition are large compared to gains due to particle infiltration. During the concentration rebound period, the opposite is true. The large variation in indoor concentration allows the effects of penetration and deposition losses to be separated by the transient, two-parameter model we employed to analyze the data. We found penetration factors between 0.3 and 1 and deposition loss rates between 0.1 and 5 h⁻¹, for particles between 0.1 and 10 μm.

INTRODUCTION

Particulate air pollution is associated with increased morbidity and mortality even at the generally low levels of air pollution in United States cities (Dockery et al., 1993, Pope et al. 1995, Samet et al., 2000). The exact compounds and/or particle size ranges responsible for these health effects have not yet been determined. The indoor environment provides a significant, if not dominant, exposure potential for particles for two primary reasons. First, people spend most of their time indoors – typically ~90 % (Jenkins et al., 1992, Robinson and Nelson, 1995).

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Second, indoor concentrations of particles of outdoor origin are estimated to be on the same order as outdoor concentrations (Wallace, 1996; Ott et al., 2000; Riley et al., 2001). Indoor concentrations of particles of outdoor origin are influenced by many building and environmental factors, such as air leakage rates and ventilation system design. Thatcher et al. (2001) identified and evaluated sources of data for those factors that affect the transport to and concentration of outdoor particles in the indoor environment. In addition to particles of outdoor origin, particles generated from indoor sources, such as tobacco smoke, cooking fumes, or pet dander, may present significant specific health concerns.

Particle deposition within the home will reduce indoor airborne concentrations of particles with both indoor and outdoor origins. For this reason, understanding deposition loss rates under typical residential conditions is important for assessing human health impacts from indoor particles. Many experiments have been performed to study particle deposition in the indoor environment (Offermann et al, 1985; Xu et al., 1994; Byrne et al., 1995; Thatcher and Layton, 1995; Fogh et al., 1997; Abt et al., 2000; Long, et al., 2000; Vette et al., 2001; Mosley et al., 2001; and Thatcher et al., 2002). Results from these studies show a wide degree of variability in deposition rate for any given particle size. This variability is due, at least in part, to variations in the conditions under which deposition rates were measured. Factors such as airflow conditions, quantity and nature of furnishings, interior surface-to-volume ratio, surface-to-air temperature differences, particle type, and measurement method may all be expected to influence the measured deposition rate.

Another key factor influencing indoor exposures to particles of outdoor origin is the effect of losses due to particle filtration by the building shell. These losses are typically quantified by the

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use of a penetration factor, P , defined as the fraction of particles in the infiltrating air that pass through the building shell. Previous experiments on penetration factors have found a large variation in values. Thatcher and Layton (1995) measured particles as a function of size and found penetration factors near 1 for particles with diameters larger than $1\ \mu\text{m}$ for the single residence studied. These results suggested that the shell of the building studied provides essentially no filtration for these particles. Wallace (1996) also calculated penetration factors very close to 1 for PM 2.5 and PM 10, based on the particle mass data from the EPA PTEAM study for a large number of households in the Los Angeles area. Cristy and Chester (1981) generated large quantities of $2\text{-}\mu\text{m}$ -diameter spores outside a trailer home, which are often poorly sealed structures, and measured the indoor concentration response. They calculated that penetration losses did not have a significant effect on indoor concentrations. Vette et al. (2001) reported penetration factors for a single house between 0.4 and 0.9 for ambient particles with diameters between 0.01 and $2.5\ \mu\text{m}$. However, they did not measure air exchange rates during the period for which the penetration rates were calculated. Abt et al. (2000) calculated a factor they termed the ‘effective penetration efficiency’, which combines both deposition and penetration losses for outdoor particles. However, their analysis did not separate the two factors. Long, et al. (2001) described an ‘infiltration factor’ which was equivalent to the ‘effective penetration efficiency’ discussed above, where the effects of deposition and penetration losses are not separated. They also determined values for deposition and penetration losses using a random effects mixed model on data from nine homes and reported penetration factors between about 0.9 and 0.3 for particles between 0.02 and $6\ \mu\text{m}$. However, their report of penetration values as low as 0.53 for a home where the “windows and doors were predominantly left open”

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seems physically unreasonable and casts doubt upon this model based method. An additional factor that can complicate interpretation of results is the use of ambient outdoor particles that are not chemically resolved and may undergo unknown reactions and/or transformations in the indoor environment. Thatcher and Layton (1995), Wallace (1996), and Vette et al. (2001) all utilized measurements of ambient particle in their experiments.

Roed and Cannell (1987) reported $P = 1$ for two radioactive isotopes (^{131}I and ^7Be) and $P = 0.53$ for a third (^{137}Cs), all assumed to be bound to particles, based on measurements in a single house. Koutrakis et al. (1992) measured PM 2.5 in 394 homes and estimated penetration factors between 0.58 and 1.04 for 8 elements primarily of outdoor origin. Their calculations assumed that all 8 elements had an average deposition velocity of 0.18 m/h – in effect, that the elements were all associated with the same size particle. If the elements were truly associated with the same particle size, then the reason for the differences in penetration factors between elements is unclear. If they are associated with different particle sizes, then the assumption of a common deposition rate, independent of particle size, will lead to large errors in the calculation of penetration factors.

Chao and Tung (2001) report $P = 0.85$ based on measurements of PM 2.5 in five homes. Their analysis assumed that indoor deposition losses (β) were negligible, which leads to errors in the value of P since the factors that affect β and P are similar. Tung et al. (1999) measured concentrations of PM 10 in an interior conference room and corridor of an office building under conditions with the HVAC off and calculated penetration factors for transport from the corridor into the adjacent room ranging from 0.69 to 0.86. Their data show that a large increase in the particle concentration in the corridor did not cause any response in the conference room

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concentration, which they attribute to the low air exchange rate (0.29 h^{-1}) between the corridor and room. However, if a transient model is constructed using their parameters, the modeled room concentration shows a substantial response to the increase in the corridor particle concentration.

A possible explanation for this discrepancy is that some other pathway, such as the HVAC ductwork, duct leaks, or the ceiling plenum, was the main source of particle-bearing air infiltrating into the conference room, not the corridor.

McMurry et al. (1985) measured indoor/outdoor (I/O) ratios in a well-sealed residence, with no known indoor sources and found no correspondence between these ratios and particle diameter for particles between 0.1 and 1 μm . In developing their approach and analysis, they assumed that the indoor and outdoor concentrations were at steady state, even though the low air exchange rates and correspondingly long residence times in this study make steady state conditions more difficult to achieve. The potential impact of assuming steady state when the system is transient can be seen in their data during a rainfall episode where the I/O ratios rise dramatically for all size ranges due to a sharp drop in outdoor concentrations. These I/O ratios decline rapidly once the rainfall episode is concluded and outdoor concentrations rebound. The changes in the I/O ratio over this period are probably influenced more by the time lag between the indoor and outdoor concentrations than by changes in the physical processes influencing indoor concentrations.

In addition to the whole house studies listed above, several studies have investigated penetration through manufactured cracks in experimental chambers. Lewis (1995) reported penetration factors between 0.97 and 0.29 for particles between 1 and 6 μm passing through a Perspex (plastic) slit 0.1 mm high and 40 mm wide with a pressure differential of 10 Pa across

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the slit. Mosely et al. (2001) passed monodispersed particles through manufactured aluminum slits 0.508 mm high and 10 cm wide. They found penetration factors between 0.02 and 0.9 for 2 μm particles and 0.001 and 0.05 for 5 μm particles, with a strong dependence on the pressure differential across the crack.

In general, investigators conducting chamber studies have reported lower penetration rates than those performing whole house studies. The reasons for this are not entirely clear. However, it is reasonable that the penetration factor should be highly influenced by the size and geometry of the infiltration route, which has not been systematically investigated for a wide variety of houses. Liu and Nazaroff (2002) performed experiments measuring penetration factors as functions of particle size, crack height, pressure drop across the crack, and crack material. They found that the height of the crack significantly influenced the penetration factor. For example, they found that for 2 μm particles traveling through a 9.4 cm long smooth crack, essentially no particles deposited in a crack 1 mm high and nearly all particles deposited in a crack 0.25 mm high. Older homes, and other homes which are not tightly sealed, may have a significant portion of their infiltrating air entering through openings around pipes and electrical outlets, poorly sealed windows, and other pathways with relatively large dimensions. The ASHRAE Fundamentals Handbook (1997) reports that a large fraction of air infiltrating into a residence can come from large openings such as those around fireplace dampers (0 to 30%) or in the heating system (3 to 28%). When large pathways are prominent, penetration factors would be expected to be close to unity. In a home without larger openings, where most of the air enters through smaller cracks, the observed penetration rates may approach those found in chamber studies.

Separating the effects of deposition and penetration in a full-scale house is difficult. Often

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assumptions are made with respect to either the deposition rate or penetration factor and the resulting parameter is then used to calculate the other factor. Alternatively, both Thatcher and Layton (1995) and Vette et al. (2001) first determined the deposition rate by elevating the concentration, measuring the particle loss rate as a function of particle size, and subtracting the exfiltration rate. The measured deposition rates were then used along with ambient measurements (with resuspension minimized) to determine the size dependent penetration rate. Using this method, any changes in the deposition loss rate or variability in outdoor particle concentrations at 'steady state' will lead to inaccuracies in the calculated penetration factor.

In the present study, we vary the indoor particle concentration over a wide range, examining cases where deposition is the dominant loss mechanism and cases where penetration losses dominate. We use a transient model to determine the combination of deposition rate and penetration factor that best fits the observed temporal data. The effect of losses due to ammonium nitrate phase changes will also be investigated and discussed. This study develops a methodology that estimates deposition and penetration losses simultaneously, without relying on an assumption of steady state. The methodology is then utilized for evaluating the loss terms in two separate buildings, with different construction details and levels of air tightness. This provides information on the potential importance of building type on penetration factors and deposition loss rates.

EXPERIMENTAL PROTOCOL

Since both deposition and penetration losses are particle size dependent and occur simultaneously, it is difficult to decouple the effects of these processes in a residence. In our experimental design the indoor concentration is varied over a wide range and we analyze events

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where deposition is the dominant loss mechanism and events where penetration losses dominate. A transient model employing a two-parameter fit is used to determine the combination of deposition rate and penetration factor that best fits the observed data.

For this study we conducted experiments in two buildings, one in Clovis, and the other in Richmond, California. At both sites, a typical experiment began with a short period of resuspension activities followed by a 1 to 2 hour period of concentration decay. For some experiments at the Clovis house, a gas-stove burner was also ignited to increase the concentration of smaller particles. The buildings were then pressurized using a High Efficiency Particle Absolute (HEPA) filter mounted in a window. The HEPA filter supplies the building with essentially particle free air (greater than 99.99% removal of $0.3 \mu\text{m}$ particles) causing the indoor particle concentration to decline to nearly zero due to exfiltration from the building. The effectiveness of the pressurization was confirmed by measuring the indoor-outdoor pressure differential across the building surfaces at various points to determine whether infiltration was occurring at any location across the building shell. When the HEPA filter was then turned off, infiltration of particle laden outdoor air resumed, and the indoor particle concentration rebounded. During the entire experiment, tracer gas measurements were made continuously at several locations within the building. Size-differentiated particle concentrations were measured every 3 minutes with particle instruments located both inside and outside the building.

The resuspension process raises indoor concentrations of particles larger than $\sim 0.2 \mu\text{m}$ in diameter by causing particles that have been deposited or tracked onto surfaces to disperse in the indoor air. It is expected that these resuspended particles will be chemically stable in indoor air and will not undergo transformation processes. During the concentration decay period, indoor

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concentrations are relatively high and losses due to deposition will be large compared to gains due to particle infiltration. The deposition rates obtained during this period are stable and reproducible. During the concentration rebound period, the opposite will be true. Changes in indoor particle concentrations will be due almost exclusively to infiltrating outdoor particles and deposition effects will be small compared to infiltration. Thus we can effectively separate penetration and deposition losses. In some cases, chemical transformation can be an important confounder; we discuss this further below.

Study Locations

To explore the effect of building characteristics on deposition losses and penetration factors, we applied this method in two separate buildings, each with very different construction. Experiments were performed in two houses: one in Richmond, California and a second in Clovis, California. The Richmond facility is a small, older building (59 m²) with wood slat siding, un-insulated walls, and double-hung wooden windows. The building is located in a relatively unsheltered location near the San Francisco Bay and is subject to winds that exhibit a significant diurnal variation, as well as occasional high wind speeds. Figure 1 shows a floor plan of the facility. All particle measurement equipment was located in room 1 and two oscillating fans sitting on the floor were used to promote mixing, one located in room 1 and the other located in the hallway.

The Clovis facility is a moderate sized home (134 m²) constructed in 1972. It has a stucco exterior and single-glazed aluminum frame windows. The house is single story, with standard height ceilings (2.4 m), a forced air heating and cooling system (which was not operated during these experiments), and ceiling fans (which were operated during the experiments to promote

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mixing). An additional oscillating fan located approximately 1.5 m from the floor was operated in the living room to disperse tracer gas and promote mixing near the particle measurement equipment. The building is located in a residential suburb of Fresno, California, surrounded by mature trees and homes of a similar height and size. The flat terrain and high level of sheltering resulted in relatively low levels of wind loading near the building. Figure 2 shows a floor plan of this facility, along with the location of equipment.

Blower door measurements, in which the amount of airflow under various imposed pressure differentials is measured, are commonly used to compare the relative 'tightness' of homes (ASHRAE Standard 136). These measurements can be used to calculate normalized leakage (NL), which is the leakage area normalized by floor area and a house height factor. The average house in the United States has a NL of $1.2 \text{ cm}^2/\text{m}^2$ Sherman and Matson (1997). The typical NL of a new house is approximately $0.55 \text{ cm}^2/\text{m}^2$ and a new well-sealed, energy-efficient house is around $0.5 \text{ cm}^2/\text{m}^2$ (Sherman and Matson, 2002). Older homes tend to have significantly larger NL than newer homes (Sherman and Dickerhoff, 1998). The Richmond house has a normalized leakage area around $2.5 \text{ cm}^2/\text{m}^2$, indicating that the structure is fairly 'leaky' and that air penetrates the building easily. The Clovis house has a relatively low normalized leakage area of $0.65 \text{ cm}^2/\text{m}^2$, indicating that the building is better sealed and may have a smaller proportion of the air leakage occurring through large penetrations in the building shell.

Measurement Equipment

The indoor particle and gas measurement instruments were located in the living room of each building. Systems to measure tracer gas concentration and pressure differentials across the building shell monitored the living room as well as several locations throughout the house, as

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shown in Figures 1 and 2. An additional set of particle measurement devices was also located outdoors at each facility. In Richmond, the outdoor instruments were located on a table approximately 30 feet upwind of the house. In Clovis, the instruments were located in a shed adjacent to the house and samples were collected through a PM 10 inlet projecting approximately 10 feet above the shed roof.

Two aerodynamic particle counters (TSI, Model APS 3320), one located indoors and one outdoors, were used to measure size distributions for particles with aerodynamic diameters between 0.5 and 10 μm . To assure that the measurements from the two instruments were comparable, measurements with the instruments sitting side-by-side were performed before, during, and after each test series. The results were compared to determine a collection efficiency ratio between the instruments for each size bin. These ratios were used to adjust the concentrations obtained by the instruments and ‘match’ the results. For the Clovis experiments, the matching experiments were performed with the outdoor instrument sampling through the PM 10 inlet manifold so that manifold losses would be included in the correction factor. The APS uses 52 size bins between 0.5 and 20 μm . For our analysis, the smallest size bin and all size bins over 10 μm were eliminated, because of poor correlation between instruments and the use of the PM 10 inlet outdoors. Data in the remaining bins were grouped to produce 12 bins between 0.5 and 10 μm . The concentration in the largest of these bins was often very low indoors, particularly in the Clovis facility, which had low air exchange rates, measuring less than 10 particles over a 3-minute sample period. We excluded periods where there were insufficient counts to provide adequate count statistics from our analysis.

Two optical particle counters (Particle Measurement System, Model LAS-X), one located

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indoors and one outdoors, were used to measure size distributions for particles with optical diameters between 0.1 and 3 μm . The instruments were ‘matched’ in a manner analogous to that used for the aerodynamic particle counters. The smallest and largest size bins were excluded due to difficulty in matching and a low number of counts indoors for the larger sizes. Calibrations using an Electrostatic Classifier (TSI Inc, Model 3071) were performed with ambient particles to determine the appropriate conversion from optical to aerodynamic size. However, the conversion from optical to aerodynamic diameter is highly dependant on particle composition and varies over time. As a consequence, variations in the chemical composition of the aerosol, such as high levels of carbonaceous aerosol, will lead to errors in the conversion from optical to aerodynamic diameter. As a result, we report our concentrations in terms of optical diameter for measurements performed with the optical counters. The optical diameter is not necessarily expected to be identical to the aerodynamic diameter for the particles studied.

In Clovis, CA, a substantial and highly variable fraction of the ambient aerosol can be composed of ammonium nitrate particles. These particles have been shown to dissociate into ammonia and nitric acid in the indoor environment (Lunden et al. 2002) and therefore cannot be readily modeled with a simple physical model that assumes that the particles are conserved. To determine the contribution of ammonium nitrate particles to the outdoor aerosol during these experiments, PM_{2.5} nitrate, carbon and sulfate were measured with 10-minute resolution using the integrated collection and vaporization method of Stolzenburg and Hering (2001). This method collects PM_{2.5} particulate matter by humidification and impaction onto a 1 mm diameter spot on a metal substrate. The sample is then analyzed by flash-vaporization and quantitation of the evolved vapor compounds. Nitrate concentrations are measured using low-temperature

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vaporization in a nitrogen carrier gas with quantitation of the evolved vapors using a chemiluminescent monitor equipped with a molybdenum converter to reduce higher oxides of nitrogen to nitric oxide. Sulfate and carbon analyses are performed using high-temperature heating, with analysis of the evolved sulfur dioxide by UV-fluorescence and carbon dioxide by nondispersive infrared absorption.

Indoor and outdoor measurements were performed simultaneously using a four-cell system. One pair of cells (one indoors and one outdoors) was used for nitrate measurements. A second pair was used for the combined measurement of carbon and sulfate. The outdoor nitrate cell and outdoor sulfate-carbon cell were housed indoors inside a box that was ventilated with outdoor air to maintain near-outdoor temperature at the point of sample collection. A more detailed description of this system can be found in (Lunden et al., 2002).

Air infiltration rates were measured using sulfur hexafluoride tracer gas techniques with a photo-acoustic infrared detector system (Bruel and Kjaer, Model 1312). For the Richmond experiments, an injection/decay method was used. In this method, a pulse of tracer gas is injected and mixed within the space. The loss of tracer gas over time is then monitored and the concentration decay rate is used to determine the infiltration rate. During the Clovis experiments, a constant injection system was used. In this method, tracer gas is injected at a constant rate before and during the experiment. The infiltration rate is calculated using a transient mass balance approach that accounts for the effects of injection rate, infiltration losses, and changes in indoor concentration. An automated, multiple channel differential pressure system (Energy Conservatory, APT 8) was used to monitor pressure differentials across the building shell at various points (see figure 2) and to assure that pressurization with the HEPA

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filtration system was always positive (inside with respect to outside) on all surfaces of the building envelope.

Analysis Method

The concentration of particles indoors is a balance between the sources and sinks of particles in the indoor environment. Figure 3 illustrates potentially important factors affecting indoor concentrations. In the most general form, the indoor concentration of particles of a specific size and composition particle can be represented by the following equation.

$$\frac{\partial C_I}{\partial t} = (C_o P - C_I) \lambda_v - C_I \beta + G + S + F + K + H \quad (1)$$

Where: C_I = indoor particle concentration at time t ($\# \text{ cm}^{-3}$),
 t = time (h^{-1}),
 C_o = outdoor particle concentration at time t ($\# \text{ cm}^{-3}$),
 P = penetration factor,
 λ_v = air exchange rate (h^{-1}),
 β = deposition loss rate (h^{-1}),
 G = generation of particles indoors ($\# \text{ cm}^{-3} \text{ h}^{-1}$),
 S = particle formation through gas-particle conversion ($\# \text{ cm}^{-3} \text{ h}^{-1}$),
 F = particle formation due to reaction ($\# \text{ cm}^{-3} \text{ h}^{-1}$),
 K = particle size change through coagulation ($\# \text{ cm}^{-3} \text{ h}^{-1}$), and
 H = particle size change through hygroscopic growth ($\# \text{ cm}^{-3} \text{ h}^{-1}$).

For the conditions and particle size ranges used in these experiments, we do not expect

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coagulation, hygroscopic growth, or formation to have a significant impact on indoor particle concentrations. In addition, care was taken to avoid indoor sources by using an unoccupied space and operating equipment that could generate particles, such as pumps, outdoors. Therefore, we assume that these effects are negligible in our analysis. During some experiments, observations indicated that the results were complicated by dissociation and vaporization of ammonium nitrate particles. Periods of high ammonium nitrate were excluded from our results. For the periods selected for our analysis, we could reduce the mass balance equation to:

$$\frac{\partial C_I}{\partial t} = C_o P \lambda_v - C_I (\lambda_v + \beta) \quad (2)$$

Under steady-state conditions, this equation is solved easily for a constant outdoor concentration and stable infiltration rate. However, in most real-world situations – as was the case for these experiments – there is significant time variation in C_o and λ_v causing the steady-state solution to be invalid. When $\partial C_I / \partial t$ is not zero, the equation can be solved using a basic ‘forward-marching’ scheme with time step, Δt , as shown below:

$$C_I(t_2) = C_I(t_1) + P C_o(t_1) \lambda_v \Delta t - C_I(t_1) (\lambda_v + \beta) \Delta t \quad (3)$$

In these experiments, the time step used was 3 minutes, corresponding to the measurement interval for the size resolved instruments.

The equation solving methodology employed the minimization of the sum of the square of the differences between the measured and modeled indoor concentration at each time step

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divided by the measured concentration at that time. Since the concentration varied over several orders-of-magnitude during each experiment (from a maximum after resuspension to a minimum during the filtration period), dividing by the measured concentration normalized the weighting of each point over the entire experiment. Although this leads to biases due to measurement errors, it was found to provide better fits over the entire data range than produced by other weighting schemes. For instance, without weighting the high concentrations periods dominated and the curve fit at lower concentrations was poor. A weighting scheme minimizing the inverse of the measured/modeled difference led to large errors at high concentrations. Individual measurements which appeared to be inaccurate due to either equipment limitations or instabilities were excluded from the analysis if they exhibited the following characteristics: (1) zero or near zero counts or (2) one or more points where counts differ by more than 50% from the counts in both the preceding and following measurements in the time series, with no physical explanation. In general, the excluded measurements represented less than 1% of the total number of points, with the exception of the largest size channel of each instrument, which exhibited very low particle counts over significant periods.

The Excel spreadsheet tool SOLVE (Microsoft Corporation) was used to determine the deposition loss rate, β , and penetration factor, P , which minimized the sum of the weighting function residuals for each particle size range. Due to bias introduced by weighting the model/measurement error by the measured concentration, the average modeled concentration tended to be slightly lower than the measured average concentration. This difference was typically less than 5%. In consideration of this bias, the goodness-of-fit was determined using two measures: the first is the measured/modeled correlation coefficient and the second is the

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difference between the measured and modeled average concentration. If the correlation coefficient was less than 0.95 or the difference between the average concentrations was more than 10% for a time series reconstruction for a given particle size range, then the fit was rejected and the results were discarded. Typically the model performed very well and few rejections were observed, for example, in the Richmond experiments, 12 of the 122 reconstructions were rejected, and only 3 of the 81 reconstructions were rejected in the Clovis data. One experiment performed at the Clovis facility was discarded due to problems associated with interference by ammonium nitrate, which will be discussed in more detail subsequently. For each particle size range, the deposition loss rates and penetration factors obtained from the reconstructions were averaged over all experiments performed at each facility. The average and standard deviation are reported for each site.

RESULTS AND DISCUSSION

The penetration rebound method produces a characteristic indoor particle profile as shown in Figure 4 for smaller particle sizes measured with the optical particle counter and Figure 5 for larger particles measured with the aerodynamic particle counter. Immediately following resuspension, indoor concentrations are significantly higher than during quiescent periods, as seen at the end of the experiment, for all but the smallest particle sizes. Similarly, during the period when the house was pressurized with filtered air, indoor concentrations are significantly lower than quiescent concentrations. This large range of indoor concentrations increases the sensitivity of the method and improves the ability to find unique solutions for the penetration factor and deposition rate. Changes in the outdoor concentration during the experiment also increase the sensitivity of the fit since deposition losses are a function of indoor concentration

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and penetration losses are a function of outdoor concentrations. Outdoor concentration variability help differentiate the effects of penetration and deposition losses. Significant changes in outdoor concentration occur over the course of one or more hours on a typical day, but they can occur more rapidly, for instance when the wind direction shifts. For particle sizes near the larger end of an instrument's range, low indoor particle counts often lead to poor count statistics and a high degree of sample-to-sample variability especially during the rebound phase or when air exchange rates are low. This variability can lead to a decrease in the sensitivity of the fit of the model to the data. Figures 4 and 5 illustrate the type of model fit achieved during the experiment. The model fits well on both the downward sloping deposition phase and on the upward sloping rebound phase, indicating that both deposition and penetration losses are appropriately described.

The sensitivity of the model fit to the penetration factor value can be seen in Figure 5. This example uses APS data from the Richmond experiments with the model constrained to a penetration factor arbitrarily fixed at 0.5 leaving the deposition rate as the sole fitting parameter. In this example where the penetration factor is lower than the 'best-fit' penetration factor, the solver reduces the modeled deposition rate to compensate for the artificially low penetration rate. This results in an over-prediction of the indoor concentration in the initial period just following resuspension, when deposition is the dominant loss mechanism. Later in the experiment, when indoor concentrations are lower, the effect is reversed, with the model under-predicting indoor concentrations. If the penetration factor were fixed above the 'best-fit' value, the opposite would be observed. These results demonstrate that constraining the penetration rate creates a situation where it is not possible to adequately fit both the concentration decay and rebound portions of an

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experiment.

At the Clovis site, the effects of ammonium nitrate vaporization indoors confounded the effects of penetration factor and deposition rate on indoor concentrations. Figures 6 and 7 illustrate this effect. As shown in Figure 6, the outdoor ammonium nitrate concentrations are low and the modeled particle concentrations match the measured particle concentrations well early in the experiment. Later, when the outdoor nitrate concentrations rise rapidly, the outdoor particle counts show a similar rise for particles in size bins between 0.3 and 1 μm , but exhibit little effect for larger or smaller particles. This suggests that outdoor ammonium nitrate particles have diameters primarily between 0.3 and 1 μm . The model predicts that indoor particle concentrations will increase substantially when the outdoor particle concentrations increase, as expected from equations 2 and 3. However, indoor measurements of indoor particle nitrate concentration and indoor particle concentrations do not show corresponding responses. This indicates that deposition and penetration losses are not the only processes affecting the indoor particle concentrations at this site during times of high outdoor ammonium nitrate concentrations. As a consequence, a simple chemical-conservative model cannot be used to represent adequately the indoor particle concentration during these periods. Methods used for assessing losses of particle nitrate and conversion to gaseous ammonia and nitric acid indoors are described in more detail by Lunden et al (2002) and Fischer et al (2002).

Determining the size distribution for ammonium nitrate and subtracting the nitrate particles from the size-resolved particle concentrations both indoors and outdoors could remove the effect of nitrate transformation. Since our time-resolved nitrate measurements are not size-resolved, this method would require a very stable nitrate size distribution, which did not occur during this

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study. Figure 7 shows indoor and outdoor particle concentrations for selected size ranges and PM_{2.5} nitrate concentrations during a period when no rebound experiments occurred. Initially the outdoor PM_{2.5} nitrate concentration is low and stable. Between approximately 0900 and 1200, a nitrate peak occurs outdoors. The outdoor particle size distributions for particles with diameters less than about a micron exhibit peaks during the nitrate episode. The response appears first in the 0.15 to 0.2 μm size bin and shifts to larger size ranges as the episode develops. By the time the outdoor PM_{2.5} nitrate concentration reaches a maximum, the peak is seen predominantly in the 0.65 to 0.80 μm size bin. Based on the response seen in the outdoor size distribution, the average size of the PM_{2.5} nitrate particles appears to increase as the nitrate peak episode progresses. These sizes are consistent with the bimodal ambient nitrate distribution reported by John et al (1990). The shifting nitrate size distribution makes it difficult to remove the effect of nitrate evaporation in the absence of size resolved nitrate measurements. The indoor size distributions indicate no corresponding increase in concentration during the nitrate peak episode.

Due to the difficulties caused by ammonium nitrate vaporization indoors, we chose to calculate penetration factors and deposition rates for the Clovis house using data only from those portions of the experiments where the outdoor concentrations of ammonium nitrate were low and relatively stable. Despite this constraint, ammonium nitrate may still exhibit a somewhat confounding effect on the results because overall particle concentrations were also low during these periods. This may result in artificially low penetration rate predictions for submicron particles in the Clovis experiments. Since the indoor particles during the deposition portion of the experiments consist mainly of resuspended and/or generated particles, calculated deposition

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rates are less likely to be affected by ammonium nitrate losses.

The best fit parameters and standard deviations for penetration factor and deposition loss rate at various particle sizes are listed in Table 1. Figures 8 and 9 compare the deposition loss rates and penetration factors obtained at the two site. The optical diameter values for the Richmond site represent the best fit achieved by applying the two parameter transient model to the data for five experiments conducted during March and April 2000. The aerodynamic data are based on only four of these experiments because the equipment failed during one of the experiments. The values for the Clovis site represent the best fit for three experiments conducted during August, September, and October 2000.

Deposition rates for the Richmond and Clovis houses are similar and fall within the range of deposition rates found by other researchers (Thatcher and Layton, 1995; Fogh et al, 1997; Mosley et al, 2001). The deposition loss rates at the Clovis residence were slightly lower for all particle sizes, but the differences were not statistically significant. Deposition rates can vary between residences and within a single residence for many reasons. Thatcher et al. (2002) showed that both the indoor air speed (related to the internal mechanical energy) and the amount of furnishings influenced deposition loss rates. Both indoor air speed and furnishing level varied between the 2 facilities. Mean air speed will also typically vary between experiments at the same facility.

Penetration factors at the Richmond house were near unity for all but the largest and smallest particle sizes. Since it is older, has a relatively high normalized leakage area, and is constructed with double hung wooden windows and wood siding, the Richmond building is presumed to have larger cracks through the building shell. Large cracks are not expected to be very efficient

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for removing particles (Liu and Nazaroff, 2001). Penetration rates for the Clovis house were lower than those at the Richmond house, ranging from ~ 0.8 for particles 0.2 to $0.5 \mu\text{m}$ to just over 0.3 for particles $5 \mu\text{m}$ and larger. This may mean that in the tighter construction of the Clovis house, the penetration pathways are smaller and more effective for removing particles. However, there are no published studies on the correlation, or lack of correlation, between normalized leakage area and average crack dimensions. For particles between 0.1 and $1.0 \mu\text{m}$ there may be a confounding effect due to ammonium nitrate dissociation in the Clovis house, since ammonium nitrate particles are in this size range—see Figure 7 and John et al (1990). This may yield calculated penetration factors that are lower than the actual penetration factor in this particular size range.

The correlation between the deposition rates calculated by the two sets of instruments (APS and LAS-X) is very good for most sizes, especially considering that the instruments measure particles in a different manner (aerodynamic and optical diameter, respectively). The standard deviations for the deposition loss rates averaged about 60% of the deposition rate value. It is not clear from this study how much of this deviation is due to measurement and fitting inaccuracies and how much is due to actual variability in the deposition rate. Previous research has indicated that deposition loss rates can vary considerably based on conditions within the interior space (Thatcher, et al. 2002). The standard deviations for the penetration factors averaged 16% of the penetration factor and once again the portion of the variability attributable to experimental uncertainty as opposed to variations in the actual derived rate is unclear. The data and model of Liu, et al. (2001 and 2002) and the experiments of Mosely et al. (2001) suggest that the penetration factor is a function of pressure differential across the crack (reflecting flow rate

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effects within the building envelope). It is not unreasonable to assume that the penetration factor in real buildings will vary with environmental conditions, as these variations induce time variations in the differential pressure across the building shell. The parameter fit is not as sensitive to penetration factor as it is to deposition rate, since the decay period - which provides the best fitting data for deposition losses- is typically longer than the rebound period - which provides the best fitting data for the penetration rate, and therefore provides more data points for the model fit. Consequently, more model fit uncertainty is expected for the penetration factor. This is especially true in the upper size channels of both instruments, which sometimes record only a few particles during a measurement period, leading to noisy data and poor count statistics that reduce the accuracy of the reconstructions. Additional work is required to obtain a better understanding of the range, variability, and particle size dependence of the penetration factor in a representative range of real buildings.

CONCLUSIONS

Deposition and penetration losses are both important factors influencing indoor particle concentrations and resulting exposures. Since these factors operate simultaneously in the indoor environment, it can be difficult to separate their effects experimentally. We have demonstrated an effective method for determining size resolved penetration factors and deposition loss rates in full-scale homes. The two houses studied represent two distinctly different portions of the housing stock and air leakage spectrum; from older style, leakier, wood siding construction to a newer style, tighter construction. Although deposition loss rates were similar for the two buildings, penetration factors were significantly different. The older home had high penetration factors (near 1 for most particle sizes) while the newer home showed significant filtration by the

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building shell (penetration factors near 0.3 for particle larger than 5 μm). Deposition rates were slightly, but not significantly, higher in the older building. Deposition rate differences can be caused by differences in indoor air speed and/or differences in surface to volume ratio.

The composition of the ambient aerosol was shown to have a significant impact on the results and their interpretation as determined from some of the field experiments where ammonia nitrate concentrations were particularly large. Changes in the physical environment and gas phase contaminant concentrations indoors can lead to transformation processes, such as ammonium nitrate dissociation, which confounds the interpretation of experiments designed to quantify aerosol losses that result from crossing the building shell. Care must be taken to identify and account for the impact of these processes, to avoid improperly attributing their effects to deposition or penetration losses. Consideration and quantification of the various loss mechanisms must also be accounted for when estimating exposure.

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Table 1: Average model fit parameters and standard deviations for deposition loss rate and penetration factor at the Richmond and Clovis residences by (a) median optical diameter and (b) median aerodynamic diameter.

a) Optical Particle Counter Results

Median Optical Diameter of Size Range

Clovis	0.13	0.175	0.225	0.275	0.35	0.575	0.725	0.9	1.125	1.375	1.75	2.25	2.75
Deposition Loss Rate (hr-1)	0.12±0.09	0.11±0.10	0.15±0.16	0.2±0.2	0.2±0.2	0.12±	0.2±0.1	0.2±0.1	0.2±0.1	0.3±0.2	0.4±0.2	0.5±0.2	0.7±0.3
Penetration Factor	0.8±0.2	0.8±0.1	0.6±0.1	0.8±0.3	0.6±0.2	0.8±0.1	0.8±0.1	0.7±0.2	0.8±0.1	0.9±0.1	0.9±0.1	0.9±0.1	0.8±0.1
Richmond	0.13	0.175	0.225	0.275	0.35	0.575	0.725	0.9	1.125	1.375	1.75	2.25	2.75
Deposition Loss Rate (hr-1)	0.3±0.3	0.3±0.3	0.2±0.2	0.2±0.2	0.2±0.2	0.2±0.1	0.4±0.3	0.5±0.4	0.5±0.2	0.9±0.6	0.7±0.2	0.9±0.5	1.1±0.1
Penetration Factor	0.8±0.2	0.93±0.08	1.00±0.08	1.0±0.1	1.0±0.1	1.1±0.2	1.01±0.06	1.0±0.1	1.0±0.1	1.0±0.2	0.9±0.2	0.9±0.2	0.87±0.07

b) Aerodynamic Particle Counter Results

Median Aerodynamic Diameter of Size Range

Clovis	0.65	0.81	1.0	1.2	1.5	1.9	2.4	2.9	3.6	4.5	5.6	7	8.7
Deposition Loss Rate (hr-1)	0.1±0.1	0.1±0.1	0.1±0.1	0.2±0.1	0.3±0.1	0.5±0.1	0.9±0.2	1.3±0.4	1.8±0.4	2.2±0.8	3.3±0.6	3.5±1.1	3.3±1.3
Penetration Factor	0.7±0.2	0.8±0.2	0.9±0.2	0.9±0.2	0.7±0.04	0.7±0.02	0.62±0.06	0.5±0.1	0.4±0.1	0.4±0.1	0.33±0.05	0.35±0.09	0.48±0.16
Richmond	0.65	0.81	1.0	1.2	1.5	1.9	2.4	2.9	3.6	4.5	5.6	7	8.7
Deposition Loss Rate (hr-1)	0.5±0.7	0.4±0.4	0.4±0.2	0.5±0.4	0.7±0.3	1.0±0.4	1.3±0.5	1.5±0.7	2.3±0.6	2.6±1.0	3.7±0.9	4.8±0.7	5.06 ^a
Penetration Factor	1.0±0.3	1.0±0.2	1.0±0.1	0.93±0.06	0.94±0.05	0.93±0.06	0.93±0.09	0.86±0.03	0.9±0.1	0.9±0.1	0.9±0.1	0.77±0.06	0.88 ^a

a The acceptance criteria for this particle size was met for only one experiment.

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FIGURE CAPTIONS

- Figure 1: Floor plan of the experimental house located in Richmond, California. Particle measurement equipment was located in room 1. Tracer gas concentrations were measured at locations denoted by stars.
- Figure 2: Floor plan of the experimental house located in Clovis, CA showing locations of ceiling fans, tracer gas measurements, and differential pressure measurements. Particle measurement equipment was located in the living room and in the outdoor shed.
- Figure 3: Schematic of particle transport and removal processes in the indoor environment.
- Figure 4: Measured and modeled indoor particle concentrations for several optical particle diameter ranges during a typical run in Richmond, CA. The larger particle size ranges show increased sample-to-sample variability due to the low number of counts per sample period that produces poor count statistics.
- Figure 5: Measured and modeled indoor concentrations for selected aerodynamic particle diameters during a typical experiment at the Richmond site. For the constrained model, the penetration factor is 0.5 and the deposition rates are determined using the same residual minimization process as used in the unconstrained results.
- Figure 6: Measured and modeled indoor concentrations for selected optical particle diameter size ranges, along with outdoor PM_{2.5} nitrate concentrations at the Clovis, CA site. The increase in outdoor PM_{2.5} nitrate does not result in the increases in indoor particle concentrations predicted by the simple physical model that assumes that particles do not undergo phase change.

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Figure 7: Indoor and outdoor concentrations for PM_{2.5} nitrate and selected optical particle diameter size ranges. The outdoor nitrate peak results an increase in the outdoor particle concentration for particles with optical particle diameters between 0.1 and 1.0 μm , but does not result in a corresponding increase in indoor concentrations.

Figure 8: Averages and standard deviations of the parameter fits at the Richmond site for (a) penetration factor and (b) deposition loss rate as a function of optical and aerodynamic particle diameter. The optical and aerodynamic diameters are not necessarily identical for a given particle.

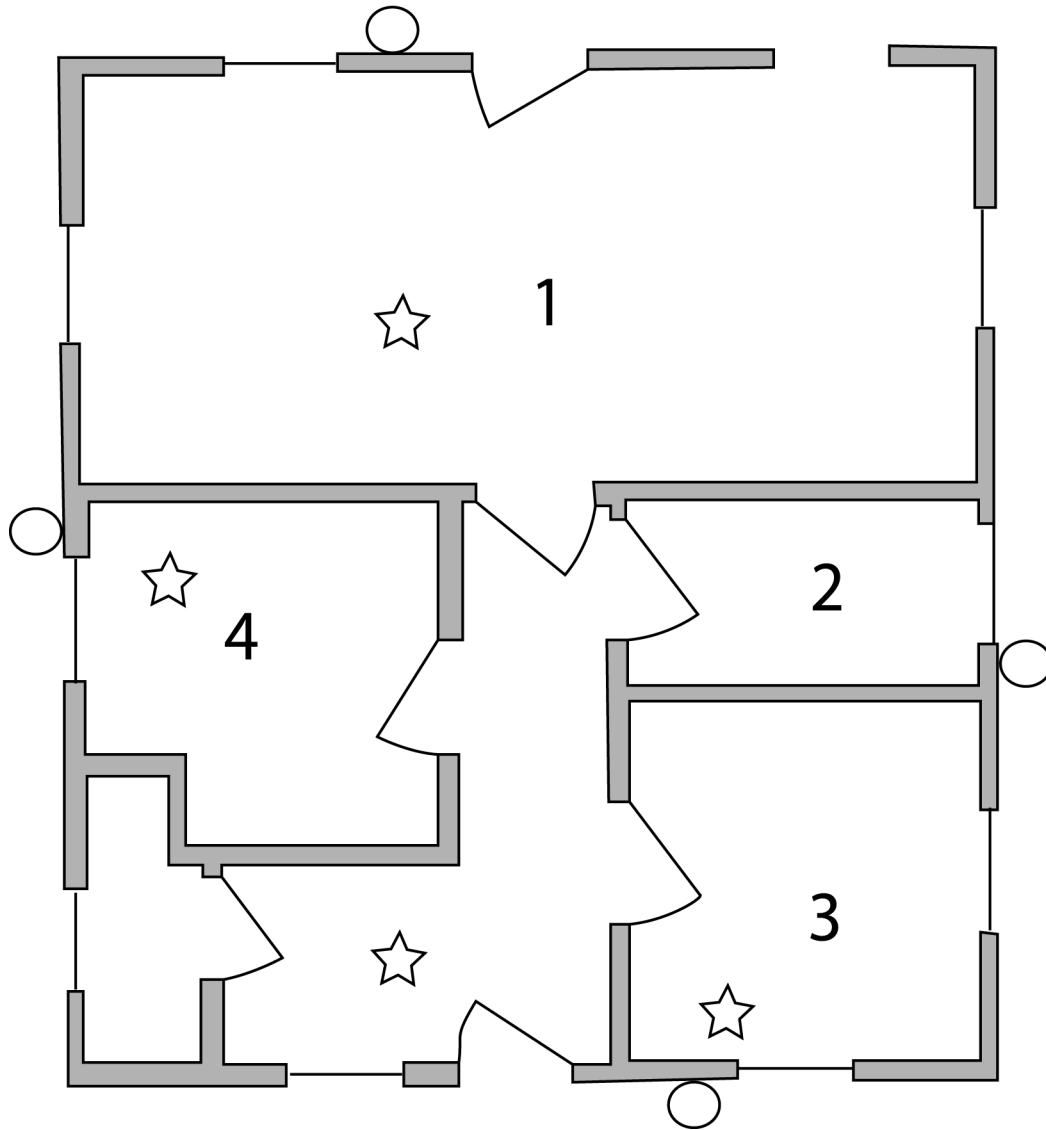
Figure 9: Averages and standard deviations of the parameter fits at the Clovis site for (a) penetration factor and (b) deposition loss rate as a function of optical and aerodynamic particle diameter. The optical and aerodynamic diameters are not necessarily identical for a given particle.

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○ Pressure Tap

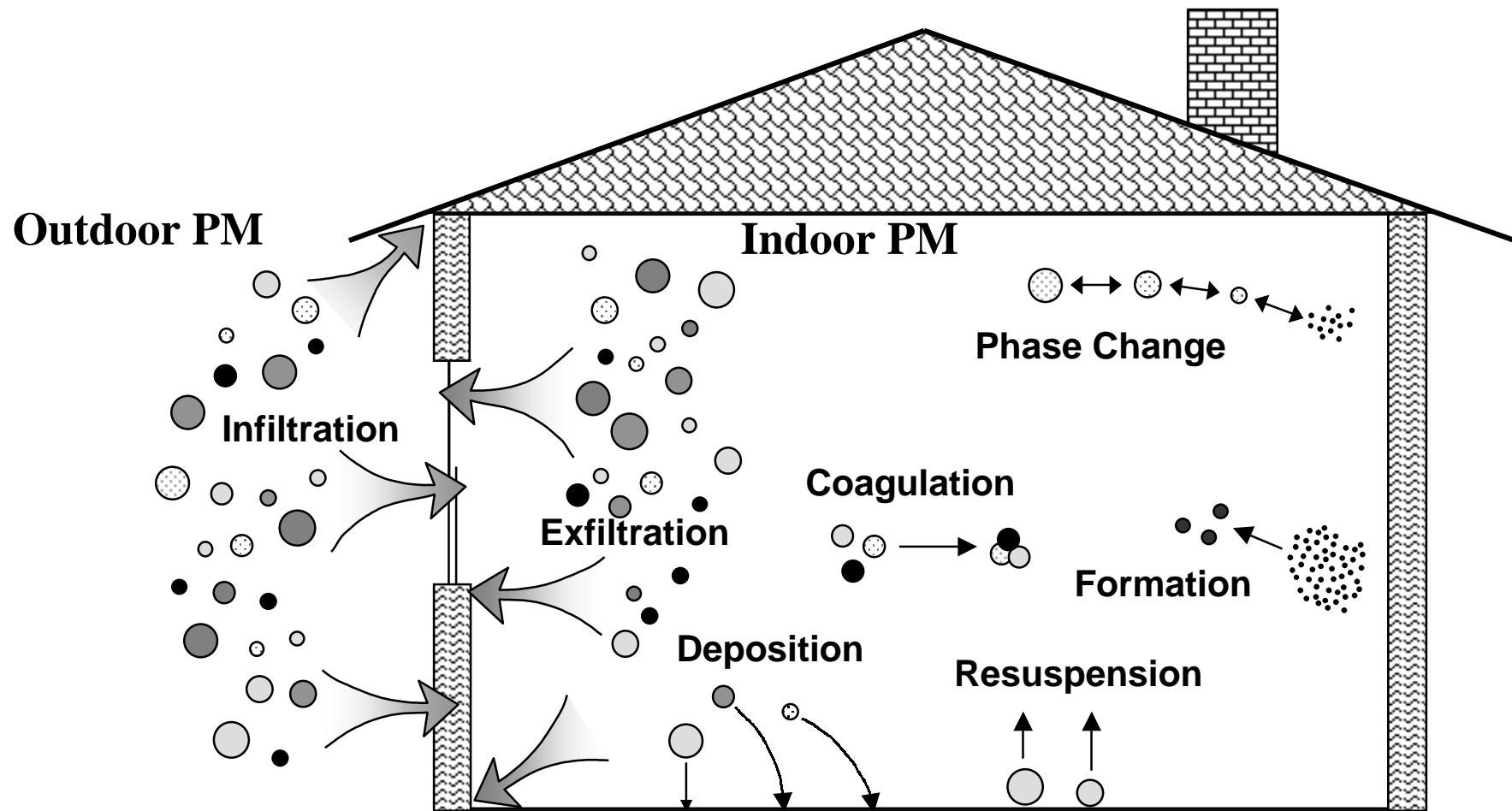
☆ Gas Detection

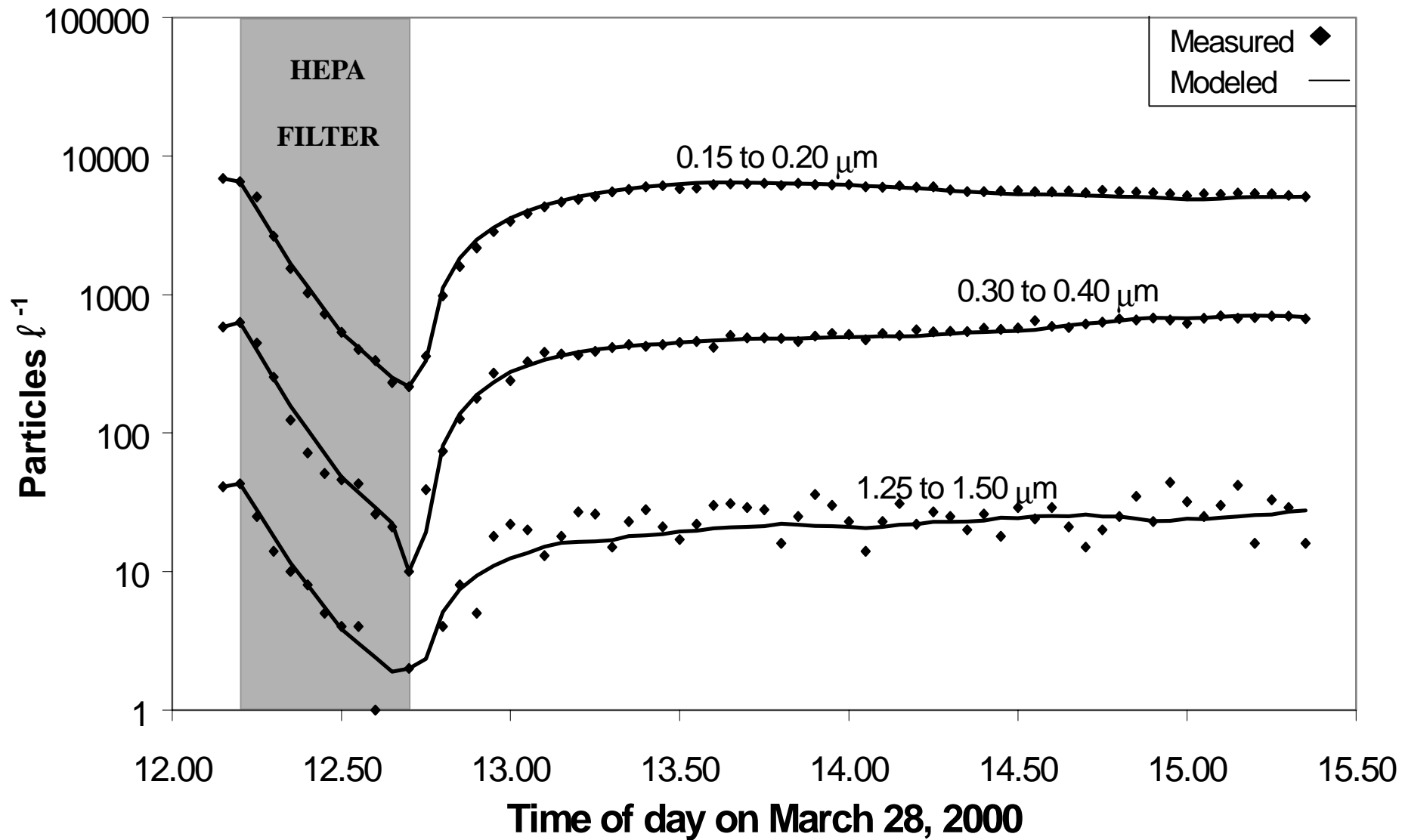
Total Area = 639 ft² (59 m²)

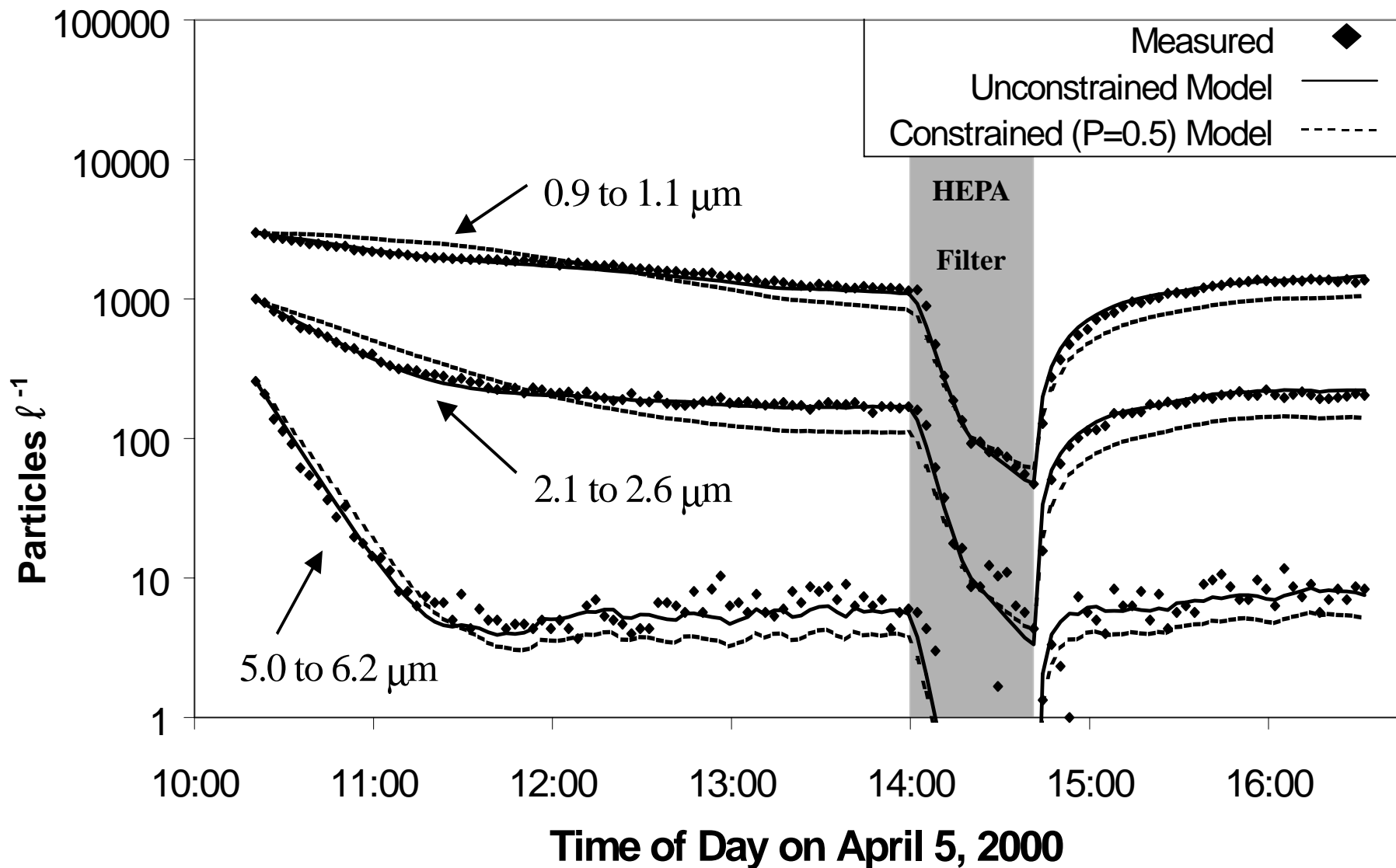


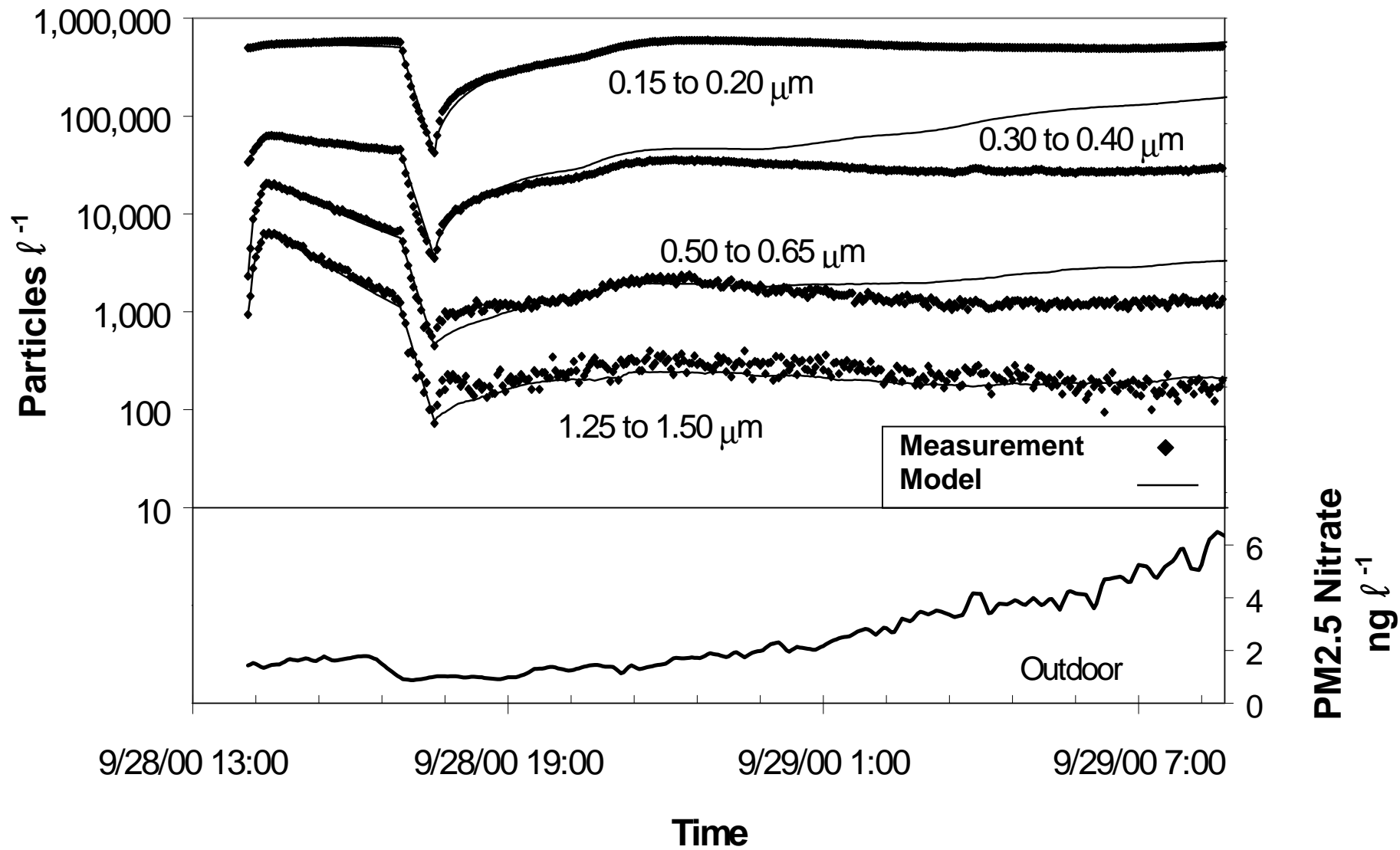
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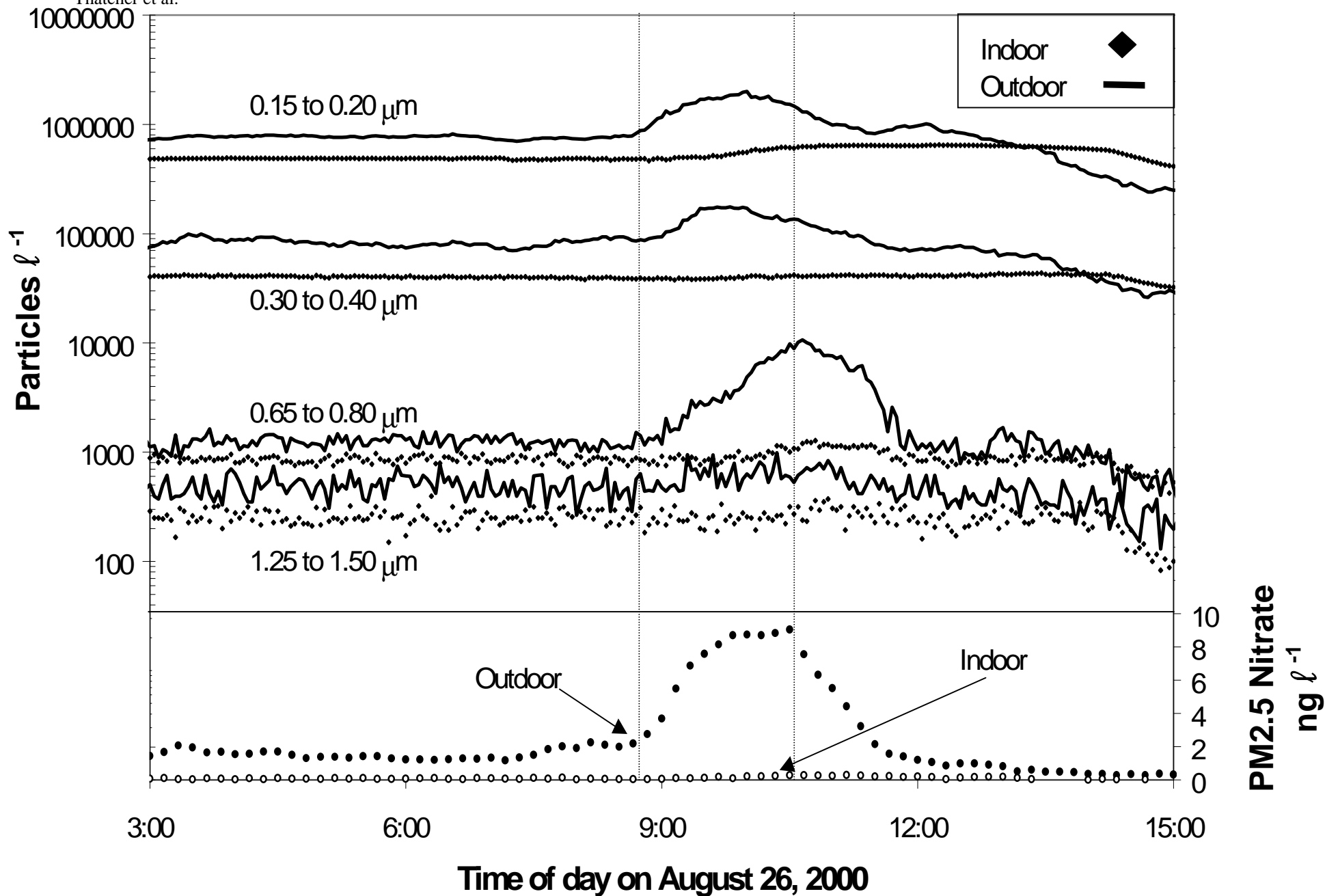








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