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Use of Real-Time Light Scattering Data To Estimate the Contribution of Infiltrated and Indoor-Generated Particles to Indoor Air

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The contribution of outdoor particulate matter (PM) to residential indoor concentrations is currently not well understood. Most importantly, separating indoor PM into indoor- and outdoor-generated components will greatly enhance our knowledge of the outdoor contribution to total indoor and personal PM exposures. This paper examines continuous light scattering data at 44 residences in Seattle, WA. A newly adapted recursive model was used to model outdoor-originated PM entering indoor environments. After censoring the indoor time-series to remove the influence of indoor sources, nonlinear regression was used to estimate particle penetration (P, 0.94 $\pm$ 0.10), air exchange rate (a, 0.54 $\pm$ 0.60 h^{-1}), particle decay rate (k, 0.20 $\pm$ 0.16 h^{-1}), and particle infiltration (F_{inf}, 0.65 $\pm$ 0.21) for each of the 44 residences. All of these parameters showed seasonal differences. The F_{inf} estimates agree well with those estimated from the sulfur-tracer method (R^2 = 0.78). The F_{inf} estimates also showed robust and expected behavior when compared against known influencing factors. Among our study residences, outdoor-generated particles accounted for an average of 79 $\pm$ 17% of the indoor PM concentration, with a range of 40−100% at individual residences. Although estimates of P, a, and k were dependent on the modeling technique and constraints, we showed that a recursive mass balance model combined with our censoring algorithms can be used to attribute indoor PM into its outdoor and indoor components and to estimate an average P, a, k, and F_{inf} for each residence.

Introduction

Epidemiological studies have shown associations between 24-h average ambient particulate matter (PM) concentrations and several adverse health effects including mortality, decrements in lung function, exacerbation of asthma, and increases in emergency room visits (*1−5*). Although ambient concentrations are poorly correlated with total personal exposure to PM (*6, 7*), some studies have demonstrated a strong correlation between ambient PM concentrations and personal exposure to PM of ambient origin (*6, 8*). There is little information on the contribution of ambient-generated PM to indoor PM concentrations.

Elderly people, a susceptible subpopulation, spend more than 70% of their time indoors at home (*9*). In the general population, a large portion of indoor PM mass is generated inside the residence from smoking, cooking, cleaning, and movement of people (*10−14*). In an elderly population, it is arguable that outdoor-generated PM contributes a larger percentage of the total indoor PM because the elderly conduct a less active lifestyle than the general population.

The chemical properties and physical characteristics of indoor-generated particles may differ from outdoor-generated particles due to differences in sources, photochemistry, and temporal and person-to-person variability (*15*). One small study has compared the toxicities of indoor- and outdoor-generated particles and found indoor-generated particles to be more toxic (*16*). Distinguishing contributions from ambient and nonambient PM to total personal exposure is important from a control and regulatory viewpoint (*15*). Several methods for estimating personal exposure to ambient-generated PM were recently published (*15, 17, 18*). The random component superposition model (*17*) was designed for estimating population exposure distributions. In contrast, Wilson et al. (*15*) and Mage (*18*) provided mass-balance methods for estimating personal exposure to PM components.

The challenging task in estimating personal exposure to ambient-generated PM is the separation of indoor PM into indoor- and outdoor-generated components. One of the most important parameters for making such a separation is the particle infiltration efficiency (F_{inf}), a unitless quantity defined as the equilibrium fraction of ambient PM that penetrates indoors and remains suspended (*15*). F_{inf} is a function of particle penetration efficiency (P), which is the fraction of ambient PM that is not removed from ambient air during its entry into the indoor volume, the particle removal rate due to diffusion or sedimentation (k), and the air exchange rate (a):

$$F_{\mathrm{inf}} = \frac{Pa}{a + k} \tag{1}$$

While a can be measured directly, estimates of P and k are more difficult to obtain. Several studies have estimated P, k, and F_{inf} in residences. The estimates of F_{inf} relied on the use of a physical model (*11, 19, 20*); a tracer with no indoor sources such as sulfate (*21, 22*); conversion of air exchange rates into F_{inf} values based on published conversion factors (*23*); or the indoor to outdoor PM ratio during nighttime, nonsource periods (*24*). These studies have provided average values across homes or individual values for a limited number of homes. No studies have made individual estimates in a large group of residences where susceptible individuals reside, including private homes, apartments, and retirement facilities. Therefore, little is presently known about the inter-home variability of P, a, k, and F_{inf}. In addition, there have been no papers published that estimate separately short-term levels of indoor- and outdoor-generated PM.

This paper takes the initial step of separating indoor light scattering measurements into indoor- and outdoor-generated

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TABLE 1. Residence and Subject Characteristics[a]

	private homes	private apts	group homes	total residences	total monitoring events
total monitored	27 (8)	12 (2)	5 (1)	44 (11)	55
season					
heating season only	13 (2)	6	1	20 (2)	22
nonheating season only	10 (2)	4	3	17 (2)	19
both seasons	4 (4)	2 (2)	1 (1)	7 (7)	14
air cleaner used					
yes	7 (2)	1 (1)	1	9 (3)	12
no	20 (6)	11 (1)	4 (1)	35 (8)	43
age of residence (yr)					
0–29	3 (3)	8 (1)	3	14 (4)	18
30–59	15 (2)	1	2 (1)	18 (3)	21
≥60	9 (3)	2	0	11 (3)	14
unknown	0	1 (1)	0	1 (1)	2
subject health status[b]					
healthy	6	2	0	8	8
COPD	7 (2)	1 (1)	4 (1)	12 (4)	16
CHD	6 (2)	9 (1)	1	16 (3)	19
asthmatic children	8 (4)	0	0	8 (4)	12
air conditioning[c]					
central	4	0	0	4	4
window	1	3	0	4	4
none	22 (8)	9 (2)	5 (1)	36 (11)	47
heating system					
forced air	22 (6)	2	0	24 (6)	30
electric space heater	2 (1)	7 (1)	3	12 (2)	14
radiator/heated floor	1	1	2 (1)	4 (1)	5
gas space heater	1 (1)	1	0	2 (1)	3
open stove	0	1 (1)	0	1 (1)	2
unknown	1	0	0	1	1

[a] Number in parentheses indicates the number of residences monitored twice. [b] healthy = elderly without cardiopulmonary disease. COPD = chronic obstructive pulmonary disease. CHD = coronary heart disease. [c] The presence of air conditioning, regardless of whether it was used.

components using data collected at 44 residences in Seattle over 2 years. We developed censoring algorithms to identify indoor sources in the indoor light scattering time-series and then applied a recursive mass balance model (*25*) to the hourly outdoor and censored indoor light scattering data. A nonlinear regression was used to estimate the average P, a, k, and F_{inf} for each residence in the recursive model. The relatively large number of residences monitored in this study allowed us to estimate the contribution of ambient PM to indoor concentrations and to evaluate the between-home variability of that contribution.

Experimental Section

Study Design. This study was a subset of a larger exposure assessment study conducted in Seattle between November 1999 and May 2001 (*26*). The monitored residences included private homes, private apartments, and group homes (Table 1). Monitoring occurred in both the heating (October–February) and nonheating (March–September) seasons. Residents included healthy elderly subjects, elderly with chronic obstructive pulmonary disease (COPD) and coronary heart disease (CHD), and child subjects with asthma. The residence and subject characteristics are shown in Table 1 for those residences that met our quality control criteria. Subjects' activities and potential PM-generating activities were recorded on a subject-administered time-location-activity diary (TAD) and on a technician-administered daily follow-up questionnaire (DFQ) (see Supporting Information).

Particle mass and light scattering were monitored during 24 10-day monitoring sessions. To account for residences that were monitored twice, we defined a "monitoring event" as the monitoring of a residence for a single 10-day monitoring session. Light scattering was monitored using the real-time Radiance nephelometers (hereafter referred to as neph; Radiance Research, Seattle, WA) both inside and immediately outside the subjects' residences during 73 monitoring events. A detailed description of this instrument and the setup were described previously (*27*). Collocated 24-h $PM_{2.5}$ mass concentration measurements were taken inside and outside the residences using the Harvard impactors ($HI_{2.5}$; *27*). Our monitoring setup minimized the effect of relative humidity on the light scattering response, and the 24-h light scattering to mass concentration relationship did not differ significantly among homes (*27*).

In addition to particle mass, 136 pairs of indoor and outdoor Teflon filters collected with $HI_{2.5}$ during 14 monitoring events (from 10 residences with 4 monitored twice) were analyzed using energy-dispersive X-ray fluorescence (XRF) for a suite of 55 trace elements including sulfur. The limit of detection for sulfur was 2.6 ng/m^3. The indoor to outdoor concentration ratio of a component of PM without indoor sources gives a direct estimate of F_{inf} for particles with the same aerodynamic diameter (*15*). Sulfur has conventionally been selected as a tracer of outdoor $PM_{2.5}$ since there are few indoor sources of sulfur (*10*, *11*), and indoor and outdoor sulfur concentrations have been shown to be highly correlated (*11*, *28*). Although studies (*10*, *22*) have identified smoking and kerosene heaters as indoor sulfur sources, neither source was present among our study subjects. Air exchange rate was measured daily at 5 residences (each monitored only once) using a perfluorinated methylcyclohexane tracer and capillary adsorption tubes (*29*).

Quality Control (QC). As this was the first study to apply portable nephelometers in a large number of residences for aerosol characterization, it was important to establish and apply vigorous, objective, and consistent QC criteria to eliminate any possible measurement errors generated from field operation problems, monitor glitches, and outliers. Our QC criteria included the following for each monitoring event: (i) achieve 50% data collection to ensure unbiased F_{inf} estimates, (ii) have a significant ($p < 0.05$) indoor to outdoor relationship during nonsource periods as used by the PTEAM study (*11*) and Long et al. (*24*), and (iii) have a median indoor to outdoor ratio <1 during nonsource periods to capture occasional neph baseline drift. We removed 6 monitoring events that did not achieve 50% data collection. The 10-min neph measurements with a light scattering coefficient greater than 6.0×10^{-3} m^{-1} (~2145 μg/m^3; neph malfunction) or values of less than -1.0×10^{-4} m^{-1} (unreasonably large negative values) were also removed (*27*). After averaging the 10-min data into 1-h segments, we examined the indoor–outdoor relationship during nighttime, nonsource periods, which were defined as times between 11 PM and 9 AM when the subject's TAD indicated that they were asleep and the DFQ did not indicate any indoor sources. During these periods, 5 monitoring events had a median hourly indoor to outdoor ratio greater than 1, and 7 monitoring events had an insignificant relationship between indoor and outdoor light scattering. In total, 18 monitoring events were removed, leaving 55 monitoring events from 44 unique residences for analysis. For these monitoring events, we converted the 24-h neph averages into $PM_{2.5}$ mass concentrations ($PM_{2.5}$ (μg/m^3) = ($b_{sp} \times 10^5 - 0.01$)/0.28), which were compared with the collocated 24-h $HI_{2.5}$ measurements. Differences that were > (75th percentile + 1.5 × interquartile range) or < (25th percentile − 1.5 × interquartile range) were removed. Outdoors, outliers were removed if the neph showed poor agreement with the central-site $HI_{2.5}$ measurements (obtained from the Washington State Department of Ecology) while the home outdoor $HI_{2.5}$ agreed well with the central-site $HI_{2.5}$. These outliers were most likely due to malfunction of the outdoor neph fan or heater. These criteria removed 53 (9.6%) of 551 d of data. Overall, 4344

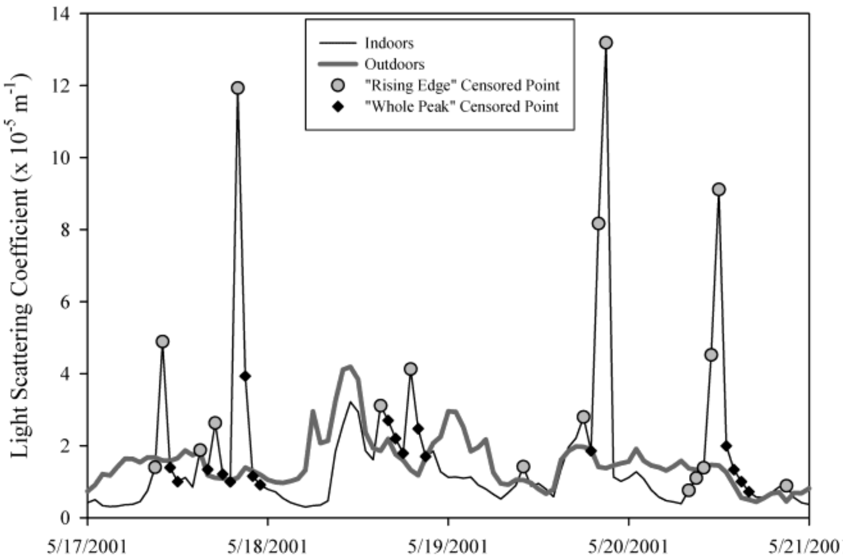

FIGURE 1. Portion of a censored indoor time−series for one monitoring event.

(28%) of the original 15 337 h of neph data were identified as problematic and excluded from the analysis. Of the 136 pairs of sulfur data, we included only days for which we also had valid indoor and outdoor neph data ($N = 107$ pairs). Five of these days were removed when data collection logs indicated that the indoor or outdoor sample had been mishandled or that the filter was torn. In addition, 4 days from 3 monitoring events at 3 separate residences with an indoor to outdoor sulfur ratio greater than 1 (range = 1.03−1.21) were also removed. Although resuspension of indoor particles could be one possible source of sulfur on these days, the average number of people in the residence and the frequency of being active indoors were no higher on these days than on other days. As our TAD and DFQ were not designed to highlight sulfur sources, it is likely that sulfur-containing sources were present but not noted in these records. The final sulfur analysis included 98 valid pairs of sulfur data from 14 monitoring events.

Recursive Model. We adapted a recursive mass balance model (RM) to the valid light scattering data (*25*). Our model uses the average light scattering values over time periods of equal duration (1 h) and assumes constant air exchange rates and well-mixed indoor air. This model states that the average indoor light scattering coefficient during time period t ($(b_{sp})_t^{in}$) is equal to the sum of a fraction of the average outdoor scattering coefficient during the same time period ($(b_{sp})_t^{out}$), a fraction of the average indoor scattering coefficient from the previous time period ($(b_{sp})_{t-1}^{in}$), and the scattering contribution from indoor sources (S_t^{in}):

$$(b_{sp})_t^{in} = a_1(b_{sp})_t^{out} + a_2(b_{sp})_{t-1}^{in} + S_t^{in} \qquad (2)$$

Parameter a_1 describes the fate of outdoor particles once they penetrate indoors:

$$a_1 = F_{inf}\{1 - \exp[-\Phi\Delta t]\} \qquad (3)$$

where Φ is the total particle loss rate (h^{-1}):

$$\Phi = a + k \qquad (4)$$

Parameter a_2 describes the decay of indoor particles:

$$a_2 = \exp[-\Phi\Delta t] \qquad (5)$$

To apply a mass balance model to b_{sp} measurements, we assumed a constant relationship between infiltrated particle mass and scattering coefficient on a 1-h basis. We minimized the influence of the indoor source term in eq 2 by censoring the indoor data and modifying points when the indoor b_{sp} levels increased rapidly without corresponding changes in the outdoor light scattering levels:

$$\frac{(b_{sp})_t^{in}}{(b_{sp})_{t-1}^{in}} > 1.5 \text{ and } \frac{(b_{sp})_t^{out}}{(b_{sp})_{t-1}^{out}} < 1.5 \text{ and}$$
$$((b_{sp})_t^{in} - (b_{sp})_{t-1}^{in}) > 10^{-5}\ \mathrm{m}^{-1}\ (\text{or} \sim 4\ \mu\mathrm{g/m}^3) \qquad (6)$$

The difference between "modifying" points and deleting them is important at times $(b_{sp})_t^{in}$ immediately following a censored peak. For such points, the $(b_{sp})_{t-1}^{in}$ value is censored but not deleted and is, therefore, available for the $(b_{sp})_{t-1}^{in}$ term in eq 2. Once a point was identified by eq 6, we continued to censor the "rising edge" of the peak in the indoor time-series as long as the indoor light scattering level continued to increase. We censored only the rising edge because at time (t) when an indoor source is shut off and the indoor concentration begins to decay, the S_t^{in} term in eq 2 becomes 0, and the particles generated by the indoor source become part of the $(b_{sp})_{t-1}^{in}$ term (i.e., part of the indoor light scattering during the previous time step). Retaining the decay of indoor peaks allows for a more accurate estimate of the total particle loss rate. Here we implicitly assume that the decay of light scattering was representative of the decay of $PM_{2.5}$ in these residences. In addition to prominent indoor-generated peaks, we also identified low-level indoor sources as

$$((b_{sp})_t^{in} - (b_{sp})_{t-1}^{in}) > 2.5 \times 10^{-6}\ \mathrm{m}^{-1}\ (\text{or} \sim 1\ \mu\mathrm{g/m}^3) \text{ and}$$
$$((b_{sp})_t^{out} - (b_{sp})_{t-1}^{out}) \leq 0 \qquad (7)$$

To validate our censoring algorithm, we also manually censored the indoor time-series for indoor sources that might not be identified by the algorithms. An example of a censored time-series is shown in Figure 1, in which indoor sources identified by the "rising edge" censoring algorithm are marked with filled circles.

To test our assumption that rising edge censoring was appropriate, we also censored the entire indoor source peak by examining each time-series and manually censoring each peak's decay ("whole peak" censoring). This whole peak technique is identified in Figure 1 with filled diamonds. The rising edge censoring process identified 13.3% of the hourly measurements, while the whole peak method identified 24.1% of the hourly measurements.

Assuming that $S_t^{in} = 0$, subtracting $(b_{sp})_{t-1}^{in}$ from both sides of eq 2 and multiplying by a censoring modifier (δ), we obtained the following equation:

$$\Delta(b_{sp})^{in} = \{a_1(b_{sp})_t^{out} + (a_2 - 1)(b_{sp})_{t-1}^{in}\}\delta \qquad (8)$$

δ was set equal to 0 when $(b_{sp})_t^{in}$ was identified as a censored point, otherwise δ was set equal to 1. This procedure allowed us to minimize the influence of the S_t^{in} term in eq 2 without deleting any data; thus, all data were available for our later re-creation of the indoor-generated and infiltrated indoor time-series (see Supporting Information).

We determined values for P, a, and k for each monitoring event by solving eq 8 using a nonlinear regression model (PROC NLIN, SAS Version 8), with bounds $0 \le P \le 1$, $a \ge 0\ h^{-1}$, and $k \ge 0\ h^{-1}$. Note that P, a, and k entered into eq 8 through eq 1 and the terms a_1 and a_2 in eqs 3–5. The nonlinear technique, which was also used in the PTEAM study (*11*), allows for the least squares error fit of parameters while setting boundaries for those parameters based on physically reasonable values. As part of our sensitivity analysis, we used the 5 monitoring events during which a was measured to evaluate the effect of inserting a measured value of a into the model and solving only for P and k. In addition to an overall average P, a, k, and F_{inf} for each monitoring event, we also calculated separate averages for open- and closed-window days during each monitoring event. An open-window day was defined as a day with any number of windows open for any length of time.

Data Analysis. We estimated F_{inf} two ways: (i) using eq 1 and the nonlinear regression parameter estimates and (ii) using a linear regression (forcing the intercept to zero) of eq 8 to solve for a_1 and a_2. F_{inf} can be calculated from

$$F_{inf} = \frac{a_1}{1 - a_2} \qquad (9)$$

However, the linear regression does not solve for P, a, and k individually. It does solve for the total particle loss rate:

$$\Phi = a + k = -\ln(a_2) \qquad (10)$$

The linear regression was expected to produce more stable estimates since only 2 parameters were being simultaneously estimated. Linear and nonlinear estimates of F_{inf} and Φ were compared to determine if the estimates differed depending on the number of parameters being estimated. F_{inf}^{S} values, calculated from the indoor and outdoor sulfur data, were used as a reference to compare with the RM F_{inf} estimates. Unless otherwise noted, the F_{inf} values presented are those estimated using nonlinear regression.

To estimate the percent contribution of infiltrated particles to the indoor PM mass concentration during each monitoring event, we multiplied the average of each event's 24-h outdoor filter samples by the estimated F_{inf} and then divided this infiltrated concentration by the average of each event's 24-h indoor filter samples. In cases where the infiltrated concentration was greater than the measured indoor concentration ($N = 8$), the infiltrated concentration was set equal to the measured indoor concentration, and the indoor-generated concentration was set equal to 0.

We attempted to validate the estimates of P, a, k, and F_{inf} by using regression models to evaluate the effect of influential factors on our modeled estimates, including season (increased F_{inf} in summer) (*24*), air conditioning or keeping windows closed (decreased F_{inf}) (*19*, *21*, *24*), building age (increased F_{inf}) (*23*), the presence of storm or double-pane windows (decreased F_{inf}) (*19*), and the use of an air cleaner.

Results and Discussion

Summary Statistics. The hourly neph light scattering data collected indoors and outdoors were log-normally distributed. The geometric mean (geometric standard deviation) of the light scattering data outdoors was $2.10 \times 10^{-5}\ m^{-1}$ (1.66), which was higher than the indoor geometric mean of $1.88 \times 10^{-5}\ m^{-1}$ (1.68) ($p < 0.001$). Indoor light scattering had a higher maximum hourly value ($100.8 \times 10^{-5}\ m^{-1}$) than that outdoors ($28.0 \times 10^{-5}\ m^{-1}$). Indoor and outdoor light scattering were only slightly correlated (Pearson's $r = 0.31$ for hourly data and 0.62 for 24-h data). When only nighttime, nonsource periods were included, the r for all hourly data increased to 0.77. The 24-h indoor–outdoor correlation was similar to the indoor–outdoor $HI_{2.5}$ correlation reported in our Seattle panel study ($r = 0.58$) (*26*) but lower than those reported for $PM_{2.5}$ gravimetric samples among similar populations in Baltimore ($r = 0.96$) and Fresno ($r = 0.93$) (*30*, *31*), probably due to the minimal number of indoor sources in these studies.

Using a nonlinear regression model to solve the RM, we estimated values of P, a, and k for each monitoring event (Figure 2). The average value ($\pm$ SD) for F_{inf} across all monitoring events was 0.65 ± 0.21. This average F_{inf} value agrees well with most previously published values (Table 2), which ranged between 0.50 in Boston, MA, for 1–2 μm particles and 0.86 for sulfate in non-air-conditioned homes in Uniontown, PA. We estimated the average value for P to be 0.94 ± 0.10, which agrees well with those reported by Thatcher and Layton (*32*) for all particle size ranges (0.3–25 μm) and by the PTEAM study for $PM_{2.5}$ and $PM_{2.5-10}$ (*33*). Our average for k, $0.20 \pm 0.16\ h^{-1}$, is near the low end of the experimental values presented in the literature (*11*, *20*, *24*, *34*). Long et al. (*24*) reported the lowest average $PM_{2.5}$ decay rate of 0.15 (summertime) and 0.10 h^{-1} (wintertime), while Wallace et al. (*34*) reported the average k in one townhouse of 0.20, 0.37, and 0.59 for particle size ranges of 0.3–0.5, 0.5–1, and 1–2.5 μm, respectively. The k was reported to be 0.39 h^{-1} for $PM_{2.5}$ and 0.16 h^{-1} for fine sulfur in the PTEAM study (*11*). Abt et al. (*20*) reported the highest k values, between 0.70 and 1.11 h^{-1}, for particle size ranging between 0.02 and 1 μm.

In our study, the estimated F_{inf} was significantly higher on open-window days than on closed-window days (Table 3), and a was marginally significantly higher ($p = 0.08$) on open-window days. This difference in a remained ($p = 0.08$) even after removing the monitoring event with the highest estimated open-window a (4.63 h^{-1}). The average k was significantly higher on closed-window days, probably due to a longer residence time when windows are closed. The estimated P did not differ based on window opening.

The average modeled value for a in our study residences, $0.54 \pm 0.60\ h^{-1}$, agrees with our measured a in 13 other Seattle homes (0.56 ± 0.27). The average a was heavily influenced by one monitoring event (subject no. 3, $a = 4.47\ h^{-1}$, open windows on 6 of 10 days), and when this monitoring event was excluded the overall mean a was $0.46 \pm 0.26\ h^{-1}$. Wallace et al. (*35*) reported a mean air exchange rate of 0.56 and 0.73 h^{-1} during the daytime and nighttime, respectively, in the same townhouse, while two other studies estimated the median air exchange rate in the United States to be 0.53 and 0.46 h^{-1} (*36*, *37*).

While the average values of P, a, and k agree well with previously published values, the SAS nonlinear regression algorithm does not produce confidence intervals for individual estimates of P, a, and k. The variation of these estimates among homes provides a crude overall uncertainty. However, if measured air exchange rates are available, nonlinear

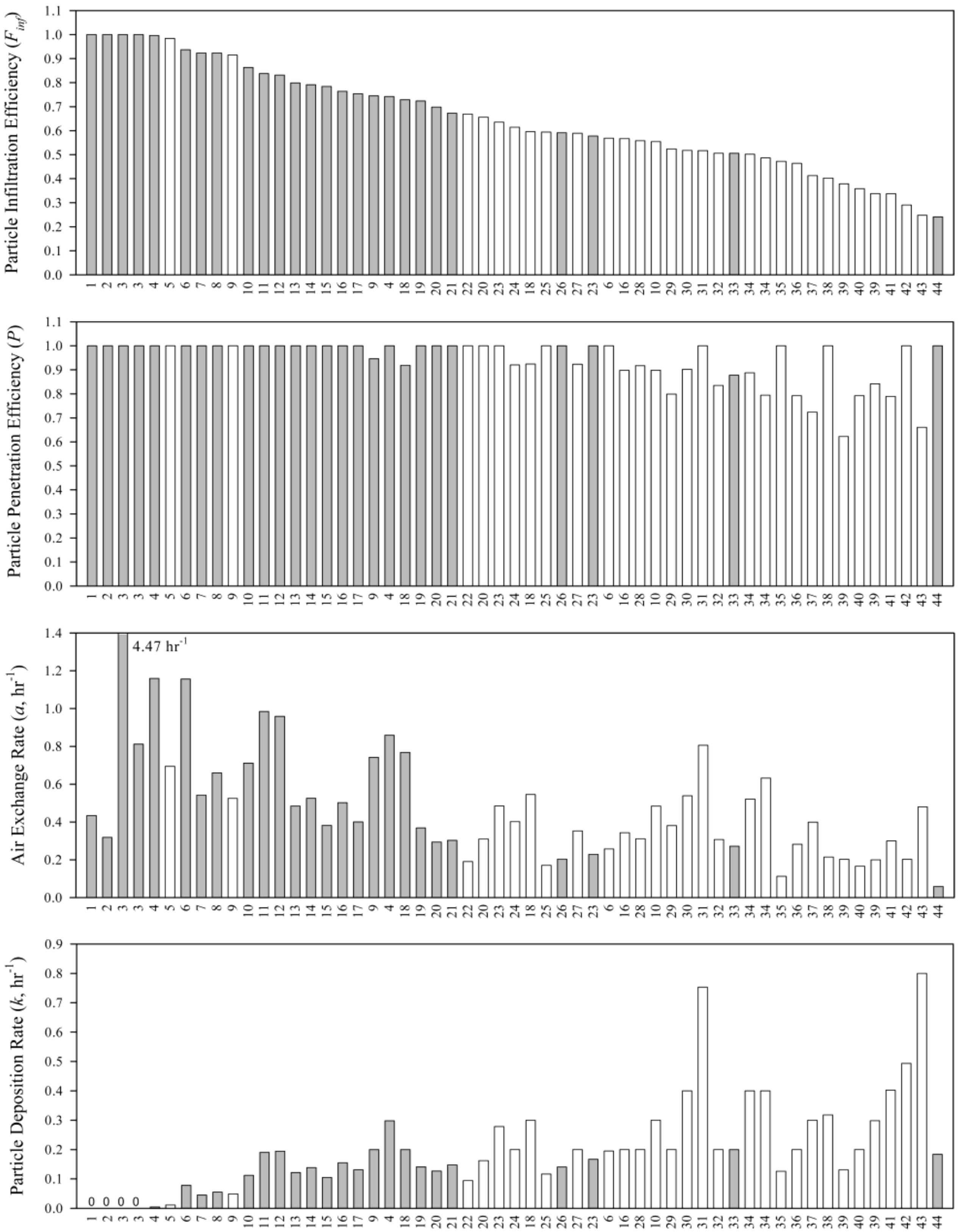

FIGURE 2. Individual estimates of F_{inf}, P, a, and k. (Note: Repeated number indicates residences monitored twice; gray bars indicate homes monitored during nonheating season.)

regression of the RM for estimating P and k could be quite useful, as the confidence intervals can be easily produced when solving with only two unknowns.

Model Validation. We examined the adequacy of our censoring techniques and the robustness of the F_{inf}, P, a, and k estimates. To assess the effectiveness of our whole peak censoring, we compared indoor to outdoor light scattering ratios estimated using whole peak censoring with those calculated using uncensored nighttime data. The estimates with the whole peak technique (mean ratio = 0.75 ± 0.25) agreed well (R^2 = 0.79) with those estimated with the uncensored data (mean ratio = 0.77 ± 0.24), indicating that

TABLE 2. Summary of Published Mean Infiltration Efficiencies (F_{inf})

location	season	no. of residences	particle size/tracer	mean F_{inf}	ref
Riverside, CA	fall	178	$PM_{2.5}$	0.7[a]	11
Uniontown, PA	summer	24	sulfate	AC = 0.69[b]	21
				non-AC = 0.86[b]	
Virginia & Connecticut	summer	58	sulfate	0.74[b]	22
Boston, MA	spring−summer & fall−winter	9	$PM_{2.5}$	0.74[b]	24
Boston, MA	summer & winter	4	PM: 0.7−1 μm	0.53[a]	20
			PM: 1−2 μm	0.50[a]	
Birmingham, AL	summer & winter	10	$PM_{2.5}$	0.66[a]	23
Six cities	all seasons	68	$PM_{3.5}$	0.70[a]	19
			sulfates	0.75[a]	
Seattle, WA	all seasons	44	light scattering	0.65[a]	this work
		10	sulfur	0.60[b]	

[a] Modeled. [b] Measured.

TABLE 3. Distribution of Parameters Estimated from Nonlinear Regression of Recursive Model on Open- and Closed-Window Days

parameter	windows	mean[c]	min.	10%	25%	50%	75%	90%	max.
F_{inf}	open[a]	0.69*	0.24	0.39	0.53	0.71	0.87	1.00	1.00
	closed[b]	0.58*	0.25	0.34	0.43	0.56	0.71	0.88	1.00
	all days	0.65	0.24	0.36	0.51	0.61	0.80	0.98	1.00
P	open[a]	0.93	0.68	0.71	0.88	1.00	1.00	1.00	1.00
	closed[b]	0.90	0.62	0.76	0.84	0.92	1.00	1.00	1.00
	all days	0.94	0.62	0.79	0.90	1.00	1.00	1.00	1.00
a (h^{-1})	open[a]	0.59**	0.05	0.19	0.31	0.44	0.67	0.98	4.63
	closed[b]	0.40**	0.10	0.17	0.20	0.31	0.57	0.77	0.93
	all days	0.54	0.06	0.20	0.28	0.40	0.63	0.86	4.46
k (h^{-1})	open[a]	0.16*	0.00	0.00	0.08	0.16	0.20	0.30	0.40
	closed[b]	0.23*	0.00	0.04	0.13	0.20	0.30	0.48	0.80
	all days	0.20	0.00	0.01	0.12	0.19	0.28	0.40	0.80

[a] Based on 44 monitoring events. [b] Based on 35 monitoring events (36 had at least one closed-window day, but one did not converge due to the smaller data sets after separating by window status). [c] *, $p < 0.05$; **, $p < 0.10$.

TABLE 4. Results of Regression Analysis[a]

parameter	effect	estimate ± SE	p value	model R^2
F_{inf}	intercept	0.57 ± 0.05	<0.001	0.50
	nonheating season	0.27 ± 0.05	<0.001	
	air cleaner used	−0.14 ± 0.05	<0.05	
	building age ≥ 45 yr	−0.10 ± 0.05	<0.05	
	storm windows	0.02 ± 0.05	0.70	
P	intercept	0.89 ± 0.03	<0.001	0.32
	nonheating season	0.11 ± 0.02	<0.001	
	air cleaner used	0.01 ± 0.04	0.76	
	building age ≥ 45 yr	−0.03 ± 0.02	0.13	
	storm windows	0.02 ± 0.02	0.45	
a	intercept	0.72 ± 0.21	<0.001	0.18
	nonheating season	0.31 ± 0.13	<0.05	
	air cleaner used	−0.30 ± 0.17	0.08	
	building age ≥ 45 yr	−0.03 ± 0.12	0.77	
	storm windows	−0.35 ± 0.20	0.07	
k	intercept	0.24 ± 0.04	<0.001	0.39
	nonheating season	−0.17 ± 0.03	<0.001	
	air cleaner used	0.05 ± 0.05	0.37	
	building age ≥ 45 yr	0.12 ± 0.04	<0.001	
	storm windows	−0.02 ± 0.04	0.54	

[a] $N = 52$. The monitoring event with $a = 4.47\ h^{-1}$ has been excluded from this analysis. Building age was unknown for two monitoring events.

indoor source peaks were adequately removed by the whole peak censoring method. To test the validity of our parameter estimates, we constructed models that regressed F_{inf}, P, a, and k against influential factors (Table 4). As expected, F_{inf} is significantly higher in the nonheating season, when P and a are higher and k is lower. Lachenmyer and Hidy (*23*) reported that older building age was associated with higher infiltration, although their results were based on measurements at 10 homes in Birmingham, AL. Our model indicates the opposite effect of building age on F_{inf} in the Seattle residences. In addition, older residences were associated with a higher mean k, possibly due to a higher surface to volume ratio in older homes (*11*). The estimated air exchange rate was lower in homes with storm or double-pane windows and in homes that used an air cleaner.

Across all monitoring events ($N = 55$), the 10-day average outdoor, indoor, infiltrated, and indoor-generated concentrations were 10.2 ± 2.8, 8.1 ± 2.2, 6.3 ± 1.8 (78% of total indoor PM), and 1.8 ± 1.8 $\mu g/m^3$ (22% of total indoor PM), respectively (Table 5). Specifically, infiltrated particles accounted for 40−100% percent of the total indoor $PM_{2.5}$ concentration, with a mean of 79 ± 17% (Table 5). For all monitoring periods, we found a significant ($p < 0.05$), inverse relationship between the fraction of hours spent cooking and the outdoor contribution to the total indoor concentration. We also found an association between the outdoor contribution to the indoor concentration and the longitudinal indoor to outdoor correlation ($p < 0.001$). Koutrakis et al. (*10*) estimated that outdoor sources were associated with 60−70% of the indoor $PM_{2.5}$ mass concentration in nonsmoking homes in two counties in New York. Abt et al. (*20*) estimated that, for particles of 0.02−0.3 and 2−10 μm, outdoor particles comprise 57−80% and 20−43% of the PM mass, respectively. The fact that indoor particles accounted for less of the indoor PM mass in our study than in other studies reflects the fact that the majority of our subjects were 65 years old or older and were less active indoors than subjects in other studies (*26*). For 8 of the 55 monitoring events in our study, we estimated that outdoor particles accounted for 100% of the indoor $PM_{2.5}$ concentration. The longitudinal indoor to outdoor correlation was higher for these monitoring events (mean $r = 0.91 \pm 0.05$) than for the remaining 47 monitoring events (mean $r = 0.69 \pm 0.27$, $p < 0.05$). We further validated our modeled results using activity data collected on the TAD and DFQ. One of these 8 monitoring events reported no cooking (the major indoor source of $PM_{2.5}$ in nonsmoking households). On average, these subjects reported cooking during 2.8 ± 2.6% of the hours as compared with 4.5 ± 2.7% for the remaining 47 monitoring events ($p < 0.10$).

Estimation of F_{inf} Using Different Techniques. The modeled F_{inf} values are comparable using three different estimating techniques. Linear regression of the recursive model (eq 8) produces estimates of F_{inf} and Φ of 0.66 ± 0.23 and 0.74 ± 0.60 h^{-1}, respectively. Individual estimates of F_{inf} and Φ are identical to those produced by nonlinear regression with the exception of 4 monitoring events with F_{inf} greater than 1 (range = 1.01−1.30), which were bounded at 1 in the nonlinear regression. This indicates that the nonlinear model

TABLE 5. Distributions of 10-day Average $PM_{2.5}$ Components for 55 Monitoring Events

concentration (μg/m^3)	mean	SD	min.	10%	25%	50%	75%	90%	max.
outdoor $PM_{2.5}$[a]	10.2	2.8	6.1	7.2	8.4	9.7	11.5	13.7	22.0
indoor $PM_{2.5}$[a]	8.1	2.2	3.0	5.6	6.7	8.0	9.6	10.6	13.4
indoor-generated indoor $PM_{2.5}$[b]	1.8	1.8	0.0	0.0	0.3	1.3	3.1	4.3	7.6
outdoor-generated indoor $PM_{2.5}$[b]	6.3	1.8	2.1	3.9	5.1	6.5	7.2	8.4	10.6
% of Indoor $PM_{2.5}$ Generated Outdoors[b]	78.7	16.9	40.2	54.0	66.6	80.5	94.5	100.0	100.0

[a] Measured using $HI_{2.5}$. [b] Modeled using F_{inf} estimates and $HI_{2.5}$ data.

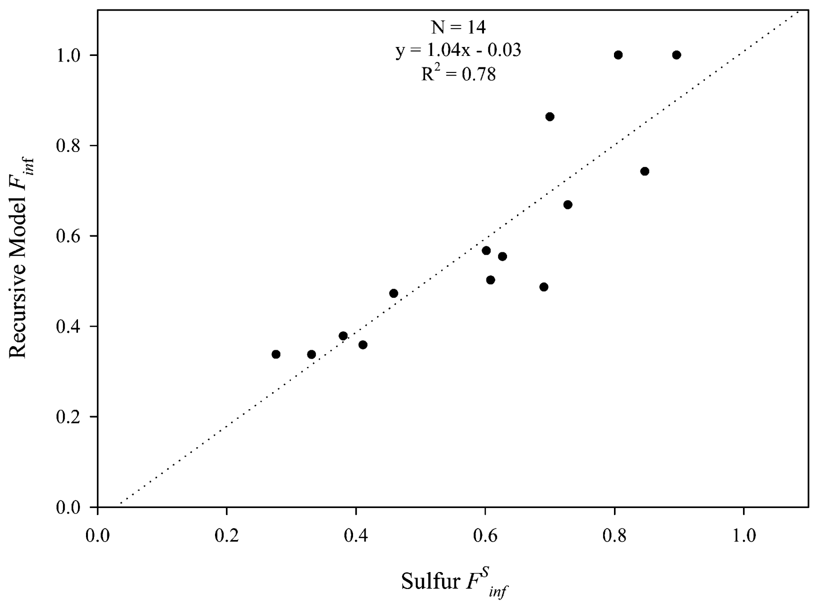

FIGURE 3. Comparison of infiltration estimated by RM and sulfur tracer.

is estimating F_{inf} and Φ as reliably as the linear model. The nonlinear model may not be sufficiently constrained to partition Φ into a and k, but we could not validate this with the current data.

The mean sulfur concentration was 0.44 $\pm$ 0.23 μg/m^3 outdoors and 0.28 $\pm$ 0.17 μg/m^3 indoors (p < 0.001). Regression results show a significant relationship between indoor and outdoor sulfur concentrations with a slope of 0.65 $\pm$ 0.01 ($R^2 = 0.78$; Figure 2, Supporting Information). For neph monitoring events during which sulfur data were available, the longitudinal R^2 (over the 10-day monitoring period for each monitoring event) between the 24-h indoor and outdoor sulfur concentrations ranged between 0.76 and 0.99, except for one residence that used an electrostatic precipitator (subject 39; $R^2 = 0.44$). These R^2 values agree with those reported in the PTEAM study (*11*), which ranged consistently between 0.8 and 0.9. The regression slope was used as an estimate of F^S_{inf} for $PM_{2.5}$ containing sulfur. Regression analysis for the RM F_{inf} estimates and sulfur F^S_{inf} estimates for 14 monitoring events showed strong agreement (Figure 3). Note that in our use of sulfur as a tracer of $PM_{2.5}$ infiltration we are assuming that sulfur infiltrates with the same efficiency as the rest of $PM_{2.5}$, which was recently validated with field monitoring data from 6 Boston homes (*38*).

Seasonal Variation. Significant seasonal differences were found for P, a, and k (p < 0.05). The average P, a, and k for residences monitored during the heating season were 0.89 $\pm$ 0.11, 0.37 $\pm$ 0.17, and 0.27 $\pm$ 0.18, respectively. During the nonheating season the average values of P, a, and k were 0.99 $\pm$ 0.03, 0.72 $\pm$ 0.82, and 0.12 $\pm$ 0.08, respectively. As expected, the average percent of days with at least one open window was lower during the heating season (42.1 $\pm$ 38.5%) than during the nonheating season (70.4 $\pm$ 37.5%, p < 0.01). Note that the seasonal difference in a remained statistically significant (p < 0.05), even when one outlier (a = 4.47 h^{-1}) was removed.

F_{inf} also showed seasonal differences (Figure 4). Residences monitored during the nonheating season had a mean F_{inf} of 0.79 $\pm$ 0.18, and residences monitored during the heating season had a mean F_{inf} of 0.53 $\pm$ 0.16 (p < 0.001). Within building types, private homes showed a significant seasonal difference in F_{inf} with a mean of 0.76 $\pm$ 0.19 during the nonheating season and 0.49 $\pm$ 0.11 during heating season (p < 0.001). The seasonal difference was also found for private apartments, with a mean of 0.82 $\pm$ 0.18 during nonheating season and 0.56 $\pm$ 0.22 during heating season (p < 0.05). Group homes did not show a statistically significant seasonal difference, due in part to the small sample size (N = 6). Considering that multiple monitoring events occurred during the same monitoring session, we evaluated the seasonal effect on F_{inf} in an ANOVA model that controlled for the session effect. The statistical significance of season remained unchanged.

Long et al. (*24*) found that during summertime all hourly F_{inf} were greater than 0.7 and in wintertime 73% of hourly F_{inf} were less than 0.7. The authors suggest that the difference is probably due to seasonal changes in home ventilation characteristics. This was supported by our finding of a higher frequency of window opening during the nonheating season and a higher F_{inf} on days with open windows (Table 3).

Sensitivity Analysis. We evaluated the effect of manually censoring rising edge points not identified by the algorithm. The data censored with the rising edge algorithm produced a mean F_{inf} of 0.66 $\pm$ 0.22, which agreed well ($R^2 = 0.99$) with the mean F_{inf} (0.65 $\pm$ 0.21) produced by manually censoring additional points. Thus, the rising edge algorithm is adequate, and manually censoring additional points is unnecessary.

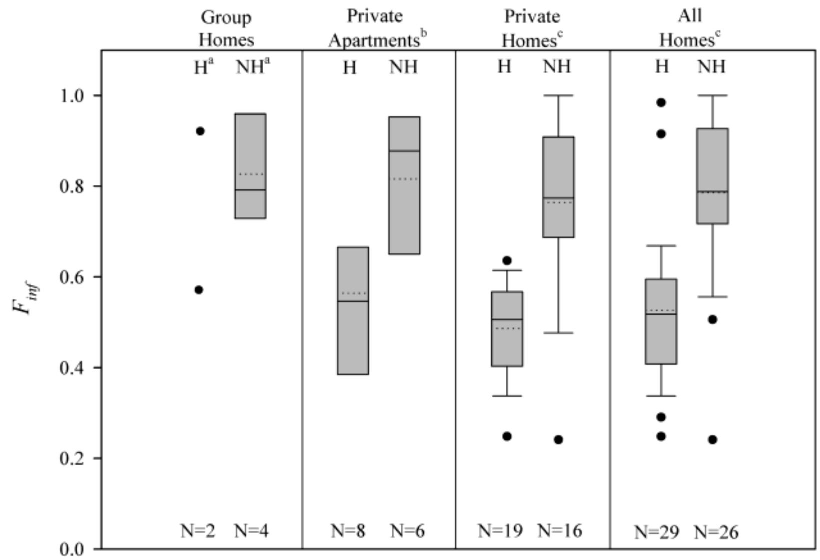

FIGURE 4. Boxplots of F_{inf} by home type and season. Key: [a] H = heating season, NH = nonheating season. [b] two-sample *t*-test; $p < 0.05$. [c] two-sample *t*-test; $p < 0.001$.

TABLE 6. Sensitivity Analysis for Parameter Estimates from Nonlinear Regression[a]

effect	category	censoring technique	no. of monitoring events	P	*a* (h^{-1})	*k* (h^{-1})	Φ (*a* + *k*; h^{-1})	F_{inf}
air exchange rate	measured	rising edge	5	0.81 (0.12)	0.51 (0.37)	0.36 (0.35)	0.87 (0.35)	0.46 (0.16)
	modeled			0.86 (0.13)	0.42 (0.15)	0.40 (0.23)	0.82 (0.32)	0.45 (0.13)
air exchange rate	measured	whole peak	5	*0.52*** (*0.16*)	0.51 (0.37)	*0.04** (*0.05*)	0.55 (0.35)	0.47 (0.12)
	modeled			*0.75*** (*0.24*)	0.27 (0.08)	*0.17** (*0.09*)	0.44 (0.17)	0.46 (0.10)
bounds used[b]	yes	rising edge	55	*0.94*** (*0.10*)	*0.54*** (*0.60*)	*0.20*** (*0.16*)	0.74 (0.59)	0.65 (0.21)
	no			*1.14*** (*0.36*)	*0.48*** (*0.51*)	*0.26*** (*0.15*)	0.74 (0.60)	0.66 (0.23)
bounds used[b]	yes	whole peak	54[c]	*0.92*** (*0.14*)	*0.41*** (*0.62*)	*0.14*** (*0.08*)	0.55 (0.62)	0.64 (0.21)
	no			*1.32*** (*0.70*)	*0.35*** (*0.53*)	*0.20*** (*0.10*)	0.55 (0.62)	0.65 (0.23)

[a] Standard deviations shown in parentheses. [b] $0 \le P \le 1$, $k \ge 0$, $a \ge 0$. [c] One monitoring event did not converge when "whole peak" censoring was used. Paired *t*-test: significant differences are italicized. **, $p < 0.01$; *, $p < 0.05$.

To assess the sensitivity of the P, a, k, and F_{inf} estimates to the model inputs, bounds, and censoring criteria, we compared values of P, a, k, and F_{inf} for each monitoring event under different modeling conditions (Table 6). First, we compared the parameters estimated with and without measured a for 5 monitoring events. The use of measured air exchange rates did not produce different estimates for any of the model parameters when rising edge censoring was used. However, when the whole peak censoring technique was used, both P and k were significantly different in the two model runs. If we assume that the use of measured values of a produces more accurate estimates of P and k because the model is only solving for 2 unknowns, then the use of rising edge censoring appears to be more appropriate because of the better agreement for P and k. We also examined the effect of using bounds in the nonlinear regression model and found that the parameters P, a, and k estimated using bounds were different from the unbounded estimates for both censoring techniques. The use of bounds produced higher estimates of P and k and lower estimates of a than the unbounded model. The bounded estimates appear more reasonable based on physical possibility and literature values. None of the modeling techniques examined in the sensitivity analysis had a significant impact on the F_{inf} or Φ estimates. This provides evidence that these estimates are robust. The individual estimates of P, a, and k, however, are less robust and are dependent on the modeling technique and the use of bounds in the nonlinear model.

Model Uncertainties. There were 4 extreme estimates when $F_{\mathrm{inf}} = 1$, $P = 1$, and $k = 0$. Because of the small likelihood of k being equal to 0 for $PM_{2.5}$, these 4 events required further examination. For these events the indoor light scattering level was greater than that outdoors for an average of 76 ± 18% of the hours, which was significantly higher ($p < 0.01$) than the mean for the remaining 51 monitoring events (26 ± 20%). The mean indoor to outdoor ratio from these 4 monitoring events, after whole peak censoring, was 1.24 ± 0.21, which was greater than the mean for the remaining 51 monitoring events (0.72 ± 0.21, $p < 0.01$). In addition, the average indoor to outdoor $HI_{2.5}$ ratio for these 4 monitoring events was 1.20 ± 0.30, again greater than the average ratio for the remaining 51 monitoring events (0.81 ± 0.26, $p < 0.01$). We tested the relationship between indoor and outdoor light scattering to mass ratios, and these 4 monitoring events did not differ from the other 51 events. These results indicate a potential problem during these 4 monitoring events that might be due to a problem with the calibration of one or both nephs. Although the nephs were all calibrated before each monitoring session and agreed well in lab collocation tests after each session (Supporting Information), a baseline drift remains a possibility.

Identifying indoor sources (or excluding daytime, indoor source periods) is a crucial prerequisite before running the recursive model. In this paper, we developed censoring algorithms for identifying indoor sources. These algorithms identified larger indoor peaks that have no corresponding large outdoor increase and low-level indoor sources with a very small increase (2.5×10^{-6} m^{-1} or approximately 1 $\mu g/m^3$) in indoor light scattering without a corresponding outdoor increase. In addition, we manually examined the time-series from each monitoring event for indoor sources not identified by the censoring algorithms. Our censoring method, however, does not identify constant indoor sources, such as those that may be generated from constantly operating furnaces. Unidentified indoor sources would be incorrectly considered to be outdoor particles that have infiltrated, thus causing an overestimation of F_{inf}. While this may be a potential problem, previously published studies indicate that PM levels resulting from indoor sources are generally "spikes" displaying a rapid increase and subsequent decay (*12*, *14*). Thus, constant indoor sources may account for a very small percentage of the total indoor contribution in most residences. This was further supported by the comparable indoor to outdoor light scattering ratios obtained using whole peak censoring and uncensored nighttime data.

Real-time data provide an excellent opportunity to re-create separate indoor time series for indoor- and outdoor-generated particles (Supporting Information). We showed that a recursive mass balance model can be successfully used to attribute indoor PM to its outdoor and indoor components and to estimate an average P, a, k, and F_{inf} for each residence. We estimated F_{inf} by applying a recursive model and nonlinear solution to light scattering measurements. These F_{inf} estimates were validated with the conventional sulfur-tracer technique. However, to apply such results to a broader population, more studies are needed to further validate our RM/light scattering results with conventional real-time gravimetric measurements.

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Supporting Information Available

Description of time activity diary and daily follow-up questionnaire, description of nephelometer performance, graph of indoor-outdoor sulfur relationship, and example of reconstructed indoor time-series. This material is available free of charge via the Internet at http://pubs.acs.org.

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