Journal of Exposure Science and Environmental Epidemiology (2007) 17, 468–477
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www.nature.com/jes

Evaluation of the recursive model approach for estimating particulate matter infiltration efficiencies using continuous light scattering data

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Quantifying particulate matter (PM) infiltration efficiencies (F_{inf}) in individual homes is an important part of PM exposure assessment because individuals spend the majority of time indoors. While F_{inf} of fine PM has most commonly been estimated using tracer species such as sulfur, here we evaluate an alternative that does not require particle collection, weighing and compositional analysis, and can be applied in situations with indoor sources of sulfur, such as environmental tobacco smoke, gas pilot lights, and humidifier use. This alternative method involves applying a recursive mass balance model (recursive model, RM) to continuous indoor and outdoor concentration measurements (e.g., light scattering data from nephelometers). We show that the RM can reliably estimate F_{inf}, a crucial parameter for determining exposure to particles of outdoor origin. The RM F_{inf} estimates showed good agreement with the conventional filter-based sulfur tracer approach. Our simulation results suggest that the RM F_{inf} estimates are minimally impacted by measurement error. In addition, the average light scattering response per unit mass concentration was greater indoors than outdoors; after correcting for differences in light scattering response the median deviation from sulfur F_{inf} was reduced from 15 to 11%. Thus, we have verified the RM applied to light scattering data. We show that the RM method is unable to provide satisfactory estimates of the individual components of F_{inf} (penetration efficiency, air exchange rate, and deposition rate). However, this approach may allow F_{inf} to be estimated in more residences, including those with indoor sources of sulfur. We show that individual homes vary in their infiltration efficiencies, thereby contributing to exposure misclassification in epidemiological studies that assign exposures using ambient monitoring data. This variation across homes indicates the need for home-specific estimation methods, such as the RM or sulfur tracer, instead of techniques that give average estimates of infiltration across homes.
Journal of Exposure Science and Environmental Epidemiology (2007) **17,** 468–477; doi:10.1038/sj.jes.7500539; published online 15 November 2006

Keywords: *particulate matter, infiltration, recursive model, light scattering, sulfur.*

Introduction

Much recent research has focused on quantifying the PM infiltration efficiency (F_{inf}) and the building characteristics and human behaviors that govern F_{inf} (Abt et al., 2000; Wilson et al., 2000; Long et al., 2001; Allen et al., 2003; Williams et al., 2003; Wallace et al., 2006; Wu et al., 2006). These studies all ultimately depend on an integrated solution of the differential mass balance equation (Nazaroff and Cass, 1989). This solution assumes a single air exchange zone throughout the house, uniform mixing of particles throughout the house, and no chemical or physical processes (e.g. coagulation or condensation) affecting the number or size of the particles. Given these assumptions, F_{inf}, defined as the fraction of the ambient concentration that penetrates indoors and remains suspended under steady-state conditions, depends on the particle penetration efficiency (P; unitless), the air exchange rate (a; h^{-1}), and the particle deposition rate (k; h^{-1}):

1. Abbreviations: a, air exchange rate (h^{-1}); a_1, coefficient describing the infiltration, deposition, and exfiltration of ambient $PM_{2.5}$; a_2, coefficient describing the deposition and exfiltration of indoor $PM_{2.5}$; AVR, sum of the absolute values of residuals; b_{sp}, particle light scattering coefficient; F_{inf}, infiltration efficiency; F_{inf}^{RM}, infiltration efficiency obtained using nephelometer data and the recursive model approach; F_{inf}^{S}, sulfur infiltration efficiency; HI, Harvard impactor; k, particle deposition rate (h^{-1}); Neph, radiance research nephelometer; OLS, ordinary least squares; P, particle penetration efficiency; PM, particulate matter; $PM_{2.5}$, particulate matter with an aerodynamic diameter less than 2.5 μm; RM, recursive model; S_i, indoor sulfur concentration; S_o, outdoor sulfur concentration; XRF, X-ray fluorescence; Φ, total particle loss rate (h^{-1})

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Received 26 June 2006; accepted 29 September 2006; published online 15 November 2006

$$F_{\text{inf}} = \frac{Pa}{a+k} \quad (1)$$

The sum of a and k is the total particle loss rate, Φ. As P and k depend on particle size (Mosley et al., 2001; Lai, 2002), F_{inf} is also particle size-dependent (Abt et al., 2000).

Quantifying F_{inf} in individual residences is important to PM exposure assessment. F_{inf} is used, in combination with time-location data, to estimate separately exposure to, and health effects associated with, outdoor-generated and indoor-generated PM (Ebelt et al., 2005; Koenig et al., 2005). Exposure studies have shown that F_{inf} varies both between residences and over time within a residence; and this variability in F_{inf} may result in exposure misclassification in epidemiologic studies that rely entirely on outdoor monitoring data to estimate exposure (Meng et al., 2005). In addition, epidemiologic studies have attempted to compare health effect estimates from different cities or from different seasons within a city (Dominici et al., 2003; Peng et al., 2005) to better understand the toxicities of major PM constituents. However, these comparisons of health effect estimates may be complicated by the variability in F_{inf} between cities and seasons.

The most common approach for estimating F_{inf} has been to use sulfur or sulfate as an ambient tracer, (Lewis, 1991; Suh et al., 1992; Leaderer et al., 1999; Geller et al., 2002; Sarnat et al., 2002; Na et al., 2004; Wallace et al., 2006; Wilson and Brauer, 2006) based on the assumptions of the absence of indoor sources (Lewis, 1991; Koutrakis et al., 1992; Ozkaynak et al., 1996; Geller et al., 2002; Na et al., 2004) and the similarity of its size distribution to $PM_{2.5}$ (Lewis, 1991; Wilson et al., 2000; Long and Sarnat, 2004). The latter assumption has been examined extensively because particles containing sulfur are typically between 0.2 and 0.7 μm (Horvath et al., 1996; Sarnat et al., 2002; Smolik et al., 2003; Martuzevicius et al., 2004). Other elements such as nickel, zinc, and iron have shown potential as tracers of various ranges of the ambient PM size distribution given detectable concentrations and no indoor sources (Koutrakis et al., 1992; Geller et al., 2002; Long and Sarnat, 2004).

As an alternative approach, we previously described the application of a recursive form of the mass balance model (recursive model; RM) to hourly indoor and outdoor light scattering data to estimate P, a, k, and F_{inf} for each of 44 residences in Seattle, WA (Allen et al., 2003). The deployment of continuous or semi-continuous (e.g. hourly) particle monitors requires neither the collection of a physical sample nor any compositional analysis. The ease of the data acquisition potentially allows F_{inf} to be estimated in more residences in the population. In addition, our approach uses censoring algorithms to identify and remove the influence of indoor particle sources; thus, the RM method does not require the exclusion of homes with indoor sources. This represents another potential advantage over the tracer approach because, although indoor sources of sulfur or sulfate are few, previous studies have reported elevated indoor concentrations in homes with smokers (Koutrakis et al., 1992), kerosene heaters (Koutrakis et al., 1992; Leaderer et al., 1999), pilot lights (Wallace et al., 2006), and humidifiers (Wallace et al., 2006). In addition, whereas sulfur is most representative of the infiltration behavior of 0.06–0.5 μm particles, the RM approach could be used to estimate the infiltration efficiencies of any particle sizes or components for which reliable continuous measurement methods exist.

Despite its promise, the application of the RM to light scattering data has some potential limitations. The recursive model cannot estimate daily values of F_{inf} due to an insufficient amount of data, particularly if indoor PM sources are frequent. Also, one must assume that the relationship between particle mass concentration and light scattering coefficient (b_{sp}) is the same indoors and outdoors. In addition, indoor sources that are not censored will be incorrectly treated as ambient particles and bias the estimates of F_{inf}. This paper presents an analysis to (1) evaluate the stability of the RM estimates of F_{inf} ($F_{\text{inf}}^{\text{RM}}$), P, a, k, and Φ in individual residences; (2) compare the $F_{\text{inf}}^{\text{RM}}$ estimates with those obtained using the sulfur ratio approach (F_{inf}^{S}) under different model and censoring scenarios; (3) compare the accuracy of the $F_{\text{inf}}^{\text{RM}}$ estimates with the accuracy of estimates from the simpler Random Component Superposition (RCS) model; and (4) evaluate the effect of indoor–outdoor differences in fine particle mass scattering efficiencies on the $F_{\text{inf}}^{\text{RM}}$ estimates.

Methods

Data Collection

The data used in this analysis were collected as part of a large PM exposure assessment study in Seattle, WA (henceforth called the "Seattle study"). The study design and data collection methods for the Seattle study have been described previously (Liu et al., 2003). In summary, indoor and outdoor $PM_{2.5}$ concentrations were measured at 108 study residences. The residences included private homes, private apartments, and group retirement homes. Monitoring occurred in both the heating (October–Febuary) and nonheating (March–September) seasons. Up to nine residences were monitored simultaneously during individual 10-day monitoring sessions and individual residences were monitored in up to three sessions. As some residences were monitored multiple times, we have previously defined a "monitoring event" as the monitoring of a single residence for a single 10-day monitoring session (i.e., one residence monitored twice is considered two monitoring events) (Allen et al., 2003).

We measured indoor and outdoor $PM_{2.5}$ on a 24-h basis during all monitoring events using 10-l/min single-stage inertial Harvard Impactors (HI; Air Diagnostics and

Engineering Inc., Naples, ME) with 37-mm Teflon filters. The precision of the HI $PM_{2.5}$ samples was 1.2 $\mu g/m^3$ (Liu et al., 2003). During 73 monitoring events, we also measured b_{sp} on a continuous basis indoors and outdoors using nephelometers (henceforth called "neph," Model 902 and 903, Radiance Research, Seattle, WA). The precision of the neph measurements was 3–8% (Liu et al., 2002). A subset of the Teflon filters was analyzed using energy-dispersive X-ray fluorescence (XRF) for a suite of 55 trace elements including sulfur. Our previous paper reported XRF-based sulfur results from 136 indoor–outdoor filter pairs from 14 monitoring events (Allen et al., 2003). This analysis includes 100 additional filter pairs from 15 monitoring events recently analyzed by XRF.

Quality Control

The quality control (QC) procedures for the filter data were previously described by Liu et al. (2003). Briefly, data were removed if the flow rate was not within 10% of the designated value or in the event of a pump failure, power outage, broken filter, or if tubing became disconnected. Consistent with our previous criterion, sulfur data were removed if the indoor sulfur concentration (S_i) exceeded the corresponding day's outdoor sulfur concentration (S_o) (Allen et al., 2003). Monitoring events with 5 or fewer valid indoor–outdoor sulfur pairs were excluded from the analysis. The final sulfur data set contained 195 daily indoor–outdoor pairs from 24 monitoring events at 21 unique residences. For continuous neph data, we used three QC criteria to include monitoring events that (1) achieved 50% data collection to ensure unbiased estimates; (2) had a significant ($P<0.05$) indoor-to-outdoor relationship during nonsource (night time) periods; and (3) had a median indoor-to-outdoor ratio <1 during nonsource periods (Allen et al., 2003). The final neph data set contained hourly indoor–outdoor pairs from 55 monitoring events at 44 unique residences (in which 24 monitoring events from 21 unique residences overlapped with those in the sulfur data set).

Recursive Modeling Approach

The RM is derived from the differential mass balance equation assuming constant a and piecewise constant outdoor concentrations during each discrete time step (Switzer and Ott, 1992). When applied to hourly light scattering data, the RM states that the average indoor particle light scattering coefficient (b_{sp}) during hour t ($(b_{sp})_t^{in}$) is equal to the sum of a fraction of the average outdoor particle scattering coefficient during the same hour ($(b_{sp})_t^{out}$), a fraction of the average indoor particle scattering coefficient remaining from the previous hour ($(b_{sp})_{t-1}^{in}$), and the scattering contribution from indoor sources (S_t^{in}) (Allen et al., 2003):

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

where

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

and

$$a_2 = \exp[-\Phi\Delta t] \quad (4)$$

As we use hourly averages of b_{sp}, $\Delta t = 1$ h and both a and k are in units of h^{-1}. This approach using hourly averaged values departs somewhat from the original derivation that assumed constant outdoor concentrations during each time step (Switzer and Ott, 1992); because the outdoor concentrations are generally slowly varying, we expected that this departure would generally have negligible effects. The influence of indoor sources is minimized by applying algorithms that identify hours when the indoor light scattering signal was influenced by indoor sources. These identified hours are then "censored" to eliminate the S_t^{in} term from Eq. (2):

$$(b_{sp})_t^{in} = a_1(b_{sp})_t^{out} + a_2(b_{sp})_{t-1}^{in} \quad (5)$$

The censored values are set to missing, and therefore these hours are not included in the regression. P, a, and k are estimated from nonlinear regression of Eq. (5) after substituting a_1 and a_2 with Eq. (3) and (4), respectively. F_{inf} can then be estimated using the P, a, and k estimates and Eq. (1) or by estimating the coefficients a_1 and a_2 via multiple linear regression of Eq. (5) and using the following relationship:

$$F_{inf} = \frac{a_1}{1 - a_2} \quad (6)$$

When the multiple linear regression intercept from Eq. (5) is forced to zero, these two methods give identical estimates of F_{inf} and Φ. Multiple linear regression can only estimate two of the three individual parameters (P, a, and k) influencing F_{inf}. As a was only measured in a small subset of the Seattle study residences, our previous work relied on nonlinear regression to estimate the three parameters simultaneously (Allen et al., 2003).

Testing the Stability of the Estimates

We evaluated the stability of the parameter estimates by performing grid searches of combinations of P, a, and k for each of the 55 monitoring events and examining the contours of the solution surfaces. These grid searches were repeated for combinations of F_{inf}^{RM} and Φ. The contour of the solution surface provided information on the uncertainty in the point estimates of P, a, k, Φ, and F_{inf}^{RM}. In addition, we used two goodness-of-fit criteria to fit the RM model and compared results for each of the 55 monitoring events. The goodness-of-fit criteria included the sum of the absolute values of the residuals (AVR) and the ordinary least squares (OLS) residuals. The AVR is less influenced by outliers than is OLS, which is the default in the nonlinear regression procedure.

We conducted simulations to test the sensitivity of the RM estimates to measurement error in the indoor and outdoor b_{sp}

data under the "best case" scenario; that is in situations where a is constant and known. Using the 10-day outdoor b_{sp} time series from one Seattle study monitoring event, we recreated the exact 10-day indoor b_{sp} time series using Eq. (5) and assuming $P = 1.00$, $a = 0.75\,h^{-1}$, and $k = 0.25\,h^{-1}$ (and therefore $F_{inf} = 0.75$). We then introduced measurement error into the b_{sp} values by adding a series of uniformly distributed random values (with mean = 0 and range = -0.18×10^{-5} to $0.18 \times 10^{-5}\,m^{-1}$) to the indoor b_{sp} values and a different series of uniformly distributed random values (also with mean = 0 and range = -0.18×10^{-5} to $0.18 \times 10^{-5}\,m^{-1}$) to the outdoor b_{sp} values. These ranges were selected to achieve average errors equal to approximately 5% of the average of the outdoor b_{sp} value used in the simulation, with 5% corresponding to the previously reported neph precision of 3–8%. After recreating the indoor b_{sp}, the first 24-h of indoor and outdoor b_{sp} data were removed to eliminate the influence of starting conditions on the simulated data. Next, we solved Eq. (5) via multiple linear regression to estimate a_1 and a_2 and estimated F_{inf} from Eq. (6). P and k were then estimated from the known value of a and the estimates of a_1 and a_2 as follows:

$$P = \left(\frac{1}{a}\right)\left(\frac{a_1}{1 - a_2}\right)(-\log(a_2)) \tag{7}$$

$$k = -\log(a_2) - a \tag{8}$$

The simulations were repeated 1,000 times, thus simulating 1,000 concurrent 10-day measurements indoors and outdoors at one residence.

As a final check on the sensitivity of the parameter estimates we compared estimates using hourly data with those obtained using 10-min data. This was necessary to evaluate the effect of using hourly data in the RM, which assumes constant outdoor concentrations during each time step.

Sulfur Tracer F_{inf} *Estimates*

Previously, reference values used to assess the accuracy of our F_{inf}^{RM} estimates were obtained by regressing the indoor sulfur concentration on the outdoor sulfur concentration at each residence and estimating F_{inf}^{S} from the regression slope (Allen et al., 2003). The sulfur slope approach was also used in a recent paper by Wilson and Brauer (2006). In contrast, Wallace et al. (2006) estimated F_{inf}^{S} by taking the average indoor sulfur concentration over all days at a residence divided by the average outdoor sulfur concentration over all days at the residence ($\overline{S_{in}}/\overline{S_{out}}$) and found this ratio provided a more trustworthy estimate of F_{inf}^{S} than the regression slope. In their sulfur tracer validation study, Sarnat and colleagues (2002) also used the indoor-to-outdoor sulfur ratio. We used the sulfur ratio as our primary method to estimate F_{inf}^{S} for our comparison with F_{inf}^{RM}. As a secondary check, we also compared F_{inf}^{RM} with F_{inf}^{S} estimated from the sulfur slope. Unless otherwise indicated, the F_{inf}^{S} results are those estimated using the sulfur ratio approach.

Evaluating Censoring Approaches for Indoor Sources

Our previous work evaluated two censoring approaches: "whole peak" and "rising edge" (Allen et al. 2003). In this paper we present the results of four combinations of multiple regression models and censoring techniques to find the most accurate estimates of F_{inf}^{RM}: (1) rising edge censoring and a regression model with no intercept term (i.e. with the intercept forced to 0); (2) rising edge censoring and a regression model with an intercept term; (3) whole peak censoring and a regression model without an intercept term; and (4) whole peak censoring and a regression model with an intercept term. We calculated the accuracy of the F_{inf}^{RM} estimates as the percent difference from the F_{inf}^{S} estimate for each monitoring event:

$$\%\ \text{difference} = \left|\frac{F_{inf}^{RM} - F_{inf}^{S}}{F_{inf}^{S}}\right| \times 100 \tag{9}$$

Evaluating the Effect of Mass Scattering Efficiencies on the F_{inf} *Estimates*

A key assumption in applying the RM to light scattering data is that the mass scattering efficiencies at individual homes are the same indoors and outdoors. First, we evaluated the relationship between b_{sp} and $PM_{2.5}$ across all homes by regressing 24-h averaged b_{sp} on the colocated $PM_{2.5}$ measurement indoors and outdoors. This provided a direct comparison with our previous pooled analysis based on the first year of data from the Seattle study (Liu et al., 2002). In addition, we calculated mass scattering efficiencies (in units of m^2/g) indoors and outdoors at each home by dividing the average b_{sp} by the corresponding $PM_{2.5}$ mass concentration measured with HI. We then repeated the F_{inf}^{RM}–F_{inf}^{S} comparisons after normalizing the indoor b_{sp} time series to the outdoor mass scattering efficiency (i.e., by dividing the indoor b_{sp} values on a given day by that day's indoor–outdoor ratio of mass scattering efficiencies).

Results

The 44 individual homes with valid indoor and outdoor neph data included 27 private homes, 12 private apartments, and five group retirement facilities. The median hourly b_{sp} values indoors and outdoors were $2.32 \times 10^{-5}\,m^{-1}$ (corresponding to $\sim 8.1\,\mu g/m^3$ based on an indoor conversion factor of $\mu g/m^3 = (b_{sp} \times 10^5 + 0.09)/0.30$) and $2.55 \times 10^{-5}\,m^{-1}$ (or $\sim 10.2\,\mu g/m^3$ based on an outdoor conversion factor of $\mu g/m^3 = (b_{sp} \times 10^5 + 0.31)/0.28$), respectively. The ratio of indoor to outdoor b_{sp} at individual homes ranged between 0.38 and 1.89, with an average of 0.97. The summary statistics of

Table 1. Indoor–outdoor relationships for 24-h $PM_{2.5}$ and sulfur concentrations during 24 monitoring events (all concentrations are in $\mu g/m^3$) sorted by descending indoor-outdoor sulfur ratio.

Monitoring event	N	$PM_{2.5}$ (measured w/harvard impactor)				Sulfur (measured by XRF)					
										I/O regression	
		Mean indoor conc.	Mean outdoor conc.	I/O corr.	I/O ratio	Mean indoor conc.	Mean outdoor conc.	I/O corr.	I/O ratio	Slope	Intercept[a]
M03	9	9.05	8.74	0.97	1.04	0.51	0.58	0.99	0.88	0.85	0.02
M01	8	11.81	10.40	0.87	1.13	0.70	0.81	0.95	0.86	0.99	−0.11
M44	10	7.56	7.23	0.35	1.04	0.39	0.48	0.96	0.81	0.93	−0.06
M02	7	7.58	7.69	0.93	0.99	0.44	0.55	0.99	0.80	0.90	−0.05
T63	10	10.56	10.05	0.75	1.05	0.39	0.51	0.98	0.77	0.85	−0.04
S21	8	13.66	9.21	0.82	1.48	0.40	0.52	1.00	0.76	0.67	**0.05**
S57	10	7.16	8.68	0.96	0.82	0.47	0.62	0.97	0.76	0.70	0.04
H25	6	5.95	6.84	0.82	0.87	0.33	0.44	0.97	0.74	0.71	0.01
H28	6	7.50	7.89	0.87	0.95	0.36	0.48	1.00	0.74	0.76	−0.01
S33	9	8.62	8.40	0.58	1.03	0.46	0.62	1.00	0.73	0.79	**−0.04**
H02	6	7.51	6.96	0.85	1.08	0.38	0.53	0.86	0.72	1.14	−0.22
H31	8	10.44	8.01	0.93	1.30	0.30	0.43	0.98	0.68	0.70	−0.01
M05	9	10.32	13.74	0.62	0.75	0.31	0.48	0.99	0.65	0.61	0.02
H05	7	10.35	14.17	0.89	0.73	0.31	0.49	0.92	0.64	0.69	−0.03
H39	10	8.47	7.59	0.49	1.12	0.26	0.41	0.98	0.64	0.60	0.02
M28	6	9.05	11.24	0.93	0.80	0.20	0.30	0.98	0.64	0.60	0.01
M29	10	6.49	10.51	0.98	0.62	0.28	0.44	0.97	0.64	0.60	0.02
M07	10	10.59	22.03	0.95	0.48	0.38	0.65	0.98	0.59	0.53	0.04
H19	9	5.88	8.73	0.63	0.67	0.23	0.41	0.86	0.55	0.70	−0.06
M26	8	7.88	12.54	0.50	0.63	0.22	0.47	0.94	0.48	0.50	−0.01
H14	7	4.63	10.94	0.83	0.42	0.21	0.44	0.88	0.47	0.38	0.04
M42	8	6.08	9.30	0.94	0.65	0.20	0.43	0.96	0.47	0.46	0.01
M18	6	6.85	15.23	0.91	0.45	0.20	0.49	0.90	0.41	0.34	0.03
F47	8	4.04	6.67	0.01	0.60	0.13	0.37	0.83	0.34	0.28	0.02
Mean	8.1	8.25	10.12	0.77	0.86	0.34	0.50	0.95	0.66	0.68	−0.01
SD	1.5	2.31	3.48	0.24	0.27	0.13	0.11	0.05	0.14	0.21	0.06
Min	6.0	4.04	6.67	0.01	0.42	0.13	0.30	0.83	0.34	0.28	−0.22
Median	8.0	7.73	8.98	0.86	0.85	0.32	0.48	0.97	0.67	0.70	0.01
Max	10.0	13.66	22.03	0.98	1.48	0.70	0.81	1.00	0.88	1.14	0.05

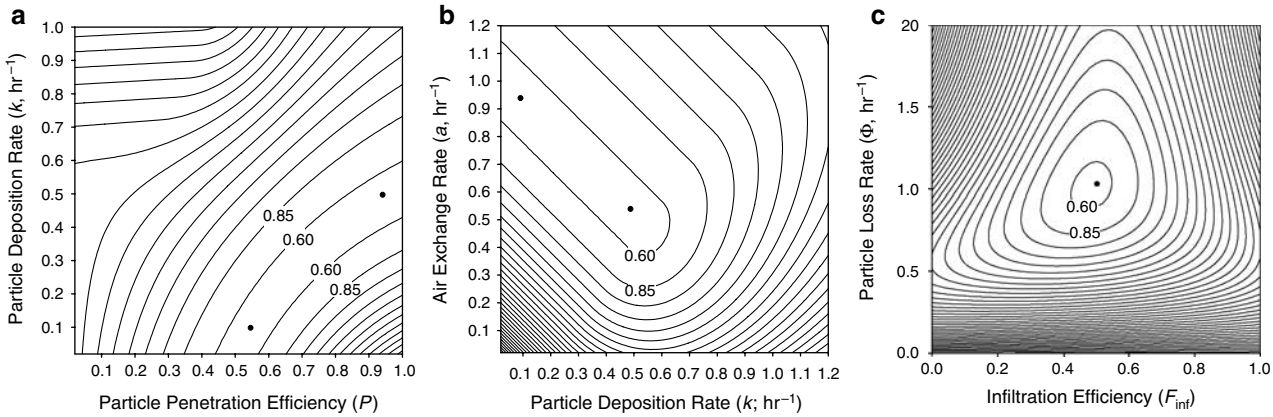
[a]Bold indicates intercept significantly ($P<0.05$) different from zero.

indoor and outdoor concentrations of gravimetric $PM_{2.5}$ and sulfur are presented in Table 1. Consistent with our previous findings, the high indoor–outdoor correlations (median = 0.97) and small (median = $0.02 \pm 0.06\,\mu g/m^3$), nonsignificant intercepts (23 of 24 not significantly > 0) indicate an absence of indoor sulfur sources in most of these residences.

Stability of the Parameter Estimates

The solution surfaces for all monitoring events had the same general appearance as that shown in Figure 1 from one representative residence. Figure 1a shows the residual sum of squares (SS) for various combinations of P and k, while Figure 1b shows the SS for combinations of k and a at this residence (in Figure 1a and b, for each combination of plotted parameters, the third parameter was set to the value that provided the best model fit). The two filled circles in Figures 1a and b indicate that two combinations of P, a, and k produced identical SS. The surfaces for the P/k and a/k combinations were flat and lacked a unique solution, whereas the solution surface for F_{inf}^{RM} and Φ showed a steeper gradient (Figure 1c) that resulted in a unique solution. This suggests larger uncertainties in the estimates of P, a, and k than in the estimates of Φ and F_{inf}^{RM}. For example, the area of the solution surface with residual sum of squares $\leq 0.60 \times 10^{-8}\,m^{-2}$ includes values of P, a, and k ranging between 0.44 and 1 (when bounded at 1), 0.38–1.14 h^{-1}, and 0–0.62 h^{-1} (when bounded at 0), respectively. In contrast, the same areas of the surface for Φ and F_{inf}^{RM} contain narrower ranges of values: 0.9–1.2 h^{-1} for Φ and 0.42–0.55 for F_{inf}^{RM}.

Figure 2 shows comparisons of the point estimates of P, a, k, Φ, and F_{inf}^{RM} for 54 monitoring events from two goodness-of-fit criteria: OLS and AVR (note that one monitoring event



Notes: (1) Plot a: for each combination of k and P, a is the value between 0.02 and 1.6 h^{-1} that produced the lowest sum of squares.
(2) Plot b: for each combination of a and k, P is the value between 0.02 and 1 that produced the lowest sum of squares.
(3) In a-c the contour lines increase by 0.25 beyond 0.60; black dots represent minimum sum of squared residuals (0.54 x 10^{-8} m^{-2}).

Figure 1. Contour plots of the residual sum of squares ($\times 10^{-8}\,m^{-2}$) for combinations of (a) k and P; (b) a and k; and (c) Φ and F_{inf} for one monitoring event.

with estimates of $a > 2\,h^{-1}$ and $k > 2\,h^{-1}$ is not shown in Figure 2). When multiple parameter combinations provided equally good model fit, Figure 2 shows the estimates from the combination with P estimated closest to 1. P, a, and k are sensitive to the goodness-of-fit criterion, whereas Φ is less sensitive and the F_{inf}^{RM} estimates are the most robust.

As expected based on the solution surfaces, the estimates of F_{inf} were much less sensitive to measurement error than the estimates of P and k (Figure 3), even assuming that a is constant and known. The mean ($\pm 2\sigma$) of the distribution of 1000 F_{inf}^{RM} estimates was 0.75 ± 0.01, whereas for P and k the estimates were 1.01 ± 0.10 and $0.26 \pm 0.10\,h^{-1}$, respectively.

The estimates of P, a, and k demonstrated some sensitivity to the averaging time of the indoor and outdoor neph data. We ran the nonlinear RM on the 10-min neph data and compared the estimates with those previously obtained using hourly data. The correlation coefficients between estimates using the two averaging times were 0.52, 0.70, and 0.64 for P, a, and k, respectively. Therefore, although hourly data and the RM do not allow for reliable estimates of P, a, and k, we cannot rule out the possibility that shorter averaging times in combination with measurements of a will allow the RM to reliably estimate P and k. F_{inf}^{RM} was much less sensitive to the averaging time; the correlation between estimates using 10-min data and those using hourly data was 0.96. The remainder of this paper will focus on hourly data, since our primary goal is to evaluate our previously used method.

Evaluating Censoring and Modeling Approaches

To identify the best method for estimating F_{inf}^{RM}, the F_{inf}^{s} estimates were compared against the F_{inf}^{RM} estimates from multiple linear regression models with and without an intercept, each using two different censoring techniques. Under both censoring scenarios, the no intercept models showed better agreement with F_{inf}^{s} (median % differences: 11.1–14.6%; R^2: 0.62–0.64) than the intercept models (median % differences: 21.0–28.0%; R^2: 0.38–0.47). The same conclusion was drawn when F_{inf}^{s} was estimated using the indoor–outdoor slope; again the no intercept models showed better agreement with F_{inf}^{s} (median % differences: 12.5–13.0%; R^2: 0.72–0.73) than the intercept models (median % differences: 18.2–19.6%; R^2: 0.52–0.65). In addition, the F_{inf}^{RM} estimates from the intercept models were more sensitive to the influence of individual hours. Using rising edge censoring and the intercept model, 37 monitoring events had at least 1 h that, when removed, resulted in a $\geq 5\%$ change in F_{inf}^{RM} estimate. In contrast, under the no intercept model 13 monitoring events had one or more such hours.

The small difference in F_{inf}^{RM}–F_{inf}^{s} agreement between the rising edge and whole peak censoring approaches under the no intercept model was due almost entirely to one monitoring event. When rising edge censoring was used in the no intercept model, the estimate of F_{inf}^{RM} for this monitoring event was 0.93, whereas whole peak censoring in the no intercept model produced an F_{inf}^{RM} estimate of 0.74. Further examination of the data from this monitoring event revealed that 1 h during the decay of a large indoor peak was highly influential on the estimate of F_{inf}^{RM}. Censoring this single influential hour in addition to rising edge censored points resulted in an F_{inf}^{RM} estimate of 0.65 (a decrease of 30%). The influential point occurred during an hour in which the indoor b_{sp} decreased from $50.2 \times 10^{-5}\,m^{-1}$ (or approximately $170\,\mu g/m^3$) to $12.1 \times 10^{-5}\,m^{-1}$ (or approximately $40\,\mu g/m^3$). This suggests that in rare cases of extremely high indoor concentrations, the decay of the indoor peak may be

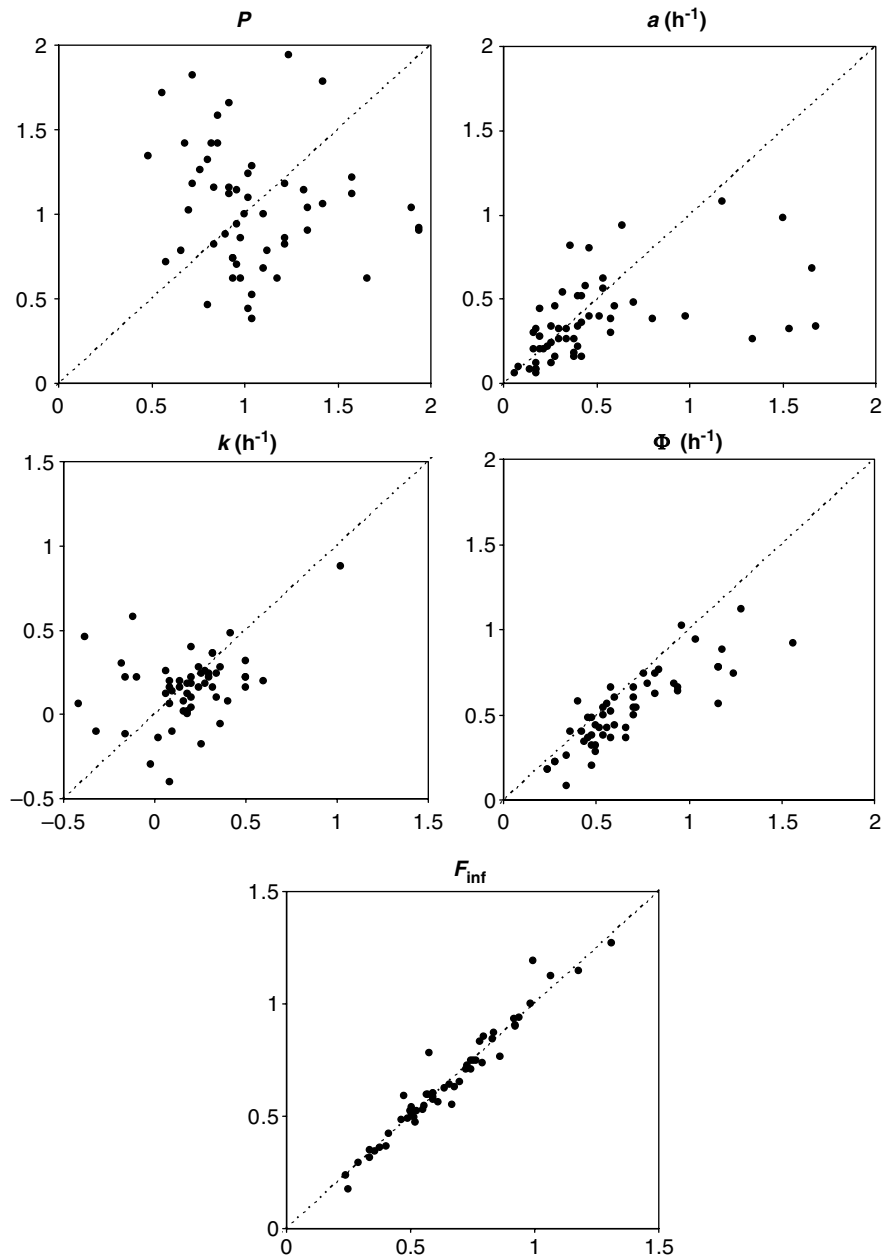


Figure 2. Comparisons of parameter estimates using different goodness-of-fit criteria and nonlinear regression to solve the RM.

particularly influential on the $F_{\mathrm{inf}}^{\mathrm{RM}}$ estimate. In addition to our rising edge censoring criteria, users of this RM method should consider the use of an additional censoring criterion (e.g. if $(b_{sp})_{t-1}^{\mathrm{in}} - (b_{sp})_{t}^{\mathrm{in}} > 10^{-4}\mathrm{m}^{-1}$) to remove the effect of such potentially influential time periods.

Comparison of the Recursive Model with the Random Component Superposition Model

Ott et al. (2000) proposed the RCS model in which indoor PM concentrations are regressed on outdoor PM concentrations; the model's slope is then interpreted as F_{inf} and the intercept indicates the indoor-generated concentration. We estimated F_{inf} in individual homes using the RCS model applied to (1) 1-h averaged neph data, (2) 24-h averaged neph data, and (3) 24-h gravimetric $PM_{2.5}$ concentrations measured with HI. We then compared these estimates with $F_{\mathrm{inf}}^{\mathrm{s}}$ to determine if the $F_{\mathrm{inf}}^{\mathrm{RM}}$ estimates were more or less accurate than those from the simpler RCS model.

The RCS model produced inaccurate estimates of F_{inf}. There was poor agreement between $F_{\mathrm{inf}}^{\mathrm{s}}$ and the RCS model

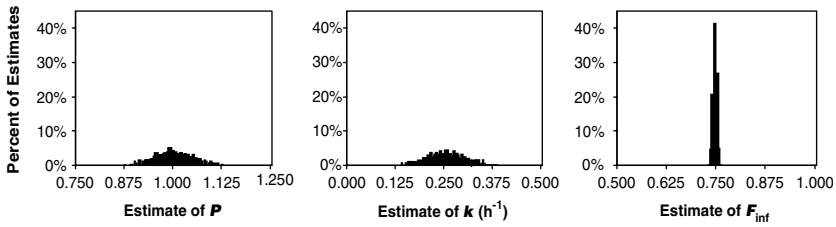


Figure 3. Distributions of 1000 estimates of P, k, and F_{inf} at 5% measurement error. True values used in simulations were $P = 1.00$, $k = 0.25\,h^{-1}$, and $F_{inf} = 0.75$.

estimates derived from hourly neph data (median % difference = 31.2%), daily neph data (38.5%), and daily $PM_{2.5}$ data (39.2%). Similarly, when F^{s}_{inf} was estimated from the indoor–outdoor sulfur slope, the agreement was again poor for RCS model estimates from hourly neph data (median % difference = 33.0%), daily neph data (23.0%), and daily $PM_{2.5}$ data (37.3%). Thus, the estimates from the RCS model were less accurate than the RM estimates, which had median % differences of 11–15%.

Effect of Mass Scattering Efficiencies on the F_{inf} *Estimates*

Differences were observed between indoor and outdoor mass scattering efficiencies. Pooling data from all monitoring events, the slope and intercept of regressing 24-h average outdoor b_{sp} on outdoor $PM_{2.5}$ were $2.81\,m^2/g$ and $-0.31 \times 10^{-5}\,m^{-1}$, respectively. Indoors, the regression slope was $2.98\,m^2/g$ and the intercept was $-0.09 \times 10^{-5}\,m^{-1}$. In an interaction model comparing the indoor and outdoor regression equations, the slopes ($P = 0.04$) and intercepts ($P < 0.01$) both showed significant differences. We calculated monitoring event-averaged efficiencies indoors and outdoors by dividing each monitoring event's 10-day average b_{sp} by the corresponding gravimetric $PM_{2.5}$ concentration measured with HI. Indoors, the average mass scattering efficiency across all 55 monitoring events was $2.83 \pm 0.42\,m^2/g$, which was higher than the outdoor average of $2.47 \pm 0.46\,m^2/g$ (paired t-test: $P < 0.001$). Indoor–outdoor ratios of scattering efficiencies ranged between 0.59 and 1.76, with a mean of 1.17. There was not a statistically significant difference between seasons; the average heating season ratio was 1.14 ± 0.20 ($N = 29$) and average nonheating season ratio was 1.21 ± 0.21 ($N = 26$; $P = 0.22$). Some of the indoor–outdoor differences in scattering efficiencies are attributable to the use in some homes of the older model 902 nephs (used indoors only), which were not calibrated directly but were adjusted to the model 903 nephs based on laboratory colocation experiments (Liu et al., 2002). For monitoring events in which 902 nephs were used (12 monitoring events) the mean indoor–outdoor scattering efficiency ratio was 1.29 ± 0.19, whereas for 903 nephs (43 monitoring events) the mean ratio was 1.14 ± 0.20 (2-sample t-test: $P < 0.05$).

We corrected for indoor–outdoor differences by calculating daily mass scattering efficiencies indoors and outdoors for each monitoring event. We then normalized the indoor mass scattering efficiency to that outdoors by dividing the indoor b_{sp} values for a given day by that day's indoor–outdoor mass scattering efficiency ratio. Using these corrected b_{sp} values in the RM resulted in improved agreement between the F^{RM}_{inf} and F^{s}_{inf} estimates (Figure 4): the median percent difference decreased from 14.6 to 10.9% and the R^2 increased from 0.62 to 0.74. Likewise, when compared to F^{s}_{inf} estimates from the sulfur slope method, use of the corrected b_{sp} values in the RM decreased the median percentage difference from 13.0 to 12.0% and increased the R^2 from 0.72 to 0.87. These results suggest that deviations from the assumption of equal light scattering-to-mass concentration relationships indoors and outdoors have a modest impact on the estimates of F^{RM}_{inf}.

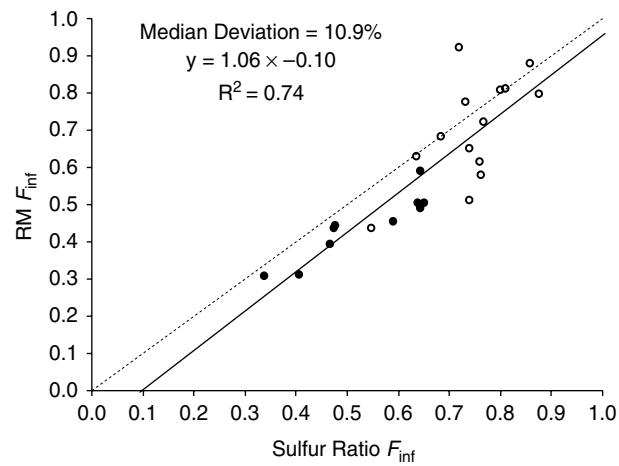


Figure 4. Relationship between F^{S}_{inf} and F^{RM}_{inf} after correcting for indoor–outdoor differences in 24-h mass scattering efficiencies.

Discussion

We have evaluated the RM technique for estimating F_{inf} (and the individual parameters that govern it) in individual homes. F_{inf} can be used in combination with time-location data to estimate ambient contribution fractions (α, the fraction of the ambient concentration to which an individual is exposed). Alternatively, α can be estimated directly as the personal-to-

ambient ratio (or slope) of an ambient tracer such as sulfur (Wilson and Brauer, 2006). While this direct estimation of α is one potential advantage of sulfur over the RM, personal PM measurements are generally not feasible on a large scale because such measurements are costly and can be burdensome for participants. Therefore, large-scale studies of personal exposure to ambient PM are likely to rely on F_{inf}, which can be estimated from indoor and outdoor data and does not require personal measurements.

Consistent with results from our earlier, preliminary sensitivity analysis (Allen et al., 2003), the individual estimates of P, a, and k were unstable. The estimates were highly dependent upon the goodness-of-fit criterion, particularly for P, and the solution surfaces for P, a, and k were quite flat, with multiple solutions producing equally good model fit. Other investigators have observed similar difficulties in estimating P and k (Bennett and Koutrakis, 2006). In contrast, the solution surfaces for F_{inf}^{RM} had much steeper gradients and had a unique solution. In addition, the estimates of F_{inf}^{RM} were more robust as they were less sensitive to the goodness-of-fit criterion.

Simulations with random 5% light scattering measurement errors resulted in precise estimates of F_{inf}, but large errors in k and P, under the best-case scenario in which a is constant and known. Our results suggest that under typical measurement conditions, estimates of P and k using the RM may be unreliable. Additional simulations at shorter time steps and with various outdoor concentration profiles are needed to determine conclusively whether the RM is able to estimate P and k.

We found that, when compared with the RCS model, the RM provided better agreement with F_{inf} as indicated by sulfur. In a study of agriculture burning exposure, Wu et al. (2006) estimated α and exposure to agriculture burning smoke using both the RCS model applied to 14-61 central site-indoor pairs of 12-h (0800–2000, 2000–0800) $PM_{2.5}$ measurements and the RM applied to hourly neph light scattering data. When compared with a specific marker (levoglucosan) for biomass burning smoke, Wu et al. (2006) reported better performance for the RM ($r = 0.75$) than the RCS model ($r = 0.58$) for estimating this ambient-generated exposure. In a recent study in North Carolina, Wallace et al. (2006) found that the RCS model applied to gravimetric $PM_{2.5}$ data was not able to reliably estimate F_{inf} for individual homes, even though up to 28 days of data were available per home. Taken together, these results suggest that the RM is a better option than the RCS model for estimating individual infiltration efficiencies.

We found slightly higher mass scattering efficiencies indoors compared to outdoors. This differs from our previous finding of no difference between the indoor and outdoor b_{sp}-$PM_{2.5}$ regression slopes based on data from the first year of the Seattle study (Liu et al., 2002). In addition to the effect of using an older model neph, this difference in mass scattering efficiencies may also be due to indoor-generated particles that scatter light more efficiently than the ambient aerosol (Brauer et al., 2000) and/or to the fact that the neph is especially sensitive to the size range in which particles infiltrate most efficiently (~ 0.1–$0.5\ \mu m$) (Waggoner and Weiss, 1980; Sarnat et al., 2006). Thus, the particle size-dependence of F_{inf} may alter the size distribution of the ambient aerosol as it comes indoors and result in an indoor aerosol with a slightly increased mass scattering efficiency. The agreement between F_{inf}^{RM} and F_{inf}^{S} improved somewhat after adjusting the indoor b_{sp} data to account for these differences in mass scattering efficiency. Therefore, although the uncorrected data produced acceptably accurate results, the application of the recursive model technique may require some colocated gravimetric measurements, particularly in settings where differences in the indoor and outdoor scattering-to-mass relationships (due to differences in particle size distribution, composition, density, etc.) are expected. For example, Sarnat et al. (2006) recently reported results from Los Angeles, an area with relatively high concentrations of outdoor ammonium nitrate. They found that volatilization of nitrate indoors led to differences in the indoor and outdoor $PM_{2.5}$ compositions. In this setting, some measurements of the indoor and outdoor mass scattering efficiencies would probably be necessary.

Although individual estimates of P, a, and k are unreliable, the RM applied to hourly data provides a reliable method for estimating particle infiltration efficiencies (the relevant parameter for estimating ambient exposure) in individual homes, even when light scattering data are used in place of fine particle mass concentrations. Although the sulfur tracer method has some important advantages, such as the ability to estimate F_{inf} on a daily basis and the ability to estimate α directly, the relative ease with which continuous data can be collected and the ability to reliably estimate F_{inf} in individual homes may make the recursive model approach a desirable alternative. This is particularly true in homes with indoor sulfur sources such as environmental tobacco smoke, kerosene heaters, pilot lights, or humidifiers.

Obtaining estimates of F_{inf} in individual homes will lead to an improved understanding of the variability in F_{inf} between homes and/or seasons, which is important for epidemiologic studies that rely on outdoor concentrations to assign exposure. Differences in F_{inf} between individual homes can contribute errors resulting in exposure misclassification in studies that rely on concentration gradients across space (i.e. between homes) or over time. The recursive model shows promise for improving our understanding of these important sources of PM exposure heterogeneity.

Acknowledgements

We are grateful to the study participants for their willingness to participate in this study and the field technicians for

collecting the data. This work was funded by the United States Environmental Protection Agency (EPA) through a cooperative agreement between the University of Washington and the EPA (CR82717701) and the EPA/UW Northwest Research Center for Particulate Air Pollution and Health (EPA Grant CR827355), and by the National Institute of Environmental Health Sciences (Grant No. P3OES07033). The views expressed in this paper do not necessarily reflect the views or policies of the EPA. Mention of trade names does not constitute endorsement or recommendation for use.

References

Abt E., Suh H.H., Catalano P., and Koutrakis P. Relative contribution of outdoor and indoor particle sources to indoor concentrations. *Environ Sci Technol* 2000: 34: 3579–3587.

Allen R., Larson T., Sheppard L., Wallace L., and Liu L.-J.S. Use of real-time light scattering data to estimate the contribution of infiltrated and indoor-generated particles to indoor air. *Environ Sci Technol* 2003: 37: 3484–3492.

Bennett D.H., and Koutrakis P. Determining the infiltration of outdoor particles in the indoor environment using a dynamic model. *Aerosol Sci* 2006: 37: 766–785.

Brauer M., Hirtle R., Lang B., and Ott W. Assessment of indoor fine aerosol contributions from environmental tobacco smoke and cooking with a portable nephelometer. *J Expos Anal Environ Epidemiol* 2000: 10: 136–144.

Dominici F., McDermott A., Zeger S.L., and Samet J.M. National maps of the effects of particulate matter on mortality: exploring geographical variation. *Environ Health Perspect* 2003: 111: 39–43.

Ebelt S.T., Wilson W.E., and Brauer M. Exposure to ambient and nonambient components of particulate matter – a comparison of health effects. *Epidemiology* 2005: 16: 396–405.

Geller M.D., Chang M.H., Sioutas C., Ostro B.D., and Lipsett M.J. Indoor/outdoor relationship and chemical composition of fine and coarse particles in the Southern California deserts. *Atmos Environ* 2002: 36: 1099–1110.

Horvath H., Kasahara M., and Pesava P. The size distribution and composition of the atmospheric aerosol at a rural and nearby urban location. *J Aerosol Sci* 1996: 27: 417–435.

Koenig J.Q., Mar T.F., Allen R.W., Jansen K., Lumley T., and Sullivan J.H., et al. Pulmonary effects of indoor- and outdoor-generated particles in children with asthma. *Environ Health Perspect* 2005: 113: 499–503.

Koutrakis P., Briggs S.L.K, and Leaderer B.P. Source apportionment of indoor aerosols in Suffolk and Onondaga Counties, New York. *Environ Sci Technol* 1992: 26: 521–527.

Lai A.C.K. Particle deposition indoors: a review. *Indoor Air* 2002: 12: 211–214.

Leaderer B.P., Naeher L., Jankun T., Balenger K., Holford T.R., and Toth C., et al. Indoor, outdoor, and regional summer and winter concentrations of PM_{10}, $PM_{2.5}$, SO_4^{2-}, H^+, NH_4^+, NO_3^-, NH_3, and nitrous acid in homes with and without kerosene space heaters. *Environ Health Perspect* 1999: 107: 223–231.

Lewis C.W. Sources of air pollutants indoors: VOC and fine particulate species. *J Expos Anal Environ Epidemiol* 1991: 1: 31–44.

Liu L.J.S., Box M., Kalman D., Kaufman J., Koenig J., and Larson T., et al. Exposure assessment of particulate matter for susceptible populations in Seattle. *Environ Health Perspect* 2003: 111: 909–918.

Liu L.J.S., Slaughter J.C., and Larson T.V. Comparison of light scattering devices and impactors for particulate measurements in indoor, outdoor, and personal environments. *Environ Sci Technol* 2002: 36: 2977–2986.

Long C.M., and Sarnat J.A. Indoor–outdoor relationships and infiltration behavior of elemental components of outdoor $PM_{2.5}$ for Boston-area homes. *Aerosol Sci Technol* 2004: 38: 91–104.

Long C.M., Suh H.H., Catalano P.J., and Koutrakis P. Using time- and size-resolved particulate data to quantify indoor penetration and deposition behavior. *Environ Sci Technol* 2001: 35: 2089–2099.

Martuzevicius D., Grinshpun S.A., Reponen T., Gorny R.L., Shukla R., and Lockey J., et al. Spatial and temporal variations of $PM_{2.5}$ concentration and composition throughout an urban area with high freeway density – the Greater Cincinnati Study. *Atmos Environ* 2004: 38: 1091–1105.

Meng Q.Y., Turpin B.J., Polidori A., Lee J.H., Weisel C., and Morandi M., et al. $PM_{2.5}$ of ambient origin: estimates and exposure errors relevant to PM epidemiology. *Environ Sci Technol* 2005: 39: 5105–5112.

Mosley R.B., Greenwell D.J., Sparks L.E., Guo Z., Tucker W.G., and Fortmann R., et al. Penetration of ambient fine particles into the indoor environment. *Aerosol Sci Technol* 2001: 34: 127–136.

Na K.S., Sawant A.A., and Cocker D.R. Trace elements in fine particulate matter within a community in western riverside county, CA: focus on residential sites and a local high school. *Atmos Environ* 2004: 38: 2867–2877.

Nazaroff W.W., and Cass G.R. Mathematical modeling of indoor aerosol dynamics. *Environ Sci Technol* 1989: 23: 157–166.

Ott W., Wallace L., and Mage D. Predicting particulate (PM_{10}) personal exposure distributions using a random component superposition statistical model. *J Air Waste Manage Assoc* 2000: 50: 1390–1406.

Ozkaynak H., Xue J., Weker R., Butler D., Koutrakis P., and Spengler J. *The Particle Team (PTEAM) Study: Analysis of the Data, Final Report, Vol III, EPA/600/R-95/098*. United States Environmental Protection Agency: Research Triangle Park, 1996.

Peng R.D., Dominici F., Pastor-Barriuso R., Zeger S.L., and Samet J.M. Seasonal analyses of air pollution and mortality in 100 US cities. *Am J Epidemiol* 2005: 161: 585–594.

Sarnat J.A., Long C.M., Koutrakis P., Coull B.A., Schwartz J., and Suh H.H. Using sulfur as a tracer of outdoor fine particulate matter. *Environ Sci Technol* 2002: 36: 5305–5314.

Sarnat S.E., Coull B.A., Ruiz P.A., Koutrakis P., and Suh H.H. The influences of ambient particle composition and size on particle infiltration in Los Angeles, CA, residences. *J Air Waste Manage Assoc* 2006: 56: 186–196.

Smolik J., Zdimal V., Schwarz J., Lazaridis M., Havranek V., and Eleftheriadis K., et al. Size resolved mass concentration and elemental composition of atmospheric aerosols over the eastern Mediterranean area. *Atmos Chem Phys* 2003: 3: 2207–2216.

Suh H.H., Spengler J.D., and Koutrakis P. Personal exposures to acid aerosols and ammonia. *Environ Sci Technol* 1992: 26: 2507–2517.

Switzer P., and Ott W. Derivation of an indoor air averaging time model from the mass balance equation for the case of independent source inputs and fixed air exchange rates. *J Exposure Anal Environ Epidemio* 1992: 2(Suppl 2): 113–135.

Waggoner A.P., and Weiss R.E. Comparison of fine particle mass concentration and light scattering extinction in ambient aerosol. *Atmos Environ* 1980: 14: 623–626.

Wallace L., Williams R., Suggs J., and Jones P. *Estimating Contributions of Outdoor Fine Particles to Indoor Concentrations and Personal Exposures: Effects of Household and Personal Activities. EPA/600/R-06/023*. US EPA, National Exposure Research Laboratory: Research Triangle Park, NC, 2006.

Williams R., Suggs J., Rea A., Sheldon L., Rodes C., and Thornburg J. The Research Triangle Park particulate matter panel study: modeling ambient source contribution to personal and residential PM mass concentrations. *Atmos Environ* 2003: 37: 5365–5378.

Wilson W.E., and Brauer M. Estimation of ambient and non-ambient components of particulate matter exposure from a personal monitoring panel study. *J Expos Sci Environ Epidemiol* 2006: 16: 264–274.

Wilson W.E., Mage D.T., and Grant L.D. Estimating separately personal exposure to ambient and nonambient particulate matter for epidemiology and risk assessment: why and how. *J Air Waste Manage Assoc* 2000: 50: 1167–1183.

Wu C.F., Jimenez J., Claiborn C., Gould T., Simpson C.D., Larson T., and Liu L.-J.S. Agricultural burning smoke in Eastern Washington: Part II. Exposure Assessment. *Atmos Environ* 2006: 40: 5379–5392.