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Estimated Hourly Personal Exposures to Ambient and Nonambient Particulate Matter Among Sensitive Populations in Seattle, Washington

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ABSTRACT

Epidemiological studies of particulate matter (PM) routinely use concentrations measured with stationary outdoor monitors as surrogates for personal exposure. Despite the frequently reported poor correlations between ambient concentrations and total personal exposure, the epidemiologic associations between ambient concentrations and health effects depend on the correlation between ambient concentrations and personal exposure to ambient-generated PM. This paper separates personal PM exposure into ambient and nonambient components and estimates the outdoor contribution to personal PM exposures with continuous light scattering data collected from 38 subjects in Seattle, WA. Across all subjects, the average exposure encountered indoors at home was lower than in all other microenvironments. Cooking and being at school were associated with elevated levels of exposure. Previously published estimates of particle infiltration (F_{inf}) were combined with time–location data to estimate an ambient contribution fraction (α, mean = 0.66 ± 0.21) for each subject. The mean α was significantly lower for subjects monitored during the heating season (0.55 ± 0.16) than for those monitored during the nonheating season (0.80 ± 0.17). Our modeled α estimates agreed well with those estimated with the sulfur-tracer method (slope = 1.08; R^2 = 0.67). We modeled exposure to ambient and nonambient PM with both continuous light scattering and 24-hr gravimetric data and found good agreement between the two methods. On average, ambient particles accounted for 48% of total personal exposure (range = 21–80%). The personal activity exposure was highly influenced by time spent away from monitored microenvironments. The median hourly longitudinal correlation between central site concentrations and personal exposures was 0.30. Although both α and the nonambient sources influence the personal–central relationship, the latter seems to dominate. Thus, total personal exposure may be poorly predicted by stationary outdoor monitors, particularly among persons whose PM exposure is

IMPLICATIONS

Many epidemiological studies have demonstrated associations between exposure to airborne particulate matter and adverse health effects. Because these studies have commonly used outdoor concentration data, a comprehensive understanding of the relationship between ambient concentrations and personal exposures is needed to estimate exposure misclassification. Separation of total personal exposure into its ambient and nonambient components will further enhance epidemiological studies in estimating toxicity and health effects from these two major PM source categories.

dominated by nonambient exposures, for example, those living in tightly sealed homes, those who cook, and children.

INTRODUCTION

Epidemiological studies have consistently shown an association between elevated 24-hr airborne particulate matter (PM) concentrations and adverse health effects. There is also recent evidence that PM concentration excursions of 1- to 4-hr may cause detrimental health effects.[1–3] These studies have typically used measurements collected outdoors at centrally located stationary monitoring sites as a surrogate for personal exposure. However, outdoor, or ambient, PM concentrations are sometimes poorly correlated with actual personal exposures.[4] In our exposure panel study in Seattle, WA, we observed a median longitudinal correlation between central site concentrations and personal exposures of 0.43, and the correlation seemed to depend on particle infiltration efficiency (F_{inf}) and the relative variability of ambient and nonambient exposures.[5] Higher correlations have been found between ambient PM concentrations and exposures to tracers of ambient-generated PM than between ambient concentrations and total personal exposures.[4,6,7] These results imply that the low correlations for total personal exposures are the result of variability in nonambient sources. An improved understanding of the relationship between ambient concentrations and personal exposures, particularly among susceptible subpopulations, has been identified as an important research need.[8]

Understanding the relationship between ambient concentrations and exposures to ambient-generated PM is ultimately important for epidemiologic and regulatory purposes.[9–11] Therefore, it is useful to separate PM exposures into ambient and nonambient components. Because these classes of particles originate from different sources, they may also have different toxicities;[12] thus, it is more appropriate to treat them as separate pollutants.[13]

To separate PM exposures into ambient and nonambient exposures, it is first necessary to estimate the fraction of the ambient PM concentration to which individuals are exposed. This ambient contribution fraction, α (sometimes called the ambient exposure attenuation factor[14]), has commonly been estimated by using the personal-to-outdoor ratio of a PM component with few indoor or personal sources, such as sulfur or sulfate.[4,6,11,15–17] Once α is estimated, it can be combined with concentration data and time–location information to separate total personal exposure into ambient and nonambient components. Few estimates of exposure to these classes of $PM_{2.5}$ and PM_{10} have been published.[11,18] In addition, despite the associations between short-term PM exposures and health effects, no studies have yet estimated separately personal exposures to, and the relation between, ambient and nonambient PM on a short-term (<24-hr) basis.

We previously estimated particle infiltration efficiencies (F_{inf}) and separated indoor $PM_{2.5}$ concentrations into outdoor- and indoor-generated components by using a recursive model.[19] This article builds on our previous work to model hourly personal exposure to ambient and nonambient PM for individual subjects. Combining these estimates with measured personal exposures, we were able to estimate PM concentrations generated by personal activities. Wilson et al. drew a distinction between the total personal activity exposure and the "personal cloud."[11] They defined the personal cloud as the difference between the concurrent personal and area-representative measurements (i.e., stationary measurements taken in the vicinity of the subject), whereas the personal activity exposure includes times when the subject is away from the stationary monitors. Real-time exposure estimates and time–location data allowed us to separate the personal activity exposure into the true personal cloud and the effect of time spent in unmonitored microenvironments.

METHODS

Data Collection

The data presented here were collected as part of a larger exposure assessment study conducted in Seattle between October 1999 and March 2002.[5] Monitoring occurred in both the heating (October through February) and the nonheating (March through September) seasons. During the first 2 years of the study (October 1999 through May 2001), monitoring was performed over 26 10-day monitoring sessions; year 3 (January 2002 through March 2002) consisted of six 5-day monitoring sessions and enrolled only subjects who had previously been monitored. Subjects were monitored in as many as 3 sessions and included elderly (>65 years old) subjects with chronic obstructive pulmonary disease (COPD) and coronary heart disease (CHD), elderly subjects with no cardiopulmonary disease (healthy), and pediatric subjects (ages 6–13) with asthma. These subjects were not selected according to a probability-based design, and therefore findings cannot be extrapolated to larger populations. Residential indoor and outdoor $PM_{2.5}$ concentrations were monitored over 24-hr with the 10-L/min Harvard Impactors ($HI_{2.5}$, Air Diagnostics and Engineering, Inc., Naples, ME) and at 10-min intervals with Radiance nephelometers (hereafter referred to as neph; model 903, Radiance Research, Seattle, WA). Personal $PM_{2.5}$ exposures were monitored over 24-hr with the 4-L/min Harvard Personal Environmental Monitors ($HPEM_{2.5}$, Harvard University, Boston, MA) and at 10-min intervals with the personal DataRAM (pDR, Thermo-MIE, Inc., Smyrna, GA) on 28 subjects. These pDR subjects were chosen based on their willingness to carry two instruments. A subset of indoor, outdoor, and personal $PM_{2.5}$ Teflon filters were further analyzed for a suite of 55 trace elements (including sulfur) with a modified long count energy dispersive X-ray fluorescence (XRF) methodology (Chester Labs, Portland, OR) for low mass samples. Because the analyses presented in this paper made use of combinations of measurements collected at different time scales (e.g., continuous and 24-hr) and in different locations (e.g., personal, indoor, and outdoor), Table 1 provides a summary of the amount of data used in each analysis after combining data sets and completing quality control (QC) procedures (described later). Because some residences and subjects were monitored more than once, we defined a "monitoring event" as the monitoring of a residence or subject for a single 5- or 10-day monitoring session (i.e., the same subject or residence monitored twice is considered two monitoring events).

The pDR is a small, lightweight nephelometer; detailed descriptions and evaluation results have previously been published.[5,20–23] In a previous article, we reported that the pDR's uncertainty and precision ranged between 0.5–1.3 $\mu g/m^3$ and 18–35%, respectively, at $PM_{2.5}$ levels ranging between 1.5 and 3.8 $\mu g/m^3$.[22] A correlation between pDR measurements and $PM_{2.5}$ gravimetric methods has been demonstrated (R^2 range = 0.44–0.66), although the pDR typically overestimates $PM_{2.5}$ gravimetric concentrations by 27–50%.[20,22,23] This overestimation is primarily caused by the fact that the pDR may respond to particles up to 10 μm and the calibration dust used by the manufacturer is denser (2.6 g/cm^3) than typical ambient aerosols. In our study, the pDR sampled passively without a size-selective inlet and was zeroed daily with a "zero bag" and filtered air pump supplied by the manufacturer.

The neph's performance has also been described elsewhere.[22] In short, the neph's uncertainty and precision ranged between 0–0.1 $\mu g/m^3$ and 3–8%, respectively, at concentrations ranging between 1.4 and 3.6 $\mu g/m^3$.[22] We have previously described the use of these neph data and a recursive model to estimate particle infiltration efficiency (F_{inf}) and separate indoor PM concentrations into indoor- and outdoor-generated components on an hourly basis.[19]

Concurrently with our monitoring work, PM was also monitored at a centrally located site, Beacon Hill, operated by the Washington State Department of Ecology. This site is approximately 5 miles from downtown Seattle and has been shown to be representative of regional $PM_{2.5}$ levels in the Seattle area.[24]

The subjects kept a time–location-activity diary (TAD), at 15-min resolution in years 1 and 2 and at 10-min resolution in year 3, and recorded the amount of time spent indoors at home, outdoors near home, in transit, at work, indoors away from home, and outdoors away from home. Their recorded activities were later coded into 54 activity categories. In addition, a daily follow-up questionnaire (DFQ) was used to gather additional information on potential particle-generating activities such as cooking (baking, frying, sautéing, broiling, and grilling) and cleaning. The TAD and DFQ entries were pooled into 30-min periods to combine with the pDR data and into 60-min periods to combine with the indoor- and outdoor-generated indoor concentration estimates.[19]

Quality Control

All samples were examined and all continuous data were screened based on our previously established QC criteria.[5,19,22] For pDR measurements, we included only days with at least 18 hr of pDR data and subjects with at least 4 such days. These criteria resulted in the removal of 6 subjects and left 246 person-days of pDR data from 38 monitoring events (from 28 unique subjects) for analysis (column 1 in Table 1). Because of the concern about a baseline or zero drift on pDRs, the mean difference between the neph and pDR measurements during nighttime (11 pm to 9 am) periods when no indoor sources were reported was used to calibrate the pDR concentrations each day. This adjustment was also necessary when we later used the pDR measurements to evaluate personal modeling results based on neph measurements. When no neph data were available, the pDR was calibrated with the 24-hr $HPEM_{2.5}$ concentration (N = 30 days). The average daily adjustment to the pDR was -0.2 ± 2.5 $\mu g/m^3$ (range = −8.7–5.5 $\mu g/m^3$). Nine days of pDR data had no valid concurrent neph or $HPEM_{2.5}$ data, and on such days we made no adjustment to the pDR data. For neph measurements, 11,753 hr (from 62 monitoring events) of the 16,473 hr of data collected from 84 monitoring events met our QC criteria[19] and were used in the recursive model to estimate F_{inf} and α (column 2 in Table 1).

The precision for the $HI_{2.5}$ and $HPEM_{2.5}$ data was 1.2 $\mu g/m^3$ and 2.2 $\mu g/m^3$, respectively, and the Pearson's r between $HI_{2.5}$ or $HPEM_{2.5}$ measurements and the collocated Federal Reference Method was ≥0.93.[5] The outdoor, indoor, and personal sulfur data were collected during 14 monitoring events (N = 94 personal–outdoor pairs and 107 indoor–outdoor pairs). The limit of detection for sulfur from the XRF analysis was 2.6 ng/m^3. We included only personal–outdoor sulfur pairs from days for which valid indoor and outdoor neph data were also available (N = 72 pairs). From these data, we removed days with personal-to-outdoor sulfur ratios greater than 1 (N = 3 pairs) and monitoring events that had fewer than 4 pairs of personal and outdoor sulfur data (N = 11 pairs). These steps left 58 pairs of personal and outdoor sulfur data for analysis (column 5 in Table 1). The data reduction for the

Table 1. Number of subjects, residences, and monitoring events by health status and season after QC procedures[a]

	Subjects Wearing pDRs	Neph Residences[b]	Subjects Wearing pDRs at Neph Residences Plus Time-Location Data[c]	Neph Residences with Personal, Indoor, and Outdoor Gravimetric Samplers Plus Time-Location Data[d]	Neph Residences with Personal and Outdoor Sulfur Data[e]	Neph Residences with Indoor and Outdoor Gravimetric Samplers, Indoor and Outdoor Sulfur Data, Plus Time-Location Data[f]	Subjects Wearing pDRs with Concurrent Central Site Neph Data[g]	Concurrent Personal, Home Outdoor, and Central Site Gravimetric Samplers Plus Neph Residences[h]
No. of subjects or sites by health status								
Healthy	5	8	2	6	0	0	5	3
COPD	10 (3)	13 (6)	6 (2)	10 (6)		1	10 (3)	7 (3)
CHD	11 (4)	17 (5)	7	16 (4)	5 (2)	5 (2)	11 (4)	14 (2)
Asthmatic	2	8 (4)	2	8 (4)	2	4 (2)	1	5 (4)
No. of subjects or sites by season								
Heating only	12 (2)	22 (3)	10	19 (3)	6 (1)	7 (2)	11 (2)	16 (3)
Nonheating only	11	13 (1)	5	11 (1)	2 (1)	2 (1)	11	7
Both seasons	5 (5)	11 (11)	2 (2)	10 (10)	0	1 (1)	5 (5)	6 (6)
Total subjects or sites	28 (7)	46 (15)	17 (2)	40 (14)	8 (2)	10 (4)	27 (7)	29 (9)
Total monitoring events	38	62	20	55	10	14	36	38
Total days	246	490	120	403	58	92	224	260

Note: A "monitoring event" refers to the monitoring of a residence or subject for a single 5- or 10-day monitoring session (i.e., the same subject or residence monitored twice is considered two monitored events); [a]Number in parentheses indicates the number of subjects or residences monitored multiple times; e.g., 10 (3) means that 3 of the 10 subjects were enrolled in more than one monitoring session; [b]Refers to residences with both indoor and outdoor neph; these data were previously used to model F_{inf};[19] [c]Data used for hourly (method 1) estimates of exposure components; [d]Data used for 24-hr (method 2) estimates of exposure components; gravimetric samplers include $HPEM_{2.5}$ and $HI_{2.5}$; [e]Data used to validate SSE model estimates of α; [f]Data used for 24-hr (method 3) estimates of exposure components and daily estimates of F_{inf}; gravimetric samplers include $HI_{2.5}$; [g]Data used to calculate hourly and 24-hr correlations between personal exposure and central site concentration. [h]Data used to calculate the effect of Ψ and α on correlations with central site; gravimetric samplers include $HPEM_{2.5}$ and $HI_{2.5}$.

indoor–outdoor sulfur data has been described previously and resulted in 98 valid indoor–outdoor sulfur pairs.[19]

Data Analysis

There were the following 4 major components to our data analysis: (1) characterize personal $PM_{2.5}$ exposure in various microenvironments and during various activities; (2) estimate an average α for each neph monitoring event, and evaluate the estimates with those determined by using the sulfur tracer method; (3) model exposure to ambient, indoor-generated, and personal activity $PM_{2.5}$; and (4) evaluate factors influencing the correlations between personal exposure and central site concentrations.

Microenvironmental Exposures. We used generalized estimating equation (GEE) models to determine the effect of microenvironments and activities on elevated particle exposures while accounting for autocorrelation and clustering.[25,26] For both total exposure (E = 30-min basis measured with the pDR) and personal activity exposure (E_{pact} = 60-min basis, discussed later) we constructed two models:

$$y = \beta_0 + \sum \beta_i x_i + \varepsilon \tag{1}$$

where y represents the exposures, β_0 are the model intercepts, x_i are the indicator variables, β_i are the model coefficients, and ε represents the model error. When examining the effect of microenvironments on exposure, x_i represented all microenvironments other than indoors at home. When examining the effect of activities on exposure, x_i represented the potential particle-generating activities. For the microenvironment and activities models, the coefficient β_0 represented the average exposure indoors at home and the average exposure during times when none of the potential particle-generating activities were reported. In our analysis, the GENMOD procedure in the SAS statistical program (Version 8, SAS Institute, Cary, NC) was used with an identity link function and an autoregressive working correlation structure.[27] Missing or deleted data were assumed to be missing at random. We included in the activity exposure models those activities that were expected to be associated with higher personal exposures, primarily those involving combustion (e.g., burning candles), resuspension (e.g., cleaning), or proximity to high particle concentrations (e.g., traveling by car or bus). In addition, because we previously found an association between being at school and elevated personal activity exposures,[5] we also included being in class or being at school during recess as variables in the exposure models.

The Ambient Contribution Fraction (α). For each subject, an average value of α, which depends on particle infiltration and the fraction of time spent outdoors, was calculated with our previously published F_{inf} estimates[19] and the TAD data:

$$\alpha = [F_o + (1 - F_o)F_{inf}] \tag{2}$$

where F_o is the fraction of time spent outdoors near home, in transit, or outdoors away from home. The F_{inf} estimates are averages for the entire 5- or 10-day monitoring event, and eq 2 assumes that F_{inf} is similar for all indoor environments encountered by the subject. The α estimates were evaluated against the exposure fraction estimates obtained by regressing the personal sulfur concentrations on the outdoor sulfur concentrations for each monitoring event.[4,6,11,16] The slope of this regression provides an estimate of α_S. Although sulfur best traces 0.06- to 0.5-μm particles, it accurately modeled the infiltration behavior of $PM_{2.5}$ in a recent study in 6 Boston homes.[17]

Modeling Exposure Components. Exposure to the ambient and nonambient $PM_{2.5}$ components was estimated with a source-specific exposure (SSE) model.[11] Total personal exposure (E) is the sum of ambient exposure (E_a) and nonambient exposure (E_{na}). Ambient exposure is equal to the ambient concentration (C_a) multiplied by the ambient contribution fraction. Thus, total personal exposure can be written as

$$E = \alpha C_a + E_{na} \tag{3}$$

Nonambient exposure (E_{na}) is the sum of exposure to indoor-generated PM (E_{ig}) and exposure to personal activity PM (E_{pact}).

E_a and E_{ig} were calculated with the recursive model F_{inf} estimates applied to hourly neph measurements (method 1) or the 24-hr $HI_{2.5}$ measurements (method 2). A subset of E_a and E_{ig} estimates were also obtained with the sulfur tracer F_{inf} applied to the 24-hr $HI_{2.5}$ measurements (method 3).

Method 1: Hourly exposure components were calculated for 20 monitoring events with the previously published reconstructed indoor-generated (C_{in}^{ig}) and infiltrated indoor concentrations (C_{in}^{inf}),[19] hourly outdoor neph data, and hourly reported F_o values.

$$E_a = (F_o)C_a + (1 - F_o)C_{in}^{inf} \tag{4}$$

$$E_{ig} = (1 - F_o)C_{in}^{ig} \tag{5}$$

It was necessary to use C_{in}^{inf} and C_{in}^{ig} because these estimates were reconstructed with the recursive model and account for the autocorrelation of hourly concentration data.[19] For 23% of the hours, C_{in}^{inf} was estimated to be greater than C_{in}. The average difference between C_{in}^{inf} and C_{in}^{ig} during these hours was 1.1 ± 1.1 μg/m^3 (minimum, 10th percentile, 90th percentile, and maximum = 0.0, 0.1, 2.4, and 9.9 μg/m^3, respectively). During these hours, C_{in}^{inf} was set equal to C_{in} and C_{in}^{ig} was set equal to 0.

Method 2: This method allowed us to estimate E_a and E_{ig} for the largest possible number of subjects. On a daily basis, E_a and E_{ig} were calculated for 55 monitoring events with estimates of F_{inf},[19] 24-hr indoor and outdoor $HI_{2.5}$ measurements, and reported daily F_o values:

$$E_a = (F_o)C_a + (1 - F_o)(C_a \times F_{inf}) \tag{6}$$

and

$$E_{ig} = (1 - F_o)[C_{in} - (C_a \times F_{inf})] \tag{7}$$

On days when ($C_a \times F_{inf}$) was greater than C_{in}, ($C_a \times F_{inf}$) was set equal to C_{in} and E_{ig} was set equal to zero. This occurred on 25% of the days. The average overestimation of the infiltrated concentration [($C_a \times F_{inf}$) − C_{in}] on these days was 1.3 ± 1.3 μg/m^3 (minimum, 10th percentile, 90th percentile, and maximum = 0.0, 0.1, 2.4, and 9.9 μg/m^3, respectively).

Method 3: The purpose of method 3 was to evaluate the method 1 estimates and the assumption of a constant F_{inf} in method 1. This method is similar to method 2 except that the F_{inf} used in eqs 6 and 7 were daily indoor-to-outdoor sulfur ratios for each subject. Again, when ($C_a \times F_{inf}$) was greater than C_{in}, ($C_a \times F_{inf}$) was set equal to C_{in} and E_{ig} was set equal to zero. This occurred on 16% of the days. The average overestimation of the infiltrated concentration on these days was 1.4 ± 1.4 μg/m^3 (minimum, 10th percentile, 90th percentile, and maximum = 0.0, 0.1, 2.4, and 9.9 μg/m^3, respectively).

Exposure that results from personal activities, E_{pact}, is defined as the difference between the measured exposure, E, and the modeled exposure, $\hat{E}$ (= $E_a + E_{ig}$). E_{pact} is thus the microenvironmental exposure encountered by subjects but not detected by stationary monitors, either because particles were generated in the immediate proximity of the subject (i.e., the personal cloud) or because the subject entered microenvironments without stationary monitors.

Longitudinal Correlations for Personal and Central Site Measurements. These longitudinal correlations (Pearson's r) were calculated with both short-term light scattering and 24-hr gravimetric data. From eq 3, the correlation between C_a and E is defined as the covariance of C_a and E, or $\alpha\sigma_{C_a}^2$, divided by the product of the standard deviations of C_a ($\sqrt{\sigma_{C_a}^2}$) and E ($\sqrt{\alpha^2\sigma_{C_a}^2 + \sigma_{E_{na}}^2}$) (assuming C_a and E_{na} are independent). Algebraic manipulation yields the following:

$$longitudinal\ r = \frac{1}{\sqrt{1 + \frac{\Psi}{\alpha^2}}} \tag{8}$$

where

$$\Psi = \frac{\sigma_{E_{na}}^2}{\sigma_{C_a}^2} \tag{9}$$

Ψ, which was given the symbol R in our previous article,[5] is a measure of the relative impact of nonambient exposure on the total personal exposure. We investigated the effects of Ψ and α on the longitudinal r for monitoring events not impacted by local outdoor sources ($p < 0.10$ for the correlation between central site $HI_{2.5}$ and home outdoor $HI_{2.5}$). Three Ψ outliers (>75th percentile + 1.5 × interquartile range) were excluded from the analysis. We evaluated the influence of subject and residential characteristics on Ψ by regressing the Ψ estimates on the percent of hours with cooking reported, the subject's age group (child or elderly adult), the use of an air cleaner, the fraction of days that the subject's home had at least one window open for any duration, the type of residence, and the season.

RESULTS

Hourly Personal Exposure Data

The 30-min pDR data collected from the 38 monitoring events over 246 person-days (N = 11,474) were right-skewed, with a geometric mean (GSD) of 6.9 μg/m^3 (2.6) (arithmetic mean ± standard deviation = 10.9 ± 19.1 μg/m^3). The ratio of the maximum 30-min concentration to the 24-hr average concentration ranged between 1.5 and 16.9 (mean = 4.3 ± 2.6, N = 246 days), indicating a wide range in the relative magnitude of the maximum daily exposure. The exposures observed in the subset of monitoring events chosen for pDR monitoring were lower than the exposures for all monitoring events observed during the first 2 years of the full Seattle panel study (geometric standard deviation [GM] personal exposures of 7.7–8.8 μg/m^3 for the adult groups based on $HPEM_{2.5}$ measurements).[5]

The relationship between the adjusted 24-hr average pDR and the collocated $HPEM_{2.5}$ concentration is shown in Figure 1. The pDR and $HPEM_{2.5}$ data agreed well with an R^2 of 0.74. The regression slope (1.30, standard error [SE] = 0.06) indicates that the pDR overestimates the $HPEM_{2.5}$ measurements by 30%. After excluding the

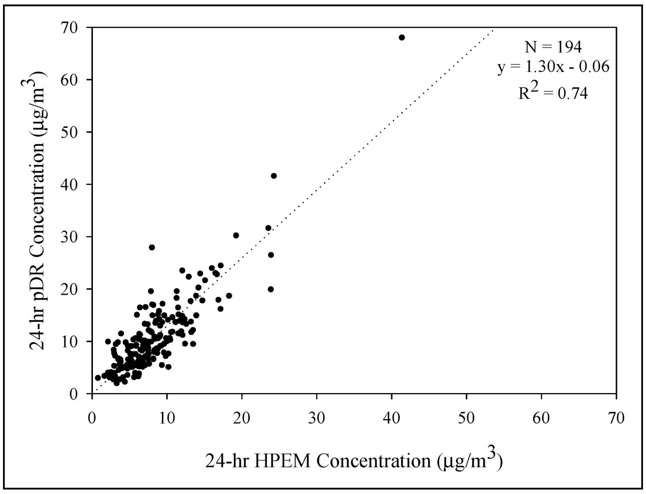

Figure 1. Relationship between collocated pDR and $HPEM_{2.5}$ concentrations.

highest pDR value, the R^2 in Figure 1 decreased to 0.66 and the slope (1.16, SE = 0.06) was still significantly greater than 1 ($p < 0.01$).

Microenvironmental Exposures

The average exposure, fraction of time spent, and fraction of exposure received in each microenvironment by health group are presented in Table 2. The amount of time spent in each microenvironment by the subset of monitoring events in the present analysis was in agreement with the results of the 109 subjects enrolled in the first 2 years of the full Seattle panel study.[5] The highest GM exposures for all four health groups were outdoors, either near home or away from home (excluding one subject who worked). PM exposure while in transit was also among the highest for pediatric subjects, possibly because of travel on diesel school buses. Adults received an average of at least 79% of their total PM exposure indoors at home, because of the large percentage of time spent there. In contrast, the two asthmatic children received half of their exposure indoors at home and 34% in other indoor environments (primarily school).

Relative to exposures encountered indoors at home, exposures at work were significantly elevated and exposures in transit were marginally elevated (Table 3a). Activities also played an important role in determining PM exposures. Table 3b shows the increase in exposure associated with various activities relative to times when none of these activities were reported. Cooking and being at school in class or recess significantly elevated personal exposure.

Ambient Contribution Fraction (α)

The average α was 0.66 ± 0.21 (range = 0.25–1.00) across all monitoring events (N = 62) and differed by season (Figure 2a) but not by health group (Figure 2b). Subjects monitored during the heating season (October through February, N = 36) had a significantly lower average α (0.55 ± 0.16) than those monitored during the nonheating season (March through September, N = 26) (0.80 ± 0.17, $p < 0.001$, 2-sample t test). This is caused by a lower average F_{inf} during the heating season.[19] The fraction of time spent outdoors (F_o) was only marginally different between the nonheating (0.08 ± 0.04) and heating seasons (0.06 ± 0.03) (p = 0.06).

For the days with concurrent personal and outdoor sulfur data (N = 58 days), the average sulfur concentrations were 0.25 ± 0.15 μg/m³ (range = 0.04–0.81 μg/m³) and 0.45 ± 0.25 μg/m³ (range = 0.18–1.22 μg/m³), respectively. The average concurrent indoor and outdoor sulfur concentrations (N = 98 days) were 0.28 ± 0.17 μg/m³ (range = 0.11–0.93 μg/m³) and 0.44 ± 0.23 μg/m³ (range = 0.14–1.22 μg/m³), respectively. The α estimates from the sulfur tracer method (α_s) averaged 0.54 ± 0.17 (range = 0.23–0.79). The longitudinal R^2 for personal and outdoor sulfur concentrations for individual subjects ranged between 0.70 and 0.99 (median R^2 = 0.94). The R^2 between the α estimates from eq 2 and the sulfur tracer α_S estimates was 0.67, with a regression slope of 1.08 (SE = 0.27; Figure 3). The slope in Figure 3 is not significantly different from 1 (p = 0.76) and the intercept is not significantly different from 0 (p = 0.65).

To further assess the effect of using an average value of F_{inf} (as opposed to a day-specific estimate) in method 1, we examined the coefficient of variation (CV) for each monitoring event with the daily F_{inf} estimates from the indoor-to-outdoor sulfur ratio. The CV ranged between 6% and 34% for individual monitoring events, with a median value of 12%. Twelve of 14 monitoring events had a CV below 25%. Between-subject variability accounted for the majority (52%) of the total variability in the daily sulfur F_{inf} estimates. This finding, combined with the previously reported high longitudinal correlations between indoor and outdoor sulfur levels,[19] justifies the assumption of constant F_{inf} within subject over time and our use of an average F_{inf} in methods 1 and 2 to model the individual exposure components.

Exposure Components

Hourly Estimates (Method 1). On average, E_a made the largest contribution to the total exposure. For the 20 monitoring events with method 1 exposure estimates, the GM hourly personal exposure measured with the pDR was 7.5 μg/m³ (2.5) (arithmetic mean = 11.1 ± 15.2 μg/m³). Across all monitoring events, the GM of hourly E_a was 3.9 μg/m³ (2.5; arithmetic mean = 5.6 ± 4.6 μg/m³). Excluding hours when E_{ig} was estimated to be zero (see description of Method 1), the GM E_{ig} concentration was 1.4 (3.4;

Table 2. Percent of time spent, average concentration, and percent exposure received in each microenvironment.

Location	% Time Spent Mean (SD)	Exposure ($\mu g/m^3$) GM (GSD)	Exposure ($\mu g/m^3$) AM (SD)	% Exposure Mean (SD)
Healthy Subjects (5 Monitoring Events; $N = 35$ Subject-Days)				
Indoors at home	84.7 (10.1)	7.3 (2.6)	10.9 (12.8)	79.5 (15.4)
Outdoors at home	1.0 (1.4)	18.4 (1.7)	26.9 (24.6)	2.4 (4.0)
In transit	4.1 (3.0)	10.1 (2.6)	17.2 (36.6)	5.8 (4.5)
At work	0.0 (0.0)	—	—	—
Outdoors away from home	1.4 (2.8)	12.1 (1.5)	13.2 (6.4)	1.6 (3.3)
Indoors away from home	8.8 (6.4)	9.2 (2.7)	14.7 (17.6)	10.6 (10.1)
CHD Subjects (16 Monitoring Events; $N = 94$ Subject-Days)				
Indoors at home	88.8 (7.4)	6.9 (2.5)	11.0 (24.9)	82.7 (13.4)
Outdoors at home	0.6 (1.1)	8.0 (2.0)	9.6 (4.9)	0.9 (1.9)
In transit	3.5 (3.2)	9.4 (2.4)	15.4 (33.8)	5.3 (5.8)
At work	0.0 (0.0)	—	—	—
Outdoors away from home	1.9 (2.6)	12.6 (2.0)	16.9 (20.1)	3.5 (5.0)
Indoors away from home	5.2 (5.0)	8.1 (2.9)	15.5 (30.5)	7.6 (9.3)
COPD Subjects (15 Monitoring Events; $N = 105$ Subject-Days)				
Indoors at home	88.0 (13.0)	5.9 (2.5)	8.7 (11.2)	79.3 (20.3)
Outdoors at home	1.3 (2.6)	13.2 (2.2)	18.1 (17.0)	2.4 (4.9)
In transit	3.3 (4.0)	11.4 (2.4)	16.0 (14.8)	6.0 (6.9)
At work	0.3 (1.9)	18.9 (1.9)	23.1 (17.0)	0.7 (4.9)
Outdoors away from home	1.4 (2.6)	12.5 (2.2)	16.8 (14.3)	2.7 (6.0)
Indoors away from home	5.7 (10.0)	10.3 (2.7)	15.7 (19.2)	9.0 (12.7)
Asthmatic Subjects (2 Monitoring Events; $N = 12$ Subject-Days)				
Indoors at home	66.9 (12.7)	8.7 (2.1)	11.5 (13.0)	49.7 (17.7)
Outdoors at home	1.1 (2.4)	18.4 (1.7)	21.1 (11.7)	2.4 (5.1)
In transit	2.9 (2.6)	17.5 (2.1)	22.4 (16.3)	5.0 (4.4)
At work	0.0 (0.0)	—	—	—
Outdoors away from home	5.9 (4.1)	17.1 (2.0)	20.0 (10.8)	8.5 (6.2)
Indoors away from home	23.2 (13.3)	16.7 (2.0)	20.7 (15.2)	34.4 (18.2)
All pDR Subjects (38 Monitoring Events; $N = 246$ Subject-Days)				
Indoors at home	86.8 (11.7)	6.5 (2.5)	10.0 (18.1)	79.2 (18.4)
Outdoors at home	1.0 (2.0)	12.7 (2.3)	18.1 (17.4)	1.8 (3.9)
In transit	3.5 (3.5)	10.5 (2.4)	16.2 (27.6)	5.6 (6.1)
At work	0.1 (1.2)	18.9 (1.9)	23.1 (17.0)	0.3 (3.2)
Outdoors away from home	1.8 (2.9)	13.0 (2.1)	17.1 (16.0)	3.1 (5.5)
Indoors away from home	6.8 (9.0)	10.1 (2.7)	16.3 (22.3)	9.9 (12.7)

arithmetic mean = 2.8 ± 7.3 $\mu g/m^3$). When the zero values of E_{ig} were included, the arithmetic mean was 2.2 ± 6.5 $\mu g/m^3$.

The hourly E_{pact} estimates were approximately normally distributed with an arithmetic mean of 3.3 ± 13.9 $\mu g/m^3$, and the estimates differed by microenvironment and activity (Table 4). The two microenvironments where PM monitoring was conducted, that is, indoors and outdoors at home, had the lowest E_{pact} levels. Note that the average E_{pact} indoors at home is influenced by approximately 900 hr of sleeping time. To estimate the personal cloud, we limited the analysis to hours when subjects were indoors or outdoors near home ($N = 2{,}276$) and during such hours the mean E_{pact} concentration was reduced to 1.9 ± 10.4 $\mu g/m^3$. This significantly ($p < 0.001$) lower average E_{pact} indicates that modeling error accounted for an average of approximately 1.4 $\mu g/m^3$ of the total E_{pact} for these subjects.

Activities also played an important role in determining E_{pact} concentrations. The results of the GEE model (Table 4) indicate that being at school (either in class or during recess), traveling by car, traveling by bus, burning candles, and sanding or carpentry were all associated with elevated E_{pact} concentrations ($p < 0.05$) relative to hours when no particle-generating activities were reported. Interestingly, most of the significant activities in the E_{pact}-activities model involved the subject spending time away from home.

The range of contributions of E_a, E_{ig}, and E_{pact} to total E for individual monitoring events were 21–80% (mean = 48%), 10–48% (mean = 21%), and 2–57% (mean = 31%), respectively. The utility of the hourly estimates of E_a, E_{ig}, and E_{pact} are shown in Figure 4 for one CHD subject. Exposure to ambient-originated PM generally dominated total personal exposure, whereas indoor and personal activity-generated PM only spiked occasionally. Figure 5 presents the averages of the hourly estimates of E_a, E_{ig}, and E_{pact} for each subject. The E_{pact} concentrations in Figure 5 have been separated into two parts: (1) E_{pact} during hours when the subject was indoors at home or outdoors near home (i.e., the personal cloud) and (2) E_{pact} resulting from modeling error (calculated as the average E_{pact} during all hours minus the average E_{pact} during hours in or near home). Note that for some subjects (particularly the two children) the E_{pact} concentrations were largely the result of time spent in unmonitored microenvironments.

Daily Estimates (Method 2). The GM personal exposure based on $HPEM_{2.5}$ for the subset of 55 concurrent

Table 3. Differences in total personal exposure measured with the pDR compared with the referent conditions and determined by the generalized estimating equations (GEE) models ($N = 38$ monitoring events).

Microenvironment or Activity	Model Estimates in $\mu g/m^3$	*p*-Value	Number of 30-min Periods with Microenvironment or Activity Reported
3a: Microenvironments			
Indoors at home (Referent condition)	10.79[a] (0.8)[b]	<0.001	9,857
At work[c]	7.88 (0.3)	<0.001	15
In transit (but not indoors away from home)[d]	1.62 (0.9)	0.07	376
In transit and indoors away from home[d]	1.58 (1.1)	0.14	111
Indoors away from home (but not in transit)[d]	1.40 (1.3)	0.28	777
Outdoors at home	0.96 (1.4)	0.50	126
Outdoors away from home	0.61 (0.9)	0.48	249
3b: Particle-generating activities			
None (referent condition)	10.77[a] (0.7)	<0.001	10,456
Burning a candle or incense	15.36 (10.2)	0.13	10
At school (in recess but not class/library)[d]	8.30 (0.5)	<0.001	16
At school (in class/library but not recess)[d]	5.76 (1.7)	<0.001	102
Cooking	5.46 (2.3)	<0.05	195
Carpentry or sanding	3.36 (2.8)	0.23	10
At school (both class/library and recess reported)[d]	2.66 (0.5)	<0.001	10
Traveling by bus	1.90 (1.5)	0.19	50
Traveling by car	0.63 (0.6)	0.27	622
Vacuuming, sweeping, or dusting	0.35 (0.4)	0.33	20

[a]Model intercept; [b]Standard error in parentheses; [c]Only one subject worked; [d]Multiple microenvironments or activities were reported during some 30-min intervals; the interaction terms take into account the most common covarying terms.

gravimetric and neph monitoring events (i.e., the monitoring events with method 2 exposure estimates) was 9.7 $\mu g/m^3$ (1.8; arithmetic mean = 11.6 ± 8.1 $\mu g/m^3$; $N = 403$ days). Personal exposure did not differ significantly by season, that is, 9.7 (1.8) $\mu g/m^3$ in the heating season ($N = 228$ days) and 9.7 (1.8) $\mu g/m^3$ in the nonheating season ($N = 175$ days; $p = 0.94$, 2-sample t test). The GM of the outdoor $HI_{2.5}$ concentration for this subset of subjects was higher during the heating season (11.0 $\mu g/m^3$ [1.8], $N = 228$ days) than during the nonheating season (7.7 $\mu g/m^3$ [1.6], $N = 175$ days, $p < 0.001$, 2-sample t test) because of wood burning in winter and less summertime photochemical particle enhancement in Seattle than in many other U.S. cities.[5] The seasonal difference in α (lower in the heating season and higher in the nonheating season; Figure 2a) counteracted the seasonal difference in the outdoor concentration, resulting in personal exposures that were similar across seasons.

The GM of E_a from method 2 was 5.8 $\mu g/m^3$ (1.7; arithmetic mean = 6.6 ± 3.6 $\mu g/m^3$). When zero values of E_{ig} were excluded, the GM was 1.4 $\mu g/m^3$ (3.7; arithmetic mean = 2.6 ± 2.9 $\mu g/m^3$). When zero values were included, the arithmetic mean decreased to 2.0 ± 2.8 $\mu g/m^3$. The daily E_{pact} estimates from method 2 were approximately normally distributed with a mean of 2.9 ± 7.2 $\mu g/m^3$. None of the exposure components showed seasonal differences. Eleven of the 55 monitoring events with method 2 estimates had an average $E_{pact} < 0$ $\mu g/m^3$. For monitoring events with an average $E_{pact} > 0$ $\mu g/m^3$ ($N = 44$), the range of contributions of E_a, E_{ig}, and E_{pact} to E for individual monitoring events were 13–90% (mean = 57%), 0–47% (mean = 17%), and 3–81% (mean = 26%), respectively.

Longitudinal Correlations of Personal Exposure with Central Monitoring Site

The median longitudinal r at a 1-hr averaging time was 0.30 (mean = 0.30 ± 0.23), and was statistically significant ($p < 0.05$) for 27 of 36 monitoring events. As expected based on the statistical process of averaging,[14] the median correlation coefficient improved with an increase in averaging time, reaching 0.62 (mean = 0.50 ± 0.46) for the 24-hr period. The 24-hr correlation was statistically significant ($p < 0.05$) for 14 of 36 monitoring events. The low correlation for hourly data implies an exposure misclassification issue in air pollution epidemiologic studies that attempt to use real-time central site measurements to evaluate health effects on a short time scale.

We examined the influence of α and nonambient sources, quantified with the estimated ratio of the variance of nonambient exposures to the variance of ambient concentrations (Ψ), on the longitudinal correlations. Forty-one monitoring events had at least four valid pairs of personal and central site gravimetric data and were not influenced by local outdoor sources. After removing 3 outliers, estimated values of Ψ ranged between 0.02 and 6.3 (mean = 1.3 ± 1.5, median = 0.66), with 47% of the monitoring events having $\Psi < 1$. First, we separated monitoring events into high and low groups for Ψ, that is, above or below the median value of 0.66. Next, using the 5- or 10-day average α estimates calculated with eq 2, we

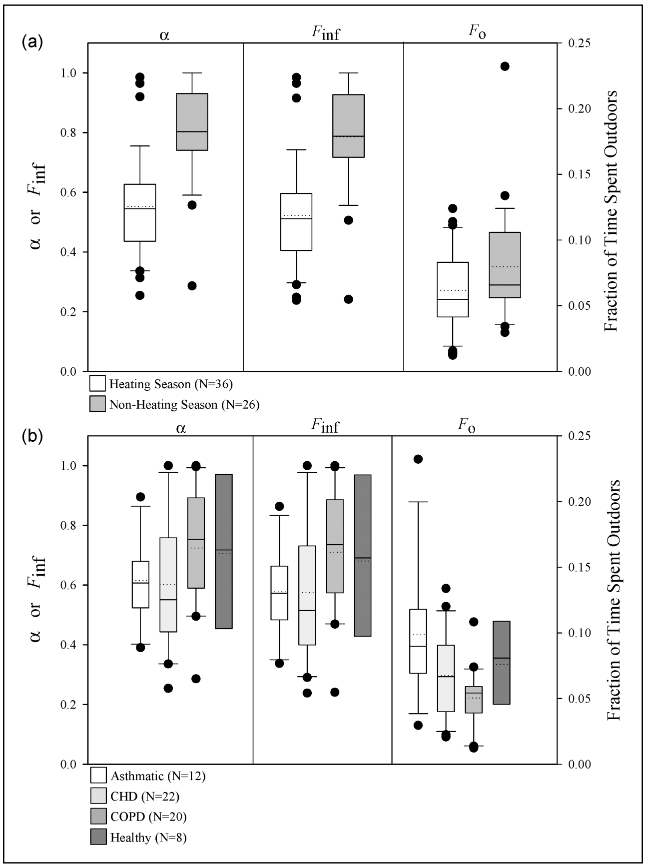

Figure 2. Ambient contribution fraction (α), particle infiltration efficiency (F_{inf}), and fraction of time spent outdoors (F_o) by (a) season and (b) health group.

stratified the monitoring events in both Ψ groups into low, middle, and high α groups based on the 33rd and 66th percentiles of each Ψ group (low Ψ group: low $\alpha < 0.51$, $0.51 \leq$ middle $\alpha \leq 0.69$, and high $\alpha > 0.69$; high Ψ group: low $\alpha < 0.52$, $0.52 \leq$ middle $\alpha \leq 0.63$, and high $\alpha > 0.63$). Figure 6 demonstrates the dependence of the longitudinal correlations between central site concentrations and personal exposures on α and nonambient exposure. Within both Ψ groups, there is a trend toward a higher median correlation between the central site and personal exposure as α increases. Within α groups, the median correlation is higher for monitoring events with a smaller relative variance in nonambient exposure, that is, lower Ψ values. Ψ seems to have more influence on the correlations than α; the median correlation for the lowest α group in the low Ψ group is approximately equal to the highest α group in the high Ψ group. Thus, PM exposure of subjects who live in well-ventilated homes would still be expected to have low longitudinal correlations with the central site measurements if nonambient sources dominate the microenvironments they encountered. We further examined personal and residential characteristics that affect the Ψ. Our multiple linear regression analysis results (Table 5) indicate that only the percent of hours with cooking reported, subject age (pediatric subjects had higher average Ψ), and the fraction of days with at least one open window were significantly ($p < 0.05$) associated with Ψ. Only 25% of the variability in Ψ was explained by our model, indicating that some important factors were unaccounted for.

Validation of Modeled Results

Figure 7 shows that the E_a and E_{ig} estimated from method 1 agreed with those calculated with method 3. Regressing the averaged hourly estimates on the daily estimates ($N = 92$ pairs from 14 monitoring events) we found the agreement for E_a to be better ($R^2 = 0.83$) than the agreement for E_{ig} ($R^2 = 0.76$ for all data; $R^2 = 0.66$ with highest E_{ig} value removed). The slightly poorer agreement for E_{ig} is probably caused by the fact that this quantity was estimated indirectly as the difference between the total indoor concentration and the infiltrated indoor concentration. The regression intercept for E_{ig} was significantly different from 0 ($p < 0.001$), but the slope was not different from 1 ($p = 0.49$). For E_a, the intercept was not different from 0 ($p = 0.52$), but the slope was significantly lower than 1 ($p < 0.001$). The correlations between the two methods provide further justification for our use of an event-averaged F_{inf} (as opposed to a day-specific F_{inf}) in the SSE model.

The median r between hourly measured (E) and modeled exposure ($\hat{E}$) was 0.59 (mean = 0.56 ± 0.28), and ranged between 0.12 and 0.96 ($N = 20$ monitoring events). To evaluate the bias from modeling exposure in environments where no measurements were taken (i.e., neither indoors at home nor outdoors near home), we limited our comparison to hours when the subjects were indoors or outdoors near home (N ranged between 55 and 196 hr for individual monitoring events). The median r

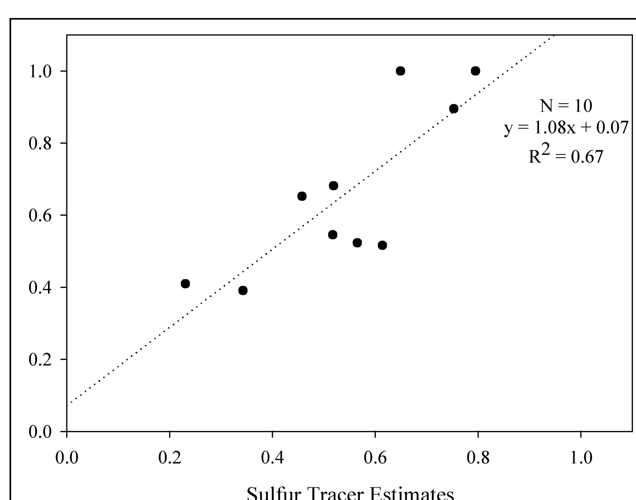

Figure 3. Comparison of ambient contribution fractions between source-specific exposure model and sulfur tracer methods.

Table 4. Differences in E_{pact} compared with the referent conditions as determined by the generalized estimating equations (GEE) models ($N = 20$ monitoring events).

Microenvironment or Activity	Model Estimates in μg/m³	*p*-Value	Number of Hours with Microenvironment or Activity Reported
4a: Microenvironments			
**Indoors at home* (referent condition)[e]	2.32[a] (0.4)[b]	<0.001	2,346
Indoors away from home (but not in transit)[d]	8.15 (1.7)	<0.001	256
At work[c]	7.44 (0.4)	<0.001	8
In transit and indoors away from home[d]	5.37 (2.6)	<0.05	18
In transit (but not indoors away from home)[d]	3.95 (1.7)	<0.05	82
Outdoors away from home	3.78 (1.7)	<0.05	55
Outdoors at home[e]	1.51 (1.7)	0.36	27
4b: Particle-generating activities			
None (Referent condition)	2.91[a] (0.4)	<0.001	2409
Burning a candle or incense	20.75 (8.5)	<0.05	6
At school (both class/library and recess reported)[d]	17.52 (2.4)	<0.001	5
At school (in recess but not class/library)[d]	16.62 (5.2)	<0.01	3
At school (in class/library but not recess)[d]	8.58 (0.7)	<0.001	33
Traveling by car	2.93 (1.4)	<0.05	236
Carpentry or sanding	2.22 (0.2)	<0.001	5
Traveling by bus	2.05 (0.5)	<0.001	9
Vacuuming, sweeping, or dusting	−0.02 (0.7)	0.98	3
Cooking	−0.96 (1.0)	0.31	70

[a]Model intercept; [b]Standard error in parentheses; [c]Only one subject worked; [d]Multiple microenvironments or activities were reported during some hours; the interaction terms take into account the most common covarying terms; [e]Stationary monitors were located in these microenvironments.

during such hours increased to 0.86 (mean = 0.72 ± 0.28, range = 0.18–0.96). The median r between E (measured with $HPEM_{2.5}$) and daily estimates of $\hat{E}$ (from method 2) was 0.75 (mean = 0.61 ± 0.39), with a range between −0.81 and 0.99 ($N = 55$ monitoring events).

DISCUSSION

Total Personal Exposures

The average daily pDR and $HPEM_{2.5}$ agreed reasonably well ($R^2 = 0.74$), although the pDR overestimated the $HPEM_{2.5}$ concentration by 30%. The positive pDR bias relative to the gravimetric method was on the low end of the range of literature values (27–50%) because we adjusted the pDR values based on colocated neph data when possible. Because the response of the pDR depends on the particle size, shape, density, and refractive index, the instrument should ideally be calibrated specifically for the aerosol being measured.[25] However, such aerosol-specific calibrations are not always feasible. Our previous studies have suggested that the bias and loss of precision introduced by using the pDR instead of gravimetric methods is an acceptable trade-off to be able to quantify short-term concentration peaks.[21–23]

Among our elderly groups, we found that an average of 88–90% of the total personal exposure was received indoors, which is in agreement with the 86–89% for elderly subjects in the Baltimore and Fresno exposure studies conducted by the U.S. Environmental Protection Agency (EPA).[20] These results indicate that elderly subjects with various health conditions across cities received a similar percent of their exposure indoors. The pediatric subjects spent more time away from home, and therefore they received a smaller percentage of their total exposure indoors at home than the elderly groups. Across all pDR subjects, the average personal exposure was lower indoors than in any of the other five microenvironments, especially in transit and at work. Similarly, in both Baltimore and Fresno, the lowest mean $PM_{2.5}$ concentration was also found when the subjects were indoors at home.[20] In our study, cooking, being in class, and being at recess were associated with increased levels of personal exposure. For healthy and more active adults in San Deigo, CA, barbecuing, yard work, construction work, and cooking were associated with elevated levels of personal PM exposure (measured with the pDR).[25]

Exposure to Ambient PM

We estimated a mean ambient contribution fraction, α, of 0.66 ± 0.21 based on time–location data and the infiltration

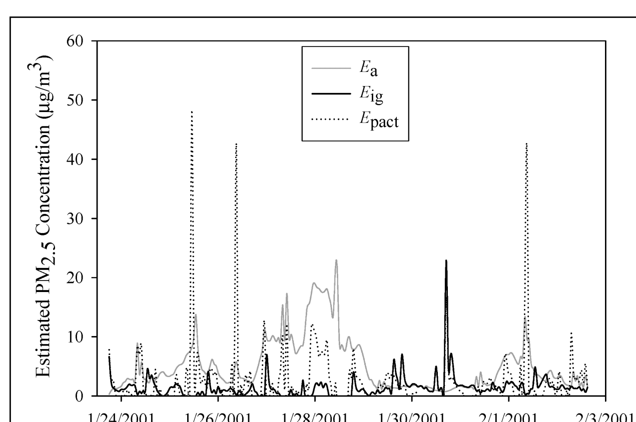

Figure 4. Example of the estimated hourly exposure to ambient-generated (E_a), indoor-generated (E_{ig}), and personal activity (E_{pact}) $PM_{2.5}$ for one subject.

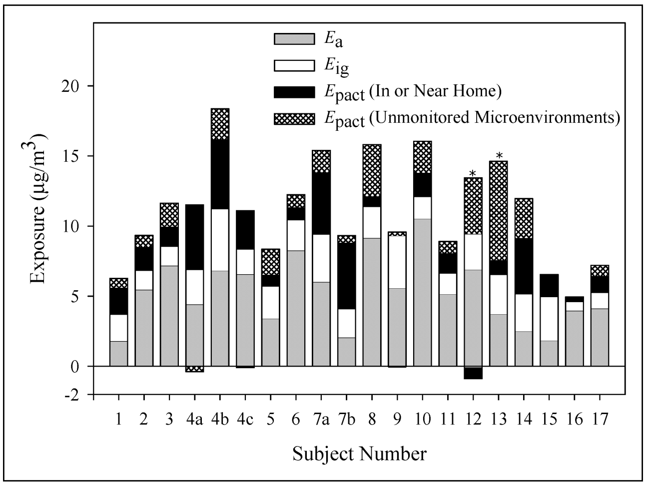

Figure 5. Average ambient, indoor-generated, and personal activity exposure and the influence of unmonitored microenvironments for each monitoring event. *Indicates children. Subjects 4 and 7 were monitored multiple times. "E_{pact} (near home)" was calculated as E (measured with pDR) − $\hat{E}(E_a + E_{ig})$ during hours when the subject was in or near home. "E_{pact} (unmonitored microenvironments)" was the difference between the average E_{pact} during all hours and the average "E_{pact} (near home)".

efficiency estimated with hourly neph measurements and a recursive model.[19] α represents the fraction of the ambient concentration to which individuals are exposed; thus, on average, the ambient-generated exposure for these subjects was about two thirds of the ambient concentration. The estimates of F_{inf} (mean = 0.63 ± 0.22) were similar to the estimates of α because of the small fraction of time spent outdoors (0.07 ± 0.04) by these subjects. α did not differ by health condition; however, α was lower during the heating season and higher during the nonheating season. The seasonal difference in α is largely caused by different residential ventilation characteristics or infiltration efficiency during the two seasons.[19] Our mean α is in the high end of the range of literature values. Sarnat et al. used sulfate as a tracer and found that summertime values for α among elderly subjects in Baltimore ranged from 0.39 in poorly ventilated indoor environments to 0.70 in well-ventilated indoor environments.[6] Landis et al. monitored 10 elderly subjects during the summer at a retirement facility in Baltimore and estimated an average α of 0.40 using sulfate.[28] Williams et al. estimated the average α for 37 adult subjects in North Carolina to be 0.47.[18] A study of adults with cardiorespiratory disease in Saint John, New Brunswick, Canada estimated an average α of 0.56 using sulfate.[15] Wilson et al.[11] used sulfur data collected in the fall in Riverside, CA,[16] and sulfate data collected in the summer from non-air-conditioned homes in Uniontown, PA,[29] and reported an average α for $PM_{2.5}$ of 0.70 and 0.88, respectively.

Our modeled α estimates agreed with α_s estimated with the sulfur tracer method (R^2 = 0.67), although this comparison is based on only 10 monitoring events. Four of the 62 estimates of α from eq 2 (2 of the 10 in Figure 3) were bounded at 1, indicating that α may be overestimated for some subjects. The personal–outdoor sulfur correlations for individual subjects (R^2 range = 0.70–0.99, median = 0.94) were in agreement with those reported in the PTEAM study (R^2 range = 0.8–0.9).[16]

E_a was estimated to contribute an average of 57% of the total exposure based on gravimetric samples (note that this includes only monitoring events with a positive average E_{pact}) and 48% based on light scattering data. These results are similar to the 47% estimated for nonsmoking adult subjects in North Carolina.[18] However, the fact that approximately 25% of the infiltrated indoor concentration estimates from both methods in our study were greater than the measured indoor concentrations provides further indication that we might be overestimating F_{inf} (and therefore α and E_a). This is consistent with the fact that we previously estimated an average α of 0.39 for the entire Seattle panel study with a fixed-effects model.[5]

The seasonal difference in α opposed the seasonal difference in outdoor $PM_{2.5}$ levels so that personal exposures did not differ between seasons in the present study. We previously reported higher exposure during the heating season among the entire Seattle panel study population.[5] Group homes were underrepresented in this subset of residences chosen for neph monitoring, and we previously reported a less pronounced seasonal difference in F_{inf} for group homes than for private homes or apartments.[19] Therefore, it is possible that the discrepancy in the seasonal effect on personal exposure between our current and former[5] work is caused by a less pronounced seasonal difference in F_{inf} (and therefore α) for the entire panel study than for the subset of residences chosen for neph monitoring.

Exposure to Personal Activity PM

Our average estimated E_{pact} concentration was 3.3 $\mu g/m^3$ using hourly light scattering data and 2.9 $\mu g/m^3$ using 24-hr gravimetric data. These E_{pact} estimates are on the low end of the range of values in the literature for young children or healthy adults,[30,31] but in agreement with the results of the entire Seattle panel study[5] and other studies of elderly subjects.[32] One advantage of our real-time data is that it allowed for estimation of E_{pact} concentrations in specific microenvironments and during specific times. To estimate the true personal cloud, we examined the E_{pact} concentrations during hours when the subjects were indoors at home or outdoors near home (where stationary nephs were located), and we found that the average E_{pact} concentrations during these hours were significantly lower than the average E_{pact} concentrations during all hours. This indicates that total E_{pact} is partly modeling errors resulting from subjects spending time away from microenvironments where stationary monitors were located. This finding helps explain why

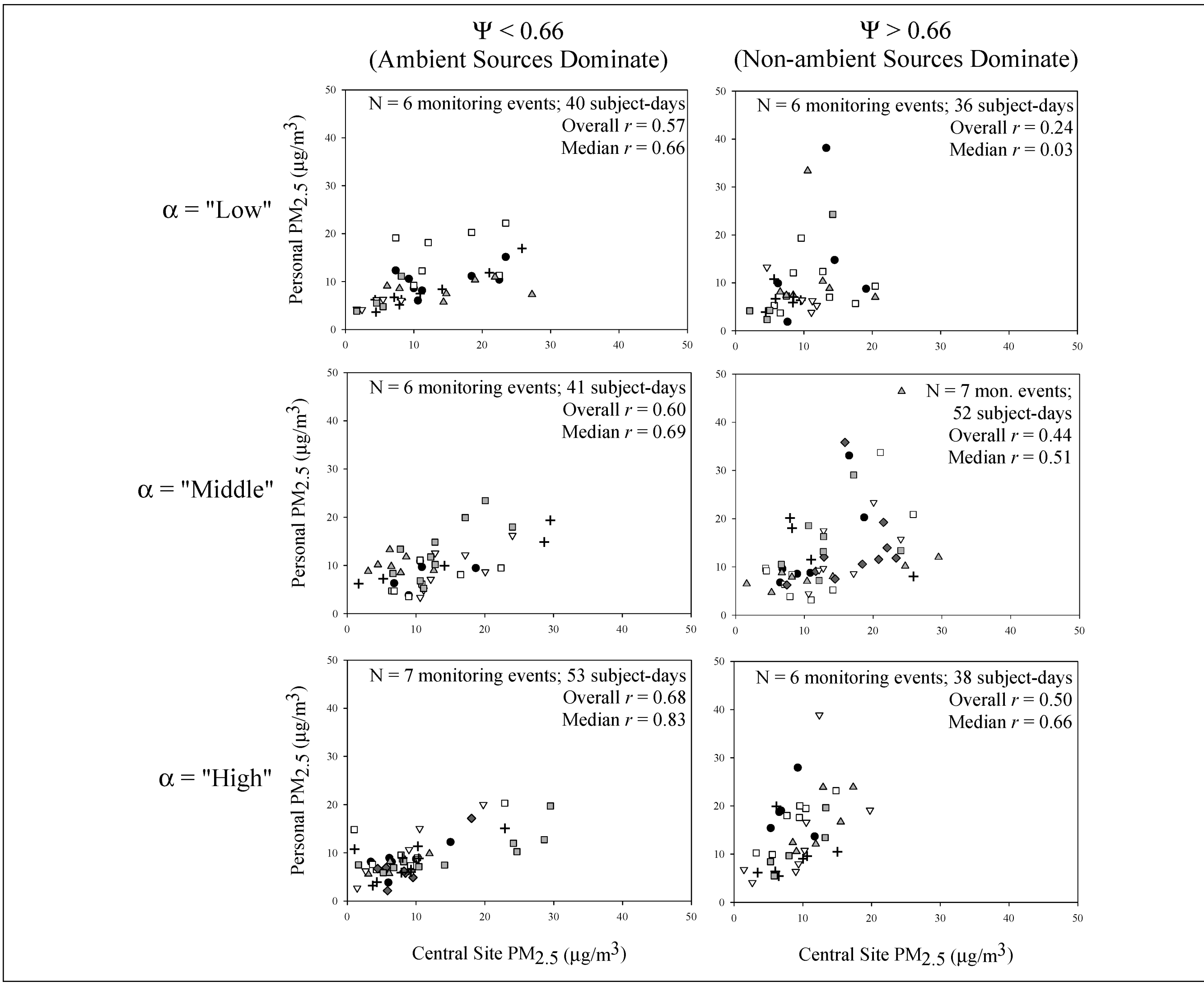

Figure 6. Relationship between central site concentration and personal exposure for 38 monitoring events stratified by α and $\Psi\left(\frac{\sigma^2_{E_{na}}}{\sigma^2_{C_a}}\right)$. (Note that the symbols in each graph represent a single monitoring event.)

children, who spent more time away from the home than elderly subjects, were found to have the highest average E_{pact} concentration of all the health groups in the larger Seattle panel study.[5] This was further confirmed by the statistically significant E_{pact} coefficients associated with being at school. It is important to note that our hourly estimates of E_{pact} involve the use of two instruments that respond differently to different aerosols. The pDR is more sensitive than the neph to particles larger than 1 μm, so the E_{pact} concentration during activities that produce particles above 1 μm is probably overestimated.[22]

Correlations between Personal Exposure and Central Site Concentrations

The median 24-hr longitudinal (Pearson's) correlation (0.62, 39% were significant at $p < 0.05$) between central site measurements and personal exposure was higher than the 0.43 for the entire Seattle panel study,[5] but within the broad range of correlations (0.0–0.86) reported in the literature.[4] Using hourly data, we found a median longitudinal r of 0.30 (75% were significant). A study of scripted activities performed by one technician in Baltimore reported hourly correlations between personal DustTrak and ambient TEOM $PM_{2.5}$ measurements ranging between 0.36 and 0.90.[33] The higher correlations were associated with outdoor environments or indoor environments with high air exchange rates. Similarly, a study in Baltimore demonstrated higher 24-hr ambient–personal correlations in well-ventilated indoor environments than in poorly ventilated ones; a finding the authors suggested is caused by the contribution of nonambient particle sources.[6] Rojas–Bracho et al. found that the influence of outdoor concentrations on personal exposures was affected by home ventilation conditions and indoor PM sources.[34] We presented a model that relates the longitudinal correlation to the relative variance of nonambient exposure and showed that an increase in the nonambient exposure contribution was associated with a decreased median longitudinal correlation. We also found a trend of increasing median correlations with increasing α. These results demonstrate that the correlations between outdoor concentrations and personal exposures are influenced by residential ventilation characteristics and particles generated indoors or from personal activities.

CONCLUSIONS

We used a source-specific exposure model to separate PM exposure into its ambient and nonambient components. This model is an effective alternative to the more common microenviromental modeling approach and allows for the separation of PM exposure into ambient and nonambient components for individual subjects. The exposure components estimated with neph data and recursive model F_{inf} estimates showed good agreement with the exposure components estimated with gravimetric and sulfur data. There was good agreement between modeled and measured total exposures (median $r = 0.59$ for hourly data), particularly during times when the subjects were in or near their residences (median $r = 0.86$). We found a considerable range in the daily contribution of ambient particles to the total personal exposure (21–80%). Although our 5- and 10-day average values of α provided useful exposure estimates, α varies over time and thus future estimates of ambient and nonambient exposure could be improved by estimating α on a shorter time scale. Real-time exposure estimates have the advantage over 24-hr estimates of allowing E_{pact} to be separated into two components—the true personal cloud and the model error caused by time spent away from monitored microenvironments. For subjects whose home indoor microenvironment has lower concentrations than other locations, as was found in elderly subjects in our study and in the U.S. Environmental Protection Agency (EPA)'s Fresno and Baltimore exposure panel studies, the total E_{pact} concentration will increase with the amount of time spent away from home if data collected indoors at home are used to represent all indoor microenvironments. We also found that total personal exposure may be poorly predicted by stationary outdoor monitors, particularly among persons living in tightly sealed homes, persons with a large nonambient exposure contribution (i.e., children and those who cook), and at a 1-hr averaging time.

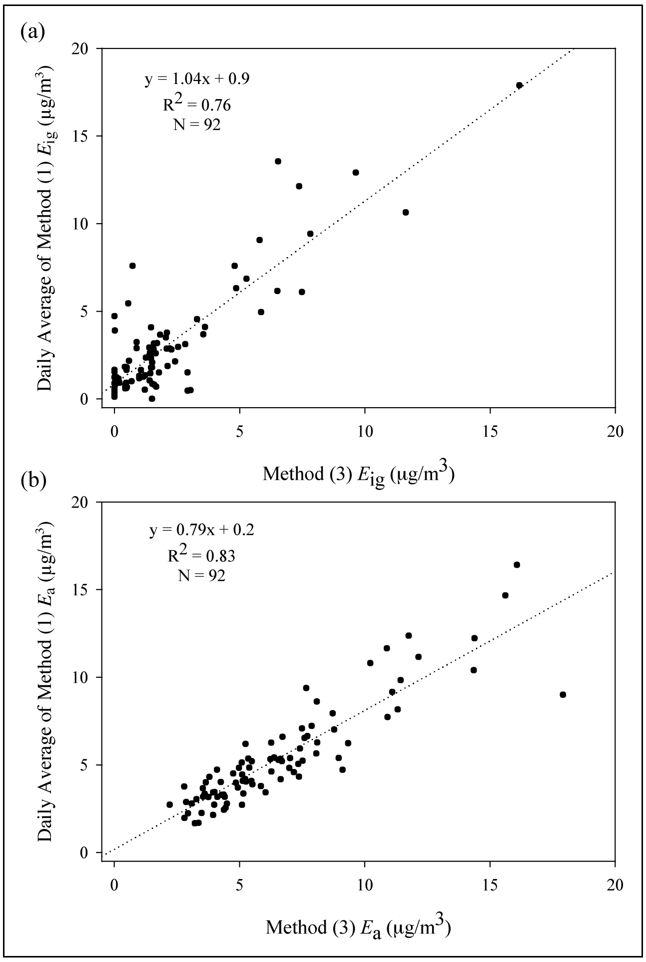

Figure 7. Comparison of method 1 and method 3 estimates of personal exposure to (a) indoor-generated $PM_{2.5}$, E_{ig}, and (b) ambient $PM_{2.5}$, E_a.

Table 5. Factors influencing the ratio Ψ as determined by multiple linear regression (model $R^2 = 0.25$).

Factor	Estimate (SE)	*p*-Value
Intercept	1.1 (0.6)	0.05
Subject characteristics		
% of days with cooking reported	0.2 (0.1)	<0.05
Use of an air cleaner[a]	0 (0.4)	0.94
Adult	Reference	
Child	1.7 (0.8)	<0.05
Residential characteristics		
Fraction of days with an open window	−1.1 (0.4)	<0.01
Private home	Reference	
Private apartment	0.6 (0.6)	0.3
Group home	0.1 (0.5)	0.77
Season		
Nonheating	Reference	
Heating	−0.4 (0.4)	0.33

[a]Air cleaner used for any number of days during the monitoring session.

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