Journal of the Air & Waste Management Association
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/uawm20

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Published online: 27 Dec 2011.

To cite this article: Jeremy A. Sarnat, Petros Koutrakis & Helen H. Suh (2000) Assessing the Relationship between Personal Particulate and Gaseous Exposures of Senior Citizens Living in Baltimore, MD, Journal of the Air & Waste Management Association, 50:7, 1184-1198, DOI: 10.1080/10473289.2000.10464165

To link to this article: http://dx.doi.org/10.1080/10473289.2000.10464165

TECHNICAL PAPER

ISSN 1047-3289 *J. Air & Waste Manage. Assoc.* **50**:1184-1198

Assessing the Relationship between Personal Particulate and Gaseous Exposures of Senior Citizens Living in Baltimore, MD

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ABSTRACT

We conducted a multi-pollutant exposure study in Baltimore, MD, in which 15 non-smoking older adult subjects (>64 years old) wore a multi-pollutant sampler for 12 days during the summer of 1998 and the winter of 1999. The sampler measured simultaneous 24-hr integrated personal exposures to $PM_{2.5}$, PM_{10}, SO_4^{2-}, O_3, NO_2, SO_2, and exhaust-related VOCs.

Results of this study showed that longitudinal associations between ambient $PM_{2.5}$ concentrations and corresponding personal exposures tended to be high in the summer (median Spearman's $r = 0.74$) and low in the winter (median Spearman's $r = 0.25$). Indoor ventilation was an important determinant of personal $PM_{2.5}$ exposures and resulting personal–ambient associations. Associations between personal $PM_{2.5}$ exposures and corresponding ambient concentrations were strongest for well-ventilated indoor environments and decreased with ventilation. This decrease was attributed to the increasing influence of indoor $PM_{2.5}$ sources. Evidence for this was provided by SO_4^{2-} measurements, which can be thought of as a tracer for ambient $PM_{2.5}$. For SO_4^{2-}, personal–ambient associations were strong even in poorly ventilated indoor environments, suggesting that personal exposures to $PM_{2.5}$ of ambient origin are strongly associated with corresponding ambient concentrations. The results also indicated that the contribution of indoor $PM_{2.5}$ sources to personal $PM_{2.5}$ exposures was lowest when individuals spent the majority of their time in well-ventilated indoor environments.

Results also indicate that the potential for confounding by $PM_{2.5}$ co-pollutants is limited, despite significant correlations among ambient pollutant concentrations. In contrast to ambient concentrations, $PM_{2.5}$ exposures were not significantly correlated with personal exposures to $PM_{2.5\text{-}10}$, $PM_{2.5}$ of non-ambient origin, O_3, NO_2, and SO_2. Since a confounder must be associated with the exposure of interest, these results provide evidence that the effects observed in the $PM_{2.5}$ epidemiologic studies are unlikely to be due to confounding by the $PM_{2.5}$ co-pollutants measured in this study.

IMPLICATIONS

Particulate epidemiologic studies typically use stationary, ambient concentrations as a surrogate for personal exposure. Questions have been raised concerning the accuracy of ambient $PM_{2.5}$ measures as a surrogate of exposure due to variability in personal–ambient associations and confounding by $PM_{2.5}$ co-pollutants. This paper examines the personal particulate and gaseous exposures for a cohort of older adults and investigates their relationship to corresponding ambient concentrations and to one another. Findings from these analyses will help interpret results from epidemiological studies.

INTRODUCTION

Associations between ambient concentrations of PM and a variety of adverse health outcomes have been well documented in numerous epidemiologic studies.[1-3] Results from recent studies indicate that $PM_{2.5}$ is the particulate size fraction most strongly associated with these observed effects.[4] Despite consistent findings from these studies, interpretation of the results has been questioned, in part due to the use of ambient concentrations as surrogates of personal exposure, and also due to concerns about confounding by $PM_{2.5}$ co-pollutants, such as $PM_{2.5\text{-}10}$, O_3, NO_2, and SO_2.

Weak correlations between ambient PM concentrations and personal exposures reported in various cross-sectional exposure studies have been offered by some as evidence that ambient concentrations are poor surrogates of personal PM exposures and that findings reported in epidemiologic studies of PM are inaccurate.[5] More recent observations examining the longitudinal associations between personal and ambient PM have muted some of this concern. These studies have found relatively strong associations between personal $PM_{2.5}$, and to a lesser degree PM_{10}, exposures and ambient concentrations over time.[6,7] Although the strength of these associations varied substantially by individual, results indicated that for certain individuals ambient PM concentrations are an appropriate surrogate for personal $PM_{2.5}$ exposures.

In one of the more recent studies by Rojas-Bracho et al.,[8] this inter-personal variation was attributed to variability in indoor $PM_{2.5}$ concentrations, as the association between personal $PM_{2.5}$ exposures and outdoor concentrations was found to be strongly correlated with corresponding associations between outdoor and indoor $PM_{2.5}$ concentrations. This finding suggests that indoor sources and indoor ventilation characteristics are important determinants of personal $PM_{2.5}$ exposures and their relationship to outdoor levels. (Since the participants spent similar amounts of time indoors, this was unable to account for the observed variability in the personal–ambient relationship.) For $PM_{2.5-10}$, the relationship between ambient concentrations and corresponding personal exposures was shown to be insignificant. Personal exposures to $PM_{2.5-10}$ and $PM_{2.5}$ were uncorrelated as well. These insignificant relationships suggested that $PM_{2.5-10}$ is unlikely to confound observed associations between ambient $PM_{2.5}$ and adverse health effects.

The Baltimore multi-pollutant exposure study builds on the work of these previous longitudinal studies by 1) further examining the influence of indoor ventilation status upon associations between personal exposures and ambient concentrations for various particulate and gaseous measures and 2) investigating the potential for confounding by several $PM_{2.5}$ co-pollutants. In order to measure gaseous and particulate pollutants simultaneously, a multi-pollutant personal sampler was developed for this study. Using these simultaneous measurements, we examined and compared associations between personal exposures and ambient concentrations for the various pollutants and determined the relative importance of indoor sources and ventilation characteristics to these associations.

This paper reviews the design of the Baltimore study and provides a characterization of the personal particulate and gaseous exposures for this sample cohort. Relationships between the personal exposures and ambient concentrations for the various pollutants are presented. In addition, factors influencing these relationships are discussed.

METHODS

Study Design

During the summer of 1998 and the winter of 1999, 24-hr integrated personal exposures to $PM_{2.5}$, PM_{10}, SO_4^{2-}, O_3, NO_2, SO_2, and select VOCs were measured for 20 older adults living in the Baltimore metropolitan area. Ten subjects participated in both the summer and winter sampling seasons, while the remaining 10 subjects participated in either the summer or winter season. Subjects were monitored for 12 days during each of the monitoring seasons. Four to six subjects were measured concurrently during each 12-day monitoring period. All summer samples were collected from June 29 to August 7, 1998. All winter samples were collected from February 2 to March 13, 1999. All subjects completed the prescribed 12-day sampling period with the exception of one individual who dropped out of the study after the first day and was subsequently replaced with an additional subject.

Subjects were recruited primarily from senior and community centers located throughout the Baltimore area. Subject selection was non-random. All subjects were nonsmokers, retired, physically healthy, and lived in non-smoking private residences (i.e., either single-family houses or apartments). All residences, except one (SA4), were equipped with central air conditioning; however, not all of these residences used air conditioning throughout the summer. The average age of the subjects was 75 (±6.8 years). The subjects were from a range of socio-economic backgrounds and geographic locations throughout Baltimore.

In addition to wearing the personal sampler, every subject completed a daily time-activity diary on each monitoring day. Subjects were given a choice of completing two types of time-activity diaries during the study. Twenty-five of the thirty subjects used a closed-form, time-based diary in which subjects recorded their activities and location in closed, time-interval spaces that were provided on the diaries. During the summer, the closed-form diary was divided into 30-min recording intervals, while in the winter, the diary consisted of 15-min intervals. The diary included several columns to allow the subject to indicate whether a specific pollutant-generating activity was being performed (i.e., dusting, vacuuming, exposure to environmental tobacco smoke [ETS], near a busy road). The remaining five subjects used an open-form, activity-based diary. Each page of the diary corresponded to a specific activity with space available on the page to indicate location and any special conditions likely to affect exposure. No formal cross-validation of the two diary formats was conducted. However, differences between the two formats were not likely to affect study results, since analyses based on activity diaries was limited.

Subjects recorded the ventilation status of every visited indoor location (e.g., windows open, air conditioning use). Ventilation status was recorded only during the summer sampling session, as it was assumed that windows were closed during the winter in Baltimore. For the summer, personal exposures were classified by the fraction of time the windows were open while a subject was in an indoor environment (F_v). This metric can be thought of as a surrogate for air-exchange rate, as several studies have shown air-exchange rates to be associated with both open-window status and air conditioning use.[8,9] General information concerning the subject's residence was also collected at the beginning of the sampling period. At the beginning of each 12-day monitoring period, field staff

recorded household characteristics that could potentially have influenced personal exposures. Among these characteristics were cooking fuel type, carpeting, garage type, air-cleaning devices, pets, and number of individuals residing in the residence.

Field staff visited the subjects daily each morning of monitoring between 7:00 a.m. and 11:00 a.m. to change sampling equipment, replace pump batteries, adjust flows, and review the completed time-activity diaries from the previous 24 hr.

Multi-Pollutant Sampler. All samples were collected using a specially designed multi-pollutant sampler, which was attached to a single, BGI model 400 sampling pump. The entire sampler weighed approximately 5 lb. Sampling inlets were placed on the shoulder strap of a backpack to correspond to the breathing zone of each subject. The sampling pump and battery pack were carried in a back or shoulder pack. Subjects were permitted to remove the pack during activities when the sampler could be damaged or during prolonged periods of inactivity (i.e., sleeping, watching TV). During periods when the sampler was removed from the subject's body, subjects were instructed to keep the sampling inlets as close as possible to their breathing zone.

The multi-pollutant sampler is a modification of an earlier, PM-only version described by Rojas-Bracho et al.[7] The performance of this modified multi-pollutant sampler has been discussed in Chang et al.[10] The sampler consisted of active samplers to collect $PM_{2.5}$, PM_{10}, and VOCs and passive samplers to collect O_3, NO_2, and SO_2 (Figure 1). To allow simultaneous $PM_{2.5}$, PM_{10}, and VOC sampling using a single pump, the sample air flow was split three ways using flow restrictors. Two Personal Environmental Monitors (PEMs), one for $PM_{2.5}$ and the other for PM_{10}, were placed, nozzle-down, into 10-cm-long PFA Teflon-coated aluminum elutriators.[11] Tests were conducted to examine the effect of the elutriators on particle collection. Nine pairs of $PM_{2.5}$ PEMs and 10 pairs of PM_{10} PEMs were collocated on subjects. For each pair, one PEM was placed in an elutriator, nozzle-down, while the other PEM was also placed downward without an elutriator. Each PEM sampled at a flow rate of 3.2 L/min ± 10%. Each subject wore the two PEMs for approximately 16 hr. A paired two-sample *t*-test was performed to examine whether the elutriator affected the measurement of $PM_{2.5}$ or PM_{10} exposures. Results from this *t*-test showed no difference between the elutriator/non-elutriator measurements for either cut size.

Particles were collected by the PEMs on 37-mm Teflon filters (37-mm Teflo, Gelman Sciences) at a flow rate of 3.2 and 2.0 L/min for $PM_{2.5}$ and PM_{10}, respectively. VOCs were collected at a flow rate of 20 cm^3/min using sorbent tubes filled with CarbopackB (Supelco; Bellefonte, PA). SO_4^{2-} concentrations were determined by extracting the $PM_{2.5}$ filters and analyzing the aqueous extract by ion chromatography.

O_3, NO_2, and SO_2 concentrations were measured using passive badge samplers. Each of the passive badge samplers contained a single cellulose filter, coated with either nitrite to collect O_3[12] or triethanolamine to collect NO_2 and SO_2.[13] To ensure a constant face velocity across the filter, each of the passive samplers were placed in the side of the elutriators, with the face of the passive sampler exposed to the sampling air stream. Filters were extracted after sampling and analyzed using ion chromatography.

In addition to personal sampling, field staff operated a pair of Harvard Impactors (HIs) at a centrally located monitoring site to measure 24-hr (8:00 a.m.–8:00 a.m.) integrated ambient $PM_{2.5}$ and PM_{10} data. All of the subjects lived within a 20-km radius of this site. O_3, NO_2, SO_2, and VOC data were obtained from stationary ambient monitoring (SAM) sites operated by the Maryland Department of the Environment and located throughout Baltimore. Ambient concentrations of O_3, NO_2, and SO_2 were measured using a UV photometric analyzer, a chemiluminiscence monitor, and a pulsed fluorescent monitor, respectively.

The Teflon filters used to collect $PM_{2.5}$ and PM_{10} were weighed in a temperature- and humidity-controlled weighing room (temperature = 18–24 °C; relative humidity = 40 ± 5%). Filters were left to equilibrate 24 hr before the initial weighing and 48 hr prior to post-sampling weighing. Each filter was weighed in duplicate both before and after sampling using a using a Mettler Model MT5 microbalance (Mettler Toledo International Inc.; Greifensee, Switzerland). The average of the two weights was used as the filter weight. When the two filter weights differed by more than 5 μg, the filter was weighed a third time, with the final value being the average of the two closest weights.

Quality Assurance. Standard quality assurance/quality control procedures were followed for this study.[14] Collected data have been assessed for their bias, precision, and completeness.

Precision and bias of the multi-pollutant sampler methods were calculated by collocating replicate, fully configured sampling backpacks at SAM sites equipped with reference sampling methods. The samplers were operated for 24 hr ± 10%. For a given pollutant, precision was estimated as the root mean squared difference between the collocated personal samplers, divided by the square root of 2. Bias for a given method was determined using the mean relative difference between the multi-pollutant sampler concentration and the corresponding mean reference method concentration. Method limit of detections (LODs) were estimated as 3 times the standard deviation of the field blanks.

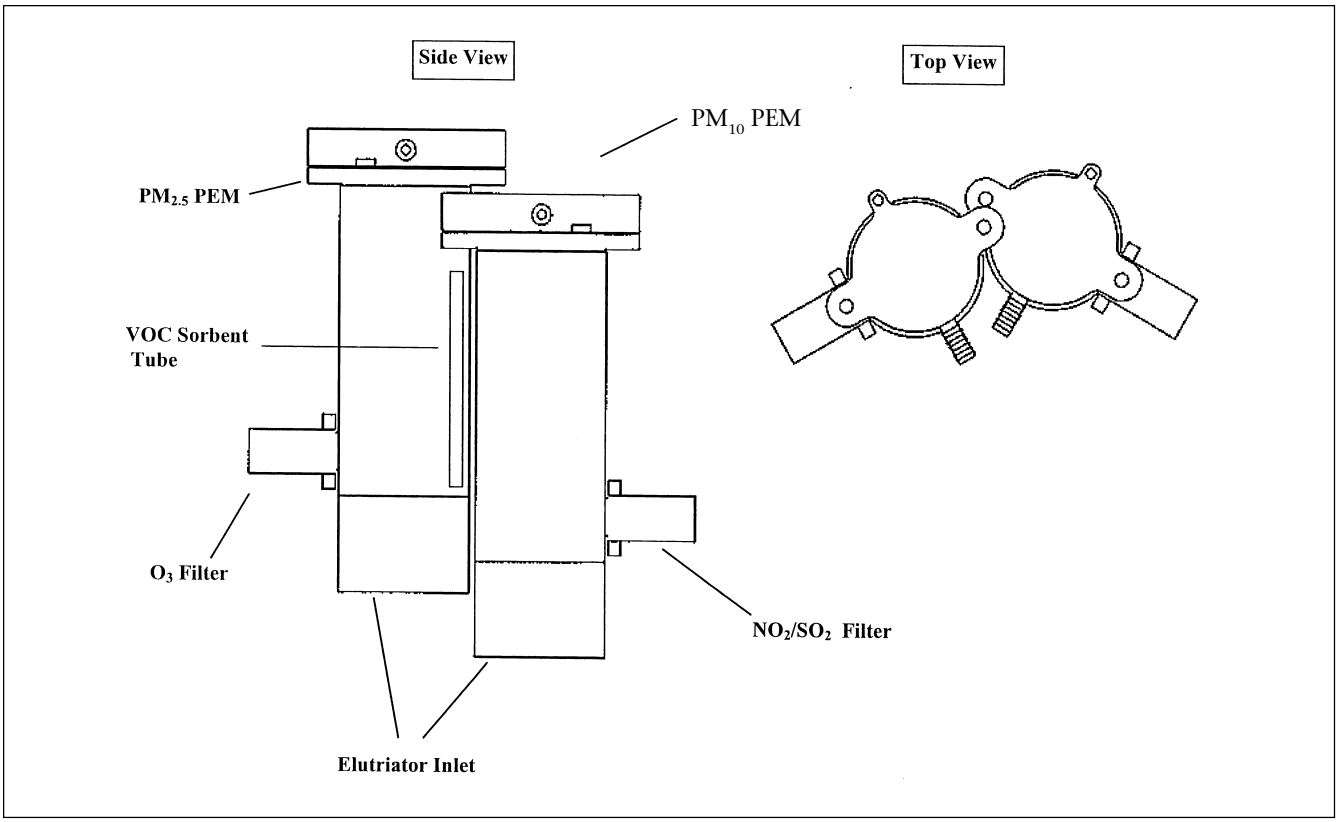

Figure 1. Diagram of multi-pollutant sampler.

Completeness was calculated as the number of samples collected divided by the target number of samples. In total, 30 12-day sampling sessions were conducted during the study. At least 9 days of valid samples were collected for each subject. Precision, bias, LOD, and completeness results for each method are listed in Table 1.

Filter Quality Assurance. The total number of field blanks equaled 19% of all personal PM filters and 14% of all outdoor PM filters. Mean blank filter weights, which were significantly different from 0 ($\alpha = 0.05$), were used to correct the particulate mass concentrations. PM masses were corrected by subtracting the mean field blank weights from the sample weights. The field blank values for the PEMs were 2.2 μg (4.0 μg) and 5.3 μg (4.5 μg) for the summer and winter seasons, respectively. Field blank value for the HIs were –0.3 μg (3.4 μg) and 1.2 μg (3.5 μg) for the summer and winter seasons, respectively. Teflon filters were also corrected for barometric pressure during weighing. During the winter sampling period, all particle filters were stored in refrigerated environments post-sampling to reduce potential volatilization from the filters.

All O_3 and NO_2/SO_2 badge samplers were refrigerated pre- and post-sampling and were shipped pre- and post-sampling in refrigerated coolers. Since blank values differed by batch, blank corrections for the gaseous pollutants were computed on a per-batch basis. For O_3, blank correction values ranged from 0.2 to 1.0 parts per billion (ppb) and 0.1 to 1.0 ppb during the summer and winter, respectively. For NO_2, blank correction values ranged from 0.2 to 0.4 ppb and 0.2 to 0.3 ppb during the summer and winter, respectively. For SO_2 badge samplers, blank correction values ranged from 0.11 to 0.18 ppb.

Data Analysis. Units for PM concentrations and exposures are reported in $\mu g/m^3$. Coarse particle concentrations, or $PM_{2.5-10}$, were calculated as the difference between PM_{10} and $PM_{2.5}$. Units for gaseous concentrations and exposures are reported in ppb. Analysis of VOC data was not included in this paper. Negative values for the gaseous pollutants and $PM_{2.5-10}$, as well as values less than their respective LODs, were included in the data analyses.

Shapiro-Wilks tests indicated that the data were not normally distributed. As a result, the median, mean, standard deviation, and maximum concentrations were used to describe the distribution of the data. Data were characterized using descriptive statistics, graphical displays, Spearman's correlation coefficients, linear regression, and mixed models. Spearman's correlation coefficient, linear regression, and mixed models were computed using the SAS system (SAS Institute Inc.; Cary, NC). Statistical significance is reported at the 0.05 level unless otherwise specified.

Analyses of the ventilation effect on associations between personal exposures and ambient concentrations

were conducted using mixed-effects models in which personal exposures were modeled as the dependent variable, ambient concentrations and ventilation status were modeled as fixed variables, and subjects were modeled as random variables to account for between-subject variation. Autocorrelation between pollutant concentrations over time was modeled using an autoregressive covariance structure. Since in the mixed model there is no equivalent single measure of goodness-of-fit, crude R^2 values were generated using simple linear regression techniques and are presented to give a rough indication of the scatter of the data around the estimated regression lines.

Exclusion of Data Points. Data points were voided due to sampling problems (e.g., pump or battery failures, tube disconnection) or laboratory analysis irregularities. Time-activity data indicated that one subject, in particular, (listed as SA4 in the summer and WA3 in the winter) was heavily exposed to ETS, since this individual spent a large fraction of time in bingo parlors where smoking was common. Summary statistics have been presented both including and excluding samples for which substantial ETS exposure was reported. Analysis of associations between personal exposures and ambient concentrations, however, did not include this subject, since collected samples did not typify exposures for a non-smoker or someone living in a residence with non-smokers.[6,15] Similarly, analysis of the effect of ventilation status also did not include data from subject SA4, due to ETS exposure, nor did it include data from subjects SB5 and SC5 due to inadequate reporting of ventilation status in their activity diaries. (The analysis of the effect of ventilation status was conducted during the summer only. As such, specific exclusion of data from WA3, a winter subject, was unnecessary.) In addition, analyses of ventilation effects were conducted both with and without an additional, single $PM_{2.5}$ observation from subject SB2, which showed an extremely high personal $PM_{2.5}$ exposure. This extreme value exerted substantial influence on the overall model results, yet no source for this elevated exposure was identified in the time-activity diaries.

RESULTS

Summary statistics for the measured ambient PM concentrations and personal exposures, stratified by size, season, and subject, are presented in Tables 2 and 3. Summary statistics for the gaseous pollutants are presented in Tables 4 and 5.

During both seasons, personal PM exposures were comparable, yet significantly lower ($p < 0.0001$) than their respective ambient concentrations for all particle cut-sizes. The sole exception to this finding was wintertime coarse PM. In all, ambient concentrations of $PM_{2.5}$, PM_{10}, $PM_{2.5-10}$, and SO_4^{2-} exceeded corresponding personal exposures 228 of 333 (68%), 206 of 314 (66%), 172 of 310 (55%), and 307 of 314 (98%) of sampled person-days, respectively. Median ambient concentrations and median personal exposures for all PM cut-sizes, except for $PM_{2.5-10}$, were significantly higher during the summer than the winter.

During both sampling seasons, personal exposures to O_3, NO_2, and SO_2 were extremely low. Seventy percent of the measured personal O_3, NO_2, and SO_2 values were below their respective LOD, even when ambient concentrations were well above their LOD.

Ambient concentrations of O_3 and NO_2 varied seasonally. As expected, O_3 levels tended to be higher in the summer while NO_2 concentrations were higher during the winter. These ambient seasonal trends were reflected, to a lesser degree, in personal exposure measurements as well.

Table 1. Method LOD, precision, bias, and completeness.

	Summer					Winter					Units
	LOD	Precision	Bias MRD (SD)[a]	Bias Reference Mean[b]	Completeness	LOD	Precision	Bias MRD (SD)[a]	Bias Reference Mean[b]	Completeness	
$PM_{2.5}$	2.6	0.9[c]	2.6 (2.6)	32.5	168/180 (93.3%)	4.0	1.2	1.7 (2.6)	20.1	165/180 (91.7%)	$\mu g/m^3$
PM_{10}	4.2	1.3[c]	0.1 (3.8)	43.2	169/180 (94.0%)	6.4	2.1	-1.0 (5.6)	30.0	168/180 (93.3%)	$\mu g/m^3$
$PM_{2.5-10}$	4.9[d]	1.5	-2.5 (3.3)	8.6	167/180 (92.8%)	7.5[c]	2.7	-2.6 (4.9)	9.1	165/180 (91.7%)	$\mu g/m^3$
SO_4^{2-}	2.6[e]	0.7[e]	N/A	N/A	168/180 (93.3%)	2.6[e]	0.7[e]	N/A	N/A	164/180 (91.1%)	$\mu g/m^3$
O_3	6.6	3.4	-0.2 (5.3)	38.4	145/180 (81.0%)	5.5	3.7	-1.0 (5.4)	21.2	165/180 (91.7%)	ppb
NO_2	5.7	2.3	0.0 (2.8)	16.0	167/180 (92.8%)	11.7	8.9	6.4 (20.5)	23.5	163/180 (90.6%)	ppb
SO_2	N/A	N/A	N/A	N/A	N/A	6.5	2.3	-3.5 (1.9)	8.1	160/180 (88.9%)	ppb

Notes: [a] Corresponds to the mean relative difference (MRD) between the multi-pollutant sampler concentration to the corresponding reference method concentration; [b] Corresponds to the mean concentration of the reference method; [c] Excludes one outlier. When outlier is included, the precision for $PM_{2.5}$ is 2.9 $\mu g/m^3$ and 2.2 $\mu g/m^3$ for PM_{10}; [d] Square root of the sum of the squares of the two single sampler LODs; [e] Estimated LOD and precision from the Harvard-EPA Annular Denuder System.

Relationship Between Personal Exposures and Ambient Concentrations

In general, the association between ambient concentrations and personal exposures to PM varied by particle cut-size and by season (Table 6, Figure 2). The strength of the association increased with decreasing particle size.

The strongest associations between personal exposures and ambient concentrations were observed for SO_4^{2-}, which can be considered a tracer of ambient particles since it is an important fine particle constituent ($d_a < 1$ μm) with few indoor sources.[16,17] When data were analyzed cross-sectionally and were treated as independent, personal SO_4^{2-} exposures and ambient concentrations were strongly associated, with ambient concentrations explaining 64% of the variation in personal SO_4^{2-} exposures ($p < 0.0001$; $N = 127$). The personal–ambient associations were even stronger when data were analyzed by individual. Twenty-one of the 28 subjects (14 subjects per season) had significant personal–ambient associations. Correlation coefficients for these subjects were stronger in the summer than the winter. This seasonal difference may be attributed to differences in indoor ventilation conditions, which were generally characterized by open windows in the summer and closed windows in the winter.[8,18]

Although weaker than that for SO_4^{2-}, the association between personal $PM_{2.5}$ exposures and ambient concentrations was also significant. This was not unexpected given that SO_4^{2-} [expressed as $(NH_4)_2SO_4$] was a major component of $PM_{2.5}$ in Baltimore, comprising 40% of the total ambient $PM_{2.5}$ mass in the summer and 26% of the total ambient $PM_{2.5}$ mass in the winter. When the data were analyzed cross-sectionally and treated as independent, ambient concentrations explained 45% of the variability in personal $PM_{2.5}$ exposures ($p < 0.0001$; $N = 133$ [$R^2 = 0.28$ including extreme value for SB2]). Again, correlations between personal exposures and ambient concentrations improved when data were analyzed by subject, as significant correlation coefficients were found for 10 of the 14 summertime subjects, resulting in a median correlation coefficient of 0.76 (Table 6). Despite the observed high median correlation coefficient, correlation coefficients for the measured individuals ranged widely ($-0.21 < r < 0.95$). This finding is consistent with previous studies.[6,19-21]

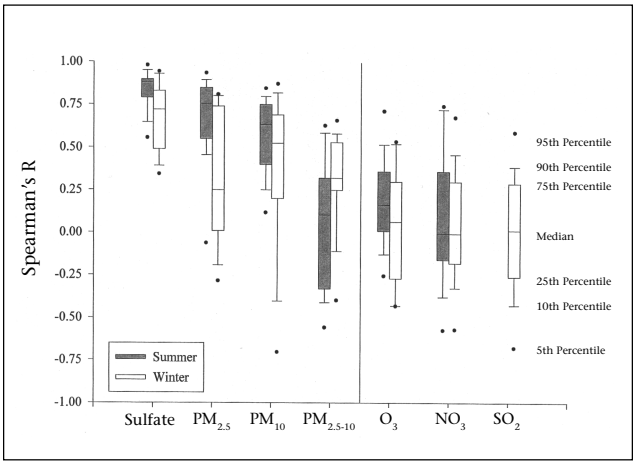

Figure 2. Distribution of personal–ambient correlation coefficients.

As was the case for SO_4^{2-}, personal–ambient correlations for $PM_{2.5}$ were lower in the winter as compared with the summer, with significant positive, correlation coefficients found for only five of the 14 winter subjects (median $r = 0.25$). This seasonal difference was independent of individual, as the seasonal variation in the personal–ambient associations was evident even for the 10 subjects participating in both sampling seasons. For these 10 subjects, the median correlation coefficient was 0.70 in the summer and 0.25 in the winter. This seasonal difference was more pronounced than that observed for SO_4^{2-}, suggesting that both indoor ventilation differences and indoor $PM_{2.5}$ emissions were responsible for the observed seasonal variation in the personal–ambient association for $PM_{2.5}$.

Consistent with previous studies,[7] personal $PM_{2.5-10}$ exposures and ambient concentrations were generally not significantly associated, as only three of the 28 subjects (14 subjects per season) had significant and positive correlation coefficients for $PM_{2.5-10}$. Insignificant personal–ambient associations for $PM_{2.5-10}$ probably reflect the contribution of indoor $PM_{2.5-10}$ sources, which include cooking, cleaning, and resuspension, to personal $PM_{2.5-10}$ exposures and also the reduced effective penetration efficiency of coarse particles indoors.[7,8,22,23]

Similarly, personal–ambient associations were weak for all of the gaseous pollutants, a result which was not unexpected since measured personal gaseous exposures were frequently below their respective limits of detection (Tables 4 and 5). In addition, spatial variability in ambient gaseous concentrations may have contributed to these weak associations, as spatial variability in outdoor gaseous concentrations may be relatively high due to local sources, such as automobiles for NO_2, or to local sinks, such as automobiles for O_3. Virtually none of the correlation coefficients for the individual-specific pairwise comparisons were significant for O_3, NO_2, or SO_2 (Table 6, Figure 2).

Ventilation Status and $PM_{2.5}$ Correlations

The influence of indoor ventilation patterns on the association between personal and ambient $PM_{2.5}$ was examined by using mixed models on data stratified into one of three indoor ventilation categories. Indoor ventilation conditions were categorized as either well ($F_v > 0.72$), moderately ($0.04 \le F_v \le 0.72$), or poorly ($F_v < 0.04$) ventilated, based on the distribution (mean ± standard deviation) of the fraction of time indoor environments were characterized by open windows (F_v). In the summer, F_v values varied

Table 2. Descriptive statistics for ambient concentrations and personal exposures: PM, summer 1998 ($\mu g/m^3$).

Subject	$PM_{2.5}$					PM_{10}					$PM_{2.5-10}$					SO_4^{2-}				
	N[a]	Med	Mean	SD[b]	Max	N[a]	Med	Mean	SD[b]	Max	N[a]	Med	Mean	SD[b]	Max	N[a]	Med	Mean	SD[b]	Max
SA1	12	15.3	16.7	5.8	26.2	12	22.8	22.5	6.3	33.7	12	6.1	5.8	2.9	11.5	12	2.7	3.0	1.6	6.5
SA2	10	27.4	29.4	8.7	43.8	11	34.4	38.7	12.6	64.2	10	6.4	6.7	3.0	12.4	10	3.7	5.0	3.5	13.8
SA4	11	78.4	80.7	41.9	134.8	11	101.0	85.4	39.1	130.9	11	2.8	4.7	8.8	22.5	11	4.8	6.4	4.5	14.8
SA5	10	24.6	28.9	12.4	49.6	10	28.0	30.9	11.0	49.6	10	1.4	2.0	2.4	6.9	10	4.8	7.5	4.9	17.9
A Ambient	12	21.7	25.3	12.8	46.6	8	28.0	33.7	15.6	55.6	8	7.1	7.0	1.1	9.0	9	7.0	9.4	7.1	21.2
SB1	11	21.0	23.5	11.0	52.3	11	36.1	36.2	10.4	59.8	11	12.8	12.6	3.8	20.0	11	5.2	5.0	3.2	13.4
SB2	10	27.4	32.8	17.6	77.9	10	27.3	33.9	18.0	78.1	10	-0.1	1.7	3.0	8.4	10	4.9	4.7	1.8	7.5
SB3	12	20.5	21.9	7.7	39.3	12	31.1	32.6	9.0	49.0	12	10.1	10.7	4.1	18.0	12	6.6	6.4	3.3	14.2
SB4	12	18.6	21.7	6.5	35.1	12	24.5	28.6	9.0	42.4	12	6.6	6.9	4.4	14.7	12	6.3	6.3	2.9	11.5
SB5	12	21.2	22.7	8.4	42.1	12	23.4	24.9	8.8	43.1	12	2.0	2.2	2.0	5.6	12	5.4	6.1	3.4	15.1
SB6	9	23.7	23.6	7.5	40.0	9	29.4	28.9	7.4	40.3	9	4.6	5.3	2.8	9.0	9	6.8	7.5	2.9	14.9
B Ambient	12	24.8	26.4	9.5	46.9	12	32.7	35.5	9.9	54.4	12	8.1	9.1	2.6	14.5	12	10.3	11.7	7.8	29.6
SC1	12	19.1	20.9	10.3	36.0	12	30.0	30.3	9.6	46.8	12	9.6	9.3	3.5	14.5	12	6.0	6.8	4.6	16.5
SC2	12	12.5	12.9	4.2	23.0	12	17.3	18.9	4.9	27.8	12	5.7	6.0	2.7	12.6	12	3.4	3.6	1.8	6.4
SC3	12	18.1	19.6	8.1	32.0	12	18.2	30.9	7.7	43.7	12	11.4	11.2	3.3	14.9	12	4.5	5.5	3.8	11.3
SC4	11	20.8	20.1	6.8	29.2	11	28.4	30.3	14.0	68.7	11	4.8	10.2	14.0	49.3	11	7.1	6.7	3.8	14.0
SC5	12	20.2	18.9	6.4	28.8	12	30.5	33.1	16.1	66.4	12	8.9	14.3	14.1	49.3	12	5.7	5.8	3.1	9.3
C Ambient	13	20.9	23.9	12.7	45.6	13	28.3	32.7	14.2	55.0	13	9.2	8.8	2.1	12.1	13	6.8	10.2	8.8	27.3
Summer Personal[c]	37	23.1	26.7	13.7	63.6	37	32.1	33.9	11.7	65.0	37	6.7	7.2	4.0	24.8	37	5.4	5.6	3.1	13.8
Summer Personal[c,d]	37	20.6	22.2	7.5	40.6	37	27.7	29.7	7.5	46.9	37	6.4	7.3	3.8	24.8	37	5.4	5.6	3.0	13.8
Summer Personal[c,e]	37	23.1	26.7	13.7	63.6	34	29.7	32.8	11.5	65.0	34	6.8	7.4	4.0	24.8	35	5.5	5.8	3.1	13.8
Summer Personal[c,d,e]	37	20.6	22.2	7.5	40.6	34	27.9	29.9	7.6	46.9	34	6.7	7.4	3.9	24.8	35	5.5	5.7	3.1	13.8
Summer Ambient	37	23.1	25.2	11.5	46.9	34	32.2	34.0	12.8	55.6	34	8.1	8.4	2.3	14.5	35	8.0	10.5	7.8	29.6

Notes: [a]N refers to sample size; [b]SD refers to arithmetic standard deviation; [c]Summer personal values were computed by calculating the mean exposures of all subjects sampled concurrently; [d]Excludes personal exposures with extreme ETS exposures; [e]Matched with ambient sampling days (i.e., personal exposures from days when no ambient monitoring was available were removed).

Table 3. Descriptive statistics for ambient concentrations and personal exposures: PM, winter 1999 ($\mu g/m^3$).

$PM_{2.5}$ Subject	N[a]	Med	Mean	SD[b]	PM_{10} Max	N[a]	Med	Mean	SD[b]	$PM_{2.5-10}$ Max	N[a]	Med	Mean	SD[b]	SO_4^{2-} Max	N[a]	Med	Mean	SD[b]	Max
WA1	12	9.5	10.4	3.4	17.7	12	16.1	19.2	11.4	54.6	12	6.4	8.8	10.7	42.0	12	1.5	1.5	0.6	2.5
WA2	11	15.6	17.0	2.5	20.9	11	35.3	34.3	5.1	41.1	11	17.1	17.3	4.9	26.0	11	2.6	2.3	0.8	3.6
WA3	10	46.5	53.4	38.7	113.1	10	58.2	66.2	40.3	124.3	10	12.6	12.8	9.4	36.0	10	3.5	3.5	1.1	6.2
WA4	9	11.3	15.3	2.9	19.8	9	18.3	20.1	4.1	27.7	9	4.8	4.9	2.3	7.8	9	2.6	2.2	0.7	2.8
WA5	11	15.7	16.6	5.5	30.4	11	25.4	26.5	7.7	40.5	11	10.1	9.9	4.2	17.0	11	2.7	2.6	0.9	4.0
A Ambient	12	22.3	22.4	5.8	32.6	11	29.3	28.4	6.3	36.3	11	5.3	6.9	3.1	11.3	11	4.4	4.1	1.0	5.7
WB1	11	11.5	12.5	4.5	20.3	11	15.8	16.4	6.3	27.1	11	4.1	3.9	2.6	7.9	10	2.1	2.1	0.9	3.8
WB2	12	14.4	21.3	14.8	57.6	12	21.2	35.1	34.4	134.4	12	5.5	13.8	32.7	117.3	12	2.4	2.4	1.2	4.2
WB3	11	43.0	46.5	24.4	88.6	11	66.8	71.3	33.8	124.6	11	27.1	24.8	11.6	43.5	11	3.1	3.1	1.4	5.2
WB4	12	23.4	26.4	13.0	52.4	12	32.5	37.4	18.3	81.9	12	9.4	11.0	6.9	29.6	12	2.3	2.3	1.1	4.1
B Ambient	12	24.8	25.1	12.0	49.0	12	35.2	33.3	17.1	73.2	12	5.6	8.6	7.0	24.2	12	5.0	4.8	2.2	8.3
WC1	12	5.2	5.9	2.9	11.9	12	12.5	12.0	5.9	23.8	12	6.2	6.1	3.5	12.2	12	0.8	0.1	0.4	1.6
WC2	10	4.1	5.5	3.8	15.1	11	6.5	7.3	3.7	17.4	10	2.4	2.0	1.1	3.4	10	1.1	1.2	0.6	2.3
WC3	10	5.7	6.6	3.8	14.6	11	9.3	9.8	5.2	20.3	10	3.1	3.5	2.4	6.4	10	1.0	1.2	0.7	2.2
WC4	10	6.9	7.1	2.7	11.3	11	9.7	10.3	3.0	14.4	10	3.4	3.6	2.5	6.6	10	1.9	1.5	0.6	2.2
WC5	12	5.6	5.8	2.4	10.7	12	6.6	7.9	3.8	14.9	12	0.9	2.1	2.4	7.3	12	1.7	1.6	0.6	2.4
WC6	12	9.7	11.2	5.0	25.1	12	15.4	18.3	8.4	37.0	12	6.0	7.1	4.6	18.6	12	1.5	1.3	0.5	2.0
C Ambient	12	11.9	14.1	6.8	26.3	11	18.3	18.0	9.4	34.0	11	3.8	4.8	3.8	9.4	11	3.4	3.0	0.9	3.7
Winter Personal[c]	36	15.4	18.5	11.2	44.0	36	24.8	28.0	16.5	69.5	36	8.2	9.6	7.9	47.6	36	1.9	2.1	1.0	4.1
Winter Personal[c,d]	36	14.5	16.6	10.2	44.0	36	23.3	26.7	17.6	90.8	36	8.3	10.2	10.9	68.2	36	1.9	2.0	1.0	4.4
Winter Personal[c,e]	36	15.4	18.5	11.2	44.0	34	24.8	28.5	16.7	69.5	34	8.2	9.7	8.1	47.6	34	1.9	2.1	1.0	4.1
Winter Personal[c,d,e]	36	14.5	16.6	10.2	44.0	34	23.3	27.2	17.9	90.8	34	8.3	10.3	11.2	68.2	34	1.9	2.1	1.0	4.4
Winter Ambient	36	20.7	5.6	49.0	9.7	34	28.2	7.5	73.2	13.4	34	5.2	-1.3	24.2	5.1	34	2.7	1.0	8.3	1.7

Notes: [a]N refers to sample size; [b]SD refers to arithmetic standard deviation; [c]Winter personal values were computed by calculating the mean exposures of all subjects sampled concurrently; [d]Excludes personal exposures with extreme ETS exposures; [e]Matched with ambient sampling days (i.e., personal exposures from days when no ambient monitoring was available were removed).

Table 4. Descriptive statistics for ambient concentrations and personal exposures: gaseous pollutants, summer 1998 (ppb).

Subject	O_3 N[a]	Med	Mean	SD[b]	Max	NO_2 N[a]	Med	Mean	SD[b]	Max
SA1	12	0.1	0.1	1.9	3.1	12	–1.2	–1.0	1.7	3.4
SA2	10	3.7	3.0	3.0	6.5	12	3.4	2.6	4.3	8.3
SA4	11	0.7	1.0	2.1	5.9	11	14.8	15.5	5.7	24.2
SA5	10	6.3	6.2	3.7	11.5	10	8.4	8.3	2.5	12.7
A Ambient	12	34.5	37.5	9.4	53.6	12	18.7	20.7	5.2	30.6
SB1	10	1.2	1.1	1.8	4.3	11	7.4	8.6	4.4	16.2
SB2	9	0.2	0.6	0.1	2.7	10	7.1	8.2	5.4	20.5
SB3	11	2.7	3.2	1.8	6.9	12	5.6	5.8	3.0	11.0
SB4	11	1.1	1.0	1.4	3.4	12	4.9	5.4	5.1	14.4
SB5	10	0.9	0.9	1.5	4.0	11	3.4	9.8	17.3	59.3
SB6	8	3.1	4.0	4.0	11.7	9	3.9	10.4	13.4	39.6
B Ambient	12	39.7	38.4	7.1		12	22.7	21.5	4.5	30.8
SC1	9	2.4	4.7	6.7	21.1	11	17.1	16.7	4.9	26.8
SC2	10	8.5	8.5	5.6	17.1	12	8.4	13.5	16.9	65.8
SC3	9	3.1	5.3	4.6	10.4	11	10.0	10.5	4.5	18.1
SC4	8	9.6	9.6	2.2	14.1	11	7.3	8.5	5.1	17.2
SC5	7	9.7	9.7	3.5	16.9	12	11.3	11.9	6.1	22.7
C Ambient	13	36.3	36.9	7.6	48.7	12	20.4	21.8	5.2	30.5
Summer Personal[c]	33	2.7	3.5	3.0	9.9	37	8.0	8.7	5.4	28.6
Summer Personal[c,d]	33	3.3	3.6	3.0	9.9	37	7.4	7.9	5.8	28.6
Summer Ambient[e]	33	35.1	37.3	8.3	53.6	37	20.4	21.4	4.9	30.8
Summer Ambient	37	36.3	37.6	7.9	53.6	37	20.4	21.4	4.9	30.8

Notes: [a]N refers to sample size; [b]SD refers to arithmetic standard deviation; [c]Summer personal values were computed by calculating the mean exposures of all subjects sampled concurrently; [d]Excludes personal exposures with extreme ETS exposures; [e]Matched with personal sampling days (i.e., ambient concentrations from days when no personal sampling was available were removed).

Table 5. Descriptive statistics for ambient concentrations and personal exposures: gaseous pollutants, winter 1999 (ppb).

Subject	O_3 N[a]	Med	Mean	SD[b]	Max	NO_2 N[a]	Med	Mean	SD[b]	Max	SO_2 N[a]	Med	Mean	SD[b]	Max
WA1	12	0.5	0.1	1.3	1.6	12	3.9	4.2	1.9	74.0	11	0.2	0.2	1.4	2.3
WA2	11	–0.3	0.0	1.7	1.0	11	16.3	17.0	24.5	3.9	10	1.1	1.2	5.0	1.8
WA3	10	0.9	0.9	4.1	1.9	9	25.9	25.5	41.4	8.1	8	–0.5	–0.7	2.3	1.5
WA4	9	0.7	0.4	3.0	1.6	9	15.4	15.2	20.0	4.5	9	1.3	0.7	2.1	1.2
WA5	11	1.0	0.6	1.8	1.0	11	19.2	19.2	29.5	5.2	11	0.4	0.0	2.5	1.8
A Ambient	12	11.2	10.6	6.3	21.2	12	24.6	24.1	4.2	30.7	12	5.9	6.6	2.6	10.4
WB1	11	0.6	0.5	2.7	1.2	10	5.7	8.6	24.4	8.5	10	0.5	0.3	2.3	1.6
WB2	12	0.5	0.5	2.5	0.9	11	11.3	18.2	54.1	17.7	11	0.4	0.3	3.5	1.4
WB3	11	0.3	0.9	3.6	1.5	10	11.3	18.0	52.6	18.6	10	0.1	–0.5	1.7	1.7
WB4	12	0.8	0.8	3.7	1.5	12	6.5	13.6	42.1	15.7	12	–0.7	–0.5	1.3	1.4
B Ambient	12	13.0	15.5	9.6	34.5	12	21.7	22.2	9.9	39.2	12	9.4	10.2	3.7	17.6
WC1	12	–2.8	–2.1	6.7	4.3	12	15.7	20.8	94.4	24.8	12	0.3	–0.1	1.8	1.5
WC2	12	2.3	–0.8	3.8	3.8	11	9.2	11.8	25.9	9.0	11	–0.3	–0.3	2.6	1.4
WC3	10	0.0	–0.2	8.5	5.0	11	12.3	13.6	32.3	9.7	11	–0.2	–0.7	1.7	1.5
WC4	11	–2.8	–2.5	0.3	2.1	11	15.1	18.9	36.7	9.2	11	–0.3	–0.8	0.7	1.3
WC5	12	-0.8	–1.3	2.8	3.1	12	25.9	30.1	54.5	11.1	12	0.8	0.5	2.7	1.6
WC6	11	0.4	1.5	10.6	5.7	11	10.0	13.1	40.6	10.3	11	–0.3	–0.7	1.4	1.4
C Ambient	12	26.5	27.3	7.0	38.5	12	23.9	21.8	5.6	28.8	12	8.0	9.8	6.3	18.8
Winter Personal[c]	36	0.3	0	1.8	2.8	36	14.3	16.3	7.9	42.7	36	–0.1	0.0	0.7	1.5
Winter Personal[c,d]		0.3	0	1.8	2.8	36	13.1	15.8	8.0	42.7	36	0.0	0.0	0.7	1.5
Winter Ambient	36	16.0	17.8	10.3	38.5	36	24.0	22.7	6.9	39.2	36	8.3	8.9	4.6	18.8

Notes: [a]N refers to sample size; [b]SD refers to arithmetic standard deviation; [c]Winter personal values were computed by calculating the mean exposures of all subjects sampled concurrently; [d]Excludes personal exposures with extreme ETS exposures.

Table 6. Correlation between personal and ambient pollutant level by subject and season.[a]

SUMMER							WINTER						
	SO_4^{2-}	$PM_{2.5}$	PM_{10}	$PM_{2.5-10}$	O_3	NO_2		SO_4^{2-}	$PM_{2.5}$	PM_{10}	$PM_{2.5-10}$	O_3	NO_2	SO_2
SA1	0.95[b]	0.55	0.5	0.32	0.36	0.72[b]	**WA1**	0.4	0.22	0.2	0.25	-0.27	0.74[b]	0.05
SA2	0.79[b]	0.85[b]	0.86[b]	0.21	0.12	0.27	**WA2**	0.54	-0.38	-0.36	0.26	-0.28	-0.01	-0.75[b]
SA5	0.9[b]	0.89[b]	0.79[b]	-0.39	0.49	-0.07	**WA4**	0.33	-0.18	-0.79[b]	-0.48	0.52	-0.18	-0.02
SB1	0.66[b]	0.65[b]	0.63[b]	0	0.21	-0.31	**WA5**	0.49	0.22	0.3	0.68[b]	0.3	0.13	0.65[b]
SB2	0.82[b]	-0.21	0.08	0.26	-0.03	-0.13	**WB1**	0.93[b]	0.8[b]	0.69[b]	0.36	0.35	0.3	-0.21
SB3	0.86[b]	0.82[b]	0.75[b]	-0.11	-0.11	0.06	**WB2**	0.88[b]	0.62	0.54	0.53	0.29	0	-0.39
SB4	0.89[b]	0.73[b]	0.59[b]	0.58[b]	0.47	0.14	**WB3**	0.95[b]	0.55	0.51	0.48	-0.43	-0.64[b]	0.36
SB5	0.89[b]	0.73[b]	0.71[b]	0.29	0.77[b]	-0.35	**WB4**	0.81[b]	-0.12	0.1	-0.07	0.18	-0.04	-0.26
SB6	0.53	0.53	0.4	0.37	0.12	-0.63	**WC1**	0.75[b]	0.74[b]	0.59	0.32	0.3	0.3	-0.22
SC1	0.99[b]	0.95[b]	0.79[b]	-0.6[b]	0.33	0.75[b]	**WC2**	0.73[b]	0.79[b]	0.89[b]	0.1	-0.43	-0.25	-0.28
SC2	0.92[b]	0.78[b]	0.29	-0.34	0.01	-0.08	**WC3**	0.71[b]	0.28	0.79[b]	0.55	-0.11	-0.29	0.14
SC3	0.9[b]	0.85[b]	0.71	-0.33	0.27	0.4	**WC4**	0.47	0.19	0.81[b]	0.57	0.54	0.17	0.35
SC4	0.87[b]	0.78[b]	0.27	-0.12	0.05	-0.16	**WC5**	0.83[b]	0.81[b]	0.69[b]	0.32	-0.05	-0.01	0.06
SC5	0.71[b]	0.55	0.64[b]	0.64[b]	-0.29	0.36	**WC6**	0.55	0.01	0.35	0.25	-0.11	0.43	0.29
Median	0.88	0.76	0.64	0.11	0.17	-0.01	**Median**	0.72	0.25	0.53	0.32	0.07	-0.01	0.02
Mean	0.83	0.68	0.57	0.06	0.20	0.07	**Mean**	0.67	0.33	0.38	0.29	0.06	0.05	-0.02
Stdev	0.12	0.29	0.23	0.38	0.28	0.40	**Stdev**	0.20	0.40	0.47	0.30	0.34	0.34	0.37
95% CI	0.07	0.15	0.12	0.20	0.14	0.21	**95% CI**	0.11	0.21	0.25	0.16	0.18	0.18	0.19

Notes: [a]Correlations represent Spearman's *r* values; [b]Significant at the $\alpha = 0.05$ level.

widely by individual for most subjects spanning ventilation categories (Figure 3). In the winter, F_v values for all subjects were assumed to fall into the poorly ventilated category. Since ventilation conditions during the summer were quantified (and not assumed) using activity data, analyses for summer and winter data were performed separately.

When summertime personal $PM_{2.5}$ exposures were measured for individuals spending time in well-ventilated indoor environments (e.g., $F_v > 0.72$), ambient concentrations were excellent predictors of personal $PM_{2.5}$ exposures (slope = 0.83, $p < 0.0001$, crude $R^2 = 0.80$) (Figure 4). The association between personal $PM_{2.5}$ exposures and corresponding ambient concentrations was weaker for the moderately ventilated category (slope = 0.59, $p < 0.0001$, crude $R^2 = 0.57$), and still weaker for the poorly ventilated category (slope = 0.46, $p < 0.0001$, crude $R^2 = 0.25$ [slope = 0.24, $p < 0.02$, crude $R^2 = 0.05$ including extreme value from SB2]). The weaker personal–ambient associations for the poorly ventilated category is reflected by the increased variability in the ratio of personal to ambient $PM_{2.5}$ for the poorly as compared to the well- and moderately ventilated groups (Figure 5). Slopes for the personal on ambient regressions (slope = 0.83, well; slope = 0.59, moderate; slope = 0.46, poor) and median personal/ambient ratios also decreased with ventilation. This may be due to lower effective penetration efficiencies for poorly ventilated indoor environments. In the winter, personal–ambient associations for $PM_{2.5}$ (both crude R^2 and slope) were similar to those for the summertime poorly ventilated category (slope = 0.47, $p < 0.0001$, crude $R^2 = 0.19$).

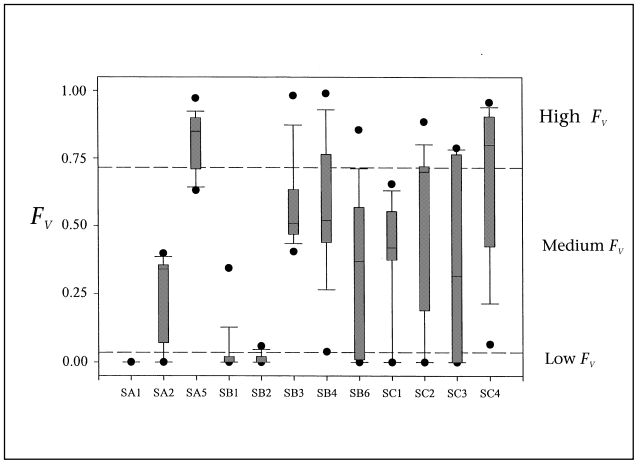

Figure 3. Range of F_v values by subject.

Ventilation status also affected the association between personal exposures and ambient concentrations for SO_4^{2-}; however, the effect was not as pronounced as that for $PM_{2.5}$. In the summer, ambient SO_4^{2-} concentrations were strongly associated with personal exposures for all three ventilation categories (crude $R^2 = 0.88$, 0.73, and 0.72 for the well, moderately, and poorly ventilated indoor environments, respectively) (Figure 6). As was the case with $PM_{2.5}$, the slopes of personal ambient level decreased with ventilation status (0.70, 0.40, and 0.39 for well, moderately, and poorly ventilated categories, respectively), which again may be due to the fact that the effective penetration efficiency for SO_4^{2-} is lower for these environments. In winter, even though ambient SO_4^{2-} concentrations were relatively low, ambient concentrations remained relatively strong predictors of personal SO_4^{2-} concentrations. Wintertime personal–ambient $PM_{2.5}$ associations (slope = 0.50, $p < 0.0001$, crude $R^2 = 0.71$)

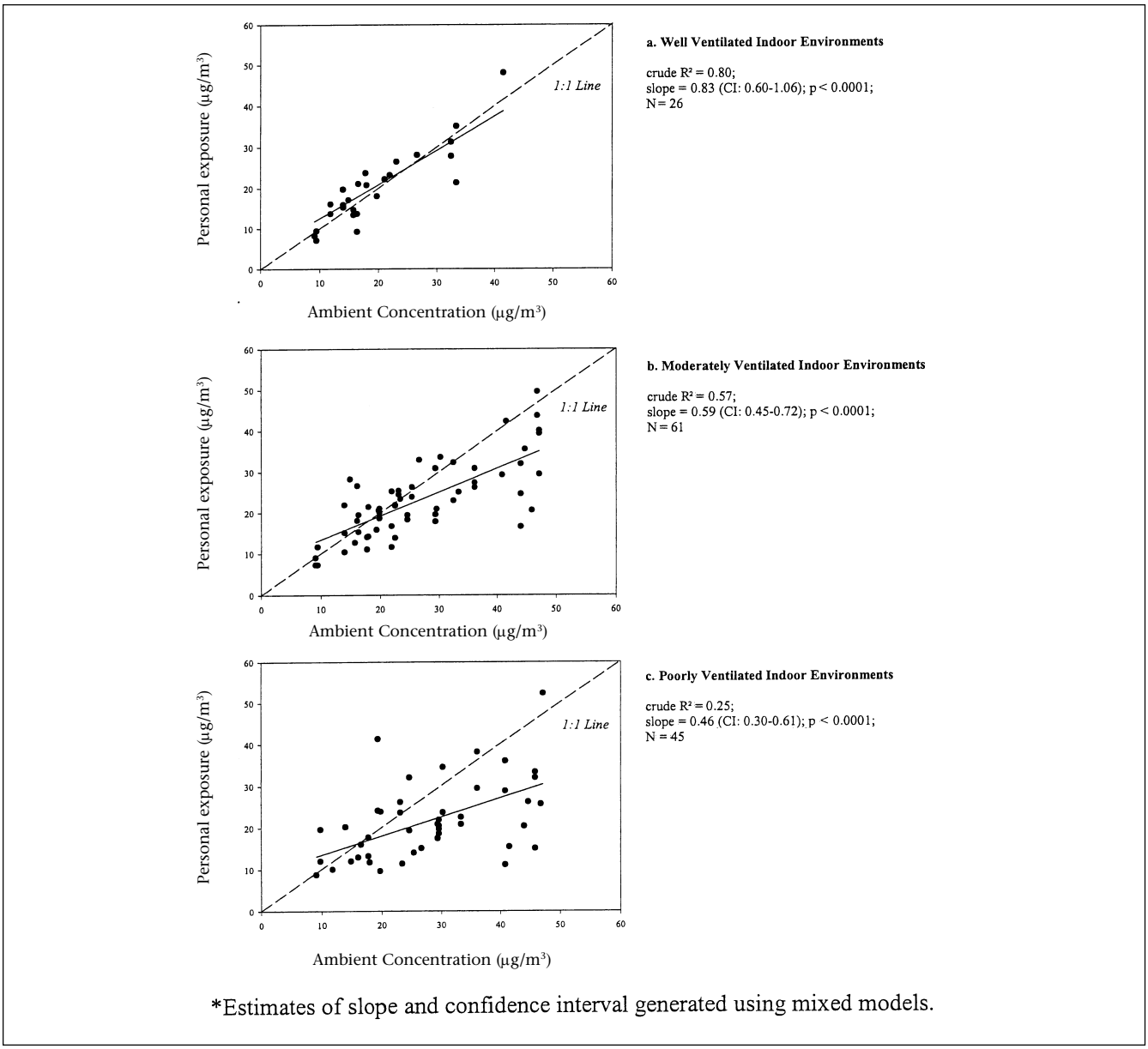

Figure 4. Personal $PM_{2.5}$ exposures vs ambient $PM_{2.5}$ concentrations: Summer 1998.*

were similar to those for the moderately and poorly ventilated categories in the summer. These results suggest that the weak personal–ambient associations observed for $PM_{2.5}$ for the poorly ventilated category are attributable primarily to the influence of non-ambient $PM_{2.5}$ sources.

The fraction of time subjects spent outdoors was added to F_v to examine whether consideration of time spent outdoors improves our ability to explain variability in the personal–ambient associations. Results from these analyses differ little from analyses involving F_v alone, suggesting that information about time spent outdoors explains little of the variability in the personal–ambient relationship. The unimportance of time spent outdoors may be attributed to the fact that subjects spent little time outdoors (approximately 5±6% of a 24-hr sampling period) and that the variation in this time was minimal.

Correlation between Ambient $PM_{2.5}$ and Personal Exposure to $PM_{2.5}$ of Ambient Origin

Personal exposure to $PM_{2.5}$ of ambient origin was estimated using the expression

$$\left(\frac{\left[SO_4^{2-}\right]_{Personal_{ij}}}{\left[SO_4^{2-}\right]_{Ambient_j}}\right)\cdot\left[PM_{2.5}\right]_{Ambient_j}$$

where $Personal_{ij}$ represents the personal exposure to SO_4^{2-} for subject i on day j and $Ambient_j$ represents the ambient concentration measured at the stationary site on day j. The "effective penetration" of ambient $PM_{2.5}$ to personal

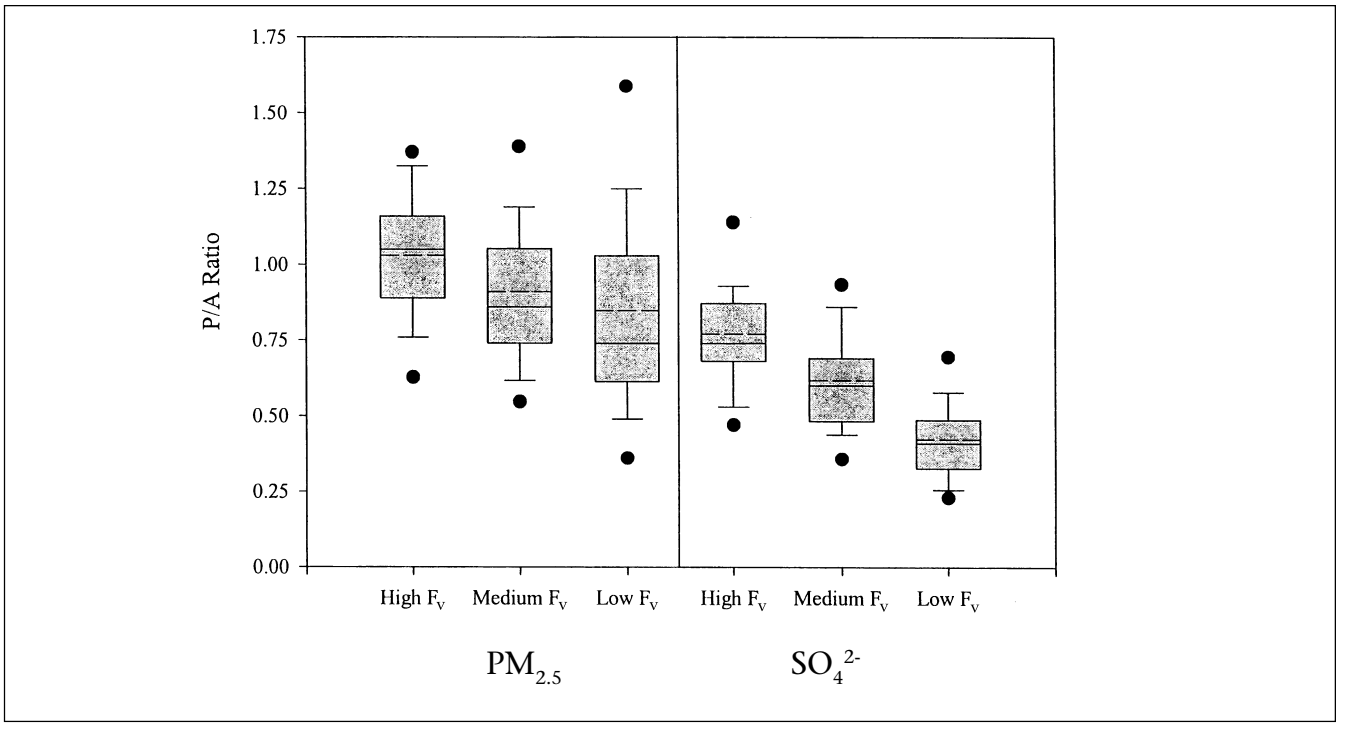

Figure 5. Personal–ambient ratios for $PM_{2.5}$ and SO_4^{2-} by ventilation status: Summer 1998.

exposures for all fine particles was assumed to equal that for SO_4^{2-}.

Estimates of personal exposure to $PM_{2.5}$ from ambient sources were plotted against total personal $PM_{2.5}$ exposures (Figure 7), where deviations from the 1:1 line reflect non-ambient source contributions to personal $PM_{2.5}$ exposures. As with previous analyses, the association between personal exposures to $PM_{2.5}$ of ambient origin and personal exposures to total $PM_{2.5}$ varied by ventilation status, with the scatter around the regression line greatest for the poorly ventilated category. Similarly, the contribution of personal $PM_{2.5}$ of ambient origin to personal $PM_{2.5}$ exposures varied by ventilation category. $PM_{2.5}$ of ambient origin comprised an average 75% (±13%) of personal $PM_{2.5}$ exposures for the well-ventilated category, 70% (±16%) for the moderately ventilated category, and 55% (±13%) for the poorly ventilated category. These differences are reflected in the intercepts of the regression of personal exposures to $PM_{2.5}$ on that for personal $PM_{2.5}$ of ambient origin, which equaled 2.7, 3.3, and 5.9 $\mu g/m^3$ for the well-, moderately, and poorly ventilated categories, respectively. The slopes for the three ventilation categories were comparable (1.2, 1.3, and 1.3 for high, medium, and low F_v categories, respectively).

Relationship between $PM_{2.5}$ and Other Pollutants

Correlations among the pollutants were examined to investigate the potential for confounding in epidemiologic studies of $PM_{2.5}$. Ambient $PM_{2.5\text{-}10}$, O_3, and NO_2 concentrations were significantly associated with ambient $PM_{2.5}$ concentrations during the summer ($r = 0.34$, $p < 0.04$; $r = 0.63$, $p < 0.0001$; and $r = 0.43$, $p < 0.008$, respectively). Significant correlations were also found between wintertime $PM_{2.5\text{-}10}$ concentrations and ambient $PM_{2.5}$ concentrations ($r = 0.57$, $p < 0.0004$). Ambient concentrations of $PM_{2.5\text{-}10}$, O_3, and NO_2 were also significantly associated with personal $PM_{2.5}$ exposures and personal exposure to $PM_{2.5}$ of ambient origin for some subjects. This was especially true for NO_2, where the personal $PM_{2.5}$ exposures for 10 of the 28 subjects (5 subjects per season) were significantly associated with ambient NO_2 concentrations (Table 7).

For personal exposures, significant correlations among the pollutants were not observed. Personal $PM_{2.5}$ exposures were not significantly associated with $PM_{2.5\text{-}10}$, O_3, NO_2, or SO_2 when analyzed either cross-sectionally, by individual, or by ventilation status. Similarly, associations between personal exposures to $PM_{2.5}$ of ambient origin and of non-ambient origin were not significant, with only one subject having a significant correlation coefficient. Personal exposures to $PM_{2.5}$ of ambient origin were also not significantly associated with personal exposures to the gaseous pollutants.

DISCUSSION AND CONCLUSIONS

Personal exposures to all of the measured pollutants tended to be lower than corresponding ambient concentrations. For PM, this finding is contrary to the results from several studies of healthy individuals, which attributed higher personal particulate exposures to the presence of a

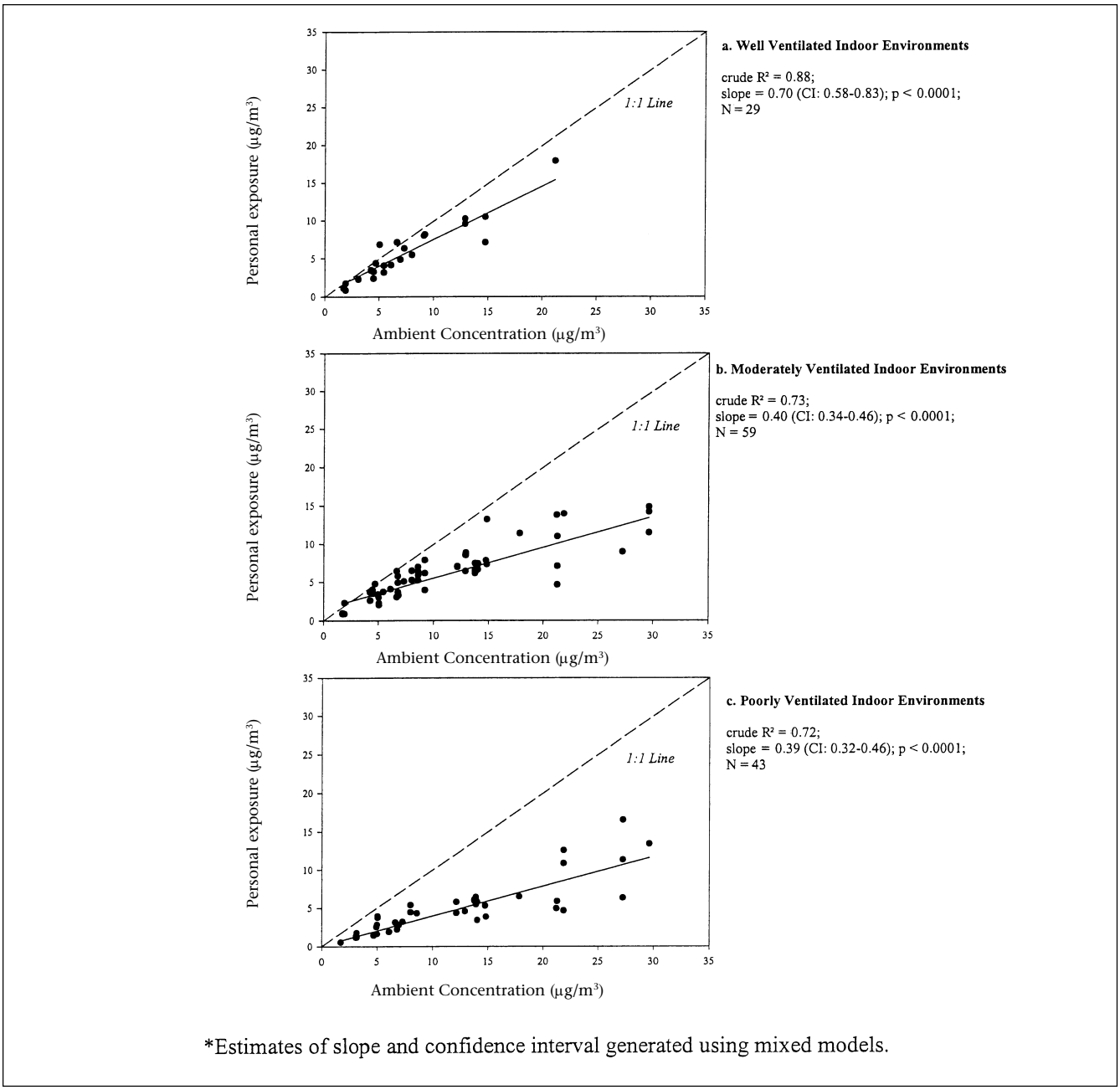

Figure 6. Personal SO_4^{2-} exposures vs ambient SO_4^{2-} concentrations: Summer 1998.*

personal particulate cloud and indoor and personal particulate sources.[7,15,20] Lower personal particulate exposures as compared with ambient concentrations, however, have been found in two studies of individuals with limited exposure to indoor particulate sources. Bahadori et al., for example, found mean personal $PM_{2.5}$ concentrations of individuals with chronic obstructive pulmonary disease to be lower than corresponding outdoor concentrations and attributed these lower exposures to the cohort's low activity level.[24] Similarly, Tamura et al. found mean personal PM_{10} exposures to be lower than corresponding outdoor levels for a cohort of older adults with limited exposure to indoor PM sources.[21] It is possible that similarly reduced exposures to indoor and personal particulate sources may also have contributed to the observed lower personal particulate exposures for our Baltimore cohort.

From the current analyses, it is clear that indoor ventilation was an important determinant of personal $PM_{2.5}$ exposures through its competing effects on effective penetration efficiency and indoor source contributions. These effects were clearly illustrated by the relationship between ventilation status and the personal–ambient associations for SO_4^{2-} and $PM_{2.5}$. This relationship was independent of subject, as evidenced by the fact that most individuals had exposures that spanned ventilation categories and by the results from the mixed models that

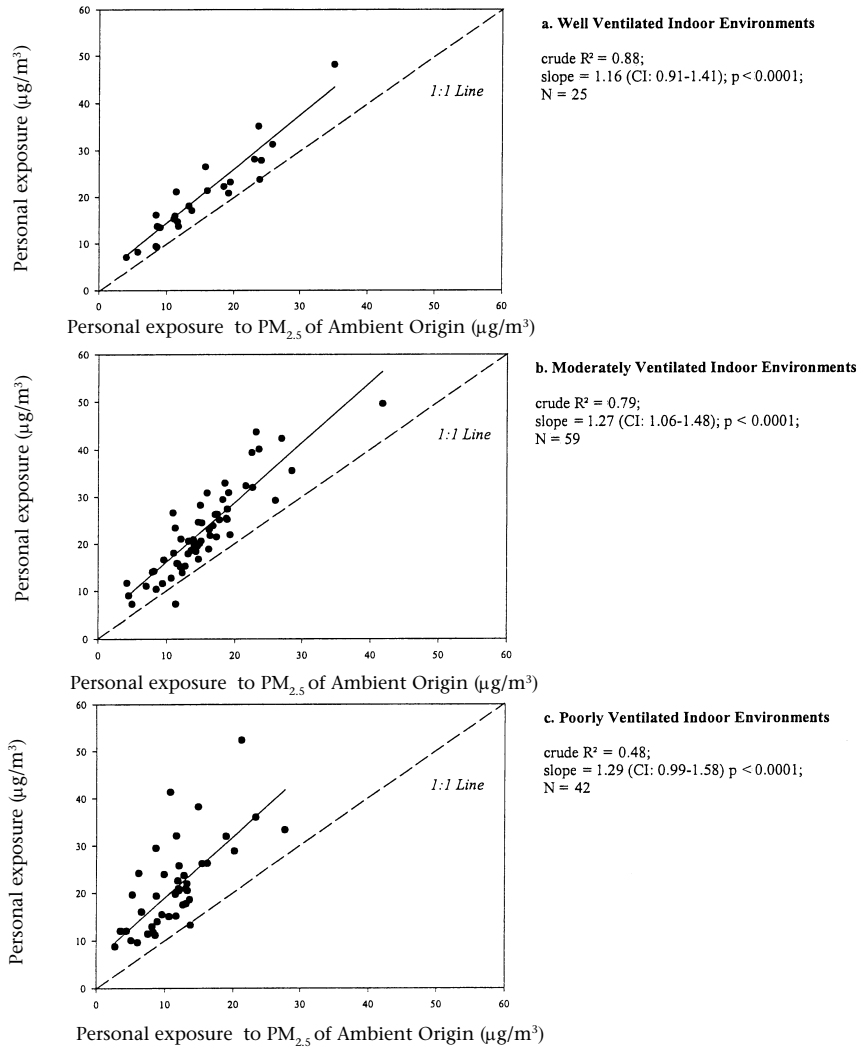

Figure 7. Personal $PM_{2.5}$ exposures vs personal exposure to $PM_{2.5}$ of ambient origin: Summer 1998.*

controlled for subject. For SO_4^{2-}, both the slopes of the personal-on-ambient regression lines and the median personal/ambient ratios decreased significantly with ventilation status, where the slopes and ratios were highest for the well-ventilated category and were lowest for the poorly ventilated category. This decrease, however, was not accompanied by a concomitant decrease in the strength of the personal–ambient relationship for SO_4^{2-}, as shown by a similar degree of scatter around the regression line for all three ventilation categories. Since SO_4^{2-} has relatively few indoor or personal sources, the observed decreases in personal–ambient slopes and ratios for SO_4^{2-} with ventilation may be attributed primarily to corresponding reduction in the effective penetration efficiency of particles from outdoor to personal environments. This reduction in the effective penetration efficiency with ventilation affected the magnitude of the personal–ambient association but, importantly, not the strength of this relationship.

The slopes and median ratios also decreased for $PM_{2.5}$, which may also be attributed to reduced effective penetration efficiencies in poorly ventilated environments. These reductions, however, were less substantial than those observed for SO_4^{2-}, probably as a result of the influence of indoor $PM_{2.5}$ sources, which have been shown in several studies to be greatest in poorly ventilated indoor environments.[7,8,23]

Evidence of the influence of indoor ventilation conditions on indoor $PM_{2.5}$ source contributions is provided by estimates of the fraction of personal $PM_{2.5}$ that is of ambient origin. As reflected by both the intercepts of the regression lines and by the ratios of personal $PM_{2.5}$ of ambient origin to total personal $PM_{2.5}$, this fraction was

Table 7. Correlations between personal $PM_{2.5}$ and ambient pollutant concentrations.[a]

		Personal $PM_{2.5}$ vs Ambient:				Personal $PM_{2.5}$ of Ambient Origin vs Ambient:		
	Subject	$PM_{2.5}$	O_3	NO_2	$PM_{2.5-10}$	O_3	NO_2	$PM_{2.5-10}$
SUMMER	SA1	0.55	0.15	0.38	–0.12	0.27	0.71[b]	0.15
	SA2	0.85[b]	0.31	0.66[b]	0.57	0.21	0.64	0.68
	SA5	0.89[b]	0.18	0.82[b]	0.64	0.33	0.81[b]	0.79
	SB1	0.65[b]	0.40	–0.15	0.38	0.89[b]	–0.74[b]	–0.03
	SB2	–0.21	–0.62	0.81[b]	0.15	0.26	0.08	0.33
	SB3	0.82[b]	0.55	–0.14	–0.04	0.52	–0.20	0.00
	SB4	0.73[b]	0.62[b]	–0.34	–0.12	0.45	–0.29	–0.14
	SB5	0.73[b]	0.45	–0.42	0.23	0.36	–0.48	0.33
	SB6	0.53	0.15	–0.38	0.12	–0.03	–0.57	0.32
	SC1	0.95[b]	0.78[b]	0.66[b]	0.65[b]	0.83[b]	0.63[b]	0.57[b]
	SC2	0.78[b]	0.68[b]	0.36	0.51	0.66[b]	0.65[b]	0.76[b]
	SC3	0.85[b]	0.78[b]	0.73[b]	0.68[b]	0.69[b]	0.71[b]	0.80[b]
	SC4	0.78[b]	0.66[b]	0.59	0.70[b]	0.50	0.50	0.51
	SC5	0.55	0.51	0.32	0.43	0.34	0.33	0.27
WINTER	WA1	0.22	–0.18	–0.26	–0.05	–0.78[b]	–0.04	–0.24
	WA2	–0.38	–0.07	–0.36	–0.70	–0.15	–0.15	0.02
	WA4	–0.18	0.67[b]	–0.22	–0.29	–0.33	0.20	0.00
	WA5	0.22	–0.43	0.61[b]	0.50	–0.72[b]	–0.09	0.40
	WB1	0.80[b]	–0.84	0.77[b]	0.41	–0.87[b]	0.53	0.66[b]
	WB2	0.62[b]	–0.32	0.59[b]	0.09	–0.76[b]	0.59[b]	0.59[b]
	WB3	0.55	–0.45	0.62[b]	0.04	–0.77[b]	0.56	0.60
	WB4	–0.12	–0.01	0.34	–0.10	–0.80[b]	0.68[b]	0.48
	WC1	0.74[b]	–0.62	–0.15	0.44	–0.64[b]	0.02	0.69
	WC2	0.79[b]	–0.88	0.17	0.77[b]	–0.87[b]	0.25	0.71[b]
	WC3	0.28	–0.42	0.03	0.57	–0.77[b]	0.30	–0.45
	WC4	0.19	–0.84	0.50	0.45	–0.72[b]	0.22	0.67
	WC5	0.81[b]	–0.62	0.08	0.81[b]	–0.76[b]	0.05	0.42
	WC6	0.01	–0.03	0.65[b]	0.37	–0.75[b]	0.19	–0.45
Median	Summer	0.76	0.48	0.37	0.41	0.41	0.42	0.33
Median	Winter	0.25	–0.43	0.26	0.39	–0.76	0.21	0.45

Notes: [a]Correlations represent Spearman's *r* values; [b]Significance at the $\alpha = 0.05$ level.

lowest for the poorly ventilated as compared with the well- and moderately ventilated categories. These results indicate that the contribution of indoor $PM_{2.5}$ sources to personal $PM_{2.5}$ exposures was highest when individuals spent little to no time in well-ventilated—or high air-exchange rate—indoor environments.

Given these results, it was not surprising that the personal–ambient association for $PM_{2.5}$ also declined with ventilation status and that this decline was more pronounced than that for SO_4^{2-}. As the contribution of particles of indoor origin to personal $PM_{2.5}$ exposures increased, the corresponding association between personal exposures and ambient concentrations became weaker. In contrast, the personal SO_4^{2-} exposures were strongly correlated with ambient concentrations for all three ventilation categories, suggesting that personal–ambient associations involving personal exposures to particles of ambient origin are strong and are less affected by indoor ventilation conditions as compared with personal exposures for particles of non-ambient origin. As a result, personal $PM_{2.5}$ exposures of individuals who spent more time in these well-ventilated indoor environments are more strongly associated with corresponding ambient concentrations.

It should be added that the above associations between personal exposure and ambient concentrations were unique to $PM_{2.5}$ and were not observed for any of the other measured pollutants. As a result, the potential for confounding appears to be limited, despite significant correlations that were observed among ambient pollutant concentrations. In contrast to ambient concentrations, neither personal exposures to total $PM_{2.5}$ nor $PM_{2.5}$ of ambient origin were significantly correlated with personal exposures to the co-pollutants (i.e., $PM_{2.5-10}$, $PM_{2.5}$ of non-ambient origin, O_3, NO_2, and SO_2). Not surprisingly, personal–ambient associations for $PM_{2.5-10}$, O_3, NO_2, and SO_2 were similarly weak and insignificant. Since a confounder

must be associated with the exposure of interest, these results provide among the first evidence from an exposure study that the effects observed in the PM epidemiologic studies are probably not due to confounding by the measured PM co-pollutants. Given the strong correlations among the ambient pollutants and the strong personal–ambient associations for $PM_{2.5}$, however, ambient co-pollutant concentrations may be appropriate surrogates for personal exposures to $PM_{2.5}$ or $PM_{2.5}$ of ambient origin for some individuals. It should be noted that other, unmeasured co-pollutants, such as carbon monoxide and specific PM components, may also act as surrogates for personal $PM_{2.5}$ or, alternatively, may confound the observed associations between ambient $PM_{2.5}$ and health effects. The importance of these unmeasured co-pollutants should be evaluated in future studies.

It is also important to note that inter-pollutant variability in method precision between PM and the gaseous pollutants and the general low level of personal gaseous exposures may account for some of this lack of correlation among personal $PM_{2.5}$ and its co-pollutants. An analysis employing more precise methods of co-pollutant sampling would improve the strength of these findings. A detailed analysis of subject time-activity diaries is expected to yield more information of the relationships outlined in this paper and the corresponding strength between ambient concentrations and personal exposures.

ACKNOWLEDGMENTS

The authors wish to thank all of the participants of the study as well as M. Wolfson, G. Allen, S. Ferguson, L.-T. Chang, P. Catalano, J. Evans, A. Wheeler, J. Baker, C. Jang, D. Millman, J. Sullivan, A. Joshi, F. Plucienik, R. Simon, M. Erdman, C. Howard-Reed, G. Lau, J. Sekula, D. Belliveau, K. Misra, N. Alba, and R. Cheung. Fixed-site monitoring data were provided, in part, by the Maryland Department of the Environment. This study was funded by the Electric Power Research Institute and the American Petroleum Institute. Additional funding for this analysis was provided by the Harvard-EPA Center on Particle Health Effects (grant #R827353-01-0).

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