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Ambient Site, Home Outdoor and Home Indoor Particulate Concentrations as Proxies of Personal Exposures

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Abstract

Despite strong longitudinal associations between particle personal exposures and ambient concentrations, previous studies have found considerable inter-personal variability in these associations. Factors contributing to this inter-personal variability are important to identify in order to improve our ability to assess particulate exposures for individuals. This paper examines whether ambient, home outdoor and home indoor particle concentrations can be used as proxies of corresponding personal exposures. We explore the strength of the associations between personal, home indoor, home outdoor and ambient concentrations of sulfate, fine particle mass ($PM_{2.5}$) and elemental carbon (EC) by season and subject for 25 individuals living in the Boston, MA, USA area. Ambient sulfate concentrations accounted for approximately 70 to 80% of the variability in personal and indoor sulfate levels. Correlations between ambient and personal sulfate, however, varied by subject (0.1 – 1.0), with associations between personal and outdoor sulfate concentrations generally mirroring personal-ambient associations (median subject-specific correlations of 0.8 to 0.9). Ambient sulfate concentrations are good indicators of personal exposures for individuals living in the Boston area, even though their levels may differ from actual personal exposures. The strong associations for sulfate indicate that ambient concentrations and housing characteristics are the driving factors determining personal sulfate exposures. Ambient $PM_{2.5}$ and EC concentrations were more weakly associated with corresponding personal and indoor levels, as compared to sulfate. For EC and $PM_{2.5}$, local traffic, indoor sources and/or personal activities can significantly weaken associations with ambient concentrations. Infiltration was shown to impact the ability of ambient concentrations to reflect exposures with higher exposures to particles from ambient sources during summer. In contrast in the winter, lower infiltration can result in a greater contribution of indoor sources to $PM_{2.5}$ and EC exposures. Placing EC monitors closer to participants' homes may reduce exposure error in epidemiological studies of traffic-related particles, but this reduction in exposure error may be greater in winter than summer. It should be noted that approximately 20% of the EC data were below the field limit of detection, making it difficult to determine if the weaker associations with the central site for EC were merely a result of methodological limitations.

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Introduction

Numerous epidemiological studies have linked ambient particle concentrations with adverse health outcomes,[1,2] particularly for sensitive populations, such as individuals with cardiovascular or respiratory disease.[2–4] Additional studies suggest that the observed health effects may be due specifically to fine particles ($PM_{2.5}$), which consist primarily of combustion-related components, such as sulfate (SO_4^{2-}), elemental carbon (EC) and organic carbon.[3,5,6]

These epidemiologic studies have relied primarily on ambient concentrations to reflect the ambient component of personal particulate exposures for their study populations. To date, only a small number of exposure assessment studies have evaluated the ability of ambient concentrations, measured at a stationary site, to predict ambient exposures. Exposure studies have identified several factors that may influence the relationship between personal exposures and ambient concentrations, including home ventilation, indoor sources, and time activity patterns.[7–9] The impact of these factors, however, is not well quantified, particularly for specific fine particle components.

In this paper we characterize personal exposures and indoor and outdoor (outside home) and ambient (at a central site) concentrations of SO_4^{2-}, $PM_{2.5}$ and EC for a panel of individuals with pre-existing cardiorespiratory disease living in the Boston, MA, metropolitan area. As part of this characterization, we examine whether ambient site, home outdoor and home indoor concentrations can serve as proxies of personal exposures to SO_4^{2-}, $PM_{2.5}$ and EC.

Methods

Study Design

Simultaneous 24-hour integrated personal, indoor, and outdoor SO_4^{2-}, $PM_{2.5}$ and EC concentrations were measured for 25 individuals living in the metropolitan Boston, MA, during winter (November 1999-January 2000) and summer (June-July 2000). Fifteen participants were monitored in each season, with five of the 25 individuals participating in both seasons. Monitoring for each individual was conducted over seven consecutive days. During a given sampling session, three to four homes were measured simultaneously, and each home had one to two individuals take part in the personal monitoring. In total, 30 seven-day sampling sessions were conducted during the study, comprising 210 sample-days.

The 24-h integrated samples were exchanged every morning of the study between approximately 7:00 and 11:00 AM. Personal samplers were worn on the shoulder strap of a backpack. Participants were asked to bring the sampler with them at all times but were allowed to put the sampler nearby during stationary activities. Indoor samplers were placed inside the main activity room of the home on a tripod, approximately one meter from doors, windows and vents. Outdoor samplers were placed in the yard on tripods, as possible at least one meter from the house, trees, etc. Inlets for both the indoor and outdoor samplers were placed approximately four feet above the ground. Corresponding 24-hour SO_4^{2-}, $PM_{2.5}$ and EC samples were collected each day, beginning at 9:00 AM, at a stationary ambient monitoring ("ambient") site located at the Harvard School of Public Health in Boston, MA. The ambient site was located approximately 20 meters above ground level, which was a higher elevation than most of the homes in this study.

Subjects were recruited through radio and newspaper advertisements, fliers in hospitals and doctors' offices, and direct recruiting at senior centers. For eight participants during winter and six during summer, personal exposure monitoring was also conducted for a partner

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living in the home. Consent forms and study procedures were approved by the Human Subjects Committee of the Harvard School of Public Health.

Sampling Methods

All personal, indoor, and outdoor samples were collected using the multi-pollutant personal sampler, which contains Harvard Personal Environmental Monitors (HPEMs).[10] The sampler consisted of individual samplers with separate lines to collect SO_4^{2-}, $PM_{2.5}$ and EC, which were connected to a single pump. Flows in each line were controlled using valves to achieve flows within 10% of the target flow rates. Sampler flows were measured both before and after sampling.

$PM_{2.5}$ samples were collected using HPEMs, small inertial impactors that collected $PM_{2.5}$ on 37-mm Teflon filters. All Teflon filters were refrigerated immediately after collection to minimize semi-volatile losses. Barometric pressure corrections were applied to each of the pre- and post-sampling weights.[11] SO_4^{2-} concentrations were subsequently determined by extracting the $PM_{2.5}$ filters and analyzing the aqueous extract by ion chromatography. EC samples were collected using a HPEM with a single pre-fired quartz filter. Collected samples were subsequently analyzed using thermal optical reflectance by the Desert Research Institute.[12]

Questionnaires

Home characteristics information was collected for each day of sampling using questionnaires that asked for information on household activities and conditions that may have affected indoor particle concentrations. Participants also completed daily time-activity diaries denoting their activities and location every 15 minutes. All questionnaires were developed and provided by the US Environmental Protection Agency.[13]

Quality Assurance

Standard methods were used to measure precision and limits of detection (LODs).[14] Data from at least 12 duplicate pairs were used to estimate precision of the sampling method for SO_4^{2-}, $PM_{2.5}$ and EC. Absolute precision was calculated using the root mean squared difference of the duplicate samples divided by $\sqrt{2}$ and is reported in $\mu g/m^3$. The LODs for SO_4^{2-}, $PM_{2.5}$ and EC were calculated as three times the method precision. The absolute precision was then divided by the mean of the duplicate samples yielding the relative precision.

Since $PM_{2.5}$ and EC field blanks were shown to be statistically different from zero, samples were corrected using the median blank correction value for $PM_{2.5}$: 3.8 μg (winter), 8.7 μg (summer); and for EC: 0.1 μg (winter), 0.4 μg (summer). The LOD and relative precision for SO_4^{2-} were 0.6 $\mu g/m^3$ and ± 6 to 8%, respectively, during both seasons. The LODs for $PM_{2.5}$ ranged from 3 to 4 $\mu g/m^3$ and precision was ±10% in both seasons, but a larger fraction of the winter than summer $PM_{2.5}$ concentrations were less than the LOD. The LODs for EC were approximately 1 $\mu g/m^3$, and about 20% of the EC samples were below the LOD during both seasons. The relative precision for EC was ±30% during winter and ±22% during summer.

Data analysis

SO_4^{2-}, $PM_{2.5}$ and EC concentrations are reported in $\mu g/m^3$. Negative pollutant concentrations and values below the method detection limit were included in the analyses.[15] It was determined that two $PM_{2.5}$ data points -one personal and one indoor- were substantially influenced by an extreme cooking event and were excluded from the statistical

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analyses. $PM_{2.5}$ and SO_4^{2-} results from two participants were also excluded due to humidifier use, which was a significant source of these particle species. Since the effects of specific sources were not a focus of this paper, these extreme points were excluded from this analysis. A subsequent paper will present results associated with specific indoor or personal sources as well as the influence of activity patterns on personal exposures.

Associations between personal, indoor, outdoor and ambient concentrations for each pollutant were characterized using univariate mixed effects models ("longitudinal models") that included a random subject term.[16] For each pollutant six longitudinal models were tested, stratified by season: (1) "personal-indoor" with personal as the dependent variable and indoor as the independent variable; (2) "personal-outdoor" with personal as the dependent variable and outdoor as the independent variable; (3) "personal-ambient" with personal as the dependent variable and ambient as the independent variable; (4) "indoor-outdoor" with indoor as the dependent variable and outdoor as the independent variable; (5) "indoor-ambient" with indoor as the dependent variable and ambient as the independent variable; and (6) "outdoor-ambient" with outdoor as the dependent variable and ambient as the independent variable. Coefficient of determination (R^2) values were obtained from these models to show the strength of the associations between personal, indoor, outdoor and ambient levels for each pollutant.[17]

Subject-specific Spearman correlation coefficients are also presented to show how the strength of the associations varied by participant. Only correlations containing four or more valid observations were used in this analysis. Consequently, $PM_{2.5}$ results for five participants are not presented ($N<4$). For SO_4^{2-}, four participants had fewer than four observations, and three participants had too few EC observations to present subject-specific results.

Results

Participant and Housing Characteristics

The median home age was more than 40 years during both seasons. During the winter, approximately half of the sampled homes were apartments as compared to one-third in summer. Similarly, half the sampled homes had gas stoves in winter and only one-third in summer. Overall the homes sampled during the summer were located farther from the central ambient monitoring site, in more rural areas with less traffic volume. Results not presented here showed that air exchange rates for most homes were relatively high with more than one-third greater than 1 air exchange per hour during winter and more than two-thirds during summer.

On average, participants spent approximately 80% of the time indoors at home with little difference by season. Time spent outdoors was significantly lower in winter compared to summer ($p = 0.03$); however, time spent outdoors was low in both seasons (mean = 2.5% or 36 minutes in winter and 6.0 % or 86 minutes in summer). Time spent indoors away from home (~7 – 11%) and time spent in transit (~5 – 7 %) were comparable between the two seasons.

Microenvironmental Pollutant Levels

Sulfate—Table 1 provides summary statistics by season for personal, indoor, outdoor and ambient concentrations of SO_4^{2-}, $PM_{2.5}$ and EC. Geometric mean SO_4^{2-} concentrations varied by microenvironment and season, ranging from 1.3 to 3.1 μg/m^3. During both seasons, personal and indoor geometric mean SO_4^{2-} concentrations were comparable as

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were outdoor and ambient SO_4^{2-} concentrations. SO_4^{2-} concentrations and exposures were higher during summer than winter.

Personal-indoor SO_4^{2-} ratios were typically close to 1, which was expected since individuals spent most of their time indoors at home (Table 2). One personal-indoor ratio was well above one for a home with very low measured indoor SO_4^{2-}. All of the indoor-outdoor SO_4^{2-} ratios were less than one, with higher ratios in summer than winter. Personal-outdoor and personal-ambient SO_4^{2-} ratios were comparable to each other during both seasons, due to the comparable SO_4^{2-}concentrations measured at the ambient and outdoor sites. Additionally, the indoor-outdoor ratios were also similar to the personal-outdoor and personal-ambient SO_4^{2-} ratios, as personal and indoor SO_4^{2-} concentrations were comparable.

$PM_{2.5}$—In contrast to SO_4^{2-}, personal $PM_{2.5}$ exposures were higher than indoor, outdoor or ambient levels during winter with geometric means ranging from 7.3 to 10.4 μg/m^3 (Table 1). The opposite was true during summer, when the personal $PM_{2.5}$ geometric mean concentration (10.0 μg/m^3) was less than the indoor, outdoor and ambient geometric mean concentrations (12.0, 12.5 and 11.8 μg/m^3, respectively). Similarly, the median personal-indoor, personal-outdoor and personal-ambient $PM_{2.5}$ ratios were greater than 1 in winter but not in summer (Table 2). While personal-outdoor $PM_{2.5}$ ratios were higher in winter than summer, indoor-outdoor ratios were lower in winter than summer. Personal-outdoor and personal-ambient $PM_{2.5}$ ratios were very similar to those found for SO_4^{2-} only during summer. This finding was likely due to the fact that indoor $PM_{2.5}$ sources have a greater impact on personal $PM_{2.5}$ exposures during winter when homes have reduced ventilation.

Elemental Carbon—The personal, indoor and ambient geometric mean EC concentrations during winter were generally comparable to those measured during summer (Table 1). However, outdoor EC was higher during winter than summer, likely due to reduced traffic impacts for the homes measured during summer. Ambient EC concentrations measured at the central site were generally lower than corresponding home outdoor EC levels in both seasons.

The median personal-indoor EC ratio was less than one during winter and close to one during summer (Table 2). During winter, median personal-outdoor and indoor-outdoor EC ratios were less than one. In contrast, median personal-ambient, indoor-ambient and outdoor-ambient ratios were all greater than one during winter, indicating higher local levels of EC compared to the central ambient site in that season. All summertime median EC concentration ratios were approximately one, similar to the SO_4^{2-} and $PM_{2.5}$ ratios seen in summer.

Home outdoor SO_4^{2-} (mostly in the form of ammonium sulfate) represented approximately 40% of home outdoor $PM_{2.5}$ in this study during both seasons. EC comprised 19 and 13% of home outdoor $PM_{2.5}$ during winter and summer, respectively. As a result, home outdoor SO_4^{2-} and EC comprised approximately 50 to 60% of home outdoor $PM_{2.5}$.

Microenvironmental Pollutant Relationships

Mixed effects regression models were used to assess the strength of the associations between pollutant levels measure in the four microenvironments. SO_4^{2-}, $PM_{2.5}$ and EC model results are presented in Table 3 for winter and Table 4 for summer. The slopes in these models provide an indication of the strength of the relationship between concentrations measured in the different microenvironments. The intercepts provide an indication on average of the mass not accounted for by the independent variable in each model. In addition, subject-

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specific Spearman correlation coefficients are provided for SO_4^{2-}, $PM_{2.5}$ and EC in Figures 1–3, respectively.

Sulfate—The regression model results for SO_4^{2-} during winter (Table 3) and summer (Table 4) show the strong associations between all of the microenvironments. Personal-indoor slopes were highest (0.9 in both seasons), and indoor SO_4^{2-}accounted for approximately 90% of the variability in personal SO_4^{2-} during both seasons. The personal and indoor models (personal-outdoor, personal-ambient, indoor-outdoor and indoor-ambient) all had slopes close to 0.4 during winter and 0.7 during summer. During winter, outdoor and ambient SO_4^{2-} accounted for approximately 70% of the variability in the personal and indoor levels, while during summer this was approximately 90%. Outdoor-ambient SO_4^{2-} models had lower slopes and R^2 values in winter compared to summer, indicating somewhat greater spatial variability in SO_4^{2-} during winter. It should be noted that only five of the homes were measured in both seasons. To address this potential limitation, the personal-ambient SO_4^{2-} models were run only on the five homes repeated during both seasons. The slopes from this more limited data set were 0.38 (95% CI: 0.30, 0.45) and 0.73 (95% CI: 0.64, 0.82) during winter and summer, respectively.

Subject-specific correlations among personal, indoor, outdoor, and ambient SO_4^{2-} concentrations presented in Figure 1 show similarly strong associations for most participants, although there was some inter-subject variability. This was especially the case for comparisons between personal or indoor with ambient SO_4^{2-} levels.

$PM_{2.5}$—The levels of $PM_{2.5}$ in the four microenvironments were also significantly associated with each other, but the effects were weaker than for SO_4^{2-} (Tables 3 and 4). While the slopes were very similar between SO_4^{2-} and $PM_{2.5}$, outdoor and ambient $PM_{2.5}$ accounted for less of the variability in personal and indoor concentrations compared to SO_4^{2-} likely due to participants' activities, especially in winter. This is further illustrated by the large intercepts for the personal models in winter. The large fraction of SO_4^{2-} comprising $PM_{2.5}$ (40%) would indicate similar model slopes; however, outdoor and ambient $PM_{2.5}$ were weaker predictors of personal and indoor $PM_{2.5}$ compared to SO_4^{2-}.

To further examine how associations varied by subject, subject-specific correlations for $PM_{2.5}$ are presented in Figure 2. As indicated by the regression model results, weaker associations were found for $PM_{2.5}$ than SO_4^{2-} in both seasons. The effect of personal activities on $PM_{2.5}$ personal models is pronounced, especially in winter when personal models showed weak or even negative associations for many of the participants.

Elemental Carbon—There were significant differences in the EC regression results compared to SO_4^{2-} and $PM_{2.5}$. During winter, the personal-outdoor and indoor-outdoor regression slopes were very similar to those for SO_4^{2-} and $PM_{2.5}$ with R^2 values of ~0.3 (Table 3). However, the models using the ambient EC data showed much higher slopes than for the outdoor models. These results indicated that significant spatial variability in winter EC may be driving this effect; however, the scatter plot presented in Figure 4a shows the limited range of EC at the ambient monitoring site compared to the outdoor sites during winter. As a result, the high slopes for the three ambient models may be due to the limited range in ambient EC concentrations.

During summer, EC showed consistently lower slopes compared to SO_4^{2-} and $PM_{2.5}$. All slopes were significant, except the indoor-ambient slope (Table 4). Outdoor and ambient EC concentrations explained approximately one-third or less of the variability in personal or indoor EC levels. Outdoor EC concentrations were better predictors of personal and indoor EC concentrations compared to ambient concentrations. For example, use of home outdoor

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EC to predict personal EC during summer increased the slope from 0.41 to 0.54 and the R^2 from 0.08 to 0.35. Figure 4b is a plot of the outdoor and ambient EC concentrations during summer, which further illustrates the relatively weak associations between the outdoor and ambient site concentrations during summer.

There was a great deal of inter-subject variability in the subject-specific EC correlations, with many participants having weak and even negative correlations, particularly during summer (Figure 3). Median correlations among personal, indoor, outdoor, and ambient EC concentrations were approximately 0.6 to 0.7 in winter and 0.2 to 0.6 in summer. Correlations for EC were weaker than those for SO_4^{2-} during both seasons, but somewhat stronger than wintertime $PM_{2.5}$ correlations.

Comparison of Personal and Partner Exposures

In both seasons, personal SO_4^{2-} exposures for individuals living in the same home were strongly correlated (personal-personal correlations in Figures 1–3), while corresponding comparisons of personal exposures for EC and $PM_{2.5}$ were significantly weaker. Using mixed models only on the subset of subjects with a partner that participated in the study, a partner's personal SO_4^{2-} exposures explained more than 90% of the variability in the study subjects' SO_4^{2-} exposures during both seasons (Table 5), and the slopes were >0.8. In contrast, the slopes and R^2 values for $PM_{2.5}$ were lower than for SO_4^{2-} and varied by season with higher slope and R^2 in summer. While personal-personal slopes and R^2 values for EC varied by season, they were higher in winter than summer. The differences in the personal-personal associations provide some indication of the effect of activity patterns on exposures, when housing characteristics remain constant, as was the case with the personal-personal comparisons. This shows that activity patterns strongly influence $PM_{2.5}$ exposures but may have less of an effect for SO_4^{2-} during both seasons and for EC during winter.

DISCUSSION

In this analysis, ambient SO_4^{2-} was strongly correlated with corresponding personal exposures and home indoor concentrations for individuals not using humidifiers, a source of indoor SO_4^{2-}, consistent with a number of previous studies in the US and Canada.[18–20] Associations with outdoor SO_4^{2-}concentrations were similar to those for ambient concentrations. The strong associations between personal SO_4^{2-} and ambient SO_4^{2-} are due to the high infiltration ratios of sulfate and the relatively low spatial variability of SO_4^{2-} in this study. High infiltration of SO_4^{2-} has been reported previously in Boston.[21] Limited spatial variability of SO_4^{2-} has also been shown in a number of previous studies in northeastern US cities[22,23] and provides an explanation for the equally strong associations between outdoor and personal SO_4^{2-}. It should be noted that there was somewhat greater spatial variability in outdoor SO_4^{2-} during the winter, likely due to reduced atmospheric mixing in colder temperatures. One outdoor monitor in particular had poor correlation with the ambient site during winter. This home was located less than 10 m from Boston Harbor, and previous studies have shown marine engines to contribute to airborne SO_4^{2-} levels,[24] which may explain this result.

The current results indicated that a seasonal difference existed in the SO_4^{2-} ratios and regression model slopes, which was likely due to lower infiltration in winter. Despite this finding, R^2 values and correlations were generally high in both seasons. Together these results suggest that personal or indoor SO_4^{2-} levels correlated consistently with outdoor or ambient levels regardless of the differences in the absolute levels in these microenvironments. This relationship may vary by geographic region as shown in Baltimore and Boston.[25] This variation may be due to factors, such as air conditioning or other housing characteristics that reduce infiltration. The infiltration into the homes in the current study

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was much greater during summer than for homes in a study in North Carolina,[26] likely due to greater air conditioning use for homes in that study and the age of the homes in the current study. While only five of the study homes were measured in both seasons, the model results indicated very similar personal-ambient associations for that subgroup compared with all of the homes measured. This indicates that the seasonal effect was not due merely to differences in the types or locations of the homes or even markedly different activity patterns for the participants that did not participant in both seasons.

While the R^2 values for the $PM_{2.5}$ were lower than those for SO_4^{2-}, the slopes of the mixed models for the $PM_{2.5}$ and SO_4^{2-} were remarkably similar. This indicates that personal and indoor $PM_{2.5}$ can be predicted using outdoor or ambient $PM_{2.5}$ albeit less accurately than for SO_4^{2-}. This is likely due to non-ambient sources of $PM_{2.5}$, such as indoor sources and resuspension from activities, although the poorer precision of the $PM_{2.5}$ measurements compared to those for SO_4^{2-} may have been a contributing factor.

A number of previous studies have shown higher personal than home indoor PM levels, referred to as a “personal cloud.”[9,26,27] There was evidence of personal cloud effects for $PM_{2.5}$ during winter based on the high personal-indoor ratios during winter. This contribution was not observed in the summer. The lack of personal cloud during summer may be due to the fact that homes were better ventilated (indoor sources contributed less to the indoor concentrations) and $PM_{2.5}$ concentrations were higher (lower relative impact of the personal cloud). Finally, the personal cloud was likely non-sulfate or non-EC-related PM for most participants, as personal-indoor SO_4^{2-} and EC ratios tended to be relatively close to or less than one.

A previous personal exposure study conducted in Boston[7] found similar correlations between personal, indoor and outdoor levels of $PM_{2.5}$ as those presented in this paper. This is in spite of the slightly higher $PM_{2.5}$ concentrations in the previous study. Mean subject-specific R^2 values for personal-indoor and indoor-outdoor $PM_{2.5}$ models were generally similar to the R^2 values presented here. Despite the non-random selection of participants in Rojas-Bracho (2000) and the current study, time spent indoors in both studies was very similar, which may account for the similarity in personal-indoor and personal-outdoor associations. In addition, the similar R^2 values for indoor-outdoor models in the two studies may indicate that housing characteristics are regionally consistent, regardless of absolute pollutant levels.

The spatial variability in EC during winter was pronounced. The ambient site showed little variability in either season. During summer, the variability at the outdoor sites was much less, but use of home outdoor monitoring data did improve the model results somewhat. These results indicate that spatial variability in traffic-related pollutants may differ by season, potentially due to meteorological factors. Previous studies have reported stronger personal-outdoor or indoor-outdoor associations for EC or black carbon than we saw in this study.[18,28–30] These weaker associations may also be due, in part, to lower EC concentrations in Boston and/or to our method imprecision as a consequence of low sampling flow rates, as the EC method was much less precise than either the SO_4^{2-} or $PM_{2.5}$ measurements.

The measurement of a second individual in the home allowed us to control for housing characteristics so we could evaluate the effect of differences in activity patterns on personal exposures. During both seasons SO_4^{2-} showed very strong associations between personal exposures of individuals living in the same home, implicating housing characteristics as the likely driving factor in determining the personal-outdoor and personal-ambient relationships. EC and $PM_{2.5}$ did not show the same strength of associations between personal exposures,

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and the strength of the relationships varied by season. Consistent with our "personal cloud" findings during winter, the personal-personal EC slope and R^2 were higher than those seen for $PM_{2.5}$, further indicating that non-EC and non- SO_4^{2-} -related compounds may predominate in the "personal cloud."

The significantly weaker associations for $PM_{2.5}$ and EC compared to SO_4^{2-} indicate that personal and household activities that generate $PM_{2.5}$ and EC, likely impact personal exposures. These can include cooking, cleaning and walking across carpet for $PM_{2.5}$[31] and candle burning for EC.[32] This can weaken the associations between personal exposures and outdoor or ambient concentrations. For $PM_{2.5}$ weaker associations compared to SO_4^{2-} are potentially due to resuspension of deposited PM in the home, proximity to indoor sources and exposures to sources when away from the home. Factors that may have led to weaker associations for EC include our method imprecision, exposures to EC during time away from home and/or proximity to EC sources in the home.

These results suggest that placement of outdoor EC monitors closer to participants' homes may reduce exposure error in epidemiological studies of EC and other traffic-related particles. This effect may be greater in winter than summer months. Infiltration was also shown to impact the ability of ambient concentrations to reflect exposures, as a strong seasonal difference in infiltration (based on indoor-outdoor sulfate ratios) was found. Greater ventilation during the summer may have resulted in significantly higher personal exposures to particles originating from ambient sources. In contrast in the winter, lower infiltration can result in a greater contribution of indoor sources to personal exposures to $PM_{2.5}$ and EC.

This study showed differences in associations by seasons, pollutant and subject; however, ambient SO_4^{2-} and $PM_{2.5}$ were generally good predictors of personal exposures. Based on these results epidemiological studies should factor in seasonal differences in personal-ambient associations, particularly in the northeaster U.S. Additionally, spatial variability in traffic-related pollutants may also vary by season, as a result, there may be more error in estimating exposures during winter than summer for these pollutants. It should also be noted that the participants in this study were not randomly selected, therefore, the results presented here may not be generalizable to a larger population.

Evaluating the relationships between microenvironmental levels of these particle species enabled us to understand some of the sources of exposure error associated with use of a central ambient monitor for estimating personal exposures. These results indicate that ambient SO_4^{2-} concentrations are good indicators of personal exposures to ambient SO_4^{2-} for individuals living in the Boston area, even though their levels may differ from actual personal exposures. The strong associations for SO_4^{2-} indicate that ambient concentrations and housing characteristics are the driving factors determining personal SO_4^{2-} exposures. For EC and $PM_{2.5}$, near home sources (e.g., local traffic), indoor sources and/or personal activities can significantly weaken associations with outdoor or ambient concentrations, although the significant fraction of EC samples below the field LOD likely contributed to these weakened associations.

Conclusions

Epidemiological studies generally rely on ambient site monitoring data to estimate the ambient component of personal particulate exposures. This study showed differences in personal-ambient associations by season, pollutant and subject; however, this analysis showed that ambient monitoring data can be used as proxies of personal exposures to pollutants, such as sulfate and $PM_{2.5}$, which have been shown to have more limited spatial

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variability over a metropolitan area. Based on these results epidemiological studies should factor in seasonal differences in personal-ambient associations, particularly in the northeastern U.S. Spatial variability in traffic-related pollutants may also vary by season, as a result, there may be more error estimating ambient exposures during winter than summer for these pollutants. However, limitations of our mesurement methods cannont be ruled out as contributing to the weaker between-site associations seen for EC.

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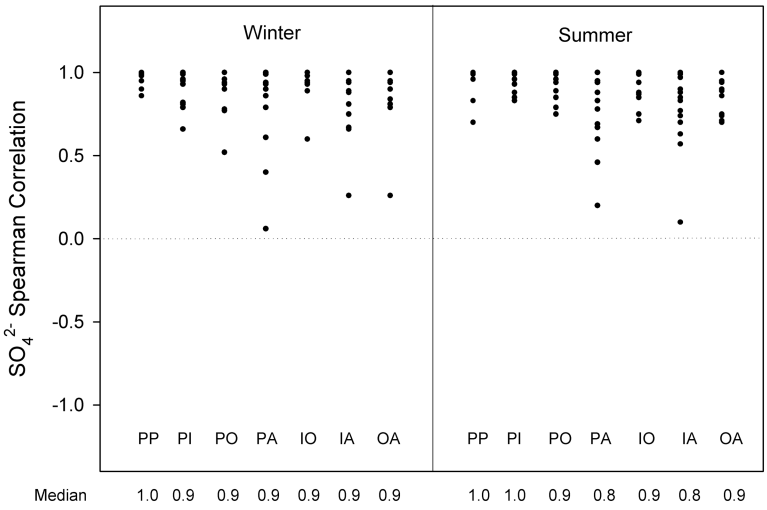

Figure 1.

Plot of subject-specific personal-personal (PP), personal-indoor (PI), personal-outdoor (PO), personal-ambient (PA), indoor-outdoor (IO) and outdoor-ambient (OA) SO_4^{2-} Spearman correlation coefficients. PP correlations are presented for the subset of subjects with a partner that also participated in the personal monitoring. Results are restricted to those individuals with at least four observations for each correlation.

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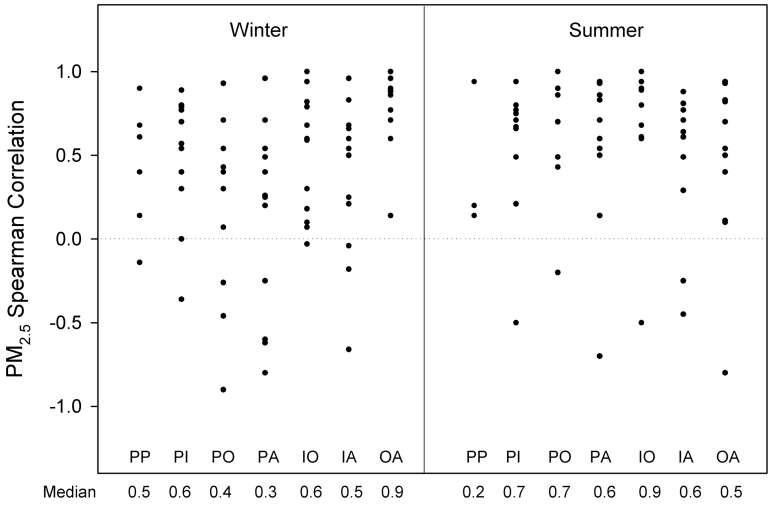

Figure 2.

Plots of subject-specific personal-personal (PP), personal-indoor (PI), personal-outdoor (PO), personal-ambient (PA), indoor-outdoor (IO) and outdoor-ambient (OA) $PM_{2.5}$ Spearman correlation coefficients. PP correlations are presented for the subset of subjects with a partner that also participated in the personal monitoring. Results are restricted to those individuals with at least four observations for each correlation.

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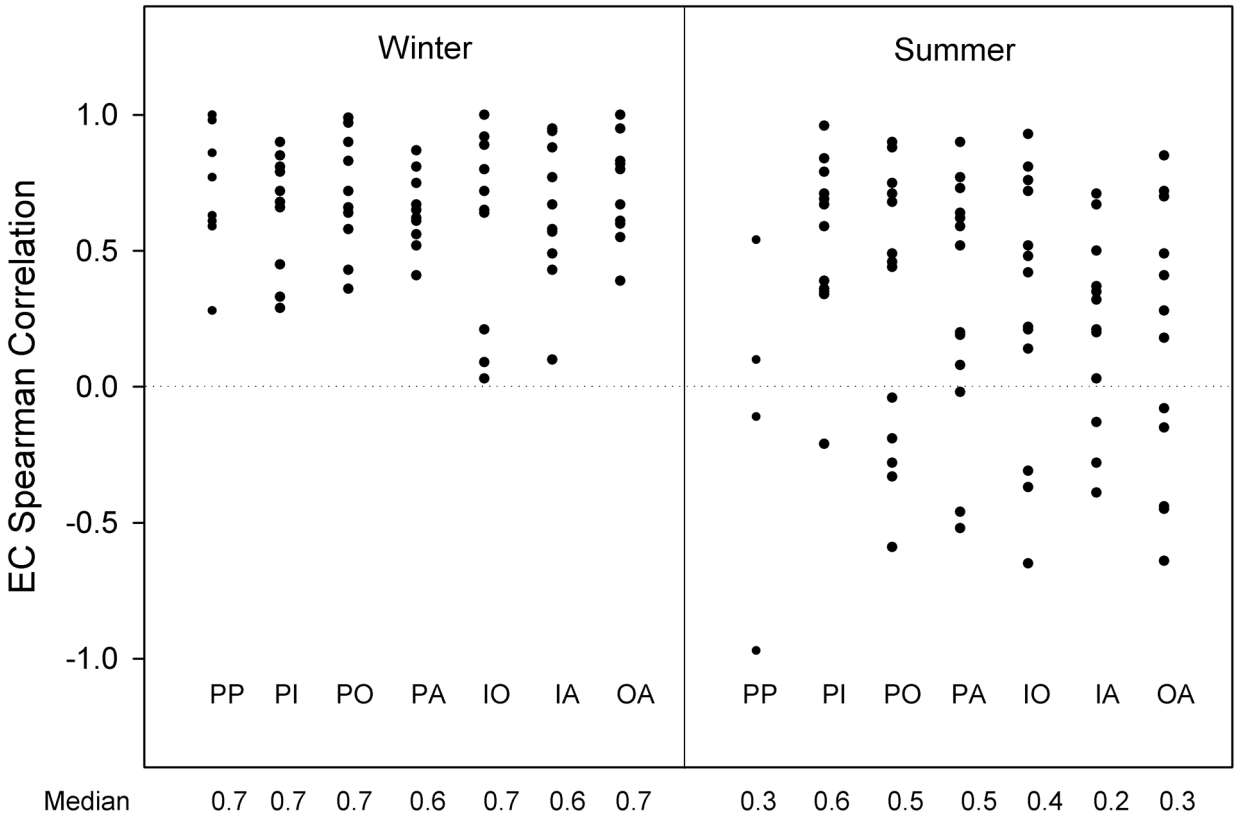

Figure 3.

Plots of subject-specific personal-personal (PP), personal-indoor (PI), personal-outdoor (PO), personal-ambient (PA), indoor-outdoor (IO) and outdoor-ambient (OA) EC Spearman correlation coefficients. PP correlations are presented only for the subset of subjects with a partner that also participated in the personal monitoring. Results are restricted to those individuals with at least four observations for each correlation.

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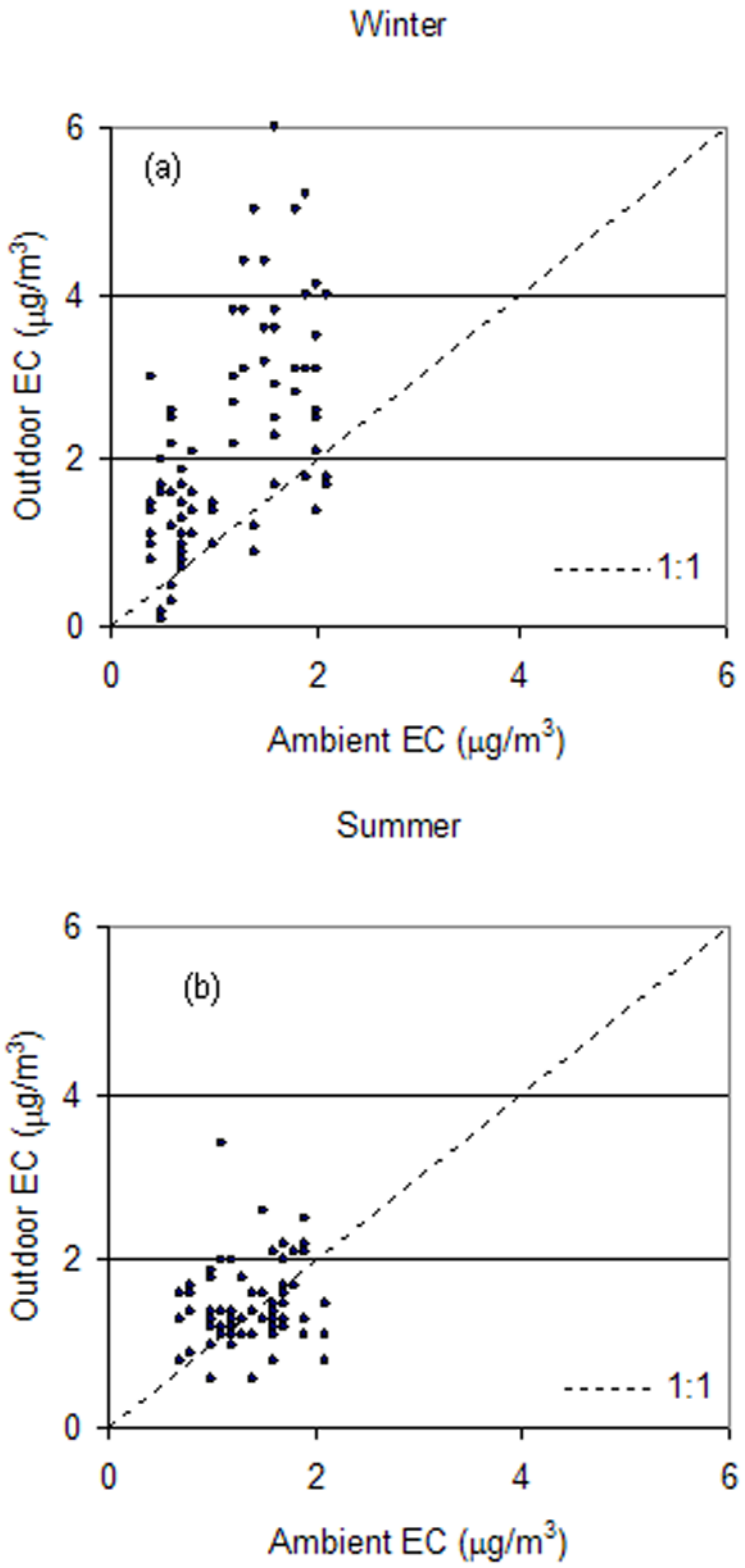

Figure 4.

Scatterplots of home outdoor EC compared to ambient site EC concentrations during winter (a) and summer (b).

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Table 1

Sampling Program

Season	Sampling Session	Start Date	End Date	Homes	Subjects	Sample Days
Winter 2000	1	11/15/99	11/21/99	3	6	21
	2	1/5/00	1/11/00	4	6	28
	3	1/15/00	1/21/00	4	6	28
	4	1/23/00	1/29/00	4	5	28
				15	23	105
Summer 2000	5	6/6/00	6/12/00	3	5	21
	6	6/15/00	6/21/00	3	4	21
	7	6/24/00	6/30/00	3	4	21
	8	7/11/00	7/17/00	3	5	21
	9	7/19/00	7/25/00	3	3	21
				15*	21	105
Total						210

[†]Five of these homes were also measured during the winter.

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Table 2

Descriptive statistics of personal, home indoor, home outdoor and stationary ambient monitoring site SO_4^{2-}, $PM_{2.5}$ and EC concentrations during winter and summer. All concentrations are in μg/m3.

Pollutant	Season	Location	N	Mean (SD) [a]	GM[b] (GSD)[c]	Min	Max	CV (%)[d]
SO_4^{2-}	Winter	Personal	80	1.4 (0.6)	1.3 (1.6)	0.3	3.6	43.2
		Indoor	85	1.5 (0.7)	1.3 (1.7)	0.2	3.4	46.0
		Outdoor	82	2.7 (1.2)	2.4 (1.7)	0.4	5.6	44.0
		Ambient	25	2.6 (1.4)	2.2 (1.9)	0.5	7.4	55.9
	Summer	Personal	100	3.1 (2.1)	2.5 (2.1)	0.3	8.5	65.8
		Indoor	103	3.1 (2.2)	2.5 (2.1)	0.4	10.7	69.1
		Outdoor	104	4.0 (2.7)	3.1 (2.2)	0.4	11.6	68.2
		Ambient	32	3.6 (2.2)	2.9 (2.1)	0.4	10.4	61.2
$PM_{2.5}$	Winter	Personal	75	12.0 (6.0)	10.4 (1.8)	1.4	30.8	50.0
		Indoor	85	10.1 (4.6)	7.3 (1.8)	2.4	28.0	60.4
		Outdoor	87	8.6 (5.2)	9.0 (1.7)	2.2	19.7	45.2
		Ambient	27	9.9 (5.1)	8.5 (1.8)	1.8	20.1	51.9
	Summer	Personal	82	10.0 (6.2)	8.5 (1.7)	2.6	35.7	62.1
		Indoor	101	12.0 (7.3)	10.3 (1.8)	1.1	45.0	60.7
		Outdoor	87	12.5 (7.6)	10.7 (1.7)	3.6	39.7	60.8
		Ambient	35	11.8 (5.5)	10.7 (1.6)	3.5	29.1	46.8
EC	Winter	Personal	88	1.6 (1.7)	1.2 (1.9)	0.0	11.6	107.2
		Indoor	95	1.9 (1.5)	1.5 (2.1)	0.1	10.7	80.8
		Outdoor	87	2.1 (1.3)	1.7 (2.2)	0.0	6.0	61.1
		Ambient	25	1.1 (0.6)	1.0 (1.7)	0.4	2.1	51.7
	Summer	Personal	99	1.4 (0.6)	1.3 (1.4)	0.6	4.7	42.4
		Indoor	100	1.5 (0.6)	1.4 (1.5)	-0.3	3.3	40.7
		Outdoor	102	1.6 (0.7)	1.4 (1.5)	0.6	4.7	46.8
		Ambient	33	1.3 (0.4)	1.3 (1.4)	0.5	2.1	28.6

[†] Arithmetic mean (standard deviation)

[‡] Geometric mean.

[§] Geometric standard deviation.

¶Percent coefficient of variation.

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Table 3

Subject-specific personal-indoor, personal-outdoor, personal-ambient, indoor-outdoor, indoor-ambient and outdoor-ambient concentration ratios by season for SO_4^{2-}, $PM_{2.5}$ and EC.

		Subject-Specific Ratios											
		Personal-Indoor		Personal-Outdoor		Personal-Ambient		Indoor-Outdoor		Indoor-Ambient		Outdoor-Ambient	
		Med	Range	Med	Range	Med	Range	Med	Range	Med	Range	Med	Range
SO_4^{2-}	Winter	0.9	0.8–1.6	0.5	0.3–0.7	0.6	0.3–0.8	0.5	0.3–0.8	0.5	0.4–0.9	1.1	0.8–1.6
	Summer	1.0	0.9–1.2	0.8	0.5–1.0	0.9	0.4–1.1	0.8	0.4–1.0	0.8	0.3–1.3	1.0	0.8–1.1
$PM_{2.5}$	Winter	1.5	0.9–2.0	1.2	0.9–1.8	1.2	0.8–1.8	0.8	0.5–1.6	0.8	0.4–1.6	1.0	0.8–1.2
	Summer	0.9	0.6–1.1	0.9	0.6–1.6	0.9	0.6–1.2	1.0	0.6–1.4	1.0	0.6–1.3	1.0	0.8–1.3
EC	Winter	0.7	0.5–1.7	0.7	0.4–3.0	1.1	0.7–4.5	0.8	0. 5–2.1	1.5	0.7–3.1	1.7	1.3–2.2
	Summer	1.0	0.7–1.3	1.0	0.5–1.2	1.0	0.9–1.3	1.0	0.6–1.2	1.0	0.7–1.6	1.0	0.8–1.9

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Table 4

Wintertime regression model results for personal-indoor, personal-outdoor, personal-ambient, indoor-outdoor, indoor-ambient and outdoor-ambient associations.

Model	Dependent Variable	Independent Variable	Slope (95% CI)	Int (95% CI)	No. Obs	R^2
Personal-Indoor	Personal SO_4^{2-}	Indoor SO_4^{2-}	**0.85 (0.73 to 0.98)**	0.2 (0.0 to 0.4)	78	0.85
	Personal $PM_{2.5}$	Indoor $PM_{2.5}$	**0.88 (0.73 to 1.03)**	**4.4 (2.2 to 6.5)**	73	0.54
	Personal EC	Indoor EC	**0.36 (0.29 to 0.44)**	**0.7 (0.4 to 1.0)**	81	0.29[a]
Personal-Outdoor	Personal SO_4^{2-}	Outdoor SO_4^{2-}	**0.39 (0.30 to 0.49)**	**0.3 (0.1 to 0.6)**	71	0.72
	Personal $PM_{2.5}$	Outdoor $PM_{2.5}$	0.36 (−0.05 to 0.78)	**8.5 (2.3 to 14.7)**	66	0.09
	Personal EC	Outdoor EC	**0.30 (0.20 to 0.41)**	**0.7 (0.4 to 1.0)**	76	0.27[a]
Personal-Ambient	Personal SO_4^{2-}	Ambient SO_4^{2-}	**0.35 (0.27 to 0.44)**	**0.5 (0.2 to 0.7)**	73	0.71
	Personal $PM_{2.5}$	Ambient $PM_{2.5}$	**0.37 (0.08 to 0.66)**	**8.1 (4.8 to 12.4)**	72	0.17
	Personal EC[†]	Ambient EC[†]	**0.60 (0.45 to 0.75)**	**0.6 (0.4 to 0.8)**	74	0.30[a]
Indoor-Outdoor	Indoor SO_4^{2-}	Outdoor SO_4^{2-}	**0.41 (0.27 to 0.54)**	**0.3 (0.0 to 0.6)**	76	0.72
	Indoor $PM_{2.5}$	Outdoor $PM_{2.5}$	**0.53 (0.28 to 0.79)**	3.2 (−1.0 to 7.4)	77	0.30
	Indoor EC[†]	Outdoor EC[†]	**0.47 (0.34 to 0.59)**	**0.7 (0.5 to 0.9)**	81	0.28[a]
Indoor-Ambient	Indoor SO_4^{2-}	Ambient SO_4^{2-}	**0.35 (0.27 to 0.42)**	**0.5 (0.3 to 0.8)**	77	0.68
	Indoor $PM_{2.5}$	Ambient $PM_{2.5}$	**0.29 (0.12 to 0.46)**	**5.4 (2.3 to 8.6)**	84	0.17
	Indoor EC[†]	Ambient EC[†]	**0.91 (0.66 to 1.16)**	**0.7 (0.4 to 0.9)**	80	0.30[a]
Outdoor-Ambient	Outdoor SO_4^{2-}	Ambient SO_4^{2-}	**0.74 (0.60 to 0.89)**	**0.7 (0.3 to 1.2)**	75	0.73
	Outdoor $PM_{2.5}$	Ambient $PM_{2.5}$	**0.71 (0.61 to 0.82)**	**2.9 (1.6 to 4.3)**	83	0.71
	Outdoor EC	Ambient EC	**1.30 (0.84 to 1.75)**	**0.7 (0.1 to 1.3)**	75	0.39

[†]All EC models exclude one home with candle burning. Including that home, the longitudinal R^2 values increased to 0.7 for the PI model and decreased to 0.0 for the PO, PA, IO and IA models.

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Table 5

Summertime regression model results for personal-indoor, personal-outdoor, personal-ambient, indoor-outdoor, indoor-ambient and outdoor-ambient associations.

Model	Dependent Variable	Independent Variable	Slope (95% CI)	Int (95% CI)	No. Obs	R^2
Personal-Indoor	Personal SO_4^{2-}	Indoor SO_4^{2-}	**0.90 (0.85 to 0.95)**	**0.2 (0.0 to 0.4)**	98	0.93
	Personal $PM_{2.5}$	Indoor $PM_{2.5}$	**0.58 (0.35 to 0.82)**	**3.0 (0.2 to 6.1)**	79	0.49
	Personal EC	Indoor EC	**0.65 (0.40 to 0.89)**	**0.5 (0.1 to 0.8)**	97	0.40
Personal-Outdoor	Personal SO_4^{2-}	Outdoor SO_4^{2-}	**0.70 (0.60 to 0.79)**	0.2 (0.0 to 0.5)	99	0.88
	Personal $PM_{2.5}$	Outdoor $PM_{2.5}$	**0.66 (0.39 to 0.93)**	1.8 (−1.3 to 4.8)	65	0.66
	Personal EC	Outdoor EC	**0.47 (0.13 to 0.80)**	**0.7 (0.3 to 1.2)**	85	0.17
Personal-Ambient	Personal SO_4^{2-}	Ambient SO_4^{2-}	**0.72 (0.57 to 0.86)**	0.3 (−0.1 to 0.8)	91	0.74
	Personal $PM_{2.5}$	Ambient $PM_{2.5}$	**0.75 (0.46 to 1.03)**	0.6 (−2.5 to 3.7)	81	0.55
	Personal EC	Ambient EC	**0.41 (0.04 to 0.78)**	**0.9 (0.4 to 1.3)**	93	0.08
Indoor-Outdoor	Indoor SO_4^{2-}	Outdoor SO_4^{2-}	**0.74 (0.63 to 0.86)**	0.1 (−0.1 to 0.4)	102	0.89
	Indoor $PM_{2.5}$	Outdoor $PM_{2.5}$	**0.74 (0.58 to 0.89)**	**2.9 (0.9 to 5.0)**	83	0.65
	Indoor EC	Outdoor EC	**0.51 (0.22 to 0.81)**	**0.7 (0.3 to 1.1)**	86	0.23
Indoor-Ambient	Indoor SO_4^{2-}	Ambient SO_4^{2-}	**0.75 (0.59 to 0.90)**	0.3 (−0.1 to 0.6)	94	0.79
	Indoor $PM_{2.5}$	Ambient $PM_{2.5}$	**0.84 (0.61 to 1.07)**	1.7 (−0.6 to 4.0)	100	0.55
	Indoor EC	Ambient EC	0.29 (−0.02 to 0.59)	**1.1 (0.6 to 1.5)**	94	0.05
Outdoor-Ambient	Outdoor SO_4^{2-}	Ambient SO_4^{2-}	**1.08 (1.02 to 1.14)**	0.0 (−0.2 to 0.1)	95	0.91
	Outdoor $PM_{2.5}$	Ambient $PM_{2.5}$	**1.10 (0.93 to 1.28)**	−0.7 (−2.4 to 1.0)	86	0.72
	Outdoor EC	Ambient EC	0.22 (−0.03 to 0.48)	**1.2 (0.8 to 1.5)**	83	0.07

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Table 6

Personal regression model results for subset of participants with second personal measurement in the home.

Season	Pollutant	Model	Slope	95% CI	Int	95% CI	No. Obs	R^2
Winter	SO_4^{2-}	Personal SO_4^{2-} = Spouse SO_4^{2-}	**0.97**	**0.77 to 1.18**	0.1	−0.2 to 0.4	47	0.87
		Personal SO_4^{2-} = Indoor SO_4^{2-}	**0.84**	**0.68 to 1.00**	0.2	−0.1 to 0.5	50	0.80
		Personal SO_4^{2-} = Outdoor SO_4^{2-}	**0.39**	**0.26 to 0.51**	**0.4**	**0.1 to 0.7**	47	0.70
		Personal SO_4^{2-} = Ambient SO_4^{2-}	**0.39**	**0.26 to 0.52**	**0.4**	**0.1 to 0.7**	47	0.73
	$PM_{2.5}$	Personal $PM_{2.5}$ = Spouse $PM_{2.5}$	**0.51**	**0.21 to 0.82**	**7.7**	**5.4 to 9.9**	46	0.24
		Personal $PM_{2.5}$ = Indoor $PM_{2.5}$	**0.87**	**0.70 to 1.05**	**4.7**	**2.0 to 7.3**	48	0.56
		Personal $PM_{2.5}$ = Outdoor $PM_{2.5}$	0.39	−0.13 to 0.90	**9.1**	**0.1 to 18.0**	44	0.10
		Personal $PM_{2.5}$ = Ambient $PM_{2.5}$	0.34	0.00 to 0.67	**9.3**	**3.3 to 15.3**	49	0.12
	EC[a]	Personal EC= Spouse EC[†]	**0.75**	**0.57 to 0.92**	**0.5**	**0.1 to 0.8**	48	0.47
		Personal EC= Indoor EC[†]	**0.33**	**0.27 to 0.39**	**0.9**	**0.5 to 1.3**	47	0.20
		Personal EC= Outdoor EC[†]	**0.30**	**0.17 to 0.42**	**0.8**	**0.4 to 1.2**	46	0.22
		Personal EC= Ambient EC[†]	**0.67**	**0.49 to 0.85**	**0.7**	**0.3 to 1.0**	43	0.31
Summer	SO_4^{2-}	Personal SO_4^{2-} = Spouse SO_4^{2-}	**0.84**	**0.77 to 0.91**	0.5	0.0 to 0.9	37	0.96
		Personal SO_4^{2-} = Indoor SO_4^{2-}	**0.85**	**0.79 to 0.91**	0.3	−0.1 to 0.6	39	0.97
		Personal SO_4^{2-} = Outdoor SO_4^{2-}	**0.69**	**0.60 to 0.77**	0.1	−0.4 to 0.7	38	0.93
		Personal SO_4^{2-} = Ambient SO_4^{2-}	**0.74**	**0.62 to 0.86**	0.1	−0.6 to 0.7	35	0.91
	$PM_{2.5}$	Personal $PM_{2.5}$ = Spouse $PM_{2.5}$	**0.61**	**0.32 to 0.90**	**2.9**	**0.1 to 5.7**	22	0.52
		Personal $PM_{2.5}$ = Indoor $PM_{2.5}$	**0.75**	**0.43 to 1.07**	0.4	−4.8 to 5.5	31	0.77
		Personal $PM_{2.5}$ = Outdoor $PM_{2.5}$	**0.80**	**0.48 to 1.11**	−0.2	−5.3 to 4.8	28	0.81
		Personal $PM_{2.5}$ = Ambient $PM_{2.5}$	**0.89**	**0.48 to 1.30**	−1.6	−6.9 to 3.7	31	0.70
	EC	Personal EC[a]= Spouse EC	**0.44**	**0.33 to 0.55**	**0.8**	**0.6 to 1.0**	37	0.27
		Personal EC[a]= Indoor EC	**0.57**	**0.26 to 0.88**	0.6	0.0 to 1.2	38	0.44
		Personal EC[a]= Outdoor EC	0.32	−0.11 to 0.75	**0.9**	**0.2 to 1.6**	39	0.22
		Personal EC[a]= Ambient EC	0.02	−0.33 to 0.37	**1.4**	**0.7 to 2.0**	36	0.00

[†]All EC models exclude one home with candle burning. Including that home, the longitudinal R^2 values increased to 0.97 and 0.91 for the PP and PI models and decreased to 0.00 for the PO and PA models