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Determinants of Indoor and Personal Exposure to PM_{2.5} of Indoor and Outdoor Origin during the RIOPA Study

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Abstract

Effects of physical/environmental factors on fine particle (PM_{2.5}) exposure, outdoor-to-indoor transport and air exchange rate (*AER*) were examined. The fraction of ambient PM_{2.5} found indoors (*F_{INF}*) and the fraction to which people are exposed (α) modify personal exposure to ambient PM_{2.5}. Because *F_{INF}*, α , and *AER* are infrequently measured, some have used air conditioning (AC) as a modifier of ambient PM_{2.5} exposure. We found no single variable that was a good predictor of *AER*. About 50% and 40% of the variation in *F_{INF}* and α , respectively, was explained by *AER* and other activity variables. *AER* alone explained 36% and 24% of the variations in *F_{INF}* and α , respectively. Each other predictor, including Central AC Operation, accounted for less than 4% of the variation. This highlights the importance of *AER* measurements to predict *F_{INF}* and α . Evidence presented suggests that outdoor temperature and home ventilation features affect particle losses as well as *AER*, and the effects differ.

Total personal exposures to PM_{2.5} mass/species were reconstructed using personal activity and microenvironmental methods, and compared to direct personal measurement. Outdoor concentration was the dominant predictor of (partial $R^2 = 30\text{--}70\%$) and the largest contributor to (20–90%) indoor and personal exposures for PM_{2.5} mass and most species. Several activities had a dramatic impact on personal PM_{2.5} mass/species exposures for the few study participants exposed to or engaged in them, including smoking and woodworking. Incorporating personal activities (in addition to outdoor PM_{2.5}) improved the predictive power of the personal activity model for PM_{2.5} mass/species; more detailed information about personal activities and indoor sources is needed for further improvement (especially for Ca, K, OC). Adequate accounting for particle penetration and persistence indoors and for exposure to non-ambient sources could potentially increase the power of epidemiological analyses linking health effects to particulate exposures.

Keywords

fine particle exposure; PM_{2.5}; air exchange rate; particle infiltration; PM and health

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Supporting Information

The frequency distribution of activity variables (Table S1) and distribution of PM_{2.5} mass and species concentrations across each activity variable (Table S2) are provided as on-line supporting information.

1. Introduction

Epidemiologic studies report positive associations between *ambient* fine particulate matter (PM_{2.5}) and adverse health effects in metropolitan areas worldwide (U.S. EPA, 2004). In these studies, the ambient PM_{2.5} concentration is used as a surrogate for the ambient PM_{2.5} *exposure*. However, people spend more than 85% of their time *indoors* (Klepeis et al., 2001). Thus the indoor environment is a main location in which exposures to ambient PM_{2.5} occur. To account for the modification of ambient PM_{2.5} with outdoor-to-indoor transit, some epidemiologic studies use air exchange rate (*AER*), or in its absence air conditioner use, as a modifier of ambient PM_{2.5} (Franklin et al., 2007; 2008). We note below that the fraction of the ambient concentration to which people are exposed (α) and the fraction of the ambient concentration found indoors (F_{INF}) are the *key* modifiers of personal exposure to *ambient* PM_{2.5}; these factors depend, in part, on *AER*. We recognize that indoor sources and personal activities, as well as ambient PM_{2.5}, contribute to *total* PM_{2.5} exposure. Herein we examine the main determinants of *AER*, α and F_{INF} , and the effects of activities and other exposure factors on indoor PM_{2.5} and *total* personal exposure. A better understanding of the drivers of *AER*, F_{INF} and α variability could help refine epidemiological study design (U.S. EPA, 2004) and aid the development of effective strategies to mitigate PM_{2.5} exposures.

The steady state mass balance equations,

$$C_i = \frac{Pa}{a+k} C_a + C_{na} = F_{INF} C_a + C_{na} = C_a + C_{na} \quad (1)$$

$$E_i = \{y + (1 - y)F_{INF}\} C_a + E_{na} = \alpha C_a + E_{na} = E_a + E_{na} \quad (2)$$

have been widely used to separate indoor PM_{2.5} (C_i , $\mu\text{g}/\text{m}^3$) and total personal PM_{2.5} exposure (E_i , $\mu\text{g}/\text{m}^3$) into two components: PM_{2.5} of ambient (or outdoor) origin (C_a in Equation 1; E_a in Equation 2, $\mu\text{g}/\text{m}^3$), and PM_{2.5} of nonambient origin (C_{na} in Equation 1; E_{na} in Equation 2, $\mu\text{g}/\text{m}^3$). The ambient component of indoor PM_{2.5} is a product of the ambient concentration (C_a , $\mu\text{g}/\text{m}^3$) and F_{INF} (dimensionless), which is a function of the penetration coefficient (P , dimensionless), air exchange rate (a , h^{-1}), and particle loss rate (k , h^{-1}). Personal exposure to ambient PM_{2.5} is a product of the ambient exposure factor (α , a.k.a. attenuation factor, dimensionless) and ambient concentration. The ambient exposure factor is a function of F_{INF} and the fraction of time a person spends outdoors (y , dimensionless).

New methods have been proposed to estimate α and F_{INF} based on measured indoor, outdoor, and personal PM mass and species concentrations (U.S. EPA, 2004; Hopke et al., 2003; Meng et al., 2005; Wilson and Brauer, 2006; Strand et al., 2007), or based on mechanisms governing PM penetration (Hering et al., 2007). However, the influence of environmental factors (e.g., temperature) and housing factors (e.g., house age, ventilation) on *AER*, α , and F_{INF} is poorly characterized.

Personal exposure to non-ambient PM_{2.5} (e.g., from indoor sources and personal activities) can also complicate the interpretation of air pollution studies and reduce the power of epidemiological findings (U.S. EPA, 2004). Personal activity information from questionnaires has been used in regression and analysis of variance models to describe sources and activities impacting exposure and indoor air quality (Koistinen et al., 2001; Lai et al., 2006; Lanki et al., 2007; Baxter et al., 2007). However, the multivariate nature of personal activities and use of this knowledge to reconstruct PM_{2.5} exposures requires further elucidation. How well can

PM_{2.5} exposure be predicted by personal activities? How important are various activities to indoor PM_{2.5} concentrations and personal exposures? Three methods frequently employed to quantify personal PM_{2.5} exposure are: 1) direct personal exposure measurements, 2) measured microenvironmental concentrations and time spent in each microenvironment, and 3) personal activity information. It is not feasible to measure personal exposures directly for a large population. It is also burdensome to apply the microenvironmental method to estimate personal exposures for a large population. Thus the personal activity method could prove more practical as a method for estimating population exposure distributions.

In this work, the effects of home characteristics and environmental factors on AER , F_{INF} and α are examined. Also, models are employed to predict indoor and total personal exposure to PM_{2.5} using personal activity information and using a microenvironmental method. This work reflects our continuing effort to understand 1) indoor and personal exposure to ambient and non-ambient PM_{2.5} and 2) exposure modifiers (e.g., Naumova et al., 2003; Meng et al., 2005; 2007; Polidori et al., 2006).

2. Methods

2.1 Sampling and Chemical Analysis

The Relationship of Indoor, Outdoor and Personal Air (RIOPA) study is documented in detail elsewhere (Weisel et al., 2005; Turpin et al., 2007). Briefly, as part of the RIOPA study (1999 summer – 2001 spring), 48-h indoor, outdoor and personal PM_{2.5} samples were collected in 374 non-smoking homes in Houston (TX), Los Angeles County (CA), and Elizabeth (NJ). Samples were analyzed for mass (gravimetric), and a subset of the samples (279 homes) were analyzed for 36 elements (XRF) and organic and elemental carbon (OC and EC, indoor and outdoor samples only; thermal-optical transmittance with adsorption artifact correction). The 48-h average indoor and outdoor temperature and AER for each home were also measured. AER was determined from house volume and the concentration of an inert non-toxic tracer released at a constant rate during sampling (Dietz et al., 1986). The maximum measurable AER was approximately 5 air changes/hr, and method precision (expressed as coefficient of variation) was 18%, based on 79 pairs of collocated samples.

2.2 RIOPA Questionnaires

Four questionnaires were developed based on the National Human Exposure Assessment Survey (NHEXAS; Sexton et al., 1995): the Baseline Questionnaire, Activity Questionnaire, Technician Walk-Through Questionnaire, and Activity Diary. Questions related to household characteristics, participant demographics and socioeconomic information were included in the Baseline Questionnaire and answered by study participants. The Technician Walk-Through Questionnaire documented the independent observations of study personnel with respect to the characteristics of the participants' household and neighborhood. The Activity Questionnaire collected information regarding when and where participants spent time and what they did during the sampling period. The Activity Diary was a 30-min resolution activity log listing the time a participant spent in each location or microenvironment.

In this analysis, questions were examined for their relevance to indoor PM_{2.5} emission, home ventilation, particle loss, particle penetration, and the fraction of time a person spent in each microenvironment. Thirty-two questions from the Activity Questionnaire and three questions from the Baseline Questionnaire were selected for use.

2.3 Estimation of F_{INF} and α

In previous work, a microscopic mixture model was used to calculate F_{INF} for each RIOPA home based on the measured indoor and outdoor PM_{2.5} elements, OC, and EC (Meng et al.,

2005; Turpin et al., 2007). Briefly, a least-trimmed squared regression (S-plus, Insightful, Inc.) was used to regress the indoor $PM_{2.5}$ species concentrations on the outdoor $PM_{2.5}$ species concentrations collected concurrently at a single home, yielding a $PM_{2.5} F_{INF}$ (slope) for each of 114 RIOPA homes. The F_{INF} mean and standard deviation were 0.69 and 0.23 respectively, across homes. Individual F_{INF} values had standard errors of 0.0002 – 0.066. The microscopic mixture model assumes steady state and that indoor and outdoor sources are independent. This model allows for sample-to-sample variations (across homes and days) in AER , particle penetration, and particle loss rate that can occur due to variations in parameters such as house structure, air conditioner use, ventilation practice, particle size distribution, particle composition, and the thermodynamic stability of particle species. The least-trimmed squared regression was used to estimate F_{INF} for each home because it is very robust with respect to outliers, and outliers occur whenever there are substantial indoor sources.

The ambient exposure factor (α) for each subject was calculated (Equation 2) using F_{INF} estimated above and the fraction of time a person spent in different microenvironments, as recorded in the Activity Diary Questionnaire. The effects of ventilation conditions and temperatures on F_{INF} , α and AER were then examined.

2.4 Analysis of Housing, Environmental Factors and Activities

The central tendency of the AER , F_{INF} , and α under different household and ventilation conditions was examined using nonparametric statistical techniques, such as Wilcoxon two-sample test and Kruskal-Wallis tests. The relationships between ambient temperature and AER , F_{INF} , and α were characterized by a nonparametric regression method, LOESS (Local Polynomial Regression Fitting), which provides great flexibility and does not assume a parametric form for the regression surface. The smoothing parameter was determined based on the optimization of the bias-corrected Akaike information criterion, and the robustness of the nonparametric fit was improved by iterative reweighting.

2.5 Reconstruction of Total Personal Exposures

Personal exposures were reconstructed with two methods: 1) microenvironmental method and 2) personal activity method. The resulting personal exposure estimates were compared with direct personal exposure measurements using Wilcoxon two-sample tests.

The microenvironmental method is given by:

$$E_t = \sum_j C_j t_j \quad (3)$$

where C_j is the $PM_{2.5}$ concentration in the j^{th} microenvironment, and t_j is the time a person spends in the j^{th} microenvironment. In this work, the microenvironmental method predicted personal exposure to $PM_{2.5}$ and associated species using residential indoor and outdoor concentrations, time spent in each microenvironment, and Equation 3.

The personal activity method predicted indoor and personal exposures using multiple linear regression (MLR) of activity variables with stepwise predictor selection, adjusting for corresponding outdoor concentrations. The 27 dichotomous activity variables used in the analysis (Table S1) are related to PM source emissions. These variables document that a certain activity/event either happened (Yes, coded as 1 in the regression model) or didn't happen (No, coded as 0) during the 48-h sampling period. Variables were first selected based on their relevance and frequency distribution across the two levels (Yes/No) of each activity variable

(Table S2). If the frequency of an activity on either level was less than 1, that variable was removed from the input even if it was relevant to PM_{2.5} exposure.

The collinearity between activity variables was then tested and outliers were identified. Questionnaire-based personal activity information has a multivariate nature due to personal/population living patterns and the design of the questionnaire (e.g. the same information might be asked multiple times in different ways). The variance inflation factor was used to identify collinearities between activity variables. No more than one of any group of highly correlated variables was retained in any final stepwise regression model. The final variance inflation factors for all variables were close to one, indicating that the issue of collinearity was addressed adequately. The detection of outliers was based on influential diagnostics (the DFFITS statistic) in the regular least square regression; outlier detection by least trimmed square regression was computationally prohibitive in this case. An outlier was defined and removed from the final input if the DFFITS value was greater than $2\sqrt{k/N}$, where k is the number of regression coefficients to be determined and N is the number of observations.

Finally, MLR with stepwise selection was conducted based on the “clean” input dataset (without outliers and collinear variables) to characterize the effects of personal activities on indoor and personal PM_{2.5} exposure. Input variables (Table S1) differ slightly for different PM species because the subset of PM samples analyzed for elements and OC/EC were not entirely identical, leading to slightly different collinearity and sample size issues. The PM species selected are those typically associated with PM sources or formation mechanisms: Ca (soil dust), K (soil dust and biomass combustion), S (secondary PM from coal and diesel combustion), V (oil combustion), OC (both primary and secondary PM) and EC (combustion) (Hopke, 1985). Nitrate was not measured.

Statistical analyses were conducted with the Statistical Analysis System (SAS version 9.1, SAS Inc, Cary, NC). The significance level was 0.05 except where specified, and mathematical transformation of data (i.e. logarithmization) was made whenever necessary to reduce the skewness of the original data and more closely meet the assumptions of the statistical models. Homes sampled a second time (~3 mo later) were treated as independent samples.

3. Results and Discussion

3.1 Effects of Home Ventilation and House Type on F_{INF} , α , and AER

As defined in Equation 1, F_{INF} is a function of three physical parameters: P , k and AER . The relationship among F_{INF} , P , k and AER (a) can be recast as

$$\frac{1}{F_{INF}} = \frac{K}{P} \cdot \left(\frac{1}{a}\right) + \frac{1}{P} \quad (4)$$

where $(1/P)$ and (k/P) can be regarded as the intercept and the slope, respectively, in the context of a regression analysis. A least trimmed square robust regression was conducted to examine the association between $1/F_{INF}$ and $1/a$ for the 114 RIOPA homes with home-specific F_{INF} (Meng et al., 2005). A positive association (P-value < 0.001) between $1/F_{INF}$ and $1/a$ was seen (Figure 1), and the population average P (0.83) and k (0.06 h^{-1}), derived from the regression intercept and slope, are similar to what we found previously for the overall RIOPA study (Turpin et al, 2007). If all homes shared the same penetration coefficient and particle loss rate, a perfect linear relationship between $1/F_{INF}$ and $1/a$ would have been observed, and AER would be an outstanding surrogate for F_{INF} . Differences in P and k across homes might contribute to the observed scatter in Figure 1 ($r^2 = 0.18$). Physically, P and k are not independent (Lunden et al., 2003). We expect that P and k are affected by factors such as home ventilation status

(e.g., Central AC Operation), type/age of construction (e.g. Building Age), particle size distribution, and the thermodynamic properties of the aerosol because these factors affect PM losses by impaction, diffusion, sedimentation and evaporation. For these reasons, below we examine the effects of several factors on the magnitude and variability of F_{INF} , α , and AER (Table 1–2).

Central AC Operation was the only factor significantly associated (negatively) with all three parameters (F_{INF} , α , and AER ; Wilcoxon two-sample tests P-value < 0.05) (Table 1). Mean F_{INF} , α , and AER were 0.57, 0.68, and 0.7 h⁻¹, respectively, for homes where a central air conditioning (AC) system was in operation; and 0.73, 0.79, and 1.3 h⁻¹, respectively, for homes where it was not. The lower F_{INF} and α for homes using central AC (compared to those that were not) could be driven by the lower AER s for these homes.

Unlike Central AC Operation, the factors Window Fan Operation, Ceiling Fan Operation, and Central AC (existence of a system) were significantly (P-value < 0.05) or marginally significantly (0.05 < P-value < 0.10) associated with changes in F_{INF} or α , but not AER . For example, Window Fan Operation was significantly associated with changes in α (P-value = 0.0069) and marginally associated with changes in F_{INF} (P-value = 0.10). The mean α was 0.85 vs. 0.75 and mean F_{INF} was 0.76 vs. 0.68 for using vs. not using a window fan, respectively, whereas the increase in AER was not significant (1.5 h⁻¹ for using vs. 1.1 h⁻¹ for not using a window fan). In contrast, Window AC Operation was a marginally significant factor associated with AER change (1.9 h⁻¹ for using vs. 1.0 h⁻¹ for not using a window AC); however, using window AC did not significantly alter F_{INF} or α .

This observation (see also Figure 1) is consistent with the expectation that exposure factors can alter F_{INF} or α not only by affecting AER but also by changing particle loss rates indoors (k) and/or during outdoor-to-indoor transport (P) (Bearg, 1993). In part this occurs because particle loss processes are complex functions of the air flow patterns (stagnant vs. turbulent), and the use of some ventilation units (e.g. ceiling fans) change the air flow pattern in a room. Therefore, the use of a ventilation unit is expected to change P , k and AER . It is important to recognize this when house type/age, home ventilation status, or AER is used as a surrogate for F_{INF} or α to classify personal exposure to ambient PM_{2.5}. Note sample sizes for some categories are small (e.g. N = 3 for using attic fan, Table 1); findings for these factors are less robust.

Outdoor temperature also affected F_{INF} , α , and AER (Figure 2). F_{INF} (and α) reaches its maximum when the outdoor temperature is approximately 20 °C, decreasing at higher and lower temperature (Figure 2b,2c). In contrast, little association was observed between AER and outdoor temperature (Figure 2a). Relationships with outdoor temperature (Figure 2a–c) were characterized by LOESS, a nonparametric regression method. LOESS was used because no parametric form of the regression surface is known, although Lai et al. (2006) found a significant association between outdoor temperature and indoor PM_{2.5} (ln-transformed) in EXPOLIS. Long et al. (2001) observed a seasonal effect of hourly F_{INF} in nine Boston residential homes, with higher F_{INF} in the summer (> 0.7) and lower in the winter (68% of hourly F_{INF} were less than 0.7).

Differences in the response of F_{INF} , α , and AER to temperature changes might be explained as follows. When the ambient temperature departs from the thermal comfort temperature, people tend to close their windows and use either cooling or heating equipment. As a result of heating or cooling, the absolute indoor-outdoor temperature difference increases, driving additional airflow. Closing the windows decreases AER (wind effect) and increases particle losses; while at the same time the increased temperature gradient increases AER (temperature effect), resulting in greater effects of temperature on P and k (and therefore F_{INF}) than on AER .

Least trimmed square regressions were applied to examine the effect of absolute indoor-outdoor temperature differences on F_{INF} (Figure 2e) and α (Figure 2f). Correlations were weak; slopes were negative and not significant at the level of 0.05. The association between AER and the absolute temperature difference was positive but not significant (Figure 2d). In contrast, Wallace et al. (2002) and Howard-Reed (2002) reported a strong linear association between the within-home AER (70 min resolution) and the absolute indoor-outdoor temperature difference ($R^2 = 0.46$, $N=543$). The differences in findings between these studies can, in part, be explained by differences in study designs. This study examined associations across homes in three distinct climates, whereas the other examined within home associations longitudinally.

To examine how much variation in F_{INF} , α , and AER can be explained by the observed or measured variables, a multivariate approach (stepwise MLR) was used to predict F_{INF} , α , and AER based on home ventilation status, building age/type, measured AER and outdoor temperature (Table 2). For F_{INF} and α , AER was the dominant predictor, accounting for 35.6% and 24.1% of variations in F_{INF} and α , respectively. Each of the other predictors accounted for less than 4% of the overall variation. The overall modeled coefficients of determination (R^2) are 49% for F_{INF} and 41% for α , respectively. For AER , the only significant predictor was Central AC Operation, explaining 6.9% of the overall variation. Thus, no simple ventilation variable was identified that was an adequate surrogate for AER . Substantially better F_{INF} prediction was achieved, with AER as the main predictor. This highlights the importance of AER measurements.

3.2 Effects of Activity Patterns on Indoor $PM_{2.5}$ and Personal Exposure

Indoor and personal $PM_{2.5}$ mass and species predictions from stepwise MLR of personal activity variables and outdoor concentrations are presented in Table 3–Table 4. The regression coefficients (and P-values), partial R^2 for each predictor, and the overall model R^2 from the final least square fit are given. Regression coefficients for activity variables are in concentration units ($\mu\text{g}/\text{m}^3$ for mass, OC, EC, and S; ng/m^3 for Ca, K, and V). They represent the increase in the PM mass or species concentration when a certain activity occurs. For example, the regression coefficient for “Incense” is $1.6 \mu\text{g}/\text{m}^3$ for indoor $PM_{2.5}$ mass and $26.8 \mu\text{g}/\text{m}^3$ for K (Table 3, $N=58$), suggesting that burning incense indoors increases the 48-h average indoor $PM_{2.5}$ mass and K by 1.6 and $26.8 \mu\text{g}/\text{m}^3$ on average, respectively, when it happens. Also presented (Table 3–4) are the median percentage contributions of outdoor $PM_{2.5}$ and each activity to the measured $PM_{2.5}$ mass/species concentrations for the study population. The median contribution of an activity to the population is determined by the product of the activity-related source strength (the regression coefficient) and the frequency distribution of the activity across the population (Bernoulli distribution for dichotomous variable; 0 when the activity didn’t occur; 1 when it did; Table S2). Even if an activity generated substantial emissions, it was not a substantial contributor to population exposure unless it was also a frequently occurring activity.

Among all predictors, outdoor concentration dominates the contribution to $PM_{2.5}$ mass and species (except for Ca), with median percentage contributions of 18.5% to 100% (Table 3–4). In addition, in all cases, the overall R^2 in the MLR models are larger than the partial R^2 for outdoor concentration (as if outdoor concentration were the only predictor), indicating that the inclusion of personal activities increases the predictive power of the model. For indoor OC, Ca, K and personal Ca, K and mass, the overall model R^2 increases by a factor of two by including personal activities as predictors, compared to the partial R^2 for outdoor concentration alone (Table 3–4). For S, V, and EC, outdoor concentration is the predominant predictor. The overall model R^2 is high for S, V, EC (dominated by outdoor sources), but still low for Ca, K, and OC (which have substantial indoor sources) despite the added predictive power of personal activities. The low R^2 for these species might occur if: 1) these species are generated by

activities that are not adequately documented in the questionnaires; 2) indoor activity-related emissions or losses are highly variable, or 3) outdoor soil dust particles are a substantial contributor to these species and their infiltration is highly variable. (Dust particles are in the coarse tail of PM_{2.5} and have low infiltration factors; Meng et al., 2007.)

Environmental tobacco smoke (ETS) has long been known to be the most important indoor particle source (Wallace, 1996). Although RIOPA subjects were non-smokers, ETS emissions were reported in a few (N=5) homes. For these homes, the median indoor PM_{2.5} (23.2 µg/m³) was almost double that of homes without ETS emissions (14.2 µg/m³) (Table S2), consistent with EXPOLIS (Lai et al., 2006).

Home cleaning processes (labeled “Sweeping”) primarily contributed to PM_{2.5} mass and dust related species (Ca, K) (Table 3–4). They also contributed to indoor OC (15%). The association of cleaning with OC might occur because OC is a constituent of house dust (Hopke et al., 2003), and/or because it is formed from the oxidation of household cleaning products (Weschler, 2001). Housework such as vacuuming has previously been linked to soil dust tracers (Ca, K) (Baxter et al., 2007). Lanki et al (2007) estimated that indoor cleaning activities increased 24-h average personal PM_{2.5} mass exposures by 2 µg/m³. Baxter et al. (2007) estimated that they increased indoor Ca and K by 3.28 and 11.8 ng/m³, respectively, when averaged over a week. The Baxter et al. (2007) finding for the effects of indoor cleaning on Ca is comparable to our results: in RIOPA, cleaning increased indoor Ca by 20.9 ng/m³ over the 48-h sampling period, which is equivalent to ~ 6.0 ng/m³ averaged over a week (assuming cleaning once per week).

Indoor heating and combustion activities (Oil furnace, Oven, Fireplace, and Incense) are associated with increased indoor and personal exposure to EC, K, S and/or V (Table 3–4). This is not surprising, since these species have been used as combustion tracers in the past. Note that Baxter et al. (2007) found an association between EC and candle burning. Cooking could be another important indoor combustion source (Wallace, 1996), although results from recent studies are mixed. No significant effects of cooking on indoor PM_{2.5} mass were reported in this study or EXPOLIS (Helsinki; Koistinen et al., 2001). In contrast, Lanki et al (2007) and Baxter et al. (2007) reported that cooking was a significant contributor to indoor PM_{2.5} (Amsterdam, Helsinki, and Boston). The differences might result from variations in cooking style, cooking time, frequency and kitchen/home design.

Travel, Cooking_outside, Woodworking, Sander, and Chainsaw contributed to increases in Ca, K and/or V. Neither time spent outdoors nor Travel affected personal PM_{2.5} exposure in EXPOLIS (Koistinen et al., 2001), but in RIOPA Travel was significantly associated with personal Ca exposure. Chainsaw and Sander use were associated with increased personal V exposure, and Outdoor Cooking with increased Ca, K and V. Ca is released from meat cooking and is a soil dust component (Watson and Chow, 2001). Note these activities were rare and therefore contributed little to the median exposure.

Exposures reconstructed using the microenvironmental method (Equation 3) were more highly correlated with direct personal exposure measurements (Figure 3) than were exposures predicted with the personal activity method (Table 4), suggesting that personal exposure variations are better captured by the microenvironmental model. The coefficients of determination (R²) for PM_{2.5} mass, S, Ca, K, and V were 28.4%, 86.2%, 26.2%, 30.5%, and 79.3%, respectively, for the microenvironmental model; they were 8.5%, 75.4%, 9.9%, 11.0% and 75.6%, respectively, for the personal activity model. For ambient generated species such as S and V, no significant differences were observed between measured (personal) and modeled (microenvironmental method) exposure concentrations according to Wilcoxon two-sample tests. In contrast, for species with strong local and indoor sources, the microenvironmental

model underestimated population mean exposures by 20 $\mu\text{g}/\text{m}^3$, 87 ng/m^3 , and 346 ng/m^3 (for $\text{PM}_{2.5}$ mass, K and Ca, respectively; P -value < 0.001). For the personal activity model, no significant differences were observed between the modeled and measured exposures since measured personal exposures were used in the model (dependent variables) to “calibrate” the contributions from personal activities.

4. Discussion and Conclusions

In this work, the ability to predict exposure to *ambient* and to *total* $\text{PM}_{2.5}$ and associated species was examined. Specifically, we examined drivers of exposure to $\text{PM}_{2.5}$ and associated species including personal activities, microenvironmental concentrations, and outdoor-to-indoor transport.

First we explored the effects of physical and environmental factors on 1) the main modifiers of exposure (F_{INF} and α), and 2) on air AER . No single variable was a good predictor of AER . Substantially better prediction of F_{INF} and α was achieved, with AER as the dominant factor (explaining 36% and 24% of the variation in F_{INF} and α , respectively). This highlights the importance of AER measurements.

There was a considerable difference in the response of F_{INF} and of AER to changes in temperature. F_{INF} was largest for ambient temperatures near 20 °C and decreases at higher and lower temperatures, whereas AER showed little sensitivity to outdoor temperature. Central AC operation was the only factor with a statistically significant effect on F_{INF} , α , and AER . Other home ventilation and building features seem to affect AER differently than they affect F_{INF} and α . These observations suggest that temperature, ventilation features, and building age/type alter particle loss processes (P , k) as well as AER . Note P and k are also physical determinants of F_{INF} and α (Equations 1,2). This is important to recognize when using a ventilation variable as an exposure modifier in PM epidemiology.

The contributions of personal activities to *total* $\text{PM}_{2.5}$ exposures were investigated using the personal activity model. Outdoor concentration was the dominant predictor of indoor concentration and personal exposure for all examined species (except Ca). Incorporation of personal activities as predictors improved the predictive power of the personal activity model, despite the fact that these activities were incorporated as binary variables. Several non-ambient PM sources were identified that were rarely encountered by RIOPA participants but dramatically increased the $\text{PM}_{2.5}$ mass/species exposures for the participants that were exposed. Most notably, engaging in woodworking and exposure to tobacco smoke increased $\text{PM}_{2.5}$ exposures by approximately 25 and 10 $\mu\text{g}/\text{m}^3$ (averaged over 48-h) respectively, when these exposures occurred. Engaging in woodworking, when it occurred, also had a large impact on K exposure (>100 ng/m^3). Several personal activities were identified that can substantially increase exposure to Ca and K. Effective control for (or accounting of) non-ambient sources in predictive exposure models could enhance the ability of epidemiologic studies to explore associations between health endpoints and exposure to individual particle-phase species.

Total personal exposure was reconstructed using personal activity and microenvironmental methods. The microenvironmental model better captured exposure variability than the personal activity method, suggesting that inter-personal variations in exposure features such as indoor source strength and exposure duration can be well captured by microenvironmental models that make use of both indoor and outdoor measurements. However, in this work the microenvironmental model did not accurately capture the *magnitude* of exposure to $\text{PM}_{2.5}$ species with large personal activity source contributions. One limitation of this study is that concentrations were only measured in two microenvironments (indoor and outdoor), and exposures in other microenvironments (e.g. office) were not represented.

The performance of the personal activity model relies on activity information collected using a questionnaire. The biggest limitation of the personal activity model is that it does not account for variations in the duration, composition, or magnitude of emissions associated with a documented activity. The personal activity model provides a single average source strength (the regression coefficient). It is possible that a more sophisticated treatment of activities and indoor sources, for example coupling highly time-resolved measurements and activity data, could substantially improve prediction for species with considerable activity-related sources. More information about sources of Ca, K, and OC is especially warranted.

RIOPA was a pooled study where many homes were sampled 1–2 times to maximize the variability in important exposure drivers. If this study instead had a longitudinal design (few people sampled many times), we would expect that a greater portion of the exposure variability would have been explained because personal activities and indoor sources vary less within subjects than between subjects and the effects of home construction on P , k and AER would be de-emphasized.

This paper highlights the importance of AER , F_{INF} , α , and PM exposure prediction with the goal of inspiring further advancements that will aid exposure mitigation and PM epidemiology. Because people spend most of their time indoors and the fraction of ambient PM that penetrates and persists in the indoor environment (F_{INF}) varies, we expect that adequately accounting for the penetration and persistence of ambient particles into indoor spaces will reduce exposure error and bias leading to narrower confidence intervals, better model fits and perhaps larger risk estimates in epidemiologic studies of *ambient* PM. Also, if a *particular* PM species were responsible for PM toxicity, exposure error and bias would be reduced if epidemiologic studies examined *total* (ambient plus non-ambient) exposure to that species. Exposure assessment is also needed to develop effective exposure mitigation strategies.

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Figure 1.

The relationship between the fraction of ambient $PM_{2.5}$ found indoors (F_{INF}) and air exchange rate (a) for the 114 RIOPA homes with home-specific $PM_{2.5}$ F_{INF} values. The regression line and the equation on the plot are the results of least trimmed square regression.

Figure 2.

The relationship between outdoor temperature and air exchange rate (a), fraction of ambient $\text{PM}_{2.5}$ found indoors, F_{INF} (b), and alpha (ambient exposure factor) (c), a LOESS smooth curve (solid line) and corresponding upper and lower 95% confidence levels (dash lines) are also shown on (a), (b), and (c); the relationship between the absolute difference of indoor and outdoor temperature and air exchange rate (d), infiltration factor (e) and alpha (f), the lines and the equations on (d), (e) and (f) are the results of the least trimmed square regression.

Figure 3.

Estimated personal exposure concentrations using a microenvironmental model and direct personal exposure measurements for PM_{2.5} mass (a) and Sulfur (b). The solid line is the least trimmed square robust regression line.

Table 1

Distribution of air exchange rate (AER), fraction of ambient $PM_{2.5}$ found indoors (F_{INF}) and fraction of ambient $PM_{2.5}$ to which people are exposed (α) by ventilation status and building characteristics

Ventilation Status and Household Characteristics				AER, h ⁻¹			F _{INF}			α				
Category	Status ^c	N	Mean	Median	Standard Deviation	P-value	Mean	Median	Standard Deviation	P-value	Mean	Median	Standard Deviation	P-value
Central AC Operation ^a	Y	28	0.7	0.6	0.5	0.0019	0.57	0.60	0.23	0.0013	0.68	0.70	0.16	0.0008
	N	84	1.3	0.9	1.0		0.73	0.74	0.22		0.79	0.83	0.17	
Window AC Operation	Y	13	1.9	1.6	1.5	0.053	0.76	0.70	0.22	0.44	0.79	0.75	0.18	0.68
	N	97	1.0	0.8	0.8		0.68	0.70	0.23		0.76	0.76	0.18	
Window Fan Operation	Y	12	1.5	1.0	1.3	0.43	0.76	0.82	0.28	0.1	0.85	0.98	0.21	0.0069
	N	99	1.1	0.8	0.9		0.68	0.68	0.22		0.75	0.75	0.17	
Ceiling Fan Operation	Y	45	1.2	0.9	1.0	0.79	0.65	0.66	0.27	0.1	0.73	0.73	0.20	0.14
	N	67	1.1	0.8	0.9		0.72	0.73	0.19		0.78	0.80	0.16	
Attic Fan Operation	Y	3	1.9	0.6	2.2	0.72	0.71	0.59	0.25	0.84	0.77	0.65	0.20	0.83
	N	109	1.1	0.8	0.9		0.69	0.70	0.23		0.76	0.76	0.18	
Building Age	< 1944	15	1.3	1.1	1.0	0.3	0.76	0.79	0.19	0.37	0.79	0.81	0.15	0.47
	1945 – 1959	25	1.4	0.9	0.9		0.75	0.71	0.16		0.81	0.81	0.12	
	1960 – 1975	12	1.0	0.9	1.0		0.64	0.61	0.28		0.71	0.69	0.19	
	1976 – 1984	16	0.8	0.7	0.5		0.71	0.76	0.22		0.77	0.85	0.18	
	> 1985	10	1.2	0.5	1.4		0.65	0.63	0.26		0.76	0.73	0.21	
Building Type	Apartment	26	1.1	0.9	0.9	0.83	0.79	0.8	0.22	0.08	0.81	0.86	0.19	0.20
	Single House	51	1.1	0.8	0.8		0.69	0.68	0.22		0.76	0.74	0.17	
	Town House	4	1.1	1.0	0.4		0.67	0.73	0.23		0.77	0.78	0.10	
	Mobile House	14	1.3	0.5	1.3		0.64	0.59	0.25		0.73	0.70	0.20	
Central AC ^b	With	32	0.8	0.8	0.5	0.23	0.63	0.66	0.2	0.03	0.72	0.72	0.16	0.05
	Without	69	1.2	0.8	1.0		0.73	0.74	0.23		0.78	0.78	0.18	

^aCentral AC Operation: Air conditioning system operation status

^bCentral AC: Building with or without a central air conditioning system installed

^cStatus: if a ventilation unit was used during the 48-h sampling time, “Y” is marked; otherwise, it is “N”.

Table 2

Using ventilation variables and outdoor temperature to predict air exchange rate (*AER*), *F_{INF}*, and α in a stepwise multilinear regression

Dependent Variables	Regression Parameters	Central AC Operation	Window AC Operation	Window Fan Operation	Ceiling Fan Operation	Attic Fan Operation	Air Exchange Rate	Outdoor Temperature	Intercept	Modeled R ²
α	Coefficient	-0.08	-0.10	0.12	-0.08	-0.13	0.08	0.01	0.66	41%
	P-value	0.004	0.004	0.002	0.002	0.081	<0.001	0.008	<0.001	
	Partial R ²	1.9%	3.3%	2.6%	3.8%	1.5%	24.1%	3.6%		
<i>F_{INF}</i>	Coefficient	-0.13	-0.10	0.07	-0.06	-0.15	0.12	0.01	0.50	49%
	P-value	<0.001	0.015	0.105	0.035	0.077	<0.001	0.001	<0.001	
	Partial R ²	3.5%	2.7%	1.2%	1.3%	1.2%	35.6%	3.2%		
<i>AER</i>	Coefficient	-0.40					N.A. ^a		1.1	6.9%
	P-value	0.004					N.A.		<0.001	
	Partial R ²	6.9%					N.A.			

^aN.A.: Not Applicable

Table 3

Predicted indoor PM_{2.5} mass and species by stepwise multilinear regression

Species	Significant Predictors ^a	Outdoor	Tobacco_home	Sweeping	Incense	Sander	Chainsaw	Cooking_inside	Diesel_parked	Intercept	Model R ^{2d}
Mass	Estimate ^b	0.5	10.3	2.2	1.6				1.3	4.9	34.9%
(N = 240)	P-value	<0.001	0.12	0.012	0.142				0.138	<0.001	
	Partial R ²	32.2%	0.6%	2.0%	0.6%				0.9%		
OC	Median										
	%Contribution ^c	52.8%	0.0%	5.5%	0.0%				0.0%		
(N = 133)	Estimate	0.4		1.7					1.2	3.2	9.0%
	P-value	0.009		0.012					0.069	<0.001	
	Partial R ²	4.2%		4.6%					2.3%		
EC	Median										
	%Contribution	21.7%		15.0%					0.0%		
(N = 168)	Estimate	0.8	0.3	0.1	0.2					26.8	75.1%
	P-value	<0.001	0.093	0.073	0.038					0.696	
	Partial R ²	74.3%	0.4%	0.5%	0.5%						
S	Median										
	%Contribution	91.6%	0.0%	6.9%	0.0%						
(N = 177)	Estimate	0.8		-0.07				0.06		-0.05	78.5%
	P-value	<0.001		0.075				0.127		0.226	
	Partial R ²	78.2%		0.3%				0.3%			
Ca	Median										
	%Contribution	104.0%		-3.0%				0.0%			
(N = 191)	Estimate			20.9		95.2				71	7.8%
	P-value			0.022		<0.001				<0.001	
	Partial R ²			2.6%		6.2%					
K	Median										
	%Contribution			8.7%		0.0%					
(N = 184)	Estimate	0.3			26.8				16.6	29.2	18.7%
	P-value	<0.001			<0.001				0.007	<0.001	
	Partial R ²	10.9%			5.8%				3.3%		
	Median										
	%Contribution	36.0%			0.0%				0.0%		

Species	Significant Predictors ^a									Model R ^{2d}
	Outdoor	Tobacco_home	Sweeping	Incense	Sander	Chainsaw	Cooking_inside	Diesel_parked	Intercept	
V	0.6					1.2			0	80.3%
(N = 174)	<0.001					0.021			0.822	
Partial R ²	80.0%					0.6%				
Median %Contribution	95.7%					0.0%				

^aExcept intercept, all regression coefficient presented are significant at 0.15 level, which is the default option in SAS.

^bNumbers are regression coefficients, in units of $\mu\text{g}/\text{m}^3$ for PM mass, OC, EC and S, and in units of ng/m^3 for others.

^cThe median of the percentage contribution of the significant factor to indoor PM mass or species across all homes.

^dAdjusted R² for the final stepwise multiple linear regression model.

Table 4
Predicted personal exposure to PM_{2.5} mass and species by stepwise multilinear regression

Species	Significant Predictors ^a	Outdoor	Sweeping	Oil_ furnace	Oven	Fire place	Incense	Travel	Cooking_ outside	Wood working	Sander	Chain saw	Diesel_ parked	Intercept	Model R ^{2d}
Mass (N = 214)	Estimate ^b	0.4	6.4							25				22.6	8.5%
	P-value	0.001	0.002							0.095				<0.001	
	Partial R ²	4.1%	4.4%							1.2%					
	Median %Contribution ^c	18.5%	7.8%							0.0%					
S (N = 190)	Estimate ^b	0.6				0.9	0.08			0.3				0.04	75.4%
	P-value	<0.001				<0.001	0.042			0.118				0.301	
	Partial R ²	72.9%				2.2%	0.5%			0.3%					
	Median %Contribution ^c	88.2%				0.0%	0.0%			0.0%					
Ca (N = 197)	Estimate ^b	0.2	115.8					91.6	160.1				62.6	116.1	9.9%
	P-value	0.054	0.003					0.05	0.011				0.107	0.029	
	Partial R ²	1.6%	4.4%					1.5%	3.6%				1.2%		
	Median %Contribution ^c	7.1%	9.7%					27.4%	0.0%				0.0%		
K (N = 190)	Estimate ^b	0.3	28.6				24.7		35.2	134.4				83.3	11.0%
	P-value	0.003	0.012				0.075		0.074	0.088				<0.001	
	Partial R ²	5.6%	3.6%				1.4%		1.4%	1.4%					
	Median %Contribution ^c	19.2%	6.3%				0.0%		0.0%	0.0%					
V (N = 190)	Estimate ^b	0.5		1.4	1.5	7.4			0.7		1.9	1.2		0.1	75.6%
	P-value	<0.001		0.035	0.025	<0.001			0.039		0.044	0.01		0.485	
	Partial R ²	69.7%		0.5%	0.6%	3.7%			0.6%		0.7%	0.8%			
	Median %Contribution ^c	80.1%		0.0%	0.0%	0.0%			0.0%		0.0%	0.0%			

^aExcept intercept, all regression coefficient presented are significant at 0.15 level, which is the default option in SAS.

^bNumbers are regression coefficients, in units of $\mu\text{g}/\text{m}^3$ for PM mass, OC, EC and S, and in units of ng/m^3 for others.

^c The median of the percentage contribution of the significant factor to indoor PM mass or species across all homes.

^d Adjusted R^2 for the final stepwise multiple linear regression model.