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Toward refined estimates of ambient PM_{2.5} exposure: Evaluation of a physical outdoor-to-indoor transport model

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Abstract

Because people spend the majority of their time indoors, the variable efficiency with which ambient PM_{2.5} penetrates and persists indoors is a source of error in epidemiologic studies that use PM_{2.5} concentrations measured at central-site monitors as surrogates for ambient PM_{2.5} exposure. To reduce this error, practical methods to model indoor concentrations of ambient PM_{2.5} are needed. Toward this goal, we evaluated and refined an outdoor-to-indoor transport model using measured indoor and outdoor PM_{2.5} species concentrations and air exchange rates from the Relationships of Indoor, Outdoor, and Personal Air Study. Herein, we present model evaluation results, discuss what data are most critical to prediction of residential exposures at the individual-subject and populations levels, and make recommendations for the application of the model in epidemiologic studies. This paper demonstrates that not accounting for certain human activities (air conditioning and heating use, opening windows) leads to bias in predicted residential PM_{2.5} exposures at the individual-subject level, but not the population level. The analyses presented also provide quantitative evidence that shifts in the gas-particle partitioning of ambient organics with outdoor-to-indoor transport contribute significantly to variability in indoor ambient organic carbon concentrations and suggest that methods to account for these shifts will further improve the accuracy of outdoor-to-indoor transport models.

Keywords

PM_{2.5} exposure; organic aerosol; gas-particle partitioning; Relationships of Indoor; Outdoor and Personal Air (RIOPA) study; Aerosol Penetration and Persistence (APP) model

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1. Introduction

While people spend the majority of time indoors (Klepeis et al., 2001), fine particulate matter (PM_{2.5}) concentrations measured at outdoor central-site monitors are commonly used as surrogates for exposure to PM_{2.5} of outdoor (ambient) origin in epidemiological studies. The use of central-site PM_{2.5} concentrations as ambient PM_{2.5} exposure surrogates inherently assumes that indoor and outdoor ambient PM_{2.5} concentrations are highly correlated. However, the fraction of ambient PM_{2.5} that penetrates and persists indoors (F) varies with multiple factors including meteorological conditions, the physical and chemical properties of ambient PM_{2.5}, housing characteristics, and home ventilation conditions (Riley et al., 2002; Nazaroff, 2004; Sarnat et al., 2006; Hering et al., 2007; Meng et al., 2009; Chen and Zhao 2011; Chen et al. 2012; Meng et al., 2012). Exposure error associated with not accounting for variability in F generally contributes to an underestimation of health effects associated with ambient PM_{2.5} exposures (Zeger et al., 2000).

In order to reduce this exposure error, practical methods to predict indoor concentrations of ambient PM_{2.5} are needed. Toward this goal, we evaluated and refined a physical mass-balance model using measurements from the Relationships of Indoor, Outdoor, and Personal Air (RIOPA) study (Weisel et al., 2005; Turpin et al., 2007). An earlier version of the model was applied in two epidemiologic studies: one that explored associations between ambient PM_{2.5} exposures and myocardial infarction (MI) and the other, associations with birth outcomes (Turpin et al., 2012; Baxter et al., 2013; Hodas et al., 2013). The work herein provides a partial validation of the exposure estimates used in those studies, while also providing new insights that are used to refine the model. This paper highlights the measurements and data most critically needed to facilitate the prediction of residential ambient PM_{2.5} exposures in epidemiological studies.

2. Methods

2.1 Modeled Indoor PM_{2.5} Concentrations

Indoor concentrations of ambient particulate sulfate, elemental carbon (EC), and organic carbon (OC) were calculated for RIOPA homes (Table S1) with a mass-balance model. The model describes the concentration of chemically non-reactive PM_{2.5} species j in indoor air ($C_{in,j}$) as a function of its outdoor concentration ($C_{out,j}$), residential air exchange rate (AER), particle penetration efficiency (P_j), and the depositional loss rate of species j in indoor air ($k_{dep,j}$; Hering et al., 2007):

$$dC_{in,j}/dt = C_{out,j} (P_j \times AER) - C_{in,j} (k_{dep,j} + AER) \quad (1)$$

Forty-eight hour average outdoor sulfate, EC, and OC concentrations and AERs measured at each RIOPA home (Table S2) were used as model inputs (nitrate was not measured during RIOPA). Details regarding RIOPA study measurements are provided in supplementary material. Briefly, AERs were measured with a perfluorocarbon tracer method (Yamamoto et al., 2010). PM_{2.5} filter samples were analyzed for EC and OC (µgC/m³) with a Sunset carbon analyzer and for sulfur by energy-dispersive XRF spectrometry and expressed as sulfate (Weisel et al., 2005; Turpin et al., 2007). OC was corrected for the adsorption artifact

by subtracting the organic mass on the backup filter (Turpin et al., 2007). Due to the long averaging time of RIOPA measurements, indoor concentrations were calculated with the time-averaged solution to equation 1:

$$C_{in,j} = C_{out,j} \left(\frac{AER \times P}{AER + k_{dep,j}} \right) \quad (2)$$

A review of published species size distributions from diverse geographic locations and seasons (supplementary material, Table S3) was conducted to identify “typical” size distributions (i.e., number of modes, mass median diameter of each mode, and the fraction of mass in each mode) for sulfate, EC, and OC. Values of k_{dep} were then selected for the mass median diameter of each size mode of each PM_{2.5} species size distribution (Table 1) using the fourth-order polynomial fit to measured particle-size-resolved deposition rates from Nazaroff, (2004). While this method provides a means to estimate reasonable values of k_{dep} , the reader should be aware that factors in addition to particle size can contribute to variability in k_{dep} (e.g. particle density, room airflow conditions; Lai and Nazaroff, 2000; Nazaroff, 2004) and there is heterogeneity in measured size-resolved particle deposition rates across studies (Nazaroff, 2004). A constant P of 0.8, the median value reported by Chen and Zhao (2011) for particles in the size range considered here, was used for all species. Like k_{dep} , many factors contribute to variability in P . For example, laboratory studies have demonstrated that the geometry and roughness of cracks in a building shell can contribute to variability in P (Liu and Nazaroff, 2001; Nazaroff, 2004; Chen and Zhao, 2011); however, these cracks have not been well characterized for individual homes and are likely to be highly variable (Nazaroff, 2004). As a result, this variability is not accounted for in our calculations. In subsequent sections, we explore other contributors to variability in P such as particle size and home ventilation conditions.

2.2 Model Evaluation

We compared modeled indoor concentrations of *ambient* sulfate, EC, and OC with the measured indoor concentrations of these PM_{2.5} species (supplementary material, Table S2) for each (occupied) RIOPA home. In epidemiologic analyses, the extent to which a model is successful in predicting exposures at the individual-subject level is described by the covariance between actual and estimated exposures. As a result, we examined correlations between measured and modeled indoor concentrations. Paired t-tests were also conducted to evaluate whether pairs of measured and modeled indoor PM_{2.5} species concentrations were significantly different at the 95% confidence level. To assess model performance at the population level, chi-square tests were used to examine whether cumulative distributions of measured and modeled indoor concentrations had the same underlying distribution at a 95% confidence level. All analyses were conducted with SAS software (version 9.3; SAS Institute Inc., Cary, NC).

Using the same methods, we also evaluated whether measured residential *outdoor* PM_{2.5} concentrations were good predictors of indoor ambient PM_{2.5} concentrations. Much of the recent work aimed at refining ambient PM_{2.5} exposure surrogates has focused on accounting for spatial variability in *outdoor* PM_{2.5} concentrations (e.g. land use regression, interpolation

methods; Jerrett et al., 2005; Hoek et al., 2008). Ambient $PM_{2.5}$ concentrations measured outside of RIOPA homes provide spatially-resolved measures of outdoor $PM_{2.5}$ concentrations. A comparison between measured residential *outdoor* $PM_{2.5}$ concentrations and modeled indoor ambient $PM_{2.5}$ concentrations evaluates whether exposure metrics that account for outdoor-to-indoor transport offer improvement over exposure metrics that account only for spatial variability in outdoor concentrations.

2.3 Attributing model-measurement differences

2.3.1 Human activities—To focus our efforts to refine the outdoor-to-indoor transport model, we explored the contributions of several factors to differences between modeled and measured indoor $PM_{2.5}$ species concentrations. First, we evaluated the extent to which model-measurement differences could be attributed to the fact that the model does not account for the effects of human activities likely to influence F . Human-activity variables that were likely to influence the efficiency with which ambient $PM_{2.5}$ penetrated and/or persisted in RIOPA homes were selected from questionnaires administered to RIOPA participants to characterize home-occupant activities during sample collection (supplementary material; Weisel et al., 2005; Meng et al., 2009). The activities were: (1) time with windows open, (2) time with central air conditioning (AC) in use, and (3) time with central heating in use. Differences between modeled and measured indoor $PM_{2.5}$ concentrations were regressed on these activity variables using multiple linear regression (MLR) with stepwise selection ($\alpha = 0.15$ for variable entrance and removal threshold; SAS version 9.3). Variance inflation factors indicated that the human activities were not significantly correlated with each other. Outliers were detected based on the student's t and a value was considered an outlier if t was greater than 2. It should be noted that outliers are likely indicators of strong indoor sources. Outlier homes were excluded from all following analyses to avoid influence of strong indoor sources of $PM_{2.5}$ on model evaluation results, as the model predicts only the contribution of *ambient* $PM_{2.5}$ to indoor concentrations and not the contribution of indoor sources.

We refined the model to account for the human-activity variables selected as significant predictors of model-measurement differences based on assumptions about the ways in which each activity variable would influence F . For homes with open windows, there is little to no removal of the particles entering the home and, thus, we assumed a penetration efficiency (P_{window}) of 1.0 (Table 1; Chen and Zhao, 2011). For homes with central AC or heating in use, a filter penetration efficiency term (P_{filter}) was multiplied by the right side of equation (2) to account for losses in the filters of central heating and cooling systems. Values of P_{filter} were selected from particle-size-resolved filtration efficiencies for residential furnace filters (assuming a pressure drop of 125 Pa across the filter to account for particle loading; Hanley et al., 1994) using the same assumptions about species size distributions as were used to select k_{dep} values (Table 1). If more than one activity occurred within a home, we accounted only for the dominant activity (i.e. the activity carried out for the longer period of time). The performance of this refined version of the model was evaluated using the same methods as described above.

2.3.2 Indoor sources of OC—Because sulfate and EC are non-volatile and have minimal indoor sources, indoor concentrations of these species are likely driven by outdoor-to-indoor transport (Sarnat et al., 2006). However, organics comprised the majority of PM_{2.5} emitted or formed inside RIOPA homes (on average, 41 – 76%; Polidori et al., 2006). Because the aim of the current modeling is to predict indoor concentrations of *ambient* PM_{2.5}, there is a need for an estimate of the measured indoor OC that can be attributed to outdoor sources. We estimated this by regressing measured indoor OC concentrations on measured outdoor OC concentrations using robust regression (SAS version 9.3). Robust regression down-weights outliers and, thus, reduces the influence of strong indoor sources on the regression equation (Ott et al., 2000; Meng et al., 2005). The intercept of the resulting regression equation provides an average indoor-source strength and the slope is a population-average estimate of F . We multiplied this population-average F by each measured outdoor OC concentration to calculate the distribution of measured indoor OC of ambient origin (Ott et al., 2000; Meng et al., 2005). When this approach was used for sulfate, which is dominated by outdoor sources, F estimated by robust regression was in good agreement with F calculated as the ratio of measured indoor to measured outdoor sulfate (supplementary material, Figure S1).

2.3.3 Uncertainty in OC size distributions—Ambient OC size distributions are more variable across sampling locations and seasons than sulfate and EC. We conducted a sensitivity analysis to explore whether uncertainty in ambient outdoor OC size distributions (and, thus, k_{dep}) was a source of error in modeled indoor ambient OC. Indoor ambient OC concentrations were calculated assuming two alternative size distributions: (1) a bimodal distribution with an ultrafine peak at 0.08 μm and a broad accumulation mode peaking at 0.4 μm with 20% and 80% of OC mass comprising each mode, respectively ($k_{dep} = 0.07 \text{ h}^{-1}$; Turpin et al., 1997; Miguel et al., 2004) and (2) a trimodal distribution with an ultrafine peak (comprising 20% of OC mass) and an accumulation mode comprised of a condensation mode (0.2 μm) and droplet mode (0.7 μm) of equal mass proportions. The k_{dep} values for alternative size distribution (2) are the same as those shown in Table 1, but the mass fractions comprising the condensation and droplet modes are different from those explored in the main analysis. Indoor OC concentrations calculated assuming each of the three size distributions were compared to evaluate the sensitivity of the model to uncertainty in k_{dep} associated with variability in OC size distributions.

2.3.4 Phase changes of ambient organics—Predicting the outdoor-to-indoor transport of particulate OC is further complicated by the fact that organics can undergo phase changes due to indoor-outdoor differences in temperature, surface area, and the availability of particulate matter for sorption (Naumova et al., 2003; Polidori et al., 2006; Lunden et al., 2008; Shi and Zhao, 2012). Because ambient OC is comprised of thousands of compounds with largely unknown identities (Goldstein and Galbally, 2007), it is not possible to calculate the change in gas-particle partitioning with outdoor-to-indoor transport from first principles. In order to explore the influence of phase changes on F , a surrogate is needed. We used 5 - 7 ring polycyclic aromatic hydrocarbons (PAHs), which were measured in the gas and particle phases inside and outside of 76 RIOPA homes (Naumova et al., 2002), for this purpose. The PAHs included were benzo[b+k]fluoranthene, benzo[e]pyrene,

benzo[a]pyrene, perylene, indeno[1,2,3-c,d]pyrene, dibenzo[a,c+g,h]anthracene, benzo[g,h,i]perylene, and coronene. These PAHs are predominantly of outdoor origin and were mainly in the particle phase under the ambient conditions measured outside RIOPA homes (Naumova et al., 2002), making them a useful surrogate for ambient particulate OC.

Using MLR, we explored the extent to which variability in measured indoor OC concentrations could be explained by (1) physical losses associated with outdoor-to-indoor transport (i.e., those already accounted for in the model) and (2) shifts in the gas-particle partitioning of ambient organics with indoor transport (using changes in partitioning of 5-7 ring PAHs). We regressed measured indoor OC on modeled indoor OC and on the indoor-outdoor difference in the pooled gas-particle partitioning coefficient (K_p) of the 5 - 7 ring PAHs ($\alpha = 0.15$ for variable entrance and removal threshold; SAS version 9.3). K_p was calculated as the ratio of the pooled concentration of PAHs in the particle phase to their concentration in the gas phase, normalized by the total $PM_{2.5}$ concentration (Pankow, 1994). No collinearity between variables was found and outliers were removed using the same criteria as described above.

3. Results and Discussion

3.1 Initial Model

Agreement between modeled indoor ambient $PM_{2.5}$ concentrations and measured indoor concentrations varied by species. While modeled indoor ambient EC concentrations were well correlated with measured indoor EC ($R^2 = 0.70$), the model generally underestimated indoor EC (Figure 1a, 2a). In fact, pairs of measured and modeled indoor EC were significantly different and modeled and measured values did not have the same underlying distribution at a 95% confidence level, suggesting that model refinements are needed to predict residential EC exposures.

For sulfate, the initial model performed reasonably well at the population level, but the model under-predicted indoor sulfate for many high concentration homes (Figure 2b). Cumulative distributions of measured and modeled indoor sulfate (Figure 1b) had the same underlying distribution ($P = 0.87$). While modeled and measured values were well correlated ($R^2 = 0.86$; Figure 2b), modeled indoor sulfate concentrations were significantly lower than measured concentrations at a 95% confidence level according to a paired t-test. Thus, the initial model could be applied to estimate residential sulfate exposures at the population level, but refinements are needed to improve exposure estimates at the individual-subject level, particularly for high-end exposures.

Measured indoor OC concentrations were not well captured by the initial model (Figure 1c, 2c), which accounts for physical losses of ambient OC during outdoor-to-indoor transport into closed homes without air conditioning, but does not account for phase changes or indoor sources. Modeled indoor particulate OC of ambient origin explained only 4% of the variability in total particulate OC measured indoors ($R^2 = 0.04$). Further, measured and modeled indoor OC concentrations were significantly different at a 95% confidence level at both the individual-(paired) and population (distribution) levels. Contributors to this poor agreement are explored below.

3.2 Model refinement: accounting for human activities

Human activities that were not accounted for in the initial model helped to explain differences between modeled and measured indoor sulfate (Table 2), but not EC and OC. For sulfate, all activity variables included in the MLR analysis were selected as significant predictors of model-measurement differences ($P < 0.15$) and together explained 31% of the variance in these differences (Table 2). We refined the model to account for these activities based on our assumptions regarding the ways in which each activity would influence F (Table 1).

Improved agreement between measured and modeled indoor sulfate at the individual-subject level was substantial (Figure 3a). Pairs of measured and modeled indoor sulfate concentrations were no longer significantly different ($P = 0.60$). Indoor sulfate concentrations modeled with the refined model explained 90% of the variance in measured indoor sulfate, compared to 86% for the initial model. While use of the refined model also improved agreement between measured and modeled indoor sulfate distributions ($P = 0.996$; Figure 3c), the initial model distribution was not significantly different from the measured distribution to begin with. Thus, while the initial model is adequate for predicting sulfate distributions, we recommend the use of the refined model when estimating residential sulfate exposures at the individual-subject level. Notably, most epidemiologic studies do not focus on sulfate exposures, but rather on exposure to total ambient $PM_{2.5}$. When using a mass-balance model like the one explored here, exposure to total ambient $PM_{2.5}$ would be calculated by summing predicted indoor concentrations of the individual species. The fact that a refined version of the model is needed to predict residential sulfate exposures has implications for the design of epidemiologic studies focused on both $PM_{2.5}$ species and total ambient $PM_{2.5}$ exposures, as it requires the collection of human activity data (e.g., pertaining to windows, air conditioning, and heating) using questionnaires or activity diaries or a method to estimate human activity patterns (e.g. sampling from a distribution of published time-activity patterns; Zhou and Zhao, 2012) over the length of the study.

While accounting for human activities in the model improved model-measurement agreement for sulfate, the examined human activities had a minimal impact on F for ambient EC. The small impact of human activities can likely be explained by the EC size distribution. Values of P_{filter} for 80 nm particles are ~90% (Hanley et al., 1994) and, thus, use of central AC or heating is expected to result in only small losses of EC. This also suggests that for the ultrafine-mode, overall penetration efficiencies may be greater than the 0.8 used in the initial calculations. We re-calculated indoor ambient EC assuming a P value of 0.9. With this refinement, the model captured indoor EC concentrations at both the population and individual-subject levels (Figure 3b, 3d). Measured and modeled EC had the same underlying distribution ($P = 0.65$), they were well correlated ($R^2 = 0.70$), and pairs of measured and modeled indoor EC were not significantly different at a 95% confidence level ($P = 0.16$). These results suggest that human activities might not need to be accounted for when calculating residential EC exposures, but that P values can vary across $PM_{2.5}$ species due to differences in size distributions.

Like EC, human activities were not selected as significant predictors of differences between measured and modeled indoor OC. Based on the assumed size distribution for OC, we would expect that the influence of human activities on F would be minimal for OC in the ultrafine mode, but similar to that for sulfate for the accumulation mode fraction. However, the effect of human activities was likely overshadowed by the substantial contribution of indoor emissions (Polidori et al., 2006) to measured indoor OC.

3.3 Accounting for indoor sources of OC

The robust-regression estimated F value for OC was 0.53, suggesting that, on average, 53% of the ambient OC penetrated and persisted indoors (Figure S2). This value is higher than the F value for OC reported in Polidori et al. (2006), which estimated contributions of outdoor-generated OC to total OC measured in RIOPA homes using a Random Component Superposition (RCS) statistical model ($F = 0.32$). Figure 4 compares cumulative distributions of ambient indoor OC estimated with the mass-balance model with our robust-regression estimate of F (a comparison with the RCS-estimated F value is available in Figure S3). Agreement between the distributions of indoor ambient OC estimated with the model and with robust regression (Figure 4; mean $\pm$ standard deviation = 2.54 ± 1.61 $\mu\text{gC}/\text{m}^3$ and 1.91 ± 1.26 $\mu\text{gC}/\text{m}^3$, respectively) is improved compared to agreement between modeled indoor ambient OC and measured (total) indoor OC (6.08 ± 3.77 $\mu\text{gC}/\text{m}^3$). However, the two distributions are still significantly different ($P = 0.0004$). Accounting for human activities and the higher penetration efficiency of ultrafine-mode particles ($P = 0.9$) in calculations of indoor ambient OC did not reduce this bias (mean $\pm$ standard deviation 2.74 ± 1.81 $\mu\text{gC}/\text{m}^3$; Figure S3).

3.4 Variability and uncertainty in OC size distributions

The model showed little sensitivity to the uncertainty in k_{dep} associated with variability in OC size distributions. Estimated indoor OC concentrations were highly correlated across the size-distributions scenarios ($R^2 > 0.99$) and distributions of modeled indoor OC were in good agreement (mean $\pm$ standard deviation = 2.54 ± 1.61 , 2.62 ± 1.66 , and 2.58 ± 1.64 $\mu\text{gC}/\text{m}^3$ for the initial and two alternative size-distribution scenarios, respectively). The OC size distributions considered here are based on measurements conducted in urban regions in which OC is comprised of a mix of locally- and regionally-generated PM (supplementary material). It is possible that these size distributions are not representative of the OC measured outside of some of the RIOPA study homes. For example, for homes in close proximity to primary $\text{PM}_{2.5}$ sources (as is the case for many RIOPA homes), the majority of OC might be in the ultrafine mode, which would result in smaller depositional losses (and possibly greater P values) than those calculated assuming that accumulation-mode OC comprised a substantial fraction of the OC. It should be noted, however, that this would result in increased calculated indoor ambient OC concentrations and the model already has an upward bias (Figure 4).

3.5 Shifts in the gas-particle partitioning of ambient OC

We did find evidence that shifts in the gas-particle partitioning of ambient OC with outdoor-to-indoor transport contributed to variability in particulate OC measured inside RIOPA

homes. The model-estimated indoor OC, which was included in the MLR analysis to represent physical particle losses associated with outdoor-to-indoor transport, was the most significant predictor of variability in measured indoor OC concentrations ($P = 0.0003$), explaining 20% of this variability ($R^2 = 0.20$). The indoor-outdoor difference in K_p for the 5 - 7 ring PAHs, which we used as a surrogate for changes in the gas-particle partitioning of ambient OC, was also selected as significant predictor of variability in indoor OC ($P = 0.05$), explaining 5% of this variability ($R^2 = 0.05$). While PAHs account for only a small fraction of total OC, this new finding for OC is consistent with previous work which demonstrated that shifts in the gas-particle partitioning of outdoor-generated PAHs with outdoor-to-indoor transport contributes substantially to variability in residential PAH exposures (Shi and Zhao, 2012; Zhou and Zhao, 2012). Much of the remaining variability can likely be attributed to indoor OC sources, which contributed to 3 - 99% of the OC in these RIOPA homes (calculated by subtracting the robust-regression estimate of indoor ambient OC from total measured indoor OC).

While physical loss was the dominant contributor to variability in indoor ambient OC, our results suggest that refining the model to account for phase changes of OC with outdoor-to-indoor transport would improve the predictive abilities of the model. This is an important area of future work that requires further characterization of the thermodynamic properties of ambient OC and a better understanding of the chemistry that occurs in indoor air, including interactions between indoor- and outdoor-emitted organics (Weschler, 2011).

3.6 Further recommendations for epidemiologic studies

Exposure research has focused on accounting for spatial variability in outdoor air pollution concentrations (e.g., use of residential outdoor concentrations rather than central-site concentrations through land use regression, interpolation between sites, etc.; Jerrett et al., 2005; Hoek et al., 2008). One objective of this study was to explore whether a model that brings the residential outdoor air pollution indoors, offers additional improvement. The mass-balance model did offer improvement over the use of measured outdoor concentrations as residential ambient $PM_{2.5}$ exposure surrogates. As noted above, EC and sulfate have minimal indoor sources (Sarnat et al., 2006) and, thus, the vast majority of the sulfate and EC measured inside RIOPA homes can be attributed to $PM_{2.5}$ of outdoor origin. As expected, measured outdoor sulfate and EC concentrations were well-correlated with measured indoor concentrations ($R^2 = 0.77$ and 0.69 , respectively), but correlations between modeled and measured indoor concentrations are even stronger ($R^2 = 0.86$ and 0.90 for the initial and refined sulfate models and $R^2 = 0.70$ for EC for both models). Measured outdoor and indoor OC were weakly correlated ($R^2 = 0.03$), undoubtedly because of the substantial contributions of indoor sources to indoor OC concentrations. The mass-balance model offered only a small improvement over measured outdoor OC concentrations ($R^2 = 0.004$). However, when the influence of indoor sources was reduced using the robust-regression estimate of F , indoor OC concentrations calculated with the mass-balance model performed better than measured outdoor OC concentrations (mean $\pm$ standard deviation = 1.91 ± 1.26 , 2.54 ± 1.61 , and $3.61 \pm 2.38 \mu\text{gC}/\text{m}^3$ for the robust-regression estimate of indoor ambient OC, modeled indoor ambient OC, and measured outdoor OC, respectively). Notably, in the two epidemiologic studies discussed above (in which a version of this mass-balance model

was used to estimate the fraction of *central-site* PM_{2.5} found in study-subject homes) the spatial resolution of residential ambient PM_{2.5} exposure estimates was identified as a possible source of error (Hodas et al., 2013). A two-step approach involving a method to account for local-scale variability in outdoor PM_{2.5} followed by the use of an outdoor-to-indoor transport model might offer the best results when predicting residential PM_{2.5} exposures.

It should also be noted that AERs measured at each individual home were used as model inputs in our calculations; however, these data are not generally available for an epidemiologic study population. The Lawrence Berkeley National Laboratory Infiltration model, which accounts for air exchange due to air flow through cracks in a residence (Sherman and Grimsrud, 1980) has recently been refined to include natural ventilation through open windows (Breen et al., 2010; Turpin et al., 2012) and can be used to calculate AER distributions for a study population using readily-available housing data (from the United States Census and the American Housing Survey) and routinely measured meteorological parameters. A refined version of the LBNL Infiltration model was used for this purpose in the two epidemiologic studies mentioned above (Baxter et al, 2013; Hodas et al., 2013).

4. Conclusions

The evaluation and refinement of an outdoor-to-indoor transport model using measured indoor and outdoor PM_{2.5} species concentrations and AERs from the RIOPA study illustrates that the modeling tools presented here offer improvement over the use of outdoor PM_{2.5} concentrations to estimate residential ambient PM_{2.5} exposure. The level of model refinement and data required to facilitate the use of this model in large epidemiologic studies varies across PM_{2.5} species. Accounting for AC and heating use and open windows led to reduced bias in predicted *F* values for sulfate at the individual-subject level, but this refinement was not needed for EC nor at the population level for sulfate. This refinement did not resolve the large model-measurement differences for OC. We did, however, find quantitative evidence that shifts in the gas-particle partitioning of ambient organics with outdoor-to-indoor transport contribute significantly to variability in *F*. Our results suggest that the collection of human activity data or a method to predict these human activity patterns can lead to substantial improvements in individual-subject level residential ambient PM_{2.5} exposure estimates. This work also highlights the need for a method to account for shifts in the gas-particle partitioning of ambient OC in outdoor-to-indoor transport models. While further refinements are recommended, this mass-balance model is a practical method that can be applied in large epidemiologic studies to predict residential ambient PM_{2.5} exposures. The input parameters (i.e. k_{dep} , P_{filter}) provided here are based on a comprehensive assessment of PM_{2.5} species size distributions and their evaluation using RIOPA data provides confidence in this version of the mass-balance model as a robust tool for reducing exposure misclassification in epidemiologic studies.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Abbreviations

F	Fraction of ambient PM _{2.5} that penetrates and persists indoors
AER	Residential air exchange rate
P	Particle penetration efficiency
k_{dep}	Indoor depositional loss rate coefficient
P_{window}	Particle penetration efficiency for homes with open windows
P_{filter}	Central air conditioning or heating filter penetration efficiency

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- Indoor ambient PM_{2.5} species concentrations were modeled for RIOPA study homes
- Modeled PM_{2.5} concentrations were evaluated against measured indoor concentrations
- Accounting for human activity reduces bias in modeled residential PM_{2.5} exposures
- Phase changes with indoor transport influence ambient organic carbon exposures

Figure 1.

Cumulative distributions: measured indoor species (solid) and indoor species of ambient origin modeled with the initial model (dashed): (a) elemental carbon (EC), (b) sulfate, and (c) organic carbon (OC).

Figure 2. Indoor $PM_{2.5}$ species concentrations ($\mu\text{g}/\text{m}^3$): modeled with the initial model and measured (a) elemental carbon (EC), (b) sulfate, and (c) organic carbon (OC). The dashed line is the 1:1 line. Note the model predicts indoor concentrations of ambient origin, whereas measurements also include the contribution from indoor sources.

Figure 3. Indoor $PM_{2.5}$ species concentrations ($\mu g/m^3$): modeled with the refined model and measured (a) sulfate and (b) elemental carbon (EC). The dashed line is the 1:1 line. Cumulative distributions: measured indoor species (solid) and indoor species of ambient origin modeled with the refined model (dashed): (c) sulfate and (d) elemental carbon (EC). Note that for sulfate the refined model accounts for human activities, while for EC it accounts for the greater penetration efficiency of ultrafine-mode particles.

Figure 4. Cumulative distributions of measured indoor organic carbon (OC) concentrations (solid), indoor OC of ambient origin estimated with the mass-balance model (dashed), and indoor OC of ambient origin estimated with the population average F value calculated using robust regression (dotted).

Table 1

Ambient PM_{2.5} species particle diameters and associated particle deposition loss rate coefficients (k_{dep}), penetration efficiencies (P), central heating and air conditioning filter penetration efficiencies (P_{filter}), and penetration efficiencies for homes with open windows (P_{window}).

	Elemental Carbon	Sulfate		Organic Carbon		
		Mode 1	Mode 2	Mode 1	Mode 2	Mode 3
Mass Fraction in Mode	1.0	0.2	0.8	0.4	0.12	0.48
Particle Diameter (μm)	0.08	0.2	0.7	0.08	0.2	0.7
k_{dep} (h^{-1})	0.05	0.05	0.13	0.05	0.05	0.13
P	0.80/0.90 ^b	0.80	0.80	0.80/0.90 ^b	0.80	0.80
P_{filter}^a	0.90	0.90	0.65	0.90	0.90	0.65
P_{window}^a	1.0	1.0	1.0	1.0	1.0	1.0

^a Refined model: activities selected as significant predictors of variability in model-measurement differences were included in the refined model.

^b Refined model: greater penetration efficiency of ultrafine-mode particles was accounted for in the refined model

Table 2

Multiple linear regression (MLR) analysis investigating the contribution of human activities to variability in model-measurement differences for sulfate.

Sulfate (n = 203)				
Selection Step	Activity	Partial R²	Model R²	P
1	Central Air Conditioning	0.20	0.20	< 0.0001
2	Open Windows	0.09	0.29	< 0.0001
3	Central Heating	0.02	0.31	0.027

Partial R² describes the variance in model-measurement differences explained by each human-activity variable individually. Model R² describes the total variance in these differences described by the full MLR model at each selection step. Indoor sulfate of outdoor origin is modeled. Measurements are of total indoor sulfate. Previous work suggests indoor sulfate is predominately of outdoor origin (Sarnat et al., 2006).