A Population Exposure Model for Particulate Matter: Case Study Results for $PM_{2.5}$ in Philadelphia, PA

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Disclaimer

The research described has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

Acronyms

CHAD = Consolidated Human Activity Database
EPA = Environmental Protection Agency
ETS = Environmental tobacco smoke
HVAC = Heating, ventilation, and air conditioning
PM = Particulate matter
MAACS = Metropolitan Acid Aerosol Characterization Study
NAAQS = National Ambient Air Quality Standards
NHAPS = National Human Activity Pattern Survey
PMSA = Primary metropolitan statistical area
RECS = Residential Energy Consumption Survey
SHEDS = Stochastic Human Exposure and Dose Simulation
TEOM = Tapered element oscillating microbalance

Abstract

A population exposure model for particulate matter (PM), called the Stochastic Human Exposure and Dose Simulation (SHEDS-PM) model, has been developed and applied in a case study of daily $PM_{2.5}$ exposures for the population living in Philadelphia, PA. SHEDS-PM is a probabilistic model that estimates the population distribution of total PM exposures by randomly sampling from various input distributions. A mass-balance equation is used to calculate indoor PM concentrations for the residential microenvironment from ambient outdoor PM concentrations and physical factor data (e.g., air exchange, penetration, deposition), as well as emission strengths for indoor PM sources (e.g., smoking, cooking). PM concentrations in non-residential microenvironments are calculated using equations developed from regression analysis of available indoor and outdoor measurement data for vehicles, offices, schools, stores and restaurants/bars. Additional model inputs include demographic data for the population being modeled and human activity pattern data from EPA's Consolidated Human Activity Database (CHAD). Model outputs include distributions of daily total PM exposures in various microenvironments (indoors, in vehicles, outdoors), and the contribution from PM of ambient origin to daily total PM exposures in these microenvironments.

SHEDS-PM has been applied to the population of Philadelphia using spatially and temporally interpolated ambient $PM_{2.5}$ measurements from 1992-93 and 1990 U.S. Census data for each census tract in Philadelphia. The resulting distributions showed substantial variability in daily total $PM_{2.5}$ exposures for the population of Philadelphia (median=20 $\mu g/m^3$; 90th percentile=59 $\mu g/m^3$). Variability in human activities, and the presence of indoor residential sources in particular, contributed to the observed variability in total $PM_{2.5}$ exposures. The uncertainty in the estimated population distribution for total $PM_{2.5}$ exposures was highest at the

upper end of the distribution and revealed the importance of including estimates of input uncertainty in population exposure models. The distributions of daily microenvironmental $PM_{2.5}$ exposures (exposures due to time spent in various microenvironments) indicated that indoor residential $PM_{2.5}$ exposures (median=13 $\mu g/m^3$) had the greatest influence on total $PM_{2.5}$ exposures compared to the other microenvironments.

The distribution of daily exposures to $PM_{2.5}$ of ambient origin was less variable across the population than the distribution of daily total $PM_{2.5}$ exposures (median=7 $\mu g/m^3$; 90th percentile=18 $\mu g/m^3$) and similar to the distribution of ambient outdoor $PM_{2.5}$ concentrations. This result suggests that human activity patterns did not have as strong an influence on ambient $PM_{2.5}$ exposures as was observed for exposure to other $PM_{2.5}$ sources. For most of the simulated population, exposure to $PM_{2.5}$ of ambient origin contributed a significant percent of the daily total $PM_{2.5}$ exposures (median=37.5%), especially for the segment of the population without exposure to environmental tobacco smoke in the residence (median=46.4%).

Development of the SHEDS-PM model using the Philadelphia $PM_{2.5}$ case study also provided useful insights into the limitations of currently available data for use in population exposure models. In addition, data needs for improving inputs to the SHEDS-PM model, reducing uncertainty and further refinement of the model structure were identified.

Introduction

Air pollution epidemiology studies have found statistically significant associations between particulate matter (PM) concentrations and acute and chronic health outcomes (Pope et al., 1995; Samet et al., 1995; Dockery and Pope, 1994; Schwartz, 1994). These studies typically use air pollution measurements from stationary air monitoring sites within a community as a surrogate for personal exposure levels for the population. To better understand the relationship between personal exposures and stationary community measurements, human exposure field studies have been conducted that measure both community PM concentrations and personal PM exposures. These PM exposure field studies have included population-based studies such as PTEAM and EXPOLIS (Clayton et al, 1993; Jantunen et al., 1998) and panel studies that target a subset of the population, such as the elderly (Ebelt et al., 2000; Evans et al., 2000; Janssen et al., 2000; Sarnat et al., 2000; Williams et al., 2000a).

Both types of studies have shown the impact of human activities on individual PM exposures (Howard-Reed et al., 2000; Oglesby et al., 2000; Sarnat et al., 2000; Williams et al, 2000b; _zkaynak et al., 1996). Personal PM exposure measurements were influenced by ambient outdoor PM as measured by a stationary community monitor, as well as other sources of PM located in indoor microenvironments. The observed relationship between personal exposure and community monitor levels has differed between studies, indicating this relationship may vary depending on the study population, location, season, and statistical method used for the analysis.

Although human exposure field studies provide important information for understanding PM exposures, they also have limitations. These studies are expensive to conduct, particularly for population-based studies that require a large number of participants. Exposure studies are also burdensome on participants, which causes problems with retaining participants and with including certain groups that may be particularly at risk for health effects, such as children and people with cardiac or lung disease. In addition, results from an exposure study may only be applicable to the individual participants in that study and not representative of the general population.

Exposure modeling can therefore be a useful tool for understanding human exposures to environmental pollutants, provided sufficient data exist to develop a model. Exposure models combine microenvironmental concentrations with human activity data to estimate personal exposures, and allow analysis of the various exposure factors that influence personal exposures. *Population* exposure models use a probabilistic approach to randomly sample from distributions of available data for each exposure factor and predict the distribution of exposures for the population of interest (Zartarian et al., 2000; Law et al., 1997; MacIntosh et al., 1995; McCurdy, 1995; Ott et al., 1988). Input distributions incorporate the variability in the exposure factor data across as many individuals and conditions as data are available. The predicted distribution of exposures provides the range in exposures for the general population or sub-population of interest, and the likelihood of exposures above a particular level. Population exposure models also have limitations, particularly when insufficient data are available to characterize variability in the exposure factors.

A population exposure model for PM, called the Stochastic Human Exposure and Dose Simulation (SHEDS-PM) model (Version 1), has been developed. The main objectives for the initial model development were:

- to predict the distribution of daily averaged total exposure to PM for the population of an urban/metropolitan area

- to estimate the contribution of PM of ambient origin to total PM exposure
- to determine the major factors that influence personal exposure to PM, and
- to identify factors that contribute the greatest uncertainty to model predictions.

To meet these objectives, a case study was selected for the SHEDS-PM model and appropriate input databases were developed.

Population exposure to $PM_{2.5}$ in Philadelphia, PA was selected for the case study. $PM_{2.5}$ measurement data were obtained from the Metropolitan Acid Aerosol Characterization Study (MAACS) conducted in Philadelphia during 1992-93 (Wilson and Suh, 1997; Burton et al., 1996). During this study, PM measurement sites were operational during multiple seasons, and the spatial distribution of these sites was fairly even across the most densely populated areas within Philadelphia County (Figure 1). In addition to the availability of $PM_{2.5}$ data, several epidemiology studies have reported an association between air pollution and daily mortality in Philadelphia (Neas et al., 1999; Kelsall et al, 1997; Samet et al., 1997; Li and Roth, 1995; Moolgavkar et al., 1995; Samet et al., 1995; Schwartz and Dockery, 1992).

This paper describes the structure of the SHEDS-PM model, the algorithms used to estimate personal exposures and the types of input data required. The specific input databases developed for the Philadelphia $PM_{2.5}$ case study are also described, including both the microenvironmental data and the population/demographic data needed by the SHEDS-PM model. Results from the Philadelphia $PM_{2.5}$ case study are presented and discussed, along with the limitations of the model and the data needs identified through the case study.

Methods

Population Exposure Model Description

SHEDS-PM is a stochastic model, where input data for exposure factors are in the form of distributions that are randomly sampled for each individual in the simulation. These input distributions represent the variability in the exposure factors across the population (Taylor, 1993). Estimates of the knowledge- or measurement-based uncertainty associated with the exposure factor inputs are also required as input (Cullen and Frey, 1999). The SHEDS-PM model utilizes two-dimensional Monte Carlo sampling of the input distributions to propagate the variability and uncertainty in the inputs through to the predicted exposure distributions (Buck et al., 2001; MacIntosh et al., 1995; Bogen and Spear, 1987). Using this technique, the predicted output from the model includes estimates of both inter-individual variability in the population and uncertainty about any specific percentile of the predicted population distribution (see Figure 1, MacIntosh et al., 1995).

SHEDS-PM estimates PM exposures for individuals within a population. The population for the simulation is generated using demographic data at the census tract level from the U.S. Census. A fixed number of individuals are simulated to represent each census tract. Characteristics of the simulated individuals are randomly selected to match the demographic proportions within the census tract for gender, age, employment status and housing type. A smoking status (smoker/non-smoker) is randomly selected for each individual in the simulation population using smoking prevalence statistics for the U.S. by gender and age. Individual diaries of human activity pattern data are then randomly assigned to each individual by selecting from diaries that match the demographic characteristics of the simulated individual. The diaries contain data on the various microenvironments individuals spend time in during a day and the various activities performed while in each microenvironment.

The SHEDS-PM model simulates individual exposures in 8 specific microenvironments, including outdoors, indoors (residence, office, school, store, restaurant, bar) and in vehicles. PM concentrations in these microenvironments are needed to determine the individual PM exposures. For the outdoor microenvironment, the model requires a spatial field of PM concentrations at each census tract. The outdoor PM concentration database can be developed from spatial interpolation of either ambient PM measurements from community air monitoring sites or atmospheric dispersion model predictions. Typically, the temporal resolution of PM data from dispersion models is greater than ambient PM measurement data (hourly vs. 24-hour integrated), but measurement data may be available for a year or longer. The current structure of the SHEDS-PM model requires the outdoor PM concentrations at each census tract to have a temporal resolution of 12 or 24 hours and seasonal data for at least one year. For each simulated individual within a census tract, the SHEDS-PM model randomly selects an outdoor PM concentration from the input database for that census tract with equal proportions across all seasons.

PM concentrations in the indoor and in-vehicle microenvironments are calculated using microenvironment-specific equations for the relationship between ambient outdoor and microenvironmental PM concentrations. For the indoor residential microenvironment, a single-compartment, steady-state mass balance equation (_zkaynak et al., 1996) is used to calculate indoor PM concentrations ($C_{residential}$) from the infiltration of ambient PM indoors ($C_{residential\text{-}ambient}$) and indoor PM sources ($C_{residential\text{-}nonambient}$) as shown below:

$$C_{residential} = C_{residential\text{-}ambient} + C_{residential\text{-}nonambient}$$

$$= \frac{P\,ach}{ach + k} C_{ambient} + \frac{E_{smk}\,N_{cig} + E_{cook}\,t_{cook} + E_{other}\,T}{(ach + k)\,V\,T} \qquad (1)$$

where: $C_{ambient}$ = ambient outdoor PM concentration (μg/m^3)

P = penetration factor (unitless)

k = deposition rate (h^{-1})

ach = air exchange rate (h^{-1})

E_{smk} = emission rate for smoking (mg cig^{-1})

N_{cig} = number of cigarettes smoked during model time step

E_{cook} = emission rate for cooking (mg min^{-1})

t_{cook} = time spent cooking during model time step (min)

E_{other} = emission rate for other sources (mg h^{-1})

T = model time step (h)

V = residential volume (m^3)

The first term in (1) describes the infiltration of ambient PM indoors. The penetration factor P is the fraction of particles that penetrate the building envelope. The deposition rate k includes all processes except air exchange that result in loss of particles (sedimentation, adsorption, absorption, etc.). The second term in (1) describes the generation of particles from indoor sources. Indoor emissions from smoking and cooking are specified in the equation, while emissions from all other indoor sources are combined due to lack of data on these types of sources. Other indoor PM sources may include combustion sources such as wood-burning fireplaces and candle burning, cleaning activities, and/or resuspension of particles due to physical activity (Abt et al., 2000; _zkaynak et al., 1996; Wallace, 1996). For smoking, the model includes only exposure to environmental tobacco smoke (ETS) and not direct inhalation from active smoking. The estimation of particles emitted from smoking within the residence is determined from the number of cigarettes smoked in the residence by the individual (if a smoker) and/or by someone else smoking in the individual's residence. Particle generation from cooking is determined using the time the individual spent preparing food from the assigned activity diary. Since preparing food may or may not include particle-generating cooking, the time spent preparing food is multiplied by a random factor (from 0 to 1.0) to estimate the time spent cooking in the mass-balance equation.

For the non-residential microenvironments (office, school, store, restaurant, bar, vehicle), PM concentrations are determined using a linear regression equation developed from analysis of concurrent indoor and outdoor PM measurement data available for these microenvironments:

$$C_{microenvironment} = \mathrm{b}_0 + \mathrm{b}_1\, C_{ambient} \qquad (2)$$

This regression equation describes the relationship between microenvironmental and ambient outdoor PM concentrations (b_1) and the contribution of indoor PM sources (b_0).

The microenvironment-specific equations used to calculate residential and non-residential PM concentrations require input data for each parameter of the equation. The input data are distributions that characterize the variability in the measurement data for a parameter. For example, variability in available data for residential volume, V, used in the mass-balance equation above can be described by a lognormal distribution with a certain geometric mean and standard deviation. Additionally, input data for a particular parameter may include more than one set of distributions when data are available to characterize the variability by different categories. For the residential volume example above, separate distributions for a number of different housing types can be constructed. The specific variability distributions used for the equation parameters in the Philadelphia $PM_{2.5}$ case study are discussed in the following section.

For each individual in the simulation, the SHEDS-PM model randomly selects values from these distributions for the equation parameters. Microenvironmental PM concentrations for each individual are calculated using the selected parameter values and the equations for the residential and non-residential microenvironments. The individual's total PM exposure is then calculated using the microenvironmental PM concentrations and the time spent in the various microenvironments from the assigned activity diary for that individual.

The daily average total exposure for individual i (E_i) is calculated from the sum of daily average exposures for the m microenvironments encountered by the individual during the averaging time for exposure (24 hours), and each daily average microenvironmental exposure (E_{ij}) is calculated as the average time-weighted PM concentration for the microenvironment over n time steps:

$$E_i = \sum_{j=1}^{m} E_{ij} = \sum_{j=1}^{m} \left(\frac{1}{T} \sum_{k=1}^{n} C_{ijk}\, t_{ijk} \right) \tag{3}$$

where: E_i = daily average total exposure for individual i (µg/m^3)
E_{ij} = daily average exposure for individual i for microenvironment j (µg/m^3)
T = averaging time for exposure (24 h)
C_{ijk} = PM concentration for individual i in microenvironment j for model time step k (µg/m^3)
t_{ijk} = time spent by individual i in microenvironment j for model time step k (h)

It is important to note that the SHEDS-PM model does not keep track of the time series of microenvironmental exposures. The time spent in a microenvironment within each model time step (t_{ijk}) is the total over the time step. Two 12-hour time steps (daytime and nighttime) were used in the daily average total exposure calculation for the Philadelphia $PM_{2.5}$ case study.

In the SHEDS-PM model, microenvironmental exposures for each individual are also separated into exposures to PM of ambient origin ($E_{ij\text{-}ambient}$) and PM from indoor sources ($E_{ij\text{-}nonambient}$). The exposures are calculated as described in (3) using the ambient and non-ambient components of the microenvironmental PM concentrations ($C_{ij\text{-}ambient}$ and $C_{ij\text{-}nonambient}$, respectively):

$$E_i = \sum_{j=1}^{m} E_{ij} = \sum_{j=1}^{m} E_{ij_{ambient}} + \sum_{j=1}^{m} E_{ij_{nonambient}} \quad (4)$$

$$= \sum_{j=1}^{m} \left(\tfrac{1}{T} \sum_{k=1}^{n} C_{ijk_{ambient}} \, t_{ijk} \right) + \sum_{j=1}^{m} \left(\tfrac{1}{T} \sum_{k=1}^{n} C_{ijk_{nonambient}} \, t_{ijk} \right) \quad (5)$$

The SHEDS-PM model calculates the contributions of ambient and non-ambient PM to the average PM concentration in each microenvironment using the first and second terms, respectively, in the residential and non-residential equations above [(1) and (2)].

The estimated daily average total PM exposures for the simulated individuals in each census tract are combined to obtain a distribution of exposures for the entire population. The resulting distribution represents a 'cross-sectional' distribution of PM exposures for the simulated population that corresponds to the time period of the ambient outdoor PM data used as input. The predicted distribution is 'cross-sectional' and not 'longitudinal', since each individual is randomly assigned an ambient outdoor PM concentration from a particular date in the input data with equal proportions across all seasons. Therefore, SHEDS-PM simulates a different set of individuals for each date in the input data, whereas a longitudinal model would simulate the same individuals over time.

The resulting output distribution for the population describes the inter-individual variability in the predicted PM exposures across the population. The SHEDS-PM model also estimates the uncertainty in the output distribution using repeated model simulations and two-dimensional Monte-Carlo sampling of the input distributions. Estimates of the knowledge- or measurement-based uncertainty associated with each variability distribution for the equation parameters are required in the form of uncertainty distributions. For instance, the uncertainty in the geometric mean of the lognormal distribution for the residential volume example given above can be estimated by a normal distribution with a certain standard deviation around the value of the geometric mean. Similarly, the uncertainty in the value of the geometric standard deviation can also be estimated by a normal distribution around the value of the geometric standard deviation. For each SHEDS-PM simulation, the uncertainty distributions are randomly sampled to select values for the mean and standard deviation of each variability distribution during that simulation. Then for each individual within that simulation, values for the equation parameters are randomly selected from these variability distributions and used to estimate individual exposures as described above. This procedure is repeated for multiple model simulations, and the different population distributions obtained from the repeated simulations are used to characterize the uncertainty for any specific percentile of the predicted PM exposure distribution for the population.

Philadelphia $PM_{2.5}$ Case Study Inputs

Inputs to the SHEDS-PM model include two types of data: microenvironmental data and population/demographic data. Microenvironmental data were obtained from available field study measurements for each microenvironment. Population data for the model were obtained from available databases such as the U.S. Census and human activity pattern surveys. The methods used to develop variability and uncertainty distributions from available measurement study data for the Philadelphia case study are based on the techniques described in Cullen and Frey (1999).

Microenvironmental Data

Outdoor Microenvironment. $PM_{2.5}$ mass data for the outdoor microenvironment were obtained from measurements at eight monitoring locations during the MAACS study in

Philadelphia (Figure 1) from May 1992 through September 1993 (Burton et al., 1996). Daily or every-other-day $PM_{2.5}$ mass measurements (24-hour) were collected at three sites for the entire 503-day study period. Daily or every-other-day $PM_{2.5}$ mass was also collected at five additional sites during the summer of 1992 and at four of those sites during the summer of 1993. TEOM measurements of hourly $PM_{2.5}$ mass were also available at one site during the summer of 1992.

To obtain census tract $PM_{2.5}$ concentrations from the Philadelphia measurement study data, a spatial and temporal interpolation technique was applied to the daily $PM_{2.5}$ mass and concurrent meteorological data (Kibria et al., 2001). An analysis of the space-time structure of the data was conducted, missing values were imputed and spatial interpolation to the 482 census tract centroids within the defined model area (Figure 1) was performed. The Bayesian interpolation technique used is a multivariate extension of the more traditional method of kriging. An hourly model was also constructed from the hourly TEOM measurement data and used to predict hourly $PM_{2.5}$ concentrations at each census tract from the interpolated daily values. The hourly values were averaged over two 12-hour periods to obtain average daytime (7am-7pm) and nighttime (7pm-7am) $PM_{2.5}$ concentrations for each census tract that were used as input to the SHEDS-PM model.

Summary statistics for the interpolated census tract $PM_{2.5}$ concentrations used as input to the SHEDS-PM model by day/night and season are shown in Table 1. On average, $PM_{2.5}$ concentrations were higher in the daytime than nighttime, and the highest concentrations occurred during the summer seasons. A 24-hour average $PM_{2.5}$ concentration above the new $PM_{2.5}$ National Ambient Air Quality Standard (NAAQS) of 65 $\mu g/m^3$ occurred for only one day during June 1993 and at less than 10% of the census tracts on that day.

The spatial variation in $PM_{2.5}$ mass concentrations for Philadelphia was also investigated. Burton et al. (1996) report that during the summer the average daily $PM_{2.5}$ mass was similar across the Philadelphia MAACS sites (18-21 $\mu g/m^3$ in summer 1992; 21-25 $\mu g/m^3$ in summer 1993) and correlation among the sites was high ($r \geq 0.70$). However, the authors did not report whether $PM_{2.5}$ concentrations were consistently higher at certain sites compared to others. To determine if such a spatial pattern existed in the data, a daily mean $PM_{2.5}$ concentration across all the sites was calculated and the ratio to the daily all-site mean was determined for each site on each sample day. Most of the calculated ratios were within ±10% of the daily all-site mean $PM_{2.5}$ concentration, but a spatial trend was evident. On average, sites closest to the city center had ratios above 1.0, while sites farther from the city center were below 1.0. The same calculation was performed using the interpolated $PM_{2.5}$ concentrations at each of the census tracts and a similar result was obtained. $PM_{2.5}$ concentrations at the census tracts closest to the urban center were typically higher than the daily mean $PM_{2.5}$ concentration for all census tracts.

Indoor-Residential Microenvironment. Input distributions used in the Philadelphia $PM_{2.5}$ case study for the parameters of the indoor residential mass balance equation are presented in Table 2. The table shows the two types of input distributions needed for each equation parameter (variability and uncertainty). Air exchange rate distributions for each season were obtained from an analysis by Murray and Burmaster (1995). These authors report distributions of the variability in measured air exchange rates for residential structures in the U.S. by region of the country and by season. Distributions for the region with the most observations in the mid-Atlantic states (PA, NY, NJ, MD) for each season were chosen for the Philadelphia case study. Uncertainty in the mean of the air exchange rate distributions was estimated using a normal distribution and the standard error of the mean ($\sigma/n^{1/2}$) as the standard deviation of the uncertainty. Uncertainty in the standard deviation of the air exchange distributions was estimated using the upper and lower bounds of a 95% confidence interval for the standard deviation in a triangular distribution. Air exchange rates were significantly higher for the summer season, with greater uncertainty in the mean of the summer distribution due to fewer data points.

Distributions of the variability in residential volumes were obtained from an analysis of residential structures in the U.S. by Murray (1997). In this analysis, data from a US Dept. of Energy population-based survey (1993 RECS database) were used to construct residential volume distributions by type of residential structure. The volume distributions were derived from floor area measurements of the heated portions of homes and assumed an 8-foot ceiling height for all structures. Residential volumes were greater for single-family, detached structures and lower for multi-family units. The ‘other’ type of residential structure includes mainly mobile homes. Uncertainty distributions for residential volumes were estimated in the same manner as for the air exchange distributions using the standard error of the mean and the upper and lower bounds of a 95% confidence interval for the standard deviation.

Distributions for residential penetration, deposition and emission rates for $PM_{2.5}$ were obtained from an analysis by _zkaynak et al. (1996). In their analysis, a mass-balance equation was solved for these unknown parameters using non-linear least-squares regression of measurement data from the PTEAM study. The measurement data were collected in 175 homes in Riverside, California for two periods over 24 hours, which allowed separate estimation of these mass-balance parameters for daytime and nighttime. In Table 2, the means and standard deviations of the variability distributions correspond to the regression coefficients and one-half of their 95% confidence limits, respectively. The variability distributions are consistent with other estimates and measured data (Thornburg et al., 2001; Abt et al., 2000; Tung et al., 1999; Klepeis et al., 1996; Thatcher and Layton, 1995). The amount of data available from these other sources was used to estimate the uncertainty associated with the values of the variability distributions. The standard deviations for the uncertainty distributions were calculated as 10%, 25% and 30% of the mean values for penetration, deposition and indoor source emission rates, respectively.

The regression analysis by _zkaynak et al. (1996) allowed distributions for parameters of the residential mass balance equation to be defined when insufficient measurement data were available to characterize the variability in these parameters. Using regression coefficients instead of measurement data to define input distributions is not ideal, particularly when values outside the physical limits for some parameters could be produced using the predicted distribution. For example, negative emission rates could be obtained from some of these distributions. Negative emission rates were set to zero when this occurred (less than 5% of the simulated individuals). In addition, daytime penetration values slightly greater than 1.0 were often obtained from that distribution, when penetration can not physically exceed 1.0. However, no significant difference between results was observed when an upper limit of 1.0 for penetration was used.

Input data for the remaining mass balance equation parameters (number of cigarettes smoked and time spent cooking) are discussed along with the other population-related data in the following section.

Non-residential Microenvironments. Input distributions used in the case study for the parameters of the linear regression equations for non-residential microenvironments are shown in Table 3. The indoor/outdoor $PM_{2.5}$ measurement data used to develop these distributions are summarized in a review article by Zufall et al. (2001). The variability distributions for $PM_{2.5}$ concentrations in non-residential microenvironments were defined as normal distributions with means calculated from the linear regression equation (2) described above and standard deviations calculated from the actual measurement data. Uncertainty distributions for the linear regression equation parameters (b_1 and b_0) were estimated from the regression coefficients and their standard errors. Although these regression equation parameters may be significantly correlated, the distributions were sampled independently. However, the distribution of predicted concentrations in each microenvironment was compared with the distribution of measured concentrations and no outlying values were found in the predicted concentrations.

For the restaurant and bar microenvironments, a similar regression equation developed by Ott et al. (1996) was used that includes smoking as an indoor source term. The smoking source term consisted of the active smoking count, *ASC*, and a corresponding PM emission rate. A uniform distribution from 0 to 3 was used for the active smoking count and the uncertainty distribution for the additional equation parameter (b_2) is shown in Table 3. In addition, a random factor was also added to select whether smoking occurred for restaurants, while smoking was always included for bars.

Values of the linear regression coefficients for offices with no smoking allowed (b_1 " std. error=0.18 " 0.06; b_0 " std. error=3.6 " 1.3) indicate that ambient outdoor $PM_{2.5}$ did not efficiently infiltrate into the office microenvironments (most likely due to HVAC systems) and minimal indoor $PM_{2.5}$ sources were present in non-smoking offices. In contrast, ambient outdoor $PM_{2.5}$ did contribute significantly to indoor $PM_{2.5}$ concentrations in schools, stores, restaurants and bars. Indoor $PM_{2.5}$ sources were also significant for these microenvironments. The regression equation for the vehicle microenvironment incorporated measurements from near roadways in addition to data collected inside vehicles. A low b_1 for the ambient $PM_{2.5}$ contribution was obtained in the fitted regression model for vehicle measurements, possibly due to limited penetration and high deposition within vehicles (Rodes et al., 1998). A high b_0 for vehicles was also obtained that accounts for the elevated concentrations often measured near roadways. Smoking exposure while in vehicles was not included in the model because information on the prevalence of smoking within vehicles was not available. This excludes a potentially high short-term exposure to non-ambient $PM_{2.5}$ from the total exposure predicted by the model. For all other indoor microenvironments not specified in the model, the parameters for one of the non-residential microenvironments were randomly selected with equal probability and used to calculate the PM concentration for the other microenvironments.

Population data

Census data. Demographic data for the population living in Philadelphia during 1992-93 were obtained from the 1990 U.S. Census. The Primary Metropolitan Statistical Area (PMSA) for Philadelphia as defined by the U.S. Census includes Philadelphia County and portions of 6 surrounding counties. This PMSA was judged too large for the case study and a smaller area was selected that included Philadelphia County and parts of 3 adjacent counties. Two of these counties are directly north and west of Philadelphia County, and the third is the city of Camden, NJ across the PA-NJ border from Philadelphia County. Figure 1 shows the area selected for the model case study. This area includes a total of 482 census tracts and has a total population of approximately 2.2 million.

The U.S. Census data provided population totals for 31 age categories for each gender, 4 employment categories for each gender, and 10 housing type categories. The relative proportions for each category within each of the 482 census tracts were calculated and used as input to the SHEDS-PM model. Analysis of these proportions by census tract indicated that the percentage of working adults varied across the modeled area, as did the proportion of elderly and children. The majority of the population of Philadelphia included in the modeled area resided in single family homes, with most of those being attached units (row houses). Large apartment complexes dominated certain census tracts and census tracts containing colleges or elderly housing were also evident.

Smoking statistics. Input data on smoking was required to determine exposure to environmental tobacco smoke (ETS) in the residence for the case study. Smoker prevalence during 1992-93 was obtained from a U.S. Dept. of Health report (National Center for Health Statistics, 1998). Only data for adults (18 years and older) were included in this report, so additional data for adolescents aged 12 to 17 during 1994-95 were obtained from another U.S. Dept. of Health report (Substance Abuse and Mental Health Services Administration, 1996). Children under age 12 were not included as possible smokers in the case study. A limited amount of smoking prevalence data was also available for Philadelphia and Pennsylvania from more recent years to compare with the U.S. statistics. In general, smoking prevalence in Philadelphia was higher than the average for Pennsylvania, and Pennsylvania was higher than the average for the U.S. Therefore, smoker prevalence in the case study could be an underestimate for the population of Philadelphia.

The smoking prevalence data used as input for the Philadelphia case study are shown in Table 4. Prevalence data from 1992 and 1993 were averaged for each age/gender category and used as the proportion of the population that were smokers (data for 1994 and 1995 were averaged for 12-17 year olds). Variability distributions were not used since U.S. smoking prevalence data were available for a number of different age/gender categories and only one data source was used for each age/gender category. Uncertainty distributions for these proportions were estimated based on the difference between the values for the two years that were averaged. The data in Table 4 show that more males were smokers than females across all adult age groups and that smoking prevalence was highest in either the 25-34 or 35-44 year-old age groups.

The number of cigarettes smoked by a smoker in their residence was obtained from questionnaire data for the National Human Activity Pattern Survey, or NHAPS (Klepeis et al., 2001; Klepeis et al., 1996; Tsang and Klepeis, 1996). NHAPS is a population-based telephone-recall survey database for the U.S. that includes more than 9,000 questionnaires and diaries. Several questions regarding smoking exposures were asked of the survey respondents. One question specifically asked smokers the number of cigarettes they smoked in their residence during the previous day. Table 5 summarizes these data by age and gender that were used as input for the SHEDS-PM model. The model randomly selected a range for the number of cigarettes smoked in the residence by a smoker from these proportions according to the age and gender of the simulated individual and then randomly sampled from a uniform distribution from the minimum to the maximum of the selected range. The data in Table 5 indicate that the older the smoker, the greater the number of cigarettes that were smoked in the residence and males generally smoked more cigarettes than females.

The prevalence of smoking by others in a residence and the number of cigarettes that were smoked according to age, gender and smoking status were also obtained from the NHAPS questionnaire data. All respondents were asked whether someone else smoked cigarettes in the respondents home during the previous day, and if yes then how many cigarettes were smoked. Tables 6 and 7 display this data used to randomly select whether the simulated individual was exposed to ETS from others smoking in their residence and the number of cigarettes they were exposed to. Smokers had higher rates of exposure to others smoking in their residence than non-smokers.

Activity pattern data. Individual diaries of human activity pattern data were obtained from EPA's Consolidated Human Activity Database (CHAD) that includes over 22,000 diary days from 10 surveys including NHAPS (McCurdy et al., 2000). The CHAD diaries are sequential records of the time spent by each survey respondent in various microenvironments and what activities were performed while in the microenvironment over flexible time intervals (1-minute minimum). The total time in each of the 8 microenvironments specified in the SHEDS-PM model for two 12-hour periods (daytime: 7 am – 7 pm; nighttime: 7 pm – 7 am) was calculated for each of the CHAD diaries and used as input to the model for the case study. Time spent in all other indoor microenvironments not specified by the model was totaled to account for all 24 hours in the day for each individual. In addition, time spent preparing food in the residence during daytime and nighttime was also totaled for use as input to the residential mass balance equation previously described.

The CHAD database also includes corresponding questionnaire data for each survey respondent. The questionnaire data used in the case study included gender, age, employment status and smoking status. These data were used to match individual diaries of human activity pattern data to each individual in the simulation population. In most cases sufficient numbers of diaries were available for random assignment to the simulated individuals within each of the various combinations of gender, age group, employment status and smoking status. However, for those cases when not enough diaries were available, duplicate copies of the diaries for the particular combination were generated until the total number of diaries needed was reached.

Results

The following results for the Philadelphia $PM_{2.5}$ case study were produced using the population and microenvironmental input data described above. Each SHEDS-PM model simulation included realizations for 500 individuals per census tract for the 482 census tracts within the defined model area for Philadelphia. Demographic characteristics were randomly selected for each individual according to the calculated proportions for each census tract from the U.S. Census data. Gender and age group were used to select a smoking status for each individual based on the U.S. smoker prevalence data. Human activity diaries from CHAD were randomly assigned by selecting from diaries that matched the gender, age group, employment status and smoking status of that individual.

Microenvironmental data were also randomly selected for each simulated individual. A pair of day/night outdoor $PM_{2.5}$ concentrations was selected for each individual from the 503 days of interpolated measurement data. Values for each microenvironment-specific equation parameter were randomly selected from the variability distributions and used along with the assigned outdoor $PM_{2.5}$ concentrations to calculate daytime and nighttime $PM_{2.5}$ concentrations in the residential and non-residential microenvironments for each individual. Daily average microenvironmental exposures for each individual were calculated using the time spent in each microenvironment during the day/night from the assigned CHAD activity diary and the day/night $PM_{2.5}$ concentrations for each microenvironment. The daily microenvironmental exposures were then summed to obtain the daily total $PM_{2.5}$ exposure for each individual. The contributions from $PM_{2.5}$ of ambient origin and indoor $PM_{2.5}$ sources to the total and microenvironmental exposures for each individual were also calculated.

Results for the simulated individuals within each census tract were combined to obtain the population distribution of daily total $PM_{2.5}$ exposures for Philadelphia for one model simulation. Uncertainty estimates for the population distribution were produced from 100 model simulations. Output from the model are summarized using cumulative frequency distributions of microenvironmental $PM_{2.5}$ concentrations, time spent in microenvironments, total and microenvironmental $PM_{2.5}$ exposures and ambient/non-ambient $PM_{2.5}$ exposures for the simulated individuals representing the population of Philadelphia.

Microenvironmental $PM_{2.5}$ Concentrations

Cumulative frequency distributions of daily average $PM_{2.5}$ concentrations within the 8 microenvironments specified in the model case study for the simulated population of Philadelphia are shown in Figure 2. The $PM_{2.5}$ concentration distribution for the outdoor microenvironment has a median of 12 $\mu g/m^3$, with a range of 2 to 67 $\mu g/m^3$. Variability in the outdoor $PM_{2.5}$ concentrations was low, with 90% of the simulation population having concentrations between 7 and 27 $\mu g/m^3$. In contrast, concentrations of $PM_{2.5}$ in the indoor-residential microenvironment varied greatly within the population. The distribution has a median of 18 $\mu g/m^3$, with 10% of the simulation population having $PM_{2.5}$ levels greater than 69 $\mu g/m^3$ in their residence due to the contribution of indoor sources such as cooking and smoking. However, approximately 40% of the simulation population was not exposed to indoor sources in their residence and the $PM_{2.5}$ concentration distribution for this segment of the population (not shown) has the same median value and low variability as observed for the outdoor $PM_{2.5}$ concentration distribution.

The $PM_{2.5}$ concentration distributions for most of the non-residential microenvironments (offices, schools, stores, and vehicles) have similar profiles in Figure 2 since they were estimated from relationships with ambient $PM_{2.5}$ levels. Office $PM_{2.5}$ concentrations were lower than outdoor levels (median=6 $\mu g/m^3$), school and store $PM_{2.5}$ concentrations were similar to outdoor levels (median=15 and 18 $\mu g/m^3$, respectively), and in-vehicle $PM_{2.5}$ concentrations were greater than outdoor levels (median=37 $\mu g/m^3$). For the office, school and store microenvironments, variability in $PM_{2.5}$ concentrations across the population was low, while in-vehicle $PM_{2.5}$ concentrations were more variable. Indoor sources of $PM_{2.5}$ such as cooking and smoking influenced the $PM_{2.5}$ concentration distributions for restaurants and bars, resulting in highly variable $PM_{2.5}$ levels in these microenvironments across the population.

Time Spent in Microenvironments

The distributions of time spent in different microenvironments per day for the simulated population of Philadelphia are presented in Table 8. The percent of the population that spent time in each microenvironment is also shown. Nearly all individuals in the simulation population (99%) spent some time in the indoor-residential microenvironment, with most spending at least 9 hours per day in their residence. Time spent in this microenvironment was also highly variable, with a median of 17.7 hours and the 90th percentile at 23 hours. Nearly two-thirds of the case study population spent some time traveling in vehicles, while just over half spent some time outdoors. Time spent outdoors for the NHAPS diary data may be negatively biased according to Klepeis et al. (2001). The results suggest that brief periods of time outdoors could be missing from these recall diaries. One quarter of the individuals who spent time outdoors, however, spent more than 3.5 hours outdoors. Only 14% of the population spent time in an office and 17% spent time in a school, but 25% of these individuals spent more than 7 hours in these microenvironments. The microenvironments specified in the SHEDS-PM model were sufficient to account for the daily activities of most individuals in the simulated population.

However, approximately 15% of the total population spent a significant amount of time (> 6 hours) within indoor microenvironments not specified in the current version of the model.

Total and Microenvironmental $PM_{2.5}$ Exposures

The cumulative frequency distribution of daily *total* $PM_{2.5}$ exposures predicted by the SHEDS-PM model for the simulated population of Philadelphia is shown in Figure 3. The median daily total $PM_{2.5}$ exposure for the population was 20 $\mu g/m^3$. Significant variability in total $PM_{2.5}$ exposures across the population was evident in Figure 3, with total $PM_{2.5}$ exposures less than 10 $\mu g/m^3$ for 10% of the population and greater than 59 $\mu g/m^3$ for the upper 10%.

Also displayed in Figure 3 are the distributions of *microenvironmental* $PM_{2.5}$ exposures (exposures due to time spent in the various microenvironments) that contributed to the total $PM_{2.5}$ exposure distribution. It is evident from this figure that indoor residential $PM_{2.5}$ exposures had the greatest influence on total $PM_{2.5}$ exposures compared to the other microenvironments. The median indoor residential $PM_{2.5}$ exposure was 13 $\mu g/m^3$ compared to medians of 3 $\mu g/m^3$ or less for outdoor, in-vehicle and indoor non-residential exposures. Microenvironmental exposures were determined from time-weighted microenvironmental concentrations; therefore, both time spent in the microenvironment and the concentration in the microenvironment contributed to the estimates of exposure. The number of hours spent in the indoor residential microenvironment was generally much higher than the other microenvironments across the population (Table 8), resulting in a large contribution to total $PM_{2.5}$ exposures from indoor residential exposures. In contrast, the contribution of indoor non-residential exposures was less, due to the low amounts of time spent in microenvironments with relatively high $PM_{2.5}$ concentrations (restaurants and bars) or the low concentrations in microenvironments where individuals spent substantial time (offices).

Due to the fact that total $PM_{2.5}$ exposures were strongly influenced by indoor residential exposures, the contribution of indoor $PM_{2.5}$ sources within the residential microenvironment was further examined. One-third of the simulation population was exposed to ETS in their residence. Distributions of indoor residential and total $PM_{2.5}$ exposures for the two-thirds of the simulation population without ETS exposure in the residence are also shown in Figure 3. The presence of a smoker in an individual's residence added approximately 10 $\mu g/m^3$ to the 75th percentile for the distribution of indoor-residential $PM_{2.5}$ exposures, and approximately 28 $\mu g/m^3$ to the 90th percentile. The predicted distributions of total $PM_{2.5}$ exposures are also significantly different for these populations. The median total $PM_{2.5}$ exposure for those without ETS exposure in their residence was 16 $\mu g/m^3$ compared to 20 $\mu g/m^3$ for the entire population, and the 90th percentile of the distribution was 32 $\mu g/m^3$ versus 59 $\mu g/m^3$.

Results from the SHEDS-PM model also included estimates of the uncertainty in the predicted exposure distributions. Uncertainty for the total $PM_{2.5}$ exposure distribution for the simulation population is displayed in Figure 4. This figure shows the range in the values of selected distribution percentiles for the 100 uncertainty iterations of the model. Uncertainty in the input distributions had an impact on the predicted total $PM_{2.5}$ exposure distribution, particularly at the high exposure tail of the distribution. The median (50th percentile) of the total $PM_{2.5}$ exposure distribution ranged from 17-25 $\mu g/m^3$ for the 100 iterations, while the 90th percentile ranged from 42-80 $\mu g/m^3$.

Ambient $PM_{2.5}$ Exposures

The SHEDS-PM model also estimated the contribution of $PM_{2.5}$ of ambient origin to total $PM_{2.5}$ exposure for the case study population. Both ambient and non-ambient $PM_{2.5}$ exposures were calculated for each microenvironment and totaled over all the various microenvironments for each individual. Distributions of daily exposure to ambient and non-ambient $PM_{2.5}$ for the simulation population are shown in Figure 5 along with the distribution of daily total $PM_{2.5}$ exposures for comparison. The median daily exposure to ambient $PM_{2.5}$ was 7 $\mu g/m^3$ for the simulation population. Variability in the distribution of ambient $PM_{2.5}$ exposures was significantly less than for the distribution of total $PM_{2.5}$ exposures, with 90% of the simulation population having ambient $PM_{2.5}$ exposures between 3 and 18 $\mu g/m^3$. Distribution statistics and percentiles are also presented in Table 9. Exposure to ambient $PM_{2.5}$ was less than the NAAQS level of 65 $\mu g/m^3$ for all the simulated individuals in this case study.

It is also evident in Figure 5 that total $PM_{2.5}$ exposures were highly influenced by exposure to non-ambient $PM_{2.5}$ (i.e. indoor sources of $PM_{2.5}$). Exposure to ETS in the indoor-residential microenvironment was again the main source of that influence. The variability in non-ambient $PM_{2.5}$ exposures was significantly less for those in the simulation population without exposure to ETS in their residence as shown in Figure 5.

The contribution of ambient $PM_{2.5}$ exposures was also calculated on a percentage basis. The distribution of daily ambient $PM_{2.5}$ exposure as a percentage of daily total $PM_{2.5}$ exposure is shown in Table 9. While the median value for the simulation population was 37.5%, the percent of total $PM_{2.5}$ exposures from ambient $PM_{2.5}$ was highly variable across the population. For one-quarter of the population, ambient $PM_{2.5}$ exposure contributed less than 20% to total $PM_{2.5}$ exposure while for another quarter of the population (upper 25% of the population distribution) ambient $PM_{2.5}$ exposure contributed more than 57% to their total $PM_{2.5}$ exposure. The percent contribution from ambient $PM_{2.5}$ exposures was higher across the entire distribution for those without ETS exposures in the residence, as shown in Table 9, and the distribution was also highly variable for this segment of the population.

The predicted exposure distributions for the simulation population were also analyzed by various demographic characteristics. For example, in Table 9 distributions for the percent of total $PM_{2.5}$ exposures from ambient $PM_{2.5}$ are displayed for selected age categories. For the youngest (0-4 and 5-11 yr) and oldest (>65 yr) in the population, ambient $PM_{2.5}$ exposures contributed more to their total $PM_{2.5}$ exposures than for the other age categories. The variability in each age group was consistently high and similar to the variability for the entire population.

Spatial Analysis of Total and Ambient $PM_{2.5}$ Exposure

An analysis of spatial trends in the $PM_{2.5}$ concentration data used as input to the SHEDS-PM model was described above. This analysis showed that $PM_{2.5}$ levels were typically higher at the census tracts closest to the urban center, but the magnitude of the differences was small. A similar spatial analysis for both total and ambient $PM_{2.5}$ exposures did not find a similar spatial pattern in exposures. The lack of spatial differences in the predicted total and ambient $PM_{2.5}$ exposures indicates that the inter-individual differences within a census tract were greater than any spatial difference in the ambient outdoor $PM_{2.5}$ concentrations between census tracts.

Apportionment of Total $PM_{2.5}$ Exposure

Results from the SHEDS-PM model were used to apportion total $PM_{2.5}$ exposures by type of microenvironment and $PM_{2.5}$ source. The apportionment as percent of total $PM_{2.5}$ exposures is summarized in Table 10 for the entire population and for those without ETS exposure in the residence. Comparison of the mean values indicates that, in general, residential exposures contributed the most to total $PM_{2.5}$ exposures, with the percent contribution from ambient and non-ambient $PM_{2.5}$ in the residence differing between the entire population and those without ETS exposure in the residence. The mean values for the percent contribution from the outdoor and non-residential microenvironments were lower than for the residential microenvironment.

Contributions from non-ambient $PM_{2.5}$ were significantly higher than for ambient $PM_{2.5}$ in non-residential microenvironments. The percentiles of the distribution for the simulated population of Philadelphia are also shown in Table 10. While the mean values provide a useful summary statistic for comparison of the apportionment, high variability in the distributions demonstrate that these mean values do not represent the population as a whole. The apportionment of total $PM_{2.5}$ exposures between microenvironments and source types (ambient/non-ambient) differed across various segments of the population due to differences in activity patterns and other exposure factors.

Discussion

The SHEDS-PM population exposure model (Version 1) was developed using a case study of $PM_{2.5}$ exposures for the population of Philadelphia. While no personal $PM_{2.5}$ exposure measurements were collected in Philadelphia during the time of the ambient $PM_{2.5}$ measurements, data from other urban areas are available for comparison with the model results. The population mean for daily total $PM_{2.5}$ exposures (30 $\mu g/m^3$) predicted by the SHEDS-PM model for Philadelphia is similar to personal $PM_{2.5}$ exposure measurement data from population-based studies in Toronto (Pellizzari et al., 1999) and Switzerland (Oglesby et al., 2000). Mean personal $PM_{2.5}$ measurements were 28.4 and 23.7 $\mu g/m^3$, respectively, for those studies. Oglesby et al. (2000) also report a mean of 17.5 $\mu g/m^3$ when no exposure to ETS occurred, which is similar to the SHEDS-PM population mean of 19.7 $\mu g/m^3$ for individuals without ETS exposure in their residence. Longitudinal studies of specific sub-populations within urban areas have also measured similar levels. For example, Rojas-Bracho et al. (2000) report the mean personal $PM_{2.5}$ measurement for 17 adults with COPD in Boston was 21.6 $\mu g/m^3$ with a range of 1 to 128 $\mu g/m^3$. However, Williams et al. (2000) report a mean personal $PM_{2.5}$ concentration of 13.0

$\mu g/m^3$ for elderly adults in Baltimore (n=23), and Evans et al. (2000) report mean personal $PM_{2.5}$ concentrations of 13.3 and 11.1 $\mu g/m^3$ for elderly adults in two studies in Fresno (n=24 and 12).

Results from the SHEDS-PM model case study indicate the importance of including estimates of input uncertainty in population exposure models along with the input variability. Uncertainty in the predicted distribution of daily total $PM_{2.5}$ exposures was highest at the upper end of the distribution where the 90^{th} percentile varied by as much as 38 $\mu g/m^3$ between the 100 iterations of the model. This also emphasizes the need to evaluate the validity of the predicted extremes in the exposure distributions.

The SHEDS-PM case study results showed that exposures to ambient $PM_{2.5}$ were much less variable across the simulation population than total $PM_{2.5}$ exposures. This suggests that human activity patterns (the microenvironments encountered and the amount of time spent in each) do not have a strong influence on ambient $PM_{2.5}$ exposures. The predicted exposures to ambient $PM_{2.5}$ were, however, dependent on the variability in the outdoor $PM_{2.5}$ concentrations. Ambient $PM_{2.5}$ exposure and outdoor $PM_{2.5}$ concentration were highly correlated (r=0.89, p<0.001) for the simulation population. Exposure to other sources of $PM_{2.5}$ contributed more to the variability in predicted total $PM_{2.5}$ exposures than ambient $PM_{2.5}$ exposure. Total $PM_{2.5}$ exposures were highly correlated with non-ambient $PM_{2.5}$ exposures for the simulation population (r=0.99, p<0.001) and not correlated with ambient $PM_{2.5}$ exposures (r=0.08). In addition, there was no significant statistical relationship between exposure to ambient $PM_{2.5}$ and exposure to non-ambient $PM_{2.5}$ (r=-0.08). Individuals in the simulation population with the same ambient $PM_{2.5}$ exposure had a wide range of exposures to non-ambient $PM_{2.5}$.

Age differences in the percent of total $PM_{2.5}$ exposures from ambient $PM_{2.5}$ for the SHEDS-PM case study indicated that certain sub-populations, such as the elderly, had a greater percentage of their total exposure from ambient $PM_{2.5}$ compared to the population as a whole, although the variability was high across all age groups. Comparisons across other demographic characteristics such as employment status or housing type are also possible with the SHEDS-PM model output. In addition, the population could be ranked by time spent in a particular microenvironment (high/medium/low) and the distributions compared.

The results from the SHEDS-PM model also allowed apportionment of the predicted total $PM_{2.5}$ exposure by microenvironment and $PM_{2.5}$ source type. The apportionment results emphasized the importance of the residential microenvironment in contributing to total $PM_{2.5}$ exposures for the simulated population. The majority of ambient $PM_{2.5}$ exposures occurred in this microenvironment compared to outdoor and non-residential $PM_{2.5}$ exposures. When indoor residential $PM_{2.5}$ sources were limited (i.e. no ETS exposure in residence), as expected, ambient $PM_{2.5}$ exposures in the residence were an even greater proportion of predicted total $PM_{2.5}$ exposures.

Limitations and data needs

Development of the SHEDS-PM model (Version 1) using the Philadelphia $PM_{2.5}$ case study has identified several key limitations, as well as data needs for improving inputs to the model, reducing uncertainty and further refining of the model structure. For instance, few studies have been conducted that provide daily PM measurements over multiple seasons and across multiple sites within an urban area. Without this type of measurement data for input to the SHEDS-PM model, statistical methods must be used to interpolate the data both spatially and temporally. Improving the capability of dispersion models to predict ambient PM concentrations within urban areas at greater spatial resolution and over a year time period, would also provide useful input data for SHEDS-PM applications.

Personal exposure studies with appropriate study designs and sufficient measurements for a thorough evaluation of the SHEDS-PM model results are also limited. While the predicted total $PM_{2.5}$ exposures for the case study are similar to available personal $PM_{2.5}$ measurements, a comprehensive population-based study in an urban area with both personal and microenvironmental sampling would allow for a full evaluation of all aspects of the model.

Certain assumptions were made in the development of the structure of the SHEDS-PM model that may have had an impact on the predicted results. For example, in using the mass balance equation to calculate residential PM concentrations it was assumed that the residence was a well-mixed single-compartment under equilibrium conditions. The time dependence of the physical factors in the equation were thereby ignored, as well as the effect of several factors on mixing, including the extent of door/window opening, indoor/outdoor differences in temperature, type of heating/cooling system, etc. However, the impact of these assumptions may be minimal considering the length of the time step for this version of the SHEDS-PM model (12 hours).

Measurement studies are needed to better characterize the physical factors governing infiltration of ambient $PM_{2.5}$ into residential microenvironments, including the influence of opening doors and windows, the type and use of HVAC systems and other characteristics of residential buildings. Diurnal patterns in residential air exchange rates may be an important factor that is currently not accounted for by the model. Also, particle-generating sources within the residence such as smoking and cooking were shown to have a significant impact on total $PM_{2.5}$ exposures and additional data are needed to reduce the uncertainty in the values used for those inputs. For example, sufficient information was not available in the human activity diary data to characterize residential cooking activities in detail. Finally, the impact of other indoor residential $PM_{2.5}$ sources also needs further investigation, including differences between age groups (elderly, children) in the resuspension of particles due to physical activity.

Currently insufficient data are available to fully apply a mass balance equation to determine PM concentrations in the non-residential microenvironments included in the SHEDS-PM model. Further measurement data are needed to characterize the important physical factors for different types of vehicles, office buildings, schools, and stores (particularly mall stores vs. stores open to outdoors). In addition, data for the other indoor microenvironments not specified in the model could also improve the exposure predictions for certain segments of the population.

The next version of the SHEDS-PM model (Version 2) is currently under development. The main objectives for this next phase in the model development include structuring the model to predict exposures on an hourly time-series basis and to estimate intake dose based on activity level and ventilation rate. These refinements to the model structure will also require input data to be on an hourly basis and therefore, more continuous measurements are needed to develop these model inputs. Adding the capability of estimating exposures for each individual over time (longitudinal exposures) to the model is also an objective and will require information on the longitudinal patterns of human activities.

The continued development and evaluation of the SHEDS-PM population exposure model is being conducted as part of EPA/ORD's effort to develop a source-to-dose modeling system. This type of exposure and dose modeling system is considered to be important for scientific and policy evaluation of the critical pathways, exposure factors and source types influencing human exposures to a variety of environmental pollutants, including particulate matter.

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Table 1. Summary statistics for the interpolated $PM_{2.5}$ concentrations ($\mu g/m^3$) at 482 census tracts within Philadelphia, PA for the 503-day MAACS study period (May 1992 – September 1993) by season and day/night.

Season		Mean	Standard Deviation	Median	Range
Winter	Day	12.2	6.8	10.2	2.0 – 47.8
	Night	10.4	4.4	9.8	2.0 – 33.5
Spring	Day	13.6	7.2	11.7	3.1 – 56.6
	Night	12.4	5.1	11.5	1.0 – 32.5
Summer	Day	19.3	11.0	17.2	4.4 – 82.7
	Night	17.3	7.2	16.3	4.6 – 51.5
Autumn	Day	14.5	7.7	12.7	4.6 – 50.4
	Night	12.7	5.1	11.7	4.3 – 29.7

Table 2. Input distributions for parameters of the mass balance equation used to determine $PM_{2.5}$ concentrations in the residential microenvironment[1].

Equation Parameter	Category	Variability Distribution	Uncertainty Distributions Mean	Standard Deviation	Ref.[2]
Air exchange rate (h^{-1})	Winter	LogN(-0.8, 0.7)[3]	N(-0.8, 0.10)	Tri(0.6, 0.7, 0.85)	a
	Spring	LogN(-0.8, 0.8)[3]	N(-0.8, 0.06)	Tri(0.7, 0.8, 0.9)	a
	Summer	LogN(-0.2, 0.7)[3]	N(-0.2, 0.26)	Tri(0.6, 0.7, 0.9)	a
	Autumn	LogN(-1.0, 0.5)[3]	N(-1.0, 0.19)	Tri(0.4, 0.5, 0.7)	a
Volume (m^3)	Single-family, detached	LogN(6.02, 0.50)[4]	N(6.02, 0.03)	Tri(0.46, 0.50, 0.55)	b
	Single-family, attached	LogN(5.79, 0.50)[4]	N(5.79, 0.09)	Tri(0.46, 0.50, 0.55)	b
	Multi-family, < 5 units	LogN(5.16, 0.44)[4]	N(5.16, 0.05)	Tri(0.37, 0.44, 0.54)	b
	Multi-family, ≥ 5 units	LogN(5.16, 0.44)[4]	N(5.16, 0.05)	Tri(0.37, 0.44, 0.54)	b
	Other	LogN(5.34, 0.36)[4]	N(5.34, 0.08)	Tri(0.28, 0.36, 0.50)	b
Penetration (unitless)	Day	N(1.00, 0.080)	N(1.00, 0.10)	N(0.080, 0.008)	c
	Night	N(0.89, 0.058)	N(0.89, 0.09)	N(0.058, 0.006)	c
Deposition (h^{-1})	Day	N(0.27, 0.098)[5]	N(0.27, 0.07)	N(0.098, 0.025)	c
	Night	N(0.39, 0.090)[5]	N(0.39, 0.10)	N(0.090, 0.023)	c
Cooking emission rate (mg min^{-1})	Day	N(1.56, 0.412)	N(1.56, 0.47)	N(0.412, 0.124)	c
	Night	N(0.69, 0.439)	N(0.69, 0.21)	N(0.439, 0.132)	c
Smoking emission rate (mg cig^{-1})	Day	N(10.9, 2.16)	N(10.9, 3.27)	N(2.16, 0.648)	c
	Night	N(16.9, 2.43)	N(16.9, 5.07)	N(2.43, 0.729)	c
Other emission rate (mg h^{-1})	Day	N(1.46, 0.833)	N(1.46, 0.44)	N(0.833, 0.250)	c
	Night	N(0.78, 0.517)	N(0.78, 0.23)	N(0.517, 0.155)	c

Notes:
[1]Shape of distribution: LogN=lognormal(geometric mean, geometric standard deviation), N=normal(mean, standard deviation), Tri=triangular(minimum, mode, maximum).
[2]References: a=Murray and Burmaster (1995), b=Murray (1997), c=Özkaynak et al. (1996).
[3]Minimum value=0.01 h^{-1}
[4]Minimum value=50 m^3
[5]Minimum value=0.1 h^{-1}

Table 3. Input distributions for parameters of the linear regression equations used to determine $PM_{2.5}$ concentrations in non-residential microenvironments[1].

Microenvironment	Variability Distribution[2]	Uncertainty Distributions		
		b_0	b_1	b_2
Vehicle (no smoking)	N($C_{\mu e}$, 12)	N(33.0, 7.2)	N(0.26, 0.14)	
Office (no smoking)	N($C_{\mu e}$, 2.9)	N(3.6, 1.3)	N(0.18, 0.06)	
School	N($C_{\mu e}$, 5.4)	N(6.8, 1.4)	N(0.60, 0.09)	
Store	N($C_{\mu e}$, 2.1)	N(9.0, 3.6)	N(0.74, 0.18)	
Restaurant/Bar[3]	N($C_{rest/bar}$, 10)	N(9.8, 0.5)	N(1.00, 0.05)	Tri(32, 40.4, 50)

Notes:
[1]Data source: Zufall et al. (2001); Shape of distribution: N=normal(mean, standard deviation), Tri=triangular(minimum, mode, maximum).
[2]Mean of distribution calculated using linear regression equation: $C_{\mu e} = b_0 + b_1 C_{ambient}$.
[3]Equation for restaurant/bar has additional term (see text): $C_{rest/bar} = b_0 + b_1 C_{ambient} + b_2 ASC$, where ASC is the active smoking count and varies uniformly from 0 to 3.

Table 4. Input data on smoking prevalence by age group and gender[1].

Age Group	Gender	Proportion that are Smokers	Uncertainty Distribution
12 – 13 yr	Both	0.100	N(0.100, 0.004)
14 – 15 yr	Both	0.201	N(0.201, 0.004)
16 – 17 yr	Both	0.293	N(0.293, 0.004)
18 – 24 yr	Males	0.284	N(0.284, 0.008)
	Females	0.239	N(0.239, 0.008)
25 – 34 yr	Males	0.315	N(0.315, 0.008)
	Females	0.287	N(0.287, 0.008)
35 – 44 yr	Males	0.324	N(0.324, 0.008)
	Females	0.273	N(0.273, 0.008)
45 – 64 yr	Males	0.289	N(0.289, 0.012)
	Females	0.245	N(0.245, 0.008)
> 65 yr	Males	0.148	N(0.148, 0.008)
	Females	0.114	N(0.114, 0.008)

Notes:
[1]Data sources: SAMHSA (1996) for children 12-17 years old; National Center for Health Statistics (1998) for adults.

Table 5. Input data on number of cigarettes smoked per day in residence by a smoker for various age groups[1].

Age Group	Range for Number of Cigarettes[2]	Male Proportion	Female Proportion
12 – 17 yr	0 - 2	0	0.14
	3 - 5	0.40	0.43
	6 - 9	0.10	0.14
	10 - 14	0.30	0.14
	15 - 24	0.20	0
	25+	0	0.14
18 – 24 yr	0 - 2	0.17	0.11
	3 - 5	0.33	0.30
	6 - 9	0.11	0.20
	10 - 14	0.24	0.23
	15 - 24	0.09	0.16
	25+	0.07	0
25 – 34 yr	0 - 2	0.09	0.06
	3 - 5	0.30	0.23
	6 - 9	0.17	0.19
	10 - 14	0.26	0.23
	15 - 24	0.17	0.23
	25+	0.01	0.06
35 – 44 yr	0 - 2	0.02	0.07
	3 - 5	0.22	0.28
	6 - 9	0.14	0.16
	10 - 14	0.25	0.27
	15 - 24	0.28	0.18
	25+	0.10	0.05
45 – 64 yr	0 - 2	0.05	0.03
	3 - 5	0.16	0.21
	6 - 9	0.13	0.15
	10 - 14	0.24	0.37
	15 - 24	0.32	0.16
	25+	0.11	0.08
≥ 65 yr	0 - 2	0.03	0.06
	3 - 5	0.10	0.21
	6 - 9	0.16	0.08
	10 - 14	0.19	0.38
	15 - 24	0.45	0.25
	25+	0.06	0.02

Notes:
[1]Data source: NHAPS questionnaire data (Klepeis et al., 1996; Tsang and Klepeis, 1996).
[2]Uniform distribution from min. to max. of range was used. The maximum number of cigarettes was 40.

Table 6. Input data on prevalence of others smoking in residence by smoking status, age group and gender[1].

Smoking Status	Age Group	Gender	Proportion With Other Smoker in Home	Uncertainty Distribution
Smoker	0 - 17 yr	Males	0.73	N(0.73, 0.035)
		Females	0.89	N(0.89, 0.045)
	18 - 64 yr	Males	0.35	N(0.35, 0.020)
		Females	0.38	N(0.38, 0.020)
	65+ yr	Males	0.16	N(0.16, 0.010)
		Females	0.28	N(0.28, 0.015)
Non-smoker	0 - 17 yr	Males	0.31	N(0.31, 0.015)
		Females	0.35	N(0.35, 0.020)
	18 - 64 yr	Males	0.10	N(0.10, 0.005)
		Females	0.12	N(0.12, 0.005)
	65+ yr	Males	0.04	N(0.04, 0.005)
		Females	0.05	N(0.05, 0.005)

Notes:
[1]Data source: NHAPS questionnaire data (Klepeis et al., 1996; Tsang and Klepeis, 1996).

Table 7. Input data on number of cigarettes smoked in residence by other smokers by smoking status and age group[1].

Smoking Status	Age Group	Range for Number of Cigarettes[2]	Male Proportion	Female Proportion
Smoker	12 – 17 yr	0 - 2	0.11	0
		3 - 5	0.11	0.25
		6 - 9	0.22	0.13
		10 - 14	0.12	0.37
		15 - 24	0.22	0.13
		25+	0.22	0.12
	18 – 64 yr	0 - 2	0.04	0.06
		3 - 5	0.14	0.16
		6 - 9	0.16	0.13
		10 - 14	0.33	0.24
		15 - 24	0.20	0.33
		25+	0.13	0.08
	≥ 65 yr	0 - 2	0.20	0.25
		3 - 5	0	0.25
		6 - 9	0	0
		10 - 14	0.20	0
		15 - 24	0.40	0.38
		25+	0.20	0.12
Non-smoker	12 – 17 yr	0 - 2	0.06	0.02
		3 - 5	0.16	0.20
		6 - 9	0.17	0.14
		10 - 14	0.24	0.22
		15 - 24	0.25	0.27
		25+	0.12	0.15
	18 – 64 yr	0 - 2	0.14	0.15
		3 - 5	0.19	0.31
		6 - 9	0.19	0.13
		10 - 14	0.22	0.19
		15 - 24	0.20	0.17
		25+	0.06	0.05
	≥ 65 yr	0 - 2	0.29	0.22
		3 - 5	0	0.28
		6 - 9	0.14	0.17
		10 - 14	0.28	0.11
		15 - 24	0.15	0.05
		25+	0.14	0.17

Notes:
[1]Data source: NHAPS questionnaire data (Klepeis et al., 1996; Tsang and Klepeis, 1996).
[2]Uniform distribution from min. to max. of range was used. The maximum number of cigarettes was 40.

Table 8. Distribution statistics for time (hours) spent per day in various microenvironments for the simulated population of Philadelphia.

Microenvironment	Percent of population (%)[1]	Mean (h)	Std. Dev. (h)	Percentiles 0	5	10	25	50	75	90	95	100
Outdoor	58	2.6	2.9	<0.1	0.1	0.2	0.6	1.5	3.5	6.6	8.8	24.0
Indoor residential	99	17.4	4.6	<0.1	9.7	11.7	14.3	17.7	21.2	23.4	24.0	24.0
Indoor office	14	3.7	3.7	<0.1	0.1	0.2	0.4	1.9	7.6	9.0	9.75	24.0
Indoor school	17	4.6	3.1	<0.1	0.1	0.3	1.5	5.3	7.0	8.1	9.0	16.6
Indoor store	34	1.5	1.8	<0.1	0.1	0.2	0.4	0.9	1.8	3.4	5.0	18.0
Indoor restaurant	22	1.3	1.6	<0.1	0.2	0.3	0.5	0.9	1.4	2.5	4.0	15.4
Indoor bar	3	2.6	2.0	<0.1	0.5	0.5	1.2	2.0	3.3	5.3	7.2	14.5
Indoor other	55	3.9	3.9	<0.1	0.2	0.5	1.1	2.5	5.6	9.3	11.4	24.0
In-vehicle	62	1.5	1.5	<0.1	0.2	0.3	0.6	1.1	1.9	3.0	4.0	22.6

Notes:
[1]Percent of population that spent time (at least 1 minute) in the microenvironment and were included in distribution statistics. For example, 42% of the population spent 0 minutes outdoors.

Table 9. Distribution statistics for total $PM_{2.5}$ exposures for the simulated population of Philadelphia.

	Mean	Std. Dev.	Percentiles 0	5	10	25	50	75	90	95	100
Total $PM_{2.5}$ Exposure ($\mu g/m^3$)											
- All	30.0	32.0	1.0	8.6	10.2	13.8	20.3	33.4	58.9	84.5	1259
- No ETS in residence	19.7	14.0	1.0	7.9	9.3	12.1	16.3	22.9	32.4	41.9	450
- From ambient $PM_{2.5}$	8.2	5.0	0.3	2.9	3.5	4.9	7.0	10.1	14.4	17.9	63
- From nonambient $PM_{2.5}$											
- All	21.9	32.0	0.0	2.5	3.5	6.0	11.2	24.4	50.5	76.6	1233
- No ETS in residence	11.6	13.5	0.0	2.1	2.9	4.7	7.8	13.5	23.0	32.7	443
Percent of Total $PM_{2.5}$ Exposure from Ambient (%)											
- All	39.6	23.1	0.1	6.5	10.2	20.2	37.5	56.8	72.6	80.5	100
- No ETS in residence	47.1	21.6	0.1	13.2	18.5	30.2	46.4	63.3	76.9	83.7	100
- By age group (yr):											
0 – 4	45.8	24.4	0.2	8.0	12.4	25.5	45.4	65.2	79.4	85.8	100
5 – 11	46.6	23.3	0.7	9.1	14.1	27.4	47.4	65.0	77.8	83.7	100
12 – 17	40.8	23.5	0.4	6.4	10.1	20.3	39.6	59.1	73.6	80.9	100
18 – 24	38.9	21.5	0.4	7.3	11.2	21.3	37.3	54.7	69.2	76.5	100
25 – 54	36.4	22.6	0.1	5.6	8.7	17.6	33.2	52.4	69.1	77.9	100
65+	42.8	23.0	0.2	8.3	12.8	24.1	41.2	60.1	75.4	82.8	100

Table 10. Distribution statistics for the apportionment of total $PM_{2.5}$ exposures by micro-environment and $PM_{2.5}$ source type for the simulated population of Philadelphia.

		Mean (%)	Std. Dev. (%)	Percentiles (%) 0	5	10	25	50	75	90	95	100
Percent of Total $PM_{2.5}$ Exposure:												
- All												
Outdoor		4.6	9.3	0	0	0	0	0.5	4.9	14.1	22.8	100
Residential	ambient	27.6	19.7	0	3.3	5.5	11.7	23.5	39.4	56.1	66.5	100
Non-residential	ambient	7.5	8.6	0	0	0	1.0	4.4	10.8	19.5	25.7	78.9
Residential	non-ambient	42.0	28.5	0	3.5	6.7	16.4	37.8	66.4	84.5	90.5	99.9
Non-residential	non-ambient	18.4	19.2	0	0	0	2.8	12.3	28.1	47.0	59.2	98.2
Total		100										
- No ETS in residence												
Outdoor		5.3	10.2	0	0	0	0	0.6	6.0	16.4	25.9	100
Residential	ambient	33.1	19.8	0	6.4	9.9	17.8	30.0	45.5	61.4	71.1	100
Non-residential	ambient	8.7	9.4	0	0	0	1.6	5.8	12.9	22.0	27.9	78.9
Residential	non-ambient	31.2	23.9	0	2.6	4.9	11.5	25.3	46.9	68.3	78.9	99.9
Non-residential	non-ambient	21.6	20.2	0	0	0	4.7	16.6	33.0	51.6	63.2	98.2
Total		100										

Figure captions:

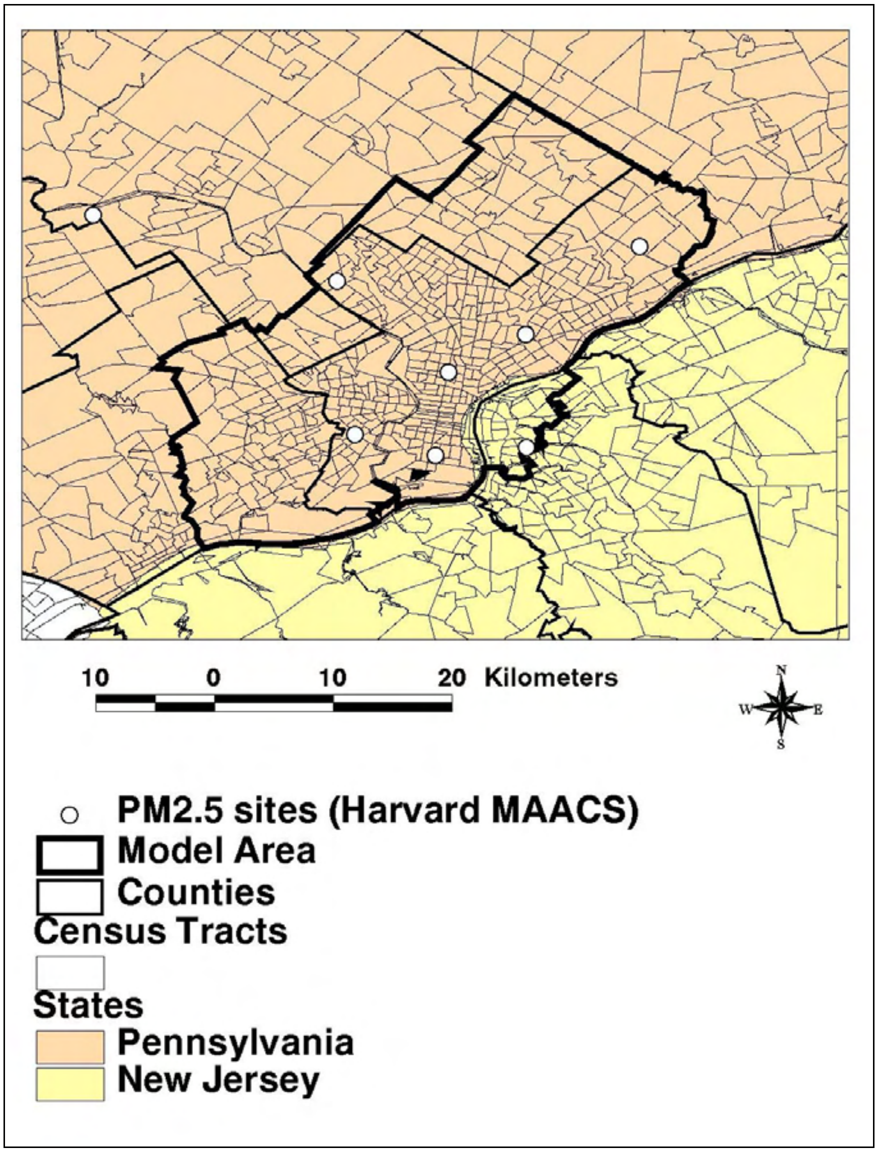
Figure 1. $PM_{2.5}$ measurement site locations, census tracts and defined model area within Philadelphia, PA for the SHEDS-PM model case study.

Figure 2. Cumulative frequency distributions of predicted daily average $PM_{2.5}$ concentrations ($\mu g/m^3$) for the 8 microenvironments included in the SHEDS-PM model case study for Philadelphia.

Figure 3. Cumulative frequency distributions of daily total and microenvironmental $PM_{2.5}$ exposures ($\mu g/m^3$) for the simulated population of Philadelphia (solid lines). Distributions of total and indoor residential $PM_{2.5}$ exposures for the population without environmental tobacco smoke (ETS) exposure in the residence (dashed lines) are also shown.

Figure 4. Horizontal box plots showing the uncertainty associated with selected percentiles of the predicted distribution of daily total $PM_{2.5}$ exposures for the simulated population of Philadelphia determined from 100 model iterations.

Figure 5. Cumulative frequency distributions of daily total, ambient and non-ambient $PM_{2.5}$ exposures ($\mu g/m^3$) for the simulated population of Philadelphia (solid lines). Distributions of daily total and non-ambient $PM_{2.5}$ exposures for the population without environmental tobacco smoke (ETS) exposure in the residence (dashed lines) are also shown.


10
0
10
20
Kilometers
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PM2.5 sites (Harvard MAACS)
Model Area
Counties
Census Tracts
States
Pennsylvania
New Jersey

Figure 1.

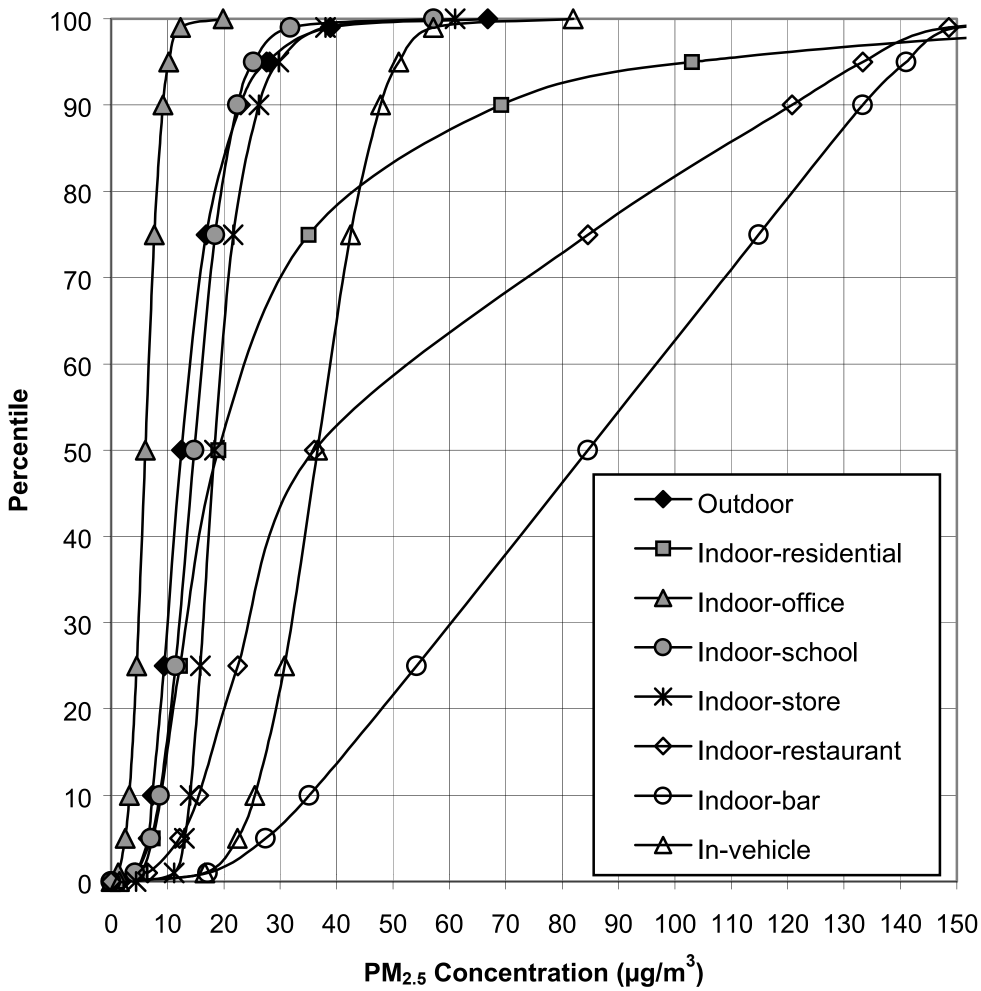
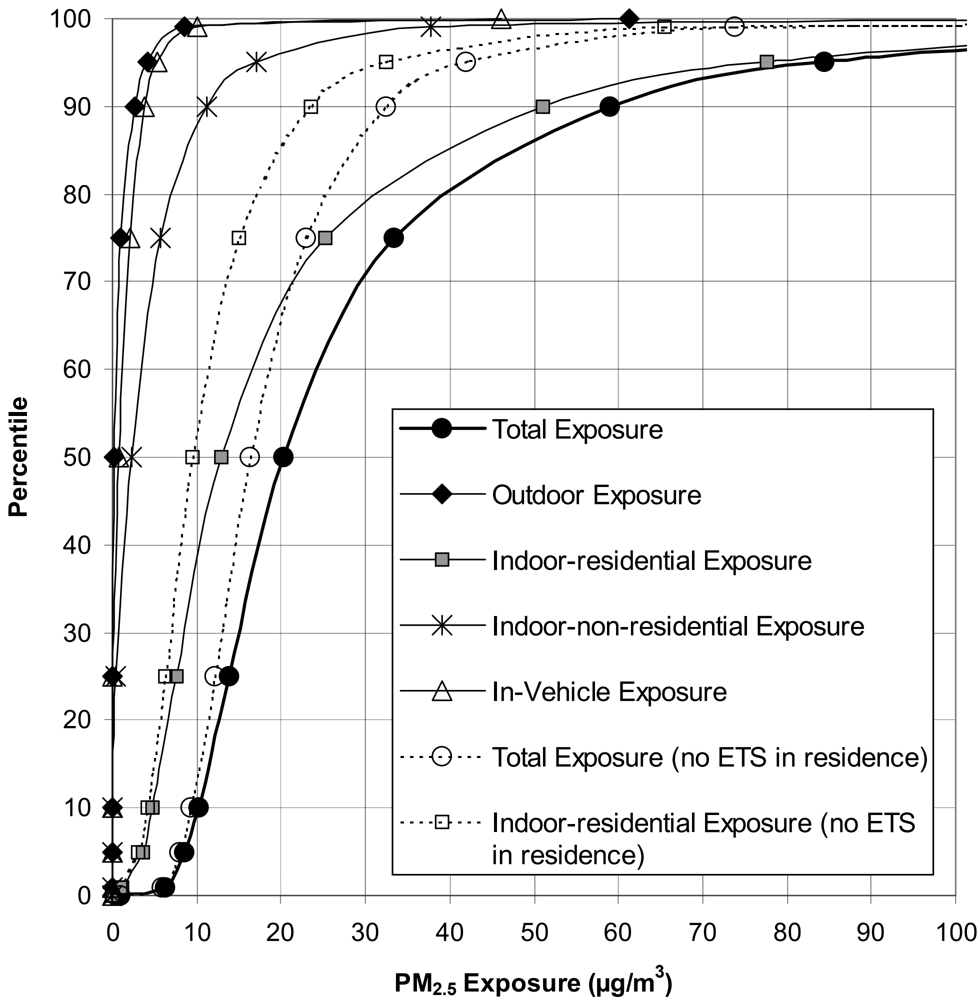

Percentile
PM2.5 Concentration (µg/m3)
Outdoor
Indoor-residential
Indoor-office
Indoor-school
Indoor-store
Indoor-restaurant
Indoor-bar
In-vehicle

Figure 2



100
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40
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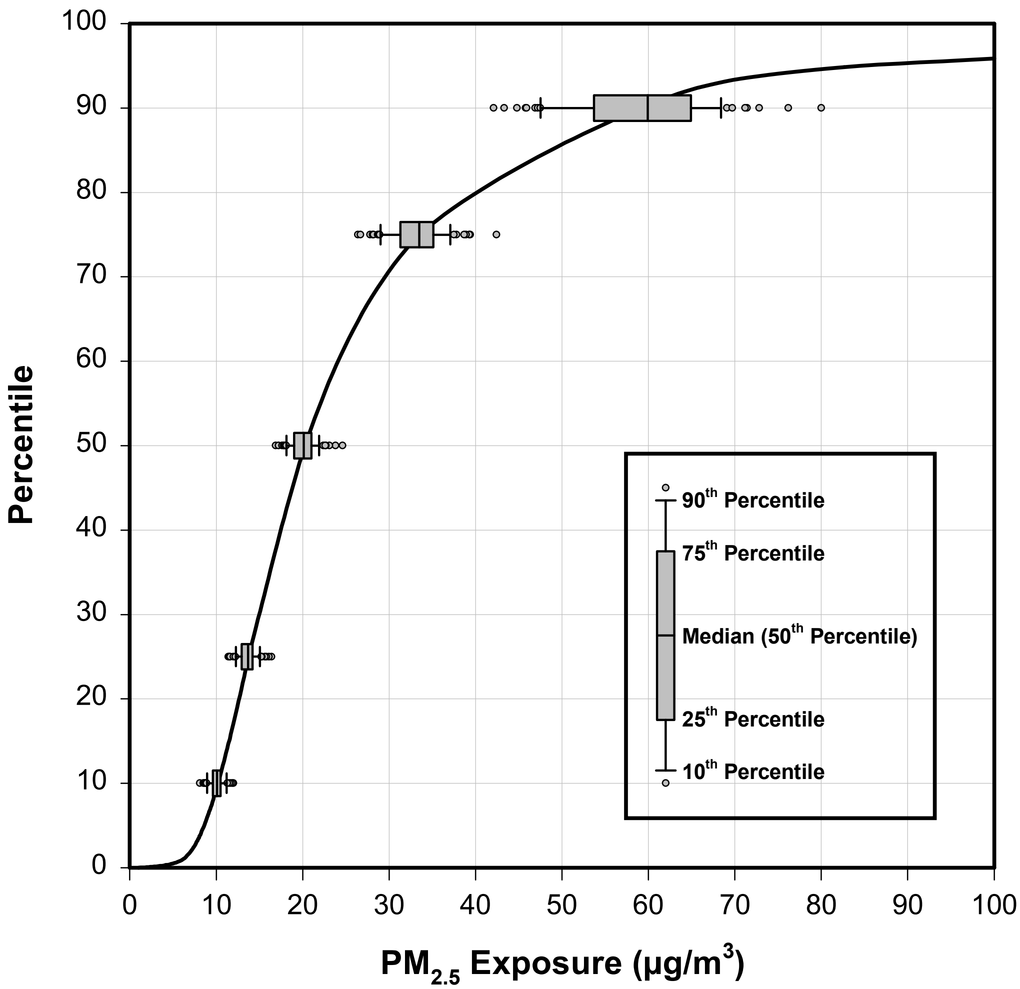
Percentile
0 10 20 30 40 50 60 70 80 90 100
PM2.5 Exposure (µg/m3)
Total Exposure
Outdoor Exposure
Indoor-residential Exposure
Indoor-non-residential Exposure
In-Vehicle Exposure
Total Exposure (no ETS in residence)
Indoor-residential Exposure (no ETS in residence)

Figure 3



100
90
80
70
60
50
40
30
20
10
0
Percentile
0
10
20
30
40
50
60
70
80
90
100
PM2.5 Exposure (µg/m3)
90th Percentile
75th Percentile
Median (50th Percentile)
25th Percentile
10th Percentile

Figure 4

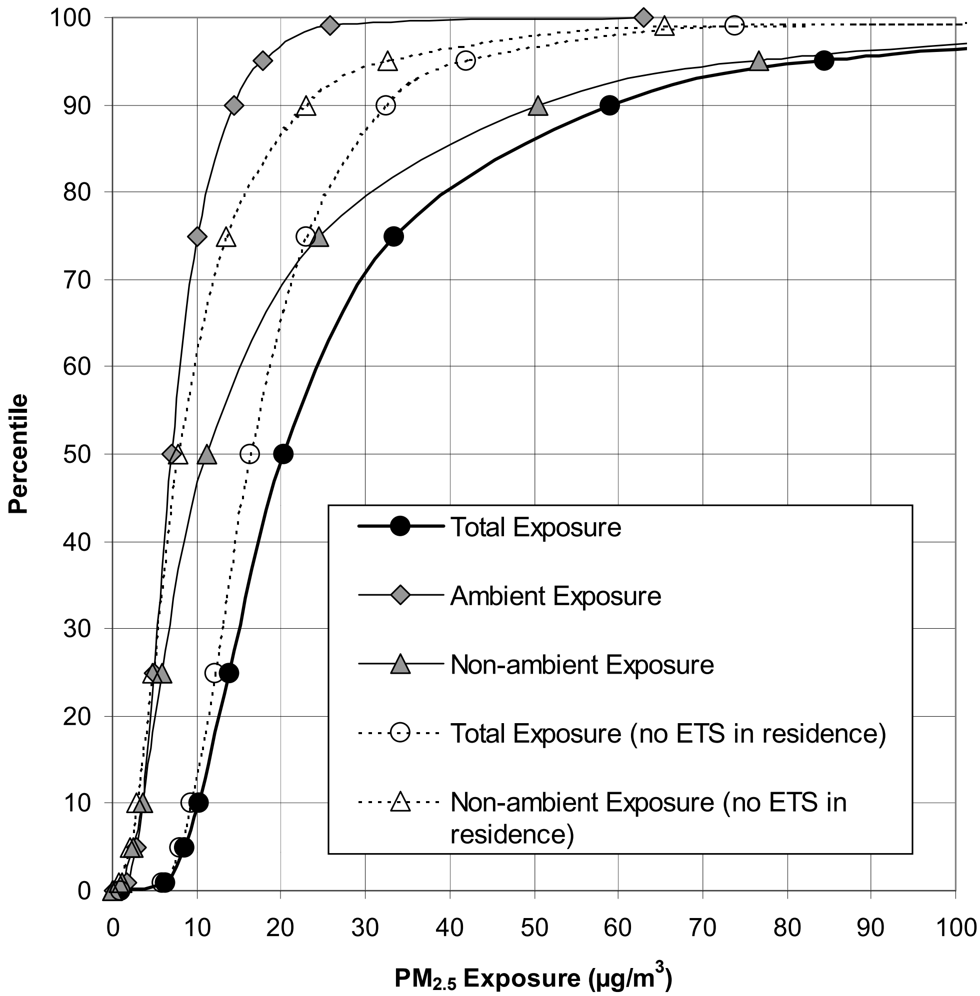

100
90
80
70
60
50
40
30
20
10
0
Percentile
0
10
20
30
40
50
60
70
80
90
100
PM2.5 Exposure (µg/m3)
Total Exposure
Ambient Exposure
Non-ambient Exposure
Total Exposure (no ETS in residence)
Non-ambient Exposure (no ETS in residence)

Figure 5