ABSTRACT

CAO, YE. Modeling of Human Exposure to Fine Particulate Matter Using a Stochastic Scenario-Based Model. (Under the direction of Dr. H. Christopher Frey).

Exposures to fine particulate matter ($PM_{2.5}$) are associated with adverse health effects. Inter-individual and geographic variability of exposure maybe associated with the health effects. Environmental tobacco smoke (ETS) is a major contributor to indoor human exposures to $PM_{2.5}$, especially for the residential and in-vehicle microenvironment. The Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM) model developed by the US Environmental Protection Agency estimates distributions of outdoor and indoor $PM_{2.5}$ exposure for a specified population based on ambient concentrations and indoor emissions sources. A critical assessment was conducted of the methodology and data used in SHEDS-PM for estimation of microenvironmental exposure to ETS. For the residential microenvironment, SHEDS uses a mass-balance approach which is comparable to best practices. Sensitivity analysis was used to determine which inputs should be prioritized for updating. Data regarding the proportion of smokers and "other smokers," and cigarette emission rate were found to be important. Geographic variability in ETS exposure was quantified based on the varying prevalence of smokers in five selected locations in the U.S.

For the in-vehicle microenvironment, a ETS mass balance model was incorporated into the SHEDS-PM model to quantify the potential magnitude and variability of in-vehicle exposures to ETS. The in-vehicle exposure also takes into account the near-road incremental $PM_{2.5}$. Air exchange rate (ACH) and the deposition rate have wider relative ranges of variation than other inputs, representing inter-

individual variability in operations, and inter-vehicle variability in performance, respectively. Results of probabilistic study indicate that ETS is a key contributor to the in-vehicle high-end exposure.

A case study of modeling exposure to $PM_{2.5}$ was conducted in New York City, Harris County in Texas, and Wake, Durham, Orange, Alamance, Guilford, and Forsyth County in North Carolina to represent different climate zones. Air exchange rate (ACH), penetration factor (P), and deposition rate (k) are the key factors affecting the fraction of ambient particles that penetrate indoors and remain suspended. Distributions of ACH, P and k are proposed in three geographic areas and seasons. Based on the sensitivity analysis, data regarding the ACH was found to be more important than P and k. Geographic and inter-individual variability in exposure are assessed in three locations using SHEDS-PM model. Geographic variability in average exposure is mainly caused by the difference of ambient air quality and ACH. Inter-individual variability in high-end exposure is mainly caused by indoor emission sources, such as smoking and cooking. The ratios of exposure to ambient concentration are significantly different from geographic areas.

Modeling of Human Exposure to Fine Particulate Matter Using a Stochastic Scenario-Based Model

by
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DEDICATION

This thesis is dedicated to my family for love and support.

BIOGRAPHY

Ye Cao was born on the 11th of September, 1985 at Shanghai, China. She obtained her Bachelor's Degree in Environmental Engineering from Qingdao Technological University, China in 2007. She worked as an environmental analyst in a venture capital firm in Shanghai, China from 2007 to 2008. In August 2008, she joined in the Department of Civil, Construction and Environmental Engineering at North Carolina State University, pursuing her Master of Science degree in Environmental Engineering. She was a research assistant in Dr. H. Christopher Frey's research group, and worked on a project of "A Spatial-Temporal Modeling Approach for Evaluating the Impact of Environmental Stressors, in Conjunction with Human Activity, on Human Health". This thesis completes the requirements for this degree.

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PART I

INTRODUCTION

1.0 INTRODUCTION

Particulate matter (PM) is an air pollution term from a mixture of solid particles and liquid droplets found in the air. The pollutant comes in a variety of sizes and can be composed of many types of materials and chemicals. Particles that are small enough to be inhaled have the potential to cause health effects. Of particular concern is a class of fine particulate matter ($PM_{2.5}$) that are 2.5 microns or less in aerodynamic diameter which gets deep into the lung.

Exposure is defined as the frequency and duration of contact between an agent and a target, with contact taking place at a contact boundary over an exposure period (EPA, 1992). Exposure to air pollutants via the inhalation pathway is often quantified as a time weighted average concentration. Epidemiology studies have found association between the exposure to $PM_{2.5}$ with adverse health outcomes, such as irritation of the airways, reduced lung function, and aggravated asthma (EPA, 2004).

Total personal exposure to $PM_{2.5}$ includes both indoor and ambient exposures. Air pollution epidemiology and exposure studies have identified Environmental Tobacco Smoke (ETS) as a major contributor to indoor air concentrations and human exposure to $PM_{2.5}$ (Wallace, 1996; Gilmour *et al.,* 2006). Smoking is associated with significantly increased risk of heart disease, stroke, lung and chronic lung diseases (Vineis *et al.,* 2005). Exposure to second-hand smoke by children is associated with reduced cognitive ability, and increased risk of serious respiratory problems and middle ear infections (DiFranza *et* *al.,* 2004; Yolton *et al.,* 2005). Therefore, it is necessary to account for the contribution of smoking to indoor $PM_{2.5}$ when estimating total exposures to $PM_{2.5}$.

Americans are estimated to spend 87% of their time indoors, 8% outdoors and 5% in vehicles such as automobiles, buses, vans, and trucks (Klepeis *et al.,* 2001). Because of the variability in air exchange rate (ACH), deposition rate (k), and small interior vehicle volume, smoking in a vehicle can potentially expose commuters and passengers to high concentrations of $PM_{2.5}$. Therefore, it is necessary to account for the contribution of smoking to in-vehicle $PM_{2.5}$ when estimating total exposures to $PM_{2.5.}$

People spend 62 to 87 percent their time in the residential microenvironment, which is critical for the daily total personal exposure of the population (Adgate *et al*., 2002; Klepeis *et al.,* 1996). A microenvironment is a physical compartment or defined space with relatively homogeneous or well characterized air pollutant concentrations (Ott *et al.,* 1992). Individual exposures to $PM_{2.5}$ occur both outdoor and indoor, and in door $PM_{2.5}$ concentrations are affected by penetrations of ambient $PM_{2.5}$ and exposures from indoor sources, such as cooking, cleaning and smoking.

Critical parameters influencing the penetrations of outdoor particles into the indoor microenvironment and remain suspended are ACH, penetration factor (P), and k (Wilson *et al.,* 2000). ACH is measured by a tracer gas, such as perfluorocarbon tracer (PFT) or sulfur hexafluoride (SF_6). P and k are difficult to measure directly, but can be estimated by fitting a mass balance model to data for paired indoor and outdoor concentration and ACH. Few observational data are available on the levels of seasonal and geographic variability of ACH, P, and k within residential home. Therefore, it is important to review and recommend values of ACH, P, and k relevant to different geographic areas.

In recent epidemiology studies, associations between exposure to $PM_{2.5}$ and health effects are quantified by the response-concentration functions in multicity studies, however exposure is not measured or estimated (EPA, 2009). These studies assumed concentration is a surrogate for exposure, but did not address whether the ratio of exposure to concentration is similar for different locations. The exposure to concentration ratio is approximately independent of concentration for ambient sources of exposure, and varies geographically depending on demographics and housing stock. Therefore, it is necessary to identify the exposure to concentration ratio in different geographic areas.

Two approaches for exposure studies are measurement and mathematical modeling. Direct measurement is the most accurate way to determine air quality, when conducted for representative samples of different microenvironments and activity patterns, but it is time consuming and costly (Wallace 1987; Thomas *et al.,* 1993; Sexton *et al.,* 1995; Jenkins *et al.,* 1996). A scenario-based inhalation exposure simulation model is intended to estimate exposures to simulated individuals by estimating the movement of such individuals through a series of microenvironments, each with its own air pollutant concentration (Ott *et al.,* 1986). The exposure of an individual during a day is based on the time-weighted concentration from the microenvironments in which the individual spent time.

The Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM) Model was developed by the US Environmental Protection Agency (EPA) (Burke *et al.*, 2001). SHEDS-PM uses a probabilistic approach to estimate distributions of outdoor and indoor $PM_{2.5}$ exposure for a population of simulated individuals based on ambient $PM_{2.5}$ concentrations and sources of indoor $PM_{2.5}$ emissions.

2.0 OBJECTIVES

The objectives of this study are to:

(1) Characterize the inter-individual and geographic variability in human exposure to $PM_{2.5}$ in ETS.

(2) Characterize the potential magnitude and variability of in-vehicle exposures to ETS.

(3) Characterize the inter-individual, geographic, and seasonal variability in daily average exposure to $PM_{2.5}$.

3.0 OVERVIEW OF RESEARCH

This section provides an overview of the research scope aimed to achieve the main objectives. In general, four principal components will be investigated:

(1) Assessment of the algorithms and inputs in the SHEDS-PM Model regarding the ETS

Currently, SHEDS accounts for ETS exposure for home, restaurant, and bar microenvironments. The algorithms and inputs regarding ETS in these microenvironments and the in-vehicle microenvironment are updated based on literature review. Sensitivity analysis is conducted to identify the key factors to which exposure is sensitive for ETS in selected microenvironments.

(2) Development of the key inputs in the residential microenvironment

Distributions of ACH, P, and k are recommended for different seasons and geographic areas based on the critical literature review. Sensitivity analysis is conducted to evaluate their importance.

(3) Inter-individual Variability in Exposure to $PM_{2.5}$

For the ETS exposure in the residential microenvironment, inter-individual variability in exposures are estimated and compared for specific sources of $PM_{2.5}$ including: (a) infiltration of outdoor air; (b) indoor sources other than smoking; and (c) ETS from smoking.

For the ETS exposure in the vehicle, inter-individual variability in exposures are estimated and compared for various vehicle operating scenarios, with different vehicle speeds, status of windows and the operations of Heating, Ventilating, and Air Conditioning (HVAC) system.

For the daily average exposures in different geographic areas, inter-individual variability in exposures is characterized based on the ratios of estimated exposure to ambient concentrations.

(4) Geographic Variability in Exposure to $PM_{2.5}$

In order to assess the geographic variability in estimated exposure to ETS, five locations were selected as a basis for comparisons from U.S. states that span the lowest to highest range of smoking prevalence.

For the daily average exposure to $PM_{2.5}$, three locations are selected that represent different climate zones in the nation. Exposure to concentration ratios are quantified for three geographic areas.

4.0 ORGANIZATION

This thesis consists of five parts, in which three journal manuscripts are included in the main body. Appendices are given at the end of the document. Each part of the dissertation contains a separate reference list.

Part I introduces the background information regarding the importance of ETS exposure in some selected microenvironments, the SHEDS-PM model, research objectives and tasks, and thesis organization.

Part II updates the SHEDS inputs regarding ETS in the selected microenvironment, and assesses the inter-individual and geographic variability in human exposure to $PM_{2.5}$ in ETS.

Part III focuses on the modeling of human exposure to in-vehicle $PM_{2.5}$ from ETS. Inter-individual and intra-vehicle variability in exposure to ETS are quantified based on both point estimates and probabilistic studies.

Part IV develops the distributions of key inputs for the residential microenvironment in the SHEDS, for different geographic areas. Inter-individual and geographic variability in daily average exposure to $PM_{2.5}$ are quantified.

Part V summarizes findings, main conclusions and recommendations of this research.

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PART II

ASSESSMENT OF INTER-INDIVIDUAL AND GEOGRAPHIC VARIABILITY IN HUMAN EXPOSURE TO FINE PARTICULATE MATTER IN ENVIRONMENTAL TOBACCO SMOKE

ABSTRACT

Environmental tobacco smoke (ETS) is a major contributor to indoor human exposures to fine particulate matter of 2.5 microns or smaller ($PM_{2.5}$). The Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM) model developed by the US Environmental Protection Agency estimates distributions of outdoor and indoor $PM_{2.5}$ exposure for a specified population based on ambient concentrations and indoor emissions sources. A critical assessment was conducted of the methodology and data used in SHEDS-PM for estimation of indoor exposure to ETS. For the residential microenvironment, SHEDS uses a mass-balance approach which is comparable to best practices. The default inputs in SHEDS-PM were reviewed and more recent and extensive data sources were identified. Sensitivity analysis was used to determine which inputs should be prioritized for updating. Data regarding the proportion of smokers and "other smokers," and cigarette emission rate were found to be important. SHEDS-PM does not currently account for in-vehicle ETS exposure; however, in-vehicle ETS-related $PM_{2.5}$ levels can exceed those in residential microenvironments by a factor of 10 or more. Therefore, a mass-balance based methodology for estimating in-vehicle ETS $PM_{2.5}$ concentration is evaluated. Recommendations are made regarding updating of input data and algorithms related to ETS exposure in the SHEDS-PM model. Inter-individual variability for ETS exposure was quantified. Geographic variability in ETS exposure was quantified based on the varying prevalence of smokers in five selected locations in the U.S.

1.0 INTRODUCTION

Epidemiological studies of health effects associated with $PM_{2.5}$ typically use ambient concentration as a surrogate for human exposures (Spengler *et al.*, 1998; Lipfert and Wyzga, 1997; Gamble, 1998). Therefore, health effects are often estimated based on concentration-response (C-R) relationships derived from such studies (Pope *et al.,* 2002; Cifuentes *et al.*, 2001; Künzli *et al.,* 2000). However, because most people spend the majority of their time indoors, the use of ambient data does not accurately represent the concentrations to which people are actually exposed. Hence, there is growing recognition of the need to quantify human exposure to $PM_{2.5}$ as an alternative basis for characterizing associated health effects (EPA, 2008).

Total personal exposure to $PM_{2.5}$, including both indoor and ambient exposures, is significantly associated with daily mortality (Calder *et al.*, 2008). Air pollution epidemiology and exposure studies have identified Environmental Tobacco Smoke (ETS) as a major contributor to indoor air concentrations and human exposure to $PM_{2.5}$ (Wallace, 1996; Gilmour *et al.*, 2006). Smoking is associated with significantly increased risk of heart disease, stroke, lung and chronic lung diseases (Kawachi *et al.*, 1997; Bonita *et al.*, 1999; Vineis *et al.,* 2005). Exposure to second-hand smoke by children is associated with reduced cognitive ability, and increased risk of serious respiratory problems and middle ear infections (Yolton *et al.,* 2005; DiFranza *et al.*, 2004; Etzel *et al.*, 1992) Therefore, it is necessary to account for the contribution of smoking to indoor $PM_{2.5}$ when estimating total exposures to $PM_{2.5}$.

A scenario-based inhalation exposure simulation model is intended to estimate exposures to simulated individuals by estimating the movement of such individuals through a series of microenvironments, each with its own air pollutant concentration (Ott *et al.,* 1986). The exposure of an individual during a day is based on the time-weighted concentration from the microenvironments in which the individual spent time. Examples of such models that incorporate ETS are the Simulation of Human Activity and Pollutant Exposure (SHAPE) model, Total Human Exposure Model (THEM), Air Pollution Exposure (APEX) model, and Stochastic Human Exposure and Dose Simulation model for Particulate Matter (SHEDS-PM) (Ott *et al.,* 1984; Klepeis *et al.,* 1994; Richmond *et al.,* 2002; Burke, 2005)

The objective of SHEDS-PM is to predict total personal exposures to $PM_{2.5}$. SHEDS-PM uses a probabilistic approach to estimate inter-individual variability in distributions of outdoor and indoor $PM_{2.5}$ exposure for a population of simulated individuals based on ambient $PM_{2.5}$ concentrations and sources of indoor $PM_{2.5}$ emissions. Currently, SHEDS accounts for ETS exposure for home, restaurant, and bar microenvironments.

The objectives of this paper are to answer four key questions:

- What are the spatial and temporal trends in factors affecting ETS exposure?
- What are the key factors to which exposure is sensitive for ETS in different microenvironments?
- What are the key factors leading to geographic and inter-individual

variability in ETS exposure?

2.0 MODELING OF ENVIRONMENTAL TOBACCO SMOKE EXPOSURE

Figure 1 illustrates the main inputs and key algorithms in SHEDS-PM for calculating indoor $PM_{2.5}$ concentrations contributable to ETS. Input data include demographic data, ambient $PM_{2.5}$ concentration, and human activity data. The demographic data used in SHEDS-PM were obtained from the US Census for the year 2000. The daily average ambient $PM_{2.5}$ concentration for each census tract for the geographic area of interest is input by the user based on ambient monitoring or air quality modeling data. The Consolidated Human Activity Database (CHAD) is comprised of U.S. human activity pattern diary data compiled based on a variety of activity studies (Klepeis et al., 1996; Johnson, 1984, 1989; Settergren et al., 1984; Wiley, 1991).

SHEDS-PM selects the US Census data for user specified census tracts and randomly generates demographically representative individuals by age and gender. The number of individuals simulated, and the distribution of age and gender, is specified by the user. Each simulated individual is randomly assigned an activity diary record from CHAD based on age and gender and other user specified matching criteria (e.g., housing type, employment status, and smoking status). A cross-sectional simulation is based on a different random sample of individuals each day, whereas a longitudinal simulation is based on one random set of individuals each of whose activity pattern is simulated from day-to-day, thereby taking into account daily dependence in activities (Burke et al., 2001). All simulations reported here are based on longitudinal simulation.

For the residential microenvironment, ETS-related inputs include the cigarette emission rate, proportions of smokers and "other smokers," and the number of cigarettes smoked. Emissions from cigarette smoking include: (1) emissions from smoking by someone who is a smoker; and (2) emissions to which a non-smoker is exposed because of smoking by others, who are referred to as "other smokers." These support assessments of ETS-based $PM_{2.5}$ exposures for smokers and non-smokers, respectively (Burke, 2005).

In the restaurant and bar microenvironments, the ETS-related inputs are Active Smoking Count (ASC) and the average incremental increase in indoor $PM_{2.5}$ concentration caused by smoking one cigarette smoking (C_{ets}). ASC is the average number of cigarettes actively smoked during a defined time interval (Ott *et al.*, 1996).

A mass balance approach is applied in the residential microenvironment based on the assumption of a single steady-state zone, and on parameters for penetration of outdoor $PM_{2.5}$, air exchange rate, deposition rate, and indoor volume. The assumption is not strictly satisfied in most cases, however, in many situations the equation provides good estimates (Nazaroff and Klepeis, 2003; Ott *et al.*, 1992; Özkaynak *et al.*, 1996a, 1996b). Exposure events are simulated for exposure time periods of typically minutes to hours, according to the duration of time spent in a microenvironment per diary sampled from CHAD.

SHEDS-PM estimates the indoor $PM_{2.5}$ concentration including ETS but does not estimate direct inhalation by a smoker from active smoking. $PM_{2.5}$ concentrations in the residential microenvironment are estimated by a mass balance (Özkaynak *et al.*, 1996a, 1996b):

$$C_{Home} = \frac{P \cdot a}{a+k} C_{ambient} + \frac{E_{cig}\ N_{cig} + E_{cook}\ t_{cook} + E_{clean}\ t_{clean} + E_{other}\ t_{other}}{(a+k)VT} \quad (1)$$

Where,

a	= air exchange rate (h^{-1});
C_{Home}	= $PM_{2.5}$ concentration in the home (µg/m^3);
$C_{ambient}$	= ambient outdoor $PM_{2.5}$ concentration (µg/m^3);
E_{cig}	= emission rate for cigarette smoking (µg/cig);
E_{cook}	= emission rate for cooking (mg/m^3);
E_{clean}	= emission rate for cleaning (mg/m^3);
E_{other}	= emission rate for all other activities (mg/m^3);
k	= deposition rate (h^{-1});
N_{cig}	= number of cigarettes smoked during model time step (cig);
P	= penetration factor (unitless);
T	= model time step (min);
t_{cook}	= duration of time spent cooking during model time step (min);

t_{clean} = duration of time spent cleaning during model time step (min);

t_{other} = duration of time spent doing other activities during model time step (min); and

V = volume of microenvironment (m^3).

The indoor $PM_{2.5}$ concentration attributable to penetration of ambient $PM_{2.5}$ is estimated based on a penetration factor, deposition rate, air exchange rate, and indoor volume. The second term in Equation (1) describes the contribution from indoor emission sources, including smoking, cooking, cleaning, and other sources.

Parameter values for Equation (1) can be assigned fixed quantities or frequency distributions. The model time step (T) is the duration of a diary event. The emission generating durations (t_{cook}, t_{clean}, t_{other}) are obtained from the CHAD database for each simulated individual. Several steps are used to calculate the number of cigarettes (N_{cig}) smoked. The daily total numbers of cigarettes smoked in the residence are assigned to an individual who is a smoker or other smoker, based on the user-specified proportions for the number of cigarettes smoked by smokers and by others, respectively. The rate of smoking in the home is based on the number of cigarettes smoked at home divided by time spent at home while not sleeping. For each diary event at home, the hourly rate is multiplied by the duration of the event in hours to estimate an average number of cigarettes (Burke *et al.*, 2001)

Due to lack of data needed to apply Equation (1) to the restaurant and bar microenvironments, a simplified approach is used instead. The simplified approach is based ona linear regression to estimate indoor concentration based on outdoor concentration, incremental impact on indoor air quality from cigarette smoking, and indoor background concentration. The indoor $PM_{2.5}$ concentration for the restaurant and bar microenvironments is (Ott *et al.*, 1996):

$$C_{rest/bar} = B + r_{I/O} \cdot C_{ambient} + ASC \cdot C_{ets} \qquad (2)$$

Where,

ASC = active smoking count, the average number of cigarettes being actively smoked in the microenvironment in a defined time interval (cig);

B = background indoor $PM_{2.5}$ concentration from indoor $PM_{2.5}$ sources (µg/m^3);

$C_{rest/bar}$ = $PM_{2.5}$ concentration for of restaurant or bar microenvironment (µg/m^3);

$C_{ambient}$ = ambient outdoor $PM_{2.5}$ concentration (µg/m^3);

C_{ets} = incremental $PM_{2.5}$ concentration caused by smoking a cigarette during a defined time interval [µg/(m^3.cig)]; and

$r_{I/O}$ = ratio of indoor concentration associated with penetration of outdoor concentration.

The first term of Equation (2) describes the non-ambient contribution to indoor $PM_{2.5}$ concentration except for ETS. The second term describes the contribution from outdoor $PM_{2.5}$. The $PM_{2.5}$ concentration attributable to ETS is described by the last term.

3.0 METHODOLOGY

The methodology includes: (1) review literature for ETS data and algorithms used in SHEDS-PM; (2) sensitivity analysis to identify the key factors to which exposure is sensitive for ETS in selected microenvironments; (3) assessment of the effect of updated data on estimated exposures; and (4) characterization of inter-individual and geographical variability associated with ETS exposure.

3.1 Review of Inputs and Algorithms

The review of existing inputs and algorithms in SHEDS-PM for estimating $PM_{2.5}$ concentration associated with ETS is based on: (a) detailed review of the SHEDS-PM model, its user guide, and the literature cited as the basis for default input assumptions; (b) published peer reviewed papers regarding SHEDS-PM and similar models; and (c) databases of the U.S. Department of Health and Human Services (DHHS) and the Substance Abuse & Mental Health Services Administration (SAMHSA).

3.2 Sensitivity Analysis

An overview of sensitivity analysis methods is given by Frey and Patil (2002). Differential Sensitivity Analysis (DSA) is applicable to linear models for which there are no nonlinear interactions between terms, as is the case for the ETS aspects of SHEDS-PM. DSA evaluates the effect on model outputs exerted by individually perturbing only one of the model inputs, while holding all other inputs at their nominal or base-case values (Cullen and Frey, 1999). Sensitivity in a model output is represented as a positive or negative percentage change compared to the nominal solution. This type of sensitivity analysis provides a measure of model responsiveness to a unit change in an input. In separate analyses, inter-individual variability in exposure is estimated based on simultaneous variation in multiple model inputs over their plausible ranges of variability.

In the DSA, selected inputs were varied by plus or minus 10 percent, which is well within the plausible range of values. In subsequent probabilistic simulations, these inputs are varied over plausible ranges. The differential sensitivity of estimated exposure for selected indoor microenvironments is based on the time-weighted daily average $PM_{2.5}$ exposures for the 50^{th}, 90^{th}, and 99^{th} percentile of simulated individuals. Because only a small fraction of the simulated population spent time in the restaurant and bar microenvironments, the sensitivity analysis for these microenvironments focuses on the 99^{th} percentile.

3.3 Assessment of Updated Inputs and Algorithms

The assessment of the effect of updated input data on estimated exposures are based on running SHEDS-PM with default data, with updated data for ETS-related inputs, and comparison of the two sets of results. Ambient $PM_{2.5}$ air quality data, demographic data, sample size, and algorithms are kept the same in both sets of simulations. Frequency distributions of air exchange rate, penetration factor, and deposition rate are used for the residential microenvironment. Vehicle air exchange rates have much more variability than those in a residential microenvironment. Therefore, a mass balance model for estimating in-vehicle microenvironment $PM_{2.5}$ concentration is separately evaluated based on sensitivity to air exchange rates.

3.4 Inter-individual Variability in Residential Exposures

To explore variability in exposure in the residential microenvironment, exposures are estimated and compared for specific sources of $PM_{2.5}$ including: (a) infiltration of outdoor air; (b) indoor sources other than smoking; and (c) ETS from smoking. Updated data regarding the proportion of smokers and "other" smokers, and cigarette emission rate, are used in the simulation.

3.5 Geographic Variability in Exposures

Smoking prevalence, housing types, and demographic factors (i.e. age, gender) vary among geographic areas. In order to assess the geographic variability in estimated exposure, five locations were selected as a basis for comparisons from U.S. states that span the lowest to highest range of smoking prevalence. Utah has only 9.3 percent proportion of smokers compared to Kentucky, with 25.2 percent, based on 2008 data. California (14.0%), New York (16.8%), and North Carolina (20.9%) are examples of varying proportions between the lowest and highest (CDC, 2009). For each state, the county with the highest population was selected as the basis for case studies, with an assumption that the county and state smoking prevalence are the same. For each county, ten census tracts were selected at random. Two case studies were conducted for each area: (1) base case without ETS; and (2) with ETS. Updated data were used for E_{cig}, ASC, and C_{ets}. To focus on the role of ETS without confounding effects of differences in actual ambient $PM_{2.5}$ concentrations, the daily average ambient $PM_{2.5}$ concentration for each census tract was assigned the same constant value of 10 g/m^3 over space and time for each area, and analysis of results focused on the incremental contribution of ETS to daily exposure.

4.0 RESULTS

Results for updated inputs and algorithms are given. An algorithm for ETS exposure in the in-vehicle microenvironment is evaluated using sensitivity analysis. Inter-individual and geographic variability in ETS-related exposure is evaluated.

4.1 Evaluation of ETS-Related Input Data

The default data in SHEDS related to smoking prevalence is representative of 1995 and 1993, for adults 18 years and older and adolescents from 12 to 17 years old, respectively. More recently available U.S. data, representative of 2007 and 2006 for adults and adolescents, respectively, on the proportion of smokers at home is lower than that of the default data, especially for adolescents aged from 12-17, because of declining trends in smoking prevalence. From 2000 to 2007, the U.S. nationwide prevalence of smoking decreased from 23.2 to 18.4% (DHHS, 2008). The default and updated inputs are compared in Table 1.

There are no recent data regarding the proportion of "other smokers" by gender. However, updated data are available for age categories. SHEDS takes into account other smokers for persons older than 12 years old for three age groups. The default proportion of persons exposed to other smokers varies from 4 to 49 percent depending on the age group. However, SHEDS does not currently estimate exposures of children 11 years of younger to other smokers. An estimated 24.9% of children in this age group are exposed to cigarette smoking at home (DHHS, 2007).

The available data regarding smoking prevalence are on an individual basis (DHHS, 2008). However, there are no available data on the proportion of residences in which smoking occurs.

The default input for the cigarette emission rate in SHEDS-PM is 10.9 mg per cigarette. Özkaynak *et al*. (1996a) estimated a cigarette emission rate of 14 ± 4 mg/cig by fitting a nonlinear regression model to average $PM_{2.5}$ concentrations for 178 homes in Riverside, CA. The mean value of $PM_{2.5}$ emission rate among the 50 top brands of cigarettes is 13.8 mg/cig, with a standard deviation of 3.1 mg/cig and a range of 8 to 23 mg/cig based on a sample size of 111 (Nelson, 1994). Based on a summary of 14 papers, Nazaroff and Klepeis (2003) reported a mean $PM_{2.5}$ emission rate of 13.7 mg/cig. Thus, the default input is lower than the mean value of cigarette emission rate based on various studies. An updated emission rate of approximately 13.8 mg/cig, which is 27 percent higher than the default, with a range from 8 to 23 mg/cig, is used here.

In 1993, an average smoker smoked 19.6 cigarettes per day (cpd), with a mean of 21.3 cpd for men and 17.8 cpd for women. In 2004, the mean was 16.8 cpd, with 18.1 cpd for men and 15.3 cpd for women (MMWR, 2005). Thus, over an 11 year period, the number of cigarettes smoked declined on average by 14 percent for women and 15 percent for men. However, updated data are not available for specific gender and age cohorts. Therefore, the default inputs used in SHEDS-PM based on NHAPS are retained. However, to assess the implications of possible reductions in cpd, a sensitivity case was conducted using the example of Wake County in which a 15 percent reduction was applied for all cohorts.

As a default, ASC for the restaurant and bar microenvironments has a uniform distribution of 0 to 3. ASC ranged from 0 to 4 cigarettes per hour with an average of 1.3 cigarettes per hour based on 1993 to 1994 data (Ott *et al.*, 1996). Assuming 16 hours of smoking per day, the ASC in 2004 is approximately 1.05 cigarettes per hour per smoker, based on 2004 data. The value of ASC appears to be decreasing with time.

The SHEDS-PM default for C_{ets} is a triangular distribution with a minimum of 32 $\mu g/m^3$, best estimate or mode of 40.4 $\mu g/m^3$, and a maximum of 50 $\mu g/m^3$ per cigarette per hour for both restaurants and bars. Ott *et al.* used a mass balance approach to estimate an incremental $PM_{2.5}$ concentration in a tavern of 42.5 $\mu g/m^3$ based on a cigarette emission rate of 2.4 mg/min, and an ASC of 1.17 cigarettes (Ott *et al.*, 1996). Based on the range of cigarette emission rate from 8 to 23 mg/cig, and assuming the duration of one cigarette smoking is 10 minutes, the estimated C_{ets} ranges from 14 to 40 $\mu g/m^3$, which overlaps with the range of defaults used in SHEDS-PM.

Based on CHAD data, the time people spend in a vehicle is almost 10 times less than that at home. However, in-vehicle $PM_{2.5}$ concentrations associated with smoking have been measured or estimated to be as high as 658 $\mu g/m^3$, depending on the status of vehicle windows and air conditioning system (Ott *et al.*, 2008). Vehicles have a wider range of air exchange rates compared to those measured in homes. The relatively high ETS $PM_{2.5}$ concentrations inside a vehicle can be attributed to the smaller interior volume (Ott *et al.*, 2008).

4.2 Sensitivity Analysis

To compare the sensitivity of estimated exposure to each of several ETS-related inputs, a typical case study was developed based on ten randomly selected census tracts in Wake

County, North Carolina. For each census tract, 10,000 individuals were simulated. For each of the residential, restaurant and bar microenvironments, sensitivity analysis was conducted based on default inputs.

Seven inputs were varied, including: (1) proportions of smokers and other smokers, cigarette emission rate, and number of cigarettes, for the residential microenvironment; and (2) ASC and C_{ets} for the restaurant and bar microenvironments. One simulation was conducted for default inputs in all three microenvironments, and 14 simulations were conducted for the upper and lower bound of each of the 7 inputs except the number of cigarette. One sensitivity case was conducted for the number cigarettes smoked per day based on 15% reduction of the default inputs. Each model run was conducted on a Windows XP Pentium 4 computer and had an approximate runtime of 400 minutes. The results for the residential microenvironment are given in Table 2. The results for restaurants and bars are given in Table 3.

Over 99 percent of the simulated population spent time in the residential microenvironment. Based on the default inputs, about 40 percent of people were estimated to be exposed to ETS. Because ETS concentration is a linear function of proportion of smokers and cigarette emission rate, therefore, the 90th and 99th percentiles of inter-individual variability in exposure vary plus or minus 10 percent for a plus or minus 10 percent variation in proportion of smokers or cigarette emission rate. For an estimated 15 percent decrease in the average cpd over time, the 90th and 99th percentiles of exposure also decrease by 15 percent. The 50th percentile of exposure is less sensitive to ETS-related inputs.

Only 22 and 4 percent of the simulated population spent time in the restaurant and bar microenvironments, respectively. According to Equation (2), the concentrations inside the restaurant and bar are linearly proportion to the product of ASC × C_{ets}. Therefore, the exposure at the 99th percentile is equally sensitive to relative variations of plus or minus 10 percent to either of these two inputs.

4.3 Assessment of the Effect of Updated Inputs and Algorithms

Comparison of estimated daily average exposures to individuals based on updated data to that based on default data is given in Table 4. For the residential microenvironment inputs, a lognormal distribution was used for air exchange rate, with geometric mean of 0.56 and geometric standard deviation of 1.84. Normal distributions were used for penetration factor and deposition rate, with means of 0.91 and 0.79, standard deviation of 0.1 and 0.31, respectively. The updated data include circa 2006 to 2007 proportions of smokers and nonsmokers, with E_{cig} = 13.8 μg/m^3, versus default circa 1995 data on the proportion of smokers and non-smokers and E_{cig} = 10.9 g/m^3. The mean exposures, and standard deviation based on updated data are 8 and 31 percent higher, respectively, than those based on defaults. Although the mean values of updated proportion of smokers and "other" smokers are 32% and 5% lower, respectively, than the defaults, the updated mean cigarette emission rate is 27% higher than the default. The overall increase in the estimated exposure despite the lower proportions of smokers and other smokers is consistent with the results of sensitivity analysis, which indicates that exposure is equally sensitive to the proportion of smokers and “other smokers,” and cigarette emission rate.

4.4 In-Vehicle Exposure to ETS

Turk developed a mass balance equation which contains both indoor and outdoor emission sources for calculating concentrations in a chamber (Turk, 1963). Ott *et al.* (1992) summarize previous studies, and describe and evaluate a mass balance equation used in a chamber. Examples of exposure models using mass balance approaches are SHAPE, THEM, APEX, Sequential Cigarette Exposure Model (SCEM), Multi-Chamber Concentration and Exposure Model (MCCEM), and European Population Particle Exposure Model (EXPOLIS) (Ott *et al.,* 1992; Koontz and Nagda, 1991; Hänninen *et al.,* 2003).

Ott *et al*. (1992) evaluates a linear regression equation for estimation of indoor respirable suspended particle (RSP) concentration based on penetration of ambient RSP. They conclude that the linear regression approach can be applied to estimate RSP concentrations from ETS in similar taverns.

A typical mass balance approach for estimation in-vehicle concentration is available in SCEM. SCEM was developed for predicting the time series of concentration in a well-mixed vehicle for any cigarette smoking activity pattern. Ott *et al.* (1992)

observe that time series of carbon monoxide and particle concentration agree well (within 5 percent) with the time series predicted by the model. The mass balance model is:

$$C_{In-Vehicle} = \frac{R_{cig} \cdot E_{cig} \cdot t}{a \cdot V \cdot T} \tag{3}$$

Where,

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

$C_{In\text{-}Vehicle}$ = $PM_{2.5}$ concentration for the in-vehicle microenvironment (µg/m^3);

E_{cig} = emission rate for cigarette smoking (µg/cig);

R_{cig} = average smoking rate (cig/h);

t = duration of active smoking of an individual (min);

T = duration of diary event in the vehicle (min); and

V = vehicle interior cabin volume (m^3).

This model is similar to the second term of Equation (1), except it does not account for deposition rate.

For the in-vehicle microenvironment, results of sensitivity analysis of Equation (3) are given in Table 5. The air exchange rate was varied, holding other inputs at their default values. The estimated in-vehicle $PM_{2.5}$ concentrations range from 8 to 201 µg/m^3

based on air exchange rates ranging from 79 to 3.0 h^{-1}. When all the windows are closed, and with the air conditioner operated in the "max" AC setting with recirculation, the $PM_{2.5}$ concentration reached the largest estimated value. At a vehicle speed of 20 mph, opening a single window from 3 inches to fully open decreased the estimated concentration by fourfold. Air exchange rate is more sensitive to the ventilation system status than to the window status and vehicle speed. With the vehicle speed at 60 mph, windows closed, switching from AC Max (with recirculation of interior air) to AC Regular (with intake of fresh air) leads to an eightfold increase in the air exchange rate, and an eightfold decrease in the in-vehicle $PM_{2.5}$ concentration. At a speed of 20 mph and with AC off, adjusting the window from 3 inches to fully open leads to a fourfold increase in the air exchange rate and decrease in in-vehicle $PM_{2.5}$ concentration. With one window open 3 inches and the AC off, increasing the vehicle speed from 20 mph to 60 mph leads to a threefold increase in the air exchange rate and decrease in in-vehicle $PM_{2.5}$ concentration.

4.5 Inter-individual Variability in Residential $PM_{2.5}$ Exposure with Default Inputs

Simulated cumulative frequency distributions (CDF) of inter-individual variability in daily average $PM_{2.5}$ exposures based on selected scenarios for the residential microenvironment are given in Figure 2. In order to focus comparisons among the exposures attributable to different emission sources, ambient $PM_{2.5}$ concentration is set to 10 μg/m^3 for 24 hours and kept constant for each simulated day. Updated inputs are used in the simulation. The $PM_{2.5}$ sources considered are ETS only, cooking only, and infiltration of ambient air only. A comparison of these three scenarios provides insight regarding their relative importance to exposure.

About 25 percent of simulated individuals are exposed to ETS in the residential microenvironment. The CDF attributable to only ETS has a mean of 7.0 μg/m^3, with a range of 0 to 519 μg/m^3, and a standard deviation of 20 μg/m^3. For an ambient concentration of 10 g/m^3, the average exposure associated with infiltration of outdoor air is 2.4 μg/m^3. The average exposure from cooking is 1.6 μg/m^3. Hence, unless ambient concentration is very high, ETS is likely to contribute a plurality or majority of the average residential indoor exposure in homes where smoking occurs. However, the contribution of ETS to individual exposure is much higher for some individuals, and is zero for households without any smoking.

4.6 Geographic Variability in Daily Average ETS-related $PM_{2.5}$ Exposure

There are geographic differences in factors such as the distribution of smoking prevalence by age and gender, distribution of the population by age and gender, and the distribution of housing stock, which affect comparisons of estimated concentrations. The variation in these factors among geographic areas is shown in Table 6. These data are among the inputs to SHEDS-PM.

The average proportion of smokers, including all age groups and both genders, ranges from 9.3 to 25.2 percent among the selected geographic areas, with Jefferson

County, KY being the highest and Salt Lake County, UT being the lowest. Jefferson County generally has the highest smoking prevalence for all age groups between ages 14 to 64 for both male and female compared to the other four geographic areas, with the exception of male smokers in Wake County, NC aged 45 to 64.

The average age of the population in Wake County, NC and Los Angeles, CA is slightly younger than in the other areas, whereas the other three areas have similar average ages. For Wake and Los Angeles, approximately 54 percent of the population is aged 12 to 34, versus only 42 percent for Jefferson County. In Jefferson County, taking into account the distribution of both smoking prevalence and of the population by age and gender, the cohorts that typically contribute the most to smoking are for ages 35 to 44 for both genders, with a nearly similar contribution from the 45 to 64 age cohorts. Although New York and Salt Lake Counties have a similar proportion of the population in the 45 to 64 age groups as Jefferson County, there is a lower contribution of these groups to ETS exposure because of lower smoking prevalence.

The distribution of housing stock varies geographically. Larger houses are estimated to have lower indoor concentrations of ETS for the same air exchange rate and emission rate compared to smaller houses. Single family homes, whether detached or attached (e.g., townhouses), tend have larger interior volumes than either multiple family homes (apartments) or mobile homes. Los Angeles, Salt Lake, and Jefferson Counties have 67 to 71 percent single family housing, versus only 46 percent for New York

County. Conversely, New York County has the highest proportion of smaller homes, at 54 percent.

Results of estimated incremental daily average $PM_{2.5}$ exposure attributable to ETS are summarized in Table 7. These average exposures range from 4.6 to 7.7 $\mu g/m^3$ among the five analyzed geographic areas. The mean exposure to ETS increases monotonically with respect to the average proportion of smokers, a metric that takes into account both the population distribution and the smoking prevalence for individual age and gender cohorts. However, housing stock is also an influential factor. For example, even though New York County has a smoking prevalence approximately one-third lower than for Jefferson County, the average ETS exposure is only 12 percent lower at least in part because of the generally smaller housing volumes.

5.0 CONCLUSIONS

SHEDS-PM default inputs regarding the proportion of smokers and "other smokers" should be updated to account for the desired time period for which exposures are simulated, since there are significant differences in these proportions over time. The default data regarding cigarette emission rate is low compared to average emission rates estimated from several studies, and thus should be updated. Furthermore, emission rates vary by approximately a factor of three among cigarettes. This variability can be accounted for as part of a probabilistic simulation of exposure. Data on the market-share weighted distribution of variability in cigarette emission rate are needed in order to allow better estimation of the contribution of ETS to indoor air. The algorithms used for ETS exposure in the residential, restaurant, and bar microenvironments are generally based on best practice.

ETS-related $PM_{2.5}$ exposure is sensitive to the proportion of smokers and "other smokers," and cigarette emission rate for the residential indoor microenvironment, and to the incremental increase in indoor $PM_{2.5}$ concentration associated with smoking a cigarette during an hour in the residential and bar microenvironments. Hence, these inputs and parameters are the ones that merit the most attention when developing input data.

For the in-vehicle microenvironment, the most sensitive parameter is the air exchange rate, which in turn depends on the status of windows, the air conditioning and heating system, and vehicle speed. An implication of the sensitivity analysis results is that in-vehicle exposure to ETS can be very high particularly in warm weather for drivers who use air conditioning on recirculation with windows closed. For some individuals, in-vehicle exposures to ETS could be a significant component of daily average exposure even though the time spent in vehicle is less than that of other indoor microenvironments.

For a population of individuals, exposure to ETS can be the largest single contributor to daily average exposure to fine particulate matter, even though only a portion of all individuals are exposed to ETS. For those who are exposed to ETS, there is a wide range of variability in such exposures.

Geographic variability in the prevalence of smokers and demographic factors such as the distribution of the population by age and gender are among factors that lead to geographic variability in daily average $PM_{2.5}$ exposures attributable to ETS. Thus, area-specific data for the proportion of smokers and for demographics should be used.

There are some limitations in available data and models that lead to recommendations for future efforts to improve ETS exposure modeling. Even though the proportion of smokers and the number of cigarettes smoked per smoker per day appear to be declining with time in the U.S., they are still significant and should be tracked consistently over time by age and gender. Some demographic factors that affect smoking prevalence, such as education or socioeconomic status, are not incorporated into existing exposure models. Exposure of young children to ETS and data on the proportion of households in which smoking occur merit quantification. Data are not currently available for avoidance behaviors, such as a non-smoker who avoids proximity to a smoker during a smoking event. Furthermore, changes in smoker activity patterns due to bans on smoking in public indoor spaces, such as whether the rate of smoking is differentially affected in other microenvironments, are not yet quantified.

Despite a variety of actions and messages aimed at reducing the prevalence of smoking, smoking nonetheless continues to be a significant source of exposure to fine particulate matter. The residential and in-vehicle microenvironments in particular are conducive to potentially high exposure concentrations.

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Table 1. Default and Updated National Average Inputs for the Proportion of smokers and "Other smokers" by age and Gender in the Stochastic Human Exposure and Dose Simulation Model for $PM_{2.5}$[a]

Proportion of Smokers (%)				
	Default[b]		Updated[c]	
Age Group	Male	Female	Male	Female
12-13	10.0	10.0	1.80	1.80
14-15	20.1	20.1	8.40	8.40
16-17	29.3	29.3	18.9	18.9
18-24	28.4	23.9	28.5	19.3
25-34	31.5	28.7	27.4	21.5
35-44	32.4	27.3	24.8	20.6
45-64	28.9	24.5	24.5	19.3
>64	14.8	11.4	12.6	8.30

Proportion of "Other Smokers" (%)					
		Default[e]		Updated[f]	
Smoking Status	Age Group	Male	Female	Male	Female
Smoker	12-17	73	89	49	62
	18-64	35	38	37	55
	>64	16	28	32	39
Non-smoker	0-11	n/a	n/a	22	35
	12-17	31	35	17	23
	18-64	10	12	5.0	9.0
	>64	4.0	5.0	4.0	6.0

[a] Data are based on household interviews of a sample of the civilian noninstitutionalized population.

[b] DHHS (1998)(37): adults 18 years and older; SAMSHA (1996): adolescents 12-17 years old.

[c] DHHS (2008)(36): adults 18 years and older; SAMSHA (2008): adolescents 12-17 years old.

[d] Klepeis and Tsang (1996).

[e] DHHS (2007).

[f] According to DHHS (2007), the proportion of other smokers for male continues to be lower than that for females, but available data are not reported by gender. However, ranges are given. To be consistent with the expected trends, the lower bound of the range of the proportion of other smokers is assigned to males, and the upper bound is assigned to females.

Table 2. Results of Sensitivity Analysis of Total Daily Exposure for SHEDS-PM for the Residential Microenvironment[a]

Input Assumption	50th Percentile $PM_{2.5}$ Exposure (µg/m^3)	90th Percentile $PM_{2.5}$ Exposure (µg/m^3)	99th Percentile $PM_{2.5}$ Exposure (µg/m^3)	% Change in 50th Percentile	% Change in 90th Percentile	% Change in 99th Percentile
Default Inputs	4.4	41	105	/	/	/
Proportion of Smokers +10%[b]	4.7	45	116	6	10	10
Proportion of Smokers -10%[b]	4.2	37	95	-6	-10	-10
Proportion of "Other Smokers" +10%[c]	4.7	45	116	6	10	10
Proportion of "Other Smokers" -10%[c]	4.2	37	96	-6	-10	-10
Emission Rate +10%[d]	4.8	45	116	8	10	10
Emission Rate -10%[d]	4.1	37	95	-8	-10	-10
Number of Cigarette -15%[e]	4.1	36	96	-9	-15	-15

[a] Simulation assumptions: 100,000 individuals, 10 randomly selected census tracts in Wake County, NC; same random seeds are used in each simulation; ambient $PM_{2.5}$ concentration: 10 µg/m^3; emission rate: 13.8 mg/cig.

[b] The proportion of smokers given in Table I for each gender and age group was multiplied by 1.1 for the +10% case and by 0.9 for the -10% case.

[c] The proportion of other smokers given in Table I for each smoking status, age group, and gender was multiplied by 1.1 for the +10% case and by 0.9 for the -10% case.

[d] The emission rate was varied from 12.4 mg/cig (-10) to 15.2 mg/cig (+10%).

[e] The average number of cigarettes smoked, as given in (Burke, 2001), is reduced by 15 percent.

Table 3. Results of Sensitivity Analysis of Total Daily Exposure for SHEDS-PM for the Restaurant and Bar Microenvironment[a]

Input Assumption	Restaurant		Bar	
	99th Percentile $PM_{2.5}$ Exposure (µg/m^3)	% Change in 99th Percentile	99th Percentile $PM_{2.5}$ Exposure (µg/m^3)	% Change in 99th Percentile
Default Inputs	12	/	10	/
ASC +10%	13	10	13	10
ASC -10%	11	-10	11	-10
C_{ets} +10%	13	10	13	10
C_{ets} -10%	11	-10	11	-10

[a] Simulation assumptions: 100,000 individuals, 10 census tracts in Wake County, NC; same random seeds are used in each simulation; ambient $PM_{2.5}$ concentration: 10 µg/m^3; ASC = 1.05 cigarette; C_{ets}= 40.4 µg/m^3 in restaurant and bar.

[b] ASC was varied from 0.95 to 1.16 cig/hr.

[c] Cets was varied from 36.4 to 44.4 µg/m^3.

Table 4. Inter-individual Variability in Estimated Residential Microenvironmental Exposures for Default and Updated Data for Smoking Prevalence and Cigarette Emission Rate[a]

Input Data	SHEDS-PM Model Output for Inter-Individual Variability in Daily Average Exposure to $PM_{2.5}$				
	50th Percentile ($\mu g/m^3$)	90th Percentile ($\mu g/m^3$)	99th Percentile ($\mu g/m^3$)	Mean ($\mu g/m^3$)	Std.Dev. ($\mu g/m^3$)
Default Inputs[b]	6.7	29	79	12	16
Updated Inputs[c]	6.0	31	100	13	21

[a] Simulation assumptions: 100,000 individuals, 10 census tracts in Wake County, NC; same random seeds are used in both simulations; ambient $PM_{2.5}$ concentration data were based on hourly data from July 1, 2002 to July 30, 2002 from the output of the CMAQ air quality model.

[b] The proportions of smokers and "other smokers" in 1995 are approximately 23% and 31%, respectively. E_{cig}=10.9 mg/cig.

[c] The proportions of smokers and "other smokers" in 2007 are approximately 18% and 28%, respectively. E_{cig}=13.8 mg/cig.

Table 5. Estimated In-vehicle $PM_{2.5}$ Concentrations for Selected ETS Exposure Scenarios[a]

Speed (mph)	Windows	Ventilation System	Air Exchange Rates[b] ACH (h^{-1})	Predicted $PM_{2.5}$ Concentrations[c] ($\mu g/m^3$)
20	One fully Open	AC Off	78.6	8
60	One Open 3 inches	AC Off	56.4	11
60	All Closed	AC Regular	38.6	16
20	One Open 3 inches	AC Off	20.9	29
0 (parked)	One Fully Open	AC Off	19.2	31
60	All Closed	AC Max	5.1	118
20	All Closed	AC Max	3.0	201

[a] Simulation assumptions: $V = 4\ m^3$, $E_{cig} = 13800$ µg/cig, $R_{cig} = 1.05$ cig/h, t = 10 min, T = 60 min, and ACH as shown.

[b] Air exchange rates were obtain from Ott *et al*. (2008).

[c] $PM_{2.5}$ concentrations are calculated based on the mass balance Equation (3).

Table 6. Factors Affecting Geographic Variability in Daily Average $PM_{2.5}$ Exposure Associated with ETS

Distribution of Smoking Prevalence by Gender (%)[a]										
	Jefferson, KY		Wake, NC		New York, NY		Los Angeles, CA		Salt Lake, UT	
Age	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female
12-13	4.1	4.1	4.0	4.0	5.1	5.1	4.1	4.1	3.1	3.1
14-15	14.4	14.4	12.0	12.0	11.6	11.6	10.6	10.6	4.4	4.4
16-17	27.9	27.9	21.0	18.0	18.1	18.1	17.1	17.1	7.9	7.9
18-24	32.7	30.6	31.3	20.9	19.3	17.3	20.0	13.5	10.1	7.5
25-34	33.6	31.5	29.0	20.6	20.4	18.4	19.9	13.6	10.9	8.3
35-44	28.8	26.7	26.4	16.0	18.1	16.0	18.4	11.9	12.5	9.9
45-64	25.7	23.6	26.8	16.4	20.0	18.1	17.1	10.6	11.9	9.3
>64	11.9	9.8	14.5	4.1	8.9	6.9	9.8	3.3	4.7	2.1
Distribution of Age by Gender (%)[a]										
	Jefferson, KY		Wake, NC		New York, NY		Los Angeles, CA		Salt Lake, UT	
Age	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female
12-13	3.6	3.4	4.5	4.8	3.8	3.9	4.7	4.6	3.1	2.9
14-15	3.5	3.3	4.9	4.3	3.7	3.9	4.6	4.4	3.7	3.3
16-17	3.5	3.5	4.6	4.7	3.5	3.7	4.6	4.5	4.1	3.9
18-24	3.2	3.2	4.5	5.5	3.5	3.6	4.5	4.5	5.4	5.6
25-34	7.4	7.6	8.5	8.5	6.8	7.5	9	8.6	7.5	7.5
35-44	10	11	6.3	5.9	9.7	11.1	8.7	8.3	8.1	7.9
45-64	10.9	11.9	9.6	9.4	11.2	12	9.5	9.5	11.5	11.5
>64	6	8	6.9	7.1	5.6	6.5	5.2	4.8	7.2	6.8

Distribution of Housing Types, Percent[b]					
	Jefferson, KY	Wake, NC	New York, NY	Los Angeles, CA	Salt Lake, UT
Single Family Detached	66	58	30	64	65
Single Family Attached	1	2	16	7	6
Multiple Family	32	31	53	27	27
Mobile Family	1	9	1	2	2

[a] DHHS (2008): adults 18 years and older; SAMSHA (2008): adolescents 12-17 years old.

[b] U.S. Census 2000.

Table 7. Simulated Incremental Daily Average $PM_{2.5}$ Exposures Associated with ETS for Selected Geographic Areas[a]

Location (County, State)	Proportion of Smokers (%)	50th Percentile[b] (µg/m³)	90th Percentile (µg/m³)	99th Percentile (µg/m³)	Mean (µg/m³)	Std.Dev.[c] (µg/m³)
Jefferson, KY	25.2	1.8	22	71	7.7	20
Wake, NC	22.1	1.4	21	72	7.0	20
New York, NY[d]	16.8	1.6	20	69	6.8	21
Los Angeles, CA[d]	14.0	1.3	17	61	6.0	18
Salt Lake, UT	9.3	1.1	13	45	4.6	14

[a] Simulation assumptions: 10,000 individuals per census tract, 10 census tracts simulated for each of Jefferson County (KY), Wake County (NC), New York County (NY), Los Angeles County (CA), and Salt Lake County (UT); ambient $PM_{2.5}$ concentration: 10 µg/m³; proportions of smokers are different for each state based on (CDC, 2009).

[b] For each percentile and for the mean, incremental exposure is calculated by the difference between exposures with and without smoking.

[c] Incremental standard deviation is calculated by the square root of the difference between the variance of exposure with and without smoking.

[d] Smoking was banned in restaurants and bars in NY and CA before 2008; therefore, ETS was not modeled in these two geographic areas in restaurants and bars.

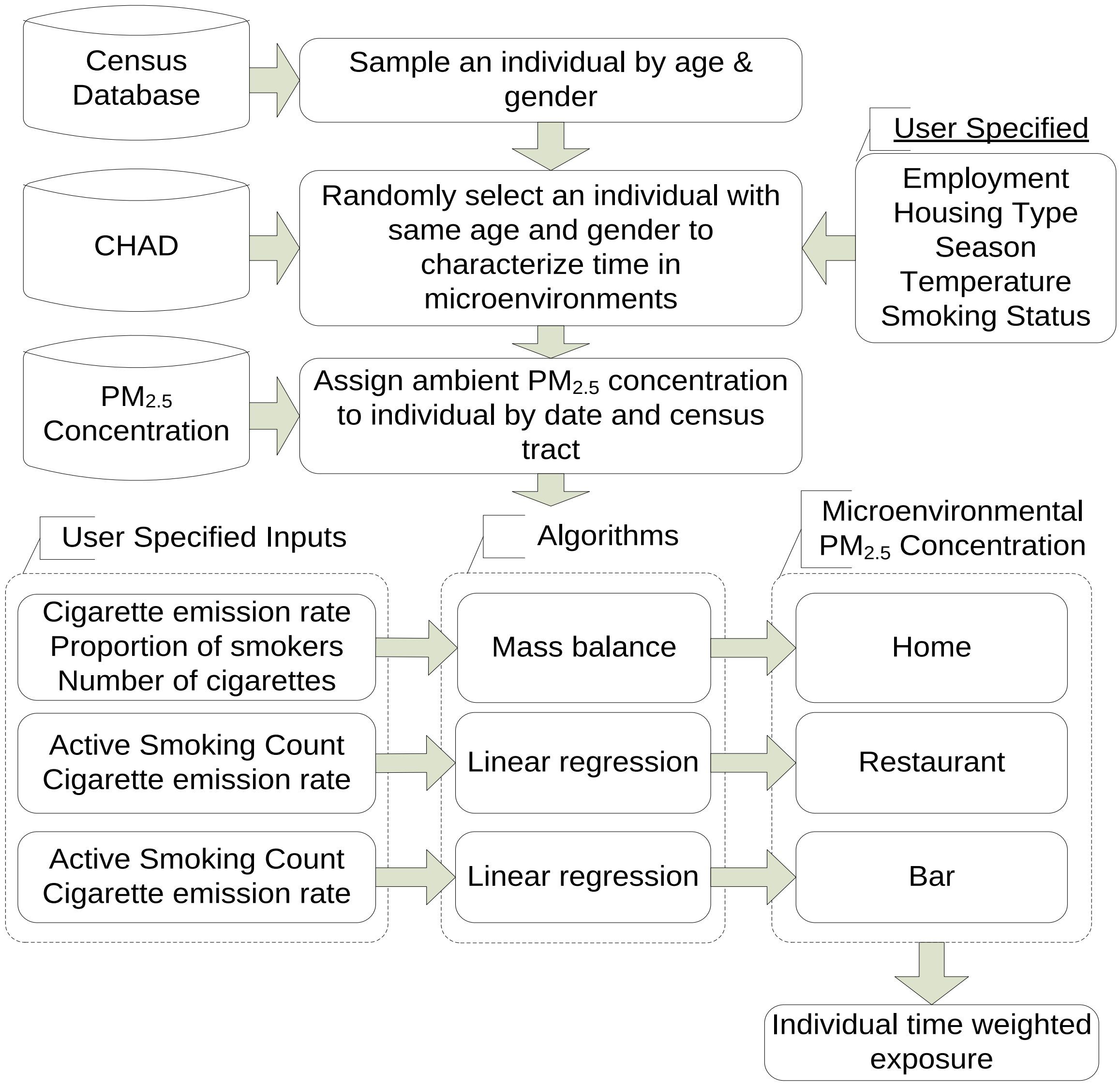
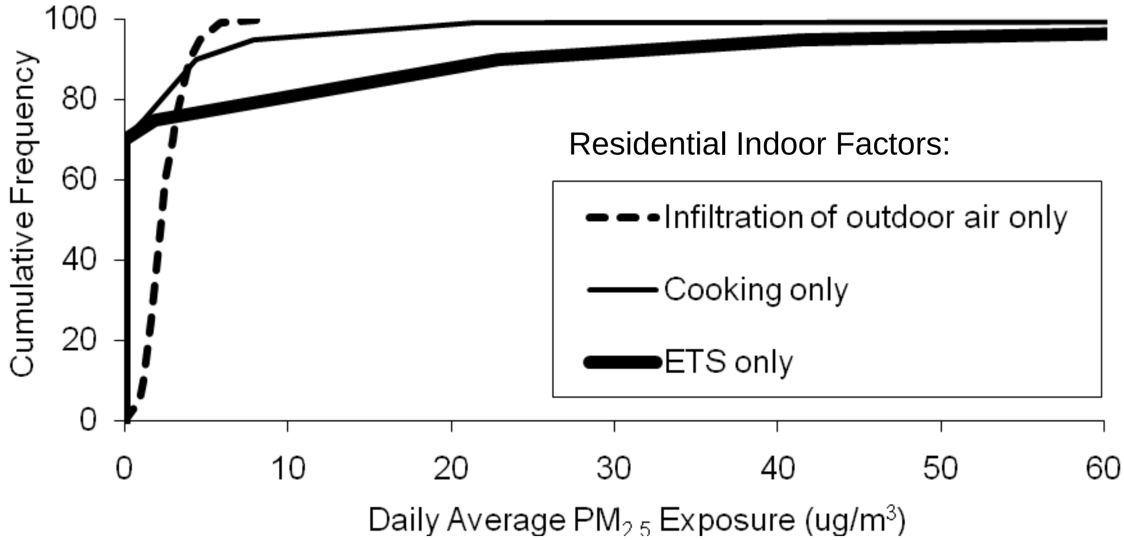


Figure 1. Conceptual Diagram of the Components of the Stochastic Human Exposure and Dose Simulation Model for Particulate Matter (SHEDS-PM) Relevant to Environmental Tobacco Smoke (ETS)



Note: Simulation assumptions: 100,000 individuals, 10 census tracts in Wake County, NC. Same random seeds are used in each simulation Emission Rate: 13.8 mg/cig. Ambient $PM_{2.5}$ concentration: 10 µg/m^3.

Figure 2. Comparison of Cumulative Frequency Distributions of Estimated Daily Average $PM_{2.5}$ Exposures (µg/m^3) in the Residential Microenvironment

PART III

MODELING OF HUMAN EXPOSURE TO IN-VEHICLE $PM_{2.5}$ FROM ENVIRONMENTAL TOBACCO SMOKE

ABSTRACT

Environmental tobacco smoke (ETS) is estimated to be a significant contributor to in-vehicle human exposure to fine particulate matter of 2.5 microns or smaller ($PM_{2.5}$). A critical assessment was conducted of a mass balance model for estimating $PM_{2.5}$ concentration with smoking in a motor vehicle. Recommendations for the range of inputs to the mass-balance model are given based on literature review. Sensitivity analysis was used to determine which inputs should be prioritized for data collection. Air exchange rate (ACH) and the deposition rate have wider relative ranges of variation than other inputs, representing inter-individual variability in operations, and inter-vehicle variability in performance, respectively. The in-vehicle ETS mass balance model was incorporated into the Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM) model to quantify the potential magnitude and variability of in-vehicle exposures to ETS. The in-vehicle exposure also takes into account the near-road incremental $PM_{2.5}$. Results of probabilistic study indicate that ETS is a key contributor to the in-vehicle high-end exposure. Sample and rank correlation coefficient indicate the slightly more importance for cigarette smoking rate than that of cigarette emission rate, and vehicle volume.

1.0 INTRODUCTION

Americans are estimated to spend 87% of their time indoors, 8% outdoors and 5% in vehicles such as automobiles, buses, vans, and trucks (Klepeis *et al.*, 2001). Because of small interior vehicle volume, smoking in a vehicle can potentially expose commuters and passengers to high concentrations of particulate matter less than 2.5 microns in aerodynamic diameter ($PM_{2.5}$). Air pollution epidemiology and exposure studies have identified environmental tobacco smoke (ETS) as a major contributor to human exposure to $PM_{2.5}$ (Wallace 1996; Gilmour *et al.*, 2006). Smoking is associated with significantly increased risk of heart disease, stroke, and chronic lung diseases (Kawachi *et al.*, 1997; Bonita *et al.*, 1999; Vineis *et al.*, 2005). Therefore, it is necessary to account for the contribution of smoking to in-vehicle $PM_{2.5}$ when estimating total exposures to $PM_{2.5}$.

In-vehicle $PM_{2.5}$ concentration includes contributions from both ambient, and non-ambient sources. The emissions from non-ambient sources, such as ETS, can potentially contribute more than ambient sources to the total in-vehicle concentration (Park *et al.*, 1998; Offermann *et al.*, 2002; Ott *et al.*, 2008).

Park *et al.* (1998) modeled the concentration of Respirable Suspended Particles (RSP) with ETS for different vehicle scenarios. Ott *et al.* (2008) used the Sequential Cigarette Exposure Model (SCEM) to estimate ETS $PM_{2.5}$ concentration in vehicles for different vehicle air exchange rates (ACH). However, there is no study in which population-based exposure models are used for the estimation of in-vehicle ETS exposure.

In this work, an in-vehicle ETS mass balance model that accounts for ETS is incorporated into a population exposure model. The potential magnitude of exposure attributable to ETS for different vehicle operating scenarios is quantified. The contribution of near-road emissions to in-vehicle exposure concentration is considered in this study. In-vehicle exposure is compared with other microenvironments.

The objectives of this paper are to: (1) review mass balance models for estimating $PM_{2.5}$ concentrations associated with ETS in vehicles; (2) recommend values of inputs for a selected mass balance model; (3) conduct sensitivity analysis to identify the key inputs for the model; and (4) characterize the potential magnitude and variability of in-vehicle exposures to ETS.

1.1 In-vehicle Exposure

Exposure is defined as the frequency and duration of contact between an agent and a target, with contact taking place at a contact boundary over an exposure period (EPA, 1992). Exposure to air pollutants via the inhalation pathway is often quantified as a time weighted average concentration.

Exposures to ETS in a motor vehicle can exceed those in residential microenvironments by a factor of 10 or more (Offermann *et al.,* 2002). A microenvironment is a physical compartment or defined space with relatively homogeneous or well characterized air pollutant concentrations (Ott *et al.,* 1992a). For just two cigarettes smoked inside a vehicle, 24-hour average personal exposure to $PM_{2.5}$

could be higher than the daily National Ambient Air Quality Standard (NAAQS) of 35 $\mu g/m^3$ (Ott *et al.*, 2008).

1.2 Measurement of In-vehicle Concentration

Two approaches for exposure studies are measurement and mathematical modeling. Direct measurement is the most accurate way to determine in-vehicle air quality, when conducted for representative samples of vehicles and activity patterns, but it is time consuming and costly (Wallace 1987; Thomas *et al.,* 1993; Sexton *et al.,* 1995; Jenkins *et al.,* 1996). In-vehicle $PM_{2.5}$ concentration can be measured using portable, battery-operated laser light scattering photometers.

Usually, average concentrations are obtained based on second-by-second data measured from two aerosol monitors, one located in each of the front and rear seats at the breathing zone in the vehicle. Depending on the status of windows and the ventilation system, the mixing of cigarette emissions takes place over a time period of 1 minute to 1 hour.

The in-vehicle ACH is measured using a tracer gas. For example, sulfur hexafluoride (SF_6) is injected into the vehicle, with all windows closed and the ventilation off, and mixed for around 15 seconds by manually fanning the in-vehicle air to obtain a uniform tracer concentration throughout the vehicle compartment. A tracer gas sample is collected and the mode of ventilation is initiated (i.e. windows open/closed, ventilation on/off). Samples are collected in 30 second intervals by drawing air into polypropylene syringes. The syringe samples are analyzed within 24 hours using a gas chromatograph equipped with an electron capture detector. The ACH is calculated directly from the decay rate of the concentration of SF_6. The average ACH from time t_0 to t_1 is (Spengler *et al.*, 2000):

$$ACH = \frac{\ln C(t_0) - \ln C(t_1)}{t_1 - t_0} \tag{1}$$

Where,

ACH	= air exchange rate (h^{-1});
C (t_0)	= initial tracer gas concentration ($\mu g/m^3$);
C (t_1)	= tracer gas concentration at time t_1 ($\mu g/m^3$);
t_0	= initial time (h); and
t_1	= final time (h).

The removal of ETS particles that deposit on surfaces in the vehicle is denoted by the deposition rate (k). The emissions from cigarettes include both CO and PM. Time series of the concentration decay curves from these two pollutants allows one to determine both ACH, and the decay rate (Φ_p). The difference between these two rates is due to the deposition of particles on the interior surfaces of the vehicle (Ott *et al.*, 1992b, 1996):

$$k = \Phi_p - ACH \tag{2}$$

Where,

Φ_p = decay rate (h^{-1});

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

The deposition rate includes the filter efficiency of the Heating, Ventilating, and Air Conditioning (HVAC) system. As an example, measurements in French high-speed train smoker cars indicated that filter efficiency can vary from 10% to 93% depending on the type of filter. A standard filter has low capture efficiency. An activated charcoal (AC) filter, a high-efficiency H10 (filter class according to European standard EN 1822) filter, and a cellulose acetate (CA) + H10 filter were found to have efficiencies of 44%, 75%, and 86%, respectively (Abadie *et al.,* 2006).

1.3 Modeling of In-vehicle Concentration

A complementary exposure assessment approach, which enables generalization to a wider range of situations, is to develop a mathematical model for predicting in-vehicle $PM_{2.5}$ concentrations that can be validated by measurements. Such a model can be combined with a scenario-based microenvironmental stochastic model that simulates the movement of individual and their exposures.

Indoor air quality models are based on a "mass balance" equation (Ott *et al.,* 2006). The indoor concentration is estimated based on: (1) an infiltration factor which represents the fraction of outdoor particles that reach the indoor environment; (2) indoor emission sources; and (3) loss of particles through deposition on surfaces, filtering or exfiltration via cracks or open windows.

The model is based on three key assumptions: (1) the indoor microenvironment is a single well-mixed zone; (2) a pollutant emitted into the indoor microenvironment mixes quickly, causing its concentration to be spatially uniform at any instant of time; and (3) a short-term change in the outdoor concentration is negligible compared to the long-term average concentrations.

SCEM, a typical mass balance model, was developed for calculating the pollutant concentrations in a well-mixed microenvironment of known volume when any cigarette smoking activity pattern occurs (Ott *et al.,* 1992b). The performance of SCEM has been evaluated in several ETS-related particle studies in microenvironments such as vehicle, tavern, and an experiment chamber (Ott *et al.,* 1992b, 1996).

2.0 METHODOLOGY

The methodology includes: (1) literature review of a mass balance model for in-vehicle air quality estimation; (2) literature review for model input data; (3) sensitivity analysis to identify key factors to which exposure is sensitive for ETS-related $PM_{2.5}$ concentration in vehicles; and (4) use of the Stochastic Human Exposure and Dose Simulation for

Particulate Matter (SHEDS-PM) model to quantify the potential magnitude and variability of in-vehicle exposures to ETS.

2.1 Mass-balance Model

Derivation and assumptions for a mass balance model of in-vehicle $PM_{2.5}$ exposure are given by Ott *et al.* (1992b, 1996). The model takes into account the average smoking rate (R_{cig}), cigarette emission rate (E_{cig}), ACH, k, and interior vehicle volume (V). R_{cig} is the average number of cigarettes being actively smoked during one hour. The application of mass balance models to estimate indoor air quality has been done in various studies conducted over the last 20 years (Spengler *et al.*, 2000; Ott *et al.*, 1992b, 2008; Nazaroff *et al.*, 1989). In order to evaluate the model, comparison of model predictions with measurements was reviewed for a variety of microenvironments.

The values of inputs for the mass balance model were identified based on published data (Ott *et al.*, 1992b, 1996, 2006, 2008; MMWR 2005; Nelson *et al.*, 1994; Repace and Lowrey 1980; Martin *et al.*, 1997). Recommendations of ranges and best estimates for each input are developed.

2.2 Sensitivity Analysis

Sensitivity analysis of an exposure model helps to identify the most significant factors that aid in risk management or that enable prioritization of additional research to reduce uncertainty in the estimates (Frey and Patil, 2002). Sensitivity analysis was conducted to assess the variability in the in-vehicle $PM_{2.5}$ concentration as a function of factors such as ACH, k, and V.

Nominal Range Sensitivity Analysis (NRSA) is used (Frey and Patil, 2002). Ranges of values for each selected input were identified based on measurement data in order to represent variability, uncertainty, or both. During the sensitivity analysis, all inputs were held at their default values except for one, which was varied by plus or minus a typical percentage. The sensitivity for each input is based on the ratio of variation in estimated $PM_{2.5}$ concentration to the estimate based on the default input. Based on comparing sensitivity for each selected input, the key inputs were identified and prioritized.

2.3 Modeling of In-Vehicle ETS Exposure in the Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM) Model

SHEDS-PM was developed by the US Environmental Protection Agency (EPA) (Burke *et al.,* 2001). SHEDS-PM uses a probabilistic approach to estimate distributions of outdoor and indoor $PM_{2.5}$ exposure for a population of simulated individuals based on ambient $PM_{2.5}$ concentrations and sources of indoor $PM_{2.5}$ emissions. Currently, SHEDS-PM accounts for ETS exposure for three microenvironments: home, restaurant, and bar.

A linear regression is used in SHEDS-PM to estimate in-vehicle $PM_{2.5}$ concentration based on ambient concentration, and in-vehicle background concentration (Burke *et al.*, 2001):

$$C_{iv} = r_{I/O} \cdot C_{amb} + B \tag{3}$$

Where,

B = background in-vehicle $PM_{2.5}$ concentration from in-vehicle $PM_{2.5}$ (μg/m^3);

C_{iv} = in-vehicle $PM_{2.5}$ concentration (μg/m^3);

C_{amb} = area-wide ambient $PM_{2.5}$ concentration (μg/m^3); and

$r_{I/O}$ = in-vehicle/area-wide ambient ratio $PM_{2.5}$ concentration.

The background in-vehicle concentration is non-zero only when there are in-vehicle sources of $PM_{2.5}$, such as smoking (Burke *et al.*, 2001).

SHEDS-PM can quantify inter-individual variability in daily average exposures, averaged over one month, taking into account non-ambient and ambient contributions to exposure in each micro-environment. The output of SHEDS-PM includes a database for each individual for each simulated day, with estimates of daily average microenvironmental exposure concentrations for ambient, non-ambient, and total exposure. The parameters used in Equation (3) are set to zero for the intercept and one for the ratio of in-vehicle to area-wide ambient concentration. In a post-processing step, the in-vehicle exposure concentration is adjusted for other ratios of in-vehicle to ambient air, and an intercept is added to take into account the contributions to in-vehicle exposure concentration from near-road emissions and from ETS in the vehicle.

A simulation case study is based on 52,500 randomly selected individuals in all census tracts of Wake County, North Carolina. Figure 1 is the conceptual diagram of the simulation of residential and in-vehicle ETS exposure in SHEDS-PM. C_{ETS} is the in-vehicle $PM_{2.5}$ concentration caused by ETS, $R_{I/O}$ is the ratio of in-vehicle concentration to near-road ambient concentration.

The proportion of smokers and non-smokers are specified in SHEDS based on 2002 data (BRFSS, 2002; Marshall *et al.*, 2006). SHEDS has an option to input data for the proportion of other smokers present at home with smokers or non-smokers. Because no recent data are available to update the defaults in SHEDS for the proportion of "other smokers," default data are used.

Based on a 1997 survey in California with a sample size of 6985, 71% of smokers actively smoked in a vehicle. Twenty-five percent of non-smokers in a vehicle were exposed to second-hand smoke from another person in the vehicle who smoked. A similar prevalence was observed in the U.S. outside of California in 2000 (Kegler and Malcoe, 2002). In North Carolina, 22% of the population are smokers. Thus, for a simulation sample size of 52,500 randomly generated individuals, 11,760 will be smokers. Of these, 8,350 are estimated to smoke while in a vehicle. Of the 40,740 non-smokers, approximately 10,185 are estimated to be exposed to in-vehicle ETS.

The exposure to ETS for a simulated individual is estimated by multiplying the SHEDS output for the daily average vehicle microenvironmental exposure by $R_{I/O}$, and subsequently adding an intercept term that accounts for the incremental portion of daily average ambient exposure associated with incremental near-road $PM_{2.5}$ concentration (C_{NR}) and C_{ETS}. C_{NR} is the concentration from vehicles operating on the roadway, and was estimated using the California LINE Source Dispersion Model, version 4 (CALINE4).

2.4 Variability of In-Vehicle ETS Exposure

In order to explore the significance of in-vehicle ETS exposure for various operating scenarios, three vehicle operating scenarios, referred to as Cases A, B, and C, with different vehicle speeds, status of windows and HVAC system operation are selected.

Results of estimated variability of daily exposure with ETS in the vehicle microenvironment were compared with that in other microenvironments, and to the total exposure, in order to assess the magnitude and variability of in-vehicle ETS exposure.

2.5 Probabilistic Study for In-Vehicle ETS Exposure

A probabilistic case study is conducted to make a preliminary quantification of inter-vehicle variability in several key parameters and to identify which of the selected parameters is the sensitive one for which more or better data would be useful. The results of the probabilistic simulation and sensitivity analysis are a basis for recommending priorities for additional data collection.

Analytica 4.2 is used in the probabilistic study. Quantities in *Analytica* can be specified using a probability distribution function (Chrisman *et al.*, 2010). Variation of C_{ETS} is estimated using Monte Carlo simulation with a simulation sample size of 18,535, this corresponds to the number of persons simulated to be exposed to ETS.

The total in-vehicle exposure is estimated by the time each individual spends in the vehicle from the Consolidated Human Activity Database (CHAD), and the total in-vehicle concentration, which including C_{iv} from SHEDS output, C_{ETS}, and C_{NR}.

Sensitivity in a model output is identified using the sample and rank correlation coefficients between the simulated frequency distribution of CETS and the simulated frequency distribution of each of the three probabilistic inputs. Sample and rank correlations provide an indication of the strength of the linear and monotonic relationships, respectively, between an input and output. (Cullen and Frey, 1999).

3.0 RESULTS

A mass balance model for estimating $PM_{2.5}$ concentration attributable to smoking in a motor vehicle was evaluated based on detailed literature review. Recommendations for the inputs for the mass balance model are given. Key inputs for the model are identified using sensitivity analysis. The magnitude and variability of in-vehicle ETS exposure is assessed.

3.1 Mass Balance Model

A mass balance model has been commonly used to model indoor air quality in a well-mixed compartment (Ott *et al.*, 2006). The ambient ETS $PM_{2.5}$ concentration is assumed to be negligible. The removal of ETS particle mass from in-vehicle air, such as because of filtering, and deposition are assumed to be proportional to the volume-average in-vehicle ETS mass concentration. Under these assumptions, the estimated in-vehicle ETS $PM_{2.5}$ concentration is (Nazaroff and Klepeis 2003):

$$C_{ETS} = \frac{R_{cig} \cdot E_{cig}}{(ACH + k) \cdot V} \tag{4}$$

Where,

ACH	= air exchange rate (h^{-1});
R_{cig}	= average smoking rate (cig/h);
C_{ETS}	= $PM_{2.5}$ concentration caused by ETS (μg/m^3);
E_{cig}	= emission rate for cigarette smoking (μg/cig);
V	= vehicle interior cabin volume (m^3); and
k	= deposition rate (h^{-1}).

All terms except ACH and V are functions of particle size.

The estimated concentrations from Equation (4) have agreed well with measurements in a variety of microenvironments. Ott *et al.* (1992b) found that predicted time series of carbon monoxide and particle concentrations agreed to within 5 percent of measured values. Klepeis *et al.* (1996) found that predicted average RSP and carbon monoxide concentrations in a public lounge agreed to within 12 percent of measurements over multiple scenarios. Ott *et al.* (1996) found that a predicted RSP concentration for a tavern agreed with the observed value within 3 percent. $PM_{2.5}$ concentrations measured in a vehicle and the concentrations predicted by SCEM agreed to within plus or minus 3 percent. Ott *et al.* (2008) concluded that there was a satisfactory fit of the model to the experimental data.

3.2 Inputs

Inputs for the mass balance model include: R_{cig}, E_{cig}, ACH, k, and V.

The estimated 1978 U.S. national average smoking rate was 2 cigarettes per hour (Repace and Lowrey, 1980). R_{cig} ranged from 0 to 4 cigarettes per hour per smoker with an average of 1.3 cigarettes per hour per smoker from 1993 to 1994 for smokers older than 18 years old (Sexton *et al.*, 1995). From 1993 through 2004, the percentage of daily smokers who smoked more than 25 cigarettes per day (cpd) (i.e., heavy smokers) decreased from 19.1% to 12.1%. The average smoking rate among daily smokers in 1993 was 19.6 cpd (21.3 cpd for men and 17.8 cpd for women) and in 2004 was 16.8 cpd

(18.1 cpd for men and 15.3 cpd for women) for smokers older than 18 years old (MMWR, 2005). Based on assuming 16-hours of smoking per day, R_{cig} in 2004 is estimated to be 1.05 cigarettes per hour per smoker. R_{cig} is decreasing with time. This is comparable to the default inputs in the SHEDS-PM Model (Burke *et al.*, 2001; Georgopoulos *et al.*, 2001), in which R_{cig} varies uniformly from 0 to 3. Because we assume smoking occurs in the vehicle, the in-vehicle R_{cig} is assumed to range from 1 to 3 cigarettes per hour. There are no data available that enable quantification of different rates of smoking in vehicles versus other microenvironments.

Typical cigarette $PM_{2.5}$ emission rates reported in the literature are from 10.8 to 22.4 mg/cig (Ott *et al.*, 1996). The mean $PM_{2.5}$ emission rate among the 50 top brands of cigarettes, representing 65.3% of the U.S. cigarette market, is 13.8 mg/cig, with a sample standard deviation of 3.1 mg/cig, a range of 8 to 23 mg/cig, and a sample size of 111 (Nelson, 1994). Nazaroff and Klepeis (2003) summarized 14 papers and report a mean $PM_{2.5}$ emission rate of 13.7 mg/cig. An average emission rate of approximately 13.8 mg/cig is assumed, with a range of 8 to 23 mg/cig.

ACH and k for a typical vehicle are given in Table 1. The deposition rate is calculated based on measured decay rates (Φ_p) and ACH based on Equation (2). Based on variation in vehicle speed, ventilation conditions, and window position, ACH and k range from 3.0 to 78.6 h^{-1}, and 4.7 to 138 h^{-1}, respectively.

V ranges from approximately 2 to 6 m^3 (Ott *et al.*, 2008). For example, the volume of a 2005 Toyota Corolla is 2.4 m^3, a 2005 Ford Taurus sedan is 2.7 m^3, a 1999

Jeep Grand Cherokee Limited is 3.0 m^3, and a 1999 Lexus RX-300 (SUV) is 5.5 m^3. There are few readily available data regarding the distribution of vehicle types. In 2002 there were 136 million passenger cars and 85 million light trucks (mostly SUVs, minivans) in the US (BTS, 2002) . If we assume a typical V= 3 m^3 for cars, and V=5 m^3 for light trucks, a best estimate is about 3.8 m^3.

3.3 Sensitivity Analysis

The best estimate and range of values for each input based on the literature review are summarized in Table 2. A second sensitivity analysis was conducted that focused specifically on the ACH, because ACH varies based on different vehicle operation scenarios. ACH assumptions for the second sensitivity analysis are summarized in Table 1. A nominal estimate of ACH and k is 39 h^{-1} and 10 h^{-1}, with vehicle speed at 60 mph, windows closed, and AC on a regular setting. Other inputs are identical to the defaults in the previous sensitivity analysis.

Since R_{cig} and E_{cig} are in the numerator of Equation (4), the estimated $PM_{2.5}$ concentration responds linearly to a percentage change in these inputs, as shown in Figure 2. Each input has a different relative range of variability, leading to differences in the range of estimated in-vehicle $PM_{2.5}$ concentrations.

For the inputs that are in the denominator of Equation (4), an increase in the input causes a decrease in estimated $PM_{2.5}$ concentrations. Based on the slopes in Figure 2, V is the most sensitive input among those in the denominator for a unit relative change. Since vehicle volume is a constant for a given vehicle, variation of this input represents inter-vehicle variability. For a unit perturbation (e.g., one percent), the $PM_{2.5}$ concentration is slightly less sensitive to ACH than to V. For a unit perturbation, the $PM_{2.5}$ concentration is approximately 4 times as sensitive to ACH as k.

However, ACH and k have wider relative ranges of variability than that of V. Variation in ACH represents variability in how drivers and passengers specify the degree of closure of windows and usage of the HVAC system. Thus, the range of ACH represents variability in operating conditions. The range of input values for ACH leads to -45% to +277% variability in estimated $PM_{2.5}$ concentration.

Results of sensitivity analysis for selected ACH scenarios are summarized in Table 1. At a vehicle speed of 20 mph, when all windows are closed, and with the air conditioner operated in the "max" AC setting with recirculation, the $PM_{2.5}$ concentration reaches the largest estimated value of 940 $\mu g/m^3$. At the same speed, opening a single window by 3 inches decreases the concentration to 170 $\mu g/m^3$. ACH is more sensitive to the ventilation system than to the window status and vehicle speed. With a vehicle speed of 60 mph, windows closed, switching from AC Max to AC Regular leads to an eightfold increase in the ACH and a factor of four decrease in the in-vehicle $PM_{2.5}$ concentration. With a speed of 20 mph and AC off, adjusting a window from 3 inches to fully open leads to a fourfold increase in the ACH and a factor of four decrease in the in-vehicle $PM_{2.5}$ concentration.

3.4 Variability of In-Vehicle ETS Exposure

ETS exposures are very high in some typical vehicle operation scenarios. Therefore, case studies were conducted to characterize the potential magnitude of in-vehicle exposures to ETS using SHEDS-PM.

The estimated in-vehicle $PM_{2.5}$ concentration due to ETS from Equation (4), and infiltration of near-road incremental $PM_{2.5}$ concentration from emissions of other vehicles on the roadway, are incorporated into Equation (3) to estimate the total in-vehicle concentration:

$$C_{it} = R_{I/O} \cdot C_{amb} + R_{I/O} \cdot C_{NR} + \frac{R_{cig} \cdot E_{cig}}{(ACH + k) \cdot V} \quad (5)$$

Where,

C_{it} = total in-vehicle $PM_{2.5}$ concentration ($\mu g/m^3$);

C_{NR} = near-road incremental $PM_{2.5}$ concentration ($\mu g/m^3$); and

$R_{I/O}$ = in-vehicle/near-road ambient ratio $PM_{2.5}$ concentration.

Equation (5) takes into account the penetration of ambient and near-road $PM_{2.5}$ from outdoor to in-vehicle, and emissions from cigarette smoking. The time weighted in-vehicle total exposure is calculated based on the fraction of the day the individual spends in the vehicle.

The contribution of in-vehicle ETS exposure to total exposure is estimated based on three case studies for Wake County, North Carolina, which contains 105 census tracts and had a 2002 population of 630,000 people. A random sample of 500 individuals in each census tract was simulated to characterize activity patterns, including smoking status, and commuting. $PM_{2.5}$ concentration data are based on daily data from July 1, 2002 to July 30, 2002, which is based on predictions of average concentrations for 12 by 12 km grid cell obtained from the Community Multiscale Air Quality (CMAQ) and combined, using Bayesian statistical methods, with monitoring data (McMillan *et al.*, 2010). The average ambient $PM_{2.5}$ concentration during this time period was 21.2 $\mu g/m^3$. The longitudinal simulation type is used in all model runs, which accounts for correlation in day-to-day activity patterns for each individual.

Cases A and B assume vehicle speeds of 20 and 60 mph, respectively. These speeds correspond to local roads and highways, respectively, which have different traffic volumes and numbers of lanes and, therefore, different near-road incremental concentrations attributable to emissions of other vehicles. Cases B and C compare HVAC system operation using outside versus recirculated air, respectively, for vehicle operation on a highway. For each scenario, point estimates of key model inputs are specified as given in Table 3. R_{cig}, E_{cig}, and V are assigned based on the literature review of the range of each input.

Liu *et al.* (2010) used CALINE4 to estimate the near-road incremental $PM_{2.5}$ concentration for selected scenarios that varied with respect to road type, atmospheric stability class, and wind speed, C_{NR} is 6.6 $\mu g/m^3$ based on a local road with 4 lanes, and 35.0 $\mu g/m^3$ based on a highway with 8 lanes. C_{NR} vary among weekday, Saturday, and Sunday in terms of the variation of vehicle miles of travel (VMT). The ratio of VMT between weekday and weekend, and Saturday and Sunday are both 1.2 (Hu and Reuscher 2001; FHWA 2005). Therefore, the C_{NR} reported by Liu *et al.* (2010) are revised by take into account variations by day of the week. C_{NR} is estimated to be 7.1, 5.9 and 4.9 $\mu g/m^3$ for weekday, Saturday, and Sunday, respectively, in Case A; and 37.3, 31.1 and 25.9 $\mu g/m^3$ for weekday, Saturday, and Sunday, respectively, in Cases B and C.

Liu *et al.,* (2010) developed an in-vehicle mass balance model for the case of recirculation of cabin air via the HVAC system and for an HVAC system using outside air. The model takes into account area-wide air quality, incremental near-road concentration, ACH due to infiltration and HVAC system, filter removal efficiency, and deposition rate. The $R_{I/O}$ for Cases A, B, and C are calculated using this model based on the ACH, k, and C_{NR} given in Table 3 and the average ambient air quality. The filter removal efficiency is set to zero, because the reported deposition rates take into account filtering (Ott *et al.,* 2008).

The model run was conducted on a Windows XP quad-processor computer and had an approximate runtime of 400 minutes. Results are given in Table 4.

In Case A, the in-vehicle exposure attributable to the penetrations from area-wide and near-road $PM_{2.5}$ ranges from 0.1 to 4.7 $\mu g/m^3$ from 5th to 99th percentiles, however, the exposure attributable to only ETS has a 99^{th} percentile up to 1180 $\mu g/m^3$, this is due to the low values of ACH, k, V, and extremely high values of R_{cig} and E_{cig}.

Because only 35 percent of the smokers and non-smokers exposed to ETS in the vehicle, ETS exposure is zero in the 50^{th} percentile. The very high end exposure of 99^{th} percentile attributable to only ETS is 11 times higher than the mean, this is because the individual spend six and half hours per day in the vehicle. Based on the CHAD data, the time people spend in the vehicle has a 95 percent frequency range of 15 to 253 minutes per day, with a mean, and a standard deviation of 96, and 103 minutes, respectively. Therefore, for some individuals, such as a truck driver, who is a heavy smoker and spend larger amount of time in the vehicle could have significantly high exposure to $PM_{2.5}$.

In Case A, the average in-vehicle exposure attributable to the penetrations from area-wide and near-road $PM_{2.5}$ is lower than those of other microenvironments. However, the average in-vehicle exposure attributable to ETS is more than 5 and 10 times higher than the residential microenvironment, and other microenvironments, respectively.

In Case B, the average in-vehicle exposure attributable to area-wide and near-road $PM_{2.5}$ is more than twice higher than that of Case A. This is because the C_{NR}, and $R_{I/O}$ of Case B is more than 5 times, and 3 percent higher than those of Case A. The average in-vehicle exposure attributable to only ETS is more than 10 times lower than that of Case A. This is caused by the higher ACH, k, V, and lower E_{cig} in Case B than those of Case A. From Case A to Case B, 70 percent increase of ACH, 66 percent increase of k, 65 percent decrease of E_{cig}, and 200 percent increase of V, lead to 93 percent decrease of in-vehicle exposure attributable to only ETS, respectively.

In Case B, the average in-vehicle exposure attributable to the penetrations from area-wide and near-road $PM_{2.5}$ is approximately equal to those of store and office, and lower than those of other microenvironments. The average in-vehicle exposure attributable to ETS is comparable to that of bar, however, higher than those of other microenvironments, except for home.

In Case C, the average in-vehicle exposure attributable to the penetrations from area-wide and near-road $PM_{2.5}$ is comparable to that of ETS. However, in Cases A and B, ETS is the dominant contributions to the total in-vehicle exposure. This is due to the higher ACH, k, and lower R_{cig} in Case C than those of Cases A and B. The high end exposures of 90th percentile in Case C are 6 and 10 factors higher than the mean, for the penetrations of area-wide and near-road $PM_{2.5}$, and ETS, respectively. From Case C to Case A, 1200 percent increase of ACH, 113 percent increase of k, 33 percent decrease of R_{cig}, 40 percent decrease of E_{cig}, and 100 percent increase of V, lead to 97 percent decrease of C_{ETS}.

In Case C, the average in-vehicle exposure attributable to the penetrations from area-wide and near-road $PM_{2.5}$, and that of ETS are comparable to those of restaurant, and all other indoor microenvironment; and lower than those of home, school, and bar.

3.5 Probabilistic Study for In-Vehicle ETS Exposure

An alternative set of case studies to Case A-P, B-P, and C-P (P for probabilistic) to explore the possible inter-vehicle variability in C_{ETS} based on the limited data now available for variability in R_{cig}, E_{cig}, and V are conducted.

A lognormal distribution is specified for E_{cig} with arithmetic mean and standard deviation of 13.8 and 3.1 mg/cig, respectively, and whose 95 percent frequency range is approximately 8 to 23 mg/cig. For R_{cig}, a uniform distribution between 1 and 3 is assumed. A triangular distribution is assigned to V, with a mode of 3.8 m^3, and a range of 2 to 6 m^3. Because ACH, k, and $R_{I/O}$ are based on the vehicle operating scenario, these inputs are set as constant values for each Case.

Results of inter-vehicle variability of daily average in-vehicle $PM_{2.5}$ exposure based on the distributions of R_{cig}, E_{cig}, and V in each case are given in Table 4 and Figure 3.

Because there is no input change between point estimates and the probabilistic study for the in-vehicle exposure attributable to the penetrations from area-wide and near-road $PM_{2.5}$, the exposures are identical.

In Case A-P, the mean of in-vehicle exposure attributable to ETS is more than 4 times lower than that of Case A. This is due to the central tendency of the distributions of R_{cig}, E_{cig} are 50 and 67 percent lower than the point estimates, and the central tendency of the distribution of V is 47 percent higher than the point estimate. All three of these lead to the decrease of the in-vehicle exposure caused by only ETS.

The ETS exposure in the 90^{th} and 99^{th} percentiles of Case A-P is lower than that of Case A. Because Case A is an extremely worse case for the ETS exposure, C_{ETS} is up to 4500 $\mu g/m^3$, which is constant and large for each simulated smokers and non-smokers who exposed to ETS. However, in Case A-P, the 95 percent frequency of C_{ETS} ranges from 0 to 1439 $\mu g/m^3$, with a mean and standard deviation of 343 and 540 $\mu g/m^3$, respectively. Since the distributions of three inputs are independent, the chance of get simultaneously high R_{cig}, $E_{cig,}$ and V is low. Therefore the standard deviation of ETS exposure in Case A is greater than that of Case A-P.

In Case B-P, the average in-vehicle exposure attributable to ETS is 71 percent lower than that of Case A-P, this is due to the 70 and 66 percent increase of ACH, and k, respectively. The reduction is than that from Case A to Case B.

The average in-vehicle exposure attributable to ETS in Case B-P is 46 percent higher than that of Case B. In Case B-P, the 95 percent frequency of C_{ETS} ranges from 0 to 865 $\mu g/m^3$, with a mean and standard deviation of 205 and 322 $\mu g/m^3$, respectively. Case B has a constant C_{ETS} of 310 $\mu g/m^3$. Therefore, the mean, 90^{th} and 99^{th} percentiles of in-vehicle exposures attributable to ETS in Case B-P are higher than those of Case B. In Case C-P, the average in-vehicle exposure attributable to ETS is more than 6 times lower than that of Case A-P, this is due to the 1200 and 113 percent increase of ACH, and k, respectively. The average in-vehicle exposure attributable to ETS in Case C-P is 8 percent higher than that of Case C. In Case C-P, the 95 percent frequency of C_{ETS} ranges from 0 to 291 $\mu g/m^3$, with a mean and standard deviation of 153 and 72 $\mu g/m^3$, respectively. Case C has a constant C_{ETS} of 140 μg/m^3. Therefore, the mean, 90th and 99th percentiles of in-vehicle exposures attributable to ETS in Case C-P are higher than those of Case C.

Depends on the variation of ACH, and k through Cases A, B, and C, the average in-vehicle exposure attributable to only ETS ranges from 4 to 24 μg/m^3, which is comparable to the exposure in home, bar, school, and restaurant, and higher than those of office, store, all other indoor and outdoor microenvironments.

In Figure 3, from 0 to 65th percentile, the daily average in-vehicle $PM_{2.5}$ exposure in Case A-P and B-P is lower than that of Case C-P. Because ETS exposure is zero for 65 percent non-smokers, the exposure for these non-smokers are dominated by the penetrations from area-wide and near-road $PM_{2.5}$ into the vehicle. Since Cases A-P and B-P has the lower $R_{I/O}$, the exposures for these two cases from 0 to 65th percentile are lower than that of Case C-P. Case B-P has higher $C_{NR,}$ and similar $R_{I/O}$ than that of Case A-P, therefore, the exposure from 0 to 65th percentile for Case B-P is higher than that of Case A-P. However, from 65th to 100th percentile, because of the highest contribution of ETS exposure in Case A-P, the daily average in-vehicle $PM_{2.5}$ exposure in Case A-P is higher than that of Cases B-P and C-P.

The rank correlation between the inputs of R_{cig}, E_{cig}, V and the outputs of C_{ETS} are 0.66, 0.56, and -0.45, respectively. Similar coefficients are found for sample correlation. These results imply that all three variables contribute significantly to variation in exposure, with slightly more importance for R_{cig}.

4.0 CONCLUSIONS

A mass balance approach for calculating the in-vehicle $PM_{2.5}$ concentration attributable to ETS is based on best practice.

ACH is distinguishably the most important input. R_{cig} is slightly more important than E_{cig}, and V, which are of comparable importance to each other. k turn to be the least important. ACH and are k are correlated, and vary based on the behavior of drivers and passengers, and the filter efficiency among different vehicles, respectively. Therefore, data regarding the in-vehicle ACH and k are required for different vehicle scenarios.

Inter-individual variability for in-vehicle ETS exposure is associated with vehicle scenarios, which tend to vary with traffic conditions, and ambient air quality, and therefore will vary seasonally and geographically. Thus, data regarding seasonal and geographic variability in vehicle operation conditions are needed.

Because of the variability in ACH, k and small vehicle interior volume, in-vehicle ETS $PM_{2.5}$ concentration can be higher than that in other microenvironments, especially for a typical scenario in summer, such as windows closed and AC on. Without the contributions from ETS, the average in-vehicle exposure is lower than all of other microenvironments, except for the vehicle with very high ACH, k and, filter efficiency.

Occupational in-vehicle exposure attributable to ETS can be more than 10 times higher than the average in-vehicle ETS exposure. Sixty five percent of people exposed to

ETS in the vehicle microenvironment are not smokers, depends on the time spend in the vehicle, the exposures for these non-smokers can be very high.

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Table 1. Air Exchange and Deposition Rates in Various Vehicle Scenarios[a]

Speed (mph)	Windows	Ventilation System	Decay Rate[b] Φ_p (h^{-1})	Air Exchange Rate[b] ACH (h^{-1})	Deposition Rate[c] k (h^{-1})	Predicted ETS $PM_{2.5}$ Concentration[d]
60	One Open 3 Inches	AC Off	194.4	56.4	138.0	37
20	One fully Open	AC Off	151.4	78.6	72.8	48
60	All Closed	AC Regular	48.8	38.6	10.2	150
20	One Open 3 Inches	AC Off	42.4	20.9	21.5	170
0	One Fully Open	AC Off	39.5	19.2	20.3	180
60	All Closed	AC Max	12.9	5.1	7.8	560
20	All Closed	AC Max	7.7	3.0	4.7	940

[a] Source: Ott *et al.* (2008)

[b] The decay rate (Φ_p) and ACH are measured

[c] Deposition rate (k) includes filter efficiency and ACH is estimated by $k = \Phi_p$-ACH.

[d] $PM_{2.5}$ concentrations are calculated based on Equation (4), and R_{cig} = 2 cig/h, E_{cig} = 13.8 mg/cig, V = 3.8 m^3, and ACH and k as shown.

Table 2. Sensitivity Analysis of the In-vehicle ETS Mass Balance Model

Inputs[a]	Nominal Value	Range of Input Values	Relative Ranges of Input of Variation[a]	Relative Ranges of Predicted In-vehicle $PM_{2.5}$ Concentration[b]
Average Smoking Rate R_{cig} (cig/h)	2	1 to 3	-50% to +50%	-50% to + 50%
Cigarette Emission Rate E_{cig} (mg/cig)	13.8	8 to 23	-42% to +67%	-42% to +67%
Air Exchange Rate ACH (h^{-1})	39	3.0 to 78.6	-92% to +103%	-45% to +277%
Deposition Rate k (h^{-1})	10	4.7 to 138	-50% to +1280%	-72% to +12%
In-vehicle Volume V (m^3)	3.8	2 to 6	-50% to +50%	-37% to +90%

[a] Relative ranges of input of variation is calculated based on the nominal values and range of input values.

[b] Relative ranges of predicted $PM_{2.5}$ concentrations is based on nominal values using Equation (4).

Table 3. Scenario Inputs for Point Estimates of In-Vehicle $PM_{2.5}$ Exposure to ETS[a]

		Case A	Case B	Case C
Speed (mph)		20	60	60
Recirculation or Outside Air		Recirculation	Recirculation	Outside
$R_{I/O}$[b]		0.39	0.40	0.80
C_{NR}[c] ($\mu g/m^3$)	Weekday	7.1	37.3	
	Saturday	5.9	31.1	
	Sunday	4.9	25.6	
ACH (h^{-1})		3.0	5.1	39
k (h^{-1})		4.7	7.8	10
R_{cig} (cig/h)		3	3	2
E_{cig} (mg/cig)		23	8	13.8
V (m^3)		2	6	4
Estimated C_{ETS}[d] ($\mu g/m^3$)		4500	310	140

[a] Vehicle speed, ACH, and k were obtained from Ott *et al.*, (2008).

[b] $R_{I/O}$: the ratio of in-vehicle concentration to near-vehicle ambient concentration, which was estimated to have the same values for weekday and weekend (Liu *et al.*, 2010).

[c] C_{NR}: in Case A, C_{NR} was estimated based on a local road with 4 lanes, wind speed of 1.0 m/s, and stability class G: in Case B, C_{NR} was estimated based on a highway with 8 lanes, wind speed of 1.0 m/s, and the stability class G; the same assumption was used for Case C as for Case B; C_{NR} for weekday, Saturday, and Sunday are estimated based on the ratio of Vehicle Miles of Travel (VMT) between weekday and weekend, Saturday and Sunday (Liu *et al.*, 2010; Hu and Reuscher, 2004; CS and TRI, 2005).

[d] C_{ETS} is calculated based on Equation (4).

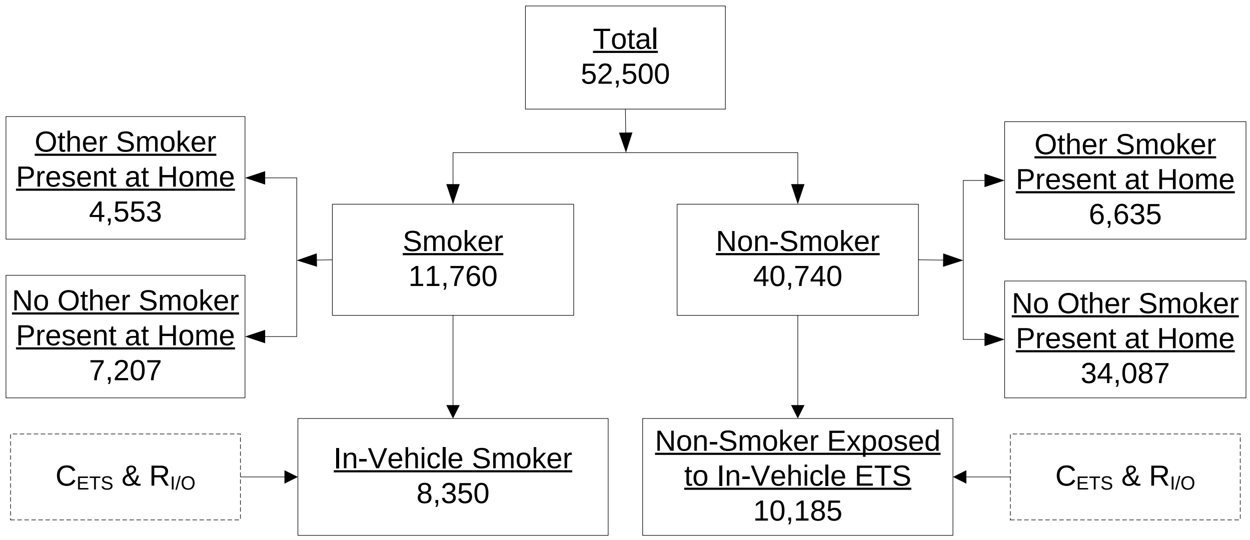
Table 4. Inter-Individual Variability of Daily Average Microenvironmental $PM_{2.5}$ Exposure for Wake County, NC Case Study[a]

Microenvironments			50th Percentile (µg/m^3)		90th Percentile (µg/m^3)		99th Percentile (µg/m^3)		Mean (µg/m^3)		S.D. (µg/m^3)	
			Area-Wide &Near-Road	ETS	Area-Wide &Near-Road	ETS	Area-Wide &Near-Road	ETS	Area-Wide &Near-Road	ETS	Area-Wide &Near-Road	ETS
In-Vehicle[b]	Point Estimates	A	0.1	0	1.5	336	4.7	1180	0.7	108	0.9	246
		B	0.2	0	2.9	23	9.1	82	1.5	7.5	1.7	17
		C	0.4	0	5.7	11	18	37	3.0	3.4	3.4	7.7
	Probabilistic Inputs[c]	A-P	0.1	0	1.5	71	4.7	267	0.7	24	0.9	60
		B-P	0.2	0	2.9	43	9.1	154	1.5	14	1.7	35
		C-P	0.4	0	5.7	11	18	41	3.0	3.7	3.4	9.2
Home			8.7		45		139		18.5		27.9	
Office			1.6		4.0		6.2		1.9		1.5	
School			4.3		8.5		13.7		4.6		3.1	
Store			0.8		4.4		13.2		1.8		2.5	
Restaurant			1.7		7.4		34.6		3.6		6.4	
Bar			6.1		22.2		49.5		9.5		10.1	
All Other Indoor			1.7		7.5		17.0		3.0		3.6	
Outdoor			0.9		5.1		14.5		2.0		3.0	

[a] The case study is based on a sample size of 52,500 individuals in Wake County, NC, using July 2002 air quality data, and the longitudinal simulation type in SHEDS-PM Model, ETS is modeled in the residential, restaurant, and bar microenvironments.

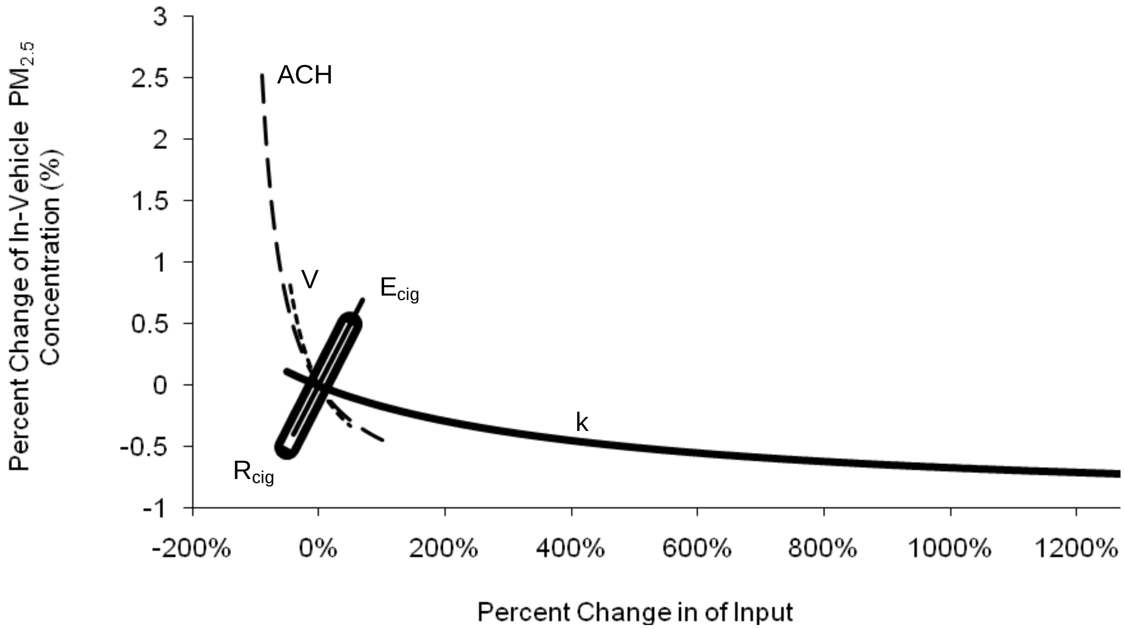
[b] In-vehicle $PM_{2.5}$ exposure include the contributions of ambient air, near-road incremental air, and in-vehicle ETS concentration.

[c] Input distribution: R_{cig}: Uniform(1,3), Uniform(minimum, maximum); E_{cig}: lognormal (13.8, 3.1), lognormal(arithmetic mean, arithmetic S.D.); Triangular(2,3.8,6), Triangular(minimum, mode, maximum).



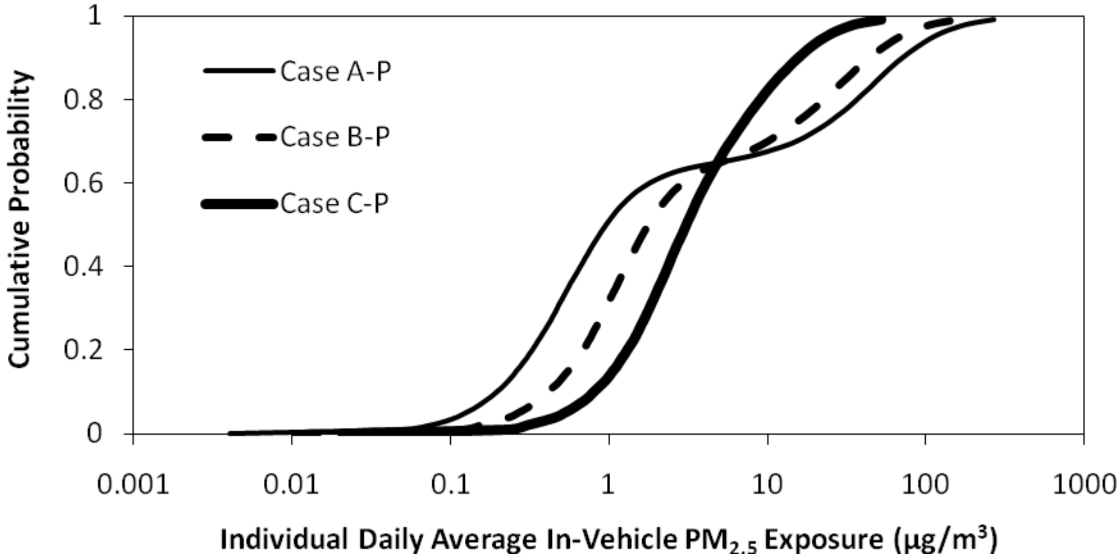
Note: C_{ETS} is in-vehicle $PM_{2.5}$ concentration caused by ETS, $R_{I/O}$ is the ratio of in-vehicle concentration to near-road ambient concentration.

Figure 1. Conceptual Diagram of the Distribution of Simulated Individuals with Respect to ETS Exposure at Home and In-Vehicle for the SHEDS-PM Case Study



Note: Percent Change of $PM_{2.5}$ Concentration (%) is the ratio of $PM_{2.5}$ concentrations based on sensitivity analysis to $PM_{2.5}$ concentrations based on default inputs.

Figure 2. Variation of In-Vehicle $PM_{2.5}$ Concentration Based on Relative Changes of Inputs in the Mass Balance Model



Note: the case study is based on a sample size of 52,500 individuals in Wake County, NC, using July 2002 air quality data, and the longitudinal simulation type in SHEDS-PM Model, In-vehicle $PM_{2.5}$ exposure include the contributions of ambient air, near-road incremental air, and in-vehicle ETS concentration.

Figure 3. Inter-Vehicle Variability of Daily Average In-Vehicle ETS Exposure to $PM_{2.5}$

PART IV

ASSESSMENT OF GEOGRAPHIC DIFFERENCES IN INTER-INDIVIDUAL VARIABILITY OF HUMAN EXPOSURE TO FINE PARTICULATE MATTER

ABSTRACT

Air exchange rate, penetration factor, and deposition rate are the key factors affecting the fraction of ambient particles that penetrate indoors and remain suspended. However, few observational data are available on the levels of seasonal and geographic variability of these factors within residential home. In order to conduct a case study of modeling exposure to $PM_{2.5}$ in New York City, Harris County in Texas, and Wake, Durham, Orange, Alamance, Guilford, and Forsyth County in North Carolina, these factors are summarized based on the critical literature review and assessment. Distributions of air exchange rate, penetration factor, and deposition rate are proposed in three geographic areas and seasons. Sensitivity analysis was used to determine which inputs should be prioritized for updating. Data regarding the air exchange rate was found to be important. Geographic and inter-individual variability of exposure to $PM_{2.5}$ is conducted by using the Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM) model, which is developed by the US Environmental Protection Agency (EPA). Results are shown in Cumulative Distribution Function (CDF) and the ratios between exposure to ambient concentrations for ambient exposure, non-ambient exposure, and total exposure. Geographic variability in daily average exposure is mainly caused by the difference of ambient air quality and air exchange rate. Inter-individual variability in high-end exposure is mainly caused by indoor emission sources, such as smoking and cooking. The ratios of exposure to ambient concentration are significantly different from geographic areas.

1.0 INTRODUCTION

Fine particulate matter ($PM_{2.5}$) includes particles that are 2.5 microns or less in aerodynamic diameter. Exposure to $PM_{2.5}$ is associated with adverse health outcomes (EPA, 2009). Hence, there is a need to quantify human exposure to $PM_{2.5}$ to support assessment of its health effects. Individual exposures to $PM_{2.5}$ occur both outdoors and indoors, and indoor $PM_{2.5}$ concentrations are affected by penetration of ambient $PM_{2.5}$ and exposures from sources such as cooking, cleaning and smoking (Lachenmyer and Hidy, 2000).

In recent epidemiology studies, associations between exposure to $PM_{2.5}$ and health effects are quantified as response-concentration functions based on multicity studies; however, exposure is not measured or estimated (EPA, 2009). These studies assumed that ambient concentration is a surrogate for exposure, but do not address whether the ratio of exposure to concentration is similar for different locations.

$PM_{2.5}$ exposure studies typically employ either direct measurement methods or estimate exposure using models. For example, Williams *et al.* (2003) performed a 1-year investigation in North Carolina of $PM_{2.5}$ and related co-pollutants to characterize the relationship between measured personal exposure versus ambient and residential $PM_{2.5}$ concentration. Mean daily personal $PM_{2.5}$ exposures were only moderately correlated to ambient $PM_{2.5}$ concentrations. Lachenmyer and Hidy (2000) conducted outdoor, indoor and personal exposure measurements for a sample population in Alabama and observed a weakly linear relationship between personal exposure and ambient $PM_{2.5}$ concentration.

Population-based exposure monitoring requires considerable resources. If sufficient data exist, a scenario-based exposure model is an economical tool for quantifying personal exposure (Burke *et al.,* 2001). Such models estimate personal exposure for simulated members of a defined population. The exposures for each individual are simulated based on the time spent in specific microenvironments. Microenvironments are outdoor and indoor places for which concentrations are well characterized. Microenvironments include home, school, store, restaurant and vehicles. Total individual exposure is calculated from the sum of the microenvironmental exposures over the course of an averaging time of interest, such as a typical weekday. As an example, the Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM) model, developed by the US Environmental Protection Agency (EPA), uses a probabilistic approach to estimate distributions of outdoor and indoor $PM_{2.5}$ exposure for a population of simulated individuals based on ambient $PM_{2.5}$ concentrations and sources of indoor $PM_{2.5}$ emissions (Burke, 2005).

People spend 70 percent of their time in the residential microenvironment (Adgate *et al*., 2002; Klepeis *et al.,* 2001). The amout of time each person spends during a typical day in each microenvironment is quantified in SHEDS-PM based on the Consolidated Human Activity Database (CHAD) (Burke, 2005). The key factors affecting the fraction of ambient particles that penetrate indoors and remain suspended are: (1) air exchange rate (ACH); (2) penetration factor (P); and (3) deposition rate (k) (Wilson *et al.*, 2000). ACH is estimated based on measurements with a tracer gas, such as perfluorocarbon tracer (PFT) or sulfur hexafluoride (SF_6). P and k are difficult to measure directly, but are typically estimated by fitting a mass balance model to data for paired indoor and outdoor concentration and ACH. Few observational data are available on seasonal and geographic variability P and k.

Ambient air quality data are obtained from the output of an air quality model such as the Community Multiscale Air Quality (CMAQ) model (Byun and Schere, 2006). The exposure model estimates the exposure based on ambient concentrations, human activity patterns, demographic data and the averaging time used. The estimated exposure can be conceptualized as based on coupling of an air quality model and an exposure model that estimates the ratio of exposure to ambient concentration (Özkaynak *et al*., 2009):

$$Exposure = C \cdot \left(\frac{E}{C} \right) \tag{1}$$

Where,

C = daily-average ambient concentration from an output of an air quality model (μg/m^3);

E/C = the ratio of daily-average exposure to daily-average ambient concentration, based on an exposure model.

The ratio E/C is approximately independent of C for ambient sources of exposure, and varies geographically depending on demographics and housing stock. If modeling resource constraints were an issue, an estimate of the distribution of inter-individual variability in E/C could be employed to estimate exposure concentrations for alternative air quality scenarios, rather than re-run the exposure model.

The objectives of this paper are to: (1) review and recommend values of ACH, P, and k for selected geographic areas; (2) conduct sensitivity analysis for ACH, P, and k to evaluate their importance; (3) evaluate geographic differences in inter-individual variability in exposure; and (4) evaluate geographic differences in the ratio of exposure to concentration.

2.0 METHODOLOGY

The methodology includes: (1) review of algorithm and ACH, P, and k for residential $PM_{2.5}$ concentration in SHEDS; (2) review of literature for ACH, P, and k; (3) sensitivity analysis of ACH, P, and k to assess their importance with respect to estimated exposure; (4) characterization of geographical variability associated with total daily average $PM_{2.5}$ exposure; and (5) characterization of the ratio of exposure to ambient concentration for ambient exposure, non-ambient exposure, and total exposure in each geographic area.

2.1 RESIDENTIAL $PM_{2.5}$ CONCENTRATION

SHEDS-PM includes a single-compartment, steady-state mass balance equation to estimate the indoor $PM_{2.5}$ concentration in the residential microenvironment (Burke *et al.* 2001). The indoor residential $PM_{2.5}$ is a combination of outdoor PM that enters indoors and PM generated by indoor emission sources such as cigarette smoking, cooking, and cleaning:

$$C_{Home} = \frac{P \cdot ACH}{ACH + k} C_{ambient} + \frac{E_{cig}\ N_{cig} + E_{cook}\ t_{cook} + E_{clean}\ t_{clean} + E_{other}\ t_{other}}{(ACH + k)VT} \tag{2}$$

Where,

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

C_{Home} = $PM_{2.5}$ concentration in the home ($\mu g/m^3$);

$C_{ambient}$ = ambient outdoor $PM_{2.5}$ concentration ($\mu g/m^3$);

E_{cig} = emission rate for cigarette smoking (μg/cig);

E_{cook} = emission rate for cooking (mg/m^3);

E_{clean} = emission rate for cleaning (mg/m^3);

E_{other} = emission rate for all other activities (mg/m^3);

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

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

P = penetration factor (unitless);

T = model time step (min);

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

t_{clean} = duration of time spent cleaning during model time step (min);

t_{other} = duration of time spent doing other activities during model time step (min); and

V = volume of microenvironment (m^3).

The air exchange rate, deposition rate and penetration factor can be specified as probability distributions. Default values of p, k, and ACH are given in Table 1.

ACH is the volume flow of air within the indoor microenvironment divided by the interior volume. ACH is affected by air leakage through cracks and crevices in the building envelope, natural ventilation through open windows and doors, and mechanical ventilation by fans (Burke, 2005).

SHEDS-PM categorizes ACH into four seasons: winter, spring, summer, and fall. The default data for ACH for these seasons was originally derived from a PFT database developed by Brookhaven National Laboratory (BNL). Murray and Burmaster (1995) analyzed the database and categorized ACH by climate region and season. However, the regional variations of ACH represented in Murray and Burmaster (1995) are not included in SHEDS by default.

The penetration factor is the ratio of particles that penetrate indoors from outdoors. The particle deposition rate refers to settling of airborne particles due to gravity and diffusion. The deposition rate depends on particle size and density and room temperature gradients and ventilation conditions (Burke, 2005). The default values of P and k in SHEDS are obtained from the Particle Total Exposure Assessment Methodology

(PTEAM) study conducted for Riverside, California, in fall of 1990 (Özkaynak *et al.* 1996).

2.2 Review of Penetration, Depostion, and Air Exchange Rates

The review of P, k, and ACH is based on: (a) detailed review of the SHEDS-PM model, its user guide, and the literature cited as the basis for default input assumptions; (b) published peer reviewed papers regarding similar models; and (c) published peer reviewed papers regarding data for ACH, P and k. Data are reviewed with respect to selected geographic areas and for four seasons.

2.3 Sensitivity Analysis

Sensitivity analysis of an exposure model helps to identify the most significant factors that aid in risk management or that enable prioritization of additional research to reduce uncertainty in the estimates (Frey and Patil, 2002). Sensitivity analysis was conducted to access the variability in daily average $PM_{2.5}$ exposure as a function of variation in P, k, and ACH.

During the sensitivity analysis, all inputs were held at their default values except for one, which was varied probabilistically. Results are shown as a Cumulative Distribution Function (CDF) of inter-individual variability in daily average exposure for simulated individuals. Based on the percent difference in the mean and standard deviation of exposure associated with comparison of alternative distributions for each selected input, the key inputs were identified and prioritized.

2.4 Geographic and Inter-Individual Variability

To assess the geographic variability in estimated exposure, three locations were selected that represent different climate zones: (1) New York City (NYC); (2) Wake, Durham, Orange, Alamance, Guilford, and Forsyth Counties in North Carolina, which includes the cities of Raleigh, Durham, Burlington, Greensboro, High Point, and Winston-Salem; and (3) Harris County in Texas, which includes Houston. Since the average ambient $PM_{2.5}$ concentration tends to be highest in the summer, air quality data for July 2002 were selected. Six counties were selected for the NC case study to represent urban areas along the I-40 highway corridor.

SHEDS-PM output includes a database for each individual for each simulated day, with estimates of daily average microenvironmental exposure concentrations for ambient, non-ambient, and total exposure. The ratio of exposure to ambient concentration for ambient exposure, non-ambient exposure, and total exposure in each geographic area are based on the pairwise estimated exposure and assigned ambient concentration for each simulated individual.

3.0 RESULTS

Recommended values of ACH, P, and k are given. Data regarding ACH are reviewed mainly based on Murray and Burmaster (1995) and the Relationship of Indoor, Outdoor and Personal Air (RIOPA) study from 1999 to 2001. Data regarding P and k are reviewed mainly based on the RIOPA and PTEAM Studies (Weisel *et al.* 2005; Özkaynak *et al.* 1996). Each input is evaluated using sensitivity analysis. Geographic and inter-individual variability in daily average exposure to $PM_{2.5}$ is evaluated.

3.1 Air Exchange Rate

Murray and Burmaster (1995) summarized ACH data compiled by BNL for 2,844 households. This is the most recently reported comprehensive analysis of such data. They stratified the data into four regional climate zones based on heating degree days. For each region and season, lognormal distributions were fit to the data to represent inter-household variability. New York State includes two regions, and NYC is in region 2.

Koontz and Rector (1995) analyzed 2,976 measurement results from the BNL database stratified by state rather than climate zone. Available ACH data for NYC from Murray and Bumaster (1995) and Koontz and Rector (1995) are compared in Table 2.

Summer estimates for Region 2 are based mostly on data from Washington State, and thus are not representative of NYC (Murray and Burmaster, 1995). Koontz and Rector (1995) report μ_g for New York State but not σ_g. However, Murray and Burmaster

(1995) report σ_g of 2.09 for Region 2, which includes New York State just north of NYC. Thus, ACH for summer in NYC is assumed to have μ_g of 0.64 with σ_g of 2.09.

For summer and Texas, μ_g and σ_g from the RIOPA and Koontz and Rector (1995) data are much lower than from Murray and Burmaster (1995). Because air conditioning usage is higher in Texas than in the northern regions (EIA, 2000), ACH is expected to be lower in the summer than for other seasons. The seasonal trend in μ_g from the RIOPA data is internally constant and thus these data are used for summer.

Wallace *et al.* (2006) measured ACH in 37 residences in the Research Triangle Park (RTP) area in North Carolina for 7 consecutive days in each of four seasons. As shown in Table 2, the highest mean value of ACH was observed in the winter and the lowest in the summer, a reversal of the typical pattern in other studies (Abt *et al.*, 2000; Long *et al.*, 2001). North Carolina is in Region 3 in Murray and Burmaster (1995). For summer, both μ_g and σ_g from Murray and Burmaster (1995) are similar to these reported by Wallace *et al.* (2006). The data from Wallace *et al.* (2006) are used for summer.

3.2 Penetration Factor and Deposition Rate

Estimated values of P and k from the RIOPA and PTEAM studies are summarized in Tables 3 and 4, respectively. Ideally, P and k should be measured and estimated for a house with no indoor emission sources. However, many of the available data appear to be for houses with indoor sources, except as noted. The presence of indoor sources will produce bias in P, including values greater than 1, and bias in k. In some cases, estimates of k are negative.

In the RIOPA study, there are a limited number of households (n=21) for which there is explicit reporting of no indoor emission sources (Weisel *et al.,* 2005). For these households, P and k are 0.78, and 0.40 h^{-1}, respectively. For 165 households, the indoor concentration was less than the outdoor concentrations, as denoted in the table as I/O<1. I/O<1 is consistent with houses that do not have indoor sources, but it is not adequate assurance that no indoor sources were present. For these data, the average values of P and k are 0.73 and 0.20 h^{-1}, respectively. Data for other RIOPA locations may include indoor sources and, thus, might be biased.

The PTEAM study was conducted in Riverside, California in fall 1990. Indoor concentrations were measured in the center of the residence and outdoor concentrations were measured nearby. Measurements were conducted daily for two consecutive 12-h periods. During each 12-h period, PFT measurements were made from which ACH was estimated.

Based on the physical constraint that $P \leq 1$, an upper bound of $P = 1$ is assumed. A lower bound of 0.7 is assumed consistent with the mean value observed for the RIOPA study for I/O<1, Elizabeth, NJ, and the lower bound of the reported 95 percent Confidence Intervals (CI)s for Los Angeles and the overall data. A nominal "best estimate" of 0.78 is assumed based on the mean of the data for which no indoor sources were reported. A triangular distribution is used for P to represent these judgments.

Triangular distributions are sometimes used to represent judgment in the absence of a probability sample of data (Cullen and Frey, 1999). In order to assess whether the estimated inter-individual variability is sensitive to the specification of the distribution for P, two alternative frequency distributions are also considered: (1) normal distribution with a mean of 0.85 and a standard deviation of 0.075; (2) normal distribution with a mean of 0.90 and a standard deviation of 0.05.

A lower bound of $k = 0.2\ h^{-1}$ is assumed consistent with the mean value observed for the RIOPA data in which $I/O \leq 1$, the lower bound of reported CIs for overall cases, and the lower bound of the CI for Los Angeles from the PTEAM study. A nominal "best estimate" of $0.40\ h^{-1}$ is assumed based on the mean of data for which no indoor sources were reported, and the mean of data from the PTEAM study. The upper bound of $k = 0.6\ h^{-1}$ is assumed based on the upper bound of the CI from the PTEAM study. A normal distribution is assumed for k with a mean of $0.40\ h^{-1}$ and standard deviation of 0.1, giving a 95 percent probability range of approximately 0.2 to $0.6\ h^{-1}$. To assess whether exposure estimates are sensitive to the specification of the distribution of k, an alternative frequency distribution is considered. A normal distribution is also assumed for k with a mean of $0.39\ h^{-1}$ and a standard deviation of 0.085.

3.3 Sensitivity Analysis

Sensitivity analysis was conducted to assess variability in the daily average $PM_{2.5}$ exposure as a function of alternative probabilistic assumptions for P, k, and ACH. The specified distributions for these inputs are summarized in Table 5.

To compare the sensitivity of estimated exposure to each of several residential microenvironmental inputs, a case study was developed based on all census tracts in Harris County, Texas. For each census tract, 1% of the population was simulated. Distributions for P and k are assumed to be the same for the three geographic areas, while ACH differs. Sensitivity analysis for ACH is conducted for summer in Harris County, Texas. Air quality data for July 2002 were used. These data are estimated based on the predictions of average concentrations for 12 by 12 km grid cells obtained from CMAQ and combined, using Bayesian statistical methods, with monitoring data (McMillan *et al.*, 2010). All exposure model runs were conducted using longitudinal simulations and the same random seed. Five Harris County simulations were conducted for various combinations of probabilistic assumptions for P, k, and ACH: (1) P_1, k_1, ACH_1; (2) P_2, k_1, ACH_1; (3) P_3, k_1, ACH_1; (4) P_1, k_2, ACH_1; and (5) P_1, k_1, ACH_2. Each model run was conducted on a Windows XP Pentium 4 computer and had an approximate runtime of 800 minutes. The results for the sensitivity analysis are given as CDFs of inter-individual variability in daily average $PM_{2.5}$ exposure based on the variation of each input.

The three alternative distributions for P shown in Figure 1(a) differ in terms of central tendency and range. However, the corresponding three simulated distributions of inter-individual variability in exposure shown in Figure 1(b) are very similar. The mean exposure differs by less than 5 percent and the standard deviations differ by less than 2 percent. Therefore, the results are not sensitive to the choice of distribution for P. Since P_3 is the only one of the three that observes the physical limit of $P \leq 1$, P_3 is chosen as the basis for further simulations.

The two alternative distributions for k differ in terms of central tendency and range as shown in Figure 2(a). However, the corresponding simulated distributions of inter-individual variability in exposure are very similar as shown in Figure 2(b). Both the mean exposure and the standard deviation of exposure differ less than 1 percent. Therefore, the results are not sensitive to the choice of distribution, and k_1 is chosen as the basis for further simulations.

The two alternative distributions of ACH for Texas differ in terms of central tendency and range as shown in Figure 3(a). The μ_g and σ_g of ACH_1 are 65 and 24 percent higher than those of ACH_2, respectively. The corresponding simulated distributions of inter-individual variability in mean exposure and standard deviation based on ACH_1 is 67 and 40 percent lower than that based on ACH_2 as shown in Figure 3(b), respectively. Therefore, the results are sensitive to the choices of distributions of ACH. ACH_2 is chosen because it is consistent with the expected seasonal trends.

3.4 Geographic Variability of Exposure to $PM_{2.5}$

A random sample of 50,000 individuals in all age groups from all census tracts in each of NYC, Harris County in Texas, and the six counties in North Carolina was simulated to characterize the geographic variability of estimated exposure to $PM_{2.5}$. $PM_{2.5}$ air quality data input to SHEDS-PM are based on daily 12 km by 12 km grid cell concentrations for July 2002 obtained from CMAQ and combined, using Bayesian statistical methods, with monitoring data (McMillan *et al.*, 2010). The average ambient $PM_{2.5}$ concentration during this time period was 20.9, 15.7, and 20.6 $\mu g/m^3$ in the NY, TX, and NC domains, respectively. Longitudinal simulation was used in all model runs.

Inputs that vary among geographic areas, including ACH, smoking prevalence, demographics, and distribution of housing types, are given in Table 6. The central tendency of summer ACH in Texas is 31 and 42 percent lower than for the NC and NYC domains, respectively. Lower ACH leads to more retention of indoor emissions, and higher non-ambient exposure. Conversely, lower ACH leads to lower penetration of ambient $PM_{2.5}$ indoors, and lower ambient exposure.

Based on the distribution of smoking by gender and age, and the population distribution by age and gender, the weighted overall prevalence of smokers are 23.4, 20.0, and 19.1 percent for NC, TX, and NYC, respectively. The smoking prevalence varies substantially among different age and gender cohorts. For example, the proportion of smokers older than 45 years old is 28 percent higher than those in other age groups in Harris County, TX. Furthermore, the proportion of time spent indoors also varies by cohort. Based on CHAD diary data, the distribution of daily average time spent outdoors, indoors, and in travel by gender and age are given in Table 7. The time spent indoors for people older than 64 years old are 16 percent higher than that of people from 14 to 15 years old. Human activity patterns have as an impact on the exposure for subgroup of population.

The average interior volume of residential housing is approximately similar for the NC and TX domains, but approximately 30 percent lower in NYC. The smaller average housing interior volume is because the proportion of multiple family (apartment) housing in NYC is more than 50 percentage points larger than that in North Carolina. The NC and TX domains have approximately two-thirds single family houses, which have larger interior volume than apartments. Large indoor volume enhances the dilution of indoor emissions. Smaller indoor volume leads to higher indoor exposure to non-ambient $PM_{2.5}$.

As shown in Table 7, when the activity patterns in Table 7 are weighted based upon the geographic location-specified population distribution, as given in Table 6, differences in activity patterns between cohorts can be pronounced in some cases. For example, even though the same activity diary data are used, because there is a larger proportion of 18 to 24 year old males in NC, the overall proportion of time spent outdoors by 18 to 24 year old males in NC is estimated to be 4 and 13 percent more than those for TX and NYC, respectively. The aggregated proportion of time spent outdoors for people older than 45 years old in NYC is estimated to be 6 percent more than those of

TX and NC domains, respectively. There is also a larger proportion of smokers older than 45 years old in NYC.

The estimated daily average ambient exposure in NYC is 12.7 $\mu g/m^3$, with a 95 percent frequency range, as shown in Figure 4(a), of 3.4 to 24.6 $\mu g/m^3$. For Harris County, the mean is 8.4 $\mu g/m^3$, with a range of 3.4 to 14.1 $\mu g/m^3$. For the six county area of North Carolina, the mean is 12.2 $\mu g/m^3$, with a range of 4.4 to 21.0 $\mu g/m^3$.

The higher estimated ambient exposure in NYC is attributed in part to higher ambient $PM_{2.5}$ concentration, which averages 25 and 1.4 percent higher than those of Texas, and North Carolina, respectively. The higher summer ACH in NYC leads to more indoor penetration of ambient $PM_{2.5}$. Ambient exposure for NC is 31 percent higher than that for TX. This is attributed in part to higher ACH and ambient $PM_{2.5}$ concentration in NC than that for Harris County.

The average non-ambient exposure in Harris County is 14 and 11 percent higher than that of the NC and NY domains. The popoulation weighted smoking prevalence in Texas is lower than that of NC. The weighted average indoor residential volume for Texas is higher than that of NC and NYC. These two factors lead to lower non-ambient exposure to $PM_{2.5}$. However, the low summer ACH in Texas leads to the overall higher non-ambient exposure. This implies that non-ambient exposure is more sensitive to ACH than to other factors.

NYC has a lower weighted average smoking prevalence and higher summer ACH than that of NC, which should lead to lower non-ambient exposure. However, smaller average indoor volume in NYC leads to overall higher non-ambient exposure to $PM_{2.5}$ than that of NC. This implies that non-ambient exposure is sensitive to the distribution of housing stock.

The mean values of total daily average exposure to $PM_{2.5}$ in NYC, six county area of North Carolina, and Harris County in Texas are 28.9, 27.8, and 26.6 $\mu g/m^3$, respectively. The comparative order of these averages is consistent with the order of average ambient exposure. However, the 90th percentile of total exposure is higher in Harris County by 3 and 7 percent compared to NYC and North Carolina, respectively. The latter trend is consistent with the trend in non-ambient exposure. Thus, geographic variability of total daily average exposure is influenced by the variability in ambient exposure. However, the geographic variation in high-end exposure is influenced by variability in non-ambient exposure. This is because the high-end of non-ambient exposures can be a factor of 2 to 10 higher than the ambient exposures. The high-end exposure are mainly caused by indoor emissions, such as, smoking and cooking. The estimated average non-ambient exposures for smokers are approximately fivefold higher than those for non-smokers in the three geographic areas.

3.5 Inter-Individual Variability in Exposure to $PM_{2.5}$

Figure 5 illustrates variability in the ratio of estimated exposure to ambient concentration for ambient exposure (E_a/C), non-ambient exposure (E_{na}/C), and total exposure (E_t/C).

As shown in Figure 5(a), except for one simulated outlier individual in each area that has E_a/C greater than 1, all simulated values of E_a/C are lower than 1. The mean values of E_a/C are 0.60, 0.58, and 0.52, with standard deviations of 0.16, 0.15, and 0.14, for the NY, NC and TX domains, respectively. The geographic differences in the mean E_a/C ratio are associated with differences in average ACH and demographics by age and gender among the three geographic areas.

NYC has the highest E_a/C ratio. Higher ACH in NYC leads to higher penetration of ambient $PM_{2.5}$ indoors, which leads to higher exposure to ambient $PM_{2.5}$. The average E_a/C in the three geographic areas increase as the mean ACH increases.

th percentile, and 90^{th} percentile of E_a/C varied approximately by 3 percent between the NYC and NC domians, 9 percent between the NC and TX domains, and 12 percent between the NYC and TX domains. The 50^{th} percentile of ACH in NYC is 16 and 42 percent higher than those of the NC and TX domains, the 90^{th} percentile of ACH in NYC is 2 and 13 percent higher than those the TX and NC domains.

The relative magnitude of E_a/C appears to be insensitive to population demographics when comparing the three geographic areas. For example, the comparative increase in mean E_a/C for NYC, NC, and TX does not correspond to a demographic trend such as the proportion of the population aged 18 to 65.

Based on Figure 5(b), the average E_{na}/C are 1.2, 1.0, and 0.9, with standard deviations of 2.1, 1.8, and 1.4, for the TX, NY and NC domains, respectively. Geographic variability in E_{na}/C is associated with ambient air quality, ACH, smoking prevalence, and indoor volume. The TX domain has the lowest ACH and the lowest average ambient air quality. Low ACH leads to more retention of indoor emissions. Thus, TX has the highest average E_{na}/C ratio. Other factors are not as sensitive as ACH and ambient air quality. The overall smoking prevalence in TX is lower than that of NC. The proportion of single family homes in TX is 67 percentage points more than that of NYC; therefore, the average indoor volume of TX is greater than that of NYC. Larger indoor volume helps the dilution of indoor emission sources, such as smoking and cooking. However, in this case, the ratio of E_{na}/C is more sensitive to ACH.

Based on Figure 6(c), the average E_t/C are 1.8, 1.6, and 1.4, with standard deviations of 2.1, 1.8, and 1.5, for the TX, NY and NC domains, respectively. The 90th percentile E_t/C ratio is consistent with the order of E_{na}/C ratio. However, in the 50th percentile, E_t/C is consistent with the order of E_a/C ratio. The 50th percentile E_t/C for NYC is 17 and 35 percent higher than those of the TX and NC domains, respectively.

4.0 CONCLUSIONS

Recommended values of P, k and ACH are given. However, there are relatively few data from which to estimate P and k. Data are needed for P and k that more clearly represent situations without indoor emissions. Furthermore, more recent data for ACH are needed to represent current housing stock in different regions.

For a single model domain, ACH is distinguishably the most sensitive input for both ambient and non-ambient exposure to $PM_{2.5}$, especially in combination with strong indoor emission sources. High ACH leads to high ambient exposure indoors but lower non-ambient exposure, and vice versa.

Approximately 60 percent of total daily average exposures to $PM_{2.5}$ are caused by non-ambient exposures for each of the three geographic areas. The extremely high daily average exposures for some individuals are mainly caused by non-ambient exposure to smoking or cooking. Geographic differences in housing stock and climate, which lead to geographic differences in ACH, account for geographic variability in the estimated ambient and non-ambient exposure to $PM_{2.5}$.

The distributions of E_a/C ratios in each geographic area indicate that simulated individuals are typically exposed to more than half of the modeled ambient concentrations. The 95 percent frequencies of E_a/C range approximately from 0.5 to 0.8. The average E_a/C ratio varies among geographic areas based on differences in housing stock, climate, and seasonal ACH. Thus, on average, individuals in different geographic areas are exposed to different proportions of ambient $PM_{2.5}$ by 13 percent.

Most epidemiology studies are using central site ambient monitoring data as a surrogate for population-level exposure. Area-specific E_a/C ratios are recommended for possible use in epidemiology studies to better address geographic and inter-individual variability in exposure to $PM_{2.5}$. The differences in average E_a/C may help to explain geographic variations in dose-response functions.

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Table 1. Default Inputs for the Residential Microenvironment in the Stochastic Human Exposure and Dose Simulation Model for Particulate Matter (SHEDS-PM)

Parameters	Distribution Type	Distribution Values
Penetration (P)	Normal	Mean=0.97, Standard Deviation=0.1
Deposition (k)	Normal	Mean=0.30 h^{-1}, Standard Deviation=0.095 h^{-1}
Air Exchange Rate[a] (ACH)	Lognormal	$Season_1$ (winter): μ_g =0.449 h^{-1}, σ_g =2.014 h^{-1} $Season_2$ (spring): μ_g =0.449 h^{-1}, σ_g =2.226 h^{-1} $Season_3$ (summer): μ_g =0.819 h^{-1}, σ_g =2.014 h^{-1} $Season_4$ (fall): μ_g =0.368 h^{-1}, σ_g =1.649 h^{-1}

[a] Sources: Burke *et al.* (2001). μ_g = Geometric Mean. σ_g = Geometric Standard Deviation.

Table 2. Summer Air Exchange Rates for New York City, Six County Area of North Carolina, and Harris County in Texas

Location	Region	Geometric Mean (h^{-1})	Geometric Standard Deviation (h^{-1})
NYC	Murray and Burmaster (1995), Region 2[a]	1.26	na
	Koontz and Rector (1995), New York State	0.64	na
TX	Murray and Burmaster (1995), Region 4[a]	1.05	2.49
	Koontz and Rector (1995), TX	0.05	na
	RIOPA study (2005), TX[b]	0.37	1.9
NC	Murray and Burmaster (1995), Region 3[a]	0.56	1.84
	RTP Panel Study (2006), NC[c]	0.54	1.70

[a] Region1: annual heating degree days ≥ 7,000; Region 2: 5,500 ≤annual heating degree days ≤ 7,000; Region 3: 2,500 ≤annual heating degree days ≤ 5,500; Region 4: annual heating degree days < 2,500 (Murray and Burmaster, 1995).

[b] Sources: Relationship of Indoor, Outdoor and Personal Air (RIOPA) study from 1999 to 2001 (Weisel et *al.*, 2005); Sample size: 63 in summer.

[c] Research Triangle Park (RTP) Panel Study (Wallace et al., 2006).

Table 3. Estimated $PM_{2.5}$ Penetration Factors From the RIOPA and PTEAM Studies

Study	Location	Sample Size	Mean Penetration Factor	Reported 95% CI for Penetration Factor	Season
Relationships of Indoor, Outdoor, and Personal Air (RIOPA) Study[a]	Overall	268	0.91	(0.71, 1.12)	spring summer fall winter
	Overall (I/O<1)[b]	165	0.73	n/a	
	Overall (No indoor sources)	21	0.78	n/a	
	Los Angeles	112	1.04	(0.75, 1.33)	
	Houston	76	1.35	(0.46. 2.23)	
	Elizabeth	80	0.73	(0.42, 1.05)	
Particle Total Exposure Assessement Methodology (PTEAM) Study[c]	Los Angeles	293	1.00	(0.89, 1.11)	fall

[a] Sources: Weisel *et al.* (2005); Turpin *et al.* (2007)

[b] I/O<1: indoor concentration is less than outdoor concentration

[c] Sources: Özkaynak *et al*. (1996)

Table 4. Estimated $PM_{2.5}$ Deposition Rates From the RIOPA and PTEAM Studies

Study	Location	Sample Size	Mean Deposition Rate (h^{-1})	Reported 95% CI for Deposition Rate	Season
Relationships of Indoor, Outdoor, and Personal Air (RIOPA) Study[a]	Overall	268	0.79	(0.18, 1.41)	spring summer fall winter
	Overall (I/O<1)[b]	165	0.20	n/a	
	Overall (No indoor sources)	21	0.40	n/a	
	Los Angeles	112	0.90	(0.53, 1.28)	
	Houston	76	0.99	(-1.38. 3.35)	
	Elizabeth	80	0.46	(-0.44, 1.36)	
Particle Total Exposure Assessement Methodology (PTEAM) Study[c]	Los Angeles	293	0.39	(0.22, 0.55)	fall

[a] Sources: Weisel *et al.* (2005); Turpin *et al.* (2007)

[b] I/O<1: indoor concentration is less than outdoor concentration

[c] Sources: Özkaynak *et al*. (1996)

Table 5. Input Assumptions for Sensitivity Analysis of P, k, and ACH

Input	Distribution
P_1	N (0.85, 0.075)[a]
P_2	N (0.90, 0.05)
P_3	Tri (0.70, 0.78, 1.0)[b]
k_1 (h^{-1})	N (0.40, 0.10)
k_2 (h^{-1})	N (0.39, 0.085)
ACH_1 (h^{-1}) in summer in TX[c]	Log (1.05, 2.49)
ACH_2 (h^{-1}) in summer in TX[d]	Log (0.37, 1.90)

[a] N~ Normal (mean, standard deviation)

[b]Tri ~ Triangular (minimum, mode, maximum).

[c] Log ~ lognormal (13.8, 3.1);

[d] Sources: Murray and Burmaster (1995)

Table 6. Factors Affecting Geographic Variability in Daily Average $PM_{2.5}$ Exposure

Air Exchange Rate for Summer (h^{-1})[a]						
New York City			Harris County, TX		Six Counties Region, NC	
$\mu_g = 0.64\ h^{-1}$, $\sigma_g = 2.09$			$\mu_g = 0.37\ h^{-1}$, $\sigma_g = 1.90$		$\mu_g = 0.54\ h^{-1}$, $\sigma_g = 1.70$	
Distribution of Smoking Prevalence by Gender and Age[b] (%)						
	New York City		Harris County, TX		Six County Region, NC	
Age	Male	Female	Male	Female	Male	Female
12-13	4.0	4.0	9.7	6.5	9.4	8.8
14-15	12.0	12.0	10.6	8.0	11.5	12.1
16-17	22.0	18.0	13.5	11.0	20.7	15.1
18-24	28.8	23.4	27.7	20.2	42.3	30.5
25-34	28.5	23.1	27.6	19.1	36.4	20.9
35-44	23.9	18.5	30.0	22.5	29.3	30.5
45-64	24.4	21.7	27.8	20.3	27.7	20.9
>64	12.0	6.6	16.7	9.2	18.5	10.5
Distribution of Population by Gender and Age[b] (%)						
	New York City		Harris County, TX		Six County Region, NC	
Age	Male	Female	Male	Female	Male	Female
12-13	3.6	3.6	3.8	3.9	3.4	3.4
14-15	3.6	3.6	3.7	3.7	3.5	3.5
16-17	3.5	3.6	3.7	3.8	3.6	3.6
18-24	3.3	3.5	3.8	3.7	3.9	3.9
25-34	7.9	8	8.4	8.6	8.3	8.4
35-44	8.8	8.9	9.9	10.2	9.2	9.3
45-64	12.3	12.4	11.3	11.4	11.8	11.9
>64	6.5	6.9	4.8	5.3	5.9	6.4
Distribution of Housing Types[c] (%)						
Housing Type[d]	New York City		Harris County, TX		Six County Region, NC	
Single Family Detached	9.5		68.3		62.5	
Single Family Attached	7.2		1.2		4.8	
Multiple Family	83.2		8.8		27.2	
Mobile Home	0.1		21.7		5.5	

[a] Distribution type: lognormal distribution, μ_g: geometric mean, σ_g: geometric standard deviation (Koontz and Rector 1995; Weisel *et al.*, 2005; Wallace *et al.*, 2006).

[b] DHHS (2008)(36): adults 18 years and older; SAMSHA (2008)(39): adolescents 12-17 years old.

[c] U.S. Census 2000. The indoor volumes are of single family detached; of single family attached; of multiple family; of mobile home.

[d] Average Indoor volume: Single Family Detached: 466 m^3; Single Family Attached: 371 m^3; Multiple Family: 241 m^3; Mobile Home: 222 m^3.

Table 7. Distribution of Daily Average Time Spent Outdoors and Indoors by Gender and Age[a]

	Time Spent Outdoors Per Day (hr) [b]		Time Spent Indoors Per Day (hr) [c]		Travel Time Per Day (hr) [d]	
Age	Male	Female	Male	Female	Male	Female
12-13	0.9	0.4	19.5	21.1	3.6	2.5
14-15	0.6	0.3	18.4	18.9	5.0	4.8
16-17	0.8	0.2	19.5	18.3	3.7	5.5
18-24	0.8	0.2	20.0	20.6	3.2	3.2
25-34	1.0	0.3	20.5	20.0	2.5	3.7
35-44	1.1	0.2	20.5	20.0	2.4	3.8
45-64	1.0	0.3	21.1	21.1	1.9	2.6
>64	1.0	0.3	21.6	22.7	1.4	1.0

[a] Source: Consolidated Human Activity Database (CHAD) (McCurdy *et al.*, 2005).

[b] Outdoor includes street, parking lot, gas station, park, playgrounds, pool, farm, and all other outdoor microenvironments

[c] Indoor includes home, office, school, store, bar, restaurant, and all other indoor microenvironments.

[d] Travel includes travel by car, truck, motorcycle, bus, train, subway, airplane, boat, walking, bicycle, and waiting for travel either indoors or outdoors.

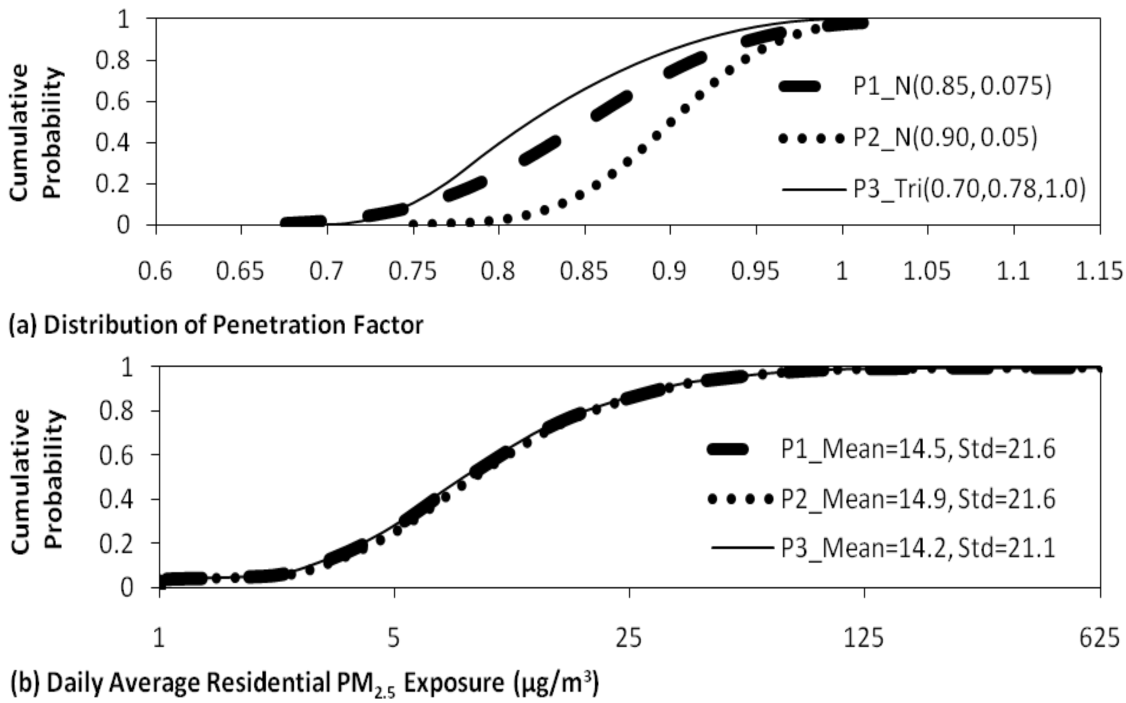


Figure 1. Variability in Penetration Factor and Daily Residential $PM_{2.5}$ Exposure for Harris County, TX for July 2002

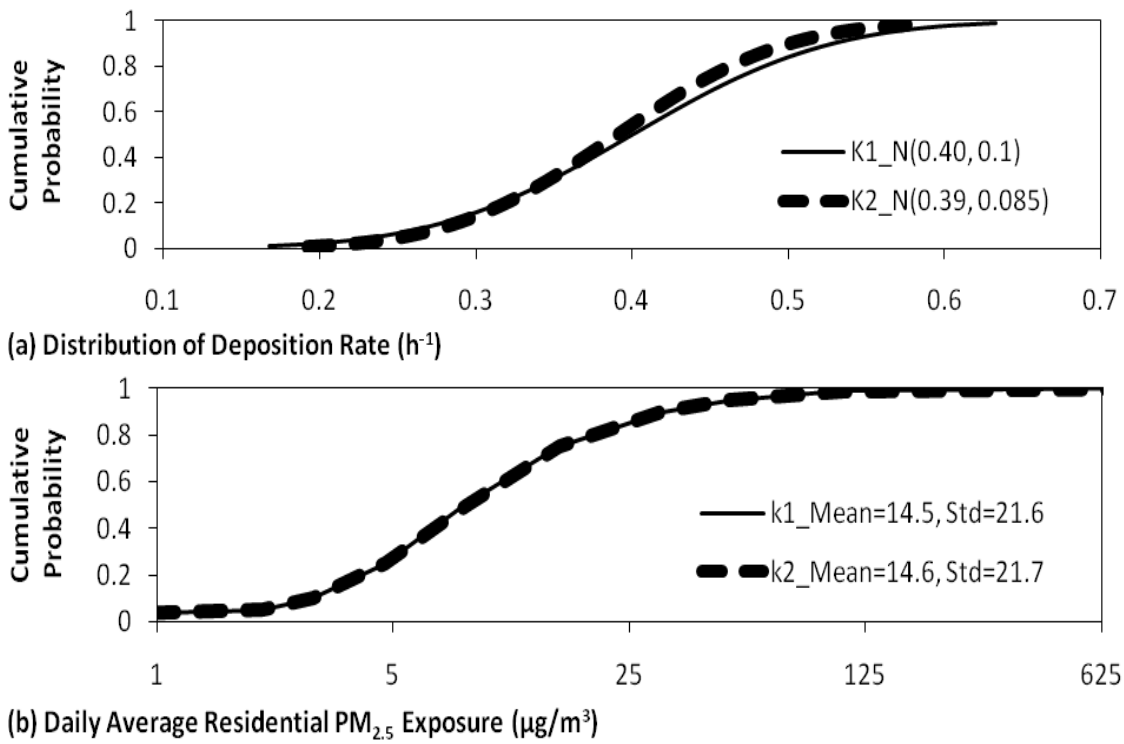


Figure 2. Variability in Deposition Rate and Daily Residential $PM_{2.5}$ Exposure for Harris County, TX for July 2002

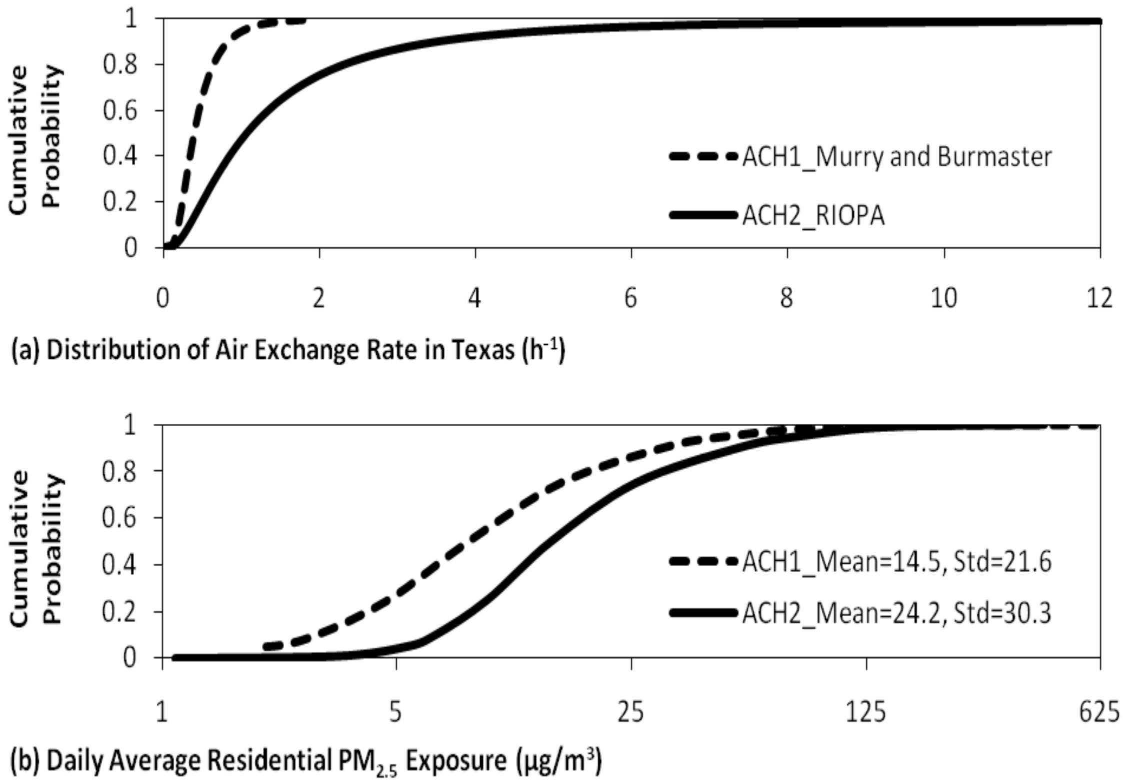


Figure 3. Variability in Air Exchange Rate and Daily Residential $PM_{2.5}$ Exposure for Harris County, TX for July 2002

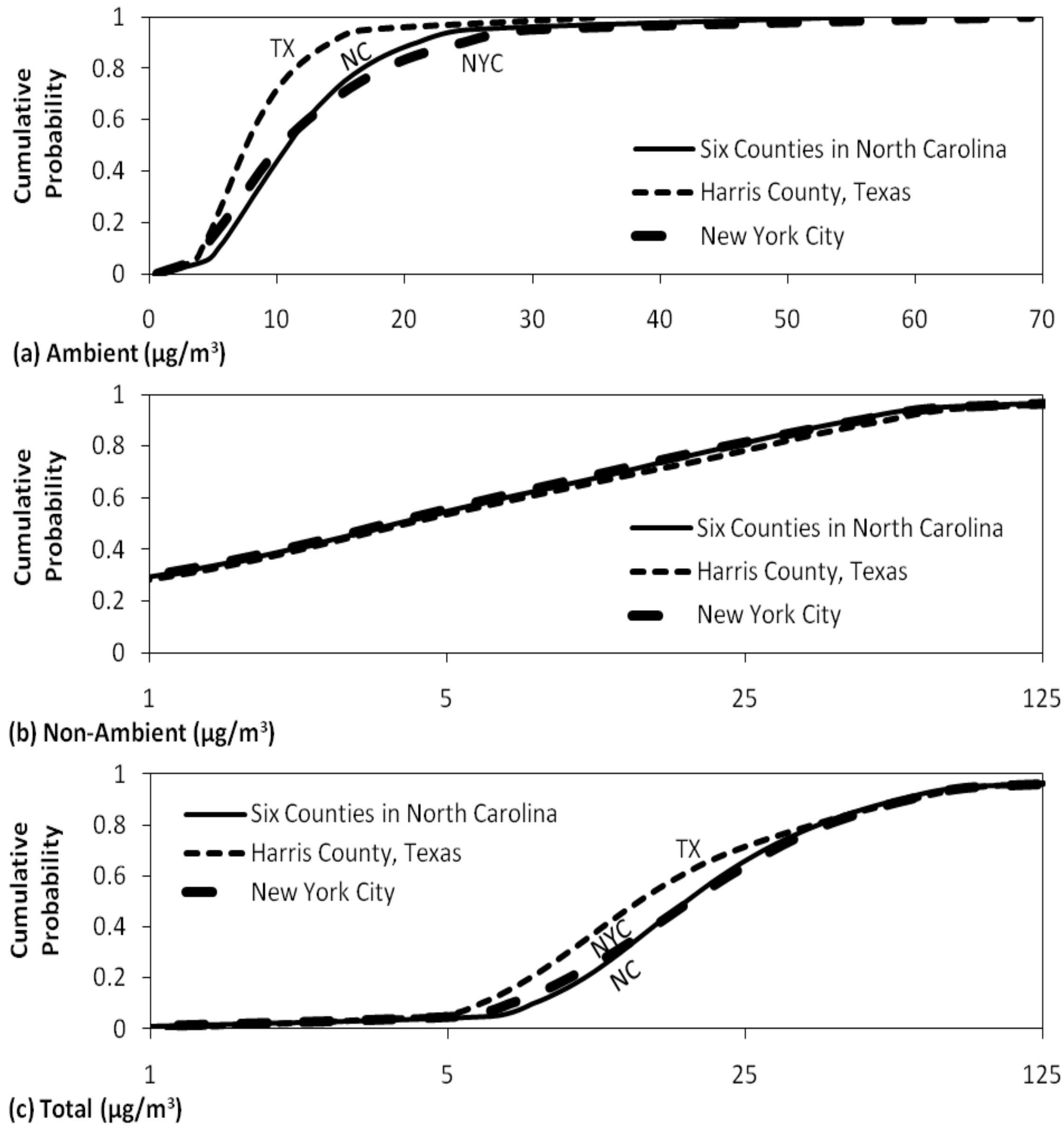
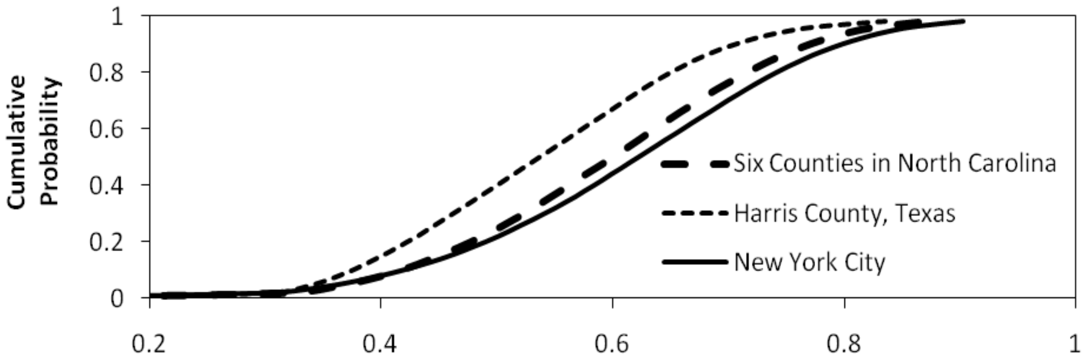
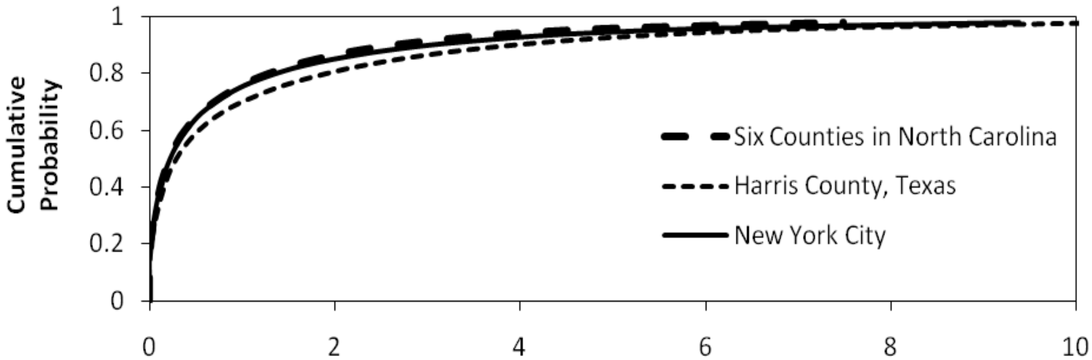


Figure 4. Geographic Variability of Inter-Individual Variability in Daily Average $PM_{2.5}$ Exposure in Summer

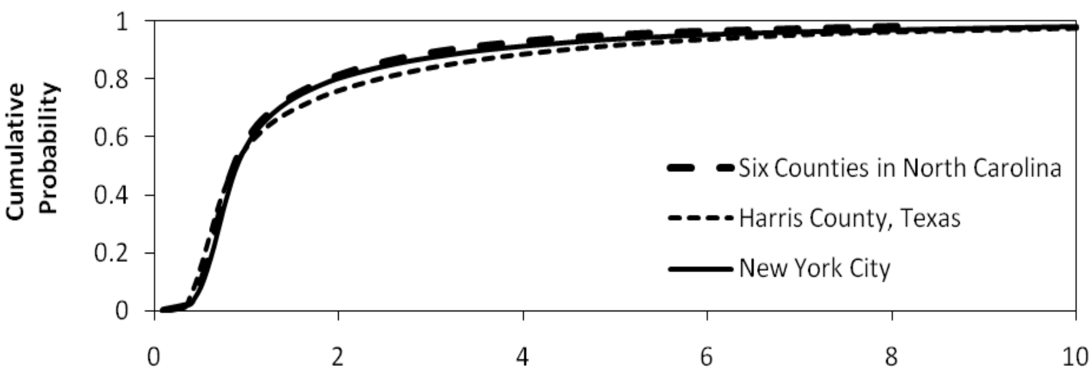
Note: in (b), non-ambient exposure is zero below 10^{th} percentile



(a) Ratio of Daily Average Ambient Exposure to Ambient Concentration (E_a/C)



(b) Ratio of Daily Average Non-Ambient Exposure to Ambient Concentration (E_{na}/C)



(c) Ratio of Daily Average Total Exposure to Ambient Concentration (E_t/C)

Figure 5. Inter-Individual Variability of Exposure to Concentration Ratio (E/C) in Summer

PART V

CONCLUSIONS AND RECOMMENDATIONS

This chapter gives key findings, conclusions and recommendations.

1.0 FINDINGS

This section presents key findings regarding the algorithms and inputs in SHEDS-PM: (1) ETS algorithms and inputs for different microenvironments; (2) in-vehicle ETS algorithms and inputs; and (3) key inputs for the residential microenvironment.

1.1 ETS Algorithms and Inputs in SHEDS-PM

A mass balance and a linear regression approach for calculating the residential, restaurant and bar $PM_{2.5}$ concentration including ETS, respectively, are generally based on the best practice.

SHEDS-PM default inputs regarding the proportion of smokers and "other smokers" should be updated to account for the desired time period for which exposures are simulated, since there are significant differences in these proportions over time. The default data regarding cigarette emission rate is low compared to average emission rates estimated from several studies, and thus should be updated.

1.2 In-Vehicle ETS Algorithms and Inputs in SHEDS-PM

A mass balance equation can be incorporated into the current linear regression model for the in-vehicle microenvironment in SHEDS, to take into account the contributions of ETS to the total exposure.

Inputs for the mass balance model include: R_{cig}, E_{cig}, ACH, k, and V. Suggested distributions of R_{cig}, E_{cig} and V are given in the thesis. Ranges of ACH and k for various vehicle operating scenarios are summarized.

1.3 Residential Microenvironment Inputs in SHEDS-PM

Data regarding ACH varies in different geographic areas. P and k varies in different seasons. Therefore, proposed inputs of P and k are summarized for four seasons. Summer ACH is summarized for three geographic areas of New York City, Harris County in Texas, and Wake, Durham, Orange, Alamance, Guilford, and Forsyth County in North Carolina.

2.0 CONCLUSIONS

This section presents key conclusions regarding inter-individual variability of $PM_{2.5}$ exposure from ETS, in-vehicle ETS exposure, and the geographic variability of $PM_{2.5}$ exposure in the NYC, TX, and NC domains.

2.1 Inter-Individual and Geographic Variability in Human Exposure to $PM_{2.5}$ in ETS

Inputs and parameters regarding the proportion of smokers and “other smokers,” and cigarette emission rate for the residential indoor microenvironment, and to the incremental increase in indoor $PM_{2.5}$ concentration associated with smoking a cigarette during an hour in the restaurant and bar microenvironments are the ones that merit the most attention when developing input data.

For a population of individuals, exposure to ETS can be the largest single contributor to daily average exposure to fine particulate matter, even though only a portion of all individuals are exposed to ETS. For those who are exposed to ETS, there is a wide range of variability in such exposures.

Geographic variability in the prevalence of smokers and demographic factors such as the distribution of the population by age and gender are among factors that lead to geographic variability in daily average $PM_{2.5}$ exposures attributable to ETS. Thus, area-specific data for the proportion of smokers and for demographics should be used.

2.2 Human Exposure to In-Vehicle $PM_{2.5}$ from ETS

ACH is distinguishably the most important input. R_{cig} is slightly more important than E_{cig}, and V, which are of comparable importance to each other. k turn to be the least important. ACH and are k are correlated, and vary based on the behavior of drivers and passengers, and the filter efficiency among different vehicles, respectively. Therefore, data regarding the in-vehicle ACH and k are required for different vehicle scenarios.

Because of the variability in ACH, k and small vehicle interior volume, in-vehicle ETS $PM_{2.5}$ concentration can be higher than that in other microenvironments, especially for a typical scenario in summer, such as windows closed and AC on.

Occupational in-vehicle exposure attributable to ETS can be more than 10 times higher than the average in-vehicle ETS exposure. Sixty five percent of people exposed to ETS in the vehicle microenvironment are not smokers, depends on the time spend in the vehicle, the exposures for these non-smokers can be very high.

2.3 Geographic Difference in Inter-Individual Variability of Human Exposure to $PM_{2.5}$

For a single model domain, ACH is distinguishably the most sensitive input for both ambient and non-ambient exposure to $PM_{2.5}$, especially in combination with strong indoor emission sources. High ACH leads to high ambient exposure indoors but lower non-ambient exposure, and vice versa.

Approximately 60 percent of total daily average exposures to $PM_{2.5}$ are caused by non-ambient exposures for each of the three geographic areas. The extremely high daily average exposures for some individuals are mainly caused by non-ambient exposure to smoking or cooking. Geographic differences in housing stock and climate, which lead to geographic differences in ACH, account for geographic variability in the estimated ambient and non-ambient exposure to $PM_{2.5}$.

The distributions of E_a/C ratios in each geographic area indicate that simulated individuals are typically exposed to more than half of the modeled ambient concentrations. The 95 percent frequencies of E_a/C range approximately from 0.5 to 0.8. The average E_a/C ratio varies among geographic areas based on differences in housing stock, climate, and seasonal ACH. Thus, on average, individuals in different geographic areas are exposed to different proportions of ambient $PM_{2.5}$ by 13 percent.

3.0 RECOMMENDATIONS

This section presents key recommendations regarding data requirement, and further studies in human exposure to $PM_{2.5}$ from ETS, in-vehicle ETS exposure, and geographic and inter-individual variability of $PM_{2.5}$ exposure.

3.1 Human Exposure to $PM_{2.5}$ in ETS

The proportion of smokers and the number of cigarettes smoked per smoker per day appear to be declining with time in the U.S., they are still significant and should be tracked consistently over time by age and gender. Some demographic factors that affect smoking prevalence, such as education or socioeconomic status, are not incorporated into existing exposure models.

Exposure of young children to ETS and data on the proportion of households in which smoking occur merit quantification. Data are not currently available for avoidance behaviors, such as a non-smoker who avoids proximity to a smoker during a smoking event. Furthermore, changes in smoker activity patterns due to bans on smoking in public indoor spaces, such as whether the rate of smoking is differentially affected in other microenvironments, are not yet quantified.

3.2 Modeling of Human Exposure to In-Vehicle $PM_{2.5}$ from ETS

Inter-individual variability for in-vehicle ETS exposure is associated with vehicle scenarios, which tend to vary with traffic conditions, and ambient air quality, and therefore will vary seasonally and geographically. Thus, data regarding seasonal and geographic variability in vehicle operation conditions are required for further investigation.

3.3 Geographic Difference in Inter-Individual Variability of Human Exposure to $PM_{2.5}$

Recommended values of P, k and ACH are given. However, there are relatively few data from which to estimate P and k. Data are needed for P and k that more clearly represent situations without indoor emissions. Furthermore, more recent data for ACH are needed to represent current housing stock in different regions.

Most epidemiology studies are using central site ambient monitoring data as a surrogate for population-level exposure. Area-specific E_a/C ratios are recommended for possible use in epidemiology studies to better address geographic and inter-individual variability in exposure to $PM_{2.5}$. The differences in average E_a/C may help to explain geographic variations in dose-response functions.