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

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Abstract

Environmental tobacco smoke (ETS) is estimated to be a significant contributor to in-vehicle human exposure to fine particulate matter of 2.5 μm 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. Cigarette smoking and emission rates, and vehicle interior volume, are also key inputs. 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 near-road incremental PM_{2.5} concentration from on-road emissions. Results of probabilistic study indicate that ETS is a key contributor to the in-vehicle average and high-end exposure. Factors that mitigate in-vehicle ambient PM_{2.5} exposure lead to higher in-vehicle ETS exposure, and vice versa.

Keywords

tobacco smoke; vehicles; exposure; PM_{2.5}; variability; inhalation

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) and with development of childhood asthma (Bukowski *et al.*, 2002).

High exposures to smoking are not only to smokers, but to other people who are in the presence of smokers, including children and elderly people (Rees and Connolly 2006).

Second-hand smoke exposures in cars to Irish children of 13 to 14 years old were found to increase the likelihood of respiratory and allergic symptoms (Kabir *et al.*, 2009) Rees and Connolly (2006) found that private passenger cars can have high levels of SHS under normal use and conclude that their data can be used to help promote smoke-free environments for children. MacKenzie and Freeman (2010) conclude that exposure to SHS poses significant risk to children, particularly since they are not able to remove themselves from such a smoky environment. Freeman *et al.* (2008) report on the history of smoking bans in cars carrying children in two Australian states, and report that fine particulate levels in a car can be twice that of a “smoky” bar. Based on these considerations, there is a need 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.

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

The objectives of this paper are to: (1) recommend values of inputs for a selected mass balance model; (2) conduct sensitivity analysis to identify the key inputs for the model; and (3) characterize the potential magnitude and variability of in-vehicle exposures to ETS.

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 (USEPA 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 µg/m³ (Ott *et al.* 2008).

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 is costly (Wallace 1987; Thomas *et al.* 1993; Sexton *et al.* 1995; Jenkins *et al.* 1996).

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. ACH is calculated based on the decay rate of the concentration of a tracer gas such as SF₆. The average ACH from time t_0 to t_1 is (Spengler *et al.* 2000):

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

Where,

ACH = air exchange rate (h⁻¹);

C (t_0) = initial tracer gas concentration (µg/m³);

C (t_1) = tracer gas concentration at time t_1 (µg/m³);

t_0 = initial time (h); and

t_1 = final time (h).

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 the deposition of particles on the interior surfaces of the vehicle (Ott *et al.* 1992b, 1996):

$$k = \Phi_p - ACH \quad (2)$$

Where,

Φ_p = decay rate (h⁻¹);

k = deposition rate (h⁻¹).

The deposition rate (k) 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 (Abadie *et al.* 2004).

Modeling of In-vehicle Concentration

In order to generalize the results of measurements, a model for in-vehicle PM_{2.5} concentrations can be combined with a scenario-based microenvironmental stochastic model that simulates the movement of individuals 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.

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). Derivation and assumptions for SCEM 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 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). Many mass balance models for estimating indoor air quality are based on SCEM. The application of mass balance models has been done in various studies conducted over the last 20 years (Nazaroff and Klepeis, 2003; Spengler *et al.* 2000; Ott *et al.* 1992b, 2008; Nazaroff *et al.* 1989)..

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.

METHODOLOGY

The methodology includes: (1) literature review for model input data; (2) sensitivity analysis to identify key factors to which exposure is sensitive for ETS-related $PM_{2.5}$ concentration in vehicles; and (3) 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.

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) was used (Frey and Patil 2002). NRSA is a relatively simple method that is easily applied. It works well with linear models and when the plausible ranges are available for each input. The results of this approach can be used to rank order key inputs. 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.

Scenario-Based Modeling of In-Vehicle ETS Exposure

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):

$$C_{iv} = r_{iv/o} \cdot C_{amb} + B \quad (3)$$

Where,

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

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

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

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

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). The ratio $r_{I/O}$ represents the proportion of $PM_{2.5}$ per unit volume that penetrates from the ambient air to the vehicle interior.

SHEDS-PM can quantify inter-individual variability in daily average exposures 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. In case study applications given later, 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 from all demographic groups and all census tracts of Wake County, North Carolina. Exposures to ambient $PM_{2.5}$ are estimated based on July 2002 gridded concentrations estimated using the Community Multiscale Air Quality (CMAQ) model, updated with available monitoring data using Bayesian statistical methods (McMillan *et al.* 2010). Demographic and housing data are sampled from a database from the 2000 U.S. Census. Human activity patterns are sampled from diaries of the Consolidated Human Activity Database (CHAD). Details of SHEDS are reported elsewhere (Burke *et al.* 2001). Figure 1 is a conceptual diagram of the simulation of residential and in-vehicle ETS exposure in SHEDS-PM.

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, 11,760 of the simulated individuals are 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 in-vehicle exposure to ETS for a simulated individual is estimated by multiplying the SHEDS output for the daily average vehicle microenvironmental exposure by the ratio of in-vehicle concentration to near-road ambient concentration ($R_{I/O}$), 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 the in-vehicle $PM_{2.5}$ concentration caused by ETS (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) (Liu *et al.* 2010).

Variability of In-Vehicle ETS Exposure

In order to explore the significance of in-vehicle ETS exposure for various operating scenarios, and develop upper-bound estimates of exposure, a conservative point estimate-based analysis was conducted for three selected vehicle operating scenarios with different vehicle speeds, status of windows and HVAC system operation.

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.

Probabilistic Study for In-Vehicle ETS Exposure

In order to consider the uncertainty and variability of possible in-vehicle ETS exposures, a probabilistic case study was conducted to quantify inter-vehicle variability in several key parameters and to identify which of the selected parameters is most sensitive and for which more or better data would be useful.

Analytica 4.2 was used in the probabilistic study. Quantities in *Analytica* can be specified using a probability distribution function (Chrisman *et al.* 2010). Uniform, lognormal, and triangular distributions are specified for R_{cig} , E_{cig} , and V , respectively. The ranges of each distribution function are given in the Results section. Variation of C_{ETS} was estimated using Monte Carlo simulation with a simulation sample size of 18,535, which corresponds to the number of persons simulated to be exposed to ETS.

Sensitivity in a model output is identified using sample and rank correlation coefficients between the simulated frequency distribution of C_{ETS} and the simulated frequency distribution of each probabilistic input. 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).

RESULTS

A mass balance model for estimating $PM_{2.5}$ concentration attributable to smoking in a motor vehicle is 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.

Mass Balance Model

The ambient $PM_{2.5}$ concentrations are dominated by ambient sources, such as power plants, motor vehicles, secondary sulfate, and secondary nitrate. Source apportionment and other air quality studies do not provide evidence of a detectable concentration of ETS in the ambient air (Chow *et al.* 2002; Laden *et al.* 2000). Therefore, 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 (Ott *et al.* 1992b; Nazaroff and Klepeis 2003):

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

Where,

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

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

C_{ETS} = $\text{PM}_{2.5}$ concentration caused by ETS ($\mu\text{g}/\text{m}^3$);

E_{cig} = emission rate for cigarette smoking ($\mu\text{g}/\text{cig}$);

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. $\text{PM}_{2.5}$ 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.

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 SHEDS-PM (Burke *et al.* 2001; Georgopoulos *et al.* 2001), in which R_{cig} varies uniformly from 0 to 3. Because we assume that a smoker will smoke, the minimum value of R_{cig} used here is increased from zero to one. There are no data available that enable quantification of different rates of smoking in vehicles versus other microenvironments.

Typical cigarette $\text{PM}_{2.5}$ emission rates reported in the literature are from 10.8 to 22.4 mg/cig (Ott *et al.* 1996). The mean $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$ 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 m³ (Ott *et al.* 2008). For example, the volume of a 2005 Toyota Corolla is 2.4 m³, a 2005 Ford Taurus sedan is 2.7 m³, a 1999 Jeep Grand Cherokee Limited is 3.0 m³, and a 1999 Lexus RX-300 (SUV) is 5.5 m³. 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 (USBTS, 2002). If we assume a typical V= 3 m³ for cars, and V=5 m³ for light trucks, a best estimate is approximately 3.8 m³.

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 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 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.

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, which is larger than for any of the other inputs.

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 µg/m³. 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. The deposition rate covaries with ACH.

Variability of In-Vehicle ETS Exposure

ETS concentrations are very high in some typical vehicle operation scenarios as illustrated in Table 1. Therefore, case studies were conducted to characterize the potential magnitude of daily average 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 (µg/m³);

C_{NR} = near-road incremental PM_{2.5} concentration (µg/m³); and

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

The model is evaluated by comparing the estimated PM_{2.5} concentration to the experimental data from Offermann *et al.*, 2003. The estimated in-vehicle PM_{2.5} concentration with ETS agreed to within 20 percent of measured values in two different ventilation modes: (1) windows open, ventilation off; and (2) windows closed ventilation on.

The time weighted in-vehicle total exposure is estimated 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. A random sample of 500 individuals in each of 105 census tracts was simulated to characterize activity patterns, including smoking status, and commuting. The average ambient PM_{2.5} concentration during the July 2002 simulation time period was 21.2 µg/m³. The longitudinal simulation type is used in all model runs, which accounts for autocorrelation 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 (Liu *et al.* 2010). 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, traffic volume, atmospheric stability class, and wind speed. Liu *et al.* (2010) reported that C_{NR} is estimated at 6.6 µg/m³ based on a local road with 4 lanes, and 35.0 µg/m³ based on a highway with 8 lanes. However, traffic volume varies based on the day of the week. The ratio of Vehicle Miles of Travel (VMT) between weekday and weekend, and Saturday and Sunday are each 1.2 (Hu and Reuscher 2001; FHWA *et al.* 2005). Therefore, the C_{NR} reported by Liu *et al.* (2010) is revised to account for variations in traffic volume by day of the week. C_{NR} is estimated to be 7.1, 5.9 and 4.9 µg/m³ for weekday, Saturday, and Sunday, respectively, in Case A; and 37.3, 31.1 and 25.9 µg/m³ 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 SHEDS-PM 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 penetration of area-wide and near-road $PM_{2.5}$ ranges from 0.4 to 4.7 $\mu g/m^3$ for the 50th to 99th percentiles, however, the ETS exposure has a 99th percentile of 1180 $\mu g/m^3$. The very large value of the high end ETS exposure is attributed to low values of ACH, k, V, and upper bound values of R_{cig} and E_{cig} .

Because only 35 percent of the simulated population is exposed to ETS in a vehicle, ETS exposure is zero in the 50th percentile. The 99th percentile of ETS exposure is 11 times higher than the mean. This variability is attributed to variation in the time spent in-vehicle based on CHAD data, which has a 95 percent frequency range of 15 to 253 minutes per day, with a mean and standard deviation of 96, and 103 minutes, respectively. The time spent in vehicle at the 99th percentile is 6.5 hours per day, which is indicative of an occupational exposure. The duration of in-vehicle travel time for non-smokers traveling with smokers in the vehicle has a 95 percent frequency range of 11 to 209 minutes. The ETS exposures to these non-smokers can be very high.

In Case A, the average in-vehicle exposure attributable to the penetration of ambient $PM_{2.5}$ is lower than those of other microenvironments. However, the average in-vehicle exposure attributable to ETS is more than 5 times higher than the total exposure in the residential microenvironment.

In Case B, the average in-vehicle exposure attributable to ambient $PM_{2.5}$ is more than twice as high as that of Case A. This is because C_{NR} and $R_{I/O}$ are higher than for Case A. Conversely, the average in-vehicle ETS exposure 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 Case A. Of the 100.5 $\mu g/m^3$ average decrease in mean ETS exposure, 67% is because of the increase in V.

In Case B, the average in-vehicle exposure attributable to ambient $PM_{2.5}$ is approximately equal to those of store and office, and lower than those of other microenvironments. The average in-vehicle ETS exposure is comparable to the total exposure in bars and higher than those of other microenvironments, except for home.

In Case C, the average in-vehicle exposure attributable to ambient $PM_{2.5}$ is comparable to that of ETS. This is due to the higher ACH, k, and lower R_{cig} in Case C than those of Cases A and B. The 99th percentile exposures to ambient $PM_{2.5}$ and ETS are factors of 6 and 11, respectively, higher than the corresponding mean values. Of the 104.6 $\mu g/m^3$ average decrease in mean ETS exposure from Case A to C, 82% is because of the increase in ACH. The average Case C total exposure of 6.4 $\mu g/m^3$ is greater than that of all microenvironments except bar and home.

Probabilistic Case Studies

An alternative set of case studies explore the possible inter-vehicle variability in C_{ETS} based on limited data available for variability in R_{cig} , E_{cig} , and V.

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 m³, and a range of 2 to 6 m³. Because

ACH, k , and $R_{I/O}$ are based on the vehicle operating scenarios of Case A, B, and C, these inputs are set as constant values for each Case. However, unlike the point estimate case studies A, B, and C in which E_{cig} , R_{cig} , and V varied, in the probabilistic case studies A-P, B-P, and C-P the same distributions of these three variables are used.

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 for Cases A-P, B-P, and C-P and as frequency distributions of inter-individual variability in Figure 3.

There is no difference between the probabilistic and point estimate case studies with respect to exposure factors related to ambient exposure. Therefore, the ambient exposure results are the same.

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 because 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 decrease of in-vehicle ETS.

The ETS exposure in the 90th and 99th percentiles of Case A-P is lower than that of Case A. Case A is based on worst case bounding estimates of R_{cig} , E_{cig} , and V , for which C_{ETS} is 4500 $\mu g/m^3$. However, in Case A-P, the 99 percent frequency range of C_{ETS} is from 400 to 2450 $\mu g/m^3$, with a mean of 970 $\mu g/m^3$. Since the distributions of R_{cig} , E_{cig} , and V are independent, the chance of simultaneously obtaining worst case combinations of high R_{cig} , high E_{cig} , and low V is low. The standard deviation of ETS exposure in Case A is greater than that of Case A-P, because Case A is based on an extreme range of either zero ETS concentration for those not exposed to a very high C_{ETS} of 4500 $\mu g/m^3$ for all who are exposed.

In Case B-P, the average in-vehicle ETS exposure is 71 percent lower than that of Case A-P. Since the inputs for R_{cig} , E_{cig} , and V are the same for Cases A-P and B-P, the decrease in average ETS exposure is attributed to the joint increase in both ACH and k .

The average in-vehicle ETS exposure in Case B-P is 46 percent higher than that of Case B. In Case B-P, the 99 percent frequency range of C_{ETS} is from 240 to 1450 $\mu g/m^3$, with a mean of 580 $\mu g/m^3$. Case B has a constant C_{ETS} of 310 $\mu g/m^3$. Therefore, the mean and upper percentiles of in-vehicle ETS exposures for Case B-P are higher than for Case B.

In Case C-P, the average in-vehicle ETS exposure is more than 6 times lower than that of Case A-P, which is due to 1200 and 110 percent increase in ACH, and k , respectively. The average in-vehicle ETS exposure in Case C-P is 8 percent higher than that of Case C. In Case C-P, the 99 percent frequency range of C_{ETS} is from 63 to 380 $\mu g/m^3$, with a mean of 150 $\mu g/m^3$. In contrast, Case C has a constant C_{ETS} of 140 $\mu g/m^3$, which is lower than the mean of Case C-P.

Depending on the variation of ACH, and k among Cases A-P, B-P, and C-P, the average in-vehicle ETS exposure ranges from 4 to 24 $\mu g/m^3$, 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.

The distributions of inter-individual variability in daily average in-vehicle exposure in Figure 3 are each based on two main components: ambient exposure and ETS exposure. In Case A-P, these two components are well separated, with a noticeable inflection point at a cumulative frequency of 0.65. ETS exposure is zero below the 65th percentile. ACH and k

for Case A-P lead to relatively low penetration of ambient $PM_{2.5}$ into vehicles; thus, the lower ambient exposure component for Case A-P has lower values than for the other cases. Conversely, the low ACH and k of Case A-P lead to retention of ETS in the vehicle cabin and relatively large values of ETS exposure above the 65th percentile.

For Case B-P, separation between the components of the population exposed only to ambient $PM_{2.5}$ versus those also exposed to ETS is also apparent, but the separation between these components is less than that for Case A-P. The higher values of ACH and k lead to greater penetration of ambient $PM_{2.5}$, and also lower ETS concentrations.

For Case C-P, ACH and k are very large compared to Cases A-P and B-P. Thus, there is greater penetration of ambient $PM_{2.5}$ but also better dispersion of ETS. Although the results of Case C-P are comprised of a mixture of subpopulations of persons not exposed to ETS and those exposed to ETS, there is substantial overlap such that these two components are not well separated as for the other cases.

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

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. ACH and k are correlated. In-vehicle ETS exposure are very sensitive to the joint changes of ACH and k . Conditions that lead to high ambient exposure in a vehicle are conducive to lowering ETS exposure, and vice versa. In order to improve the accuracy of the estimates of in-vehicle ETS exposure, data regarding ACH and k are required for different vehicle operating scenarios, such as the vehicle speed, and the status of windows and HVAC system.

In-vehicle ETS exposure varies 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.

For a typical summer scenario in which vehicle windows are closed and AC is on, in-vehicle ETS exposure is higher than that in other microenvironments, such as office, store, and outdoor, by a factor of 10. The average in-vehicle ambient exposure is lower than all of other microenvironments, except for the scenarios with very high ratio of in-vehicle to ambient $PM_{2.5}$ concentration. Examples of the latter are for situations in which windows are open or the HVAC system is using outside air.

Smoking under both open and closed ventilation conditions resulted in increased exposure to $PM_{2.5}$. However, ETS exposures are higher under the closed ventilation condition than with the ventilation from outside. This indicates that opening windows or maintaining adequate ventilation in a vehicle help to reduce the exposure.

Occupational in-vehicle exposure attributable to ETS can be more than 10 times higher than the average in-vehicle ETS exposure. Depending on the time spent in the vehicle, the ETS exposures to non-smokers who are in the presence of a smoker can be very high. Although smoking is a personal choice, ETS exposure to non-smokers, especially for children is of concern.

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FIGURE 1. Conceptual Diagram of the Distribution of Simulated Individuals with Respect to Environmental Tobacco Smoke 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, In-vehicle PM_{2.5} exposure include the contributions of ambient air, near-road incremental air, and in-vehicle ETS concentration.

FIGURE 3.
Inter-Individual Variability of Daily Average In-Vehicle Exposure to PM_{2.5}

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

^aSource: Ott *et al.*(2008).

^bThe decay rate (Φ_p) and ACH are measured.

^cDeposition rate (k) includes filter efficiency and ACH is estimated by $k = \Phi_p \cdot ACH$.

^dPM_{2.5} concentrations are calculated based on Equation (4), and $R_{cig} = 2$ cig/h, $E_{cig} = 13.8$ mg/cig, $V = 3.8$ m³, and ACH and k as shown.

TABLE 2

Sensitivity Analysis of the In-vehicle ETS Mass Balance Model

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

^aSources: Repace and Lowrey 1980; Sexton et al. 1995; MMWR 2005; Burke et al. 2001; Georgopoulos et al. 2001.

^bSources: Ott *et al.* 1996; Nelson 1994; Nazaroff and Klepeis 2003.

^cSources: Ott *et al.* (2008).

^dSources: Ott *et al.* (2008); USBTS 2002.

^eRelative ranges of predicted PM_{2.5} concentrations is based on nominal values using Equation (4).

TABLE 3Scenario 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 (μg/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 (μg/m ³)		4500	310	140

^aVehicle 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; FHWA, 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 Studies^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	0.4	0	1.5	336	4.7	1180	0.7	108	0.9	246
		B	1.1	0	2.9	23	9.1	82	1.5	7.5	1.7	17
		C	2.1	0	5.7	11	18	37	3.0	3.4	3.4	7.7
	Probabilistic Inputs ^c	A-P	0.4	0	1.5	71	4.7	267	0.7	24	0.9	60
		B-P	1.1	0	2.9	43	9.1	154	1.5	14	1.7	35
		C-P	2.1	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		

^aThe 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. ETS is modeled in the residential, restaurant, and bar microenvironments.

^bIn-vehicle PM_{2.5} exposure include the contributions of ambient air, near-road incremental air, and in-vehicle ETS concentration.

^cInput distributions: R_{cig} ~ Uniform (1.3); E_{cig} ~ lognormal (13.8, 3.1); V ~ Triangular (2.3, 8.6). For Uniform distribution, parameters are minimum and maximum. For Lognormal distribution, parameters are the arithmetic mean and arithmetic standard deviation. For Triangular distribution, the parameters are minimum, mode, and maximum.