ABSTRACT

JIAO, WAN. Assessment of Population and Microenvironmental Exposure to Fine Particulate Matter ($PM_{2.5}$). (Under the direction of Dr. H. Christopher Frey).

A positive relationship exists between fine particulate matter ($PM_{2.5}$) exposure and adverse health effects. $PM_{2.5}$ concentration-response functions used in the quantitative risk assessment were based on findings from human epidemiological studies that relied on area-wide ambient concentrations as surrogate for actual ambient exposure, which cannot capture the spatial and temporal variability in human exposures. The goal of the study is to assess inter-individual, geographic and seasonal variability in population exposures to inform the interpretation of available epidemiological studies, and to improve the understanding of how exposure-related factors in important exposure microenvironments contribute to the variability in individual $PM_{2.5}$ exposure. Typically, the largest percentage of time in which an individual is exposed to $PM_{2.5}$ of ambient origin occurs in indoor residence, and the highest ambient $PM_{2.5}$ concentrations occur in transportation microenvironments because of the proximity to on-road traffic emissions. Therefore, indoor residence and traffic-related transportation microenvironments were selected for further assessment in the study.

Population distributions of individual daily $PM_{2.5}$ exposures were estimated for the selected regions and seasons using the Stochastic Human Exposure and Dose Simulation Model for Particulate Matter (SHEDS-PM). For the indoor residence, the current practice by assuming the entire residence to be one large single zone for calculating the indoor residential $PM_{2.5}$ concentration was evaluated by applying an indoor air quality model, RISK, to compare indoor $PM_{2.5}$ concentrations between single-zone and multi-zone scenarios. For

the transportation microenvironments, one field data collection focused on in-vehicle microenvironment and was conducted to quantify the variability in the in-vehicle $PM_{2.5}$ concentration with respect to the outside vehicle concentration for a wide range of conditions that affect intra-vehicle variability in exposure concentration, including ventilation air source, window status, fan setting, AC utilization, vehicle speed, road type, travel direction, and time of day. Another field data collection measured $PM_{2.5}$ exposure concentrations on pre-selected routes across transportation modes of pedestrian, bus, and car to quantify the variability in the transportation mode concentration ratios, and identify factors affecting variability in traffic-related concentrations.

In general, population daily average exposure to ambient $PM_{2.5}$ is less than the ambient concentration by approximately half. The ratio of $PM_{2.5}$ ambient exposure to ambient concentration (E_a/C) varies by individual, geographic area and season, as a result of regional differences in housing stock and seasonal differences in air exchange rates (ACH). For the indoor residence, the single-zone assumption is biased when any non-ambient source is presented. Bias correction factors are developed for cooking and smoking scenarios, separately, to improve the concentration estimates. Correction factors are most sensitive to changes in ACH but relatively insensitive to variations in source emission rate and duration. In a SHEDS-PM case study, the population daily average total exposure increased by 17% after applying correction factors. Transportation mode exposure concentrations are sensitive to mode, and are affected by factors such as vehicle ventilation and proximity to on-road emission sources. The in-vehicle to outside vehicle concentration (I/O) ratio is highly

sensitive to whether windows are open or, for closed windows, to whether fresh air or recirculating air is used.

Both model simulations and field studies are needed to inform better understanding of human exposure. Exposure, and not just concentration, should be considered in developing risk management strategies to reduce uncertainty in health effect estimates, and to identify highly exposed groups and possible exposure reduction strategies.

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Assessment of Population and Microenvironmental Exposure to Fine Particulate Matter ($PM_{2.5}$)

by
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DEDICATION

This dissertation is dedicated to my parents for their long-term unselfish love and support to me. My wise dad, a professor with full of wisdom; my lovely mom, a doctor with the biggest love in the world, you were, are and will always be my sun in my life path. I hope you both would feel proud of your only daughter when she really grows up.

BIOGRAPHY

Wan Jiao was born in Xi'an, one of the oldest cities in China. She received her Bachelor of Science degree in Environmental Engineering (valedictorian) from Tianjin University of Science and Technology in 2007. To broaden and deepen her world views, Wan further obtained her dual Master of Arts degree in Sociology through a joint program offered by University of Saskatchewan, Canada, and Xi'an Jiaotong University, China in 2010. Being prepared with a multi-disciplinary knowledge background, she joined in the Department of Civil, Construction, and Environmental Engineering at North Carolina State University, Raleigh, NC, in August 2010, to pursue her PhD degree under the supervision of Dr. H. Christopher Frey. From either model simulations or field measurements, her research mainly focuses on population and micro-environmental exposure assessments for criteria pollutants such as fine particulate matter ($PM_{2.5}$) and carbon monoxide (CO). She has also participated in emission measurements for on-road light duty gasoline vehicles.

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PART I INTRODUCTION

1.1 Introduction

Particulate matter (PM) pollution is a mixture of microscopic solids and liquid droplets suspended in the air. PM comes in a variety of sizes and can be made up of many types of components, including acids (sulfates and nitrates), organic chemicals, metals, soil, or dust particles, and allergens (fragments of pollen or mold spores) (U.S. EPA, 2013). Of particular concern is a class of particles known as fine particulate matter ($PM_{2.5}$) that is comprised of particles 2.5 micrometers or smaller in aerodynamic diameter. They are small enough to enter the lungs and cross the blood-air barrier in the alveoli. Based on review of numerous studies, the U.S. Environmental Protection Agency (EPA) has identified causal associations between exposure to $PM_{2.5}$ and adverse human health effects including premature death in people with heart or lung disease, nonfatal heart attacks, irregular heartbeat, aggravated asthma, decreased lung function, and increased respiratory symptoms (U.S. EPA, 2009a). People with heart or lung diseases, children, and older adults are the most vulnerable subpopulations to be affected by particle pollution exposure (U.S. EPA, 2009a).

Exposure is defined as the contact of a chemical, physical, or biological agent with the outer boundary of a human body (U.S. EPA, 1992). Exposure differs from internal dose, which is the amount of a chemical absorbed upon crossing the boundary to biologically significant sites within the body (U.S. EPA, 1992). While exposure can be conceptually characterized as a linear function of concentration and time, the description of internal dose is more complex because internal dose may depend on inter-individual variability in physiological or other factors and is not usually just a simple linear function of concentration (U.S. EPA, 1992).

A challenging aspect of air pollution health effects studies is to properly quantify the exposures of individuals in the population. Ambient $PM_{2.5}$ concentrations are affected by meteorology and by changes in emission rates, and locations of emission sources. However, actual $PM_{2.5}$ exposure to ambient origin depends on the amount of time an individual spends in different microenvironments. Microenvironments are surroundings that can be treated as homogeneous or well characterized with respect to the concentrations of an agent (U.S. EPA, 1992). Microenvironments include various indoor locations (e.g. home, work, school, restaurant, and store), outdoors, in transit, and others. For indoor microenvironments, a portion of ambient $PM_{2.5}$ penetrates and deposits to interior surfaces.

Exposure concentration is the concentration with which a person comes into contact (U.S. EPA, 1992). An individual's time-weighted $PM_{2.5}$ exposure can be quantified as the airborne $PM_{2.5}$ exposure concentration integrated over a given time period based on the person's activities (U.S. EPA, 2009a):

$$E_T = \int C_j dt \tag{1}$$

Where E_T = total exposure over a time period of interest, C_j = airborne PM concentration at microenvironment j, and dt = portion of the time period spent in microenvironment j. As indicated in Figure I-1, E_T can be decomposed to account for exposure to PM of ambient (E_a) and non-ambient (E_{na}) origin:

$$E_T = E_a + E_{na} \tag{2}$$

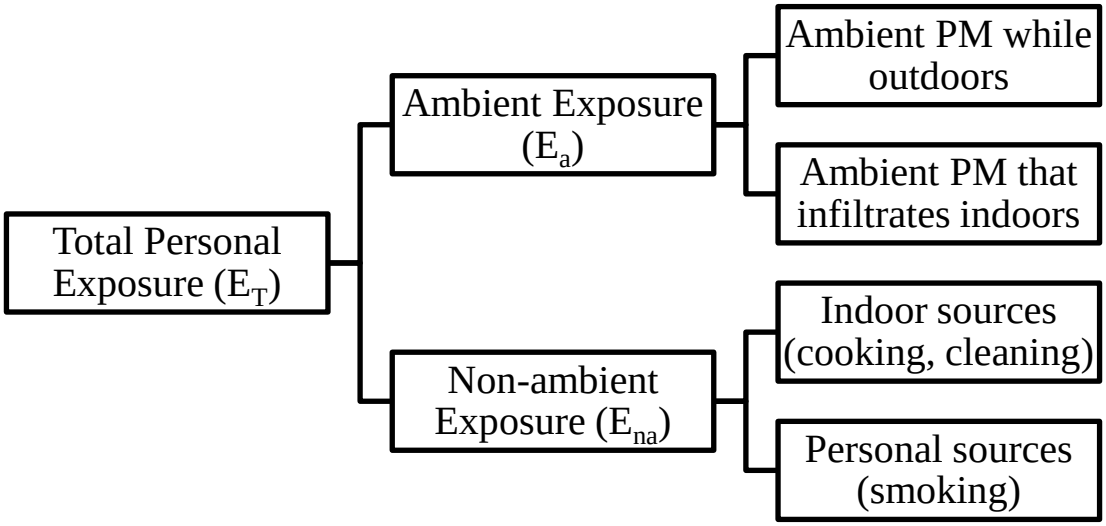

Figure I-1. Decomposition of Personal Exposure to PM

Previous studies generally found that individual daily values of total and non-ambient $PM_{2.5}$ exposure were poorly correlated with the daily ambient concentrations, but the individual daily ambient exposures were highly correlated with ambient concentrations (U.S. EPA, 2004). Thus, the separation of total PM exposures into ambient and non-ambient components reduces potential uncertainties in the analysis and interpretation of PM health effects data (U.S. EPA, 2009a).

Ambient PM sources include industrial and mobile source emissions, re-suspended dust, biomass combustion, and secondary formation. Non-ambient sources include smoking, cooking, home heating, cleaning, and indoor air chemistry (U.S. EPA, 2009a). PM concentrations from both ambient and non-ambient origin subject to spatial and temporal variability, and can affect exposure and resulting health effect estimates. In addition, the chemical composition of $PM_{2.5}$ can vary depending on the emission source, proximity to

sources, or as a result of formation of secondary $PM_{2.5}$. Furthermore, $PM_{2.5}$ of different chemical compositions can have different size ranges and thus may penetrate differently from outdoors to indoors (U.S. EPA, 2009b).

Population exposure assessment will be useful in characterizing the relationship between ambient concentration and population exposure, and the variability in population exposures. Both of these are important to the interpretation of community time-series epidemiological studies for establishing regulatory standards, such as the National Ambient Air Quality Standards (NAAQS). In the last revision of the PM NAAQS, EPA did not include an exposure assessment. This is in part because the health risks estimated using ambient concentrations provided adequate information on the change in health risks associated with a change in ambient concentrations, and a perception of the need for more research to provide insights on population exposures to identify various personal and building-related factors that may account for variability in $PM_{2.5}$-associated health risks (U.S. EPA, 2009a).

Since health outcomes depend on many community and individual level exposure factors, it is important for exposure assessment to focus on either vulnerable subpopulations that are more likely to receive higher exposure, or susceptible subpopulations that manifest more severe health effects for a given exposure due to personal, environmental, and socio-economic factors, including age, gender, race, socio-economic position, and pre-existing diseases (EPA, 2009a). For example, active smoker, occupational cooker, and people living near road are vulnerable subpopulations, while children, elderly, and people with pre-existing diseases are susceptible subpopulations.

The current NAAQS for $PM_{2.5}$ were based on evidence of health effects associated with short- and long-term exposure to fine particles from epidemiologic studies, and ambient concentration is typically used as a surrogate for personal exposure (U.S. EPA, 2009a). However, in a four-city exposure study, average personal exposures to ambient origin varied by individual, city and season, and were substantially less than the ambient concentrations (Sarnat *et al.*, 2009). Differences between personal ambient exposure and ambient concentration can result in the ordinal ranking of the mean concentrations and exposures in each of the locations or seasons that are not the same, which can lead to biased health risk estimates since the true relationships between exposure and response are not reflected in the concentration-response (C-R) functions.

Since people spend majority of daily time indoors, on average for a population, the daily exposure to particles of ambient origin is typically less than the ambient concentration, and the difference contributes to exposure errors (Zeger *et al.*, 2000). Using ambient $PM_{2.5}$ concentrations as a surrogate for the community average personal exposure to ambient $PM_{2.5}$ will negatively bias the estimation of health risk coefficients by the ratio of $PM_{2.5}$ ambient exposure to ambient concentration (E_a/C) (Chang *et al.*, 2012). These findings indicated how the selection of exposure metric can impact the risk estimates, and underscore the importance of characterizing the E_a/C ratio to accurately estimate exposure to ambient PM.

The E_a/C ratio depends on individual activity patterns, and factors affecting particle infiltration to indoor microenvironments, such as air exchange rate, penetration and deposition rate. Inter-individual variability in the estimated E_a/C ratio typically varies from 0

to 1. Assessing inter-individual variability in E_a/C will acknowledge variability in exposures within a population, and help to identify high-end exposure subgroups.

U.S.-based multicity epidemiological studies generally found that $PM_{2.5}$ effects differ by season and region, with greater effects observed in eastern U.S. and during warmer months in spring and summer (U.S. EPA, 2009a). For example, Franklin *et al.* (2008) analyzed $PM_{2.5}$ monitoring and daily mortality data in 25 cities nationwide between years 2000 to 2005, and reported a 80% higher effect estimate in the east (0.92% increase in non-accidental deaths associated with a 10 $\mu g/m^3$ increase in 2-day averaged $PM_{2.5}$ concentration) than in the west (0.51%). Spring showed the highest effect of 1.88% among all seasons, which was a factor of 11.5 higher than that of the lowest winter season (0.19%). An inverted U-shape curve was identified to illustrate the relationship between $PM_{2.5}$ health effect estimates and seasonally-averaged ambient temperature (Franklin *et al.*, 2008). Temperature was used as a surrogate for building ventilation, which varies by season and affects particle penetration to indoor microenvironments. When temperatures were moderate between 10 to 25 ℃, the estimated effects of non-accidental mortality were above the average effect, while effects were below average for both low and high temperatures.

Bell *et al.* (2008) further quantified the short-term effects of $PM_{2.5}$ on cardiovascular or respiratory hospitalizations among the elderly by geographic region and season in the US, and found the strongest evidence was in the Northeast and in winter for both causes. For lag-0 cardiovascular admissions, the largest and the lowest estimated effects among geographic regions for a given season such as winter varied by 1.6 times from 2.01% in the Northeast to 0.76% in the Southwest. In the same region such as for the Northeast, the effect in the winter

season (2.01%) was about 2.7 times higher than for the summer (0.55%). Heterogeneity in $PM_{2.5}$ health effects may result from seasonal and regional differences in emissions and particle chemical constituents, and differences in exposure factors other than concentration, including differences in home ventilation, indoor versus outdoor activity patterns, and population demographics (Bell *et al.*, 2008).

While Franklin *et al.* (2008) and Bell *et al.* (2008) used measured ambient PM concentration data for risk estimates, Chang *et al.* (2012) estimated the short-term effects of personal exposure to ambient $PM_{2.5}$ in New York City for years 2001 to 2005 using two exposure metrics, including the fused Community Multiscale Air Quality model (CMAQ) data and the estimated exposure from an exposure simulator SHEDS-PM. Risk estimates associated with exposures were greater than that associated with concentrations, which indicated a negative bias in the concentration-response function when ambient levels were used as a proxy for exposure. The magnitude of the bias approximated the ratio between daily concentration and exposure level. When stratified by season, the risks of mortality associated with $PM_{2.5}$ were higher and only statistically significant during the summer months. Such result agreed with previous seasonal analysis of PM_{10} and mortality in Northeastern US (Peng *et al.*, 2005). This highlights the need to characterize geographic and seasonal variability in the E_a/C ratio on a population basis to aid in the interpretation of epidemiologic findings.

There have been studies looking at the E_a/C ratio in exposure for pollutants such as ozone, CO and SO_2, by location and season (U. S. EPA, 2008; U. S. EPA, 2013; U. S. EPA, 2010b). For ozone, the population average E_a/C ratios are typically 0.1 to 0.3. Higher E_a/C

ratios for ozone are generally observed with increasing time spent outside and higher air exchange rate. For CO, the average E_a/C ratios are around 1. For SO_2, as a result of low ambient SO_2 concentrations and the limitations of passive sampling, only two studies have reported average E_a/C ratios, with values ranging from 0.08 to 0.13. Differences in the E_a/C ratio between pollutants are mainly related to pollutant-specific physical or chemical removal processes. E_a/C ratios for ozone tend to be low because ozone is a highly reactive oxidant that is removed from air by surface interactions and airborne constituents (U.S. EPA, 2013). In contrast, CO is relatively inert and has a much longer lifetime in air (U.S. EPA, 2010b). SO_2 is highly soluble and thus can be removed by reactions on indoor surfaces, especially those that are moist (U.S. EPA, 2008).

None of these characteristics applies to $PM_{2.5}$. $PM_{2.5}$ is not as chemically reactive as ozone, as soluble as SO_2, nor removed as slowly as CO. Thus, the E_a/C ratio for $PM_{2.5}$ tends to be substantially different than the E_a/C ratios for these other pollutants. The main removal process for $PM_{2.5}$ is deposition. Also, unlike the gaseous pollutants, the penetration efficiency of $PM_{2.5}$ from outdoors to indoor microenvironments is less than one, because of physical processes such as impaction and interception (U.S. EPA, 2009a).

Due to the limitation of field studies, there is a need for research to develop modeling approaches for exposure assessment. EPA has used the Air Pollution Exposure (APEX) model for estimating human population exposure to ozone and CO, and produced substantial information and data to support model development (U.S. EPA, 2013; U.S. EPA, 2010b). Although EPA recommended using APEX and/or the Stochastic Human Exposure and Dose Simulation model for Particulate Matter (SHEDS-PM) for PM population exposure

assessment (Glen *et al.*, 2012; Burke and Vedamtham, 2009), there has not yet been a systematic modeling analysis to quantify the E_a/C ratio for $PM_{2.5}$ by region and season. A goal of the study is to provide an example of the application of simulation-based exposure modeling to quantification of variability in $PM_{2.5}$ exposure, especially the E_a/C ratio.

Another important issue in PM epidemiological studies is to determine whether the C-R relationship is linear across the full concentration range or if there are concentration ranges that exhibit nonlinearity. Evidence suggests that the C-R relationship for $PM_{2.5}$ is consistent with a linear no-threshold model at low levels (annual average concentrations from 5 to 30 $\mu g/m^3$), but flattens out to be log-linear at higher long-term average concentrations on the order of 100 $\mu g/m^3$ or more, such as for an active tobacco smoker or a person exposed to indoor solid-fuel combustion (Pope *et al.*, 2009; Ezzati and Kammen, 2001). Therefore, it is important to accurately quantify non-ambient exposure from indoor emissions and assess the incremental health effect of $PM_{2.5}$ from non-ambient exposure relative to a baseline of the relatively lower ambient exposure.

Individual exposures to $PM_{2.5}$ occur both outdoors and indoors. Although NAAQS regulates particles in outdoor air, not indoors, it is still important to consider indoor air. Decisions on the level at which to set the standards will depend partly on considerations of the impact of outdoor air on indoor air and human exposure. Indoor $PM_{2.5}$ concentrations are affected by the penetration of ambient $PM_{2.5}$, which leads to ambient exposure that takes place indoors; and non-ambient sources such as cooking and smoking, which leads to non-ambient exposure that can be much higher than ambient exposure (Wilson *et al.*, 2000). Since people spend at least half of their time per day in their residence (Stallings *et al.*,

2002), indoor residential $PM_{2.5}$ exposure has a substantial influence on total $PM_{2.5}$ exposure. Based on a SHEDS-PM case study in Philadelphia, indoor residential exposure of ambient and non-ambient origins accounted for 28% and 42%, respectively, on total $PM_{2.5}$ exposure of the population (Burke *et al.*, 2001). However, ambient and non-ambient particles differ in sources and sizes, and likely to differ in composition and biologic properties as well (Abt *et al.*, 2000; Long *et al.*, 2001). Thus, characterizing the potential health impacts associated with ambient and non-ambient particles within the indoor residence, separately, may help to reduce uncertainty in health risk estimates.

The current practice in stochastic population exposure models such as the Air Pollution Exposure (APEX) and Stochastic Human Exposure and Dose Simulation (SHEDS) typically assumes the home residence to be a single, well-mixed zone when calculating residential exposure. However, indoor emissions from cooking or smoking typically occur in a specific room, and the indoor mixing of $PM_{2.5}$ from indoor emissions is mostly limited to that room initially. Therefore, the bias in non-ambient exposure concentration associated with the assumption of one large single zone within a home should be evaluated. In addition, there is a need to know how parameters such as air exchange rate, volume and type of the house, and emission rate and duration of non-ambient sources affect inter-individual variability in the distribution of indoor $PM_{2.5}$ concentrations. Better quantification of dispersion of pollution within the indoor residential microenvironment is critical for $PM_{2.5}$ exposure assessment.

Studies of personal exposure to ambient $PM_{2.5}$ generally found that the largest percentage of time in which an individual is exposed to $PM_{2.5}$ of ambient origin occurs

indoors but the highest ambient $PM_{2.5}$ exposure concentrations occur in on-road or near-road transportation microenvironments (U.S. EPA, 2009a). Typical personal transport in the U.S. include modes such as personal car, referred to as "in-vehicle," transit bus and pedestrian walking, which comprise more than 90% of total trips (U.S. DOT, 2001). In the U.S., the average one-way daily commuting travel time is 25.5 minutes, and 86% of trips to work are via personal vehicle (McKenzie and Rapino, 2011). Depending on traffic flow, meteorological conditions, vehicle emission rates, whether windows are open, operation of the vehicle heating, ventilation, and air conditioning (HVAC) system, and time spent in-vehicle, in-vehicle exposure may account for 10 to 20 percent of total daily average $PM_{2.5}$ exposure (Liu and Frey, 2011). The mode share of public transportation and pedestrian walking are comparably higher in large metro areas or areas with at least one large college or university that had high proportions of college-aged students (McKenzie and Rapino, 2011). A closer examination of PM concentrations on or near roadway is essential to accurately estimating individual exposure, and to identifying key factors affecting variability in exposures that may aid in developing improved exposure model estimates and effective control strategies.

1.2 Objectives

The objectives of this study are to:

(1) Assess inter-individual, geographic and seasonal variability in population $PM_{2.5}$ exposures;

(2) Improve $PM_{2.5}$ exposure concentration estimates for the indoor residential microenvironment; and

(3) Evaluate variability in $PM_{2.5}$ exposure concentrations for the transportation microenvironments.

1.3 Organization

This dissertation consists of six parts. As an overview graph, Figure I-2 illustrates how parts of the dissertation are related with each research objective. Appendices are given at the end of the document. Each part of the dissertation contains a separate reference list.

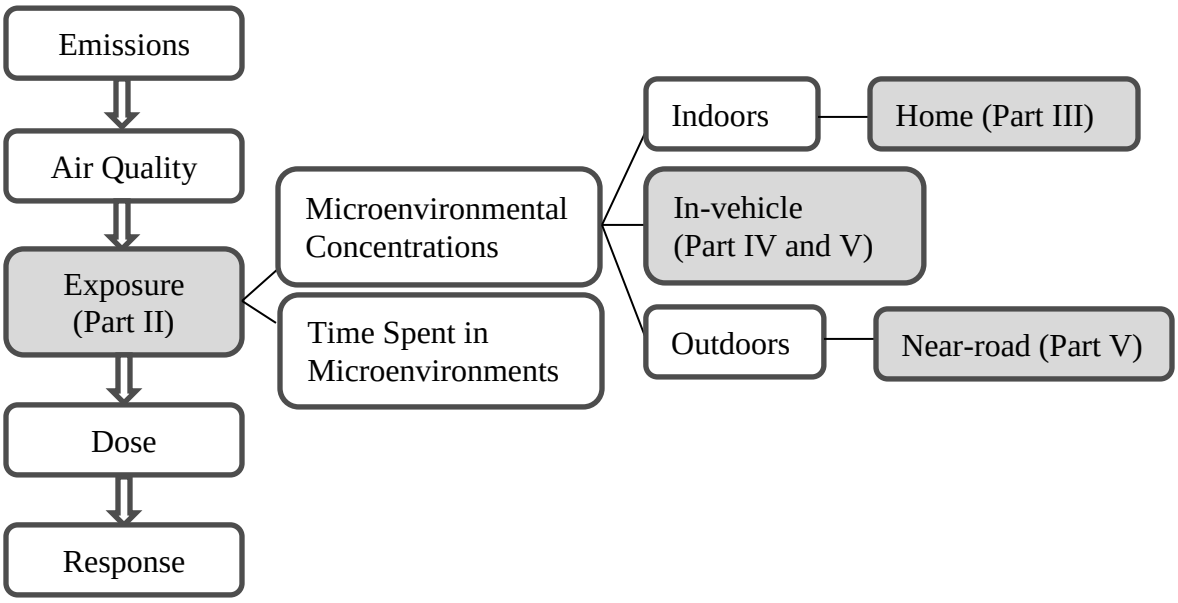

Figure I-2. Overview of Research Scope

Part I introduces the background information regarding $PM_{2.5}$ exposure, as well as general exposure concepts, research motivation and objectives, and dissertation organization.

Part II assesses inter-individual, geographic, and seasonal variability in estimated population exposure to $PM_{2.5}$ using an exposure simulation model.

Part III develops a method to estimate correction factors for bias correcting the single-zone estimates of indoor residential $PM_{2.5}$ concentrations to account for indoor emission sources such as cooking and smoking.

Part IV demonstrates a method for quantifying the ratio of in-vehicle to near-vehicle exposure concentrations of airborne $PM_{2.5}$.

Part V compares $PM_{2.5}$ concentrations in selected transportation modes including car, bus and walking, and identifies factors affecting in-transit exposure.

Part VI discusses and summarizes main conclusions of this research.

Three appendices are provided. Appendix A gives supporting information for Part II. Appendix B gives supporting information for Part III. Appendix C gives supporting information for Part IV. Appendix D compares the predicted exposures with ambient $PM_{2.5}$ concentrations across years.

PART II ASSESSMENT OF INTER-INDIVIDUAL, GEOGRAPHIC, AND SEASONAL VARIABILITY IN ESTIMATED HUMAN EXPOSURE TO FINE PARTICLES*

* This manuscript was published in *Environmental Science & Technology*, 2012, 46 (22): 12519-12526.

Abstract

Health effects associated with ambient fine particle ($PM_{2.5}$) exposure are typically estimated based on concentration-response (C-R) functions using area-wide concentration as an exposure surrogate. Persons 65 and older are particularly susceptible to adverse effects from $PM_{2.5}$ exposure. Using a stochastic microenvironmental simulation model, distributions of daily $PM_{2.5}$ exposures were estimated based on ambient concentration, air exchange rate, penetration factor, deposition rate, indoor emission sources, census data, and activity diary data, and compared for selected regions and seasons. Even though the selected subpopulation spends an average of over 20 hours per day indoors, the ratio of daily average estimated exposure to ambient concentration (E_a/C) is approximately 0.5. The daily average E_a/C ratio varies by a factor of 4 to 5 over a 95% frequency range among individuals, primarily from variability in air exchange rates. The mean E_a/C varies by 6 to 36% among selected NC, TX and NYC domains, and 15 to 34% among four seasons, as a result of regional differences in housing stock and seasonal differences in air exchange rates. Variability in E_a/C is a key factor that may help explain heterogeneity in C-R functions across cities and seasons. Priorities for improving exposure estimates are discussed.

2.1 Introduction

According to the U.S. Environmental Protection Agency (EPA) Integrated Science Assessment (ISA) for Particulate Matter, a causal or likely to be causal relationship exists between short-term (daily) human exposure to fine particulate matter ($PM_{2.5}$) and several health effects, such as mortality, cardiovascular and respiratory morbidity (U.S. EPA, 2009a). Children and adults 65 years and older are the two most susceptible subpopulations (U.S. EPA, 2009a). The proportion of older adults in the US population will increase from 13% in 2011 to 20% in 2030 (U.S. Census Bureau, 2000). Thus, PM-related health incidents could occur more frequently in the future.

$PM_{2.5}$ ambient concentration is frequently used in epidemiological studies as a surrogate for personal exposure to ambient $PM_{2.5}$ (U.S. EPA, 2009a). However, actual exposure depends on the amount of time an individual spends in different microenvironments, which are surroundings that can be treated as homogeneous or well characterized in the concentrations of an agent (U.S. EPA, 1992). Microenvironments include various indoor locations (e.g. home, work, school, restaurants, stores), outdoors, in transit, and others (Wallace *et al.*, 2006). On average, people spend more than 80 percent of daily time indoors and two thirds of daily time at home (Klepeis *et al.*, 2001). For indoor microenvironments, a portion of ambient $PM_{2.5}$ penetrates indoors, and some deposits to interior surfaces (Wallace *et al.*, 2006). For some cases, the individual indoor $PM_{2.5}$ concentrations could be higher than ambient because of particle re-suspension resulting from human activity (U.S. EPA, 2007). However, SHEDS-PM does not model re-suspension. On average for a population, the daily exposure to particles of ambient origin is typically less

than the ambient concentration, and the difference contributes to exposure error (Carroll *et al.*, 1995; Zeger *et al.*, 2000). Using ambient $PM_{2.5}$ concentrations as a surrogate for the community average personal exposure to ambient $PM_{2.5}$ will bias the estimation of health risk coefficients by the ratio of ambient $PM_{2.5}$ exposure to ambient $PM_{2.5}$ concentration (E_a/C) (U.S. EPA, 2009a).

Findings from $PM_{2.5}$ exposure panel studies indicate substantial variability in 24-h average personal $PM_{2.5}$ exposures among US regions (Wallace and Williams, 2005; Williams *et al.*, 2003; Weisel *et al.*, 2005; Sarnat *et al.*, 2005). Individuals are exposed to $PM_{2.5}$ of both ambient and nonambient origin, and both sources of $PM_{2.5}$ may contribute to adverse health outcomes. Total and nonambient PM exposure are poorly correlated with ambient PM concentration (Wilson *et al.*, 2000). Ambient PM concentrations are affected by meteorology and by changes in emission rates and locations of emission sources, whereas nonambient PM concentrations are influenced by daily activities of people. The E_a/C ratio depends on housing type and activity patterns, air exchange rate, and PM deposition rate. It varies between 0 and 1 among individuals, and varies among cities and seasons (Sarnat *et al.*, 2007). Since concentration-response functions are typically heterogeneous across cities and seasons, characterizing the factors that influence E_a/C could be useful in correcting for exposure error.

Inter-individual variability in exposure may also be influenced by the sampling time interval. Many time-series studies examine the associations of short-term health outcomes based on a 24-hr interval (U.S. EPA, 2010a). There is some evidence, based on analysis of PM_{10} data, that mortality or morbidity on a given day is influenced by prior PM exposures up

to more than a month before the date of death (Schwartz, 2000). Thus, averaging times other than 24-hr averages should be considered to provide appropriate modeling results for sub-acute or chronic health effects applications (Özkaynak *et al.*, 2009). Therefore, quantification of the sensitivity of inter-individual variability in exposure with respect to different averaging times may help to better inform health effects studies.

$PM_{2.5}$ exposure can be estimated based on field study or simulation models. Measured exposure data mainly come from either a few multi-city probability-based field studies, such as the Relationship between Indoor, Outdoor and Personal Air (RIOPA) study, the Detroit Exposure Aerosol Research Study (DEARS), and the Canadian Windsor Ontario Exposure Assessment Study (WOEAS), or longitudinal panel studies focused on small numbers of exposed individuals in susceptible subpopulations, such as the elderly (Weisel *et al.*, 2005; Williams *et al.*, 2009; Wheeler *et al.*, 2011; Williams *et al.*, 2000).

Field studies are time-consuming and expensive to conduct, which limits their sample sizes. As a result, measurements may only represent participants in that study and may not necessarily be generalizable. In comparison, a scenario-based exposure model can be an alternative tool for quantifying human exposure on a population basis (Burke *et al.*, 2001). For example, the U.S. Environmental Protection Agency has developed the Stochastic Human Exposure and Dose Simulation model for Particulate Matter (SHEDS-PM). SHEDS-PM is used here for simulating exposure because it has undergone several evaluation and validation studies (Burke *et al.*, 2002; Cao and Frey, 2011; Liu *et al.*, 2010; Cao and Frey, 2011).

As summarized by Sarnat *et al.* (2007), several measurement studies have been conducted to estimate the associations between $PM_{2.5}$ personal ambient exposure and ambient concentration, which can be used to estimate the E_a/C ratio. Although there have been studies that quantified geographic and seasonal variability in the E_a/C ratio for other pollutants, such as ozone, CO, and SO_2, there has not been systematic quantification of variation in for $PM_{2.5}$ using a consistent framework (U.S. EPA, 2012; U.S. EPA, 2010b; U.S. EPA, 2008). E_a/C ratios differ substantially among pollutants depending on chemical reactivity, solubility, and pollutant lifetime. The average E_a/C ratios for highly reactive ozone are typically 0.1 to 0.3, for relatively unreactive CO are around 1, and for highly soluble SO_2 are from 0.08 to 0.13. In contrast, the E_a/C ratio for $PM_{2.5}$ depends on factors such as interception and impaction that affect penetration from outdoors to indoors, and indoor deposition rate (U.S. EPA, 2009a).

The objectives of this paper are to: (1) assess the sensitivity of inter-individual variability in exposures with respect to averaging time; (2) evaluate geographic differences in inter-individual variability in exposures; and (3) evaluate seasonal differences in inter-individual variability in exposures.

2.2 Methodology

The methodology includes: (1) scenario-based exposure modeling; (2) study design and identification of data sources for the case study; and (3) analysis of exposure model output.

2.2.1 Scenario-based Exposure Modeling

Burke *et al.* (2001) evaluated SHEDS-PM using measurement data from a PM panel study conducted in the Research Triangle Park, NC area. The model predictions of individual and population exposures to $PM_{2.5}$ are generally consistent with the estimates derived from the personal measurement data. Other studies have reviewed the algorithms and input data for SHEDS-PM, and recommended improvements are incorporated here. Cao and Frey reviewed default Environmental Tobacco Smoke (ETS) related inputs and recommended updates to input data for the proportion of smokers and other smokers, and for cigarette emission rates (Cao and Frey, 2011a). Liu *et al.* reviewed the algorithms and default inputs for the in-vehicle microenvironment, and proposed an alternative approach which integrates a dispersion model and a mass balance approach (Liu *et al.*, 2010). Cao and Frey recommended updates to the distributions of ACH, P, and k for selected areas and seasons (Cao and Frey, 2011b).

Estimates of inter-individual variability in outdoor and indoor microenvironmental $PM_{2.5}$ exposures for a simulated population are based on user-supplied ambient air quality data, model-incorporated census data, and human activity data for the selected geographic domain (Burke and Vedamtham, 2009). Individuals are randomly generated from the year 2000 US Census to represent the population in a selected area. For each simulated individual, the amount of time spent in each microenvironment is derived from the Consolidated Human Activity Database (CHAD) (McCurdy *et al.*, 2000). Algorithms are applied to estimate $PM_{2.5}$ concentrations in each microenvironment based on ambient concentration and indoor

sources. Simulated microenvironments include outdoors, home, office, school, store, restaurant, bar and vehicle.

For the residential microenvironment, SHEDS-PM utilizes a single-compartment, steady state mass balance equation to calculate residential $PM_{2.5}$ concentration (Burke and Vedamtham, 2009). The contribution from indoor emission sources such as smoking, cooking, cleaning and other sources is quantified. The housing type categories in SHEDS-PM are single-family detached, single-family attached, multiple family, mobile home and other. Based on the US Census 2000 Housing Survey, lognormal distributions are used for each housing type in SHEDS-PM to represent inter-home variability in indoor volume. The key mass balance inputs include ACH, P and k. ACH is the volume flow of air within the indoor microenvironment divided by the interior volume. It 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 (Özkaynak *et al.*, 1996). Mechanical ventilation would lower indoor ambient PM exposure because a substantial portion of indoor air is recirculated and, consequently, ACH is smaller. Distributions of ACH by area and season reflect the variability in ACH caused by different ventilation practices used and housing types/volumes of each individual. P is the portion of particles that pass through the building from outdoors. Deposition rate, k, is the settling of airborne particles onto indoor surfaces (Weisel *et al.*, 2005).

2.2.2 Study Design

The focus here is to quantify possible regional and seasonal variability in estimated exposure. Adults over 65 years old are selected because they are a susceptible subpopulation with respect to $PM_{2.5}$ exposure (U.S. EPA, 2009a). To address regional differences, three urban areas are chosen to represent diverse southeast, south central and northeast US climate zones. These areas include: (1) a six-county area in North Carolina along Interstate 40, comprised of Wake, Durham, Orange, Alamance, Guilford, and Forsyth Counties, that includes the cities of Raleigh, Durham, Burlington, Greensboro, High Point, and Winston-Salem; (2) Harris County in Texas, including the city of Houston; and (3) New York City, including Bronx, New York, Kings, Queens, and Richmond Counties. Approximately 50,000 individuals 65 years and older are simulated from all census tracts of each area for a period of 30 days in each season. To address seasonal differences, one month from each of four seasons is selected, including April for spring; July for summer; October for fall; and December for winter. SHEDS-PM model version 3.7 was used to run all cases. Typically it takes approximately 5 to 6 hours of CPU time to simulate exposures for 50,000 individuals for a 30-day time period.

Daily average $PM_{2.5}$ air quality data are obtained from U.S. EPA based on predictions of 2002 average concentrations for 12 km by 12 km grid cells from the Community Multiscale Air Quality (CMAQ) model, which were updated with available monitoring data using Bayesian statistical inference (McMillan *et al.*, 2010). Demographic distributions by age, gender and housing type are sampled from year 2000 US Census data. CHAD diary data are matched to each simulated individual based on gender, age, day type, and smoking status.

ETS is modeled in residential, restaurant, and bar microenvironments. The proportions of smokers and non-smokers are estimated based on region-specific 2002 data (State Center for Health Statistics, 2002; Marshall *et al.*, 2006; U.S. DHHS, 2006; NYC Department of Health and Mental Hygiene, 2002). Input parameters for residential microenvironments, such as ACH, P, k, and cigarette emission rate, and for the in-vehicle microenvironment, are specified based on related literature (Cao and Frey, 2011a; Liu *et al.*, 2010; Cao and Frey, 2011b; Özkaynak *et al.*, 1996; Murray and Burmaster, 1995; Koontz and Rector, 1995). Parameters of other microenvironments are based on Burke *et al.* (2001).

2.2.3 Statistical Analysis Methods

SHEDS-PM outputs are processed and analyzed using Predictive Analytics SoftWare (PASW) 18.0. The SHEDS-PM model quantifies time spent by each simulated individual in different microenvironments and the corresponding microenvironmental $PM_{2.5}$ concentrations, and estimates daily average E_a, E_{na}, and E_t for each simulated individual. The E_a/C ratio is calculated for each simulated individual for each simulated day.

Variability in exposure among individuals on a given day is compared to variability for a single individual over time to assess the relative importance of inter- versus intra-individual variability. The variability among individuals and over time is quantified based on coefficients of variation (CV). The NC domain spring case is selected as an example. The reported CV of inter-individual variability is an average of daily CVs of inter-individual variability over simulated days. The reported CV of intra-individual variability is an average of individual CVs of intra-individual variability in exposure over simulated individuals.

To examine the sensitivity of inter-individual variability with respect to averaging time, the daily E_a/C ratio is further averaged by individual over one month to represent a longer term. Both daily and monthly ratios of E_a/C are compared for each geographic area and season. The analysis included comparisons of 95% frequency ranges inferred from simulated cumulative distribution functions (CDFs) of inter-individual variability in exposure, and comparisons of the effect of longitudinal versus randomized day-to-day sampling of activity dairies on the distribution of monthly average exposures. Individual daily exposure values are extracted from SHEDS-PM output to construct a 50,000 individual by 30 day exposure matrix. For each day, correlations of inter-individual variability in exposure are calculated with each other day. Days are categorized by day type, including weekday, Saturday and Sunday. For each of the 30 days, the average correlation with all other days of the same type, and with all days of each of the other two types, are estimated.

To assess the geographic and seasonal differences in inter-individual variability in exposures, daily ratios of E_a/C, E_{na} and E_t are compared among geographic areas for the same season and among multiple seasons for the same area.

2.3 Results

2.3.1 Key Inputs

This section assesses SHEDS-PM inputs that affect inter-individual variability in exposure. The average daily ambient $PM_{2.5}$ concentrations were highest in July compared to the other months for all regions.

Values of ACH, P, and k for the residential microenvironment are shown in Table II-1. Distributions of variability in ACH by geographic region are based on the Relationship of Indoor, Outdoor and Personal Air (RIOPA) study (Weisel *et al.*, 2005) from 1999 to 2001, Murray and Burmaster (1995), Koontz and Rector (1995), and Wallace *et al.* (2006). Distributions of variability in P and k are based on the PTEAM and RIOPA studies (Özkaynak *et al.*, 1996; Weisel *et al.*, 2005). The distribution types are selected based on Cao and Frey (2011b) and are explained in the Appendix A. Sensitivity analysis by Cao and Frey (2011b) assessed the effects of ACH, P and k on estimated human exposures. ACH is the most sensitive input for both ambient and non-ambient exposure to $PM_{2.5}$, whereas the results are not sensitive to the choice of distribution for P and k.

Table II-2 shows other input factors that affect inter-individual exposure variability, such as demographics, smoking prevalence, housing types, and human activity patterns. Females account for approximately 60 percent of the total elderly population in each area. Exposure to indoor sources of $PM_{2.5}$ is typically higher as the housing interior volume decreases (Cao and Frey, 2011b). The population weighted average time spent in different microenvironments differs by gender.

Table II-1. Residential Microenvironment Input Parameters

Parameter	Distribution Type [a]	Location [b]	Season	Value [c]
Penetration (P)	Triangular	ALL	ALL	Min= 0.70, Mode= 0.78, Max= 1.0
Deposition (k)	Normal	ALL	ALL	$\mu = 0.40\ h^{-1}$, $\sigma = 0.01\ h^{-1}$
Air Exchange Rate (ACH)	Lognormal	NC domain	Winter	$\mu_g = 0.38\ h^{-1}$, $\sigma_g = 1.80\ h^{-1}$
			Spring	$\mu_g = 0.31\ h^{-1}$, $\sigma_g = 2.31\ h^{-1}$
			Summer	$\mu_g = 0.54\ h^{-1}$, $\sigma_g = 1.70\ h^{-1}$
			Fall	$\mu_g = 0.49\ h^{-1}$, $\sigma_g = 1.62\ h^{-1}$
		Harris County, TX	Winter	$\mu_g = 0.56\ h^{-1}$, $\sigma_g = 2.20\ h^{-1}$
			Spring	$\mu_g = 0.38\ h^{-1}$, $\sigma_g = 1.80\ h^{-1}$
			Summer	$\mu_g = 0.37\ h^{-1}$, $\sigma_g = 1.90\ h^{-1}$
			Fall	$\mu_g = 0.65\ h^{-1}$, $\sigma_g = 1.80\ h^{-1}$
		NYC	Winter	$\mu_g = 0.45\ h^{-1}$, $\sigma_g = 2.03\ h^{-1}$
			Spring	$\mu_g = 0.40\ h^{-1}$, $\sigma_g = 1.82\ h^{-1}$
			Summer	$\mu_g = 0.64\ h^{-1}$, $\sigma_g = 2.09\ h^{-1}$
			Fall	$\mu_g = 0.22\ h^{-1}$, $\sigma_g = 1.72\ h^{-1}$

[a.] Triangular distribution parameters are the minimum, mode, and maximum; normal distribution parameters are the mean μ and standard deviation σ; lognormal distribution parameters are the geometric mean μ_g and standard deviation σ_g. The selection of distribution types are based on Cao and Frey (2011) and is described in the Appendix A.

[b.] NC includes Wake, Durham, Orange, Alamance, Guilford, and Forsyth Counties; TX includes Harris County; NYC includes Bronx, New York, Kings, Queens, and Richmond Counties.

[c.] Sources: Cao and Frey (2011); P, k: Weisel *et al.* (2005), Özkaynak *et al.* (1996); ACH: NC- Murray and Burmaster (1995), Wallace *et al.* (2006); TX- Murray and Burmaster (1995), Weisel *et al.* (2005) ; NYC- Murray and Burmaster (1995), Koontz and rector (1995).

Table II-2. Exposure Model Demographic Input Data Regarding Population Distribution, Smoking Prevalence, Housing Types, and Activity Patterns

Distribution of Population by Age and Gender [a] (%)						
Age Group	Six-County Region, NC		Harris County, TX		New York City	
	Male	Female	Male	Female	Male	Female
65-69	12.8	15.5	14.3	17.0	12.0	15.7
70-74	10.9	14.9	11.5	15.0	10.2	14.9
75-79	8.2	12.7	8.3	12.1	7.8	12.8
80-84	4.5	8.9	4.2	7.6	4.7	9.0
85-89	2.1	5.6	2.0	4.7	2.5	5.8
90-94	0.6	2.5	0.6	2.0	0.9	2.6
≥95	0.2	0.8	0.2	0.6	0.3	0.9
Total	39.2	60.8	41.4	58.9	38.4	61.6

Distribution of Smoking Prevalence in Older Adults by Gender [b] (%)						
	Six County Region, NC		Harris County, TX		New York City	
Age Group	Male	Female	Male	Female	Male	Female
≥ 65	18.5	10.5	12.0	4.6	9.3	10.4

Distribution of Housing Types [c] (%)			
	Six County Region, NC	Harris County, TX	New York City
Single Family Detached	58.8	62.2	13.2
Single Family Attached	4.6	4.6	9.9
Multiple Family	31.3	29.6	76.8
Mobile Home	5.3	3.5	0.1

Distribution of Older Adult Daily Activity Patterns by Gender [d] (hr/day)						
	Time Spent Outdoors		Time Spent Indoor		Time Spent in Vehicle	
Age	Male	Female	Male	Female	Male	Female
≥ 65	2.1	0.8	20.7	22.3	1.2	0.9

[a.] Source: U.S. Census 2000.

[b.] Sources: NC and TX – Behavioral Risk Factor Surveillance System (BRFSS), 2002; NYC – Epiquery, 2002.

[c.] Source: U.S. Census 2000 – Housing Survey. 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.

[d.] Source: Consolidated Human Activity Database (CHAD) (McCurdy *et al.*, 2005). Outdoor includes street, parking lot, gas station, park, playgrounds, pool, farm, and all other outdoor microenvironments; indoor includes home, office, school, store, bar, restaurant, and all other indoor microenvironments; in vehicle includes travel by car, truck, motorcycle, bus, train, subway, airplane, boat, walking, bicycle, and waiting for travel either indoor or outdoor.

2.3.2 Output

Inter-individual variability in estimated exposure to $PM_{2.5}$ is compared with respect to averaging time, area, and season.

Averaging Time

Inter- and intra-individual variability in exposure are compared based on the CV of inter- and intra-individual variability in daily exposure, respectively, as detailed in Table A-3 of the Appendix A. Inter-individual variability results in part from spatial variation in ambient concentration. However, inter-individual variability is also influenced by other factors such as ACH and activity. For the NC spring case, on average, CVs of inter- and intra-individual variability in daily average C are 0.11 and 0.32, respectively. Thus, C varies relatively little over space and among exposed individuals on a given day, and varies more substantially with time from day-to-day, which is consistent with other studies that are summarized in the PM ISA (U.S. EPA, 2009a). However, inter-individual variability in exposure is influenced by variability in other factors in addition to C. The CV of inter-individual variability in E_a is 0.37, which is much larger than that for C. The CV of intra-individual variability in E_a is 0.45, which is 18% higher than for inter-individual variability. Thus, exposure varies more over time than it does between individuals on a given day. The CV of E_a/C is similar (about 0.3) for both inter- and intra-individual variability, indicating that factors influencing E_a other than C, such as ACH, P, and k have similar variability among individuals or over time.

Inter-individual variability in estimated exposure varies with respect to averaging time. When comparing daily and monthly average distributions of inter-individual variability

in estimated E_a/C for each geographic area and season, they have the same mean as expected, but the standard deviation of the daily average E_a/C is approximately twice that of the monthly average. The 95% frequency interval for the daily average E_a/C ratio typically ranges from 0.2 to 0.9, whereas the range for the monthly average is approximately 0.3 to 0.7, as shown in Figure II-1 for the NC domain in spring. The variation in E_a/C is reduced as the averaging time increases, since day-to-day variations are averaged out for each individual. For the NC spring case, the correlations of inter-individual variability in daily E_a/C among days of the same day type are around 0.4, whereas there is little correlation among days of different day types.

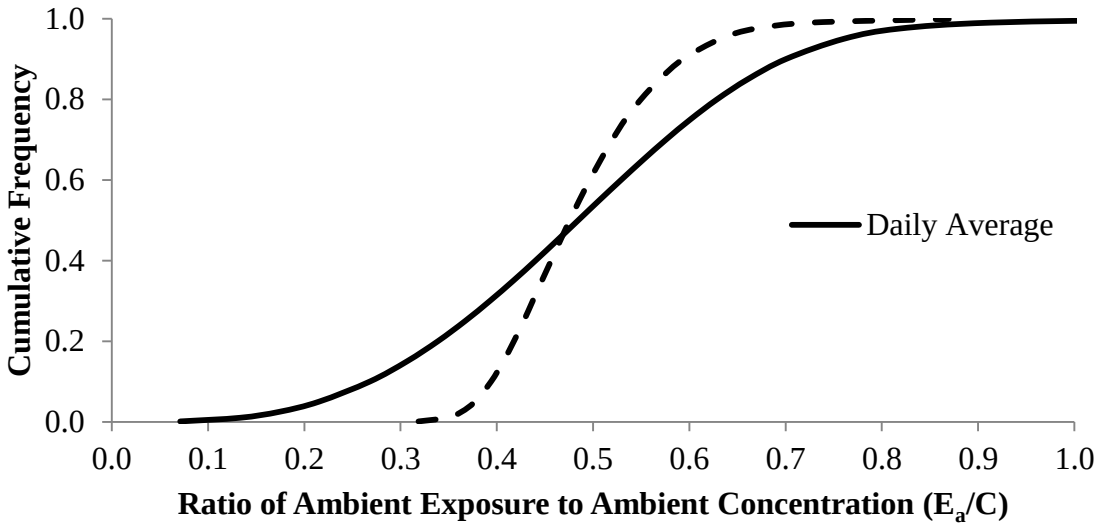

Figure II-1. Comparison of Inter-Individual Variability in the Ratio of Estimated Ambient Exposure to Ambient Concentration (E_a/C) for Selected Averaging Times, NC domain, Spring 2002

The range of the distribution of monthly average E_a/C is influenced in part by the method used in SHEDS-PM for longitudinal sampling of activity diaries. To explore how

sensitive the results are to this sampling method, an alternative NC spring case was analyzed in which individuals were randomly sorted from day-to-day, thus leading to no significant correlation in day-to-day comparisons of inter-individual variability. Because the longitudinal simulation is based on positive correlations from day-to-day, it is expected to provide a wider range of monthly average inter-individual variability than a randomized simulation. The difference in results between the two cases indicates whether longitudinal simulation has a significant effect. For the randomized case, the 95% frequency range in inter-individual variability of monthly average E_a/C is from 0.43 to 0.55, which is about one-third the range of the longitudinal case. Thus, the results are sensitive to the approach used for longitudinal simulation.

Geographic Variability

Inter-individual variability in exposure differs among climate zones. Figure II-2 presents the CDFs of the estimated daily average E_a/C for each area and season. Although the range is similar among areas considered, the mean values differ. As indicated in Table 3, the simulated elderly adults are exposed to approximately half of the modeled ambient $PM_{2.5}$ concentrations. The highest and the lowest mean E_a/C among geographic areas for a given season differ by 6% to 16% except in fall, which has a larger difference of 36%. The geographic differences in the mean E_a/C ratio are associated with differences in ACH. Lower ACH leads to less penetration of ambient $PM_{2.5}$, which results in lower indoor ambient exposure to $PM_{2.5}$. Therefore, areas with lower ACH typically have lower E_a/C ratios. For example, Harris County, TX has more air conditioning usage in the summer (U.S. EPA,

Table II-3. Geographic and Seasonal Variability in Exposure [a]

Area[b]	Season	E_a/C[c]		E_a[c] ($\mu g/m^3$)		E_{na}[c] ($\mu g/m^3$)		E_t[c] ($\mu g/m^3$)	
		Mean	StdDev	Mean	StdDev	Mean	StdDev	Mean	StdDev
NC domain	winter	0.52	0.15	6.2	3.7	12.9	29.3	19.1	29.4
	spring	0.49	0.17	6.2	3.0	13.9	32.2	20.1	32.1
	summer	0.57	0.13	11.7	6.3	11.0	24.9	22.6	25.5
	fall	0.56	0.14	7.6	4.5	11.5	26.0	19.1	26.3
Harris County, TX	winter	0.58	0.16	6.2	3.3	8.6	21.4	14.8	21.5
	spring	0.51	0.14	8.0	3.8	10.2	25.7	18.2	25.8
	summer	0.51	0.15	8.0	3.8	10.0	24.2	18.0	24.4
	fall	0.60	0.13	8.0	3.5	8.0	19.3	16.0	19.5
NYC	winter	0.54	0.15	7.1	3.9	13.0	31.2	20.1	31.2
	spring	0.52	0.15	6.2	4.3	13.7	31.8	20.0	32.0
	summer	0.59	0.15	12.2	8.5	11.2	27.3	23.4	28.2
	fall	0.44	0.17	4.6	3.3	17.7	41.5	22.3	41.6

[a.] Mean and standard deviation are based on individual daily average exposures obtained using the Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM).
[b.] NC includes Wake, Durham, Orange, Alamance, Guilford, and Forsyth Counties; TX includes Harris County; NYC includes Bronx, New York, Kings, Queens, and Richmond Counties.
[c.] E_a/C: ratio of ambient exposure to ambient concentration; E_a: ambient exposure ($\mu g/m^3$); E_{na}: non-ambient exposure ($\mu g/m^3$); E_t: total exposure ($\mu g/m^3$).

2012), which leads to lower air exchange and consequently lower E_a/C ratio than NC and NYC.

The average E_{na} for the simulated population differs among areas, as shown in Table II-3. The differences in the mean values among regions for a given season vary from 11% to 55% for the total population. The major factors that contribute to the geographic variability in estimated E_{na} are smoking prevalence, indoor volume, and ACH. A higher proportion of smokers leads to higher non-ambient $PM_{2.5}$ emissions. Smaller indoor volume leads to less dilution of indoor $PM_{2.5}$ sources. Lower ACH results in longer retention of indoor emissions.

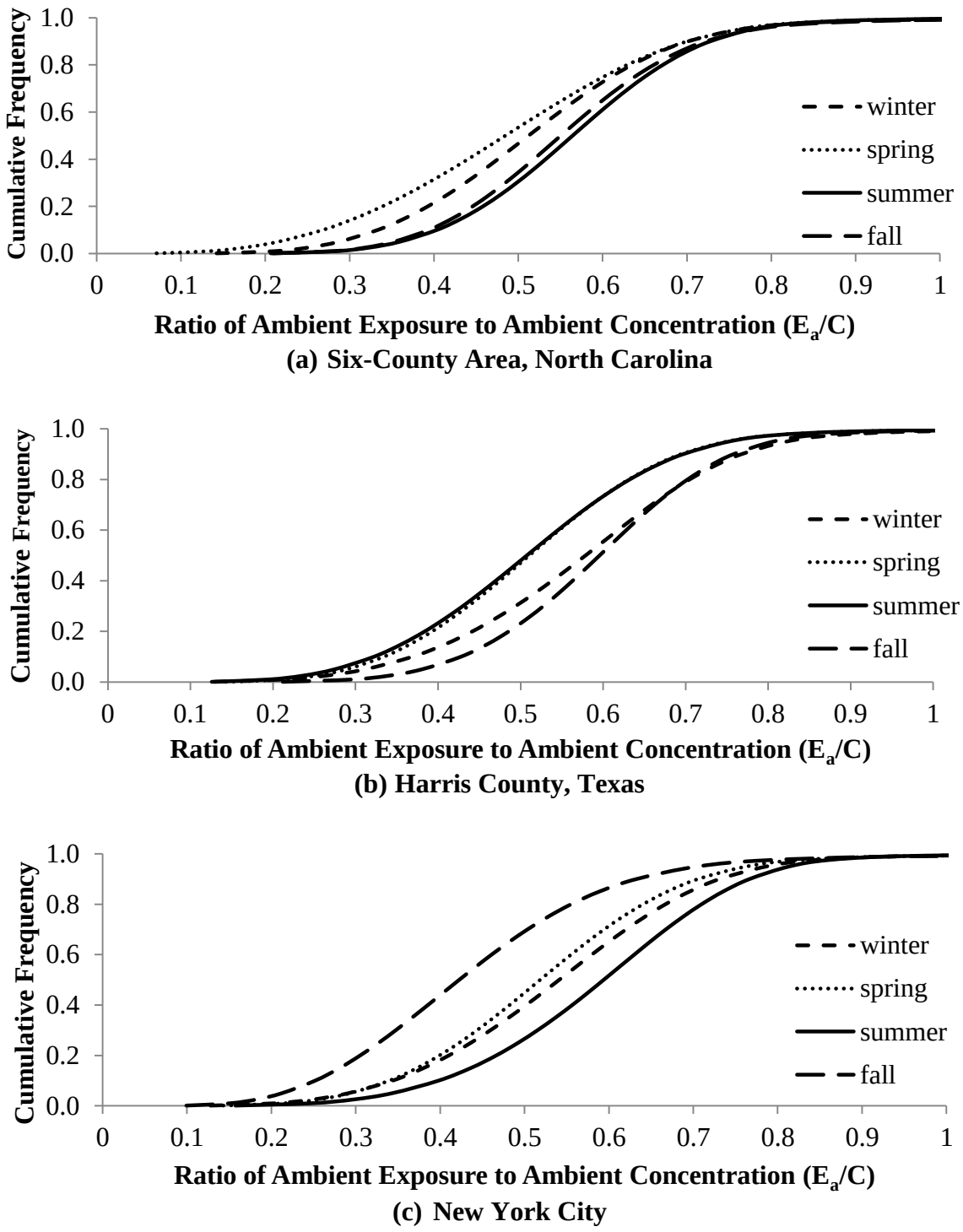

Figure II-2. Geographic and Seasonal Variability in the Ratio of Estimated Daily Ambient Exposure to Ambient Concentration for the NC domain, Harris County, and NYC, 2002

Generally, E_{na} is most sensitive to ACH. Areas with lower ACH tend to have higher average E_{na}. However, the combined effect of lower smoking prevalence and higher indoor housing volume sometimes can be more influential than ACH, as illustrated by the TX domain in summer. This domain has the smallest proportion of smokers and largest average indoor volume. Although the summer ACH is much less for the TX versus NC or NYC domains, the population average E_{na} for the TX domain is the lowest.

The daily average E_t varies by 10% to 28% among geographic areas for any given season for the total population. However, non-smokers not exposed to ETS have much lower average E_t than persons with ETS exposure. For people with ETS exposure, approximately 85% of daily E_t is non-ambient. Thus for those with ETS exposure, the average E_t is highly influenced by the non-ambient exposure level. This is because the upper-tail values of E_{na} (above the 90th percentile) are usually 2 to 10 times higher than for E_a.

Seasonal Variability

In the same region, the average daily E_a/C ratio differs among seasons by 16% in the NC domain and by 34% in NYC, as shown in Table II-3. Many dwellings in NYC are not air conditioned, and residents tend to open windows more in the summer. In contrast, in Harris County, the average E_a/C is 15% lower in summer than in the fall. For the NC domain, the E_a/C ratios are similar between summer and fall. The seasonal difference in the E_a/C ratio is partly associated with more widespread air conditioner use in TX and NC (U.S. EPA, 2012).

Mean values of E_{na} vary by 21% to 37% by region among seasons. Because several factors affecting the average estimated daily E_{na} are similar within a region, such as smoking

prevalence, indoor volume, demographics, and individual activity patterns, seasonal differences in E_{na} are most sensitive to seasonal differences in ACH.

The seasonal variability in estimated average daily E_t is not as pronounced compared to geographic variability. The difference in the average daily E_t among seasons for a given region ranges from 15% to 19%, which is mainly attributed to seasonal variations in ACH. For people exposed to ETS, because E_t is dominated by the contribution of non-ambient exposure, higher ACH leads to lower E_t. But for non-smokers, E_{na} only accounts for about 40% in their daily E_t on average. Therefore, higher ACH leads to higher E_t for non-smokers.

2.4 Discussion and Conclusions

As expected, the ambient air quality data in grid cells used as input to the SHEDS-PM model typically exhibited rather low spatial variation within a geographic domain on each day, with CV < 0.2 for 11 of the 12 area and season cases studied. Daily average ambient exposures are significantly lower than ambient concentrations and vary by season and location. Furthermore, there is substantial inter-individual variability in exposure that is not explained by ambient concentration alone.

A limitation not readily addressed by CHAD is the possible seasonal differences in activity patterns. Based on analysis of CHAD, as detailed in Table A-7 of the Appendix A, differences in the time spent indoors or time spent outdoors are found to be statistically significant depending on the gender, region, and season. However, these results are based on aggregation of CHAD data across large geographic regions that represent generic climate zones. There is insufficient data in CHAD from which to quantify differences in activity

pattern by gender, region, and season for the specific geographic areas that are the focus here. Clearly, there is a need to further develop CHAD to contain diaries representative of geographic areas and seasons of interest, and to refine the exposure model to take ambient conditions into account when sampling diaries.

The CV in time spent outdoors among individuals was found to be similar to the CV for 11 individuals for whom more than 4 days of diaries were available in CHAD. Thus, the limited evidence suggests that activity patterns are repeatable from day-to-day and similar among individuals, at least for the selected subpopulation. The simulation model appropriately accounts for similarity in day-to-day activity.

Non-ambient exposure to $PM_{2.5}$ is approximately uncorrelated with ambient concentration (average r_p = -0.002, as detailed in Table A-6 of the Appendix A), which is consistent with other studies that are summarized by EPA (U.S. EPA, 2009a). Average levels of E_{na} vary by area and season mainly because of differences in ACH. For people exposed to ETS, E_{na} is the dominant contributor to E_t.

The E_a/C ratio varies by individual, geographic area, season, and spatial-temporal averaging times. Therefore, quantification of the E_a/C ratio has implications for air pollutant exposure assessment, risk management, and epidemiologic studies. The daily E_a/C ratio differs by a factor of 4 to 5 over a 95% frequency range among individuals, indicating that some people are very highly exposed compared to others because of factors other than ambient concentration. The E_a/C ratio varies among individuals because of differences in activity patterns, housing characteristics, and seasons. The day-to-day variation of estimated individual daily average E_a is highly correlated ($r_p > 0.8$) with that of C. Even though biased,

the use of ambient concentration as a surrogate for ambient exposure in epidemiology studies may still account for temporal trends in exposure.

The distribution of E_a/C ratios in each area and season implies that, in general, exposure to ambient $PM_{2.5}$ is less than the ambient concentration. On average, exposures to simulated individuals are approximately half of the ambient concentrations. These findings indicate that concentration-response functions developed in epidemiological studies using ambient concentration as surrogate for exposure are biased when compared to exposure concentration.

Exposure, and not just concentration, should be considered in developing risk management strategies to reduce uncertainty in health effect estimates, and to identify highly exposed groups and possible exposure reduction strategies. High-end daily average ambient exposures among individuals are influenced by factors other than high ambient concentration, such as ACH by location and season. ACH is related to housing type and ventilation practices used. E_a/C is well correlated (r_p=0.5 to 0.6) with ACH, but has little correlation with ambient concentration C. Thus the distribution of inter-individual variability in the E_a/C ratio can be used to identify the need for providing advisory information to the public. Such information might include, for example, advice to reduce ventilation with outside air on high ambient $PM_{2.5}$ days.

The average E_a/C ratio illustrates the potential exposure error related to epidemiologic estimates of concentration-response (C-R) ratios. Regional or seasonal differences in the average E_a/C ratio may help to explain variations in concentration-response relationships between cities and seasons, because the estimated health effect parameter is a function of the

E_a/C ratio. The range in mean E_a/C among studied areas and seasons is from 0.44 to 0.60. The difference between these ratios is statistically significant based on the simulated results and represents a 36 percent relative difference. Because $E_a/C \geq 0$ and typically $E_a/C < 1$ for most individuals, the population mean of E_a/C is constrained and therefore the range of possible difference in average E_a/C ratio is also constrained. Region and season-specific E_a/C ratios are recommended as a factor to consider when interpreting heterogeneity in epidemiologic studies.

2.5 Acknowledgements

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PART III METHOD FOR BIAS CORRECTION OF ESTIMATED INDOOR RESIDENTIAL SINGLE-ZONE $PM_{2.5}$ CONCENTRATION TO ACCOUNT FOR INDOOR EMISSION SOURCES

Abstract

A positive relationship exists between fine particulate matter ($PM_{2.5}$) exposure and adverse health effects. Since people spend substantial daily time at home, residential indoor $PM_{2.5}$ exposure can have a large influence on total $PM_{2.5}$ exposure. The Stochastic Human Exposure and Dose Simulation model for Particulate Matter (SHEDS-PM) is a probabilistic tool for estimating population distributions of $PM_{2.5}$ exposure. SHEDS-PM assumes the home residence to be a single, well-mixed zone when calculating residential $PM_{2.5}$ concentration. However, indoor emissions from cooking or smoking typically occur in a specific room, and the indoor mixing of $PM_{2.5}$ from indoor emissions is mostly limited to that room initially. The bias in non-ambient exposure concentration associated with the assumption of one large single zone within a home is evaluated by applying an indoor air quality model, RISK, to compare indoor $PM_{2.5}$ concentrations for single-zone and multi-zone scenarios. Bias correction factors are estimated for cooking and smoking scenarios, separately, based on the ratio of multi-zone activity time-weighted average zonal concentrations divided by the single-zone average concentration. Correction factors are most sensitive to changes in air exchange rate whereas they are relatively insensitive to variations in source emission rate and duration. In a SHEDS-PM case study, the daily average total exposure increased by 17% after applying correction factors. Since emissions from cooking or smoking can contribute to high exposures for home residents, these results can assist in assessing the upper percentiles of population exposures and in informing effective management of indoor exposures.

3.1 Introduction

Epidemiologic studies have demonstrated a positive relationship between ambient fine particulate matter ($PM_{2.5}$) concentration and adverse health effects, such as cardiovascular and respiratory morbidity and mortality (U.S. EPA, 2009a). Individual exposures to $PM_{2.5}$ occur both outdoors and indoors. Indoor $PM_{2.5}$ concentrations are affected by the penetration of ambient $PM_{2.5}$, which leads to ambient exposure that takes place indoors; and non-ambient sources such as cooking and smoking, which leads to non-ambient exposure (Wilson *et al.*, 2000). Since people spend at least half of their time per day in their residence (Stallings *et al.*, 2002), indoor residential $PM_{2.5}$ exposure has a substantial influence on total $PM_{2.5}$ exposure. Therefore, quantification of dispersion of pollution within the indoor residential microenvironment is critical for $PM_{2.5}$ exposure assessment.

3.1.1 Scope

$PM_{2.5}$ exposure studies employ either direct measurement (personal exposure monitoring) or exposure models (Weisel et al, 2005; Wheeler *et al.*, 2011; Williams *et al.*, 2008). Population-based exposure monitoring studies require a large number of participants and significant resources. If sufficient input data exist, a scenario-based exposure model can be used to quantify human exposure on a population basis. Exposures for each simulated individual are estimated based on the time an individual spends in specific microenvironments. Microenvironments are locations for which $PM_{2.5}$ concentrations can be characterized, such as outdoor, home, school, in-vehicle, restaurant, bar, and others (U.S. EPA, 1992).

Various modeling tools are available to estimate human exposure to $PM_{2.5}$, including the Air Pollutant Exposure Model (APEX) (Glen *et al.*, 2012) and the Stochastic Human Exposure and Dose Simulation Model for Particulate Matter (SHEDS-PM) (Burke and Vedamtham, 2009). SHEDS-PM is selected because it has previously undergone evaluation and has been applied in recent assessment of $PM_{2.5}$ exposure (Burke *et al.*, 2001; Cao and Frey, 2011a; Cao and Frey, 2011b; Jiao *et al.*, 2012).

Key SHEDS-PM inputs include human activity data from the Consolidated Human Activity Database (CHAD), demographic and housing type data from the US Census, air quality data and microenvironment-specific inputs (Burke and Vedamtham, 2009). For the indoor residential microenvironment, SHEDS-PM uses a single-compartment, steady-state mass-balance equation to predict indoor $PM_{2.5}$ concentrations (Burke and Vedamtham, 2009). Air exchange rate (ACH), indoor residential volume, deposition factor and penetrate rate are factors that affect the infiltration of ambient $PM_{2.5}$ and the dilution of non-ambient $PM_{2.5}$:

$$C_r = \left(\frac{P \cdot ACH}{ACH + k} \right) C_a + \frac{E_{smk} N_{cig} + E_{cook} t_{cook}}{\left(ACH + k \right) VT} \tag{3-1}$$

Where,

C_r = indoor residential $PM_{2.5}$ concentration ($\mu g/m^3$)

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

P = penetration factor (unitless)

k = deposition factor (hr^{-1})

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

V = residential volume (m^3)

T = model time step

E_{smk} = emissions rate for cigarette smoking (mg/cig)

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

E_{cook} = emission rate for cooking (mg/min)

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

The first term in Equation (3-1) describes the amount of outdoor $PM_{2.5}$ that infiltrates into the residential microenvironment. The second term describes the $PM_{2.5}$ generated from indoor sources. Equation (3-1) can be extended to include additional indoor sources such as use of cleaning products.

SHEDS-PM assumes that the entire residential unit is one compartment. The single-zone assumption may be reasonable for estimating ambient exposure indoors, because the estimate of indoor concentration of ambient pollution is not sensitive to indoor volume based on Equation (3-1). However, in the presence of an indoor emission source, such as cooking or smoking, the non-ambient exposure will be underestimated (Dimitroulopoulou *et al.*, 2006; Klepeis and Nazaroff, 2006). The division of a house into several rooms affects the distribution of $PM_{2.5}$ concentration. For example, people who are cooking or smoking may be exposed to much higher concentrations in the room where the emission occurs, whereas the concentration may be substantially lower in other rooms. Therefore, the use of a single-zone assumption may lead to significant biases in the estimated exposures. A quantitative comparison between estimated single and multi-zone indoor concentrations is needed. Cooking can be a key source of indoor $PM_{2.5}$, especially affecting the concentration in the

kitchen and the adjacent zones (Cao and Frey, 2011a). Indoor residential smoking is also a potential key source of indoor $PM_{2.5}$ (Klepeis and Nazaroff, 2006). Therefore, the comparison should be carried out based on room-specific emissions from cooking and smoking.

Because the data needs and modeling requirements for simulating multiple zones are significant, it is not practical to implement a multi-zone model directly in SHEDS-PM. Instead, the development of a correction factor may enable a bias correction from the single-zone indoor $PM_{2.5}$ concentration estimates. A correction factor is the ratio of time-weighted average $PM_{2.5}$ concentration in the multi-zone versus single-zone approach.

3.1.2 Objectives

The objectives of this work are to:

(1) Compare indoor residential multi-zone and single-zone modeling approaches;

(2) Develop a bias correction factor to infer indoor exposure concentration from a single-zone simulation;

(3) Assess the sensitivity of correction factor with respect to variation in key inputs; and

(4) Demonstrate the implementation of correction factor.

3.2 Methodology

The methodology includes: (1) review inputs used for estimating indoor residential $PM_{2.5}$ concentration; (2) use an indoor air quality model to compare indoor concentration between single- and multi-zone approaches; (3) develop a correction factor to enable bias correction

to the single-zone estimation; (4) conduct sensitivity analysis using the indoor air quality model to assess the impact of input variation on the correction factor; (4) implement the correction factors with SHEDS-PM; and (5) conduct bounding case analyses to demonstrate the sensitivity of exposure estimates to bias correction.

3.2.1 Mass Balance Inputs

Based on Equation (3-1), the fraction of outdoor $PM_{2.5}$ that infiltrates indoors depends on ACH, k, and P. In addition, parameters such as indoor volume, emission rate and duration from indoor emission sources such as cooking and smoking are critical in estimating indoor residential $PM_{2.5}$ concentration from indoor emissions.

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 (Liu and Nazaroff, 2001). Season-specific air exchange rates are used in SHEDS-PM for the mass balance equation to calculate indoor residential PM concentrations. The default data for ACH for these seasons were originally derived from a perfluorocarbon tracer (PFT) database developed by Brookhaven National Laboratory (BNL). Murray and Burmaster (1995) analyzed the database and categorized ACH by climate region and season. The particle deposition rate k refers to settling of airborne particles due to gravity and diffusion. k depends on particle size, density, room temperature gradients and ventilation conditions (Liu and Nazaroff, 2001). P is the fraction of particles that is not removed from ambient air during its entry into indoors (Wilson *et al.*, 2000) However, there are relatively few data from

which to estimate k and P (Cao and Frey, 2011b). The default values of k and P in SHEDS-PM were obtained from the Particle Total Exposure Assessment Methodology (PTEAM) study conducted for Riverside, California, in the fall of 1990 (Özkaynak *et al.*, 1996).

Cao and Frey (2011b) recommended distributions of ACH, P and k based on results of literature review and sensitivity analysis. Data regarding ACH were reviewed mainly based on Murray and Burmaster (1995) and the Relationship of Indoor, Outdoor and Personal Air (RIOPA) study from 1999 to 2001 (Weisel *et al.*, 2005). Data regarding P and k were reviewed mainly based on the RIOPA (Özkaynak *et al.*, 1996) and PTEAM (Weisel *et al.*, 2005) studies. ACH is distinguishably the most sensitive input for both ambient and non-ambient $PM_{2.5}$ exposure, whereas exposures are relatively insensitive to variations in k and P (Cao and Frey, 2011b).

SHEDS-PM has default values for average indoor residential volumes of different housing stocks in US. These volumes are based on the US Census 2000 Housing Survey (U.S. Department of Commerce, 2013). Typical housing types include single-family detached house, single-family attached house, multi-family apartment, and mobile home.

The typical amount of time that an average individual spends cooking was inferred from CHAD based on diaries that have activity codes for “prepare food” (code = 11100 or 11110). On average, people spent one hour per day for cooking. Although adult females cook an average of one half hour longer per day than adult males, the 5th and 95th percentiles of cooking time did not show any difference by age group or gender. Thus, the duration of cooking was assumed to be one hour for a typical cooking scenario. A default emission rate of 1.56 mg/min was assumed for cooking (Burke *et al.*, 2001).

Since CHAD does not contain information on smoking activity, the assumptions about the typical duration of smoking activity and number of cigarettes smoked were based on literature review. A typical smoker smokes 1.5 packs (30 cigarettes) every day (Nazaroff and Singer, 2004) with an average cigarette smoking duration of 10 min (Ott *et al.*, 2003). Based on the summary of CHAD activity code of "sleep or nap" (code = 14500), people of all ages on average spent 9 hours per day for sleeping. Thus, assumption was made that there are approximately 15 waking hours during which the daily total of 30 cigarettes were smoked. Thus, approximately 2 cigarettes were smoked per hour by an individual. Based on CHAD location codes, the distribution of personal time-activity patterns were categorized into home, outdoors, in-vehicle and other indoors microenvironments, as shown in Table III-1. The home microenvironment is further categorized, in most dairies, into kitchen, living room, bedroom, other specific rooms, and general. "General" is an unspecified location within the home. Approximately 99% of CHAD diaries have location information that identifies the home generally or specific zones within the home. For these diaries, people spent about 16 hours per day in their residence on average. Thus, an assumption was made that a typical individual spends 8 hours per day outside of the residence (9 am to 5 pm) and was awake until 10 pm. Under this scenario, he/she had 5 waking hours at the residence in the evening during which 10 cigarettes were smoked. An emission rate of 13.56 mg/cig was assumed for smoking (Cao and Frey, 2011a).

Table III-1. Distribution of Individual Daily Time Spent in Consolidated Human Activity Database (CHAD)[a]

Information Reported by Microenvironment	Number of Diaries in CHAD	Mean (hr/day)	Median (hr/day)	Standard Deviation (hr/day)
Home: diaries have any location information (specific[b] or general[c])	21455	16.3	16.0	4.6
Home: diaries have both specific and general location information	9946	17.1	17.0	4.3
Home: diaries have only specific location information	7515	15.2	14.8	4.8
Home: diaries have only general information	3994	16.4	16.0	4.4
Home: diaries have activity in kitchen	14208	1.4	1.0	1.3
Home: diaries have activity in living room	14297	3.7	2.9	3.1
Home: diaries have activity in bedroom	17196	10.0	10.0	3.0
Home: diaries have activity in other specific rooms	11995	1.1	0.5	1.6
All outdoors	13470	2.5	1.5	2.7
All in-vehicle	18295	1.5	1.1	1.6
Other indoors	17945	6.1	6.0	4.1

a. The current version of CHAD in SHEDS-PM model contains 21,667 diaries in total. Each diary contains multiple locations information that the individual spent time in, including indoors, outdoors and in-vehicle microenvironments.

b. "Specific" means the diary indicates the specific home location that the individual spent time in, such as kitchen, living/family room, bedroom, dining room, bathroom, study/office, basement, and utility/laundry room.

c. "General" means the diary uses location codes such as "residence, general" (code = 30000), "your residence" (code = 30010), "residence, indoor" (code = 30020), and "your residence, indoor" (code = 30120) to indicate the individual's location at home.

3.2.2 Indoor Air Quality Model

To compare the estimated $PM_{2.5}$ concentration between single- and multi-zone approaches, an indoor air quality (IAQ) model which can simulate $PM_{2.5}$ concentrations under different

emission scenarios and work with either single- or multi-zone approach should be selected. A number of multi-zone airflow models have been developed during the past three decades and have been reviewed and evaluated (Feustel and Dieris, 1992; Emmerich, 2001; Sparks *et al.*, 1991). Examples of such models are CONTAM (Dols *et al.*, 2000), COMIS (Feustel, 1999), and RISK (Sparks, 2005). These multi-zone models use similar equations for predicting indoor air, and are able to simulate both mechanical and natural ventilation in the ventilation system.

RISK model

The RISK IAQ model incorporates results from EPA research on sources, sinks, ventilation, and air cleaners on indoor air quality (Sparks, 2005). RISK can estimate indoor concentrations based on indoor emission sources. Therefore, RISK was used to compare estimated $PM_{2.5}$ concentrations between single- and multi-zone approaches under different emission scenarios.

The RISK model was evaluated by EPA via comparison of model predictions of indoor concentration versus time to experimental data from an EPA IAQ test house. Five sets of experiments covering a wide range of sources, including moth cakes, kerosene heater, dry cleaned clothing, aerosol spray product, and wet products were conducted to evaluate the impact of indoor sources on IAQ. In all cases the concordance between predictions and measurements was verified with an overall coefficient of determination (r^2) of 0.96 (Sparks *et al.*, 1991).

Computational study design

To compare the estimated indoor $PM_{2.5}$ concentrations between single and multi-zone approaches, two modeling cases were formulated in RISK for a typical floor plan of each of the four housing types included in SHEDS-PM and for indoor source emissions scenarios for cooking and smoking. The average single-zone volume used for each housing type in SHEDS-PM equals the average volume of each categorized housing type from the US Census 2000 Housing Survey (U.S. Department of Commerce, 2013). Persily *et al.* (2008) identified a collection of about 200 homes representing 80% of the US housing stock based on two residential housing surveys, the US Department of Energy Residential Energy Consumptions Survey (RECS) and the US Census American Housing Survey (AHS). For each housing type, the floor plan from Persily *et al.* (2008) with indoor volume approximately equal to the average volume from AHS was selected. Multi-zone characteristics regarding the number and volume of rooms were inferred from each selected floor plan. The sum of the volume of all rooms equals the volume used for the single-zone.

For a traditional detached house floor plan (volume = 463 m^3), the kitchen, living room, and dining room are separated by walls. Ten zones were assumed, including living room, kitchen, dining room, den, master bedroom, front bedroom, middle bedroom, master bathroom, main bathroom and hall. For a contemporary attached house floor plan (volume = 371 m^3), the kitchen, living room, and dining room spaces are open with respect to each other. Thus, seven zones were assumed, including living area, master bedroom, front bedroom, middle bedroom, master bathroom, main bathroom and hall.

For the apartment housing type (volume = 242 m^3), six zones were assumed, including living area, master bedroom, front bedroom, master bathroom, main bathroom and hall. For the mobile home (volume = 222 m^3), seven zones were identified, including living area, master bedroom, front bedroom, middle bedroom, main bathroom, master bathroom and hall.

Based on typical heating, ventilation, and air conditioning (HVAC) design practice, five house volumes per hour of air flow are assumed to be distributed to rooms via a supply forced-air HVAC fan (Klepeis and Nazaroff, 2006; Bearg, 1993). In addition to forced-air ventilation, natural ventilation, such as through cracks and crevices in the building envelop, was simulated based on the product of ACH and each room volume.

Multi- and single-zone approaches were compared for three indoor emissions scenarios: (1) no indoor emissions sources; (2) indoor emissions from cooking; and (3) indoor emissions from cigarette smoking. For the no source scenario, the only contributor to indoor $PM_{2.5}$ was penetration of outdoor $PM_{2.5}$, which was assigned an ambient concentration of 10 $\mu g/m^3$. For the cooking and smoking scenarios, the outdoor $PM_{2.5}$ concentration was set to zero.

To aid in specifying a typical individual activity pattern for each emissions scenario, time spent in specific microenvironments was characterized based on CHAD. Table III-1 shows the average individual daily time spent in different rooms. On average, people spent approximately 16 hours at home, of which the average time spent was 1 hour in the kitchen, 4 hours in the living room, 10 hours in the bedroom and 1 hour in other specific or “general” rooms. Figure III-1 characterizes the time spent in each specific microenvironment by hour

of day. Assumptions regarding indoor emissions and activity scenarios were made based on the distribution of activity patterns. A typical individual was assumed to spend one hour in the kitchen from 5 pm to 6 pm, three hours in the living room from 6 pm to 9 pm, and a waking hour in the master bedroom after 9 pm. Therefore, for the cooking scenario, an individual was assumed to cook during the hour in the kitchen. For the typical smoking scenario, the smoker was assumed to smoke in the kitchen for one hour, in the living room for three hours, and in the master bedroom for one hour.

Estimated indoor concentrations were compared for single-zone and multi-zone cases. For the multi-zone case, residential indoor exposure was estimated based on a time-weighted average of the predicted zonal concentrations taking into account the duration of emissions and the time spent by the individual in each zone. For example, a person who cooked or smoked was exposed to the concentration in the zone where these emissions occurred. For times during the day in which emissions did not occur, an average concentration from all zones in the house was used to represent the concentration in the multi-zone approach. The use of an all-zones average concentration is reasonable if the coefficient of variation (CV) of inter-zonal variability in $PM_{2.5}$ concentrations at each simulated time point is small, which was verified.

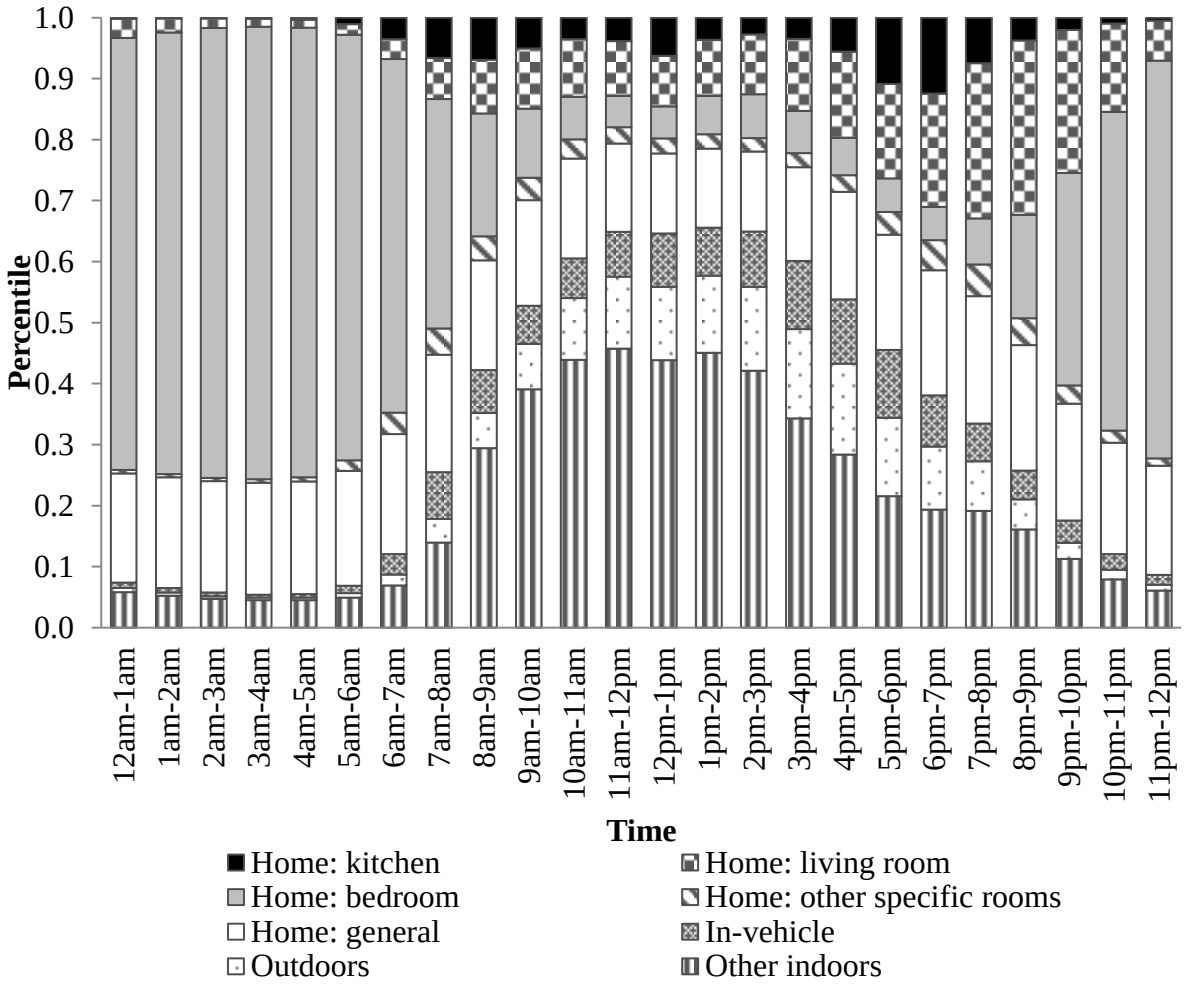


Figure III-1. Distribution of Individual Daily Time-weighted Activity Patterns in Consolidated Human Activity Database (CHAD), All Ages and Genders[a]

[a.] Percentiles of time spent in each location for every one hour is calculated by the sum of time spent in that location divided by the sum of time spent in all locations during that hour.

3.2.3 Correction Factor

Based on Equation (3-1), the indoor concentration of ambient origin, which is only from the penetration of outdoor $PM_{2.5}$, does not depend on indoor volume. Therefore, no correction is needed for the indoor concentration of $PM_{2.5}$ of ambient origin. However, a correction is

needed for contributions to indoor concentration that are from indoor sources such as cooking and smoking because these depend on the effective volume of dispersion. Correction factors were estimated for cooking (CF_{cook}) and smoking (CF_{smk}) scenarios, separately, based on the activity time-weighted average of zonal concentrations from the multi-zone model results divided by the average concentration from the single-zone case for the same time period:

$$CF_{cook} \text{ or } CF_{smk} = \frac{\sum_{i}^{I}(C_{i,t} \times t_i)/T}{C_{s,t} \times t_s / T} = \frac{\sum_{i}^{I}(C_{i,t} \times t_i)}{C_{s,t} \times t_s} \tag{3-2}$$

Where,

CF_{cook} = correction factor for emission source cooking

CF_{smk} = correction factor for emission source smoking

i = the room where people spent time

I = total number of rooms where people spent time

$C_{i,t}$ = average $PM_{2.5}$ concentration of room i in the multi-zone approach (μg/m^3)

t_i = time spent in room i in the multi-zone approach

$C_{s,t}$ = average $PM_{2.5}$ concentration estimated by the single-zone approach (μg/m^3)

t_s = time spent in any location in the house for the the single-zone approach

t_s = $\sum t_i$

T = duration of one day, 24 hour

Correction factors apply to the respective terms in the mass balance equation that deal with the relevant non-ambient emission source. The application of the correction factor to the mass balance is:

$$C_{bias-corrected} = \left(\frac{P \cdot ACH}{ACH + k}\right) C_a + \frac{E_{cook} t_{cook}}{(ACH + k)VT} \times CF_{cook} + \frac{E_{smk} N_{cig}}{(ACH + k)VT} \times CF_{smk} \quad (3\text{-}3)$$

Where,

$C_{bias\ corrected}$ = bias-corrected indoor $PM_{2.5}$ concentration ($\mu g/m^3$)

CF_{cook} = correction factor for cooking

CF_{smk} = correction factor for smoking

3.2.4 Sensitivity Analysis

Nominal range sensitivity analysis (NRSA) (Frey and Patil, 2002) was applied to identify key inputs to which the correction factor was most sensitive and to assess the effect of inter-individual variability in time-activity patterns on personal exposure levels. ACH, k, cooking emission rate, cooking duration, smoking emission rate, and number of cigarettes smoked were varied individually, while holding all other inputs unchanged from the baseline typical scenario case.

For ACH, 2.5th and 97.5th percentile values were inferred from the lognormal distributions indicated by Cao and Frey (2011b) for use as the lower and upper limits, respectively, in the sensitivity analysis. The 5th, 10th, 90th, and 95th percentiles were also included to enable evaluation of nonlinearity in concentration versus ACH. For k, 0.2 and 0.6 h^{-1} were chosen as the range, compared to the value of 0.4 h^{-1} in the baseline case (Cao and Frey, 2011b). For cooking, a range of emissions rates from 1.0 to 2.3 mg/min was assumed based on the PTEAM study (Klepeis and Nazaroff, 2006). For cooking duration, the 2.5th, 5th, 10th, 90th, 95th and 97.5th percentiles of individual daily average cooking time from CHAD

were used to represent inter-individual variability. For smoking emission rate, a lower limit of 8 mg/cig and an upper limit of 23 mg/cig were assumed (Burke *et al.*, 2001). The number of cigarettes smoked was varied from 1 to 3 cig/hr to account for inter-individual variability in time-activity patterns for smoking (Nazaroff and Singer, 2004).

3.2.5 Implementation of Correction Factors

The implementation of correction factors was demonstrated via an example case study in SHED-PM. Population exposures were simulated for 50,000 individuals over a one month period in July, 2002, for a six county domain along the I-40 corridor in North Carolina. SHEDS-PM was run twice to account for residential non-ambient source from cooking and smoking scenarios, separately. To repeat the same sample of activity diaries for both simulations, the same random seed was used. Estimated indoor residential non-ambient concentrations from these two scenarios were added to account for each individual's exposure to indoor sources. Results were separated by housing type. Correction factors were applied to each simulated person by emission source and housing type. Exposure concentrations were compared before and after the application of correction factors.

3.2.6 Low Exposure Bounding Case

In reality, a household can have multiple residents. A situation could happen in which only one person cooks or smokes, and other persons in the house may intentionally choose to reduce their $PM_{2.5}$ exposure by avoiding the room in which emissions occur. Thus, as a low exposure bounding case analysis, the exposure concentrations for a hypothetical person who

avoids non-ambient emissions from others in the house were compared between multi- and single-zone approaches. For each housing type simulated, the high exposure-avoiding person was assumed to stay in the front bedroom during all non-ambient emission occurrences. Correction factors for these avoidance cases were developed for each emission scenario and housing type. Sensitivity analysis was conducted to assess the relative importance of mass balance inputs on the multi-zone to single-zone concentration ratio.

3.3 Results

Single- and multi-zone approaches for estimating indoor residential $PM_{2.5}$ concentrations under different emission scenarios were modeled using RISK version 1.9. Concentrations were estimated based on a model default 0.2 hour time interval for each assumed zone during the day.

3.3.1 Comparison between Single- and Multi-Zone Concentrations

When no indoor sources were present, the difference in estimated $PM_{2.5}$ concentrations between the single- and multi-zone approaches was within 5%, with an absolute difference as low as 0.15 $\mu g/m^3$. The similarity in estimated indoor concentration between these two approaches indicates that the assumption of a single well-mixed zone is acceptable if there are no indoor emission sources.

However, when indoor cooking or smoking emissions were included, the multi-zone approach estimated 7% to 204% higher daily average $PM_{2.5}$ exposure concentrations than the single-zone, depending primarily on factors such as housing type and ACH. As an example,

results are detailed here for the traditional single-family house. The housing dimensions and room specifications are specified in Table III-2. Table III-3 presents mass balance equation inputs used in baseline cases and sensitivity analysis of the RISK model simulations.

Table III-2. Housing Dimensions and Room Specifications for Typical Traditional Single-family Detached House

Room	Volume[a] (m^3)	HVAC Flows[b] (m^3/hr)
Den	23	(+) 110
Kitchen	72	(+) 360
Dining Room	27	(+) 135
Living Room	108	(+) 540
Hall	37	(-) 2125
Middle Bedroom	33	(+) 160
Front Bedroom	35	(+) 170
Master Bedroom	86	(+) 430
Main Bathroom	14	(+) 70
Master Bathroom	29	(+) 145
Total	463	

[a.] The total house volume of 463 m^3 is based on results of the US Census 2000 Housing Survey.

[b.] Forced air flow for supply (+) or return (-) to/from the house heating, ventilation, and air conditioner (HVAC) system is approximately five house volumes per hour.

Table III-3. Mass Balance Inputs to RISK Model for Cooking and Smoking Scenarios

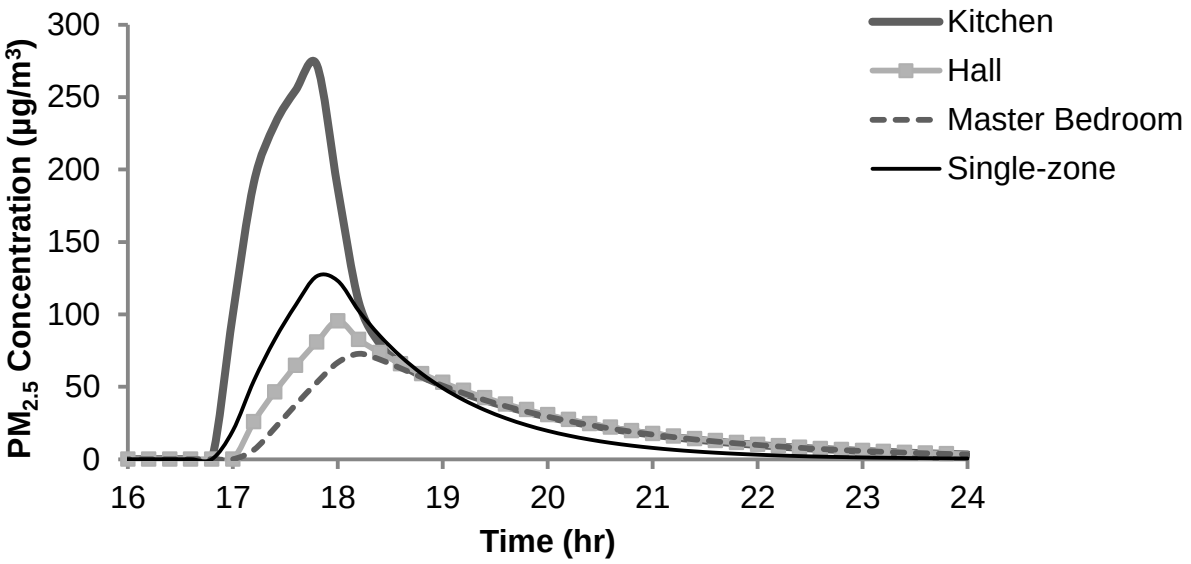
Parameters	Baseline	Sensitivity	
		Lower	Upper
Cooking emission rate (E_{cook}, mg/min)	1.56[a]	1.00[b]	2.30[b]
Duration of cooking (t_{cook}, min)[c]	60	5	240
Smoking emission rate (E_{smk}, mg/cig)[d]	13.56	8.00	23.00
Number of cigarettes smoked (N_{cig})[e]	10	5	15
ACH (h^{-1})[f]	0.5	0.06	2.71
k (h^{-1})[d]	0.4	0.2	0.6

Sources: [a.] Burke *et al.* (2001)
[b.] Ozkaynak *et al.* (1996)
[c.] Consolidated Human Activity Database (CHAD)
[d.] Cao and Frey (2011)
[e.] Nazaroff and Singer (2004)
[f.] Murray and Burmaster (1996), Cao and Frey (2011)

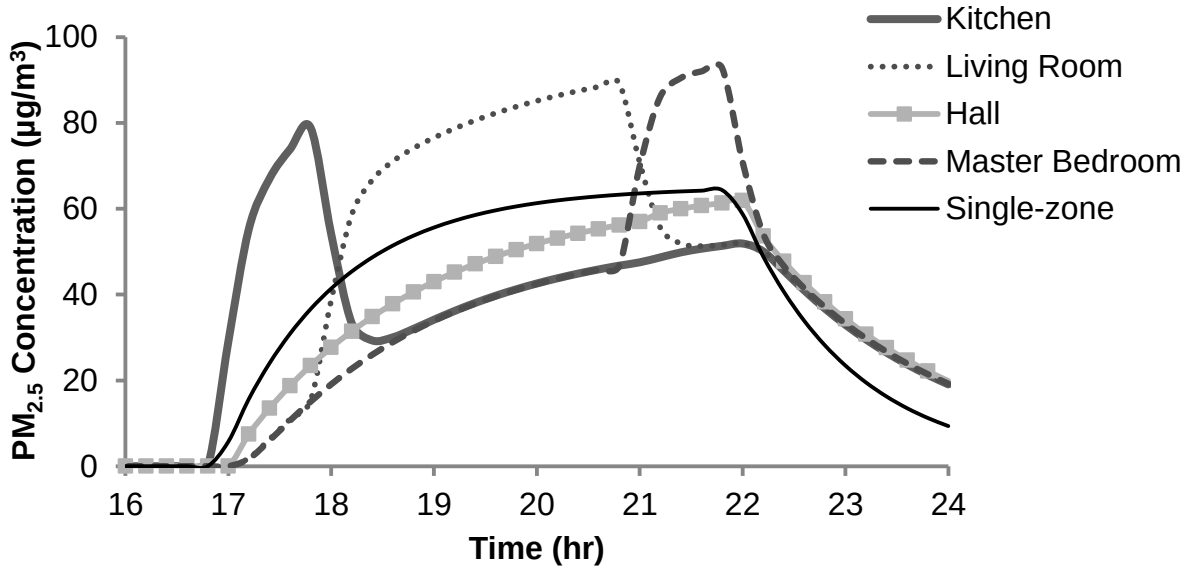
The daily average exposure concentrations are compared between activity-based multi-zone and single-zone estimates in Table III-4 for typical cooking and smoking scenarios. The multi-zone approach estimated 14% to 77% higher exposure concentrations than the single-zone approach. Estimated exposure concentrations to $PM_{2.5}$ varied by room inside a house, depending on where and how long the non-ambient sources were emitting, as well as personal activity across rooms. For the cooking scenario, since non-ambient emissions only occurred in the kitchen, and $PM_{2.5}$ diluted quickly after the cooking event and became homogeneous across rooms, the activity-based daily average multi-zone exposure concentration was approximately equal to the daily average microenvironmental concentration of the kitchen for each housing type. However, for the smoking scenario, exposure estimates from the multi-zone approach were higher than any daily average single room concentration. Room-specific concentrations depend on the duration of emissions inside the room, as well as the ACH.

Cooking

To compare the estimated indoor residential zonal $PM_{2.5}$ concentrations for the cooking scenario, results for the traditional detached house are shown in Figure III-2(a). The peak $PM_{2.5}$ concentration was estimated to be 270 $\mu g/m^3$ in the kitchen, considerably higher than those estimated for other rooms, which ranged from 70 to 95 $\mu g/m^3$. A lag effect of about 10 to 20 minutes on peak $PM_{2.5}$ concentrations was observed in rooms other than kitchen. The



(a) Cooking



(b) Smoking

Figure III-2. Comparion of Estimated $PM_{2.5}$ Concentrations between Single- and Multi-zone Assumptions, Cooking and Smoking Scenarios, Traditional Single-family Detached House

Table III-4. Comparisons of Daily Average Zonal Concentrations (μg/m^3), Typical Exposure[a]

Scenario	Housing Type	Daily Average Exposure Concentrations		Daily Average Multi-zone Microenvironmental Concentrations			
		Activity-based Multi-Zone Average	Single-Zone Average	Kitchen	Living Room	Master Bedroom	Front Bedroom
Cooking	Traditional Single-family House	16.6	9.4	17	7.8	7.8	7.8
	Contemporary Single-Family House	13.4	11.7	13.5	13.5	9.4	9.4
	Multifamily Apartment	21	17.9	21.2	21.2	14.4	14.4
	Mobile home	22.8	19.5	23	23	15.8	15.8
Smoking	Traditional Single-family House	20.5	13.6	14.1	16.7	13.1	11.4
	Contemporary Single-Family House	21.1	16.9	18.9	18.9	16.6	14.1
	Multifamily Apartment	29.6	25.9	26.9	26.9	22.1	19.1
	Mobile home	35.9	28.3	32.2	32.2	28	23.9

[a.] Based on the typical exposure activity pattern in which cooking occurs in kitchen or kitchen/living room combined area from 5 pm to 6 pm; and smoking occurs in kitchen or kitchen/living room combined area from 5 pm to 6 pm, in living room from 6 pm to 9 pm, in master bedroom from 9 pm to 10 pm.

dispersion of $PM_{2.5}$ from kitchen to other rooms was affected by the air flow rate between rooms and their proximity to the kitchen. The single-zone approach had an estimated peak indoor $PM_{2.5}$ concentration of 130 μg/m^3, which was approximately half of the peak concentration predicted for the kitchen in the multi-zone case. However, the peak single-zone concentration was substantially higher than for other rooms in the multi-zone case.

For a typical person who cooks one hour a day, the daily time-weighted average $PM_{2.5}$ exposure concentration estimated by the multi-zone approach was 17 μg/m^3, compared to 9 μg/m^3 from the single-zone approach. The time-weighted multi-zone concentration was based on the concentration in the kitchen during cooking, and average concentrations from all zones for all other times. For each simulated time except when cooking, $PM_{2.5}$

concentrations were approximately similar across zones. The CV for inter-zonal variations in concentration decreased from 0.1 within the first half hour after cooking activity ended to only 0.01 when room-specific $PM_{2.5}$ concentrations became homogenous across zones approximately a half hour after cooking activity ended. Therefore, the use of an all-zones average concentration as the generic exposure concentration for exposures that takes place a half hour after cooking does not introduce significant error. Overall, the single-zone approach under-estimated personal exposure by approximately 43% versus the multi-zone approach. Likewise, the single-zone approach underestimates the personal $PM_{2.5}$ exposure concentration by 13% in the attached house, 15% in the multi-family apartment, and 15% in the mobile home than the multi-zone approach under typical cooking scenario. The lower percentages for these housing types were mainly because they have a larger kitchen/living room combined area instead of an isolated smaller kitchen in the traditional detached house.

Smoking

The variations of estimated zonal indoor residential $PM_{2.5}$ concentrations under the typical smoking scenario for the traditional detached house are shown in Figure III-2(b). The estimated peak $PM_{2.5}$ concentrations were 80 $\mu g/m^3$ in the kitchen, 90 $\mu g/m^3$ in the living room, and 95 $\mu g/m^3$ in the master bedroom. In contrast, the single-zone approach had an estimated peak indoor $PM_{2.5}$ concentration of 65 $\mu g/m^3$. The daily time-weighted average $PM_{2.5}$ exposure concentration predicted for the multi-zone approach was 21 $\mu g/m^3$, compared to 14 $\mu g/m^3$ for the single-zone approach. Thus the single-zone assumption underestimated the daily exposure concentration by 34%. The combined kitchen/living room area in other

housing types has larger volumes than the single kitchen in detached house, which facilitates pollutant deposition and dilution within the room. Therefore, the single-zone prediction of personal daily $PM_{2.5}$ exposure concentration was 20% lower in the attached house, 13% lower in the apartment, and 21% lower in the mobile home than that of the multi-zone approach.

3.3.2 Development of Correction Factors

No correction was needed for predicting residential $PM_{2.5}$ concentrations of ambient origin. However, for residences with non-ambient cooking or smoking emissions, Table III-5 shows the inferred cooking and smoking correction factors for each selected housing type for both typical exposure and exposure avoidance scenarios, separately. For the traditional single family detached house, the cooking correction factor estimated for a typical exposure scenario is 1.77, and the smoking correction factor is 1.51. Correction factors were sensitive to the room volume where emissions occurred. For example, the traditional house had a separate kitchen with smaller area compared to other housing types. Thus, higher time-weighted exposure concentrations were found during cooking, which resulted in a larger correction factor than for other housing types.

Table III-5. Correction Factors for Different Housing Types, Baseline Case

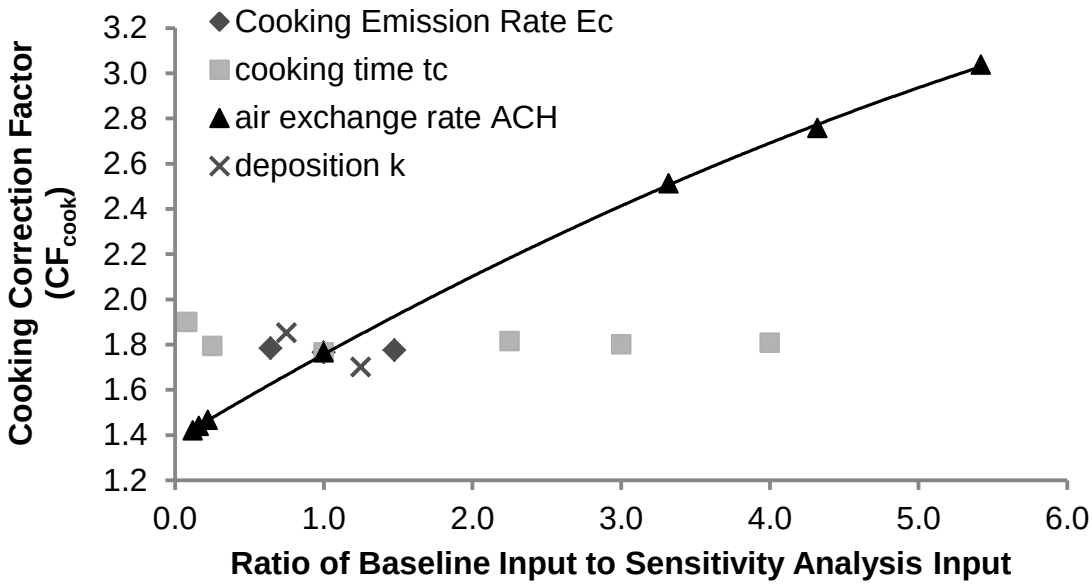
Housing Type	Typical Exposure		Low Exposure (Avoidance)	
	Cooking	Smoking	Cooking	Smoking
Traditional Single-family House	1.77	1.51	0.8	0.8
Contemporary Single-Family House	1.15	1.25	0.8	0.8
Multifamily Apartment	1.17	1.14	0.8	0.8
Mobile home	1.17	1.27	0.8	0.8

For the exposure avoidance case studies, the correction factors in Table III-5 are less than one. The $PM_{2.5}$ concentration in the room where emissions occur was up to a factor of 30 higher versus the front bedroom during an emissions event. However, differences in zonal concentrations became smaller as the pollutant gradually diluted into other rooms of the house. Under the typical cooking or smoking emission scenarios, the daily time-weighted exposure avoidance activity pattern average $PM_{2.5}$ concentrations predicted by the multi-zone approach for all four housing types were approximately 20 percent lower than the concentrations estimated by the single-zone approach for all selected housing types. Therefore, for individuals who attempt to avoid exposure to indoor sources, the single-zone approach will over-estimate personal exposure by about 20% than the multi-zone approach in the baseline case.

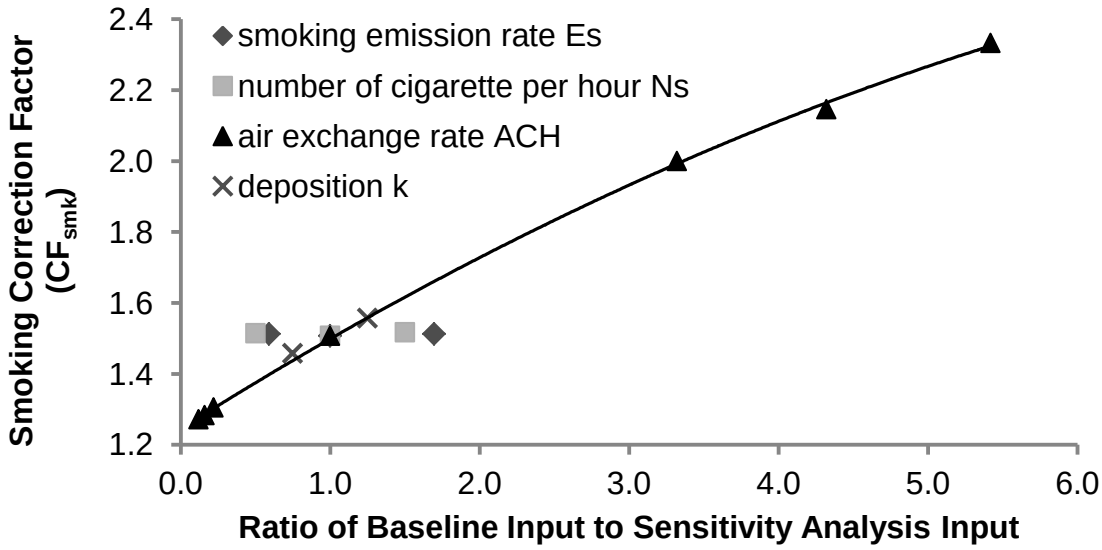
3.3.3 Sensitivity of Correction Factor to Input Variation

To assess the relative effect of inputs on correction factors, and to account for effects of inter-individual variability in personal activity, sensitivity analysis was conducted. Results of

sensitivity analysis for the traditional detached house under the typical exposure scenario are shown in Figure III-3 as an example. Changes in source emission rate, and cooking duration



(a) Cooking



(b) Smoking

Figure III-3. Sensitivity Analysis of Correction Factor, Cooking and Smoking Scenarios, Traditional Single-family Detached House

time or number of cigarettes smoked resulted in little variation in the correction factors, mainly because these parameters were linearly proportion to non-ambient concentration based on Equation (3-1). In either the multi-zone or single-zone approach, the ratio of non-ambient concentration between the input-varied case and the baseline case equaled the ratio of varied input divided by the base-case input. Thus, the relative proportion of multi-zone versus single-zone concentration stayed the same and was not sensitive to these input variations. However, a positive non-linear relationship was observed between correction factors and ACH, mainly because non-ambient concentration is not linearly related to ACH. The correction factor for the 97.5th percentile ACH was 114% higher for cooking and 55% higher for smoking, compared to the 2.5th percentile of ACH.

For all four housing types simulated, correction factors of the bounding exposure avoidance cases all varied within a much smaller range of 0.6 to 0.9 for changes in ACH between the 2.5th percentile and the 97.5th percentile. Correction factors were relatively insensitive to variations in other inputs.

Polynomial equations for correction factors versus ACH were fit to the sensitivity analysis results for typical exposure and avoidance scenarios, separately, as summarized in Table III-6. Taking a typical exposure scenario in a traditional house for an example, CF_{cook} can be inferred from:

$$CF_{cook} = -0.067 \times ACH^2 + 0.79 \times ACH + 1.38 \qquad (3\text{-}4)$$

For a representative residential ACH of 0.5, the inferred CF_{cook} of the detached house is 1.76.

Table III-6. Relationships between Correction Factor (CF_{cook} or CF_{smk}) and Air Exchange Rate (ACH) for Selected Housing Types[a]

CF_{cook} or $CF_{smk} = a(ACH)^2 + b(ACH) + c$												
Housing Type	Typical Exposure						Low Exposure (Avoidance)					
	Cooking (CF_{cook})			Smoking (CF_{smk})			Cooking (CF_{cook})			Smoking (CF_{smk})		
	a	b	c	a	b	c	a	b	c	a	b	c
Traditional Single-family House	– 0.067	0.79	1.38	– 0.049	0.53	1.25	0.019	– 0.17	0.91	0.015	– 0.15	0.91
Contemporary Single-Family House	– 0.0074	0.14	1.12	– 0.022	0.26	1.12	0.032	– 0.22	0.94	0.018	– 0.17	0.91
Multifamily Apartment	– 0.020	0.22	1.09	– 0.006	0.23	1.10	0.020	– 0.17	0.91	0.034	– 0.20	0.90
Mobile home	– 0.013	0.19	1.10	0.22	0	1.14	0.023	– 0.18	0.91	0.013	– 0.15	0.91

[a.] Each relationship was inferred based on 7 cases of ACH.

3.3.4 Implementation of Correction Factors

The daily average residential non-ambient concentration estimated using the single-zone approach in SHEDS-PM was 28.0 μg/m^3, of which 4.9 μg/m^3 came from cooking and 23.1 μg/m^3 from smoking. After bias corrections using equations in Table III-6 by emission scenario and housing type, the estimated residential concentration of non-ambient origin increased by 31% to 36.8 μg/m^3. Since most people spend the majority of their time at home, changes in indoor residential concentration have a significant impact on daily total exposure levels. The corrected daily average total exposure was 36.3 μg/m^3, which is 17 percent higher that of 30.9 μg/m^3 before bias correction.

3.4 Conclusions

Residential non-ambient $PM_{2.5}$ concentrations from indoor sources predicted by a single-zone approach are underestimated. The assumption of a well-mixed single-zone was found to be reasonable in the case of no indoor sources. However, in the presence of cooking or smoking, the assumption was not valid. An approach for applying a cooking or smoking correction factor to the original single-zone mass balance equation was demonstrated for a traditional single-family detached house. The correction factor is most sensitive to variations in ACH. Polynomial relationships were identified for each of several housing types for cooking and smoking emission scenarios, separately. Correction factors address the difference in estimated indoor exposure concentrations between the multi- and single-zone approaches, and can be easily applied to the single-zone results based on relationships with ACH. The

estimated individual daily average total exposure levels increased with the application of correction factors.

The results show the importance of indoor sources on the estimated total daily average exposure to $PM_{2.5}$, which can contribute to extremely high daily average exposures for some individuals due to non-ambient emissions from cooking or smoking. Identification and appropriate quantification of these emissions in different housing types can assist in assessing upper percentiles of population exposures and identifying homes where the health risks are likely to be highest.

Distributions of indoor $PM_{2.5}$ concentrations with respect to a range of different emissions scenarios and individual time-activity patterns are critical for the effective health risk assessment of indoor air. Based on the results of sensitivity analysis, the potential for reducing indoor concentrations and exposures is not only limited to control of outdoor and indoor sources. Building characteristics such as ACH and room size have a significant effect on both mean and peak exposures. The magnitude of non-ambient exposure for a given indoor source strength is inversely proportional to ACH and to interior room volume. Thus, for indoor air research and policy, the role of building characteristics in reducing indoor human exposure to $PM_{2.5}$ merits further consideration .

The current U.S. national standard to prevent $PM_{2.5}$ adverse effects is based on 24 hour average ambient concentration. However, peak 1 hour average indoor concentrations were estimated to be high for both cooking and smoking scenarios. The potential health and policy significance associated with such peak concentrations merits further assessment. Persons within a house who are not engaged in emissions producing activities have the

option of engaging in exposure avoidance behavior. For example, results of bounding cases considered here indicate that exposures from non-ambient sources can be reduced for persons not engaged in emissions producing activities by changing their indoor location. However, those engaged in activities such as cooking would have to reduce their emissions or increase indoor ventilation to reduce exposure.

3.5 Acknowledgements

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PART IV METHOD FOR MEASURING THE RATIO OF IN-VEHICLE TO NEAR-VEHICLE EXPOSURE CONCENTRATIONS OF AIRBORNE FINE PARTICLES♦

♦ This manuscript was recommended by the Transportation Research Board's Transportation and Air Quality Committee for publication in *Transportation Research Record: Journal of the Transportation Research Board*, 2013.

Abstract

Human exposure to fine particulate matter ($PM_{2.5}$) is causally linked to cardiovascular and pulmonary diseases. In-vehicle exposure may account for 10 to 20 percent of daily average exposure. However, exposure models are typically based on area-wide air quality data that poorly predict in-vehicle concentration. A practical method is demonstrated for conducting field measurements to quantify the ratio of in-vehicle to outside vehicle concentration (I/O) for a wide range of conditions that affect intra-vehicle variability in exposure concentration. A field data collection study design is developed based on sources of intra-vehicle variability in I/O, including ventilation air source, window status, fan setting, AC utilization, vehicle speed, road type, travel direction, and time of day. Three replicates of measurements were made for 16 combinations of these factors on 110 miles of roads comprised of eight one-way routes between typical commuter origin/destination pairs. Two portable particle monitors recorded in-vehicle and near-vehicle ambient concentrations on one minute averages for four particle size ranges. The comparability of the monitors was quantified. Near-vehicle concentrations varied with road type, time of day, and traffic conditions. The I/O ratio was approximately independent of near-vehicle concentration, and varied with window status, source of ventilation air (fresh or recirculation), and, for cases with recirculation and closed windows, fan setting and air conditioning use. The study design can be extended to additional vehicles to account for potential sources of inter-vehicle variability. Data collected here can be used to improve exposure simulation models.

4.1 Introduction

Human exposure to fine particulate matter of less than 2.5 microns in aerodynamic diameter ($PM_{2.5}$) is causally linked to cardiovascular and pulmonary diseases (U.S. EPA, 2009a). Since the ambient concentrations of particulate matter (PM) can be higher on or near roadways, the time spent in or near vehicles can contribute disproportionately to $PM_{2.5}$ exposure (Liu and Frey, 2011). In 2007, approximately 90% of U.S. commuters drove to work (U.S. DOT, 2012). Based on the National Human Activity Pattern Survey (NHAPS), Americans spent average 1.3 hours per day in a vehicle (Klepeis *et al.*, 2001).

Depending on traffic flow, meteorological conditions, vehicle emission rates, whether windows are open, operation of the vehicle heating, ventilation, and air conditioning (HVAC) system, and time spent in-vehicle, in-vehicle exposure may account for 10 to 20 percent of total daily average $PM_{2.5}$ exposure (Liu and Frey, 2011). In-vehicle concentrations (C_{IV}) are dependent on near-vehicle ambient concentration (C_{NV}) along a roadway, which in turn is influenced by local traffic, and on the penetration of ambient air into the vehicle. However, many population-based exposure models estimate C_{IV} based on a ratio to a fixed site monitor (FSM) (U.S. EPA, 2009a).

For example, the Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM) model estimates C_{IV} based on a user-specified ratio of in-vehicle to either FSM concentration or area-wide ambient concentration estimated from an air quality model (Burke and Vedamtham, 2009). However, a FSM can be far from the roadway and is not a good indicator of C_{NV} (Adams *et al.*, 2001; Gulliver and Briggs, 2004; Kaur *et al.*, 2007). C_{IV} is more accurately estimated based on C_{NV} rather than FSM or area-wide

concentration. Therefore, for exposure modeling purposes, it is more accurate to estimate ambient concentrations based on a parametric model for the ratio of C_{IV}/C_{NV}, defined here as the I/O ratio k' (Liu and Frey, 2011):

$$C_{iv} = k'C_{amb} + k'C_{icr} + b + \varepsilon \tag{4-1}$$

Where,

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

k' = ratio of in-vehicle to near-vehicle ambient $PM_{2.5}$ concentration, I/O ratio

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

C_{icr} = incremental component of ambient concentration on a roadway ($\mu g/m^3$)

b = in-vehicle non-ambient $PM_{2.5}$ concentration ($\mu g/m^3$)

ε = unbiased error term for residual variability in in-vehicle $PM_{2.5}$ concentration

The focus of this work is to demonstrate an approach for measuring the I/O ratio for $PM_{2.5}$ and other selected PM size ranges.

Several studies have focused on the ratio of C_{IV} to FSM concentrations, with a wide range of variability in findings. These ratios are far in excess of unity in some cases, and much lower in others (Adams *et al.*, 2001; Riediker *et al.*, 2003). More fundamentally, the I/O ratio is sensitive to the air exchange rate (ACH). ACH is ambient air flow into the vehicle divided by vehicle cabin volume. Variability in ACH was measured by Ott *et al.* (2008) based on controllable factors such as window status, HVAC fan setting, and AC on or off. Based on earlier mass balance models (Switzer and Ott, 1992; Allen *et al.*, 2007; Abi-

Ester and El-Fadel, 2008), Liu and Frey (2011) developed a mass balance model to predict the I/O ratio. They estimated I/O ratios for $PM_{2.5}$ from 0.82 to 0.99 for cases with high advection of outside air, and from 0.5 to 0.8 for cases with HVAC air recirculation and closed windows. The estimated I/O ratio was not sensitive to variation in particle deposition rate.

Empirical studies of other particle size ranges and other exposure microenvironments illustrate factors that might affect the I/O ratio. Zhu *et al.* concluded that the interior to outdoor concentration ratio for ultrafine particles (UFPs) of 100 nanometers aerodynamic diameter or less does not depend on the magnitude of the outdoor concentration (Zhu *et al.*, 2007). Rather, factors determining the ratio are a function of particle size and vehicle characteristics. Residential housing indoor to outdoor ratios ranged from 0.8 to 1.0 for UFPs, 0.7 to 0.8 for $PM_{2.5}$, and 0.1 to 0.2 for larger particles (Long *et al.*, 2001). Thus, as particle size increases, the I/O ratio is expected to decrease. However, the relative variation in the I/O ratio for vehicles may differ from that for houses. For example, vehicles have higher passive ventilation as a result of driving speed (Fletcher and Saunders, 1994).

The focus here is on development and demonstration of a practical method for conducting field measurements to quantify the I/O ratio for a wide range of conditions that affect intra-vehicle variability. The latter include the source of HVAC air intake, status of windows, HVAC fan setting, AC utilization, and passive ventilation. Thus, the objectives are to: (1) assess the variability in $PM_{2.5}$ concentrations for near-vehicle and in-vehicle microenvironments; (2) assess the variability in the I/O ratio; (3) assess repeatability among replicate measurements; and (4) compare the I/O ratio among particle size ranges.

4.2 Methodology

Measurement of in-vehicle PM exposure includes: (1) preparation of instruments; (b) study design for the field data collection; and (3) data quality assurance and analysis of results.

4.2.1 Instruments

The TSI DustTrak monitor is a battery operated, light-scattering laser photometer that records real-time readings proportion to aerosol mass concentrations for a specified particle size range. DustTrak monitors have been widely used in previous studies (Wu *et al.*, 2002; Zhu *et al.*, 2006; Diapoulu *et al.*, 2007; Wang *et al.*, 2009). The DustTrak DRX Aerosol Monitor 8533 simultaneously measures size-segregated mass fraction concentrations corresponding to PM_1, $PM_{2.5}$, PM_4, and PM_{10}. The DustTrak was factory-calibrated to Arizona Test Dust (ISO 12103-1). One DustTrak sampled C_{IV} near the driver breathing zone. A second sampled C_{NV} via a tube routed through a passenger window. The operational temperature of the DustTrak 8533 is from 0 °C to 50 °C (TSI, 2012). Temperatures inside and outside the vehicle were within this range. The outside tube was taped onto the vehicle window so that the inlet was oriented in the down-wind direction. The DustTrak uses a pump to actively sample at 3.0 L/min, and thus draws a continuous sample. To evaluate whether the orientation of the sampling probe could lead to bias in results, an experiment with the external probe inlet oriented in the up-wind direction was done for cases with very high air exchange rate. The window gap around the tube was sealed with duct tape. An averaging time of one minute was used for both DustTraks.

Vehicle cabin temperature (T) and relative humidity (RH%), were recorded for one minute averages using a HOBO U14 logger. The position and movement of the vehicle was recorded at 1 Hz using a Garmin 76CSx GPS with barometric altimeter.

4.2.2 Study Design

The key goal is to quantify the I/O ratio for PM_1, $PM_{2.5}$, PM_4, and PM_{10} based on the most significant controllable or observable sources of variability. The focus here is on intra-vehicle variability, which is variability during the course of operating a single vehicle on a trip. All vehicles are expected to be subject to similar sources of intra-vehicle variability. Thus, the study design can be replicated later with more vehicles to address inter-vehicle variability.

Sources of Intra-Vehicle Variability

Intra-vehicle variability is the variation in C_{NV}, C_{IV}, and I/O from the perspective of the vehicle as it moves along a path. Based on prior studies concluding that I/O is independent of C_{NV} (Liu and Frey, 2011; Zhu *et al.*, 2007), the study design is conceptually divided into two components: (1) controllable factors affecting variability in C_{IV} and I/O; and (2) uncontrollable but potentially observable factors affecting variability in C_{NV}. The key controllable sources of intra-vehicle variability include HVAC air source, window status, HVAC fan setting, and air conditioning utilization. Vehicle speed, which affects passive ventilation, is potentially controllable but for safety reasons has to be in harmony with existing traffic flow. Thus, real-world speed is observable but not fully controllable. The

range of variability encountered for uncontrollable factors is influenced by choices for surrogate study design factors, including choices of routes, travel directions, and time of day.

Air Source The driver usually can control whether the HVAC system intakes fresh ambient or recirculated cabin air, with or without concurrent AC operation. For closed windows, use of recirculated air leads to lower infiltration of pollution from outside the vehicle (Zhu *et al.*, 2007; Hill and Gooch, 2007). Air recirculation reduces penetration of particle-laden fresh air into the cabin and enhances the particle deposition to interior surfaces (Riediker *et al.*, 2003). Use of fresh substantially increases the bulk flow of particle-laden ambient air into the cabin (Abi-Esber and El-Fadel, 2008).

The location and performance of a cabin air filter can affect the I/O ratio. If a large portion of cabin air flow is through the filter, then the filter may have a significant role in lowering C_{IV} and I/O. However, there is a lack of published data on the in-use efficiency of such filters.

Window Status Opening a window while a vehicle is moving substantially increases the air exchange rate (Ott *et al.*, 2008; Knibbs *et al.*, 2009; Abi-Esber and El-Fadel, 2012). For example, opening a window three inches can increase ACH by an order-of-magnitude. In the limit, having all windows fully open would be expected to produce I/O approaching unity.

Fan Setting There has been relatively little evaluation of the role of the HVAC fan setting on ACH, C_{IV}, or I/O. For fresh air and fan off, there can be significant airflow through the

HVAC ducts from passive ventilation, depending on vehicle speed (Fletcher and Saunders, 1994). For a stationary or slow moving vehicle, increasing the fan speed with fresh air would be expected to increase the duct air flow, thereby increasing C_{IV} and I/O. If recirculating air is used, then passive ventilation may be negligible unless there is a poor seal in the duct damper.

Air Conditioning C_{IV} tends to decrease in association with AC utilization (Chan *et al.*, 2002; Tsai *et al.*, 2008). This could be in part because vehicles equipped with AC may be more likely to have a cabin air filter. For example, PM_{10} concentrations in non-air conditioned taxis and buses were 72% and 59% higher, respectively, than those with AC (Chan and Chung, 2005). Temperature may be a factor for some particle size ranges. For example, higher UFP number concentrations are typically observed in the winter compared to the summer, because of an increase in nucleation mode particle formation (Zhu *et al.*, 2004). However, studies that quantified correlation between ambient temperature and in-vehicle $PM_{2.5}$ concentration did not report a significant effect (Zagury *et al.*, 2000; Kaur and Nieuwenhuijsen, 2009).

Vehicle Speed ACH increases with vehicle speed, especially when a window is open or if fresh air intake is used (Knibbs *et al.*, 2009). Passive ventilation is air movement through an open duct because of pressure differences. An empirical estimate of ACH for passive ventilation is sensitive to vehicle speed (Fletcher and Saunders, 1994):

$$ACH = 0.60V^{1.25} \quad (4\text{-}2)$$

Where,

ACH = air exchange rate (1/hr)

V = vehicle speed (m/s)

For example, a vehicle traveling at 65 mph would have an estimated passive ventilation ACH of 39 hr^{-1}.

Road Type and Time of Day Spatial variability in C_{NV} is influenced by number of lanes of traffic, traffic flow, vehicle emission rates, wind speed, and atmospheric stability (Liu and Frey, 2011). The surrogate study design variables of the choice of time of day, route, and travel direction can influence whether free-flow or congested traffic is likely to be encountered.

C_{NV} can vary from one route to another, depending in part on traffic density and dispersion characteristics of the road (Briggs *et al.*, 2008). Vehicle exhaust pollutant concentrations, such as for NO_x and $PM_{2.5}$, are typically elevated during congested traffic (Janssen *et al.*, 2003). C_{NV} will differ by road types that have different levels of traffic, which is the number of vehicles passing a particular point along a route, such as urban freeways versus rural arterials (Chan *et al.*, 1991). In-vehicle UFP number concentrations were strongly associated with traffic density (Kaur and Nieuwenhuijsen, 2009; Briggs *et al.*, 2008). However, in-vehicle $PM_{2.5}$ exposure levels have not been found to be significantly influenced by traffic density and road type in several empirical studies (Kaur and Nieuwenhuijsen, 2009; Adams *et al.*, 2002; Kingham *et al.*, 2011). Whether a statistically significant finding can be

observed depends in part on the variability in conditions encountered. Wind speed is important in pollutant dispersion and dilution. However, wind speed appears to have little effect on in-vehicle $PM_{2.5}$ levels (Kaur and Nieuwenhuijsen, 2009). Studies have considered the impact of ambient temperature and relative humidity on in-vehicle $PM_{2.5}$ exposure typically do not report a significant effect (Zagury *et al.*, 2000; Kaur and Nieuwenhuijsen, 2009).

Sources of Inter-Vehicle Variability

Zhu *et al.* indicate that vehicle age might be an important factor affecting the I/O ratio (Zhu *et al.*, 2007). However, they measured only three vehicles and thus could not produce a statistically significant finding. Knibbs *et al.* conducted ACH measurements on six vehicles (Knibbs *et al.*, 2009). Airflow infiltration was found to be partly related with vehicle age. Deteriorating HVAC duct damps and seals on windows and doors may lead to higher ACH.

Prioritizing Factors for Study Design.

The priority here is to focus on plausible combinations of key sources of intra-vehicle variability and to demonstrate a method for data collection based on an intensive set of field measurements collected with one vehicle. The methodology can easily be extended later to additional vehicles to address inter-vehicle variability.

The key factors taken into account include air, window status, fan setting, AC utilization, and selection of routes, as shown in Table IV-1. There are eight cases based on the HVAC air source of fresh air, and eight cases based on recirculating air. Because prior

studies have established that any amount of window opening substantially increases ACH and I/O, only one open window case is considered for each air source. The fan level is varied among four settings, including off, low, second-highest, and high. There are comparison cases for AC off and AC on.

Table IV-1. Details of the Field Study Design for Air Source, Window Position, Fan Level, AC setting and Route

Case	Air Source	Window	Fan Level	AC	Route	Road Type[a]
1-1	fresh air	passenger window open 3"	0	off	A-out	NF
1-2	fresh air	closed	0	off	A-in	NF
1-3	fresh air	closed	1	off	1-out	10%NF / 90%F
1-4	fresh air	closed	1	on	1-in	90%F / 10%NF
1-5	fresh air	closed	3	off	C-out	half NF/half F
1-6	fresh air	closed	3	on	C-in	half F / half NF
1-7	fresh air	closed	4	off	3-out	NF
1-8	fresh air	closed	4	on	3-in	NF
2-1	recirculation	passenger window fully open	0	off	A-out	NF
2-2	recirculation	closed	0	off	A-in	NF
2-3	recirculation	closed	1	off	1-out	10%NF / 90%F
2-4	recirculation	closed	1	on	1-in	90%F / 10%NF
2-5	recirculation	closed	3	off	C-out	half NF/half F
2-6	recirculation	closed	3	on	C-in	half F / half NF
2-7	recirculation	closed	4	off	3-out	NF
2-8	recirculation	closed	4	on	3-in	NF

a. "NF" refers to non-freeway (local road), "F" refers to freeway (highway).

Eight one-way routes were selected to observe variation in driving speed and traffic conditions, as shown in Figure IV-1. These routes are between typical commuting origin and destination (O/D) pairs of NC State to north Raleigh, and north Raleigh to Research Triangle Park (RTP). The routes include varying mixes of freeway and non-freeway road types as

indicated in Table IV-1. Each route has directional traffic flow, enabling data collection under congested and uncongested conditions in a given direction depending on time of day.

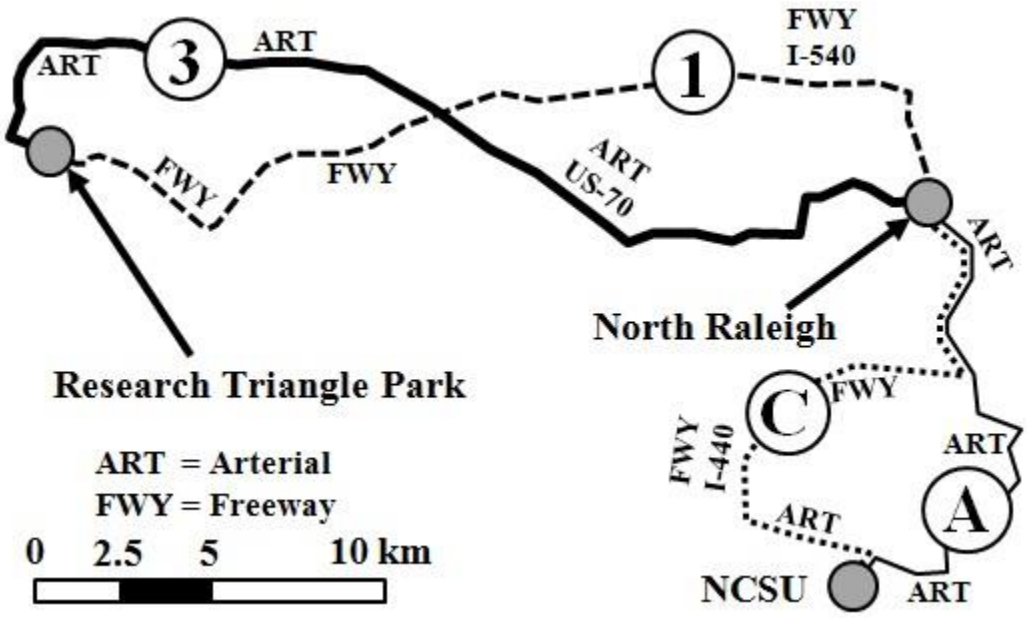


Figure IV-1. Study Area Map

The study design shown in Table IV-1 was repeated three times. It took approximately 3 to 4 hours to drive the 110 miles of the selected routes. Late morning to mid-day off-peak and afternoon peak traffic flow periods were sampled. For fresh air cases, data were collected from 9 am to 1 pm on Tuesday, July 3, and from 3 pm to 7 pm on Thursday, June 28 and Friday, June 29. For recirculation cases, data were collected from 9 am to 1 pm on Thursday, July 5, and from 3 pm to 7 pm on Wednesday, June 27 and Tuesday, July 3. The vehicle used for data collection was a two-door 2008 Mitsubishi Eclipse coupe with AC and cabin air filter.

4.2.3 Data Quality Assurance and Analysis of Results

Quality assurance includes instrument comparison, synchronization of data, and range and consistency checks. No outlier or inconsistent data was found.

To enable comparisons between the two DustTrak monitors, a size specific comparison factor was developed for each particle size range (PM_1, $PM_{2.5}$, PM_4, and PM_{10}) prior to field data collection. The two monitors were placed side by side in various microenvironments, including outdoors, in-vehicle, a kitchen while cooking, and a bedroom. Simultaneous one minute average measurements were conducted for at least one hour in each microenvironment. Paired sample T-tests were performed to determine if the average difference of concentrations between the monitors was significantly different from zero. The comparison factor for each size range was developed based on linear regression. The monitor with the more recent manufacture date was selected as the reference. Readings from the second monitor were corrected to the reference monitor using the comparison factor.

For each replicate of each case given in Table IV-1, average C_{NV}, C_{IV}, and I/O were calculated for each size range based on concentrations averaged over each sampling period. Pearson correlation was used to assess the linear dependence between C_{IV} and C_{NV} within each case. Means and 95% confidence intervals were reported.

4.3 Results

This section presents results for: (1) the monitor comparison factors; (2) comparisons of C_{NV} and C_{IV} concentrations; and (3) comparisons of I/O.

4.3.1 Comparison Factors

Figure IV-2 shows a parity plot of $PM_{2.5}$ concentrations measured in several microenvironments using the two monitors. Measured concentrations vary widely from 15 to 110 μg/m^3. The slope of the best fit line in Figure IV-2 is 0.973 with a 95% confidence interval of 0.971 to 0.975. Thus, the slope is significantly different from, but very close to, one. The high coefficient of determination (R^2= 0.993) indicates that the two monitors produce consistent and repeatable readings. The slope of 0.973 is used as the comparison factor to adjust the readings of one monitor for comparison to the other. The size-specific comparison factors for PM_1, PM_4, and PM_{10} are 0.971, 0.966, and 0.976, respectively.

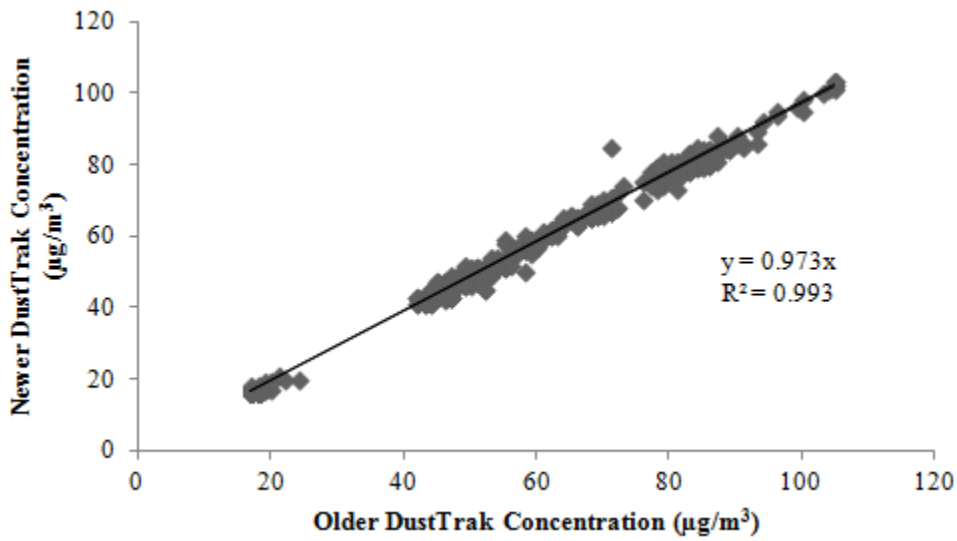


Figure IV-2. Comparison of $PM_{2.5}$ Measurements between Two DustTrak DRX 8533 Monitors, n=600

For cases with very high air exchange rate, we expect the readings for both DustTraks to be identical. If outside air pressure had unduly influenced measured concentration, the

average ratio of in-vehicle to outside (I/O) concentration would substantially differ from 1 in these cases. However, we observed that for cases with an open window and fresh air intake, the I/O ratio was 0.98 ± 0.01, and with window fully open and recirculated air, the I/O ratio was 0.97 ± 0.03. These observed ratios are very close to 1, as expected. In supplemental measurements with the external probe inlet oriented in the upwind direction, the average I/O ratio was 1.04 ± 0.05 with window-opening and fresh air intake setting. This is not statistically different from the expected I/O ratio of 1. Thus, there is no evidence of bias associated with outdoor air sampling.

4.3.2 Temporal Trends

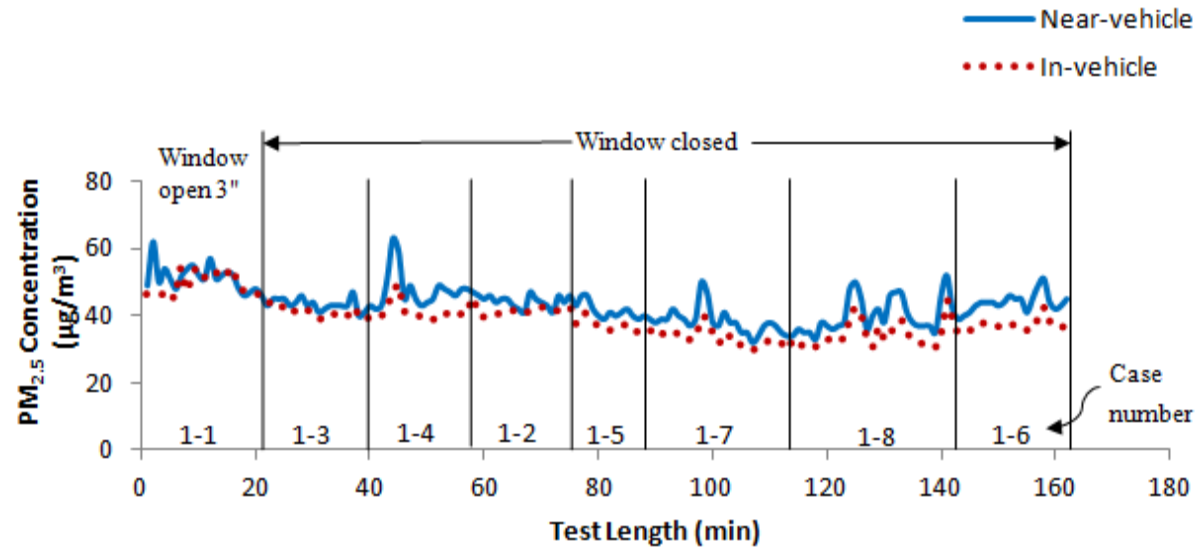
Examples of the field measurements of C_{NV} and C_{IV} are illustrated in Figure IV-3 for $PM_{2.5}$ based on data collected on Jun 29 for fresh air cases and July 3 for recirculation cases, both in the afternoon. During the early afternoon on both days, C_{NV} typically ranged from 40 to 60 $\mu g/m^3$. At about 5 pm and later, concurrently with Cases 1-8 and 1-6, C_{NV} increases on average on June 29 and for Cases 2-8 and 2-6 ranged from 40 to 100 $\mu g/m^3$ on July 3. The latter is associated with a mix of lighter workday but heavier holiday traffic. Thus, time of day is associated with differences in C_{NV}, and time of day is a reasonable surrogate for prevailing traffic flow conditions. The highest C_{NV} occurred during travel from RTP to north Raleigh in Case 2-8 along route 3-in. This route includes highly congested signalized multilane divided arterials, which may account for the high concentration observed at approximately 140 to 150 minutes in Figure IV-3(b). The short duration peaks in C_{NV} are associated with stop and go traffic in queues at red phase traffic signals.

C_{IV} is similar to C_{NV} for situations in which a window was partially or fully opened (Cases 1-1 and 2-1, respectively). When fresh air was used, C_{IV} was similar to C_{NV} to within about 10% in most cases, as illustrated in Figure IV-3(a). Peaks in C_{IV} were typically somewhat smaller in magnitude and delayed by about one minute compared to those for C_{NV}, as indicated at approximately 45, 100, 125, 140, and 155 minutes.

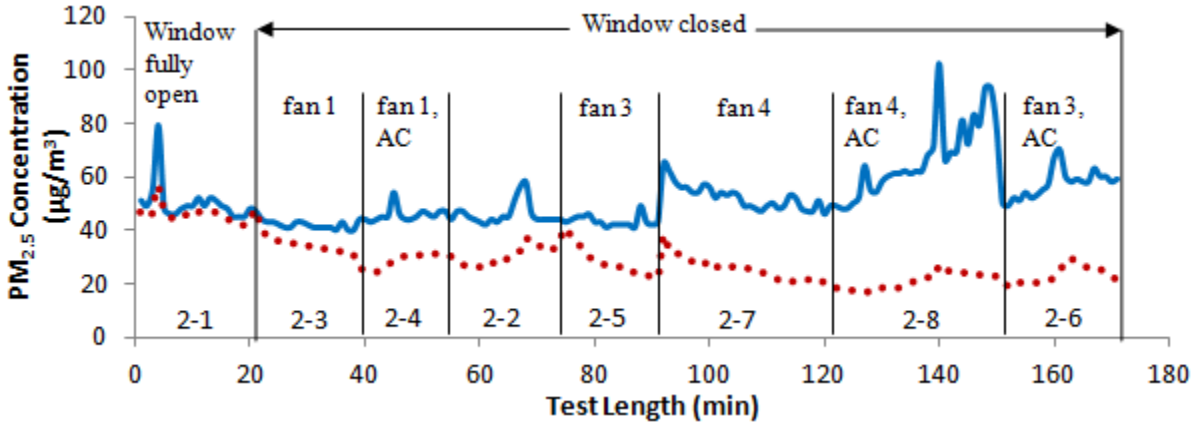
When recirculation was used with closed windows, C_{IV} differed substantially from C_{NV}, as shown in Figure IV-3(b). The largest differences between the concentrations at high fan setting (Case 2-7) or combinations of high fan setting and air conditioning (Cases 2-6 and 2-8). Large short duration peaks in C_{NV} had little effect on C_{IV}.

4.3.3 Variation in Near-Vehicle Concentration: Time of Day and Traffic

The three-day averages of daily average C_{NV} are shown in Table IV-2 for each of the 16 cases. The standard deviations quantify variability in daily averages among the three measurement days. All data were collected during a period of record high temperatures in the Research Triangle Park, NC area: the average ambient temperature was 94 ^{o}F as recorded at RDU airport (which is near several of the routes), with an hourly standard deviation of 4 ^{o}F. Thus, the ambient temperatures were approximately similar throughout data collection. The in-vehicle temperature averaged 99 ^{o}F, with hourly variability of approximately 7 ^{o}F. Thus, the range of variation in in-vehicle temperature is not large and is not likely to affect particle formation, agglomeration, or removal processes. The vehicle was driven at the speed limit or with prevailing traffic. The average speed for travel during Cases 1-3, 1-4, 2-3, and 2-4



(a) Fresh air cases (1-1 through 1-8)



(b) Recirculation cases (2-1 through 2-8)

Figure IV-3. Example Comparisons of Simultaneous Measurements for Near-Vehicle and In-Vehicle $PM_{2.5}$ Concentrations with Different Sources of Forced Air, on Afternoons of June 29 for (a) and July 3 for (b)

ranged from 47 to 55 mph on routes comprised primarily of interstate roads. Case 2-8 had an average speed of only 28 mph, which was influenced by congested traffic in the afternoon.

Table IV-2. Average Near-Vehicle and In-Vehicle $PM_{2.5}$ Concentration ($\mu g/m^3$) ± Standard Deviation ($\mu g/m^3$), Average I/O Ratio ± 95% Confidence Interval by Route and Ventilation Condition

Case	Air Source[a]	Window[b]	Fan[c]	AC	Route	Road Type[d]	Average Speed (mph)[e]	Near-Vehicle Concentration C_{NV} ($\mu g/m^3$)	In-Vehicle Concentration C_{IV} ($\mu g/m^3$)	I/O Ratio (C_{IV}/C_{NV})
1-1	F	3"	0	off	A-out	NF	25	52 ± 1.2	51 ± 1.7	0.98 ± 0.010
1-2	F	closed	0	off	A-in	NF	37	47 ± 3.7	44 ± 3.6	0.94 ± 0.001
1-3	F	closed	1	off	1-out	10%NF / 90%F	50	46 ± 3.2	43 ± 2.8	0.93 ± 0.007
1-4	F	closed	1	on	1-in	90%F / 10%NF	47	47 ± 0.5	42 ± 1.2	0.89 ± 0.044
1-5	F	closed	3	off	C-out	half NF / half F	39	45 ± 4.7	41 ± 4.3	0.91 ± 0.002
1-6	F	closed	3	on	C-in	half F / half NF	30	46 ± 2.9	40 ± 3.7	0.87 ± 0.037
1-7	F	closed	4	off	3-out	NF	41	41 ± 4.1	37 ± 4.4	0.90 ± 0.020
1-8	F	closed	4	on	3-in	NF	31	44 ± 5.4	38 ± 4.8	0.86 ± 0.002
2-1	R	fully	0	off	A-out	NF	26	45 ± 7.2	42 ± 4.9	0.93 ± 0.028
2-2	R	closed	0	off	A-in	NF	32	43 ± 4.9	28 ± 3.3	0.65 ± 0.003
2-3	R	closed	1	off	1-out	10%NF / 90%F	52	40 ± 2.9	32 ± 3.5	0.80 ± 0.033
2-4	R	closed	1	on	1-in	90%F / 10%NF	55	43 ± 4.2	28 ± 0.7	0.65 ± 0.035
2-5	R	closed	3	off	C-out	half NF / half F	36	42 ± 1.7	27 ± 1.9	0.64 ± 0.012
2-6	R	closed	3	on	C-in	half F / half NF	30	67 ± 14.1	21 ± 2.6	0.31 ± 0.082
2-7	R	closed	4	off	3-out	NF	33	45 ± 10.4	21 ± 6.1	0.47 ± 0.031
2-8	R	closed	4	on	3-in	NF	28	63 ± 4.3	19 ± 2.7	0.30 ± 0.024

a. Air source of forced air includes fresh air (F) and recirculation (R).
b. Window position includes passenger window open 3 inch (3”), passenger window fully open (fully), and closed.
c. Fan setting is indicated by levels: off (0), lowest (1), second-highest (3), and highest (4).
d. Road type includes non-freeway (NF) and freeway (F).
e. Estimated based on average ratio of route length divided by the time spent in the route.

C_{NV} varied significantly among cases. The highest case average C_{NV} of 79 μg/m^3 was 98% higher than the lowest of 40 μg/m^3. Variations in C_{NV} result from differences in traffic, as inferred from the vehicle turning movement counts (TMCs) data from NC Department of Trasnportation (DOT) at major signalized intersections at different times of day. Cases 2-6 and 2-8 had substantially higher average C_{NV} than other cases. These routes are highly congested during afternoon rush hour, leading to high on-road concentrations. The concentrations on these same routes in Cases 1-6 and 1-8 were not as high because the traffic was less congested compared to those of cases 2-6 and 2-8.

4.3.4 Variation in In-Vehicle Concentration: Windows, Recirculation, and Fresh Air

C_{IV} varied with respect to different ventilation conditions. As expected, for cases with open windows, C_{IV} was similar to C_{NV}. For cases with fresh air intake and windows closed, average C_{IV} varied within a relatively narrow range of 37 to 44 μg/m^3, compared to C_{NV} ranging from 41 to 47 μg/m^3. For cases with recirculating air and windows closed, average C_{IV} varied substantially, from 19 to 32 μg/m^3. The corresponding range of C_{NV} was 40 to 79 μg/m^3.

Fan Speed

The role of fan speed in affecting infiltration of ambient air was assessed by estimating the linear correlation between minute by minute C_{IV} and C_{NV} for each fan setting and each measurement day. For Cases 1-3 and 1-4 that used fresh air and the lowest fan setting, the correlation varied from 0.55 to 0.59 among the three measurement days. For the fresh air

second highest fan setting cases, the correlation varied from 0.71 to 0.89, and for the highest fan setting varied from 0.90 to 0.93. As expected, the trend of higher correlation with higher fan settings indicates an influential role in mixing between the ambient and in-vehicle microenvironments. When using recirculated air, increasing fan setting is not strongly correlated with minute-by-minute covariation of C_{IV} and C_{NV}, but it is associated with a clear decreasing trend in the average C_{IV}.

Air Conditioning

AC operation did not substantially affect C_{IV} for the cases with fresh air intake and closed windows. However, it made a substantial difference for situations with recirculating air. For example, for Cases 2-3 and 2-4 with closed windows and a low fan setting, C_{IV} with AC off was only 8 $\mu g/m^3$ lower than C_{NV}, but was 15 $\mu g/m^3$ lower when the AC was on. As the fan setting increased, the differences became more pronounced. For example, for Cases 2-7 and 2-8, at the highest fan setting, the concentration difference was 24 $\mu g/m^3$ with AC off and 60 $\mu g/m^3$ with AC on.

Passive Ventilation

Case 1-2 is the only case that focuses on passive ventilation. Even though windows were closed, C_{IV} was only 3 $\mu g/m^3$ less than C_{NV}. The average vehicle speed was 37 mph. The estimated passive ventilation ACH based on Equation (2) is about 20 hr^{-1}. This is a high value of ACH that would lead to a large I/O ratio.

4.3.5 Variability in the Average In-Vehicle to Near-Vehicle $PM_{2.5}$ Concentration (I/O) Ratio

Average $PM_{2.5}$ I/O ratios for different cases, along with 95% confidence intervals, are shown in Table IV-2. For cases with high advection of outside air, either by window-opening or fresh air ventilation, the average I/O ratios ranged from 0.86 to 0.98. The passive ventilation case also had a high I/O ratio (0.94). With windows closed and recirculation, the average ratio ranged from 0.30 to 0.80, depending on window position, fan level and AC setting. Venting by fresh air resulted in a 49% average higher concentration ratio compared to recirculation.

I/O ratios were found to have an inverse relationship with fan level. However, the recirculation cases were more sensitive to fan level than the fresh air cases. For fan levels increasing from 1 to 4, the average I/O ratios for the recirculation cases decreased by 49%, in comparison with an average decrease of only 4% for fresh air cases.

Similarly, the recirculation cases were more affected by AC utilization than the fresh air cases. When controlling for other factors, for recirculation cases there was an average 30% decrease in I/O ratios for AC on versus off, versus an average decrease of only 4% for the fresh air cases.

Although day-to-day variability in traffic congestion led to variations in C_{NV}, the day-to-day I/O ratios were relatively constant. On July 3, traffic was highly congested during cases 2-7 and 2-8, with an average C_{NV} of 70 $\mu g/m^3$. In contrast, on June 27, traffic was much lighter, with an average C_{NV} that was 34% lower at 46 $\mu g/m^3$. The congested versus moderate traffic I/O ratios were 0.48 and 0.45, respectively, for Case 2-7 and 0.31 and 0.29,

respectively, for Case 2-8. Thus, the I/O ratios varied much less, on a relative basis, than C_{NV}.

Table IV-3 summarizes case average I/O ratios for particle sizes ranging from PM_1 to PM_{10}. On average, the ratios are approximately the same for PM_1 and $PM_{2.5}$ for each case, and decrease slightly with larger particle size ranges. However, the relative decrease in the ratios with increasing particle size is only about 1 to 2 percent on a relative basis. Thus, while the qualitative trend is consistent with expectations that the I/O ratio decreases as particle size increases, the quantitative trend is weak. On average, the I/O ratios for the recirculation versus fresh air cases, excluding open window cases, are 37 to 38 percent lower among the size ranges.

4.3.6 Benchmarking the $PM_{2.5}$ I/O Ratios

The distributions of average $PM_{2.5}$ concentration ratios from 0.30 to 0.98 for different ventilation conditions were generally comparable to the range of 0.43 to 0.99 predicted by Liu and Frey for approximately similar cases (Liu and Frey, 2011).

Table IV-3. Average I/O Ratios of In-Vehicle to Near-Vehicle Concentrations for Selected Ventilation and Route Cases for Particle Sizes Ranging from PM_1 to PM_{10}

Case	Air Source[a]	Window[b]	Fan Level[c]	AC	Route	Road Type[d]	Average I/O Ratio			
							PM_1	$PM_{2.5}$	PM_4	PM_{10}
1-1	F	3"	0	off	A-out	NF	0.97	0.98	0.97	0.99
1-2	F	closed	0	off	A-in	NF	0.94	0.94	0.93	0.95
1-3	F	closed	1	off	1-out	10%NF / 90%F	0.93	0.93	0.92	0.92
1-4	F	closed	1	on	1-in	90%F / 10%NF	0.89	0.89	0.88	0.87
1-5	F	closed	3	off	C-out	half NF / half F	0.91	0.91	0.89	0.88
1-6	F	closed	3	on	C-in	half F / half NF	0.87	0.87	0.86	0.84
1-7	F	closed	4	off	3-out	NF	0.90	0.90	0.89	0.88
1-8	F	closed	4	on	3-in	NF	0.87	0.86	0.86	0.84
2-1	R	fully	0	off	A-out	NF	0.96	0.93	0.93	0.93
2-2	R	closed	0	off	A-in	NF	0.66	0.65	0.65	0.68
2-3	R	closed	1	off	1-out	10%NF / 90%F	0.81	0.80	0.80	0.80
2-4	R	closed	1	on	1-in	90%F / 10%NF	0.67	0.65	0.66	0.66
2-5	R	closed	3	off	C-out	half NF / half F	0.65	0.64	0.64	0.64
2-6	R	closed	3	on	C-in	half F / half NF	0.32	0.31	0.31	0.31
2-7	R	closed	4	off	3-out	NF	0.47	0.47	0.46	0.46

a. Air source of forced air includes fresh air (F) and recirculation (R).
b. Window position includes passenger window open 3 inch (3"), passenger window fully open (fully), and closed.
c. Fan setting is indicated by levels: off (0), lowest (1), second-highest (3), and highest (4).
d. Road type includes non-freeway (NF) and freeway (F).

4.4 Conclusions

A method for simultaneous measurement of PM concentrations in various size ranges for in-vehicle and near-vehicle ambient microenvironments was demonstrated. There is no evidence of bias in the outside air measurement due to probe inlet orientation. The key result from the measurements is the I/O ratio, which can be used as the slope in a mechanistic-based approach to estimate PM exposure for the in-vehicle microenvironment, as detailed in Liu and Frey (2011). The I/O ratio is highly sensitive to whether windows are open or, for closed windows, to whether fresh or recirculating air is used. For recirculating air cases, the

fan setting and AC utilization are also important. Passive ventilation is important when the vehicle is operating at road speeds with the fresh air intake setting, even if windows are closed and the fan is off.

C_{NV} is influenced by on-road traffic, which in turn depends on road type, route, travel direction, and time of day. C_{IV} is most sensitive to ventilation conditions, such as window position and the source of forced air. For recirculating air, C_{IV} is also sensitive to the HVAC fan setting and whether the AC is on.

The I/O ratio is not sensitive to traffic, as time of day was used as a surrogate for traffic volume based on vehicle turning movement counts data. Differences in the ratio between particle size ranges were found to be small. However, whether this is an artifact of the instrument sampling and analysis method, the local particle size distribution, or a valid finding would require further research.

Three replicate measurements were made for each case. The 95% confidence intervals for the concentration ratio indicated that measurement results are repeatable and robust for each ventilation case studied. In particular, the day-to-day variability in the I/O ratios for any given case and particle size was much smaller on a relative basis than the variability in the constituent concentration measurements. Thus, the I/O ratio appears to be approximately independent of the magnitude of the C_{NV}.

The study design focused specifically on sources of intra-vehicle variability as a result of controllable or observable conditions that affect C_{NV}, C_{IV}, and the I/O ratio while a vehicle operates on one more paths. There are also factors that might explain inter-vehicle variability, such the effect of vehicle age on the integrity of seals around dampers, doors, and

windows. There is not much evidence yet that vehicle size within the range of passenger car sizes would have a significant effect on the I/O ratio, but factors such as these can easily be explored by extending the study design to include additional vehicles.

4.5 Acknowledgements

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PART V COMPARISON OF FINE PARTICULATE MATTER EXPOSURE CONCENTRATIONS FOR SELECTED TRANSPORTATION MODES

Abstract

Daily commutes may contribute disproportionately to overall daily exposure to urban air pollutants such as fine particulate matter ($PM_{2.5}$). $PM_{2.5}$ concentrations were measured and compared across pedestrian, bus, and car modes during lunchtime and afternoon rush hour within a three-week time period on pre-selected round trip routes in Raleigh, NC. Variability in the transportation mode concentration ratios of $PM_{2.5}$ was quantified and factors affecting variability in concentrations were identified. Transportation mode exposure concentrations are sensitive to mode, and are affected by factors such as vehicle ventilation and proximity to on-road emission sources. In general, pedestrian and bus modes had higher $PM_{2.5}$ concentrations than car mode. Near-road pedestrian $PM_{2.5}$ concentrations generally co-varied with FSM measurements. Field studies such as this are needed to develop data for input to population-based stochastic exposure simulation models to more accurately predict transportation mode exposure concentrations.

5.1 Introduction

Air pollutants emitted from vehicle exhaust, such as fine particulate matter ($PM_{2.5}$) can cause adverse health effects, including cardiovascular and respiratory diseases and mortality (U.S. EPA, 2009a). In the U.S., the average one-way daily commuting travel time is 25.5 minutes, and 86% of trips to work are via personal vehicle (McKenzie and Rapino, 2011). The transport microenvironment is proximate to higher on-road traffic emissions than other microenvironments (Adams *et al.*, 2001; Chan *et al.* 2002; Gulliver and Briggs, 2004). Personal transport includes modes such as personal car, referred to as 'in-vehicle,' transit bus, pedestrian, and others.

Ambient $PM_{2.5}$ concentration is influenced by direct emissions from on-road mobile sources such as cars and trucks, and from secondary formation based on precursor emissions such as sulfur dioxide, nitrogen oxides, volatile organic compounds and ammonia (Gertler *et al.*, 2000). In the US, mobile sources contribute approximately 14% of primary $PM_{2.5}$ emissions and 65% of primary NO_x emissions (Rao *et al.*, 2008).

Measured ambient pollutant concentrations from fixed site monitors (FSMs) are typically used as surrogates for personal exposure in epidemiologic studies (U.S. EPA, 2009a). In addition, for population-based exposure models such as the Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM) model and the Air Pollution Exposure (APEX) model, in-vehicle concentrations are estimated based on a ratio of in-vehicle to FSM or area-wide concentration (Burke and Vedamtham, 2009). APEX also includes estimation of near-road concentrations using a scaling factor based on FSM or area-wide concentration data (Glen *et al.*, 2012). However, FSMs can be far from a particular

roadway and may not be a good indicator of on-road or near-road air quality. FSMs may have little association with individual exposure (Kulkarni and Patil, 1999; Chan *et al.*, 2000). Additional factors, such as personal time-activity patterns, choice of transportation mode, and proximity to the pollutant source affect transportation exposure (Kaur *et al.*, 2007).

Exposure concentration is a concentration with which a person comes into contact (U.S. EPA, 1992). Typically, the largest percentage of time in which an individual is exposed to $PM_{2.5}$ of ambient origin occurs indoors but the highest ambient $PM_{2.5}$ exposure concentrations occur in transportation microenvironments (U.S. EPA, 2009a). Some studies assessed exposure concentrations for three or more transportation modes, but either these modes were not measured at similar time, sample size was limited, or the study was not conducted in the U.S. (Kingham *et al.*, 1998; Morabia *et al.*, 2009; de Nazelle *et al.*, 2012). There is need for comparison of exposure concentrations for multiple transportation modes with more samples based on measurements conducted under approximately similar conditions for factors such as meteorology, vehicle fleet composition, and routes.

The objectives here are to: (1) measure $PM_{2.5}$ concentrations across transportation modes; (2) determine factors affecting these exposure concentrations; and (3) evaluate whether an FSM is an appropriate surrogate for near-road concentration.

5.2 Methodology

The methodology includes: (1) study design for field data collection; (2) preparation of instruments; and (3) data quality assurance, and analysis.

5.2.1 Study Design

The study design is developed based on the most significant controllable or observable factors affecting exposure in transportation microenvironments.

Factors Affecting Variability in Transportation Exposure Concentration

Comparisons of transportation exposure concentration across studies are difficult because of the variety of study designs, which differ with respect to study area, transportation modes, choice of routes, vehicle ventilation settings, monitoring instruments used, and weather conditions (de Nazelle *et al.*, 2012; Kaur and Nieuwenhuijsen, 2009; Chan *et al.*, 1999).

Transportation Mode Typical personal transportation modes in the U.S. include cars, bus, and walking, comprising more than 90% of total trips (U.S. DOT, 2013). Modes closest to traffic emission sources experience the highest concentrations of traffic-related pollutants such as $PM_{2.5}$ (de Nazelle *et al.*, 2012; Karner *et al.*, 2010).

In-vehicle $PM_{2.5}$ exposure concentrations are typically comparable to or less than that of pedestrians walking on the same route (Briggs *et al.*, 2008; Gulliver and Briggs, 2007). Jiao and Frey (2013) found that, under different ventilation conditions, the in-vehicle to outside-vehicle $PM_{2.5}$ concentration (I/O) ratio for a sample car varied from 0.30 to 0.98. The lower ratios were based on closed windows and use of recirculating air. In-bus concentration is influenced by door opening at bus stops, which may introduce outside air, and passenger movement inside the bus, which combined with air movement may lead to re-suspension of previously deposited particles (Huang and Hsu, 2009).

Vehicle age might affect in-vehicle $PM_{2.5}$ concentrations. Deteriorating HVAC duct dampers and seals on windows and doors may lead to higher airflow infiltration (Zhu *et al.*, 2007).

Pedestrians using roadside sidewalks may experience peaks in concentrations associated with waiting at a traffic light, proximity to a construction area, or proximity to other pedestrians who are smoking (Kaur *et al.*, 2005). Pedestrian proximity to a road influences the concentration encountered (Buonanno *et al.*, 2011).

Traffic Volume Traffic volume can affect transportation PM exposure (de Nazelle *et al.*, 2012; Briggs *et al.*, 2008; Chan *et al.*, 1991; Buonanno *et al.*, 2011; Koushiki *et al.*, 1992; Brunekreef *et al.*, 2003). Carslaw *et al.* (2007) indicate that light-duty vehicle count is a more important determinant of CO concentration than heavy-duty vehicle count, because gasoline-powered vehicles emit more CO. In contrast, diesel vehicles are a more significant contributor to primary $PM_{2.5}$ concentrations (Richmond-Bryant *et al.*, 2009).

Road and Facility Type Variations in in-vehicle concentrations may be related to lane choice, proximity to other vehicles, and speed (Chan *et al.*, 1991). PM concentrations can vary from one route to another, depending in part on traffic density and vehicle operations (Briggs *et al.*, 2008). In-vehicle CO concentrations differ between relatively congested and uncongested roads, and between tunnels, street canyons and open well-ventilated roads (Chan and Chung, 2003). For example, in-vehicle CO concentrations were higher on urban roadways with high traffic and stop signs than on rural roads (Chan *et al.*, 1991).

Meteorology Factors such as wind speed and direction, temperature, relative humidity and precipitation are often evaluated with respect to exposure concentrations (Kaur and Nieuwenhuijsen, 2009; Briggs *et al.*, 2008; Buonanno *et al.*, 2011; Kingham *et al.*, 2011; Alm *et al.*, 1999; Zagury *et al.*, 2000; Gómez-Perales *et al.*, 2004). An increase in wind speed typically results in a decrease in near-road ambient $PM_{2.5}$ concentration (Kaur *et al.*, 2007; Zagury *et al.*, 2000; Gómez-Perales *et al.*, 2004). However, wind speed may have little effect on in-vehicle $PM_{2.5}$ concentration (Kaur and Nieuwenhuijsen, 2009), which may depend more on vehicle ventilation conditions. The effect of wind direction on exposure is related to road and building configuration (Kaur *et al.*, 2007). For a wide street or flat area, pollutant concentrations dilute quickly in comparison with street canyons (Buonanno *et al.*, 2011). Variation in ambient temperature and relative humidity appears not to significantly affect transportation-related $PM_{2.5}$ concentration (Kaur and Nieuwenhuijsen, 2009; Zagury *et al.*, 2000). Singh *et al.* (2006) reported increased particle number concentrations during afternoon rush hour particularly during warmer months.

Incorporating Factors into Study Design

The key factors taken into account include selected transportation modes, vehicle ventilation status, vehicle age, time of day (surrogate for traffic volume), facility type, travel direction, and meteorological conditions (temperature, humidity, wind speed, and wind direction). Car, bus and pedestrian modes were selected for travel on a predefined urban unrestricted route with measurements taken during weekday lunchtime (11: 30 AM – 1: 30 PM) and afternoon rush hour (4 PM – 6 PM). The measurements were made at or near NC State and were

expected to contain hot spots where $PM_{2.5}$ concentration may be relatively high, such as roads with moderate to high traffic in peak traffic hours.

As shown in Figure V-1, the route includes a 4-mile roundtrip on two major arterial urban roadways in Raleigh, NC. Car and bus modes started the outbound trip at point 1 along Avent Ferry Road to point 2, and returned to point 1 followed by a short segment to point 4. The pedestrian route was selected to enable roundtrip travel in approximately 30 minutes and, thus, is shorter than the car and bus route at 1.3 miles roundtrip. The pedestrian route begins at point 1, goes to point 3, returns to point 1 and then goes to point 4. The roadways along the route differ with respect to traffic volume (Annual Average Daily Traffic counts vary from 9300 to 33000) and number of lanes (2 to 7, depending on location). The choice of time of day, route, and travel direction influence whether free-flow or congested traffic is likely to be encountered.

Data were collected during a three-week period between March 25 and April 12, 2013. Students from the CE 479/579 Air Quality class at NC State formed 11 groups to measure transportation exposure concentrations. Each group had a personal vehicle. On Apr 8, two personal vehicles were used for data collection at different times of day. Thus, the study collected valid data for 12 vehicles, ranging in model year from 2000 to 2011. With some exceptions, each group collected data for each selected transportation mode for both specified time periods in a day.

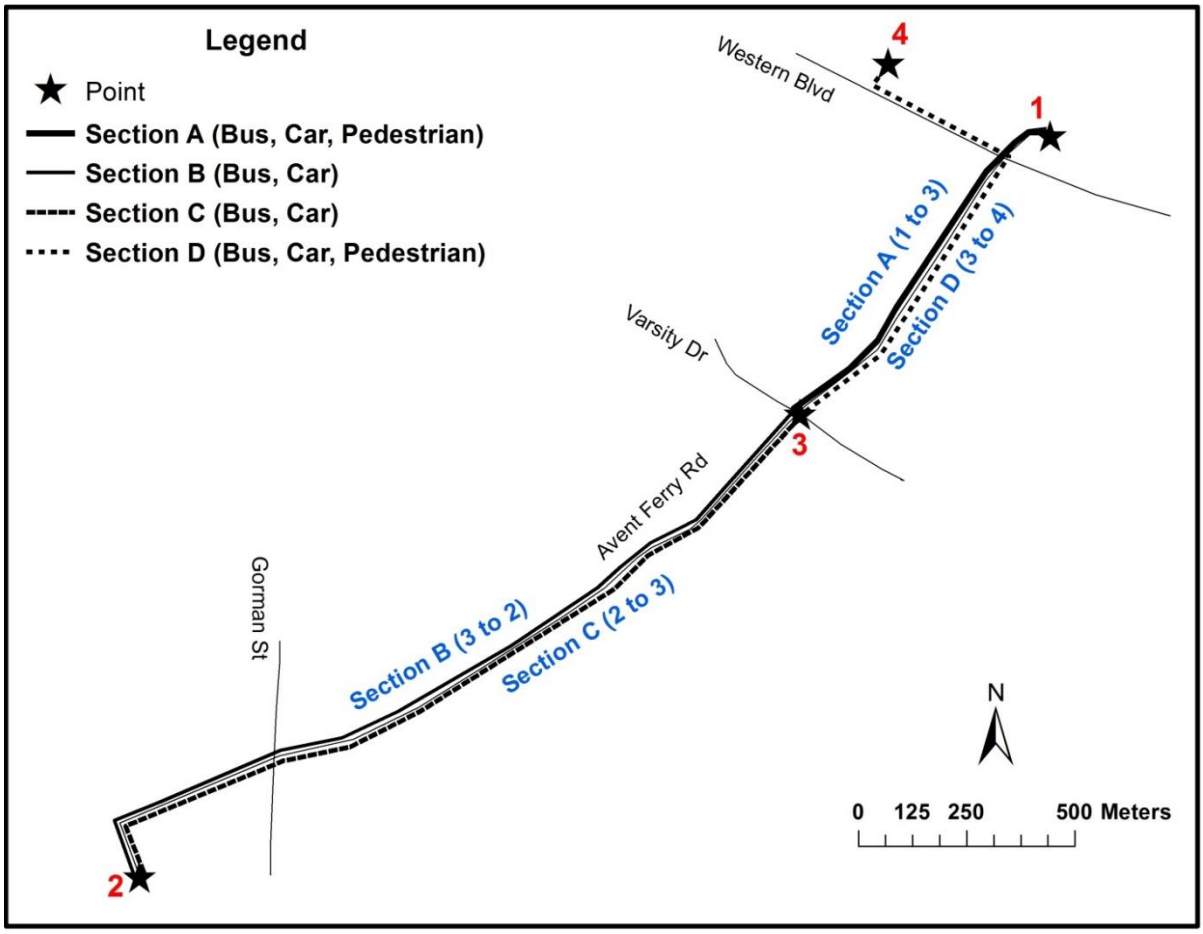

Figure V-1. Study Area and Route Map in Raleigh, NC

Logsheets were used to record events that might be related to elevated concentrations, such as: vehicle in front has visible plume or is a heavy duty diesel vehicle; a vehicle with visible plume or plume that has noticeable odor, a group of vehicles, or a noticeably old model year passenger car has passed by. To enable comparisons between data collection runs, the windows of all cars were closed and recirculating air with median fan speed (50% of maximum fan speed) was used, with one exception noted later. This combination of ventilation conditions is expected to have an average in-vehicle to near-vehicle $PM_{2.5}$

concentration (I/O) ratio of about 0.4 to 0.6 depending on air conditioning utilization (Jiao and Frey, 2013). During bus transportation, factors that may be related to in-cabin concentration were noted, such as fan or AC operation, location of the instruments in the bus, and passenger counts. Pedestrians collected data on sidewalks within 2 meters of the edge of roads. For each transportation mode, a one-minute traffic count indicating numbers of passing cars and diesel trucks was made at or in proximity to each major intersection.

5.2.2 Instruments

Several instruments were packaged into a tote bag for data collection, including a TSI DustTrak 8520 aerosol monitor, a HOBO U14 temperature and relative humidity logger, a Garmin 76CSx GPS with barometric altimeter, and a UNI-T anemometer. The DustTrak is a battery operated, light-scattering laser photometer that records real-time aerosol concentrations for a specified particle size range (Chan *et al.*, 2002; de Nazelle *et al.*, 2012). The DustTrak was factory-calibrated to the Arizona Test Dust (ISO 12103-1) (TSI, 2010), and subjected to daily zero checks during measurements. A sampling tube was attached to a strap of the sampling bag so that the inlet was near a person's breathing zone. An averaging time of ten-seconds was used for the DustTrak.

Ambient temperature and relative humidity were recorded for ten-second averages using a HOBO U14 logger. Position and elevation were recorded at 1 Hz using the GPS with barometric altimeter. Wind speed at four intersections was measured during pedestrian travel for one minute each using a UNI-T anemometer, or was quantified based on hourly average

ambient wind speed and direction data from the Lake Wheeler Road Field Lab, which is the meteorology station nearest to NC State.

To enable comparisons between pedestrian concentration and FSM data, hourly average FSM data were obtained from the NC Division of Air Quality. The Millbrook monitoring site is the closet FSM to NC State (7.7 miles in linear distance) and is the only station in Wake County that provides ambient $PM_{2.5}$ concentrations and meteorology data. Ambient $PM_{2.5}$ concentrations were measured using a Federal Reference Method (FRM)-designated Beta Attenuation Monitor (BAM) 1020. The Millbrook station is within 100 feet of a 5-lane local road with annual average daily traffic count (AADT) of 17,000, within 0.4 miles of a major arterial road with AADT of 44,000 and 1.5 miles from an interstate highway with AADT of 61,000. Near-road air quality studies generally indicate that concentrations of traffic related air pollutants decay to background within a few hundred meters from a road depending on the specific pollutant. However, $PM_{2.5}$ had smaller spatial gradients (Karner *et al.*, 2010). Thus, traffic on the 5-lane local road is expected to affect $PM_{2.5}$ concentrations at the FSM. However, the more distant major arterial and highway are expected to have less impact.

Since previous studies indicate that a DustTrak typically overestimates $PM_{2.5}$ concentrations compared to EPA FRM samplers by a factor of 2 to 3 (Kingham *et al.*, 2006; Yanosky *et al.*, 2002), correction factors were developed based on a linear regression analysis of two-day 8-hour side-by-side measurements using the DustTrak next to the Millbrook site. These corrections were used when comparing concentration measurements to the FSM.

5.2.3 Data Quality Assurance and Analysis of Results

Quality assurance includes instrument calibration, data synchronization, and range and consistency checks. All instruments were synchronized each day using the GPS clock as the reference. Data from multiple instruments were synchronized using SAS 9.3 and merged into one dataset on a 1-Hz basis. Negative concentration values were replaced with zero.

The test routes were divided into four sections to facilitate comparisons between transportation modes. Sections A (point 1 to 3) and B (point 3 to 2) represent the outbound vehicle trips whereas sections C (point 2 to 3) and D (point 3 to 4) are inbound. The pedestrian trips took place only on Sections A and D. These sections differ in traffic volume and have intersections with roads that differ with respect to numbers of through and turning lanes. Means and standard deviations of $PM_{2.5}$ concentrations for each transportation mode by section at each time of day were estimated. Since concentrations measured in the car with recirculating air setting were expected to vary the least, the car mode was used as a basis to calculate pedestrian-to-car and bus-to-car concentration ratios for each route section at each time of day. Correlations between average pedestrian concentrations with individual meteorological and traffic parameters were analyzed.

To provide insight regarding spatial variability in concentration, data visualization includes $PM_{2.5}$ concentration versus distance plots for each transportation mode by each time of day, route section, and trip direction on each day. Using pedestrian measurement as a surrogate for near-road concentration, the concordance of FSM to near-road concentrations was evaluated based on scatter plots of average pedestrian concentration and FSM data for each day and time period.

Data collection went smoothly in most cases. There are a few instances where data were not collected. An in-vehicle measurement intended for Mar 25 at lunchtime did not occur because the vehicle driver was not available. A scheduled Apr 4 measurement of pedestrian mode was cancelled due to rain. A portion of the Apr 10 afternoon bus mode data was missing because of instrument mis-operation.

5.3 Results

This section presents results for: (1) factors affecting variability in transportation mode concentrations; (2) variability in transportation mode concentration ratios; (3) comparing the instruments to the FSM; and (4) comparison of near-road pedestrian concentration and FSM data.

5.3.1 Factors Affecting Variability in Transportation Mode Concentrations

Table V-1 shows the average $PM_{2.5}$ concentrations by time of day, route section, and transportation mode. The coefficient of variation (CV) is the ratio of standard deviation divided by the mean, which quantifies the relative variability in the average when comparing across days. There are differences in average concentration by time of day and among transportation modes. Afternoon during rush hour generally had higher $PM_{2.5}$ concentrations than lunchtime.

For $PM_{2.5}$, the pedestrian mode averaged only 5% higher concentration than the bus mode, and thus pedestrian and bus concentrations were similar. However, the $PM_{2.5}$ pedestrian mode concentration averaged 42% higher compared to the car mode. Thus, the

car mode had a ratio of approximately 0.7 on average compared to the near-road concentration. The I/O ratio for the car mode is probably lower than this, since the $PM_{2.5}$ concentration over the road, in contact with the vehicle, may be higher than at roadside. Thus, the result is consistent with an expected I/O ratio of 0.4 to 0.6.

Example time plots of transportation exposure concentrations are illustrated in Figure V-2. For $PM_{2.5}$, the short-period $PM_{2.5}$ peaks observed in bus and pedestrian modes were mainly associated with congested traffic at red phase traffic signals at three major intersections, especially when there were diesel trucks nearby. $PM_{2.5}$ exposure concentrations in the car mode varied the least among the three transportation modes along the route.

The sampling period average temperature for the pedestrian mode was 67 ± 23 °F (mean $\pm$ standard deviation), and the average relative humidity was 29 ± 15 %. Wind speed varied from 0.6 to 5.9 m/s. One-minute traffic counts at three major intersections were generally 40% higher in the afternoon than at lunchtime. Table V-2 shows the Spearman (rank) correlations between average pedestrian concentrations for each of the two times of day and measured meteorological and traffic parameters, as well as correlations between corresponding FSM concentrations with meteorological parameters measured at the FSM site.

Table V-1. Summary of Average Measured $PM_{2.5}$ Concentrations by Time of Day, Route Section, and Transportation Mode

Time of Day	Route Section	Transportation Mode	N	$PM_{2.5}$ Mean ($\mu g/m^3$)	$PM_{2.5}$ CV
Lunchtime (11:30 AM – 1:30 PM)	A	Bus	11	8	0.5
		Car	10	8	0.7
		Pedestrian	10	10	0.6
	B	Bus	11	9	0.5
		Car	10	8	0.7
	C	Bus	11	9	0.5
		Car	10	7	0.6
	D	Bus	11	9	0.6
		Car	10	6	0.6
		Pedestrian	10	11	0.6
Afternoon (4:00 PM – 6:00 PM)	A	Bus	11	13	0.6
		Car	11	8	0.9
		Pedestrian	10	11	0.9
	B	Bus	11	12	0.5
		Car	11	8	1.0
	C	Bus	11	12	0.6
		Car	11	9	1.0
	D	Bus	10	11	0.6
		Car	11	8	1.1
		Pedestrian	10	12	0.9

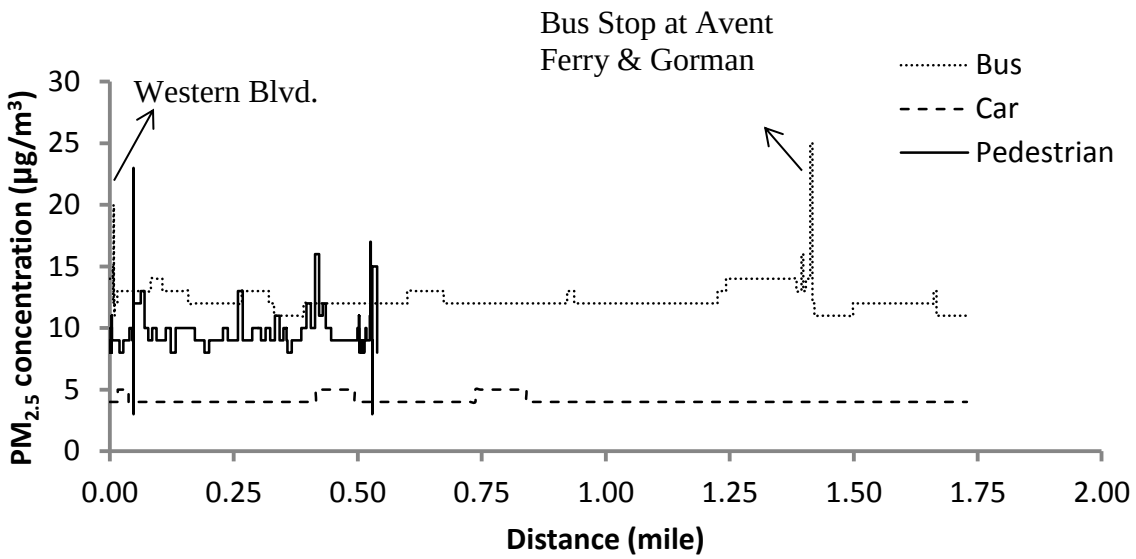

Figure V-2. Example Comparison of Distance versus Concentration for Bus, Car, and Pedestrian mode on Apr 3 Lunchtime, Outbound (Sections A and B)

Insignificant correlation was found between pedestrian $PM_{2.5}$ exposure concentrations versus wind speed and traffic counts, but there were statistically significant correlations between temperature and $PM_{2.5}$ concentration based on both the FSM and exposure measurements. In the Raleigh area, ambient $PM_{2.5}$ levels tend to be higher on warmer days, perhaps as a result of more emissions-producing activities, and with increased formation of secondary particles (Zagury *et al.*, 2000). Temperature and traffic are collinear, with a statistically significant correlation of 0.42.

Table V-2. Spearman Correlation Results[a]

(a) Between sampling period average[a] near-road pedestrian concentration by time of day (n = 20) and measured meteorology, traffic parameters (n = 20)

Parameters	Pedestrian $PM_{2.5}$ Concentration
Temperature	0.709*
Relative humidity	0.375
Wind speed	0.190
Total traffic count	0.078

[a.] Since the pedestrian concentration on Apr 4 was not available due to rain, the results were based on 10 lunchtime periods and 10 afternoon periods, of which the pedestrian sampling time within each time period was approximately 30 minutes. The correlations were based on averages over the 30 minutes for each of the 20 sampling periods.

(b) Between sampling period average[b] fixed site monitor (FSM) concentration (n=20) and FSM-based ambient meteorology parameters (n = 20)

Parameters	FSM $PM_{2.5}$ Concentration
Temperature	0.653*
Relative humidity	0.195
Wind speed	-0.156

*. Correlation is significant at the 0.05 level (2-tailed).

[b.] For each test day, hourly average FSM data that included the sampling period were selected. For the lunchtime, a sampling period average was calculated based on hourly FSM data from 11 AM to 2 PM, and an afternoon sampling period average was calculated based on hourly FSM data from 4 PM to 6 PM. The results were based on 10 lunchtime period averages and 10 afternoon period averages. FSM data on Apr 4 were not used because there was no comparable pedestrian measurement at the study site. The correlations were based on averages over 3 hours for lunchtime and 2 hours for afternoon for the total of 20 sampling periods.

5.3.2 Variability in Transportation Mode Concentration Ratios

Quantification of ratios between exposure concentrations of combinations of pairwise combinations of transportation modes provides insights regarding which modes tend to have higher concentrations, irrespective of variability in meteorology or other factors. Figure V-3 shows distributions of concentration ratios for pairwise combinations of transportation modes by time of day and route section. Both pedestrian-to-car and bus-to-car $PM_{2.5}$ concentration

ratios were expected to be one or greater, mainly because recirculating air was used for 11 out of 12 cars. Among individual days and time periods, 76% of pedestrian-to-car ratios and 72% of bus-to-car ratios were above 1. On average, over all days and time periods, the pedestrian-to-car and bus-to-car $PM_{2.5}$ ratios were 1.5 and 1.3, respectively.

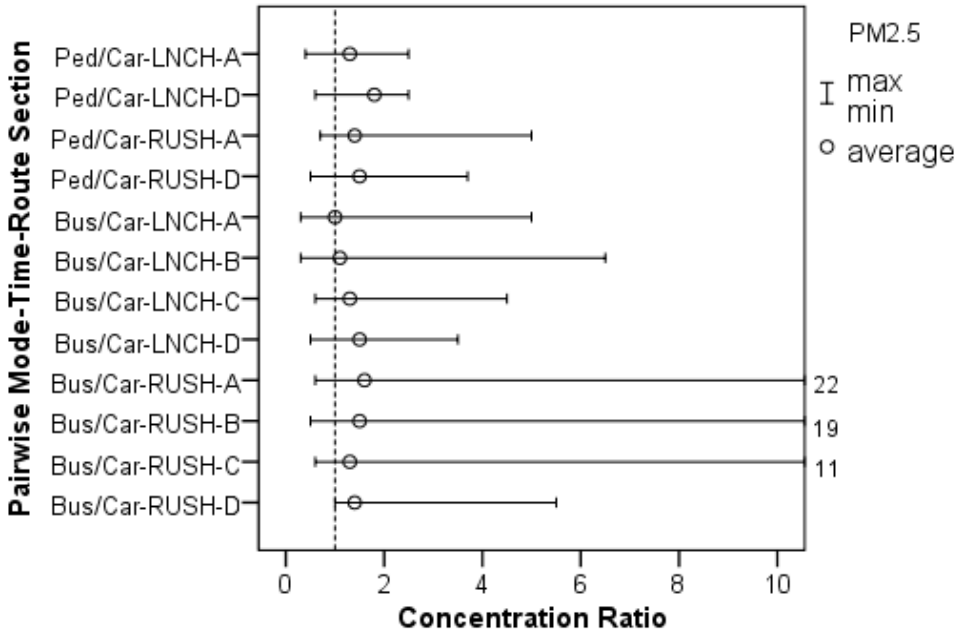

Figure V-3. Distributions of the Ratios of Average $PM_{2.5}$ Concentrations for Pairwise Comparisons of Pedestrian-to-Car and Bus-to-Car by Time of Day and Route Section ("Ped" for Pedestrian Mode; "LNCH" for Lunchtime, "RUSH" for Afternoon)

There were three cases during afternoon rush hour where the maximum bus-to-car $PM_{2.5}$ concentration ratios for a given section were very high, shown as the off-scale numerical values in Figure V-2. These three cases are from April 5, during which the in-bus concentrations were relatively high, at 11 to 22 $\mu g/m^3$ among sections A, B, and C, compared to a relatively very low in-vehicle concentration that averaged only 1 $\mu g/m^3$. Within the same period, in-bus concentrations were also 4.5 times higher than the pedestrian mode. Although

the Apr 5 afternoon traffic count was 94% higher than at lunchtime, there was no increase in the average pedestrian concentration of 5 μg/m^3 between time periods. The bus was observed to be almost full during the outbound trip. Thus, the elevated concentration might be related to passenger movements when alighting or boarding. A peak $PM_{2.5}$ concentration of 36 μg/m^3 occurred when the bus stopped to let off passengers in section A near the intersection of arterials with 7-lane and 6-lane roadways. The specific reason for the extremely low in-vehicle concentration is not clear; it might be related to in-cabin air filter efficiency.

The bus-to-pedestrian $PM_{2.5}$ concentration ratios were typically around 1, suggesting these two modes received similar concentration. In comparison, the car mode typically had the lowest exposure concentrations among the three modes. As an exception, a 2002 Jeep Wrangler was not equipped with air recirculation and had a convertible "soft-top." Thus, the doors and windows of this vehicle were more poorly sealed than for other cars. Higher in-vehicle $PM_{2.5}$ concentrations were found for this vehicle compared to the other two modes, regardless of the route section or time of day.

5.3.3 Comparison between Pedestrian Concentration and FSM

Based on side-by-side measurements with the FSM, the hourly-average DustTrak $PM_{2.5}$ concentrations were consistently higher than the corresponding hourly-average FSM concentrations. Site-specific calibration factor for DustTrak 8520 was 2.04. The FSM is located beside a middle school that has diesel school bus activity during weekdays, and the FSM concentrations might be influenced by nearby traffic on arterial roads somewhat similar

to those of the study site. Thus, the FSM was expected to have similar but possibly higher $PM_{2.5}$ concentrations than the pedestrian concentration measured in the study area.

Scatter plots in Figure V-4 compare the bias-corrected average near-road pedestrian $PM_{2.5}$ concentrations to FSM data for each day and time period. For $PM_{2.5}$, the correlation was 0.66. However, the FSM measured value of 30 $\mu g/m^3$ is unusual and may be an artifact of an unknown localized event, such as an unusual set of passing school buses or trucks. If this data point is removed, the correlation coefficient increases to 0.91. On average, the corrected near-road pedestrian $PM_{2.5}$ concentration was 43% lower than the FSM concentration.

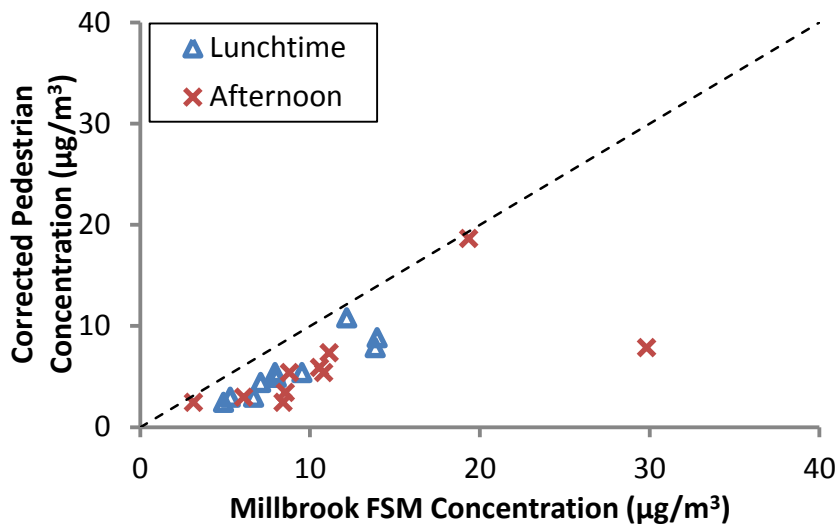

Figure V-4. Comparisons between Daily Average Near-Road Pedestrian $PM_{2.5}$ Concentration and Millbrook Ambient FSM Data

5.4 Conclusions

In general, pedestrian and bus modes had higher $PM_{2.5}$ concentrations among the measured transportation modes. Passing trucks were associated with many peaks in $PM_{2.5}$ exposure

concentration. The average in-car $PM_{2.5}$ concentration was the lowest among the modes because the selected ventilation conditions helped to prevent ingress of particles. Vehicle age might be a reason to affect in-vehicle concentrations. Measurements were not conducted simultaneously among the three transportation modes. Thus, factors such as traffic volume, wind speed and direction were not exactly the same. However, measurements for each of three modes were conducted within a one and half hour time period. The meteorological and traffic parameters were relatively stable during this time.

Whether FSM is an appropriate surrogate for personal near-road exposure concentration depends on the proximity of FSM to traffic, as well as similarity in traffic patterns. There is strong concordance between the ambient $PM_{2.5}$ concentrations measured in the study area versus the FSM, indicating that there is area wide co-variation in $PM_{2.5}$ concentrations. While both sets of measured data were obtained near arterial roads, the concordance between them implies either that both sites have similar vehicle mix and comparable traffic activity, or that the measured concentrations are more influenced by upwind sources and secondary particle formation than from near-field sources. However, there is evidence from the time plots from the study area measurements that individual peaks in transportation mode concentrations are associated with passing vehicles or local traffic activity at intersections.

Field studies such as this are needed to develop data for input to population-based stochastic exposure simulation models to more accurately predict transportation mode exposure concentrations. The field measurement results illustrate that exposure to air pollution depends on choice of transportation mode, and that the comparison of modes differs

for different pollutants. More accurate data on transportation mode exposures are needed in exposure simulation models to support assessments of the contribution of transportation activity to total exposure and to determine whether additional policies or actions are warranted to reduce such exposures. Examples might include development of information to inform individual choices of transportation mode, encouraging transit agencies to reduce exposures on buses via changes in ventilation practices, or measures that reduce tailpipe emissions especially during transient vehicle operation near intersections.

5.5 Acknowledgements

Although the research described in the article has been funded wholly or in part by the U.S. Environmental Protection Agency's STAR program through grant RD 83386301, it has not been subjected to any EPA review and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred. The Spring 2013 CE 479/579 Air Quality class conducted data collection. Rebecca Duffy in NC Department of Transportation provided traffic volume data, and Dr. Wayne Cornelius in NC Division of Air Quality provided the Millbrook ambient site data.

PART VI CONCLUSIONS

6.1 Findings

The study sets out to explore variability in human exposure to $PM_{2.5}$ from population and personal levels. Key findings are presented in this section.

6.1.1 Inter-Individual, Geographic, and Seasonal Variability in Estimated Human $PM_{2.5}$ Exposure

Using a stochastic exposure simulation model SHEDS-PM, distributions of daily $PM_{2.5}$ exposures were estimated based on ambient concentration, air exchange rate (ACH), penetration factor, deposition rate, indoor emission sources, census data, and activity diary data, and compared for selected regions and seasons.

There is substantial inter-individual variability in exposure that is not explained by ambient concentration alone, as demonstrated in the distribution of E_a/C ratios. The mean E_a/C varies by 6% to 36% among selected NC, TX and NYC domains, and 15% to 34% among four seasons, mainly as a result of regional differences in housing stock and seasonal differences in ACH. The estimated daily E_a/C ratio differs by a factor of 4 to 5 over a 95% frequency range among individuals because of differences in activity patterns, housing characteristics and related ACH. The distribution of E_a/C ratios in each area and season implies that, in general, exposure to ambient $PM_{2.5}$ is less than the ambient concentration by approximately half. High-end daily average E_a among individuals are influenced by factors other than high ambient concentration, such as ACH by location and season. E_a/C is correlated (r_p=0.5 to 0.6) with ACH, but has little correlation ($r_p \approx -0.2$) with ambient concentration C. Average levels of E_{na} also vary by area and season mainly because of

differences in ACH. For people exposed to ETS, non-ambient exposure is the dominant contributor to E_t.

6.1.2 Improvement in Estimated Indoor Residential $PM_{2.5}$ Concentration to Account for Indoor Emission Sources

By applying an indoor air quality model, RISK, indoor residential $PM_{2.5}$ exposure concentration was quantified for single- and multi-zone emission scenarios. The assumption of a well-mixed single-zone used in exposure simulation models was found to be reasonable in case of no indoor sources. However, in the presence of cooking or smoking, the assumption was not valid. An approach for applying a cooking or smoking correction factor to the original single-zone mass balance equation was demonstrated. The correction factor is most sensitive to variations in ACH. Polynomial relationships were identified for each of several housing types for cooking and smoking emission scenarios, separately. Correction factors address the difference in estimated indoor exposure concentrations between the multi- and single-zone approaches, and can be applied to the single-zone results based on relationships with ACH. In a SHEDS-PM case study, the daily average total exposure increased by 17% after applying correction factors.

6.1.3 Variability in $PM_{2.5}$ Exposure Concentrations for the Transportation Microenvironments

A method for simultaneous measurement of PM concentrations in various size ranges for in-vehicle and near-vehicle ambient microenvironments was demonstrated in Part IV. The key

result from the measurements is the in-vehicle to outside-vehicle concentration (I/O) ratio, which ranges from 0.30 to 0.98 for a single vehicle and is sensitive to whether windows are open or, for closed windows, to whether fresh or recirculating air is used. For recirculating air cases, the fan setting and AC utilization are also important.

Near-vehicle concentration (C_{NV}) is influenced by on-road traffic, which in turn depends on road type, route, travel direction, and time of day. In-vehicle concentration (C_{IV}) is highly sensitive to ventilation conditions, such as window position and the source of forced air. For recirculating air, C_{IV} is also sensitive to the heating, ventilation and air conditioning (HVAC) fan setting and whether the air conditioning (AC) is on.

The I/O ratio is less sensitive to traffic and time of day but varied with window status, source of ventilation air (fresh or recirculation), and, for cases with recirculation and closed windows, fan setting and air conditioning use. Differences in the ratio between particle size ranges were found to be small. The 95% confidence intervals for the concentration ratio based on three replicate measurements indicated that measurement results are repeatable and robust for each ventilation case studied.

With the aim of assessing the variability in commuter exposure to on-road and near-road particles, a field data collection study design was developed in Part V based on factors that may affect transportation exposure concentrations, such as transportation mode, time of day, traffic volume, meteorology, and road type. $PM_{2.5}$ concentrations were measured and compared across pedestrian, bus, and car modes during lunchtime and afternoon rush hour within a three-week time period on pre-selected round trip routes in Raleigh, NC.

Variability in the transportation mode concentration ratios of $PM_{2.5}$ was quantified and factors affecting variability were identified. The results indicate that exposure concentrations were largely influenced by the transportation mode selected. In general, pedestrian and bus modes had higher $PM_{2.5}$ concentrations among the measured transportation modes. Passing trucks were associated with many peaks in $PM_{2.5}$ exposure concentration. There is strong concordance between the ambient $PM_{2.5}$ concentrations measured in the study area versus the FSM, indicating that there is area wide co-variation in $PM_{2.5}$ concentrations.

6.2 Conclusions

This section presents key conclusions regarding inter-individual, geographic and seasonal variability in estimated human $PM_{2.5}$ exposure, improvements in estimated indoor residential exposure concentration, and variability in the transportation microenvironments exposure concentration.

6.2.1 Inter-Individual, Geographic, and Seasonal Variability in Estimated Human $PM_{2.5}$ Exposure

The E_a/C ratio varies by individual, geographic area, season, and spatial-temporal averaging times. The distribution of E_a/C ratios in each area and season implies that, in general, exposure to ambient $PM_{2.5}$ is less than the ambient concentration. On average, exposures to simulated individuals are approximately half of the ambient concentrations. These findings indicate that concentration-response functions developed in epidemiological studies using

ambient concentration as surrogate for exposure are biased when compared to exposure concentration. However, since the day-to-day variation of estimated individual daily average E_a is highly correlated ($r_p > 0.8$) with that of daily average C, even though biased, the use of ambient concentration as a surrogate for ambient exposure in epidemiology studies may still account for temporal trends in exposure.

E_a/C is well correlated with ACH, but has little correlation with ambient concentration C. ACH is related to housing type and ventilation practices used. Thus the distribution of inter-individual variability in the E_a/C ratio can be used to identify the need for providing advisory information to the public. Such information might include, for example, advice to reduce ventilation with outside air on high ambient $PM_{2.5}$ days.

Assessing population-level exposure at urban scale is particularly important for time-series epidemiologic studies, which can provide information on the relationship between health effects and community-average exposure, rather than variations in individual exposure. Region and season-specific E_a/C ratios are recommended as a factor to consider when interpreting heterogeneity in epidemiologic studies. For example, epidemiologic studies found that warmer months (spring and summer) and the eastern U.S. were typically associated with higher $PM_{2.5}$ health effects. Geographically, E_a/C ratios estimated in the study for the summer season are the highest in NYC, followed by NC and TX area. Similarly, the risk estimates associated with same-day cardiovascular admissions in the Bell *et al.* (2008) study were the highest in the Northeast region, followed by Southeast, Southwest and Northwest regions. Seasonally, E_a/C ratios estimated in the study for NYC are the highest during summer, followed by winter, spring, and fall. In comparison, Bell *et al.* (2008)

reported the lag-0 cardiovascular admission risks in the Northeast region were the highest during winter, followed by fall, spring, and summer. When compared with another study by Chang *et al.* (2012), the mortality associated with $PM_{2.5}$ exposure in NYC were the highest in summer, followed by fall, winter, and spring.

Different exposure metrics, either air monitoring data or fused CMAQ data, were used for risk estimates in previous epidemiologic studies. In addition, other risk factors such as influenza, might affect the health effect during the winter season. Thus, a direct comparison between E_a/C ratios estimated in the study and previous risk estimates are difficult to have completely consistent results. However, the estimated E_a/C ratios generally supported findings from previous epidemiologic studies for higher risk effects estimated during warmer months and in the eastern US. Heterogeneity in health effects could partly be explained by differences in exposure factors, such as ACH, individual activity patterns and home air conditioning usage. Differences in chemical composition and particle size by season and location might also contribute to additional variation in the health effects.

6.2.2 Improvement in Estimated Indoor Residential $PM_{2.5}$ Concentration to Account for Indoor Emission Sources

Part III shows the importance of indoor sources to the estimated total daily average exposure to $PM_{2.5}$. Indoor source emissions can contribute to high-end personal exposures for some individuals. Identification and appropriate quantification of these emissions for different housing types can assist in assessing upper percentiles of population exposures and identifying homes where the health risks are likely to be highest.

Based on the sensitivity analysis, the potential for reducing indoor concentrations and exposures is not only limited to control of outdoor and indoor sources. Building characteristics such as ACH and room size have a significant effect on both mean and peak exposures. The magnitude of non-ambient exposure for a given indoor source strength is inversely proportional to ACH and to interior room volume. Thus, for indoor air research and policy, the role of building characteristics in reducing indoor human exposure to $PM_{2.5}$ merits further consideration, such as size of building, efficiency of building equipment and systems, age and construction characteristics.

Persons within a house who are not engaged in emissions producing activities have the option of engaging in exposure avoidance behavior. For example, results of bounding cases considered here indicate that exposures from non-ambient sources can be reduced for persons not engaged in emissions producing activities by changing their indoor location. However, those engaged in activities such as cooking would have to reduce their emissions or increase indoor ventilation to reduce exposure.

6.2.3 Variability in $PM_{2.5}$ Exposure Concentrations for the Transportation Microenvironments

The in-vehicle to near-vehicle concentration (I/O) ratio measured in Part IV can be used as the slope in a parametric model (Equation IV-1) to more accurate estimates of PM exposure for the in-vehicle microenvironment. The 95% confidence intervals for the in-vehicle to near-vehicle concentration (I/O) ratio indicated that measurement results in Part IV are repeatable and robust for each ventilation case studied. In particular, the day-to-day

variability in the I/O ratios for any given case and particle size was much smaller on a relative basis than the variability in the constituent concentration measurements. Thus, the I/O ratio appears to be approximately independent of the magnitude of near-vehicle concentration (C_{NV}). Differences in the ratio between particle size ranges were found to be small. However, whether this is an artifact of the instrument sampling and analysis method, the local particle size distribution, or a valid finding would require further research.

The study design focused specifically on sources of intra-vehicle variability as a result of controllable or observable conditions that affect C_{NV}, C_{IV}, and the I/O ratio while a vehicle operates on one more paths. There are also factors that might explain inter-vehicle variability, such as the effect of vehicle age on the integrity of seals around dampers, doors, and windows. There is not much evidence yet that vehicle size within the range of passenger car sizes would have a significant effect on the I/O ratio, but factors such as these can be explored by extending the study design to include additional vehicles.

In Part V, the average in-car $PM_{2.5}$ concentration was the lowest among the modes because the selected ventilation conditions helped to prevent ingress of particles. Measurements were not conducted simultaneously among the three transportation modes. Thus, factors such as traffic volume, wind speed and direction were not exactly the same. However, measurements for each of three modes were conducted within a one and half hour time period and the meteorological and traffic parameters were relatively stable during this time. Whether FSM is an appropriate surrogate for personal near-road exposure concentration depends on the proximity of FSM to traffic, as well as similarity in traffic patterns. While both sets of measured data were obtained near arterial roads, the concordance

between the ambient $PM_{2.5}$ concentrations measured in the study area versus the FSM implies either that both sites have similar vehicle mix and comparable traffic activity, or that the measured concentrations are more influenced by upwind sources and secondary particle formation than from near-field sources.

While the number of scientific investigations on traffic-related air pollutants is increasing, data for exposure assessment purposes are still not abundant. There is a clear need for data to better assess air pollution exposures from traffic source. Such data will help improve understanding of the impacts of major roadways on air quality in communities, and provide key information in health studies that associated with traffic-related air pollutants.

6.3 Recommendations

This section presents recommendations for $PM_{2.5}$ exposure assessments, including future research and data needs, and policy implication and recommendations.

6.3.1 Future Research and Data Needs

Exposure assessment of $PM_{2.5}$ may need to be source specific (traffic, cigarette smoking, gas cooking), population specific (susceptible groups like children or elderly), and geographic area or season specific (because of the heterogeneity of effect estimates) to provide the basis to develop policy options and reduction strategies. The most effective way to develop and select risk reduction strategies may be to attribute exposures and associated risks to microenvironments, activities and emission sources. With the aim of reducing public health risks, the relative contributions of the sources to microenvironments and

microenvironments to exposures, and the relevance of these exposure sources to health should be evaluated.

From a scenario-based exposure modeling perspective, since people spend the majority of their daily time at home, the indoor residential microenvironment tends to be the most significant contributor to both population average and personal exposure. Among all indoor microenvironmental inputs to SHEDS-PM, air exchange rate (ACH) is the most important factor that affects inter-individual, seasonal and geographic variability in estimated population or personal exposure to $PM_{2.5}$. ACH varies by area and season, and is related with housing type and age, as well as individual ventilation practices used. Thus, for future studies estimating urban-scale or microenvironmental $PM_{2.5}$ exposure, it is important to characterize or select appropriate distributions or values of ACH that are most relevant to the ventilation practice and housing distribution of study area(s) and season(s).

Heterogeneity in the E_a/C ratio due to $PM_{2.5}$ compositional differences has not been addressed in the study because of the lack of sufficient input data. As it is highly plausible that the chemical composition may be another predictor of health effects other than particle size, studies of source apportionment on $PM_{2.5}$ exposure are needed in the future to relate adverse health effects to $PM_{2.5}$ sources and to develop effective source control strategies for reducing particle pollution health risks.

The stochastic human exposure model such as SHEDS-PM creates sequences of activities for each simulated individual by sampling human activity data from CHAD. A limitation not readily addressed by CHAD is the possible seasonal and geographic differences in activity patterns. There is insufficient data in the current CHAD from which to

quantify differences in activity pattern by gender, region, and season for the specific geographic areas. There is a need to augment CHAD to contain diaries representative of geographic areas and seasons of interest.

In addition, some issues that are not quantifiable with the current data and model might also be important for personal exposure. Correlations between certain human activities and microenvironmental concentrations are not captured in current exposure simulation models. For example, smoking or cooking may lead to an individual opening a window, which in turn affects the amount of outside air coming into the residence. Variations in concentration caused by possible urban canyon effects or pollutant transport within large buildings on a smaller spatial resolution are not adequately addressed by using area-wide input concentrations in the model.

Field studies are needed to develop data for inputs to population-based stochastic exposure simulation model such as SHEDS-PM to more accurately predict microenvironmental exposures. To be specific, it is necessary to determine the relationship between ambient outdoor and indoor $PM_{2.5}$ concentrations given the greater amount of time people spend indoors. For the indoor residence, there are relatively few data from which to estimate particle infiltration. Thus, data are needed for particle penetration (P) and deposition rate (k) that more clearly represent situations without indoor source emissions for different particle size and composition. For the in-vehicle microenvironment, there are also factors that might explain inter-vehicle variability in the in-vehicle to near-vehicle concentration ratio, such as the effect of vehicle age on the integrity of seals around dampers, doors, and

windows. Factors such as these can be explored by extending the study to include additional vehicles.

6.3.2 Policy Implications

Exposure, and not just concentration, should be considered in developing risk management strategies to reduce uncertainty in health effect estimates, and to identify highly exposed groups and possible exposure reduction strategies. The E_a/C ratio addresses an important source of uncertainty in risk estimates by characterizing one of the exposure errors due to the use of area-wide concentration to represent population $PM_{2.5}$ exposure. City- and season-specific E_a/C ratios are recommended as a factor to consider when interpreting differences in health effects stratified by areas and seasons, which illustrates regional or seasonal differences in climate and housing that may affect factors such as time spent outdoors or indoors and air conditioning usage. Further, distributions of individual daily average E_a/C ratio, especially the high end percentiles, may be combined with ambient concentrations to aid in identifying alternative 24-hour standard level in the next PM NAAQS review to provide better protection for effects associated with short-term $PM_{2.5}$ exposure. The NAAQS currently regulate PM in the ambient air only. In general, ambient exposure is a fraction of ambient concentration, and total exposure is often greater than ambient concentration because of non-ambient source emissions. Both ambient and non-ambient PM can exert adverse health effects. The health risks posed by air pollution are determined by the personal exposure of individuals and not simply by pollutant concentrations in the ambient air. Since there is some evidence of a non-linear C-R

relationship at high $PM_{2.5}$ concentrations, the appropriate quantification of non-ambient emissions within the indoor residential microenvironment has implications for evaluating and understanding the shape of the C-R curve and the potential presence of a threshold for $PM_{2.5}$ health impacts from high level exposures.

For non-ambient emissions, although smoking in public places is regulated in most states in U.S., no standards are available to regulate indoor non-ambient PM mass. EPA or other environmental regulatory authorities should inform the public regarding PM risks from non-ambient indoor generating sources as well, such as cooking and smoking. Gaseous emissions from materials or products that are associated with non-ambient exposure should be reduced as these emissions are precursors to secondary $PM_{2.5}$ formation. Individual non-ambient exposure should be mitigated or avoided by increasing room ventilation rate and intentionally avoiding contact with emission sources.

In addition, some other policy initiatives for homes may have adverse effects on indoor air. For example, while energy efficiency incentives may reduce building energy use as a result of tighter building shells, they may lead to the accumulation of indoor pollutants, if no compensatory strategies are taken, such as utilizing active ventilation. Further research is needed on how to improve ventilation efficiency within the home while minimizing the energy use. For exposure health effect studies, since indoor sources contributes largely to total $PM_{2.5}$ exposures, it may be useful to separate personal exposure into ambient and non-ambient components, and to examine the associations of health effect with ambient concentration, ambient exposure, non-ambient exposure, and total exposure, respectively, to reduce estimates uncertainty.

FSM networks are irreplaceable for monitoring ambient $PM_{2.5}$ trends in both short- and long-term. However, the proximity of the FSM to the pollutant source and the dispersion conditions determine the magnitude of difference between the concentrations experienced by individuals and those measured at FSM. It would be ideal of detailed personal exposure can be measured for everyone in a population, but clearly, this is not plausible. The most cost-effective, efficient and convenient method in the long-run appears to continue having data from FSM to represent personal exposure concentrations, but this requires a better representativeness of FSM data to account for personal ambient exposure concentrations, especially for the urban transport microenvironments. FSM networks can incorporate more FSMs to be situated in a surrounding area of traffic to better account for on-road emissions. Such data can be used to further develop, calibrate, and verify exposure assessment method and models, and contribute to the development of appropriate health protection regulations and policy.

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APPENDICES

Appendix A Supporting Information for Part II

This SI provides supplemented texts, tables and figures to further describe inter-individual, geographic, and seasonal variability in estimated population $PM_{2.5}$exposure, including additional information on background motivation of assessing $PM_{2.5}$ E_a/C ratio; justifications for inputs (ACH, P, k) selection; inter- and intra-individual variability in non-ambient exposure (E_{na}) and total exposure (E_t); discussion of seasonal variability in elderly activity patterns; discussion of autocorrelation in elderly activity patterns; discussion of potential modeling assumptions that may cause decreased variability in the exposure results; discussion of the difference in mean E_a/C ratios; other SHEDS-PM input parameters for microenvironmental $PM_{2.5}$ concentrations (Table A-1), excluding residential microenvironment; summary of 2002 average daily ambient $PM_{2.5}$ concentrations to the SHEDS-PM model input (Table A-2); summary of inter- and intra-individual coefficient of variations (CVs) for NC Domain, Spring 2002 (Table A-3); paragraphs and tables about correlations of total Exposure (E_t) by day type for NC domain, Spring 2002 (p.SI-9, p.SI-10, Table A-4), and summary of average ratio of ambient exposure to ambient concentration (E_a/C) and non-ambient exposure (E_{na}) by day type for all cases (Table A-5); correlations coefficients between daily average exposures and concentrations (Table A-6); comparison of distribution of elderly activity based on time for males and females in winter and summer (Mean $\pm$ 95% Confidence Interval) (Table A-7); cumulative distribution functions (CDFs) of the geographic and seasonal variability in the daily total exposure (E_t), non-ambient exposure (E_{na}), and ambient exposure (E_a) in NC domain, Harris County, and NYC in 2002 (Figure A-1 – Figure A-3); CDFs of the geographic and seasonal variability in the monthly average ratio of ambient exposure to ambient concentration (E_a/C) in NC domain, Harris County, and

NYC in 2002 (Figure A-4); and distributions of inter-individual (Figure A-5) and intra-individual (Figure A-6) coefficients of variation (CVs) of total exposure (E_t) for NC domain, Spring, 2002.

1. Background and Motivation for Assessing $PM_{2.5}$ E_a/C Ratio

There have been studies looking at the trend in exposure for pollutants, such as ozone, CO and SO_2, by location and season (U.S. EPA, 2012; U. S. CO, EPA, 2010b; U. S. EPA, 2008). They found that for ozone, the population average E_a/C ratios are typically 0.1 to 0.3. Higher E_a/C ratios for ozone are generally observed with increasing time spend outside, higher air exchange rate, and in seasons other than winter. For CO, the average E_a/C ratios are around 1. For SO_2, as a result of low ambient SO_2 concentrations and the limitations of passive sampling, only two studies have reported average E_a/C ratios, with values ranging from 0.08 to 0.13. Differences in the E_a/C ratio are mainly related with pollutant-specific physical or chemical removal processes. E_a/C ratios for ozone tend to be low because ozone is a highly reactive oxidant that is removed from air by surface interactions and airborne constituents (U.S. EPA, 2012). In contrast, CO is relatively inert and has a much longer lifetime in air (U.S. EPA, 2010b). SO_2 is highly soluble and thus can be removed by reactions on indoor surfaces, especially those that are moist (U. S., EPA, 2008).

None of these characteristics applies to $PM_{2.5}$. $PM_{2.5}$ is not as chemically reactive as O_2, as soluble as SO_2, nor removed as slowly as CO. Thus, the E_a/C ratio for $PM_{2.5}$ tends to be substantially different than the E_a/C ratios for these other pollutants. The main removal process for $PM_{2.5}$ is deposition. Also, unlike the gaseous pollutants, the penetration efficiency of $PM_{2.5}$ from outdoors to indoor microenvironments is less than one, because of physical processes such as impaction and interception (ISA for PM, EPA, 2009).

Ozkayanak *et al.* (2009) analyzed coupled model uncertainties in the source-to-dose modeling of human exposures to ambient $PM_{2.5}$, and concluded that exposure and dose models are quite complex.

EPA has used the APEX model for estimating human population exposure to ozone and CO and produced substantial information and data to support model development (U. S. EPA, 2012; U. S. EPA, 2010). There has not yet been a systematic analysis to quantify the E_a/C ratio for $PM_{2.5}$ by region and season. In the last revision of the PM standard, EPA did not include PM exposure modeling in part because of a perception of the need for more research to develop the exposure modeling approaches for $PM_{2.5}$. A goal of this work is to provide an example of the application of simulation based exposure modeling to quantification of regional and seasonal differences in $PM_{2.5}$ exposure.

2. Justifications for Inputs (ACH, P, k) Selection

2.1. Value

Data for ACH, P and k are based on published peer reviewed papers and are with respect to the selected geographic areas and four seasons. Details on inputs selection can be found in Cao and Frey (2011). Cao and Frey (2011) reviewed literature regarding distributions of air exchange rate (ACH), penetration (P), and deposition (k), and recommended values to be used for the three areas studied.

Data regarding ACH were based on studies published from 1995 to 2006, such as Murray and Burmaster (1995), the RIOPA study (2005), Koontz and Rector (1995), and Wallace *et al.* (2006). The frequency distributions of ACH among studies of different publication years were similar by location and season. Thus, the publication date is not a significant factor affecting the quantified distributions of ACH.

The physical constraint of P is less than 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 (2005) for I/O<1, and the lower bound of the reported 95 percent Confidence Intervals (CI)s for 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 lower bound of $k = 0.2\ h^{-1}$ is assumed consistent with the mean value observed for the RIOPA data in which I/O ≤ 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.

2.2. Distribution Type

For ACH, Murray and Burmaster (1995) summarized ACH data compiled by BNL for 2,844 households. 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. For P, a triangular distribution is used to represent the judgments in the absence of a probability sample of data (Cullen and Frey, 1999). For k, 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}.

2.3. Sensitivity analysis

Cao and Frey (2011) did a sensitivity analysis to assess the relative importance of ACH, P and k in estimating human exposure. ACH is the most sensitive input for both ambient and nonambient exposure to $PM_{2.5}$, whereas the results are not sensitive to the choice of distribution for P and k.

3. Inter- and Intra-individual Variability in Non-ambient Exposure (E_{na}) and Total Exposure (E_t)

For estimated $PM_{2.5}$ non-ambient exposure based on simulation results, the CV of inter-individual variability is 54% higher than for intra-individual variability. The higher CV for inter-individual variability is mainly associated with differences between those exposed to ETS versus those not exposed. To provide insight regarding differences in non-ambient exposure, three subgroups are defined: (1) non-smokers without ETS exposure at home; (2) non-smokers with ETS exposure at home because of another person who smokes; and (3) smokers. Groups 2 and 3 have approximately 7 and 11 times higher daily average E_{na} than Group 1, respectively. Compared to the total population, the CV of inter-individual variability for Group 1 is 13% lower and for Groups 2 and 3 is approximately 50% lower. Inter-individual variability in the total population is associated with large differences in mean exposures among the three groups; the variability within each group is much less than that of the total population. The CV of inter-individual variability in E_t is approximately 40% lower in each subgroup than in the total population.

The CV of intra-individual variability in E_{na} for Group 1 does not differ much from that of the total population, but is approximately 50% lower for Groups 2 and 3. Activity patterns for the two groups with ETS exposure are similar over time for each individual. For total exposure, the CV of intra-individual variability in Group 1 is similar to that of the total population, whereas it is approximately 20% lower for Groups 2 and 3. The decreased intra-

individual variability in E_t for the latter subgroups results from within-group homogeneity in ETS exposure.

To evaluate whether inter-individual variability in exposure differs from one day to another and help to explain the difference in variability in exposure between short and long averaging times, a day-by-day matrix of E_t is constructed to perform correlation analysis between day types. Three types of days are simulated, including weekday, Saturday, and Sunday. Correlations of E_t between day types indicate whether inter-individual variability in exposure differs from one day to another. Day types include weekday, Saturday, and Sunday. Strong positive correlations are found to be significant at the 0.01 level for all day type combinations, with correlation coefficients ranging from 0.72 to 0.82.

Inter-individual variability in E_t is sensitive to differences between those exposed to ETS versus those not exposed. For Group 1, correlations between days within the same day type are strong ($r_p > 0.7$) while correlations between different day types are weak ($r_p < 0.1$). For the other two groups, correlations are strong both within the same day type and between different day types (r_p varies from 0.58 to 0.75). Strong correlation within the same day type is a result of how CHAD diaries are sampled by SHEDS-PM. Each individual is assigned the same diary for each day of given type. Strong covariations between different day types for smokers are a result of similarity in the estimated number of cigarettes smoked from day-to-day. Smokers tend to have high smoking exposure each day irrespective of any other activity.

4. Discussion of Seasonal Variability in Elderly Activity Patterns

Based on the version of CHAD that is used by the SHEDS-PM model, there are a total of 21,667 diaries for all ages. For people over 65 years old, there are 1,810 diaries, with 659 male diaries and 1,151 female diaries. The total number of elderly individuals sampled in CHAD is 1,615, including 588 males and 1,027 females. Thus, most of the studies that comprise CHAD are based on only one diary day per person. The National Human Activity Pattern Study collected 1-day activity diaries from participants and is the largest and most geographically diverse survey included within CHAD. A Baltimore study included within CHAD sampled several elderly people for four to nine days. The Baltimore study has 85 elderly diaries. Thus, the Baltimore study does not comprise a substantial portion of CHAD dairies for those 65 and older. Therefore, the diary assigned to each simulated individual in SHEDS-PM will not be overwhelmed by the possible autocorrelation in elderly activity patterns from the Baltimore study.

To assess seasonal and regional differences in elderly activity patterns, the diaries of people 65 years and older were extracted from CHAD by gender, state and month. The average aggregated time spent of those individuals for indoors, outdoors, and in-vehicle microenvironments were compared. Since activity patterns tend to differ by gender, climate zone and season, the comparison is stratified by gender and by climate region between winter and summer seasons. The criteria for defining climate regions and seasons are based on Murray and Burmaster (1995). States with annual heating degree days higher than 5,500 are categorized as the colder climate zone, whereas those less than 5,500 are categorized as the

warmer climate zone. Winter includes December, January and February. Summer includes June, July and August. There are relatively few relevant CHAD diaries from which to assess this hypothesis. The available sample size is 153 and 162 for males in winter and summer, respectively, and 271 and 270 for females in winter and summer, respectively. The daily average aggregated time spent of these elderly people among indoors, outdoors, and in-vehicle microenvironments are summarized in Table A-7.

When comparing the daily average aggregated time spent in different microenvironments by the elderly in CHAD, as indicated in Table A-7, we found that: (1) for both climate regions, people have a tendency to spend more time indoors in winter, and increased time outdoors in summer; (2) in each of the two seasons studied, females tend to spend more time indoors than males, and conversely males spend more time outdoors than females; (3) there is not much difference in time spent in vehicle between seasons and between climate regions; (4) for the colder climate zones, males spend two hours more time indoors in winter than in summer, whereas the seasonal difference in time spent indoors is only 50 minutes for females; (5) for the warmer climate zones, males spend about one hour more time indoors in winter than in summer, whereas the seasonal difference in time spent indoors is 40 minutes for females; and (6) t-test results indicate that time spent indoors differs significantly at $p=0.05$ between genders for a given season and climate zone, and between seasons for a given gender and climate zone. Time spent outdoors also differs significantly between genders at a given season and climate zone, and between seasons for a given gender and climate zone. In winter, time spent indoors differs significantly between climate zones for both females and males, but the region difference in time spent outdoors is

only significant for females. In summer, no significant regional difference in elderly activity patterns is found for any gender. Time spent in-vehicle does not differ significantly across season and regions, or between genders. Thus, there is limited quantitative indication of the potential for seasonal or climatic differences in time spent indoors or outdoors. However, the available sample sizes of diaries with respect to the combinations of locations and seasons that are the focus of this paper are not sufficient for the purpose of quantifying such differences, nor is it possible to take into account regional differences at the level of the specific study domain geographic areas that are the focus of this paper.

5. Discussion of Autocorrelation in Elderly Activity Patterns

To evaluate whether there is autocorrelation in daily activities for the elderly, we extracted elderly individuals from CHAD who have been tracked for over 4 days and compared the day to day activities for each individual. There were only 11 individuals that met this criteria, and they all came from the Baltimore study. There are 72 daily diaries for these individuals, and each individual had 4 to 9 days of diaries. The CVs of intra-individual variability in time spent indoors from day-to-day ranged from from 0 to 0.19 among the 11 individuals. For 7 of the individuals, the CV was less than 0.04. This limited sample provides some evidence that activity patterns are repeated from day-to-day, at least for these particular individuals."

Based on 1,810 diaries in CHAD for people 65 years and older, the inter-individual variability in the amount of time spent indoors has a CV of 0.12. This CV is similar to the range of intra-individual variability observed for the 11 individuals for whom multiple diaries were available. There is relatively little variability in activity patterns when comparing individuals, and when comparing the same individual for multiple days. Thus, the limited evidence regarding longitudinal activity patterns indicates that activity patterns are highly repeatable from day-to-day. Because the activity patterns are also similar when comparing individuals, the simulation results are based on repeatable activity patterns.

6. Discussion of Potential Modeling Assumptions that May Cause Decreased Variability in the Exposure Results

Some potential modeling assumptions that may cause decreased variability in the exposure results are identified and discussed. These include the size of the grids used in the air quality models, housing volume distributions, air exchange rates, longitudinal diary sampling method, and CHAD sample size for the population of interest.

NYC is covered by 12 12 km × 12 km grid cells. The input daily average PM2.5 concentrations are from EPA CMAQ grid cells data. In order to be processed by SHEDS-PM, grid cell concentrations are fused into census tract level. The spatial variation in daily ambient concentrations among census tracts are small, as indicated in part (b) of Table A-2. $PM_{2.5}$ is comprised of several key components, including sulfate, nitrate, elemental carbon, organic carbon, and others. Sulfate and nitrate are secondary particles that are formed from upwind precursors. Therefore, the PM concentrations associated with secondary particles tend to be spatially uniform. Portions of the other PM components are transported from upwind, whereas some may be emitted directly in the study location. In principal, smaller grid cells would lead to more estimated spatial variability in ambient concentration of primary PM, as long as the emission inventories for primary PM emissions are accurate. However, as the spatial resolution of an emission inventory is increased, uncertainty regarding the localized primary emission rates increases. Emission inventory are more robust when averaged over a larger geographic area. Given that a substantial portion of PM components have little spatial variability, and the trade-off in accuracy and uncertainty of

estimating primary emissions with smaller grid sizes, it is not likely that a grid size of 12 km by 12 km is introducing significant bias to the results.

The housing volume for each exposed individual was sampled from lognormal distributions of housing volume based on housing type. Thus, inter-individual variability in exposure is influenced by inter-home variability in size, and this is not a factor that could lead to decreased variability in exposure results.

Area- and season-specific ACH distributions are used in SHEDS-PM simulations to represent geographic and seasonal variability in ACH. For a single individual, ACH is sampled independently from day to day. For a single day, inter-individual variability is estimated taking into account the full estimated range of ACH for each housing type. For averaging times of longer than one day, SHEDS-PM samples independently from the applicable housing type ACH distribution for each individual. The average of independently sampled ACHs from day to day for an individual will trend toward the mean of the ACH distribution and consequently result in less variability. If ACH could be sampled in a person-specific (autocorrelation) manner, the variability in exposures over time would become larger. Thus the current sampling method for ACH in SHEDS-PM will underestimate inter-individual variability in exposure for averaging times of more than one day. However, the variability in the daily exposure results will not be affected since ACH is sampled based on defined distributions on each day.

The CHAD database contains very limited data for longitudinal activity patterns. Thus, SHEDS-PM simulates longitudinal activity using a stratified sampling approach. In this approach, an individual is categorized by age and sex. Each day, a diary is sampled at

random from the set of diaries of individuals of the same age range and sex. As noted in Point 5, based on analysis of a limited number of elderly individuals for whom approximately multiple days of diaries are available, there tends to be little variation in the day-to-day activity pattern. Furthermore, there tends to be little variation in the activity pattern when comparing different individuals. Given this structure of the diary data, SHEDS-PM in effect samples repeatable activity patterns from day to day for each individual.

To assess whether the limited elderly people in CHAD will cause decreased variability in exposure estimate, elderly diaries were extracted from CHAD for analysis. There are 1,810 diaries in CHAD for people 65 and older, including 659 for males and 1,151 for females. The main sources of uncertainty in estimates of activity patterns based on this sample of diaries are: (a) are the diaries a random sample?; (b) are there errors in recording the activity patterns?; and (c) random sampling error based on sample size and variability in the sample, if the diaries were randomly sampled. Most likely, the diaries are not truly a random sample, and thus there could be bias in the mean estimate of time spent in each microenvironment or in the distribution of time among microenvironments. This type of error cannot be corrected merely by obtaining more diaries, but rather by changing the sampling method. It is unclear if the bias is positive or negative, and the bias is difficult to quantify. If diaries were not correctly completed, there can be errors in the inferred activity patterns. These errors could occur in any part of the diary and thus their combined effect on the activity estimates may tend to be random. Confidence intervals on the mean time spent in each microenvironment can be estimated if an assumption is made that the diaries are a

random sample with no recording errors. However, the confidence intervals may be robust to some degree of deviation from these assumptions.

A limitation regarding how well human activity is characterized is that seasonal or regional patterns are not adequately addressed in the current SHEDS-PM simulation. As indicated in Table A-7, although the results indicate some seasonal or regional differences in human activity patterns, the limited sample size weakens the conclusion, and data specific to the geographic regions that are the focus of this paper are insufficient to quantify differences for the specific case studies developed in this work. There is not sufficient geographic coverage of diaries in CHAD to specifically represent a given location. If people in a given season or location spend more time outdoors, the average E_a/C ratio for that specific season or domain would increase. Thus, due to the limited number of people in CHAD from which to draw from when assigning diaries, estimated results may not fully account for possible seasonal or geographic differences in activity patterns. Substantially more data is needed to better quantify such differences.

While it is always preferred to have more diaries, a truly random sample of 30 or more diaries would tend to give a representative characterization of variability within a homogeneous category of individuals (e.g., age, sex, health status, climate zone, land-use patterns, and so on). It is difficult to reach a clear conclusion as to the potential direction of bias of the available sample.

In summary, our findings are that: (a) the 12 km x 12 km grid cells are not likely to reduce variability in the estimates compared to smaller grid cells without a trade-off in increased uncertainty of the estimate: (b) distributions of housing size are used and thus do

not contributed to decreased variability in exposure estimates; (c) ACH distributions sampled on a given day will appropriate account for inter-individual variability, but that the day-to-day sampling approach leads to reduced variability in longer term average exposure estimates; (d) the limitations of the longitudinal diary building method are not likely to substantially affect results for the 65 and older population, given similarities between intra-individual and inter-individual variability in activity patterns; and (e) the limited number of relevant diaries available in CHAD does not necessarily imply that variability is underestimated, but implies that there is more uncertainty in the estimate than if a larger number of diaries could be available.

7. Discussion of the Difference in Mean E_a/C Ratios

The range in means from the lowest 0.44 to the highest 0.60 is not a small difference. The E_a/C ratio typically varies from 0 to 1. The simulated sample size for individual daily exposure is approximately 1,500,000, which is 50,000 individuals times 30 days in each case. Thus, the absolute range of the mean of E_a/C over all individuals and days is constrained. For example, the TX domain fall case has a mean E_a/C of 0.60, with a standard deviation of 0.13 of individual daily exposure. The 95% confidence interval for the mean, taking into account the exact sample size of 1,584,410, is $0.60 \pm 2.0 \times 10^{-4}$. Thus differences between the mean E_a/C ratios are statistically significant. In this context, a difference in mean E_a/C of 0.60 versus 0.44 is relatively substantial. There is a 36% difference between 0.60 and 0.44.

Table A-1. Other Input Parameters in SHEDS-PM for Microenvironmental $PM_{2.5}$ Concentrations, excluding Residential Microenvironment[a]

Microenvironment	Calculation Scheme	Parameter	Distribution Type	Distribution Values
All Outdoor	Scaling Factor	Factor	Fixed	1.0
Office	Linear Regression	Slope	Fixed	0.18
		Intercept	Fixed	3.6
		Residuals	Normal	μ =0, σ =2.9
School	Linear Regression	Slope	Fixed	0.6
		Intercept	Fixed	6.8
		Residuals	Normal	μ =0, σ =5.4
Store	Linear Regression	Slope	Fixed	0.75
		Intercept	Fixed	9.0
		Residuals	Normal	μ =0, σ =2.1
Restaurant	Linear Regression	Slope	Fixed	1.0
		Intercept	Fixed	9.8
		Residuals	Normal	μ =0, σ =10
Bar	Linear Regression	Slope	Fixed	1.0
		Intercept	Fixed	9.8
		Residuals	Normal	μ =0, σ =10
All Other Indoor	Linear Regression	Slope	Fixed	0.85
		Intercept	Fixed	8.4
		Residuals	Normal	μ =0, σ =4
All In-vehicle	Linear Regression	Slope	Fixed	1
		Intercept	Fixed	0
		Residuals	Normal	μ =0, σ =6.64

[a.] Source: Burke, J.M., M.J. Zufall and H. Ozkaynak (2001). "A Population Exposure Model For Particulate Matter: Case Study Results for $PM_{2.5}$ in Philadelphia, PA," *Journal of Exposure Analysis and Environmental Epidemiology*, 11(6), 470-489.

Table A-2. Summary of 2002 Average Daily Ambient $PM_{2.5}$ Concentrations to the SHEDS-PM Model Input (μg/m^3)[a]

(a) Mean Values

Time of Year	Location					
	NC Domain		Harris County, TX		NYC	
	Mean	StdDev	Mean	StdDev	Mean	StdDev
April (Spring)	12.8	4.1	15.6	5.9	12.0	7.5
July (Summer)	20.7	9.9	15.7	6.7	20.9	13.3
October (Fall)	13.7	7.5	13.5	5.1	10.7	6.6
December (Winter)	12.1	6.1	10.8	4.9	13.2	6.1

[a.] Means are based on averages of 12 × 12 km grid cell concentrations; standard deviations are for daily averages in each grid cell among all days of the month (μg/m^3).

(b) Standard Deviations

Time of Year	Location		
	NC Domain	Harris County, TX	NYC
	StdDev[b]		
April (Spring)	1.4	2.6	1.2
July (Summer)	9.8	2.7	2.5
October (Fall)	1.9	2.0	1.1
December (Winter)	1.7	1.6	1.4

[b.] Average standard deviations for spatial variation on each day (μg/m^3).

Table A-3. Summary of Inter- and Intra-individual Coefficient of Variations (CVs), NC Domain, Spring 2002

(a) Total

CV	C	E_a	E_a/C	E_{na}	E_t	ACH
Inter-individual	0.11	0.37	0.35	2.32	1.61	1.01
Intra-individual	0.32	0.45	0.31	1.06	0.46	0.91

(b) Sub-groups

Non-smokers and No ETS Exposure at Home

CV	E_{na}	E_t
Inter-individual	2.03	0.97
Intra-individual	1.20	0.48

Non-smokers Exposed to ETS at Home

CV	E_{na}	E_t
Inter-individual	1.24	1.06
Intra-individual	0.48	0.35

Smokers

CV	E_{na}	E_t
Inter-individual	1.06	0.95
Intra-individual	0.46	0.37

Table A-4. Correlation of Inter-individual Variability in Exposures between Pairs of Day Types, NC Domain, Spring 2002[a]

Total Exposure (E_t)

	Correlation Coefficient [b]		
Day Type	Weekday	Saturday	Sunday
Weekday	0.807	0.730	0.723
Saturday	-	0.809	0.734
Sunday	-	-	0.815

Ambient Exposure (E_a)

	Correlation Coefficient [b]		
Day Type	Weekday	Saturday	Sunday
Weekday	0.342	0.030	0.026
Saturday	-	0.390	0.025
Sunday	-	-	0.423

Non-ambient Exposure (E_{na})

	Correlation Coefficient [b]		
Day Type	Weekday	Saturday	Sunday
Weekday	0.799	0.724	0.716
Saturday	-	0.802	0.727
Sunday	-	-	0.809

Ratio of Ambient Exposure to Ambient Concentration (E_a/C)
Original Unsorted

	Correlation Coefficient [b]		
Day Type	Weekday	Saturday	Sunday
Weekday	0.361	0.011	0.012
Saturday	-	0.428	0.013
Sunday	-	-	0.460

Randomly Sorted

Day Type	Correlation Coefficient [b]		
	Weekday	Saturday	Sunday
Weekday	0.000	0.000	-0.001
Saturday	-	0.002	0.000
Sunday	-	-	-0.001

[a.] Individual daily exposure values in the NC domain, spring case are extracted from SHEDS-PM output to construct a 50,516 individuals by 31 days matrix. For each day, correlations of inter-individual variability in exposure are calculated with each other day. Days are categorized by day type, including weekday, Saturday and Sunday. For each of the 31 days, the average correlations with all other days of the same day type, and with all days of each of the other two types, are estimated. A 31-day by 3-day type matrix is constructed. Correlation coefficients in the table above are based on averages of correlation coefficients of days with the same day type from the day-day type matrix.

[b.] All values are significant at the p = 0.01 level.

(b) Correlation of Total Exposures (E_t) in Sub-groups

Non-smokers and No ETS Exposure at Home

Day Type	Correlation Coefficient		
	Weekday	Saturday	Sunday
Weekday	0.746	0.086	0.074
Saturday	-	0.729	0.075
Sunday	-	-	0.705

Non-smokers Exposed to ETS at Home

Day Type	Correlation Coefficient		
	Weekday	Saturday	Sunday
Weekday	0.749	0.702	0.704
Saturday	-	0.748	0.703
Sunday	-	-	0.730

Smokers

Day Type	Correlation Coefficient		
	Weekday	Saturday	Sunday
Weekday	0.653	0.647	0.608
Saturday	-	0.676	0.626
Sunday	-	-	0.682

Table A-5. Summary of Average Ratio of Ambient Exposure to Ambient Concentration (E_a/C), Non-ambient Exposure (E_{na}), and Total Exposure (E_t) by Day Type, Season, and Geographic Area[a]

Area	Season	Day Type	E_a/C	E_{na} (μg/m^3)	E_t (μg/m^3)
NC Domain	Winter	Weekdays	0.52	13.2	19.0
		Saturdays	0.51	13.0	19.8
		Sudays	0.53	11.8	19.1
	Spring	Weekdays	0.48	14.1	20.2
		Saturdays	0.48	13.8	20.9
		Sudays	0.50	12.6	18.7
	Summer	Weekdays	0.56	11.1	22.8
		Saturdays	0.57	11.0	22.6
		Sudays	0.58	10.0	21.9
	Fall	Weekdays	0.56	11.7	18.8
		Saturdays	0.55	11.5	19.1
		Sudays	0.57	10.5	20.6
Harris County, TX	Winter	Weekdays	0.57	8.8	14.5
		Saturdays	0.57	8.6	17.4
		Sudays	0.59	7.7	14.1
	Spring	Weekdays	0.51	10.4	18.5
		Saturdays	0.52	10.2	18.1
		Sudays	0.53	9.0	16.2
	Summer	Weekdays	0.51	10.1	17.9
		Saturdays	0.51	10.1	18.3
		Sudays	0.53	8.8	13.3
	Fall	Weekdays	0.59	8.1	16.1
		Saturdays	0.60	7.9	16.3
		Sudays	0.61	7.1	14.9
NYC	Winter	Weekdays	0.54	13.3	20.0
		Saturdays	0.54	12.9	21.6
		Sudays	0.56	11.7	19.1
	Spring	Weekdays	0.52	13.9	20.8
		Saturdays	0.52	13.8	19.0
		Sudays	0.54	12.4	16.6
	Summer	Weekdays	0.59	11.4	23.7
		Saturdays	0.59	11.3	18.7
		Sudays	0.60	10.0	26.4
	Fall	Weekdays	0.44	18.0	22.7
		Saturdays	0.44	17.7	22.1
		Sudays	0.45	16.0	20.2

[a.] E_a/C: ratio of ambient exposure to ambient concentration; E_{na}: nonambient exposure (μg/m^3); E_t: total exposure (μg/m^3).

Table A-6. Correlations Coefficients between Daily Average Exposures and Concentrations[a]

Area	Season	Sample Size	C vs. E_a	C vs. E_{na}	C vs. E_t	E_a vs. E_{na}	E_a vs. E_t	E_t vs. E_{na}
NC Domain	Winter	1,504,680	0.846	-0.005	0.100	-0.380	0.086	0.992
	Spring		0.659	-0.001	0.060	-0.070	0.023	0.996
	Summer		0.886	-0.004	0.214	-0.034	0.212	0.969
	Fall		0.899	-0.006	0.150	-0.028	0.146	0.985
Harris County, TX	Winter	1,533,300	0.832	-0.005	0.123	-0.054	0.100	0.988
	Spring		0.787	0.000	0.116	-0.035	0.113	0.989
	Summer		0.804	-0.003	0.136	-0.040	0.133	0.804
	Fall		0.848	-0.001	0.153	-0.036	0.146	0.983
NYC	Winter	1,570,710	0.832	0.000	0.105	-0.055	0.071	0.992
	Spring		0.887	0.003	0.122	-0.029	0.105	0.991
	Summer		0.912	-0.006	0.269	-0.047	0.256	0.953
	Fall		0.835	0.002	0.068	-0.025	0.055	0.997

[a.] C: ambient concentration; E_a: ambient exposure; E_{na}: non-ambient exposure; E_t: total exposure.

Table A-7. Comparison of Distribution of Elderly Activity Based on Time for Males and Females in Winter and Summer (Mean ±95% Confidence Interval)

(1) Elderly from colder climate zones[a]

Sex	Male		Female	
Season	Winter	Summer	Winter	Summer
Sample Size	72	76	115	157
Indoors (minutes)	1,335 ±30	1,195 ±56	1,375 ±15	1,327 ±19
In-Vehicle (minutes)	55 ±19	70 ±21	51 ±13	57 ±12
Outdoors (minutes)	67 ±34	175 ±53	13 ±8	56 ±16

[a] These data are based on CHAD diaries for persons 65 years and older from regions with colder climate that have annual heating days greater than 5,500, including KY, IL, IN, MO, OH, TN, WV, IA, MI, MN, WI, CT, DE, ME, MD, MA, NH, NJ, NY, PA, RI, VT, ID, OR, WA, MT, NE, ND, SD, WY, NV, UT, CO.

(2) Elderly from warmer climate zones[b]

Sex	Male		Female	
Season	Winter	Summer	Winter	Summer
Sample Size	81	86	156	113
Indoors (minutes)	1,284 ±37	1,228 ±39	1,338 ±19	1,297 ±29
In-Vehicle (minutes)	76 ±21	65 ±18	57 ±13	56 ±13
Outdoors (minutes)	79 ±28	147 ±35	43 ±15	87 ±27

[b] These data are based on CHAD diaries for persons 65 years and older from regions with warmer climate that have annual heating degree days less than 5,500, including AL, FL, GA, NC, SC, VA, AZ, NM, CA, AR, LA, KS, MS, OK, TX.

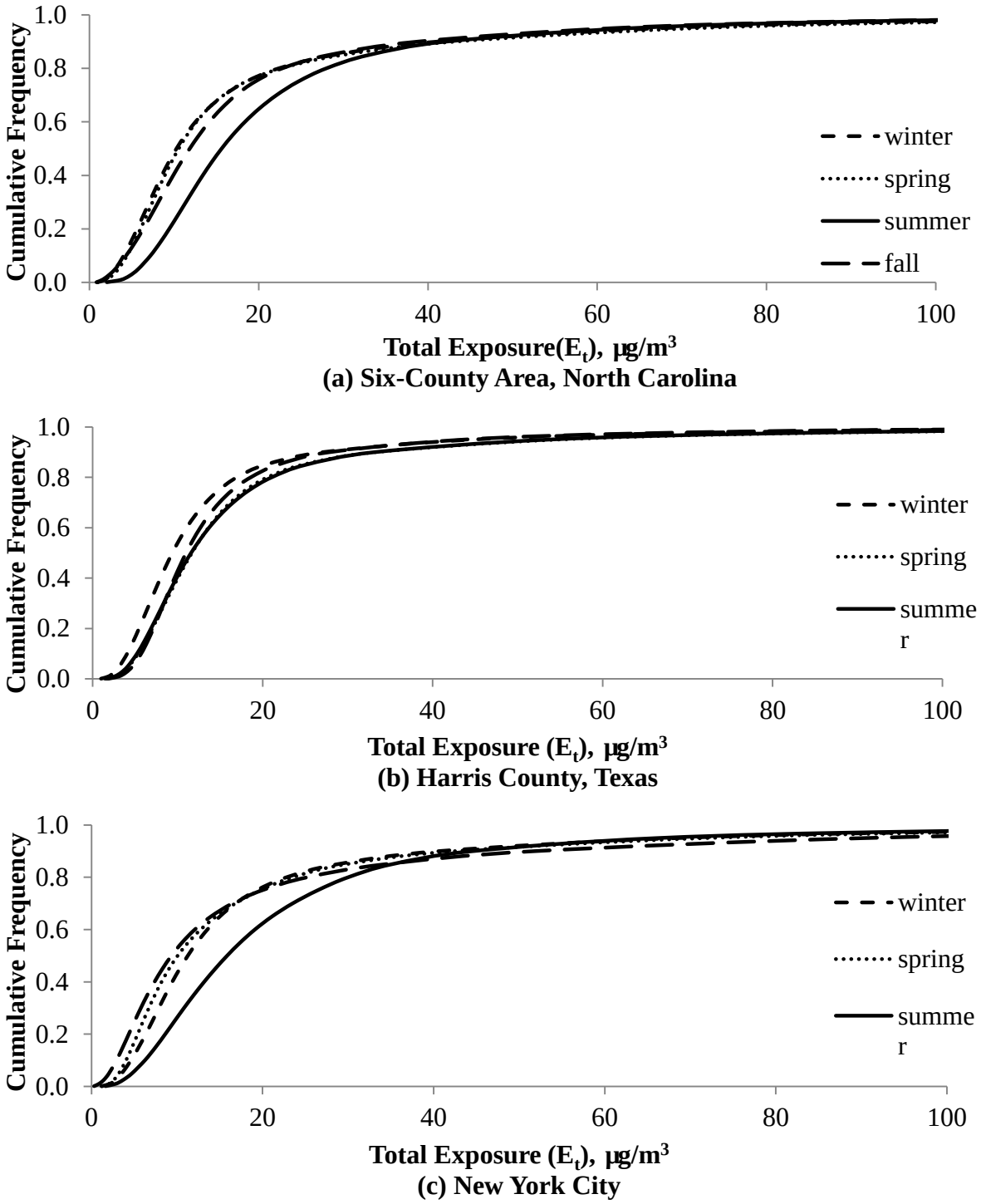

Figure A-1. Cumulative Distribution Functions (CDFs) of Inter-individual Variability in Daily Total Exposure (E_t) in NC Domain, Harris County, and NYC in 2002 for Four Seasons (µg/m^3)

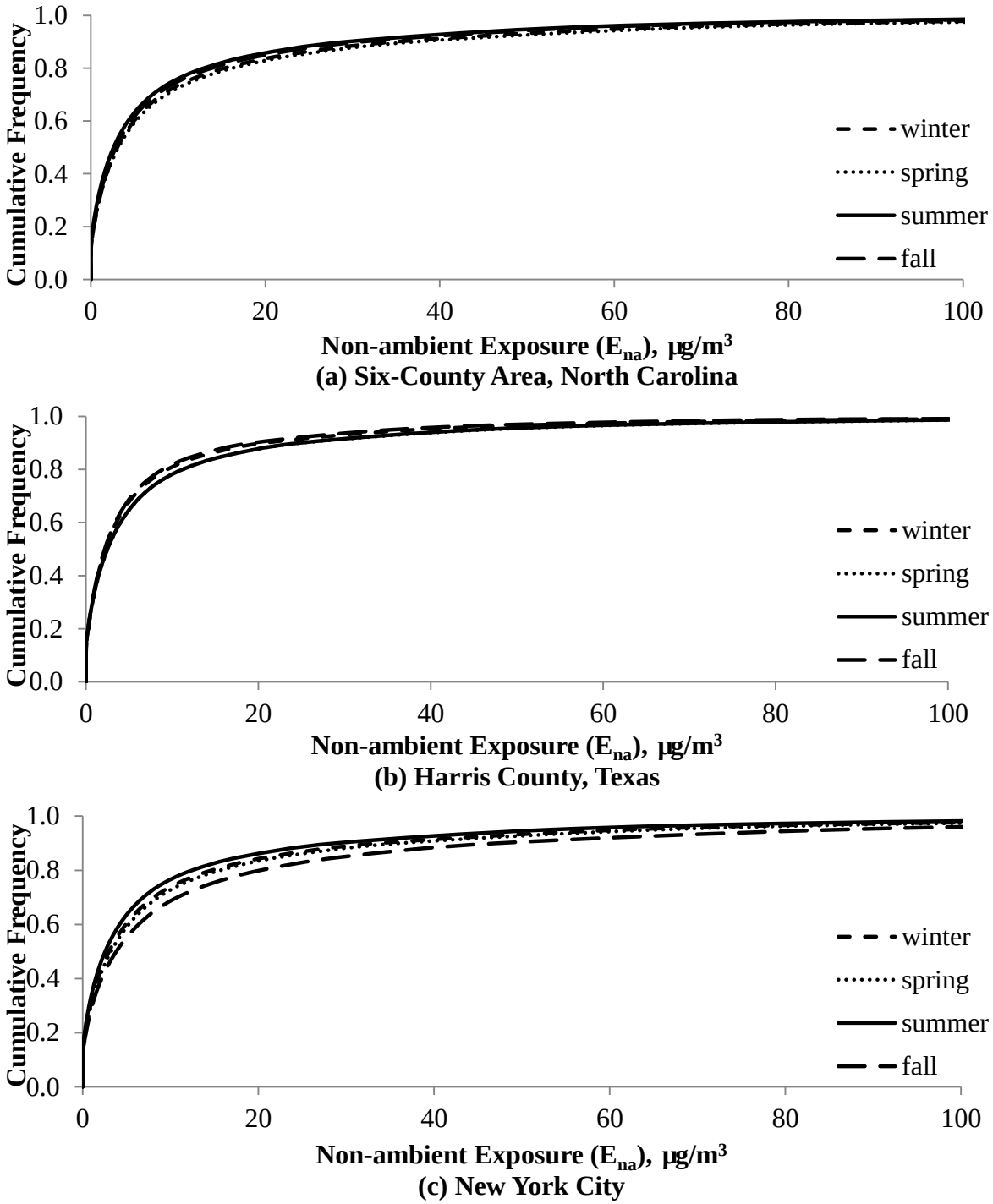

Figure A-2. Cumulative Distribution Functions (CDFs) of Inter-individual Variability in Daily Non-ambient Exposure (E_{na}) in NC Domain, Harris County, and NYC in 2002 for Four Seasons (µg/m^3)

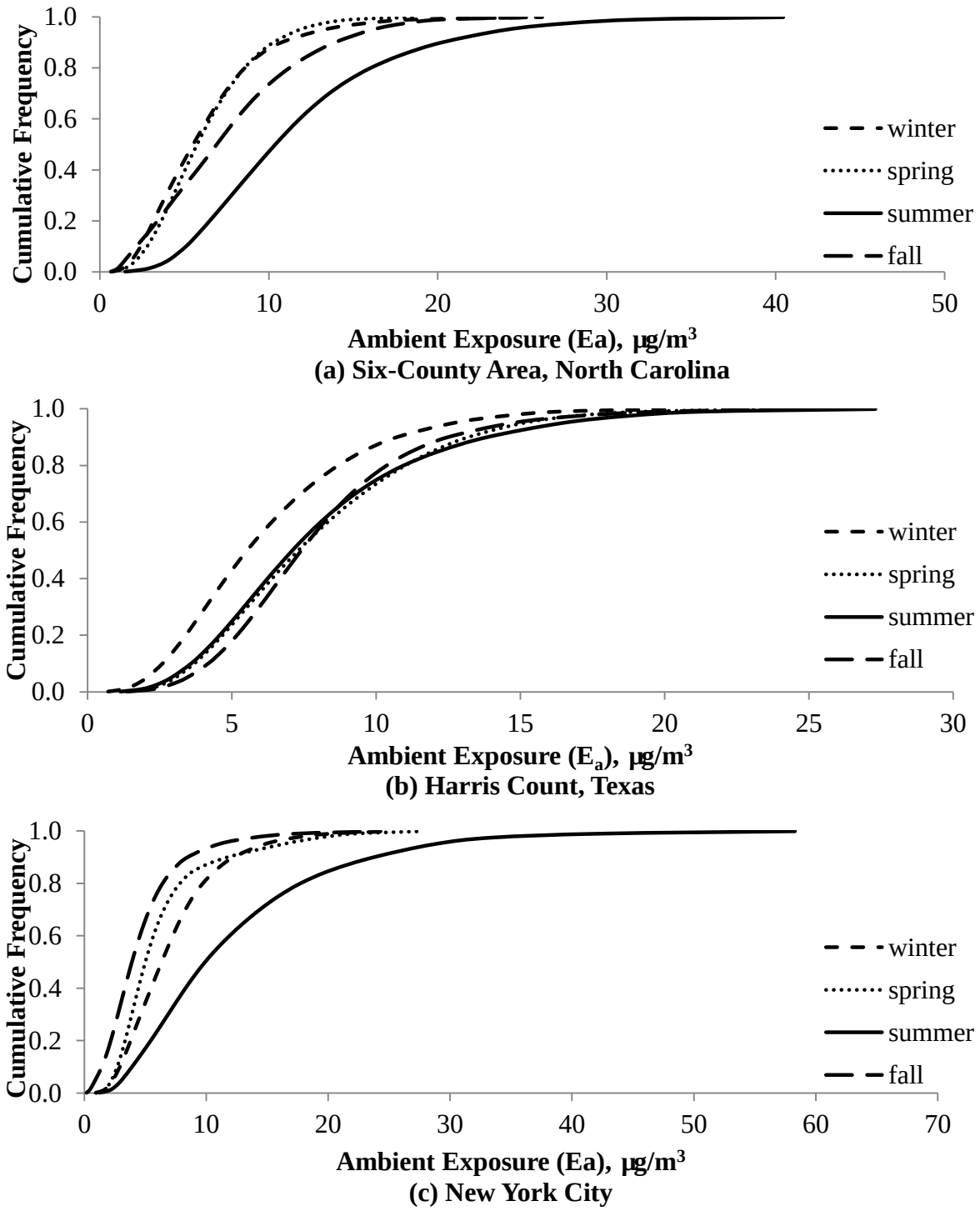

Figure A-3. Cumulative Distribution Functions (CDFs) of Inter-individual Variability in Daily Ambient Exposure (E_a) in NC Domain, Harris County, and NYC in 2002 for Four Seasons (µg/m^3)

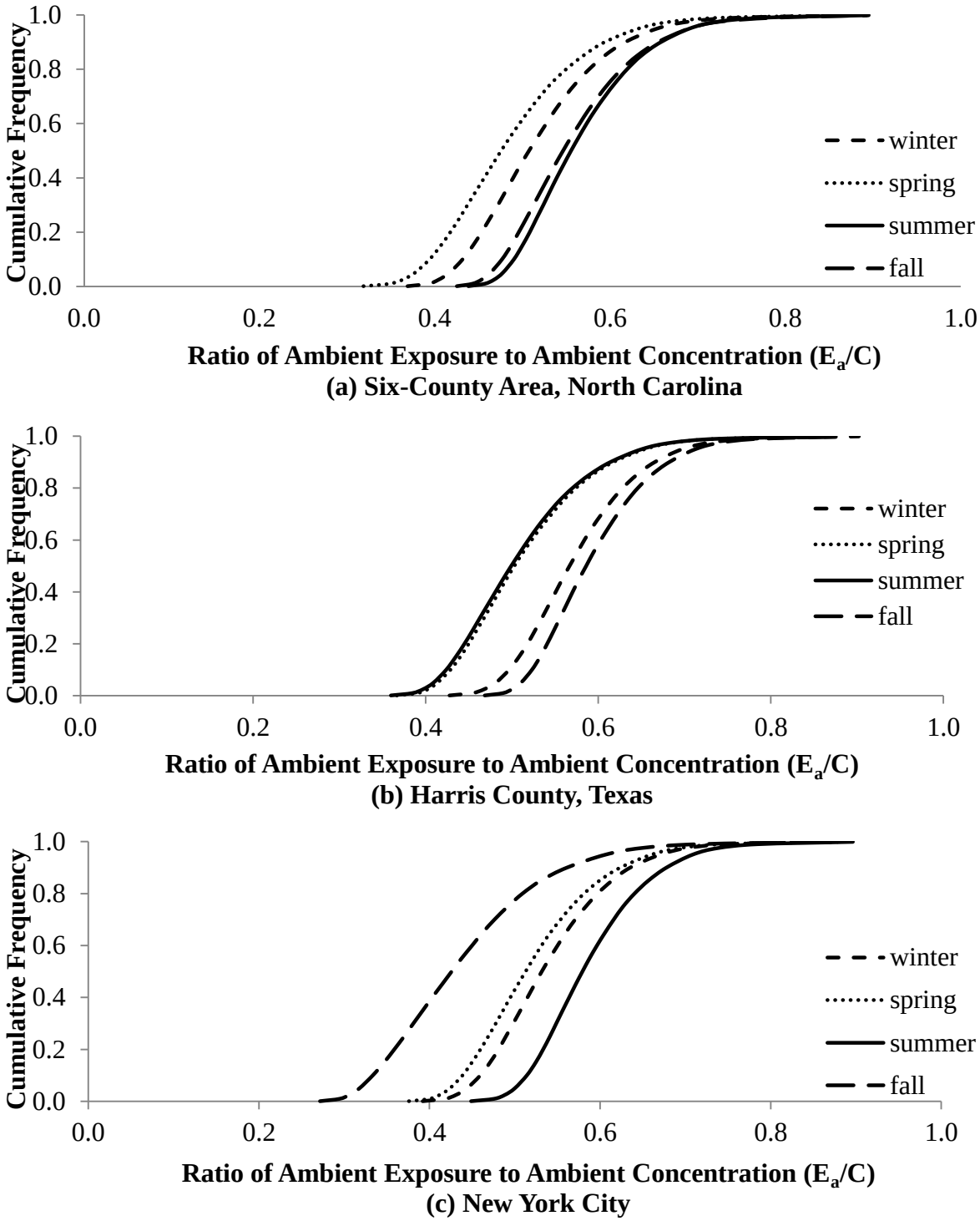

Figure A-4. Cumulative Distribution Functions (CDFs) of Inter-individual Variability in Monthly Average Ratio of Ambient Exposure to Ambient Concentration (E_a/C) in NC Domain, Harris County, and NYC in 2002 for Four Seasons

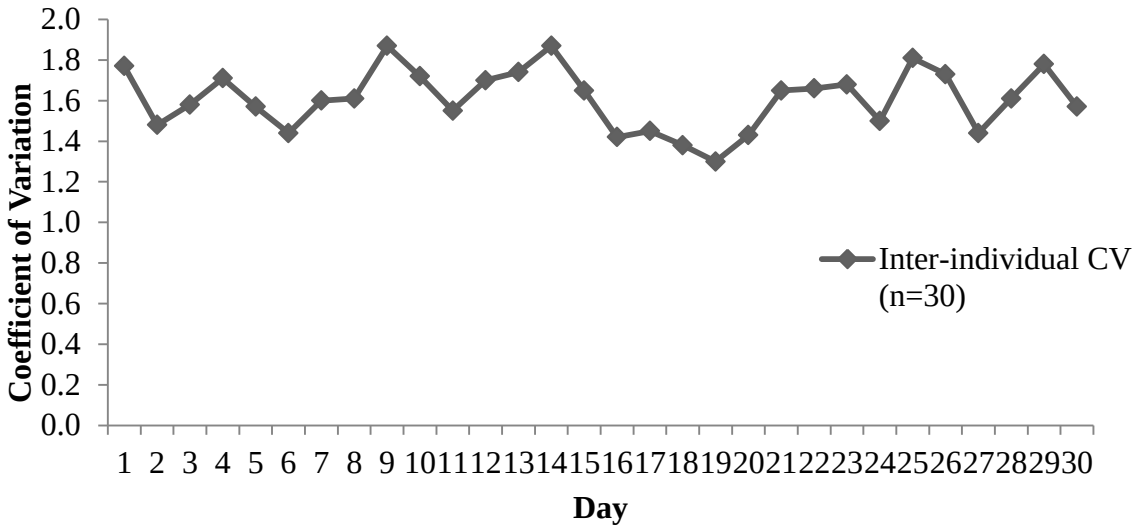

Figure A-5. Coefficient of Variation (CV) for Inter-Individual Variability in Total Exposure (E_t) for Each Simulated Day of Spring NC Domain Exposure Case Study, 2002

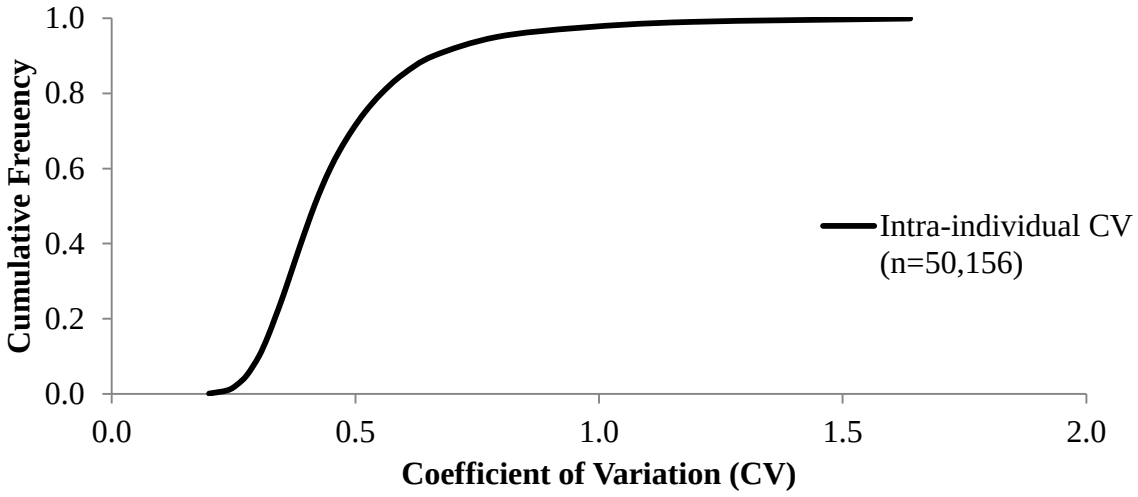

Figure A-6. Cumulative Distribution Function of Coefficient of Variation of Intra-individual Variability in Total Exposure (E_t) among 31 Simulated Days for Spring NC Domain Case Study, 2002

Appendix B Supporting Information for Part III

Table B-1. Distribution of Cooking Time[a] in Consolidated Human Activity Database (CHAD)

(1) All Ages and Genders (total number of diaries in CHAD = 7286)

Mean (hr/day)	1.0
Median (hr/day)	0.7
Standard Deviation (hr/day)	1.0
5th percentile (min/day)	5
95th percentile (min/day)	165

(2) By Age Group and Gender

Age Group	Gender	Number of Diaries in CHAD	Mean (hr/day)	Median (hr/day)	Standard Deviation (hr/day)	5th percentile (min/day)	95th percentile (min/day)
0-17	Male	404	0.4	0.3	0.4	5	155
	Female	646	0.6	0.4	0.6	5	155
18-64	Male	1554	0.7	0.5	0.8	5	165
	Female	3681	1.2	0.9	1.2	5	165
≥ 65	Male	243	0.8	0.5	0.8	5	165
	Female	758	1.2	1.0	1.0	5	165

[a.] Cooking time is summarized by the CHAD activity code of 11100 or 11110 ("prepare food").

Table B-2. Distribution of Individual Daily Time Spent in Consolidated Human Activity Database (CHAD)[a] by Age Group and Gender
(1) 0-17 years old
a. Male

Location	Number of Diaries in CHAD	Mean (hr/d)	Median (hr/d)	StD (hr/d)
Home: diaries have any location information (specific[b] or general[c])	4901	16.6	16.4	4.3
Home: diaries have both specific and general location information	3083	17.3	17.0	3.9
Home: diaries have only specific location information	1338	15.4	15.4	4.6
Home: diaries have only general information	480	15.7	15.2	4.5
Home: diaries have activity in kitchen	3755	1.1	1.0	0.8
Home: diaries have activity in living room	3676	2.9	2.3	2.5
Home: diaries have activity in bedroom	4391	11.1	11.0	2.7
Home: diaries have activity in other specific rooms	3993	5.8	5.8	4.0
All outdoors	3479	2.7	2.0	2.4
All in-vehicle	4111	1.2	0.9	1.2
Other indoors	3993	5.8	5.8	4.0

b. Female

Location	Number of Diaries in CHAD	Mean (hr/d)	Median (hr/d)	StD (hr/d)
Home: diaries have any location information (specific[b] or general[c])	4597	16.6	16.5	4.5
Home: diaries have both specific and general location information	2911	17.2	17.0	4.2
Home: diaries have only specific location information	1214	15.4	15.4	4.9
Home: diaries have only general information	472	16.3	16.1	4.6
Home: diaries have activity in kitchen	3537	1.1	1.0	0.8
Home: diaries have activity in living room	3288	2.8	2.2	2.4
Home: diaries have activity in bedroom	4098	11.1	11.2	2.8
Home: diaries have activity in other specific rooms	2354	0.8	0.5	0.9
All outdoors	2912	2.3	1.8	2.3
All in-vehicle	3891	1.2	1.0	1.3
Other indoors	3837	6.1	6.0	4.1

(2) 18-64 years old

a. Male

Location	Number of Diaries in CHAD	Mean (hr/d)	Median (hr/d)	StD (hr/d)
Home: diaries have any location information (specific[b] or general[c])	4557	14.3	13.7	4.5
Home: diaries have both specific and general location information	1024	14.7	14.0	4.6
Home: diaries have only specific location information	2412	13.8	13.3	4.5
Home: diaries have only general information	1121	15.0	14.0	4.6
Home: diaries have activity in kitchen	2346	1.2	0.8	1.3
Home: diaries have activity in living room	2789	4.1	3.5	3.2
Home: diaries have activity in bedroom	3332	8.5	8.4	2.9
Home: diaries have activity in other specific rooms	2770	1.3	0.5	2.0
All outdoors	2913	3.1	1.6	3.5
All in-vehicle	4142	2.0	1.4	2.2
Other indoors	3962	7.1	7.8	4.2

b. Female

Location	Number of Diaries in CHAD	Mean (hr/d)	Median (hr/d)	StD (hr/d)
Home: diaries have any location information (specific[b] or general[c])	5599	16.4	16.0	4.6
Home: diaries have both specific and general location information	2107	16.7	16.8	4.5
Home: diaries have only specific location information	1806	15.2	14.5	4.7
Home: diaries have only general information	1686	17.3	17.5	4.3
Home: diaries have activity in kitchen	3211	1.8	1.3	1.6
Home: diaries have activity in living room	3153	4.0	3.3	3.0
Home: diaries have activity in bedroom	3847	9.2	9.0	2.9
Home: diaries have activity in other specific rooms	3298	1.3	0.8	1.7
All outdoors	3155	1.8	1.0	2.3
All in-vehicle	4925	1.6	1.2	1.6
Other indoors	4878	6.1	6.0	4.1

(3) ≥ 65 years old

a. Male

Location	Number of Diaries in CHAD	Mean (hr/d)	Median (hr/d)	StD (hr/d)
Home: diaries have any location information (specific[b] or general[c])	656	18.0	18.4	4.3
Home: diaries have both specific and general location information	219	18.3	18.7	3.9
Home: diaries have only specific location information	337	17.8	18.0	4.6
Home: diaries have only general information	100	18.2	18.7	3.9
Home: diaries have activity in kitchen	453	1.9	1.4	1.8
Home: diaries have activity in living room	492	6.1	5.4	3.9
Home: diaries have activity in bedroom	539	8.9	8.8	2.7
Home: diaries have activity in other specific rooms	379	1.9	1.0	2.6
All outdoors	432	3.2	2.1	3.1
All in-vehicle	478	1.7	1.2	1.7
Other indoors	470	3.8	2.8	3.3

b. Female

Location	Number of Diaries in CHAD	Mean (hr/d)	Median (hr/d)	StD (hr/d)
Home: diaries have any location information (specific[b] or general[c])	1145	19.8	20.6	3.7
Home: diaries have both specific and general location information	602	20.1	20.8	3.6
Home: diaries have only specific location information	408	19.8	20.6	3.7
Home: diaries have only general information	135	20.1	20.8	3.6
Home: diaries have activity in kitchen	906	2.4	1.8	2.1
Home: diaries have activity in living room	899	6.4	6.0	3.7
Home: diaries have activity in bedroom	989	9.3	9.0	3.0
Home: diaries have activity in other specific rooms	716	1.6	1.0	2.1
All outdoors	579	1.6	1.0	1.9
All in-vehicle	748	1.4	1.0	1.4
Other indoors	805	3.6	2.7	3.2

a. The current version of CHAD in SHEDS-PM model contains 21,667 diaries in total. Each diary contains multiple locations information that the individual spent time in, including indoors, outdoors and in-vehicle microenvironments.

b. "Specific" means the diary indicates the specific home location that the individual spent time in, such as kitchen, living/family room, bedroom, dining room, bathroom, study/office, basement, and utility/laundry room.

c. "General" means the diary uses location codes such as "residence, general" (code = 30000), "your residence" (code = 30010), "residence, indoor" (code = 30020), and "your residence, indoor" (code = 30120) to indicate the individual's location at home.

Table B-3. Housing Dimensions and Room Specifications

"Contemporary" Single-family House:

Room	Volume[a] (m^3)	HVAC Flows[b] (m^3/hr)
Kitchen/Living Area	161	(+) 810
Hall	42	(-) 1645
Middle Bedroom	37	(+) 185
Front Bedroom	37	(+) 180
Master Bedroom	64	(+) 320
Main Bathroom	13	(+) 65
Master Bathroom	17	(+) 85
Total	371	

Multi-family Apartment:

Room	Volume[a] (m^3)	HVAC Flows[b] (m^3/hr)
Kitchen/Living Area	98	(+) 490
Hall	20	(-) 1115
Front Bedroom	44	(+) 220
Master Bedroom	49	(+) 245
Main Bathroom	15	(+) 75
Master Bathroom	17	(+) 85
Total	242	

Mobile Home:

Room	Volume[a] (m^3)	HVAC Flows[b] (m^3/hr)
Kitchen/Living Area	93	(+) 460
Hall	15	(-) 1030
Middle Bedroom	29	(+) 145
Front Bedroom	29	(+) 145
Master Bedroom	34	(+) 170
Main Bathroom	7	(+) 35
Master Bathroom	15	(+) 75
Total	222	

[a.] The total house volume for each housing type is based on results of the US Census 2000 Housing Survey.

[b.] Forced air flow for supply (+) or return (-) to/from the house HVAC system is approximately five house volumes per hour

Appendix C Supporting Information for Part IV

Chi-Square Test of Independence between Concentrations of Different Ventilation Conditions

1. This analysis is to provide rationale for using standard deviations in Table IV-2 to represent the variability in average near-vehicle and in-vehicle concentrations among the three sample days for different ventilation conditions combinations (case 1-1 to 2-8). Chi-square test is selected because the sampling method was random sampling, and the test variables under study (sample day, ventilation conditions) were categorical.
2. Test hypothesis

   H_0: Concentration measured at each sample day and ventilation conditions are independent

   H_1: Concentration measured at each sample day and ventilation conditions are not independent
3. Two 16 × 3 contingency tables (ventilation case × near-vehicle concentration on each sample day, and ventilation case × in-vehicle concentration on each sample day) was constructed for chi-square test.
4. Chi-square test using SAS software:

```
proc import out=work.test
datafile='C:\Users\vicki\Desktop\test.txt';
getnames=yes;
run;

proc print data=test;run;

proc freq data=test;
table Case*Near/chisq;
weight count_Near;
run;

proc freq data=wan;
table Case*In/chisq;
```

```
weight count_In;
run;
```

5. Results:
   (1) Near-vehicle concentration:

Statistic	DF	Value	Prob
Chi-Square	30	7.5581	1.0000
Likelihood Ratio Chi-Square	30	7.5709	1.0000
Mantel-Haenszel Chi-Square	1	0.0141	0.9056
Phi Coefficient		0.0571	
Contingency Coefficient		0.0570	
Cramer's V		0.0404	

   (2) In-vehicle concentration:

Statistic	DF	Value	Prob
Chi-Square	30	1.5293	1.0000
Likelihood Ratio Chi-Square	30	1.5361	1.0000
Mantel-Haenszel Chi-Square	1	0.1143	0.7353
Phi Coefficient		0.0303	
Contingency Coefficient		0.0303	
Cramer's V		0.0214	

6. Conclusion:
   Since P(Chi-Square) for both near- and in-vehicle concentrations are > 0.05. We cannot reject the null hypothesis (H_0). The concentrations measured on each sample day are independent of the combination of ventilation conditions. Therefore, using standard deviations to represent the variability in near- and in-vehicle concentrations are reasonable.

Appendix D Comparison of Predicted Exposures versus Ambient Fine Particulate Matter Concentrations

INTRODUCTION

Fine particulate matter ($PM_{2.5}$) is comprised of particles 2.5 micrometers or smaller in aerodynamic diameter. They are small enough to pass enter the lungs and cross the blood-air barrier in the alveoli. Based on review of numerous studies, the U.S. Environmental Protection Agency (EPA) has identified causal associations between exposure to $PM_{2.5}$ and adverse human health effects including premature death in people with heart or lung disease, nonfatal heart attacks, irregular heartbeat, aggravated asthma, decreased lung function, and increased respiratory symptoms.[1] People with heart or lung diseases, children, and older adults are the most vulnerable subpopulations to be affected by particle pollution exposure.[1]

A challenging aspect of air pollution health effects studies is to properly quantify the exposures of individuals in the population. Ambient $PM_{2.5}$ concentrations (C) are affected by meteorology and by changes in emission rates and locations of emission sources. However, actual $PM_{2.5}$ exposure to ambient origin (E_a) depends on the amount of time an individual spends in different microenvironments. Microenvironments are surroundings that can be treated as homogeneous or well characterized with respect to the concentrations of an agent.[2] Microenvironments include various indoor locations (e.g. home, work, school, restaurant, and store), outdoors, in transit, and others. For indoor microenvironments, a portion of ambient $PM_{2.5}$ penetrates and deposits to interior surfaces. Since people spend majority of daily time indoors, on average for a population, the daily exposure to particles of ambient origin is typically less than the ambient concentration, and the difference contributes to exposure errors.[3] Using ambient $PM_{2.5}$ concentrations as a surrogate for the community average personal exposure to ambient $PM_{2.5}$ will bias the estimation of health risk

coefficients by the ratio of $PM_{2.5}$ ambient exposure to ambient concentration (E_a/C).[1] The E_a/C ratio depends on housing type and activity patterns, air exchange rate, and PM deposition rate. Inter-individual variability in the estimated E_a/C ratio typically varies from 0 to 1, and the population average value of the ratio differs by region and season mostly because of differences in average residential air exchange rate.[4]

Many time-series studies examine the associations of short-term health outcomes based on a daily 24-hour interval.[5] Thus, quantification of inter-individual variability in daily average exposure, as well as ambient concentration, may help to better inform health effects studies and evaluate key sources of variability related to PM risk. Since direct measurements of individual exposure are not available with sufficient spatial and temporal coverage, few existing studies have compared variability in daily average exposures over years. Knowledge of how exposure varies by year is critical to decision making in risk management, especially for reducing variations in exposures or reducing the frequency and magnitude of high exposures.

U.S. EPA has developed the Stochastic Human Exposure and Dose Simulation Model for Particulate Matter (SHEDS-PM).[6] SHEDS-PM employs a probabilistic approach to estimate population distributions of inter-individual variability in daily average exposure using data regarding human activity patterns, microenvironmental concentrations, census population demographics and housing types for the selected geographic domain. SHEDS-PM is used here for estimating population daily average exposure for multiple years.

Three research questions will be addressed: (1) how much is variability in estimated daily average exposure to ambient air pollution influenced by variability in ambient concentration compared to other exposure factors?; (2) what is the inter-annual variation in the annual distribution of daily average exposure?; and (3) what key factors and values of these key factors lead to high exposure?

METHODOLOGY

The methodology includes: (1) scenario-based exposure modeling; (2) study design and identification of data sources for the case study; and (3) analysis of SHEDS-PM output.

Modeling Approach

In SHEDS-PM, age, sex, employment status, and housing type for each simulated individual are sampled from the year 2000 US Census to represent the population in a selected area. For each simulated individual, the amount of time spent in each microenvironment is obtained from diaries in the Consolidated Human Activity Database (CHAD).[7] The diaries are selected to match the age, sex, and smoking status of the simulated individual. Algorithms are applied to estimate $PM_{2.5}$ concentrations in each microenvironment based on ambient concentration and indoor sources. For the residential microenvironment, a single-compartment, steady state mass balance equation is used. Key factors that influence the fraction of ambient $PM_{2.5}$ concentration which penetrates and remains in the residential microenvironment are: (1) air exchange rate (ACH); (2) penetration factor (P); and (3) deposition rate (k). Distributions of ACH by area and season reflect the variability in ACH

caused by different ventilation practices used and housing types. SHEDS-PM has undergone substantial validation and evaluation. Related studies were reviewed in Jiao *et al.*.[4]

Study Design

The focus here was to compare variability in estimated daily average exposure with input ambient concentration for each of several selected years. Adults over 65 years old were selected because they are a susceptible subpopulation with respect to $PM_{2.5}$ exposure.[1] Three counties in the New York City area were selected, including Bronx, Queens and New York Counties. Daily exposures for a random sample of approximately 50,000 individuals over 65 years old were simulated from these three counties for each year between the years of 2002 to 2006.

Air quality data were from the Community Multi-scale Air Quality modeling system (CMAQ) based on the prediction of average concentrations for 12 km by 12 km grid cells, as described in Byun and Schere.[8] Distributions of ACH, P, and k were updated from default values based on literature review and are listed in Table C-1.

Lognormal ACH distributions were developed based on data for New York City from the Toxic Exposure Assessment: A Columbia-Harvard Study (TEACH-NYC) for cold and warm day temperature categories.[11] Days with daily average temperatures less than 65°F are defined as “cold,” whereas days higher or equal than 65°F are defined as “warm.” Since the current version of SHEDS-PM cannot assign ACH distribution based on daily temperature, SHEDS-PM was run twice to simulate ACH distributions of “cold” and “warm” categories, separately. Post-processing steps were done in SAS 9.3 software to select the appropriate

ACH distributions for each simulated day depending on the daily average temperature from the National Climate Data Center.

Statistical Analysis

The SHEDS-PM output includes estimated daily average values of ambient (E_a), non-ambient (E_{na}), and total exposure (E_t) for each simulated individual on each simulated day, and time spent in each microenvironment. Exposures are calculated based on individual time spent in each microenvironment and microenvironment-specific concentrations. All outputs were processed and analyzed using SAS 9.3 software. Ratios of E_a/C for each simulated individual were calculated from daily average ambient exposure divided by input daily average ambient $PM_{2.5}$ concentration in the same census tract as the individual's home. For each year, variability in estimated daily average E_a was compared with variability in daily average C, and the daily average ratio of E_a/C based on means and coefficients of variation (CVs). Distributions of daily average E_a, C and E_a/C were further compared by season and across counties by year.

RESULTS

SHEDS-PM version 3.7 was used to run all cases. The model runtime per case depends on the sample size, number of census tracts, and simulated time period selected. Typically it takes approximately 10 hours of wall clock time to simulate exposures to 50,000 individuals for a one year time period.

Variability in Daily Average Exposure versus Concentration

To compare the variability in daily average exposure with concentration, the means and CVs of estimated daily average C, E_a, and E_a/C ratio for the simulated populations are shown in Table C-2. The overall daily average E_a for the simulated population was 9.8 $\mu g/m^3$ or about 65% of the overall daily average C. CVs of overall daily average C, E_a, and E_a/C are 0.69, 0.72 and 0.22, respectively. The CV of 0.22 for the variability in overall daily average E_a/C ratio indicated that E_a is not a simple linear function of C. E_a is also influenced by ACH in a home and personal activity patterns.

Differences between the highest and lowest estimated annual average of daily values of C, E_a and E_a/C among years varied by 11.7%, 12.3% and 1.5%, respectively. Since the annual average E_a/C ratios were relatively constant over each year, the annual average E_a by year was consistent with changes in C. Higher values of C often resulted in higher E_a. Both annual average C and E_a were highest in 2003 and lowest in 2005. The small variation in daily average E_a/C from year to year was because factors affecting this ratio, such as ACH, housing type and activity patterns, were assumed to be relatively unchanged across years in the simulation.

For any single year, daily average C, E_a and E_a/C had seasonal fluctuations, especially when comparing summer to the other three seasons. As an example, Table C-3 shows the estimated daily average C, E_a and E_a/C ratio by county for winter and summer seasons in 2002. Winter and summer were selected because they represent “cold” and “warm” temperature categories. There was a clear seasonal difference in C and E_a within each county. Winter has an average daily C of about 40% higher than the summer average, while the

difference in E_a is about 30%. The increased amount of CMAQ predicted $PM_{2.5}$ concentrations during wintertime could be due to a combination of factors including wood burning during winter months,[12] as well as effects of temperature inversions on vertical mixing processes in urban area. The E_a/C ratio differed by approximately 10% between summer and winter seasons. Seasonal variation in the estimated daily average E_a/C ratio was mainly related to differences in ACH distributions by temperature, but was also affected by individual seasonal activity patterns.

For each season compared, Bronx County had the lowest average C and E_a, while the highest seasonal average values were in New York County. Many studies have indicated that $PM_{2.5}$ ambient concentrations exhibited rather low spatial variation within a geographic domain on each day.[1] However, based on CMAQ predicted concentrations used here, the county and seasonal average concentrations differed by 36% when comparing New York and Bronx counties for each of the winter and summer seasons. Based on Air Quality System (AQS) monitoring data, the county and seasonal average concentrations in AQS data differed by 14% in winter and 3% in summer when comparing New York and Bronx counties. When comparing CMAQ with Air Quality System (AQS) monitoring data, CMAQ tended to under-estimate summer concentrations and to over-predict winter concentrations. Based on analysis of CMAQ results for other years, similar prediction errors were observed. The reasons for CMAQ prediction errors may include improper characterization of the weaker mixing in winter than summer, incorrect temporal emission allocations, and differences in the science process treatments of surface deposition processes, vertical turbulent transport, and secondary aerosol formulation.[13] Since the same ACH distributions were used for these

counties, E_a/C was essentially identical between counties within each season. Thus variations in seasonal average E_a between New York and Bronx counties of 36% were mainly determined by C.

Factors Affecting High-End Daily Average Exposure

The variability in daily average E_a is primarily influenced by spatial variation in C..The day-to-day variation of estimated E_a is highly correlated (r_p=0.8) with C. However, variability in E_a is also influenced by factors such as ACH, P and k among individuals or over time. E_a/C is correlated with ACH (r_p=0.5-0.6). Thus the seasonal difference in E_a/C ratio was associated with differences in ACH. This is related to individual ventilation practices. For example, residents in NYC tend to open windows more in the hot summer than in the colder winter. Lower ACH leads to less infiltration of ambient $PM_{2.5}$, which results in lower E_a/C and lower indoor ambient exposure to $PM_{2.5}$. The daily E_a/C ratio differs by a factor of 4 to 5 over a 95% frequency range among individuals, indicating that some people are very highly exposed compared to others because of factors other than ambient concentration, such as ACH by season or location.

Potential Bias in CMAQ

To assess the CMAQ model performance, CMAQ estimated concentrations were compared with AQS data from FSMs. For long-term annual averages, CMAQ predictions is similar to FSM data, with mean bias of 1.0 $\mu g/m^3$ and normalized mean bias of 7.4% for years 2002 to 2006. However, for short-term daily averages, CMAQ tended to under-estimate summer

concentrations and over-predict winter concentrations. Take the year of 2002 as an example, the bias varied by season over three counties from -25% in summer to 30% in winter, and varied by county over winter and summer, from -13% in Bronx to 10% in New York County. The higher bias in the winter and lower (usually negative) bias in the summer suggested potential issues related to mixing, emission temporal allocations, or other CMAQ model processes or inputs (Tesche *et al.*, 2006).

To compare differences in estimated exposure between using CMAQ and FSM data, Table D-3(b) shows E_a estimated by multiplying FSM concentration with E_a/C ratios derived in Table D-3(a). FSM-based estimates of E_a were 25% lower in winter than summer, compared to CMAQ-based E_a of 30% higher in winter than summer. In addition, FSM-based estimates of E_a were 14% higher for New York County than Bronx County in winter, and 1% higher in summer, compared to CMAQ-based E_a of 36% higher in both seasons. Thus, uncertainty exists in CMAQ concentration estimates, which will further affect exposure model prediction. The appropriateness of using CMAQ to estimate ambient concentration mainly depends on metric of concern, acceptable level of uncertainty, population of interest, and study design (Bravo *et al.*, 2012). CMAQ can be updated with FSM data using Bayesian techiniques to lessen bias when estimating seasonal and spatial variation in C (McMillan *et al.*, 2010).

Limitations

Some potential modeling assumptions that may cause decreased variability in the exposure results are identified and discussed. The input $PM_{2.5}$ concentrations used were from CMAQ

12 km by 12 km grid cell predictions. Thus the variations in concentration caused by possible urban canyon effects or pollutant transport within large buildings on a smaller spatial resolution are not addressed. Near roadway exposures were not well represented by grid cell concentrations, as area-wide ambient concentrations are often much lower near roadways. City-specific activity patterns are not yet adequately addressed. There is not sufficient geographic coverage of diaries in CHAD to specifically represent a given location such as NYC. Thus, estimated exposure results may not fully account for possible geographic or seasonal differences in activity patterns. Furthermore, there is possible bias in CMAQ concentrations, which should be considered when interpreting exposure results.

SUMMARY

In comparison with CMAQ input ambient concentrations, exposure estimates from SHEDS-PM have more variability in daily average values. SHEDS-PM uses information about demographics and activity patterns for the population of interest, resulting in a more complete characterization of population exposure. Results indicate that variation in exposure to pollutants ambient origin is mostly affected by variation in ambient concentration, but is also influenced by other exposure factors. CMAQ performance varied by time interval, season, and location. Not much inter-annual variation in daily average exposure was estimated, as factors affecting exposure such as ACH, housing type and activity patterns were assumed to be relatively stable across years. Seasonal and inter-county variability in CMAQ-based estimates of E_a were not accurate. FSM-based estimates indicate higher E_a in summer than winter, and much less inter-county variability in E_a. However, E_a/C was

unaffected by input concentration selection. Other than C, seasonal difference in E_a was mainly associated with differences in ACH. Some people are highly exposed because of ACH and individual activity patterns.

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Table D-1. Residential microenvironment input parameters

Parameter	**Distribution Type**[a]	**Temperature**	**Value**[b]
Penetration (P)	Triangular	All	Min=0.7, Mode=0.78, Max=1.0
Deposition (k)	Normal	All	$\mu=0.40h^{-1}$, $\sigma=0.1h^{-1}$
Air Exchange Rate (ACH)	Lognormal	Cold (< 65°F)	$\mu_g=0.78$, $\sigma_g=1.94$
		Warm (≥ 65°F)	$\mu_g=1.44$, $\sigma_g=2.14$

[a.] Triangular distribution parameters are the minimum, mode, and maximum; normal distribution parameters are the mean μ and standard deviation σ; lognormal distribution parameters are the geometric mean μ_g and geometric standard deviation σ_g.

[b.] Sources: P, k: Weisel *et al.* (2005),[9] Özkaynak *et al.* (2006);[10] ACH: Jones *et al.* (2012).[11]

Table D-2. Variability in estimated daily average ambient concentration (C), ambient exposure (E_a), and ambient exposure to concentration ratio (E_a/C) by year.

Year	**C**		**E_a**		**E_a/C**	
	Mean (μg/m$^{3)}$	CV	Mean (μg/m^3)	CV	Mean	CV
Overall	15.1	0.69	9.8	0.72	0.65	0.22
2002	15.6	0.65	10.2	0.69	0.66	0.22
2003	16.2	0.68	10.4	0.72	0.65	0.22
2004	14.7	0.69	9.5	0.72	0.66	0.22
2005	14.5	0.69	9.3	0.72	0.65	0.23
2006	14.7	0.72	9.6	0.75	0.66	0.22

Table D-3. Variability in ambient concentration (C), ambient exposure (E_a), and ambient exposure to concentration ratio (E_a/C) by county for winter and summer[a], in year 2002.

(a) CMAQ

	Bronx County				New York County				Queens County			
	Winter		Summer		Winter		Summer		Winter		Summer	
	Mean	CV	Mean	CV	Mean	CV	Mean	CV	Mean	CV	Mean	CV
C, µg/m^3	17.5	0.61	12.3	0.61	23.8	0.56	16.7	0.58	19.1	0.54	13.2	0.58
E_a, µg/m^3	11.0	0.66	8.5	0.64	15.0	0.61	11.5	0.61	12.1	0.59	9.2	0.61
E_a/C	0.63	0.22	0.70	0.20	0.63	0.22	0.69	0.19	0.63	0.22	0.70	0.19

[a.] Winter includes December, January and February; summer includes June, July and August.

(b) FSM

	Bronx County		New York County		Queens County	
	Winter	Summer	Winter	Summer	Winter	Summer
	Mean	Mean	Mean	Mean	Mean	Mean
C, µg/m^3	15.4	18.8	17.6	19.3	13.6	17.6
E_a, µg/m^3	9.7	13.2	11.1	13.3	8.5	12.3
E_a/C	0.63	0.70	0.63	0.69	0.63	0.70

[a.] Winter includes December, January and February; summer includes June, July and August.