ULTRAFINE PARTICLE CONCENTRATION INSIDE VEHICLES:
MODELS FOR EXPOSURE ASSESSMENT

by

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A Dissertation Presented to the
FACULTY OF THE USC GRADUATE SCHOOL
UNIVERSITY OF SOUTHERN CALIFORNIA
In Partial Fulfillment of the
Requirements for the Degree
DOCTOR OF PHILOSOPHY
(ENVIRONMENTAL ENGINEERING)

August 2012

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DEDICATION

To, My Parents
ACKNOWLEDGEMENTS

I would like to acknowledge the guidance and advice received during my graduate student career from:

Dr. Constantinos Sioutas
Department of Civil and Environmental Engineering, University of Southern California

Dr. Scott A Fruin
Department of Preventive Medicine, Division of Environmental Health, University of Southern California
Dr. Katharine F. Moore
Department of Civil and Environmental Engineering, University of Southern California

I would like to specially mention the assistance received from Dr. Sandrah P. Eckel, Department of Preventive Medicine, Division of Biostatistics, University of Southern California, for the development of models and my colleagues at Aerosol Lab, University of Southern California for assisting with field-work.

Lastly, I would like to thank Dr. Ralph Delfino at University of California, Irvine, and co-Principal Investigator for the research grant that funded this work, and Air Resources Board California for funding this work through grants 05-317 and 07-310. I would also like to mention financial support received for roadway sampling through NIEHS grant 1K25ES019224-01.
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ABSTRACT

Despite the proven toxicity of ultrafine particles (UFP), epidemiological studies focusing on them are rare. This is at least in part because accurate exposure assessment for such studies is challenging due to the ubiquitous and dynamic nature of UFP, and successful epidemiological studies will depend on the ability to characterize personal exposure in several key microenvironments. The in-vehicle or in-transit microenvironment is one such microenvironment. This is because for large sections of the population, highest UFP concentrations are often encountered during travel, i.e., where close proximity to relatively undiluted vehicular emissions occurs.

This thesis focuses on developing useful models for linking roadway UFP concentrations to in-vehicle concentrations. Even when roadway concentrations are known, correct assignment of exposure requires an additional parameter, the inside-to-outside UFP ratio (I/O), which reflects the combined effects of multiple mechanisms that either limit the pollutant penetration inside vehicles or cause loss therein. I/O ratios can vary from nearly zero to one, within an individual vehicle, depending on the ventilation condition, fan setting and speed. It also varies widely within a vehicle fleet due to differences in vehicle age, model and manufacturer. However, it is not feasible to measure I/O ratios for thousands of individual vehicles, as is required in large epidemiological studies, hence the need for accurate predictive models.
Results from a test fleet representative of the U.S. fleet in terms of age, mileage, volume and manufacturer distribution, suggest that UFP I/O ratio is strongly dependent on air exchange rate (AER) of the vehicle, which in turn is determined by ventilation mode, ventilation fan setting, driving speed, cabin volume and age of the vehicle. The UFP I/O ratios measured under recirculation (RC) ventilation mode were far lower than those under outside air intake (OA) conditions due to typically an order of magnitude lower AERs under RC mode than under OA mode. Predictive models developed for AER and I/O ratios were able to account over 70% of variation in measured data. All the predictive variable values are easily obtainable through questionnaire, which will be useful to exposure and epidemiological studies.

As a demonstration of model usefulness and scalability to dynamic real world scenarios, UFP I/O ratio distributions were further linked to UFP concentrations measured on-road on multiple routes (freeways and arterial roads) in Los Angeles. In-vehicle concentrations varied by over a magnitude due to inter-vehicle variability and ventilation choice (windows open, RC or OA), and inter-vehicle variation in I/O ratios was often larger than the variability in on-road concentrations. In general, exposure concentration increased two to four fold when ventilation mode was switched from RC to OA. In a real-world setting, 1) ventilation setting, 2) roadway type (arterial or freeway) and 3) the totality of vehicle and driving characteristics (age, manufacturer and speed) each are all of roughly similar impact on in-vehicle UFP concentration.
The research described in this thesis demonstrates that the in-vehicle microenvironment is unique in its extreme variability, but that in-vehicle UFP concentrations are predictable based on easily obtainable information. Furthermore, the key physical mechanism determining in-vehicle concentrations is AER, which too is predictable. Therefore, determining UFP exposures in vehicles, previously a difficulty and key gap in UFP exposure assessment, is now possible. The work can also serve as a basis for other particulate pollutants and reactive gaseous species in in-vehicle and other indoor environments.
CHAPTER 1: INTRODUCTION

1.1 ULTRAFINE PARTICLES: SOURCES AND NATURE

Ultrafine particles (UFP), defined as particles having aerodynamic diameter less than 100 nanometers, are ubiquitous in urban atmospheres given the multiplicity of their sources (Shi et al., 2001). A dominant primary source of UFP in urban areas is fuel combustion emission by vehicles (Shi et al., 2001; Fine et al., 2004), and as much as 80% of emitted particles can be in the UFP size range (Morawska et al., 1998; Shi et al., 2001; Sioutas et al., 2005; Moore et al., 2009). Cass et al. (2002) reported that in South Coast Air Basin that surrounds Los Angeles mobile and stationary fuel combustion sources having the most significant contribution. Percentage contribution of mobile sources to ambient UFP was estimated as 43%, stationary source fuel combustion as 32%, another 10% from non-highway mobile sources and 7% from industrial sources. They further suggest that for those in proximity to trafficked areas, far more than 43% UFP exposure may be of mobile-vehicle emission origin. In addition to primary, or direct, ultrafine particle emissions, photochemical reactions in the atmosphere may also be responsible for the formation of secondary UFP. Long-range transport as well as photochemical particle formation in the atmosphere can significantly increase particle number concentrations downwind of urban areas (Kim et al., 2002; Fine et al., 2004; Hudda et al., 2010).
UFP are dynamic and transient, undergoing simultaneous transformations via multiple mechanisms like dilution, condensation and evaporation, nucleation, turbulent transport, deposition and heterogeneous chemical reactions (Zhang et al., 2004a). As a consequence of rapid processing, some UFP have short lifetimes and concentration gradients can be strong (Sioutas et al., 2005). Furthermore, these mechanisms not only change the numerical concentration, but also the nature of the ultrafine particles (Zhang et al., 2004b). Zhu et al. (2002a, 2002b) along with numerous other studies (Westerdahl et al., 2005; Chan et al. 1991; Duffy and Nelson 1997) have shown that UFP concentrations in vicinity of freeways are significantly higher, up to 10 times the ambient levels, and typically decay exponentially with distance from the freeways.

1.2 Ultratine Particles: Health Effects and Epidemiological Study Needs

Numerous recent studies investigating associations between particulate pollution and health effects have attributed higher risk to ultrafine particles on per mass basis (Oberdörster et al., 1995; Donaldson et al., 1998; Delfino et al., 2005; 2009) compared to particles of greater diameters. Even though the exact mechanism remains under investigation, oxidative stresses caused by UFP are believed to be involved (Cho et al., 2005; Delfino et al., 2005; Sioutas et al., 2005). In-vitro toxicological studies have also shown that UFP have higher oxidative potential and can penetrate and destroy mitochondria within epithelial cells (Li et al., 2003) and have the highest associated reactive oxygen species amongst particle size fractions (Cho et al., 2005). Given the large
numbers, surface areas and high pulmonary deposition efficiency of UFPs, they have the potential for being carriers of toxic air pollutants and redox active compounds to cardiovascular system (Delfino et al., 2005). Recently based on an expert panel study, Knol et al. (2009) reported that the “highest rated likelihood pathway” involved respiratory inflammation and subsequent thrombotic effects.

Despite sufficient evidence of UFP toxicity, epidemiological evidence for health effects of UFP is limited and no studies exist that examine long-term exposure (Hoek et al. 2010). The few epidemiological studies that do exist have examined short-term exposures’ association with acute respiratory response and lung function (Ibald-Mulli et al., 2002), and only two studies have linked UFP to daily mortality and hospital admissions (Wichmann et al., 2000; Stozel et al., 2007). No clear dose-response relationship yet exists for health impact assessment, partly due to the difficulty of accurately assessing exposure concentrations, possible differences in UFP toxicity by source, and a lack of long-term studies. Hoek et al. (2010) quantified effect of UFP on all-cause mortality and reported a 0.43% decrease in mortality for a 1000 particle per cubic centimeter decrease in UFP concentration, but underscored the existence of high uncertainty. Given the weight of toxicological evidence and ubiquity of high UFP exposures in urban areas and near combustion sources like traffic, there exists an important need to assess the health risks posed by UFP. This requires filling the
exposure assessment gap – through accurate determination of exposure concentrations in a multitude of microenvironments.

1.3 Ultrafine Particles: Exposure Assessment

Accurate exposure assessment for UFP is challenging due to several reasons. Firstly, due to their dynamic nature, concentration prediction or measurement is challenging, with direct measurements requiring intensive and impractical numbers of samples for large cohorts in epidemiological studies. Secondly, exposure assessment not only requires time-activity information but also direct or indirect estimates of concentrations in all key microenvironments. Ubiquitous presence of UFP and highly variable concentrations within some key microenvironments such as inside vehicles, make this task challenging. However, accurate assessment of risk will depend on the ability to characterize individual exposure within these very key and highly variable microenvironments; where high and disproportionate exposure occurs for large sections of the population. There is also a need to determine how large, spatially dispersed populations experience UFP exposures over prolonged periods of study, i.e., the need to determine community level exposures (Sioutas et al., 2005).

1.4 Importance of In-Vehicle Microenvironment

A microenvironment can be defined as an environment of homogenous pollutant concentration for the given period of time. The ‘microenvironmental’ approach has
been in practice since 1972 when it was proposed by Fugas, M. (Fugas, 1976) and has since been refined by various researchers (Duan, 1982; Ott, 1981; 1982a; 1982b). The importance of a microenvironment in terms of contribution to total exposure will depend on the concentration difference observed between the microenvironment and the ambient, as well as the time spent within it (Ott, 1982a).

Given the rapidly decaying and reactive nature of ultrafine particles (UFP, aerodynamic diameter less than 100 nanometer), the exposure to UFP in microenvironments that are nearer to the source can form a significant fraction of the total dose. Thus, in terms of assessing exposure, or linking the emissions to inhalation, microenvironments placed at two extremes in terms of both concentrations and time spent are of special interest.

At one of these extreme is the need to determine how large, spatially dispersed populations experience ultrafine exposures over prolonged periods of study, i.e., the need to determine community level exposures (Sioutas et al., 2005). Klepeis et al., (2001) reported in National Human Activity Pattern Survey (based on more than 9000 respondents) that on an average in U.S., people spend 68.7% of their time in indoor residential setting (total time spent indoors was 86.9%). For California, the numbers are similar. On an average in California, 983 minutes in a day are spent indoors in a residence compared to 996 nationwide. In another study conducted by Jenkins et al., (1992) focusing on California, it was reported that 62% time was spent indoors in residences. Results from this study are summarized in Figure 1.1.
Figure 1.1: Time spent in different microenvironments

At the other extreme is enclosed transit environment. On an average 95 min per day are spent in the in-vehicle microenvironment, about a tenth of the residential microenvironment (Klepeis et al. 2001). Jenkins et al., (1992) reported that Californians spend 7% time in enclosed transit. This, when compounded by the predictions of increasing commuting times in Los Angeles, (Southern California Association of Governments predicts that commuting times will double by 2020 due to population growth in the Los Angeles (LA) area (SCAG 2003)), underscore the need for assessment of personal exposure to UFP in vehicular microenvironment.
Fruin et al. (2008) calculated that 33-45% of UFP exposure occurs while driving in Los Angeles, taking other micro-environmental concentrations and time-activity patterns into account, but ignoring in-vehicle particle losses. Zhu et al. (2007) suggest approximately 10–50% of total daily exposure to UFP from traffic occurs within the in-vehicle microenvironment. In suburban locations of less traffic and the most comprehensive study identified, Wallace and Ott (2011) estimated a 17% contribution of in-vehicle microenvironment to total UFP exposure.

Though studies attempting in-vehicle exposure assessment are few, certain observations can be drawn. Firstly, there is disparity among the percentage contribution of in-vehicle microenvironment to the total exposure, which goes beyond the variation expected due to roadway environment differences and differences in time in transit. This disparity arises in part due to inaccurate assumptions made about in-vehicle to roadway concentration (I/O) ratios for vehicles. For example, Zhu et al. (2007) assumed it to be 0.6, despite reporting significantly different I/O ratios even in the limited results from three vehicles. Fruin et al. (2008), for simplicity, assumed it to be one, such as is the case when vehicle windows are open. Such assumptions fail to fully capture an important source of in-vehicle UFP variability or bias exposure estimates upward. Secondly, ascertaining the percentage exposure concentration in in-vehicle (or any other microenvironment) requires a priori knowledge of concentration in all or at least the important microenvironments. Wallace and Ott (2011) assessed personal exposure through extensive measurements, but their results, though otherwise accurate, cannot
be generalized widely because the range of variation of UFP concentrations on subject-to-subject basis is very wide, specially so for dynamic environments like in-vehicle or residential.

1.5 Research Objectives

The main objective of this research work was to develop models that can be used to estimate in-vehicle UFP number concentration so that accurate exposure assessment in this microenvironment becomes possible. A parameter defined as inside-to-outside ratio (I/O, which is simply the ratio of in-vehicle to roadway UFP concentration), if quantified and characterized for real-world conditions, can be used to calculate in-vehicle concentration for given roadway concentration. Towards this end, the research was carried out meeting the following objectives:

i. Prioritize the influence of vehicle related factors such as age, mileage, manufacturer, type, etc., on I/O ratio in a set of carefully selected vehicles that are positioned at the ends of normal range for these variables.

ii. Make particle-size resolved measurements to establish if the effect of aforementioned factors were comparable across the UFP size range of interest.

iii. Characterize the influence of driving related factors such as speed, ventilation setting and fan strength preference on I/O ratios, in addition
to the important vehicle related variables in a large and representative set of vehicles.

iv. Quantify these various influences on a dependent variable – air exchange rate (AER), commonly used in mechanistic models.

v. Develop empirical models for both AER and I/O ratios based on variables for which information can be gathered via survey questionnaires in large epidemiological studies.

vi. Demonstrate the scalability and usefulness of these models in real-world dynamic conditions.

1.6 Thesis Layout

Chapter 2 describes the methods used to measure AERs, describes the vehicles tested for this work, and presents the predictive models for AER. Chapter 3 discusses the influence of various factors, both vehicle and driving related, that impact the I/O ratios. Chapter 4 presents predictive models for I/O ratios. Chapter 5 examines the influence of real-world variation in input predictive variables, and demonstrates the scalability of the models to dynamic roadway environment at fleet-wide level. Chapter 6 summarizes the principal findings and applications of this work, and discusses gaps in extant literature on UFP exposure assessment and future work.
CHAPTER 2: PREDICTIVE MODEL FOR VEHICLE AIR EXCHANGE RATES BASED ON A LARGE, REPRESENTATIVE SAMPLE

2.1 INTRODUCTION

Large sections of population are exposed to highest concentrations of fuel combustion related pollutants during travel, i.e., when in closest proximity of undiluted emission. Roadway concentration of traffic-related pollutants is typically an order of magnitude higher than urban ambient concentrations (Chan et al., 1991; Westerdahl et al., 2005). Given the high concentration level in this microenvironment, even a small amount of time such as the average 80 min per day spent by people in the U.S. in the in-vehicle microenvironment contributes disproportionately to total exposure (Klepeis et al., 2001). Jenkins et al. (1992) reported that Californians spend 7% of their time or 100 minutes in enclosed transit. In order to accurately assess total exposure, this microenvironment needs to be correctly accounted, which requires knowledge of pollutant concentration on roadways, as well as fraction thereof that gets inside the vehicle cabin.

Exchange of air between the vehicle cabin and roadway, quantified as air exchange rate (AER, the number of times vehicle cabin air is replaced by roadway air in an hour) drives the influx of pollutants into the cabin. Pollutant concentration inside a vehicle generally matches the roadway concentration when there is sufficiently high air turnover between the vehicle and roadway. This occurs whenever windows are open, often when
outside air is drawn into the vehicle through the ventilation system, or when the vehicle is sufficiently leaky. However, under conditions that limit exchange of air between the vehicle cabin and roadway, there can be significant reduction in particle mass and number may occur due to losses to vehicle’s internal surfaces (Ott et al., 1992). Such conditions usually only occur for newer cars, for which door seals and insulation are tightest, and/or at low speeds where airflow dynamics are not producing large differences in pressure around the vehicle.

If the air exchange rate (AER) of a vehicle is known, the particle losses can be estimated; however, AERs are usually not known, and are highly variable even for the same vehicle, as they vary widely with speed (Knibbs et al., 2009; Ott et al., 2008; Rodes et al., 1998). For example, Knibbs et al. (2009) found AERs to vary from 1 to 33 air changes per hour (h\(^{-1}\)) across six cars at a speed of 37 miles h\(^{-1}\). AERs further vary by up to a magnitude for the same vehicle depending on the ventilation setting. Knibbs et al. (2009) showed that even at lowest fan settings, AERs under outside air intake ventilation mode were typically a magnitude higher than those under recirculation setting. Ott et al. (2008) observed that opening the windows by just 3 inches increased AERs 8–16 times.

Few studies have characterized AERs. The largest study to date has been Knibbs et al. (2009) who measured AER using SF\(_6\) as a tracer gas for six vehicles spanning an age range of 18 years at various speeds and under different ventilation settings. In addition to the findings cited above for speeds of 37 miles h\(^{-1}\), they found AERs to range from 2.6
to 47 h\(^{-1}\) (mean: 18) at 68 miles h\(^{-1}\). They also tested stationary vehicles and reported AERs within 0.1–3.3 h\(^{-1}\) range with five cars having AERs < 1 h\(^{-1}\). Ott et al. (2008) reported AERs in the range of 1.6 to ~40 h\(^{-1}\) for four vehicles up to six years old and at speeds ranging from 20 to 72 miles h\(^{-1}\) using CO as a tracer gas. They also provide an excellent review of previous studies on the subject. Other than the work by Fletcher and Saunders (1994), Rhodes et al. (1998), Kvisgaard and Pejtersen et al. (1999), Ott et al. (1992; 1994; 2008), Batterman et al. (2006) and Knibbs et al. (2009) – a total of 16 cars tested – others have only tested AERs in stationary vehicles, and not during on-road conditions, where most of the travel time exposure occurs.

Because vehicle AER varies more than an order of magnitude among vehicles, a large sample number is necessary to fully characterize vehicle AERs. Further, as it is impractical to measure either AER for large numbers of subjects’ vehicles as required in an epidemiological study addressing drive-time exposure, predictive models are needed for estimating AER. The purpose of this study was to test a sufficiently large number of cars in order to develop robust predictive models of AER under both outside air intake (OA) conditions and recirculation (RC) conditions as a simple function of readily available information, such as vehicle age, mileage, manufacturer, and average speed.

In this chapter, results are presented from AER measurements conducted in 67 vehicles, chosen to represent the California fleet with regard to age, vehicle type, and manufacturer. These results more than quadruple the number of vehicle AERs reported
in the literature and provide for the first time a sample of vehicles that is large enough to be considered reasonably representative of the current fleet of California vehicles or perhaps U.S. vehicles. This work also demonstrated that Carbon dioxide can be used to calculate vehicle AER, and it is a relatively straightforward and accurate alternative to the use of other tracer gases, which require specialized measurement instruments. The ease of this method enabled the testing of large number of vehicles.

2.2 METHODS

2.2.1 VEHICLE SELECTION

Vehicles were selected to approximate the distribution of the California fleet in terms of vehicle size type (e.g., subcompact, compact, midsize, etc.), mileage, and age. Vehicle size data were based on the data set of the 2002 report by the California Department of Motor Vehicles to the California Air Resources Board in support of their mobile source Emission Factors model (EMFAC) database), the latest available at the time of initial study design. Data on fleet mileage and age were based on 2009 data. Target numbers of test vehicles for each size category were calculated based on the frequency of these size categories multiplied by the fraction of the fleet that was 5 years old or newer (30%), 6–14 years (53%), and 15 years or older (17%). Within these categories, an attempt was also made to select vehicles from the models having the largest sales in California (e.g., Toyota Corolla, Honda Civic, etc.) but there were no specific requirements by manufacturer. All vehicles tested are listed in the Table 2.1.
### Table 2.1: List of vehicles tested in the study.\(^1\)

<table>
<thead>
<tr>
<th>Manufacturer</th>
<th>Model</th>
<th>Year</th>
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<th>Model</th>
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<td>Honda</td>
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<td>Chrysler</td>
<td>300*</td>
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<td>Honda</td>
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\(^1\) Ultrafine particle measurements were conducted in the models marked with *
2.2.2 *INSTRUMENTS*

Carbon dioxide (CO$_2$) was measured both inside and outside the vehicle simultaneously using two TSI Q-Traks, model 7565 (TSI Inc., MN, USA) and one LI-COR Li-820 units (LI-COR Biosciences, NE, USA). Both units use a non-dispersive infrared detection technique, but the Li-820 unit is pump driven, thus allowing a faster response time than the Q-Trak unit, e.g., several seconds versus 20 seconds. Table 2.2 provides more details about the instruments and their settings. All instruments used for a test were run simultaneously and ambient concentrations before and after a run were checked for consistency. An on-board GPS device (Garmin GPSMAP 76CSC) recorded the location and speed of the car at one second intervals. All instruments were synced to within one second of the time recorded by GPS.

Table 2.2: Instruments used for the study.

<table>
<thead>
<tr>
<th></th>
<th>TSI Q-TRAK 7565</th>
<th>LICOR LI-820</th>
<th>Garmin GPSMAP 76CSC</th>
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<tr>
<td><strong>Range</strong></td>
<td>0-5000 ppm</td>
<td>0-20000 ppm</td>
<td></td>
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<tr>
<td><strong>Resolution</strong></td>
<td>1 ppm</td>
<td>0.1 ppm</td>
<td>0.1 mph</td>
</tr>
<tr>
<td><strong>Accuracy</strong></td>
<td>±3% of reading or ±50 ppm, whichever is greater</td>
<td>&gt;4% of the reported value</td>
<td>0.05 m/sec</td>
</tr>
<tr>
<td><strong>Response Time</strong></td>
<td>20 seconds</td>
<td>-</td>
<td>1 sec</td>
</tr>
<tr>
<td><strong>Logging Interval</strong></td>
<td>10 seconds</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td><strong>Averaging Interval</strong></td>
<td>10 seconds</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td><strong>Calibration Frequency</strong></td>
<td>~ 50 hours of operation</td>
<td>Factory Calibration before and after the sampling campaign</td>
<td>-</td>
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<tr>
<td><strong>Time-Sync Frequency</strong></td>
<td>~ &lt; 10 hours of operation</td>
<td>At the beginning of each car tested</td>
<td>-</td>
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</table>
2.2.3 **Carbon Dioxide as a Tracer Gas**

CO₂ was chosen as a tracer gas for its low toxicity, ease of measurement, and its ready availability when using car occupants as the source. At a fixed vehicle speed (and hence fixed AER), in-vehicle CO₂ concentrations change until an equilibrium concentration is reached whereby the production of CO₂ from vehicle occupants is balanced by the losses of CO₂ due to replacement of high CO₂ concentration air within the vehicle with low CO₂ concentration from outside. This difference is typically hundreds or thousands of parts per million (ppm) of CO₂, so it is easy to measure with high relative accuracy. We achieved well-mixed conditions with a fan inside the vehicle. This was verified for each test by checking agreement with Q-Trak and Li-820 instruments placed in different locations within the vehicle, i.e., the front and back seats of the vehicle.

2.2.4 **Speed and Routes Driven**

Routes were carefully chosen to allow nearly constant speeds for a time period sufficiently long to acquire stable AERs and in-vehicle concentrations. To achieve constant speeds of 55 miles h⁻¹, freeways were driven during conditions of free-flowing traffic. To achieve constant speeds of 30–35 miles h⁻¹, runs were either made in a large cemetery or a continuous loop around the Rose Bowl in Pasadena. Both of these routes allowed fairly short laps to prevent long duration in one direction, thus canceling any effect of wind direction and velocity on AER. Furthermore, there was minimal vehicular traffic on the roads at both locations during the times the tests were conducted. This minimized changes in outside CO₂ due to the presence of exhaust plumes from other
vehicles. Finally, to incorporate measurements under non-steady speeds in this modeling data reported by Knibbs et al. (2009; 2010) from Sydney (Australia) was included in the data set.

2.2.5 MATHEMATICAL EQUATIONS AND ASSUMPTIONS UNDER RECIRCULATION CONDITIONS

AER increases whenever pressure differences and/or turbulence around the vehicle increases, such as caused by increase in driving speed. However, for a given vehicle speed (strictly speaking, the pressure difference between the cabin and roadway and air speed around the vehicle shell), the AER is nearly constant and the CO\textsubscript{2} concentrations inside the vehicle will eventually reach an equilibrium value. But until the equilibrium is reached, the mass balance equation, Equation 1, applies

\[
\left( \frac{dC_{in}}{dT} \right) V = S + (C_{amb} - C_{in})(AER_s)V \quad \text{or}
\]

\[
\left( \frac{dC_{in}}{dT} \right) = \frac{S}{V} + (C_{amb} - C_{in})(AER_s) \quad \text{... (1)}
\]

where \( S/V \) is the vehicle-volume-specific source strength in ppm per hour, \( C_{amb} \) and \( C_{in} \) are the outdoor and in-vehicle CO\textsubscript{2} concentrations (ppm), respectively, and AER\( s \) is the speed and vehicle-specific air exchange rate (h\textsuperscript{-1}).

At equilibrium, Equation 1 becomes

\[
\left( \frac{dC_{in}}{dT} \right) = 0 = \frac{S}{V} + (C_{amb} - C_{eq})(AER_s) \quad \text{... (1.a)}
\]

which can be rewritten as

\[ AER_s = (S/V)/(C_{amb} - C_{eq}) \quad \text{... (1.b)} \]
Assuming a small rate of air exchange when the car is stationary, with interior air well mixed, the vehicle-specific source term can be determined by the initial build-up rate of CO$_2$ when inside and outside CO$_2$ concentrations are similar, i.e., the ((C$_{amb}$ - C$_{in}$)AER) term in Equation 1 is much smaller than the S/V term. For example, for <10 ppm difference in inside versus outside CO$_2$, and an AER of 2 h$^{-1}$, the ((C$_{amb}$ - C$_{in}$)AER) term is 20 ppm per hour per unit volume change, compared to a typical build-up rate of 15000 ppm per hour per unit volume for two occupants, or less than one percent. Under these conditions, Equation 1 becomes:

$$\left( \frac{dC_{in}}{dT} \right) \approx \frac{S}{V}$$

Or, any change in-cabin concentration results from the release of CO$_2$ by occupants and equals the volume normalized source strength.

2.2.5.1 Determination of Source Strength

The CO$_2$ source strength was determined by measuring the build-up rate of CO$_2$ from two occupants inside the vehicle when the vehicle was first sealed and the inside CO$_2$ concentrations were close to ambient concentrations. At the start of the test, any small rates of air exchange had little effect on inside CO$_2$ concentrations since the inside and outside CO$_2$ concentrations were similar. Furthermore, at this time the CO$_2$ concentration build-up is very linear and it is easily and accurately determined as the slope of the CO$_2$ concentration versus time (See Equation 1.c). Eventually, as the inside CO$_2$ concentrations reach high levels, build-up rates slow down and become nonlinear,
but this typically requires at least several minutes. Since physical activity before, during, and after the runs was minimal, the CO\(_2\) source strength reflected the resting (inactive) metabolism rates of the occupants and was therefore constant for the hour or two of the measurements. CO\(_2\) build-up rates were measured before and after the run in 10 vehicles.

2.2.5.2 DETERMINATION OF EQUILIBRIUM CONCENTRATION

Equilibrium CO\(_2\) concentrations were determined for constant speeds of 20, 35, and 55 miles h\(^{-1}\) with windows closed, ventilation set to air recirculation, and the fan setting set to either midway/medium or the closest possible to a midway setting. For a smaller subset of vehicles, AER was also determined for stationary vehicles and at speeds exceeding 65 miles h\(^{-1}\). Equilibrium CO\(_2\) concentrations were determined when the maximum fluctuation in in-cabin concentration was less than 50 ppm for at least the last 10 minutes at the test speed. For the median equilibrium value recorded in this study, 50 ppm fluctuation corresponded to 2.1%.

2.2.5.3 CO\(_2\) EQUILIBRIUM CRITERIA

Time series plots of speed and CO\(_2\) were aligned and adjusted to take into account any differences in instrument clock time or response time. Alignments were made based on events that caused a rapid concentration change, such as opening a window opening that rapidly reduced in-vehicle CO\(_2\). Where the in-vehicle CO\(_2\) concentration met the less than 50 ppm change criterion for a given speed, the exact equilibrium concentration
was determined at the time where CO₂ concentrations had less than 2% standard deviation for at least 20 data points (i.e., more than three minute of data). Concurrent outside CO₂ concentration was then subtracted (See Equation 1.b). For the 20 and 35 miles h⁻¹ speed, the outside CO₂ concentrations at both the Rose Bowl and the cemetery were very stable, but the outside CO₂ concentrations on freeways for the 55 miles h⁻¹ condition were not. Therefore, freeway CO₂ concentrations were averaged over the previous two minutes for each equilibrium value chosen.

2.2.6 **Mathematical Equations, Assumptions and Measurements under Outside Air Intake Conditions**

It was consistently observed that when the ventilation mode was set to intake outside air or the windows were open, AER was extremely high. To characterize these high AERs, we measured AER in eight stationary vehicles. Since AERs were much higher, they required higher starting CO₂ concentrations that cannot be readily reached by occupants. CO₂ from a pressurized cylinder (industrial grade) was used to produce the necessary high starting concentrations. CO₂ concentrations exceeding 12000 ppm were built inside the vehicle cabin and then CO₂ supply was cut-off. An exponential drop in CO₂ concentration under such a condition occurs due to continual injection of air from outside (having lower concentration) in to the cabin by the vehicle’s ventilation system. This physical phenomenon can be mathematically expressed as Equation 2.

\[ C_{in,t} = (C_{in,0} - C_{amb}) e^{-(AER)t} \]  \hspace{1cm} \text{(2)}
where $C_{in,t}$ is the in-cabin concentration at a time $t$ and $C_{in,0}$ is the in-cabin concentration at the beginning of the concentration decay experiment. On rearranging Equation 2 becomes:

$$\ln\left(\frac{C_{in}}{C_{in,0} - C_{amb}}\right) = -(AER)t \quad \ldots (2. a)$$

The rate of change of natural log of ratio of in-cabin concentration to the difference of initial and ambient concentration is equal to the AER at this ventilation mode. AER was measured at all possible ventilation fan settings in a vehicle while it was stationary. A term, fan fraction, was then defined as the ratio of the selected fan setting to the total number of options for fan setting to quantify the fan setting for the models. For example, if a vehicle had seven fan setting options and was operated at the third strongest option, the fan setting was set to $3/7$ (or, $= 0.43$). Measurement at each fan setting was repeated three times and the standard deviation was less than 5% amongst repeated measurements and the average of these three tests was used for further analysis. Mobile tests were not possible at such high concentrations of CO$_2$. To reflect the effect of speed, data from Knibbs et al. (2009; 2010) was added to the data set.

2.2.7 Predictive Model

Results for the 67 vehicles tested (and additional six vehicles tested in Sydney), generally at three different speeds, were modeled to test the predictive power of the following candidate independent variables: ventilation fan (fraction of maximum setting), vehicle age (years), mileage (thousands of miles), speed (miles h$^{-1}$), interior volume (ft$^3$), and the
product of coefficient of drag \( (C_d) \) and frontal area \( (m^2) \) along with pair-wise interactions between vehicle speed, age, and fan setting, and between \( C_d \) and frontal area of the vehicle. Squared and cubed terms for mileage, age, and speed were included to account for any nonlinear effects. Manufacturer variables which included specific vehicle manufacturer categories such as Ford, GM, Toyota, Nissan, Honda, and 'other' as well as broader categories such as U.S. and non-U.S. or U.S., Japan, and 'other' were also tested. Vehicles were also grouped by the source of the vehicle (i.e., CARB, rental agency, or student volunteers) and tested for differences. Speed was included; both as a predictive variable and as a stratifying variable, i.e., data were analyzed separately for a given speed. Because AER results had a strong rightward skew, a natural log transformation was used. Predicted values on the original scale can be recovered using the equation \( AER = \exp(\ln AER) \).

AER was strongly related to vehicle speed in particular under recirculation conditions. However, even after adjusting for speed, repeated measurements of AER on the same vehicle have some degree of correlation. For example, a leaky vehicle will consistently show higher AERs than average across all speeds. This violates the assumption of independent observations in multiple linear regression (MLR) models. To account for the presence of unknown within-vehicle correlation, generalized estimating equation (GEE) models (Liang et al., 1986) were used. MLR models were also fit to compare results across modeling techniques.
Parsimonious GEE and MLR models were obtained by backward stepwise selection in which variables were retained if they improved $R^2$ (MLR) or were statistically significant (GEE) at $p = 0.05$ value. Residuals from both models were inspected to check model assumptions. $R^2$ was calculated for the GEE model by taking the square of the Pearson correlation coefficient between observed and model-predicted values of natural log transformed AER. Model fit was assessed by adjusted $R^2$ and by leave-one-vehicle-out cross-validated adjusted $R^2$, which provides a more reliable estimate of the predictive ability of the same model fit to a new dataset containing information on different vehicles. For each model, significance of an indicator variable for Sydney data was also evaluated. Residuals were inspected to assess model assumptions of linearity, normality, and homoskedasticity.

2.3 RESULTS AND DISCUSSION

2.3.1 VEHICLES TESTED

Achieving a representative sample of vehicles for testing was a primary objective of this study since representativeness enhances the utility of predictive models of AER. We selected 67 vehicles to represent the U.S. in terms of vehicle age and size type based on Environmental Protection Agency (EPA) classes as shown in Figure 5.3. The age distribution of the cars tested is presented in Figure 5.4 against a background of the age distribution of the U.S. fleet.
2.3.2 *CONSISTENCY OF CO₂ BUILD–UP RATES*

Build up rates varied from 3100-26200 ppm hr⁻¹, with the median being 13400 ppm hr⁻¹. Figure 2.1 shows the range of measured CO₂ build-up rates for the most frequent combination of occupants. (Build-up rate varies for different combinations of occupants since CO₂ production will depend on a person’s metabolism, body size, etc.) The inverse relationship between build-up rate and vehicle interior volume is as expected for constant source strength. Differences in source strength for a given person from day to day were observed to be small. Repeated tests of build-up rate conducted across different days showed excellent consistency, as shown in Figure 2.2 for ten vehicles. Repeated tests agreed with each other to within 2%.

![Figure 2.1: Car volume versus observed Build-up rates for a subset of data.](image)
2.3.3 AER RESULTS AND UNCERTAINTIES

Figure 2.3: Typical time-series plot for runs conducted at Cemetery along with the initial build up and freeway run under recirculation mode. Average speed during Freeway run was $56 \pm 6$ miles h$^{-1}$ for stable portion highlighted in black). The second black highlight corresponds to stable values during $32 \pm 6$ miles h$^{-1}$ and $20 \pm 3$ miles h$^{-1}$ speed runs.
Figure 2.4: Typical time-series plot for CO₂ concentration decay inside vehicle cabin under outside air ventilation mode.

A typical time-series plot of in-vehicle and outside CO₂ concentration and speed is shown in Figure 2.3. As shown in this plot, the CO₂ build-up rate at the beginning of the test is quite linear. Figure 2.3 also depicts how the various in-vehicle CO₂ concentrations at different speeds show an underlying logarithmic change that eventually reaches a steady equilibrium concentration despite the small irregularities in speed under recirculation ventilation mode. In Figure 2.3, the percent standard deviations of the in-vehicle CO₂ concentration were 1.0, 1.5, and 1.1% at 20, 35, and 55 miles h⁻¹, respectively, while the outside CO₂ concentration standard deviations were 4, 7, and 1.4%, respectively. The resulting AER at 55 miles h⁻¹ was 13.6 h⁻¹. For the in-vehicle CO₂ concentration deviation equal to 1.1% (13 ppm), the resulting AER values range from 13.4–13.8, or standard deviation equals ±1.5%. Similarly, the change in AER values for
35 and 20 miles h\(^{-1}\) due to observed deviations in inside CO\(_2\) concentrations were ± 2% and 1%, respectively. The equilibrium CO\(_2\) concentrations were repeatable to within 2%. The Q-Trak and Li-820 manufacturer stated accuracies were (3% and 4%, respectively, in the range of measurements taken. Root-mean-square error propagation resulted in a maximum CO\(_2\) build-up rate uncertainty of 5.2%, an equilibrium concentration maximum uncertainty of 4.5%, and an AER maximum uncertainty of 7.5%. The highest uncertainty was for 35 miles h\(^{-1}\), due to challenges in maintaining this speed on arterial roads.

Typical decay curve for in-cabin CO\(_2\) concentration during outside air intake ventilation conditions are shown in Figure 2.4 for three fan settings (low, medium and high in a Ford Contour 1999 model that only had three options for fan setting). Root-mean-square error propagation resulted in a maximum uncertainty of 6.8% in AER determination. Up to first 180 three minutes of data were used to determine decay rates and AERs by fitting a least square regression to the data with \(r^2\) for linear fit exceeding 0.98.

Figure 2.5 shows the results for all cars tested at each speed. Under recirculation (RC) conditions, the large vehicle-to-vehicle differences are readily apparent, as is the strong dependence of AER on speed. The median AER in 73 vehicles under the RC setting was 6.0 h\(^{-1}\) (inter quartile range: 3.6-10.3 h\(^{-1}\)). Under outside air (OA) settings, AERs were up to a magnitude higher, and the median AER was 63 h\(^{-1}\) under OA conditions (inter
quartile range: 47-83 h\(^{-1}\)). A strong positive correlation with the fan setting (average \( r^2 = 0.99\) was observed at OA mode. However the effect on fan setting on AER under recirculation was not as strong and varied with the vehicle. The effect of fan setting on AER under RC mode was tested for nine vehicles which spanned most of the range of measured AERs. The average increase in AER was 13% at 20 miles h\(^{-1}\) and 13.7% at 35 miles h\(^{-1}\). These results are shown in Figure 2.6, where AERs at full and medium fan setting for both 20 and 35 miles h\(^{-1}\) speed are plotted.

Figure 2.5: AER results for all 67 vehicles tested.
2.3.4 **Predictive Model for \( \text{Ln(AER)} \) at RC and OA Setting**

The GEE model gave the following Equations 1 and 2 for predicting \( \text{Ln(AER)} \) under RC and OA condition, respectively.

**Equation 1: \( \text{Ln(AER)} \) under RC conditions**

\[
\text{Ln}(\text{AER}) = 2.79 + (0.019 \times \text{speed}) + [0.015 \times \text{age} - 3.3 \times 10^{-3} \times \text{age}^2]
+ [-0.023 \times \text{vol} - 6.6 \times 10^{-5} \times \text{vol}^2] + \text{Manuf Adjustment}
\]

where the manufacturer adjustment is -0.71 for German vehicles and -0.39 for Japanese vehicles. If the speed is zero, a -0.51 factor should be added.
Equation 2: $\ln(AER)$ under OA conditions

$$
\ln(AER) = 4.20 + [(1.88 \times \text{fan strength}) + (-0.92 \times \text{fan strength}^2)] \\
+ (0.0048 \times \text{speed}) + (-0.0073 \times \text{vol})
$$

where the coefficients for fan strength and fan strength$^2$ should be 0.40 and 0.13, respectively, at zero speed, and the speed coefficient should be -0.32 at zero speed.

These models had good fit and are likely to have a similar ability to predict AER in different datasets (GEE adjusted $R^2$: 0.68 for RC and 0.79 for OA; GEE cross-validated adjusted $R^2$: 0.60 for RC and 0.73 for OA). MLR and GEE regression coefficients were similar, but the GEE confidence intervals (which appropriately account for the correlation of repeat measurements on the same vehicles) were roughly a third larger than MLR intervals (Tables 2.3 and 2.4). In all AER model runs, an indicator variable for Sydney data was not significant. Predicted $\ln(AER)$ versus measured $\ln(AER)$ values have been plotted in Figure 2.7 and Figure 2.8 for RC and OA conditions, respectively.

The predicted AER under RC conditions is plotted against the two most significant determinants of AER, speed and age, in Figure 2.9 (a). Model results suggest that an 11 year old vehicle (~75$^{th}$ age percentile) has an AER that is about 1.5 times higher than that of a 4 year old vehicle (~25$^{th}$ age percentile). Furthermore, AER during typical freeway driving speed (65 miles h$^{-1}$) is expected to be 1.8 times higher than under typical arterial driving conditions (35 miles h$^{-1}$).
The two surfaces plotted in Figure 2.9 (a) represent the extremes of other model inputs under RC conditions: a U.S. manufactured sub-compact vehicle (85 ft$^3$ cabin) and a German manufactured large vehicle (120 ft$^3$ cabin), the range of AER variation that can be expected due to manufacturer and volume. A U.S. manufacturer’s vehicle is expected to have an AER nearly 50% higher than a Japanese vehicle and about twice as high as a German manufactured vehicle, for given cabin volume, age and speed. It is interesting that cabin volume was negatively than correlated with AER when expressed in units of air changes per hour under RC conditions.

Figure 2.7: Predicted values for AER plotted against two most significant variables under RC and OA ventilation modes.
Table 2.3: AER under RC Model Coefficients, Confidence Intervals, and p-Values

|                  | Estimate | Std.err | Wald     | Pr(>|W|) | Confidence Intervals |
|------------------|----------|---------|----------|----------|----------------------|
| Intercept        | 2.79     | 0.36    | 62       | 4.10E-15 | 2.1                  |
| speed > 0 (miles h⁻¹) | 0.019    | 0.0013  | 223      | < 2e-16  | 0.017                |
| speed = 0        | -0.51    | 0.12    | 19       | 1.60E-05 | -0.75                |
| age (yr)         | 0.015    | 0.031   | 0.24     | 0.62     | -0.046               |
| age² (yr²)       | 0.0033   | 0.0017  | 4.0      | 0.045    | -                    |
| vol (ft³)        | -0.023   | 0.0049  | 21       | 4.00E-06 | -0.033               |
| vol² (ft⁶)       | 0.000066 | 0.000015| 18       | 1.90E-05 | 0.000037             |
| Manuf: Japan     | -0.39    | 0.12    | 11       | 0.00091  | -0.63                |
| Manuf: Germany   | -0.71    | 0.25    | 8.1      | 0.0045   | -1.2                 |
| MLR              |          |         |          |          |                      |
| Intercept        | 2.97     | 0.28    | 10.75    | < 2e-16  | 2.43                 |
| speed > 0 (miles h⁻¹) | 0.018    | 0.0020  | 8.93     | < 2e-16  | 0.014                |
| speed = 0        | -0.49    | 0.11    | -4.34    | 2.00E-05 | -0.71                |
| age (yr)         | 0.010    | 0.019   | 0.53     | 0.59313  | -0.027               |
| age² (yr²)       | 0.0039   | 0.0011  | 3.67     | 0.00029  | 0.0018               |
| vol (ft³)        | -0.025   | 0.0037  | -6.77    | 6.80E-11 | -0.032               |
| vol² (ft⁶)       | 0.000074 | 0.000013| 5.69     | 3.00E-08 | 0.000048             |
| Manuf: Japan     | -0.34    | 0.070   | -4.86    | 1.90E-06 | -0.48                |
| Manuf: Germany   | -0.88    | 0.12    | -7.26    | 3.30E-12 | -1.11                |

Figure 2.8: Predicted ln(AER) versus measured ln(AER) under recirculation ventilation mode
Table 2.4: AER under OA Model Coefficients, Confidence Intervals, and p-Values

| GEE                  | Estimate | Std.err | Wald  | Pr(>|W|) | Confidence          |
|----------------------|----------|---------|-------|----------|---------------------|
| Intercept            | 4.2      | 0.24    | 295   | < 2e-16  | 3.7                 |
|                      |          |         |       |          | 0.7                 |
| fan strength         | 1.88     | 0.14    | 170   | < 2e-16  | 1.6                 |
|                      |          |         |       |          | 0.43                |
| fan strength²        | -0.92    | 0.11    | 70    | < 2e-16  | -1.1                |
| speed = 0            | -0.32    | 0.09    | 11    | 0.0007   | -0.50               |
| speed > 0 (miles h⁻¹)| 0.0048   | 0.0013  | 14    | 0.0002   | 0.0023              |
|                      |          |         |       |          | 0.0038              |
| vol (ft³)            | -0.0073  | 0.0019  | 15    | 0.0001   | -0.011              |
|                      |          |         |       |          | 0.0056              |
| fan strength at speed| 0.40     | 0.26    | 2.5   | 0.12     | -0.10               |
|                      |          |         |       |          | 0.76                |
| fan strength² at speed| 0.13  | 0.20    | 0.45  | 0.50     | -0.26               |
|                      |          |         |       |          | 0.59                |

MLR

| Intercept            | 4.2      | 0.14    | 29    | < 2e-16  | 3.9                 |
|                      |          |         |       |          | 4.5                 |
| fan strength         | 2.3      | 0.40    | 5.7   | 0        | 1.5                 |
|                      |          |         |       |          | 3.1                 |
| fan strength²        | -1.3     | 0.36    | -3.7  | 0.00040  | -2.1                |
| speed = 0            | -0.34    | 0.15    | -2.3  | 0.023    | -0.63               |
| speed > 0 (miles h⁻¹)| 0.0043   | 0.0014  | 3.0   | 0.0028   | 0.0015              |
| vol (ft³)            | -0.0074  | 0.00080 | -8.8  | 0        | -0.0090             |
|                      |          |         |       |          | -0.0057             |

Figure 2.9: Predicted ln(AER) versus measured ln(AER) under outside air intake ventilation mode
For example, an 85 ft\(^3\) vehicle is expected to have an AER, which is 2.2 times that in a 120 ft\(^3\) vehicle (or 1.6 times higher if AER units are ft\(^3\) h\(^{-1}\)). To provide a typical AER value under RC conditions for reference, a seven-year-old vehicle (50\(^{th}\) age percentile, U.S. manufactured, and 110 ft\(^3\), the average U.S. fleet cabin volume) would have an AER of 3.7 h\(^{-1}\) at 35 miles h\(^{-1}\) and 6.7 h\(^{-1}\) at 65 miles h\(^{-1}\).

Under OA conditions, fan strength explained the most variability in lnAER, followed by speed. For example, increasing a four-setting fan from low (0.25) to medium (0.5) to highest (1.0) increased the AER by a factor of 1.3 and 1.7, respectively. In comparison, increasing the driving speed, the second most significant variable, from arterial to freeway speeds only increased the AER by 1.2. Vehicle cabin volume was also found to be significant, with higher volume vehicles having lower predicted AER (h\(^{-1}\)). An 85 ft\(^3\) sub-compact vehicle had 1.3 times higher AER than a 120 ft\(^3\) large sedan. Fan strength and zero speed interaction terms, while not significant individually, were significant as a pair, so included in the OA model. Figure 2.9 (b) shows the model predictions plotted against the two most significant determinants of AER under OA conditions; ventilation fan strength and vehicle speed, for a sub-compact (85 ft\(^3\)) and large sedan vehicle (120 ft\(^3\)), thus capturing the full range of AERs that can be expected under OA condition. For the previously mentioned reference vehicle travelling at 35 and 65 miles h\(^{-1}\), AER would be 72 h\(^{-1}\) and 83 h\(^{-1}\), respectively, at the middle fan setting, roughly an order of magnitude higher than under RC conditions.
2.4 CONCLUSIONS

The predictive model explained greater than 70% of the variation in the observed AERs under both ventilation conditions, from <2 h$^{-1}$ to up to 150 h$^{-1}$, but only requires variables that are easily obtainable through questionnaire or survey. AER was found to be a predictable function of vehicle age (or mileage), speed, and to a lesser extent, cabin volume and manufacturer. While fan speed setting was relatively unimportant under recirculation setting it was the strongest determinant of AER under outside air intake conditions. Age, manufacturer and volume can be obtained via collecting just a single survey data like the vehicle identification number. Average speed can also be estimated by survey for typical commutes to work, and from city averages for other trips based on home location. Ventilation setting preference would be a necessary survey component as driving with ventilation set to outside air or with windows open generally produces AERs that are an order of magnitude higher, with correspondingly low particle loss rates.
CHAPTER 3: VEHICLE AND DRIVING CHARACTERISTICS THAT INFLUENCE PARTICLE NUMBER CONCENTRATIONS IN VEHICLES

3.1 INTRODUCTION

The proximity of vehicles to relatively undiluted emissions from other vehicles on freeways and busy roadways leads to significantly elevated pollutant concentrations inside vehicle cabins compared to other indoor environments. Consequently, a disproportionate share of total personal exposure can occur while driving, especially for pollutants emitted mostly by vehicles, like ultrafine particles (particles smaller than 100 nm, UFP). Fruin et al. (2008) calculated that 33-45% of UFP exposure occurs while driving in Los Angeles, taking other micro-environmental concentrations and time-activity patterns into account, but ignoring in-vehicle particle losses. In suburban locations of less traffic, Wallace and Ott (2011) estimated a 17% contribution of in-vehicle microenvironment to total UFP exposure.

Despite its importance as a route of exposure, the contribution of the in-transit vehicular microenvironment remains largely uncharacterized, in part due to the difficulty of characterizing the large differences in air exchange rate (AER), which drives particle influx rates and varies not only from vehicle to vehicle but also across different driving conditions. As described in the previous chapter, to better characterize AERs, we (Fruin et al., 2011) tested 59 vehicles and reported that AER under recirculation ventilation conditions can be reliably predicted based on vehicle’s age, mileage, driving
speed and manufacturer ($r^2 = 0.7$). Under outside air intake conditions, for the eight vehicles tested, we found AERs to be an order of magnitude higher than at recirculation settings. We also found that at the outside air ventilation setting, AER was driven by fan speed and not vehicle speed. In a similar but smaller study, Knibbs et al. (2009) reported AER values at higher fan settings during outside air conditions to be 73% higher than at lowest fan setting for six vehicles.

Particle influx and removal rates result from a complicated interaction among multiple factors, including AER, physical characteristics of the vehicle; particle size and in-cabin filter efficiency. Any accurate determination of the relative influence of each factor requires experiments where factors are systemically varied to observe effects under real driving (aerodynamic) conditions; else AERs are not realistic. Several recent studies (Gong et al., 2009; Xu and Zhu, 2009; Xu et al., 2011) addressing in-vehicle particle losses have not used realistic driving, in favor of artificial air movement, or have relied on measurements that are difficult to make outside a laboratory, such as filter efficiency tests. Furthermore, these studies have suffered from small sample sizes, ranging from only one to three vehicles (Gong et al., 2009; Xu and Zhu, 2009).

Of the few studies that have used real driving conditions, Pui et al. (2008) and Qi et al. (2008) demonstrated a dramatic particle number concentration reduction in-cabin during recirculation ventilation settings in two vehicles, although AERs were not reported. Zhu et al. (2007) observed particle losses of about 85% at recirculation
settings in three vehicles but AER was not measured and variable speeds during the tests would have resulted in variable AERs. Zhu et al. (2007) was also the only study identified that made size-resolved particle concentration measurements but only up to 217 nm. Furthermore, they reported the fractional losses to be primarily dependent on particle size and vehicle characteristics.

The most useful study from an exposure assessment perspective has been by Knibbs et al. (2010) who measured the inside-to-outside UFP concentration ratios in five vehicles and reported a high correlation between these ratios and AER \( (r^2 = 0.81) \). They reported an average particle reduction of 0.69 during recirculation settings at low fan setting and 0.08 at outside air intake, but did not associate the losses with particle size or influence of specific removal mechanisms like cabin filtration.

The goal of this study was to quantify the effects on particle reduction rates due to changes in 1) ventilation setting; 2) measured AER; 3) fan setting; 4) filter condition; 5) driving speed; and 6) easily-obtainable vehicle characteristics such as age and mileage (which affect AER); and to determine the relative importance of each of these variables. It is the first study to combine measurements of AER and particle losses as a function of particle size.
3.2 METHODS

3.2.1 VEHICLE SELECTION AND CONDITIONS TESTED

Six vehicles were selected such that AERs at recirculation ventilation settings spanned the inter-quartile range of AERs (4.5 - 13 h\(^{-1}\), median 7.4 h\(^{-1}\)) measured in Fruin et al. (2011), Hudda et al. (2012) and (described in Chapter 2). Two vehicles (a 1999 Ford Contour and 2001 Ford Escort) were more than 10 years old when tested, while four newer vehicles (a 2010 Toyota Prius, 2010 Scion Xb, 2009 Toyota Matrix and 2009 Honda Civic) were 3-14 months old. All six vehicles were evaluated at both recirculation (RC) and outside air intake (OA) ventilation conditions. At each ventilation condition, experiments were conducted in both stationary and mobile mode at both medium and high fan speed settings.

At RC setting, a total of 42 conditions were evaluated. Experiments measuring particle losses and AER were conducted on six vehicles at seven AERs each (resulting from a combination of ventilation fan setting and driving speed). At OA settings, a total of 34 combinations of vehicle/ speed/ fan strength were evaluated. However, because it has been previously demonstrated that fan setting and not vehicle speed determines AER at OA ventilation settings (Fruin et al., 2011), simultaneous particle loss and AER measurements were conducted for a subset of conditions (10 out of 34), i.e., in stationary vehicles at various fan settings. Particle loss measurements in moving vehicles were assumed to have the similar AER for a given fan speed. Air conditioning was kept on during all experiments, except for those at which the fan was off (6 out of
total 76 conditions evaluated). Relevant vehicle characteristics are summarized in the following Table 3.1.

Table 3.1: List of vehicles tested.

<table>
<thead>
<tr>
<th>Vehicle Model</th>
<th>Mileage</th>
<th>In-Cabin Filter</th>
<th>Vehicle Age (Years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ford Contour</td>
<td>1999</td>
<td>115990</td>
<td>Yes</td>
</tr>
<tr>
<td>Ford Escort</td>
<td>2001</td>
<td>127280</td>
<td>Yes</td>
</tr>
<tr>
<td>Toyota Prius</td>
<td>2010</td>
<td>3210</td>
<td>Yes</td>
</tr>
<tr>
<td>Toyota Matrix</td>
<td>2009</td>
<td>26125</td>
<td>Yes</td>
</tr>
<tr>
<td>Toyota Scion Xb</td>
<td>2010</td>
<td>24068</td>
<td>Yes</td>
</tr>
<tr>
<td>Honda Civic</td>
<td>2009</td>
<td>22000</td>
<td>Yes</td>
</tr>
</tbody>
</table>

3.2.2 *PARTICLE CONCENTRATION MEASUREMENTS*

Particle number concentration measurements were made using a condensation particle counter (CPC, TSI Inc. Model 3007, size range 10 nm to 1000 nm) and number concentration measurements by size using a Scanning Mobility Particle Sizer (SMPS, TSI Inc. Differential Mobility Analyzer Model 3080 and Model 3022a CPC). The measured mobility diameter \(D_p\) range was 14-750 nm. However, data are presented only for the 14-400 nm range because above 400 nm, relatively few particles were counted and the resulting concentrations had large uncertainties. Furthermore, since roadway particles between 14-25 nm are volatile, concentrations in this size range are exceptionally variable in on-road environments. Reported results for this size range should be interpreted with caution.
Experiments were conducted at 0, 20 and 35 miles h\(^{-1}\), with speed recorded each second by a Garmin GPSMAP unit 76CSC. Experiments at 20 and 35 miles h\(^{-1}\) were conducted while driving at constant speed around the Rose Bowl Stadium in Pasadena CA, a three mile long loop with little vehicular traffic. Freeway speeds were not evaluated in this study due to the rapidly changing traffic and particle number and size distributions typically present on Los Angeles freeways.

All in-vehicle measurements were made with windows fully closed. Outside vehicle concentrations were assumed to be equal to roadway concentrations, and were measured for 10-20 minutes before and after the in-vehicle measurement period, with windows fully open to allow the outside air to pass freely through the vehicle. Earlier in the study, it was verified that open window conditions allow accurate measurement of roadway particle size distributions and number concentrations by comparing simultaneous measurements with two CPCs, one measuring concentrations with a 1 m long inlet sampling air right outside the vehicle, and the other CPC, sampling in the middle of the backseat of the vehicle. Ambient particle number concentrations and size distributions were also measured using a stationary monitor at a position central to the run loop. All ambient concentrations were stable to within 10%, before, during, and after a run.

Inside-to-outside particle concentration ratio (I/O) was calculated for all measurements, to provide a measure that reflects particle removal indoors. I/O ratio was calculated
after concentrations were stable in the vehicle over 15 minutes or more of sampling. I/O ratios reported for specific size ranges are equivalent to the average of mobility-diameter-specific I/O ratios within the range.

3.2.3 Air Exchange Rate Measurements

During both mobile and stationary conditions, air exchange occurs between the vehicle cabin and the outside environment through leaks in the body of the vehicle (door seals, window cracks, etc.) and through the ventilation system, when it is set to draw outside air into the cabin. This air exchange continually replenishes the vehicle cabin with pollutants/particles from the outside environment, hence is an essential measurement in any study of in-vehicle exposure.

AERs were determined at RC conditions using CO₂ as a tracer gas and two occupants as a stable source of CO₂ generation (described in detail in Section 2.2.4, Chapter 2). Build-up rates of CO₂ concentration were used to determine the CO₂ rate of emission, and equilibrium CO₂ concentrations at fixed speeds then allowed calculating the AER at that speed. Under OA conditions, AERs are much higher, requiring higher starting CO₂ concentrations that cannot be readily reached by occupants. As it was observed that speed plays an insignificant role in affecting AER compared to fan setting at OA conditions, only stationary tests were necessary to characterize AER, and CO₂ from a pressurized cylinder was used to produce the necessary high starting concentrations.
(described in detail in Section 2.2.5, Chapter 2). Table 3.2 and 3.3 list the AERs for the vehicles at various conditions tested.

Table 3.2: AER rates (h\(^{-1}\)) at recirculation setting for the vehicles tested.

<table>
<thead>
<tr>
<th>Speed (miles h(^{-1}))</th>
<th>Fan</th>
<th>Ford Contour</th>
<th>Ford Escort</th>
<th>Toyota Prius</th>
<th>Toyota Matrix</th>
<th>Honda Civic</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>No</td>
<td>2.1</td>
<td>2.20</td>
<td>0.53</td>
<td>0.27</td>
<td>1.70</td>
</tr>
<tr>
<td>0</td>
<td>Medium</td>
<td>3.1</td>
<td>3.50</td>
<td>0.83</td>
<td>0.53</td>
<td>2.40</td>
</tr>
<tr>
<td>0</td>
<td>Full</td>
<td>6.2</td>
<td>5.40</td>
<td>1.50</td>
<td>1.40</td>
<td>3.20</td>
</tr>
<tr>
<td>20</td>
<td>Medium</td>
<td>11.2</td>
<td>8.10</td>
<td>3.00</td>
<td>3.50</td>
<td>4.10</td>
</tr>
<tr>
<td>20</td>
<td>Full</td>
<td>14.1</td>
<td>9.50</td>
<td>3.70</td>
<td>4.80</td>
<td>4.70</td>
</tr>
<tr>
<td>35</td>
<td>Medium</td>
<td>16.0</td>
<td>11.5</td>
<td>3.70</td>
<td>4.50</td>
<td>5.10</td>
</tr>
<tr>
<td>35</td>
<td>Full</td>
<td>19.0</td>
<td>13.5</td>
<td>4.30</td>
<td>5.70</td>
<td>6.30</td>
</tr>
</tbody>
</table>

Table 3.3: AER rates (h\(^{-1}\)) at outside air intake setting for the vehicles tested. \(^2\)

<table>
<thead>
<tr>
<th></th>
<th>Ford Contour</th>
<th>Ford Escort</th>
<th>Toyota Scion xB</th>
<th>Toyota Prius</th>
<th>Toyota Matrix</th>
<th>Honda Civic</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Fan</strong></td>
<td><strong>AER</strong></td>
<td><strong>AER</strong></td>
<td><strong>AER</strong></td>
<td><strong>AER</strong></td>
<td><strong>AER</strong></td>
<td><strong>AER</strong></td>
</tr>
<tr>
<td>1/4</td>
<td>36.0</td>
<td>1/3</td>
<td>51.0</td>
<td>1/4</td>
<td>20.0</td>
<td></td>
</tr>
<tr>
<td>2/4*</td>
<td>65.0</td>
<td>2/3*</td>
<td>64.0</td>
<td>2/4*</td>
<td>35.0</td>
<td></td>
</tr>
<tr>
<td>3/4</td>
<td>94.0</td>
<td>3/3(^$)</td>
<td>83.0</td>
<td>3/4</td>
<td>50.0</td>
<td></td>
</tr>
<tr>
<td>4/4(^$)</td>
<td>117.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Honda Civic</strong></td>
<td><strong>AER</strong></td>
<td><strong>AER</strong></td>
<td><strong>AER</strong></td>
<td><strong>AER</strong></td>
<td><strong>AER</strong></td>
<td><strong>AER</strong></td>
</tr>
<tr>
<td>2/12</td>
<td>57.0</td>
<td>1/7</td>
<td>23.0</td>
<td>1/4</td>
<td>20.0</td>
<td></td>
</tr>
<tr>
<td>4/12</td>
<td>72.0</td>
<td>2/7</td>
<td>36.0</td>
<td>2/4*</td>
<td>35.0</td>
<td></td>
</tr>
<tr>
<td>6/12*</td>
<td>93.0</td>
<td>3/7</td>
<td>48.0</td>
<td>3/4</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>8/12</td>
<td>112.0</td>
<td>4/7*</td>
<td>59.0</td>
<td>4/4(^$)</td>
<td>71.0</td>
<td></td>
</tr>
<tr>
<td>10/12</td>
<td>125.0</td>
<td>5/7</td>
<td>69.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12/12(^$)</td>
<td>141.0</td>
<td>6/7</td>
<td>84.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>7/7(^$)</td>
<td></td>
<td></td>
<td>97.0</td>
<td></td>
</tr>
</tbody>
</table>

\(^2\) *Medium fan setting, \(^\$\) *Full fan setting
3.3 RESULTS AND DISCUSSION

3.3.1 EFFECT OF AIR EXCHANGE RATE ON I/O RATIOS

Figure 3.1: I/O ratio dependence on AER for 25-400 nm particles under re-circulation (RC) and outside air (OA) ventilation setting.

At RC ventilation conditions, the 42 AERs across the six vehicles tested varied from less than 1 to 19 h⁻¹. At OA setting, measured AERs varied from 20 to 145 h⁻¹. Any significant increase in AER resulted in an increase in particle influx rate and increased I/O ratios. Figure 3.1 presents I/O ratio results under both ventilation conditions, and illustrates the strong dependence of I/O ratio on AER, which is exhibited most distinctively in the difference between the I/O ratios at RC and OA ventilation settings. The decrease in I/O ratio with increasing AER was less dramatic at OA conditions compared to RC conditions.
Across the size range (25-400 nm) an increase in AER elevated I/O ratio, but the effect was strongest for particles above 200 nm.

The only other study to report both I/O ratio and AER, Knibbs et al. (2010), reported a high correlation between I/O ratio for UFP and AERs ($r^2 = 0.81$). In the present study, at RC the $r^2$ between I/O ratio (using a CPC 3007, the same instrument used by Knibbs et al. (2010)) and AER was 0.80, indicating that AER is the most significant determinant of I/O ratio at RC ventilation conditions. The average I/O ratio under RC conditions was 0.17 ± 0.13. In contrast, at OA conditions, the I/O ratios averaged 0.67 ± 0.10. The average $r^2$ between I/O ratios and AER under OA conditions was 0.75 ($r^2$ values for all but one vehicle were 0.9 or above). On average for the six vehicles tested, a switch in ventilation condition from RC to OA increased I/O ratio by nearly a factor of four.

3.3.2 Effect of Vehicle Speed and Age on AER and I/O Ratios

Under RC conditions, an increase in speed increased AER and I/O ratios. On average, a 10 miles h$^{-1}$ increase in speed resulted in a 1.65 h$^{-1}$ increase in AER and a 0.035 increase in I/O ratio. Speed affected AER more for older vehicles (+2.4 h$^{-1}$/10 miles h$^{-1}$) compared to the newer vehicles (+1.2 h$^{-1}$/10 miles h$^{-1}$), similar to the results reported by Knibbs et al. (2009). As a result, I/O ratio, which depends strongly on AER, increased with speed at twice the rate for older vehicles (-0.05 /10 miles h$^{-1}$, Pearson $r^2 = 0.78$) than for newer vehicles (-0.025 /10 miles h$^{-1}$, Pearson $r^2 = 0.20$).
Despite these differences by vehicle age, an overall strong correlation was observed between AER and speed as well as I/O ratios and speed, as well as AER. In Fruin et al. (2011) an $r^2$ equivalent of 0.92 was calculated between AER and speed for a much larger fleet of vehicles and for speeds up to 70 miles h$^{-1}$ using a Generalized Estimating Equations (GEE) (Liang et al., 1986). (GEE techniques account for correlated measurements within a vehicle; for example, a tight vehicle with lower AER will consistently have lower I/O ratios across each speed compared to a leakier vehicle with higher AER). Among the six vehicle data, using GEE technique, 82% of the variation in AER could be accounted for by speed ($p$ value $= 2.5 \times 10^{-9}$). Furthermore, nearly all variation ($r^2 = 0.98$, $p$-value $= 6.9 \times 10^{-7}$) in I/O ratios at RC setting was explained by changes in AER. Given the consistent and strong correlations between AER and speed (Knibbs et al., 2009, Fruin et al, 2011; this study) and between I/O ratios and AER at RC setting (Knibbs et al., 2009, this study), these results can be expected to extrapolate well to the higher AERs of vehicles travelling at higher freeway speeds (65–70 miles h$^{-1}$) from the speeds tested in this study, i.e., 35 miles h$^{-1}$ and less.

In contrast, under OA conditions, no definitive association between speed and I/O ratios could be discerned for the six vehicles tested. Since AER at OA is driven by mechanical ventilation rather than speed-driven pressure differences outside the vehicle shell, this lack of association is not surprising. Knibbs et al. (2009) have previously shown that at OA conditions, for the newer four of total six vehicles tested, the association between AER and speed was weak (linear regression on all four vehicles: $AER = 0.15 \times \text{Speed}$
[miles h\(^{-1}\)] + 51, \(r^2 = 0.06\). For the oldest two (10 and 19 year old) of the total six vehicles tested, the relationship was stronger (AER = 0.59 * Speed [miles h\(^{-1}\)] + 52, \(r^2 = 0.52\)). This suggests that for older vehicles, the AER at OA may increase at speeds higher than those tested in this study, which should elevate I/O ratios. However, for the two oldest vehicles in this study (10 years of age), any increase in AER with speeds between 1 to 35 miles h\(^{-1}\) did not cause a noticeable change in I/O ratios.

Most real-world driving involves constantly changing speeds due to traffic conditions and widely varying roadway particle number concentrations. To illustrate how well the steady speed/AER and relatively stable roadway concentration condition results apply to such changing speed and concentration conditions, measurements were made during two 15-minute duration trips on a freeway (I-110) and an arterial road (Figueroa Street, downtown Los Angeles) in a Toyota Matrix 2009. No attempt was made to maintain steady speeds during these runs, and average speed was 55 miles h\(^{-1}\) and 27 miles h\(^{-1}\) on the freeway and arterial road, respectively. The ventilation setting was set to OA, leading to moderately high AER (35 h\(^{-1}\)) that would allow rapid influx of roadway concentration into the vehicle cabin and reflect the unsteady roadway environment, as shown in Figure 3.2. It should be noted that under low AER conditions, in-cabin concentrations are fairly steady due to limited influx of particles. The average I/O ratio calculated from average in-cabin and roadway particle number concentrations agreed almost perfectly with the I/O ratios predicted from the I/O ratio versus speed regression equation based on measurements at steady speeds.
3.3.3 **Effect of Particle Size on I/O Ratios**

Figure 3.2: Agreement between measured I/O during variable speed driving and regression-predicted I/O.

Figure 3.3: Size range specific I/O ratios at three speeds and two ventilation conditions tested. The dashed lines join values from the same vehicle.
Other than AER, particle size itself can be expected to play a role in attenuation, since particle infiltration, surface deposition and filtration efficiency are functions of particle size (Liu and Nazaroff, 2003; Nazaroff 2004; Hinds 1999). I/O ratios for the five size ranges plotted in Figure 3.3 show the lowest attenuation in-cabin (or highest I/O ratios) for particles in the size range 200-400 nm. Similar values for the least attenuated particle size have been reported in other indoor environments (Nazaroff 2004). The lowest I/O ratios were consistently associated with the UFP size range. This is consistent with the high losses expected for smaller particles due to higher diffusion rates (Hinds 1999).

For a given ventilation and speed combination, particle size-specific I/O ratios were found to be similar across size for newer vehicles and only moderately different across size for older vehicles. At RC, the I/O ratios for 100-200 and 200-400 nm were respectively 0.04 and 0.07 higher than the average I/O ratio (0.16 ± 0.09) for ultrafine range. At OA, the differences in size specific I/O ratios were more accentuated than at RC. The I/O ratios for both 100-200 and 200-400 nm were 0.06 higher than the average I/O ratio (0.75 ± 0.12) for ultrafine range. The difference in I/O ratios across size was much less than the difference in I/O ratios across ventilation conditions, evident in Figure 3.3.
### 3.3.4 Effect of Ventilation Fan Setting on I/O Ratios

![Graph showing comparison of I/O ratios at different speeds and fan settings.]

**Figure 3.4**: Comparison of I/O ratios at different speeds and fan settings.

Figure 3.4 shows that increasing the ventilation fan setting from medium to full elevates the I/O ratios at both RC and OA, but this reduction was far more pronounced at OA than at RC. Under RC conditions, the attenuation reduction inside the cabin (thus the elevated I/O ratios) is somewhat counter-intuitive, since increasing fan setting might be thought to increase particle removal via greater rates of airflow through the in-cabin filter, when present. Also, the deposition of particles on cabin surfaces increases with increase in in-cabin air velocity (Gong et al., 2009). However, fan setting has been previously demonstrated to increase AER for older vehicles (Fruin et al., 2011; Knibbs et al., 2009), likely due to leaks into the ventilation system, and filtration efficiency.
decreases at greater air flow velocities associated with higher fan settings as well (Qi et al., 2008). Apparently, these mostly offset the particle reductions expected due to greater volumes of air being filtered at high fan settings under RC conditions. However, higher fan settings resulted in increased losses for particles smaller than 50 nm, perhaps due to the increased turbulence in the ventilation system at higher fan settings.

At OA, an increase in fan speed settings strongly increased AER. The I/O ratios at full fan setting under OA conditions were as much as double that at medium fan setting and AERs at full fan setting were about 65% higher than at medium fan. Thus, ventilation fan setting is a key predictor of I/O ratios under OA ventilation conditions.

3.3.5 Effect of Cabin Air Filter and Loading on I/O Ratios

In order to determine the effect of cabin filters on particle removal and resultant I/O ratios, measurements were conducted under several filter conditions: no filter, used (loaded), and new, at both OA and RC setting in three stationary vehicles. The Ford Contour’s in-use filter had been operational for 36 months at the time of testing and was heavily loaded. The Honda Civic’s filter had been in operation for ~14 months at the time of testing and was moderately loaded, and the Prius’s filter had been in use for ~3 months, and was lightly loaded. All new filters were standard replacement cabin air filters bought from an auto parts store (all either brand STP or Purolator).

The presence or absence of the filter, or its loading, was observed to have only a small effect on I/O ratios. Used, loaded filters provided only moderately lower or comparable
I/O ratios to a new filter. The results in Figure 3.5 for a Toyota Prius are typical and show that overall particle loss is not significantly affected by the presence of a new filter or even a loaded filter at OA settings. Without any filter, I/O ratio was only moderately higher (maximum difference observed was 0.1).

![Figure 3.5: I/O ratios by filter condition or absence under OA conditions in a 2010 Toyota Prius.](image)

Furthermore, the effect of several different filter loadings as characterized by in-vehicle pressure drop for the same vehicle was also investigated under OA ventilation conditions. Increased loading and resulting reduced pore size and flow rates, were found to decrease I/O ratios for UFP, but no significant changes were observed for particles exceeding 100 nm in size. Four loaded filters and a brand new filter were placed into Honda Civic 2010 model and tested at OA setting. Experiments were
conducted in a stationary vehicle. Highest pressure drop (plotted as a subset in Figure 3.6) was observed for the heaviest loaded filter L4, while values for moderately loaded filters L1, L2 and L3 were comparable (Figure 3.6). Under OA setting, the I/O ratios decreased with filter loading for UFP particles (>100 nm) from 0.36 (± 0.05) for new filter to 0.36 (± 0.04), 0.42 (± 0.05), 0.49 (± 0.05) and 0.54 (± 0.05) for L1, L2, L3 and L4, respectively. A large increase in filter loading resulted in significantly better filtration efficiency for nano-particles (<50 nm), as seen in the abrupt change of I/O values for L3 and L4 in Figure 3.6.

This implies that the particle removal due to filtration is a small fraction of the total particle attenuation, and that most of the attenuation is probably due to turbulent surface deposition in the ventilation system or vehicle surface itself. Pui et al. (2008) reported 19% particle loss in a Toyota Camry in the absence of filter and suggested intrinsic losses in the ventilation system as an attenuation mechanism. In our tests 23, 40 and 40% particle losses were observed for the Honda Civic, Toyota Prius and Ford Contour with no filter, respectively. I/O ratios based on total particle number concentration are reported in Table 3.4 for three vehicles by filter presence and condition. Furthermore under RC settings, the I/O ratios in the absence of a filter were on average only 5% higher than with a filter in place. Although the presence and condition of the filter were insignificant to I/O ratio, it somewhat affected the time that the system requires to achieve the maximum attainable attenuation and stable I/O
ratios. Similar increases in the time required to reach maximum attenuation have been reported by Pui et al. (2008).

Figure 3.6: Effect of filter loading on particle removal for filters tested in Honda Civic vehicle under OA conditions.

Figure 3.7: Effect of presence of filter in RC ventilation mode. \( C_0 \) is the concentration at the beginning of experiment, i.e., at time \( t = 0 \) and is equal to the ambient concentration.
### Table 3.4: I/O ratios for three filter scenarios.

<table>
<thead>
<tr>
<th></th>
<th>Recirculation Setting</th>
<th>Outside Air Setting</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>No Filter</td>
<td>New Filter</td>
</tr>
<tr>
<td><strong>Honda Civic</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.08 ± 0.02</td>
<td>0.04 ± 0.01</td>
</tr>
<tr>
<td><strong>Ford Contour</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.11 ± 0.02</td>
<td>0.07 ± 0.01</td>
</tr>
<tr>
<td><strong>Toyota Prius-1</strong></td>
<td>0.05 ±0.01</td>
<td>0.03 ± 0.01</td>
</tr>
<tr>
<td><strong>Toyota Prius-2</strong></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Measurements made in two Prius 2010 (vehicle 1 had ~3,200 miles and vehicle 2 had ~11,000 miles) to check for the effects of vehicle make.

Some recent studies (Xu and Zhu, 2009; Xu et al., 2011) attribute all particle losses in the ventilation system to filtration and incorporate them into models by using filter efficiency as the removal mechanism. If the presence of cabin filters actually plays a minor role in the attenuation of particles inside the vehicles, as observed in our measurements, a much larger component of attenuation occurs due to losses onto cabin and ventilation system surfaces. These different loss mechanisms should therefore be differentiated in models since they likely differ under different ventilation and driving conditions. Furthermore, any quantitative modeling should account for the intrusion flow of outside air into the ventilation system at RC. Knibbs et al. (2009; 2010) and Fruin et al. (2011) report that at RC, increases in fan setting increase the AER. They also reported that this increase seems to increase with age of the vehicle and could vary considerably by vehicle. Less than 15% increase on average was reported by Fruin et al.
(2011) when fan setting was changed from medium to full, but this became as large as 40% for the older vehicles.

3.4 CONCLUSIONS

Predicting particle exposure inside vehicles requires determining ventilation setting first and foremost (i.e., OA or RC), due to its large impact on AER. Under OA conditions, fan setting is the most dominant variable, and I/O ratio was approximately 0.6 and fairly independent of speed. Under RC conditions, I/O ratio has a large range and varies from 0.5 to zero, depending on AER, which can be predicted by speed and vehicle age and mileage (Fruin et al. 2011). Under open window conditions, I/O ratios approach one, i.e., in-cabin concentrations frequently equal roadway concentration.

Difficult to obtain information, such as state of in-cabin filter loading, does not appear to be a crucial factor in assessing I/O ratios and the resulting in-vehicle particle exposures. It also does not appear that changes in on-road size distribution have a large impact on I/O ratios. Figure 3.8 exhibits the I/O ratio differences for four widely different hypothetical size distributions having number concentration mode less than 25 nm (fresh vehicle exhaust plume), 25-50 nm (on-road diluted plumes), 50-100 nm (aged vehicle emissions) and 100-200 nm (aged aerosol observed as urban background). The largest difference in I/O ratio occurs between aged and fresh aerosol, changing the overall I/O ratio by no more than 0.1 at OA and less than 0.05 at RC. Considering that on-road particle size number concentrations are dominated by ultrafine particles
(Morawska et al., 1998), I/O ratio measurements based only on total number concentration (e.g., a total particle count from CPC) would be expected to (and were observed to) produce I/O ratio measurements nearly identical to an average I/O ratio resulting from a number-weighted average I/O ratio across multiple size ranges. Therefore, all of the variables needed to estimate I/O ratio within 10% or less can be obtained through questionnaires given to vehicle owners.

Figure 3.8: Impact of change in particle size distribution on number concentration weighed I/O ratios.
Figure 3.9: Progression of particle loss within vehicles at recirculation and outside air intake condition. The subscripts in the legend indicate the experiment time during which the scan was made. The measured AER at 35 miles h\(^{-1}\), medium fan, recirculation mode was 7.3 h\(^{-1}\) and at 0 miles h\(^{-1}\), medium fan outside air intake mode was 93 h\(^{-1}\).

Lastly, for the case of short duration trips, the equilibration conditions reported in this study for I/O ratio may not be reached. The particle attenuation reported here was attained typically within 5 to 10 minutes at OA conditions and 15 to 20 minutes at RC conditions, as shown in Figure 3.9. The benefit derived from higher reduction in particle exposure with the use of RC setting may be reduced if short trips are taken repeatedly. The dynamic nature of particle attenuation should be considered in assessing short trip exposures.
CHAPTER 4: PREDICTIVE MODEL FOR ULTRAFINE PARTICLE NUMBER CONCENTRATION IN-CABIN-TO-ROADWAY RATIOS BASED ON A LARGE, REPRESENTATIVE SAMPLE

4.1 INTRODUCTION

Exposure to traffic related pollutants has been associated with detrimental health outcomes like asthma, exacerbation of adverse respiratory (Brauer et al., 2002; Gauderman et al., 2005; McConnell et al., 2006; Gan et al., 2011) and cardiovascular outcomes (Delfino et al., 2005), coronary artery atherosclerosis (Araujo et al., 2008; Künzli et al. 2010), and an increase in mortality (Hoek et al., 2002; Stölzel et al., 2007; Wichmann et al., 2000). The particular components of traffic emissions responsible for causing adverse health effects are not known (Sioutas et al., 2005; Delfino et al., 2005), but ultrafine particles (UFP), defined as particles having aerodynamic diameter less than 100 nm, are of particular interest due to their large cumulative surface area, ability to trans-locate through the epithelium, as well as their elevated proportion of organic material and metals that results in high oxidative potential (Li et al., 2009; Delfino et al., 2005; Morgan et al., 2011).

Numerous studies (e.g., Leung and Harrison 1999; Westerdahl et al., 2005) have shown that UFP concentrations on or in the vicinity of roadways are frequently almost one order of magnitude higher than ambient levels. This has important implications for exposure assessment. For example, less than 10% of daily time spent in vehicular transit
microenvironments (Klepeis et al., 2001) has been estimated to contribute 35-50% of total UFP exposure by Fruin et al. (2008) for Los Angeles residents under open window conditions, and 17% by Wallace and Ott (2011) for more suburban locations. However, large variations in exposure inside vehicles are expected to occur not only due to differences in roadway environments, but also because inside-to-outside (I/O) ratios (i.e., in-vehicle to roadway concentration ratios) vary vehicle to vehicle from nearly zero to one (Knibbs et al., 2010; Hudda et al., 2011).

Some studies that have measured UFP I/O ratios or particle number concentrations inside vehicles include Pui et al. (2008) and Qi et al. (2008), who investigated in-cabin air filter efficiency, and found large UFP reductions inside two new vehicles under recirculating conditions, although air exchange rate (AER) or speed was not reported. Zhu et al. (2007) also reported large reductions in in-vehicle UFP concentrations in three vehicles. Other recent studies (Gong et al., 2009; Xu et al., 2009) that have produced mechanistic models of particle losses inside vehicles relied on parameters like particle penetration through cracks and surface deposition rates, which while useful, are not obtainable outside of a laboratory setting.

Two recent studies have simultaneously measured both I/O and AER, and shown that I/O ratio is strongly dependent on air exchange rate (AER), which is defined as the number of times per hour vehicle cabin air is replaced by roadway or
outside air. Knibbs et al. (2010) reported an $r^2$ of 0.81 (Pearson correlation coefficient) between AER and I/O ratios and Hudda et al. (2011) reported an $r^2$ of 0.75 or 0.80, depending on ventilation choice. Both of these studies performed measurements under real driving conditions (multiple speed and ventilation conditions) and found that ventilation preference (windows open, outside air intake or in-cabin air recirculation) and ventilation fan setting strongly influence AER and the resulting I/O ratio. Prior to Knibbs et al. (2010), no in-vehicle UFP I/O ratio results had been reported in a form that can be generalized.

As it is impractical to measure either the I/O ratio (or AER) for large numbers of subjects’ vehicles as required in an epidemiological study addressing drive-time exposure, predictive models are needed for estimating I/O ratios. If these models could be based on information that can be collected via questionnaire, for example, they would be useful tools for more accurately estimating personal UFP exposures. The purpose of this study was to measure UFP I/O ratios in a sufficiently large number of vehicles to develop accurate predictive models for assessing drive-time UFP exposure based on easy-to-obtain information.

4.2 Methods

4.2.1 Vehicle Selection and Ventilation Conditions Tested

Vehicles were selected to provide a wide distribution of age and mileage, which are both important factors affecting AER, albeit highly correlated. See Chapter 2 for more
details on test fleet distribution. In general, the measurement dataset adequately covered the full range of speed, vehicle, and ventilation settings necessary for optimum model performance. Finally, to incorporate both all ventilation conditions and particle measurements under non-steady speeds in this modeling, data from six vehicles tested in Sydney (Australia) (Knibbs et al. (2009; 2010)) was added to that collected in 67 vehicles in Los Angeles. All together, data from measurements performed in 73 vehicles was modeled. Vehicles in were selected from different size categories (sub-compact, compact, mid-size, etc.) in proportions similar to their presence in U.S. fleet. Table 2.1 in Chapter 2 summarizes the vehicle make, model, age and mileage at the time of testing.

Measurements were made with the air conditioning system operating at both ventilation setting options: recirculation (RC), where the in-cabin air is re-circulated, and outside air intake (OA), where outside air is drawn into the vehicle cabin and passed through a filter (if present). Almost all vehicles tested in Los Angeles had cabin air filters in the ventilation loop (i.e. capable of filtering air under both RC and OA). Only two of the vehicles tested in Sydney had filters, and these were of the basic 'pollen filter' type, and filtered OA but not RC air. This resulted in slightly higher I/O ratios in the vehicles tested in Sydney as cabin filters typically reduce I/O ratios by 0.1 or less (Hudda et al., 2011). Overall, I/O ratio was measured for 241 combinations in 43 vehicles (110 at RC and 131 at OA setting).
4.2.2 **PARTICLE CONCENTRATION AND I/O RATIO MEASUREMENT**

In order to maintain steady air exchange rates (AER), measurements were made while driving at near constant speeds that ranged from 20–65 miles h\(^{-1}\). Experiments at speeds up to 35 miles h\(^{-1}\) were conducted around the Rose Bowl, Pasadena – a 3.3 mile continuous loop, where vehicular traffic was light. Measurements at speeds ranging from 55–70 miles h\(^{-1}\) were made on Freeways I-10, CA-60 and I-605 during free flowing traffic conditions. An on-board Global Positioning System (GPS) device (Garmin GPSMAP 76CSC) recorded the location and speed of the car at one second interval. The AER determination procedure is detailed in Chapter 2 (Also, Fruin et al. (2011) and Hudda et al. (2011)) and those for Sydney data in Knibbs et al. (2009). Fruin et al. (2011) have previously demonstrated very good agreement between their predictions of results reported by Knibbs et al. (2009) (Pearson \(r^2 = 0.83\)), despite the use of different tracer gases.

Particle number concentrations measurements were performed with a TSI Model 3007 Condensation Particle Counter (CPC) with a 50% lower size detection limit of 10 nm. Two CPCs were used to measure inside and outside concentrations simultaneously, except for a small number of measurements where inside and outside measurements were conducted in an alternating manner. For those tests, on road concentrations were shown to be stable during the measurement periods. The CPC recording outside concentrations pulled through a meter long conductive silicon tube during mobile tests.
and particle losses were insignificant. Inside concentration measurements required no tubing. Measurements in Sydney were conducted using a single CPC that alternated between inside and outside measurements every 20-25 seconds. Only the last 10 seconds of data in each 20–25 second block was used in further analyses in order to allow for sample clearance time and instrument response.

In-vehicle to roadway concentration ratios were determined as the average value observed for at least 10 minutes of measurement after a stable value had been attained (i.e., a standard deviation less than 5%). Data quality assurance comprised regular flow and zero reading checks. All instruments used were run simultaneously before and after test runs to check for consistency of response and ambient concentrations. All instruments were synced to within one second of the time recorded by GPS.

4.2.3 **Predictive Models**

Models were developed to predict I/O ratios for UFPs under both RC and OA conditions, using the following as candidate independent variables: measured and predicted air exchange rate (AER), ventilation fan (fraction of maximum setting), vehicle age (years), mileage (thousands of miles), speed (miles h⁻¹), manufacturer (United States, Japan or Germany/Other), interior volume (ft³), and the product of coefficient of drag (C_d) and frontal area (A, m²) along with pair-wise interactions between vehicle speed, age, and fan setting, and between C_d and frontal area of the vehicle. Ventilation fan fraction was defined as the ratio of the selected fan setting to the total number of options for fan
setting. For example, if a vehicle had seven fan setting options and was operated at the third strongest option, the fan setting was set to 3/7 (or, = 0.43) in the models.

For I/O ratios, which varied between 0 and 1, a logit transformation (the natural log of \([I/O]/ [1-(I/O)]\)) was used as an outcome in prediction models, often more appropriate for fractions. Predicted values on the original scales can be recovered using the equation \(I/O = \exp \text{ (logit I/O)}/ (1+\exp \text{ (logit I/O)})\).

Because multiple measurements of I/O ratio and AER were performed in each vehicle at different speeds and/or ventilation settings, these repeated measurements were sometimes correlated. This correlation violates the assumption of completely independent observations in multiple linear regression (MLR) models, and MLR models fit to correlated data have unbiased regression coefficients but incorrect standard errors (Diggle et al., 2002). To account for correlated observations, we present results from Generalized Estimating Equation (GEE) models (Liang et al., 1986) for continuous outcomes, with an exchangeable correlation structure (assumes that there is a single, uniform correlation between all repeated measurements from the same vehicle, after controlling for the included predictor variables) and robust sandwich estimates of regression coefficient standard errors (produces valid standard errors even if the covariance structure is mis-specified). MLR estimated regression coefficients were similar to those from GEE and are provided for comparison. Model fit was assessed by adjusted R\(^2\) and by leave-one-vehicle-out cross-validated adjusted R\(^2\), which provides a
more reliable estimate of the predictive ability of the same model fit to a new dataset containing information on different vehicles.

All-subset MLR was used to identify the most important set of predictor variables. From this set, a parsimonious GEE model was developed that included all lower-ordered terms of any interactions or squared variables, had high cross-validated adjusted R², statistically significant predictor effects (α = 0.10), and satisfied linear model assumptions. For each model, significance of an indicator variable for Sydney data was also evaluated. Residuals were inspected to assess model assumptions of linearity, normality, and homoskedasticity.

4.3 RESULTS AND DISCUSSION

4.3.1 IN-VEHICLE-TO-ROADWAY CONCENTRATION RATIOS

The I/O ratios measured under RC conditions were far lower than those under OA conditions due to lower AERs under RC (Hudda et al., 2011). The median I/O ratio value at RC was 0.11 (inter-quartile range: 0.07–0.22) compared to 0.66 at OA (inter-quartile range: 0.53–0.80). The median AER value at RC was 6.0 h⁻¹ (inter quartile range: 3.6–10 h⁻¹) compared to 63 h⁻¹ for OA (inter quartile range: 47–83 h⁻¹). The maximum uncertainty associated with AERs was 7.5%, using root mean square error propagation accounting for both instrument accuracy and stability of continuous measurements for AER measurements. The uncertainties of the I/O ratios were slightly less, 7%, since it is only determined by the stability of continuous measurements for I/O ratio. Figure 4.1
shows the distributions of AER and I/O ratio results and their transformed values, under both RC and OA ventilation mode. The measurements in Los Angeles and Sydney have been differentiated in the sub-figures.

Figure 4.1: Distributions and relationship between measured AER and measured I/O ratios used in models.

4.3.2 PREDICTIVE MODEL FOR LOGIT (I/O) UNDER RC AND OA SETTING

I/O ratios under both ventilation conditions were modeled together, using a binary indicator variable for RC setting, (i.e., variable RC = 1 under RC setting and zero otherwise). The resultant Equations 1 and 2 from the GEE model for predicting I/O ratios under RC and OA conditions are as follows:

Equation 1: Logit (I/O) under RC conditions

\[
\text{logit}(I/O) = -(0.29 + 2.93) + 0.54 \times \text{fan strength} + 0.025 \times \text{speed} \\
+ (0.017 + 0.086) \times \text{age}
\]
Equation 2: Logit (I/O) under OA conditions

\[ \text{logit}(I/O) = -0.29 + 0.54 \times \text{fan strength} + 0.025 \times \text{speed} + 0.017 \times \text{age} \]

This estimated model (Table 4.1) also had good fit and predictive ability (GEE adjusted \( R^2: 0.79 \); GEE cross-validated adjusted \( R^2: 0.76 \) ) using only vehicle age, speed, ventilation fan strength, and ventilation setting. For the I/O ratio modeling, the indicator variable for Sydney was significant (p-value: 0.02), but including this variable did not substantially improve the predictive ability of the model (GEE cross-validated adjusted \( R^2: 0.78 \) ). This variable was omitted in the final model since it would not be useful to other users of the model. After stratifying on ventilation setting, and including fan strength, age and speed as predictor variables (most of which are strong predictors of

Table 4.1: I/O model parameters, confidence intervals and p-values for a model that uses ventilation setting, vehicle speed and age as predicting variables.

| GEE Model          | Estimate | Std.err | Wald     | Pr>|W>|     | Confidence Intervals |
|--------------------|----------|---------|----------|-----------|----------------------|
|                    |          |         |          |           | 2.5%  | 97.5%                |
| Intercept          | -0.29    | 0.19    | 2.4      | 1.20E-01  | -0.65  | 0.078                |
| fan strength       | 0.54     | 0.21    | 6.8      | 9.10E-03  | 0.14   | 0.95                 |
| speed (miles h\(^{-1}\)) | 0.025   | 0.0028  | 81       | <2E-16    | 0.019  | 0.030                |
| age (yr)           | 0.017    | 0.02    | 0.84     | 3.60E-01  | -0.020 | 0.055                |
| RC                 | -2.95    | 0.14    | 468      | <2E-16    | -3.2   | -2.7                 |
| RC X age (yr)      | 0.086    | 0.019   | 20       | 7.00E-06  | 0.048  | 0.12                 |

| MLR Model          | Estimate | Std.err | Wald     | Pr>|W>|     | Confidence Intervals |
|--------------------|----------|---------|----------|-----------|----------------------|
|                    |          |         |          |           | 2.5%  | 97.5%                |
| Intercept          | -0.31    | 0.14    | -2.2     | 3.30E-02  | -0.59  | -0.026               |
| fan strength       | 0.30     | 0.17    | 1.8      | 7.40E-02  | -0.029 | 0.62                 |
| speed (miles h\(^{-1}\)) | 0.033   | 0.0027  | 12       | <2e-16    | 0.028  | 0.039                |
| age (yr)           | 0.042    | 0.011   | 3.9      | 1.30E-04  | 0.021  | 0.063                |
| RC                 | -2.93    | 0.13    | -22      | <2e-16    | -3.19  | -2.67                |
| RC X age (yr)      | 0.06     | 0.02    | 3.9      | 1.30E-04  | 0.031  | 0.093                |
AER) in the models for logit (I/O), additional inclusion of AER itself did not explain an important amount of additional variability in logit (I/O) Models including AER as a variable have been presented in the following section. See Figure 4.2 for predicted logit (I/O) versus measured logit (I/O) plot.

Figure 4.2: Model predicted logit (I/O) versus measured logit (I/O).
Figure 4.3: Predicted values for I/O ratios (from a model that uses ventilation setting, vehicle speed and age as predicting variables) versus two most important model variables for each mode. Bottom subsets show actual measurements versus surface of median model predictions.

Figure 4.3 (a) shows the full range of I/O ratios that can be expected in vehicles up to 20 years old and travelling at speeds up to 75 miles h⁻¹, with age and speed being the most important predictors. Under RC ventilation conditions, I/O ratios can be expected to vary from less than 0.1 to nearly 0.8 in the leakiest vehicles (old and travelling at high speeds). The two surfaces mark the upper (full fan) and lower limits (low fan setting, equal to 0.33) of variation that can be expected due to the third most significant variable, fan strength. Under RC conditions, fan setting was relatively unimportant. For
the entire range of age/speed plotted in Figure 4.3 (a), fan strength made an average
difference of only 0.07 ± 0.02 in I/O ratio.

In contrast, under OA conditions, I/O ratios were most strongly dependent on vehicle
speed and fan speed. I/O ratios were higher but had a smaller range compared to RC
conditions, varying from 0.5 – 0.9 (Figure 4.3 (b)). The plotted surfaces in Figure 4.3 (b)
mark the lower (25th age percentile) and upper bounds (75th age percentile) of predicted
I/O ratios due to variation in the third variable, vehicle age, though the distinction is
barely discernible. Age (by itself) under OA was not significant and made a maximum
difference of 0.03 in I/O ratios predicted using Equation 2. The lower subset figures
show measured I/O ratios plotted along with median predicted surface to show
modeled data fit.

We performed a sensitivity analysis based on maximum expected variable measurement
uncertainty: 5 miles h⁻¹ uncertainty in speed, 1 year in vehicle age, and a 10%
uncertainty in fan speed based on fraction of maximum. These uncertainties led to 8.0,
5.6, and 3.4% difference in I/O ratio, respectively. These relatively modest changes
reflect maximum uncertainties. Typical uncertainties would tend to be smaller, and
since independent, would tend to cancel each other out (i.e., they are just as likely to be
positive as negative). However, the model predictions may be less accurate for vehicles
older than 15 years and at speeds exceeding 60 miles h⁻¹ due to limited coverage of
measured data for such conditions.
4.3.3 *Predictive Model for Logit (I/O) Based on AER*

Additionally, models were fitted for predicting I/O using AER as a predictive variable. The AER input was obtained using Equations 1 and 2 in Chapter 2. The predictive equations were as follows:

**Equation 3: Predictive Model for logit (I/O) under RC conditions using AER input**

\[
\text{logit}(I/O) = -3.37 + 0.017 \times \text{speed} + 0.071 \times \text{age} + 0.30 \times \text{fan strength} + 0.41 \times \ln (AER)
\]

**Equation 4: Predictive Model for logit (I/O) under OA conditions using AER input**

\[
\text{logit}(I/O) = -1.70 + 0.017 \times \text{speed} + 0.020 \times \text{age} + 0.30 \times \text{fan strength} + 0.41 \times \ln (AER)
\]

The GEE coefficients, confidence intervals and p values for both the models are listed in Table 4.2. GEE models built on AER input account 79% of the variability (Cross validated $R^2$ was 0.76), similar to the models built on driving/vehicle inputs. However, the predictions were slightly different due to different input variables. Equation 3 (RC conditions) takes vehicle volume and manufacturer into account but does not require fan strength as an input. Equation 4 (OA conditions) in addition to the variables accounted in Equation 2 also accounts vehicle cabin volume.
### Table 4.2: I/O model parameters, confidence intervals and p-values for a model that additionally includes AER as a predicting variable.

| GEE Model | Estimate | Std.err | Wald | Pr(>|W|) | Confidence Intervals |
|-----------|----------|---------|------|---------|----------------------|
|           |          |         |      |         | 2.5% | 97.5% |
| Intercept | -1.70    | 0.34    | 25   | 5.90E-07| -2.36 | -1.03 |
| fan strength | 0.30    | 0.25    | 1.4  | 2.30E-01| -0.19 | 0.80 |
| speed (miles h\(^{-1}\)) | 0.017  | 0.0028  | 39   | 5.40E-10| 0.012 | 0.023 |
| age (yr) | 0.020    | 0.021   | 0.91 | 3.39E-01| -0.021 | 0.061 |
| RC | -1.6     | 0.36    | 21   | 4.20E-06| -2.34 | -0.94 |
| RC X age (yr) | 0.051  | 0.024   | 4.5  | 3.40E-02| 0.0037 | 0.098 |
| ln(AER) | 0.41     | 0.10    | 17   | 3.80E-05| 0.21 | 0.60 |

| MLR Model | Estimate | Std.err | Wald | Pr(>|W|) | Confidence Intervals |
|-----------|----------|---------|------|---------|----------------------|
|           |          |         |      |         | 2.5% | 97.5% |
| Intercept | -1.36    | 0.486   | -2.790 | 5.63E-03| -2.31 | -0.40 |
| fan strength | 0.071   | 0.192   | 0.370 | 7.12E-01| -0.31 | 0.45 |
| speed (miles h\(^{-1}\)) | 0.029  | 0.003   | 9.030 | < 2E-16| 0.023 | 0.036 |
| age (yr) | 0.040    | 0.011   | 3.720 | 2.50E-04| 0.019 | 0.061 |
| RC | -1.96    | 0.449   | -4.360 | 1.90E-05| -2.8 | -1.1 |
| RC X age (yr) | 0.040  | 0.019   | 2.150 | 3.29E-02| 0.0033 | 0.08 |
| ln(AER) | 0.31     | 0.14    | 2.26 | 2.50E-02| 0.039 | 0.57 |

Either of the models may be used to generate I/O estimate within 0.1, under RC ventilation setting (up to freeway speeds and 75th age percentile), but it is preferable to use the model that includes AER as a predictive variable, as it incorporates previously developed linear combinations of the candidate predictor variables. However, use of the simpler model (Equations 1 and 2) does offer the advantage of a very small set of input variables – only four (ventilation choice, fan setting, age of the vehicle and speed – rather easy to gather for large epidemiological studies as opposed to six for predicting I/O using Equations 3 and 4. Of the additional variables, through manufacturer is easy to obtain, estimation of volume will require knowledge of vehicle model year, model subtype and access to cabin volume information for all or most model types. Under OA
conditions, it is suggested that under conditions of steady speed driving (during non-rush hours on freeway and less frequently signaled arterial roads) Equation 2 be used. Equation 4 should be preferred to predict I/O ratios when travelling conditions included short trips, congested traffic, stop-and-go mode or frequent stops at signals on arterial roads.

![Figure 4.4: Predicted values for I/O ratios (from a model that additionally includes AER as a predicting variable) versus two most important model variables for each mode.](image)

Figure 4.4 shows predictions based on Equations 3 and 4 for two contrasting input variable scenarios under both ventilation conditions. Under both ventilation conditions the two surfaces represent the upper and lower bounds based on vehicle manufacturer and/or volume. I/O ratios were predicted to be 0.4 or higher, using Equation 4, at low or stronger fan settings under OA conditions. Under RC conditions, only the vehicles older
than 75th age percentile (11 years in current U.S. fleet) and travelling at speeds greater than 35 miles h⁻¹ are expected to have I/O ratios over 0.4.

4.4 CONCLUSIONS

Models have been developed for predicting UFP in-vehicle to roadway concentration ratios (I/O) based on simple variables like driving preferences and vehicle characteristics. Inclusion of ventilation fan speed, vehicle age or mileage, vehicle air exchange rate (AER) and driving speed explained greater than 70% of the variability in measured UFP I/O ratios. In general, factors that increase air exchange AER increase UFP I/O ratio, and AER alone explains 66% of the variability in I/O ratios. Vehicle age was significant and positively correlated with I/O ratios under recirculation ventilation setting (RC), but age was not significant under fresh air intake setting (OA). Under OA conditions, fan strength was positively correlated with I/O ratios and the most significant determinant of I/O ratio. Under both ventilation settings, an increase in vehicle volume decreased I/O ratios, indicating vehicle class/model type would be another important variable in exposure analysis.
CHAPTER 5: REAL-WORLD IN-VEHICLE ULTRAFINE PARTICLE EXPOSURE DISTRIBUTIONS USING ON-ROAD MEASUREMENTS

5.1 INTRODUCTION

Exposure to vehicle exhaust has been associated with detrimental health effects in numerous epidemiological studies (HEI, 2010). Highest exposure concentrations and disproportionate fraction of total exposure is likely to occur either during travel (inside vehicles or other enclosed transit) or near roadways (Stein et al., 2007). U.S. travel surveys indicate a 10% increase in commute times between 1995 and 2001 (Hu and Reuscher, 2004). Increasing travel times and emerging evidence linking exposure to vehicular traffic with exacerbation of cardiovascular diseases including triggering myocardial infarction (Peters et al., 2004) among other adverse health outcomes (HEI, 2010), increases the importance of in-vehicle/in-transit microenvironment for exposure studies (Knibbs et al., 2011).

However, accurate exposure assessment in this microenvironment is challenging due to several reasons. Roadway environment is very dynamic. Sharp concentration gradients exist at small spatial scales and local concentrations are strongly dependent on local traffic characteristics like vehicle type mix (e.g., gasoline or diesel fueled engines), driving modes (exhaust composition and concentration levels vary with engine load), fleet age (better control technology is reducing emissions from newer vehicles), etc., in addition to being strongly influenced by local meteorology. This makes concentration
prediction challenging and direct on-road measurements effort intensive and impractical for large cohorts in epidemiological studies. Further, personal exposure assessment not only requires time-activity information but also direct measurements or indirect estimates of not the roadway but within vehicle cabin concentrations.

In enclosed environments like the in-vehicle, vehicle cabin concentration does not always equal the roadway concentration. Pollutant concentrations, especially for particulate species like ultrafine particles (UFP, aerodynamic diameter less than 100 nm) are frequently lower inside vehicles (at multiple ventilation settings) than on roadways because pollutants are often lost during and post infiltration (Hudda et al., 2011; Knibbs et al., 2010). However, they may still exceed ambient concentrations by manifolds (Chan et al., 1991; Rodes et al, 1998; Leung and Harrison 1998). Prior to this work, some studies (Zhu et al., 2007, Pui et al., 2008, Brown et al., 2012) have reported in-vehicle UFP concentrations but results cannot be generalized. No study was identified that can systemically quantify the interaction of roadway concentration variation with vehicle characteristics on in-vehicle concentration.

In general, it is important to identify the most influential parameters and their relative impact in order to assess exposure for epidemiological studies. For example frequently at recirculation (RC) ventilation mode, inside concentrations is half or lower than concentration observed at outside air (OA) intake ventilation mode. A typical two-fold difference in roadway concentration between peak and off-peak traffic hours may
therefore be of minor consequence for exposure assessment if a vehicle is driven in RC mode since vehicle characteristics will overshadow roadway concentration differences. With information of ventilation mode lacking, it would be impossible to draw correct conclusions on in-vehicle concentrations and associate them with any health end points in an epidemiological study.

Correct determination of in-vehicle concentration requires an additional parameter, inside-to-outside ratio (I/O), which can accurately modify roadway concentrations to reflect the combined effects of multiple mechanisms that either limit the pollutant penetration inside vehicles or cause loss therein. For a given vehicle, the I/O ratio depends on ventilation setting preferences and driving speed. Furthermore, the inter-vehicle difference in I/O, due to age and make/manufacturer type variation is very significant. As a result, I/O ratios can vary from nearly zero to one; both within an individual vehicle and within a fleet.

This doctoral work focused on establishing a link between roadway concentrations and in-vehicle concentration. This chapter demonstrates the effects of that association for UFP by combining previously developed I/O ratio models (Chapter 4) with real on-road measurements to generate in-vehicle concentration estimates. As a proof-of-concept and a demonstration of model application and scalability, in-vehicle UFP concentration distribution were generated for both individual vehicles and a typical fleet,
corresponding to real-world on-road concentrations measured in Los Angeles. This technique is applicable to other pollutants if I/O ratios are known.

5.2 METHODS

5.2.1 INSTRUMENTS AND SAMPLING ROUTES

A hybrid vehicle (2010 Honda Insight) was used as a mobile measurement platform to measure on-road concentrations. UFP concentration was measured using a Condensation Particle Counter (CPC Model 3007, TSI Inc., MN, USA) which sampled air from a duct installed across the rear windows. Instrument was periodically calibrated and time was synchronized to be within 1 second of the Global Positioning System (GPS) device time (Garmin GPSMAP 76CSC). Quality assurance procedures included regular flow and zero reading checks.

To generate UFP concentration distributions expected on roadways, on-road concentrations were measured on five Los Angeles freeways – CA SR-110, I-110, I-405, I-710 and CA SR-91, and 50 mile length stretch of arterial roads. The mobile measurement platform was driven in the central freeway lane, when possible. In general, sampling periods were well distributed to cover both rush and non-rush hour activity.
5.3 RESULTS AND DISCUSSION

5.3.1 EFFECT OF VARIABLE SPEED AND ROADWAY TYPE ON I/O RATIO

During real-world driving, speeds are often unsteady and vary mainly due to differences in roadway type being driven (unrestricted access freeways versus restricted access arterial roads) and traffic intensity (free flowing during off-peak hours versus congested during peak traffic hours). Since I/O ratios are dependent on and positively correlated with vehicle speed, variable speed during trips results in a distribution of I/O ratios for the trip rather than a singular value. This follows from the unsteady and varying AERs. The I/O ratio distribution reflects the variation in speed if ventilation conditions are unchanged during the trip.

To investigate the influence of unsteady speed on I/O ratios during real world driving, I/O ratio distributions were calculated for typical speed frequency distribution during peak (4–5 PM) and off-peak traffic hours (4–5 AM) for both roadway types – freeways and arterial roads. The probability distributions for speed are plotted in Figure 5.1 and are same as the input distributions in EPA Motor Vehicle Emission Simulator (MOVES), with 16 speed bins covering 0–75 miles h\(^{-1}\) range (http://www.epa.gov/otaq/models/moves/tools/averagespeedconverter_mobile6.xls). The hours plotted had the highest contrast. The weighted average speeds for arterial roads during peak and off-peak traffic hours were 27 and 30 miles h\(^{-1}\), respectively, and 27 and 52 miles h\(^{-1}\) for freeways.
Higher speeds during off-peak traffic hours resulted in higher I/O ratios, and the difference in average I/O ratio (frequency weighed) was less than 0.05 on arterial roadways, under both RC and OA ventilation modes. This difference was up to or less than 0.15 for freeways, resulting from a greater difference in peak and off-peak traffic speed distribution on freeways. The resulting I/O ratio distributions are plotted in Figure 5.1 for a vehicle of median age (7 years for U.S. fleet, EPA 2010) operating at medium fan (fan strength = 0.5).

![Figure 5.1: I/O ratio distribution in a median aged vehicle operating at medium fan setting corresponding to typical distribution of driving speed on urban freeways and arterial roads during peak (4-5 AM) and off-peak (4-5 PM) traffic conditions.](image)

In absolute terms, the difference in I/O ratios during peak and off-peak hours was greater for older vehicles, higher fan settings and under OA ventilation condition. However, in terms of percentage change in I/O ratio, the trend was reverse. Most
significant change in I/O ratios was observed under RC ventilation mode for new vehicles operation at low fan settings. Average I/O ratios are summarized in Table 5.1 for other vehicles (25th and 75th age percentiles) and ventilation fan settings (low and full fan).

Furthermore, the disparity between freeway and arterial I/O ratios was greater at RC than OA and the difference between average I/O on freeways and arterial roads during peak traffic hour was negligible (about 0.01) due to little or no difference in speed distribution on these two roadways during rush hours. However, during off-peak traffic hours the difference was significant, about 0.07-0.08. This has important implications for exposure assessment. During commute hours, which generally correspond to peak traffic hours, the aspect of roadway type that is of greater consequence and will drive the concentration variability is the difference in pollutant concentration and not the travel speed variability resulting from different roadway types. However, during off-peak hours, the differences in travel speed on different roadway types will significantly contribute to the variability in concentrations.

5.3.2 EFFECT OF CHANGE IN VENTILATION SETTING ON I/O RATIOS

Even after integrating the effect of unsteady speeds (by weighing I/O ratios by their frequency), the most significant difference in resultant I/O ratio is caused by ventilation setting choice – recirculation versus outside air intake. For typical speed distributions on freeways and arterial roads during all hours of the day, the range of I/O ratios under
each ventilation mode did not overlap for vehicles up to 15 year old and speeds up to 75 miles h\(^{-1}\). I/O ratios are expected to vary between 0.1–0.5 under RC conditions and 0.6–0.8 under OA conditions.

Figure 5.2: Average I/O ratios for vehicles ages spanning the inter-quartile range of US fleet and full range of ventilation fan setting for rush hour related speed distribution under both ventilation modes.

Table 5.1: Average I/O ratios for typical speed distributions during peak and off-peak traffic hours on urban arterial roads and freeways.

<table>
<thead>
<tr>
<th>Arterial Roads</th>
<th>Fan Traffic</th>
<th>Low (20%)</th>
<th>Medium (50%)</th>
<th>High (100%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Non-rush</td>
<td>Rush</td>
<td>Non-rush</td>
<td>Rush</td>
</tr>
<tr>
<td>RC Age: 2</td>
<td>0.12</td>
<td>0.10</td>
<td>0.13</td>
<td>0.11</td>
</tr>
<tr>
<td>Age: 7</td>
<td>0.18</td>
<td>0.16</td>
<td>0.21</td>
<td>0.18</td>
</tr>
<tr>
<td>Age: 11</td>
<td>0.25</td>
<td>0.22</td>
<td>0.28</td>
<td>0.24</td>
</tr>
<tr>
<td>OA Age: 2</td>
<td>0.67</td>
<td>0.63</td>
<td>0.71</td>
<td>0.66</td>
</tr>
<tr>
<td>Age: 7</td>
<td>0.69</td>
<td>0.65</td>
<td>0.73</td>
<td>0.68</td>
</tr>
<tr>
<td>Age: 11</td>
<td>0.71</td>
<td>0.66</td>
<td>0.74</td>
<td>0.70</td>
</tr>
</tbody>
</table>

Freeways

<table>
<thead>
<tr>
<th>Ventilation Condition</th>
<th>Fan Traffic</th>
<th>Low (20%)</th>
<th>Medium (50%)</th>
<th>High (100%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Non-rush</td>
<td>Rush</td>
<td>Non-rush</td>
<td>Rush</td>
</tr>
<tr>
<td>RC Age: 2</td>
<td>0.17</td>
<td>0.11</td>
<td>0.19</td>
<td>0.12</td>
</tr>
<tr>
<td>Age: 7</td>
<td>0.25</td>
<td>0.16</td>
<td>0.29</td>
<td>0.18</td>
</tr>
<tr>
<td>Age: 11</td>
<td>0.34</td>
<td>0.22</td>
<td>0.37</td>
<td>0.25</td>
</tr>
<tr>
<td>OA Age: 2</td>
<td>0.76</td>
<td>0.62</td>
<td>0.78</td>
<td>0.65</td>
</tr>
<tr>
<td>Age: 7</td>
<td>0.77</td>
<td>0.64</td>
<td>0.80</td>
<td>0.67</td>
</tr>
<tr>
<td>Age: 11</td>
<td>0.78</td>
<td>0.65</td>
<td>0.81</td>
<td>0.68</td>
</tr>
</tbody>
</table>
5.3.3 Fleet-wide distributions of AER and I/O ratio

To calculate individual in-vehicle UFP exposures, the models presented in Chapters 2 and 4 for prediction of AER and I/O ratios require six inputs: (a) ventilation mode; (b) fan setting; (c) manufacturer; (d) vehicle age; (e) vehicle volume; and (f) speed. For individual vehicles, all the vehicle related inputs (manufacturer, age, volume) would be singular fixed values. Ventilation setting related inputs (ventilation mode and fan setting), can be determined even if they vary during the trip through driving habit questionnaire. Lastly speed, which is expected to be unsteady and variable, too can be determined either through measurements or data made available by transportation agencies. But the variability in speed leads to a spread in I/O ratios, as opposed to a singular average value.

However, at fleet wide level all the required input variables would either have multiple possibilities (for example, three distinct manufacturers were identified as being significant in the models) or distributions (for example, age or volume of the vehicles). Each predictive variable will contribute a significant spread in I/O ratios expected in the fleet, and consequently UFP concentrations. Both population-size distributions of in-vehicle UFP exposure and the distribution of predicted AER and I/O ratios in a fleet of vehicles can be computed using input variable distributions. As an example, probability distributions for AER and I/O ratio (predicted using Equations 1 and 2 in both Chapter 2 and 4) were computed for a fleet of sedan type vehicles.
EPA uses a standard vehicle size class definition based on cabin volume to classify vehicles (http://www.fueleconomy.gov/feg/info.shtml). The fraction of US fleet in each of three classes is available in summarized reports (http://www.epa.gov/otaq/fetrends.htm). Passenger cars are classified into three broad categories based on cabin volume: small, mid-size or large. Since the fraction of US fleet in each of these broad categories varies with age of the fleet, so will the average volume. However for simplicity, through the variation in fractional distribution for each year was taken into consideration, the size class average cabin volume was assumed to equal the average for the entire period. Regression on 1981-current year data results in average volume for each of these categories, which equals 99 ft$^3$, 112 ft$^3$ and 135 ft$^3$, for small, mid-size and large sized vehicles. Volume distribution for the last decade is shown in Figure 5.3. The fraction of passenger car fleet not shown in Figure 5.3 comprised of vans and truck and only sedan-type vehicles or passenger car were considered for this analysis.
For fan setting, it was arbitrarily assumed that an equal fraction of vehicles were being driven at three fan settings, low (fan setting = 0.33), medium (0.67) and highest (1.0).

The current fractions of manufacturer share in U.S. car market were used (44.5% U.S., 42% Japanese and 13.5% German/other). These fractional distributions can be further resolved for each year of the fleet. For age, EPA MOVES default age class bins (of one
year increment) and fractional distribution of fleet in each bin for passenger cars were used as inputs for age distribution (http://www.epa.gov/otaq/models/moves/tools/reg-distrib-converter-veh16-20100209.xls). Age distribution of U.S. fleet over the last decade is plotted in Figure 5.4. For comparison, the distribution of vehicles in the test fleet for developing the models presented in Chapters 2 and 3 has also been plotted. The aforementioned distributions were coupled with speed distributions (discussed in Section 5.3.1) for arterial roads and freeways for two different traffic intensity scenarios i.e., peak and off-peak traffic hours. Results are interpreted presuming that the changes in traffic intensity only affect travel speed for the vehicle fleet and no other model inputs.

Figure 5.5: Distribution for AER and I/O ratio for a fleet similar to U.S. passenger car fleet in terms of manufacturer’s market share, vehicle volume and age.
The resulting fractions of vehicles having a specific AER or I/O ratio are plotted in Figure 5.5 for both ventilation choices (RC and OA). Several important observations can be made from Figure 5.5. Foremost, at fleet-wide level even though roadway type and associated speed differences affect AER and I/O ratios, the most significant difference occurs due to ventilation setting choice. Further under RC conditions, 80% of the fleet is expected to have I/O ratio between 0.15 and 0.5—significant protection—under all road types and speeds, but for OA conditions, 80% all vehicles are expected to have I/O ratios from 0.65 to 0.85, only moderately reduced concentrations. Looked at another way, under RC conditions, the fraction of vehicle fleet that will experience cabin concentrations lower than half of on-road concentrations exceeds 80%, but virtually none of the fleet is expected to have I/O ratios less than 0.5 under OA conditions.
5.3.4 **EXPECTED IN-CABIN CONCENTRATIONS FOR GIVEN ROADWAY CONCENTRATIONS**

The ultimate goal of generating predictive models for I/O ratios is to be able to predict in-cabin concentrations from roadway concentrations (calculated as Concentration\textsubscript{in-cabin} = I/O x Concentration\textsubscript{roadway}). To illustrate, representative probability distributions of UFP concentrations were generated from sampling on arterial roads and freeways in Los Angeles and are shown in Figure 5.6 during the peak traffic period. In turn, these distributions were joined with the I/O ratio distributions in Figure 5.5 to generate distributions of UFP concentrations inside the U.S. vehicle fleet if driven on those Los Angeles roads.

![Figure 5.6: Expected in-cabin concentration for US like vehicle fleet travelling on Los Angeles arterial roads and freeways](image)

Comparison of the measured roadway concentrations and the predicted in-cabin concentrations under RC and OA conditions shown in Figure 5.6 suggests that for the
range of fleet vehicle characteristics such as age and mileage (e.g., 25th to 75th percentile differences for a ventilation setting and road type), we would expect a two to three-fold range in in-vehicle UFP exposures (more under RC than OA), while the differences due to ventilation mode selection alone for a given vehicle on either road type were larger, with factors ranging from two to four. The increase in speed going from arterial to freeway speeds, along with increase in on-road concentrations on freeways, increased in-vehicle UFP exposure for a given vehicle at either ventilation mode by nearly three-fold. Therefore, in a real-world setting, we observe that 1) ventilation setting, 2) roadway type (arterial or freeway) and 3) the totality of vehicle and driving characteristics (age, manufacturer and speed) each are all of roughly similar impact on in-vehicle UFP concentration. Significantly lower exposures occur during RC ventilation and driving on arterial roads. UFP concentrations on arterial roads at OA ventilation setting was similar to driving on freeways at RC ventilation setting.

5.4 CONCLUSIONS

Scalability of the models developed during this doctoral work was demonstrated at fleet-wide level and in dynamic roadway environments. For exposure assessment, the most important parameter would be the ventilation setting mode preference, followed by other vehicle and driving characteristics. If more than one ventilation mode is used frequently or comparably, an approximate division of time between them would help in
accurate determination of exposure level. Given the relatively moderate influence of variation in trip speed on the I/O ratios, using an average speed for the trips, as opposed to finely resolved actual measurements of subject’s trip, would result in I/O ratios that are accurate to within 0.1, under most frequent real-world driving conditions.

On combining I/O results with on road UFP concentration distributions measured on Los Angeles roadways, in-cabin UFP concentrations during freeway driving were up to three times that of arterial driving, but the switch from OA ventilation condition to RC dropped the in-vehicle concentration on either road type two- to four-fold. Since, during rush hour traffic the speed distribution on both freeways and arterial roads can be similar; the difference in I/O ratios resulting from driving on different roadway types will be most consequential during off-peak hours. Furthermore, I/O ratios are higher during off-peak hours when travel speeds are higher. However, the positive difference in roadway concentrations (if higher during periods of peak traffic) may more than compensate for the lower I/O ratios at lower speeds. In such a case, roadway concentration differences will drive in-vehicle concentration variation.
CHAPTER 6: PRINCIPAL FINDINGS, APPLICATION AND FUTURE RESEARCH

6.1 PRINCIPAL FINDINGS

The principal findings of this thesis work are as follows:

I. Despite its complexity, in-vehicle microenvironment can be accurately characterized in terms of air exchange rate (AER) and ultrafine particle (UFP) vehicle-to-roadway concentration (I/O) ratios.

II. The foremost determinant of in-vehicle concentrations is the vehicle AER. AER is responsible for influx of pollutants from the roadway into the vehicle and also influences the rate of particle removal within the vehicle.

   a. AER depends on multiple factors, though primarily influenced by vehicle’s ventilation mode setting. The influence of other factors on AER is disparate with respect to ventilation mode.

   b. Under recirculation (RC) ventilation mode, the key determinants of AER are speed and age of the vehicle, followed by vehicle volume. AERs varied from < 2 h⁻¹ in stationary vehicles to about 50 h⁻¹ in oldest vehicles (over 15 year) at highest speeds tested (65–70 miles h⁻¹). The influence of ventilation fan setting on AER under RC was non-uniform across the test fleet.
c. Under outside air (OA) intake mode, the key determinant of AER is the ventilation fan setting. In comparison, other factors – driving speed and vehicle volume, have a relatively moderate influence. The influence of vehicle age on AER under OA was unclear, but a positive correlation is likely.

d. AER under OA conditions exceeds that under RC condition by an order of magnitude or more within the same vehicle.

e. When windows were kept open, AER is even higher than under OA mode and in-vehicle concentrations equal roadway concentrations.

III. At AERs from less than 2 h\(^{-1}\) to up to 150 h\(^{-1}\) measured in this study, in-vehicle UFP concentrations are still on average lower than roadway concentrations and resultant in-vehicle-to-roadway concentration or inside-to-outside (I/O) ratios were less than one. It is so because multiple mechanisms either limit the UFP penetration inside vehicles or cause loss therein via surface deposition, filtration etc.

IV. UFP I/O ratios measured in the study varied from nearly zero to almost one, and were highly positively correlated with AER.

a. The median I/O ratio value at RC was 0.11 (inter-quartile range: 0.07–0.22) compared to 0.66 at OA (inter-quartile range: 0.53–0.80).

b. AER alone accounted for over 66 % of variation in I/O ratios.
c. Further inclusion of other variables like age of vehicle, speed, ventilation fan strength in predictive models explained over 70% of variability.

d. Similar to AER, influence of certain variables (age, fan strength) was disparate under the two ventilation modes.

e. Presence, absence or loading level of cabin air filter did not influence the UFP I/O ratios significantly, in the vehicles tested. However, significant UFP losses still occur in the ventilation system, likely via surface loss mechanism.

f. For particles in the 25 – 200 nm size range, little difference in I/O ratios was observed owing to difference in size. Number concentration weighed average I/O was within 0.1 of size resolved I/O ratio.

V. Predictive models for AER and I/O ratios require variable inputs that are obtainable through questionnaire.

a. These variables, namely, vehicle age (or mileage), vehicle make and manufacturer (volume can be found based on these), average driving speed, ventilation mode and fan setting preference, can be easily obtained.

b. These models will be useful to exposure and epidemiological studies, since measurements in individual vehicles for large cohorts are not feasible and cost prohibitive.
VI. For a fleet similar in its age, cabin volume, make and manufacturer distribution to U.S. passenger car fleet, and travelling in UFP concentration environments similar to major Los Angeles freeways and arterial roads

a. I/O ratios will be higher if vehicle is driven on freeways than on arterial roads, because of higher driving speeds.

b. In-cabin UFP concentrations during freeway driving were up to three times that of arterial driving.

c. A switch from OA ventilation condition to RC reduces the in-vehicle concentration on either road type two- to four-fold.

d. Using an average speed for the trips, as opposed to finely resolved actual measurements of subject’s trip, would results in I/O ratios that are accurate to within 0.1, under most frequent real-world driving conditions.

e. Higher speeds during off-peak traffic hours resulted in higher I/O ratios.

However, this does not necessarily imply higher resultant in-cabin concentrations. Higher pollutant concentration during peak traffic hours may more than compensate for lower I/O ratios.

VII. Accurate assessment of risk posed by UFP exposure will depend on the ability to characterize exposure during microenvironments like in-vehicle where peak, disproportionate, and widely varying exposure occurs.
6.2 Application of this Work to Ultrafine Exposure Assessment

6.2.1 Applications

This thesis work enables reliable prediction of UFP concentration in the in-vehicle microenvironment. It is also the first work that makes it possible for researchers to estimate vehicle AERs or UFP I/O ratios instead of having to measure them directly. This work can be directly applied to estimate exposure concentration in at least two distinct sizes of subject populations. First, these models can be used in studies that investigate exposure related hypothesis at population-mean levels. For example, studies that investigate the difference in exposure resulting from mobility patterns or travel mode choice (vehicles vs. rails or buses). Second, these models can be used to assess personal exposure of individuals in an epidemiological cohort. One example might be a study comparing UFP exposures of pregnant women who stay at home to pregnant women who work outside the home and have higher UFP exposures because of their commute times.

Though the application of these particular models is limited to in-vehicle microenvironment, certain findings have relevance to multiple traffic-related pollutants. For example, the relation between AER and I/O ratios for other pollutants with decay rates related to surface losses (e.g., black carbon, PM$_{2.5}$, particle-bound PAHs, etc) is expected to be as predictable as it is for UFP. Additional similarities between AER and I/O ratios may also extend to other indoor environments if AER can be sufficiently accurately determined. For example, a recent study by Breen et al., (2010), determined
home AERs based on questionnaire data and meteorology. It should be pointed out that additional research is needed to produce accurate models to predict on-road concentrations, particularly on arterial roads. The models described in this thesis depend on knowing the on-road concentration, and uncertainties in the on-road concentrations transfer directly to in-vehicle concentration uncertainty. Though roadway concentrations can be measured with relative ease, as has been demonstrated by many studies that use mobile platforms, it would be more cost effective to have models that accurately predict roadway concentrations than need to measure the roadways driven by any epidemiological cohort, and that is one next logical research problem that should be addressed as an extension of this thesis work.

6.2.2 Gaps in Extant Literature

Certain critical gaps impede the accurate assessment of total exposure to UFP. Foremost is the lack of good predictive models for roadway UFP concentration under all conditions including driving on smaller roadways. Arterial roadways account for about half of in-vehicle travel and no study has been identified that addresses arterial roadways.

Other than roadway concentration, ambient concentration determination is another important aspect of total UFP exposure assessment. Primary UFP concentrations can vary widely at small spatial scales (within one or even several miles) in locations with high traffic intensity (Moore et al., 2009) while secondary UFP from photochemistry can
exhibit homogeneity at large spatial scales (over tens of miles) (Hudda et al., 2010). At both spatial scales they exhibit significant diurnal and seasonal variations. As a result, the spatial heterogeneity varies by time of day and season, and also inhibits the use of few stationary monitoring site data as measures of ambient. Measurement of accurate subject level ambient UFP concentration for an epidemiological study would prove to be expensive and inefficient.

The one microenvironment where ambient concentration plays the most critical role is the outdoor microenvironment, where about 5% of daily time is spent (Jenkins et al., 1992). For subjects residing in close proximity of roadways, roadway concentration would be of greater significance than ambient for assessing exposure concentration in outdoor microenvironment. For subjects living further away from roadways, ambient concentrations are likely to be low and exposure to them might not constitute a significant fraction of total exposure. It remains to be determined how important a role ambient concentration will play and if it is reasonable to accept high uncertainty in ambient concentration.

However it is likely, and Wallace and Ott (2011) results suggest that the highest variability in UFP concentration is associated with the indoor microenvironment – be it home or work. Since most of peoples’ time is spent indoors, it is the one of the most crucial microenvironments to assess accurately. Presence of multiple sources of UFP, and their varying importance and prevalence on subject-to-subject basis complicates
characterization, but it is possible. Multiple studies have measured UFP concentration in indoor environments due to common sources like cooking, vacuuming, printing etc., and emissions from these sources can be modeled. For example, emission from cooking appliances can be treated similar to vehicle emission factors and then correlated with AER and home volume to estimate exposure concentrations. In summary, avenues exist for characterizing indoor environment for assessing UFP exposure, that do not rely solely on individual measurements.

6.2.3 **RECOMMENDATIONS FOR FUTURE RESEARCH**

Several lines of research pursuits immediately follow the work presented in this thesis. First, it is reasonable to conclude that if UFP I/O ratios can be accurately predicted from easily-obtained information about vehicle characteristics and driver behavior, I/O ratios for other particulate species that are less dynamic and volatile than UFP, like PM$_{2.5}$, black carbon and particle bound polycyclic aromatic hydrocarbons (PB-PAH) can also be accurately modeled for exposure estimation during in-vehicle travel. It is suggested that these tests first be conducted in a careful but small selection of vehicles (similar to Chapter 3). Preliminary tests for PM$_{2.5}$ indicate that the range of I/O ratios is very narrow and will be determined by ventilation mode and filtration efficiency; BC and PB-PAH (which correlate well with UFP under most circumstances) I/O ratios are most significantly determined by ventilation setting choice. Along with establishing these relationships, modeling of roadway concentrations should also be pursued. Arterial
roadways in particular have been little studied and measurements indicate a much more transient and challenging set of conditions are necessary to characterize. Finally, it is now possible and would be interesting to implement the models described in this thesis to estimate UFP exposures for a commuter cohort and evaluate the possible association between UFP exposure and health outcomes.
BIBLIOGRAPHY


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