Inhalation of Vehicle Emissions in Urban Environments

by

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B.S.E. (Princeton University) 1996
M.S. (University of California, Berkeley) 2002

A dissertation submitted in partial satisfaction of the

requirements for the degree of

Doctor of Philosophy

in

Energy and Resources

in the

GRADUATE DIVISION

of the

UNIVERSITY OF CALIFORNIA, BERKELEY

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Spring 2005

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ABSTRACT

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This dissertation explores the relationship between motor vehicle emissions and the human inhalation intake of these emissions. Motor vehicles are ubiquitous to urban areas throughout the world. In most urban areas, vehicle emission are a significant contributor to air pollution problems. Inhalation of vehicle emissions has been shown to cause a number of adverse health effects. Better understanding of the relationship between emissions and inhalation will aid in designing effective strategies to reduce air

pollution health effects. Understanding the emission-to-inhalation relationship is also important for estimating the total health impacts attributable emissions from a specific air pollution source, such as motor vehicles in a specific city.

Three objectives of his dissertation are (1) to quantify the emission-to-inhalation relationship in a way that is useful to other air quality and health researchers and to policy analysts; (2) to employ a variety of analytic approaches, in order to understand better the relative strengths and weakness of each approach; and (3) to demonstrate that the conclusions one draws from air quality analyses may depend on whether one uses as the figure-of-merit inhalation of air pollution, as is done for analyses in this work, or other common air quality metrics such as mass emission rate, ambient concentrations, or concentration at the location of the maximally exposed individual.

The methods employed here include several data analysis and modeling approaches. The data analyses incorporate a range of inputs, including results from air dispersion models of varying sophistication, tracer-gas experiments, and "tracers of opportunity" (gases that are emitted primarily by one source or source category). The inhalation model that I develop in Chapter 6 simulates the movement of people through an urban area, tracking the individual or population inhalation rate during simulated activities (e.g., shopping, driving, cooking).

The specific research topics considered in this work are as follows. In the first portion of this dissertation (Chapters 1 – 5), I generate estimates for a summary inhalation metric, called intake fraction, for vehicle emissions in urban areas. In the second portion (Chapters 6 – 7), I first develop a mobility-based GIS inhalation model for

urban air pollution, and then, separately, consider how changes in urban population and land area would influence population inhalation of private passenger emissions.

Intake fraction is the fraction of emissions from some source that are cumulative inhaled by an exposed population. As an example of how intake fraction values might be used, the health effects attributable to an emissions source or source category can be estimated as the product of the mass emission rate, the intake fraction (mass inhaled per mass emitted), and the toxicity (impact per mass inhaled). Intake fraction values will, in general, vary over time and among source categories, source locations, and pollutants. Intake fraction can be used as a potential basis to rank sources when prioritizing emission control strategies. All else being equal, the public health benefit per amount of emission reduced would be larger for an emission source with a large intake fraction than for a source with a small intake fraction.

In Chapter 1, I provide background and framing for the research. In Chapter 2, I explore the use of intake fraction. This chapter gives examples of how and why intake fraction varies among sources; discusses how this variability may be exploited to increase the health effectiveness of air pollution policy; and, illustrates types of intake and intake fraction analyses one might carry out, depending on the information available.

In Chapter 3, I provide an estimate of the intake fraction for nonreactive vehicle emissions in California's South Coast Air Basin (SoCAB). This estimate incorporates several inputs, including (1) measured ambient concentrations of benzene and carbon monoxide (CO), two pollutants that are primarily emitted by motor vehicles; (2) US Census data indicating population densities throughout the SoCAB; (3) time-activity pattern data indicating the amount of time people spend indoors, outdoors, and inside

vehicles; (4) microenvironment factors for benzene and CO, which indicate how concentrations may differ in specific microenvironments as compared to the nearby ambient concentration; and (5) the time-varying population-average breathing rate. The annual average intake fraction for nonreactive gaseous vehicle emissions in the SoCAB is estimated as 48 per million, with an uncertainty of ~ 33%. This value means that 48 g of emissions are collectively inhaled by the population per million g emitted.

The SoCAB is an important case study, in part because of its large population size (~15 million people, or 1 in 19 US residents). It is also atypical of US urban areas because of its high population density (~860 person km^{-2}) and because of the typically poor air dispersion owing to relatively frequent low inversion heights. In Chapter 4, I estimate the central tendency and main range for the intake fraction of vehicle emissions in urban areas throughout the US. I employ three independent approaches. First, I use a one-compartment mass-balance model of an urban area to combine meteorological data on wind speed and mixing heights with demographic data on urban population and land area. Second, I use a statistical model that relates observed ambient concentrations of carbon monoxide (CO) to motor vehicle emission factors for CO. Third, I evaluate model input and output for the US EPA's National-scale Air Toxics Assessment (NATA), the EPA's main nationwide air dispersion and exposure model. These approaches incorporate measurements and models, and range in analytic complexity from straightforward to sophisticated. There is broad consistency among the results that these approaches yield. Combining the results of these three investigations, I estimate that the population-weighted annual-average mean intake fraction for nonreactive gaseous vehicle emissions in US urban areas is ~ 14 per million, with a confidence interval of ~ 50%. This value is

about three times lower than the intake fraction value of 48 per million estimated for the SoCAB.

These annual average intake fraction values (48 per million for the SoCAB and 14 per million for the mean value among US urban areas) represent fleet-wide average values. The intake fraction of emissions from a specific vehicle or class of vehicles will, in general, differ from this fleet-wide average. For example, all else being equal, the intake fraction of nonreactive vehicle emissions will be higher if a vehicle operates and emits pollution near population centers (e.g., neighborhood delivery trucks) than if it operates and emits pollution along rural highways. Another reason why intake fraction may differ among vehicles is "self-pollution," which occurs when a portion of the emissions from a vehicle migrate to inside that vehicle. Experiments were conducted by others, wherein a tracer gas (SF_6) was injected at a known flow rate into the exhaust manifold of a school bus, and at the same time concentrations of SF_6 were recorded inside the bus. Six buses, representing a range of vehicle ages, were tested with windows open and closed, along actual school bus routes in the SoCAB. In Chapter 5, I analyze results from these tracer gas experiments to estimate children's school bus self-pollution intake fraction, i.e., the fraction of a school bus's emissions that are inhaled by students riding on that bus. The average value across the six buses and all bus runs is 27 per million; values were higher with windows closed rather than open, and for older rather than newer buses. When considering the emissions from a specific bus, the mass of pollution collectively inhaled by students on that bus is comparable to, and in many cases greater than, the mass of pollution collectively inhaled by all other residents in an urban area.

In Chapter 6, I develop a GIS-based inhalation model for the SoCAB, and investigate the importance of mobility on estimated inhalation rates for several air pollutants. This investigation represents a new and promising approach for inhalation intake analyses. The four main inputs to the model are (1) spatially and temporally disaggregated estimates of ambient concentrations of specific air pollutants; (2) geo-coded time-location-activity survey data indicating individuals' location (latitude and longitude) throughout the day; (3) microenvironment factors, which account for differences between the estimated ambient concentration and the exposure concentration attributable to outdoor pollution in locations such as indoors, outdoors, and in-vehicle; and (4) breathing rates, which vary by age, gender, and activity level. Model output is the estimated inhalation intake rate (μg d^{-1}) for each modeled pollutant and each simulated person-day. Results indicate that mobility influences daily intake rates by less than a factor of two for most individuals. I also explore how inhalation intake rates differ among ethnic and income groups. For the five pollutants considered in this chapter, differences in median intake rate vary by 10 – 60% among the four ethnic groups considered (White, Hispanic, African-American, and Asian/Pacific Islander).

In Chapter 7, I explore the impact of urban population and land area on inhalation intake of vehicle emissions. Changes in population density can impact emissions, owing to changes in average daily distance traveled per person, and also intake fraction, owing to changes in the proximity between people and emissions. The main research question I consider in this chapter is the following: if increasing population density reduces emissions but increases intake fraction, does per capita inhalation intake of vehicle emissions increase or decrease with increasing population density? The research

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approach employs a one-compartment mass-balance model as an archetypal representation of a hypothetical urban area. This approach clarifies underlying relationships, aids in elucidating causal connections, and permits the problem to be analytically tractable. I find that the impact of population density on inhalation intake of vehicle emissions depends on how much emissions change in response to a change in population density (the "density-emissions elasticity"). To use infill development (i.e., increase population density) to reduce inhalation of vehicle emissions, urban planners must strive to achieve large magnitude density-emissions elasticity values, so that vehicle emissions are significantly reduced by density increases.

In Chapter 8, I provide a summary of this dissertation, suggestions for future research, and concluding remarks. Overall, this dissertation presents new information and new ways of thinking about the relationship between vehicle emissions and inhalation of these emissions. The tools developed and results presented may be useful in health risk assessment, in policy and economic analyses such as cost-benefit and cost-effectiveness analyses, in considering the goals and impacts of transportation and land-use planning, and in designing effective intervention strategies to reduce the health effects of atmospheric pollutants. The emerging field of exposure science has developed tools, metrics, and approaches that are ready to be integrated more fully into air quality research and management. I make several specific suggestions are made for future investigations. For example, the diurnal profile of population breathing rate is important for determining daily inhalation intake of air pollution, yet there is little information available from which to estimate this parameter. Previous research has explored the influence of urban population density on distance traveled by private passenger vehicles. Given the

importance of diesel emissions to air quality and health, this same parameter should be

estimated for diesel vehicles.

ACKNOWLEDGMENTS

This dissertation represents the culmination of several years of effort. I would not have been able to make it here without the significant support others have given me over the years. I am indebted to many people.

To my dissertation committee, thank you. This group includes Professors Rob Harley, Betty Deakin, Tom McKone, and Bill Nazaroff. All four have offered guidance and feedback as I formulated and carried out the research summarized here. The insights I have gained are in large part because of their inputs. Thank to Betty especially for encouraging me to broaden my dissertation by exploring urban planning questions (Chapters 6 and 7 of this dissertation). I began working with Tom and Bill in my first year at Berkeley, and it has been a wonderful experience. In truth, I cannot image a better pair of advisors. I have learned from their ability to ask insightful, probing questions that demand serious scholarship to address, yet address real-world issues. I feel fortunate to have worked with two such broadly curious scholars, who have challenged me to think deeply about a variety of topics, and who embody the interdisciplinary that first attracted me to my home department, the Energy and Resources Group. I have learned from them to tackle problems at all levels – from large-scale issues such as global sustainability, to the physics and biology of what happens to air pollutants after they are inhaled. Like most students, I began my PhD hoping to find a topic that was relevant and worthy of my attention for several years, so that my dissertation would be of use and of interest to others. I thank them for their help, input, and encouragement towards this goal. I am

especially indebted to Bill, my dissertation chair, for his excellent mentorship over the years. I thank him for being demanding and nurturing; both serious and light-hearted, and above all for his selfless devotion to mentoring his apprentices. Bill leads by example; the care and attention he gives to all of his professional activities, from mentoring student to crafting research articles to delivering seminars and class lectures, will continue to be a source of inspiration to me throughout my career. I owe much of my scholarly accomplishments to his support and advising, and I am deeply grateful.

Several others help me along the way. Thanks to Bill Riley and Jamie Fine for their excellent informal mentoring during my first couple years at Berkeley. Thanks also to Brett Singer and Michael Sohn. Bill Riley completed the EMFAC runs whose results are used in Chapter 3. Within the Energy and Resources Group, I would like to thank the staff for doing an excellent job keeping ERG up and running, and the faculty and the participants of the PhD seminar for their constructive feedback on my research. Many of the ideas contained in this dissertation were spurred on by interdisciplinary suggestions at the ERG PhD seminar. Thanks especially to my ERG Masters/PhD cohort – Rebecca, Leah, and Garvin – for helpful feedback on my research and for making ERG a fun community to be a part of. Thanks especially to Garvin for many fruitful and enjoyable discussions, on research and otherwise. I appreciate having been able to share so many aspects of my graduate experience with Garvin, including research, sports (squash and biking), classes, organic California produce, advising, music, and even housing. I could not have asked for a better officemate, classmate, neighbor, "lab spouse," and partner-in-crime. Thanks to Bev for making 605 Davis Hall a fun place to do (or not do) research,

and to Dev, Julie, Seema, Shannon, Sharon, for making the Airheads wing of Davis Hall a great place to take a break from research.

As I finish my 21st year of being a student, thank you to my family for their many years of encouragement. My father told me long ago that he thought I would enjoy getting a PhD. My response, as a scientist, was to gather empirical evidence to test this hypothesis. Having conducted the experiment (i.e., finishing grad school), I conclude that he was right: being a student is tremendous fun. Do I have to leave? Thanks to Leni for being the best older sister that a kid brother could want. To my wife, Jess, thank you for your love, patience, and support, and for making it all worthwhile in the first place.

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Chapter 1: Introduction

This dissertation explores human inhalation intake of motor vehicle emissions in urban environments. This chapter highlights the motivation for studying these topics, provides background information that frames this investigation, and outlines the flow of ideas contained in the remaining chapters.

The scope (and the title) of this work is comprised of three themes: *urban environments*, *vehicle emissions*, and *inhalation*. I begin by considering each of these themes.

Urban environments

Human societies are rapidly becoming more urbanized (United Nations, 2004). Currently, over 90% of global population growth occurs in urban areas. The global population is on a cusp, with approximately an equal number of urban- and rural-dwellers. By 2008, this balance is expected to tip, after which urban populations will exceed rural populations.

The US urban population has outnumbered the rural population since 1919 (US Census, 2003). Today there are about 4 urban residents per rural resident. About half or more of the US population lives in metropolitan areas with more than one million people each.

In general, delineating the precise boundaries of an urban area is somewhat arbitrary. Political entities, such as cities, towns, and counties, have clear boundaries, but often researchers want to classify areas as urban or rural based on geographic attributes such as population density rather than political boundaries. The US Census offers a definition based on population density: boundaries of urban regions "encompass densely settled territory, which consists of (1) core census block groups or blocks that have a population density of at least 1,000 people per square mile and (2) surrounding census blocks that have an overall density of at least 500 people per square mile" (US Census, 2004a).

Understanding the increasing degree of urbanization and its consequences is a topic of significant research by urban sociologists, demographers, and economists. Urban populations are growing because of natural population growth (i.e., birth rates exceed death rates) and because of rural-to-urban migration. Two hypotheses explaining the latter trend are (1) employment opportunities may be greater – or perceived to be greater – in urban areas than in rural areas; and (2) after evaluating the amenities (e.g., social interactions, educational opportunities), disamenities (e.g., crime, crowding), and standard of living in urban versus rural areas, individuals may prefer living in urban areas (Renkow and Hoover, 2000; Sato and Yamamoto, 2005). The relative importance of these two factors will, in general, vary over time and among societies and individuals (Colwell et al., 2002; Spilimbergo and Ubeda, 2004; Tacoli, 1998). Factors attracting employers to urban areas include agglomeration economies and access to labor, markets, and transportation infrastructure (Cohen and Paul, 2005; Mori and Nishikimi, 2002).

Urban environments are important to study for several reasons. Because a large fraction of people live and will live in urban areas, urban environments represent the conditions facing much of humanity as a whole, both now and even more so in the future. Many environmental insults occur in urban areas, because this is where much of human activity and anthropogenic emissions occur. Urban environments are important to air quality and health research because a significant fraction of air pollution that can affect health is emitted and inhaled in urban areas.

Motor vehicles and vehicle emissions

Mobility – the ability to travel when, where and how one wishes – is an integral aspect of modern society, and a cornerstone of the liberties afforded all citizens in free societies. Societies existed for millennia without motor vehicles, yet it is difficult to image modern society without vehicles, or, more broadly, without freedom of mobility. The freedom offered by vehicles captures the imagination, as exemplified by the following urbanist vision from *The Radiant City*:

> "The cities will be part of the country; I shall live 30 miles from my office in one direction, under a pine tree; my secretary will live 30 miles away from it too, in the other direction, under another pine tree. We shall both have our own car. We shall use up tires, wear out road surfaces and gears, consume oil and gasoline. All of which will necessitate a great deal of work... enough for all" (Le Corbusier, 1967).

The myriad of benefits offered by motor vehicles are difficult to fully quantify. How does one evaluate the benefits of travel: of visiting friends and relatives; of pursuing one's livelihood; of obtaining food and medicine from around the globe; and, of partaking in

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social activities such as shopping, entertainment, and dining? Motor vehicles have transformed our society in a way unmatched by most technologies.

The benefits and freedoms enjoyed by car owners, combined with rising affluence, has caused per capita car ownership and use to skyrocket (Abu-Eisheh and Mannering, 2002; Dargay and Gately, 1999; Riley, 2002). In the US, the number of licensed motor vehicles (230 million) outnumbers not only the number of licensed drivers (191 million), but also the number of people of driving age (16 and older; 224 million) (US DOT, 2003a). As one measure of the importance of mobility, consider the annual energy consumption for transportation. In the US, transportation consumes ~100 MJ person^{-1} d^{-1} (http://www.eia.doe.gov). (Among the four energy consumption sectors – residential commercial, industrial, and transportation – in the US, transportation is second in size after industry, accounting for ~27% of energy consumption.) This value of 100 MJ person^{-1} d^{-1} is about ten times the typical energy consumed as food per person per day. Walking consumes ~0.21 MJ km^{-1} (Ackermann et al., 1998), so if a person were to expend 100 MJ walking, she would travel ~480 km. (Traveling 480 km in 24 hours would mean averaging 4.8 minutes per mile, a pace that is maintainable in running by world-class athletes for at best a few hours. For example, the world record for a marathon is a few minutes over 2 hours, corresponding to an average pace of 4.7 minutes/mile.) Considering travel by bicycle yields an analogous result: At ~0.13 MJ km^{-1} by bicycle (Ackermann et al., 1998), expending 100 MJ d^{-1} cycling would involve traveling ~780 km d^{-1}, a rate achievable only by a few world-class athletes (http://www.ultracycling.com). Thus, motor vehicles allow us to expend an amount of

energy not only that we do not wish to expend ourselves, but also that most people are not physically capable of expending. Clearly, the benefits of vehicles are large.

Yet, there are costs to our transportation system as well. Safety is a major concern, with traffic collisions causing death and serious injury. Ease of travel allows people to chose to live in the suburbs, but the resulting urban form may foster feelings of isolation rather than of community (Putnam, 2001). Freeways are noisy and a nuisance to the communities they disrupt. Cities designed to be efficient for the movement of cars are not necessarily pleasant places to live and work. The prospect of a city of several million people achieving Le Corbusier's integrated urban/rural vision seem implausible in the foreseeable future. A practical limitation is motor vehicle congestion, which is an issue for urban areas around the world. A study of 75 urban areas in the US estimated the total annual cost of congestion at $70 billion, or $520 per person (Texas Transportation Institute, 2003).

Transportation systems deleteriously impact human health and the environment in several ways. Wetlands are paved over to build new roads. Rainwater runoff, washing over roads and parking lots, carries to local water bodies vehicle pollutants such as fuel, lubricating oil, and tire and brake dust. Petroleum exploration and drilling disturbs wilderness environments. Road construction and vehicle manufacture consumes significant amounts of materials and energy.

One important impact of transportation systems is the health impact attributable to human inhalation of vehicle emissions. In the US, about 126 million people, or 44% of the total population, live in areas where atmospheric concentrations of one or more criteria pollutants exceed the National Ambient Air Quality Standards (US EPA, 2003b).

Atmospheric concentrations of criteria pollutants in California's South Coast Air Basin (SoCAB), which includes Los Angeles, exceed these air quality standards approximately one out of three days (CARB, 2005).

Vehicle emissions are a significant contributor to urban air pollution throughout the world (Faiz, 1993). Vehicle emissions include criteria pollutants, such as carbon monoxide and particulate matter; species that react to form criteria pollutants, such as nitrogen oxides (NO_x) and volatile organic compounds (VOCs), both of which can lead to ozone and particulate matter formation; and several hazardous air pollutants, such as benzene and butadiene. The portion of total year-2002 emissions in the US National Emission Inventory that are attributable to transportation sources is 77% for CO, 54% for NO_x, and 44% for VOCs (US EPA, 2003c).

Exposure to traffic emissions has been shown to cause a variety of acute and chronic health impacts (Brook et al., 2004). For example, maternal exposure to vehicle emissions is associated with adverse birth outcomes such as low birth weight and premature birth (Wilhelm and Ritz, 2003). Chronic exposure to traffic exhaust is associated with leukemia and other childhood cancers (Feychting et al., 1998; Knox and Gilman, 1992; Pearson et al., 2000; Savitz and Feingold, 1989). There is an association between exposure to traffic and onset of a myocardial infarction within the subsequent one to three hour period (Peters et al., 2004). Vehicles contribute significantly to population exposure to particulate matter (PM), which has been associated with lung cancer and premature mortality (Dockery et al., 1993; Laden et al., 2000; Pope, 2000; Pope et al., 2002; Pope et al., 1995). A study of ambient concentrations and attributable risk in California's South Coast Air Basin found that mobile sources contribute almost

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70% of the estimated lifetime ambient air pollution cancer risk (Morello-Frosch et al., 2001). Quantifying the emission-to-inhalation relationship is important for better understanding air pollution health effects and for designing effective mitigation strategies.

The ways in which current and future transportation systems satisfy transportation demand will have significant implications for air quality. Because transportation emissions are a significant contributor to urban air pollution, the technologies and infrastructure that make up the current and future transportation system will likely impact overall air quality in most urban areas. Conversely, the air quality impacts of motor vehicles are one of several factors that influence transportation planning decisions. Because transportation is one of the main aspects of urban air quality management, efforts to improve air quality are one of a few key motivators for transportation planning.

Historically, significant reductions in vehicle emissions are attributable to technical advances, including catalytic converters, on-board diagnostics, inspection and maintenance programs, and fuel reformulation. In designing strategies to meet air quality objectives, one would reasonably expect that technology can and should play an important role. At the same time, technical fixes are not always available, nor are they necessarily the easiest, most reliable, or least expensive solution. Ideas for ways to improve the transportation system may come from a variety of other fields as well, including economics, transportation planning, political science, business, and sociology.

Inhalation

In contrast to many other types of environmental contamination such as groundwater pollution, there is no "remediation" of urban air pollution. Once pollution is emitted to urban air, there is no practical way to actively remove a significant fraction of that pollution. Instead, one can only wait for removal by natural processes such as advection and wet and dry deposition. Improving urban air quality occurs by reducing emissions. Because the typical residence time of air in an urban area is less than one day (see Chapter 4 and Appendix I of Chapter 3), emission reductions lead to improvements in urban air quality literally overnight. In general, it is not considered practical to eliminate all emissions. An important air quality policy goal, then, is effective *prioritization* of emission reductions. There are many sources of emissions in a typical urban area, and policy makers are tasked with choosing which sources to control and by how much. Because the most important reason for regulating urban air pollution is to protect public health, health impact is should be a very important factor for prioritizing emission reductions. Much of the research in this dissertation investigates analytic tools, data analysis techniques, and models to help prioritize emission reductions in terms of their potential for health benefits.

Inhalation intake of air pollution is a better proxy than mass emissions or ambient concentrations for the health impacts of a specific emission source. Yet, air pollution regulations in the U.S. focus on limiting emissions and controlling ambient concentrations. For example, the Clean Air Act regulates criteria pollutants based on ambient concentrations, as given by National Ambient Air Quality Standards (NAAQS), and hazardous air pollutants based on emission control technologies. There is the

potential for air pollution policy to be more effective at improving public health by focusing some attention on limiting intake or health impacts rather than simply restricting emissions or ambient concentrations. (In understanding the policy implications of the results I present in this dissertation, one should recognize that environmental policy is not always 100% rational or even coherent. Decisions are based not only on scientific understanding, but also on a number of factors including costs, public opinion, and politics. My dissertation can help inform policy decisions, even recognizing that such decisions result from a variety of inputs.)

The health effects attributable to an air pollution source or source category, such as a power plant or motor vehicles, may be estimated as the product of three terms: emissions (mass emitted per time), intake fraction (mass inhaled per mass emitted), and toxicity (health impact per mass inhaled). In conducting a health risk assessment for motor vehicle emissions, using this paradigm, vehicle emissions can be determined by laboratory testing and by on-road remote sensing. Intake fraction can be estimated using air dispersion models, biomarkers, tracer gas experiments, and "tracers of opportunity" (i.e., exploiting measurements of pollutants that are primarily emitted from motor vehicles). Intake fractions depend on the size of the exposed population, the proximity of that population to the emission sources, and the persistence of pollutants in the environment. Toxicity and other dose-response information can be determined in a laboratory by exposing animals, cells, or humans to pollutants, or based on epidemiological studies of "real world" human exposures. This dissertation explores intake fraction. It also explores attributable intake, which can be determined as the product of emissions and intake fraction.

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Dissertation outline

 The remaining chapters in this dissertation are organized as follows. Chapters 2-5 focus on intake fraction, while Chapters 6 and 7 explore intake. Chapter 2 provides an overview of the intake fraction metric, including how it is calculated and how it could be used in formulating air pollution policy. Chapters 3, 4, and 5 estimate intake fraction values for specific situations: Chapter 3 focuses on vehicle emissions in California's South Coast Air Basin; Chapter 4 covers vehicle emissions in urban areas throughout the U.S.; and, Chapter 5 investigates the issue of school bus self-pollution, which refers to exhaust from a school bus migrating to inside that bus.

 People are mobile. Chapter 6 describes the methods and results for a Geographic Information System (GIS) inhalation assessment for the South Coast that accounts explicitly for the movement of people within an urban area during each day. Chapter 7 investigates how urban population and land area influence population intake of vehicle emissions. At issue is whether changes to urban population density can reduce transportation emissions and/or inhalation intake of these emissions. The investigation in this chapter uses an idealization of a hypothetical urban area. Chapter 8 provides a summary, suggestions for future research, and conclusions.

 In terms of the spatial scale of analysis, Chapter 3 begins at a logical starting place for investigating urban air pollution: a single urban area. Chapter 4 considers a larger scale, investigating many urban areas. Chapter 5 considers a narrower scale, focusing on a specific vehicle type (school buses). Chapters 6 and 7 return to the scale of a single urban area to explore various aspects of urban-scale inhalation modeling

(Chapter 6) and to investigate broadly the role of urban-scale planning goals (e.g., sprawl versus infill) on inhalation of vehicle emissions (Chapter 7).

The main disciplines underlying this dissertation are air quality engineering, environmental health, and urban planning. These disciplines arose to solve complex sets of problems facing society, mainly in urban environments. Their objectives, and the broad objectives of this dissertation, include understanding how systems work and also how to propose and evaluate potential solutions to urban environmental concerns. While most of the research in this dissertation quantifies and documents current conditions, there is a secondary emphasis on predicting how things may change in the future and on suggesting opportunities for effective policy interventions.

Chapter 2: Overview of intake fraction and its use in air-pollution policy

Introduction

The effectiveness of air quality regulations is sometimes evaluated in terms of changes in emissions rates. Indirectly, the effects of such reductions may be observed through changes in air concentrations as measured at ambient monitoring stations. Regulators commonly assume that decreases in ambient air concentrations cause commensurate decreases in human exposure. However, this is not necessarily the case, because personal exposures can vary substantially from what ambient air monitors indicate. For example, concentrations of pollutants from motor vehicles are higher in vehicles than in ambient air. As another example, exposure concentrations from second-hand cigarette smoke (also called environmental tobacco smoke, ETS) are significantly higher if the cigarette is smoked indoors rather than outdoors, even though the indoor and outdoor releases would yield comparable attributable concentrations at an ambient monitoring site. The intake fraction is orders of magnitude greater for indoor releases than for outdoor releases of primary pollutants (Lai et al., 2000).

This chapter presents ideas about how to prioritize emission reductions based on their effectiveness in reducing exposures. One might consider, for example, the location of the emissions source, the surrounding population densities, and the factors affecting dilution of the emissions. This approach contrasts with a more typical approach in

common regulatory practice, which would evaluate emissions based on mass emitted or based on the contribution to concentration measured at an ambient monitor.

Because intake fraction is a relatively new idea, the use and implications of intake fraction values have not been fully explored in the literature. This chapter aims to help fill this gap. This chapter describes in broad brush many aspects of intake fraction, including typical values and how the use of intake fraction in policy analyses may depend on what information is available. Throughout this chapter, specific examples are used to illustrate ideas, but these examples are not intended to be exhaustive.

Background

As highlighted in Chapter 1, an important air quality policy goal is effective *prioritization* of emission reductions. There are many sources of urban air pollution in each urban area, and policy makers are tasked with choosing which sources to control and by how much. Because the most important reason for regulating urban air pollution is to protect public health, environmental health impact should be an important factor when prioritizing emission reductions.

One way to estimate the environmental health impact of a pollution source or source class is as the product of three terms: emission rate (mass per time), intake fraction (mass inhaled per mass emitted), and toxicity (health impact per mass inhaled). In the ideal situation, one would know all three terms for all major emission sources. However, as I describe below, one can make effective prioritization decisions without complete information. This chapter focuses on understanding and using the second term in this

relationship (intake fraction). In many cases, there is a complex or non-linear relationship between health and these three factors. Such nonlinearity is discussed below.

There are several advantages to presenting exposure analyses in terms of intake fraction. Intake fraction is a tangible concept that can increase understanding and improve intuition about exposure assessments. As an exposure assessment metric, intake fraction can be used as a diagnostic tool to help corroborate empirical and modeled exposure assessments, and it can be used to summarize the importance of various transport and exposure pathways for each chemical being studied. Intake fraction is an excellent metric for technical and nontechnical researchers to understand exposure assessment results. Policy makers want to make progress towards efficiently and effectively reducing human exposure to hazardous air pollutants, and the intake fraction is a valuable metric for understanding how reductions in emissions relate to reductions in intake.

The term "intake fraction" was first introduced in the literature in 2002 (Bennett et al., 2002). However, there is a much longer history of the idea of quantitatively relating pollutant emissions to inhalation intake, as reviewed by Evans et al. (2002). Smith and colleagues have explored policy implications of intake fraction and related concepts (e.g., Smith, 1995; Smith, 2002; Smith and Edgerton, 1989). Although not yet large, the literature on intake fraction is diverse, addressing primary and secondary pollutants (Levy et al., 2003), inhalation and other intake pathways (MacLeod et al., 2004), and varied sources such as motor vehicles (Evans et al., 2002), power plants (Evans et al., 2002; Levy et al., 2003), and dry cleaners (Evans et al., 2000).

Intake fraction should be understood as a metric, not as a method. Like emissions and concentrations, it can be determined using several methods. Broadly, there are two approaches for quantifying the emission-to-intake relationship: models and measurements. During the past several decades, much work in air-quality engineering has developed and used these approaches to understand emission-to-airborne concentration relationships. The methods developed and the results obtained can also be used to inform the emission-to-intake relationship.

What is intake fraction?

Intake fraction summarizes in a compact and transparent form the quantitative relationship between emissions and inhalation of those emissions. Intake fraction is useful in connecting emissions to effects because mass inhaled is a better indicator of potential adverse health impacts than either mass emitted or airborne concentration.

More generally, the emission-to-effects relationship involves a series of causally related steps. As illustrated in Figure 2-1 (adapted from Smith, 1993), emissions are diluted, transported, and/or transformed to generate the pollutant concentrations that people breathe. Human contacts with concentrations constitute exposures, and inhalation of pollutants results in intake. Pollutant transfer into the body of an exposed individual leads to doses to organs and other physiological targets, which in turn can elevate the risk of adverse health effects. Intake fraction quantitatively summarizes an important portion of this chain of events by describing as a single number the emission-to-intake relationship.

Intake fraction can be determined through several different methods. Investigations that generate intake fraction results can range from straightforward to complicated, and can depend on modeling, experimental measurement, or both. Simple intake fraction calculations, when performed similarly across different sources, can in many cases produce reasonably accurate estimates of relative levels of inhalation exposure.

Intake fraction for a primary pollutant is the total mass inhaled from an emission source, divided by the total mass emitted from that source:

$$\text{Intake fraction} = \frac{\text{pollutant mass inhaled}}{\text{pollutant mass emitted}} \qquad (2\text{-}1)$$

The emission source evaluated in the denominator can be a single emitter, such as an industrial stack, or a broad source class, such as motor vehicles. When considering an entire population, the value of the numerator would be the cumulative pollutant mass inhaled by all exposed individuals. When considering a subpopulation or an individual, the value in the numerator would be the pollutant mass inhaled by that subpopulation or individual. Intake fraction depends primarily on three types of parameters: those that influence dilution, such as meteorology; those that reflect the proximity of people to the emissions, such as population density; and those that reflect persistence of a pollutant in the atmosphere, such as pollutant removal rates. Therefore, intake fraction tends to vary with location and may also vary with time. For example, if two emission sources emit the same mass of a pollutant, but one source is in a densely populated urban area while the

other is in a rural area, the first source will have a higher associated intake fraction because there are more people in the vicinity of the emissions. On the other hand, intake fraction for ground-based ambient pollutant releases is smaller during periods of rapid mixing and dispersion, such as sunny and windy days, than during stagnant atmospheric conditions.

One important attribute of intake fraction is that it can be applied to multiple pollutants from a source, rather than only to specific pollutants. That is, if two pollutants are emitted from the same source and have the same transport and transformation characteristics, then their intake fraction values will be the same, even if their chemical composition and mass emission rates are very different. A second important attribute of intake fraction is that it can be applied to multiple sources. That is, pollutants from different sources or source classes can have similar intake fraction values if governing factors (e.g., size of the exposed population; proximity between emissions and exposed population; environmental fate and transport characteristics) are similar. As more studies of intake fraction are completed, a compendium of intake fraction results can be developed that would provide useful guidance on expected values for similarly situated sources not yet assessed.

Air pollution includes both primary and secondary contributions. Primary pollution refers to species that are in the same chemical form when inhaled as when emitted. Secondary pollutants are formed in the atmosphere from gaseous precursors. The meaning of an intake fraction value, for example as defined in Equation 2-1, is unambiguous for primary pollutants. In contrast, since the chemical form of a secondary pollutant is different as inhaled than as emitted, the definition of intake fraction for

secondary pollutants requires a more detailed specification. If one were interested, for example, in exposure to secondary particulate matter originating from nitrogen oxide emissions from combustion sources, then the intake fraction might usefully be defined as the attributable mass of particulate oxidized nitrogen inhaled per unit mass of nitrogen in NO_x emitted. Tracking a specific chemical element from source to receptor when specifying the intake fraction of a secondary pollutant can assist in preserving across different pollutant forms important characteristics of the source-receptor relationship that are central to the intake fraction metric. This approach of tracking a specific element when estimating intake fraction is possible for some pollutants, such as particulate nitrate (where nitrogen emissions could be tracked), but not for other pollutants, such as ozone. Ozone is formed in the atmosphere from a complex series of reactions involving nitrogen oxides (NO_x) and reactive organic gases (ROG). The chemical structure of ozone is three oxygen atoms (O_3), but these atoms cannot meaningfully be traced back to a specific emission source: oxygen (O_2), which comprises 21% of the atmosphere, is available to form ozone whenever reactions with NO_x and ROGs occur. Further development of the idea of incremental reactivity (i.e., the change in ozone concentration attributable to a change in precursor emissions) (Martien et al., 2003) may yield a meaningful basis for quantifying ozone intake fractions.

Typical intake fraction values

Population intake fraction values for air pollutants vary over several orders of magnitude, depending on characteristics of the release environment and the pollutant. Typical values for some important release categories are known, as presented in Figure 2-

2. For outdoor releases in urban areas, intake fraction values are typically in the range 1 –
100 per million. An intake fraction of one per million means that for every million grams
emitted, one gram is collectively inhaled by the exposed population. This intake fraction
value also means that to reduce inhalation intake by one gram through emission control
would require reducing emissions by one million grams.

Emissions that are not inhaled eventually degrade or otherwise leave the
atmosphere. A common degradation mechanism for anthropogenic air toxics is attack by
a hydroxyl radical (OH). The rate of decay depends on the concentration of hydroxyl and
other reactive compounds and on the reactivity of the emitted species. A few pollutants
do not degrade in the troposphere. These pollutants may mix throughout the troposphere
via advection, and then migrate via molecular diffusion and weak air currents to the
upper stratosphere, where degradation occurs via ultraviolet radiation. Air pollutants can
also be removed from the atmosphere via wet and dry deposition to land and water. There
can be rapid degradation on vegetation surfaces, in soil, and in surface water for
deposited pollutants. Some emissions enter ecosystem food chains, which is especially a
concern for bio-accumulative compounds.

For a single species, intake fraction values may be different for secondary and
primary components. For example, because almost all secondary PM is in the fine mode,
the intake fraction could be large because the particles are highly persistent. At the same
time, however, secondary PM does not exhibit the same proximity effect that primary PM
does, because secondary PM takes time to form from precursor emissions. In addition,
the conversion from precursor emissions to secondary PM is incomplete, further reducing
the intake fraction. For example, consider secondary ammonium nitrate particles. Nitric

oxide (NO) emissions are oxidized in the atmosphere to nitrogen dioxide (NO_2), and then to nitric acid (HNO_3). If the nitric acid encounters ammonia (NH_3), the two species can react to form particle-phase ammonium nitrate (NH_4NO_3). In general, only some portion of NO and NH_3 emissions ultimately form PM, which reduces the intake fraction of secondary PM associated with NO_x emissions. The effect of the factors above on intake fraction will vary among areas and over time. Evans et al. (2002), studying conditions throughout the US, found that intake fraction values for power plants and urban and rural vehicle emissions were between one and two orders of magnitude less for secondary PM than for primary PM.

Determining intake fraction

This dissertation analyzes results from both models and measurements. Models to calculate intake fraction range from simple, one-compartment representations of an urban area, to complex, three-dimensional urban airshed models or multi-compartment regional multimedia fate models. Measurements include tracer gas experiments as well as utilization of "tracers-of-opportunity" (i.e., chemical compounds that act as a "fingerprint" for an emission source).

The following factors are likely to be important when estimating intake fraction for urban emissions:

- population density and the size and location of the exposed population relative to the source;
- meteorological conditions controlling atmospheric transport and dispersion, such as wind speed and mixing height, and pollutant release height;

- pollutant persistence, which depends on the rate of removal mechanisms such as deposition; and,

- the presence of simultaneous indoor releases (e.g., self-pollution), if any.

The use of intake fraction in policy decisions

This section highlights several ways that the intake fraction could be used to assist in making air pollution policy decisions. The way that one might use intake fraction in such decisions depends in part on the availability of information. I describe below how intake fraction can provide useful information in a policy decision, depending on what information is available. I then provide specific examples of the use of intake fraction to prioritize emission reductions.

How intake fraction can be used depends on available information and the policy question

The health impact attributable to an emission source can be expressed as the product of emissions, intake fraction, and toxicity. However, there are many situations in which one can use intake fraction to assist in setting priorities for emission control without full information about emissions and toxicity. For example, each of the next four paragraphs presents a situation wherein different pieces of information are available. In each case, I assume that there are two emission sources, and that the policy question at hand is to prioritize between these two sources as the target of emission reductions. I

further assume here that the pollutant of concern exhibits a linear, no-threshold dose-response relationship.

1. When all three terms – emissions, intake fraction, and toxicity – are known, one can estimate the overall health impact from the two sources. The emission source with the higher health impact would be identified as a higher priority for control. If information about the costs of control technologies is also known, then one could prioritize emission reductions based on a cost-effectiveness analysis. In this case, one could seek to maximize the reduction in adverse health effects per unit cost (Smith, 1995; Smith and Edgerton, 1989).

2. When only emissions and intake fractions are known, one could prioritize emission source reductions based on total emissions, but using the intake fraction values as multipliers. This approach is useful when comparing two sources of a specific species. For example, if the intake fraction is two times greater for emission source *A* than for emission source *B*, an emission reduction of 1 kg from *A* could be given the same policy "priority" as an emission reduction of 2 kg from *B*. In application to particulate matter, this approach implicitly assumes that — in the absence of information to the contrary — all particulate matter should be treated as equally toxic, regardless of its source. Under this assumption, inhalation intake becomes a suitable proxy for adverse effect.

3. When only intake fractions and control costs are known, one can carry out certain cost-effectiveness analyses. For example, if control costs per kg *emitted* are the same for emission sources *A* and *B*, but the intake fraction is larger for *A*, then the control cost per kg *inhaled* is less for *A* than for *B*. All else being equal,

controlling emissions from *A* would be a higher priority than controlling

emissions from *B*.

4. Finally, when only intake fractions are known, one can compare sources that are

    similar in other ways. For example, comparing natural-gas power plants in

    different locations, one could prioritize for control the emissions location with the

    higher intake fraction.

Using intake fraction to prioritize among emission sources

Several examples are presented below of how intake fraction might be used to

prioritize among emission sources, based on attributes of the emissions or the

environment. These comparisons are summarized in Table 2-1.

Urban versus rural emissions. Intake fraction depends in part on the proximity

between an emission source and the population. All else being equal, urban emissions

tend to have a higher intake fraction than rural emissions because of the closer proximity

of urban sources to large numbers of people. The intake fraction difference between

urban and rural areas can be greater than an order of magnitude, indicating that the

location of emissions can significantly influence the public health impact of those

emissions. For example, based on one-compartment model results in Chapter 4, the

intake fraction is estimated to be ~5 times larger in a moderate-sized city such as

Sacramento, California, than in a small city such as Chico, California, owing to

differences in the population and size of the urban area. The difference in intake fraction

for emissions in Sacramento as compared to a typical rural area would be even greater than a factor of 5.

When comparing two emission sources, a full analysis of intake fraction and health impacts would require air dispersion modeling and geographic information about where people live. The one-compartment model approach, discussed in more detail in Chapter 4, suggests that an estimate of the difference in intake fraction between two situations can sometimes be made based on the difference in linear population density, which is the urban population divided by the square root of the urban land area. Thus linear population density could be used in a first-level analysis to "weight" emissions. To illustrate, because of differences in linear population density (given in Table 4-1), reducing emissions by a certain mass amount per year is expected to yield about 5 times the cumulative public health benefit if it occurs in Sacramento than if it occurs in Chico.

Time-of-day and time-of-year. Meteorology varies diurnally and seasonally. Low wind speeds and low mixing heights tend to increase the intake fraction for ground-level emissions, as compared with high wind speeds and high mixing heights. For example, using Gaussian plume dispersion modeling, Lai et al. (2000) found that atmospheric stability class can affect short-term intake fraction by as much as an order of magnitude. Wind speed and direction may influence intake fraction by a similar amount.

Meteorological patterns also affect long-term average intake fraction values. For example, results presented in Chapter 3 indicate that the intake fraction for motor vehicles in the South Coast Air Basin is about 2 times higher in winter than in summer. The main reason for this trend, explored in Chapter 4, is that mixing heights tend to be

higher in summer than in winter. This same seasonal trend has been observed to be common in cities throughout the United States. Diurnal meteorological patterns, with lower mixing heights during the night than during the day, suggest that intake fractions are higher for emissions that occur at night and in the early morning rather than during the middle of the day.

Household wood stove smoke is an example of an emission source for which time patterns in emissions are likely to have an important influence on intake fraction. Because stoves operate more at night than during the day, and in winter rather than during summer, their intake fraction is higher than would be the case if they did not exhibit diurnal or seasonal emission patterns.

Stack height. Just as the geographic location in the horizontal plane of an emission source influences those emission's proximity to people and, thus, the intake fraction, so too does stack height. Tall stacks loft pollution high into the air, often reducing the intake fraction because of the significant dilution that occurs by the time the plume reaches the ground. (Because pollutants released from a tall stack take time to reach the ground, high attributable concentrations may occur significantly downwind of the release location. Population density may be significantly higher or lower downwind of the release than near the release.) Evans et al. (2002) calculated intake fractions for emissions from 40 power plants in the US, and found that stack height can make an order of magnitude difference in intake fraction. Lai et al. (2000), using a Gaussian plume model to analyze hypothetical stack emissions, reported similar results.

On-road sources. People in urban areas typically spend some time in or near vehicles each day. For example, focusing on urban diesel PM emissions, one can use published data to estimate the typical intake fraction differences between on-road sources and other sources. Supporting calculations for this paragraph are in the appendix for this chapter. Recent measurements for diesel suggest a factor of 4 – 14 difference between in-vehicle and nearby ambient concentrations (Fruin et al., 2004). The result is that the ~6% of time (80 minutes per day) people spend on average in vehicles (Klepeis et al., 2001) would contribute ~25 – 54% of total exposure to diesel PM, rather than 6% (Appendix, line 5d). This range of values matches the range of 28 – 55% obtained from a more detailed analysis by Fruin et al. (2004). If it is assumed that 25% of diesel PM emissions are from on-road sources (CARB, 2000c), then on-road sources contribute ~39 – 63% (rather than 25%) to total diesel PM exposure (Appendix, line 6c). On average, on-road sources are estimated to contribute between 1.9 and 5.1 times more diesel PM inhalation per unit emissions than other sources (Appendix, line 8). Thus, from an exposure standpoint, on-road diesel particle emissions should be given a "weighting" of ~1.9 – 5.1 relative to off-road diesel sources. The width of this range reflects uncertainty in diesel PM concentrations in vehicles relative to those in the ambient environment. For comparison, the appendix presents similar calculations for benzene, a pollutant for which in-vehicle and ambient concentrations are more certain, and for which the corresponding intake "weighting" for on-road sources is estimated to be ~1.3. This weighting value is lower for benzene (~ 1.3) than for diesel PM (~ 1.9 – 5.1) in part because outdoor benzene, unlike outdoor diesel PM, is assumed to penetrate the building envelope without loss.

Self-pollution. Combustion sources typically possess an exhaust system to deliver emissions to ambient air. The exhaust manifold of a car conducts effluents from the engine to the tailpipe; wood stoves emit their waste gases through a chimney that runs from the fireplace to the rooftop. Generally, exhaust systems work well but not perfectly, and a small fraction of emissions may enter the indoor or in-vehicle environment. Such leaks lead to a condition known as "self-pollution."

Intake fractions for pollutants that enter occupied buildings or vehicles are much higher than intake fractions for outdoor air emissions. For example, consider a one-compartment model applied to a single household. Assuming three people occupy the household, a building volume of 350 m^3, and an air exchange rate of between 0.2 h^{-1} and 2 h^{-1}, the intake fraction for an indoor release of a conserved pollutant is between 0.3% and 3% (i.e., between 3,000 per million and 30,000 per million) (Lai et al., 2000). These values are 50 – 500 times greater than the intake fraction estimated in Chapter 4 for the Los Angeles Metropolitan Area using the one-compartment model (60 per million) and 800 – 8000 times greater than the similar estimate for Chico, California (4 per million).

An approximate estimate of the intake fraction in vehicles can be derived using the same approach used by Lai et al. (2000) for a household. Typical values for a vehicle are 1.6 individuals per vehicle (US DOT, 2003b), an air exchange rate of ~ 36 h^{-1} (Hayes, 1991; Johnson, 2002), and an interior volume of 2.5 – 3.5 m^3 (Johnson, 2002). (Air exchange rates in vehicles vary by more than an order of magnitude, depending on factors such as the vehicle speed, wind speed, window position (open or closed), and vent position (open or closed). The air exchange rate of 36 h^{-1} represents the best estimate for

typical conditions in vehicles (Johnson, 2002).) These values yield an intake fraction of 1.0% – 1.4%, which lies within the range estimated for the indoor intake fraction, 0.3% – 3%.

Because intake fractions are so much larger for indoor and in-vehicle releases than for outdoor releases, even a small amount of self-pollution can significantly increase the intake fraction associated with sources like motor vehicle exhaust and residential wood combustion. As detailed in Chapter 5, school buses illustrate the high impacts possible from self-pollution. Tracer-gas experiments conducted on six buses indicate that the self-pollution intake fraction is significant for school buses, to a degree that depends on the age of the bus. The self-pollution intake fraction reported in Chapter 5 is 70 per million for the oldest bus investigated (model year: 1975), and averages 20 per million for the remaining five buses (model years between 1985 and 2002). These values indicate that for a school bus operating in the South Coast Air Basin, the cumulative mass of pollution inhaled by the roughly 40 students on a bus is similar to the cumulative mass of pollution inhaled by all of the other 15 million exposed individuals in the air basin. Making the reasonable assumption that the self-pollution intake fraction does not depend on the size of an urban area, for buses operating in a small city such as Chico, the majority of the inhalation intake of emissions from a school bus might well be experienced by the individuals riding that bus. Self-pollution offers the potential to be a "low-hanging fruit" for realizing exposure reduction benefits. Addressing bus self-pollution could markedly reduce the inhalation intake of diesel PM, even before bus emission reductions are achieved.

Coarse particles versus accumulation mode particles. Accumulation mode particles are more persistent than coarse mode particles (Seinfeld and Pandis, 1998). Coarse mode particles deposit onto stationary surfaces as a result of gravitational settling and inertial impaction, which reduces their characteristic residence time in air. As a result, all else being equal, the intake fraction is greater for accumulation mode particles than for coarse particles. The fraction of PM that is in the coarse mode is likely to be higher for physically generated PM (e.g., wind-blown soil and dust from car tires and brakes) than for combustion-generated PM (e.g., vehicle emissions, power plant emissions).

A rough estimate, using a one-compartment model of an urban area and assuming settling velocities (units: cm s^{-1}) of 0.03 and 3 for fine (PM$_{2.5}$) and coarse (PM$_{10-2.5}$) particles, respectively (Seinfeld and Pandis, 1998), indicates that the urban intake fraction is ~80% less for coarse PM than for fine PM. This value is derived from Equation 3A-3 in Chapter 3 Appendix I, and uses (1) the deposition velocity divided by the mixing height as an estimate for the first-order rate constant, k, and (2) the value 480 m^2 s^{-1} for wind speed times mixing height (i.e., the median x-axis value in Figure 4-4). Deposition velocity can vary over a wide range, depending not only on particle size, but also on airflow conditions and land surface roughness. This calculation illustrates that deposition may be important as a removal mechanism for coarse particles, especially when considering a large air basin. (This analysis does not incorporate indoor-outdoor ratios for coarse versus fine PM, which influence intake rates. In general, when ambient pollution migrates to indoor environments, the fraction of particle mass removed is greater for coarse particles than for fine particles. Accounting for this difference would further

reduce the intake fraction for coarse particles relative to the intake fraction for fine

particles.)

      Primary versus secondary PM. Evans et al. (2002), studying conditions

throughout the US, found that intake fraction values for power plants and urban and rural

vehicle emissions were between one and two orders of magnitude less for secondary PM

than for primary PM. This large difference is a result of two factors: (1) the greater time

required for secondary PM to form and the resulting greater dilution that occurs between

emissions and exposure, and (2) a significant mass fraction of precursor emissions do not

actually form secondary PM because of chemical equilibrium and kinetic constraints and

because of loss mechanisms for gaseous precursors. The difference in intake fraction

between primary PM and secondary PM varies significantly among locations because of

differences in dilution rates, population proximity, and particle formation rates. The

findings in Evans et al. (2002) suggest that reducing PM inhalation intake by a specific

amount would require much greater mass emission reductions if policies target precursor

emissions that form secondary PM, rather than targeting primary emissions. Even so,

targeting precursor emissions is still appropriate. For US power production, mass

emissions of SO_x and NO_x, which are $PM_{2.5}$ precursors, exceed primary $PM_{2.5}$ emissions

by several orders of magnitude.

Environmental justice

      Understanding and addressing distributional issues related to air pollution

exposure is an important aspect of air quality management. Air pollution control policies

need not only to reduce the total health impact of emissions, but also to ensure that the distribution of burden among the population is not unfair or unjust. Throughout most of this document, intake fraction is based on the total population intake. However, intake fraction can be apportioned by population subgroup, even down to the level of an individual. So, for example, if the population is divided into a set of groups, the total population intake fraction can be considered as the sum of the partial intake fractions associated with each group. One indicator of environmental justice would be the degree to which partial intake fractions per capita are consistent among different demographic indicators. For example, Heath et al. (2003) considered how intake fraction for an electricity generation station depends on the station's location. They present, for specific locations in California, the percentage of the downwind population that is white versus non-white, and the percentage of total intake that occurs in the white and non-white populations. In 3 of the 5 cases studied, non-white populations are significantly more exposed than white populations. For example, for a hypothetical small-scale electricity generator ("distributed generation") located in downtown Los Angeles, 32% of the exposed (i.e., downwind) population is non-white, but this population accounts for 69% of the total intake (Heath et al., 2003). Chapter 6 of this dissertation considers environmental issues related to inhalation of urban air pollutants in the South Coast. That chapter considers intake rate rather than intake fraction.

When considering environmental justice and air pollution, the population could be divided into subpopulations using a number of attributes, including ethnicity, gender, neighborhood, income, age, and health status. As information emerges about different degrees of susceptibility of demographic subgroups to air pollution exposure, intake

fraction analyses can be conducted to highlight the levels of exposure that these subgroups encounter in relation to other parts of the population. Doing so would enable additional control effort to be targeted at protecting those who are most vulnerable to air pollution.

Intake fractions and health risk assessments

The source-by-source approach used in a conventional health risk assessment is designed to accommodate a small number of large sources for which the local impacts are large. While it is possible to complete a conventional HRA for a situation involving many individuals and many sources, this approach becomes less efficient as the number of sources and individuals increases. For a distributed source, such as motor vehicles in the SoCAB, whose pollution reaches many millions of people, it is important to consider the cumulative impact of all vehicles on the entire population. The evaluation of the health risks associated with motor vehicles represents a different context from situations typically evaluated in a health risk assessment, and evaluating the intake fraction represents a useful step in quantifying these risks.

To use population intake as part of a risk assessment, it would first be necessary to convert the units for existing toxicity factors from risk per concentration to risk per intake. For example, benzene's concentration-based unit risk for leukemia is 8.3×10^{-6} per $(\mu g/m^3)$, meaning that for each 1 $\mu g/m^3$ increase in the population average lifetime benzene exposure will lead to an additional 8.3 leukemia cases per million people so exposed (US EPA, 1993). Similarly, each 1 $\mu g/m^3$ increase in the lifetime exposure for an individual will increase the risk (probability) of contracting leukemia by 8.3 per million.

This same lifetime of exposure, at an inhalation rate of 12.2 m^3 d^{-1} (Layton, 1993), will lead to a lifetime intake of 0.31 g. Thus, the intake-based unit risk is 27×10^{-6} per gram (i.e., 8.3×10^{-6} divided by 0.31 g), meaning that each additional 1.0 gram inhaled over a lifetime will increase the risk of leukemia by 27 per million. If the dose-response curve is linear, with no threshold, then the intake-based unit risk represents the cancer risk independent of whether the intake occurs in one individual or many individuals. (This assumption is not valid across all possible exposure scenarios. It is mathematical result of the common linear no-threshold dose-response assumption, and it may reasonably hold for a range of common exposure scenarios.)

One method for characterizing environmental health risks is in terms of the risk to the "maximally exposed individual" (MEI). This approach is common in conventional health risk assessments (HRAs). The MEI for a specific source is a hypothetical person who spends all of his or her time at the location of that source's maximum impact. For example, for a power plant, the MEI might be a hypothetical person who spends 100% of his or her time close to the plant and in the downwind direction. Usually the health risk to this hypothetical MEI is significantly larger than the true risk to any real individual. If decision makers evaluate emissions sources solely in terms of the risk to a hypothetical MEI, then sources that have a large localized impact may be deemed unacceptable, whereas sources that have a moderate impact over a large number of people may be deemed acceptable. An unintended consequence of this approach is that a single large source that yields an unacceptable MEI risk can be deemed more acceptable if it is divided into several smaller sources, each with smaller MEI risks. The conclusion – that a larger number of smaller MEI risks is acceptable – may be reached *even though the total*

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risk to the exposed population could remain unchanged or even increase through the process of splitting a large source into many smaller ones.

A second method commonly used in a conventional HRA is to identify the number of people above a certain concentration or risk level. This method is more useful than the MEI approach at capturing the cumulative population burden, but it is still not complete. Two sources could have the same number of people above a certain threshold, while exposing the above-threshold and the below-threshold populations to very different concentrations. In addition, similar to the MEI approach, a single source split up into many little sources may cease to expose any individual above a certain level, even if doing so does not decrease the total population health burden.

Intake fractions are used to calculate the total population intake, which (for some pollutants) may be more closely associated with the cumulative risk to the population or with the population disease burden. This focus on cumulative population risk is often not included in conventional HRAs. For a compound exhibiting a linear, no threshold, dose-response relationship, the population's health risk is directly proportional to the total population intake. The use of intake fractions in risk assessments shifts the framework from one based on the risk to an individual or group of individuals to one based on the risk burden to an entire population or to a subpopulation.

Intake fractions represent a novel way of quantifying answers to environmental health questions. Intake fraction values can be useful for comparing pollutant impacts across diverse source categories. For example, using total population intake as a metric facilitates comparing motor vehicles to tobacco smoke as sources of exposure to benzene (Bennett et al., 2002; Nazaroff and Singer, 2004). Intake fraction could be used to

compare environmental impacts of transportation options such as train, bus, automobile, and ferry (van Wee et al., 2005). Intake fraction offers a top-down measure that summarizes exposure differences among pollutant and source types, accounting for issues such as the proximity between emissions and the receptors and the persistence of a pollutant in the environment. Nevertheless, the intake fraction does have limitations; these are presented in the second subsection below.

Additional uses for the intake fraction

This dissertation investigates people's inhalation of air pollution. However, the iF can be applied to other media, exposure pathways, or endpoints (MacLeod et al., 2004). Intake fractions can be a useful way to organize understanding about the complex emissions-to-intake relationship for multi-media, multi-pathway compounds. For example, semivolatile organic compounds (SVOCs) are inhaled as air pollution, and they are ingested via fruits and vegetables after depositing onto crops (Lobscheid et al., 2004). Intake fraction analyses can be constructed that account for multiple exposure pathways.

Intake fraction is among the many metrics that can be used to compare and corroborate model predictions. As I have done in Chapter 3, iFs can be used to pose the question of whether additional levels of sophistication in a model or an analysis add value or modify the results. Given the increasing cost, complexity, and in some cases potential for unseen errors associated with increasingly detailed analyses, iFs can be useful in evaluating the level of detail that is appropriate for a specific situation.

Limitations to using intake fraction

Intake fraction represents a particular way of organizing information. As such, it is useful in some but not all situations. Here are three important limitations to the intake fraction metric.

(1) Intake fraction is less useful for pollutants with a nonlinear rather than a linear dose-response relationship. In regulatory contexts, urban air toxics are typically assumed to have a linear dose-response relationship, but there may be cases when one wants to consider pollutants that exhibit threshold effects or non-linear dose-response relationships. For these pollutants, the intake fraction is still useful for understanding the relationship between emissions and inhalation intake. However, quantitatively estimating population health impact would require additional information. For example, for a pollutant with a linear dose-response relationship, but a threshold below which no health damage occurs, a modified intake fraction could be defined which incorporated the sum of intake above the threshold rather than total intake. Such potential applications of the intake fraction concept have not yet been substantially explored in the literature, nor are they addressed in this dissertation. In situations where the dose-response is highly nonlinear, population intake fraction may offer little or no useful insight.

(2) Intake fraction does not preserve information about the time pattern of exposure. Dose rate may be an important aspect of the health impact of certain specific pollutants. For example, the same dose of carbon monoxide that causes death if inhaled over a duration of 30 minutes might cause no impacts if inhaled over a duration of 10 hours.

(3) Intake fraction is not used in extant regulations. Specific analyses that are required in regulations, such as permitting decisions, may not find intake fraction to be useful. (At the same time, when regulators have flexibility in deciding how to meet air quality objectives, intake fraction may be useful as one component in a larger analysis.)

Conclusions

This chapter introduced the metric intake fraction, and explored how it might be used in air quality analyses. Typical intake fraction values for ambient emissions in urban areas are in the range $1 - 100$ per million, meaning that $1 - 100$ g are cumulatively inhaled by urban residents, per million grams emitted. Intake fraction values vary over time and by location, with values being higher for indoor than for outdoor emissions and also higher for urban than for rural emissions. Intake fraction values may be used in health risk assessment, cost-benefit analyses, environmental justice analyses, and in prioritizing emissions sources in terms of the impact on population inhalation of emissions. This chapter provides several examples of analyses and comparison that could be done using intake fraction, depending in part on what information is available. The chapter also explores attributes of emission sources (e.g., stack height, diurnal timing of emissions) that may influence intake fraction. Intake fraction is more applicable when considering pollutants with a linear, no-threshold dose-response, and less useful when considering pollutants with highly nonlinear dose-response relationships.

Table 2-1: Summary of intake fraction comparisons

Comparison	Key difference(s)	Typical difference in intake fraction values[a]
Urban versus rural emissions	Population density; proximity between emissions and people	Intake fraction is an order of magnitude larger for a large city than for a small city, and an order of magnitude larger for a small city than for a rural area (see Chapter 4).
Summer versus winter; night versus day	Dilution rate; atmospheric stability	Intake fraction is up to an order of magnitude larger for stable than for unstable atmospheric conditions (Lai et al., 2000). Intake fraction is ~2 times higher in winter than summer (see Chapters 3 and 4).
Stack height	Proximity between people and emissions; dilution reduces exposures	Intake fraction is up to an order of magnitude less for a tall stack than for a short stack or no stack (Evans et al., 2002; Lai et al., 2000).
On-road versus off-road	Proximity between people and emissions	Urban intake fraction is approximately 1.3 – 5 times larger for on-road sources as for off-road sources. (See calculations in the appendix to this chapter.) The comparable difference in rural on-road versus off-road intake fraction is unknown.
Self-pollution	Outdoor versus indoor or in-vehicle environment. Reduced dilution rate inside a vehicle or a building.	The rate of self-pollution is unknown for many sources. Potentially, self-pollution can increase intake fraction by more than an order of magnitude. For school buses in urban areas, self-pollution is expected to increase intake fraction by a factor of 2 – 10 (see Chapter 5).

Table 2-1 (Cont.)

Comparison	Key difference(s)	Typical difference in intake fraction values[a]
Coarse versus fine particles	Atmospheric persistence	Intake fraction is larger for fine particles than for coarse particles. A typical difference is a factor of ~3. (See calculations in this chapter.)
Primary versus secondary pollutants	Proximity between emissions and people; multiple chemical fates for species that are precursors to secondary PM	May be one or two orders of magnitude difference, with intake fraction larger for primary PM than secondary PM (Levy et al., 2003). Difference between primary and secondary gaseous pollutants has not been explored in the literature.
Indoor versus outdoor	Persistence of the pollutant in people's breathing zone; dilution rate	On average, indoor intake fraction is two to three orders of magnitude greater for indoor releases than for outdoor releases (Lai et al., 2000; Smith, 1993; Smith, 2002).

(a) Except for the row labeled "stack height", this table focuses on ground-level releases of pollutants. Except for the row labeled "primary versus secondary", this table focuses on conserved pollutants.
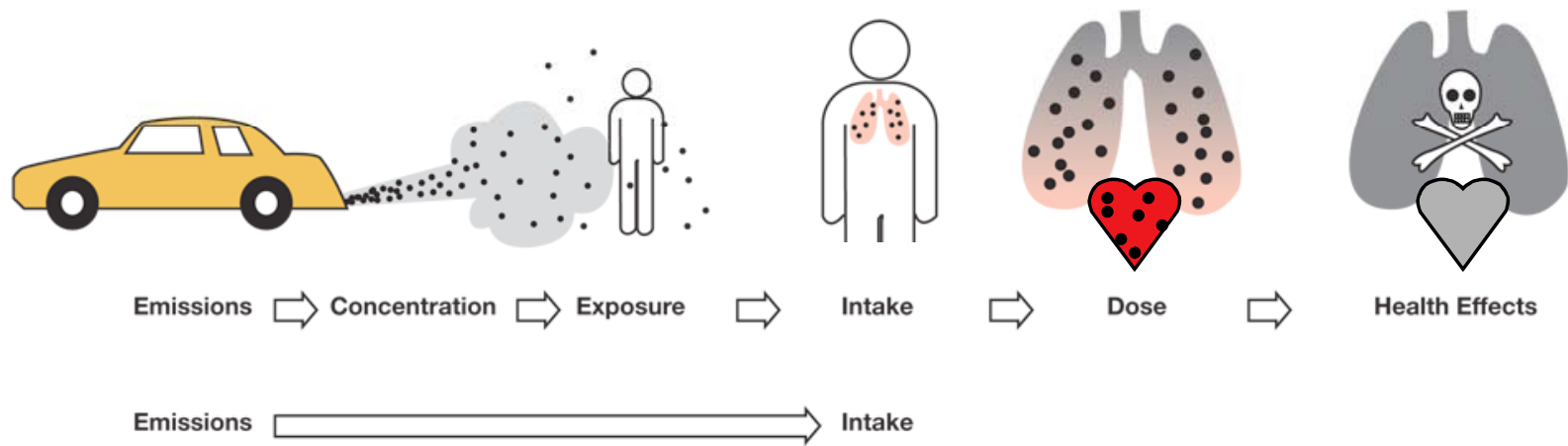
Figure 2-1: Emission-to-health effects relationships. Intake fraction summarizes the relationship between emissions and intake (figure after Smith, 1993).
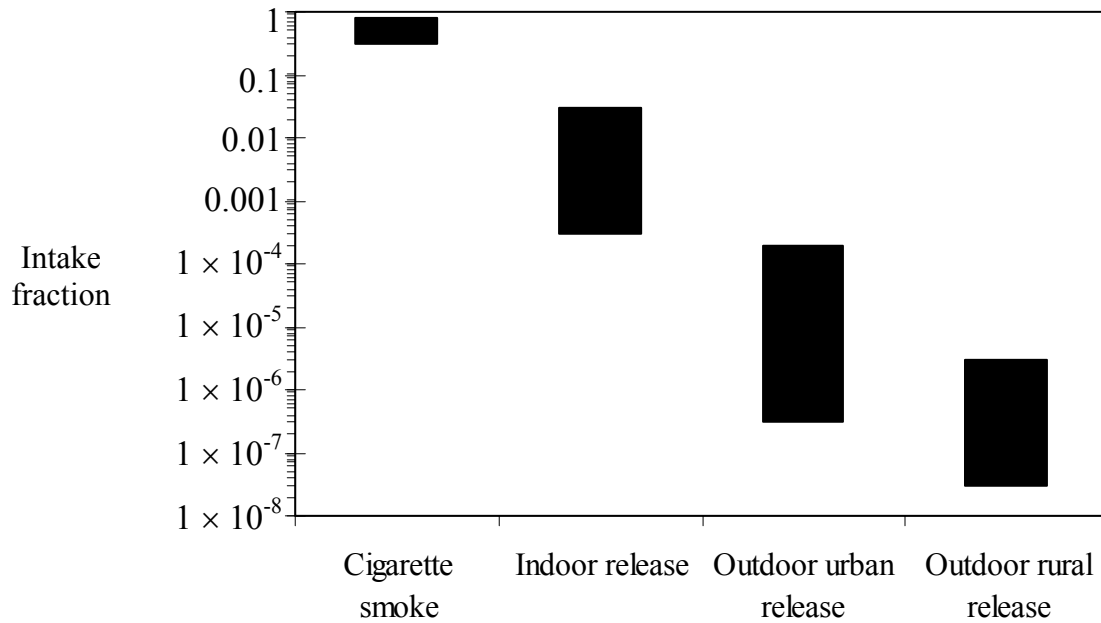
Figure 2-2: Typical intake fraction values for various emission situations. Values shown are illustrative and approximate, rather than definitive. Based on Lai et al. (2000).

Appendix: Calculations involving intake fraction for on-road versus not-on-road emission sources

This appendix presents and discusses calculations regarding the intake fraction of diesel PM and benzene emissions from on-road versus not-on-road emission sources. Calculations and data sources are given in Table 2A-1. Note that "not-on-road" is not the same as "off-road". The latter term is commonly used to indicate a subset of mobile sources (e.g., all-terrain vehicles operating on trails). I use the former term here to indicate sources, mobile or not, that are not on roads (e.g., also including an "area" source such as small industrial shops). For the purposes of this calculation, it is not necessary to distinguish, for example, between off-road mobile and area sources; both are considered here as not-on-road sources.

The table in this appendix contains calculations for both diesel PM and benzene. Cigarette smoke and other indoor sources of benzene are significant contributors to total population benzene inhalation (Wallace, 1996). This analysis does not consider indoor sources because it focuses on exposure to ambient pollution.

The main input parameters in Table 2A-1 are (1) the portion of emissions that are from on-road sources, (2) the percent of time spent in three microenvironments (in-vehicle, indoors, outdoors), (3) the ratio of in-vehicle concentration to ambient concentration, and (4) the ratio of indoor concentrations to ambient concentrations, considering only ambient sources. These four parameters are labeled as rows 1 – 4 in Table 2A-1. The table presents a range of values for the third input (ratio of in-vehicle concentration to ambient concentration), and point estimates for the remaining 3

parameters. Here, square brackets ("[ ]") are used to denote labeled row numbers in Table 2A-1.

The calculations are based on the assumption that ambient concentrations have been normalized to 1.0. All concentrations and intakes in the table are expressed relative to this (unitless) value. The breathing rate, which is not specified, is assumed to be constant over time.

The first calculation [rows 5a–5d] estimates the portion of population intake attributable to in-vehicle exposures [5d]. This value is equal to the in-vehicle intake [5a] divided by total intake [5c]. In-vehicle intake [5a] is estimated as the product of time spent in vehicles [2a] and the ratio of in-vehicle concentrations to ambient concentrations [3]. (Recall that the ambient concentration is assigned a normalized value of 1.0.) Not-in-vehicle intake is calculated as the time spent indoors times the ratio of indoor concentration to ambient concentration, plus the time spent outdoors. Total intake [5d] is the sum of in-vehicle [5b] and not in-vehicle intake [5c].

The second calculation [6a–6c] estimates intake attributable to on-road and not-on-road sources. This calculation assumes that all exposures in-vehicle that are in excess of ambient concentrations are attributable to vehicles, and that exposures to ambient pollution are attributable to vehicles and non-vehicles based on the fraction of emissions. The former assumption is robustly valid, almost by definition. The latter assumption is a reasonable basis for making these calculations. The intake attributable to not-on-road sources [6a] is equal to the portion of emissions that are from not-on-road sources (i.e., 1.0 minus the portion of emissions that are from on-road sources) times the intake of ambient pollution (which is a mixture of on-road and not-on-road sources). The estimated

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intake of not-on-road pollution by design excludes the higher intake rates in vehicles because these are assumed to be attributable to vehicles. This intake of ambient pollution is the sum of the time spent in-vehicles, the time spent outdoors, and the product of the time spent indoors times that ratio of indoor concentration to ambient concentration. The intake attributable to on-road sources [6b] is the total intake [5c] minus the intake attributable to not-on-road sources [6a]. The table also gives the portion of total intake that is attributable to on-road sources (i.e., the intake attributable to on-road sources divided by the total intake) [6c].

The third calculation [7a-7b] estimates the relative intake fraction for both on-road and not-on-road sources. The values presented in this portion of the table are not intake fraction values. Rather, intake fraction for the two sources (on-road and not-on-road) would be proportional to the values given in the table. This approach allows one to estimate relative intake fractions. The relative intake fraction for not-on-road sources [7a] is the intake attributable to not-on-road sources [6a] divided by the portion of emissions from not-on-road sources (i.e., 1.0 minus the portion of emissions from on-road sources [1]). The relative intake fraction for on-road sources [7b] is the intake attributable to on-road sources [6b] divided by the portion of emissions from on-road sources [1].

Finally, the fourth calculation [8] presents the ratio of intake fraction values for on-road and not-on-road sources. This value is the ratio of the two relative intake fraction values presented in the third calculation [7a and 7b].

The results in Table 2A-1 indicate that the diesel PM intake fraction is estimated to be 1.9 - 5.1 times as high for on-road sources as for other sources. Two reasons for this difference between vehicles and other sources are (1) proximity (on average, the in-

vehicle environment is closer than other environments to vehicle emissions) and (2) that buildings offer more protection against outdoor PM than do vehicles. There is uncertainty in the in-vehicle/ambient concentration ratio, as indicated by the two estimates presented above. Values are presented here for benzene as a comparison. There is less uncertainty in this ratio for benzene than for diesel PM. While both vehicle benzene and vehicle diesel PM have a proximity effect [3], only vehicle diesel PM has a building protection effect [4]. The benzene intake fraction is estimated to be 1.3 times as high for on-road sources as for other sources.

There is significant uncertainty in the portion of diesel PM from on-road sources versus from off-road sources [1]. If this value were 50% (rather than 25%), then the diesel PM intake fraction would be estimated to be 1.5 - 3.0 times as high (rather than 1.9 - 5.1 times) for on-road sources as for other sources. There is also uncertainty in the ratio of diesel PM concentrations in-vehicle versus in ambient air [3]. If this value were 3, as employed in Chapter 6, rather than 4, as employed in estimate #1 in Table 2A-1, then the diesel PM intake fraction would 1.6 times as high (rather than 1.9 times) for on-road sources as for other sources.

Table 2A-1: Summary of calculations involving intake fraction for on-road versus non-on-road emission sources

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Input data

			References/ Notes
1)	Portion of emissions that are from on-road sources		
	benzene	66%	(a)
	diesel PM	25%	(b)

2) Time-activity information for California
The "percent" values, rather than the "minutes/day" values are used below.

		minutes/day	percent	
2a)	in-vehicle	80	5.6%	(c)
2b)	indoors	1252	86.9%	(c)
2c)	outdoors	108	7.5%	(c)
	TOTAL	1440	100.0%	

3) Ratio of in-vehicle concentration to ambient concentration

	estimate #1	estimate #2	
benzene	4	4.5	(d)
diesel PM	4	14	(d)

4) Ratio of indoor concentration to ambient concentration, considering only ambient sources

benzene	100%	(e)
diesel PM	67%	(f)

Calculations

Assume the ambient concentration is normalized to 1.0. All concentrations are relative to this (unitless) value. Assume a constant (unspecified) breathing rate.

5) Intake apportioned by receptor location

5a) *In-vehicle* $= (2a) \times (3)$

	estimate #1	estimate #2
benzene	0.22	0.25
diesel PM	0.22	0.78

5b) *Not in-vehicle* $= (2b) \times (4) + (2c)$

	estimate #1	estimate #2
benzene	0.94	0.94
diesel PM	0.66	0.66

Table 2A-1 (Cont.)

5c) *Total* = **(5a)** + **(5b)**

	estimate #1	estimate #2
benzene	1.17	1.19
diesel PM	0.88	1.44

5d) *Portion of total intake attributable to in-vehicle exposures* = **(5a)** / **(5c)**

	estimate #1	estimate #2
benzene	19%	21%
diesel PM	25%	54%

6) Intake apportioned by emission location (i.e., on-road versus not-on-road sources)

Assumes that all exposures in-vehicle in excess of ambient concentrations are attributable to vehicles, and that exposures to ambient pollution are attributed to vehicles and to non-vehicles based on the fraction of emissions.

6a) *Attributable to sources other than on-road* = $(1.0 - (1)) \times ((2a) + (2c) + (2b) \times (4))$

	estimate #1	estimate #2
benzene	0.34	0.34
diesel PM	0.53	0.53

6b) *Attributable to on-road sources* = **(5c)** - **(6a)**

	estimate #1	estimate #2
benzene	0.83	0.86
diesel PM	0.34	0.90

6c) *Portion of intake attributable to on-road sources* = **(6b)** / **(5c)**

	estimate #1	estimate #2
benzene	71%	72%
diesel PM	39%	63%

7) Relative intake fraction

The values below are not intake fraction values. Rather, intake fraction for the two sources would be proportional to the values below. This approach allows us to estimate relative intake fractions.

7a) *Sources other than on-road* = **(6a)** / (1.0 - **(1)**)

	estimate #1	estimate #2
benzene	1.00	1.00
diesel PM	0.71	0.71

Table 2A-1 (Cont.)

7b) *On-road sources* = **(6b) / (1)**

	estimate #1	estimate #2
benzene	1.25	1.29
diesel PM	1.38	3.60

Conclusion

8) Ratio of intake fraction values: on-road sources relative to other sources

=**(7b) / (7a)**

	estimate #1	estimate #2
benzene	1.3	1.3
diesel PM	1.9	5.1

Notes

(a) US EPA (1993).

(b) CARB (2000c).

(c) Table 6 of Klepeis et al. (2001).

(d) The bases for the estimates of the ratio of in-vehicle concentration to ambient concentration (line 3) are as follows. For benzene, the values of 4 and 4.5 are from Chapter 3 and Rodes et al. (1998), respectively. For diesel PM, the values of 4 and 15 reflect the range of values reported by Fruin et al. (2004).

(e) Benzene, as a nonreactive gas, penetrates the building envelope without loss or removal. Based on mass balance, the long-term average concentration for ambient benzene is the same indoors as outdoors.

(f) Source: Fruin et al. (2004). The value of 0.67 for the ratio of indoor to ambient concentrations for diesel PM (Fruin et al., 2004) is consistent with indoor/outdoor

(I/O) ratios given by Riley et al. (2002) for elemental carbon of 0.73 for a typical infiltration ventilation residence, 0.38 for a residence with central air conditioning, and 0.63 for an office building with a 40% ASHRAE filter. For example, consider the following "back-of-the-envelope" approximation of the average indoor/outdoor concentration ratio. In 1999, ~36% of occupied housing units in the Los Angeles-Long Beach Metropolitan Area had central air conditioning (AC) (US HUD, 2001). Assume that (1) this value applies to all residences, (2) AC is used 5 months per year, and (3) during these five months, AC is on for 12 hours per day. Based on these assumptions, on average (36% × (5/12) × (12/24)=) 7.5% of housing units are using central air and the remainder (92.5%) are not. If people spend 1/3 of their indoor time in an office (I/O = 0.63), and 2/3 of their indoor time in a residence (I/O = 7.5% × 0.38 + 92.5% × 0.73 = 0.70), then the occupancy-weighted average I/O ratio for a building would be 0.68, which agrees closely with the value of 0.67 used above. This "back-of-the-envelope" calculation suggests that the value of 0.67 given by Fruin et al. (2004) is reasonable.

Chapter 3: Intake fraction of vehicle emissions in California's South Coast Air Basin

Introduction

Although strongly associated with photochemical smog and its harmful components, such as ozone and NO_x, motor vehicles also contribute significantly to ambient concentrations of hazardous air pollutants and certain primary criteria pollutants, such as carbon monoxide. Nationwide, on-road motor vehicles contributed 48% of US benzene emissions in 1996 and 51% of US carbon monoxide (CO) emissions in 1999 (US EPA, 2001c). In the South Coast Air Basin (SoCAB) of California, on-road motor vehicles contributed 70% and 80%, respectively, of total benzene and CO emissions (CARB, 2000b; SCAQMD, 2000). Previous investigations have emphasized the importance of motor vehicles as a dominant source of exposure to ambient benzene and CO (Flachsbart, 1999b; Fruin et al., 2001; Law et al., 1997; Macintosh et al., 1995).

In this chapter, I estimate intake fraction values for benzene and CO from motor vehicles in the SoCAB (see Figure 3-1) during 1996 – 1999. Prior to this research, no published report had analyzed ambient concentration data to quantify the intake fraction

(iF). Two previous investigations quantified the iF for motor vehicles based on air dispersion modeling. Evans et al. (2002) used a trajectory model, with 448 grid cells of 10,000 km^2 each, to calculate iFs for motor vehicle emissions on 40 highway segments throughout the United States. For primary PM$_{2.5}$, they report iFs of 3–18 per million for urban locations. Nigge (2001) combined two air dispersion models to calculate iFs of nine primary pollutants from point sources in Germany. For short-range transport (within 100 km), he used a Gaussian plume model. For long range transport (>100 km) he used a trajectory model with 10,000-km^2 grid cells. Intake fraction results are presented by Nigge for three pollutants: acetaldehyde (3–14 per million), PM$_{2.5}$ (8–18 per million), and PM$_{10}$ (3–12 per million). These results, which Nigge argues are applicable to motor vehicles, are similar to those of Evans et al. (2002). In contrast with these two studies, we estimate intake based on ambient monitoring data, and we explicitly include near-source exposures. Our research focuses on an urban area (17,460 km^2) that would occupy less than two grid cells in the trajectory models employed by Evans et al. (2002) and Nigge (2001).

Methods

For inhalation of a primary pollutant from an emission episode, the intake fraction can be expressed as follows (Lai et al., 2000):

$$\text{Intake fraction (iF)} = \frac{\text{Attributable intake}}{\text{Attributable emissions}} = \frac{\int_{T_1}^{\infty} \left(\sum_{i=1}^{P} (C_i(t) \cdot Q_i(t)) \right) dt}{\int_{T_1}^{T_2} E(t) \, dt} . \quad (3\text{-}1)$$

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Here, T_1 and T_2 are the starting and ending times of the emission episode; P is the number of people in the exposed population; $Q_i(t)$ is the breathing rate for individual i at time t ($m^3 h^{-1}$); $C_i(t)$ is the incremental concentration at time t in individual i's breathing zone that is attributable to the emission source ($g\ m^{-3}$); and $E(t)$ is that source's emissions at time t ($g\ h^{-1}$). In practice, the integral in the numerator would not be evaluated out to infinite times, but only until the incremental concentration attributable to the source of interest becomes negligibly small. For this study, it is only necessary to evaluate this integral from T_1 to T_2, because the duration of that interval (four years) is several orders of magnitude longer than the time scale for pollutant transport through an urban air basin (less than a day).

The method I employ for calculating the intake fraction requires information on three parameters or parameter groups: emissions, population size and breathing rate, and exposure concentration. Each of these parameters is discussed below. If there were no spatial or temporal variability in the exposure concentration, the intake fraction could be evaluated from a simple form of Equation 3-1. It would be computed as the product of the population size, the average breathing rate, and the average incremental exposure concentration attributable to a specific source, divided by the total emission rate for that source. However, a more detailed analysis is required for two reasons. First, publicly available concentration data come from monitoring stations that record ambient concentrations (most monitoring stations are located on the roof of a building), rather than from exposure concentrations. Second, spatial and temporal correlations among population density, breathing rates and exposure concentrations may increase the actual population intake (Hayes and Marshall, 1999).

Emissions

Emissions data, which are shown in Figure 3-2, are based on the California Air Resources Board's (CARB's) EMFAC database and model (CARB, 2000a). EMFAC calculates evaporative and exhaust emissions from on-road mobile sources based on monthly estimates of vehicle-miles traveled and of the age distribution of the vehicle fleet. Exhaust emissions are estimated from dynamometer tests, which are run according to Federal Testing Procedure (FTP) protocols, and from CARB's database of time spent in various operating modes, such as idling, accelerating, and startup. Evaporative emissions include drips, leaks, and "breathing losses" due to diurnal heating and cooling of the gas tank and the engine. Benzene is present in both evaporative and exhaust emissions, because it is a constituent of gasoline and also a product of incomplete combustion. Carbon monoxide is only present in exhaust emissions because it is formed during incomplete combustion but is not a gasoline constituent.

EMFAC directly estimates CO and total organic gas (TOG) emissions; it does not differentiate among the hydrocarbons that make up TOG emissions. I calculated benzene emissions by applying data from recent tunnel studies, which indicate that benzene is 3.3% of the TOG from exhaust emissions and 0.5% of the TOG from evaporative emissions (Kirchstetter et al., 1999a; Kirchstetter et al., 1999b). These two benzene percentages (3.3% for exhaust; 0.5% for evaporation) are assumed here to be constant over time. In reality, these percentages will vary diurnally and seasonally because they depend on the temperature (which differentially impacts the volatility of various ROGs) and on fuel composition (which changes seasonally). Gasoline was reformulated in

California in the mid-1990's, reducing benzene levels in gasoline and therefore also in evaporative emissions. EMFAC-estimated emissions used here average 495 tonnes per month for benzene and 196,500 tonnes per month for CO. For benzene, exhaust emissions are about an order of magnitude greater than evaporative emissions. EMFAC-estimated emissions used here are 92% exhaust and 8% evaporative.

Initially there were discrepancies between the bottom-up approach of EMFAC and the top-down approach of fuel-based emissions inventories (Fujita et al., 1992; Harley et al., 1997; Pierson et al., 1990; Singer and Harley, 1996). The former is based on scaling up the emissions from a sampling of individual motor vehicles, in terms of the emissions per km times the total km driven (Horie, 1995). The latter is based on the total fuel consumption times the emissions per liter (Singer and Harley, 2000). Recent versions of EMFAC agree with the fuel-based emission inventory to within about 20% (Singer and Harley, 2000).

Population size and breathing rate

The SoCAB is home to ~15 million people occupying 17,460 km^2 (6745 $miles^2$). In contrast, the population of California is ~34 million, and the population of the U.S. is ~285 million. Thus, the South Coast contains 44% of the population of California, and one in 19 US residents. The average population density is 860 people km^{-2} (2,200 people $mile^{-2}$).

Using an approach based on metabolic activity, Layton (1993) estimates age- and gender-specific breathing rates. To use the breathing rates in the intake fraction analysis, I first determined the population average breathing rate, and then estimated the diurnal

profile of breathing rate. Population average breathing rate is calculated from the data in Layton (1993) based on the fraction of the US population in 5-year age bins (under 5 years old, between 5 and 9 years old, between 10 and 14 years old, etc.) (US Census, 2001). The estimated population average breathing rate is 12.2 m^3 d^{-1}. As a comparison, I also calculated the lifetime average breathing rate for an individual, based on the data in Layton (1993) and assuming a lifetime duration of 75 years. To three significant figures, the result of this second calculation is the same as for the first calculation (12.2 m^3 d^{-1}). In contrast with the value of 12.2 m^3 d^{-1} used in this chapter, risk assessments often use a higher breathing rate (e.g., 20 or 25 m^3 d^{-1}) to account for interindividual variability and to provide a conservative intake estimate.

The next step is to determine the diurnal breathing rate profile (i.e., allocate the total volume of air breathed per day – 12 m^3 – to each 1-hour time step). To my knowledge, there are no available estimates of the diurnal profile for the population average breathing rate, nor are there data that would yield a robust estimate. In this dissertation, I present two independent estimates for this information, one in this chapter and one in Chapter 6. Layton (1993) provides the breathing rate and amount of time spent per day for five activity levels: sleep, light, moderate, hard, very hard. For the analysis in this chapter, I estimate the diurnal profile for the population average breathing rate by allocating total hours spent at each activity level to each 1-hour period during the day. This allocation is based on my own best estimate of the likelihood of a specific activity level occurring in each hour. Figure 3-3 presents the resulting diurnal profile, in terms of the population average breathing rate and the activity level. If better estimates of the

diurnal population average breathing rate become available, it would be straightforward to update my analysis to include this information.

Exposure concentration

Exposure concentrations are calculated from ambient concentrations, the time spent in various microenvironments (i.e., time-activity patterns), and the concentrations in these microenvironments. Each of these three parameters is discussed below. Monthly average ambient and exposure concentrations attributable to motor vehicles are shown in Figures 3-4a and 3-4b, respectively. Figure 3-5 shows the typical diurnal pattern for breathing rate and exposure concentration.

Ambient concentrations

The South Coast Air Quality Management District (SCAQMD) measures and records ambient pollutant concentrations at 34 air quality monitoring stations distributed throughout the South Coast Air Basin (SoCAB). During 1996 – 1999, 20 of these stations recorded one-hour average CO concentration every hour, for a total of 623,534 measurements. Six of these stations recorded 24-hour average benzene concentration approximately twice per month, for a total of 518 measurements. Additional information on the ambient concentration data is given in Table 3-1.

Monitoring station data and year-2000 population densities for the US Census tracts containing monitoring stations are combined to yield a population-weighted ambient concentration. That is, when determining the average ambient concentration, the

concentration recorded at each monitoring station is multiplied by a weighting factor. The weighting factor used is the population density in the census tract containing the monitor. Visual inspection of a map showing census tract population density and the locations of ambient monitor stations did not reveal a systematic bias between the population densities of the tracts containing monitors, as compared to the tracts surrounding each monitor. A more sophisticated approach would be to generate a three-dimensional surface of concentrations (i.e., concentrations throughout the two-dimensional ground surface), based on kriging or other interpolation methods. The additional resources necessary to implement such an approach are not justified for this analysis because there is only a modest degree of spatial heterogeneity in the ambient data. For example, the difference between the highest and lowest annual average ambient CO concentration is a factor of four (0.54 ppm at El Torro versus 2.3 ppm at Lynwood). The coefficient of variability among the stations (i.e., the standard deviation divided by the average) is only 0.33, again indicating only modest spatial heterogeneity. This result for the South Coast appears to hold for other urban areas throughout the US (see Chapter 4).

Because hourly ambient concentrations are available for CO but not benzene, I estimate hourly ambient benzene concentrations by applying the characteristic daily profile for CO concentrations in each month and year to the 24-hour average benzene concentration. The typical daily CO profile is shown in Figure 3-6. This approach assumes that benzene and CO exhibit similar profiles over the hours of the day. Expectations suggest that this assumption is approximately true, since both pollutants are nonreactive and emitted by motor vehicles, but it is not rigorously correct because CO comes from exhaust emissions while benzene comes from both exhaust and evaporative

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emissions. Evaporative benzene emissions peak during hot afternoons, while CO emissions peak during "cold start" conditions on cold mornings. The approach I have used, which does not account for these differences in the diurnal pattern between benzene and CO, is the best currently possible. If more detailed data were available on hourly ambient benzene concentrations, the analysis could be readily refined to incorporate the additional information.

One of the practical data-analysis challenges I encountered is accounting for concentrations that were below the detection limit, which occurred 5-6% of the time in the ambient CO and benzene data sets. There are several ways to utilize data with a significant fraction of nondetects. A straightforward method is to replace all non-detects with an arbitrary value, such as half the detection limit, two standard deviations below the mean, or zero. A more robust method is to replace each nondetect with a randomly generated value below the detection limit, based on statistical properties (e.g., geometric mean and geometric standard deviation) of the data above the detection limit. I tested both methods, and, for both CO and benzene, the different methods do not change the mean concentration significantly. This finding results from two attributes of this system: the data have a small fraction of nondetects, and the detection limit is small relative to the average measured values.

Time-activity patterns

Pollutant intake depends on time-activity patterns, which indicate how much time is spent in various microenvironments. Using National Human Activity Pattern Survey (NHAPS) data (Klepeis et al., 2001) provided directly to me by Klepeis, I examined three

microenvironments: in a vehicle, in a residence with an attached garage, and in all other

locations, whether indoor or outdoor. In a separate analysis, described below, I also

account for exposures in indoor locations that are immediately downwind of a freeway.

For the first microenvironment, I used data for the NHAPS category "in/near

vehicle." This category includes any outdoor activity that takes place inside or nearby a

transportation vehicle, such as riding in a vehicle, waiting for a bus/train/automobile, and

walking on a sidewalk. For the second microenvironment, I combined an estimate for the

Los Angeles-Long Beach Metropolitan Area that 60% of people have an attached garage

(US HUD, 2001) with NHAPS data on time spent in a residence. All other time is

allocated to the third microenvironment, which includes both outdoor (not in/near a

vehicle) and indoor (without an attached garage) locations. Of the 1.30×10^{20} person-

hours available annually to the 15 million residents of the SoCAB, 7% is spent in-vehicle,

41% is spent inside a residence with an attached garage, and the remainder (52%) is spent

elsewhere. Other microenvironments that have been used in benzene and CO exposure

assessments, such as bars or houses with natural gas cook stoves, are not needed to study

exposure only to motor vehicle emissions (Fruin et al., 2001; Kirchstetter et al., 1996;

Macintosh et al., 1995; Ott et al., 1992). Levels of benzene and CO from vehicles are not

expected to vary systematically across such microenvironments (e.g., in homes with

natural gas stoves as compared to homes with electric stoves).

Microenvironment concentrations

In locations that are in close proximity to motor vehicles, the exposure

concentration tends to be higher than the measured ambient concentration. The estimated

average concentration in each of four microenvironments (in-vehicle, in a residence with an attached garage, in a building near a freeway, and all other locations) is presented below. The results below are consistent with an EPA study that uses 37 microenvironments (US EPA, 2001a). The population-average increase in the exposure concentration over the ambient concentration, owing to microenvironmental enhancements, is presented in Figure 3-5 as the normalized exposure concentration.

In-vehicle concentrations. Concentrations of vehicle pollutants such as benzene and CO are higher in vehicles than at ambient monitoring stations owing to differences in proximity to the emissions source. When in traffic, a person is potentially exposed to pollutants from (1) ambient urban air, (2) surrounding vehicles, and (3) the vehicle in which that person is riding. The first type of pollution, ambient pollution, would be recorded at ambient monitoring sites, and is attributable to vehicle emissions throughout an urban area and upwind of an urban area. Ambient concentrations vary with time of day, day of week, and season, because of differences in emissions and meteorology. The second type of pollution is attributable the vehicle passing through "micro-plumes" from the nearby surrounding vehicles. Time-average concentrations attributable to these micro-plumes will vary, depending on wind speed and direction, traffic density and speed, distances between vehicles, and emissions from neighboring vehicles. The third type of pollution, termed "self-pollution", is explored in more detail in Chapter 5 for the case of children's inhalation of school bus emissions. It is generally believed that self-pollution is relatively minor for most private passenger vehicles. Here, it is necessary to know how average concentration in vehicles differ from average ambient concentrations, but it is not

necessary to disaggregate in-vehicle concentrations into the three types of pollution listed above (ambient, nearby vehicle, self-pollution).

Flachsbart (1995; 1999a) reviewed 14 studies reporting in-vehicle and ambient concentrations of CO. All 14 studies reported concentrations to be higher in-vehicle than in ambient air, to a degree that is relatively consistent over time and among cities. The ratio of in-vehicle concentration to ambient concentration is found to be in the range $2 - 5$ for most studies, with an average value of 3.5. The three studies conducted in Los Angeles, California, reported values of 2.3, 2.9, and 3.9 for this ratio. For benzene, Weisel et al. (1992) reported this ratio as 5.8 in winter and 10.0 in summer, for an annual average of 7.9. These values from Weisel et al. (1992) are consistent with results from the multi-city TEAM study (Wallace, 1996), which reported typical benzene concentrations 6.0 $\mu g\ m^3$ in ambient air and $30 - 40\ \mu g\ m^3$ in vehicles, suggesting a ratio of $5.0 - 6.7$. The average value for this ratio from among the studies conducted in Los Angeles (values: 2.3, 2.9, 3.9, 7.9) is 4. Based on this value, I assume here that in-vehicle concentrations are 4 times higher than ambient concentrations.

Concentrations in residences with an attached garage. In a residence with an attached enclosed garage, vehicle emissions into the garage can migrate into the household via airflow coupling between the garage and the living space of the house (CMHC, 2001; Lansari et al., 1996). This coupling can increase in-residence concentrations of vehicle pollutants (Graham et al., 2004; Thomas et al., 1993). The seasonal and diurnal profiles of in-garage emissions will differ for CO and benzene. As a product of incomplete combustion, CO is only emitted when the engine is operating (all

CO emissions are tailpipe emissions). Benzene emissions include tailpipe and non-tailpipe components. In-garage tailpipe emissions occur when a vehicle enters and exits the garage, and whenever the vehicle is warming-up or otherwise idling in-garage. Tailpipe emissions of CO and benzene are greater in the morning before the car leaves, when the car is idled in-garage to warm up the engine and before the catalytic converter is warm enough to function fully, than in the evening after the car returns home, when the catalytic converter is warm and the engine is likely to be turned off soon after entering the garage. Non-tailpipe benzene emissions (e.g., breathing losses) can occur whenever the vehicle is present, whether or not the engine is operating, and also potentially when the car is not present (e.g., evaporative emissions of gasoline drips and leaks on the garage floor). In general, evaporative benzene emissions are greatest during high ambient temperatures (daytime in the summer), and breathing loses are greatest for days with a wide diurnal temperature range.

Evidence suggests that coupling between in-garage emissions and a household may, under certain circumstances, be significant. A clear reminder of this fact is the accidental CO poisonings that have been attributed to high-emitter vehicle emissions (Marr et al., 1998). In addition, measurements of household CO levels indicate that CO concentrations increase following in-garage emissions (IES, 1995).

One recent study that explored this issue measured indoor, in-garage, and outdoor concentrations of CO and several VOCs (Liu et al., 2004). Unfortunately, the results from that study are not directly applicable to the current investigation, for two reasons. First, that study considered homes in Anchorage, Alaska, in winter. Few study conditions (e.g., meteorology, personal activities, home design) in the US would be *less* representative of

Southern California. This issue of applicability to conditions in Southern California applies generally to other studies conducted in cold climates (e.g., Graham et al., 2004). Second, by design, garage VOC emissions in the Anchorage study included gasoline equipment (e.g., lawn mower, snow blower) and other VOCs stored in the garage. The Anchorage study, like several other similar studies (e.g., Thomas et al., 1993), did not disaggregate indoor VOC concentrations attributable to garage emissions into vehicles and non-vehicle components, yet this desegregation would be necessary for me to directly use the data. Nevertheless, the Anchorage study (Liu et al., 2004) concluded that CO emissions from cars did not significantly increase long-term indoor CO concentrations. If this conclusion holds for Anchorage, where "warming up" a car is significantly more likely than in Southern California, then it is likely to hold for the South Coast.

Based on limited tailpipe CO emissions in garages (as evidenced by the results from Anchorage) and no evaporative CO emissions, I estimate that motor vehicle-caused CO concentrations in all houses (with or without an attached garage) are the same as the local ambient value. The brief period of time that people spend in-garage, between exiting the vehicle and entering the house, is not expected to significantly increase the total population inhalation of CO or benzene.

I estimate here that residences with an attached garage have vehicle-associated benzene concentrations that are ~20% higher than the ambient counterparts. This value is derived from measured benzene concentrations in 300 California homes during the period December 1991 – April 1992 (IES, 1995). On average, the difference between indoor and outdoor concentrations was reported as 0.06 ppb for no attached garage, 0.2 ppb for an

attached garage with no parked car, 1.1 ppb for one car, and 1.9 for two cars (Fruin et al., 2001). These values indicate that vehicle emissions in garages, excluding non-vehicle in-garage benzene emissions, contributed ~0.8 ppb to indoor benzene concentrations, or a ~20% increase over typical ambient benzene levels of 3.4 ppb in the South Coast at the time.

Indoor concentrations near freeways. People in houses and other buildings in the vicinity of freeways will tend to experience higher concentrations than the ambient concentrations at monitoring stations because of the close proximity to a relatively major emissions source. I analyze time spent indoors near freeways separately because this microenvironment is not included in the NHAPS data. As with the previous microenvironments, the key questions are "How much higher are typical concentrations, compared to the ambient concentration?" and "How much time does the population spend in this class of microenvironments?"

For the approach employed here, answers to these questions can be derived from experiments or models, or both. Similar to the houses with an attached garage microenvironment, using experiments to measure the impact of a freeway in a way that the results could be generalized would require careful experimental design and would likely be highly data-intensive. Alternatively, a modeling approach can more easily estimate concentrations downwind of a generic freeway. Combining Gaussian plume model results for a line source with data showing in-vehicle concentrations as four times ambient concentrations (Fernandezbremauntz and Ashmore, 1995; Flachsbart, 1995), I estimate that average concentrations of motor vehicle emissions near a major roadway are

twice the basin-wide ambient concentration. This estimate accounts for the rapid decrease in concentration immediately downwind of a major source due to atmospheric dispersion. "Near" is defined here by the distance downwind of a freeway wherein the observed concentration is significantly higher than the ambient concentration because of that roadway's emissions. Drivas and Shair (1974) found this distance to be less than 100 m. This result agrees broadly with the Gaussian plume dispersion equation for a line source (Nazaroff and Alvarez-Cohen, 2001), which indicates that this distance is typically less than 300 m. Both of these analyses assumed the wind is perpendicular to the freeway. Since all other wind directions will result in lower values for this characteristic distance, 200 m represents a reasonable upper bound for the average characteristic distance. Zhu et al. (2002) measured CO concentrations at 17, 20, 30, 90, 150, and 300 m downwind and 200 m upwind from the center of a Los Angeles freeway (Interstate 710). Their results are consistent with Drivas and Shair (1974) and with Nazaroff and Alvarez-Cohen (2001): the on-road spike in CO concentrations diminishes after 100–300 m. Similarly, the East Bay Children's Respiratory Health Study reported that concentrations of several vehicle-related pollutants "were higher at schools located within 300 m downwind of a freeway compared with those at schools upwind or further from major traffic sources" (Kim et al., 2004).

Combining the characteristic distance of 200 m with the length of freeways in the SoCAB, 3316 km (2061 miles) (Bhat, 2001) yields 660 km^2 of "near-freeway" land, or 4% of the total area of the SoCAB. For this portion of the analysis, I assume that the population density is uniform throughout the basin, and therefore ~4% of the people in the SoCAB are in buildings near freeways at any given time. While the assumption of

uniform population density is rough, it is sufficiently accurate for this calculation as my final results are not sensitive to this parameter. Although there are major roads that are not freeways, I have not accounted for them explicitly in this analysis because their impact on concentrations is partially reflected in the ambient concentration data.

Concentrations in other locations. In all locations other than the three microenvironments already discussed, the exposure concentration is taken to be equal to the measured ambient concentration. Both benzene and CO are relatively nonreactive gases, and outdoor concentrations readily penetrate into indoor environments without loss. Indoor environments may have additional sources of benzene or carbon monoxide, such as gas stoves, cigarette smoke, or, at an ice skating rink, Zamboni emissions, but the existence of these sources does not alter exposure to on-road vehicle emissions. (The existence of these indoor sources makes it difficult to design exposure monitoring studies to directly confirm predicted iF values for vehicle emissions of benzene and CO.)

Results

As shown in Figure 3-2, basinwide emissions are relatively constant throughout the year. However, ambient concentrations of benzene and CO (Figures 3-4a and 3-4b) are about twice as high in winter as in summer. The varying concentration-to-emissions ratio generates a similar seasonal pattern in the intake fraction, as is shown in Figure 3-7. This variability is a consequence of varying seasonal meteorological patterns. Atmospheric transport and dispersion are lower on average during the winter because of the weaker incident solar radiation. Poorer pollutant transport means that the same

emissions of primary pollutants will lead to higher ambient concentrations. Both the wind speed and the atmospheric mixing height are lower in winter (NREL, 1995), leading to higher concentrations in winter. Episodes of extreme air pollution, such as the infamous "killer fog" that occurred in London during December 5 – 9, 1952, are associated with periods of highly stable meteorological conditions. This seasonal concentration pattern for primary pollutants – with higher concentrations in the winter – is the reverse of the pattern for ozone and other photochemical smog constituents, which have higher concentrations in the summer owing to much higher solar radiation.

Based on the methodology laid out above, including microenvironments, hourly ambient concentrations, breathing rates, and time-activity pattern data, I calculate annual average intake fractions of 46 per million for CO and 49 per million for benzene. These estimates mean that approximately 50 grams of primary motor vehicle pollutants are inhaled for every million grams of primary pollutants emitted in the South Coast Air Basin. The iF is slightly higher for benzene than for CO, owing to the slightly increased exposures from attached garages, but this difference is small compared to the seasonal variability for both benzene and CO. As is shown in Figure 3-7, wintertime iFs are almost twice summertime iFs; this pattern is due to the seasonal variability in ambient concentrations (shown in Figure 3-4a). Intake fraction estimates by month are given in Appendix II for this chapter. Using 48 months of data, with a single intake fraction calculated for each month, I calculate a standard deviation in the monthly average iF of 20 per million for benzene and 15 per million for CO. Note that these standard deviations represent variations in the monthly intake fraction, not uncertainties associated with the estimate.

Because there are significantly more concentration data for CO than benzene, the monthly-average CO concentration measurements are more consistent from one year to the next. Consequently, my results show about twice as much interannual variability in the iF for benzene as for CO.

Discussion

My results are consistent with existing research. Based on previous publications, I expected the time-average iF for an outdoor release in an urban area to be on the order of $1 - 100$ per million. For example, using Gaussian plume equations, Lai *et al.* (2000) calculated iF values of $4 - 230$ per million for episodic outdoor sources, depending on the meteorology, population density, and urban area. Smith (1993) reported 20 per million as an order-of-magnitude estimate for outdoor ground-level emission sources in urban settings. Evans *et al.* (2000) used a Gaussian plume model to calculate an intake fraction of $3.6 - 13$ per million for ambient dry cleaner emissions in the US (excluding indoor exposures, such as to workers and customers). (Note that to compare the Evans *et al.* (2000) results with other values listed in this chapter, I have modified here their reported iF of $6 - 22$ per million to account for the different breathing rate they used, $20 \, \text{m}^3 \, \text{d}^{-1}$ rather than $12.2 \, \text{m}^3 \, \text{d}^{-1}$.) Schauer *et al.* (1996) reported a value of 0.4 ($\mu\text{g m}^{-3}$) per (t d^{-1}) for the ratio of ambient concentration increase to emissions for elemental carbon from diesel exhaust in downtown Los Angeles[1]. Multiplying this value by an inhalation rate of $12.2 \, \text{m}^3 \, \text{d}^{-1}$ and a population of 7 million for the ~1600 km^2 region they used yields an

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[1] Owing to a typographical error, the units in the publication are given as $\mu\text{g/m}^3$ per kg/day.

iF of about 34 per million for this urban emission source. A study of Taipei City, Taiwan, stated that because of government efforts over the past two decades to relocate point sources to outside the city, over 99% of urban CO emissions are from motor vehicles (Chen et al., 2002). They present modeled and measured CO concentrations of 1.1 ppm, a population of 2.6 million people, and CO emissions of 400,000 tonnes y^{-1}. Using a breathing rate of 12.2 m^3 d^{-1}, their results indicate an iF of 39 per million. Consistency between previous findings and the results presented here substantiates the general accuracy of my results and reinforces the potential utility of the intake fraction concept for air pollution exposure assessments.

In addition, the close agreement between the iFs for benzene and CO also reinforces the validity of the intake fraction approach. CO and benzene from motor vehicle emissions should have similar iFs because they have similar fate and transport characteristics in the atmosphere. The dominant removal mechanism from the air basin for nonreactive gases is advection, and nonreactive gases penetrate the building envelopes without impedance or removal. I characterize CO and benzene as relatively nonreactive because their characteristic lifetimes in urban atmospheres (882 hours for CO (CARB, 1999) and 490 hours for benzene (US EPA, 1993)) are significantly greater than the typical residence time of air in the air basin (7 – 16 hours, see Appendix I for this chapter).

Within a specific air basin, the iF for any gas emitted from a broadly distributed outdoor urban source should be similar to the iF for CO and benzene if its characteristic lifetime is significantly greater than ~8 daylight hours. A gas emitted from a distributed

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source with a lifetime less than ~8 hours will have a smaller iF because some of the emissions will degrade before people inhale them.

For emissions with a relatively short lifetime (less than ~1 hour), a significant fraction of the total intake will occur during near-source exposures, such as the time spent in vehicles. For such a compound, it is more difficult to deduce the average concentration to which people are exposed from a limited number of ambient monitoring stations.

Intake fraction results presented in this chapter are not directly applicable to other urban locations. Differences in the intake fraction could arise because of differences in meteorology, such as the wind speed, rate of dispersion, and mixing height, or because of differences in the city, such as the size and population density. Chapter 4 provides estimates of intake fraction for motor vehicle emissions in other urban areas throughout the US.

Applying the results for benzene and CO to other chemicals

Understanding about exposures is built on models and measurements. Measurements are necessary to provide input data and to validate models. Properly validated models allow the testing of a variety of questions and hypotheses for which experiments are difficult or impossible to undertake.

Intake fractions facilitate the application of insights gained in one investigation to other, related, investigations. For example, models and measurements can be used to calculate intake fraction values, which can then be applied to new situations or compounds beyond those specifically modeled or measured. In Table 3-2, I have applied my results for benzene and CO to several chemicals that are emitted from motor vehicles.

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The estimation of exposure to these compounds owing to motor vehicles would be difficult based on a pure monitoring approach. Intake fractions represent an innovative and practical method for estimating the total population intake to these chemicals due to motor vehicles.

Table 3-2 lists the emissions, as given by the 1996 Toxic Emissions Inventory; intake fraction, as calculated in this chapter; and intakes for six compounds emitted from motor vehicles. The iF value of 0.0048%, or 48 per million, is only valid for primary pollutants that are nonreactive on the time scale of an urban air basin. Table 3-2 shows how intake fractions can be decreased, using the equations in Appendix I for this chapter, to account for the presence of removal reactions. (The benzene emissions shown in Table 3-2 are from the published 1996 inventory, rather than directly from EMFAC, to maintain consistency with the data on other chemicals listed in the table.) The total population inhalation burden presented in this table could be used in several analyses, e.g., in a health risk assessment, to track changes in intake rates over time, to compare against other urban areas, or to compare the intake rates attributable to outdoor and indoor pollution.

Comparison to ambient concentration analysis

As a comparison with my main intake fraction estimate of 48 per million, I completed a second analysis using the average ambient concentration as a direct surrogate for the exposure concentration. For this simplified analysis, I computed the intake as the product of the monthly average ambient concentration, the fraction of emissions attributable to motor vehicles (70% for benzene and 80% for CO (CARB,

2000b; SCAQMD, 2000)), the population size, and the monthly breathing rate per person.

The resulting intake fraction estimates are 33 ± 14 per million for benzene and 32 ± 11

per million for CO, or 32% less than obtained by the more detailed analysis. The standard

deviations presented here indicate the monthly variability in the values, rather than the

confidence interval or associated level of error.

The simplified analysis, which is summarized in Table 3-3, agrees well with the

values of 34 and 39 presented above for downtown Los Angeles and Taipei City,

respectively (Chen et al., 2002; Schauer et al., 1996). Those estimates reflect a similar

level of simplification in their analysis (i.e., the Schauer et al. (1996) and Chen et al.

(2002) studies investigated ambient concentrations rather than population intakes).

The more detailed analysis accounts for several factors that may be important in

determining the total intake, such as variability in concentration and breathing rates, and

elevated concentrations in certain microenvironments. However, considerably more input

data and processing time are required to complete the more complicated analysis. My

study suggests that if urban population exposures for motor vehicle emissions are needed

to within a error bound of approximately 50%, then the less complex analysis may be

adequate. Additional detailed studies of other sources and other urban air basins are

necessary to confirm this inference.

Exposures occurring outside the air basin

The intake fraction results determined in this study only account for exposures

that occur within the same air basin as the emissions. In reality, some exposure will occur

outside of that air basin because of atmospheric transport of the pollutants from one air

basin to another. I used a material-balance equation based on a box model to estimate

exposures occurring *outside* the SoCAB that are attributable to motor vehicle emissions

occurring *inside* the SoCAB. The details of this analysis, including the equations and data

used, are given in Appendix I to this chapter. I examine both a conserved pollutant and a

reactive pollutant that has a characteristic lifetime of 80 hours. The characteristic reaction

lifetime for benzene in Los Angeles (i.e., accounting for chemical reactions but not loss

by advection) is 80 hours in the summer, and 900 hours (37 days) in the winter (US EPA,

1993). The rate of degradation via attack by the hydroxyl radical will decrease once the

air leaves the urban air basin, because of lower hydroxyl radical concentrations in less

polluted environments. Consequently, the lifetime of benzene will increase after leaving

the SoCAB in the summer. Thus, a lifetime of 80 hours, when applied to areas downwind

of the South Coast, represents a compound that is more reactive than benzene in the

summer.

I first look at the regional intake outside the SoCAB by assuming that motor

vehicle emissions from inside the SoCAB mix throughout the two air basins that border

the South Coast to the east (the Salton Sea Air Basin and the Mojave Desert Air Basin).

These two air basins combined occupy 84,240 km^2, or about five times the land area of

the South Coast, and they are inhabited by 1.3 million people, or less than one-tenth the

population of the South Coast (CARB, 2002b). Next, I estimate the contribution of

SoCAB motor vehicle emissions to nationwide exposure, using an area of 9,157,000 km^2

and a population of 281,422,000 people (US Census, 2002).

For both the regional and nationwide analyses, I assume a population breathing

rate of 12.2 m^3 person^{-1} d^{-1} and a wind speed of between 3.36 and 5.50 m s^{-1}. These wind

speeds are the 5% and 95% values of the distribution of harmonic mean wind speed for the 75 cities listed in EPA's SCRAM database (US EPA, 2002). They represent the average throughout the mixing depth, rather than being the surface wind speed. For the regional box, I use a mixing height of between 600 and 3,000 m, representing an assumption that the pollutant mixes throughout 5% to 25% of the troposphere. In comparison with the lower estimate of 600 m, the elevation difference between Beaumont, California (located inside the air basin, near the eastern border) and Coachella, California (located outside of the air basin, also near the eastern border) is ~ 820 m. The mountain peaks surrounding Beaumont are significantly higher than Beaumont, so the altitude drop upon leaving the air basin is greater than 820 m in many places. Thus, 600 m represents a reasonable lower estimate for the mixing height of air exiting the South Coast. In addition, the range of 600 – 3,000 meters for the Salton Sea and Mojave Desert Air Basins is consistent with current research by Heath on typical mixing heights for Bakersfield, CA. The interquartile values (i.e., the 25th and 75th percentile values) for a derived Typical Meteorological Year are 630 – 2,870 m (Heath, 2005). For the national box, I use a mixing height of 3,000 to 12,000 meters, representing an assumption that the pollutant mixes throughout 25% to 100% of the troposphere.

For a conserved pollutant, I calculate incremental iFs (in units of per million) of 0.04 – 0.45 regionally and 0.20 – 1.85 nationally. For a reactive pollutant (k^{-1} = 80 h) I calculate incremental iFs of 0.03 – 0.31 per million regionally and 0.07 – 0.34 per million nationally.

The reactive pollutant (k^{-1} = 80 h) would be considered moderately reactive when compared to the residence time of air in the US air basin (150 – 360 hours). However, it

would be considered relatively nonreactive when compared to the residence time of air in the regional air basin (15 – 34 hours). This distinction leads to an important result. The regional iF for the reactive pollutant is only 20 – 40% less than the regional iF for the conserved pollutant. On the other hand, the national iF for the reactive pollutant is 3 to 5 times lower than the national iF for the conserved pollutant. All else being equal, a more reactive compound will tend to have a smaller intake fraction.

Combining the regional and nation wide intakes, I estimate a total incremental iF of 0.10 – 0.65 per million for a reactive pollutant and 0.24 – 2.30 per million for a nonreactive pollutant. These results for exposures *outside* the SoCAB are 20 – 500 times less than the iF for exposures that occur *inside* the SoCAB. Consequently, for the case being studied, regional and national intakes of primary and reactive pollutants are significantly less than intra basin intakes of urban emissions.

The analysis of downwind air basins presented above is based on the prevailing wind direction, which is an offshore wind (i.e., from the west or southwest). This prevailing wind direction dominates the wind-rose, with 51% of hours in a Typical Meteorological Year (TMY) having wind from $220 – 280^{\circ}$ (Heath et al., 2003), where 90° indicates wind blowing from the East. During other hours, however, air pollution may enter air basins other than the Salton Sea and Mojave Desert. One way to investigate this issue would be to consider a Lagrangian model, which would follow a parcel of air migrating from the South Coast to other air basins. This approach could account for direct and indirect migration of air between air basins. For example, an air parcel could travel directly from the South Coast to San Diego, or it could migrate from the South Coast to the ocean, then southeast over the ocean, then eastward to San Diego.

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My analysis here employs a simple analysis that accounts for direct flow between air basins and ignores indirect flow patterns. In this analysis, wind blowing perpendicular to the shore leads to pollutant transport to the Salton Sea and Mojave Desert Air Basins, while wind parallel to the shore leads to pollutant transport to the South Central Coast Air Basin (population: 1.54 million people; area: 20,450 km^2) or the San Diego Air Basin (3.15 million people; 10,890 km^2) (CARB, 2005). The interquartile range for the term *uH*, based on the distribution of values from TMY data for San Diego, is 840 – 3,320 m^2 s^{-1} (Heath, 2005). Applying this value to San Diego and the South Central Coast, and emplying the same one-compartment model used for the Salton Sea and Mojave Desert, I estimate that the intake fraction (units: per million) is 1.3 – 5.1 in the San Diego Air Basin and 0.5 – 1.8 in the South Central Coast. Next, I assume here that wind from 220 – 280$^°$ transports pollutants to San Diego, while wind from 100 – 170$^°$ transports pollutants to the South Central Coast. An analysis of TMY data for Los Angeles indicates that the former condition annually occurs during 9.9% of the hours and the latter condition occurs during 13.8% of the hours (Heath et al., 2003). Combining these values, the time-weighted average intake fraction (per million) for nonreactive South Coast vehicle emissions being inhaled by residents of the four air basins surrounding the South Coast is 0.4 – 2.5. This range is similar to the range presented above of 0.2 – 2.3, which accounts for intakes in the Salton Sea and Mojave Desert but not San Diego or the South Central Coast. Thus, I conclude that for vehicle emissions in the South Coast, the inhalation rate is significantly larger for South Coast residents than for residents of the four nearby air basins.

As a comparison, the box model is also used in Appendix I to predict the intake fraction for exposures occurring inside the SoCAB. The results are in the range 12 to 82 per million, which evenly brackets the values of 46 and 49 per million presented above for CO and benzene. This consistency lends support to the validity of the box model for estimating incremental intake fractions.

Uncertainty and sensitivity analysis

My research quantifies the population intake of motor vehicle emissions, a parameter that is not amenable to direct measurement. An uncertainty analysis is important to determine the confidence that can be placed in the results and methodology. In addition to the uncertainty analysis presented here, the uncertainty in the results could also be ascertained by comparing these results with future research that uses an alternative method to quantify the population intake of motor vehicle emissions.

Systematic errors can reduce the accuracy of a measurement and random errors can reduce the precision of a measurement. Systematic errors refer to biases that lead to consistent under- or over-estimation, while random errors are fluctuations that lead to a specific measurement being too high or too low. For example, if a laboratory technique to measure the concentration of a specific compound in an air sample has random errors but no systematic errors, then duplicate measurements of a single air sample will yield different results while the average of many duplicate samples will approach the correct answer as the number of samples increases. Such measurements are characterized as accurate but not precise, because the average is correct even though any individual measurement is not necessarily correct. In contrast, if a laboratory technique has

systematic errors but no random errors, then duplicate measurements of a single sample will yield similar results, all having a similar level of error. Such measurements are precise but not accurate, because even though there is a high level of consistency between measurements, the average of several measurements is different from the correct value.

Among the data that I use in this investigation, four parameters dominate in influencing the results. These four parameters are emissions from motor vehicles, ambient concentrations, the size of the population, and the breathing rate. In addition, a second set of parameters is relatively important, though less so than the previous four. This second list is concentrations in vehicles, time spent in vehicles, concentrations inside residences with an attached garage, and population density at each of the ambient monitoring stations.

Below, I first complete an uncertainty analysis for the four input parameters that dominate my results (emissions, ambient concentrations, population size, and breathing rate). Then, I conduct an approximate sensitivity analysis for the four additional parameters (concentrations in vehicles, time in vehicles, concentrations in residences with an attached garage, and population density at each monitoring station). Finally, I provide a qualitative discussion of methodological uncertainties.

Uncertainty analysis

Emissions. The EMFAC emissions model, which yields monthly emissions estimates, is maintained by the California Air Resources Board (CARB). CARB does not provide an uncertainty estimate for the EMFAC results. A comparison between EMFAC and a fuel-based emission inventory (Singer and Harley, 2000) indicates agreement to

~20%, with EMFAC yielding lower estimates. This level of agreement is a significant improvement from previous versions of EMFAC, which were a factor of 2 – 4 lower than fuel-based emission inventories (Fujita et al., 1992; Harley et al., 1997; Pierson et al., 1990; Singer and Harley, 1996).

Ambient concentrations. Because of the relatively large number of measurements (623,534 CO measurements and 518 benzene measurements), random error in individual measurements will tend to average out and not bias the final result. However, the potential exists for systematic error in the measurement technique.

The CO and benzene monitoring stations are audited on a regular basis using a laboratory-prepared calibration sample of a known concentration. During the years considered (1996 – 1999), audits of monitors throughout California yielded an average percent difference between the calibration sample and the monitor's measurement of 0.5% and -11% for CO and benzene, respectively (CARB, 2001; Miguel, 2002). These audits indicate that CO monitors have a high degree of accuracy while benzene monitors tend to underestimate the true concentration somewhat.

Population size. The US Census Bureau does not directly report uncertainty. An indirect indicator of the level of accuracy in their data is the undercount rate, which is the amount by which survey tally results are increased to account for people not surveyed. Publicly available census data have been corrected to account for the undercount rate. In California and the US, the undercount rates are 2.7% and 1.6%, respectively. In Los Angeles, Orange, Riverside, and San Bernardino Counties, the rates are 3.3%, 2.1%,

2.4%, and 2.6%. These values indicate a relatively high degree of precision and accuracy in census results, as compared with other major input parameters in this analysis.

Breathing rate. There is no direct information on the accuracy and precision of the breathing rate data I use (Layton, 1993). Variability in breathing rates offers a proxy for the level of accuracy. Lifetime average rates for men and women are 14.1 and 10.2 m^3 d^{-1}, respectively. These two values are 2.0 m^3 d^{-1} different from the average breathing rate of 12.2 m^3 d^{-1}. Breathing rates for people 18 and under are 11.2 m^3 d^{-1} while breathing rates for people over 18 are 12.5 m^3 d^{-1}. These two values are 1.0 and 0.4 m^3 d^{-1} different, respectively, from the average breathing rate.

A second estimate of the level of accuracy in the breathing rate data comes from comparing against other available published values. Average breathing rates reported in alternative sources (units: m^3 d^{-1}) include 12 (US EPA, 1997), 15 (Marty et al., 2002), and 17 (OEHHA, 1996). However, there are two aspects of the analysis yielding the value of 17 m^3 d^{-1} which are incorrect. First, it is obtained as the product of the reported adult average body weight (63 kg) and the reported lifetime-average (i.e., adult plus child) breathing rate per kg body weight (0.271 m^3 kg^{-1} d^{-1}). A more accurate approach would account explicitly for breathing rate differences between children and adults. For example, dividing the population into children ($\leq$ 12 years old) and adults (>12 years old), using values reported by OEHHA (1996) for children (body weight = 18 kg; breathing rate per kg body weight = 0.452 m^3 kg^{-1} d^{-1}) and adults (body weight = 63 kg; breathing rate per kg body weight = 0.232 m^3 kg^{-1} d^{-1}), and assuming a lifetime of 70 y (i.e., the population is 17% children, 83% adults) yields a population average breathing

rate of 13.5 m^3 d^{-1}. This value is significantly closer than 17 m^3 d^{-1} to the value of 12.2

reported by Layton (1993) and used in this chapter. The second error in the OEHHA

(1996) analysis is that the reported body weight values significantly underestimate

current body weights. Results from the Centers for Disease Control's (CDC's) National

Health and Nutrition Examination Survey during 1999 – 2002 indicate that the population

average body weight is 28 kg, not 18 kg, for children (< 12 years old), and 79 kg, not 63

kg, for adults (> 12 years old) (Ogden et al., 2004). Incorporating these updated body

weight values yields a population-average breathing rate 17.4 m^3 d^{-1}, a value that is close

to the reported value of 17.1 m^3 d^{-1}. However, based on the two errors identified here and

the lack of consistency between the values in OEHHA (1996) and those reported

elsewhere (Layton, 1993; Marty et al., 2002; US EPA, 1997), I would assign a low

confidence level to the value of 17.1 m^3 d^{-1}, and would not consider this value to be a

useful indicator of uncertainty in the estimate of 12.2 m^3 d^{-1} from Layton (1993).

Based on the within-population variability reported by Layton (1993) and

variability among alternative data sources, I estimate here that the population-weighted

breathing rate is approximately 12.2 +2/-1 m^3 d^{-1}.

Combined uncertainty. Combining the uncertainties reported above for the four

main calculation inputs (emissions, concentrations, population, breathing rate) yields the

following estimates for the range of plausible intake fraction results. A lower-bound

estimate for both benzene and CO is determined by considering the case where the

breathing rate is too high by 8%, the census values are too high by 3.3%, the

concentrations are correct, and the emission inventory is too low by 20%. In this case, the

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actual iF for benzene and CO would be 28% lower than my results. An upper-bound estimate for the benzene iF is determined by considering the case where the breathing rate is too low by 16%, the census values are too low by 3.3%, the benzene concentrations are too low by 11%, and the emission inventory is correct. In this case, the correct benzene iF result would be 26% higher than my result. Finally, an upper-bound estimate for the CO iF is determined by considering the case where the breathing rate is too low by 16%, the census values are too low by 3.3%, the CO concentrations are correct, and the emission inventory is correct. In this case, the correct CO result would be 12% higher than my results. These values are likely to over-estimate uncertainty because they are derived by assuming that the errors line up at maximum possible amounts. Based on the values reported above (lower-bound uncertainty of 28%; upper-bound uncertainty of 26% and 12% for benzene and CO, respectively), I conclude that my results are accurate to approximately $\pm$ 33% or better, and that the CO results are somewhat more certain than the benzene results because of greater accuracy in the ambient concentration data and because spatial and temporal coverage of the data is more sparse for benzene than for CO.

Sensitivity analysis

The above estimate of 33% uncertainty includes the four factors that most strongly influence the results. However, it does not include the uncertainty in several inputs, such as time-activity pattern data and concentrations in microenvironments, because the level of uncertainty associated with these data has not been quantified. Among the data for which uncertainty has not been quantified above, the most important

values are the concentration and time spent in vehicles, the concentration inside

residences with an attached garage, and the population density around the air basin

throughout the day. A crude sensitivity analysis indicates the potential importance of

uncertainty in these variables. The results of this sensitivity analysis are shown in Table

4. For example, changing the in-vehicle concentration by 20% yields a 6% change in the

intake fraction results. Doubling the population density at any one specific monitoring

station changes the intake fraction results by between –2% and 8%. Note that one station

(Station #60371301, located at 11220 Long Beach Boulevard in Lynwood, California)

has both the highest population density and the highest annual average concentration.

Changes to the population density for this station have a larger impact on the intake

fraction than changes to other stations.

Methodological uncertainties

The estimate of 33% uncertainty based on primary data inputs does not include

the potential for methodological errors, which can be especially difficult to quantify in a

rigorous manner. For example, I use census data on population density to population-

weight the ambient concentration measurements. These data account for where people

live, but not where they travel during the day (i.e., downtown to shop or work). The issue

of how mobility influences the estimated population inhalation intake of air pollution is

addressed in Chapter 7. A second example of a potential methodological error in this

work is how I implemented the NHAPS data on time spent in vehicles. Specific data on

time spent "in an automobile" rather than simply "in a vehicle" (which includes trains

and airplanes) is not currently available in NHAPS. By including time spent in trains and

airplanes with time spent in a motor vehicle, I overpredict the average exposure to motor vehicle exhaust. However, because most travel time in the South Coast is spent in private motor vehicles, the magnitude of this error is likely to be small, and unlikely to significantly impact my results. A third example of a potential methodological error is the use of data from monitoring stations to represent ambient concentrations of benzene and CO. Monitoring stations offer the most comprehensive ambient concentration data set available. Nevertheless, the methodology used in this report, which estimates exposure concentrations based on monitoring station data, may under- or overestimate exposures. A limited number of monitoring stations might not accurately capture the true average ambient concentration, either because there are not enough monitoring stations or because they are not well situated throughout the air basin.

The close agreement in the calculated intake fractions for benzene and CO indicates that certain components of the underlying data are likely to be reasonably accurate. In addition, the agreement between the results presented in this work and in other works provides general support for the broad accuracy of the results reported here. However, the close agreement in the benzene and CO results does not intrinsically lend support to calculations that are applied to both chemicals. For example, if the population size I use is incorrect, it will have the same effect on both the benzene and the CO results. EMFAC-estimated emissions may be similarly biased for benzene as for CO. For example, if high-emitting vehicles (i.e., "gross polluters") are underrepresented in the EMFAC-modeled vehicle fleet, then EMFAC could underestimate emissions for both pollutants.

Conclusions

Intake fraction (iF) characterizes the fraction of a pollutant's emissions inhaled by people. The iF presented in this chapter is for population exposure to atmospheric emissions from motor vehicle emissions in the South Coast Air Basin (SoCAB). However, iF can also be applied to individuals or subpopulations, and it can involve a multi-pathway, multi-media exposure assessment. Based on population-weighted ambient concentrations of benzene and CO, time-activity pattern data, a microenvironment analysis, and population average breathing rates, I calculated an iF of 48 per million. This value means that 0.0048% of SoCAB nonreactive emissions for motor vehicles are inhaled. The results for CO and benzene are consistent with each other (within 5%) and with previous intake fraction studies, thereby lending support to the results and to the intake fraction approach.

Table 3-1: Summary of ambient concentration data

	Carbon monoxide	Benzene
Number of data points	623,534	518
Percent non-detects	5%	6%
Precision	0.1 ppm	0.1 ppb
Detection limit	0.1 ppm	0.2 - 0.5 ppb
Average value	1.20 ppm	1.29 ppb

Table 3-2: Inhalation of various motor vehicle emissions[1], South Coast Air Basin, 1996 – 1999

		1,3-butadiene	acetaldehyde	benzene	formaldehyde	styrene	acrolein
SoCAB emissions from motor vehicles[2]	t y^{-1}	1067	1235	5482	3963	291	8
Lifetime[3]	h	6	39	490	12	23	17
Reaction rate constant[4]	d^{-1}	4.1	0.6	0.05	2.1	1.0	1.4
Reactivity correction[5]	-	28 - 47 %	71 - 85 %	97 - 99 %	43 - 63 %	60 - 78 %	52 - 72 %
iF for this pollutant	per million	13 - 22	34 - 41	46 - 47	20 - 30	28 - 37	25 - 34
Population intake	kg y^{-1}	14 - 24	42 - 50	253 - 257	80 - 119	8 - 11	0.2 - 0.3

Notes:

(1) These results are for inhalation of primary motor vehicle emissions in the South Coast Air Basin. Degradation of primary emissions is included, but secondary formation is not.

(2) Source: California 1996 Toxics Emission Inventory (http://www.arb.ca.gov/toxics/cti/cti.htm). Motor vehicle emissions in SoCAB are 42% of California motor vehicle emissions (Singer and Harley, 2000).

(3) Source for styrene and acrolein: CARB Toxic Air Contaminant Fact Sheets (http://www.arb.ca.gov/toxics/tac/toctbl.htm). Source for remaining compounds: EPA 1993 Motor Vehicle-Related Air Toxics Study (http://www.epa.gov/otaq/toxics.htm).

(4) The reaction rate constant (k) is the reciprocal of the lifetime.

(5) The reactivity correction, which is derived in Appendix I for this chapter, is multiplied by the intake fraction for a conserved pollutant to account for the presence of removal mechanisms other than advection. The intake fraction for a conserved pollutant is taken as 47.5 per million, which is the average of the results for benzene and carbon monoxide presented in the report. The range of values for the reactivity correction in this table are based on high and low values for the residence time of air in the air basin. The residence time of air in SoCAB is estimated in Appendix I for this chapter as between 7 and 16 hours.

Table 3-3: Simplified intake fraction analysis

		Carbon Monoxide	Benzene
Concentration	ppm	1.20	0.00129
Concentration	$\mu g \ m^{-3}$	1410	4.22
Ambient concentration attributable to motor vehicles	-	80%	70%
Breathing rate	$m^3 \ d^{-1}$	12.2	12.2
Population	people	1.5×10^7	1.5×10^7
Intake	$g \ month^{-1}$	6.3×10^6	1.6×10^4
Emissions	$g \ month^{-1}$	2.0×10^{11}	5.0×10^8
Intake fraction	per million	32	33

Table 3-4: Sensitivity analysis

Parameter	Change in parameter	Change in intake fraction
Concentration in vehicles	+ 20%	+ 6%
	- 20%	- 6%
	+ 100%	+ 30%
	- 100%	- 30%
Time spent in vehicles	+ 20%	+ 6%
	- 20%	- 6%
	+ 100%	+ 30%
	- 100%	- 30%
Concentration in residences with an attached garage	+ 20%	+ 8%
	- 20%	- 8%
	+ 100%	+ 40%
	- 100%	- 40%
Population density at each specific monitoring station	+ 100%	- 2% to + 8%
	- 100%	- 10% to + 2%
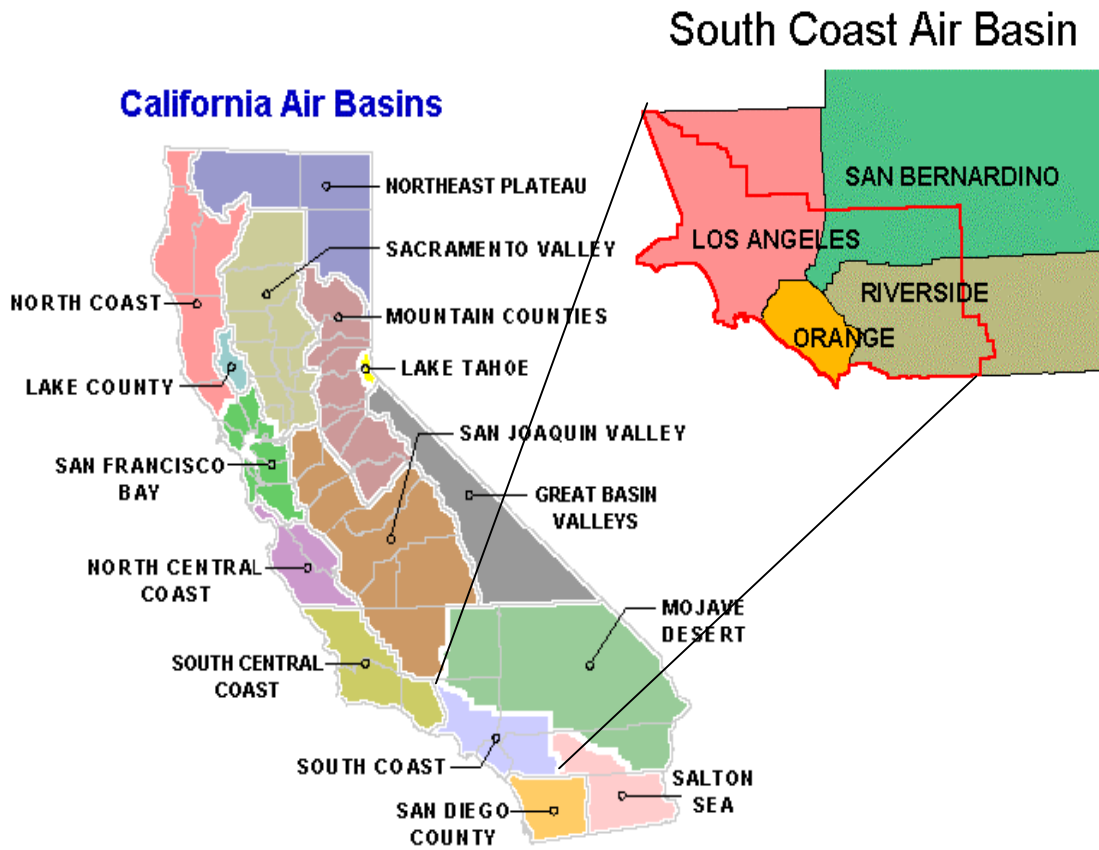
Figure 3-1: California's 15 air basins, highlighting the South Coast Air Basin counties.

From http://www.arb.ca.gov/emisinv/maps/ statemap/abmap.htm.
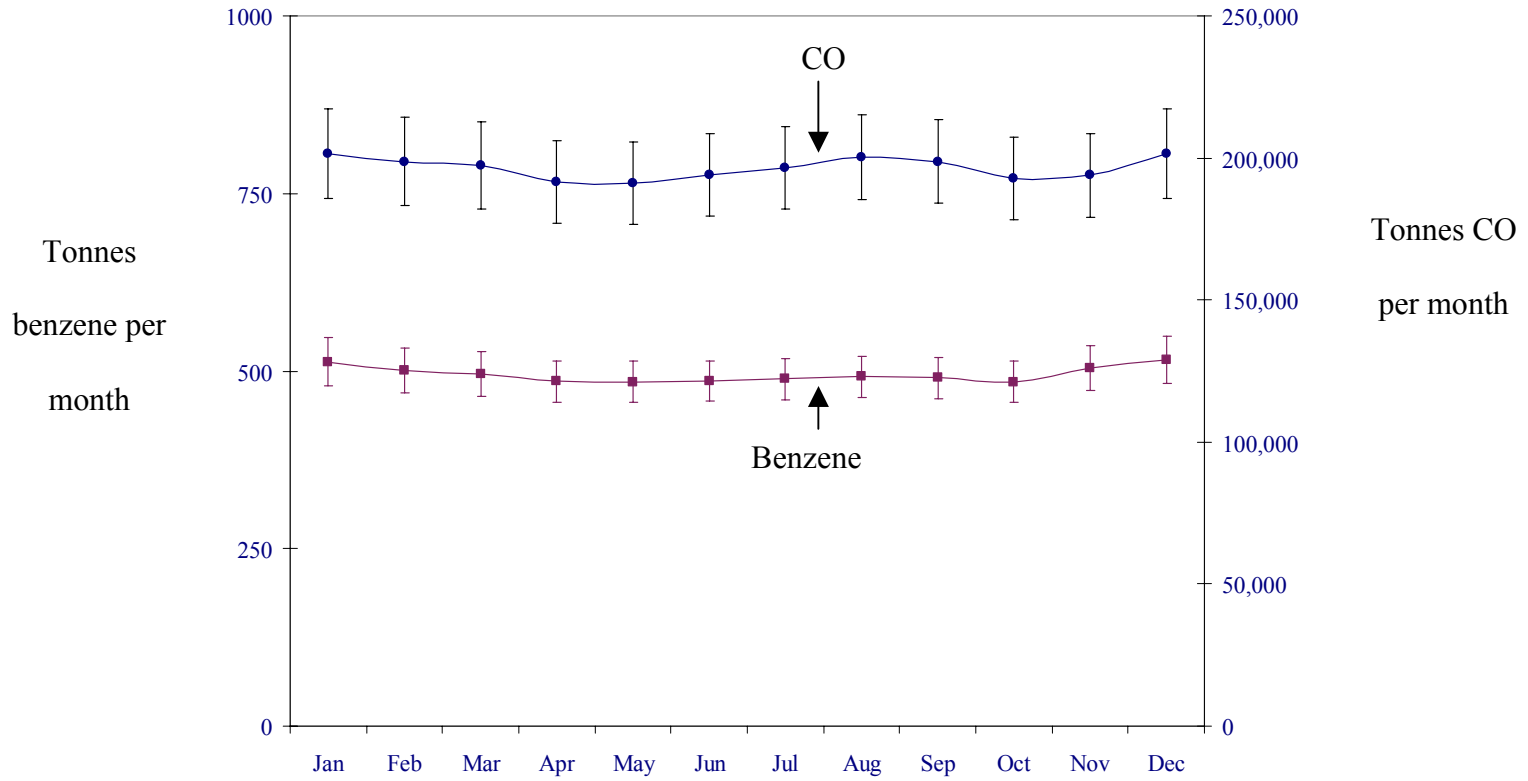
Figure 3-2: Motor vehicle emissions in the SoCAB 1996 – 1999.
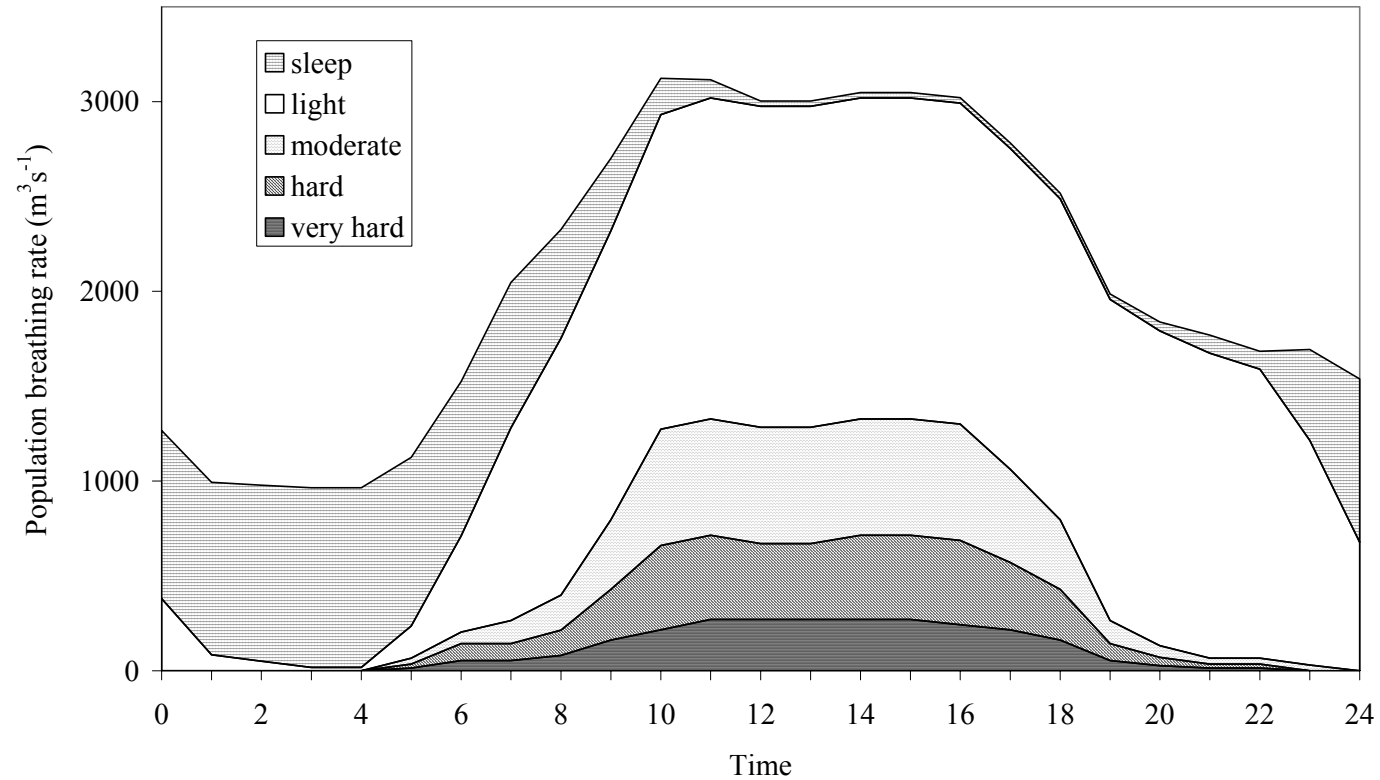
Figure 3-3: Hourly breathing rate by time of day and activity intensity. Layton (1993) gives breathing rates for five activity levels (sleep, light, moderate, hard, very hard) and the total number of hours spent in each of those activities. This figure shows how these rates and hours were divided into the hours of the day.
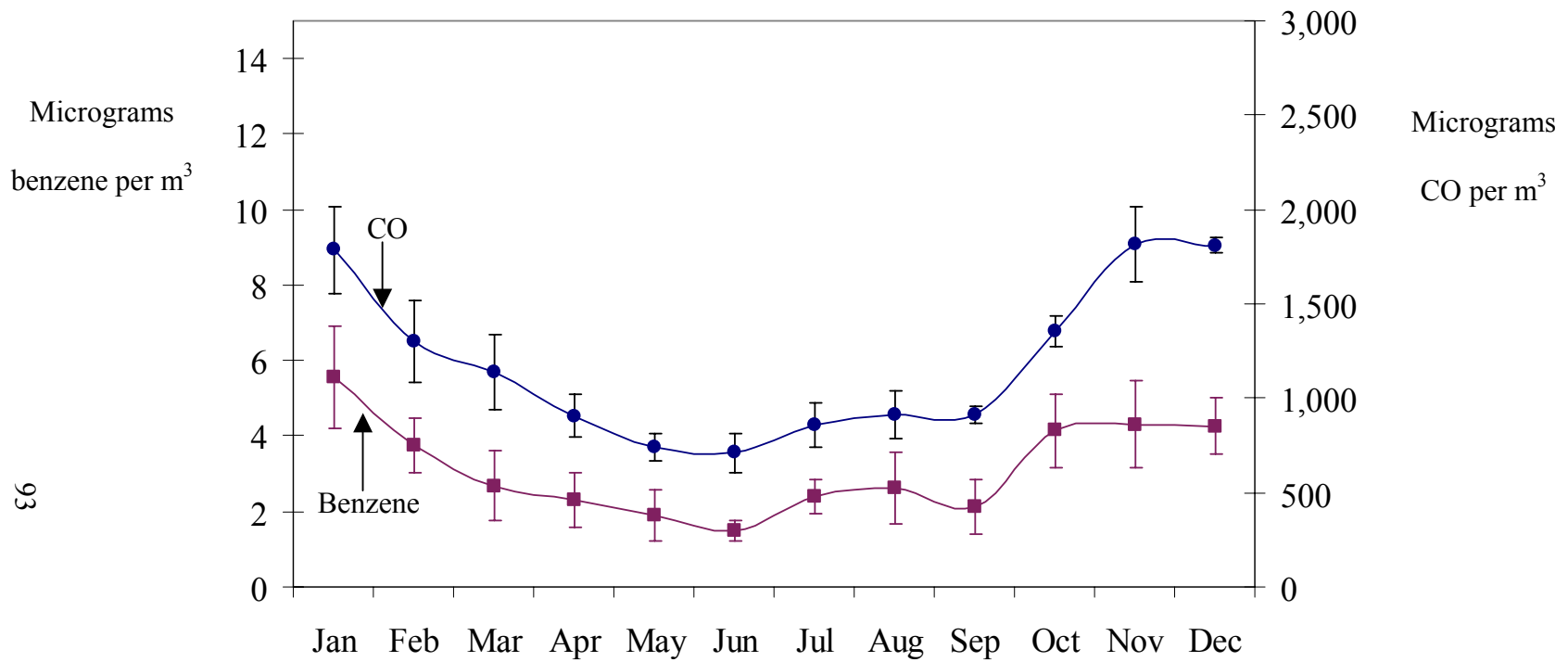
Figure 3-4a: Monthly average ambient concentration attributable to motor vehicles in the SoCAB 1996 – 1999. The ambient

concentration attributable to motor vehicles, which is based on ambient air monitoring station data, shows a "U-shaped" profile due to

the predominant meteorology. Summer conditions tend to disperse primary pollutants more efficiently than winter conditions.
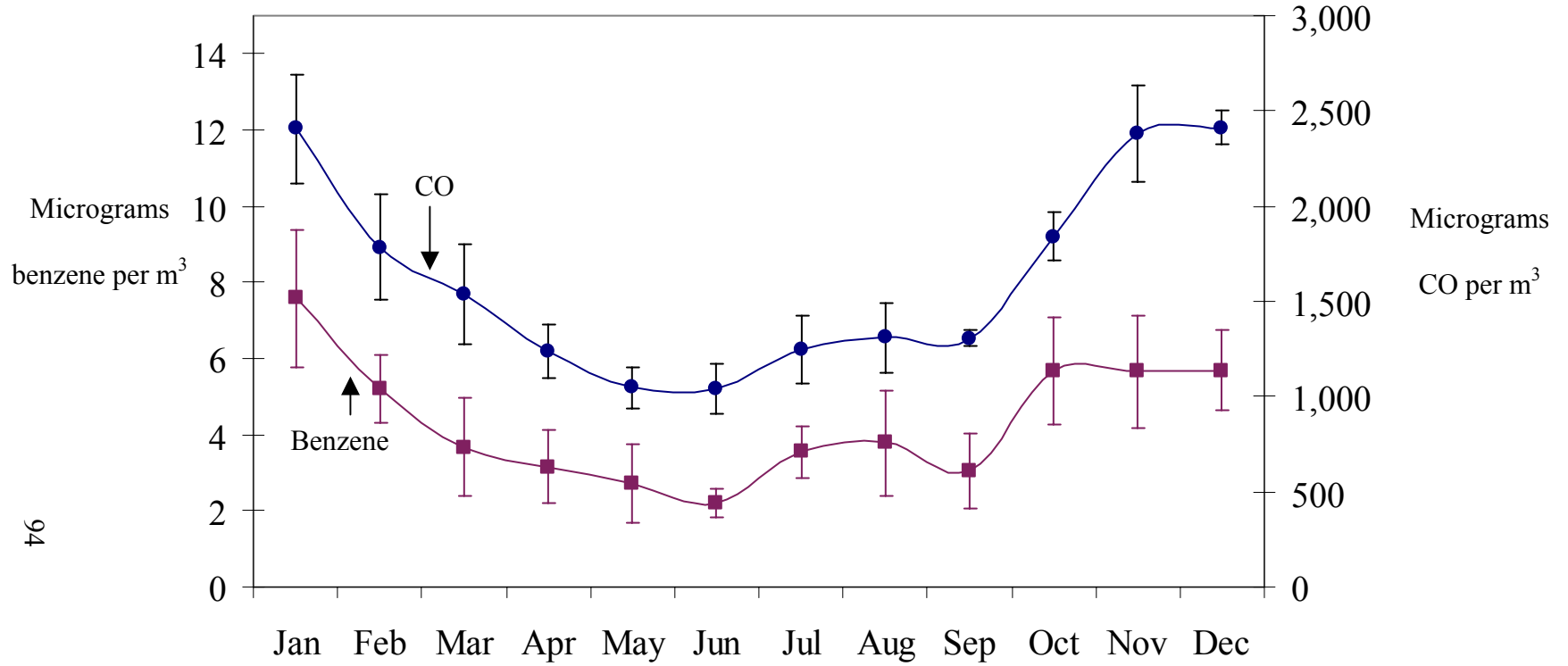
Figure 3-4b: Exposure concentration attributable to motor vehicles in the SoCAB 1996 - 1999. Exposure concentration to benzene and CO attributable to motor vehicles shows the same pattern as the ambient concentrations in Figure 3-4a.
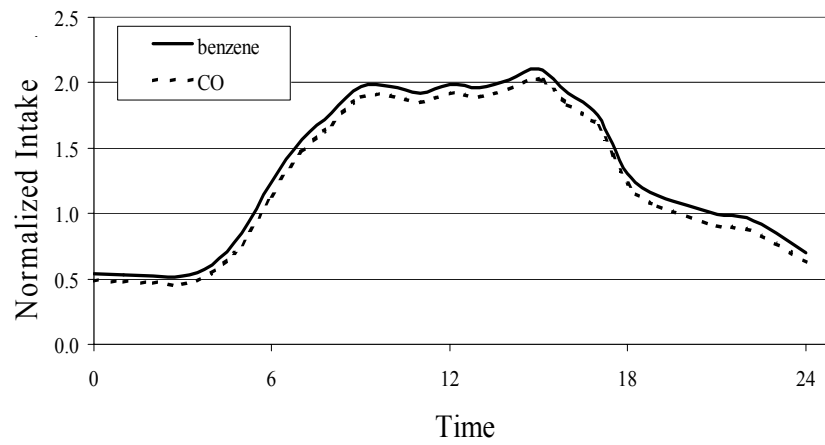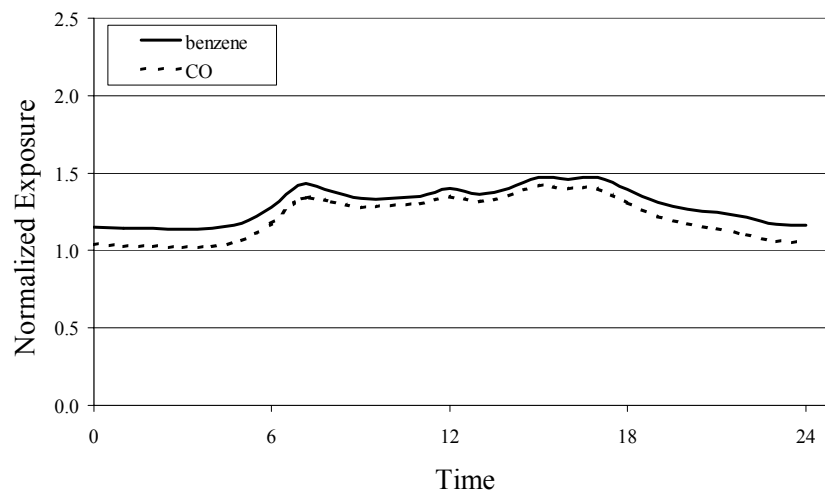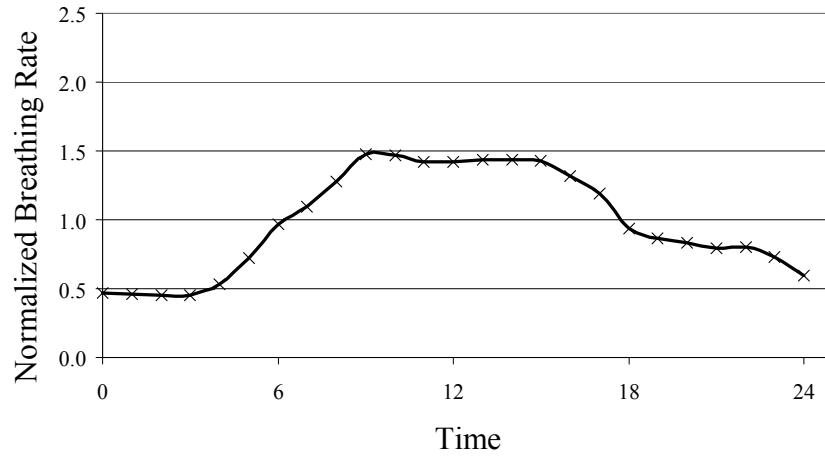
Figure 3-5. (Caption is on next page.)

Figure 3-5: Hourly variability in breathing rate, exposure, and intake. Normalized breathing rate is the hourly breathing rate divided by the average breathing rate. Normalized exposure concentration is the exposure concentration divided by the ambient concentration. Normalized intake, which is the product of the normalized breathing rate and the normalized exposure concentration, indicates the increase in the true intake, as compared to the simplified analysis, owing to the combined influence of microenvironments and breathing rates. This figure shows that the population-average exposure concentration is greater than the ambient concentration at all times. Normalized benzene and CO intake is greater than one during the daytime, indicating that during the daytime, population intake is greater than ambient concentration times the average breathing rate. At night, population intake is less than ambient concentration times average breathing rate.
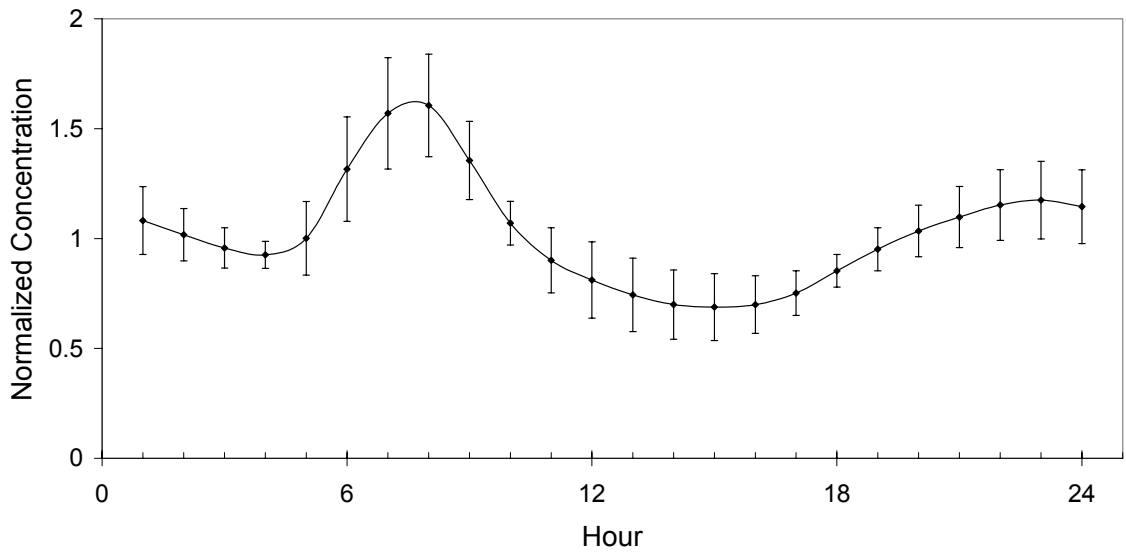
Figure 3-6: Typical ambient daily carbon monoxide concentration profile. Normalized concentration is the average ambient concentration in each hour divided by the overall average ambient concentration. Concentrations are highest during the morning commute, when emissions are high and dispersion is slow.
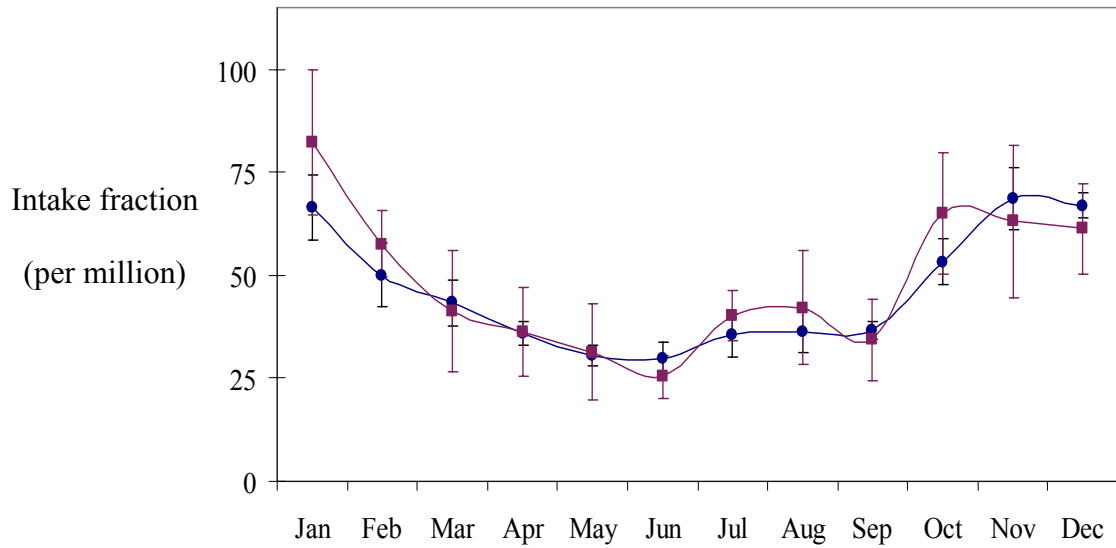
Figure 3-7: Monthly average intake fraction for nonreactive motor vehicle emissions in the SoCAB 1996 – 1999. Intake fractions for benzene and CO show the same pattern seen in Figures 3-4a and 3-4b. Consistency between the values for benzene and CO lends support to the intake fraction concept.

Appendix I: One-compartment model for estimating intake fraction

Here I use a one-compartment model to estimate the intake fraction for a conserved and for a reactive compound. This approach indicates the influence of various factors on the calculated intake fraction. In a one-compartment model, the air in the basin is assumed to be well mixed and pollutant concentrations are assumed to be at steady state (Figure 3A-1).

The total population intake rate, I (g s^{-1}), that is attributable to an emission source is

$$I = CQ_BP, \hspace{4cm} 3A\text{-}1$$

where C is the incremental concentration (g/m^3) attributable to the emission source, Q_B is the population average breathing rate per person (m^3 person^{-1} s^{-1}), and P is the population. The steady-state mass balance for a one-compartment model is given in Equation 3A-2:

$$E = kCV + QC, \hspace{4cm} 3A\text{-}2$$

where E is the emission rate (g/s), k is the first-order reaction rate constant (s^{-1}), V is the volume (m^3) of the compartment (i.e., the air basin), and Q is the flow of air through the compartment (m^3 s^{-1}). For a conserved pollutant, k=0. The first term in Equation 3A-2 (kCV) is the loss rate owing to reaction, and the second term (QC) is the removal rate by

advection. For a square-plan, one-compartment model of an urban area, the flow of air through the compartment may be estimated as $uHA^{0.5}$, where u is the wind speed (m s^{-1}), H is the atmospheric mixing height (m), and A is the urban land area (m^2). Intake fraction (iF; unitless) is the total attributable intake rate divided by the total attributable emission rate. This ratio may be expressed incorporating Equations 3A-1 and 3A-2 as

$$ iF = \frac{CQ_BP}{E} = \frac{Q_BP}{kV+Q} = \frac{Q_B\rho\tau}{H(k\tau+1)}. \qquad \text{3A-3} $$

Here, ρ is the population density (m^{-2}), which is equal to P/A, and τ is the residence time (s) of air in the air basin, which may be expressed as $V\,Q^{-1}$ or estimated as $A^{0.5}\,u^{-1}$.

For a relatively nonreactive compound, $k\tau \ll 1$. In this case, Equation 3A-3 reduces to iF $\sim Q_B\rho\tau/H$. For a highly reactive compound, $k\tau \gg 1$. In this case, Equation 3A-3 reduces to iF $\sim (Q_B\rho) / (Hk)$. A compound is defined as being moderately reactive when $\tau \sim (1/k)$, i.e., when the reciprocal of the reaction rate constant is similar in magnitude to the residence time. In Equation 3A-3, the term $(k\tau+1)^{-1}$ stands out as a "reactivity correction term." If the intake fraction is known for a nonreactive compound, multiplying by this term will yield an intake fraction estimate that accounts for the reactivity of a specific compound. This approach is used in Table 3-2.

Equation 3A-3 can be rewritten as

$$ \text{Intake Fraction} = \frac{Q_B\rho}{H}T \qquad \text{(3A-4)} $$

where $T = (\tau)/(k\tau+1)$ is the characteristic time for the pollutant to be removed from the system. For a nonreactive compound, the characteristic time simplifies to $T = \tau$. For a highly reactive compound, the characteristic time becomes $T = k^{-1}$.

Table 3A-1 first uses iF = $(Q_B P) / (uHA^{0.5})$ to calculate the intake fraction for a conserved pollutant, and then applies the "reactivity correction term" $(k\tau+1)^{-1}$ to predict the intake fraction for a reactive pollutant with k^{-1} = 80 hours. For the values in this table, the reactivity term varies from 18% to 92%, meaning that the intake fraction for the reactive pollutant is 18 – 92% of the intake fraction for a conserved pollutant.

The first entry for the SoCAB columns in Table 3A-1 represents the product of the wind speed and mixing height. The values used in Table 3A-1 (195 – 1,300 m^2 s^{-1}) are the 5% and 95% values from the distribution of harmonic means for the 75 cities in the EPA's SCRAM database of mixing heights (US EPA, 2002). The mixing heights for the downwind regional intake (600 – 6,000 m) represent mixing occurring throughout 5% – 50% of the troposphere, and the mixing heights for the US intake (3,000 m – 12,000 m) represent the assumption of mixing throughout 25% – 100% of the troposphere. The wind speeds (2.36 – 5.50 m s^{-1}) represent the 5% and 95% distribution of harmonic means for the 75 cities in the EPA's SCRAM database of wind speeds. These wind speeds are the average over the mixing height rather than being the surface wind speed. As is discussed in the text, the reaction rate (k^{-1} = 80 hours) represents a compound that is more reactive than benzene in the summer.

The iF for a conserved pollutant in the SoCAB is estimated to be in the range 12 – 82 per million using the box model. This range includes the main results of ~48 per

million for benzene and CO, and the results presented in Table 3 of ~33 per million from

the simplified analysis. Consistency between the box model and these two results lends

support to the validity of the box model approach for approximating the intake fraction

for broadly distributed emissions of nonreactive pollutants into outdoor urban air.

Table 3A-1: Intake fractions using a box model

		SoCAB		Downwind Regional		Nationwide	
H	m			600	6,000	3,000	12,000
u	m s^{-1}			2.36	5.50	2.36	5.50
Hu	m^2 s^{-1}	195	1,300	1,416	33,000	7,080	66,000
A	m^2	1.7×10^{10}	1.7×10^{10}	8.4×10^{10}	8.4×10^{10}	9.2×10^{12}	9.2×10^{12}
$A^{0.5}$	m	1.3×10^{5}	1.3×10^{5}	2.9×10^{5}	2.9×10^{5}	3.0×10^{6}	3.0×10^{6}
P	people	1.5×10^{7}	1.5×10^{7}	1.3×10^{6}	1.3×10^{6}	2.81×10^{8}	2.81×10^{8}
Q_B	m^3 d^{-1} person^{-1}	12.2	12.2	12.2	12.2	12.2	12.2
PQ_B	m^3 s^{-1}	2.1×10^{3}	2.1×10^{3}	1.8×10^{2}	1.8×10^{2}	4.0×10^{4}	4.0×10^{4}
$UHA^{0.5}$	m^3 s^{-1}	2.6×10^{7}	1.7×10^{8}	4.1×10^{8}	9.6×10^{9}	2.1×10^{10}	2.0×10^{11}
iF[a]	-	8.2×10^{-5}	1.2×10^{-5}	4.5×10^{-7}	1.9×10^{-8}	1.9×10^{-6}	2.0×10^{-7}
iF[a]	**per million**	**82**	**12**	**0.45**	**0.019**	**1.85**	**0.20**
k^{-1}	h	80	80	80	80	80	80
k	s^{-1}	3.5×10^{-6}	3.5×10^{-6}	3.5×10^{-6}	3.5×10^{-6}	3.5×10^{-6}	3.5×10^{-6}
u	m s^{-1}	2.36	5.50	2.36	5.50	2.36	5.50
T	h	16	7	34	15	356	153
$(kt+1)^{-1}$	-	84%	92%	70%	85%	18%	34%
iF[b]	**per million**	**69**	**11**	**0.31**	**0.016**	**0.34**	**0.07**

(a) Intake fraction for a nonreactive compound
(b) Intake fraction for a reactive compound with $k^{-1} = 80$ h.

Appendix II: Intake fraction results for the South Coast by month

Table 3A-2, presented next, summarizes the estimated intake fractions in each month during 1996–1999 (inclusive), for carbon monoxide and benzene.

Table 3A-2: Intake fraction results for the South Coast by month

		Intake Fraction (per million)	
Year	Month	Carbon monoxide	Benzene
1996	1	68.4	98.4
1996	2	48.2	58.2
1996	3	42.8	29.3
1996	4	37.5	36.1
1996	5	28.0	22.3
1996	6	31.0	24.3
1996	7	34.3	42.0
1996	8	35.7	57.3
1996	9	34.1	44.0
1996	10	51.0	77.9
1996	11	61.0	39.7
1996	12	62.9	63.2
1997	1	55.2	58.9
1997	2	54.5	68.2
1997	3	51.3	50.2
1997	4	32.4	31.3
1997	5	34.3	48.2
1997	6	24.4	18.7
1997	7	31.3	47.6
1997	8	32.1	36.2
1997	9	37.1	38.0

Table 3A-2 (Cont.)

| Year | Month | Intake Fraction (per million) | |
		Carbon monoxide	Benzene
1997	10	48.1	60.4
1997	11	71.9	78.1
1997	12	66.0	49.0
1998	1	70.0	78.5
1998	2	40.0	48.7
1998	3	38.4	57.7
1998	4	38.8	51.2
1998	5	29.9	25.2
1998	6	31.5	27.5
1998	7	42.8	37.7
1998	8	43.8	49.1
1998	9	36.5	20.8
1998	10	53.0	46.2
1998	11	77.5	57.1
1998	12	69.5	75.4
1999	1	73.0	92.9
1999	2	57.6	55.6
1999	3	41.0	28.4
1999	4	35.3	26.1
1999	5	29.9	29.7
1999	6	32.8	31.1
1999	7	33.2	33.6
1999	8	34.2	26.3
1999	9	39.0	34.7
1999	10	61.3	75.6
1999	11	63.8	77.6
1999	12	69.1	57.9
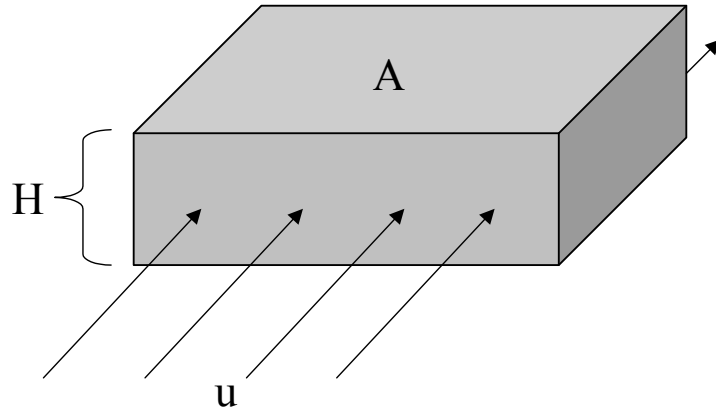
Figure 3A-1: Box model representation of an urban airshed. Shown are wind speed (u), mixing height (H), and urban land area (A).

Chapter 4: Intake fraction of nonreactive vehicle emissions in US urban areas

Introduction

The previous chapter presented a detailed estimate of intake fraction of vehicle emissions in California's South Coast Air Basin. The South Coast is an important case study: it is home to approximately 1 in 19 US residents, and air pollution levels there are among the worst in the US. However, conditions in the South Coast are not typical of US urban areas. This chapter expands on the previous one by considering the intake fraction of vehicle emissions in urban areas throughout the US. Here, I develop and implement methods for efficiently estimating intake fraction for distributed ground-level releases of nonreactive pollutants in urban environments. The methods incorporate three types of models: the straightforward one-compartment model described in Appendix I of Chapter 3 (Benarie, 1980), an empirical model describing measured carbon monoxide concentrations in 14 cities (Glen et al., 1996), and the US EPA's most sophisticated national-scale exposure model (US EPA, 2004).

Methods

Intake fraction is the fraction of emissions that are taken in by people. For inhalation of a primary pollutant, intake fraction is defined in the previous chapter, in Equation 3-1. Three approaches for estimating intake fractions for vehicular emissions in urban air basins are described and applied in the following subsections. In all three cases, the population average breathing rate, Q, is taken as 12.2 m^3 person^{-1} d^{-1}, based on metabolic activity studies (Layton, 1993). In this chapter, I estimate intraurban intake fractions, i.e., those associated with urban residents' inhalation of emissions that occurred in the same urban area. This approach represents an important and logical step towards a complete treatment, which would also quantify downwind intakes for urban areas throughout the US. Downwind intakes for the South Coast were estimated in Chapter 3 as significantly smaller than intraurban intakes. This attribute does not hold for all urban areas. The closer spacing of cities in the Eastern US and elsewhere in many parts of the world would yield higher extraurban intake fractions.

One-compartment model

I use a one-compartment model (Benarie, 1980; Lyons et al., 2003) to combine meteorological data on wind speed and mixing heights with demographic data on urban population and land area. This model is often assumed to be too simple to offer reasonable estimates of ambient concentrations in urban areas. It does not offer many of the capabilities of more sophisticated models, such as predicting spatial variability in ambient concentrations. However, for conserved or slowly reacting emissions from

broadly distributed ground-level sources, the one-compartment model may offer a reasonably accurate estimate of spatially averaged concentrations in an urban area. In Chapter 3, I reported that for the South Coast Air Basin, the range of values reported for the long-term basin-wide average intake fraction of benzene and CO, as estimated by the one-compartment model, bracketed the value estimated based on a detailed analysis incorporating measured concentrations and microenvironments.

The one-compartment model assumes that air concentrations are uniform throughout an air basin. To explore the accuracy of this assumption, I analyzed year-2002 annual average CO concentrations at the 497 monitoring stations in the US EPA AIRData database (http://www.epa.gov/air/data). I chose CO because it is nonreactive, because there are a large number of monitoring stations in the US, and because most urban CO emissions are attributable to motor vehicles (US EPA, 2001c). First, I removed from the dataset the 60 monitors that did not meet EPA's reliability criterion (> 75% reporting rate). Then, I removed the 30 monitors that did not have an associated metropolitan statistical area (MSA) code. Among the remaining 407 monitors, 189 (46%) are located in one of the 28 MSAs with five or more monitors. I evaluated intra-MSA variability among these 189 monitors. The coefficient of variability (the standard deviation divided by the mean) for each MSA has a small average value of 0.31 (range: 0.13-0.53). Furthermore, the concentration difference between a monitor and the associated MSA average is always less than 65%. Low intra-MSA variability in annual average ambient CO concentrations suggests that the one-compartment model is useful for estimating the average emissions-to-concentration relationship for primary nonreactive vehicle emissions in urban areas.

Examples of pollutants that are reasonably modeled as conserved when considering ambient concentrations include benzene, carbon monoxide, and primary $PM_{2.5}$. Advection is the dominant removal mechanism for conserved pollutants because the time they take to react or deposit is considerably longer than the residence time of air in an urban basin, which may be estimated as $A^{0.5} u^{-1}$. Here, A is urban land area (m^2) and u is wind speed averaged over the mixing height (m s^{-1}). (This approach assumes that (1) the characteristic length scale for wind traversing the urban area is $\sim A^{0.5}$, and (2) air parcels that exit the urban area do not re-enter. These assumptions are reasonable for the purposes here for urban areas whose 2-dimensional map projection is continuous and in a shape that is simple, such as a square or circle, rather than sinuous or contorted.) The population-weighted average value for $A^{0.5}$ for US urban areas is 49 km (US DOT, 2003a) (the unweighted average for $A^{0.5}$ is 20 km), and the harmonic mean wind speed in the US is 3.4 m s^{-1} (US EPA, 2002), indicating that the characteristic residence time of air in a US urban area is $\sim 2 - 4$ h. The lifetime of many air pollutants is much greater than 4 h (Atkinson, 1994). For example, of the 130 toxic air contaminants that have a half-life listed in the California Air Resources Board's contaminants summary database (www.arb.ca.gov/toxics/tac/txctbl2.htm), 81% have a half-life of more than 10 h.

For a square-plan, one-compartment model, the intake fraction of nonreactive pollutant emissions is calculated using Equation 4-1 (Lai et al., 2000):

$$iF_{compartment} = \frac{QP}{uH\sqrt{A}}.$$
(4-1)

Here, $iF_{compartment}$ is the intake fraction (unitless) estimated by means of the one-compartment model, Q is the population average breathing rate (m^3 person^{-1} s^{-1}), P is the

population, and H is the atmospheric mixing height (m). Equation 4-1 is derived from a mass balance. The main assumptions in the derivation are that air in an urban area is well-mixed, and that either the system is at steady-state or concentrations are not strongly correlated over time with breathing rates. (The impact on exposures of temporal correlations between diurnal breathing rate and diurnal concentrations is explored in Chapter 6.) I assume here that deposition and chemical reactions occur slowly compared to advection, but as is shown in Chapter 3 (Appendix I), it is straightforward to extend the approach to incorporate first-order decay processes.

The variables in Equation 4-1 can be clustered into three parameter groups. The first parameter group (which I term "linear population density"), $P A^{-0.5}$, is an attribute of a city's urban form, i.e., the way in which the urban area is laid out. The second parameter group ("normalized dilution rate"), uH, is an attribute of the meteorology. Normalized dilution rate ($m^2 s^{-1}$) is the volumetric airflow rate out of the basin ($m^3 s^{-1}$) divided by air basin width (m). The final parameter is the population average breathing rate, Q.

Linear population density (people m^{-1}) values are calculated from year-2002 population and land area data for the 379 urban areas in the US with more than 50,000 people (US DOT, 2003a). These 379 urban areas contain 63% of the US population. Harmonic mean normalized dilution rates ($m^2 s^{-1}$) are calculated from twice-daily derived values of wind speeds and mixing heights for the 75 meteorological stations in the US EPA's Support Center for Regulatory Air Models (SCRAM) database (US EPA, 2000; US EPA, 2002). Wind speeds in this database are the average speed over the mixing height (US EPA, 2002). The mean and median values of the meteorological stations' harmonic mean normalized dilution rate, uH (units: $m^2 s^{-1}$), are 610 and 480, respectively. The linear

population density and normalized dilution rate data are presented in Figure 4-1. In Figure 4-2, the linear population density data is compared against population in that urban area.

Using Equation 4-1, I combine each of the 379 linear population densities with each of the 75 annual harmonic mean normalized dilution rates, yielding 28,425 estimates of intake fraction. This method of combining the two datasets implicitly assumes that the SCRAM data are representative of meteorological conditions throughout the US and that the DOT data are representative of urban areas throughout the US. Visual inspection of a US map showing the locations of the 75 meteorological stations and the 379 cities did not reveal a systematic location bias.

As a comparison with Figures 4-1 through 4-5, which apply to US urban areas, Table 4-1 presents linear population density and intake fraction values for select urban areas in California. Intake fraction values in Table 4-1 are based on Equation 4-1, using a breathing rate of 12.2 m^3 d^{-1} $person^{-1}$ and a normalized dilution rate of 480 m^2 s^{-1} (i.e., 41.5 million m^2 d^{-1}). Linear population density, and thus estimated intake fraction, varies among California urban areas by more than an order of magnitude.

Empirical model

The empirical model developed by Glen et al. (1996) estimates ambient concentrations of carbon monoxide (CO), which is a good tracer for nonreactive vehicle emissions. As a statistical model based on measured concentrations, this approach offers a good complement to the two other methods presented in this chapter. The empirical model focuses explicitly on vehicle emissions, incorporating US EPA's MOBILE5 emission

factors (www.epa.gov/otaq/m5.htm). The model offers good predictions of observed data, based on only a few empirically determined parameters.

For the years 1984 – 1991, Glen et al. (1996) compared monthly average ambient concentrations of CO in 15 US cities with meteorological data and MOBILE5 emission factors. They report the following empirical relationship:

$$C_{i,n} = k_n E_{i,n} \exp\left\{-\frac{H_{i,n}}{h^*} - \frac{u_{i,n}}{u^*}\right\} \qquad (4\text{-}2)$$

Here, $C_{i,n}$ is the modeled ambient CO mole fraction (ppm) in month i for city n; k_n is an empirically determined constant (ppm mile g^{-1}) for city n; $E_{i,n}$ is the average CO emission factor (g mile^{-1}) in month i for city n; $H_{i,n}$ and $u_{i,n}$ are the average mixing height (m) and wind speed (m s^{-1}), respectively, in month i for city n; and h^* and u^* are empirically-determined constants with units of length (m) and speed (m s^{-1}), respectively, used to make dimensionless the argument in the exponential. They report one value for h^* (1626 m) and for u^* (9.55 m s^{-1}) and the following information for each city: k_n, mean summer and winter wind speed and mixing height, and modeled and measured CO concentration time series.

For each of the 15 cities analyzed by Glen et al., I calculate the winter and summer intake fraction using Equation 4-3, which is derived from Equations 3-1 and 4-2.

$$\text{iF}_{\text{empirical}} = \left(\frac{QP}{D_{VMT}} \phi k_n \exp\left\{-\frac{H_{i,n}}{h^*} - \frac{u_{i,n}}{u^*}\right\}\right)\left(\frac{0.00125 \text{ g}}{\text{m}^3 \text{ ppm}}\right). \qquad (4\text{-}3)$$

Here, D_{VMT} is the total daily vehicle-miles travelled (VMT) in an urban area (mile d^{-1}), ϕ is the fraction of ambient concentrations attributable to motor vehicle emissions (unitless), and 0.00125 converts the CO mole fraction (ppm) to CO concentration (g m^{-3}). For the

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years 1984 – 1991, ϕ is ~0.7 based on US nationwide data (US EPA, 2003c). Population

and VMT data are taken from US DOT (2003a).

National-Scale Air Toxics Assessment (NATA)

The National-Scale Air Toxics Assessment (NATA) estimates year-1996 population

inhalation of atmospheric emissions in the US (US EPA, 2004). To my knowledge, NATA

is the most comprehensive national-scale exposure model available. Two main steps within

NATA are important here. First, the ASPEN Gaussian plume dispersion model uses

meteorological data and the year-1996 National Toxics Inventory to estimate ambient

concentrations in all US census tracts. Next, a probabilistic exposure model combines (1)

ASPEN-estimated ambient concentrations, (2) time-activity information for 30 hypothetical

individuals from each of 10 cohorts (5 age groups, two genders) and (3) estimates of

differences between ambient and microenvironment exposure concentrations. The results

are summarized as the population average incremental exposure concentration attributable

to four source categories (point, area, on-road mobile, and off-road mobile) in two county

types (urban and rural). I estimate NATA intake fractions for urban on-road mobile sources

based on two conserved pollutants: benzene and diesel particulate matter.

Intake fraction is estimated from the NATA values using Equation 4-4:

$$iF_{NATA} \quad = \quad \frac{CQP}{E} \tag{4-4}$$

Here, C is the mean urban attributable exposure concentration (g m^{-3}), and E is the

emission rate from on-road mobile sources (g h^{-1}). Consistent with the EPA's caveat that

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NATA results are more meaningful when aggregated rather than presented for individual counties, I present here the mean intake fraction among US urban counties. Because the NATA exposure concentrations are the mean values across census tracts, they are approximately population-weighted values, as US Census tracts are sized to contain ~4,000 people each (US Census, 2004b).

Results and discussion

Intake fraction values

Intake fraction values vary among urban areas. The first two methods I use (the one-compartment model and the empirical model) provide information about this variability. Table 4-2 presents population-weighted and unweighted intake fraction results. The unweighted mean, for example, is the mean value of the intake fraction among urban areas (i.e., giving equal weighting to each urban area). These values are applicable when considering each US urban area as a distinct unit. The population-weighted mean weights the intake fraction value for each urban area based on urban population (i.e., giving equal weight to each person). These values are applicable for population-weighted measures including total US urban environments.

Unprocessed intake fraction results from the one-compartment model are presented in Figure 4-3. Figure 4-4 presents isopleths of one-compartment-model-derived intake fraction values as a function of linear population density ($PA^{-0.5}$) and normalized dilution rate (uH). At a given pair of percentiles, intake fraction is larger for the population-weighted values than for the unweighted values. Figure 4-5 is a bubble plot of results from

the empirical model, with the icon area proportional to the intake fraction. Each of the 15 cities in the empirical model is represented by two icons (summer and winter).

Comparisons among the three methods

Intake fraction values calculated by the three methods employed in this chapter are consistent with each other. The range of intake fraction values (units: per million) is broader for the one-compartment model (0.1 – 280) than for the empirical approach (5.7 – 54). One reason for this difference is that the one-compartment approach considers significantly more urban areas than the empirical approach (379 versus 15 urban areas). In addition, as applied here, the one-compartment model estimates values for all possible combinations of linear population density ($PA^{-0.5}$) and normalized dilution rate (uH), rather than incorporating only the one, true set of meteorological conditions found in any urban area. Hence, it is more appropriate to consider the one-compartment model results presented in this chapter in terms of central tendencies (e.g., median and inter-quartile range) rather than for extreme values (e.g., maximum and minimum).

Unweighted intake fraction values for the empirical model are larger than for the one-compartment model mainly because of differences in urban population size. The mean urban population is 5.5 times larger for the 15 cities in the empirical model than for the 379 urban areas in the one-compartment model (1.2 million versus 220,000 people). The best-fit relationship in Figure 4-2 indicates that linear population density is proportional to $P^{0.59}$. Based on this relationship, the population difference of 1.2 million versus 220,000 would yield a factor of 2.7 difference in the linear population density values, and thus, in the expected mean unweighted intake fraction. Consistent with this expectation, the difference

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in unweighted mean intake fraction between the empirical model (15 per million, summer and winter combined) and the one-compartment model (5.3 per million) is a factor of 2.8.

For comparison, I also calculated the NATA-derived mean urban intake fraction value for a reactive vehicle emission, 1,3-butadiene (characteristic lifetime ≈ 6 h (US EPA, 1993)). The result, 3.1 per million, is less than the NATA-derived intake fraction for benzene (7.0 per million) because chemical reactions remove a portion of the 1,3-butadiene from ambient air.

Results presented in this chapter, like those presented in Chapter 3, support the idea that a one-compartment model can yield reasonably accurate results for investigations of typical intake fraction values. NATA accounts for several factors that the one-compartment model does not, such as concentration differences in microenvironments and spatial heterogeneities in emissions and ambient concentrations. The more sophisticated approach employed by NATA allows it to address questions the one-compartment model cannot. Nevertheless, the two approaches yield similar results for the primary research objective of this chapter.

Intake fraction in urban areas not studied in this work

One method for estimating the intake fraction in a specific urban area would be to scale the results from this chapter up or down based on the linear population density. For example, based on the one-compartment model results, when considering an urban area with a linear population density that is two times greater than the US DOT (2003a) median value of 9.5 people m^{-1}, the intake fraction would be estimated as ~6 per million (i.e., two times greater than the unweighted one-compartment model median intake fraction value of

3.0 per million). For the South Coast Air Basin (linear population density $\approx$ 120 people m^{-1}), this approach suggests a value of 38 per million, which is close to the value of 48 per million presented in Chapter 3. If the linear population density is not known, the intake fraction for an urban area can be approximated from the urban population (P) using the following relationship: intake fraction $\approx$ 0.0025 $P^{0.59}$, where intake fraction is in units of per million. This relationship combines Equation 4-1, the empirical relationship in Figure 4-2, a breathing rate of 12.2 m^3 d^{-1} person^{-1}, and a normalized dilution rate of 480 m^2 s^{-1}.

Uncertainty

I discuss here uncertainty in the input parameters and in the methods used. For the one-compartment and empirical models, method uncertainty is expected to be larger than input uncertainty. While rigorous uncertainty bounds are not known for the input parameters used in these two approaches, most of the input data (e.g., population, land area, wind speed) have relatively tight confidence intervals. An exception is the vehicle CO emission factor, for which the uncertainty is a factor of ~2 (Singer and Harley, 1996). These two methods do not account for differences between ambient and exposure concentrations, such as those occurring while traveling in a vehicle, nor do they incorporate emissions' spatial and temporal variability. To the extent that such factors increase the estimated inhalation intake rate, these two methods may underestimate the true intake fraction value. The NATA approach accounts for these two factors, which reduces method uncertainty, but in doing so, it increases both the number of input parameters and the input uncertainty. Most of the uncertainty information that NATA provides is qualitative. One exception is uncertainty in modeled ambient concentrations: comparisons between modeled

and measured concentrations indicate that the model under-predicts ambient concentrations by ~40% for conserved gases and by a factor of ~5 for particles (US EPA, 2004). Because intake fraction incorporates the ratio of concentrations to emissions, differences between measured and modeled concentrations may or may not lead to errors in the NATA-derived intake fraction estimates presented in this work. If these differences were attributable to the air dispersion model, then the NATA-derived intake fraction values presented in this work would be too low by ~40% for conserved gases and by a factor of ~5 for particles. In contrast, if these differences were attributable to errors in the emission inventory, then they would not indicate errors in the NATA-derived intake fraction values. The EPA considers the latter case to be more likely than the former (US EPA, 2004).

Comparing results among the three methods provides information about the overall method uncertainty. Combining results from the three methods with equal weight, the mean population-weighted annual average intake fraction for conserved gaseous vehicle emissions is estimated to be ~14 per million. The mean values from the three methods (excluding diesel PM because it is not a nonreactive gas) are within ~50% of this average. Thus, I estimate method uncertainty to be roughly 50%. Further investigations, applying additional methods to urban areas throughout the US, are necessary to confirm the intake fraction results and the uncertainty estimates presented in this work.

Total intake for urban emissions is the sum of the intakes within and downwind of the urban area. The results presented in this chapter only quantify intraurban intake. Previous work indicates that for urban vehicle emissions, the downwind intake may either be small relative to, or be comparable to, intraurban intake. Not enough work has yet been done to arrive at firm conclusions. Evans et al. (2002) found that 50% of the total

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inhalation of urban highway emissions occur within 100 km of the source. Greco et al. (2004) found that 41% of people in the US live in counties where a majority of the total inhalation intake of mobile source emissions occurs within their county borders. For vehicular emissions in California's South Coast air basin, I report in Chapter 3 that the downwind increment of intake is a few orders of magnitude smaller than the intake within the air basin.

Seasonal variability

I investigate seasonal variability for the 15 cities in Glen et al. (1996) and for six cities from the SCRAM database: Waycross, Georgia; Denver, Colorado; Atlantic City, New Jersey; Oakland, California; Peoria, Illinois; and Tucson, Arizona. These six cities were chosen to span a range of climates throughout the US. Following the approach by Glen et al., I group the SCRAM data into four summer months (May-August) and four winter months (November-February).

The analysis reveals that intake fraction values are, on average, higher in winter than in summer. The median intake fraction value calculated using the empirical model is ~40% larger in winter than in summer. (One city of the fifteen in the empirical model does not follow this trend: the calculated intake fraction for Buffalo, New York, is essentially the same in summer as in winter.) For five of the six SCRAM stations, the average normalized dilution rate is greater in summer than in winter, causing the calculated intake fraction to be 50 – 200% larger in winter than in summer. (At the sixth station, Peoria, Illinois, the normalized dilution rate is 80% larger in winter than in summer.) These estimates are consistent with the finding reported in Chapter 3 that the vehicle intake fraction in the South Coast Air Basin is ~100% larger in winter than in summer.

For these 21 cities, seasonal variability in the calculated intake fraction is more attributable to changes in mixing height than to changes in wind speed. On average, mixing heights are 90% higher in summer than in winter. This seasonal trend, with mixing heights being higher in summer than in winter, occurs in all 21 cities. In contrast, wind speeds for the 21 cities change by an average of 20% between summer and winter. For 10 of the 21 cities, wind speed is larger in summer than in winter; for the remaining 11 cities, the reverse is true.

Conclusions

I have used three independent methods to characterize intake fraction for nonreactive vehicle emissions in US urban areas. These three methods incorporate empirical results and models with different levels of sophistication. Intake fraction varies among locations, based on factors such as meteorology, linear population density, and the spatial distribution of emissions. Population-weighted annual-average mean intake fractions for nonreactive gaseous vehicle emissions in US urban areas are estimated to be ~14 per million, with an uncertainty of approximately 50%. Intraurban removal mechanisms, such as chemical reactions (as for 1,3-butadiene) and physical removal as air migrates from outdoors to indoors (as for diesel PM), reduce the intake fraction. Seasonal average intake fractions are usually higher in winter than in summer, owing primarily to changes in atmospheric mixing height.

Intake fraction is a useful metric for health risk assessments, cost-benefit analyses, and other investigations that require a summary of the emission-to-intake relationship. Earlier work (Bennett et al., 2002; Evans et al., 2002; Lai et al., 2000) highlighted the

merits of compiling intake fraction values and methods for various sources and pollutants. This investigation contributes to that goal.

Table 4-1: Linear population density values for selected California metropolitan areas, with estimated intake fraction values

Metropolitan Statistical Area (MSA)	Linear population density[a] (people m⁻¹)	Estimated intake fraction[b] (per million)
Los Angeles-Long Beach-Santa Ana	163	48
San Francisco-Oakland	74	22
San Diego	65	19
San Jose	54	16
Sacramento	48	14
Riverside-San Bernardino	44	13
Fresno	28	8.3
Oxnard	26	7.5
Bakersfield	20	5.9
Santa Barbara	16	4.8
Simi Valley	14	4.0
Lodi	14	4.0
Vacaville	12	3.6
Chico	11	3.2
Santa Cruz	9.8	2.9
San Luis Obispo	8.7	2.5
Redding	6.9	2.0

(a) Source: US DOT (2003a).

(b) Intake fraction estimates in this table are based on Equation 4-1, using $Q = 12.2$ m³ person⁻¹ d⁻¹, $uH = 41.5 \times 10^6$ m² d⁻¹, and the linear population density values in this table.

Table 4-2: Estimated annual average intake fraction (per million) for urban vehicle emissions using the three methods employed in this chapter

	One-compartment model	Empirical model, summer	Empirical model, winter	NATA, benzene	NATA, diesel PM
Range of values	0.1 – 280	5.7 – 31	7.7 – 54	-	-
Population-weighted					
mean	21	10	15	7.0	4.4
median	12	9.3	13	-	-
inter-quartile range[a]	5.1 – 25	8.4 – 11	11 – 16	-	-
10%-trimmed range[b]	2.4 – 50	7.6 – 15	11 - 29	-	-
Unweighted					
mean	5.3	12	19	-	-
median	3.0	9.3	13	-	-
inter-quartile range[a]	1.8 – 5.6	8.2 – 13	11 – 24	-	-
10%-trimmed range[b]	1.1 – 11	6.2 – 19	11 – 33	-	-

(a) The inter-quartile range is the range of values excluding the top 25% and bottom 25% of the distribution.
(b) The 10%-trimmed range is the range of values excluding the top 10% and bottom 10% of the distribution.
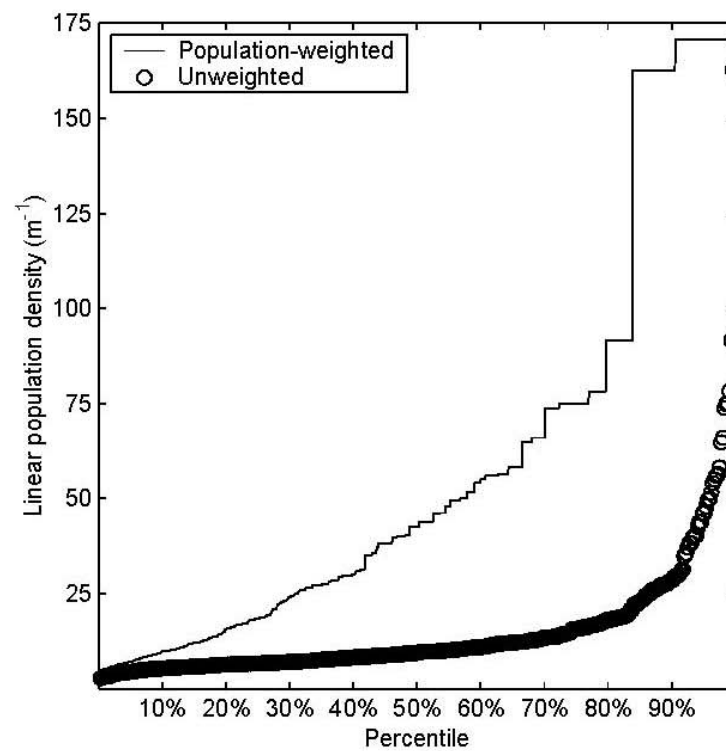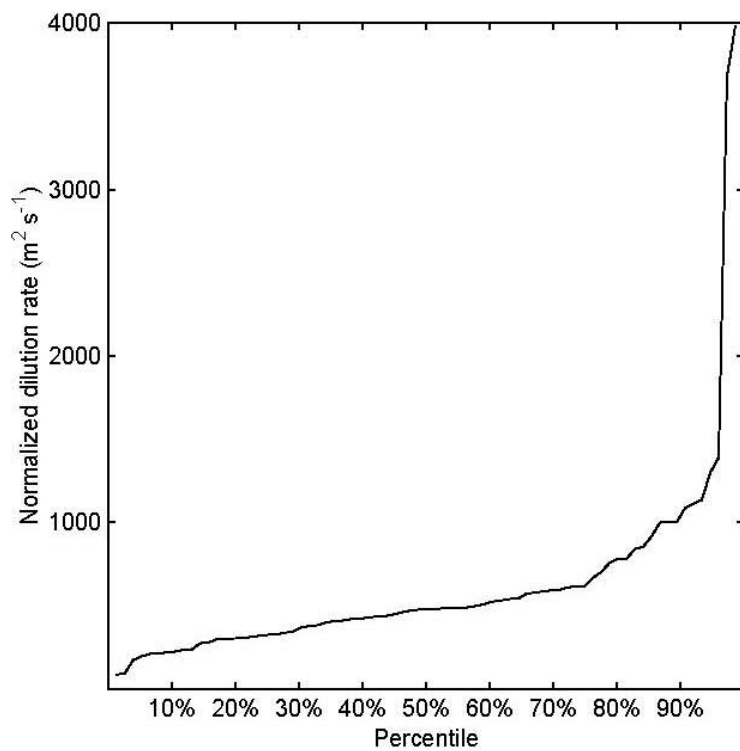
Figure 4-1: Distributions of values for normalized dilution rate (m^2 s^{-1}; left plot) and linear population density (m^{-1}; right plot). These two variables are input parameters to the one-compartment model.
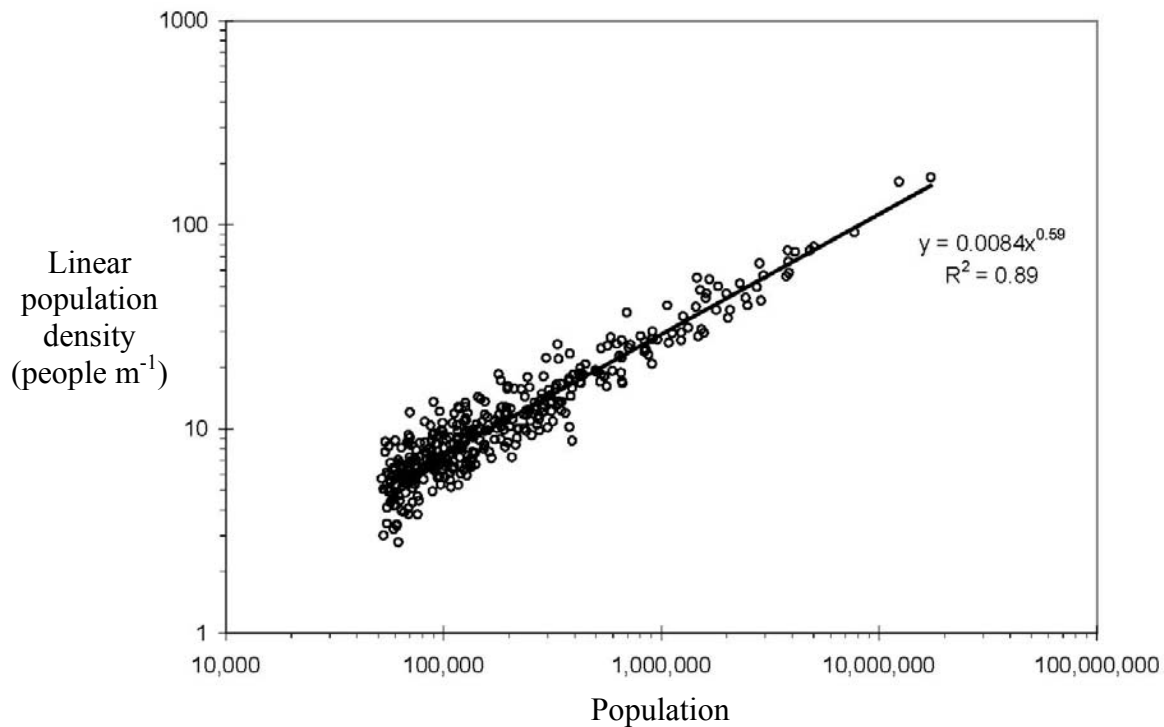
Figure 4-2: Relationship between linear population density and population for the 379 urban areas in the US with more than 50,000 people. Linear population density is the population divided by the square root of the land area. Urban areas with large populations tend to have large linear population densities, suggesting that increasing the urban population size increases the intake fraction. Data are from US DOT (2003a).
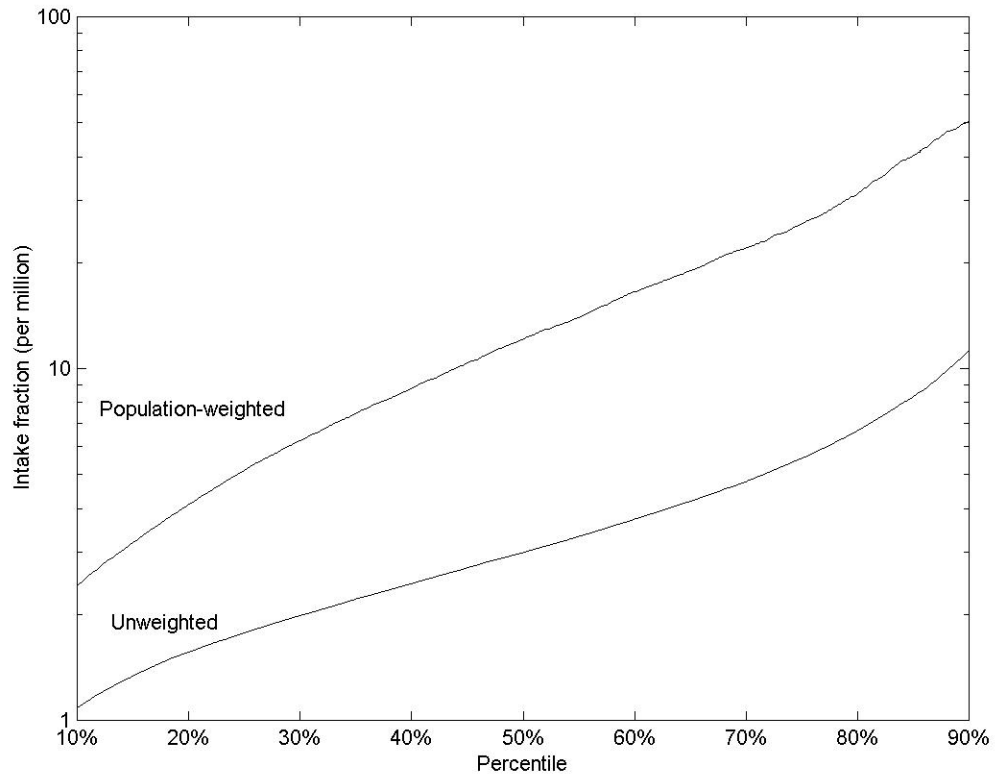
Figure 4-3: 10%-trimmed distribution of annual average intake fraction values for distributed ground-level releases of nonreactive pollutants in US urban areas using the one-compartment model.
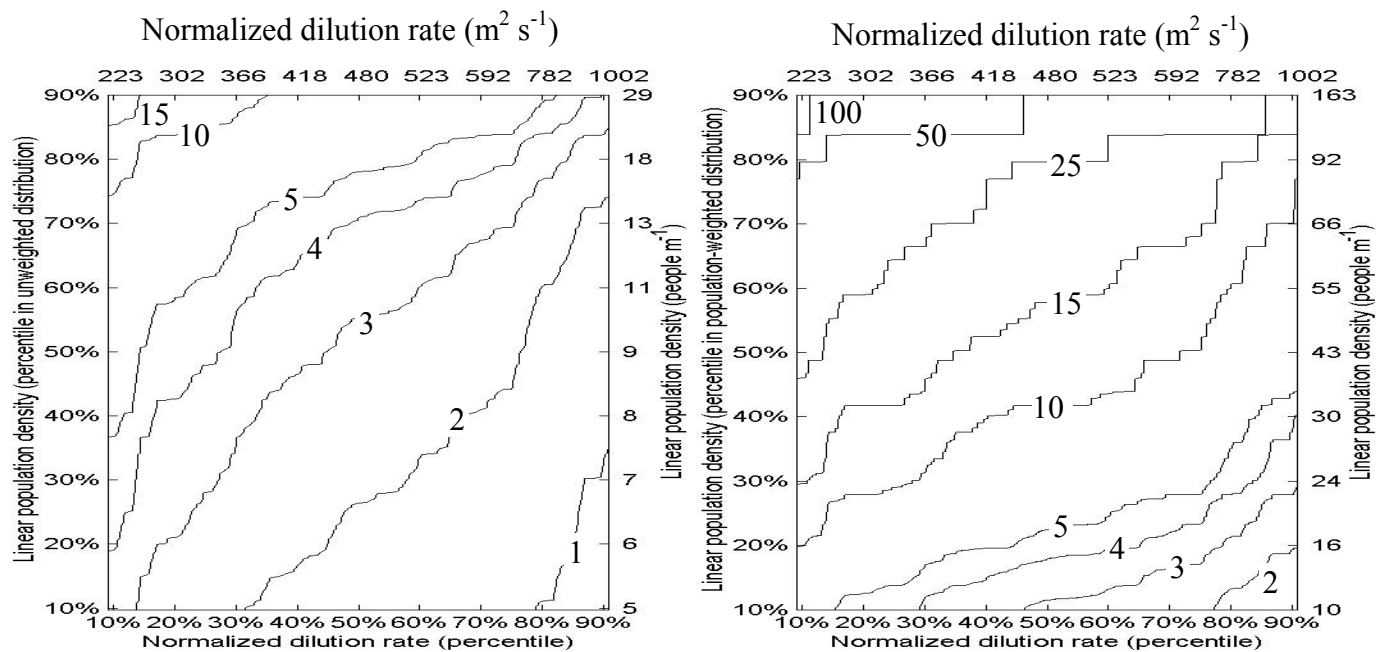
Figure 4-4: Isopleths of 10%-trimmed annual average intake fraction values (per million) for vehicle emissions in US urban areas based on the one-compartment model. In the left figure (the unweighted plot), the linear population density percentile values on the y-axis represent the distribution among urban areas in the US DOT (2003a) database. In the right figure (the population-weighted plot), the linear population density percentile values on the y-axis represent the distribution among people living in urban areas.
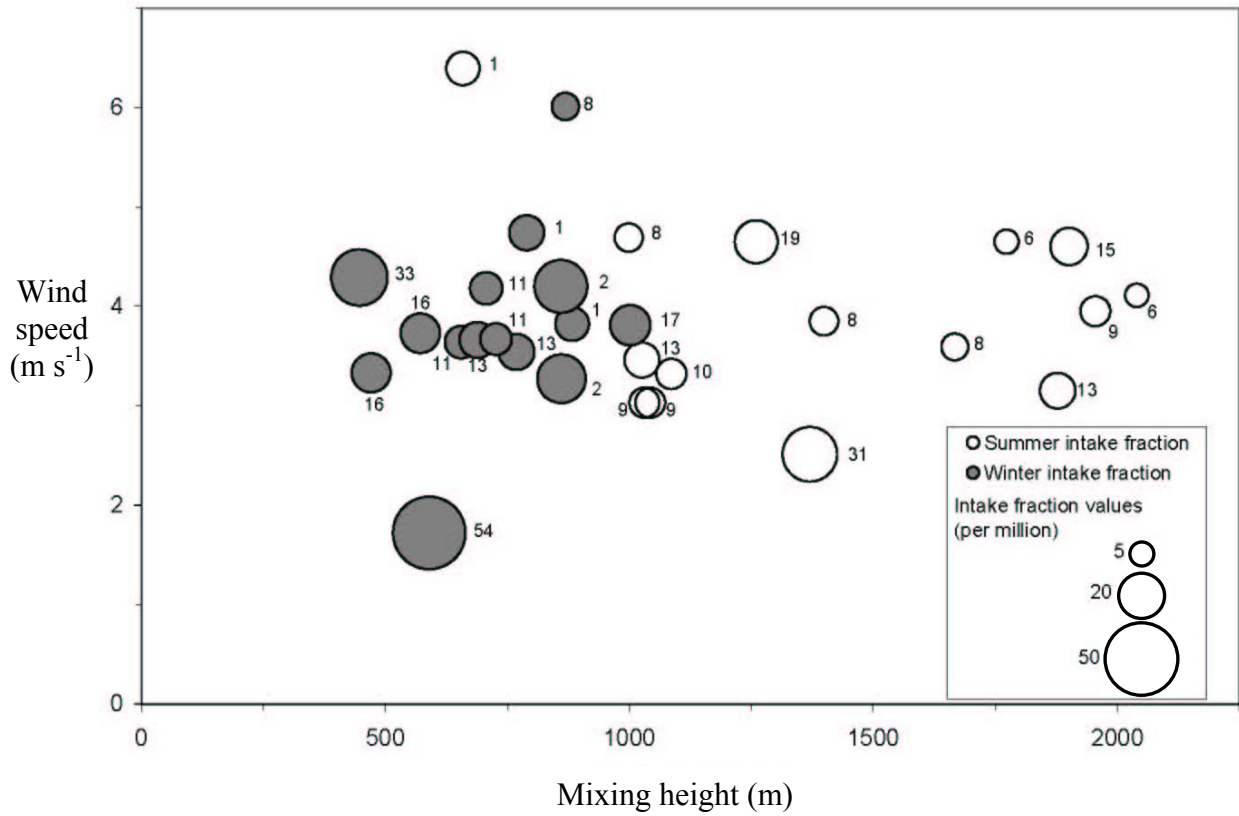
Figure 4-5: Summer and winter seasonal-average wind speed and mixing height (harmonic means) and intake fraction (iF) values (numerical labels) for vehicle emissions in the 15 urban areas in the empirical model. The area of each circle is proportional to the intake fraction value.

Chapter 5: Self-pollution intake fraction of school bus emissions

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Introduction

The two preceding chapters presented intake fraction estimates for urban areas. In this chapter, I focus on the intake fraction in a specific microenvironment, a specific population, and a specific vehicle type. Here, I investigate children's inhalation of vehicle emissions from the school bus in which they are riding.

As discussed in Chapters 2 and 3, concentrations of vehicular pollutants are higher in and near vehicles than at centrally located monitors (Flachsbart, 1995; Gulliver and Briggs, 2004; Rodes et al., 1998; Wallace, 1991; Wallace, 1996). Recent work confirms that pollution from other vehicles is important on school buses, especially in explaining short-term variability in on-board concentrations (e.g., particle concentrations increase after passing a diesel truck with visible emissions) (Sabin et al., 2005). Proximity to other vehicles is one reason for elevated in-vehicle concentrations (Rodes et al., 1998). However, a fraction of the pollution inside a school bus is attributable to the bus itself. This chapter investigates this type of pollution – which I term vehicle "self-pollution" – for students commuting on a school bus. As a major form of children's

transportation, school bus emissions represent a potentially important source of children's exposure to vehicle pollutants. Self-pollution increases exposure to vehicular emissions for those children riding a school bus.

School bus emissions, like other vehicle emissions, are a health concern in urban areas. However, school bus emissions are especially salient for two reasons. First, almost all bus engines are diesel engines. Diesel particulate matter (DPM) is estimated to cause a majority of the cancer risk attributable to ambient air pollution in the South Coast Air Basin (SCAQMD, 1999). Second, an important objective of air quality regulations is to reduce exposures, especially for sensitive subpopulations. Children are believed to be especially susceptible to air pollution because of their high inhalation rates and lung surface area per body weight, narrow lung airways, low lung clearance rates, and immature immune systems (CARB, 2002c; Dockery et al., 1989; Lipsett, 1995; Thurston, 2000).

Using results from previous tracer-gas experiments (Behrentz et al., 2004; Fitz et al., 2003), I estimate the fraction of emissions inhaled by the population of school bus riders (the intake fraction, iF) and by an average individual bus rider (the average individual intake fraction, iF_i). In addition to assisting in characterizing exposures and elucidating causative factors, this information can aid in designing effective exposure reduction strategies.

Methods

Tracer gas experiment

Tracer gas experiments were performed on six buses while traveling on an in-use

school bus route that covered highly urbanized areas of south-central Los Angeles and

suburban areas of West Los Angeles. Researchers from the University of California at

Los Angeles made measurements during seven runs with closed windows and nine runs

with open windows in April, May, and June, 2002. Table 5-1 summarizes the

characteristics of these runs. Experimental methods are described elsewhere (Behrentz et

al., 2004; Fitz et al., 2003). In each bus run, a mass flow controller metered the delivery

of a tracer gas, sulfur hexafluoride (SF_6), from a high-concentration cylinder into the

bus's exhaust system. On-board SF_6 concentrations were measured at two locations (front

and rear) with an electron capture detection analyzer (AeroVironment Model CTA 1000).

Model years of the buses were 1975, 1985, 1993, 1998 (two buses), and 2002. These

buses included two older (year-1975 and 1985) high-emitting (HE) diesel buses, two

diesel buses more representative (RE) of current fleets, one particle trap-outfitted (TO)

diesel bus, and one bus powered by compressed natural gas (CNG).

Intake fraction

Intake is the mass of a pollutant that is taken in by an exposed individual or

population. For inhalation of air pollution, intake rate (g min^{-1}) is the product of

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volumetric breathing rate (L min⁻¹) and exposure concentration (g L⁻¹). I calculate intake

fraction for school bus self-pollution, iF_{SP}, using Equation 5-1:

$$iF_{SP} = \frac{Q_B P C}{E}.$$ (5-1)

Here, Q_B is the average breathing rate (m³ person⁻¹ min⁻¹), P is the average number of

people on a school bus, C is the temporally and spatially averaged on-board SF_6

concentration (g m⁻³) during a bus run, and E is the experimental SF_6 mass emission rate

(g min⁻¹) into the bus's exhaust during a bus run, calculated as the product of the

concentration of SF_6 (g L⁻¹) in the high-pressure SF_6 gas cylinder and the metered flow

rate of gas (L min⁻¹) from the cylinder to the bus's exhaust.

The variables in Equation 5-1 can be grouped into two terms. The first term, $Q_B P$

(units: L min⁻¹), is the volume of air inhaled per minute by the exposed population. The

second term, $C E^{-1}$ (min L⁻¹), indicates the magnitude of self-pollution, as measured by a

tracer gas. I define a self-pollution term, S, as $C E^{-1}$, which is proportional to the mass

fraction of emissions that enter the bus (M_f):

$$S = \frac{C}{E} = \frac{M_f \tau}{V_{bus}}.$$ (5-2)

Here, V_{us} is the interior volume of the bus (L), and τ is the mean residence time of air

inside the bus (min). The residence time of air inside the bus depends on the rate of air

exchange between inside and outside air, which in turn depends on factors such as the

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window position (open or closed), vehicle speed, and wind speed (Fitz et al., 2003; Park et al., 1998).

Values for the self-pollution term, S, were calculated from reported tracer gas experiments for school-bus commutes (Fitz et al., 2003), as given in Table 5-1. Data on breathing rate and number of passengers are not available for students on a school bus. Based on children's metabolic rates at rest and at light activity levels (Layton, 1993), I estimate that children's average breathing rate on a school bus is between 7.2 and 22 L min^{-1}. Activities given in Layton (1993) that correspond to this range of breathing rates include "sweeping", "preparing vegetables", and "cooking". The middle estimate, used in the analyses below, is the average of these two values (14.6 L min^{-1}). I estimate that the average number of children on a school bus is 40, and in the range 20 to 50. The middle estimate equals the number of public school students transported by buses in California during the 2000-2001 school year (964,815 students) divided by the number of buses available (24,497 buses) (School Transportation News, 2003). On any given day, some of the 964,815 students will not ride a bus, and not all of the buses in the fleet will be used. The time spent on a bus varies among students.

Results

Table 5-1 summarizes the SF_6 measurement results and the calculated intake fractions for each bus run (i.e., each row represents a single tracer-gas experiment). Figure 5-1 depicts the self-pollution term, S, for the six buses studied. Self-pollution is substantial for all six buses.

Bus age and window position affect the magnitude of self-pollution, with older buses and closed-window buses having higher self-pollution levels. On average, S values are ~2 times higher with windows closed than with windows open. The importance of window position increases with bus age: the difference between open and closed windows is ~20% for the newer buses and a factor of ~3 for the older buses. Similarly, the importance of bus age increases when windows are closed: the difference in average S value between the oldest (model year 1975) and the newer buses (model year 1993 and later) is a factor of ~2 with windows open and a factor of ~6 with windows closed.

Total intraurban inhalation intake for school bus emissions has two components: inhalation intake by passengers (self-pollution) and inhalation intake by all other people (excluding self-pollution). Intake fraction (iF) is equal to self-pollution intake fraction (iF_{SP}) plus intake fraction excluding self-pollution (iF_{non-SP}). These two components (iF_{SP} and iF_{non-SP}) are presented separately in Figure 5-2. Values for iF_{SP} are estimated from the tracer gas experiments (Behrentz et al., 2004; Fitz et al., 2003) analyzed here. Values for iF_{non-SP} for primary, nonreactive pollutants are estimated as the fleet-wide average intake fraction for motor vehicle emissions of carbon monoxide in the South Coast Air Basin (46 per million). This value, derived in Chapter 3, accounts for spatial variability in population density and ambient concentrations; temporal variability in concentrations and breathing rates; and microenvironments such as in- and near-vehicle and indoors near a freeway. The true non-self-pollution intake fraction for buses may differ from the value of 46 per million derived in Chapter 3 because school-bus travel patterns differ from the fleet average. For example, the timing of emissions during the day and the spatial location of the emissions (e.g., freeway versus highway) differ between school buses and

the fleet average. However, there is not available information to estimate readily how these differences in the spatial and temporal emission patterns influence intake fraction.

Self-pollution intake fraction is estimated in this work from ~90-minute tracer gas experiments, while the estimate of non-self-pollution intake fraction is based on annual exposures in the South Coast. Results from these two methods can be compared because the analyses are independent of exposure duration, which is true for two reasons. First, the periods analyzed are much longer than the residence times of air in the respective environments. The tracer gas experiments were performed for much longer than the residence time of air in a bus (Fitz et al., 2003), and the South Coast analysis considered a much longer time period than the residence time of air in an air basin (see Chapter 3). Second, in this work, I do not analyze temporal variability in self-pollution intake fraction (i.e., when using Equation 5-1, I incorporate average, rather than time-varying, values for the parameters).

Figures 5-2 and 5-3 depict intake fraction and average individual intake fraction for nonreactive gaseous emissions. Because the self-pollution term is similar in magnitude for the newer buses, these four buses are combined in Figures 5-2 and 5-3. Average values across all bus runs are 27 per million for school bus self-pollution intake fraction, 73 per million for school bus intake fraction (i.e., including self-pollution and non-self-pollution intakes), and 0.7 per million for school bus self-pollution individual intake fraction.

Figures 5-2 and 5-3 use the arithmetic mean of front and rear S values for the newer buses and for the oldest bus. Error bars assume that uncertainty in the average number of students on a school bus (P) and in students' average breathing rate (Q_B) align

to yield the maximum uncertainty in intake fraction and individual intake fraction. The iF values in Figure 5-2 indicate that for every million grams of a primary conserved pollutant emitted by a school bus, a total of ~46 g are inhaled by the ~15 million people in the South Coast Air Basin who are not riding that bus, and that the passengers inhale an additional 10 – 54 g for a newer school bus, and 13 – 94 g for an older school bus.

The values in Figure 5-2 apply to nonreactive gasous emissions, but a similar comparison that focused on diesel $PM_{2.5}$ (DPM) would be likely to yield similar results. For example, I estimate here that the value for iF_{non-SP} for primary $PM_{2.5}$ is 33 per million. This value combines the estimate of 46 per million for iF_{non-SP} for primary, nonreactive pollutants (shown in Figure 5-2) with the estimate that people spend 87% of time indoors where ambient $PM_{2.5}$ concentrations are 67% of their value outdoors (see Chapter 2, Appendix). Thus, the non-self-pollution intake fraction value for $PM_{2.5}$ is approximately 71% (i.e., 87% × 67% + (100% - 87%)) of the same value for a nonreactive gas. The difference (33 per million versus 46 per million) is attributable to the protection buildings offer against ambient $PM_{2.5}$. Furthermore, I estimate here that the school bus self-pollution intake fraction is approximately the same for $PM_{2.5}$ as for nonreactive gases, for two reasons. (1) Cracks and leaks in a school bus tend to be relatively large (large enough to allow in visible light), and thus are unlikely to remove a significant fraction of $PM_{2.5}$ as air migrates into the bus. (2) The residence time of air on a bus is short enough that only a small fraction of the $PM_{2.5}$ would be removed in-vehicle (e.g., via deposition).

Figure 5-3 presents average individual intake fractions. Average individual intake fraction is equal to intake fraction divided by the number of people exposed. The right-most bar represents a typical person's exposure to emissions from an average vehicle in

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the South Coast Air Basin. The remaining values are for a student's exposure to emissions from the school bus on which they commute. For example, for every million grams of a primary, conserved pollutant emitted by an old bus with closed windows, ~2 g are inhaled by the average person on that bus, and ~2×10^{-6} g are inhaled by the average person who is not riding on that bus. The difference in intake fraction among the cases in Figure 5-2 (iF_{SP} for a newer and older bus, and iF_{non-SP}) is less than one order of magnitude, but the size of the exposed population varies by more than five orders of magnitude (~40 people for self-pollution, and ~15 million people for non-self-pollution). Therefore, the difference in individual intake fraction values between self-pollution and non-self-pollution is between 5 and 6 orders of magnitude. That is, the emission-to-individual-intake ratio is 10^5–10^6 times greater for children inhaling their own school bus's emissions than for the average South Coast resident inhaling emissions from a single average school bus. Even with the order of 10^4 school buses operating daily in the South Coast Air Basin, the individual inhalation intake from all school buses is much larger for bus-riding school children owing to vehicle self-pollution than for the average urban resident.

Discussion

Self-pollution intake

The relationship between reductions in emissions and reductions in a child's intake varies significantly among sources. For example, reducing annual DPM emissions by 1 tonne would reduce an average exposed child's annual intake of DPM by 3×10^{-3}

mg if the reduction comes from ambient emissions from a typical diesel vehicle, by 400 mg if the reduction comes from the child's school bus if he rides a newer bus, or by 1,000 – 2,000 mg if the reduction comes from the child's school bus in the case that he rides an older bus. In other words, because of self-pollution, school buses are orders of magnitude more effective than an average vehicle at delivering emissions to bus-riding children's lungs. Reducing an exposed child's intake by a specific amount would require 4 to 6 orders of magnitude greater emission reduction if control strategies target typical diesel vehicles (e.g., heavy duty diesel trucks) than if strategies target self-pollution from that child's school bus.

Health risk assessment

An estimate of the overall health risk can be made based on the intake fraction values above. DPM emissions from a diesel bus are estimated by the US EPA MOBILE6.2 emission factor models as 0.5 g km^{-1} for year-2004 buses and 2.0 g km^{-1} for year-1985 and older buses (US EPA, 2003a). (An alternative US EPA model, PART5, gives vales that are 40 – 50% lower than these values from MOBILE6.2. EPA considers the values from MOBILE6.2 to be more reliable than those from PART5 (US EPA, 2003a).) The concentration-based lung cancer unit risk factor for lifetime exposure to DPM is 0.0003 (μg m^{-3})$^{-1}$ (CARB, 1998; OEHHA, 1998). This value means that the lung cancer risk attributable to a lifetime average exposure to 1 μg m^3 of DPM is 0.0003, or 300 per million. Based on a lifetime average breathing rate of 12.2 m^3 d^{-1} and a lifetime duration of 70 y (i.e., based on a breathing rate of 312,000 m^3 lifetime^{-1}), the

concentration-based unit risk factor can equivalently be expressed as an intake-based unit risk factor of 0.0010 g^{-1}. This value means that the lung cancer risk attributable to a lifetime inhalation intake of 1 g of DPM is 0.001, or 1000 per million. Based on the common assumption of a linear dose-response for cancer risk, this intake-based unit risk factor holds whether one considers the intake by an individual or by a group.

According to the US EPA's Clean School Bus program website (www.epa.gov/cleanschoolbus), school buses drive more than 6.4 billion km per year, and 24 million children per day ride a school bus. Combining this distance traveled (6.4 × 10^9 km y^{-1}) with the range of MOBILE6.2 emission factors for new and old buses (0.5 – 2.0 g km^{-1}), total DPM emissions from school buses are estimated at 3,000 – 13,000 t y^{-1}. If the average self-pollution intake fraction is 27 per million, then the cumulative inhaled mass of DPM by children is 90 – 360 kg y^{-1}. Using an intake-based toxicity of 0.001 g^{-1}, the estimated number of lung cancer cases attributable to self-pollution intakes is 90 – 350 y^{-1}. For each year of school bus fleet operation, an estimated lifetime burden of 90 – 350 lung cancer cases is estimated to occur among school children owing to vehicle self-pollution. The average annual risk per child, assuming an exposed population of 24 million bus riders, is 4 per million – 14 per million. The total risk attributable to 13 years of exposure (i.e., during the age range of 5 – 18 years old) is 52 per million – 180 million. Assuming that the person does not have significant school-bus ridership after age 18, these values represent the lifetime average lung cancer risk attributable to an individual's school bus. These values are broadly consistent with an extant health risk assessment that estimated that the lifetime lung cancer risk attributable to riding a school bus as a child is ~30 per million (CARB, 2003a). These lung cancer risk values are well above the "de

minimus" risk level of 1 per million identified in the 1990 Clean Air Act. These values represent average risks for a large population. Risks are highest for the most exposed students, including those who ride the buses with the highest emissions and the highest self-pollution levels, and those riders with the longest exposure duration (i.e., the longest commute). Because of the assumptions involved in these health risk assessment calculations, they represent only a magnitude estimate of the health risks. Nevertheless, the estimate is useful for understanding the scale of the problem.

Control strategies

The school bus microenvironment contributes significantly to children's estimated total inhalation intake of DPM. Approximately 90% of school bus fuel consumption is diesel (Davis and Diegel, 2003). On commute days, for newer and year-1975 buses, students commuting on school buses have 34% and 70% higher 24-hour total intakes of DPM than car-commuters, respectively (CARB, 2003a). The daily inhalation intake by a child of emissions from the one school bus on which he or she commutes is between ~7 and ~70 times greater than the average daily inhalation intake by a typical South Coast resident of emissions from all school buses.

Both emissions and self-pollution intake fraction are higher for old buses than for new buses. The difference between newer (model year 1993 and later) and older buses (model years 1975 and 1985), for windows closed, is a factor of ~2 for average iF values (63 per million versus 140 per million) and ~5 for average iF_{SP} values (17 per million versus 94 per million). The emission factor difference between newer and older buses, according to EPA's MOBILE6 emission model, is approximately a factor of 10 (US

EPA, 2003a). The correlation between vehicle age, vehicle emissions, and both iF and iF$_{SP}$ suggests that older buses should be a much higher emission reduction priority than newer buses. Based on the results I presented in this chapter, even if vehicle emission control technologies were implemented on older buses, such that emissions from all buses were equal, self-pollution intake would still be higher on older buses than on newer buses because of the higher iF$_{SP}$ values.

Inhalation intake equals emissions times intake fraction. Intake control strategies should aim to reduce both emissions and intake fractions. Intake fraction can be reduced, for example, by decreasing the use of older school buses and by better decoupling the exhaust flow from air flowing into the bus. Improved understanding of self-pollution mechanisms may suggest additional exposure control strategies. Opening windows, which reduces τ, may reduce self-pollution.

It is important to identify the mechanism of self-pollution (CARB, 2003b). Many emission control technologies are applied at the end of the tailpipe. If the dominant mechanism for self-pollution transport is post-tailpipe, then end-of-tailpipe technologies will reduce all attributable exposures, including self-pollution. But, if the dominant mechanism for self-pollution occurs before emissions exit the tailpipe, then end-of-tailpipe technologies will not reduce attributable exposures from self-pollution.

Variability and uncertainty

Relative to other urban areas in the US, the South Coast Air Basin has a large population size (15 million people) and generally unfavorable atmospheric mixing and pollutant transport conditions (CARB, 2002a). Both factors increase intake fraction

associated with ambient emissions. Average non-self-pollution intake fraction, iF_{non-SP}, in most urban areas is less than the value of 46 per million used in this chapter for CO in the South Coast. The value of 46 per million for the South Coast is derived in Chapter 3. Results in Chapter 4 indicate an intake fraction of 14 per million for nonreactive, gaseous vehicle emissions as a population-weighted mean for all US urban areas. Similarly, based on Eulerian air dispersion modeling, and not accounting for microenvironmental exposures, Evans et al. (2002) reported values for iF_{non-SP} in the range 3–18 per million for primary $PM_{2.5}$ emitted by motor vehicles in urban areas. Thus, the average self-pollution intake fraction among all bus runs in this study (27 per million) is larger than the non-self-pollution intake fraction for nonreactive vehicle emissions in a typical US urban area. Stated differently, when considering two groups in a typical urban area – students who ride a school bus and everyone else – the total mass of bus pollution inhaled by school bus riders likely exceeds the total bus pollution inhaled by the remaining public, despite school bus riders representing a small proportion of the population.

Self-pollution intake fraction will vary based on factors such as the window position, bus speed, wind speed and direction, and the bus's shape and structural integrity. The results presented in this work are averages over the conditions tested. Given the small sample size in the original tracer gas study (six buses, 16 runs), results presented in this chapter should be considered indicative, rather than representative of the entire bus fleet. Additional research is needed to refine and extend the results presented here, for example, by employing additional buses, bus types, and operating conditions.

Uncertainty and variability in SF_6 emission rates and on-board concentrations do not affect the results significantly (CARB, 2003a; Fitz et al., 2003). There is uncertainty

in the average activity level, and therefore also in the average breathing rate, for students on buses. Based on the range of breathing rates presented in Layton (1993), uncertainty in the average breathing rate is estimated as approximately a factor of 2.

There is also uncertainty regarding the average number of students on each bus. The estimate used above, 40 children per bus, assumes that all buses are being used, that all students are attending school each day, and that students ride the bus for the entire duration of the route. The capacity on a typical school bus is larger than 40 students. For example, one school bus manufacturer, Blue Bird ®, sells three makes of standard size school bus: the "All American" with forward engine (three possible wheelbases; seating capacity ranges between 72 passengers and 89 passengers), the "All American" with rear engine (four possible wheelbases; seating capacity ranges from 63 to 84 passengers), and the "Vision" (five possible wheelbases; seating capacity ranges from 48 to 78 passengers). An estimate of average number of students on a bus would need to account for travel time with no students, e.g., between the bus garage and the start of the bus route. As a back-of-the-envelope estimate, assume that (1) the average capacity for a school bus is 70 students, and (2) that a bus operates for an equal duration at each of three capacity factor levels: 0%, 50%, and 90%. In this case, the average capacity factor is 46% and the time-average utilization is 33 students per bus. This value is in the range of values used in this chapter (20 – 50 students per bus), and is within 20% of the best estimate value of 40 students per bus. The level of uncertainty in this estimate (40 students per bus) is difficult to determine, but is estimated to be approximately a factor of 2 or less. Variability and uncertainty in breathing rate and number of students on a school

bus affect the detailed numerical results, but do not alter the broad conclusions I draw from these results.

Conclusions

Vehicle self-pollution occurs when a vehicle's emissions migrate inside that vehicle's passenger compartment. This chapter presented values for two parameters: vehicle self-pollution intake fraction (iF$_{SP}$), which is the total fraction of a vehicle's emissions inhaled by all people in the vehicle, and vehicle self-pollution individual intake fraction (iF$_{i,SP}$), which is the fraction of a vehicle's emissions inhaled by an individual in the vehicle. I used results from tracer-gas experiments in California's South Coast Air Basin (SoCAB) to quantify students' iF$_{SP}$ and iF$_{i,SP}$ for school-bus emissions. Six buses were studied during nine runs with windows open and seven runs with windows closed. The resulting iF$_{SP}$ values (units: per million; min = 10, max = 94, mean = 27) indicate that the total mass of a bus' exhaust inhaled by students commuting on it is comparable in magnitude to the total mass of that bus' exhaust inhaled by all other people in the SoCAB. Reported iF$_{i,SP}$ values (units: per million; min = 0.2, max = 2.4, mean = 0.7) indicate that average per capita inhalation of emissions from any single bus is $10^5 - 10^6$ times greater for a student on that school bus than for a typical resident in the SoCAB. Vehicle self-pollution rate varies with bus window position (open or closed) and bus manufacture year. The results presented in this chapter can contribute to the development of cost-effective strategies to reduce children's exposure to school-bus emissions. The results indicate, for example, that even if emission reductions were many times more expensive per gram emitted for school buses than for an average vehicle, it would still be

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less expensive per gram inhaled by a student to reduce emissions from school buses than from an average vehicle.

Table 5-1: Self-pollution intake fraction and individual intake fraction results[*]

Model year	Window position	Bus designation	S, front $(10^{-9}$ min $L^{-1})$	S, rear $(10^{-9}$ min $L^{-1})$	S, average $(10^{-9}$ min $L^{-1})$	iF_{SP} (per million)	$iF_{i,SP}$ (per million)
1975	Open	HE3	76	82	79	46	1.2
1975	Closed	HE3	100	220	160	94	2.4
1985	Open	HE2	18	28	23	13	0.33
1985	Closed	HE2	70	100	86	50	1.3
1993	Open	RE2	47	8	27	16	0.40
1993	Open	RE2	23	22	23	13	0.33
1993	Closed	RE2	19	28	23	14	0.34
1993	Closed	RE2	14	21	17	10	0.25
1998	Open	RE1	83	102	93	54	1.4
1998	Open	RE1	17	35	26	15	0.38
1998	Open	TO	14	20	17	10	0.25
1998	Open	TO	29	27	28	16	0.41
1998	Closed	TO	35	44	40	23	0.58
1998	Closed	TO	31	35	33	19	0.48
2002	Open	CNG	25	23	24	14	0.35
2002	Closed	CNG	25	41	33	19	0.48
		Average	**39**	**52**	**46**	**27**	**0.67**

[*] HE2, HE3 = high emitter diesel school buses; RE1, RE2 = representative diesel school buses; TO = particle-trap outfitted diesel school bus; CNG = compressed natural gas school bus; S = the self-pollution term, calculated as the ratio of the on-board SF_6 concentration (g L^{-1}) to the SF_6 emission rate (g min^{-1}); iF_{SP} = self-pollution intake fraction; $iF_{i,SP}$ = average self-pollution individual intake fraction. Based on Equations 5-1 and 5-2, iF_{SP} is calculated as the product of the average breathing rate, the estimated population on the bus, and the self-pollution term, S. The values presented here assume an average breathing rate of 14.6 L min^{-1} and an average population of 40 people on each bus.

(a)

(b)
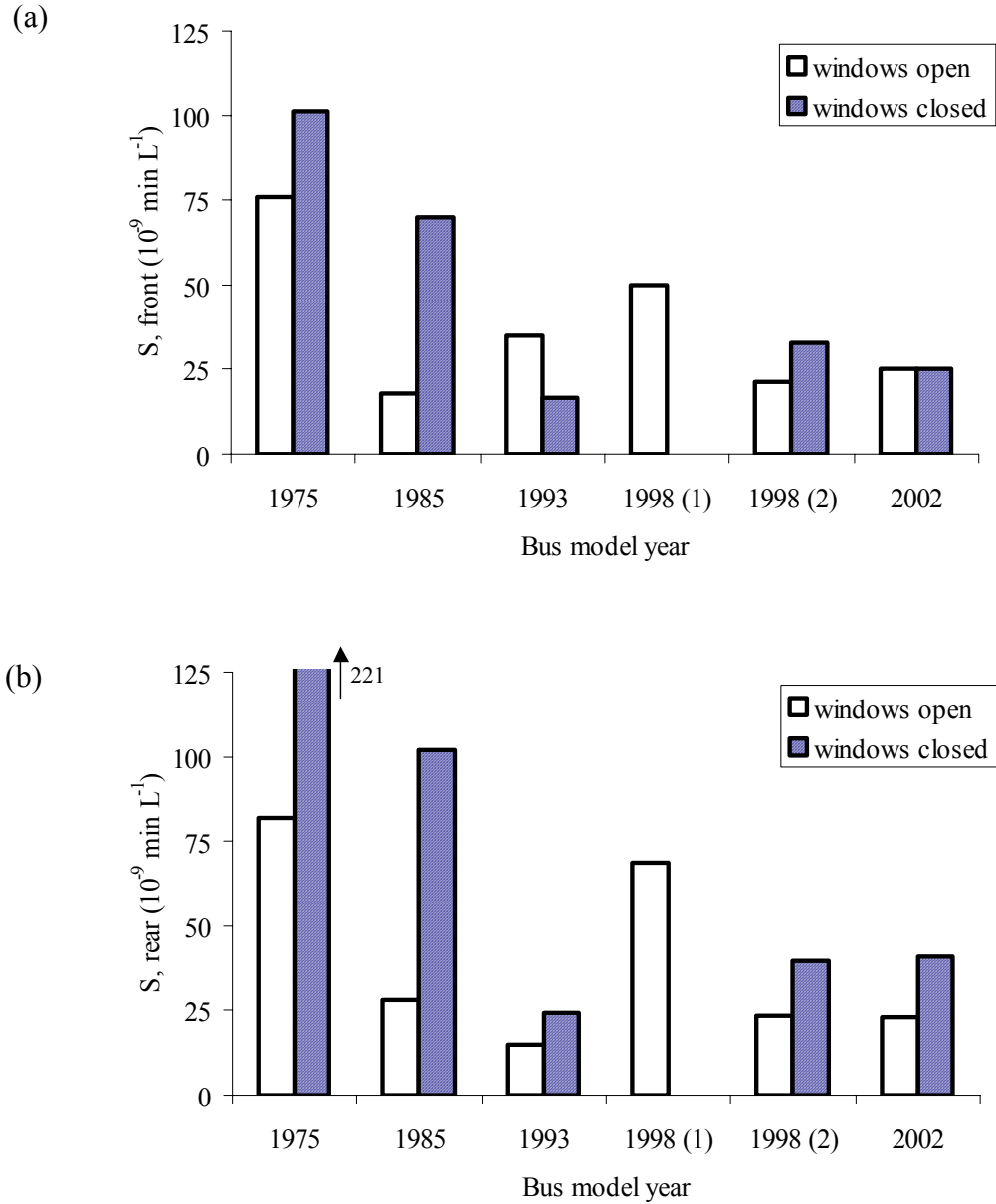
Figure 5-1: Self-pollution term, $S\ (=C\,E^{-1})$, versus bus model year for the front (upper plot) and rear (lower plot) of each bus, and with open and closed windows. Older buses have higher self-pollution levels than newer buses, but the level is still significant for newer buses. To facilitate comparisons between the two graphs, the scales were chosen such that one of the values in the lower plot (year-1975, closed windows; $S_{rear} = 221 \times 10^{-9}$ min l^{-1}) extends off the plot.
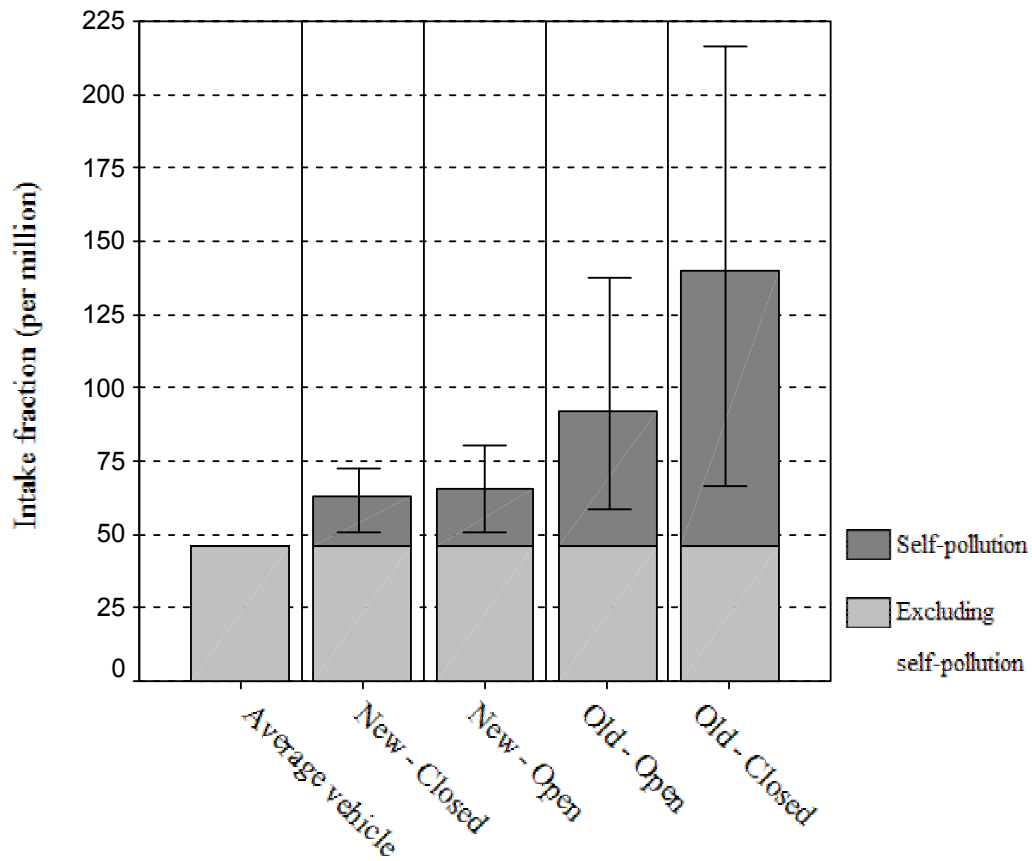
Figure 5-2: Intake fraction for an average vehicle and for school buses in the South Coast. Intake fraction is the sum of self-pollution intake fraction and non-self-pollution intake fraction. X-axis category labels refer to the bus model year ("new" means 1993 or later; "old" means 1985 or earlier) and the window position (open or closed). The error bars indicate uncertainty in the self-pollution term, owing to uncertainty in the breathing rate and in the number of people on the bus and assuming that uncertainty in these two parameters align to yield the maximum possible uncertainty.
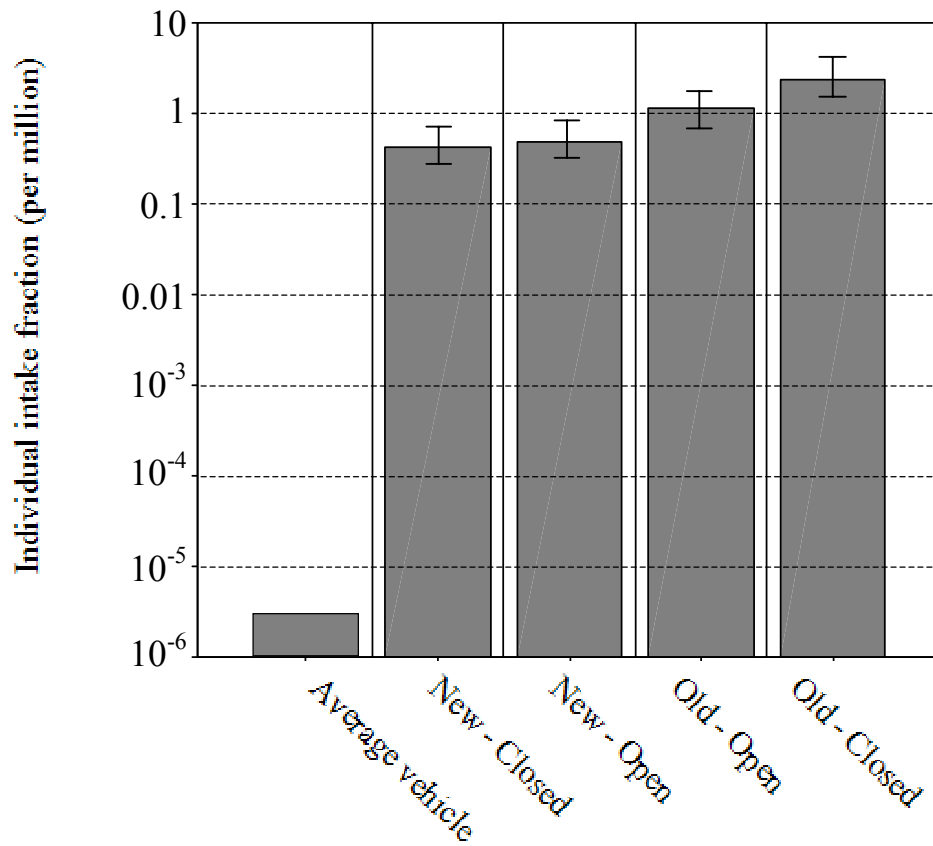
Figure 5-3: Individual intake fraction for an average vehicle and for school buses in the South Coast. The first (left-most) value is for a typical person's inhalation of emissions from an average vehicle without self-pollution. The remaining values are for a student's inhalation of emissions from the school bus on which they commute. Note the log scale. Error bars and X-axis labels are analogous to those in Figure 5-2.

Chapter 6: Mobility-based GIS model of inhalation intake of air pollution in the South Coast Air Basin

Introduction

One of the main challenges in environmental health is accurately estimating pollutant intake. For example, environmental epidemiology relies on exposure assessment to determine dose-response relationships, and health officials use exposure levels to estimate the total health impact of air pollution. Understanding and addressing distributional issues, such as correlations between exposure and demographic attributes such as ethnicity and income, is important for establishment of equitable policy goals.

Current air pollution exposure models rarely, if ever, account for population mobility. This statement includes those models considered and developed in the preceding chapters of this dissertation. For example, using home-based census data (as is done in Chapter 3) implicitly assumes that people spend 100% of their time at home. Yet, we know that people move within an air basin for employment, shopping, recreation, and for other purposes. In doing so, they encounter pollutant concentrations that, in general, may differ from those at their home. This chapter addresses whether mobility matters for air-pollutant exposure analyses. While Chapters 2–5 emphasized intake fraction, this chapter and the next one focus on population inhalation of air pollution.

I use data from a large (~29,000 person-days) time-location-activity survey in Southern California along with spatially and temporally resolved modeled ambient

concentrations to estimate population inhalation of air pollutants of outdoor origin. Each activity is geocoded, providing people's location (latitude and longitude) over time. Spatially and temporally explicit ambient concentration estimates are combined with microenvironment factors and breathing rates to yield the inhalation intake rate. The results are evaluated to determine correlations between air-pollution intake rate and demographic attributes such as race and income.

The research I describe in this chapter offers two significant advancements to exposure modeling science. First, I incorporate time-varying breathing rates, which makes more accurate the connection between exposures and intakes. In addition to presenting the diurnal breathing rate profile estimated in this chapter, I also explore explicitly the impact of time variability in population breathing rate on inhalation intake of air pollutants. To my knowledge, prior to the research presented in this dissertation, this issue has been raised in only one extant publication. Hall et al. (1993) used time-varying breathing rates in their assessment of inhalation intake of ozone and PM_{10} in the South Coast. Unfortunately, Hall et al. (1993) do not give sufficient detail to understand what was done or to recreate their work. (They devote less than one sentence to this issue.) Second, the model presented in this chapter explicitly incorporates individuals' movement throughout an urban area. Most exposure analyses do not address the issue of mobility (e.g., Fruin et al., 2001; Lu et al., 2005; Wu et al., 2005). A few publications have pointed to its importance; Hayes (1989) may have been the first to do so. Based on a generalized exposure-modeling framework, he showed that mobility could impact modeled exposure rates. Boudet et al. (2001) explored how proximity to roads influences the relationship between ambient and exposure concentrations for PM_{10} and $PM_{2.5}$. Their analysis included both home and work locations.

To my knowledge, only one exposure modeling study, by Gulliver and Briggs (2005), tracks individuals' exposures as they move through predicted concentration fields. They report journey-time exposure (i.e., exposures during travel activities) to PM_{10} for 5 days for 50 children living and walking to school in Northampton, England. The research presented in the chapter goes much further in evaluating the effects of mobility on exposures, by incorporating a significantly larger sample and longer duration (~29,000 versus 250 person-days) and by modeling inhalation intake in all microenvironments during each day rather than modeling exposures in one microenvironment (in-transit).

Methods

Inhalation intake of air pollution is calculated in this chapter using Equation 6-1:

$$I = \int_{T_1}^{T_2} Q(a(t)) C_{amb}(x, y, t) \gamma_{\mu}(t) dt, \qquad (6\text{-}1)$$

where I is the mass of pollutant inhaled (μg) by an individual between times T_1 and T_2 (h); $Q(a(t))$ is the volumetric breathing rate ($m^3 h^{-1}$), which depends on the person's activity level, $a(t)$, which changes over time, t; $C_{amb}(x,y,t)$ is the ambient (i.e., outdoor) concentration ($\mu g\ m^{-3}$) near the person, which is a function of location (x,y) and time; and $\gamma_{\mu}(t)$ is a dimensionless factor for each microenvironment, μ, that accounts for differences between the ambient concentration and the exposure concentration in that microenvironment. In practice, the above integral is evaluated as a series of sums over discrete time intervals. These intervals are chosen such that the three variables (breathing

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rate, ambient concentration, and microenvironment factor) are reasonably represented as constant for the duration of each interval. The input data for these three variables, and my methods for combining them, are described next.

Breathing rates

I use age-, gender-, and activity-specific breathing rates from metabolic activity studies (Layton, 1993). Age and gender are recorded in the transportation survey. Survey activity data indicate when people are exercising, but otherwise do not provide information on metabolic level. Individuals are assigned an exercise breathing rate during time spent exercising. If the individual was at home during 11 PM – 7 AM, I use sleeping breathing rates. All other activities (e.g., shopping, employment, household chores, etc.) are assigned a light activity breathing rate. The calculated average breathing rate (units: m^3 d^{-1} person^{-1}) for the sample is 13.0, which lies between the population average estimates of 12 and 15 given by Layton (1993) and Marty et al. (2002), respectively.

Ambient concentrations

The ambient concentration assigned to an individual is determined from the individual's location at a particular time and from the output of a spatially and temporally explicit model of urban air toxics in the South Coast Air Basin. Ambient concentrations used in this study are from the CAMx air quality model (www.camx.com). The air dispersion modeling period is April 1, 1998 – March 31, 1999, and the location is the South Coast Air Basin modeling domain (Figure 6-1). CAMx is a three-dimensional

Eulerian grid model that incorporates emissions, advection, and chemical reaction. Ground-level ambient concentrations are given each hour for each 2 km × 2 km grid cell in the 120 km × 210 km domain. Details about model formulation, model validation, and the broader Multiple Air Toxics Exposure Study (MATES) are available elsewhere (ENVIRON, 2002; Morris and Jia, 2003; SCAQMD, 1999).

I chose CAMx for two reasons. First, the model output offers significantly more spatial and temporal resolution than ambient concentration data. The extant ambient monitoring network for toxics is not sufficiently detailed to offer robust answers to the issues I address in this chapter. Second, the model output covers a full year and includes important toxic pollutants, such as diesel particulate matter and benzene, in addition to a key criteria pollutant, ozone. In this work, I focus on five of the species modeled in CAMx: benzene, 1,3-butadiene, ozone, fine particulate matter emitted from diesel engines ($DPM_{2.5}$), and hevalent chromium in the form of fine particulate matter (Cr-$PM_{2.5}$). These five species incorporate different types of pollutants (primary and secondary; vehicle-dominated and point-source-dominated emissions; toxic and criteria pollutants) and have been determined to be significant contributors to the total health impact of ambient air pollution in the South Coast Air Basin (SCAQMD, 1999; US EPA, 2004). Based on preliminary work, results for the five species are representative of all 13 toxic species modeled in CAMx.

Travel survey data

Individuals' locations are from geocoded activity diaries from the Southern California Association of Government (SCAG) year-2000 transportation survey of

40,376 individuals. The travel survey incorporates an area larger than the South Coast Air

Basin modeling domain (SoCAB). I removed from the dataset the 15,188 records (38%)

for people who live or otherwise spent time outside the SoCAB, leaving 25,188 records

(62%) of people who spent 100% of their travel diary time within the SoCAB. I removed

an additional 120 records (0.5% of 25,188) that contained erroneous or missing data or

that yielded infeasible results, leaving the records for 25,068 individuals used in the

present study. Most (21,391 out of 25,068, or 85%) of the survey respondents in the data

subset that I used have one 24-hour weekday record. The rest (15%) have a 48-hour

record that includes one weekend day. Thus, there are a total of 28,745 person-days in the

dataset. The records come from 11,780 households. Diary entries start and end at 3:00

AM, the approximate time when the fewest trips occur (US DOT, 2003b). There are an

average of 4 activities per person-day.

The travel survey data were collected during three phases, corresponding to the

following approximate dates: March 1, 2000 – June 30, 2000 (17 weeks), September 1,

2001 – December 15, 2001 (15 weeks), and January 15, 2002 – June 30, 2002 (24

weeks). It is not possible to match travel survey records directly to dates from the

ambient concentration model results for two reasons. First, the survey period (year-2000

through 2002) is after the modeling period (year-1998 and 1999). Second, the survey data

provide the day-of-week and the survey phase, but not the date. To address these two

limitations, travel survey data and ambient concentration fields were matched using a

random selection process that preserves fidelity to day-of-week and time-of-year. For

example, to determine the ambient concentrations for a Tuesday travel survey record in

the second phase, I use model results for one Tuesday, chosen at random, between
August 27, 1998 and December 16, 1998.

An alternative approach to using travel survey data would be to use results from
travel demand models. These models simulate the flow of people between origins and
destinations, and predict attributes of the transportation system, such as congestion,
vehicle speeds, and public transportation usage. Such models are often used as part of a
common four-step approach for transportation demand modeling.

(1) *Trip generation*. Here, the total number of trips to be taken is estimated, based
on the number of people and the number of destinations such as shopping and
employment.

(2) *Trip distribution*. Here, the total trips estimated in step 1 are allocated
spatially. This allocation is often done based on a gravity-based model, which
assumes that the likelihood for an origin to be associated with a potential
destination is inversely proportional to the distance (in length and/or time)
between the origin and the destination. The results from this step are the
number of trips per hour for each origin-destination pair.

(3) *Mode choice*. Here, the trips modeled in step 2 are allocated to the available
modes, such as automobile, bus, train, bicycle, and walking. Typically, when
allocating a trip to a mode, a logit model is used to predict people's behaviors,
based on a comparison among the mode options of the time and financial cost
and mode availability. The results from this step are the modes associated with
each of the trips predicted in step 2.

(4)   *Network assignment*. Here, trips are assigned to the extant transportation

network. For example, trips by automobile are assigned to the network of

streets.

Travel demand models are commonly used in transportation planning, especially for

regulatory purposes. Their strength, from a research standpoint, is that they are

comprehensive and provide unique information about the transportation system. Their

disadvantages include that they are time consuming and cumbersome to run, and they

involve a number of assumptions that are difficult to test about individuals' behavior and

about the transportation network. Another disadvantage, based on the goals of the

investigation presented in this chapter, is that they describe the statistical movement of

whole population, rather than the actual behavior of specific individuals. For example, I

analyze below correlations between air pollution inhalation rate and demographic

attributes such as individuals' ethnicity and income. I would not be able to conduct this

same analysis using results from a typical transportation demand model.

Combining travel survey data with ambient concentration estimates

The method for determining the ambient concentration depends on whether a

person is traveling during a specific time. During non-travel activities, the ambient

concentration assigned to a person is the ambient concentration for the CAMx grid cell in

which they are located. During transportation activities, people may travel through

multiple grid cells. The survey provides the time and location for the origin and the

destination, but not the route traveled. I model people as moving in a straight line at a

constant speed from their origin to their destination. Assigned ambient concentrations

during travel are the concentrations in each of the CAMx grid cells the person traverses during their trip, for the duration spent in that cell. In general, real trips do not conform to this straight line, constant velocity assumption. However, the additional resources necessary to relax this assumption are not justified because of uncertainty in determining a person's true route between two locations and because of limited temporal and spatial resolution in the CAMx results (i.e., grid cells are 2 km × 2 km). The impact of this assumption on estimated population intake of air pollution is expected to be small for the pollutants studied. Time spent traveling is only 4–7% of the total day. Estimates of daily travel time include ~55 min for the US (US DOT, 2003b), ~80 min for the US and California (Klepeis et al., 2001), and ~92 minutes for California (Jenkins et al., 1992).

Figure 6-2 summarizes the SCAG travel survey data in terms of the cumulative distribution plot of distance from home. Three lines are shown in Figure 6-2, corresponding to the distributions at 3:30 AM and 3:30 PM and the daily maximum distance away from home. The two times (3:30 AM and 3:30 PM) correspond approximately to the times with the fewest and most trips: 0.1% of trips begin during 3:00 – 4:00 AM, and 8.4% of trips begin during 3:00 – 4:00 PM (US DOT, 2003b). Figure 6-2 indicates that at 3:30 AM, most people (~98%) are at home (i.e., the distance from home is approximately zero). At 3:30 PM, 58% of people are home, 22% are not at home but are less than 10 km from home, and the rest (20%) are more than 10 km from home. The line in Figure 6-2 corresponding to the maximum distance from home indicates that during an average travel diary day, 27% of people do not leave home (defined here as having a maximum distance from home of less than 1 km), 38% leave home but never travel more than 10 km from home, and the rest (35%) travel to at least one location that is more than 10 km from home.

Microenvironments

The concept of microenvironments is used to account for times when the exposure concentration is different from the nearby ambient concentration. For example, concentrations of primary vehicle emissions such as benzene tend to be higher in a vehicle than in nearby ambient air because the in-vehicle microenvironment tends to be in closer proximity to other vehicles' emissions than the mean location in an air basin. In contrast, attributable concentrations are lower indoors than in ambient air for pollutants such as ozone that are removed chemically or physically within indoor environments or as air migrates from outdoors to indoors. The attributable concentration in a microenvironment is estimated as the product of the ambient concentration and the applicable microenvironment factor.

Microenvironment factors used in this work are presented in Table 6-1. This work evaluates exposure to outdoor air pollution and therefore does not incorporate indoor sources in the microenvironment factors. I use four microenvironments: outdoor, indoors in a residence, indoors in a non-residence, and in or near motor vehicles. The exposure concentration for all pollutants in the outdoor microenvironment is the ambient concentration. Benzene and butadiene can penetrate the building envelope without significant loss. For these two gases, the time-average indoor (residential and nonresidential) concentration attributable to ambient emissions is taken to be equal to the time-average outdoor concentration, and hence the corresponding microenvironment factors are equal to 1.0.

For other species and microenvironments considered in this work, the microenvironment factor is determined stochastically, with values chosen from a distribution representing uncertainty and variability in the relationship between the ambient and the exposure concentrations. For example, benzene and butadiene are primarily emitted by vehicles and concentrations are higher in-vehicle than in ambient air averaged over a modeling grid cell. The microenvironment factor used for a specific in-vehicle activity is chosen at random from a triangular distribution, with minimum = 2, peak = 4, and maximum = 6 (Flachsbart, 1995; Flachsbart, 1999a; Rodes et al., 1998), denoted "Tri(2, 4, 6)".

The in-vehicle microenvironment factor employed in Chapter 3 for benzene and CO is also 4.0. Strictly, the interpretation of the in-vehicle microenvironment factor is slightly different in this chapter as compared to Chapter 3. Here, the in-vehicle microenvironment accounts for differences between on-road concentrations and model-predicted ambient concentration within a 2 km × 2 km grid cell. In Chapter 3, the in-vehicle multiplier accounted for differences between on-road concentrations and the general urban ambient air, as measured at monitor stations. Information is not currently available that allows distinct, robust estimates to be generated separately for these two parameters; hence, they are both taken as having a best estimate value of 4.0 in this dissertation.

Ozone concentrations tend to be lower in vehicles than in ambient air because of the close proximity to fresh nitric oxide emissions. Nitric oxide titrates ozone to oxygen and nitrogen dioxide. I use a Tri(0.15, 0.20, 0.60) distribution for the microenvironment factor for ozone in vehicles based on the empirical findings of Chan et al. (1991). Ozone

concentrations also tend to be lower in buildings than outdoors because ozone production rates are reduced in the absence of direct sunlight and because ozone reacts with indoor surfaces such as carpets, walls, and furniture (Weschler, 2000). The ozone microenvironment factor in nonresidential buildings is taken to be a Tri(0.3, 0.5, 0.8) distribution (Weschler, 2000). For residences, I use ozone microenvironment factors derived from indoor and outdoor ozone measurements taken at 126 homes in the Los Angeles area (Avol et al., 1998). I divided this dataset, which consists of 235 indoor-outdoor ratios, into two subsets: the 159 values taken during April 15 through October 15 ("summer"; range of values: 0.0% – 99.7%; mean = 34%), and the 76 measurements taken during October 15 thought April 15 ("winter"; range: 0.0% – 71.0%; mean = 11.7%). When calculating the exposure concentration, an indoor-outdoor ozone ratio is chosen at random for each residence from the appropriate set of indoor-outdoor ratios, based on the travel diary date.

The microenvironment factors used in this work for PM ($DPM_{2.5}$ and $Cr\text{-}PM_{2.5}$) in nonresidential buildings are 0.63 +/- 0.11 for $DPM_{2.5}$ and 0.72 +/- 0.053 for $Cr\text{-}PM_{2.5}$ (Riley et al., 2002). These distributions are derived from a mass-balance model that accounts for infiltration, ventilation, makeup and recirculation airflow, and particle filtration and deposition for generic $PM_{2.5}$ (for $Cr\text{-}PM_{2.5}$) and for elemental carbon (for $DPM_{2.5}$) (Riley et al., 2002). These distributions are treated as normal, with the indicated means and standard deviations, and with a maximum value of 1.0. (Truncating values above 1.0 does not significantly impact the distribution: the probably of generating a value greater than 1.0 for the indicated means and standard deviations is less than 0.04%.) The in-vehicle microenvironment factor for $DPM_{2.5}$ is modeled as a Tri(2,3,6)

distribution. To evaluate the microenvironment factor for $PM_{2.5}$ in residences, I employed

a mass-balance modeling approach, as shown in Equation 6-2 (Burke et al., 2001):

$$\gamma_{residence, PM2.5} = \frac{Pa}{a+k}.$$

(6-2)

Here, $\gamma_{residence, PM2.5}$ is the microenvironment factor for ambient $PM_{2.5}$ (i.e., $DPM_{2.5}$ or Cr-

$PM_{2.5}$) in residential buildings (dimensionless), P is the penetration efficiency of $PM_{2.5}$

(dimensionless), a is the air exchange rate (h^{-1}), and k is the particle removal rate by

means of deposition or filtration indoors (h^{-1}). Penetration efficiency is estimated to be

1.0 for $PM_{2.5}$, based on the work of Ozkaynak et al. (1996) and Riley et al. (2002).

Measured deposition rates have been reported to be 0.39 ± 0.16 h^{-1} (normal distribution;

minimum value = 0.0) for $PM_{2.5}$ (Ozkaynak et al., 1996). Values of the geometric mean

(GM) and geometric standard deviation (GSD) for air-exchange rates in the Los Angeles

area, based on a dataset of 1,444 measurements (Wilson et al., 1996), are GM = 0.55 h^{-1},

GSD = 1.97 during the six winter months, and GM = 1.05 h^{-1}, GSD = 2.39 during the six

summer months. For each residence, values for a and k were randomly generated from

these distributions, and then the residence microenvironment factor for $DPM_{2.5}$ and Cr-

$PM_{2.5}$ were calculated using Equation 6-2, with a maximum value of 1.0. (Truncating

values above 1.0 does not significantly impact the distribution: the probability of

generating a value greater than 1.0 is ~0.2% for both summer and winter.) Arithmetic

mean and standard deviations for the resulting values for $\gamma_{residence, PM2.5}$ are 0.61 ± 0.06 in

winter and 0.71 ± 0.07 in summer. These values are consistent with the value of 67%

used in Table 2A-1 (Chapter 2 Appendix) as the indoor-outdoor ratio for ambient $PM_{2.5}$ (Fruin et al., 2004), and with the "back-of-the-envelope" estimate in Note (f) for Table 2A-1, derived from values reported by Riley et al. (2002), that this ratio is 68%.

Results and discussion

Inhalation rates

Model results yield the inhalation intake rate ($\mu g \ d^{-1}$) for each person in the dataset for each of the five species (benzene, butadiene, ozone, $DPM_{2.5}$, and $Cr-PM_{2.5}$). Table 6-2 provides statistics summarizing the inhalation rates. This table also presents statistics summarizing other model parameters, such as individuals' breathing rates and daily travel patterns.

Intake rates presented in this work only account for inhalation of air pollution of ambient origin. Indoor emissions such as environmental tobacco smoke (ETS) also contribute to total population intake rates for certain pollutants, such as benzene and butadiene (Fruin et al., 2001; Nazaroff and Singer, 2004). Unlike in Chapter 3, emissions to attached garages, which can be an important contributor to population inhalation of vehicle emissions (Fruin et al., 2001; Graham et al., 2004; Thomas et al., 1993), are not included here. The TEAM series of studies on benzene found that for non-smokers, vehicle emissions contribute the majority of total exposure, while ETS only contributes only ~ 10% (Wallace, 1996). For smokers, cigarettes contribute ~ 90% of benzene exposures. TEAM results suggest that smokers bear roughly half of the total benzene exposure in the US (Wallace, 1996). Ozone is emitted by printers and photocopiers

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(Tuomi et al., 2000) and by some indoor air "purifying" devices (Phillips et al., 1999). While the model developed and implemented in this chapter does not account for indoor emissions, it does account for inhalation indoors of ambient pollution.

Mean intake rates (μg d^{-1}) for the five pollutants are 67 for benzene, 7.3 for butadiene, 47 for DPM$_{2.5}$, 0.0016 for CrPM$_{2.5}$, and 120 for ozone. To explore the reliability of these results, and as a "reality check" to determine whether other investigations corroborate this one, I analyzed modeled average ambient concentrations from two independent investigations: the MATES II study of the South Coast Air Basin, which uses a 3-D Eulerian air dispersion model (SCAQMD, 1999) and values for California in the EPA's National-scale Air Toxics Assessment (NATA), which uses a Gaussian plume model (US EPA, 2004). To estimate intake rates based on model output from these two studies, I used a breathing rate of 13 m^3 d^{-1}, and I used modeled ambient concentrations as a proxy for exposure concentrations. The results of this analysis yields the following per capita intake rates (μg d^{-1}): for benzene: 41 (MATES) and 20 (NATA); for butadiene: 4.4 (MATES) and 0.9 (NATA); for DPM$_{2.5}$: 46 (MATES) and 33 (NATA); and for CrPM$_{2.5}$: 0.0031 (MATES). (There were not estimates for hexavalent chromium in NATA. Neither NATA nor MATES evaluated ozone.) The results from my analyses are within a factor of two or less of the MATES results. The NATA results are significantly less than those reported here and in MATES. One reason for this difference is that MATES and this study investigate a highly urbanized environment (the South Coast), whereas NATA results incorporate all of California, including both urban and rural areas. Another reason is that, based on model-measurement comparisons, NATA results underestimate ambient concentration; for gaseous species, modeled concentrations

are too low by ~40% on average (US EPA, 2004). As a third "reality check", the results

presented in Chapter 3 can generate an estimate of population intake of benzene in the

South Coast. Based on values given in Chapter 3 for benzene (vehicle emissions = 16

tonnes d^{-1}; intake fraction = 48 per million; population = 15 million), mean per capita

intake of vehicular benzene is 53 µg d^{-1}. The method I employ in Chapter 3 to estimate

this intake fraction value incorporates ambient concentration data for benzene in the

South Coast, as measured at ambient monitoring stations. As described in Chapter 3,

vehicle benzene emissions represent 70% of total benzene emissions. Assuming that the

intake fraction for the remaining ambient benzene emissions is also approximately 48 per

million, mean per capita benzene intake is estimated by this approach to be ~76 µg d^{-1},

which is within 12% of the value reported in this chapter (67 µg d^{-1}). Thus, the results

presented in this chapter are reasonably consistent with the three previous investigations

considered here. In the Uncertainty section (below), I compare measured and CAMx-

modeled ambient concentrations.

Figure 6-3 presents cumulative distribution plots for the five species. A lognormal

distribution would appear as a straight line when presented in these plots. Some of the

distributions in these plots are approximately linear, indicating a nearly lognormal

distribution. Statistical tests of the data (Kolmogorov-Smirnov and Cramer-von Mises)

reject the strict assumption of lognormality for all five plots. Because of the large size of

the dataset, such tests are more sensitive to deviations from lognormality than is desired

for some applications.

Health risks

There are a number of health risks expected from inhaling the pollutants modeled in this work. Diesel $PM_{2.5}$ is believed to cause lung cancer; benzene is a known human carcinogen, and causes leukemia; ozone exposures can cause asthma attacks and bronchitis. Considering the combined impact of these five species considered, significant health impacts are expected at these exposure levels (SCAQMD, 1999).

Diesel $PM_{2.5}$ is believed to cause a majority of the air pollution lung cancer health risk in the South Coast (SCAQMD, 1999). Based on a chronic exposure mortality risk factor of 3×10^{-6} ($\mu g\ d^{-1})^{-1}$ (Lloyd and Cackette, 2001), the average intake of diesel $PM_{2.5}$ (47 $\mu g\ d^{-1}$), is estimated to cause approximately 2,100 deaths annually among the 15 million residents of the South Coast. My estimates for diesel PM inhalation rates are ~2 times higher than extant statewide estimates for California (CARB, 2000c; Fruin et al., 2004). For example, Fruin et al. (2004) estimated an average diesel PM exposure concentration of $1.7 – 2.7\ \mu g\ m^3$, or $22 – 35\ \mu g\ d^{-1}$ if one assumes a population average breathing rate of $13\ m^3\ d^{-1}$. There are several factors that contribute to the difference between the results in Fruin et al. (2004) and those presented here. (1) This study focuses on a single urban area rather than the whole state. Exposures are expected to be higher in the SoCAB than elsewhere in the state for reasons discussed in Chapter 4. (2) I include personal mobility and time-varying breathing rates, which increase intake rates by ~10% each. (3) My method incorporates intraurban spatial variability in population density rather than using one average ambient concentration for the air basin.

Evaluating three common exposure analysis assumptions

Three assumptions commonly made in exposure assessments – for example, when using ambient concentrations as a surrogate for exposure concentrations – are to ignore the influence on exposures of (1) population mobility, (2) diurnal variability in breathing rates, and (3) microenvironments. The approach employed in this chapter allows me to explicitly evaluate the consequences of these three assumptions. To do so, I estimated intake rates using five parallel approaches: (1) incorporating the three common assumptions listed above ("base case"); (2) accounting for people's movement throughout the air basin but not for variability in breathing rates or for microenvironments ("mobile"); (3) accounting for microenvironments but not mobility or breathing rate variability ("microenvironments"); (4) accounting for diurnal variability in breathing rates, based on activity data, but not microenvironments or variability ("breathing rates"); and, (5) accounting for breathing rate variability, microenvironments, and mobility ("all"). The default approach for results presented in this chapter is "all". Figure 6-4 compares the results from these analyses in terms of the mean change in intake for each individual and for each chemical relative to the base case. For example, for butadiene, incorporating microenvironments in the analysis increases an average individual's estimated intake rate by an average of 30%. This increase is attributable to the amplified exposure concentration in the in-vehicle microenvironment. For chromium $PM_{2.5}$, incorporating microenvironments reduces individuals' intake rates by 31% on average. This decrease is attributable to reduced concentrations of ambient particles in the indoor microenvironment. For all five pollutants, accounting for mobility increases

the mean intake rate, indicating that, on average, people travel to locations where the ambient air pollution concentration is higher than at their home.

The three factors explored here (mobility, microenvironments, and temporally varying breathing rates) exhibit temporal correlations. For example, typical diurnal trends are that mobility rates, breathing rates, and the portion of time in a non-residence microenvironment are higher during the day than during the night. Because of these temporal correlations, the combined effect of the three factors is not, in general, equal to the direct sum of the effects of each factor taken individually (see Figure 6-4). The three factors, taken together, impact the individual's estimated intake rates by an average of 4 - 69%. Accounting for the three factors together reduces intake rates by 59% for ozone, increases intake rates by 40% and 69%, respectively, for benzene and butadiene, and causes only small changes (~5% or less) in the intake rates for chromium $PM_{2.5}$ and diesel $PM_{2.5}$.

Figure 6-4 gives the *mean* influence among individuals of each refinement on the intake rate. The influence of each refinement on the estimated intake rate for specific individuals varies and – for some factors – can either increase or decrease intake rates. Accounting for mobility increases the estimated intake rate for individuals who travel to areas whose concentrations are higher than at home, and it decreases the estimated intake rate for individuals who travel to areas whose concentrations are lower than at home. For most people (~90% among the five pollutants), accounting for mobility changes the estimated intake rate by 30% or less. The remaining people (~10%), for whom accounting for mobility changes the estimated intake by more than 30%, are more mobile than the average individual. For example, for $DPM_{2.5}$, the average daily distance traveled

is three times greater for the latter group than for the former group (68 km versus 21 km), and the average daily maximum distance from home is four times greater for the latter group than for the former group (33 km versus 8 km). These findings indicate, as expected, that mobility influences a person's inhalation intake of air pollution more for people who travel a lot during the day than for people who travel only a little.

The influence of mobility on inhalation rate varies among subpopulations

Results presented in this chapter suggest that mobility leads to only moderate changes in population-average exposure concentrations. Not surprisingly, the effect of mobility on intake rates is higher for highly mobile individuals than for the general population. Mobility is correlated with demographic attributes: on average, low-income and minority (non-Caucasian) individuals tend to travel less than the population average.

To explore whether the importance of mobility varies among subpopulations, I completed the analyses yielding Figure 6-4, but restricting the sample population first to individuals in households with an income of less than $50,000, then to Hispanic individuals in households with an income of less than $50,000. In both cases, the impact of mobility on exposure concentration is less for the subpopulation than for the whole population. For example, for the whole population, the highest impact of mobility on mean intake rate among the five pollutants is 17% (butadiene; see Figure 6-4). The highest impact of mobility on mean intake rate is 7% for household income <$50,000, and 4% for Hispanic individuals with household income <$50,000. These two subpopulations are less mobile, on average, than the population as a whole. Consequentially, the importance of mobility is reduced for these subpopulations relative

to the whole population. To elaborate, average values for distance traveled (DT) and maximum distance from home (MDFH) for the whole population (n=28,745) are (units: km) 29.5 and 12.2, respectively. Using these two metrics, individuals with household income <$50,000 (n=12,309) are about 21% less mobile than the whole population: DT and MDFH values for this group are 23.3 and 9.6, respectively. Similarly, Hispanic individuals with household income <$50,000 (n=5065) are about 33% less mobile than the whole population: DT and MDFH values for this group are 19.5 and 8.2, respectively.

Uncertainty

In this section, I address uncertainty in the four main inputs: breathing rate, people's location, microenvironmental concentrations, and ambient concentrations. I use metabolic activity rate studies to estimate breathing rate from age, gender, and exercising status (Layton, 1993). As discussed in Chapter 3, population average breathing rates are ~20% lower using the values in Layton (1993) than using common alternatives (Marty et al., 2002; OEHHA, 1996; US EPA, 1997).

The diurnal breathing rate profile for this investigation is presented in Figure 6-5. To my knowledge, estimates of this population-average diurnal profile have not been published by other researchers. In Chapter 3, I estimated this profile based on data in Layton (1993) and my own estimates of the likelihood of each activity occurring in each hour. In this chapter I estimated activity level for each individual and in each time based on the survey data. Enough information is provided in the survey data to yield a reasonable estimate of the diurnal breathing rate profile (especially in light of the limited information available about this topic), but enough information is missing from the

survey data that these estimates should be considered approximate rather than definitive. For example, survey responses indicate when a person exercises, but not his or her bedtime and waking time. Based on the results resented in this chapter, generating a more robust estimate of the diurnal profile of population breathing rate would be worthwhile.

Travel diaries are likely to underestimate mobility because of trip underreporting, which is estimated at ~10 – 15% of trips (Clarke et al., 1981), and because comparatively mobile individuals may be underrepresented in survey data since they are harder to contact (Schafer, 2000). As discussed below, future studies using global positioning systems (GPS) may significantly reduce uncertainty associated with the location component of exposure models.

The indoor-outdoor concentration ratio for ambient pollutants varies among microenvironments and pollutants, from near-zero to 100% (Avol et al., 1998; Riley et al., 2002). The Monte Carlo approach I used to estimate exposures in microenvironments incorporates available information about variability in the indoor-outdoor ratio. Particle decay rate values, k, used in this work were determined by Ozkaynak et al. (1996) from cotemporal measurements indoors and nearby outdoors, assuming particle penetration, P, equals unity. Empirical (Liu and Nazaroff, 2003; Long et al., 2001) and modeling (Liu and Nazaroff, 2001) studies of $PM_{2.5}$ suggest P values less than unity, typically between 0.6 and 1.0. However, to the extent that P is less than unity, this fact is accounted for in k values that are determined experimentally, e.g., using Equation 6-2 (Kopperud et al., 2004). That is, when analyzing cotemporal indoor and nearby outdoor measurements to determine P and k, allowing $P \leq 1$ rather than assuming $P = 1$ yields lower values of P and higher values of k. Kopperud et al. (2004) found that the fit of the curve, and the

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utility of the resulting model for estimating indoor/outdoor relationships, is not significantly diminished by assuming $P = 1$. The indoor-outdoor concentration ratios for ambient pollutants employed here (average values: 0.61 ± 0.06 in summer, 0.74 ± 0.07 in winter) are broadly consistent with empirical evidence: reported values for elemental carbon are in the range 0.5 (LaRosa et al., 2002) to $0.8 - 0.9$ (Geller et al., 2002; Na and Cocker, 2005).

Uncertainty in ambient concentration estimates varies among the five pollutants studied (ENVIRON, 2002). Modeled benzene concentrations have a near-zero bias, whereas modeled butadiene concentrations underestimated measured concentrations by ~40%. Modeled hexavalent chromium concentrations underestimate measurements by a factor of ~4, but it is unclear whether this difference is attributable to errors in measurements, in the air dispersion model, or in the emission inventory (ENVIRON, 2002). Diesel PM is a complex mixture of species. Consequentially, direct measurement of ambient diesel PM is not possible. Because approximately two-thirds of ambient elemental carbon in the SoCAB is attributed to diesel emissions (SCAQMD, 1999), elemental carbon serves as a reasonable proxy for $DPM_{2.5}$ when assessing model performance (ENVIRON, 2002). Employing this approach, modeled $DPM_{2.5}$ concentrations are estimated to be ~15% higher than ambient concentrations (ENVIRON, 2002). CAMx model performance evaluation is not available for ozone (Morris, 2004). In Table 6-3, I present basinwide annual average concentrations for the five species considered in this chapter. I calculate these average concentrations from the CAMx model output that is used as input for the inhalation model presented in this chapter.

Eulerian air dispersion models such as CAMx implicitly assume that concentrations are uniform throughout each grid cell. When considering concentrations in a grid cell that are attributable to emissions in the same grid cell, this spatial smoothing of sub-grid processes can cause near-source concentrations to be underestimated.

Reported inhalation intake rate estimates for the ~25,000 people surveyed are not necessarily representative of intake rates by the ~15 million people in the South Coast Air Basin. For example, relative to US Census data for the Los Angeles Metropolitan Statistical Area, survey respondents have a higher proportion of Whites (51% for the survey versus 22% for the Los Angeles MSA), a lower proportion of Hispanics (27% versus 43%), Asian/Pacific Islanders (6% versus 11%), and people who listed their ethnicity as "other" (3% versus 17%), and a comparable portion of African-Americans (7% for both the survey and the MSA). Development of appropriate weighting factors for each individual, which would allow "scale-up" from the survey population to the whole population, is a challenging task that is beyond the scope of the present work. Such weighting factors would incorporate, for the survey population and the whole population, attributes that correlate with intake, such as proximity to emissions, mobility, ethnicity, and weekend versus weekday differences.

Weekend/weekday differences in inhalation rate

Most (85%) of the survey respondents in this investigation recorded only weekday activities. Because air pollutant concentrations in Southern California (Marr and Harley, 2002; Qin et al., 2004) and human activities (Klepeis et al., 2001) exhibit weekend/weekday patterns, the results presented here are not necessarily representative

of weekend exposures. As expected, modeled exposures vary between weekend and weekday. Average modeled ozone intake is about 7% higher on weekends than on weekdays, which is consistent with ambient ozone concentrations being higher on weekends than weekdays (Marr and Harley, 2002). Average modeled exposures for the remaining four pollutants are lower on weekends than on weekdays, which is consistent with lower emissions for these pollutants owing to reduced commercial and personal activity. The weekend/weekday intake rate difference is about 9% for benzene and butadiene, and about 23% for $DPM_{2.5}$ and $CrPM_{2.5}$. Consistent with values reported elsewhere (Marr et al., 2002), survey respondents' average travel distance is about 10% less on weekends than weekdays.

Inhalation rate by ethnic and income group

Intake rates are correlated with demographic attributes such as race and income. Figures 6-6 and 6-7 present the 50th and 90th percentile intake rates, respectively, as a function of subpopulation, for four ethnic groups and two income levels. These plots exclude the 14% of respondents who did not provide their ethnicity or household income and the 3% of respondents who listed "other" as their ethnicity. Two types of differences are immediately apparent in this figure: among demographic groups (ethnicity and income) and among pollutants (mainly, ozone versus the other four pollutants). For the four primary pollutants, median and 90th percentile intake rates are lower for Whites, and higher for Hispanics, African-Americans, and Asians/Pacific Islanders, than for the population as a whole. Ozone inhalation intake rates exhibit the opposite trend. Results in Figures 6-6 and 6-7 indicate that, for the four primary pollutants, individuals in higher

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income households have lower intake rates than individuals in lower income households. For ozone, the reverse is true.

Figures 6-8 and 6-9 elaborate on the results in Figures 6-6 and 6-7. Whereas Figures 6-6 and 6-7 present differences among ethnic and income groups at the 50th and 90th percentiles, Figures 6-8 and 6-9 present these differences throughout the distribution. The interpretation of the values presented in Figures 6-8 and 6-9 is similar to in Figures 6-6 and 6-7. For example, in Figure 6-8, the plot presenting distributions for Whites has a y-axis value of 82% for ozone at the 5th percentile x-axis value. This value indicates that comparing the ozone inhalation rate distribution for Whites and for the whole population, the 5th percentile value for whites is 82% higher than the 5th percentile value for the whole population. The main trends in Figures 6-8 and 6-9 are the same as in Figures 6-6 and 6-7. For primary pollutants, inhalation rates are lower for Whites than non-Whites and for high-income than for low-income. For ozone, the reverse pattern holds: inhalation rates are higher for Whites than non-Whites and for high-income than for low-income. Two exceptions to this broad trend are (1) that Asian/Pacific Islander inhalation rates for ozone are roughly equal to the population values throughout much of the distribution, and (2) that African-American inhalation rates for benzene are only slightly below the population values at the upper end of the distribution.

The demographic make-up differs between the overall survey population and the survey subpopulation with above-average inhalation rates. For example, the percent of survey respondents that are White is 52%. In contrast, the upper quartile for benzene and diesel PM inhalation rates are only 41% White and 39% White, respectively.

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In all likelihood, the main factor underlying these trends in pollution inhalation rate by ethnic and income group is proximity to emission sources. On average, non-Whites are in closer proximity to emission sources than Whites are, and therefore non-Whites have higher exposures and higher intake rates for primary pollutants than do Whites (Gunier et al., 2003; Houston et al., 2004; Pastor et al., 2004). Proximity to sources would also explain why the ozone intake rate is higher for Whites than for non-Whites: Because advection moves air masses during the time required for precursor emissions to form ozone, peak ozone concentrations are *not* proximate to emission sources but rather occur in downwind locations. Similarly, residents in lower-income households are closer to emission sources, on average, than residents of higher income households (Gunier et al., 2003; Houston et al., 2004; Pastor et al., 2004), which may account for the observed differences in inhalation intake rates between income levels for both primary and secondary pollutants.

Although not accounted for in my model, a second causal factor that could also cause correlations between income level and ambient air pollution intake rates is household air-exchange rate. Older, "leakier" residences (i.e., residential buildings with a higher-than-average air-exchange rate) are more likely to be occupied by low-income than high-income families. The higher-than-average air exchange rates in older buildings cause them to offer less protection against particles and ozone than do newer and well-maintained buildings (Equation 6-2). All else being equal, the inhalation intake rate for particles and ozone would be higher in a leaky building than in a tight building. Similarly, there is likely to be a correlation between household income and use of air conditioning versus natural ventilation (i.e., open windows). To my knowledge, the

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literature correlating exposures with demographic attributes such as race and income has not yet quantified the impact of this issue.

A comparison between Figure 6-6 and Figure 6-7 indicates that, on average and for the case study considered in this work, median and 90th percentile exposure levels differ more among ethnic groups than between high- and low-income households. (The value $50,000 is used here as the divider between "high" and "low" household annual income levels because this value reflects a specific question included as part of the transportation survey.) Variability in intake rates among the population, and correlations between intake rates and demographic attributes such as race and income, are important aspects of air quality. Environmental policy seeks not only to reduce the population-average health risk attributable to air pollution, but also to ensure that specific subpopulations are not unduly burdened, relative to the population as a whole. Such concerns are components of the broader theme of environmental justice.

The results presented in this work are generally consistent with those presented elsewhere. Several studies have reported higher exposures for low-income groups and non-Whites for primary pollutants (Brown, 1995; Schweitzer and Valenzuela, 2004), including populations in California in general (Gunier et al., 2003; Pastor et al., 2004) and specifically in Southern California (Morello-Frosch et al., 2001; Morello-Frosch et al., 2002). Consistent with the findings in this chapter, the proportion of upper income households and Whites are observed to be higher in high-ozone areas downwind of New York City and Philadelphia than in the urban areas where ozone precursors are emitted (Liu, 1996). Ozone levels in the SoCAB were found to be positively correlated with the percentage of Whites in the community (Brajer and Hall, 2005; Korc, 1996). However, in

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contrast with the results presented in this chapter, others have found that ozone levels are inversely correlated with income (Brajer and Hall, 2005; Korc, 1996).

All else being equal, the ambient concentration difference between near-source and not-near-source would be larger for rapidly decaying emissions (e.g., primary ultrafine particles) than for slowly decaying pollutants (e.g., benzene). Based on differences among ethnic and income groups in proximity to emissions, I would expect that the difference in inhalation intake rates between Whites and non-Whites would be greater for rapidly decaying emissions of primary pollutants than for slowly decaying emissions.

The results support this expectation. For example, based on rate constants given in Table 3-2, the characteristic lifetimes for benzene and butadiene are 20 d and 6 h, respectively. Butadiene may be considered to be a modestly reactive (but not highly reactive) pollutant; benzene is nearly nonreactive. On the time scale of air in the South Coast Air Basin, between ½ and ¾ of emitted butadiene decays, but only $1 - 3\%$ of benzene decays. The microenvironment factors (Table 6-1) are identical for benzene and butadiene. Differences in inhalation rates among ethnic and income groups are larger for butadiene than for benzene. The difference in median inhalation rate between Whites and the whole population is 13% for benzene versus 21% for butadiene. The difference in median inhalation rate between individuals in household income above $50,000 and the whole population is 3.2% for benzene versus 5.8% for butadiene. Similar comparisons at the 90th, rather than 50th, percentile yield similar results. These values support the hypothesis that differences among ethnic and income groups would be greater for rapidly

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decaying pollutants such as primary ultrafine particles than for the primary species evaluated in this chapter.

Future research

Estimating the importance of mobility requires understanding temporal and spatial variability in pollutant concentrations and in people's locations. Increasing the spatial and temporal precision of ambient concentration estimates may require considerable effort by air quality researchers; the financial costs associated with these advances appear unlikely to drop significantly in the near future. In contrast, future research may be able to estimate people's locations to greater accuracy and precision than in this study. Global Positioning System (GPS) costs have dropped rapidly in recent years, as evidenced by the fact that the U.S. Federal Communications Commission requires that cellular phone companies be able to locate most calls to within 50 or 100 m for 911 Emergency Response purposes (www.fcc.gov/911/enhanced). If privacy concerns can be addressed, there exists the potential of inexpensive, widespread deployment of GPS devices to record volunteers' time-location patterns for air pollution exposure research purposes. Coupling GPS devices with personal sampling equipment could generate a rich air pollutant exposure dataset (Phillips et al., 2001).

Conclusions

The use of census data, or other home-based data, is common in exposure assessment and environmental epidemiology. This chapter investigates the question of

whether this assumption is appropriate. I conduct a mobility-based exposure assessment for ~29,000 individuals in California's South Coast Air Basin. I estimate inhalation intake rate for five pollutants: benzene, butadiene, hexavalent chromium $PM_{2.5}$, diesel $PM_{2.5}$, and ozone. Accounting for mobility increases the population mean intake rate by 5 – 30% among the five pollutants. The combined effect on estimated mean intake rate of mobility, temporally varying breathing rates, and microenvironments is between 4% and 93% for the five pollutants considered here.

Median intake rates vary among demographic groups for the two demographic attributes studied here (ethnicity and income). For the four primary pollutants studied, median intake rates are higher for non-White individuals than for White individuals, and higher for individuals in households with less than $50,000 income than for houses with more than $50,000 income. For ozone, the reverse is true. For the case study considered in this chapter, differences in median intake rates among subpopulations are greater when considering ethnicity than when considering income category.

As highlighted in Chapters 3 and 4, the South Coast is an important locale for studying air pollution exposure, but its attributes such as size, population, and meteorology are not representative of other US urban areas. Investigations of other urban areas would usefully refine and extend the results presented in this chapter.

My expectations for how findings in other US urban areas would differ from the results presented in this chapter for the South Coast are as follows. Relative to the South Coast, I would expect that in a monocentric urban area, a greater fraction of commuters would exhibit the stereotypical suburban-downtown commute pattern; that the emissions and ambient concentration gradient between downtown and suburbs would be steeper and

more persistent during the day; that ethnic and income groups would be more spatially segregated (i.e., the spatial autocorrelation would be higher). As a result of these differences, relative to the South Coast, I would expect that in a monocentric urban area, mobility would be more important when estimating population inhalation rate, and inhalation intake rates would differ more among demographic groups. While urban areas are increasingly transitioning from monocentric to polycentric, my expectation is that most urban areas in the US lag behind the South Coast in the degree to which this shift in urban form has already occurred. Thus, comparing the South Coast to a hypothetical monocentric urban area is a reasonable basis for generating hypotheses about how the results in the South Coast would differ from similar results in other urban areas.

Table 6-1: Summary of microenvironment factors[a]

	in-vehicle	indoor, residence	indoor, other
Benzene	Tri(2,4,6)[b]	1[c]	1[c]
Butadiene	Tri(2,4,6)[b]	1[c]	1[c]
Chromium PM$_{2.5}$	1[c]	Using Equation 6-2, $P = 1$, $k = 0.39$ +/- 0.16 h^{-1}, and geometric means (GM) and geometric standards deviations (GSD) for a are GM = 0.55 h^{-1}, GSD = 1.97 in winter and GM = 1.05 h^{-1}, GSD = 2.39 in summer [d]	0.72 +/- 0.053 [e]
Diesel PM$_{2.5}$	Tri(2,3,6)[f]	Same as for chromium PM$_{2.5}$	0.63 +/- 0.11 [e]
Ozone	Tri(0.15,0.20,0.60)[g]	Randomly select a value from an empirical dataset, based on the season (summer/winter)[h]	Tri(0.3,0.5,0.8)[i]

[a] This table lists three of the four microenvironments employed in this chapter. The factor for the fourth microenvironment, outdoors, is 1.0 by definition.

[b] Flachsbart (1995), Flachsbart (1999a), Rodes et al. (1998), Chapter 3. "Tri(2,4,6)" indicates a triangular probability distribution; the lowest value of the distribution is 2; the most common value is 4; the maximum value is 6.

[c] The time-average concentration for a nonreactive pollutant such as benzene is the same indoors as outdoors. While butadiene has a moderate reactivity on the time scale of air in an urban area, for processes such as migration of outdoor air to indoor environments, it can be considered as nonreactive.

[d] Burke et al. (2001), Ozkaynak et al. (1996), Wilson et al. (1996). The parameter k is assumed to have a normal distribution with indicated mean and standard deviation, and with a lower bound limit of zero. The parameter a is assumed to have a lognormal distribution. Derived values for the microenvironment factor have an upper bound limit of 1.0.

[e] Riley et al. (2002). This microenvironment factor is taken as a normal distribution with indicated mean and standard deviation, and with an upper bound limit of 1.0.

[f] CARB (2004), Flachsbart (1995), Flachsbart (1999a), Rodes et al. (1998), Chapter 3.

[g] Chan et al. (1991).

[h] Avol et al. (1998).

[i] Weschler (2000). See also note (b).

Table 6-2: Summary statistics[a] for time-location-activity survey data and for inhalation intake rate[b] for the five chemicals studied in this chapter

Variable		Percentile					Mean	Std	GM	GSD
		10th	25th	50th	75th	90th				
Number of household vehicles		1	1	2	2	3	2.0	1.1	1.9	1.6
Survey respondent's age	(y)	7	19	37	54	70	38	23	28	2.5
Breathing rate	$(m^3 d^{-1})$	9.46	10.5	12.1	15.0	16.3	13.1	4.4	12.6	1.3
Total distanced traveled	$(km d^{-1})$	0	0	13	38	79	29	46	23	3.1
Maximum distance from home	(km)	0	0	5	15	33	12	18	9.3	3.1
Inhalation intake rate										
Ozone	$(\mu g d^{-1})$	11	30	68	160	280	120	170	52	6.2
Benzene	$(\mu g d^{-1})$	19	30	53	89	130	67	54	51	2.1
Butadiene	$(\mu g d^{-1})$	0.80	1.9	5.1	10	16	7.3	7.6	4.1	3.4
Diesel $PM_{2.5}$	$(\mu g d^{-1})$	8.8	15	30	63	110	47	51	30	2.6
Chromium $PM_{2.5}$	$(ng d^{-1})$	0.18	0.38	0.87	1.9	3.7	1.6	2.6	0.83	3.4

(a) Abbreviations used in this table are Std for standard deviation, GM for geometric mean, and GSD for geometric standard deviation.

(b) Values estimate inhalation intake of pollutants of ambient origin. Indoor emissions are excluded.

Table 6-3: Basin-wide annual average ground-level ambient concentrations for the five species studied in this chapter

	Mean concentration[a]	Mean intake-relevant exposure concentration[b]	Ratio of mean intake-relevant exposure concentration to mean ambient concentration
Benzene (ppt)	670	1600	2.4
Butadiene (ppt)	65	240	3.7
Chromium $PM_{2.5}$ ($\mu g\ m^3$)	74×10^{-6}	120×10^{-6}	1.6
Diesel $PM_{2.5}$ ($\mu g\ m^3$)	2.1	3.6	1.7
Ozone (ppb)	28	4.5	0.16

(a) Based on CAMx model output for April 1, 1998 through March 31, 1999. The South Coast Air Basin Modeling Domain is 25,200 km^2 (210 km × 120 km), and contains 6,300 grid cells of size 2 km × 2 km.

(b) Intake-relevant concentration is the concentration that, when multiplied by the mean breathing rate (here, 13.1 $m^3\ d^{-1}$), yields the mean intake rate (given in Table 6-2).

Figure 6-1: Map of California, indicating the South Coast Air Basin modeling domain.
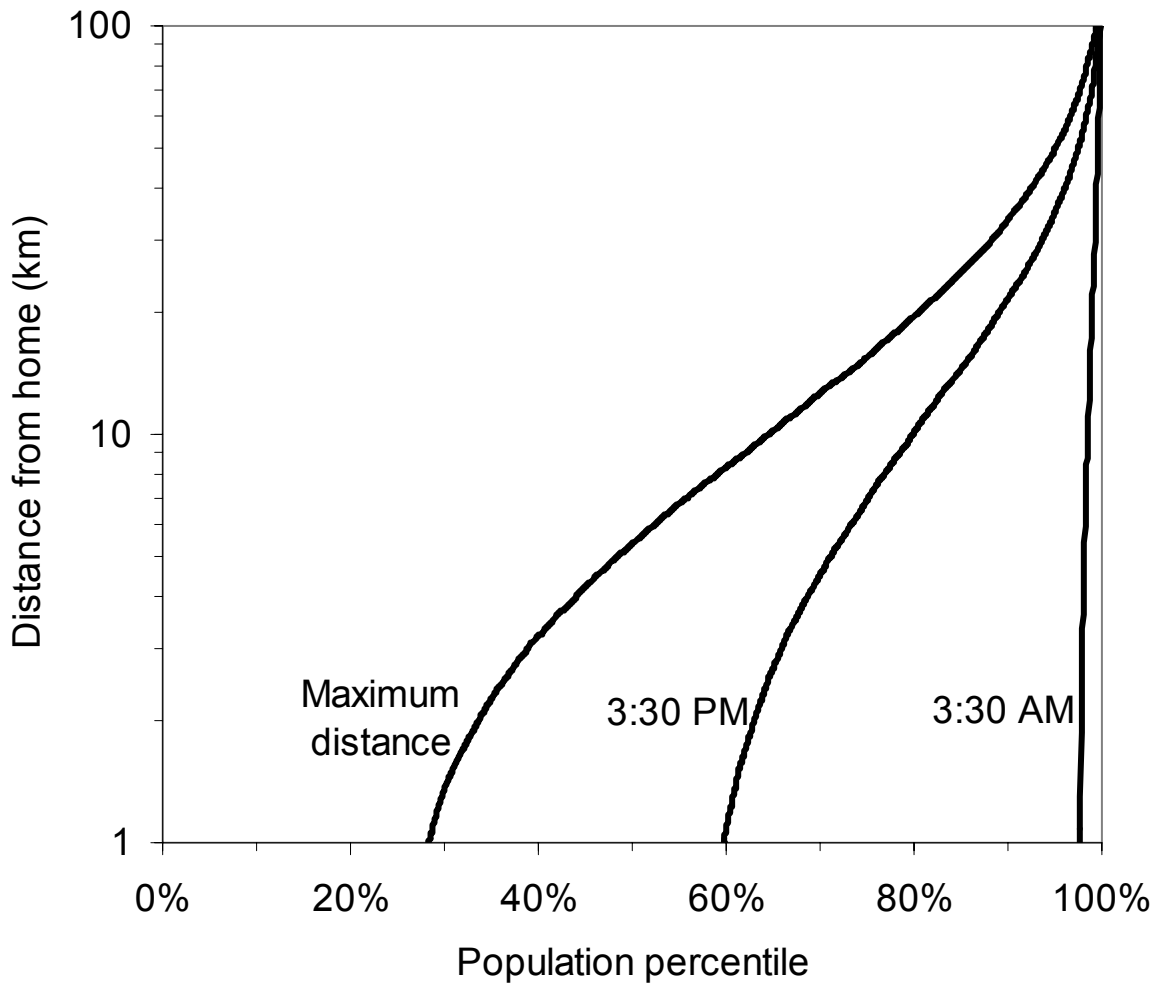
Census tracts are also shown on this map.

Figure 6-2: Cumulative distribution plot of distance from home, based on the South Coast Association of Governments travel survey data, for the 28,745 person-days simulated. Three distributions are shown: at 3:30 AM, at 3:30 PM, and the daily maximum distance away from home. For example, at 3:30 PM, 58% of people are home (defined here as being less than 1 km from home), 22% are not at home but are less than 10 km from home, and the rest (20%) are more than 10 km from home.
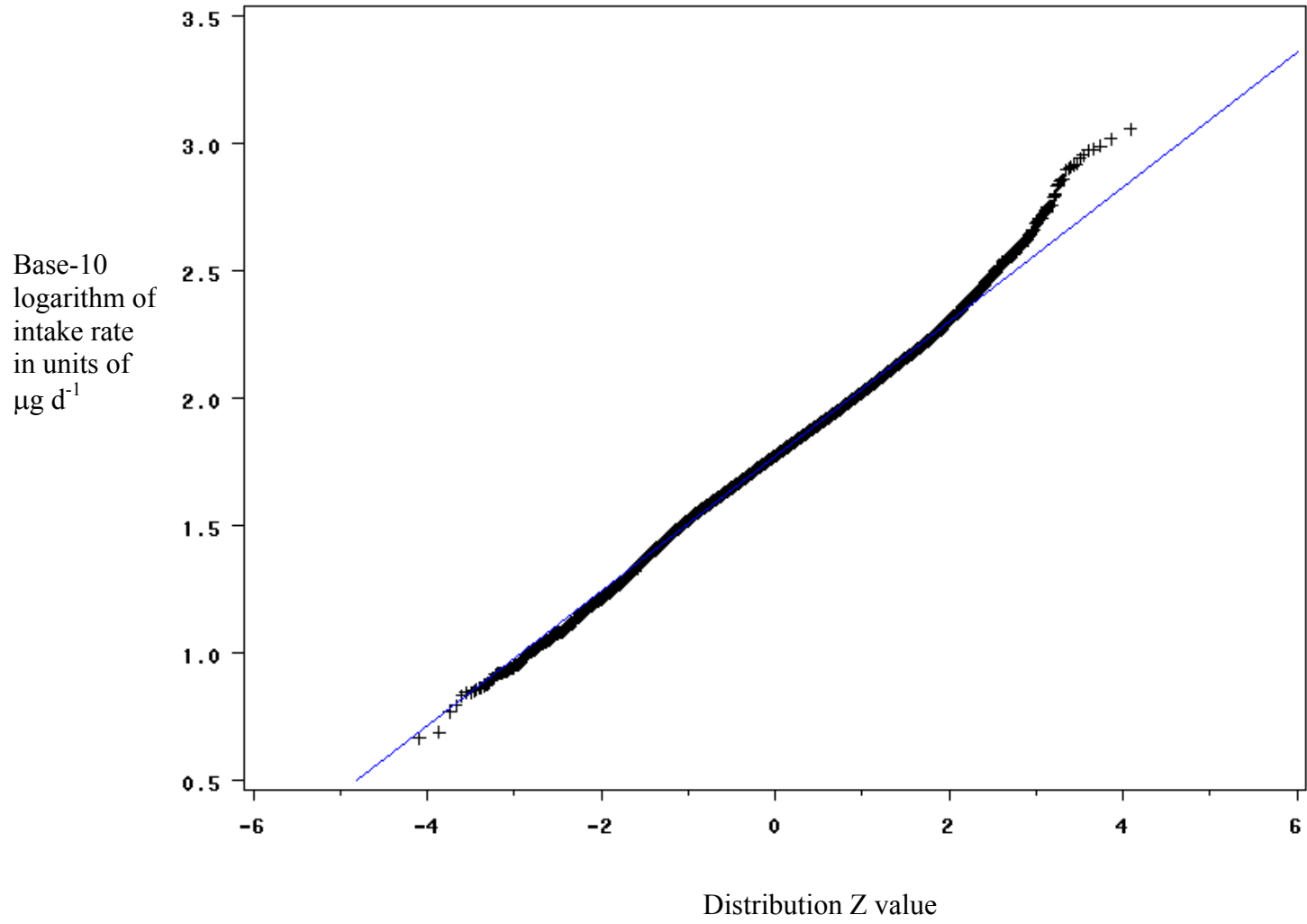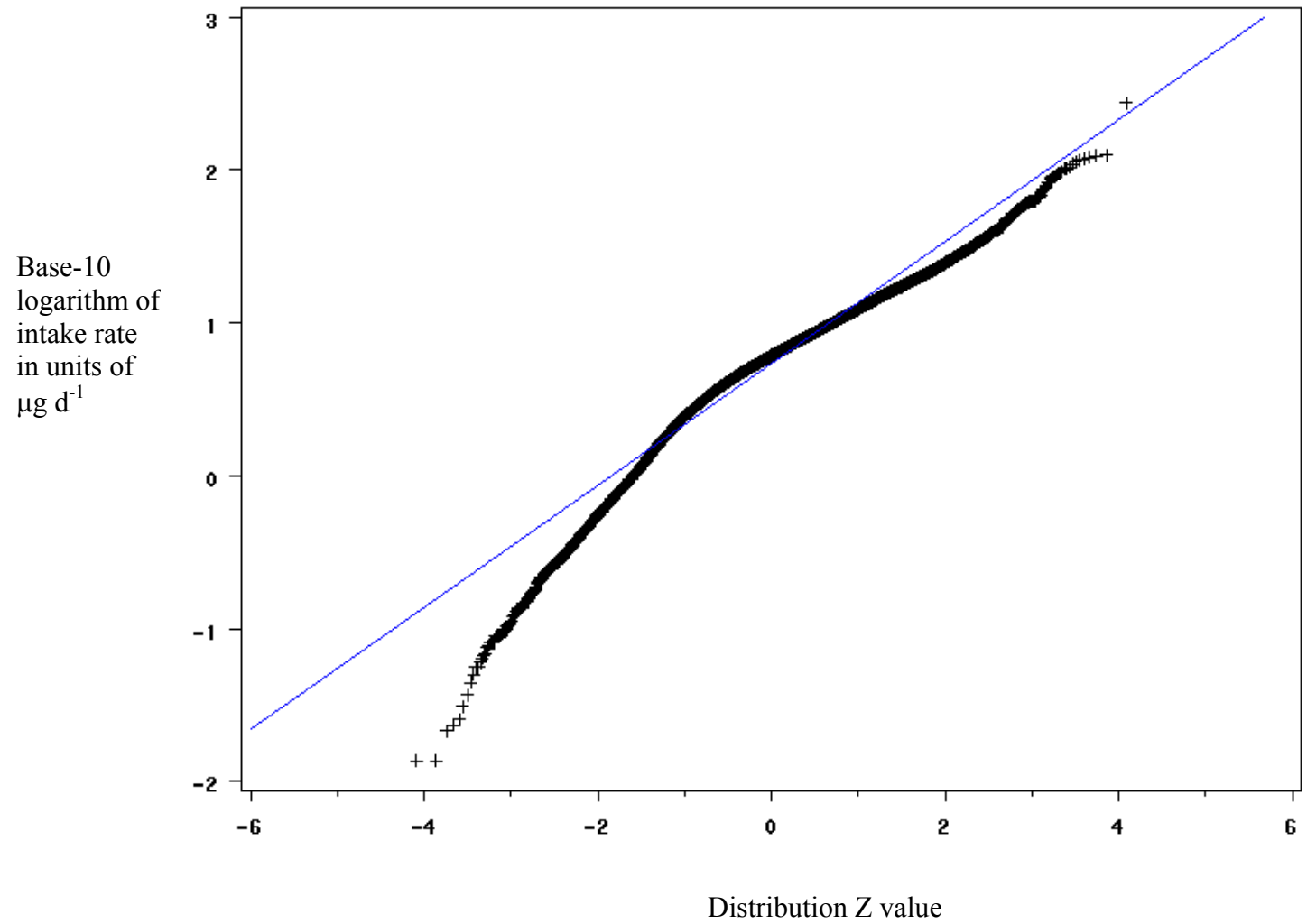
Figure 6-3a: benzene

Base-10 logarithm of intake rate in units of $\mu g \ d^{-1}$

Distribution Z value

Figure 6-3b: 1,3-butadiene
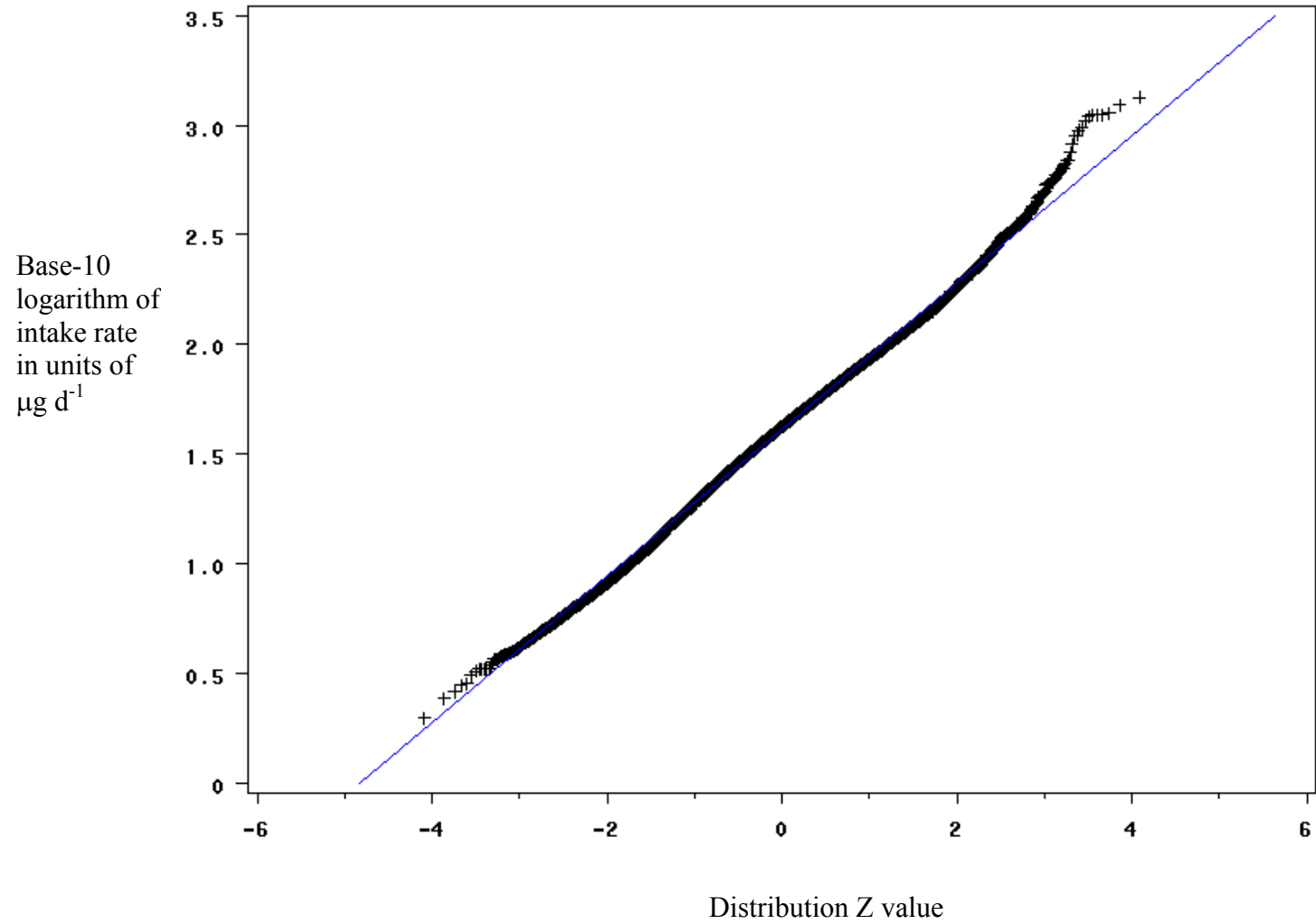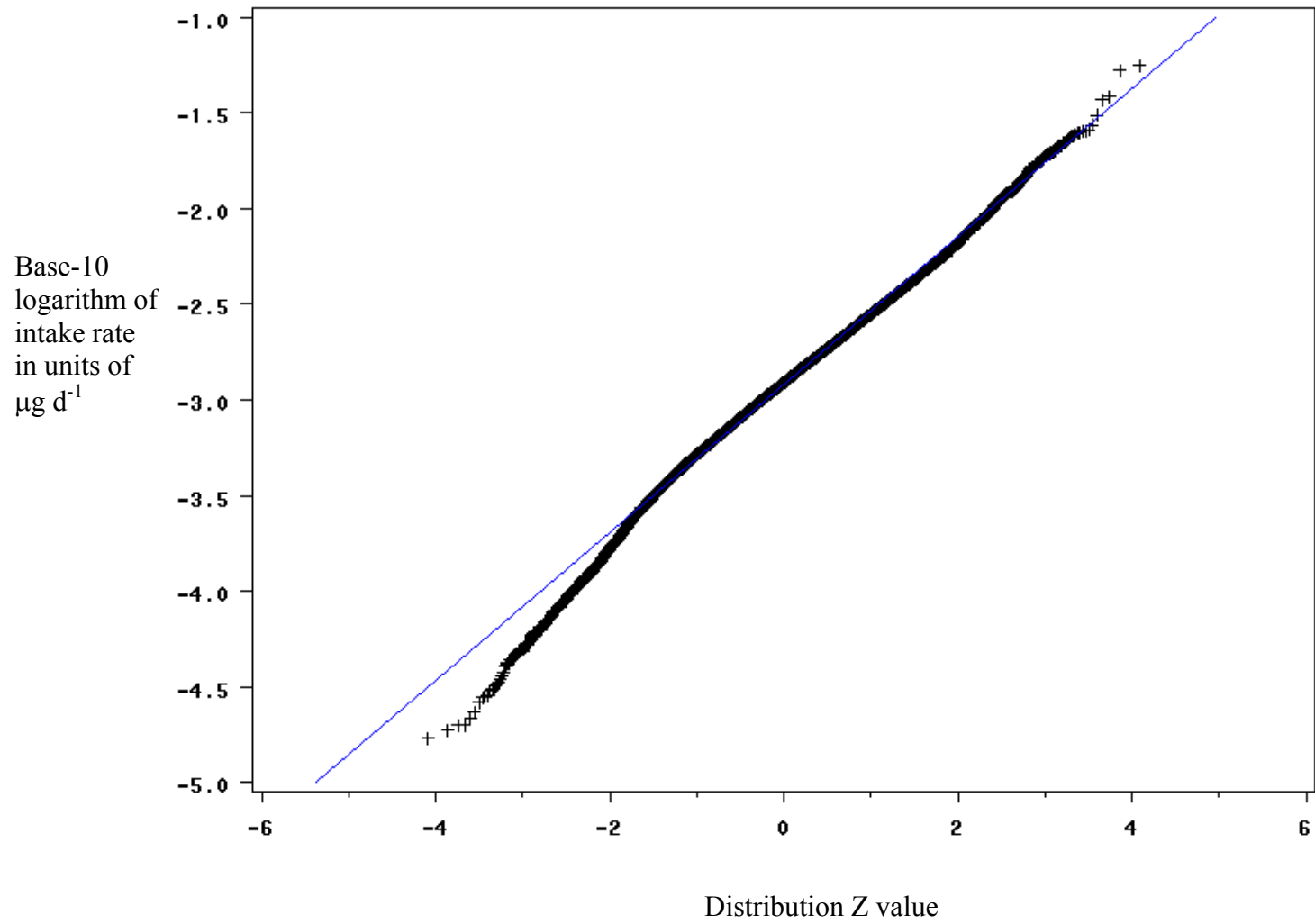
Figure 6-3c: diesel PM$_{2.5}$

Figure 6-3d: hexavalent chromium PM$_{2.5}$
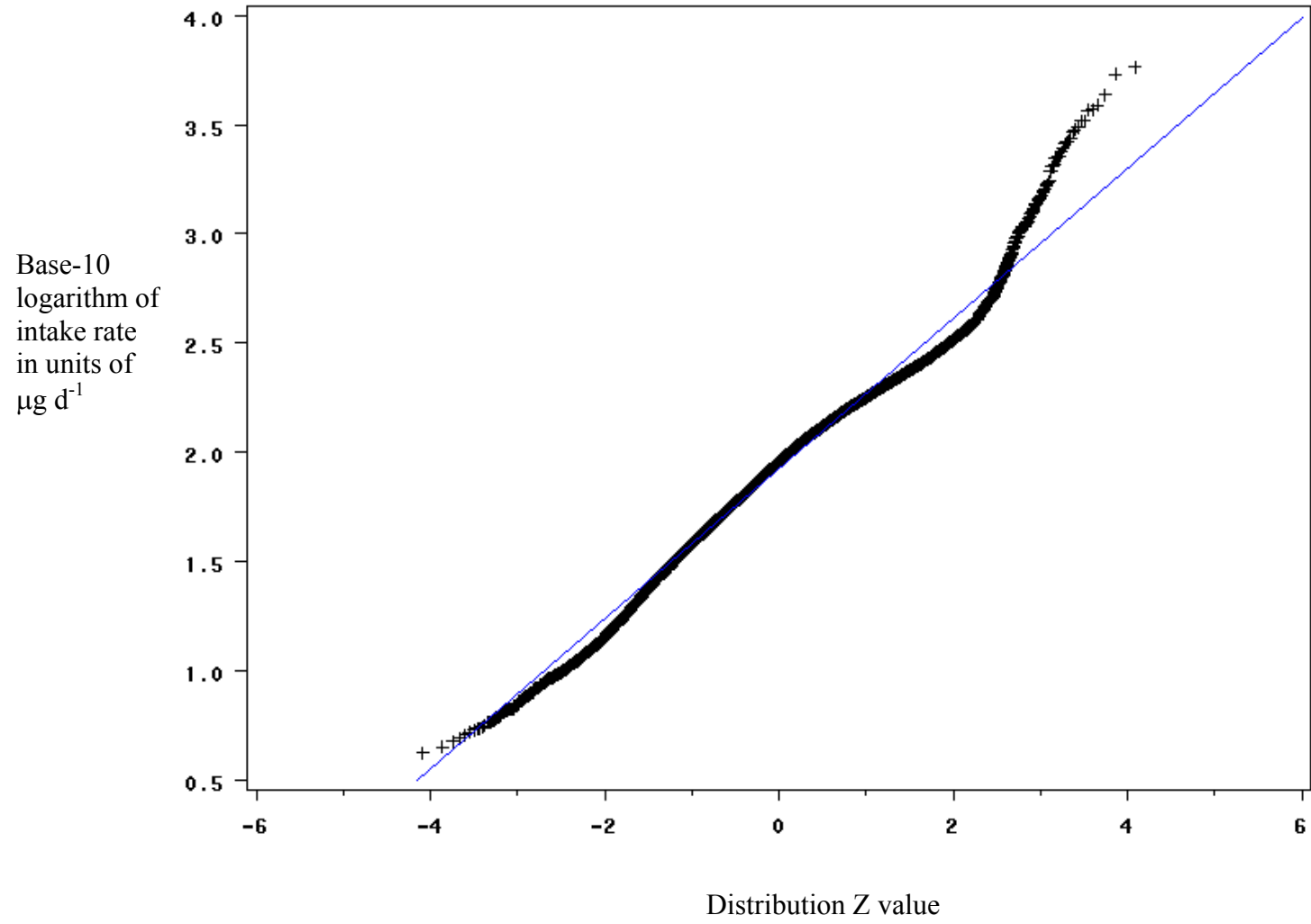
Figure 6-3e: ozone

(Figure 6-3 appears on the preceding pages.)

Figure 6-3: Cumulative distribution plots of inhalation intake rate for the 28,745 person-days simulated, for each of the five chemicals studied (benzene, butadiene, diesel $PM_{2.5}$, hexavalent chromium $PM_{2.5}$, and ozone. The y-axis values, which are dimensionless, are the $\log_{10}$ of the inhalation intake rate in units of $\mu g \ d^{-1}$. The x-axes are the Z values (quantiles) of the distribution. For completeness, these plots present all model results. However, the accuracy is lower when predicting the tails of the distribution than when predicting the central tendency or main range.
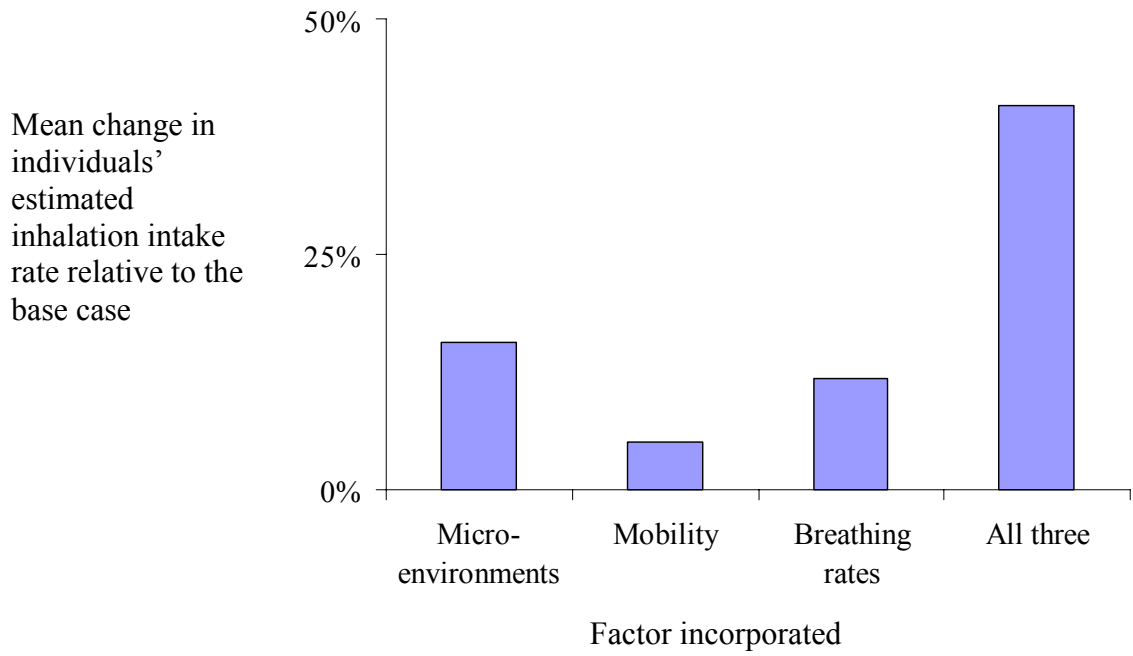
Figure 6-4a: benzene
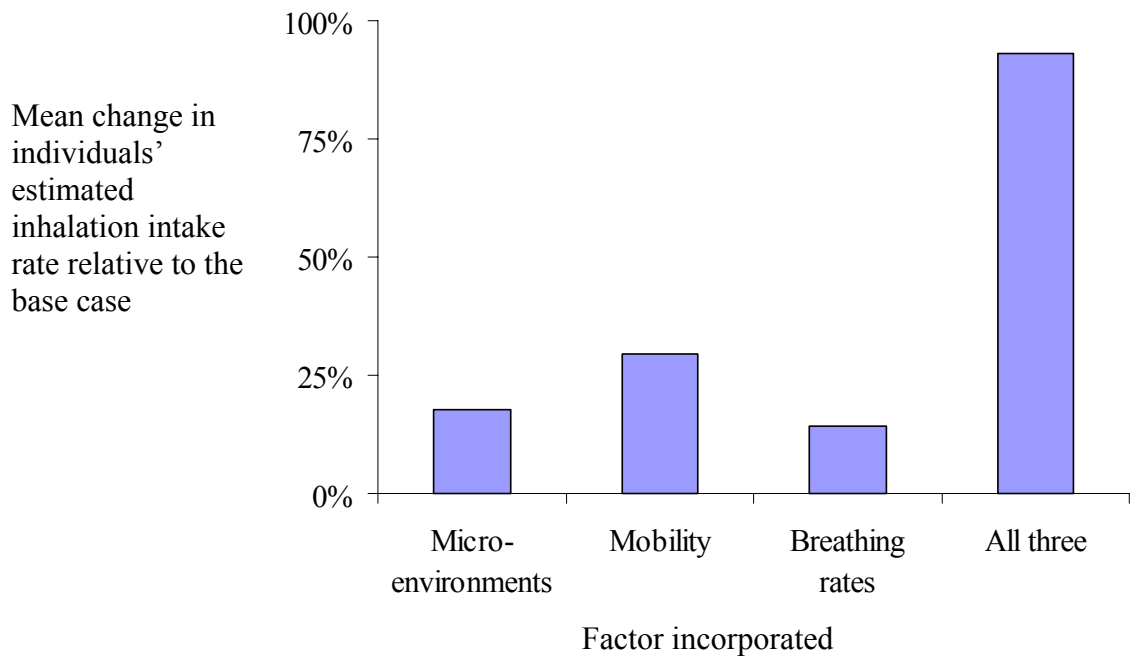
Figure 6-4b: 1,3-butadiene

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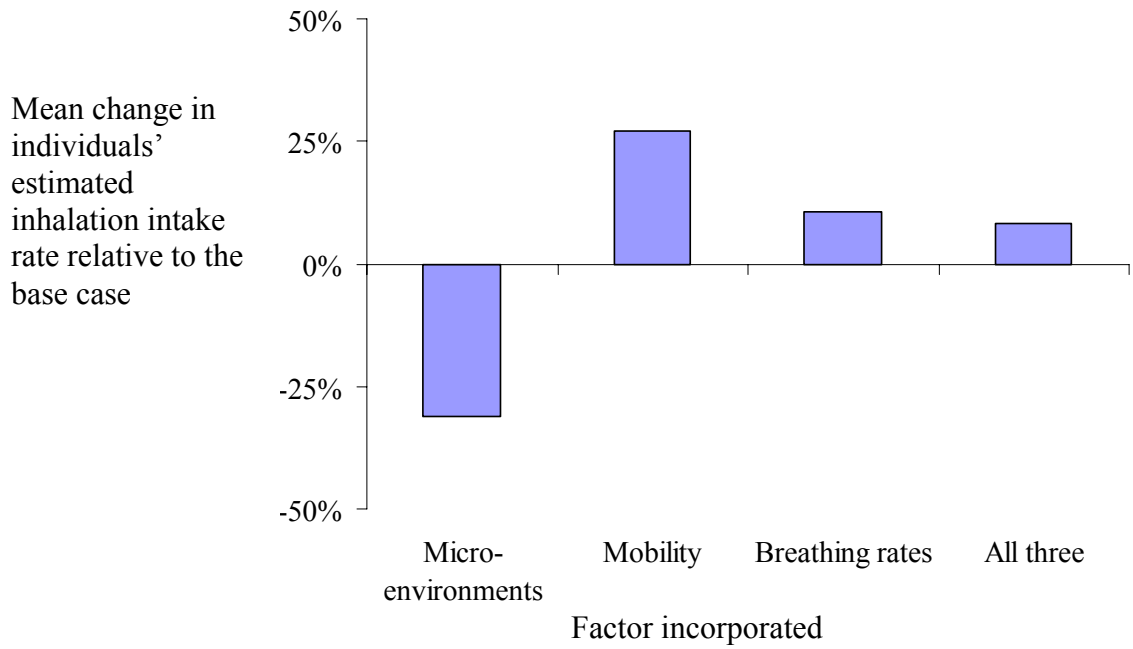
Figure 6-4c: diesel PM$_{2.5}$
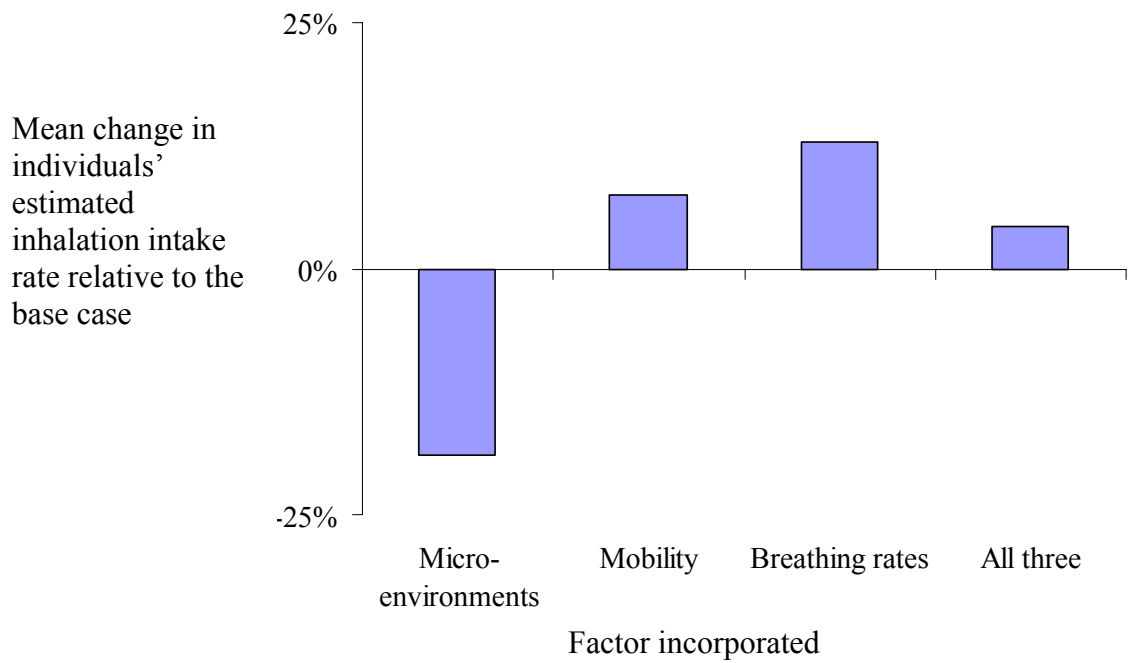
Figure 6-4d: hexavalent chromium PM$_{2.5}$
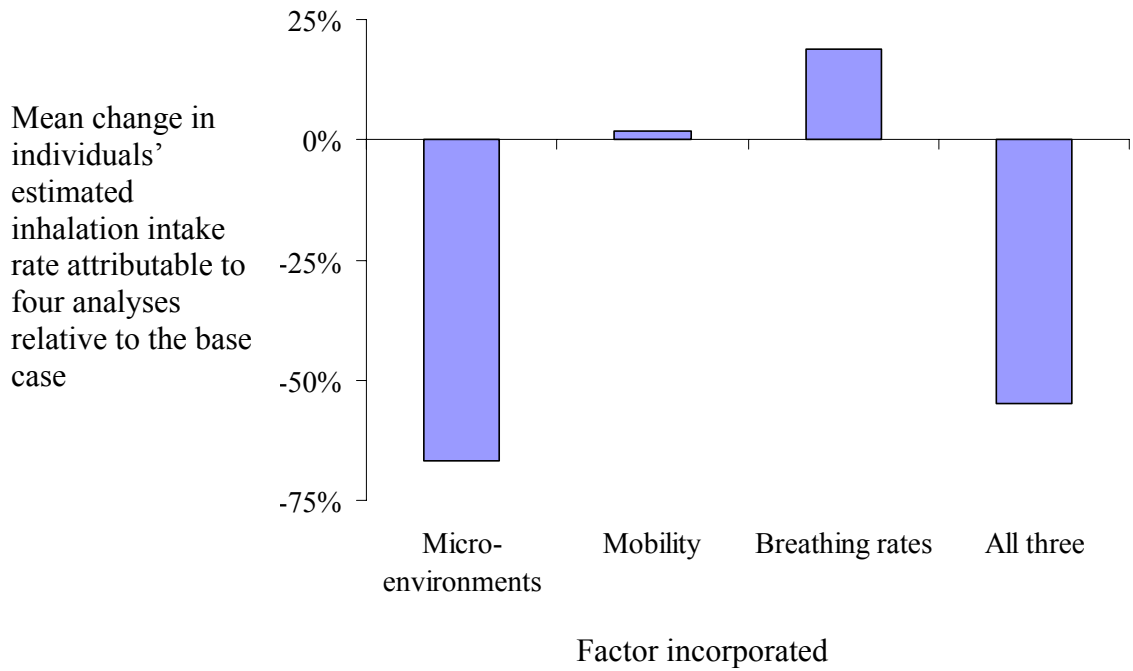
Figure 6-4e: ozone

(Figure 6-4 consists of five plots; see also preceding pages.)

Figure 6-4: Mean change in individuals' estimated inhalation intake rate attributable to incorporating one or more of three factors relative to the base case (see text), for each of the five chemicals studied. For example, accounting for microenvironments decreases the estimated intake rate for ozone, but increases the estimated intake rate for benzene. For butadiene, accounting for mobility is the most important of the three factors, increasing the estimated intake rate by ~30%.
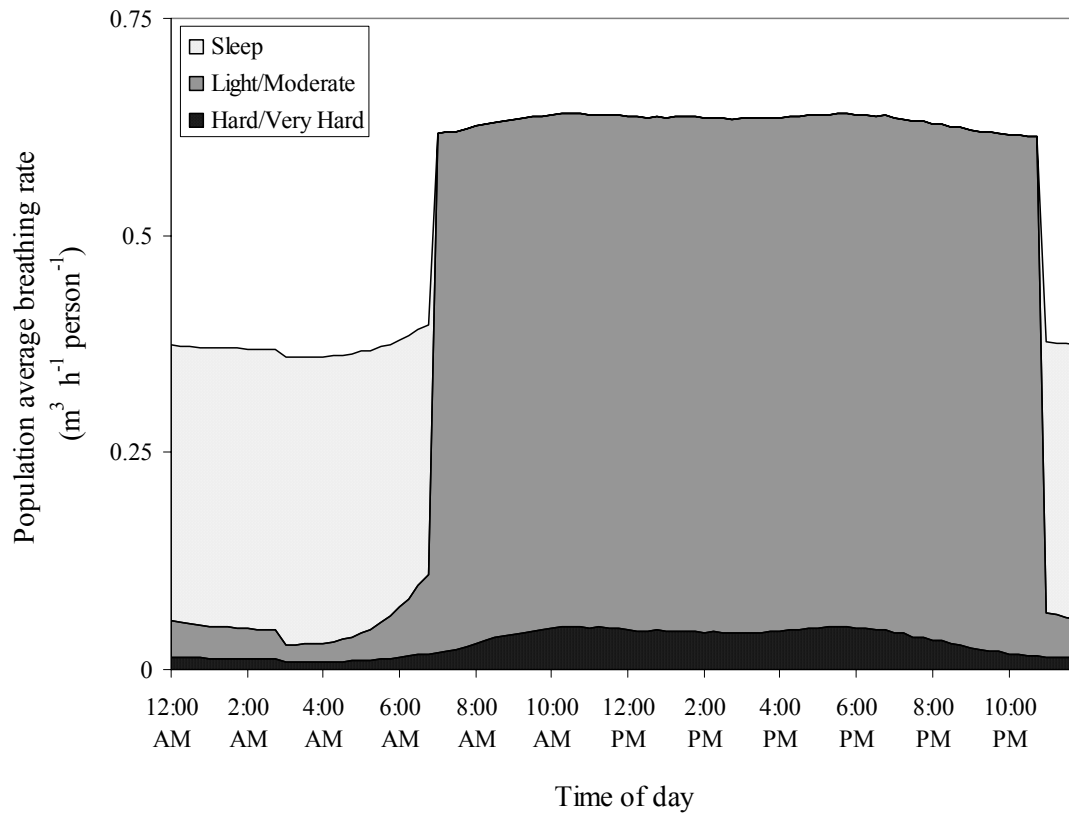
Figure 6-5: Estimated diurnal profile of the population mean breathing rate for the 28,745 person-days simulated. The daily average breathing rate per person is 13 m^3 d^{-1} $person^{-1}$. This estimate is derived from the time-activity survey data used in this work.
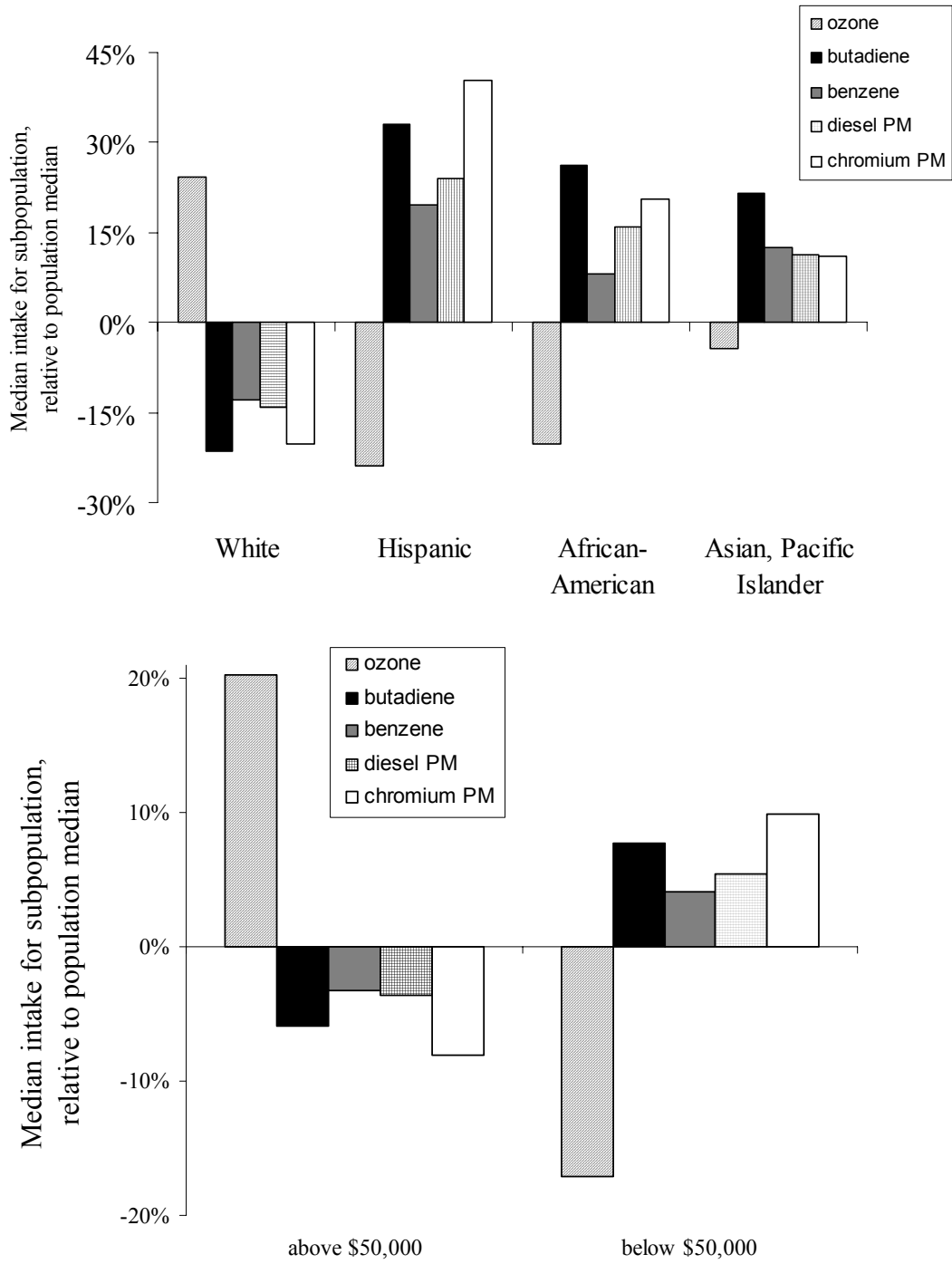
Figure 6-6: Estimated median inhalation intake rate for the subpopulation, relative to the population median, based on ethnicity (upper plot) and household income category (lower plot).
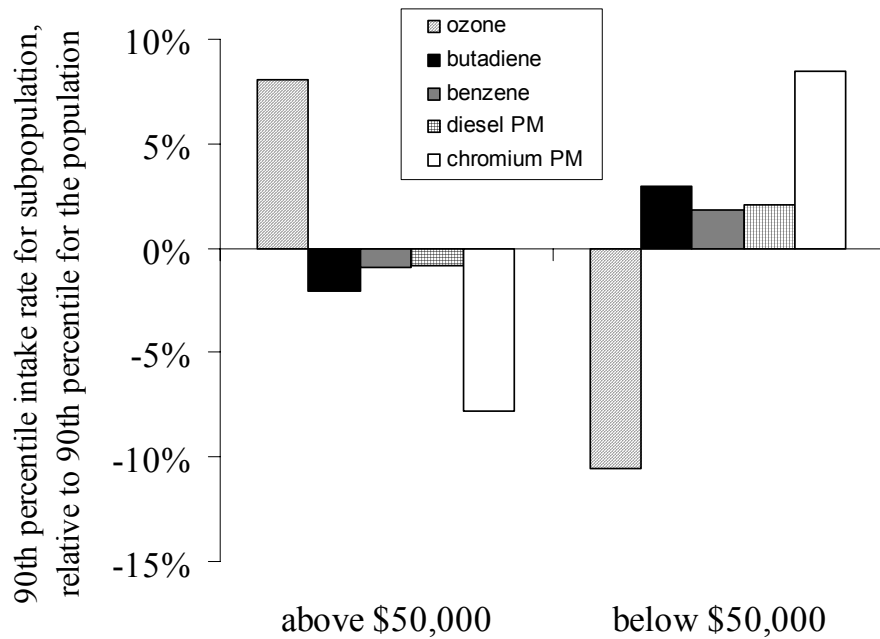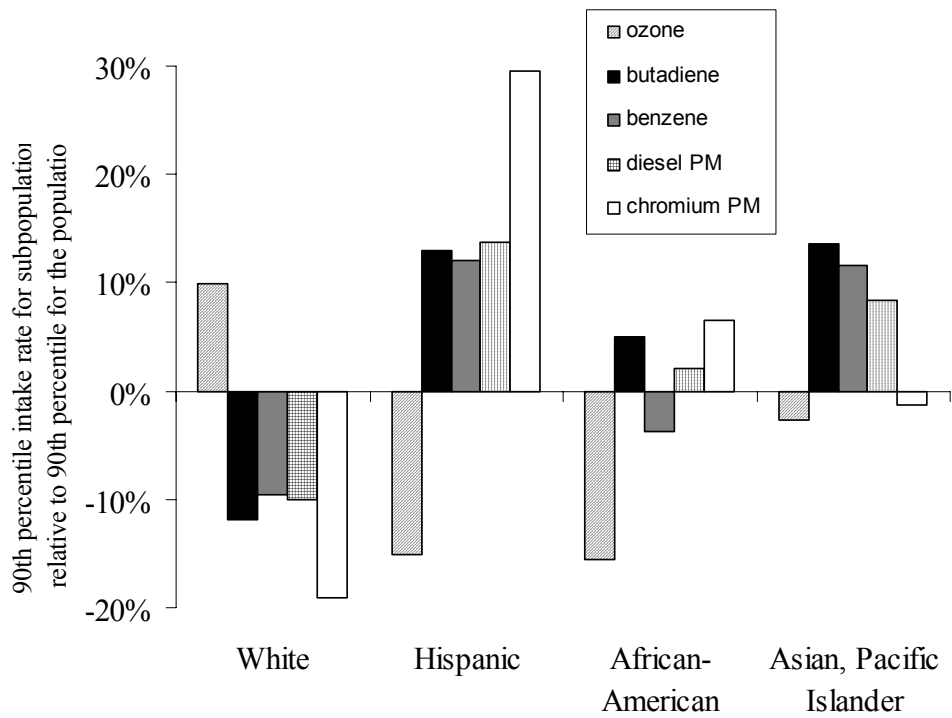
Figure 6-7: Estimated 90th percentile inhalation intake rate for the subpopulation, relative to the population 90th percentile, based on ethnicity (upper plot) and household income category (lower plot).
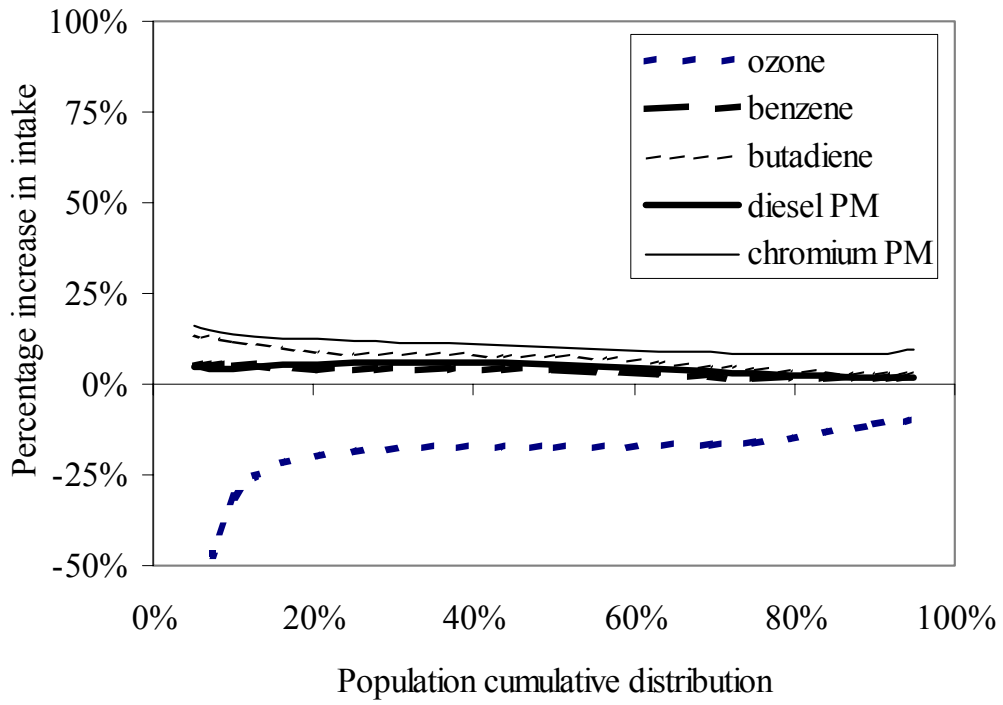
Figure 6-8a: Household income below $50,000
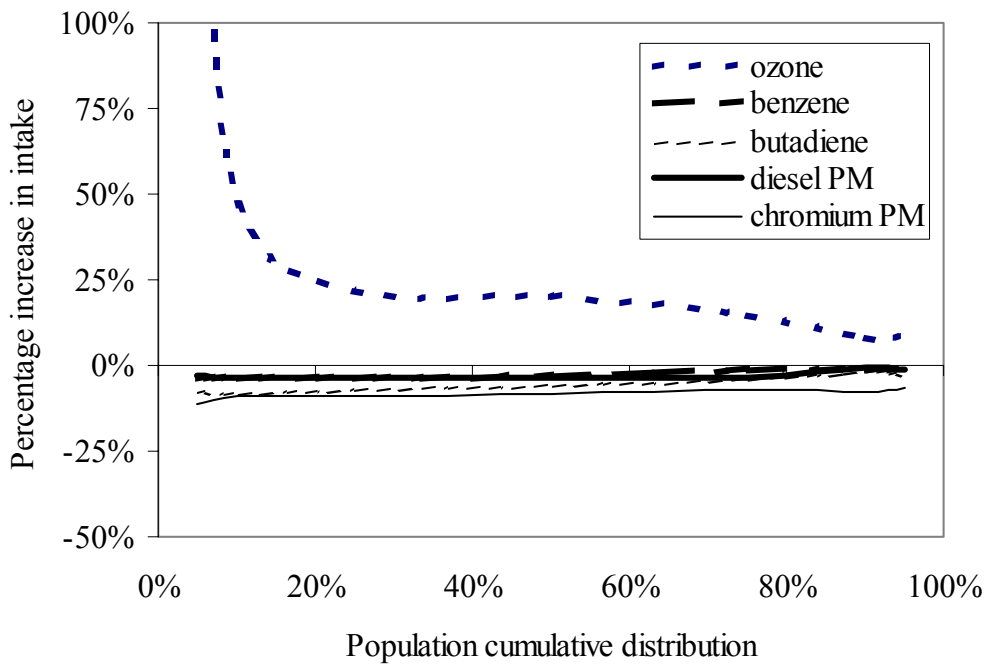
Figure 6-8a: Household income above $50,000

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Figure 6-8: Estimated intake rate distribution by household income class, relative to the population. Values show indicate, for each pollutant, the subpopulation intake rate at each point on the distribution relative to the intake rate for the whole population at the same cumulative percentage. For example, for the subpopulation with household income more than $50,000, for ozone, the y-axis value is 15% at the x-axis value 75%. This means that the 75th percentile intake rate for this subpopulation is 15% more than the 75th percentile intake rate for the whole population. Intake rates for the four primary pollutants (benzene, diesel PM, chromium PM, and butadiene) are higher for the subpopulation with lower household income than for the subpopulation with higher household income. For ozone, the reverse is true.
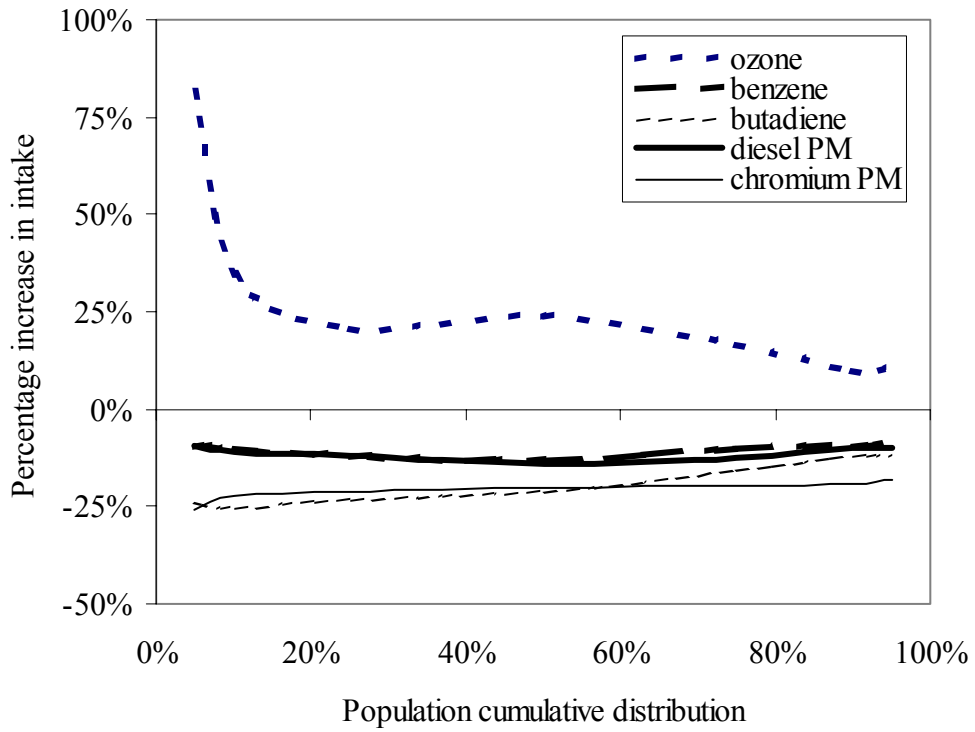
Figure 6-9a: White
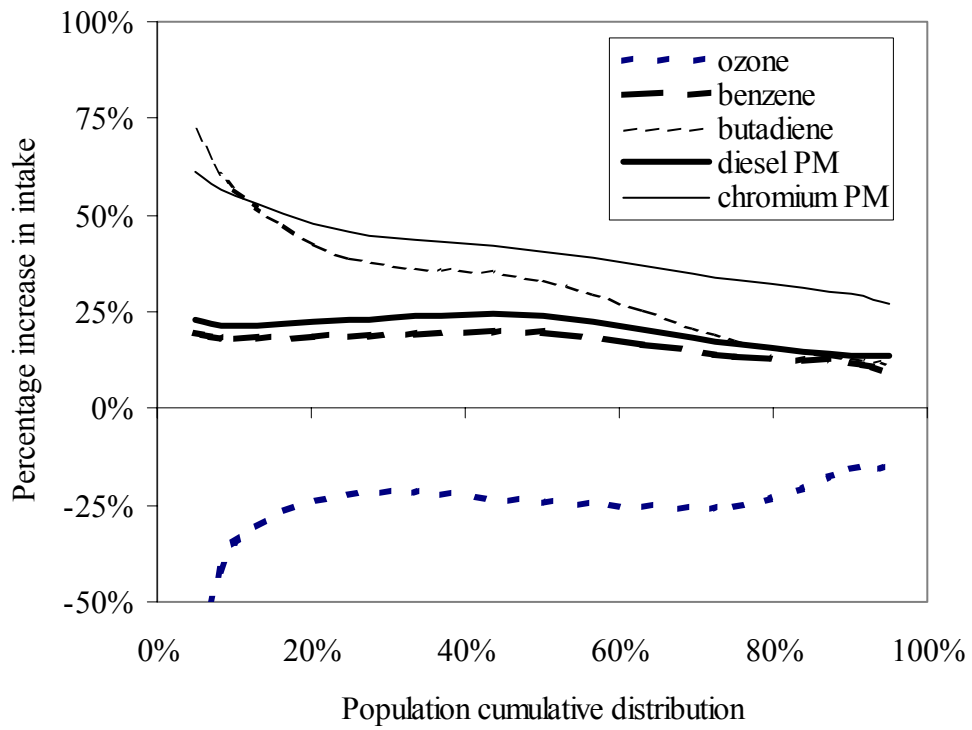
Figure 6-9b: Hispanic
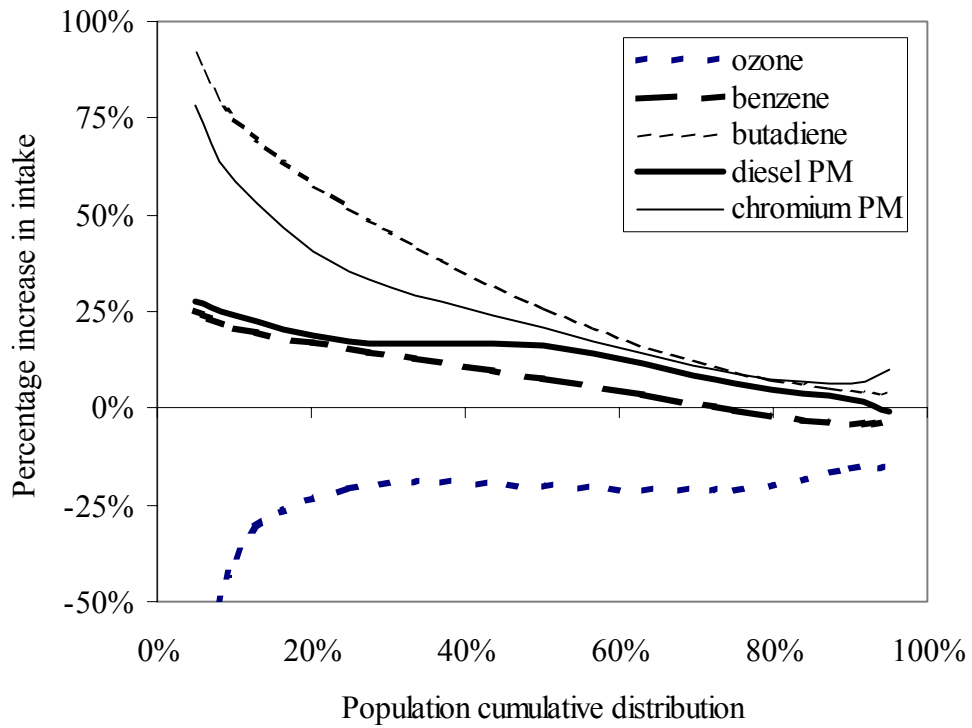
Figure 6-9c: African-American
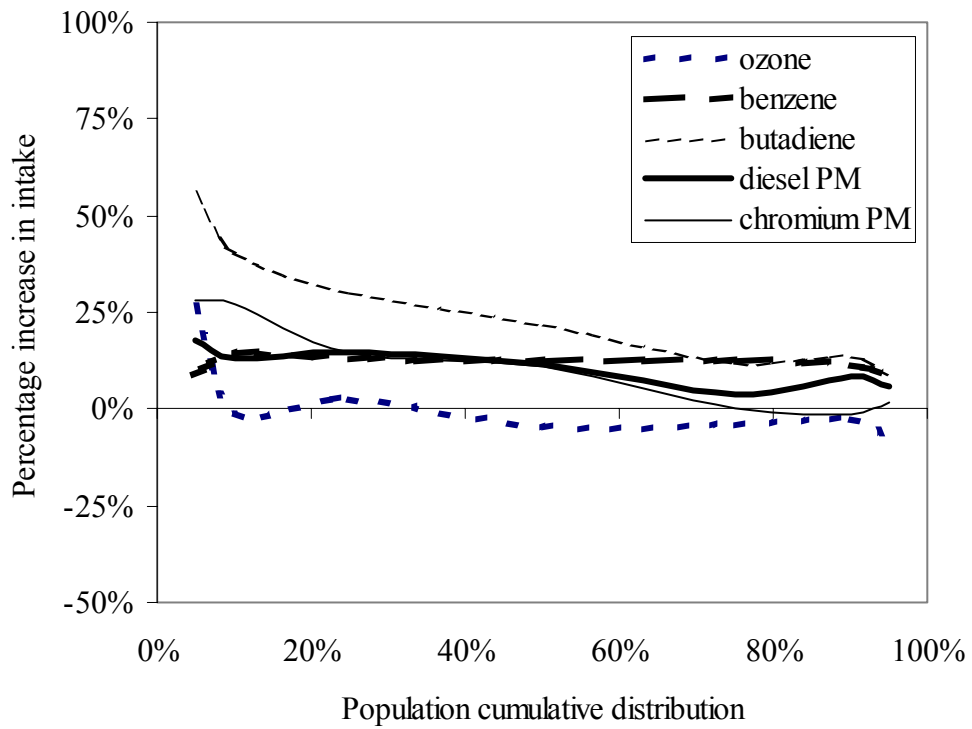
Figure 6-9d: Asian/Pacific Islander

Figure 6-9: Estimated intake rate distribution by ethnicity, relative to the population intake rate. Values indicate, for each pollutant, the subpopulation intake rate at each point on the distribution relative to the intake rate for the whole population at the corresponding point of the cumulative distribution. For example, for Whites, for ozone, the y-axis value is 17% at the x-axis value 75%. This means that the 75th percentile intake rate for this subpopulation is 17% more than the 75th percentile intake rate for the whole population. In general, intake rates for the four primary pollutants (benzene, diesel PM, chromium PM, and butadiene), intake rates are higher for non-Whites than for Whites. For ozone, the reverse is true.

Chapter 7: Effects of urban population and land area on inhalation intake of vehicle emissions

Introduction

This chapter considers whether and how urban planning might be enlisted as a tool to improve air quality and health. Specifically, I explore here how changes in urban land area and population density would affect population inhalation of vehicle emissions. Effective air quality management would likely involve multiple strategies for improving air quality and health. Urban planning represents only one class of approaches from among the many technical, economic, and social strategies available.

Previous investigations of urbanization and health have typically fallen into one of two camps: "urban penalty" research, which documents ways in which bringing people closer together (i.e., increasing urbanization) increases health risks and disease rates, and "sprawl penalty" research, which documents the environmental and social health implications of migration to suburbs and exurbs. There are currently calls to move beyond these two camps, instead investigating more holistically the relationship between urban living conditions and health (Freudenberg et al., 2005). The end goal, of course, is not only to study the system but also to uncover possible intervention strategies that

would improve living conditions and health. This chapter explores how one aspect of living conditions – population density – relates to inhalation intake of vehicle emissions, a proxy for the health effects of these emissions. The findings presented below indicate that there may be either an urban penalty or a sprawl penalty, depending on specific attributes of the infill or sprawl.

Traditionally, air quality engineers have investigated the connection between transportation demand (measured, for example, in terms of total vehicle-miles traveled) and emissions, and between emissions and ambient concentrations. Recently, air quality managers have begun to consider the extent to which urban planning may reduce transportation demand and motor vehicle emissions. Increasing population density is expected to reduce average daily vehicle-kilometers traveled in private motor vehicles per capita (VKT) for several reasons (Ewing and Cervero, 2001). For example, increasing population density increases accessibility: people in more dense areas do not need to travel as far to reach common destinations such as stores, schools, and employment centers (Cervero, 1997; Levinson, 1998). Public transit and non-motorized private transportation such as walking and biking have higher mode shares in more densely populated regions (Crane, 2000; Messenger and Ewing, 1996). Certain disincentives to driving, such as congestion delays and limited parking availability, occur more frequently in densely populated areas.

Urban planners are interested in the air quality and health benefits of their activities for several reasons. In the US, the free market is often considered the default mechanism for societal decisions. Alternative mechanisms, such as regulations and "planning", may be seen as requiring case-by-base justification for why the free market is

insufficient. The health impacts of air pollution, as a major externality of the transportation system, are a politically and economically defensible reason why urban planning is necessary. That is, environmental health issues are important in this context because they help legitimize transportation and land-use planning.

A broad definition for infill development is "any type of new development that occurs within existing built-up areas" (US EPA, 1999a). The potential association between density and VKT has led some planners in urban areas with an increasing population to implement policies that encourage infill development rather than sprawl (APA, 2002; Burchell et al., 2002; US EPA, 2001b). To understand the air-quality impacts of such policies, two questions arise: (1) Under what circumstances does increasing population density reduce vehicle emissions? (2) Under what circumstances does reducing emissions by increasing population density reduce people's inhalation intake of vehicle emissions? A few publications have commented on these questions. An international study of motor vehicle use concluded that "whilst *per capita* [transportation] emissions may be higher in the low-density automobile-dependent regions, the rate of [transportation] emissions per urbanized hectare [is] clearly lower. We thus have the situation in the high-density cities… where emissions output is highly concentrated. This leads to more concentrated impacts and higher exposure…" (Kenworthy and Laube, 2002). Cervero (2000) summarizes the dilemma: "exposure levels (and thus health risks) are lower with sprawl, but tailpipe emissions and fossil-fuel consumption are greatly increased."

Many urban areas are growing in population or land area or both, and this growth may impact emissions and emissions-to-intake relationships. Such impacts will vary with

urban conditions (e.g., urban population) and with the nature of growth. To my knowledge, no prior research has quantified how changes in urban land area and urban population would affect the population inhalation of transportation emissions. Nor has previous research addressed the necessary conditions such that increased population density is accompanied by reduced inhalation of vehicle emissions. This chapter contributes to filling these gaps. In addition to offering insights for air quality management and urban planning, the results in this chapter can inform expectations in the absence of active planning.

I start with the premise, discussed in Chapters 1 and 2, that population inhalation of vehicle pollutants is more appropriate than emissions, ambient concentrations, or conditions for the maximally exposed individual, as an indicator of the total public health impacts attributable to air pollution. I develop and present an exploratory analysis that considers a hypothetical, idealized representation of an urban area. Using this representation, I investigate, quantitatively and parametrically, how three changes in urban land area and urban population influence population inhalation of motor vehicle emissions: (1) increasing population while land area remains constant (denoted "infill" in this chapter), (2) increasing land area while population remains constant ("sprawl"), and (3) increasing land area and population while density remains constant ("constant-density growth"). Note that as employed here, these terms have a narrower and more precisely defined scope than in common usage.

There is debate in the literature as to whether and how much population density and other aspects of urban form influence VKT. Some investigations have found that increasing density reduces VKT while others have found no connection (Badoe and

Miller, 2000). Some research suggests that the correlation between density and VKT is not causal, but rather that density is a proxy for income, which is itself causally connected to VKT (Boarnet and Sarmiento, 1998). Others disagree, finding that both density and income are important (Kenworthy and Laube, 2002). In this chapter, I do not take a position on this debate. Because there is variability and uncertainty in the impact of density on VKT and vehicle emissions (Badoe and Miller, 2000; Gordon and Richardson, 1997), I allow a range of values (including zero) for the density-emissions elasticity, and I identify the minimum elasticity necessary for a given change in urban population and land area to reduce intake.

Methods

Because this investigation represents the first attempt to quantify the relationship between urban population density and the inhalation intake of primary traffic-related air pollutants, I aim for a direct approach that clarifies underlying relationships, aids in elucidating causal connections, and permits the problem to be analytically tractable. I consider population density, passenger vehicle emissions, attributable ambient concentrations for primary pollutants, and the resulting attributable intake per capita. Below I describe my method for connecting these elements of the source-intake relationship for primary pollutants from motor vehicles.

Density-emissions elasticity

Population density has the potential to influence vehicle emissions (Holtzclaw et al., 2002) as well as the fraction of emissions inhaled by people (Lai et al., 2000). Population density is a key aspect of urban form, and one that can be influenced by urban planning.

If there were no relationship between density and VKT, then an increase in population density would cause an increase in both transportation emissions per km^2 and per capita inhalation of transportation emissions. On the other hand, if an increase in population density were to result in a reduction in per capita emissions, then the same two variables (emissions per km^2 and per capita inhalation of emissions) might either increase or decrease, depending on the density-emissions elasticity. Equation 7-1 defines density-emissions elasticity (ε_e) and density-VKT elasticity (ε_v):

$$\varepsilon_e \equiv \frac{dE/E}{d\rho/\rho}, \qquad \varepsilon_v \equiv \frac{dD_{VKT}/D_{VKT}}{d\rho/\rho}. \tag{7-1}$$

Here, E is the total vehicle emission rate of a pollutant (g s^{-1}), ρ is the population density (people km^{-2}), and D_{VKT} is the average daily per capita vehicle-distance traveled (km person^{-1} d^{-1}). If ε_e is negative and large in magnitude, then increasing population density could reduce both vehicle emissions and per capita inhalation of vehicle emissions. However, if the magnitude of ε_e is small (but still negative), then increasing population density could reduce vehicle emissions yet increase per capita inhalation of vehicle

emissions. In this investigation, I allow ε_e to vary, and explore the resulting relationship

between changes in population, land area and per capita inhalation of vehicle emissions.

Pollutant classification

The relationship between emissions and inhalation intake depends, among other

factors, on the dynamic behavior of the air pollutant. Pollutants are classified as *primary*

or *secondary*, according to whether they are emitted directly from sources or are formed

in the atmosphere from precursors (Seinfeld and Pandis, 1998). Pollutants are further

classified as *nonreactive* or *reactive* according to their level of atmospheric reactivity.

For the present purposes, a nonreactive pollutant is one for which the pollutant's

characteristic atmospheric lifetime owing to loss by chemical reaction or deposition is

significantly greater than the characteristic residence time of air in an urban basin

(typically of the order of several hours; see Chapter 4).

Vehicular emissions of concern include primary nonreactive species (e.g., CO and

benzene), primary reactive species (e.g., 1,3-butadiene and ultrafine particles), and

secondary reactive species (e.g., ozone and NO_2). The analysis in this chapter focuses on

primary nonreactive pollutants as the logical and important first step toward a complete

treatment of all pollutant classes. In the discussion, I outline how one would extend the

methods to address secondary and reactive primary pollutants.

Ambient concentrations

In this chapter, I use a one-compartment model (Benarie, 1980; Lyons et al., 2003) to describe the relationship between emissions and ambient concentrations. The strengths and limitations of this model are discussed in Chapter 4. The limitations of the one-compartment model are such that the results reported here should be considered as preliminary and suggestive rather than conclusive.

The steady-state mass-balance equation for a square-plan, one-compartment model yields the following expression for attributable concentration of a primary nonreactive pollutant:

$$C = \frac{E}{uH\sqrt{A}} = \frac{FD_{VKT}P}{uH\sqrt{A}} \times \frac{1}{86400} . \qquad (7\text{-}2)$$

Here, C is the average ambient concentration attributable to vehicles (g m^{-3}), u is the wind speed (m s^{-1}), H is the mixing height (m), A is the urban land area (m^2), F is the average motor vehicle emission factor (g km^{-1}), P is the population size, and 86400 converts time units from seconds to days. The group (uH) indicates how rapidly local meteorology dilutes and removes emissions from an area; the group ($P\,A^{-0.5}$) is linear population density; and, the group (FV) is the average per capita emission rate.

Intake

Given Equation 7-2, average daily per capita intake of motor vehicle emissions, I (units: g person^{-1} d^{-1}), can be estimated as

$$I = QC = \frac{QFD_{VKT}P}{uH\sqrt{A}} \times \frac{1}{86400} .$$

(7-3)

Here, Q is the average breathing rate for an individual (m^3 person^{-1} d^{-1}).

Of the variables that urban planning might influence, I explore three: D_{VKT}, P, and A. I define a normalized intake (I^*, units: d^{-1}) to highlight the influence on intake of these three variables:

$$I^* \equiv I\left(\frac{uH}{QF} \times 86400\right) = \frac{D_{VKT}P}{\sqrt{A}} .$$

(7-4)

Although potentially important, I do not explore here intraurban concentration variability, the influence of urban population and area on emission factors (e.g., by changing traffic flow conditions), or the role of urban form on mixing height (e.g., via the urban heat island effect).

Exposure concentrations can be subdivided based on the distance to the responsible emission source: e.g., global (>3000 km), regional (150–3000 km), urban (5–150 km), local (200 m – 5 km), and microenvironmental (3–200 m) (Colvile et al., 2003; Watson and Chow, 2001). For the analysis presented here, I consider exposures from urban and local emissions. The importance of regional and global emissions will depend on the pollutant and the emission rate upwind of an urban area (Tsuang et al., 2003). An urban area's population and land area are unlikely to strongly affect exposures attributable to emissions from upwind sources. The importance of microenvironmental factors depends on the amount of time spent in a microenvironment and the concentration difference between a microenvironment and ambient air. Exposures in near-source microenvironments contribute a greater fraction of total intake for rapidly decaying

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primary pollutants (e.g., ultrafine PM) than for nonreactive species. Because of the transport and dispersion that occurs during the interval between precursor release and secondary pollutant formation, local and microenvironment emissions will be less important for secondary pollutants that take ~0.5 hour or more to form than for primary pollutants.

Both intraurban concentration heterogeneity and microenvironments might play important roles influencing the relationship between urban form and inhalation intake of primary vehicle emissions. However, in addition to the analysis of ambient CO monitoring station data presented in Chapter 4, other evidence also indicates that average outdoor concentrations are relatively homogeneous for primary nonreactive pollutants from motor vehicles. For example, Chapter 3, which investigated population exposure to benzene and CO from vehicle emissions in California's South Coast Air Basin, presented results for two analyses. The first analysis accounted for spatial variability of population density and ambient concentrations; temporal variability of concentrations and breathing rates; and microenvironments such as in- and near-vehicle and indoors near a freeway. The second analysis considered only the air basin-wide annual average ambient concentration. Estimated average intake values in the second analysis were ~70% of the values in the first analysis, indicating that the ambient concentration analysis captured most of the average population exposure to motor vehicle emissions. In a second example, Watson and Chow (2001), studying conditions in Mexico City, reported that "65% of the 24-hr black carbon was part of the urban mixture, 23% originated in the neighborhood surrounding the monitor, and only 12% was contributed from nearby sources [within ~1 km]." For primary nonreactive pollutants, if there are removal

mechanisms as air moves from outdoors to indoors (e.g., ventilation system air filters that can remove diesel PM), then the average attributable exposure concentration will be less than the average attributable ambient concentration. But if such removal mechanisms do not exist (e.g., for CO), then the average attributable exposure concentration will more nearly equal the average attributable outdoor concentration. In addition to these considerations, the present study explores how *changes* in urban population and area lead to *changes* in inhalation. This approach reduces the importance to the results of differences between the average attributable ambient concentration and the average attributable exposure concentration.

Results

Changes in urban population and area

Figure 7-1 illustrates the three changes in urban population and area considered (infill, sprawl, and constant-density growth). I present the effect of increases in urban population and area on per capita inhalation of vehicle emissions; a reduction would cause the opposite effect. Equations describing the three changes in urban population and area are given in Table 7-1. The entries in Table 7-1 follow from Equations 7-1 and 7-3 and from the assumption that, among the variables considered, per capita transportation emissions are only a function of population density. The entries do not assume any specific functional form for the density-emissions relationship.

Figure 7-2 summarizes key results. For the system considered here, constant-density growth always increases per capita intake. Infill and sprawl may either increase or

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decrease per capita intake, depending on the density-emissions elasticity. Infill reduces

per capita intake when ε_e is less than -1.0. Sprawl reduces per capita intake when ε_e is

greater than -0.5.

Rather than plotting numerical values on the ordinate axes, Figure 7-2 shows

mathematical expressions. To calculate the value for the derivatives in a specific city, one

needs to know values of parameters such as the city's population and land area. The term

on the ordinate axis of the $\partial I/\partial P$ plot (Figure 7-2, left) contains $A^{-0.5}$, indicating that — all

else being equal — changes in per capita intake attributable to changes in population

would be more significant in small cities than in large cities. The term on the ordinate

axis of the $\partial I/\partial A$ plot (Figure 7-2, right) contains $P\,A^{-1.5}$, indicating that — all else being

equal — changes in per capita intake attributable to changes in land area would be more

significant in densely populated small cities than in sparsely populated large cities.

Table 7-2 presents the results in terms of an important policy question: which

change in urban population and land area minimizes per capita intake? The answer

depends on density-emissions elasticity, ε_e, and on whether population is increasing,

decreasing, or remaining constant. For example, consider the case of an increasing

population. If ε_e is less than -0.5 then infill minimizes per capita intake; if ε_e is greater

than -0.5 then constant-density growth minimizes per capita intake.

Density-emissions elasticity

The analysis in this chapter develops results that depend on the relationship

between population density and transportation emissions. Only a few studies have

investigated this relationship. A comparison between two Nashville neighborhoods found

that one neighborhood is 68% more dense, has 25% fewer VKT, and has 7% less toxic-

emissions per capita per day from vehicles, than the other (NRDC, 2003). These findings

suggest $\varepsilon_e = -0.10$ and $\varepsilon_v = -0.37$. The study did not consider changes in population

intake. Using an international dataset, Newman and Kenworthy (1989) reported a

density-fuel consumption elasticity of between -0.4 and -0.5. Fuel consumption is likely

a better surrogate for vehicle emissions than distance traveled (Pokharel et al., 2002;

Singer and Harley, 1996). On-road remote sensing techniques used to determine vehicle

emissions in these studies may prove valuable in direct investigations of density-

emissions elasticity.

Because data from empirical studies of ε_e are sparse, I use empirical information

about ε_v as a surrogate. The relationship between ε_e and ε_v is

$$\frac{\varepsilon_e}{\varepsilon_v} = \frac{\dfrac{dE}{dD_{VKT}}}{\dfrac{E}{D_{VKT}}} = \frac{F^*}{F}, \qquad (7\text{-}5)$$

where F^* (g km^{-1}) is the marginal change in emissions attributable to a marginal change

in VKT. Using reported values for ε_v in place of robust estimates for ε_e assumes $F^* \approx F$,

i.e., that F is not strongly dependent on population density. Because density and other

urban-form attributes affect congestion (Dunphy and Fisher, 1996) and because emission

factors are related to average speed (Kean et al., 2003; Ntziachristos and Samaras, 2000),

distance traveled is an imperfect indicator of emissions. I expect in many situations that

the density-emissions elasticity would be greater than the density-VKT elasticity (e.g., if

both terms were negative, I expect the density-emissions elasticity would be less negative

than the density-VKT elasticity). Because of start-up emissions (Heeb et al., 2003), reductions to average trip length would reduce emissions less than it would reduce VKT. Furthermore, increasing density may increase congestion and driver aggressiveness, which would increase emission factors (De Vlieger et al., 2000). If future research better quantifies the relationship between density and emissions, that information could be applied directly to the approach presented in this investigation to yield refined results.

There is debate in the literature regarding the nature of the density-VKT relationship. Some investigations have found little or no relationship between density and VKT, suggesting that ε_e may be approximately zero, while other investigations have found a strong relationship between density and VKT (Badoe and Miller, 2000; Mindali et al., 2004). Published ε_v values are between 0 and –0.7 (Holtzclaw et al., 2002). Empirical evidence of density-VKT elasticity comes from both intra- and interurban comparisons. Figure 7-3 presents an interurban comparison of density and VKT (US DOT, 2003c). These data exhibit a clear inverse relationship and suggest $\varepsilon_v \approx -0.3$. A 1996 study of four areas in Toronto (urban core, core ring, inner suburbs, and outer suburbs) found that urban core residents traveled half as far (motorized distance traveled) and had about four times the residential density (persons per sq. km. of urbanized land) as outer suburb residents (CST, 1998), suggesting that $\varepsilon_v = -0.5$. Transportation demand modeling of two hypothetical housing developments in each of three US metropolitan areas (Montgomery County, Maryland; San Diego, California; and West Palm Beach, Florida) concluded that VKT would be 40 – 50% lower for infill than for "greenfield" development (US EPA, 1999b). Holtzclaw (1991; 1994) reported that ε_v is between –0.3 and –0.5 after accounting for demographic variables such as income and cars per

household. Internationally, a strong relationship has been observed between urban density and travel patterns (Kenworthy et al., 1999). For example, in a comparison of 100 cities worldwide, Kenworthy and Laube (2002) concluded, "The data show how the higher car use cities are low in population density and more decentralized… while the higher density and more centralized cities have reduced car use per person."

Empirical elasticity values cited here are from intra- and interurban comparisons, rather than from changes over time in a single urban area. By comparing available estimates for density-VKT elasticity with the results presented in this work, I implicitly assume that existing intra- and interurban cross-sectional data are informative about the longitudinal conditions that would apply in any given urban area. This assumption is common in the literature, but, to my knowledge, it has not been rigorously tested.

Comparing the analyses with reported values for ε_v, I find that whether infill is an effective strategy for minimizing intake of vehicle emissions depends on the circumstances. Within the range of reported ε_v values, infill and constant-density growth both tend to increase per capita intake. If the elasticity is strong ($\varepsilon_v < -0.5$), then the intake increase is less for infill than for constant-density growth. However, in the case of weak elasticity ($\varepsilon_v > -0.5$), the reverse is true. On the basis of the available evidence, it appears that merely increasing population density, while holding constant all other aspects of urban form, will likely not reduce VKT enough to reduce average per capita intake of primary motor-vehicle emissions. Rather, to reduce inhalation intake of air pollutants emitted from motor vehicles, the analysis presented in this chapter suggests that infill development must include urban design features that strengthen the density-

VKT relationship, such that the density-emissions elasticity satisfies the condition $\varepsilon_e <$ −0.5.

Discussion

Considering a specific urban area

Applying the intake results presented here to a specific urban area would require an estimate of ε_e or ε_v. The results presented in Table 7-1 and Figure 7-2 do not depend on a specific functional form for ε_e or ε_v. However, estimating ε_e or ε_v for a given situation may require specifying this function.

Empirical studies of the density-VKT relationship often report results as "doubling density reduces VKT by X%." These observations can be represented mathematically using the following two-parameter relationship:

$$D_{VKT} = k\rho^{\left[\frac{\log\left(1-\frac{X\%}{100\%}\right)}{\log(2)}\right]}.$$

(7-6)

Here, k is a constant (km person^{-1} d^{-1}), and X is the percent reduction in VKT attributable to a doubling of population density. The exponent in Equation 7-6 is the density-VKT elasticity (ε_v). For example, if doubling density reduces VKT by 40%, then $\varepsilon_v = -0.74$. As an alternative to Equation 7-6, Holtzclaw et al. (2002) suggested the following three-parameter relationship:

$$D_{VKT} = a(\rho + b)^c.$$

(7-7)

Here, *a*, *b*, and *c* are empirical constants.

To compare the two functional forms found in the literature (Equations 7-6 and 7-7), I determined the correlation parameters for the neighborhood-scale data used by Holtzclaw et al. (2002) and for the urban-scale data reported by the US Department of Transportation (US DOT, 2003c). The neighborhood-scale dataset contains VKT and density for each traffic analysis zone in three urban areas (Chicago, Los Angeles, and San Francisco). The urban-scale dataset contains VKT and density for the 47 urban areas in the US with population greater than 750,000. Correlation parameters for the two- and three-parameter density-VKT equations, and a summary of the input datasets used to derive these parameters, are presented in Table 7-3. I report the neighborhood-scale density-VKT relationship for three cities (Chicago, San Francisco, Los Angeles). I also report the urban-scale density-VKT relationship for two representative urban areas (Atlanta and New York) from among the 47 urban areas in the dataset. There is almost no difference in the goodness-of-fit parameter (r^2) for the two- and three-parameter equations.

Table 7-3 also contains changes in normalized intake attributable to the three hypothesized changes in urban population and area. Intake differences in Table 7-3 between the two- and three-parameter equations are <14% and <4% for the neighborhood- and urban-scale datasets, respectively.

Figure 7-4 presents the relationship between elasticity and population density for the functional fits to the empirical neighborhood-scale data presented in Figure 7-3b. Elasticity is independent of density for the two-parameter equation. However, for the

three-parameter equation, elasticity magnitude increases as density increases ($\varepsilon_v =$ $c/(1+(b/\rho))$).

The normalized intake results in Table 7-3 and Figure 7-2 provide relative estimates of the exposure impact of changes in urban population and area. To quantify intake (Equation 7-3) for a specific pollutant in a specific location, one must specify average breathing rate (Q), average emission factor (F), and typical meteorological conditions in terms of wind speed and mixing height (uH). Appropriate values for these parameters are presented next.

Estimates of the US population-average breathing rate vary. In this dissertation, I have used the value 12.2 m^3 d^{-1} person^{-1} (Layton, 1993). Alternative values (units: m^3 person^{-1} d^{-1}) found in the literature include 12 (US EPA, 1997), 15 (Marty et al., 2002), and 17 (OEHHA, 1996). As discussed in Chapter 3, I do not consider the reported value of 17 m^3 person^{-1} d^{-1} (OEHHA, 1996) to be a reliable estimate of the population breathing rate, nor do I consider it to be a useful indicator of the uncertainty in the value of 12.2 m^3 d^{-1} person^{-1} from Layton (1993). Emission factors are available for many pollutants, based on techniques such as on-road measurements and laboratory dynamometer tests. There can be significant variability and uncertainty in estimates of F (Abu-Allaban et al., 2003). An estimate of the overall average value of F can be obtained as the ratio of total vehicle emissions to total VKT. For example, dividing reported year-2000 $PM_{2.5}$ tailpipe emissions for gasoline vehicles in California's South Coast Air Basin (6.2×10^6 g d^{-1}) (CARB, 2000b) by the total distance traveled by gasoline vehicles (5.1×10^8 km d^{-1}) (CARB, 2002a) yields a value of F for tailpipe fine particulate matter of ~12 mg km^{-1}. This value is consistent with experimentally measured values (Abu-Allaban et al., 2003).

Meteorology varies among locations and times. In Chapter 4, I compute the harmonic mean value of *Hu* for each of the 73 meteorological stations in the EPA SCRAM database (www.epa.gov/ttn/scram). The median value among the stations is ~500 m^2 s^{-1}. Combining the above values, for tailpipe emissions of $PM_{2.5}$, I^* can be converted to *I* by multiplying by 4.2×10^{-9} mg $person^{-1}$.

Results in Table 7-3, combined with conversion factors such as those given above, can provide information that is helpful to cost-benefit analyses and to understanding the health impacts of urban development. For example, the value in Table 7-3 for infill development in Atlanta, $\partial I^*/\partial P|_A = 0.55$ d^{-1} $person^{-1}$, is converted to $\partial I/\partial P|_A = 2.3 \times 10^{-9}$ mg d^{-1} $person^{-2}$ for $PM_{2.5}$. This means that if the population of Atlanta were to increase by 100,000 people via infill development, I estimate that the average increase in inhalation intake of tailpipe emissions of $PM_{2.5}$ would be 0.2 μg $person^{-1}$ d^{-1}. Per Table 7-3, if the same population growth were to occur via infill development in New York City, then the expected average increase in per capita inhalation intake of $PM_{2.5}$ would be 3 times lower. Based on results presented in Chapter 3 and 6, the effects of microenvironments, time-varying breathing rates, and individual mobility on estimated average per capita inhalation would be expected to modify these results in detail but not in the main. The impact of these three factors (microenvironments, time-varying breathing rates, and individual mobility) on comparisons between urban areas (e.g., the factor of 3 difference between the effect of population growth in Atlanta versus New York City) is expected, in most cases, to be modest.

Considering specific pollutants or pollutant classes

The analysis presented in this investigation is directly applicable to inhalation of primary conserved passenger-vehicle emissions, such as benzene and CO. The results can also inform considerations beyond this subset of pollutants. For example, at equal emission rates, the average ambient concentration of a primary conserved pollutant would be higher than for a primary reactive pollutant. All else being equal, intake for a primary nonreactive pollutant is an upper-bound estimate of intake of primary, reactive (or depositing) pollutants. Similarly, the estimated *change* in intake of a primary nonreactive pollutant that results from a change in urban form (e.g., as given in Table 7-3) is an upper bound estimate of the *change* in intake of a primary reactive pollutant.

For rapidly reacting pollutants (i.e., those for which the characteristic reaction time is much less than the time for removal from the air basin by advection), concentrations are likely to exhibit a high degree of spatial heterogeneity. For all primary vehicle pollutants, concentrations will be higher near roadways than elsewhere, but the concentration difference between near-source and not-near-source areas would be greater for rapidly reacting pollutants than for nonreactive pollutants. One implication of this difference is that, when estimating population inhalation of vehicle emissions, proximity to the emission source is more important for rapidly reacting pollutants than for slowly reacting pollutants. A second implication is that the difference between the population average exposure and exposures for people who live or work in proximity to major roadways will be greater for rapidly reacting pollutants than for slowly reacting pollutants.

Two important pollutants associated with transportation are diesel PM (predominantly from non-passenger vehicles) and ozone (a highly reactive, secondary pollutant). To my knowledge, estimates of density-emissions elasticity for diesel PM do not exist, and I do not expect ε_e for passenger vehicles to be an accurate estimator of ε_e for diesel PM. Because diesel vehicle emissions are concentrated near specific land uses (e.g., highways and freight centers), I expect ambient concentrations in US urban areas to be more spatially heterogeneous for diesel emissions than for passenger vehicle emissions (SCAQMD, 1999). The density-emissions elasticity for diesel PM may be negative, because increasing population density is likely to increase the efficiency with which organizations can deliver the goods and services that require diesel consumption. However, there is currently no good basis for estimating this parameter.

The approach for primary pollutants developed in this investigation could be extended to secondary pollutants (Marquez and Smith, 1999). For example, investigations of how changes in VKT affect ozone concentrations can yield a pseudo-emission factor, defined as the attributable change in the average mass of ozone in an urban area divided by the change in VKT (Carter, 1989). Similar metrics could be explored for changes in the size of an urban area or the spatial distribution of precursor vehicle emissions. Factors influencing such metrics include climate and meteorology, topography, total precursor emissions (i.e., including non-vehicle emissions), and the spatial and temporal distribution of emissions and of changes in emissions. Vehicle emissions may reduce ozone concentrations locally (because fresh NO emissions remove ozone) but increase ozone concentrations in areas that are downwind of the emissions. Average ozone concentrations are lower indoors than outdoors because the absence of

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direct sunlight reduces ozone formation and because reactions with indoor surfaces increase ozone destruction (Weschler, 2000).

Non-health impacts of vehicle emissions

The health effects attributable to inhalation of emissions are only one of many impacts associated with motor vehicles and urban form (Delucchi, 1996). Emissions occur throughout the lifecycle of all components of the transportation infrastructure, including vehicles, fuels, and roads. Impacts of the transportation system include local and global environmental damage (e.g., habitat loss, urban heat island effects, and global climate change). Among non-pollution health effects, urban form may influence exercise levels, obesity, mental health, and other "quality of life" issues (Frank and Engelke, 2001; Frumkin, 2002).

Actions that reduce one impact might not reduce other impacts. As an example, Table 7-4 presents policies that influence greenhouse gas (GHG) and toxic emissions, and population inhalation of vehicle emissions. Some actions exhibit co-benefits between these impacts; others exhibit trade-offs.

Other issues

An important limitation to the approach employed here is the assumption that individuals are exposed to the same attributable concentration. Differences in exposures among individuals and among subpopulations are important components of society's overall air quality concerns. (This topic is explored in Chapter 6.) While the results of

this investigation indicate that sprawl may reduce total population inhalation of motor vehicle emissions, the exposure change is not expected to be uniform across the population. Sprawl may reduce the population average exposure while increasing exposures for persons living near transportation corridors, especially if people living at the urban edge commute to downtown locations.

A second important limitation is that, unlike in Chapters 3 and 6, I use here the average ambient concentration as a proxy for the average exposure concentration. In some situations (e.g., benzene concentrations in vehicles), attributable exposure concentrations are likely to be greater than attributable ambient concentrations; in other situations (e.g., particulate matter in a mechanically ventilated building), the reverse is true. In a specific urban area, correlations are likely among population density, building type and age, the ratio of indoor-to-outdoor pollution concentrations, and time spent in or near vehicles. Such considerations may be important in understanding a specific individual's or subpopulation's exposures.

Finally, my analysis does not address the effects of changes in fuels and vehicle technologies. Aggressive programs have led to demonstrable and substantial reductions in on-road emissions of many criteria and toxic air pollutants (Kean et al., 2001; Kean and Harley, 2000). In cases where inhalation intake of vehicle emissions is not reduced by infill development alone, combining infill with efforts to further reduce vehicle emissions may permit overall inhalation intake to decrease. Historically, vehicle improvements over time have substantially reduced emissions per mile, thereby offsetting increases in VKT (Kahn, 2000).

Conclusions

Urban land area and population change over time, with or without planning. I

analyze the impact of changes in land area and population on per capita inhalation of

primary passenger vehicle emissions. Depending on the density-emissions elasticity (ε_e),

infill development has the potential to reduce motor vehicle emissions yet increase per

capita inhalation of these emissions, while sprawl has the potential to increase vehicle

emissions but reduce inhalation of these emissions. Under the idealized conditions

considered here, for ε_e greater than –0.5, constant-density growth and sprawl minimize

intake for increasing and constant population, respectively. For ε_e less than –0.5, infill

and contraction minimize intake for increasing and constant population, respectively.

Data on density-emissions elasticity (ε_e) are lacking, but published values for density-

VKT elasticity (ε_v) are between 0 to –0.7. To interpret the model results (which are based

on ε_e), I assumed that ε_v is a reasonable proxy for ε_e, and also that data on ε_v from cross-

sectional studies provides useful predictive information for describing changes in

response to growth over time in any given urban area. To the extent that these

assumptions are reasonably accurate, then merely increasing population density while all

other aspects of urban form are unchanged appear unlikely to reduce VKT enough to

reduce average per capita intake of motor vehicles emissions. Rather, to reduce health

impacts of transportation emissions relative to constant-density growth, infill

development would have to include urban design features that strengthen the density-

VKT relationship, such that the condition $\varepsilon_e < -0.5$ is satisfied.

An ultimate goal in air quality management is to minimize adverse health effects

of air pollution. In the case of motor vehicle emissions, major progress has been achieved

through technological developments such as fuel reformulation and on-board emission controls. Urban planning may also reduce vehicle emissions and their associated health effects. To do so will require a better understanding of the relationships among urban form, vehicle use, vehicle emissions, and inhalation intake of those emissions. This investigation offers early progress toward such understanding.

Table 7-1: Mathematical description of the three changes in urban population and area[a]

Name of change in urban form	Change in urban population, area, and density	Incremental change in normalized pollutant intake associated with incremental change in urban population and area
Infill	Population increases; land area is constant; density increases.	$$\left.\frac{\partial I^*}{\partial P}\right|_A = \frac{D_{VKT}(1+\varepsilon_e)}{\sqrt{A}}$$
Sprawl	Population is constant; land area increases; density decreases.	$$\left.\frac{\partial I^*}{\partial A}\right|_P = -\frac{PD_{VKT}}{2A^{1.5}}\left[2\varepsilon_e + 1\right]$$
Constant-density growth	Population increases; land area increases; density is constant.	$$\left.\frac{\partial I^*}{\partial P}\right|_\rho = \frac{1}{\rho}\left.\frac{\partial I^*}{\partial A}\right|_\rho = \frac{D_{VKT}}{2\sqrt{A}}$$

(a) Here, I^* is the normalized intake (d^{-1}), P is the population, A is the urban land area (km^2), D_{VKT} is the average daily per capita vehicle-kilometers traveled, ε_e is the density-emission elasticity defined in Equation 7-1, and ρ is the population density (km^{-2}).

Table 7-2: The change in urban population and area that minimizes intake, depending on the density-emissions elasticity and the change in population[a]

	Population is increasing	Population is constant	Population is decreasing
$\varepsilon_e < -0.5$	Infill ($dA/dt = 0$)	Contraction ($dA/dt < 0$)	Constant-density contraction ($d\rho/dt = 0$)
$\varepsilon_e > -0.5$	Constant-density growth ($d\rho/dt = 0$)	Sprawl ($dA/dt > 0$)	Constant-land-area contraction ($dA/dt = 0$)

(a) Here, t = time (y), ε_e = density-emissions elasticity, A = land area (km^2), and ρ = population density (km^{-2}).

Table 7-3: Two- and three-parameter density-VKT equations and attributable changes in normalized intake[a]

	Neighborhood-scale data			Urban-scale data	
	Chicago	Los Angeles	San Francisco	Atlanta	New York City
Population (million)	7.3	14.0	5.9	3.0	17.1
Land area (km^2)	9,700	23,400	17,700	4,600	10,300
Average density (km^{-2})	753	597	336	650	1,660
Total vehicle-kilometers traveled per day (million)	136	256	112	162	424
Average vehicle-kilometers traveled per capita per day	29.9	29.5	30.4	54.4	24.8
Number of data points	31.5	1471	1048	47	
Using $D_{VKT}= k\rho^e$					
k	69	52	56	335	
e	–0.10	–0.07	–0.08	-0.31	
r^2	0.57	0.20	0.27	0.35	
ε_v	–0.10	–0.07	–0.08	-0.31	
Infill ($\partial I^*/\partial P|_A$, units: d^{-1} person^{-1})	0.27	0.18	0.21	0.55	0.17
Sprawl ($\partial I^*/\partial A|_P$, units: d^{-1} km^{-2})	-91	-49	-32	-97	-76
Constant-density growth ($\partial I^*/\partial P|_\rho$, units: d^{-1} person^{-1})	0.15	0.10	0.11	0.40	0.12
Using $D_{VKT}= a(\rho+b)^c$					
a	2100	1800	2900	343	
b	1800	2800	4200	20	
c	-0.51	-0.48	-0.52	-0.32	
r^2	0.74	0.31	0.43	0.35	
ε_v	-0.15	-0.08	-0.04	-0.306	-0.312
Infill ($\partial I^*/\partial P|_A$, units: d^{-1} person^{-1})	0.26	0.18	0.22	0.56	0.17
Sprawl ($\partial I^*/\partial A|_P$, units: d^{-1} km^{-2})	-80	-48	-35	-101	-76
Constant-density growth ($\partial I^*/\partial P|_\rho$, units: d^{-1} person^{-1})	0.15	0.10	0.11	0.40	0.12

(a) Here, ε_e = density-emissions elasticity, I^* = normalized intake (d^{-1}), P = population, A = land area (km^2), D_{VKT} = vehicle-km traveled per person per day and ρ = population density (km^{-2}).

Table 7-4: Examples of actions that increase and reduce two impacts from vehicles[a]

		CO$_2$ and toxic emissions	
		Reduction	Increase
Inhalation of emissions	Reduction	Increased fuel-efficiency	Sprawl, if $-0.5 < \varepsilon_e < 0$
	Increase	Infill development, if $-1.0 < \varepsilon_e < 0$	Reduced fuel-efficiency

(a) Here, ε_e = density-emissions elasticity.
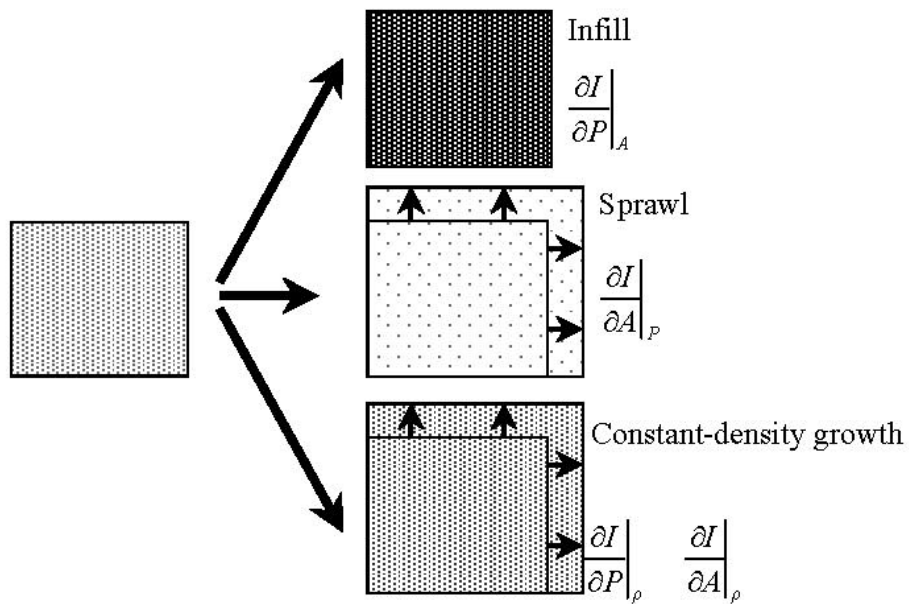
Figure 7-1: Three changes in urban population (*P*) and urban area (*A*) investigated in this chapter, in terms of the impact on the incremental change in per capita intake (*I*). The first change (infill, $\partial I/\partial P|_A$) is population increase at constant land area. The second change (sprawl, $\partial I/\partial A|_P$) is land area increase at constant population. The third change (constant-density growth, $\partial I/\partial A|_\rho$) is increase in population and land area, at constant population density. Not shown is the opposite of sprawl: a land area decrease at constant population (contraction).
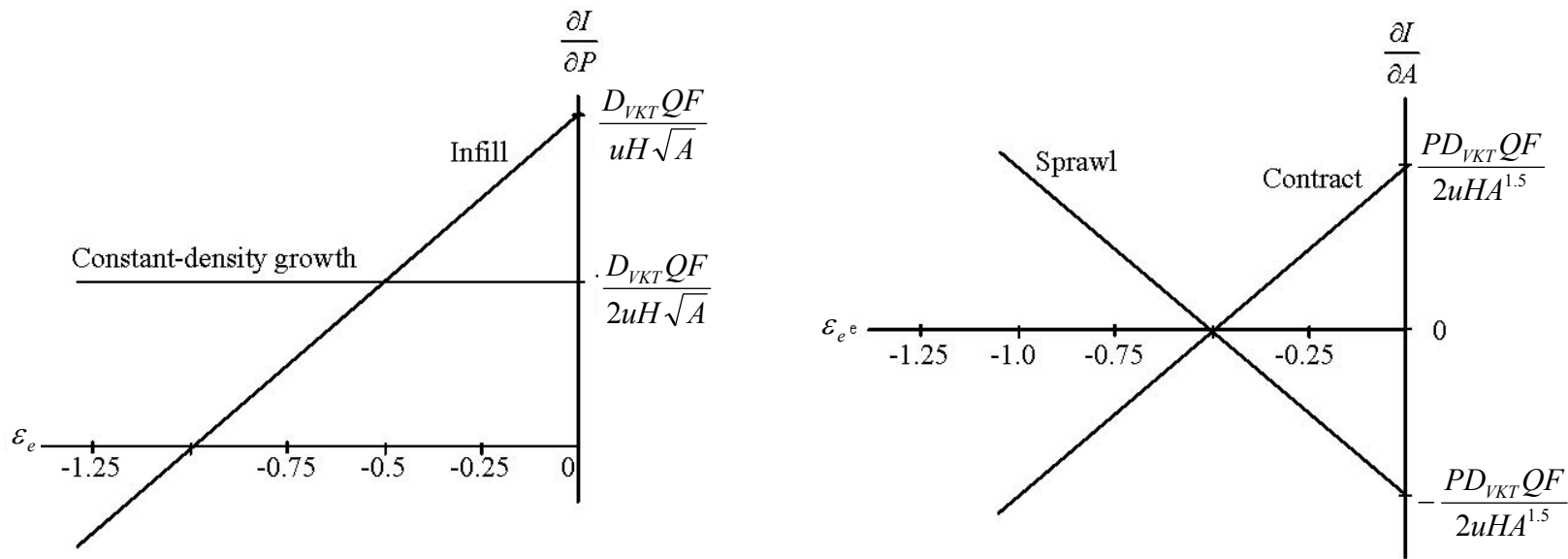
Figure 7-2: Influence of density-emissions elasticity (ε_e) on the incremental change in per capita intake (I) with respect to a change in urban population (P) or urban area (A). The left plot ($\partial I/\partial P$) shows the impact of increasing population on intake when urban land area is constant (infill) and when population density is constant (constant-density growth). The right plot ($\partial I/\partial A$) shows the impact of increasing (sprawl) and decreasing (contraction) urban land areas on intake when population is constant. In each plot, the change in urban form that minimizes intake is the lower line. A negative value on the ordinate axis indicates an absolute reduction in I.
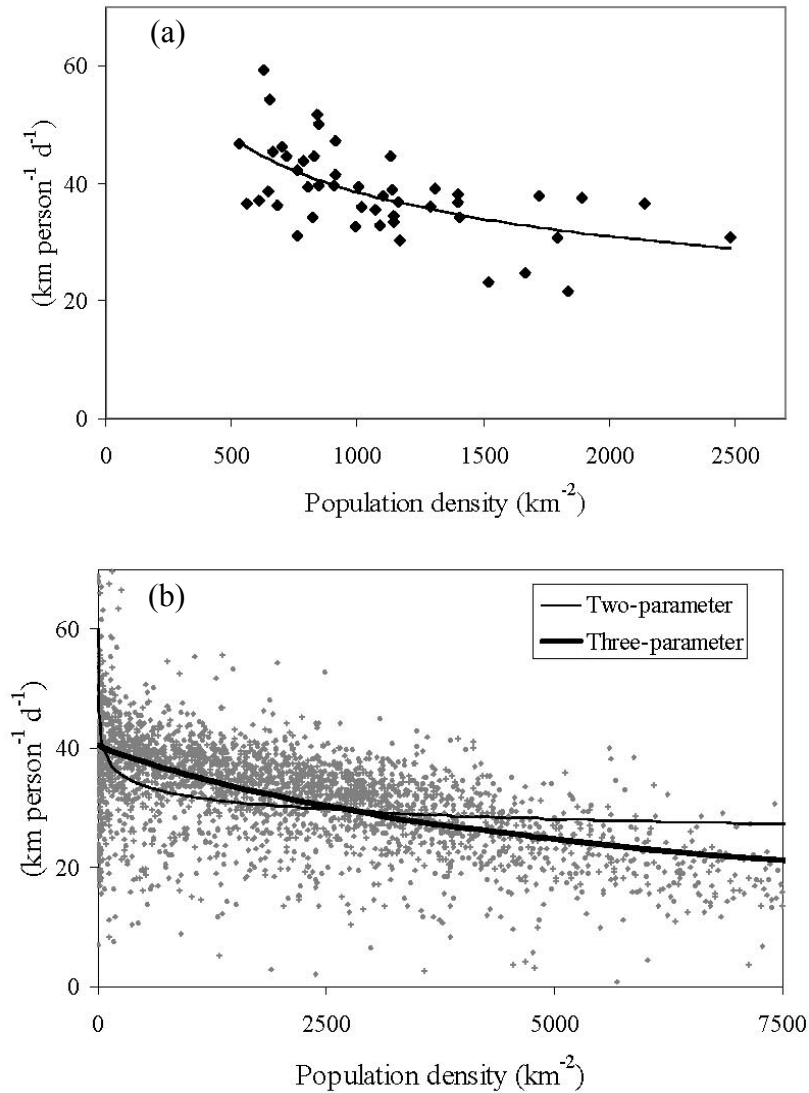
Figure 7-3: Comparisons of population density and average daily per capita vehicle-km traveled. Figure 7-3a presents data for the 47 urban areas in the US with population exceeding 750,000. For this dataset, the two- and three-parameter regression lines are indistinguishable. Figure 7-3b presents data for the 2,834 Traffic Analysis Zones in the Chicago, Los Angeles and San Francisco metropolitan areas. Not plotted are the 5% of the population density values that are greater than 7,500 km^{-2} and the 0.8% of the VKT values that are greater than 65 km $person^{-1}$ d^{-1}. Both datasets show an inverse relationship, with more dense areas having lower per capita VKT.

Figure 7-4: Density-VKT elasticity as a function of population density, based on data for the 2,834 Traffic Analysis Zones in the Chicago, Los Angeles and San Francisco metropolitan areas. Elasticity is independent of density with the two-parameter regression. With the three-parameter regression, elasticity is seen to increase in magnitude as population density increases.

Chapter 8: Conclusions

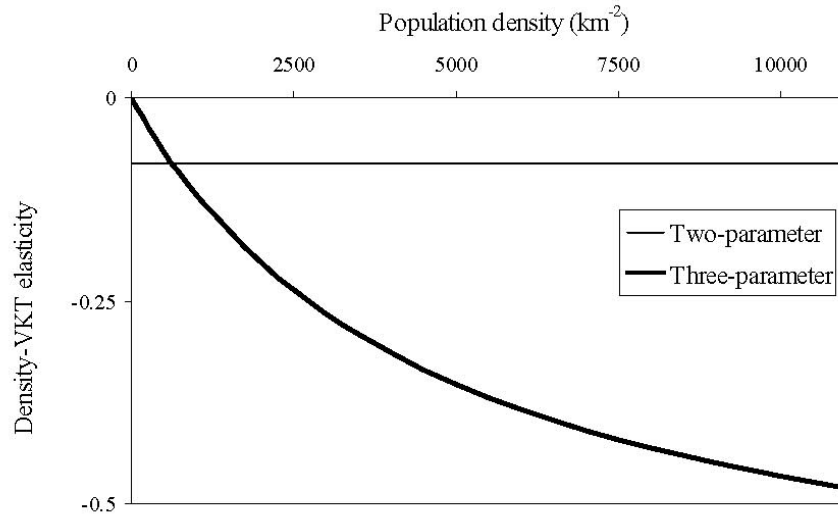This dissertation explores several aspects of human inhalation intake of urban air pollutants, focusing mainly on vehicle emissions. In this chapter, I summarize the findings from each previous chapter, discuss possible topics for future research, and then offer closing remarks.

Summary

In Chapter 1, I provide motivation and background for the topics being explored. In Chapter 2, I introduce intake fraction as an emerging intake metric useful for health risk assessment and other science and policy analyses. Intake fraction, which is the fraction of emissions that are taken in (inhaled), is a measure of the "exposure efficiency" for a specific pollutant and source. In Chapter 3, I estimate the intake fraction for vehicle emissions in California's South Coast Air Basin. The South Coast is an important case study because of the large population size (~15 million people) and because of the relatively high air-pollution levels. The investigation incorporates time-activity pattern data, an analysis of microenvironments such as in-vehicle and in-residence with an attached garage, and ambient monitoring station data on benzene and carbon monoxide during 1996-1999. The resulting estimate indicates that for every tonne of a primary, nonreactive pollutant emitted by a vehicle in the South Coast, ~50 grams are inhaled. The South Coast is an important case study, but it is not representative of typical US urban

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areas. In Chapter 4, I explore intake fraction for vehicle emissions in urban areas throughout the US. The reported median value, 14 per million, is ~30% of the value of ~50 per million reported for the South Coast. Analyzing recent tracer-gas experiments in Chapter 5, I estimate that the self-pollution intake fraction for school bus emissions is ~30 per million. Based on these values, self-pollution – a micro-scale, vehicle-specific issue – is expected to be a significant contributor to the total health impact of school bus exhaust.

The following comparison highlights the utility of these intake fraction results. Consider the three groups that inhale the exhaust from a specific school bus operating in an urban area: (1) the students on the bus, (2) people in vehicles near the bus, and (3) all other people in the urban area. The mass of the bus's pollution inhaled by the students on the bus is comparable to, or in many cases greater than, the mass of the bus's pollution inhaled by the latter two groups. For nonreactive gaseous emissions, these three groups inhale ~30 g, ~3 g, and ~13 g, respectively, per million grams emitted. For $PM_{2.5}$ these three groups are estimated here to inhale ~30 g, ~3 g, and ~9 g, respectively, per million grams emitted. For the third group, intake fraction is lower for $PM_{2.5}$ than for nonreactive gaseous emissions (9 per million versus 13 per million) because of the protection that buildings offer for particles. These calculations are derived from values in the Chapter 2 Appendix, from the ambient vehicle intake fraction of 14 per million reported in Chapter 4, and from the self-pollution intake fraction of ~30 per million presented in Chapter 5. The values given here are intended to be illustrative rather than definitive.

This comparison, illustrated for diesel $PM_{2.5}$ in cartoon format in Figure 8-1, yields two important findings. First, on average, a given mass of diesel emission

reduction is expected to have approximately three times more health benefit if it comes from school buses than if it comes from vehicles without self-pollution (43 versus 16 g per tonne for gaseous pollutants; 39 versus 12 g per tonne for $PM_{2.5}$). Identifying this three-fold increase in effectiveness required investigating inhalation intake rate, rather than only emissions or ambient concentrations. Second, reducing self-pollution offers the potential to be a relatively simple and straightforward way to reduce inhalation of school bus emissions, even without necessarily reducing school bus emissions. Efforts to reduce self-pollution directly could significantly reduce total inhalation intake of school bus emissions, even if emissions were to remain constant.

As I discuss in Chapter 2, the utility of intake fraction values increases when one can compare values across sources and situations. The results presented in this work offer progress towards a compendium of intake fraction values for a variety of sources and emission scenarios.

In Chapter 6, I estimate air pollution inhalation for 25,068 travel survey respondents in the South Coast. This investigation represents a new and promising method for exposure analysis. It uses individuals' latitude and longitude to track the nearby modeled ambient concentration; microenvironment factors and ambient concentration to estimate the exposure concentration; and, activity level to determine breathing rate. Inhalation intake rate is the product of breathing rate and exposure concentration. In Chapter 6, I explore the importance of the following three issues when estimating an individual's inhalation intake of air pollution. (1) People often change location during each day, as they go to work, shop, etc.; this movement is not accounted for in conventional exposure assessments, such as those that use US Census data to

determine location. (2) People's breathing rates are correlated with ambient concentrations of many urban air pollutants: both are typically higher during the day, when people are active, than at night, when people tend to be asleep. Ignoring this correlation, for example by assuming a constant breathing rate over time, may bias downward estimates of inhalation intake. (3) People spend time indoors and in vehicles, where the exposure concentration for pollutants emitted from vehicles may differ from the nearby ambient concentration. Depending on the pollutant, average exposure concentrations may be greater than or less than average ambient concentrations. These results are useful to exposure modelers, who would like to know what level of detail is necessary when building an exposure model, and what level of error may be introduced by not incorporating one or more of these three issues. Among the five pollutants investigated, the three issues considered (mobility, time-varying breathing rates, and microenvironments) influence the estimated intake rate by up to a factor of 2. This chapter also briefly explored correlations between individuals' inhalation intake rate and ethnicity and income category. These results, indicating total inhalation intake rate for the case study population and for specific subpopulations (e.g., specific ethnic groups), are useful to air quality managers, health risk assessors, and public health officials who seek to design effective intervention strategies to reduce the health effects of air pollution. Using the approach presented in this chapter, one could track changes over time in the per capita air pollution inhalation rate and in the distributions among the population of these intake rates. One could also model the effect on intake rates and intake rate distributions of potential transportation planning options or potential air pollution control strategies.

Considering an idealization of a hypothetical urban area, I explore in Chapter 7 the impact of changes in urban population and land area on inhalation intake of vehicle emissions. I find that in some cases, a specific change in urban form (e.g., infill development when the density-emissions elasticity is negative and is greater than 0.5 in magnitude) may reduce per capita vehicle emissions yet increase per capita inhalation of these emissions. The investigation and results are potentially useful and important to establishing and motivating urban planning as a tool for air quality management. In addition, these results may be beneficial to air quality managers evaluating technical and policy options for meeting air quality objectives.

In this dissertation, I present new methods and new insights about population inhalation of vehicle emissions. Broadly, the results suggest that air quality engineers and managers can and should consider inhalation of air pollution, not only emissions or ambient concentrations. For example, the comparison of intake fraction values for school buses and other vehicles yielded suggestions for effective interventions to reduce exposures. Further investigation of the emission-to-intake relationship will likely yield additional potential intervention strategies. The emerging field of exposure science is developing tools, metrics, and approaches that are ready to be integrated more fully into air quality research and management. Doing so will make the field more rigorous and more relevant.

Future research

Below, I highlight areas for further research that are suggested by the investigations in this dissertation.

Future investigations could extend the exploration in Chapter 2 of the policy implications of intake fraction. One next step would be to generate intake fraction estimates for all emission sources in a specific area. One could then rank emission sources in terms of (1) mass emissions, and (2) mass inhaled, which is estimated as the product of emissions and intake fraction. This approach could help environmental policy focus on reducing inhalation intake of air pollution. Another next step would be to incorporate intake fraction estimates into pollution trading regimes. The marginal impacts of emissions vary in space and time, yet most pollution markets do not account for this variability. The idea of "trading ratios" has been suggested, whereby the value of an emission credit would depend on the time and location of the emission or emission reduction. Intake fraction values could offer a useful basis for determining trading ratios.

In Chapter 3, I calculate intake fraction of vehicle emissions in the South Coast. Several other intake fraction investigations are worthwhile. For example, the approach I employed in Chapter 3 could be applied to vehicle emission in other urban areas, including areas outside the US. I have focused on vehicle emissions; future work should explore other source categories, such as large point sources, area sources, and indoor releases. In this dissertation, intake fraction has only been applied to primary pollutants. Future research could extend the intake fraction concept to secondary pollutant such as ozone, which is formed as a result of chemical reactions among precursor emissions.

An analysis that is missing from the investigations in this dissertation is a robust comparison against measured exposures. Calculating intake fraction from measured exposures is challenging because most pollutants have multiple sources. Direct measurement of intake fraction is not possible for most sources. Nevertheless, estimates

could be made from available evidence. For example, to estimate the intake fraction for vehicle benzene, one could start with measured benzene exposures, and then subtract exposures attributable to non-vehicle sources (e.g., environmental tobacco smoke). This approach could be used to corroborate the results presented in Chapter 3 for vehicle benzene intake fraction in the South Coast.

School bus self-pollution was quantified in Chapter 5, but in general self-pollution has not been well studied. There is the potential that school bus self-pollution could be easily addressed through minor structural modifications to the buses, but more work is needed to generate and test potential ideas. The range of values for self-pollution in private passenger vehicles is important but unknown. Self-pollution is an idea that extends beyond vehicles; for example, residential woodstoves likely exhibit self-pollution.

In Chapter 6, I explored the impact on estimated intake rate of three exposure assessment attributes: microenvironments, time-varying breathing rates, and mobility. The first issue is the topic of significant research, but the second and third issues had received almost no attention before this dissertation. These topics are an important gap in the literature. One basic piece of information that is missing from the literature is the diurnal profile of population breathing rate. I presented two estimates for this information (Chapters 3 and 6). A more robust investigation would be reasonably straightforward (i.e., about the scope of a journal article) and highly useful to the exposure analysis community.

In Chapter 7, I explored the impact on a hypothetical urban area of changes in urban population and land area. This investigation suggests the idea of exploring how

intake fraction has changed over time in specific urban areas. I have done an preliminary investigation into this topic, and found that over time scales of several decades, vehicle intake fraction increases in some urban areas and decreases in others, but the average intake fraction value is relatively constant over time. More work is necessary to further explore this topic. The research in Chapter 7 highlighted the need to better understand two parameters and their determinants: the density elasticity of emissions and the density elasticity of vehicle-km traveled. In addition, these elasticity values have been estimated for private passenger vehicles but not for diesel vehicles. Given the importance of diesel emissions to air quality and health, these elasticity parameters should be explored for diesel vehicles.

Transportation planning and inhalation of vehicle emissions

Globally, many cities are growing rapidly. Urban areas in developing countries are increasingly interested in improving air quality. As these trends continue, demand for urban-scale air quality solutions will increase. Consider, for example, three options that may help meet air quality objectives: reducing emissions from private passenger vehicles via technological changes (e.g., fuel reformulation), increasing availability of mass transit, and using transportation and land use planning to reduce transportation demand (as indicated, for example, by average annual distance traveled per capita in a specific urban area). For an urban area prepared to spend money and political capital improving air quality, which option offers the most effective way to reduce human inhalation intake of air pollution? The ideas explored in this dissertation provide useful background and strategies for framing and exploring this question. While reducing passenger vehicle

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emissions has been a main focus of air quality management in the US, the cost-effectiveness of other options such as improved mass transit should also be considered, especially in rapidly urbanizing developing countries (McKinley et al., 2005).

Broadly, air quality management may be divided into technical (i.e., technology-based) and nontechnical options. Technical options include catalytic converters, on-board diagnostics, and fuel reformulation. Nontechnical options, such as urban planning and cross-subsidies for public transportation, aim to shift individuals' behavior. Technical changes to engines and fuels have successfully reduced per-mile emissions for many pollutants. However, there are several concerns with air quality management approaches that focus almost exclusively on technical options to the exclusion of nontechnical options. First, it remains an open question whether opportunities for easy improvement have been used up. Second, emission reduction equipment, when deployed at the scale of millions of cars, sometimes fails to perform as well as initially predicted. Third, technical options have not reduced total fuel consumption or carbon dioxide emissions (though hybrids offer the potential to do so, or at least slow the growth in consumption).

Technical options are often considered to offer immediate and verifiable change, unlike many nontechnical options (e.g., changes in urban form occur over times scales of decades). To some extent this is true: a technology such as a catalytic converter can change a vehicle's emissions as soon as implemented (and, at typical vehicle turn-over rates, within several years for a fleet), offering immediate air quality benefits. The effectiveness of a catalytic converter can be tested in a laboratory. At the same time, however, technologies often perform better in laboratory tests than when used in the "real world." In addition, the time scale and effort necessary to deploy functioning control

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technologies to a large proportion of a vehicle fleet can be multiple decades. For example, EPA began serious efforts to control diesel emissions in the 1990's, following the passage of the 1990 Clean Air Act Amendments. Final diesel rules were passed in 2000 and 2001, and standards for engines and fuels will take effect during 2006 – 2012 (http://www.epa.gov/cleandiesel). Fleet turnover is anticipated to take one or two decades more. Thus, the time scale for this technical shift, multiple decades, is comparable to the time scale that would be necessary for shifts in urban form. Urban form should not be dismissed out of hand as requiring too much time, relative to technical options, to cause change. Urban planning and other approaches that aim to shift individuals' behavior have the potential to be more cost effective than technical options, and they should be one tool among many used to improve urban air quality.

Urban engineering and sustainability

Broadly, the research in this dissertation is situated in the field of urban engineering, which has as its objective designing urban infrastructure systems that improve human health and the environment and promote quality of life. The core tools for this field are derived from civil and environmental engineering, public health, and urban planning. In US universities, these three fields are typically separate departments, but alternatives are possible. For example, at the University of Tokyo, considered to be the leading university in Japan, the Department of Urban Engineering, formed in 1962, combines urban planning with environmental engineering (http://www.due.t.u-tokyo.ac.jp).

Three major transportation air pollution challenges are (1) to reduce the total health impacts of vehicle exhaust, (2) to address distributional issues among subpopulations, and (3) to reduce or offset climate change impacts of transportation energy consumption. Transforming current motor vehicles into ones with no use-phase toxic emissions would address two of these three issues, although upstream health impacts (e.g., at a power plant for electric cars) may continue to be of substantial concern. Transforming the basis of entire energy supply system from fossil fuels to renewable energy would address all three issues, assuming low lifecycle emissions from alternative energy supply technologies.

Reducing urban air pollution is an important and worthwhile goal for improving the quality of life for urban residents. I have written elsewhere about my views on the difference between sustainability and quality of life improvements (Marshall and Toffel, 2005). I believe that (1) goals for our society extend well beyond merely sustaining itself. Technological and urban systems can and must strive towards constant improvement, addressing flaws such as urban air pollution. (2) Population is a key component of sustainability. Any nonzero level of per-capita energy consumption is ultimately unsustainable if population grows without bound. (3) The challenge is to transform the society and technologies we currently have into those we want to have and need to have to be sustainable. I believe that the future is not predetermined, but rather is a result of our collective action (or inaction). Our actions are influenced by the available information, by social values and norms, and by the physical limitations of the global environment. With finite resources, we will need to make tradeoffs as we decide which path to take to get from where we are to where we want to be. The insights offered in this

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dissertation contribute some drops to the sea of knowledge that will help us make this

transition.
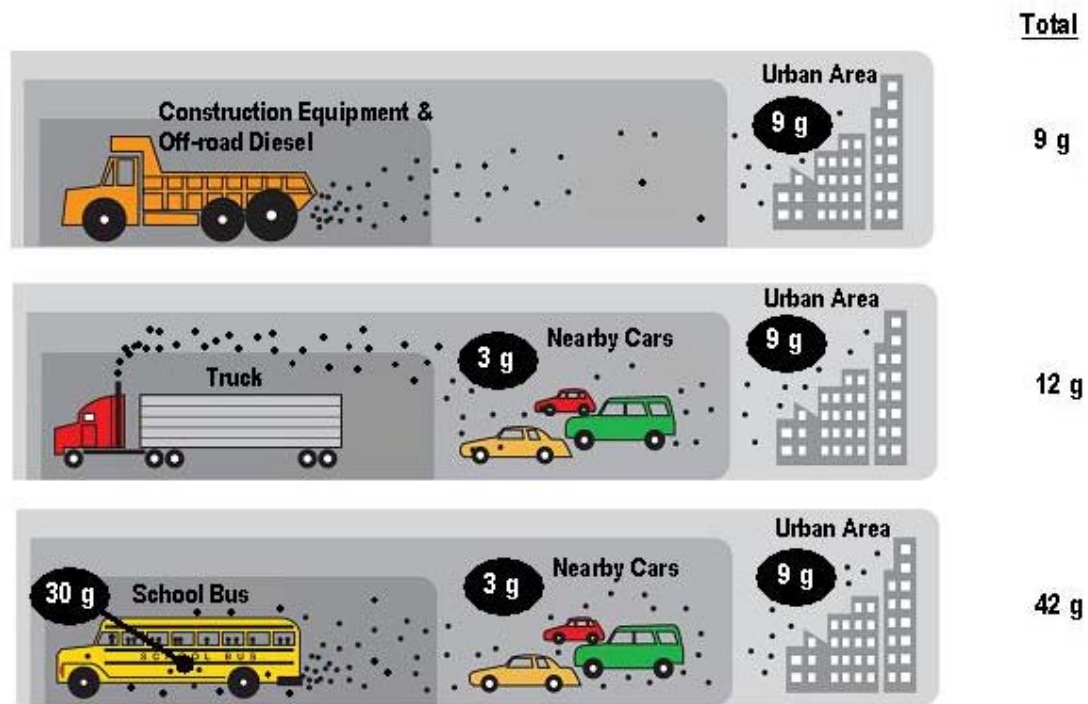
Figure 8-1: Diesel PM$_{2.5}$ intake fraction for three mobile sources. Values shown are the mass of PM$_{2.5}$ emissions inhaled in different locations, per million grams emitted. For example, owing to self-pollution, ~ 30 grams are inhaled by students on a school bus, per million grams emitted. For each of the three source types, there will some residences near the source and some further away. On average, the cumulative intake fraction for the whole exposed population is ~ 9 per million for US urban areas. Values are approximate and illustrative, rather than definitive. Illustration courtesy of The Linus Group, Berkeley, CA.

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