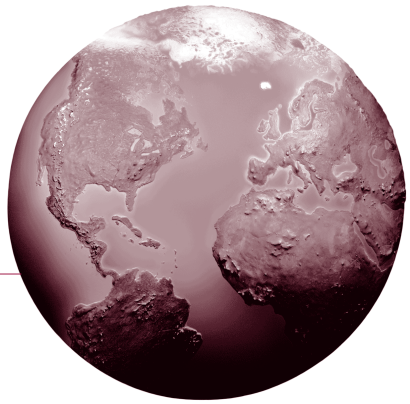

HEALTH EFFECTS INSTITUTE

Number 130 Part I
November 2005

RESEARCH REPORT

Relationships of Indoor, Outdoor, and Personal Air (RIOPA)

Part I. Collection Methods and Descriptive Analyses

Clifford P Weisel, Junfeng (Jim) Zhang, Barbara J Turpin, Maria T Morandi, Steven Colome, Thomas H Stock, Dalia M Spektor, and Others

Includes a Commentary by the Special Review Panel

Number 7
November 2005

MICKEY LELAND NATIONAL URBAN AIR TOXICS RESEARCH CENTER

HEALTH
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The Health Effects Institute was chartered in 1980 as an independent and unbiased research organization to provide high quality, impartial, and relevant science on the health effects of emissions from motor vehicles, fuels, and other environmental sources. All results are provided to industry and government sponsors, other key decisionmakers, the scientific community, and the public. HEI funds research on all major pollutants, including air toxics, diesel exhaust, nitrogen oxides, ozone, and particulate matter. The Institute periodically engages in special reviews and evaluations of key questions in science that are highly relevant to the regulatory process. To date, HEI has supported more than 220 projects at institutions in North America, Europe, and Asia and has published over 160 Research Reports and Special Reports.

Typically, HEI receives half of its core funds from the US Environmental Protection Agency and half from 28 worldwide manufacturers and marketers of motor vehicles and engines who do business in the United States. Other public and private organizations periodically support special projects or certain research programs. The research reported here was funded with the Mickey Leland National Urban Air Toxics Research Center. Regardless of funding sources, HEI exercises complete autonomy in setting its research priorities and in reaching its conclusions.

An independent Board of Directors governs HEI. The Institute's Health Research Committee develops HEI's five-year Strategic Plan and initiates and oversees HEI-funded research. The Health Review Committee independently reviews all HEI research and provides a Commentary on the work's scientific quality and regulatory relevance. Both Committees draw distinguished scientists who are independent of sponsors and bring wide-ranging multidisciplinary expertise.

The results of each project and its Commentary are communicated widely through HEI's home page, Annual Conference, publications, and presentations to professional societies, legislative bodies, and public agencies.

The Mickey Leland National Urban Air Toxics Research Center (NUATRC or the Leland Center) was authorized under the Clean Air Act Amendments of 1990 and established in 1991 to develop and support research into potential human health effects of exposure to air toxics in urban communities. The Center released its first Request for Applications in 1993. The aim of the Leland Center has been to build a research program structured to investigate and assess the risks to public health that may be attributed to air toxics. Projects sponsored by the Leland Center are designed to provide sound scientific data useful for researchers and for those charged with formulating environmental regulations.

The Leland Center is a public–private partnership in that it receives support from government sources and from the private sector. Thus, government funding is leveraged by funds contributed by organizations and businesses, which enhances the effectiveness of the funding from both stakeholder groups. The US Environmental Protection Agency has provided the major portion of the Center's government funding to date; a number of corporate sponsors, primarily in the chemical and petrochemical fields, have also supported the program.

A nine-member Board of Directors oversees the management and activities of the Leland Center. The Board also appoints the thirteen members of a Scientific Advisory Panel who are drawn from government, academia, and industry. These members represent such scientific disciplines as epidemiology, biostatistics, exposure assessment, toxicology, and medicine. The Scientific Advisory Panel provides guidance in formulating the Center's research program and conducts peer reviews of results from the Center's completed projects.

The Leland Center is named for the late United States Congressman George Thomas "Mickey" Leland from Texas who sponsored and supported legislation to reduce the problems of pollution, hunger, and poor housing that unduly affect residents of low-income urban communities.

MICKEY LELAND NATIONAL URBAN AIR TOXICS RESEARCH CENTER

STATEMENT

Synopsis of the RIOPA Research Report Part I

HEI HEALTH EFFECTS INSTITUTE

Pollutants in Indoor, Outdoor, and Personal Air: Collection Methods and Descriptive Analyses

BACKGROUND

Urban populations are exposed to a complex mixture of possibly toxic pollutants generated and emitted by a variety of outdoor and indoor sources. These pollutants occur naturally or result from human activities; they may be present in the form of gases, liquid droplets, or solid particles. The US Environmental Protection Agency (EPA) defines an air toxic as any substance known or suspected to cause harm to humans or the environment. The Clean Air Act Amendments of 1990 list 188 air toxics as hazardous air pollutants; these include volatile organic compounds (VOCs), carbonyls (aldehydes and ketones), and components often associated with particulate matter (PM). The Amendments require the EPA to evaluate the possible health risks from air toxics and, if appropriate, control their ambient levels. To achieve this objective, the EPA identified pollutants that may be most hazardous to health and categorized them as urban air toxics (emitted from all sources) or mobile-source air toxics; some pollutants appear on both lists. Currently, the EPA regulates ambient levels of fine PM through the National Ambient Air Quality Standards for $PM_{2.5}$ (PM of 2.5 µm or smaller).

Understanding personal exposures to both air toxics and PM—and how different sources contribute to individual exposures—has been considered an important first step in assessing the possible public health risks from these species in the urban environment. The Relationships of Indoor, Outdoor, and Personal Air (RIOPA) study was designed to provide such information for a large number of VOCs and carbonyls, including some that are listed as urban and mobile-source air toxics, and for $PM_{2.5}$.

APPROACH

The investigators measured indoor, outdoor, and personal exposure concentrations of 16 VOCs, 10 carbonyls, and $PM_{2.5}$ during two 48-hour sampling periods in different seasons between the summer of 1999 and the spring of 2001. The study included 100 homes with 100 adult residents in each of three cities with different air pollution sources and weather conditions: Los Angeles CA, Houston TX, and Elizabeth NJ. Homes were selected by distance from various sources.

In this report the investigators (1) compare concentrations of the pollutants measured in indoor, outdoor, and personal air (within the subject's breathing zone), and in vehicles for carbonyls; (2) examine the effects of city, season, type of home, and other variables on measured concentrations; and (3) quantify how much outdoor sources contributed to the indoor concentrations using measurements of outdoor–indoor air exchange rates.

The VOCs measured include

- some on the EPA's list of urban air toxics (benzene, carbon tetrachloride, chloroform, trichloroethylene);
- some on the EPA's list of mobile-source air toxics (benzene, chloroform, ethyl benzene, MTBE, *m- & p*-xylenes, *o*-xylene, styrene, and toluene); and
- some that originate primarily from indoor sources (α-pinene, β-pinene, and *d*-limonene).

The carbonyls measured include

- some from the EPA's lists of urban air toxics and mobile-source air toxics (acetaldehyde and formaldehyde);
- several that are present at low levels in mobile-source emissions (acrolein, butyraldehyde, crotonaldehyde, hexaldehyde, isovaleraldehyde, propionaldehyde, and valeraldehyde); and
- two that are primarily formed as a result of photochemical reactions with hydrocarbons (glyoxal and methylglyoxal)

The investigators used passive organic vapor monitors to collect VOC samples. For carbonyls,

This Statement, prepared by the Health Effects Institute and the National Urban Air Toxics Research Center, summarizes a research project funded jointly by HEI and NUATRC. It was conducted by Drs Clifford P Weisel, Junfeng (Jim) Zhang, and Barbara J Turpin of the Environmental and Occupational Health Sciences Institute, Piscataway NJ. The following Research Report (HEI Number 130; NUATRC Number 7) contains both the detailed Investigators' Report and a Commentary on the study prepared by a Special Review Panel from both funding organizations.

they used two sampling methods: a conventional active sampler and a new passive sampler that was developed as part of the study. The new sampler performed better for several carbonyls and was used most; therefore the Investigators Report presents only the analyses and conclusions based on the passive samples. For $PM_{2.5}$, indoor and outdoor samples were collected on filters mounted in a Harvard impactor; personal samples were collected on smaller filters mounted in a personal monitor.

RESULTS AND INTERPRETATION

The homes and subjects selected did not proportionally represent the greater population. Rather, homes close to sources were preferentially sampled in order to examine the impact of possibly high exposures. In addition, the characteristics of the subjects and the homes differed among cities. Thus comparing results among the three areas, extrapolating the numeric results obtained in this study to the general population, or attributing them to a given city or region must be considered with caution.

The analyses of the aggregate data suggest some trends that will need to be verified with more detailed analyses. With a few exceptions, mean and median personal exposure and indoor concentrations of VOCs and carbonyls were higher than the outdoor concentrations within each city and for the whole data set. Personal $PM_{2.5}$ concentrations were higher than indoor and outdoor concentrations. The finding that personal exposure concentrations were higher than outdoor concentrations for many compounds indicates that indoor sources contribute to, and in some cases dominate, personal exposures; this is consistent with results from other studies.

Several VOCs were present only at low levels in all environments and were not detected in many outdoor samples. The species detected in more than 60% of outdoor samples common to all three cities were MTBE, carbon tetrachloride, benzene, ethyl benzene, *m- & p*-xylenes, and *o*-xylene. MTBE had the highest outdoor concentrations. Although cities with different types of sources were chosen and homes near sources were preferentially sampled, the ranges of outdoor VOC concentrations were generally similar in the three cities. The median outdoor concentrations of carbonyls were more variable than VOCs across the cities (with the exception of formaldehyde).

Indoor concentrations of several VOCs and carbonyls differed among cities. The species with the highest indoor concentrations were the VOCs MTBE, toluene, α-pinene, and *d*-limonene and the carbonyls formaldehyde, acetaldehyde, and acetone. Personal exposure concentrations for several VOCs and some carbonyls also differed among cities.

Among the three cities, differences in indoor and outdoor $PM_{2.5}$ levels were slight, but differences in personal $PM_{2.5}$ exposures were more pronounced.

The analyses of the outdoor contributions to indoor air suggested that some VOCs (MTBE, benzene, carbon tetrachloride, and trichloroethylene) were primarily generated outdoors and contributed 90% to 100% of the indoor concentrations. Outdoor concentrations of other VOCs (chloroform, α-pinene, β-pinene, and *d*-limonene) and most carbonyls (including formaldehyde, acetaldehyde, and hexaldehyde) contributed less to indoor air (13% to 43% of indoor concentrations). The carbonyls that contributed most were acrolein, crotonaldehyde, and propionaldehyde (50% to 63%). For $PM_{2.5}$, outdoor air contributed 60% of the indoor concentration.

CONCLUSIONS

The RIOPA study generated a rich database on the concentrations of air toxics and $PM_{2.5}$ for a large number of subjects and their homes. Few investigators have looked at personal, indoor, and outdoor concentrations of a suite of VOCs, carbonyls, and $PM_{2.5}$ in a large set of subjects in multiple urban centers. (The information on $PM_{2.5}$ composition [published as Part II of this Research Report] provides needed information about exposure to the components of PM.)

With a few exceptions, median indoor, outdoor, and personal air concentrations of the various compounds were similar for the three cities. This was unexpected given the wide variety of pollutant sources. Both the higher concentrations of species in personal samples compared with outdoor samples and the contributions of outdoor air to indoor concentrations of each species confirm and extend earlier findings.

Future analyses of this data set will help clarify the impact of proximity to sources and the individual factors associated with high personal exposure levels. Overall, the data collected in the RIOPA study increase the database on the distribution of concentrations for many air toxics and $PM_{2.5}$ and supply data for assessing whether these levels are of health concern.

Cameographics, Union ME, Compositor. Printed at Capital City Press, Montpelier VT.
Library of Congress Catalog Number for the HEI Report Series: WA 754 R432.
The paper in this series meets the minimum standard requirements of the ANSI Standard Z39.48-1984 (Permanence of Paper) effective with Report 21 in December 1988; and effective with Report 92 in 1999 the paper is recycled from at least 30% postconsumer waste with Reports 25, 26, 32, 51, 65 Parts IV, VIII, and IX, 91, 105, 117, and 128 excepted. These excepted Reports are printed on acid-free coated paper.

MICKEY LELAND NATIONAL URBAN AIR TOXICS RESEARCH CENTER

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Environmental and Occupational Health Sciences Institute, Piscataway NJ; School of Public Health and Robert Wood Johnson Medical School, University of Medicine and Dentistry of New Jersey, Piscataway NJ; Department of Environmental Sciences, Rutgers University, New Brunswick NJ; School of Public Health, University of Texas, Houston TX; Integrated Environmental Science, Irvine CA; and Rand Corporation, Santa Monica CA

STATEMENT

This Statement by the Special Review Panel is a nontechnical summary of the Investigators' Report and the Commentary.

PREFACE

INVESTIGATORS' REPORT

When the study was completed, the investigators submitted a final report to the funding organizations. The Investigators' Report was first examined by ouside technical reviewers and a biostatistician. The report and the reviewers' comments were then evaluated by members of the Special Review Panel, who had no role in selecting or managing the project. During the review process, the investigators had an opportunity to exchange comments with the Special Review Panel and, if necessary, revise the report.

Continued

COMMENTARY Special Review Panel

The Commentary about the Investigators' Report was prepared by a Special Review Panel from both funding organizations and their staffs. Its purpose is to place the study into a broader scientific context, to point out its strengths and limitations, and to discuss remaining uncertainties and implications of the findings for public health.

RELATED HEI AND NUATRC PUBLICATIONS

Publishing history: This document was posted as a preprint on *www.healtheffects.org* and on *www.sph.uth.tmc.edu/mleland* and then finalized for print.

Citation for whole document:

Weisel CP, Zhang J, Turpin BJ, Morandi MT, Colome S, Stock TH, Spektor DM, and Others. November 2005. Relationships of Indoor, Outdoor, and Personal Air (RIOPA): Part I. Collection Methods and Descriptive Analyses. HEI Research Report 130; NUATRC Research Report 7. Heath Effects Institute, Boston MA; Mickey Leland National Urban Air Toxics Research Center, Houston TX.

When specifying a section of this report, cite it as a chapter of the whole document.

PREFACE to *Relationships of Indoor, Outdoor, and Personal Air (RIOPA)*

Relationships of Indoor, Outdoor, and Personal Air (RIOPA) is a study funded jointly by NUATRC and HEI. It was designed to provide information about the concentrations of volatile organic compounds (VOCs), carbonyls, and particulate matter (PM) in indoor, outdoor, and personal air samples for adults and children living in three urban centers with different pollutant sources and weather. It is composed of three related projects separately funded.

In December of 1996, NUATRC issued Request for Applications 96-01, "Personal Exposures to Air Toxics in Urban Environments". This Request invited research that would help to understand (1) personal exposures to air toxics and PM, and (2) how those exposures relate to daily activities and to outdoor and indoor sources of pollutants. In response, Clifford Weisel (at the University of Medicine & Dentistry of New Jersey and at the Environmental and Occupational Health Sciences Institute [EOHSI]) proposed to monitor indoor, outdoor, and personal exposures to VOCs in 100 homes with 100 adult subjects and 50 children in each of three cities: Elizabeth NJ, Houston TX, and Los Angeles CA. The proposal also included, for half the homes, measurements of indoor and outdoor concentrations of some aldehydes and PM with an aerodynamic diameter of 2.5 µm or less ($PM_{2.5}$). Coinvestigators were Junfeng (Jim) Zhang (affiliated with the same institutions); Barbara Turpin (at EOHSI and Rutgers University); Thomas Stock and Maria Morandi (at the University of Texas), Steven Colome (Integrated Environmental Services), and Dalia Spector (Rand Corporation). This first study was funded by NUATRC in 1997.

Also in 1997, HEI issued RFA 97-2, "Assessing Personal Exposure to Selected Aldehydes Using Chemical and Biological Techniques", seeking studies to define human exposure to several environmental aldehydes through the use of area or personal monitors. In 1998 HEI funded Dr Junfeng (Jim) Zhang of EOHSI as principal investigator to expand the Weisel study by (1) increasing the number of carbonyl compounds measured, (2) collecting samples for carbonyls indoors and outdoors for the remaining half of the homes, and (3) adding personal samples of carbonyls for all subjects and inside vehicles.

In 1998, HEI issued RFA 98-1A, "Characterizing Exposure to Particulate Matter", requesting studies to characterize personal exposure to PM in different indoor and outdoor environments and geographic locations and also to determine the composition of these particles. That year HEI funded Dr Barbara Turpin of Rutgers University as principal investigator to (1) add measurements of $PM_{2.5}$ in personal air samples for the subjects in the 50 homes for which Dr Weisel had collected indoor and outdoor samples, and (2) measure the composition of the particles in all indoor, outdoor, and personal air samples collected.

Because the two HEI studies complemented and extended the initial NUATRC study, the two organizations treated the three studies as one so that the data could be analyzed and presented in a coherent manner. Due to the large set of data and analyses, the Investigators' Final Report was divided into *Part I: Collection Methods and Descriptive Analyses* (for VOCs, carbonyls, $PM_{2.5}$ concentrations; this volume) and *Part II: Analyses of Concentrations of Particulate Matter Species* (the compositional analysis of $PM_{2.5}$; in press). The Investigators' Final Report was examined by external peer reviewers; the Report and the reviewers' comments were then evaluated by a Special Review Panel composed of members of the HEI Review Committee and the NUATRC Scientific Advisory Panel. The Special Review Panel developed the Commentary in collaboration with scientists from HEI and NUATRC.

SPECIAL REVIEW PANEL

John Bailar
University of Chicago, Emeritus; Scholar in Residence, National Academy of Sciences [NUATRC]

Annette Guiseppi-Elie
Principal Consultant, Du Pont Corporate Remediation Group [NUATRC]

Brian Leaderer
Susan Dwight Bliss Professor, Department of Epidemiology and Public Health, Yale University School of Medicine [NUATRC]

Edo Pellizzari
RTI Fellow, Analytical and Environmental Sciences, Director for Proteomics, RTI International [HEI]

Nancy Reid
University Professor, Department of Statistics, University of Toronto [HEI]

INVESTIGATORS' REPORT

Relationships of Indoor, Outdoor, and Personal Air (RIOPA)

Part I. Collection Methods and Descriptive Analyses

Clifford P Weisel, Junfeng (Jim) Zhang, Barbara J Turpin, Maria T Morandi, Steven Colome, Thomas H Stock, Dalia M Spektor, and Others

ABSTRACT

This study on the relationships of indoor, outdoor, and personal air (RIOPA*) was undertaken to collect data for use in evaluating the contribution of outdoor sources of air toxics and particulate matter (PM) to personal exposure. The study was not designed to obtain a population-based sample, but rather to provide matched indoor, outdoor, and personal concentrations in homes that varied in their proximity to outdoor pollution sources and had a wide range of air exchange rates (AERs). This design allowed examination of relations among indoor, outdoor, and personal concentrations of air toxics and PM across a wide range of environmental conditions; the resulting data set obtained for a wide range of environmental pollutants and AERs can be used to evaluate exposure models.

Approximately 100 households with residents who do not smoke participated in each of three cities in distinct locations expected to have different climates and housing characteristics: Elizabeth, New Jersey; Houston, Texas; and Los Angeles County, California. Questionnaires were administered to characterize homes, neighborhoods, and personal activities that might affect exposures. The concentrations of a suite of volatile organic compounds (VOCs) and carbonyl compounds, as well as the fraction of airborne particulate matter with a mass median aerodynamic diameter ≤ 2.5 µm ($PM_{2.5}$), were measured during continuous 48-hour sessions in which indoor, outdoor, and personal air samples were collected simultaneously. During the same 48-hour period, the AER (exchanges/hr; $x\ hr^{-1}$) was determined in each home, and carbonyl compounds were measured inside vehicle cabins driven by a subset of the participants. In most of the homes, measurements were made twice, during two different seasons, to obtain a wide distribution of AERs.

This report presents in detail the data collection methods, quality control measures, and initial analyses of data distributions and relations among indoor, outdoor, and personal concentrations. The results show that indoor sources dominated personal and indoor air concentrations of many measured VOCs and carbonyl compounds. For several measured species, personal concentrations were higher than either indoor or outdoor concentrations, indicating the presence of some sources closely related to personal activities. For some species there were no significant indoor sources in the majority of the homes; thus indoor concentrations were mainly determined by outdoor concentrations in these homes. The range of distributions of air concentrations for the measured VOCs, formaldehyde and acetaldehyde, $PM_{2.5}$, and AERs were generally consistent with values reported previously in the literature. Thus associations derived from or models based on this data set that may link the influence of outdoor sources with indoor air concentrations of air toxics and $PM_{2.5}$ can be relevant to other urban settings.

The simultaneous measurements of indoor concentrations, outdoor concentrations, AERs, and room volumes allowed the use of a mass balance model, under the steady-state approximation, to mechanistically examine the relative contributions of indoor and outdoor sources to measured indoor concentrations on a home-by-home basis. Estimated indoor source strengths for VOCs and carbonyl compounds varied widely from home to home, consistent

* A list of abbreviations and other terms appears at the end of the Investigators' Report.

This Investigators' Report is Part I of a Research Report published by the Health Effects Institute (Report 130) and the Mickey Leland National Urban Air Toxics Research Center (Report 7). The Report also includes a Commentary written by a Special Review Panel jointly selected by both organizations, a Preface, and a Statement synopsis of the research project. Correspondence concerning the Investigators' Report may be addressed to Dr Clifford P Weisel (*weisel@eohsi.rutgers.edu*) or Dr Junfeng (Jim) Zhang (*jjzhang@eohsi.rutgers.edu*), Environmental and Occupational Health Sciences Institute, Piscataway NJ 08854.

(Health Effects Institute) Although this document was produced with partial funding by the United States Environmental Protection Agency under Assistance Award R82811201 to HEI, it has not been subjected to the Agency's peer and administrative review and therefore may not necessarily reflect the views of the Agency, and no official endorsement by it should be inferred. The contents of this document also have not been reviewed by private party institutions, including those that support the Health Effects Institute; therefore, it may not reflect the views or policies of these parties, and no endorsement by them should be inferred.

(Mickey Leland National Urban Air Toxics Research Center) This project has been authorized by the Clean Air Act Amendments of 1990 (Title III, Section 301/p) and funded wholly or in part by the United States Environmental Protection Agency under Assistance Agreement R828678 to the Mickey Leland Center. The contents of this document do not necessarily reflect the views and policies of the Environmental Protection Agency, nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

with the indoor–outdoor concentration patterns, as shown in scatter plots. The indoor source estimations agreed with published values for $PM_{2.5}$ and with the general understanding of sources of VOCs and carbonyl compounds. The source strengths reported here, derived from hundreds of homes, are an important contribution to the literature on exposure to air toxics. For the first time for many compounds, these estimates present a cohesive set of measurements across a range of air toxics in paired indoor, outdoor, and personal samples along with AER and questionnaire results that can be used for future analyses of indoor air quality. The estimation of outdoor contributions to measured indoor concentrations provides insights about the relative importance of indoor and outdoor sources in determining indoor concentrations, the main determinant of personal exposure for most of the measured compounds.

In this report simple statistical tests mainly of the pooled data were used to analyze differences by sampling site, emission source type, season, home type, and home age. Paired adult–child personal concentrations within the same home were also compared using the pooled data set. These analyses generated some intriguing results that warrant more in-depth investigation in the future.

INTRODUCTION

BACKGROUND

The US Environmental Protection Agency (EPA) classifies a chemical as an air toxic (also termed hazardous air pollutant) if its presence in the atmosphere is associated with adverse health outcomes (Morello-Frosch et al 2000). Urban communities are often exposed to a complex mixture of air toxics that includes compounds in solid, liquid, and gas phases, generated in different microenvironments, and emitted by a variety of sources. The mixture includes a large number of VOCs and carbonyl compounds (aldehydes and ketones). (According to their volatilities, gas-phase carbonyl compounds belong to the VOC category; however, they are conventionally referred to as their own chemical class. Hence, we present carbonyl compounds, or carbonyls, as a separate group in this report.) Also included in the air toxics mixture are semivolatile compounds and elements that comprise fine airborne PM, of which one respirable fraction is $PM_{2.5}$.

Toxicologic and clinical studies performed under controlled exposure conditions, as well as epidemiologic studies in occupational settings and communities, have been used to investigate the toxicity and health effects of many components of this complex mixture. Exposure to high concentrations of several air toxics has been associated with neurologic, teratologic, carcinogenic, and cardiovascular effects (Liber et al 1989; Cohen et al 1992; Koren et al 1992; Leikauf 2000; Parent et al 1992; Dockery et al 1993; Lovett et al 1999; Morello-Frosch et al 2000; Pope 2000; Samet et al 2000; Boj et al 2003; Bolt HM 2003; Delfino et al 2003). However, little has been done to characterize possible health effects of exposures to air toxics at concentrations that approach environmental levels (Bascom et al 1996; Suh et al 2000). The general population, including more susceptible groups such as children, older persons, and individuals with compromised health, is exposed to environmental-level air toxics every day; therefore, it is important to develop appropriate control strategies and regulations for air toxics. The largest data gap in the risk assessment of air toxics, however, appears to be characterization of exposure for the general population.

Air toxics are emitted into the outdoor air from many different sources. During 1993 in the United States, 3.7 million tons of air toxics were emitted, with approximately 41% derived from mobile sources, 35% from area sources, and 24% from local stationary sources (US EPA 1993). Nearly 50 million people have been estimated to live in locations where outdoor concentrations of one or more air toxics may exceed levels of concern for cancer and noncancer health effects in humans. Air toxics emitted outdoors from local sources in urban settings present a potential for inhalation exposure. Some air toxics that are not generated locally can be transported through regional, national, or global air sheds, depending upon their atmospheric residence times, and contribute to inhalation exposure. Air toxics emitted from indoor sources and personal activities add to the potential exposure burden. A comparison of total air toxics emissions by state indicates that, as expected, heavily industrial and highly populated areas have the highest emissions (US EPA 2001; Reynolds et al 2003); these areas include the three urban locations examined in this study.

SPECIFIC AIMS

The overall objectives were to investigate the relations of indoor, outdoor, and personal air concentrations of VOCs, carbonyl compounds, and $PM_{2.5}$, and in-vehicle concentrations of carbonyl compounds; and to quantify the outdoor contribution to indoor and personal air concentrations of the measured pollutants. Homes located close to outdoor sources of target compounds were oversampled to examine these relations in situations with high contributions of outdoor sources to exposures. For each household, data were collected on VOCs, carbonyl compounds, $PM_{2.5}$, AERs, temperature, relative humidity, personal activities of participants, and home characteristics.

$PM_{2.5}$ samples were also analyzed for a suite of chemical species (these data are the subject of Part II of this Research Report [Turpin et al 2005]). Elizabeth, New Jersey; Houston, Texas; and Los Angeles County, California were selected for sampling. These three geographically distinct locations have different climates and housing characteristics. Measurements were made across seasons and in homes with a wide distribution of AERs.

Some pollutant transport models and exposure models have been evaluated using air pollutant concentrations and exposure measurements provided by this and other similar studies. Linking such models would be the most comprehensive approach to predicting population exposures to outdoor sources and to developing effective strategies for reducing personal and community exposures to air toxics (Georgopoulos et al 1997; Jurvelin et al 2001, 2003). Although this modeling effort was beyond the scope of the current project, one goal of our study design was to gather data that could be used for this purpose.

The specific aims of this study were to:

1. compare indoor, outdoor, and personal air concentrations of the pollutants measured (and in-vehicle concentrations for carbonyl compounds);
2. examine the effects of variables such as season, home type, and city on measured concentrations and the relation between indoor and outdoor concentrations;
3. quantify the contribution of outdoor sources to indoor concentrations of the measured pollutants; and
4. determine indoor source strengths of the measured pollutants that are primarily generated indoors.

This report does not address one original aim, which was to evaluate outdoor air toxic concentrations as a function of proximity of homes to specific sources of individual compounds or groups of compounds (such as proximity to a dry cleaning establishment for tetrachloroethylene, or to a gas station for a suite of aromatic VOCs and methyl *tert*-butyl ether [MTBE]). In order to perform a robust analysis of the effects of proximity to outdoor sources, we would need additional data (eg, source strength and meteorologic parameters) that have not yet been incorporated into this data set. This is a subject for future data gathering and analysis.

STUDY DESIGN

DESIGN FEATURES

Attempts were made to measure indoor, outdoor, and personal concentrations of 18 VOCs, 17 carbonyl compounds, and $PM_{2.5}$ mass concentrations. The target VOCs were either air toxics commonly found in urban settings (1,3-butadiene, methylene chloride, MTBE, chloroprene, chloroform, carbon tetrachloride, benzene, trichloroethylene, toluene, tetrachloroethylene, ethyl benzene, *m*- & *p*-xylenes (which coelute), *o*-xylene, styrene, and *p*-dichlorobenzene) or precursors of aldehydes and PM in outdoor or indoor air (*d*-limonene, α-pinene, and β-pinene) (Fan et al 2003). The carbonyl compounds selected included six that had been targeted by HEI's RFA 97-2 (formaldehyde, acetaldehyde, acrolein, crotonaldehyde, glyoxal, and methylglyoxal) and several others that are expected to be relatively abundant in the air (acetone, propionaldehyde, benzaldehyde, and hexaldehyde) (Zhang et al 1994a,b).

The major features of the study design follow.

1. Approximately 100 homes were selected from each of the three cities. Some were immediately adjacent to known outdoor sources of a target compound, and others were some distance from sources. The homes located particularly close to outdoor sources were oversampled to examine possibly high-level contributions of outdoor sources to exposures.
2. From May 1999 through February 2001, each home was measured twice (except for those that left the study), with a time interval of at least 3 months between measurements to examine possible seasonal effects. Because of budgetary constraints, PM measurements were conducted in a subset of the homes (about 50% in each city).
3. Each measurement session was a continuous 48-hour period. Indoor, outdoor, and personal measurements were made simultaneously.
4. Each home had at least one adult subject who provided personal measurements. Children were recruited as subjects as well to the maximum possible extent.
5. During the 48-hour measurement period, a subset of the adult subjects (68 total for all three cities) participated in the in-vehicle measurements of carbonyl compounds.

In summary, this study was not designed to obtain a population-based sample (the number of homes sampled, the participant selection criteria, and the recruiting procedures did not meet the criteria for population-based sampling), but rather to provide matched indoor, outdoor, and personal concentrations in homes with varying proximities to outdoor pollution sources and with a wide range of AERs. These data can be used in mechanistic examinations of relations among indoor, outdoor, and personal concentrations.

STUDY SITES

The three study sites are described in detail in Appendix A. Briefly, Elizabeth is a municipality of 110,000 in New Jersey. It has a high population density and forms an urban continuum with other cities in the region. Within Elizabeth and in adjacent communities, there are a multitude of outdoor air toxic sources including emissions from an industrial complex and an incinerator, numerous commercial sources (eg, gasoline stations, dry cleaners, refinishing shops, and small factories), and mobile sources from congested local streets and several major highways that pass through the community. Newark Liberty International Airport borders Elizabeth on its north side. Home selection included residences on the same block or within one to two blocks of some of these source types, with the exception of the airport. Homes farther from the sources were selected from the western section of Elizabeth, which has fewer commercial and industrial facilities and lower traffic density. Homes were selected throughout the year in all sections of Elizabeth, so no intentional imbalance in homes by source type should be present in the seasonal data for Elizabeth.

In Houston, petrochemical facilities were the major source type targeted. The Houston metropolitan area has the largest density of petrochemical complexes in the world. Different units within these facilities may process crude petroleum for fuel production, but they also produce chemicals including plastics and solvents. Thus emissions from these facilities come not only from the types of chemicals expected from fuel production, but also from the raw materials and processes involved in the production of chemicals and plastics. In addition, most of the large facilities are surrounded by highways and major access roads, so some contribution from mobile sources would also be expected in these areas. The approach used in Houston to target households for participation was to focus on those areas with large petrochemical complexes (Appendix A), and in each such area to target households near the source and further away from the source. To the extent it was possible, both locations of households were monitored within any given area during the same time frame.

In Los Angeles County, vehicular emissions from major freeways were the primary source type targeted. Sampling was conducted at four locations in Los Angeles County that are influenced by major freeways: West Los Angeles, Pico Rivera, Burbank, and Santa Clarita. Again, homes were selected according to their proximity to a major freeway (nearby or further away; Figure A.4). All sampling locations were within 4 km of an ambient air monitoring station operated by the South Coast Air Quality Management District.

DATA COLLECTION METHODS

SUBJECT RECRUITMENT

Before subject recruitment commenced, the field protocol and the consent form designed were approved by the Institutional Review Boards of the University of Medicine and Dentistry of New Jersey, Rutgers University, and the University of Texas. Human consent procedures met governmental guidelines. Informed consent was obtained from each participant and a parent or guardian for minors.

Once the targeted areas were identified, mailings were sent and follow-up telephone calls were made to specific homes to identify and recruit subjects. In Los Angeles, for example, mailings were sent to homes randomly selected for participation from within the four preselected geographic areas (Appendix A). Although this was adequate in LA, in Elizabeth and Houston the frequency of acceptance by those contacted in this manner was low. Therefore other methods to locate individuals in designated areas were developed, including door-to-door canvassing, seeking support from community and religious leaders in the targeted neighborhoods, giving interviews about the study to the local newspapers, radio, and television stations, making presentations at community centers, and using word-of-mouth contacts through local organizations. Some details about subject recruiting techniques and lessons learned in the pilot phase can be found in Appendix B.

When an individual in a selected residence was contacted, the first step was to determine if the following study criteria were met: the household residents did not smoke, there was an adult in the household who would be home for more than 10 hours per day and was willing to wear the personal sampler; and the possible subject was not planning on moving within the next 3 months so a repeat visit could be done. If these requirements were met, then an appointment was made for the study staff to visit the home. In any home where a child lived, if the adults gave permission, the child was asked if he or she wanted to participate by wearing a personal monitor (passive samplers only for children between 8 and 15 years old or active and passive samplers for children older than15). For many subjects, after an individual expressed willingness to participate, initial recruitment and retention through the second visit was time-consuming and required many telephone calls and visits.

SURVEY INSTRUMENT: QUESTIONNAIRES

The questionnaires used were adapted from National Human Exposure Assessment Survey (NHEXAS) questionnaires (Sexton et al 1995). By using or modifying the questions

relevant to the study objectives, we developed three questionnaires (Appendix H, available on request): the Baseline Questionnaire; the Technician Walk-Through Questionnaire; and the Activity Questionnaire. The Baseline Questionnaire includes sections on household and participant characteristics, demographics of the participant, family income, housing characteristics, facilities and usage, personal exposure activities before the study period, and a few questions on the respiratory health status of participants. The Technician Walk-Through Questionnaire includes an evaluation of the home and its usage, a floor plan, and a description of the neighborhood and possible local stationary sources of air toxics. The Activity Questionnaire includes a 48-hour activity log listing the time spent in each microenvironment and a detailed series of questions related to activities, duration of activities, and use of consumer products.

Each questionnaire was translated into Spanish, and a Spanish-speaking field staff member was available for each household where Spanish was the native language. All three questionnaires were evaluated in the pilot study, and their lengths were reduced to make administration of the questionnaires more feasible in the main study (Appendix B).

Analyses of the questionnaire data are not included in this report.

GENERAL APPROACH FOR FIELD DATA AND SAMPLE COLLECTION

The fieldwork included collecting (1) indoor, outdoor, and personal air samples of VOCs and carbonyl compounds in all of the homes (306 in all cities); (2) indoor, outdoor, and personal air samples of $PM_{2.5}$ in a subset of approximately 50% of the homes; and (3) in-vehicle air samples of carbonyl compounds in a subset of adult subjects (68 from all cities). The fieldwork also included administering the three questionnaires and measuring AER, temperature, and relative humidity in all the homes.

During the first home visit, the study was explained, informed written consent to collect the samples and questionnaire responses was obtained, the Baseline Questionnaire and Technician Walk-Through Questionnaire were completed, the tracer gas sources for the AER measurement were placed (see the section Measurement of AER), and an appointment was made to set up the samplers. Implementation of the fieldwork strictly followed a single set of detailed field protocols in all three cities. To conduct the fieldwork, four different forms were used: (1) an informed consent form to confirm that the participant had proper information on the study and had provided informed consent to participate; (2) a subject fee payment voucher; (3) a sampling information form to collect data on all types of samples; and (4) a form (originated during field preparation) to document chain of custody of samples.

The indoor samplers were placed in a rack assembled in the main living area of each home. The rack was between 1 and 2 m above the floor and at least 1 m from a wall. The samplers were positioned as far as possible from any indoor emission sources, such as portable heaters, fireplaces, and kitchen stoves (cooking can generate some of the targeted compounds). The outdoor samplers were placed in a secure location (sheltered from rain and direct sun), which was selected as representative of the air surrounding the residence. The preferred outdoor location was approximately 1 to 2 m above the ground and at least 1 m away from the outside wall or other objects. Access to an electrical power source was necessary. The use of a low roof or a patio outside windows was considered acceptable for outdoor sampler placement if security concerns or practical logistics required it.

A personal air sampling set was designed to hold and carry all the monitors (for $PM_{2.5}$, VOCs, and carbonyl compounds) such that the sampling inlets were as close as possible to the participant's breathing zone. The set included a bag containing a personal air pump, a bag containing the battery packs for the pump, and holders to keep the monitor inlets near the breathing zone. The passive samplers and active sampler inlets were attached to the participant's collar or to a shirt or jacket pocket (Figure 1).

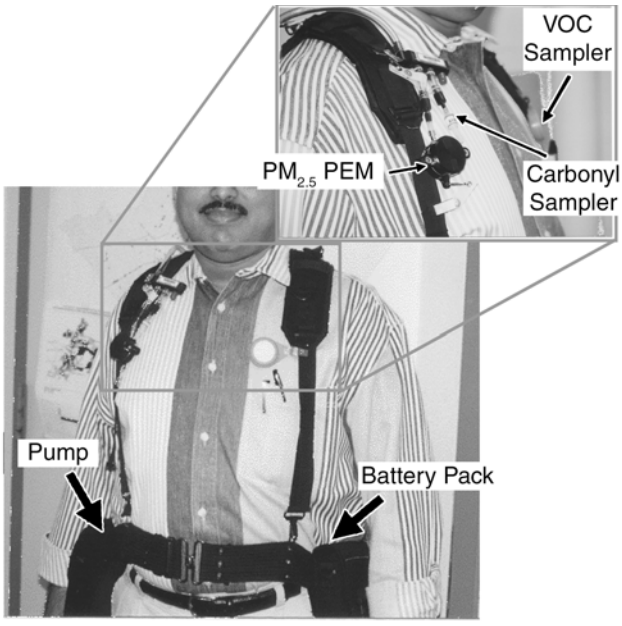

Figure 1. Personal air sampling set showing the harness equipped with a BGI personal sampling pump, battery pack, tubing, $PM_{2.5}$ PEM, VOC sampler (OVM 3500 badge), and carbonyl sampler (active method). (Children < 15 years old wore only passive VOC and carbonyl samplers clipped to a shirt; not shown.)

Labels with sequential bar code numbers were used to identify each sample and placed on the sample container and related sampling form. The sequential number (and manufacturer-provided identification label for the VOC badges) was the only information recorded for the VOC badges, carbonyl cartridges, or PM filter containers; the samples were analyzed in a blind fashion, that is, without the analyst knowing whether the sample was a blank or an indoor, outdoor, or personal air sample. Colored labels were used for different sample types to facilitate the proper labeling of the samples (eg, blue for VOCs, red for PM, green for carbonyl compounds, and black for equipment).

Specific guidelines were prepared and provided to the participants and the laboratory and field personnel to help ensure that all procedures were followed strictly. The guidelines included a checklist for the field staff, a spare parts list, a participant's guide for handling the personal air monitoring set, a field guide, a technician's guide for loading the filters and completing prefield setup, a technician's guide for unloading the filters, and a list of postfield procedures. In addition, appendices to the guidelines included sampling information forms for collecting data; chain-of-custody forms; and standard operating procedures giving detailed directions on coding, labeling, and tracking samples, weighing, shipping, mailing, and storing filters, cleaning the pump, and repacking and reconditioning the adsorbent trap.

MEASUREMENT OF AERs

AERs were measured using a technique developed by Dietz and coworkers (1986) for the determination of total exchange of indoor air with outdoor air in relatively small enclosures such as homes, apartments, or small offices. As the number of air changes per hour increases, the steady-state concentration of an indoor tracer gas decreases. AERs have previously been reported to be quantifiable over the range of air changes from 0.10 hr^{-1} to 2.5 hr^{-1}; and the limit of detection of air changes to be about 3.0 hr^{-1} (Dietz et al 1986). In this study we increased the source strength of the tracer gas in order to detect up to 5.0 hr^{-1}. The measurement of AER was accomplished using perfluorinated methylcyclohexane (PMCH) as the tracer gas, under steady-state conditions; the passive sampling device was a capillary absorption tube (CAT; 6.35-cm length × 0.6-cm OD × 0.4-cm ID), containing a small amount of a carbonized adsorbing material sandwiched in the middle by stainless steel screens. Samples were extracted and analyzed using a gas chromatograph (GC) and electron capture detector.

During the first visit, four PMCH sources were placed in different rooms of the residence to distribute the tracer compound throughout the entire structure. The sources were placed in locations where they were unlikely to be picked up or misplaced by a resident. Whenever possible, the sources were placed in an area that allowed the tracer to disperse evenly and be transported throughout the dwelling. Among the areas avoided were windows and doors where there were strong drafts or winds, stairways (which have increased vertical air movement due to thermal effects), walls or cubbyholes, sources of heat or cold, and appliances such as refrigerators and dehumidifiers that contain Freon, a possible interfering agent for the PMCH analysis. The PMCH sources were placed in the residence at least 48 hours before sampling with the CAT to ensure that the tracer would reach steady state inside the residence. The CAT was never placed closer than 2 m from a PMCH source. Once the location for the CAT was selected, sampling was activated by removing the cap on the numbered end of the glass tube and positioning it with the open end facing down or sideways, to minimize collection of settled particles. CATs were activated at the start and capped at the end of the air sampling. During the final visit to pick up all samples, the CATs were recapped and the field logs were completed.

Indoor and outdoor temperatures were recorded using a sensor containing a data logger. These sensors were set to read the temperature every 10 minutes during the sampling period. The volume (occupied space) of each home was measured using a tapeless ultrasonic tool or a walking tape. Unfinished basement or attic space that was not routinely used during the sampling was not included in the total volume.

The PMCH sources and CATs were never transported to the field together or on the same day. The sources were shipped in ziplock plastic bags in a box within a box. The CATs were kept in ziplock bags with activated carbon to protect them (in Houston, 4-oz polypropylene bottles with screw caps were used instead of ziplock bags). These bags and bottles were placed in a box within a box with additional papers impregnated with activated carbon for shipping. Field blanks of CATs were transported to the sampling location and treated the same as samples except that they were opened once and then resealed. Collocated CATs (about 15%) were used to establish precision.

The PMCH sources and CATs were supplied under a contract with Harvard University (Robert Weker's laboratory). The Harvard laboratory also checked emission rates for the sources and analyzed the CATs. In preparing the standard calibration curve, a series of CATs were infused with known amounts of PMCH using a PMCH generator. The amount of PMCH adsorbed on CATs was determined using a GC and electron capture detector system (Varian model 6000) (Dietz et al 1986). Carrier gas was 5% H_2 and

95% N_2. The temperature of the special external Porapak GC column was monitored by a thermocouple with the signal (0 to 2 mV) displayed on a stripchart recorder.

The AER was determined with the following equation:

$$AER = (n \times R_{Perm} \times R_{CAT} \times T_{CAT}) / (V_{PMCH} \times V_{Home}),$$

where n is the number of PMCH sources used, R_{Perm} is the source permeation rate (ng/min; determined with the same method as the source emission rate), R_{CAT} is the CAT collection rate (0.008308 L/hr), T_{CAT} is the CAT exposure time (minutes), V_{PMCH} is the volume (pL) of PMCH found on the CAT (calculated using the standard GC calibration curves), V_{Home} is the home volume (ft^3) and necessary conversion factors (60 min/hr, 28.3 L/ft^3, 109 ng/g, 1000 pL/nL, PMCH molecular weight = 350 g/mol or ng/nmol, PMCH molecular volume = 24.45 L/mol or nL/nmol). Details of the method detection limit (MDL), precision, and other quality control measures are presented in the section Quality Control Measures and Data Correction.

MEASUREMENT OF VOCs

Two types of 3M organic vapor monitor (OVM) badges were evaluated in the pilot study (Appendix B); on the basis of the results, the OVM 3500 badges were used to collect VOC samples. VOCs diffuse through a fixed windscreen at a known rate and then are adsorbed onto pads impregnated with activated carbon. Each badge was removed from its sealed container, a label was placed on the back, and the corresponding bar code label was placed on the sampling information sheet. The badges for the indoor and outdoor samples were clipped to the rack or platform that held the sampling equipment. The participant wore the personal sampler on clothing or on the sampling vest or sampler holder such that the windscreen (sampling side) faced outwards and was not covered by any clothing (see Figure 1). The need to leave the sampling surface exposed to the air was related to the participant verbally and written in the instructions provided to the participant. The personal sampler was worn whenever the participant was awake, except while showering or bathing. During these activities and while sleeping, the participant placed the sampler in a location that could practically represent the breathing zone (eg, bedside table while sleeping). After sampling was completed, the sampler was retrieved from the subject or sampling setup, capped with the plastic cover that was kept in the original container, and placed back into that container. The containers were transported to the laboratory in a cooler containing blue ice and stored in a refrigerator in the laboratory until analysis.

The charcoal pad of each badge sample was removed from the OVM 3500 badge using clean tweezers with Teflon-coated tips. The procedure for analyzing the target VOCs was based on a previously described method (Chung et al 1999a,b). Extraction was performed using a high-purity solvent mixture of acetone and carbon disulfide (2:1) containing a surrogate (4-bromofluorobenzene) added to the solvent mixture before extraction and internal standards (bromochloromethane, chlorobenzene, *d*-5-difluorobenzene, and 1,4-difluorobenzene) added to the badge extract. Calibration curves were prepared for each VOC from commercially purchased certified standards (Accustandards, New Haven CT). The standard solutions were specially prepared by the manufacturer using the same low-benzene carbon disulfide as was used in the extraction of the badges for all target VOCs. (1,3-Butadiene was prepared in methanol solution because it is largely insoluble in carbon disulfide.)

The standards were supplied in individual 1-mL glass ampules and diluted with high purity acetone and carbon disulfide solvent to achieve the desired concentrations for preparing calibration curves. The internal standards (a solution of bromochloromethane, chlorobenzene, *d*-5-difluorobenzene, and 1,4-difluorobenzene in methanol [Supelco, Bellefonte PA]) were diluted 1:10 for use as a working solution; this was added to all sample extracts. The purity of each lot of acetone and carbon disulfide was tested prior to its use to confirm that it did not contain more than trace amounts of any of the target compounds. Each acetone–carbon disulfide solvent mixture was freshly prepared before use in the extraction and analyzed for all of the target compounds using the same method as for analyzing the samples. If any of the target compounds were present at more than 0.05 ng/µL in the solvent mixture, then a new solvent mixture was prepared.

In an amber-glass vial, VOCs were extracted from the charcoal pads with 1 mL of the high-purity acetone–carbon disulfide solvent mixture by sonication for 45 minutes in an ultrasonic bath. Ice was added to the bath water to minimize any temperature increase and resulting evaporation of the solvent. After the ultrasound extraction was complete, a 200-µL aliquot of the extract was pipetted into another amber-glass vial with a 250-µL conical glass insert containing 10 µL of the internal standard solution. Each vial was immediately capped with a Teflon-lined cap and analyzed using an autosampler and sequencing software on a Hewlett Packard 6980/5973 GC and mass selective detector system. Laboratory blanks and controls were included in the sequence after every 10 sample vials. Duplicates and replicates were prepared for analysis as well.

Compounds were separated using a Restek 624 GC column (60-m length × 0.25-mm ID × 1.4-µm OD) and the following GC conditions. The injection port and transfer line temperatures were 180°C and 280°C, respectively. The

GC oven temperature program sequence consisted of an initial temperature of 40°C held for 12 minutes, then a ramped increase of 8°C/min to 100°C with a 6-minute hold, followed by a rate increase of 15°C/min, to a final temperature of 200°C, with a final holding time of 4 minutes. A 1-µL sample was injected by the autosampler with initial purge for 1.0 minute and a solvent peak delay of 4 minutes. The inlet was pressure programmed from 3 psi, held for 0.5 minute to 16.1 psi, corresponding to 1.0 mL/min column flow rate. Solvent delay was set to 4 minutes, and the detector was turned off from 5.5 minutes to 11.2 minutes to avoid the solvent peak. This step was necessary as 1,3-butadiene elutes at 4.7 minutes, ahead of the solvent peak. The detector was set to scan from 35 to 200 atomic mass units at a rate of 3.9 scans per second with a threshold level of 500 at 2100 electron multiplier voltage (emv). The mass selective detector was operated at 167°C with a rough pump vacuum of 0.040 mm Hg (40 mtorr). The analysis time per extract was 36 minutes. The data collected were analyzed and quantified using a calibration curve consisting of a minimum of five concentrations made freshly using the certified standard solutions of the target VOCs. The calibration curves before and after cleaning the mass spectrometer were compared and verified with an external standard check at two concentrations prior to running the samples. The calculated concentration of the external standards had to be within 20% of the expected value or a new calibration curve was prepared.

After the sample was analyzed with gas chromatography–mass spectrometry (GC-MS), each chromatograph was analyzed using Hewlett Packard Chemstation software and tabulated using an Excel file format to calculate the mass per milliliter of target VOCs in each extract solution. The OVM bar code numbers, rather than the OVM identification label supplied by the manufacturer, were used to identify the samples so that the analyst did not know the type of sample being analyzed until the laboratory quality control procedures were completed. After analytical results were final, they were linked to other spreadsheets containing sample information such as sample type, sampling time, temperature, average blank values, and compound diffusion rates (as reported by 3M Company 1993) to calculate temperature-corrected air concentrations. The field blank was subtracted from each sample before the sample concentration was calculated (see the section Quality Control Measures and Data Correction).

All the Elizabeth VOC samples were analyzed in the Environmental and Occupational Health Sciences Institute (EOHSI) laboratory in New Jersey, all the Houston VOC samples were analyzed in the University of Texas laboratory, and the Los Angeles samples were distributed (about evenly) between the two laboratories for analysis. An interlaboratory comparison was conducted in which the same extracts of VOC standards and samples were analyzed by both laboratories. The comparison results are presented in Appendix C. Agreement of better than 20% was found for most compounds.

MEASUREMENT OF CARBONYL COMPOUNDS

The measurement of carbonyl compounds was done using both active and passive sampling. When we submitted the proposal and started the study, there were no passive samplers for carbonyl compounds available that suited our needs. During the early phase, the conventional active sampling method based on 2,4-dinitrophenylhydrazine (DNPH) was used to collect indoor, outdoor, and in-vehicle samples and a personal sampling system was designed that could collect carbonyl compounds and $PM_{2.5}$ samples simultaneously (Appendix B). Meanwhile, Dr J Zhang and colleagues at EOHSI continued developing and evaluating the passive aldehydes and ketones sampler (PAKS).

The PAKS evaluation consisted of tests under different environmental conditions in the laboratory and collocation of PAKS with DNPH active samplers in some of the homes. The results indicated that for both stationary and personal sampling, the PAKS is a valid passive sampler for collecting carbonyl compounds over 24 to 48 hours (Zhang et al 2000). Field evaluation revealed several advantages of the PAKS method. Mainly, the PAKS worked substantially better for acrolein and crotonaldehyde; it eliminated the possibility of pump malfunctioning, which substantially increased the number of valid samples; and its use significantly reduced the burden on participants and the workload on the field personnel. Thus carbonyl compounds were measured using the PAKS-based passive method during the later stage of the study, after the funding agencies had approved. However, the active method was used throughout the study for in-vehicle measurements because those sampling periods usually lasted less than 3 hours, which was not enough time to collect a sufficient quantity for subsequent chemical analysis using the passive method. Details of PAKS development and evaluation are presented in Appendix D.

Active Sampling Method

The active sampling method for carbonyl compounds had been used at EOHSI previously, and details of the method can be found in earlier publications (Zhang et al 1994a,b, 2000). Briefly, batches of Sep-Pak C_{18} cartridges (Waters Corp, Milford MA) were coated with DNPH prepared in the laboratory and stored in a freezer before use. The DNPH-coated cartridges were used to collect carbonyl

samples at a desired sampling flow rate for different types of samples (Table 1). The flow rate for all sampling was less than 1 L/min to avoid potential breakthrough (Zhang et al 1994a). (The results of breakthrough tests showed insignificant breakthrough for all measured carbonyl species.) Indoor and outdoor active sampling of carbonyl compounds was achieved with a Buck SS pump (AP Buck, Orlando FL) that was placed in the instrument rack. For personal sampling, breathing-zone air was pulled through a DNPH-coated cartridge, with or without a personal $PM_{2.5}$ sampler in parallel (see the section Measurement of $PM_{2.5}$ Mass), using a BGI 400S pump system (BGI, Waltham MA) worn in the sampling bag on the participant's hip (Figure 1 and Appendix B).

The in-vehicle, intermittent carbonyl sampler consisted of a BGI pump inside a pressed-polystyrene–board box, bonded with methylene chloride. A nylon rope that passed through two holes drilled in the bottom of the box was used to hang the pump box from the passenger seat headrest (Figure 2). In-vehicle sampling took place only while participants were driving the car. To compensate for this shorter sampling period, which would result in a lesser volume of air sampled and, ultimately, in sample loading on the cartridges that would be insufficient to meet the limit of detection, the BGI pump was calibrated to the higher flow rate of 700 to 800 mL/min. Further, a check valve was placed on the inlet of the sampling cartridge to prevent passive sampling of the DNPH-coated cartridge when the BGI pump was turned off. Participants were questioned about their vehicle activities to complement the information on the in-vehicle sampling data sheet and verify that they had completed it correctly. The sheet, which subjects filled out before and after their driving activities, included pump on and pump off time increments, road type traveled, whether anyone was smoking in the vehicle during the time of sampling, whether the windows were open or closed, and what, if any, ventilation was used. This sheet was clipped to the sample box, with a pen for the subjects' convenience. Sampling instructions were also attached to the

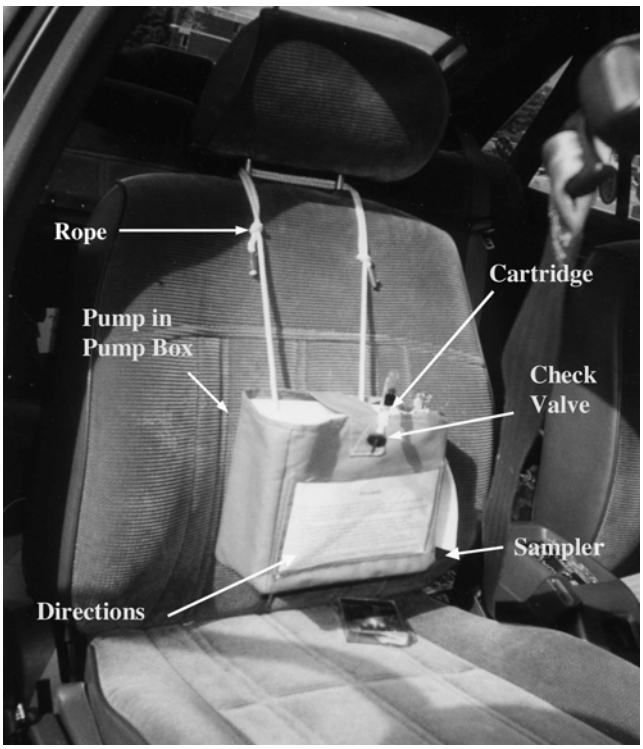

Figure 2. In-vehicle sampler positioned in the front passenger's seat.

Table 1. Sampling Methods for Carbonyl Collection

Sample Type	Flow (cm^3/min)	Pump Type	Collection Duration	Collection Medium
Active				
Indoor, outdoor	~200	Buck SS	~48 hr	DNPH coated C_{18} cartridge
Personal adult and child[a]	~50–80	BGI personal	~48 hr	DNPH coated C_{18} cartridge
In-vehicle	~700–800	BGI personal	Driving duration within 48-hr period[b]	DNPH coated C_{18} cartridge
Passive (PAKS)				
Indoor, outdoor, personal adult and child	NA[c]	NA	~48 hr	Modified C_{18} cartridge coated with DNSH

[a] Only children older than 15 years wore active samplers.

[b] The sampling pump was turned on only when the participant was driving his or her vehicle. A check-valve was placed on the end of the sampling cartridge to prevent contamination when the pump was not turned on. The range of sampling time was 55 to 459 minutes.

[c] NA indicates not applicable.

pump box to remind participants of the sampling procedures (see Figure 2).

When sampling was completed, the exposed cartridges (samples) were capped and shipped, along with field blanks (unopened cartridges), to the laboratory in a cooler with blue ice packs. The cartridges were extracted with acetonitrile (ACN) immediately after they arrived in the laboratory. The extracts were analyzed using a high-pressure liquid chromatography (HPLC) system with a reverse-phase Nova-Pak C_{18} column (3.9 × 150 mm; Waters Corp). The mobile-phase gradient program used was 100% of solvent A (water-ACN-tetrahydrofuran [THF] 60:30:10) held for 5 minutes; then programmed to 100% solvent B (ACN-water 60:40) in 28 minutes and held at 100% for 10 minutes; and then programmed back to 100% A in 5 minutes. The flow rate of the mobile phase was kept constant at 1 mL/min. The sample injection volume was 20 µL. The UV detector was set at 365 nm. Carbonyl compound concentrations were determined through calibration curves prepared using standard solutions of DNPH–carbonyl derivatives purchased commercially (Supelco, Accustandards). All sample concentrations were corrected for field blanks and carbonyl recovery (see the section Quality Control Measures and Data Correction).

All samples collected by the active method in Elizabeth and Los Angeles were analyzed in the EOHSI laboratory and the Houston samples were analyzed in the University of Texas laboratory. Some extracts of DNPH-carbonyl derivatives were analyzed in both laboratories. Results of the interlaboratory comparisons are presented in Appendix C. The results from the two laboratories agreed very well for most of the quantified carbonyl compounds. For two dicarbonyl compounds, glyoxal and methylglyoxal, a systematic difference occurred. Investigation into this issue revealed that the University of Texas laboratory had improperly converted concentrations of DNPH derivatives of these two dicarbonyl compounds to concentrations of the parent carbonyl compounds. Therefore, all the University of Texas laboratory glyoxal and methylglyoxal concentrations were corrected using the regression equations generated from the interlaboratory comparison results (Appendix C). No corrections were made for other carbonyl compounds.

Passive Sampling Method

The passive sampling method has been described in detail previously (Zhang et al 2000). Briefly, the PAKS, a tube-type diffusive sampler, was prepared by coating a custom-made C_{18} cartridge (Supelco) with an ACN solution of dansylhydrazine (DNSH) (Aldrich Chemical Co, Milwaukee WI). A batch of DNSH-coated cartridges were dried in a vacuum desiccator for 48 hours then individually wrapped and stored in a freezer. The shipping procedure was the same for the PAKS cartridges as for the DNPH cartridges.

For personal sampling, the cap of a PAKS was removed and the PAKS was clipped to the collar or shirt pocket of a subject, along with the OVM 3500 badge for passive VOC sampling. The participant ID, cartridge ID, start date and time, and end date and time were recorded on a sampling sheet accompanying each sampler. At the end of the sampling period, the PAKS was removed from the subject and securely capped. When a PAKS was used to collect carbonyl compounds in indoor air and outdoor air, it was simply placed in the selected sampling location with the cap open.

The exposed PAKS and unexposed field blanks were extracted with 2 mL ACN. The extracts were analyzed at EOHSI using an HPLC system consisting of a Waters 600E System Controller and a Waters 712WISP Autosampler 4100 Programmable Fluorescence Detector. The analytical column used was a Nova-Pak C_{18}. The mobile-phase program used was a linear gradient of 100% solution A (32% ACN; 68% water containing 1.6 g/L of KH_2PO_4) to 100% solution B (70% ACN; 30% water containing 1.6 g/L of KH_2PO_4) in 20 minutes; then from 100% B back to 100% A in 10 minutes; and then held at 100% A for 10 minutes. The mobile-phase flow rate was 1.0 mL/min. The injection volume was 20 µL. The fluorescence detector was set at an excitation wavelength of 240 nm and an emission wavelength of 470 nm. DNSH-carbonyl derivative standards were prepared in situ by spiking a known amount of carbonyl compounds into the DNSH-coated C_{18} cartridges. The spiked cartridges, treated and extracted in the exact same manner as the samples, served as external standards for identification and quantification of the carbonyl compounds.

The sampling rate of PAKS for each measured carbonyl compound was determined from a series of chamber experiments or from theoretical calculation based on the Fick Law. We found that the experimentally determined values and the calculated values agreed well (Zhang et al 2000). The sampling rates, as a function of temperature, used for the calculation of carbonyl concentrations in air are shown in Table 2. All concentrations were corrected for field blank levels and carbonyl recoveries in the same way as for the DNPH-based active method (see the section Quality Control Measures and Data Correction).

MEASUREMENT OF $PM_{2.5}$ MASS

Indoor and outdoor samples of $PM_{2.5}$ for analysis of gravimetric mass concentration were collected at 10 L/min on 37-mm stretched Teflon filters (2-µm pore; R2PJ037; Pallflex Gelman Scientific, Ann Arbor MI) mounted in Harvard impactors downstream of the single-jet impactor with a 2.5-µm cutpoint.

Table 2. PAKS Sampling Rate[a] at 25°C (298K)

Carbonyl[b]	Sampling Rate (mL/min)
Formaldehyde	7.43
Acetaldehyde	5.07
Acetone	4.76
Acrolein	3.92
Propionaldehyde	4.97
Crotonaldehyde	3.33
Benzaldehyde	3.21
Hexaldehyde	3.25
Glyoxal	5.15
Methylglyoxal	4.38

[a] Sampling rate (SR) at any temperature t (°C) can be obtained from the following equation:

$$(SR)_t = (SR)_{298} \times \left(\frac{298}{273+1}\right)^{1.5}$$

[b] Sampling rates of glyoxal and methylglyoxal were determined by spiking a known amount of each carbonyl onto a cartridge. Sampling rates of the other carbonyls were determined by exposing the cartridges to known concentrations of the carbonyls for a defined duration in a test chamber.

Personal samples were collected on 25-mm stretched Teflon filters (3-µm pore; R2PI025; Pallflex Gelman Scientific) with modified MSP personal environmental monitors (PEMs; MSP Co, Minneapolis MN) for 48 hours. The PEM has a 10-jet impactor upstream of the filter that is designed to provide a 2.5-µm cutpoint when a 0.4-L/min flow is maintained through each jet. Two jets were blocked to achieve a 2.5-µm cutpoint at 3.2 L/min. PEMs were also modified to hold a 25-mm filter, rather than a 37-mm filter, to reduce detection limits for mass and other species. Air was pulled through the PEM, and in some cases through an active carbonyl sampler parallel to it, using a BGI pump (see Figure 1).

Filters were loaded and unloaded in the samplers in the laboratory and the samplers were checked for leaks. They were transported to the field with a field blank that was placed in the sampling rack and then returned to the laboratory with the samples. Flow rates were measured at the beginning and end of each sampling period, and samplers were checked for leaks at the end of the sampling period if the flow rate had changed by more than 5%. All collected samples and field blanks were returned to the laboratory in coolers with blue ice packs and stored frozen until analysis.

Duplicate indoor and outdoor samples (35 pairs) were collected with pairs of Harvard impactors placed simultaneously on the indoor and outdoor sampling racks. In addition, 14 samples were collected with PEMs that were placed simultaneously on the indoor sampling rack with the Harvard impactor.

All the filters (samples and field blanks) were weighed on a microbalance (Cahn C-30, Cahn Instruments, Cerritos CA; or Mettler MT5, Mettler Toledo, Columbus OH) in an EPA-audited laboratory at EOHSI according to the EPA protocols. Each filter was equilibrated before and after sampling for at least 24 hours at approximately 40% relative humidity and 20° to 23°C, and weighed twice under those conditions. For postcollection analysis, conditions were 30% to 40% relative humidity and 20° to 23°C, within 5% relative humidity and 2°C of the conditions for precollection analysis. The precollection and postcollection analyses were conducted by the same operator on the same balance, with few exceptions. The balance was calibrated daily before filters were weighed with a 200 ± 0.025-mg primary mass standard traceable to US National Institute of Standards and Technology (NIST) mass standards, and an independent standard (50 mg) was analyzed after every 10 filters. At least one laboratory blank was weighed daily.

QUALITY CONTROL AND DATA MANAGEMENT

Quality Control Measures and Data Correction

A number of quality control measures were in place to assess measurement and analytical precision, accuracy, and detection limits. (1) Field blanks were prepared identically to sample substrates, transported to the field, stored on the indoor sampling rack, returned, and analyzed with samples. (2) A positive control was employed for VOCs and carbonyl compounds. (3) A known quantity of target species was spiked on prepared substrates, which were transported to the field, stored on the indoor sampling rack, returned, and analyzed with samples. (4) Approximately 10% of samples were analyzed in replicate, and independent standards were analyzed in addition to calibration standards. (5) Duplicate samples were acquired from collocated samplers. (6) Side-by-side measurements were made at study residences with collocated passive and active samplers for carbonyl compound and collocated PEMs and Harvard impactors for $PM_{2.5}$ monitoring. Quality control measures are summarized in Tables 3 through 8.

Field blank distributions (Table 3) were used to determine MDLs and blank corrections. If there was a batch-to-batch variation in blank concentrations, then we used batch means; otherwise, we used the overall means for blank subtraction. When measurable blank levels were present for a compound, the MDLs were expressed as 3 × SD of the field blanks. For compounds that were not detected in the field blanks, the MDL was calculated as 3 × SD of seven replicate injections of a low-level standard. Analytical precision (as a measure of instrumental reproducibility) was expressed as a pooled coefficient of variation (CV) of replicate sample analyses (Table 4). Measurement precision (as a measure of

Table 3. MDLs[a]

	Active Sampling Method[b]			Passive Sampling Method[c]	
	Indoor & Outdoor	Personal	In-Vehicle	EOHSI	University of Texas
VOCs					
1,3-Butadiene				3.1	4.0
Methylene chloride				2.1	0.29
MTBE				0.68	0.39
Chloroprene				0.51	0.51
Chloroform				0.42	0.28
Carbon tetrachloride				0.27	0.34
Benzene				1.1	0.54
Trichloroethylene				0.44	0.24
Toluene				6.7	7.1
Tetrachloroethylene				0.42	0.23
Ethyl benzene				0.74	0.23
m- & *p*-Xylenes				1.4	0.65
o-Xylene				0.85	0.29
Styrene				0.84	0.34
α-Pinene				1.27	0.21
β-Pinene				2.04	0.28
d-Limonene				1.01	0.43
p-Dichlorobenzene				0.91	0.75
Carbonyls					
Formaldehyde	0.96	1.75	4.65	0.28 or 0.10[d]	
Acetaldehyde	0.75	1.37	3.63	0.74	
Acetone	2.75	5.04	13.38	0.40	
Acrolein	0.57	1.04	2.76	0.14	
Propionaldehyde	0.52	0.95	2.53	0.05	
Crotonaldehyde	0.51	0.93	2.48	0.13	
Benzaldehyde	1.03	1.88	4.99	0.24	
Hexaldehyde	0.59	1.09	2.88	0.20	
Glyoxal	0.90	1.65	4.39	0.06	
Methylglyoxal	0.53	0.96	2.56	0.09	
$PM_{2.5}$	0.47	1.4	NA		

[a] Detection limits (in μg/m^3) were estimated as 3 × the SD of the field blank. (All samplers for field blanks were placed indoors.) When the species was absent in the field blank, a low-concentration calibration standard was used to determine detection limits. NA indicates not applicable.

[b] For carbonyl compounds, air concentration detection limit (μg/m^3) for active (DNPH) method was estimated using the average sample volume collected in the field for each sample type. (Average sampling volumes [m^3]: Indoor and outdoor samples, 0.417; personal samples, 0.228; in-vehicle samples, 0.086.) The results shown here are from the EOHSI laboratory; the results from the University of Texas laboratory were similar.

[c] The air concentration detection limit (μg/m^3) for the passive (DNSH) method was estimated using a nominal 48-hour sampling period.

[d] The detection limit was 0.28 μg/m^3 for the first batch of cartridges prepared with unpurified DNSH (10/13/99–5/1/2000); it was 0.10 for the second batch of cartridges prepared with purified DNSH (5/1/2000–2/7/2001). The remaining values in this column are for all samples (10/13/1999–2/7/2001).

Table 4. Species Analytical Precision (Instrumental Reproducibility)[a]

Measure	Active % (n)[a]	Passive % (n)[a]
AER		6–7
VOCs		
1,3-Butadiene		60 (44)
Methylene chloride		24 (44)
MTBE		8.6 (44)
Chloroprene		16 (44)
Chloroform		16 (44)
Carbon tetrachloride		9.7 (44)
Benzene		3.7 (44)
Trichloroethylene		9.8 (44)
Toluene		4.1 (44)
Ethyl benzene		3.2 (44)
m- & p-Xylenes		7.1 (44)
o-Xylene		5.8 (44)
Styrene		11 (44)
p-Dichlorobenzene		15 (44)
Carbonyls		
Formaldehyde	7.2 (10)	6.4 (20)
Acetaldehyde	14.7 (10)	5.8 (20)
Acetone	15.0 (10)	4.7 (20)
Acrolein	10.3 (10)	5.0 (20)
Propionaldehyde	14.2 (10)	3.4 (20)
Crotonaldehyde	14.7 (10)	6.3 (20)
Benzaldehyde	21.9 (10)	7.5 (20)
Hexaldehyde	29.0 (10)	7.4 (20)
Glyoxal	24.1 (10)	8.4 (20)
Methylglyoxal	17.4 (10)	11.1 (20)
$PM_{2.5}$	< 1 (60)	NA

[a] Values are expressed as the pooled CV of replicate laboratory analyses (%). Total number of replicate analyses is given in parentheses. NA indicates not applicable.

method reproducibility) was expressed as a pooled CV of collocated (duplicate) sample concentrations (Table 5). The pooled CV is given by the pooled SD (σ_{Pooled}) divided by the mean value of the pairs. For the general case

$$\sigma_{Pooled} = [\ \Sigma(n_i - 1)\ \sigma_i^2 / \Sigma(n_i - 1)\ ]^{1/2}\ , \quad (1)$$

and for paired data

$$\sigma_{Pooled} = [\ \Sigma d_i^2 / 2n]^{1/2}\ , \quad (2)$$

where σ_i is the SD of replicate set i, d_i is the difference between paired i values, n_i is the number of data points used to calculate σ_i, and n is the number of pairs.

Analytical accuracy for carbonyls (as a measure of instrumental accuracy) was calculated from the analysis of independent standards, expressed as the percentage of difference between measured and spiked species concentration or mass (Table 6). Field positive-control results (Table 7) were calculated from the analysis of samplers that had been spiked with a known quantity of target species and placed with field samplers for the designated sampling duration (48 hours nominal; VOCs and carbonyl compounds only). The results are expressed as the percentage of difference between measured and spiked species mass. Extraction efficiency for VOCs and two carbonyls (glyoxal and methylglyoxal) was calculated as the ratio of the measured quantity to the quantity spiked on the sampler and reflects only the recovery for the extraction process (Table 8). For the remainder of the carbonyls, recovery is expressed as the ratio of the measured concentration to the concentration generated in a gas test chamber and reflects recovery for both the collection and extraction processes.

AERs The ability to measure AERs is limited by the amount of PMCH on the CATs. The lower the collected PMCH, the higher the AER. Therefore, the detectable limit for PMCH determines the maximum AER measurable for a given residence and sampling protocol. The distributions of the blanks for the AERs for the three cities were tested by analysis of variance (ANOVA) and no differences were found in the mean values of field blanks among the cities. Accordingly, field blank values were pooled, and the mean blank value of 0.54 pL was subtracted from all sample CATs. The overall MDL of PMCH, expressed as 3 × SD of the field blanks (n = 158), was 4.67 pL. This enabled measurement of AER values up to approximately 5 hr^{-1}. Analytical precision was 6% to 7%, as a CV of replicate analyses of standards similar in volume to the samples (20 to 40 pL) and run during analysis of samples. Recoveries of 90% to 100% reported by the Harvard laboratory were based on analysis of known atmospheres generated using sources with known PMCH permeation rates and controlled air flow across the sources, and verified with standard injections into the GC flow path. The measurement precision was 16% expressed as a pooled CV of collocated samples (see Table 5).

VOCs Several VOCs had no measurable blank contributions. The MDLs for most VOCs were approximately 0.4 to 1 μg/m^3 for samples measured at EOHSI and 0.2 to 0.5 μg/m^3 for samples measured at University of Texas (for nominal 48-hour samples) (see Table 3 and Appendix C). The compounds with higher MDLs were 1,3-butadiene (which eluted close to the solvent peak and resulted in an increased signal-to-noise ratio in the chromatographic trace) and methylene chloride, toluene, β-pinene, and the xylenes (which had measurable blank levels owing to contributions from either the

Table 5. Species Measurement Precision (Method Reproducibility)[a]

Measure	Active % (*n*)	Passive[b] % (*n*)	Indoor Passive % (*n*)	Outdoor Passive % (*n*)	Personal Passive % (*n*)
AER		16			
VOCs					
Methylene chloride		ND	ND	ND	ND
MTBE		22 (151)	27 (54)	16 (62)	24 (36)
Chloroform		23 (103)	23 (44)	ND (29)	13 (31)
Carbon tetrachloride		16 (145)	25 (51)	17 (62)	11 (33)
Benzene		17 (156)	19 (56)	16 (64)	20 (37)
Trichloroethylene		ND	ND	ND	ND
Toluene		40 (145)	37 (55)	ND	43 (37)
Tetrachloroethylene		25 (141)	41 (50)	ND	20 (37)
Ethyl benzene		19 (148)	27 (55)	27 (58)	17 (36)
m- & *p*-Xylenes		18 (154)	29 (55)	23 (63)	19 (37)
o-Xylene		21 (153)	27 (55)	16 (62)	17 (37)
Styrene		38 (102)	34 (41)	ND	40 (35)
α-Pinene		37 (138)	51 (55)	69 (49)	20 (35)
β-Pinene		42 (87)	40 (47)	ND	29 (33)
d-Limonene		23 (120)	23 (53)	ND	14 (32)
p-Dichlorobenzene		8 (116)	4 (46)	ND	63 (29)
Carbonyls					
Formaldehyde	8.0 (11)	19 (108)	13 (41)	21 (41)	22 (26)
Acetaldehyde	14 (11)	30 (108)	33 (41)	23 (41)	16 (26)
Acetone	18 (10)	22 (105)	17 (40)	32 (39)	20 (26)
Acrolein	14 (5)	29 (86)	29 (31)	31 (33)	27 (22)
Propionaldehyde	19 (11)	27 (108)	28 (41)	27 (41)	24 (26)
Crotonaldehyde	15 (7)	26 (92)	22 (37)	35 (32)	22 (23)
Benzaldehyde	10 (11)	20 (108)	21 (41)	19 (41)	17 (26)
Hexaldehyde	10 (10)	19 (97)	14 (37)	24 (38)	21 (22)
Glyoxal	14 (10)	21 (108)	23 (41)	18 (41)	17 (26)
Methylglyoxal	10 (10)	19 (104)	17 (39)	21 (41)	17 (24)
$PM_{2.5}$[c]	17 (35)	NA			

[a] Values are the pooled CVs of pairs of collocated field measurements expressed as percentages. The total number of pairs is given in parentheses. ND indicates that CVs were not determined because more than half the values were below detection. NA indicates not applicable.

[b] These CVs are based on the total of all indoor, outdoor, and personal samples.

[c] Evaluated for Harvard impactors only, not for PEMs.

solvent or the OVM charcoal pad background). In particular, toluene had the highest blank contributions and a detection limit near 7 $\mu g/m^3$, which is higher than values reported in studies that used active sampling methods. However, this method sensitivity was sufficient to detect toluene in most samples because it had the highest air concentration of all of the VOCs measured.

Comparison of field blanks with laboratory blanks showed few, if any, differences for the majority of the VOCs (data not shown), indicating that little contamination occurred during handling and shipment. The species extraction efficiencies determined by both laboratories (see Table 8) were similar to those that the manufacturer reported at higher concentrations and generally exceeded 90%. Styrene, 1,3-butadiene, and chloroprene had extraction efficiencies closer to 70%, which might reflect chemical reactions and therefore losses, of those compounds on the OVM badge charcoal pads. Subsequent studies by Drs Morandi and Stock (personal communication from Dr Maria T Morandi 1997) showed that 1,3-butadiene and chloroprene were not stable on the OVM badges after sampling at environmental concentrations and that the losses from the badges increased with time. The concentrations

Table 6. Carbonyl Analytical (Instrumental) Accuracy[a]

Carbonyl	Indoors and Outdoors Active (*n*)	Passive (*n*)
Formaldehyde	8.5 (180)	5.2 (35)
Acetaldehyde	10 (180)	4.1 (35)
Acetone	10 (180)	14 (35)
Acrolein	8.7 (180)	16 (35)
Propionaldehyde	9.9 (180)	7.6 (35)
Crotonaldehyde	13 (180)	25 (35)
Benzaldehyde	11 (180)	10 (35)
Hexaldehyde	13 (167)	8.2 (35)
Glyoxal	19 (175)	14 (35)
Methylglyoxal	17 (166)	17 (35)

[a] Values are expressed as the average percentage of difference between measured and spiked samples calculated with the formula: average of (100 × |spiked − measured| / spiked). The values for the average of spiked samples were taken from analyses of standards (spiked samples) independent of analyses of calibration standards.

of 1,3-butadiene and chloroprene were not reliable and therefore are not reported here.

The analytical precision was less than 20% for all compounds (see Table 4) except for 1,3-butadiene (with stability problems on the OVM badge) and methylene chloride (with inconsistent blank values).

The overall measurement precision of the samples (see Table 5) was similar for the three types of samples collected—outdoor, indoor, and personal. The overall precision derived from pooled samples of all types ranged from 8% to 42%, as determined by analysis of the collocated samples. Some individual compounds had poorer precision for some sample types. There are two possible mathematical reasons for the lower precision. Even though the absolute error was small, a high percentage of deviation could occur for compounds if many of the samples were close to the MDL, as was the case for several of the chlorinated compounds. A high CV was also found for compounds that had one or two concentrations that were orders of magnitude above the majority of the sample concentrations, because 20% variation at high concentration could be magnified when the square of those differences was divided by the overall mean of the sample concentrations. This was the case for α-pinene and β-pinene, *d*-limonene, and *p*-dichlorobenzene. Measurement precision was lower for toluene and methylene chloride than for the other compounds, probably because background contributions from the OVM badge charcoal pad were higher and more variable than for the other compounds.

Table 7. Field Positive-Control Samples[a]

	Active (%)	Passive (%)
VOCs		OVM Badge (*n* = 12)
Methylene chloride		40
MTBE		25
Chloroform		44
Carbon tetrachloride		37
Benzene		33
Trichloroethylene		37
Toluene		32
Tetrachloroethylene		30
Ethyl benzene		29
m- & *p*-Xylenes		36
o-Xylene		29
Styrene		45
α-Pinene		23
β-Pinene		24
d-Limonene		29
p-Dichlorobenzene		27
Carbonyls	DNPH (*n* = 5)	DNSH (*n* = 17)
Formaldehyde	10	12
Acetaldehyde	21	34
Acetone	7.4	32
Acrolein	100	43
Propionaldehyde	2.6	11
Crotonaldehyde	77	29
Benzaldehyde	—[b]	15
Hexaldehyde	—	14
Glyoxal	61	13
Methylglyoxal	36	24

[a] Values are expressed as the average percentage of difference between measured and spiked samples calculated with the formula: average of (100 × |spiked − measured| / spiked). Values reflect an average of the blanks from all three cities.

[b] Dash indicates no data.

The results from the VOC field positive-control samples show the average differences between the spiked amount and the measured amount; differences ranged from 23% for α-pinene to 45% for styrene (Table 7). Extraction efficiencies, shown in Table 8, can be converted to the same expressions as for Table 7; for these extraction experiments, the average differences ranged from 0% (extraction efficiency = 100%) to 40% (for styrene tested in the University of Texas laboratory). The differences for the field positive-control samples were generally larger than the differences for the extraction efficiency experiments. This suggests that VOC losses might have occurred during the transport and field storage processes; the field positive-control samples

Table 8. Species Extraction Efficiency and Recovery[a]

Species	Active % (n = 6)	Passive EOHSI % (n = 6)	Passive University of Texas % (n = 15)
VOCs[a]			
Methylene chloride		120	90
MTBE		83	99
Chloroform		95	100
Carbon tetrachloride		130	96
Benzene		71	95
Trichloroethylene		87	97
Toluene		110	98
Tetrachloroethylene		98	91
Ethyl benzene		90	97
m- & p-Xylenes		87	82
o-Xylene		83	84
Styrene		71	60
α-Pinene		—	100
β-Pinene		—	100
d-Limonene		—	100
p-Dichlorobenzene		110	75
Carbonyls[b]			
Formaldehyde	81	101	
Acetaldehyde	96	87	
Acetone	109	80	
Acrolein	20	60	
Propionaldehyde	85	108	
Crotonaldehyde	39	76	
Benzaldehyde	95	98	
Hexaldehyde	86	94	
Glyoxal[a]	87	90	
Methylglyoxal[a]	93	95	

[a] Extraction efficiency is reported as the percentage of measured amount versus spiked amount.

[b] Recovery is reported as the percentage of the measured concentration versus the gas concentration in the chamber.

would have included any effects of transport and storage on individual badges sent to the field throughout the study; whereas the extraction efficiency experiments were conducted in batches and were prepared and analyzed in the laboratory within a short time period, generally a single day.

Carbonyl Compounds The distribution of all the field blanks indicated there were no batch-to-batch differences in DNPH cartridge (active sampler) field blank concentrations. Therefore, the overall means from all the blank cartridges were used to determine the MDLs and to correct the blank levels. However, the blank formaldehyde concentrations of DNSH cartridges were lower later in the study (when the passive PAKS were used) than during the first third of the study (when only active samplers were used). Other carbonyl compounds either were not detected in the field blanks or had no batch-to-batch differences. The decrease in formaldehyde blank values on the later batches of DNSH cartridges was due to the change in the cartridge-coating procedure. Initially, high-purity DNSH reagent (> 97% purity) was used directly to prepare the coating solution without further purification in our laboratory. To further reduce the impurity levels, we recrystallized the DNSH reagent in HPLC-grade ethanol and ACN. Within the cartridges prepared by the same DNSH-coating procedure, no significant batch-to-batch differences in field blanks were found. Therefore we had two overall PAKS blank concentration means (and thus two estimates of the MDLs) for formaldehyde, one for all the cartridges prepared with unpurified DNSH and the other for all the cartridges prepared with purified DNSH.

For the active method the analytical precision (Table 4) was generally comparable to the measurement precision (Table 5), indicating that the deployment of DNPH cartridges to the field (including sampler handling, transport, and storage) and recovery of the cartridges did not result in significant additional error. In contrast, the passive method had lower measurement precision (higher CV) than analytical precision, indicating that the field deployment process increased the variability of the DNSH cartridge blank levels, possibly through contamination. These findings are reasonable not only because the configuration of the PAKS allowed more molecular diffusion to occur (even when it was sealed during field deployment) compared with DNPH cartridges, but also because the fluorescence-based analysis of DNSH-carbonyl derivatives was more sensitive and thus able to capture a smaller variability than the UV-based analysis of DNPH-carbonyl derivatives.

Analytical (instrumental) accuracies (measured as the average percentage of difference between the measured amount and the spiked amount) for the active DNPH-based method ranged from 8.5% for formaldehyde to 19% for glyoxal. For the passive DNSH-based method, these ranged from 4.1% for acetaldehyde to 25% for crotonaldehyde (see Table 6). Neither method performed better than the other in terms of analytical accuracy.

Positive field controls (spiked cartridges) were used to evaluate the potential loss of species collected on the sampling substrate. For acrolein and crotonaldehyde, both the positive control results (Table 7) and the recovery results (Table 8) indicated a larger difference between the measured and spiked amounts than for the other carbonyl compounds

in general. This is consistent with the stability test results (Appendix D) showing that acrolein and crotonaldehyde disappeared rapidly from DNPH cartridges. Because all the samples were collected during a 48-hour (nominal) period and several hours passed before the sampled cartridges were extracted in the laboratory, acrolein and crotonaldehyde might have been lost partially or completely during sample transportation and even during sample collection. Hence we decided to exclude the concentrations of acrolein and crotonaldehyde measured using the active method from data analysis. We also decided not to report the hexaldehyde concentrations measured using the active method because of problems in accurately quantifying hexaldehyde in a fraction of these samples. Other carbonyl compounds analyzed using the active method were *o*-tolualdehyde, *m- & p*-tolualdehydes (which coeluted), and dimethylbenzaldehyde. They were detected in less than 10% of samples, however, so no statistical analyses were performed for these carbonyl compounds (in addition, these compounds were not specific research targets of the funding agencies).

$PM_{2.5}$ Mass MDLs for $PM_{2.5}$ mass concentrations, determined from a total of 452 field blanks, were 13 µg (1.4 µg/m^3) for personal mass concentrations and 15 µg (0.47 µg/m^3) for indoor and outdoor mass concentrations (see Table 3). Field blank weights before and after transport to the field were not significantly different according to a paired *t* test with 95% confidence intervals (CIs). Therefore no blank subtraction was performed for $PM_{2.5}$ mass measurements. All $PM_{2.5}$ mass concentrations were above MDLs. Any uncertainties in $PM_{2.5}$ mass measurements were introduced by sample handling, transport, storage, and sampling methods, rather than by analytical uncertainties; this is evidenced by extremely small estimates of analytical precision, that is, less than 1% (see Table 4). Analysis of 35 pairs of collocated Harvard impactors indicated that for indoor and outdoor $PM_{2.5}$ concentrations, measurement precision was 17% (Figure 3).

Sampling considerations limited the accuracy with which $PM_{2.5}$ was measured. It is well known that the collection of fine PM on a sampling substrate, changes in relative humidity, and changes in temperature alter the equilibrium partitioning of semivolatile PM species such as water vapor, ammonium nitrate, and semivolatile organic compounds. Some effort has been made to standardize analytic conditions by using an EPA filter-weighing protocol.

Figure 4 shows highly correlated $PM_{2.5}$ mass concentrations (r^2 = 0.98) measured with PEM and Harvard impactors placed together in the indoor sampling racks of 14 study homes. According to a *t* test on the log-transformed data, however, mass concentrations measured with the PEMs were significantly greater at the 95% confidence level than those measured with the Harvard impactors. During collocated sampling, the mean and median concentrations measured were 16.5 and 11.6 µg/m^3, respectively, for the Harvard impactor, and 19.5 and 13.5 µg/m^3, respectively, for the PEM. At the median personal $PM_{2.5}$ concentration of 37.6 µg/m^3, regressions with and without outliers suggested that the difference between the samplers was 1% with outliers included and 16% without them.

This level of accuracy is reasonable considering PM measurement precision. Intersampler differences of this size are not unusual in the measurement of $PM_{2.5}$. The intersampler differences could be due to differences in the shapes of the collection efficiency curves for the 2.5-µm impactor precut, differences in bounce from the impactor plates, or differences in volatile losses. The Harvard impactor had a single-jet impactor inlet and a face velocity of 16 cm/sec, whereas the modified PEM was operated with an 8-jet impactor inlet and a face velocity of 11 cm/sec.

Because the Harvard impactor has had a longer history in the field, had lower uncertainties owing to the higher flow rate, and has been compared with the $PM_{2.5}$ Federal

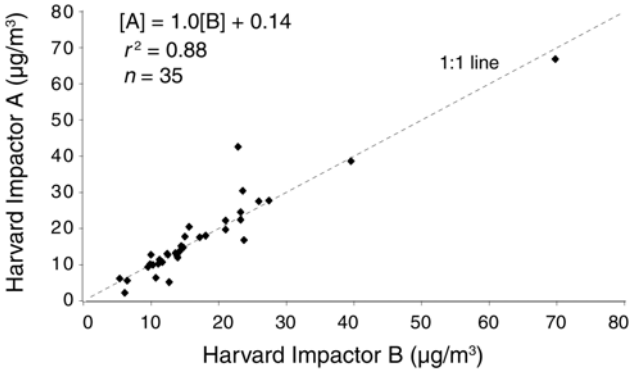

Figure 3. Indoor and outdoor $PM_{2.5}$ concentrations from collocated Harvard samplers.

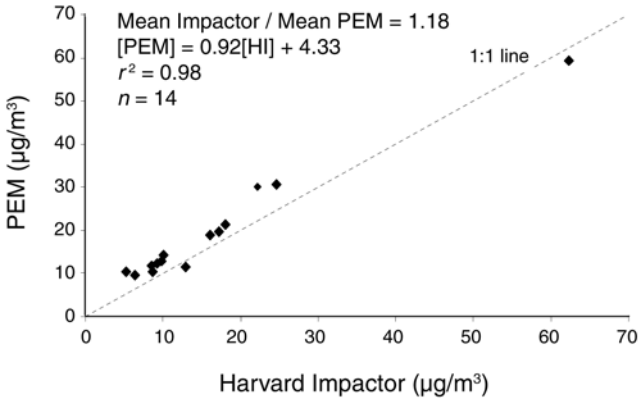

Figure 4. $PM_{2.5}$ mass measurements from collocated Harvard impactor and PEM.

Reference Method sampler (Allen et al 1997), it might be appropriate to calibrate the PEM to agree with the Harvard impactor. In this study, however, the PEM sampler was not "calibrated" to the Harvard impactor because the scarcity of PEM–Harvard comparison data above 30 μg/m^3 would make the accuracy of the correction uncertain for high-level measurements. Indoor, outdoor, and personal mass concentrations reported were all actual, measured values. The effect that intersample calibration would have on the results, however, is noted in the report where applicable.

Data Management System and Quality Control Measures

Everything from the collection of samples and questionnaire information to the consolidation and entry of such items into a compiled database was done in an organized manner, as illustrated in Figure 5. Whenever possible, questionnaire responses were entered directly into a notebook computer in the field. These questionnaire data and the associated database were stored securely either in a filing cabinet in a locked room or in a password-protected computer. By securing the data and ensuring accessibility to only the designated field technician and the study's principal investigators, subjects' identities have been completely protected in compliance with human subject guidelines.

Whenever samplers and substrates were provided to a field technician, chain of custody documentation was initiated. The laboratory technician signed and dated the chain-of-custody form and provided it with the samplers and substrates to the field technicians. Likewise, the field technicians signed and dated the chain-of-custody form to confirm receipt of the material and to record the dates of sampler deployment, sample collection, and return of the samples to the laboratory. The receiving laboratory's technician confirmed receipt and recorded when the samples were extracted, analyzed, and stored.

As part of the field sampling and data collection process, the field technician completed the sampling information sheets containing the home identification number, sampler type, sampler location, and sampling start and finish dates, times, and flow rates and distributed them directly to the appropriate principal investigators. In order to ensure that the analytical procedure was blind and consequently unbiased, the principal investigators stored these

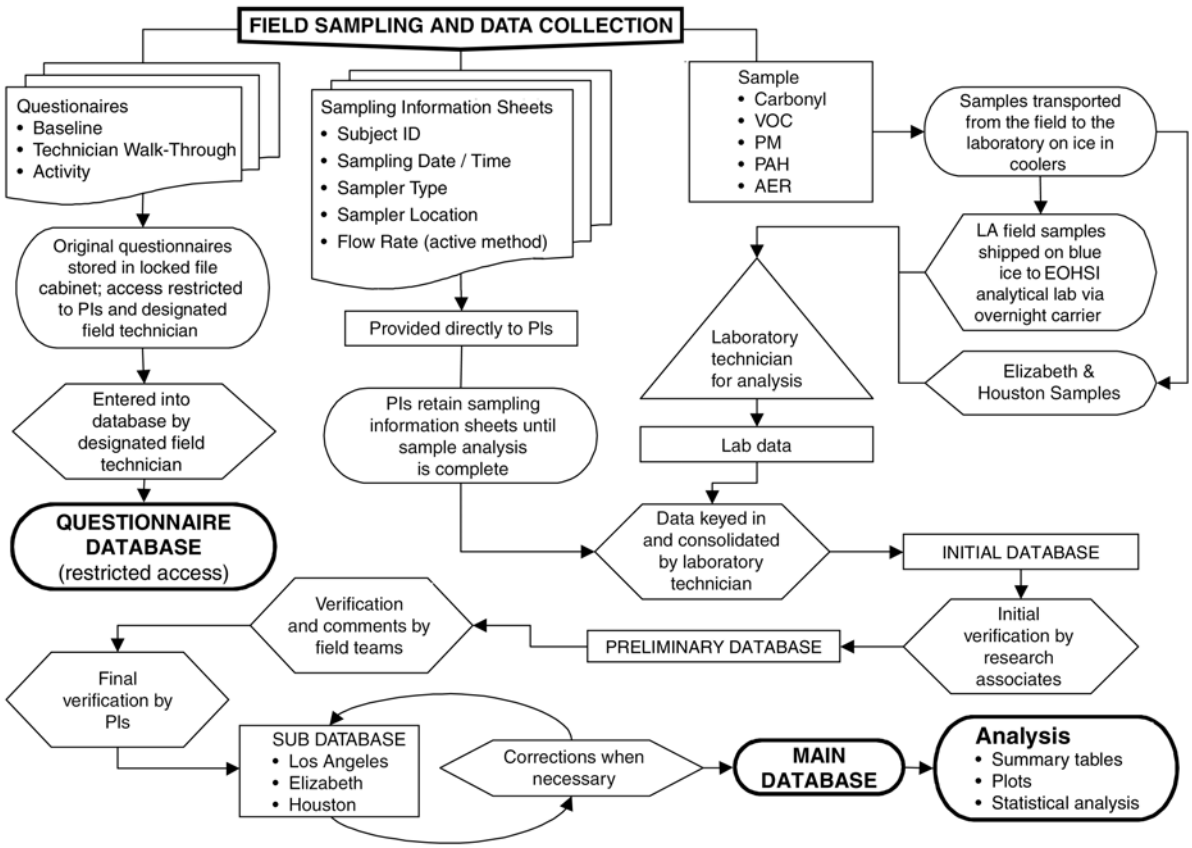

Figure 5. Flow diagram illustrating the transfer of information from field sampling to database formation and the quality control process.

documents until all of the samples from the respective homes were analyzed. After all home samples had been analyzed, the investigators provided the sampling information sheets to the laboratory technician, who keyed the information into an initial database. Some notes from the sampling information sheets were used to determine the validity of the samples such as observation notes (eg, sampling pump was off, as observed at "take down"), short sampling duration (< 42 hours), or large flow change (> 15%).

When the initial database was completed, a designated research associate who is experienced in analyzing the specific type of data reviewed the database. This review included cross-checking any keyed data entries against the original forms on which data has been recorded. The research associate double-checked all of the calculations used to transform the analytical data into the reported outdoor air concentrations. Finally, random data were confirmed by reapplying all of the calculations to the original analytical data. After the designated research associate completed the verification, the initial database was then classified as the preliminary database.

The field teams validated the preliminary database by reviewing the field sampling information and confirming the calculations that incorporated the information from the field sampling sheets. The field team then made any corrections necessary and noted the change, which was reported back to the originator for further confirmation of the needed correction. After the field teams made their comments and corrections, the principal investigators checked the data randomly by cross-referencing the electronic data for the chosen samples with the respective original data from the analytical results or sampling information sheets.

DATA ANALYSIS METHODS

DESCRIPTIVE ANALYSES

A number of descriptive analyses were performed to (1) present some basic characteristics of the data set, such as distributions of measured concentrations, AERs, and categorical data; (2) compare indoor, outdoor, and personal air concentrations (and in-vehicle concentrations for carbonyl compounds); (3) compare personal concentrations of adult–child pairs living in the same home; and (4) examine the effects of a number of variables (eg, season, home type, and city) on measured concentrations and indoor–outdoor relations. The techniques used in these descriptive analyses included univariate distribution analysis and bivariate scatter plotting. The analyses were done on a compound-by-compound basis. In addition to the analyses of the pooled data (all the data), analyses were done by several stratifying variables (eg, city, season, and home type). When duplicate samples obtained using the same sampling method were available, only the results from the primary samples were used in the data analyses. For PM samples obtained simultaneously using collocated Harvard impactors and PEMs, the indoor and outdoor concentrations were derived from the Harvard impactor data, whereas the personal air concentrations were derived from the PEM data. The carbonyl results from collocated DNPH and DNSH samples are presented separately, by measurement method (active or passive), in the tables and figures.

When concentrations were below the MDLs, we used one half of MDLs as censored data that were used in the analyses. The primary method used to test differences between paired data (indoor, outdoor, adult personal, and child personal concentrations) was an incomplete randomized block mixed model (SAS, Version 8, Cary NC) in which "home ID" was treated as a random effect. The error correlations between each pair of samples were allowed to differ by including a repeated statement with an unstructured covariance matrix in the SAS script. This method was used to minimize potential within-home correlations (between the first and the second measurements). This method is relatively insensitive to data below detection limits.

When the mixed model was used to compare paired adult–child personal concentrations within each home, the variations in concentrations from different children in the same home were taken into account because each home was considered as a block (33 homes had more than one child participating). Log transformation was carried out on the concentration data before using them in the mixed model. The Kruskal-Wallis test was used to examine the effects of variables such as season, city, and home characteristics. This nonparametric one-way ANOVA method was used because the data for most of the measured species generally did not meet the normal or log-normal distribution assumption of ANOVA or other types of parametric statistical methods.

MASS BALANCE ANALYSIS MODEL

The goal of the mass balance analysis was to estimate the contributions of indoor and outdoor sources to indoor concentrations. Assuming that a home can be approximated as a completely mixed reactor, the steady-state indoor concentration of an air contaminant is the sum of two terms: (1) the contribution derived from outdoor sources (outdoor concentrations, penetration, air exchanges, decay) and (2) the concentration derived from indoor sources (source strength, home volume, air exchanges, decay). Thus,

$$C_{In} = [aP/(a + k)]\ C_{Out} + (S/V)\ [1/(a + k)]\ , \quad (3)$$

where C_{In} (μg/m^3) is the steady-state indoor concentration of each species measured, C_{Out} (μg/m^3) is outdoor concentration, P (fraction between 0 and 1) is penetration through the building envelope), a (hr^{-1}) is the AER, k (hr^{-1}) is the decay rate due to deposition and reaction, S (μg/hr) is the indoor source strength, and V (m^3) is home volume.

For many nonreactive VOCs, the decay rates (k) are expected to approach zero because they have minimal losses due to diffusion to and reactions with surfaces. When k is 0 at steady state and P is 1, the slope of the regression C_{In} on C_{Out} is unity. Further, for homes with few or no indoor sources for a particular compound, the second term approaches zero. In this case (ie, homes for which outdoor sources drive indoor concentrations), indoor and outdoor air concentrations are expected to be distributed around the 1:1 line in an indoor–outdoor scatter plot.

The terms in equation 3 that were not measured were P, k, and S. In general, home-to-home variations in P and k values are relatively small for nonreactive pollutants. Therefore we applied the same P and k values across the homes to calculate home-specific indoor source strength (S) and outdoor contributions to indoor concentrations. From equation (3), we derived S as

$$S = V(a + k)C_{In} - aPVC_{Out}\ .$$

To calculate the fractional outdoor contribution to the indoor concentration, we applied the formula

$$aP\ [1/(a + k)]\ [C_{Out} / C_{In}]\ ,$$

inserting compound-specific values of P and k based on published data for the compound under analysis or for a compound with similar chemical properties if no data were available for the compound itself (Nazaroff and Cass 1986; Özkaynak et al 1996; Lachenmyer and Hidy 2000). In addition, for $PM_{2.5}$, we used P and k values estimated using nonlinear regression (NLIN procedure in SAS, Cary NC) of C_{Out}, C_{In}, and a for all homes in equation 3. The sensitivity of P and k to different reasonable assumptions was examined for the $PM_{2.5}$ mass data as well.

RANDOM COMPONENT SUPERPOSITION STATISTICAL MODEL

The random component superposition (RCS) model proposed by Ott and associates (2000) is based on the assumption of linear superposition of the outdoor and indoor components of exposure and the lack of correlation between these two components. It takes a similar form to the mass balance model (equation 3), as seen in equation 4:

$$H = \alpha B + A\ , \quad (4)$$

where H (μg/m^3) is the indoor concentration in a home (C_{In} in equation 3); αB is the contribution of outdoor sources to a home (the first term of equation 3): α is the dimensionless infiltration factor {$[aP/(a + k)]$ in equation 3}, and B (μg/m^3) is the outdoor concentration for a home (C_{Out} in equation 3); and A (μg/m^3) is the concentration derived from indoor sources {$[(S/V)\ [1/(a + k)]$} in equation 3).

This model assumes a lack of correlation between the concentrations resulting from indoor and outdoor sources. It does not provide estimates of indoor and outdoor contributions for individual homes. Instead it provides estimates of the distributions of these quantities for the entire set of homes. This model has been used previously (Ott et al 2000) to estimate the distributions of indoor and outdoor source contributions for pooled PM_{10} concentration data (ie, collected across many homes at different times) for which AER information was not available.

To examine the compatibility of the mass balance analysis and RCS model results, we estimated the distribution of outdoor and indoor contributions to indoor $PM_{2.5}$ concentrations obtained using the two models. The RCS model assumes a linear superposition of the outdoor and indoor contributions to indoor $PM_{2.5}$ and lack of correlation between these two components. Each home shares the same infiltration factor. It doesn't account for the variations in AERs from home to home.

RESULTS AND DISCUSSION

HOME CHARACTERISTICS AND DEMOGRAPHIC INFORMATION

In total, 306 homes were sampled: 105 homes in Los Angeles, 95 in Elizabeth, and 106 in Houston. The attributes of the homes shown in Table 9 are those most directly relevant to the analysis included in this report and were derived from the subjects' responses to several questions in the Baseline Questionnaire (Appendix H). Houston had the highest proportion of mobile homes; and no mobile homes were sampled in Elizabeth. More recently built homes (1995 to 2000) were found in Los Angeles than in the other two cities. Renovation within the year prior to sampling was defined in the Baseline Questionnaire as, "In the past year has there been a major renovation to this home or apartment, such as adding a room, putting up or taking down a wall, replacing windows, or refinishing floors." About 20% to 30% of the households reported such renovations in the past year.

Table 9. Number of Homes by City and Classified by Home Characteristics

Characteristic	Los Angeles	Elizabeth	Houston	Total
Number of homes	105	95	106	306
Home type				
Single-family	52	25	69	146
Multiple-family	4	6	1	11
Apartment	46	62	3	111
Mobile home	3	—	28	31
Don't know or missing data[a]	—	2	5	7
Year the home was built				
1995–2000	26	2	3	31
1985–1994	4	4	16	24
1975–1984	12	2	17	31
1960–1975	20	7	22	49
1945–1959	26	11	19	56
1900–1944	12	29	4	45
Before 1900	—	5	—	5
Don't know or missing data[a]	5	35	25	65
Renovations in year before sampling[b]				
Yes	23	33	33	89
No	78	58	68	204
Don't know or missing data[a]	4	4	5	13
Attached garage				
Yes	31	10	63	104
No	74	85	43	202
Presence of carpet(s) indoors				
Yes	17	16	10	43
No	79	68	81	228
Don't know or missing data[a]	9	11	15	35

[a] Subject chose the "Don't know" option to answer the question, or did not respond to the question (missing data).

[b] Renovation was described in the baseline questionnaire as, "In the ***past year*** has there been a major ***renovation*** to this house or apartment, such as adding a room, putting up or taking down a wall, replacing windows, or refinishing floors?"

A total of 309 adults and 118 children (ages 8 to 18) living in the 306 homes participated in personal air sampling. The adult subjects were evenly distributed across the three city locations. However, owing to differences among populations within the recruitment areas, the number of child subjects in Houston was more than three times the number of child subjects in Los Angeles or Elizabeth. Table 10 presents a summary of some demographic data (eg, gender, age, ethnicity, education level, and work status) provided by the participants in the Baseline Questionnaire (Appendix H). We purposely recruited subjects who were at home most of the time to evaluate the relations of indoor, outdoor, and personal air concentrations. Because a higher proportion of women were at home for most of the time than men, women subjects predominated the study. Because this was not a population-based study, we did not try to match the demographic and socioeconomic status of the population from city to city or between subjects living close to and far from outdoor sources with a city. As shown in Table 10, the distributions of ethnic backgrounds, education levels, and other personal characteristics were not even across the three cities. More subjects with higher levels of education participated in Los Angeles than in Elizabeth and Houston. No Mexican Americans were among the Elizabeth subjects.

SUBJECT RETENTION AND DATA COMPLETENESS

The number of homes to which first and second visits were made and the number of personal samples that were collected are provided in Tables 11 and 12. In Elizabeth, Houston, and Los Angeles, we retained 84%, 77%, and 88% of the subjects, respectively, for the repeat visit. Reasons for refusal of the second visit included (in order of frequency) loss of interest, burden too large, moved from the home, and illness.

The analyses reported here for active samples are based on only valid samples for which flow rate changes during sampling were less than 15% and collection times were longer

Table 10. Number of Participants in the Personal Sampling Portion by City and Demographic Group[a]

	Los Angeles		Elizabeth		Houston	
Demographic Group	Adult	Child	Adult	Child	Adult	Child
Number	105	23	101	22	103	73
Age[b]						
Mean	44	12	46	12	46	10
Minimum	20	7	17	8	23	6
Maximum	86	19	89	17	83	19
Gender						
Male	41	14	22	9	16	38
Female	64	9	79	13	87	35
Total	105	23	101	22	103	73
Cultural background[c]						
White	57	4	19	2	45	11
African American	—	—	8	2	3	—
American Indian	—	—	1	—	—	—
Asian or Pacific Islander	19	3	1	—	—	—
Mexican American	15	7	—		51	59
Hispanic white	8	3	28	7	3	3
Hispanic black	1	2	1	—	—	—
Hispanic other	—	2	44	9	2	
Other	6	2	—	—	—	—
Total	106	23	102	20	104	73
Highest level of education completed						
No schooling or kindergarten only	1		—		1	
Primary or middle school	2		14		11	
Some high school	2		12		15	
High school graduate	10		27		15	
Some college or technical school	28		23		31	
Undergraduate degree received	17		9		7	
Some graduate school	13		2		7	
Graduate degree received	32		8		3	
Total	105		95		90	
Work status						
Adult working full time	38		23		4	
Adult working part time	12		15		7	
Student, working	21		5		1	
Student, not working	4		—		1	
Self-employed working at home or homemaker	12		21		59	
Out of work just now but usually employed	1		6		3	
Retired	17		9		21	
Disabled or unable to work	—		5		5	
Total	105		84		101	

[a] Missing information was not included in this summary. A dash indicates no subjects in that group.

[b] Age was determined as of December 31, 2000.

[c] Some subjects selected multiple answers in responding to the question about cultural background.

Table 11. Number of Valid Measurements Collected for Chemical Species by City[a]

	Visits		Indoor		Outdoor		Personal Adult or (Child)		In-Vehicle	
	1st	2nd	1st	2nd	1st	2nd	1st	2nd	1st	2nd
Variable[b]										
Homes										
Elizabeth	101	82								
Houston	106	92								
Los Angeles	105	70								
Participants										
Elizabeth	120	93								
Houston	169	165								
Los Angeles	119	89								
Main Measurements[c]										
AER										
Elizabeth	87	76								
Houston	88	76								
Los Angeles	103	79								
VOCs										
Elizabeth	318	258	100	83	100	83	95 (24)	76 (17)	NA	
Houston	380	351	105	93	105	93	105 (64)	94 (71)	NA	
Los Angeles	315	242	98	76	98	77	100 (19)	75 (14)	NA	
Carbonyls[d]										
Elizabeth	305	256	93	84	98	85	101 (21)	75 (20)	7	3
Houston	309	307	79	80	74	80	77 (49)	85 (66)	20	13
Los Angeles	374	285	99	84	99	76	116 (11)	84 (13)	41	31
$PM_{2.5}$										
Elizabeth	167	132	53	43	57	46	44 (13)	33 (10)	NA	
Houston	176	141	59	47	63	47	53 (1)	45 (2)	NA	
Los Angeles	203	148	70	54	69	52	64 (0)	41 (1)	NA	

[a] The number of samples does not include either collocated, duplicate, or field blank samples. NA is not applicable.

[b] Homes and Participants indicate the number of homes or participants sampled (first visit) and the number of homes or participants sampled twice (second visit) for at least some air toxics.

[c] For each class of air toxics, the total number of measurements (sum of indoor, outdoor, personal, and in-vehicle) during all first and second visits, and the number of samples collected by type of sample are listed.

[d] With the exception of the in-vehicle samples, two types of samplers (DNSH and DNPH) were used to collect the carbonyl samples. The breakdown of these samples is shown in Table 12.

than 42 hours (87.5% of the target duration of 48 hours). For Elizabeth, Houston, and Los Angeles, respectively: totals comprised about 82%, 83%, and 91% of collected $PM_{2.5}$ samples; use of the passive sampler resulted in higher percentages of valid VOC samples (99.8%, 98%, and 91%); and for carbonyls, 88%, 86%, and 94% of the samples were valid.

To evaluate compliance of the subjects in carrying the personal samplers, we included a motion sensor in each pack with an active sampler. The sensor data could be reviewed to verify that the pack was not merely left in one spot for the entire sampling period. Four Elizabeth participants appeared not to have worn the sampler, and these samples were suspect as to whether they represented personal air samples. (Similar evaluations have not yet been done on the Los Angeles and Houston samples.) For children younger than 15 years, who wore only the passive samplers (for both VOCs and carbonyls), compliance was reviewed with both child and parent. As these samplers presented little strain on the participant, agreement of compliance by the parent and child seemed sufficient.

Table 12. Number of Valid Carbonyl Samples Collected by City, for each Sample Type[a] During Both First and Second Visits

City	Sampler Type	Visits		Indoor		Outdoor		Personal Adult or (Child)	
		1st	2nd	1st	2nd	1st	2nd	1st	2nd
Elizabeth	Active	94	33	27	11	33	12	27 (7)	7 (3)
	Passive	219	231	66	73	65	73	74 (14)	68 (17)
Houston	Active	80	22	28	6	24	6	28 (0)	9 (1)
	Passive	199	289	51	74	50	74	49 (49)	76 (65)
Los Angeles	Active	103	56	32	17	31	11	40 (0)	18 (0)
	Passive	222	211	67	67	68	65	76 (11)	66 (13)

[a] The in-vehicle sample type is not included here because all in-vehicle carbonyl samples were collected only on the DNPH sampling media (active sampling method).

AERs

A total of 739 nonblank CAT samples were collected during the main part of the study; of these, 112 were quality control samples (duplicates) and 103 were invalid because they were below the MDL, information on home volume was lacking, or there was a field failure. After having further excluded 15 measurements with AER values greater than 5 hr^{-1} (measurement upper limit), we obtained 509 AER measurements that were used for analysis.

The homes had AER values ranging from 0.14 hr^{-1} to 4.75 hr^{-1} in Los Angeles, 0.11 hr^{-1} to 4.48 hr^{-1} in Elizabeth, and 0.08 hr^{-1} to 4.3 hr^{-1} in Houston. The Houston homes had a median AER of 0.47 hr^{-1}, substantially lower than the Los Angeles median AER of 0.87 hr^{-1} and the Elizabeth median AER of 0.88 hr^{-1}. This was due to the higher prevalence of air conditioner use in the Houston homes (as judged by Baseline Questionnaire data).

Figure 6 shows box plots of AER values for each city by season. The seasons were defined according to the calendar year 2001. For the Houston homes, the median AER was higher during the fall and winter than during the spring and summer. In contrast, the Los Angeles homes had the lowest median AER during the winter and the highest during the spring. The Elizabeth homes showed higher median AERs in the summer and winter than in the spring and fall. When more detailed home parameters, such as the presence of central air conditioning or the home structure, were not considered, the indoor–outdoor temperature difference appeared to be a possible explanation for the main AER patterns observed in the three locations. During seasons when the indoor–outdoor temperature difference was greater (and thus the infiltration and exfiltration by convection was greater), the median AER values tended to be greater, suggesting that convection may have been a dominating mechanism of air exchange for the homes.

Figure 7 shows AER distributions by home type within each city. Elizabeth did not have mobile homes or trailers. In Los Angeles and Houston, the mobile homes appeared to have a slightly higher AER median than the other types of homes. (However, there were only five mobile homes in Los Angeles.) In Los Angeles, the median AER seemed to be higher for single-family houses than for multiple-family homes or apartments. A within-city comparison of buildings by age showed that the category of newest homes (built after 1995) was associated with the lowest median AER values in all three cities (Figure 8).

In an AER study conducted during the winter of 1991–1992 in the Los Angeles area (Wilson et al 1996), mean, median, and SD of AER values were 0.79 hr^{-1}, 0.64 hr^{-1}, and 0.57 hr^{-1}, respectively; whereas in our study the wintertime mean, median, and SD of AER values for the Los Angeles homes were 0.83 hr^{-1}, 0.76 hr^{-1}, and 0.47 hr^{-1}, respectively. Pandian and colleagues (1998) summarized nationwide residential AER values using 4590 measurements from different studies. New Jersey and Texas were included in the northeast and southeast regions, respectively. Mean, median, and SD of the AER values were 0.55 hr^{-1}, 0.42 hr^{-1}, and 0.47 hr^{-1}, respectively, for the northeast region, and 0.71 hr^{-1}, 0.62 hr^{-1}, and 0.56 hr^{-1} for the southeast region. In our study, the AER values in Houston (mean, median, and SD of 0.66 hr^{-1}, 0.47 hr^{-1}, and 0.64 hr^{-1}, respectively) were similar to the southeast region survey data from Pandian and associates, but the AER values for Elizabeth homes (mean, median, and SD of 1.20 hr^{-1}, 0.88 hr^{-1}, and 0.97 hr^{-1}, respectively) were considerably higher than the northeast region survey data. One possible reason for this difference was that we used a

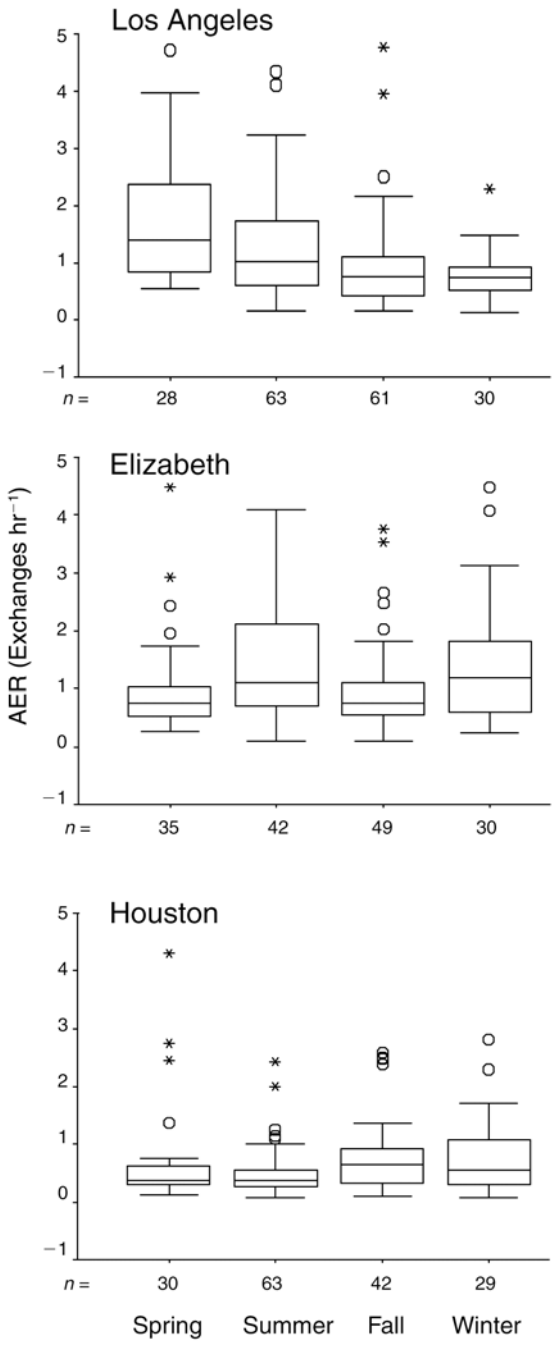

Figure 6. Seasonal variations of AERs in Los Angeles, Elizabeth, and Houston. *n* is the number of samples analyzed for each season. The box plots summarize the median, lower quartile, upper quartile, lower range, and upper range. White circles (○) represent outliers between 1.5 and 3 box lengths from the upper or lower edge of the box. Asterisks (*) represent extreme values more than 3 box lengths from the upper or lower edge of the box. Spring was defined as March 21 to June 20; summer, June 21 to September 20; fall, September 21 to December 20; winter, December 21 to March 20.

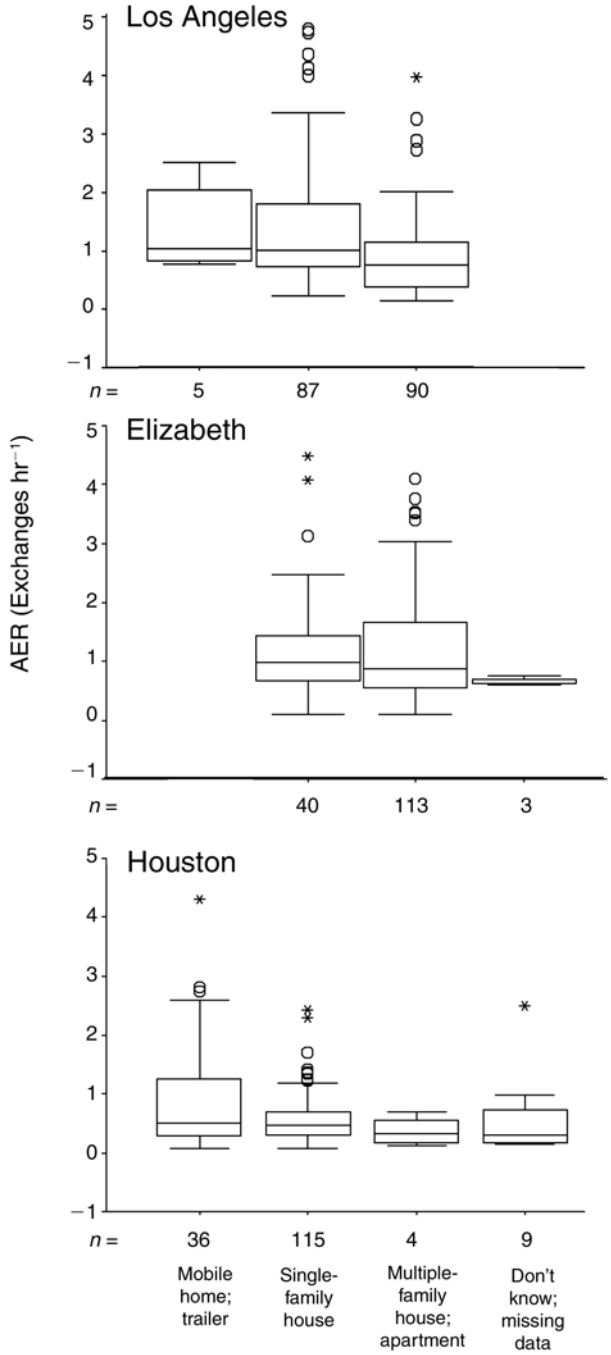

Figure 7. AER variations with different home types in Los Angeles, Elizabeth, and Houston. *n* is the number of samples analyzed for each type of dwelling. The box plots summarize the median, lower quartile, upper quartile, lower range, and upper range. White circles (○) represent outliers between 1.5 and 3 box lengths from the upper or lower edge of the box. Asterisks (*) represent extreme values more than 3 box lengths from the upper or lower edge of the box.

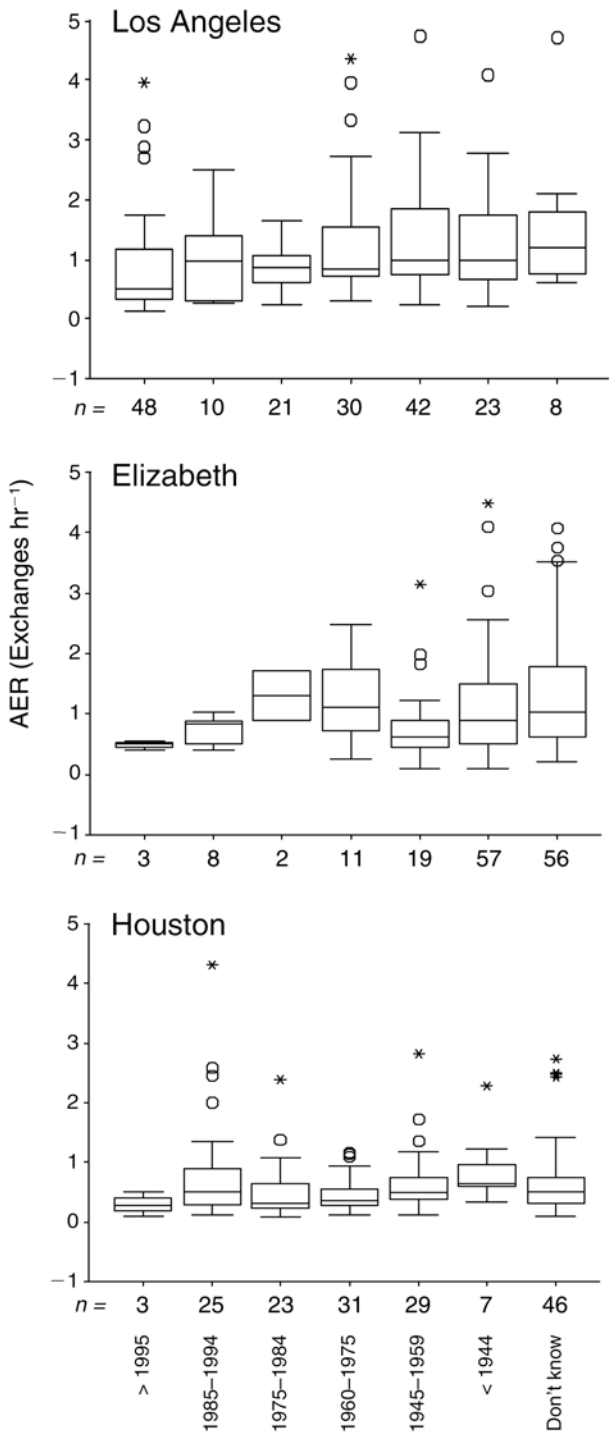

Figure 8. AER variations by building age in Los Angeles, Elizabeth, and Houston. Years when the homes were built are shown on the x-axis. *n* is the number of samples analyzed for each time span. Box plots summarize the median, lower quartile, upper quartile, lower range, and upper range. White circles (○) represent outliers between 1.5 and 3 box lengths from the upper or lower edge of the box. Asterisks (*) represent extreme values more than 3 box lengths from the upper or lower edge of the box.

maximum measurable AER (5 hr^{-1}) that was considerably higher than had been used for the previous studies. The highest AER reported by Pandian and coworkers (1998) was approximately 2 hr^{-1}. Also, AERs in New Jersey are expected to vary considerably from area to area with the age of the homes. The homes in urban Elizabeth are primarily older, whereas many newer homes are found in suburban areas of New Jersey.

INDOOR, OUTDOOR, AND PERSONAL CONCENTRATIONS AND THEIR RELATIONS

Descriptive summaries of all the data (pooled) are presented by outdoor concentrations (Table 13), indoor concentrations (Table 14), adult personal concentrations (Table 15), child personal concentrations (Table 16), and in-vehicle concentrations (Table 17). Results from univariate analyses for all the measured pollutants are presented in Appendix E. These include data distributions by city, personal concentrations (child versus adult), season, and home type for indoor concentrations. The data are discussed in this section by the generic type of the pollutants measured (VOCs, carbonyl compounds, and $PM_{2.5}$).

VOCs

Less than 50% of indoor, outdoor, or personal samples had methylene chloride and trichloroethylene above the MDLs. Styrene was detected in only 29% and 61% of outdoor and indoor samples, and in 69% and 83% of adult and child personal samples, respectively. Chloroform, toluene, α-pinene, β-pinene, *d*-limonene, and *p*-dichlorobenzene were detected in less than 60% of outdoor samples but in the majority of the personal and indoor samples (see Tables 13–16). With the exception of carbon tetrachloride and tetrachloroethylene, the personal and indoor VOC concentrations were higher than outdoor concentrations. The differences were all statistically significant ($P < 0.05$), according to results from the incomplete randomized block mixed model described in the section Descriptive Analyses.

The individual compound concentrations were highest in the personal air samples and next highest in the indoor air samples, especially for compounds that have consumer uses, such as *p*-dichlorobenzene, α-pinene, *d*-limonene, and tetrachloroethylene.

An intercity comparison of outdoor VOC concentrations, using the Kruskal-Wallis test at $\alpha = 0.05$, showed significant differences for all the VOCs reported here (Appendix E, Table E.1). Because the homes selected were not a population-based sample of the three cities and there are underlying differences in the homes and climates of the

Table 13. Descriptive Summary of Outdoor Air Concentrations (μg/m^3)

Species[a]	n[b]	Mean	SD	Median	Percentile				Percentage Above LOD
					1st	5th	95th	99th	
VOCs	555								
Methylene chloride		0.95	2.24	0.84	0.04	0.07	2.46	9.32	31.2
MTBE		8.10	9.99	5.32	0.43	0.44	22.1	51.2	94.6
Chloroform	554	0.32	0.99	0.17	0.08	0.09	0.76	2.35	20.8
Carbon tetrachloride		0.71	1.31	0.64	0.13	0.34	1.00	1.58	95.7
Benzene		2.15	2.11	1.68	0.41	0.48	5.16	11.1	79.8
Trichloroethylene		0.30	1.30	0.12	0.04	0.05	0.79	1.90	33.7
Toluene		7.09	6.47	5.42	1.30	2.82	19.6	32.0	41.1
Tetrachloroethylene		1.00	2.17	0.56	0.09	0.11	3.17	7.75	44.3
Ethyl benzene		1.29	1.87	0.93	0.15	0.30	3.04	7.05	78.7
m- & p-Xylenes		3.57	4.15	2.49	0.25	0.53	10.0	19.1	94.8
o-Xylene		1.48	3.90	0.96	0.10	0.17	3.23	7.17	91.7
Styrene		0.48	2.08	0.17	0.07	0.11	1.29	4.15	29.2
α-Pinene		0.89	4.18	0.32	0.04	0.07	1.90	16.5	50.6
β-Pinene		0.53	2.20	0.18	0.10	0.12	1.43	7.35	17.7
d-Limonene		2.39	6.26	1.27	0.24	0.28	6.54	37.8	12.8
p-Dichlorobenzene		2.25	17.15	0.72	0.09	0.19	3.66	18.3	24.7
Carbonyls (passive method)	395								
Formaldehyde		6.38	2.52	6.53	1.16	2.17	10.1	12.4	99.7
Acetaldehyde		6.94	4.96	5.44	0.32	1.46	15.0	25.9	98.0
Acetone		9.75	69.4	4.39	0.20	0.20	19.6	55.3	91.4
Acrolein[c]		6.28	101	0.47	0.06	0.07	4.60	11.9	67.3
Propionaldehyde		1.57	1.14	1.37	0.02	0.06	3.69	5.37	93.9
Crotonaldehyde		0.77	5.29	0.26	0.06	0.06	1.97	3.85	61.8
Benzaldehyde		2.03	1.27	1.87	0.12	0.13	4.22	5.93	93.9
Hexaldehyde		2.31	3.21	2.06	0.06	0.25	4.68	6.49	99.0
Glyoxal		1.81	0.90	1.82	0.06	0.45	3.48	4.33	99.0
Methylglyoxal		2.05	1.07	2.05	0.10	0.28	3.99	5.12	96.5
Carbonyls (active method)	117								
Formaldehyde		3.82	3.02	3.00	0.25	0.32	12.9	14.6	91.5
Acetaldehyde		3.21	1.65	2.91	0.33	0.88	7.04	8.27	82.9
Acetone		1.73	1.21	1.32	0.31	0.54	4.25	5.69	32.5
Propionaldehyde		0.99	0.87	0.85	0.10	0.22	2.35	7.27	86.3
Benzaldehyde		2.56	2.18	2.07	0.31	0.34	7.42	8.79	66.7
Glyoxal		0.49	0.36	0.47	0.04	0.04	1.02	2.35	70.9
Methylglyoxal		2.26	14.7	0.28	0.00	0.00	1.03	129	43.6
Butyraldehyde		0.86	0.44	0.82	0.08	0.28	1.60	3.01	42.7
Isovaleraldehyde		0.54	0.58	0.38	0.06	0.08	1.44	4.29	10.3
Valeraldehyde		0.78	0.52	0.65	0.09	0.24	2.05	2.48	36.8
$PM_{2.5}$[d]	334	18.1	10.7	15.5	5.44	6.52	33.9	71.0	100

[a] The fractions of nondetected samples were > 90% for *o*-tolualdehyde (MDL = 0.29 μg/m^3), *m*- & *p*-tolualdehydes (MDL = 0.15 μg/m^3), and dimethylbenzaldehyde (MDL = 0.25 μg/m^3). Their distributions are not reported here.

[b] Total samples for each compound within a group, unless otherwise noted.

[c] The high standard deviation of acrolein is caused by one extreme balue (2018 μg/m^3) in the data set.

[d] After removing two outliers, outdoor $PM_{2.5}$ mean ± SD is 15.4 ± 9.4 μg/m^3.

Table 14. Descriptive Summary of Indoor Air Concentrations (μg/m^3)

Species[a]	n[b]	Mean	SD	Median	Percentile				Percentage Above LOD
					1st	5th	95th	99th	
VOCs	554								
Methylene chloride		2.31	10.6	0.84	0.04	0.11	7.50	33.7	44.9
MTBE	553	11.8	27.3	5.98	0.44	0.44	36.0	196	93.1
Chloroform		1.86	2.97	0.92	0.11	0.17	6.34	14.8	79.4
Carbon tetrachloride		0.71	0.98	0.62	0.13	0.27	1.10	2.03	94.4
Benzene		3.50	5.15	2.19	0.48	0.48	10.0	36.4	85.4
Trichloroethylene		0.46	1.56	0.12	0.04	0.05	1.36	7.84	41.3
Toluene		15.4	24.4	10.1	2.83	3.02	39.8	122	70.0
Tetrachloroethylene		1.81	4.48	0.56	0.10	0.11	6.01	20.9	62.8
Ethyl benzene		2.52	4.74	1.46	0.32	0.36	7.62	26.7	86.3
m- & p-Xylenes		7.33	15.9	4.07	0.25	0.70	22.2	75.2	97.3
o-Xylene		2.48	4.77	1.46	0.17	0.36	7.24	22.6	95.3
Styrene		1.41	4.26	0.50	0.11	0.16	5.13	23.5	61.4
α-Pinene		4.87	13.6	1.22	0.04	0.07	18.1	78.6	89.7
β-Pinene		4.80	11.0	1.47	0.12	0.18	20.4	62.2	75.3
d-Limonene		31.0	107	9.67	1.10	1.27	103	273	81.0
p-Dichlorobenzene		68.9	304	1.44	0.19	0.29	344	1790	58.8
Carbonyls (passive method)	398								
Formaldehyde		21.6	7.13	20.1	11.1	12.9	32.5	53.8	100
Acetaldehyde		23.2	18.6	18.9	3.24	8.01	55.1	119	99.7
Acetone		14.0	21.7	8.25	0.20	1.12	45.8	128	97.2
Acrolein		1.71	7.65	0.62	0.07	0.07	5.27	14.8	71.6
Propionaldehyde		2.05	2.03	1.76	0.03	0.23	3.77	8.23	96.7
Crotonaldehyde		0.70	0.87	0.45	0.06	0.06	2.61	4.42	70.1
Benzaldehyde		3.02	1.35	2.90	0.12	0.98	5.38	7.30	97.7
Hexaldehyde		4.53	2.74	3.79	0.85	1.63	9.52	13.5	99.7
Glyoxal		2.60	0.94	2.55	0.25	1.12	4.38	5.14	99.7
Methylglyoxal		2.86	1.84	2.72	0.24	1.05	4.72	5.39	99.2
Carbonyls (active method)	121								
Formaldehyde		25.2	13.5	23.4	0.41	7.12	55.3	72.5	98.3
Acetaldehyde		11.9	10.5	9.23	0.36	0.90	35.0	48.1	86.8
Acetone		2.59	4.75	1.46	0.42	0.57	7.79	36.9	33.1
Propionaldehyde		2.11	1.36	1.99	0.09	0.24	4.72	7.40	91.7
Benzaldehyde		2.13	1.83	1.59	0.33	0.44	6.54	10.3	76.9
Glyoxal		0.87	0.56	0.80	0.03	0.07	1.76	3.10	76.9
Methylglyoxal		2.74	10.2	1.19	0.00	0.00	4.22	88.4	66.9
Butyraldehyde		1.41	0.81	1.20	0.26	0.54	3.18	4.81	62.0
Isovaleraldehyde		1.12	0.83	0.97	0.07	0.13	2.85	4.88	56.2
Valeraldehyde		2.22	1.68	1.75	0.19	0.54	5.88	10.2	71.1
$PM_{2.5}$	326	17.6	12.6	14.4	3.16	4.86	40.3	69.3	100

[a] The fractions of nondetected samples were > 90% for *o*-tolualdehyde (MDL = 0.29 μg/m^3), *m- & p*-tolualdehydes (MDL = 0.15 μg/m^3), and dimethylbenzaldehyde (MDL = 0.25 μg/m^3). Their distributions are not reported here.

[b] Total samples for each compound within a group, unless otherwise noted.

Table 15. Descriptive Summary of Adult Personal Air Concentrations (μg/m^3)

Species[a]	n[b]	Mean	SD	Median	Percentile 1st	Percentile 5th	Percentile 95th	Percentile 99th	Percentage Above LOD
VOCs	545								
Methylene chloride		3.04	17.1	0.84	0.04	0.13	7.39	32.9	47.2
MTBE	544	14.8	42.7	7.14	0.44	0.94	42.7	129	96.0
Chloroform	542	4.20	52.6	1.04	0.14	0.17	6.34	17.4	84.9
Carbon tetrachloride		0.79	2.44	0.61	0.13	0.27	1.08	2.00	94.3
Benzene		3.64	5.30	2.39	0.48	0.48	10.7	27.4	87.5
Trichloroethylene		0.95	8.83	0.13	0.04	0.05	1.88	13.3	49.0
Toluene		19.2	37.3	12.2	2.81	3.02	50.2	138	73.9
Tetrachloroethylene		7.14	112	0.61	0.10	0.13	7.21	57.4	69.5
Ethyl benzene		2.79	5.13	1.68	0.36	0.36	7.48	28.8	87.3
m- & p-Xylenes		8.07	15.5	4.42	0.25	0.93	22.7	75.2	97.8
o-Xylene		2.89	5.58	1.73	0.17	0.47	8.14	23.0	97.8
Styrene		1.51	4.32	0.57	0.13	0.17	5.51	21.4	69.0
α-Pinene	544	4.21	11.1	1.21	0.05	0.07	17.6	39.4	90.1
β-Pinene		5.48	13.1	1.65	0.11	0.18	22.4	72.4	78.9
d-Limonene		41.2	239	11.8	1.27	1.27	112	287	85.0
p-Dichlorobenzene		56.7	229	1.88	0.18	0.35	314	1480	63.5
Carbonyls (passive method)	409								
Formaldehyde		21.7	9.03	20.5	9.62	12.4	34.0	45.4	100
Acetaldehyde		22.9	14.9	18.7	5.12	8.12	53.8	86.1	99.8
Acetone		25.9	112	8.36	0.20	1.74	57.7	700	98.0
Acrolein		12.9	138	0.51	0.07	0.07	5.12	11.2	68.5
Propionaldehyde		2.00	1.11	1.91	0.03	0.29	3.92	5.26	96.3
Crotonaldehyde		1.23	6.34	0.44	0.06	0.06	2.57	8.61	67.7
Benzaldehyde		3.36	1.99	3.04	0.12	1.11	6.45	10.9	97.1
Hexaldehyde		5.26	7.08	4.17	0.70	1.63	9.64	24.0	100
Glyoxal		2.64	1.36	2.44	0.67	1.21	4.29	7.02	100
Methylglyoxal		2.75	1.08	2.71	0.36	1.11	4.79	5.57	100
Carbonyls (active method)	129								
Formaldehyde		26.3	14.3	23.5	1.62	7.93	53.1	88.0	99.2
Acetaldehyde		15.9	11.2	13.5	0.58	1.78	41.7	54.7	92.2
Acetone		6.58	14.3	2.93	0.75	0.91	21.2	107	49.6
Propionaldehyde		3.07	3.15	2.44	0.27	0.54	6.59	23.0	93.0
Benzaldehyde		2.25	2.08	1.61	0.38	0.59	7.12	12.2	48.1
Glyoxal		0.95	0.66	0.90	0.07	0.10	2.05	3.54	64.3
Methylglyoxal		2.81	9.87	1.64	0.00	0.00	4.41	83.9	66.7
Butyraldehyde		2.06	1.29	1.85	0.09	0.60	3.94	8.43	59.7
Isovaleraldehyde		1.34	0.93	1.23	0.07	0.10	2.96	4.40	51.2
Valeraldehyde		3.09	3.94	2.18	0.24	0.65	7.05	32.0	73.6
$PM_{2.5}$	280	36.3	23.7	30.6	5.62	12.7	87.4	139	100

[a] The fractions of nondetected samples were > 90% for *o*-tolualdehyde (MDL = 0.29 μg/m^3), *m- & p*-tolualdehydes (MDL = 0.15 μg/m^3), and dimethylbenzaldehyde (MDL = 0.25 μg/m^3). Their distributions are not reported here.

[b] Total samples for each compound within a group, unless otherwise noted.

Table 16. Descriptive Summary of Child Personal Air Concentrations (μg/m^3)

Species[a]	n[b]	Mean	SD	Median	Percentile				Percentage Above LOD
					1st	5th	95th	99th	
VOCs	209								
Methylene chloride		1.66	6.51	0.69	0.04	0.06	5.25	16.7	64.6
MTBE		11.7	22.1	7.03	0.44	0.56	30.2	193	94.7
Chloroform		2.03	3.63	1.14	0.14	0.17	7.47	16.4	88.0
Carbon tetrachloride		0.56	0.17	0.56	0.13	0.21	0.83	0.97	91.9
Benzene		4.16	5.57	2.79	0.36	0.48	12.0	43.6	94.3
Trichloroethylene		0.31	0.89	0.12	0.04	0.04	0.89	7.08	34.9
Toluene		18.4	27.8	12.2	1.44	2.94	57.2	220	80.9
Tetrachloroethylene		2.81	15.9	0.56	0.09	0.12	7.34	81.8	76.6
Ethyl benzene		3.34	6.35	1.95	0.30	0.36	10.3	54.2	94.3
m- & p-Xylenes		8.87	16.7	5.15	0.43	1.38	28.2	63.1	98.6
o-Xylene		2.91	4.88	1.96	0.11	0.52	7.97	22.2	97.1
Styrene		1.69	4.37	0.67	0.14	0.16	6.89	30.6	83.3
α-Pinene		3.48	5.06	1.42	0.06	0.08	15.3	25.4	96.2
β-Pinene		5.32	6.23	2.85	0.18	0.18	18.2	29.8	86.1
d-Limonene		32.1	49.7	17.4	1.27	1.27	111	168	93.3
p-Dichlorobenzene		122	314	4.18	0.27	0.44	979	1460	76.1
Carbonyls (passive method)	169								
Formaldehyde		20.8	7.18	20.1	7.09	11.4	33.9	47.4	100
Acetaldehyde		24.9	19.0	20.0	2.81	8.12	65.9	112	100
Acetone		29.1	98.4	11.5	1.41	4.25	81.0	759	99.4
Acrolein		10.9	105	0.87	0.06	0.07	8.04	504	75.7
Propionaldehyde		5.15	34.1	2.33	0.07	0.93	4.87	141	98.2
Crotonaldehyde		1.50	6.51	0.58	0.06	0.06	2.84	55.9	75.7
Benzaldehyde		3.18	1.77	2.89	0.12	1.15	5.59	12.8	97.0
Hexaldehyde		6.44	11.5	4.68	0.96	2.22	12.1	78.8	100
Glyoxal		2.93	1.84	2.78	0.77	1.25	4.25	15.1	100
Methylglyoxal		3.09	1.34	2.92	0.11	1.46	5.36	9.19	98.8
Carbonyls (active method)	11								
Formaldehyde		24.3	15.1	18.8	10.3	10.3	62.4	62.4	100
Acetaldehyde		22.6	15.3	21.1	4.33	4.33	47.0	47.0	100
Acetone		15.6	18.9	6.91	1.03	1.03	51.4	51.4	63.6
Propionaldehyde		4.88	5.55	3.54	1.36	1.36	20.6	20.6	100
Benzaldehyde		2.31	2.12	1.60	0.59	0.59	7.44	7.44	54.5
Glyoxal		1.27	0.94	0.99	0.14	0.14	3.59	3.59	90.9
Methylglyoxal		2.90	4.26	1.53	0.00	0.00	15.1	15.1	72.7
Butyraldehyde		2.50	2.19	2.35	0.67	0.67	8.07	8.07	72.7
Isovaleraldehyde		1.56	1.07	1.39	0.16	0.16	3.41	3.41	72.7
Valeraldehyde		5.74	9.27	3.68	0.61	0.61	33.2	33.2	81.8
$PM_{2.5}$	27	51.5	30.1	39.2	19.9	20.3	136	150	100

[a] The fractions of nondetected samples were > 90% for *o*-tolualdehyde (MDL = 0.29 μg/m^3), *m- & p*-tolualdehydes (MDL = 0.15 μg/m^3), and dimethylbenzaldehyde (MDL = 0.25 μg/m^3). Their distributions are not reported here.

[b] Total samples for each compound within a group.

three regions, the differences measured indicate that caution is needed when attempting to combine the samples into a single data set. In general, differences in home types and climatic conditions are important when using the data to evaluate exposure models that are intended to be applicable to a wide range of conditions.

Compared with the Elizabeth and Houston homes, the Los Angeles homes had markedly higher median outdoor concentrations of MTBE, *m- & p*-xylenes, and *o*-xylene. Measured concentrations of these three VOCs may reflect larger influences of mobile sources on the homes sampled in Los Angeles. Similar comparisons showed significant intercity differences in indoor concentrations of all the VOCs except toluene. The Los Angeles homes had the highest median indoor concentration of MTBE and *o*-xylene. This finding is consistent with the results from the intercity comparison of outdoor VOC concentrations, suggesting these compounds were mainly generated from mobile sources.

Intercity differences in personal concentrations were observed for the majority of the VOCs, with the exceptions of toluene for both the adult and child samples, carbon tetrachloride for the adult samples, and MTBE and α-pinene for the child samples (see Tables E.3 and E.4). The Houston homes had strikingly higher indoor and personal median concentrations of several VOCs, mainly of indoor origin (*d*-limonene, *p*-dichlorobenzene, and β-pinene), than the Los Angeles and Elizabeth homes, at least in part because of the lower AERs in the Houston homes.

A comparison of paired adult–child personal concentrations within the same home showed significant differences, at $\alpha = 0.05$, for only two VOCs (MTBE and toluene) in Los Angeles and none in Elizabeth (Appendix E, Tables E.6 and E.7). Personal concentrations of MTBE were significantly higher for the Los Angeles adults, whereas personal concentrations of toluene were significantly higher for the Los Angeles children (Table E.6). In contrast, in Houston, personal concentrations of 9 of the 16 VOCs were significantly higher for adults than for children (Table E.8). The reasons for these observations need to be further examined.

In Los Angeles, median outdoor concentrations of MTBE, benzene, ethyl benzene, *m- & p*-xylenes, and *o*-xylene were all markedly higher in fall and winter than in spring and summer (Table E.9). In Elizabeth, only *m- & p*-xylenes and *o*-xylene had higher median outdoor concentrations in fall and winter (Table E.10). In contrast, in Houston, the highest median concentrations of MTBE, benzene, and *m- & p*-xylenes all appeared in fall, whereas the next-highest median concentrations of these three VOCs all appeared in summer (Table E.11).

In Los Angeles, the seasonal patterns for indoor concentrations of MTBE, benzene, ethyl benzene, *m- & p*-xylenes, and *o*-xylene were the same as those for outdoor concentrations; that is, they were higher in fall and winter than in spring and summer (Table E.12). The indoor median concentration of chloroform, however, appeared to be markedly lower in the spring than in other seasons. In Elizabeth, statistically significant seasonal differences were found for a number of VOCs, but few of the indoor median concentrations differed significantly (except for the benzene indoor median concentrations, which were lowest in summer and highest in winter by nearly a factor of 2; see Table E.13). In Houston, the seasonal pattern of indoor MTBE concentrations was the same as the outdoor pattern, consistent with the predominantly outdoor origin of this VOC. Seasonal differences in indoor concentrations were also found for styrene and α-pinene in the Houston homes (Table E.14).

Compared with indoor and outdoor VOC concentrations, overall in all three cities, adult personal VOC concentrations showed greater seasonal differences for more compounds. This may reflect not only seasonal variations in sources and meteorologic effects, but also seasonal variations in personal activities. For example, seasonal differences were found for benzene and toluene in Los Angeles (Table E.15).

The sample sizes from which child personal concentration data were obtained in Los Angeles and Elizabeth were too small for meaningful interseason comparisons. In the Houston child personal concentration data (not shown), five VOCs had significant seasonal differences, benzene had a much higher median concentration in fall and winter than in spring and summer, and concentrations of chloroform and β-pinene were higher in summer than in other seasons.

Home type significantly affected indoor concentrations of most VOCs in Los Angeles, about half of the VOCs in Houston, and only MTBE (with $P = 0.05$) in Elizabeth (Tables E.18 through E.20). In most Los Angeles homes, the highest median concentrations were found in multiple-family homes or apartments, which had the lowest AERs of the three home types (see Figure 7). In Houston, the highest median concentrations of most VOCs were found in single-family homes, which had a lower median AER than that for mobile homes.

Scatter plots (Appendix F) of the indoor and outdoor air concentrations provide qualitative insights into the influence of indoor and outdoor sources on indoor concentrations and on the proportion of the homes for which infiltration from outdoor air is the major source of air pollutants indoors. Similarly, the scatter plots of the personal and outdoor air concentrations and the personal and

indoor air concentrations indicate whether outdoor air, indoor sources, personal activities, or some combination of these are the dominant contributors to inhalation exposure. Data points that lie close to and are randomly distributed around the 1:1 line indicate a strong association between the air concentrations portrayed on the two axes; data points that fall along either the indoor or the personal air axis indicate that indoor emissions or personal activities dominate the indoor and personal air concentrations, respectively, and data points that follow a straight line with a slope less than unity in the indoor–outdoor air plots suggest the absence of indoor sources and losses of the substance during the penetration process and within the indoor environment.

Within the data set, some pollutants appeared to be predominately influenced by indoor sources, and others by outdoor sources, or by personal activities (see Appendix F). Some compounds were substantially influenced by more than one category. For example, for carbon tetrachloride and MTBE, the vast majority of data points fell along the 1:1 line on the indoor–outdoor plots (Appendix F), indicating that outdoor air sources dominated the indoor air concentration for a majority of the homes, although a few of the indoor air data points for these compounds were elevated. The vast majority of the respective concentrations of chloroform, styrene, α-pinene, β-pinene, *d*-limonene, and *p*-dichlorobenzene, except for those near the MDLs, were nearly parallel to the indoor air axis on the indoor–outdoor plots, consistent with indoor sources being the major contributors for these compounds (Appendix F).

A portion of the data points on the scatter plots for methylene chloride, benzene, tetrachloroethylene, trichloroethylene, ethyl benzene, *m- & p*-xylenes, and *o*-xylene were randomly distributed about the 1:1 line, and another portion showed highly elevated indoor compared with outdoor air concentrations. This pattern was best explained by some homes having few to no indoor emissions of these compounds and other homes having large indoor sources; the transition region on the scatter plots indicates homes that had similar contributions from outdoor air and indoor emissions. The highest indoor air concentrations in this group were from homes that showed major deviations from the 1:1 line where indoor sources overwhelmed contributions from outdoors.

Most of the toluene data were skewed toward higher indoor air concentration levels, suggesting an indoor source for this compound in most homes. In some homes personal concentrations of toluene were substantially higher than indoor concentrations (Figure 9). The indoor sources, which vary by compound, include attached garages for aromatic compounds; dry-cleaned clothing for tetrachloroethylene; use of chlorinated drinking water for

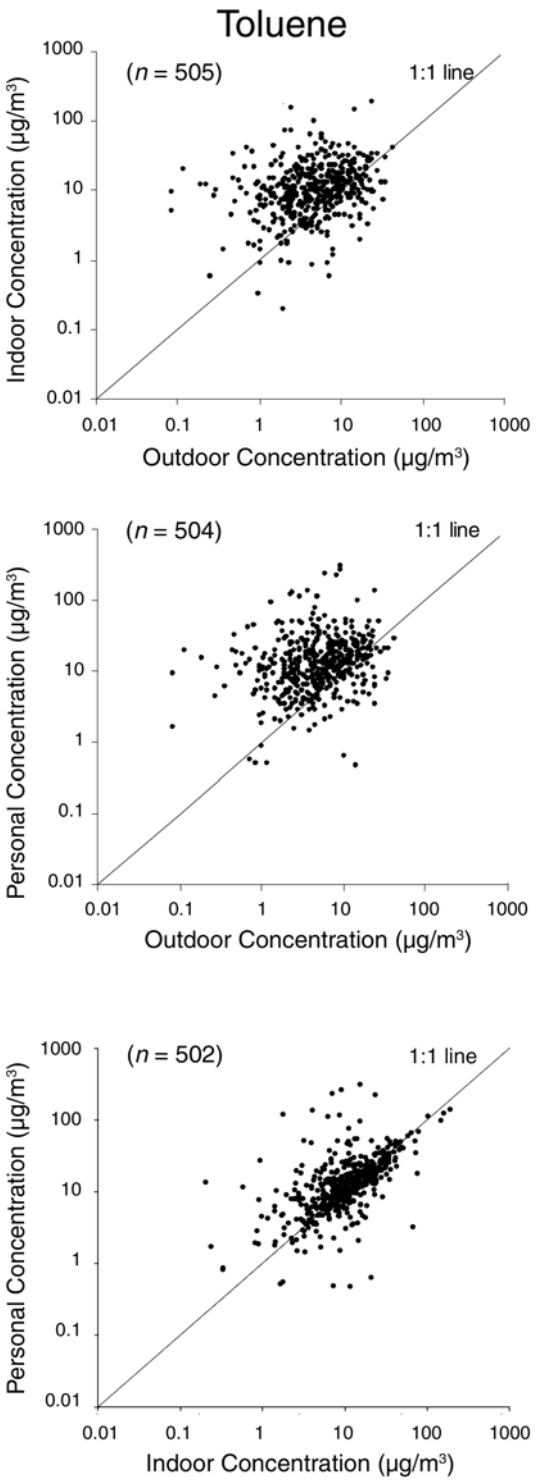

Figure 9. Toluene air concentrations for indoor–outdoor, personal–outdoor, and personal–indoor comparisons. Shown on logarithmic scales.

chloroform; cleaning products for toluene, trichloroethylene, and methylene chloride; and air fresheners for α-pinene, β-pinene, *d*-limonene, and *p*-dichlorobenzene. The questionnaire information could be used to conduct a full evaluation of indoor sources in the future, but this was beyond the scope of the current analyses.

The data points on the scatter plots for personal and indoor concentrations for all compounds were generally distributed around the 1:1 line. Thus, regardless of whether the major contributions to indoor VOC concentrations were from infiltration of outdoor air or emissions from indoor sources, indoor air appeared to be the dominant medium for inhalation exposure to VOCs. Compounds with strong indoor–outdoor air correlations also showed strong associations between the personal–outdoor concentrations, as evidenced by a close fit to the 1:1 line, but with more scatter than was observed for the personal–indoor relations. For compounds with little association between the indoor and outdoor concentrations, associations between the personal and outdoor concentrations were weaker.

Individual data points also deviated from the 1:1 line in the personal–indoor concentrations. When the indoor air concentration was higher than the personal air concentration, it was likely that there were emissions into the indoor air when the individual was not at home; when the personal air concentration was higher, it was likely that the exposure occurred away from home or by an activity that generated emissions very close to the participant's breathing zone. At a later time, these deviations can be explored using responses to the Activity Questionnaire and data from the activity log to evaluate what activities may have affected individual inhalation exposures. Reviewing the scatter plots was an important first step in understanding the data and can provide a guide for developing more advanced statistical data analysis, such as multivariate analysis, to understand what sources dominate inhalation exposures, and what the mechanisms control transport of outdoor-generated compounds to the indoor environment and to individuals.

The ability to compare the indoor, outdoor, and personal VOC concentration measurements obtained in this study with those reported in other studies is limited by the variability in monitoring methods, length of sampling, and reporting modes of different investigators. In addition, there have been comparatively few studies as large as this one in which indoor, outdoor, and personal air concentrations of the same VOCs were measured simultaneously. For example, methylene chloride has not been typically included in large personal exposure studies. In the Toxic Exposure Assessment: A Columbia/Harvard Study (TEACH study) in New York City and Los Angeles, Kinney and associates (2002) reported mean outdoor concentrations of methylene chloride ranging from 1.96 $\mu g/m^3$ (winter) to 1.10 $\mu g/m^3$ (summer), with corresponding indoor concentrations of 6.18 and 1.1 $\mu g/m^3$, and personal air concentrations of 3.8 and 9.3 $\mu g/m^3$, respectively. Those mean concentrations are within the same range as the ones we calculated; however, as for many of the VOCs measured, the distribution of concentrations in both studies was highly skewed.

Outdoor concentrations of chloroform have been reported to vary between 0.3 and 0.7 $\mu g/m^3$ in the total exposure assessment methodology (TEAM)–California study (Wallace 1987; Wallace 1991); 1.57 $\mu g/m^3$ mean (0.86 $\mu g/m^3$ median) in the NHEXAS Region 5 study (US EPA Region 5 [northern Midwest]; Clayton et al 1999); and 0.33 to 33 $\mu g/m^3$ (mean winter to summer) in the TEACH study (Kinney et al 2002); the corresponding values in this study are consistent with those reported from the TEAM and TEACH studies. The indoor concentrations we measured are consistent with levels reported from the NHEXAS Region 5 study. Personal concentrations from our study are consistent with those reported from the TEACH study, and slightly higher than those reported from the TEAM–California and the NHEXAS Region 5 studies.

Carbon tetrachloride mean concentrations in our study were consistently below 1 $\mu g/m^3$ and thus consistent with levels reported from the TEAM–California and TEACH studies; concentrations reported from the TEAM–New Jersey study for this compound were slightly higher (Wallace et al 1985).

For trichloroethylene, mean outdoor concentrations in this study are comparable to those reported from the TEACH study and slightly lower than levels reported from the NHEXAS Region 5 and TEAM–California studies. Mean indoor concentrations from this study are higher than those found in the Air Pollution Exposure Distributions of Adult Urban Populations in Europe (EXPOLIS)–Helsinki study (Edwards et al 2001), but lower than those reported from the TEACH and NHEXAS Region 5 studies. Mean personal concentrations for this VOC in our study are rather similar to indoor and outdoor levels; they are also higher than those reported from the EXPOLIS–Helsinki study, but lower than those from the TEAM–California, TEACH, NHEXAS Region 5, and the German Environmental Survey 1990/92 (GerES II) (Hoffmann et al 2000) studies.

For tetrachloroethylene, mean indoor and outdoor levels we measured are generally lower than those reported from the TEAM–California, TEAM–New Jersey, TEACH, and NHEXAS Region 5 studies; the personal levels are all comparable.

For MTBE, ours is the largest study in which community indoor, outdoor, and personal air measurements have been taken. This compound was found ubiquitously in all three locations and in all the measured microenvironments; levels were typically above MDLs and usually followed the pattern of decreasing values from personal to indoor to outdoor concentrations. Other studies (Vayghani and Weisel 1999; Vainiotalo et al 1999) have found typically higher nonoccupational exposures at gas stations and during refueling. Such exposure measurements, however, are not comparable to the integrated 48-hour measurements obtained in this study.

Mean benzene levels in our study were generally lower than those reported from the TEAM study, probably reflecting the decrease in benzene use since the late 1980s; in our study, indoor and personal concentrations were at the low end or lower than those reported from the TEACH, NHEXAS Region 5, and GerES II studies. It is important to emphasize, however, that the current study excluded households with smokers.

The comparisons described above for benzene also reflect the comparative patterns seen for the levels of ethyl benzene, the xylenes, and toluene. The personal levels of these compounds were highest in the EXPOLIS and GerES II studies, probably reflecting higher source strengths in Europe than in the United States and the inclusion of smokers in those studies. Toluene personal air concentrations were particularly high in GerES II study, with mean values approximately six times higher than those in our study. Overall, GerES II reported the highest personal exposures to aromatic hydrocarbons, which are generally associated with motor vehicle emissions and environmental tobacco smoke.

For *p*-dichlorobenzene, mean outdoor concentrations in this study are similar to those found in the other studies referenced above. However, both the indoor and personal mean concentrations (although not the medians) were appreciably higher than those reported in other studies except the TEACH study. This may reflect comparatively high indoor and automobile interior use of solid deodorants among some sectors of the population.

For *d*-limonene, mean personal concentrations measured in our study are strikingly similar to those measured across European (EXPOLIS, GerES II) and US homes (NHEXAS Region 5, TEACH); this probably reflects the extensive and uniform use of this chemical in consumer products.

α-Pinene levels are also comparable across the US studies (NHEXAS Region 5, TEACH, RIOPA) but lower than levels reported in the European studies (EXPOLIS, GerES II).

Carbonyl Compounds

We primarily used the passive method for measuring carbonyl compounds, and most of our analyses were based on those data. Ten compounds were measured and analyzed using this method (see Tables 13–16).

Those ten compounds and six additional ones were also measured using the active method. Among those 16 compounds, acrolein and crotonaldehyde had very low recoveries; another unknown peak in the chromatograph interfered with sample analysis of hexaldehyde; and *o*-toluladehyde, *m*- & *p*-toluladehydes, and dimethylbenzaldhyde were detectable in only about 10% of the samples. Therefore, these compounds were dropped from the data analysis (see the section Quality Control Measures and Data Correction), leaving 10 compounds analyzed that had been measured with the active method. In addition, the small sample size and unbalanced data structure of the data collected using the active method resulted in very limited analyses of these data.

All carbonyl compounds measured with the passive method, except acrolein and crotonaldehyde, were detected in more than 90% of indoor, outdoor, and personal samples (see Tables 13–16). The incomplete randomized block mixed model was used to analyze the eight carbonyls with detection frequencies greater than 90% in indoor, outdoor, and personal samples. The results indicated that all eight compounds had significantly higher indoor and adult personal concentrations than outdoor concentrations ($P < 0.05$).

A comparison of indoor and personal concentrations indicated that adult personal concentrations of acetone were significantly higher than indoor concentrations for some homes, with largely scattered personal–indoor relations across all the homes (Appendix F). The comparison also showed that for benzaldehyde more homes had higher personal adult concentrations than indoor concentrations (Figure 10). The data pattern for glyoxal was similar to that for benzaldehyde (Figure F.2).

In general the relation patterns were similar for the same carbonyl compounds measured using two different methods, although the proportion of the data points above MDLs was smaller for the active method data than for the passive method data.

In-vehicle measurements were made only using the active method. Of the in-vehicle samples, only formaldehyde and acetaldehyde were detected in more than 60% of the samples (Table 17) owing to the high MDLs of the short-duration in-vehicle measurements. In-vehicle concentrations of formaldehyde and acetaldehyde had ranges wider than those for other indoor, outdoor, and personal concentrations; and their median in-vehicle concentrations were

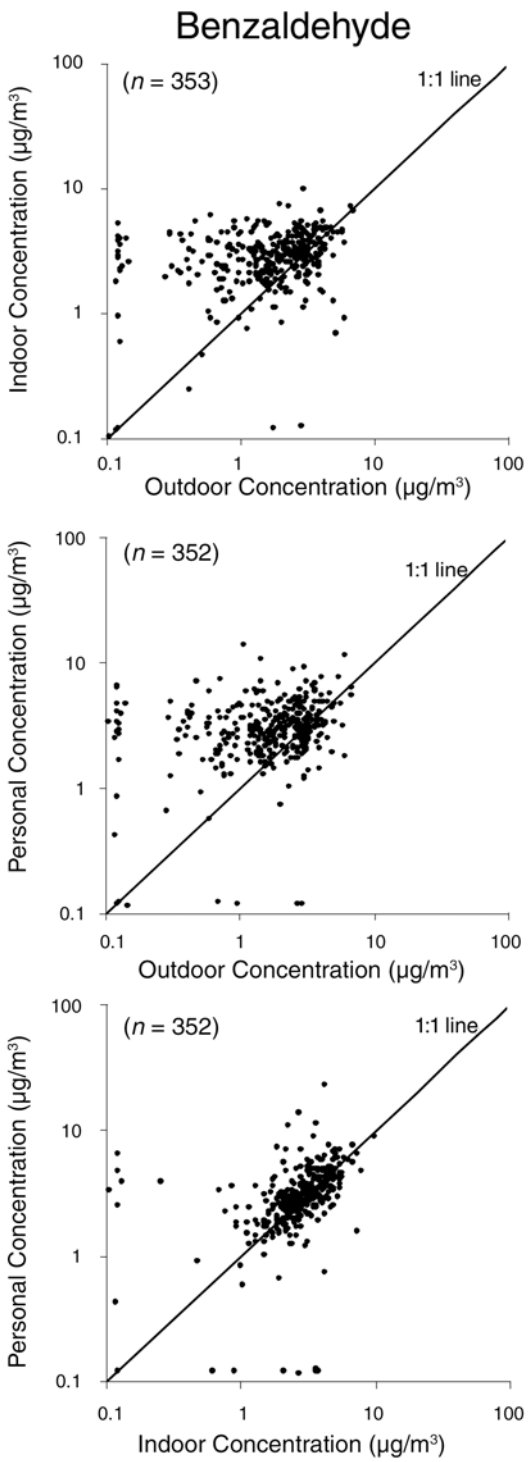

Figure 10. Benzaldehyde air concentrations for indoor–outdoor, personal–outdoor, and personal–indoor comparisons. Data derived only from samples collected using the passive method; shown on logarithmic scales.

Table 17. Descriptive Summary of In-Vehicle Concentrations of Carbonyls (µg/m³)[a]

Carbonyl	Mean	SD	Median	Percentile				Percentage Above LOD
				1st	5th	95th	99th	
Formaldehyde	39.7	104	20.2	1.39	6.07	121	950	92.2
Acetaldehyde	25.2	135	5.92	0.73	1.91	43.2	1240	63.5
Acetone	20.9	103	4.08	0.74	1.16	45.0	941	22.6
Propionaldehyde	6.11	31.6	1.71	0.36	0.44	12.0	291	42.6
Benzaldehyde	19.0	124	2.26	0.41	0.67	29.0	1130	2.6
Glyoxal	3.03	10.6	1.01	0.17	0.23	9.45	84.8	28.7
Methylglyoxal	2.86	9.39	1.43	0.01	0.01	8.91	85.8	26.1
Butyraldehyde	21.0	129	2.50	0.02	0.15	43.2	1180	33.9
Isovaleraldehyde	6.86	31.1	1.29	0.07	0.10	17.9	282	20.9
Valeraldehyde	23.3	149	2.53	0.09	0.41	47.2	1360	38.3

[a] In-vehicle carbonyl concentrations were measured using the active method only. $n = 115$ for each analysis.

higher than the median outdoor concentrations, perhaps owing to the infiltration of on-road emissions or to the outgassing of materials inside the cabin's interior.

The vast majority of formaldehyde concentrations, except for those near the MDL, were nearly parallel to the indoor concentration axis on the indoor–outdoor plot, nearly parallel to the personal concentration axis on the personal–outdoor plot, and fell along the 1:1 line on the personal–indoor plot, suggesting that indoor sources were the dominant contributors to measured personal concentrations (Figure 11). Similar patterns were observed for acetaldehyde (for most of the homes), butyraldehyde, isovaleraldehyde, valeraldehyde, and hexaldehyde (Appendix F).

For acrolein, crotonaldehyde, and propionaldehyde, a large portion of the data points were randomly distributed about the 1:1 line and another portion of the data showed highly elevated indoor air concentrations compared with outdoor air concentrations. This pattern suggests that indoor emissions of these compounds were insignificant in the majority of the homes, but significant in some of the homes.

A comparison of outdoor concentrations, based on the Kruskal-Wallis test, showed significant intercity differences for all the carbonyl compounds other than formaldehyde (Appendix E, Table E.1) based on the results from the passive method. Median outdoor concentrations for the majority of the compounds were lowest in Elizabeth. Elizabeth also has the lowest average outdoor temperature and the least amount of solar radiation, on an annual basis, and thus the lowest level of photochemical reaction activities that could lead to the formation of certain carbonyl compounds.

Indoor concentrations showed significant intercity differences for all the carbonyls except formaldehyde and benzaldehyde; overall, the highest median indoor concentrations were observed in Houston (Table E.2).

Adult personal concentrations showed significant intercity differences for all carbonyl compounds except formaldehyde and benzaldehyde, with highest median concentrations observed in Houston (Table E.3). In contrast, significant intercity differences in child personal concentrations were observed only for acetaldehyde, acetone, hexaldehyde, and methylglyoxal.

More than 40% of the in-vehicle concentrations of all carbonyl compounds except formaldehyde were below the MDLs (data obtained by the active method only). Therefore, intercity comparisons were done only for formaldehyde concentrations, which differed significantly among the three cities; the highest median concentration was observed in Los Angeles (Table E.5). This observation, consistent with the observation that the highest median outdoor formaldehyde concentration was in Los Angeles,

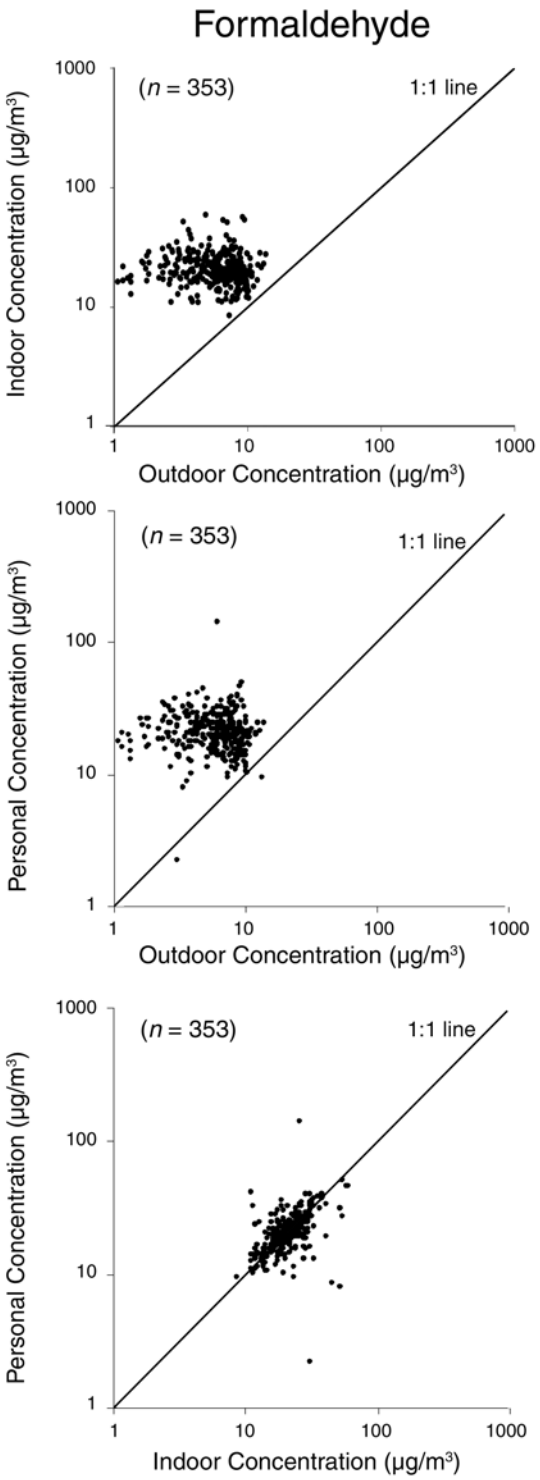

Figure 11. Formaldehyde air concentrations for indoor–outdoor, personal–outdoor, and personal–indoor comparisons. Data derived only from samples collected using the passive method; shown on logarithmic scales.

indicates the importance of mobile emissions for outdoor and on-road concentrations of formaldehyde.

Significant paired adult–child differences were observed for only a few carbonyl compounds: propionaldehyde and hexaldehyde in Los Angeles homes (Table E.6), glyoxal in Elizabeth homes (Table E.7), and propionaldehyde in Houston homes (Table E.8). In all these cases median concentrations were higher for the child subjects. The reasons underlying these observations need to be further evaluated.

In Los Angeles, seasonal differences in outdoor concentrations were significantly different for 7 of 10 measured carbonyl compounds (Table E.9). Among these seven, three (propionaldehyde, hexaldehyde, and methylglyoxal) had the highest median concentrations measured in spring and summer.

In Elizabeth, acrolein and crotonaldehyde were not examined for seasonal differences because of low detection rates. The other eight carbonyl compounds showed significant seasonal differences in outdoor concentrations (Table E.10). Some had the highest median concentrations in summer, some in spring, and some in winter.

In Houston, with the exception of acrolein and crotonaldehyde, the carbonyl compounds showed significant seasonal differences in outdoor concentration (Table E.11), but no clear seasonal trends were found.

These seasonal differences may not only reflect varied meteorologic conditions among the three cities, but also seasonal variations in primary and secondary sources of carbonyl compounds. Comparing microenvironments, more carbonyl compounds showed significant seasonal differences in outdoor concentrations than among indoor and adult personal concentrations. However, comparing locations, more compounds had significant seasonal differences in indoor and adult personal concentrations in Elizabeth than in Los Angeles and Houston (Tables E.12–E.17).

Unlike the results for most of the VOCs, home type had significant effects on indoor concentrations of very few carbonyl compounds (four in Los Angeles, one in Elizabeth, and one in Houston) (Tables E.18–E.20). In Los Angeles, the highest indoor median concentrations of acetaldehyde and hexaldehyde were found in multiple-family homes or apartments, and the highest median concentration of benzaldehyde was found in mobile homes or trailers. In Elizabeth, the median concentration of hexaldehyde was lowest in single-family homes. In Houston the median concentration of crotonaldehyde was highest in single-family homes.

This study's findings on carbonyl compounds are consistent with those from a number of studies conducted in recent years. Indoor measurements have often targeted formaldehyde because it is usually present at higher concentrations indoors than outdoors. In addition to formaldehyde and acetaldehyde, other aldehydes have been measured occasionally. For example, a study conducted in Xalapa, Mexico, measured formaldehyde, acetaldehyde, acetone, propionaldehyde, and butyraldehyde concentrations in a variety of indoor environments including residences, offices, and museums (Baez et al 2003). Clarisse and associates (2003) reported indoor concentrations of several aldehydes in typical Paris dwellings. Acrolein, crotonaldehyde, glyoxal, and methylglyoxal have been measured in the outdoor air of Los Angeles (Grosjean and Grosjean 1996); propionaldehyde, 2-furaldehyde, butyraldehyde, benzaldehyde, *iso*-valeraldehyde, valeraldehyde, and hexaldehyde have been measured in both indoor and outdoor air of New Jersey and Boston homes (Zhang et al 1994a; Reiss et al 1995). Jurvelin and colleagues (2001, 2003) examined the relations of indoor, outdoor, and personal exposure to carbonyl compounds for residents of 15 homes in Helsinki, Finland, as a part of the EXPOLIS study.

When comparing previously reported indoor and outdoor concentrations with each other, we found that almost all the measured carbonyl compounds were present at higher concentrations indoors than outdoors (Table 18). Because of this result and because people spend a large portion of time indoors, exposure to carbonyl compounds would be expected to be derived mainly from indoor concentrations. This expectation is supported by the personal–indoor concentration relations observed for most of the carbonyl compounds measured in this study.

$PM_{2.5}$ Mass

Study-wide median indoor, outdoor, personal–adult, and personal–child $PM_{2.5}$ mass concentrations were 14.4 $\mu g/m^3$, 15.5 $\mu g/m^3$, 30.6 $\mu g/m^3$, and 39.2 $\mu g/m^3$, respectively. Personal $PM_{2.5}$ concentrations were significantly higher than indoor concentrations and outdoor concentrations as determined by one-way ANOVA and the Sheffe test ($P < 0.001$) performed on the log-transformed data.

No statistically significant differences were found between indoor and outdoor concentrations. Personal concentrations were also more variable than both indoor and outdoor concentrations according to the Levene test ($\alpha = 0.05$) for the overall study data and for data segregated by city, with the exception of the Los Angeles outdoor concentrations. Indoor concentrations for the Houston homes were more variable than outdoor concentrations, whereas no significant difference in the variance of indoor and outdoor concentrations was found for the Los Angeles homes and the Elizabeth homes (Levene test; $\alpha = 0.05$).

Table 18. Multistudy Comparisons of Carbonyl Concentrations (μg/m^3) Measured in Residential Settings[a]

	Suburban NJ (Zhang et al 1994a)			Boston MA (Reiss et al 1995)						Helsinki, Finland (Jurvelin et al 2003)			Los Angeles, Elizabeth, and Houston (This Study)		
	6 Homes Summer (2:30–7:30 PM) Data from 1992			9 Homes Summer (24-hr average) Data from 1993			4 Homes Winter (24-hr average) Data from 1993–1994			15 Homes Summer (48-hr average) Data from 1996–1997			~100 Homes per City Year-Round (48-hr average) Data from 1999–2001		
Carbonyl	Indoor	Outdoor	I/O	Indoor	Outdoor	I/O	Indoor	Outdoor	I/O	Indoor	Outdoor	I/O	Indoor	Outdoor	I/O
Formaldehyde	66.6	15.3	4.3	21.96	6.04	3.63	19.7	3.18	6.2	40.8	3.19	26	13.6	3.80	3.6
Acetaldehyde	5.30	4.74	1.1	20.59	6.29	3.27	9.16	1.98	4.6	18.2	2.70	11	11.5	2.69	4.3
Acetone	—	—	—	10.96	5.26	2.08	14.9	4.16	3.6	74.4	4.03	78	16.4	3.43	4.8
Propionaldehyde	2.72	3.01	0.9	2.05	1.45	1.41	4.50	1.96	2.3	2.16	0.33	10	2.60	0.50	5.2
Benzaldehyde	1.64	1.08	1.5	2.8	2.13	1.31	2.21	0.35	6.4	2.82	1.47	3.1	3.03	1.82	1.7
Hexaldehyde	5.22	2.45	2.1	4.87	2.11	2.37	8.98	0.65	13.8	16.4	3.76	6.7	5.71	0.12	46.7
Butyraldehyde	1.94	1.47	1.3	1.35	0.7	1.93	1.65	0.38	4.3	2.00	1.06	2.2	1.82	0.76	2.4
Isovaleraldehyde	1.44	1.30	1.1	0.97	0.24	4.04	0.42	0.04	12.0	—	—	—	1.23	0.14	8.8
Valeraldehyde	3.19	2.81	1.1	2.17	0.71	3.05	3.86	0.32	12.2	4.57	1.27	3.4	3.37	2.81	1.2

[a] Dash indicates no data.

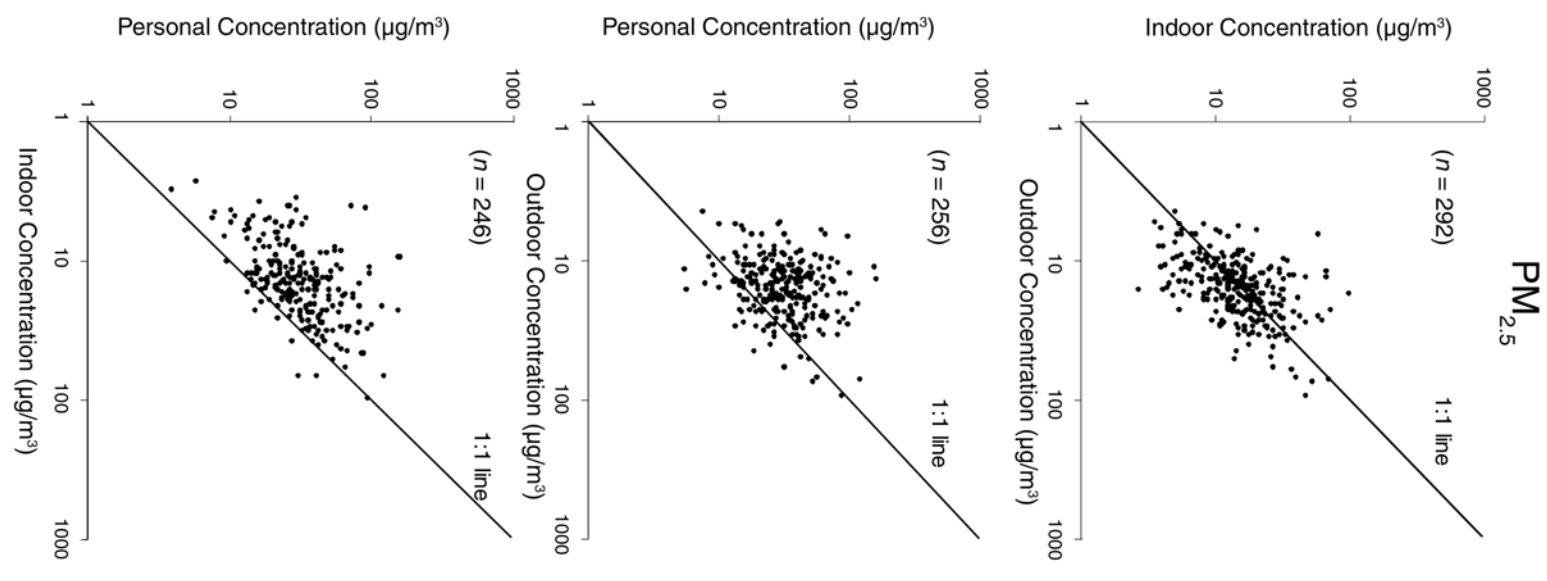

Figure 12. $PM_{2.5}$ air concentrations for indoor–outdoor, personal–outdoor, and personal–indoor comparisons. Shown on logarithmic scales.

Analysis using the incomplete randomized block mixed model showed, however, that personal concentrations were higher than indoor and outdoor concentrations for all three cities, and outdoor concentrations were higher than indoor concentrations for the Elizabeth and Los Angeles homes, as well as for the overall pooled data set. The same conclusions were obtained when only the first-visit sample from each home was used in the analysis, confirming that the conclusions were not artifacts of within-home correlation.

Figure 12 shows scatter plots of indoor, outdoor, and personal $PM_{2.5}$ concentrations, and Table 19 provides CVs. Pooled indoor, outdoor, and personal $PM_{2.5}$ mass concentrations were only poorly to moderately correlated (r^2 = 1% to 19% for Elizabeth and Houston; r^2 = 21% to 44% for Los Angeles), reflecting daily and home-to-home variations in indoor source strength, AER, and personal activities. As one would expect, correlations between indoor and outdoor concentrations were much stronger for homes in which the ratio of indoor to outdoor mass concentrations was less than 1 (r^2 = 43% to 80%; indoor-to-outdoor concentrations [I/O] less than 1 were found in 54% to 71% of homes by city). The higher correlations presumably resulted from low indoor source strengths or high AER values (or both) in these homes. Correlations of outdoor or indoor $PM_{2.5}$ concentrations with personal $PM_{2.5}$ concentrations were not much stronger for these homes than for all homes.

The mean outdoor $PM_{2.5}$ concentration for the Los Angeles homes (19.2 μg/m^3) in this study was similar to that measured in the 1999 wintertime $PM_{2.5}$ exposure studies in Fresno, California (20.5 μg/m^3; Vette et al 2001). The mean (19.2 μg/m^3) and median (16.1 μg/m^3) values from our study were much smaller than $PM_{2.5}$ mass concentrations in the 1990 Particle Total Exposure Assessment Methodology (PTEAM) study in Riverside, California (mean, 48.9 μg/m^3 for daytime and 50.5 μg/m^3 for nighttime; median, 35.5 μg/m^3 for daytime and 35.0 μg/m^3 for nighttime) (Clayton et al 1993). Also the outdoor $PM_{2.5}$ concentrations for the Los Angeles homes in this study were less variable than the PTEAM samples (RIOPA SD 13.3 μg/m^3, and CV 69%; PTEAM daytime SD 37.6 μg/m^3, and CV 77%; PTEAM nighttime SD 40.3 μg/m^3, and CV 80%; Clayton et al 1993). The differences between our findings and those from the PTEAM study are likely to have resulted from differences in sampling strategies, study locations, and study years. Riverside is at the eastern edge of the Los Angeles Basin, a receptor of aged pollution transported across the basin. In contrast, the homes in our study were located in the western half of the basin, closer to primary pollution sources. Air quality in the Los Angeles Basin has also improved over the last 10 years, although declines in PM concentrations are more modest than declines in ozone concentrations.

The annual average central monitor $PM_{2.5}$ mass concentration was 16.4 μg/m^3 in Elizabeth, New Jersey, for July 1997 to June 1998 (Chuersuwan and Turpin 2000), lower than the mean 48-hour outdoor concentration of 20.4 μg/m^3 that we measured. The difference between the central monitoring result and our result may arise from different sampling (year-round versus intermittent), sampling (rooftop versus yard), and strategies.

Comparisons can also be drawn with studies conducted in other locations. Lachenmyer and Hidy (2000) reported 48-hour average $PM_{2.5}$ mass concentrations in Birmingham, Alabama, of 12.2 μg/m^3 for outdoor and 11.2 μg/m^3 for indoor concentrations in winter 1998, and 26.5 μg/m^3 for outdoor and 16.1 μg/m^3 for indoor concentrations in summer 1997. Median indoor, outdoor, and personal concentrations in the Toronto exposure study were 15.4, 13.2, and 18.7 μg/m^3, respectively (Clayton et al 1999). Median concentrations in the EXPOLIS study (Helsinki, Finland; 1996–1998; Koistinen et al 2001) were 11.7 μg/m^3 for indoor, 7.3 μg/m^3 for outdoor, and 21.6 μg/m^3 for personal concentrations. The EXPOLIS study included smokers,

Table 19. Coefficients of Determination (r^2) for $PM_{2.5}$ Concentrations

Group	Homes[a]	Indoor vs Outdoor	Personal vs Indoor	Personal vs Outdoor
All cities	All	0.18	0.20	0.05
	I/O < 1	0.71	0.15	0.10
Los Angeles	All	0.44	0.27	0.21
	I/O < 1	0.80	0.40	0.33
Elizabeth	All	0.12	0.19	0.05
	I/O < 1	0.66	0.16	0.09
Houston	All	0.06	0.13	0.007
	I/O < 1	0.43	0.03	0.02

[a] I/O indicates r^2 for homes in which the ratio of indoor to outdoor $PM_{2.5}$ is less than 1.

however, whereas our study did not. According to our Activity Questionnaire, exposure to passive tobacco smoke did occur, but rarely. One subject that reported passive tobacco smoke exposure had a personal $PM_{2.5}$ mass concentration of 96.5 μg/m^3, which was higher than the 95th percentile of overall personal measurement data; another subject had a concentration of 66.0 μg/m^3, which was higher than the 90th percentile of the overall data set. Other activities such as cooking could also cause higher personal concentrations. Future data analyses will address the contribution of personal activities to $PM_{2.5}$ personal exposure levels.

In the present study, personal/outdoor and personal/indoor concentration ratios for $PM_{2.5}$ were higher than the same ratios reported by others. In our study, personal concentrations were measured using the PEM, whereas indoor and outdoor concentrations were measured using the Harvard impactor. A systematic difference was observed in the comparison of the PEM and Harvard impactor (see the section Quality Control Measures and Data Correction). However, in any analysis that involved both Harvard impactor data and PEM data, we compared the unadjusted personal concentration results with the results obtained when the PEM data were adjusted according to the results from regression of the PEM data against the collocated Harvard impactor data. The main findings were consistent regardless of whether or not the personal concentration data were adjusted, suggesting that the systematic difference between the PEM method and Harvard impactor method may not be the main cause of the higher personal–indoor or personal–outdoor ratios observed in our study compared to those reported by others.

As shown under Home Characteristics and Demographic Information, the subjects in our study were predominantly women, mainly housewives, who spent more time inside their residences than the general population. Many of these subjects might perform cooking and cleaning activities more frequently or for longer periods per day than the general population. These activities are known sources of personal PM exposure and could lead to higher personal concentrations than indoor concentrations of $PM_{2.5}$.

CONTRIBUTIONS OF INDOOR AND OUTDOOR SOURCES TO INDOOR CONCENTRATIONS

Table 20 presents the distributions of indoor source strengths (S in μg/hr) and the fractional outdoor contributions to indoor concentrations $\{aP\,[1/(a + k)]\,[C_{Out}/C_{In}]\}$, both estimated using equation 3 on a home-by-home basis. For $PM_{2.5}$, the results were estimated using a variety of values for penetration through the building envelope (P) and decay rate due to deposition and reaction (k), that is, sensitivity analyses. The indoor-to-outdoor concentration ratios (I/O, or C_{In}/C_{Out}) were calculated from the measured home-specific concentration data. (Only paired indoor–outdoor data, including censored data, were used in the calculations.)

Some estimates of indoor source strength, as shown in Table 20, appeared to be negative values; and some estimates of fractional outdoor contributions to indoor concentrations appeared to be greater than 1. If all assumptions of equation 3 are met, then the minimum S value should be zero and the maximum fractional outdoor contribution should be unity. Physically implausible values of S and fractional outdoor contributions occurred in some homes with I/O greater than 1. This result reflects possible errors associated with measurements of indoor and outdoor concentrations and AERs, or potential errors associated with assumptions for equation 3 (eg, P and k values, steady-state approximation). It is likely that k values may be quite different for some homes with special characteristics (distributions of k values across the homes will be examined in future analyses). P (penetration) values may also vary across homes but are expected to be less variable than k values. In the current analysis, for convenience, we used the same P and k values for each measured species across all the homes.

The funds available allowed only limited sensitivity analyses, and only for the $PM_{2.5}$ data. Similar analyses should be done for the VOCs and carbonyl compounds. In addition, the estimated S values, as well as assumed P and k values, should be validated; this can be done in future analyses using the questionnaire data on home characteristics, for example.

VOCs

When using equation 3 to calculate S and the fractional outdoor contributions to indoor concentrations, we assumed P was 1 and k was 0 for all the VOCs across all homes. This assumed that losses of the measured VOC species, both during outdoor-to-indoor transport (penetration) and within the indoor environment, were negligible. Except for highly reactive compounds (eg, ozone, $P \sim 0.8$), most gases should have P values equal or close to unity. Within-home decay rate (k) was determined by surface deposition and chemical reaction. The surface deposition of gases is mainly driven by their water solubility. The VOCs measured were nonpolar organic gases that have low solubility in water. Therefore, unless they can be removed via chemical reactions or are absorbed into furnishings, they should have k values close to zero.

Previous studies of indoor chemistry have shown that only unsaturated VOCs (ie, styrene, α-pinene, β-pinene, and d-limonene in our VOC list) can react with ozone at a

rate comparable to a typical AER. Hence the assumption that k is 0 is perhaps improper only for these unsaturated VOCs when indoor ozone levels are elevated. For example, k values would be 0.36 hr^{-1} for d-limonene and 0.15 hr^{-1} for α-pinene when the ozone concentration is 20 ppb, according to reaction rate calculations (Fan et al 2003). Even for these more reactive VOCs, assuming k is 0 should not result in a large error in estimating S and the fractional outdoor contributions, as demonstrated in the estimates for $PM_{2.5}$ under different P and k values (Table 20) (Indoor chemistry, however, may play an important role in generating secondary indoor pollutants, which could include carbonyl compounds and PM. The database we gathered, coupled with ozone concentrations measured in the neighborhoods of the homes, can be used at some future time to estimate contributions of indoor ozone–VOC reactions to measured indoor concentrations of certain carbonyl compounds and $PM_{2.5}$ [Fan et al, 2003].)

The results showed large home-to-home variations in both source strength (S) values and the fractional outdoor contributions for all the VOCs. Overall, the calculated source strengths support the interpretations from the scatter plots (Figure F.1). Reviewing the median values, we found that chloroform, α-pinene, β-pinene, and d-limonene had high indoor source strengths, low fractional outdoor contributions, and high I/O values. Chloroform, a byproduct of water chlorination, can be readily released into indoor air through volatilization from tap water during showering, bathing, washing, and cooking. The three terpenes (α-pinene, β-pinene, and d-limonene) are used commonly in terpene-based solvents and can be found in air fresheners and as fragrances in consumer products. Although p-dichlorobenzene had a median S value much smaller than that of d-limonene, it had a relatively high 75th percentile value and the highest 95th percentile value, which reflects strong sources of this compound in a few homes. This finding is reasonable because not all the households used mothballs and deodorizers, the dominant sources of p-dichlorobenzene.

At least half of the homes had no indoor sources of MTBE, carbon tetrachloride, and trichloroethylene. These compounds have few known indoor sources. Attached garages and within-home storage of gasoline might account for high levels of MTBE and other gasoline-derived compounds in some homes. (This can be further evaluated using the questionnaire data.)

The median fractional outdoor contributions to indoor concentrations for compounds with dominant indoor or dominant outdoor sources are consistent with what is known about these compounds. For the compounds with dominant indoor sources, the values ranged from 13% for d-limonene to less than 50% for chloroform, α-pinene, β-pinene, and p-dichlorobenzene. For the compounds with dominant outdoor sources (MTBE, carbon tetrachloride, and trichloroethylene), the median outdoor contribution to indoor concentrations was approximately 100%.

Carbonyl Compounds

Carbonyl compounds are more polar and more water-soluble than the VOCs measured. Thus we expected a slightly lower value for penetration through the building envelope (P) and higher indoor decay rate (k). In the current analysis we assumed unity for the P values of all the measured carbonyl compounds ($P = 1$ for nonreactive gases). The surface deposition velocity of formaldehyde measured in a test room has been reported to be 0.005 ± 0.003 cm/sec (Nazaroff and Cass 1986), or 0.36/hr for typical homes with a nominal surface-to-volume ratio of 2/m. Because we could not find previously reported k values for the other carbonyl compounds, we used k equal to 0.36 hr^{-1} for all the carbonyl compounds analyzed. Compared with formaldehyde, the actual k values may be higher for those carbonyl compounds with higher water solubility, or lower for those with lower water solubility. Nevertheless, as judged by the analysis of $PM_{2.5}$ under a variety of P and k values (see Table 20), we expected those differences to insignificantly change the estimated source strengths and fractional outdoor contributions.

Formaldehyde and acetaldehyde had large S values even at the 25th percentiles. This is consistent with high I/O values for these two compounds. Except for acrolein and crotonaldehyde, with I/O medians of approximately 1, all the other carbonyl compounds had median S values above 170 µg/hr and median I/O values between 1 and 2 based on the results from the passive sampling method (Table 20). Fractional outdoor contributions to measured indoor concentrations, at median level, ranged from 19% for formaldehyde to 63% for acrolein. Source strengths of the same compounds derived from the active method data and the passive method data showed similar patterns for all the carbonyls except benzaldehyde, although the samples collected using the two different methods were mainly from different homes.

These results support our general understanding of carbonyl sources. Carbonyl compounds in the air are produced from both primary and secondary sources. Primary sources emit carbonyl compounds directly into the air, whereas secondary sources are those in which carbonyl compounds are formed through atmospheric chemical reactions. Emissions from the incomplete combustion of fuels and waste materials contain carbonyl compounds

Table 20. Distributions of Estimated Source Strengths and Fractional Outdoor Contributions to Measured Indoor Concentrations Along with I/O Ratios

	Indoor Source Strength (µg/hr)[a]				Fractional Outdoor Contribution to Indoor Concentration[a]				I/O Concentration Ratio			
Species	*n*	25th Percentile	Median	75th Percentile	*n*	25th Percentile	Median	75th Percentile	*n*	25th Percentile	Median	75th Percentile
VOCs[b]	486								486			
MTBE		< 0.0[c]	0.0	320	504	0.65	1.0	>1.0[c]		0.75	1.00	1.53
Chloroform		23	110	250	464	0.09	0.2	0.57		1.74	4.87	10.8
Carbon tetrachloride		< 0.0[c]	< 0.0[c]	6.7	498	0.89	1.0	>1.0[c]		0.83	0.98	1.12
Benzene		< 0.0[c]	28	190	517	0.52	0.9	1.0		0.96	1.12	1.94
Trichloroethylene		0.0	0.0	1500	377	0.30	1.0	1.0		1.00	1.00	3.35
Toluene		0.0	470	57	515	0.48	0.65	1.0		1.00	1.54	2.10
Ethyl benzene		0.0	51	180	514	0.37	0.74	1.0		1.00	1.35	2.73
m- & p-Xylenes		<0.0[c]	110	390	517	0.40	0.71	>1.0[c]		0.95	1.41	2.47
o-Xylene		<0.0[c]	42	140	517	0.41	0.74	>1.0[c]		0.93	1.34	2.43
Styrene		0.0	21	81	434	0.26	0.58	1.0		1.00	1.72	3.87
α-Pinene		0.0	160	520	513	0.08	0.31	1.0		1.00	3.19	12.4
β-Pinene		0.0	140	470	439	0.05	0.20	1.0		1.00	5.06	18.4
d-Limonene		300	1300	3300	476	0.04	0.13	0.45		2.20	7.44	26.1
p-Dichlorobenzene		0.0	46	840	503	0.09	0.50	1.0		1.00	2.01	10.8
Carbonyls (passive method)	353				353				353			
Formaldehyde		2488	3890	6101		0.11	0.19	0.29		2.32	3.24	5.17
Acetaldehyde		2017	3182	4995		0.11	0.21	0.39		1.80	3.22	5.69
Acetone		430	920	2252		0.15	0.36	0.68		1.06	1.70	4.06
Acrolein		< 0.0[c]	35.5	220		0.19	0.63	1.15		0.50	1.01	3.04
Propionaldehyde		31.4	179	358		0.25	0.50	0.89		0.80	1.23	2.19
Crotonaldehyde		< 0.0[c]	28.3	133		0.16	0.61	1.15		0.52	1.01	3.90
Benzaldehyde		161	303	611		0.23	0.43	0.69		1.02	1.46	2.51
Hexaldehyde		344	596	988		0.17	0.30	0.52		1.38	1.93	3.23
Glyoxal		152	309	536		0.29	0.44	0.61		1.07	1.44	2.08
Methylglyoxal		153	314	517		0.29	0.45	0.65		1.01	1.34	2.09

[a] Estimates were calculated using equation 3 on a home-by-home basis. *n* is the total samples for each compound within a group, unless otherwise noted. NA indicates not available.

[b] Methylene chloride was not included because a high proportion of samples were below the MDLs.

[c] These physically implausible values are generated by errors associated with measurements of indoor and outdoor concentrations and AERs or errors associated with assumptions of *P* and *k* values and steady-state approximations.

[d] Paper A: Özkaynak et al 1996. Paper B: Lachenmyer and Hidy 2000.

[e] RIOPA A: *P* and *k* values were derived from the whole data set. RIOPA B: *P* and *k* values were derived from subset of homes with I/O ratios < 1 and with no indoor PM sources reported on questionnaires.

Table 20 (*continued*). Distributions of Estimated Source Strengths and Fractional Outdoor Contributions to Measured Indoor Concentrations Along with I/O Ratios

	Indoor Source Strength (μg/hr)[a]				Fractional Outdoor Contribution to Indoor Concentration[a]				I/O Concentration Ratio			
Species	*n*	25th Percentile	Median	75th Percentile	*n*	25th Percentile	Median	75th Percentile	*n*	25th Percentile	Median	75th Percentile
Carbonyls (active method)	78				78				78			
Formaldehyde		2790	4599	6864		0.06	0.10	0.14		4.83	6.69	11.2
Acetaldehyde		953	1687	2421		0.09	0.21	0.39		1.72	2.89	5.71
Propionaldehyde		163	251	437		0.16	0.29	0.44		1.44	2.24	3.72
Benzaldehyde		< 0.0[c]	92.5	275		0.24	0.66	1.64		0.41	0.89	2.73
Glyoxal		45.7	107	175		0.23	0.38	0.61		1.07	1.97	2.91
Methylglyoxal		55.8	184	369		0.09	0.22	0.54		1.15	3.20	7.18
Butyraldehyde		65.2	136	271		0.24	0.46	0.62		1.04	1.53	2.42
Isovaleraldehyde		41.2	130	216		0.13	0.35	0.61		1.00	2.10	4.65
Valeraldehyde		124	266	464		0.12	0.24	0.46		1.34	2.59	5.13
$PM_{2.5}$	262				262							
Paper A[d]: $P = 1$, $k = 0.39$		< 0.0[c]	640	2100		0.51	0.79	1.0		NA	NA	NA
Paper B[d] $P = 1$, $k = 0.6$		360	1100	3000		0.39	0.65	0.83		NA	NA	NA
RIOPA A[e]: $P = 0.91$, $k = 0.79$		940	2000	3900		0.46	0.56	0.93	268	0.63	0.90	1.20
RIOPA B[e]: $P = 0.78$, $k = 0.40$		210	1300	2200		0.33	0.61	0.74	165	0.54	0.70	0.87

[a] Estimates were calculated using equation 3 on a home-by-home basis. *n* is the total samples for each compound within a group, unless otherwise noted. NA indicates not available.

[b] Methylene chloride was not included because a high proportion of samples were below detection limits.

[c] These physically implausible values are generated by errors associated with measurements of indoor and outdoor concentrations and AERs or errors associated with assumptions of *P* and *k* values and steady-state approximations.

[d] Paper A: Özkaynak et al 1996. Paper B: Lachenmyer and Hidy 2000.

[e] RIOPA A: *P* and *k* values were derived from the whole data set. RIOPA B: *P* and *k* values were derived from subset of homes with I/O ratios < 1 and with no indoor PM sources reported on questionnaires.

and hydrocarbons that can be oxidized to form carbonyl compounds in the atmosphere, resulting in elevated carbonyl concentrations in polluted urban ambient air (Baugh et al 1987; Calvert and Madronich 1987; Liu et al 1999).

On-road emissions of carbonyl compounds from vehicle exhausts are one of the major sources of ambient carbonyls in urban areas (Grosjean et al 2001; Kean et al 2001; Destaillats et al 2002). There is some possibility that changes in fuel composition, specifically replacing high-octane fuels with substitutes such as methanol and ethanol, will increase carbonyl emissions (Tanner et al 1988; Williams et al 1990; US National Research Council 1991; Ho and Winer 1998; Magnusson et al 2002; Zervas et al 2002). (Future analysis should determine whether there are differences in fuel composition and control devices among the three cities and, if so, whether this is a reason for higher outdoor and in-vehicle concentrations of formaldehyde that we observed in Los Angeles.)

Some carbonyl compounds are released into occupational and residential indoor air settings from building materials, furniture, and consumer products (Bravo et al 1990; Muramatsu et al 1990). Direct emissions from household products and materials have been identified as major sources of some aldehydes and ketones in indoor air. Particleboard and medium-density fiberboard, popular building and furnishing materials in the United States, are reported to be possibly important contributors of indoor carbonyl compounds. In one study (Baumann et al 2000), emissions of small straight-chain aldehydes, such as hexaldehyde, valeraldehyde, octanal, and nonanal, were found to generally exceed emissions of other compounds and accounted for more than 50% of total VOC emissions from wood materials.

Surface coatings constitute another source of carbonyl compound emissions. Results from chamber studies of different types of furniture coatings indicate that aliphatic and aromatic aldehydes (eg, benzaldehyde) are among the most prevalent compounds emitted by these coating materials (Salthammer 1997). The use of alkyd paint indoors can produce odorous aldehydes during the drying process (Chang and Guo 1998; Hancock et al 1989). When alkyd paints are applied to a surface at room temperature, the unsaturated fatty acids, used as additives in paints, react with atmospheric oxygen to produce hydroperoxides. These hydroperoxides can generate free radicals and at the same time can be decomposed by fragmentation resulting in by-products, mainly aldehydes and a small quantity of ketones and alcohols (Fortmann et al 1998; Afshari et al 2003). It is also well known that reactions of ozone with indoor alkenes (eg, *d*-limonene and α-pinene) produce carbonyl compounds (Zhang et al 1994a,b; Weschler et al 1992, Weschler 2000; Reiss et al 1995). Researchers found that ozone can react with the compounds emitted from carpets to form formaldehyde, acetaldehyde, and other C_5- to C_{10}-aldehydes (Weschler et al 1992; Morrison and Nazaroff 2002). Therefore, home renovation, such as installing new carpets and applying new paints, may significantly increase indoor concentrations of carbonyl compounds. Formaldehyde, acetaldehyde, valeraldehyde, hexaldehyde, other less volatile aldehydes, and terpenes (precursors of certain aldehydes in the presence with ozone) were found to be the predominant air pollutants in newly built homes (Hodgson et al 2002).

Certain consumer products, such as cleaning agents, air fresheners, nail polish remover, deodorants, perfumes, glues, and highlighter pens, can be sources or precursors of carbonyl compounds (Collins and Mitchell 1975; Weschler 2000). Tobacco smoke is a major indoor source of carbonyls. It has been reported that burning one cigarette can generate 1310 µg of formaldehyde and 2150 µg of acetaldehyde (Daisey et al 1998). Butyraldehyde, propionaldehyde, acrolein, and crotonaldehyde have also been found in tobacco smoke (Poirier et al 2002). (The current study recruited only participants who did not smoke and lived in households with no resident smokers. However, passive exposures to environmental tobacco smoke were likely to affect some personal measurements. In the future, the RIOPA questionnaire and diary data can be used to assess the possible impact of carbonyl concentrations from environmental tobacco smoke on personal exposure.) Other activities that may elevate indoor levels of carbonyl compounds include burning candles and incense as well as high-temperature cooking (Lin and Tang 1994; Lau et al 1997; Lin and Liou 2000; Schauer et al 2002). One suggested future analysis of the data gathered in this study is an in-depth examination of the impact of indoor sources and activities on indoor and personal concentrations of carbonyl compounds.

$PM_{2.5}$ Mass

The distributions of indoor source strengths and fractional outdoor concentrations to measured indoor concentrations are shown in Table 20 for four sets of values for penetration through the building envelope (P) and decay rate due to deposition and reaction (k). The first two sets of P and k values were taken from the literature and represent the higher and lower ends of published data. The other two sets were derived from our data set, as explained below.

Table 21 presents the results of parameter estimation from the nonlinear regression (NLIN in SAS, Cary NC) of equation 3 using measured indoor and outdoor concentrations and AERs to find a single estimate of P and k for all homes together and for Los Angeles, Elizabeth, and Houston homes separately. Typically in solving this kind of an equation, P is unconstrained; this provides a "reasonable"

estimation of *P*. However if that estimation is not physically meaningful (eg, greater than 100%), then *P* must be constrained between 0 and 1 to arrive at a valid solution. Therefore, we tested *P* and *k* in both conditions to ensure that our results were both valid and robust.

P values estimated in this study (0.73 to 1.0) are comparable to estimates from other studies of $PM_{2.5}$, which reported *P* values of 0.84 to 1.0 (Koutrakis et al 1992; Thatcher and Layton 1995; Özkaynak et al 1996; Lachenmyer and Hidy 2000; Long et al 2001; Winkle and Scheffe 2001).

The indoor particle decay rate (*k*) is a function of many factors including home surface-to-volume ratio, housing structure, near-surface air flows, turbulence, and particle size distribution. The *k* value obtained from the nonlinear regression procedure was identified as an "average" value for all homes in our study.

The use of a single value of *k* for all homes introduces an uncertainty in estimating *S* and the outdoor contribution. The degree of uncertainty depends on the relative magnitudes of *k* and the AER. When the AER is very low, *k* is a very important determinant of the outdoor contribution. At an AER of 1 hr^{-1}, changing *k* from 0.79 hr^{-1} to 0.4 hr^{-1} with no change in *P* changes the median outdoor contribution from roughly 55% to nearly 70%. The value of *k* was fairly sensitive to the inclusion or removal of homes with indoor sources (which generated outliers in the regression). However, the distribution of outdoor contribution to indoor concentration was very stable and consistent for these reasonable but different analytic approaches (ie, approaches yielding $P = 0.78$ to 1.0 and $k = 0.39$ hr^{-1} to 0.79 hr^{-1}).

The estimated average *k* value for $PM_{2.5}$ for the homes in our study was 0.79 hr^{-1} (95% CI = 0.18, 1.4). Özkaynak and associates (1996) estimated *k* as 0.39 hr^{-1} (95% CI = 0.22, 0.55) for the PTEAM study. Lachenmyer and Hidy (2000) estimated *k* to be 0.6 hr^{-1} with a range of 2.0 hr^{-1}. Abt and colleagues (2000) and Vette and coworkers (2001) estimated *k* values as a function of particle size using real-time particle monitors. In Fresno, *k* was estimated to be 0.5 hr^{-1} for particles 0.1 μm in diameter and 3.5 hr^{-1} for particles 2.5 μm in diameter (Vette et al 2001); in Boston, the lowest *k* was 0.7 hr^{-1} for particles 0.4 to 0.5 μm in diameter, and the highest was 1.2 hr^{-1} for particles 2 to 3 μm in diameter (Abt et al 2000).

Outdoor contributions to indoor concentrations calculated with the RCS model were based on the statistical inferences of regression analysis. Indoor–outdoor associations could be affected by extreme values (outliers), such as a high indoor concentration on a day with a low outdoor concentration, or vice versa. Therefore we identified outliers and evaluated their influence on infiltration factor or attenuation factor in the RCS model. A value was considered an outlier if the absolute residual of that data point, calculated with the Student *t* test, was larger than 3. In evaluating the outdoor $PM_{2.5}$ contributions to indoor concentrations, seven outliers were identified. After removing those outliers, the infiltration factor $[aP/(a + k)]$ changed by 0.01. After outliers were removed the outdoor contributions to indoor concentrations increased by 2%.

To examine the compatibility of results from the mass balance and RCS models, we applied each model to estimate the distribution of outdoor and indoor contributions to indoor $PM_{2.5}$ concentrations for all homes (268 total) with valid indoor and outdoor $PM_{2.5}$ and AER data.

With the RCS model, the infiltration factor (α; the slope of the regression of indoor and outdoor $PM_{2.5}$ concentrations) was 0.46 for those 268 homes. The distribution of outdoor contributions to indoor concentrations was calculated by multiplying the infiltration factor by the measured outdoor $PM_{2.5}$ mass concentration (*B*) for each home. The difference between the measured indoor $PM_{2.5}$ mass concentration and the calculated outdoor contribution was the indoor contribution.

Table 21. Summary of Parameter Estimation by NLIN Regression of Equation 3 Using $PM_{2.5}$ Data

Group	*N*	Boundary Condition[a]	*P* (Penetration)	95% CI of *P*	*k* (Decay Rate; hr^{-1})	95% CI of *k* (hr^{-1})
All cities	268	Yes	0.91	0.71 , 1.12	0.79	0.18 , 1.41
		No	0.91	0.71 , 1.12	0.79	0.18 , 1.41
Los Angeles	112	Yes	1.00	1.00 , 1.00	0.90	0.53 , 1.28
		No	1.04	0.75 , 1.33	0.98	0.28 , 1.69
Elizabeth	80	Yes	0.73	0.42 , 1.05	0.46	−0.44 , 1.36
		No	0.73	0.42 , 1.05	0.46	−0.44 , 1.36
Houston	76	Yes	1.00	1.00 , 1.00	0.99	−1.38 , 3.35
		No	1.35	0.46 , 2.23	1.18	−1.57 , 3.92

[a] "Yes" means penetration (*P*) was estimated with boundary condition $P \in [0,1]$. "No" means no boundary conditions constrained the estimation of *P*.

With the mass balance model, we calculated the values of the infiltration factor [$\alpha = aP/(a + k)$] for each of the 268 homes using the measured AER values for each home, and P and k. Estimates of k were derived (as previously described in the section Mass Balance Analysis) by fitting the mass balance model to the measured quantities of AERs and indoor and outdoor concentrations. The values of the infiltration factor calculated from the mass balance model were approximately normally distributed (by Kolmogorov-Smirnov test; $\alpha = 0.05$ and $P > 0.15$) with a mean of 0.46 and a SD of 0.16, which is in excellent agreement with the fixed RCS model infiltration factor of 0.46. Again, the difference between the measured indoor $PM_{2.5}$ concentration and the calculated outdoor contribution was the indoor contribution.

Figure 13 shows the cumulative lognormal distributions of indoor contributions and outdoor contributions to indoor concentrations. Two curves in Figure 13 reflect results from the mass balance model (variable-slope infiltration factor α) and RCS model (fixed-slope infiltration factor α). Good agreement was found between these two approaches, particularly for distribution means. The difference between distribution means from those two models was less than 1 μg/m^3, for both outdoor and indoor contributions to indoor $PM_{2.5}$ concentrations.

The RCS model is not designed to predict the indoor and outdoor contributions for individual homes; nevertheless, we found that the CV for the outdoor contribution to indoor concentrations for the two models was 26% when results for 268 homes were compared on a home-by-home basis. The CV for the indoor contribution to indoor concentrations was 24%. Results were highly correlated, with a coefficient of determination (r^2) greater than 75%, for both outdoor and indoor contributions to indoor concentrations. Figure 14 shows the results of the paired data comparison. In the RCS model, a single fixed infiltration

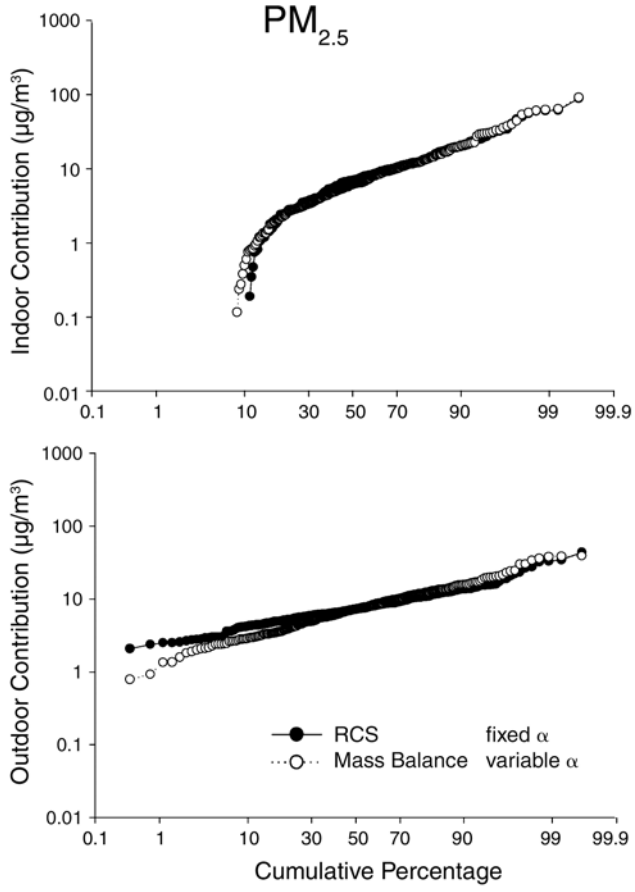

Figure 13. Contributions to indoor concentrations of $PM_{2.5}$ from indoor (top) **and outdoor** (bottom) **sources calculated using the mass balance model with measured AERs (variable-slope α) and the RCS model (fixed-slope α).** Shown on logarithmic scales.

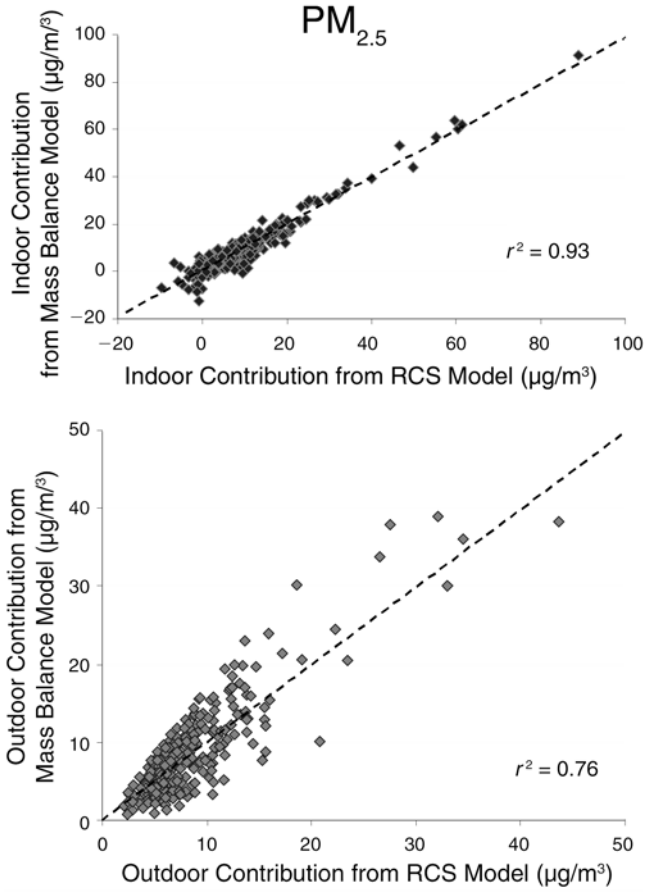

Figure 14. Comparison of mass balance model and RCS model for determining indoor and outdoor source contributions to indoor concentrations of $PM_{2.5}$.

factor was applied to all homes. However, this quantity was affected by AER, particle decay rate, and penetration coefficient, all factors that vary from home to home. This method comparison illustrates the degree of uncertainty introduced when indoor and outdoor contributions are estimated without an AER measurement, which was perhaps the most variable parameter in this study. The home-to-home variability in particle penetration and decay (P and k) values was not considered, which increases the uncertainty somewhat. Nevertheless, the 26% uncertainty in the outdoor contribution to indoor concentrations is not excessive.

SUMMARY AND RECOMMENDATIONS

This study successfully collected and analyzed air toxic concentrations, AER data, and questionnaire responses related to personal and household activities for approximately 100 households in three distinct cities. The measured pollutants included a variety of air toxics and other species relevant to health issues, classified into three generic categories: VOCs, carbonyl compounds, and $PM_{2.5}$. This represents the first comprehensive collection of data on carbonyl compounds in indoor, outdoor, and personal air, as well as inside vehicle cabins; furthermore, data were collected throughout the year in each city. An important feature was that we measured concentrations in homes with a wide range of AERs so the data can be used to develop and evaluate exposure models.

The distributions of measurements for VOCs, formaldehyde, acetaldehyde, and $PM_{2.5}$ air concentrations and the AERs were generally consistent with values reported previously from other studies. Thus associations or models that are based on this data set and seek to link outdoor sources with indoor air concentrations of air toxics could be relevant to general urban settings. However, the three cities we studied differed substantially in terms of demographics, housing characteristics, climate, and pollutant sources; in addition, our subject base was not representative of the general population. Consequently, the data reported here should be compared with data from other studies with caution.

Using scatter plots and simple statistical tests, we examined relations between indoor and outdoor, personal and outdoor, and personal and indoor concentrations. Many VOCs and carbonyl compounds had dominant indoor sources that mainly determined personal concentrations. For a few compounds, personal concentrations were higher than either indoor or outdoor concentrations, indicating the presence of some sources closely related to personal activities. Some compounds had no significant indoor sources in the majority of the homes, and thus indoor concentrations were mainly determined by outdoor concentrations penetrating these homes. More in-depth analysis of these relations can be done by incorporating detailed questionnaire and time and activity data.

In this stage, simple statistical analyses, mainly of the pooled data, were used to examine differences by city, season, home type, home age, and paired adult–child personal concentrations within the same home. These analyses generated some intriguing results that warrant further investigation in the future. The results from the analyses presented in this report should be considered preliminary and tentative; more in-depth analyses may identify subtle differences and provide explanations for some of the observations reported here.

The simultaneous measurements of indoor and outdoor concentrations and AERs enabled us to use a steady-state mass balance model to estimate indoor source strengths and the relative importance of indoor and outdoor sources to the measured indoor concentrations. Estimated indoor source strengths exhibited large home-to-home variations for VOCs and carbonyl compounds, consistent with the observed indoor–outdoor concentration patterns. We calculated source strengths for many compounds never before analyzed, and derived them from hundreds of homes. This is an important contribution to the literature on air toxics. Further, this data set shows potential for future indoor air quality analysis and exposure modeling that has been previously unavailable. These indoor source estimations agreed with previously published values for $PM_{2.5}$ and with our general understanding of sources of VOCs and carbonyl compounds. The estimation of outdoor contributions to measured indoor concentrations provides insights about the relative importance of outdoor and indoor sources in determining indoor concentrations, the main determinant of personal exposure for most of the measured compounds.

We intentionally selected homes according to their proximity to a variety of outdoor stationary and mobile air toxic sources because one of the original study objectives was to examine the effects of outdoor source proximity on indoor and personal air measurements. However, funds were insufficient to include the necessary meteorologic data and geographic information system (GIS) techniques to estimate proximity. The measured concentrations and collected relevant questionnaire data, when coupled with emission source data for air toxics and meteorologic data, can be used in future analyses to assess the impact of distances between homes and sources on measured indoor, outdoor, and personal concentrations.

ACKNOWLEDGMENTS

We are grateful to all of the participants in the study and we appreciate their continued hospitality. We gratefully acknowledge the hard work of all the students and technicians in the field and laboratories involved in this study. In addition, we thank Dr Charles J Weschler of the University of Medicine and Dentistry of New Jersey for his comments on the mass balance analysis of indoor sources, and Dr Wayne Ott of Stanford University for his review of the RCS model and mass balance model analysis. We are indebted to the guidance of Dr Glen Cass (who passed away after outstanding service to his students), the aerosol science community, and HEI.

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APPENDIX A. Field Sampling Sites

ELIZABETH, NEW JERSEY

Elizabeth is a municipality of 110,000 with a high population density. The population has a mixture of lower- and middle-income residents and a diverse ethnic makeup. Its homes are typical of the northeast: a mixture of older homes (approaching 100 years in age) and newer single-family and multiple-family, detached and semidetached houses, and apartment buildings.

Within Elizabeth and adjacent communities there are a multitude of outdoor air toxics sources. The cities of Linden to the south and Newark to the north (Figure A.1) have a number of industrial complexes and an incinerator. The major metropolitan airport in Newark borders Elizabeth on the north.

Elizabeth has industrial emissions sources within its municipal boundaries: along the eastern section is a shipping area and an industrial complex known as Bayway, which also contains a small number of homes. Numerous small commercial enterprises (Figure A.2), including gasoline stations, dry cleaners, refinishing shops, and small factories, are interspaced on residential streets to serve the local population.

A number of congested local streets and major highways pass through Elizabeth; the New Jersey Turnpike is on the

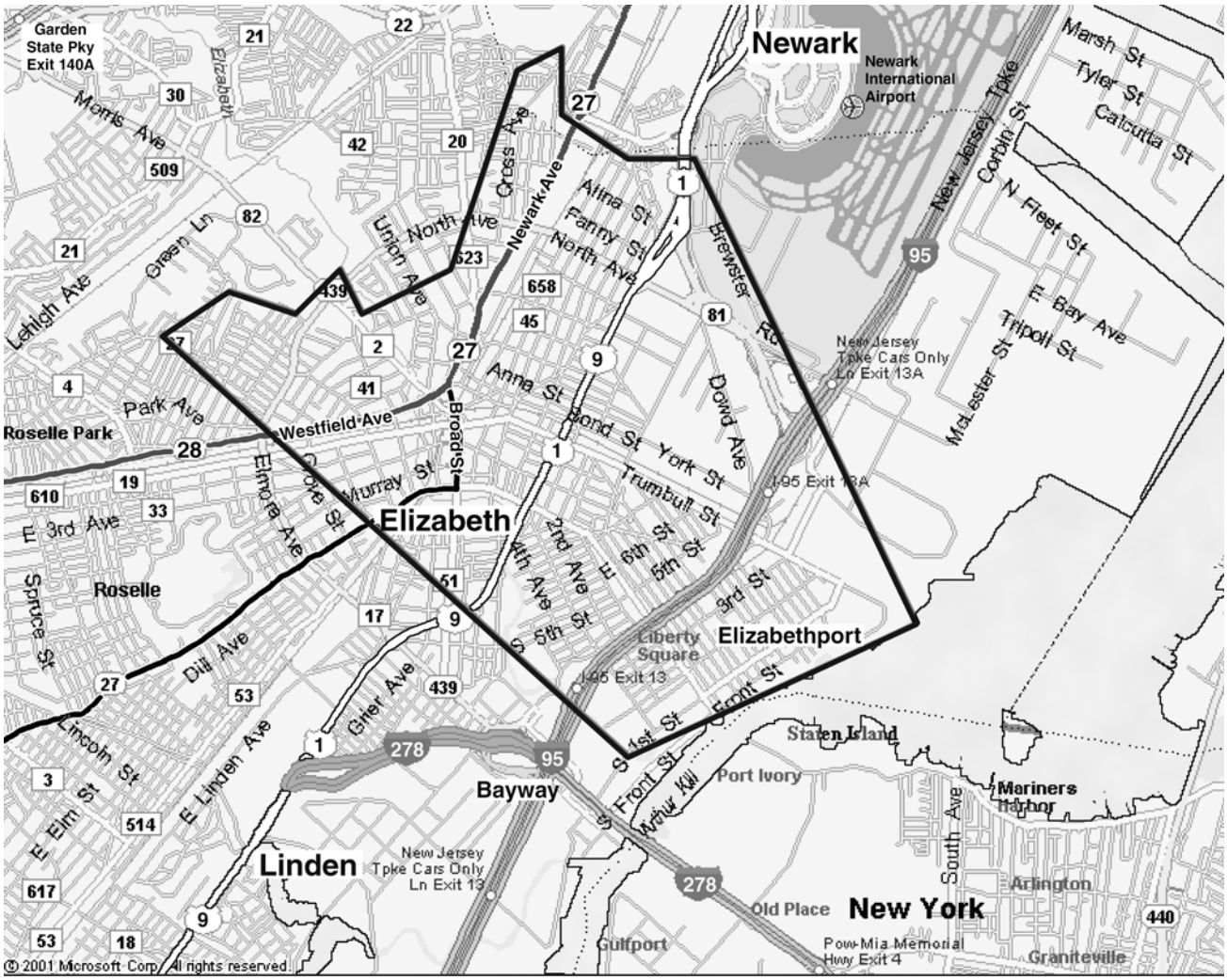

Figure A.1. Location of Elizabeth sampling site in relation to surrounding communities in New Jersey. All participating households were within the outlined perimeter (approximated). (Adapted from a map by Microsoft 2001.)

east (within the study area) and the Garden State Parkway is on the west (outside the study area); these are the two major north-south toll roads in New Jersey (Figure A.1). In addition, US Highway 1/9 bisects Elizabeth; this is a major north-south freeway in New Jersey and carries a large amount of both passenger and truck traffic, including traffic to and from the Newark airport (Figure A.1). Some residences immediately front Route 1/9. In addition, State Routes 27 and 28 pass through the center of the city.

Homes selected for this study included residences on the same block or within one or two blocks (< 200 m) of each of these stationary and traffic sources. The exception is the Newark airport, which has undeveloped land between it and the homes in Elizabeth, although many homes are under the flight paths of planes.

Homes farther from the sources were selected from the western section of Elizabeth (Figure A.2), which has fewer commercial and industrial facilities and lower traffic density, though few, if any, areas in Elizabeth are more than several blocks away from at least one well-used road. Homes were selected throughout the year in all sections of Elizabeth.

HOUSTON, TEXAS

In Houston, we targeted petrochemical facilities as the primary source of pollutants. The Houston metropolitan area has the largest density of petrochemical complexes in

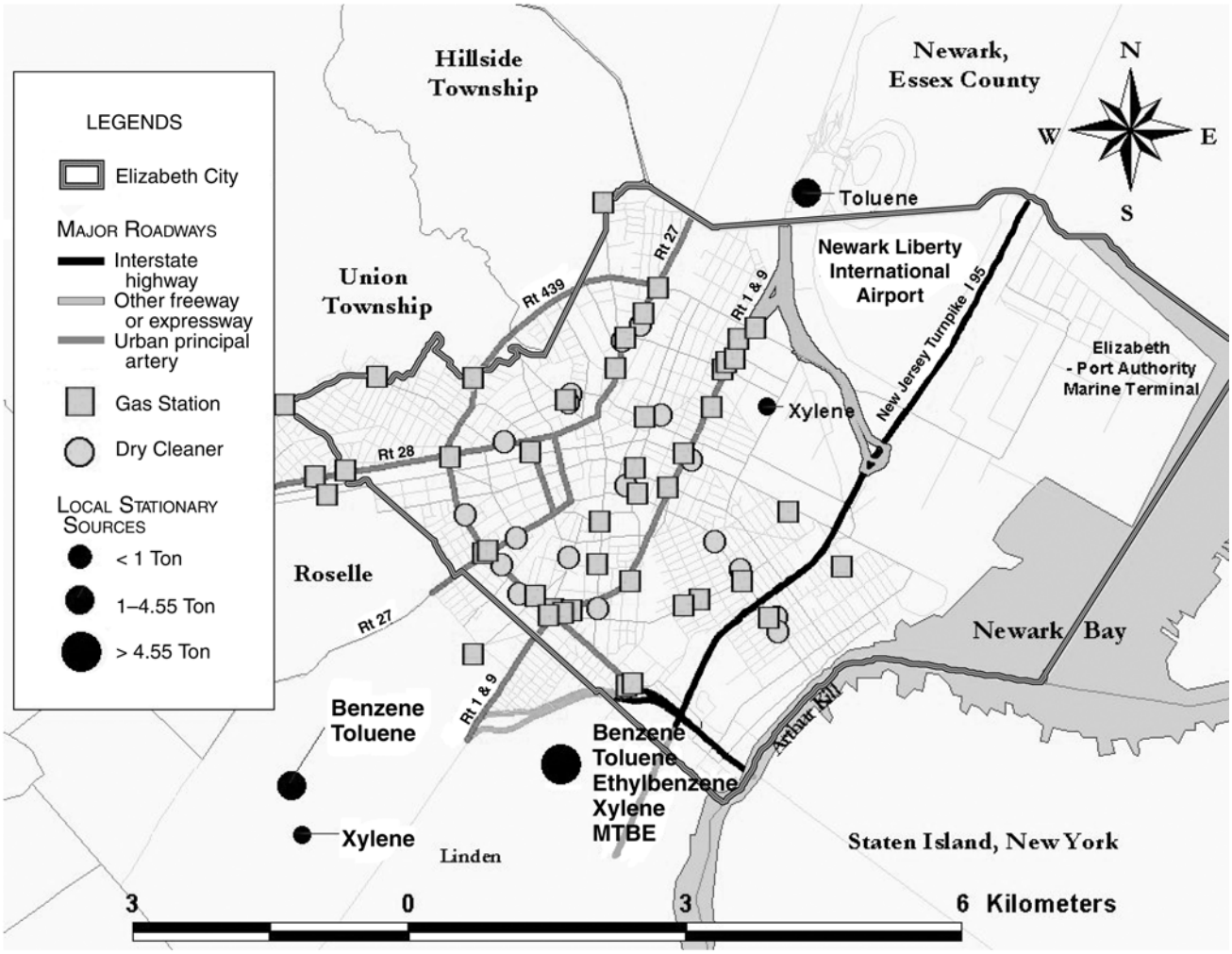

Figure A.2. Elizabeth site showing source locations throughout the sampling area. All participating households were within the outlined perimeter (approximated).

the world. Different units within these facilities may process crude petroleum for fuel production, but they also produce chemicals including plastics and solvents. Thus emissions from these facilities come not only from the type of chemicals expected in fuel production, but also from the raw materials and processes involved in the production of chemicals and plastics.

For homes to potentially participate in the study, we focused on areas around large petrochemical complexes, and then chose households near to and away from the source in each area. The areas from which participants were recruited (Figure A.3) are described below.

Houston Ship Channel

A dense band of petrochemical facilities surround the shipping channel that connects the City of Houston with the Gulf of Mexico. Most residences in the area are older homes built in the 1940s and 1950s, typically of wood and clapboard construction, with crawl spaces beneath them rather than slab foundations. Few have central air conditioning or heating systems. The population in the area consists mainly of young Hispanic families with low to low-middle incomes and retired persons on fixed incomes.

Pasadena

This area is to the immediate north and south of I-225 (called the 225 corridor), which runs east and west from Houston. A string of large petrochemical complexes lie parallel to the north side of I-225 and within easy access of this highway. Most residential and commercial areas are immediately south of I-225. This area is also largely Hispanic, particularly in the zones closer to I-225. The Hispanic population consists of families with young children, in many cases of low- to middle-income levels. The homes are a mix of those built in the 1940s, early 1950s, and later. Most are single-family wood frame homes; the newer ones have brick veneer on the outside walls. In some cases the residents have had central air conditioning and heating systems installed. In areas that are farther away from the 225 corridor, also on the south side, the population consists of primarily white families with children or retired individuals who have higher incomes. The homes are also single-family but newer and larger, than those closer to the highway; they are more typical of the residential middle-class areas in Houston.

Galena Park

This area is north of I-225 and south of I-10. It has similar characteristics to Pasadena with respect to some of the housing, but with pockets of homes with middle- to upper-income residents. These are typically larger homes of two or three floors, built in the 1980s or later, with brick or stone veneers on the outside walls. There are also pockets of trailer homes. The population is a mix of Hispanic and white families.

Channelview

This area is west of Galena Park and south of I-10. It shares some of the characteristics of Galena Park, but has a larger proportion of white residents.

Baytown

This is the location of one of the two largest petrochemical facilities of a major oil company. The area around the petrochemical plant, toward the center of the town district, has low-income residents in single-family houses; many homes in this area are in disrepair or abandoned. Wealthier, middle- or upper-class residential areas are both around the plant and farther away. These homes are of more recent construction, typically have two or three levels, and are very large.

Medical Center

This area does not have any chemical facilities; the major outdoor emission source is local motor vehicle traffic. The area's population is composed of a mix of ethnic backgrounds, as well as a mix of residence types, including single-family houses and large apartment complexes.

LOS ANGELES, CALIFORNIA

The four Los Angeles County sampling areas, each near a major freeway, are shown in Figure A.4. All sampling locations were within 4 km of an air monitoring station operated by the South Coast Air Quality Management District.

West Los Angeles

The majority of sampling was conducted in neighborhoods in West Los Angeles (Figure A.5) because that area has the highest daily vehicle count and is relatively free from the influence of local stationary sources. This location is within 1 km of the 405 and 10 freeways, near the intersection of these two major arteries.

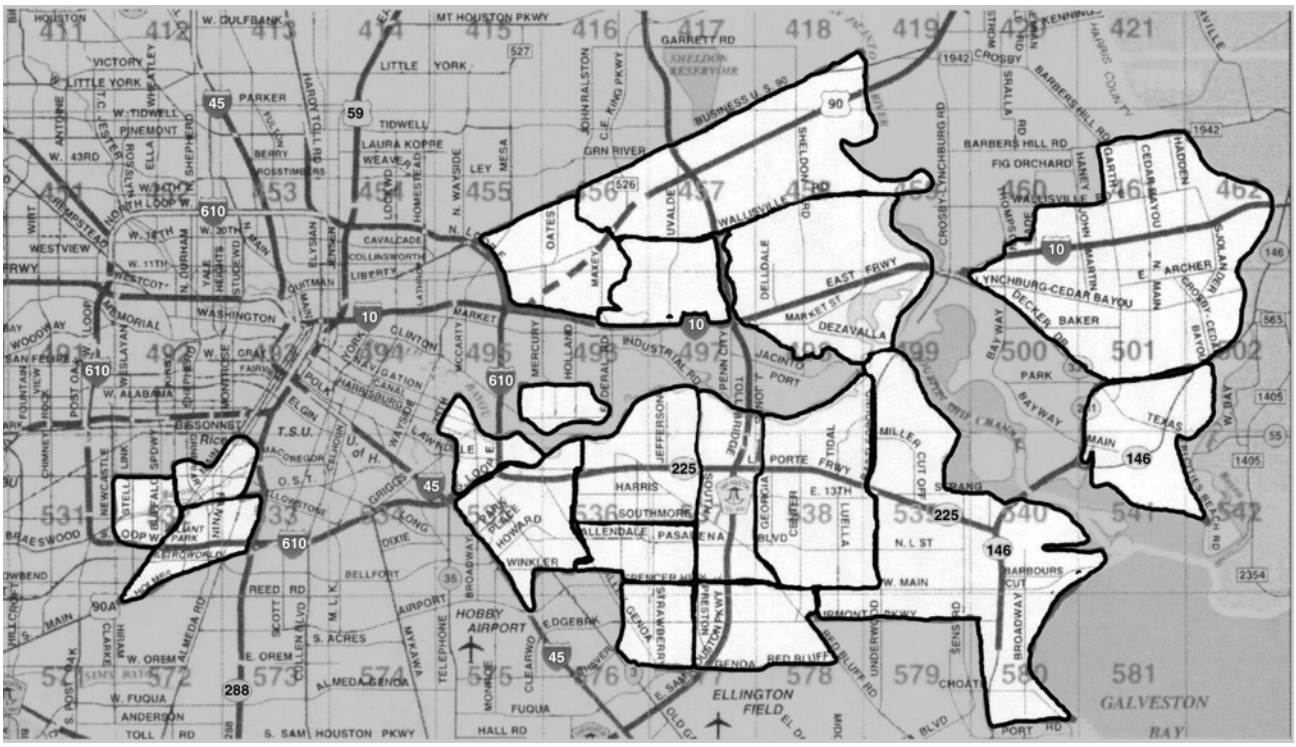

Figure A.3. Houston site showing the general location of the individual sampling communities in relation to the surrounding area. Actual sampling locations, segregated by ZIP code, are outlined.

Pico Rivera

Pico Rivera is in central Los Angeles County and located on the 605 freeway (Figure A.6). Heavy-duty diesel trucks use this freeway to distribute goods from the port of Los Angeles.

Burbank

This community is north of downtown Los Angeles on the 101 freeway (Figure A.7). Traffic volumes are lower than those in West Los Angeles and Pico Rivera.

Santa Clarita

This community is farther north of Burbank on the 101 Freeway (Figure A.8). The air monitoring station is on the edge of the town of Newhall, and our sample was actually drawn from the neighboring community of Santa Clarita. Traffic volumes are lower than those in Burbank, but this area is still on a major north-south artery for the State of California.

Figure A.4. Los Angeles County communities sampled: Santa Clarita, Burbank, West Los Angeles, and Pico Rivera. Major roadways show the areas where participants were recruited.

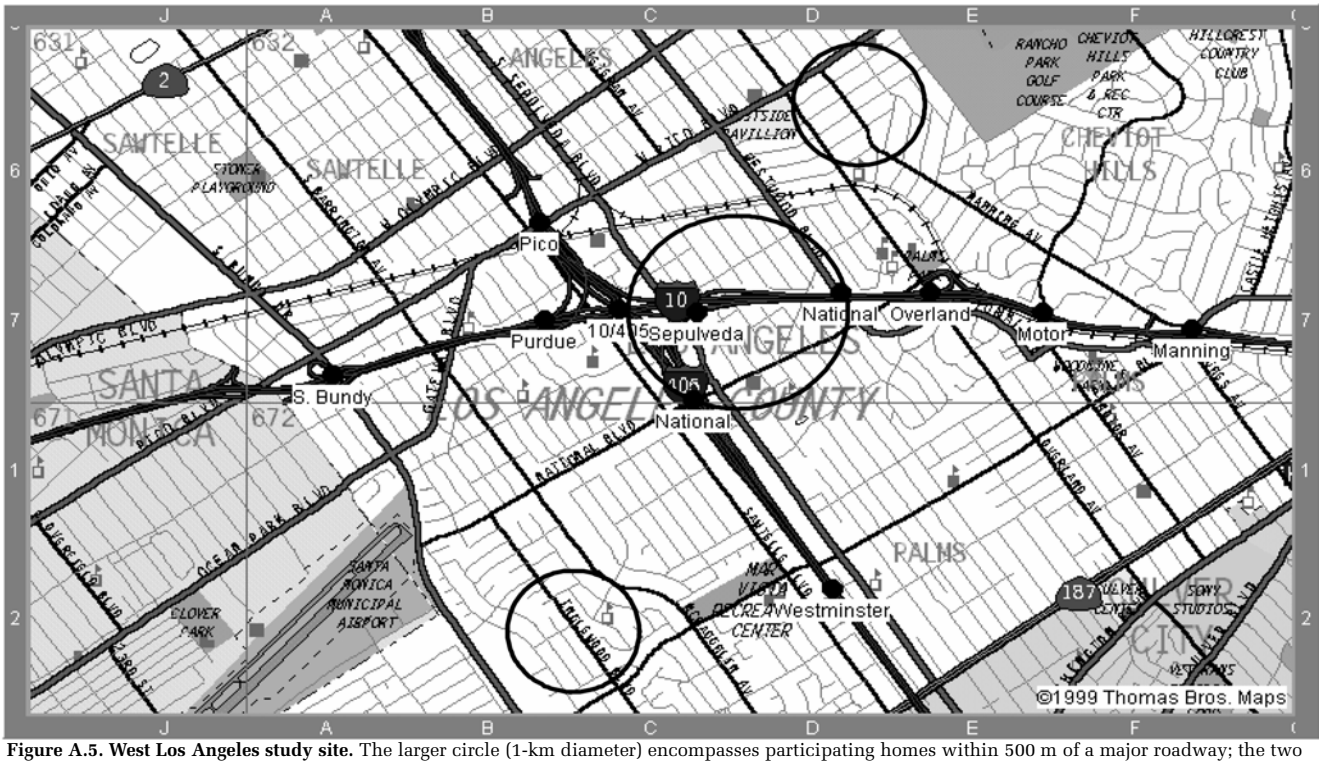

Figure A.5. West Los Angeles study site. The larger circle (1-km diameter) encompasses participating homes within 500 m of a major roadway; the two smaller circles (0.6-km diameter) encompass homes more than 500 m away. The nearest ambient central-site monitoring station is located approximately 1 km north of the northernmost homes sampled and is outside the scale of the figure. (Adapted from Thomas Bros Maps 1999.)

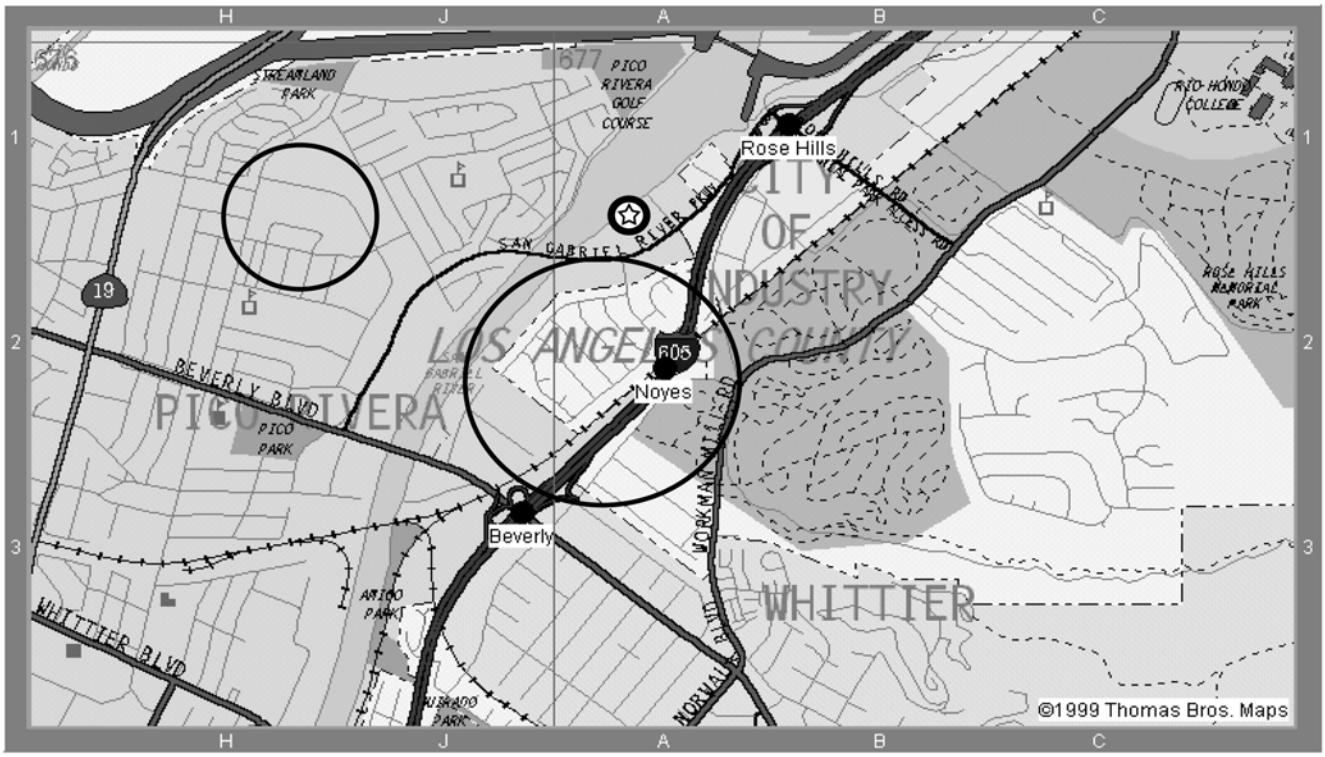

Figure A.6. Pico Rivera study site. The larger circle (1-km diameter) encompasses participating homes within 500 m of a major roadway; the smaller circle (0.6-km diameter) encompasses homes more than 500 m away. ★ indicates the nearest ambient central-site monitoring station. (Adapted from Thomas Bros Maps 1999.)

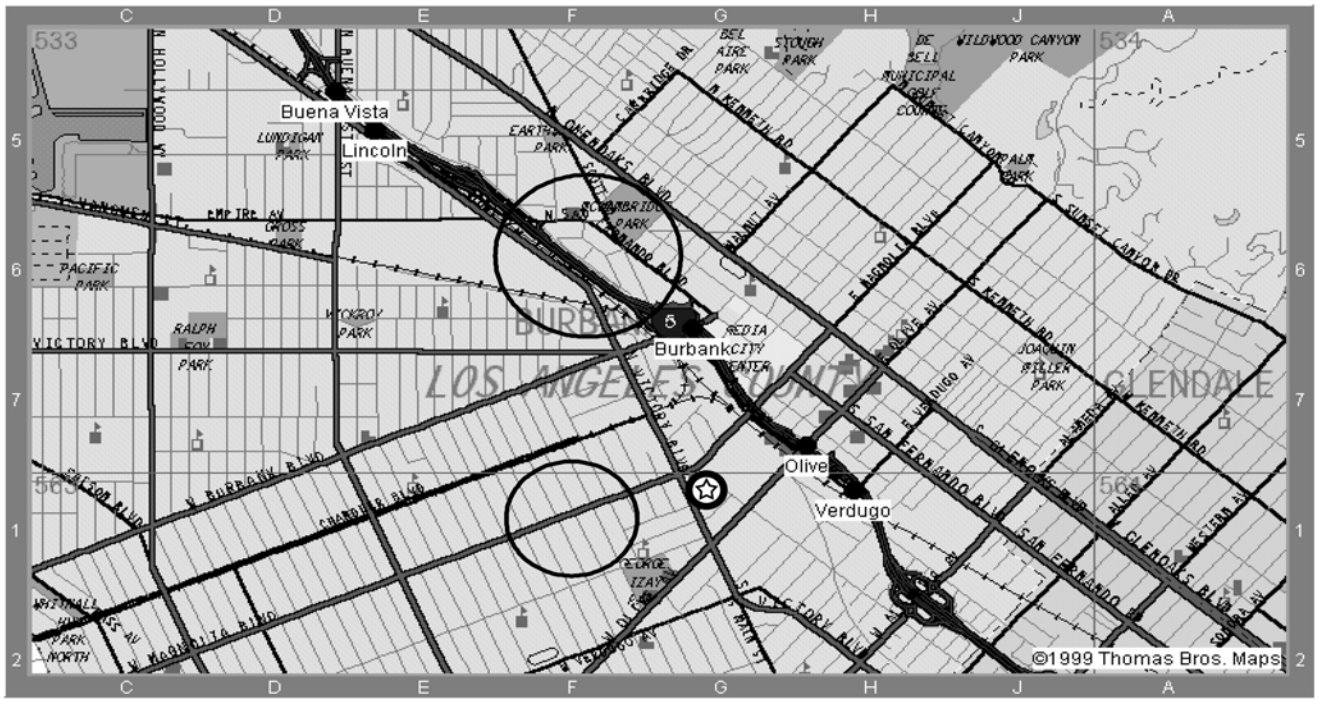

Figure A.7. Burbank study site. The larger circle (1-km diameter) encompasses participating homes within 500 m of a major roadway; the smaller circle (0.6-km diameter) encompasses homes more than 500 m away. ⛤ indicates the nearest ambient central-site monitoring station. (Adapted from Thomas Bros Maps 1999.)

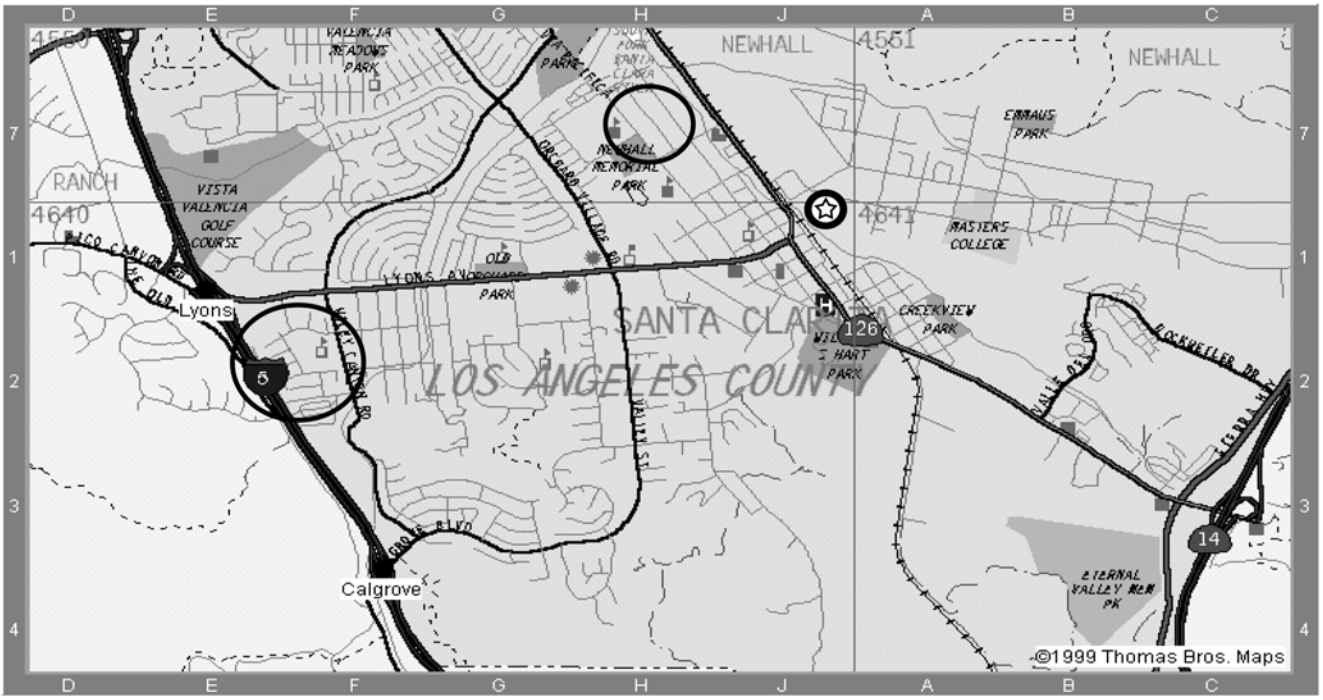

Figure A.8. Santa Clarita study site. The larger circle (1-km diameter) encompasses participating homes within 500 m of a major roadway; the smaller circle (0.6-km diameter) encompass homes more than 500 m away. ⛤ indicates the nearest ambient central-site monitoring station. (Adapted from Thomas Bros Maps 1999.)

APPENDIX B. Pilot Study

SUMMARY

A pilot project was conducted during the first year of this study (1999) to test and optimize each component of the field work and laboratory procedures. In the pilot study, participants were recruited from 10 homes in each of the three locations (Elizabeth, New Jersey; Houston, Texas; and Los Angeles County, California); questionnaires were administered to the participants; and samples were collected from each home. The pilot study was done in two phases. Initially, samples were collected in Elizabeth and Houston. These were evaluated and a number of problems identified concerning pump sampling protocols and the length of the questionnaire. Changes were made and the pilot was then conducted in Los Angeles, after which final adjustments were made before starting the main study of 100 homes in each of the three cities.

During the pilot study, funding received from HEI permitted expanding the original study design funded by the National Urban Air Toxics Research Center as follows: for carbonyl compounds, samples were collected in all households rather than half, and personal and in-vehicle samples were also collected; for $PM_{2.5}$, indoor, outdoor, and personal samples were collected for all households rather than half; and for half of the homes in each city, $PM_{2.5}$ samples were analyzed for organic carbon, elemental carbon, trace metals, sulfur and functional groups, and polycyclic aromatic hydrocarbons (PAHs), in addition to mass. (The results of the $PM_{2.5}$ speciation analyses will be presented in a subsequent report.)

The major findings from the pilot study were

- the proposed sampling and analytic methods were sufficient to measure the target species in the existing environmental concentration range;
- the responses to questionnaires designed for this study and the personal and indoor samples could be collected from the population with the assigned staff; and
- recruitment of participants in each city was possible, but the effort required to recruit a population-based sample in each city was beyond the resources available and not justified in terms of accomplishing the main goal of the study.

The main goal was to evaluate the effect of proximity to outdoor sources on indoor and personal air concentrations and the mechanistic associations between outdoor emissions and exposures. This objective required recruiting participants who lived in residences (1) with no smokers, (2) that reflected a variety of housing characteristics, and (3) with a wide range of AERs. The objective also required oversampling homes close to outdoor sources. A population-based sample of 100 homes in each city that met these criteria would represent a relatively small portion of the population in each city. Consequently, the results about distributions of indoor air concentrations and exposures could not be extrapolated to each urban population directly. However, the findings from the analysis of the relations between indoor, outdoor, and personal air concentrations of air toxics and the resulting models could be extended beyond the population included in the study. Thus the full study was not a random selection of homes, but rather the result of targeted recruitment.

EVALUATION OF VOC METHODS

The performance of two passive VOC monitors, the OVM 3500 and OVM 3520 (3M Company, St Paul MN), was compared for the 18 target VOCs to determine if breakthrough of these compounds would occur on the front pad of the 3520 OVMs. To maximize comparability of the VOC results, the two laboratories analyzing the VOCs (EOHSI and University of Texas) followed identical procedures and used a common standard operating procedure. To further facilitate the implementation of a single method, Masoud Afshar, the senior chemist from University of Texas, went to EOHSI to coordinate the exact procedures of OVM analyses. In addition, both laboratories used the same source of supplies (solvents and standards) and instrumentation. Optimizing the analytic procedure and selecting and screening solvents minimized field and laboratory blank contributions; and, for most compounds, MDLs at concentrations below the level of micrograms per cubic meter were achieved.

EVALUATION OF PUMPS FOR SAMPLING CARBONYL COMPOUNDS AND $PM_{2.5}$

The initial personal sampling pumps selected were those used in NHEXAS. Upon evaluating these pumps, we determined that they would not provide continuous 48-hour sampling for carbonyl compounds and $PM_{2.5}$. During the Elizabeth and Houston pilot projects, we also evaluated Buck GENIE personal sampling pumps (AP Buck). These are designed to operate continuously on a single battery pack for more than 24 hours; they sample PM at a flow rate of 3 to 4 L/min, carbonyl compounds through a Sep-Pak cartridge at a flow rate of 60 to 100 cm^3/min. Pump noise was also considered and adding noise-damping materials was necessary. Reliability of these pumps, which were being operated beyond their original design capabilities, was somewhat limited.

During the Los Angeles pilot project, a BGI personal sampling pump system was also evaluated. The BGI pump system operated consistently at the correct flow rate, and the ease with which the battery could be changed made it practical to use for 48 hours of personal sampling. Initially the study participant had to change the battery after 24 hours of use. Early in the study, BGI provided longer-life batteries, so a single one could be used for the entire monitoring period.

After evaluating whether a single pump could be used for both personal PM and carbonyl compound samples, it was decided that separate pumps would be used for each sample type. The same pump and C_{18} Sep-Pak cartridges coated with DNPH were used to collect indoor and outdoor carbonyl compound samples. Subsequent to the pilot study, a passive carbonyl compound sampler with a DNSH coating was developed and used for most of the main study.

Standard sampling techniques were used and verified during the pilot study for indoor and outdoor $PM_{2.5}$ sample collection at 10 L/min on a 37-mm stretched Teflon filter downstream of a single-jet impactor using Harvard impactors. The personal samples were collected on 25-mm stretched Teflon filters, initially with Buck personal pumps and subsequently with BGI pumps. The MDL for $PM_{2.5}$ area samples (on 37-mm filters) was 27 $\mu g/m^3$ for indoors and 1.0 $\mu g/m^3$ for outdoors based on Elizabeth field blanks (3 SDs of the distribution of field blank concentrations). For personal samples (on 25-mm filters), this corresponds to an MDL of 11 $\mu g/m^3$ for indoors and 1.3 $\mu g/m^3$ for outdoors. Additional quality control checks for weighing reduced the MDLs for 37-mm filters in half for the main study.

EVALUATION OF AER METHOD

The AER measurements were made using the perfluorocarbon trace method originally developed by Brookhaven National Laboratories. No significant problems were observed with the placement of tracer sources or receptors (CATs) in the field, and acceptable field blank levels were obtained.

EVALUATION OF QUESTIONNAIRES

The NHEXAS questionnaires were used as a basis for our questionnaires. By adapting the NHEXAS questions (ie, only using the questions that were relevant to this study's objectives), we developed three distinct questionnaires: the Baseline Questionnaire; the Technician Walk-Through Questionnaire; and the Activity Questionnaire, which included a time diary.

Each questionnaire was translated into Spanish, and a Spanish-speaking field staff member was available for each household where Spanish was the native language. The time required to complete the questionnaires during the Elizabeth and Houston pilot studies was deemed to be too long. The number of questions was reduced, and the modified questionnaires were completed during the Los Angeles County pilot study without problems.

OVERALL PILOT STUDY RESULTS

Several problems were encountered during the first phase of the pilot study in Elizabeth and Houston. The major issues were:

- the flow rate of the sampler for indoor and outdoor carbonyl compounds was not stable;
- the questionnaires were too long to maintain the interest of some participants until completion;
- the functioning of the personal pump was erratic;
- the battery pack was heavy;
- the holder for the personal pump could cause the flow to be restricted when the participant moved, which resulted in a flow rate change; and
- the connection between the pump and battery pack was unreliable, which caused the battery pack to fail to recharge (this was identified midway through the pilot study).

Several modifications were made to alleviate these problems:

- separate pumps were used for the $PM_{2.5}$ and carbonyl compound samplers for indoor and outdoor sampling;
- the questionnaires were shortened and their format was changed to reduce the burden on the subjects and ease subsequent data entry;
- the personal sampling pump was changed;
- the pump holders were redesigned; and
- the VOC air concentration results from OVM 3500 with one charcoal pad and from OVM 3520 with two charcoal pads were not statistically different, so the OVM 3500 could be used; this was preferable because it required one half the time for analysis.

These changes were tested during the second phase of the pilot study in Los Angeles and samples were successfully collected and analyzed. Overall, we were able to recruit people in the desired communities and collect samples from the selected homes. The measurement of the air toxics was practical with the selected sampling and analytical methods.

APPENDIX C. Interlaboratory Comparisons

The two laboratories at EOHSI and at University of Texas analyzed the VOC samples and carbonyl compound samples collected using the active (DNPH-based) method. Interlaboratory comparisons were conducted as part of the quality control protocol.

CARBONYL COMPOUND ANALYSES

The active method samples collected in Elizabeth, Houston, and Los Angeles were extracted in the EOHSI laboratory, the University of Texas laboratory, and a University of California-Los Angeles laboratory, respectively. The Elizabeth and Los Angeles samples were analyzed at the EOHSI laboratory, and the Houston samples were analyzed at the University of Texas laboratory. To ensure consistency between the EOHSI and the University of Texas laboratories, we exchanged extracts of samples, field blanks, and standards for independent analyses at both laboratories. (The two laboratories used similar HPLC-UV techniques.) When the concentration data from the University of Texas laboratory were regressed against the concentration data from the EOHSI laboratory, the nondetectable values were excluded. The regression results (Table C.1) indicated reasonably good interlaboratory agreement for most of the quantified carbonyl compounds; however, systematic differences occurred for the two dicarbonyl compounds, glyoxal and methylglyoxal, as demonstrated by the high values of r^2 with slopes largely different from 1. Investigation into this issue revealed that the University of Texas laboratory had improperly converted concentrations of

Table C.1. Interlaboratory Comparisons on Analysis of DNPH-Carbonyl Derivatives in ACN Solution

Derivative	*n*	Slope	SE of Slope	Intercept	SE of Intercept	r^2
University of Texas vs EOHSI						
Formaldehyde	21	1.171	0.061	0.051	0.032	0.951
Acetaldehyde	19	1.118	0.048	0.028	0.019	0.969
Acetone	19	1.081	0.087	0.068	0.037	0.900
Acrolein	5	0.989	0.030	0.029	0.020	0.997
Propionaldehyde	5	0.956	0.017	0.033	0.011	0.999
Crotonaldehyde	3	1.034	0.015	0.020	0.013	1.000
Benzaldehyde	5	1.006	0.051	0.057	0.033	0.993
Hexaldehyde	4	1.024	0.029	0.143	0.020	0.998
Glyoxal	5	2.080	0.033	−0.012	0.024	0.999
Methylglyoxal	4	2.361	0.099	−0.009	0.070	0.997
Butyraldehyde	4	1.001	0.053	0.036	0.039	0.994
Valeraldehyde	5	0.909	0.032	0.131	0.020	0.996
DGA vs EOHSI[a]						
Formaldehyde	12	1.045	0.013	−0.008	0.020	0.998
Acetaldehyde	12	0.982	0.054	0.013	0.046	0.970
Acetone	12	1.063	0.023	−0.009	0.012	0.995
Propionaldehyde	11	0.862	0.034	0.003	0.006	0.986
Crotonaldehyde	7	0.612	0.359	0.002	0.005	0.368
Benzaldehyde	11	0.969	0.134	−0.017	0.041	0.853
Hexaldehyde	10	0.675	0.124	0.072	0.086	0.786
Glyoxal	10	0.558	0.074	−0.016	0.006	0.875
Methylglyoxal	10	0.408	0.077	0.008	0.009	0.777
Butyraldehyde	10	0.943	0.034	0.006	0.004	0.990
Isovaleraldehyde	9	0.318	0.076	0.009	0.006	0.717
Valeraldehyde	10	0.712	0.040	−0.008	0.009	0.976

[a] DGA is Daniel Grosjean and Associates laboratory.

DNPH derivatives of these two dicarbonyl compounds to concentrations of the parent carbonyl compounds. Therefore, we corrected all the University of Texas glyoxal and methylglyoxal concentrations using the regression equations generated from the interlaboratory comparison results (see Table C.1).

We also conducted an interlaboratory comparison with Daniel Grosjean and Associates on the analysis of DNPH-carbonyl derivatives. In this effort, 12 extracts (three field blanks, two indoor samples, two outdoor samples, two in-vehicle samples, and three personal samples) were prepared by the EOHSI laboratory, and aliquots were analyzed by the two laboratories. The Daniel Grosjean and Associates laboratory used an LC-MS technique, whereas the EOHSI laboratory used a LC-UV method (as described in this report). The concentration data from the Daniel Grosjean and Associates laboratory were regressed against the concentration data from the EOHSI laboratory (values below the limit of detection were excluded). The regression results (see Table C.1) indicated excellent interlaboratory agreement for the following carbonyl compounds: formaldehyde, acetaldehyde, acetone, propionaldehyde, benzaldehyde, and hexaldehyde. The reason for poorer agreement of results for the other measured carbonyl compounds needs to be further investigated.

VOC ANALYSES

The VOC analyses were compared by exchanging passive method samples from the OVM badges between the EOHSI laboratory and the University of Texas laboratory. This resulted in the same extracts being analyzed by both laboratories. The results of the two laboratories were then compared (Table C.2). A regression fit was determined for the paired data (values below the MDL were excluded). The slopes of all but three of the VOCs were between 0.8 and 1.2 with an r^2 greater than 0.95, indicating biases of less than 20% and a high correlation between the results from the two laboratories. The exceptions were methylene chloride, styrene, and toluene. Inconsistent field blank contributions were observed for methylene chloride at EOHSI, which may have resulted in elevated levels of methylene chloride in some extracts after they were analyzed at EOHSI. It is not clear why styrene values indicated a bias of a factor of 2; thus those numbers need to be reviewed carefully before they are used in other analyses. Toluene had a lower correlation than optimal, but the slope was in the acceptable range, suggesting no bias but a lower overall agreement between the two laboratories than for the other compounds.

MDLs were estimated periodically during the study (Table C.3). The estimations were based on procedures suggested by the EPA (Chapter 1 of the Code of Federal Regulations 40 EPA 1996) whereby the critical t value (3.143 for seven replicates) is multiplied by the standard deviation of (a) the response of at least seven standards near the instrumental standard detection, or (b) seven measurements of field blank levels when detectable quantities are present in the field blanks. In practice, we used 3 × SD

Table C.2. Interlaboratory Comparisons on Analyses of VOC Extracts at University of Texas and EOHSI[a]

VOC	Slope	SE of Slope	Intercept	SE of Intercept	r^2
Methylene chloride	0.62	0.07	1.1	0.37	0.77
MBTE	1.08	0.04	0.009	0.041	0.96
Chloroform	1.39	0.08	0.006	0.056	0.92
Carbon tetrachloride	1.16	0.01	0.040	0.007	0.99
Benzene	1.16	0.06	0.092	0.054	0.95
Trichloroethylene	1.14	0.02	0.013	0.017	0.99
Toluene	1.12	0.11	0.19	0.28	0.81
Teterachloroethylene	1.16	0.04	0.072	0.037	0.97
Ethyl benzene	0.829	0.026	0.15	0.03	0.98
m- & *p*-Xylenes	0.985	0.039	0.18	0.09	0.96
o-Xylene	0.942	0.028	0.11	0.02	0.99
Styrene	2.28	0.03	0.084	0.014	0.99
p-Dichlorobenzene	1.06	0.05	0.11	0.05	0.95

[a] n = 26 for all comparisons.

Table C.3. Summary Statistics for 48-Hour MDLs (μg/m^3) of VOCs by Analytical Laboratory

	EOHSI (n = 852)[a]			University of Texas (n = 1011)[a]		
VOC	Mean	Minimum	Maximum	Mean	Minimum	Maximum
Methylene chloride	2.13	0.88	3.18	0.29	0.07	1.07
MTBE	0.68	0.32	2.35	0.38	0.17	1.12
Chloroform	0.42	0.10	0.63	0.28	0.15	0.50
Carbon tetrachloride	0.27	0.11	1.33	0.34	0.14	0.56
Benzene	1.13	0.64	1.34	0.54	0.15	0.86
Trichloroethylene	0.44	0.11	0.88	0.24	0.08	0.63
Toluene	6.73	2.22	10.7	7.12	0.90	15.0
Tetrachloroethylene	0.42	0.12	0.56	0.22	0.01	0.59
Ethyl benzene	0.74	0.21	0.65	0.22	0.11	0.68
m- & *p*-Xylenes	1.39	0.69	1.78	0.65	0.16	2.51
o-Xylene	0.85	0.38	2.16	0.29	0.09	0.81
Styrene	0.84	0.15	1.33	0.34	0.07	1.59
β-Pinene	2.04	0.71	2.92	0.28	0.08	1.52
d-Limonene	1.01	0.28	2.12	2.09	0.44	7.20
α-Pinene	1.27	0.53	3.36	0.74	0.15	3.38
p-Dichlorobenzene	0.91	0.28	1.14	0.43	0.18	0.75

[a] n is the number of blanks analyzed throughout the study.

of at least seven measurements of field blanks or low-level standards. For compounds present in the field blanks, specific VOC laboratory blank concentrations for each laboratory were statistically compared to the pooled field blank values previously run. Laboratory blanks and field blanks were pooled if the field and laboratory blank concentrations were not statistically different ($P < 0.05$). Each batch of either field or laboratory blanks was compared to the pooled data to determine whether the batch blank values were significantly different. If they were different (eg, because of a change in the lot of OVMs that may have had different background levels), MDLs were based on pooled values from the field blanks for the new batch of OVMs. This approach resulted in variable MDLs during the sampling period. For compounds not present in the field blanks, for which MDLs were estimated on the basis of repeated analysis of the lowest standard, the MDLs varied because of slight changes in analytic conditions. Evaluation of the solvent blanks and badge extraction blanks indicated that the majority of the blank contribution was from the badge, some contributions of aromatic compounds were from the carbon disulfide, and occasionally other compounds were from exposure of the solvents to air.

The presence of methylene chloride in field and laboratory blanks was found predominately at EOHSI and varied over time, which suggested contamination from the laboratory air present in the building. The differences in MDLs between the two laboratories reflect differences in (a) the sensitivity of the GC-MS procedure used by the two laboratories (even though they both used the same model GC-MS) or (b) the contributions from field or laboratory blanks. These differences represent variability of no more than several hundredths of a nanogram per injection into the GC-MS when no measurable blank levels were present, and several tenths of a nanogram per badge when blanks were present. The exceptions to this were methylene chloride (at EOHSI) and toluene (at both laboratories), which had laboratory or field blank values close to, and in a few cases exceeding, 1 ng/1 mL injection into the GC. These differences existed across both laboratories and within each laboratory over time.

APPENDIX D. Method Development and Validation of the PAKS

A passive sampler is usually preferable to an active sampler in studies involving personal measurements because passive monitors generally increase subject participation, decrease the number of samples lost to pump failure, and are more readily accepted by participants, especially small children.

At the time we submitted the proposal and started this study, only one type of commercial passive sampler was available; it was designed to measure formaldehyde in general environmental settings. The GMD badge (developed by GMD Systems of Bacharach; Pittsburgh PA) uses a DNPH-coated C_{18} cartridge as the collection medium. Carbonyl concentrations are determined by extracting DNPH-carbonyl derivatives formed on the DNPH-coated cartridge with ACN and analyzing the extracts using HPLC-UV (personal communication from Dr Maria T Morandi 1997). Although a study to evaluate GMD badges had corroborated the dosimeter's suitability for microenvironmental and personal air measurements of formaldehyde, other aldehydes were not detected by the GMD badge, probably owing to the lack of sensitivity for other aldehydes that are typically present at much lower concentrations than formaldehyde (Zhang et al 1994a; Morandi et al 1997). Thus we did not use the GMD passive badge for this study.

At the commencement of this project, Dr Zhang and coworkers were in the process of developing a new passive sampler designed to measure multiple carbonyl compounds at typical environmental levels based on 24 to 48 hours of sample collection. The PAKS uses a fluorogenic reagent, DNSH, to derivatize aldehydes (Nondek et al 1992). Similar to the DNPH derivatives of carbonyl compounds, the DNSH derivatives of carbonyl compounds can be separated with a reverse-phase HPLC column. However, the use of fluorescence detection, compared with the UV detection method used for the DNPH derivatives, can greatly enhance the sensitivity and selectivity of the DNSH technique.

During the pilot study and early phase of the full study, Zhang and coworkers continued to develop and evaluate the PAKS. The sampler was evaluated in the laboratory under a range of face velocities, temperatures, changes in relative humidity, carbonyl concentrations, and sampling durations. The evaluation results indicated that the PAKS is a valid passive sampler for 24- to 48-hour collection of carbonyl compounds for stationary and personal sampling (Zhang et al 2000).

In addition to the evaluations reported previously (Zhang et al 2000), a number of tests were made to evaluate the stability of DNSH-carbonyl derivatives on the collection medium and in the extract solution compared with those of DNPH-carbonyl derivatives. A large number of indoor, outdoor, and personal samples were also collected using collocated DNSH passive samplers (the PAKS) and DNPH active samplers in each of the three urban areas, Elizabeth, Houston, and Los Angeles. Results from both the laboratory tests and the field evaluation led to the conclusion that the DNSH passive method had several advantages over the DNPH active method for obtaining indoor, outdoor, and personal measurements of carbonyl compounds within this study.

First, most of the carbonyl compounds (including the six aldehydes targeted by HEI's RFA 97-2) detected using the active DNPH sampler with subsequent HPLC-UV analysis were also detected using the passive sampler coupled with HPLC–fluorescence analysis. Table D.1 shows the results from regression of the active method data against the passive method data. In general, the two methods agreed reasonably well for formaldehyde and acetaldehyde with high

Table D.1. Linear Regression Results from the Comparison Between Collocated Samples Obtained by the DNPH Active and the DNSH Passive Sampling Methods

Carbonyl	n [a]	Slope	SE of Slope	Intercept	SE of Intercept	r^2
Formaldehyde	86 (1)	1.181	0.077	0.092	1.604	0.737
Acetaldehyde	70 (16)	0.868	0.060	0.260	0.999	0.757
Acetone	29 (30)	0.585	0.083	1.004	0.604	0.646
Propionaldehyde	42 (39)	0.846	0.131	0.503	0.314	0.509
Benzaldehyde	43 (34)	0.753	0.097	0.429	0.315	0.596
Glyoxal	52 (26)	0.323	0.057	0.484	0.129	0.392
Methylglyoxal	44 (30)	0.644	0.136	0.880	0.366	0.347

[a] Nine pairs of data in total were excluded because of a difference in sampling duration of more than 2 hours between the active method and the passive method. Number in parentheses represents the number of pairs removed because at least one of the compound concentrations for that pair was below the MDL.

r^2 and slope values close to unity. For propionaldehyde and benzaldehyde, although the regression slopes are 0.846 and 0.753, respectively, the r^2 values are only 0.509 and 0.596, respectively, which indicates large variation between measurements obtained by the two methods. The two methods did not agree well for acetone, glyoxal, and methylglyoxal; concentrations measured using the passive method were generally higher. The difference between MDLs of these two methods makes the passive method more accurate in detecting low concentrations of carbonyl compounds than the active method. Differences in stability of carbonyl derivatives on the collection media and in the extracts may contribute, in part, to the differences in concentrations measured using the two different methods (Figures D.1 and D.2). Because the DNPH-based active method is known to give poor results for the measurement of acrolein and crotonaldehyde, comparisons were not made for these two unsaturated carbonyls.

Second, the PAKS worked substantially better for acrolein and crotonaldehyde than the DNPH sampler. This finding is supported by the stability test results and recoveries. The recoveries of acrolein and crotonaldehyde for the PAKS method were 60.3% and 76.3%, respectively, and those for the DNPH method were 20.0% and 38.6%, respectively (see Table 8). The low recoveries for the DNPH method may be related to the instability of DNPH derivatives of these unsaturated carbonyl compounds on the collection medium (C_{18} cartridge). In contrast, the DNSH derivatives of acrolein and crotonaldehyde were substantially more stable on the cartridge. In ACN extracts, the stability of DNSH derivatives was comparable to that of DNPH derivatives (see Figure D.1). With the DNPH method, acrolein and crotonaldehyde were detected in 4% and 24% of samples, respectively. The use of the passive sampler increased this percentage to 71% for acrolein and 69% for crotonaldehyde.

Finally, the use of the PAKS eliminated the potential for pump malfunctioning. Hence, the number of valid samples increased from 68% for the active method to 90% for the passive method. The use of the PAKS also reduced considerably the burden on participants and the workload on field personnel.

We submitted a special report evaluating the DNSH passive method to both HEI and the Mickey Leland National Urban Air Toxics Research Center to request substituting the DNPH active sampling method with the DNSH passive method for all types of carbonyl measurements except the in-vehicle measurements. Upon the approval of both funding agencies, the investigators started using the new protocol in May 2000. This change was consistent with the approved original project plan, in which we had indicated that we would switch the active method to the passive method at a point when the passive sampler proved to be reliable and appropriate to meet the study's objectives. This protocol change increased the overall number of valid samples, improved the data quality, especially for acrolein and crotonaldehyde, and expedited completion of the project.

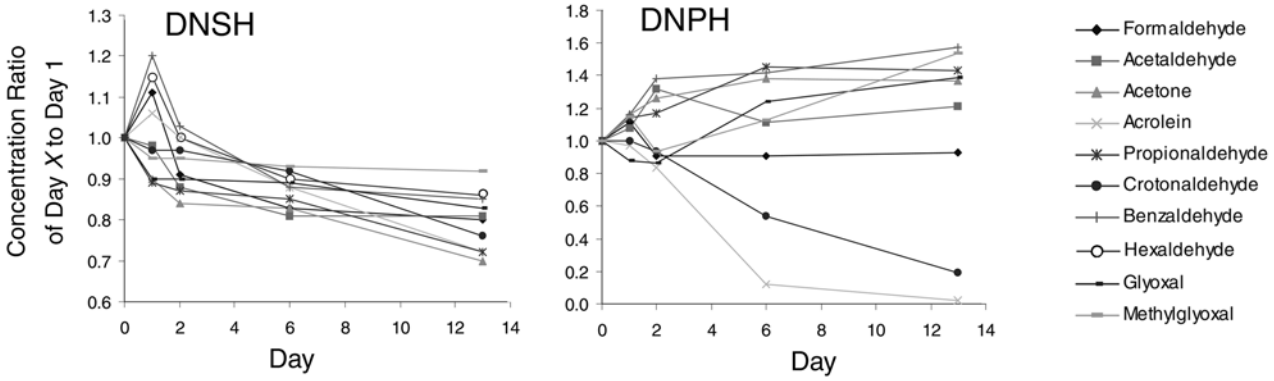

Figure D.1. Stability of DNPH- and DNSH-derived carbonyls on collection media (C_{18} cartridges), as a function of storage duration, compared with the concentration measured on Day 1.

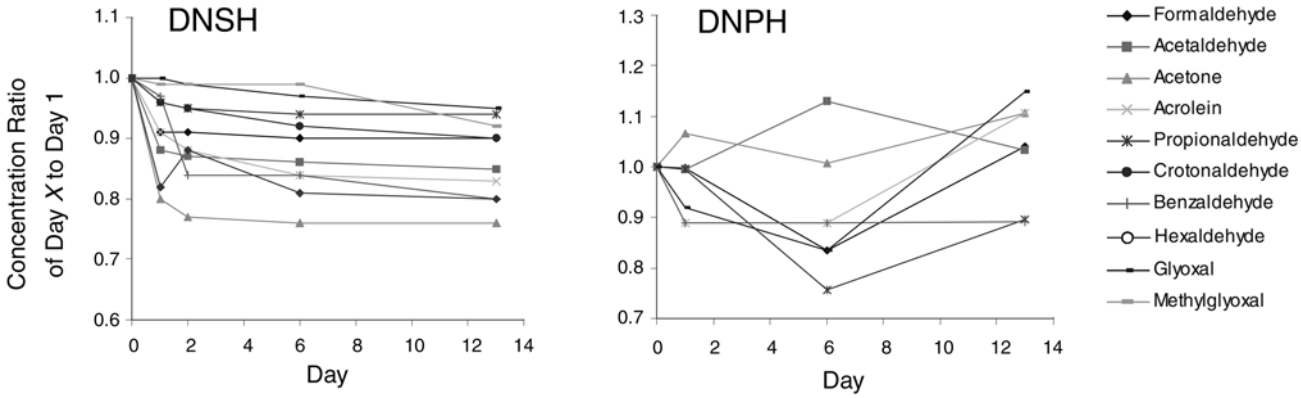

Figure D.2. Stability of DNPH- and DNSH-derived carbonyls in ACN extracts, as a function of storage duration, compared with the concentration measured on Day 1.

Table E.1. Descriptive Summary of Outdoor Air Concentrations (μg/m^3) by City[a]

	Los Angeles					Elizabeth					Houston					
Species	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*P* Value[b]
VOCs	175					182					198					
Methylene chloride		1.39	3.23	0.84	42.9		1.30	2.11	0.84	11.5		0.24	0.19	0.15	38.9	NR
MBTE		10.8	11.4	8.31	97.1		5.76	5.34	4.32	90.1		7.88	11.3	4.52	96.5	< 0.01
Chloroform		0.39	0.77	0.17	38.9	181	0.30	0.68	0.17	11.0		0.26	1.34	0.15	13.6	NR
Carbon tetrachloride		0.68	0.23	0.63	96.6		0.84	2.28	0.69	92.9		0.63	0.16	0.62	97.5	< 0.01
Benzene		2.52	2.35	1.98	81.1		1.44	1.57	1.22	58.2		2.48	2.17	1.94	98.5	< 0.01[c]
Trichloroethylene		0.21	0.31	0.11	30.9		0.57	2.23	0.38	63.7		0.13	0.09	0.12	8.6	NR
Toluene		8.96	8.67	6.65	47.4		6.77	5.75	3.02	40.7		5.75	3.99	5.36	35.9	NR
Tetrachloroethylene		1.82	1.91	1.30	76.6		1.05	3.09	0.56	23.6		0.22	0.19	0.14	34.8	NR
Ethyl benzene		1.62	1.52	1.30	81.1		1.34	2.75	0.99	58.8		0.94	0.79	0.79	94.9	< 0.01[c]
m- & p-Xylenes		4.91	5.25	3.56	92.6		3.23	4.30	2.34	94.5		2.69	2.17	2.23	97.0	< 0.01
o-Xylene		1.80	1.65	1.40	90.9		1.70	6.53	0.94	90.7		0.99	0.84	0.80	93.4	< 0.01
Styrene		0.58	1.00	0.17	34.9		0.53	3.48	0.17	18.1		0.35	0.36	0.22	34.3	NR
α-Pinene		1.72	6.62	0.32	53.7		0.77	3.18	0.32	18.1		0.26	0.29	0.11	77.8	NR
β-Pinene		0.64	1.49	0.20	25.7		0.64	3.49	0.18	17.0		0.33	0.58	0.25	11.1	NR
d-Limonene		3.65	9.08	1.27	24.6		2.46	5.53	1.27	9.3		1.21	2.51	0.94	5.6	NR
p-Dichlorobenzene		1.38	2.11	0.72	30.9		3.77	26.9	0.72	9.9		1.62	12.4	0.33	32.8	NR
Carbonyls (passive method)	133					138					124					
Formaldehyde		6.51	2.33	6.52	100.0		6.35	2.81	7.09	99.3		6.30	2.42	6.16	100.0	0.50
Acetaldehyde		5.89	2.71	5.27	97.7		8.88	6.50	7.86	98.6		5.93	4.16	4.70	95.2	< 0.01
Acetone		4.78	4.43	4.19	90.2		3.70	3.18	3.00	85.5		21.8	123	6.52	99.2	< 0.01
Acrolein		1.02	2.31	0.40	67.7		0.89	1.29	0.39	59.4		17.9	181	0.95	75.8	0.01[c]
Propionaldehyde		2.04	1.36	1.82	97.0		1.21	0.95	1.06	90.6		1.47	0.87	1.34	94.4	< 0.01
Crotonaldehyde		0.54	0.67	0.33	66.2		0.39	0.73	0.17	55.1		1.45	9.37	0.35	64.5	0.04[c]
Benzaldehyde		2.54	1.33	2.67	92.5		1.64	1.22	1.42	91.3		1.91	1.06	1.64	98.4	< 0.01
Hexaldehyde		2.49	1.38	2.23	100.0		1.45	1.00	1.35	98.6		3.09	5.33	2.52	98.4	< 0.01
Glyoxal		1.96	0.89	2.00	98.5		1.53	0.87	1.38	100.0		1.97	0.86	1.90	98.4	< 0.01
Methylglyoxal		2.18	1.15	2.07	97.0		1.74	1.02	1.81	94.2		2.27	0.97	2.29	98.4	< 0.01

[a] *n* is the total samples for each compound within a group, unless otherwise noted.

[b] *P* values based on Kruskal-Wallis test for intercity differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported because the proportion of nondetected samples was > 40%.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.1 (*continued*). Descriptive Summary of Outdoor Air Concentrations (μg/m^3) by City[a]

	Los Angeles					Elizabeth					Houston					
Species	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*P* Value[b]
Carbonyls (active method)	42					45					30					
Formaldehyde		6.12	3.58	4.81	97.6		2.75	1.67	2.39	95.6		2.23	1.47	2.08	76.7	< 0.01
Acetaldehyde		3.42	1.56	3.29	97.6		2.88	1.47	2.47	86.7		3.41	1.97	3.10	56.7	0.19
Acetone		1.98	1.42	1.51	64.3		1.89	1.00	1.53	20.0		1.16	1.00	0.71	6.7	NR
Propionaldehyde		0.71	0.32	0.70	78.6		0.88	0.37	0.79	100		1.55	1.48	1.34	76.7	< 0.01[c]
Benzaldehyde		2.11	2.13	1.17	61.9		2.69	2.03	2.47	80		2.98	2.41	2.51	53.3	0.06[c]
Glyoxal		0.53	0.39	0.49	69.0		0.60	0.35	0.54	91.1		0.26	0.24	0.20	43.3	< 0.01[c]
Methylglyoxal		0.45	0.28	0.28	38.1		0.41	0.24	0.44	55.6		7.54	28.74	0.11	33.3	NR
Butyraldehyde		0.72	0.34	0.76	59.5		0.70	0.27	0.65	53.3		1.29	0.47	1.09	3.3	NR
Isovaleraldehyde		0.50	0.39	0.39	11.9		0.57	0.44	0.43	8.9		0.55	0.93	0.22	10.0	NR
Valeraldehyde		0.58	0.30	0.58	47.6		0.62	0.36	0.59	40		1.27	0.64	1.12	16.7	NR
$PM_{2.5}$	121	19.2	13.3	16.1	100	103	20.4	10.7	18.2	100	110	14.7	5.75	13.2	100	< 0.01

[a] *n* is the total samples for each compound within a group, unless otherwise noted.

[b] *P* values based on Kruskal-Wallis test for intercity differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported because the proportion of nondetected samples was > 40%.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.2. Descriptive Summary of Indoor Air Concentrations (μg/m^3) by City[a]

	Los Angeles					Elizabeth					Houston					
Species	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*P* Value[b]
VOCs	174					182					198					
Methylene chloride		1.72	2.83	0.84	50.6		1.63	4.20	0.84	15.9		3.46	17.1	0.44	66.7	NR
MTBE		13.2	33.1	7.44	92.0	181	7.38	9.58	5.03	87.8		14.7	31.9	5.82	99.0	< 0.01
Chloroform		1.58	2.13	0.92	81.0		1.64	3.46	0.74	67.6		2.31	3.07	1.32	88.9	< 0.01[c]
Carbon tetrachloride		0.79	1.69	0.58	92.5		0.66	0.30	0.63	91.8		0.68	0.28	0.62	98.5	0.07
Benzene		3.00	5.00	2.05	83.9		2.52	3.97	1.65	70.9		4.85	5.93	3.06	100.0	< 0.01[c]
Trichloroethylene		0.25	0.83	0.11	34.5		0.95	2.51	0.43	69.8		0.18	0.26	0.12	21.2	NR
Toluene		16.5	33.6	10.6	67.8		12.7	11.6	9.74	74.2		16.8	23.4	10.3	68.2	0.39[c]
Tetrachloroethylene		3.29	7.07	1.66	83.3		1.26	1.99	0.56	38.5		1.02	2.42	0.29	67.2	NR
Ethyl benzene		2.45	3.51	1.45	85.6		2.30	5.71	1.29	72.0		2.78	4.72	1.68	100.0	< 0.01[c]
m- & p-Xylenes		6.88	9.33	4.16	94.8		6.52	19.0	3.18	96.7		8.47	17.2	4.55	100.0	< 0.01
o-Xylene		2.45	3.13	1.64	94.3		2.17	5.31	1.18	93.4		2.80	5.40	1.53	98.0	< 0.01
Styrene		1.24	2.07	0.49	56.9		1.40	4.08	0.17	45.6		1.58	5.64	0.67	79.8	NR
α-Pinene		5.87	14.6	1.12	86.8		3.93	10.8	1.45	81.3		4.86	14.8	0.14	100.0	< 0.01[c]
β-Pinene		2.98	9.22	0.79	66.1		3.24	11.2	0.69	63.7		7.84	11.6	4.03	93.9	< 0.01[c]
d-Limonene		22.0	45.0	7.31	77.6		14.7	24.5	6.71	69.8		54.0	170	20.8	94.4	< 0.01[c]
p-Dichlorobenzene		38.8	316	0.76	50.0		29.3	121	0.72	48.9		132	390	2.02	75.8	NR
Carbonyls (passive method)	134					139					125					
Formaldehyde		21.5	7.44	19.0	100.0		22.4	7.09	21.2	100.0		20.9	6.79	19.8	100.0	0.11
Acetaldehyde		24.1	13.3	20.7	100.0		16.3	8.94	14.3	100.0		30.0	27.1	22.3	99.2	< 0.01
Acetone		8.52	9.68	6.30	95.5		11.6	17.3	7.04	96.4		22.4	30.9	12.9	100.0	< 0.01
Acrolein		1.21	2.27	0.47	73.1		0.96	1.33	0.49	63.3		3.08	13.3	0.92	79.2	< 0.01[c]
Propionaldehyde		1.91	1.12	1.70	97.0		1.98	3.13	1.45	94.2		2.28	0.91	2.36	99.2	< 0.01
Crotonaldehyde		0.58	0.77	0.37	67.9		0.69	0.92	0.40	66.9		0.83	0.92	0.55	76.0	0.03[c]
Benzaldehyde		3.00	1.47	2.94	94.0		3.05	1.31	3.18	99.3		3.02	1.28	2.70	100.0	0.63
Hexaldehyde		3.96	1.82	3.51	99.3		3.81	2.31	3.21	100.0		5.95	3.42	5.66	100.0	< 0.01
Glyoxal		2.63	1.01	2.57	99.3		2.38	0.95	2.32	100.0		2.82	0.79	2.72	100.0	< 0.01
Methylglyoxal		3.09	2.78	2.83	99.3		2.33	1.02	2.47	98.6		3.21	0.98	3.22	100.0	< 0.01

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for intercity differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported because the proportion of nondetected samples was > 40%.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.2 (*continued*). Descriptive Summary of Indoor Air Concentrations (μg/m^3) by City[a]

	Los Angeles					Elizabeth					Houston					
Species	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*P* Value[b]
Carbonyls (active method)	49					38					34					
Formaldehyde		28.0	13.4	26.0	100		21.9	11.2	20.5	100		24.9	15.5	21.0	94.1	0.07
Acetaldehyde		12.5	9.79	10.5	98.0		10.6	10.6	8.69	86.8		12.6	11.4	8.79	70.6	0.35
Acetone		1.56	1.30	0.72	46.9		3.90	7.68	1.46	15.8		2.60	3.12	2.19	32.4	NR
Propionaldehyde		2.14	1.31	1.90	95.9		2.02	1.49	1.91	97.4		2.17	1.32	2.36	79.4	0.71[c]
Benzaldehyde		1.83	1.21	1.59	93.9		1.93	1.30	1.62	81.6		2.79	2.75	1.33	47.1	0.85[c]
Glyoxal		0.87	0.43	0.84	89.8		1.23	0.63	1.22	92.1		0.48	0.36	0.47	41.2	< 0.01[c]
Methylglyoxal		1.48	0.88	1.44	85.7		1.97	1.70	1.77	78.9		5.42	19.0	0.42	26.5	< 0.01[c]
Butyraldehyde		1.22	0.56	1.11	79.6		1.39	0.85	1.12	78.9		1.71	0.98	1.51	17.6	0.03[c]
Isovaleraldehyde		0.99	0.71	0.86	57.1		1.06	0.73	0.94	63.2		1.38	1.04	1.39	47.1	NR
Valeraldehyde		1.98	1.24	1.71	85.7		2.18	1.71	1.58	76.3		2.61	2.12	2.02	44.1	0.32[c]
$PM_{2.5}$	124	16.2	9.38	14.5	100	96	20.1	15.5	15.7	100	106	17.1	12.7	13.4	100	

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for intercity differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported because the proportion of nondetected samples was > 40%.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.3. Descriptive Summary of Adult Personal Air Concentrations (μg/m^3) by City[a]

	Los Angeles					Elizabeth					Houston					
Species	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*P* Value[b]
VOCs	175					171					199					
Methylene chloride		3.74	25.0	0.84	51.4		1.64	3.83	0.84	11.1		3.62	15.6	0.51	74.4	NR
MTBE		12.2	13.5	8.51	97.1	170	14.7	65.6	5.51	91.8		17.1	33.9	7.32	98.5	< 0.01
Chloroform		8.49	92.4	0.87	84.6	168	2.02	4.21	0.85	75.6		2.27	2.96	1.33	93.0	< 0.01[c]
Carbon tetrachloride		0.85	3.16	0.58	93.7		0.87	2.96	0.64	88.9		0.67	0.23	0.62	99.5	0.07
Benzene		3.10	6.52	2.28	84.6		2.80	4.19	1.76	76.0		4.82	4.76	3.13	100.0	< 0.01[c]
Trichloroethylene		0.39	1.29	0.12	45.7		2.38	15.6	0.50	76.0		0.21	0.57	0.12	28.6	NR
Toluene		18.9	49.2	11.9	70.9		20.8	38.6	11.3	76.0		18.0	20.1	13.1	74.9	0.52[c]
Tetrachloroethylene		3.75	9.58	1.72	85.7		17.3	200	0.56	44.4		1.39	4.75	0.36	76.9	NR
Ethyl benzene		2.36	3.59	1.67	84.6		2.91	6.89	1.40	76.0		3.06	4.45	1.83	99.5	< 0.01[c]
m- & *p*-Xylenes		7.07	9.74	4.54	96.6		7.93	20.3	4.04	96.5		9.07	14.7	5.10	100.0	< 0.01
o-Xylene		2.57	3.41	1.85	97.1		3.07	8.05	1.56	95.9		3.02	4.40	1.80	100.0	< 0.01
Styrene		1.16	1.95	0.49	60.0		1.75	5.58	0.36	53.8		1.61	4.59	0.84	89.9	< 0.01[c]
α-Pinene		3.87	8.05	0.96	85.7		5.03	16.5	1.96	82.9		3.83	6.85	0.14	100.0	< 0.01
β-Pinene		2.72	8.55	0.73	65.1	170	5.09	16.2	0.96	73.7		8.26	12.9	4.16	95.5	< 0.01[c]
d-Limonene		48.2	387	7.65	80.0		18.0	32.1	8.39	74.9		55.0	153	22.4	98.0	< 0.01[c]
p-Dichlorobenzene		14.9	63.2	1.15	52.6		26.5	114	1.85	56.7		120	351	3.42	78.9	NR
Carbonyls (passive method)	142					142					125					
Formaldehyde		21.7	6.48	20.3	100.0		21.9	6.92	20.6	100.0		21.6	12.9	20.4	100.0	0.33
Acetaldehyde		22.7	11.2	20.1	99.3		17.3	9.18	15.1	100.0		29.4	20.3	22.3	100.0	< 0.01
Acetone		9.04	10.6	6.75	97.2		29.3	129	8.22	97.9		41.2	146	13.4	99.2	< 0.01
Acrolein		1.12	1.62	0.47	70.4		0.74	1.09	0.26	55.6		40.0	249	0.91	80.8	< 0.01[c]
Propionaldehyde		2.15	1.15	2.10	97.2		1.69	0.97	1.51	97.2		2.18	1.13	2.19	94.4	< 0.01
Crotonaldehyde		0.63	0.72	0.42	70.4		0.63	1.06	0.30	59.2		2.59	11.3	0.63	74.4	< 0.01[c]
Benzaldehyde		3.21	1.50	3.11	95.8		3.33	1.67	3.20	95.8		3.56	2.69	2.90	100.0	0.64
Hexaldehyde		4.28	1.87	3.95	100.0		4.10	2.42	3.58	100.0		7.70	12.1	5.38	100.0	< 0.01
Glyoxal		2.66	1.11	2.52	100.0		2.37	0.80	2.22	100.0		2.92	1.94	2.64	100.0	< 0.01
Methylglyoxal		2.89	1.10	2.83	100.0		2.30	0.92	2.28	100.0		3.11	1.04	2.98	100.0	< 0.01

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for intercity differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported because the proportion of nondetected samples was > 40%.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.3 (*continued*). Descriptive Summary of Adult Personal Air Concentrations (μg/m^3) by City[a]

	Los Angeles					Elizabeth					Houston					
Species	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*P* Value[b]
Carbonyls (active method)	58					34					37					
Formaldehyde		25.9	11.0	25.3	98.3		26.7	16.1	22.7	100		26.7	17.3	22.3	100	0.73
Acetaldehyde		13.7	7.61	11.5	96.6		16.9	15.0	13.6	91.2		18.4	11.6	15.1	86.5	0.23[c]
Acetone		5.20	5.34	3.72	70.7		7.14	15.0	2.79	32.4		8.24	21.7	2.60	32.4	NR
Propionaldehyde		2.36	1.40	2.12	87.9		3.05	3.96	2.32	100		4.21	3.96	3.20	94.6	< 0.01[c]
Benzaldehyde		1.74	1.74	1.23	50.0		2.15	1.81	1.49	70.6		3.15	2.53	2.09	24.3	NR
Glyoxal		0.89	0.49	0.79	60.3		1.35	0.70	1.17	91.2		0.68	0.68	0.47	45.9	NR
Methylglyoxal		1.79	0.84	1.76	81.0		2.66	2.61	2.28	82.4		4.56	18.3	0.37	29.7	NR
Butyraldehyde		1.51	0.68	1.45	72.4		1.83	1.55	1.46	79.4		3.14	1.12	3.06	21.6	NR
Isovaleraldehyde		1.07	0.71	1.11	51.7		1.40	0.89	1.25	61.8		1.71	1.13	1.53	40.5	NR
Valeraldehyde		2.13	1.31	2.00	82.8		3.48	6.33	2.18	94.1		4.24	3.59	3.30	40.5	NR
$PM_{2.5}$	105	29.2	14.8	26.5	100	77	44.8	29.9	37.4	100	98	37.2	23.8	31.6	100	< 0.01

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for intercity differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported because the proportion of nondetected samples was > 40%.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.4. Descriptive Summary of Child Personal Air Concentrations (μg/m^3) by City[a]

	Los Angeles					Elizabeth					Houston					
Species	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*P* Value[b]
VOCs	33					41					135					
Methylene chloride		1.28	1.40	0.84	51.5		2.09	3.30	0.84	22.0		1.62	7.87	0.36	80.7	NR
MTBE		8.35	6.86	5.87	97.0		9.24	10.51	6.63	82.9		13.2	26.5	7.19	97.8	0.33
Chloroform		0.92	0.97	0.71	69.7		2.61	6.16	1.06	75.6		2.12	2.92	1.41	96.3	< 0.01c
Carbon tetrachloride		0.48	0.18	0.51	84.8		0.62	0.22	0.63	92.7		0.56	0.14	0.56	93.3	< 0.01
Benzene		2.23	1.36	2.21	87.9		2.76	4.00	1.97	80.5		5.07	6.37	3.96	100.0	< 0.01[c]
Trichloroethylene		0.19	0.20	0.11	33.3		1.01	1.85	0.52	75.6		0.13	0.11	0.11	23.0	NR
Toluene		15.6	14.9	9.83	69.7		26.3	49.4	11.3	82.9		16.7	19.8	12.6	83.0	0.93[c]
Tetrachloroethylene		5.01	15.2	1.55	90.9		1.57	2.97	0.56	41.5		2.65	18.3	0.39	83.7	NR
Ethyl benzene		1.86	1.15	1.69	87.9		3.41	9.24	1.53	82.9		3.67	6.01	2.08	99.3	< 0.01[c]
m- & *p*-Xylenes		4.64	3.03	4.13	93.9		10.7	31.5	3.90	97.6		9.36	11.5	5.53	100.0	0.03
o-Xylene		1.77	0.90	1.72	90.9		3.28	9.12	1.48	97.6		3.08	3.40	2.01	98.5	0.06
Styrene		1.20	1.76	0.59	69.7		2.18	4.75	0.55	70.7		1.66	4.69	0.78	90.4	0.08[c]
α-Pinene		1.52	2.15	0.57	84.8		4.06	4.13	2.18	92.7		3.78	5.69	0.14	100.0	< 0.01
β-Pinene		1.13	1.35	0.56	54.5		3.19	5.73	1.23	75.6		6.99	6.44	4.46	97.0	< 0.01[c]
d-Limonene		10.1	8.35	7.55	84.8		20.0	23.5	14.5	78.0		41.2	58.4	24.5	100.0	< 0.01[c]
p-Dichlorobenzene		1.41	1.88	0.72	30.3		36.4	159	1.46	51.2		177	369	10.2	94.8	NR
Carbonyls (passive method)	24					31					114					
Formaldehyde		22.4	6.87	24.0	100.0		20.5	6.33	21.2	100.0		20.6	7.46	19.6	100.0	0.33
Acetaldehyde		26.6	20.5	23.1	100.0		18.2	7.53	16.0	100.0		26.4	20.5	20.5	100.0	0.02
Acetone		12.4	16.4	7.51	95.8		14.5	13.7	9.32	100.0		36.6	119	13.3	100.0	< 0.01
Acrolein		0.82	1.08	0.52	62.5		1.74	2.16	0.85	87.1		15.5	128	1.00	75.4	0.15[c]
Propionaldehyde		2.78	1.95	2.30	100.0		2.26	1.12	2.04	96.8		6.44	41.6	2.50	98.2	0.28
Crotonaldehyde		0.56	0.43	0.54	75.0		1.05	1.05	0.58	80.6		1.82	7.89	0.57	74.6	0.48[c]
Benzaldehyde		2.84	1.84	3.25	79.2		3.18	1.11	3.15	100.0		3.25	1.91	2.79	100.0	0.54[c]
Hexaldehyde		3.86	1.67	3.80	100.0		4.85	1.93	4.52	100.0		7.41	13.8	5.06	100.0	0.01
Glyoxal		2.79	0.82	2.92	100.0		2.55	0.78	2.65	100.0		3.05	2.17	2.79	100.0	0.26
Methylglyoxal		3.34	1.54	2.82	100.0		2.59	1.09	2.22	100.0		3.17	1.34	3.01	98.2	0.02
$PM_{2.5}$	1	40.2	—[d]	40.2	100	23	54.0	32.0	39.2	100	3	36.6	7.97	39.1	100	NR

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for intercity differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

[d] Dash indicates no data.

Table E.5. Descriptive Summary of In-Vehicle Air Concentrations (μg/m^3) of Carbonyls Measured Using the Active Method by City[a]

	Los Angeles					Elizabeth					Houston					
Species	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*n*	Mean	SD	Median	Percentage Above the LOD	*P* Value[b]
Carbonyls (active method)	72					10					33					
Formaldehyde		38.0	36.5	25.6	98.6		14.9	9.30	12.9	100		50.9	189	10.2	75.8	< 0.01[c]
Acetaldehyde		6.92	6.29	5.47	86.1		5.67	4.48	4.10	20		70.9	249	9.43	27.3	NR
Acetone		6.68	7.57	3.33	27.8		6.60	4.21	5.32	0		56.1	190	5.45	18.2	NR
Propionaldehyde		1.92	1.54	1.49	33.3		2.42	1.51	2.24	100		16.4	58.4	2.75	45.5	NR
Benzaldehyde		2.37	2.23	1.66	2.8		1.79	1.14	1.45	0		60.7	229	5.58	3.0	NR
Glyoxal		1.70	1.89	1.21	33.3		1.10	0.70	1.04	30		6.52	19.4	0.41	18.2	NR
Methylglyoxal		2.15	1.88	1.68	27.8		1.31	0.65	1.36	30		4.85	17.3	0.50	21.2	NR
Butyraldehyde		3.54	9.38	1.73	45.8		2.39	1.55	2.23	60		64.6	238	6.41	0	NR
Isovaleraldehyde		3.91	11.0	1.62	25		1.97	1.99	1.07	40		14.8	55.6	1.15	6.1	NR
Valeraldehyde		3.64	8.57	1.92	50		2.35	1.44	1.73	60		72.6	275	4.95	6.1	NR

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for intercity differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported because the proportion of nondetected samples was > 40%.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.6. Descriptive Summary of Paired Adult–Child Personal Concentrations (μg/m^3) in Los Angeles[a]

	Adult				Child				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	20				33				
Methylene chloride		1.20	0.66	0.85		1.28	1.40	0.84	NR
MTBE		11.8	10.0	7.47		8.35	6.86	5.87	0.01
Chloroform		0.85	0.77	0.60		0.92	0.97	0.71	0.48[c]
Carbon tetrachloride		0.53	0.24	0.52		0.48	0.18	0.51	0.81
Benzene		2.74	1.72	2.38		2.23	1.36	2.21	0.19[c]
Trichloroethylene		0.19	0.15	0.12		0.19	0.20	0.11	NR
Toluene		12.0	9.72	7.19		15.6	14.9	9.83	0.04[c]
Tetrachloroethylene		6.45	17.0	1.82		5.01	15.2	1.55	0.18[c]
Ethyl benzene		1.83	1.06	1.72		1.86	1.15	1.69	0.91[c]
m- & *p*-Xylenes		5.25	3.83	4.30		4.64	3.03	4.13	0.29
o-Xylene		1.97	1.10	1.92		1.77	0.90	1.72	0.57
Styrene		1.61	2.16	0.59		1.20	1.76	0.59	0.45[c]
α-Pinene		1.55	2.24	0.58		1.52	2.15	0.57	0.76[c]
β-Pinene		0.87	0.94	0.53		1.13	1.35	0.56	0.59[c]
d-Limonene		8.87	7.60	7.37		10.1	8.35	7.55	0.16[c]
p-Dichlorobenzene		1.32	1.45	0.72		1.41	1.88	0.72	NR
Carbonyls (passive method)	15				20				
Formaldehyde		19.7	6.49	20.3		24.2	6.03	25.2	0.10
Acetaldehyde		21.7	12.6	18.3		21.8	6.92	23.1	0.42
Acetone		17.1	19.9	8.09		13.1	17.9	7.35	0.19
Acrolein		0.69	0.79	0.62		0.90	1.16	0.52	0.67[c]
Propionaldehyde		1.86	1.56	1.59		2.38	1.02	2.06	0.02
Crotonaldehyde		0.48	0.41	0.45		0.60	0.44	0.54	0.51[c]
Benzaldehyde		3.15	2.01	2.77		3.05	1.79	3.30	0.55
Hexaldehyde		3.79	1.63	3.01		4.24	1.49	3.96	0.03
Glyoxal		2.35	0.55	2.42		2.79	0.86	2.92	0.19
Methylglyoxal		2.81	1.29	2.70		3.03	1.01	2.74	0.19

[a] Only paired adult–child samples within the same home are presented here. *n* is the total samples for each compound within a group. $PM_{2.5}$ data are not reported because no valid paired adult–child samples were found.

[b] *P* values based on results of incomplete randomized block mixed model. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.7. Descriptive Summary of Paired Adult–Child Personal Concentrations (μg/m^3) in Elizabeth[a]

Species	Adult				Child				*P* Value[b]
	n	Mean	SD	Median	*n*	Mean	SD	Median	
VOCs	29				30				
Methylene chloride		2.51	4.30	0.84		2.25	3.59	0.84	NR
MTBE	28	8.38	6.01	7.03		9.43	11.6	7.44	0.74
Chloroform		3.05	8.45	1.36		2.92	7.03	1.15	0.64[c]
Carbon tetrachloride		0.60	0.26	0.56		0.65	0.21	0.63	0.16[c]
Benzene		3.13	5.02	1.89		3.12	4.60	2.03	0.78[c]
Trichloroethylene		1.19	3.00	0.52		1.22	2.13	0.56	0.84[c]
Toluene		22.4	47.1	12.2		23.7	43.3	11.2	0.69[c]
Tetrachloroethylene		1.77	4.02	0.56		1.62	3.46	0.56	0.58[c]
Ethyl benzene		4.09	11.7	1.74		4.10	10.8	1.59	0.32[c]
m- & *p*-Xylenes		13.0	39.8	4.17		12.8	36.7	3.84	0.81
o-Xylene		4.05	11.4	1.75		3.94	10.6	1.58	0.98
Styrene		1.99	3.98	0.50		1.51	2.86	0.56	NR
α-Pinene		3.70	3.78	1.93		3.70	3.77	2.19	0.62[c]
β-Pinene		7.05	24.6	1.28		3.85	6.55	1.25	0.91[c]
d-Limonene		12.9	14.1	8.10		16.2	13.9	13.6	0.08[c]
p-Dichlorobenzene		10.1	26.8	0.72		45.6	186	2.21	NR
Carbonyls (passive method)	22				24				
Formaldehyde		22.3	7.81	21.1		21.8	6.03	22.0	0.77
Acetaldehyde		19.0	7.38	17.0		17.9	6.76	17.5	0.85
Acetone		17.4	28.8	9.20		13.2	13.6	7.75	0.66
Acrolein		0.79	1.03	0.42		1.32	2.06	0.71	NR
Propionaldehyde		1.97	1.12	1.68		2.32	1.32	1.97	0.41
Crotonaldehyde		0.65	0.67	0.34		1.04	1.01	0.61	NR
Benzaldehyde		3.40	1.32	3.35		3.50	1.13	3.46	0.47
Hexaldehyde		5.55	2.53	4.60		4.89	2.01	4.52	0.19
Glyoxal		2.28	0.78	2.21		2.55	0.67	2.66	0.03
Methylglyoxal		2.63	0.86	2.49		2.93	1.17	2.53	0.83
$PM_{2.5}$	11	44.1	20.0	39.5	11	50.8	28.2	38.9	0.23

[a] Only paired adult–child samples within the same home are presented here. *n* is the total samples for each compound within a group, unless otherwise noted.

[b] *P* values based on results of incomplete randomized block mixed model. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.8. Descriptive Summary of Paired Adult-Child Personal Concentrations (μg/m^3) in Houston[a]

	Adult				Child				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	79				135				
Methylene chloride		3.03	14.4	0.52		1.62	7.87	0.36	0.10[c]
MTBE		14.8	26.2	8.06		13.2	26.5	7.19	< 0.01
Chloroform		2.65	3.35	1.61		2.12	2.92	1.41	< 0.01
Carbon tetrachloride		0.61	0.11	0.59		0.56	0.14	0.56	0.01
Benzene		5.16	5.33	3.50		5.07	6.37	3.96	< 0.01
Trichloroethylene		0.13	0.07	0.12		0.13	0.11	0.11	NR
Toluene		18.5	17.6	15.6		16.7	19.8	12.6	0.11
Tetrachloroethylene		0.95	2.35	0.36		2.65	18.26	0.39	0.02[c]
Ethyl benzene		3.24	3.39	2.32		3.67	6.01	2.08	0.23[c]
m- & *p*-Xylenes		9.10	10.85	5.74		9.36	11.45	5.53	0.63
o-Xylene		3.07	3.41	1.97		3.08	3.40	2.01	0.47
Styrene		2.40	6.83	0.88		1.66	4.69	0.78	0.02
α-Pinene		4.20	6.43	1.12		3.78	5.69	0.14	0.72
β-Pinene		8.43	7.54	6.84		6.99	6.44	4.46	< 0.01
d-Limonene		48.1	46.4	32.1		41.2	58.4	24.5	0.01
p-Dichlorobenzene		167	395	5.98		177	369	10.2	< 0.01[c]
Carbonyls (passive method)	61				107				
Formaldehyde		21.1	7.3	20.0		20.6	7.44	19.8	0.08
Acetaldehyde		28.2	18.1	22.3		26.6	21.1	20.6	0.16
Acetone		65.2	207	14.4		37.9	122	13.3	0.09
Acrolein		80.1	353	0.80		16.3	132	0.94	0.39[c]
Propionaldehyde		2.19	1.24	2.20		6.67	42.9	2.50	0.02
Crotonaldehyde		4.28	16.0	0.58		1.88	8.14	0.54	0.54[c]
Benzaldehyde		3.67	2.45	2.91		3.25	1.90	2.80	0.20
Hexaldehyde		9.13	16.9	4.84		7.40	14.2	5.10	0.52
Glyoxal		3.08	2.65	2.57		3.06	2.23	2.78	0.52
Methylglyoxal		3.05	1.12	3.03		3.21	1.37	3.10	0.85
$PM_{2.5}$	3	45.1	27.2	59.3	3	36.6	7.97	39.1	NR

[a] Only paired adult–child samples within the same home are presented here. *n* is the total samples for each compound within a group.

[b] *P* values based on results of incomplete randomized block mixed model. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.9. Descriptive Summary of Outdoor Air Concentrations (μg/m^3) in Los Angeles by Season[a]

	Spring				Summer				Fall				Winter				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	21				54				55				18				
Methylene chloride		0.59	0.35	0.53		0.83	0.44	0.84		2.43	5.58	0.84		1.37	1.21	0.90	NR
MTBE		5.16	2.83	4.59		9.66	14.53	5.93		15.2	12.0	11.4		10.7	7.98	9.13	< 0.01
Chloroform		0.17	0.03	0.17		0.64	1.32	0.25		0.37	0.27	0.33		0.17	0.08	0.12	NR
Carbon tetrachloride		0.66	0.12	0.63		0.69	0.19	0.65		0.64	0.22	0.61		0.66	0.15	0.62	0.78
Benzene		1.33	0.74	1.21		1.56	1.25	1.25		3.92	3.22	3.14		3.18	1.51	3.63	< 0.01[c]
Trichloroethylene		0.31	0.55	0.14		0.13	0.06	0.11		0.17	0.16	0.10		0.12	0.08	0.11	NR
Toluene		7.03	5.88	5.57		7.21	8.58	6.62		10.1	9.90	7.53		9.41	5.64	8.08	NR
Tetrachloroethylene		0.56	0.25	0.56		1.39	1.28	0.76		2.83	2.68	2.10		1.76	1.25	1.34	NR
Ethyl benzene		0.83	0.42	0.79		1.18	1.13	0.98		2.48	2.08	1.96		1.61	0.92	1.63	< 0.01[c]
m- & *p*-Xylenes		2.53	1.33	2.33		3.69	3.93	3.01		8.05	7.03	5.86		6.17	3.43	6.46	< 0.01
o-Xylene		0.88	0.42	0.90		1.33	1.31	1.04		2.73	2.13	2.32		2.11	1.20	2.16	< 0.01
Styrene		0.23	0.09	0.19		0.40	1.03	0.17		1.17	1.26	0.74		0.27	0.22	0.17	NR
α-Pinene		0.24	0.10	0.23		4.58	11.46	0.64		0.38	0.58	0.32		0.37	0.30	0.32	NR
β-Pinene		0.23	0.11	0.23		0.98	2.30	0.22		0.74	1.27	0.26		0.22	0.06	0.19	NR
d-Limonene		1.88	1.09	1.93		6.68	13.21	3.11		3.18	8.85	1.27		1.01	0.15	0.95	NR
p-Dichlorobenzene		0.77	0.49	0.70		2.19	2.73	1.38		1.25	2.37	0.72		0.73	0.30	0.74	NR
Carbonyls (passive method)	16				54				46				17				
Formaldehyde		6.17	2.28	5.97		6.69	1.83	6.73		7.06	2.56	7.06		4.77	2.46	3.59	0.01
Acetaldehyde		4.47	1.71	5.10		6.10	2.48	5.41		6.72	2.95	5.63		4.30	2.52	4.09	0.01
Acetone		4.96	2.70	4.26		3.51	2.73	3.23		6.00	6.15	4.47		5.38	3.76	5.38	0.01
Acrolein		0.49	0.92	0.07		0.96	1.59	0.49		1.11	3.09	0.42		1.49	2.72	0.72	0.05[c]
Propionaldehyde		3.24	1.95	2.81		2.17	1.23	2.22		1.66	0.91	1.61		1.57	1.47	1.28	< 0.01
Crotonaldehyde		0.64	1.16	0.07		0.62	0.72	0.43		0.43	0.32	0.45		0.51	0.66	0.24	0.37[c]
Benzaldehyde		2.86	1.20	2.68		2.78	1.23	2.91		2.20	1.24	2.66		2.44	1.83	2.18	0.17
Hexaldehyde		3.84	1.81	4.12		2.82	1.24	2.68		1.67	0.70	1.53		2.35	1.45	2.26	< 0.01
Glyoxal		2.09	0.49	2.05		1.85	0.85	2.01		2.14	0.93	2.21		1.70	1.11	1.56	0.07
Methylglyoxal		2.39	0.52	2.32		2.48	1.25	2.25		1.78	1.04	1.62		2.07	1.26	1.67	0.01
$PM_{2.5}$	25	16.4	6.9	15.2	34	19.8	10.2	17.3	35	25.2	18.6	19.0	27	13.4	9.62	10.7	< 0.01

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for interseason differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.10. Descriptive Summary of Outdoor Air Concentrations (μg/m^3) in Elizabeth by Season[a]

	Spring				Summer				Fall				Winter				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	37				53				55				37				
Methylene chloride		0.90	0.28	0.84		1.56	3.35	0.84		1.33	1.42	0.84		1.29	1.67	0.84	NR
MTBE		5.47	4.92	5.32		5.26	4.97	4.19		6.65	5.77	5.22		5.44	5.64	2.80	0.5
Chloroform		0.51	1.39	0.17		0.29	0.37	0.17		0.22	0.22	0.17	36	0.22	0.17	0.17	NR
Carbon tetrachloride		0.71	0.12	0.70		0.68	0.21	0.73		0.72	0.27	0.77		1.36	5.05	0.56	< 0.01
Benzene		1.09	0.74	1.04		0.93	0.77	0.48		1.73	1.04	1.75		2.11	2.88	1.62	NR
Trichloroethylene		0.36	0.19	0.38		0.45	0.47	0.43		0.39	0.30	0.37		1.25	4.89	0.33	0.99[c]
Toluene		8.14	6.35	3.02		4.85	3.71	3.02		6.14	4.24	3.02		9.07	8.10	6.50	NR
Tetrachloroethylene		0.67	0.36	0.56		0.72	0.39	0.56		1.13	0.80	0.56		1.78	6.77	0.56	NR
Ethyl benzene		0.80	0.47	0.82		0.96	0.88	0.36		1.45	0.98	1.48		2.27	5.81	1.26	NR
m- & p-Xylenes		2.63	1.23	2.26		2.28	2.82	1.81		3.77	2.66	2.70		4.41	8.12	3.00	< 0.01
o-Xylene		0.84	0.35	0.87		2.28	11.03	0.72		1.46	0.85	1.15		2.09	6.01	1.16	< 0.01
Styrene		0.19	0.14	0.17		0.22	0.21	0.17		0.38	0.37	0.17		1.55	7.69	0.17	NR
α-Pinene		0.43	0.47	0.32		0.57	0.81	0.32		0.58	0.63	0.32		1.67	6.92	0.32	NR
β-Pinene		0.57	1.13	0.18		0.38	1.09	0.18		0.24	0.17	0.18		1.66	7.54	0.18	NR
d-Limonene		1.81	2.28	1.27		2.47	5.75	1.27		1.76	2.31	1.27		4.16	9.42	1.27	NR
p-Dichlorobenzene		2.52	9.89	0.72		1.88	3.83	0.72		0.94	1.54	0.72		11.9	58.7	0.72	NR
Carbonyls (passive method)	23				46				39				30				
Formaldehyde		7.83	2.03	8.49		5.18	2.97	4.99		6.14	2.55	6.66		7.29	2.65	8.32	< 0.01
Acetaldehyde		10.3	4.53	10.1		9.47	6.56	9.21		10.5	8.22	6.08		4.80	2.41	3.88	< 0.01
Acetone		4.41	2.61	4.85		2.58	2.27	2.04		4.40	4.19	2.47		3.95	2.94	3.39	0.03[c]
Acrolein		1.57	1.99	0.07		1.01	1.19	0.63		0.38	0.46	0.14		0.84	1.26	0.35	NR
Propionaldehyde		1.95	0.85	1.75		1.46	0.85	1.36		0.51	0.49	0.40		1.16	1.06	0.95	< 0.01
Crotonaldehyde		0.30	0.48	0.07		0.50	1.12	0.21		0.28	0.29	0.19		0.43	0.49	0.27	NR
Benzaldehyde		1.83	1.00	1.59		2.41	1.19	2.41		1.04	0.73	1.10		1.09	1.29	0.68	< 0.01
Hexaldehyde		2.35	0.63	2.33		1.88	0.75	1.79		0.84	0.92	0.58		0.90	0.85	0.77	< 0.01
Glyoxal		2.09	0.70	2.34		1.83	1.04	1.69		1.06	0.57	1.03		1.25	0.59	1.21	< 0.01
Methylglyoxal		2.47	0.83	2.41		2.08	0.89	2.03		0.97	0.80	0.81		1.67	0.93	1.67	< 0.01
$PM_{2.5}$	20	19.2	7.69	17.2	24	19.5	8.18	18.4	34	18.7	11.1	14.9	25	24.7	13.5	22.5	0.05

[a] *n* is the total samples for each compound within a group, unless otherwise noted.

[b] *P* values based on Kruskal-Wallis test for interseason differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.11. Descriptive Summary of Outdoor Air Concentrations (μg/m^3) in Houston by Season[a]

Species	Spring				Summer				Fall				Winter				*P* Value[b]
	n	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	
VOCs	38				76				48				36				
Methylene chloride		0.13	0.08	0.11		0.22	0.19	0.11		0.35	0.19	0.36		0.27	0.22	0.14	NR
MTBE		4.55	4.00	3.72		9.94	15.4	5.09		7.85	6.53	6.36		7.11	10.76	3.42	< 0.01
Chloroform		0.18	0.11	0.17		0.42	2.15	0.15		0.18	0.07	0.15		0.14	0.10	0.12	NR
Carbon tetrachloride		0.64	0.23	0.61		0.60	0.12	0.59		0.64	0.07	0.64		0.68	0.22	0.64	< 0.01
Benzene		2.16	1.84	1.71		2.44	1.91	1.93		3.18	3.01	2.42		1.98	1.39	1.62	< 0.01[c]
Trichloroethylene		0.19	0.16	0.14		0.13	0.04	0.12		0.10	0.08	0.06		0.12	0.06	0.12	NR
Toluene		7.46	5.15	5.70		5.47	3.59	4.35		4.63	2.60	3.80		6.01	4.42	4.46	NR
Tetrachloroethylene		0.16	0.13	0.12		0.22	0.17	0.13		0.28	0.23	0.19		0.21	0.23	0.12	NR
Ethyl benzene		0.78	0.64	0.59		1.02	0.81	0.89		1.14	0.93	0.83		0.70	0.62	0.49	NR
m- & *p*-Xylenes		1.87	0.93	1.64		3.05	2.70	2.28		3.03	1.72	2.53		2.37	2.19	1.52	< 0.01
o-Xylene		0.68	0.34	0.54		1.21	1.10	1.01		1.06	0.62	0.85		0.77	0.67	0.53	< 0.01[c]
Styrene		0.31	0.37	0.23		0.39	0.31	0.28		0.44	0.50	0.17		0.18	0.09	0.16	NR
α-Pinene		0.24	0.20	0.14		0.33	0.34	0.14		0.26	0.30	0.10		0.12	0.16	0.06	NR
β-Pinene		0.23	0.07	0.25		0.38	0.86	0.21		0.36	0.33	0.26		0.32	0.38	0.19	NR
d-Limonene		1.67	3.41	0.86		1.36	3.19	0.94		1.18	0.58	1.20		0.42	0.30	0.25	NR
p-Dichlorobenzene		0.81	0.97	0.61		3.20	19.92	0.30		0.59	0.64	0.39		0.54	1.14	0.17	NR
Carbonyls (passive method)	19				63				35				7				
Formaldehyde		7.39	2.95	7.23		6.27	2.19	6.19		5.64	2.25	5.17		6.82	3.01	5.22	< 0.01
Acetaldehyde		3.86	4.25	2.92		4.20	2.51	3.75		9.50	3.99	11.23		9.22	3.69	10.63	< 0.01
Acetone		10.7	14.46	4.52		5.15	3.54	4.72		56.6	230	15.72		28.1	39.7	7.16	0.03
Acrolein		2.84	1.45	2.92		0.90	1.12	0.34		60.2	341	1.32		0.40	0.32	0.36	0.06[c]
Propionaldehyde		1.99	0.73	1.96		1.05	0.64	1.12		1.93	1.00	1.79		1.47	0.39	1.38	< 0.01
Crotonaldehyde		0.48	0.65	0.07		0.52	0.50	0.40		3.90	17.55	0.40		0.21	0.16	0.23	0.26[c]
Benzaldehyde		2.12	0.87	1.90		1.80	1.10	1.55		2.00	1.14	1.63		1.86	0.78	1.65	< 0.01
Hexaldehyde		3.91	1.12	4.01		2.81	1.02	2.64		3.39	9.95	1.61		1.82	0.57	1.92	< 0.01
Glyoxal		2.01	0.30	1.90		1.86	0.67	1.86		2.10	1.26	1.98		2.27	1.03	2.21	< 0.01
Methylglyoxal		2.43	0.31	2.43		2.34	0.81	2.47		2.20	1.42	1.85		1.52	0.21	1.53	< 0.01
$PM_{2.5}$	23	14.3	5.67	13.5	37	15.4	5.46	15.6	20	17.2	7.49	13.5	30	12.5	3.99	11.7	0.23

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for interseason differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.12. Descriptive Summary of Indoor Air Concentrations (μg/m^3) in Los Angeles by Season[a]

	Spring				Summer				Fall				Winter				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	22				54				55				18				
Methylene chloride		1.99	3.69	0.84		0.91	0.55	0.84		2.21	3.14	1.03		3.27	5.04	1.47	NR
MTBE		23.7	73.3	5.99		12.3	35.5	5.24		13.1	9.65	9.78		8.89	6.47	6.09	< 0.01
Chloroform		1.43	4.19	0.39		1.73	1.98	1.03		1.83	1.59	1.27		1.45	1.36	1.04	< 0.01[c]
Carbon tetrachloride		1.22	2.90	0.59		0.98	2.38	0.62		0.60	0.16	0.57		0.61	0.15	0.56	0.61
Benzene		3.83	9.50	1.66		2.30	6.07	1.37		3.52	1.91	3.27		3.81	2.68	3.34	< 0.01[c]
Trichloroethylene		0.23	0.17	0.14		0.38	1.47	0.11		0.16	0.13	0.10		0.18	0.13	0.12	NR
Toluene		27.2	61.7	11.2		16.6	43.9	7.44		14.3	9.77	11.4		16.0	14.3	12.5	0.08[c]
Tetrachloroethylene		1.69	4.15	0.56		2.50	3.89	1.59		5.24	11.2	2.23		2.18	1.26	1.92	< 0.01[c]
Ethyl benzene		1.74	2.35	1.15		2.13	3.59	1.14		3.25	4.54	2.26		2.59	2.43	1.69	< 0.01[c]
m- & p-Xylenes		5.81	7.57	3.71		7.32	12.60	3.79		8.30	7.68	7.11		9.27	8.95	6.47	< 0.01
o-Xylene		2.03	3.00	1.33		2.61	4.43	1.41		2.77	2.39	2.30		2.88	2.33	2.20	< 0.01
Styrene		0.53	1.01	0.19		0.59	1.15	0.32		2.42	2.88	0.92		0.70	0.56	0.60	NR
α-Pinene		5.28	21.0	0.79		11.0	20.4	1.67		2.93	4.87	0.80		5.15	11.1	1.22	0.02[c]
β-Pinene		4.95	20.8	0.25		2.99	5.36	0.84		1.99	2.23	1.12		5.63	14.0	0.68	0.04[c]
d-Limonene		21.3	40.8	7.10		27.6	57.9	11.4		20.7	45.7	4.48		21.7	25.3	7.18	0.05[c]
p-Dichlorobenzene		3.21	5.67	0.69		90.8	554	1.66		30.2	115	0.72		4.00	5.04	1.16	NR
Carbonyls (passive method)	16				55				46				17				
Formaldehyde		18.8	4.31	18.6		20.4	5.19	18.8		21.2	5.72	19.9		28.7	13.9	25.3	0.17
Acetaldehyde		21.5	8.43	20.0		24.0	15.4	18.8		24.8	11.3	23.1		24.9	15.6	22.5	0.74
Acetone		5.02	2.21	5.07		8.43	10.9	5.23		8.69	6.82	7.15		11.6	14.9	8.57	0.05
Acrolein		0.50	0.55	0.07		1.52	2.57	0.79		0.83	1.41	0.38		1.88	3.61	0.50	0.05[c]
Propionaldehyde		2.40	1.64	2.04		1.98	1.15	1.71		1.65	0.82	1.66		1.93	1.04	1.42	0.49
Crotonaldehyde		0.96	1.63	0.35		0.62	0.63	0.48		0.46	0.45	0.35		0.45	0.53	0.07	0.56[c]
Benzaldehyde		2.41	1.02	2.63		3.00	1.27	2.74		2.99	1.61	3.17		3.63	1.86	3.40	0.13
Hexaldehyde		4.04	1.66	4.16		4.29	1.99	3.50		3.37	1.45	3.41		4.42	1.99	3.61	0.10
Glyoxal		2.69	0.75	2.69		2.45	0.99	2.37		2.81	0.99	2.69		2.65	1.28	2.48	0.43
Methylglyoxal		4.95	7.54	2.93		3.01	0.92	2.90		2.51	0.80	2.43		3.18	1.35	3.06	0.01
$PM_{2.5}$	23	16.7	12.5	14.7	38	17.2	6.23	16.1	37	18.6	10.6	14.8	26	10.8	5.89	9.78	< 0.01

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for interseason differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.13. Descriptive Summary of Indoor Air Concentrations (μg/m^3) in Elizabeth by Season[a]

	Spring				Summer				Fall				Winter				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	37				53				56				36				
Methylene chloride		2.33	8.38	0.84		1.44	2.20	0.84		1.61	2.52	0.84		1.23	1.02	0.84	NR
MTBE		8.31	12.9	4.37		6.09	8.92	4.13		7.82	6.90	5.81	35	7.63	10.3	5.03	0.14[c]
Chloroform		1.88	2.20	0.84		2.20	5.78	0.43		1.31	1.63	0.76		1.09	1.48	0.59	0.33[c]
Carbon tetrachloride		0.59	0.20	0.57		0.69	0.35	0.63		0.67	0.26	0.69		0.64	0.37	0.64	0.17
Benzene		2.64	5.03	1.28		1.52	1.53	1.07		2.82	2.83	2.22		3.38	6.09	2.11	< 0.01[c]
Trichloroethylene		0.62	0.68	0.38		0.91	2.13	0.45		1.38	3.83	0.59		0.68	1.33	0.10	0.26[c]
Toluene		17.1	15.7	11.6		10.7	12.6	8.13		12.4	8.00	11.0		11.6	8.94	8.41	0.02[c]
Tetrachloroethylene		1.03	0.98	0.56		1.03	0.79	0.56		1.82	3.28	1.22		0.94	1.02	0.56	NR
Ethyl benzene		3.82	11.2	1.02		1.35	1.58	0.86		1.92	1.46	1.64		2.75	5.31	1.42	0.03[c]
m- & p-Xylenes		12.4	37.9	3.99		3.15	3.43	2.37		5.53	4.73	4.43		6.91	16.7	3.02	< 0.01
o-Xylene		3.61	10.9	1.11		1.30	1.48	0.84		2.10	1.45	1.68		2.10	3.82	1.18	< 0.01
Styrene		1.25	3.93	0.17		0.99	3.66	0.17		2.11	5.25	0.47		1.03	2.40	0.27	NR
α-Pinene		4.05	10.1	1.27		3.17	5.27	1.27		3.09	3.09	2.41		6.24	21.0	1.36	0.08[c]
β-Pinene		0.78	1.19	0.18		1.90	3.22	0.82		2.64	7.45	0.80		8.66	22.4	0.71	0.01[c]
d-Limonene		11.0	17.5	5.02		13.2	23.3	3.26		16.7	25.2	8.28		17.8	30.7	8.08	0.12[c]
p-Dichlorobenzene		27.2	69.8	3.03		50.5	200	0.72		15.0	56.7	0.72		22.2	73.3	2.11	NR
Carbonyls (passive method)	23				46				39				31				
Formaldehyde		25.3	8.62	25.4		20.9	5.16	20.2		24.0	6.82	22.3		20.4	7.80	19.4	< 0.01
Acetaldehyde		16.0	4.66	14.9		15.6	7.04	14.2		18.5	13.1	15.2		14.9	7.25	13.9	0.50
Acetone		9.45	9.61	6.60		8.85	9.32	6.09		7.23	5.23	5.70		22.8	31.3	10.7	< 0.01
Acrolein		1.39	2.13	0.07		1.17	1.44	0.69		0.51	0.54	0.32		0.90	0.96	0.63	0.14[c]
Propionaldehyde		2.38	1.29	2.22		1.98	2.79	1.46		0.94	0.69	0.96		3.01	5.38	1.73	< 0.01
Crotonaldehyde		1.21	1.60	0.07		0.75	0.86	0.50		0.47	0.44	0.32		0.50	0.57	0.33	0.59[c]
Benzaldehyde		2.54	0.93	2.39		2.92	1.15	2.76		3.33	1.30	3.59		3.27	1.67	3.30	0.07
Hexaldehyde		5.26	2.76	4.11		4.19	2.37	3.55		2.65	1.80	2.45		3.63	1.67	3.15	< 0.01
Glyoxal		2.78	0.54	2.81		2.14	1.14	2.21		2.10	0.84	2.11		2.79	0.79	2.40	< 0.01
Methylglyoxal		2.78	0.72	2.66		2.39	0.92	2.60		1.77	1.07	1.90		2.63	1.00	2.62	< 0.01
$PM_{2.5}$	20	24.7	20.4	18.0	23	16.4	7.40	16.2	29	18.7	15.8	13.9	24	21.5	16.1	14.1	0.46

[a] *n* is the total samples for each compound within a group, unless otherwise noted.

[b] *P* values based on Kruskal-Wallis test for interseason differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.14. Descriptive Summary of Indoor Air Concentration (μg/m^3) in Houston by Season[a]

	Spring				Summer				Fall				Winter				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	38				76				48				36				
Methylene chloride		1.48	3.87	0.23		6.00	26.6	0.49		2.14	5.20	0.44		1.96	6.63	0.44	NR
MTBE		11.6	38.5	4.51		21.0	41.8	6.13		11.4	11.3	7.42		8.86	9.81	4.93	< 0.01
Chloroform		1.86	2.88	0.90		3.12	3.44	2.14		2.04	3.15	1.16		1.43	1.77	0.81	< 0.01[c]
Carbon tetrachloride		0.64	0.19	0.64		0.71	0.36	0.64		0.64	0.22	0.60		0.71	0.23	0.64	0.27
Benzene		3.56	3.03	2.39		5.34	6.45	3.21		6.11	8.07	3.91		3.47	2.25	2.82	0.09
Trichloroethylene		0.17	0.07	0.13		0.19	0.28	0.12		0.14	0.28	0.07		0.19	0.32	0.12	NR
Toluene		18.7	32.0	6.23		20.5	28.6	10.4		13.7	8.07	12.3		10.9	8.23	8.09	0.12[c]
Tetrachloroethylene		0.91	2.74	0.13		1.17	2.50	0.34		0.66	1.04	0.25		1.32	3.15	0.30	NR
Ethyl benzene		2.87	5.30	1.44		2.98	3.48	1.87		2.36	2.79	1.74		2.84	7.66	1.30	0.09[c]
m- & *p*-Xylenes		8.63	20.19	3.99		9.12	13.0	4.94		6.48	7.35	5.25		9.61	28.3	4.30	0.13
o-Xylene		2.68	5.23	1.21		3.17	4.13	1.80		2.01	1.94	1.64		3.21	9.59	1.38	0.08
Styrene		2.19	7.32	0.87		2.24	7.41	0.92		0.87	0.83	0.59		0.50	0.52	0.41	< 0.01[c]
α-Pinene		7.18	15.14	1.24		7.33	20.7	0.87		2.04	3.50	0.10		0.97	1.68	0.08	< 0.01
β-Pinene		7.39	8.49	3.26		9.16	13.9	5.04		6.77	7.18	3.97		6.97	13.8	2.34	0.08
d-Limonene		96.7	337	26.8		31.7	38.7	20.0		71.2	157	26.6		32.9	55.4	14.0	0.32
p-Dichlorobenzene		143	370	1.89		134	422	2.42		124	376	3.91		124	372	1.30	NR
Carbonyls (passive method)	20				64				34				7				
Formaldehyde		20.4	3.61	19.3		20.3	5.70	20.2		23.0	9.64	22.6		16.7	3.20	15.7	0.18
Acetaldehyde		26.6	7.27	24.3		35.9	35.7	22.3		21.5	9.58	18.7		26.2	16.1	21.1	0.04
Acetone		28.5	33.2	16.1		15.6	25.2	9.85		29.6	36.3	18.6		32.9	37.1	17.1	< 0.01
Acrolein		2.08	1.34	2.07		1.29	1.58	0.68		7.50	25.1	1.44		0.89	0.66	0.71	0.02[c]
Propionaldehyde		2.51	0.90	2.50		2.19	0.92	2.33		2.35	0.93	2.47		2.14	0.65	2.31	0.49
Crotonaldehyde		0.81	1.21	0.21		0.66	0.59	0.56		1.22	1.19	0.89		0.61	0.20	0.52	0.25[c]
Benzaldehyde		3.24	1.27	2.79		2.82	1.15	2.58		3.19	1.52	2.82		3.34	1.02	3.02	0.19
Hexaldehyde		6.54	2.11	6.21		6.54	3.40	6.07		4.69	3.62	3.69		5.00	4.35	2.41	< 0.01
Glyoxal		2.63	0.41	2.66		2.75	0.77	2.69		3.05	0.91	3.02		2.80	0.98	2.69	0.29
Methylglyoxal		3.10	0.47	3.05		3.03	0.88	3.12		3.76	1.20	3.65		2.56	0.81	2.60	0.01
$PM_{2.5}$	24	17.0	13.2	12.6	37	16.1	15.3	10.8	19	20.9	11.6	16.1	26	16.0	8.77	13.3	0.07

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for interseason differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.15. Descriptive Summary of Adult Personal Concentrations (μg/m^3) in Los Angeles by Season[a]

	Spring				Summer				Fall				Winter				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	22				55				53				18				
Methylene chloride		1.97	2.98	0.84		1.09	1.47	0.84		8.82	45.1	1.29		2.88	3.71	1.79	< 0.01
MTBE		15.5	28.0	6.57		10.1	10.1	7.92		14.8	10.4	11.4		10.1	7.58	6.97	0.01
Chloroform		56.2	261	0.57		1.96	3.00	0.92		1.72	1.55	1.37		1.36	1.24	0.79	0.01
Carbon tetrachloride		2.48	8.89	0.59		0.65	0.23	0.62		0.59	0.15	0.57		0.60	0.15	0.58	0.62
Benzene		5.89	17.8	2.08		1.76	1.19	1.53		3.63	1.93	3.55		3.69	2.19	3.14	< 0.01
Trichloroethylene		1.25	3.36	0.28		0.30	0.60	0.11		0.24	0.45	0.11		0.17	0.10	0.13	< 0.01
Toluene		44.2	134	16.8		10.1	8.10	7.01		17.2	15.8	12.9		21.5	16.5	15.6	< 0.01
Tetrachloroethylene		4.51	16.9	0.57		2.04	1.61	1.61		6.21	13.1	2.23		2.05	1.08	2.19	< 0.01
Ethyl benzene		2.72	6.56	1.16		1.57	1.27	1.27		3.13	4.48	2.28		2.48	1.63	2.05	< 0.01
m- & *p*-Xylenes		9.46	21.3	4.26		5.37	4.53	4.34		9.44	8.61	7.66		9.14	6.29	7.52	< 0.01
o-Xylene		3.38	8.01	1.46		1.93	1.44	1.64		3.20	2.71	2.57		2.90	1.80	2.63	< 0.01
Styrene		0.48	0.44	0.37		0.51	0.77	0.19		2.35	2.90	0.95		0.66	0.40	0.57	< 0.01
α-Pinene		3.87	11.8	0.85		6.16	9.97	1.47		2.27	4.99	0.32		2.70	4.82	0.84	< 0.01
β-Pinene		1.49	3.16	0.60		2.43	5.04	0.73		3.43	12.6	1.27		5.56	12.2	0.78	0.45
d-Limonene		247	1087	8.10		21.3	42.6	9.19		18.5	42.8	5.21		29.2	53.6	8.63	0.09
p-Dichlorobenzene		6.18	18.0	0.71		15.6	49.8	1.88		28.0	102	0.72		3.14	3.42	1.16	0.70
Carbonyls (passive method)	26				56				44				16				
Formaldehyde		21.3	6.94	20.2		22.3	5.57	20.7		21.6	5.76	20.5		20.7	10.1	18.3	0.76
Acetaldehyde		23.8	6.89	24.1		23.3	13.2	18.3		21.1	9.84	18.5		23.4	13.2	24.4	0.08
Acetone		6.26	3.13	6.46		8.48	11.3	5.17		9.85	8.39	7.35		13.3	18.2	7.68	0.12
Acrolein		0.96	1.27	0.46		1.46	1.69	0.79		0.67	0.96	0.29		1.42	2.76	0.15	0.06[c]
Propionaldehyde		2.83	0.92	2.80		2.02	0.79	2.09		1.94	1.34	1.98		2.03	1.63	1.66	< 0.01
Crotonaldehyde		0.75	0.94	0.13		0.65	0.60	0.46		0.53	0.77	0.28		0.69	0.55	0.72	0.20[c]
Benzaldehyde		2.86	1.19	2.69		3.07	1.10	2.97		3.49	1.62	3.46		3.50	2.48	3.18	0.17
Hexaldehyde		5.04	1.80	4.74		4.51	1.67	4.10		3.36	1.21	3.20		4.76	3.04	4.14	< 0.01
Glyoxal		2.82	1.01	2.53		2.41	0.93	2.49		3.01	1.15	2.79		2.33	1.47	2.06	0.02
Methylglyoxal		3.30	0.83	3.28		2.94	1.12	2.88		2.59	1.09	2.29		2.85	1.28	2.37	0.01
$PM_{2.5}$	24	28.0	12.5	27.1	29	29.2	13.4	27.4	26	36.1	18.3	32.6	26	23.3	12.1	18.8	< 0.01

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for interseason differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.16. Descriptive Summary of Adult Personal Concentrations (μg/m^3) in Elizabeth by Season[a]

	Spring				Summer				Fall				Winter				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	32				51				55				33				
Methylene chloride		1.99	6.04	0.84		1.74	4.34	0.84		1.67	2.71	0.84		1.10	0.60	0.84	0.44
MTBE		9.62	8.91	7.61		7.30	10.8	4.67		25.5	113	6.01	32	13.1	23.2	5.12	0.15
Chloroform		2.00	1.81	1.64	50	2.65	7.02	0.73	53	1.91	2.55	1.05		1.24	1.46	0.56	0.10
Carbon tetrachloride		0.66	0.27	0.68		1.40	5.40	0.62		0.70	0.35	0.68		0.56	0.31	0.55	0.13
Benzene		2.75	4.25	1.72		1.71	2.28	1.21		3.46	4.81	2.18		3.46	5.02	2.11	< 0.01
Trichloroethylene		0.74	1.41	0.43		5.44	28.3	0.50		1.31	2.45	0.68		1.04	3.39	0.10	0.01
Toluene		27.0	49.2	16.1		18.8	46.0	8.58		20.0	32.5	12.7		19.0	20.9	11.6	0.03
Tetrachloroethylene		1.27	1.52	0.56		53.1	366	0.56		3.07	8.69	1.34		1.23	1.60	0.56	0.01
Ethyl benzene		4.07	11.2	1.24		1.92	3.99	1.01		3.15	7.39	1.85		2.90	3.17	1.61	< 0.01
m- & p-Xylenes		13.2	38.2	4.17		4.73	8.03	2.81		8.22	18.16	5.63		7.27	9.22	3.70	0.01
o-Xylene		3.88	10.9	1.52		2.12	4.00	1.12		3.61	10.51	2.00		2.86	3.90	1.36	0.01
Styrene		1.61	3.81	0.38		1.12	3.29	0.17		2.84	8.66	0.48		1.02	2.22	0.35	0.30
α-Pinene		3.66	7.53	1.52		4.07	7.55	1.21		3.68	3.80	2.63		10.2	35.6	2.48	0.07
β-Pinene		0.76	0.78	0.49		3.32	6.85	0.81		3.82	10.33	1.30		14.1	32.1	1.66	< 0.01
d-Limonene		15.4	26.5	7.79		13.8	24.3	5.96		19.1	26.9	10.7		25.3	50.4	12.4	0.06
p-Dichlorobenzene		44.2	128	2.97		41.0	173	0.72		14.3	51.2	1.57		7.37	13.5	2.48	0.09
Carbonyls (passive method)	18				43				49				32				
Formaldehyde		26.1	6.51	24.8		21.3	5.72	19.6		22.3	6.58	21.3		19.9	8.24	17.9	< 0.01
Acetaldehyde		17.1	3.53	17.5		17.5	8.93	15.2		19.4	11.1	16.1		14.1	7.80	11.5	0.03
Acetone		17.4	24.0	9.10		29.2	111	7.57		41.2	194	7.00		17.9	25.2	10.3	0.22
Acrolein		0.66	1.30	0.07		0.70	0.97	0.29		0.50	1.01	0.07		1.21	1.12	0.99	NR
Propionaldehyde		2.38	1.09	2.23		1.47	0.72	1.53		1.57	1.07	1.34		1.78	0.91	1.51	0.01
Crotonaldehyde		0.87	1.16	0.07		0.56	0.92	0.30		0.69	1.35	0.28		0.48	0.54	0.33	NR
Benzaldehyde		2.94	1.04	2.73		3.37	1.37	3.24		3.74	2.00	3.47		2.87	1.68	2.89	0.13
Hexaldehyde		5.78	3.72	4.71		4.36	1.87	3.86		3.42	2.31	2.98		3.85	1.87	4.00	< 0.01
Glyoxal		2.85	0.66	2.77		2.26	0.87	2.20		2.26	0.71	2.16		2.40	0.83	2.25	0.01
Methylglyoxal		2.82	0.70	2.55		2.48	0.76	2.49		1.90	0.92	2.01		2.40	1.02	2.06	< 0.01
$PM_{2.5}$	15	50.6	33.2	34.2	19	42.0	19.3	39.5	27	41.9	29.9	29.7	16	47.5	37.9	36.3	0.36

[a] *n* is the total samples for each compound within a group, unless otherwise noted.

[b] *P* values based on Kruskal-Wallis test for interseason differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

Table E.17. Descriptive Summary of Adult Personal Concentrations in Houston by Season[a]

	Spring				Summer				Fall				Winter				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	38				76				50				35				
Methylene chloride		1.54	4.38	0.29		6.74	24.5	0.52		1.79	3.94	0.50		1.72	4.40	0.51	0.30
MTBE		13.3	36.2	4.97		23.4	44.3	8.57		16.4	22.0	8.45		8.84	8.15	5.60	0.01
Chloroform		1.98	3.02	1.07		3.03	3.55	1.99		1.94	2.34	1.30		1.39	1.70	0.80	0.01
Carbon tetrachloride		0.64	0.15	0.61		0.70	0.31	0.64		0.64	0.14	0.62		0.68	0.21	0.62	0.91
Benzene		3.57	2.78	2.53		5.33	5.25	3.21		6.11	6.02	4.14		3.22	1.78	2.86	0.01
Trichloroethylene		0.26	0.54	0.15		0.20	0.47	0.12		0.11	0.07	0.09		0.32	1.02	0.12	< 0.01
Toluene		18.5	24.1	12.5		20.4	23.5	12.3		15.3	7.99	13.6		16.2	19.7	13.4	0.69
Tetrachloroethylene		0.83	2.02	0.28		2.08	7.26	0.39		0.75	1.06	0.36		1.38	2.64	0.40	0.04
Ethyl benzene		2.86	3.99	1.83		2.98	2.99	1.90		4.00	6.92	2.02		2.10	2.63	1.42	0.03
m- & *p*-Xylenes		8.20	14.8	3.84		9.05	10.63	5.86		11.4	21.6	5.94		6.73	9.64	4.11	0.05
o-Xylene		2.63	3.76	1.36		3.21	3.86	1.89		3.49	6.00	1.99		2.34	3.45	1.67	0.04
Styrene		1.86	5.09	0.82		2.06	6.03	1.04		1.48	2.91	0.86		0.56	0.32	0.47	< 0.01
α-Pinene		3.87	9.27	0.10		5.42	7.71	0.75		2.89	4.39	1.23		1.65	2.99	0.09	0.01
β-Pinene		8.73	12.49	3.38		9.26	11.7	5.54		6.68	6.69	4.31		7.82	20.5	2.83	0.04
d-Limonene		85.0	270	34.0		34.4	41.2	20.0		74.1	179	30.3		39.7	60.3	17.5	0.26
p-Dichlorobenzene		134	348	2.80		122	361	3.61		127	396	5.43		88.5	267	1.83	0.92
Carbonyls (passive method)	20				62				37				6				
Formaldehyde		20.7	6.20	20.4		21.7	16.7	20.5		22.9	8.21	21.3		15.1	2.21	14.6	0.05
Acetaldehyde		26.9	8.27	26.4		32.3	25.0	22.1		27.2	16.7	21.0		21.1	7.20	22.5	0.46
Acetone		17.9	18.8	11.6		15.7	23.7	9.13		98.8	260	22.3		26.9	26.6	12.5	< 0.01
Acrolein		1.73	0.95	1.69		1.25	1.62	0.71		132	448	2.40		0.41	0.37	0.30	< 0.01[c]
Propionaldehyde		2.77	1.03	2.57		1.97	0.94	2.05		2.27	1.42	2.20		1.89	0.59	1.95	0.07
Crotonaldehyde		0.86	0.96	0.49		0.81	0.77	0.63		6.86	20.3	0.87		0.50	0.38	0.53	0.32[c]
Benzaldehyde		3.38	1.54	2.96		3.33	3.04	2.49		4.06	2.76	2.96		3.33	0.75	3.24	0.09
Hexaldehyde		7.60	4.07	6.24		6.36	3.45	5.94		10.4	21.5	3.55		5.17	3.37	3.46	< 0.01
Glyoxal		2.73	0.46	2.73		2.56	0.84	2.53		3.57	3.29	2.64		3.19	0.93	3.37	0.26
Methylglyoxal		3.06	0.39	3.03		2.77	0.89	2.73		3.70	1.28	3.35		3.10	0.92	3.36	< 0.01
$PM_{2.5}$	21	33.3	20.5	29.6	30	28.9	17.0	26.2	23	53.9	31.8	42.5	24	34.9	17.1	32.8	0.01

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for interseason differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.18. Descriptive Summary of Indoor Concentrations (μg/m^3) in Los Angeles by Type of Home[a]

	Single-Family House				Multiple-Family House or Apartment				Mobile Home or Trailer				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	81				88				5				
Methylene chloride		2.04	3.45	0.84		1.48	2.18	0.84		0.74	0.31	0.84	NR
MTBE		15.9	46.1	7.43		11.2	14.4	7.56		4.86	2.16	4.65	0.3
Chloroform		1.04	2.27	0.55		2.12	1.92	1.43		0.74	0.51	0.59	< 0.01[c]
Carbon tetrachloride		0.94	2.46	0.54		0.67	0.23	0.62		0.68	0.22	0.57	< 0.01
Benzene		3.20	6.91	1.97		2.87	2.36	2.21		1.93	0.95	1.34	0.49[c]
Trichloroethylene		0.32	1.21	0.11		0.20	0.18	0.11		0.15	0.08	0.13	NR
Toluene		17.6	47.6	8.22		15.6	12.5	12.9		15.2	11.3	12.4	0.02[c]
Tetrachloroethylene		2.63	4.26	1.26		4.06	9.02	2.04		0.56	0.26	0.56	< 0.01[c]
Ethyl benzene		2.11	3.10	1.25		2.79	3.89	1.83		2.03	2.36	1.05	0.04[c]
m- & *p*-Xylenes		6.38	10.5	4.02		7.58	8.32	5.11		2.64	0.78	2.46	0.04
o-Xylene		2.32	3.73	1.58		2.64	2.55	2.06		1.23	0.84	0.89	0.03
Styrene		1.12	2.14	0.30		1.39	2.05	0.65		0.39	0.32	0.20	NR
α-Pinene		4.24	16.5	0.78		7.64	13.0	2.10		1.28	1.93	0.32	< 0.01[c]
β-Pinene		2.82	11.1	0.55		3.18	7.41	1.10		1.93	2.47	1.20	0.01[c]
d-Limonene		16.9	36.7	4.93		27.2	52.2	10.3		10.6	3.59	9.01	0.05[c]
p-Dichlorobenzene		69.9	458	0.72		12.3	59.9	1.53		0.93	0.46	0.72	NR
Carbonyls (passive method)	66				65				3				
Formaldehyde		21.2	7.62	19.0		21.4	6.42	19.0		30.3	19.0	24.9	0.66
Acetaldehyde		21.4	13.2	17.8		27.3	12.9	24.6		15.0	10.8	17.3	< 0.01
Acetone		6.96	8.56	5.04		9.29	8.13	6.87		25.8	34.5	7.26	0.02
Acrolein		0.99	2.03	0.41		1.42	2.54	0.50		1.44	1.11	1.25	0.27[c]
Propionaldehyde		2.06	1.27	1.72		1.81	0.92	1.72		0.81	0.70	1.04	0.10
Crotonaldehyde		0.50	0.60	0.25		0.60	0.57	0.48		2.14	3.59	0.07	0.30[c]
Benzaldehyde		2.67	1.49	2.64		3.27	1.39	3.13		4.46	0.36	4.45	0.01
Hexaldehyde		3.38	1.46	3.37		4.54	1.93	4.23		4.22	2.90	2.83	< 0.01
Glyoxal		2.66	0.92	2.64		2.62	1.07	2.37		2.05	1.79	2.59	0.87
Methylglyoxal		3.44	3.82	2.83		2.79	0.93	2.84		1.85	1.53	2.44	0.34
$PM_{2.5}$	70	16.4	9.40	14.5	34	16.4	11.2	14.4	3	16.7	4.69	18.3	0.02

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for type-of-home differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.19. Descriptive Summary of Indoor Concentrations (μg/m^3) in Elizabeth by Type of Home[a]

	Single-Family House				Multiple-Family House or Apartment				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	45				125				
Methylene chloride		1.39	2.48	0.84		1.74	4.81	0.84	NR
MTBE		4.85	4.73	4.13	124	8.58	11.00	5.74	0.05[c]
Chloroform		1.12	1.72	0.53		1.77	3.92	0.84	0.11[c]
Carbon tetrachloride		0.68	0.28	0.66		0.65	0.32	0.63	0.38
Benzene		1.64	1.35	1.30		2.89	4.67	1.87	0.24[c]
Trichloroethylene		0.54	0.69	0.29		1.13	2.96	0.45	0.12[c]
Toluene		11.0	10.1	8.41		13.3	12.4	10.1	0.47[c]
Tetrachloroethylene		1.14	0.93	0.56		1.31	2.33	0.56	NR
Ethyl benzene		2.03	4.75	0.91		2.49	6.28	1.42	0.21[c]
m- & *p*-Xylenes		5.81	15.0	2.59		6.97	21.1	3.29	0.35
o-Xylene		1.84	3.36	1.10		2.37	6.09	1.23	0.3
Styrene		0.99	2.78	0.17		1.39	4.01	0.17	NR
α-Pinene		2.81	4.67	1.45		4.29	12.7	1.41	0.78[c]
β-Pinene		2.77	6.72	0.76		3.58	12.9	0.62	0.85[c]
d-Limonene		11.9	19.0	6.18		15.9	26.9	6.68	0.81[c]
p-Dichlorobenzene		16.5	66.0	0.72		35.5	140	1.94	NR
Carbonyls (passive method)	36				95				
Formaldehyde		22.2	6.04	22.0		22.5	7.49	21.0	0.86
Acetaldehyde		15.5	5.76	14.5		16.7	10.1	14.2	0.87
Acetone		9.80	8.25	7.71		12.6	20.2	6.41	0.49
Acrolein		0.97	1.49	0.56		0.98	1.30	0.49	0.93[c]
Propionaldehyde		1.72	2.02	1.59		2.11	3.57	1.41	0.71
Crotonaldehyde		0.85	1.06	0.56		0.64	0.88	0.32	0.11[c]
Benzaldehyde		3.04	1.13	3.39		3.09	1.40	3.18	0.93
Hexaldehyde		3.08	1.87	2.85		4.06	2.45	3.40	0.01
Glyoxal		2.28	0.99	2.39		2.38	0.95	2.29	0.78
Methylglyoxal		2.30	1.04	2.43		2.34	1.03	2.50	0.92
$PM_{2.5}$	20	19.1	15.0	14.6	52	19.2	16.0	14.6	0.43

[a] *n* is the total samples for each compound within a group, unless otherwise noted.

[b] *P* values based on Kruskal-Wallis test for type-of-home differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

Table E.20. Descriptive Summary of Indoor Air Concentrations (μg/m^3) in Houston by Type of Home[a]

	Single-Family House				Multiple-Family House or Apartment				Mobile Home or Trailer				
Species	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median	*P* Value[b]
VOCs	128				5				48				
Methylene chloride		3.60	17.3	0.53		8.16	17.3	0.46		3.70	19.50	0.23	NR
MTBE		19.5	38.6	7.00		3.10	1.95	3.71		5.20	4.09	4.24	< 0.01
Chloroform		2.11	2.46	1.34		2.54	0.81	2.48		3.06	4.63	1.01	0.38[c]
Carbon tetrachloride		0.68	0.24	0.62		0.77	0.30	0.77		0.69	0.38	0.64	0.9
Benzene		5.56	7.02	3.48		1.57	0.43	1.55		3.63	2.46	2.55	0.03
Trichloroethylene		0.18	0.30	0.12		0.21	0.06	0.23		0.17	0.18	0.12	NR
Toluene		15.9	18.8	11.1		24.2	27.9	13.9		19.8	34.7	6.80	0.67[c]
Tetrachloroethylene		1.17	2.81	0.29		1.85	1.25	1.80		0.61	1.57	0.15	NR
Ethyl benzene		2.81	4.68	1.82		1.84	0.53	1.95		3.16	5.74	1.46	0.55[c]
m- & *p*-Xylenes		8.69	16.4	5.39		4.95	1.33	4.73		9.59	22.6	2.79	< 0.01
o-Xylene		2.92	5.52	1.83		1.72	0.37	1.77		3.04	6.23	1.08	0.01
Styrene		1.19	5.22	0.58		1.28	0.62	1.33		2.92	7.55	1.07	< 0.01[c]
α-Pinene		3.98	6.16	0.75		1.34	2.78	0.13		9.03	28.0	0.27	0.33
β-Pinene		7.06	10.7	3.73		13.4	9.50	12.0		9.55	14.7	3.72	0.18
d-Limonene		58.4	208	18.6		36.0	26.3	23.5		52.9	63.3	29.4	0.3
p-Dichlorobenzene		128	412	1.89		58.1	25.1	52.3		175	406	6.15	NR
Carbonyls (passive method)	82				2				31				
Formaldehyde		21.3	5.90	21.3		14.2	0.11	14.2		19.0	5.52	18.4	0.06
Acetaldehyde		29.5	27.7	21.4		48.9	43.0	48.9		31.9	28.5	24.2	0.28
Acetone		20.4	27.2	13.1		14.3	2.52	14.3		24.3	37.5	12.4	0.97
Acrolein		3.98	16.3	1.08		0.27	0.28	0.27		1.47	1.70	0.73	0.17[c]
Propionaldehyde		2.33	0.84	2.38		1.19	0.04	1.19		2.13	1.04	2.31	0.17
Crotonaldehyde		1.03	1.02	0.67		0.26	0.27	0.26		0.36	0.32	0.28	< 0.01[c]
Benzaldehyde		2.92	0.96	2.72		2.35	0.21	2.35		3.16	1.58	2.68	0.64
Hexaldehyde		5.53	2.15	5.57		5.06	0.30	5.06		6.90	4.71	6.11	0.67
Glyoxal		2.81	0.77	2.76		2.03	1.12	2.03		2.79	0.78	2.69	0.52
Methylglyoxal		3.22	1.06	3.15		2.41	1.61	2.41		3.27	0.81	3.32	0.62
$PM_{2.5}$	46	14.9	11.6	11.4	2	8.21	4.82	8.21	24	19.6	12.2	14.6	< 0.01

[a] *n* is the total samples for each compound within a group.

[b] *P* values based on Kruskal-Wallis test for type-of-home differences in medians. $P < 0.05$ indicates the difference is significant at $\alpha = 0.05$. NR indicates not reported either because the proportion of nondetected samples was > 40% or because the sample size was too small.

[c] Proportion of nondetected samples was between 10% and 40%.

APPENDIX F. Scatter Plots of Measured Concentrations

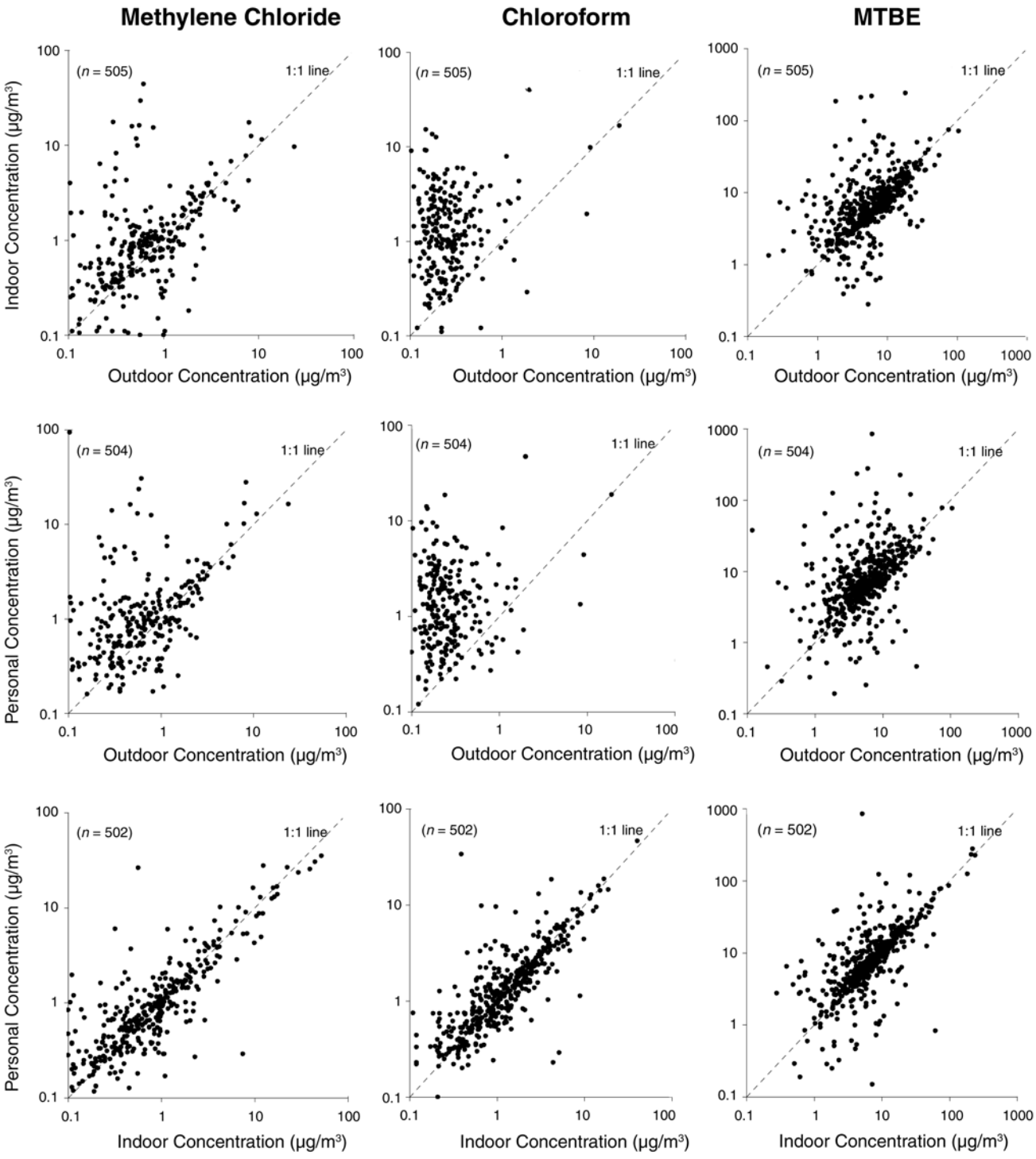

Figure F.1. Concentrations of each of 16 VOCs to demonstrate indoor–outdoor, personal–outdoor, and personal–indoor comparisons. Each compound is identified at the top of the column. All comparisons are shown on logarithmic scales; note that some axis scales may differ within a column.

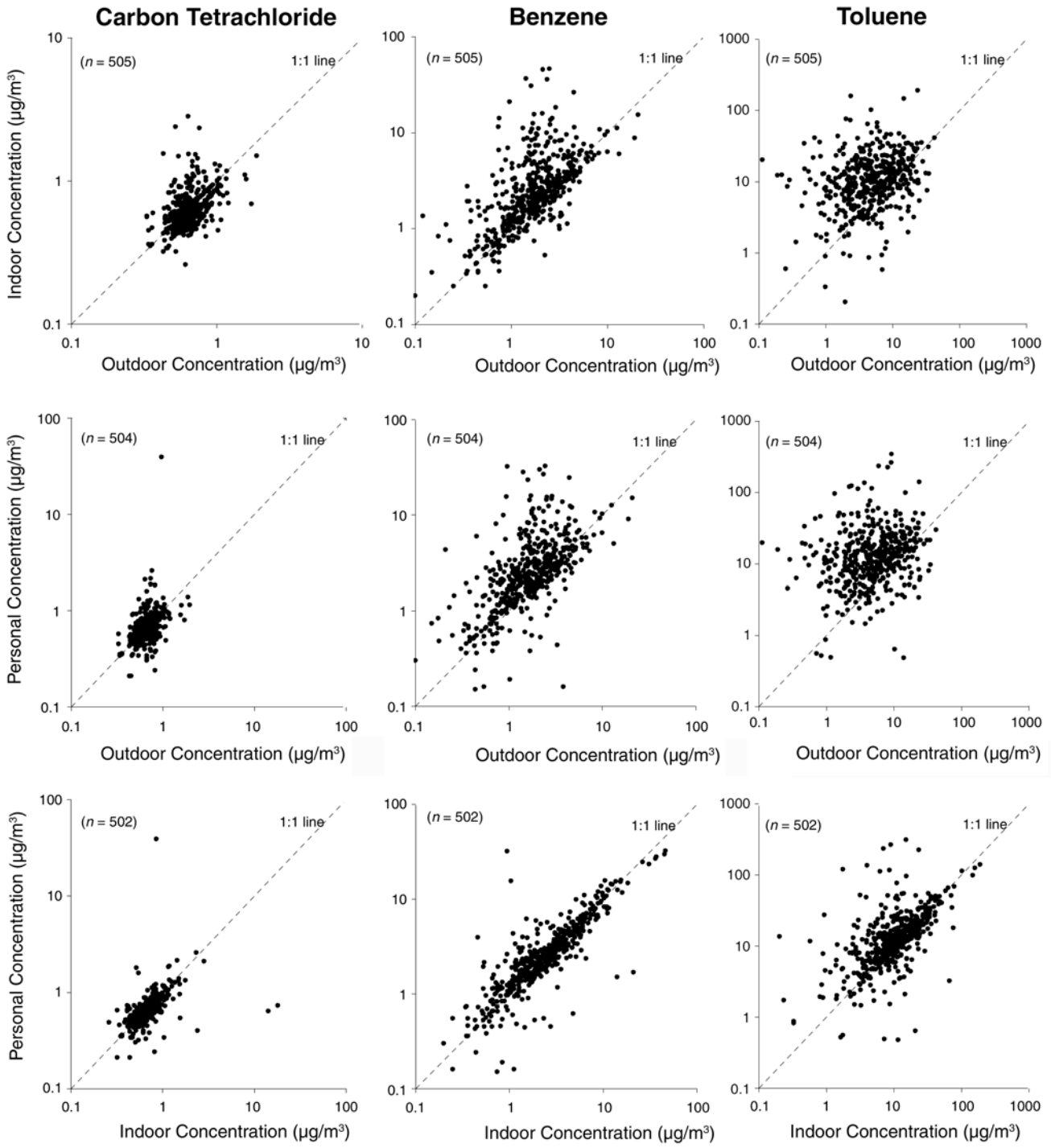

Figure F.1. ***Continued.***

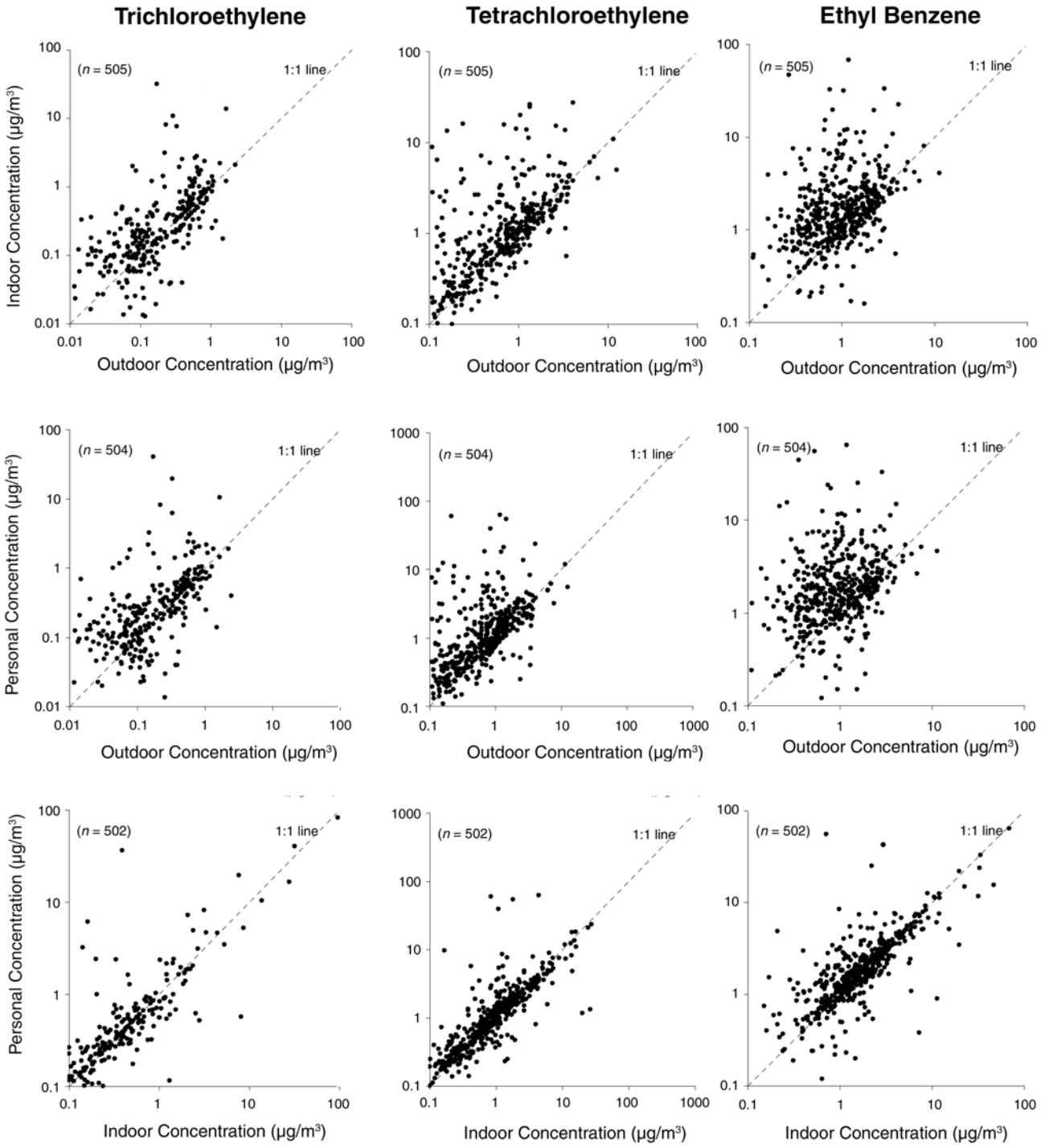

Figure F.1. ***Continued.***

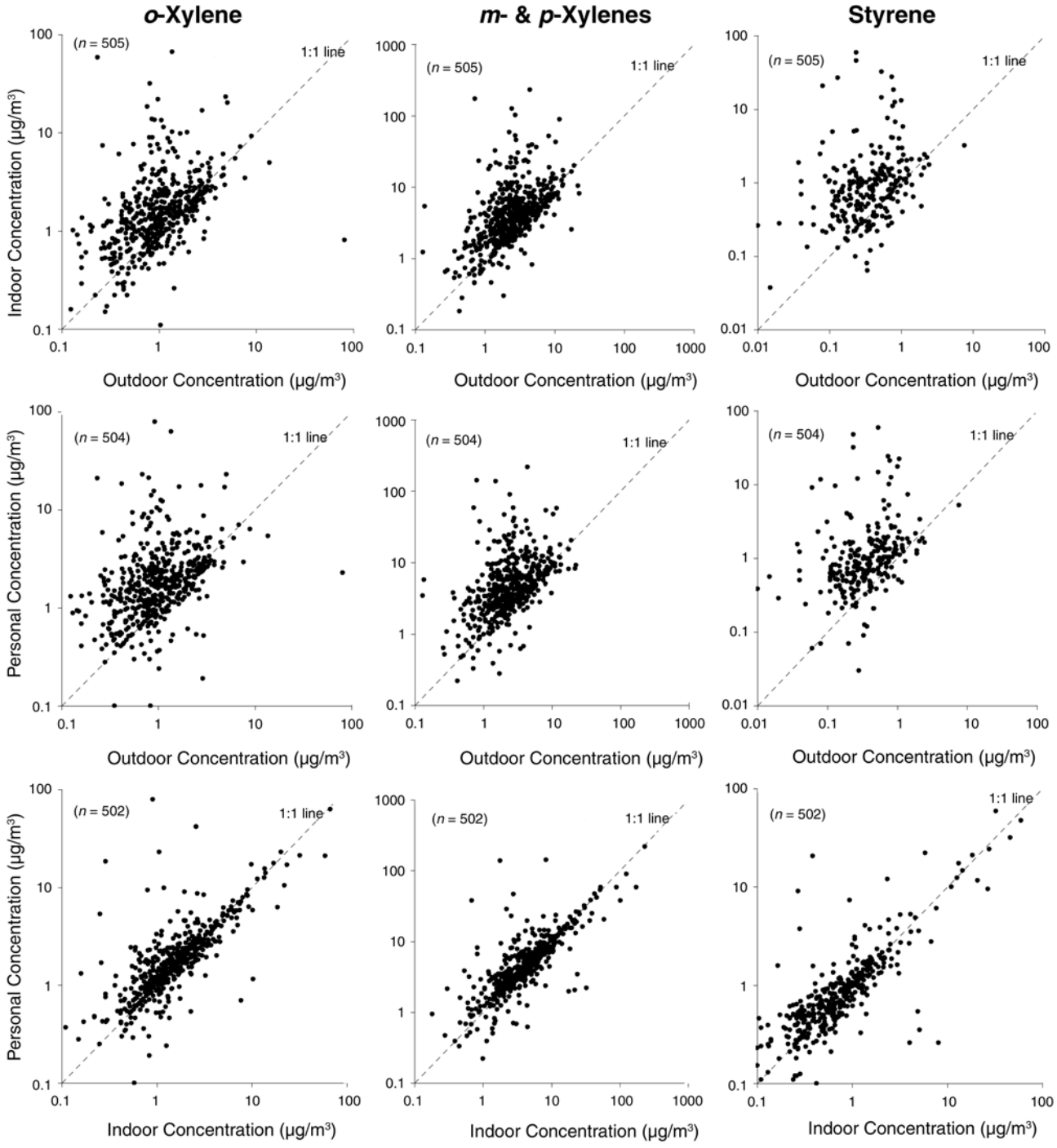

Figure F.1. ***Continued.***

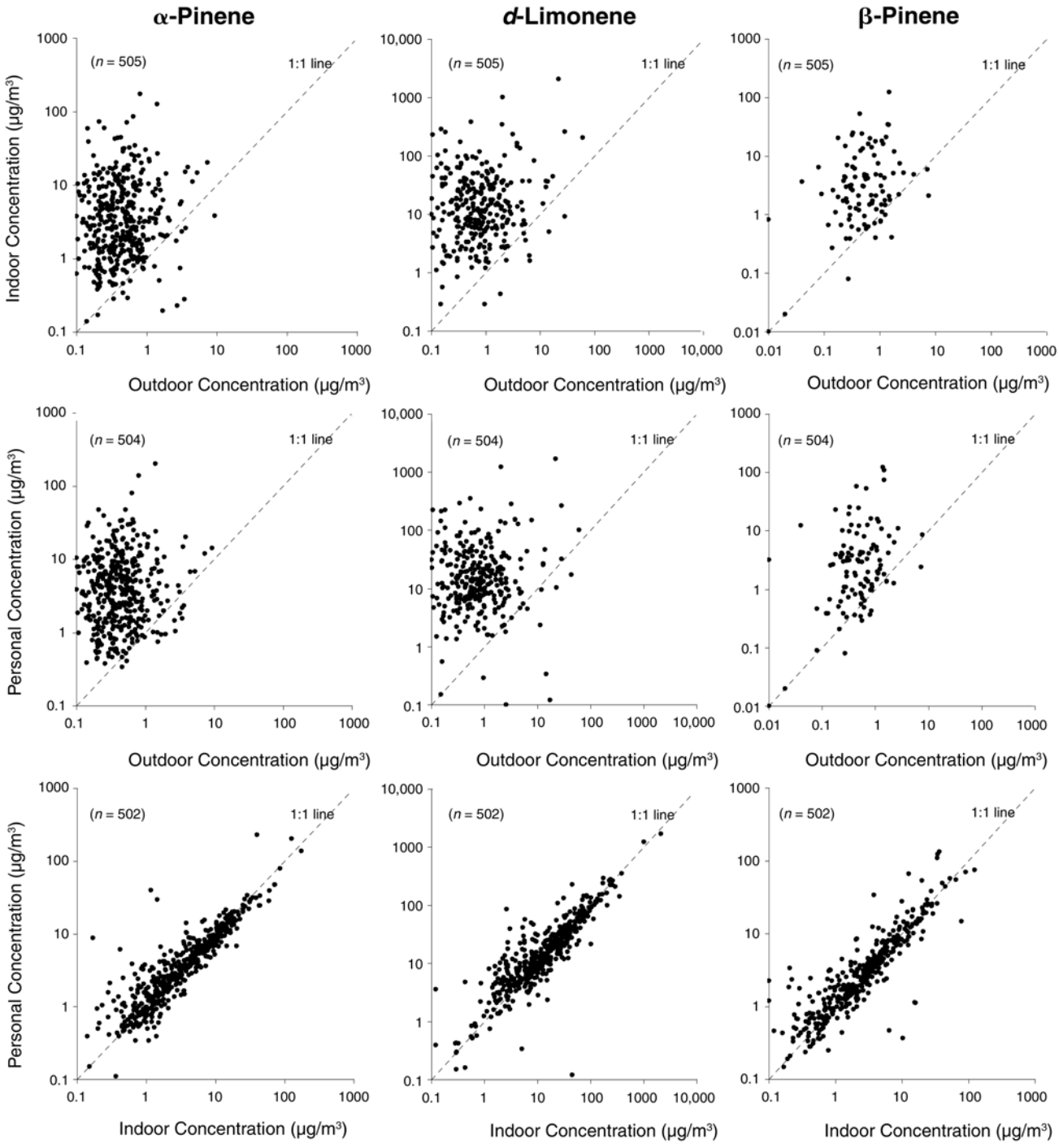

Figure F.1. ***Continued.***

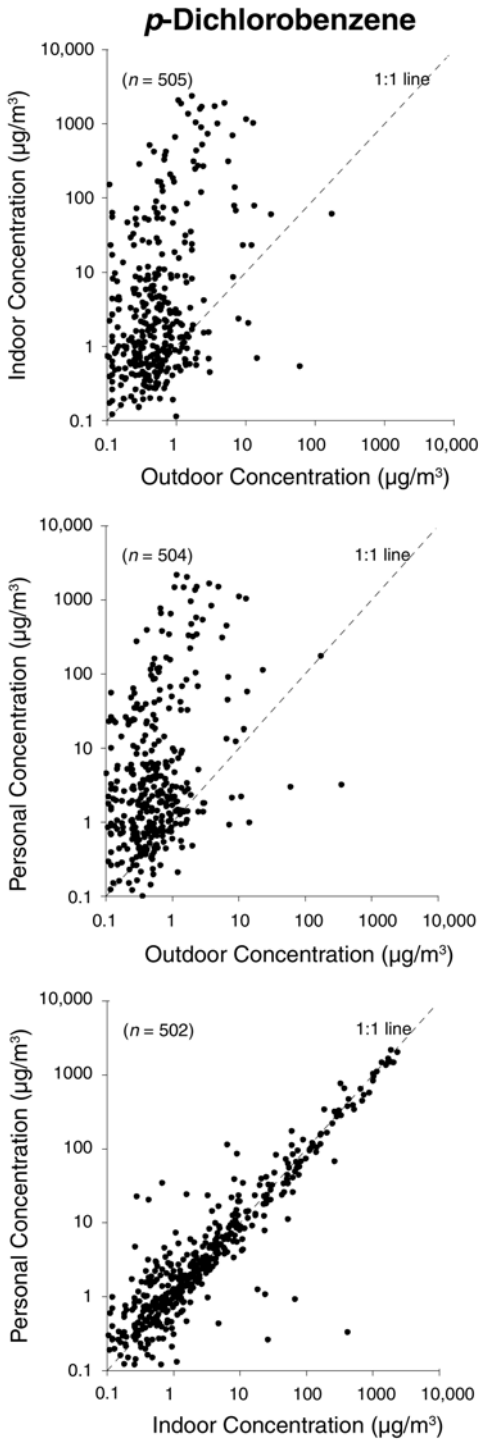

Figure F.1. ***Continued.***

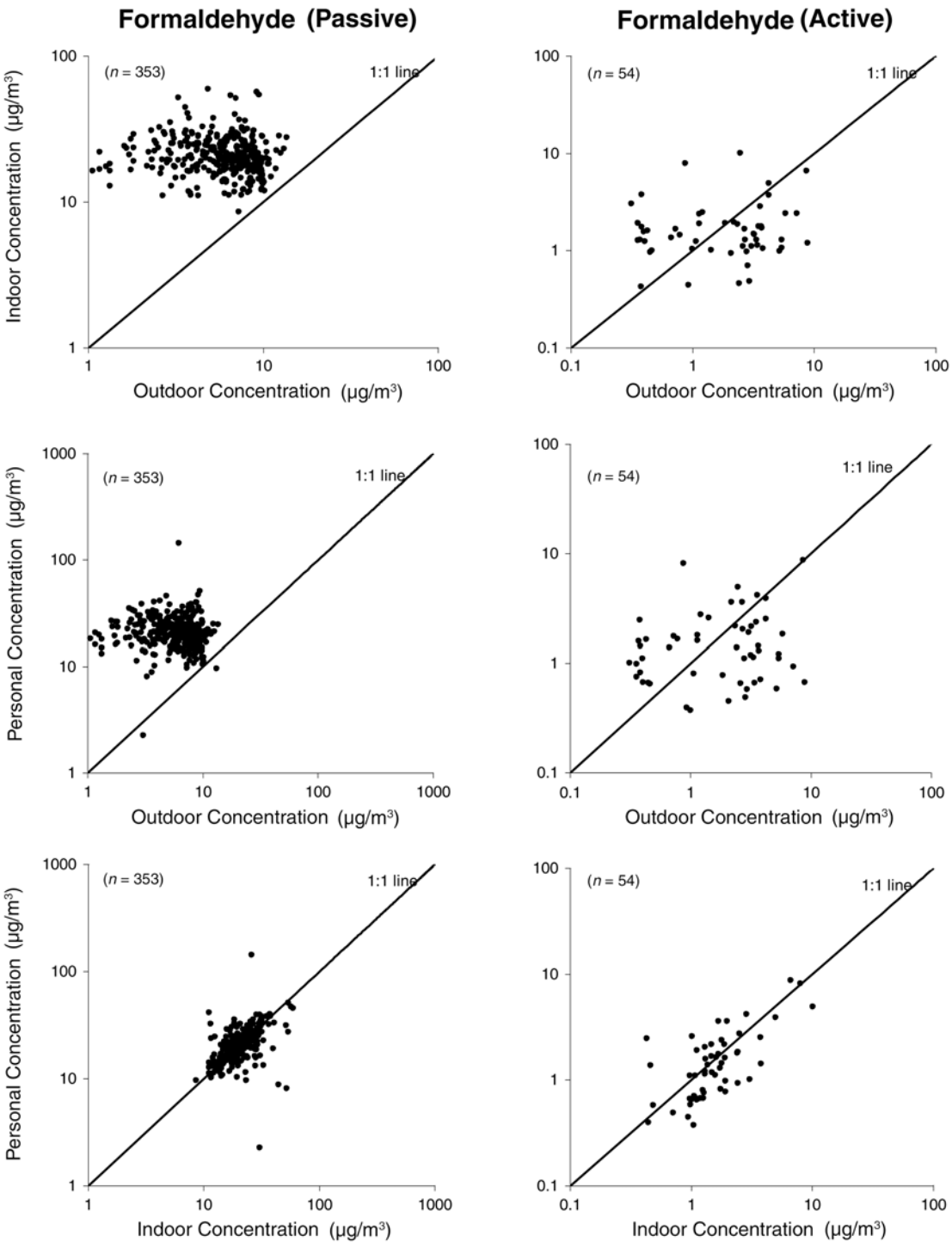

Figure F.2. Concentrations of each of 15 carbonyls obtained using the passive or active method or both and of $PM_{2.5}$ to demonstrate indoor–outdoor, personal–outdoor, and personal–indoor comparisons. Each carbonyl and the measurement method (or $PM_{2.5}$) is identified at the top of the column. All comparisons are shown on logarithmic scales; note that some axis scales may differ within a column or between comparative columns.

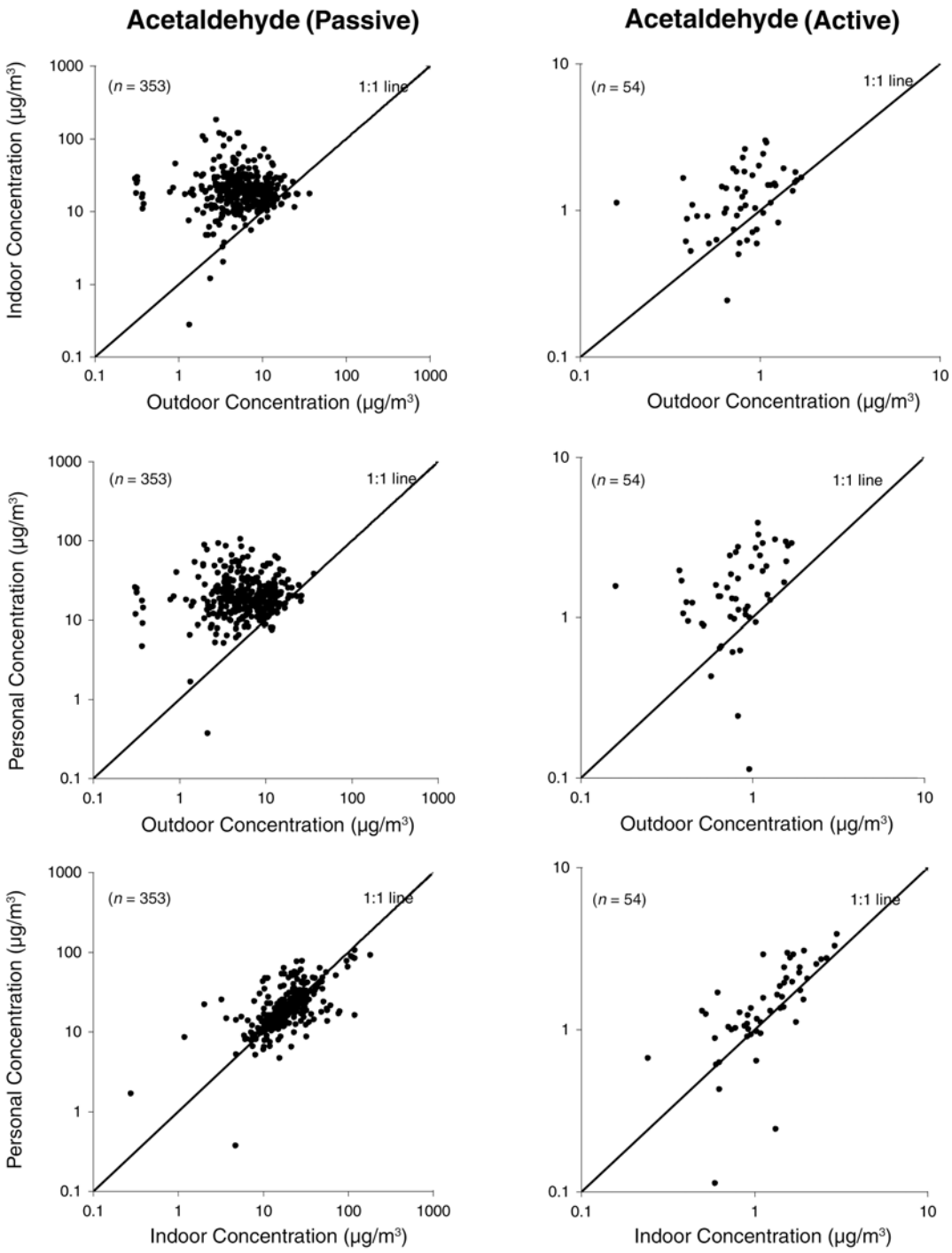

Figure F.2. ***Continued.***

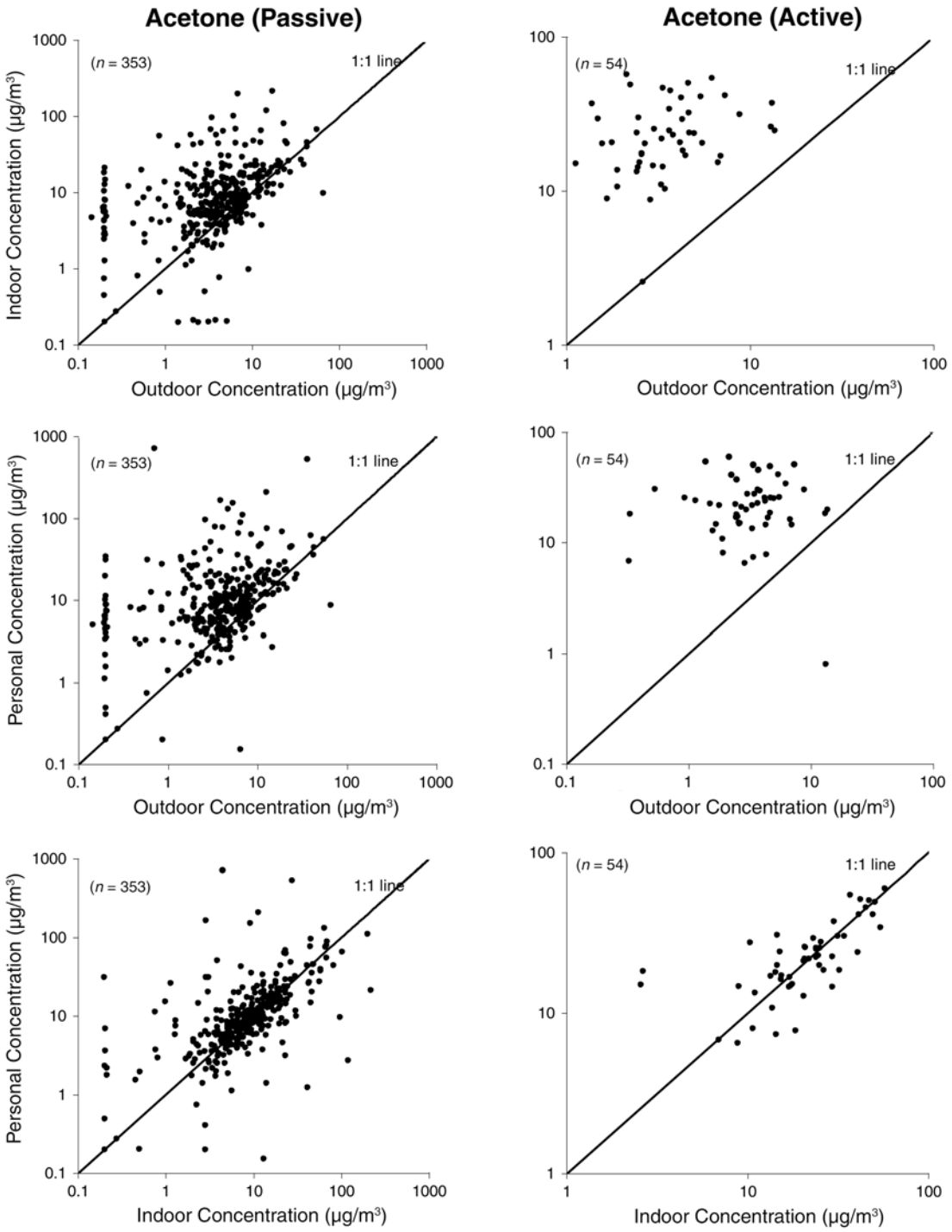

Figure F.2. ***Continued.***

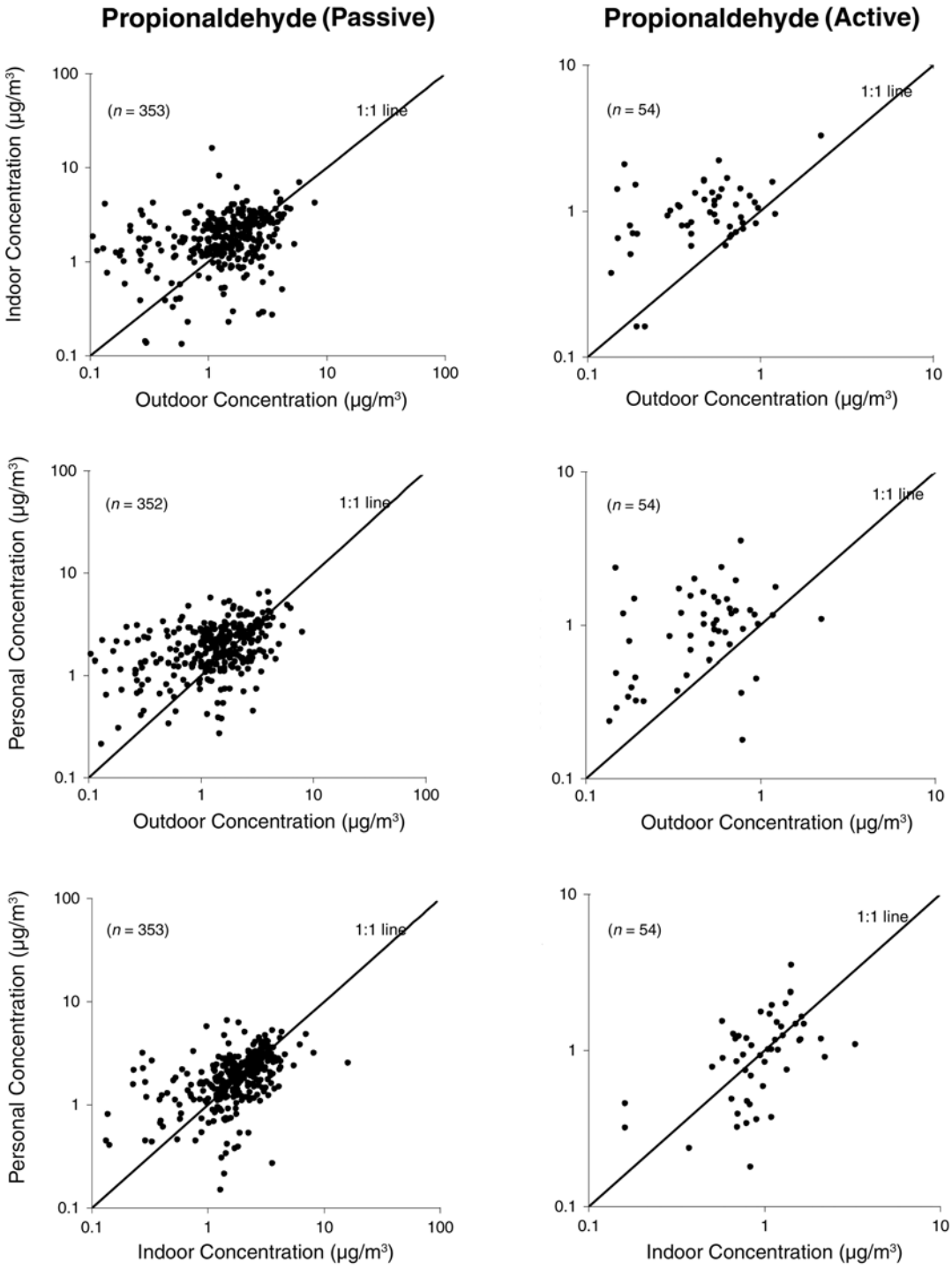

Figure F.2. ***Continued.***

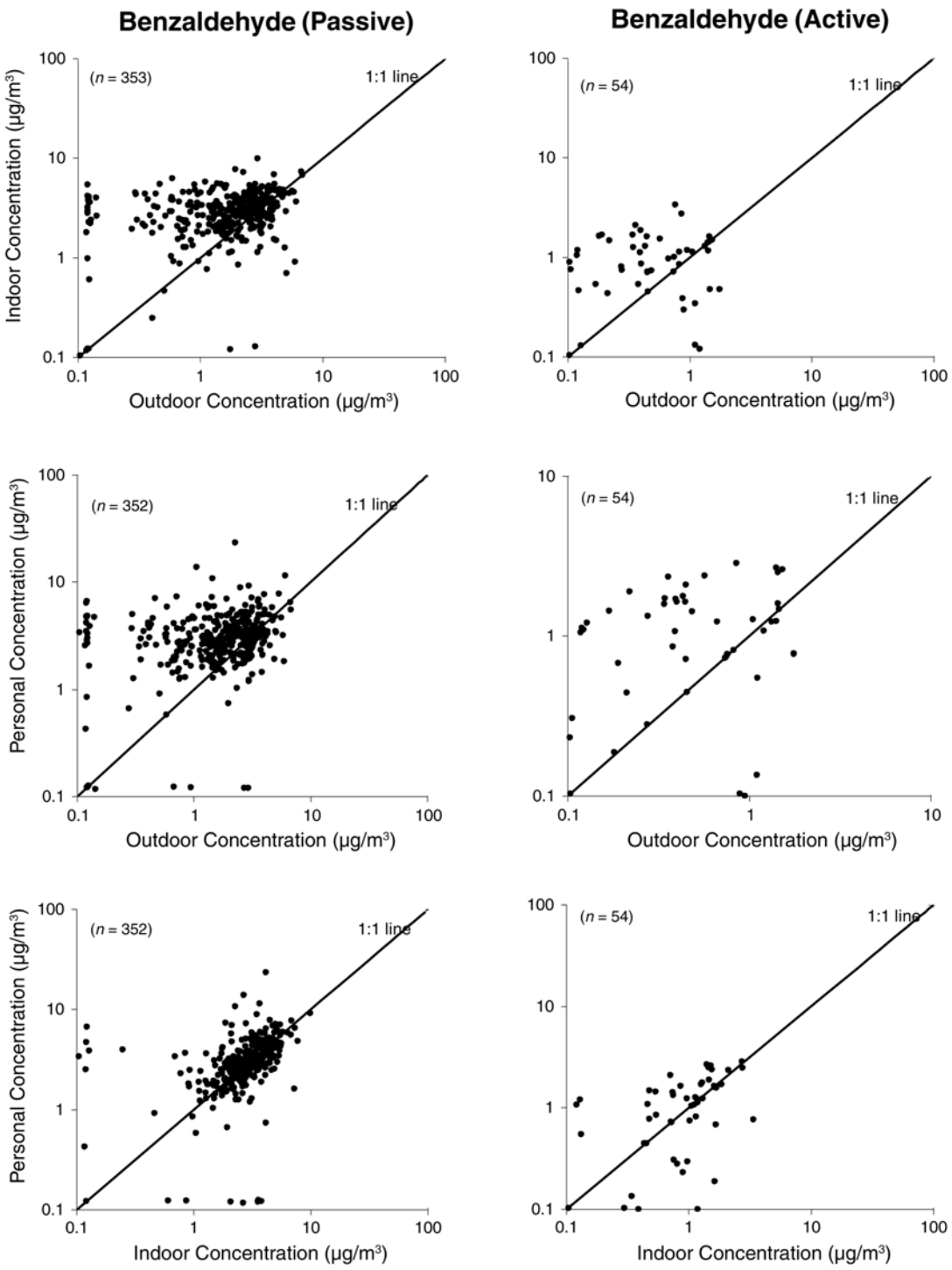

Figure F.2. ***Continued.***

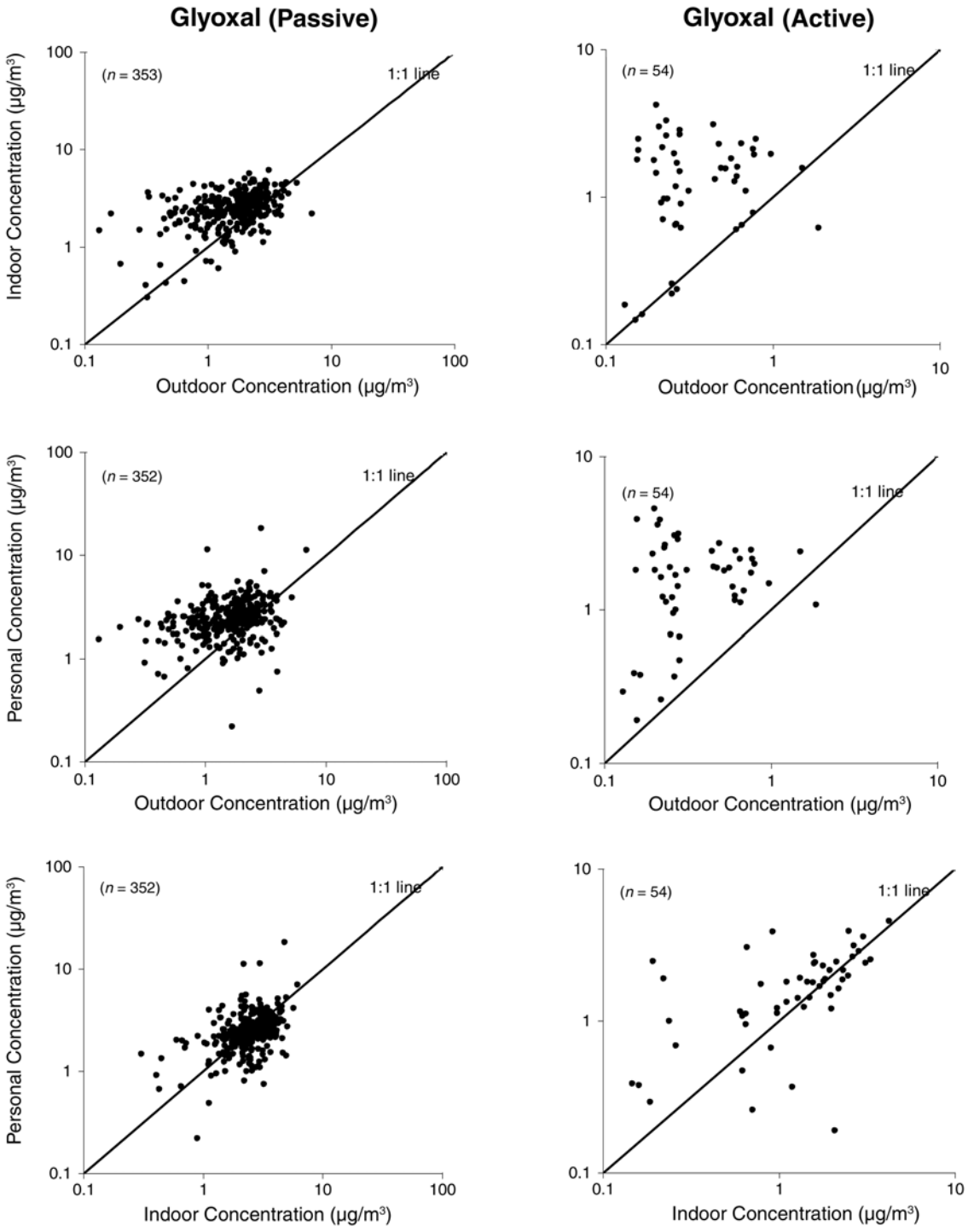

Figure F.2. ***Continued.***

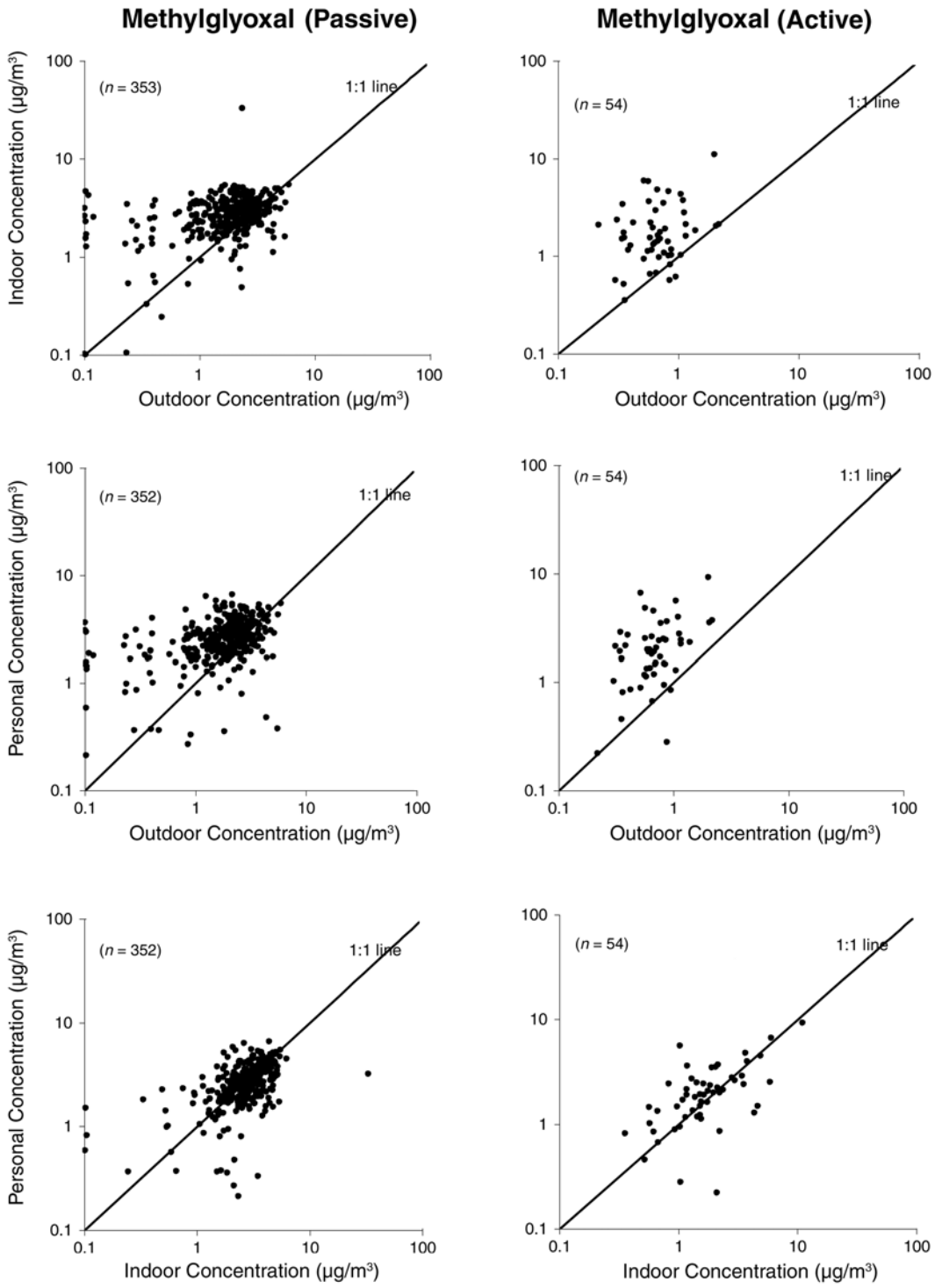

Figure F.2. ***Continued.***

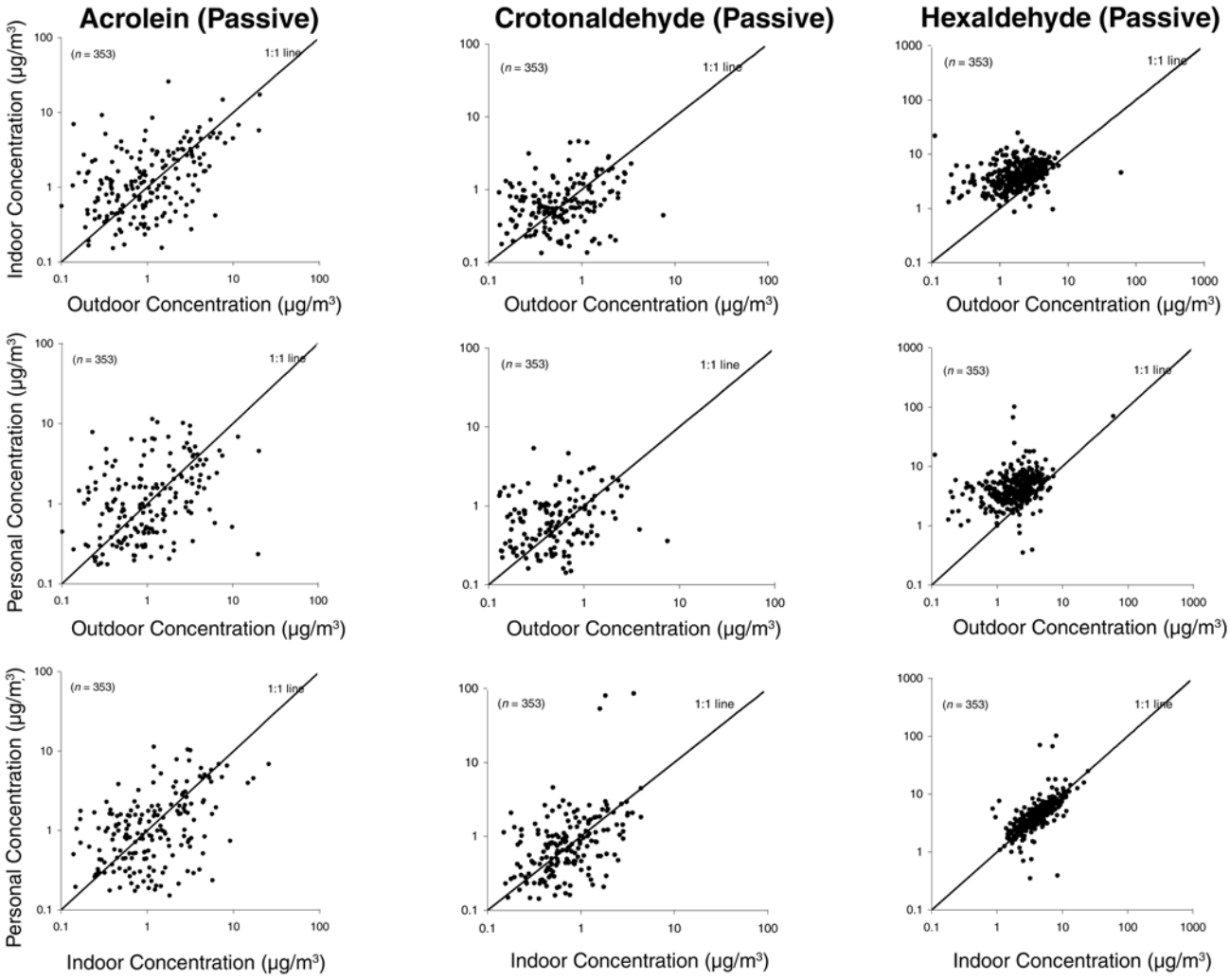

Figure F.2. ***Continued.***

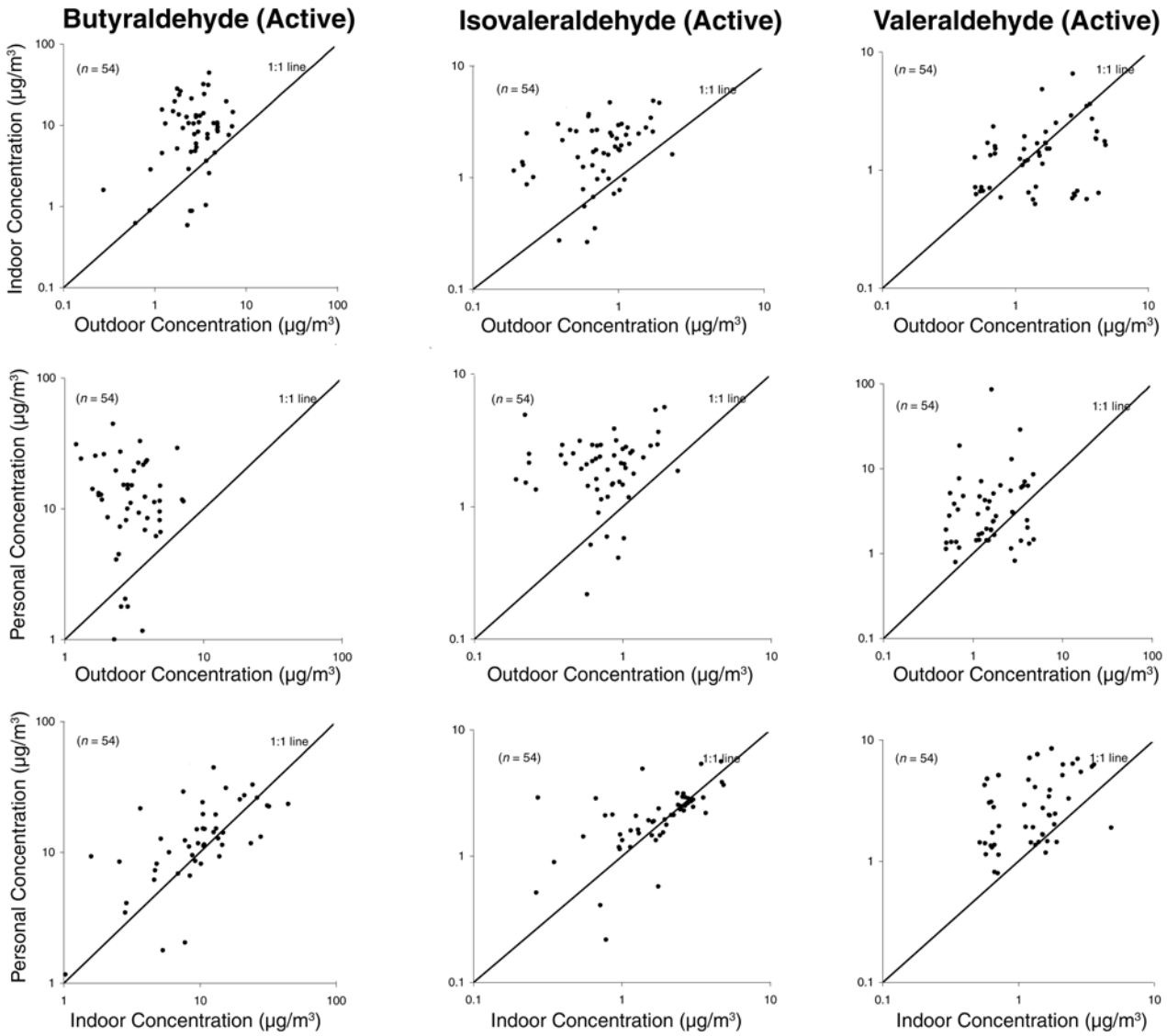

Figure F.2. ***Continued.***

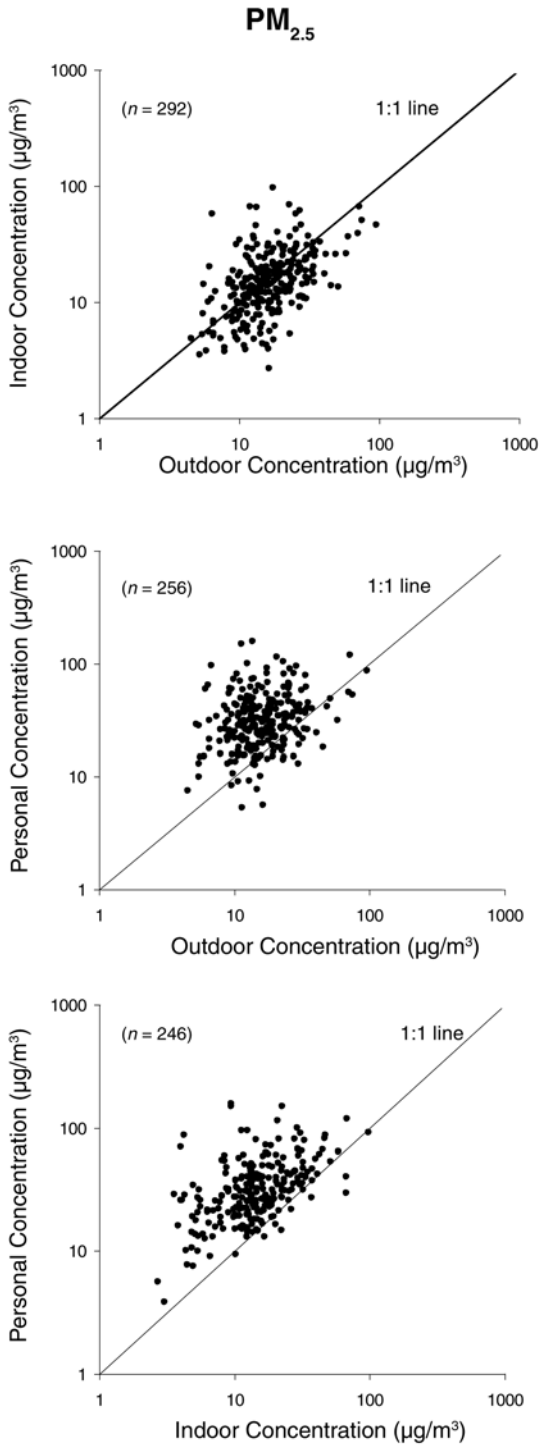

Figure F.2. ***Continued.***

APPENDIX G. HEI and NUATRC Quality Assurance Statement

The RIOPA study was simultaneously performed over a multiyear period in three different geographic areas of the United States. An audit team was selected by the sponsoring agencies (the Health Effects Institute and the Mickey Leland National Urban Air Toxics Research Center) to provide external quality assurance and feedback to both the agencies and the participating investigators. The audit team consisted of two individuals: Kochy Fung, who has an extensive background in methods development and analytic determination of gas-phase species, and Edward Avol, who has many years of experience in environmental health field sampling and in human health research conducted both in communities and the laboratory. On-site audits of the field investigative teams (in Elizabeth NJ, Houston TX, and Los Angeles CA) were performed in each study location by one or both members of the audit team.

Two sets of on-site audits were conducted in the course of study operations. The initial set was performed in 1999, early into actual field operations. A second series of on-site closeout audits were performed in 2003 to verify data sets, track randomly selected subjects through the data collection process, and confirm the status of archival storage for all components of the data set, field logs, and sample measurements.

Database development and management, study sample preparation and laboratory processing, and field operation elements of the study were all carefully evaluated firsthand by the auditors. Study standard operating procedures were reviewed and compared to actual operations. Field investigative teams were accompanied by auditors in each of the three geographic locations during actual study deployments to verify procedural compliance and observe study field operations. Data management activities were also reviewed at each of the three research centers. Editing, acceptance, validation, and data processing activities were recreated using randomly selected values in the data set. Archival storage, preservation, and access to the data set were also investigated.

Audit reports were prepared, submitted in written form to sponsoring agencies and discussed with study investigators immediately after on-site audits to the respective sites.

Ed Avol

Kochy Fung

Quality Assurance Auditors for RIOPA

Date	Study Location
September 20–23, 1999	Los Angeles CA; UCLA field and data site audit
October 26–28, 1999	Elizabeth NJ; Rutgers field and data site audit
October 3–6, 1999	Houston TX; Houston Medical Center field & data site audit
November 9–12, 2003	Elizabeth NJ; Rutgers field and data site audit
November 25, 2003	Los Angeles CA; UCLA field and data site audit
December 14–15, 2003	Houston TX; Houston Medical Center field and data site audit

APPENDIX AVAILABLE ON REQUEST

The following materials may be requested by contacting the Health Effects Institute at Charlestown Navy Yard, 120 Second Avenue, Boston MA 02129-4533, +1-617-886-9330, fax +1-617-886-9335, or email (*pubs@healtheffects.org*). Please give (1) the first author, full title, and number of the Research Report and (2) title of the appendix requested.

APPENDIX H. Baseline, Technician Walk-Through, and Activity Questionnaires

ABOUT THE AUTHORS

Clifford P Weisel, PhD, is a professor of environmental and occupational medicine at Robert Wood Johnson Medical School-University of Medicine and Dentistry of New Jersey and the Deputy Director of the Exposure Measurement and Assessment Division of EOHSI. He is a member of the graduate faculty of the School of Public Health of University of Medicine and Dentistry of New Jersey and the Environmental Sciences Graduate Program of Rutgers, The State University of New Jersey, where he codirects the Joint UMNDJ-RU graduate degree in Exposure Assessment.

Junfeng (Jim) Zhang, PhD, is an associate professor of the environmental and occupation health department at the School of Public Health-University of Medicine and Dentistry of New Jersey and a member of the Exposure Measurement and Assessment Division of EOHSI. He also holds an appointment in the Environmental Sciences Graduate Program of Rutgers, The State University of New Jersey.

Barbara J Turpin, PhD, is an associate professor of environmental sciences at Rutgers, The State University of New Jersey, and is a member of the Exposure Measurement and Assessment Division of EOHSI. She directs the Air Pollution section of the Cooperative Extension of New Jersey Agricultural Station.

Maria T Morandi, PhD, is an assistant professor of public health at the University of Texas in Houston.

Steven Colome, PhD, is principal of Integrated Environmental Science and an adjunct professor of public health at the University of California at Los Angeles.

Thomas H Stock, PhD, is an associate professor of public health at the University of Texas in Houston.

Daila M Spektor, PhD, was an affiliate of Integrated Environmental Science and a senior research scientist at Rand Corporation. Dr Spektor died in 2002.

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ABBREVIATIONS AND OTHER TERMS

ACN	acetonitrile
AER	air exchange rate
ANOVA	analysis of variance
CAT	capillary absorption tube
CI	confidence interval
CV	coefficient of variation
DNPH	2,4-dinitrophenylhydrazine
DNSH	dansylhydrazine
EOHSI	Environmental and Occupational Health Sciences Institute
EXPOLIS	Air Pollution Exposure Distributions of Adult Urban Populations in Europe [study]
EPA	Environmental Protection Agency (US)
GC	gas chromatography
GC–MS	gas chromatography–mass spectrometry
GerESII	German Environmental Survey 1990/1992 [study]
HAPs	hazardous air pollutants
HPLC	high-pressure liquid chromatograph
I/O	indoor-to-outdoor concentration ratio
LC	liquid chromatography
LOD	limit of detection
MDL	method detection limit
MS	mass spectrometry
MTBE	methyl *tert*-butyl ether
NHEXAS	National Human Exposure Assessment Survey
OVM	organic vapor monitor
PAH	polycyclic aromatic hydrocarbon
PAKS	passive aldehydes and ketones sampler
PEM	personal environmental monitor
PM	particulate matter
$PM_{2.5}$	particulate matter with a mass median aerodynamic diameter $\leq$ 2.5 µm
PMCH	perfluorinated methylcyclohexane
PTEAM	particle total exposure assessment methodology [study]
r^2	coefficient of determination
RCS	random component superposition [model]
RIOPA	relationships of indoor, outdoor, and personal air [study]
TEACH	Toxic Exposure Assessment: A Columbia/Harvard study
TEAM	total exposure assessment methodology [study]
VOC	volatile organic compound

COMMENTARY
Special Review Panel

INTRODUCTION

Urban populations are exposed to a complex mixture of possibly hazardous air pollutants generated and emitted by a variety of outdoor and indoor sources or formed as the result of chemical transformations in the atmosphere. These pollutants occur naturally or may be the result of human activities; they may be present in the form of gases, liquid droplets, or solid particles. Outdoor sources can be large or small, stationary or mobile, and can affect both localized and widespread areas. Indoor sources include building materials, carpets, household chemicals, and heating, cooking, and cleaning activities.

The US Environmental Protection Agency (EPA*) defines an air toxic as any substance in the air that is known or suspected to cause harm to humans or the environment. The 1990 Clean Air Act Amendments list 188 air toxics as hazardous air pollutants (HAPs) that the EPA is required to evaluate and, if appropriate, control. These include a large number of volatile organic compounds (VOCs), including carbonyls (aldehydes and ketones), and components often associated with particulate matter (PM).

Epidemiologic and animal studies have shown associations between exposure to many of these air toxics and a variety of adverse health effects (reviewed in Leikauf 1992; EPA 1993; Heseltine et al 1993; Snyder 2000; Delfino 2003). Data collected in several outdoor and indoor locations across the United States have shown that most of the VOCs are present in both environments (Shah and Singh 1988). Studies of targeted populations determined that indoor sources of air toxics contribute to personal exposures and in most cases outweigh the impact of emissions from outdoor sources (Wallace et al 1987).

The US EPA also regulates the maximum ambient concentrations of PM, which in urban air includes particles of varying sizes and composition. The main components of PM are elemental and organic carbon, inorganic ions (ammonium sulfate and ammonium nitrate), and metallic ashes. Many epidemiologic studies have shown an association between exposure to PM and increased mortality and morbidity (EPA 2004). However, lack of information on several important aspects of exposure to particles (including the characteristics that may be associated with the effects observed) complicates interpreting this research, assessing human risk, and designing control strategies.

Obtaining a clear understanding of personal exposure in relation to ambient pollutant concentrations and how different sources contribute to individual exposure has been considered an important first step in assessing the possible public health risks posed by air toxics and PM in the urban environment.

The intent of the Relationships of Indoor, Outdoor, and Personal Air (RIOPA) study was to define the relation between indoor, outdoor, and personal exposure concentrations of a large number of air toxics, particulate matter 2.5 μm or smaller in aerodynamic diameter ($PM_{2.5}$), and components of $PM_{2.5}$ in well-defined populations selected according to the distance of their residences from point (large stationary sources), area, and mobile sources.

The Preface to this Research Report describes the application and selection processes through which the three components of the RIOPA study were funded. Due to the large set of data and analyses, the Investigators' Final Report was divided into *Part I: Collection Methods and Descriptive Analyses* (for VOCs, carbonyls, $PM_{2.5}$ concentrations; this volume and the subject of this Commentary) and *Part II: Analyses of Concentrations of Particulate Matter Species* (the compositional analyses of $PM_{2.5}$; in press).†

The Commentary is intended to aid the sponsors of NUATRC and HEI and to inform the public by highlighting the strengths of the study, pointing out alternative interpretations, and placing the research into scientific and regulatory perspective.

* A list of abbreviations and other terms appears at the end of the Investigators' Report.

This document has not been reviewed by public or private party institutions, including those that support HEI and NUATRC; therefore, it may not reflect the views of these parties, and no endorsements by them should be inferred.

† The RIOPA study resulted from three applications: "Relationship Among Indoor, Outdoor, and Personal Exposures to Air (RIOPA Study)" to NUATRC with Dr Clifford Weisel as principal investigator; "Personal and Microenvironmental Measurements of Human Exposures to Multiple Aldehydes in Three Distinct Urban Areas" to HEI with Dr Junfeng (Jim) Zhang as the principal investigator; and "Contributions of Outdoor PM Sources to Indoor Concentrations and Personal Exposures: A Three-City Study" to HEI with Dr Barbara Turpin as the principal investigator. Dr Weisel's portion of the study began in December 1997, Dr Zhang's portion in June 1998, and Dr Turpin's portion in October 1998. Total NUATRC expenditures for the Weisel portion were $1,512,327. Total HEI expenditures for the Zhang and Turpin portions were $1,961,153. An integrated draft Investigators' Report was received for review by the HEI and NUATRC Special Review Panel in March 2002. A revised report, received in January 2004, was accepted for publication in May 2004. (See also *Part II. Analyses of Concentrations of Particulate Matter Species* for a complete presentation of Dr Turpin's analyses of PM.) During the review process, the Special Review Panel and the investigators had the opportunity to exchange comments and to clarify issues in both the Investigators' Report and in the Special Review Panel's Commentary.

REGULATORY BACKGROUND

The Clean Air Act of 1970 and subsequent amendments provide a framework for regulating the concentrations of certain pollutants in source emissions and in ambient air. Many regulatory actions pertain to the two groups of pollutants examined in this study, air toxics and PM.

AIR TOXICS

The Clean Air Act Amendments of 1990 require the EPA to promulgate standards based on maximum achievable reductions and generally achievable control technology (Section 112 [d]) for all large or major stationary (point) sources and some small area sources of the 188 HAPs (US Congress 1990). In addition, the EPA is to promulgate standards to address the risks remaining after technology-based standards are imposed. This phase of the regulatory response is being implemented through programs such as the Integrated Urban Air Toxics Strategy and the Residual Risk Program (Section 112 [f]; EPA 1999).

The Integrated Urban Air Toxics Strategy identifies 33 HAPs plus diesel exhaust PM as high-priority urban air toxics because of their emission levels (regardless of sources) or because they pose the greatest estimated threat to public health (EPA 1999). The EPA strategy is to (1) ensure 75% reduction in cancer incidence from exposure to pollutants from stationary sources and a substantial reduction in the risk of noncancer health effects; (2) focus on sensitive populations or on geographic areas with heavy concentrations of emissions (hot spots); and (3) ensure that area sources that account for 90% of the total emissions of urban HAPs are subject to maximum and generally achievable control technology standards.

Section 112 of the Clean Air Act Amendments of 1990 also required the EPA to regulate, or consider regulating, air toxics from motor vehicles by setting standards for fuels or vehicles or both. The EPA has thus developed a list of 21 air toxics that originate, at least in part, from mobile sources; inclusion in the list takes into account motor vehicle emissions data and health assessment information (EPA 2001). Not all of the 21 mobile-source air toxics are included in the list of 33 high-priority urban air toxics (EPA 1999); however, with the exception of diesel exhaust PM, they are all in the list of 188 HAPs originally established by the Amendments of 1990. Because of expected reductions in air toxics as a result of existing and future regulatory programs for fuels and emissions, the EPA did not specify standards to control mobile-source emissions as part of the regulations imposed in 2001 (EPA 2001); but the Agency is currently reviewing the information on air toxics and is expected to propose a new rule in 2006.

The California Air Resources Board also has developed a list of toxic air contaminants to consider for possible control measures (California Air Resources Board 1999). Currently, 113 compounds (including diesel exhaust PM) are on this list.

In accordance with the EPA rules, state and regional agencies undertake monitoring programs to measure ambient concentrations of a variety of air toxics in communities within their jurisdictions. In some regions, private monitoring networks have also been established. In addition, the EPA maintains a National Emissions Inventory (*www.epa.gov/ttn/chief/net/*) with input from state and local agencies and industries. The database includes, by source, estimates of annual emissions of HAPs and criteria pollutants (those for which the EPA has set health-based standards) in each area of the country. Data from the Inventory are used for modeling air dispersion, developing regional strategies, setting regulations, assessing air toxics risks, and tracking trends in emissions over time.

Among the species measured in the RIOPA study are VOCs included in the list of the EPA's 33 urban air toxics (benzene, carbon tetrachloride, chloroform, methylene chloride, tetrachloroethylene, and trichloroethylene); VOCs primarily from mobile sources (benzene, ethyl benzene, methyl *tert*-butyl ether [MTBE], styrene, toluene, *o*-xylene, and *m*- & *p*-xylenes); and other VOCs that originate primarily from indoor sources (*d*-limonene, α-pinene, and β-pinene). Some of the carbonyls measured are included in both of the EPA's lists of urban and mobile-source air toxics, and also originate from indoor sources (acetaldehyde and formaldehyde); some are not listed as HAPs and are primarily from mobile sources (butyraldehyde, crotonaldehyde, hexaldehyde, isovaleraldehyde, propionaldehyde, and valeraldehyde) or are formed from photochemical reactions with hydrocarbons (glyoxal and methylglyoxal). Commentary Table 1 lists the compounds measured and which are classified as urban air toxics or mobile-source air toxics by the EPA or as toxic air contaminants by the California Air Resources Board. The sidebar describes the major sources of the species measured in the RIOPA study.

PM

PM is generated by combustion, mechanical processes, and many other methods. Based on aerodynamic diameter in ambient air, particulate volume distribution is conventionally classified in three size modes: coarse particles (> 1 μm), fine particles (0.1 to 1 μm), and ultrafine particles (< 0.1 μm) (EPA 2004). Because of concerns about health effects of exposure to particles, the EPA regulates

Commentary Table 1. Compounds Measured in RIOPA and Their Designations As Air Toxics by the EPA and the California Air Resources Board

Compounds	EPA Urban Air Toxics[a]	EPA Mobile-Source Air Toxics[b]	CARB Toxic Air Contaminants[c]
VOCs			
1,3-Butadiene[d]	√	√	√
Methylene chloride	√	—	√
MTBE	—	√	√
Chloroprene[d]	—	—	—
Chloroform	√	—	√
Carbon tetrachloride	√	—	√
Benzene	√	√	√
Trichloroethylene	√	—	√
Toluene	—	√	√
Tetrachloroethylene	√	—	—
Ethyl benzene	—	√	√
m- & p-Xylenes	—	√	√
o-Xylene	—	√	√
Styrene	—	√	√
α-Pinene	—	—	—
β-Pinene	—	—	—
d-Limonene	—	—	—
p-Dichlorbenzene	—	—	√
Carbonyls (passive method only)			
Formaldehyde	√	√	√
Acetaldehyde	√	√	√
Acetone	—	—	—
Acrolein	√	√	√
Propionaldehyde	—	—	—
Crotonaldehyde	—	—	—
Benzaldehyde	—	—	—
Hexaldehyde	—	—	—
Glyoxal	—	—	—
Methylglyoxal	—	—	—

[a] EPA 1999.

[b] EPA 2001a.

[c] California Air Resources Board 1999.

[d] Data not reported in the Investigators' Report due to low recovery.

ambient mass concentrations of fine PM through the National Ambient Air Quality Standards for $PM_{2.5}$ (EPA 1997). The current standards are being reviewed.

The EPA also regulates PM emitted directly from mobile and stationary sources. Like ambient standards, emission standards are based on the mass (weight) of particles and do not take into account particle composition, which depends on the sources and compounds present during the generation process. Information on the composition of PM to which people are exposed is needed to help determine whether certain components are more strongly associated with health outcomes than others.

SCIENTIFIC BACKGROUND

Estimates of human exposure to air contaminants have generally relied on measurements at fixed sampling locations, but the relation of such measurements to personal exposure is uncertain. Assessments of personal exposure could be improved by considering, for example, the characteristics of specific indoor and outdoor environments, the time and activity patterns of individuals (Wallace et al 1989), and the impact of indoor sources of air pollution (Samet et al 1987; Stolwijk 1990; Weschler et al 1990; Wallace 1991). Most urban residents spend more than 90% of their time indoors, and indoor environments have contaminant concentrations different from, and often higher than, the immediate outdoor environment due to indoor sources (Spengler and Sexton 1983).

The concept of the microenvironment was developed to allow better estimates of an individual's daily pollutant exposure. Total personal exposure on a given day is the sum of exposures received in each of the microenvironments the person has passed through in that day multiplied by the time spent in each environment. By placing monitors in microenvironments visited by individuals, both indoors and outdoors, personal exposures to a pollutant could be estimated more accurately. This approach was used in the Harvard Six Cities Study (Ferris et al 1979), a comprehensive longitudinal study of the respiratory health effects of air pollutants generated by burning fossil fuels.

Improvements in technology, though, have further improved assessments by allowing study subjects to wear or carry small collection devices that measure some toxic air pollutants as concentrations in the individual's breathing zone (referred to as personal air). Such personal monitoring samplers were used by investigators in the Total Exposure Assessment Methodology (TEAM) studies that the EPA conducted throughout the United States from 1980 through 1988 (Wallace et al 1985, 1987, 1991; Wallace 1993; Ozkaynak et al 1996; Rappaport and Kupper 2004). The TEAM studies investigated exposures of individuals to toxic chemicals through air, water, and food pathways. Data on personal exposures to VOCs were collected in a group of more than 1000 persons in ten US cities recruited using a probability-based sampling framework (in which the participants proportionally represent the greater population).

SOURCES OF VOCs, ALDEHYDES, AND PM

The species measured in the RIOPA study are grouped into categories based on their chemical structures. Many have common sources, such as cigarette smoke. Most species are generated both indoors and outdoors, but some only outdoors (MTBE) and some only indoors (the terpenes).

This brief description of sources of was compiled from

1. the Agency for Toxic Substances and Disease Registry (*www.atsdr.cdc.gov/toxpro2.html*);
2. the EPA National Air Toxics Assessment (*www.epa.gov/ttn/atw/nata/pollinf2.html*);
3. the Hazardous Substances Data Bank (*http://toxnet.nlm.nih.gov/cgi-bin/sis/htmlgen?HSDB*); and
4. the California Office of Environmental and Health Hazard Assessment (*www.oehha.org/air/toxic_contaminants/tactable.html*).

Benzene, Toluene, Ethyl Benzene, and the Xylenes

These compounds are constituents of petroleum products, particularly gasoline, jet fuel, and kerosene. As pollutants, they result from combustion and are emitted from vehicle tailpipes, oil refineries, and hazardous waste sites; they also evaporate from vehicles and from solvents in paints and coatings.

MTBE

MTBE is added to gasoline to increase octane levels and reduce emissions of carbon monoxide. Its addition also reduces emissions of benzene but increases emissions of formaldehyde and acetaldehyde. Exposures to MTBE result from both tailpipe and evaporative emissions (including vapors during refueling). The Clean Act Amendments of 1990 mandated that MTBE be used in geographic areas with high carbon monoxide concentrations (which started in 1992) and in reformulated gasoline in areas with high ozone concentrations (which started in 1995). The three cities in the RIOPA study were located in states where gasoline with MTBE is used (Health Effects Institute 1996).

Chlorinated Compounds

Carbon tetrachloride, used primarily as a solvent, refrigerant, and propellant, is released by industries and oil refineries. Because of it's harmful effects, it has been banned from consumer products and is only used in industrial processes.

Tetrachloroethylene (perchloroethylene) is released by waste incinerators and by industries in which it is used as a general solvent. It is widely used in dry cleaning operations and can be released from dry-cleaned clothing inside homes.

Trichloroethylene is used mainly as a solvent to remove grease from metal parts, but it is also an ingredient in adhesives and paint removers.

Chloroform in the air is generally a by-product of chlorine use. Pulp and paper mills where chlorine is used as a bleach and water treatment plants where chlorine is used as a disinfectant are the primary sources of chloroform; it is also emitted from chemical industries that use chlorine to make other chemicals. Chlorine is present in household bleach products and as a solvent in a variety of household products; it can also derive from chlorinated water.

1,3-Butadiene, Styrene, and Chloroprene

1,3-Butadiene is derived from the incomplete combustion of petroleum-derived fuels. Mobile sources are responsible for most emissions; area and point sources include petroleum refineries, residences, and industrial operations. Vehicles with malfunctioning catalysts emit much higher concentrations than vehicles with functioning catalysts. Other butadiene sources are manufacturers of tires, other rubber products, and latex paint.

Styrene is released from mobile sources and by facilities that manufacture tires and other rubber products, plastics, and resins.

Chloroprene is released from facilities that use it to produce rubber and rubber goods, sealants, and adhesives.

Terpenes and *p*-Dichlorobenzene

Terpenes are a broad category of organic solvents obtained from naturally occurring oils. α-*Pinene*, β-*pinene*, and d-*limonene* are obtained from pine and citrus oils. Pinene is the major oil constituent of turpentine. These chemicals have very strong odors and are used in household products as cleaners and deodorizers.

p-*Dichlorobenzene* is used primarily to make mothballs and solid ambient deodorizers, from which it is released as a vapor by sublimation at indoor temperatures.

Carbonyls

Aldehydes Several aldehydes, most importantly *formaldehyde, acetaldehyde, acrolein, crotonaldehyde*, and *propionaldehyde*, are present in emissions from motor vehicles, power plants using fossil fuels, incinerators, wood combustion, cigarettes, and cooking. They are also naturally formed in ambient air through photochemical oxidation of hydrocarbons in the presence of hydroxyl radicals and ozone (Grosjean et al 1983; Atkinson 1990). Photochemical production of aldehydes is more predominant than direct emissions, particularly during the daytime hours and on warm sunny days (Harley and Cass 1994).

In urban areas *glyoxal* and *methylglyoxal* are primarily derived from the photochemical reactions of toluene and other aromatic compounds (Dumdei and O'Brien 1984). *Formaldehyde* is released from building materials, carpets, ordinary paper products, and indoor combustion sources. *Acetaldehyde* and *isovaleraldehyde* are used in producing perfumes.

Ketones (Acetone) Acetone occurs naturally in plants, trees, volcanic gases, forest fires, and as a product of the breakdown of body fat. It is used to make plastics, fibers, drugs, and chemicals such as sealants, and to dissolve other substances. It is present in vehicle exhaust, tobacco smoke, and in the air around landfill sites. Industrial processes contribute more acetone to the environment than do natural processes.

PM

PM is the product of the incomplete combustion of fossil fuels. It is also produced as a result of vehicular brake wear, soil erosion, and sea spray. PM can be generated by outdoor sources and indoor activities (such as cooking, vacuuming, or burning incense and candles). Based on aerodynamic diameter in urban air, PM volume distribution is conventionally classified in three size modes: coarse particles (> 1 μm), fine particles (0.1 to 1 μm), and ultrafine particles (< 0.1 μm). Maximum outdoor ambient concentrations of fine PM are regulated by the EPA through the National Ambient Air Quality Standards for $PM_{2.5}$ (EPA 1997).

Data on personal exposures to PM_{10} were collected from about 180 persons in one city (Riverside CA). The TEAM studies showed that, for the most prevalent VOCs and for PM, personal exposure concentrations were consistently higher than either indoor or outdoor concentrations; indoor concentrations dominated personal exposures, which implies that indoor sources (such as consumer products) and personal activities contribute substantially to total exposures (Wallace et al 1991).

When NUATRC released the RFA that resulted in funding the RIOPA study, the EPA was conducting the National Human Exposure Assessment Survey (NHEXAS) to provide scientists and regulators with information about personal exposures through inhalation, ingestion, and dermal routes. That study involved a probability-based sample of 500 individuals in six Midwestern states; the investigators measured personal exposures to a suite of chemicals including VOCs, particles, metals, and pesticides; they acquired data from direct exposure measurements and from biomarkers that could indicate the internal dose that may result from those exposures (Clayton et al 1999; Pellizzari et al 2001b).

In summary, at the time the RIOPA study was funded (see the Preface to this Research Report), few studies had acquired concomitant measurements of various pollutants outdoors, indoors, and in personal exposures for several cities, or had provided information about how outdoor sources contribute to indoor air pollutant concentrations.

TECHNICAL EVALUATION

STUDY DESIGN AND OBJECTIVES

The RIOPA study measured personal exposures and outdoor and indoor air concentrations of $PM_{2.5}$ and selected VOCs and carbonyls for adults and children. Sampling was conducted during two 48-hour sampling periods in different seasons between the summer of 1999 and the spring of 2001. The study was designed to address the hypothesis that outdoor sources contribute a significant proportion of the pollutant concentrations in the indoor and personal air for residents who live near those sources. The study included approximately 100 homes and 100 adult residents of those homes in each of three urban centers with different weather conditions and air pollution source profiles: Los Angeles CA, dominated by mobile sources; Houston TX, dominated by large industrial stationary and area sources (with a portion contributed from mobile sources); and Elizabeth NJ, with a mixture of mobile, point, and area sources. Samples of VOCs, carbonyls, and $PM_{2.5}$ were collected inside and outside the homes and in subjects' personal air (breathing zone). The subjects carried personal samplers during their daily activities. In a subset of homes, the personal exposures of one or more children were monitored; in-vehicle exposures to carbonyls were also measured for some residents.

The specific aims of the portion of the RIOPA study reported here were to:

1. Compare indoor, outdoor, and personal air concentrations (and in-vehicle concentrations for carbonyl compounds) of the pollutants measured.
2. Examine the effects of season, home type, and other variables on measured concentrations.
3. Quantify (1) the contribution of outdoor sources to indoor concentrations and (2) the indoor source strength of the measured pollutants using measured air exchange rates (AERs).

The investigators also planned to estimate the fractional concentration that outdoor sources contribute to indoor concentrations as a function of distance from those sources and to collect a data set that could be used to address important questions about exposure assessment in the future. The analysis of the effect of distance from sources was not conducted as part of this study due to time and budgetary constraints. However, this evaluation is currently under way and the results of the initial analyses are being published elsewhere.

METHODS

Subject Recruitment and Home Survey

The investigators targeted geographic areas of each city according to their distance from sources; they used a variety of methods to recruit the subjects from these areas, such as direct mail, contacts through religious or community leaders, and word-of-mouth. Potential subjects who met predetermined selection criteria were invited to participate in the study and asked to fill out questionnaires to provide information on personal characteristics, their homes, and nearby pollutant sources. Informed consent was obtained from each of the adult participants and from a parent or guardian for each child.

Exposure Monitoring

The indoor sampler was clipped to a rack placed in the main living area inside the home and the outdoor sampler in a secure location outside the home. Participants wore personal samplers on their clothing or in an attached sampler

holder; they filled out an activity questionnaire during the 48-hour monitoring period. Field blanks, collected using capped samplers stored in the indoor sampling rack, were transported and analyzed with the field samples. The samplers used are listed in Commentary Table 2 and briefly described below.

For VOCs, the investigators used the passive organic vapor monitor (OVM 3500) that contains a single pad impregnated with activated carbon. Collected VOCs were extracted from the badges and analyzed by a gas chromatograph and mass selective detector system. The measured values were corrected by subtracting the field blank values. An interlaboratory study was conducted to ascertain the comparability of analyses of VOCs conducted by the Environmental and Occupational Health Sciences Institute (EOHSI) and the University of Texas.

For carbonyls, the investigators used two sampling methods. In the first year of the study, they used the conventional active method consisting of a cartridge impregnated with the carbonyl-reactive compound 2,4-dinitrophenylhydrazine (DNPH) connected to a pump. They subsequently switched to a new passive sampler developed and validated by Dr Zhang as part of the RIOPA study: the passive aldehydes and ketones sampler (PAKS), which uses a cartridge coated with a fluorogenic reagent, dansylhydrazine (DNSH). Regardless of the sample collection method, separation of the species in the extract was done by high performance liquid chromatography. The use of fluorescence detection for the DNSH-carbonyl derivatives was expected to provide better sensitivity and selectivity than the ultraviolet detection used for the DNPH derivatives. In addition, several carbonyls are more stable on the DNSH substrate than on the DNPH substrate. All measurements were corrected for field blank values and recovery rate (as shown in Table 8 of the Investigators' Report and described later in this Commentary). Both the Report and Commentary describe and discuss primarily the results obtained with the passive DNSH sampler because it performed better for some carbonyls and was used most.

Commentary Table 2. Samplers Used in the RIOPA Study

Sampling Period[a]	VOCs	Carbonyls	$PM_{2.5}$
Outdoor	Passive OVM badge	Passive Active	Harvard impactor
Indoor	Passive OVM badge	Passive Active	Harvard impactor
Personal	Passive OVM badge	Passive Active	PEM
In vehicles	Not measured	Active	Not measured

[a] All samplers operated for 48 hours except for those in vehicles; those operated for up to 8 hours.

For $PM_{2.5}$, indoor and outdoor samples were collected on Teflon filters mounted in a Harvard impactor. Personal $PM_{2.5}$ samples were collected on smaller Teflon filters mounted in the personal environmental monitor (PEM). Both samplers separate $PM_{2.5}$ by means of an impactor inlet. All filters were weighed in an EPA-audited laboratory. Field blank values were very low and were not subtracted from the $PM_{2.5}$ mass measurements. Agreement between the Harvard impactor and the personal monitor was determined by comparing the concentrations measured by collocated samplers.

Air concentrations of the target species were calculated using the analyte values determined from the samplers, the sampling duration of 48 hours, and the sampling rate.

Determining Air Exchange Rates

The investigators measured the number of air exchanges per hour in each home during the two seasons when indoor air was sampled in that home. An AER estimate is expressed as the number of times in 1 hour that the volume of indoor air is replaced by outdoor air. The AER was measured by releasing a tracer gas, perfluorinated methylcyclohexane, inside the home for 48 hours and collecting it on a passive capillary absorption tube. The method was able to detect up to 5.0 air exchanges per hour.

DATA QUALITY

The investigators conducted sampling and laboratory analyses to evaluate the performance of the samplers and the analytical methods used. The main quality control measurements and analyses they conducted included determining the method detection limit (MDL, the minimum concentration of a compound that can be measured and reported with 99% confidence), analytic precision (the variation in the analytic method under constant [laboratory] conditions), measurement precision (the variation of the analyses of individual species from duplicate samplers), and analytic accuracy (expressed as the percentage of difference between spiked and measured concentrations of independent standards). For each species, when concentrations of a compound were measurable on field blanks, the MDLs were expressed as 3 times the standard deviation of the field blank values.

Field positive controls, which consisted of samplers spiked with known amounts of each species, were placed in the indoor sampling racks to determine the amount of a species lost during the sampling period. Extraction efficiency of VOCs, determined by spiking a VOC badge with a known

quantity of each VOC, was expressed as the ratio of the measured concentration to the quantity spiked on the sampler. Species recovery for all carbonyls (except glyoxal and methylglyoxal) was calculated by exposing the active DNPH or the passive DNSH carbonyl sampler to a gaseous carbonyl in a test chamber and determining the ratio of the measured concentration to the known concentration generated in the chamber.

Summaries of the number of samples collected, MDLs, and comparisons of methods and samplers are provided below.

VOCs

The investigators measured 18 VOC species in about 550 indoor, outdoor, and personal samples. However, the stability of 1,3-butadiene and chloroprene on the OVM badges decreased significantly with time and the results of these constituents are not included in the analyses. For most VOCs (Table 3) the MDL ranged from 0.21 to 2.1 μg/m^3, but it was higher for 1,3-butadiene (3.1 to 4.0 μg/m^3) and toluene (6.7 to 7.1 μg/m^3). The MDL values determined at EOHSI were higher than those determined at the University of Texas by a factor of 2 for most VOCs, with the exception of methylene chloride, for which the MDL was about 7-fold higher at EOHSI (2.1 versus 0.29 μg/m^3).

The interlaboratory comparison of the analyses of sample extracts showed that, for most compounds, the difference (based on the slope of the measurements in the two laboratories) was less than 20% (Appendix Table C.2).

Some VOCs (chloroform, styrene, α-pinene, *d*-limonene, and *p*-dichlorobenzene) were detected in less than 40% of the outdoor samples from all three cities (Appendix Table E.1). The proportion of VOCS that were detectable in less than 40% of outdoor samples was 9 of 16 in Houston, 8 of 16 in Elizabeth, and 6 of 16 in Los Angeles.

Carbonyls

The investigators measured 16 carbonyls using the active DNPH method, but six were excluded from the analyses. These were *o*-tolualdehyde, *m*- & *p*-tolualdehydes, and dimethylbenzaldehyde due to low percentages of samples with detectable levels; acrolein and crotonaldehyde because of low recovery; and hexaldehyde because it could not be clearly identified. In total, each carbonyl species assessed with the active method was measured in 117 outdoor, 121 indoor, and 129 personal samples (Table 12).

The investigators measured 10 carbonyls using the passive DNSH method and reported data for all of them. In total, each carbonyl species assessed with the passive method was measured in 395 outdoor, 398 indoor, and 409 personal samples (Table 12). Seven compounds (formaldehyde, acetaldehyde, acetone, propionaldehyde, benzaldehyde, glyoxal, and methylglyoxal) were measured with both methods. Acrolein, crotonaldehyde, and hexaldehyde were measured only with the passive method.

The MDL values were low in general (Table 3). Exceptions were the acetone MDL values for all types of samples collected with the active method (2.75 to 13.38 μg/m^3) and the MDL values of all other carbonyls collected in vehicles (2.48 to 4.99 μg/m^3). All ten carbonyls measured with the passive method were detected in more than 90% of the all samples collected, except for acrolein (detected in 56% to 81% of samples; Tables E.1, E.2, and E.3) and crotonaldehyde (detected in 55% to 76% of samples). For carbonyls in indoor, outdoor, and personal samples analyzed with the active method, the percentage in which compounds were detected was lower except for formaldehyde and acetaldehyde.

Laboratory tests showed that more than 80% of most carbonyls were recovered with both samplers. Exceptions were acrolein and crotonaldehyde, for which recovery was 20% and 39%, respectively, for the active DNPH method and 60% and 76% for the passive DNSH method; this confirms that these species lack stability on DNPH (Table 8).

A side-by-side comparison of the two samplers showed that they agreed well for formaldehyde and acetaldehyde (r^2 of the regression line of 0.7), but poorly for other carbonyls measured (acetone, propionaldehyde, benzaldehyde, glyoxal, and methylglyoxal; r^2 ranging from 0.3 to 0.6). The slope of the regression line indicated that for all the species except formaldehyde, the passive sampler yielded higher values than the active sampler. (See Appendix D of the Investigators' Report.) The investigators attributed these differences to the stability of the carbonyl derivatives. They could also be due to errors in determining the sampling rate for the passive sampler and the different types of calibration standards used (described in the Methods section of the Report).

$PM_{2.5}$

The investigators report the $PM_{2.5}$ concentrations for 334 outdoor, 326 indoor, and 280 personal samples (Tables 13, 14, and 15). All $PM_{2.5}$ mass determinations exceeded the MDL (which was also very low). When the personal monitor and Harvard impactor were collocated indoors at 14 homes for comparison, the personal monitor consistently yielded $PM_{2.5}$ concentrations that were 18% higher than those from the Harvard impactor (based on the regression analysis; Figure 4). At a median concentration of 38 μg/m^3, the difference ranged between 1% and 16%.

DATA ANALYSIS

The Investigators' Report includes several descriptive univariate distribution analyses and bivariate scatter plots showing (1) some basic characteristics of the RIOPA data set; (2) indoor, outdoor, and personal exposure concentrations (and in-vehicle air concentrations for carbonyl compounds); (3) possible trends in the data related to season and to housing characteristics; and (4) personal concentrations of paired adult–child data. Individual species data from the three cities were combined for these analyses. (Data for each city are presented in Appendix E).

Standard paired comparison methods were used to estimate differences between paired data (such as outdoor and indoor concentrations, or adults and children). Rank tests (Kruskal-Wallis) were used for comparisons of subpopulations when the measurements did not appear to be normally distributed and the nonnormality could not be removed by a log transformation; otherwise *t* tests were used.

A mass balance model was used to estimate the contributions of indoor and outdoor sources to the indoor concentration for each species. The model represents the indoor concentration as the sum of two terms: (1) the concentration contributed by outdoor sources, and (2) the concentration contributed by indoor sources. The contribution from outdoor sources to indoor air (referred to as the fractional outdoor contribution) was calculated from the outdoor concentration of a given species corrected for the infiltration factor and was expressed as the fraction of the species' total indoor concentration. The home-specific infiltration factor was determined from the measured AER and the published data on species-specific decay rate and penetration through the building envelope. The concentration derived from indoor sources was calculated as the difference between the measured indoor concentration and the concentration contributed from outdoor sources. The indoor source contribution is dependent on the indoor source strength (*S*), the home volume, the AER, and the species decay rate. Using the model's equation, the investigators derived *S* (the rate at which a pollutant is generated from an indoor source in µg/hr).

For $PM_{2.5}$, the random component superposition (RCS) statistical model proposed by Ott and colleagues (2000) was used as a comparison to the mass balance model. Unlike the mass balance model, the RCS model uses the same infiltration factor for all homes and does not take into account differences in AER. In this case, the infiltration factor is represented by the slope of the regression of indoor and outdoor concentrations for all homes. The intercept of the line represents the indoor contribution.

RESULTS

Study Subjects and Housing Characteristics

The RIOPA study included 306 homes, 309 adults, and 118 children (including some siblings) in Elizabeth, Houston, and Los Angeles between 1999 and 2001. Overall retention of adult subjects for the repeat visit was between 77% and 88%. The percentage of children recruited varied among cities, with Houston having 62% of the child subjects, Los Angeles 19%, and Elizabeth 19%.

Among the three cities, the age ranges of subjects were similar, but differences in gender, cultural background, education, and work history were substantial (Table 10). Likewise, the proportions of the three main home types (single-family homes, mobile homes and trailers, and apartments) varied among cities (Table 9).

Indoor, Outdoor, and Adult Personal Exposure Concentrations in the Three Cities

For most VOCs and carbonyls, mean and median personal and indoor concentrations were similar, and both were higher than outdoor concentrations (see Commentary Table 3 for a summary within each city and for the whole data set). Exceptions were (1) median outdoor concentrations of acrolein, which in Houston were higher than indoor and personal concentrations; and (2) median and mean personal concentrations of acetone, which in Elizabeth and Houston were higher than indoor concentrations.

For $PM_{2.5}$, mean and median personal exposure concentrations were higher than indoor and outdoor concentrations, which were similar.

For all species, personal samples showed the largest differences between mean and median values, and they showed a larger range of measured concentrations than indoor and outdoor samples. In personal samples, compounds with the widest range of values were MTBE, tetrachloroethylene, *d*-limonene, *p*-dichlorobenzene, toluene, acrolein, and acetone. Acetone was especially high in personal samples from Elizabeth and Houston, and acrolein was especially high in personal samples from Houston.

For outdoor samples of VOCs, median concentrations differed slightly by city. Compared with Houston and Elizabeth, Los Angeles had a higher percentage of outdoor VOC samples with detectable concentrations; and it had higher outdoor concentrations of some VOCs, including MTBE, ethyl benzene, and the xylenes. All of these VOCs are emitted primarily from motor vehicles, gas stations, and oil refineries. For outdoor samples of carbonyls among the three cities, median concentrations were more variable than those for VOCs (with the exception of formaldehyde and acetone concentrations, which were similar in all three cities).

Commentary Table 3. Mean and Median Pollutant Concentrations in Outdoor, Indoor, and Personal Air (μg/m^3) by City

	Los Angeles			Elizabeth			Houston		
Species	Outdoor	Indoor	Personal	Outdoor	Indoor	Personal	Outdoor	Indoor	Personal
VOCs									
Methylene chloride	1.39/0.84	1.72/0.84	3.74/0.84	1.30/0.84[a]	1.63/0.84[a]	1.64/0.84	0.24/0.15[a]	3.46/0.44	3.62/0.51
MTBE	10.8/8.31	13.2/7.44	12.2/8.51	5.76/4.32	7.38/5.03	14.7/5.51	7.88/4.52	14.7/5.82	17.1/7.32
Chloroform	0.39/0.17[a]	1.58/0.92	8.49/0.87	0.30/0.17[a]	1.64/0.74	2.02/0.85	0.26/0.15[a]	2.31/1.32	2.27/1.33
Carbon tetrachloride	0.68/0.63	0.79/0.58	0.85/0.58	0.84/0.69	0.66/0.63	0.87/0.64	0.63/0.62	0.68/0.62	0.67/0.62
Benzene	2.52/1.98	3.00/2.05	3.10/2.28	1.44/1.22	2.52/1.65	2.80/1.76	2.48/1.94	4.85/3.06	4.82/3.13
Trichloroethylene	0.21/0.11[a]	0.25/0.11[a]	0.39/0.12	0.57/0.38	0.95/0.43	2.38/0.50	0.13/0.12[a]	0.18/0.12[a]	0.21/0.12[a]
Toluene	8.96/6.65	16.5/10.6	18.9/11.9	6.77/3.02	12.7/9.74	20.8/11.3	5.75/5.36[a]	16.8/10.3	18.0/13.1
Tetrachloroethylene	1.82/1.30	3.29/1.66	3.75/1.72	1.05/0.56[a]	1.26/0.56[a]	17.3/0.56	0.22/0.14[a]	1.02/0.29	1.39/0.36
Ethyl benzene	1.62/1.30	2.45/1.45	2.36/1.67	1.34/0.99	2.30/1.29	2.91/1.40	0.94/0.79	2.78/1.68	3.06/1.83
m- & p-Xylenes	4.91/3.56	6.88/4.16	7.07/4.54	3.23/2.34	6.52/3.18	7.93/4.04	2.69/2.23	8.47/4.55	9.07/5.10
o-Xylene	1.80/1.40	2.45/1.64	2.57/1.85	1.70/0.94	2.17/1.18	3.07/1.56	0.99/0.80	2.80/1.53	3.02/1.80
Styrene	0.58/0.17[a]	1.24/0.49[a]	1.16/0.49	0.53/0.17[a]	1.40/0.17	1.75/0.36	0.35/0.22[a]	1.58/0.67	1.61/0.84
α-Pinene	1.72/0.32	5.87/1.12	3.87/0.96	0.77/0.32[a]	3.93/1.45	5.03/1.96	0.26/0.11	4.86/0.14	3.83/0.14
β-Pinene	0.64/0.20[a]	2.98/0.79	2.72/0.73	0.64/0.18[a]	3.24/0.69	5.09/0.96	0.33/0.25[a]	7.84/4.03	8.26/4.16
d-Limonene	3.65/1.27[a]	22.0/7.31	48.2/7.65	2.46/1.27[a]	14.7/6.71	18.0/8.39	1.21/0.94[a]	54.0/20.8	55.0/22.4
p-Dichlorobenzene	1.38/0.72[a]	38.8/0.76[a]	14.9/1.15	3.77/0.72[a]	29.3/0.72[a]	26.5/1.85[a]	1.62/0.33[a]	132/2.02	120/3.42
Carbonyls (passive method)									
Formaldehyde	6.51/6.52	21.5/19.0	21.7/20.3	6.35/7.09	22.4/21.2	21.9/20.6	6.30/6.16	20.9/19.8	21.6/20.4
Acetaldehyde	5.89/5.27	24.1/20.7	22.7/20.1	8.88/7.86	16.3/14.3	17.3/15.1	5.93/4.70	30.0//22.3	29.4/22.3
Acetone	4.78/4.19	8.52/6.30	9.04/6.75	3.70/3.00	11.6/7.04	29.3/8.22	21.8/6.52	22.4/12.9	41.2/13.4
Acrolein	1.02/0.40	1.21/0.47	1.12/0.47	0.89/0.39	0.96/0.49	0.74/0.26[a]	17.9/0.95	3.08/0.92	40.0/0.91
Propionaldehyde	2.04/1.82	1.91/1.70	2.15/2.10	1.21/1.06	1.98/1.45	1.69/1.51	1.47/1.34	2.28/2.36	2.18/2.19
Crotonaldehyde	0.54/0.33	0.58/0.37	0.63/0.42	0.39/0.17[a]	0.69/0.40	0.63/0.30	1.45/0.35	0.83/0.55	2.59/0.63
Benzaldehyde	2.54/2.67	3.00/2.94	3.21/3.11	1.64/1.42	3.05/3.18	3.33/3.20	1.91/1.64	3.02/2.70	3.56/2.90
Hexaldehyde	2.49/2.23	3.96/3.51	4.28/3.95	1.45/1.35	3.81/3.21	4.10/3.58	3.09/2.52	5.95/5.66	7.70/5.38
Glyoxal	1.96/2.00	2.63/2.57	2.66/2.52	1.53/1.38	2.38/2.32	2.37/2.22	1.97/1.90	2.82/2.72	2.92/2.64
Methylglyoxal	2.18/2.07	3.09/2.83	2.89/2.83	1.74/1.81	2.33/2.47	2.30/2.28	2.27/2.29	3.21/3.22	3.11/2.98
$PM_{2.5}$	19.2/16.1	16.2/14.5	29.2/26.5	20.4/18.2	20.1/15.7	44.8/37.4	14.7/13.2	17.1/13.4	37.2/31.6

[a] Proportion of detected samples was ≤ 40%.

Several VOCs were present only at low levels in all environments and were not detected in many outdoor samples. The species detected in more than 60% of outdoor samples common to all three cities were MTBE, carbon tetrachloride, benzene, ethyl benzene, *m*- & *p*-xylenes, and *o*-xylene. MTBE had the highest outdoor concentrations.

Indoor concentrations of several VOCs and carbonyls differed among cities. The species with the highest indoor concentrations were the VOCs MTBE, toluene, and *d*-limonene and the carbonyls formaldehyde, acetaldehyde, and acetone.

Compared with Los Angeles and Elizabeth homes, Houston homes had several-fold higher indoor mean and median concentrations of several VOCs (including some of indoor origin such as β-pinene, *d*-limonene, and *p*-dichlorobenzene) and of some carbonyls (acetone and acrolein).

Personal exposure concentrations for several VOCs and some carbonyls also differed, especially between Houston and the other two cities. In particular, Houston subjects had very high personal exposures to β-pinene, *d*-limonene, *p*-dichlorobenzene, and several carbonyls; these reflect higher indoor concentrations.

Among the three cities, indoor and outdoor $PM_{2.5}$ concentrations differed only slightly, but differences in personal exposures were more pronounced.

In-Vehicle Concentrations of Carbonyls

Most in-vehicle samples were collected in Los Angeles (72), followed by Houston (33), and Elizabeth (10). Formaldehyde was detected in 76% to 100% of the samples in each city; acetaldehyde was detected in less than 30% of samples from Elizabeth and Houston but in 86% of samples from Los Angeles.

For the three cities combined, the in-vehicle concentrations of these two species (formaldehyde and acetaldehyde) had wider ranges than the indoor, outdoor, and personal exposure concentrations. The in-vehicle concentrations of formaldehyde (mean 39.7 $\mu g/m^3$; median 20.2 $\mu g/m^3$) and acetaldehyde (mean 25.2 $\mu g/m^3$; median 5.92 $\mu g/m^3$) were higher than the outdoor concentrations (3 $\mu g/m^3$ approximate mean and median for both species). This trend persisted when data were broken down by city.

Comparison of Personal Exposures for Adults and Children

For all three cities combined: (1) personal VOC samples were collected from 107 children during the first visit and 102 children during the second visit; (2) personal carbonyl samples were collected from 81 and 99 children, respectively; and (3) personal PM samples were collected from 14 and 13 children, respectively. The majority of children that contributed VOC and carbonyl data were in Houston (65% and 63%, respectively), whereas most of the children that contributed $PM_{2.5}$ data were in Los Angeles (85%). The authors reported small but significant differences between paired adult–child concentrations for some carbonyls (acrolein and formaldehyde) and some VOCs (MTBE and toluene). In general, however, the median adult and child personal exposures were similar for all species. For $PM_{2.5}$ the sample size for children was insufficient for analysis.

Air Exchange Rates

Homes had AERs that ranged from 0.14/hr to 4.75/hr in Los Angeles, 0.11/hr to 4.48/hr in Elizabeth, and 0.08/hr to 4.3/hr in Houston. The median AER was substantially lower for Houston homes (0.47/hr) than for Los Angeles homes (0.87/hr) and Elizabeth homes (0.88/hr). In Houston the median AER was higher in winter and fall; in Los Angeles, in spring; and in Elizabeth, in summer and winter. Some differences in AERs were noted among types of homes. For example, mobile homes (in Houston and Los Angeles) had slightly higher median AERs than the other types of homes. Single-family homes in Los Angeles had higher median AERs than multiple-family homes or apartments. In all three cities the newest homes (built after 1995) had the lowest median AERs; Los Angeles had the highest proportion of new homes.

Outdoor Contributions to Indoor Concentrations of Pollutants

For all three cities combined, the mass balance model showed that, for the VOC species MTBE, carbon tetrachloride, and trichloroethylene, 100% of indoor concentrations were contributed by outdoor air; for benzene it was 90%. Accordingly, these species also had the lowest indoor source strengths. Those with the lowest outdoor contributions were *d*-limonene, β-pinene, α-pinene, and chloroform (13% to 31%). For the remaining VOCs, the median fractional outdoor contribution to indoor concentrations ranged from 50% to 74%.

For carbonyls, the fractional outdoor contributions to indoor concentrations were lower than for VOCs (19% to 61%) and the indoor source strengths were higher. Of the carbonyls, formaldehyde and acetaldehyde had the lowest fractional outdoor contributions (and the highest indoor source strengths) and acrolein, crotonaldehyde, and propionaldehyde had the highest outdoor contributions.

For the whole $PM_{2.5}$ data set, the median fractional outdoor contribution to indoor concentrations was 56% or 61%; these fractions depended on the values used to correct for penetration through the building envelope and as the decay factor.

DISCUSSION AND CONCLUSIONS

The investigators measured the concentrations of air toxics (VOCs and carbonyls) and of $PM_{2.5}$ in three urban locations with different types of pollutant sources in order to understand how specific sources may impact indoor and personal exposure. The study was carefully conducted with good quality-control procedures and quality assurance confirmation by an independent auditor. It is unique in that a large number of compounds, including many carbonyls not measured in previous studies, were analyzed indoors, outdoors, and in personal samples from the same groups of subjects and their homes in three cities.

For each city and for the three cities combined, the Investigators' Report (1) documents important features of the study population (such as age, gender, and type of housing); (2) describes data quality; and (3) provides summary statistics and distributions of measured concentrations of VOCs, carbonyls, $PM_{2.5}$ mass, and residential AERs. It also includes the in-vehicle concentrations of selected carbonyls. Finally, the Investigators' Report provides estimates of the contributions of outdoor pollutants to indoor concentrations for all homes in the three cities combined. The compositional analysis of $PM_{2.5}$ samples, which forms Part II of this Research Report (*Part II. Analyses of Concentrations of Particulate Matter Species*), adds new information on the relations of PM components in indoor, outdoor, and personal air samples for this large group of subjects. The RIOPA data set extends our understanding of personal exposures to air toxics and PM and will be an important resource for future analyses and research.

The RIOPA study was designed to evaluate the impact of residential proximity to sources of pollutants, which was the main criterion by which geographic areas and clusters of homes were selected. These analyses are currently under way and will be published elsewhere.

The investigators also collected information on the subjects' activity patterns (time spent in various locations and activities) and factors that might influence the outdoor/indoor ratio of pollutants (distance from outdoor sources, type and usage of residential ventilation systems, and indoor sources).

SELECTION OF HOMES AND SUBJECTS

The selection of homes and subjects was not probability-based; rather, homes close to outdoor sources were preferentially sampled in an effort to examine the impact of possibly high exposures (described in Appendix A). A clear definition of the preferential sampling scheme would have been helpful. For example, the authors note that homes in Elizabeth were chosen within 200 m of the sources, which included gas stations, dry cleaners, and highway Routes 1/9 and 27. For future analyses of the association between exposure and source proximity, it is important that actual distances between homes and sources be determined and a consistent categorization of proximity be developed. This will help establish gradients of the air toxics concentrations associated with their sources.

Subjects were recruited using several methods of convenience. Between 22% and 33% of subjects and homes left the study before the second round of sampling. A discussion of the success rate of the different recruiting approaches and of the possible biases associated with the loss of subjects would provide insights into designing better recruitment methods in the future.

The types of housing sampled and the characteristics of the subjects (such as gender, cultural background, and education level) differed across cities and the proportions were not representative of the general population. Up to 80% of the adults were female. None of the subjects smoked. Information about the subject's possible exposure to environmental tobacco smoke was collected on the questionnaires, but the Investigators' Report does not discuss this.

Because of these differences, comparing results among the three areas, extrapolating the numeric results obtained in this study to the general population, or attributing them to a given city or region must be considered with caution.

DATA QUALITY

The overall quality of data for most air toxics measured was good. The quality is similar to that of other studies: the NHEXAS study of exposure to VOCs conducted in a group of adults and their homes in six Midwestern states (Pellizzari et al 2001b); a group of children and their homes in the Minneapolis–St Paul area (Adgate et al 2004); and a study of exposure to VOCs in two communities with different types of sources in South Baltimore (Buckley 2005).

For most VOCs, the mean MDLs were below 2.1 $\mu g/m^3$, but were higher for 1,3-butadiene (3.1 to 4 $\mu g/m^3$) and toluene (6.7 to 7.1 $\mu g/m^3$) (Table 3). The two laboratories that conducted the analyses differed in their determinations of MDLs for most VOCs. Given this, it would have been better if only one laboratory had performed the analyses and thus

achieved more consistent detection limits. However, it is well known that MDL values are variable and can be affected by the volume of air collected, the instrument used for collection, and the solvent (which may have background levels of the analyte of interest). A thorough discussion of the issues related to variability in the MDLs would have been helpful in light of the potential this data set has for further analyses. Details on measured concentrations below the MDL (which were all reported as half the MDL) would have provided more information on the variance of the low values and the effects of this variance on the estimates reported in the study.

The range of MDLs for most VOCs appears to be consistent with those reported in the studies mentioned above. The problem of a high MDL range for toluene has been reported by Chung and colleagues (1999b) and by Adgate and associates (2004). Others, however, have reported lower ranges possibly because of using longer sampling times (Pellizzari et al 2001b).

The OVM badge used to measure VOCs, which has been characterized for performance by some investigators of the RIOPA study (Chung and colleagues 1999a,b), appears to have performed well for several VOCs, but less well for others. For example, the low carbon tetrachloride values (0.6 to 0.8 μg/m^3 for all measured concentrations) appropriately reflect the fact that this chemical has been banned from consumer products. The ratios of outdoor concentrations of *m- & p*-xylene to *o*-xylene to ethyl benzene of 3:1:1 are consistent with those in other studies (Wallace et al 1987; Adgate et al 2004; Mukerjee et al 2004; Sexton et al 2004). However, toluene (generally the most abundant of the benzene, toluene, ethyl benzene, and xylene species) was detected in less than 60% of the outdoor samples in each of the three cities; this may have resulted from high blank values and a high MDL. The instability of 1,3-butadiene and chloroprene on the OVM substrate prevented measuring them; in agreement, however, none of the other studies that used the OVM badge have reported concentrations of 1,3-butadiene.

As part of the original OVM evaluation, Chung and coworkers (1999b) found that recoveries of compounds from OVM badges were specific to each compound and were generally lower at low concentrations and at high temperatures and humidity; they were similar to the recoveries from an active charcoal sampler at the tested concentration of 200 μg/m^3 under similar conditions and yielded comparable measurements. The exceptions were 1,3-butadiene and styrene, for which the OVM badge performed better than the active charcoal sampler. In the NHEXAS study in Arizona, Gordon and colleagues (1999), however, found that the OVM badge appeared to underestimate concentrations by about 40% compared with an active VOC sampling method for the two species examined (benzene and toluene). This was also corroborated by Kinney and associates (2005) when they collocated OVM badges with active thermal desorption tubes. The lower concentrations detected with OVM badges was attributed, at least in part, to the uncertainty in the sampling rate used to calculate the concentrations from the badges. Sampling rate can vary depending on air velocity across the face of the badge and can be affected by temperature and humidity (Chung et al 1999b). These factors could contribute to uncertainty about the concentrations reported from the RIOPA study. A method to validate the sampling rates for OVM badges should be considered.

In contrast with these studies that suggest a negative bias between OVM badges compared with active sampling methods, a comparison between a dual-pad OVM badge and a continuous gas-chromatograph sampler yielded a good agreement for toluene, benzene, and *o*-xylene. This suggests that the dual-pad OVM was capturing material that may have volatilized from the single-pad OVM used in RIOPA and other studies described above (Mukerjee et al 2004).

For carbonyls, the MDL values found with both the active DNPH and passive DNSH methods were less than 1.9 μg/m^3; exceptions were acetone (by both methods) and all the carbonyls from in-vehicle samples (measured with the active method only) (Table 3). Possible reasons for these exceptions are not discussed in the report. The passive method for measuring carbonyls provided better recovery of acrolein and crotonaldehyde than the active method (Table 8). The passive sampler was developed as part of this study; its performance has not been characterized by other researchers or in other studies.

No federal reference methods are available against which the VOC and carbonyl samplers could have been evaluated. Their performance varies across studies and the levels reported depend on the estimated sampling rate used (which is difficult to determine for passive samplers [and may vary from person to person]) and on different methods of correcting final concentrations for extraction efficiency or recovery (the RIOPA VOC data were not corrected). Also, different studies have used different samplers, sampling protocols, and populations. Thus the absolute values obtained in this study cannot be directly compared with those obtained in other studies.

For $PM_{2.5}$ measurements data quality was good: The MDL was very low and measurement precision was around 17% (this is intermediate among values reported in other studies; Thomas et al 1993; Ozkaynak et al 1996; Williams et al 2000). A comparison between the two $PM_{2.5}$ samplers showed the personal monitor consistently registered 1% to

16% higher concentrations than those measured with the Harvard impactor. A similar positive bias of the personal monitor compared with the Harvard impactor has been reported by Williams and colleagues (2000), Liu and coworkers (2003), and Geyh and associates (2004). Note, however, that biases may be associated with the Harvard impactor as well. Because neither the personal monitor nor the impactor have been compared with a federal reference method in this study, we do not know the performance of each sampler or the appropriate corrections to make.

CONCENTRATIONS MEASURED

The investigators provide summaries of the mean and median indoor, outdoor, and personal air concentrations and the range of values obtained for 16 VOCs, 10 carbonyls, and $PM_{2.5}$ (Tables 13, 14, and 15). The analyses of the aggregate data suggest some trends that will need to be verified by more detailed analyses. The distribution of values indicates a large range of concentrations of the measured compounds.

Several VOCs were present only at low levels in all environments and were not detected in many of the samples collected in outdoor air. For some of the analyses, the investigators excluded species for which more than 60% of the values were below the MDL, but did not explain the rationale for the choice of this cut point. The species detected in more than 40% of the outdoor samples in all three cities were MTBE, carbon tetrachloride, benzene, ethyl benzene, *m- & p*-xylenes, and *o*-xylene. Median and mean outdoor concentrations of MTBE, ethyl benzene, *m- & p*-xylenes, and *o*-xylene, which originate primarily from mobile sources, were slightly higher in Los Angeles samples than in Elizabeth and Houston samples. Although cities with different types of sources were chosen and homes near sources were preferentially sampled, generally the range of values measured was remarkably similar.

In contrast, large intercity differences in the concentrations of many of the VOCs measured in this study have been noted in the more in-depth analyses of the TEAM study data from five cities (Rappaport and Kupper 2004). In the more recent Toxic Exposure Assessment: A Columbia/Harvard (TEACH) study of levels of VOCs and carbonyls outside and inside homes of inner-city high school students in New York City and Los Angeles, the authors found a clear city-to-city difference: for most VOCs considered to be predominantly from outdoor sources, the median outdoor levels were twice as high in Los Angeles as in New York (Sax et al 2004).

VOC concentrations (including those for benzene, toluene, ethyl benzene, and xylene) were higher indoors and in personal exposure concentrations than in outdoor air, which suggests that they were, at least in part, generated indoors. The species with the highest indoor concentrations were MTBE, toluene, α-pinene, and *d*-limonene. The investigators concluded that the higher indoor concentrations of MTBE may be associated with evaporative emissions from vehicles parked in attached garages. This conclusion is reasonable because (1) the mass balance model yielded indoor source strengths of zero, and (2) there are no known indoor sources of MTBE. Nevertheless, this conclusion could be strengthened by additional analyses including type of home.

Measurement of carbonyl concentrations was a unique feature of this study. This is the first study to measure concentrations of acrolein and crotonaldehyde in indoor, outdoor, and personal air samples. These are highly reactive species produced from atmospheric reactions of VOCs. Some variability was noted in the outdoor levels of all carbonyls among the three cities, perhaps as a result of differences in atmospheric reactions, fuel composition, and stationary sources; but the roles of these variables were not determined. Intercity differences in indoor levels were also found. In particular, outdoor and indoor concentrations of acetone and acrolein were higher in Houston than in the other two cities, possibly due to higher outdoor temperatures and consequently higher photochemical activity. It is not clear what outdoor sources contributed to the high outdoor concentrations of acetone.

Like VOC levels, median indoor carbonyl levels were generally higher than outdoor levels, which is consistent with recent measurements made by Sax and coworkers (2004) in New York City and Los Angeles homes. Indoor and personal median exposure levels were similar to each other for all carbonyls in all three cities with the exception of mean concentrations of acetone, which were higher in personal air samples than in indoor air, especially in Elizabeth and Houston. A possible source of acetone in personal air samples is the subject's own breath, because acetone is a product of a number of endogenous metabolic reactions; but the difference among the cities' outdoor concentrations argues against this as the sole determinant of exposure.

For the carbonyls formaldehyde and acetaldehyde, concentrations in vehicles were substantially higher than outdoors; this result needs to be considered in the context of the sampling strategy. First, the in-vehicle data were collected during short periods while driving (up to 8 hours) whereas the outdoor levels were integrated measurements made over 48 hours. Second, the outdoor samples were obtained outside the homes of the subjects, which may or may not have been near traffic. Finally, the in-vehicle measurements would have been affected by traffic patterns and time of day, but these are not discussed in the report.

For $PM_{2.5}$, outdoor and indoor concentrations differed little among the cities and between indoor and outdoor levels in each city. In a study of the homes of asthmatic children in seven US cities, Wallace and coworkers (2003) also noted small variations in the indoor and outdoor $PM_{2.5}$ concentrations across cities and suggested that the sources of indoor concentrations do not vary substantially from one city to the next.

Personal exposure concentrations of $PM_{2.5}$ varied among cities. The ratio of personal exposure to outdoor median $PM_{2.5}$ concentrations ranged from 1.6 in Los Angeles to 2.3 in Elizabeth and 2.4 in Houston; these ratios were higher than those measured in similar studies of large groups of adults in Toronto (Pellizzari et al 1999) and in Indianapolis (Pellizzari et al 2001a). In those studies, the authors attributed the differences between personal and outdoor levels mostly to smoking tobacco. Although the subjects in the RIOPA study did not smoke, information about their possible exposure to environmental tobacco smoke was collected on questionnaires but not discussed in the report. The authors note that their subjects were mostly women who spent much of their time at home, where they were exposed to PM while cooking and cleaning; these activities would have more impact on personal exposure levels than on general indoor levels of PM. Possible exposures outside the homes could also contribute to differences in personal exposure levels, however. The difference between the levels measured indoors by collocated personal monitors and Harvard impactors (in which measurements from the personal monitor were consistently higher) may have contributed to the difference observed, but would not by itself be enough to explain it.

OUTDOOR CONTRIBUTIONS TO INDOOR CONCENTRATIONS OF POLLUTANTS

The investigators used a mass balance model to determine, for each species, its indoor source strength and the fraction of the indoor concentration that was contributed by outdoor air. For this calculation they measured the AER for each home and estimated physical parameters of penetration and decay for each species.

The ranges of measured AERs were similar in the three cities, but the median rates were lower in Houston (0.47/hr) than in Los Angeles (0.87/hr) and Elizabeth (0.88/hr), possibly because air conditioners were more prevalent in the Houston homes. Further studies of air conditioner use may clarify this. Seasonal effects on AERs varied across cities, but no consistent pattern was observed. The authors noted that the difference between indoor and outdoor temperatures was a fairly good predictor of the AER pattern observed (higher indoor–outdoor temperature differences were associated with higher AERs) and hypothesized that convection may be the dominant mechanism of air exchange. This hypothesis is probably correct, but factors such as the placement of the tracer gas source, the collector used, the type of home, and whether windows were opened may all affect AERs. Uncertainty associated with the calculated rates was not determined. Wallace and colleagues (1991) discerned an effect of indoor–outdoor temperature differences on AERs as well, but found that having windows opened also had an effect. Sax and coworkers (2004) noted differences in AERs between New York and Los Angeles homes, even though the seasonal patterns in the two cities were similar.

The results from the mass balance model show that some of the measured VOCs (MTBE, benzene, carbon tetrachloride, and trichloroethylene) were primarily from outdoor sources, which contributed 90% to 100% of the indoor concentrations. Outdoor concentrations of other VOCs (chloroform, α-pinene, β-pinene, and *d*-limonene) and most carbonyls (including formaldehyde, acetaldehyde, and hexaldehyde) were primarily from indoor sources and outdoor sources contributed between 13% and 43% of the indoor concentrations. The carbonyls with the highest outdoor contributions to indoor concentrations were acrolein (63%), crotonaldehyde (61%), and propionaldehyde (50%). These values were fairly well correlated with the indoor source strengths. For $PM_{2.5}$, the mass balance model indicated an outdoor contribution of 60% of the indoor concentrations.

Overall, the outdoor contributions to indoor concentrations for VOCs, carbonyls, and $PM_{2.5}$ were consistent with those reported by several other studies. The source strength estimates for VOCs and several aldehydes were also in very good agreement with measurements reported for the same species in the TEACH study (Sax et al 2004).

To evaluate the impact of penetration and decay parameters on the estimated fractional outdoor $PM_{2.5}$ contribution to indoor concentrations, the investigators used different sets of values for these parameters without changing the AER. Overall, the median outdoor contribution ranged from 56% to 79% of indoor concentrations, which indicates that changing these parameters had some impact on the final results.

For $PM_{2.5}$ the authors also compared the results of the mass balance model with those obtained by applying the RCS model, which assumes a single infiltration factor determined as the slope of the regression of indoor and outdoor $PM_{2.5}$ concentrations. (The infiltration factor, referred to as the "attenuation factor" by Ott and colleagues [2000], accounts for deposition of particles as the outdoor air infiltrates indoors.) For all homes the infiltra-

tion factor was 0.46, which agrees with the 0.46 figure obtained with the mass balance model. This indicates that, in these homes, about half of the outdoor particles penetrated indoors. The agreement between the two models for $PM_{2.5}$ reflects that the calculation of the outdoor contribution for the whole data set was not substantially improved by using a home-specific infiltration factor based on the AER (as in the mass balance model) compared with using a constant infiltration factor (as in the RCS model). This conclusion was reached also by Ott and colleagues (2000); however, the RCS model does not provide information about the indoor source strength. In the Inner City Air Pollution Study (Wallace et al 2003), the investigators used the RCS model and found a range of infiltration factors across six cities and reported an average infiltration factor of 0.5 for all cities combined; this is consistent with the finding from the RIOPA study.

Similar comparisons of the mass balance and RCS models have not been conducted for air toxics. However, as shown in Table 20, using the mass balance model to determine the outdoor contributions to indoor concentrations resulted in median values that were in good agreement with those that can be estimated from the inverse of the ratios of the median indoor to the median outdoor concentrations. This agreement suggests that the RCS model (using the regression of all indoor and outdoor concentrations) would provide a reasonable approximation of the outdoor contributions of air toxics as well.

The analyses just described focused on the outdoor contributions to indoor concentrations of each pollutant. From the scatter plots of the measured concentrations of each species (Figures 9 through 12 and Appendix Figures F.1 and F.2), the investigators drew some conclusions regarding the outdoor and indoor contributions to personal air samples. The plots between personal exposure levels and outdoor levels for some VOCs and some carbonyls (MTBE, carbon tetrachloride, benzene, acrolein, and crotonaldehyde) showed many data points distributed along the 1:1 line for a large proportion of the subjects. This suggests that outdoor sources of these compounds were important contributors to personal exposures. The weaker correlations between personal exposure levels and outdoor levels for all other VOCs and carbonyls and for $PM_{2.5}$ suggest that personal exposures to these species were derived primarily from indoor sources (and personal activities). (These results agree with those from the mass balance model.) If supported by results from similar analyses with different populations, the findings about outdoor versus indoor contributions to personal exposure could have implications for the design and specifications of models currently used to estimate exposure to these air toxics.

Despite the broad agreement with other studies, the results of the mass balance model used in RIOPA may not be applicable to the general population or even to populations in the three cities. First, several critical assumptions were made regarding the penetration and decay parameters used in the model. Second, there could have been errors in determining AERs. Third, the high percentage of outdoor samples in which several VOCs were nondetectable would yield an unrealistic indoor–outdoor ratio for those VOCs. Finally, the investigators combined all the homes in the three cities in this analysis, which may have masked differences related to regional variations in housing characteristics and sources of pollutants.

ADDITIONAL ANALYSES

The RIOPA data set is rich with information for understanding the sources of air toxics and their effect on personal exposure; its full potential has yet to be realized. Analysis of source proximity is an important future goal, but additional data will be needed. These include meteorologic data and geographic identifiers of the potential sources of pollutants around the homes.

The existing data can also be used to characterize distributions of the measured concentrations by a variety of factors: for example, in-vehicle concentrations by season and traffic density indicators (if available); contribution of time spent in vehicles to personal exposure; relation between outdoor residential levels and central site monitors; and personal exposure concentrations by employment status and by time spent indoors, outdoors, and in vehicles.

Furthermore, the data collected in the RIOPA study can be used to evaluate (1) relations between concurrent measurements, such as spatial differences between outdoor measurements of different species, (2) relations between concurrent personal, indoor, and outdoor measurements of the same species, and (3) contributions of outdoor pollutant levels to indoor concentrations and personal exposure by city, type of housing, and the distance of residences from sources. In addition, it is important to identify and assess individual factors that may be associated with high personal exposures to describe possible subgroups that may be at greater risk.

CONCLUSIONS

This study generated a large database on the concentrations of air toxics and $PM_{2.5}$ for a large number of subjects and their homes selected on the basis of distances from various sources. Using passive samplers to measure air toxics enabled a large data set of concurrent measurements to be collected from indoor, outdoor, and personal air

samples. The VOC sampler performed well for most species; however uncertainties remain about outdoor levels of several VOCs because their low ambient levels, high limits of detection, or low extraction efficiencies made them difficult to measure. The inability of the passive OVM badge to measure 1,3-butadiene is also a limitation. These uncertainties suggest that new technologies or improvements in air sampling and analytic methods will be needed to draw further conclusions about some of these compounds.

The newly developed passive carbonyl sampler appeared to perform well for most species. $PM_{2.5}$ active samplers performed well, but a small bias was noted between the personal exposure monitor and the Harvard impactors used for outdoor and indoor measurements.

The data presented in the Investigators' Report are the results of descriptive analyses for each species measured. Values were highly variable for all species within and across the three cities. However, the overall relations that compare indoor, outdoor, and personal air samples for most compounds were similar for all three cities. This was unexpected given the wide variety of pollutant sources and weather. With a few exceptions, mean and median personal exposure and indoor levels of VOCs and carbonyls were similar and higher than the outdoor levels within the whole data set and within individual cities. Mean and median personal $PM_{2.5}$ exposure concentrations were higher than indoor and outdoor levels, and indoor and outdoor levels were very similar. The finding that personal exposure levels were higher than outdoor levels of these compounds is consistent with those of many other studies. Personal exposures of adults and children were similar for most compounds. The clustering of homes based on proximity to pollutant sources within a defined geographic area differed by city, as did the home-sampling schemes. Because the population of the study was not a probability-based sample, the results may not be extrapolated to the general population or attributed to a city or a region.

Time and budget constraints did not allow full use of the database; for example, determining how levels of different pollutants are related to each other and nearby sources for each single home was not done. As a first step in determining how types of sources may impact personal exposure, the investigators calculated the contributions of outdoor air to indoor air concentrations for each species for all homes combined. Several compounds with highly correlated and similar outdoor and indoor levels, low indoor source strengths, and high fractional outdoor contributions were identified as primarily of outdoor origin (eg, MTBE, carbon tetrachloride, trichloroethylene). Another group of compounds with elevated indoor levels compared with outdoor levels, high indoor source strengths, and low fractional outdoor contributions (eg, 1,2-dichlorobenzene, chloroform, styrene, α-pinene, β-pinene, *d*-limonene, formaldehyde, acetaldehyde, butyraldehyde, isovaleraldehyde, valeraldehyde, and hexaldehyde) were identified as primarily indoor origin. A third group of compounds (including *m- & p*-xylenes, *o*-xylene, propionaldehyde, acrolein, crotonaldehyde, glyoxal, methylglyoxal, and $PM_{2.5}$) showed intermediate values for indoor source strengths and for the fractional outdoor contributions to indoor concentrations, which indicates they are derived from both indoor and outdoor sources.

The results of the RIOPA study confirm and extend earlier findings by others for VOCs and $PM_{2.5}$ and have yielded new information for a large number of carbonyls. Few investigators have looked at personal, indoor, and outdoor concentrations of a suite of VOCs, carbonyls, and $PM_{2.5}$ in the same large set of subjects in multiple urban centers. The information on $PM_{2.5}$ composition in the RIOPA study (Part II of this Research Report) provides needed information about exposure to the components of PM. Overall, the data collected in the RIOPA study increase the database on the distribution of levels of a large number of air toxics and $PM_{2.5}$; these data can be used to assess whether these levels pose health concerns, to understand the sources of air toxics, and how individual factors may be associated with high exposures.

ACKNOWLEDGMENTS

The Special Review Panel thanks the ad hoc reviewers for their help in evaluating the scientific merit of the Investigators' Report. The Committee is also grateful to Lata Shirnamé-Moré (NUATRC) and Debra Kaden (HEI) for their oversight of the study, to Maria Costantini (HEI) and Lata Shirnamé-Moré for their assistance in preparing its Commentary, to Virgi Hepner and Genevieve MacLellan for science editing the Investigators' Report and this Commentary, and to Melissa R Harke, Kasey Oliver, Quentin Sullivan and Ruth E Shaw for their roles in publishing this Research Report.

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INSTITUTE

Charlestown Navy Yard
120 Second Avenue
Boston MA 02129-4533 USA
+1-617-886-9330
www.healtheffects.org

Number 130 Part I
November 2005

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Number 7
November 2005

PO Box 20286
Houston TX 77225-0286 USA
713.500.3450 Phone
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www.sph.uth.tmc.edu/mleland