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RESEARCH REPORT

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

Part II. Analyses of Concentrations of Particulate Matter Species

Barbara J Turpin, Clifford P Weisel, Maria Morandi, Steven Colome, Thomas Stock, Steven Eisenreich, Brian Buckley, and Others

Includes a Commentary by the Special Review Panel

Number 10
August 2007

MICKEY LELAND NATIONAL URBAN AIR TOXICS RESEARCH CENTER

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The Health Effects Institute is a nonprofit corporation chartered in 1980 as an independent research organization to provide high-quality, impartial, and relevant science on the effects of air pollution on health. To accomplish its mission, the Institute

- Identifies the highest-priority areas for health effects research;
- Funds and oversees the conduct of research projects;
- Provides intensive independent review of HEI-supported studies and related research;
- Integrates HEI's research results with those of other institutions into broader evaluations; and
- Communicates the results of HEI research and analyses to public and private decision makers.

Typically, HEI receives half of its core funds from the US Environmental Protection Agency and half from the worldwide motor vehicle industry. Frequently, other public and private organizations in the United States and around the world also support major projects or certain research programs. HEI has funded more than 250 studies in North America, Europe, and Asia that have produced important research to inform decisions regarding carbon monoxide, air toxics, nitrogen oxides, diesel exhaust, ozone, particulate matter, and other pollutants. The results of these studies have been published in more than 200 Research and Special Reports.

HEI's independent Board of Directors consists of leaders in science and policy who are committed to the public–private partnership that is central to the organization. The Health Research Committee solicits input from HEI sponsors and other stakeholders and works with scientific staff to develop the Five-Year Strategic Plan, select research projects for funding, and oversee their conduct. The Health Review Committee, which has no role in selecting or overseeing studies, works with staff to evaluate and interpret the results of funded studies and related research.

All project results and HEI Commentaries are widely communicated through HEI's website (*www.healtheffects.org*), annual conferences, publications, and presentations to 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 II

HEI HEALTH EFFECTS INSTITUTE

Pollutants in Indoor, Outdoor, and Personal Air: Composition of Particulate Matter

INTRODUCTION

Many epidemiologic studies have shown an association between exposure to particulate matter (PM) and increased morbidity and mortality. These types of studies often use ambient (outdoor) concentrations measured at fixed monitoring sites as a surrogate for personal exposure. However, the adequacy of this surrogate measure continues to be an important research and policy question, despite much recent research to address it. The factors that influence the relation between outdoor particle concentrations and personal exposure need to be better understood. This involves assessing: the similarities and differences in levels and characteristics of particles in various microenvironments; how outdoor particles contribute to indoor concentrations; and how individual activity patterns influence personal exposure and resulting dose.

HEI and NUATRC sought to fund research to (1) characterize personal exposure to particles in different indoor and outdoor microenvironments and in geographic locations that differ in types and sources of particles, topography, and climate; and (2) identify distinctive characteristics of particles that would improve exposure estimates in epidemiologic studies. Ideally, studies to address the second objective would determine particle characteristics (eg, concentration, size, and composition) and describe the relation between overall personal exposure and the surrogate measures of exposure used in many epidemiologic studies.

The Relationships of Indoor, Outdoor, and Personal Air (RIOPA) study was designed to provide such information for $PM_{2.5}$ (PM of 2.5 µm or smaller in aerodynamic diameter), a large number of volatile organic compounds (VOCs), and carbonyls. Dr Turpin's component of the larger project focused on $PM_{2.5}$ species—key constituents of $PM_{2.5}$ that include sulfur, organic and elemental carbon, polycyclic aromatic hydrocarbons (PAHs), chlordanes, trace elements, and functional groups (atoms attached to carbon that can influence a molecule's behavior). These analyses are presented here in Part II of this Research Report; Part I presents the analyses for the VOCs and carbonyls.

APPROACH

The RIOPA study addressed the hypothesis that outdoor sources contribute a substantial proportion of the pollutant concentrations in the indoor air and personal air (breathing zone) for residents who live near those sources. 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 approximately 100 homes with 100 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 their distance from various sources. Approximately 300 residents in 300 homes participated in the full RIOPA study; samples from 219 homes and their residents were analyzed for $PM_{2.5}$ and its components.

Dr Turpin and colleagues aimed to: (1) characterize and compare indoor, outdoor, and personal $PM_{2.5}$ mass composition; (2) estimate the contribution of outdoor $PM_{2.5}$ and its components to indoor concentrations and to personal exposures using residential air exchange rates (AERs); and (3) conduct exploratory analyses of indoor and personal $PM_{2.5}$ concentrations to identify particulate sources.

$PM_{2.5}$ filter samples were collected inside and directly outside each home. Organic $PM_{2.5}$ sampling artifacts were also measured. Gas- and particle-phase samples were collected for measurement of selected semivolatile organic compounds. Personal

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 Dr Barbara J Turpin at Rutgers University, New Brunswick NJ. The following Research Report (HEI Number 130 Part II; NUATRC Number 10) contains both the detailed Investigators' Report and a Commentary on the study prepared by a Special Review Panel from both funding organizations.

$PM_{2.5}$ filter samples were collected using a personal environmental monitor worn by each participant.

Samples or subsets of samples were analyzed for $PM_{2.5}$ mass, elements, organic and elemental carbon, functional groups, PAHs, and chlordanes.

AERs, expressed as the number of indoor air volumes replaced each hour by outdoor air, were measured using a technique developed specifically for application to relatively small spaces, including homes. Investigators measured the number of air exchanges per hour at each home during each sampling period.

The investigators used AERs to calculate the contribution of outdoor air to indoor $PM_{2.5}$ mass using three methods, each with increasingly more realistic assumptions: one that assumed the infiltration factor was constant across homes; one that assumed the infiltration factor varied according to measured AERs for each home; and one that estimated an independent infiltration factor for each home and sampling day using measured $PM_{2.5}$ species, AER, and housing characteristics.

RESULTS AND INTERPRETATION

A number of analyses quantified and compared indoor, outdoor, and personal exposure concentrations of $PM_{2.5}$ and its components. Some key results are summarized below.

When data from all three cities were combined, the median $PM_{2.5}$ concentrations indoors and outdoors were about the same and personal concentrations were about twice as high.

Among the cities and within each city, indoor and outdoor particle concentrations differed little, whereas differences in personal exposures were more pronounced.

The ratio of personal exposure to outdoor median concentrations varied among cities; it was notably lowest in Los Angeles (1.6 vs 2.3 in Elizabeth and 2.4 in Houston). This variation could reflect differences in the strength of indoor sources, AERs, and personal activities. The degree of correlation between indoor and outdoor concentrations did not have much impact on correlations with personal $PM_{2.5}$ concentrations.

When specific constituents of $PM_{2.5}$ were assessed, organic matter dominated $PM_{2.5}$ concentrations both indoors and outdoors. Differences in the composition of outdoor, indoor, and personal $PM_{2.5}$ were observed, however. Indoor organic $PM_{2.5}$ concentrations were nearly twice as high as outdoor concentrations, which indicates the importance of indoor sources.

Similarly, chlordane concentrations were higher indoors than outdoors. This is most likely due to strong indoor emissions from volatilization of termiticides used during home construction.

In contrast, elemental carbon concentrations indoors and outdoors were well correlated, with indoor concentrations generally lower than outdoor concentrations. This suggests that indoor emissions of elemental carbon were low.

The concentrations of PAHs were substantially more variable indoors than outdoors. Phenanthrene was consistently the largest measured contributor to PAH mass in both indoor and outdoor air.

The methods used to estimate how much outdoor sources of $PM_{2.5}$ contributed to indoor concentrations produced broadly consistent results: over 60% of indoor concentrations in Los Angeles, 70% in Elizabeth, and over 40% in Houston. $PM_{2.5}$ of outdoor origin contributed much less to personal $PM_{2.5}$ exposure—approximately 25% to 33%.

As shown above, outdoor contributions to indoor concentrations were much lower for Houston homes than for those in Los Angeles and Elizabeth, and the same pattern was observed for the outdoor contribution to personal exposure. The investigators suggest that this difference could be attributed to the more common use of air conditioning in Houston, which tends to reduce air exchanges; they did not test this hypothesis, however.

The investigators attempted to characterize a source of exposure error in epidemiologic time-series studies, namely variations in particle infiltration behavior. Three approaches were used to explore how AERs, particle properties, and housing characteristics can influence particle infiltration. When used in conjunction with concentrations measured at fixed monitoring sites, information on AERs can minimize uncertainty in estimates of exposure to $PM_{2.5}$ of outdoor origin.

CONCLUSIONS

Dr Turpin and her colleagues have contributed important information by (1) characterizing and comparing the composition of indoor, outdoor, and personal $PM_{2.5}$ in the three cities; and (2) estimating the contribution of outdoor $PM_{2.5}$ and its components to

indoor and personal exposures. This is one of the most comprehensive studies to characterize $PM_{2.5}$ exposures and one of the first to measure $PM_{2.5}$ functional groups.

Although the lack of a population-based sampling strategy limits the generalizability of the results for broad epidemiologic analyses, the compositional data provide insight on exposure to $PM_{2.5}$ constituents for a large number of subjects and homes selected on the basis of distance from various outdoor sources.

This study has generated a rich database that can be used to identify what levels of exposure could be related health concerns, the sources of air toxics, and factors associated with high exposures. HEI and NUATRC are currently developing additional opportunities to explore aspects of these data.

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STATEMENT

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

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 outside 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

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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 outdoor, indoor, 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 and Dentistry of New Jersey and at the Environmental and Occupational Health Sciences Institute [EOHSI]) proposed to monitor outdoor, indoor, 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 measurements of outdoor and indoor concentrations of some aldehydes and PM with an aerodynamic diameter of 2.5 µm or less ($PM_{2.5}$) for half the homes. Coinvestigators were Junfeng (Jim) Zhang (affiliated with the same institutions); Barbara Turpin (EOSHI and Rutgers University); Thomas Stock and Maria Morandi (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", which sought 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 outdoors and indoors 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", which requested studies that would characterize personal exposure to PM in different indoor and outdoor environments and geographic locations and also 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, and $PM_{2.5}$ concentrations; published in 2005) and *Part II: Analyses of Concentrations of Particulate Matter Species* (the compositional analysis of $PM_{2.5}$; this volume). 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

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INVESTIGATORS' REPORT

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

Part II. Analyses of Concentrations of Particulate Matter Species

Barbara J Turpin, Clifford P Weisel, Maria Morandi, Steven Colome, Thomas Stock, Steven Eisenreich, Brian Buckley, and Others

ABSTRACT

During the study Relationships of Indoor, Outdoor, and Personal Air (RIOPA*), 48-hour integrated indoor, outdoor, and personal air samples were collected between summer 1999 and spring 2001 in three different areas of the United States: Elizabeth NJ, Houston TX, and Los Angeles County CA. Air samples suitable for analyzing particulate matter 2.5 µm or smaller in aerodynamic diameter ($PM_{2.5}$) were collected in 219 homes (twice in 169 homes). Indoor and outdoor air samples suitable for gas-phase and particle-phase organic analyses were collected in 152 homes (twice in 132 homes). Samples or subsets of samples were analyzed for $PM_{2.5}$ mass, organic functional groups, elements, organic carbon (OC), elemental carbon (EC), gas-phase and particle-phase polycyclic aromatic hydrocarbons (PAHs), and chlordanes. Air exchange rate (AER), temperature, and relative humidity were measured for each residence; questionnaire data and time-activity information were collected from the participants.

Median indoor, outdoor, and personal $PM_{2.5}$ mass concentrations were 14.4, 15.5, and 31.4 µg/m^3, respectively. Personal $PM_{2.5}$ concentrations were significantly higher and more variable than indoor and outdoor concentrations. Several approaches were applied to quantify indoor $PM_{2.5}$ of ambient (outdoor) and nonambient (indoor) origin, some using $PM_{2.5}$ mass concentrations and others using $PM_{2.5}$ species concentrations. PM of outdoor origin was estimated in three ways using increasingly accurate assumptions. Comparing estimates from the three approaches enabled us to quantify several types of errors that may be introduced when central-site PM concentrations are used as surrogate estimates for PM exposure. Estimates made using individual measurements produced broader distributions and higher means than those made using a single infiltration factor for all homes and days. The best estimate (produced by the robust regression approach) of the mean contribution of outdoor $PM_{2.5}$ to the indoor mass concentration was 73% and to personal exposure was 26%. Possible implications of exposure error for epidemiologic assessments of PM are discussed below.

Organic particulate matter was the major constituent of $PM_{2.5}$ generated indoors. After correcting for artifacts, it constituted 48%, 55%, and 61% of $PM_{2.5}$ mass inside study homes in Los Angeles, Elizabeth, and Houston, respectively. At least 40% but probably closer to 75% of this organic matter, on average, was emitted or formed indoors. Functional group analysis provided some insights into the composition and properties of the indoor-generated organic $PM_{2.5}$. Chlordane, a very minor but mutagenic semivolatile organic mixture previously used as a termiticide, was found to be mostly of indoor origin. High emission rates were most frequently found in homes built from 1945 to 1959.

Analysis of the change in gas–particle partitioning during transport of outdoor PAHs to indoor environments illustrated that chemical thermodynamics can alter the concentration and composition of outdoor PM as it is transported indoors. (This has been previously noted for nitrate [Lunden et al 2003].) In epidemiologic studies that rely on central-site monitoring data, such transformations may result in measurement error, and this possibility warrants further investigation.

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

This Investigators' Report is Part II of a Research Report published by the Health Effects Institute (Report 130) and the Mickey Leland National Urban Air Toxics Research Center (Report 10). The Report also includes a Commentary written by a Special Review Panel jointly selected by both organizations, a Preface, and a Statement synopsis about the research project. Correspondence concerning the Investigators' Report may be addressed to Dr Barbara J Turpin (*turpin@envsci.rutgers.edu*), Department of Environmental Sciences, Rutgers University, 14 College Farm Road, New Brunswick NJ 08901.

(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.

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INTRODUCTION

BACKGROUND

Numerous epidemiologic studies have shown a positive association between outdoor PM concentrations and cardiovascular and respiratory morbidity and mortality (Norris et al 1999; Schwartz et al 1999, 2002; Klemm et al 2000; Goldberg et al 2001; US Environmental Protection Agency [EPA] 2004). Adverse effects have been more closely associated with fine particles ($PM_{2.5}$) than coarse particles (PM_{10}) (Schwartz and Dockery 1996; Wilson and Suh 1997; Schwartz et al 1999; Klemm et al 2000). Because exposure is necessary to establish a causal association between ambient PM exposure and adverse health effects (Zartarian et al 1997), these epidemiologic findings have prompted initiation of many exposure studies (Wallace 1996). Personal exposure studies, however, have consistently found poor correlations between ambient $PM_{2.5}$ concentrations and individuals' personal exposure (Sexton et al 1984; Spengler et al 1985; Morandi et al 1988; Wallace 1996; Pellizzari et al 1999; Lachenmyer and Hidy 2000; Oglesby et al 2000; Meng et al 2005).

The poor correlations in personal exposure studies were initially used to argue that ambient PM measurements are a poor surrogate for exposure to PM and to question the conclusions of epidemiologic studies (Spengler et al 1985; Lipfert and Wyzga 1997; Gamble 1998). In response, Wilson and Suh (1997) and Mage and associates (1999) characterized the seeming contradiction between the exposure studies and epidemiologic findings as a logical syllogism. They argued that ambient particles differ substantially in composition and properties from particles generated in other microenvironments, and that epidemiologic studies use central-site ambient PM concentrations as a surrogate for exposure to PM of outdoor origin, not as a surrogate for total PM exposure. This work motivated additional exposure analyses to (1) determine where and how people are exposed to particles outdoors and in microenvironments, (2) quantify exposure errors that arise from using a central-site PM concentration as an exposure surrogate, and (3) understand the effect of such errors on epidemiologic conclusions (Leaderer et al 1999; Abt et al 2000b; Lachenmyer and Hidy 2000; Oglesby et al 2000; Ott et al 2000; Patterson and Eatough 2000; Sarnat et al 2000, 2002; Williams et al 2000a; Koponen et al 2001; Adgate et al 2002, 2003; Kousa et al 2002). Estimates of PM of ambient origin can also be used to test and refine predictive exposure models. Ultimately, validated predictive models that link sources of particles to exposures will facilitate the development of more effective strategies for public health protection.

People are exposed to particles generated indoors, outdoors, in other microenvironments, and through personal activities. Particles generated through different mechanisms vary in composition and presumably in toxicity (Monn and Becker 1999; Long et al 2001b). Thus for decades chemical characterization has been used to resolve ambient PM source contributions and to develop effective strategies for controlling PM levels outdoors. Despite concerns about the health effects of PM exposure, much less has been done to chemically characterize typical or high-level personal exposures and microenvironmental PM concentrations.

The major components of ambient $PM_{2.5}$ are sulfate, nitrate, OC, ammonium, and water (EPA 2004). Soil dust, EC, and other trace elements are also present in small quantities. In the United States, sulfate dominates the aerosol mass in the east, whereas nitrate and OC are the largest constituents in the west. Secondary sulfate and nitrate are formed in the atmosphere from reactions involving precursor gases such as sulfur dioxide, ammonia, and oxides of nitrogen (Seinfeld and Pandis 1998). Sulfate concentrations are highest in the summer; in the winter, lower sulfate concentrations and lower temperatures can result in higher nitrate concentrations. EC is emitted in particulate form (primary) from sources of combustion. Both primary OC from emissions and secondary OC formed in the atmosphere are present in ambient air (Turpin et al 2000).

Ambient organic $PM_{2.5}$ is composed of thousands of compounds with a wide variety of vapor pressures and chemical properties. Typically, rigorous molecular-level analysis of ambient samples can account for only 10% to 30% of the organic $PM_{2.5}$ mass (Turpin and Lim 2001; Rogge et al 1993). Alkanes, aldehydes, alkenes, carboxylic acids, ketones, and PAHs are among the commonly identified species in outdoor $PM_{2.5}$ (Turpin et al 2000). The evidence that oligomers also form in the atmosphere and contribute to ambient $PM_{2.5}$ mass concentrations is growing (Jang et al 2002). Secondary organic PM contains more polar and hygroscopic compounds than primary organic PM, and is a larger contributor in the summer than in the winter.

Given that $PM_{2.5}$ generated outdoors infiltrates indoors, PM constituents found outdoors will presumably also be found indoors. Several investigators have measured sulfate indoors, and a few have measured EC (Landis et al 2001; Geller et al 2002; Gotschi et al 2002). Although sulfate concentrations are typically lower indoors, they are highly correlated with outdoor concentrations (eg, Leaderer 1999; Landis et al 2001; Sarnat 2002). Limited but compelling evidence suggests that ammonium nitrate is rapidly lost in indoor environments (Lunden et al 2003).

Trace elements, although they make up a small fraction of PM mass, are the most commonly measured constituents of indoor PM (Koutrakis et al 1992; Özkaynak et al 1996; Conner et al 2001; Chao and Wong 2002; Graney et al 2004) because they are useful for tracing sources.

The largest uncertainties in the chemical characterization of $PM_{2.5}$ are the quantitation and speciation of the organic fraction. Sampling artifacts interfere with accurate measurements of total organic PM (Turpin et al 2000). Landis and coworkers (2001) illustrated the difficulty this posed in characterizing $PM_{2.5}$ inside a Baltimore retirement facility: Because sampling artifacts were not taken into account, measured OC was equal to 168% of measured fine particle mass. This problem, also encountered in personal exposure measurements, has been recognized for many years by investigators working on ambient PM.

PAHs account for only a small fraction of organic $PM_{2.5}$, yet they are the most studied particulate organic compounds in indoor environments (Dubowsky et al 1999; Fischer et al 2000; Liu et al 2001); many have been shown to be suspected or known carcinogens. PAHs arise from a variety of combustion processes, including operating motor vehicles (Harrison et al 1996; Dickhut et al 2000; Kavouras et al 2001), generating power via combustion of coal and oil (Harrison et al 1996; Rogge et al 1997), incineration (Harrison et al 1996; Kavouras et al 2001), and burning wood (Benner et al 1995; Rogge et al 1998). PAHs are transported from outdoor to indoor environments (Dubowsky, et al 1999; Fischer et al 2000). They are also generated indoors by cooking (Rogge et al 1991; Dubowsky et al 1999), smoking (Mitra and Ray 1995), and burning natural gas (Rogge et al 1993; Mitra and Ray 1995), wood (Oanh et al 1999; McDonald et al 2000), and candles and incense (Lau et al 1997; Li and Ro 2000). Measurements of other organic PM constituents in indoor samples are extremely limited (Weschler and Fong 1986; Kavouras and Stephanou 2002).

Epidemiologic and exposure studies have generated great interest in characterizing indoor particles because people spend most of their time indoors. US residents spend approximately 87% of a day indoors, 7% in vehicles, and only 6% outdoors (Robinson and Nelson 1995). Typically, indoor PM consists of outdoor particles that infiltrate indoors and remain suspended (EPA 2004), primary particles emitted indoors (Abt et al 2000a), and sometimes secondary particles formed indoors through reactions of gas-phase precursors emitted both indoors and outdoors (Wallace 1996; Weschler and Shields 1997; Wainman et al 2000).

When indoor PM sources are present, indoor concentrations can be substantially higher than outdoor PM concentrations (Kamens et al 1991; Leaderer et al 1994; Özkaynak et al 1996; Wallace 1996;; Pellizzari et al 1999; Lachenmyer and Hidy 2000; Patterson and Eatough 2000; Conner et al 2001; Winkle and Scheff 2001; Adgate et al 2002, 2003; Kousa et al 2002; EPA 2004). Activities that can increase PM from indoor sources include smoking, cooking (especially with gas stoves), and cleaning, washing, and walking because they release dust and chemicals from furniture and floors (Yocom 1982; Özkaynak et al 1996; Chao et al 1997; Jones et al 2000).

Outdoor $PM_{2.5}$ is also a main contributor to indoor particle concentrations in both naturally and mechanically ventilated structures (Thatcher and Layton 1995; Abt et al 2000a). Outdoor particles can enter indoor environments by convective flow (eg, through an open window) or by diffusional flow (ie, infiltration) through cracks and fissures in the barrier of the building envelope. A growing number of studies have estimated contributions of outdoor sources to indoor $PM_{2.5}$ concentrations (Leaderer et al 1999; Oglesby et al 2000; Ott et al 2000; Sarnat et al 2002). Some evidence indicates that outdoor PM has a greater effect on indoor particle levels in homes near local sources (Daisey et al 1989; Fischer et al 2000), despite other reports that $PM_{2.5}$ concentrations were fairly uniformly distributed across different cities (Wilson and Suh 1997; Pellizzari et al 1999; Oglesby et al 2000).

Numerous studies have found that personal exposure concentrations are usually higher than either indoor or outdoor concentrations (Özkaynak et al 1996; Pellizzari et al 1999, 2001; Evans et al 2000; Lachenmyer and Hidy 2000; Oglesby et al 2000; Rojas-Bracho et al 2000; Sarnat et al 2000; Williams et al 2000a,b; Rodes 2001; Adgate et al 2002, 2003). This has led to the term *personal cloud* to describe the elevated PM concentrations found near individuals. A personal cloud occurs, at least in part, because indoor PM sources are usually associated with personal activities, which results in elevated concentrations near people. Time spent in other microenvironments, such as in transit vehicles, can also contribute to personal exposures that are elevated compared with indoor and outdoor concentrations. Some studies involving subjects who spend considerable amounts of time indoors in locations with few indoor PM sources have found personal concentrations that are lower than outdoor concentrations (Evans et al 2000; Sarnat et al 2000). Measurements of the personal cloud for older persons with chronic obstructive pulmonary disease (approximately 6 to 11 $\mu g/m^3$ for PM_{10} and 6 $\mu g/m^3$ for $PM_{2.5}$) are much lower than those for healthy individuals (approximately 27 to 56 $\mu g/m^3$ for PM_{10} and

11 to 27 μg/m^3 for $PM_{2.5}$; Wallace 2000); measurements of the personal cloud for $PM_{2.5}$ are lower than those for PM_{10} (Wallace 2000; Rodes 2001).

Speciation data for personal PM samples are limited, but a number of studies have measured sulfate and trace elements (Dockery and Spengler 1981; Özkaynak et al 1996; Pellizzari et al 1999; Sarnat et al 2000). Such analyses have shown that elevated personal exposures to PM_{10} can be explained, at least in part, by elevated concentrations of soil dust in personal samples (15% from indoor soil and 30% from resuspended indoor soil; Yakovleva and Hopke 1999).

SPECIFIC AIMS

The overall goal of $PM_{2.5}$ analysis in the RIOPA study was to improve the understanding of sources and mechanisms responsible for $PM_{2.5}$ exposure; this information would then facilitate developing effective strategies for public health protection. The specific aims for analyzing $PM_{2.5}$ data from study homes were:

1. to characterize and compare indoor, outdoor, and personal $PM_{2.5}$ mass composition;
2. to quantify the contribution of $PM_{2.5}$ of outdoor origin to indoor $PM_{2.5}$ concentrations and to personal $PM_{2.5}$ exposure; then to consider implications for predicting exposure and applying epidemiologic assessment methods; and
3. to further characterize the sources of indoor $PM_{2.5}$ concentrations and personal exposure (exploratory).

STUDY DESIGN

The design for the full RIOPA study is described in detail in Part I of this Research Report (Weisel et al 2005) and by Weisel and colleagues (2004). The study was undertaken both to investigate the relations between indoor, outdoor, and personal air concentrations for a variety of contaminants, and to evaluate the contribution of outdoor sources to personal contaminant exposure. Sampling was conducted during summer 1999 through spring 2001, indoors and outdoors at approximately 100 homes in each of three geographically distinct locations with different climates and housing characteristics; these conditions provided a wide distribution of AERs and compound infiltration mechanisms. This study design enabled us to examine the mechanisms that influence the relations among indoor, outdoor, and personal air contaminants. The study was not designed to obtain a population-based sample (the number of homes sampled, the participant selection criteria, and the recruiting procedures do not meet the criteria for population-based sampling), but rather to provide matched personal, indoor, and outdoor concentrations to facilitate mechanistic analyses. Homes particularly close to outdoor sources were oversampled to examine potentially high-level contributions of outdoor sources to personal exposure.

The concentrations of 18 volatile organic compounds, 17 carbonyl compounds, and $PM_{2.5}$ mass and more than 23 $PM_{2.5}$ species were measured with 48-hour resolution in indoor, outdoor, and personal air samples collected simultaneously. OC, EC, and 30 gas-phase and particle-phase PAHs were measured in indoor and outdoor samples collected concurrently. Questionnaires were administered to participating residents to characterize homes, neighborhoods, and personal activities that might affect exposures; subjects did not smoke. AER, temperature, and relative humidity were also measured in each home. In each city the study aimed to collect samples twice from approximately 100 homes that varied in proximity to sources, and PM measurements in about half of those homes.

Part I of this Research Report describes the study design and data collection methods in detail (Weisel et al 2005). It focuses on participants' characteristics and activities and presents measurements of AERs, volatile organic compounds, carbonyls, and $PM_{2.5}$ mass concentrations. The current report (Part II) highlights analysis and interpretation of $PM_{2.5}$ mass and species concentrations. To provide a context for the $PM_{2.5}$ results reported herein, sampling locations, recruiting procedures, housing characteristics, and demographic information about participants are briefly described below. In addition, measurement of AERs is briefly summarized.

STUDY SITES

The study sites were Elizabeth NJ, Houston TX, and Los Angeles County CA. Briefly, Elizabeth is a municipality of 110,000 that is contiguous with other cities in the region. It has a high population density with single and multifamily detached and semidetached homes as well as apartment buildings. The types of homes are typical of many areas in the northeast with many homes approaching 100 years in age and some newer homes. Elizabeth has a diverse racial and ethnic makeup with mainly lower-income and middle-income families. English and Spanish are the predominant languages.

Ambient air toxic sources within Elizabeth and in adjacent communities include industrial sources, an incinerator, numerous commercial sources (eg, gasoline stations, dry cleaners, refinishing shops, and small factories), and mobile sources from a number of congested local streets and major highways intersecting the area. A major metropolitan airport, Newark Liberty International Airport, borders Elizabeth on its north side. A major seaport is in the

eastern section of Elizabeth. Homes selected for the study included some on the same block as or within one or two blocks of local PM sources, with the exception of the airport. Homes farther from sources were selected from the western section of the city, which has fewer commercial and industrial facilities and lower traffic density. Homes were selected throughout the year in all sections of the city so no intentional seasonal imbalance in proximity to source type would be present in the data.

The Houston metropolitan area has the largest density of petrochemical complexes in the world. Some units within these facilities process crude petroleum for fuel production, and others produce chemicals including plastics and solvents. Most facilities are surrounded by highways and major access roads. Areas with large petrochemical complexes were identified, and homes near sources as well as homes farther away from sources were sampled within each area and, as much as possible, homes within any given area were monitored during the same time frame. Areas sampled were (1) the Houston Ship Channel; (2) Pasadena, located along interstate highway I-225; (3) Galena Park, north of I-225 and south of I-10; (4) Channelview, west of Galena Park and south of I-10; (5) Baytown; and (6) the Medical Center. With the exception of the Medical Center, these areas all include major chemical facilities. All have many single-family homes and low- and middle-income residents. Some areas also include upper-class residential neighborhoods. All areas include residents whose first language is English or Spanish.

Sampling was conducted at four locations in Los Angeles County: West Los Angeles, Pico Rivera, Burbank, and Newhall, each of which is intersected by at least one major freeway. Homes were selected at different distances from the freeways. All sampling locations were within 4 km of an ambient air monitoring station operated by the South Coast Air Quality Management District. Most of the Los Angeles sampling was conducted in West Los Angeles because that area has the highest daily vehicle count and is relatively free from the influence of point sources; it is near the intersection of I-405 and I-10. Pico Rivera is in central Los Angeles County on the 605 freeway, a road used by heavy-duty diesel trucks distributing goods from the port of Los Angeles. Burbank is north of downtown Los Angeles on the 101 freeway. Traffic volumes are lighter than those in West Los Angeles and Pico Rivera. Newhall is farther north of Burbank on the 101 freeway and traffic volumes are lower still; nevertheless, I-101 is a major north–south artery for the State of California. Primary languages spoken in these areas are English and Spanish.

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 areas were identified, subjects were recruited through requests to community and religious leaders, mailings, telephone calls, door-to-door canvassing, and by word of mouth. To qualify for the study, possible subjects had to live in a home with no smokers, spend at least 10 hours at home on a typical day, and not plan to move in the next 3 months. Recruiting and field contact with participants (including administration of questionnaires) were done in Spanish and English, according to the wishes of the participant.

HOUSING AND DEMOGRAPHIC CHARACTERISTICS

Samples were collected at 306 homes; some attributes of the homes are shown in Table 1. These were derived from the subjects' responses to several questions in the Baseline Questionnaire. (Questionnaires are available on request as Appendix H to Part I of this Research Report.) Houston had the highest proportion of participants living in mobile homes; no mobile homes were sampled in Elizabeth. Los Angeles participants lived in more recently built homes (1995 to 2000) than did participants in the other two cities. The Baseline Questionnaire included the following question: "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?" Responses indicated that about 20% to 30% of the homes had been renovated in the past year.

A total of 309 adults and 118 children (ages 8 to 18) living in the 306 homes participated in personal air sampling. Table 2 presents data on the age, gender, ethnicity, education level, and work status of participants; data were obtained from the Baseline Questionnaire. We purposely recruited subjects who typically spent at least 10 hours per day at home so that in-home measurements would be relevant to the evaluation of indoor, outdoor, and personal air relations. Because a higher proportion of women we contacted spent this amount of time at home, women subjects outnumbered men in the study. As shown in Table 2, the distributions of ethnic backgrounds, education levels, and other demographic descriptors differed among the cities. More subjects with higher levels of education participated in Los Angeles than in Elizabeth or Houston. Elizabeth had

Table 1. 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 either 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?"

many Hispanic participants, but no Mexican Americans. African American participants were few in all three cities. Roughly half of Los Angeles and Houston participants were white, whereas a minority of Elizabeth participants were white.

MEASUREMENT OF AERs

AERs were measured using a technique developed for determining total exchange of indoor air with outdoor air in relatively small enclosures such as homes, apartments, or small offices (Dietz et al 1986). As the number of air changes per hour increases, the steady-state concentration of an indoor tracer gas decreases. In this study we increased the source strength of the tracer gas in order to detect air exchanges up to 5/hour (AERs are shown as 5.0 hr^{-1}). AER was determined by emitting perfluorinated methylcyclohexane (PMCH) as the tracer gas at a known emission rate and measuring its steady-state concentration with a passive capillary absorption tube (CAT). CAT samples were analyzed by gas chromatography with an electron capture detector. The timing and location of CAT placement and quality control measures are described in detail in Part I of this Research Report (Weisel et al 2005). Indoor and outdoor temperatures were recorded every 10 minutes during sampling. The volume of occupied space in each home was measured using a tapeless ultrasonic tool or a walking tape. An unfinished basement or attic space that was not routinely used during the sampling was not included in the total home volume. The PMCH sources and CATs were supplied under a contract with Harvard University (Robert Weker's laboratory). The Harvard

Table 2. 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.

laboratory also checked emission rates for the sources and analyzed CATs. The AER was determined as follows:

$$\text{AER} = (n \times R_{\text{Perm}} \times \text{R}_{\text{CAT}} \times T_{\text{CAT}}) / (V_{\text{PMCH}} \times V_{\text{Home}}), \quad (1)$$

where n is the number of PMCH sources used, R_{Perm} is the source permeation rate (ng/min), R_{CAT} is the CAT collection rate (0.008308 L/hr), T_{CAT} is the CAT exposure time (minutes), V_{PMCH} is the volume of PMCH (picoliters) found on the CAT (calculated using standard gas chromatography calibration curves), and V_{Home} is the home volume (cubic feet). Figure 1 shows box plots of AERs for each city by season. The season was defined according to the 2001 calendar. For Houston homes the median AER was higher during the fall–winter months than during the spring–summer months. In contrast, Los Angeles homes had the lowest median AER during the winter and the highest median AER during the spring. The Elizabeth homes showed higher median AERs in the summer and winter than in the spring and the fall.

$PM_{2.5}$ SAMPLING, MEASUREMENT, VALIDATION, AND QUALITY CONTROL

The information presented here is described in detail in Part I of this Research Report (Weisel et al 2005) and by Weisel and colleagues (2004).

Concurrent indoor, outdoor, and personal measurements were made in 306 homes between summer of 1999 and spring of 2001. The study plan called for PM sampling and species analysis in about 150 homes; ultimately, however, $PM_{2.5}$ samples were collected in 219 homes. Of the 309 adults and 118 children who participated in personal air sampling for volatile organic compounds, 208 adults (145 twice) and 23 children (14 twice) also participated in $PM_{2.5}$ sampling. A 48-hour collection time was used for all chemical measurements to improve quantitation of trace-level species.

Indoor, outdoor, and personal samples suitable for analysis of $PM_{2.5}$ mass, functional groups, and elements were collected by a sampler using a Teflon filter in 219 homes; 169 of these homes were sampled a second time at least 3 months later (Figure 2). Indoor and outdoor samples suitable for $PM_{2.5}$ organic and elemental carbon (OC and EC) and trace level organic analyses were collected at the same time by a different sampler using a QFF in 152 (132 sampled twice) of the 219 homes. Samples from all 219 homes and participants were analyzed for mass and functional groups (elemental structures attached to carbon that can influence a molecule's behavior). Of the 219 homes, a subset of samples

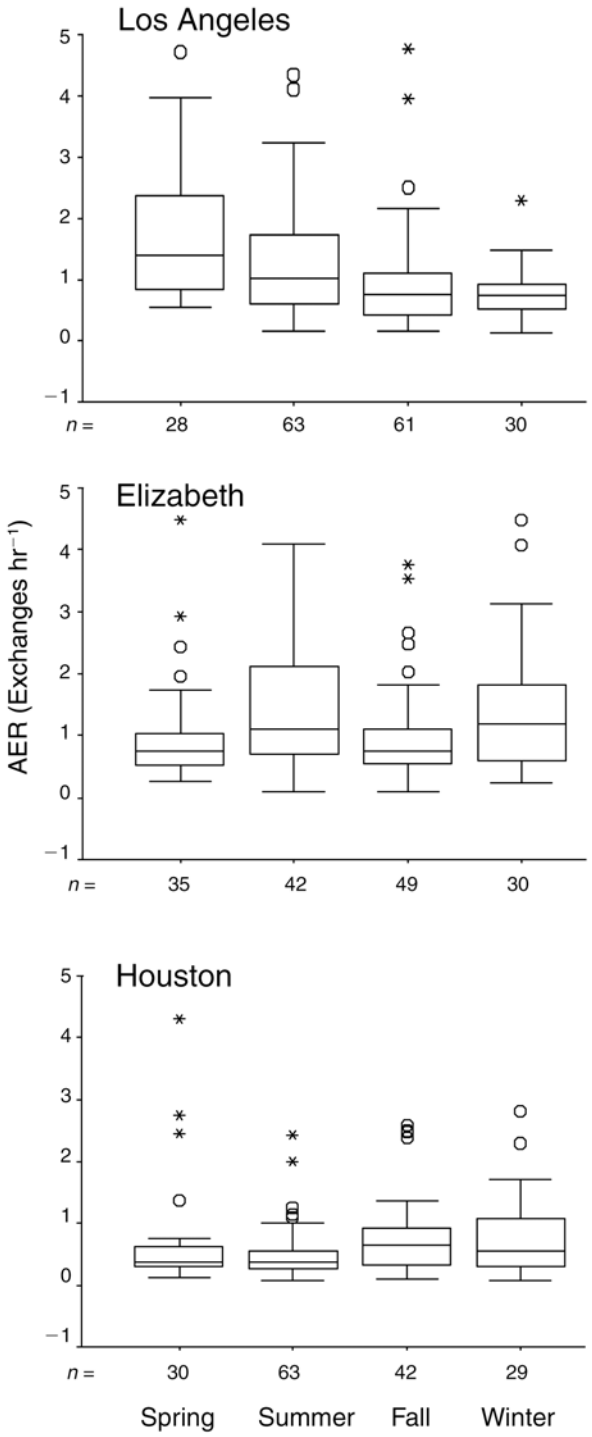


Figure 1. Seasonal variations of AERs in Los Angeles, Elizabeth, and Houston. *n* values beneath the *x* axes are the numbers of samples analyzed for each season. The box plots summarize the median, lower quartile, upper quartile, lower range, and upper range. White circles (○) represent outliers between 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 2. Sample analysis flow chart. Numbers of homes where samples were collected are in parentheses (first sampling/second sampling). Teflon filter samples were collected in 219 homes, and a second sample was collected in 169 of these homes (219/169). All of these samples were analyzed for mass and for functional groups by FTIR spectroscopy. Samples from a subset of these homes (162/120) were analyzed for elements by XRF and by ICP–MS. Quartz fiber filter samples also were collected in a subset (152/132) of the 219 homes and analyzed for OC and EC (99/74) and for PAHs and chlordanes (99/58). Samples analyzed for OC and EC were also analyzed for PAHs and chlordanes, mass, elements (by ICP–MS and XRF), and functional groups (by FTIR spectroscopy).

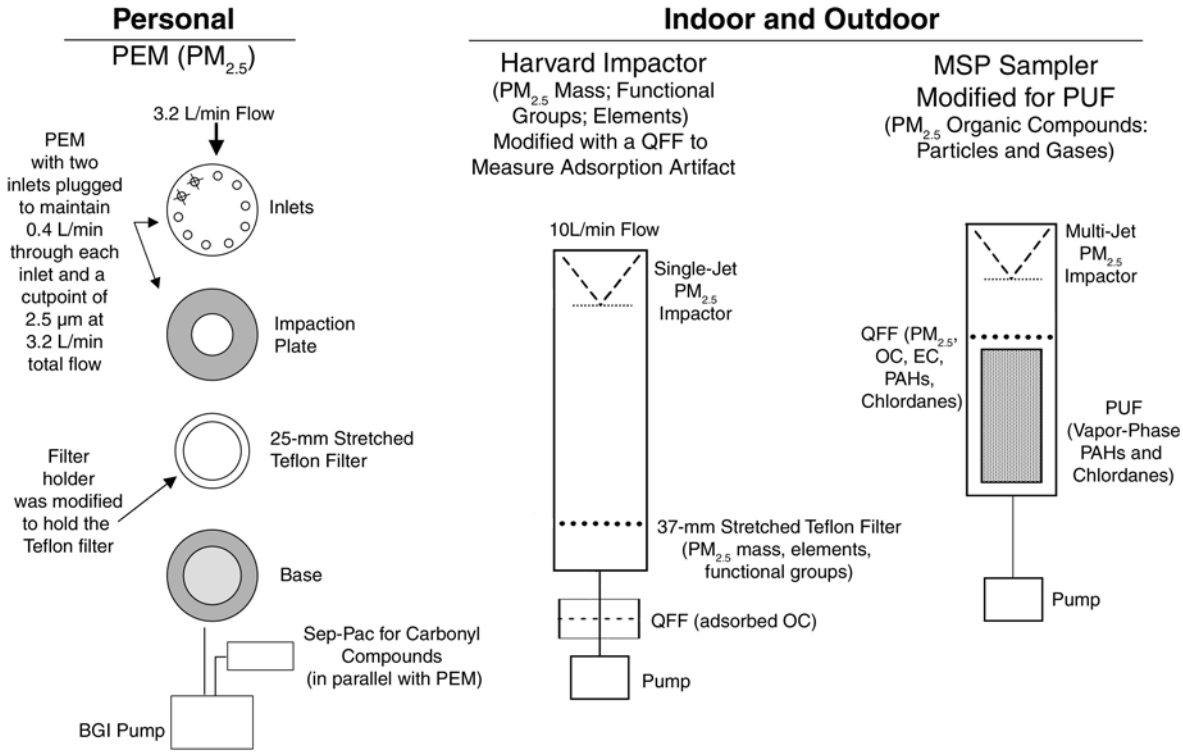


Figure 3. Schematics of the PEM, Harvard Impactor, and MSP sampler. The PEM was worn by participants to collect personal $PM_{2.5}$ samples. The Harvard impactor collected indoor and outdoor samples for $PM_{2.5}$ mass, functional groups, and chemical elements and included a backup filter to estimate adsorbed OC on the MSP QFF. The modified MSP sampler collected indoor and outdoor samples for particulate OC and EC and trace-level organic compounds and gases.

from 162 homes (120 sampled twice) were analyzed for elements. Of these, 99 homes (74 sampled twice) were also analyzed for OC and EC and these 99 homes (58 sampled twice) were analyzed for PAHs and chlordanes. Samples for detailed chemical analysis were selected to obtain a balance of homes across states and near to and farther from identified sources.

$PM_{2.5}$ SAMPLING

Personal and microenvironmental (indoor and outdoor) $PM_{2.5}$ samplers are illustrated in Figure 3. Each personal sample was collected using an MSP (MSP Co, Minneapolis MN) personal environmental monitor (PEM). The PEM has a 10-jet impactor inlet designed to provide a particle cutpoint of 2.5 µm in aerodynamic diameter when 0.4 L/min flow is maintained through each jet. For this study two jets were blocked to achieve the same cutpoint at 3.2 L/min

total flow. The PEM was also modified to hold a stretched 25-mm Teflon filter (3-μm pore), rather than a 37-mm filter, to obtain better species detection limits. Flow was drawn through the PEM, and in some cases through an active carbonyl sampler connected in parallel, using an AFC 400S pump (BGI, Waltham MA). PEMs were placed on the front strap of a harness near the breathing zone of the participant. The sampling bag, worn on the participant's hip or back, contained the pump, battery pack, and a motion sensor (HOBO, Onset Computer Corp, Bourne MA). Participants were instructed to wear the PEM near their breathing zone, or to keep it nearby when remaining stationary for long periods, such as during sleep.

Indoor samplers were placed in the main living area of the home (excluding the kitchen) and outdoor samplers were placed in secure locations in the front or back yard. Both indoor and outdoor samplers were mounted 1 m to 2 m from the floor and at least 1 m from walls or other structures.

Indoor and outdoor samples to be analyzed for $PM_{2.5}$ mass, functional groups, and chemical elements were collected with a Harvard impactor: a 37-mm stretched Teflon filter (2-μm pore; Pallflex Gelman Scientific, Ann Arbor MI) located downstream of a single-jet impactor with a 2.5-μm aerodynamic diameter cutpoint at 10 L/min. Collection time was 48 hours.

Concurrently, indoor and outdoor samples to be analyzed for particulate OC and EC and for trace-level organic compounds were collected using a modified MSP microenvironmental $PM_{2.5}$ sampler. This sampler was modified to hold a polyurethane foam (PUF) adsorbent (diameter, 25 mm; height, 100 mm) for collecting vapor-phase semivolatile organic compounds. The PUF was placed downstream of a multiple-jet impactor inlet with a 2.5-μm aerodynamic diameter cutpoint (at 10 L/min flow; 25 cm/sec face velocity) and a 37-mm quartz fiber filter (QFF). Samples were collected on QFFs for analysis of particulate OC, EC, PAHs, and chlordanes, and on PUFs for gas-phase PAHs and chlordanes.

Before sampling QFFs were prebaked at 550°C for 2 hours and stored at room temperature in Petri dishes lined with aluminum foil. PUF cartridges were hand-washed in tap water containing Alconox detergent, rinsed in deionized water followed by acetone, then sequentially extracted in a Soxhlet apparatus with acetone (24 hours) and petroleum ether (24 hours), and dried in a vacuum desiccator for 48 hours at ambient temperature. Cleaned PUF cartridges were stored at room temperature in prebaked (400°C) glass jars covered with lids lined with aluminum foil.

QFFs are typically used for measuring particulate OC and EC because they can withstand the high temperatures of thermal-optical analysis. However, in addition to collecting particulate carbon with approximately 100% efficiency, the filter surfaces also adsorb some organic vapors. Left uncorrected, this adsorption artifact typically results in a 30% to 50% overestimation of outdoor particulate OC concentrations (Turpin et al 2000). Little is known about the magnitude of the adsorption artifact indoors. However, indoor concentrations of organic gases are frequently higher than outdoor concentrations, which suggests that the artifact could be even more substantial indoors than outdoors.

The size of the adsorption artifact depends on the filter surface area, sampling face velocity, and the concentration and properties of the semivolatile organic vapors. In addition, changes in temperature and in organic vapor concentrations during sampling disturb the equilibrium between the gas-phase compound that passes through the filter and the organic material sorbed to the filter and particles. This provides a driving force for further adsorption or volatilization of collected semivolatile organic matter, which leads to a sample that is weighted toward the conditions at the end of the sampling period (Turpin et al 2000).

Because of the magnitude of the adsorption artifact, efforts to measure outdoor particulate OC concentrations frequently involve minimizing the adsorption artifact through the use of a denuder or by estimating the adsorption artifact by measuring the OC collected concurrently on a dynamic blank. Specifically, particle-free ambient air is sampled in the same location, at the same face velocity, on a QFF downstream of a Teflon filter (Turpin et al 2000). In this study, a 37-mm QFF was placed downstream of the Teflon filter in a Harvard impactor, which was collocated with the MSP sampler. This backup QFF on the Harvard impactor provided an estimate of the organic vapor adsorbed on the QFF in the MSP sampler.

In the laboratory, filters were loaded, unloaded, and checked for leaks. 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 measured flow rate had changed by more than ±5%. A field blank of each filter type was transported with samples to the field, kept near the indoor or outdoor sampler during sample collection, and stored and analyzed with field samples from concurrently measured homes. Duplicate samples were collected with pairs of collocated Harvard impactors at 35 homes and pairs of collocated MSP samplers at 31 homes. In addition, 14 samples were collected with PEMs mounted next to the indoor Harvard impactors to compare sampler performance. Collected samples and field blanks were returned to the laboratory in coolers with blue ice packs and stored frozen (−4°C) until analysis.

A variety of methods were used to document the sample collection and analysis process. A form to document chain

of custody was initiated with filter preparation and transported with the filters through analysis. Prepared filters were placed in Petri dishes labeled with a number and bar code. Identical labels were taped to the outside of the Petri dish. When a filter was loaded into the sampler, another label was applied to the outside of the sampler.

A field sheet form was used to guide the field technician through the process of measuring and recording critical data about the sampling, such as flow rates, start and stop times, and comments about factors that could affect sample validity. Upon return from the field, sample and blank filters were returned to their original labeled Petri dishes, and field data were entered into the electronic database. A second researcher later checked these entries against the original field sheets.

After validation of sample analyses, field data and analytical data were merged by sample identification number to provide sample volumes and information needed to determine sample validity and to calculate concentrations. One researcher was responsible for providing filters to the field team, receiving collected samples and blanks from the field, storing filters, and providing samples and blanks to analysts. This made it possible to conduct blind analyses.

SAMPLE VALIDATION

Sample validation required that flow rates changed less than 15% during sampling and that collection times exceeded 42 hours (87.5% of target duration). Field sheet comments were also taken into consideration during sample validation. For example, a sample was invalid if field comments suggested that the equipment malfunctioned or that the subject did not wear the personal monitor. Of the $PM_{2.5}$ samples collected on Teflon filters, 91%, 82%, and 83% were deemed valid in Los Angeles, Elizabeth, and Houston, respectively. Of the samples collected on QFFs, a total of 91%, 94%, and 94% were deemed valid in Los Angeles, Elizabeth, and Houston, respectively. Invalidation of analytical results was infrequent and did not lead to a significant decrease in the completeness of the data set because enough substrate or extract was available that invalid analyses were rerun.

Table 3. Number of Teflon Filter Samples Analyzed for $PM_{2.5}$ Component Category

Location	Mass	Functional Groups (FTIR)	Elements	
			XRF	ICP–MS
Los Angeles				
Indoor	131	131	106	106
Outdoor	130	130	103	103
Personal	126	126	96	96
Elizabeth				
Indoor	117	117	83	83
Outdoor	117	117	79	79
Personal	137	137	89	89
Houston				
Indoor	127	127	86	86
Outdoor	128	128	84	84
Personal	128	128	82	82

SAMPLE ANALYSIS

Figure 2 is a flow chart of the $PM_{2.5}$ sampling and chemical analysis strategies. Tables 3 and 4 provide, respectively, the number of Teflon filter and QFF samples analyzed by each method. Samples for species analysis were selected in such a way as to construct, to the extent possible, a database of homes that is complete with respect to concurrent indoor, outdoor, and personal species concentrations and is balanced across cities, seasons, and proximity of homes to identified sources.

$PM_{2.5}$ MASS

All Teflon filters were weighed on a microbalance (C-30, Cahn Instruments, Cerritos CA; or MT5, Mettler Toledo, Columbus OH) in an EPA-audited laboratory at the Environmental and Occupational Health Sciences Institute according to EPA protocols for $PM_{2.5}$ mass. Filters were equilibrated before and after sampling for 24 hours at 30% to 40% relative humidity and 20°C to 23°C. Conditions for postcollection analysis were within 5% relative humidity and 2°C of those for precollection analysis for each filter.

Table 4. Number of QFF Samples Analyzed for $PM_{2.5}$ Component Categories

Location	PAHs and Chlordanes	OC	EC
Los Angeles			
Indoor	61	44	44
Outdoor	61	44	44
Elizabeth			
Indoor	51	60	60
Outdoor	51	60	60
Houston			
Indoor	45	69	69
Outdoor	45	69	69

Temperature and relative humidity were recorded continuously in the weighing room. The balance was calibrated daily before weighing filters with a primary mass standard (200 ± 0.025 mg) traceable to US National Institute of Standards and Technology (NIST) mass standards. An independent standard (50 mg) was analyzed after every ten filters to evaluate analytical accuracy. At least one laboratory blank was also weighed daily. All filters were weighed twice.

The limits of detection for $PM_{2.5}$ mass concentrations, calculated as 3 × SD of the field blanks, were 0.55 μg/m^3 for indoor and outdoor samples and 1.4 μg/m^3 for personal samples. Field blank weights were not significantly different before and after transport to the field according to a paired *t* test with $\alpha = 0.05$ ($n = 452$, $P = 0.24$). Therefore, no blank subtraction was performed for $PM_{2.5}$ mass measurements. All $PM_{2.5}$ mass concentrations were above detection limits.

Uncertainties in mass concentrations were clearly dominated by sampling uncertainties, as evidenced by very high estimates of analytical precision (better than 1% as judged by replicate sample analysis) and analytical accuracy (replicate analyses of the 50-mg standard had a SD of 0.002 mg). Overall measurement precision for indoor and outdoor mass concentrations was 17%, as a coefficient of variation (CV), as judged by analysis of 35 pairs of collocated Harvard impactors inside and outside of homes (Figure 4). Overall measurement accuracy for atmospheric PM is usually on the order of 15% to 20% and is limited by sampling artifacts.

During 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 ammonium nitrate, semivolatile organic compounds, and water. Though such changes were not controlled during sampling, they were minimized during analysis by using the EPA weighing protocol (EPA 1997).

Figure 5 shows $PM_{2.5}$ mass concentrations measured with PEM and Harvard impactor samplers collocated in indoor sampling racks of 14 homes. Shapiro-Wilk tests suggest that both data sets are consistent with a log-normal distribution ($\alpha = 0.05$; $P = 0.40$ for Harvard impactor and $P = 0.05$ for PEM). Concentrations measured by the collocated Harvard impactor and PEM were highly correlated (coefficient of determination [r^2] = 98%; 92% without highest point), suggesting that the PEM had good precision.

The mass concentrations measured with the PEMs were significantly greater at the 95% confidence level than those measured with the Harvard impactors according to a *t* test on the log-transformed data. The median concentrations measured during the collocated sampling were 13.5 μg/m^3 for the PEM and 11.6 μg/m^3 for the Harvard impactor; the means of collocated measurements were 19.5 and 16.5 μg/m^3, respectively. The linear least-squares regression of PEM mass measurements on collocated Harvard impactor (HI) measurements is

$$[\text{PEM}] = 0.92[\text{HI}] + 4.33, \quad (2)$$

where the 95% confidence intervals (CIs) for slope and intercept are 0.81–1.02 and 2.03–6.63, respectively. Note that the CI for the slope includes 1.0. The regression equation is fairly sensitive to individual data points.

These data were not used to "calibrate" one sampler against the other because the scarcity of PEM and Harvard impactor data above 30 μg/m^3 would make the accuracy of this correction uncertain for high-level exposures. According to equation 2, the relative bias between samplers at the mean personal exposure of 37.6 μg/m^3 is 1.4 μg/m^3 (4%); or 0.32 μg/m^3 (0.9%) without outliers defined by the Dixon test ($\alpha = 0.05$). This uncertainty is

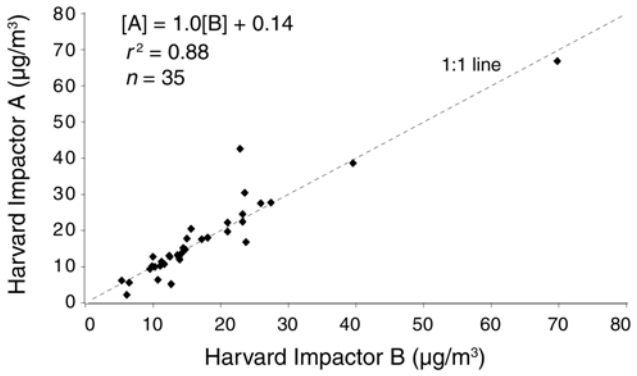


Figure 4. Indoor and outdoor $PM_{2.5}$ mass concentrations from 35 pairs of collocated Harvard impactors.

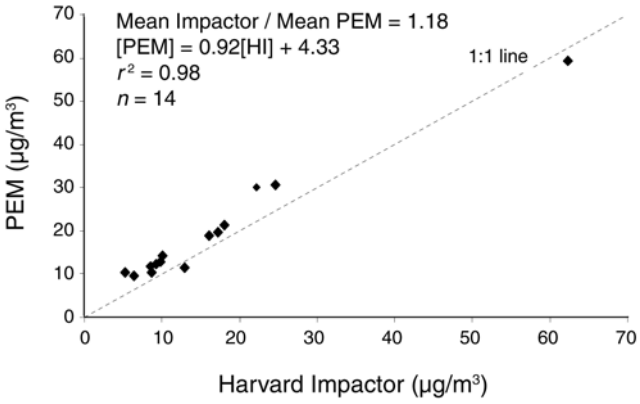


Figure 5. $PM_{2.5}$ mass concentrations from 14 pairs of collocated Harvard impactors and PEMs.

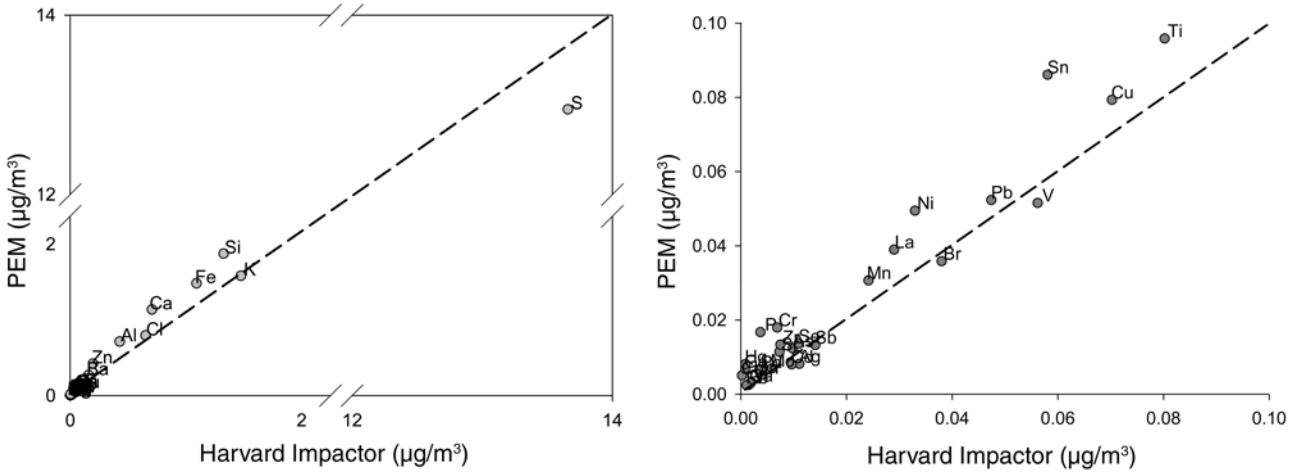


Figure 6. Mean concentrations (above MDLs) of $PM_{2.5}$ elements from collocated samplers. Elements were determined by XRF analysis. All elements (left); expanded scale (right).

reasonable considering PM mass measurement precision. Intersampler differences of this size are not unusual for collocated measurements of $PM_{2.5}$, which can result from differences in the shapes of the collection efficiency curves for the 2.5-µm impactor cutpoint, differences in bounce from the impaction plates, and differences in volatile losses. The Harvard impactor has a single-jet impactor inlet and a face velocity of 16 cm/sec, whereas the PEM was operated with an 8-jet impactor inlet and a face velocity of 11 cm/sec. Samples obtained at low face velocity are less susceptible to volatilization (Turpin et al 2000).

Species concentration data provide further insights into the intersampler differences. Figure 6 shows mean elemental concentrations obtained by x-ray fluorescence (XRF) analysis from the collocated PEM and Harvard impactor (see the section $PM_{2.5}$ Sampling, Measurement, Validation, and Quality Control / $PM_{2.5}$ Elements / XRF). The Shapiro-Wilk test was used to assess the distribution of elements and suggested that some elements were neither normally nor log-normally distributed. For these elements a Wilcoxon test was used to compare the results; for the remainder a paired *t* test was used.

Measured PEM concentrations were significantly higher than measured Harvard impactor concentrations for most soil elements: aluminum, silicon, calcium, titanium, iron, and zinc (Figure 6 top). In contrast, the PEMs yielded slightly lower concentrations of sulfur (Figure 6 top) and vanadium (Figure 6 bottom), which are accumulation-mode elements. $PM_{2.5}$ samplers collect only the smallest of the coarse-mode soil dust aerosol, so the mass of collected soil dust is particularly sensitive to the shape of the inlet collection efficiency curve. These results suggest that differences in the shape of the collection efficiency curves for the 2.5-µm impactor inlets could explain the intersampler differences in $PM_{2.5}$ mass concentrations.

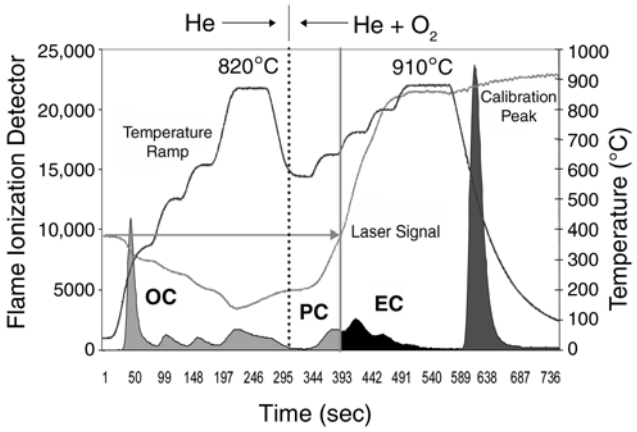


Figure 7. Example thermogram from analysis of OC and EC by thermal-optical transmittance in a Sunset Laboratory Carbon Analyzer (Birch and Cary 1996).

The discrepancy between Harvard impactor and PEM results introduces uncertainty in the magnitude of the difference between personal exposures and microenvironmental concentrations, rather than in the correlation between these measurements. The effect of this uncertainty on subsequent analyses is minimal and is noted below.

$PM_{2.5}$ OC AND EC

Both OC and EC were measured by thermal-optical transmittance in a Sunset Laboratory Carbon Analyzer (Birch and Cary 1996). An example analysis thermogram is shown in Figure 7. Briefly, air was purged from the analyzer after a 1-cm^2 punch of sampled QFF was loaded in the laboratory analyzer. The QFF was then heated stepwise in a helium environment to 820°C to volatilize OC. After removing OC, EC was eluted by combustion in 2% oxygen in helium while heating stepwise to 910°C. All evolved carbon was converted to methane and measured with a

flame ionization detector (FID). A calibration gas with a known amount of methane was automatically injected in the last step of the analysis for quantitation.

During analysis, some OC was pyrolytically converted to EC, which reduced the transmittance through the filter. Correction for pyrolysis was made by monitoring the transmittance of light through the filter using a diode laser and a photodetector. The amount of carbon that has been pyrolytically converted to EC is considered to be the amount of EC that must be removed to return the transmittance to its initial analysis value (often called the OC–EC split point). This pyrolysis correction assumes that either the pyrolytically generated EC is removed first, or the original EC and the pyrolytically generated EC have the same absorptivity (Turpin et al 1990). OC is then equal to the carbon removed in helium plus the EC removed before the laser regains its prepyrolysis value. EC is the remaining carbon removed in helium–oxygen. Carbonate carbon was not separately determined because previous studies have found that ambient particulate carbonate carbon concentrations are minimal (Mueller et al 1972; Nunes and Pio 1993; Ohta et al 1998). Any carbonate carbon present is expected to be reported as OC (Birch and Cary 1996).

Ultra-high-purity helium, 10% oxygen in helium, and 5% methane in helium (certified grade), ultra-high-purity hydrogen, and zero-grade air were used for carbon analysis. Pressurized gases from Matheson Gas Products (Montgomeryville PA) were used without further purification except that the helium passed through a series of oxygen traps (4002; 4004; Alltech, Deerfield IL) to remove trace amounts of oxygen before use.

Regular quality control checks were made for system contamination, variations in FID response across an analyses, analytical precision, and analytical accuracy. Instrument blanks were measured daily during sample analysis; no system contamination was found. Detection limits, expressed as 3 × SD of the field blanks, were 0.3 $\mu g/m^3$ for OC and 0.07 $\mu g/m^3$ for EC. OC and EC were above detection limits in all samples.

Analytical precision, expressed as a CV of replicate analysis of 10% of all samples, was 5% for OC and 9% for EC. Analytical accuracy was 3.5% (for OC and EC), based on daily analysis of sucrose solutions spiked on a QFF. Variability in detector sensitivity (FID response) across an analysis was examined by analyzing an instrument blank and automatically injecting a known amount of methane during the helium and the helium–oxygen segments of the analysis. This quality control check was performed daily. The FID response did not vary by more than ±5% between analysis segments.

Internal calibration was performed by switching in-line a loop of tubing containing methane in helium (approximately 5%, certified). The exact loop volume varies from instrument to instrument. We verified the volume in our laboratory by manually injecting known volumes of certified calibration gas with a gas-tight syringe during an instrument blank analysis and comparing these areas to the internal calibration peak area. This quality control check was run approximately once every 3 months. Over the time the samples were analyzed, this was conducted using two independent calibration gas standards. The loop volume of this instrument was 1.3 ± 0.2 mL. This provided 34 ± 0.4 μg of carbon in the internal calibration peak given a calibration tank with exactly 5% methane in helium.

Another parameter important to the accurate separation of OC and EC is the transit time. The photodetector responds instantly when EC is formed on or is evolved from the filter. In contrast, the corresponding change in the FID signal is delayed by the transit time of gases from the filter to the FID, which was 11 seconds.

Analytical accuracy and precision are well-defined, measurable quantities; however, overall measurement accuracy and precision for OC and EC are more difficult to assess. Total carbon (OC + EC) is a well-defined quantity. However, OC and EC are somewhat operationally defined. EC is composed mostly of carbon atoms; OC contains considerably more hydrogen, oxygen, and perhaps other constituents. There is no well-defined separation between the two. Different analytical methods and protocols to separate OC and EC are based on different principles and yield somewhat different OC–EC splits (Turpin et al 2000; Lim et al 2003a). Ideally (assuming the OC evolution temperature is high enough and the assumptions underlying the pyrolysis correction are accurate), thermal-optical analysis methods define OC as carbon that can volatilize in the absence of oxygen, and EC as carbon that requires oxygen to evolve (ie, through combustion). However, somewhat different OC–EC splits can occur with thermal-optical methods using different temperature protocols or pyrolysis correction methods (Schauer et al 2003).

Accurate collection of organic PM is also hampered by the fact that many organic compounds are semivolatile and partition between the gas and particle phases. Sampling alters the organic gas–particle equilibrium, introducing positive and possibly negative organic artifacts (Heubert and Charlson 2000; Turpin et al 2000). The success of the sampling strategy at minimizing or quantifying these artifacts affects the accuracy of OC and total carbon measurements. For example, for outdoor low-volume $PM_{2.5}$ samples, typically 30% to 50% of the organic material collected on a QFF is adsorbed vapor (Turpin et al 2000).

These sampling and analytical issues make it particularly important that the data for analyses be obtained from a single collection and analysis protocol. Measurements made using different collocated samplers and analyzed with different methods provide an estimate of the precision with which carbonaceous PM can be measured. The intermethod precision is on the order of 5% for total carbon and is not much greater for OC. The intermethod precision for EC is considerably greater; for example, it was 34% during the Carbonaceous Methods Intercomparison Study and 20% to 200% during the Atlanta Supersite Experiment (Turpin et al 2000; Solomon et al 2003). For this study, the within-method measurement precision was calculated from MSP samplers collocated outdoors at homes (n = 30). These measurements yielded pooled CVs of 4% for OC and 7% for EC, suggesting that the measurement precision was comparable to the analytical precision.

Particulate OC and EC concentrations reported for this study are in micrograms of carbon per cubic meter of air. A QFF was placed behind the Teflon filter in the Harvard impactor (ie, the dynamic blank) for 89% of all samples. This provided a measure of the field blank plus the adsorption of organic vapors on the MSP QFF. Reported particulate OC was then equal to OC on the MSP QFF minus OC on the concurrent Harvard impactor backup QFF. For samples without a corresponding backup QFF, the magnitude of the adsorption artifact was estimated as described in the Organic Aerosol Sampling Artifacts section of the Results and Discussion.

$PM_{2.5}$ ELEMENTS

XRF

Selected indoor, outdoor, and personal samples (from Teflon filters) were analyzed with an energy-dispersive XRF spectrometer (Delta 770, Kevex, Thermo-Fisher Scientific, Waltham MA) equipped with a water-cooled end-window x-ray tube with a rhodium anode and a peak operating power of 60 kV and 3.3 mA (Chester LabNet, Portland OR). Standard operating procedure XR-002.01 was followed during XRF analysis. In this protocol each sample has a 48-minute contact time. Standard reference materials NIST 1832 and 1833, field blanks, and replicates (10%) were analyzed with samples. A total of 36 elements were analyzed by XRF (Ag, Al, As, Ba, Br, Ca, Cd, Cl, Co, Cr, Cu, Fe, Ga, Ge, Hg, In, K, La, Mn, Mo, Ni, P, Pb, Pd, Rb, S, Sb, Se, Si, Sn, Sr, Ti, V, Y, Zn, and Zr).

Analytical accuracy ranged from 94% to 104%, based on certified NIST 1832 and 1833 values. The average field blank was not subtracted from the samples because the field blank collections were generally low. Therefore, method detection limits (MDLs) were defined as 3 × SD of the field blanks plus the average field blank (79 field blanks for indoor and outdoor samples, and 57 field blanks for personal samples). Analytical precision was expressed as a CV based on duplicate analysis of 10% of the samples; it was better than 10% for most elements. Overall measurement precision for most elements was better than 20% for indoor and outdoor samples, based on 33 pairs of collocated Harvard impactor samples inside and outside study homes.

ICP–MS

Teflon filter samples (and blanks) that were analyzed by XRF were subsequently analyzed by ICP–MS. ICP–MS analyses were performed because, for a number of source tracers, ICP–MS detection limits tend to be substantially lower than those for XRF; ICP–MS also distinguishes isotopes.

The filters were digested in closed Teflon vessels (6 mL, Savillex Corp, Minnetonka MN), cleaned in a microwave with 0.5 mL Optima HNO_3 (Thermo-Fisher Scientific, Waltham MA), followed by 0.5 mL deionized water, in closed 50-mL centrifuge tubes (VWR, Westchester PA). Specifically, a Teflon filter sample, 1 mL Optima HNO_3 (Thermo-Fisher Scientific), and 0.5 mL Utrex II H_2O_2 (JT Baker, Phillipsburg NJ) was added to each Teflon vessel. The vessel was sealed with a socket-type cap (Savillex Corp) designed for high-pressure applications.

Initially, digestion was performed without H_2O_2 and with a five-stage digestion procedure (10 min/stage; Appendix A), based on our previous experience. Initial samples were cooled, transferred to 50-mL centrifuge tubes, and diluted to 20 mL. However, the low sample loadings in this study led us to revise the analytical method to reduce detection limits. With the revised protocol, recoveries were slightly improved, and detection limits were dramatically reduced, as shown in Appendix A. A total of 173 indoor and outdoor and 88 personal samples were analyzed using the initial digestion protocol, whereas 374 indoor and outdoor and 180 personal samples were analyzed using the optimized final digestion protocol.

The final digestion protocol had eight stages (500 watts; 5 min/stage; 10% power increments from 40% to 70% for the first four stages and 60% for the last four stages). Samples were digested in a microwave oven (MDS-2000, CEM Corporation, Matthews NC) operated under time–power control mode. In each batch, 32 samples and field blanks, one urban PM standard (NIST 1648), a standard aqueous solution (NIST 1643), one solvent blank, and two laboratory blanks (one 37-mm blank Teflon filter and one 25-mm blank Teflon filter) were digested. After digestion, samples were cooled and transferred to precleaned 15-mL centrifuge tubes.

Samples and controls were analyzed with a Thermo Elemental Plasma Quad3 ICP–MS and ASX-500 autosampler

(CETAC Technologies, Omaha NE). Table 5 provides instrument operating parameters. For every six to eight samples, a 10-ppb solution made from NIST traceable SM-1811-001 and SM-1811-002 (high-purity element solutions containing 23 elements) was run as a quality control sample. If the quality control sample was not within ± 20% of the certified value for target elements, the instrument was recalibrated and the batch was reanalyzed.

Table 5. ICP–MS Operating Parameters

Torch	VG quartz
Nebulizer	Concentric
RF power	1350 W
Reflected power	Zero
Sample delivery rate	0.8 mL/sec
Sampler cone	1.0-mm orifice
Skimmer	0.7-mm orifice
Dwell time	320 msec/1000 msec
Acquire time	1 min/4 min
Cooling argon gas	14 L/min
Auxiliary argon gas	0.88 L/min
Nebulizer argon gas	0.73 L/min

In total, 22 elements were quantified by ICP–MS (Ag, As, Ba, Be, Bi, Cd, Co, Cr, Cs, Cu, Ga, Mn, Ni, Pb, Rb, Se, Sr, Ti, Tl, U, V, Zn). Most of these were also analyzed by XRF (except Be, Bi, Cs, Tl, and U). Accuracy was determined by comparisons with certified results from standard solution (NIST 1643) and urban PM standard (NIST 1648) to reflect digestion and matrix-extraction recoveries, respectively. Recoveries for most "extractable" elements were between 91% and 103% with ICP–MS. However, digestion recoveries for soil elements, such as chromium and titanium, are considerably lower with this method (in contrast, XRF spectroscopy is particularly well suited for soil elements). Improved digestion recovery for soil elements by ICP–MS might require the addition of an acid, such as hydrofluoric acid, to solubilize the silicates. Detection limits are expressed as 3 × SD of the field blank plus the mean field blank value. Blank values were not subtracted from the data.

Table 6 shows the percentage of data above detection limits for each element analyzed by ICP–MS. Analytical

Table 6. Percentage of Samples Above Detection Limits, by Element, When Analyzed by ICP–MS[a]

	Indoor		Outdoor		Personal	
Element	> 1 SD	> 3 × SD	> 1 SD	> 3 × SD	> 1 SD	> 3 × SD
Ag	54.6	17.4	45.0	13.7	59.4	16.7
As	96.8	88.0	99.5	94.2	85.0	32.2
Ba	99.5	99.5	99.5	99.5	99.4	97.2
Be	12.4	2.7	14.3	1.6	8.3	0.0
Bi	78.4	48.4	79.4	57.4	62.8	26.7
Cd	84.9	60.9	85.7	62.1	77.2	33.3
Co	68.6	27.7	78.8	42.6	40.0	7.8
Cr	55.7	8.2	55.0	10.0	62.2	20.6
Cs	11.4	1.6	13.8	3.2	14.4	0.0
Cu	86.5	42.9	84.1	50.0	92.2	53.3
Ga	93.5	77.2	99.5	93.7	81.1	46.7
Mn	99.5	99.5	99.5	99.5	100.0	99.4
Ni	9.7	5.4	11.6	3.2	56.7	25.6
Pb	99.5	94.6	99.5	97.9	100.0	98.3
Rb	88.1	44.0	98.4	64.7	61.7	17.2
Se	72.4	33.7	90.5	63.7	3.3	1.7
Sr	61.6	16.3	78.3	34.2	75.6	16.7
Ti	97.8	77.2	99.5	91.1	98.9	85.0
Tl	47.6	7.1	56.1	10.5	8.9	0.0
U	2.7	2.2	3.7	1.6	8.3	0
V	99.5	97.8	99.5	99.5	98.9	94.4
Zn	91.4	48.4	94.7	68.9	100.0	95.0

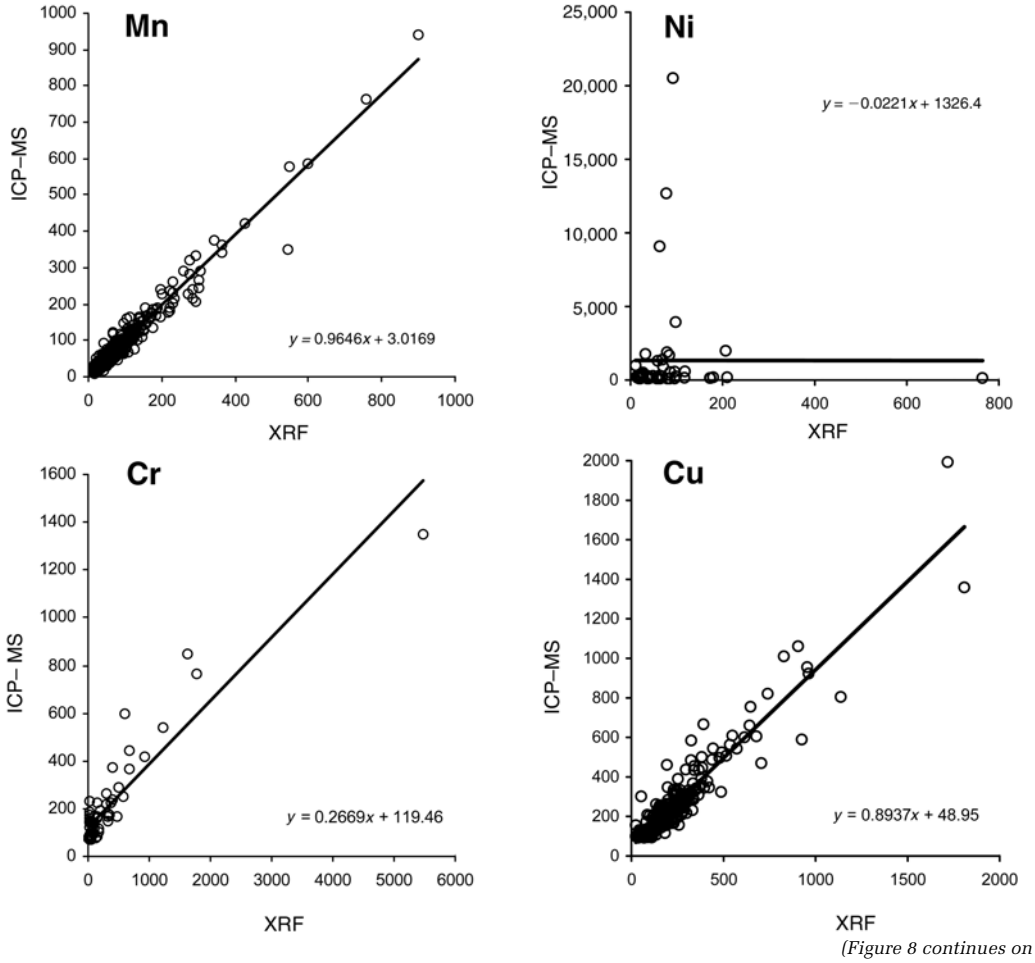
[a] Shown are percentages greater than the reported detection limit (3 × SD of the blank) and percentages higher than 1 SD of the blank for indoor, outdoor, and personal samples.

precision, expressed as pooled CVs of replicate sample analyses (10% replicates), was within 20%, with the exception of that for nickel, which is affected by the loss of nickel from the instrument core. Measurement precision was 4% (cesium) to 30% (copper) based on analysis of 34 collocated indoor and outdoor samples.

Results from the final ICP–MS protocol are compared below with XRF results. We found good agreement between XRF and ICP–MS for most elements of interest. The data analyses described below were conducted for elements identified using XRF. Isotope information provided by ICP–MS results might prove to be useful in future research into source apportionment. If future analyses are conducted with element data obtained using both the original and final optimized ICP–MS protocols, then care must be taken to properly address the difference in detection limits.

Concentrations measured by ICP–MS were compared with those measured by XRF for the 13 elements that had at least 10 pairs of data above detection limits (Ti, V, Cr, Mn, Ni, Cu, As, Se, Rb, Sr, Cd, Ba, and Pb). High correlation coefficients (r = 0.90 to 0.98) were obtained for 10 elements (V, Cr, Mn, Cu, As, Rb, Sr, Cd, Ba, and Pb). In contrast, XRF and ICP–MS results were more poorly correlated for nickel (r = 0.00), titanium (r = 0.78), and selenium (r = 0.47). Titanium and selenium are soil elements, and are difficult to extract without hydrofluoric acid. In addition, selenium is subject to interference. Nickel is a component of instrument core, and its loss during analysis introduces considerable analytical uncertainty.

The slopes of the Deming regressions (Deming 1943) of ICP–MS measurements on XRF measurements were close to 1, which suggests that the two methods agree well (Figure 8).



(Figure 8 continues on next two pages)

Figure 8. Concentrations of the 13 elements that were measured by both XRF and ICP–MS (ng/m^3). The equations and lines express the Deming regression of the ICP–MS measurement on the XRF measurement (for measurements above MDLs). Note that x and y axes differ within and between panels.

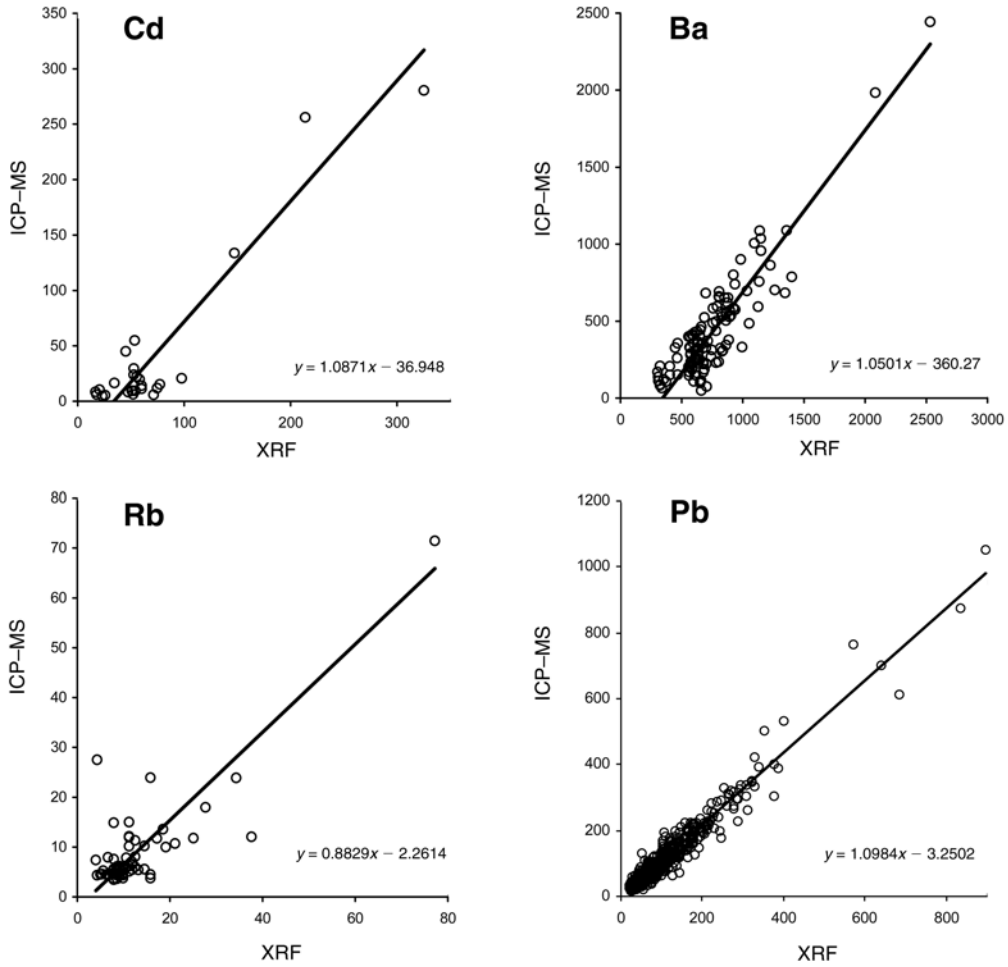


Figure 8. *(continued).*

Nine of 13 elements for which data were compared had slopes of 0.88 to 1.10. Many regression programs assume the uncertainties in the *x* variable are negligible. The Deming regression, however, allows uncertainties in *x* and *y* to be designated. The uncertainty was designated to be the measurement precision (%) of each element.

$PM_{2.5}$ FUNCTIONAL GROUPS

All particle samples from Teflon filters were analyzed by FTIR spectroscopy before precollection weighing and after postcollection weighing. Filters were analyzed directly without extraction or other sample preparation using a Mattson 100 Research Series Spectrometer (ATI Mattson, Madison WI) containing a deuterated triglycine sulfate detector. Filters were scanned 200 times at 4/cm resolution, producing an infrared absorbance spectrum from 450/cm to 4000/cm. To obtain the final sample spectrum, the precollection scan was subtracted from the postcollection scan using WinFIRST 3.61 software (ATI Mattson, Madison WI).

Filters were analyzed in the same orientation before and after sampling by aligning a mark scribed on the polypropylene ring with a mark on the filter holder. This improves the subtraction of the Teflon spectrum from the sample (Krost and McClenny 1994). Instrument background spectra were taken every half-hour. Every day the instrument bench was reset to maintain an energy throughput (peak-to-peak ratio) of at least 4.2 V; a standard-thickness polystyrene film provided by Mattson was scanned to monitor drift and changes in instrument sensitivity. The instrument automatically uses a helium–neon (He–Ne) laser as an internal standard to maintain wave number alignment.

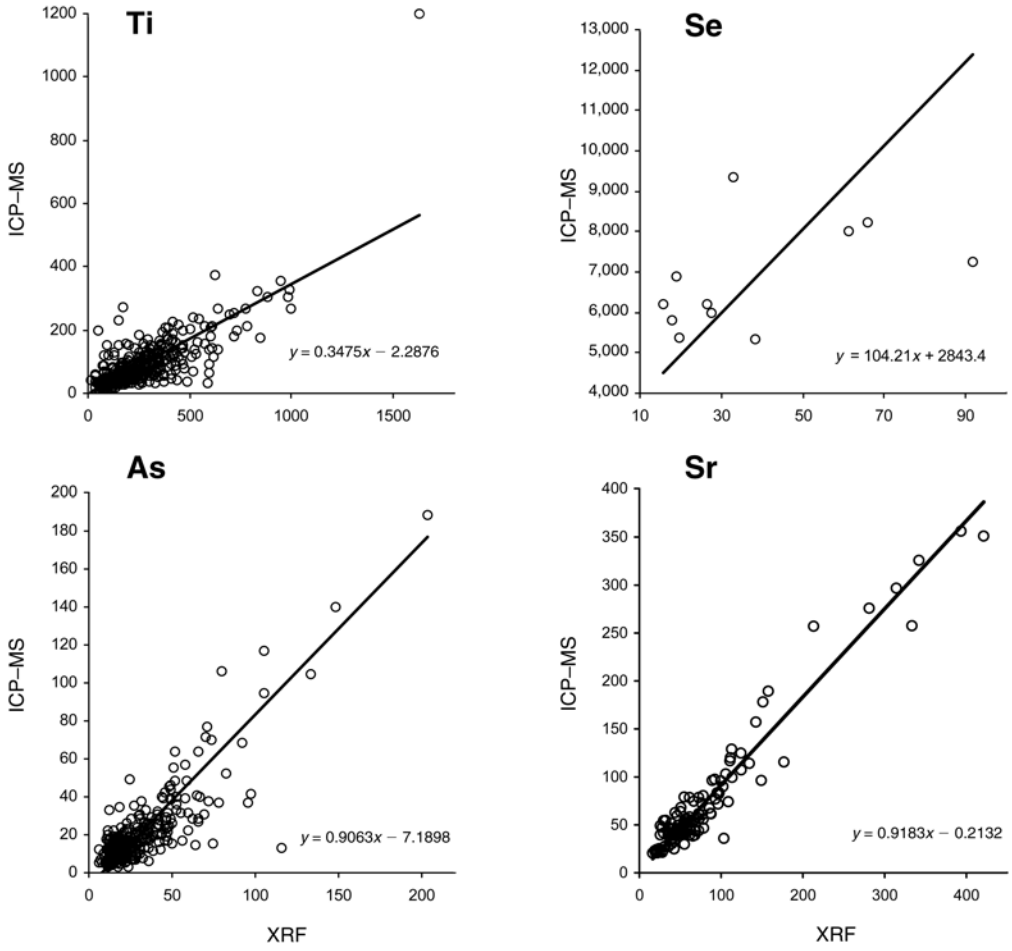


Figure 8. *(continued).*

Functional groups were identified from the aerosol literature (Allen et al 1994; Blando et al 1998; Carlton et al 1999), spectroscopy literature (Colthup et al 1990; Socrates 1994), and spectra of aerosol standards generated in our laboratory.

PAHs AND CHLORDANES

The indoor and outdoor concentrations of 30 PAHs and six chlordane species were measured in both gas and particle phases in 157 samples (99 homes of which 58 were sampled twice). PAHs are formed through combustion processes. Chlordanes are persistent organic pollutants that were used in the United States until 1988, initially as broad-spectrum agricultural insecticides and then later as termiticides in the subsurface of new construction. Both chlordanes and PAHs are semivolatile, meaning they partition between the gas and particle phases. Samples selected for trace-level organic analysis were extracted and analyzed by gas chromatography–mass spectrometry (GC–MS).

To determine analytical recoveries, before extraction PUF samples were spiked with 100 µL and QFF samples were spiked with 1 µL of a surrogate standard consisting of 1000 ng/mL of each of the following perdeuterated PAHs: acenaphthene-d_{10}, anthracene-d_{10}, fluoranthene-d_{10}, and benzo[*e*]pyrene-d_{12}. The amounts added—100 ng per compound for PUFs and 1 ng per compound for QFFs—were close to the mass of PAHs in the samples. The PUFs were extracted statically in glass columns (ID 30 mm × 120 mm, with 2-mm Teflon stopcock) for 1 hour with 40 mL of the mixture of hot (50°C) hexane and dichloromethane (DCM) (4:1 by volume).

The extracts were drained into collection flasks, and the PUFs were rinsed twice with 20 mL of the hot hexane and DCM mixture; rinses were combined with the extracts. Each QFF sample was split in two portions. Two 1-cm^2 punches of each filter were reserved for thermal-optical carbon analysis. The remaining substrate was spiked with the surrogate standard and extracted twice for 35 minutes with 25 mL of DCM under ultrasonic agitation.

The PUF and QFF extracts were concentrated by rotary evaporation (Büchi RE 111; BÜCHI Labortechnik AG, Flawil, Switzerland), followed by further concentration under a gentle nitrogen stream, and cleaned on microcolumns (I.D. 5 mm × 100 mm) of silicic acid to remove interfering polar compounds. Silica gel (60 to 200 mesh) was baked at 400°C for 8 hours, cooled in a desiccator for 1 hour, and deactivated with 5% deionized H_2O. The column was rinsed with 2 mL of hexane–DCM (9:1 by volume). The samples were added to the column and eluted with 8 mL of 9:1 hexane–DCM. Collected samples were reduced to approximately 0.05 mL by evaporation under a gentle stream of nitrogen. An internal standard solution (100 µL for PUF and 1 µL for QFFs) consisting of 1000 ng/mL of naphthalene-d_8, phenanthrene-d_{10}, pyrene-d_{10}, and benzo[*a*]pyrene-d_{12} was added to concentrated samples, and the QFF extracts were concentrated further to approximately 0.01 mL. The PAH internal standard that was nearest in chromatographic retention time was used for chlordane quantification.

The samples were analyzed on a Hewlett Packard 6890 gas chromatograph equipped with HP 5973 mass selective detector operated in selected ion monitoring mode. Compounds were separated on a high-resolution capillary column (J&W Scientific, Folsom CA; I.D. 0.25 mm; length 30 m) with DB-5 as the stationary phase (film thickness, 0.25 µm). Helium (chromatographic grade) was used as the carrier gas at a flow rate of 1.2 mL/min. The pressure in the column was maintained at 9.86 psi. The inlet was operated in the pulsed splitless mode at 300°C; the injection volume was 1 µL. The initial temperature (50°C) was held for 1.1 minutes, after which the temperature was raised using three sequential temperature ramps: first at 25°C/min to 125°C, then at 8°C/min to 260°C, and finally at 3.5°C/min to 300°C; the final temperature was held for 10 minutes. The analysis time for one sample was about 43 minutes.

The sum of masses for all PAHs (ΣPAH mass) measured in the PUF field blanks ranged from 0.039 to 190 ng, whereas in the PUF study samples it ranged from 52 to 31,500 ng. In the QFF field blanks, ΣPAH mass ranged from 0.16 to 9.2 ng; in the QFF study samples, it ranged from 0.36 to 800 ng. The MDL for an individual PAH was defined as 3 × SD of the mean PAH mass in the field blanks. Because the means of PAH masses in the field blanks collected in different cities were similar and no significant difference was found between the field blanks collected indoors and outdoors, media-specific MDLs were calculated using all field blanks and applied to samples collected in each of the three cities.

Laboratory blanks (clean PUFs and QFFs) and reference standards were extracted and analyzed with every 14 samples. The ΣPAH mass in the laboratory blanks ranged from below the MDL to 59 ng for the PUFs and from below the MDL to 5.6 ng for the QFFs and accounted for less than 5% of the corresponding ΣPAH mass in the PUF and QFF samples. The sum of masses for all chlordanes (Σchlordane mass) in laboratory blanks ranged from below the MDL to 0.26 ng for PUFs and from below the MDL to 0.77 ng for QFFs and on average accounted for less than 30% of the Σchlordane mass in the PUF and QFF samples. Most of this was driven by oxychlordane, which was below the MDL in more than 94% of particle samples and in approximately 28% of gas-phase samples. Samples were not corrected for laboratory blanks.

NIST standard reference material 1649a (Urban Dust) was analyzed to validate the QFF analyses for 16 PAHs and three chlordane species. A solution of 37 individual PAHs (Ultra, Sigma-Aldrich, Supelco, St Louis MO; and Cambridge Isotope Laboratories, Andover MA) in hexane and a solution of six chlordane species (Supelco) in hexane were used to validate the PUF analyses. Each material (the standard solution or NIST Standard Reference Material 1649a—Urban Dust—Organic Compounds) was added to the clean matrices (PUF or QFF) and then processed as a regular sample to determine the analytical accuracy of the method. Analytical precision was determined from duplicate GC–MS analysis of 10% of the samples. Measurement precision was determined from analysis of collocated samples.

In the GC–MS analysis, response factors for individual PAHs compared with those for the internal standard were determined from analyzing the calibration solution containing 43 PAHs, including deuterated PAHs. A separate solution containing six chlordane species was used in an identical fashion for concurrent analysis of chlordane species. Analytical recoveries of the surrogate PAHs in the PUF samples (n = 130) and QFF samples (n = 137) were 83% ± 17% PUF and 62% ± 12% QFF for acenaphthene-d_{10}, 91% ± 16% PUF and 69% ± 7.8% QFF for anthracene-d_{10}, 85% ± 10% PUF and 83% ± 7.3% QFF for fluoranthene-d_{10}, and 82% ± 12% PUF and 90% ± 8.9% QFF for benzo[*e*]-pyrene-d_{12}. All samples were corrected for surrogate recoveries by dividing the concentrations of individual PAHs and chlordane species by the recovery of a surrogate PAH with the closest molecular weight.

Significant breakthrough (23% to 56% expressed as percentage of the PAH mass on the backup PUF) was observed for the PAHs with lowest molecular weights: naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLR), and 1-methylfluorene (1-MFL). The concentrations of these PAHs were not reported. Breakthrough of chlordane species was not significant, as evidenced by minimal individual compound masses (less than 1%) for each of the six chlordane species measured on the backup PUF. Backup PUFs were collected at Houston homes outdoors in the summertime, when breakthrough was most likely to be a problem.

Because chrysene (CHR) and triphenylene (Tr) coelute in GC–MS analysis, results are reported as a sum of the two compounds (CHR/Tr). For the same reason, dibenzo[*a,c*]anthracene and dibenzo[*a,h*]anthracene are reported as a sum (DBA). Because of the occasional low resolution of the peaks corresponding to benzo[*b*]fluoranthene and benzo[*k*]fluoranthene, the two PAH isomers are also reported as a sum (BFLTs). Substantial interference of 2-methylphenanthrene with an unidentified compound was observed in approximately half of the PUF samples; thus 2-methylphenanthrene was excluded from data analyses.

DATA ANALYSIS METHODS

Data were analyzed using SAS 8.0 (SAS Institute, Cary NC), SPSS 10.0, Excel, and Access (Office 2000; Microsoft, Redmond WA). The pooled CV (%), used above to characterize precision, is defined as the pooled standard deviation (σ_{pooled}) divided by the mean of pooled measurements. For paired data, $\sigma_{pooled} = [\Sigma d_i^2/2n]^{1/2}$, where d is the difference between paired i values and n is the number of pairs.

To allow subpopulation means to be compared, each subpopulation distribution (or log-transformed distribution) was examined using a Shapiro-Wilk test ($\alpha = 0.05$) to identify subpopulations that are statistically different from normal (or log-normal). These subpopulations were compared with a Kruskal-Wallis nonparametric test. Remaining comparisons were made using t tests or analysis of variance (ANOVA) tests ($\alpha = 0.05$) on the original data or the log-transformed data, as appropriate.

Data below detection limits were included as reported, rather than replacing these values with half the detection limit, for the purpose of calculating summary statistics. For species for which more than 40% of the data were below detection limits, only graphical or descriptive analyses were conducted. Data analysis of species for which 10% to 40% of the data were below detection limits was limited to methods that can accommodate censored data.

Although some homes had two measurements, the intercorrelation between the multiple measurements is not expected to be strong enough to affect statistical analyses performed in this project because the second measurement was taken at least 3 months later than the first measurement. For example, outdoor $PM_{2.5}$ mass concentrations for the first and second visit are poorly correlated (approximately -0.05 to -0.10) and not significant ($\alpha = 0.05$). This is also true of indoor $PM_{2.5}$ mass concentrations. Analyses were conducted with all measurements and repeated for first samples only. The results were not meaningfully different (both sets of results are reported). Subsequent analyses of $PM_{2.5}$ species data were conducted without considering the fact that some homes had multiple measurements.

RESULTS AND DISCUSSION

The overall objective was to improve characterization and prediction of exposure to $PM_{2.5}$ (of indoor and outdoor origin) and further assess the assumptions that underlie current $PM_{2.5}$ epidemiology. Sample collection was designed to include homes with varying AERs in different geographic areas and across seasons, and varying exposures at homes particularly close to and farther from primary $PM_{2.5}$ sources in order to evaluate different exposure concentrations. Speciation studies provided information about $PM_{2.5}$ sources and transport.

$PM_{2.5}$ mass and species concentrations and AERs (for homes with PM sampling) are shown by city in Tables 7, 8, and 9 and by city and season in Appendix C (available on request). Species mass balances were constructed to characterize the composition of indoor, outdoor, and personal $PM_{2.5}$.

Organic carbon, a major component of $PM_{2.5}$, is subject to sampling artifacts; these have been studied extensively in outdoor aerosol research (Heubert and Charlson 2000; Turpin et al 2000), but have only recently been recognized by the exposure assessment community. In this study indoor and outdoor carbon measurements were accompanied by measurements to assess and correct for sampling artifacts so that $PM_{2.5}$ composition would be accurately portrayed. These results will be useful when assessing sampling artifacts in other similar studies.

Results suggest that organic compounds are major contributors to $PM_{2.5}$ emitted or formed indoors and outdoors. Organic $PM_{2.5}$ comprises thousands of compounds spanning a wide variety of vapor pressures and chemical properties. Typically, rigorous molecular-level analyses can account for only 10% to 30% of the organic $PM_{2.5}$ mass (Rogge et al 1993; Turpin and Lim 2001). This study used a combination of total carbon, functional groups, and molecular-level analytical

Table 7. Mean and Median Indoor, Outdoor, and Personal Concentrations of $PM_{2.5}$ Species for Los Angeles Study Homes[a]

	Outdoor		Indoor		Personal Child		Personal Adult	
Species	Mean	Median	Mean	Median	Mean	Median	Mean	Median
$PM_{2.5}$ Mass ($\mu g/m^3$)	19.2	16.1	16.2	14.5	40.2	40.2	29.2	26.5
Carbon ($\mu gC/m^3$)								
EC	1.4	1.2	1.3	1.1				
OC	4.1	3.6	5.4	4.7				
Elements (ng/m^3)								
Ag	0.5	0.3	0.7	0.5	ND	ND	0.7	0.4
Al	24.7	12.7	25.4	16.3	377.9	377.9	75.1	43.4
As	0.5	0.4	0.5	0.4	0.3	0.3	0.5	0.4
Ba	22.9	20.7	17.2	17.0	39.8	39.8	31.7	25.9
Br	5.3	4.7	4.2	3.8	5.6	5.6	6.0	3.8
Ca	80.9	71.5	114.4	78.9	761.2	761.2	264.5	160.8
Cd	0.4	0.1	0.5	0.3	ND	ND	0.7	ND
Cl	62.0	21.1	35.4	22.9	246.9	246.9	80.0	50.2
Co	ND	ND	0.0	ND	ND	ND	0.1	ND
Cr	0.6	0.4	0.9	0.5	1.8	1.8	2.8	1.3
Cu	5.5	4.2	5.4	4.9	17.3	17.3	17.5	7.3
Fe	162.9	149.7	109.5	108.4	477.9	477.9	189.3	154.6
Ga	0.1	ND	0.1	ND	ND	ND	0.2	ND
Ge	0.1	ND	0.1	ND	ND	ND	0.3	0.2
Hg	0.1	ND	0.1	ND	ND	ND	0.3	ND
In	0.3	ND	0.3	ND	ND	ND	0.5	ND
K	74.1	65.5	75.2	70.0	339.3	339.3	117.3	100.8
La	2.3	0.3	2.5	0.0	0.1	0.1	3.9	1.1
Mn	2.9	2.6	2.0	2.0	7.0	7.0	3.1	2.5
Mo	0.4	0.3	0.3	0.2	ND	ND	0.2	ND
Ni	2.0	1.6	1.6	1.5	3.8	3.8	3.9	2.6
Pb	4.7	4.2	3.7	3.6	5.0	5.0	4.8	4.0
Pd	0.3	ND	0.4	0.1	0.9	0.9	0.7	0.4
P	0.1	ND	0.3	ND	6.2	6.2	1.4	ND
Rb	0.1	ND	0.1	ND	0.6	0.6	0.2	0.1
S	1022.9	825.5	916.7	614.4	516.3	516.3	895.3	713.9
Sb	2.1	1.8	1.8	1.5	1.7	1.7	1.9	1.5
Se	1.4	0.9	0.8	0.6	0.6	0.6	0.9	0.7
Si	128.9	107.9	128.6	108.6	901.2	901.2	273.3	191.3
Sn	7.9	5.0	6.8	4.8	6.0	6.0	9.0	6.9
Sr	1.8	1.4	1.5	1.2	6.2	6.2	3.2	1.9
Ti	10.4	9.6	10.9	9.1	44.7	44.7	20.3	15.8
V	5.3	4.5	4.2	3.6	0.8	0.8	3.8	3.2
Y	0.1	ND	0.1	ND	0.2	0.2	0.2	ND
Zn	16.4	13.6	15.7	12.0	29.6	29.6	76.5	28.7
Zr	0.5	0.5	0.9	0.8	2.6	2.6	6.5	1.6

Table continues next page

[a] AER: mean = 1.2; median = 0.9. ND = not detected.

Table 7 (*continued*). Mean and Median Indoor, Outdoor, and Personal Concentrations of $PM_{2.5}$ Species for Los Angeles Study Homes[a]

	Outdoor		Indoor		Personal Child		Personal Adult	
Species	Mean	Median	Mean	Median	Mean	Median	Mean	Median
PAHs (ng/m^3)								
1-MA	1.0	0.87	2.5	2.2				
1-MP	0.83	0.62	1.5	1.3				
2-MA	0.23	0.091	0.61	0.38				
3,6-DMP	0.40	0.32	0.70	0.62				
4,5-MP	0.67	0.58	0.97	0.62				
9,10-DMA	0.032	0.014	0.086	0.043				
9-MA	0.033	0.010	0.19	0.11				
ANT	0.67	0.44	1.0	0.47				
BaA	0.076	0.046	0.037	0.029				
BaFLR	0.10	0.086	0.071	0.057				
BaP	0.10	0.050	0.074	0.041				
BbFLR	0.053	0.045	0.044	0.023				
BeP	0.15	0.11	0.11	0.068				
BFLTs	0.29	0.19	0.19	0.15				
BghiP	0.45	0.27	0.34	0.18				
BNT	0.025	0.019	0.016	0.014				
CHR/Tr	0.25	0.24	0.16	0.16				
COR	0.49	0.23	0.36	0.20				
CPP	0.071	0.038	0.057	0.037				
DBA	0.015	0.0084	0.014	0.0088				
DBT	0.97	0.69	2.5	2.0				
FLT	1.9	1.5	1.6	1.2				
IP	0.37	0.19	0.28	0.18				
PER	0.020	0.012	0.019	0.011				
Phe	11	8.4	16	13				
PYR	1.8	1.6	1.9	1.5				
RET	0.25	0.15	0.45	0.35				
Chlordanes								
OXY	0.017	0.014	0.025	0.018				
TC	0.356	0.257	5.097	0.815				
CC	0.259	0.180	2.695	0.496				
MC5	0.079	0.059	0.557	0.119				
TN	0.176	0.137	1.703	0.380				
CN	0.030	0.022	0.148	0.041				

[a] AER: mean = 1.2; median = 0.9. ND = not detected.

Table 8. Mean and Median Indoor, Outdoor, and Personal Concentrations of $PM_{2.5}$ Species for Elizabeth Study Homes[a]

	Outdoor		Indoor		Personal Child		Personal Adult	
Species	Mean	Median	Mean	Median	Mean	Median	Mean	Median
$PM_{2.5}$ Mass ($\mu g/m^3$)	20.4	18.2	20.1	15.7	54.0	39.2	44.8	37.4
Carbon ($\mu gC/m^3$)								
EC	1.4	1.3	1.4	1.1				
OC	3.3	3.0	7.9	5.4				
Elements (ng/m^3)								
Ag	0.9	0.9	0.9	0.9	1.3	1.4	1.6	1.6
Al	84.9	15.5	13.2	3.4	103.1	89.3	92.3	56.2
As	1.2	1.0	0.9	0.8	1.1	0.7	0.9	0.8
Ba	21.2	17.6	10.8	10.3	17.1	18.1	18.4	18.1
Br	4.1	3.5	3.1	2.5	3.7	3.4	3.4	2.8
Ca	164.9	55.6	71.6	42.1	364.7	286.5	388.8	188.3
Cd	0.6	0.0	0.6	0.3	0.4	ND	0.7	0.2
Cl	129.1	6.8	55.3	12.2	239.1	179.2	185.2	93.6
Co	0.1	ND	0.1	ND	0.5	0.0	0.5	0.1
Cr	7.1	1.5	4.0	0.8	5.9	4.2	5.4	1.7
Cu	11.0	6.9	11.9	4.3	25.9	13.8	17.6	10.7
Fe	278.4	150.9	74.3	58.0	203.8	194.9	208.4	143.0
Ga	0.5	0.5	0.4	0.5	0.8	1.2	0.8	0.9
Ge	0.2	0.1	0.2	0.1	0.3	0.3	0.4	0.3
Hg	0.2	ND	0.2	ND	0.4	0.1	0.3	ND
In	0.3	0.0	0.3	ND	0.2	ND	0.6	ND
K	88.0	58.4	110.0	49.1	211.8	147.8	170.9	106.6
La	2.8	0.5	3.0	1.1	4.5	1.0	3.7	1.3
Mn	5.6	3.7	2.2	1.8	4.1	4.0	4.0	3.3
Mo	0.5	0.4	0.3	0.2	0.1	ND	0.2	ND
Ni	5.3	3.7	3.1	2.6	7.1	5.3	7.4	4.6
Pb	7.5	6.5	5.3	4.2	6.7	5.6	11.4	6.7
Pd	0.3	0.0	0.2	ND	0.4	ND	0.3	ND
P	0.1	ND	0.6	ND	3.1	ND	1.7	ND
Rb	0.2	0.1	6.1	ND	0.3	0.1	0.2	0.0
S	1288.8	1154.0	1011.4	861.3	833.8	755.1	973.6	828.0
Sb	2.1	1.4	1.2	0.8	2.4	1.4	1.3	1.2
Se	1.5	1.3	0.9	0.7	0.8	0.7	0.8	0.7
Si	287.7	107.2	87.7	67.1	346.4	325.4	416.4	230.4
Sn	5.5	3.9	3.9	3.3	7.4	6.2	5.0	3.9
Sr	1.3	0.6	0.4	0.3	1.6	1.4	1.9	1.0
Ti	13.5	6.6	4.2	3.9	28.2	28.1	22.8	17.4
V	6.6	3.9	4.2	2.5	3.8	3.4	3.6	2.5
Y	0.1	ND	0.0	ND	0.1	ND	0.2	ND
Zn	48.1	24.6	112.3	14.3	61.6	41.8	215.2	41.3
Zr	1.3	0.8	0.7	0.5	8.7	7.6	15.2	5.3

Table continues next page

[a] AER: mean = 1.2; median = 0.9. ND = not detected.

Table 8 (*continued*). Mean and Median Indoor, Outdoor, and Personal Concentrations of $PM_{2.5}$ Species for Elizabeth Study Homes[a]

	Outdoor		Indoor		Personal Child		Personal Adult	
Species	Mean	Median	Mean	Median	Mean	Median	Mean	Median
PAHs (ng/m^3)								
1-MA	2.2	2.1	3.4	3.1				
1-MP	1.7	1.5	2.4	2.0				
2-MA	0.89	0.57	0.97	0.50				
3,6-DMP	0.86	0.75	0.93	0.87				
4,5-MP	1.5	1.2	1.2	1.0				
9,10-DMA	0.032	0.020	0.081	0.044				
9-MA	0.051	0.030	0.12	0.11				
ANT	1.7	1.3	1.1	1.0				
BaA	0.21	0.11	0.088	0.059				
BaFLR	0.26	0.21	0.17	0.13				
BaP	0.22	0.12	0.14	0.092				
BbFKR	0.13	0.086	0.052	0.036				
BeP	0.26	0.21	0.14	0.12				
BFLTs	0.53	0.42	0.32	0.25				
BghiP	0.54	0.33	0.37	0.26				
BNT	0.044	0.030	0.027	0.022				
CHR/Tr	0.45	0.36	0.28	0.21				
COR	0.56	0.29	0.36	0.24				
CPP	0.11	0.041	0.072	0.040				
DBA	0.023	0.014	0.014	0.010				
DBT	2.2	1.6	3.5	3.0				
FLT	5.6	3.8	3.6	2.5				
IP	0.55	0.32	0.32	0.21				
PER	0.045	0.027	0.029	0.023				
Phe	29	20	41	21				
PYR	3.8	3.0	2.9	2.3				
RET	0.22	0.14	0.82	0.71				
Chlordanes								
OXY	0.014	0.012	0.029	0.018				
TC	0.239	0.080	1.447	0.449				
CC	0.183	0.057	1.000	0.291				
MC5	0.019	0.010	0.167	0.052				
TN	0.082	0.033	0.581	0.159				
CN	0.007	0.004	0.048	0.014				

[a] AER: mean = 1.2; median = 0.9. ND = not detected.

Table 9. Mean and Median Indoor, Outdoor, and Personal Concentrations of $PM_{2.5}$ Species for Houston Study Homes[a]

	Outdoor		Indoor		Personal Child		Personal Adult	
Species	Mean	Median	Mean	Median	Mean	Median	Mean	Median
$PM_{2.5}$ ($\mu g/m^3$)	14.7	13.2	17.1	13.4	36.6	39.1	37.2	31.6
Carbon ($\mu gC/m^3$)								
EC	0.7	0.7	0.7	0.5				
OC	3.2	2.3	7.2	5.4				
Elements (ng/m^3)								
Ag	0.7	0.7	0.9	0.8	0.9	0.9	1.4	1.3
Al	105.5	21.6	41.8	29.4	188.5	188.5	181.1	149.9
As	1.0	0.9	0.9	0.6	2.1	2.1	1.2	1.1
Ba	14.6	13.6	11.5	11.5	23.9	23.9	24.0	23.0
Br	3.9	3.9	3.4	2.3	4.0	4.0	4.3	3.5
Ca	137.3	109.2	104.8	81.3	513.7	513.7	611.6	497.4
Cd	0.2	ND	0.4	0.1	0.2	0.2	0.6	0.1
Cl	33.4	6.0	111.5	28.9	215.1	215.1	237.1	155.5
Co	0.4	ND	0.3	ND	0.4	0.4	0.5	0.2
Cr	1.1	0.6	1.2	0.5	0.3	0.3	2.3	1.6
Cu	1.9	1.5	4.5	2.7	4.0	4.0	12.6	10.4
Fe	118.2	72.2	49.4	42.2	145.3	145.3	171.5	136.8
Ga	0.2	ND	0.1	ND	0.5	0.5	0.1	ND
Ge	0.1	0.1	0.2	0.1	0.2	0.2	0.2	0.1
Hg	0.2	0.0	0.2	ND	0.6	0.6	0.3	ND
In	0.2	ND	0.3	0.0	0.3	0.3	0.4	ND
K	97.6	82.9	88.5	64.9	157.2	157.2	182.9	153.9
La	2.5	0.7	2.2	0.4	ND	ND	3.3	0.3
Mn	4.3	3.2	2.1	1.5	3.3	3.3	4.3	3.8
Mo	0.4	0.3	0.3	0.2	ND	ND	0.2	ND
Ni	2.2	1.8	1.2	1.0	1.1	1.1	4.1	3.3
Pb	2.8	2.4	1.8	1.5	2.0	2.0	3.9	3.0
Pd	0.3	0.0	0.3	ND	0.6	0.6	0.4	ND
P	0.1	ND	2.4	ND	4.9	4.9	3.6	0.1
Rb	0.1	0.0	0.1	0.1	0.6	0.6	0.3	0.2
S	1158.3	1086.0	651.6	577.4	457.3	457.3	700.4	635.0
Sb	1.7	1.1	0.9	0.7	ND	ND	1.9	1.8
Se	0.6	0.6	0.3	0.3	0.5	0.5	0.4	0.3
Si	261.9	121.1	166.1	147.3	513.8	513.8	574.7	460.1
Sn	2.7	2.3	3.1	2.8	4.5	4.5	4.9	4.5
Sr	0.7	0.5	0.5	0.5	1.9	1.9	2.3	2.0
Ti	8.9	4.7	5.6	4.5	22.1	22.1	28.9	20.9
V	5.4	4.2	2.7	2.0	1.9	1.9	2.6	2.1
Y	0.1	ND	0.1	ND	0.2	0.2	0.2	ND
Zn	13.7	9.0	54.2	7.5	24.7	24.7	70.9	34.2
Zr	0.4	0.2	0.8	0.7	5.2	5.2	11.6	5.8

Table continues next page

[a] AER: mean = 0.7; median = 0.5. ND = not detected.

Table 9 (*continued*). Mean and Median Indoor, Outdoor, and Personal Concentrations of $PM_{2.5}$ Species for Houston Study Homes[a]

	Outdoor		Indoor		Personal Child		Personal Adult	
Species	Mean	Median	Mean	Median	Mean	Median	Mean	Median
PAHs (ng/m^3)								
1-MA	1.8	1.1	5.0	4.9				
1-MP	1.1	0.82	2.9	2.9				
2-MA	0.29	0.19	0.67	0.49				
3,6-DMP	0.66	0.47	1.3	1.2				
4,5-MP	1.2	0.93	1.3	1.3				
9,10-DMA	0.11	0.020	0.24	0.10				
9-MA	0.094	0.023	0.20	0.15				
ANT	1.0	0.69	1.7	0.97				
BaA	0.057	0.031	0.062	0.026				
BaFLR	0.20	0.13	0.15	0.12				
BaP	0.078	0.049	0.072	0.027				
BbFLR	0.078	0.052	0.051	0.033				
BeP	0.085	0.053	0.080	0.038				
BFLTs	0.20	0.14	0.20	0.091				
BghiP	0.17	0.074	0.25	0.046				
BNT	0.042	0.026	0.031	0.029				
CHR/Tr	0.67	0.50	0.46	0.31				
COR	0.13	0.049	0.35	0.036				
CPP	0.037	0.014	0.095	0.0090				
DBA	0.012	0.0067	0.014	0.0040				
DBT	2.1	1.5	5.1	4.2				
FLT	3.9	3.1	3.0	2.4				
IP	0.18	0.082	0.29	0.060				
PER	0.014	0.011	0.022	0.011				
Phe	22	15	32	25				
PYR	2.8	2.4	2.9	2.4				
RET	0.73	0.45	1.2	0.85				
Chlordanes								
OXY	0.011	0.010	0.068	0.015				
TC	0.177	0.085	4.737	1.521				
CC	0.115	0.061	3.139	0.973				
MC5	0.030	0.017	0.551	0.181				
TN	0.078	0.042	1.744	0.564				
CN	0.011	0.007	0.132	0.062				

[a] AER: mean = 0.7; median = 0.5. ND = not detected.

tools to derive further insights into the sources and composition of OC because it is a major and chemically complex constituent of $PM_{2.5}$.

Epidemiologic studies use measurements from outdoor central-site monitors as surrogates for personal exposure to $PM_{2.5}$ of outdoor origin. To better understand the sources of exposure and to assess the validity of exposure surrogates used in epidemiologic studies, it is useful to separate indoor and personal $PM_{2.5}$ concentrations into PM of outdoor origin and PM of indoor origin. Several approaches have been applied to achieve this; some use $PM_{2.5}$ mass concentrations and others use $PM_{2.5}$ species concentrations. The impact of various assumptions on the distribution of exposures was used to better understand what key parameters are needed for exposure prediction and the impact of exposure misclassification on epidemiologic results.

One goal of this project was to explore the suitability of the data set for source apportionment. Throughout this work speciation data provided insights regarding $PM_{2.5}$ sources. In addition, speciation data were prepared for positive matrix factorization (PMF), a factor analysis method that takes into consideration measurement uncertainty. Factor analysis identifies factors of covariant species. Species can vary together because they are emitted from the same source type, because they are transported together from the same source region or microenvironment, or because they are emitted or formed with the same temporal pattern.

Preliminary PMF analyses were conducted on the indoor $PM_{2.5}$ data. The PMF results are exploratory and therefore are not reported here. Nevertheless, the work thus far has shown that the data are of sufficient quality to identify at least seven factors, two representing indoor sources and five representing outdoor sources. On the basis of these results, we are confident that future efforts to expand the analysis across the three study cities, finalize the results, and conduct a sensitivity analysis would be worthwhile. In addition to the direct benefits of source apportionment, we expect the source apportionment results to aid the validation of an aerosol model to improve prediction of exposure to PM of outdoor origin.

INDOOR, OUTDOOR, AND PERSONAL $PM_{2.5}$ MASS COMPOSITION

$PM_{2.5}$ Mass Concentrations and AERs

Median indoor, outdoor, and personal $PM_{2.5}$ mass concentrations were 14.4 $\mu g/m^3$, 15.5 $\mu g/m^3$, and 31.4 $\mu g/m^3$, respectively. $PM_{2.5}$ mass concentrations and AERs were approximately log-normally distributed, as suggested by a Shapiro-Wilk test on the log-transformed data ($\alpha = 0.05$). Personal $PM_{2.5}$ concentrations were significantly greater than indoor and outdoor concentrations as determined by one-way ANOVA and the Scheffe test ($\alpha = 0.05$; $P < 0.0001$) performed on the log-transformed data. This was still true when PEM and Harvard impactor measurements were "harmonized" using equation 2. Personal concentrations were also more variable than both indoor and outdoor concentrations according to a Levene test ($\alpha = 0.05$) for the overall study data and for data segregated by city, with the exception of Los Angeles outdoor concentrations. Indoor concentrations for Houston homes were more variable than outdoor concentrations (the Levene test; $\alpha = 0.05$).

For each home, indoor, outdoor, and personal concentrations were compared using an incomplete randomized block mixed model (SAS Institute, Version 8) by treating the specific home identification number as a random block effect, and sample categories (indoor, outdoor, and personal concentrations) as treatment effects. 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 (Proc Mixed, and type = un for error structure). The added power obtained by pairing indoor, outdoor, and personal concentrations from the same home verified that personal concentrations were higher than indoor and outdoor concentrations for all three cities, and revealed that outdoor concentrations were significantly higher than indoor concentrations for Elizabeth and Los Angeles homes, as well as for the overall data set. The same conclusions were obtained when only the first sample from each home was used in the analysis; this confirmed that the conclusions are not artifacts of within-home correlation.

The observation of personal exposure concentrations that exceed indoor and outdoor concentrations is consistent with the findings of many other studies. The average $PM_{2.5}$ personal cloud concentration for this study was 17 $\mu g/m^3$. The review by Wallace (2000) reported $PM_{2.5}$ personal cloud values of 11 to 27 $\mu g/m^3$ for healthy populations, and 6 $\mu g/m^3$ for populations with chronic obstructive pulmonary disease. Personal concentrations could be higher than residential indoor and outdoor concentrations because the participant spent time in another, higher-concentration microenvironment (eg, a smoky bar or restaurant), or in closer proximity to indoor sources than the indoor monitor (eg, while cooking).

Although smokers were effectively excluded from this study (as validated by the personally administered Activity Questionnaire), passive exposure to environmental tobacco smoke (ETS) was a potential contributor to personal exposures. As part of the Activity Questionnaire, participants were asked if they had been in an area where smoking

occurred during the sample collection period. Questionnaire responses suggested that passive tobacco smoke exposure influenced fewer than 15 samples. It is unlikely that ETS exposure influenced the median personal exposures for the study, but it could be a significant contributor to the highest exposure concentrations. For example, two subjects who reported ETS exposure had personal exposure concentrations of 96.5 μg/m^3 and 66.0 μg/m^3; these concentrations were greater than the 95th and 90th percentiles of measured personal exposure concentrations. Other personal activities can also have a considerable influence on personal exposures.

Scatter plots in Figure 9 show pairs of indoor, outdoor, and personal PM2.5 concentrations; Table 10 provides coefficients of determination. 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), which reflects 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 $PM_{2.5}$ mass concentrations was less than one (r^2 = 43% to 80%; I/O < 1 in 54% to 71% of homes by city). The higher correlations occurred presumably because of low indoor source strengths or high AERs (or both) in these homes. Correlations of outdoor or indoor $PM_{2.5}$ concentrations with personal $PM_{2.5}$ concentrations were not much greater for these homes than for all homes.

Table 10. Coefficients of Determination (r^2) Between Indoor, Outdoor, and Personal $PM_{2.5}$ Concentrations[a]

Homes	Indoor vs Outdoor	Personal vs Indoor	Personal vs Outdoor
Overall study			
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 where I/O $PM_{2.5}$ ratio is < 1.

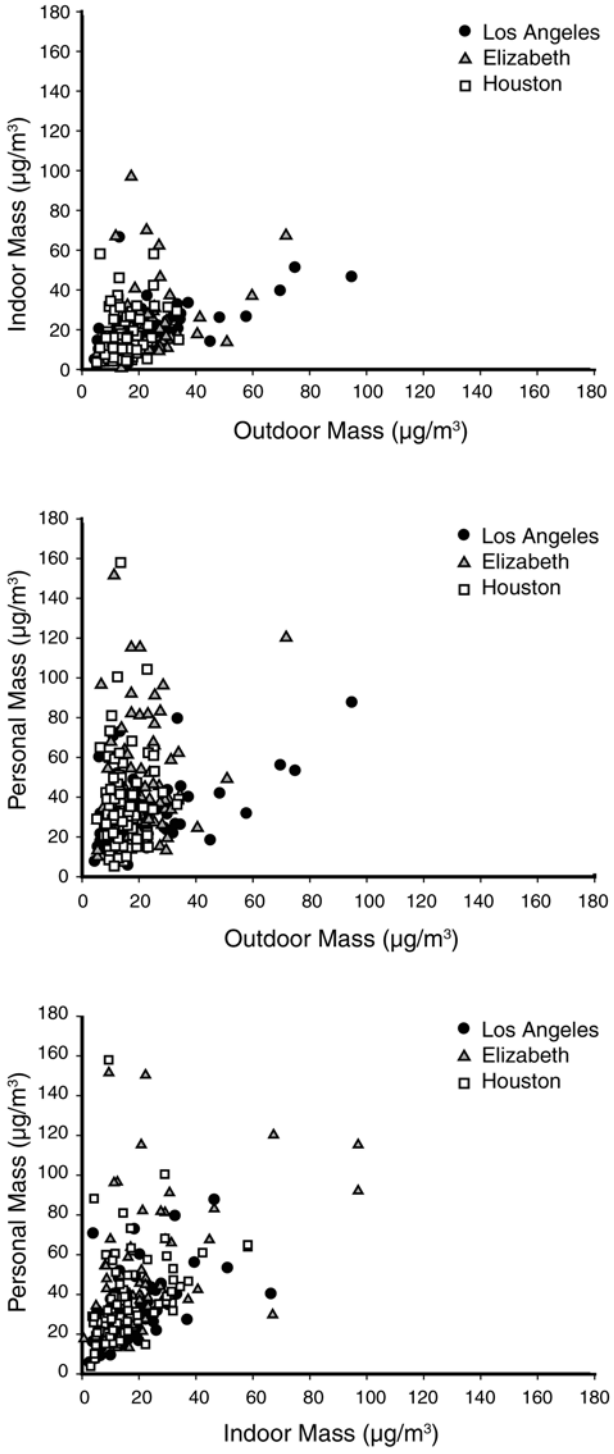


Figure 9. Comparison of indoor–outdoor, personal–outdoor, and personal–indoor $PM_{2.5}$ mass concentrations in Los Angeles, Elizabeth, and Houston homes.

The mean outdoor $PM_{2.5}$ concentration for the Los Angeles samples (19.2 μg/m^3) was similar to that measured in the winter 1999 $PM_{2.5}$ exposure studies in Fresno, California (20.5 μg/m^3; Vette et al 2001). However, the outdoor $PM_{2.5}$ mass concentrations in the current study (mean, 19.2 μg/m^3; median, 16.1 μg/m^3) were much lower than those in the fall 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 mass concentrations for Los Angeles samples in the current study were less variable than PTEAM study samples (σ = 13.3 μg/m^3 or 69% in this study; σ = 37.6 μg/m^3 or 77% for the daytime and 40.3 μg/m^3 or 80% for the nighttime in the PTEAM study; Clayton et al 1993).

Los Angeles indoor concentrations in the current study were higher than the Fresno concentrations (9.7 μg/m^3 and 8.0 μg/m^3 for winter and spring, respectively) and much lower than the PTEAM study concentrations (48.2 μg/m^3 and 36.2 μg/m^3 for daytime and nighttime, respectively).

The differences between findings in the current study and 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 pollutants transported across the basin. In contrast, the homes in this study are in the western half of the Los Angeles Basin, closer to primary sources. Air quality in the Los Angeles Basin has also improved over the last 10 years, although PM concentrations have declined more modestly than ozone concentrations. In addition, the PTEAM study included homes with smokers.

The annual average central-site monitor $PM_{2.5}$ mass concentration in Elizabeth was 16.4 μg/m^3 for the period July 1997 to June 1998 (Chuersuwan and Turpin 2000), which is close to the outdoor residential median concentration of 18.2 μg/m^3 measured in this study, and somewhat lower than the mean of 20.4 μg/m^3.

Comparisons can also be drawn with studies conducted in other locations. For Birmingham, Alabama, Lachenmyer and Hidy (2000) reported 48-hour average $PM_{2.5}$ mass concentrations of 12.2 μg/m^3 outdoors and 11.2 μg/m^3 indoors in winter 1998, and 26.5 μg/m^3 outdoors and 16.1 μg/m^3 indoors 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 (Pellizzari et al 1999). In the EXPOLIS (Air Pollution Exposure Distributions of Adult Urban Populations in Europe) study in Helsinki, Finland, 1996–1998 (Koistinen et al 2001), median indoor, outdoor, and personal concentrations were 11.7 μg/m^3, 7.3 μg/m^3, and 21.6 μg/m^3, respectively, for subjects who smoked; indoor and personal concentrations for subjects who did not smoke were 6.9 μg/m^3 and 7.8 μg/m^3, respectively.

AERs for Los Angeles homes in this study (mean, median, and SD of 1.22 hr^{-1}, 0.93 hr^{-1}, and 0.87 hr^{-1}, respectively) were similar to those measured in a Los Angeles survey during 1984 and 1985 (mean, median, and SD of 1.51 hr^{-1}, 1.07 hr^{-1}, and 1.47 hr^{-1}, respectively; Wilson et al 1996). In a wintertime 1991–1992 study of AERs in the Los Angeles area, mean, median, and SD of AERs were 0.79 hr^{-1}, 0.64 hr^{-1}, and 0.5 hr^{-1}, respectively, whereas the wintertime mean, median, and SD of Los Angeles AERs for samples in this study were 0.83 hr^{-1}, 0.76 hr^{-1}, and 0.47 hr^{-1}, respectively (Pandian et al 1998).

Pandian and associates (1998) summarized 4590 AER measurements made in residences nationwide during different studies. New Jersey and Texas were included in the northeast and southeast regions of that study. Mean, median, and SD 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 (after removal of two outliers). In the current study, AERs in Houston (mean, median, and SD of 0.71 hr^{-1}, 0.46 hr^{-1}, and 0.73 hr^{-1}, respectively) were similar to the southeast region survey data, but those in Elizabeth (mean, median, and SD of 1.22 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 the difference in study findings is the considerable difference in the maximum measurable AER between studies. AERs are calculated from the home volume and the concentration of a perfluorocarbon tracer emitted at a known rate (Dietz et al 1986; Weisel et al 2004). Lower concentrations correspond to higher AERs, so the MDL of the perfluorocarbon tracer results in an upper detection limit for AER measurements of 5 air changes per hour. The highest AER reported by Pandian was approximately 2 changes per hour. Thus, it is quite possible that AERs reported for Elizabeth in the current study are higher than those previously reported for New Jersey because we were able to measure AERs across a larger dynamic range. Also, AERs in New Jersey are expected to vary considerably between areas that have primarily older homes (built 1910 to 1940), such as Elizabeth, and other areas that have primarily homes built after 1940 or after 1970. AERs in older homes (eg, those sampled in this study) are likely to be higher than AERs for homes built in the late 1900s. (Note: Only AERs for homes with PM sampling are included in these statistics.)

Organic Aerosol Sampling Artifacts

Numerous organic compounds partition between the gas and particle phases. Their vapor pressure, the ambient temperature, and the quantity and properties of the PM into or onto which they sorb all affect the partitioning between phases. During sampling, the particle phase is collected by pulling the vapor phase through an initially clean filter with a surface area for adsorption that is much larger than the surface area of the particles that are ultimately collected. The amount of organic vapor that adsorbs on the QFF largely depends on the face velocity through the filter, sampling duration, temperature, and the composition and concentration of atmospheric organic vapors.

For samples collected outdoors in a manner similar to that used in the current study, typically 30% to 50% of measured OC is adsorbed vapor (Turpin et al 2000). If uncorrected, this would result in a substantial overestimate of particulate OC concentrations. In the current study, as in many other outdoor air pollution studies (Turpin et al 2000), the magnitude of the adsorption artifact was estimated by collecting a dynamic blank concurrently with the sample. Specifically, a QFF sampled air downstream of the Teflon filter in the Harvard impactor. The amount of adsorbed vapor on this backup QFF provided an estimate of the quantity of adsorbed organic vapor on the concurrently collected QFF in the MSP sampler.

Figure 10 shows the percentage of measured OC (ie, gases + particles collected on the QFF in the MSP sampler) that is adsorbed vapor (ie, vapor adsorbed to the concurrently collected backup QFF in the Harvard impactor). This OC artifact is expressed as a percentage of measured OC. As has been shown elsewhere, the bias introduced by the adsorption artifact becomes less important (a smaller percentage of the sample) as the loading of PM increases. At small sample loadings, adsorbed vapors can dominate the sampled mass. Measured OC concentrations on the MSP QFF tended to be higher indoors than outdoors. At the median OC concentrations of 8.2 μgC/m^3 indoors and 5.0 μgC/m^3 outdoors, the percentages of measured OC that were adsorbed vapor indoors (36%) and outdoors (37%) were nearly identical. However, at any single measured OC concentration, the indoor artifact was larger than the outdoor artifact. This suggests that organic vapors indoors have a greater tendency to adsorb to QFFs than organic vapors outdoors, presumably owing to differences in source mix and composition. Adsorption artifact behavior did not appear to differ substantially among the three cities.

The adsorption artifact results for this study are consistent in magnitude and functional dependence with those in previous outdoor studies (Turpin et al 2000; Lim et al 2003b). Uncorrected, an artifact of this magnitude would

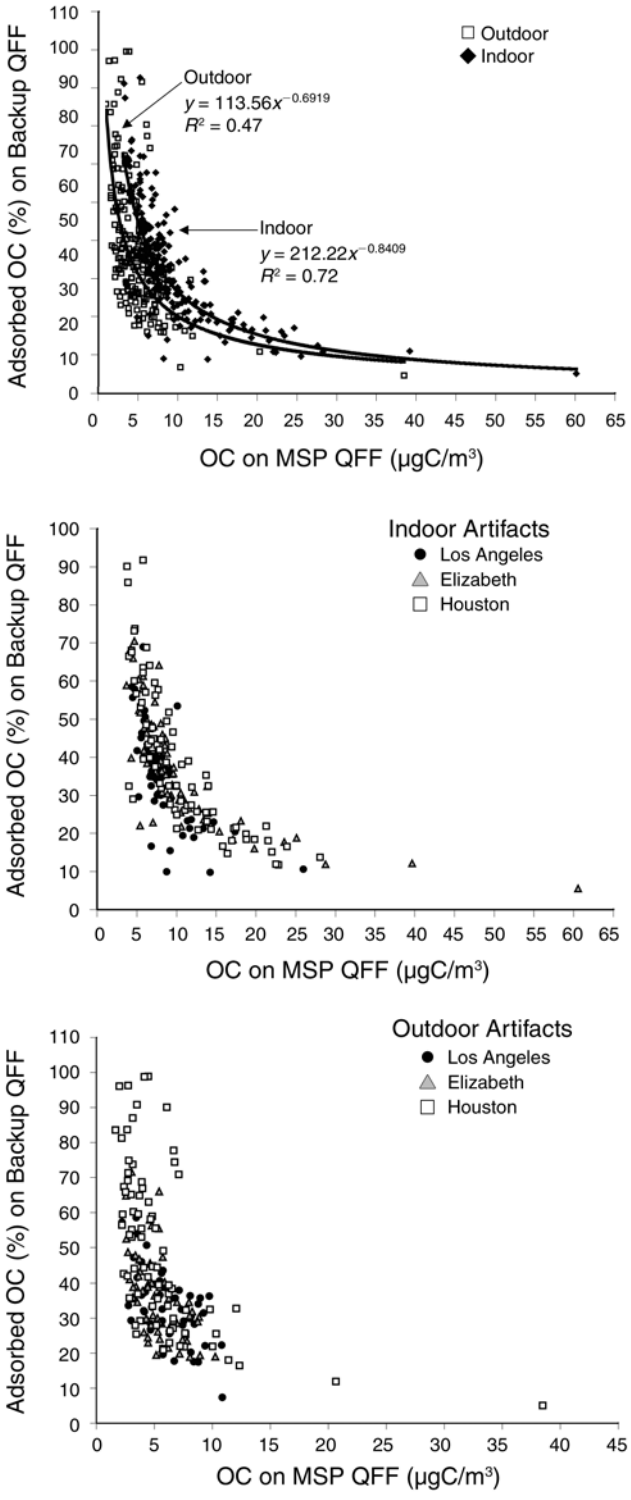


Figure 10. Percentage of measured OC (gas phase + particle phase) that is adsorbed vapor, an indicator of OC artifact. To determine adsorbed vapor, OC collected on the backup QFF in the Harvard impactor was divided by OC collected on the QFF in the collocated MSP sampler. Top: Indoor and outdoor samples, all cities. Middle: Indoor samples only, by city. Bottom: Outdoor samples only, by city.

clearly result in substantial bias in reported particulate OC concentrations. This finding could explain some previous exposure study reports (eg, Landis et al 2001) of personal $PM_{2.5}$ carbon concentrations that exceeded the total $PM_{2.5}$ mass concentrations. (Note that particulate OC concentrations used in data analyses reported here were corrected for the adsorption artifact on a sample-by-sample basis. At some homes backup filters were not used. For those samples the equations in Figure 10a were used to estimate the adsorption artifact, which resulted in larger uncertainties for these estimates.)

Other positive and negative sampling artifacts that were not estimated in this study can also occur due to changing ambient conditions during sampling, a pressure drop across the sampling filter, or chemical interactions. Turpin and coworkers (2000) have presented an extensive discussion of sampling artifacts affecting particulate OC. Briefly, although chemical interactions during sampling have been shown to alter the concentrations of individual organic compounds, there is little evidence that such reactions significantly alter total particulate OC concentrations. In addition, the pressure drop across a 37-mm QFF at 10 L/min is quite small, and the volatile losses induced by this pressure drop are calculated to be small compared with the adsorption artifact described above (McDow and Huntzicker 1990). Changes in microenvironmental conditions during sampling, such as temperature, relative humidity, and gas-phase concentrations, can result in additional positive (adsorption) and negative (volatilization) sampling artifacts because they alter the equilibrium between the gas phase (passing through the filter) and the sorbed phase (on collected particles and filter). Thus changes in microenvironmental conditions yield a sample that is weighted toward the conditions present at the end of the sampling period.

Species Mass Balance

In Figure 11 the mean species contributions to the indoor and outdoor $PM_{2.5}$ mass concentrations are shown by city. Table 11 provides the indoor and outdoor species contributions for the homes in the highest and lowest 25th percentiles of outdoor $PM_{2.5}$ mass. These results illustrate the importance of indoor sources of organic PM and are consistent with the substantial loss of particulate nitrate

Table 11. Mean Indoor and Outdoor Species Contributions for Homes in the Highest and Lowest 25th Percentiles by Outdoor $PM_{2.5}$ Mass[a]

Category	Mass Concentration	Soil	Sulfate	OM	EC	Other[b]
Highest 25th Percentile						
Los Angeles						
Indoor	27.0	1.0	7.3	13.0	1.2	4.5
Outdoor	35.6	0.7	7.7	6.1	1.2	19.9
Elizabeth						
Indoor	38.7	1.0	5.3	21.6	1.8	9.0
Outdoor	26.2	1.3	7.5	7.1	1.8	8.5
Houston						
Indoor	32.1	1.2	3.6	21.6	0.9	4.8
Outdoor	23.1	1.7	6.7	6.1	0.8	7.8
Lowest 25th Percentile						
Los Angeles						
Indoor	9.3	0.8	2.1	5.2	1.1	0.1
Outdoor	10.9	0.9	2.9	4.9	1.1	1.1
Elizabeth						
Indoor	8.1	0.3	2.6	4.3	0.8	0.1
Outdoor	8.1	0.4	3.0	3.1	0.8	0.8
Houston						
Indoor	8.1	0.6	2.4	4.2	0.5	0.4
Outdoor	8.8	0.6	3.3	2.1	0.4	2.4

[a] All values are given in $\mu g/m^3$. Soil is sum of oxides; sulfate is ammonium sulfate; OM is estimated as 1.4 × OC; "other" is the difference between the measured mass and the sum of the measured species.

[b] The major component of this category is expected to be ammonium nitrate.

indoors in California homes proposed by Lunden and colleagues (2003).

Sulfur determined from XRF was assumed to be in the form of ammonium sulfate, and OC concentrations were multiplied by 1.4 to estimate particulate organic matter (OM; 1.4 is an estimate of the proportion of average organic molecular weight per carbon weight, OM/OC; Turpin and Lim 2001). Soil dust concentrations were calculated as the sum of the oxides of aluminum, silicon, calcium, titanium, iron, and potassium (Brook et al 1997; Lee et al 2002). These assumptions are common in $PM_{2.5}$ species mass balance calculations. In the eastern United States, the sulfate contribution could be somewhat overestimated by assuming that sulfate was completely neutralized. A previous evaluation of assumptions in species mass balances (Turpin and Lim 2001) suggested that an average organic molecular weight

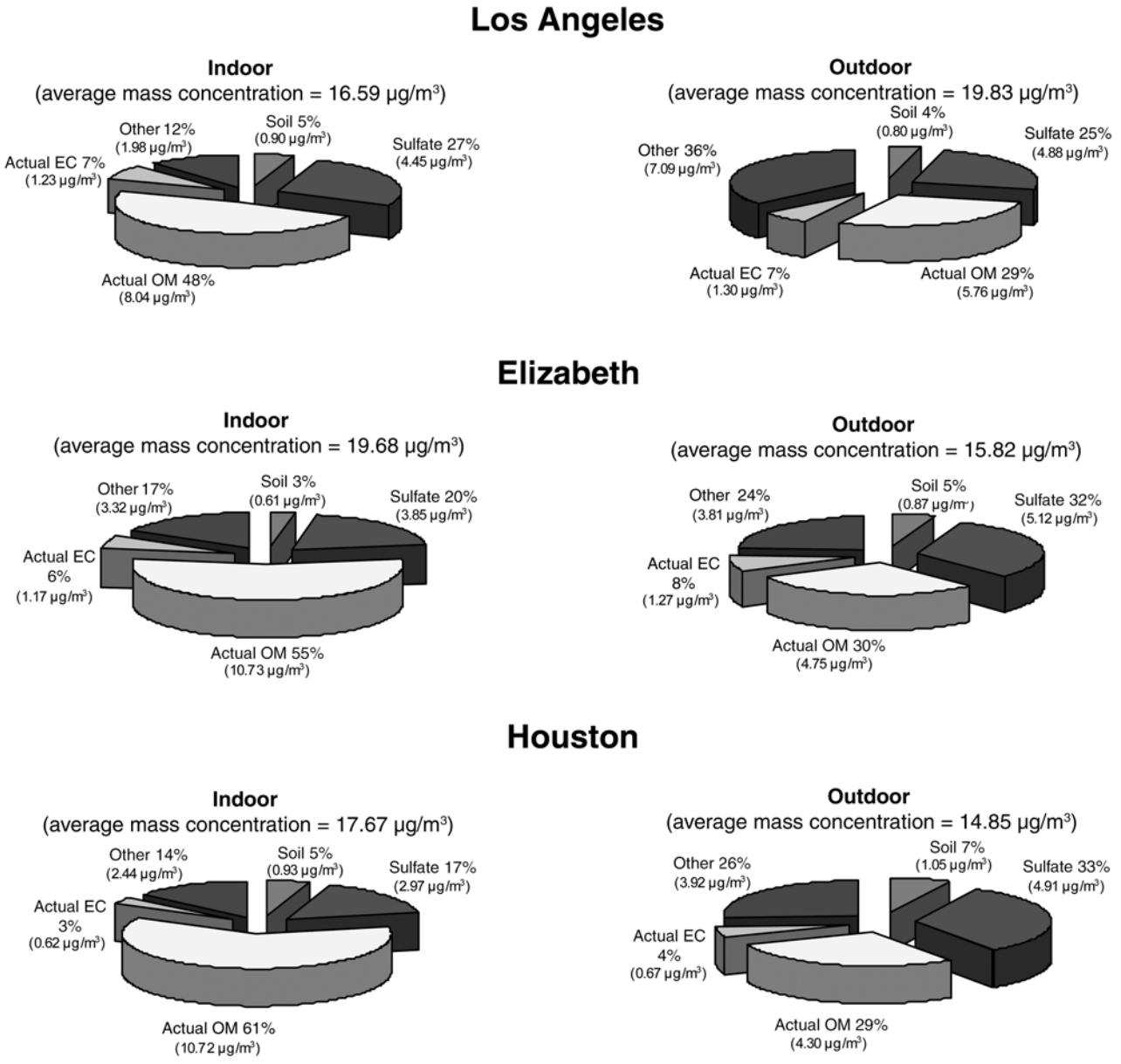


Figure 11. Mean species contributions, by percentage and concentration, to indoor and outdoor $PM_{2.5}$ mass concentrations for Los Angeles, Elizabeth, and Houston homes. Soil is sum of oxides; sulfate is ammonium sulfate; OM was estimated as 1.4 × OC; "other" is the difference between the measured mass and the sum of the measured species (ammonium nitrate is expected to be the major component of this category). n = 125 homes.

per carbon weight of 1.4 to 1.6 is reasonable in urban areas. The only major PM constituents not measured in this study were ammonium nitrate and water. These are the main constituents of the category called "other", which constitutes the difference between the mean $PM_{2.5}$ mass concentration and the sum of measured species.

Outdoor mass balance results in this study are in reasonable agreement with those in other urban studies. The category designated "other," which includes nitrate, was a larger contributor to $PM_{2.5}$ mass in Los Angeles samples than in samples from the other two cities (Figure 11), and was a larger contributor to outdoor $PM_{2.5}$ mass on high $PM_{2.5}$ days than on low $PM_{2.5}$ days (Table 11). Sulfate contributed a larger percentage to Elizabeth and Houston samples than to Los Angeles samples, and the soil contribution was slightly larger in Houston samples than in both other cities (Figure 11).

The most notable observation in the species mass balance was that the mean particulate OM concentration indoors (OM = 9.8 $\mu g/m^3$) was nearly twice the mean outdoor concentration (OM = 4.9 $\mu g/m^3$). In contrast, the mean EC concentration was 1.1 $\mu g/m^3$ indoors and outdoors. In fact, for Elizabeth and Houston homes, the concentrations and percentage contributions of all species except OM were the same or somewhat smaller indoors than outdoors (Figure 11). The elevated concentrations of organic PM indoors suggest that it was emitted or formed indoors in sufficient quantities to substantially alter the concentration and composition of $PM_{2.5}$ indoors, where people spend most of their time.

The results of indoor–outdoor comparisons for Los Angeles homes were somewhat different. Like Elizabeth and Houston homes, Los Angeles homes had substantially higher concentrations of organic PM indoors. In addition, for Los Angeles homes the concentration (and percentage contribution) of "other" to the $PM_{2.5}$ mass concentration was substantially smaller indoors (2.0 $\mu g/m^3$; 12%) than outdoors (7.1 $\mu g/m^3$; 36%; Figure 11), a difference that was particularly pronounced on high $PM_{2.5}$ days (Table 11). Because the largest component of "other" is expected to be ammonium nitrate, this finding is consistent with the modeling and controlled experimental results of Lunden and colleagues (2003). Their studies suggest that losses of nitric acid to indoor surfaces drive a redistribution of nitrogen from the particle phase (ammonium nitrate) to the gas phase (nitric acid) as it is transported indoors from outdoors. The lower contribution of "other" in Los Angeles samples (compared with those from Houston and Elizabeth) more than makes up for the higher contribution of OM, so the *percentage* contribution of ammonium sulfate was actually slightly higher indoors, despite the fact that the mean ammonium sulfate concentration was slightly lower indoors. The loss of "other" PM provides some evidence that the composition of *indoor* $PM_{2.5}$ of *outdoor origin* can differ substantially from that of outdoor residential and central-site $PM_{2.5}$, and that the relation between central-site $PM_{2.5}$ mass and indoor $PM_{2.5}$ of outdoor origin might not vary linearly in locations where ammonium nitrate is a major outdoor $PM_{2.5}$ constituent.

ORIGIN AND COMPOSITION OF ORGANIC $PM_{2.5}$

The contribution of organic compounds to $PM_{2.5}$ exposure, though substantial, is complex and poorly understood (EPA 2004). Therefore, we designed data analyses to provide more insight into the origin and composition of organic PM in study homes. This report presents one of the first assessments of the contributions of indoor and outdoor sources to indoor concentrations of particulate OC. Molecular-level analysis of atmospheric organic PM is typically able to identify only 10% to 30% of the organic mass. However, FTIR spectroscopy provides functional group information on the entire sample. FTIR spectroscopy can be used to assess the polarity and chemical functionality of the aerosol; it is useful from the standpoint of understanding aerosol properties and behavior; and it provides some insights into aerosol sources.

Some molecular-level tracers, in particular PAHs and chlordanes, have also been measured in this study to aid in source identification. These constitute only a small fraction of $PM_{2.5}$ mass. Several are identified air toxics.

Indoor and Outdoor Contributions to Carbon

Inside homes, particulate OM constituted these percentages of $PM_{2.5}$ mass: 48% ± 16% in Los Angeles, 55% ± 18% in Elizabeth, and 61% ± 22% in Houston (OM was estimated as 1.4 × OC; see Figure 11 and Table 11). The indoor and outdoor concentrations of OC and EC are displayed by city in Figure 12. Particulate OC was substantially higher indoors than outdoors for many Los Angeles, Elizabeth, and Houston homes. In addition, indoor and outdoor OC concentrations were poorly correlated (r^2 = 0.01). These observations suggest that many homes had substantial indoor sources of particulate OC. In contrast, with a few exceptions, paired indoor and outdoor EC concentrations were similar. After removing one to three outliers, within-city indoor and outdoor EC concentrations were reasonably well correlated (r^2 = 43% to 79%). Only two homes had dramatically higher EC concentrations indoors than outdoors, suggesting that substantial indoor emissions of EC were rare.

Primary OC in the particle phase can be directly emitted indoors from sources such as cooking, and secondary particulate OC can be formed in indoor air as a result of reactions involving reactive gas-phase organic compounds and ozone (Weschler and Shields 1997). Outdoors, OC also has primary sources, and photochemical reactions can generate substantial secondary OC when conditions are favorable (Turpin and Huntzicker 1995; Lim and Turpin 2002; Pandis et al 1992). EC is formed through incomplete combustion and is a good tracer for primary, combustion-generated OC. It is also frequently used as a tracer for diesel PM.

Figure 13 shows that the correlation between OC and EC was stronger outdoors than indoors. In addition, the ratio of OC to EC was higher indoors than outdoors. Assuming that all EC originated outdoors, a weaker indoor correlation and a higher indoor ratio of OC to EC is consistent with a substantial indoor source of OC.

The mean contributions of indoor and outdoor sources to indoor OC concentrations were estimated using the random component superposition (RCS) statistical model (Ott et al 2000). This approach and a variety of others are discussed in detail in the section Results and Discussion / Outdoor Contributions to Indoor and Personal $PM_{2.5}$. Briefly, the RCS model provides a constant infiltration factor from the linear regression of indoor OC concentrations on outdoor OC concentrations. The product of this

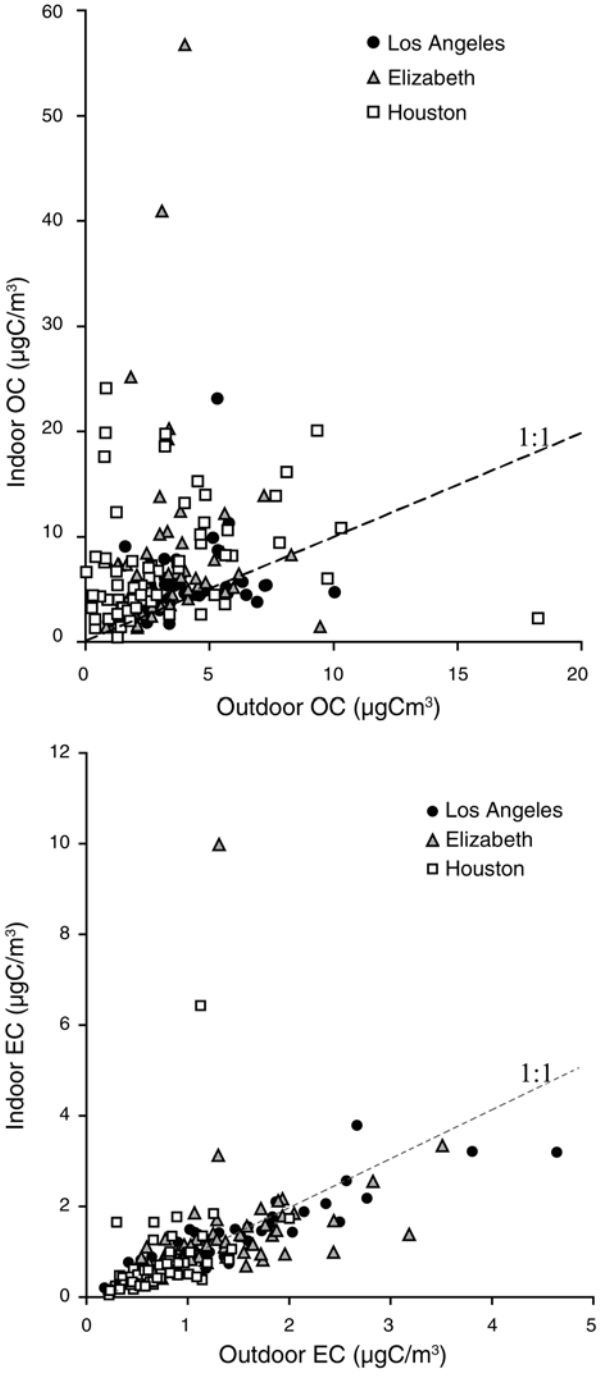


Figure 12. Indoor and outdoor concentrations of OC and EC for Los Angeles, Elizabeth, and Houston homes. Note that the axis lengths differ between panels.

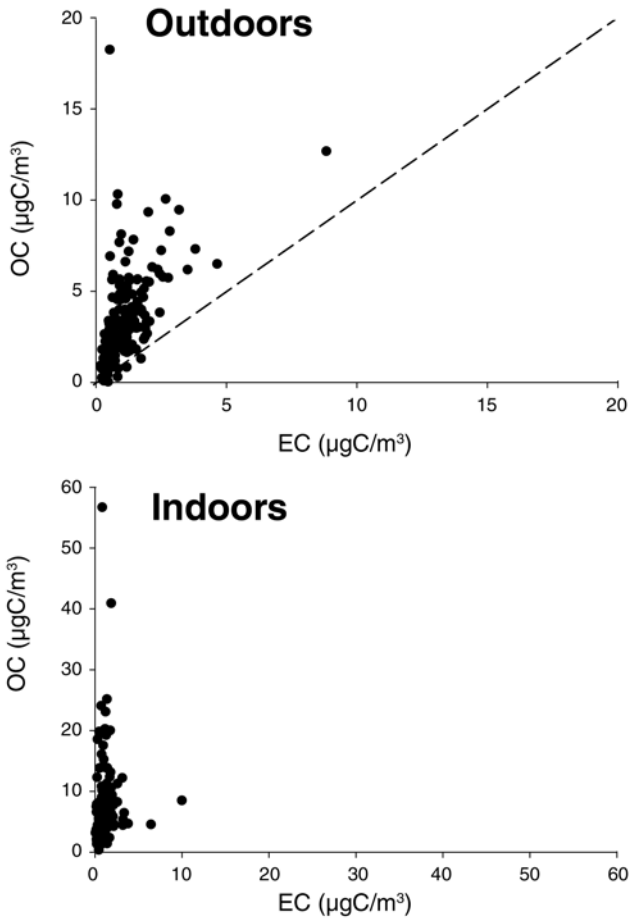


Figure 13. OC and EC concentrations outdoors and indoors. n = 173.

infiltration factor and each outdoor concentration provides an estimate of the distribution of OC of outdoor origin for the homes. The distribution of indoor contributions to indoor OC concentrations is given by the difference between the measured indoor OC concentration and the OC of outdoor origin calculated for each home.

The RCS model assumes a linear superposition of OC of outdoor origin and OC of indoor origin and a lack of correlation between these two components. Using this approach 76%, on average, of OC found indoors was emitted or formed indoors, rather than being transported indoors from outdoor sources. Although the uncertainties around this number have not been explored, this finding is reasonable, especially in light of the following lower-bound calculation. If the penetration of particles through the building envelope was 1.0 and there were no particle losses indoors, then 41% of indoor OC would be emitted or formed from indoor sources at the mean indoor OC concentration of 7.00 μgC/m^3 (OM = 9.80 μg/m^3) and mean outdoor OC concentration of 3.49 μgC/m^3 (OM = 4.88 μg/m^3).

Organic Functional Groups

FTIR spectroscopy provides functional group and bond information about the entire $PM_{2.5}$ sample without any chemical preparation. It is semiquantitative, nondestructive, and provides what is sometimes described as a "chemical snapshot" of the aerosol. FTIR spectroscopy has been used to gain insights into the origin and behavior of outdoor organic PM by examining the polarity and size distributions of compound classes (Pickle et al 1990; Mylonas et al 1991; Blando et al 1998). Carlton and associates (1999) demonstrated the application of FTIR spectroscopy to exposure assessment. To our knowledge the current study is the first to use FTIR spectroscopy for $PM_{2.5}$ exposure analysis.

FTIR Spectroscopy Results The spectra in Figure 14 are from individual home samples and illustrate FTIR absorbances typically observed in this study. Note that the Teflon filters absorbed infrared light strongly at about 1213/cm and 1152/cm, making peak identification between about 1280/cm and 1080/cm uncertain despite subtraction of the Teflon spectrum from the sample spectrum. Subtraction of the Teflon spectrum sometimes left spurious positive or negative features near the smaller Teflon absorbances at 640/cm, 554/cm, and 517/cm but did not interfere with the identification of sulfate at about 618/cm.

Sulfate, ammonium, nitrate, silicate, carbonyl, organic nitrate, amide, and aliphatic absorbances were observed in study spectra. A carbonyl absorbance commonly peaked near 1720/cm (Figure 14 panel B). This feature is consistent with the presence of particle-phase aliphatic aldehydes, ketones, and carboxylic acids, which all absorb in the range of 1710 to 1730/cm. Frequently a "shoulder" on the peak at 1720/cm indicated the presence of a peak centered near 1740/cm, which appeared much more frequently in indoor and personal samples than in outdoor samples (see magnified insets in Figure 14 panels C and D).

Occasionally (in fewer than 10 spectra) a small carbonyl absorbance was observed close to 1800/cm (not shown). High wave number carbonyl peaks appeared almost exclusively in indoor and personal samples. This peak is usually caused by the presence of more complex compounds that contain carbonyls, such as cyclic or aromatic ketones and halogenated carbonyls (Colthup et al 1990; Socrates 1994).

Absorbances corresponding to the stretching of aliphatic hydrocarbon (CH) bonds were clearly distinguished in some spectra, and are represented by the sharp doublet of peaks near 2900/cm. Spectra in Figure 14 (panels A and B) have weak CH absorbances, and those in Figure 14 (panels C and D) have strong CH absorbances. Strong CH absorbances are rarely seen in studies of outdoor fine particles, though CH functionalities are common to organic particulate compounds (Rogge et al 1993; Allen et al 1994; Blando et al 1998; Schauer et al 1999). In this study only 3% of outdoor samples had strong CH absorbances. In contrast, 57% of indoor and 59% of personal samples had strong CH absorbances. This suggests that PM emissions in many of the homes had a strong aliphatic character. A small, more rounded peak near 3060/cm to 3070/cm, consistent with the presence of aromatic hydrocarbons, was sometimes detectable (see Figure 14 panels A and B). Aromatic hydrocarbons are more easily detected by GC–MS than by FTIR spectroscopy.

Differences Between Indoor, Outdoor, and Personal Samples In order to more quantitatively describe the differences between indoor, outdoor, and personal spectra, all study spectra were grouped into four categories delineated by combinations of strong and weak aliphatic absorbance and the presence or absence of amide. Figure 15 shows the number of spectra in each of the four categories for outdoor, indoor, and personal samples. A total of 97% of the outdoor $PM_{2.5}$ spectra had weak CH absorbance and no detectable amide absorbance. In all, 44% of homes with this typical outdoor $PM_{2.5}$ spectrum also had weak aliphatic absorbance and no detectable amide absorbance in the indoor spectra, suggesting that there were no detectable indoor sources of these functionalities in 44% of the homes. This means that at least 56% of the homes had indoor organic PM sources that substantially altered the composition of $PM_{2.5}$ exposures. Most of these indoor

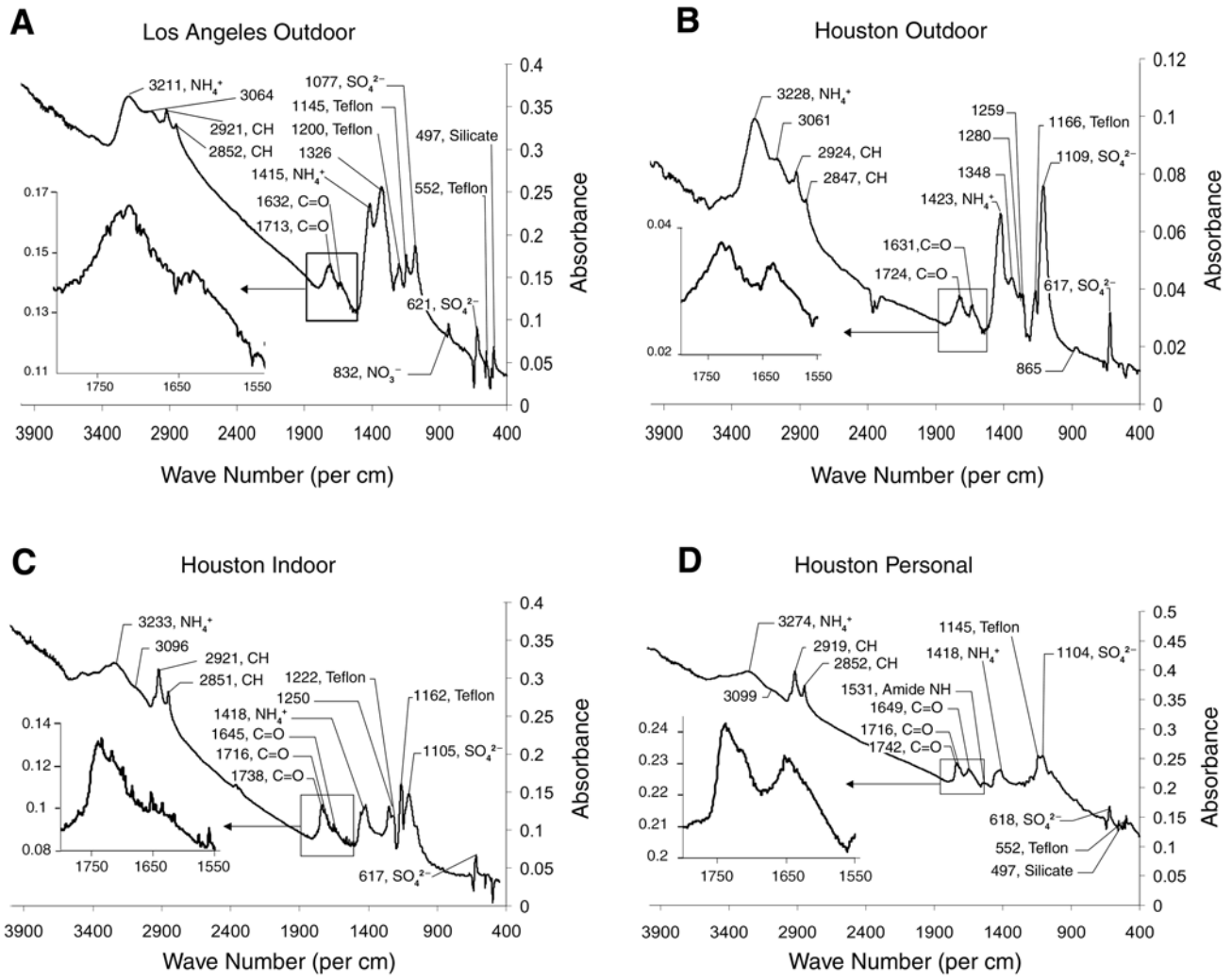


Figure 14. Typical FTIR spectra of particle samples from individual homes. Spectra provide functional group and bond information. (A) Los Angeles home 29 outdoor sample. Houston home 210 (B) outdoor sample, (C) indoor sample, and (D) personal sample. Note the different scales on the *z* axes.

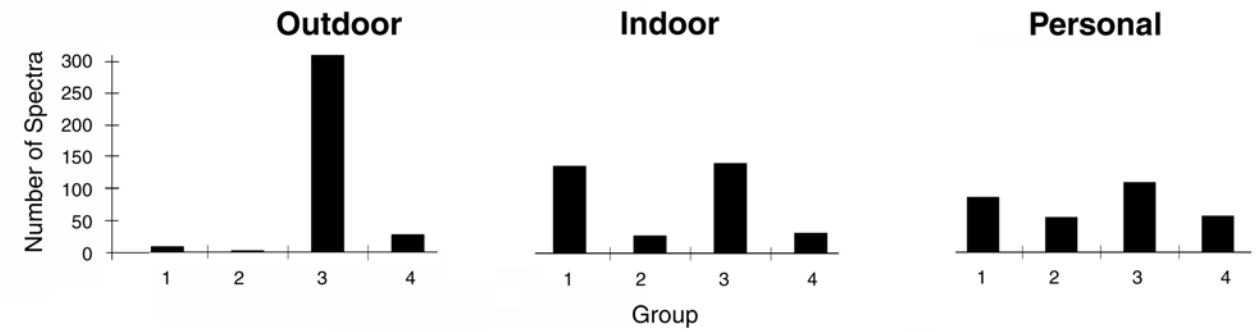


Figure 15. Number of spectra in outdoor, indoor, and personal samples in each of the four categories: (1) no amide, strong CH absorbance; (2) amide present, strong CH absorbance; (3) no amide, weak CH absorbance; (4) amide present, weak CH absorbance.

spectra differ due to the presence of strong CH absorbances. Specifically, 49% of the homes with weak aliphatic and no detectable amide absorbances in the outdoor spectrum had strong aliphatic absorbances in the indoor spectrum; 12% had detectable amide absorbances in indoor spectra.

Taken as a whole the outdoor $PM_{2.5}$ spectra in this study are similar to those observed previously (eg, Pickle et al 1990; Blando et al 1998). The indoor and personal spectra in this study suggest that indoor sources and personal exposures were frequently modified by added organic material with a strong aliphatic character. Sometimes these exposures also included amide and carbonyl absorbances that differed significantly from those in outdoor spectra (ie, 1740/cm). The spectrum of $PM_{2.5}$ of indoor origin for each home can be obtained by subtracting the outdoor spectrum from the indoor spectrum.

Figure 16 shows the difference between indoor and outdoor spectra for one Houston home, in which the outdoor spectrum from Figure 14 panel B was scaled to the 618/cm sulfate absorbance of the indoor spectrum from Figure 14 panel C. Assuming there are no indoor sources of sulfate and that penetration and loss-rate coefficients are similar for the different functional groups in the $PM_{2.5}$ spectra, this difference would represent the spectrum of indoor PM of indoor origin. Figure 17 shows the personal cloud for one Houston participant, which was constructed from the difference between the personal spectrum (Figure 14 panel D) and the indoor spectrum (Figure 14 panel C). These analyses provide evidence that the contributions of indoor sources alter not only the quantity, but also the character and properties of organic PM to which people are exposed.

Gas-Phase and Particle-Phase PAHs and Chlordanes

PAHs and chlordanes constitute only a small fraction of atmospheric $PM_{2.5}$. However, analysis of these and other selected trace organic compounds has proven useful in understanding the behavior, sources, and fate of airborne particles. Profiles of PAHs have been used with other tracers in source apportionment and to understand gas–particle partitioning of semivolatile organic compounds. Some PAHs and chlordanes are also of concern because they are mutagenic and persistent in the environment (EPA 1997b; WHO 2003). Here we report some insights obtained by analysis of indoor and outdoor samples for 30 PAHs and six chlordane compounds.

PAHs PAHs arise from a variety of combustion processes and are therefore common in both outdoor and indoor environments. The sources that contribute the largest percentage of PAHs to the atmosphere include motor vehicles, power generation via combustion of coal and oil, incineration, and burning wood (Benner et al 1995; Harrison et al 1996; Rogge et al 1997, 1998; Durlak et al 1998; Marr et al 1999; Simcik et al 1999; Dickhut et al 2000; Kavouras et al 2001). In indoor environments, PAHs are generated from cooking, smoking tobacco products, and burning natural gas, wood, candles, and incense, and are transported from the outdoors (Rogge et al 1991, 1998; Mitra and Ray 1995; Lau et al 1997; Dubowsky et al 1999; Oanh et al 1999; Li and Ro 2000; McDonald et al 2000). A number of studies have reported PAH concentrations in indoor air and attributed them to both indoor and outdoor sources (Daisey et al 1989; Chuang et al 1991; Ando et al 1996; Li and Ro 2000, Liu et al 2001). Most studies that investigated the relation

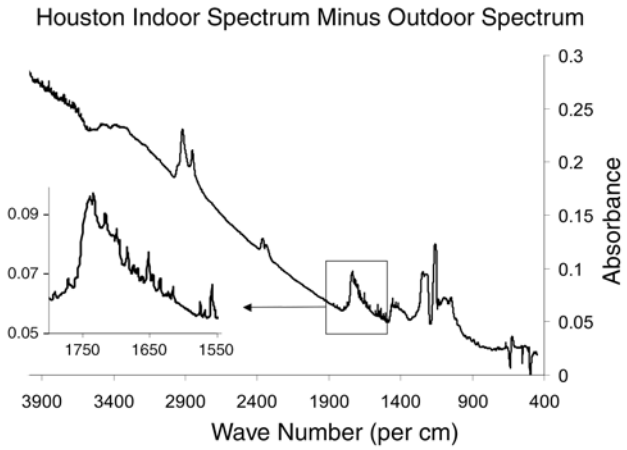


Figure 16. Spectrum of $PM_{2.5}$ of indoor origin for Houston home 210, obtained by subtracting the outdoor spectrum (Figure 14 panel B) from the indoor spectrum (Figure 14 panel C).

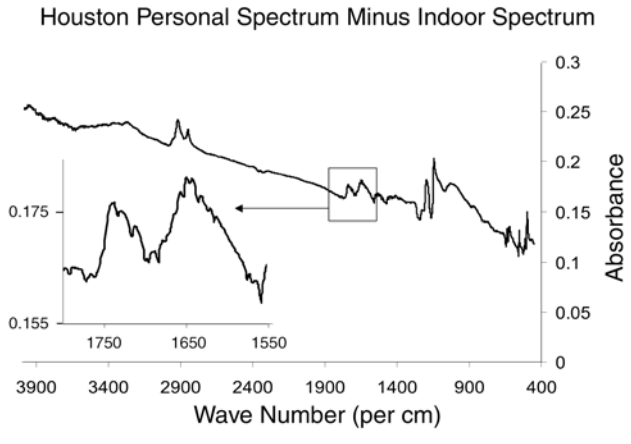


Figure 17. Spectrum of the personal cloud for a Houston participant, obtained by subtracting the indoor spectrum for home 210 (Figure 14 panel C) from the personal spectrum in home 210 (Figure 14 panel D).

between indoor PAH concentrations and outdoor pollution sources have focused on traffic-related emissions (Minoia et al 1997; Dubowsky et al 1999; Fischer et al 2000; Kingham et al 2000). For example, emissions from traffic were found to be the main outdoor source of indoor PAHs in urban, semiurban, and suburban locations around Boston, Massachusetts (Dubowsky et al 1999). Few studies have examined the indoor–outdoor relations of PAH concentrations with respect to other types of outdoor sources. A comprehensive assessment of indoor PAH concentrations in urban areas with different climates and the contribution of outdoor sources to indoor concentrations would be an important addition to the present understanding of human exposure.

The main objective of the PAH component of this study was to characterize exposure to PAHs. PAH data presented here were used to (1) assess the indoor and outdoor PAH concentrations in three geographically distinct urban areas characterized by different climates and types of dominant emission sources, (2) examine the relation between the indoor and outdoor PAH concentrations, and (3) examine indoor exposure to outdoor PAHs. Comparisons of PAH concentrations and PAH profiles were conducted on log-transformed data by ANOVA, *t* test, and the Scheffe test ($\alpha = 0.05$), as appropriate. Log-transformed distributions of data subsets used in comparisons were consistent with a normal distribution according to a Shapiro-Wilk test.

The concentrations of gas-phase and particle-phase PAHs are summarized in Figure 18. The ΣPAH concentration on the *y* axis represents the sum of the concentrations of all 30 individual PAHs. The total (gas phase + particle phase) ΣPAH concentrations in outdoor samples ranged from 1.5 to 64 ng/m^3 in Los Angeles, from 10 to 160 ng/m^3 in Houston, and from 12 to 200 ng/m^3 in Elizabeth. The variability in the PAH concentrations was substantially larger indoors than outdoors. The total (gas phase + particle phase) ΣPAH concentrations in indoor samples ranged from 7.0 to 220 ng/m^3 in Los Angeles, from 3.1 to 310 ng/m^3 in Houston, and from 19 to 350 ng/m^3 in Elizabeth. Gaseous compounds, which on average composed 90% to 97% of the total PAH mass measured in the samples, drove the variability in PAH concentrations in both the outdoor and indoor samples.

The mean ΣPAH concentration outside homes differed significantly by city. The gas-phase mean ΣPAH concentration was lowest in Los Angeles samples, whereas the lowest particle-phase ΣPAH concentrations were in Houston samples. These differences could be due to different dominant emission sources of PAHs in the cities and temperature-driven differences in gas and particle distribution. Because

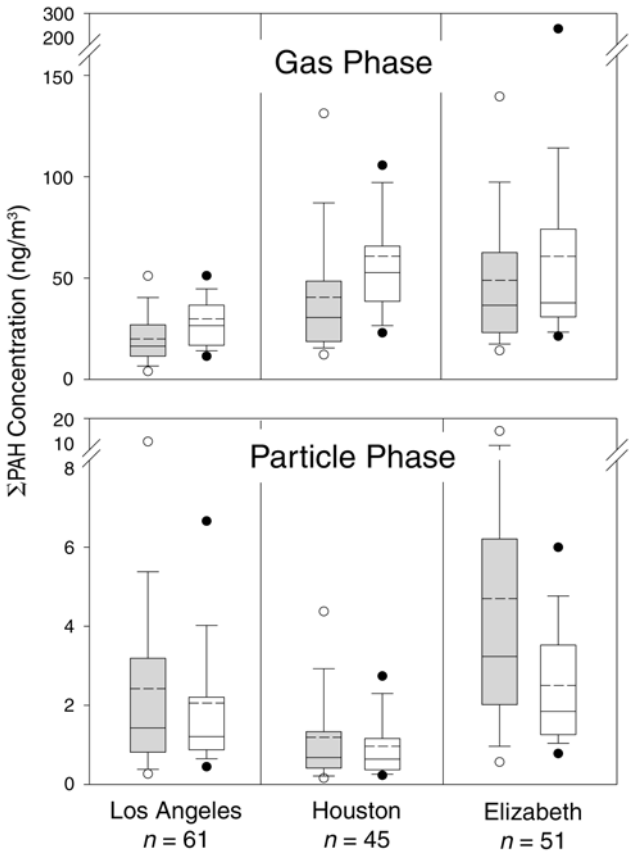


Figure 18. ΣPAH concentrations for gas phase and particle phase outdoors (gray bars) and indoors (white bars). Boxes show 25th to 75th percentiles, whiskers are 10th and 90th percentiles, and circles are 5th and 95th percentiles. Solid and dashed lines inside the boxes show the median and mean values, respectively.

of these differences, the data were further analyzed separately by city.

Figure 19 shows the relative contributions of individual PAHs to the total (gas phase + particle phase) PAH mass. Phenanthrene (Phe) was the largest contributor to the ΣPAH mass in the outdoor and indoor air in each city. This was followed by the sum of four methylated derivatives of Phe and anthracene (ANT) (2-MA, 1-MA, 1-MP, and 9-MA). The PAH profiles of low molecular weight PAHs with 3 or 4 rings (from dibenzothiophene [DBT] to benzo[*b*]naphtho[2,1-*d*]-thiophene [BNT], MW = 184 to 234) were not significantly different in the samples from the three cities.

In contrast, significant differences existed for high molecular weight PAHs with 5 to 7 rings (from BFLTs to coronene [COR], MW = 252 to 300). The percentage of PAHs with 5 to 7 rings in the ΣPAH mass was lowest in Houston samples, as seen in Figure 19. High molecular weight PAHs in the outdoor air were dominated by benzo[*g,h,i*]perylene (BghiP)

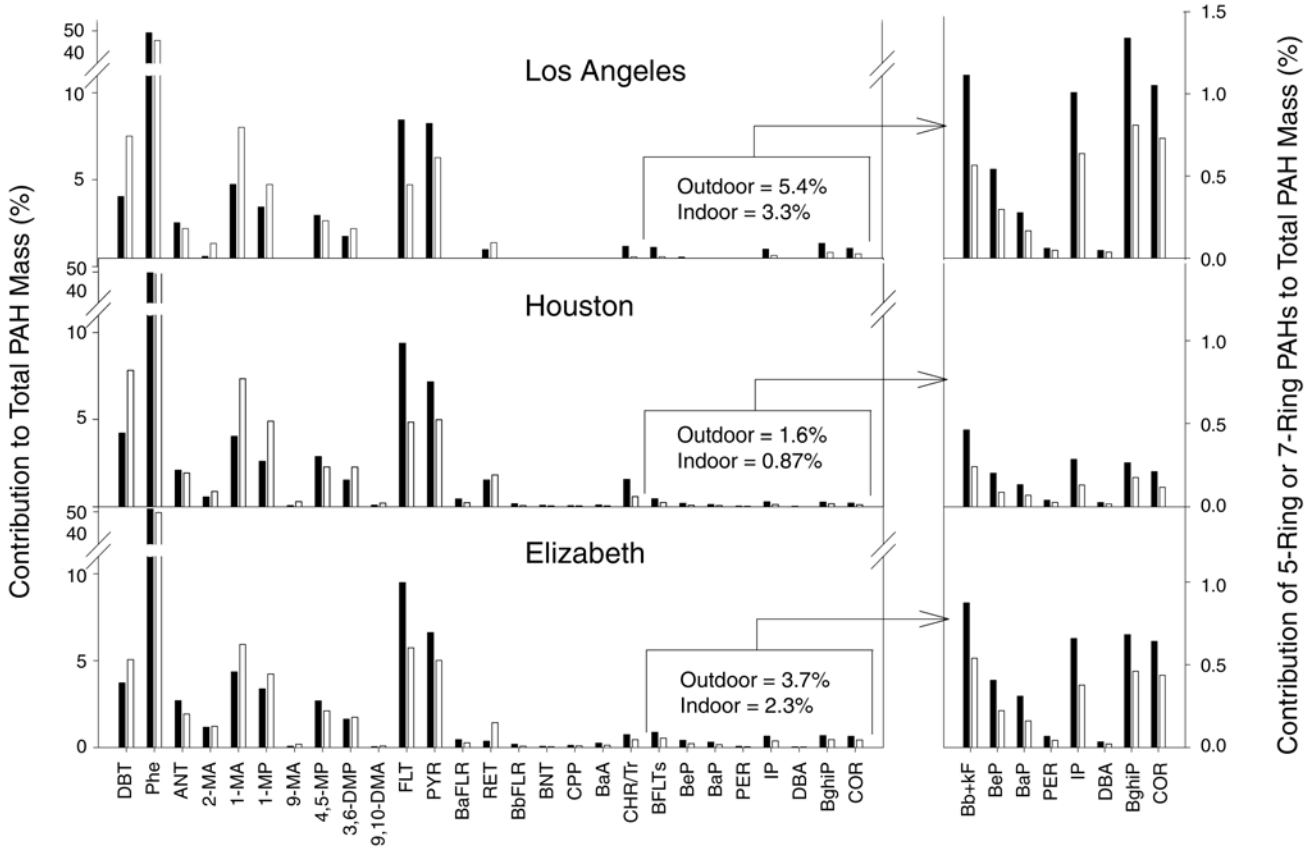


Figure 19. Geometric mean contribution of each PAH to total (gas phase + particle phase) PAH mass (%) for outdoor samples (black bars) and indoor samples (white bars). The scale for high molecular weight compounds is expanded at the right.

and COR in Los Angeles samples and by BFLTs in Houston samples; in Elizabeth outdoor air samples, contributions of indeno[1,2,3-*c,d*]pyrene (IP), BghiP, and COR were approximately equal. Significantly different outdoor air profiles of PAHs with 5 to 7 rings suggest different dominant PAH sources in the three cities. The indoor air profiles of these PAHS were similar to the outdoor profiles in each city, which suggests that outdoor sources dominated the indoor concentrations of PAHs with 5 to 7 rings.

The outdoor PAH concentrations measured in this study were compared with the ambient PAH concentrations reported for the same geographic areas in other studies (Table 12). The PAH concentrations measured in Houston during this study are in good agreement with those measured previously in Seabrook, Texas (Park et al 2001), which is about 40 km southeast of Houston. The PAH concentrations measured in Elizabeth during this study are comparable with those measured previously in Jersey City, New Jersey (Eisenreich et al 2001), which is about 10 km northeast of Elizabeth. The PAH concentrations measured in Los Angeles during this study are consistent with those measured in 1993 (Fraser et al 1998) with the exception of Phe concentrations, which were lower in this study.

As seen in Table 12, the coupled indoor and outdoor concentrations of low molecular weight PAHs measured in this study are comparable with concentrations in other studies in Columbus, Ohio (Mitra and Ray 1995) and Taipei, Taiwan (Li and Ro 2000), and considerably lower than concentrations in Hangzhou, China (Liu et al 2001). The indoor and outdoor concentrations of high molecular weight PAHs, associated predominantly with the particle phase, are similar to PAH concentrations measured in Huddersfield, England (Kingham et al 2000), and lower, on average, than concentrations in Columbus, Ohio (Mitra and Ray 1995), Amsterdam, The Netherlands (Fischer et al 2000), Pavia, Italy (Minoia et al 1997), and Taipei, Taiwan (Li and Ro 2000).

Table 12. The Concentrations of Selected PAHs (Gas Phase + Particle Phase) in Indoor and Outdoor Air: Comparison with Other Studies

		Outdoor Concentration (ng/m^3)				
Site Location	*n*	Phe	PYR	BFLTs	BghiP	Reference
Los Angeles[a]	61	9.1 (1.1–33)	1.5 (0.048–5.9)	0.19 (0.014–1.8)	0.24 (0.0054–3.1)	This study
Houston[a]	45	18 (1.7–97)	2.1 (0.87–15)	0.13 (0.013–1.3)	0.076 (0.0018–1.9)	This study
Elizabeth[a]	51	23 (2.9–140)	2.9 (0.53–21)	0.38 (0.013–1.9)	0.30 (0.0018–2.4)	This study
Los Angeles[b]	NA[c]	50 (3.6–140)	7.2 (0.65–26)	0.22 (0.00–1.1)[d]	0.77 (0.03–4.2)	Fraser et al (1998)
Seabrook TX[b]	NA	12 (0.65–58)[e]	3.3 (0.084–19)[e]	0.14 (0.015–0.63)[f]	0.058 (0.007–0.30)[f]	Park et al (2001)
Jersey City NJ[b]	58	15 (3.4–34)[e]	2.1 (0.16–4.3)[e]	0.55 (0.0052–3.1)[f]	0.37 (0.0052–2.1)[f]	Eisenreich et al (2001)

		Outdoor Concentration (ng/m^3)		Indoor Concentration (ng/m^3)		
		Phe	BaP	PHE	BaP	
Los Angeles[a]	19	9.1 (1.1–33)	0.049 (0.0023–1.0)	12 (5.0–180)	0.045 (0.0040–0.57)	This study
Houston[a]	21	18 (1.7–97)	0.037 (0.0026–0.48)	27 (11–240)	0.028 (0.0036–1.1)	This study
Elizabeth[a]	15	23 (2.9–140)	0.13 (0.0030–0.89)	27 (4.4–330)	0.081 (0.0064–0.63)	This study
Columbus OH[g]	8	30	0.27	84.5	0.44	Mitra and Ray (1995)
Huddersfield England[h]	13	NA	0.035 ± 0.053	NA	0.0090 ± 0.015	Kingham et al (2000)
Amsterdam, The Netherlands[b]	18	NA	0.82 (0.25–2.3)	NA	0.49 (0.15–1.1)	Fischer et al (2000)
Pavia, Italy[b]	8	NA	1.19 (0.68–2.85)	NA	0.11 (< MDL–0.21)	Minoia et al (1977)
Taipei, Taiwan[i]	14	21 ± 4.9	1.7 ± 2.2	20 ± 4.6	1.7 ± 2.4	Li and Ro (2000)
Hangzhou, China[h]	8	660 ± 600	NA	530 ± 460	NA	Liu et al (2001)

[a] Geometric mean (range).

[b] Arithmetic mean (range).

[c] NA = data not available.

[d] Only concentration of benzo[*k*]fluoranthene reported.

[e] Concentration range for gas phase only.

[f] Concentration range for particle phase only.

[g] Arithmetic mean.

[h] Arithmetic mean ± SD.

[i] Geometric mean ± geometric SD.

The indoor–outdoor (I/O) ratios of total (gas phase + particle phase) PAH concentrations measured in this study are presented in Figure 20. The reference line represents I/O equal to 1. I/O values > 1 suggest the presence of indoor sources. I/O values < 1 can occur in the absence of indoor sources or in the presence of indoor sources if the penetration of PAHs through the building envelope is < 1, or if the loss rate indoors is significantly > 0, which are both likely to be true for particle-phase PAHs.

In general, the I/O values were higher for low molecular weight PAHs that are present predominantly in the gas phase. (This is consistent with findings in other studies [Mitra and Ray 1995; Li and Ro 2000]). Median indoor concentrations of 3-ring Phe and ANT and their alkylated derivatives, 3-ring DBT, and 4-ring 4,5-methylenephenanthrene [4,5-MP]) (MW = 178 to 206 and 234) exceeded median outdoor concentrations. For example, in 120 of 157 homes the I/O for 1-MA was > 1, and in 81 homes the I/O was > 2. This suggests that indoor sources were important contributors to low molecular weight PAH concentrations.

Considerably lower I/O values were observed for PAHs with 4 rings: fluoranthene (FLT), pyrene (PYR), benzo[*a*]fluorene (BaFLR) and benzo[*b*]fluorene (BbFLR), benzo[*a*]anthracene (BaA), CHR/Tr, and BNT, and for 5-ring cyclopenta[*c,d*]pyrene (CPP) (MW = 202 to 234), which are distributed between the gas and particle phases. The median I/O values of these PAHs were < 1. The I/O for PYR, for example, was ≤ 1 in 84 of 157 homes. However, for several

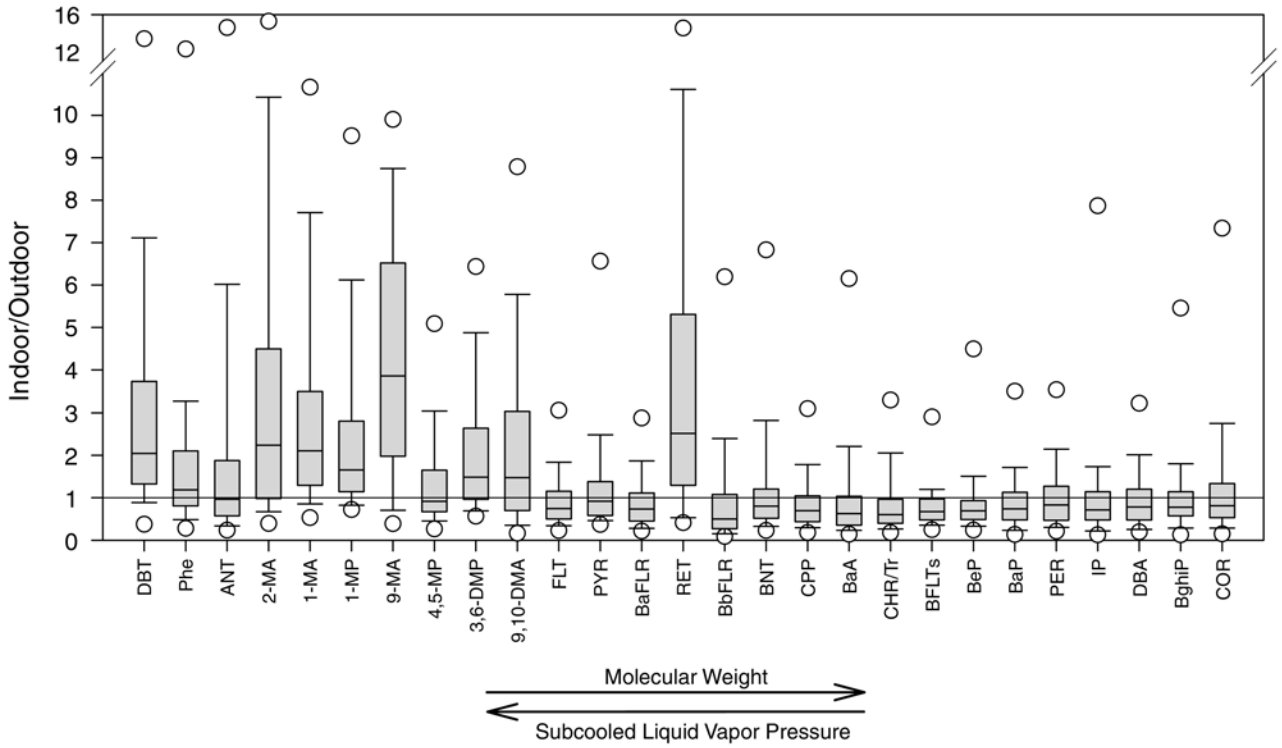


Figure 20. Indoor–outdoor ratios of measured PAH concentrations (gas phase + particle phase). Boxes show 25th to 75th percentiles, whiskers are 10th and 90th percentiles, and circles are 5th and 95th percentiles. Solid and dashed lines inside the boxes show the median and mean values, respectively. Reference line shows I/O = 1.

PAHs with 4 to 5 rings, 95% of the I/O values were > 3, verifying that indoor sources of 4-ring PAHs were present in at least some homes.

The lowest I/O values were observed for high molecular weight PAHs with 5 to 7 rings (MW = 252 to 300), which are associated predominantly with the particle phase: BFLTs, benzo[*e*]pyrene (BeP), benzo[*a*]pyrene (BaP), perylene (PER), IP, DBA, BghiP, and COR. In most cases, indoor concentrations of these PAHs were lower than outdoor concentrations. As an example, the I/O for BghiP was ≤ 1 in 92 of 157 homes. The median I/O was < 1 for these compounds.

These observations suggest that in most homes outdoor sources could be the dominant cause of the high molecular weight PAHs, found mostly in the particle phase. This is supported by the strong within-home correlations between the concurrently measured indoor and outdoor concentrations of the eight PAHs with 5 to 7 rings. Within-home correlations of the indoor and outdoor concentrations across all measured PAH species were significant ($P < 0.05$) for 134 (85%) of 157 homes; in 42% of homes r^2 values between indoor and outdoor PAH concentrations were greater than 0.90.

Figure 21 shows the regression of the indoor concentration on the outdoor concentration across homes for low molecular weight Phe and high molecular weight BghiP. Low correlations and large intercepts were observed for indoor and outdoor concentrations of Phe; the correlation was not significant for Houston homes. The large degree of data scatter and large number of values above the 1:1 line suggest that indoor sources of Phe were important and variable. In contrast, strong correlations were observed between the indoor and outdoor concentrations of the high molecular weight compound BghiP. The strong correlations and low intercepts suggest that outdoor sources were substantially more important for this high molecular weight compound.

PAH Gas–Particle Partitioning Semivolatile compounds like PAHs partition between the gas and particle phases according to their vapor pressure and concentration, atmospheric temperature, and the concentration and properties of the PM. Partitioning occurs through both adsorption on the surface of particles and absorption into suitable particulate material (Pankow 1987, 1994). Gas–particle partitioning of PAHs has been extensively studied in different outdoor urban, remote, and coastal environments because the gas–particle phase distribution has a dramatic impact on the atmospheric lifetime and fate of PAHs (Ligocki and Pankow 1989; Foreman and Bidleman 1990; Cotham and Bidleman 1995; Harner and Bidleman 1998; Simcik et al

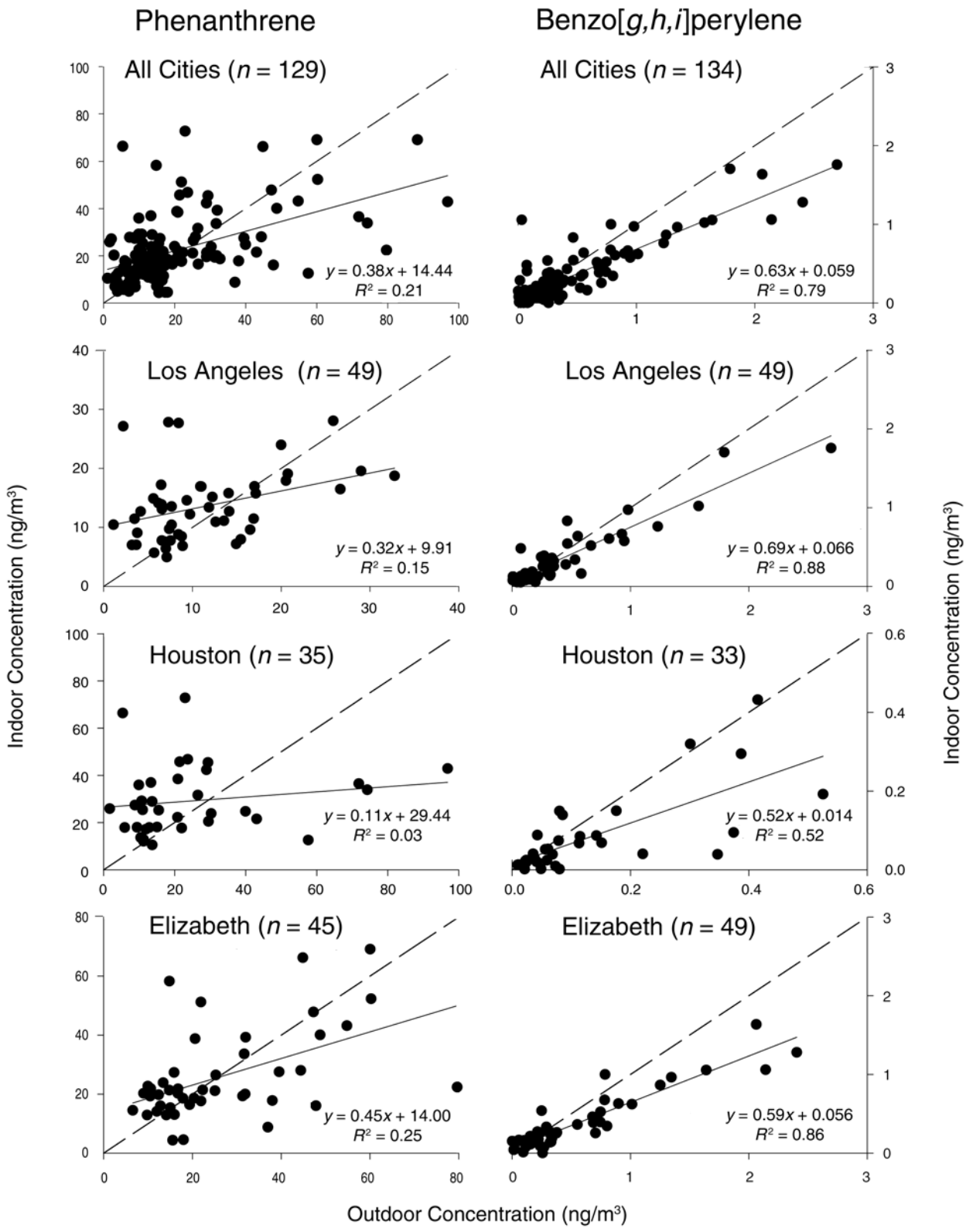


Figure 21. Indoor and outdoor concentrations of phenanthrene (low molecular weight) and benzo[*g,h,i*]perylene (high molecular weight), regression equations, and coefficients of determination (r^2) for all homes and for Los Angeles, Houston, and Elizabeth homes. Line is 1:1; *n* is the number of homes.

1998; Offenberg and Baker 2002). The efficiency and location of semivolatile organic compound deposition in the respiratory tract is also strongly dependent on gas–particle partitioning (Pankow 2001). Most of the research that addresses the partitioning of semivolatile organic compounds in indoor air focuses on interaction of the compounds with indoor surfaces (eg, van Loy et al 2000).

Outdoor-to-indoor transport of PAHs is often accompanied by changes in air temperature, the introduction of freshly emitted PM, and possibly the introduction of PAHs emitted from indoor sources (Conner et al 2001). These changes will drive the redistribution of transported PAHs between the gas and particle phases as a new equilibrium is established. A better understanding of these effects and the underlying mechanisms driving partitioning will improve estimates of PAH contributions from outdoor sources and the understanding of PAH partitioning and persistence indoors. The data from the paired indoor–outdoor air samples collected during this study provided a unique opportunity to examine changes in gas–particle partitioning of PAHs between indoor and outdoor environments. Specific objectives of this work were to compare gas–particle partitioning of PAHs in different atmospheric environments, to examine the effect of changes in temperature and $PM_{2.5}$ composition on PAH partitioning, and to look for insights into the mechanisms driving partitioning of PAHs in outdoor and indoor air.

This analysis examines a subset of gas-phase and particle-phase PAH concentrations measured in the indoor and outdoor air of 76 study homes (28 in Los Angeles, 28 in Houston, and 20 in Elizabeth; total of 152 samples). The gas-phase concentrations of the five PAHs with 6 to 7 aromatic rings (MW = 276 to 300, log of subcooled liquid vapor pressure [$\log p_L^\circ = -8.23$ to -5.62) were below the MDLs in 93% of the measurements. Therefore, gas–particle partitioning was examined for 20 PAHs with 3 to 5 rings (MW = 178 to 252, $\log p_L^\circ = -6.54$ to -0.80): Phe, ANT, 1-methylphenanthrene (1-MP), 1-MA and 2-methylanthracene (2-MA), 4,5-MP, 3,6-dimethylphenanthrene (3,6-DMP), FLT, PYR, BaFLR and BbFLR, retene (RET), BaA, CHR/Tr, BFLTs, BeP, BaP, and PER. For each PAH, p_L° was derived from Offenberg and Baker (1999). Statistical analyses were performed using SPSS 10.0 software.

For the homes examined in this study, 48-hour average temperatures in the outdoor air ranged from 11°C to 25°C in Los Angeles, from 9.4°C to 30°C in Houston, and from 1.7°C to 30°C in Elizabeth. In the indoor air, average temperatures ranged from 17°C to 29°C across the three cities. Temperature variability within the 48-hour sampling periods was about 10°C for the outdoor samples and 6°C for the indoor samples. The fraction of PAHs associated with $PM_{2.5}$ ($\phi_{2.5}$) was defined as the quantity collected on the filter divided by the quantity collected on filter and adsorbent. It ranged from 0.00033 to 0.022 for Phe (MW = 178, $\log p_L^\circ = -2.16$ to -0.80) to 0.85 to 1.0 for COR (MW = 300, $\log p_L^\circ = -8.23$ to -6.19).

The partitioning of PAHs between the gas and particle phases was parameterized using the gas–particle partition coefficient K_p ($m^3/\mu g$; Yamasaki et al 1982; Pankow 1987), defined as follows:

$$K_p = \frac{F_{2.5}/PM_{2.5}}{A}, \quad (3)$$

where $F_{2.5}$ and A (ng/m^3) are the PAH concentrations on the $PM_{2.5}$ QFF (particle phase) and on the adsorbent (gas phase), respectively. $PM_{2.5}$ ($\mu g/m^3$) is the $PM_{2.5}$ mass concentration. The propagated precision (ie, from random errors) in the gas–particle partition coefficients (K_p) ranged from 31% to 48% for all PAHs except CHR/Tr, BFLTs, and BaP; for these three compounds the uncertainty was 62% to 71% (Naumova et al 2003). The higher uncertainties for the latter PAHs are associated with greater uncertainties in the gas-phase measurements. Systematic errors in partition coefficients were dominated by sampling artifacts that occur when the sampled air is often not in equilibrium with the collection substrate. Naumova and associates (2003) examined these errors in detail, including calculating adsorption (positive) artifacts using the method of Mader and Pankow (2001).

Regardless of whether PAHs partition primarily by adsorption on the particle surface or by absorption into the organic PM, the partition coefficients of homologue compounds tend to be inversely proportional to the subcooled liquid vapor pressure of the compounds (Yamasaki et al 1982; Ligocki and Pankow 1989; Foreman and Bidleman 1990; Cotham and Bidleman 1995; Harner and Bidleman 1998; Simcik et al 1998; Offenberg and Baker 2002):

$$\log K_p = m \log p_L^\circ + b, \quad (4)$$

where m and b are, respectively, the slope and intercept of the linear regression.

Linear regressions of the log of the measured gas–particle partition coefficient ($\log K_{p,meas}$) on $\log p_L^\circ$ yielded significant (95% confidence) slopes and intercepts for all samples, with r^2 of 0.90 ± 0.060. Linear regression plots of $\log K_{p,meas}$ on $\log p_L^\circ$ for the individual samples (n = 1847) are presented in Figure 22. The slopes, m, ranged from -1.19 to -0.445; the intercepts, b, ranged from -6.22 to -3.38. Regression statistics by city and indoor or outdoor category are summarized in Table 13.

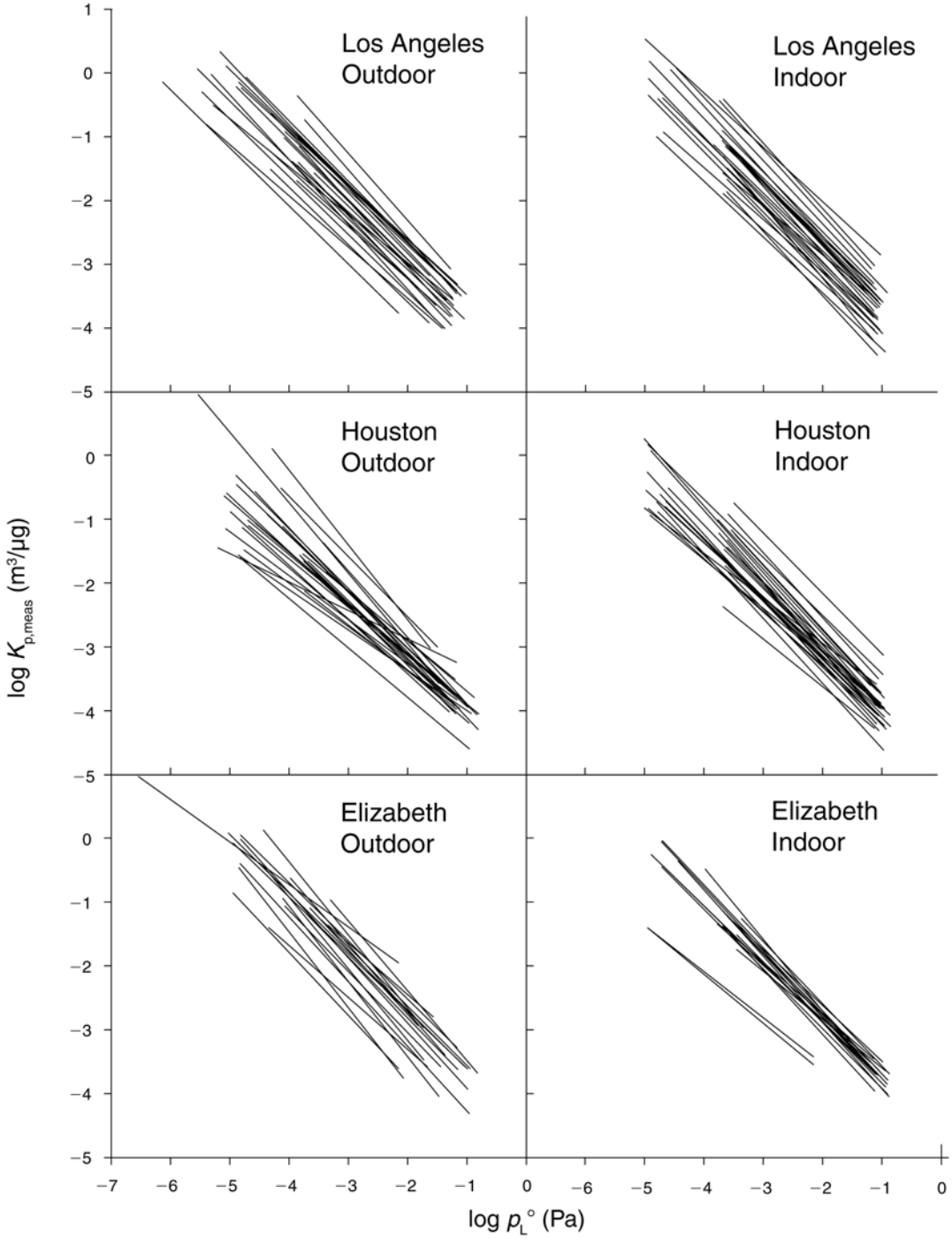


Figure 22. Linear regressions of log $K_{p,meas}$ on log p_L° for each individual sample. All measured PAHs are regressed; underlying data are not shown.

Table 13. Summary Statistics of Linear Least Squares Regression[a] of Log $K_{p,meas}$ on Log p_L° for PAHs from Each Home Individually

		Slope				Intercept			
Category	*n*	Mean	SD	Minimum	Maximum	Mean	SD	Minimum	Maximum
Los Angeles									
Outdoor	28	− 0.913	0.0740	− 1.06	− 0.753	− 4.80	0.367	− 5.73	− 4.30
Indoor	28	− 0.947	0.0778	− 1.14	− 0.763	− 4.70	0.394	− 5.51	− 3.72
Houston									
Outdoor	28	− 0.847	0.148	− 1.17	− 0.445	− 4.82	0.312	− 5.34	− 3.76
Indoor	28	− 0.936	0.0961	− 1.08	− 0.706	− 4.90	0.324	− 5.60	− 4.05
Elizabeth									
Outdoor	20	− 0.977	0.134	− 1.19	− 0.664	− 4.89	0.635	− 6.22	− 3.38
Indoor	20	− 0.922	0.102	− 1.12	− 0.723	− 4.73	0.234	− 5.20	− 4.36

[a] Log $K_{p,\,meas}$ = $\boldsymbol{m}$ log p_L° + b, for individual samples. $\boldsymbol{m}$ is slope; b is intercept; n is number of measurements.

The slopes and intercepts for individual samples determined in this study were comparable to the slopes and intercepts for PAHs reported for other urban areas: Portland, Oregon (m = −0.88, b = −5.38; Ligocki and Pankow 1989); Denver, Colorado (m = −0.760, b = −5.10; Foreman and Bidleman 1990); Chicago, Illinois (m = −0.690 and −0.638, b = −4.61 and −3.47; Cotham and Bidleman 1995; Simcik et al 1998); and Manchester, England (m = −0.688, b = −5.13; Lohmann et al 2000). Strong linear relations between log $K_{p,meas}$ and log p_L° suggest that PAHs in the outdoor air were close to equilibrium and PAHs transported from outdoor to indoor air rapidly approached new equilibrium with indoor emissions and conditions. This is consistent with the conclusion that PAHs rapidly equilibrate between the gas and particle phases stated by Kamens and colleagues (Kamens et al 1995; Kamens and Coe 1997).

Variations of the regression parameters (m and b) in the outdoor samples (Table 13, Figure 22) indicated that differences in gas–particle partitioning of PAHs within each city were on the same order as the differences between the cities. In the indoor samples variability among these parameters was about as high as variability in the outdoor samples. This justified regressing all data together, as shown in Figure 23. The resulting single-parameter linear regression (SLR),

$$\log K_{p,meas} = -0.860 \log p_L^\circ - 4.67 \quad (r^2 = 0.775), \qquad (5)$$

had a 95% prediction interval for log $K_{p,meas}$ of about 2 log units. The 2 orders of magnitude span of partition coefficients at any given value of vapor pressure is greater than the estimated systematic and random errors and could be related to differences in aerosol surface area of the particles

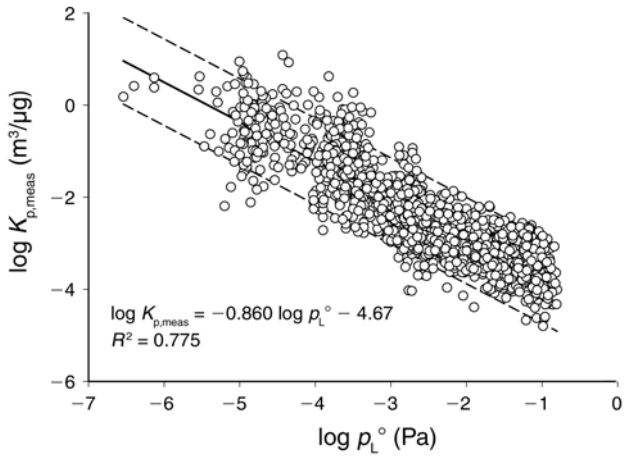


Figure 23. Log $K_{p,meas}$ and log p_L° from all PAH measurements from all homes. Solid line is the least squares regression. Dashed lines define the 95% CI. Also provided is the regression equation and coefficient of determination (r^2). n = 1808.

in case of *adsorption,* organic matter content in case of *absorption* (Pankow 1994), and sampling temperature (Pankow and Bidleman 1991), assuming that the PAHs are at equilibrium.

The complex effect of temperature and aerosol composition on gas–particle partitioning of PAHs was examined by stepwise multiple linear regression (MLR) on the pooled data set. The purpose of the MLR analysis was to establish an empirical relation that would allow for more accurate prediction of partition coefficients for PAHs in indoor and outdoor air when temperature and aerosol characteristics are known. Offenberg and Baker (2002) have used MLR for exploring gas–particle partitioning of PAHs on size-segregated aerosol. Here, regression analysis was performed in the form

$$\log K_{p,meas,SD} = I + A \log p_L^\circ(25°C) + B\,T + C\,f_{EC} + D\,f_{OC}, \quad (6)$$

where A, B, C, D, and I are fit parameters, $p_L^\circ(25°C)$ is the subcooled liquid vapor pressure of the compound at 25°C, T is ambient indoor or outdoor temperature, and f_{EC} and f_{OC} are the fractions of EC and OC in $PM_{2.5}$ mass, respectively. The form of equation 6 was guided by the absorption theory of Pankow and Bidleman (1991), which delineates the dependence of partitioning on p_L° and T, and predicts that PAHs absorb into organic PM. The EC term was included because Dachs and Eisenreich (2000) concluded, in a regional atmospheric study, that the EC concentration was a better predictor of PAH partitioning than the OC concentration.

Separation of $\log p_L^\circ$ into two variables, $\log p_L^\circ(25°C)$ and T (average over the sampling period), was necessary to consider the separate effect of a change in temperature on partitioning during the outdoor-to-indoor transport; that is, temperature affects the partitioning directly and also indirectly by changing compound vapor pressures. To maintain standard conditions, the measured partition coefficients were converted to standard temperature (25°C) in micrograms per cubic meter using the ideal gas law. The fraction of OC, f_{OC}, was used rather than the more commonly used fraction of organic matter (f_{OM}) in order to avoid introducing uncertainties in converting from OC to OM (Turpin and Lim 2001). Neither OC nor PAH concentrations were corrected for adsorption artifacts, in keeping with the body of literature in the PAH partitioning field. However, the resulting uncertainties in K_p have been assessed (Naumova et al 2003). The analysis of OC sampling artifacts in the current study indicated that the percentage corrections for the gas adsorption artifact for indoor and outdoor OC samples were similar. Therefore, measured OC was a reasonable surrogate for particulate OC in the MLR.

The pooled data set for the MLR analysis included 1808 measurements of PAH partition coefficients, for which the corresponding measurements of T, f_{OC}, and f_{EC} were available. No two independent variables were highly correlated. The result of the MLR analysis was the estimated regression hyperplane

$$\log K_{p,meas,SD} = 8.398 - 0.888 \log p_L^\circ(25°C) - 0.0456T + 3.686 f_{EC} + 0.469 f_{OC} \qquad \left(R^2 = 0.845\right). \quad (7)$$

All partial regression coefficients were significantly different from zero. The summary statistics for the MLR given by equation 7 are presented in Table 14, which also includes the results of the SLR given by equation 5 for comparison. Because of the complexity of graphical presentation of the five-dimensional plot, the regression of equation 7 is illustrated by partial regression plots, shown in Figure 24. Each partial plot shows the relation between one independent variable and $\log K_{p,meas,SD}$ by means of the scatter plot of the residuals (actual values minus predicted values) of these two variables. Each partial plot is characterized by the partial R^2, which shows the net association between an independent variable (designated as a subscript on R^2) and a dependent variable as determined by MLR. Note that partial R^2 in MLR is different from R^2 in SLR because it estimates the effect of independent variable, X_1, on dependent variable, Y, through the relation of X_1 with other independent variables (X_2, X_3, etc).

Table 14. Multiple Linear Regression of Log $K_{p,meas,SD}$ on Log p_L°, T, f_{OC}, or f_{EC} for All PAH Measurements and Single-Parameter Regression Results[a]

Dependent Variable	Independent Variable	Coefficient	Coefficient Estimate	SD	*P* Value	95% CI	Total R^2	Partial R^2
MLR Model	n = 1808							
$\log K_{p,meas,SD}$	Constant	I	8.398	0.604	< 0.001	(7.213 to 9.582)	0.845	
	$\log p_L^o(25°C)$	A	− 0.888	0.009	< 0.001	(− 0.907 to − 0.870))		0.837
	T	B	− 0.0456	0.002	< 0.001	(− 0.050 to − 0.042)		0.212
	f_{EC}	C	0.469	0.055	< 0.001	(0.360 to 0.577)		0.117
	f_{OC}	D	3.686	0.238	< 0.001	(3.219 to 4.153)		0.038
SLR Model	n = 1847							
$\log K_{p,meas}$	Constant	b	− 4.671	0.028	< 0.001	(− 4.727 to − 4.616)	0.775	
	$\log p_L^o$	m	− 0.860	0.011	< 0.001	(− 0.881 to − 0.839)		—

[a] n is number of measurements.

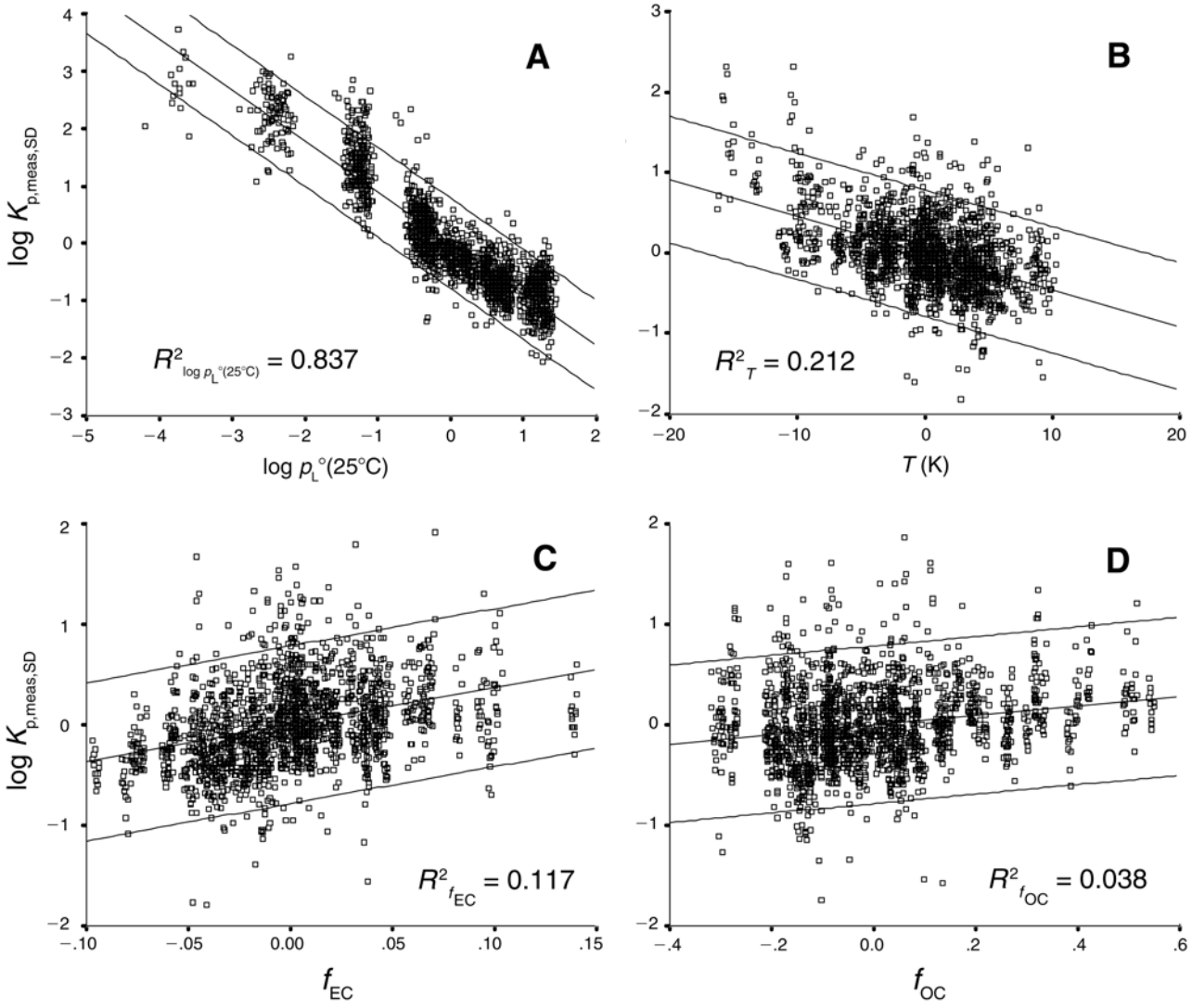


Figure 24. Partial regression plots illustrating MLR results of equation 6. Each partial plot shows the relation between log $K_{p,meas,SD}$ and log $p_L°$ (panel A), T (panel B), f_{EC} (panel C), or f_{OC} (panel D), by means of a scatter plot of residuals. R^2 values are the partial coefficients of determination. Outer rules define the 95% CI.

Expanding the regression of log $K_{p,meas}$ versus log $p_L°$ to include T, f_{OC}, and f_{EC} as independent variables significantly improved the prediction of log $K_{p,meas}$. In the SLR, 78% of variability in log $K_{p,meas}$ was explained by the estimated regression line. Regression on four predictors increased the explained variance in log $K_{p,meas,SD}$ to 85%. The SE of the estimate for the partition coefficient decreased from 0.470 in the SLR to 0.400 in the MLR.

The improved predictive capability of the MLR results can be seen by comparing calculated and measured partition coefficients. For example, the measured log $K_{p,meas}$ for FLT in the indoor air of a Los Angeles residence was −3.60 on July 13 to 14, 1999. The SLR model that used $p_L°$(25°C) as the single predictor yielded a value of −2.93 for log K_p; the 95% CI was −3.85 to −2.00 (1.85 log units). The MLR model predicted a partition coefficient of −3.19 using measured $p_L°$(25°C), T, f_{EC}, and f_{OC}, with a 95% prediction interval of −3.98 to −2.41 (1.54 log units).

The direction of the effect of each predictor on the partition coefficients is seen in Figure 24. Log $p_L°$(25°C) and T exhibit negative effects on $K_{p,meas,SD}$; that is, an increase in either variable leads to a decrease in the thermodynamic partition coefficient and the fraction in the particle phase. In contrast, an increase in variables f_{EC} and f_{OC} tends to produce an increase in K_p (ie, partitioning to the particle phase). The latter is logical because an increase in carbon

content of the aerosol provides more media to which PAH molecules can sorb. The relative importance of each predictor is shown not only by the amount of explained variance but also by the absolute change in the partition coefficients due to environmentally relevant changes in this variable. For example, an increase in f_{EC} by 0.01 and an increase in f_{OC} by 0.1 have about the same effect on log $K_{p,meas,SD}$ as a decrease in temperature by 1 K.

As before, compound log $p_L°$(25°C) was the most important predictor (Figure 24, panel A), accounting for an 84% reduction in the unexplained variance in log $K_{p,meas,SD}$ when T, f_{OC}, and f_{EC} were held constant. A unit increase in log $p_L°$(25°C) resulted in a decrease in the partition coefficient by 0.888 log units. Note that this represents the change in the partition coefficient with respect to change in log $p_L°$ from compound to compound, neglecting changes in $p_L°$ due to changes in temperature.

Temperature was the second most important predictor of the partition coefficient (Figure 24, panel B). The variation in temperature explained 21% of the variance of log $K_{p,meas,SD}$. According to the regression, a 1-K increase in temperature will result in a decrease in log $K_{p,meas,SD}$ by 0.0456 log units when all other parameters are held constant. A practical illustration of the effect of temperature is the outdoor-to-indoor transport of PAHs when, for example, the outdoor temperature is 0°C and the indoor temperature is 20°C. If f_{OC} and f_{EC} remain constant, then log $K_{p,meas,SD}$ for each PAH will decrease by 0.912 log units as the PAH is transported indoors. For example, given a partition coefficient for BaA in the outdoor air of 0.80 $m^3/\mu g$, in the indoor air it would become 0.091 $m^3/\mu g$ owing to the change in temperature only. Assuming further that the $PM_{2.5}$ concentration was 20 $\mu g/m^3$ in both indoor and outdoor air, the fraction of BaA in the particle phase, $\phi_{2.5}$, would decrease from 0.94 in the outdoor air (0°C) to 0.64 in the indoor air (20°C).

The slopes 3.686 and 0.469 (Figure 24, panels C and D, respectively) denote the increase in log $K_{p,meas,SD}$ for each additional increase in f_{EC} and f_{OC}, respectively, to $PM_{2.5}$ mass. The f_{EC} is a more significant predictor of the partition coefficient than the f_{OC}. Variations in f_{EC} explained 12% of the variance of log $K_{p,meas,SD}$ that was unexplained by other predictors, whereas variations in f_{OC} explained only 4%.

This finding is in qualitative agreement with that of Dachs and Eisenreich (2000). Because EC is highly correlated with (and is a good tracer of) primary combustion-generated OC, this result suggests that PAHs more readily sorb to primary combustion-generated aerosol (OC or EC) than to other types of OC. This conclusion is logical for both indoor and outdoor environments. Secondary organic PM, which is fairly polar, is unlikely to be a good substrate for absorption of nonpolar PAHs. Indoor aerosol is often more enriched in OC than outdoor aerosol, whereas EC concentrations are usually similar or slightly lower indoors than outdoors. This suggests that the additional OC indoors is dominated by noncombustion primary sources such as cooking at moderate temperatures or by secondary formation or both. The results of the MLR suggest that the indoor-generated OC is a less favorable substrate for PAH sorption than combustion aerosol. Enrichment of the indoor aerosol in noncombustion OC appears to increase partitioning of PAHs to the particle phase, but to a smaller degree than it would if the added OC was generated directly by combustion.

Environmental conditions important for partitioning (T, f_{EC}, and f_{OC}) often change drastically during the outdoor-to-indoor transport of air, causing PAHs transported indoors to reequilibrate between the gas and particle phases. Application of the MLR model to the paired indoor–outdoor samples helps to explain the processes by which PAHs approach equilibrium in the indoor air.

Figure 25 shows the measured outdoor and indoor fractions of $PM_{2.5}$ mass in the particle phase ($\phi_{2.5}$) for selected PAHs in the paired indoor–outdoor samples collected in a Los Angeles home December 13 to 14, 1999. It also shows the predicted indoor $\phi_{2.5}$ that would result if all indoor PAHs originated outdoors and the partitioning were altered by the measured outdoor-to-indoor change in T, f_{EC}, and f_{OC} independently and together. The largest effect on the partitioning of BaA, for example, was the decrease in $\phi_{2.5}$ from 0.70 (measured outdoor fraction) to 0.42 (modeled indoor fraction) caused only by the 11.3°C increase in temperature indoors. The increase in f_{EC} by 0.068 and in f_{OC} by 0.40, when considered alone, led to increases in $\phi_{2.5}$ of 0.11 and 0.082, respectively. The net effect of BaA being transported from outdoors to indoors at that home was a 4% decrease of the fraction of BaA in the particle phase. The modeled indoor gas–particle partitioning was in good agreement with the measured indoor gas–particle partitioning for all PAHs at this Los Angeles home. This example illustrates the importance of considering the aerosol characteristics in comparing gas–particle partitioning of PAHs in different atmospheric environments and is a reminder that chemical thermodynamics can alter the relation (in terms of mass, composition, and properties) between measurements of $PM_{2.5}$ taken at central-site monitors and exposure to $PM_{2.5}$ of outdoor origin.

The MLR results depend on the assumption that the overall random errors in $p_L°$, T, f_{EC}, and f_{OC} are roughly equal. Our current understanding of these errors suggests this is a good assumption. Systematic bias, for example

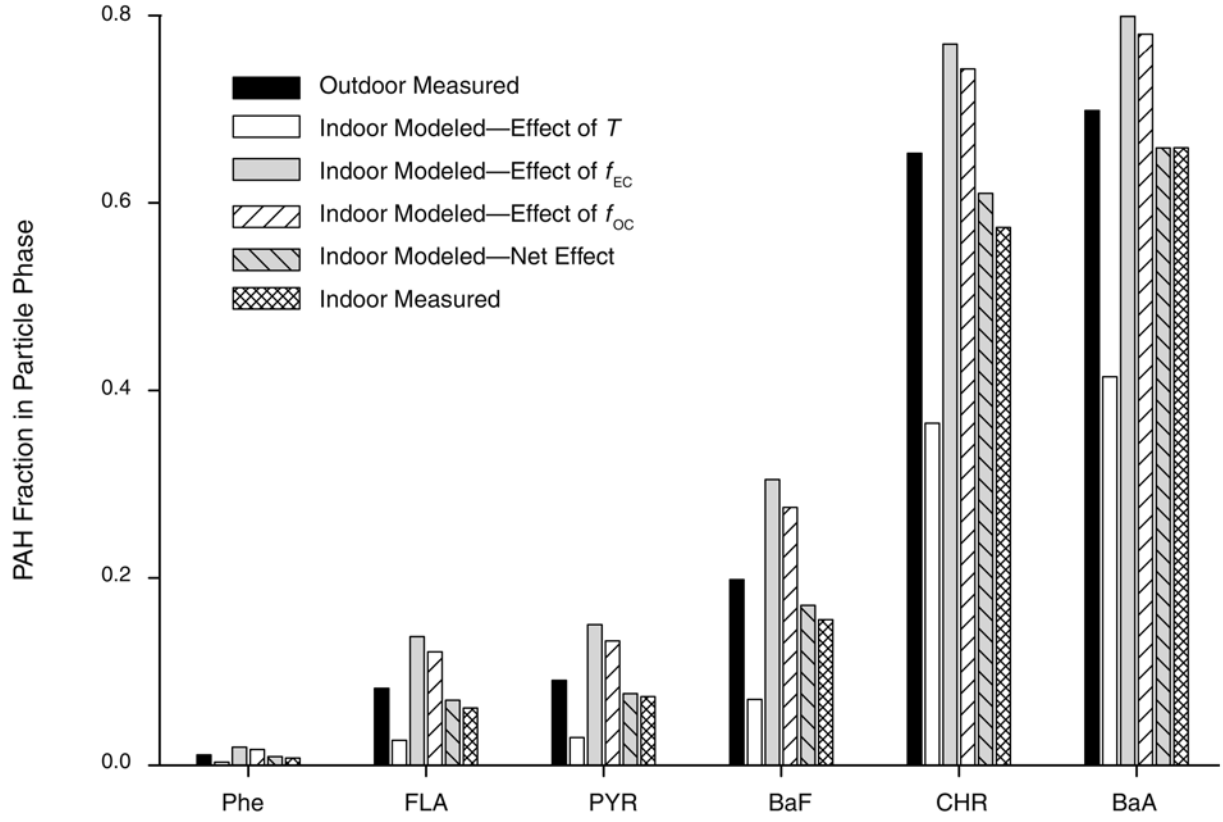


Figure 25. Measured and calculated fractions in particle phase for selected PAHs in paired indoor–outdoor samples collected in a Los Angeles home December 13 to 14, 1999. Shown are the predicted changes in partitioning when outdoor aerosol is adjusted to indoor T, indoor f_{EC}, and indoor f_{OC}, and when all three adjustments are made (net effect). Shown also are the fraction in the particle phase measured in the indoor and outdoor samples.

due to OC sampling artifacts, cannot affect the explained variance in the measured partition coefficients. An important limitation of the MLR model is the assumption that the estimated effect of each predictor is independent of other variables. For example, it assumes that the estimated effect of f_{EC} or f_{OC} on the partition coefficients is constant across the compound class. This does not agree with the findings of some studies that examined the processes underlying adsorptive and absorptive partitioning. Computations based on group contribution methods indicate that activity coefficients of PAHs in different types of organic matter can vary within the compound class (Jang et al 1997). Goss and Schwarzenbach (1998) have shown that adsorptive interactions of PAHs with surfaces depend on compound vapor pressure and electron donor properties of aromatic rings, which in turn depend on vapor pressure. The MLR model explained more variance in the partition coefficients than the SLR model did, but left 16% of the variance unexplained.

Chlordanes Chlordane species, including *cis*-chlordane (CC) and *trans*-chlordane (TC), *cis*-nonachlor (CN) and *trans*-nonachlor (TN), oxychlordane (OXY), and MC5, comprise only a small fraction of the organic phase of ambient $PM_{2.5}$. These polychlorinated compounds are, however, among pollutants of concern for human health owing to their carcinogenic and mutagenic properties. Chlordanes are components of Technical Chlordane, a pesticide that was produced and used in North America until 1997. Technical Chlordane is a mixture of approximately 140 compounds. It contains the major components TC (13%), CC (11%), TN (5%), and heptachlor (5%), as well as more than 30 less abundant chlordanes, chlordenes, and nonachlors (Dearth and Hites 1991; Buser and Muller 1993; Mattina et al 1999; Jantunen et al 2000).

Chlordane was first synthesized in 1944 (Dearth and Hites 1991) and thereafter introduced as an agricultural pesticide in the United States and around the world. It gained widespread use as a broad-spectrum pesticide from the 1940s through the 1960s. Regulations on chlordane use were initiated in 1974, and by 1983 the only remaining application of chlordane in the United States was as a termiticide, primarily in new building construction (Aigner

et al 1998). In 1988 the termiticide registration was cancelled (EPA 1988) and sales and use in the United States were halted on April 15. The major producer (Velsicol Chemical Company) voluntarily halted global production in 1997 (Pesticide Action Network of North America 1997).

Because of the thermodynamic properties of chlordane, illustrated by its chemical stability, vapor pressure, the Henry Law constant, octanol–water partition coefficient, and octanol–air partition coefficient, chlordane species are ubiquitous in the environment. They have been found in the air and biota of the Arctic, Antarctic, and many other regions of the world (Norstrom et al 1988; Bidleman et al 2002). Further, these compounds tend to bioconcentrate and biomagnify in the aquatic food web (Muir et al 1988, 1996; Kucklick et al 1996; Kucklick and Baker 1998). Chlordanes have also been found in human serum, human breast milk, and human adipose tissue (Barquet et al 1981; Murphy and Harvey 1985; Skaare et al 1988; Walker et al 2003). In fact, some aboriginal peoples have a dietary intake of chlordanes (including nonachlors and OXY) through traditional foods that exceeds the tolerable daily intake value (van Osstdam et al 1999).

In the atmosphere chlordanes partition between the gas and particle phases in accord with atmospheric temperature, compound vapor pressure, and particle properties. Several studies have characterized concentrations of chlordane species in outdoor air (eg, Hoff et al 1996; Bidleman et al 1995, 1998, 2002; Oehme et al 1995; Jantunen et al 2000). Generally, atmospheric concentrations are positively correlated with outdoor air temperature, indicating that air–surface exchange of chlordane species is a major determinant of outdoor concentrations. Until the mid-1990s chlordanes were still in active use as termiticides in indoor environments. Concentrations of chlordane species in indoor air and their attribution to indoor and outdoor sources have been characterized in a few studies (Wallace 1996; Jantunen et al 2000), the most comprehensive of which was the Nonoccupational Pesticide Exposure Study (NOPES) conducted in 1988 (Whitmore et al 1994).

The main objective of the chlordane component of the current study was to characterize indoor and outdoor concentrations of chlordanes and improve understanding of how indoor and outdoor sources contribute to residential indoor concentrations. To this end, indoor chlordane emission rates were calculated for study residences, providing data to improve human exposure prediction.

The outdoor and indoor total (gas phase + particle phase) Σchlordane concentrations, defined as the sum of the concentrations of TC, CC, TN, and CN in both the gas and particle phases, are presented in Figure 26. The total Σchlordane concentrations in the outdoor samples ranged from 36 to 4270 pg/m^3 in Los Angeles, from 8 to 11,000 pg/m^3 in Elizabeth, and from 62 to 1770 pg/m^3 in Houston. The corresponding indoor total Σchlordane concentrations ranged from 37 to 111,500 pg/m^3 in Los Angeles, from 260 to 31,800 pg/m^3 in Elizabeth, and from 410 to 38,900 pg/m^3 in Houston. Geometric mean concentrations were higher in indoor air than in outdoor air for all three cities: 1980 vs. 580 pg/m^3 in Los Angeles; 1300 vs. 170 pg/m^3 in Elizabeth; and 4180 vs. 280 pg/m^3 in Houston. The outdoor Σchlordane concentrations were not significantly different between the three cities according to ANOVA ($\alpha = 0.05$) of log-transformed data. Variations in the chlordane concentrations in the outdoor and indoor samples were driven by the gaseous compounds, which comprised on average ~90% of the total chlordane mass measured in the samples.

The outdoor concentrations measured in the three study cities are only slightly lower than high levels measured in the southern United States (South Carolina in 1994–1996 and Alabama in 1996–1997; Bidleman et al 1998; Jantunen et al 2000), yet they are higher than levels measured over and near the Laurentian Great Lakes (Cortes et al 1998; McConnell et al 1998). The concentrations reported here are 1 or 2 orders of magnitude higher than measurements taken within approximately 3 years in Arctic, high Arctic, and Antarctic atmospheres (Bidleman et al 2002).

The indoor air concentrations of chlordane species measured in this portion of the study were somewhat lower than those measured in NOPES (120 homes in Jacksonville, Florida, and Springfield, Massachusetts; Whitmore et al 1994). The indoor chlordane concentrations in NOPES ranged from 220 to 324 ng/m^3 in Jacksonville and from

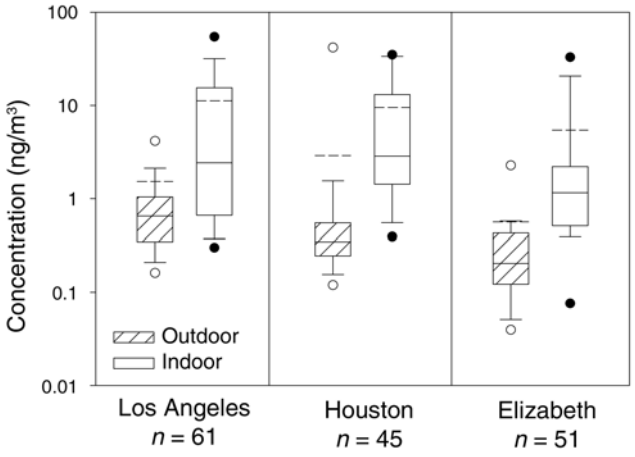


Figure 26. Outdoor and indoor total (gas phase + particle phase) Σchlordane concentrations (sum of TC, CC, TN, and CN). Boxes show 25th to 75th percentiles, whiskers are 10th and 90th percentiles, and circles are 5th to 95th percentiles. Solid and dashed lines inside the boxes show the median and mean values, respectively.

34.8 to 199 ng/m^3 in Springfield. For both cities average indoor chlordane concentrations were higher than corresponding outdoor concentrations. A few measurements of chlordanes made in areas of historical agricultural (outdoor) chlordane usage showed extremely high indoor concentrations of these species (Jantunen et al 2000). In a detailed investigation of a single house, Wallace (1996) found a trend of chlordane concentrations increasing from the second floor down to the basement, suggesting that the primary source was volatilization from the foundation or basement of this home.

Indoor total (gas phase + particle phase) chlordane concentrations often exceeded outdoor concentrations at study homes (Figures 26 and 27). The indoor Σchlordane concentration was greater than the outdoor concentration for 99 of 108 homes that had paired indoor and outdoor total Σchlordane concentrations. Similarly, in 103 of 112 homes for which there were indoor and outdoor concentrations above the MDL for gas and particle phases, indoor TC exceeded outdoor. Of these, there were 95 homes for which the I/O for TC was greater than 2, and 46 homes for which it was greater than 10. Likewise, in 100 of 112 homes the I/O for CC was greater than 1. Of these, there were 84 homes for which the I/O for CC was greater than 2, and 35 homes for which it was greater than 10. This is strong evidence of the presence of current sources of chlordanes indoors.

Indoor emission rates can be estimated by treating the home as a space with a fixed air volume in which the species of interest is completely mixed. If one assumes that mixing is perfect and instantaneous and that factors affecting indoor concentrations are constant or change slowly throughout the monitoring period, then the change in the indoor concentration with time can be described as follows:

$$VdC_{In}/dt = PaVC_{Out} - VaC_{In} + Q_{In} - kVC_{In}, \quad (8)$$

where a is the AER (hr^{-1}), P is the penetration factor (penetration through the building envelop), k is the loss rate constant (decay rate due to deposition and reaction; hr^{-1}), C_{In} and C_{Out} are concentrations measured indoors and outdoors (ng/m^3), Q_{In} is the indoor source strength or emission rate (ng/hr), and V is the total home air volume(m^3).

At steady state ($dC_{In}/dt = 0$), equation 8 becomes

$$C_{In} = [Pa/(a + k)]C_{Out} + (Q_{In}/V)/(a + k), \quad (9)$$

where the first term is the outdoor contribution and the second is the contribution from indoor sources (ng/m^3). This equation can be rearranged to yield indoor chlordane emission rates (Q_{In}; ng/hr). Outdoor half-lives for chlordane species are on the order of approximately 7 to 10 days, which translate into reaction rate constants of approximately 0.003/hr (Atkinson 1987; Scheringer 1997). AER was measured for each home.

As a limiting case, a value of unity was chosen for the penetration factor, P, and zero for the loss rate constant, k. These are common assumptions for nonpolar gases; more than 90% of chlordanes in study samples were in the gas phase. Particle-phase chlordanes are expected to have somewhat lower penetration coefficients and higher loss rates. Thus the assumptions used provide lower-bound estimates of indoor chlordane emission rates at study homes. The average indoor emission rate was 1517 ± 3292 ng/hr. For some homes this lower-bound approach estimated negligible indoor emissions; however, four homes had emission rates greater than 14,000 ng/hr.

The era of home construction appeared to play a role in the indoor chlordane emission rate. Five of the eight highest indoor emission rates for Σchlordanes (all > 5000 ng/hr) occurred in homes that were constructed between 1945 and 1959 (Figure 28).

In summary, strong indoor chlordane sources existed in a fraction of the study homes in each city. Calculated indoor source strengths averaged approximately 1000 ng/hr across all homes, which translates into a total mass flux of approximately 0.26 g from each home over a period of 30 years, assuming constant emission. This estimated emission, which is several orders of magnitude less than the typical application of several kilograms, indicates that these strong indoor emissions may continue for many years. High emission rates were most often calculated for homes built during the period from 1945 to 1959. Indoor sources of chlordanes are likely to include volatilization from residues of termiticide applications indoors and infiltration from subsurface and foundation applications during home construction.

OUTDOOR CONTRIBUTIONS TO INDOOR AND PERSONAL $PM_{2.5}$

Quantitative assessment of the outdoor and indoor contributions to indoor $PM_{2.5}$ concentrations and personal exposures can be used to better understand the implications of exposure errors for epidemiologic findings and to develop effective strategies for controlling outdoor exposures and mitigating indoor exposures. In addition, because $PM_{2.5}$ of outdoor origin and of indoor origin have been shown to differ in composition, it is possible that their health effects also differ. Below, estimates of the outdoor and indoor contributions to personal and indoor $PM_{2.5}$ concentrations are provided. Then, the mass concentration and percentage of indoor $PM_{2.5}$ that originates outdoors, calculated in three ways with increasingly accurate assumptions, are used to examine, in part, the use of central-site $PM_{2.5}$ as a surrogate for exposure. The focus on

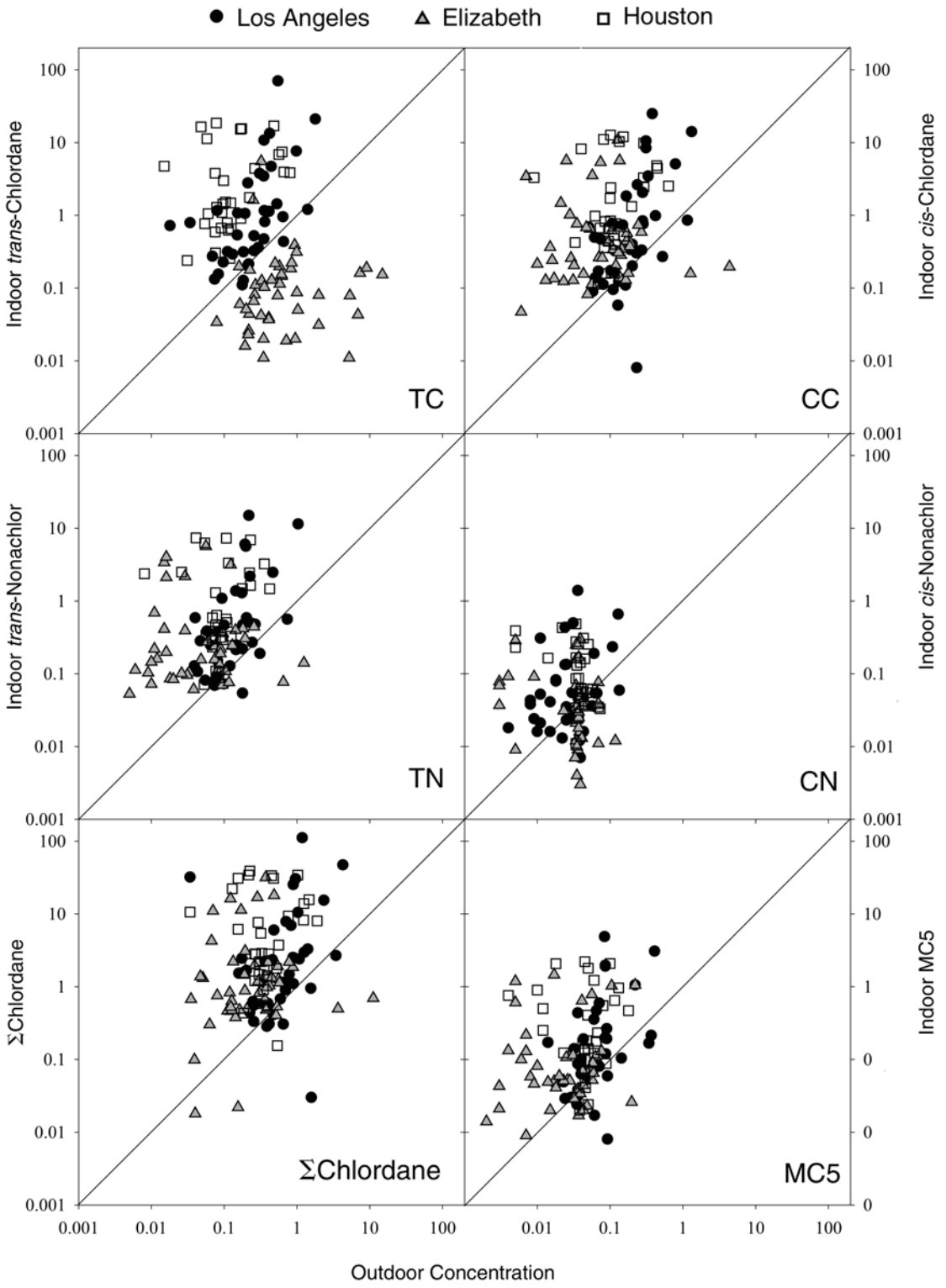


Figure 27. Indoor and outdoor total (gas phase + particle phase) concentrations (in ng/m^3) of chlordane species: TC, CC, TN, CN, Σchlordane, and MC5.

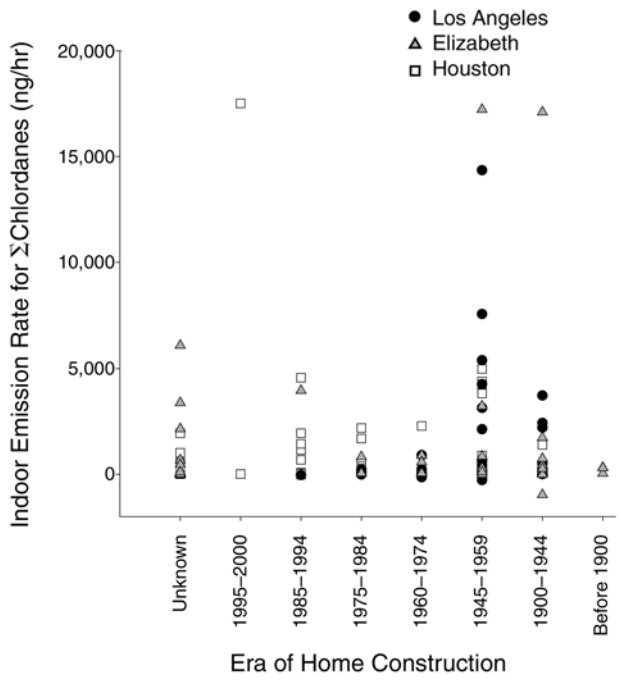


Figure 28. Indoor emission rate (ng/hr) of Σchlordanes listed by era of home construction for Los Angeles, Elizabeth, and Houston homes.

indoor $PM_{2.5}$ is warranted because people spend most of their time indoors.

Assuming perfect instantaneous mixing and assuming that factors affecting indoor concentrations are constant or change slowly throughout the monitoring period, the indoor $PM_{2.5}$ mass concentration can be described with a single-compartment mass balance model as shown in equation 8.

At steady state this equation becomes

$$\begin{aligned} C_{\mathrm{In}} &= \frac{PaC_{\mathrm{Out}}}{a+k} + \frac{Q_{\mathrm{In}}/V}{a+k} \\ &= F_{\mathrm{Inf}}C_{\mathrm{Out}} + C_{\mathrm{IgI}} \\ &= C_{\mathrm{IgO}} + C_{\mathrm{IgI}}, \end{aligned} \tag{10}$$

where C_{In} and C_{Out} are $PM_{2.5}$ concentrations (μg/m^3) measured indoors and outdoors, P is penetration coefficient (dimensionless), a is AER (hr^{-1}), k is particle loss rate (hr^{-1}), Q_{In} is indoor source strength or emission rate (μg/hr), V is home volume (m^3), F_{Inf} is infiltration factor (dimensionless), C_{IgI} is indoor $PM_{2.5}$ (μg/m^3) generated indoors, and C_{IgO} is indoor $PM_{2.5}$ (μg/m^3) generated outdoors.

The indoor concentration is the sum of two terms. The first term describes the contribution of outdoor $PM_{2.5}$ to the indoor $PM_{2.5}$ concentration (outdoor contribution), and the second term is the contribution of indoor sources to the indoor concentration (indoor contribution). In reality, AER, P, k, and F_{Inf} are different from home to home and from species to species.

The outdoor contribution to indoor $PM_{2.5}$ was calculated in three ways with increasingly accurate assumptions. The first approach (RCS model) assumes that F_{Inf} is constant across homes as would be the case if central-site $PM_{2.5}$ measurements were a perfect surrogate for $PM_{2.5}$ of outdoor origin. The second approach (mass balance model) uses the measured AER for each home and assumes that the penetration of particles into the home (P) and loss rate coefficient (k) of particles indoors are constant across the homes. In this way F_{Inf} varies only with AER. The third approach (robust regression) uses all measured major $PM_{2.5}$ species to calculate home-specific values for F_{Inf}. These infiltration factors take into account the possibility that building construction, ventilation practices, particle size distributions, and particle chemistry or thermodynamics might vary across homes and days, which would introduce home-to-home and day-to-day variations in particle infiltration behavior.

Comparison of the more realistic estimate of PM of outdoor origin provided by the robust regression approach and the estimate obtained from the RCS model illustrates several types of error encountered when epidemiologic studies use central-site PM measurements as a surrogate for personal exposure. Variations in the amount of time spent indoors might also be expected to broaden the distribution of personal exposures to PM of outdoor origin, generating further exposure error with use of a central-site surrogate. We examined the effects of these different assumptions on the distribution of PM of outdoor origin across the homes.

Personal exposure to PM of outdoor origin and PM of indoor origin can be described as follows:

$$\begin{aligned} E_t &= E_{\mathrm{gO}} + E_{\mathrm{gI}} \\ &= \{y + (1-y)[Pa/(a+k)]\}C_{\mathrm{Out}} + E_{\mathrm{gI}} \\ &= \alpha C_{\mathrm{Out}} + E_{\mathrm{gI}}, \end{aligned} \tag{11}$$

where E_{t} (μg/m^3) is total time-averaged personal $PM_{2.5}$ exposure, E_{gO} (μg/m^3) is time-averaged personal exposure to $PM_{2.5}$ generated outdoors, E_{gI} is time-averaged personal exposure to $PM_{2.5}$ generated indoors (μg/m^3), y is the fraction of time a person spent outdoors (dimensionless), and α is the attenuation factor (dimensionless).

Estimates with Constant Infiltration and Attenuation Factors

To estimate the mean and distribution of outdoor and indoor contributions to indoor $PM_{2.5}$, the RCS statistical model proposed by Ott and associates (2000) regresses the

measured outdoor concentrations (C_{Out}) on indoor concentrations (C_{In}). The result is a constant infiltration factor (F_{Inf}), as shown in equation 10. The product of this infiltration factor with each outdoor concentration (C_{Out}) provides an estimate of the mean and distribution of the outdoor contributions for the homes. The mean and distribution of indoor contributions are given by the difference between this quantity and the indoor concentration (C_{In}). This model is not used to estimate indoor and outdoor contributions for *individual* homes, in part because the use of a single attenuation factor does not account for the large home-to-home variations in actual AERs. This model assumes (1) linear superposition of the outdoor and indoor components of the calculation, and (2) lack of correlation between these two components.

The RCS model is also sometimes used to estimate the outdoor and indoor contributions to personal exposure. This is done by regressing time-averaged personal exposure (E_t) on the outdoor concentration (C_{Out}), as shown in equation 11. The slope of the regression is the attenuation factor (α). The products of this constant attenuation factor with each of the outdoor concentrations (C_{Out}) provide an estimate of the mean and distribution of the outdoor contributions to personal exposure for the population of homes. The difference between this quantity and personal exposure (E_t) gives the mean and distribution of indoor contributions. Again, the model assumes (1) linear superposition of the outdoor and indoor components of exposure, and (2) lack of correlation between these two components.

The RCS statistical model was used to obtain a constant infiltration factor (outdoor–indoor) and attenuation factor (outdoor–personal) for the homes with measured $PM_{2.5}$ mass concentrations. These were then used to provide distributions (across homes) of outdoor and indoor contributions to indoor $PM_{2.5}$ concentrations and personal $PM_{2.5}$ exposures.

Outdoor contributions to indoor concentrations and personal exposures calculated using the RCS model are based on the statistical inferences of regression analysis. Indoor–outdoor or personal–outdoor relations could be affected by extreme values (outliers), such as a high indoor exposure on a day with a low outdoor concentration or vice versa. For this reason outliers were identified and their influence on the infiltration factor or attenuation factor in the RCS model was evaluated. A value was considered an outlier if the absolute studentized residual of that data point was larger than 3. In evaluating the outdoor $PM_{2.5}$ contribution to the indoor $PM_{2.5}$ concentration, seven outliers were identified. After removing those outliers, F_{Inf} changed by 0.01. In evaluating the outdoor $PM_{2.5}$ contribution to personal exposure, four outliers were found. Elimination of these outliers changed the attenuation factor by 0.05. Eliminating outliers increased the RCS-estimated mean outdoor contribution to indoor exposure concentration by 0.1 μg/m^3 and RCS-estimated mean outdoor contribution to personal exposure concentration by 0.9 μg/m^3.

Estimates with Constant Penetration and Loss Rate Coefficients and Measured AERs

In this work the mass balance model was used to estimate the outdoor and indoor contributions to indoor $PM_{2.5}$ concentrations and personal $PM_{2.5}$ exposures. Both the RCS and mass balance models used measured $PM_{2.5}$ mass concentrations. The primary difference between the two models is that the mass balance model takes into consideration the measured AER, which varies considerably from home to home. The terms in equation 10 that were not measured in the current study were P, k, and Q_{In}. In the mass balance model, population-averaged P and k were obtained by fitting measured indoor and outdoor $PM_{2.5}$ concentrations and AERs to the mass balance equation (equation 10) using nonlinear regression (NLIN procedure in SAS, Cary NC). This resulted in the estimation of single values for P and k for the data set. Outdoor and indoor contributions to indoor $PM_{2.5}$ concentrations and personal exposures were then calculated for each home or participant assuming these constant values of P and k, and using the measured $PM_{2.5}$ concentrations, AERs, and time spent indoors (from activity diaries) for each residence and subject.

Parameter Estimation Table 15 presents the fitted values of P and k from the nonlinear regression of C_{In}, C_{out}, and a

Table 15. Fitted Values of P and k Coefficients from the Nonlinear Regression of C_{In} on C_{Out} and a Using the Mass Balance Model With and Without Bounding[a]

Number of Samples and Boundary Condition[b]	P	95% CI of P	k (hr^{-1})	95% CI of k
Overall (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] Shown are solutions with and without bounding $P\epsilon[0,1]$ for all data and for each city separately. Physically, P must be between 0 and 1.

[b] Yes means parameters are estimated with boundary condition $P\epsilon$ [0,1]; no means no boundary conditions are constrained for parameter estimation.

using the mass balance model both with and without bounding $P \in [0,1]$ for all data and for each city separately. Physically, the penetration coefficient, *P*, must be between 0 and 1. Comparison of bounded and unbounded results shows that estimates of *P* and *k* are linked; therefore, the true values of each cannot be known without independent knowledge of the other. Also, there are several reasons to expect that *P* and *k* will vary from day to day and home to home. Nevertheless, this approach represents an improvement over the RCS model because the variability in AER is considered. Regression of all homes yielded an estimated *P* of 0.91 (95% CI, 0.71–1.12) and *k* of 0.79 hr^{-1} (95% CI, 0.18–1.41).

In reality, the penetration coefficient, *P*, varies with particle size and home structure. The indoor particle loss rate, *k*, is determined by many factors, such as surface-to-volume ratio, housing structure, near-surface air flows, turbulence, and particle size distribution. The use of air conditioning results in higher particle loss rates. The value of *k* obtained from the nonlinear regression procedure is an average value for all homes. The use of a single value of *k* for all homes probably introduces the largest uncertainty in the mass balance results. The effects of AER and particle loss rate on the fraction of outdoor $PM_{2.5}$ found indoors (infiltration factor) for all data are illustrated in Figure 29. The two curves in Figure 29 show the infiltration factor as a function of AER assuming a loss rate of 0.79 hr^{-1} (the value estimated from all homes) or 0.4 hr^{-1} (half the estimated value). Each point on the plot gives the infiltration factor for a single home at the measured AER (*a*), a fixed particle loss rate (*k*), and a penetration coefficient equal to 0.91 (the *P* calculated using all study homes). If AER is very small, then *k* is an important determinant of the infiltration factor. At AER of 1 hr^{-1}, changing *k* from 0.79 hr^{-1} to 0.4 hr^{-1} changes the infiltration factor from roughly 55% to nearly 70%.

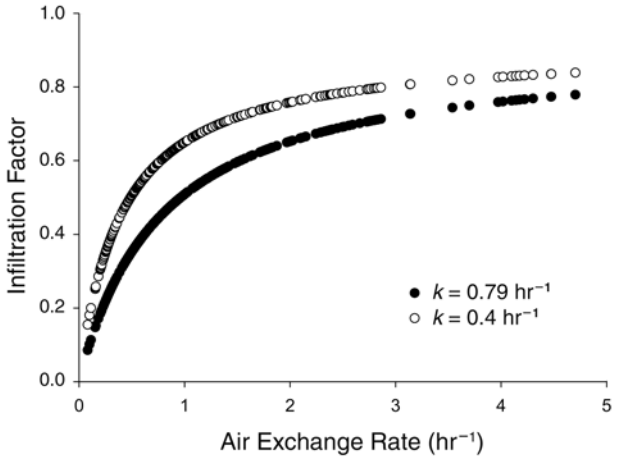


Figure 29. Infiltration factor estimated using the mass balance model as a function of AER assuming particle loss rates of 0.79 hr^{-1} (value estimated from all homes) and 0.4 hr^{-1} (half the estimated value).

The particle penetration and loss rate values estimated here are in reasonable agreement with those in other $PM_{2.5}$ studies. Koutrakis and coworkers (1992) estimated a value for *P* of 0.84. Experiments by Thatcher and Layton (1995) yielded a *P* of 1, and Özkaynak and colleagues (1996) also obtained *P* of 1 (95% CI, 0.89–1.11) through statistical data analysis. Lachenmyer and Hidy (2000) calculated a *P* of 0.95, and Winkle and Scheff (2001) obtained a *P* of 0.89 from parameter estimation. Long and associates (2001a) estimated a larger value for *P* in the summer (1.11 ± 0.10) and a smaller one in the winter (0.54 ± 0.02). The estimated value of *k* for $PM_{2.5}$ for homes in the current study was 0.79 hr^{-1} (95% CI, 0.18–1.4 hr^{-1}). Özkaynak and colleagues (1996) estimated a *k* of 0.39 hr^{-1} [0.22–0.55 hr^{-1}] for the PTEAM study. Lachenmyer and Hidy (2000) estimated a *k* of 0.6 hr^{-1} with a range of 2.0 hr^{-1}. Abt and coworkers (2000b) and Vette and coworkers (2001) estimated loss rates 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; in Boston, the lowest *k* was 0.7 hr^{-1} for particles 0.4 to 0.5 µm in diameter, and the highest *k* was 1.2 hr^{-1} for particles 2 to 3 µm in diameter. In all these studies, the values for *P* and *k* were either estimated by the nonlinear regression, similar to the current study (Özkaynak et al 1996; Lachenmyer and Hidy 2000), or measured in controlled chambers or controlled residences.

The mean contribution of outdoor sources to indoor $PM_{2.5}$ concentrations estimated using the mass balance approach was 8.7 µg/m^3 or 60% for all homes (67%, 70%, and 41% for Los Angeles, Elizabeth, and Houston homes, respectively; Table 16). The median contribution of outdoor

Table 16. Outdoor Contributions to Indoor and Personal $PM_{2.5}$ Concentrations Using the Mass Balance Model

Category (*n*)	Mean (µg/m^3) Contribution	Mean (%) Contribution
Overall study		
Indoor (268)	8.7	60
Personal exposure (197)	9.3	26
Los Angeles homes		
Indoor (112)	10.2	67
Personal exposure (85)	8.3	33
Elizabeth homes		
Indoor (80)	9.5	70
Personal exposure (53)	8.2	22
Houston homes		
Indoor (76)	5.4	41
Personal exposure (59)	8.5	21

sources to indoor $PM_{2.5}$ concentrations was estimated to be 7.2 μg/m^3 or 56% for all homes (63%, 52%, and 33% for Los Angeles, Elizabeth, and Houston homes, respectively).

The mean outdoor contribution to personal $PM_{2.5}$ exposure estimated using the RCS model was 25% for all homes (33%, 33%, and 13% for Los Angeles, Elizabeth, and Houston homes, respectively). These values are consistent with the results from the mass balance approach (26% for all homes, and 33%, 22%, and 21% for Los Angeles, Elizabeth, and Houston homes, respectively; Table 16). Harmonizing PEM and Harvard impactor measurements using equation 2 would increase the mean percentage contribution of outdoor to personal $PM_{2.5}$ exposure from 26% to 27% (mass balance) over all homes.

Sensitivity Analysis The sensitivity of the mass balance results to the choice of particle penetration and loss rate coefficients is shown in Figure 30. It gives the mean (panel A) and median (panel B) percentage of contributions from outdoor $PM_{2.5}$ to the indoor $PM_{2.5}$ concentration for Los Angeles, Elizabeth, and Houston homes individually and together. Seven alternatives (designated A through G) are presented: bars A–C are based on data from other studies; bars D–G are based on data from this study. For bar A, P (1) and k (0.39 hr^{-1}) were suggested by the PTEAM study. For bar B, P (0.95) and k (0.62 hr^{-1}, the average of winter and summer values) were taken from Lachenmyer and Hidy (2000). For bar C, P (1.0) and k (0.5 hr^{-1}) were based on other reasonable estimates in the literature (Thatcher and Layton 1995; Abt et al 2000b; Long et al 2001a; Thornburg et al 2001; Vette et al 2001). For bar D, P (0.91) and k (0.79 hr^{-1}) were estimated from all homes in the current study. For bar E, city-specific estimates of P and k were determined separately from the study homes (Table 15, results with boundary conditions).

For bars F and G, we attempted to eliminate homes with substantial indoor sources before the nonlinear regression was performed to find estimates of P and k. This was done because the goal was to find P and k, which were assumed to be independent of the indoor source strength, and because the indoor source strength, which is highly variable from home to home, introduces scatter that makes estimation of P and k more difficult. In this approach, the variability in the indoor source term from home to home is substantially reduced, but so is the number of homes. For bar F, only homes in which the indoor $PM_{2.5}$ concentration was less than the outdoor concentration were used, yielding $P = 0.73$ and $k = 0.19$ hr^{-1} ($n = 165$). For bar G, questionnaire data were used to eliminate homes with identified indoor sources. Of the remaining 23 homes, two had indoor $PM_{2.5}$ concentrations greater than the corresponding outdoor concentrations. A nonlinear regression of the remaining 21 homes yielded $P = 0.78$ and $k = 0.40$.

Figure 30 is intended to illustrate the sensitivity of results to uncertainties in P and k. Literature-based values of P and k are likely to have less accurate results than estimates based on data from this study (D and E) because P and k will vary with type of home, climate, and particle source mix. The comparison presented in Figure 30 suggests that uncertainties in P and k led to uncertainties on the order of 20% in the mean outdoor contribution to indoor $PM_{2.5}$.

To examine the compatibility of results from the mass balance and RCS statistical models, the distribution of outdoor and indoor contributions to indoor $PM_{2.5}$ concentrations for 268 indoor–outdoor pairs of PM samples and corresponding AERs was estimated using equation 10, measured indoor and outdoor concentrations, and either (1) the infiltration factor obtained from the RCS model or (2) the measured AER along with particle penetration and loss rate coefficients obtained from the mass balance model. This was done to see how much the use of actual AERs would change the mean and distribution of $PM_{2.5}$ of outdoor origin. The infiltration factor (the slope of the regression of indoor on outdoor $PM_{2.5}$ concentrations) was 0.46 (RCS model). The values of $Pa/(a + k)$ calculated from the mass balance model were approximately normally distributed (by the Shapiro-Wilk test with $\alpha = 0.05$ and $P > 0.15$) with a mean of 0.46 and SD of 0.16; this is consistent with the fixed RCS attenuation factor of 0.46.

Figure 31 shows the cumulative log-normal distributions of indoor (panel A) and outdoor (panel B) contributions to indoor $PM_{2.5}$. The two curves reflect results from the mass balance model (variable infiltration factor) and RCS model (fixed infiltration factor). 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. Introducing the actual AERs (mass balance model) provided a broader distribution of values of PM of outdoor origin across homes.

The RCS model is not designed to predict the indoor and outdoor contributions for individual homes; however, we found that the CV for the outdoor contribution to indoor $PM_{2.5}$ for the two models was 26% when results were compared for the 268 indoor–outdoor pairs of samples. The CV for the indoor contribution to indoor $PM_{2.5}$ was 24%. Results were reasonably well correlated, with r^2 greater than 75%, for both outdoor and indoor contributions to indoor concentrations.

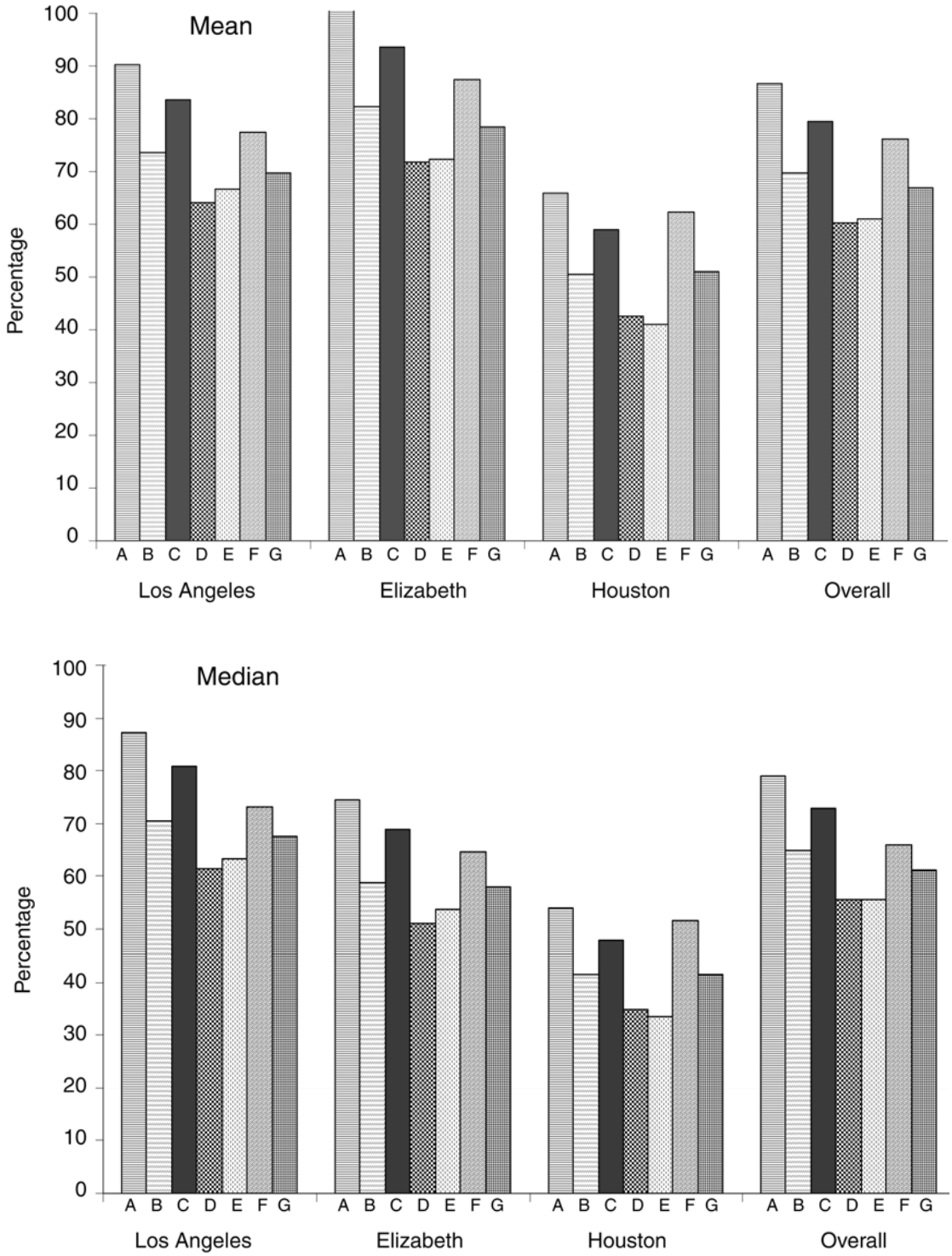


Figure 30. Sensitivity of the mass balance model results to the choice of particle penetration and loss rate coefficients. Mean (top) and median (bottom) percentage of contribution from outdoor $PM_{2.5}$ to indoor $PM_{2.5}$ using seven scenarios: (A) $P = 1$, $k = 0.39$ (from the PTEAM study); (B) $P = 0.95$, $k = 0.62$ (from Lachenmyer and Hidy 2000); (C) $P = 1$, $k = 0.5$ (based on published reasonable estimates; see text); (D) $P = 0.91$, $k = 0.79$ estimated from all homes in this study; (E) P and k estimated from homes separated by city (Table 17); (F and G) P and k calculated from a subset of homes with fewer sources (see text). Values in D or E should be considered best estimates.

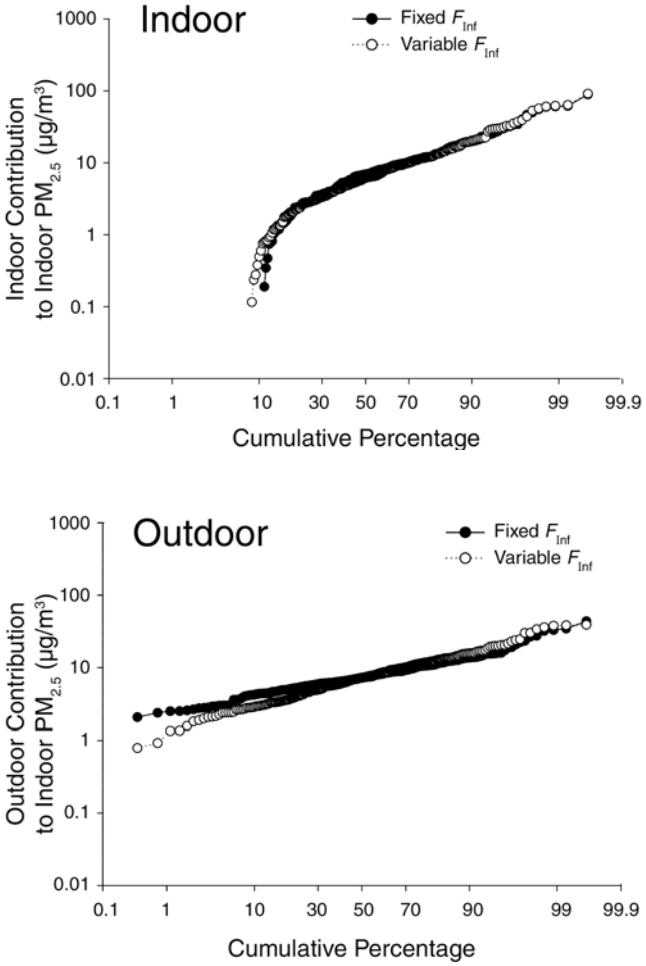


Figure 31. Cumulative log-normal distributions of indoor and outdoor contributions to indoor $PM_{2.5}$ mass concentrations. The two curves represent results from the mass balance model (variable F_{Inf}) and RCS model (fixed F_{Inf}). n = 268 paired indoor–outdoor samples.

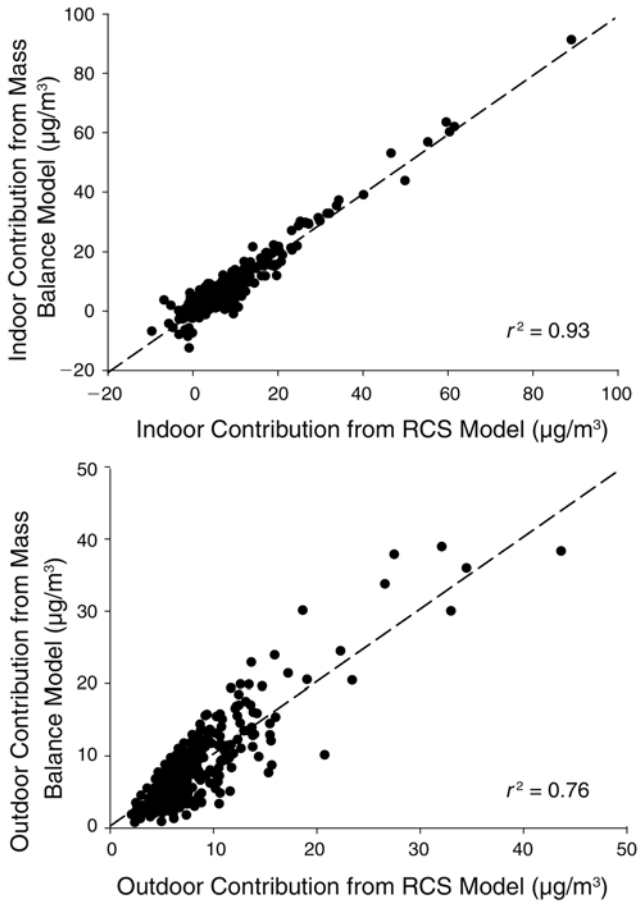


Figure 32. RCS and mass balance model estimates of indoor (top) and outdoor (bottom) contributions to indoor $PM_{2.5}$ mass concentrations. Note that axes differ in length between the panels. n = 268 paired indoor–outdoor samples.

Figure 32 shows the results of the paired data comparison. In the RCS model a single fixed infiltration factor was applied to all homes. However, in reality this quantity is affected by AER, particle loss rate, and penetration coefficient, all factors that vary from home to home. This comparison of methods suggests that estimating indoor and outdoor contributions without using measured AERs introduced an uncertainty of 26% in the estimate of PM of outdoor origin.

Estimating the Infiltration Factor for Each Home

Outdoor contributions to indoor $PM_{2.5}$ concentrations were calculated by estimating a separate infiltration factor for each home using $PM_{2.5}$ species data. Spatial and temporal variations in the $PM_{2.5}$ source mix and variations in proximity to fresh emissions result in variations in particle composition. Because different particle formation mechanisms yield different particle size distributions, variations in source mix and proximity of homes to outdoor sources result in home-to-home and day-to-day differences in particle size distributions. These variations cause the penetration and loss rate coefficients to vary because particle removal processes are a strong function of particle size. Although the mass balance approach accounts for variations in AERs, it cannot accommodate variations in particle infiltration behavior due to variations in particle properties or variations in home structure, both of which can cause particle penetration and loss rate to vary with home and day. Measurements of $PM_{2.5}$ species made at the same time in the same home were used to estimate the infiltration factor for each home on the day the measurements were made.

Figure 33 shows the indoor and outdoor concentrations of measured species for two study homes, plotted separately. The robust regression line is included. The data presented for each home are individual $PM_{2.5}$ species measured concurrently. Twenty-two species were used in this method (EC, OC, Al, As, Br, Ca, Cl, Cr, Cu, Fe, K, Mn, Ni, Pb, S, Se, Si, Sr, Ti, V, Zn, and Zr). Some species had substantial indoor sources, as evidenced by indoor concentrations that far exceed their outdoor concentrations. Other species appeared to be distributed around a regression line.

As seen in equation 10, the indoor $PM_{2.5}$ concentration can be described as a function of the outdoor concentration, AER, particle penetration, particle loss rate, and indoor contribution. All concurrently measured species in the same home are affected by the same AER at any given time. If all species also had the same penetration and loss rate coefficients and no indoor sources, then species measured concurrently in the same home would be perfectly correlated and the slope of the indoor species concentrations on the outdoor species concentrations would represent the infiltration factor for that home on that day. In reality, PM species might have different penetration and loss rate coefficients (eg, by having differing size distributions) and some species have indoor sources.

Infiltration factors were estimated for each home, allowing for indoor sources of several species and for differences in species penetration and loss rate coefficients as long as indoor and outdoor contributions are independent. The use of many PM species, and not only one or two species (for example, sulfur) is helpful because the infiltration behavior is unlikely to be the same for each species. For example, combustion-derived materials like EC are unlikely to have the same size distribution or infiltration behavior as species formed in the atmosphere like sulfate, or species formed through mechanical abrasion like soil elements. Some researchers have used the I/O of sulfur concentrations to describe the infiltration behavior of PM mass. In the current study we found that indoor and outdoor sulfur values were well correlated ($r^2 = 0.70$), but outdoor sulfur and outdoor $PM_{2.5}$ mass concentrations were poorly correlated ($r^2 = 0.33$). The poor correlation with PM mass, along with the knowledge that different PM species can have quite different size distributions, suggests that the use of multiple species, rather than sulfur alone, would be a better method for characterizing the infiltration behavior of $PM_{2.5}$ in this study.

A robust regression method, called least-trimmed squared regression (S-Plus, Insightful, Seattle WA), was used to regress the indoor species concentrations on the concurrently collected outdoor species concentrations, yielding a $PM_{2.5}$ infiltration factor for each home. In this analysis outliers are species for which indoor sources contributed significantly to their concentrations. Therefore it is desirable to considerably down-weight outliers in the regression used to estimate the infiltration factor. The least-trimmed squared regression is robust with respect to outliers in the response and predictor variables, even when as many as half of the data points are outliers (Rousseeuw and Leroy 1987).

The least-trimmed squared regression was used to estimate the $PM_{2.5}$ infiltration factor for 114 indoor–outdoor pairs of PM species measurements. The mean, median, and SD of the infiltration factors were 0.69, 0.70, and 0.23, respectively. The product of the estimated home infiltration factor and the corresponding measured outdoor $PM_{2.5}$ concentration is the contribution of outdoor $PM_{2.5}$ to the indoor $PM_{2.5}$ concentration (ie, PM of outdoor origin in $\mu g/m^3$) for that home. The mean, median, and SD

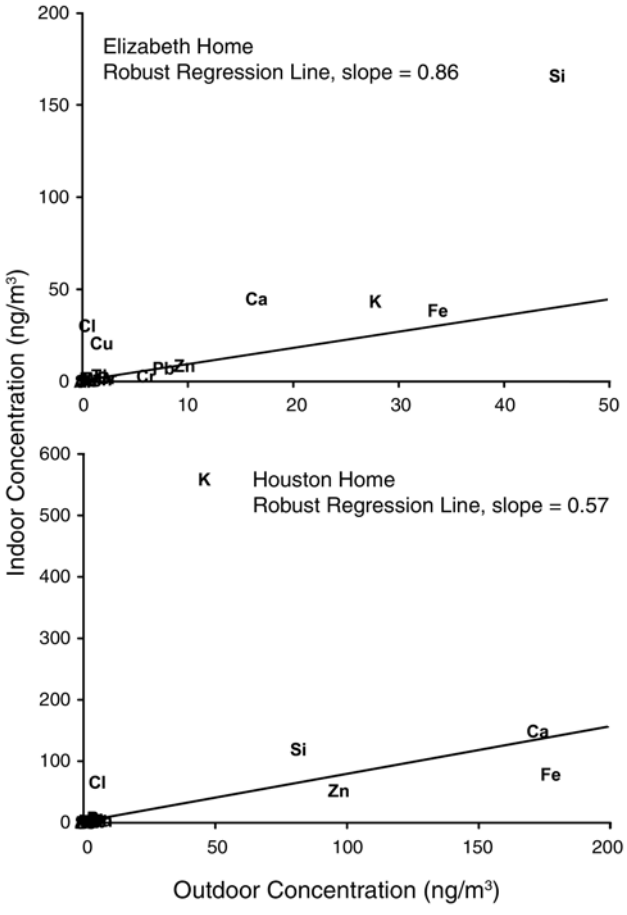


Figure 33. PM species at two homes: home 23 in Elizabeth and home 36 in Houston. Each data point represents a single PM species collected concurrently. Si (top) and K (bottom) are substantially above the regression line, presumably because they have indoor sources.

of percentage of indoor PM that originated outdoors were 73%, 74%, and 36%, respectively, using this approach. The mean outdoor contribution was higher than means obtained by the RCS and mass balance models.

The line of triangles in Figure 34 shows the cumulative distribution of PM of outdoor origin using the least-trimmed squared regression. Once again, this method takes into consideration home-to-home variations in AER, particle penetration, and particle loss rate that can occur due to variations in parameters such as home structure, season, air conditioner use, and particle size distributions (ie, source mix and source proximity). It assumes that indoor and outdoor source contributions are independent, that indoor mixing is perfect and instantaneous, and that factors affecting indoor concentrations are constant or change slowly throughout the monitoring period. This method provides an estimate of the outdoor contribution for the measured home on the day it was sampled.

Comparisons between the RCS and mass balance model results show how the modeled distribution of PM of outdoor origin changed when actual variations in AER were taken into consideration. Comparisons between mass balance model and robust regression results show how the modeled distribution of PM of outdoor origin changed when particle penetration and loss rate were allowed to vary from home to home and day to day. The modeled values of PM of outdoor origin derived from RCS, mass balance, and robust regression approaches are based on increasingly accurate assumptions. Central-site PM measurements would be a perfect surrogate for exposure to PM of outdoor origin if central-site PM mass and species concentrations were related to exposure by a constant, similar to the constant infiltration factor of the RCS model. When indoor concentrations of PM of outdoor origin are modeled using a constant infiltration factor, rather than values of particle penetration, particle loss rate, and AER that reflect home-by-home and day-by-day differences, the exposure errors introduced represent a subset of the exposure errors introduced when central-site PM is used as a surrogate for exposure to PM of outdoor origin. This work provides some insight into those errors.

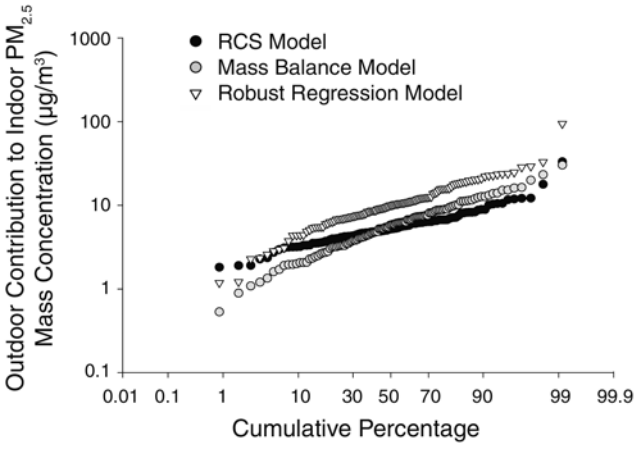


Figure 34. Indoor $PM_{2.5}$ of outdoor origin as determined by RCS model, mass balance model, and robust regression. RCS assumes a constant infiltration factor across all homes. Mass balance uses measured AER in each home but constant particle penetration (*P*) and loss rate (*k*) coefficients across all homes. Robust regression allows *P, k,* and AER to vary with home. Results provided for all three approaches are for the same 114 paired indoor–outdoor samples.

IMPLICATIONS OF RESULTS

EXPOSURE ASSESSMENT

In summary, these results suggest that outdoor $PM_{2.5}$ was the largest source of indoor particles for the mean study home. Outdoor $PM_{2.5}$ was a much smaller source of particles for personal exposure: On average, only about 25% of personal exposure was contributed by outdoor air. The most likely explanation for this finding is that people are exposed to PM through activities that put them in proximity to many sources at home, outdoors, and in other microenvironments (eg, in transit); the concentrations at multiple exposure sites are not well represented by measurements taken at central-site ambient $PM_{2.5}$ monitors.

Mean and median outdoor contributions to indoor and personal concentrations appeared to be similar for the Los Angeles and Elizabeth homes but much lower for the Houston homes. This finding is consistent with our expectations because of the prevalence of air conditioner use in Houston homes. A vast majority of homes in Texas have air conditioning that is used during the summer. In this study, samples were taken across the year. More than 23% of Houston homes, but less than 3% of Los Angeles and about 18% of Elizabeth homes, used air conditioning during sampling. The AER tends to be lower in homes being air-conditioned. As shown by equation 10 and Figure 29, a decrease in the AER resulted in a decreased outdoor contribution to indoor $PM_{2.5}$ concentrations.

In this set of analyses we first assumed a constant infiltration factor across homes, then allowed AER to vary, and finally allowed particle penetration and loss rate coefficients along with AER to vary from home to home. With the increasing degrees of freedom and increasingly accurate assumptions, the distribution of values of PM of outdoor origin became broader. In addition, the mean value of PM of outdoor origin increased when particle penetration and loss rate coefficients were allowed to vary. This increase in the mean is probably a result of the fact that the mean infiltration factor is not equal to the infiltration factor calculated from the means of the parameters particle penetration, particle

loss rate, and AER. The study results are important in part because they demonstrate that PM emitted and formed outdoors is a substantial source of PM exposure.

Because people spend a large majority of their time indoors and $PM_{2.5}$ mass concentrations are largely homogeneous across urban areas, this assessment of residential indoor–outdoor relations provides insights into exposure errors that are introduced when concentrations of $PM_{2.5}$ measured at central sites are used as a surrogate for exposure to $PM_{2.5}$ of outdoor origin. The SD of outdoor contributions across homes increased with increasingly accurate assumptions (ie, decreasing constraints on particle infiltration behavior). This work illustrates several ways in which the use of central-site $PM_{2.5}$ mass concentrations as an exposure surrogate underestimates the distribution of exposures to PM of outdoor origin. Particle infiltration behavior appears to be a substantial contributor to exposure error. AER, particle properties, and housing characteristics all appear to contribute substantially to home-by-home and day-by-day variations in infiltration.

$PM_{2.5}$ EPIDEMIOLOGY

Epidemiologic studies use $PM_{2.5}$ measured at outdoor central-site monitoring stations as a surrogate for exposure to PM of outdoor origin. A perfect surrogate would be perfectly covariant with exposures to PM of outdoor origin. In reality, the relation between exposure to PM of outdoor origin and central-site $PM_{2.5}$ is likely to vary across time (eg, season) and among people for several reasons: (1) $PM_{2.5}$ concentrations vary spatially; (2) the amount of time people spend in different microenvironments varies between people and across time; (3) AER, particle penetration, and particle loss rates vary among homes and seasons; (4) the fine-particle size distribution, and therefore penetration and decay rate, vary with PM source mixture; and (5) some particle-phase species undergo transformations with outdoor-to-indoor transport.

In this project PM of outdoor origin was estimated using three approaches, all in relation to residential outdoor PM concentrations. The distribution across homes of the contribution of PM of outdoor origin to indoor PM concentrations is shown for all three approaches using the same set of data in Figure 34. These results can be used to explain the impacts of some critical assumptions on epidemiologic measurement error. For example, the distribution derived with a constant infiltration factor can be used to represent the assumption made in epidemiologic studies that central-site $PM_{2.5}$ is a perfect surrogate for exposure to PM of outdoor origin. If central-site $PM_{2.5}$ is a perfect surrogate, then it can be assumed that results varied across homes only because homes were sampled on different days. When home-by-home variations in the AER were taken into consideration, the distribution of values of PM of outdoor origin broadened. The final analysis suggests that accounting for other differences between homes, such as particle size distributions or home structure, broadens the distribution further. Spatial variations in $PM_{2.5}$ concentrations and interpersonal variations in the time spent in each microenvironment are likely to broaden the distribution as well. These results suggest that epidemiologic studies that use central-site PM as a surrogate for exposure to PM of outdoor origin will (1) underestimate the distribution of exposures encountered by the study population, which will introduce a random exposure error, and (2) produce larger uncertainties in the relative risk factors for $PM_{2.5}$ exposure than would occur if the studies had used more accurate exposure measures. To restate this, a more accurate assessment of $PM_{2.5}$ exposure would produce smaller uncertainties and a larger likelihood that relative risk factors would be significant. Exposure variability across the population alone will not lead to misclassification of risk in longitudinal epidemiology.

In epidemiologic studies of chronic exposure, time-averaged $PM_{2.5}$ concentrations are assigned retrospectively as surrogates for $PM_{2.5}$ dose. It is possible, at least in theory, for the mean outdoor $PM_{2.5}$ concentration to be higher in City A than in City B, but for the mean exposure to outdoor $PM_{2.5}$ to be higher in City B than in City A because of a difference in particle infiltration behavior. Such a difference could result from variations between the cities in particle properties, ventilation practices, or home structures. For example, City A might be located in Texas and City B in California. In this study, the particle infiltration factors estimated for Houston homes were smaller than those for Los Angeles homes, presumably because of the more extensive use of air conditioning in Houston homes. Such a situation could result in a surrogate (a single time-averaged central-site $PM_{2.5}$ concentration) that does not vary with actual exposure to PM of outdoor origin (represented by the mean of the exposure distribution). Under certain situations seasonal variations in particle infiltration behavior in longitudinal (time-series) epidemiologic studies could cause misclassification of risk as well.

The above results can also be used to understand the impacts of certain critical assumptions on epidemiologic measurement error. For example, the distribution derived using a constant infiltration factor can serve to represent the assumption made in many epidemiologic studies that central-site $PM_{2.5}$ measurements are a perfect surrogate for exposure to PM of outdoor origin. When home-by-home variations in the AER were taken into consideration, the distribution of values of PM of outdoor origin was broadened

by 4%. Accounting also for home-by-home variations in particle properties and housing properties broadened the distribution by 10%. Spatial variations in outdoor $PM_{2.5}$ concentrations and interpersonal variations in the time spent in various microenvironments are also likely to broaden exposure distributions.

This work provides an assessment of some selected types of exposure error. Exposure errors result in larger uncertainties in relative risk factors for $PM_{2.5}$ than would occur if epidemiologic studies had more accurate exposure measures. Systematic variations in infiltration behavior (ie, with time in longitudinal studies, or spatially in chronic epidemiology) could, under certain circumstances, lead to misclassification of risk. Although this study focused on $PM_{2.5}$ mass, changes in particle composition with outdoor-to-indoor transport should inform studies that address the question: What is it about atmospheric PM that is responsible for the adverse health effects?

Some issues not explored in this work could introduce other types of errors and warrant further investigation. If concentrations of PM generated indoors and generated outdoors were covariant, the risk due to indoor PM sources could be improperly attributed to outdoor PM. Such covariance might occur, for example, if conditions contributing to high ambient PM concentrations alter behaviors in such a way as to increase concentrations of or exposure to PM generated indoors. The relation between ambient PM and exposure to PM of outdoor origin does appear to differ with location. For example, high outdoor temperatures were associated with elevated PM concentrations in all three study locations. However, high temperatures in Los Angeles typically lead to opening windows, which increased the infiltration of outdoor $PM_{2.5}$ into indoor environments and enhanced the impact of outdoor PM on exposure. In contrast, high temperatures in Houston increased air conditioning use and time spent indoors, which damped the impact of outdoor PM.

Another potential issue complicating the relation between central-site PM measurements and personal exposure is that changes in gas–particle partitioning, thermodynamics, and reactions in the indoor environment can alter PM of outdoor origin. Certainly, as discussed above, outdoor ozone can penetrate indoors and react with indoor alkene emissions, forming PM that is neither entirely of indoor nor entirely of outdoor origin. Also, Lunden and colleagues (2003) have shown that the equilibrium between gaseous nitric acid and particulate ammonium nitrate is disturbed indoors, leading to dramatic losses of particulate nitrate indoors. Nitrate was not measured in this study, although the species mass balance results for Los Angeles are consistent with the findings of Lunden and colleagues (2003). Gas–particle partitioning of organic compounds is also altered with changes in temperature and particle properties encountered during transport from outdoor to indoor spaces, as demonstrated above with PAHs. The effects of these transformations on PM epidemiology warrant further investigation.

SUMMARY

People are exposed to PM generated indoors, outdoors, in other microenvironments, and through personal activities. Particles generated through different mechanisms differ in composition and, presumably, toxicity. Recent epidemiologic studies using concentrations from central-site PM monitors as a surrogate for exposure to PM of outdoor origin report increased respiratory and cardiovascular morbidity and mortality with increased PM concentrations. (Norris et al 1999; Schwartz et al 1999, 2002; Klemm et al 2000; Goldberg et al 2001; EPA 2004). These epidemiologic findings provide motivation to characterize exposure to PM of outdoor origin and to explore the relation between central-site PM measurements and this quantity. Further motivation comes because strategies for mitigating exposures differ substantially for substances emitted into the outdoor environment and those generated indoors or in other microenvironments.

In the RIOPA study, the indoor, outdoor, and personal concentrations and composition of $PM_{2.5}$ and selected semivolatile species were measured and used to estimate the outdoor contributions to indoor concentrations and personal $PM_{2.5}$ exposure, and to provide a better mechanistic understanding of $PM_{2.5}$ exposure. The data analysis process yielded insights into the composition and sources of indoor-generated $PM_{2.5}$ and the personal cloud. Finally, implications for PM epidemiology were assessed. The results can be used to test and refine predictive models that link sources of particle emissions to exposures and to facilitate the development of effective strategies for protecting public health.

During the RIOPA study, 48-hour integrated indoor, outdoor, and personal $PM_{2.5}$ samples were collected in Elizabeth, Houston, and Los Angeles, between summer 1999 and spring 2001. Indoor and outdoor air samples suitable for gas-phase and particle-phase organic analyses were also collected. All samples were analyzed for $PM_{2.5}$ mass and functional groups. Subsets of samples were analyzed for $PM_{2.5}$ elements, OC, EC, and gas-phase and particle-phase PAHs and chlordanes. Volatile organic compounds, aldehydes, AER, temperature, and relative humidity were also measured (a 48-hour collection time was used to

improve quantitation of trace-level species). Questionnaire data and time-activity information for 309 participants were collected. The study goal of $PM_{2.5}$ sampling and species analysis in 150 homes was exceeded.

Median indoor, outdoor, and personal $PM_{2.5}$ mass concentrations were 14.4, 15.5, and 31.4 $\mu g/m^3$. Personal $PM_{2.5}$ concentrations were significantly greater and more variable than indoor and outdoor concentrations. Several approaches were applied to quantifying the distributions of $PM_{2.5}$ of outdoor and indoor origin, some using $PM_{2.5}$ mass concentrations and others using $PM_{2.5}$ species concentrations. Stepwise changes in the distributions with improved mechanistic assumptions were used to provide insights relevant to PM epidemiology. Estimates of PM of outdoor origin made with more accurate assumptions had broader distributions than would be obtained by using central-site data as a surrogate for PM exposure. Thus we conclude that using such a surrogate for PM exposure in epidemiologic studies will underestimate the actual distribution of exposures. This would lead to wider confidence intervals around relative risk factors than one would obtain if more accurate exposure measures were used, and would make it less likely to define a significant association between PM and health effects. The best estimate of the mean outdoor percentage of contribution to the indoor $PM_{2.5}$ mass concentration for homes was 73%. The outdoor contribution to the indoor $PM_{2.5}$ mass concentration was lower for Houston homes than for Elizabeth and Los Angeles homes, presumably because of the greater use of air conditioning in Houston. The mean outdoor contribution to personal exposure was estimated to be 26%.

Indoor and outdoor $PM_{2.5}$ species mass balances suggest that organic matter is the major constituent of $PM_{2.5}$ generated indoors. Organic matter (corrected for artifacts) constituted 48%, 55%, and 61% of $PM_{2.5}$ mass inside Los Angeles, Elizabeth, and Houston homes, respectively. At least 40% but probably closer to 75% of this organic matter, on average, was emitted or formed indoors. Functional group analysis, sampling artifact assessments, and analysis of the gas–particle partitioning of PAHs suggest that the composition and properties of the indoor-generated organic $PM_{2.5}$ differed substantially from that found outdoors. For example, indoor organic PM appeared to be a less favorable substrate for absorption of PAHs than outdoor organic matter on a per-mass basis. In addition, semivolatile organic species indoors showed a greater propensity for adsorption on the sampling QFF than those outdoors at the same concentration. Many indoor and personal samples showed strong aliphatic peaks and shifts in the carbonyl absorbance not found in outdoor $PM_{2.5}$. Some indoor and personal samples also contained amide. One likely source of particulate amide is cooking meat. Chlordane, a minor but mutagenic semivolatile organic mixture, was found to be mostly of indoor origin, with indoor source strengths on the order of 1500 ng/hr. Indoor sources of chlordane are likely to include volatilization from residues of termiticides applied indoors and infiltration from subsurface and foundation applications (typically several kilograms) during home construction. High emission rates were found most frequently in homes built from 1945 to 1959.

The change in gas–particle partitioning with transport of outdoor PAHs to the indoor environment was analyzed, and the results illustrate that chemical thermodynamics can alter the concentration and composition of outdoor PM as it is transported indoors. Although $PM_{2.5}$ nitrate was not measured, the study's species mass balance results provide indirect evidence that $PM_{2.5}$ nitrate is largely lost during outdoor-to-indoor transport, as previously reported for one home by Lunden and colleagues (2003). This could result in dramatic changes in the mass and composition of outdoor-generated $PM_{2.5}$ in California homes. The impact of such transformations on epidemiologic measurement error warrants further investigation.

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APPENDIX A. Initial Microwave Digestion Protocol

The microwave digestion protocol was initially performed without H_2O_2 and with a five-stage procedure that used power increments of 10% and a time duration of 10 min per stage. As described in the text section $PM_{2.5}$ Measurement and Quality Control / ICP–MS, an eight-stage digestion procedure was substituted. Tables A.1 and A.2 compare the percentages of recovery and the detection limits of both protocols, and Figure A.1 compares the percentages of samples above detection limits.

Table A.1. Average Percentage of Recovery Using Initial Digestion Protocol and Optimized Final Digestion Protocol

Element	Initial Protocol (%)	Final Protocol (%)
Ag	90.2	110.6
As	99.7	103.5
Ba	73.8	74.2
Cd	99.0	96.5
Co	74.0	78.0
Cr	22.1	26.2
Cs	61.9	68.6
Cu	91.6	109.4
Mn	92.3	104.0
Ni	82.6	127.5
Pb	78.5	73.4
Rb	38.8	74.8
Ti	22.9	23.3
U	77.2	75.0
V	73.2	89.0
Zn	85.0	105.2

Table A.2. Detection Limits Using Initial Digestion Protocol and Optimized Final Digestion Protocol

Element	Initial Protocol (ng)	Final Protocol (ng)
Ag	5.34	7.24
As	18.1	2.95
Ba	38.8	16.8
Be	14.3	3.00
Bi	3.46	3.27
Cd	27.9	5.61
Co	10.4	4.71
Cr	191	137
Cs	2.64	6.85
Cu	199	150
Ga	5.30	3.38
Mn	79.5	10.5
Ni	3391	1306
Pb	375	18.6
Rb	3.80	3.27
Se	9.97	21.1
Sr	64.8	38.1
Ti	449	27.6
Tl	3.13	2.76
U	3.43	3.53
V	118	4.34
Zn	3218	416

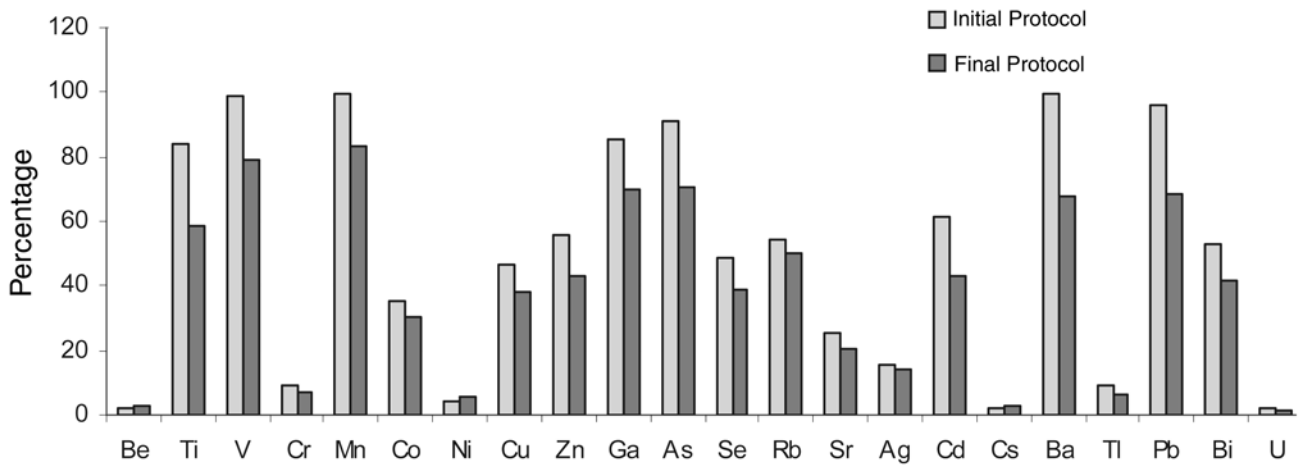

Figure A.1. Percentage of samples above detection limits (3 × SD of the field blank) as determined by initial digestion protocol and optimized final digestion protocol.

APPENDIX B. 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 and one remote audit 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 close-out 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. A final audit was performed to review the final database and check for consistency in calculations, derivations, and data-tracking.

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. Final reports for both the particulate and gaseous components of the study were reviewed by the auditors for completeness and accuracy. The Final Reports appear to be accurate representations of the study.

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
November–December, 2005	Remote audit of Turpin et al RIOPA PM database

APPENDIX AVAILABLE ON THE WEB

The following material is available on the HEI website *http://pubs.healtheffects.org*. They may also 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 C. Indoor, Outdoor, and Personal Concentrations of $PM_{2.5}$ Species by City and Season

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ABBREVIATIONS AND OTHER TERMS

AER	air exchange rate
ANOVA	analysis of variance
CAT	capillary absorption tube
CH	aliphatic hydrocarbon bond
CI	confidence interval
CV	coefficient of variation
DCM	dichloromethane
EC	elemental carbon
EPA	Environmental Protection Agency (US)
FID	flame ionization detector
FTIR	Fourier transform infrared (spectroscopy)
GC–MS	gas chromatography–mass spectrometry
HNO_3	nitric acid
H_2O_2	hydrogen peroxide
ICP–MS	inductively coupled plasma–mass spectrometry
MDL	method detection limit
MLR	multiple linear regression
MW	molecular weight
NAAQS	National Ambient Air Quality Standard
NIST	National Institute of Standards and Technology
NOPES	Nonoccupational Pesticide Exposure Study
NUATRC	National Urban Air Toxics Research Center
OC	organic carbon
OM	organic matter (OC × average organic molecular weight per carbon weight)
PAH	polycyclic aromatic hydrocarbon
PEM	personal environmental monitor
$PM_{2.5}$	particulate matter 2.5 μm or smaller in aerodynamic diameter
PMCH	perfluorinated methylcyclohexane
PMF	positive matrix factorization
ppb	parts per billion
PTEAM	Particle Total Exposure Assessment Methodology [study]
PUF	polyurethane foam
QFF	quartz fiber filter
r	correlation coefficient, bivariate analyses
r^2	coefficient of determination, bivariate analyses
R^2	coefficient of determination, multivariate analyses
RCS	random component superposition [statistical model]
RIOPA	Relationships of Indoor, Outdoor, and Personal Air
SD	standard deviation
SLR	single-parameter linear regression
Σchlordane	sum of chlordane masses or concentrations
ΣPAH	sum of PAH masses or concentrations
XRF	x-ray fluorescence

PAHs Measured

ACE	acenaphthene
ACY	acenaphthylene
ANT	anthracene
BaA	benzo[*a*]anthracene
BaFLR	benzo[a]fluorene
BaP	benzo[*a*]pyrene
BbFLR	benzo[b]fluorene
BeP	benzo[*e*]pyrene
BFLTs	benzo[*b*+*k*]fluoranthene
BghiP	benzo[*g,h,i*]perylene
BNT	benzo[*b*]naphtho[2,1-*d*]thiophene
CHR/Tr	chrysene and triphenylene
COR	coronene
CPP	cyclopenta[*cd*]pyrene
DBA	dibenzo[*a,c*+*a,h*]anthracene
DBT	dibenzothiophene
9,10-DMA	9,10-dimethylanthracene
3,6-DMP	3,6-dimethylphenanthrene
FLT	fluoranthene
FLR	fluorene
IP	indeno[1,2,3-*cd*]pyrene
1-MA	1-methylanthracene
2-MA	2-methylanthracene
9-MA	9-methylanthracene
1-MFL	1-methylfluorene
1-MP	1-methylphenanthrene
4,5-MP	4,5-methylenephenanthrene
NAP	naphthalene
PER	perylene
Phe	phenanthrene
PYR	pyrene
RET	retene

Chlordanes Measured

CC	*cis*-chlordane
CN	*cis*-nonachlor
MC5	a chlordane species
OXY	oxychlordane
TC	*trans*-chlordane
TN	*trans*-nonachlor

Equation Terms

a	air exchange rate
α	attenuation factor
A	PAH concentration on the adsorbent (gas phase)
C_{In}	indoor $PM_{2.5}$ concentration
C_{Out}	outdoor $PM_{2.5}$ concentration
C_{IgI}	indoor $PM_{2.5}$ concentration generated indoors
C_{IgO}	indoor $PM_{2.5}$ concentration generated outdoors
E_t	time-averaged personal exposure to $PM_{2.5}$
E_{gO}	time-averaged personal exposure to $PM_{2.5}$ generated outdoors
E_{gI}	time-averaged personal exposure to $PM_{2.5}$ generated indoors
$\phi_{2.5}$	fraction of PAHs associated with $PM_{2.5}$ (particle phase)
$F_{2.5}$	PAH concentration on the $PM_{2.5}$ QFF (particle phase)
f_{EC}	fraction of elemental carbon in $PM_{2.5}$ mass
F_{Inf}	infiltration factor; fraction of outdoor $PM_{2.5}$ that is found indoors
f_{OC}	fraction of organic carbon in $PM_{2.5}$ mass
k	loss rate coefficient
K_p	gas–particle partition coefficient
$\log K_{p,meas}$	log of measured gas–particle partition coefficient
$\log K_{p,meas,SD}$	log of $K_{p,meas}$ adjusted to temperature = 25°C
P	penetration coefficient
p_L°	subcooled liquid vapor pressure
Q_{In}	indoor source strength or emission rate
V	home volume
y	fraction of time subject spends indoors

COMMENTARY
Special Review Panel

INTRODUCTION

Particulate matter (PM*) is a complex mixture of particles that vary in size and composition and are generated by combustion, atmospheric reaction, and mechanical processes. Epidemiologic and animal studies have shown associations between exposure to PM and a variety of adverse health effects (reviewed in Leikauf 1992; US Environmental Protection Agency [EPA] 1993; Heseltine et al 1993; Snyder 2000; Delfino et al 2003; EPA 2004; Schlesinger et al 2006). Because of concerns about health effects, the EPA regulates ambient concentrations of fine PM (smaller than 2.5 μm in aerodynamic diameter [$PM_{2.5}$]) through the National Ambient Air Quality Standards (NAAQS) (EPA 1997a) and emissions of PM from mobile and stationary sources. At present, regulations are based on the mass (weight) of particles and do not take into account particle composition, which depends on the sources. More detailed information on composition is needed to help determine whether certain PM components are more strongly associated with adverse health outcomes than the conventional measure of PM mass.

SIZE AND COMPONENTS OF PM

Particle size is generally classified by aerodynamic diameter into coarse (>2.5–10 μm), fine (0.1–2.5 μm), and ultrafine (< 0.1 μm) fractions. The most common indicator of fine particles is $PM_{2.5}$. Depending upon sources and the changes they undergo in the atmosphere, particles also vary in chemical composition and other physical, chemical, and biological properties and are not uniform among geographic regions with different sources, climates, and topography. These geographic, size, and compositional considerations could explain some of the discrepancies among results from epidemiologic studies (Schwartz et al 1996; Fairley 1999; Burnett et al 2000; Castillejos et al 2000; Gwynn et al 2000; Hoek et al 2000; Ostro et al 2000).

Fine particles are derived mainly from direct emissions from combustion processes, such as diesel and gasoline engines, burning wood, and burning coal for power generation and industrial processes (such as smelters, cement plants, and paper mills). They are mostly composed of sulfate and nitrate particles, which are generated through conversion of primary sulfur and nitrogen oxide emissions, and secondary organic aerosols from semivolatile organic compound emissions. PM also contains organic and elemental carbon (OC and EC), ammonium ions, and various transition elements.

Fine particles differ from coarse particles because they remain suspended in the air for longer periods of time, penetrate more easily into indoor environments, and are transported over long distances. They are of particular health concern because they can penetrate deeper into the lung and are retained longer in the alveolar region (Schwartz and Neas 2000). Furthermore, they may be more toxic because they consist of sulfates, nitrates, acids, and metals, including transition metals.

HEALTH EFFECTS OF EXPOSURE TO PM

Recent research has provided new insight into how PM may exert its effects through biological and chemical mechanisms of action. These include exacerbation of existing pulmonary disease, oxidative stress, and inflammation; changes in cardiac autonomic function; alterations in vasculature; translocation of PM across internal biological barriers; reduced defense mechanisms; and lung damage (Pope and Dockery 2006).

Whereas earlier research focused on respiratory effects of PM exposure, research in the past decade has focused on the observed association between PM inhalation and cardiovascular effects, including cardiac irregularities and the onset of myocardial infarction (Pope and Dockery 2006). Both animal studies (in which animals inhale particles or particles are instilled into the lungs) and human studies have revealed direct alterations in cardiac autonomic functions (including decreased heart rate variability) in response to PM exposures (Watkinson et al 1998; Godleski et al 2000; Gold et al 2000; Schwartz et al 2001).

A recent review (Pope and Dockery 2006) notes that epidemiologic studies over the past 10 years show statistically significant associations between cardiopulmonary mortality and daily concentrations of $PM_{2.5}$ and PM_{10}. Long-term exposures result in larger effects than short-term exposures across all studies. The health effects of PM appear to depend on both the concentrations and the duration of exposure: Repeated or prolonged exposures to high concentrations over many years have, to date, shown higher associations with adverse effects than less frequent, short-term exposures to high concentrations.

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

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Some components of $PM_{2.5}$ have been well studied because of their potentially toxic effects—specifically, soluble transition metals (Dreher et al 1996; Costa and Dreher 1997). Several studies in humans and other species have identified a possible role of metals in inducing PM-related effects (Schlesinger et al 2006). Short-term exposure of rodents to high concentrations of nickel and vanadium or of residual oil fly ash induced inflammatory, respiratory, and cardiovascular responses, including cardiac arrhythmias (Watkinson et al 1998; Campen et al 2001). (Residual oil fly ash is an emission from power plants that is rich in particles containing metals, especially iron, nickel, and vanadium. The concentrations and proportions of metals are much higher than those found in ambient air.) Another study (Ghio and Devlin 2001) found that particles collected when metal concentrations (specifically iron, copper, zinc, lead, and nickel) were high induced a greater inflammatory response in human lungs than when metal levels were low.

Ambient air also contains many different organic compounds associated with combustion particles. However, with the exception of diesel exhaust, much less research has been conducted to investigate the health effects of these compounds. Diesel exhaust particles are reported to enhance the induction of at least some characteristics of the allergic response in humans and other species (Muranaka et al 1986; Diaz-Sanchez et al 1996). Some in vitro studies have shown that an organic fraction extracted from these particles enhances the synthesis of immunoglobulin E, a key mediator of the allergic response (Takenaka et al 1995; Tsien et al 1997). In addition, a similar organic extract of diesel exhaust particles has been reported to have cytotoxic effects in macrophages and epithelial cells in vitro (Nel et al 2001).

EXPOSURE TO PM

Although many epidemiologic studies have shown an association between exposure to PM and increased morbidity and mortality (EPA 2004), a lack of information on important factors that may influence exposure complicates interpreting this research, assessing human risk, and designing control strategies. In 1997, the EPA promulgated new NAAQS for PM, which included 24-hour and yearly standards for $PM_{2.5}$ (EPA 1997a). In 2006, the EPA reviewed these NAAQS and retained the annual standard and tightened the 24-hour standard. The NAAQS are based on measurements of $PM_{2.5}$ taken at defined outdoor monitoring sites in the United States; the extent to which these ambient (outdoor) measurements can be used as an adequate surrogate for personal exposure has been an important research and policy question.

An important step, therefore, toward understanding the health effects is to characterize personal exposure to PM and its components. Personal exposure includes exposure experienced outdoors and in all the different microenvironments (eg, residential dwellings, workplaces, public buildings, traffic) where people spend their time. Exposures may vary substantially due to housing characteristics, behavioral factors (such as smoking habits, exercise, and cooking and cleaning activities), proximity to sources, and time spent in different locations. Because obtaining direct measurements of personal exposure is complex and very costly, however, an exposure surrogate for personal PM exposure—usually the outdoor concentrations measured at fixed-site monitors—is used by researchers and policymakers.

Results from air pollution exposure and epidemiologic assessment studies suggest that measurements of ambient fine particles (but not gases) are strong proxies of corresponding personal exposures. However, the strength of the association between personal exposure and ambient particle concentrations varies according to particle composition and outdoor sources, household characteristics such as home ventilation (Sarnat et al 2006), and the strength of indoor sources (Brunekeef et al 2005; Janssen et al 2005).

When HEI and the Mickey Leland National Urban Air Toxics Research Center (NUATRC) issued their respective Requests for Applications (RFA 98-01 and RFA 96-01), the overall objectives were to (1) characterize personal exposure to particles in different indoor and outdoor microenvironments and in geographic locations that differ in types and sources of particles, topography, and climate; and (2) identify distinctive characteristics of particles that would improve exposure estimates in epidemiologic studies. Ideally, studies to address the second objective would determine particle characteristics (eg, concentration, size, and composition) and describe the relation between overall personal exposure and the surrogate measures of exposure that are typically used in epidemiologic time-series studies. These studies would address questions about similarities or differences in levels and characteristics of particles in various microenvironments, how outdoor particles contribute to indoor concentrations, and how individual activity patterns influence personal exposure and the resulting dose.

The overall aim of the Relationships of Indoor, Outdoor, and Personal Air (RIOPA) study was to examine the influence of outdoor sources on indoor concentrations and personal exposures for a suite of volatile organic compounds, aldehydes, and $PM_{2.5}$ in three cities with different air source profiles: Los Angeles CA, Houston TX, and Elizabeth NJ. A unique feature of the study plan was that it would provide a speciation profile of $PM_{2.5}$; that is, an estimate of chemical

composition including the quantification of key PM constituents such as trace elements, sulfate, nitrate, and carbon.

The project was jointly funded and overseen by HEI and NUATRC. 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 number of analyses conducted, the Investigators' Final Report is presented in two parts: *Part I. Collection Methods and Descriptive Analyses* (for volatile organic compounds, carbonyls, and $PM_{2.5}$ concentrations; Weisel et al 2005) and *Part II. Analyses of Concentrations of Particulate Matter Species* (this volume and the subject of this Commentary).†

The Commentary is intended to place the research into broader scientific and regulatory context, highlight the strengths and limitations of the study, point out interpretations of the results in addition to those presented by the investigators, and discuss remaining uncertainties and implications of the findings for public health.

TECHNICAL EVALUATION

AIMS AND OBJECTIVES

Dr Turpin and her colleagues measured concentrations and composition of $PM_{2.5}$ in indoor, outdoor, and personal samples collected for 48 hours. This component of the RIOPA study aimed to: (1) characterize and compare indoor, outdoor, and personal $PM_{2.5}$ mass composition; (2) estimate the contribution of outdoor $PM_{2.5}$ and its components to indoor concentrations and personal exposures using residential air exchange rates (AERs); and (3) conduct initial source apportionment analyses of indoor and personal exposure $PM_{2.5}$ concentrations.

Measurements were conducted over all seasons in three US cities with high concentrations of $PM_{2.5}$ and different outdoor source profiles that included emissions from petrochemical industries, roadways, and commercial establishments.

HOUSEHOLD AND SUBJECT SELECTION

The RIOPA study was designed to address the hypothesis that outdoor sources contribute a substantial proportion of the pollutant concentrations in the indoor air and personal air (breathing zone) for residents who live near those sources. The study included approximately 100 homes selected because of their proximity to outdoor sources, and 100 residents of those homes, in each of three urban centers with different air pollution source profiles and weather conditions: Los Angeles CA is dominated by mobile sources (several major freeways). Houston TX is dominated by large industrial point and area sources (petrochemical complexes) and mobile sources (major freeways). Elizabeth NJ has a mixture of mobile, point, and area sources (state turnpikes, industrial complexes, incinerator, airport, shipyard, and small commercial enterprises such as gasoline stations, dry cleaners, and small factories). The selection of homes and subjects was not designed to provide a population-based sample. Rather, homes close to outdoor sources were preferentially selected so the contribution of potentially high ambient sources could be explored.

Target geographic areas in each city were selected because of their relative proximity to known sources of air pollution. Areas included were close to and far from interstate highways in Los Angeles, industrial zones of Houston, and dense commercial traffic in Elizabeth. Study subjects were recruited (in Spanish and English) via phone calls, household visits, direct mail, religious or community leaders, and word-of-mouth. Selection criteria were that no smokers lived in the home, one adult would be at home for more than 10 hours per day, and the subject did not plan to move within the next 3 months. Questionnaires were used to gather demographic data, exposure history, and information on the activities of the participants, as well as the characteristics of their homes and nearby sources. Most participants were adults, although children in some homes were also recruited (especially in Elizabeth). Because of the small numbers of children, however, specific results were not presented in great detail in the Investigators' Report. Informed consent was obtained for each participant.

† 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. (See also *Part I. Collection Methods and Descriptive Analyses* [Drs Weisel, Zhang, and Turpin] for a complete presentation of study design, data collection, and analysis of volatile organic compounds, aldehydes, and particulate matter.)

Dr. Turpin's 3-year study, "Contributions of Outdoor PM Sources to Indoor Concentrations and Personal Exposures: A Three-City Study", started in July 1998 with a 3-month pilot study to evaluate the proposed participant recruitment strategy, sampling methods, and analytical techniques. The contract for the 2-year, 9-month main study began in October 1998. The study was later extended by 3 months to include a report writing period. Total expenditures were $1,002,460. The draft Investigators' Report from Dr Turpin and colleagues was received for review in February 2004. After the external reviewers and the HEI/NUATRC Special Review Panel reviewed the report, a revised report was received in December 2004 and was accepted for publication in January 2005. 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 the Commentary.

Additional details on sample selection are provided in Part I of this Research Report (Weisel et al 2005).

EXPOSURE ASSESSMENT METHODS

$PM_{2.5}$ Mass

To measure $PM_{2.5}$ mass, functional groups, and elements, samples were collected on Teflon filters mounted in a Harvard impactor (flow rate of 10 L/min) placed inside and directly outside of each home. Personal samples were collected on smaller Teflon filters mounted in the personal environmental monitor (PEM) worn by each participant. The PEM is a lightweight sampler with a $PM_{2.5}$ size-selective impactor inlet that samples at a flow rate of 3.2 L/min. Harvard impactors and PEMs were collocated to determine agreement between the two types of samplers. All filters were weighed in an EPA-audited laboratory at the Environmental and Occupational Health Sciences Institute according to EPA protocols.

OC, EC, and Trace-Level Organic Compounds

To analyze the carbonaceous particle components, $PM_{2.5}$ samples were collected indoors and outdoors concurrently using a modified MSP microenvironmental $PM_{2.5}$ sampler (MSP Co, Minneapolis MN) operating at a flow rate of 10 L/min. This sampler was modified to hold a polyurethane foam (PUF) adsorbent for collecting vapor-phase semivolatile organic compounds. The PUF was placed downstream of a multiple-jet impactor inlet with a 2.5-μm aerodynamic diameter cut-point and a 37-mm quartz fiber filter (QFF).

QFFs typically collect particulate carbon with approximately 100% efficiency; however, the filter surface also adsorbs some organic vapors, which results in an overestimate of OC concentrations (known as an adsorption artifact). To determine the magnitude of the adsorption artifact, the Harvard impactor was modified to hold a backup QFF located downstream of the Teflon filter. By comparing the organic vapor adsorbed on both QFFs, the investigators were able to correct for the organic vapors adsorbed to filter surfaces.

DETERMINING AERs

AERs are expressed as the number of indoor air volumes replaced each hour by outdoor air. They were measured using a technique developed specifically for application to relatively small spaces, including homes. A tracer gas, perfluorinated methylcyclohexane (PMCH), is emitted inside the home and then collected on a passive capillary absorption tube. By increasing the source strength of the tracer gas, up to five air exchanges per hour can be detected. The investigators measured the number of air exchanges in each home during the two seasons when the air sampling was conducted in that home.

SAMPLE ANALYSIS

Samples or subsets of samples were analyzed for $PM_{2.5}$ mass, elements, OC and EC, functional groups, polycyclic aromatic hydrocarbons (PAHs), and chlordanes using the methods briefly summarized in Commentary Table 1.

DATA QUALITY

Sample Validation

Samples were considered valid if the flow rate had changed less than 15% during sampling and if the collection time exceeded 42 hours (87.5% of target duration). Samples were invalidated if field comments suggested, for example, that the equipment malfunctioned or the subject had not worn the personal monitor. For $PM_{2.5}$ samples collected on Teflon filters, 91%, 82%, and 83% were valid in Los Angeles, Elizabeth, and Houston, respectively; 91%, 94%, and 94% of samples collected on QFFs were valid.

Quality Control

The investigators conducted field tests and laboratory analyses to evaluate the performance of the samplers and the analytical methods used in the study. They estimated the method detection limit (MDL; the minimum concentration of a compound that can be measured and reported with 99% confidence), analytical precision (variation in the analytical method under constant conditions), measurement precision (variation of the analyses of individual species from collocated field samplers), and analytical accuracy (ability of the method or sampler to correctly measure the "true" amount of a species or an accepted reference value).

DATA ANALYSIS

Comparing Indoor, Outdoor, and Personal Concentrations

Comparisons between subpopulations were made on the original or log-transformed data, as appropriate. Scatter plots were also used to provide a visual estimate of the relations between concentrations from different sources. Where more than 40% of the data were below detection limits, only graphic or descriptive analyses were conducted.

Commentary Table 1. Summary of Methods for Sample Analysis

Compounds	Methods	Comments
$PM_{2.5}$ mass	EPA protocols for $PM_{2.5}$ mass	Teflon filters were weighed on a microbalance (C-30, Cahn Instrument Inc, Cerritos CA; MT5, Mettler Toledo Inc, Columbus OH) in an EPA-audited laboratory.
Organic and elemental carbon (OC and EC)	Thermal-optical transmittance in a Carbon Analyzer (Sunset Laboratory)	Correction for pyrolytic conversion of OC to EC during analysis was made by monitoring the transmittance of light through the filter with a diode laser and a photodetector.
36 Elements: Ag, Al, As, Ba, Br, Ca, Cd, Cl, Co, Cr, Cu, Fe, Ga, Ge, Hg, In, K, La, Mn, Mo, Ni, P, Pb, Pd, Rb, S, Sb, Se, Si, Sn, Sr, Ti, V, Y, Zn, and Zr	X-ray fluorescence (XRF) spectrometry	
22 Elements: Ag, As, Ba, Be, Bi, Cd, Co, Cr, Cs, Cu, Ga, Mn, Ni, Pb, Rb, Se, Sr, Ti, Tl, U, V, and Zn	Inductively coupled plasma–mass spectrometry (ICP–MS)	
Functional groups	Fourier transform infrared (FTIR) spectroscopy	To the best of our knowledge, this was the first use of FTIR spectroscopy in a $PM_{2.5}$ exposure study.
30 PAHs and 6 chlordanes	Gas chromatography–mass spectrometry (GC–MS)	Gas- and particle-phase species were collected on a polyurethane foam and quartz fiber filter, respectively.

The indoor, outdoor, and personal concentrations were compared within a home by using an incomplete randomized block model. Multiple measurements from the same household were made at least three months apart and showed very little correlation. In light of this result, measurements made in the same home were treated independently.

Quantifying the Outdoor Contribution to Indoor $PM_{2.5}$ Concentrations

Indoor concentrations are a sum of concentrations resulting from outdoor and indoor sources. At a steady state, the indoor $PM_{2.5}$ mass equation can be described with a single-compartment mass balance model:

$$C_{In} = \frac{PaC_{Out}}{a+k} + \frac{Q_{In}/V}{a+k} = F_{Inf}C_{Out} + C_{IgI} = C_{IgO} + C_{IgI},$$

where C_{In} and C_{Out} are $PM_{2.5}$ concentrations ($\mu g/m^3$) measured indoors and outdoors, P is penetration coefficient (dimensionless), a is AER (hr^{-1}), k is particle loss rate (hr^{-1}), Q_{In} is indoor source strength or emission rate ($\mu g/hr$), V is home volume (m^3), F_{Inf} is infiltration factor (dimensionless), C_{IgI} is indoor $PM_{2.5}$ concentrations ($\mu g/m^3$) generated indoors, and C_{IgO} is indoor $PM_{2.5}$ concentrations ($\mu g/m^3$) generated outdoors. The AER, penetration, decay rate, and infiltration factor (a function of penetration and particle loss [or decay] rate) may differ from home to home and from species to species.

The investigators calculated the outdoor contribution to indoor $PM_{2.5}$ mass using three methods with increasingly more realistic assumptions:

1. A random component superposition (RCS) model, which assumes that the infiltration factor is constant across homes. This would be the case if fixed-site measurements of $PM_{2.5}$ were a perfect surrogate for $PM_{2.5}$ of outdoor origin.
2. A mass balance model, which assumes that the infiltration factor varies according to measured AERs for each home; and
3. A robust regression approach, which uses all measured $PM_{2.5}$ species to calculate the infiltration factor for each home. Specifically, indoor species concentrations were regressed on concurrently collected outdoor species concentrations using least-trimmed squared regression (S-Plus, Insightful, Seattle WA) to obtain a $PM_{2.5}$ infiltration factor.

Gas–Particle Partitioning of PAHs

Data from the paired indoor and outdoor air samples were used to examine changes in gas–particle partitioning of PAHs between indoor and outdoor environments. The effects of temperature and aerosol composition were examined using stepwise multiple linear regression.

RESULTS

Commentary Table 2 presents a selection of summary statistics for pollutant concentrations.

Indoor, Outdoor, and Personal $PM_{2.5}$ Mass Concentrations

Combined across all three cities, the median mass concentrations of $PM_{2.5}$ for indoor, outdoor, and personal exposure were 14.4, 15.5, and 31.4 μg/m^3. On average, personal exposures exceeded indoor concentrations by 17 μg/m^3.

Outdoor and indoor concentrations differed little among the cities, and indoor and outdoor levels in each city were similar. Indoor concentrations for Houston homes were more variable than outdoor concentrations. (See Commentary Table 2.)

Pooled indoor, outdoor, and personal $PM_{2.5}$ concentrations were only poorly to moderately interrelated: 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. This difference in ratios could reflect variations in indoor source strength, AERs, and personal activity. Correlations were much stronger when the ratio of indoor to outdoor mass concentration was < 1, presumably because of low indoor source strengths, high AERs, or both within these homes. The degree of correlation between indoor and outdoor concentrations did not have much impact on correlations with personal $PM_{2.5}$ concentrations.

$PM_{2.5}$ Composition

Organic PM constituted approximately one-half of indoor $PM_{2.5}$ concentrations and approximately one-third of outdoor concentrations. Comparing indoors with outdoors, the mean organic PM concentration indoors was nearly twice that outdoors; this ratio indicates the importance of indoor sources of organic PM.

This was also demonstrated when the RCS model was used to estimate the mean contributions of indoor and outdoor sources to indoor OC concentrations: on average, 69% of indoor OC was emitted or formed indoors.

For each species other than organic PM, the indoor concentration and percentage it contributed to the total $PM_{2.5}$ concentration were the same as or less than outdoor levels.

OC and EC EC concentrations indoors and outdoors were well correlated, with outdoor concentrations being higher than indoor concentrations. This suggests that most EC penetrated from outdoors and indoor emissions of EC were low (see Commentary Table 2). In comparison, many homes seemed to have substantial indoor sources of OC. The fact that the OC/EC ratio was stronger indoors than outdoors is consistent with this observation.

Commentary Table 2. Selected Pollutant Concentrations in Outdoor, Indoor, and Personal Air, by City[a]

	Los Angeles				Elizabeth				Houston			
	n	Mean	SD	Median	*n*	Mean	SD	Median	*n*	Mean	SD	Median
$PM_{2.5}$ mass concentration (μg/m^3)												
Outdoor	121	19.2	13.3	16.1	103	20.4	10.7	18.2	110	14.7	5.75	13.2
Indoor	124	16.2	9.38	14.5	96	20.1	15.5	15.7	106	17.1	12.7	13.4
Personal adult	105	29.2	14.8	26.5	77	44.8	29.9	37.4	98	37.2	23.8	31.6
Personal child	1	40.2	—	40.2	23	54.0	32.0	39.2	3	36.6	7.97	39.1
Elemental carbon (μg/m^3)												
Outdoor	44	1.4	0.9	1.2	59	1.4	0.6	1.3	69	0.7	0.3	0.7
Indoor	44	1.3	0.8	1.1	59	1.4	1.3	1.1	69	0.7	0.8	0.5
Organic carbon (μg/m^3)												
Outdoor	44	4.1	1.9	3.6	59	3.3	1.7	3.0	69	3.2	3.1	2.3
Indoor	44	5.4	3.4	4.7	59	7.9	9.1	5.4	69	7.2	5.4	5.4

[a] *n* = number of samples.

Functional Groups Functional groups are elemental structures attached to carbon that can influence a molecule's behavior. Analyses revealed sulfate, ammonium, nitrate, silicate, carbonyl, organic nitrate, amide, and aliphatic groups. The presence of particle-phase aliphatic aldehydes, ketones, and carboxylic acids (all of which absorb in the wave number/cm range of 1710 to 1730 in spectroscopic analysis) is suggested by a carbonyl absorbance that commonly peaks near 1720/cm. High wave number carbonyl peaks appeared almost exclusively in indoor and personal samples, which indicates the presence of more complex carbonyl-containing compounds, such as cyclic or aromatic ketones and halogenated carbonyls.

Absorbances that correspond to the stretching of aliphatic hydrocarbon (CH) bonds were clearly distinguished in some spectra and are represented by the sharp doublet of peaks near 2900/cm. Strong CH absorbances were found in 57% and 59% of indoor and personal samples, respectively, suggesting that PM emissions within many homes had a strong aliphatic character. Only 3% of outdoor samples, however, had strong CH absorbances; this finding is consistent with other studies in which strong CH absorbances have rarely been identified in outdoor samples of fine particles.

The spectra obtained were grouped into four categories on the basis of strong or weak aliphatic hydrocarbon absorbance and the presence or absence of amide. Of the outdoor $PM_{2.5}$ spectra, 97% had weak CH absorbance and no detectable amide absorbance. Of these homes, 44% also showed the same pattern in indoor spectra. The remaining 56% of homes had indoor organic PM sources that substantially altered the composition of $PM_{2.5}$ exposures: 49% had strong aliphatic absorbances in the indoor spectrum, and 12% had detectable amide absorbances.

PAHs The sum of masses for all measured PAHs is designated as ΣPAH mass. The ΣPAH mass concentrations in residential outdoor samples ranged from 1.5 to 64 ng/m^3, 12 to 200 ng/m^3, 10 to 160 ng/m^3 in Los Angeles, Elizabeth, and Houston, respectively. The variability in the PAH concentrations was substantially larger indoors than outdoors: indoor concentrations ranged from 7.0 to 220 ng/m^3, 19 to 350 ng/m^3, and 3.1 to 310 ng/m^3 in Los Angeles, Elizabeth, and Houston respectively. Phenanthrene was the largest contributor to mass in the outdoor and indoor air in each city (note, however, that naphthalene, typically a large contributor to ΣPAH mass, had to be excluded from their dataset due to sampling complications). Gaseous compounds were the driving factor of the variability in PAH concentrations because they made up, on average, 90% to 97% of total (gas-phase + particle-phase) PAH mass.

The mean PAH concentration profiles outside study homes in each city were significantly different from each other, which is likely to be due to different emission profiles and temperature-driven differences in gas–particle distribution. The lowest gas-phase mean ΣPAH concentration was in Los Angeles samples, and the lowest particle-phase mean ΣPAH concentration was in Houston samples.

High-molecular-weight PAHs, found mostly in the particle phase, seemed to be dominated by outdoor sources. In addition, profiles of 5- to 7-ring PAHs in the outdoor air suggest different dominant PAH sources in each of the three cities. Indoor sources were important contributors to low-molecular-weight PAH concentrations.

Gas–Particle Partitioning of PAHs PAHs seemed to equilibrate rapidly between the gas and particle phases, as suggested by high correlations between the log of the measured gas–particle partition coefficient and the log of the subcooled liquid vapor pressure. PAHs in the outdoor air were close to equilibrium, and PAHs transported from outdoor to indoor air rapidly approached new equilibrium with indoor emissions and conditions.

Differences in gas–particle partitioning of PAHs within each city were on the same order of magnitude as differences between the cities.

Vapor pressure was the most important predictor of partition coefficient, followed by temperature. Vapor pressure and temperature exhibited negative effects; that is, an increase in either variable was associated with a decrease in the thermodynamic partition coefficient and the fraction of the PAH in the particle phase. On the other hand, increases in EC and OC fractions in $PM_{2.5}$ mass exhibited positive effects: They were associated with increased partitioning to the particle phase.

Chlordanes Chlordanes are components of Technical Chlordane, which was widely used as a pesticide and termiticide in North America from the 1940s through the 1960s. Chlordane is a likely carcinogen by all routes of exposure (EPA 1997b). Its use as a broad-spectrum pesticide was regulated starting in 1974, but it was still being used as a termiticide in indoor environments in the 1990s.

The total (gas phase + particle phase) geometric mean concentrations of chlordanes in the outdoor samples ranged from 36 to 4270 pg/m^3 in Los Angeles, from 8 to 11,000 pg/m^3 in Elizabeth, and from 62 to 1770 pg/m^3 in Houston. The corresponding indoor total chlordane concentrations ranged from 37 to 111,500 pg/m^3 in Los Angeles, from 260 to 31,800 pg/m^3 in Elizabeth, and 410 to 38,900 pg/m^3 in Houston. Concentrations were higher in

indoor air than outdoor air in all three cities (1980 vs 580 pg/m^3 in Los Angeles; 1300 vs 170 pg/m^3 in Elizabeth; 4180 vs 280 pg/m^3 in Houston). The outdoor total chlordane concentrations were not significantly different among the three cities.

For 99 out of 108 homes with paired indoor and outdoor total (gas phase + particle phase) chlordane concentrations, the indoor concentration was greater than the outdoor concentration. For 103 out of 112 homes with paired indoor and outdoor concentrations above the MDL for gas and particle phases, the indoor concentration of *trans*-chlordane (a stereoisomer of chlordane) exceeded the outdoor concentration. Of these 103 homes, the indoor/outdoor ratio for *trans*-chlordane at 95 homes was greater than 2, and at 46 homes it was greater than 10. Variations in the chlordane concentrations in the outdoor and indoor samples were driven by gaseous chlordane species, which comprised approximately 90% of the chlordane mass measured in the samples.

Elements The elemental concentrations, used to construct indoor and outdoor species mass balances for $PM_{2.5}$ and to obtain home-specific estimates of infiltration factors, were presented in tabular form. Summary statistics for indoor, outdoor, and personal (adult) concentrations were provided for each element by state. (See Appendix C to the Investigators' Report, which is available on request.)

Outdoor Contribution to Indoor and Personal Concentrations: Comparison of Analytical Approaches

For all homes combined, the penetration coefficient was estimated by robust regression analysis to be 0.91 (95% confidence interval [CI], 0.71–1.12) and the particle loss rate to be 0.79/hr (95% CI, 0.18–1.41]. For 114 homes, paired indoor and outdoor $PM_{2.5}$ species measurements were obtained. Using the robust regression approach, the median infiltration factor estimated from these homes was 0.70 with an SD of 0.23.

Outdoor Contribution to Indoor $PM_{2.5}$ Concentrations

The mean contribution of outdoor sources to indoor $PM_{2.5}$ mass concentrations estimated using the mass balance model was 8.7 μg/m^3 or 60% for all study homes (67%, 70%, and 41% for Los Angeles, Elizabeth, and Houston homes, respectively; see Commentary Table 3). Using the robust regression approach, the percentage of indoor $PM_{2.5}$ that originated outdoors had a mean, median, and SD of 73%, 74%, and 36%, respectively. This regression method is able to provide an estimate of the outdoor contribution for each measured home on the day it was sampled. The two methods produced broadly consistent results, although the robust regression approach estimates were somewhat higher.

Outdoor Contribution to Personal $PM_{2.5}$ Exposure

Comparisons between the results from the RCS and mass balance models showed how the distribution of $PM_{2.5}$ of

Commentary Table 3. Outdoor and Indoor Contributions of $PM_{2.5}$ Mass to Indoor Concentration and Personal Exposure, by City

	Los Angeles	Elizabeth	Houston
Air Exchange Rate (1 exchange/hr)[a]			
Mean, Median (SD)	1.22, 0.93 (0.87)	1.22, 0.88 (0.97)	0.71, 0.46 (0.73)
Outdoor Contribution (%)			
To indoor concentrations			
Mass balance model	67	70	41
To personal exposure			
Mass balance model	33	22	21
Random component superposition model	33	33	13
Indoor Contribution (% by mass balance model)			
To indoor concentrations	33.4	29.7	59.0
To personal exposure	66.8	77.5	78.5

[a] The number of indoor air volumes replaced each hour by outdoor air.

outdoor origin changed when actual variations in AERs were taken into consideration. The mean outdoor contribution to personal $PM_{2.5}$ exposure estimated using the RCS model was 25% for all study homes (33%, 33%, and 13% for Los Angeles, Elizabeth, and Houston homes, respectively; see Commentary Table 3). Similarly, the mass balance model estimated values of 26% for all study homes (33%, 22%, and 21% for Los Angeles, Elizabeth, and Houston homes, respectively). Here too, the methods produced broadly consistent results.

Comparisons between the results from the mass balance model and the robust regression approach showed how the distribution of $PM_{2.5}$ of outdoor origin changed when the infiltration factor was allowed to vary from home to home and day to day. The distribution appeared to be more sensitive to variations in AER than to other potential influences on infiltration, such as particle and housing characteristics. When the AER was very low, the decay rate had a large inverse influence on the infiltration factor. For example, at 1 air exchange per hour, changing the decay rate from 0.79/hr to 0.4/hr changed the infiltration factor from approximately 55% to almost 70%.

DISCUSSION

$PM_{2.5}$ CONCENTRATIONS

Differences Among Cities

On the basis of $PM_{2.5}$ measurements, outdoor and indoor particle concentrations differed little within each city and among all cities. In a study of the homes of children with asthma in seven US cities, Wallace and coworkers (2003) also found 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 among cities.

Unlike outdoor and indoor measurements in the RIOPA study, both personal exposure concentrations of $PM_{2.5}$ and the ratio of personal to outdoor concentrations varied among the three cities. Adult personal exposures (with means of 29.2 $\mu g/m^3$ in Los Angeles, 44.8 $\mu g/m^3$ in Elizabeth, and 37.2 $\mu g/m^3$ in Houston) were greater and more variable than outdoor and indoor concentrations. The ratio of personal exposure to outdoor median concentrations ranged from 1.6 in Los Angeles to 2.3 in Elizabeth and 2.4 in Houston. These ratios were similar to those measured in other studies of large groups of adult nonsmokers in Toronto (Pellizzari et al 1999) and in Indianapolis (Pellizzari et al 2001). (The subjects in the RIOPA study did not smoke; information about their possible exposure to environmental tobacco smoke was collected on questionnaires but was not addressed in the study.)

Differences in climate could contribute to regional variations in housing characteristics and sources of pollutants. For example, the prevalence of air conditioning use, open windows, and burning wood indoors could be driven by weather conditions; and wind direction could affect how pollutants travel from outdoor sources. Data from homes in all three cities were combined for this analysis, however, which may have masked any differences among cities related to climate.

Mean and median outdoor contributions to indoor and personal concentrations were much lower for Houston homes than for those in Los Angeles and Elizabeth. The authors suggest this disparity could be attributed to the higher prevalence in Houston of air conditioning, which tends to reduce AERs. This is consistent with findings from other studies that suggest the infiltration of ambient $PM_{2.5}$ indoors is lower in homes with air conditioning than in homes without (Suh et al 1992). Although the investigators collected the necessary data on air conditioning use, they did not test this hypothesis. This is unfortunate because the impact of air conditioning use on outdoor $PM_{2.5}$ infiltrating indoors has possible implications for health effects research. For example, air conditioning use has been found to modify how exposure to ambient PM_{10} affects hospital admissions for heart and lung disease (Janssen et al 2002).

Organic PM

Organic PM dominated $PM_{2.5}$ concentrations both indoors and outdoors. Unlike other species, indoor concentrations were substantially higher than outdoor concentrations. This suggests that organic PM is likely to be the major species of $PM_{2.5}$ that is generated indoors.

Personal Exposure

The subjects of this study were mostly women who spent much of their time at home, where they were exposed to PM while cooking and cleaning; these activities could have more impact on personal exposure than on general indoor levels of PM. Possible exposures outside the homes could also contribute to differences in personal exposure levels.

The amount by which personal exposures exceed indoor concentrations is often referred to as the "personal cloud effect" (Özkaynak et al 1996). The personal cloud is thought to arise from activities that resuspend particles that have settled out of the air, resulting in increased exposures beyond measured indoor concentrations. The difference

between indoor concentrations and personal exposures measured in this study was 17 μg/m^3. Although human activity results primarily in the resuspension of coarse particles, fine particles also contribute to the personal cloud effect (Ferro et al 2004). A better characterization of the personal cloud would be informative for future studies of exposure and health effects, particularly because the particle size is known to influence particle deposition within the respiratory system.

METHODOLOGIC ISSUES OF DATA QUALITY

Differences Between Samplers

Although $PM_{2.5}$ mass concentrations measured by collocated PEMs and Harvard impactors were strongly correlated (R^2 = 0.98), the mass concentrations measured by PEMs were significantly higher, as seen in median (13.5 and 11.6 μg/m^3) and mean (19.5 and 16.5 μg/m^3) concentrations measured during collocated sampling.

Harvard impactors and conventional dichotomous samplers have been compared elsewhere, especially for estimating exposures to particle concentrations higher than 30 μg/m^3 (Thomas et al 1993). $PM_{2.5}$ samplers are designed to collect only the smaller end of the size distribution of respirable particles. Because of differences in efficiencies for collecting various sizes of particles, intersampler differences in results are magnified at higher concentrations. Only the absolute difference between measured concentrations will be affected, however, and not their correlation.

The authors hypothesized that differences between measurements made using Harvard impactors and PEMs could be due to how particles bounce from the impaction plates, losses from volatility, or both. The Harvard impactor has a single-jet impactor inlet with a face velocity of 16 cm/sec, whereas the PEM has an 8-jet inlet with a face velocity of 11 cm/sec. Samples obtained at low face velocity are less susceptible to volatilization (Turpin et al 2000).

In this study, concentrations measured using PEMs were significantly higher than the concentrations measured by collocated Harvard impactors for most soil elements (aluminum, silicon, calcium, titanium, iron, and zinc), but lower for accumulation mode elements such as sulfur and vanadium. The magnitude of the difference remained relatively constant over the range of concentrations observed.

The intersampler differences observed in this study could have resulted in a bias between samplers of approximately 1.4 μg/m^3 (4%) at the mean personal exposure measured. Intersampler differences alone are too small to explain the differences between measured personal exposures and indoor concentrations.

Adsorption Artifact

QFFs were used for measuring EC and OC because of their tolerance for thermal-optical analysis; however, the high specific surface area of the filters allows gas-phase semivolatile compounds to adsorb. For 89% of the samples collected by Harvard impactors, a QFF was placed behind the Teflon filter to estimate the absorption artifact (a filter–filter system). The reported particulate OC is thus the difference between the OC collected on the QFF in the MSP sampler and the OC on the backup QFF in the Harvard impactor.

In this study the percentage of adsorbed vapor was substantial for both indoor and outdoor samples in all cities combined (36.4% vs 36.6%, respectively); the adsorption artifact across the three cities differed little. Because the concentrations of OC were generally higher indoors, they tended to have higher absolute adsorption artifacts than outdoor concentrations. The indoor adsorption artifact was 2.9 μg/m^3, or 16.2% of total indoor $PM_{2.5}$ mass concentration; the outdoor adsorption artifact was 1.8 μg/m^3, or 10.8% of the total outdoor $PM_{2.5}$ mass concentration.

For the 11% of QFF samples without concurrent backup filters, values based on the amount of adsorbed vapor in available paired samples were used to estimate the adsorption artifact. This method may have introduced uncertainty into the results.

Although this approach partially corrects for an adsorption artifact, it does not account for volatilization (any particle-phase mass that evaporates from the front QFF during sampling). Furthermore, if particles evaporate from the Teflon filter and readsorb on the backup QFF, or if the pressure drop across the Teflon filter strips OC from the backup QFF, then error will be introduced in the absorption correction. The filter–filter correction is an improvement over no correction, but positive or negative errors to the observed particle-phase concentration can remain (Volckens and Leith 2002).

OUTDOOR CONTRIBUTION TO INDOOR AIR

Differences in the Three Approaches Used

The outdoor contribution to indoor and personal $PM_{2.5}$ concentrations was calculated using RCS, mass balance, and robust regression approaches. All three models assume that (1) outdoor-generated and indoor-generated $PM_{2.5}$ are independent, (2) indoor air is perfectly and instantaneously mixed, and (3) factors that affect indoor concentrations are constant or change slowly throughout the monitoring period.

The infiltration factor accounts for particle loss as the outdoor air penetrates indoors, particle introduction and loss through ventilation, and particle losses indoors. The RCS model assumes that one infiltration factor is applicable for all homes in all cities; it is determined as the slope of the regression of indoor on outdoor $PM_{2.5}$ concentrations. The mass balance model uses the actual AER and mass concentrations for each home to calculate a home-specific infiltration factor and results in a broader distribution of outdoor contributions to indoor $PM_{2.5}$. The robust regression model uses PM speciation data to estimate specific infiltration factors for each home on the day sampled.

Outdoor Contribution to Indoor Concentrations

In this study, both the RCS and mass balance models estimated an infiltration factor of 0.46 for all homes, which indicates that about half of the outdoor particles penetrated indoors. The agreement between the mass balance and RCS models for $PM_{2.5}$ suggests that the mean estimate was not substantially improved by using home-specific AERs (as in the mass balance model) rather than a constant infiltration factor (as in the RCS model). The absence of a measured AER, however, contributed an additional 26% uncertainty in the estimate of the outdoor contribution to indoor $PM_{2.5}$ concentrations.

A similar conclusion was found in an exposure study conducted in Phillipsburg NJ, Riverside CA, and Toronto, Canada (Ott et al 2000), although that study did 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. They reported an average infiltration factor of 0.5 for all cities combined, which is also consistent with the findings reported here. Another study in Vancouver, British Columbia, reported a similar average infiltration factor of 0.56 (Wilson and Brauer 2006). (That study also estimated that approximately 71% of the ambient $PM_{2.5}$ concentration was responsible for 44% of total personal exposure.)

Despite the broad agreement with other studies, the results of the mass balance model used in the RIOPA study may not be applicable to the general population or even to populations in the three cities studied because of the critical assumptions made about the infiltration and decay parameters used in the models. The investigators conducted a sensitivity analysis to determine how these parameters could influence the estimated outdoor contribution to indoor $PM_{2.5}$. Their results suggest that uncertainties in infiltration and decay could have been responsible for approximately 20% of the uncertainty in the mean outdoor contribution.

In contrast to the RCS and mass balance model, the robust regression approach estimated a much higher mean outdoor contribution to indoor $PM_{2.5}$ (73% compared with 43% for mass balance and 40% for RCS). The robust regression analysis was limited, however, because nitrate—a major species—was not measured. Nevertheless, subsequent sensitivity analyses performed by adding indoor and outdoor nitrate data from a California home in another study with similar atmospheric conditions suggested that this limitation may not have affected results (Meng et al 2005).

PM infiltration might change with different size distributions and sources. Combustion-derived materials (eg, EC) do not have the same size distribution and infiltration behavior as other species that are present in the atmosphere (eg, sulfur) or are formed from mechanical abrasion (eg, soil elements). Therefore, introducing more PM species into an analytical model (such as robust regression) allows for more variation in infiltration due to factors in addition to AER, such as particle penetration and decay rates. Such approaches would enable a more accurate estimation of the outdoor contribution to indoor $PM_{2.5}$ exposure.

Outdoor Contribution to Personal Exposure

For the mean outdoor contribution to personal $PM_{2.5}$ exposure, the estimates from the RCS and mass balance models were similar for 268 indoor–outdoor pairs of PM samples and the corresponding AERs.

IMPLICATIONS FOR EPIDEMIOLOGY

The selection of homes and subjects in this study was not population-based; rather, homes close to sources were preferentially selected in an effort to examine the effects of potentially high exposures to outdoor sources. This selection method limits the extent to which results can be generalized to other populations. If the study had been designed to represent the population distribution in each city, oversampling with respect to potential exposure to sources would have enabled epidemiologists to generalize the results.

The possibility that exposure error may arise from using data from ambient fixed-site monitors as a surrogate for exposure to PM of outdoor origin has been well acknowledged (Bates 2000). In this study, the investigators attempted to characterize the exposure error by exploring how additional information on AERs, home-to-home variations in particle composition, and infiltration factors may impact exposure estimates. They did not, however, suggest how incorporating information on these home characteristics could minimize exposure error in future epidemiologic studies.

CONCLUSIONS

Dr Turpin and her colleagues have made an important contribution by successfully achieving the first two of their objectives: (1) characterizing and comparing the composition of indoor, outdoor, and personal $PM_{2.5}$ in the three cities; and (2) estimating the contribution of outdoor $PM_{2.5}$ and its components to indoor and personal exposures. Indeed, this is one of the most comprehensive studies to characterize $PM_{2.5}$ exposures and one of the first to measure PM functional groups. The investigators did not, however, include the results of their exploratory source apportionment of personal and indoor $PM_{2.5}$ concentrations in this report.

Although the lack of a population-based sampling strategy limits the generalizability of the results for broad epidemiologic analyses, the compositional data can provide insight on exposure to PM components for a large number of subjects and homes selected on the basis of distances from various outdoor sources.

This study generated a rich database that can be used to identify what levels of exposure could pose health concerns, the sources of air toxics, and factors associated with high exposures. Some possible ways this database could be used are:

- a detailed analysis of elemental species;
- source apportionment;
- an analysis of how morphological characteristics of particles contribute to personal exposure;
- further descriptive analyses beyond those provided in the Investigators' Report; and
- additional modeling to (1) integrate information on housing characteristics and seasons, and (2) assess how pollutant levels and sources are related within individual homes.

HEI and NUATRC are currently developing additional opportunities to explore aspects of these data.

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