RESEARCH REPORT

HEALTH
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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

S T A T E M E N T

Synopsis of the RIOPA Research Report Part II

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 imila6.7(and difnof99(i)D0.0(m TJ12.96 0 TD0.0092 Tc-0.0005 Tw(fund research to (1))Tj-14.07

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 D0fif

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

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CONTENTS

$PM_{2.5}$

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,

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 [illegible] respectively.

$_{2.5}$ 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.

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

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

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. Homesd

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 middanysHof the t Tw3k2cepM-3.5r.6(d ma1)n JEmna1h;rat-ath1t Twre ideiv6(d6 ie0

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]

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

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

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

where n is the number of PMCH sources used, R

Figure 2. Sample analysis flow chart. Numbers of homes where samples were collected are in pa

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

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 continu-

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 / $PM_{2.5}$ J5.48 Tw[(the iQ2i)-4

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

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 particu-

(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.

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 “extract

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).

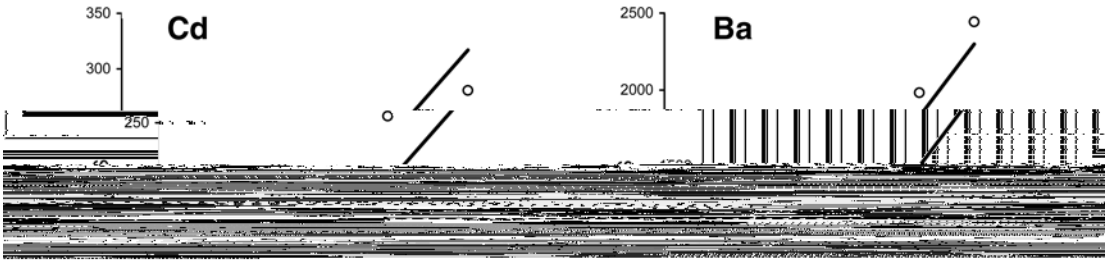


Figure 8. *(continued).*

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.

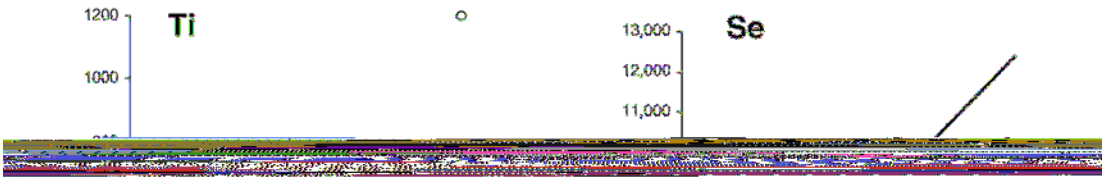


Figure 8. *(continued).*

Functional groups were identified from the aerosol literature (Allen et al 1994; Blando et al 1998; Carlton et al

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

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 $PM_{2.5}$

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

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								

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

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)								

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 $PM_{2.5}$

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 some-

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 th

clearly result in substantial bias in reported particulate OC

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 ove

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 cat-

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

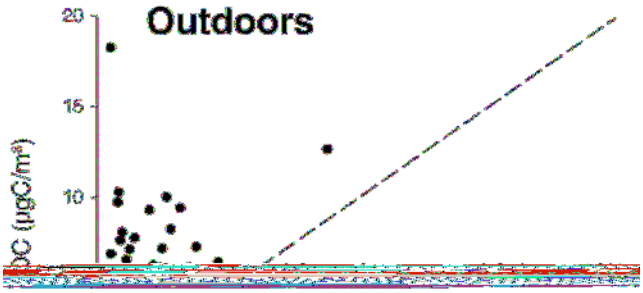


Figure 12. Indoor and outdoor concentrations of OC and EC for Los Angeles, Elizabeth, and Houston homes. Note that the axis lengths differ between panels.

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

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 absor-

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

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*

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.7(of resentennence oe)]TJces we

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:

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

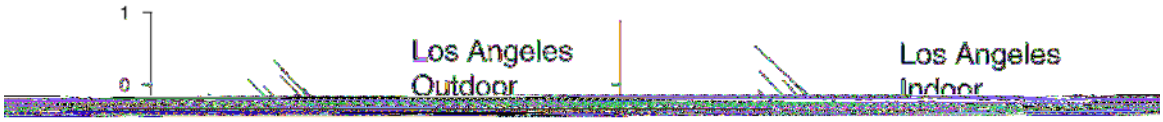
where $F_{2.5}$ and A (ng/m^3) are the PAH concentrations on the $PM_{2.5}$ QFF (particle phase) and on the adsorbent (gas phase), respectively. $PM_{2.5}$ ($\mu g/m^3$) is the $PM_{2.5}$ mass concentration. The propagated precision (ie, from random errors) in the gas–particle partition coefficients (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.


1
0
Los Angeles
Outdoor
Los Angeles
Indoor

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 (

(6)

where A, B, C, D, and I are fit parameters, $p_L^\circ(25°C)$ is the sub-

Expanding the regression of log $K_{p,meas}$ versus log p_L° to include

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

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

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 coeffi

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

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.

measured outdoor concentrations (C_{Out}

using the mass balance model both with and without

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

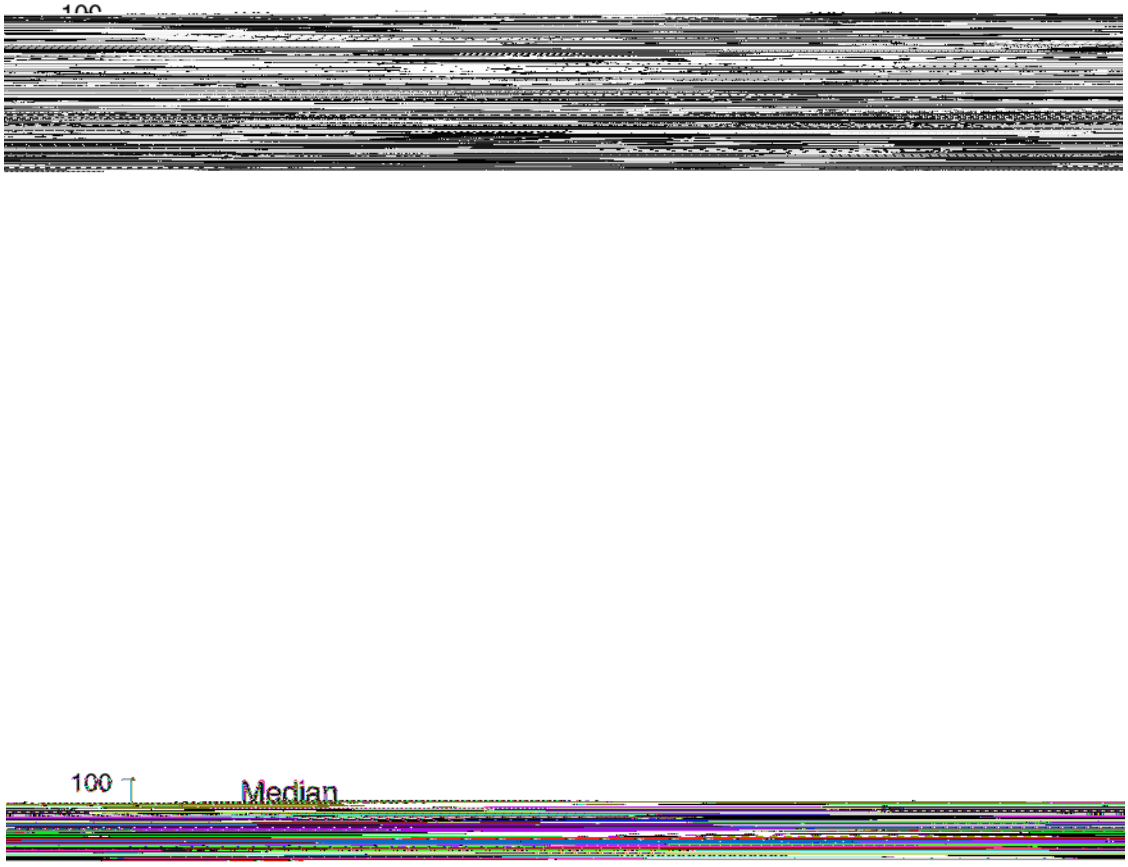


Figure 30. Sensitivity of the mass balance model results to the

by 4%. Accounting also for home-by-home variations in particle properties and housing properties broadened the distribution by 10%. Spatial variations in outdoor $PM_{2.5}$

indoor, and personal $PM_{2.5}$ exposure. Atmos Environ 36:3255–3265.

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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
age o10le A.1ion Mea- M3d2i1.224562T94014 CiatTDiati0 To47.4.6802.P.1-24.106.2(i-%Jo47[(aT9406.T63of 8E( )Ra34dsa7T9)6.o47

APPENDIX B. HEI and NUATRC Quality Assurance Statement

The RIOPA study was simultaneously performed over a

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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, su

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. How(riv7)-1 Scy-(2.15mm4(s)3((emst)6c

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

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-

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:

where C_{In} and C_{Out} are $PM_{2.5}$ concentrations ($\mu g/m^3$

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 $\mu g/m^3$

Functional Groups Functional groups are elemental structures attached to carbon that can influence a mole-

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

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 $PM_{2.5}$

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

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

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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