Journal of Exposure Science and Environmental Epidemiology (2006) 16, 321–331
© 2006 Nature Publishing Group All rights reserved 1559-0631/06/$30.00
www.nature.com/jes

Fine organic particulate matter dominates indoor-generated $PM_{2.5}$ in RIOPA homes

ANDREA POLIDORI[a], BARBARA TURPIN[a,b], QING YU MENG[a], JONG HOON LEE[a,2], CLIFFORD WEISEL[b], MARIA MORANDI[c], STEVEN COLOME[d], THOMAS STOCK[c], ARTHUR WINER[e], JIM ZHANG[b], JAYMIN KWON[b], SHAHNAZ ALIMOKHTARI[b], DEREK SHENDELL[e,3], JENNIFER JONES[e], CORICE FARRAR[e] AND SILVIA MABERTI[c]

[a]*Environmental Sciences, Rutgers University, New Brunswick, New Jersey 08901, USA*
[b]*Environmental and Occupational Health Science Institute, Piscataway, New Jersey 08854, USA*
[c]*School of Public Health, University of Texas, Houston, Texas 77030, USA*
[d]*Integrated Environmental Sciences, Irvine, California 92612, USA*
[e]*Environmental Science and Engineering Program, University of California, Los Angeles, California 90095, USA*

Residential indoor and outdoor fine particle ($PM_{2.5}$) organic (OC) and elemental carbon (EC) concentrations (48 h) were measured at 173 homes in Houston, TX, Los Angeles County, CA, and Elizabeth, NJ as part of the Relationship of Indoor, Outdoor and Personal Air (RIOPA) study. The adsorption of organic vapors on the quartz fiber sampling filter (a positive artifact) was substantial indoors and out, accounting for 36% and 37% of measured OC at the median indoor (8.2 μg C/m^3) and outdoor (5.0 μg C/m^3) OC concentrations, respectively. Uncorrected, adsorption artifacts would lead to substantial overestimation of particulate OC both indoors and outdoors. After artifact correction, the mean particulate organic matter (OM = 1.4 OC) concentration indoors (9.8 μg/m^3) was twice the mean outdoor concentration (4.9 μg/m^3). The mean EC concentration was 1.1 μg/m^3 both indoors and outdoors. OM accounted for 29%, 30% and 29% of $PM_{2.5}$ mass outdoors and 48%, 55% and 61% of indoor $PM_{2.5}$ mass in Los Angeles Co., Elizabeth and Houston study homes, respectively. Indirect evidence provided by species mass balance results suggests that $PM_{2.5}$ nitrate (not measured) was largely lost during outdoor-to-indoor transport, as reported by Lunden et al. This results in dramatic changes with outdoor-to-indoor transport in the mass and composition of ambient-generated $PM_{2.5}$ at California homes. On average, 71% to 76% of indoor OM was emitted or formed indoors, calculated by (1) Random Component Superposition (RCS) model and (2) non-linear fit of OC and air exchange rate data to the mass balance model. Assuming that all particles penetrate indoors ($P = 1$) and there is no particle loss indoors ($k = 0$), a lower bound estimate of 41% of indoor OM was indoor-generated (mean). OM appears to be the predominant species in indoor-generated $PM_{2.5}$, based on species mass balance results. Particulate OM emitted or formed indoors is substantial enough to alter the concentration, composition and behavior of indoor $PM_{2.5}$. One interesting effect of increased indoor OM concentrations is a shift in the gas-particle partitioning of polycyclic aromatic hydrocarbons (PAHs) from the gas to the particle phase with outdoor-to-indoor transport.
Journal of Exposure Science and Environmental Epidemiology (2006) **16,** 321–331. doi:10.1038/sj.jes.7500476; published online 15 March 2006

Keywords: *OC, organic PM, indoor air, sampling artifacts, organic aerosol, $PM_{2.5}$.*

Introduction

Since United States residents spend approximately 87% of their day indoors (Robinson and Nelson, 1995; Klepeis et al., 2001), understanding the composition, behavior and origin of indoor fine particulate matter ($PM_{2.5}$) is important to exposure characterization and mitigation. Typically, indoor $PM_{2.5}$ consists of ambient (outdoor) particles that infiltrate indoors and remain suspended, particles emitted indoors (primary), and $PM_{2.5}$ formed indoors (secondary) from precursors emitted both indoors and outdoors (Weschler and Shields, 1997; Weschler, 2004). When indoor sources are present, indoor PM concentrations can be substantially higher than outdoor PM concentrations (USEPA, 2004). These sources include smoking, cooking, gas stoves, other combustion, cleaning, washing and human activities that resuspend $PM_{2.5}$ (Yocom, 1982; Ozkaynak et al., 1996; Chao et al., 1997; Jones et al., 2000). Outdoor $PM_{2.5}$ is also a major contributor to indoor particle concentrations (Thatcher and Layton, 1995; Abt et al., 2000; USEPA, 2004).

Organic compounds are a large, complex, and poorly understood contributor to indoor and outdoor $PM_{2.5}$ (USEPA, 2004). Organic $PM_{2.5}$ is comprised of hundreds

1. Address all Correspondence to: Professor B. Turpin, Environmental Sciences, 14 College Farm Rd, New Brunswick, NJ 08901, USA.
Tel.: +1 732 932 9540. Fax: +1 732 932 8644.
E-mail: turpin@aesop.rutgers.edu
2. Current address: Department of Chemical Engineering, Clarkson University, Potsdam, NY 13699, USA.
3. Current address: Department of Environmental Public Health Sciences, Georgia State University, Atlanta, GA 30302, USA.
Received 21 September 2005; accepted 15 December 2006; published online 15 March 2006

of compounds spanning a wide range of vapor pressures and chemical properties. Typically, 10% to 70% of *ambient* $PM_{2.5}$ is organic, but despite substantial effort only 10% to 30% of ambient organic $PM_{2.5}$ is typically identified at the molecular-level (Rogge et al., 1993; Turpin et al., 2000). Accurate measurement of particulate organic $PM_{2.5}$ is also hampered by sampling artifacts (Turpin et al., 2000; Suburamanian et al., 2004), which have been extensively studied outdoors but largely ignored indoors. A few *indoor* organic PM species are relatively well studied (i.e., polycyclic aromatic hydrocarbons, PAHs; Dubowsky et al., 1999; Sakai et al., 2002; Naumova et al., 2002, 2003). However, little is known about organic $PM_{2.5}$ concentrations, composition, behavior and sampling artifacts in residential indoor environments. Coupled residential indoor and outdoor $PM_{2.5}$ organic carbon (OC), elemental carbon (EC) and/or black carbon (BC) concentrations have been measured in a few studies (Funasaka et al., 2000; Landis et al., 2001; Geller et al., 2002; Gotschi et al., 2002; Kinney et al., 2002; Li and Lin, 2003; Ho et al., 2004; Cao et al., 2005). These studies have yielded valuable insights. However, in all of these studies, interpretation of organic $PM_{2.5}$ results was limited by the presence of unquantified organic sampling artifacts. In fact, in one study, total particulate carbon indoors was 174% of the indoor $PM_{2.5}$ mass, presumably due to sampling artifacts (Landis et al., 2001).

Quartz fiber filters (QFF) are typically used for the measurement of particulate OC and EC because they can withstand the high temperatures of thermal-optical analysis. In addition to collecting particulate carbon with approximately 100% efficiency, some organic vapors also adsorb to the QFF surfaces. Left uncorrected, this adsorption artifact typically results in an overestimation of ambient particulate OC concentrations by 30% to 50%. (see the review by Turpin et al. (2000) for an extensive discussion of organic PM sampling artifacts). Little is known about the magnitude of the adsorption artifact indoors. However, indoor concentrations of organic gases are frequently higher than outdoor concentrations, suggesting that the artifact could be even more substantial indoors than outdoors. Also, indoor (and personal) sampling often has shorter collection times and lower flow rates (not true for this study), which result in samples where the adsorption artifact is smaller but is a larger fraction of the measured OC mass (Turpin et al., 2000). The size of the adsorption artifact depends on the filter surface area, sampling face velocity, the concentration and properties of the semi-volatile organic vapors, and the concentration of species competing for adsorption sites (i.e., water vapor). It is possible that a high enough pressure drop could develop across a heavily loaded filter to induce volatile losses of collected organic material (negative artifact). This artifact is difficult to measure, but experiments and calculations suggest that pressure-drop induced volatile losses are negligible for sampling conditions similar to the current study and other typical indoor studies (i.e., no denuder and low face velocity) (McDow and Huntzicker, 1990). In addition, changes in temperature and in organic vapor concentrations during sampling can disturb the equilibrium between the gas-phase passing through the filter and the organic material sorbed to the filter and particles. This provides a driving force for additional adsorption or for volatilization of the collected semi-volatile organic matter, leading to a sample that is weighted toward the conditions at the end of the sampling period. The magnitude of this effect could not be measured with the experimental design used in this study. Two recent outdoor studies with comparable face velocities have estimated volatilization of organic PM from undenuded QFFs to be on the order of 10% overall (Mader et al., 2003; Subramanian et al., 2004).

Typically, the contribution of the adsorption artifact to the measured ambient particulate OC concentration is minimized through the use of a denuder and/or estimated by using a concurrently collected "dynamic blank" (collection of particle-free ambient air in the same location at the same face velocity, for example, on a QFF downstream of a Teflon or QFF; Turpin et al., 2000). To estimate the adsorption artifact, the front (Teflon or quartz fiber) filter removes the particles so that the backup QFF only collects adsorbed vapor. The OC on this filter is an estimate of organic vapors adsorbed on the concurrently collected sampling QFF. Because a Teflon filter adsorbs very little, gas phase — adsorbed phase partitioning in the vicinity of a Teflon filter is expected to reach equilibrium rapidly so that the backup QFF "sees" the same concentration of adsorbable vapors as the concurrently collected sampling QFF (Mader and Pankow, 2001). The presence/absence of particles on the filter does not appear to alter the size of the adsorption artifact, presumably because the particles are already close to equilibrium with the surrounding gas when they are collected and the surface area of collected particles is extremely small relative to the surface area of a QFF (Turpin et al., 1994). In the current work, a dynamic blank (QFF behind a Teflon filter) was collected concurrently with each OC measurement to estimate and correct for the adsorption artifact.

During the Relationship of Indoor Outdoor and Personal Air study (RIOPA), 48 h integrated indoor and outdoor $PM_{2.5}$ samples were collected in non-smoking residences in Elizabeth, NJ, Houston, TX, and Los Angeles County, CA, between summer 1999 and spring 2001. Sampling was conducted in three geographically distinct locations with different climates and housing characteristics and across all seasons to create a data set with a wide distribution of air exchange rates, conducive to a mechanistic examination of the data. Some homes were particularly close to identified outdoor sources, while others were farther away. Questionnaires were administered to characterize homes, neighborhoods and activities that might affect exposures. Air exchange rate, temperature and relative humidity were also

measured in each home. $PM_{2.5}$ samples or subsets of samples were analyzed for mass (Meng et al., 2005a), functional groups (Reff et al., 2005), elements (Meng et al., 2005b), OC and EC, gas and particle phase PAHs (Naumova et al., 2002, 2003), and Chlordanes (Offenberg et al., 2004). Gas phase aldehydes (Liu et al., 2006) and volatile organic compounds (Weisel et al., 2004) were also measured. The RIOPA study design is described in detail by Weisel et al. (2004).

In this paper, indoor and outdoor $PM_{2.5}$ OC and EC concentrations are reported and used to provide new insights into the origin of organic particulate matter in residences. In the process, micro-environmental organic $PM_{2.5}$ sampling artifacts are estimated; indoor and outdoor species mass balances are constructed, and the contributions of indoor and outdoor sources to indoor OC mass concentrations are quantified.

Methods

Sampling

$PM_{2.5}$ OC and EC were measured concurrently inside and outside of 173 RIOPA homes. These measurements are well distributed across warm and cool seasons and across the three sampling locations. $PM_{2.5}$ samples collected at these homes were also analyzed for mass, functional groups and elements, including sulfur. Particulate nitrate was not measured. A 48-h collection time was used for all RIOPA chemical measurements to improve quantitation of trace-level species. The indoor samplers were placed in the main living area of the home, excluding the kitchen, and the outdoor samplers were placed in secure locations in the front or back yard. Both indoor and outdoor samplers were mounted 1 to 2 m from the floor and at least 1 m from walls or other structures.

Micro-environmental $PM_{2.5}$ samplers are illustrated in Figure 1. Indoor and outdoor samples for analysis of $PM_{2.5}$ OC, EC and trace-level organic compounds (PAHs and Chlordanes; described elsewhere) were collected at 10 l/min (25 cm/s face velocity) on 37 mm QFF (2500QAT-UP; Pallflex Gelman Scientific, Ann Arbor, MI, USA) using MSP Microenvironmental $PM_{2.5}$ samplers (MSP, Minneapolis, MN, USA). All MSP samplers were modified to hold a polyurethane foam (PUF) adsorbent (diameter 25 mm, height 100 mm) for collection of vapor-phase PAHs and Chlordanes downstream of the multiple jet 2.5 μm cutpoint impactor inlet and QFF. QFFs were pre-baked at 550°C for 2 h and stored at room temperature in Petri dishes lined with pre-baked aluminum foil. Indoor and outdoor samples for $PM_{2.5}$ mass, elemental and functional group analyses were collected concurrently on 37 mm stretched Teflon filters (2 μm pore; Pallflex Gelman Scientific, Ann Arbor, MI, USA) located downstream of a single-jet 2.5 μm aerodynamic diameter cutpoint impactor (Harvard Impactor) operated

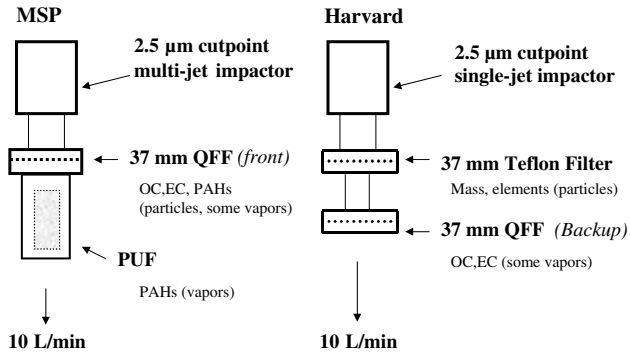

Figure 1. Micro-environmental $PM_{2.5}$ samplers used both indoors and outdoors. MSP samplers were modified to hold a polyurethane foam adsorbant (PUF) behind a 37 mm quartz fiber filter (QQF). The MSP QFF collects particles with essentially 100% efficiency and adsorbs some organic vapors. The MSP QFF was analyzed for organic carbon (OC), elemental carbon (EC) and polycyclic aromatic hydrocarbons (PAHs). The PUF was analyzed for gas-phase PAHs. The Harvard Impactor held a Teflon filter followed by a QFF. The Teflon filter collects particles and was analyzed for mass and elements. The Harvard QFF (backup) samples particle-free ambient air and provided an estimate of organic vapor adsorption on the MSP QFF (front) filter.

at 10 l/min. A 37 mm QFF was located behind the Teflon filter. The OC on this Teflon-quartz backup filter was subtracted from the concurrently collected MSP QFF to correct for the adsorption of organic vapors. More detailed explanations of sampling artifacts are provided in the Introduction and in Turpin et al. (2000).

Filters were loaded, unloaded, and leak checked in the laboratory. Air flow-rates were measured at the beginning and at the end of each sampling period, and samplers were leak-checked at the end of the sampling period if the measured flow rate had changed by more than $\pm 5\%$. In total, 93% of MSP samples were valid. A field blank for each filter type was transported with the 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 Harvard Impactors and MSP samplers collocated at 35 and 31 study homes, respectively. Collected samples and field blanks were returned to each laboratory, shipped overnight to NJ in coolers with blue ice packs, and stored frozen (−4°C) until analysis. A field sheet form was used to guide the field technician through the process of measuring and recording critical data, such as flow rates, start/stop times, and comments that could affect sample validity.

Analyses

Analysis of samples for mass and elements is described in detail elsewhere (Meng et al., 2005a, b). We used these measurements to construct $PM_{2.5}$ species mass balances. Since the focus of the present study is carbonaceous $PM_{2.5}$, a description of RIOPA carbon analysis is provided below.

OC and EC were measured by thermal-optical transmittance in a Sunset Laboratory Carbon Analyzer (Birch and Cary, 1996). Briefly, air is purged from the analyzer after a 1-cm^2 punch of sampled QFF is loaded. The QFF is then heated in a helium (He) environment, stepwise to volatilize OC. After removal of OC, EC is eluted by combustion in 2% oxygen (O_2) in He while heating stepwise. In this study, top He and He–O_2 temperatures of 820°C and 910°C were used to match the anticipated but not yet finalized protocol for EPA Speciation Trends Network (STN) samples. Evolved carbon is converted to methane (CH_4) and measured with a flame ionization detector (FID). A calibration gas with a known amount of CH_4 (nominally 5% CH_4 in He, certified) is automatically injected by switching a loop of calibration gas on-line in the last step of each analysis for quantitation. During analysis, some OC is pyrolytically converted to EC, which is light absorbing. Correction for pyrolysis is made by monitoring the transmittance of light through the filter using a diode laser and photodetector.

A comprehensive set of quality control checks were made during RIOPA carbon analysis. Instrument blanks were measured daily during sample analysis; no system contamination was found. The variability in detector sensitivity (FID response) within an analysis was also measured daily by running an analysis with automatic calibration gas injection in He and He — O_2 analysis segments. Detector variability was less than 5%. The instrument's calibration loop volume (1.3 ± 0.02 ml for this instrument) was verified every 3 months with manual injection of certified calibration gas and analysis of sucrose standards. This yields 32.5 ± 0.5 μg of carbon (μg C) in the internal calibration peak, given a calibration tank with exactly 5% CH_4 in He. The transit time of evolved material from the filter punch to the FID was stable, at 11 s. (The photodetector responds instantly when EC is formed or evolved, but the corresponding change in the FID signal is delayed by the transit time.)

Detection limits, expressed as three times the standard deviation of the field blanks, were 0.3 μg C/m^3 for OC and 0.07 μg C/m^3 for EC. (It should be noted that detection limits would have been even lower had baked substrates been stored cold before, as well as after, sampling.) All OC and EC measurements except four (three outdoor OC values and one indoor EC value) were above detection limits (see Table 1 for details). Analytical precision, expressed as the pooled coefficient of variation (CV) of replicate analyses of 10% of all samples, was 5% for OC and 9% for EC. Analytical accuracy for total carbon (TC = OC + EC) was 3.5%, based on daily analyses of sucrose standard solutions spiked on a QFF. Measurement precision was 4% for OC and 7% for EC, calculated as the pooled CV of measurements from MSP samplers collocated outdoors at RIOPA study homes. Thus, the (within-method) measurement precision was comparable to the analytical precision.

Results and discussion

Organic Aerosol Sampling Artifacts

Figure 2 shows the percentage of measured front filter OC (i.e., gases plus particles collected on the QFF in the MSP

Table 1. Particulate OC and EC concentrations (μg C/m^3)

Species	Location	Category	*N*	Below DL*	Front filter	Backup filter	Particulate Carbon (= front – backup)		
					Mean	Mean	Mean	SD	Median
OC	Los Angeles Co.	Indoor	44	0	8.08	2.65	5.43	3.41	4.68
OC	Los Angeles Co.	Outdoor	44	0	6.03	1.91	4.12	1.88	3.64
OC	Elizabeth	Indoor	60	0	11.08	3.18	7.90	9.07	5.39
OC	Elizabeth	Outdoor	60	0	5.06	1.73	3.33	1.73	3.02
OC	Houston	Indoor	69	0	10.51	3.29	7.22	5.35	5.35
OC	Houston	Outdoor	69	3	5.43	2.22	3.21	3.07	2.25
OC	All Locations	Indoor	173	0	10.09	3.09	7.00	6.59	5.30
OC	All Locations	Outdoor	173	3	5.45	1.97	3.48	2.40	3.03
EC	Los Angeles Co.	Indoor	44	0	1.34	0.07 ND	1.27	0.81	1.12
EC	Los Angeles Co.	Outdoor	44	0	1.51	0.10	1.41	0.91	1.16
EC	Elizabeth	Indoor	60	0	1.42	0.06 ND	1.36	1.28	1.10
EC	Elizabeth	Outdoor	60	0	1.43	0.07 ND	1.36	0.64	1.29
EC	Houston	Indoor	69	1	0.74	0.05 ND	0.69	0.82	0.48
EC	Houston	Outdoor	69	0	0.75	0.05 ND	0.70	0.34	0.65
EC	All Locations	Indoor	173	1	1.13	0.06 ND	1.07	1.04	0.87
EC	All Locations	Outdoor	173	0	1.18	0.07 ND	1.11	0.71	0.96

Number of samples (*N*), number of front filter measurements below detection limits (DL), measured front filter, measured backup filter, and particulate OC and EC concentrations (= front filter – backup filter) in μgC/m^3. SD, standard deviation. ND, below detection limit (0.30 and 0.07 μgC/m^3 for OC and EC, respectively.

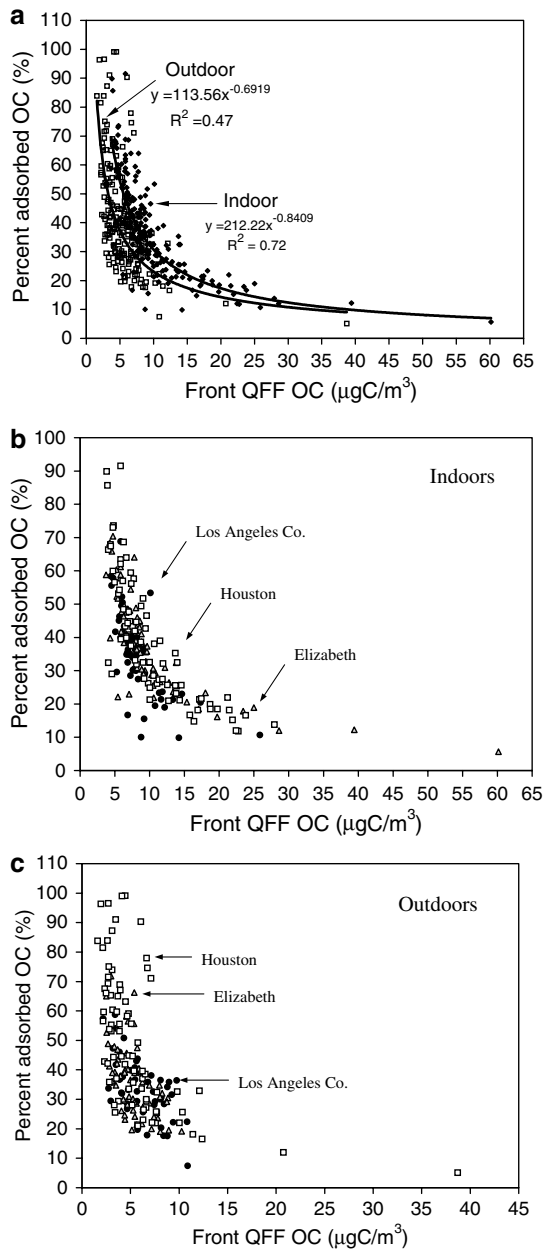

Figure 2. Percentage of measured OC (i.e., gases plus particles) that was adsorbed vapor: (a) indoor and outdoor samples, all study homes; (b) indoor only; and (c) outdoor only.

sampler) that was adsorbed vapor (i.e., OC measured on the concurrently-collected Harvard impactor backup QFF). The bias introduced by the adsorption artifact becomes less important (i.e., a smaller percentage of the sample) as the loading of $PM_{2.5}$ increases. At small sample loadings adsorbed vapors can dominate the sampled mass. In the RIOPA study, front filter OC concentrations tended to be higher indoors than outdoors. At the median front filter OC concentration (i.e., 8.2 μg C/m^3 indoors and 5.0 μg C/m^3 outdoors), the percentage of measured OC that was adsorbed vapor was 36% indoors (corresponding to 3.1 μg C/m^3) and 37% outdoors (corresponding to 1.8 μg C/m^3). However, at any single front filter OC concentration, the indoor artifact was larger than the outdoor artifact. For example, an indoor measured OC concentration of 8 μg C/m^3 is about 36% adsorbed vapor, whereas an outdoor measured OC concentration of 8 μg C/m^3 is about 27% adsorbed vapor. This suggests that organic vapors indoors have a greater tendency to adsorb to the QFF than organic vapors outdoors, presumably due to differences in the source mix and composition of volatile and semivolatile organic compounds. Furthermore, adsorption artifact behavior did not appear to be substantially different between Los Angeles Co., Elizabeth, and Houston study homes. Differences in 48-h mean temperature did not help explain the remaining variance (not shown). The sampling artifact findings are consistent with previous outdoor studies in magnitude and functional dependence (Turpin et al., 2000; Lim et al., 2003; Polidori et al., 2006). Clearly, if uncorrected an artifact of this magnitude would result in substantial bias in the reported particulate OC concentrations. This finding could explain the results obtained in some previous exposure studies that reported concentrations of indoor $PM_{2.5}$ carbon that exceeded the total indoor $PM_{2.5}$ mass concentrations (e.g., Landis et al., 2001).

Particulate OC and EC Concentrations

Particulate OC concentrations (μg C/m^3) below are corrected for the adsorption artifact on a sample-by-sample basis by subtracting the backup QFF (Harvard Impactor) from the concurrently collected front QFF (MSP sampler). In cases where backup filters were not collected (11% of samples), the equations in Figure 2a were used to estimate the magnitude of the adsorption artifact. Table 1 provides a summary of RIOPA particulate OC and EC concentrations.

Species Mass Balance

In Figure 3, the mean species contributions to the indoor and outdoor $PM_{2.5}$ mass concentrations are shown by location. The indoor and outdoor species contributions for the homes in the highest 25th percentile by outdoor $PM_{2.5}$ mass concentration are provided in Figure 4. These figures illustrate the importance of indoor sources of organic PM, and are consistent with a substantial loss of particulate nitrate indoors in California homes, as proposed by Lunden et al. (2003).

Sulfur from XRF analyses was assumed to be in the form of ammonium sulfate and OC concentrations were multiplied by 1.4, an estimate of the average organic molecular weight per carbon weight in urban areas (Turpin and Lim, 2001), to yield particulate organic matter (OM). Soil dust concentrations were calculated as the sum of the oxides of Al, Si, Ca, Ti, Fe, and K (Brook et al., 1997; Lee et al., 2002). Ammonium nitrate and water (not measured) were the main

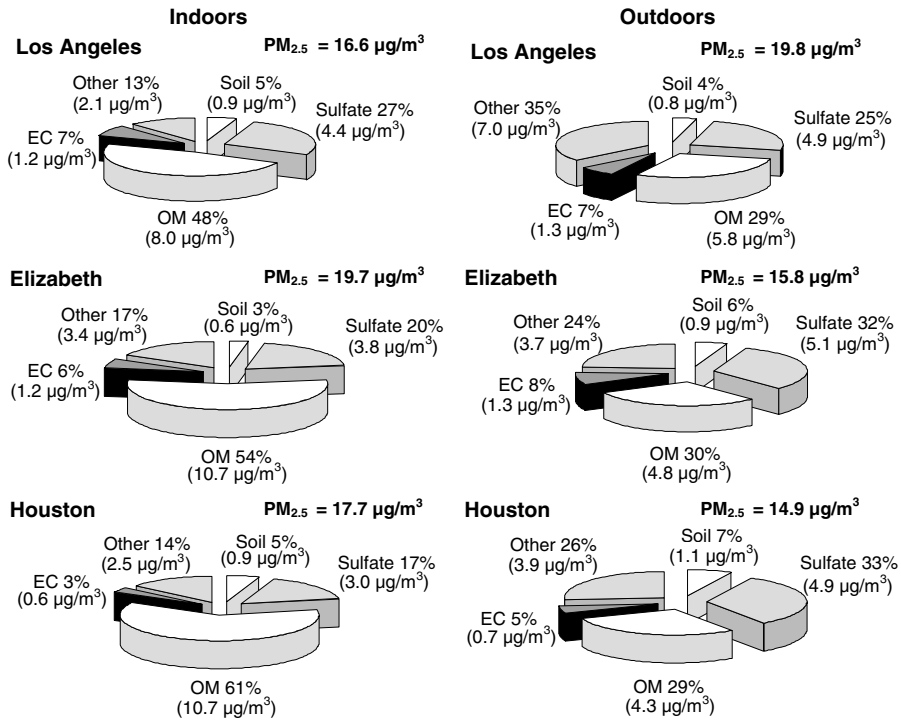

Figure 3. Mean species contributions (% and $\mu g/m^3$) to indoor and outdoor $PM_{2.5}$ mass concentration for Los Angeles Co., CA, Elizabeth, NJ and Houston, TX study homes. Soil is sum of oxides; sulfate is ammonium sulfate; organic matter is $OM = 1.4 \times OC$; EC is elemental carbon; "other" is the difference between the measured $PM_{2.5}$ mass and the sum of the measured species. The major components of "other" are expected to be ammonium nitrate and water. Shown are the means of all samples subjected to complete speciation. Plots represent 125 homes.

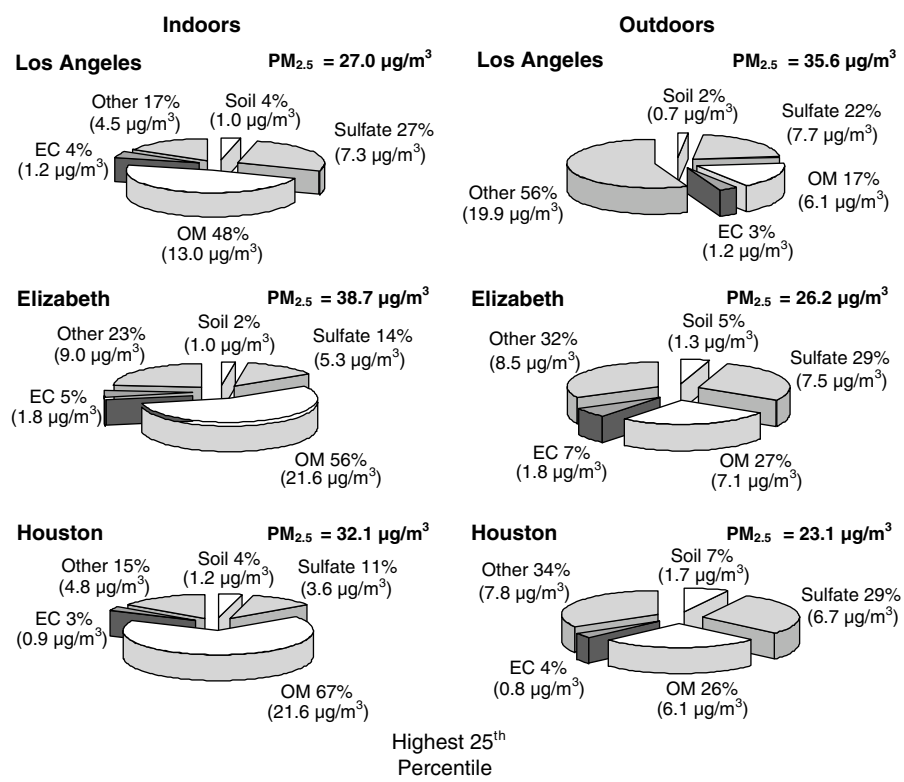

Figure 4. Mean indoor and outdoor species contributions (% and $\mu g/m^3$) for homes in the highest 25th percentile by outdoor $PM_{2.5}$ mass concentration for Los Angeles Co., CA, Elizabeth, NJ, and Houston, TX study homes.

components of the category called "other" (i.e., the difference between the $PM_{2.5}$ mass concentration and the sum of the measured species concentrations).

Outdoor mass balance results are in reasonable agreement with other urban studies (Andrews et al., 2000; USEPA, 2004). "Other" was a larger contributor to outdoor $PM_{2.5}$ mass concentration in Los Angeles Co. samples than in Houston and Elizabeth samples (especially on high $PM_{2.5}$ days; Figure 4), reflecting higher nitrate concentrations in California. Sulfate was a larger percentage contributor to Elizabeth and Houston outdoor samples than to Los Angeles Co. outdoor samples, as expected.

The most notable observation in the species mass balance results is the much larger contribution of particulate OM indoors than outdoors. Particulate OM (mean $\pm$ 95 percent confidence intervals) constituted $48 \pm 16\%$, $54 \pm 18\%$, and $61 \pm 22\%$ of $PM_{2.5}$ mass concentration inside Los Angeles Co., Elizabeth, and Houston study homes, respectively (Figure 3). The mean particulate OM concentration indoors ($9.8\,\mu g/m^3$) was double the value outdoors ($4.9\,\mu g/m^3$). In contrast, the mean EC concentration was $1.1\,\mu g/m^3$ indoors and outdoors. The elevated concentration of particulate OM indoors suggests that organic $PM_{2.5}$ was emitted or formed inside the RIOPA homes in sufficient quantities to substantially alter the concentration and the composition of $PM_{2.5}$ indoors. This is consistent with the finding of Geller et al. (2002), who reported that OC concentrations inside Coachella Valley, CA, USA homes were 77% higher on average than the corresponding concentrations outdoors.

Interestingly, the indoor–outdoor comparison for Los Angeles Co. homes is somewhat different. Like in Elizabeth and Houston, Los Angeles Co. study homes had substantially higher concentrations of particulate OM indoors. However, the concentration (and percentage contribution) of "other" was substantially smaller indoors ($2.1\,\mu g/m^3$; 13%) than outdoors ($7.0\,\mu g/m^3$; 35%). This difference was particularly pronounced on high $PM_{2.5}$ days (Figure 4). Since the largest component of "other" is expected to be ammonium nitrate, this finding is consistent with modeling and controlled experimental results reported by Lunden et al. (2003). They suggested 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 reduced contribution of "other" to Los Angeles Co. samples more than makes up for the increased contribution of OM, so the percent contribution of ammonium sulfate was actually slightly higher indoors despite the fact that the mean ammonium sulfate concentration was lower indoors. The loss of "other" $PM_{2.5}$ provides some evidence that the composition of "indoor $PM_{2.5}$ of outdoor origin" can differ substantially from that of residential-outdoor and central-site $PM_{2.5}$. This suggests that central site outdoor $PM_{2.5}$ 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. Implications to PM epidemiology warrant investigation.

Indoor and Outdoor Contributions to Carbon

Figure 5 shows the indoor and outdoor concentrations of OC and EC. Particulate OC was substantially higher indoors than outdoors for many Los Angeles Co., Elizabeth, and Houston study homes (Figure 5a). In addition, indoor and outdoor OC concentrations were poorly correlated ($R^2 = 0.01$). These observations suggest that many RIOPA study homes had substantial indoor sources of particulate OC. In contrast, with a few exceptions, paired indoor and outdoor EC concentrations were similar (Figure 5b). After removing 1 to 3 outliers, within-city indoor and outdoor EC concentrations were reasonably well correlated ($R^2 = 0.43$–0.79). Only two homes had dramatically higher EC concentrations indoors than outdoors, which suggests that substantial indoor emissions of EC occurred rarely. This is consistent with the finding of Funasaka et al. (2000), who reported that EC concentrations inside and outside roadside homes in Osaka, Japan were well correlated ($r = 0.86$). In

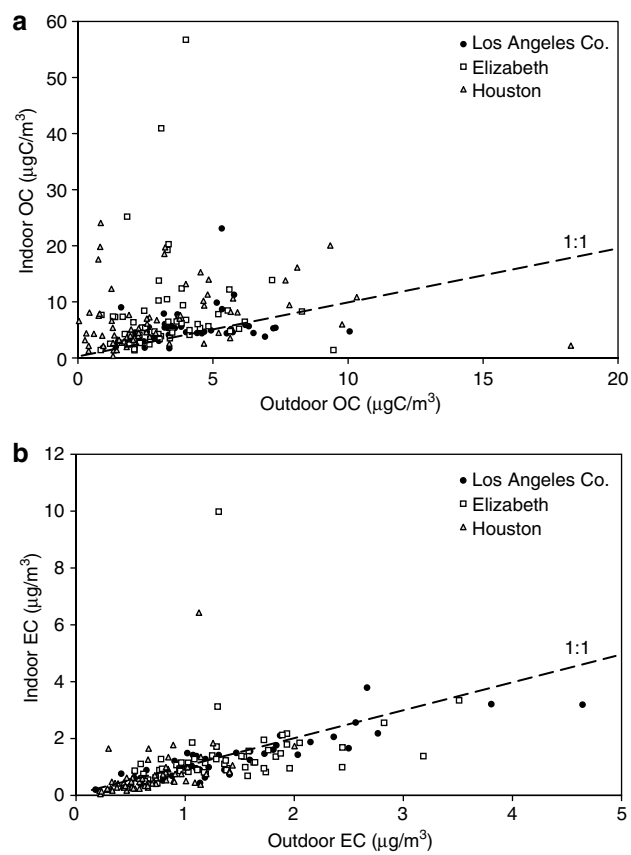

Figure 5. Indoor and outdoor OC (a) and EC (b) concentrations measured during RIOPA at Los Angeles Co., Elizabeth, and Houston study homes.

addition, the correlation between outdoor OC and EC ($R^2 = 0.23$) was higher than the indoor OC–EC correlation ($R^2 = 0.01$), and the OC/EC ratio was higher indoors (mean OC/mean EC = 6.5) than outdoors (mean OC/mean EC = 3.1). Assuming that EC originated predominantly outdoors, a weaker indoor correlation and a higher indoor OC/EC ratio are also consistent with the presence of indoor sources of particulate OC. Ho et al. (2004) also found higher OC/EC ratios indoors (mean OC/EC = 2.7) than outdoors (mean OC/EC = 2.0) in Hong Kong. Additional evidence of indoor-generated organic PM is provided by Reff et al. (2005), who found that Fourier transform infrared (FTIR) absorbances attributed to aliphatic hydrocarbon and amide functional groups were enhanced in most indoor RIOPA samples relative to absorbances in concurrently collected outdoor samples. Indoors, particulate OC can be emitted directly in the particle phase (i.e., primary) from sources including cooking, and can be formed in indoor air (i.e., secondary) as a result of reactions involving gas-phase organic compounds and ozone (Weschler and Shields, 1997; Fan et al., 2003; Nazaroff and Weschler, 2004; Weschler, 2004). Outdoors, OC also has primary sources and photochemical reactions can generate substantial secondary OC when conditions are favorable (Pandis et al., 1992; Turpin and Huntzicker, 1995; Lim and Turpin, 2002). EC is formed through incomplete combustion, and has been used as a tracer for primary, combustion-generated OC and for diesel exhaust PM.

The mean contributions of indoor and outdoor sources to indoor OC concentrations were estimated using (1) the Random Component Superposition (RCS) Statistical Model (Ott et al., 2000) and (2) a single compartment mass balance model. Assuming perfect instantaneous mixing and assuming that factors affecting indoor concentrations are constant or change slowly throughout the monitoring period, the steady-state indoor particulate OC concentration can be described as the sum of OC generated outdoors and OC generated indoors as follows:

$$C_{\mathrm{in}} = \frac{PaC_{\mathrm{out}}}{a+k} + \frac{Q_{\mathrm{i}}/V}{a+k} = F_{\mathrm{inf}}C_{\mathrm{out}} + C_{\mathrm{ig}} = C_{\mathrm{og}} + C_{\mathrm{ig}} \quad (1)$$

where C_{in} is the indoor particulate OC concentration, C_{out} is the outdoor-residential particulate OC concentration, F_{inf} is the dimensionless infiltration factor, C_{ig} is the concentration of indoor-generated OC found indoors, and C_{og} is the concentration of outdoor-generated OC found indoors. In the mass balance model, F_{inf} is given by $Pa/(a+k)$, where P is the dimensionless penetration coefficient, a is the air exchange rate (h^{-1}), and k is the particle loss rate (h^{-1}). Also in the mass balance model, C_{ig} is $Q_{\mathrm{i}}/V(a+k)$, where Q_{i} is the indoor source strength (μg/h), and V is the house volume (m^3).

In the RCS approach, a constant F_{inf} was obtained across all homes from the linear regression of the indoor OC concentrations (C_{in}) on the outdoor OC concentrations (C_{out}). The product of F_{inf} and each outdoor OC concentration (F_{inf} C_{out}) provided an estimate of the distribution of outdoor OC found inside study homes (C_{og}). The distribution of indoor-generated OC found inside study homes (C_{ig}) was given by the difference between C_{in} and C_{og} on a home-by-home basis. The RCS model assumes a linear superposition of OC of outdoor and indoor origin and a lack of correlation between these two components. Results can be easily influenced by outliers, and it must be recognized that, in reality, particle infiltration is not constant across homes nor with time. Using this method, 76%, on average, of OC found indoors was emitted or formed indoors, rather than being transported inside from outdoor sources. After removing two outliers from the data set the F_{inf} was recalculated and decreased by 0.01 (from 0.33 to 0.32). The RCS-estimated mean indoor contribution to indoor OC (C_{ig}) decreased by 0.5 μg C/m^3 (from 5.9 to 5.4 μg C/m^3), and the mean percentage indoor contribution to indoor OC remained 76%. Outliers were considered to be data with standardized residuals greater than 5. Results without outliers are presented herein.

In the mass balance approach, C_{og} values for each home were calculated using the concurrently-collected air exchange rate (a) and outdoor OC concentration (C_{out}) and assuming $P = 0.99$ and $k = 1.17\ \mathrm{h}^{-1}$ for all homes. Constant values of k and P were calculated by fitting measured C_{in}, C_{out}, and a to Eq. (1) using non-linear regression analysis (NLIN, SAS Version 8.02; SAS Institute, Cary, NC, USA; Meng et al., 2005b). C_{ig} values were then calculated as the home-by-home difference between C_{in} and C_{og}. It must be noted that estimates of P and k obtained by nonlinear regression are not truly independently determined. Also, in reality, P and k will vary from home to home and from day to day. P varies with particle size and house structure, while k is mainly determined by surface-to-volume ratio, housing structure, near-surface air flows, turbulence, and particle size distribution. Using the mass balance approach 71%, on average, of OC found indoors was emitted or formed indoors, rather than being transported inside from outdoors. After removing two outliers (i.e., data with standardized residuals greater than 5) from the dataset the mean F_{inf} was recalculated and remained the same (0.41). The mass balance-estimated mean indoor contribution to indoor OC (C_{ig}) decreased by 0.6 μg/m^3 (from 5.6 to 5.0 μg C/m^3), and the mean percentage indoor contribution to indoor OC remained 71%. Results without outliers are presented herein.

Although the uncertainties inherent in these two approaches must be recognized, these findings are reasonable, especially in light of the following lower-bound calculation. Assuming that all outdoor particles penetrated indoors through the building envelope ($P = 1$) and that there were no particle losses indoors ($k = 0$), an average of 41% (3.9 μg C/m^3) of indoor OC was emitted or formed indoors

according to the mass balance model. This represents a lower bound estimate of indoor OC of indoor origin.

Figure 6 shows the cumulative log-normal distribution of the outdoor-residential contribution ($\mu g\,C/m^3$) to indoor OC obtained with the RCS model (fixed F_{inf}), the mass balance model (variable F_{inf}), and an upper-bound case ($F_{inf} = 1$). Results are summarized in Table 2. Taken together these results highlight the importance of indoor sources of particulate OC.

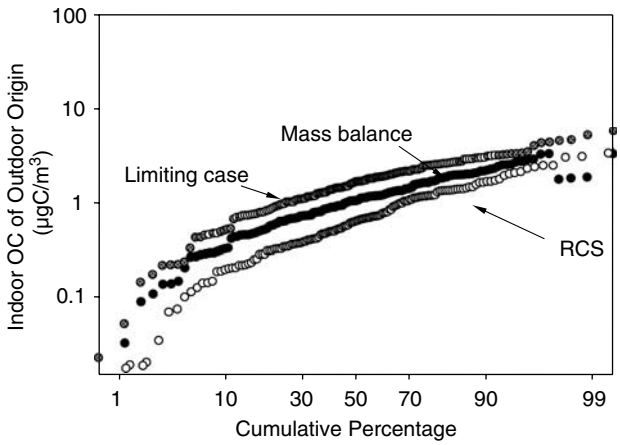

Figure 6. Cumulative percentage distributions of ambient (outdoor-residential) contributions ($\mu g\,C/m^3$) to indoor OC obtained by using the RCS model (fixed F_{inf}), the mass balance model (variable F_{inf}), and a bounding case ($F_{inf} = 1$).

Conclusions

This paper provides (1) one of the first assessments of the contributions of indoor and outdoor sources to indoor-residential concentrations of particulate OC; (2) an assessment of sampling artifacts that have hampered recent efforts at PM speciation; and (3) some insights into changes in particle properties with outdoor-to-indoor transport. A thorough analysis of the RIOPA study OC and EC sampling artifacts showed that, if uncorrected, adsorption artifacts could lead to substantial overestimation of particulate OC both in indoor and outdoor environments (the median percentage of measured OC that was adsorbed vapor was 36% indoor and 37% outdoor). $PM_{2.5}$ species mass balances suggested that OM dominated indoor $PM_{2.5}$ mass and was a major component in the outdoor samples. Particulate OM (corrected for artifacts) constituted 48%, 54% and 61% of $PM_{2.5}$ mass inside Los Angeles Co., Elizabeth and Houston study homes, respectively. While $PM_{2.5}$ nitrate was not measured in RIOPA, Los Angeles Co. species mass balance results are consistent with large nitrate losses during outdoor-to-indoor transport, as reported by Lunden et al. (2003). This suggests that dramatic changes in the mass and composition of outdoor-generated $PM_{2.5}$ can occur with outdoor-to-indoor transport in areas where nitrate is a major component of $PM_{2.5}$ (e.g., California). Since many epidemiological studies to date have used outdoor, central-site monitors as surrogates for personal exposure to $PM_{2.5}$ of outdoor origin, the impact of such transformations on epidemiological measurement errors warrants further

Table 2. Contributions of outdoor air and indoor sources to indoor OC (C_{og} and C_{ig}, respectively) estimated using three different methods: limiting case ($F_{inf} = 1$), RCS model (fixed F_{inf}), and mass balance model (variable F_{inf})

Method	Statistics	C_{og} ($\mu g\ C/m^3$)	C_{ig} ($\mu g\ C/m^3$)	C_{og} (%)	C_{ig} (%)
Limiting case					
$N = 173$	Mean	3.1	3.9	59	41
$P = 1$	Median	3.0	2.0	56	44
$k = 0$	5th percentile	0.7	0.0	7	0
$F_{inf} = 1$	95th percentile	5.9	16.2	100	93
RCS					
$N = 171$	Mean	1.1	5.4	24	76
	Median	1.0	3.8	18	82
	5th percentile	0.2	1.0	3	49
$F_{inf} = 0.32$	95th percentile	2.5	17.2	51	97
Mass balance					
$N = 150$	Mean	1.5	5.6	29	71
$P = 0.99$	Median	1.1	3.6	25	75
$k = 1.17$	5th percentile	0.2	0.9	2	25
$F_{inf} = 0.41$[a]	95th percentile	3.7	18.4	75	98

P and k values for OC are also reported for each method.
[a]For the Mass Balance Model, the mean, median, 5th percentile, and 95th percentile for the infiltration factor (F_{inf}) were 0.41, 0.39, 0.15, and 0.70, respectively.

investigation. Our modeling suggested that at least 41%, but more likely 71% to 76% of OC found indoors was emitted or formed indoors, rather than being transported inside from outdoors. Reff and co-workers (Reff et al., 2005; Reff, 2005) reported that RIOPA indoor $PM_{2.5}$ samples were enhanced in aliphatic and amide functionalities relative to concurrently collected RIOPA outdoor $PM_{2.5}$ samples. Additionally, while investigating the gas-particle partitioning of PAHs, Naumova et al. (2003) concluded that one effect of increased indoor carbon concentrations was a shift in the partitioning of PAHs from the gas to the particle phase. These results further highlight the importance of indoor sources of particulate OC.

Acknowledgements

The RIOPA study was funded by the Health Effects Institute, the Mickey Leland National Urban Air Toxics Center, the New Jersey Center for Environmental Indicators (CEI), the NIEHS Center of Excellence, and the NJ Agricultural Experiment Station. We gratefully acknowledge the hospitality of the RIOPA participants, the hard work of all the students and technicians in the field and laboratories of the RIOPA investigators, and Marta Chicano, Eric Spring, and Sandra Lake for their valuable contributions. Research described in this article was conducted, in part, under contract to the Health Effects Institute (HEI), an organization jointly funded by the United States Environmental Protection Agency (EPA: Assistance Agreement R828112) and automotive manufacturers. The contents of this article do not necessarily reflect the views of HEI, nor do they necessarily reflect the views and policies of EPA or of motor vehicle and engine manufacturers.

References

Abt E., Suh H.H., Allen G., and Koutrakis P. Characterization of indoor particle sources: a study conducted in the metropolitan Boston area. *Environ Health Persp* 2000: 108: 35–44.

Andrews E., Saxena P., Musarra S., Hildemann L.M., Koutrakis P., McMurry P.H., Olmez I., and White W.H. Concentration and composition of atmospheric aerosols from the 1995 SEAVS experiment and a review of the closure between chemical and gravimetric measurements. *J Air Waste Manage Assoc* 2000: 50: 648–664.

Birch M.E., and Cary R.A. Elemental carbon-based method for monitoring occupational exposures to particulate diesel exhaust. *Aerosol Sci Technol* 1996: 25: 221–241.

Brook J.R., Dann T.F., and Burnett R.T. The relationship among TSP, PM_{10}, $PM_{2.5}$, and inorganic constituents of atmospheric particulate matter at multiple Canadian locations. *J Air Waste Manage* 1997: 47: 2–19.

Cao J.J., Lee S.H., Chow J.H., Cheng Y., Ho K.F., Fung K., Liu S.X., and Watson J.G. Indoor/Outdoor relationships for $PM_{2.5}$ and associated carbonaceous pollutants at residential homes in Hong-Kong — case study. *Indoor Air* 2005: 15: 194–204.

Chao C.Y.H., Tung T.C.W., and Burnett J. Influence of different indoor activities on the indoor particulate levels in residential buildings. *Indoor Built Environ* 1997: 7: 110–121.

Dubowsky S.D., Wallace L.A., and Buckley T.J. The contribution of traffic to indoor concentrations of polycyclic aromatic hydrocarbons. *J Exposure Anal Environ Epidemiol* 1999: 9: 312–321.

Fan Z.H., Lioy P., Weschler C., Fiedler N., Kipen H., and Zhang J.F. Ozone-initiated reactions with mixtures of volatile organic compounds under simulated indoor conditions. *Environ Sci Technol* 2003: 37: 1811–1821.

Funasaka K., Miyazaki T., Tsuruho K., Tamura K., Mizuno T., and Kuroda K. Relationship between indoor and outdoor carbonaceous particulates in roadside households. *Environ Sci Technol* 2000: 110: 127–134.

Geller M.D., Chang M.H., Sioutas C., Ostro B.D., and Lipsett M.J. Indoor/outdoor relationship and chemical composition of fine and coarse particles in the Southern California deserts. *Atmos Environ* 2002: 36: 1099–1110.

Gotschi T., Oglesby L., Mathys P., Monn C., Manalis N., Koistinen K., Jantunen M., Hanninen O., Polanska L., and Kunzli N. Comparison of black smoke and $PM_{2.5}$ levels in indoor and outdoor environments of four European cities. *Environ Sci Technol* 2002: 36: 1191–1197.

Ho K.F., Cao J.J., Harrison R.M., Lee S.C., and Bau K.K. Indoor/outdoor relationships of organic carbon (OC) and elemental carbon (EC) in $PM_{2.5}$ in roadside environment of Hong Kong. *Atmos Environ* 2004: 38: 6327–6335.

Jones N.C., Thornton C.A., Mark D., and Harrison R.M. Indoor/outdoor relationships of particulate matter in domestic homes with roadside, urban and rural locations. *Atmos Environ* 2000: 34: 2603–2612.

Kinney P., Chillrud S., Ramstrom S., Ross J., and Spengler J. Exposure to multiple air toxics in New York city. *Environ Health Perspec* 2002: 110(Suppl. 4): 539–546.

Klepeis N.E., Nelson W.C., Ott W.R., Robinson J.P., Tsang A.M., Switzer P., Behar J.V., Hern S.C., and Engelmann W.H. The national human activity pattern survey (NHAPS): a resource for assessing exposure to environmental pollutants. *J Exposure Anal Environ Epidemiol* 2001: 11: 231–252.

Landis M.S., Norris G.A., Williams R.W., and Weinstein J.P. Personal exposures to $PM_{2.5}$ mass and trace elements in Baltimore, MD, USA. *Atmos Environ* 2001: 35: 6511–6524.

Lee J.H., Yoshida Y., Turpin B.J., Hopke P.K., Poirot R.L., Lioy P.J., and Oxley J.C. Identification of sources contributing to Mid-Altantic regional aerosol. *J Air Waste Manag Assoc* 2002: 52: 1186–1205.

Li C.S., and Lin C.H. Carbon profile of residential indoor PM_1 and $PM_{2.5}$ in the subtropical region. *Atmos Environ* 2003: 37: 881–888.

Lim H.J., and Turpin B.J. Origins of primary and secondary organic aerosol in Atlanta: results of time-resolved measurements during the Atlanta supersite experiment. *Environ Sci Techn* 2002: 36: 4489–4496.

Lim H.J., Turpin B.J., Russell L.M., and Bates T.S. Organic and elemental carbon measurements during ACE-ASIA suggest a longer atmospheric lifetime for elemental carbon. *Environ Sci Technol* 2003: 37: 3055–3061.

Liu W., Zhang J., Zhang L., Turpin B., Weisel C., Morandi M., Stock T., Colome S., and Korn L. Estimating contributions of indoor and outdoor sources to indoor carbonyl concentrations in three urban areas of the United States. *Atmospheric Environ* 2006, accepted.

Lunden M.M., Revzan K.L., Fischer M.L., Thatcher T.L., Littlejohn D., Hering S.V., and Brown N.J. The transformation of outdoor ammonium nitrate aerosols in the indoor environment. *Atmos Environ* 2003: 37: 5633–5644.

Mader B.T., and Pankow J.F. Gas/solid partitioning of semivolatile organic compounds (SOCs) to air filters. 3. An analysis of gas adsorption artifacts in measurements of atmospheric SOCs when using Teflon membrane filters and quartz fiber filters. *Environ Sci Technol* 2001: 35: 3422–3432.

Mader B.T., Schauer J.J., Seinfeld J.H., Flagan R.C., Yu J.Z., Yang H., Lim H.J., Turpin B.J., Deminter J.T., Heidemann G., Bae M.S., Quinn P., Bates T., Eatough D.J., Huebert B.J., Bertram T., and Howell S. Sampling methods used for the collection of particle-phase organic and elemental carbon during ACE-Asia. *Atmos Environ* 2003: 37: 1435–1449.

McDow S.R., and Huntzicker J.J. Vapor adsorption artifact in the sampling of organic aerosol: face velocity effects. *Atmos Environ* 1990: 24A: 2563–2571.

Meng Q.Y., Turpin B.J., Korn L., Weisel C.P., Morandi M., Colome S., Zhang J.F.J., Stock T., Spektor D., Winer A., Zhang L., Lee J.H., Giovanetti R., Cui W., Kwon J., Alimokhtari S., Shendell D., Jones J., Farrar C., and Maberti S. Influence of ambient (outdoor) sources on residential indoor and personal $PM_{2.5}$ concentrations: Analyses of RIOPA data. *J Exposure Anal Environ Epidemiol* 2005a: 15: 17–28.

Meng Q.Y., Turpin B.J., Polidori A., Lee J.H., Weisel C., Morandi M., Colome S., Stock T., Winer A., and Zhang J. $PM_{2.5}$ of ambient origin: estimates and exposure errors relevant to PM epidemiology. *Environ Sci Technol* 2005b: 39: 5105–5112.

Naumova Y.Y., Eisenreich S.J., Turpin B.J., Weisel C.P., Morandi M.T., Colome S.D., Totten L.A., Stock T.H., Winer A.M., Alimokhtari S., Kwon J., Shendell D., Jones J., Maberti S., and Wall S.J. Polycyclic aromatic hydrocarbons in the indoor and outdoor air of three cities in the US. *Environ Sci Technol* 2002: 36: 2552–2559.

Naumova Y.Y., Offenberg J.H., Eisenreich S.J., Meng Q.Y., Polidori A., Turpin B.J., Weisel C.P., Morandi M.T., Colome S.D., Stock T.H., Winer A.M., Alimokhtari S., Kwon J., Maberti S., Shendell D., Jones J., and Farrar C. Gas/particle distribution of polycyclic aromatic hydrocarbons in coupled outdoor/indoor atmospheres. *Atmos Environ* 2003: 37: 703–719.

Nazaroff W.W., and Weschler C.J. Cleaning products and air fresheners: exposure to primary and secondary air pollutants. *Atmos Environ* 2004: 38: 2841–2865.

Offenberg J.H., Naumova Y.Y., Turpin B.J., Eisenreich S.J., Morandi M.T., Stock T., Colome S.D., Winer A.M., Spektor D.M., Zhang J., and Weisel C.P. Chlordanes in the indoor and outdoor air of three US cities. *Environ Sci Technol* 2004: 38: 2760–2768.

Ott W., Wallace L., and Mage D. Predicting particulate (PM_{10}) personal exposure distributions using a random component superposition statistical model. *J Air Waste Manage Assoc* 2000: 50: 1390–1406.

Ozkaynak H., Xue J., Spengler J., Wallace L., Pellizzari E., and Jenkins P. Personal exposure to airborne particles and metals: results from the particle TEAM study in Riverside, California. *J Exposure Anal Environ Epidemiol* 1996: 6: 57–78.

Pandis S.N., Harley R.A., Cass G.R., and Seinfeld J.H. Secondary organic aerosol formation and transport. *Atmos Environ* 1992: 26A: 2269–2282.

Polidori A., Turpin B.J., Lim H.J., Cabada J.C., Subramanian R., Robinson A.L., and Pandis S.N. Local and regional secondary organic aerosol: Insights from a year of semi-continuous carbon measurements at Pittsburgh. *Aerosol Sci Technol* 2006, submitted.

Reff A. PM_{25} exposure assessment using FTIR spectroscopy. PhD Dissertation Rutgers University: New Brunswick, NJ, 2005.

Reff A., Turpin B.J., Porcja R.J., Giovennetti R., Cui W., Weisel C.P., Zhang J., Kwon J., Alimokhtari S., Morandi M., Stock T., Maberti S., Colome S., Winer A., Shendell D., Jones J., and Farrar C. Functional group characterization of indoor, outdoor, and personal $PM_{2.5}$: results from RIOPA. *Indoor Air* 2005: 15: 53–61.

Robinson J., and Nelson W.C. *National Human Activity Pattern Survey Data Base*. USEPA: Research Triangle Park, NC, 1995.

Rogge W.F., Mazurek M.A., Hildemann L.M., and Cass G.R. Quantification of urban organic aerosols at a molecular level: identification, abundance and seasonal variation. *Atmos Environ* 1993: 27(Part A): 1309–1330.

Sakai R., Siegmann H.C., Sato H., and Voorhees A.S. Particulate matter and particle-attached polycyclic aromatic hydrocarbons in the indoor and outdoor air of Tokyo measured with personal monitors. *Environ Res* 2002: 89: 66–71.

Subramanian R., Khlystov A.Y., Cabada J.C., and Robinson A.L. Positive and negative artifacts in particulate organic carbon measurements with denuded and undenuded sampler configurations. *Aerosol Sci Technol* 2004: 38(Suppl. 1): 27–48.

Thatcher T.L., and Layton D.W. Deposition, resuspension, and penetration of particles within a residence. *Atmos Environ* 1995: 29: 1487–1497.

Turpin B.J., Hering S.V., and Huntzicker J.J. Investigation of organic aerosol sampling artifacts in the Los Angeles Basin. *Atmos Environ* 1994: 28: 3061–3071.

Turpin B.J., and Huntzicker J.J. Identification of secondary organic aerosol episodes and quantitation of primary and secondary organic aerosol concentrations during PAQS. *Atmos Environ* 1995: 29: 3527–3544.

Turpin B.J., and Lim H.J. Species contributions to $PM_{2.5}$ mass concentrations: revisiting common assumptions for estimating organic mass. *Aerosol Sci Technol* 2001: 35: 602–610.

Turpin B.J., Saxena P., and Andrews E. Measuring and simulating particulate organics in the atmosphere: problems and prospects. *Atmos Environ* 2000: 34: 2983–3013.

USEPA. *Air Quality Criteria for Particulate Matter*. US Environmental Protection Agency: Research Triangle Park, NC, 2004.

Weisel C.P., Zhang J., Turpin B.J., Morandi M.T., Colome S., Stock T.H., Spektor D.M., Korn L., Winer A., Alimokhtari S., Kwon J., Mohan K., Harrington R., Giovanetti R., Cui W., Afshar M., Maberti S., and Shendell D. The relationships of indoor, outdoor and personal air (RIOPA) study: study design, methods, and quality assurance/control results. *J Exposure Anal Environ Epidemiol* 2004: 15: 123–137.

Weschler C.J. Chemical reactions among indoor pollutants: what we've learned in the new millennium. *Indoor Air* 2004: 14(Suppl. 7): 184–194.

Weschler C.J., and Shields H.C. Potential reactions among indoor pollutants. *Atmos Environ* 1997: 31: 3487–3495.

Yocom J.E. Indoor-outdoor air quality relationships, A critical review. *J Air Pollution Control Assoc* 1982: 32: 500–520.