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Indoor/Outdoor Relationships, Trends, and Carbonaceous Content of Fine Particulate Matter in Retirement Homes of the Los Angeles Basin

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ABSTRACT

Hourly indoor and outdoor fine particulate matter ($PM_{2.5}$), organic and elemental carbon (OC and EC, respectively), particle number (PN), ozone (O_3), carbon monoxide (CO), and nitrogen oxide (NO_x) concentrations were measured at two different retirement communities in the Los Angeles, CA, area as part of the Cardiovascular Health and Air Pollution Study. Site A (group 1 [G1]) was operated from July 6 to August 20, 2005 (phase 1 [P1]) and from October 19 to December 10, 2005 (P2), whereas site B (group 2 [G2]) was operated from August 24 to October 15, 2005 (P1), and from January 4 to February 18, 2006 (P2). Overall, the magnitude of indoor and outdoor measurements was similar, probably because of the major influence of outdoor sources on indoor particle and gas levels. However, G2 showed a substantial increase in indoor OC, PN, and $PM_{2.5}$ between 6:00 and 9:00 a.m., probably from cooking. The contributions of primary and secondary OC (SOA) to measured outdoor OC were estimated from collected OC and EC concentrations using EC as a tracer of primary combustion-generated OC (i.e., "EC tracer method"). The study average outdoor SOA accounted for 40% of outdoor particulate OC (40–45% in the summer and 32–40% in the winter). Air exchange rates (hr^{-1}) and infiltration factors (F_{inf}; dimensionless) at each site were also determined. Estimated F_{inf} and measured particle concentrations were then used in a single compartment mass balance model to assess the contributions of indoor and/or outdoor sources to measured indoor OC, EC, $PM_{2.5}$, and PN. The average percentage contributions of indoor SOA of outdoor origin to measured indoor OC were ~35% (during G1P1 and G1P2) and ~45% (for G2P1 and G2P2). On average, 36% (G2P1) to 44% (G1P1) of measured indoor OC was composed of outdoor-generated primary OC.

IMPLICATIONS

In the Los Angeles basin, outdoor SOA represents a substantial contributor to both indoor and outdoor OC from summer to winter. This is an important component of PM, because recent epidemiologic studies have linked exposure to SOA to respiratory inflammation. Also, $PM_{2.5}$ and its carbonaceous components (i.e., OC and EC) are characterized by different $F_{in[inf]f}$, and indoor sources of $PM_{2.5}$ and OC might be substantial. Thus, the composition of indoor and outdoor particles is different, and outdoor $PM_{2.5}$ concentrations may not adequately represent personal exposure to outdoor-infiltrated $PM_{2.5}$ in indoor environments. Implications to PM epidemiology warrant further investigation.

INTRODUCTION

Numerous epidemiologic studies have found associations between atmospheric aerosol concentrations and both acute and chronic adverse respiratory and cardiovascular effects.[1] Exposure to fine particulate matter ([PM] $PM_{2.5}$) and its components have also been investigated in many toxicological studies on the following: (1) human volunteers exposed to concentrated outdoor PM under controlled conditions[2]; (2) in vivo laboratory animal studies; and (3) in vitro tissue studies using well-characterized particles containing individual compounds or source mixtures.[3] $PM_{2.5}$ properties and components that are believed to be responsible for the observed adverse health effects include mass, surface area, size, metals, acids, organic compounds (organic carbon [OC]), elemental carbon [EC], sulfate and nitrate salts, peroxides, soot, and bioaerosols.[1,4]

The air quality standards established by the U.S. Environmental Protection Agency in 1997 were primarily based on epidemiologic studies conducted at stationary outdoor monitoring sites. However, a significant portion of human exposures to $PM_{2.5}$ occurs indoors where people spend ~85–90% of their time.[5,6] Thus, understanding the composition, behavior, and origin of indoor $PM_{2.5}$ is important to exposure characterization and mitigation.

Typically, indoor $PM_{2.5}$ consists of ambient (outdoor) particles that have infiltrated indoors, particles emitted indoors (primary), and particles formed indoors (secondary) from precursors emitted both indoors and outdoors.[7–9] Because of indoor sources such as cooking, smoking, gas stoves, cleaning, washing, and other human activities, $PM_{2.5}$ concentrations can be substantially higher indoors than outdoors.[1,7] A few recent studies have demonstrated that indoor sources make a substantial contribution to the indoor concentrations of $PM_{2.5}$ and its components, often >50%.[1,9–11] Because outdoor particles can enter the building envelope by convective flow (e.g., open windows) or by diffusional flow/infiltration (e.g., cracks and fissures), outdoor $PM_{2.5}$ is also a major contributor to indoor particle concentrations.[1,12,13] Recent epidemiologic panel studies have demonstrated the usefulness of separating total personal particle exposures into their ambient (outdoor origin) and nonambient (indoor generated) components. Typically only associations between adverse health outcomes and ambient particle exposures have been found.[14,15]

Organic compounds make an important but poorly understood contribution to indoor and outdoor $PM_{2.5}$ and are believed to be a key factor in causing adverse health effects.[1] They consist of OC and EC and are composed of hundreds of individual compounds with different physical and chemical properties. Although EC is produced only during incomplete combustion and emitted directly in the particle phase, indoor and outdoor OC are both emitted from combustion sources (primary organic aerosols) and formed from semivolatile and low-volatility products of chemical reactions involving reactive organic gases (secondary organic aerosols [SOAs]).[16] Quantifying the SOA contribution to measured OC both indoors and outdoors is important to linking the organic PM concentration to its emissions and precursors and to developing effective control strategies for PM.

The present work was funded by the National Institutes of Health (NIH) and was conducted within the Cardiovascular Health and Air Pollution Study (CHAPS), a multidisciplinary project of which the goals are to investigate the effects of microenvironmental exposures to PM on cardiovascular outcomes in elderly retirees affected by coronary heart disease (CHD). The elderly population with CHD is likely to be among the most vulnerable to the adverse effects of particulate air pollutants.

In this paper, hourly indoor and outdoor $PM_{2.5}$, OC, EC, particle number (PN), ozone (O_3), carbon monoxide (CO), and nitrogen oxide (NO_X) concentrations were measured at two different retirement communities in the Los Angeles area and used to provide new insight into the following: (1) the relationships between indoor and outdoor $PM_{2.5}$, its components, and their seasonal variations, as well as their association with gaseous copollutants; (2) the contributions of primary OC and SOA to measured outdoor OC; and (3) the relative importance of outdoor and indoor PM sources to measured indoor OC, EC, $PM_{2.5}$, and PN concentrations. The results obtained in this paper will be used to determine personal exposure to outdoor-infiltrated $PM_{2.5}$ and its particulate components and to indoor-generated $PM_{2.5}$ and its particulate components in elderly retirees with a history of CHD.

METHODS

Study Design

As a part of CHAPS, the physical and chemical characteristics of indoor and outdoor $PM_{2.5}$ were investigated at two different retirement communities in Southern California. Site A for subject group 1 (G1) was located ~30 mi east of downtown Los Angeles, in a residential area, ~2 mi away from any major freeways and close to a construction site. Site B for subject group 2 (G2) was located ~5 mi east of downtown Los Angeles, ~0.1 mi south of a major freeway. Two 6-week sampling campaigns were conducted at each site; site A (G1) was operated from July 6 to August 20, 2005 (phase 1 [P1]), and from October 19 to December 10, 2005 (phase 2 [P2]), whereas site B (G2) was operated from August 25 to October 15, 2005 (P1), and from January 4 to February 18, 2006 (P2). Thus, we were able to study the seasonal variations in the indoor/outdoor relationships of $PM_{2.5}$ and its components.

Two identical sampling stations were installed at each location, one indoors and one outdoors. The indoor sampling station at site A was located in a recreational area of the first community's main building, adjacent to a construction site where work was ongoing. The indoor sampling area at site B was situated in the dinning room of the second community's main building. At both sites, the outdoor station, set up inside a movable trailer, was positioned within 300 m from the indoor station.

Instrumentation

At both indoor and outdoor sampling areas, a water-based condensation particle counter (CPC Model 3785, TSI Inc), providing continuous (1 min) PN concentrations (operating flow rate = 1 L/min), and a semicontinuous OC_EC analyzer (Model 3F, Sunset Laboratory Inc.) were operated side by side. The two CPCs were examined at the University of Southern California laboratory before being deployed in the field and showed high internal precision. The OC_EC analyzers were placed downstream of a $PM_{2.5}$ cyclone and collected samples at an approximate flow rate of 8 L/min. Particulate OC and EC were measured in hourly cycles (i.e., sampling time = 45 min; analysis time = 15 min). A multichannel parallel carbon plate diffusion denuder (provided by the manufacturer) was placed upstream of the OC_EC instrument to remove most of the organic vapors in the sampled air. The setup and the standard operating procedure for the semicontinuous carbon analyzer are described in more details in Arhami et al.[17] A modified National Institute for Occupational Safety and Health (NIOSH) analysis protocol was used here to evolve particulate OC and EC. This protocol consists of four temperature steps in the He analysis segment and allows for the separation of particulate OC into four response peaks representing different volatility fractions of OC.[18,19] These four OC peaks are designated and recorded as peak 1 to peak 4 (OC_1 to OC_4). For the purposes of this study, OC_2, OC_3, and OC_4 were summed (OC_{2-4}) and considered as the least volatile OC fractions, whereas OC_1 represented the most volatile OC fraction. The internal precision of the two OC_EC analyzers (examined before the beginning of CHAPS by running them side by side) was high (R^2 of 0.98 and 0.97 for thermal OC and EC, respectively). A detailed description of all of the

quality control and quality assurance analyses performed with the semicontinuous carbon analyzer is reported in the Supplemental Information along with the corresponding results.

Hourly $PM_{2.5}$ mass concentrations were measured by Bete-attenuation mass monitors (BAMS) (Model 1020, Met One Instruments Inc.) at a flow rate of 16.7 L/min. Two BAMs were used at each of the indoor and outdoor sampling stations to examine the uncertainty of the collected data. Continuous (1-min) NO and NO_2 measurements were obtained both indoors and outdoors by using Thermo Environmental NO_x Analyzers (Model 42, Thermo Environmental Instruments Inc.). Dasibi Carbon Monoxide Analyzers (Model 3008, Dasibi Environmental Corp.) were implemented to measure continuous (1-min) indoor and outdoor CO levels. Continuous (1-min) outdoor O_3 concentrations were also monitored at each sampling station by using API Ozone Analyzers (Model 400A, Teledyne Technologies Inc.).

Data Analysis

To match the OC, EC, and $PM_{2.5}$ measurements, only hourly arithmetic averages of the highly resolved PN and gaseous copollutants (CO, NO_x, and O_3) concentrations were considered. Then, a comprehensive indoor and outdoor database was constructed for each group and phase of CHAPS to analyze the relationships between measured indoor and outdoor particulate and gaseous species and to facilitate the overall data analysis.

The contributions of primary OC and SOA to measured outdoor OC were estimated from collected OC and EC concentrations using EC as a tracer of primary combustion-generated OC (i.e., "EC tracer method").[20–23] This method assumes that primary OC and EC are emitted from the same combustion sources. Data points characterized by high CO and NO peaks, mainly observed during rush hour traffic, were used to identify periods dominated by primary sources, when SOA is less likely to be formed. By regressing the OC and EC data collected during these time periods, the characteristic primary OC/EC ratio for each month of CHAPS was determined. Because a conventional linear least-squares regression assumes that there are uncertainties only in the dependent variable, a Deming linear least-squares regression[24,25] was used instead, and the uncertainties in OC and EC were assumed equal. Thus, primary OC (OC_{pri}) and SOA can be estimated by the following expressions:

$$OC_{pri} = a \times EC + b \tag{1}$$

$$SOA = OC - OC_{pri}, \tag{2}$$

where $a = (OC/EC)_{pri}$ = characteristic primary OC/EC ratio for the study area, and b = noncombustion primary OC. Typically, the SOA values estimated through this method vary with season and location and are generally higher during the afternoon hours of summertime photochemical smog episodes (e.g., in the Los Angeles basin) and at locations that are recipients of long-distance transport (e.g., the Eastern United States).

The indoor-outdoor air exchange rates ([AER] hr^{-1}) at each site were estimated from indoor CO measurements collected during periods affected by a dominant indoor source. Only time periods when the CO concentration peaked at values significantly higher than the background CO level and was followed by a nonsource period (mostly observed in the morning and probably associated with cooking activities) were considered in our calculations. Assuming an exponential decay of particles, that AER and outdoor concentrations are constant during the decay period, and that indoor concentrations are well mixed, then:

$$C_t = e^{-(AER + k)t}\, C_0 \tag{3}$$

or

$$\ln C_t = -(AER + k)t + \ln C_0, \tag{4}$$

where C_t is the indoor CO concentration after time t (after the decay period), C_0 is the initial peak CO concentration (right after CO emission), and k is the indoor loss rate for particles or gases (hr^{-1}).[13] Because k is rather negligible for CO, it was possible to estimate the AERs for the two sites directly from the above-mentioned eq (4) by regressing $ln\ C_t$ over $ln\ C_0$.

The infiltration factor (F_{inf}, defined as the equilibrium fraction of ambient particles that penetrate indoors and remain suspended)[26] is a key determinant of the indoor concentrations of particulate species. F_{inf} is described by the following equation:

$$F_{inf} = P(AER)/(AER + k) \tag{5}$$

where P is the penetration coefficient (dimensionless). F_{inf} for $PM_{2.5}$ varies with particle composition, particle size and volatility, surface-to-volume ratio of the indoor sampling location, and indoor air speed. F_{inf} is typically highest for nonvolatile species such as EC.[27,28] To estimate F_{inf} for OC, EC, $PM_{2.5}$, and PN, two different techniques were used: an analysis of the indoor/outdoor concentration ratios and the recursive model (RM) developed by Allen et al.[29] In the first approach, hourly indoor/outdoor ratios (I/O) for each particulate species were determined at times when no indoor particle sources, such as cooking or cleaning, were likely to be present (i.e., only I/O ratios ≤ 1 were considered). Daily F_{inf} estimates were then obtained by averaging these segregated hourly I/O ratios. Mean F_{inf} for each group and phase of the study was also determined by averaging the corresponding daily values. To verify these results, the same analysis of the I/O concentration ratios was then repeated by using only nighttime data (from 12:00 midnight to 6:00 a.m.), because at this time, resident activities causing indoor particle generation were expected to be minimal. Conversely, the RM method, which has been validated recently for estimating F_{inf} for $PM_{2.5}$ from hourly light scattering data,[30] states that, for a particular species of interest, the average indoor concentration during hour t (C_t^{in}) is equal to the sum of a fraction of the average outdoor concentration during the

same hour (C_t^{out}), a fraction of the average indoor concentration remaining from the previous hour (C_{t-1}^{in}), and the contribution from indoor sources (S_t^{in}):

$$C_t^{in} = a_1 C_t^{out} + a_2\, C_{t-1}^{in} + S_t^{in} \tag{6}$$

where

$$a_1 = F_{inf}(1 - a_2) \tag{7}$$

and

$$a_2 = \exp[-(\mathrm{AER} + k)\Delta t] \tag{8}$$

Algorithms are used to identify and minimize the influence of hours when the indoor concentration is influenced by indoor sources, thus eliminating the S_t^{in} term from eq 6:

$$C_t^{in} = a_1\, C_t^{out} + a_2\, C_{t-1}^{in} \tag{9}$$

The coefficients a_1 and a_2 are then estimated via multiple linear regression of eq 9, and F_{inf} is calculated from a_1 and a_2 using the following relationship:

$$F_{inf} = \frac{a_1}{1 - a_2} \tag{10}$$

Finally, a single compartment mass balance model[9–11] was used to assess the mean contributions of indoor and outdoor sources to measured indoor OC, EC, $PM_{2.5}$, and PN concentrations. Under the assumption of perfect instantaneous mixing and that the factors affecting the indoor concentrations are constant or change slowly with time, the steady state indoor concentration of any particulate species can be described by the following eq:

$$C_{in} = \frac{P(\mathrm{AER})C_{out}}{\mathrm{AER} + k} + \frac{Q_i/V}{\mathrm{AER} + k} = F_{inf}C_{out} + C_{ig} = C_{og} + C_{ig} \tag{11}$$

where C_{in} is the indoor concentration of the species of interest (micrograms per cubic meter), C_{out} is the corresponding outdoor concentration (micrograms per cubic meter), F_{inf} is the corresponding infiltration factor (estimated for each species as described previously; dimensionless), C_{ig} is the indoor-generated concentration for the same species found indoors, and C_{og} is the outdoor-generated concentration for the same species found indoors. Typically, in the mass balance model, F_{inf} is given by eq 5, and C_{ig} is expressed by $Qi/V(a+k)$, where Q_i is the indoor source strength (micrograms per hour), and V is the house volume (cubic meter).

RESULTS AND DISCUSSION

Particle and Gaseous Measurements

The minimum, maximum, average, and standard deviation of all hourly particle and gas data obtained for all groups and phases of CHAPS are presented in Table 1. Overall, the magnitude of indoor measurements was similar to that of outdoor measurements for most phases of the study, which highlights the major effect of outdoor sources on indoor levels.[9,11,31,32] Although at site B (G2) a wider range in indoor $PM_{2.5}$, PN, and OC concentrations was observed compared with the outdoor levels, the average indoor and outdoor concentrations were still comparable. The difference between indoor and outdoor average concentrations was lowest for CO and NO_x (except

Table 1. Mean ± 1σ (standard deviation), minimum, and maximum of all hourly particle and gas indoor and outdoor data collected during each group and phase of CHAPS.

Mean ± 1σ Min to Max	G1P1		G2P1		G1P2		G2P2	
	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
OC (μgC/m³)	5.91 ± 2.06 2.26–16.35	5.21 ± 1.80 1.72–18.17	6.75 ± 4.55 2.24–61.75	7.07 ± 3.13 1.54–22.20	5.46 ± 2.68 0.67–26.92	5.43 ± 2.89 0.97–29.10	5.25 ± 3.86 0.92–73.21	7.06 ± 3.32 0.45–23.20
EC (μgC/m³)	1.43 ± 1.03 0.20–8.53	1.42 ± 0.73 0.20–5.98	1.24 ± 0.76 0.20–5.5	1.76 ± 1.21 0.20–10.03	1.42 ± 0.84 0.20–5.61	1.65 ± 1.05 0.20–8.87	1.18 ± 0.79 0.20–5.76	1.68 ± 1.16 0.20–6.48
OC_1 (μgC/m³)	2.97 ± 1.03 1.47–8.53	2.27 ± 0.76 0.52–6.54	3.3 ± 2.1 1.25–34.08	3.16 ± 1.34 0.91–9.98	2.35 ± 0.99 0.12–10.47	2.23 ± 1.24 0.49–12.79	2.41 ± 1.77 0.62–38.13	2.82 ± 1.32 0.43–8.66
OC_{2-4} (μgC/m³)	2.88 ± 1.34 0–10.74	2.78 ± 1.36 0–11.57	3.32 ± 2.58 0–27.65	3.41 ± 2.25 0.91–15.47	2 ± 2.07 0–17.57	2.67 ± 1.94 0.49–17.29	2.44 ± 2.24 0–35.07	3.83 ± 2.38 0–15.04
PN (ptcl#/cm³)	10,643 ± 4826 2166–43,140	13,044 ± 3442 2577–23,854	13,424 ± 4776 307–2E + 05	14,017 ± 6297 738–61,001	13,724 ± 9871 2517–2E + 05	13,542 ± 5499 2649–44,523	19,000 ± 28,400 2191–4E + 05	22,732 ± 8923 3524–62,588
$PM_{2.5}$ (μg/m³)	19.73 ± 9.66 1–194	30.83 ± 11.43 6–72	37.51 ± 27.26 5–197	24.86 ± 13.47 1–81	22.16 ± 15.15 1–136	24.82 ± 22.42 1–132	12.66 ± 9.58 1–82	19.65 ± 17.07 1–100
CO (ppm)	0.64 ± 0.23 0.1–2.1	0.68 ± 0.32 0–2	0.78 ± 0.42 0.1–3.9	0.65 ± 0.39 0.1–3	0.71 ± 0.39 0–2.9	0.7 ± 0.4 0–4.2	0.98 ± 0.61 0–4	0.81 ± 0.56 0.1–3.4
NO_X (ppb)	38.81 ± 24.66 5–163	40.57 ± 22.92 8–184	35.56 ± 34.53 0–254	58.79 ± 44.02 6–300	59.19 ± 36.34 2–260	57.58 ± 38.73 3–272	84.63 ± 65.03 1–484	84 ± 61.85 2–356
Ozone (ppb)		30.25 ± 26.97 2–122		22.3 ± 20.6 1–112		14.08 ± 12.77 0–66		22.66 ± 12.46 12–61

NO_x in G2P1), which suggests the absence of an important indoor source of these two gases and that their penetration losses were not significant. Conversely, the differences between indoor and outdoor particle levels were higher, probably because of the presence of indoor sources and/or changes in concentration because of transportation of particles indoors from outdoors.

The average hourly diurnal variations for data collected during the first phase of CHAPS at site A (G1P1; from July 6 to August 22, 2005) are shown in Figure 1. Generally, indoor and outdoor particle and gas concentrations tracked each other well, with a better agreement for gases. Slightly higher OC levels were measured indoors, mainly because of indoor contributions of the most volatile OC fraction (OC_1; not shown). Indoor EC, $PM_{2.5}$, and PN concentrations slightly increased from 10:00 a.m. to 12:00 p.m., probably because of the emission contributions of a construction site located right outside the indoor sampling area. Significant EC, PN, $PM_{2.5}$, CO, and NO_x peaks occurred concurrently outdoors and indoors during the morning rush hour traffic, suggesting that outdoor primary pollutants were important contributors to the indoor air. Indoor and outdoor OC concentrations increased from noon to 4:00 p.m.,

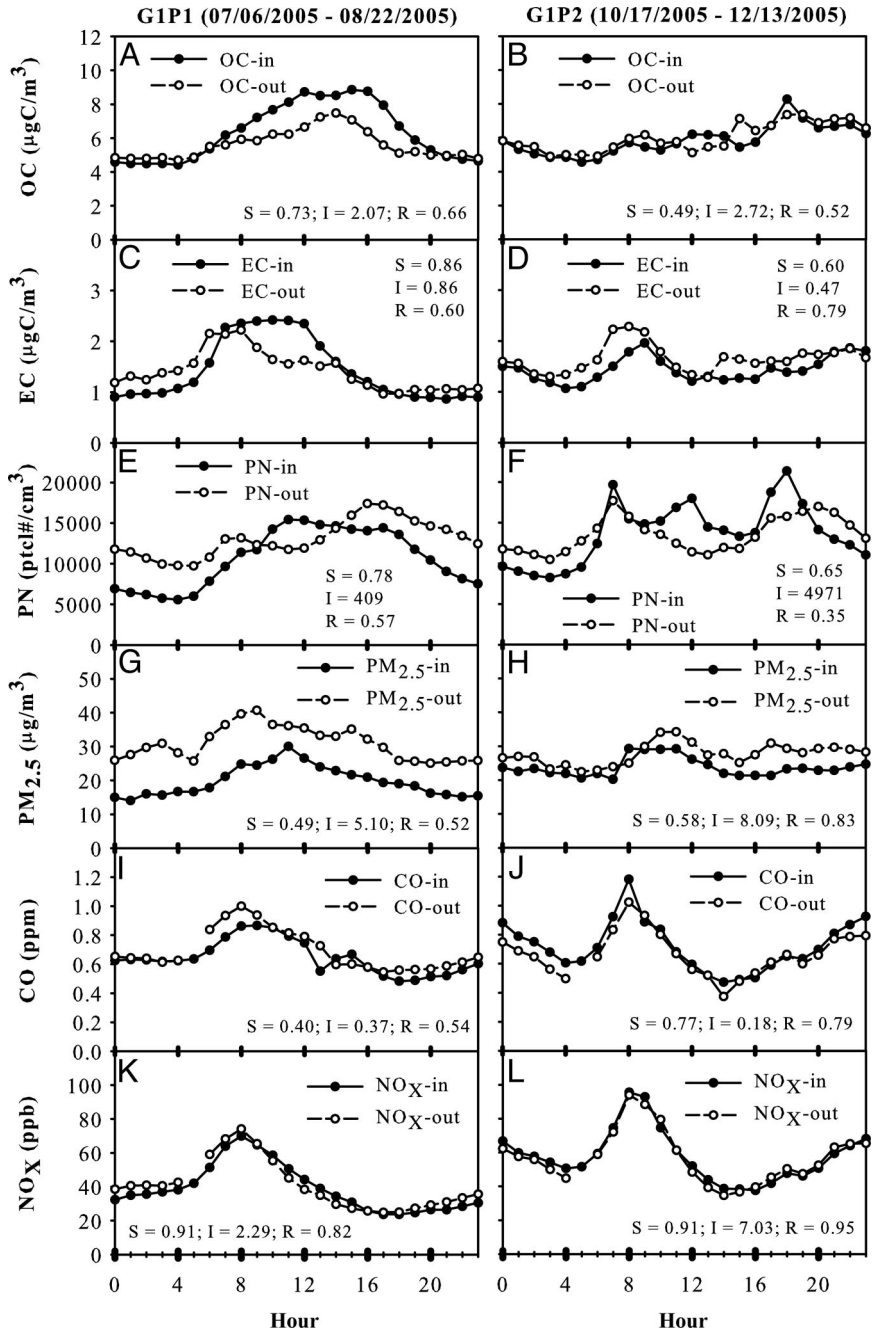

Figure 1. Average hourly diurnal variations for particle and gas data collected during the first and the second phase of CHAPS at site A (G1P1 and G1P2, respectively). The slope (*S*), intercept (*I*), and Pearson correlation coefficient (*R*) for all indoor vs. outdoor concentrations are also reported. (A) G1P1 OC; (B) G1P2 OC; (C) G1P1 EC; (D) G1P2 EC; (E) G1P1 PN; (F) G1P2 PN; (G) G1P1 $PM_{2.5}$; (H) G1P2 $PM_{2.5}$; (I) G1P1 CO; (J) G1P2 CO; (K) G1P1 NO_x; (L) G1P2 NO_x.

probably as a result of photochemical OC formation in the afternoon. This mechanism of particle generation may also explain the increase in outdoor PN and $PM_{2.5}$ between 3:00 and 5:00 p.m. that was not observed for EC, CO, and NO_x.

The average indoor and outdoor diurnal patterns at site A during the second phase of the study (G1P2; from October 17 to December 13, 2005) generally followed the same trends as those observed during G1P1 (Figure 1), which strengthen the hypothesis that at this location the majority of indoor particles and gases originated from outdoor sources. During G1P2, the CO and NO_X outdoor concentrations increased at midnight, most likely because of a lowered mixing height. This caused a subsequent increase in indoor gaseous levels.

Measurement results at site B during the first phase of CHAPS (G2P1, from August 23 to October 15, 2005) showed a substantial morning peak in indoor OC, PN, and $PM_{2.5}$ (Figure 2) between 6:00 and 9:00 a.m., probably from cooking activities in the kitchen adjacent the indoor sampling site where breakfast, lunch, and dinner were all cooked at this time by using gas stoves/ovens. Interestingly, cooking did not affect EC, CO, and NO_X, of which the indoor and outdoor levels were mostly influenced by

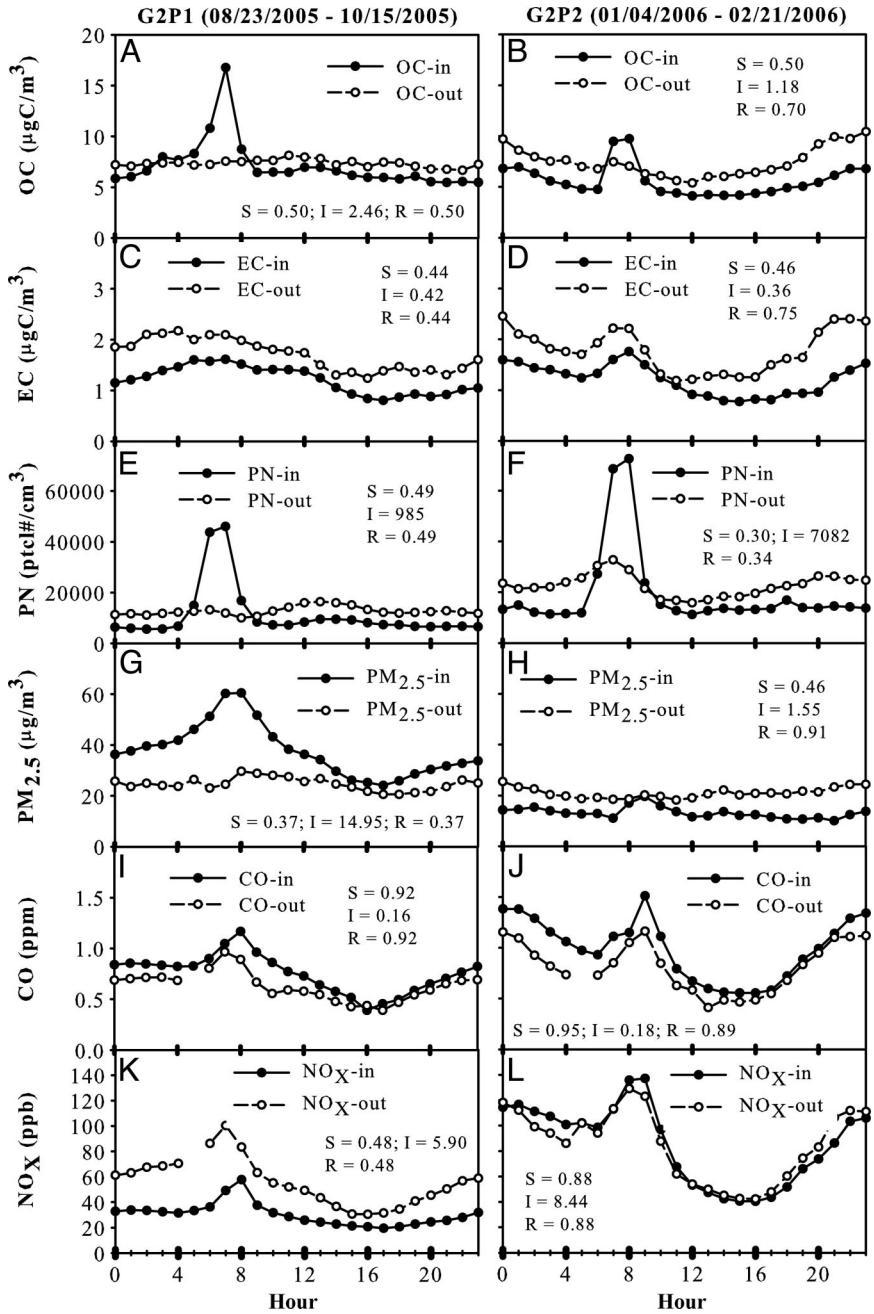

Figure 2. Average hourly diurnal variations for particle and gas data collected during the first and the second phase of CHAPS at site B (G2P1 and G2P2, respectively). The slope (*S*), intercept (*I*), and Pearson correlation coefficient (*R*) for all indoor vs. outdoor concentrations are also reported. (A) G2P1 OC; (B) G2P2 OC; (C) G2P1 EC; (D) G2P2 EC; (E) G2P1 PN; (F) G2P2 PN; (G) G2P1 $PM_{2.5}$; (H) G2P2 $PM_{2.5}$; (I) G2P1 CO; (J) G2P2 CO; (K) G2P1 NO_x; (L) G2P2 NO_x.

Table 2. Pearson correlation coefficients between particle concentrations ($PM_{2.5}$ and PN) and gas levels (CO and NO_X) for indoor and outdoor data collected during each group (G) and phase (P) of CHAPS.

	PN-CO		$PM_{2.5}$-CO		PN-NO_X		$PM_{2.5}$-NO_X	
Variable	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
G1P1	0.37	0.21	0.22	0.05	0.33	0.22	0.10	0.15
G1P2	0.07	0.38	0.40	0.28	0.15	0.49	0.04	0.07
G2P1	0.28	0.45	0.32	0.21	0.40	0.47	0.15	0.07
G2P2	0.11	0.49	0.37	0.44	0.19	0.57	0.15	0.35

morning rush hour traffic. With the exception of these morning peaks, indoor and outdoor particle and gas concentrations tracked each other well, although indoor levels were generally lower than outdoor levels. This suggests that indoor sources of OC, PN, and $PM_{2.5}$ were not significant other than during the morning cooking events and that indoor EC mainly originated from outdoor sources. Indoor cooking affected the concentrations of the least and the most volatile OC fractions (OC_1 and OC_{2-4}, respectively) equally (not shown).

Similar indoor and outdoor trends in particle and gas concentrations, including a morning increase in OC, PN, and $PM_{2.5}$ because of cooking, were also observed in the second phase of the study at site B (G2P2, from January 4, 2006, to February 21, 2006; Figure 2). Indoor and outdoor diurnal patterns for EC, CO, and NOx were virtually identical (no significant indoor sources) and peaked in the morning during rush hour traffic. Ground-level concentrations of all particulate and gaseous species increased significantly at midnight because of a decrease in the mixing height and an increase in the atmospheric stability, typical of wintertime conditions. Indoor particle mass levels were lower during G2P2 than during G2P1; the reasons for this discrepancy are unclear. Despite the substantial morning increase in indoor OC, PN, and $PM_{2.5}$ at site B because of cooking, the average indoor concentrations of these three species were not much higher than those measured outdoors (Table 1). This was the result of the smaller contribution of indoor sources during the rest of the day, which lowered the average indoor concentrations. Also, these observations are consistent with the low AER and F_{inf} values estimated in this work and discussed in a subsequent section.

Figures 1 and 2 also include the slope (S), intercept (I) and Pearson correlation coefficient (R) for indoor versus outdoor concentrations of all measured particulate and gaseous species and for all groups and phases of CHAPS. At site B, where indoor cooking significantly affected $PM_{2.5}$, PN, and OC concentrations, the morning data (5:00 to 9:00 a.m.) were not considered in the calculation of S, I, and R. Indoor gas concentrations usually showed the highest degree of correlation with the corresponding outdoor levels with S close to 1 in most cases, confirming that indoor NO_x and CO were mostly of outdoor origin. For OC, EC, $PM_{2.5}$, and PN, a positive I was obtained, suggesting the presence of significant indoor background concentrations for all particulate species. Indoor OC, OC_1, and OC_{2-4} measurements correlate well with the corresponding outdoor data, confirming the important effect of outdoor OC sources on indoor levels. A high R for the EC I/O correlations was always observed because indoor sources of EC were negligible at both sites. The highest slopes for EC were found during G1P1, probably because of a substantial outdoor contribution from the diesel vehicles operating at the construction site located right outside the indoor sampling station and between the retirement community building and the outdoor monitoring site. Therefore, the concentration of EC impacting the building shell was greater than that measured by the outdoor site. This, rather than an indoor source of EC, is thought to explain the indoor EC being higher than the outdoor EC for G1P1. Representative hourly diurnal variations of the $PM_{2.5}$, OC, and EC concentrations (micrograms per cubic meter; nonaveraged data) measured during each group and phase of CHAPS are reported in the Supplemental Information.

The correlation among gas levels, $PM_{2.5}$, and PN concentrations for indoor and outdoor data are presented in Table 2. Overall, the results show a weak Pearson correlation coefficient between particles and gases indoors. However, some modest correlations (R = ~0.4–0.5) between PN and NO_x and between PN and CO are observed outdoors during most phases of CHAPS, probably because these three species were mainly emitted from the same source (e.g., motor vehicles emission). These correlations are slightly higher at site B, which was located closer to a freeway.

Primary OC and SOA Estimations Outdoors

Figure 3 shows all semicontinuous outdoor OC and EC values obtained on July 2005 (G1P1). Data points were segregated into measurements dominated by primary

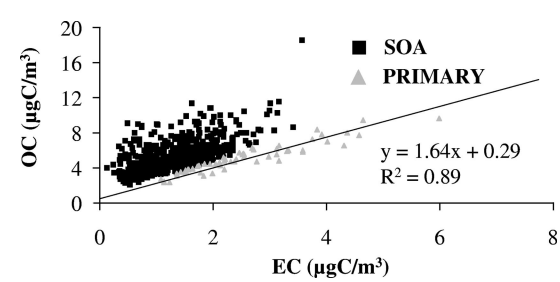

Figure 3. Semicontinuous outdoor OC and EC measurements made from July 6–31, 2005, at site A (G1P1). Black rectangles represent data influenced by a moderate or high probability of SOA formation. Gray triangles are measurements dominated by primary emissions. The line, equation, and coefficient of determination (R^2) were calculated by regressing only OC and EC data labeled as "PRIMARY" (a Deming regression was used). Similar scatter plots were obtained for each month of CHAPS.

emissions (gray) and measurements affected by SOA formation (black). Similar plots were obtained for each month of the study. By regressing OC on EC using only data dominated by primary emissions, we estimated the characteristic primary OC/EC ratios (*a* in eq 1) and noncombustion primary OC (*b* in eq 1) for each month during CHAPS (Table 3). Carbon data corresponding with time periods affected by rain were not considered in the determination of *a* and *b* because of the possibility of differential wet scavenging.[21,23] The average coefficient of determination (R^2) for all monthly regressions was 0.87 ± 0.06 (1σ), which adds confidence to our results.

Thus, using eqs 1 and 2 and the estimated *a* and *b* values, monthly averaged outdoor primary OC and SOA concentrations were obtained (Table 3). Polidori et al.[23] calculated that the variability in primary OC and SOA determined by using the method described above is ~10%. This estimate of precision only includes "model" uncertainties, and it would be higher if measurement uncertainties were also taken into account. Typically, uncertainties for primary and secondary OC calculations are on the order of 10–40%.[22,23]

During CHAPS, the average SOA concentration was 2.50 ± 1.94 (1σ) μgC/m^3, which represents 40% ± 22 (1σ) of the study average particulate OC. These results are likely to be representative of the entire San Gabriel Valley, for the SOA estimations presented in this work only refer to outdoor data, and none of the two CHAPS sites was affected by any specific local sources other than traffic emissions. The highest monthly average SOA concentration (3.30 μgC/m^3, or 40% of measured OC) was estimated between February 2 and 16, 2006 (G2P2), whereas the lowest average SOA concentration (1.88 μgC/m^3, or 35% of measured OC) was estimated between November 1 and December 9, 2005 (G1P2; see Table 3 for details).

Interestingly, the summertime percentage contributions of SOA to particulate OC obtained during CHAPS (40–45%) are higher than those estimated by Turpin and Huntzicker[20] in Claremont, CA (where SOA exceeded 40% of the daily OC only during the afternoon photochemical smog episodes) but are in good agreement with those obtained in Atlanta, GA, by Lim and Turpin[33] (44%) and in the Pittsburgh, PA, area by Cabada et al.[22] (35%) and Polidori et al.[23] (38%). In each case, a similar decision strategy was used to identify time periods dominated by primary OC and to estimate SOA. Thus, these recent results are quite consistent and suggest that, in the summertime, SOA represents a substantial fraction of measured OC both in the East and in the West of the United States. Figure 4a shows the time averaged diurnal pattern for estimated primary OC and SOA concentrations during G1P1 (typical of summertime conditions). The corresponding measured outdoor CO and O_3 concentrations are also reported. As expected, primary OC, CO, and NO (not shown) peaked between 5:00 and 11:00 a.m. because of rush hour traffic, whereas SOA and O_3 peaked between 1:00 and 7:00 p.m. because of the high photochemical activity occurring locally. CO and NO are considered tracers of local and regional combustion, whereas O_3 is used as a tracer for photochemical reactions.

During the wintertime, both the average SOA concentration (2.36 μgC/m^3 in January and 3.30 μgC/m^3 in February) and the average percentage contributions of SOA to particulate OC (32% in January and 40% in February; G2P2) were comparable to the corresponding average values estimated in the summertime (2.18 μgC/m^3 or 43% of measured OC in July and 2.16 μgC/m^3 or 40% of measured OC in August; G1P1; Table 3), but had very different diurnal patterns. These wintertime SOA results are significantly higher than those reported for the San Joaquin Valley, CA, by Strader et al.,[34] where SOA formation accounted for ~25% of wintertime OC, and for Pittsburgh, where the average wintertime SOA concentration was 24% of measured OC.[23] The time-averaged diurnal pattern for estimated primary OC and SOA concentrations during G2P2 (representative of wintertime conditions) is shown in Figure 4b. The corresponding measured CO and O_3 concentrations are also reported. Typically, the concentrations of primary OC, CO, and NO (not shown) tracked one another well across the day and peaked in the early morning (between 5:00 and 11:00 a.m.; because of rush hour traffic) and late at night (between 8:00 p.m. and 2:00 a.m.; most likely because of increased stability and low mixing heights). The average primary OC concentrations were higher during G2P2 (4.62 μgC/m^3 and 4.32 μgC/m^3 for January and February, respectively) than during G1P1 (2.64 μgC/m^3 and 3.06 μgC/m^3 for July and August, respectively), suggesting that primary combustion sources of OC were dominant in the wintertime. Periods of high wintertime SOA concentrations (as high as 9–12 μgC/m^3) typically occurred in the

Table 3. Estimates of the characteristic primary OC/EC ratios (*a* = slope), noncombustion primary OC (*b* = intercept), and coefficient of determination (R^2) for each month of CHAPS calculated by regressing OC on EC (Deming regression) considering only outdoor data dominated by primary emissions.

Variable	From	To	*a*	*b* (μgC/m^3)	R^2	Primary OC (μgC/m^3)	SOA (μgC/m^3)	SOA (%)
G1P1	07/06/05	07/31/05	1.64	0.29	0.89	2.64	2.18	43
G1P1	08/01/05	08/19/05	2.27	0.00	0.87	3.06	2.16	40
G2P1	08/23/05	09/30/05	2.08	0.19	0.86	3.39	3.01	45
G2P1	10/01/05	10/14/05	2.21	0.04	0.94	4.95	3.10	42
G1P2	10/17/05	10/31/05	2.04	0.10	0.93	2.88	2.55	45
G1P2	11/01/05	12/09/05	1.86	0.00	0.85	3.18	1.88	35
G2P2	01/05/06	01/31/06	2.46	0.00	0.91	4.62	2.36	32
G2P2	02/01/06	02/16/06	2.39	0.18	0.74	4.32	3.30	40

Notes: a and *b* were used to estimate outdoor primary OC and outdoor SOA concentrations (μgC/m^3) and the percentage contribution of SOA to measured outdoor OC (SOA %).

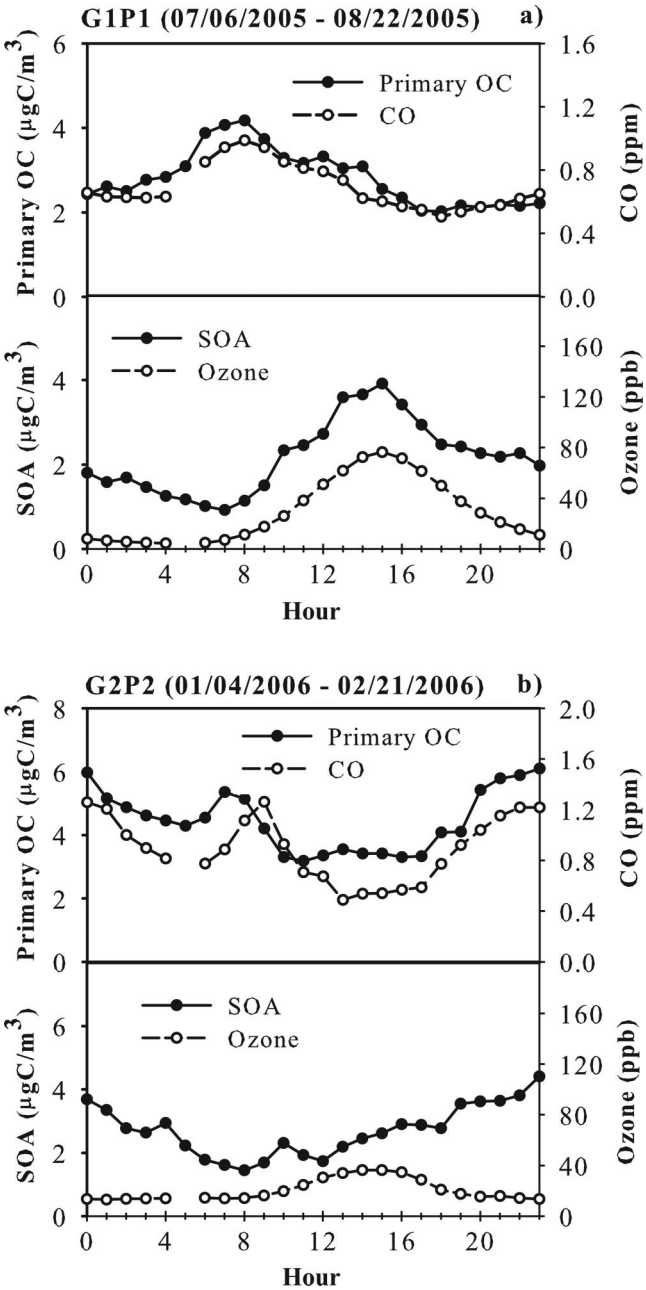

Figure 4. Time averaged diurnal patterns for estimated outdoor primary OC and SOA concentrations during (a) G1P1, typical for summertime conditions, and (b) G2P2, representative of wintertime conditions. The corresponding average outdoor CO and O_3 concentrations are also reported.

late afternoon or at night (Figure 4b). Strader et al.[34] suggested that, under suitable conditions (clear skies, low horizontal winds, and low mixing height), SOA concentrations to levels as high as 15–20 μgC/m^3 could be produced in the wintertime, mainly because of the oxidation of aromatics. Species such as toluene, xylenes, trimethylbenzenes, naphthalenes, and 1,3,5-trimethylbenzene are predicted to produce ~75% of all SOA under these conditions.

The meteorological conditions observed during G2P2 (especially in February) were extremely favorable for SOA production with afternoon temperatures that reached 30 °C on several occasions and nighttime temperatures as low as 5 °C. Under these circumstances, if highly reactive SOA precursors were accumulated within the San Gabriel Valley, significant amounts of SOA could be formed in the afternoon because of photochemical activity and at night because of a shifting of the gas-particle partitioning equilibrium toward the particulate phase because of the temperature decrease.[35] In addition, a decrease in the mixing height could also contribute to the accumulation of SOA precursors at night, accelerating the rate of SOA formation. However, it has to be recognized that these high wintertime SOA values could be, at least in part, an artifact of the EC-tracer method, because the primary OC/EC ratio is, in fact, not constant, as assumed by the method, but varies between sources and is influenced by meteorology, diurnal fluctuations in emissions, and the influence of local sources.[34] For example, an overestimation of the SOA concentrations could occur when the influence of a large primary source with no temporal regularity and a high OC/EC ratio (e.g., wood burning) were not taken into account in the determination of the primary OC/EC ratio. In fact, the OC/EC ratio for biomass burning can be >10.[36] Although CO was considered as a primary combustion tracer in the determination of the primary OC/EC ratios, the possibility that a fraction of the estimated wintertime SOA is really primary OC from biomass combustion cannot be entirely ruled out. The contribution of other processes such as nighttime chemistry and fog/cloud processing to SOA formation remains uncertain.

The fall period (most of G2P1 and G1P2 measurements) was characterized by average SOA concentrations ranging between 3.10 μgC/m^3 (October 1–14, 2005) and 1.88 μgC/m^3 (November 1 to December 9, 2005), corresponding to 42% and 35% of measured OC, respectively (Table 3). The daily primary OC and SOA concentration dynamics for G2P1 were more comparable to those observed in the summer (G1P1), with similar afternoon O_3 and SOA maxima, but slightly higher nighttime SOA increases and higher CO and primary OC morning and nighttime peaks (not shown). Likewise, G1P2 concentration dynamics were more comparable to those observed in the winter (G2P2) but with smaller nighttime SOA increases and smaller CO and primary OC morning and nighttime peaks (not shown).

AER Estimates

The average AER for each group and phase (0.25 hr^{-1} ± 0.04 (1σ), 0.28 hr^{-1} ± 0.06, 0.33 hr^{-1} ± 0.07, and 0.31 hr^{-1} ± 0.10 for G1P1, G2P1, G1P2, and G2P2, respectively) were quite constant throughout the year and similar for both G1 and G2 retirement communities. The generally low estimated AERs are consistent with the structural characteristics of the sampling sites (G1 was a recreational area and G2 a dining hall, both in the middle of the retirement homes), the low number of open windows and doors, and the presence of central air conditioners. These results are comparable to overnight AER high-resolution (3 min) measurements obtained by Sarnat et al.[28] in 17 Southern California homes using a constant sulfur hexafluoride (SF_6) source in conjunction with SF_6

monitors. By using the same methodology, the median summertime AER measured in Pennsylvania residences was ~0.30 hr^{-1} for air-conditioned homes and ~2 hr^{-1} for non-air-conditioned homes.[37] The R^2 for the regression lines used to calculate the AERs presented here (see eq 4) was always >0.9, which adds confidence to our results.

Infiltration Factor Estimates

The average F_{inf} estimates (calculated by both the I/O concentration ratio and the RM approaches) for OC, EC, $PM_{2.5}$, and PN concentrations for each group and phase of CHAPS are reported in Table 4. In general, for G1 and G2, the F_{inf} results were similar across P1 (summer and fall) and P2 (fall and winter), which is consistent with no seasonal changes in home dynamics and ventilation conditions as indicated by the rather constant AERs calculated throughout the study and discussed above. The average F_{inf} results were highest for EC (ranges across methods were 0.70–0.82, 0.67–0.74, 0.77–0.80, and 0.64–0.69 for G1P1, G2P1, G1P2, and G2P2, respectively) and OC (ranges were 0.83–0.98, 0.74–0.77, 0.82–0.87, and 0.61–0.67 for G1P1, G2P1, G1P2, and G2P2, respectively). For EC, this is likely because of the fact that EC is nonvolatile, is found mostly in the 0.1–0.4 μm range,[38,39] and, thus, is capable of infiltrating through the building envelope with great efficiency. The equally high F_{inf} estimated for OC suggests that the particle size range of this important $PM_{2.5}$ component was probably similar to that of EC and that it was mainly composed of organic compounds with relatively low vapor pressure. This is also consistent with our observations throughout the study that a substantial fraction of outdoor OC consisted of SOA, whose size distribution is generally concentrated in the lower sizes of the accumulation mode[40] (characterized by a nighttime F_{inf} of ~0.7[26,28]), and is typically composed of highly polar organics.[41–43] It should be noted that lower F_{inf} (OC) values may have been obtained if our indoor sampling sites had been located in environments characterized by a higher surface to volume ratio ([S/V] e.g., a fully furnished apartment as opposed to a recreational area or a dining room), because the depositional loss rate (k) increases with increasing S/V of the studied indoor location.[44] During CHAPS, for particulate species that are known to be composed of both semivolatile and volatile compounds (e.g., OC), this might translate in an overestimation of the corresponding F_{inf}. A somewhat lower average F_{inf} was obtained for $PM_{2.5}$ (ranges across methods were 0.52–0.74, 0.45–0.60, 0.52–0.62, and 0.38–0.45 for G1P1, G2P1, G1P2, and G2P2, respectively) reflecting the possible effects of volatile and semivolatile species on F_{inf}. For example, particulate compounds, such as ammonium nitrate, which account for 35–49% of the outdoor $PM_{2.5}$ mass in the Los Angeles basin,[45–48] volatilize upon building entry and contribute to lower the average F_{inf} of $PM_{2.5}$. The average F_{inf} for ammonium nitrate reported by Sarnat et al.[28] in a recent study conducted in Southern California homes was 0.18.

Finally, the F_{inf} estimates for PN concentration were 0.59–0.69, 0.46–0.55, 0.77–0.80, and 0.54–0.63 during G1P1, G2P1, G1P2, and G2P2, respectively. We hypothesize that the somewhat lower F_{inf} (PN) calculated at site B (G2) was caused, at least in part, by a higher fraction of sub-100-nm particles in the sampled aerosol because of the close proximity (<1 mi) of this site to a major highway. Lower F_{inf} values for PN at site B (G2) are consistent with the lower penetration of sub-100-nm particles indoors because of diffusional losses,[26,28] as well as losses because of evaporation of volatile species associated with this size range.[49]

With the exception of F_{inf} for PN concentration, the average F_{inf} results for OC, EC, and $PM_{2.5}$ estimated during CHAPS are in good agreement with those obtained in several previous studies conducted in other parts of the United States.[11,27,28] Because F_{inf} varies with both aerosol composition (e.g., changes in the outdoor concentration of labile species, such as ammonium nitrate) and the AER, diurnal variations of these variables are likely to affect particle infiltration. However, the standard deviation of the daily averaged F_{inf} estimates for OC, EC, PN, and $PM_{2.5}$ (also reported in Table 4) were small within each group and phase of the study, probably because of the structural characteristic of the G1 and G2 sampling sites (e.g., presence of central air conditioners and low number of open windows and doors). Most importantly, our findings indicate that $PM_{2.5}$ and its carbonaceous components (e.g. OC and EC, both of which compose a substantial portion of the $PM_{2.5}$ mass[11]) are characterized by different F_{inf} values. This implies that the composition of indoor and outdoor particles is different and that ambient $PM_{2.5}$ concentrations may not adequately represent personal exposures to outdoor-infiltrated $PM_{2.5}$ in indoor environments.[11,14,27,31,32]

Table 4. Average infiltration factors estimates (F_{inf}) for OC, EC, $PM_{2.5}$, and PN calculated through an analysis of the I/O concentration ratios and by using a recursive model (RM) technique.

		I/O (all data)[a]		I/O (nighttime)[b]		
Variable	Species	F	SD	F_{inf}	SD	RM F_{inf}
G1P1	OC	0.86	0.05	0.83	0.09	0.98
	EC	0.73	0.07	0.70	0.12	0.82
	PN	0.69	0.03	0.59	0.05	0.66
	$PM_{2.5}$	0.71	0.10	0.74	0.13	0.52
G2P1	OC	0.77	0.09	0.76	0.08	0.74
	EC	0.71	0.05	0.67	0.08	0.74
	PN	0.55	0.07	0.51	0.10	0.46
	$PM_{2.5}$	0.60	0.05	0.54	0.08	0.45
G1P2	OC	0.82	0.07	0.82	0.10	0.87
	EC	0.77	0.06	0.80	0.07	0.79
	PN	0.80	0.06	0.77	0.08	0.78
	$PM_{2.5}$	0.59	0.07	0.62	0.11	0.52
G2P2	OC	0.64	0.10	0.67	0.10	0.61
	EC	0.64	0.07	0.69	0.10	0.64
	PN	0.63	0.06	0.54	0.07	0.55
	$PM_{2.5}$	0.45	0.06	0.45	0.08	0.38

Notes: SD = standard deviation; [a]All I/O ratios ≤ 1 were used; [b]Only nighttime (12:00–6:00 a.m.) I/O ratios ≤ 1 were used.

Indoor and Outdoor Contributions to Measured Indoor Species Concentrations

By multiplying the measured outdoor 1-hr OC, EC, $PM_{2.5}$, and PN concentrations (C_{out}) by the corresponding average F_{inf} estimates reported in Table 4, we determined the indoor contribution of outdoor origin for each particulate species (C_{og}) and for each group and phase of CHAPS. The

resulting indoor contributions of indoor origin (C_{ig}) were then estimated by subtracting C_{og} from C_{in} on a sample-by-sample basis. Figure 5 shows the calculated C_{ig} concentrations for OC (Figure 5a), EC (Figure 5b), $PM_{2.5}$ (Figure 5c), and PN concentrations (Figure 5d) expressed as a percentage of the corresponding measured indoor concentrations (C_{in}) and averaged throughout G1P1, G2P1, G1P2, and G2P2. Columns refers to C_{ig} values obtained by using F_{inf} estimates from the I/O ratio method considering all of the hourly I/O data ≤1 (black), the I/O method accounting only for nighttime ratios ≤1 (gray), and the RM approach (darker gray).

Our results indicate that, on average, 16–26% (1.06–1.63), 18–20% (1.69–1.80), 20–23% (1.17–1.33), and 13–17% (1.03–1.23) of measured indoor OC was emitted or formed indoors during G1P1, G2P1, G1P2, and G2P2, respectively (the corresponding ranges of average indoor-generated OC concentrations in micrograms of carbon per cubic meter are reported in parentheses; Figure 5a). These calculations suggest that, although the G2 indoor site was characterized by higher indoor morning OC peaks because of cooking, the overall contribution of indoor sources to measured indoor OC was actually higher at the G1 site. These results are lower than those obtained by Polidori et al.[11] during the Relationship of Indoor, Outdoor and Personal Air (RIOPA) Study, where the average C_{ig} for OC varied between 40% and 70%. The lower C_{ig} OC estimates obtained here are consistent with the prevailing use of central air conditioning at both G1 and G2 indoor sites and may also be because of differences in home dynamics between the RIOPA and the CHAPS sampling locations (personal residences and common areas for retirees, respectively) and exposure groups. The CHAPS subjects consisted of retirees with compromised health, whose indoor activity levels are likely to be much lower than those of the RIOPA group.

The average percentages of measured indoor EC that were generated indoors were 17–25% (0.37–0.48), 11–16% (0.16–0.22), 21–23% (0.27–0.30), and 20–22% (0.23–0.27) for G1P1, G2P1, G1P2, and G2P2, respectively (ranges of average indoor-generated EC in micrograms of carbon per cubic meter in parentheses; Figure 5b). These values are quite close to the detection limit for EC for semicontinuous carbon measurements, typically around 0.15–0.35 $\mu gC/m^3$ (as determined by Lim et al.[21]), and suggest that indoor sources of EC were not an important contributor to measured indoor EC during CHAPS. These outcomes are consistent with I/O EC ratios close to or slightly lower than unity obtained in several previous studies conducted both in California[50,51] and around the world.[52,53]

The mass balance model results also showed that, on average, 6–21% (1.85–5.33), 24–38% (5.02–7.47), 42–51% (8.26–10.31), and 21–30% (2.82–4.03) of measured indoor $PM_{2.5}$ was emitted or formed indoors during G1P1, G2P1, G1P2, and G2P2, respectively (the corresponding ranges of average indoor-generated $PM_{2.5}$ concentrations in micrograms per cubic meter are reported in parentheses; Figure 5c). These outcomes are somewhat difficult to interpret and suggest that the seasonal emission/formation of indoor $PM_{2.5}$ from indoor sources was highly variable. It is important to recognize that the $PM_{2.5}$ concentrations measured indoors during G2P2 were unusually low compared with the corresponding outdoor $PM_{2.5}$ concentrations and with the G2P1 $PM_{2.5}$ data. Whether or not this was because of a malfunctioning of the indoor BAMs or seasonal changes in home dynamics and ventilation conditions between G2P1 and G2P2 remains unclear.

The average percentages of measured indoor PN concentration that was emitted/formed indoors were 14–22% (2235–3169), 17–26% (4618–5493), 17–19% (3258–3527), and 13–21% (6841–8010) for G1P1, G2P1, G1P2, and

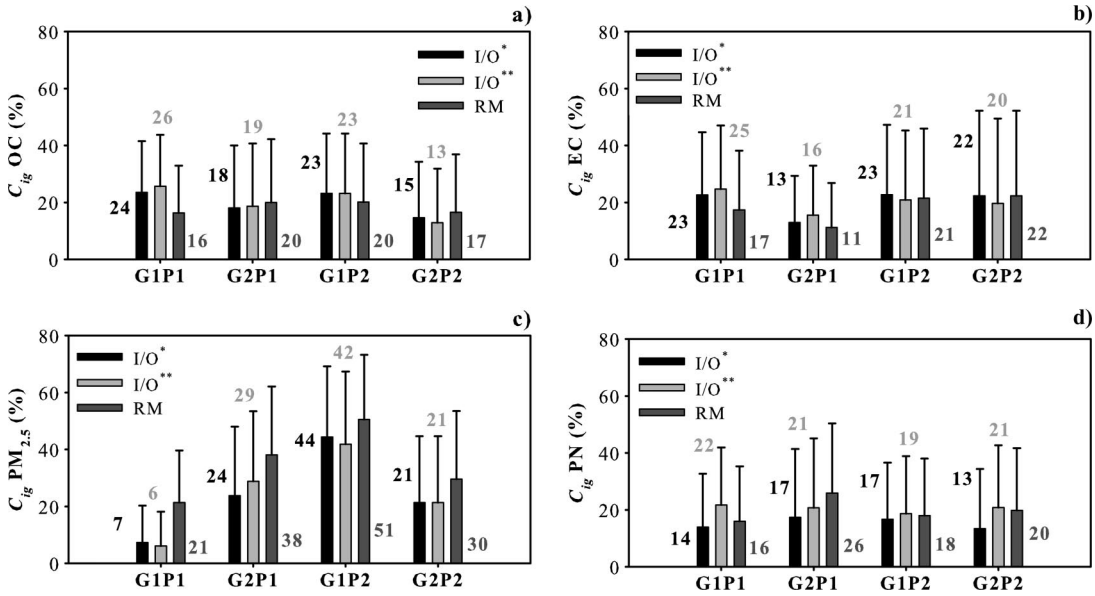

Figure 5. Calculated indoor concentrations of indoor origin (C_{ig}) for OC (a), EC (b), $PM_{2.5}$ (c), and PN (d) expressed as a percentage of the corresponding measured indoor concentrations (C_{in}) and averaged throughout G1P1, G2P1, G1P2, and G2P2. Columns refer to C_{ig} values obtained by using F_{inf} estimates from the I/O ratio method considering all hourly ratios ≤1 (I/O∗), the I/O method accounting only for nighttime ratios ≤1 (I/O∗*), and the recursive model (RM) technique. Error bars represent +1σ (1 standard deviation) of all hourly C_{ig} estimates within each group (G) and phase (P).

G2P2, respectively (ranges of average indoor-generated PN per cubic centimeter reported in parentheses; Figure 5d). These results suggest that the PN concentration of indoor origin increased from summer to fall (at the G1 site) and from fall to winter (at the G2 site). The seasonal increase in C_{ig} for PN concentration was probably because of the use of indoor fan heaters during the wintertime. A recent study conducted by He et al.[54] in 15 Australian houses demonstrated that the use of a fan heater elevates the indoor submicrometer PN concentration levels by more than five times over a 48-hr period but does not affect significantly the levels of indoor $PM_{2.5}$ mass. Other indoor activities, such as cooking, might have increased the indoor levels of PN concentrations by a substantial amount.

By using the same mass balance approach, we also estimated the average amount of outdoor SOA and outdoor primary OC that penetrated inside G1 and G2 indoor sites (C_{og} SOA and C_{og} primary OC, respectively) during each phase of CHAPS. For these calculations, we assumed that F_{inf} for SOA was equal to 0.86, the average summertime value for OC during G1P1 (see Table 4). As illustrated in Figure 6, the average percentage contribution of indoor SOA of outdoor origin to measured indoor OC, C_{og} SOA (%), was rather constant throughout the study within each group, varying from 33% (1.89) to 35% (1.60) for G1P1 and G1P2, respectively, and from 46% (2.60) to 45% (2.34) for G2P1 and G2P2, respectively (the corresponding average concentrations in micrograms of carbon per cubic meter are reported in parentheses). When varying F_{inf} for SOA of ± 0.1, the corresponding average C_{og} SOA (%) for all of the groups and phases fluctuated by $\leq 5\%$. Figure 6 also shows that, on average, 44%, 36%, 42%, and 40% of measured indoor OC was composed of outdoor-generated primary OC during G1P1, G2P1, G1P2, and G2P2, respectively. These C_{og} primary OC (%) values were determined from the mass balance equation as follows:

$$C_{og}\ primary\ OC\ (\%) = 100 - C_{ig}\mathrm{OC}\ (\%) - C_{og}\mathrm{SOA}(\%) \tag{12}$$

To the best of our knowledge, these results are among the first to quantify the contributions of outdoor-generated SOA and primary OC to indoor OC and to demonstrate

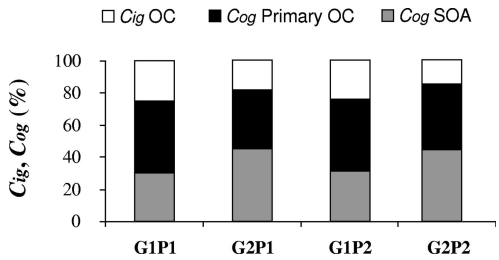

Figure 6. Estimated indoor primary OC and indoor SOA concentrations of outdoor origin ("C_{og} Primary OC" and "C_{og} SOA," respectively) expressed as a percentage of the corresponding measured indoor concentrations (C_{in}) and averaged throughout G1P1, G2P1, G1P2, and G2P2. Estimated average indoor OC concentrations of indoor origin (C_{ig} OC) are also reported.

their importance in indoor environments. These outcomes will be used by CHAPS investigators to clarify the links between exposure to $PM_{2.5}$ of indoor and outdoor origin and their effects on cardiovascular outcomes. In the Los Angeles basin, outdoor primary OC particles are mainly emitted from motor vehicle exhausts, are mostly found in the ultrafine mode, are composed of well-known carcinogenic components/species, such as diesel particles and polycyclic aromatic hydrocarbons, and are more likely to deposit in the lower airways than coarse particles.[55–57] On the other hand, a growing body of evidence is suggesting that exposure to SOA (mostly composed of highly polar organic compounds) is linked to respiratory inflammation through the generation of reactive oxygen species.[57,58]

The single compartment mass balance model presented in this work allows for a straightforward estimation of the C_{ig} and C_{og} concentrations for a given F_{inf} value. Although the uncertainties inherent in the approach proposed here must be acknowledged (e.g., F_{inf} was considered to be constant within each group and phase of the study), the estimated C_{ig} and C_{og} values for OC, EC, $PM_{2.5}$, PN, SOA, and primary OC seem reasonable when compared with the relatively limited data in the available literature.

CONCLUSIONS

This study was conducted in the Los Angeles basin at two retirement communities. Measured indoor and outdoor concentrations of $PM_{2.5}$, OC, EC, PN, O_3, CO, and NO_X were generally comparable, although at G2, a substantial peak in indoor OC, PN, and $PM_{2.5}$ (probably from cooking) was typically observed between 6:00 and 9:00 a.m. The study average percentage contribution of outdoor SOA to outdoor particulate OC (representative for the San Gabriel Valley) was 40% and varied between 40% and 45% in the summer (during G1P1) and 32% and 40% in the winter (during G2P2). Quantifying the SOA contribution to measured OC is important for the following reasons: (1) to test evolving predictive SOA models; (2) to link the organic PM concentration to its emissions and precursors; and (3) to develop effective control strategies for PM. The low AERs (0.25–0.33 hr^{-1}) calculated for G1 and G2 are consistent with the structural characteristics of the sampling sites, the low number of open windows and doors, and the presence of central air conditioners. F_{inf} values were determined for OC, EC, $PM_{2.5}$, and PN by using different methods, including the RM developed by Allen et al.[29] Here, the RM approach, validated recently for estimating F_{inf} for $PM_{2.5}$ from light scattering data,[30] has been applied to OC, EC, and PN data for the first time. F_{inf} estimates were highest for EC (a nonvolatile species mostly found in the 0.1–0.4 μm range[38,39]) and also for OC (probably because of the substantial percentage mass contribution from SOA throughout CHAPS). Lower F_{inf} values were obtained for $PM_{2.5}$ and PN, because these compounds are composed of both volatile and nonvolatile inorganic and organic components. Estimated F_{inf} and measured particle concentrations were then used in a single compartment mass balance model to assess the mean contributions of indoor and outdoor sources to measured indoor OC, EC, $PM_{2.5}$, and PN. We found that

13–17% (G2P2) to 16–26% (G1P1) of measured indoor OC was emitted or formed indoors. Although the G2 indoor site was characterized by higher indoor morning OC peaks because of cooking, the overall contribution of indoor sources to measured indoor OC was higher at the G1 site. These results are consistent with low indoor activity levels at both retirement communities and with the prevailing use of central air conditioning. Our modeling results also showed that the measured indoor $PM_{2.5}$ emitted or formed indoors was highly variable (from 6–21% at G1P1 to 42–51% at G1P2). The average percentage contribution of indoor SOA of outdoor origin to measured indoor OC varied from ~35% (at site 1) to ~45% (at site 2). Also, outdoor-generated primary OC composed, on average, 36–44% of measured indoor OC during G2P1 and G1P1, respectively. These results are among the first to quantify the contributions of outdoor-generated SOA and primary OC to indoor OC and to demonstrate their importance in indoor environments. The outcomes presented here will be used by CHAPS investigators to determine the relationship between cardiovascular outcomes and hourly retirement community exposures by each resident to $PM_{2.5}$ of indoor and outdoor origin.

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