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The contribution of traffic to indoor concentrations of polycyclic aromatic hydrocarbons[†]

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A photoelectric aerosol sensor (PAS) was used to measure real-time indoor concentrations of polycyclic aromatic hydrocarbons (PAHs) at three residences. Semi-quantitative measurements of total indoor particle-bound PAH and temperature were collected continuously every minute for approximately 2 weeks at each location. The purpose of this study was to examine the effect of traffic on indoor concentrations of PAHs. This was accomplished by collecting indoor measurements at an urban, semi-urban, and suburban residential location with varying levels of, and proximity to, traffic. Since the homes were occupied, the effects of cooking, the dominant indoor source, were also examined among the three nonsmoking households. The results indicate that traffic was the main outdoor source of PAH concentrations measured indoors for all locations. In fact, a significant (p<0.001) traffic-related trend in weekday PAH concentration was detected with a geometric mean concentration at the urban location (31 ng/m^3) nearly two times that at the semi-urban location (19 ng/m^3) and over three times larger than the suburban location (8.0 ng/m^3), once adjusted for indoor sources. Hourly average concentration profiles also revealed weekday rush hour peaks of PAHs at all locations. No pronounced peaks and significantly lower concentrations (10, 10, and 4.9 ng/m^3) were seen during the weekends for all locations i.e., the urban, semi-urban and suburban locations, respectively. Indoor sources including frying/sautéing, broiling, and candle-burning were characterized by peak concentration, duration of PAH elevation, and potential dose. This analysis suggests that cooking, and especially frying/sautéing, may be an important source of indoor PAH concentrations.

Keywords: *cooking, exposure, indoor air, polycyclic aromatic hydrocarbons (PAH), potential dose, traffic.*

Introduction

With a rising concern over human exposure to airborne particles, there is a growing interest in the chemical constituents of particulate matter, including high molecular weight polycyclic aromatic hydrocarbons (PAHs) (US EPA, 1996). High molecular weight PAHs are primarily sorbed to the particulate respirable fraction (e.g., <2.5 μm) and many have been identified as mutagens (Lewtas et al., 1992) and carcinogens (Chiddihy et al., 1984; Rannug and Sundvall, 1985; Cassarett and Doull, 1991; DHHS, 1995). Because of this toxicity and the ubiquitous occurrence of PAHs throughout the environment, exposure to PAHs is of significant public health concern. Environmental exposure occurs primarily through dietary ingestion and secondarily through inhalation (Buckley and Lioy, 1992; Buckley et al., 1995). More recent data suggest that nondietary ingestion of contaminated house dust may also be an important route of exposure especially for children due to their increased contact with house dust and increased hand-to-mouth activity (Wilson et al., 1994; Roberts and Dickey, 1995). The indoor air-inhalation pathway is of particular concern with respect to respiratory effects including lung cancer. PAHs from mobile sources have been shown to be more mutagenic than PAHs from other sources such as wood stoves (Lewtas et al., 1992; Cupitt et al., 1994).

PAHs are products of incomplete combustion and are often generated through open burning, incineration, industrial power generation, and vehicle emissions (DHHS, 1995). While these particular sources are found outdoors, indoor sources must also be considered. In fact, it has been well-documented that indoor PAH concentrations are influenced by both indoor and outdoor sources. Activities such as cooking, residential heating, smoking cigarettes, and burning candles are known to generate significant levels of PAHs (Waldman et al., 1990; Chuang et al., 1991; Nabinger et al., 1995; Ott and Klepeis, 1995; Ott et al., 1994; Wallace et al., 1997). As people spend approximately

1. Abbreviations: PAH, polycyclic aromatic hydrocarbons; PAS, photoelectric aerosol sensor.

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89% of their time indoors (National Research Council, 1981), both indoor and outdoor sources of PAH exposure must be characterized in order to fully evaluate exposures and the means for their control and mitigation.

Indoor sources of PAHs are often transient in nature and as a result, they are difficult to characterize using traditional integrating sampling methods. Instead, a monitoring strategy is needed to characterize the emission variability with time. In this study, a photoelectric aerosol sensor (PAS) was used to provide real-time measurements. In particular, the PAS monitor generates semi-quantitative measurements of total PAH bound to the surface of particles less than 2.5 μm in diameter. Therefore, the instrument is particularly well suited for particles generated from mobile sources as these particles typically range in size from 0.01 to 0.1 μm (US EPA, 1993) and are coated with an outer layer of PAHs (Steiner et al., 1992).

The purpose of this study was to evaluate the impact of traffic on indoor concentrations of particle-bound PAHs. This goal was met by examining temporal and spatial patterns of real-time indoor PAH concentrations at three residences with varying proximity to and density of traffic sources, as defined by local traffic counts. In order to isolate the effect of traffic, it was necessary to identify and adjust for the contribution of cooking to indoor PAH concentrations as the dominant indoor source in the nonsmoking residences studied.

Methods

Airborne PAH concentrations were measured semi-quantitatively at an urban, semi-urban, and suburban residence (one location at a time) during the summer of 1997. These residences were selected based on their proximity to roadways of varying traffic density. Traffic counts for those roadways in close proximity to each residence were gathered from local and state agencies to validate residential classification into urban, semi-urban, and suburban categories. In addition, measurements of wind speed and direction were obtained from nearby Massachusetts Department of Environmental Protection monitoring stations to verify that the prevailing winds did not confound the effect of traffic on indoor air. PAS measurements were taken continuously at the homes to generate daily indoor air concentration profiles. Source generation journals were kept to identify periods of indoor combustion activities.

Site Descriptions

Urban Location Sampling was conducted at a Massachusetts Institute of Technology (MIT) dormitory situated outside of Kenmore Square in Boston, Massachusetts. This location was chosen for its close proximity to many high traffic roads. The back of the dormitory is located just 30 ft south of Storrow Drive, a six-lane highway with average daily traffic counts of 81,770 and 68,660 for weekdays and weekends, respectively, as recorded in May of 1997 by the Massachusetts Highway Department (Massachusetts Highway Department, 1998). Also nearby (0.2 miles) is the Massachusetts Turnpike, an eight-lane interstate highway, which averaged 104,862 vehicles per day in this area during 1997 (Massachusetts Turnpike Authority, 1998). Other neighboring sources of traffic include the six-lane Beacon Street (0.2 miles) and Commonwealth Avenue (0.1 miles). Beacon Street and Commonwealth Avenue were reported to have 8,599 and 29,358 vehicles, respectively, traveling through Kenmore Square (0.2 miles to the southeast) during an 11-h period (7 AM to 6 PM) in 1988 (Boston Transportation Department, 1998). As this section of Beacon Street services only one-way traffic, while the stretch closest to the sampling location supports traffic in two directions, we have assumed that the true traffic count for this road is double the value reported above. Therefore, we have estimated that the daily traffic counts for the four major thoroughfares near this residence were approximately 233,000 and 220,000 cars and trucks each weekday and weekend, respectively. No known industrial sources were within 1/2 mile of the residence.

The dormitory is comprised of 11 bedrooms, five bathrooms, a poolroom, storage area, and laundry room for a total volume of approximately 1540 m^3. These rooms are situated on the perimeter of a large spiral staircase, resulting in a hollow atrium from the first floor to the fourth. A back stairwell connects the basement level to the rest of the house. All monitoring occurred on the third floor within the central atrium on a shelf at approximately 2 m above the floor and about 30 cm from the wall.

Samples were collected for 14 days from July 17 to 24, 1997 and from August 10 to 15, 1997. During these periods, approximately 25 residents occupied the house. There were no air conditioners in this residence so ventilation occurred by open windows and fans. Most bedroom doors were left open. These factors are likely to have led to a high rate of air exchange at this location.

Semi-urban Location This three-bedroom apartment is located approximately 1 mile to the west of the urban location in Brookline, MA. While this residential area is slightly removed from the high traffic roads, the apartment is still in close proximity to the Massachusetts Turnpike (0.4 miles), Commonwealth Avenue (0.3 miles), Storrow Drive (0.4 miles), and Beacon Street (0.3 miles). In addition, the residence is 0.4 miles from Harvard Street, a thoroughfare similar in size to Beacon Street and Commonwealth Avenue.

The average daily traffic count for this section of the Massachusetts Turnpike was reported to be 79,511 during

1997 (Massachusetts Turnpike Authority, 1998). All other roads were assigned the same traffic densities as those for the urban location. Since no traffic counts were available for Harvard Street, we have assumed that the count for this road is approximately equal to the average of those counts reported for Beacon and Commonwealth. In total, the estimated average traffic counts on the five major roads near this residence were 231,000 and 218,000 cars and trucks per day for weekdays and weekends, respectively. Although these traffic densities were similar to the urban residence, the two sites differed in their proximity to the major roadways. The semi-urban location was 0.3–0.4 miles distant from the roadways, while the urban location ranged from 30 ft to 0.2 miles from roadways. No known industrial sources were within 1/2 mile from the residence.

This residence is located on the fourth floor of a 412-unit apartment complex and faces an outdoor parking lot. The apartment is 240 m^3 in volume and consists of three bedrooms, one and a half baths, a living room, dining area, and kitchen. The monitor was located on the living room floor roughly 30 cm from the wall.

This residence was occupied by four adults during most of the sampling period. Sampling occurred during June 13–18, July 3–14, and August 28–29, 1997. During these 20 sampling days, residents controlled the indoor environment with zero to five air-conditioning units or open windows. The bedroom doors were left open approximately 70% of the time.

Suburban Location The suburban sampling was conducted in a private home in Wellesley, Massachusetts, approximately 15 miles west of downtown Boston. The home borders natural conservation land and the surrounding area is a quiet residential neighborhood. The nearest road of high traffic density is the four-lane Route 9 located 0.8 miles to the south of the house. Traffic counts for Route 9 indicate that an average of 50,700 vehicles traveled this section of roadway daily during 1996 (Massachusetts Highway Department, 1998). Weston Road, with daily traffic counts ranging from 2,891 to 4,791, as measured in January of 1997 (Town of Wellesley, 1998), is located 0.6 miles to the west. A similarly traveled road, Cliff Road, is also located 0.8 miles to the east. Other traffic sources, including the Massachusetts Turnpike (107,225 average vehicles per day, as counted in 1997) and Route 16 (23,600 average vehicles per day, as counted in 1997) were located over 1.2 miles from the residence (Massachusetts Turnpike Authority, 1998; Massachusetts Highway Department, 1998). In total, we have estimated that this site was impacted by an approximate traffic count of 60,000 vehicles within a 1-mile radius. The density of traffic at this location was roughly 25% of the urban locations. No known industrial sources were in the vicinity (at least 1 mile).

The home is a split-level house with approximately 600 m^3 in volume. Including the basement, the house has four levels. There are four bedrooms, two and a half baths, a living room, dining room, eat-in kitchen, sunroom, library, laundry room, basement playroom, and work room. There is also an attic and an attached two-car garage; however, since these two areas are typically sealed off from the remainder of the house, they were not included in the estimation of house volume. The PAS monitor was placed at floor level in the doorway between the sunroom and dining rooms on the ground floor.

Sampling occurred on June 18–22 and July 29–August 10, 1997 for a total of 18 days. Two residents occupied the house during most sampling days and open windows or air conditioners (four units total) were used to ventilate the house. All doors between the top two stories were kept open and the air exchange rate was expected to be similar to that of the semi-urban location.

PAH Monitor

A Gossen PAS model 1002i PAH Ambient Analyzer™ (Ecochem Technologies, West Hills, CA) was used to detect total particle-bound PAHs. The monitor's principle of operation is based on photoelectric ionization. Air is drawn through an electrostatic filter at a rate of 4 l/min to remove all charged particles. The resulting neutral air stream is then passed through an ionization chamber and exposed to ultra violet radiation at 6.7 eV. Since most particle-bound PAHs are ionized at this energy, free electrons are produced. An applied electric field and simple diffusion remove the liberated electrons and all neutral particles that are not ionized at 6.7 eV. Charged particles coated in PAHs are then collected on a filter inside an aerosol electrometer and a current is generated proportional to the quantity of all ionized PAHs (Niessner et al., 1989). This signal represents total PAH, i.e., it does not indicate individual PAHs.

Monitor Strengths The PAS monitor is most effective for detecting PAH species with four or more rings, since they have lower photoelectric thresholds than the smaller PAHs. Efficiency is also maximized for those particles less than 1–2 μm in diameter, as electron recapture is negligible (Wilson et al., 1994). Since particles generated from mobile sources were shown to have a mass median diameter peak of 0.02 μm (US EPA, 1993) and are enriched in high molecular weight PAHs (Steiner et al., 1992), this monitor is particularly well-suited for the detection of traffic-related PAHs.

Potentially, the most important characteristic of the monitor is its ability to provide real-time measurements effectively for combustion sources. This allows for temporal resolution and source identification that cannot be achieved by traditional methods. In addition, the limits of detection (LODs) for these monitors typically range between 3 and 10 ng/m^3.

Monitor Limitations The PAS monitors use ionization to detect PAHs; there is an inherent difficulty in calibration due to differing responses from different species of PAHs as well as different particle characteristics (Niessner, 1986; Niessner and Wilbring, 1989). In fact, sources with varying compositions and particle properties may require entirely different calibrations. However, as appropriate standards are not yet available, it currently is not possible to externally calibrate the instrument. Due to this limitation, the PAS instrument is considered to provide semi-quantitative measurements. Furthermore, while PAHs represent a chemical class made up of hundreds of compounds, the PAS provides a measure of the total particle-bound PAH concentration without distinguishing between specific compounds.

An additional source of variability relates to the 6.7 eV ionization energy and the specificity of the PAS monitor. Specificity is an issue for the PAS 1002i because approximately 100 compounds other than PAHs have ionization potentials less than 6.7 eV. While most or possibly all of these compounds are rare or nonexistent in indoor air, the possibility still remains that some aerosols could form a positive interference. For example, Wallace et al. (1997) reports that the 2000 series monitor with a less energetic light source (5.6 eV) often did not respond to cooking events while a side-by-side 1002i monitor did respond. This difference in monitor response might be attributable to a lack of specificity by the 1002i monitor or alternatively a lack of sensitivity by the 2000 monitor. The monitors were, however, in good agreement for traffic-related PAHs.

Supporting Evidence for Current Use Several previous authors have used this technology to measure PAHs generated from various combustion products (Niessner et al., 1989; Leonardi et al., 1990; McDow et al., 1990; Wilson et al., 1994; DHHS, 1995; Ott and Klepeis, 1995; Buckley and Ott, 1996; Wallace et al., 1997). In fact, for typical combustion sources, the detector signal has been shown to be highly correlated (r^2=0.998) with the amount of PAH absorbed onto the particle surface (Niessner, 1986; Niessner et al., 1989). Furthermore, when comparing the PAS detector response to traditional methods, moderate to high correlations were found for an oil stove (r^2=0.99, p=0.05), automobile exhaust (r^2=0.84, p=0.05) (McDow et al., 1990), indoor air microenvironments (r^2=0.985, n=12) (Wilson et al., 1994), and benzo[a]pyrene in cigarette smoke (r^2=0.88, n=20) (Niessner and Walendzik, 1989).

Previous work also indicates that the conversion factor for the PAS monitor is relatively consistent for a given combustion source. In fact, based on the former studies, Wilson et al. (1994) inferred a universal conversion factor of 1–3 μg m^{-3} pA^{-1}. This conversion factor is reported to be independent of source type within a factor of 2 (Wilson et al., 1994). For the purposes of this study, a general conversion factor of 1 μg m^{-3} pA^{-1} has been adopted. Although the PAS is considered semi-quantitative due to limitations in calibration, instrument response within and between similar monitors has been shown to be consistent for a given source type.

Data Collection

Measurements of total particle-bound PAH and temperature were collected every minute to generate daily profiles of indoor PAH levels at an urban, semi-urban, and suburban location. Each residence was sampled for two or three nonconsecutive periods so as to minimize possible bias due to temporal changes. The total sampling time was approximately 2 weeks per location.

All monitoring occurred in central rooms which were both near the kitchen (if applicable) and frequently used by the occupants. As previous findings have indicated that fairly good mixing occurs in most homes (Chuang et al., 1991), it can be expected that these rooms were representative of the whole living unit.

Prior to data collection, the PAS monitor was zeroed and the mercury lamp was allowed to warm for 30 min. While no site calibration was possible, the equipment was factory-calibrated.

The DataBear™ CO Monitor (Langan Products, San Francisco, CA) was used as a temperature sensor and data-logger for the PAS monitor. The sophisticated data-logging capabilities of this monitor allowed for simultaneous storage of PAH and temperature at 1-min intervals. The temperature sensor showed good agreement with both a standard mercury thermometer and a digital thermometer.

Measurements of wind direction and wind speed were obtained from MADEP monitoring stations located in Lynn and Waltham, Massachusetts. Lynn is a coastal community just north of Boston that was used to represent the prevailing winds at the urban and semi-urban location. Waltham is located approximately 15 miles west of Boston and was used to represent the meteorological conditions at the suburban location. All data were recorded in hourly measurements continuously for the months of June, July, and August of 1997.

Daily activity journals were used to record cooking and combustion events in the semi-urban and suburban locations. The participants were asked to record the time, duration, and type of event. At the urban location, a residential survey was conducted in place of a journal, since there were no cooking facilities and because full participation by the 25 residents could not be expected. Furthermore, the effect of any indoor source would be mitigated by the residence's large volume and high air-exchange rate. This was tested by conducting three source simulations to verify that indoor sources did not affect indoor air concentrations at the urban residence.

Statistical Analysis

Univariate statistical analyses were conducted using Excel version 5.0A for the Power Macintosh. All values of zero reported by the monitor were replaced by 1 ng/m^3 so that the data could be logarithmically transformed. Identical analyses were conducted by replacing the zeros with 1/2 and 1/(2)$^{1/2}$ of the upper and lower limit LODs (3 and 7.5). Assigning these different values to the monitor's reported zero concentrations had no effect on this study's conclusions.

Results and discussion

Figure 1 illustrates the trends in weekday hourly PAH averages for all locations following adjustment for indoor sources, as discussed in the indoor source section presented below. These results clearly indicate that traffic is the dominant outdoor source. Most pronounced is the effect of the morning rush hour. Morning peaks are seen at all locations and decrease in height with increasing distance from traffic sources. At the urban location, this peak begins at 5:30 AM and reaches an apex of 65 ng/m^3 at 8:30 AM. Not surprisingly, the semi-urban location peaks simultaneously, although the height is smaller by 15 ng/m^3. The morning peak of 20 ng/m^3 recorded at the suburban location is less dramatic and appears later in the morning (i.e., 9:30 AM). All locations exhibit a lull around noon. The urban and semi-urban locations exhibit a slow increase in PAH levels shortly after noon that peak at approximately 5 PM. A second peak at 10 PM was also seen at the urban location. Evidence of an evening rush hour at the suburban location is far weaker. There is a consistent elevation between 7 PM and 8 PM, but its value is only 5 ng/m^3 above background. In general, the lowest PAH concentrations of the day occur between 1:30 AM and 4:30 AM.

Figure 2 shows the weekend hourly PAH averages following adjustment for indoor sources. July 4th was included in this category as it is a national holiday and no commuter traffic was expected. This figure illustrates no PAH trends in time. This lack of rush hour peaks during the weekend is even further evidence that traffic is indeed the predominant outdoor source of indoor PAH exposure. It is also interesting to notice that the disparity between locations is much smaller during the weekends, unlike the weekdays, where clear differences are seen between locations. This suggests that PAHs from traffic sources are a dominant contributor to indoor PAH levels, particularly in urban areas.

Qualitative evaluation of these hourly average concentrations by location suggests that the urban location had the highest levels of PAHs. While these levels are similar to those at the semi-urban location, the suburban location clearly has much lower PAH concentrations. Table 1 presents PAH indoor air descriptive statistics for each location. These results have been adjusted for cooking

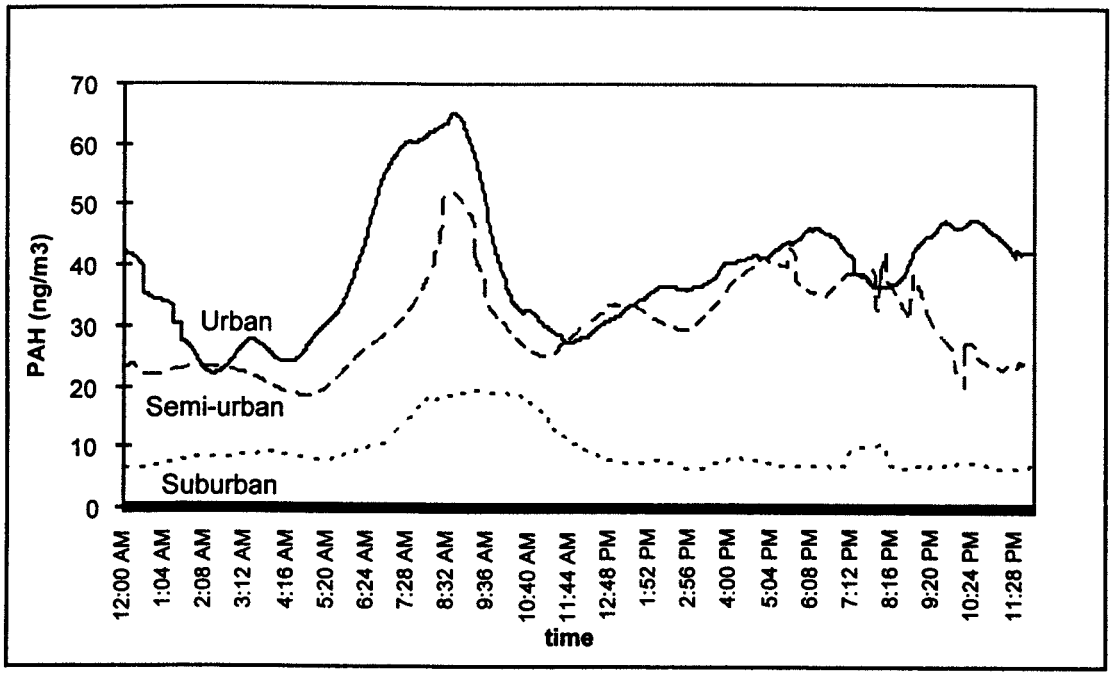

Figure 1. Weekday hourly PAH average indoor air concentrations (ng/m^3) due to outdoor sources (i.e., the effects of all indoor sources have been removed) at an urban, semi-urban, and suburban residence.

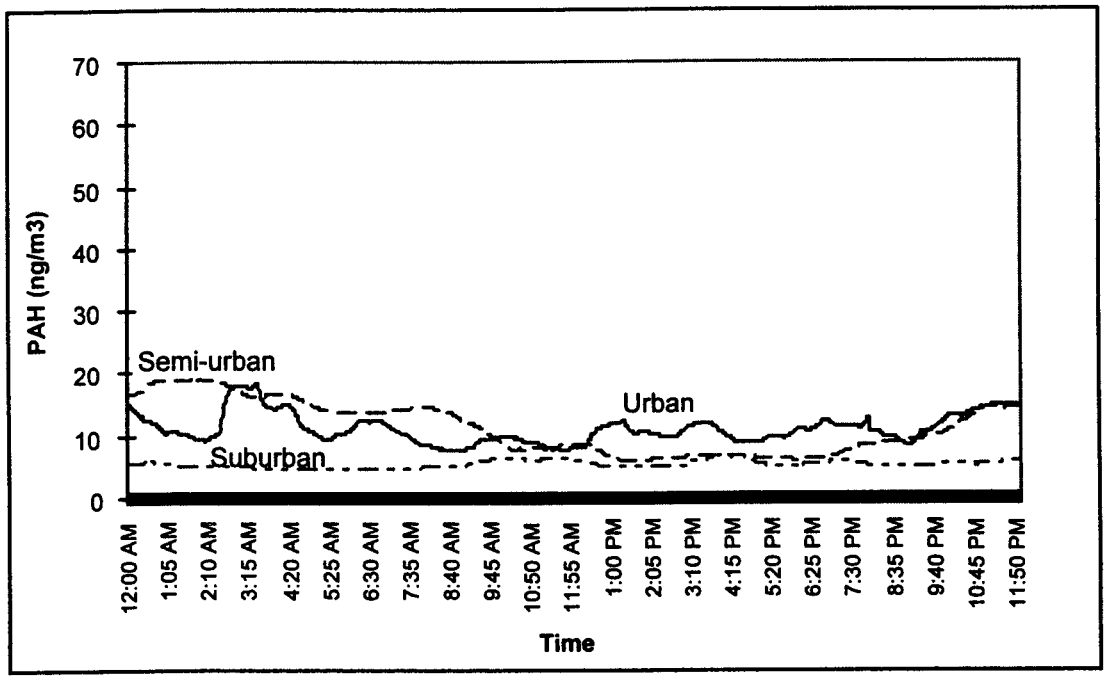

Figure 2. Weekend hourly indoor air PAH average concentrations (ng/m^3) due to outdoor sources (i.e., the effects of all indoor sources have been removed) at an urban, semi-urban, and suburban residence.

(further discussion in the indoor source section presented below) and are presented separately for weekdays and weekends due to the influence of day of the week on PAH concentration profiles.

At all locations, the geometric means of total particle-bound PAH were found to be lower than the arithmetic means, implying that the data are best fit by a right-skewed distribution such as the log normal. This finding is consistent with previous observations of indoor PAH concentrations (Waldman et al., 1990). Comparing weekday means, the data suggest that the urban location has the highest geometric mean PAH concentration (31 ng/m^3) followed by the semi-urban (19 ng/m^3) and suburban (8.0 ng/m^3) locations. During the weekends, these differences in concentration were less pronounced and the overall geometric mean concentrations were lower than the respective weekday concentrations (i.e., 10, 10, and 4.9 ng/m^3 at the urban, semi-urban, and suburban locations, respectively).

Performing a two-sided *t*-test for unequal variances on the log-transformed data indicated that all locations were statistically different from one another during the weekdays ($p<0.001$). For the weekends, no statistical difference ($p\leq0.05$) was observed between the urban and semi-urban

Table 1. Summary indoor PAH concentrations (ng/m^3) attributable to traffic at three residences (indoor source contributions removed).

Location	*n*	Minimum	Maximum	Mean	Standard deviation	Geometric mean	Geometric standard deviation
Weekday							
Urban	13,378	5	255	39	25	31*	2.0
Semi-urban	11,073	5	150	26	25	19*	2.2
Suburban	13,143	<LOD	85	9.3	5.7	8.0*	1.7
Weekend							
Urban	2734	5	90	11	5.9	10	1.5
Semi-urban	9912	<LOD	35	11	6.3	9.8	1.7
Suburban	7865	<LOD	30	5.1	1.9	4.9*	1.5

Notes: *n*=number of measurements; LOD=limit of detection.
*Location concentration (within weekday or weekend) is significantly different ($p<0.001$) from other locations based on two-sided *t*-test with unequal variances.

locations; however, the PAH concentrations at the suburban location were significantly lower ($p<0.001$) than both the urban and semi-urban locations. Weekend concentrations at all three locations were significantly less (two-sided *t*-test; $p<0.001$) than weekdays by a factor of 2–3. This difference between the workweek and weekends provides further evidence of an effect due to traffic.

Indoor Sources

In order to isolate the effect of traffic on indoor PAH concentrations, the data and analyses presented above were adjusted for indoor sources. This adjustment was performed by identifying, quantifying, and removing periods of indoor PAH generation from the concentration profile. This analysis was useful not only in isolating the effect of traffic, but also in providing an assessment of the indoor source contribution among these nonsmoking households.

For the purposes of this analysis, combustion events were determined to generate an increase in PAH concentrations if there was an observed increase in the PAS signal of at least 15 ng/m^3 during the period in which the event was reported to have occurred in the combustion activity journals. The increase of 15 ng/m^3 was compared with the average PAH concentration during the 20 min prior to the event (background). The value 15 ng/m^3 was chosen to distinguish an event from random fluctuations in PAH concentrations and was obtained empirically from inspection of the exposure profile.

Since it was not feasible for the occupants of the urban location to maintain a record of cooking or combustion events, a verbal survey was conducted. This survey revealed that few indoor sources were present. Specifically, it was reported that two toaster ovens were occasionally used, candles were sometimes lit by a fourth-floor resident, and one resident smoked cigarettes on the outside stoop. In order to assess the impact of these sources on indoor PAH concentrations, three exposure simulations were performed: burning toast, burning candles, and smoking outside. Each of these tests was conducted at the normal source location. None of the three simulations elicited a monitor response. This result is likely a function of the extremely large volume of the house, the high air-exchange rate, and/or the small size of the PAH source. Under such conditions, these data suggest that indoor sources of PAH at the urban location were negligible.

Inspection of the concentration profiles for the semi-urban and suburban residences, as defined above, indicated that the semi-urban location was the only residence to be affected by indoor combustion sources. At this location, cooking event peaks were detected for nine of 12 frying/sautéing events, three of five baking events, one of two broiling events, and one of 13 boiling events. No PAH generation was detected for any of the 16 cooking events at the suburban location. Therefore, adjustment was performed on the PAH concentration profiles at the semi-urban location by removing periods of indoor PAH generation, as previously defined, from the exposure profiles. While this procedure allows for comparison between locations, it likely caused an underestimation of mean daily PAH at the semi-urban location, since cooking frequently occurred during rush hour. The urban and suburban locations were unaffected. At the semi-urban location, measurements collected on August 28 and 29, 1997 were also removed from the analysis, since the parking lot of the apartment complex was being resurfaced during those days and asphalt contains PAHs.

The differences between PAH generation during cooking events at the semi-urban and suburban locations are most likely explained by differences in cooking styles. For example, residents at the suburban location cooked using baking and boiling in contrast to frequent stir-frying by the semi-urban residents. Another possible explanation for the differences in PAH generation is the stove type. The semi-urban location was equipped with a gas range, while the suburban location was electric. While one previous study showed that electric stoves resulted in higher PAH generation than gas stoves, it failed to consider the differences in cooking styles between locations (Chuang et al., 1991). As no other studies were found which addressed this issue, the results appear to be inconclusive.

Further evidence of the importance of indoor combustion sources at the semi-urban residence can be found in the high variability in the PAH concentration profile seen at the semi-urban location during both weekdays and weekends. Arithmetic standard deviations of 38 and 15 ng/m^3 and geometric standard deviations of 2.4 and 1.8 were recorded for this location during weekdays and weekends respectively. Furthermore, the highest PAH peaks of all locations were found at this location. Concentrations reached 680 and 675 ng/m^3 during the weekdays and weekends, respectively. In contrast, peak PAH concentrations at the suburban residence only reached 255 and 90 ng/m^3 and 85 and 30 ng/m^3 at the urban residence on weekdays and weekends, respectively.

Table 2 lists the peak PAH release, peak PAH concentration, and total observed duration for each semi-urban combustion event. These indoor source events were further classified into categories of frying/sautéing, baking, boiling, broiling, and candle-burning. While baking and boiling may not directly generate PAHs, they may be generated indirectly from accidental spills, overcooking, or the use of dirty supplies or appliances. Cigarette smoke was not included in this study as a combustion category as all locations were nonsmoking residences.

Peak PAH release was calculated by multiplying the maximum elevation in PAH concentration by the volume of the residence. This calculation assumes complete mixing

Table 2. Peak PAH release (μg), concentration (ng/m^3) and total observed duration (min) for indoor combustion events at the semi-urban location.

Category	Peak release (μg)	Peak concentration (ng/m^3)	Total observed duration (min)
Fry/sauté	160	670	42
Fry/sauté	140	595	61
Fry/sauté	130	535	147
Fry/sauté	120	520	76
Fry/sauté	50	210	153
Fry/sauté	35	150	225
Fry/sauté	18	75	87
Candles	18	75	56
Fry/sauté	17	70	140
Candles	12	50	50
Bake	11	45	72
Boil	8.4	35	37
Bake	7.2	30	32
Bake	6.0	25	124
Broil	6.0	25	26
Fry/sauté	3.6	15	29
Fry/sauté (3)[a]	<LOD	<LOD	–
Bake (2)[a]	<LOD	<LOD	–
Broil (1)[a]	<LOD	<LOD	–
Boil (12)[a]	<LOD	<LOD	–

LOD=Limit of detection.
[a]Events that generated no PAH have been listed together with the number of events in parentheses.

and was performed to facilitate comparison with other studies. Peak PAH concentration was also reported. The observed duration of PAH generation for a given event was determined by the length of time between the initial elevation in PAH and the time at which the PAH levels returned to background. If, however, the PAH concentrations increased after the documented end of the event, this new rise in PAHs was not attributed to the event. In other words, once an unrelated increase of PAHs was seen, the event was assumed to have ended despite the fact that PAH levels had not yet returned to background.

The results indicate that frying/sautéing yielded the greatest generation of PAH, with a maximum peak release of 160 μg and an average peak release of 56 μg. These PAH releases were roughly ten to 20 times greater than that of any other source. Candle-burning generated the second highest elevation in PAH followed by baking, boiling, and broiling with maximum peak releases of 18, 11, 8.4, and 6.0 μg, respectively. Cooking events ranged in duration from 29 to 225 min for frying/sautéing, 50 to 56 min for candle-burning, 32 to 124 min for baking, 37 min for boiling, and 26 min for broiling.

Potential PAH dose was calculated by integrating the increase in PAH concentration above background over the duration of the event, dividing by the exposure duration, and multiplying by an average inhalation rate of 20 m^3/day (0.0139 m^3/min) (DHHS, 1995). Average potential dose represents the average potential dose for all events under a given category. Potential dose is intended to provide a rough indication of the burden to a human for a given event. In fact, this quantification is not intended to be absolute, but rather it is meant to provide a means for comparing the potential risk of different source types. Potential dose is a better estimate of risk, since it includes an estimation of increased PAH levels, length of exposure, and human inhalation rates (Lioy, 1990).

These results indicate that average potential dose is highest for frying/sautéing (72 ng) followed by baking (12 ng), candle-burning (7.8 ng), broiling (1.8 ng), and boiling (0.72 ng). Thus, the overall order of importance between combustion categories differs between peak PAH and potential dose due to the incorporation of exposure time. While frying/sautéing generated the largest peak exposure and potential dose, baking has now become more important than candle-burning due to duration. Candle-burning had a larger peak exposure, but the length of exposure was higher for baking. Boiling became the least significant source when an averaging function was performed for average potential dose. Since only one of 13 boiling events created a peak, the significance of boiling was greatly diminished.

These observations regarding the contribution of cooking to indoor PAH concentrations should be considered as preliminary evidence of a significant source. Strict numerical interpretation of the data must be viewed cautiously due to limitations of potential dose calculations discussed earlier and the lack of verification of the PAS monitor with analytical chemistry. However, since these and earlier results indicate that cooking may substantially increase human exposures to PAH, more research is needed to better evaluate the contribution of cooking to a person's total inhaled dose.

Variability Due to Physical Parameters

While the PAH concentrations for the three locations are clearly different, there exists the possibility that the effect of interest (i.e., traffic) is confounded by other differences between the locations. Temperature and humidity were examined as additional potential confounders. While average temperatures between the locations varied between 76° and 89°F, a linear regression suggested that PAS response was unrelated to temperature for all locations (r^2=0). This finding is supported by previous studies showing no significant trends between temperature and PAS response or PAH concentrations under normal environmental conditions (Wilson et al., 1994; Lee et al., 1998). Wilson et al. (1994) also reports that PAS response was unaffected by relative humidity. Furthermore, in this study, no consistent increases or decreases in PAH concentrations were seen between periods with or without

air conditioner use. Although air-exchange rates were not measured, this factor would not be expected to affect the long-term 24-h integrated measurements upon which this study's conclusions are based.

Measurements of wind direction and speed indicate that the prevailing wind was predominantly from the west at all three locations. This result strengthens the comparison between locations, since traffic to the west is well-correlated to the assigned location category. For example, within 1 mile directly west of the urban location is the Massachusetts Turnpike (0.5 miles) and Commonwealth Avenue (0.4 miles). Harvard Street runs directly west of the semi-urban location at a distance of 0.6 miles and due west of the suburban location (0.6 miles) is Weston Road. Average wind speed was slightly higher at the semi-urban and urban residences than at the suburban residence.

Comparisons to Previous Work

The mean PAS concentrations in the range of 5 to 35 ng/m^3 reported for the current study are considerably lower than mean levels of 128 and 150 ng/m^3 reported for driving in traffic in North Carolina and California, respectively (Buckley and Ott, 1996). Wilson et al. (1994) reported very similar concentrations (5 to 40 ng/m^3) of those seen in this study for a home impacted by outdoor diesel construction machinery. Wallace et al. (1997) reported mean PAH levels (sampling every minute using the PAS 2000 series) as 48, 55, 34, and 22 ng/m^3 in a suburban private home for fall, winter, spring, and summer, 1996–1997, respectively. The higher levels in fall and winter were attributed to wood-burning in neighboring homes. For each of these studies, the same calibration factor of 1 μg m^{-3} pA^{-1} was employed.

Conclusions

The results of this study indicate that both outdoor and indoor sources contribute to particle-bound PAH concentrations in indoor air. Weekday concentration profiles indicate that traffic was the primary outdoor source determinant for indoor PAH levels. Graphs of PAH versus time indicated distinct rush hour PAH peaks during weekdays and none during weekends. Also, the differences in average concentrations found at each residence were consistent with traffic levels.

Following adjustment for indoor sources, outdoor contributions during weekdays were highest at the urban location (31 ng/m^3) followed by the semi-urban location (19 ng/m^3) and finally, the suburban location (8.0 ng/m^3, $p<0.01$). During weekends, only the suburban location (4.9 ng/m^3) was statistically different from the other locations (9.8 and 10 ng/m^3, $p<0.01$). Thus, this study qualitatively demonstrates that traffic has a significant and measurable impact on levels of indoor PAH. Additional studies are needed in order to build on this qualitative assessment to quantify the impact of traffic on residential indoor PAH. Quantitative assessment may also require consideration of air-exchange rates and site-specific meteorology.

The largest indoor source in these nonsmoking households was found to be cooking despite the fact that PAHs were not generated during every event. Frying/sautéing events generated the highest levels of PAH followed by candle-burning, baking, one boiling event, and broiling. Potential dose was also highest for frying/sautéing. Potential doses attributable to other indoor sources were far less important.

Additional research is needed to further evaluate the contribution of cooking sources to indoor airborne PAH concentrations. The current study provides preliminary evidence that cooking may contribute substantially to daily PAH exposure. Specifically, characteristics which influence the extent of PAH formation should be investigated, including the impact of gas versus electric ranges. Additionally, there is a need for a personal monitoring strategy to accurately assess the added burden of PAHs from exposure to combustion or cooking events.

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