Coal-Tar-Based Parking Lot Sealcoat: An Unrecognized Source of PAH to Settled House Dust

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Despite much speculation, the principal factors controlling concentrations of polycyclic aromatic hydrocarbons (PAH) in settled house dust (SHD) have not yet been identified. In response to recent reports that dust from pavement with coal-tar-based sealcoat contains extremely high concentrations of PAH, we measured PAH in SHD from 23 apartments and in dust from their associated parking lots, one-half of which had coal-tar-based sealcoat (CT). The median concentration of total PAH (T-PAH) in dust from CT parking lots (4760 μg/g, $n = 11$) was 530 times higher than that from parking lots with other pavement surface types (asphalt-based sealcoat, unsealed asphalt, concrete [median 9.0 μg/g, $n = 12$]). T-PAH in SHD from apartments with CT parking lots (median 129 μg/g) was 25 times higher than that in SHD from apartments with parking lots with other pavement surface types (median 5.1 μg/g). Presence or absence of CT on a parking lot explained 48% of the variance in log-transformed T-PAH in SHD. Urban land-use intensity near the residence also had a significant but weaker relation to T-PAH. No other variables tested, including carpeting, frequency of vacuuming, and indoor burning, were significant.

Introduction

Settled house dust (SHD) is an important source for indoor exposure to numerous contaminants (*1*), particularly for children (*2, 3*), who spend a substantial amount of time on the floor and who put their hands and objects into their mouths (*4*). Numerous studies have documented occurrence of polycyclic aromatic hydrocarbons (PAHs) in SHD in the United States and Europe (e.g., refs *5–12)*. Exposure to PAHs is of concern because PAHs are ubiquitous in the urban environment and because several have been identified as probable carcinogens (B2 PAHs) (*13*).

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There are numerous potential indoor and outdoor sources of PAHs to SHD, which is a complex mixture of biological material, particulate deposition of indoor aerosols, and particles tracked in from the outdoors (*14*). PAHs are formed during the incomplete combustion of carbonaceous material, including wood, coal, food, motor oil, and gasoline. Researchers, however, have remarked on the lack of success in identifying the principal sources contributing to the PAH content of SHD (*1, 9*). Maertens et al. (*9*) compiled data for PAH composition and concentrations in SHD from 18 published studies and investigated relations between PAHs and numerous site attributes and lifestyle variables. They determined that only tobacco smoking (significant in urban homes only) and home location (urban vs rural) were related to PAH content, and that the relations were weak. The significance of tobacco smoking as a factor affecting PAH concentrations has been corroborated by some studies (*10, 12, 15*) but not by others (*5, 11*). At least one other study (*12*) found that rural areas had lower concentrations of PAHs in SHD than did urban areas, although only two samples from rural areas were analyzed. Other factors, such as heating with coal (*10*), vehicle emissions (*10*), and carpeting (*11*), cited as potential explanatory variables for differences in PAH concentrations, have not been demonstrated to be significant.

A recently identified outdoor source of PAHs to the environment (*16, 17*)—coal-tar-based pavement sealcoat—has not been considered in any previous investigations of PAHs in SHD. Sealcoat is the black liquid that is sprayed or painted on the asphalt pavement of many parking lots, driveways, and playgrounds in the U.S. and Canada in an attempt to improve appearance and increase pavement longevity. There are two principal formulations of sealcoat: one with a refined coal-tar-emulsion (RT-12 grade) base and one with an asphalt-emulsion base. Coal tar is a known carcinogen that is more than 50% PAH by weight (*18*); sealcoat with a coal-tar base typically is 15 to 35% refined coal tar. The median PAH concentration (sum of 16 parent PAHs) for coal-tar-based and asphalt-based sealcoat products has been reported to be >50 000 and 50 μg/g, respectively (*19*). In the United States, coal-tar-based sealcoat is used predominantly east of the Continental Divide, where the median concentration of PAHs in dust swept from parking lots in six cities was 2200 μg/g (*17*). In the western United States, although coal-tar-based sealcoat is available, use of the asphalt-based product dominates, and the median concentration of dust from sealcoated parking lots in three western cities was 2.1 μg/g (*17*). Sealcoat abrades into mobile particles that can be carried offsite by water, wind, or mechanical tracking (e.g., tires, snowplows) (*20*).

In this study, we investigated the relation between PAH concentrations in SHD and several potential explanatory factors, including sealcoated parking lot surfaces, by analyzing dust from ground-floor apartments in Austin, TX, and from the associated apartment-complex parking lots, one-half of which had coal-tar-based sealcoat. Although the use of coal-tar-based sealcoat was banned in Austin in 2006 (*21*), on the basis of a rapid screening test (Supporting Information) we determined that more than one-half of parking lots screened for possible inclusion in this study were still coated with coal-tar sealcoat applied before the ban. For each residence, data were collected on a wide range of lifestyle variables and site characteristics that might affect PAH concentrations. The resulting information was analyzed statistically to determine those factors related to concentrations of PAHs in SHD.

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TABLE 1. Independent Variables Tested for Relation to Total PAH in Settled House Dust and Parking Lot Dust[a]

binary response variables	scalar response variables
sample site location (indoors or outdoors)	urban land-use intensity
parking lot surface type (coal-tar sealcoat or not)	distance from parking lot to front door
shoe wear indoors	proportion of sampling area carpeted
park bicycle indoors	days since sampling area last vacuumed
barbecue grill use	amount of time windows left open
regularly burn candles, incense, or oil lamp	frequency of fireplace use
allow pet to go outdoors	number of trips daily to and from front door to parking lot
desktop computer in living area	degree of sealcoat wear
	distance from major roadway

[a] Summary of ancillary data is given in Supporting Information.

Methods

Site Selection and Sample Collection. Dust was collected in April through July, 2008, from 23 ground-floor apartments and the associated parking lots in Austin, TX. No particular subpopulation or geographic area was targeted, and the apartments are believed to be reasonably representative of apartments in the central Texas area. None of the households included children, although this was not intentional. Apartment complexes were prescreened for parking lot type, with the objective of choosing about one-half of the residences with coal-tar-sealcoated ("coal tar," or CT) parking lots and one-half with another surface type (asphalt sealcoat, unsealcoated asphalt, or concrete; "not coal tar," or NCT). Coal-tar and asphalt sealcoat were distinguished at the reconnaissance level by use of the "coffee/tea test," which is based on the solubility of sealcoat scrapings in mineral spirits (Supporting Information for description and validation). On the basis of this preliminary test, 11 of the residences were hypothesized to be in apartment complexes with CT parking lots and 12 in complexes with NCT parking lots. Of the 12 NCT parking lots, seven were unsealcoated asphalt pavement, two were concrete, and three were asphalt pavement with asphalt-based sealcoat.

Apartment residents provided information on a variety of lifestyle activities and actions that might influence PAH concentrations, including tobacco smoking, cooking and vacuuming habits, wearing of shoes indoors, and burning of candles and incense. Physical characteristics of the apartments were noted, including the nature of indoor furnishings, carpeting, heating and cooking sources, and nearby businesses. All apartments had central heat (either gas or electric) and air conditioning. The distance from the parking lot to the front door was measured, distance from the residence to the nearest major road (defined as four-lane) was measured from aerial images, and land use within a 250-m radius of each apartment was determined from a GIS data set (*22*). See Supporting Information for a summary list of ancillary data and observations.

Dust was collected using a Model HVS3 High-Volume Surface Sampler—the American Society for Testing Materials (ASTM) standard method for recovering SHD for chemical analysis—following the methods recommended by the manufacturer for collection of indoor dust (*23*). SHD was collected from entryway and adjacent living room floors. Outdoor dust was collected from parking spaces, avoiding painted lines and drip areas. The indoor area sampled ranged from 1.6 to 13 m^2 (median of 3.6 m^2) and the outdoor area sampled ranged from 2.0 to 7.5 m^2 (median of 4.8 m^2). After collection, the dust was weighed and the coarse (>0.5 mm) fraction removed by sieving; only the remaining fraction was analyzed. The mass of sieved dust collected indoors ranged from 0.36 to 35 g (median of 4.8 g) and outdoors from 2.4 to 112 g (median of 11 g). Samples were transported on ice, stored at 4 °C, and shipped on ice to the U.S. Geological Survey National Water Quality Laboratory (NWQL) in Lakewood, CO, for analysis of PAH.

Chemical Analysis and Quality Control. At the NWQL, samples were prepared for gas chromatograph/mass spectrometer (GC/MS) analysis according to (24) and analyzed by GC/MS, either with the MS operated in the electron impact, full-scan mode or in the selected ion monitoring (SIM) mode. Additional information on chemical analysis and quality control are in the Supporting Information.

Total PAH (T-PAH) was determined as the sum of 16 parent PAH corresponding to the 16 PAH priority pollutants identified by the U.S. Environmental Protection Agency: anthracene, benzo[*a*]pyrene, fluoranthene, naphthalene, benz[*a*]anthracene, phenanthrene, pyrene, fluorene, acenaphthene, acenaphthylene, benzo[*b*]fluoranthene, benzo[*ghi*]perylene, benzo[*k*]fluoranthene, chrysene, dibenz[*a,h*]anthracene, and indeno[1,2,3-*cd*]pyrene. For the summation of T-PAH, nondetections were assigned a value of zero. The summation also was done with nondetections assigned the value of the detection level; similar results for the statistical tests were obtained and are not reported here.

Statistical Methods. Normality of data was evaluated on the basis of visual inspection of probability plots (*25*). Nonparametric statistics were used on untransformed data to compute summary statistics. All other analyses were done using parametric statistics on log-transformed data. The Student's *t*-test was used to compare population means and to test effects of independent variables with a binary response (e.g., indoors vs outdoors); linear regression was used to test effects of independent variables with a scalar response (e.g., distance from residence to a major roadway) (Table 1). Analysis of Variance (ANOVA) was used to quantify strength of effects and interactions of binary variables. For all statistical tests, an effect was assumed to be significant at $p < 0.05$. *Statistica 8.0* (StatSoft, Inc.) was used for all statistical tests.

Results

Concentrations of PAH. Concentrations of T-PAH ranged over 4 orders of magnitude (1.04 to 11 300 μg/g, Table 2) and were log-normally distributed. Results for the individual 16 parent PAH are given in Table S1 of the Supporting Information. Benzo[*a*]pyrene (BaP) and the sum of the seven B2 PAHs (probable human carcinogens, as identified by the U.S. Environmental Protection Agency (*13*): benzo[*a*]pyrene, benz[*a*]anthracene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, chrysene, dibenzo[*a,h*]anthracene, and indeno[1,2,3-*cd*]pyrene) showed a similarly wide range of concentrations (Table 2 and Table S1 of the Supporting Information). Analytical difficulties with dibenzo[*a,h*]anthracene prevented detection of this compound in all but one sample, likely resulting in a modest underestimate of T-PAH and the total B2 carcinogenic PAHs.

TABLE 2. Concentrations of Benzo[*a*]pyrene (BaP), Sum of 16 Parent PAH (T-PAH), and Sum of the Seven B2 PAH Measured in Settled House Dust (SHD) and in Dust from the Associated Parking Lot[a]

		BaP (μg/g)		T-PAH (μg/g)		B2 PAH (μg/g)	
pavement surface type	parking lot designation	SHD	parking lot	SHD	parking lot	SHD	parking lot
coal-tar-sealcoated asphalt	CT	3.42	149	44.5	2300	21.8	1050
coal-tar-sealcoated asphalt	CT	15.2	671	191	10 300	94.1	4230
coal-tar-sealcoated asphalt	CT	10.9	305	152	5070	70.3	2040
coal-tar-sealcoated asphalt	CT	4.04	131	54.6	2010	29.4	1110
coal-tar-sealcoated asphalt	CT	14.3	36.7	214	591	104	282
coal-tar-sealcoated asphalt	CT	1.21	26.8	19.6	387	8.62	191
coal-tar-sealcoated asphalt	CT	1.41	21.4	26.4	405	18.2	166
coal-tar-sealcoated asphalt	CT	7.33	518	137	11 300	53.8	4020
coal-tar-sealcoated asphalt	CT	4.50	285	64.3	4760	30.1	2080
coal-tar-sealcoated asphalt	CT	4.44	555	129	8900	47.0	3940
coal-tar-sealcoated asphalt	CT	24.2	511	335	6960	156	3330
unsealed concrete	NCT	0.15	1.05	1.94	15.1	0.98	7.51
unsealed asphalt	NCT	1.36	2.97	18.3	48.7	9.35	20.1
asphalt-sealcoated asphalt	NCT	3.91	0.60	43.0	10.9	22.2	5.36
asphalt-sealcoated asphalt	NCT	0.58	0.30	6.05	4.43	3.17	2.36
unsealed asphalt	NCT	1.50	0.50	22.2	5.89	8.54	2.96
unsealed asphalt	NCT	2.05	3.36	27.1	42.0	11.7	22.1
unsealed concrete	NCT	12.4	0.49	194	7.79	85.8	3.60
unsealed asphalt	NCT	0.06	0.14	1.04	2.28	0.45	1.19
unsealed asphalt	NCT	0.26	0.19	4.12	3.32	1.81	1.43
unsealed asphalt	NCT	0.23	1.35	3.81	17.4	1.68	8.76
asphalt-sealcoated asphalt	NCT	0.30	0.56	4.11	10.2	2.27	5.20
unsealed asphalt	NCT	0.25	0.06	3.32	1.10	1.58	0.42

[a] CT, apartment complex has a coal-tar-sealcoated parking lot; NCT, apartment complex does not have a coal-tar-sealcoated parking lot.

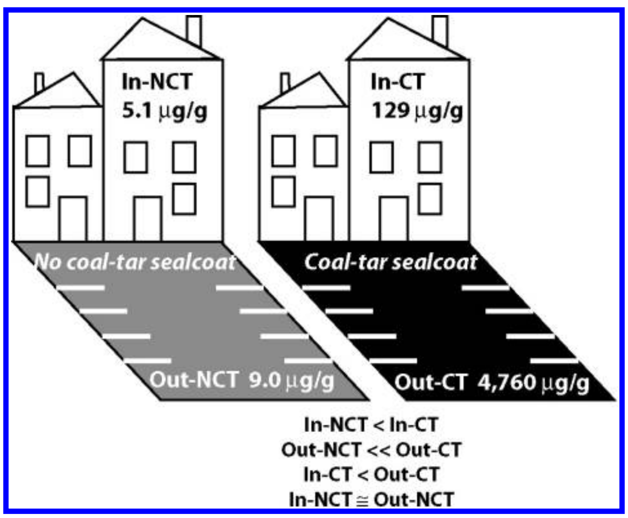

FIGURE 1. Comparison of median concentrations of sum of 16 PAHs (T-PAH) in dust vacuumed from inside 23 apartments and from their parking lots.

Relations Between Sample Location, Parking Lot Surface Type, and PAH Concentrations. T-PAH concentrations were different depending on the sample collection location (outdoors vs indoors) and the presence or absence of coal-tar-based sealcoat on the associated parking lot (Figures 1 and 2). The concentrations of T-PAH measured in parking lot dust (outdoors) form two distinct groups with no overlap: those in dust vacuumed from parking lots with coal-tar sealcoat (hereinafter, Out-CT) versus without coal-tar sealcoat (hereinafter, Out-NCT) − the lowest concentration of T-PAH measured in an Out-CT sample was eight times higher than the highest concentration measured in an Out-NCT sample. Overall, T-PAH measured in Out-CT dust was higher than that measured in Out-NCT dust by a factor of 530 (median concentrations of 4760 and 9.0 μg/g, respectively), a difference that greatly exceeds the sampling variability and analytical uncertainty as reflected by a median relative percent difference (RPD) of 23% for an outdoor replicate sample (details in the Supporting Information). Within Out-NCT samples, there was no significant difference in mean log T-PAH concentrations in dust from pavement sealed with asphalt-based sealcoat (median concentration 10.2 μg/g, $n = 3$) and unsealed pavement (asphalt or concrete pavement; median concentration 7.8 μg/g, $n = 9$) ($p = 0.94$).

The concentration of T-PAH measured in SHD from apartments with CT parking lots (hereinafter, In-CT) was significantly higher than that measured in SHD from apartments with NCT parking lots (hereinafter, In-NCT) by a factor of 25 (median concentrations of 129 and 5.1 μg/g, respectively). This difference greatly exceeds the sampling variability and analytical uncertainty as reflected by a median RPD of 47% for indoor replicate samples (details in the Supporting Information). In this sample set, however, there was some overlap: four In-NCT samples had T-PAH concentrations that were greater than the minimum concentration measured in In-CT samples. One of those four samples was an extreme outlier, with a concentration 4.5 times higher than the next highest In-NCT concentration and 38 times higher than the median In-NCT concentration (Figure 2).

Location of sample collection (indoors vs outdoors) was significant only for apartments with CT parking lots, for which the median T-PAH concentration in outdoor samples was higher than in indoor samples by a factor of 37. For apartment complexes with NCT parking lots, there was no significant difference in T-PAH concentrations between indoor and outdoor dust samples.

Pavement surface type (CT vs NCT) was a predominant factor affecting the concentration of PAH in both outdoor dust and SHD, as determined by ANOVA. Pavement surface type explained 86% of the variance in log T-PAH in parking lot dust, and 48% of the variance in log T-PAH in SHD. Log T-PAH measured in samples of SHD was linearly related to that measured in samples of parking lot dust ($r^2 = 0.59$, Figure 3). This relation arises to some degree because concentrations of T-PAH are low in In-NCT and Out-NCT samples and are

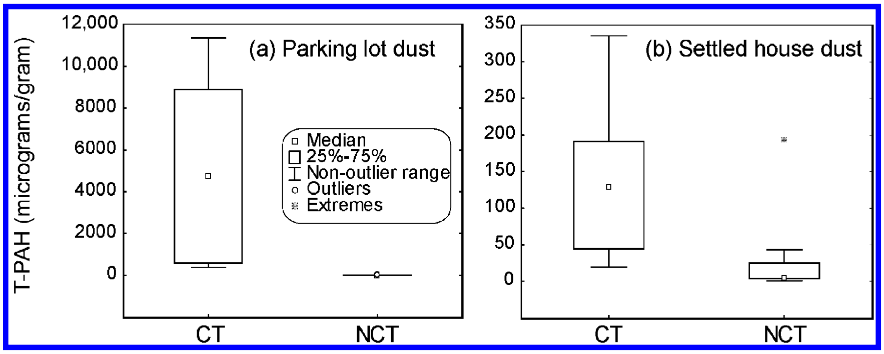

FIGURE 2. Comparison of concentrations of the sum of 16 parent PAH (T-PAH) measured in dust vacuumed from (a) parking lots with coal-tar sealcoat (CT) and without coal-tar sealcoat (NCT), and (b) in settled house dust from the associated apartments.

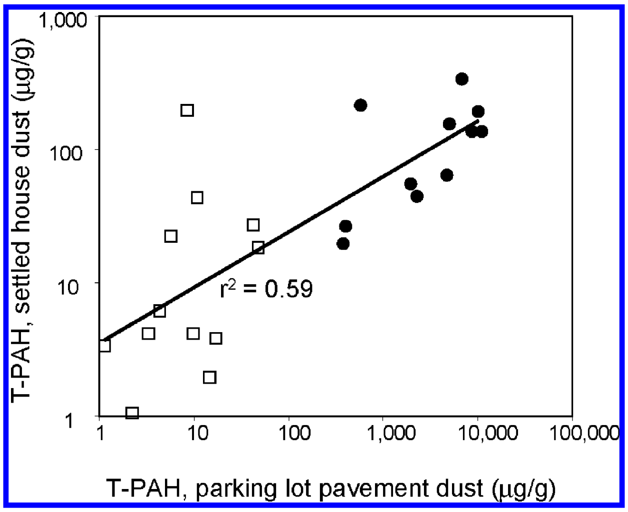

FIGURE 3. Relation between sum of 16 parent PAH (T-PAH) in dust vacuumed from parking lot pavement and in settled house dust. Apartments with coal-tar-sealcoated parking lots (●), and those with parking lots without coal-tar sealcoat (□).

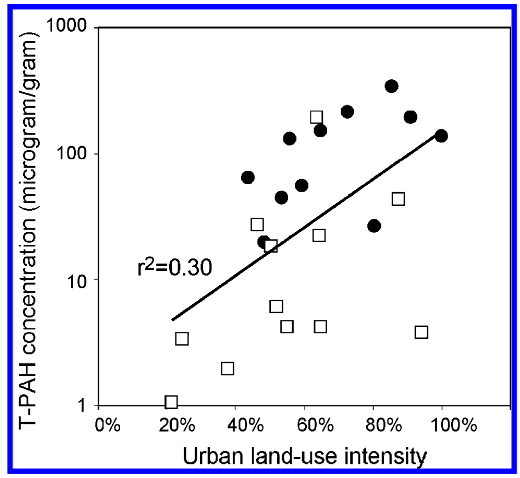

FIGURE 4. Relation between urban land-use intensity (sum of multifamily residential, commercial, office, warehouse, and streets and roads) and sum of 16 parent PAH (T-PAH) in settled house dust. Apartments with coal-tar-sealcoated parkings lots (●), and those with parking lots without coal-tar sealcoat (□).

high in In-CT and Out-CT samples. However, when CT and NCT samples are considered separately, log T-PAH in In-CT and Out-CT samples also are linearly correlated ($r^2 = 0.43$), although log T-PAH in In-NCT and Out-NCT samples are not.

Other Potential Explanatory Factors and Relation to PAH Concentrations in Settled House Dust. Tobacco smoking and land use (amount of urbanization) are two variables often cited as affecting PAH concentrations in SHD. For this study, effects of tobacco smoke could not be tested statistically because of the small number of smokers participating, of which only one smoked indoors. Of the three residences with a smoker, two had CT parking lots and one had an NCT parking lot. Concentrations of T-PAH in the two In-CT samples from residences with a smoker (which included the indoor smoker) were the lowest of the 11 In-CT samples, and concentrations of T-PAH in the In-NCT sample from a residence with a smoker was the fifth highest of the 12 In-NCT samples. Thus, for this study, there was no indication that environmental tobacco smoke contributed to elevated concentrations of PAHs in SHD.

The relation between intensity of urban land use surrounding the apartment and T-PAH in SHD was examined using linear correlation on log-normalized T-PAH concentration. Urban land-use intensity was defined as the percentage of land use consisting of the sum of multifamily residential, commercial, office, warehouse, and streets and roads within a 250-m radius of the apartment, and ranged from 22 to 100%. There was a statistically significant relation between log T-PAH concentration in SHD and urban land-use intensity ($r^2 = 0.30$, Figure 4). Urban land-use intensity might not be independent of parking lot surface type, however. The mean urban land-use intensity for NCT apartments was 55% and for CT apartments was 69%, although the difference between the means was not significant. Further, the residuals are not randomly distributed: Residuals for NCT samples are mostly negative and for CT samples are mostly positive, indicating that both urban land-use intensity and parking lot surface type likely explain some of the variability in T-PAH concentration.

Accordingly, parking lot surface type and urban land use were entered as independent variables into a multiple linear regression, with log T-PAH as the dependent variable. For indoor samples, both variables were significant and together explained 60% of the variance, following the equation

$$y = 0.26 + 0.83x_1 + 1.25x_2 \qquad (1)$$

where y is log T-PAH concentration, x_1 is parking lot surface type (0 for apartments with NCT parking lots and 1 for apartments with CT parking lots), and x_2 is land-use intensity (fractional from 0−1). Addition of an interactive term did not result in an improved adjusted r^2, so it was not retained in the model. For outdoor samples, only the independent variable parking lot surface type (x_1) was significant ($r^2 = 0.88$).

Numerous additional potential explanatory variables, categorical and scalar, were tested to determine whether they were related to T-PAH concentrations in SHD (Table 1), both with all indoor samples combined and separately by associated parking lot surface type (In-NCT and In-CT). The only significant relations were between T-PAH concentration and distance from the front door to the parking lot for In-

NCT samples (positive relation, $r^2 = 0.45$), and between T-PAH concentration and the proportion of the vacuumed area carpeted for both In-NCT (inverse relation, $r^2 = 0.38$) and In-CT (positive relation, $r^2 = 0.45$) samples.

Discussion

PAH Concentrations in Parking Lot Dust. The extremely elevated concentrations of T-PAH in dust vacuumed from CT parking lots and much lower concentrations in dust from NCT lots are consistent with those reported previously (*17*). Concentrations of T-PAH for Out-CT samples (median of 4760 μg/g) are similar to those reported for dust swept from sealcoated parking lots in six central and eastern U.S. cities (median of 2200 μg/g) (*17*). Similarly, T-PAH concentrations in Out-NCT samples (median 9.0 μg/g) were similar to those in dust swept from unsealcoated parking lots in the same six cities (median 27 μg/g), and to dust from sealcoated and unsealcoated parking lots in three cities in the western United States, where coal-tar-based sealcoat use is minimal (median 1.5 μg/g) (*17*). The wide range in Out-CT T-PAH concentrations (387 to 11 300 μg/g) (Table 2) likely results from differences in sealcoat age, formulation and application, parking lot use, and amount of dilution (e.g., by soil), among other factors. On the basis of ANOVA and regression, virtually all of the variance in T-PAH concentrations measured in parking lot dust for this study could be explained by the presence or absence of coal-tar sealcoat. We note that T-PAH associated with dust from CT parking lots remains high even though the minimum sealcoat age is ~2.5 y (use of CT sealcoat was banned in Austin in August 2006).

There was no statistically significant difference between T-PAH concentrations in dust from typical non-CT pavement surface types (concrete, unsealcoated asphalt, and asphalt-based sealcoat; range of 1.10 to 48.7 μg/g). This result was different from that of ref *16*, who found higher T-PAH associated with particles in runoff from parking lots with asphalt-based sealcoat relative to that from unsealed parking lots. The difference likely is because the asphalt-based sealcoat on the lots tested by (*16*) had been applied over worn coal-tar sealcoat, whereas the asphalt-based sealcoat on the parking lots tested for this study had been applied over new asphalt pavement.

Coal-tar-sealcoated parking lots and driveways represent a large reservoir of mobile PAHs because of the extensive area they cover and the elevated concentrations of T-PAH associated with them. Of four mapped watersheds in Texas, sealcoated parking lots constituted 1 to 2% of the total watershed area (*16*). In the town of Lake in the Hills, Illinois, a Chicago suburb, sealcoated pavement constituted 4% of the watershed area; 42% of parking lot area in the watershed was sealcoated, and 89% of driveway area was sealcoated (U.S. Geological Survey, unpublished data, 2009). The T-PAH concentration in dust from CT parking lots, generally in the range of 1000−10 000 μg/g, exceeds by 1 to 3 orders of magnitude that in other outdoor sources that might contribute PAHs to SHD, including tire particles (226 μg/g (*26*)); road dust (59 μg/g (*26*)); asphalt (2.25 μg/g (*27*)); and diesel engine (17.5 μg/g (*28*)) and gasoline engine (35.0 μg/g (*28*)) emissions. Parking lot and driveway pavement are the surfaces on which many people walk directly before entering their residence. Abraded sealcoat particles on parking lots thus are likely to be tracked indoors on shoes or bare feet, bicycle tires, and even pets' feet.

PAH Concentrations in Settled House Dust and Contribution from Coal-Tar Sealcoat. The relation between PAH concentrations in SHD and the presence or absence of CT sealcoat on the associated parking lot provides insight regarding the wide range in PAHs reported for other studies. The mean and median concentrations of T-PAH for In-NCT samples from this study are consistent with those for mean or median T-PAH in SHD reported by other investigators. For example, the arithmetic mean for T-PAH computed for In-NCT (27.4 μg/g, sum of 16 parent PAHs) is similar to the arithmetic mean (28 μg/g, sum of 18 PAHs) reported for total PAH in SHD from a compilation of 18 studies (*9*). Mean or median total PAH concentrations cited by other recent publications are similarly low, for example, 6.4 μg/g (sum of 18 PAH (*10*)), 5.1 μg/g (sum of 18 PAH (*12*)), and 29.3 μg/g (sum of 13 PAH (*11*)).

In contrast, the mean T-PAH in In-CT samples (124 μg/g) exceeds not just the mean or median but also the maximum T-PAH reported by most studies (e.g., 26.7 μg/g (*7*), 55.1 μg/g (*29*), 21.0 μg/g (*10*)). It is of the same order of magnitude as the maximum (325 μg/g) reported by Maertens et al. (*11*) but is similar to the mean reported by Chuang et al. (121 μg/g (*30*), as reported in (*9*)) for a study in Ohio. These researchers were not able to identify a source for such elevated PAH concentrations in SHD, which exceed those in most candidate PAH source materials. Our results indicate that coal-tar sealcoat might be that source.

Regional variations in use of coal-tar-based sealcoat might explain some differences between values reported in other studies. For example, Butte (*31*) notes that the 95th percentile of BaP in SHD in the United States was higher than that in SHD in Germany by a factor of as much as about 12, even though sampling and sample preparation techniques were identical, but can offer no explanation. Pavement sealcoat is not used in Germany, whereas coal-tar-based sealcoat is available in all 50 U.S. states and Canada (*17*). Lewis et al. (*8*) speculate that the cause of the difference between the mean concentration of B2 PAHs reported for Columbus, OH (72 μg/g), and for Seattle, WA (11 μg/g), might be different types of home heating or geographic differences in exposure to tobacco smoke. We suggest that the difference might be attributable to regional differences in sealcoat use.

Relation of PAH in Settled House Dust to Coal-Tar-Based Sealcoat and Other Explanatory Factors. Of the numerous explanatory variables tested for relation to PAH concentrations in SHD (all samples combined), only two were significant: presence or absence of coal-tar-based sealcoat on the associated parking lot and urban land use within a 250-m radius of the residence. Individually, presence or absence of a CT parking lot explained 48% of the variance in T-PAH in SHD, and the intensity of urban land use explained 30%. When combined in multiple linear regression, together they explained 60% of the variance in T-PAH concentrations in SHD. This percentage is much more than other independent variables previously identified, such as environmental tobacco smoke and urban location, which individually explained 10 and 13% of total PAH variance, respectively (*9*).

A strong control exerted by parking lot surface type is reasonable, given the greatly elevated concentration of T-PAH in Out-CT samples: A contribution of only about one part Out-CT in 38 parts SHD is necessary to elevate T-PAH in SHD from the median In-NCT to the median In-CT concentration (5.1 and 129 μg/g, respectively) measured in this study. The stronger relation between urban location and T-PAH concentration in SHD determined in this study relative to that cited for earlier studies might result from a more precise and scalar quantification of land use in the area immediately surrounding the residence. Most elements of the urban land-use intensity metric (multifamily residential, commercial, office, warehouse, and streets and roads) are likely to have PAH sources, including sealcoated parking lots that might contribute to PAHs in SHD by offsite transport by wind and tracking. We note, however, that vehicle emissions do not appear to contribute significantly to PAH in SHD, as there was no significant relation between distance to a major roadway and T-PAH in SHD.

We tested many other of the potential factors previously hypothesized to affect PAH concentrations in SHD (Table 1), including amount of carpeting (*11*), frequency of vacuuming (*11*), and presence of a desktop computer (*15*). None of the factors had a significant relation to T-PAH concentration except proportion of sampling area carpeted and distance from the front door to the parking lot and then only for subsets of the population. For carpeting, the direction of the relation was positive for In-CT and negative for In-NCT. Carpeting might more efficiently trap abraded particles tracked in from outdoors, including coal-tar-sealcoat particles in the In-CT samples (increasing T-PAH concentrations), and relatively uncontaminated particles in the In-NCT samples (diluting T-PAH concentrations). The positive relation between distance from the front door to the parking lot and T-PAH in In-NCT samples might result from the same process − fewer uncontaminated particles tracked in to dilute T-PAH. The lack of a significant relation between the other potential explanatory variables and T-PAH concentration does not necessarily mean there is no relation but rather might arise because that relation is relatively weak and the sample size relatively small.

A single extreme outlier in the In-NCT sample set (T-PAH of 194 μg/g) raises the question of indoor PAH sources. The T-PAH concentration in the SHD sample was about 25 times higher than in the associated parking lot dust sample (7.8 μg/g), but no indoor source of elevated PAH could be identified − residents were nonsmokers, used electricity for heating and cooking, did not use a barbecue or fireplace, and rarely burned candles or incense. One indoor source of T-PAH that might contribute to such elevated concentrations in SHD is coal-tar adhesive in flooring. Hansen and Volland (*32*) reported high concentrations (149 to 762 μg/g) of PAHs in SHD associated with coal-tar-based flooring adhesives in Germany but only where parquet flooring had been damaged. Similar coal-tar adhesives were used for linoleum in the United States into the 1980s (David Nick, DPNA International Inc., written communication, 2009). The apartment identified in this study was built in 1980 and had a wood-laminate floor, although at the time of sampling the condition of the flooring was not inspected for damage. Of the four other NCT apartments in this study with wood-laminate or linoleum flooring, only one was built prior to 2000 and it did not have elevated T-PAH concentrations.

PAH Concentrations in Settled House Dust from Apartments with Coal-Tar-Sealcoated Parking Lots in Relation to Health Guidelines and Nondietary Ingestion. The only existing guideline for PAH concentrations in SHD is 10 μg/g for BaP, issued by the German Federal Environmental Agency's Commission for Indoor Air Quality (*32*) in response to concerns about coal tar in wood-flooring adhesive. BaP concentrations measured in SHD for this study exceeded that guideline in four of 11 In-CT samples (36%) and one of 12 In-NCT samples (the outlier previously discussed, 8%). In parking lot dust, BaP in all 11 of the Out-CT samples exceeded the guideline, in some cases by a factor of 50 or more, but did not exceed the guideline in any of the 12 Out-NCT samples. Although outdoor dust might not result in the same type of exposure as SHD, outdoor activities such as basketball playing or chalk drawing on CT parking lots, driveways, and playgrounds might result in exposure levels of BaP that would be considered elevated in relation to the German guideline.

Maertens et al. (*11*) calculated the excess cancer risk resulting from nondietary ingestion of carcinogenic (B2) PAHs in SHD during preschool years. They reported that 10% of the households they sampled had a concentration of B2 PAHs that exceeded 40 μg/g, resulting in an excess cancer risk of greater than 1×10^{-4} for a "high" dust ingestion rate scenario of 0.1 g/day. Of the 11 apartments with CT parking lots sampled for this study, six (55%) had a concentration of B2 PAHs that exceeded 40 μg/g (Table 2), indicating that use of coal-tar sealcoat on parking lots and driveways is related to elevated concentrations of carcinogenic PAHs in SHD.

The high concentrations of B2 PAHs in SHD from apartments with CT parking lots imply that past evaluations might have underestimated the importance of nondietary ingestion of PAHs by preschoolers for many residences. Studies that have evaluated the relative importance of dietary and nondietary exposure to PAHs have been based on concentrations of B2 PAHs in SHD of 1.73 μg/g (ref *7*; estimated to constitute about 24% of total dose to preschoolers) and 0.925 μg/g (ref *29*; estimated to constitute about 40% of total dose to preschoolers). The minimum concentration of B2 PAHs measured in In-CT samples for this study (8.6 μg/g) exceeds those concentrations by a factor of 5 or 9, and the median concentration (47.0 μg/g) exceeds them by a factor of 27 or 51. This indicates that, for residences with coal-tar-sealcoated parking lots or driveways, nondietary exposure to B2 PAHs might represent the most important exposure pathway for children living in those residences.

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Supporting Information Available

Results of coffee/tea test for determination of sealcoat type, summary of ancillary data, analytical methods and quality control, and table with concentrations of 16 parent PAHs in SHD and parking lot dust. This material is available free of charge via the Internet at http://pubs.acs.org.

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