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PAHs Underfoot: Contaminated Dust from Coal-Tar Sealcoated Pavement is Widespread in the United States

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We reported in 2005 that runoff from parking lots treated with coal-tar-based sealcoat was a major source of polycyclic aromatic hydrocarbons (PAHs) to streams in Austin, Texas. Here we present new data from nine U.S. cities that show nationwide patterns in concentrations of PAHs associated with sealcoat. Dust was swept from parking lots in six cities in the central and eastern U.S., where coal-tar-based sealcoat dominates use, and three cities in the western U.S., where asphalt-based sealcoat dominates use. For six central and eastern cities, median ΣPAH concentrations in dust from sealcoated and unsealcoated pavement are 2200 and 27 mg/kg, respectively. For three western cities, median ΣPAH concentrations in dust from sealcoated and unsealcoated pavement are similar and very low (2.1 and 0.8 mg/kg, respectively). Lakes in the central and eastern cities where pavement was sampled have bottom sediments with higher PAH concentrations than do those in the western cities relative to degree of urbanization. Bottom-sediment PAH assemblages are similar to those of sealcoated pavement dust regionally, implicating coal-tar-based sealcoat as a PAH source to the central and eastern lakes. Concentrations of benzo[*a*]pyrene in dust from coal-tar sealcoated pavement and adjacent soils greatly exceed generic soil screening levels, suggesting that research on human-health risk is warranted.

Introduction

Contamination of urban aquatic sediments by PAHs, which represent the largest class of suspected carcinogens (*1*), has been increasing in the United States during the last 20–40 years (*2, 3*). PAHs in the environment largely are a product of the incomplete combustion of petroleum, oil, coal, and wood (*4*). Sources in the urban environment include industrial emissions and wastes (*5*); home heating with fuel oil, wood, and coal; power plants (*6*); vehicles (*7, 8*); and pavement sealants, also known as sealcoat (*9*). In a study of PAH sources in Austin, Texas, particles in runoff from parking lots treated with coal-tar-based sealcoat had a mean total PAH concentration of 3500 mg/kg, 65 times greater than that in particles from concrete and asphalt parking lots that were not sealcoated (*9*). On the basis of comparison with suspended sediment concentrations, loads, and chemical assemblages in streams, the study concluded that sealcoat was a major source of PAHs to streams in the four watersheds studied. Recent studies have documented adverse biological effects in some Austin streams receiving runoff from coal-tar sealcoated lots (*10*), and demonstrated altered survival, growth, and development in a model amphibian species (*Xenopus laevis*) exposed to sediment spiked with coal-tar-based sealcoat (*11*).

Most sealcoat products have either a refined-coal-tar or asphalt (crude oil) base. The coal-tar varieties typically are 15–35% coal tar, a known carcinogen with extremely high concentrations of PAHs (*12*). The City of Austin reported a median concentration of the sum of 16 PAHs (dry weight basis) for coal-tar-based sealcoat products of more than 50,000 mg/kg and a median for asphalt-based sealcoat products of about 50 mg/kg (*13*). A recent informal survey on the Internet (June 5, 2008) located sealcoat applicators in all 50 U.S. states and Canada (see Supporting Information for Internet sites accessed). Although national use is not reported, the sealcoat industry estimates that in the State of Texas 225 million L of refined coal-tar-based sealcoat are applied annually ((*10*) and references therein), and the New York Academy of Sciences reported estimated annual use of coal-tar-based sealcoat in the New York harbor watershed of approximately 5.3 million L (*14*). Anecdotal reports (e.g., Web sites, blogs, commercial availability, comments by industry representatives) indicate that coal-tar-based sealcoat dominates use east of the Continental Divide (central and eastern U.S) and asphalt-based sealcoat dominates use west of the Continental Divide (western U.S.).

High concentrations of PAHs in particles washed from coal-tar sealcoated parking lots in Austin raise two questions. First, are similarly high PAH concentrations associated with sealcoated pavement in other U.S. cities? Second, does use of coal-tar-based sealcoat lead to contamination of aquatic sediments? To answer these questions, the U.S. Geological Survey (USGS) collected dust from sealcoated and unsealcoated pavement in Austin and eight other U.S. cities; samples were collected in the watersheds of lakes sampled by the USGS National Water-Quality Assessment (NAWQA) Program (Figure 1). The primary objectives were to characterize concentrations of PAHs in dust from sealcoated and unsealcoated pavement at the national scale and to evaluate PAH concentrations in lake sediments in the context of regional differences in sealcoat use. An additional objective of the study was to investigate potential off-site transport of PAHs by transport modes other than runoff. To address this

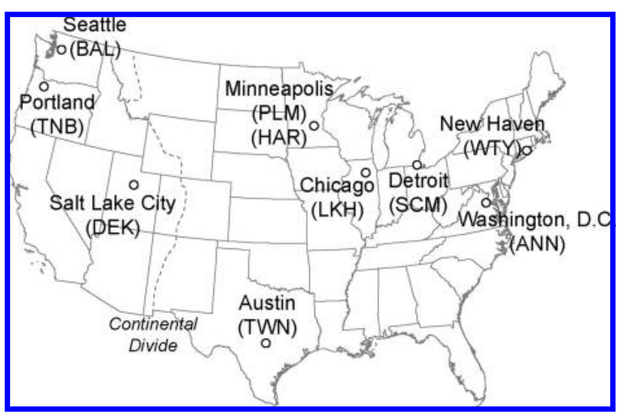

FIGURE 1. Cities where samples of pavement dust and other pavement-related solids were collected. Abbreviations (e.g., DEK) identify each watershed where dust and/or scrapings samples and lake-sediment cores were collected (Table 1).

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TABLE 1. Number of Dust Samples Collected by City and Lake Watershed

city and state	suburb, if applicable	lake watershed	NAWQA lake ID	no. of samples from sealcoated pavement	no. of samples from unsealcoated pavement
Seattle, WA	Mountlake Terrace	Lake Ballinger	BAL	9	1
Portland, OR	Beaverton	Tanasbrook Ponds	TNB	2	1
Salt Lake City, UT		Decker Lake	DEK	1[a]	1[a]
Minneapolis, MN	Brooklyn Center	Palmer Lake	PLM	1[a]	1[a]
Minneapolis, MN		Lake Harriet	HAR	1[a]	1[a]
Chicago, IL	Lake in the Hills	Lake in the Hills	LKH	7	2
Detroit, MI	Commerce	S. Commerce Lake	SCM	1[a]	1[a]
New Haven, CT		Lake Whitney	WTY	1[a]	1[a]
Washington, DC	Reston, VA	Lake Anne	ANN	1[a]	
Austin, TX		Town Lake	TWN	6	

[a] Sample is a composite of dust from three parking lots of the same type in the lake watershed indicated.

objective at a reconnaissance level, samples of soil and street dust were collected near sealcoated and unsealcoated parking lots in Lake in the Hills, Illinois, a suburb of Chicago.

Materials and Methods

Methods, quality-control results, and full data for the dust and pavement-related solids described in this paper are presented in ref *15* and briefly summarized here. Cities in this study are those where lake sediment cores were sampled by the NAWQA Program in 2004−2007 (seven cities) or previously (Lake Anne, Reston, VA, 1996; Town Lake, Austin, 1998). The design and methods for the NAWQA lakes study are presented in ref *16* and analytical methods are presented in *16* or are the same as described here and in ref *15* for dust samples. Parking lots chosen for dust sampling serve multifamily residential housing, schools, office parks, or retail businesses; none serve industrial facilities. Two residential driveways also were sampled. In five cities composite samples of dust from three parking lots of the same surface type (sealcoated or not sealcoated) were analyzed, and in four cities samples from individual lots were analyzed (Table 1). Individual lots were sampled in some cities to better understand variation in PAH concentrations among sealcoated pavements in the same area. In Austin, dust samples were collected from six individual lots, two of which were known to be sealcoated with coal-tar-based sealcoat and four with asphalt-based sealcoat. In Lake of the Hills, in addition to samples from individual parking lots, dust samples from two sealcoated driveways, dust from (unsealcoated) roads adjacent to sealcoated and unsealcoated pavement, and soil adjacent to sealcoated and unsealcoated pavement also were collected and analyzed (*15*).

Dust samples from driveways and parking lots were collected by sweeping areas of several square meters using a soft, clean, nylon brush and a clean plastic dustpan (Figure S1, Supporting Information). Areas sampled generally were in drive lanes; areas with oil staining or heavy accumulations of sediment, such as near curbs, were avoided. Brushes and dustpans were discarded after collection of each sample analyzed. Dust samples were sieved using a 0.5-mm stainless steel mesh to remove coarse sand, gravel, and debris. Details of street dust and soil sampling are presented in ref *15*. Samples were placed in clean glass jars and shipped chilled to the USGS National Water Quality Laboratory for analysis.

Samples were analyzed for PAHs using pressurized liquid extraction and gas chromatography/mass spectrometry (GC/MS) (*17*), with modifications for some samples as described in *15* and references therein. Quality control consisted of analysis of surrogate compounds added to each environmental sample and analysis of spiked samples and blank samples concurrent with analysis of each set of environmental samples. Quality-control data are presented in ref *15*. Total PAH (ΣPAH) is defined here as the sum of concentrations of 12 parent PAHs: naphthalene, acenaphthylene, acenaphthene, 9H-fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[*a*]anthracene, chrysene, benzo[*a*]pyrene, and dibenzo[*a,h*]anthracene. These are the PAHs used in the consensus-based sediment-quality-guideline probable effect concentration (PEC) (*18, 19*), with the exception of 2-methylnaphthalene, which was not quantified in this study. ΣPAH as reported here treats nondetections as zero values.

Results

Concentrations of dust from pavement in the central and eastern U.S. contrast sharply with those in the western U.S. (Figure 2). For the six central and eastern U.S. cities, a region where coal-tar-based sealcoat is reported to dominate use, the median ΣPAH concentration (computed as the median of the median value for each city) in dust from sealcoated lots was 2200 mg/kg. In contrast, the median ΣPAH concentration in dust from unsealcoated lots (collected in four of the six central and eastern cities) was 27 mg/kg, a factor of about 80 lower. This considerable difference cannot be attributed to other sources of PAHs, such as fallout of industrial emissions, exhaust particles, tire-wear residue, or leaking motor oil, because PAHs from such sources are equally likely to occur on both unsealcoated and sealcoated lots. The ΣPAH concentrations reported here are consistent with ΣPAH concentrations in particles in runoff from coal-tar sealcoated and unsealcoated lots in Austin of 3500 and 54 mg/kg, respectively (*9*). Two of the dust samples collected in the central and eastern cities were from sealcoated driveways of single-family homes in suburban Chicago. ΣPAH concentrations in these samples (5800 and 9600 mg/kg) exceeded those in all of the parking lot dust samples.

The results from the western cities tell a different story (Figure 2). Concentrations of ΣPAH from sealcoated and unsealcoated lots in the three western cities were low (13 mg/kg or less); the single exception in Seattle (one of nine sealcoated lots sampled in that city) of 850 mg/kg indicates use of coal-tar-based sealcoat on this lot. The low ΣPAH concentrations for most sealcoated lots in the western cities are consistent with reports that asphalt-based sealcoat use dominates in the western U.S.

There is substantial variability in ΣPAH concentrations in dust from sealcoated pavement within regions, with a range in concentrations in the central and eastern cities (median of each city with all lots included) of 345 to 3400 mg/kg and from 0 (nondetection of all compounds) to 5.9 mg/kg in western samples. Numerous factors likely affect variability within a region or even within a watershed, including sealcoat type, sealcoat age, climate, and parking lot characteristics such as slope and use. Nonetheless, concentrations in dust

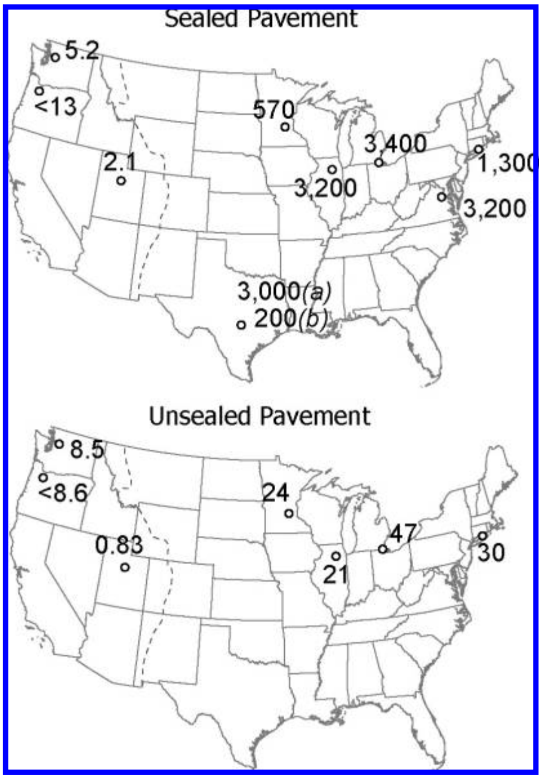

FIGURE 2. ΣPAH concentration (mg/kg) in samples of dust from sealcoated and unsealcoated pavement. Concentrations shown either are a median if multiple samples analyzed or the result for a composite sample from three pavements of the same type (see Table 1 for details). Two values are shown for Austin, TX: (a) median of two lots reportedly sealcoated with coal-tar-based sealcoat; (b) median of four lots reportedly sealcoated with asphalt-based sealcoat.

TABLE 2. ΣPAH Concentrations in Dust Swept from Pavement, in Dust Swept from Adjacent (Unsealcoated) Streets, and in Soil Adjacent to Pavement in Lake in the Hills

dust site	street dust, mg/kg	sealcoated pavement dust, mg/kg	soil, mg/kg
LKH 1		3,200	
LKH 2		2,800	
LKH 3		5,200	
LKH.PLS1	310	2,200	23
LKH.PLS4		3,200	140
LKH.PLS2	51	9,600	
LKH.PLS3	130	5,800	
		unsealcoated pavement dust, mg/kg	
LKH 4		23	
LKH.PLU1	8.0	18	10

from central and eastern sealcoated pavement are significantly (Mann−Whitney U test, $p = 0.02$) and substantially (1000×) greater than concentrations in dust from western sealcoated pavement.

For the assessment of offsite transport of PAHs, soil and street dust were collected near sealcoated parking lots and driveways and near an unsealcoated parking lot in the watershed of Lake in the Hills. None of the streets was sealcoated. Concentrations of PAHs in soil and street dust near sealcoated pavement exceeded those near unsealcoated pavement by a factor of from 6.4 to 39 (street dust) and 2.3 to 14 (soil) (Table 2).

Discussion

At the national scale, one result stands out from the sampling of pavement dust: ΣPAH concentrations in dust from sealcoated pavement in the central and eastern U.S. cities greatly exceed those in dust from unsealcoated pavement in the same cities and from sealcoated and unsealcoated pavement (with a single exception) in the western U.S. cities (Figure 2). In the central and eastern cities, the median ΣPAH concentration in dust from sealcoated pavement exceeded that from unsealcoated parking lots by a factor of about 80. In contrast, ΣPAH concentrations in dust from sealcoated and unsealcoated pavement in the three western cities are similar and about 1000 times lower than in dust from sealcoated pavement in the central and eastern cities. The elevated PAH concentrations in dust from sealcoated pavement in all six central and eastern cities where samples were collected, in contrast to the western cities where asphalt-based sealcoat dominates use, indicate that PAH-contaminated dust associated coal-tar sealcoated pavement occurs across a large part of the United States. The 1000:1 east/west ratio is comparable to the ratio of ΣPAH concentrations in refined coal-tar-based sealcoat products to those in asphalt-based sealcoat products (median ΣPAH of more than 50,000 and 50 mg/kg, respectively (*13*)). Concentrations of PAHs in dust from sealcoated and unsealcoated pavement in all of the central and eastern U.S. cities sampled are consistent with concentrations reported from Austin (*9*).

Mahler et al. (*9*) reported that particles in runoff from parking lots with coal-tar-based sealcoat might account for the majority of stream PAH loads in the Texas watersheds sampled, raising the following question: Is use of coal-tar-based sealcoat affecting water quality at a national scale? We examine this by comparing PAH concentrations in lake sediment in the watersheds where dust samples were collected, relative to urban land use, and by comparing PAH assemblages of the lake sediments to those of the dust.

PAH data from the top sample (ranging from 1 to 5 cm thickness) of a sediment core are available for lakes in the 10 watersheds for which pavement dust PAH data are presented here (Supporting Information Table S1). PAH concentrations in lake sediment have been shown to correlate strongly to percent urban land use in the watershed at the national scale (*3*). The relation to land use is not as evident for PAH in these 10 lakes because of the sparser data set, but there is a clear separation between central and eastern lakes and western lakes, with higher PAH concentrations in the central and eastern U.S. for a given amount of urban land use (Figure 3). ΣPAH in sediment at the tops of cores from three of the seven lakes in the central and eastern cities exceed the PEC, the concentration above which adverse effects to benthic biota are expected (*19*). Elevated PAH concentrations and adverse effects on benthic communities downstream from runoff from coal-tar sealcoated parking lots have been reported for some Austin streams (*10*).

PAHs comprise a large group of compounds, and PAH assemblage often is used to infer PAH sources (*20*). Differences in PAH assemblages can be investigated by computing ratios of selected PAHs; two ratios that have been identified as indicators of coal tar as a PAH source are fluoranthene/pyrene (F:P) and benzo[*a*]pyrene/benzo[*e*]pyrene (A:E) (*5, 21*). These ratios were effective for distinguishing PAH from coal-tar-based sealcoat from other combustion PAH sources in Austin (*9*). A graph of F:P versus A:E shows similarity between dust and lake sediment regionally and difference between the two regions (Figure 4). Most central and eastern dust and lake samples plot near each other and near mean values for runoff particles from coal-tar sealcoated pavement in Austin (*9*), and closer to a coal-tar standard-reference material (SRM) (*22*) than do Western dust and lake samples. Western dust

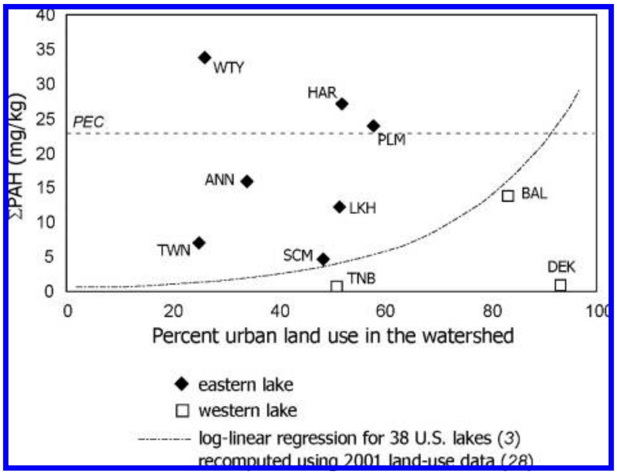

FIGURE 3. ΣPAH concentration at the tops (ranging from 1 to 5 cm depth) of lake sediment cores in watersheds where dust samples were collected, in relation to urban land use (2001 data) (*30*). Abbreviations for lakes are as shown in Figure 1 and in Table 1; PAH data are available in Supporting Information Table S-1. The dashed curve is a log-linear regression of ΣPAH versus percent urban land use for 38 lakes distributed across the U.S. (*3*), recomputed using the 2001 land-use data (*30*). Probable Effect Concentration (PEC) (*19*) is indicated by the dashed line.

and lake samples plot separately from the central and eastern samples, away from the coal-tar SRM and closer to other urban PAH source materials (*7, 23*–*25*). The similarity of the PAH assemblages of lake sediment to those of dust within each region and to different PAH source materials is additional evidence that PAH loading to lakes in the central and eastern cities includes a substantial contribution from abraded coal-tar sealcoat. In contrast, in the watersheds of the western cities, where coal-tar sealcoat use is minimal and PAH concentrations relative to urban land use are lower, the contribution of other PAH sources to lake sediment is more evident.

The elevated concentrations of PAHs in dust from sealcoated pavement in central and eastern cities cannot be attributed to urban sources of PAHs invoked in the past, such as used motor oil; burning of wood, coal, and oil; tire-wear particles; and vehicle exhaust (*6*–*8*). As all of these sources are expected to affect both sealcoated and unsealcoated pavement, they cannot explain the large difference (80×) in concentrations from sealcoated and unsealcoated pavements in the central and eastern cities. Furthermore, solely on the basis of concentration, these other urban PAH source materials cannot account for the high levels of PAHs in many of these dust samples because PAH concentrations in the dust samples exceed those in the reputed sources. Outside of coal tar and creosote (produced along with coal tar in the coking of coal), the urban source with the highest PAH concentration is used motor oil (about 600 mg/kg), followed by tire-wear particles (about 85–226 mg/kg) (*26*). Even in their pure form, undiluted by uncontaminated soil or other materials, these sources have PAH concentrations less than those measured in most dust samples swept from sealcoated pavement in the central and eastern cities (Figure 2). In essence, adding used motor oil or tire-wear particles to these dust samples would lower (dilute) the PAH concentrations. Scraping samples of coal-tar sealcoat from parking lots indicate that the dried and weathered product contains about 13,500 mg/kg PAH (median of samples from Austin (*9*) and Milwaukee (*15*), $n = 10$), and thus abraded sealcoat can account for the PAH concentrations in the dust samples even after substantial dilution by uncontaminated soil, sand, and organic debris.

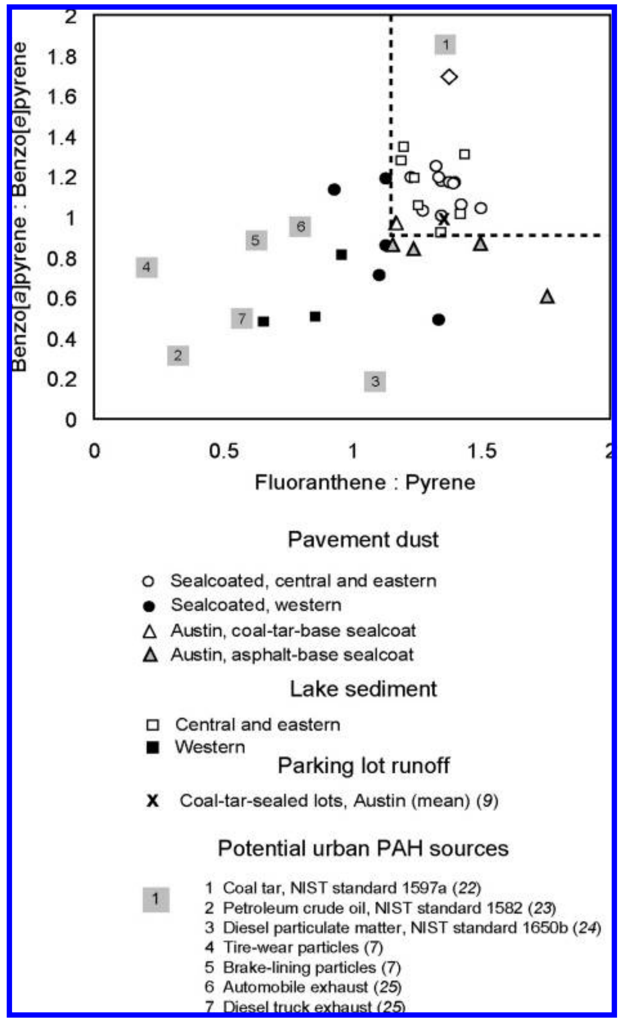

FIGURE 4. Comparison of source-indicator ratios of PAHs in dust samples, bottom sediment from lakes in the watersheds where dust was collected, and documented urban sources of PAHs. Dashed line indicates approximate separation between samples from central and eastern cities and those from western cities.

Regional differences in PAH concentrations in dust from unsealcoated pavement and the results of the soil and street dust sampling from Lake in the Hills indicate offsite transport of PAHs from coal-tar sealcoated pavement. In addition to the east–west difference in ΣPAH concentrations in dust from sealcoated pavement, there is an east–west difference (Mann–Whitney U test, $p = 0.03$) in ΣPAH concentrations in dust from unsealcoated pavement (medians of 27 and 0.83 mg/kg, respectively). We hypothesize that this difference occurs because PAH-contaminated dust from coal-tar sealcoated parking lots is being transported to unsealcoated parking lots. Additionally several of the dust samples from unsealcoated pavement in the central and eastern cities have a PAH assemblage similar to that for dust samples from sealcoated pavement in the same region (*15*), consistent with offsite transport of PAHs from coal-tar sealcoated pavement. Higher PAH concentrations in soil and street dust samples collected near sealcoated pavement in Lake in the Hills relative to concentrations in those collected near unsealcoated pavement are direct evidence of this process (Table 2). There are many ways that dust can be transported offsite in addition to runoff, including wind, snow plows, and vehicles. Visual evidence of offsite transport includes observation of fine black flecks in gutters and on sidewalks adjacent to sealcoated pavement, and dark staining and fine black flecks on unsealcoated roads at the exits from some sealcoated parking lots (Supporting Information, Figure S2a and b).

The elevated concentrations of PAHs in dust swept from coal-tar sealcoated pavement raise the question of human-health risk, particularly as use of sealcoat is not confined to commercial parking lots but includes use on playgrounds and residential driveways (Supporting Information, Figure S2c and d). Two of the dust samples analyzed for this study were collected from sealcoated driveways of single-family residences in Lake in the Hills. ΣPAH concentrations in these samples (5800 and 9600 mg/kg) exceed those in all of the other dust samples collected for this study. Concentrations of benzo[*a*]pyrene, considered the most potent carcinogen in PAH mixtures (*27*), in these samples are 597 and 357 mg/kg; the median of 477 mg/kg is more than twice the median of 201 mg/kg (computed as the median of the median value for each city) in dust from sealcoated pavement for the six central and eastern cities. The median concentration of benzo[*a*]pyrene in the two driveway samples is 5300 times greater than the benzo[*a*]pyrene generic soil screening level (SSL) of 0.09 mg/kg used by the U.S. Environmental Protection Agency Superfund Program (*28*) and is 95 times greater than a less conservative benzo[*a*]pyrene soil guideline of 5 mg/kg proposed by Fitzgerald et al. (*27*). A summary of research on mutagenic hazards of settled house dust concluded there was “substantially elevated risk” corresponding to the 95th percentile or greater PAH content in the dust as summarized from 18 studies—the 95th percentile concentration of benzo[*a*]pyrene was 13.0 mg/kg (*29*). Although pavement dust is not soil or settled house dust, there are pathways for human exposure and ingestion of pavement dust, for example by playing basketball on a sealcoated driveway. Comparison of the results from this study to these guidelines and risk assessment suggests that research is warranted on human-health risks associated with exposure to pavement sealcoated with coal-tar-based sealant.

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Supporting Information Available

Internet links to sealcoat industry Web sites demonstrating the availability of these products throughout the U.S. and in Canada, photographs of sampling and of sealcoated pavement, and PAH data for lakes used in Figures 3 and 4. This material is available free of charge via the Internet at http://pubs.acs.org.

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