Agenda Item #7
July 17, 2012

Public Hearing

MEMORANDUM

July 13, 2012

TO: County Council

FROM: Robert H. Drummer, Senior Legislative Attorney
Amanda Mihill, Legislative Attorney AMihill

SUBJECT: **Public Hearing:** Bill 21-12, Erosion, Sediment Control and Stormwater Management – Coal Tar Pavement Products

Bill 21-12, Erosion, Sediment Control and Stormwater Management – Coal Tar Pavement Products, sponsored by Councilmembers Rice, Navarro, Elrich, Riemer, and Ervin was introduced on June 19, 2012. A Transportation, Infrastructure, Energy & Environment Committee worksession is tentatively scheduled for July 26 at 2 p.m.

Bill 21-12 would prohibit the use and sale of coal tar pavement products in the County and require enforcement by the Department of Environmental Protection. Attached on ©5 is a recent study describing the problems caused by polycyclic aromatic hydrocarbons (PAHs) released into the environment through the use of a coal tar pavement product. Coal tar and coal-tar pitch are Group 1 carcinogens and the Environmental Protection Agency classifies 7 PAH compounds as probable human carcinogens.

This packet contains:	Circle #
Bill 21-12	1
Legislative Request Report	4
American Chemical Society Study	5

F:\LAW\BILLS\1221 Erosion, Sediment Control-Coal Tar\Public Hearing Memo.Doc

Bill No. 21-12
Concerning: Erosion, Sediment Control and Stormwater Management – Coal Tar Pavement Products
Revised: June 5, 2012 Draft No. 4
Introduced: June 19, 2012
Expires: December 19, 2013
Enacted:
Executive:
Effective:
Sunset Date: None
Ch. _____, Laws of Mont. Co. _________

COUNTY COUNCIL
FOR MONTGOMERY COUNTY, MARYLAND

By: Councilmembers Rice, Navarro, Elrich, Riemer, and Ervin

AN ACT to:
(1) prohibit the use and sale of coal tar pavement products in the County;
(2) require enforcement by the Director of the Department of Environmental Protection;
(3) amend the titles of Chapter 19; and
(4) generally amend the County laws regarding water quality.

By amending the titles of Chapter 19 and adding
  Montgomery County Code
  Chapter 19, Erosion, Sediment Control and Storm Water Management
  Article VI. General.
  Section 19-68

By renumbering
  Montgomery County Code
  Chapter 19, Erosion, Sediment Control and Storm Water Management
  Article VI. General.
  Sections 19-68 and 19-69

Boldface	*Heading or defined term.*
Underlining	*Added to existing law by original bill.*
[Single boldface brackets**]**	*Deleted from existing law by original bill.*
Double underlining	*Added by amendment.*
[[Double boldface brackets**]]**	*Deleted from existing law or the bill by amendment.*
* * *	*Existing law unaffected by bill.*

The County Council for Montgomery County, Maryland approves the following Act:

Sec. 1. The titles in Chapter 19 are amended, a new Section 19-68 is added, and Sections 19-68 and 19-69 are renumbered as follows:

Chapter 19, Erosion, Sediment Control and [Storm Water] Stormwater Management.

* * *

Article II. [Storm Water] Stormwater Management.

* * *

19-68. Coal tar pavement products.

(a) *Definitions.* As used in this Section:

Coal tar pavement product means a material that contains coal tar and is intended to cover an asphalt or concrete surface, including a driveway or parking area.

Director means the Director of the Department of Environmental Protection or the Director's designee.

(b) *Use of coal tar pavement products prohibited.*

(1) A person must not use a coal tar pavement product in the County.

(2) Both the property owner and the applicator have violated this Section if a coal tar pavement product is applied to an asphalt or concrete surface on the property.

(c) *Sale.* A person must not sell or offer for sale a coal tar pavement product in the County.

(d) *Enforcement.* The Director must:

(1) publish a list of alternative products for use on asphalt and concrete that do not contain coal tar; and

(2) generally enforce this Section.

[19-68] 19-69. Authority of department of environmental protection.

* * *

[19-69] <u>19-70.</u> Violations.

Any violation of this Chapter is a Class A violation. However, notwithstanding Section 1-19, the maximum penalty for a civil violation of Article I is $1,000 for an initial or repeat offense. Each day a violation continues is a separate offense.

* * *

Approved:

Roger Berliner, President, County Council Date

Approved:

Isiah Leggett, County Executive Date

This is a correct copy of Council action.

Linda M. Lauer, Clerk of the Council Date

LEGISLATIVE REQUEST REPORT

Bill 21-12
Erosion, Sediment Control and Stormwater Management – Coal Tar Pavement Products

DESCRIPTION:	Bill 21-12 would prohibit the use of coal tar pavement products in the County and require the Department of Environmental Protection to enforce this law.
PROBLEM:	Coal tar and coal-tar pitch are Group 1 carcinogens and the Environmental Protection Agency classifies 7 polycyclic aromatic hydrocarbons (PAH) compounds as probable human carcinogens. Of all known PAH sources, the highest concentrations are in coal tar and related compound creosote.
GOALS AND OBJECTIVES:	To prohibit the use of coal tar pavement products.
COORDINATION:	Department of Environmental Protection
FISCAL IMPACT:	To be requested.
ECONOMIC IMPACT:	To be requested.
EVALUATION:	To be requested.
EXPERIENCE ELSEWHERE:	To be researched.
SOURCE OF INFORMATION:	Bob Drummer, 240-777-7895
APPLICATION WITHIN MUNICIPALITIES:	To be researched.
PENALTIES:	Class A violation.

Environmental Science & Technology

Feature

pubs.acs.org/est

Coal-Tar-Based Pavement Sealcoat and PAHs: Implications for the Environment, Human Health, and Stormwater Management

Barbara J. Mahler,[†,*] Peter C. Van Metre,[†] Judy L. Crane,[‡] Alison W. Watts,[§] Mateo Scoggins,[∥] and E. Spencer Williams[⊥]

[†]U.S. Geological Survey, Austin, Texas 78754, United States
[‡]Minnesota Pollution Control Agency, St. Paul, Minnesota 55155-4194, United States
[§]University of New Hampshire, Durham, New Hampshire 03856, United States
[∥]City of Austin, Austin, Texas 78767, United States
[⊥]Baylor University, Waco, Texas 76798, United States

INTRODUCTION

Driveways and parking lots are common features of cities, suburbs, and small towns. Most single-family residences in the U.S. have paved driveways, and we encounter parking lots at multifamily residences, schools, offices, and commercial businesses. Most people in developed countries, when outdoors, probably spend as much time walking on pavement as on any other type of surface.

There are differences among paved surfaces, however. Most pavement is concrete or asphalt. The asphalt pavement of many parking lots, driveways, and even some playgrounds in North America is sprayed or painted with a black, shiny coating referred to as "sealcoat," "pavement sealant," or "driveway sealer" (Figure 1A). Sealcoat is marketed as improving pavement appearance and increasing pavement longevity.[1] In addition to making pavement black, however, one type of commonly used pavement sealcoat contains refined coal tar and is a potent source of polycyclic aromatic hydrocarbons (PAHs).[2−8] The contribution of pavement sealcoat to PAH contamination of soils, lakes, and homes has only recently been recognized.[4−6]

Coal-Tar-Based Sealcoat: A Newly Identified Source of PAHs. The two primary sealcoat product types on the market are refined coal-tar-pitch emulsion and asphalt emulsion. Coal-tar pitch, a known (Group 1) human carcinogen,[9] is the residue remaining after the distillation of crude coal tar (a byproduct of the coking of coal), and contains about 200 PAH compounds.[10] Most coal-tar-based sealcoat products consist of 20−35% coal-tar pitch as the binder. Asphalt is the residue remaining after the distillation of crude oil and is the binder in asphalt-based sealcoat products. Although the two sealcoat product types are similar in appearance, PAH concentrations in coal-tar-based sealcoat are about 1000 times higher than those in asphalt-based sealcoat[11] (Table 1).

In the U.S., coal-tar-based sealcoat is used primarily east of the Continental Divide, and asphalt-based sealcoat is used primarily west of the Continental Divide.[3] Coal-tar-based sealcoat also is used in Canada.[12] Geographic differences in use in North America likely are a historical and economic artifact of the location of most coal-tar-distillation plants near steel mills, which historically were (and are) in the central and eastern United States. An estimated 85 million gallons (320 million liters) of coal-tar-based sealcoat are used annually in the United States.[11]

The pavement sealcoat issue has been evolving since 2000, when PAH concentrations were discovered to be increasing in many urban lakes across the United States,[15] even as concentrations of other contaminants like lead, polychlorinated biphenyls (PCBs), and DDT were decreasing.[16,17] This was an apparent reversal from earlier reports that PAH concentrations in the U.S. were decreasing in response to reduced emissions from power plants and industries.[18,19] The earlier studies, however, had focused on lakes in undeveloped watersheds, whereas the upward trends in PAHs were in lakes in urban and suburban watersheds. This meant, first, that reductions in PAH emissions caused by changes in home-heating and power-generation technology had been eclipsed in urban areas by some other urban source of PAHs,[15] and second, that this other source was specific to urban and suburban areas.

A breakthrough in understanding urban sources of PAHs came in 2003, when staff with the City of Austin, TX, noted elevated PAH concentrations ($\Sigma PAH_{16} > 1000$ mg/kg) in some sediment samples collected from small tributaries and drainages in largely residential areas.[20] Concentrations of PAHs this high are typical of contaminated soils at some manufactured gas plant Superfund sites,[21] but cannot be accounted for by common urban sources (e.g., tire wear, vehicle emissions, asphalt).[2] City of Austin staff connected the dots and hypothesized that the source of the elevated PAHs was particles eroded from parking lots that were coated with coal-tar-based sealcoat.[22] Since that time, an understanding has emerged of relations between coal-tar-based pavement sealcoat and PAHs in the environment.

Published: January 24, 2012

⑥

dx.doi.org/10.1021/es203699x | Environ. Sci. Technol. 2012, 46, 3039−3045

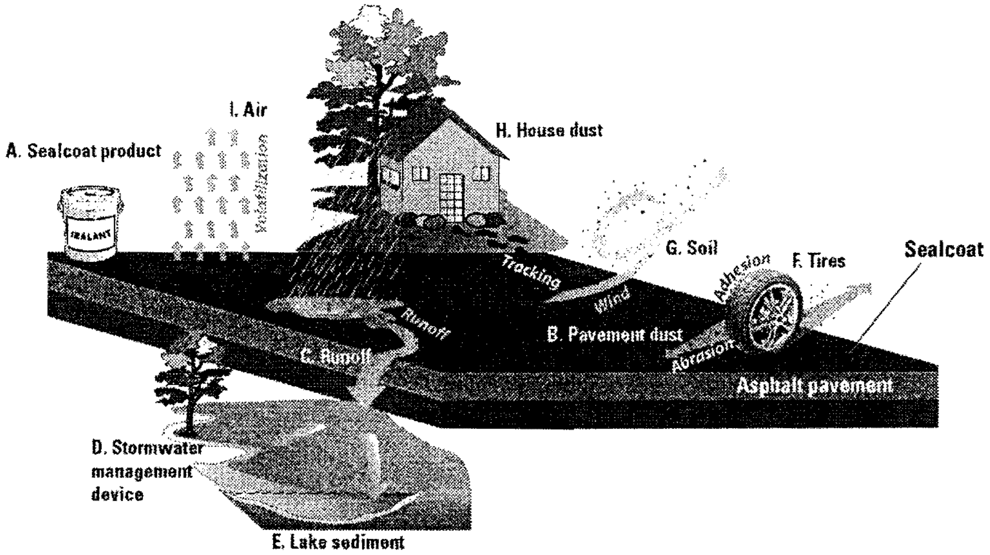

Figure 1. PAHs from coal-tar-based pavement sealcoat are transported by different pathways to various environmental compartments. Once dry, the sealcoat product (A), which contains high concentrations of PAHs, is abraded into a powder and becomes part of the dust on the pavement (B). That dust is transported by storm runoff (C) to stormwater management devices (D) or to receiving streams and lakes (E). Parking lot dust also adheres to tires (F) that track it onto unsealed pavement, and wind and runoff transport the dust to nearby soils (G). Dust particles also are tracked on shoes into residences, where they become incorporated into house dust (H). Volatile PAHs in coal-tar-based sealcoat are released into the air (I). PAH concentrations associated with each compartment and literature sources are provided in Table 1.

WHAT ARE POLYCYCLIC AROMATIC HYDROCARBONS (PAHS)? PAHs are a large group of organic compounds composed of two or more fused benzene rings arranged in various configurations. Those with a low molecular weight (two or three benzene rings) tend to be more volatile, soluble, and biodegradable than those with a higher molecular weight (four or more benzene rings). PAHs occur naturally in coal and petroleum products and are formed by the incomplete combustion of organic matter, from fossil fuels to wood to cigarettes. PAHs have many urban sources, including used motor oil, automobile exhaust, industrial atmospheric emissions, tire particles, and asphalt.[13,14] PAHs always occur as a mixture of different PAH compounds, and are ubiquitous in the urban environment. Of all known PAH sources, the highest concentrations are in coal tar and the related compound creosote. Most laboratories analyze only a subset of PAHs, and concentrations of total PAHs are reported as the sum of the subset analyzed as described in Table 1.

Migration of PAHs from Sealcoated Surfaces into the Environment. Sealcoat doesn't remain on the pavement surface indefinitely, and different applicators recommend reapplication from every 1 to 2 years (e.g., ref 23) to every 3 to 5 years (e.g., ref 24). Tires and snowplows, in particular, abrade the friable sealcoat surface into fine particles.[5,11] The overall annual loss of sealcoat from parking lots in a warm climate is about 2.4% of total sealcoat applied, with wear being most rapid (about 5% per year) in driving areas.[11] Higher wear rates have been noted in a cold-weather climate.[7] The mobilized sealcoat particles and associated PAHs are transported to various environmental compartments (Figure 1, Table 1).

The first compartment is the dust on the pavement surface itself, generated as the sealcoat is abraded from the surface (Figure 1B). Concentrations of PAHs in fine particles (dust) on pavement with coal-tar-based sealcoat are hundreds of times higher than those in dust on concrete pavement or on asphalt pavement that is unsealed or that has asphalt-based sealcoat[3−5] (Table 1). PAHs in dust on sealcoated pavement in the central and eastern U.S. are about 1000 times higher than in dust on sealcoated pavement in the western U.S., supporting anecdotal reports of geographic differences in product use[3] (Figure 2).

Stormwater runoff transports abraded sealcoat particles off sealed pavement (Figure 1C, Table 1). The PAH concentration measured in particles in runoff from parking lots with coal-tar-based sealcoat (3500 mg/kg) was 65 times higher on average than the concentration in particles in runoff from unsealed asphalt and cement lots.[2] Concentrations in unfiltered stormwater runoff from coal-tar-sealcoated pavement are particularly elevated during the months following sealcoat application. The mean ΣPAH_{16} in stormwater runoff from a coal-tar-sealcoated parking lot during the 3 months following sealcoat application was 1357 μg/L and the 3-month mean during the following two years ranged from 17 to 116 μg/L.[7] This relatively elevated concentration persists for years—the median ΣPAH_{18} in stormwater runoff from a parking lot in Madison, WI, 5 years after the last application of coal-tar-based sealcoat, was 52 μg/L.[25] That concentration is about 10 times higher than that in runoff from a mixed-use strip mall, arterial street, and unsealed parking lot (4.8−5.7 μg/L), more than 20 times higher than in runoff from a minor arterial street and a commercial rooftop (1.8−2.4 μg/L), and about 1000 times higher than in runoff from a residential feeder street (0.05 μg/L).[25]

In many communities, the first stop for stormwater runoff is a stormwater-retention pond or other stormwater-management device (Figure 1D), where suspended sediment and associated contaminants settle out. Stormwater ponds are designed to efficiently collect sediment-associated contaminants, which creates an unintended problem for many municipalities because PAHs accumulate in pond sediment. In 5 of 10 ponds sampled in the Minneapolis-St. Paul, MN, metropolitan area, concentrations

Table 1. Concentrations of PAHs as Reported in the Literature for Environmental Compartments Shown in Figure 1, and Definitions of PAH Summations Used

environmental compartment (Figure 1)	medium	PAH concentration (median or mean) in coal-tar-based sealcoat or affected medium	PAH concentration (median or mean) in asphalt sealcoat, affected medium, or associated with unsealed pavement	summation[a]	units	reference
A	sealcoat products	66 000	50	ΣPAH_{16}	mg/kg	11,22
B	pavement dust	2200	11	ΣPAH_{12}	mg/kg	3
		4760	9	ΣPAH_{16}	mg/kg	4
		685	<1	ΣPAH_{16}	mg/kg	5
C	runoff, particles	3500	54	ΣPAH_{12}	mg/kg	2
	runoff, unfiltered water[b]	71	2	ΣPAH_{16}	μg/L	7
		52	5	ΣPAH_{18}	μg/L	25
D	stormwater-management-device sediment	646	2	ΣPAH_{16}	mg/kg	5
E	lake sediment[c]	33	0.4	ΣPAH_{CMB}	mg/kg	6
F	tires	1380	3	ΣPAH_{16}	mg/kg	5
G	soil[d]	105	2	ΣPAH_{16}	mg/kg	5
H	settled house dust	129	5	ΣPAH_{16}	mg/kg	4
I	air (0.03 m from pavement), 3–8 years after sealing	1320	66	ΣPAH_8	ng/m³	28
	air (1.28 m from pavement), 3–8 years after sealing	138	26	ΣPAH_8	ng/m³	28
	air (0.03 m from pavement), 1.6 h after sealing	297 000	66	ΣPAH_8	ng/m³	29
	air (1.28 m from pavement), 1.6 h after sealing	5680	26	ΣPAH_8	ng/m³	29

[a] ΣPAH_{12} is the sum of concentrations of the 12 parent PAH (naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[*a*]anthracene, chrysene, benzo[*a*]pyrene, and dibenz[*a*,*h*]anthracene), which are those PAHs used in computation of the probable effects concentration (PEC) sediment-quality guideline,[41] less 2-methylnaphthalene. ΣPAH_{16} is the sum of the concentrations of the 16 priority pollutants identified by the U.S. Environmental Protection Agency,[42] equal to the sum of ΣPAH_{12} and concentrations of benzo[*b*]fluoranthene, benzo[*ghi*]perylene, benzo[*k*]fluoranthene, and indeno[1,2,3-*cd*]pyrene. ΣPAH_{18} is equal to ΣPAH_{16} plus concentrations of 1-methylnaphthalene and 2-methylnaphthalene. ΣPAH_{CMB} is the sum of concentrations of phenanthrene, anthracene, fluoranthene, pyrene, benz[*a*]anthracene, chrysene, benzo[*a*]pyrene, benzo[*b*]fluoranthene, benzo[*ghi*]perylene, benzo[*k*]fluoranthene, indeno[1,2,3-*cd*]pyrene, and benzo[*e*]pyrene. ΣPAH_8 is the sum of concentrations of phenanthrene, anthracene, 4,5-methylphenanthrene, 1-methylphenanthrene, fluoranthene, pyrene, chrysene, and benzo[*b*]fluoranthene. On the basis of PAH data from primarily combustion sources presented in Mahler et al.,[4] ΣPAH_{12} is about 70–75% of ΣPAH_{16}. ΣPAH_{18} is similar to ΣPAH_{16}, as the additional compounds in the summation either are not detected or are detected at very low concentrations.[2,25] [b] Collected >3 months after sealcoat application. [c] Means for urban lakes with >70% PAH from sealcoat and 0–20% from sealcoat. [d] Concentration in soil adjacent to a sealed parking lot.

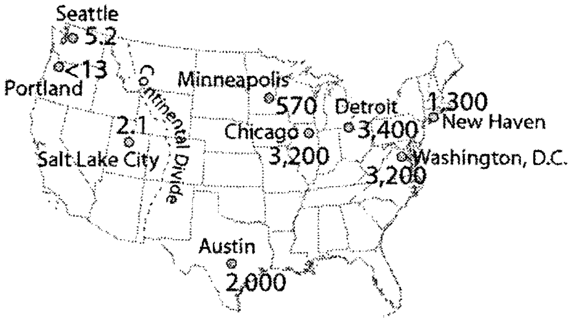

Figure 2. PAHs in dust swept from sealcoated parking lots show a striking geographic difference. PAH concentrations in dust from parking lots in central and eastern U.S. cities, where coal-tar-based sealcoat is commonly used, are about 1000 times higher than in the western U.S., where asphalt-based sealcoat is more commonly used. Concentrations are the sum of 12 PAHs (ΣPAH_{12}), in mg/kg. (Figure adapted from ref 3, Figures 1 and 2).

of PAHs in sediment exceeded Minnesota's Level 2 Soil Reference Value of 3 mg/kg benzo[*a*]pyrene equivalents (BaPeq), greatly increasing the cost for disposal.[26] Even a small amount of sealcoated pavement can be the dominant source of PAHs to sediment that collects in stormwater-management devices, as demonstrated at the University of New Hampshire Stormwater Center.[5] Sediment collected from a stormwater-management device receiving runoff from a parking lot with coal-tar-based sealcoat contained ΣPAH_{16} of 393–1180 mg/kg; sediment in devices receiving mixed runoff (4% sealed pavement and 96% unsealed pavement) contained 61–638 mg/kg ΣPAH_{16}; and sediment in a device in the center of an adjacent unsealed lot contained less than 4 mg/kg ΣPAH_{16}.[5]

Some sealcoat particles that are not trapped by stormwater ponds or other collection devices are transported down streams and rivers to lakes, where they are deposited in lake sediment (Figure 1E). Do the PAHs associated with the particles constitute a majority of PAHs in urban lake sediments, and might coal-tar-based sealcoat account for many of the upward trends in PAHs reported by Van Metre et al.?[15] An initial indication comes from a comparison of PAH ratios, or "fingerprints", of the dust collected from parking lots in nine U.S. cities to that of PAHs in sediment from lakes in the same watersheds.[3] In the central and eastern U.S., PAH fingerprints of lake sediment and dust from sealcoated parking lots were similar, and were different from fingerprints of lake sediment and dust in the western U.S., reflecting regional differences in sealcoat product type used. A more sophisticated source-apportionment method—a statistical approach that quantifies the contribution of sources with known PAH profiles to an environmental receptor—was used to quantify the contribution of identified urban PAH sources to PAHs in bed sediment in 40 U.S. urban lakes.[6] Coal-tar-based sealcoat was estimated to contribute about one-half of the PAHs in the lake sediment, when averaged across the 40 lakes; vehicle-related sources and coal combustion also were important contributors. PAH concentrations in lake sediment and the proportion contributed from coal-tar-based sealcoat were greater in the central and eastern U.S. than in the western U.S. Using sediment cores, trends in PAHs were investigated for eight urban lakes; of the six with significant upward trends, source apportionment indicated that coal-tar-based sealcoat was the cause of the trend in all six of them.

Turning our attention back to sealed pavement, dust from pavement with coal-tar-based sealcoat contaminates nearby unsealed pavement, with concentrations decreasing with distance from the sealed pavement.[5] A petrographic analysis of dust from unsealed pavement in Fort Worth, TX, found that coal-tar pitch was the dominant (92%) source of PAHs in the dust.[8] Particles are transported by adhesion to vehicle tires and by wind from sealed to unsealed surfaces—ΣPAH_{16} in particles swept from tires driven over sealed lots were 400 times higher than in particles swept from tires driven over unsealed lots[5] (Table 1, Figure 1F). Transport of abraded coal-tar-based sealcoat particles by wind and tires might be one reason why PAH concentrations in dust from unsealed parking lots in the central and eastern U.S. (median ΣPAH_{12} 27 mg/kg), where coal-tar-based sealcoat is predominantly used, are significantly higher than those in dust from unsealed parking lots in the western U.S. (median ΣPAH_{12} 0.8 mg/kg), where the asphalt-based product is predominantly used.[3]

PAHs in particles abraded from coal-tar-based sealcoat also are transported by wind, runoff, and snow removal to nearby soils (Table 1, Figure 1G). ΣPAH_{16} in surface soil adjacent to coal-tar-sealed lots at the University of New Hampshire was as high as 411 mg/kg, and concentrations decreased with distance from the sealed lot to less than 10 mg/kg.[5] The highest concentrations were measured in areas where snow was piled adjacent to the lots during the winter months—snowplows were scraping the sealcoat off with the snow. PAHs in surface soils from commercial areas in Fort Worth, TX, were dominantly (88%) from coal-tar pitch.[8]

PAHs from pavement sealed with coal-tar-based sealcoat can contaminate the indoor environment (Figure 1H) as well as the outdoor environment. In a study in Austin, TX, apartments with parking lots with coal-tar-based sealcoat had ΣPAH_{16} in house dust that was 25 times higher, on average, than ΣPAH_{16} in house dust from apartments with parking lots with other surface types (concrete, unsealed asphalt, or asphalt-based sealcoat)[4] (Table 1). The presence or absence of coal-tar-based sealcoat on the apartment complex parking lot was strongly correlated with PAH concentrations in house dust. Although tobacco smoking, candle and incense burning, and barbecue and fireplace use have been suggested to affect PAH concentrations in house dust, Mahler et al.[4] found no relation between any of these and PAH concentrations in the house dust. Concentrations of individual PAHs in house dust collected from apartments in Austin adjacent to pavement with coal-tar-sealcoated parking lots were about 140 times higher than those measured in a study of house dust in California.[27] Lower concentrations of PAHs in house dust in California are consistent with the very low concentrations of PAHs measured in pavement dust in the western U.S. (Figure 2), where coal-tar-based sealcoat is not commonly used.

In addition to contaminating stormwater, sediment, soil, and house dust, PAHs from coal-tar-based sealcoat contaminate air (Figure 1I). Several of the lower molecular weight PAHs in coal-tar-based sealcoat are volatile, which is why sealed parking lots and driveways frequently give off a strong smell. A recent study[28] reported that the flux of ΣPAH_8 from in-use parking lots with coal-tar-based sealcoat of various ages (mostly more than 3 years old) was 60 times higher than that from unsealed pavement on average. A second study[29] reported that ΣPAH_8 in air just after sealcoat application was hundreds to thousands of times higher than that above unsealed parking lots (Table 1), and that one-quarter to one-half of the PAHs in the applied sealcoat were lost to the atmosphere during the first 16 days following application. A mass balance indicated that ΣPAH_8 emissions from new applications of coal-tar-based sealant each year are larger than annual vehicle emissions of PAHs for the U.S.[29]

Biological Concerns. The detrimental effects of PAHs on terrestrial and aquatic ecosystems are well documented.[30] For example, when fish are exposed to PAHs, they exhibit chronic effects, including fin erosion, liver abnormalities, cataracts, skin tumors, and immune system impairments leading to increased susceptibility to disease.[31] When benthic macroinvertebrates—insects and other organisms that live at the bottom of rivers and lakes and that make up the base of the aquatic food chain—are exposed to PAHs, they are susceptible to a number of detrimental effects, including inhibited reproduction, delayed emergence, sediment avoidance, and mortality.[31] The most important mechanism by which acute effects occur in benthic invertebrates is a nonspecific narcosis-like mode of action that results in the degradation of cell membranes.[32] Ultraviolet (UV) radiation greatly increases the toxicity of PAHs in a wide variety of aquatic organisms.[33−36]

As the importance of coal-tar-based sealcoat as a source of PAHs has emerged, several studies have looked at potential biological effects of this particular source of PAHs. When sediment was spiked with coal-tar-based sealcoat to provide a range of environmentally relevant PAH concentrations, frogs (*Xenopus laevis*) had stunted growth or delayed development at 30 mg/kg ΣPAH_{16}, and complete mortality occurred at the highest treatment of 300 mg/kg ΣPAH_{16}.[37] Salamanders (*Ambystoma maculatum*) and newts (*Notophthalmus viridescens*) exposed to sediment contaminated with coal-tar-based sealcoat at PAH concentrations similar to the highest treatment in the frog study had stunted growth, difficulty swimming or righting themselves, and liver problems.[38,39] These effects were magnified by the addition of UV light.[38] At the community level, macroinvertebrate communities exposed to sediment spiked with coal-tar-based sealcoat had significant decreases in species abundance and richness at ΣPAH_{16} concentrations exceeding 300 mg/kg.[40] Similarly, in a study of urban streams, aquatic invertebrate communities downstream from parking lots with coal-tar-based sealcoat suffered losses of abundance and diversity along a gradient of increasing total PAH concentration, starting near the ΣPAH_{12} probable effects concentration (PEC) value of 22.8 mg/kg.[20,41] These studies demonstrate that PAHs in sediment contaminated by coal-tar-based sealcoat are bioavailable and that environmentally relevant concentrations adversely affect amphibians and benthic communities, two robust indicators of aquatic ecosystem health. The finding of adverse biological effects to biota when exposed to sediment with PAH concentrations near the PEC has widespread relevance: Of the 40 U.S. urban lakes investigated by Van Metre and Mahler,[6] sediment in the nine lakes with the greatest mass loading of PAHs from coal-tar-based sealcoat had concentrations of PAHs that exceeded the PEC.

Human-Health Concerns. Coal tar and coal-tar pitch are listed as Group 1 (carcinogenic to humans) carcinogens,[9] and the U.S. EPA currently classifies seven PAH compounds as probable human carcinogens (Group B2): benz[*a*]anthracene, benzo[*a*]pyrene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, chrysene, dibenz[*a,h*]anthracene, and indeno[1,2,3-*cd*]-pyrene.[42] Coal tar itself is a powerful mutagen: The mutagenicity index for coal tar is about 1000 times that of asphalt cements.[43] However, although coal-tar-based sealcoat has been on the market since at least 1960,[6] little has been published to date about the contribution of the sealcoat to PAH exposures and the associated potential for adverse human-health outcomes.

The elevated concentrations of PAHs in house dust, soil, air, water, and sediment associated with coal-tar-based sealcoat raise the possibility of several complete exposure pathways for humans. Incidental ingestion of house dust and soil is particularly relevant for small children, who put their hands and objects into their mouths. A recent study[44] reported that children living in homes adjacent to pavement with coal-tar-based sealcoat likely are exposed to about 14-fold higher doses of PAHs through ingestion of house dust than are children living in residences adjacent to unsealed pavement, and that exposure from ingestion of PAH-contaminated house dust is estimated to be more than double that from diet, even under conservative assumptions. Ingestion of contaminated soil is another way that children might be exposed to PAHs from coal-tar-based sealcoat, particularly given that ingestion rates of soil typically exceed those of house dust.[45] Incidental ingestion of dust directly from sealed pavement also might be important, because the extremely high concentrations of PAHs measured in these materials (Table 1) could translate to substantial doses from miniscule exposures. On a long-term basis, nondietary ingestion of PAH-contaminated house dust and soil likely are the most important routes of exposure, but a complete human-health risk analysis is required before the cancer risk associated with ingestion of these media can be quantified.

Other routes of exposure to coal-tar-based sealcoat, in addition to ingestion, might have implications for human health. Relatively high acute exposures might occur from inhalation of wind-blown particles or fumes that volatilize from sealed parking lots, especially during sealcoat application. Sealcoat applicators, in particular, might be subject to substantial inhalation exposures, but such exposures have not yet been characterized. Other potential routes include skin contact with sealcoat and abraded sealcoat particles and contaminated soil, sediment, dust, and water. Such exposures likely would be relatively infrequent and short-term. However, PAHs are readily absorbed through the skin,[46] and circumstances that increase the frequency or magnitude of exposure events, such as daily activity on pavement treated with coal-tar-based sealcoat, might be associated with increased cancer risk.

Regulatory and Retail Actions. Research to date, as documented here, provides a compelling weight-of-evidence that coal-tar-based sealcoat products are an important source of PAHs to our environment. A patchwork of actions has been taken to either ban or restrict the use of coal-tar-based sealcoat in the United States. The first ban was implemented by the City of Austin, TX, in 2006.[47] As of January 2012, 15 municipalities and two counties in four states (Minnesota, New York, Texas, and Wisconsin), the District of Columbia, and the State of Washington had enacted some type of ban, affecting nearly 10.4 million people.[48] Other local and state jurisdictions have used voluntary or limited-use restrictions for certain groups (e.g., city government) to discourage the use of coal-tar-based sealcoat.[48]

Minnesota, in particular, has been actively engaged in this issue after municipalities contacted state agencies and the Minnesota Legislature for assistance addressing PAH-contaminated stormwater pond sediment.[49] Costs for disposing of this sediment could reach $1 billion if PAHs in sediment in just 10% of the estimated 20 000 municipal stormwater ponds in the Minneapolis-St. Paul, MN, metropolitan area exceed Minnesota's Level 2 human-health risk-based Soil Reference Value of 3 mg/kg BaPeq[50] (Donald Berger, Minnesota Pollution Control Agency, written communication, 2011). The Minnesota Legislature passed a bill in 2009 that provides small grants to local governments for use in treating or disposing of contaminated sediment in stormwater ponds, provided that the governments restrict the use of undiluted coal-tar-based sealcoat.[49] As of January 2012, 13 municipalities had passed ordinances and three municipalities have received grants for remediation of stormwater ponds.

Several national and regional hardware and home-improvement retailers have voluntarily ceased selling coal-tar-based driveway-sealer products.[48] Some private applicators have chosen to use only asphalt-based sealcoat (e.g., refs 51,52). Many professional sealcoating companies in areas unaffected by bans or restrictions use coal-tar-based sealcoat, however, and coal-tar-based sealcoat products are readily available online for purchase by homeowners.

No action has been taken at a federal level to restrict the use of coal-tar-based sealcoat. Coke product residues, such as coal tar, are not classified as hazardous waste under the Resource Conservation and Recovery Act if the product is recycled.[53] This exemption allows coal-tar pitch to be used in the production of aluminum (~95% of use), commercial carbon, built-up roofing, and pavement sealcoat.[54]

Because PAHs are a ubiquitous and persistent class of urban contaminants, a decade or more might be required to assess the effectiveness of bans, restrictions, and/or changes in the retail availability of coal-tar-based sealcoat on reducing PAH concentrations in urban water bodies. Research on trends in the occurrence of PCBs and DDTs supports this concern. Following national bans on use of PCBs and DDT in the 1970s, it was 10−15 years before concentrations in lakes and reservoirs decreased by one-half.[17,55] Unlike these chemicals, all sources of PAHs in urban watersheds will not be eliminated by banning coal-tar-based sealcoat. However, reductions in PAH loads over time might be sufficient to provide more options for disposal of dredged material from stormwater ponds and navigation channels and reduce risk to terrestrial and aquatic ecosystems and human health.

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: bjmahler@usgs.gov.

Notes

The authors declare no competing financial interest.

(9)

REFERENCES

(1) Dubey, G. *Understanding how sealcoating works ... and how it can save you money*; http://pavementpro.org/understanding.htm.

(2) Mahler, B. J.; Van Metre, P. C.; Bashara, T. J.; Wilson, J. T.; Johns, D. A. Parking lot sealcoat: An unrecognized source of urban polycyclic aromatic hydrocarbons. *Environ. Sci. Technol.* **2005**, *39*, 5560−5566.

(3) Van Metre, P. C.; Mahler, B. J.; Wilson, J. T. PAHs underfoot: Contaminated dust from coal-tar sealcoated pavement is widespread in the United States. *Environ. Sci. Technol.* **2009**, *43* (1), 20−25.

(4) Mahler, B. J.; Van Metre, P. C.; Wilson, J. T.; Musgrove, M.; Burbank, T. L.; Ennis, T. E.; Bashara, T. J. Coal-tar-based parking lot sealcoat: An unrecognized source of PAH to settled house dust. *Environ. Sci. Technol.* **2010**, *44*, 894−900.

(5) *Polycyclic aromatic hydrocarbons released from sealcoated parking lots — A controlled field experiment to determine if sealcoat is a significant source of PAHs in the environment*; University of New Hampshire Stormwater Center: Durham, NH, 2010.

(6) Van Metre, P. C.; Mahler, B. J. Contribution of PAHs from coal-tar pavement sealcoat and other sources to 40 U.S. lakes. *Sci. Total Environ.* **2010**, *409*, 334−344.

(7) Watts, A. W.; Ballestero, T. P.; Roseen, R. M.; Houle, J. P. Polycyclic aromatic hydrocarbons in stormwater runoff from sealcoated pavements. *Environ. Sci. Technol.* **2010**, *44*, 8849−8854.

(8) Yang, Y.; Van Metre, P. C.; Mahler, B. J.; Wilson, J. T.; Ligouis, B.; Razzaque, M.; Schaeffer, C.; Werth, C. The influence of coal-tar sealcoat and other carbonaceous materials on polycyclic aromatic hydrocarbon loading in an urban watershed. *Environ. Sci. Technol.* **2010**, *44* (23), 1217−1233.

(9) *Monographs on the Evaluation of Carcinogenic Risks to Humans Supplement 7: Coal-Tar Pitches (Group 1)*; International Agency for Research on Cancer: Lyon, France, 1987; http://monographs.iarc.fr/ENG/Monographs/suppl7/Suppl7-57.pdf.

(10) *Toxicological profile for wood creosote, coal tar creosote, coal tar, coal tar pitch, and coal tar pitch volatiles*; U.S. Department of Health and Human Services, Agency for Toxic Substances and Disease Registry: Atlanta, GA, September 2002; http://www.atsdr.cdc.gov/ToxProfiles/tp85.pdf.

(11) Scoggins, M.; Ennis, T.; Parker, N.; Herrington, C. A photographic method for estimating wear of coal tar sealcoat from parking lots. *Environ. Sci. Technol.* **2009**, *43* (13), 4909−4914.

(12) *Reconnaissance study of coal tar sealcoat application in Toronto and an estimate of related PAH emissions*; Diamond Environmental Group, Department of Geography and Department of Chemical Engineering, University of Toronto: Toronto, ON, 2011. Prepared for Environment Canada.

(13) Rogge, W. F.; Hildemann, L. M.; Mazurek, M. A.; Cass, G. R.; Simoneit, B. R. T. Sources of fine organic aerosol. 3. Road dust, tire debris, and organometallic brake lining dust: roads as sources and sinks. *Environ. Sci. Technol.* **1993**, *27* (9), 1892−1904.

(14) Yunker, M. B.; Macdonald, R. W.; Vingarzan, R.; Mitchell, R. H.; Goyette, D.; Sylvestre, S. PAHs in the Fraser River basin: A critical appraisal of PAH ratios as indicators of PAH source and composition. *Org. Geochem.* **2002**, *33*, 489−515.

(15) Van Metre, P. C.; Mahler, B. J.; Furlong, E. T. Urban sprawl leaves its PAH signature. *Environ. Sci. Technol.* **2000**, *34*, 4064−4070.

(16) Mahler, B. J.; Van Metre, P. C.; Callender, E. Trends in metals in urban and reference lake sediments across the United States, 1970−2001. *Environ. Toxicol. Chem.* **2006**, *25* (7), 1698−1709.

(17) Van Metre, P. C.; Mahler, B. J. Trends in hydrophobic organic contaminants in urban and reference lake sediments across the United States, 1970−2001. *Environ. Sci. Technol.* **2005**, *39*, 5567−5574.

(18) Furlong, E. T.; Cessar, L. R.; Hites, R. A. Accumulation of polycyclic aromatic hydrocarbons in acid sensitive lakes. *Geochem. Cosmochim. Acta* **1987**, *51* (11), 2965−2975.

(19) Gschwend, P. M.; Hites, R. A. Fluxes of polycyclic aromatic hydrocarbons to marine and lacustrine sediments in the northeastern United States. *Geochem. Cosmochim. Acta* **1981**, *45* (12), 2359−2367.

(20) Scoggins, M.; McClintock, N.; Gosselink, L.; Bryer, P. Occurrence of polycyclic aromatic hydrocarbons below coal-tar-sealed parking lots and effects on stream benthic macroinvertebrate communities. *J. N. Am. Benthol. Soc.* **2007**, *26* (4), 694−707.

(21) Haeseler, F.; Blancher, D.; Druelle, V.; Werner, P.; Vandecasteel, J.-P. Ecotoxicological assessment of soils of former manufactured gas plant sites: Bioremediation potential and pollutant mobility. *Environ. Sci. Technol.* **1999**, *33* (24), 4379−4384.

(22) *PAHs in Austin, Texas, Sediments and Coal-Tar Based Pavement Sealants, Polycyclic Aromatic Hydrocarbons*; City of Austin Watershed Protection and Development Review Department: Austin, TX, 2005; http://www.ci.austin.tx.us/watershed/downloads/coaltar_draft_pah_study.pdf.

(23) The Blacktop Guy - FAQ. http://www.blacktopguy.com/faq.php#_13.

(24) A-Line Asphalt Products Frequently Asked Questions. http://www.alineasphaltproducts.com/faq.html.

(25) Selbig, W. R. *Concentrations of Polycyclic Aromatic Hydrocarbons (PAHs) in Urban Stormwater, Madison, Wisconsin, 2005−08*; Open File Report 2009−1077; U.S. Geological Survey: Denver, CO, 2009; http://pubs.usgs.gov/of/2009/1077/pdf/ofr20091077.pdf.

(26) Polta, R.; Balogh, S.; Craft-Reardon, A. *Characterization of stormwater pond sediments. Final project report*, EQA Report 06−572; Environmental Quality Assurance Department, Metropolitan Council Environmental Services: St. Paul, MN, 2006; http://www.metrocouncil.org/Environment/sediment/FinalReport.pdf.

(27) Whitehead, T.; Metayer, C.; Gunier, R. B.; Ward, M. H.; Nishioka, M. G.; Buffler, P.; Rappaport, S. M. Determinants of polycyclic aromatic hydrocarbon levels in house dust. *J. Expo. Sci. Env. Epid.* **2011**, *21* (March/April), 123-132.

(28) Van Metre, P. C.; Majewski, M. S.; Mahler, B. J.; Foreman, W. T.; Braun, C. L.; Wilson, J. T.; Burbank, T. Volatilization of polycyclic aromatic hydrocarbons from coal-tar-sealed pavement. Chemosphere **2012**, http://dx.doi.org/10.1016/j.chemosphere.2011.12.072.

(29) Van Metre, P. C.; Majewski, M. S.; Mahler, B. J.; Foreman, W. T.; Braun, C. L.; Wilson, J. T.; Burbank, T. PAH volatilization following application of coal-tar-based pavement sealant. *Atmos. Environ.* **2012**, http://dx.doi.org/10.1016/j.atmosenv.2012.01.036.

(30) Douben, P. E. T. *PAHs: An Ecotoxicological Perspective*; John Wiley & Sons Ltd.: West Sussex, England, 2003.

(31) Toxicological profile for polycyclic aromatic hydrocarbons; http://www.atsdr.cdc.gov/toxprofiles/tp.asp?id=122&tid=25.

(32) Van Brummelen, T. A.; et al. Bioavailability and ecotoxicity of PAHs. In *PAHs and Related Compounds. The Handbook of Environmental Chemistry*; Neilson, A.; Hutzinger, O., Eds.; Springer Verlag: Berlin, Germany, 1998; Vol. 3, part J.

(33) Ireland, D. S.; Burton, G. A. Jr.; Hess, G. G. In situ toxicity evaluations of turbidity and photoinduction of polycyclic aromatic hydrocarbons. *Environ. Toxicol. Chem.* **1996**, *15*, 574−581.

(34) Diamond, S. A.; Milroy, N. J.; Mattson, V. R.; Heinis, L. J.; Mount, D. R. Photoactivated toxicity in amphipods collected from polycyclic aromatic hydrocarbon-contaminated sites. *Environ. Toxicol. Chem.* **2003**, *22*, 2752−2760.

(35) Diamond, S. A.; Mount, D. R.; Burkhard, L. P.; Ankley, G. T.; Makynen, E. A.; Leonard, E. N. Effect of irradiance spectra on the photoinduced toxicity of three polycyclic aromatic hydrocarbons. *Environ. Toxicol. Chem.* **2000**, *19*, 1389−1396.

(36) Ankley, G. T.; et al. Assessing risks from photoactivated toxicity of PAHs to aquatic organisms. In *PAHs: An Ecotoxicological Perspective*; Douben, P. E. T., Ed.; John Wiley & Sons Ltd.: West Sussex, England, 2003; pp 275−296.

(37) Bryer, P. J.; Elliott, J. N.; Willingham, E. J. The effects of coal tar based pavement sealer on amphibian development and metamorphosis. *Ecotoxicology* **2006**, *15*, 241−247.

(38) Bommarito, T.; Sparling, D. W.; Halbrook, R. S. Toxicity of coal-tar pavement sealants and ultraviolet radiation to *Ambystoma maculatum*. *Ecotoxicology* **2010**, *19*, 1147−1156.

(39) Bommarito, T.; Sparling, D. W.; Halbrook, R. S. Toxicity of coal-tar and asphalt sealants to eastern newts, *Notophthalmus viridescens*. *Chemosphere* **2010**, *81* (2), 187–193.

(40) Bryer, P. J.; Scoggins, M.; McClintock, N. L. Coal-tar based pavement sealant toxicity to freshwater macroinvertebrates. *Environ. Pollut.* **2009**, *158* (5), 1932–1937.

(41) MacDonald, D. D.; Ingersoll, C. G.; Berger, T. A. Development and evaluation of consensus-based sediment quality guidelines for freshwater ecosystems. *Arch. Environ. Contamin. Toxicol.* **2000**, *39*, 20–31.

(42) Integrated Risk Information System (IRIS); http://cfpub.epa.gov/ncea/iris/index.cfm.

(43) Detection of coal tar materials in asphalt pavements using chemical and biological methods; http://www.asphaltinstitute.org/public/engineering/PDFs/Environmental/Detection_Coal_Tar_Materials_Asphalt_Pavements_Using_Chem_Bio_Methods.pdf.

(44) Williams, E. S.; Mahler, B. J.; Van Metre, P. C. Coal-tar pavement sealants might substantially increase children's PAH exposures. *Environ. Pollut.* **2012**, http://dx.doi.org/10.1016/j.envpol.2012.01.

(45) *Exposure Factors Handbook: 2011 ed.*; EPA/600/R-090/052F; U.S. Environmental Protection Agency: Washington, D.C., 2011; http://www.epa.gov/ncea/efh/pdfs/efh-complete.pdf.

(46) Roy, T. A.; Krueger, A. J.; Taylor, B. B.; Mauro, D. M.; Goldstein, L. S. Studies estimating the dermal bioavailability of polynuclear aromatic hydrocarbons from manufactured gas plant tar-contaminated soils. *Environ. Sci. Technol.* 1998, *32* (20), 3113–3117.

(47) *An ordinance amending the city code to add a new Chapter 6-6 relating to coal tar pavement products, creating offenses, and providing penalties*, Ordinance No. 2005117-070, 2005; http://www.ci.austin.tx.us/edims/document.cfm?id=94225.

(48) Status of actions regarding use of coal tar-based sealants; http://www.pca.state.mn.us/index.php/view-document.html?gid=16180.

(49) Crane, J. L.; Grosenheider, K.; Wilson, C. B. *Contamination of Stormwater Pond Sediments by Polycyclic Aromatic Hydrocarbons (PAHs) in Minnesota—The Role of Coal Tar-based Sealcoat Products as a Source of PAHs*; Minnesota Pollution Control Agency: St. Paul, MN, 2010, document tdr-g1-07; http://www.pca.state.mn.us/index.php/view-document.html?gid=12960.

(50) Stollenwerk, J.; Smith, J.; Ballavance, B.; Rantala, J.; Thompson, D.; McDonald, S.; Schnick, E. *Managing dredged materials in the State of Minnesota*; Minnesota Pollution Control Agency: St. Paul, MN, 2011, document wq-gen2-01; http://www.pca.state.mn.us/index.php/view-document.html?gid=12959.

(51) South GA Sealcoating and Striping Sealer performance F.A.Q.; http://www.sgasealcoating.com/sealcoating_faq.htm#5.

(52) A Better Driveway. Driveway sealcoating – now environmentally friendly; http://www.abetterdriveway.com/residential_services.

(53) Hazardous Waste Recycling Exemptions; http://www.epa.gov/osw/hazard/recycling/regulations.htm.

(54) Valle, S.; Panero, M. A.; Shor, L. *Pollution prevention and management strategies for polycyclic aromatic hydrocarbons in the New York/New Jersey Harbor*; Harbor Consortium of the New York Academy of Sciences: New York, NY, 2007.

(55) Van Metre, P. C.; Wilson, J. T.; Callender, E.; Fuller, C. C. Similar rates of decrease of persistent, hydrophobic contaminants in riverine systems. *Environ. Sci. Technol.* **1998**, *32* (21), 3312–3317.

(11)