Agenda Item 5
September 11, 2012
Action

September 7, 2012

MEMORANDUM

TO: County Council

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

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

Transportation, Infrastructure, Energy & Environment Committee recommendation (3-0): enact Bill 21-12.

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 public hearing was held on July 17, at which speakers testified in support and opposition to Bill 21-12 (see select written correspondence at ©39-54). A Transportation, Infrastructure, Energy and Environment Committee worksession was held on July 26.

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 ©10 is a recent study describing the problems caused by polycyclic aromatic hydrocarbons (PAH's) 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.

Background

What are coal tar, coal tar sealants, and polycyclic aromatic hydrocarbons (PAH)? *Coal tar* is a byproduct of the coking of coal for the steel industry and coal-tar pitch is the residue that remains after the distillation of coal tar. Coal-tar pitch is at least 50% PAH's by weight and is known to cause cancer in humans.

Pavement sealant is a liquid that is sprayed or painted on some asphalt pavement to protect the pavement surface. Sealcoat products generally have a coal-tar-pitch or asphalt base. Coal-tar-based sealcoat products are usually 20-35% coal-tar pitch and have about 1,000 times more PAH's than sealcoat products with an asphalt base.

Polycyclic aromatic hydrocarbons (PAH's) are a group of chemical compounds that form when anything with a carbon base is burned (e.g., wood, gasoline, cigarettes, meat). PAH's are also in materials (tires, coal tar).[1] Of all known PAH sources, the highest concentrations are in coal tar and the related compound creosote.[2]

How do PAH's get released into the environment? As the U.S. Geological Survey and the American Chemical Society explain, sealcoat does not stay on a pavement permanently; sealcoat manufacturers recommend reapplication every 1-2 or 3-5 years, depending on the product used. Vehicle tires erode the sealcoat and it breaks into small particles. These particles can be washed off the roadway by rain and carried through storm drains into streams, ponds, and stormwater management devices. Other particles can be blown offsite by wind or tracked indoors on the soles of shoes.[3]

What are the environmental and health concerns with coal tar and PAH's? Some PAH's are toxic to fish, amphibians, and plants. Studies have looked at the effects of coal-tar-based sealcoat. When sediment was spiked with coal-tar-based sealcoat, frogs had stunted growth, delayed development, and mortality; salamanders had stunted growth, difficulty swimming or righting themselves, and liver problems. As noted above, coal tar and coal-tar pitch are Group 1 carcinogens and the Environmental Protection Agency classifies 7 PAH compounds as probable human carcinogens.[4]

At the public hearing, Bob Hoyt, Director of the Department of Environmental Protection, endorsed Bill 21-12 on behalf of the County Executive. Mr. Hoyt noted that DEP recently sampled the sediment from Lake Whetstone and Gunners Lake and found that the levels of PAH's in the sediment exceeded the State's standard for restoring contaminated properties for residential use. Mr. Hoyt noted that the PAH's in the sediment increase the costs of managing the sediment after it is dredged by an estimated $1,120,000 for those two lakes alone (©39-40).

At a June 29 meeting of the Council sitting as the Board of Health, Councilmember Ervin asked the County Health Officer to comment on the health effects of coal tar products. Dr. Tillman, County Health Officer, forwarded studies by EPA and the Centers for Disease Control/Agency for Toxic Substances (see cover pages and summary on ©55-59), but declined to offer an "informed opinion" herself and deferred to DEP's expertise on water quality issues.

How does the USGS link PAH's in the environment to coal tar sealants? As described on ©17, the USGS collected sediment cores from 40 lakes across the country and analyzed the samples for PAH's. USGS then determined the contribution of PAH's from different sources by using a chemical mass-balance model, which is based on chemical "fingerprinting". According

[1] *Coal-Tar-Based Pavement Sealcoat, Polycyclic Aromatic Hydrocarbons (PAH's), and Environmental Health.* U.S. Geological Survey. Available at: http://pubs.usgs.gov/fs/2011/3010/pdf/fs2011-3010.pdf (©17-22).
[2] *Coal-Tar-Based Pavement Sealcoat and PAH's: Implications for the Environment, Human Health, and Stormwater Management*, American Chemical Society. Available at: http://pubs.acs.org/doi/pdfplus/10.1021/es203699x (©10-16).
[3] http://pubs.usgs.gov/fs/2011/3010/pdf/fs2011-3010.pdf; http://pubs.acs.org/doi/pdfplus/10.1021/es203699x.
[4] http://pubs.acs.org/doi/pdfplus/10.1021/es203699x.

to the USGS analysis, on average coal-tar-based sealcoat accounts for half of all PAH's in the lakes.

Presenting another viewpoint is a study on ©25. This study, funded by an industry organization, the Pavement Coatings Technology Council (PCTC), concluded that "sediments' PAH profiles are no more similar to refined tar sealers than they are to a number of other environmental inputs. While refined tar sealers were not eliminated as a potential source in some locations, forensic methods did not differentiate their contribution from other sources of PAH's, indicating refined tar sealers are not a unique or readily quantifiable source of PAH's to the urban environment." On its website, the Minnesota Pollution Control Agency noted data-quality problems with this study's environmental forensic analysis and concluded that the study made inaccurate comparisons between sediment data and stormwater runoff data in its analyses.[5]

Are there alternatives to the use of coal tar sealants? As the Environmental Protection Agency noted, there are available alternatives to the use of coal-tar-based sealants (©23). These alternatives include asphalt-based sealers, which contain 0.03% to 0.66% PAH's dry weight (compared to 3.4% to 20% PAH's dry weight for coal-tar-based sealants). Additional alternatives are permeable asphalt, gravel, and concrete.

What have other jurisdictions done? According to the blog, *Coal Tar Free America*,[6] several jurisdictions have restricted or banned the use of coal tar sealants (©37). Notable examples of jurisdictions that have banned the product include Austin, Texas; Suffolk County, New York; Washington, D.C.; and Washington State. Additionally, according to various sources, the Home Depot and Lowes stores throughout the U.S. have stopped selling these products.

Issues/Committee Recommendation

1. *Should the use of coal tar sealants be banned?* The opposition to Bill 21-12 has chiefly come from individuals and companies in the industry who are concerned about the economic effect of a potential ban (see testimony, ©39). The County Finance Department indicated that they do not expect Bill 21-12 to have an economic impact (©9). The manufactures of sealcoat products that contain coal tar may experience negative economic impact if private contractors in the County stop using those products.

A 2007 report by the Washington State Department of Transportation included the following data regarding costs of sealcoat:

Product	Cost per Gallon	Performance History
Coal Tar Pitch	$3.00	8-10 years
Coal Tar Blend	$1.80	6-8 years
Asphalt Emulsion	$1.65	4-6 years

[5] http://www.pca.state.mn.us/index.php/water/water-types-and-programs/stormwater/municipal-stormwater/coal-tar-based-sealcoat-minnesota-local-government-faqs.html

[6] The blog is at: http://coaltarfreeamerica.blogspot.com/. The blog is written by Thomas Ennis, Sustainability Officer at the City of Austin, Texas.

Councilmembers should weigh the environmental and health benefits of banning coal tar products against the economic impacts that the ban could have. Committee members were persuaded by the information about environmental effects provided by the US Geological Survey and the American Chemical Society. **Committee recommendation (3-0):** enact Bill 21-12.

2. ***Should the ban be limited to coal tar emulsion products?*** Some companies suggested that refined coal tar should not be subject to the coal tar ban because refined coal tar is different than crude coal tar. The USGS fact sheet on ©22 notes that coal-tar pitch is "refined" into 12 different viscosities, including RT-12, which is used in coal-tar-based sealcoats. Council staff has not found any evidence that "refined coal tar" should be excluded from the scope of this Bill.

The Council received correspondence from the manufacturer of PaveRx, seeking an amendment to the proposed ban to prohibit only coal-tar emulsion sealers. The manufacturer argues that the properties of PaveRx are different because it made of refined coal tar and doesn't chip, flake, dust, peel, or spall and therefore doesn't raise the same environmental issues. However, the manufacturer states that PaveRx must be reapplied every 5-7 years.[7] In Council staff's understanding, this essentially means that the product will still be eroded over time by vehicle tires and other means. In other words, the product may last longer than regular coal-tar-based sealcoat but will still contribute to PAH's being released into the environment.

The Committee did not recommend excluding this product from Bill 21-12. Councilmembers could consider whether to allow a waiver from the coal tar ban **if** the applicant for the waiver can convince DEP that their product does not release PAH's into the environment. If Councilmembers want to allow such a waiver, Council staff recommends inserting the following new subsection (d):

> <u>(d)</u> <u>The Director may waive the prohibitions of subsections (b) and (c) for a product if the applicant for a waiver shows that ordinary use of the product does not result in the immediate or eventual release of measurable quantities of polycyclic aromatic hydrocarbons into the air, water, ground, or sediments.</u>

The Committee did not discuss this language. After the worksession, Mike Leaman, President of Total Asphalt, submitted a letter requesting that Bill 21-12 be amended to include this language (©67).

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[7] The website of another company, HASCO Inc, indicates that PaveRx should be reapplied every 4-5 years. See http://www.hasco2000.com/html/property_maintenance.html.

This packet contains:

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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.

BILL 21-12

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ROCKVILLE, MARYLAND

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MEMORANDUM

July 11, 2012

TO: Roger Berliner, President, County Council

FROM: Jennifer A. Hughes, Director, Office of Management and Budget
Joseph F. Beach, Director, Department of Finance

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

Attached please find the fiscal and economic impact statements for the above-referenced legislation.

JAH:nm

c: Kathleen Boucher, Assistant Chief Administrative Officer
Lisa Austin, Offices of the County Executive
Joy Nurmi, Special Assistant to the County Executive
Patrick Lacefield, Director, Public Information Office
Alex Espinosa, Office of Management and Budget
Stan Edwards, Department of Environmental Protection
Greg Ossont, Department of General Services
Patricia Brennan, Department of Health and Human Services
Angela Dizelos, Office of Management and Budget
Naeem Mia, Office of Management and Budget
David Platt, Department of Finance

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

1. **Legislative Summary**
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 (DEP). Coal tar and coal-tar pitch are Group 1 carcinogens and the Environmental Protection Agency classifies seven (7) polycyclic aromatic hydrocarbons (PAH) compounds as probable human carcinogens. Of all known PAH sources, the highest concentrations (and greatest danger to human life) are in coal tar and related compounds.

2. **An estimate of changes in County revenues and expenditures regardless of whether the revenues or expenditures are assumed in the recommended or approved budget. Includes source of information, assumptions, and methodologies used.**
Alternatives to coal tar-based sealants are readily available on the market. Neither the Department of General Services (DGS) nor the Department of Transportation (DOT) use coal tar-based sealant products for any County activities.

Based on comparable bans of coal tar-based sealants in Washington, DC and Austin, TX, DEP estimates approximately 10 or less enforcement actions per year. Assuming ten (10) enforcement actions per year and lab testing services at a cost of $400 per action, the estimated fiscal impact to expenditures is $4,000 per year, in the first year of the ban. This number of enforcement actions is not anticipated to result in any staffing impact to DEP.

DEP cannot currently estimate a fiscal impact to expenditures resulting from outreach efforts to advertise the ban. However, DC has a similar ban and has previously contacted regional distributors, home improvement and hardware stores, trade associations, contractors, and utility companies (including PEPCO and Washington Gas); many of these parties are believed to have taken steps to find alternative products. Therefore, DEP believes that the County's outreach efforts should be relatively easier.

Section 19-70 of the proposed bill authorizes a *maximum* penalty of $1,000 for an initial or repeat offense. However, DEP does not anticipate that all violations will result in a citation; some violations may only carry a notice or warning (which does not carry a monetary penalty). Based on a conservative estimate, DEP estimates a total of $9,000 in revenues from penalties (see chart directly below).

Year	Revenues	Number of enforcement actions and citations*
1	$5,000	10 enforcement actions, 5 of which result in citations
2	$3,000	3 citations
3	$1,000	1 citation
4	$0	Coal tar products are no longer on the market
5	$0	Coal tar products are no longer on the market
6	$0	Coal tar products are no longer on the market
Total revenues	*$9,000*	*9 citations over 6-years at max. penalty*

* Assumes each citation is at the maximum penalty of $1,000.

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3. **Revenue and expenditure estimates covering at least the next 6 fiscal years.**
Revenues over the next 6 fiscal years are estimated to be $9,000 (see chart in item #2 above).

The number of enforcement actions is expected to decrease over subsequent fiscal years as users switch to alternative products. The total impact to expenditures over the next six fiscal years is $5,600 (see chart directly below).

Year	Expenditures	Number of enforcement actions and testing*
1	$4,000	10 enforcement actions/testing
2	$1,200	3 enforcement actions/testing
3	$400	1 enforcement actions/testing
4	$0	Coal tar products are no longer on the market
5	$0	Coal tar products are no longer on the market
6	$0	Coal tar products are no longer on the market
Expenditures	*$5,600*	*14 enforcement actions over 6-years at max. penalty*

* Assumes that all enforcement actions involve testing at cost of $400 per test. However, not all enforcement actions are likely to result in citations.

Although the cost of outreach cannot be estimated at this time, the fiscal impact of outreach is expected to be limited to the first year of the bill's implementation.

4. **An actuarial analysis through the entire amortization period for each bill that would affect retiree pension or group insurance costs.**
Not applicable.

5. **Later actions that may affect future revenue and expenditures if the bill authorizes future spending.**
The bill does not authorize future spending.

6. **An estimate of the staff time needed to implement this bill.**
Additional staff time required by this bill cannot be estimated, but DEP will utilize existing staff resources to absorb the additional workload.

7. **An explanation of how the addition of new staff responsibilities would affect other duties.**
DEP will utilize existing staff resources to absorb the additional workload.

8. **An estimate of costs when an additional appropriation is needed.**
Additional appropriations are not needed.

9. **A description of any variable that could affect revenue and cost estimates.**
Variables that could affect cost estimates include the cost and scope of outreach, which cannot be estimated at this time. The number of enforcement actions in any given year is also subject to wide variability.

10. Ranges of revenue or expenditures that are uncertain or difficult to project.
Although the bill allows for penalties of up to $1,000 per violation (per day), actual penalties may be lower. Furthermore, not all enforcement actions result in citations. In addition, the cost of outreach cannot be estimated at this time.

11. If a bill is likely to have no fiscal impact, why that is the case.
Not applicable.

12. Other fiscal impacts or comments.
Assumptions and estimates regarding revenues and expenditures are approximate only. DEP cannot estimate with certainty the number of enforcement actions performed in a given year and the number of citations issued.

Both the Department of General Services (DGS) and the Department Health and Human Services (HHS) report no fiscal impacts to their budgets resulting from this bill.

13. The following contributed to and concurred with this analysis:
Stan Edwards, Department of Environmental Protection
Greg Ossont, Department of General Services
Patricia Brennan, Department of Health and Human Services
Angela Dizelos, Office of Management and Budget
Naeem Mia, Office of Management and Budget

Jennifer A. Hughes
Jennifer A. Hughes, Director
Office of Management and Budget

7/11/12
Date

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

Background:

This proposed legislation would prohibit the use and sale of coal tar pavement products in the County and require enforcement by the Department of Environmental Protection. Coal tar and coal-tar pitch are Group 1 carcinogens. Currently, the use of these products is prohibited in the District of Columbia.

1. The sources of information, assumptions, and methodologies used.

Based on information provided to the Department of Finance (Finance), major retailers such as The Home Depot do not sell coal tar pavement products in the County. It is not known if private contractors currently use these products in the County, however, both the Department of Transportation and the Department of Permitting Services believe that the price difference between coal tar pavement products and those that do not include coal tar is not significant enough to have an economic impact on that sector.

2. A description of any variable that could affect the economic impact estimates.

The price of coal tar pavement products vis a vis those manufactured without coal tar is the only variable.

3. The Bill's positive or negative effect, if any on employment, spending, saving, investment, incomes, and property values in the County.

There is likely no economic effect at all, since the price of these products (coal tar and non-coal tar) is not materially different.

4. If a Bill is likely to have no economic impact, why is that the case?

See #3 above.

5. The following contributed to and concurred with this analysis: David Platt and Mike Coveyou, Finance.

Joseph F. Beach, Director
Department of Finance

7/5/12
Date

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[⊥]

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■ 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 (ΣPAH_{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

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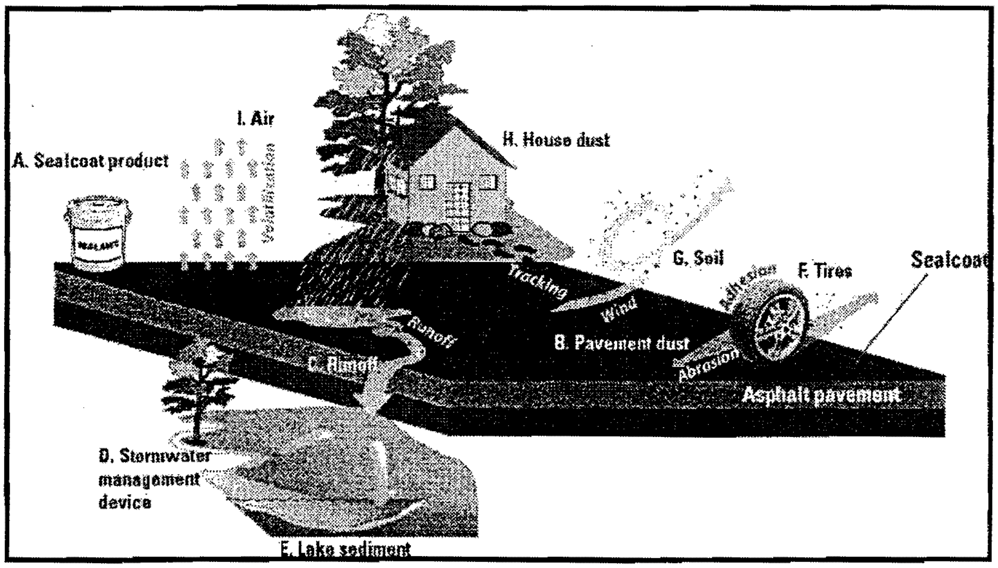

Figure 1. PAHs from coal-tar-based pavement sealcoat are transported by different pathways to various environmental compartments. Once dry, the sealcoat product (A), which contains high concentrations of PAHs, is abraded into a powder and becomes part of the dust on the pavement (B). That dust is transported 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 $\mu g/L$ and the 3-month mean during the following two years ranged from 17 to 116 $\mu g/L$.[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 $\mu g/L$.[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 $\mu g/L$), more than 20 times higher than in runoff from a minor arterial street and a commercial rooftop (1.8−2.4 $\mu g/L$), and about 1000 times higher than in runoff from a residential feeder street (0.05 $\mu g/L$).[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

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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/m^3	28
	air (1.28 m from pavement), 3–8 years after sealing	138	26	ΣPAH_{8}	ng/m^3	28
	air (0.03 m from pavement), 1.6 h after sealing	297 000	66	ΣPAH_{8}	ng/m^3	29
	air (1.28 m from pavement), 1.6 h after sealing	5680	26	ΣPAH_{8}	ng/m^3	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.

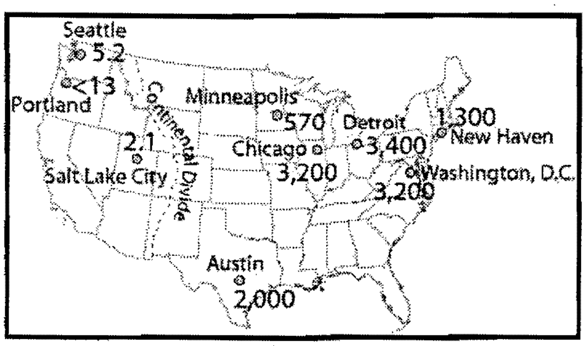

Figure 2. PAHs in dust swept from sealcoated parking lots show a striking geographic difference. PAH concentrations in dust from parking lots in central and eastern U.S. cities, where coal-tar-based sealcoat is commonly used, are about 1000 times higher than in the western U.S., where asphalt-based sealcoat 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

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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

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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.

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Notes

The authors declare no competing financial interest.

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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 seal-coated 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.

15

(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. 20051117-070, 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.

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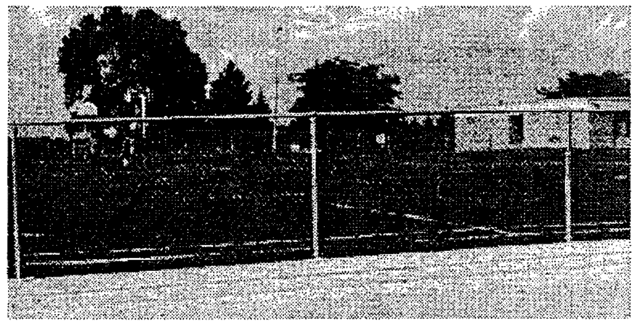
USGS
science for a changing world

Coal-Tar-Based Pavement Sealcoat, Polycyclic Aromatic Hydrocarbons (PAHs), and Environmental Health

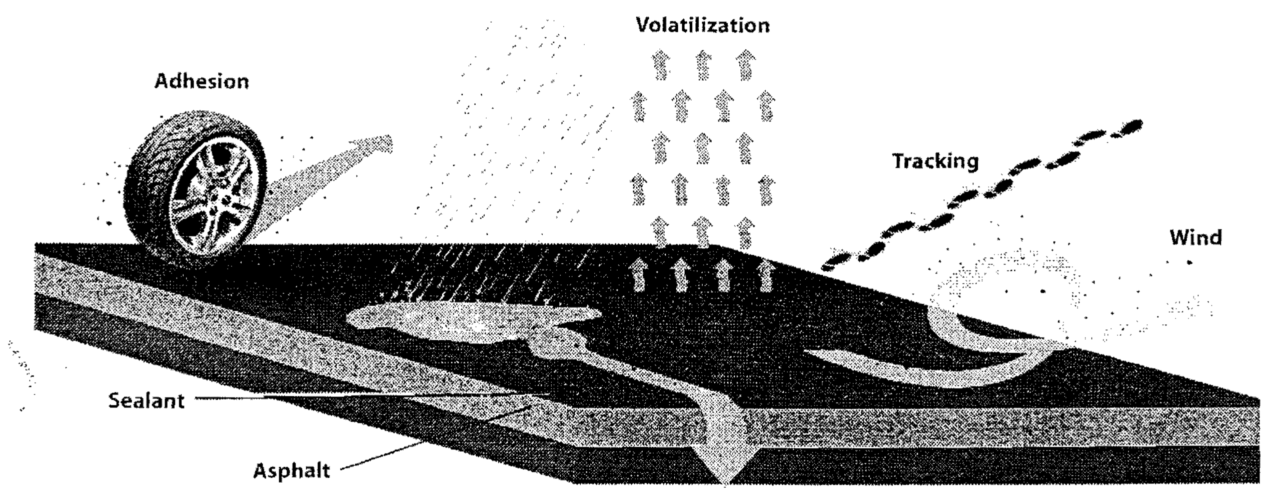
Studies by the U.S. Geological Survey (USGS) have identified coal-tar-based sealcoat—the black, viscous liquid sprayed or painted on asphalt pavement such as parking lots—as a major source of polycyclic aromatic hydrocarbon (PAH) contamination in urban areas for large parts of the Nation. Several PAHs are suspected human carcinogens and are toxic to aquatic life.



Sealcoat is the black, viscous liquid sprayed or painted on the asphalt pavement of many parking lots, driveways, and playgrounds.

Key Findings

- Dust from pavement with coal-tar-based sealcoat has greatly elevated PAH concentrations compared to dust from unsealed pavement.
- Coal-tar-based sealcoat is the largest source of PAH contamination to 40 urban lakes studied, accounting for one-half of all PAH inputs.
- Coal-tar-based sealcoat use is the primary cause of upward trends in PAHs, since the 1960s, in urban lake sediment.
- Residences adjacent to parking lots with coal-tar-based sealcoat have PAH concentrations in house dust that are 25 times higher than those in house dust in residences adjacent to parking lots without coal-tar-based sealcoat.
- PAHs move from a sealcoated surface into our environment by many mechanisms: storm runoff, adhesion to tires, wind, foot traffic, and volatilization.



Original graphic courtesy of Aaron Hicks, City of Austin, Texas.

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What are Sealcoat, PAHs, and Coal Tar?

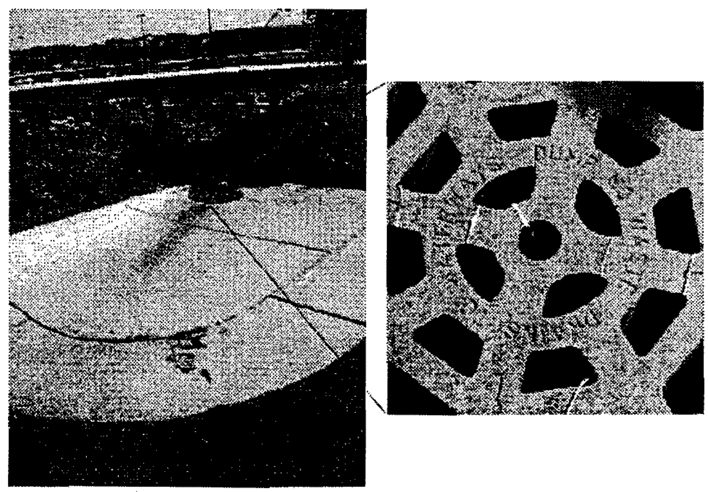
Pavement sealcoat (also called sealant) is a black liquid that is sprayed or painted on some asphalt pavement. It is marketed as protecting and beautifying the underlying pavement, and is used commercially and by homeowners across the Nation. It is applied to parking lots associated with commercial businesses, apartment and condominium complexes, churches, schools, and business parks, to residential driveways, and even to some playgrounds. Most sealcoat products have a coal-tar-pitch or asphalt (oil) base. Coal-tar-based sealcoat is commonly used in the central, southern, and eastern United States, and asphalt-based sealcoat is commonly used in the western United States.

PAHs are a group of chemical compounds that form whenever anything with a carbon base is burned, from wood and gasoline to cigarettes and meat. PAHs also are in objects and materials, such as automobile tires and coal tar, the production of which involves the heating of carbon-based materials. PAHs are of environmental concern because several are toxic, carcinogenic, mutagenic, and/or teratogenic (causing birth defects) to aquatic life, and seven are probable human carcinogens (U.S. Environmental Protection Agency, 2009).

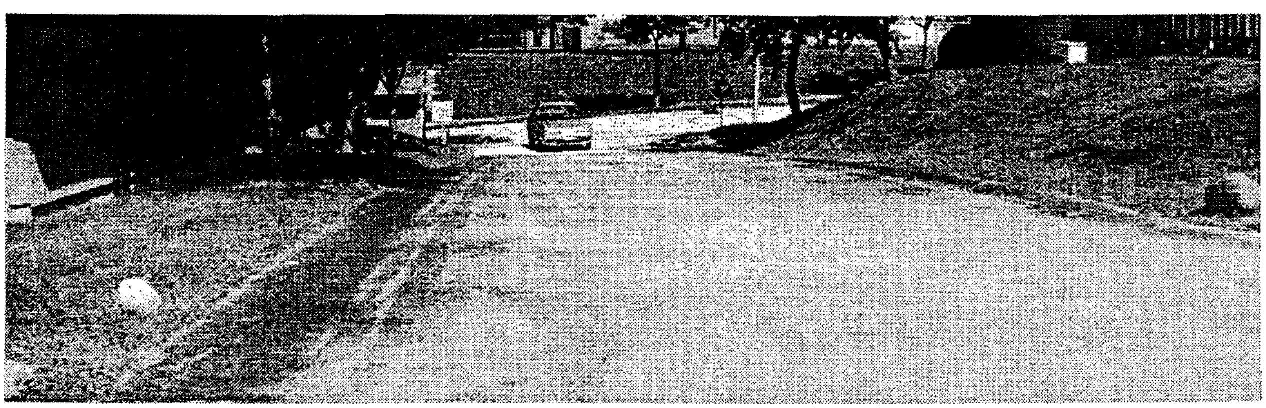
Coal tar is a byproduct of the coking of coal for the steel industry and coal-tar pitch is the residue remaining after the distillation of coal tar. Coal-tar pitch is 50 percent or more PAHs by weight and is known to cause cancer in humans (International Agency for Research on Cancer, 1980). Coal-tar-based sealcoat products typically are 20 to 35 percent coal-tar pitch. Product analyses indicate that coal-tar-based sealcoat products contain about 1,000 times more PAHs than sealcoat products with an asphalt base (City of Austin, 2005).

How does Sealcoat get from Driveways and Parking Lots into Streams and Lakes, Homes, and the Air?

Friction from vehicle tires abrades pavement sealcoat into small particles. These particles are washed off pavement by rain and carried down storm drains and into streams. Other sealcoat particles adhere to vehicle tires and are transported to other surfaces, blown offsite by wind, or tracked indoors on the soles of shoes. Some of the PAHs in sealcoat volatilize (evaporate), which is why sealed parking lots and driveways frequently give off a "mothball" smell. Sealcoat wear is visible in high traffic areas within a few months after application, and sealcoat manufacturers recommend reapplication every 2 to 4 years.



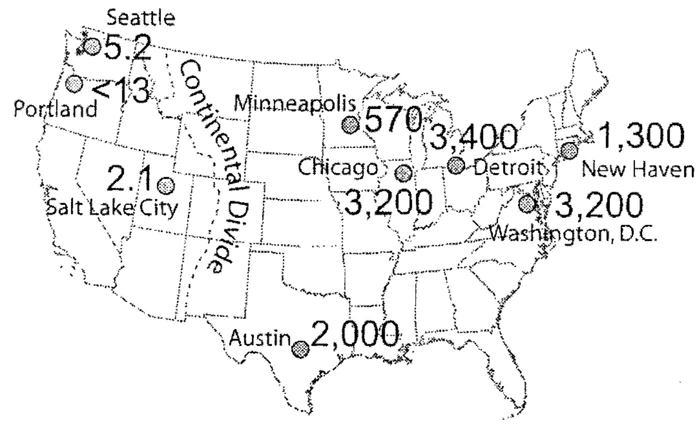
Runoff from sealcoated pavement (black surface) enters storm drains that lead to local streams. Drain grate (inset) is marked "DUMP NO WASTE" and "DRAINS TO WATERWAYS."



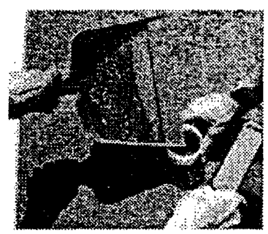
Gray asphalt pavement shows through where sealcoat has worn off the driveway of an apartment complex.

The East-West Divide
Regional Product Use Translates to Large Differences in PAH Concentrations

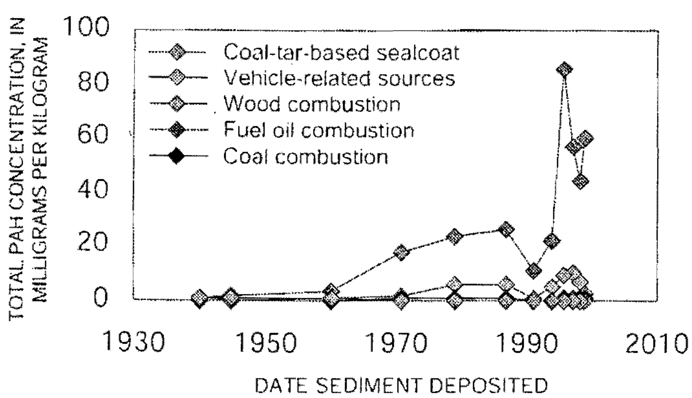
Does product type really matter? PAH concentrations in the coal-tar-based sealcoat product are about 1,000 times higher than in the asphalt-based product (more than 50,000 milligrams per kilogram [mg/kg] in coal-tar-based products and 50 mg/kg in asphalt-based products [City of Austin, 2005]). Anecdotal reports, such as Web sites, blogs, and comments by industry representatives, indicate that the coal-tar-based product is used predominantly east of the Continental Divide and the asphalt-based product is used predominantly west of the Continental Divide. During 2007–08, the USGS swept dust from sealcoated and unsealcoated parking lots in nine cities across the United States and analyzed the dust for PAHs. For six cities in the central and eastern United States, the median PAH concentration in dust from sealcoated parking lots was 2,200 mg/kg, about 1,000 times higher than in dust from sealcoated parking lots in the western United States, where the median concentration was 2.1 mg/kg. Although both product types are available nationally, these results confirm the regional difference in use patterns (Van Metre and others, 2009).



Concentrations of PAHs in dust swept from sealed parking lots in central and eastern U.S. cities, where coal-tar-based-sealcoat use dominates, were about 1,000 times higher than in western U.S. cities, where asphalt-based-sealcoat use dominates. Concentrations shown on the map are the sum of 12 PAHs, in milligrams per kilogram (Van Metre and others, 2009).



"Fingerprinting" Shows that Coal-Tar Sealant is the Largest Source of PAHs to Urban Lakes

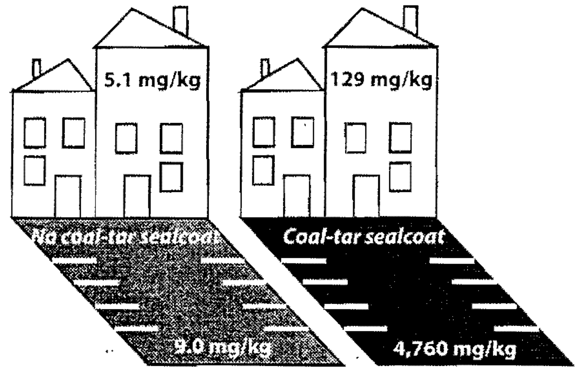


Coal-tar-based sealcoat (orange symbol) is the largest contributor to increasing concentrations of PAHs in Lake Killarney, Orlando, Florida, as determined by chemical fingerprinting. Similar patterns were seen in lakes across the central and eastern United States (Van Metre and Mahler, 2010).

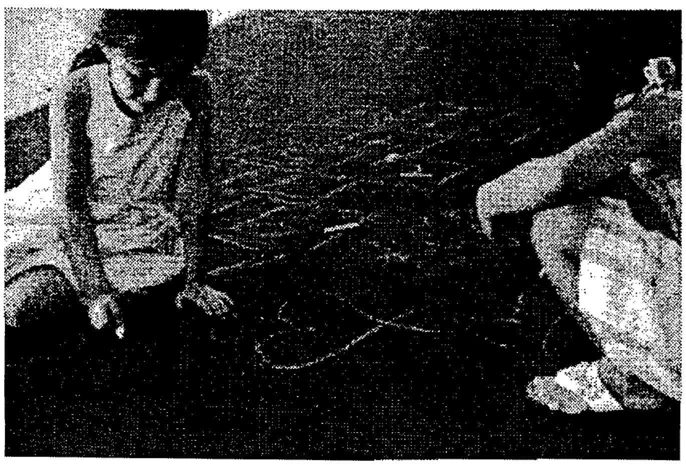
PAHs are increasing in urban lakes across the United States. To better understand why this might be happening, USGS scientists collected sediment cores from 40 lakes in cities from Anchorage, Alaska, to Orlando, Florida, analyzed the cores for PAHs, and determined the contribution of PAHs from many different sources by using a chemical mass-balance model. The model is based on differences in the chemical "fingerprint" of PAHs from each source. Coal-tar-based sealcoat accounted for one-half of all PAHs in the lakes, on average, while vehicle-related sources accounted for about one-fourth. Lakes with a large contribution of PAHs from sealcoat tended to have high PAH concentrations; in many cases, at levels that can be harmful to aquatic life. Analysis of historical trends in PAH sources to 8 of the 40 lakes indicates that sealcoat use is the primary cause of increases in PAH concentrations since the 1960s. Identifying where PAHs are coming from is essential for developing environmental management strategies (Van Metre and Mahler, 2010).

From Outside to Inside
Coal-Tar Pavement Sealant Linked to PAHs in House Dust

House dust is an important source for human exposure to many contaminants, including PAHs. This is particularly true for small children, who spend time on the floor and put their hands and objects into their mouths. In 2008, the USGS measured PAHs in house dust from 23 ground-floor apartments and in dust from the apartment parking lots. Apartments with parking lots with coal-tar-based sealcoat had PAH concentrations in house dust that were 25 times higher, on average, than concentrations in house dust from apartments with parking lots with other surface types (concrete, unsealed asphalt, and asphalt-based sealcoat). PAH concentrations in the dust from the parking lots with coal-tar-based sealcoat were 530 times higher, on average, than concentrations on the parking lots with other surface types.



Apartments with coal-tar-based sealcoat on the parking lot had much higher concentrations of PAHs, both in indoor dust and in parking lot dust, than apartments with an unsealed asphalt or concrete parking lot or with a parking lot with asphalt-based sealcoat. Concentrations shown are for the sum of the 16 U.S. Environmental Protection Agency priority pollutant PAHs (Mahler and others, 2010), in milligrams per kilogram (mg/kg).



Photograph obtained from Jupiter Images.

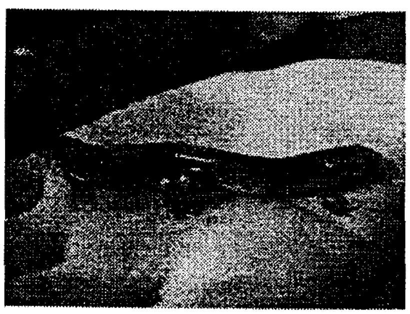
What about other sources of PAHs? Although tobacco smoking, candle and incense burning, and barbecue and fireplace use have been suggested to affect PAH concentrations in house dust, this study found no relation between any of these, or the many other factors considered, and PAH concentrations in the house dust. The presence or absence of coal-tar-based sealcoat on the apartment complex parking lot was strongly correlated with PAH concentrations in house dust; the only other variable that was related to PAH concentrations in house dust was urban land-use intensity (the percentage of land near the apartment dedicated to multifamily residential, commercial, office, warehouse, or streets) (Mahler and others, 2010).

There are no U.S. health-based guidelines for chronic exposure to PAHs in house dust. The only existing guideline is for a single PAH—benzo[*a*]pyrene—issued by the German Federal Environment Agency Indoor Air Hygiene Commission (Hansen and Volland, 1998). The guideline advises minimizing exposure to concentrations of benzo[*a*]pyrene greater than 10 mg/kg in dust to avoid adverse health effects. That guideline was exceeded for 4 of the 11 apartments with coal-tar-sealcoated parking lots and for 1 of the 12 apartments with a parking lot with a different surface type. Also of concern is exposure to the sealcoated pavement surfaces themselves through play activities. Dust on some of the sealcoated parking lots had a concentration of benzo[*a*]pyrene that was more than 50 times higher than the German guideline.

Photograph courtesy of CLEARCorps, Durham, North Carolina.

Our Environment and Us
What are the Concerns?

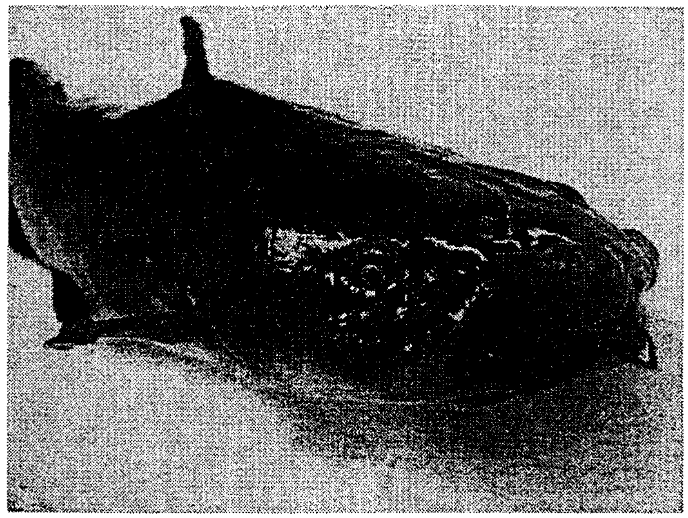
Some PAHs are toxic to mammals (including humans), birds, fish, amphibians (such as frogs and salamanders), and plants. The aquatic invertebrates—insects and other small creatures that live in streams and lakes—are particularly susceptible to PAH contamination, especially those that live in the mud where PAHs tend to accumulate. These invertebrates are an important part of the food chain and are often monitored as indicators of stream quality (analogous to the "canary in the coal mine" concept). Possible adverse effects of PAHs on aquatic invertebrates include inhibited reproduction, delayed emergence, sediment avoidance, and mortality. Possible adverse effects on fish include fin erosion, liver abnormalities, cataracts, and immune system impairments. The Probable Effect Concentration (PEC) of 22.8 mg/kg of total PAHs (MacDonald and others, 2000)—a widely used sediment quality guideline that is the concentration in bed sediment expected to have harmful effects on bottom-dwelling biota—is exceeded in one-third of the central and eastern U.S. urban lakes where PAH sources were studied.



When turned over, red spotted newts that had been exposed to sediment contaminated with coal-tar-based sealcoat had difficulty righting themselves (Bommarito and others, 2010b). Poor reflexes could result in decreased survival. Photograph by Megan Gibbons, Birmingham-Southern College.

Scientific studies have shown a relation between coal-tar-based pavement sealcoat and harmful effects on aquatic life.

- Aquatic communities downstream from stormwater runoff from sealcoated parking lots were impaired (Scoggins and others, 2007).
- Salamanders and newts exposed to sediment contaminated with coal-tar-based sealcoat had stunted growth, difficulty swimming or righting themselves, and liver problems (Bommarito and others, 2010a, b).
- Frogs exposed to sediment contaminated with coal-tar-based sealcoat died, had stunted growth, or developed more slowly than usual (Bryer and others, 2006).



Tumors in brown bullhead catfish from the Anacostia River, Washington, D.C., are believed to be related to elevated PAH concentrations (Pinkney and others, 2009). Photograph by A.E. Pinkney.

Human health risk from environmental contaminants usually is evaluated in terms of exposure pathways. For example, people could potentially be exposed to PAHs in sealcoat through ingestion of abraded particles from driveways, parking lots, or play grounds, or through skin contact with the abraded particles, either directly or by touching toys or other objects that have been in contact with the pavement. Inhalation of wind-blown particles and of fumes that volatilize from sealed parking lots are other possible pathways. PAHs in streams and lakes rarely pose a human health risk from contact recreation or drinking water because of their tendency to attach to sediment rather than to dissolve in water.

Skin contact is one way humans can be exposed to PAHs. Parking lots and driveways with coal-tar-based sealcoat have concentrations of PAHs hundreds to thousands of times higher than those with asphalt-based sealcoat or no sealcoat. Photograph obtained from Corbis Images, Inc.

FAQ

Q) *What is coal tar?*

A) Coal tar is a thick, black or brown liquid that is a byproduct of the carbonization of coal for the steel industry or the gasification of coal to make coal gas.

Q) *What is the difference between crude coal tar, coal-tar pitch, and "refined" coal tar?*

A) Coal-tar pitch is the residue that remains after various light oils are distilled from crude coal tar for commercial use. The coal-tar pitch is then separated (refined) into 12 different viscosities, RT–1 (the most fluid) through RT–12 (the most viscous). RT–12 is the viscosity used in coal-tar-based pavement sealcoat.

Q) *How can I tell if a product contains coal tar?*

A) To determine if the product has a coal-tar base, look for the Chemical Abstracts Service (CAS) number 65996–93–2 on the product Material Safety Data Sheet (MSDS). The words "coal tar," "refined coal tar," "refined tar," "refined coal-tar pitch," or other similar terms may be listed on the MSDS or on the product container.

Q) *Is sealcoat used on roads?*

A) Use on roads is extremely rare. Occasionally a private property, such as a housing development, will choose to have the roads sealcoated.

Q) *Is use of coal-tar-based sealant regulated?*

A) Several jurisdictions, including the City of Austin, Texas, the City of Washington, D.C., Dane County, Wisconsin, and several suburbs of Minneapolis, Minnesota, have banned use of coal-tar-based sealcoat. Similar bans are under consideration in other jurisdictions.

For more information on USGS research on PAHs and coal-tar-based sealcoat go to *http://tx.usgs.gov/coring/allthingssealcoat.html.*

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References

Bommarito, T., Sparling, D.W., and Halbrook, R.S., 2010a, Toxicity of coal-tar pavement sealants and ultraviolet radiation to *Ambystoma Maculatum*: Ecotoxicology, v. 19, no. 6, p. 1,147–1,156.

Bommarito, T., Sparling, D.W., and Halbrook, R.S., 2010b, Toxicity of coal-tar and asphalt sealants to eastern newts, *Notophthalmus viridescens*: Chemosphere, v. 81, no. 2, p. 187–193.

Bryer, P.J., Elliott, J.N., and Willingham, E.J., 2006, The effects of coal tar based pavement sealer on amphibian development and metamorphosis: Ecotoxicology, v. 15, no. 3, p. 241–247.

City of Austin, 2005, PAHs in Austin, Texas, sediments and coal-tar based pavement sealants: Watershed Protection Department, 55 p., accessed September 14, 2010, at *http://www.ci.austin.tx.us/watershed/downloads/coaltar_draft_pah_study.pdf.*

Hansen, D., and Volland, G., 1998, Study about the contamination of PAH in rooms with tar parquetry adhesive: Otto-Graf-Journal, v. 9, p. 48–60.

International Agency for Research on Cancer, 1980, Coal tars and coal tar pitches: accessed September 14, 2010, at *http://ntp.niehs.nih.gov/ntp/roc/eleventh/profiles/s048coal.pdf.*

MacDonald, D.D., Ingersoll, C.G., and Berger, T.A., 2000, Development and evaluation of consensus-based sediment quality guidelines for freshwater ecosystems: Archives of Environmental Contamination and Toxicology, v. 39, p. 20–31.

Mahler, B.J., Van Metre, P.C., Wilson, J.T., Musgrove, M., Burbank, T.L., Ennis, T.E., and Bashara, T.J., 2010, Coal-tar-based parking lot sealcoat—An unrecognized source of PAH to settled house dust: Environmental Science and Technology, v. 44, p. 894–900.

Pinkney, A.E., Harshbarger, J.C., and Rutter, M.A., 2009, Tumors in brown bullheads in the Chesapeake Bay watershed—Analysis of survey data from 1992 through 2006: Journal of Aquatic Animal Health, v. 21, p. 71–81.

Scoggins, M., McClintock, N., Gosselink, L., and Bryer, P., 2007, Occurrence of polycyclic aromatic hydrocarbons below coal-tar-sealed parking lots and effects on stream benthic macroinvertebrate communities: Journal of the North American Benthological Society, v. 26, no. 4, p. 694–707.

U.S. Environmental Protection Agency, 2009, Integrated Risk Information System (IRIS): accessed September 14, 2010, at *http://cfpub.epa.gov/ncea/iris/index.cfm.*

Van Metre, P.C., and Mahler, B.J., 2010, Contribution of PAHs from coal-tar pavement sealcoat and other sources to 40 U.S. lakes: Science of the Total Environment, v. 409, p. 334–344.

Van Metre, P.C., Mahler, B.J., and Wilson, J.T., 2009, PAHs underfoot—Contaminated dust from coal-tar sealcoated pavement is widespread in the United States: Environmental Science and Technology, v. 43, no. 1, p. 20–25.

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—B.J. Mahler and P.C. Van Metre

THE GREAT LAKES BINATIONAL TOXICS STRATEGY

The Facts about Coal Tar Sealants (CTS)

WHAT ARE COAL TAR SEALANTS?

Coal tar sealants (CTS) are used to protect, maintain and beautify asphalt pavement for driveways and parking lots.

WHY ARE COAL TAR SEALANTS USED?

Asphalt pavement develops cracks over time, and sealants may help protect the pavement surface. However, CTS are only one method of maintaining pavement.

PROBLEMS WITH COAL TAR SEALANTS

- **Coal tars and coal tar pitches are "*known to be human carcinogens*" according to the U.S. Dept. of Health and Human Services**
- **CTS contain 3.4% to 20% polycyclic aromatic hydrocarbons (PAHs) dry weight. PAHs are toxic to aquatic life, and several are suspected human carcinogens.**
- **CTS are a source of PAHs in stormwater runoff**
- **PAH "hot spots" are found in streams adjacent to parking lots using CTS**
- **CTS contribute more PAHs to runoff than viable alternatives**

LIMITATIONS OF COAL TAR SEALANTS

- **Tend to dry, shrink and crack with time**
- **Need reapplication about every 2 to 5 years, depending on wear**
- **Can cause surfaces to become slippery when wet**

ALTERATIVES TO COAL TAR SEALANTS

- **Consider using asphalt-based sealers, which contain 0.03% to 0.66% PAHs, much less than CTS**
- **Evaluate using permeable asphalt, which does not need sealed and allows stormwater to infiltrate**
- **Explore using gravel and concrete, which do not require sealant and reduce the urban heat island effect**
- **Promote shared driveways and parking lots to reduce the need for paved surfaces**

BE INFORMED

For more information about the effect of coal tar sealants on the environment visit http://water.usgs.gov/nawqa/asphalt_sealers.html or contact Barbara Mahler of the U.S. Geological Survey at bjmahler@usgs.gov.

BE PROACTIVE

Municipalities may choose to restrict the sale and/or use of CTS in their community. It is already happening! The City of Austin, TX and Dane County, WI have banned the use and sale of CTS. Visit http://www.cityofaustin.org/watershed/coaltar_ban.htm for more information.

BE CREATIVE

Consider the alternatives. Grid gravel and pervious concrete provide added benefits such as stormwater management, groundwater recharge, durability, and an enhanced aesthetic quality.

The Great Lakes Binational Toxics Strategy is committed to reducing or eliminating persistent toxic substances, especially those which bioaccumulate, from the Great Lakes Basin.

For more information see http://epa.gov/greatlakes/bns/.

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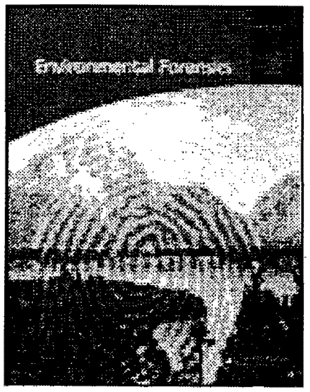

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Forensic Assessment of Refined Tar-Based Sealers as a Source of Polycyclic Aromatic Hydrocarbons (PAHs) in Urban Sediments

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Forensic Assessment of Refined Tar-Based Sealers as a Source of Polycyclic Aromatic Hydrocarbons (PAHs) in Urban Sediments

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Atmospheric deposition of particles and their subsequent transport by stormwater are a major source of polycyclic aromatic hydrocarbons (PAHs) in urban sediments. Recently, the results of forensic analysis have been used to promote a hypothesis that refined tar-based pavement sealers (RT-sealers) are another significant source. To evaluate this hypothesis, a suite of forensic methods was applied to a wider range of PAH data for this study. Sediments PAH profiles are no more similar to RT-sealers than they are to a number of other environmental inputs. While RT-sealers were not eliminated as a potential source in some locations, forensic methods did not differentiate their contribution from other sources of PAHs, indicating RT-sealers are not a unique or readily quantifiable source of PAHs to the urban environment.

Keywords: polycyclic aromatic hydrocarbons (PAHs), coal tar, refined tar, pavement sealers, sediments

Polycyclic aromatic hydrocarbon compounds (PAHs) are ubiquitous in the environment and are commonly found in aquatic sediments (Stout et al., 2004; Rodenburg et al., 2010). Given the multitude of natural and anthropogenic sources that may contribute PAH compounds to sediments, identifying and characterizing PAH sources has been the subject of significant research. Efforts to evaluate contributions of various petrogenic (fossil fuel-derived) and pyrogenic (high temperature and combustion-derived) sources have consistently identified atmospheric deposition as a significant source of PAHs to soils, paved areas, and sediments in most urban environments (Hwang and Foster, 2006; Li et al., 2003; Lima et al., 2005; Simcik et al., 1996; Stein et al., 2006; Su et al., 2000; Van Metre et al., 2000; Yunker et al., 2002). Specifically, the higher molecular weight PAHs typical of combustion-derived particulate matter, consistent with motor exhaust, coal combustion products, or wood smoke, have been found to dominate PAH profiles in sediments that are impacted by "urban background" sources (Stout et al., 2004).

A number of studies have demonstrated a link between atmospheric sources and PAHs in sediments. Evaluation of PAH chemistry in sediment from lakes, creeks, and reservoirs from across the United States report temporal links between changes in PAH concentrations and increased automobile use and vehicle emissions (Simcik et al., 1996; Stein et al., 2006; Su et al., 2000; Van Metre et al., 2000; Dickhut et al., 2000). In the upper Midwest, the mass and chemistry of PAHs in lake sediment could be linked to specific atmospheric sources associated with activities such as steel production and motor vehicle use (Su et al., 2000, Simcik et al., 1999). Automotive emissions have been shown to be a major source of particulate PAHs in aquatic systems in the Los Angeles basin (Stein et al., 2006) and San Francisco Bay Area (Tsai et al., 2002). Yunker et al., (2002) demonstrated a link between sediment chemistry and atmospheric sources throughout a regional watershed.

Since 2005, several studies have hypothesized that refined tar-based pavement sealer (RT-sealer) is another potentially significant source of PAHs to urban sediment (Mahler et al. 2005; Van Metre et al., 2009; Yang et al., 2010; Van Metre and Mahler 2010; 2011; Watts et al., 2010). The hypothesis is based on observations of elevated concentrations of PAHs in particles and runoff associated with RT sealer-treated parking lots and comparison of PAH compositions in sediment and potential source samples. Mahler et al. (2005) presented data suggesting that mean PAH concentrations of particles (μg PAH per kg particle) associated with RT-sealed parking lots was up to 65 times as high as the concentration of particles associated with non-sealed lots. During artificial rainfall events, the mean yield (μg PAH/m^2) within sealed lots was up to 44 times that of the unsealed lots. Offsite flux during actual rain events was not measured, but PAH concentrations decreased with distance from the source (McClintock et al., 2005). In a study conducted by a different USGS research team (Selbig, 2009), the mean PAH concentration (μg/L) in actual runoff from a sealed lot was six times that of an unsealed lot, but less than 2.5 times the concentration of runoff from a local roadway. Maximum total PAH

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concentrations for runoff from the sealed lot, 96 μg/L, and roadway, 98μg/L, were similar.

Three PAH diagnostic ratios, fluoranthene (Fl)/pyrene (Py), benzo[a]pyrene (BaP)/benzo[e]pyrene (BeP), and indeno[1,2,3-cd]pyrene (IDP)/benzo[ghi]perylene (BGP), were used to indicate similarities and differences between parking lot and sediment sample chemistries (Mahler et al., 2005; Van Metre et al. 2009). While there were overlaps in the observed ratios for the sealed lot particles and sediments, other potential sources that often have similar ratios were not considered. As noted by DeMott and Gauthier (2006), ratios did not overlap between lots and sediments from the same urban area.

Van Metre et al. (2009) evaluated PAH concentrations and composition of parking lot samples and sediments from 10 urban watersheds. Higher particulate PAH concentrations measured in dust collected from sealed lots in the eastern United States compared to the west were attributed to a greater use of RT-sealers in the east. Independent data on the relative use of sealer types by region were not presented. A Fl/Py versus BaP/BeP double ratio plot was used to suggest similarities between eastern sediments and particles from RT–sealed lots.

In Van Metre and Mahler (2010), the United States Environmental Protection Agency (US EPA) chemical mass balance (CMB; Coulter, 2004) model was used to estimate the relative contribution of RT-sealers versus other PAH sources in the sediments of 40 urban lakes. The model inputs were the PAH profiles of five standardized source types including RT-sealers, vehicle emissions, and wood, oil, and coal combustion-related samples. Two significant problems exist with the RT-sealer inputs used in the model. First, the authors admit that they assumed the parking lots sampled were treated with RT-sealer, and secondly, the data were prescreened to select inputs that were the most statistically similar to the sediment data set. Coal- and vehicle emissions-related source types were not the results of individual samples, but the averages of data from the literature. It appears that the averages were calculated by summing the published average concentrations of subclasses of coal or vehicle emission sources and then dividing by the total number of published sources. Because this approach results in weighting the influence of each subclass by numbers of publication, and not by their environmental contribution, it is unclear that the calculated value represents any real source. The authors of this study have concerns about the results of this PAH apportionment exercise because it appears that model requirements of source sufficiency and stability were violated (Galarneau, 2008; US EPA, 2004). These critical assumptions for receptor models require that all potential significant sources have been considered, and that the chemistries of the identified sources are stable. The use of source chemistry at the point of emission instead of atmospheric deposition ignores the effect of chemical reactions that are known to occur in the atmosphere (Galarneau, 2008; Katsoyiannis et al., 2011; Ravindra et al., 2008). Atmospheric reactions such as photolytic decay, with half-lives as short as 1 or 2 hours, and processes including nonequilibrium gas/particle partitioning complicate the application of receptor models for PAH source apportionment (Gordon, 1988). Models such as the CMB do not identify sources, but only statistically fit mixtures of sources identified by the modeler to receptor chemistry, so they have been described as "biased by *a priori* assumptions as to the number and nature of the contributing sources" (Galarneau, 2008, p. 8146).

Because of uncertainties associated with the results of any one single method, it is important to develop multiple lines of evidence when using environmental forensics to characterize source contributions (Stout and Graan, 2010). This approach is especially true when potential sources have similar chemistries such as those consisting of largely weathered pyrogenic PAHs. The goal of this study is to evaluate the hypothesis concerning the role of RT-sealers as a source of PAHs in urban sediments by applying multiple methods and considering a wider range of environmental samples representing potential contributing inputs. Possible outcomes of such analyses are that the results either support the hypothesis that RT-sealers are a dominant source of sediment PAHs (Mahler et al., 2005), fail to support it, or disprove the hypothesis. Results that fail to support a hypothesis do not mean it is incorrect, only that other explanations can account for the observed effects.

Experimental Section

Environmental chemical forensic methods are based on comparing the chemistry of the medium of interest, in this case sediment, with the chemistries of potential sources (Li et al., 2003; Su et al., 2000; Burns et al., 1997). For sediments, the term *source* can have two meanings, one of which is the *processes that create the chemicals of interest*, such as coal combustion or vehicle emissions; the second describes the *particulate matter that transports chemicals from the broader environment to sediments*. Each has advantages and disadvantages in source allocation. While there may be a better understanding of the processes resulting in the emission sources, they can be site specific and do not account for changes that may occur between the source location and sediment. The opposite is true for environmental particles: Whereas all the processes resulting in the observed PAH chemistry may not be understood; they better represent results of both generation and fate. Because of uncertainties in primary source characteristics and the changes that occur as a result of reactions in the atmosphere (Gordon, 1988; Galarneau, 2008; Ravindra et al., 2008; Golomb et al., 2001), data on a range of environmental particulate materials were evaluated as potential sources of PAHs. These sources included fresh RT-sealer, particles from RT-sealed lots, atmospheric particles, coal combustion and traffic related emissions, road dirt, roof dust, urban soil, and highway runoff. PAH data for both sediments and sources were compiled from the literature. Asphalt-based sealers were not included in this study because they have not been suggested as a significant source of PAHs (Mahler et al., 2005). A list of the data sets used is shown in Table 1. Except for fresh sealer material and a coal tar standard, the data were derived from analysis of environmental

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Table 1. Data sources used in this evaluation

Material	Reference	n [a]	PAH16? [b]	Fl/Py[c] Min	Fl/Py[c] Max	ID/BgP[d] Min	ID/BgP[d] Max
Coal tar	1 NIST 1597a [e]	1	Y	1.36		1.10	
Refined tar-based pavement (RT) products	2 Mahler et al. (2005)	6	Y	1.26	1.30	0.77	0.81
RT-sealed lots	3 Mahler et al. (2004)	20	Y	1.24	1.66	0.82	1.48
	4 Selbig (2009)	15	Y	1.26	1.64	0.82	0.95
Air particles	5 NIST 1649b [e]	1	Y	1.25		0.77	
	6 Simcik et al. (1999)	2[f]	N	1.18	1.25	0.85	1.29
	7 Li et al. (2003)	6[g]	Y	0.58	2.52	0.25	1.18
Roofs	8 Van Metre and Mahler (2003)	6	N	1.18	1.27	NA	NA
	9 Selbig (2009)	8	Y	1.23	1.30	0.81	1.00
Roads	10 Van Metre and Mahler (2003)	3	N	1.18	1.38	NA	NA
	11 Selbig (2009)	11	Y	1.28	1.67	0.78	0.95
	12 Breault et al. (2005)	5	N	1.27	1.52	0.61	1.38
Soils	13 Wilson et al. (2006)	6	Y	1.12	1.34	1.06	1.57
	14 Polta et al. (2006)	4	Y	0.40	1.10	0.50	1.00
Sediments	15 Wilson et al. (2006)	12	Y	1.23	1.42	0.90	1.18
	16 Polta et al. (2006)	50	Y	0.00	1.42	0.19	3.00
	17 Van Metre et al. (2009)	8	Y	0.66	1.48	0.47	1.55
	18 Van Metre and Mahler. (2010)	40[h]	N	0.72	1.52	0.55	1.78

Note: NA = Not measured
[a] Number of samples
[b] Does data set include all 16 of the priority pollutant PAHs yes (y) or no (n)?
[c] Fluoranthene / pyrene ratio
[d] Indeno[1,2,3-cd]pyrene / benzo[ghi]perylene ratio
[e] National Institute of Standards and Technology (NIST), Gaithersburg, MD
[f] Both results are the mean of multiple samples from Figure 2 in Simcik et al. (1999).
[g] Each sample represents an average of multiple samples for both traffic and coal combustion–related emissions.
[h] Each sample presents an average of three samples.

samples. While most of the studies analyzed particles, some such as Selbig (2009) analyzed unfiltered runoff, which would include both dissolved and particulate-associated constituents. Because organic carbon/water partitioning coefficients for the compounds used in the forensic analysis range from 10^4 to more than 10^6 (Hawthorne et al., 2007), the PAH profiles of the unfiltered samples were assumed to represent the particulate phase. Particulate bound PAHs have been shown to dominate over the dissolved phase compounds in environmental samples (Hwang and Foster, 2006).

The exact number and identity of PAH compounds analyzed differed among the studies evaluated, but typically most or all of PAHs of the 16 priority pollutant of the US EPA were included. Because of detection limit issues, fewer PAHs were reported in some cases. Individual sample data were typically available, but in some cases results were reported as the mean of a set of samples (Simcik et al., 1999; Li et al., 2003; Van Metre and Mahler, 2010). Where a concentration was listed as an estimated value or qualified with "J," it was included in the forensic analysis. To avoid skewing results based on detection limit issues, individual samples were excluded from the analysis if fewer than ten PAHs were detected. PAH diagnostic ratios were not calculated for samples where an analyte of interest was undetected. To standardize results of analysis among samples of different media and different contaminant levels, individual compound concentrations were converted to the relative fraction of the total PAHs, Ci, where the concentration of each compound, $[\text{PAH}]i$, is divided by the sum of the individual PAH concentrations, as shown in Equation (1):

$$Ci = [\text{PAH}]i / \Sigma\text{PAH}x \qquad (1)$$

Forensic Analysis

The data evaluation included diagnostic double ratio plots, (Boehm, 2006; Mahler et al., 2005) where the ratio of two PAHs was plotted on the x-axis and the ratio of a second pair of PAHs was plotted on the y-axis. Potential differences were identified by comparing the coordinates of samples to each other, to known sources, and to published values. Based on Mahler et al. (2005), the two PAH diagnostic ratios selected were the 4-ringed compounds Fl and Py and the 6-ringed IDP and BgP. Both ratios are commonly used in the PAH forensic literature to identify sediment sources (Stout et al., 2004; Yunker et al., 2002). While BaP/BeP has also been used to evaluate the influence of coal tars (Mahler et al., 2005; Van Metre et al., 2009), BeP data are available in fewer published studies because BeP is not one of the priority pollutant PAHs. When using double-ratio plots for source identification, it is critical to include an appropriate range of potential source materials to minimize the chance of misidentification (Yunker et al., 2002).

Pearson correlations were used to evaluate the similarities between sediments and sources (Yang et al., 2010; Van Metre and Mahler, 2010). For each sample pair, the *Ci*-values of each compound in one sample were set as the x values, while the *Ci*-values of each compound of the paired sample were set as the corresponding y values. The average *Ci* result of each source type was used. The student t test was used to compare data sets consisting of the r values between each source and sediments from 40 urban lakes with the null hypothesis that the mean of the Pearson's correlation population for each source type was the same with 95% confidence level. The Pearson correlation r was determined using statistical algorithms in Microsoft Excel 2007 (Microsoft, Redmond, WA) Pro-UCL (EPA, Washington, DC) was used to conduct the t tests.

Principal component analysis (PCA) is a statistical technique commonly used to compare sediment samples and suspected source materials (Stout and Graan, 2010; Sofowote et al., 2008). The objective of PCA is to reduce the dimensionality of data sets with a number of interrelated variables by transforming the data into uncorrelated principal components that account for the observed variance (Johnson et al., 2007). By plotting the results of each sample against the primary and secondary factors, more and less similar samples are identified. PCA also allows for identification of the compounds that contribute to the observed differences between the samples. To allow inclusion of sediment data from Van Metre and Mahler (2010), 11 priority pollutant PAHs were used as the input. PCA was conducted using Systat 12 (Systat Software, Chicago, IL).

The receptor model Unmix 6.0 (Norris et al., 2007) was used to evaluate the sediment data presented in Van Metre and Mahler (2010). The inputs were either the 120 samples from 40 lakes (3 samples per lake) or the 122 samples from the extended analysis of eight of these lakes (12−19 samples per lake). Unmix solves a general mixture problem where the data are assumed to be a positive linear combination of an unknown number of sources of unknown composition. Using concentration data for a given selection of chemical species, the model estimates the number of sources, source compositions, and source contributions to each sample (Norris et al., 2007). Like the CMB, Unmix was developed to evaluate atmospheric sources for air pollution monitoring, but similar approaches have been used to evaluate sediment source data (Bzdusek et al., 2004). A critical difference between the two models is that the chemistry of potential sources is not an input to Unmix.

Results and Discussion

The results of forensic analysis depend on the chemistry of the samples considered, so it is important to understand the overall nature of the PAH chemistry. Evaluation of the PAH concentration histograms can suggest whether a sample contained PAHs from a petrogenic or pyrogenic source(s) and whether the sample had weathered. PAH histograms also provide qualitative information about similarities and differences between samples and sources. Challenges arise when potential sources are similar, as it possible to misattribute the contribution of these sources.

Figure 1 contains average compositional PAH histograms for a number of environmental inputs and urban sediments. The three sediments and the modeled RT-sealer contribution are from Van Metre and Mahler (2010). The similarity among the different source types stands out, especially in the patterns of the 4- to 6-ringed compounds. This pattern is consistent with PAHs originating from pyrogenic sources. The patterns for all these materials are well known in the sediment literature, and are consistent with what is typically called "urban background" (Stout et al., 2004, p. 2987). Similarities between RT-sealers and other environmental samples are not surprising, even if sealers are not the source, because this pattern represents the balance between the relative forces that generate and decay PAHs.

The histograms indicate that fresh refined tar-based sealers have a greater concentration of the lower molecular weight PAH compounds such as naphthalene, acenaphthene, fluorene, and anthracene than environmental samples from studies listed in Table 1. These four lower molecular weight PAHs are depleted relative to the fresh product in most samples, including dust collected at lots sealed with refined tar-based sealers. Differences in PAH compositional patterns of fresh product and samples from sealed lots can be explained by the weathering of the lighter compounds (Burns et al., 1997).

Double-ratio plots for 40 urban lakes, RT-sealed lots, and other environmental sample types are shown in Figure 2. Table 1 shows the range of diagnostic ratios for each sample type. A regional trend is observed in these lake sediments, with samples from the central United States more toward the upper right corner, samples from the west toward the lower left, and eastern samples between the two. Van Metre et al. (2009) suggested that such a trend could be explained by an unreferenced claim of lower use of RT-sealers in the west compared to the other two regions (Van Metre et al., 2009). Other regional differences, such as the concentration of coal-based electricity generation, might also account for the results. The apparent regional trend may be an experimental artifact. In another study, 50 samples collected from 10 ponds in a single metropolitan area (Polta et al., 2006) had a similar range of ratios as those from the three regions (Figure 3).

To more closely evaluate the double ratio results of the RT-sealer and other environmental samples, sediment data were removed from Figure 4. Samples from RT-sealed lots in Texas and Wisconsin grouped closely with material such as roof dust and highway runoff. While the possibility of the presence of some sealer in these materials cannot be eliminated, it seems unlikely for the roof dust, which is not in direct contact with tires that might have driven on sealed pavement. If just these two diagnostic ratios are considered, one could argue that there may be more similarity between the test plot samples with freshly applied sealer and the sediment samples from the central United States, but forensic evaluation requires the use of multiple methods and, as will be seen in the PCA result, unique chemical

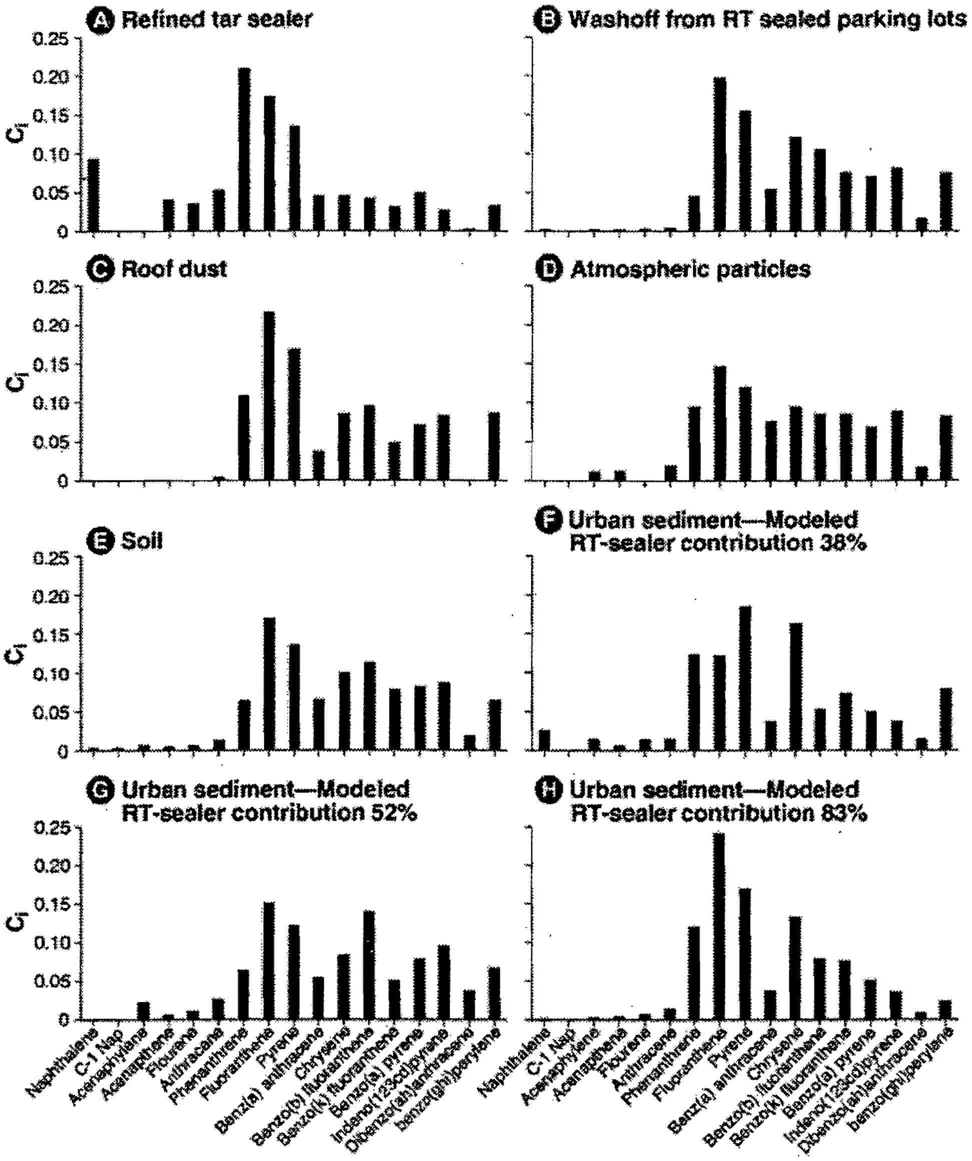

Figure 1. Polycyclic aromatic hydrocarbons (PAHs) concentration histograms for five environmental inputs and three urban sediments. The modeled refined tar-based pavement sealers (RT-sealers) contribution is from Van Metre and Mahler (2010).

similarity between sealers and sediment samples from the central United States is not supported.

In prior studies (Mahler et al., 2005; Van Metre et al., 2009), PAH ratio and double ratio analyses have been a primary forensic methods applied to evaluate the hypothesis concerning sealers. Because of uncertainties introduced by overlapping ranges for various sources, these types of ratio analyses are more useful for distinguishing between clearly different sources (such as petrogenic and pyrogenic) than for differentiating among similar ones such as a wide variety of pyrogenic sources of PAHs (Stout et al., 2004; Boehm, 2006). A number of combustion sources have been shown to have Fl/Py ratios consistent with the range for coal tar and RT-sealers reported by others and included in this paper (Costa and Sauer, 2005; Lima et al., 2005; Yunker et al. 2002). All appropriate potential sources must be included when evaluating the relative contribution of each to environmental sinks such as sediment. Yunker et al. (2002) considered more than 20 source classes and a variety of chemical ratios in an attempt to link combustion sources to sediment chemistry throughout a regional watershed, and argued that a limited assessment can result in misleading relationships between PAH sources and sinks. In another study, 18 potential source types were considered when evaluating the origin of PAHs in sediment samples (Burns et al., 1997).

The results of the CMB model (Van Metre and Mahler, 2010) and double ratio analyses are combined in Figure 5. If results of the CMB model were consistent with results of the double ratio method, one would expect there to be a relationship

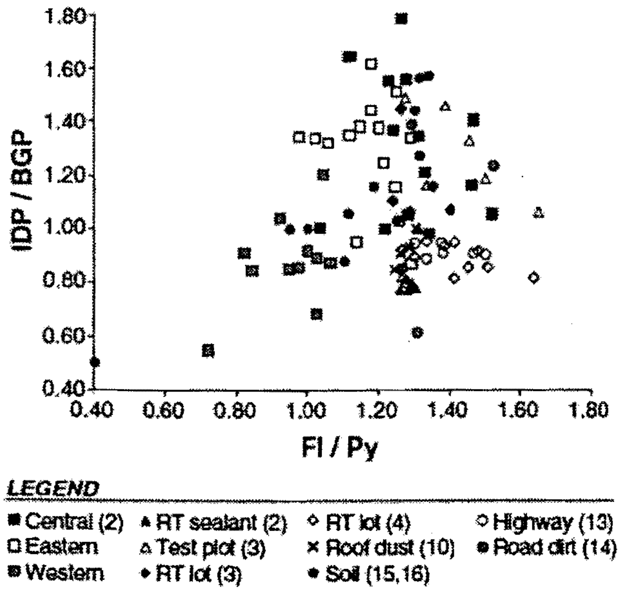

Figure 2. Polycyclic aromatic hydrocarbon (PAH) double-ratio plot comparing urban sediments with RT sealer, dust from sealed parking lots, atmospheric particles, roof dust, road runoff, and urban sediments. The sediments are classified by regions as identified in Van Metre and Mahler (2010). The numbers in parenthesis refer to the reference in Table 1.

between the PAH ratios and modeled fraction of sealer contribution. No relationship was noted as samples within each of four classes of percent RT-sealer contribution were calculated using the CMB model (≤25%, 26–50%, 51–75%, ≥76%) are spread across the range of PAH ratios. A similar lack of consistency has also been demonstrated between CMB results and the diagnostic PAH ratios Fl/Py versus BaP/BeP (O'Reilly et al., 2011).

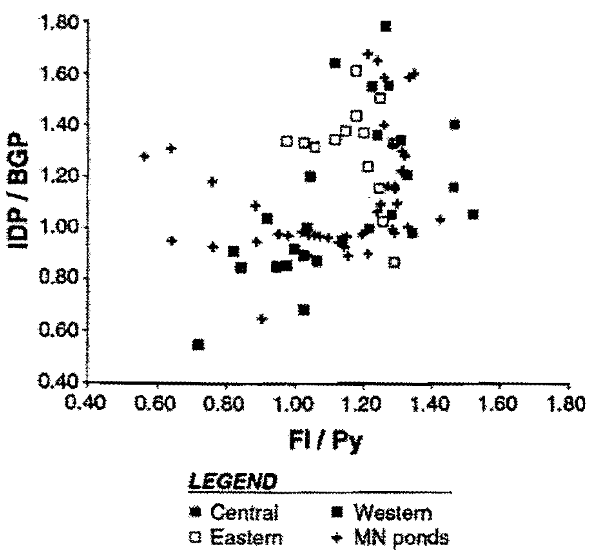

Figure 3. The range of polycyclic aromatic hydrocarbon (PAH) ratios for 10 ponds from one metropolitan area is similar to the range for samples taken across the country.

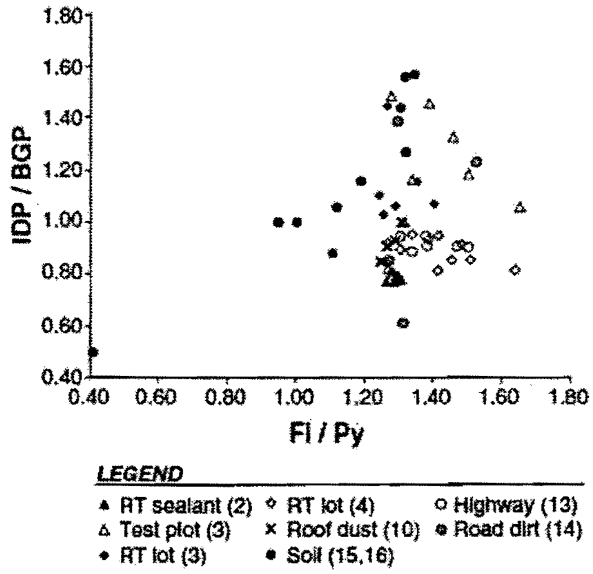

Figure 4. Double-ratio plot highlighting the similarities between environmental samples that may represent sources of polycyclic aromatic hydrocarbons (PAHs) to urban sediments. The numbers in parenthesis refer to the reference in Table 1.

Based on the selection of sources, it is not surprising that CMB results suggest that sealers are a major contributor to sediments. As noted by the authors, for most model runs they selected sealer sources for which they only assumed the presence of sealer, and of which the average Pearson coefficient (r) between source and the 40 lake sediments was greater than 0.95. The remaining sources had r of 0.83 (wood smoke), 0.67 (tunnel air), 0.55 (coal emissions), and 0.43 (fuel oil combustion). Sealer sources with average Pearson coefficients of 0.62, 0.55, and 0.38 were reported by the authors but the results of CMB modeling with these sources were not presented (Van Metre and Mahler, 2010). Using the compiled data set showed similar

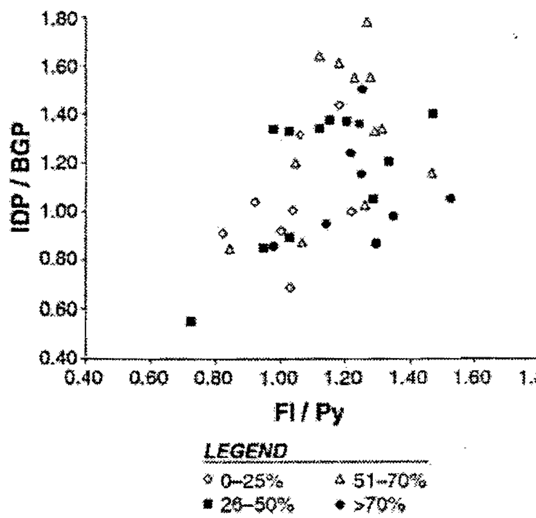

Figure 5. Comparison of double-ratio plot results to chemical mass balance (CMB) model output.

Table 2. t-test[a] results for comparison of Pearson coefficients for each lake sediment and source type.

		Product ($n = 6$)	IL Air ($n = 2$)	WI Roof ($n = 8$)	WI Lots ($n = 7$)	WI Highway ($n = 11$)	Texas Soil ($n = 6$)	Texas Lots ($n = 8$)	Texas Test Plots ($n = 8$)
Reference # from Table 1	Average r (Range)	0.32 (−0.3–0.69)	0.78 (0.27–0.95)	0.76 (0.42–0.93)	0.75 (0.33–0.91)	0.89 (0.24–0.99)	0.82 (0.48–0.94)	0.88 (0.28–0.99)	0.77 (0.07–0.96)
2	Product ($n = 6$)		Different ($p = 0.000$)	Different ($p = 0.000$)	Different ($p = 0.000$)	Different ($p = 0.000$)	Different ($p = 0.000$)	Different ($p = 0.000$)	Different ($p = 0.000$)
6	IL Air ($n = 2$)			*Same* (*p* = *0.618*)	*Same* (*p* = *0.533*)	Different ($p = 0.026$)	Different ($p = 0.000$)	Different ($p = 0.000$)	*Same* (*p* = *0.934*)
9	WI Roof ($n = 8$)				*Same* (*p* = *0.308*)	Different ($p = 0.011$)	Different ($p = 0.000$)	Different ($p = 0.000$)	*Same* (*p* = *0.644*)
4	WI Lots ($n = 7$)					*Same* (*p* = *0.188*)	Different ($p = 0.002$)	Different ($p = 0.003$)	*Same* (*p* = *0.678*)
11	WI Highway ($n = 11$)						Different ($p = 0.023$)	Different ($p = 0.041$)	*Same* (*p* = *0.115*)
13	Texas Soil ($n = 6$)							*Same* (*p* = *0.776*)	Different ($p = 0.001$)
3	Texas Lots ($n = 8$)								Different ($p = 0.003$)
3	Texas Test Plots ($n = 8$)								

[a] The comparison of Pearson's coefficients of each source and sediment by source type was based on the null hypothesis that the means of populations of the Pearson's coefficients for each source type are the same.

r values for soils from Fort Worth, TX (0.89), sealed lot dust from Austin, TX (0.88), and highway runoff from Madison, WI (0.82). The average r was also similar between sediments and sealed lot dust from Madison (0.75), sealer test plots in Austin (0.77), atmospheric particles from Chicago, IL (0.78), and roof dust from Madison (0.76). To further compare how these sources correlate with lake sediments, data sets consisting of the Pearson coefficient between each source and all 40 lakes were generated for the seven sources. Similarity between these data sets was evaluated using the student t test (Table 2). Consistent with the null hypothesis, no statistical difference was shown between the Pearson coefficients for each lake sediment and Austin sealed lots or Fort Worth soil, or between the sediments and Madison sealed lots, roof dust, highway runoff, and test plots in Austin. These results highlight the potential for biasing the outcome of receptor modeling by limiting the potential sources (Galarneau, 2008), and prescreening inputs to select specific source characteristics.

Whether using PAH ratios or more advanced receptor modeling such as the CMB, there are a number of challenges in linking PAHs in sediments to particular sources. A critical assumption in both modeling and forensic ratio analyses is that all potential significant sources have been considered, and that the chemistries of the identified sources are stable (Galarneau, 2008; Stout and Graan, 2010). To minimize these concerns, some investigators include ten or more well characterized sources even when applying receptor models to a single area (Boehm et al. 2001; Burns et al., 2006). Van Metre and Mahler (2010) focus on results from model runs where the inputs for RT-sealers were prescreened and only four other source types were used. As some of these other inputs were the geometric means of literature data compiled by another author, it is not possible to know how representative they are to actual source chemistries.

As described in a recent review (Galarneau, 2008) highlighting the potential pitfalls of PAH source apportionment, the use of source chemistry profiles based on samples collected near the point of emission fails to take into consideration changes caused by a variety of processes that affect the source material after environmental release, such as phase partitioning, differential settling, and photochemical-biodegradative reactions. Not only have these processes been shown to modify PAH profiles (Ravindra et al., 2008), they are of particular concern with PAHs because of differences in source characteristics, reactivity, and transport based on particulate size. Even within atmospheric studies, the use of emission chemistry in PAH source allocation may only be appropriate under limited conditions (Katsoyiannis et al., 2011). Because PAH profiles are known to change between emission and deposition in sediments, inclusion of decay rates should be considered when applying receptor models (Golomb et al., 2001; Xue et al., 2010).

A difference between multivariate methods, such as PCA or Unmix, and the CMB model is that pre-selection of a potential source is not required. The results of PCA using the entire sample set are presented in Figure 6. Sediment samples from Van Metre and Mahler (2010) are identified by the relative fraction of sealer contribution estimated by CMB. A first step in evaluating PCA is to review the factor-loading chart that indicates the influence of the individual compounds. As shown in Figure 7a, the less stable 3-ringed PAHs are in the upper left quadrant whereas the more stable 6-ringed compounds are toward the lower right.. The 4-ringed PAHs are spread perpendicular to the line between the 3- and 6-ringed compounds, with the 5-ringed group toward the right. This finding suggests that weathering

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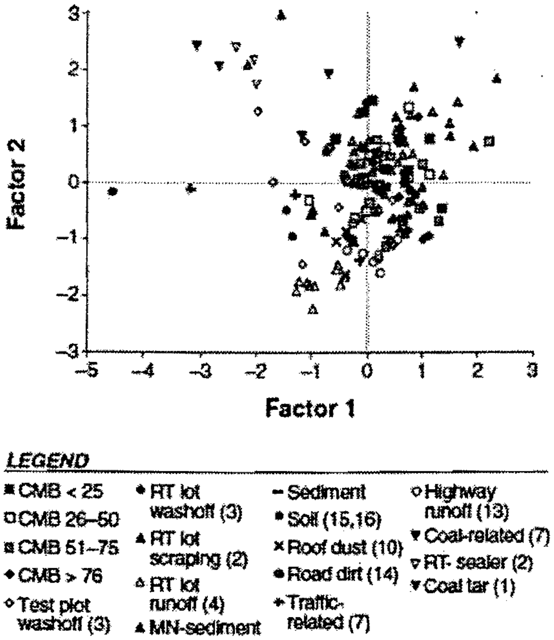

Figure 6. Principal component analysis of samples from refined tar-based pavement sealed (RT-sealed) parking lots, atmospheric particles, roof dust, road runoff, and urban sediments. The sediments are separated by results of the CMB model. Factors 1 and 2 explained 31 and 17% of variance, respectively. The numbers in parenthesis refer to the reference in Table 1.

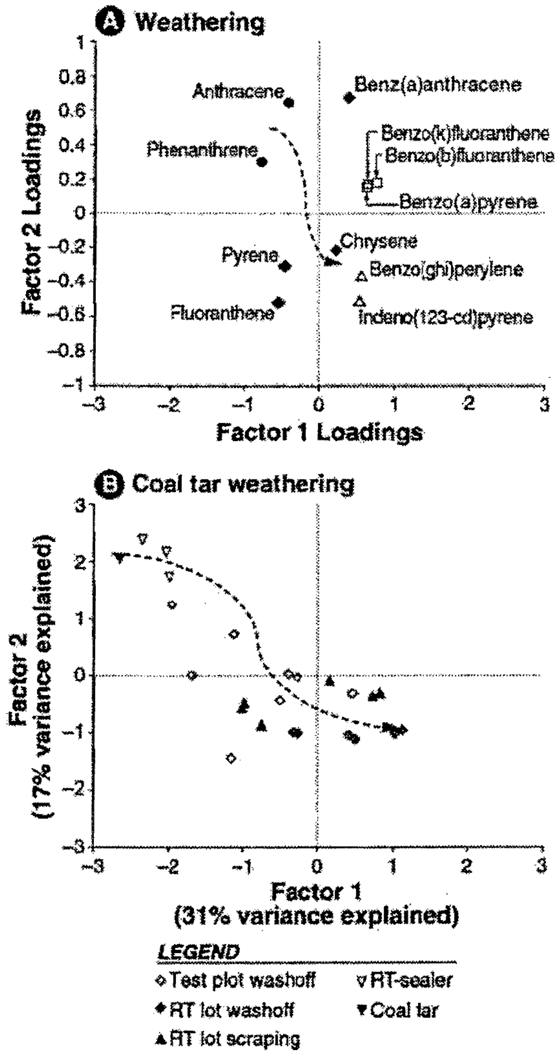

Figure 7. Illustration of impact of weathering of coal tar and refined tar-based pavement sealant (RT-sealant): (a) loadings for factors 1 and 2 for PCA shown in Figure 6; and (b) plot of factors 1 and 2 including only the coal tar and RT-sealant samples.

will result in a trend from the upper left toward the lower right. Such a trend is seen with the known RT-sealer samples with the fresher materials toward the upper left and the more aged toward the lower right quadrant (Figure 7b). The use of weathered sealer samples and unweathered emission source samples as CMB inputs may have skewed the output toward a higher estimated sealer contribution.

To allow clearer comparison of the various environmental inputs, Figure 8 shows the same information as Figure 6 but with most sediment samples removed. Samples from the sealed lots, and many of those from roof dust, soil, and highway runoff are in the lower half near the center of the plot. While some sediment samples overlap with all these sample types, most sediment results are in the upper right quadrant. Looking at the results from the 40 lake sediments, there might be a trend with samples having a lower modeled sealer contribution being more in the upper right quadrant, and samples with greater modeled contribution being in the lower right, but as with the double ratio plots, there is overlap among all four groups. The PCA results do not support the finding of a regional trend in sediment chemistry suggested by ratio analysis (Figure 9), or that sealers are a unique source of PAHs in sediments.

Unmix was unable to find a solution using 12 PAHs, but could find solutions if the three-ringed phenanthrene and anthracene were excluded. Two source profiles were identified for the 40 lakes sample set and three sources for the 8 lake samples (Figure 10). The two source profiles from the 40 lakes set are similar, with Source 1 having a greater fraction of IDP and BGP. The characteristics of Sources 1 and 2 of the 8 lake data results are even more similar, falling in between the chemistry of the 40 lakes Sources 1 and 2. The profile of the 8 lakes Source 3 is

Table 3. Pearson coefficients between potential environmental sources and the source profiles determined by Unmix using the 40 and eight lakes sample sets from Van Metre and Mahler (2010). The highest coefficient with each Unmix source profile is bolded.

Source	40LS1	40LS2	8LS1	8LS2	8LS3
Coal-related	0.05	**0.86**	0.65	0.53	**0.91**
Traffic-related	0.52	0.69	0.70	0.70	0.76
Wood	0.34	0.80	0.68	0.62	0.86
Fuel oil	**0.78**	0.29	0.68	0.54	0.33
Texas lot dust	0.52	0.84	0.82	0.84	0.86
Highway	0.70	0.68	**0.86**	0.83	0.73
Roof dust	0.66	0.69	0.84	0.78	0.75
Soil	0.44	0.89	0.82	**0.89**	0.89
Coal tar sealant	0.37	0.79	0.76	0.59	0.86

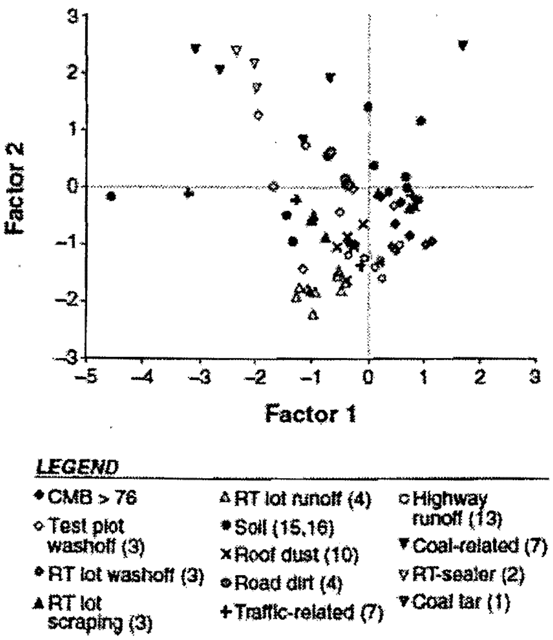

Figure 8. Principal component analysis from Figure 6 with most sediment sample points removed to allow comparison of the sources and other environmental samples. The numbers in parenthesis refer to the reference in Table 1.

weighted toward the lighter PAHs. Using these three sources profiles, the relationship between the predicted and measured concentrations exceeded 0.95 for all of the PAHs.

Since Unmix does not identify the sources, Pearson coefficients were used to compare the results to the potential sources (Table 3). For the 40 lakes data, Source 1 had the greatest similarity to the fuel oil combustion profile used as a input to the CMB (0.79), while Source 2 had the greatest similarity to the coal-related sources (0.86) and Austin lot dust (0.84). The three source profiles from the eight lakes run were most similar to

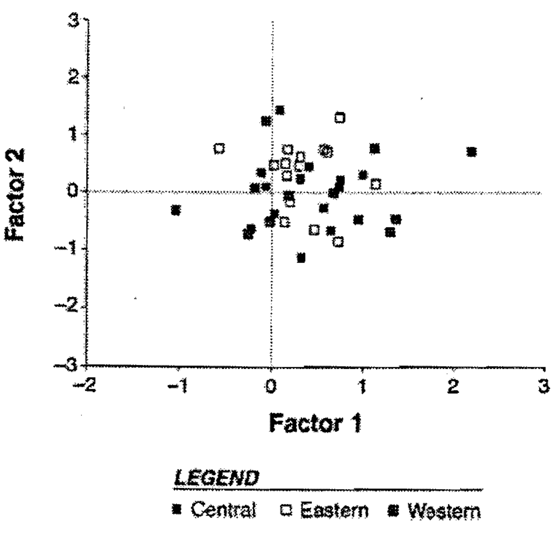

Figure 9. Principal component analysis results from Figure 6 highlighting the sediment results by regions identified by Van Metre and Mahler (2010).

highway runoff (Source 1, 0.86), soil (Source 2, 0.89), and coal–related sources (Source 3, 0.91). Pearson coefficients between the first two sources and Austin lot dust, highway runoff, roof dust, and soil were similar. If this evaluation had suggested that the PAH profiles identified by Unmix represented unique and independent sources, additional analysis that considered the confidence intervals of the output would be required for source identification (Norris et al., 2007; Pancras et al., 2011).

While Unmix characterized two or three source types, it is likely that there are more sources of PAHs in aquatic systems. What the results suggest is that the chemical profile of urban sediments is consistent with a mix of sources that differ in the fraction of lighter and heavier pyrogenic PAHs. These differences can be the result of both source-specific chemistry and the extent of weathering that occur between the source and deposition in sediment. Parsing out the relative contribution of these sources is challenging when using any of the forensic methods discussed.

Because of similarity in their PAH composition, multiple lines of environmental forensic data analyses are needed to examine possible linkages between sources and environmental samples, especially when investigating potential contributions of pyrogenic PAH sources to distal sediments (Boehm, 2006; Stout et al., 2001). Identification of a distinguishing factor among sources with otherwise similar pyrogenic PAH compositions may provide a sufficient basis for linking environmental samples to a source (Boehm et al., 1997). While the PAH histograms of fresh RT-sealer product suggest some potential distinguishing factors (i.e., greater relative amounts of the 2- and 3-ringed PAHs), this particular feature becomes less distinct as the sealer weathers and the lighter PAHs are lost. Because the variability within source type histograms and the source ratios themselves (Table 1) are similar to those between sources, no unique chemical indicator of RT-sealer is clearly identified within this limited set of 16 compounds used to link impacts to sediments. The evaluation of non-PAH source tracers may be required to accurately characterize the sources of PAHs. Such source-specific tracers may be present in RT-sealers, but no such data were found in the literature. In the case of other potential sources of PAHs, for example, in atmospheric source apportionment, hopanes and steranes indicate motor vehicle exhaust whereas levoglucosan and resin acids are indicative of biomass combustion (Fraser and Laskhmanan, 2000). Similarly, a number of markers have been identified that can assist in identifying coal combustion sources (Oros and Simoneit, 2000). If such differential signals are identified within otherwise similar chemical profiles, models such as CMB or Unmix may be sufficient to estimate the contribution of uniquely identifiable sources of PAHs (Burns et al., 1997).

Consistent with the approach of Van Metre and Mahler, data from numerous lakes were evaluated together. This is different from the typical application of forensic methods to a single watershed or water body (Li et al., 2003; Sofowote et al., 2008; Stout and Graan 2010; Su et al., 2000). Both the CMB and Unmix models were developed for use within a single source zone

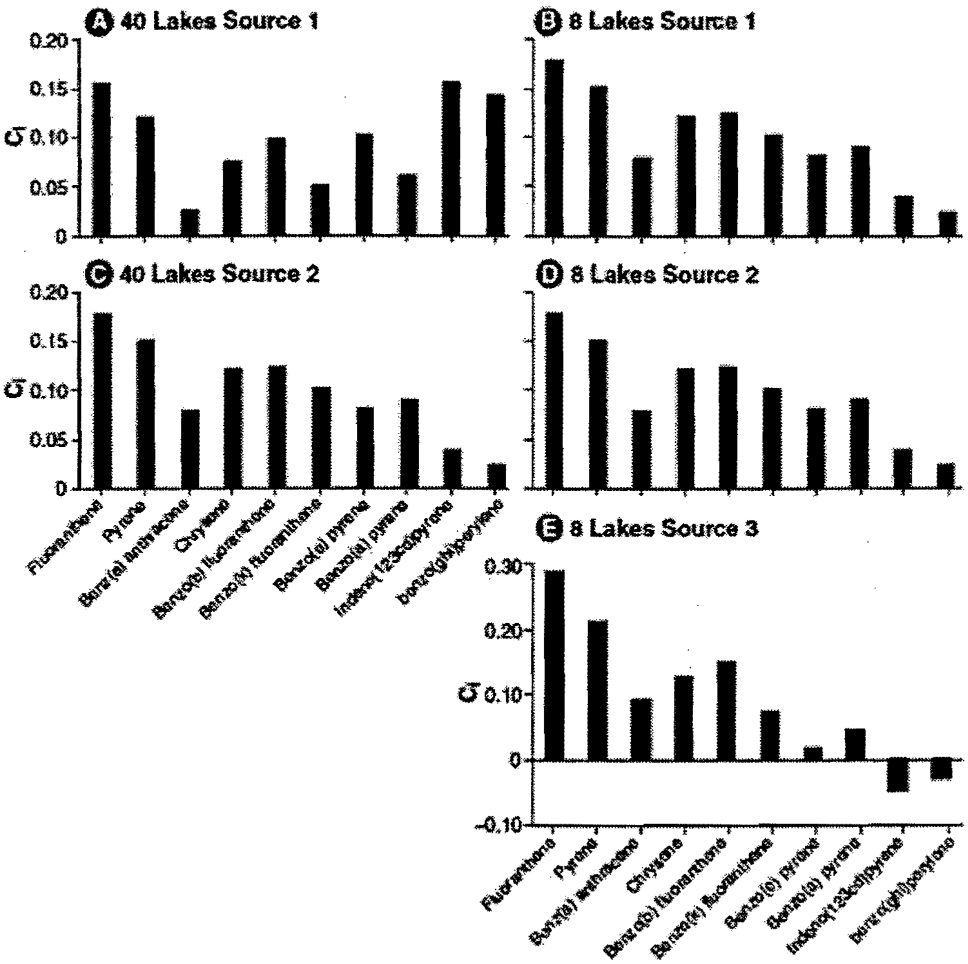

Figure 10. Source profiles determined by Unmix using the 40 and 8 lakes sample sets from Van Metre and Mahler (2010).

(Coulter, 2004; Norris et al., 2007), and the results of methods such as ratio analysis are more meaningful when local differences in PAH profiles are considered (Stout et al., 2004). Given that the type and influence of sources is site specific and the variability in sediment chemistry indicated by the 40 lakes data set, the ability to accurately identify sources by applying forensic methods to a multisite data set is unproven. This is especially true when trying to evaluate the contribution of sources with similar PAH chemistry.

A hypothesis is a proposed explanation for observed phenomenon that is typically tested by attempting to demonstrate the null hypothesis. A hypothesis is supported if the phenomenon could occur only if the proposed explanation is correct, while the null hypothesis is appropriate if other explanations cannot be eliminated. The goal of this study was to evaluate the hypothesis that RT-sealers are a dominant (Mahler et al., 2005) or substantial (Van Metre et al., 2009) source of PAHs to urban sediments. The hypothesis would be supported if the PAH profile in the lakes studied could not be explained without inclusion of the sealants as a source. The results of this study indicate that while RT-sealer cannot be eliminated as a PAH source, sediment chemistry can be explained in the absence of any contribution from sealers. While Van Metre and Mahler's work has identified similarities between the PAH profiles of RT-sealer and urban sediments, such profiles are not unique, so the similarity does not prove that one is the source of the other.

Acknowledgment

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References

Boehm, P. D. 2006. Polycyclic aromatic hydrocarbons (PAHs). In *Environmental Forensics—A Contaminant-Specific Approach*, eds., R. Morrison and B. Murphy, New York, NY: Elsevier.

Boehm, P. D., Douglas, G. S., Burns, W. A., Mankiewicz, P. J., Page, D. S., and Bence, A. E. 1997. Application of petroleum hydrocarbon chemical fingerprinting and allocation techniques after the *Exxon Valdez* oil spill. *Marine Pollution Bulletin* 34(8): 599–613.

Boehm, P. D., Page, D. S., Burns, W. A., Bence, A. E., Mankiewicz, P. J., and Brown, J. S. 2001. Resolving the origin of the petrogenic hydrocarbon background in Prince William Sound, Alaska. *Environmental Science and Technology* 35(3): 471–479.

Breault, R. F., Smith, K. P., and Sorenson, J. R. 2005. *Residential street-dirt accumulation rates and chemical composition, and removal efficiencies*

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by mechanical- and vacuum-type sweepers, New Bedford, Massachusetts, 2003–04. United States Geological Survey, Reston, VA. *Scientific Investigations Report 2005–5184*.

Burns, W. A., Mankiewicz, P. J., Bence, A. E., Page, D. S., and Parker, K. R. 1997. A principal-component and least-squares method for allocating polycyclic aromatic hydrocarbons in sediment to multiple sources. *Environmental Toxicology and Chemistry* 16(6): 1119–1131.

Burns, W. A., Mudge, S. M., Bence, A. E., et al. 2006. Source allocation by least–squares hydrocarbon fingerprint matching. *Environmental Science and Technology* 46(21): 6561–6567.

Bzdusek, P. A., Christensen, E. R., Li, A., and Zou, Q. M. 2004. Source apportionment of sediment PAHs in Lake Calumet, Chicago: Application of factor analysis with nonnegative constraints. *Environmental Science and Technology* 38: 97–103.

Costa, H. J., and Sauer, T. C. 2005. Forensic approaches and considerations in identifying PAH background. *Environmental Forensics* 6:9–16.

Coulter, C. T. 2004. *EPA-CMB 8.2 Users' Manual*, EPA-452/R-04–011. Washington, DC: U.S. Environmental Protection Agency.

DeMott, R. P., and Gauthier, T. D. 2006. Comment on parking lot sealcoat: An unrecognized source of urban polycyclic aromatic hydrocarbons. *Environmental Science and Technology* 40(11): 3657–3658.

Dickhut, R. M., Canuel, E. A., Gustafson, K. E., et al. 2000. Automotive sources of carcinogenic polycyclic aromatic hydrocarbons associated with particulate matter in the Chesapeake Bay Region. *Environmental Science and Technology* 34:4635–4640.

Fraser, M. P., and K. Lakshmanan. 2000. Using levoglucosan as a molecular marker for the long-range transport of biomass combustion aerosols. *Environmental Science and Technology* 34:4560–4564.

Galarneau, E. 2008. Source specificity and atmospheric processing of airborne PAHs: Implications for source apportionment. *Atmospheric Environment* 42(11): 8139–8149.

Golomb, D., Barry, E., Fisher, G., Varanusupakul, P., Koleda, M., and Rooney, T. 2001. Atmospheric deposition of polycyclic aromatic hydrocarbons near New England coastal waters. *Atmospheric Environment* 35(36): 6245–6258.

Gordon, G. E. 1988. Receptor models. *Environmental Science and Technology* 22:1132–1142.

Hawthorne, S. B., Grabanski, C. B., and Miller, D. J. 2007. Measured partition coefficients for parent and alkyl polycyclic aromatic hydrocarbons in 114 historically contaminated sediments: Part 2. Testing the $K_{OC}K_{BC}$ two carbon-type model. *Environmental Toxicology and Chemistry* 26(12): 2505–2516.

Hwang, H-M., and Foster, G. D. 2006. Characterization of polycyclic aromatic hydrocarbons in urban stormwater runoff flowing into the tidal Anacostia River, Washington, DC, USA. *Environmental Pollution* 140:416–426.

Johnson, G. W., Ehrlich, R., Full, W., and Ramos, S. 2007. Principal components analysis and receptor models in environmental forensics. In *Introduction to Environmental Forensics*, 2nd ed., eds., B. L. Murphy and R. D. Morrison, Burlington, MA: Elsevier Academic Press.

Katsoyiannis, A., Sweetman, A. J., and Jones, K. C. 2011. PAH molecular diagnostic ratios applied to atmospheric sources: A critical evaluation using two decades of source inventory and air concentration data from the UK. *Environmental Science and Technology* 45:8897–8960.

Li, A., Jang, J.-K., and Scheff, P. A. 2003. Application of EPA CMB8.2 model for source apportionment of sediment PAHs in Lake Calumet, Chicago. *Environmental Science and Technology* 37:2958–2965.

Lima, A. L. C., Farrington, J. W., and Reddy, C. M. 2005. Combustion-derived polycyclic aromatic hydrocarbons in the environment—A review. *Environmental Forensics* 6:109–131.

Mahler, B. J., Van Metre, P. C., and Wilson, J. T. 2004 [revised 2007]. Concentrations of polycyclic aromatic hydrocarbons (PAHs) and major trace elements in simulated rainfall runoff from parking lots, Austin, TX, 2003 (version 3). *U.S. Geological Survey Open–File Report 2004–1208*, 87 pp.

Mahler, B. J., Van Metre, P. C., Bashara, T. J., Wilson, J. T., and Johns, D. A. 2005. Parking lot sealcoat: An unrecognized source of urban polycyclic aromatic hydrocarbons. *Environmental Science and Technology* 39:5560–5566.

McClintock, N. L. Turner, M., Gosselink, L., and Scoggins, M. 2005. *PAHs in Austin, Texas sediments and coal-tar based pavement sealants polycyclic aromatic hydrocarbons*. Austin, TX: City of Austin Watershed Protection and Development Review Department, Environmental Resources Management Division.

Norris, G., Vedantham, R., Duvall, R., and Henry, R. C. 2007. *EPA Unmix 6.0 fundamentals and user guide*. EPA/600/R–07/089. Washington DC: U.S. Environmental Protection Agency.

O'Reilly, K., Pietari, J., and Boehm, P. 2011. Comment on PAHs underfoot: Contaminated dust from coal-tar seal coated pavement is widespread in the U.S. *Environmental Science and Technology* 45:3185–3186.

Oros, D. R., and Simoneit, B. R. T. 2000. Identification and emission rates of molecular tracers in coal smoke particulate matter. *Fuel* 79: 515–536.

Pancras, P. J., Vedantham, R., Landis, M. S., Norris, G. A., and Ondov, J. M. 2011. Application of EPA Unmix and nonparametric wind regression on high time resolution trace elements and speciated mercury in Tampa, Florida aerosol. *Environmental Science and Technology* 45(8): 3511–3518.

Polta, R., Balogh, S., and Craft-Reardon, A. 2006. *Characterization of stormwater pond sediments final project report*. Metropolitan Council Environmental Services, Environmental Quality Assurance Department, EQA Report 06–572. Available at: http://www.metrocouncil.org/Environment/sediment/FinalReport.pdf

Ravindra, K., Sokhi, R., and Van Grieken, R. 2008. Atmospheric polycyclic aromatic hydrocarbons: Source attribution, emission factors and regulation. *Atmospheric Environment* 42:2895–2921.

Rodenburg, L. A., Valle, S. N., Panero, M. A., Muñoz, G. R., and Shor, L. M. 2010. Mass balances on selected polycyclic aromatic hydrocarbons in the New York–New Jersey Harbor. *Journal of Environmental Quality* 39:642–653.

Selbig, W. R. 2009. *Concentrations of polycyclic aromatic hydrocarbons (PAHs) in urban stormwater, Madison, Wisconsin, 2005–08*. Reston, VA: U.S. Geological Survey. Open-File Report 2009–1077, 46 p.

Simcik, M. F., Eisenreich, S. J., Golden, K. A., et al. 1996. Atmospheric loading of polycyclic aromatic hydrocarbons to Lake Michigan as recorded in sediments. *Environmental Science and Technology* 30(10): 3039–3046.

Simcik, M. F., Eisenreich, S. J., and Lioy, P. J. 1999. Source apportionment and source/sink relationships of PAHs in the coastal atmosphere of Chicago and Lake Michigan. *Atmospheric Environment* 33: 5071–5079.

Sofowote, U. M., McCarry, B. E., and Marvin, C. H. 2008. Source apportionment of PAH in Hamilton Harbour suspended sediments: Comparison of two factor analysis methods. *Environmental Science and Technology* 42:6007–6014.

Stein, E. D., Tiefenthaler, L. L., and Schiff, K. 2006. Watershed-based sources of polycyclic aromatic hydrocarbons in urban storm water. *Environmental Toxicology and Chemistry* 25(2): 373–385.

Stout, S., and Graan, T. 2010. Quantitative source apportionment of PAHs in sediments of Little Menomonee River, Wisconsin: Weathered creosote versus urban. *Environmental Science and Technology* 44(8): 2932–2939.

Stout, S. A., Magar, V. S., Uhler, R. M., Ickes, J., Abbott, J., and Brenner, R. 2001. Characterization of naturally-occurring and anthropogenic PAHs in urban sediments—Wycoff/Eagle Harbor Superfund Site. *Environmental Forensics* 2:287–300.

Stout, S. A., Uhler, A. D., and Emsbo-Mattingly, S. D. 2004. Comparative evaluation of background anthropogenic hydrocarbons in surficial sediments from nine urban waterways. *Environmental Science and Technology* 38(11): 2987–2994.

Su, M.-C., Christensen, E. R., Karls, J. F., Kosuru, S., and Imamoglu, I. 2000. Apportionment of polycyclic aromatic hydrocarbon sources in Lower Fox River, USA, sediments by a chemical mass balance model. *Environmental Toxicology and Chemistry* 19(6): 1481–1490.

Tsai, P., Hoenicke, R., Yee, D., Bamford, H. A., and Baker, J. E. 2002. Atmospheric concentrations and fluxes of organic compounds in the northern San Francisco Estuary. *Environmental Science and Technology* 36: 4741–4747.

U.S. Environmental Protection Agency (US EPA). 2004, December. *Protocol for Applying and Validating the CMB Model for $PM_{2.5}$ and VOC.* Research Triangle Park, NC: Office of Air Quality Planning & Standards. EPA-451/R-04-001.

Van Metre, P. C., and Mahler, B. J. 2003. The contribution of particles washed from rooftops to contaminant loading to urban streams. *Chemosphere* 52:1727–1741.

Van Metre, P. C., and Mahler, B. J. 2010. Contribution of PAHs from coal-tar pavement sealcoat and other sources to 40 U.S. lakes. *Science of the Total Environment* 409:334–344.

Van Metre, P. C., and Mahler, B. J. 2011. Response to comment on PAHs underfoot: Contaminated dust from coal-tar seal coated pavement is widespread in the U.S. *Environmental Science and Technology* 45:3187–3188.

Van Metre, P. C., Mahler, B. J., and Furlong, E. T. 2000. Urban sprawl leaves its PAH signature. *Environmental Science and Technology* 34:4064–4070.

Van Metre, P. C., Mahler, B. J., and Wilson, J. T. 2009. PAHs underfoot: Contaminated dust from coal-tar sealcoated pavement is widespread in the United States. *Environmental Science and Technology* 43(1): 20-25.

Watts, A. W., Ballestero, T. P., Roseen, R. M., and Houle, J. P. 2010. Polycyclic aromatic hydrocarbons in runoff from sealcoated pavements. *Environmental Science and Technology* 44(23):8849–8854.

Wilson, J. T., Van Metre, P. C., Werth, C. J., and Yang, Y. 2006. *Particle-associated contaminants in street dust, parking lot dust, soil, lake-bottom sediment, and suspended and streambed sediment, Lake Como and Fosdic Lake watersheds, Fort Worth, Texas, 2004.* U.S. Geological Survey, Data Series 211, 24 p.

Xue L., Lang, Y., Liu, A., and Liu, J. 2010. Application of CMB model for source apportionment of polycyclic aromatic hydrocarbons (PAHs) in coastal surface sediments from Rizhao offshore area, China. *Environmental Monitoring and Assessment* 163:57–65.

Yang, Y., Van Metre, P. C., Mahler, B. J., et al. 2010. Influence of coal–tar sealcoat and other carbonaceous materials on polycyclic aromatic hydrocarbon loading in an urban watershed. *Environmental Science and Technology* 44(4): 1217–1233.

Yunker, M. B., Macdonald, R. W., Vingarzan, R,. Mitchell, R. H., Goyette, D., and Sylvestre, S. 2002. PAHs in the Fraser River basin: A critical appraisal of PAH ratios as indicators of PAH source and composition. *Organic Geochemistry* 33:489–515.

Actions to Restrict or Discontinue the Use of Coal Tar-Based Sealants in the United States

Current as of May 15, 2012

Action	State/District	Jurisdiction or Company*	2010 Population**
Ban or Ordinance	District of Columbia	Washington	601,723
	Minnesota	Buffalo	15,453
		Cannon Falls	4,083
		Centerville	3,792
		Circle Pines	4,918
		Eden Prairie	60,797
		Edina	47,941
		Falcon Heights	5,321
		Golden Valley	20,371
		Inver Grove Heights	33,880
		Little Canada	9,773
		Maplewood	38,018
		New Hope	20,339
		Prior Lake	22,796
		Rosemount	21,874
	Minnesota Population Affected 414,855	Roseville	33,660
		Shoreview	25,043
		Vadnais Heights	12,302
		Waconia	10,697
		White Bear Lake	23,797
	New York	Suffolk County	1,493,350
	Texas	Austin	790,390
		Bee Cave	3,925
	Washington	Statewide	6,724,540
	Wisconsin	Dane County	488,073
		Total Population Affected	10,516,856
Restricted Use Jurisdictions	Massachusetts	Andover Wetlands	
		Commonwealth Wetlands	
		Sudbury	
	North Carolina	Boone	
Government Use Restrictions†	California	California Department of Transportation	
	Illinois	City of Lake in the Hills	
		City of Spring Grove	
		DuPage County/Salt Creek Watershed	
		McHenry County	
	Minnesota	All State Agencies	
	Missouri	City of Springfield	
Home Improvement Stores Which Have Stopped Selling Coal Tar-Based Sealants	Nationwide Distribution	Ace Hardware, Do It Best, Lowe's, The Home Depot, True Value, United Hardware	
	Regional Distribution	Agway, Menards	

*sources: combination of Google searches, accessing Coal Tar Free America Blog (http://coaltarfreeamerica.blogspot.com/p/cts-bans.html), personal interviews, evaluating Material Safety Data Sheets for sealant products, and in-store visits conducted by Judy L. Crane, Ph.D.

**source: 2010 Census Interactive Population Search webpage (http://2010.census.gov/2010census/popmap/).

†most state Departments of Transportation no longer use coal tar-based sealants (AASHTO 2011; http://environment.transportation.org/pdf/communities_of_practice/stormwatercopjan2011.pdf)

DISCLAIMER: This table was originally prepared by Judy Crane, Ph.D. to support a feature article in *Environmental Science and Technology* on "Coal-tar-based pavement sealcoat and PAHs: Implications for the environment, human health, and stormwater management" (Mahler et al. 2012). Due to the difficulty involved with tracking restricted use jurisdictions and government use restrictions of coal tar-based sealants, this information may not be updated on a routine basis. In addition, municipalities in Minnesota are not required to inform the Minnesota Pollution Control Agency of ordinances they pass.

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Testimony of Bob Hoyt, Director
Department of Environmental Protection
On behalf of County Executive Isiah Leggett
Regarding Bill 21-12, Erosion, Sediment Control and Stormwater Management - Coal Tar Pavement Products

July 17, 2012

Good afternoon. My name is Bob Hoyt, Director of the Department of Environmental Protection (DEP). Thank you for the opportunity to testify on behalf of the County Executive in support of Bill 21-12, which would ban the sale or use of pavement products containing coal tar.

Coal tar sealants have become an environmental and economic concern to DEP within the past year. DEP is responsible for the structural maintenance of stormwater management ponds that have been transferred into our water quality protection charge program. Often the structural maintenance that we perform includes dredging the sediment that has accumulated at the bottom of the ponds.

In preparation for dredging, we recently sampled the sediment from Lake Whetstone. The results revealed that the sediment contained polycyclic aromatic hydrocarbon (PAH) above the State's standard for restoring contaminated properties for residential use. DEP also sampled the sediment from another stormwater pond, Gunners Lake, and found levels of PAHs above the standard, as well.

The PAHs in the sediment that we are finding are not at levels that present immediate human health risks, but they do present environmental problems and substantially increase the costs of managing the sediment after it is dredged. DEP is working with the Maryland Department of the Environment to identify acceptable disposal options, but we can no longer treat the sediment as clean fill. Preliminary estimates indicate that PAH contaminated soil may increase disposal costs by approximately 300 percent over clean fill. For Gunners Lake and Lake Whetstone the increased cost could well be $660,000 (14,200 cubic yards) and $460,000 (10,000 cubic yards), respectively.

Given the expense that PAH contamination adds to our program costs, we looked to identify the source of this contamination. A variety of studies, most notably those conducted by the U.S. Geological Survey (USGS), have identified coal tar-based pavement sealants as "a major source of PAH contamination in urban areas." USGS studies indicate that coal tar-based sealants are a large source of PAH contamination in urban lakes, and that residences adjacent to parking lots with coal tar-based sealants have PAH concentrations in house dust that are 25 times higher than those in house dust in residences adjacent to parking lots without coal-tar based sealants. Studies from Austin, Texas (which banned coal tar products in 2006) found that coal tar-based sealant products contain about 1,000 times more PAHs than sealant products with an asphalt base. It should also be noted that PAHs bound to sediments persist longer than PAHs in water or air. According to the U.S. Environmental Protection Agency, PAHs are problematic because several are toxic, carcinogenic, mutagenic, and/or teratogenic to aquatic life, and at least seven are probable human carcinogens.

The use of coal tar-based sealants has been banned in several jurisdictions around the country, including Washington, D.C., which enacted its ban in 2009. In response to concerns about the environmental effects of coal tar, major retailers like Home Depot and Lowe's no longer carry coal tar-based sealants. As a result, the majority of do-it-yourselfers are already using products that do not contain coal tar. Although some commercial applicators have continued to use coal tar-based products, there are asphalt based alternatives already available.

Thank you for the opportunity to testify on behalf of the County Executive in support of Bill 21-12. DEP is looking forward to continuing to work with Councilmember Rice and the other co-sponsors on this important issue. I would be happy to address any questions the Council may have.

ASPHALT & CONCRETE
MAINTENANCE, INC.
Preserve your Assets.
66 MURRAY PLACE YORK, PA 17403 P: 717-846-7908 F: 717-845-5161 WWW.TOTALASPHALT.COM

July 5 2012

Dear Councilmember,

Total Asphalt would like to object to Montgomery County's proposed ban on coal tar products (Bill 21-12). We are a regional contractor who does work in Montgomery County and throughout the state of Maryland. We use refined coal tar in our pavement rejuvenator, we are not, however, a coal tar emulsion.

Our product, Paverx, has been examined and determined not to be in violation of Federal air and water regulations, in fact it doesn't even qualify as a hazardous waste (see attached). Our product is a rejuvenator that drives the material into the pavement making it an integral part of the pavement. As our warranty states, it doesn't chip, flake, dust, peel or spall, eliminating any environmental concern.

We are in a different category than the traditional seal coats because of our product's chemical composition and how it reacts to existing asphalt.

Our product also has value to the broader concept of pavement preservation and leaves a significantly smaller carbon footprint than the creation and laying of new asphalt.

Pavement preservation requires fewer materials and less energy, and it creates less emissions and risk to the environment and the public.

The proposed ban would adversely impact our business and deny our clients in Montgomery County a cost-effective alternative to repaving, which they've been choosing to do for 20 years with Total Asphalt.

Given the high cost of liquid asphalt and the sustainable benefits of preservation, eliminating this choice for local property owners, both residential and commercial, is an expensive proposition that does nothing to address the very issue the council is now considering.

Additionally, The Council's proposed legislation seems precipitous given the legitimate debate over the science used to support this ban. At the very least, there are serious questions about methodology and selective results without proper context.

In fact, the alternative – an asphalt-based emulsion – has none of the durability or rejuvenating characteristics of our material. It will wear off the surface just as quickly as traditional seal coats. This recommendation again puts the council at odds with what appears to be its intent.

Our material is specified by the Federal Aviation Administration and a recognized by the Federal Highway Administration as a viable alternative to repaving and part of the broader picture of appropriate asset management and sustainability.

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In essence, please don't throw the baby out with the bath water.

Given the obvious harm it would cause my business, and for the reasons listed above, I ask that the Council reconsider its proposed ban and consider our rejuvenator as separate from coal-tar based seal coats and allow us to continue doing business in Montgomery County.

Sincerely,

Michael R. Leaman

Mike Leaman

President, Total Asphalt

30 Ivan Allen, Jr. Boulevard, Northwest • Suite 700 • Atlanta, GA 30308-3036 www.balch.com

BALCH
& BINGHAM LLP

RICHARD EDWARD GLAZE, JR.
ATTORNEY AT LAW
t (404)962-3566
f (866) 661-3268
e rglaze@balch.com

February 13, 2012

Mr. David Rigsbee
President/CEO
Chemtek/AeroGroup
Research Triangle Park, NC 27709

Re: Paverx Memorandum of Law

Dear David:

Attached is our memorandum of law regarding EPA regulation of Paverx. Thank you for the opportunity to be of service.

Sincerely,

Richard Edward Glaze, Jr.

REG/kp
Attachment
cc: Jim Hollis (w/attachment)

30 Ivan Allen, Jr. Boulevard, Northwest • Suite 700 • Atlanta, GA 30308-3036 www.balch.com

BALCH
& BINGHAM LLP

RICHARD EDWARD GLAZE, JR.
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MEMORANDUM

TO: David Rigsbee, President/CEO - Chemtek/AeroGroup

FROM: Richard Edward Glaze, Jr.

DATE: February 13, 2012

RE: Regulation of Paverx Pavement Protectant and Rejuvenator by EPA

Introduction. This memorandum evaluates whether Paverx Pavement Protectant and Rejuvenator, if applied in accordance with manufacturer recommendations, is regulated by environmental laws administered by the United States Environmental Protection Agency (EPA). In conducting the evaluation, we considered only whether the application of the product under routine conditions, in accordance with the manufacturer's specifications, would cause violations of the Federal environmental laws that would most likely apply to a chemical-based construction product like Paverx. We examined the Federal Water Pollution Control Act, 33 U.S.C. § 1251, *et seq.* (Clean Water Act or CWA); the Clean Air Act, 42 U.S.C. § 7401, *et seq.* (CAA); the Resource Conservation and Recovery Act, 42 U.S.C. § 6901, *et seq.* (RCRA); the release reporting requirements of the Comprehensive Environmental Response Compensation and Liability Act, 42 U.S.C. 6903(a) (CERCLA); and the Emergency Planning and Community Right-to-Know Act. 42 U.S.C. § 11004(a) (EPCRA). We express no opinion on state environmental laws, which vary from state to state. In conducting this evaluation, we reviewed the specifications for the Paverx product, the procedures for its application, and laboratory data showing concentrations of potentially hazardous constituents. We have not made an independent determination as to the accuracy of the manufacturer's specifications or the laboratory data, although we have not seen any evidence that they are inaccurate. Based on our review, and under the circumstances and subject to the caveats set forth below, it is our opinion that the application of Paverx in accordance with the manufacturer's specifications would not cause violations of the aforementioned statutes under normal conditions and under the current policies for the enforcement of and application of the statutes by EPA. A discussion of the relevant legal requirements and the rationale for our opinions are set forth below.

Paverx. Paverx is a product used to rejuvenate, seal and protect surfaces paved with asphalt, including airport runways and parking lots. Paverx is applied using an asphalt distributor that sprays the product onto asphalt surfaces at computer controlled rates of 0.05 to 0.075 gallons per square yard. Paverx is a coal-tar based product and, as such, contains low concentrations of certain chemicals commonly found in paving products including asphalt. The concentrations of the chemicals in Paverx, as applied, are much too low, however, to trigger regulation under laws administered by EPA.

The Clean Water Act. The Federal Water Pollution Control Act, commonly referred to as the Clean Water Act, provides civil and criminal penalties and injunctive relief for violations of its provisions. Potential violations include those related to discharges of pollutants, infractions of permit conditions, and falsifying monitoring data or methods. Of most concern when using a chemical product are prohibited discharges of the material or its byproducts into bodies of water. It is a violation of the CWA to discharge a "pollutant," such as a chemical substance, into jurisdictional waters via a "point source" without a permit for the discharge. Jurisdictional waters include most lakes, streams, creeks and wetlands. Other potentially regulated discharge violations include improper discharges into sewer systems in violation of standards for the protection of water treatment systems. Avoiding violations of the CWA requires an applicator of Paverx to prevent the discharge of the product or constituents of the product into jurisdictional waters. This includes taking care to not discharge the product or its constituents into storm drains and sewer systems that could result in discharges into jurisdictional waters. It is our understanding that, when applied according to the manufacturer's specifications, Paverx will not escape into surrounding waters, storm drains or sewer systems and will therefore not cause violations of the CWA.

Release Reporting Statutes. The primary release reporting statutes of CERCLA and EPCRA require a person who knows of a release of a "reportable quantity" of a "hazardous substance" "into the environment" to report the "release immediately." 42 U.S.C. § 9603. *See also* 42 U.S.C. § 11004(a). The term "environment" is defined under CERCLA to include "surface water, ground water, land surface or subsurface strata, or ambient air ...." 42 U.S.C. § 9601(8). A 'release' under CERCLA is "any spilling, leaking, pumping, pouring, emitting, emptying, discharging, injecting, escaping, leaching, dumping or disposing into the environment . . ." 42 U.S.C. §9601(22). The list of "hazardous substances" under CERCLA includes certain chemicals that are contained in Paverx in very low concentrations. When used correctly, Paverx is not being "released into the environment" as defined by CERCLA because the rejuvenator/sealant is applied directly to the surface of existing pavement and not released into water, land surface or subsurface strata. Paverx remains on the asphalt it is rejuvenating and hardens into a flexible surface, penetrating the asphalt only enough for proper rejuvenation and adhesion. A discharge of materials that remains entirely off of the ground, without releasing a reportable quantity of hazardous substance into the ground, air or water is not a "release" into the "environment" under CERCLA. Analytical testing has shown that the quantities of Paverx applied at any one time do not contain sufficient amounts of chemicals to constitute "reportable quantities" of hazardous substances even if their application were considered to be a "release into the environment."[1] It is our opinion that the proper application of Paverx does not constitute a reportable release under CERCLA or EPCRA.

Hazardous Wastes. At the federal level, hazardous wastes are regulated under RCRA. Generally speaking, to be regulated, a material must first be determined to be a "waste." To be considered a waste, a material must have been discarded. 40 C.F.R § 261.2(a)(1). Paverx as applied is not being discarded and is therefore not a waste. Moreover, analytical data show that even if disposed of, Paverx would not be regulated as a hazardous waste because it does not contain sufficient concentrations of chemicals, or have other characteristics, that would cause it to be considered "hazardous" under RCRA. *See generally*, 40 C.F.R. § 261, Subpart C.

Clean Air Act. The federal Clean Air Act is a statute that applies to numerous sources of air pollutants. Much of the CAA is directed at geographically - oriented sources of air pollution which by law are not regulated unless they emit sufficient quantities of certain pollutants.[2] Although the CAA *does* regulate "mobile sources," the mobile source statutes and regulations apply primarily to modes of transportation that emit pollutants, such as cars, trucks, and airplanes and would not apply to the application of Paverx. See 42 U.S.C. § 7521 – 7628. It is our opinion that the minor air emissions caused by the application of Paverx in accordance with the manufacturer's specifications, under normal conditions, would not trigger application of the provisions of the Federal Clean Air Act.

Caveats.

1. State laws. In virtually every State, the primary federal environmental laws are to some degree also enforced by state or local agencies that have been authorized to enforce the laws by EPA. Most states also have additional environmental laws, the scope of which can vary widely, and which could impose additional obligations on users of Paverx. This memorandum does not address separate state laws, which are not within the scope of this letter.

[1] One analytical report reviewed by the undersigned was of analysis conducted by Microbac Laboratories, Inc. of Wilson, N.C. on February 22, 2011.

[2] For example, CAA Title V permitting requirements apply to stationary sources that emit 10 tons per year or more of a single hazardous air pollutant or 25 tons per year or more of any combination of HAP's.

2. Use of this letter. While this memorandum constitutes our independent opinion on the matters expressly discussed, the memorandum was prepared for Chemtek, Inc., which is a client of Balch& Bingham LLP. The opinions expressed in this memorandum should not be considered, and are not intended, to constitute legal advice for any other entity or person. Nothing in this memorandum should be construed to create an attorney - client relationship between Balch & Bingham LLP and any entity or person. Balch & Bingham LLP disclaims any liability for reliance upon the opinions expressed herein and encourages all entities or persons to retain their own counsel to independently evaluate the use of Paverx under Federal and state environmental laws for the specific use intended, as well as other potentially applicable laws, rules or regulations that are beyond the scope of this memorandum.

3. Scope of advice. The opinions expressed in this memorandum are limited to those statutes expressly discussed. The advice contained herein is based on application of Paverx to an asphalt surface and is not intended to address all potential applications of Paverx beyond the scope of asphalt rejuvenation. We express no opinion regarding the use of Paverx in a manner not in accordance with the manufacturers instructions provided with the product. Moreover, we disclaim any responsibility for the misunderstanding or intentional misapplication of applicable laws, rules or regulations by government agencies or other persons or entities. Finally, this memorandum contains our opinions on the applicable Federal environmental laws as those laws currently exist and are interpreted. We disclaim any responsibility for new interpretations, rules or statutory changes, or additional regulatory requirements that could arise in the future. Any entity must ensure for itself that its application of Paverx complies with applicable Federal and state environmental laws for the specific application it employs at the time it is being applied.

Conclusions. It is our opinion that if Paverx, as currently formulated, is applied to existing asphalt surfaces in accordance with the manufacturer's recommendations, employing appropriate precautions to avoid the discharge of the material onto bare earth, vegetated areas or into water, it will not under normal circumstances cause violations of the current Federal Water Pollution Control Act, Clean Air Act, Resource Conservation and Recovery Act, or the release reporting requirements of the Comprehensive Environmental Response Compensation and Liability Act and the Emergency Planning and Community Right-to-Know Act.

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July 10, 2012

To Whom it May Concern:

I am the Regulatory Affairs Director for Chemtek, the manufacturer of PAVERX, which is used by Total Asphalt to rejuvenate pavements in Montgomery County Maryland.

The USGS has recently been studying the effects of coal tar sealers, and found pollution by a class of pollutants called PAHs in nearby streams and runoff from the pavement. However, the class of sealers they studied are very different from PaveRx, and the results will not be the same for the following reasons. The coal tar emulsion sealers they studied:

- were based on crude coal tar, which contains many more pollutants than the refined tar used for PaveRx,
- stay on the surface only,
- does not improve the flexibility of the pavement, causing it to continue to break/flake off,
- flake off from the pavement relatively easily, and therefore
- must be reapplied frequently.

In contrast, the properties of PaveRx are very different. PaveRx

- is based on refined coal tar, the processing of which removes the vast majority of the compounds of concern,
- soaks in to the pavement to significant depths, softening the asphalt and preventing evaporation,
- does not flake off, and
- needs only be reapplied every 5-7 years.

For this reasons, we ask that you not ban all coal tar products, only coal tar emulsion sealers.

Michael G. Kinnaird, Ph.D.

BILL 21-12

BD
cc

Marin, Sandra

From: Shawn Campbell [shawn.campbell@seaboardasphalt.com]
Sent: Tuesday, July 17, 2012 11:31 AM
To: Montgomery County Council
Subject: FW: Seaboard Asphalt opposes Bill 21-12
Attachments: Presentation to respond to Montgomery County Council Bill 21-12.doc

RECEIVED
2012 JUL 17 PM 4:23
069453

Shawn Campbell
Seaboard Asphalt Products Company
3601 Fairfield Road
Baltimore, MD 21226
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E sales@seaboardasphalt.com
W www.seaboardasphalt.com

From: Shawn Campbell [mailto:shawn.campbell@seaboardasphalt.com]
Sent: Tuesday, July 17, 2012 11:26 AM
To: county.council@montgomerycounty.md.gov
Subject: REF: Seaboard Asphalt opposes Bill 21-12

Montgomery County Council President:

I would like to submit the attached decent to Montgomery County Council Bill 21-12.

I would like to suggest the following outlined items for opposition:

1. United Steel Worker Union support for our opposition.
2. Loss of manufacturing jobs in Maryland
3. Loss of contracting jobs in Maryland
4. Language in Bill 21-12 is vague and could lead to unconstitutional searches and fines for Montgomery County citizens.

I would like to suggest that the Council table this legislation until further review with Industry where the science of the Bill and the language of its content can be reviewed co-operatively to benefit the Citizens of Montgomery County without undue harm to business and without constitutional violations.

If you have any questions, please do not hesitate to contact me.

Thank You,

Shawn Campbell
Seaboard Asphalt Products Company
3601 Fairfield Road
Baltimore, MD 21226
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F 410-355-5864
E sales@seaboardasphalt.com
W www.seaboardasphalt.com

My name is Shawn Campbell and I am the Vice President of Seaboard Asphalt.

Seaboard Asphalt is a coal tar manufacturing company located in Baltimore City within the Port of Baltimore and in an empowerment zone.

Seaboard Asphalt has been manufacturing at our current location since 1929 (over 80 years) and we oppose Montgomery County Council Bill 21-12 because it unfairly targets union manufacturing facilities. Seaboard Asphalt also opposes the language within Bill 21-12 can lead to significant additional costs to Montgomery County and burdensome fines and regulations on citizens as well as be unconstitutional where citizens are submitted to undue searches. This Bill by qualifying a violator of this proposed Bill under Section 19-68(b)(2) "is applied"---does this allow for Montgomery County to search and sample properties for violators and issue fines in accordance. Who is the regulating body; who qualifies the test results; who does the sampling?

Seaboard Asphalt is a union facility employing workers of the United Steel Worker Union Local 9477. Seaboard has the full support of the United Steel Workers Local 9477 in opposing this Bill. The United Steel Workers have pledged their support to defeat the incorporation of this legislation as written. This Bill and Amendments would force our company to shut down the production line for coal tar products and therefore directly leading to the layoff of more than half of our employees.

Several other applications currently have no alternative—for example---waster water treatment facilities; military applications and other Federal Specified applications. This Bill would force the production and jobs for this material out of Maryland to other States or overseas to supply material for these exemptions. This

Bill would directly take jobs away from the United Steel Workers Local 9477.

Seaboard Asphalt currently has 600 open accounts purchasing coal tar products in Maryland. Over half of these businesses are small minority businesses. Each of these facilities on average employee (5) people. This Bill would directly lead to the immediate loss of work of 3000+ people in the State of Maryland; if they could not use or purchase coal tar products. As well; these companies would not have the funds available to re-tool their application equipment to accommodate other products or use in other industries. This Bill would also directly and significantly affect the local companies that provide pallets, labels, pails and other products to Seaboard in support of our production needs for coal tar coatings.

I have reviewed the economic impact on our company. I have reviewed re-tooling our manufacturing facility to accommodate other products. I have determined the cost would exceed $2,000,000.00 and almost 5 years to accomplish. I have determined that it would be more cost effective for us to close our current facility in Maryland and re-locate to another State if this Bill is passed as is.

I would like to suggest that a committee should be formed to meet and work with industry to develop a plan; review the economic impact; review the science and re-write this legislation to reduce its impact on unions, minorities, small businesses and the citizens of Montgomery County.

KÆ K. A. E. PAVING CONSULTANTS, INC.

P. O. Box 1126
Wexford, PA 15090
(412) 721-9212
Fax: (724) 935-4367

kaepaving@consolidated.net

July 16, 2012

TO WHOM IT MAY CONCERN:

I am the President of K.A.E. Paving Consultants, Inc, a small business in the paving industry located in Pittsburgh PA . We supply products serving clients in Montgomery County. I write in opposition to Bill 21-12, Erosion, Sediment Control and Stormwater Management - Coal Tar Pavement Products. We are as concerned as you about the environment; but Bill 21-12 does not address any environmental problem that actually exists in the County so its only impact would be to cause economic harm to individuals and businesses and damage the County's reputation as a place to do business.

Our reasons for opposing Bill 21-12 are as follows:

1. Refined tar sealers have been commercially available for 75 years and have not been associated with any health or environmental problems in Montgomery County , in the State of Maryland, in the Chesapeake Bay watershed or anywhere else in the United States;

2. The proposed ban would not solve or even be a part of the solution of any existing problem in Montgomery, as no problems have been identified. It may open further problems associated with flat roofs, shingled roofs and sealing in-ground piping - all of which would be included in the sealing;

3. There is no alternative product. The US Army Corps of Engineers and the US Air Force have identified the usual proposed substitutes as materials that "harden the pavement" This includes asphalt emulsion sealcoats, the common proposed substitute. These cause the pavements to age prematurely, thus necessitating replacement more often. More PAH's are

created from quarrying, transporting stone, heating and blending asphalt and stone, transporting Hot Mix Asphalt (HMA) and laying the HMA. These alternatives cause more PAH's. Proper maintenance technologies would lower the creation of these PAH's.

Thank you for your consideration. Feel free to contact me if you need clarification on this issue.

Very truly yours,

Arthur J. McGovern

Arthur J. McGovern
President

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

To: Montgomery County Council, July 17, 2012

From: Dennis Barnes, on behalf of North Village Home Corporation (NVHC),
Montgomery Village, Md. 20886

As former NVHC President and 17 year elected Board member, there are a number of concerns regarding the proposed Bill, 21-12.

The impact of this approved bill would have very serious fiscal ramifications for the NVHC Board and the 950 HOA residents. The Board is the owner of all the roads in the HOA and is responsible for road maintenance including paving. Paving the entire network of community roads, based on private contractor services, requires a minimum $200,000 outlay.

The Board is confronting currently, significant resident fee delinquencies and foreclosures due to fiscal problems stemming from the economy which requires careful prioritizing of maintenance expense. Many of our residents earn a low income and some reside in Section 8, HOC subsidized town houses. The replacement of the coal tar process with asphalt based sealers while desirable would be extremely costly since such a change would require reapplication from 2-3 times. Unfortunately, the County has essentially eliminated roadway reimbursements for these paving investments. It should also be noted, that the lack of adequate county police service, due to their budgetary difficulties, has imposed on the Board the need for contract security personnel, an additional Board fiscal challenge.

The NVHC Board has a fiscal integrity responsibility regarding the governance of community property which involves maintenance of community esthetics, parks, and certainly the roads, to sustain property values and appearance. When the Board is faced with a shortfall of revenue, it must prioritize expenses. In the case of roads and paths, repair often must be delayed, consequently they fall into a state of disrepair. This situation obviously becomes exasperated if Bill 21-12 is approved. More research and analysis is needed on less expensive measures that can be applied if coal tar is to be eliminated which would than make it feasible for the NVHC Board to support this legislation.

In conclusion, please table this proposed bill and continue to study products that are better suited for HOA's facing economic challenges and until county reimbursements for road work can be initiated once again. Thank you.

Bill 21 – 12
Erosion, Sediment Control and Stormwater Management – Coal Tar Pavement Products
DHHS Comments, July 26, 2012

- I and staff from DHHS have reviewed the information submitted on the record for Bill 21-12 which would prohibit the use and sale of coal tar pavement products in the County and require enforcement by the Department of Environmental Protection. We have also reviewed 1) a 2011 EPA report, "Assessment of Water Quality of Runoff from Sealed Asphalt Surfaces" and 2) a 2009 Center for Disease Control/Agency for Toxic Substances and Disease Registry (CDC/ATSDR) study on Toxicity of Polycyclic Aromatic Hydrocarbons (PAHs). Please see copies attached.

- Based on preliminary review of these materials, we have concluded that DHHS does not have the in-house expertise or resources to provide an informed opinion on the level of health risk posed to individuals who may be exposed to polycyclic aromatic hydrocarbons (PAHs) found in coal-tar based sealants.

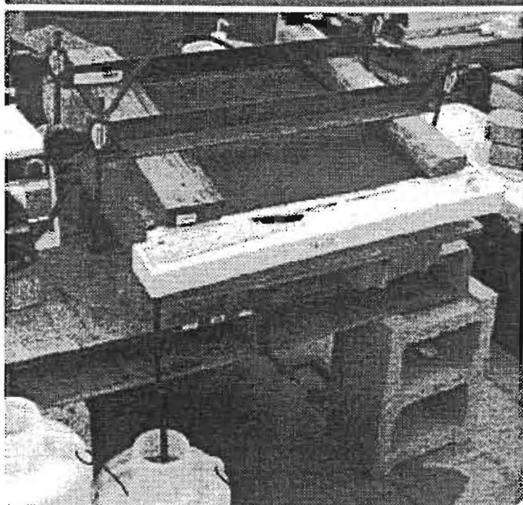
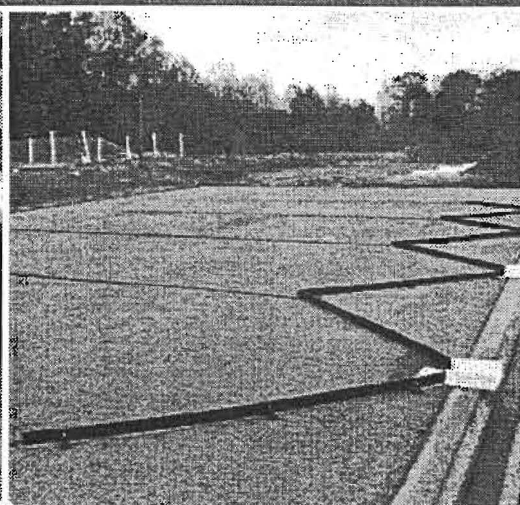
- The literature related to health impacts from PAH exposure that we were able to review from CDC/ATSDR cites that "[i]t is difficult to ascribe observed health effects in epidemiological studies to specific PAHs because most exposures are to PAH mixtures." Moreover CDC/ATSDR does not provide clear guidance on assessing the proportion of PAH concentrations in sediment that is the result of a specific known source of PAHs such as burning fuels, burning refuse, motor vehicle exhaust, pavement sealant, and cigarette smoke (CDC/ATSDR). In the absence of guidelines established by recognized regulatory authorities, our local health department acting on its own cannot provide the guidance requested by Council.

- DHHS defers to the expertise of DEP on water quality issues, the monitoring process, and the detection of PAHs in storm water management ponds. We also defer to their evaluation of the adverse environmental and fiscal impacts of the migration of PAHs into county stormwater management ponds.

EPA
United States
Environmental Protection
Agency

EPA/600/R-10/178 | September 2011 | www.epa.gov/nrmrl

Assessment of Water Quality of Runoff from Sealed Asphalt Surfaces


SCIENCE

Office of Research and Development
National Risk Management Research Laboratory - Water Supply and Water Resources Division

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Executive Summary

Background

Determining the impact of wet-weather discharges on receiving water quality remains an elusive goal given the various potential pollutants in the urban environment and the common practice for many storm drainage systems to discharge to the nearest receiving water with little or no treatment. In general, stormwater runoff from roads and parking lots has been shown to have high levels of pollutants and has been documented to be toxic to both freshwater and marine organisms. This document could be used to assist in the determination of the potential impact to receiving water quality from stormwater runoff in urban areas due to asphalt sealant use.

Sealants as Potential Sources of Water Quality Impairments

Asphalt pavement sealants are applied to parking lots and driveways to enhance appearance and protect surfaces and are widely used in commercial and residential products. Due to the tendency of these coatings to wear over time, manufacturers recommend reapplication of sealants to surfaces every two to three years. There are two types of sealcoats generally used in the U.S. today: asphalt emulsion and coal tar emulsion. Coal tar has been shown to have a detrimental effect on the overall health of a variety of aquatic organisms. Recent literature has suggested that coal tar-based asphalt sealants have impacted survival and development of amphibians, embryo and larval mortality in fish, and growth and biodiversity of macroinvertebrates and benthic phytoplankton. The primary components of coal tar that are presumably responsible for these toxic effects are polycyclic aromatic hydrocarbons (PAHs).

Why EPA?

This research project was conducted by the Water Supply and Water Resources Division of the Office of Research and Development's National Risk Management Research Laboratory. PARS Environmental Inc., an on-site contractor at EPA's Urban Watershed Research Facility (UWRF) in Edison, New Jersey, performed sampling, analysis and logistical support. The UWRF had an existing unsealed asphalt parking lot specifically designed to assist scientists and engineers in the collection of runoff. The parking lot was modified specifically for this project so that separate sections of runoff from test plots could be collected concurrently.

Analysis

The primary analyses conducted in this study were for the PAH content of the collected samples. A range of other water quality constituents were also measured. Toxicity analysis through the use of a Microtox® screening unit was also performed, though the results of these analyses were inconclusive due to matrix effects of the prepared samples. These toxicity results are therefore not reported.

Experimental Design

The project was initiated with the development of bench-scale testing. The project culminated with a full-scale, six-month study of three asphalt test plots with different or no surface treatments: coal tar emulsion sealant, asphalt emulsion sealant, and an unsealed control. Both the bench- and full-scale studies were tested over a time period of 1 to 30 days after application of sealants. The full-scale study had additional testing of test surfaces at six months.

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Results

The products examined in this study are a subset of the products available on the market and do not represent all products. Asphalt emulsion- and coal tar emulsion-based sealcoat products are the most widely used in the U.S. Coal tar products have PAH levels about 1,000 times higher than the asphalt sealcoat (Mahler et al., 2005). Precise national use is not known; however, USGS data suggest that asphalt-based sealcoat is more commonly used in the western U.S. and coal tar-based sealcoat use is more common in the other regions of the U.S. (Van Metre et al., 2005). There may be differences in water quality parameters observed in the runoff from surfaces of other sealants; therefore, the results herein cannot be translated across sealant product lines.

Results of the full-scale study indicate that PAHs are present in the runoff of surfaces coated with sealants. The PAH concentrations in the runoff were observed to decrease with time. When focusing on samples immediately after recommended curing time (24 hrs), there are correspondingly higher concentrations of PAHs. The asphalt emulsion and unsealed control surfaces did not contain concentrations of PAHs of the same order of magnitude as found in the runoff from the coal tar sealant plot.

Conclusions and Recommendations

PAHs were observed in the runoff from all three testing surfaces. The findings were consistent between the full-scale and the bench scale studies.

- The coal tar-sealed surfaces released 100 to 1000 times more PAHs to the runoff than the other surfaces. This release of PAHs from the surface to the runoff diminished with time. There was a measurable shift in the individual PAH components in the runoff, with fewer lower molecular weight PAHs observed in the runoff over time.

- The initial wetting after sealing may be the most crucial flush of PAHs into the environment.

- Additional testing is warranted on a representative variety of asphalt emulsion products. Even though low levels of PAH were observed in relation to the coal tar sealant runoff, increased total organic carbon (TOC) and chemical oxygen demand (COD) loadings were observed for the initial runoff samples collected, indicating an increased organic chemical load being released.

- Measurement of COD and TOC water quality parameters cannot be used as surrogates to identify potential release of PAHs from sealed surfaces.

- It is recommended that toxicity assays be performed with a variety of representative organisms (invertebrates, amphibians, fish, etc.) using standard procedures. This would require significant technical and financial resources. This more intensive toxicity testing is needed in order to more fully understand the effects of exposure to runoff from sealed asphalt surfaces. The literature lacks an in-depth study of sealant runoff examining both coal tar sealants or asphalt emulsion alternatives and the potential for acute toxicity, or lack thereof, to aquatic organisms in the water.

Agency for Toxic Substances and Disease Registry (ATSDR)
Case Studies in Environmental Medicine
Toxicity of Polycyclic Aromatic Hydrocarbons (PAHs)

Course: WB 1519
Original Date: July 1, 2009
Expiration Date: July 1, 2012

Table of Contents

Key Concepts	• Because of combustion of fossil fuels and organic waste, PAHs are ubiquitous in the environment • Studies show that certain PAH metabolites interact with DNA and are genotoxic, causing malignancies and heritable genetic damage in humans. • In humans, heavy occupational exposure to mixtures of PAHs entails a substantial risk of lung, skin, or bladder cancer.
About This and Other Case Studies in Environmental Medicine	This educational case study document is one in a series of self-instructional modules designed to increase the primary care provider's knowledge of hazardous substances in the environment and to promote the adoption of medical practices that aid in the evaluation and care of potentially exposed patients. The complete series of Case Studies in Environmental Medicine is located on the ATSDR Web site at http://www.atsdr.cdc.gov/csem/. In addition, the downloadable PDF version of this educational series and other environmental medicine materials provides content in an electronic, printable format, especially for those who may lack adequate Internet service.
How to Apply for and Receive Continuing Education Credit	See Internet address www.atsdr.cdc.gov/csem/conteduc.html for more information about continuing medical education credits, continuing nursing education credits, and other continuing education units.

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June 26, 2009

District Bans Coal-Tar Pavement Products

Media Contact: Alan Heymann (202) 741-2136

WASHINGTON, DC – Effective July 1, the District of Columbia has made it illegal to use, sell or permit the use of coal-tar pavement products. These products typically come in the form of pavement sealants and pavement dressing conditioners. Non-coal-tar alternatives are readily available. The purpose of the ban is to keep toxic chemicals in coal tar from poisoning local streams and threatening the Anacostia and Potomac Rivers and Chesapeake Bay.

"It's rare that we have a chance to knock out this kind of pollution in one fell swoop," said DDOE Director George S. Hawkins. "Our nation has made substantial progress, but now that we've discovered what's in coal tar and what it does, we have a rare opportunity to protect our waterways relatively easily."

Recent scientific studies have shown that concentrations of toxic polycyclic aromatic hydrocarbons (PAHs) in dust from parking lots sealed with coal-tar products are about 80 times higher than in dust from unsealed parking lots. Rain washes these toxic PAHs from coal-tar sealant off paved surfaces and into rivers and streams. Research suggests that total PAH loads washed off parking lots could be reduced by as much as 90 percent if parking lots were unsealed.

Dust from parking lots sealed with coal tar has more than 3 times the concentration of toxic PAHs as undiluted used motor oil, which has long been considered a leading urban source of PAHs. Other long-recognized urban sources of PAHs include tire particles, vehicle exhaust, and atmospheric deposition from sources like coal-fired power plants.

Property owners and contractors should avoid using products with listed ingredients including the words "coal," "tar," "refined coal tar pitch," or "RT-12." Ingredients should be listed either on the product container itself or on the Material Safety Data Sheet (MSDS) that should be available through both contractors and distributors.

The penalty for using, selling or allowing the use of coal-tar products is a fine of up to $2,500 per day. The coal-tar ban is part of the Comprehensive Stormwater Management Enhancement Amendment Act of 2008, which Mayor Adrian M. Fenty signed into law January 23, 2009.

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Chicago Tribune

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New doubts cast on safety of common driveway sealant

Recommend 18

Extremely high levels of toxic chemical in coal tar found in booming suburb

January 18, 2011 | By Michael Hawthorne, Tribune reporter

If a company dumped the black goop behind a factory, it would violate all sorts of environmental laws and face an expensive hazardous-waste cleanup.

But playgrounds, parking lots and driveways in many communities are coated every spring and summer with coal tar, a toxic byproduct of steelmaking that contains high levels of chemicals linked to cancer and other health problems.

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Nearly two decades after industry pressured the U.S. Environmental Protection Agency to exempt coal tar-based pavement sealants from anti-pollution laws, a growing number of government and academic studies are questioning the safety of the widely used products. Research shows that the tar steadily wears off and crumbles into contaminated dust that is tracked into houses and washed into lakes.

In Lake in the Hills, a fast-growing McHenry County suburb about 50 miles northwest of Chicago, researchers from the U.S. Geological Survey found that driveway dust was contaminated with extremely high levels of benzo(a)pyrene, one of the most toxic chemicals in coal tar. The amount was 5,300 times higher than the level that triggers an EPA Superfund cleanup at polluted industrial sites.

High levels also were detected in dust collected from parking lots and driveways in Austin, Texas; Detroit; Minneapolis; New Haven, Conn., and suburban Washington, D.C. By contrast, dramatically lower levels were found in Portland, Ore.; Salt Lake City and Seattle, Western cities where pavement sealants tend to be made with asphalt instead of coal tar.

The findings raise new concerns about potential health threats to people and aquatic life that went undetected for years.

"This is a real eye-opener, even for scientists who work frequently with these chemicals," said Barbara Mahler, a USGS researcher involved in the studies. "Such high concentrations usually are found at Superfund sites, but this could be your church parking lot or your school playground or even your own driveway."

About 85 million gallons of coal tar-based sealants are sold in the United States each year, according to industry estimates. There are no comprehensive figures on where it is applied, but in Lake in the Hills, researchers determined that 89 percent of the driveways are covered in coal tar.

Manufacturers promote the sealants as a way to extend the life of asphalt and brighten it every few years with a fresh black sheen. Contractors spread a mixture of coal tar, water and clay using squeegee

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machines and spray wands, or homeowners can do it themselves with 5-gallon buckets bought at hardware stores.

The makers of coal tar sealants acknowledge that the products contain high levels of benzo(a)pyrene and other toxic chemicals known collectively as polycyclic aromatic hydrocarbons, or PAHs. But they deny their products are responsible for the chemical contamination found in government studies, saying it could be coming from vehicle exhaust or factory emissions that travel long distances and eventually settle back to earth.

As more research identifies coal tar sealants as a top source of PAH-contaminated driveway dust and lake sediment, manufacturers have started to fund their own research to question the findings. Lobbyists also are offering contractors free admission to an upcoming seminar that promises to show them ways to "protect the industry," including a promotional DVD they can use to "help market sealcoating to your customers."

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"Nobody in our industry wants to hurt anybody," said Anne LeHuray, executive director of the Pavement Coatings Technology Council, an industry trade group. "The science is still evolving. If our products are a source, they are a very localized source."

The supply chain for the sealants begins at about two dozen factories, most of them around the Great Lakes or in western Pennsylvania, that bake coal into high-energy coke used in steel production. Companies figured out a century ago that much of the waste could be refined and sold to make other products, and they started adding it to pavement sealants after World War II.

One of the biggest suppliers is Koppers, a Pittsburgh-based company that processes coal tar at a plant in west suburban Stickney. The plant made about a third of the nation's refined coal tar in 2007, most of it used in aluminum production, according to an industry slide presentation. A company spokesman declined to comment.

Coal tar remains in widespread use even though its dangers have been known for centuries. During the late 1700s, many chimney sweeps exposed to tar in coal-heated London developed scrotal cancer, and decades later doctors determined that workers who coated railroad ties with tar-based creosote had high rates of skin cancer.

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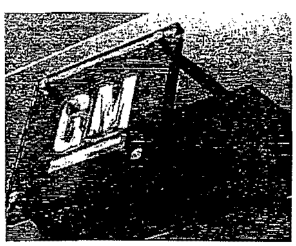

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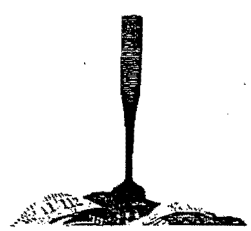

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Chicago Tribune
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Study points to dangers of children's exposure to coal tar sealants

Study comes amid attempts across U.S. to ban use on playgrounds, parking lots and driveways

February 15, 2012|By Michael Hawthorne, Tribune reporter

Children living next to driveways or parking lots coated with coal tar are exposed to significantly higher doses of cancer-causing chemicals than those living near untreated asphalt, according to a study that raises new questions about commonly used pavement sealants.

Researchers from Baylor University and theU.S. Geological Survey also found that children living near areas treated with coal tar-based sealants ingest twice as many polycyclic aromatic hydrocarbons, or PAHs, from contaminated dust tracked into their homes than they do from food.

The peer-reviewed study, and other new research documenting how coal tar sealants emit high levels of troublesome chemicals into the air, comes amid attempts by several cities in the Midwest, South and East to ban the products' use on playgrounds, parking lots and driveways. Some major retailers have pulled the products from their shelves, but coal tar sealants remain widely available elsewhere.

"There's been a long-held assumption that diet is the major source of exposure for children," said Peter Van Metre, a USGS scientist who co-authored the studies. "But it turns out that dust ingestion is a more significant pathway."

About 85 million gallons of coal tar-based sealants are sold in the United States every year, mostly in states east of the Continental Divide, according to industry estimates. The sealants, promoted as a way to extend the life of asphalt and brighten it every few years with a fresh black sheen, are sprayed by contractors or spread by homeowners using 5-gallon buckets.

During the past decade, studies have identified coal tar sealants as a major source of PAHs, toxic chemicals that can cause cancer and other health problems. Coal tar can contain as much as 50 percent PAHs by weight, substantially more than alternatives made with asphalt.

Anne LeHuray, executive director of the Pavement Coatings Technology Council, an industry trade group, said she was reviewing the new findings.

"It appears they have some other agenda here, which is to ban coal tar-based pavement sealants," she said of the government scientists.

LeHuray and other industry representatives have argued that vehicle exhaust, wood smoke and grilled hamburgers are more significant sources of the toxic chemicals than coal tar.

But the latest USGS research estimates that annual emissions of PAHs from the application of coal tar-based sealants exceed the amount from vehicle exhaust. Two hours after application, emissions were 30,000 times higher than those from unsealed pavement, one of the new studies found. Parking lots with 3- to 8-year-old sealant released 60 times more PAHs to the air than parking lots without sealant.

The studies are published in the scientific journals Chemosphere, Atmospheric Environment and Environmental Pollution.

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Lake County News-Sun
A Chicago Sun-Times Publication

Driveway sealers may be cancer time bombs

By Frank Abderholden

fabderholden@stmedianetwork.com

Last Modified: Jun 3, 2012 01:28AM

LIBERTYVILLE — The controversy over whether or not coal tar in driveway sealants are dangerous to your health and the environment came to Lake County on Tuesday at an informational meeting led by a McHenry County water resources manager.

Cassandra McKinney shared information on legislative initiatives in Illinois and across the nation to ban coal tar sealants used to give driveways and parking lots a fresh new sheen and lengthen the life span of the asphalt. Approximately 85 million gallons are used each year in the U.S..

McKinney said polycyclic aromatic hydrocarbons (PAHs) are formed by the incomplete combustion of organic materials. They are used in sealants for their binding properties, according to a sealant company owner who did not want his name used. He complains other sealants don't last.

There are actually thousands of different kinds of PAHs and the U.S. Environmental Protection Agency lists 16 as priority pollutants, chemical pollutants EPA regulates. Opponents of PAH bans point out human exposure to PAHs also comes from car exhaust, grilled meats and vegetables, wood fires, prairie burns, tobacco and other sources of combustion.

McKinney said that recent studies of the effect of coal tar sealant flakes on frogs in a lab showed that they took longer to hatch and were developmentally smaller. The study showed that at higher levels of the tar, the frogs died. Another study found high PAH in sediments were linked to mouth tumors in fish.

McKinney cited research that said in the past scientists assumed PAH exposure in children was due mainly to food. But a Baylor University study found children in an apartment complex next to parking lots treated with coal tar sealants were likely to receive twice as much PAHs from incidental ingestion of house dust than their diet. Those children ingested 14 times as much as children living next to unsealed lots.

While car exhaust is a big producer of PAHs, the U.S. Geological Survey recently stated in a scientific journals Chemosphere and Atmospheric Environment that coal tar sealants were emitting PAHs at rates that may be greater than the annual emissions from vehicles in the US.

The Agency for Toxic Substances and Disease Registry (ATSDR), a federal public health agency of the U.S. Department of Health and Human Services, says that PAHs are reasonably anticipated to be cancer causing to humans. It also noted they are used in medicines, dyes, plastics and pesticides.

It said while animal studies showed it harmed the immune and reproductive systems of mice, but the effects have not been seen in people. "PAHs can be harmful to your health under some circumstances," reads the public health statement on the compound. Exposure through breathing or skin contact for long periods to mixtures containing PAHs and other compounds could cause cancer to develop.

Anne LeHuray, executive director of the Pavement Coatings Technology Council, argues the seal coat industry is not the primary source of PAHs and shouldn't be made a scapegoat for PAHs in the environment. "No one is surprised that refined tar-based sealant contains PAHs. PAHs are everywhere in the environment," she said.

So where does that leave the consumer and government officials? Both Home Depot and Lowes stopped selling coal tar-based driveway sealant. Asphalt sealant still has coal tar, "but a significantly lower percentage," said McKinney — 50 milligrams per kilogram compared to 50,000 for coal tar sealants.

Federal legislation that would ban it is in committee and in Illinois there is a House bill in committee that is exploring changing the rules so that non-home rule communities could ban it.

McHenry County restricts its use and Lake County is looking at the issue, according to Michael Adams, senior biologist for the Health Department and Community Health Center.

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July 30, 2012

Councilmember Berliner,

I'd like to formally request that language recommended by staff for Bill 21-12 (subsection D) – application for a waiver – be written into the bill. (See T & E Packet for July 26 worksession, page 4)

Total Asphalt is a regional contractor who uses a "rejuvenator," not a seal coat, to preserve asphalt pavements. We have worked in Montgomery County for 20 years.

The product we use, PAVERX, while containing refined coal tar is not in the same chemical class as the coal tar emulsions or traditional seal coats that Bill 21-12 addresses.

This is a defining and important distinction: Our product does not migrate from the surface with evaporation and wear, but stays inert in the asphalt. Our product penetrates the pavement and becomes an integral part – a fusing if you will.

Without the ability to flake, chip, peel , dust or spall, migration into storm water and sediment is taken off the table as an issue, which has been Council's and the T&E committee's chief concern.

The language in subsection D states:

"The Director may waive the prohibitions of subsections b) and (c) for a product if the applicant for a waiver shows: that ordinary use of the product does not result in the immediate or eventual release of measurable quantities of polycyclic aromatic hydrocarbons into the air, water, ground, or sediments."

We are simply asking for this section to be written into the bill so that we have a chance to prove our case to the DEP and be given fair consideration.

Thank you for your time.

Sincerely,

Mike Leaman

President, Total Asphalt

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