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United States Senate
COMMITTEE ON ENVIRONMENT AND PUBLIC WORKS
WASHINGTON, DC 20510–6175

September 6, 2006

The Honorable Stephen L. Johnson
Administrator
U.S. Environmental Protection Agency
Ariel Rios Building
1200 Pennsylvania Avenue, NW
Washington, DC 20460

Dear Administrator Johnson:

We are writing to you regarding the results of recent collaborative studies conducted by the City of Austin, Texas, and the United States Geological Survey (USGS) that have identified coal-tar based sealcoat - the black, shiny surface often applied to asphalt pavement - as a significant and previously unrecognized source of extremely elevated concentrations of polycyclic aromatic hydrocarbons (PAHs) in streams. PAHs are suspected human carcinogens and are toxic to aquatic life. We believe that the findings of these studies have major implications for the City of Austin and the rest of the country because these sealants are used nationwide.

The studies show that runoff from parking lots sealed with coal tar-based sealant had PAH concentrations 65 times higher than concentrations in runoff from unsealed parking lots. In the greater Austin area, an estimated 660,000 gallons of coal-tar sealant are applied annually. PAHs are increasing in Town Lake sediments, which receives drainage from many of Austin's urban creeks. (The attached articles contain specific information about the levels of PAHs detected in selected Austin streams.) Biological studies conducted by the City indicate that these sealants are toxic to aquatic life at levels found in local waterways and are degrading the health of Austin's creeks, as indicated by a loss of species and decreased numbers of organisms. As a result of these findings, the City banned the use of coal-tar sealants in November 2005.

According to studies conducted by the USGS of 38 reservoirs and lakes sampled in 18 metropolitan areas across the nation from 1996 – 2001, PAHs in lakes and rivers are increasing. The biggest increases are in areas with watersheds affected by urban sprawl. For example, PAHs increased ten-fold in Lake in the Hills (suburban Chicago, Illinois) as the watershed rapidly developed. This information raises important local and national policy questions about the use of sealants and methods to prevent contaminated runoff from reaching urban water bodies.

In 1992, the EPA excluded coke product residues, including coal tar, from classifications as hazardous wastes if they are recycled. As a result, under the Resource Conservation Recovery

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Act (RCRA), these coal-tar sealants are considered products that contain recycled coal tar and are therefore not regulated. In the July 26, 1991 notice of proposed rulemaking (56 FR 35758) that discussed this recycling exemption for coke product residues that are blended with coal tar product that is sold, EPA stated: "For these reasons, the Agency believes that reinsertion of these residuals into coke ovens and mixing of these residuals with coal tar to be sold as a product are recycling practices that do not increase the levels of hazardous constituents in the final coke by-product, and therefore do not pose any significantly increased risk to human health and the environment." In light of the studies conducted by the USGS and the City of Austin that show that coal-tar based sealants contribute to PAH contamination in urban and suburban water bodies, we request that the EPA revisit this rulemaking and determine whether coke product residues blended with coal tar and sold as a product should be regulated under RCRA.

Additionally, we would like your responses to the following questions:

1. What action is the EPA taking to investigate the application of coal-tar based sealant to asphalt as a major source of PAH contamination in water bodies across the country?

2. Please specify the EPA's current strategy for controlling PAHs in urban environments. How will identification of this new source of PAH contamination influence future strategies?

3. How is the EPA working with its Regional Offices, the States, stormwater liaisons at the federal and local levels, and trade associations to advise communities about this research, its potential implications for aquatic wildlife, and the existence of safer sealant alternatives?

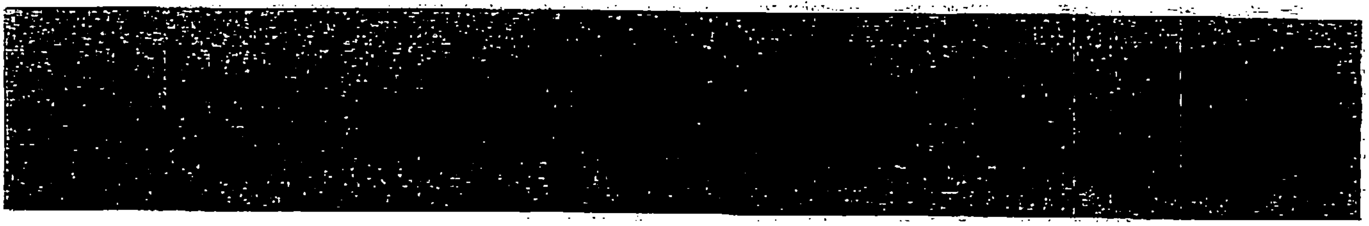
We urge the EPA to perform a national study and report its findings on how coal-tar based sealants increase PAHs in water bodies, and the effects of these PAHs on human health and the environment. We appreciate your immediate response to this matter.

Sincerely,

Jim Jeffords

John Warner

Attachments:
(1) The Coal Tar Facts, www.cityofaustin.org
(2) Van Metre, P.C.; Mahler, B.J.; 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.
(3) Parking-Lot Sealcoat: A Major Source of PAHs in Urban and Suburban Environments, U.S. Geological Survey, Congressional Briefing, December 2, 2005.



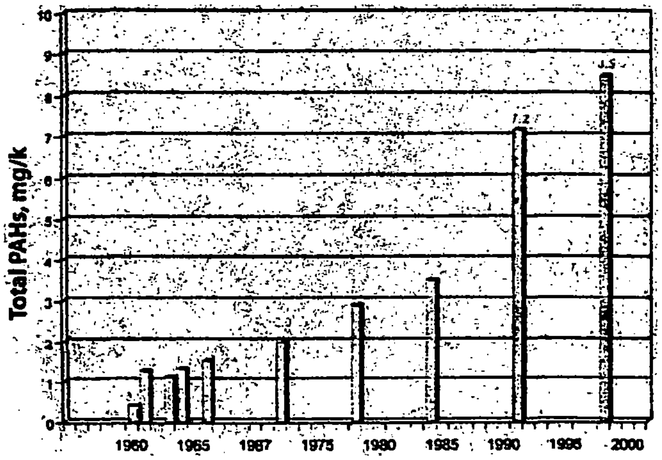
What are coal tar sealants?

Coal tar sealants are surface finishes for parking lots, driveways and airports. They contain varying concentrations of coal tar depending on product and formulation.

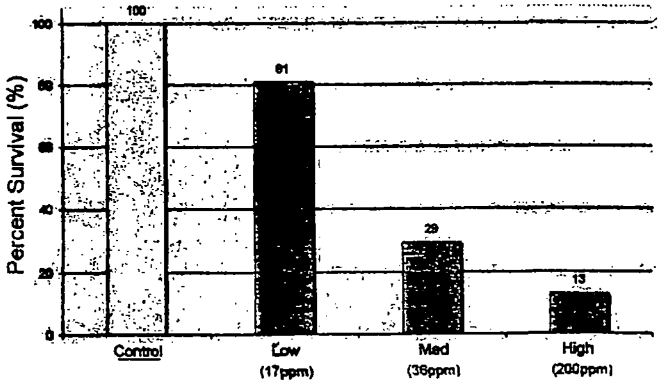
What's the problem?

Coal tar sealants contain extremely high levels of PAHs (Polycyclic Aromatic Hydrocarbons). PAHs are a group of chemicals formed during the incomplete burning of coal, gasoline, wood, garbage or other organic substances, such as tobacco and charbroiled meat. Because the sealants wear off the asphalt surface, recommendations call for reapplying them every two to three years. An estimated 660,000 gallons are applied annually in the Austin area. As the sealants wear off pavement surfaces, stormwater washes the particles into local waterways. PAHs are increasing in Town Lake sediments, where many of Austin's urban creeks drain to. City staff has documented that coal tar sealants are toxic to aquatic life at levels found in local waterways and are degrading the health of Austin's creeks.

PAH Trends in Town Lake Sediment (Core) Samples 1960 - 2000



Toxicity of Coal Tar Sealants in Sediments to Aquatic Organisms *(Hyallela azteca)* at three treatment levels (low, med, high)



What are the alternatives?

Asphalt based sealants are a comparable alternative offering a level of protection similar to coal tar sealants with lower levels of PAHs. Sealant products* that do not contain coal tar include:

RETAIL:

- Henry PM2000 Premium Driveway sealer/Filler
- Henry Elastomeric Emulsion Crack Filler

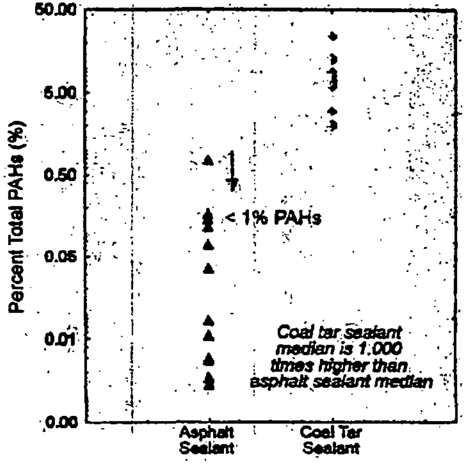
COMMERCIAL/WHOLESALE:

- Paveshield
- Jennite Asphalt Emulsion Pavement Sealer
- Gilsonite Asphalt Driveway Sealer

For updates see:
www.cityofaustin.org/watershed/coaltar_altproducts.htm

*There may be other sealant products available that do not contain coal tar so please read labels carefully. Listing of a specific product trade name does not constitute an endorsement of its use.

PAH Concentration in Asphalt Based and Coal Tar Based Sealants (Note: Percentages are on a log scale)



For additional information and data see
www.cityofaustin.org/watershed/coaltar_main.htm

Watershed Protection
Development Review

512-974-2550

Data provided by United States Geological Service

USGS
science for a changing world

Prepared in cooperation with the City of Austin, Texas

Parking-Lot Sealcoat: A Major Source of PAHs in Urban and Suburban Environments

Collaborative studies by the City of Austin and United States Geological Survey (USGS) have identified coal-tar based sealcoat—the black, shiny emulsion painted or sprayed on asphalt pavement such as parking lots—as a major and previously unrecognized source of polycyclic aromatic hydrocarbon (PAH) contamination in some Austin area stream sediments. PAHs are suspected human carcinogens and are toxic to aquatic life. The studies show that runoff from coal-tar based sealcoated parking lots has concentrations of PAHs that are about 65 times higher than concentrations in particles washed off parking lots that have not been sealcoated. Biological studies, conducted by the City of Austin in the field and in the laboratory, indicate that localized PAH levels in sediments contaminated with abraded sealcoat are toxic to aquatic life and are degrading aquatic communities, as indicated by loss of species and decreased numbers of organisms. Identification of this source may help to improve future strategies for controlling PAHs in urban water bodies across the Nation where parking lot sealcoat is used.

Sealcoat is used commercially and by homeowners across the Nation. It commonly is applied to parking lots associated with commercial businesses (including strip malls and shopping centers); apartment and condominium complexes; churches, schools, and business parks; and residential driveways. The City of Austin, Texas, estimates that about 600,000 gallons of sealcoat are applied every year in the greater Austin area.

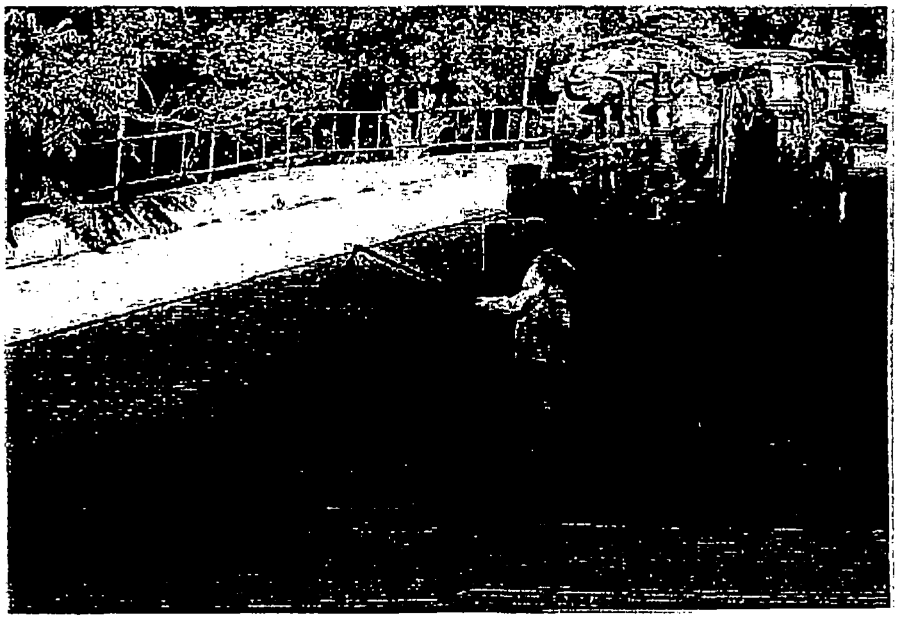
National use numbers are not available; however, use-patterns suggest that asphalt-based sealcoat is more commonly used on the West Coast and coal-tar based sealcoat is more commonly used in the Midwest, the South, and on the East Coast.

What are PAHs, coal tar, and sealcoat?

Polycyclic aromatic hydrocarbons (or PAHs) are a group of organic contaminants that form from the incomplete combustion of hydrocarbons, such as coal and gasoline. PAHs are an environmental concern because they are toxic to aquatic life and because several are suspected human carcinogens.

Coal tar is a byproduct of the coking of coal, and can contain 50 percent or more PAHs by weight.

Sealcoat is a black liquid that is sprayed or painted on asphalt pavement in an effort to protect and beautify the asphalt. Most sealcoat products are coal-tar or asphalt based. Many coal-tar sealcoat products contain as much as 30 percent coal tar by weight. Product analyses by the City of Austin indicated that coal-tar sealant products had median concentrations of total PAHs about 70 times higher than asphalt-based sealants.



Runoff from coal-tar based sealcoated parking lots has concentrations of PAHs that are about 65 times higher than concentrations in particles washed off parking lots that have not been sealcoated.

U.S. Department of the Interior
U.S. Geological Survey

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Prepared for a congressional briefing in Washington D.C. on December 2, 2005

How does sealcoat get from parking lots into the environment?

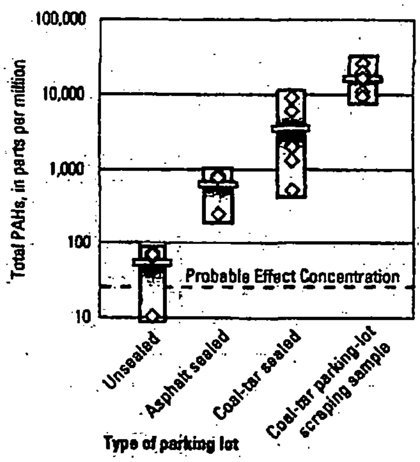
Vehicle tires abrade parking lot sealcoat into small pieces. These small particles are washed off parking lots by precipitation into storm sewers and streams. Sealcoat "wear and tear" is visible in high traffic areas within a few months after application. Sealcoat manufacturers recommend reapplication every 2 to 3 years.

What are environmental and human-health concerns associated with PAHs?

PAHs are toxic to mammals (including humans), birds, fish, amphibians, invertebrates, and plants. Possible effects of PAHs on aquatic invertebrates include inhibited reproduction, delayed emergence, sediment avoidance, and mortality, and possible adverse effects on fish include fin erosion, liver abnormalities, cataracts, and immune system impairments. PAHs tend to attach to sediments; the Probable Effect Concentration (PEC)—a widely used sediment-quality guideline that is the concentration of a contaminant in bed sediment expected to adversely affect benthic (or bottom-dwelling) biota—is 22.8 milligrams per kilogram (mg/kg) for total PAH. Studies by USGS and City of Austin did not evaluate human-health risk from exposure to sealcoat. Human-health risk from environmental contaminants is often evaluated in terms of exposure pathways. For example, people could potentially be exposed to PAHs in sealcoat through skin contact with abraded particles from parking lots, inhalation of wind-blown particles, and inhalation of fumes that volatilize from sealed parking lots. PAHs in streams and lakes rarely pose a human-health risk via drinking water because of their tendency to attach to sediment rather than dissolve in water. In addition, because PAHs do not readily bioaccumulate within the food chain, possible human-health risks associated with consumption of fish are low.

How did USGS study parking-lot runoff?

USGS researchers sampled runoff at 13 parking lots representing a range of different sealant types in Austin. They also took scraping-samples of parking lot surfaces to compare source materials to wash-off particulates. Source materials and wash-off particulates were analyzed for a suite of PAHs, major elements, and trace elements. USGS researchers sprayed water on four different types of parking-lot surfaces in Austin: lots sealed with coal-tar based sealcoat (top photo), lots sealed with asphalt-based sealcoat, unsealed asphalt lots, and unsealed concrete lots. The runoff was collected behind spill berms, pumped into containers (middle photo) and filtered through Teflon filters to collect the particulates for analysis (bottom photo). The particulates, the filtered water, and samples of sealcoat scraped from the parking-lot surfaces were analyzed for PAHs at the USGS National Water Quality Laboratory. Concentrations and yields (the amount of PAHs coming off each lot) were used to determine levels of contamination in runoff from each type of lot and the importance of sealed lots as a source of PAHs to urban streams.

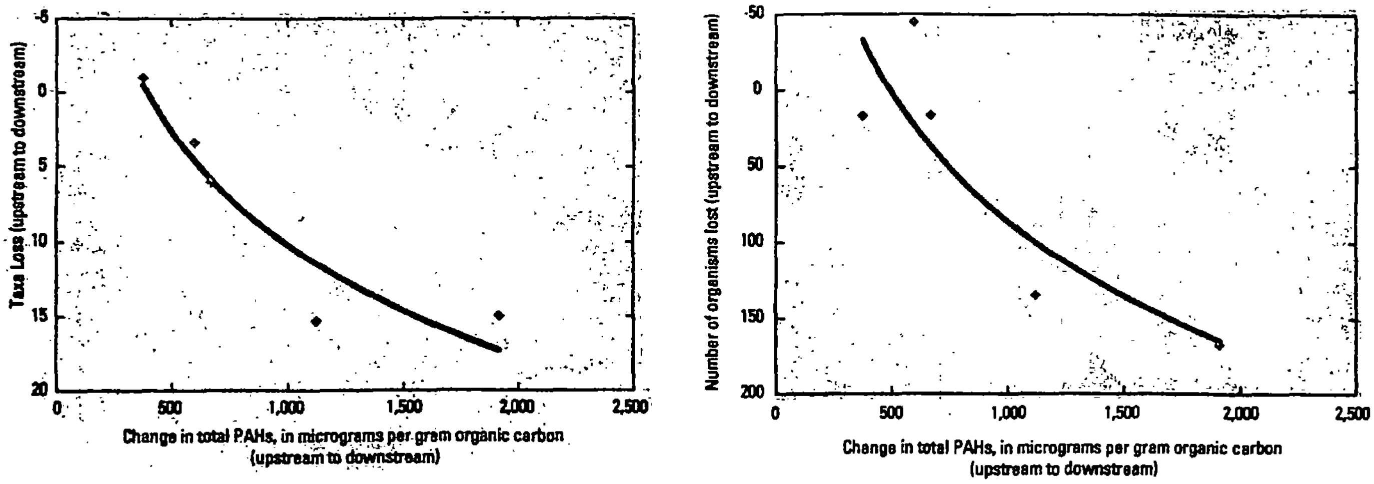


Concentrations of total PAHs in particulates in runoff from sealed parking lots greatly exceeded concentrations from unsealed parking lots. The bar on each graph is the mean concentration.

What concentrations of PAHs are in runoff from sealed and unsealed parking lots?

Concentrations of PAHs were much higher in runoff from parking lots sealed with coal-tar based sealcoat than from all other types of parking-lot surfaces. Specifically, the average concentration in runoff from coal-tar sealed lots was 3,500 mg/kg, about 65 times higher than the average concentration in particles washed off parking lots that were not sealcoated (54 mg/kg). The average concentration in particles washed off parking lots sealed with asphalt-based sealcoat was 620 mg/kg, about 6 times less than coal-tar based sealcoat, but still 10 times higher than the concentration from unsealed parking lots.

Concentrations of PAHs in particles washed off each of the different surface types—including the unsealed parking lots—exceeded the PEC of 22.8 mg/kg. This is not surprising because runoff from parking lots is likely to contain PAHs from many sources, including leaking motor oil, tire particles, vehicle exhaust, and atmospheric deposition. However, the large differences between concentrations associated with sealed and unsealed parking lots indicate that abraded sealcoat is a major and previously unrecognized contributor to PAH contamination in urban and suburban water bodies.



Field assessments in selected Austin streams showed loss of species (taxa) and decreases in the number of aquatic organisms downstream of coal-tar sealed parking lots that can be, in large part, explained by increases in total PAHs.

How do PAHs from sealcoat impact the quality and biology of streams?

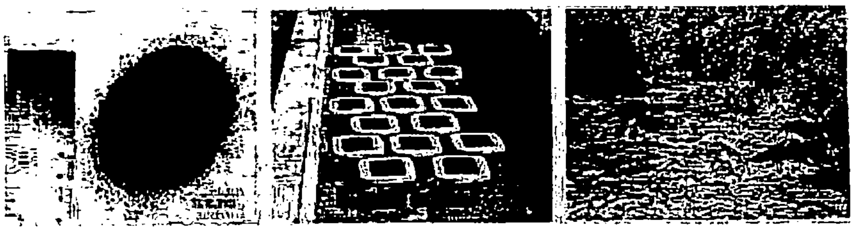
Studies by USGS scientists demonstrated possible connections between PAHs in particles washed from sealed parking lots and PAHs in suspended sediment in four streams in Austin and Fort Worth, Texas. Findings showed that PAHs in suspended sediments in the streams were chemically similar to those in runoff from parking lots sealed with coal-tar based sealcoat. Analysis of the total mass of PAHs expected to wash off sealed parking lots and the total mass of PAHs measured in suspended sediments in the four streams after rainstorms indicated that runoff from sealed parking lots could account for the majority of PAH loads to the streams.

Apart from the sealcoating itself, unsealed and sealed parking lots receive PAHs from the same urban sources—tire particles, leaking motor oil, vehicle exhaust, and atmospheric deposition—yet the average yield of PAHs from sealed parking lots was 50 times greater than that from unsealed lots. What would be the effect on PAH loading to the streams if parking lots were not sealed? Estimates from the USGS study indicate that total loads of PAHs coming from parking lots in the studied watersheds would be reduced to about one-tenth of their current loads if all of the parking lots were unsealed.

Studies by City of Austin biologists showed that sealcoat particles entering streams could be adversely affecting aquatic communities. Specifically, toxicity testing of organisms in the laboratory showed large increases in mortality as sealcoat amounts/concentrations were increased, and that coal tar sealants in sediments were toxic to aquatic life at PAH concentrations observed in Austin waterways. Controlled experiments that used aquariums with diverse natural biological communities showed significant biological degradation in response to sealcoat additions. Finally, field assessments in selected Austin streams showed loss of species and decreases in the number of aquatic organisms downstream from inflows of runoff from sealed parking lots. The impacts coincided with increases in concentrations of PAHs in stream sediments below sealed parking lots. Overall, City of Austin scientists have reported PAH contamination at levels predicted to be toxic to benthic invertebrates in over 13 percent of sampled Austin creeks.

How did City of Austin scientists conduct biological studies?

City of Austin biologists conducted laboratory and field studies to evaluate the effects of sealcoated parking lots on aquatic communities in area streams. These studies included toxicity testing in controlled laboratory experiments that exposed organisms to sediments spiked with coal-tar and asphalt-based sealcoat (left photo); controlled experiments that used aquariums with diverse natural biological communities to which sealcoat was added (middle photo); and field assessments of aquatic communities in streams upstream and downstream from inflows of runoff from sealed parking lots (right photo).



How do these findings apply to urban lakes and reservoirs?

PAHs in lakes and reservoirs across the Nation are increasing, as indicated by USGS studies of 38 reservoirs and lakes sampled in 18 metropolitan areas across the country from 1996 to 2001. Sediment cores (vertical tubes of mud) were collected from reservoir and lake bottoms (see photo below); analysis of these cores provides a reconstruction of historical water quality over time, much like using tree rings to reconstruct historical climate. Runoff carries soil, debris, and attached contaminants to lakes and reservoirs, which settle to the bottom; as the sediment builds up, changes in water quality are recorded in the successive sediment layers.

USGS findings show that concentrations of total PAHs in the majority of lakes and reservoirs in urban and suburban areas across the Nation increased significantly from 1970 to 2001. The increases were greatest in lakes with rapidly urbanizing watersheds (urban sprawl); for example, over the last 10 years PAHs increased tenfold in Lake in the Hills (suburban Chicago, Illinois) as the watershed rapidly developed. Further study is needed to assess direct links between the use of sealcoat and PAH trends in these lakes.

What are the implications of these studies?

The study of parking-lot surfaces by the USGS and the City of Austin show that abraded sealcoat could be a major source of PAHs to urban and suburban water bodies in watersheds where sealcoat is used. Such findings may have implications that extend beyond Texas as sealcoat is used nationwide. Identification of this major new source may influence future strategies for controlling PAHs in urban environments. In the past, sources of PAHs in urban watersheds were thought to be dominated by numerous nonpoint sources, such as leaking motor oil, tire wear, vehicular exhaust and atmospheric deposition. Such sources are difficult to quantify or control because of their diffuse, nonpoint nature. In contrast, sealed parking lots contribute to urban stormwater runoff (see photo below), and the use of sealcoat is voluntary and controllable. Possible alternatives to coal-tar based sealcoating of parking lots include the use of concrete and unsealed asphalt pavement and the use of asphalt-based sealants that contain lower levels of PAHs.

Currently, coal-tar based sealcoat is not federally regulated. In 1992, the U.S. Environmental Protection Agency (USEPA) excluded coke product residues, including coal tar, from classification as hazardous wastes if they are recycled. Under the Resource Conservation and Recovery Act, coal-tar based pavement sealants are products that contain recycled coal tar and, therefore, are not regulated. Further studies are needed to evaluate potential impacts on the aquatic environment in other parts of the country.

Contacts for additional information

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Austin, Texas 78754-4733

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Mateo Scoggins
City of Austin,
Watershed Protection and Development Review Department
505 Barton Springs Road, 11th Floor
Austin, Texas 78704

(512) 974-1917 or
mateo.scoggins@ci.austin.tx.us

Links to related publications, data and maps

City of Austin Coal Tar Sealant Information–
http://www.ci.austin.tx.us/watershed/bs_coaltar.htm

USGS frequently asked questions–
http://water.usgs.gov/nawqa/asphalt_sealers.html

Basic information on the toxicity of PAHs to biological organisms, U.S. Environmental Protection Agency (USEPA)–
http://www.epa.gov/R5Super/ecology/html/toxprofiles.htm#pahs

General information on PAH exposure, Agency for Toxic Substances and Disease Registry (ATSDR)–
http://www.atsdr.cdc.gov/toxprofiles/phs69.html

References

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 PAHs: Environmental Science and Technology, vol. 39, no. 15, p. 5560-5566.

Van Metre, P.C. and Mahler, B.J., 2005. Trends in hydrophobic organic contaminants in urban and reference lake sediments across the United States, 1970-2001: Environmental Science and Technology, vol. 39, no. 15, p. 5567-5574.

City of Austin. 2005. PAHs in Austin, Texas. (*http://www.ci.austin.tx.us/watershed/downloads/coaltar_draft_pah_study.pdf*)

August 1, 2005

ENVIRONMENTAL Science & Technology

http://pubs.acs.org/est

Parking Lot Sealcoat: *An Unrecognized* SOURCE *of* URBAN PAHs

Verifying Ballast-Water Treatment Performance

Microarray Analysis of Toxicogenomic Effects of Peracetic Acid on *P. aeruginosa*

PUBLISHED BY
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Environ. Sci. Technol. 2005, *39*, 5560−5566

Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons

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United States Geological Survey, 8027 Exchange Drive, Austin, Texas 78754, and City of Austin Watershed Protection Department, P.O. Box 1088, Austin, Texas 78767

Polycyclic aromatic hydrocarbons (PAHs) are a ubiquitous contaminant in urban environments. Although numerous sources of PAHs to urban runoff have been identified, their relative importance remains uncertain. We show that a previously unidentified source of urban PAHs, parking lot sealcoat, may dominate loading of PAHs to urban water bodies in the United States. Particles in runoff from parking lots with coal-tar emulsion sealcoat had mean concentrations of PAHs of 3500 mg/kg, 65 times higher than the mean concentration from unsealed asphalt and cement lots. Diagnostic ratios of individual PAHs indicating sources are similar for particles from coal-tar emulsion sealed lots and suspended sediment from four urban streams. Contaminant yields projected to the watershed scale for the four associated watersheds indicate that runoff from sealed parking lots could account for the majority of stream PAH loads.

Introduction

Concentrations of polycyclic aromatic hydrocarbons (PAHs)—a group of widely recognized aquatic contaminants (*1*) comprising numerous carcinogens (*2*)—have been increasing in recent decades in many urban lakes, particularly in areas undergoing rapid urban growth (*3*). PAHs adversely affect mammals (including humans), birds, fish, amphibians, invertebrates, and plants; in the aquatic environment, the effects of PAHs on invertebrates include inhibited reproduction, delayed emergence, sediment avoidance, and mortality, and the effects on fish include fin erosion, liver abnormalities, cataracts, and immune system impairments (*4*). Numerous sources of PAHs to urban runoff have been identified, including automobile exhaust, lubricating oils, gasoline, tire particles, erosion of street material, and atmospheric deposition (*5*−*8*), but uncertainty remains as to their relative importance. Investigations of urban sources of PAHs have thus far overlooked a potentially major source: parking lot sealants, also called "sealcoat". Our objective in this study was to evaluate the contribution of PAHs from sealed parking lots to urban streams.

In the United States and Canada, sealcoat is applied to many parking lots and driveways in an effort to protect the underlying asphalt pavement and enhance appearance. The two primary sealcoat materials on the market are refined coal-tar-pitch-based emulsion and asphalt-based emulsion. Although similar in appearance (glossy black), coal tar and asphalt have different molecular structures stemming from their origins: coal tar is a byproduct of the production of coke from coal, whereas asphalt is derived from the refining of crude petroleum. Coal tar, a known human carcinogen, is 50% or more PAHs by weight (*2*); the predominant constituents of asphalt are bitumens, complex mixtures of hydrocarbons that include asphaltenes, saturates, aromatics, and resins (*9*). Coal-tar-emulsion- and asphalt-emulsion-based sealcoats typically contain 20−35% of the emulsion.

Parking lot sealants are used extensively in the United States and Canada. Although national use figures are not available, the *Blue Book of Building and Construction* (*10*), a directory for the construction industry, lists over 3300 pavement sealant companies in 28 U.S. states. One company advertises the application of 1.7 billion liters to date worldwide (*11*), and another reports having sealed over 33 million square meters (*12*). The City of Austin, population 650000 (2000 census), estimates that about 2.5 million liters of sealcoat is used annually in this city (*13*).

Sealcoat abrades from the parking lot surface relatively rapidly, and reapplication is recommended every two to three years (*14*). In 2003, the City of Austin identified abraded parking lot sealcoat as a possible source of high concentrations of PAHs in streambed sediment (*15*). Here we present evidence suggesting that parking lot sealcoat could indeed be the dominant source of PAHs to watersheds with residential and commercial development.

Experimental Section

Sample Collection. We compared concentrations and yields of particulate PAHs in simulated runoff from parking lots sealed with coal-tar-based sealcoat, from lots sealed with asphalt-based sealcoat, and from unsealed asphalt and cement lots. Thirteen urban parking lots, representing a range of sealant types that are currently in use in Austin, TX, were sampled (Table 1). In addition, four test plots, each about 120 m^2, were sampled. Three of the test plots were sealed just prior to testing, and one was left unsealed (asphalt surface). The test plots are at the Robert Mueller Municipal Airport, Austin, TX, which has been closed since 1999. A full description of the sampling is given in ref *16*. In brief, 50 m^2 areas of each parking lot and the test plots were sprinkled with 2 mm of distilled water (100 L over a 50 m^2 area) to simulate a light rain, and concentrations of PAHs were analyzed in particles filtered from the runoff. The study focused on the particulate fraction, as PAHs in urban runoff, particularly those of higher molecular weight, are mostly associated with particulates (*7*, *17*); for selected samples (test plots and seven parking lots), the dissolved phase also was analyzed. The testing followed a minimum of 5 days with no rainfall. The parking lots were sampled once, and the test plots were sampled three times over a 6 week period. Water was sprayed from a plastic hand-held sprayer at a rate of about 7 L/min from a height of about 0.75 to 1 m. Spill berms were used at the down-slope end of the delineated area to gather water, which was then pumped into high-density polyethylene (HDPE) containers (Figure S1, Supporting Information). Recovery of water and observations about losses of water to wetting and leakage under the berms were noted. The water was returned to the laboratory, poured into a 50 L churn to keep the sample well mixed, and filtered through 0.45 μm pore size PTFE filters. The filters were

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† United States Geological Survey.

‡ City of Austin Watershed Protection Department.

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TABLE 1. Sampling Site Characteristics

site name	surface type	date of sealant application	sampling date	study component
$\mathrm{CT_{TP}1}$	coal-tar emulsion sealant	Aug 5–6, 2003	8/21/2003	test plot
$\mathrm{CT_{TP}2}$	coal-tar emulsion sealant	Aug 5–6, 2003	8/21/2003	test plot
$\mathrm{AS_{TP}}$	asphalt emulsion sealant	Aug 5–6, 2003	8/21/2003	test plot
$\mathrm{UNSAS_{TP}}$	unsealed asphalt pavement	Aug 5–6, 2003	8/21/2003	test plot
$\mathrm{CT_{TP}1}$	coal-tar emulsion sealant		9/9/2003	test plot
$\mathrm{CT_{TP}2}$	coal-tar emulsion sealant		9/9/2003	test plot
$\mathrm{AS_{TP}}$	asphalt emulsion sealant		9/9/2003	test plot
$\mathrm{UNSAS_{TP}}$	unsealed asphalt pavement		9/9/2003	test plot
$\mathrm{CT_{TP}1}$	coal-tar emulsion sealant		9/26/2003	test plot
$\mathrm{CT_{TP}2}$	coal-tar emulsion sealant		9/26/2003	test plot
$\mathrm{AS_{TP}}$	asphalt emulsion sealant		9/26/2003	test plot
$\mathrm{UNSAS_{TP}}$	unsealed asphalt pavement		9/26/2003	test plot
$\mathrm{AS_{PL}1}$	asphalt emulsion sealant	June 2003	9/7/2003	parking lot
$\mathrm{AS_{PL}2}$	asphalt emulsion sealant	June 2003	9/7/2003	parking lot
$\mathrm{AS_{PL}3}$	asphalt emulsion sealant	July 2003	9/28/2003	parking lot
$\mathrm{CT_{PL}1}$	coal-tar emulsion sealant	March 2003	9/7/2003	parking lot
$\mathrm{CT_{PL}2}$	coal-tar emulsion sealant	July 2003	9/28/2003	parking lot
$\mathrm{CT_{PL}3}$	coal-tar emulsion sealant	July 2003	9/28/2003	parking lot
$\mathrm{CT_{PL}4}$	coal-tar emulsion sealant	July 2003	9/30/2003	parking lot
$\mathrm{CT_{PL}5}$	coal-tar emulsion sealant	July 1999	9/30/2003	parking lot
$\mathrm{CT_{PL}6}$	coal-tar emulsion sealant	Nov 2000	9/30/2003	parking lot
$\mathrm{UNSAS_{PL}1}$	unsealed asphalt pavement		9/8/2003	parking lot
$\mathrm{UNSAS_{PL}2}$	unsealed asphalt pavement		9/30/2003	parking lot
$\mathrm{UNSCON_{PL}1}$	unsealed concrete pavement		9/8/2003	parking lot
$\mathrm{UNSCON_{PL}2}$	unsealed concrete pavement		9/8/2003	parking lot

massaged inside locking bags to remove retained particles, as described in ref *18*, and the recovered particulates were submitted as chilled slurries in clean glass vials to the U.S. Geological Survey National Water Quality Laboratory (NWQL) for analysis. In some cases the filtrate also was shipped, in chilled and clean amber glass bottles, to the NWQL for analysis of dissolved PAH. One or more samples of unfiltered water were collected from the churn for measurement of suspended sediment concentration (SSC), used to determine the mass of sediment recovered during each test. Although the 2 mm of simulated rain was not enough to wash off all of the mobile sediment, the recovered water was visibly clearer toward the end of each application. In samples from the five sites in which SSC was measured in the first 50 L and final 50 L of water, SSC decreased by a mean of 65% (range of 39–84%). We therefore assumed that the tests recovered most of the sediment that would be mobilized from the parking lot surfaces by a rain event, regardless of magnitude. Large, intense storms, however, likely would generate a higher yield of sediment.

The test plot and parking lot scrapings were obtained by scraping a small area (less than 0.25 m^2) with a metal paint scraper. The particulates removed were brushed onto a piece of new cardstock and then into a cleaned glass jar. The paint scraper was cleaned between sites, and a new brush was used at each site. Scrapings were examined by light and electron microscopy (Figure S2, Supporting Information), and submitted to the NWQL for PAH analysis.

Computation of Yields. Losses of water to wetting and losses of water and sediment leaking under the berms were estimated. Recovery of water ranged from 19 to 85 L with a median of 58 L. The lowest recoveries were from flat, unsealed asphalt lots, and the highest recoveries were from sealed lots and cement lots with gentle slopes. On the basis of recoveries and field observations, it was concluded that about 18 L of water was retained on the surface of sealed lots and cement lots and that the remainder of the water loss was a result of leakage past the berms. It was assumed that no yield of particles was associated with the water volume lost to surface wetting and that water leaking past the berms had the same SSC and contaminant levels as recovered water. For unsealed asphalt lots, the loss to wetting was estimated as 36 L for a maximum potential recovery of 64 L. Thus, to estimate the total yield of sediment from each lot, SSC was multiplied by the assumed maximum recovery (82 L for sealed and cement lots and 64 L for unsealed asphalt lots) to account for recovered water and leakage past the berms. Yields of PAH were estimated by multiplying the total yield of sediment times particle concentrations.

Chemical Analysis. Samples were prepared by extracting about 0.5 g dry weight of sample using pressurized solvent extraction at 120 and 200 °C with a mixture of water and isopropyl alcohol. The samples were extracted at each temperature at a pressure of 13800 kPa. Surrogate compounds were added to the sample prior to extraction to verify method recoveries. The extract was cleaned up using polystyrene divinylbenzene and Florisil solid-phase extraction cartridges. The extract was concentrated, solvent exchanged to ethyl acetate, and diluted to 10 mL. An internal standard mixture was added to an aliquot of the extract, and the extract was analyzed by full-scan gas chromatography/mass spectrometry (GC/MS). Difficult sample matrices were diluted before the full-scan analysis, and diluted surrogates were estimated in the samples.

Compound identifications were based on comparison of peak retention times and mass spectra to those of authentic standard compounds for the target compounds. Response factors were calculated for each compound from a set of calibration standards. Quantitation was carried out following the methods of Olson et al. (*19*). For PAHs in the particulate phase, the estimated method reporting limit (MRL) is 5 μg/kg for a 25 g sample. As less than 25 g was extracted, the MRL was raised accordingly, on a sample-by-sample basis. In some cases, MRLs were raised because of background interferences.

Dissolved-phase samples were analyzed following the method described by Fishman and Friedman (*20*), with the difference that continuous liquid–liquid extraction was substituted for use of the separatory funnel. In brief, 1 L samples fortified with surrogate compounds were extracted by continuous liquid–liquid extraction for 6 h under acidic and then basic conditions. Internal standards were added and sample extracts concentrated to 1 mL. Samples were analyzed by GC/MS in electron impact mode. Sample identifications were made by matching retention times and mass spectra with those of standard compounds. Quantitation involved use of internal standards and calibration curves generated by standard compounds of known amounts.

Quality control (QC) consisted of environmental and internal laboratory samples. Two duplicate environmental samples for particulate analysis of PAH were collected. For one of the sets of duplicates, ΣPAH differed by 8%; for the second (which had ΣPAH > 4000 mg/kg), ΣPAH differed by 54%. In the equipment blank analyzed for dissolved PAH, three parent PAHs were detected at concentrations about half that of the environmental sample with the lowest concentrations, and less than 1% that of the environmental sample with the highest concentrations.

Laboratory QC samples for particulate PAH analyses consisted of analysis of spiked samples, blanks, and samples of certified reference material (CRM). The median recovery for the six spiked samples was 76%. For the six laboratory blanks, an analyte was detected in 85 of 336 possible cases. The detected concentrations ranged from 0.1% to 3.5% of that in the environmental sample with the lowest concentration for that analyte. For the two analyses of CRM, the recoveries were within the NWQL-established acceptable range for 83% of the cases.

Three commercially available asphalt-based emulsion sealcoat products and six coal-tar-based emulsion sealcoat products were analyzed at DHL Analytical, Round Rock, TX, using EPA method SW 8270 (*21*). In each case, the product sample was taken directly from the container. Concentrations of 16 parent PAHs were determined. The sealants analyzed were not necessarily the same as those applied to the test plots or on the parking lots in use, although there was some overlap (product AS_PA was used on test plot AS_{TP}; product CT_PF was used on test plot $CT_{TP}2$) (Table 2).

Results

Concentrations and yields of total particulate PAH and total dissolved PAH in the runoff and total PAH in the scrapings were computed and compared between parking lot surface types (Table 2). The total particulate PAH (ΣPAH) concentration was computed for each sample as the sum of naphthalene, 2-methylnaphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[*a*]anthracene, chrysene, benzo[*a*]pyrene, and dibenz[*ah*]anthracene, which are the same as those used for the consensus-based sediment quality guidelines of MacDonald et al. (*22*). For unsealed parking lots (asphalt pavement and concrete combined), the mean ΣPAH was 54 mg/kg (range of 7.2–75 mg/kg), more than twice the probable effect concentration sediment quality guideline of 22.8 mg/kg (*22*) (Table 2), and in the range of those found by others in urban and roadway runoff (e.g. refs *23–25*). However, the mean ΣPAH concentration from the asphalt-sealed parking lots was more than 10 times higher (mean of 620 mg/kg, range of 250–830 mg/kg) than that from unsealed parking lots, and the mean ΣPAH concentration from the coal-tar-sealed parking lots was 65 times higher (mean of 3500 mg/kg, range of 520–9000 mg/kg) (Table 2; complete concentration data are given in ref *16*). ΣPAH concentrations in runoff from coal-tar-sealed lots were significantly higher than in runoff from other surface types (Kruskal–Wallis test of comparisons, hypothesis of no difference between groups rejected for $p < 0.05$). PAH concentrations from coal-tar-sealed lots also were much higher, in most cases by orders of magnitude, than PAH concentrations in other urban sources such as tire particles, motor oil, and weathered asphalt (Figure 1; note that this figure uses a different summation of PAH). ΣPAH concentrations in runoff from the sealed test plots were generally lower than those from the sealed parking lots, but the difference was not statistically significant, and concentrations from unsealed surfaces, with the exception of one outlier, were similar for test plots and parking lots.

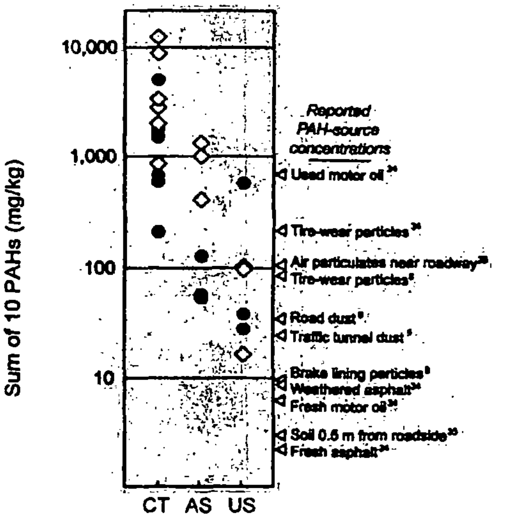

FIGURE 1. Sum of 10 PAHs (fluoranthene, pyrene, benz[*a*]anthracene, benzo[*a*]pyrene, benzo[*e*]pyrene, indeno[1,2,3-*cd*]pyrene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, and benzo[*ghi*]perylene) in particles in runoff from simulated rainfall on coal-tar emulsion sealcoat (CT), asphalt emulsion sealcoat (AS), and unsealed cement and asphalt (US) parking lots (◇) and test plots (●). Parking lots were sampled once, and test plots were sampled three times. Concentrations for other PAH sources reported in the literature also are indicated. These 10 PAHs were summed for this graph to facilitate comparison between experimental and reported concentrations.

Concentrations of ΣPAH in the scrapings ranged from 9500 to 83000 mg/kg for coal-tar-emulsion-sealed surfaces (including test plots) and from 110 to 2000 mg/kg for asphalt-emulsion-sealed surfaces (Table 2). Scrapings of two unsealed asphalt parking lots had ΣPAH concentrations of 7.1 and 20 mg/kg. Scrapings were observed under light and electron microscropy (Figure S2, Supporting Information).

Concentrations of total dissolved PAH (ΣPAH_{diss}, computed as the sum of the same PAHs as in ΣPAH excluding 2-methylnaphthalene; nondetections treated as zeros) for the test plots were about an order of magnitude greater in samples from the coal-tar-sealed test plots than concentrations in samples from the asphalt-sealed test plot, which in turn were about an order of magnitude greater than those from the unsealed test plot (Table 2). Nine of the 16 PAHs analyzed for were detected (complete data are in ref *16*). Higher weight PAHs—benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, indenopyrene, benzo[*ghi*]perylene, benz[*a*]anthracene, and dibenz[*ah*]anthracene—were not detected at laboratory reporting levels ranging from 1.7 to 3.4 μg/L. Four PAHs (acenaphthylene, acenaphthene, chrysene, and fluorene) were detected only in runoff from the coal-tar-sealed test plots; anthracene was detected in runoff from all the sealed test plots but not from the unsealed site. A similar suite of PAHs were detected at those parking lots for which the filtrate was analyzed (Table 2; complete data are in ref *16*).

Concentrations of ΣPAH in the commercially available sealant products and surface scrapings exceeded those of

TABLE 2. Concentrations of PAH in Washoff Samples, Scrapings, and Unapplied Sealcoat Product[a]

	washoff samples		scrapings	product	
	ΣPAH, mg/kg	ΣPAH_{diss}, µg/L	ΣPAH, mg/kg		ΣPAH(dry), mg/kg
		Test Plots			
$CT_{TP}1$, 8/12/03		21	83000	$CT_{P}A$	34000
$CT_{TP}1$, 8/21/03	1700	14		$CT_{P}B$	113000
$CT_{TP}1$, 9/9/03	530			$CT_{P}C$	202000
$CT_{TP}1$, 9/26/03	460	6.9		$CT_{P}D$	86000
				$CT_{P}E$	49000
$CT_{TP}2$, 8/12/03		11	11000	$CT_{P}F$	61000
$CT_{TP}2$, 8/21/03	1200	7.3			
$CT_{TP}2$, 9/9/03	4000			$AS_{P}A$	6600
$CT_{TP}2$, 9/26/03	140	3.8		$AS_{P}B$	1300
				$AS_{P}C$	300
AS_{TP}, 8/12/03		1.3	110		
AS_{TP}, 8/21/03	96	1.2			
AS_{TP}, 9/9/03	40				
AS_{TP}, 9/26/03	28	0.64			
$UNSAS_{TP}$, 8/12/03		0.16			
$UNSAS_{TP}$, 8/21/03	410	0.34			
$UNSAS_{TP}$, 9/9/03	25				
$UNSAS_{TP}$, 9/26/03	14	0.17			
		Parking Lots			
$CT_{PL}1$	2000	NA	25000		
$CT_{PL}2$	9000	5.4	15000		
$CT_{PL}3$	2000	7.1	11000		
$CT_{PL}4$	1300	12	9500		
$CT_{PL}5$	520	2.3	9900		
$CT_{PL}6$	5900	16	17000		
$AS_{PL}1$	250	NA	340		
$AS_{PL}2$	830	NA	2000		
$AS_{PL}3$	770	5.1	420		
$UNSCON_{PL}1$	75	NA	NA		
$UNSCON_{PL}2$	69	NA	NA		
$UNSAS_{PL}1$	64	NA	7.1		
$UNSAS_{PL}2$	7.2	0.24	20		

[a] Sums are as defined in the text. NA = not analyzed, CT = coal-tar-based emulsion, AS = asphalt-based emulsion, UNSAS = unsealed asphalt pavement, and UNSCON = unsealed concrete pavement.

the particulates in the washoff. Concentrations of ΣPAH in commercially available coal-tar-based sealcoat products ranged from 3.4 to 20 wt %, compared to 0.03 to 0.66 wt % for asphalt-based sealcoat products analyzed (Table 2; complete data in Table S1, Supporting Information).

Yields of ΣPAH (mass of ΣPAH per unit area of parking lot) computed for the simulated rainfall followed patterns similar to those of concentrations. Complete data for yields can be found in Table S2 in the Supporting Information. As with the concentrations, there was a wide range in the yields for a given surface type, in most cases more than an order of magnitude. The mean yield from coal-tar-sealed lots exceeded that from asphalt-sealed lots by more than a factor of 2, although this difference was not statistically significant (Kruskal–Wallis test of comparison, $p < 0.05$). However, the mean yield from sealed lots (asphalt and coal tar combined) exceeded that from unsealed lots by a factor of 50, and the difference was statistically significant.

Discussion

Runoff from parking lots typically is contaminated with PAHs from leaking motor oil, tire particles, vehicle exhaust, and atmospheric fallout, and it is not surprising that the mean concentration of ΣPAH in particles washed off each of the different surface types exceeded the probable effect sediment quality guideline. However, the large differences between concentrations for the different surface types suggest that abraded sealant products are a potentially important (and heretofore unrecognized) contributor to PAH contamination in urban and suburban water bodies.

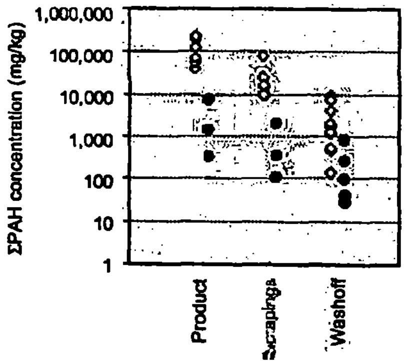

FIGURE 2. Comparison of ΣPAH concentrations in commercially available sealcoat products, scrapings from parking lots, and particles in washoff from parking lots for coal tar (◇) and asphalt (●) based sealants.

Comparison of Medium, Aging, and Vehicle Use on Concentrations and Yields. For both coal-tar- and asphalt-emulsion-based sealants, the ΣPAH concentration decreased from the unapplied sealant products to the scrapings to the washoff samples, as did the difference in concentration between the coal-tar-based and asphalt-based sealant samples (Figure 2). The difference in the median ΣPAH concentration between the coal-tar-based and asphalt-based sealants was 70-fold for the products analyzed and decreased to 40-fold for the scrapings and to a factor of about 8 for the washoff samples. Although the chemical changes between the product pre- and postapplication were not the focus of this study, the decrease in ΣPAH concentrations from the scrapings to the washoff particulates and the magnitude of the difference between the coal-tar-sealed lots and the asphalt-sealed lots can be attributed to dilution of abraded particles with less contaminated street dust and the greater abrasion of the asphalt-sealed compared to the coal-tar-sealed surfaces. A simple mass balance, assuming dilution of the coal tar scrapings (median ΣPAH concentration of 13000 mg/kg) by street dust (median ΣPAH concentration of 50 mg/kg) at a proportion of 1 part abraded particles to 7 parts street dust, results in the concentration found in the washoff. If the proportion of abraded particles is increased for the asphalt lots on the basis of the increased yields measured for asphalt-sealed lots (assuming that the greater median particle yield of 320 mg/m^2 from asphalt-sealed lots versus 200 mg/m^2 from coal-tar-sealed lots results from increased abrasion), the concentration found in the washoff from asphalt-sealed lots is well approximated.

The effect of aging of sealants on concentration over the short term (7 weeks) was evident at the test plots (Figure 3a). Overall, the concentration of ΣPAH and ΣPAH_{diss} in the washoff from each test plot decreased over the 7 week period following application. In one instance ($CT_{TP}2$, second sampling of washoff) ΣPAH exceeded that previously sampled, but in all cases the concentration at the end of the period was less than that at the beginning. The PAH assemblage changed over the same period as well, as represented by a comparison of higher molecular weight (MW) to lower MW PAHs. In the particulate samples the ratio of higher MW PAHs (represented by benzo[*a*]pyrene + chrysene) to the lower MW PAHs (represented by fluorene + phenanthrene; these two PAHs were chosen as they were detected in most of the samples) increased at all of the sealed test sites. As the lower MW PAHs are more volatile and soluble than the higher MW PAHs, volatilization and leaching of the lower MW PAHs

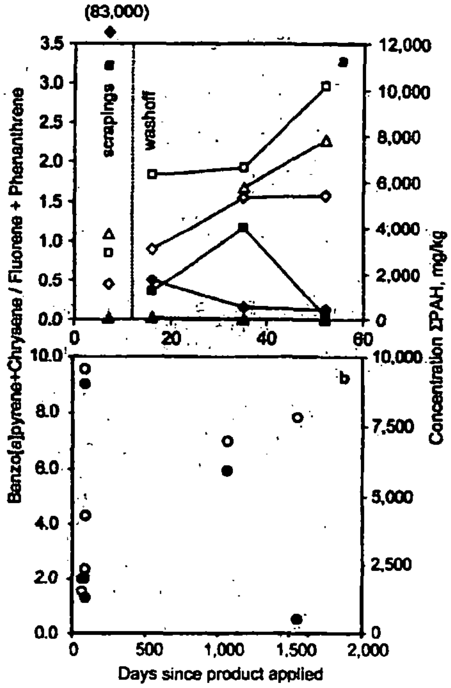

FIGURE 3. ΣPAH concentrations (closed symbols) and PAH ratios of higher and lower molecular weight PAHs (open symbols) (a) in scrapings and particles washed off coal-tar-sealed test plots (□, ◇) and an asphalt-sealed plot (△) and (b) as they relate to the age of coal-tar sealant in samples from parking lots in use.

from the newly applied sealant might be responsible for some of the decrease in concentration. For the parking lots, only coal-tar-sealed lots represented a range of ages, and for these lots there was no relationship between concentration or higher MW to lower MW PAH ratio and age of the sealant (Figure 3b). This might be because the parking lots were each sampled only once; the very wide range in PAH content between products, even those of a similar kind, may mask the effect of aging when time-series data are not available. Although the data are limited, they suggest that lots with older sealant tend to have a higher ratio of higher MW to lower MW PAHs, and that that ratio may reach a plateau after a period of time.

Comparison of the yields from the parking lots to those from the test plots, which receive no vehicle traffic, demonstrates the importance of abrasion of sealcoat by vehicles on ΣPAH yield: the mean ΣPAH yield was 20 and 160 times greater for the coal-tar-sealed and asphalt-sealed parking lots, respectively, than for the analogous test plots. This does not appear to be attributable to use patterns, although traffic counts were not made: the coal-tar-sealed lots are a mix of lots in constant use throughout the day (e.g., shopping center) and those with all-day parking (e.g., office), which are assumed to receive less use than those in constant use; all of the asphalt-sealed lots are all-day parking.

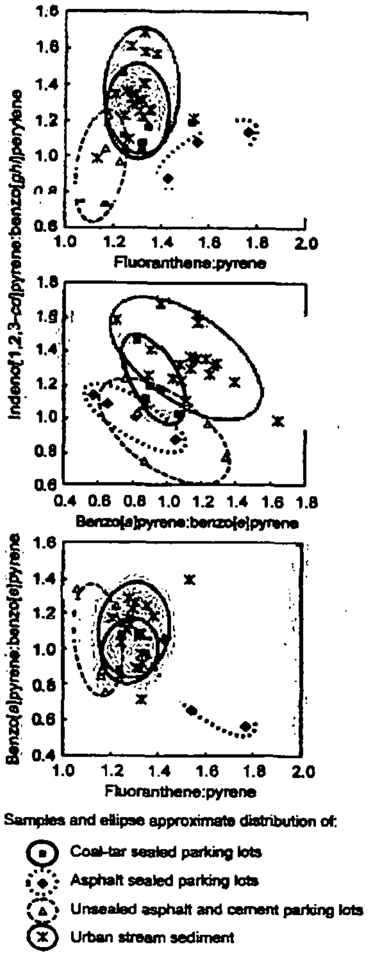

FIGURE 4. Comparison of indicator ratios of PAHs in particles washed from parking lots with coal-tar emulsion sealcoat, asphalt emulsion sealcoat, and unsealed asphalt pavement and concrete pavement, and in suspended sediment collected from four urban streams after storms.

Environmental Implications. Given the extremely elevated concentrations of PAHs in particles washed from sealed parking lots, how important is this contribution to the total mass of PAHs in urban streams? To answer this question, we compared the PAH assemblages and estimated PAH loads associated with particulates in parking lot runoff to those associated with suspended sediment collected during storm flow in four streams: Williamson Creek (Austin, TX) (*18*) and influent streams to Echo Lake, Fosdic Lake, and Lake Como (Fort Worth, TX) (*26*). These four streams are in highly urbanized watersheds (land use for the Austin watershed is about 65% urban, and for the three Fort Worth watersheds is more than 90% urban; full land use is given in Table S3, Supporting Information); the streams are ephemeral, and urban runoff is assumed to comprise a large component of storm flow.

PAHs comprise a large group of compounds, and PAH assemblage is often used to infer PAH sources (*27*). Differences in PAH assemblages can be investigated by computing the ratios of selected PAHs (*28, 29*). The best indicator ratios of coal tar as a PAH source have been identified as fluoranthene:pyrene, indeno[1,2,3-*cd*]pyrene:benzo[*ghi*]perylene, and benzo[*a*]pyrene:benzo[*e*]pyrene (*30, 31*). In graphs that combine these ratios, similarities and differences between parking lot and stream samples are apparent (Figure 4): ratios in the urban stream sediment group match those in runoff from coal-tar-sealed lots more closely than they do those from asphalt-sealed lots and from unsealed lots (asphalt and cement). We found these ratios were far more effective at distinguishing between the different parking lot samples and stream samples than ratios indicative of combustion versus noncombustion sources, or other approaches such as comparison of parent compound distribution (*32*). Although alkylated PAH homologues were analyzed (including C1–C5 homologues of the MW 128, 178, 202, 228, and 252 PAHs), their interpretation did not assist in discriminating between PAHs from the different parking lot surfaces.

The relative amount of similarity between groups of samples, as defined by the ratios, was quantified through

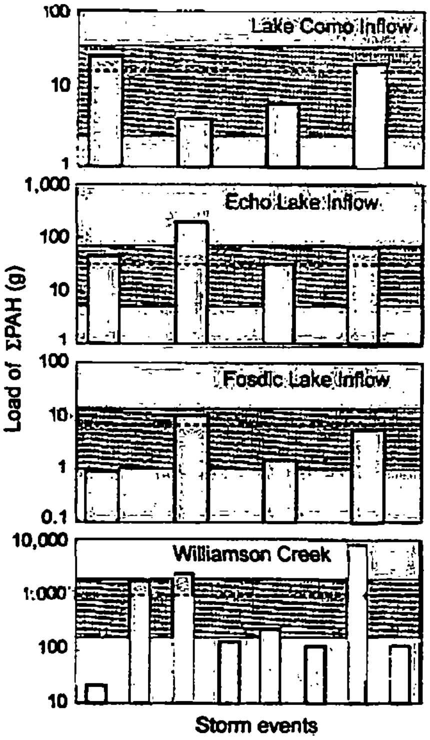

FIGURE 5. Comparison of estimated event loads of ΣPAH from sealed parking lots and measured instream storm-event loads for four urban watersheds. The interquartile range of estimated loads is shown in gray shading, on the basis of 25th and 75th percentile yields computed for sealed parking lots; the mean estimated load is indicated by a dashed line. Measured instream loads for four to eight individual events are shown as bars.

discriminant function analysis. In discriminant function analysis, each significant independent variable adds to discrimination between multiple groups. The three ratios (fluoranthene:pyrene, indeno[1,2,3-*cd*]pyrene:benzo[*ghi*]-perylene, and benzo[*a*]pyrene:benzo[*e*]pyrene) were entered into the analysis as the independent variables, with the different types of samples (coal-tar-emulsion-sealed lots, asphalt-emulsion-sealed lots, unsealed lots, and urban stormflow stream sediments) defining four groups of dependent variables. All three variables were shown to contribute significantly to discrimination between the groups ($p < 0.001$). The distances between the centroids of the groups were determined by computing the squared Mahalanobis distance, which is a measure of the distance between two points in the space defined by two or more correlated variables. The centroid of the group defined by the suspended sediment from urban streams is closest to the centroid of the coal-tar-based sealant group, next closest to that of the unsealed lot group, and farthest from that of the asphalt-based sealant group (squared Mahalanobis distances of 5.7, 13.0, and 25.0, respectively). Thus, on the basis of the three ratios diagnostic of coal-tar sources, the PAH assemblage of the suspended sediment from the urban streams most closely resembles that of the coal-tar-based sealant group, supporting the hypothesis that coal-tar-based sealants are an important source of PAHs in urban streams.

Moving to a mass-balance approach at the watershed scale for each of the four urban watersheds, we compared measured storm-event stream loads of ΣPAH to those estimated to be contributed by sealed parking lots. Digital land-use maps that included parking lots were provided by the Cities of Austin and Fort Worth and were updated using recent aerial photography and site inspections. Sealed and unsealed lots were identified by site inspection. We computed the hypothetical storm-event load generated by sealed parking lots in each watershed by multiplying the mean yield for sealed parking lots (coal-tar and asphalt emulsion sealcoat combined) determined from the runoff experiments by the sealed parking lot area of each watershed. We assumed that the 2 mm of water applied for the field tests mobilized all available particles, and that all runoff from parking lots entered storm sewers and was delivered to the stream. Although there is substantial variation in event loads for each stream (*18*, *26*), for all four watersheds the estimated ΣPAH loads contributed by sealed parking lots are similar in magnitude to measured stream loads, even though sealed parking lots cover only 1−2% of each watershed (Figure 5). These results might explain why an investigation carried out in Marquette, MI, found that runoff from commercial parking lots contributed 64% of the PAH load to the urban watershed studied (*33*).

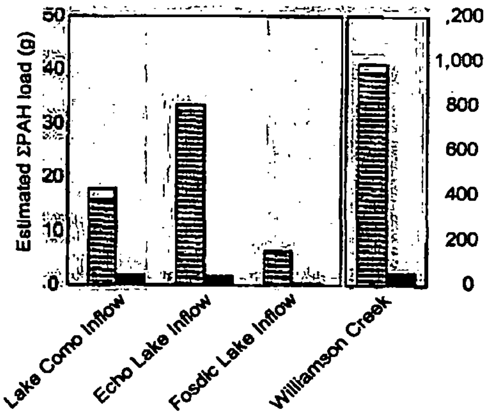

FIGURE 6. Comparison of event loads of ΣPAH for four urban watersheds estimated for parking lots in their current (2004) state (sealed by gray bars and unsealed by white bars) and projected loads if all existing parking lots were unsealed (black bars). Loads were estimated on the basis of the yields from the runoff experiments and the area of parking lots in each watershed.

What would be the effect on PAH loading to these watersheds if parking lots were not sealed? For each of the four watersheds, we compared the ΣPAH load contributed by parking lots (computed on the basis of the aerial extent of unsealed and sealed parking lots) to that obtained by applying the average yield for unsealed lots to all parking lots (Figure 6). We estimate that the ΣPAH load from parking lots in these watersheds would be reduced to 5−11% of the current loading if all lots were unsealed.

With the exception of the sealcoat itself, unsealed parking lots receive PAHs from the same urban sources as do sealed parking lots—e.g., tire particles, leaking motor oil, vehicle exhaust, atmospheric fallout—yet the average yield of PAHs from sealed parking lots is 50 times greater than that from unsealed lots. PAH assemblages and estimated loads further suggest that sealed parking lots could be dominating PAH loading in watersheds with commercial and residential land use. The implications of these results extend beyond Texas to the rest of the United States and Canada, where parking lot sealcoat is used extensively, and to other countries where sealcoat is being introduced. Previously identified urban sources of PAHs, such as automobile exhaust and atmospheric deposition, have been difficult to control or even quantify because of their nonpoint nature. In contrast, sealed parking lots are point sources, and use of the sealant is voluntary and controllable.

Acknowledgments

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Supporting Information Available

Two figures and three tables. This material is available free of charge via the Internet at http://pubs.acs.org.

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