PAHs in the Environment: Trends and Sources
Barbara Mahler and Peter Van Metre
U.S. Geological Survey
Springfield, Missouri, August 3, 2010
NAWQA – National Water Quality Assessment Program

- **STATUS** – characterize water quality nationally
- **TRENDS** – describe trends, or lack of trends
- **UNDERSTANDING** – identify and explain major factors controlling water quality
Paleolimnology

If it’s persistent and sticks to sediment ...

... we can see trends in sediment cores
Chemistry of PAHs

phenanthrene + naphthalene + pyrene = coronene + benzo[a]pyrene
PAHs are increasing in urban lakes


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Water Resources Discipline, United States Geological Survey, 8077 Exchange Drive, Austin, Texas 78754-4733

A shift in national policy toward stronger environmental protection began in the United States in about 1970. Conversely, urban land use, population, energy consumption, and vehicle use have increased since then. Thus, to assess the effects of these changes on water quality, the U.S. Geological Survey used sediment cores to reconstruct water-quality histories for urban and reference lakes across the United States. Cores were age-dated, and concentration profiles of polycyclic aromatic hydrocarbons (PAHs) and chlorinated hydrocarbons were tested statistically. Significant trends in total DDT, \( \mu \text{g} \text{ DDE} \), and total PCBs were all downward. Trends in chlordane were either steady between upward and downward, and trends in PCBs were mostly upward. Significant trends did not occur in about one half of cases tested. Concentrations of \( \mu \text{g} \text{ DDE} \), \( \mu \text{g} \text{ DDD} \), and PCBs were not as likely to exceed the probable effect concentration (PEC) at a sediment quality guideline, in sediments deposited in the 1980s as in 1985–1995, whereas PAHs were twice as likely to exceed the PEC in the more recently deposited sediments. Concentrations of all contaminants evaluated correlated strongly with urban land use. Upward trends in PAH concentrations, the strong association of PAH with urban settings, and rapid urbanization occurring in the United States suggest that PAHs could surpass chlorinated hydrocarbons in the threat they pose to aquatic biota in urban streams and lakes.

Federal environmental policy in the United States changed markedly in about 1970 with the establishment of the U.S. Environmental Protection Agency (1970) and the passage of the Clean Water Act (1972), the Safe Drinking Water Act (1974), the Toxic Substances Control Act (1976), and other laws (1). Improving water quality is one objective of these actions. Consequently, increases in urban land use and development, energy use, and vehicle use in the United States could lead to degradation of water quality. Identifying water-quality trends can provide measures of the success or failure of implementing the Clean Water Act and provide a warning of unacceptable degradation. Understanding trends also can improve our understanding of cause and effect relations between human activities and water quality and can suggest new development strategies for reducing adverse human effects on the environment.

The U.S. Geological Survey is using paleolimnology, the reconstruction of water-quality histories from age-dated sediment cores, to evaluate water-quality trends across the United States. Organic compounds that are chemically persistent and strongly hydrophobic are often preserved in the sediments, thus creating a record of historical water quality. Downward trends in polychlorinated biphenyls (PCBs) and DDT since the 1960s, for example, have been documented in a variety of environmental systems (1, 4). Trends in chlorinated and polycyclic aromatic hydrocarbons (PAHs) have been presented by many investigators but are more variable (5, 6). Many studies have addressed trends in these hydrophobic organic compounds (POCs); however, most are local in scale or focus on only a few water bodies in one region (e.g., 3–5, 7–9).

This study determined trends in persistent HOCS since 1970 using sediment cores collected from 38 lakes across the United States. The study was conducted by the U.S. Geological Survey National Water-Quality Assessment (NAWQA) Program (10). The primary objectives were to identify trends in HOCS in urban and undeveloped reference settings across the U.S. and, to the extent possible, to determine the causes of these trends. To our knowledge, this is the first attempt to apply a consistent paleolimnological approach to identifying trends in numerous HOCS across the United States.

Experimental Section

Study Design. Sediment cores were collected from 38 lakes in the United States between 1986 and 2001 (28 reservoirs and 10 natural lakes, hereafter referred to as lakes except where the distinction is relevant, age dated, analyzed chemically, and tested for trends as described in the following sections. Information for more information on lakes sampled). The NAWQA design provided a national framework for identifying potential study areas (10). The selection of urban areas in this study was based on the combination of NAWQA study units, metropolitan statistical areas, and other criteria (12). Urban areas were selected to represent a diversity of ecosystems where a majority of United States cities and urban populations are located. Lakes in or near the five most populous metropolitan areas in the United States (New York, Los Angeles, Chicago, Houston, and Dallas) were included in the study. Although it is not a probabilistic design, it does provide a geographically diverse coverage of United States cities.

Lakes were chosen for sampling on the basis of lake and watershed size, age of the lake (<40 years or more for reservoirs), and the amount and age of development of the watershed. The majority of lakes sampled have relatively small watersheds (7% of the watersheds sampled have an area of less than 100 km²), although drainage area to lake surface area ratios (DASAs) varied greatly, with generally larger ratios for urban lakes and smaller ratios for reference lakes. This bias was designed, with an objective in sampling the urban lakes being to represent historical trends in endogenous inputs to urban streams and an objective in the reference lakes being to represent historical trends in atmospheric deposition. Consistent inputs to lakes with well-developed basins and development in the watershed are typically dominated by lateral inputs from urban areas (13, 14). Lakes with small DASAs often have contaminant

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1. Federal Water Pollution Control Act (1972), the Safe Drinking Water Act (1974), the Toxic Substances Control Act (1976), and other laws (1).

2. U.S. Geological Survey, 8077 Exchange Drive, Austin, TX 78754-4733.


4. Environmental Science & Technology • VOL. 34, NO. 15, 2000

5. 5567–5574.

Upward trends in PAHs

Van Metre and Mahler, 2005, Environmental Sci. & Tech.
City of Austin monitors stream-bed sediment

- Extremely high (>1,500 mg/kg) PAHs in some small drainages
- Compare to Probable Effect Concentration (PEC) of 23 mg/kg
What are sources of PAHs?
### Sources of PAH: concentrations (mg/kg)

<table>
<thead>
<tr>
<th>Source</th>
<th>Concentration (mg/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tire wear particles</td>
<td>175 (av of 3 studies)</td>
</tr>
<tr>
<td>Road dust</td>
<td>59</td>
</tr>
<tr>
<td>Brake lining particles</td>
<td>9</td>
</tr>
<tr>
<td>Air particles, major roadway</td>
<td>104</td>
</tr>
<tr>
<td>Fresh asphalt</td>
<td>2</td>
</tr>
<tr>
<td>Weathered asphalt</td>
<td>9</td>
</tr>
<tr>
<td>Fresh motor oil</td>
<td>7</td>
</tr>
<tr>
<td>Used motor oil</td>
<td>726</td>
</tr>
<tr>
<td>Diesel engine particles</td>
<td>17.5</td>
</tr>
<tr>
<td>Gasoline engine particles</td>
<td>35</td>
</tr>
<tr>
<td>Coal-tar-based pavement sealcoat (average of 4 products)</td>
<td>92,000</td>
</tr>
</tbody>
</table>
Coal-tar sealcoat = Used motor oil
Clearing up the Confusion

- Coal-tar-based pavement sealer is made from high temperature coal-tar pitch.
- High temperature coal-tar pitch goes by many different names: Road Tar Emulsion Base, Road Tar, Refined Tar, RT-12 Emulsion Tar, Coal Tar Pitch, Pavement Sealer Base and RT-12.
COAL-TAR PITCH

CAS # 65996-93-2 = coal-tar pitch

## International Chemical Safety Cards

### Pitch

<table>
<thead>
<tr>
<th>TYPES OF HAZARD/EXPOSURE</th>
<th>ACUTE HAZARDS/SYMPTOMS</th>
<th>PREVENTION</th>
<th>FIRST AID/FIRE FIGHTING</th>
</tr>
</thead>
<tbody>
<tr>
<td>FIRE</td>
<td>Combustible</td>
<td>NO open flames</td>
<td>Foam, dry powder, carbon dioxide</td>
</tr>
<tr>
<td>EXPLOSION</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>INHALATION</td>
<td>Sneezing, Cough</td>
<td>Closed system and ventilation</td>
<td>Fresh air, rest</td>
</tr>
<tr>
<td></td>
<td>(See EFFECTS OF LONG-TERM OR REPEATED EXPOSURE)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>SKIN</td>
<td>MAY BE ABSORBED</td>
<td>Protective gloves, protective clothing</td>
<td>Rinse and then wash skin with water and soap</td>
</tr>
<tr>
<td></td>
<td>(Redness, burning sensation)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>EYES</td>
<td>Redness, Pain</td>
<td>Safety goggles, eye protection in combination with breathing protection</td>
<td>First rinse with plenty of water for several minutes (remove contact lenses if easily possible), then take to a doctor</td>
</tr>
<tr>
<td>INGESTION</td>
<td>See EFFECTS OF LONG-TERM OR REPEATED EXPOSURE</td>
<td>Do not eat, drink, or smoke during work. Wash hands before eating</td>
<td>Give plenty of water to drink. Refer for medical attention</td>
</tr>
</tbody>
</table>

### SPILLAGE DISPOSAL

Sweep spilled substance into sealable containers. Carefully collect remainder, then remove to safe place. Do NOT let this chemical enter the environment. (Extra personal protection: A/P2 filter respirator for organic vapour and harmful dust)

### STORAGE

Separated from strong oxidants. Separated from food and feedstuffs

### PACKAGING & LABELLING

Do not transport with food and feedstuffs.

**Note:** H, T symbol

**R:** 45

**S:** 53-45
MATERIAL SAFETY DATA SHEET

Manufacturer:

Emergency Phone No:

Information Phone No:

Date Of Preparation: July 18, 1996

Date Supersedes: April 19, 1994

SECTION I - IDENTIFICATION

Product Name: STAR SEAL - ASPHALT PAVEMENT SEALER

Chemical Family: Refined Coal Tar Pitch Emulsion

Chemical Name: Proprietary

Prepared by:

H.M.I.S

Health = 1

Fire = 1

Reactivity = 1

SECTION II - INGREDIENTS

<table>
<thead>
<tr>
<th>Ingredients</th>
<th>CAS NO.</th>
<th>WT%</th>
<th>Exposure Limits</th>
<th>OSHA PEL</th>
<th>ACGIH TLV</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Hazardous Ings</strong></td>
<td><strong>65996-93-2</strong></td>
<td><strong>29-32</strong></td>
<td>0.2 mg/m3 (Volatile)</td>
<td>0.2 mg/m3 (Volatile)</td>
<td></td>
</tr>
<tr>
<td>Coal Tar Pitch</td>
<td>65996-93-2</td>
<td>29-32</td>
<td>0.2 mg/m3 (Volatile)</td>
<td>0.2 mg/m3 (Volatile)</td>
<td></td>
</tr>
</tbody>
</table>

Listed in SARA Title III, Section 313- No.

STEML  - N/A

LC 50 - N/A

LD 50 - N/A

<table>
<thead>
<tr>
<th>Other Ings</th>
<th>CAS NO.</th>
<th>WT%</th>
<th>Exposure Limits</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clay</td>
<td>1332-58-7</td>
<td>18-20</td>
<td>N/A</td>
</tr>
</tbody>
</table>

STEL - 5 MG/M3 (DUST)

LC 50 - N/A

LD 50 - N/A

Water            | 7732-18-5 | 48-50| N/A             |

Listed in SARA Title III, Section 313 - No.

STEL - N/A

LC 50 - N/A

LD 50 - CTI OVER 320,000

* N/A = NOT AVAILABLE OR APPLICABLE

Total weight percentage of all the listed ingredients could be below 100, indicating other unlisted ingredients that are not considered hazardous by any federal (OSHA, WHMIS, SARA), any state or province or local Right-To-Know Regulations.
MATERIAL SAFETY DATA SHEET

Date last revised: 10-01-93

I. General Information

Chemical Name & Synonyms: Dispersion of refined coal tar and mineral fillers in water.
Trade Name & Synonyms: "federal" concentrate

Hazardous Materials Identification System (HMIS)

<table>
<thead>
<tr>
<th>HEALTH</th>
<th>FLAMMABILITY</th>
<th>REACTIVITY</th>
<th>PERSONAL PROTECTION</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>0</td>
<td>0</td>
<td>C</td>
</tr>
</tbody>
</table>

Proper DOT Shipping Name: None
DOT Hazard Classification: None

Manufacturer: [redacted]
Manufacturer's Phone #: [redacted]

II. Hazardous Ingredients

<table>
<thead>
<tr>
<th>Ingredient</th>
<th>CAS NO.</th>
<th>Percent</th>
<th>Exposure Limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Refined Coal Tar</td>
<td>65996-93-2</td>
<td>31 - 34</td>
<td>0.2 mg/m³ OSHA PEL, benzene soluble fraction 8 hr work shift avg.</td>
</tr>
</tbody>
</table>

III. Physical Data

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Boiling Point (°F)</td>
<td>IBP 212°F</td>
</tr>
<tr>
<td>Specific Gravity (H₂O=1)</td>
<td>1.2</td>
</tr>
<tr>
<td>Vapor Pressure (mm Hg.)</td>
<td>Not Determined</td>
</tr>
<tr>
<td>Percent Volatile by Volume</td>
<td>Not Determined</td>
</tr>
<tr>
<td>Vapor Density (Air =1)</td>
<td>&gt;1</td>
</tr>
<tr>
<td>Evaporation Rate (butyl acetate=1)</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Solubility in Water</td>
<td>Dispersible, not soluble.</td>
</tr>
<tr>
<td>pH</td>
<td>7.4</td>
</tr>
</tbody>
</table>

Appearance & Odor

Viscous brown black liquid with musky coal tar smell.

IV. Fire & Explosion Hazard Data

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flash Point (Test Method)</td>
<td>greater than 140°F (PMCC)</td>
</tr>
<tr>
<td>Auto Ignition Temperature</td>
<td>Not determined</td>
</tr>
<tr>
<td>Note: Product is a aqueous dispersion and does not support combustion.</td>
<td></td>
</tr>
<tr>
<td>Flammable Limits</td>
<td>LEL</td>
</tr>
<tr>
<td>Not Applicable.</td>
<td>UEL</td>
</tr>
</tbody>
</table>

Extinguishing Media

Water, chemical foam, CO₂, or dry chemical for dried film.

Special Fire Fighting Procedures
MATERIAL SAFETY DATA SHEET
Date Prepared: 03/16/2000

1. Chemical Product and Company Identification

Product Identifier: [Redacted]
Manufacturer: [Redacted]

Emergency Telephone Numbers
Emergency Contact Customer Service
Emergency Phone: 1-800-543-7077

2. Composition/Information on Ingredients

<table>
<thead>
<tr>
<th>COMPONENT</th>
<th>CAS#</th>
<th>% BY WT</th>
<th>TLV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Refined Coal Tar</td>
<td>68996-93-2</td>
<td>&lt; 34</td>
<td>0.2 mg/m³*</td>
</tr>
<tr>
<td>Ball Clay</td>
<td>1332-38-7</td>
<td>&lt; 30</td>
<td>0.1 mg/m³</td>
</tr>
</tbody>
</table>

* TWA, coal tar volatiles benzene soluble fraction, OSHA PEL
** Respirable Crystalline Quartz

3. Hazards Identification

Emergency Overview

Immediate concerns: Studies by Koppers Industries, Inc. (Using Refined Coal Tar Emulsion Safely, 1991) show that emissions during manufacture and application of coal tar emulsion are well below OSHA exposure limits. Avoid prolonged and repeated skin contact. Dermatitis may result from exposure of individuals with sensitive skin. Refined coal tar is a collection of organic compounds, primarily polynuclear aromatic hydrocarbons ranging from one ringed to 30 or 40 ringed in size. It is estimated that as many as 5,000 compounds may be present. Some of these polynuclear aromatic
Coal-tar pitch (by any name) is classified as a known human carcinogen.

What is the difference between crude coal tar and “refined” coal tar?

- The **fractional distillation** of crude coal tar yields light oil, middle oil, heavy oil, and anthracene oil; the **residue is called pitch**.

- On further distillation a large number of substances are obtained, about 200 of which have been isolated. They are used as dyes and in medicines. [Hutchinson Encyclopaedia]
The facts on benzo[a]pyrene and other B2 (carcinogenic) PAHs

- Coal-tar sealcoat products contain about 0.5% BaP (average of four products)
- One of 7 PAHs classified as probable human carcinogens ("B2 PAH")

benzo[a]pyrene, benz[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, chrysene, dibenzo[a,h]anthracene, and indeno[1,2,3-cd]pyrene
USGS - COA Joint Study

- Sample runoff from 13 parking lots
- Analyzed particles and water for PAHs
PAHs in Parking Lot Runoff Particles

Reilly Site, MN
3,000 max
Black River, OH
1,100

Superfund Sites

Applied over coal-tar-sealcoated pavement
PAHs in Parking Lot Runoff Particles

- **Used Oil**: 730 mg/kg
- **Tires**: 80-200 mg/kg
- **Asphalt**: 2-10 mg/kg
- **Other urban sources**

## Total PAH (mg/kg)

- **Unsealed**: 54
- **Asphalt sealed**: 620
- **Coal-tar sealed**: 3,500
- **Coal-tar scrape**: 15,000

PEC = 23
Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons

BARBARA J. MAHLER, *†
PETER C. VAN METRE, ‡
THOMAS J. BASHARA, ¶
JENNIFER T. WILSON, † AND DAVID A. JOHNS 

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)—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 650,000 (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 m², were sampled. Three of the test plots were sealed
9 U.S. Cities: Pavement Dust PAH (mg/kg)

Sealed
- Asphalt: 5.2, 2.1, <13, 1,200, 3,800, 570, 3,400, 1,300, 3,200
- Coal tar: <8.5

Unsealed
- Asphalt: <8.5, 0.8
- Coal tar: 24, 21, 47, 30, 54
PAHs Underfoot: Contaminated Dust from Coal-Tar Sealcoated Pavement is Widespread in the United States

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U.S. Geological Survey, Austin, Texas

Received July 29, 2008. Revised manuscript received September 22, 2008. Accepted September 24, 2008.

We reported in 2005 that runoff from parking lots treated with coal-tar-based sealcoat was a major source of polycyclic aromatic hydrocarbons (PAHs) to streams in Austin, Texas. Here we present new data from nine U.S. cities that show nationwide patterns in concentrations of PAHs associated with sealcoat. Dust was swept from parking lots in six cities in the central and eastern U.S., where coal-tar-based sealcoat dominates use, and three cities in the western U.S., where asphalt-based sealcoat dominates use. For six central and eastern cities, median ΣPAH concentrations in dust from sealed and unsealed pavement are 2200 and 27 mg/kg, respectively. For three western cities, median ΣPAH concentrations in dust from sealed and unsealed pavement are similar and very low (2.1 and 0.8 mg/kg, respectively). Lakes in the central and eastern cities where pavement was sampled have bottom sediments with higher PAH concentrations than do those in the western cities relative to degree of urbanization. Bottom-sediment PAH assemblages are similar to those of sealed pavement dust regionally, implicating coal-tar-based sealcoat as a PAH source to the central and eastern lakes studied. Recent studies have documented adverse biological effects in some Austin streams receiving runoff from coal-tar sealcoated lots (10), and demonstrated altered survival, growth, and development in a model amphibian species (Xenopus laevis) exposed to sediment spiked with coal-tar-based sealcoat (11).

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

High concentrations of PAHs in particles washed from coal-tar sealcoated parking lots in Austin raise two questions. First, are similarly high PAH concentrations associated with sealcoated pavement in other U.S. cities? Second, does use of coal-tar-based sealcoat lead to contamination of aquatic sediments? To answer these questions, the U.S. Geological Survey (USGS) collected dust from sealcoated and unsealcoated pavement in Austin and eight other U.S. cities; samples were collected in the watersheds of lakes sampled by the USGS National Water-Quality Assessment (NAWQA) Program.
23 ground-floor apartments
Ancillary Information Gathered including

- Smoking
- Incense/candles
- Fireplace use
- Type of stove/heat
- Shoe wear in house
- Indoor/outdoor pets
- Distance to major roadway
- Intensity of urbanization
Median total PAH $[\mu g/g]$

- **In-NCT (No coal-tar sealcoat)**
  - $n=12$
  - Median: 5.1
  - $\text{Out-NCT} = 9.0$

- **In-CT (Coal-tar sealcoat)**
  - $n=11$
  - Median: 129
  - $\text{Out-CT} = 4,760$

Equations:
- $\text{In-CT} = 25 \times \text{In-NCT}$
- $\text{Out-CT} = 530 \times \text{Out-NCT}$
How do pavement dust samples compare?

No coal-tar sealcoat (NCT)
- 15
- 49
- 11
- 4
- 6
- 42
- 8
- 2
- 3
- 17
- 10
- 1

MEDIAN = 9 µg/g

Coal-tar sealcoat (CT)
- 2,300
- 10,300
- 5,070
- 2,010
- 591
- 387
- 405
- 11,300
- 4,760
- 8,900
- 6,960

MEDIAN = 4,760 µg/g
Coal-Tar-Based Parking Lot Sealcoat: An Unrecognized Source of PAH to Settled House Dust

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Despite much speculation, the principal factors controlling concentrations of polycyclic aromatic hydrocarbons (PAH) in settled house dust (SHD) have not yet been identified. In response to recent reports that dust from pavement with coal-tar-based sealcoat contains extremely high concentrations of PAH, we measured PAH in SHD from 23 apartments and in dust from their associated parking lots, one-half of which had coal-tar-based sealcoat (CT). The median concentration of total PAH (T-PAH) in dust from CT parking lots (4760 µg/g, n = 11) was 530 times higher than that from parking lots with

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

A recently identified outdoor source of PAHs to the environment (16, 17)—coal-tar-based pavement sealcoat—has not been considered in any previous investigations of PAHs in SHD. Sealcoat is the black liquid that is sprayed or painted on the asphalt pavement of many parking lots, driveways, and playgrounds in the U.S. and Canada in an attempt to improve appearance and increase pavement longevity. There are two principal formulations of sealcoat: one with a refined coal-tar-emulsion (RT-12 grade) base and one with an asphalt-emulsion base. Coal tar is a known carcinogen that is more than 50% PAH by weight (18); sealcoat with a coal-tar base typically is 15 to 35% refined coal tar. The median PAH concentration (sum of 16 parent PAHs) for coal-tar-based and asphalt-based sealcoat products has been reported...
Increasing trends in PAHs

Van Metre and Mahler, 2005, Environmental Sci. & Tech.
“Contribution of PAHs from Coal-Tar Pavement Sealcoat and Other Sources to 40 U.S. Lakes” (Submitted to *Science of the Total Environment*).

- Start with source profiles and receptor profiles.
- CMB combines sources to best match the receptor profile.
- Results are the contribution of each source to each sediment sample.
Sources Considered

- **Coal combustion**
  - Power plant emissions
  - Residential heating
  - Coke oven

- **Vehicle related**
  - Diesel vehicle emissions
  - Gasoline vehicle emissions
  - Traffic tunnel air
  - Used motor oil
  - Tire particles
  - Asphalt

- **Fuel-oil combustion**

- **Wood burning**
  - Pine-wood soot particles

- **Coal-tar-sealcoat related**
  - NIST coal tar standard
  - Sealcoat products
  - Sealcoat scrapings
  - Sealcoat dust (average, 6 cities)
  - Sealcoat dust, Austin
PAH Source Apportionment to 40 U.S. Lakes

Mass PAH contribution from coal-tar-based sealcoat
PAH Trends in New Urban Lakes

BAL, Wash.

TWN, Tex.

LKH, Ill.

CYN, Calif.

ANN, Va.

KIL, Fla.

Coal-tar-based sealcoat
Vehicle-related sources
Oil combustion
Wood combustion
Coal combustion
All urban is not equal

- Tanasbrook Pond: 844 people/km, \( \Sigma \text{PAH} 1.34 \text{ mg/kg} \)
- Palmer Lake: 939 people/km, \( \Sigma \text{PAH} 34.1 \text{ mg/kg} \)
- Decker Lake: 2,090 people/km, \( \Sigma \text{PAH} 0.76 \text{ mg/kg} \)
- Lake Anne: 2,095 people/km, \( \Sigma \text{PAH} 17.0 \text{ mg/kg} \)
PAHs are increasing in urban lakes nationally.

Coal-tar-based sealcoat has exceptionally high PAH concentrations, particles are mobile, and use is extensive.

PAHs in house dust are elevated where coal-tar-based sealcoat is used.

Coal-tar-based sealcoat is the largest contributor of PAHs to urban lakes.