PAVEMENT COATINGS
TECHNOLOGY COUNCIL

PARKING LOT SEALANTS
AND
POLYCYCLIC AROMATIC HYDROCARBONS (PAHS)

Refined coal tar-based sealants and PAHs

- Coal tar is a by-product of the production of coke used in making iron and steel. Coal tar consists of a mixture of naturally occurring compounds containing hydrogen and carbon that are common to all organic fuels (coal-, petroleum-, wood-based fuels). Some of these hydrocarbons belong to a class of materials called polycyclic aromatic hydrocarbons (PAHs). More about PAHs later.
- Refined coal tar-based sealant emulsions are made from refined coal tar. The concentration of PAHs in the refined coal tar used to make sealants is less than that in unrefined coal tar. Historically, refined coal tar-based sealants have been more commonly used in regions of the country where coke plants are present.
- In addition to refined coal tar, refined coal tar-based sealant emulsions contain clay and emulsifiers (emulsifiers are ingredients used to bind together substances that don't normally mix, such as oil and water). Prior to application, the refined coal tar sealant emulsion is further diluted by addition of sand and water.
- The sources of PAHs in soils and sediments have been the subject of an enormous volume of research over the past three decades. Identification of sources and apportioning specific sources to specific occurrences of PAHs has developed into its own scientific discipline called "Environmental Forensics."
- One recent publication has suggested that refined coal tar-based sealants applied to parking lots may be a significant source of PAHs in Austin, Texas. This suggestion only can be evaluated in highly localized instances, and only when all local sources PAHs have been evaluated. Unfortunately, the authors of the Austin study did not evaluate any possible source other than parking lots, even though data exist that old industrial facilities that formerly operated next to Austin's streams and ponds are a source of the PAHs in sediments.
- A follow up study in Austin, Texas that looked at PAHs in sediments before and three years after a municipal ban on the use of refined coal tar-based sealants showed no discernable differences either in the sources or amounts of PAHs in local streams (Gauthier and DeMott, 2008; DeMott and others, submitted for publication to the Journal of Environmental Forensics).
- A study is in progress at the University of New Hampshire, where refined coal tar-based sealant was applied to test parking lot. The study to date demonstrates the importance of adhering to manufacturer's directions during sealant application. Refined coal tar-based sealants are an emulsion, which means that,

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like Jello®, the material must gel or cure. Manufacturers direct that sealants be applied when the weather is warm and no rain is in the forecast. Unfortunately, the sealant was applied to the University of New Hampshire test parking lot on a day when temperatures were not optimal and it rained within hours of application. Photographs taken by the researchers clearly show that the sealant did not cure properly. Nevertheless, data reported so far show that the washed off sealant was trapped near the point of application with surprising efficiency by vegetation in the runoff stream.

- Other than the controversial single study in Austin, no data exist to corroborate that refined coal tar-based sealants are a significant source of PAHs in any sediment.
- This is not to say that no PAHs in the environment come from refined coal tar-based sealants. PAH sources are everywhere that organic fuels are used, and the number of potential sources is very large.

What are PAHs

- Polycyclic Aromatic Hydrocarbons (PAHs) are a class of natural organic chemical compounds consisting of carbon and hydrogen atoms combined in thousands of different ways. A few hundred of the more common combinations have been named, and are sometimes considered separate chemicals.
- PAHs occur as complex mixtures and not as single compounds. PAHs are primarily introduced into the environment as by-products of incomplete combustion. These combustion sources are numerous, including natural sources such as wildfires as well as industrial processes, transportation, energy production and use, food preparation, smoking tobacco, and disposal activities such as open trash burning. Combustion sources are said to produce *pyrogenic* PAHs (that is, PAHs from burning). *Petrogenic* PAHs (literally, PAHs from rocks) are found in coal and crude oil.
- If you want to make your own PAHs, try any of the following: smoke a cigarette, barbeque a steak, burn toast, start composting, make a campfire, burn oil for heat, run an internal combustion engine, run a jet engine, light a candle, light an acetylene torch without adding oxygen.
- PAHs are lipophilic, meaning that they like oil more than water. In the environment, PAHs are thus not found in clean water but instead occur in soil and sediments.
- PAHs are present in soils and sediments in urban environments; PAH concentrations have been shown to increase as the population of the urban area increases (for example, see Hafner and others, 2005).
- Unlike metals, PAHs are organic compounds (contain carbon) and therefore can degrade in the environment. In aquatic environments, PAHs biodegrade (that is, are broken down by micro-organisms) to a greater or lesser degree depending on a wide variety of factors. PAHs can also be degraded by sunlight and atmospheric oxidation. Degradation is more rapid for some PAHs than for others so that, in the context of a human life time, some PAHs have been described as persistent.

- PAHs can enter the environment from "point sources," such as municipal or industrial outfalls, or "non-point sources," such as rainfall, runoff, or atmospheric deposition.

What's the problem with PAHs?

- The U.S. Environmental Protection Agency (EPA) lists 16 PAHs as Priority Pollutants. Seven of the 16 are listed in the U.S. Report on Carcinogens (http://ntp.niehs.nih.gov/ntp/roc/toc11.html):
  - one is classified as a known human carcinogen,
  - seven are classified as likely to be human carcinogens, which generally means that tumors have been found in experimental animals exposed to high concentrations of the substance.

  Nine of the 16 are not classified as carcinogens. The U.S. Report on Carcinogens list is consistent with the list developed by the World Health Organization's International Agency for Research on Cancer.
- Because PAHs are practically insoluble in water, they tend to be concentrated in soils and sediment. In aquatic environments, bottom-living and bottom-feeding organisms can come in contact with PAH-contaminated sediment. Impacts of PAHs on the health of aquatic organisms are widely studied and many books and scholarly papers are available on the topic. In general, PAHs with lower molecular weights can be acutely toxic when present at elevated concentrations but are generally noncarcinogenic to aquatic organisms. PAHs with higher molecular weights are generally not acutely toxic to aquatic organisms, but a number of them are classified as possible carcinogens.
- Interactions between aquatic organisms and PAHs in sediments are exceedingly complex. Scientists who have worked extensively on the problem agree that the occurrence of adverse biological effects are difficult to predict using only concentration data. The likelihood of adverse biological effects at any particular location depends on a host of factors, including the sensitivity of species present and the bioavailability of the PAHs at the location. Bioavailability in turn depends on factors such as physicochemical properties, geochemical and biological factors, and even the source of the PAHs. For example, there is some evidence that PAHs from coal-related sources are less bioavailable than petroleum-related sources, which are thought to be more readily desorbed from sediment particles (Paine and others, 1996).

How do PAHs get into streams?

- PAHs get into streams primarily through atmospheric deposition and rainfall runoff. Atmospheric deposition of particulate matter introduces PAHs from both distant and nearby sources into streams. Atmospheric PAHs are from vehicle exhaust, forest fires and other combustion sources. Rainfall runoff introduces more locally derived materials from surfaces such as roads, parking lots and roofs, picking up PAHs from tire particles, leaking gasoline and motor oil and roofing materials as well as an additional component of atmospheric particles. In formerly industrialized urban areas, PAHs were introduced into local streams via

old industrial facilities such as manufactured gas plants (MGPs), wood treating facilities and a limitless array of possible sources that processed or used organic fuels (coal, petroleum, wood).

- The sources of PAHs in soils and sediments have been the subject of an enormous volume of scientific research over the past three decades. Identification of sources and apportioning specific sources to specific occurrences of PAHs has developed into its own scientific discipline called "Environmental Forensics." In a recent book about Environmental Forensics, ways of identifying PAH sources were reviewed (Boehm, 2006).
- One recent publication has suggested that refined coal tar-based sealants applied to parking lots may be a significant source of PAHs in Austin, Texas. This study in Austin (Mahler and others, 2005, 2007) unfortunately was limited, failing to include other local sources of PAHs and even disregarding the four former manufactured gas plant (MGP) facilities located at and near the sites of PAH contamination. According to a 2003 article in a local newspaper, the *Austin Statesman*, "*The chemical fingerprint of the contamination at the hillside above Barton Springs pool and in the creek is identical to that of coal gasification wastes…*" Coal gasification wastes are found at MGP facilities.
- A follow-up study in Austin that looked at PAHs in sediments before and three years after a municipal ban on the use of refined coal tar-based sealants showed no discernable differences either in the sources or amounts of PAHs in local streams (Gauthier and DeMott, 2008; DeMott and others, submitted for publication to the journal Environmental Forensics).

At what concentration do PAHs affect stream aquatic organisms?

- The effects of PAHs in sediments on aquatic organisms have been the subject of an enormous volume of scientific research over the past three decades.
- Some research has found that PAHs adversely impact the health of aquatic organisms and/or ecosystems. Other research has found no impact or even beneficial impacts.
- One study (Paine and others, 1996) concluded that PAHs derived from coal tar-based sources have less adverse effects than PAHs from other sources.
- One study in sediments in Austin, Texas waterways attributed most if not all adverse impacts to an ecosystem to a single source: refined coal tar-based sealants. It remains unclear why the authors of this study did not consider other PAH sources known to contribute to PAHs in sediments in Austin.
- Many governments have published guidelines and reference concentrations for the amount of PAHs in sediments that are expected to limit possible adverse effects to organisms or ecosystems. One such guideline is the *Probable Effect Concentration* (PEC) [reference uncertain] for PAHs, which sets 22.8 mg/kg as the concentration below which adverse impacts to bottom-dwelling organisms are expected to be minimal.

What are options for reducing or preventing impacts from PAHs in paving materials?

- While there is no evidence that refined coal tar-based sealants are important sources of PAHs in NC sediments, and reduction in use of refined coal tar-based sealants is unlikely to result in any noticeable decrease in PAHs either already in NC sediments or that is deposited in sediments in the future, PCTC members recommend that manufacturer's specifications be followed to limit the possibility of environmental releases. Specific recommendations include:
  - Make sure no significant rainfall is forecast for at least 48 hours after application of the sealcoat;
  - Only apply sealcoat when temperatures are above 60° Fahrenheit and rising throughout the application period;
  - Take appropriate measures to ensure that newly applied sealcoat does not impinge on adjacent surfaces or enter storm or sewer drains; and
  - Make sure that no traffic can access the newly sealcoated surface for at least 12 hours.
- Asphalt-based sealants also contain PAHs. As documented in the same University of New Hampshire study described above, asphalt-based sealants are another source of PAHs in the environment. Manufacturer's specifications must also be followed during application of asphalt-based sealcoat.
- Asphalt on high speed roadways, on roofs and other surfaces are sources of PAHs, as are gasoline spills, oil and lubricant leaks and materials abraded from tires. To minimize PAHs in the environment, avoid spills and leaks and make sure equipment is in optimal working order.
- Concrete parking lots do not require sealants, but do collect PAHs from spills, leaks, abrasion and atmospheric deposition which may be washed into streams during rain events. To minimize PAHs in the environment, avoid spills and leaks and make sure equipment is in optimal working order.
- A North Carolina State University study in Wilmington, NC, found a reduction in the concentration of PAHs from parking lot runoff after treatment by a vegetated bioretention cell. This is similar to the attenuation noted in the University of New Hampshire study. Installing bioretention cells (also called rain gardens, or plant beds) to treat parking lot runoff may reduce PAHs, as well as other pollutants, in stormwater runoff.
- Some proprietary stormwater management devices are marketed to reduce organic toxins such as PAHs.

Have any local governments responded to the research on PAHs and coal-tar based sealants?

- The City of Austin, Texas passed an ordinance in November 2005 prohibiting the use and sale of refined coal tar-based sealants. Sediment samples collected before and about three years after the ban found no reduction in the amounts or types of PAHs in Austin sediments.
- Some other local governments (Dane County [Madison], Wisconsin and Washington, DC) have passed similar bans. In both Wisconsin and Washington, the ban was based on the study in Austin, Texas. There is no evidence that

refined coal tar-based sealants have discernably contributed PAHs to sediments in either Madison or Washington, DC.

Selected References

Boehm, P.D. 2006. Polycyclic aromatic hydrocarbons (PAHs). pp. 313–337. *In*: Environmental forensics: A contaminant specific approach. R.D. Morrison and B.L. Murphy (eds). Elsevier Academic Press, New York, NY.

Boehm P.D., C.P. Loreti, A.B. Rosenstein, and P.M. Rury. 2002. A guide to polycyclic aromatic hydrocarbons for the non-specialist. Publication Number 4714. American Petroleum Institute, Washington, DC.

Burgess, R.M. (2007). Evaluating Ecological Risk to Invertebrate Receptors from PAHs in Sediments at Hazardous Waste Sites. U.S. Environmental Protection Agency Document EPA/600/R-06/162, ERASC-011, External Review Draft, January 2007.

Canadian Council of Ministers of the Environment. 1999. Canadian sediment quality guidelines for the protection of aquatic life: Polycyclic aromatic hydrocarbons (PAHs). *In*: Canadian environmental quality guidelines, 1999, Canadian Council of Ministers of the Environment, Winnipeg.

DeMott, R.P., Gauthier, T.D., Wiersema, J.M. and Crenson, G. (Submitted). PAHs in Austin Sediments after a Ban on Pavement Sealers. Submitted to Environmental Forensics, 2009.

Douben, P.E.T., Editor (2003). PAHs: An Ecotoxicological Perspective (Ecological & Environmental Toxicology Series). Wiley, 404 pp.

Gauthier, T.D. and DeMott, R.P. (2008). Analysis of PAH Concentrations Detected in Austin Texas Stream Sediments Following a Ban on the Use of Coal Tar Sealers [abstract]. Society of Environmental Toxicology and Chemistry Annual Meeting Abstracts with Program, November 2008.

Hafner, WD, Carlson, D, and Hites, R, 2005. Influence of Local Human Population on Atmospheric Polycyclic Aromatic Hydrocarbon Concentrations. Environmental Science & Technology, v. 39 pp. 7374-7379.

Neilson, A.N., Editor (1997). PAHs and Related Compounds (The Handbook of Environmental Chemistry) (Part 3I). Springer, 436 pp.

Paine, M.D., P.M. Chapman, P.J. Allard, M.H. Murdoch, and D. Minifie (1996). Limited bioavailability of sediment PAH near an aluminium smelter: Contamination does not equal effects. Environ. Toxicol. Chem. 15(11):2003–2018.

U.S. Department of Health and Human Services, Public Health Service, Agency for Toxic Substances and Disease Registry (ATSDR) ToxFAQ for Polycyclic Aromatic Hydrocarbons (PAHs). Available at: http://www.atsdr.cdc.gov/tfacts69.html.

U.S. Department of Health and Human Services, Public Health Service, Agency for Toxic Substances and Disease Registry (ATSDR). Toxicological Profile for Polycyclic Aromatic Hydrocarbons. ATSDR/TP-90-20 (1990).

Ward, Mike (2003). Sources of Toxic Waste: Austin's 19th century coal gas plants provided light, left pollution. *Austin Statesman*, Sunday January 19, 2003.