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White Paper

Coal Tar Sealants: Impacts to Human Health and the Environment

For Southeastern Wisconsin Watersheds Trust, Inc

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1.0 Introduction

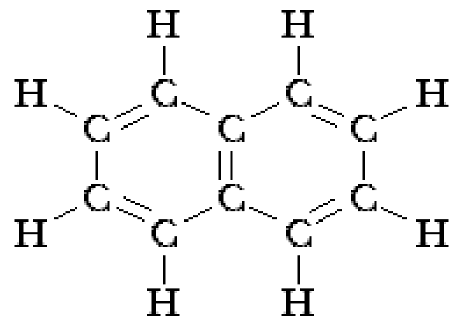
Coal tar is a steel industry byproduct resulting from the coking of coal. Coal tar pitch is the residue left after distillation of coal tar, which is incorporated into commercially available sealants applied to driveways and parking lots. Polycyclic aromatic hydrocarbons (PAHs), a component of coal tar, form when a carbon based substance is burned. Many of the PAHs found in coal tar are listed as human carcinogens. Moreover, some PAHs in the aquatic environment are found to be toxic to or otherwise adversely impact mammals, birds, fish, invertebrates and plants.

In a summary of national PAH studies, the United States Geologic Survey (USGS, Fact Sheet 2011-3010) reports that one half of PAHs in sediments in lakes can be traced to coal tar. Recent studies of Milwaukee area sediments (Baldwin, et al) determined that PAHs derived from coal tar are commonly found at toxic levels, with elevated PAH levels found in stream sediments throughout Milwaukee County. Similar studies in Minnesota have found elevated PAH levels in stormwater pond sediments, requiring excavated sediments to be categorized as hazardous waste materials when these facilities are maintained.

This White Paper will illustrate the potential and existing human health and aquatic environment impacts caused by the use of coal tar sealants, with specific reference to the Milwaukee area. Elimination of this source of harmful PAHs will significantly reduce the accumulation of this pollutant in the environment and result in remediation efforts that are effective over the long term.

2.0 Coal Tar Sealants are a Significant Source of PAHs in the Environment

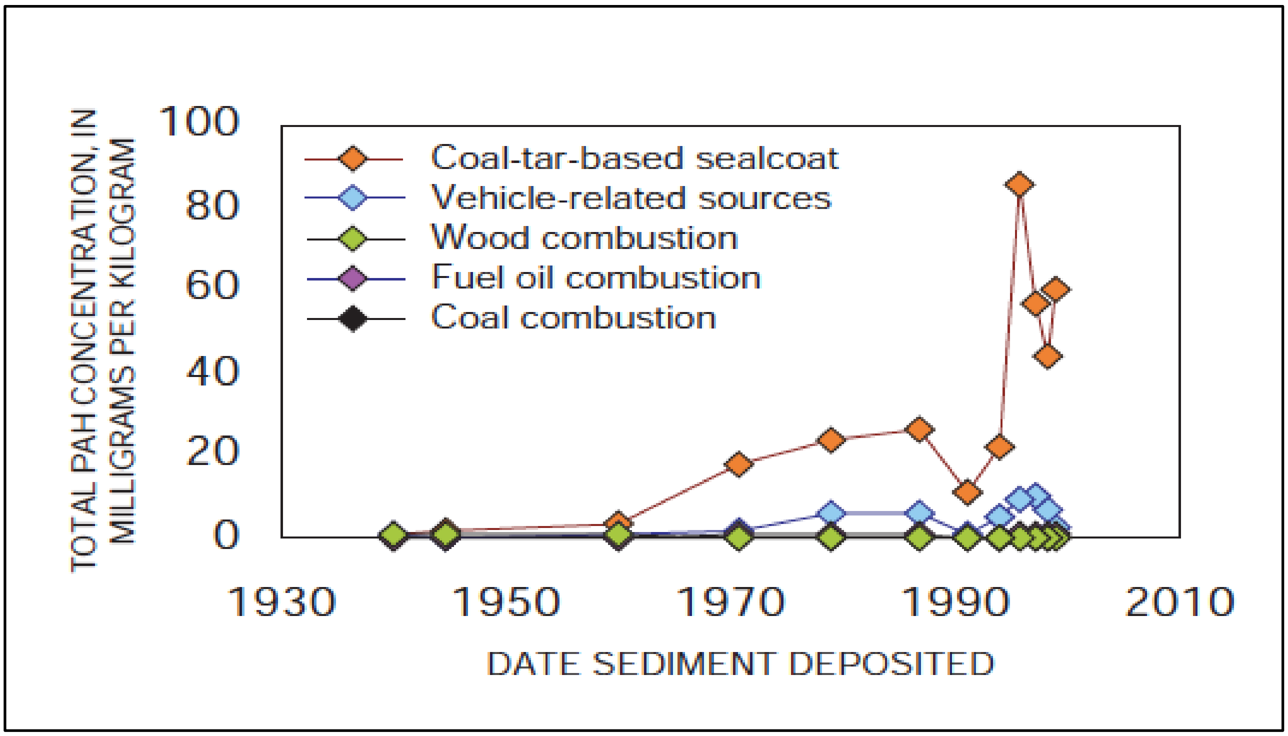
Polycyclic aromatic hydrocarbons are group of organic compounds (>200) containing only carbon and hydrogen atoms arranged around two or more aromatic (benzene - C_6H_6) rings (example provided below).



PAHs are a combustion byproduct and, as such, there are numerous sources that can contribute to their presence in the environment. Anthropogenic sources, in addition to the coal tar sealants, include vehicle-related emissions and tire wear, as well as wood, fuel oil and coal combustion. There are natural sources as well. Each PAH behaves differently and results from a different process. There are also phytogenic PAH compounds, derived from plants; petrogenic PAH compounds, derived from petroleum products; and pyrogenic PAH compounds, which result from the burning of carbon sources (wood, coal, creosote, coal-tar). Each PAH compound can be "fingerprinted" in a manner that identifies its source.

An example of the relative contribution of PAH sources is provided in a study of PAHs found in sediments obtained from Lake Killarney, located in Orlando, FL. In this study, the analysis of different strata, obtained from sediment cores, illustrated both the increase in PAH concentrations over time, and the increasing predominance of coal tar based seal coat as the primary PAH source (Source: USGS, Coal-Tar-Based Pavement Sealcoat, Polycyclic Aromatic Hydrocarbons (PAHs), and Environmental Health, Ref. 3**)**.

Figure: Coal-tar-based sealcoat concentrations of PAHs in Lake Killarney, Orlando, Florida, as determined by chemical fingerprinting.



A recent study of Milwaukee stream sediments, obtained from 40 locations, determined that PAHs were present in most samples and the highest concentrations are found where a significant portion of the drainage area land uses are parking lots and commercial development (Baldwin et al).

PAH Levels in Asphalt-Based and Coal-Tar-Based Sealcoat

Pavement sealcoat is a commercial product that has been applied to many asphalt parking lots, driveways, and playgrounds in southeastern Wisconsin in an effort to protect and beautify the underlying asphalt. It has rarely been used on public roads. Most sealcoat products applied throughout the United States are either coal-tar or asphalt emulsion, although some alternative products now are available. Coal tar and coal-tar pitch have extremely high concentrations of PAHs as do coal-tar-based sealcoat products, which typically are 20 to 35 percent coal tar or coal-tar pitch. Asphalt and asphalt-based sealcoat products have much lower concentrations of PAHs. Coal-tar-based sealcoat is commonly used in Wisconsin, except in communities where they have been banned.

Following pavement application, the sealcoat product gradually abrades to a powder and becomes a component of pavement dust, which can then be transported by rainfall runoff to stormwater management facilities and receiving water bodies (rivers and lakes). Pavement dust also adheres to tires that track it onto unsealed pavement, and foot traffic that carry it into nearby residential homes, where it is incorporated into house dust. Finally, wind and runoff also transport the pavement dust to adjacent soils.

A comparison of PAH concentrations (Summation of 12 PAH compounds, mg/kg from B. Mahler SETAC presentation 2014) for different sources is illustrated in Table 1. From the table below, dust particles from coal tar sealants have significantly greater PAH concentrations than other common sources, including alternative sealants, that can impact human receptors or organisms in the aquatic environment.

Table 1.

Fresh Asphalt	**1.5**
Weathered Asphalt	**3**
Fresh Motor Oil	**4**
Brake Particles	**16**
Road/Tunnel Dust	**24**
Tire Particles	**80**
Diesel Engine	**102**
Gasoline Engine	**370**
Used Motor Oil	**440**
Asphalt Based Pavement Sealant	**50**
Coal Tar Based Pavement Sealant	**70,000**

The potential for coal tar based sealants to contribute PAHs to urban waterways is significant because of the high concentrations and widespread use and PAHs are persistent in the environment. Once in the environment, they can be broken down by ultraviolet light, or in soil and sediment by a mixed microbial population. However, degradation, if conditions are favorable, takes weeks to months.

3.0 Potential Risks to Human Health

Elevated PAH concentrations associated with a variety of settings, and their potential human health impacts, have been investigated and reported by the USGS, other government agencies, and academic institutions, as summarized below. [Ref. 3 and 10]

PAHs from coal-tar-based sealcoat contaminate house dust

From Table 1, dust particles from surfaces sealed with coal tar products will have significantly higher concentrations of PAHs than dust from other common sources. In a study of 23 ground-floor apartments in Austin, Texas, house dust PAH levels were 25 times higher in apartments with parking lots sealed with a coal tar based product, as compared to apartments with parking lots with other surface types (concrete, unsealed asphalt, and asphalt based sealcoat). No relation was found between PAHs in house dust and other possible indoor PAH sources such as tobacco smoking and fireplace use.

House dust is an important pathway for human exposure to many contaminants, including PAHs. This is particularly true for small children, who spend time on the floor and put their hands and objects into their mouths. Research has shown that a preschooler living in a residence adjacent to coal-tar-sealed pavement who has relatively low hand-to-mouth activity consumes about 2.5 times more PAHs from house dust than from their diet. For the more active preschooler, whose hand-to-mouth activity is higher, the PAH intake from house dust is nearly 10 times more than the PAH intake from their diet.

Living adjacent to coal-tar-sealed pavement increases cancer risk

The USGS partnered with a human-health-risk analyst to estimate the excess lifetime cancer risk associated with the ingestion of house dust and soil for people living adjacent to parking lots with and without coal-tar-based sealcoat. Where, excess cancer risk is the extra risk of developing cancer caused by exposure to a toxic substance. The excess cancer risk for people living adjacent to coal-tar-sealed pavement (1.1 cancer incidences for every 10,000 individuals exposed) was 38 times higher, on average (central tendency), than for people living adjacent to unsealed pavement. The central tendency excess cancer risk estimated for people living adjacent to coal-tar-sealed pavement exceeds the threshold generally considered by the EPA as making remediation advisable.

The assessment used measured concentrations of the PAHs in house dust and soils adjacent to coal-tar-sealed pavement, and established house dust and soil ingestion rates to estimate the excess cancer risk. Much of the estimated excess risk comes from exposures to PAHs in early childhood (6 years and under). The study did not consider the excess cancer risk associated with exposure to the seal coated pavement itself, which has PAH concentrations 10 or more times greater than in adjacent residence house dust or soils.

American Medical Association policy

Given the above findings, the American Medical Association (AMA) has adopted a new policy aimed at reducing or ending the use of common coal tar based sealcoats, which advocates for legislation that bans the use of pavement sealcoats containing PAHs, or requires the use of sealcoat products containing reduced PAH concentrations. According to the International Agency for Research on Cancer, PAH compounds have been proven to be carcinogenic, mutagenic and teratogenic to humans.

Per AMA Board member Albert J. Osbahr Ill, MD (ref 10), "Whether they are sending their children to a playground or repairing a driveway, Americans are potentially being exposed to harmful carcinogens in coal-tar-based sealcoats. Even if one's exposure is limited, as sealcoats erode over time, PAHs leach into the water, soil and air, finding their way into sediment and eventually into aquatic wildlife. We must take action to either eliminate the use of PAH altogether or dramatically reduce its concentration in coal-tar sealcoats."

4.0 Potential Risks to Aquatic Life

Elevated PAH concentrations associated with a variety of settings, and their potential ecosystem health impacts, have been investigated and reported by the USGS, other government agencies, and academic institutions, as summarized below. [Ref. 3]

Runoff from coal-tar-sealed pavement is acutely toxic to aquatic biota

Exposure to runoff from coal-tar-sealed pavement collected up to 42 days after sealcoat application resulted in 100 percent mortality to two commonly tested laboratory organisms: day-old fathead minnows and water fleas. In contrast, minnows and water fleas exposed to runoff from unsealed pavement experienced no more than 10 percent mortality. When the minnows and water fleas were also exposed to simulated sunlight, which intensifies the toxicity of some PAHs, runoff collected 111 days (more than 3 months) after sealcoat application caused 100 percent mortality to both species, and caused 100 percent mortality to water fleas even when diluted to 10 percent of its original strength.

The USGS collected samples of runoff from 5 hours to111 days following sealcoat application to pavement by a professional applicator. Total PAH concentrations varied relatively little, as rapid decreases in concentrations of low molecular weight and nitrogen-substituted PAHs were offset by increases in high molecular weight PAHs. These results demonstrate that runoff from coal-tar-sealed pavement continues to contain elevated concentrations of PAHs and related compounds long after a 24-hour curing time.

A subsequent study by researchers at the National Oceanic and Atmospheric Administration (NOAA) and the U.S. Fish and Wildlife Service found that coal-tar-sealed runoff is acutely lethal to juvenile Coho salmon and causes a wide spectrum of abnormalities to zebrafish embryos. They also reported that filtration of the runoff through a bioretention system substantially reduced toxicity.

Runoff from coal-tar-sealed pavement damages DNA and impairs DNA repair (ref 3)

DNA in rainbow trout liver cells was damaged due to simultaneous exposure to runoff from coal-tar-sealed pavement and simulated sunlight, even when the runoff was diluted to 1 percent of its initial concentration. The test assessed two types of DNA damage: strand breaks and alkylated bases. Although cells can repair some DNA damage, a second experiment demonstrated that cells exposed to the coal-tar-sealed runoff had an impaired capacity to perform at least one type of DNA repair. The combination of DNA damage and impaired repair capacity intensifies the potential for long-term damage to cell health. DNA damage has many possible consequences, including aging, cell death, and mutations. Mutations can affect the function of genes and can potentially lead to cancer.

Air-Quality Concerns (ref 3)

Although unseen, releases of PAHs to the atmosphere (volatilization) from freshly coal-tar-sealed pavement are tens of thousands of times higher than from unsealed pavement. Volatilization is a potential human-health concern because inhalation is an important pathway for human exposure to PAHs. Although volatilization decreases rapidly over the weeks following application, it nonetheless continues long after application. In fact, PAH releases to the atmosphere from parking lots sealed 3 to 8 years previously, were on average 60 times higher than PAH releases from unsealed pavement. Nationwide, the combined PAH releases each year from newly applied coal-tar-based sealed pavement are estimated to exceed annual vehicle emissions of PAHs.

5.0 Cost for Removal

Stormwater pond dredging costs due to PAH Contamination (Minnesota Example) [Ref. 6]

Inver Grove Heights, a suburb of St. Paul, Minnesota, has 589 stormwater drainage basins, including many that are "internally drained" due to the city's kettle moraine topography. These ponds typically have a 30 to 40-foot deep bowl-shaped catchment. Beginning in 2009, the city began inspecting its basins as part of routine maintenance required by its Municipal Separate Storm Sewer System (MS4) permit. Inspections revealed that many basins were significantly filled with sediment, reducing their holding capacity.

Twelve ponds were tested for PAH contamination. Three of the 12 ponds were found to be contaminated with PAHs, with two having high PAH concentrations. These findings led the city to ban coal tar-based pavement sealcoats in November 2011. The city embarked on its first PAH clean-up effort by targeting the smaller of the two ponds found to be highly contaminated. This small project required the removal and landfilling of only 50 yards of sediment at a cost of $60 per yard ($3,000 total).

The second pond however had significant more sediment accumulation, with a full clean-out estimated to yield 7,000 yards of sediment. At a cost of $65 per yard, this option would have cost $455,000 for disposal alone. To reduce this cost, the city chose to dredge only the area around the inlet of 2,300 yards of sediment. Ultimately, a licensed landfill within the city limits agreed to accept the contaminated sediment for use as daily cover in a lined cell, reducing disposal costs to $36 per yard. The close proximity of the landfill also reduced transport costs. While the project did not fully restore the basin's storage capacity, the final project cost of $155,000 included:

- Evaluating and testing sediment;
- Engineering, contracting, and work plans;
- Site preparation (including temporary access roads and storage areas);
- Mobilization and overhead;
- Excavating, transport and disposal of sediment;
- Site restoration.

City staff estimate that over 140 basins will show some PAH contamination; with nearly 100 having high levels of contamination. The local landfill will not have enough space to hold these additional sediments, meaning that disposal costs will jump from $36 to $65 per ton (or higher), not including additional transportation costs. Estimates for removing PAH contamination from all basins in the City range from $1.5 to $4.0 million.

6.0 Product Alternatives to Coal Tar Sealant

As reported by the State of Minnesota Pollution Control Agency (PCA) [Ref. 11], the most common and cheapest alternative to coal tar sealants now on the market is petroleum asphalt-based sealcoat (CAS number 8052-42-4). Asphalt sealcoats contain PAHs, but at a much lower concentration (as small as 1/1000th the PAH level of coal tar sealcoats). Good asphalt sealcoat emulsions are considered very affordable, provide a black appearance for 1-2 years, and provide less-visible protection for 2 to 4 years if properly applied. Typically, they are permitted in locations that enforce a coal tar sealant ban.

Asphalt sealcoat data sheets typically state an asphalt content ranging from 10% to 30%. A 30% asphalt content is recommended. Polymer content of 2.5% to 5.0% will aid drying, add flexibility, and assist in aggregate retention (chip or gravel) when applying a chip seal.

Other alternatives such as Gilsonite®, acrylic and agricultural oil-based seals contain fewer or no PAHs, but tend to be higher in price. Also, these alternative products have less of a performance record, as compared to asphalt seals. However, these products and their cost-effectiveness should still be considered, since research and market conditions continue to evolve.

It is recommended that the consumer review data sheets, and contact providers and/or manufacturers to inquire about key protection ingredients and recommendations pertaining to optimal application. The following is noted:

- For asphalt fog seal-sealcoat products, the binder component (asphalt) content provided on data sheets typically ranges from 10% and 30%. However, an asphalt content between 25% and 30% is highly recommended.
- The asphalt emulsion for chip seals, which includes a layer of gravel, should be around 65% residual asphalt.
- For asphalt-based sealcoat emulsions, the data sheet will provide the water content, but the manufacturer may also provide guidance pertaining to the necessary dilution required for application, whether completed by spraying or spreading. Be aware of these guidelines, but also recognize that the dilution may vary with the applicator selected for use. Following application and curing, the sealcoat should have a uniform coverage and appearance, with all aggregate and spaces in-between covered, without gaps or pinholes.
- Polymers are commonly used in sealcoats but are not considered essential. Rubbery polymers (i.e., styrene/butadiene, isoprene, and neoprene) increase flexibility, and provide resistance to heat and cold extremes. Polymers also help the sealcoat set faster, an essential feature when rain, cold or darkness are approaching; and in commercial parking lots, where traffic use requires timely restoration. If desired, the selected asphalt sealcoat should contain 2.5% to 5.0% polymers. It is noted that highway departments routinely specify a polymer-modified binder in fog and chip seals.
- Clay, mineral, quartz or similar materials add strength to the sealcoat, and are typically provided at approximately 20% by weight.
- If desired, carbon black, slate and other mineral additives will darken the coating.
- Data sheets do not include sand, which may be added by the contractor (3 to 6 pounds per gallon) to fill a rough surface, or to provide improved traction to newer or smoother pavement. It is noted however, that as the sealcoat binder begins to wear, the sand will be released, causing greater abrasion beneath the tires and accelerating the wear rate of the coating.

For larger commercial parking lots, chip seals containing gravel are slightly more expensive than sealcoats without stone, but may last about twice as long.

Gilsonite® sealcoats typically have some petroleum asphalt content, and are another low-PAH option. However, they may also include naptha, mineral spirits or other solvents that may present a local regulatory concern, as related to ground-level ozone or smog formation. It is often recommended that this product only be applied on unsealed or asphalt-sealed pavement; not on coal tar or acrylic-sealed surfaces. Gilsonite-based sealcoats may be approximately twice the cost of asphalt-based sealcoats. Experiences vary on how much additional life they provide.

The no-PAH options include acrylic-based and agricultural oil-based sealcoats, and cement-based micro-layers. These are not as readily available as asphalt-based sealcoats, and there is less long-term experience or research demonstrating how they protect and preserve pavement. This is especially true for retail acrylics applied by homeowners. Even though somewhat higher in initial cost than asphalt-based sealcoats, professionally-formulated and -applied non-asphalt sealcoats may be worth considering in some applications.

With zero-PAH products, it is important to note that there is uncertainty regarding the longevity of the surface treatment. Ideally, advice can be obtained from one or more sources experienced with these newer products. Also, be sure to follow manufacturer's guidance regarding surface compatibility (type and condition) and rates of application. Should these products be improperly applied, they may ruin the pavement or reduce its life.

Product warranties, if offered by the manufacturer or contractor, vary in length (1 to 10 years). If a warranty is offered, it should be obtained in writing. Compare product claims whenever possible.

7.0 Case for Banning Coal Tar Sealant Use

From the above, discontinuing use of coal tar sealants will:

- Reduce human health risk, especially for children
- Improve aquatic habitats – especially long term following sediment remediation and other watercourse improvements projects in the urban environment
- Dramatically reduce future costs associated with for maintaining stormwater and other wet weather management facilities, and,
- Alternative sealants with significantly lower impacts are available at comparable costs.

While these factors apply nationally, these actions are particularly important to the greater Milwaukee area, given the sediment accumulation that occurs within the downstream limits of the Milwaukee, Menomonee and Kinnickinnic River watercourses. Therefore, eliminating coal tar sealants in the Milwaukee area is expected to have a dramatic long-term environmental benefit, and help facilitate related environmental projects, such as dam removal and delisting the Milwaukee Estuary as a designated Area of Concern.

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