Coal-tar-based Pavement Sealants – A Potent Source of PAHs

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Pavement sealants are applied to the asphalt pavement of many parking lots, driveways, and even playgrounds in North America (Figure 1), where, when first applied, they render the pavement glossy black and looking like new. Sealant products used commercially in the central, eastern, and northern United States typically are coal-tar-based, whereas those used in the western United States typically are asphalt-based. Although the products look similar, they are chemically different. Coal-tar-based pavement sealants typically are 25-35 percent (by weight) coal tar or coal-tar pitch, materials that are known human carcinogens and that contain high concentrations of polycyclic aromatic hydrocarbons (PAHs) and related chemicals (unless otherwise noted, all data in this article are from Mahler et al. 2012 and references therein).

PAHs are a large group of organic chemicals created by heating or burning material that contains carbon; 16 PAHs are classified as U.S. Environmental Protection Agency Priority Pollutants, six are classified as probable human carcinogens, and one (benzo[*a*]pyrene) is classified as a known human carcinogen. The many sources of PAHs to the urban environment span a wide range of PAH concentrations and include tire particles, used motor oil, and diesel and gasoline engine exhaust (Figure 2). Of known urban PAH sources, coal tar and the related compound creosote have the highest PAH concentrations. Coal-tar-based pavement sealant products contain, on average, about 70,000 mg/kg polycyclic aromatic hydrocarbons (PAHs), on the order of 1,000 times higher than asphalt-based products, which typically contain about 50 mg/kg PAHs.

Pavement sealant is not permanent – the sealant must be reapplied every few years, because it is removed from the pavement by wear and tear from traffic, weathering, and chemical processes. There are at least three ways that pavement sealant (and the PAHs and other chemicals it contains) can leave the pavement surface and enter the surrounding environment: by eroding into small, mobile particles; by dissolving into water; and by volatilizing into air. Where do PAHs derived from sealant go? The answer depends on the process involved. Pavement sealant is worn by vehicle tires into a fine powder that

Figure 1. Pavement sealant is commonly used to seal parking lots, playgrounds, and driveways throughout the United States. Sealants used in the central, northern, eastern, and southern United States typically contain coal tar or coal-tar pitch, both of which are known human carcinogens. Photos by the U.S. Geological Survey.

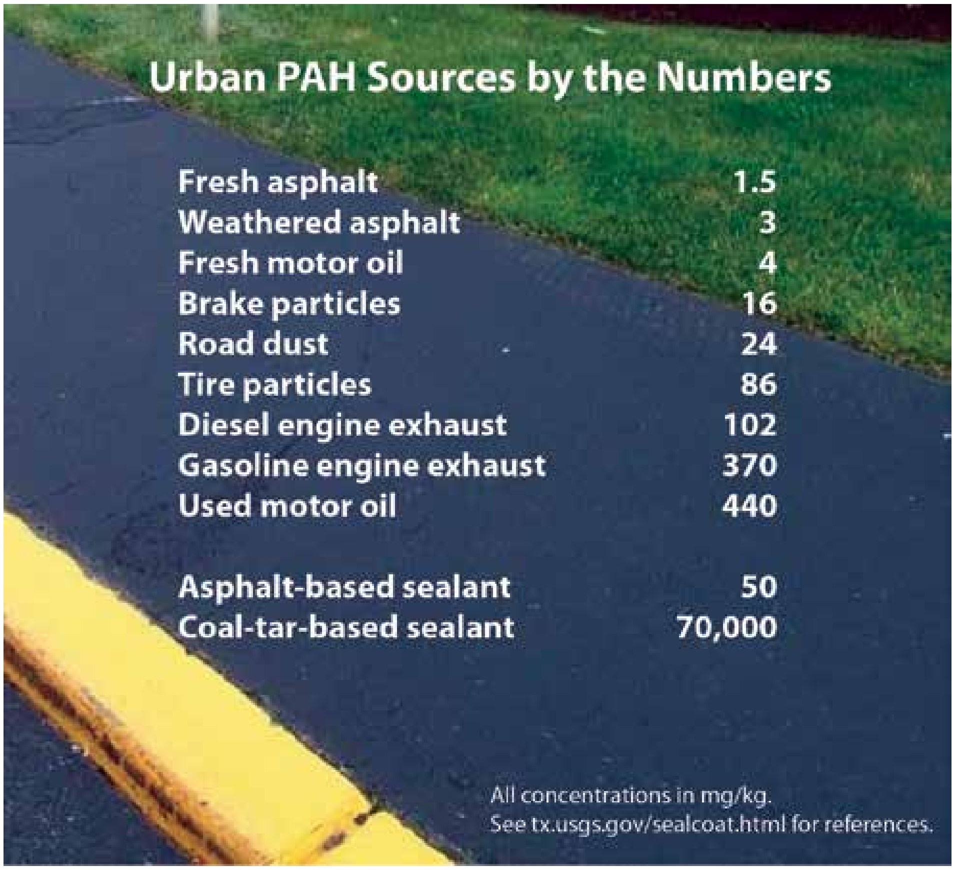

Figure 2. PAH concentrations in typical urban sources. The concentrations shown are a mean or median from as many as six studies.

is transported by stormwater runoff to streams or lakes, or by wind to adjacent soils or other impervious surfaces. Eroded sealant particles can also stick to shoes and be transported indoors. Some PAHs can dissolve into stormwater, especially just after application, and be transported, along with eroded sealcoat particles, to nearby streams and lakes. PAHs also can be released directly into the atmosphere (volatilization). The high PAH concentrations in coal-tar-based sealant can result in high concentrations of PAHs in a number of environmental settings, with potential adverse effects for human and ecosystem health (see other articles on health and ecosystem effects in this issue).

How can we evaluate whether coal-tar-based sealants are an important source of PAH contamination to the environment? One straightforward way is to compare PAH concentrations in pavement dust, runoff, soil, sediment, house dust, and air on or near coal-tar-sealed pavement with PAH concentrations in those same media on or near unsealed asphalt pavement or asphalt-sealed asphalt-based sealant (Figure 3).

Pavement Dust and Runoff

The abrasive action of vehicle tires and snowplows grinds dried sealant on the pavement surface into small particles that mix with other dust on the pavement. In southern, central, and eastern U.S. cities, where coal-tar-based sealant use dominates, dust on sealed pavement has about 1,000 times higher concentrations of PAHs than dust on sealed pavement in western U.S. cities, where asphalt-based sealant use dominates (Figure 4). Concentrations of PAHs on pavement with coal-tar-based sealant generally are in the thousands of mg/kg, comparable to concentrations in soils at some Superfund sites. Further, PAH levels in dust on sealed parking lots in the eastern U.S. can be hundreds of times higher than concentrations in dust on unsealed parking lots in the same watersheds. All of these parking lots, sealed and unsealed, share other sources of urban PAHs – vehicle exhaust, leaking motor oil, tire particles, atmospheric deposition – the only difference is the presence or absence of coal-tar-based sealant.

Pavement dust is mobile – it collects on the pavement surface and at curbs and is readily transported by runoff down storm sewers (Figure 4). When researchers measured PAH concentrations in particles transported by simulated runoff from six coal-tar-sealed parking lots, the mean concentration was 3,500 mg/kg, whereas the mean PAH concentration of particles in runoff from unsealed parking lots (asphalt or concrete) was 54 mg/kg (Mahler et al. 2005). For context, the concentration at which PAH concentrations are expected to harm bottom-dwelling aquatic life is 23 mg/kg.

PAH concentrations in stormwater runoff are highest in the months following sealant application and decrease with time, but even years after application PAH concentrations remain much higher than those in runoff from unsealed pavement, as demonstrated by studies in Wisconsin and New Hampshire. In Madison, WI, the median PAH concentration in unfiltered runoff six years after application of coal-tar sealcoat to a commercial parking lot was 52 μg/L, about 20-1000 times higher than concentrations in runoff collected from a minor arterial street, a commercial rooftop, and a residential street (0.05-2.4 μg/L). During the three months following application of coal-tar-based sealant to a parking lot at the University of New Hampshire, the mean PAH concentration in unfiltered runoff measured by the University of New Hampshire Stormwater Center (UNHSC) was 1,357 μg/L, and decreased over the next two years to a three-month mean of 17–116 μg/L (Figure 5). PAH-contaminated runoff can be acutely and chronically toxic to fish and other aquatic biota, as described in "Toxicity of Coal Tar Pavement Sealant to Aquatic Animals," page 23 this issue.

In some cases, runoff is collected in a stormwater management device, such as a retention pond, to improve water quality by retaining suspended particles. A study by the UNHSC demonstrated that sediment collected in a stormwater management device draining a coal-tar-sealed parking lot contained 393-1,180 mg/kg PAHs, and sediment collected in a device draining an adjacent unsealed lot contained about 2 mg/kg PAHs. The

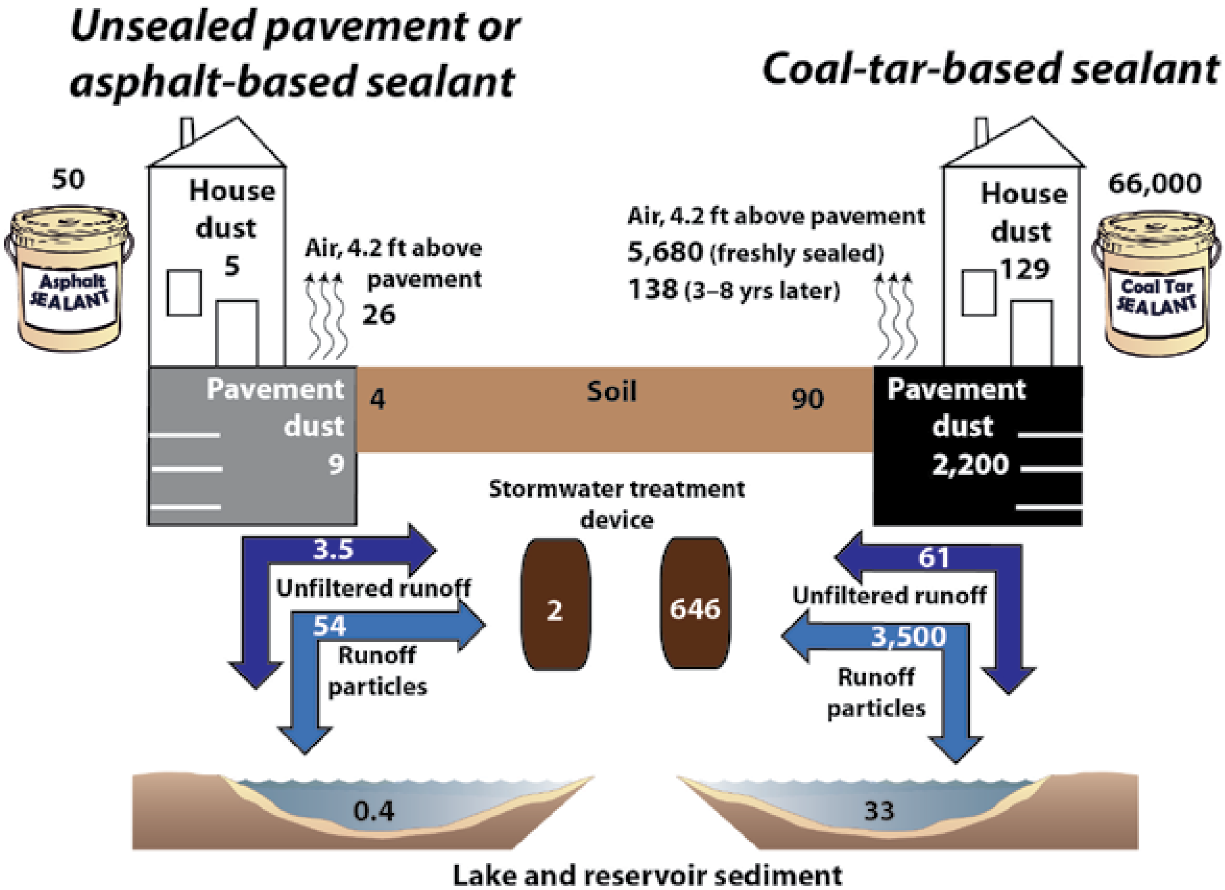

Figure 3. Concentrations of PAHs associated with unsealed or asphalt-sealed pavement (left) are many times lower than those associated with coal-tar-sealed pavement (right). PAHs from sealed pavement are transported to the environment by a variety of pathways, including stormwater runoff to stormwater treatment devices, lakes, and streams; windblown transport to soils; tracking of dried particles indoors; and release by volatilization into air. Humans are exposed to PAHs in pavement dust, soil, house dust, and air; aquatic biota are exposed to PAHs in aquatic sediment, such as in lakes and reservoirs. Concentrations shown are in units of milligrams per kilogram (mg/kg), with the exception of unfiltered runoff (µg/L) and air (ng/m³). Concentration data are from Mahler et al. (2012) (and references therein) or studies cited in this article, and in many cases are the median of multiple studies.

efficient collection of PAH-contaminated sediment in stormwater retention ponds or other devices can have unintended consequences for a municipality because elevated concentrations of PAHs and other contaminants can greatly increase the cost for sediment disposal. Costs for disposing of PAH-contaminated sediment in stormwater ponds in the Minneapolis-St. Paul area are estimated to be $40–50 per cubic yard, or about $125,000 per pond, depending on pond size (Judy Crane, Minnesota Pollution Control Agency, written communication, 2015). That translates to an estimated cost of as much as $1 billion if just ten percent of the ponds in Minnesota contain PAH concentrations that exceed the state's Level 2 human-health risk-based value (Donald Berger, Minnesota Pollution Control Agency, written communication, 2011).

Soil

Contaminated pavement dust can be washed by runoff or blown by wind onto nearby soils. PAH concentrations in soil adjacent to sealcoated pavement in a Chicago, IL, suburb were 23 and 140 mg/kg, 2.3 to 14 times higher than in soil adjacent to unsealed pavement (10 mg/kg) (Van Metre et al. 2009). Composite soil samples from two commercial districts in Fort Worth, TX, where coal-tar sealants were present on some parking lots, had a mean total PAH concentration of about 90 mg/kg, whereas composite soil samples from nearby residential areas, where sealants were not observed, had a mean concentration of about 4 mg/kg (Wilson et al. 2006). Similarly, PAH concentrations in soil adjacent to a coal-tar-sealed parking lot studied by the UNHSC were as high as 411 mg/kg, and decreased with distance from the parking lot to less than 10 mg/kg. The highest PAH concentrations were measured in soil in areas where snowplows had piled snow containing pavement dust and sealant particles scraped off with the snow during the winter.

Lake and Stream Sediment

PAH-contaminated sediment that is not trapped by stormwater ponds can be transported to streams and lakes. The contribution of coal-tar-based sealant to PAHs in lake sediment can be evaluated by a variety of approaches, including "environmental forensics" (the application of statistical methods to evaluate the chemical content of the source and the sediment), microscopic identification of particles, and land-use analysis. An example of environmental forensics is a comparison of the relative proportions of different PAHs – the "fingerprint"

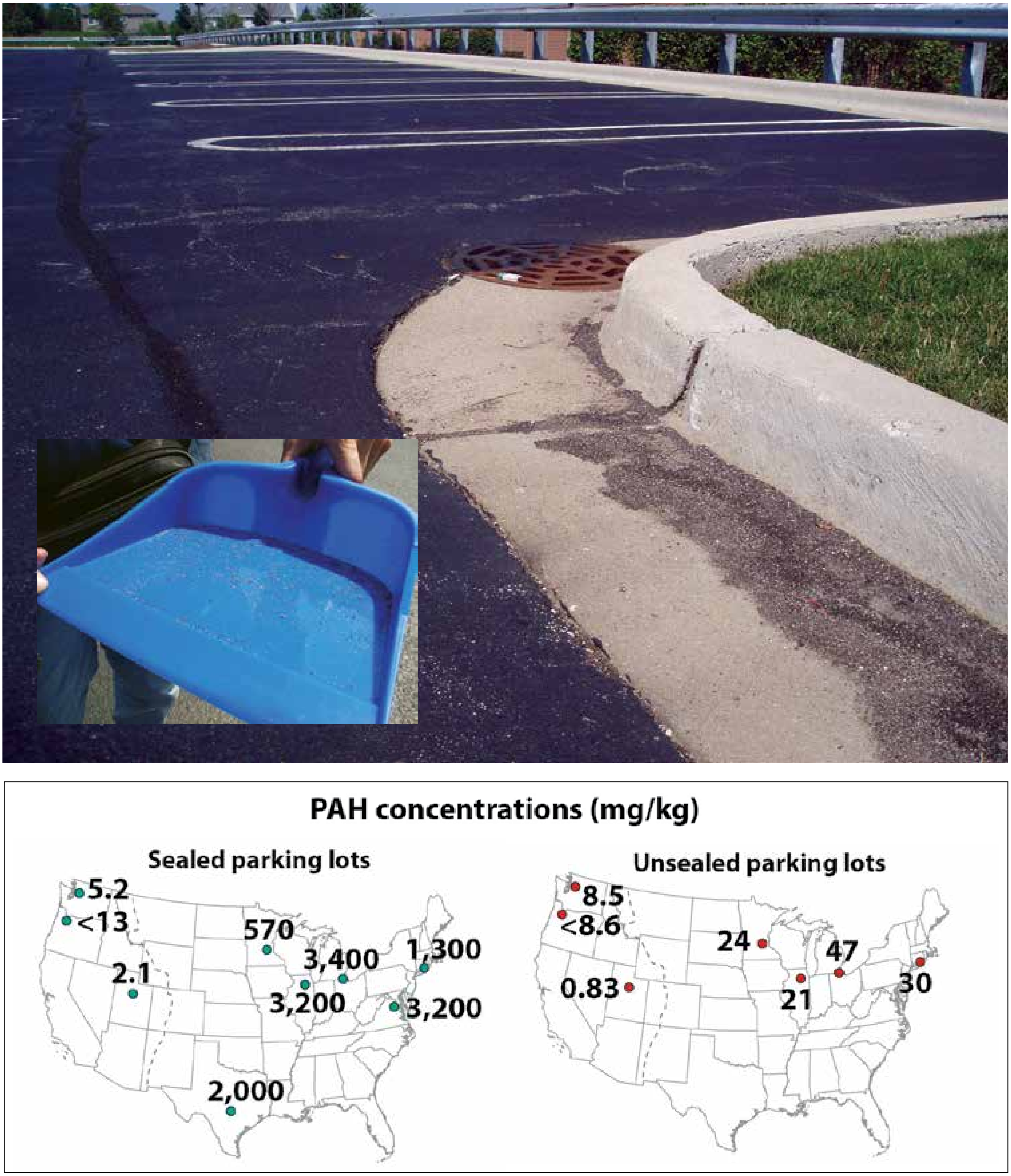

Figure 4. Parking lot "dust" contains abraded sealant (black particles, top). The dust collects on the parking lot surface and along curbs, where it can be carried by runoff down storm drains. PAH concentrations in dust swept from sealed parking lots in the eastern United States, where coal-tar-based sealant is predominantly used, were about 1,000 times higher than in dust from sealed parking lots in the west, where asphalt-based sealant is predominantly used. PAH concentrations in dust on sealed lots in the eastern U.S. also were as much as 100s of times higher than concentrations in dust on unsealed lots in the same watersheds, indicating that the sealant is the principal source of the elevated PAH concentrations (Van Metre et al. 2009). Photo by the U.S. Geological Survey.

Figure 5. Researchers at the University of New Hampshire Stormwater Center simulate runoff on coal-tar-sealed pavement for measurement of polycyclic aromatic hydrocarbons (PAHs). Photo by the University of New Hampshire Stormwater Center.

– in dust collected from parking lots in U.S. cities to fingerprints in sediments from lakes in the same watersheds. For central and eastern U.S. watersheds, the fingerprints of sealed pavement dust and lake sediment were similar, and were different from those in western U.S. watersheds, where the asphalt-based product is used. A more quantitative approach – a statistical method known as source-apportionment modeling – estimated that coal-tar-based sealant contributed about one-half of the PAHs to 40 U.S. urban lakes studied by the U.S. Geological Survey; the other major contributors were vehicles and coal combustion. The topic of PAH sources to lakes and streams is further discussed in “Trends and Sources of PAHs to Urban Lakes & Streams,” page 8 this issue.

House Dust

Coal-tar-based sealant can cause indoor as well as outdoor contamination. Abraded sealant particles can stick to the bottoms of shoes and be tracked indoors, where they become incorporated into house dust. In a study of 23 ground-floor apartments in Austin, TX, apartments with coal-tar-sealed parking lots had house dust with PAH concentrations that were 25 times higher on average than apartments with parking lots that were unsealed or that were sealed with an asphalt-based product (Figure 3). The study found no relation between PAH concentrations in house dust and other factors such as tobacco smoking, barbecue and fireplace use, and candle and incense-burning.

These results are of concern because ingestion of house dust is well recognized as a pathway for human exposure to chemicals, especially for toddlers, who play on the floor and put their hands and objects into their mouths. This topic is discussed in more detail in “Human Health Concerns Associated with Exposure to PAHs & Coal-Tar-Sealed Pavement,” page 19 this issue.

Air

Some of the PAHs in coal-tar-based sealant are released into air during and after application (Figure 3) through a process called volatilization. Many PAHs, including the seven classified as probable or known human carcinogens, are volatile to some degree. Airborne PAHs are of concern because inhalation is another important pathway for human exposure. Although unseen, airborne releases of PAHs from freshly applied coal-tar-based sealant are on the order of 45,000 $\mu g/m^2/hr$, which is tens of thousands of times higher than releases from unsealed asphalt. Taken across the entire United States, emissions of PAHs to air from newly applied coal-tar-based sealant are estimated to exceed those from motor vehicles.

The concentrations of PAHs in air above freshly coal-tar-sealcoated pavement decrease rapidly during the weeks following application, but even years later remain several times higher than in air over unsealed pavement (Figure 3). Air at approximate breathing height above coal-tar-sealed pavement (4.2 feet, or 1.28 meters) even in suburban areas contains PAH concentrations that rival or exceed those in highly industrialized areas. For example, in a study that measured volatilization of PAHs from parking lots that had been treated with coal-tar-based sealant years previously, the mean concentration of the PAH pyrene was about 2-19 times higher than pyrene concentrations measured in air samples from urban industrial sites from New Jersey and Chicago, USA (Van Metre et al. 2012).

A Scientific Consensus

Independent research by scientists and engineers from academic institutions and government agencies demonstrates that coal-tar-based sealant is a potent source of PAHs to water, dust, soil, stream and lake sediment, and air. The comparison of PAH concentrations in settings where coal-tar-based sealant is or is not applied provides an unambiguous picture of the importance of coal-tar-based sealants as a source of PAHs to our urban and suburban environments.

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Additional information is available at USGS Research: PAHs and Coal-Tar-Based Pavement Sealcoat, http://tx.usgs.gov/sealcoat.html.

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