



Mildly Contaminated Soil Distribution Assessment

Final Report

Prepared by Sanborn, Head & Associates, Inc., for the New Hampshire Department of Transportation in cooperation with the U.S. Department of Transportation, Federal Highway Administration

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16. Abstract The Department's Divisions of Operations and Project Development generate large volumes of mildly contaminated soil (MCS) and Limited Reuse Soil (LRS). These MCSs/LRSs contain non-point sourced anthropogenic impacts, with contaminant concentrations above natural background conditions. Additionally, available data indicates the presence of several metals and Polycyclic Aromatic Hydrocarbons (PAHs) at concentrations that exceed criteria that are protective of public health and the environment. Removal and disposal of the MCS/LRS at a solid waste management facility in strict accordance with the NH Solid Waste Rules (Env-Sw), as administered by the NHDES, represents a significant financial burden to the Department. Based on the research reviewed, as part of this Technical Guidance, PAHs in MCS/LRS tend to stay adsorbed to the organic carbon content of the soils and show low potential for leachability, as supported by NHDES's modeling of potential mobility of PAHs such as benzo[a]pyrene.			
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TECHNICAL GUIDANCE
POLYCYCLIC AROMATIC HYDROCARBONS (PAHS)
IN LIMITED REUSE SOIL

*New Hampshire Department of Transportation
Concord, New Hampshire*

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REFERENCES

1.0 INTRODUCTION

Sanborn, Head & Associates, Inc (Sanborn Head) prepared this Technical Guidance on Polycyclic Aromatic Hydrocarbons (PAHs) in Limited Reuse Soil (LRS) on behalf of the New Hampshire Department of Transportation (NHDOT). This Technical Guidance summarizes the literature review completed by Sanborn Head to research the extent, mobility and transport, and bioavailability/bioaccessibility of PAHs in LRS.

LRS is soil along roadways that may contain concentrations of regulated compounds elevated above naturally occurring background concentrations, and therefore, has limited reuse potential. The soil has not been impacted by a known point discharge of oil or hazardous waste but may contain elevated concentrations of manmade constituents such as PAHs.

PAHs, specifically benzo[a]pyrene, are the focus of this Technical Guidance because the contaminant was identified at relatively high concentrations (i.e., compared to typical risk-based guidance values) along roadways and is understood to be traffic related, based on an extensive assessment of elevated concentrations of traffic-related hazardous constituents in road shoulder soils during 2015, completed by the State of Oregon Department of Transportation (ODOT). The ODOT evaluation was used to create a management plan for roadway surface soils when removal is required for construction projects. While both lead and benzo[a]pyrene were identified as constituents that were traffic related and found in levels exceeding soil standards, the lead levels are understood to be a result of Oregon specific historic contamination (White 2016). Therefore, lead is not considered in this review.

The extent of LRS along roadways in New Hampshire has yet to be defined, however it is an emerging concern throughout the United States. The NHDOT, along with several other states, are leading the effort to address and mitigate the potential impacts of LRS and protect the environment.

2.0 POLYCYCLIC AROMATIC HYDROCARBONS (PAHs)

PAHs are a group of organic compounds that consist of two or more fused aromatic rings and are formed during the incomplete combustion of organic materials like wood, coal, oil, gasoline, solid waste, and tobacco, and can be found in materials such as crude oil, waste oil, coal tar, and creosote. There are over 100 different chemicals that are considered PAHs and they commonly occur as a mixture of more than one chemical (ATSDR 1995). The United States Environmental Protection Agency (USEPA) has included 16 PAHs on the Priority Pollutant List found in 40 CFR Part 423, Appendix A. This list is comprised of pollutants that are regulated by the USEPA and could pose a significant potential threat to human health as a result of both their toxicity and potential for human exposure. The 16 PAHs are listed below for reference and are commonly used as indicators of total PAH contamination in soil.

Lower Molecular Weight (LMW):

- acenaphthene (3-rings);
- acenaphthylene (3-rings);
- anthracene (3-rings);
- fluorene (3-rings);
- naphthalene (2-rings); and
- phenanthrene (3-rings).

Higher Molecular Weight (HMW):

- benz[a]anthracene (4-rings);
- benzo[b]fluoranthene (5-rings);
- benzo[k]fluoranthene (5-rings);
- benzo[g,h,i]perylene (6-rings);
- benzo[a]pyrene (5-rings);
- chrysene (4-rings);
- dibenz[a,h]anthracene (5-rings);
- fluoranthene (4-rings);
- indeno[1,2,3-c,d]pyrene (6-rings);
- pyrene (4-rings).

PAHs are often divided into two groups, as shown above, based on their molecular weight and corresponding physical and chemical properties. LMW PAHs include those with 2 or 3 aromatic rings and may cause acute toxicity to aquatic organisms but are considered noncarcinogenic. The HMW PAHs are those with 4 to 7 aromatic rings and tend to include those constituents that are considered probable human carcinogens (Duan 2015).

In general, LMW PAHs are relatively soluble in water, while HMW PAHs tend to be more hydrophobic (i.e. repelled from water) and insoluble, which results in a higher likelihood that the HMW PAHs will adsorb to soil organic matter and be observed primarily with particulates (Ukiwe 2013, Wilson 1993,). Both the solubility and vapor pressure (volatility) of PAHs decrease with an increase in molecular weight (Ukiwe 2013, Abdel Shafy 2016, Duan 2015). These characteristics tend to result in HMW PAHs being less susceptible to degradation and remediation, and hence more persistent in the environment.

As discussed in Section 1.0, the HMW PAH benzo[a]pyrene has been shown to be a traffic related contaminant present in roadside soils. Benzo[a]pyrene is also commonly used as an environmental indicator for the presence of HMW PAHs and is carcinogenic. As such,

the following sections will pay particular attention, when applicable, to the research focused on this constituent and HMW PAHs more broadly.

2.1 Fate in the Environment

PAHs are generally found in two separate phases in the environment, the vapor phase and the solid phase (i.e., PAHs may comprise or be adsorbed onto particulate matter) (Abdel-Shafy 2016, ATSDR 1995). PAHs often enter the environment through a release to air, however they can also enter surface waters through the discharges from industrial and wastewater treatment plants, highway/urban runoff, and direct air deposition. At industrial and/or hazardous waste sites, PAHs can also be released directly to soils. In broad terms, PAHs that are released to the atmosphere will be removed by wet and dry deposition onto soil, water, and vegetation, and have both short and long range transport. PAHs in surface waters may volatilize, photolyze, oxidize, biodegrade, bind/adsorb to suspended particles or sediments, or accumulate in aquatic organisms; while PAHs in soils may also volatilize (Wilson 1993), degrade, or accumulate in the soil itself or accumulate in vegetation (ATSDR 1995).

The fate of PAHs in the environment will depend on the properties of both the chemical and the environment they have been released to.

2.2 Exposure Pathways

Because PAHs are prevalent throughout the environment, human exposure can occur in a variety of ways, but is most commonly caused by inhalation of vapors or particles in the air. Common human activities/sources that contribute to PAH exposure include: cigarette smoke, vehicle exhaust, asphalt roads, coal, coal-tar, wildfires, agricultural burning, residential wood burning, and municipal and industrial waste incineration (ATSDR 1995). PAH exposure can even occur when cooking and eating meat or other food at high temperatures (e.g., grilled), or eating contaminated food. The magnitude of human exposure to PAHs depends strongly on factors such as lifestyle (e.g., diet, tobacco smoking), occupation, and living conditions (e.g., urban versus rural setting, domestic heating, and cooking methods).

Because PAH exposure commonly occurs through the inhalation of air, the lungs are the main pathway for PAHs to enter the body for most people, however little is known about how the lung absorbs PAHs. Absorption of PAHs through ingestion (i.e., drinking water and swallowing food, soil, or dust particles that contain PAHs) and dermal contact is assumed to be limited (i.e., compared to via inhalation), but can be influenced by other simultaneous chemical exposure. Once in the body, PAHs can migrate to all the tissues of the body that contain fat, but are usually found in the kidneys, liver, and fat (with smaller amounts stored in the spleen, adrenal glands, and ovaries). Based on animal studies, intestinal absorption of PAHs is believed to be highly dependent on the presence of bile.

PAHs will be metabolized by the body into different substances, some of which are more harmful than the original PAH, and some that are less harmful. PAHs also tend to leave the body within a few days through feces and urine (ATSDR 1995).

2.3 Health Effects

The major concern regarding PAH contamination in the environment and resulting exposure is that some PAHs are considered probable human carcinogens (i.e., substances capable of causing cancer) and currently include:¹

- benz[a]anthracene;
- benzo[a]pyrene;
- benzo[b]fluoranthene;
- benzo[k]fluoranthene;
- chrysene;
- dibenz[a,h]anthracene; and
- indeno[1,2,3-c,d]pyrene.

These PAHs, as well as others, are regulated by the USEPA and other governmental agencies to protect human health (ATSDR 1995), and are typically weighted in risk assessments relative to benzo[a]pyrene using toxic equivalency factors (TEFs).

Importantly, the USEPA Integrated Risk Information System (IRIS) released an updated toxicological review of benzo[a]pyrene in January 2017 which is likely to influence views on risks associated with environmental PAHs. The USEPA document updates its review of the publicly available studies to assess the potential adverse health effects of the compound and characterize exposure-response relationships. A summary of the USEPA review follows.

At sufficiently high doses, oral exposure to benzo[a]pyrene has been observed to cause developmental, neurobehavioral, reproductive, and immunotoxic effects in animals and humans. Adverse non-cancer health effects of inhalation exposure to benzo[a]pyrene are similar and include observations of developmental and reproductive toxicity in animal and human studies. As a suspected human carcinogen, high oral doses to benzo[a]pyrene have caused tumors in the forestomach, liver, oral cavity, jejunum or duodenum, and auditory canals of rats. Carcinogenic impacts of the chemical from inhalation have been observed in hamsters and include squamous cell neoplasia in the larynx, pharynx, trachea, nasal cavity, esophagus and forestomach (IRIS 2017).

While benzo(a)pyrene is still considered a likely carcinogen, USEPA has decreased its published estimate of the oral cancer potency slope factor from 7.3 kg-d/mg to 1.0 kg/d-mg. As noted above, the USEPA considers benzo[a]pyrene as an index chemical to derive relative potency factors to estimate the carcinogenicity of other

¹ Additionally, naphthalene is ranked as a possible (Class C) human carcinogen, but EPA provides no quantitative assessment of its cancer potency in the Integrated Risk Information System database. EPA proposed in 2010 to revise the Toxicity Equivalency Factor (TEF) approach to include a larger number of PAHs (<https://cfpub.epa.gov/ncea/risk/recordisplay.cfm?deid=194584>), but has not pursued the matter beyond receipt of comments.

PAHs. By implication, the update to the benzo(a)pyrene potency factor is likely to result in decreased cancer risk estimates for PAHs, and increases in target risk-based concentrations in soil and other environmental media (IRIS 2017).

For non-cancer endpoints, the impact of the USEPA review is an increase in both the reference doses (RfDs) for oral exposure (0.0003 mg/kg-day) and inhalation exposure (0.000002 mg/kg-day), which in effect, reduces estimates of potential health hazards associated with benzo[a]pyrene (and other PAHs, by TEF analogy) (IRIS 2017).

Based on this new review, USEPA indicates that humans should limit their oral exposure/dose of benzo[a]pyrene to 0.0003 mg/kg-day to avoid adverse non-cancer health effects. At an incidental soil ingestion rate of 50 mg each day, a benzo[a]pyrene equivalent concentration of 420 mg/kg is necessary for a 70 kg adult to reach the non-cancer health effects reference dose of 0.0003 mg/kg-d. However, notwithstanding consideration of background exposure and child exposure factors, it is likely that carcinogenic risks will continue to drive risk-based levels of concern for PAHs in soil (IRIS 2017).²

Therefore, using the procedures and assumptions outlined in Appendix B of NHDES's Risk Characterization and Management Policy for contaminated soils, a benzo[a]pyrene equivalent concentration of 0.9 mg/kg in soil is necessary for a resident (age 2 - 31) to reach an incremental cancer risk of 1×10^{-6} (one per million) with the recent updates to toxicity values.³ This concentration is not significantly higher than the 0.7 mg/kg soil concentration already implemented by NHDES as the S-1/Soil Remediation Standard, which regulates the allowable concentrations of contaminants in soil (see Section 4.3.2 for additional information).

As typical with many chemicals that pose a risk to human health, the young and old populations are potentially more susceptible to adverse health effects, especially those with conditions that make them susceptible to induction or nutritional deficiencies, those with genetic diseases that influence the efficiency of DNA repair, and those with immunodeficiency due to age or disease. Additionally, people who smoke, have a history of excessive sun exposure, liver and/or skin disease, and/or women (especially those of child bearing age) are more susceptible to the adverse health effects of PAHs (ASTR 1995).

² Exposure to the level of the RfD of 0.0003 mg/kg-d for a period of 30 years over a 70-year average lifetime corresponds to an incremental cancer risk of 1.3×10^{-4} , or 130 per million, using the updated cancer potency slope factor of 1 kg-d/mg – well above the 1 or 10 per million risks that are typically used as levels of regulatory concern.

³ Per default NHDES assumptions, exposure frequency is assumed to be 160 days per year, with assumed incidental soil ingestion rates of 200 mg by young children (2 to 6 years old) and 100 mg by older children and adults (for a total exposure period of 30 years). Dermal exposure is also considered at substantial (conservative) rates.

3.0 SOURCES AND EXTENT OF PAHS IN ROADSIDE SOILS

The extent of PAH contamination within soils adjacent to asphalt pavement (i.e. roadways, driveways, parking lots) has been extensively researched since the 1980s, and is a current emerging concern throughout the United States. It is generally accepted that vehicle emissions and deposition of these airborne particulates that contain PAHs along the roadside is a major source of contamination for soils adjacent to roadways. Transport of larger particulates from roadways to surrounding soils through stormwater flow, or snow removal operations (i.e. mechanical means) in northern climates may also be a source of PAH contamination. These larger particulates that are being transported may contain levels of PAHs from pieces of asphalt, small pieces of tire, motor oil, or coal-tar-based sealants. PAHs have been identified at ppm-levels in crumb rubber products (Zhang 2008), indicating their likely presence in tires, and therefore as a possible source of PAHs to roadside soil through tire wear.

While parking lots and paved driveways are impacted by the PAH sources described above, it is the use of coal-tar-based sealants on these surfaces that is considered the major source of PAH contamination in the sediments around these urban features. The United States Geological Survey (USGS) has completed extensive research related to the use of these sealants and the associated environmental and health impacts.

A brief discussion of the potential PAH concentrations in asphalt, coal-tar-based sealants, and vehicle emissions is provided below, followed by a more in depth review of research completed on the extent of PAHs in roadside soils as it relates to NHDOT's concerns regarding LRS.

3.1 PAH Concentrations in Asphalt

Overall, there is limited research available on PAH concentrations in asphalt. Asphalt (i.e., bitumen) is the non-distillable fraction of crude oil and is a complex mixture comprised mostly of carbon and hydrogen. It can be added to aggregates to create asphalt concrete for construction of asphalt paved roadways. Kriech et al. (2002) observed levels of total PAHs in the range of 1.9 to 66 mg/kg in asphalt pavements, which are in agreement with the typical level of PAHs found in petroleum-based asphalt sealant (i.e., 50 mg/kg) (Mahler 2016). Fluorene, phenanthrene, anthracene, pyrene, benzo[a]anthracene, chrysene, 5-Methylchrysene, and benzo[g,h,i]perylene were all observed by Kriech et al. (2002) in detectable quantities, though benzo[a]pyrene and dibenz[a,h]anthracene, the PAHs with the highest TEFs, were not detected. In addition, the Asphalt Institute has summarized the levels of commonly measured PAHs in various bitumens in a 2015 report of the bitumen industry, which included detectable levels of naphthalene, fluorene, phenanthrene, pyrene, benzo[e]pyrene, and benzo[g,h,i]perylene (Asphalt 2015). This report also noted that crude oil, and therefore bitumen, generally contains low levels of PAHs. Sadecki et al. (1996) and Kriech et al. (2002) also observed low potential for leachability of these PAHs in asphalt pavement to surrounding soils/water, with only naphthalene and phenanthrene observed at low levels in 3 of the 10 samples analyzed by Kriech et al.

3.2 PAHs in Coal-Tar-Based Pavement Sealants

The United States Geological Survey (USGS) has completed extensive research on the impacts of using coal-tar-based sealants on pavements because they are considered a major source of PAH contamination in urban areas. PAHs are transported from these sealed pavements through stormwater runoff, adhesion to tires or feet, wind, and volatilization. While not the focus of this Technical Guidance, it is important to note that these coal-tar-based sealants are the largest source of PAH contamination to 40 urban lakes researched by USGS (accounting for 50 percent of all PAH inputs) and the use of these sealants since the 1960s is the primary cause of the trend of increasing PAH concentrations in urban lake sediment (Watts 2010, USGS 2011)

Additionally, these sealants wear into small particles with high concentrations of PAHs that can be tracked into homes and incorporated into house dust. This can be a major pathway for human exposure and research has shown that PAH levels can be 25 times higher in house dust for an apartment adjacent to a parking lot sealed with coal-tar-based sealant as compared to a parking lot that has not been sealed.

Coal-tar-based sealants typically contain 50,000 to 100,000 mg/kg total PAHs, while petroleum asphalt-based sealants (used primarily west of the Continental Divide of the United States) typically contain about 50 mg/kg total PAHs (Mahler 2016). Based on their elevated concentrations of PAHs, and the potential for significant environmental contamination, states such as Minnesota and Washington have banned the use of these coal-tar-based sealants while more local bans have been implemented in jurisdictions/towns/counties in other states like Massachusetts and Texas (Minnesota 2016).

3.3 PAH Concentrations in Vehicle Emissions

Vehicle emissions have been studied extensively for a variety of compounds and are highly regulated by the USEPA. A wide range of PAHs can be found in the particulate matter of vehicle emissions depending on the type and age of the vehicle, type and brand of fuel, operating conditions, and time of year (Chellam 2005, Cadle 1999, Marr 1999). Correlation has been observed between increased PAH concentrations and increased particulate matter emission rates (Cadle 1999). Generally, all of the 16 USEPA priority PAHs were observed in emissions of particulate matter and vapor with a trend towards higher concentrations of the more volatile LMW PAHs (Cadle 1999, Marr 1999). Both fluoranthene and pyrene were identified as potential molecular markers for distinguishing between diesel emissions and gasoline emissions, as these PAHs were observed at increased levels in diesel emissions as compared to the emissions from gasoline (Marr 1999, Cadle 1999). In fact, diesel particulate matter is reported to contain 208-558 ppm of benzo(a)pyrene, as well as substantial concentrations of the other potentially carcinogenic PAHs (USEPA 2002).

3.4 Extent of PAHs in Roadside soils

The horizontal and vertical extents of PAHs and other contaminants in roadside soils (i.e., LRS) have been researched in a variety of countries around the world, with a large portion

of the research completed in Europe. Results from multiple research efforts, summarized in Table 1 below, indicate that the highest concentrations of PAHs were measured within a few meters of the roadways with a steep/exponential reduction in concentrations beyond that distance (Benfenati 1992, Harrison 1985, Johnsen 2006a, Clement 2015, Dierkes 1999, Pathirana 1994).

Table 1. Summary of Reviewed Research Related to Extent of PAHs in Roadside Soils

Location	Road Name/Type	Daily Traffic	Source
Italy	Highway	18,000	(Benfenati 1992)
Italy	State Road	11,500	(Benfenati 1992)
England	M6 Motorway	-	(Harrison 1985)
Denmark	Motorway 14	35,800	(Johnsen 2006a)
France	A71 Motorway	-	(Clement 2015)
Germany	Highway	5,200 - 107,600	(Dierkes 1999)
Australia	Kessels Road	-	(Pathirana 1994)

Recent research from southern China concluded that the degree of traffic congestion, rather than the traffic volume influenced the concentrations of 5 and 6 ring PAH compounds in roadside soils (Liu 2016). The researchers hypothesized that this was due to brake and tire wear, and incomplete fuel combustion resulting from the driving patterns of traffic congestion conditions (i.e., frequent stops).

Topographic and vegetative features of the surrounding area may also impact deposition of PAHs and the extent of contamination along roadways. Researchers in Germany concluded that embankments influenced the extent of PAH deposition (Diekes 1999). Multiple other researchers have found that forest/trees along the roadside may act like a “green barrier” and limit the extent of PAH contamination (Clement 2015, Zehetner 2009, Wilcke 2000). It was theorized that the wax-covered coniferous needles of the study forest acted like a filter for PAHs transported within airborne soot particles from vehicle emissions (Zehetner 2009). This “filter” process resulted in increased PAH concentrations at the road-forest interface.

Vertical extent of PAHs was found to be limited within roadside soils, with the highest PAH concentrations within the first layer of soil (i.e. 5 to 10 cm below ground surface) followed by a rapid decrease with depth (Dierkes 1999, Pathirana 1994).

We note that conditions (i.e., type of vehicles, type of fuel, roadway design, roadway construction materials, travel patterns, vehicle emission standards, etc.) in Europe and other countries may not be fully representative of those in the United States, and more specifically, New Hampshire.

3.5 PAH Background Concentrations

When assessing the horizontal and vertical extent of PAHs and other contaminants in roadside soils (i.e, LRS), it is important to know what the PAH background concentration in the existing soils may be. We understand that the potential for PAH contamination in soils

may be widespread due to the variety of anthropogenic sources in both rural and urban areas. At this time, there is no PAH background concentration data in New Hampshire, however there is limited data for the surrounding states of Maine, Massachusetts, and Vermont. A brief summary of this data is provided in Table 2 below. We note that the three states used varying methods to gather and analyze this data and it may not be comparable.

Table 2. Summary of Background Concentrations of “Residential/Rural” Soil (Mass DEP 2002, AMEC 20012, VTDEC 2015)

State	Average Total PAH Concentration (mg/kg)	Average Benzo[a]pyrene Concentration (mg/kg)
Maine ⁴	4.7	0.5
Massachusetts ⁵	26.5	2.0
Vermont ⁶	0.18	0.02

4.0 MOBILITY AND TRANSPORT

The mobility and transport of contamination in the environment is influenced by a variety of factors, such as environmental conditions (e.g., pH, presence of other chemicals, oxidation-reduction potential, groundwater geochemistry, organic matter content, microorganisms) and the physical and chemical properties of the contaminant itself. Mobility can be defined as the potential for a contaminant to migrate from a source. In the case of PAH soil contamination, mobility is most influenced by the organic carbon content of the soil and the hydrophobic nature of many PAHs (Wilcke 2000, Abdel-Shafy 2016, Wang 2001, Wang 2013, Ahens 2004, Abdel-Shafy 2016, Nam 2008, Xiao 2014, Maxin 1995, Li 2009). Both of these influences relate to the solubility in water, and adsorption and desorption processes for PAHs that dictate the bioavailability and transport of the contaminant in soil.

The sections below summarize the research that has been reviewed to assess the influence of particle size and organic carbon content of soils on the mobility and transport of PAH contamination. We have also reviewed research completed related to the leaching potential of PAHs during laboratory conditions.

4.1 Influence of Soil Particle Size

There has been extensive research into the correlation of particle size of contaminated soil and PAH concentration with varying conclusions. The strong influence of organic carbon content and quality in soils, which will be discussed in further detail in Section 4.2, can inhibit observing correlations or patterns related to particle size and PAH concentration in the soil. Wang et al. (2013) and Ahrens et al. (2004) found that soil PAH concentrations

⁴ Values are the sum of the average concentrations of each of the individual PAHs analyzed, from Table 4-1.

⁵ Values are the sum of the provided concentrations of each of the individual PAHs analyzed, from Table 1.

⁶ Values are the sum of the average concentration of each of the individual PAHs analyzed, from the analytical data reports.

were unevenly distributed among particle-size fractions and were more influenced by soil organic carbon content.

In general, PAHs can accumulate in the silt, clay, and sand fractions of contaminated soil and the HMW PAHs are found in larger proportions in the smaller particle size fractions of the soil. LMW PAHs are conversely found in larger proportions in the larger particle size fractions of the soil (Wilcke 2000). A summary of the soil particle sizes that were found to contain higher PAH concentrations is provided in Table 3.

Table 3. Soil Particle Sizes that Contained Higher Concentrations of PAHs

Location	Size of Particle (µm)	Description	Source
Boston Harbor, Massachusetts	>250	Sand	(Wang 2001)
California, Wisconsin, and New York Harbor Sediments	-	Low density material	(Ghosh 2003)
Norwegian Harbor Sediments	>75	Sand	(Oen 2006)
Auckland, New Zealand Harbor Sediments	125-150	Fine sand	(Ahrens 2004)
Auckland, New Zealand Harbor Sediments	-	Low density material	(Ahrens 2004).
China	75-2,000	Fine/coarse sand	(Wang 2013)
Beijing, China	250-500	Medium sand	(Li, 2009)
Yangtze River Delta, East China	-	Low density material	(Ni 2008)
Bangkok, Thailand		Low density material	(Muller 2000)

As noted in the table above, PAH concentrations were analyzed for the low density fractions (i.e., the fraction of soil separated through flotation methods) of contaminated soils in four cases. This fraction of the soil, while encompassing a relatively small proportion of the soil mass, was shown to contain the highest concentrations of PAHs and organic carbon (Ahrens 2004, Ni 2008, Muller 2000). The affinity of PAHs to adsorb to this organic carbon rich portion of the sediments may relate to PAHs tending to accumulate in the organic carbon rich topsoil layers of surface soils in the environment (Wilcke 2000). Wang et al. (2001) noted that organic matter from charcoal, plant detritus, and some fecal pellets from organisms in the soils sorbed PAHs better and stronger than other organic matter associated with clay minerals.

When a correlation can be made between PAH concentration and particle size, it is typically also noted that the organic matter associated with the different particle size fractions is probably controlling the PAH concentration in that fraction of the soil, rather than the size of the particles (Wang 2001, Ahrens 2004, Wang 2013, Wilcke 2000).

4.2 Influence of Organic Carbon Content

The organic carbon content of soils may influence PAH mobility and transport in contaminated soils to the largest extent. A variety of research has been completed that observed a statistically significant relationship between PAH concentration in soils and the total organic carbon (TOC) content of the soil (Nam 2008, Xiao 2014, Maxin 1995, Li 2009, Wilcke 2000). In conjunction with this observation, some researchers have concluded that rather than TOC concentration, it may be the black carbon (BC) content of the soils that dictates the corresponding PAH adsorption and concentration (Johnsen 2006a, Johnsen 2006b, Li 2009, Wilcke 2000, Oen 2006). BC is particulate matter that is formed by the incomplete combustion of fossil fuels, biofuels, and biomass. Therefore, PAHs emitted in vehicle emissions may combine with BC and become strongly adsorbed to the particles before deposition in roadside soils occurs. This could render the PAHs inaccessible to soil microbes, and potentially humans as well, which will lead to increasing concentrations in soils over time (Johnsen 2006a). The molecular weight of the individual PAHs will dictate some of this behavior and affinity to be adsorbed to the organic carbon content in the soil. Typically, the HMW PAHs have a greater tendency to adsorb to the organic carbon content (Maxin 1995, Johnsen 2006a).

It should be noted that while there is general acceptance of the organic carbon content of soils influence over PAH concentration in the soil, there has been some research that was unable to draw this conclusion. Surface soil samples collected from roadsides in Shanghai, China found no correlation between PAHs and total organic carbon content (Jiang 2009). Additional research out of Shanghai China agreed with this research, and suggested that the lack of correlation specifically to total organic carbon could be due to the PAHs being so strongly adsorbed and impacted by the black carbon content only of the soils (Wang 2015). Xiao et al. (2014) concluded that the anthropogenic source of the PAHs may affect the concentrations in soils rather than the organic carbon content (Xiao 2014).

4.3 PAH Desorption

While we understand that PAHs tend to adsorb strongly to organic carbon within soils and have low solubility in water, PAHs are still encountered in some sub-soils (Wilcke 2000). Johnsen et al. (2006b) observed LMW PAHs (i.e., higher solubility PAHs) leaching below the top horizon of a soil column, however the HMW PAHs (i.e., lower solubility and carcinogenic compounds) were only found within the top horizon. Therefore, desorption, whereby a substance is released from a surface, is occurring for some PAHs in soils. However, Wilcke et al. (2000) has suggested that an alternative method of PAH movement in soils may be the transport of PAHs associated with dissolved organic matter, or particle-associated PAH transport of HMW PAHs rather than actual desorption. In support of this theory, research from France suggests that infiltration of water in soil may solubilize PAHs which are then leached downward in soil, however during this vertical transport, PAHs may be re-adsorbed by the soils in the deeper horizon. This results in accumulation of PAHs in deeper soils without actual leaching of the PAHs out of the soil column (Saison 2004).

To assess the potential PAH desorption/mobility in soils and consequent groundwater contamination potential, a variety of leaching tests have been completed by researchers in recent years and are summarized below.

4.3.1 Leaching Tests

Column leaching tests were completed on silty-loam soils from Berlin by Zand et al. (2010) and in agreement with the tendency of PAHs to stay adsorbed to organic carbon, only about 2-4% of the total initial amount of PAHs were leached from the soil. However, the leaching behavior of the individual PAH compounds was more varied, with the initial released amounts decreasing with increasing molecular weight of the PAH (i.e., with increasing hydrophobicity). As such, benzo[a]pyrene concentrations were undetectable in the leachate from the experiment (Zand 2010).

Soil from a former gasworks plant in Sweden was also tested using a column leaching test method, under field-like conditions. In this experiment, only 0.3% of the initial amounts of PAHs in the soil were leached. This test ran for over 1,600 hours with similar conclusions as above with regard to release amounts and PAH molecular weight. Benzo[a]pyrene and benzo[g,h,i]perylene were both undetectable in the leachate from the experiment (Enell 2004). It was concluded that PAHs in aged contaminated soils were not expected to be released to any great extent by leaching with only water.

A vertical soil column experiment was completed on soils from the United Kingdom with simulated rainwater conditions. Consistent with the above summarized research, the only PAH that was found in the leachate after 26 days was naphthalene, which is consistent with the low solubility characteristics of the HMW PAHs. These researchers were able to assess that only 0.2-2.3% of the PAHs were transported below the surface layer of the soil column. These results led to the conclusion that surface applied PAHs reaching and contaminating a groundwater aquifer was unlikely (Revitt 2014).

Jonker et al. (2005) found that combustion-derived PAHs are strongly sorbed to particulates (i.e. soot), which results in very slow desorption kinetics. It was hypothesized that it could take millennia for the removal of these types of PAHs from soil. After 120 minutes of supercritical fluid extraction testing on “traffic soot” samples from a manufactured gas plant, less than 1% of the carcinogenic PAHs, relative to the total adsorbed mass of PAHs, were released.

There was only one study at a former manufactured gas plant site, in Santa Barbara, California, that concluded a higher amount of total PAHs, 10% of the total, were leachable with less than 3% of the benzo[a]pyrene detected in the leachate (Stroo 2000). This soil was also aged with a higher abundance of HMW PAHs.

4.3.2 NHDES Leaching-Based Soil Values

The New Hampshire Department of Environmental Services (NHDES) has established leaching-based soil concentration values for contaminants of concern that are intended to be protective of groundwater quality. These values are used in conjunction with direct contact risk-based (DCRB) soil concentrations and estimated quantitation limits to regulate

the allowable concentration of contaminants in soil.⁷ The NHDES uses three different soil categories (S-1, S-2, and S-3) to describe the range of potential human exposure situations at varying types of locations for contaminants. S-1 standards, which are based on a conservative residential exposure scenario as well as protection of groundwater, and practical laboratory quantitation limits, have been implemented as the Soil Remediation Standards (SRS) (New Hampshire Code of Administrative Rules Env-Or 606.19). Hence, SRS values are at or below the concentrations of contaminants in soil that are protective of groundwater quality (i.e., such that the soil will not leach a sufficient mass of a contaminant which would result in a violation of the NHDES ambient groundwater quality standards [AGQS] - New Hampshire Code of Administrative Rules Env-Or 603.03) (NHDES 2013).

The S-1 residential exposure scenario used above is protective of young children (2-6 years old) who may come into direct contact (i.e., dermal and ingestion) with soil through playing in a yard for non-carcinogenic effects, and protective of residents (2 to 31 years old) who may come into contact with soil through playing in a yard (for young children), and by working/gardening in a yard (for adults) for carcinogenic effects (NHDES 2013)

The leaching-based soil values calculated by NHDES, and used as one of the criteria to establish the SRS (as described above) were developed using the Seasonal SOIL Compartment Model (SESOIL) and 3-Dimensional Waste Transport in Aquifers (AT123D) contaminant transport models with generic site characteristic inputs considered reasonably conservative for New Hampshire conditions. The model assumed a water table approximately 3 m below ground surface and a contaminated area of 10 square meters with contamination residing 1-2 m below ground surface at a concentration of 10 parts per million (ppm). The drinking water well of concern was modeled 10 m down gradient from the contaminated area (NHDES 2013). The modeled peak groundwater concentration was used to calculate a dilution and attenuation factor (DAF), from which the leaching-based soil value could be derived.

Results for the USEPA's list of 16 PAHs in the table below indicate that most of these PAHs displayed negligible contaminant migration (NCM), which means that the modeled release rate into groundwater from the vadose zone was not measurable within 30 years. These modeled and state approved results support the findings of the leachate testing described in Section 4.3.1. Acenaphthene, acenaphthylene, fluorene, and naphthalene were the only PAHs on the list to exhibit leaching potential, however none of these contaminants are considered carcinogenic to humans (NHDES 2013).

⁷ NHDES also accounts for naturally occurring background concentrations of selected metals.

Table 3. Leaching-Based Soil Values for Select PAHs (NHDES 2013)

PAH	Time to Reach Peak Groundwater Concentration (yrs)	Leaching-based Soil Value (mg/Kg)	Probable/Possible Human Carcinogen?
acenaphthene	28	344.26	Unknown-
acenaphthylene	30	488.37	NC
anthracene	-	NCM	NC
benz[a]anthracene	-	NCM	Yes
benzo[b]fluoranthene	-	NCM	Yes
benzo[k]fluoranthene	-	NCM	Yes
benzo[g,h,i]perylene	-	NCM	NC
benzo[a]pyrene	-	NCM	Yes
chrysene	-	NCM	Yes
dibenz[a,h]anthracene	-	NCM	Yes
fluoranthene	-	NCM	NC
fluorene	11.6	77.35	NC
indeno[1,2,3-c,d]pyrene	-	NCM	Yes
naphthalene	8.6	5.06	Yes
phenanthrene	-	NCM	NC
pyrene	-	NCM	NC

NC - not classifiable as to human carcinogenicity.

NCM - negligible contaminant migration.

Unknown - no classification for carcinogenic effects.

5.0 BIOAVAILABILITY AND BIOACCESSIBILITY

Based on the research discussed throughout this Technical Guidance, we understand that the bioavailability and bioaccessibility of PAHs is widely considered to be low due to the contaminants inclination to be adsorbed strongly to particulates and organic carbon in soils (Duan 2015, Muller 2000). Bioaccessibility can be defined as a sum of the actual bioavailable PAH concentrations and the amount of PAHs that are potentially bioavailable over time (Johnsen 2006b). This bioavailability/bioaccessibility of PAHs is dependent on the hydrophobicity of the individual PAH and how it is bound to the soil. The more water soluble the PAH, the higher the uptake may be for vegetation, however the reverse is true for organisms like earthworms (Wilcke 2000). Research from Denmark demonstrated only about 1-5% of the PAH concentration in soils was accessible to soil bacteria, believed to be due to strong adsorption of the PAHs to the black carbon within the soil (Johnsen 2006a).

There is the potential for PAH contamination in foods (i.e. fruits and vegetables) grown in contaminated soil, although bioavailability of PAHs is relatively low. Voutsas et al. (1998) and Kipopoulou et al. (1999) assessed the dietary intake of PAHs from cabbage, carrots, leeks, lettuce, and endives grown in contaminated soil from an industrial area in northern Greece. LMW PAHs were detected at higher levels than HMW PAHs, in part due to their higher water solubility in soil, but also more importantly due to atmospheric deposition. The PAHs in the vegetable tissues were more correlated to the concentrations observed in the atmosphere rather than those observed in the soil. This suggests that deposition from the atmosphere was the principal pathway for accumulation of PAHs in plants. This finding is supported by the concept that LMW PAHs are more likely to be present in a gaseous phase in the atmosphere rather than in soil.

While HMW PAHs are also present in the atmosphere, they are generally associated with larger particulates that will only be superficially deposited on the plant foliage and can be washed away by rainwater. The gaseous LMW PAHs can more easily penetrate the cuticular surface of foliage, which also leads to the highest PAH concentrations observed in the leafy vegetables (i.e., vegetables with larger surface areas exposed to deposition) (Wilcke 2000, Voutsas 1998, Kipopoulou 1999). However, research from France has noted that PAH soil contamination may be more correlated to PAH concentrations in the vegetable when looking specifically at root vegetables. This is due to the strong adsorption of PAHs to the root epidermis rather than actual uptake of the PAHs (Fismes 2002). In support of this conclusion, Samsøe-Petersen et al. (2002) found that peeling potatoes and carrots significantly reduced the concentrations of benzo[a]pyrene and indeno[1,2,3-c,d]pyrene. Voutsas et al. (1998) noted that washing vegetables with water and peeling root vegetables may reduce PAH concentrations in the consumed foods, however PAH concentrations may still remain even after this typical household preparation.

Therefore, to reduce potential exposure to PAH contamination, the public should make sure to wash vegetables thoroughly with water, and peel vegetables when applicable, especially when consuming the root of any plant. While we have focused on PAH contamination of vegetables, these practices should also help reduce the risk associated with other potential pollutants in vegetables.

6.0 STATE REGULATION

The NHDES exempts the collection, storage, transfer, processing, treatment, and disposal of waste concrete, cement, brick, other inert masonry materials, and bituminous concrete (i.e. asphalt pavement) from the requirements of a solid waste permit under Env-Sw 302.03(b)(9). This exemption requires that the materials are fully cured, the waste is actively managed (and management practices comply with universal facility requirements in Env-Sw 1000), and the waste is free of any materials or substances that have the potential to leach contaminants to groundwater or surface water, or to emit pollutants to the air, including lead paint, asbestos, and chemicals (see Env-SW 302.03(b)(9) for additional requirements). As such, NHDES does not oversee cured asphalt in NHDOT roadways (whether in situ or buried), until the materials are removed from the original location. However, it should be noted that if the bituminous concrete is ground or pulverized, it is no longer exempt from a solid waste permit when landfilled

In New Hampshire, the only unregulated soils are virgin soils/rocks from gravel pits and quarries, or virgin excess soils, free of anthropogenic impacts. As such, soil along the roadsides that has been contaminated (i.e., LRS) may be considered contaminated if containing concentrations greater than background/naturally occurring levels. Contamination is defined in Env-Or 602.07 as the “presence of any regulated contaminant, as defined herein, other than naturally occurring substances at naturally occurring or background levels, in soil, groundwater, soil gas, air, sediment, surface water, construction/excavation debris, or any other material at a concentration that has the potential to adversely affect human health or the environment”. Background is further defined as the “concentration of a chemical in the environment that would exist at a site in the absence of a discharge, including chemicals that are ubiquitous and consistently present at or in the vicinity of the site”. This may include petroleum residues that are incidental to the normal operation of motor vehicles and asphalt pavement and petroleum compounds contained in associated sub-base materials.

While the LRS may be considered “background” while in place and not under the specific purview of NHDES, the moment that soil is moved, it becomes a regulated soil that must be managed on-site in a manner consistent with Env-Or 600, transported off-site to a permitted disposal/treatment facility, or the area should be operated as a soil landfill, under a NHDES solid waste facility permit.

As an alternative, NHDOT is currently working with NHDES to request waivers from some of the above requirements of the solid waste rules. Both NHDOT and NHDES understand the unique nature of LRS, the potential prevalence and persistence of the material around roadways, and the limited potential for adverse human health and environmental effects from exposure/leaching of PAHs. NHDOT already implements dust control via water application at worksites to help reduce the impacts of work along roadways (i.e., reduce the potential for inhalation and/or transport of particulates that contain PAHs).

7.0 SUMMARY

LRS along roadways has the potential to contain varying levels of PAHs and other contaminants. However, PAHs are prevalent in the environment and can even be found at low levels in samples of rural/residential soil. The USGS has concluded that coal-tar-based pavement sealants, used predominantly on driveways and parking lots, are a major source of PAH contamination in urban areas. It is these sources in close proximity to residential areas that may cause more adverse human health effects than LRS along roadways. Based on the research reviewed as part of this Technical Guidance, PAHs in LRS tend to stay adsorbed to the organic carbon content of the soils and show low potential for leachability, as supported by NHDES’s modeling of potential mobility of PAHs such as benzo[a]pyrene.

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