

# Assessment of Water Quality of Runoff from Sealed Asphalt Surfaces



# SCIENCE

# **Assessment of Water Quality of Runoff from Sealed Asphalt Surfaces**

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## **Abstract**

This report discusses the results of runoff tests from recently sealed asphalt surfaces conducted at the U.S. Environmental Protection Agency's (EPA) Urban Watershed Research Facility (UWRF) in Edison, New Jersey. Both bench-scale panels and full-scale test plots were evaluated. Full-scale tests were performed on an asphalt portion of the UWRF parking lot; no parking was allowed on any of the surfaces to minimize cross-contamination from other sources. A variety of water quality analyses of the runoff were conducted. The whole water sample was analyzed rather than analyzing the particle and dissolved phases separately. The primary measurement was polycyclic aromatic hydrocarbons (PAHs). Sealants applied to asphalt surfaces leached measurable quantities of PAHs. Results indicated that the time from the initial sealant application is a major factor in observed PAH concentration in runoff. The highest PAH concentrations measured were in initial runoff samples where sampling was performed twenty-four hours after application of sealants to the asphalt surface. Toxicity screening assays produced inconclusive data due to matrix effects of prepared samples.

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## Foreword

The U.S. Environmental Protection Agency (EPA) is charged by Congress with protecting the Nation's land, air, and water resources. Under a mandate of national environmental laws, the Agency strives to formulate and implement actions leading to a compatible balance between human activities and the ability of natural systems to support and nurture life. To meet this mandate, EPA's research program is providing data and technical support for solving environmental problems today and building a science knowledge base necessary to manage our ecological resources wisely, understand how pollutants affect our health, and prevent or reduce environmental risks in the future.

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Sally C. Gutierrez, Director  
National Risk Management Research Laboratory

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## Acronyms and Abbreviations

APWA	= American Public Works Association
AR	= Annual Runoff
BOD	= Biochemical Oxygen Demand
CF	= Correction Factor
COD	= Chemical Oxygen Demand
C <sub>PAH</sub>	= Polycyclic Aromatic Hydrocarbon Concentration
CWA	= Clean Water Act
DO	= Dissolved Oxygen
EPA	= United States Environmental Protection Agency
K <sub>OW</sub>	= Octanol Water Partitioning Coefficient
NPDES	= National Pollutant Discharge Elimination System
NPS	= Nonpoint Source
NRMRL	= National Risk Management Research Laboratory
NURP	= Nationwide Urban Runoff Program
ORD	= Office of Research and Development
ORISE	= Oak Ridge Institute of Science and Education
PAH	= Polycyclic Aromatic Hydrocarbon
PEC	= Probable Effect Concentration
SC	= Stormwater Center (of University of New Hampshire)
SM	= Standard Methods
SOP	= Standard Operating Procedure
TMDL	= Total Maximum Daily Load
TOC	= Total Organic Carbon
TPAH	= Total Polycyclic Aromatic Hydrocarbons
TSS	= Total Suspended Solids
U.S.	= United States
USGS	= U.S. Geological Survey
UWRF	= Urban Watershed Research Facility
VSS	= Volatile Suspended Solids
WQS	= Water Quality Standards
WWF	= Wet-Weather Flows

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## Executive Summary

### ***Background***

Determining the impact of wet-weather discharges on receiving water quality remains an elusive goal given the various potential pollutants in the urban environment and the common practice for many storm drainage systems to discharge to the nearest receiving water with little or no treatment. In general, stormwater runoff from roads and parking lots has been shown to have high levels of pollutants and has been documented to be toxic to both freshwater and marine organisms. This document could be used to assist in the determination of the potential impact to receiving water quality from stormwater runoff in urban areas due to asphalt sealant use.

### ***Sealants as Potential Sources of Water Quality Impairments***

Asphalt pavement sealants are applied to parking lots and driveways to enhance appearance and protect surfaces and are widely used in commercial and residential products. Due to the tendency of these coatings to wear over time, manufacturers recommend reapplication of sealants to surfaces every two to three years. There are two types of sealcoats generally used in the U.S. today: asphalt emulsion and coal tar emulsion. Coal tar has been shown to have a detrimental effect on the overall health of a variety of aquatic organisms. Recent literature has suggested that coal tar-based asphalt sealants have impacted survival and development of amphibians, embryo and larval mortality in fish, and growth and biodiversity of macroinvertebrates and benthic phytoplankton. The primary components of coal tar that are presumably responsible for these toxic effects are polycyclic aromatic hydrocarbons (PAHs).

### ***Why EPA?***

This research project was conducted by the Water Supply and Water Resources Division of the Office of Research and Development's National Risk Management Research Laboratory. PARS Environmental Inc., an on-site contractor at EPA's Urban Watershed Research Facility (UWRF) in Edison, New Jersey, performed sampling, analysis and logistical support. The UWRF had an existing unsealed asphalt parking lot specifically designed to assist scientists and engineers in the collection of runoff. The parking lot was modified specifically for this project so that separate sections of runoff from test plots could be collected concurrently.

### ***Analysis***

The primary analyses conducted in this study were for the PAH content of the collected samples. A range of other water quality constituents were also measured. Toxicity analysis through the use of a Microtox® screening unit was also performed, though the results of these analyses were inconclusive due to matrix effects of the prepared samples. These toxicity results are therefore not reported.

### ***Experimental Design***

The project was initiated with the development of bench-scale testing. The project culminated with a full-scale, six-month study of three asphalt test plots with different or no surface treatments: coal tar emulsion sealant, asphalt emulsion sealant, and an unsealed control. Both the bench- and full-scale studies were tested over a time period of 1 to 30 days after application of sealants. The full-scale study had additional testing of test surfaces at six months.

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## ***Results***

The products examined in this study are a subset of the products available on the market and do not represent all products. Asphalt emulsion- and coal tar emulsion-based sealcoat products are the most widely used in the U.S. Coal tar products have PAH levels about 1,000 times higher than the asphalt sealcoat (Mahler et al., 2005). Precise national use is not known; however, USGS data suggest that asphalt-based sealcoat is more commonly used in the western U.S. and coal tar-based sealcoat use is more common in the other regions of the U.S. (Van Metre et al., 2005). There may be differences in water quality parameters observed in the runoff from surfaces of other sealants; therefore, the results herein cannot be translated across sealant product lines.

Results of the full-scale study indicate that PAHs are present in the runoff of surfaces coated with sealants. The PAH concentrations in the runoff were observed to decrease with time. When focusing on samples immediately after recommended curing time (24 hrs), there are correspondingly higher concentrations of PAHs. The asphalt emulsion and unsealed control surfaces did not contain concentrations of PAHs of the same order of magnitude as found in the runoff from the coal tar sealant plot.

## ***Conclusions and Recommendations***

PAHs were observed in the runoff from all three testing surfaces. The findings were consistent between the full-scale and the bench scale studies.

- The coal tar-sealed surfaces released 100 to 1000 times more PAHs to the runoff than the other surfaces. This release of PAHs from the surface to the runoff diminished with time. There was a measurable shift in the individual PAH components in the runoff, with fewer lower molecular weight PAHs observed in the runoff over time.
- The initial wetting after sealing may be the most crucial flush of PAHs into the environment.
- Additional testing is warranted on a representative variety of asphalt emulsion products. Even though low levels of PAH were observed in relation to the coal tar sealant runoff, increased total organic carbon (TOC) and chemical oxygen demand (COD) loadings were observed for the initial runoff samples collected, indicating an increased organic chemical load being released.
- Measurement of COD and TOC water quality parameters cannot be used as surrogates to identify potential release of PAHs from sealed surfaces.
- It is recommended that toxicity assays be performed with a variety of representative organisms (invertebrates, amphibians, fish, etc.) using standard procedures. This would require significant technical and financial resources. This more intensive toxicity testing is needed in order to more fully understand the effects of exposure to runoff from sealed asphalt surfaces. The literature lacks an in-depth study of sealant runoff examining both coal tar sealants or asphalt emulsion alternatives and the potential for acute toxicity, or lack thereof, to aquatic organisms in the water.

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## Chapter 1 Introduction

### Background

The Clean Water Act (CWA) is the Nation's primary mechanism for protecting and improving water quality with the broad purpose of the CWA "to restore and maintain the chemical, physical, and biological integrity of the Nation's waters," (33 U.S.C § 1251 (a)). Several sections of the CWA apply to urban runoff, both as point and nonpoint sources of pollution.

Point sources, including municipal and industrial stormwater discharges, are controlled by the National Pollution Discharge Elimination Program (NPDES) permits (33 U.S.C § 1342 (p)). Because of the difficulty in identifying specific origins of pollution associated with nonpoint sources (NPS), mitigation of associated pollution is approached with management strategies. The CWA allows for both environmental quality and technology-based (treatment processes and best management practices) approaches for controlling water pollution. States are required to develop and adopt water quality standards (WQS) that specify the designated uses of each water body, and determine specific criteria deemed necessary to protect or achieve those designated uses. The CWA requires states to develop and implement WQS in accordance with EPA regulations and guidance.

Implementation of water quality programs at the federal, state, and local levels, along with accompanying research, that address both point and nonpoint sources continue to evolve with the cumulative knowledge of the impacts of urban development. Stormwater runoff from impervious surfaces in urban areas has emerged as a potential threat to water quality. Pollution problems stemming from these wet-weather flows (WWFs) are a challenge throughout the Nation. National estimates have projected costs for WWF pollution abatement in the tens of billions of dollars (APWA, 1992).

A variety of physical, chemical, and biological processes influence the type and the degree of impacts that urban WWFs can have on receiving-water capacity and aquatic ecosystems by directly or indirectly affecting the concentration of pollutants and organisms present (House et al., 1993). These processes include transport (by advection or diffusion), sedimentation, erosion, sorption, pH impacts, gas exchange, oxygen demanding pollutants, die-off and growth of organisms, bio-accumulation of contaminants in the food-chain, and species selection (Lijklema et al., 1993). Pratt et al. (1981) identified two plausible means, immediate and long-term, in which urban runoff can impact receiving waters and the aquatic ecosystems. Discharging untreated urban runoff containing solids, toxins, nutrients, and organic oxygen-demanding pollutants can have an immediate, i.e., "shock-loading effect," on receiving waters. The second long-term effect is the accumulation of contaminated sediments or the contamination of existing sediments. During a storm event, the depletion of oxygen in receiving water due to organic loading may occur; however, it is more likely to be a problem several days later due to increased sediment oxygen demand (Pitt 1979).

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Runoff from roads and parking lots has been shown to contain high levels of pollutants and to be toxic to both freshwater and marine organisms (Maltby et al., 1995; Pitt et al., 1995; Herricks et al., 1997; and Greenstein et al., 2004). Runoff has been known to be toxic for some time. Specifically, Spiegel et al. (1984) sampled receiving water and rainwater, combined sewage, and urban runoff. Nineteen out of 85 samples induced a detectable mutagenic response as measured by the Ames test (Ames, 1971). The greatest response was due to urban runoff as nine (47%) of these 19 samples were urban stormwater runoff, with 57% of these samples indicating a strong dose-related response. However, there is limited research on the potential toxicity of the individual components of pollution in urban stormwater runoff.

One component of urban stormwater runoff that may contribute to toxic loading is runoff from surfaces coated with asphalt sealants, which are often applied to asphalt parking lots and driveways for aesthetic purposes. Runoff from sealed surfaces may contain polycyclic aromatic hydrocarbons (PAHs). PAHs are a large class of typically hydrophobic compounds with varying degrees of water solubility (Aldstadt et al., 2002). PAHs are known carcinogens and are known to be toxic to aquatic life (EPA 1984; Long and Morgan 2000; and Ankley et al., 2003). It has also been known for some time that PAHs are a component of urban runoff (EPA 1983, and Pitt et al., 1995).

The overall objective of this project was to qualify and quantify pollutants in the runoff from an asphalt surface sealed with either coal tar or asphalt emulsions. This study concentrated on the immediate effect on water quality of the runoff from sealed asphalt surfaces and did not address the long-term issue of PAHs accumulating in sediment. Bench- and full-scale tests were performed at EPA's Urban Watershed Research Facility (UWRF) located on the grounds of the Edison Environmental Center in Edison, NJ. The UWRF has a parking lot, constructed in 1999, with drainage channels to assist researchers in collecting runoff samples.

This study used collected rainwater for all tests. Unlike other studies cited in literature that have allowed traffic on the surfaces, the parking lot surfaces were not parked or driven on for the duration of this study. This lot was parked on prior to the study, but was cleaned prior to study initiation. This avoided confounding variables such as surface abrasion, tire wear, and automotive exhaust.

### **Literature Review: Asphalt Sealant Studies**

Asphalt sealants are commonly used in the United States due to the prevalence of asphalt driveways and parking lots. Because of its tendency to abrade with time and use, asphalt sealants are used repeatedly over the same surface. It is often recommended that the sealants be reapplied to driveways and parking lots every two to three years (Dubey 1999). Scoggins et al. (2009) reported that an estimated 320 million liters of coal tar sealant are sold each year. Crenson (2007) used a conservative application rate of 1 liter per meter squared ( $L/m^2$ ). This leads to 160,000 kilograms (kg) of PAHs delivered to the environment each year if a sealant abrasion rate of  $0.51 \text{ g}/m^2/\text{year}$  is used (Scoggins et al., 2009). Coal tar emulsion sealants can contain up to 35% refined coal tar, which is made up of 50% PAHs by mass (NIST 2006). Asphalt emulsion sealants do not contain coal tar and usually contain less than 35% petroleum asphalt by weight; however, it is suspected that PAHs are present in these types of sealants as well, because asphalt contains PAHs (Mahler et al., 2004 and Wess et al., 2004). PAH concentrations in asphalt emulsion sealant have been measured at approximately 1,000 times less than those in coal tar sealant (Mahler et al., 2005).

Recent literature has suggested that coal tar-based sealants have contributed to alterations in the survival, growth, and development of amphibians (Bryer et al., 2006 and Bommarito et al., 2010). In one study, dried coal tar sealant flakes were added to water containing frog embryos (Bryer et al., 2006). The coal tar sealant flakes were added in low (approximately 3 ppm TPAH, where TPAH was total PAH, a sum of 16 parent PAHs), medium (approximately 30 ppm TPAH), and high concentrations (approximately 300 ppm TPAH), as well as a control with no sealant flakes. No frog embryos survived the high treatment, but the other treatments showed that embryos exposed to the sealant took longer to hatch and were developmentally delayed when compared to those in the control treatments. The study's authors could not directly link the toxicity seen to PAH concentrations, but inferred the link due to the large percentage that PAHs contributed to the makeup of sealants (20-35%).

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It has also been shown that coal tar contributes to embryo and larval mortality in fish (Kocan et al., 1996) and that coal tar inhibits the growth and biodiversity of macroinvertebrates and benthic phytoplankton (Oberholster et al., 2005). Crunkilton et al. (1997) reported PAH concentrations in runoff and observed that even low concentrations harmed organisms by weakening their immune systems and changing their metabolisms.

Greenstein et al. (2004) also examined runoff from parking lots after simulated rainfall and quantified PAHs, but did not note whether the parking lots were sealed. Lots were separated into low versus high use and maintained versus unmaintained for the purpose of examining the effect of antecedent dry period on the toxicity of runoff from the parking lots. Toxicity was evaluated using a purple sea urchin fertilization test. Every sample showed positive toxicity results. Unpublished data from these authors' laboratory showed that PAH concentrations must be greater than 100 micrograms per liter ( $\mu\text{g/L}$ ) to show toxicity to the sea urchin sperm, but concentrations in the sampled runoff, from what is assumed to be unsealed asphalt parking lots, were below 30  $\mu\text{g/L}$ . All toxicity seen in those samples was attributed to high concentrations of zinc.

## ***Case Studies***

### **Case Study – U.S. Geological Survey: Austin, TX**

A recent U.S. Geological Survey (USGS) study showed that runoff from sealed parking lots could account for a majority of PAH loadings to urban watersheds (Mahler et al., 2004 and 2005). A variety of surfaces were examined for PAHs in simulated runoff: coal tar-sealed asphalt lots; asphalt emulsion-sealed asphalt lots; unsealed asphalt lots; and unsealed concrete lots. There were four test plots that had no traffic and 13 in-use parking areas. The test plots were sealed prior to the study, while the active lots were studied in their “as is” condition, with the sealant application dates known. Each parking area was sprayed with distilled water to simulate a light rain. The active lots were sampled once each, while the test plots were sampled three times each. The parking areas were also scraped to collect solid particles for direct examination of the lot surfaces (no wetting). PAHs in sealant products were analyzed by the City of Austin by painting the sealant product on glass, allowing it to dry for three days, and measuring the PAH concentration of the dried sealant after it had been scraped from the glass.

The highest PAH concentrations were seen in the products themselves, followed by the scrapings, and then the distilled water wash-off. In the wash-off samples, independent of sealant type, the PAH concentrations in the sediment (particle-phase) were several orders of magnitude larger than those in the water column (dissolved-phase). This research defined the term “probable effect concentration” (PEC). The PEC, the concentration above which adverse effects on benthic biota are expected to occur, was 22,800  $\mu\text{g/kg}$  ( $\Sigma\text{PAH}_{\text{part}}$ ) for the particle phase (MacDonald et al., 2000). The PAH concentrations from the test plot samples all exceeded the PEC. Coal tar sealant runoff exceeded the PEC by a factor of about 150, whereas those from unsealed pavement exceeded the PEC by a factor of about two. Dissolved phase coal tar runoff averaged 9  $\mu\text{g/L}$ . The in-use parking lots generally showed greater PAH concentrations than the test plots with no traffic, generally 20 to 150 times greater, with the coal tar-sealed lots showing higher concentrations than the asphalt sealed and unsealed. The unsealed asphalt lots and unsealed concrete lots showed  $\Sigma\text{PAH}_{\text{part}}$  concentrations on the order of 70,000  $\mu\text{g/kg}$ , which were also above the PEC.

### **Case Study – University of New Hampshire Stormwater Center Study: Durham, NH**

Researchers at the Stormwater Center (SC) at the University of New Hampshire undertook a large-scale study (Watts et al., 2010) similar to the study done by the USGS. The purpose of the study was to examine PAH export from three test parking lots; one sealed with a coal tar-based sealant, one sealed with asphalt-based product, and one unsealed control lot. However, according to the study, it was discovered that both of the sealant lots were coated with coal tar sealant and one lot was left unsealed. All parking lots were traditional asphalt surfaces and the lots were in use for the duration of the study. The coal tar-based sealcoat was applied to the two coated parking areas prior to the beginning of the study and sampling commenced thereafter. Precipitation events generated stormwater runoff for this study. Dependent on rainfall events, samples were taken prior to sealing and then routinely after the sealant application. The study also examined PAH concentrations downstream of the sites to see how far the “reach” of the runoff extended.

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Both water column samples and surface sediment samples were taken, and the ambient air was also sampled for PAHs as part of another study. All three study lots were monitored over a two-year period.

The study calculated the mass total  $\Sigma$ PAHs using the measured flow volume and concentrations of the stormwater runoff from the test lots. The two sealed lots had a mass of 9.8-10.8 kg total  $\Sigma$ 16PAHs per hectare exported in stormwater runoff and 0.34 kg total  $\Sigma$ 16 PAHs per hectare from the unsealed control. The surface sediment sample PAH concentrations were very low prior to the sealcoat application, spiked soon after application, and had decreased from those peak values after twelve months. The coal tar-based sealants showed PAH concentrations that were significantly higher than those of the control (unsealed) parking lot in all sample phases (dissolved, particle, surface sediments).

### **Case Study – U.S. Geological Survey: Madison, WI**

This study examined PAH concentrations in runoff from six urban source areas, including sealed and unsealed asphalt parking lots (Selbig, 2009). The study analyzed runoff from one sealed parking lot and two unsealed, while also investigating streets, roofs, and a mixed-use strip mall. The runoff from the strip mall consisted of combined runoff from the roof, parking lot, sidewalks, and grassy areas. The strip mall parking lot was sealed, but the authors stated that the type of sealant was unknown. The three parking lots examined were each at least five years old and the sealed lot had been coated with a coal tar-based product. The other two lots were maintained only with asphalt-based crack filler. Stormwater runoff from precipitation events was the source water for this study. Sampling events varied for each location, ranging between nine and twenty-seven events from 2005 to 2008. Whole water samples were taken and processed without filtering.

Results showed that runoff from the sealed parking lot had the highest total PAH concentrations; with a mean concentration of 54  $\mu\text{g/L}$ . For most individual PAH compounds, the concentrations observed from the sealed parking lot were significantly greater than those seen in the unsealed lots. The runoff from the roof had the lowest total PAH concentrations, with a mean concentration of 3.4  $\mu\text{g/L}$ ; many of the individual compounds detected were at or near the detection limit (detection limits ranged from 0.04 – 0.5  $\mu\text{g/L}$  depending on the individual PAH under consideration).

### **Case Study – City of Austin: Austin, TX**

Scoggins et al. (2007) studied the occurrence of PAHs in receiving waters downstream from coal tar-sealed parking lots and their effects on stream benthic macroinvertebrate communities. The study matched pairs of upstream (control) and downstream (treatment) sites that were immediately above or below coal tar-sealed parking lots. Organisms were collected in both riffles and pools and nine stream community metrics were used to compare the upstream and downstream sites. Within-habitat variability was not addressed in this study and replicate samples at one site were composited. Sediment PAH concentrations were also measured upstream and downstream of the parking lots.

Total PAH concentrations were markedly higher at the downstream sites compared to the upstream ones in five of the seven sites. Four biological measures indicated that the benthic macroinvertebrate community in pools was degraded at the downstream sites relative to the upstream sites, while three biological measures indicated degradation in riffles. Taxon richness was the most robust metric used in the study. The decrease in taxon richness indicated species loss downstream compared to upstream, which can lead to decreased ecosystem function.

The authors stated that the elevated PAH concentrations could be a primary cause of the degraded streams studied, but other WWF effects, e.g., other pollutants and increased flow from impervious cover, may also be impacting the macro-invertebrate population of the downstream sites. The authors also concluded that current PEC values for PAHs in sediment may not be sufficient to predict toxic effects of bioavailability.



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## Chapter 2 Methods

### Bench-Scale Sealant Study

A bench-scale study was performed prior to the full-scale investigation to provide preliminary results regarding polycyclic aromatic hydrocarbon (PAH) concentrations. Hot-mix asphalt was poured into three open wood frames that were 60 centimeters (cm) x 60 cm wide and 10 cm deep (Figure 1) in March 2007. The runoff area of the asphalt panels was 43 cm x 48 cm due to internal framing to prevent leakage on the edges and to assist in collection of runoff. The asphalt panels were stored in a greenhouse at EPA's research facility in Edison, NJ, in order to protect the panels from precipitation, but to allow exposure to varying temperatures. Figure 2 shows the asphalt panels in the greenhouse prior to application of sealants. All of the asphalt panels were swept, vacuumed, and washed before application of the sealants to two of the asphalt panels.

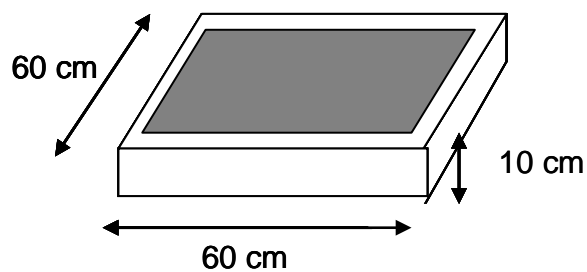


Figure 1. Schematic of asphalt panel and frame.



Figure 2. Three asphalt panels during preparation of study.

Rainwater was collected in a conical 7570 Liter (L) (2000 gallon) tank at the UWRF and transferred to the greenhouse where it was homogenized. Sealants were brushed onto the panels. One asphalt panel was sealed with coal tar sealant, one panel was coated with asphalt emulsion sealant, and one panel surface was left unsealed and treated as a control. Two coats of sealant were applied per manufacturer instructions. During the runoff experiments, the sealed panels were supported at a 14 degree ( $^{\circ}$ ) angle while the control panel was raised to 20 $^{\circ}$  to ensure runoff as some water was observed to infiltrate through the unsealed panel. Each panel was pre-wetted to encourage even sheetflow over the entire width of the panel during the experiment. A 43 millimeter (1.7 inch) rain event was simulated for each asphalt frame, equal to a volume of 10 L (2.4 gallons). This rainwater application mimics the volume of a storm event that falls between a one-year and two-year event for New Jersey (NJDEP, 2004). Collected rainwater was pumped with a peristaltic pump from a carboy to a spray bar apparatus that delivered the water to the panels. The rainwater was applied for twenty minutes, and all runoff was collected from the bottom of the sloped sample surface in a catch trough that drained through a rubber hose to a carboy (Figure 3). The first experiment with applied rainwater was performed after the manufacturer-recommended 24-hour sealant curing time. This sampling process was repeated the next day, and seven and thirty days after sealing.



Figure 3. Sealed asphalt panel with rainwater delivery apparatus, angled to promote runoff.

The asphalt panels were porous, as hand tamping the asphalt in the wooden frames did not produce sufficient compaction; however, this was not a problem for the panels that had sealant applied to them. Runoff losses for the coal tar and asphalt emulsion panels were less than 30 percent (%) and some of this loss was due to a poor seal between the panel and the wooden box, not infiltration. However, the control was losing a majority of rainwater applied, i.e., 60%, despite a higher elevation angle to promote runoff. Some of the rainwater leaked out the sides of the asphalt panels, as well. In response to this, a square panel was cut out of the asphalt from the driveway of the research facility. This cutout was poured the same day as the asphalt panels during pavement installation at the UWRP and was subject to compaction by a steam roller. This cutout asphalt panel did not infiltrate runoff, and was used as an additional unsealed control panel for the later bench-scale tests, i.e., seven and thirty days after sealing.

### **Full-Scale Sealant Study**

Applied runoff of collected rainwater was sampled from three asphalt test plots with different or no surface treatments. One test plot was coated with coal tar sealant, another plot was coated with asphalt emulsion sealant, and the third plot remained unsealed and served as the control. Sampling was also performed in a composite tank that combined the runoff from the three treatments. The sealants used in the full-scale study were the same products as the ones used in the bench-scale study. Comparisons of runoff PAH concentrations are presented as well as other standard water quality parameters. The first sampling event was performed one day after application of sealcoats to pavement, followed by events two, seven, 30, and 162 days after application.

### ***Rainwater Collection***

The full-scale study used the asphalt half of a parking lot at EPA's research facility. The lot, constructed in 1999 as a runoff testing platform for two surfaces, asphalt and concrete, is shown in Figure 4, from an aerial view facing generally north. Each surface slopes to a central drain, with asphalt runoff flowing south and concrete runoff flowing north. Once in the central drain, runoff flows into a collection chamber to the left (note: blue tarp to left of shed).

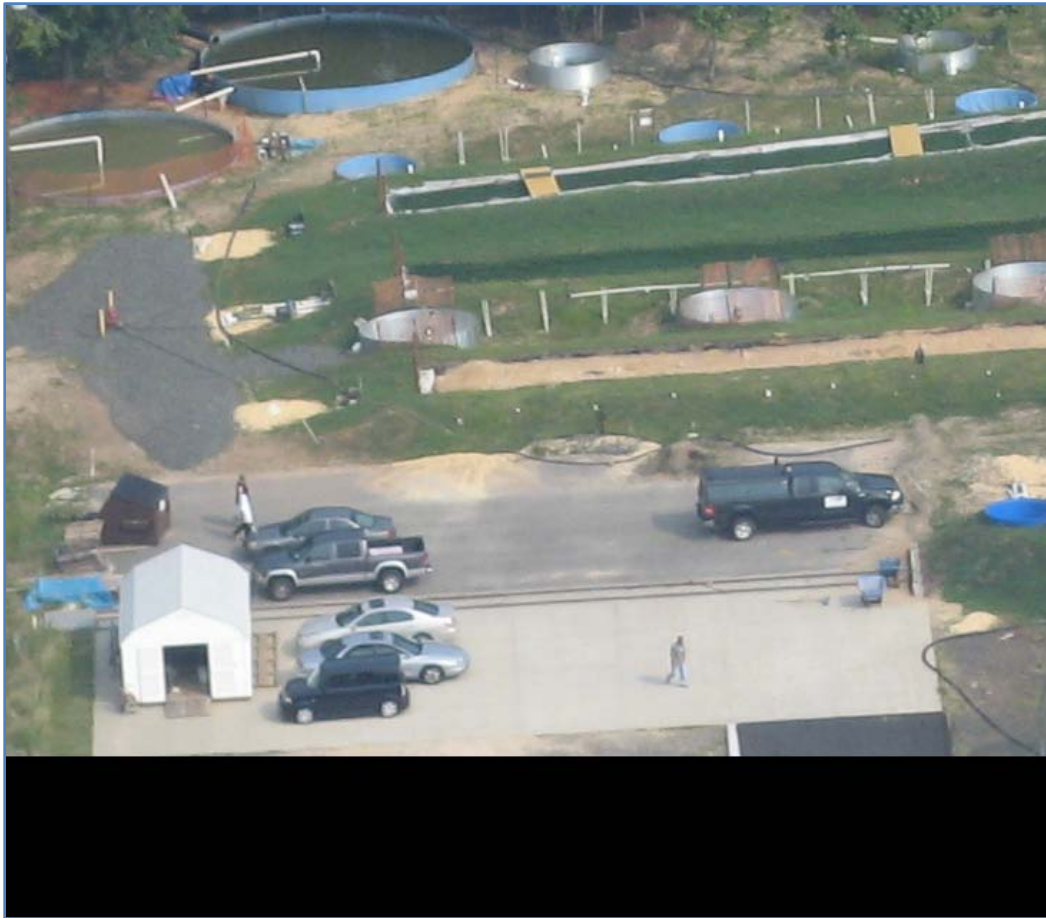


Figure 4. Aerial view of EPA's Urban Watershed Research Facility parking lot.

Rainwater for the full-scale study was captured by placing plastic sheeting on the parking lot and using the central drain to collect the rainwater (Figures 5, 6, and 7). The total area used to collect rainwater from the parking lot was approximately 260 meters squared ( $m^2$ ) (2800  $ft^2$ ). The rainwater was stored and homogenized in lined steel tanks. After sufficient rainwater was collected to apply during experiments, the plastic was taken off the lot and preparations for the experimental study were undertaken. During the study, no cars were allowed to park on the asphalt half of the parking lot; staff parking on the concrete side only.



Figure 5. Parking lot covered in plastic sheeting for rainwater collection.



Figure 6. Parking lot central drains covered in plastic for rainwater collection.



Figure 7. Parking lot drain outlet into sampling box for rainwater collection.

### ***Test Plot Configuration***

A schematic of the experimental design of the parking lot is presented in Figure 8. Each test plot was separated from the next with dividers. The dividers for the test plots were constructed of UV-resistant high-density polyethylene (HDPE) plastic that were laid flat and attached to the asphalt with screws and silicone to provide a water-tight seal. At the bottom of each test plot, the HDPE strips were placed vertically and angled to direct the runoff flow to a ramp where the runoff could either enter the common drain or be collected for sampling (Figure 9). Four test plots were built in the anticipation that an extra test plot might be required for further testing (Figure 10). Each test plot was 3.8 m x 7.6 m, giving a surface area of 29 m<sup>2</sup>. The test plots were swept, vacuumed, and power washed before application of the sealants.

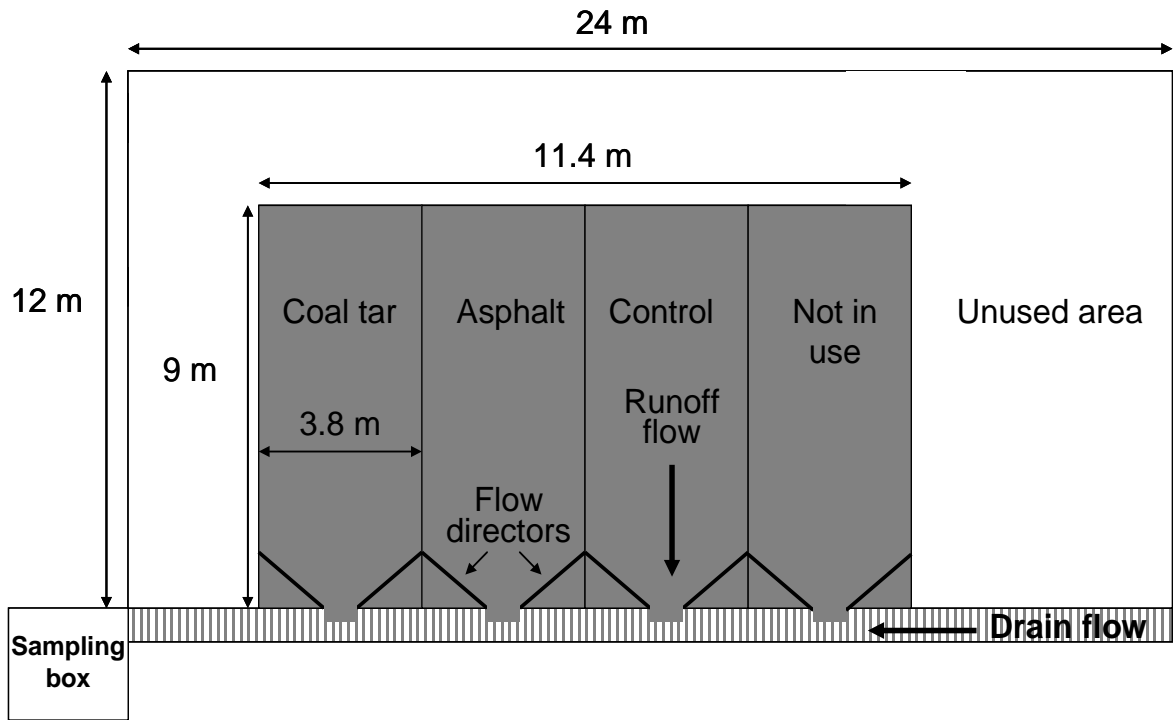


Figure 8. Schematic of the full-scale sealant study.



Figure 9. Asphalt test plot showing the flow directors and the ramp into the drain.



Figure 10. The four test plots at EPA's Urban Watershed Research Facility prior to sealing.

### ***Study Details***

Sealants were applied when the air temperature was 13° Celsius (C) (55° Fahrenheit). Manufacturers specify a minimum air temperature of 10°C (50° Fahrenheit) for application. The sealant was applied with a combination of brushes and squeegees; a new set of tools was used for each sealant. One test plot was sealed with coal tar sealant, another plot was sealed with asphalt emulsion sealant, and another test plot was left unsealed as a control.

One coat of sealant was applied to each of the two test plots that were being tested (coal tar and asphalt emulsion). Manufacturer instructions recommend the application of two coats for best results, but there was not sufficient sealant of either type to fulfill this requirement. One 5-gallon bucket of each sealant type should have been more than sufficient to cover the surface area of one test plot, assuming the asphalt was previously sealed. However, the UWRF parking lot was never sealed and would have required additional sealant to achieve two coats.

It is suspected that the age and wear condition of the UWRF parking lot asphalt led to the reduced coverage of the sealants, with the asphalt being rough and becoming slightly porous with time. This led to the sealant soaking into the asphalt rather than just coating the surface. For both types of sealants, the manufacturer's instructions on the containers recommended a minimum 24-hr cure after application before returning to general usage of surface.

After the 24-hr cure time, each test plot was pre-wetted prior to the simulated rain event to encourage an even sheet flow. The pre-wetting did not result in runoff, but some of this water was captured with the initial runoff that was sampled. The previously collected rainwater was applied using a hose and an attached spray nozzle. An in-line paddle wheel flowmeter measured the volume applied.

Approximately 436 L (115 gallons) was delivered over one hour for each asphalt test plot. This volume was equivalent to 15 mm (0.6 inches) rain event, which is approximately half the New Jersey water quality design storm (NJDEP, 2004) and ensured equal application to each test plot. Samples were collected at the discharge point from each test plot (white ramp as pictured in Figure 9) after running off the asphalt but before reaching the common drain.



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Samples of the mixed flow were collected at a catch basin where all the runoff was collected. The runoff was then pumped into the composite tank and combined to simulate mixing of runoff from a variety of surfaces. This was done in order to compare runoff from sealed asphalt surfaces to runoff that has been mixed and transported downstream.

Samples were collected immediately after runoff appeared at the collection point (time zero), at 30 minutes, and at 60 minutes. For the first sampling event, one day after sealing, the time-sliced samples were collected separately; but for all other events (two, seven, 30 and 162 days after sealant application), the time-sliced samples were composited. The first sampling event was broken down into several time slices with the intent to discern if the runoff from the first flush had measurable differences.

There was no rain during the first week after the sealant application. Three small (less than 2.5 mm) rain events and one large rain event (13 mm) occurred after the seven-day event and before the 30-day event. There were numerous precipitation events between 30 and 162 days, but these were not quantified. No samples were taken during any of these precipitation events.

## **Analytical Procedures**

### ***PAH Analyses***

The runoff samples were analyzed for PAH concentrations by EPA Method 8270C “Semi-volatile organic compounds by gas chromatograph-mass spectrometer (GC-MS)” (EPA 1996). Lists of the 17 PAHs examined during this study are presented in Table 1 and include the 16 PAHs that are classified as EPA Priority Pollutants, as well as 2-methylnaphthalene (EPA 2009). Most of the PAHs examined here are considered semi-volatile, while naphthalene and 2-methylnaphthalene are considered volatile. The table shows the individual PAH analytes, their detection limits, and the lowest standard used in the generation of a calibration curve. The method detection limit for  $\sum$ PAH for these 17 compounds was calculated as the sum of the individual detection limits, which was 0.29 microgram ( $\mu\text{g}$ )/L.

### ***Water Quality Parameters***

In addition to PAH, the samples were also tested for a variety of water quality parameters including: total suspended solids (TSS), volatile suspended solids (VSS), total organic carbon (TOC), chemical oxygen demand (COD), pH, and Microtox® toxicity. Both TOC and COD assess the amount organic matter in a water samples. The COD test is used as a surrogate for biochemical oxygen demand (BOD), as COD is a shorter, easier test to run. COD represents the amount of oxidizable material in the water, which in turn represents the potential for reducing dissolved oxygen (DO) in the receiving water. The full name of the analytical procedures and method references for the testing for these parameters can be found in Table 2. The Microtox® analyses were performed but the results were inconclusive and are not reported.

Table 1. Polycyclic aromatic hydrocarbons, detection limits and lowest standards.

Polycyclic Aromatic Hydrocarbon	Detection Limit (µg/L)	Lowest Standard (µg/L)
Naphthalene	0.02	1.00
2-Methylnaphthalene*	0.01	1.00
Acenaphthylene	0.01	1.00
Acenaphthene	0.02	1.00
Fluorene	0.01	1.00
Phenanthrene	0.03	1.00
Anthracene	0.02	1.00
Fluoranthene	0.02	1.00
Pyrene	0.02	1.00
Benzo[a]anthracene	0.02	1.00
Chrysene	0.02	1.00
Benzo[b]fluoranthene	0.02	1.00
Benzo[k]fluoranthene	0.01	1.00
Benzo[a]pyrene	0.02	1.00
Indeno[1,2,3-cd]pyrene	0.01	1.00
Dibenz[a,h]anthracene	0.01	1.00
Benzo[g,h,i]perylene	0.02	1.00

\* 2-Methylnaphthalene is not an EPA priority pollutant.

Table 2. Analytical procedures.

Parameter	Method	SOP	Discrete Bottle Identifier
Total Organic Carbon (TOC)	SM 5310-TOC		None (composite only)
Microtox® toxicity <sup>1</sup>	SM 8010-F		Composite unless otherwise noted
Total Suspended Solids (TSS)	SM 2540-D	30	None (composite only)
Volatile Suspended Solids (VSS)	SM 2540-E	30 <sup>2</sup>	None (composite only)
Chemical Oxygen Demand (COD)	SM 5220-COD-D	55	None (composite only)
Polycyclic Aromatic Hydrocarbons (PAHs)	SM 6440-C/EPA Method 8270C		Composite unless otherwise noted

<sup>1</sup> Method number refers to general toxicity guidance only

<sup>2</sup> Residue from TSS analysis will be treated according to SM 2540-E

SOP = Standard Operating Procedure

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## Chapter 3 Results

### Bench-Scale Study Results

#### *Polycyclic Aromatic Hydrocarbon (PAH) Results*

A list of the 17 polycyclic aromatic hydrocarbons (PAHs) examined during the bench-scale study is presented in Table 3. This table shows the range of PAH concentrations found in runoff samples from each panel for the bench-scale study, where “n” is the total number of samples taken. Many individual PAHs were found to be above the detection limit, but below the lowest standard so these reported concentrations may not be as reliable as those that fall within the points on the calibration curve. Non-detect results were counted as zeroes in the sum PAH ( $\Sigma$ PAH) values.

PAH results of the collected runoff are shown in Figure 11. Results are shown as the  $\Sigma$ PAH of 17 semi-volatile PAHs. Note that the concentrations are shown on a log scale. The whole water sample was analyzed rather than analyzing the particle and dissolved phases separately. Three samples were taken for each sealed asphalt panel for each rainwater application event. The unsealed panel had two samples taken at each event, with the exception of the first event, which had three samples taken; this is why no error bars are shown in Figure 11. The results below are daily averages of the samples and the error bars are the 95 percent (%) confidence intervals for the coal tar and asphalt emulsion runoff. The coal tar runoff samples contained 16 of the 17 analyzed PAHs in the one-day sample, while five of the 17 PAH analytes were detected in the one-day asphalt emulsion runoff sample, and eight of the 17 PAHs were found in the one-day control sample.

The runoff from the coal tar-sealed asphalt panel had the highest PAH concentrations at every sampling event for the bench-scale study. The PAH concentrations in the runoff samples from the coal tar panel were more than two orders of magnitude higher than those of the asphalt emulsion and unsealed panel. PAH concentrations observed in the runoff generally decreased with time. The asphalt emulsion and the unsealed panels both showed low  $\Sigma$ PAH concentrations and were not significantly different from each other (least square mean test,  $p = 0.1973$ ). The runoff from the coal tar panel had significantly higher means than the unsealed or asphalt emulsion panels (least square mean test,  $p < 0.0001$  for both comparisons).

The individual PAH compounds that contributed to the total PAH concentrations for the runoff samples were variable among the three test plots. The PAHs in the asphalt emulsion sealant runoff samples consisted of only five of the 17 PAH compounds examined, and all of those are considered low-molecular weight and more soluble and prone to volatilization. The coal tar runoff samples contained 16 of the 17 compounds analyzed, but the constituency was dominated by phenanthrene (44%). The unsealed control runoff contained eight of the 17 analytes, with phenanthrene contributing 30% to the total PAH concentration. Figure 12 shows a breakdown of individual contribution of PAHs to the  $\Sigma$ PAH in the runoff on a percent basis for the sample numbers (n) taken 24 hours after sealant application ( $n = 2$  for unsealed,  $n = 3$  for asphalt emulsion and coal tar). The runoff in the bench-scale study showed the presence of

PAHs with samples from the coal tar-sealed panel yielding the highest values; maximum ΣPAHs = 164 micrograms /liter (µg/L).

Table 3. Polycyclic aromatic hydrocarbons (PAHs) observed ranges for the bench-scale study runoff.

Polycyclic Aromatic Hydrocarbon	Observed Range (µg/L)		
	Unsealed n = 8	Asphalt emulsion sealant n = 12	Coal tar sealant n = 12
Naphthalene	ND – 0.04	0.02 – 0.04	0.17 – 8.45
2-Methylnaphthalene*	ND – 0.02	ND – 0.02	0.03 – 1.26
Acenaphthylene	ND	ND	ND
Acenaphthene	ND – 0.06	ND – 0.02	0.32 – 22
Fluorene	0.02 – 0.06	ND – 0.03	0.54 – 27.3
Phenanthrene	0.03 – 0.13	0.03 – 0.09	3.29 – 72.4
Anthracene	ND – 0.04	ND	0.35 – 12
Fluoranthene	ND – 0.04	ND – 0.02	0.85 – 13.4
Pyrene	ND – 0.03	ND	0.58 – 6.67
Benzo[a]anthracene	ND – 0.04	ND – 0.03	0.03 – 0.98
Chrysene	ND – 0.05	ND – 0.03	0.07 – 0.57
Benzo[b]fluoranthene	ND – 0.05	ND – 0.03	0.02 – 0.19
Benzo[k]fluoranthene	ND – 0.05	ND – 0.03	0.02 – 0.19
Benzo[a]pyrene	ND – 0.04	ND – 0.02	ND – 0.18
Indeno[1,2,3-cd]pyrene	ND – 0.04	ND – 0.03	ND – 0.13
Dibenz[a,h]anthracene	ND – 0.05	ND – 0.03	ND – 0.14
Benzo[g,h,i]perylene	ND – 0.04	ND – 0.03	ND – 0.12

\* 2-Methylnaphthalene is not an EPA priority pollutant

ND = Non-detect

n = Sample size

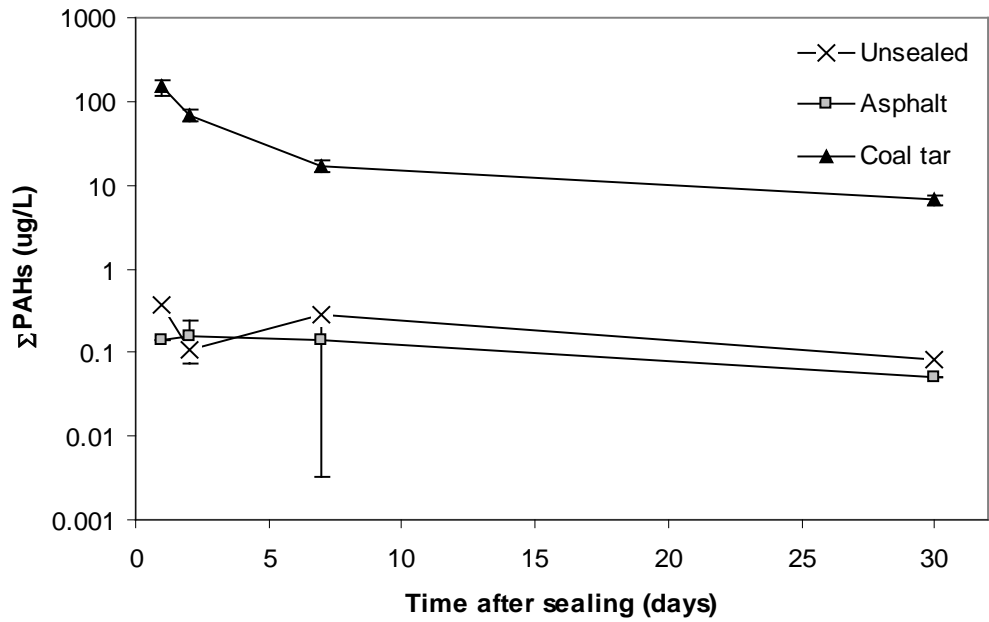


Figure 11. Summation of 17 polycyclic aromatic hydrocarbons ( $\Sigma$ PAH) concentrations for four sampling events of bench-scale study.

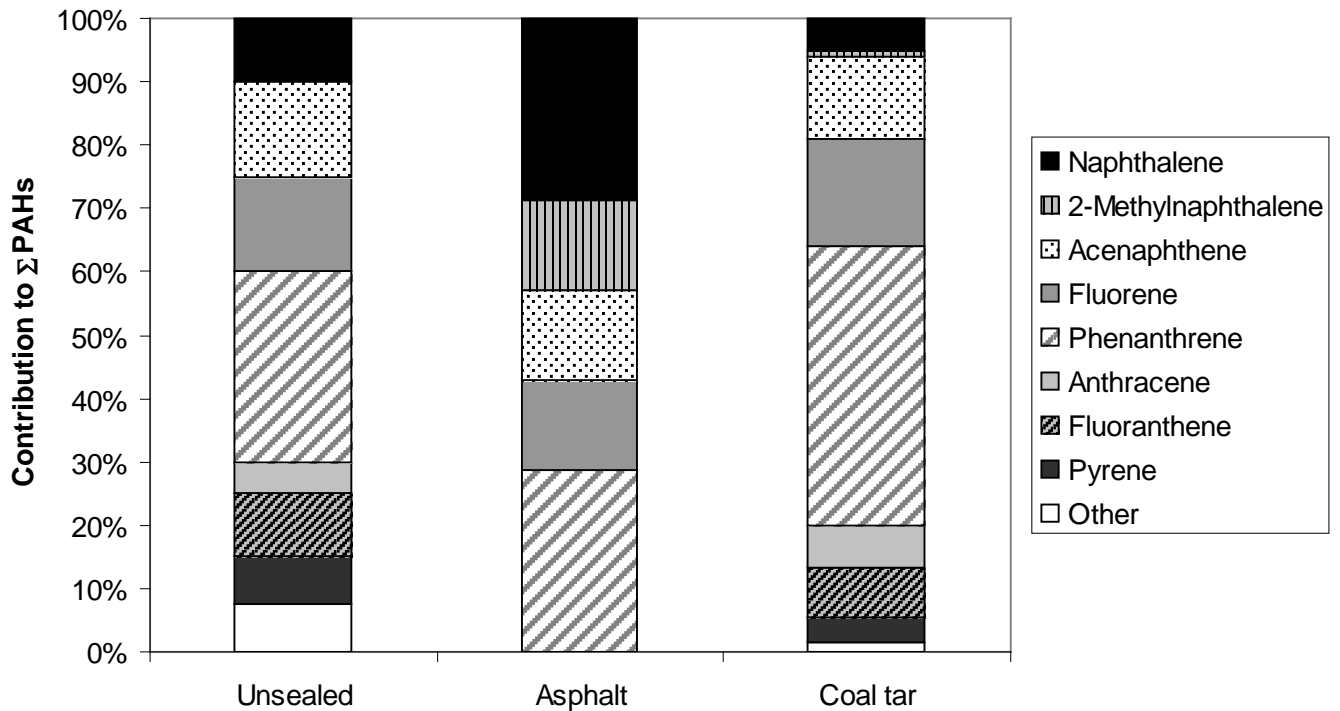


Figure 12. Percent contribution of individual to sum of polycyclic aromatic hydrocarbons (PAHs) of bench-scale study.

### Water Quality Results

The water quality parameters of the chemical oxygen demand (COD) and total organic carbon (TOC) were especially interesting for the one-day sampling event (Figure 13). The COD values for all three panels were elevated above concentrations typically measured in stormwater runoff samples. Lager et al. (1977) found a mean COD concentration of 115 mg/L with a range of values of 48-170 mg/L. All exceeded the 90% observed range for urban sites of 140 mg/L, (EPA, 1983). The asphalt emulsion runoff samples exceeded 500 mg/L, which for comparative purposes, is more typical of a COD concentration of medium-strength untreated domestic wastewater (Tchobanoglous and Burton, 1991) than runoff. The asphalt emulsion COD runoff concentrations dropped dramatically the first day after sealant application.

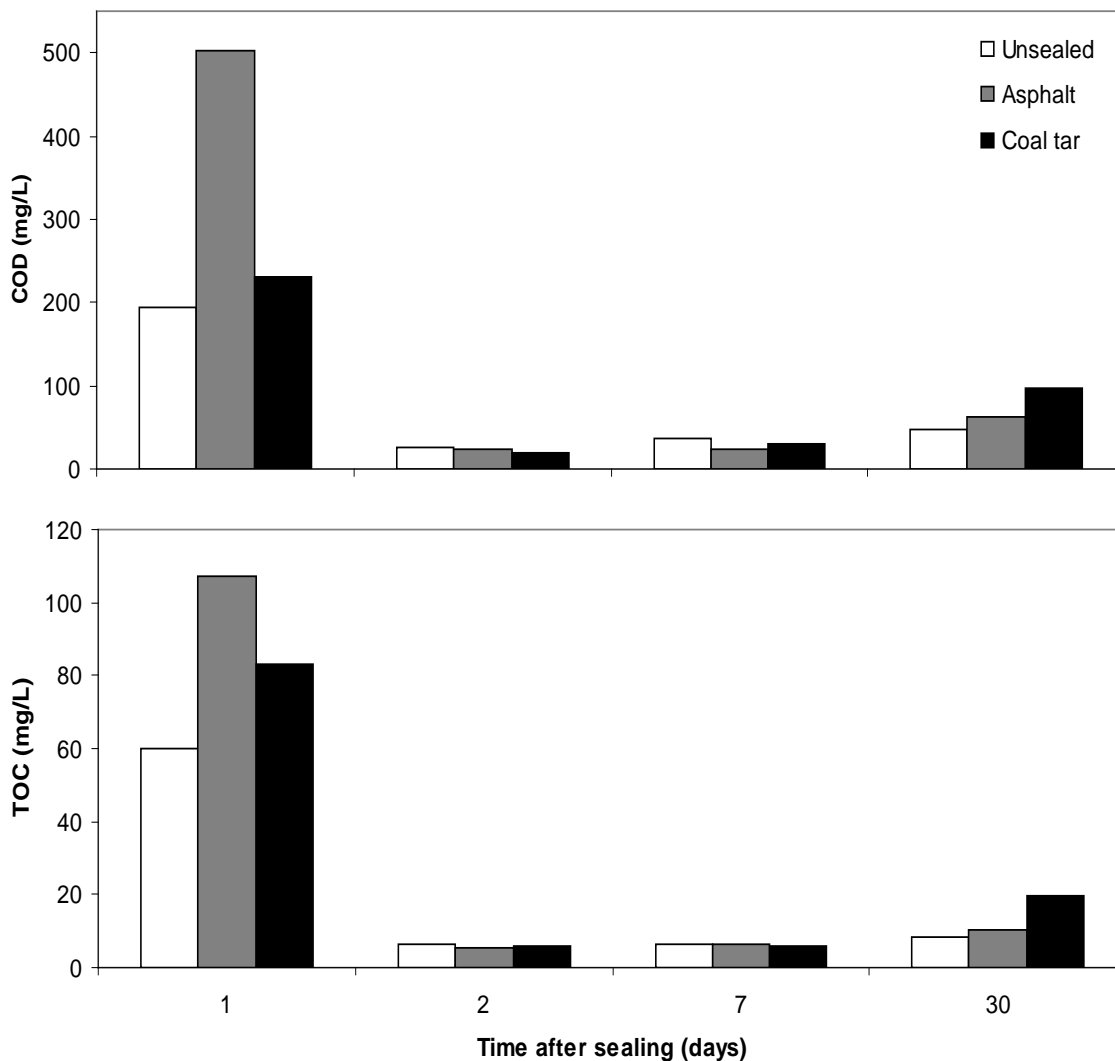


Figure 13. Total organic carbon (TOC) and chemical oxygen demand (COD) concentrations of bench-scale study.

The TOC concentrations follow a similar pattern to the COD values, producing runoff of both the coal tar and asphalt emulsion sealant in a range of untreated wastewater from weak, 80 mg/L to medium, 160 mg/L (Tchobanoglous and Burton, 1991). The direct impact to receiving water of increased COD and TOC loading is difficult to predict as both

analyses convert organic matter regardless of “the biological assimilability of the substances” (Sawyer and McCarty, 1978); specifically, PAHs and the other organic components of the sealants may not exert a large oxygen demand due to potential toxicity of PAHs and limited biodegradability of other organic components. Total suspended solids (TSS) concentrations ranged from 1.4 – 9.5 mg/L, which is surprisingly low but can be attributed to the preparation of the surfaces, i.e., cleaning before sealing as per manufacturer instructions.

## Full-Sale Study Results

### *Polycyclic Aromatic Hydrocarbon (PAH) Results*

A list of the 17 PAHs examined during the full-scale study is presented in Table 4. This table shows the range of PAH concentrations found in runoff samples from each plot in the full-scale study, where “n” is the number of samples taken. Many individual PAHs were found to be above the detection limit, but below the lowest standard; therefore, these reported concentrations may not be as reliable as those that fall within the points on the calibration curve. Non-detect results were counted as zeroes in the  $\sum$ PAH values.

Table 4. Polycyclic aromatic hydrocarbons (PAHs) observed ranges for the full-scale study runoff.

Polycyclic Aromatic Hydrocarbon	Observed Range (µg/L)		
	Unsealed n = 11	Asphalt emulsion sealant n = 15	Coal tar sealant n = 15
Naphthalene	ND – 0.16	ND – 0.44	0.86 – 19.4
2-Methylnaphthalene*	ND – 0.04	ND – 0.1	0.14 – 2.48
Acenaphthylene	ND – 0.02	ND – 0.03	0.02 – 0.15
Acenaphthene	ND – 0.2	ND – 0.84	1.3 – 25.4
Fluorene	0.02 – 0.5	0.03 – 1.14	3.47 – 40.4
Phenanthrene	0.16 – 1.13	0.39 – 1.64	33.1 – 125
Anthracene	ND – 0.08	0.02 – 0.19	3.04 – 16.1
Fluoranthene	0.04 – 1.04	0.07 – 0.47	12 – 28.8
Pyrene	0.02 – 0.81	0.05 – 0.37	4.43 – 15.8
Benzo[a]anthracene	ND – 0.45	ND – 0.14	0.28 – 3.28
Chrysene	ND – 0.71	0.02 – 0.23	0.4 – 2.59
Benzo[b]fluoranthene	ND – 0.58	ND – 0.22	0.11 – 1.59
Benzo[k]fluoranthene	ND – 0.56	ND – 0.18	0.1 – 1.65
Benzo[a]pyrene	ND – 0.51	ND – 0.18	0.08 – 2.08
Indeno[1,2,3-cd]pyrene	ND – 0.4	ND – 0.1	0.04 – 1.18
Dibenz[a,h]anthracene	ND – 0.15	ND – 0.04	0.02 – 0.59
Benzo[g,h,i]perylene	ND – 0.44	ND – 0.11	0.03 – 1.17

\* 2-Methylnaphthalene is not an EPA priority pollutant  
 ND = Non-detect  
 n = Sample size

Results of the runoff are shown in Figure 14 as the  $\Sigma$ PAH sum of 17 semi-volatile PAHs. Note that the concentrations are shown on a log scale. The whole water sample was analyzed rather than analyzing the particle and dissolved phases separately. The method detection limit for  $\Sigma$ PAH for these 17 compounds is 0.29  $\mu\text{g/L}$  (the sum of the individual detection limits). Individual non-detect results were counted as zeroes in the  $\Sigma$ PAH values. The analysis of the coal tar runoff samples detected all 17 PAHs in every sample. Three runoff samples were taken for each sealed test plot (coal tar and asphalt emulsion) and the composite tank for each rainwater application event; the unsealed plot had two runoff samples taken at each event. The results below are averages of the collected samples and the error bars are the 95% confidence intervals. The composite tank was not sampled at 162 days because the general mixing trends were known by that time.

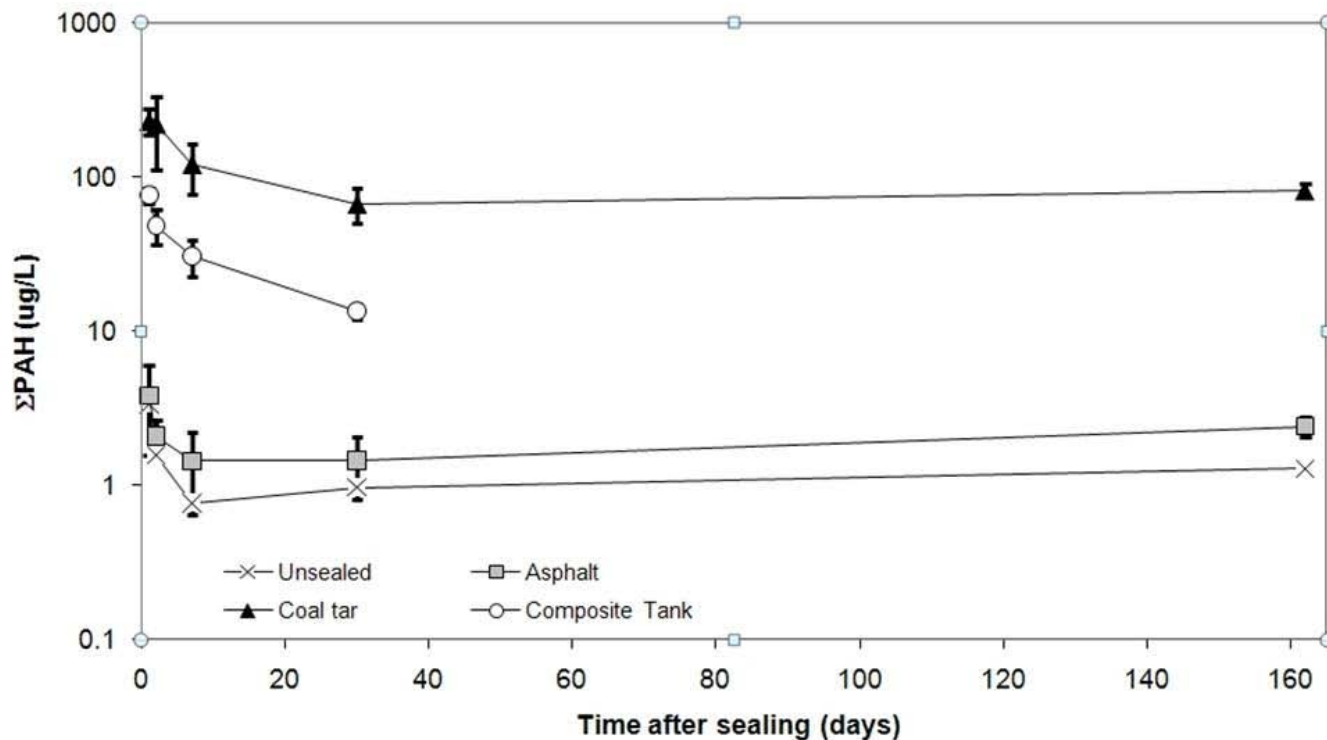


Figure 14. The sum of the polycyclic aromatic hydrocarbon concentrations ( $\Sigma$ PAH) in runoff for the five sampling events of full-scale study ( $\Sigma$ PAH = 17).

The runoff from the coal tar-sealed asphalt plot had the highest PAH concentrations at every sampling event. The PAH concentrations of the runoff from the coal tar plot were more than an order of magnitude higher than runoff concentration from the asphalt emulsion section. Runoff from the asphalt emulsion and the unsealed plots both showed low  $\Sigma$ PAH concentrations, and concentrations were significantly different from each other (Least square mean test,  $p = 0.0006$ ). The asphalt emulsion and coal tar runoff samples were significantly different from each other with regard to PAH concentrations (Least square mean test,  $p < 0.0001$ ). The coal tar runoff samples were also significantly different from those from the unsealed test plot (Least square mean test,  $p < 0.0001$ ). The results from the composite tank were generally about a third of the coal tar runoff values, as expected because the tank samples were a combination of the unsealed (low PAH concentrations), the asphalt emulsion (low PAH concentrations), and the coal tar runoff. PAH concentrations in the runoff generally decreased rapidly over the first three sampling events.



The individual PAH compounds that contributed to the  $\Sigma$ PAHs for each runoff sample were variable for the one-day sampling event (Figure 15). More PAH compounds were found in the asphalt emulsion runoff samples here in the full-scale study than in the bench-scale; however, the presence of 2-methylnaphthalene was conspicuously diminished. In the bench-scale, 2-methylnaphthalene contributed 14% to the total PAHs for the asphalt emulsion runoff samples, while in the full-scale study it contributed less than 2%. The coal tar runoff samples contained each of the 17 compounds analyzed, but the constituency was dominated by phenanthrene (47%). The unsealed control contained all 17 analytes, with only phenanthrene contributing more than 16% to the total PAH concentration; the concentrations were very low compared to those in the runoff from the coal tar-sealed test plot. The composite tank samples were dominated by the coal tar runoff with regard to PAH constituents. Figure 16 compares the individual PAH concentrations in the runoff for the three surfaces. PAHs are presented in order of elution (low molecular weight on the left, to increasing molecular weight on the right).

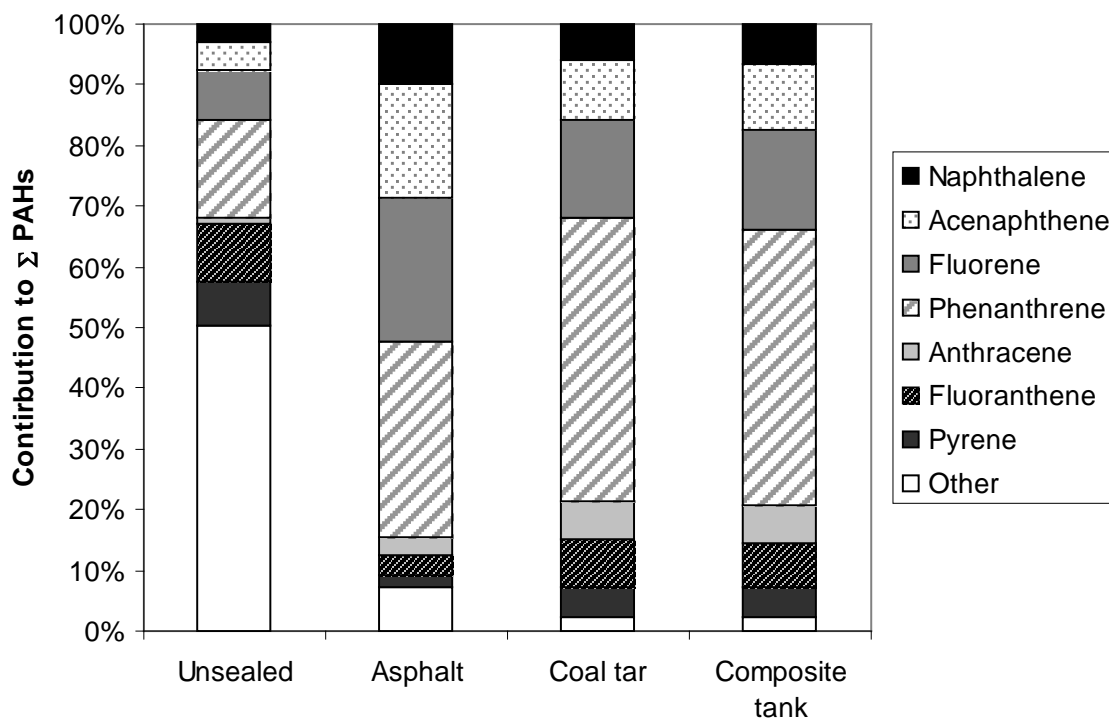


Figure 15. Percent contribution to the sum of polycyclic aromatic hydrocarbons ( $\Sigma$ PAH) for individual polycyclic aromatic hydrocarbons in runoff for the one-day sampling event for the full-scale study.

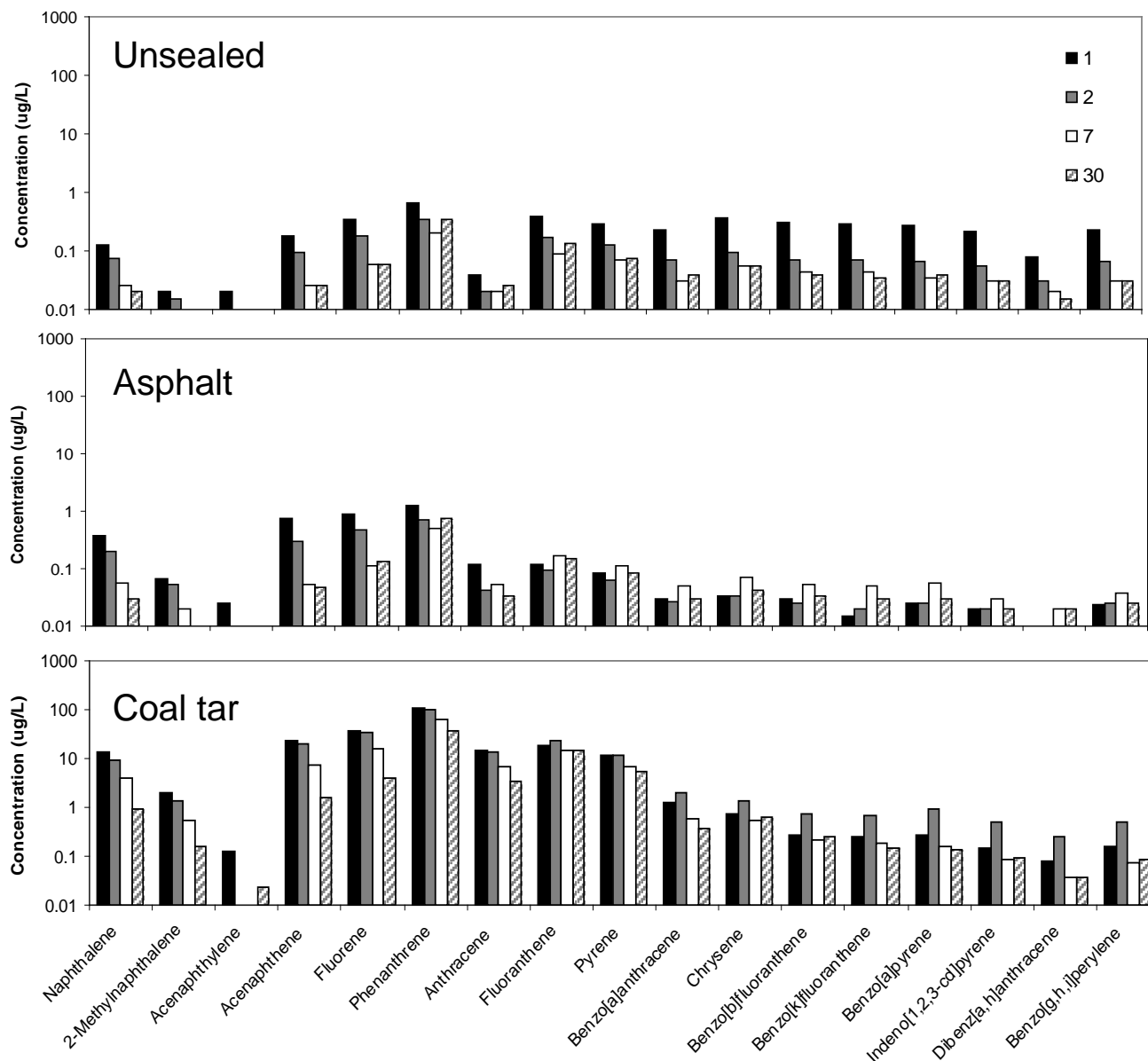


Figure 16. Polycyclic aromatic hydrocarbon concentrations in runoff for each compound for the 1-, 2-, 7-, and thirty-day sampling events for each surface of full-scale study.

The individual PAH makeup of the runoff samples changed with time as shown in Figure 17, which shows the compounds in order from lower to higher molecular weights. The runoff from the sealed surfaces lost the lower molecular weight PAHs as the sealant aged. For runoff samples from the asphalt emulsion-sealed plot and the coal tar-sealed plot, the contributions of naphthalene, 2-methylnaphthalene, and acenaphthene decreased from one to 162 days after sealing. In the runoff samples from the asphalt emulsion-sealed plot, the higher molecular weight compounds comprised a greater percentage of the total PAHs in the last sample, which was five and one-half months after sealing. Watts et al. (2010b) observed a similar pattern and attributed it to weathering.

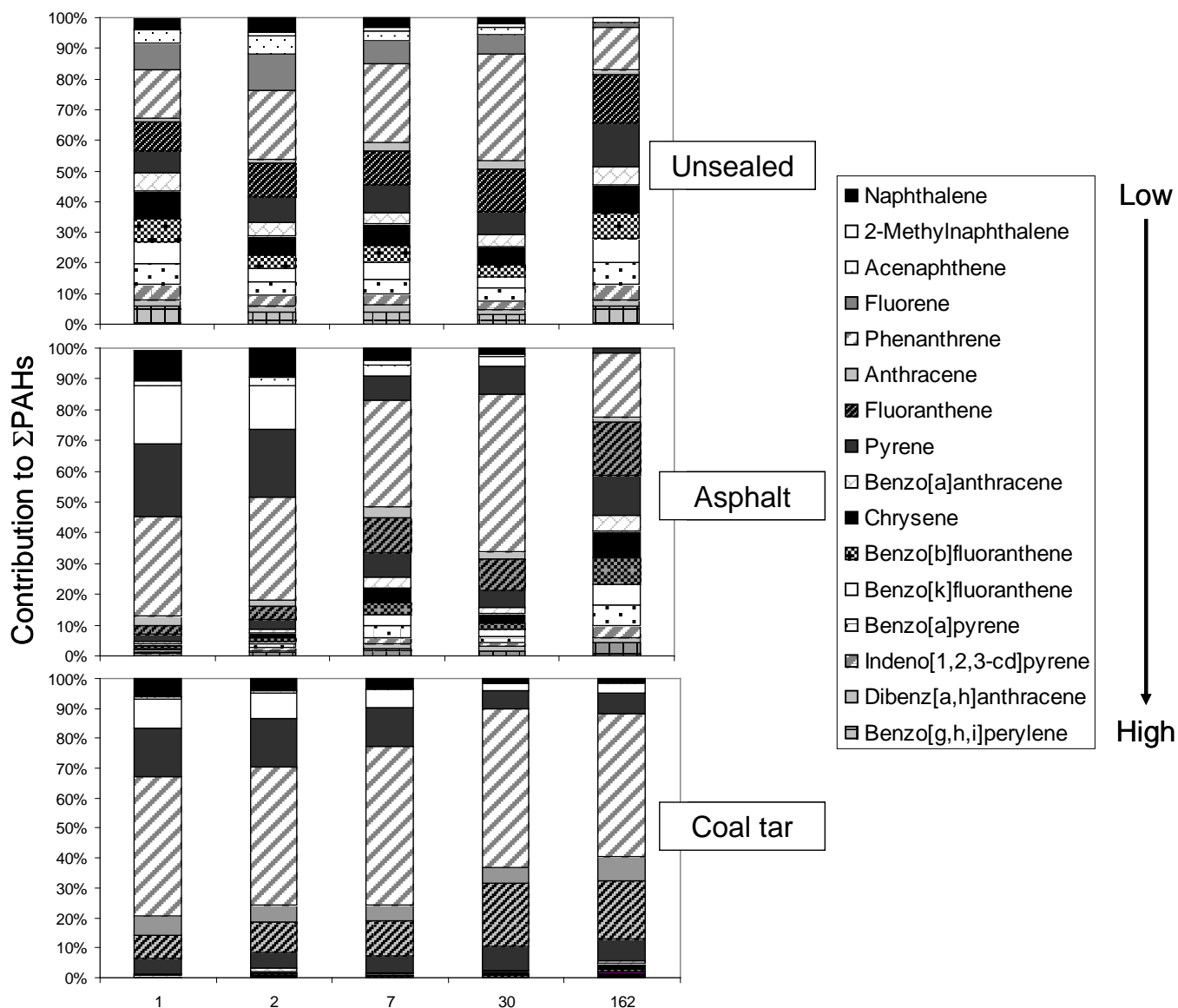


Figure 17. Percent contribution of individual polycyclic aromatic hydrocarbons (PAHs) in runoff to the total PAHs observed ( $\Sigma$ PAH) over all sampling events for the full-scale study.

### Water Quality Results

The water quality parameters of COD and TOC presented in Figure 18 (note: scales of y-axes are different) showed values that were more typical of urban runoff than the very high concentrations seen in the bench-scale (Figure 13). As noted previously, the composite tank was not sampled at the 162-day sampling event. The TOC results are characteristic of urban stormwater runoff (Winer, 2000). COD and TOC concentrations in the runoff were relatively consistent over the first 30 days, with the exception of the asphalt emulsion, which peaked on the initial sampling time. Not including the initial sample of the asphalt emulsion sealant runoff, both COD and TOC concentrations peaked at the 162-day sampling event, which may be indicative of the degradation of recalcitrant organic compounds over a longer time period; general accumulation of organic matter via atmospheric deposition; or wind-swept local transport. This is supported by the TSS results, which also peaked at the 162-day sampling event.

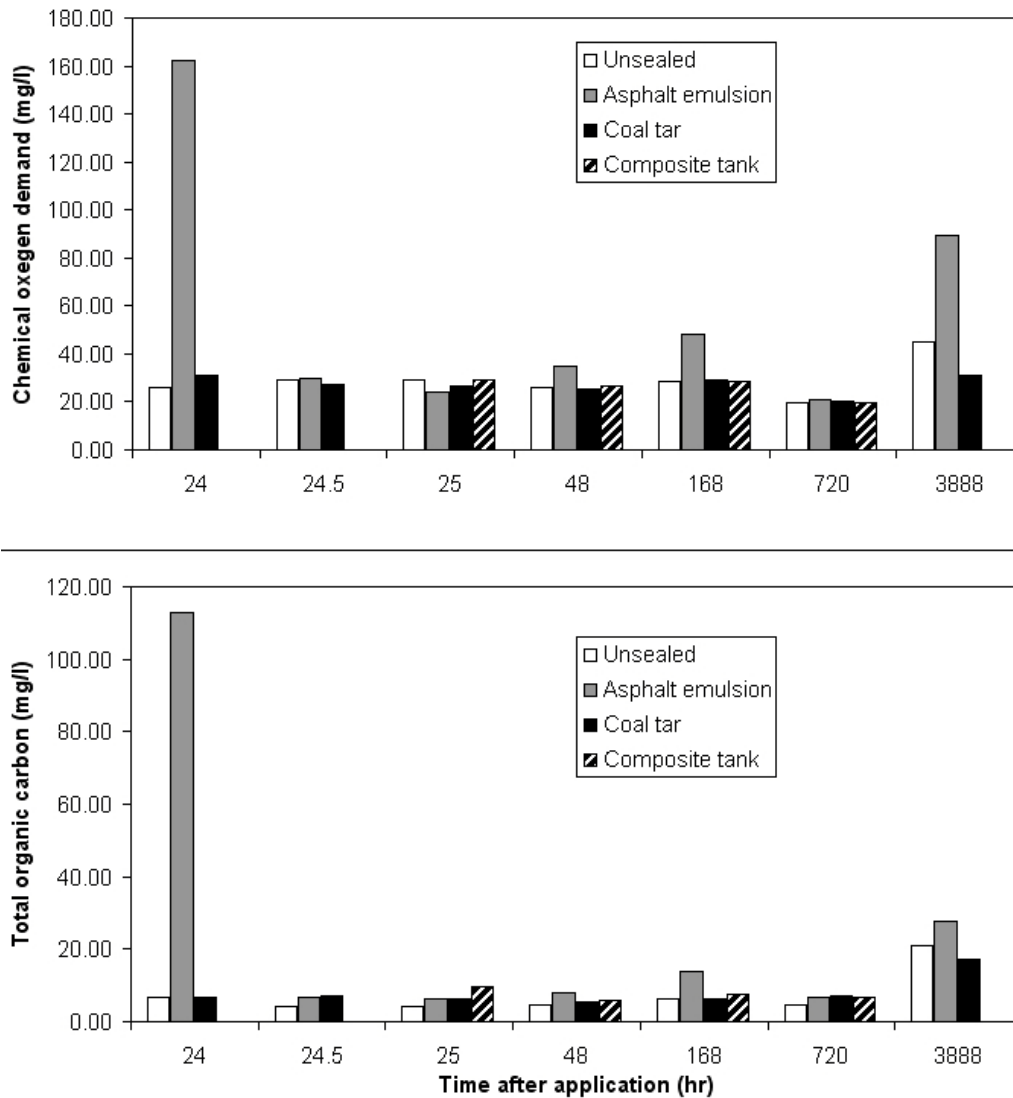


Figure 18. Total organic carbon and chemical oxygen demand concentrations in runoff for full-scale study.

TSS concentrations were generally very low for the first sampling events (1-, 2-, 7-, and 30-day samples) and concentrations ranged from 2.8 – 14.8 mg/L. Peak TSS concentrations in the runoff were seen at the 162-day sampling event for all plot types, with concentrations of 25.7 mg/L for the unsealed, 82.2 mg/L for the asphalt emulsion, and 22.5 mg/L for the coal tar sealant. The high TSS concentrations seen at the long-term sampling event most likely represent the accumulation of atmospheric deposition of particles and other material, the degradation of the sealant, or the degradation of the asphalt surface over 4-5 months.

While TSS values were low, the observed ratio of volatile suspended solids (VSS) to TSS in sampled runoff was high. This is an indication of an organic component loading in the runoff. The composite tank averaged 0.96 on the first day and ranged from 0.69 to 1.0 for the study. Similarly, coal tar runoff was 0.81 the first day and ranged from 0.54 to 0.94 for the study; asphalt emulsion runoff was initially 0.84, ranging from 0.43 to 0.98; and the unsealed runoff was initially 0.89 and ranged from 0.44 to 0.96.

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## Chapter 4 Discussion

### Comparison to Literature Values

#### *Polycyclic Aromatic Hydrocarbon (PAH) Concentrations*

The maximum PAH concentrations in this study are comparable to those reported in other studies for sealed asphalt sites. Figure 19 shows concentrations for this and two other studies. The University of New Hampshire (UNH) data are from Watts (2009, 2010a) and the U.S. Geological Survey-Wisconsin (USGS-WI) data are from Selbig (2009).

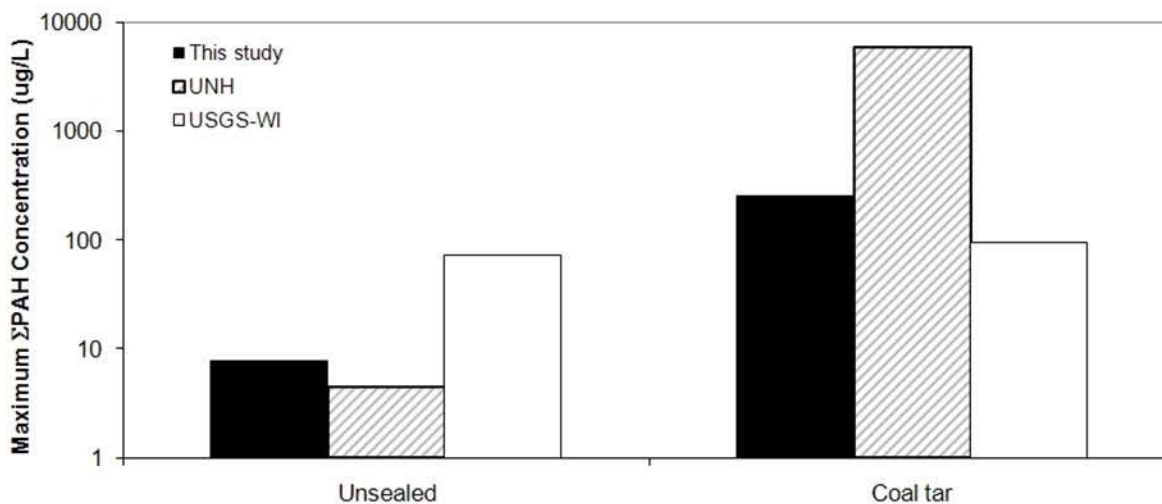


Figure 19. Maximum sum of polycyclic aromatic hydrocarbon ( $\Sigma$ PAH) concentrations in runoff for full-scale study and two other studies.

The USGS study examined runoff from one coal tar-sealed parking lot and two unsealed parking lots. The sum of PAHs for the full-scale research associated with this study consisted of 17 compounds, 16 for the UNH study, and 18 for the USGS-WI study. It should be noted that during the UNH and USGS-WI studies, the parking lots were active and parking was allowed, while parking was prohibited for the duration of this study. The EPA study results represent the PAH runoff that could be expected from runoff and weathering without contributions from vehicular use.

A survey of in-use parking lot runoff in Alabama showed that the mean total PAH concentration, based on seven reported PAHs, was 286 µg/L (Pitt et al., 1995). In comparison, the sum of the day one sampling event in this study for these same seven PAHs was much lower: 4.11 µg/L for the unsealed and 48 µg/L for the coal tar-sealed runoff.

The above discussion only relates the observed concentrations from this and other studies. Because of the variations in urban runoff, due to variability in usage conditions and climate (e.g., antecedent moisture conditions and rainfall intensity), the NURP study (EPA, 1983) recommended comparing loadings (not concentrations).

In the runoff samples from the asphalt emulsion-sealed plot, the higher molecular weight compounds comprised a greater percentage of the total PAHs in the last sample. This is most likely due to the low molecular weight PAHs volatilizing off earlier, leaving the heavier PAHs to contribute a higher percentage to the total PAH concentration. Similar observations were made by Mahler et al. (2005), who reported a general (but not uniform) decrease in the concentration of PAHs in both the particulate and dissolved phase in runoff from test plots (no vehicle traffic) and a decrease in the ratio of the low molecular weight to high molecular weight PAHs, which was attributed to volatilization and leaching.

The varied PAH makeup of the unsealed control samples is potentially from some combination of atmospheric deposition; the asphalt surface itself; and/or possibly wind-driven cross-contamination from the nearby coal tar emulsion- and asphalt emulsion-sealed test plots. The concentrations seen in the unsealed runoff and the asphalt emulsion runoff samples are only slightly higher than detection limits, while those of the coal tar runoff samples are two orders of magnitude higher.

### ***Water Quality Parameters and Polycyclic Aromatic Hydrocarbon (PAH) Concentrations***

The chemical oxygen demand (COD) concentrations seen in EPA’s full-scale study here are generally high compared to the total suspended solids (TSS) concentrations, which is unusual for urban runoff. Table 5 compares COD concentrations, TSS values, and the COD/TSS ratio of several urban sites and EPA’s full-scale study. The observations in this study are unusual in comparison to other literature values as COD values are exceeding TSS values. The initial runoff from the asphalt emulsion sealant had the highest observed COD values.

Table 5. Mean total suspended solids, chemical oxygen demand, and ratios for runoff from literature and this study.

<b>Location</b>	<b>Total Suspended Solids (mg/L)</b>	<b>Chemical Oxygen Demand (mg/L)</b>	<b>Chemical Oxygen Demand Total Suspended Solids Ratio</b>
<b>Urban Stormwater<sup>1</sup> (mean)</b>	415	113	0.27
<b>NURP<sup>2</sup> (median)</b>			
Residential	101	73	0.72
Mixed	67	65	0.97
Commercial	69	57	0.83
Open/Nonurban	70	40	0.57
<b>NSQD<sup>3</sup> (median)</b>	58	53	0.91
<b>EPA Full-scale runoff</b>			
Unsealed (mean)	12	29	2.3
Asphalt emulsion (mean)	25	55	2.2
Coal tar (mean)	11	27	2.4

<sup>1</sup> Lager et al. (1997)

<sup>2</sup> EPA (1983) Nationwide Urban Runoff Program (NURP)

<sup>3</sup> Pitt et al. (2004) National Stormwater Quality Database (NSQD)

Crunkilton et al. (1997) noted a strong correlation between TSS and PAHs in runoff samples, but also observed dissolved PAHs in waters that tested toxic, although PAHs were only one of several observed potential toxicants in

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the runoff. In toxicity testing of PAH-laden stormwater runoff, Ireland et al. (1996) observed a reduction in toxicity when the organic PAH containing fraction was removed from the samples. They also observed in all runoff samples where TSS were removed by filtration, an increase in toxicity indicating the PAHs were either in solution or sorbed to organic carbon. The high COD/TSS ratio for this study indicates that most of the constituents in the runoff are either in the aqueous phase or bound to colloidal material that passes through traditional TSS filters. While particulate release due to vehicular abrasion is demonstrated in other studies (Mahler et al., 2004 and 2005), even without abrasion sealants may release PAHs.

PAH concentrations were regressed against COD concentrations, TOC concentrations, and volatile suspended solids (VSS) concentrations. None of these regressions showed a correlation, despite literature suggesting that VSS is a good surrogate parameter for organic compounds that have an octanol-water partition coefficient ( $K_{ow}$ ) greater than  $10^5$  (Novotny and Olem, 1994). Ten of the 17 PAH compounds analyzed in this study have a  $K_{ow}$  coefficient greater than  $10^5$ . This lack of relationships among PAHs and other water quality parameters (COD, TOC, and VSS) suggests that these water quality parameters are not surrogates for PAHs.

### **Instituting Stormwater Controls and Management Options for Sealed Surfaces**

In the full-scale testing, runoff collected from the coal tar-sealed surface indicated greater PAH concentrations than the asphalt emulsion and the unsealed runoff. Pitt et al. (1995) observed that treatment processes, particularly sediment removal can reduce toxicity of runoff. Therefore, routing, such as travel time, distance and disconnection (i.e., not directly connecting impervious surfaces to receiving water bodies), and storage and treatment can have a dramatic effect on the content of PAHs in a runoff discharge. Treatment by sedimentation, however, does not actually reduce the toxicity of the PAHs, which will still be present in the aqueous phase or in the sediments. These sediments need to be managed and disposed of properly. The long-term accumulation of PAHs in the sediment potentially leads to chronic toxicity effects. Prevention of PAH contaminated particles from entering the waterway through application of stormwater best management practices (BMPs), such as berms or stormwater ponds, was demonstrated by Bommarito et al. (2010) and Crane et al. (2010), respectively. Any resuspension of sediments in a stormwater control can ultimately reach a receiving water body, therefore transferring the sediment load to the receiving water body.

Routing and treatment of stormwater runoff appear to be effective controls in reducing the toxicity of runoff in general. PAHs are assumed to be bound to particulates, which could imply that sedimentation might treat runoff and remove PAHs. This study indicates that there is increased risk in the period immediately after sealant curing when the PAHs may not be associated with sediments. PAHs in the dissolved-phase or particle free state are not readily removed by conventional treatment methods (Crisafulli et al., 2008) used in stormwater (e.g., sedimentation, filtration) or even advanced methods used in wastewater treatment plants (e.g., coagulation, flocculation). In PAH-contaminated wastewater (specifically creosote-contaminated), fixed-film bioreactors removed and degraded the PAHs, while only removal by sorption was observed in wetlands (Tremaine et al., 1994). Biodegrading was easier for the lower molecular weight PAHs and was less effective on larger molecular weight PAHs. The current suite of recommended stormwater BMPs are passive systems that are not subject to the same level of oversight and operational rigor as wastewater treatment plants, the latter of which appears necessary to thoroughly treat PAHs.

There are still many parts of the country where stormwater routing is insufficient in both time and length to reduce toxicity to receiving waters. Directly connected surfaces can deliver large volumes and concentrations of toxic substances in runoff during and after a sufficient rainfall event. In a study by Scoggins et al. (2007), PAHs were measured in the receiving water downstream of parking lots with a corresponding reduction in the biodiversity of benthic communities.

Due to the expense of retrofitting control measures into existing stormwater sewage and discharge systems, many municipalities may opt for banning coal tar-based sealants due to PAH content and related chronic toxicity effects on the environment. An alternative is to implement retrofitted stormwater controls at the point of discharge; however, there is currently no authorizing regulatory framework nationally. Implementation of a retrofitted stormwater treatment and control system would require a municipality to manage and control sediments collected in the control

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system, including those sediments contaminated by PAHs. Crane et al. (2010) found high enough concentrations of PAHs in stormwater BMPs to require contaminated sediment disposal, an additional expense to municipalities.

### **Sealant Application Issues**

The Pavement Coatings Technology Council has issued a fact sheet regarding the correct application of sealant in order to reduce PAH impacts to receiving waters. The recommendations include: ensuring that no significant rainfall will occur within forty-eight hours after sealant application; applying sealant when temperatures are higher than 15°C and rising; and prohibiting parking for at least twelve hours after sealant application (Pavement Coatings Technology Council, 2010). These recommendations may differ from the manufacturer's instructions given on the containers for sealant products.

A regional analysis dividing the continental U.S. into eight regions found that the mean interval for runoff-producing rainfall events in the summer ranges from a low of 76 hours in the Northeast, to a high of 425 hours in the arid Southwest; annual means range from 73 hours to 277 hours (Driscoll et al., 1986). All eight regions have coefficient of variations exceeding values of 1, which indicates that the standard of deviation of the predicted period between storms never drops below that predicted period. One therefore cannot statistically predict the period between rainfall events to a precision that would eliminate the potential for runoff. Sub-regional and local analysis may increase the precision of inter-event period, but broad metrics like these are typically used in stormwater routing and treatment design.

### **Future Research Needs**

The literature is still lacking an in-depth study of asphalt sealant runoff examining both coal tar sealants or asphalt emulsion alternatives and the potential for acute toxicity, or lack thereof, to aquatic organisms in the water. Unless runoff samples are measured for PAH content and tested for toxicity, there is no direct linkage to the acute toxicity threat to aquatic life in receiving waters due to the prevailing assumption in the literature that PAHs are attached to particulate matter, and that the resulting water quality issue is primarily a problem of contaminated sediment and associated chronic toxic effects. The needed study must evaluate multiple screening studies for health endpoints so as to pass adequate judgment on the toxicity discussion.

Alternatives to asphalt- and coal tar-based sealants are limited. There are acrylic co-polymers available on the market but they are more expensive; although they claim to be less toxic, the actual toxicity of these alternatives is unknown. Some organic concrete sealants include acrylic sealers, epoxy coatings, urethane sealants, polyureas, and polyaspartics but these may not be relevant to sealing asphalt. The ability of these concrete sealants to be effective on unsealed asphalt surfaces is unknown. Future research needs include finding an inexpensive alternative to either asphalt- or coal tar-based products (i.e., new soy-based sealants).



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