



June 1, 2020

#### VIA ELECTRONIC SUBMISSION

EPA Headquarters Office of Water Office of Wastewater Management (4203M) 1200 Pennsylvania Avenue NW Washington, D.C. 20460

> Re: Joint Comments of the Pavement Coatings Technology Council and the Coke Oven Environmental Task Force on National Pollutant Discharge Elimination System: 2020 Issuance of the Multi-Sector General Permit for Stormwater Discharges Associated with Industrial Activity Docket ID No. EPA-HQ-OW-2019-0372

Dear Sir or Madam:

The Pavement Coatings Technology Council ("PCTC") and the Coke Oven Environmental Task Force ("COETF") appreciate the opportunity to file the following comments regarding the U.S. Environmental Protection Agency's ("EPA") Draft National Pollutant Discharge Elimination System ("NPDES") General Permit for Stormwater Discharges From Industrial Activities, 85 Fed. Reg. 12288 (March 2, 2020) (referred to here as the "MSGP" or the "2020 Proposed MSGP" or the "Proposal"). These comments focus on one issue: EPA's proposal, in the 2020 Proposed MSGP, to establish eligibility criteria related to the application of refined coal-tar sealcoat ("RTS"). This proposal is flawed for numerous legal, policy, scientific, and industry-related reasons, and EPA should not finalize it.

#### **The Commenters' Interest**

The PCTC is an industry trade association that represents North American manufacturers of asphalt- and refined coal tar-based pavement sealants (ABS and RTS, respectively) and their suppliers. Sealant manufacturers are predominantly small, family-owned businesses.

The COETF, which is managed by the American Coke and Coal Chemicals Institute ("ACCCI"), was formed in 1996 by ACCCI's members that produce metallurgical coke via the "by-product recovery" process, to address collaboratively the major environmental issues affecting them. The COETF represents five of the six domestic companies that produce metallurgical coke via this process, including three integrated steel companies that operate six "by-product recovery" coke plants (20 batteries) and two independently owned/operated "merchant" companies that operate two "by-product recovery" coke plants (4 batteries). One of the by-products that all these plants produce is crude coal tar, which is purchased by tar refiners, among others, as critical

Docket ID No. EPA-HQ-OW-2019-0372 June 1, 2020 Page 2

feedstock for the production of refined tar products. One of their refined tar products is RT-12 grade, which is a critical feedstock - base tar - used in producing RTS.

#### **Background on RTS Eligibility Provisions in 2020 Proposed MSGP**

On June 16, 2015, EPA published in the Federal Register a notice that it had issued the Multi-Sector General Permit ("2015 MSGP"). Several interest groups filed petitions for review of the 2015 MSGP, which were consolidated in the United States Court of Appeals for the Second Circuit.<sup>1</sup> Before the parties filed any briefing in the MSGP Litigation, they entered into settlement discussions to resolve the Petitioners' challenges to the 2015 MSGP. One of the Petitioners' issues involved MSGP permittees' use of coal tar sealants. Accordingly, in the settlement agreement resolving that litigation, dated August 16, 2016 ("2016 Settlement"), EPA agreed "to propose for comment a condition of eligibility that operators who, during their coverage under the next MSGP, will use coal tar sealant to initially seal or to re-seal pavement and thereby discharge polycyclic aromatic hydrocarbons (PAHs) in stormwater are not eligible for coverage under the MSGP and must either eliminate such discharge or apply for an individual permit." 2020 Proposed MSGP Fact Sheet, at 3. EPA also agreed "to take final action" on its proposal.

The 2020 Proposed MSGP includes a proposed new eligibility criterion related to the use of coal-tar sealcoat under the MSGP, which provides as follows:

To be eligible for coverage under this permit, you must not have any stormwater discharges from paved surfaces that will be initially sealed or re-sealed with coaltar sealcoat where industrial activities are located during your coverage under this permit. EPA may authorize your coverage under this permit if you eliminate such discharge or EPA will notify you that an individual permit application is necessary per Part 1.3.7.

Regarding the proposed eligibility criterion related to the use of coal-tar sealant, EPA has requested comment on:

- Whether the permit should include an eligibility criterion related to the application of coal-tar sealcoat to paved areas where industrial activities are located;
- Any studies that provide data on the level of PAHs from coal-tar sealed pavements, the sources of measured PAHs in the aquatic environment, the levels of PAHs in fish and seafood, and associated chemical and biological impacts that may occur via stormwater discharges; and,
- Whether or to what extent requiring facilities to implement specific stormwater control measures under the MSGP to control and treat PAH-laden discharges from surfaces paved with coal-tar sealcoat is an appropriate alternative to the proposed eligibility criterion and, if so, what those control measures should be.

<sup>&</sup>lt;sup>1</sup> Waterkeeper Alliance, Inc., et al., v. U.S. EPA, Case Nos. 15-2091 (lead), 15-2259 (consolidated). 15-2428 (consolidated), and 15-3315 (consolidated) (collectively, "the MSGP Litigation").

Docket ID No. EPA-HQ-OW-2019-0372 June 1, 2020 Page 3

#### **Concerns with RTS Eligibility Provisions in 2020 Proposed MSGP**

EPA's proposal to bar facilities using RTS from eligibility under the MSGP amounts to a *de facto* ban on RTS usage. This proposal is flawed for numerous legal, policy, cost, scientific, and product-related reasons. Those flaws in the EPA RTS proposal are described in detail in the appendices to these comments.

Appendix A focuses on legal and policy-related problems with the proposal, including the major cost impacts that will result from it. Appendix B describes the scientific problems with the proposal. And, Appendix C discusses why, despite EPA's assertions to the contrary, there are no alternative products with performance and cost similar to coal tar sealant, making it the preferred choice among pavement sealants. Our major points in each of these areas are summarized below:

Legal and Policy-Related Concerns

- <u>EPA lacks legal authority to condition permit eligibility on RTS use</u>. The net impact of EPA's action, if finalized, would result in a *de facto* ban on RTS usage. EPA lacks legal authority to ban any product under the Clean Water Act (CWA)
  - <u>EPA lacks legal authority to ban products through the CWA</u>. While the CWA authorizes EPA to establish effluent limitations designed to meet water quality standards within a receiving body, it does not authorize the agency to impose past, present or future product-usage permit conditions, especially when it cannot quantify any water quality benefits.
  - <u>EPA's proposal is inconsistent with its CWA authority.</u> The CWA, through the NPDES permit program, limits the discharge of pollutants from "point sources" into waters of the United States, based upon the capabilities of the practices or technologies available to control such discharges. EPA cannot regulate an activity or "meddle in the facility." It can regulate only an existing pollutant discharge from a point source.
  - EPA's proposal is inconsistent with its own industrial stormwater permitting program. In regulating industrial stormwater through permits, EPA has followed an orderly, structured process that is guided by the structure of the CWA and EPA's own regulations. EPA's proposal to focus here on runoff of RTS from parking lots is inconsistent with that structure and with the information that EPA has collected over the course of implementing the MSGP.
  - <u>EPA has not gone through the regulatory steps required to establish</u> <u>Technology-Based Effluent Limitations (TBELs) for PAHs</u>. For technology-based controls, the CWA authorizes EPA to promulgate effluent limitations guidelines or other technology-based controls, including non-numeric measures. Historically, EPA has implemented technologybased controls through imposing "best management practices" (BMPs) in the MSGP. While EPA can establish technology-based limitations on

discharges of pollutants, it must leave facilities free to choose the means by which they meet such limitations.

- <u>EPA must operate within its CWA authority to impose water quality-based</u> <u>controls</u>. Rather than adopting the eligibility provision to address potential pollutants associated with RTS being used at regulated industrial sites, in areas of industrial activities, EPA should rely upon its authorized powers to control pollutant discharges. Here, EPA has not attempted to promulgate WQBELs for PAHs.
  - <u>The 2020 Proposed MSGP is inconsistent with the 2016 Settlement</u>. The 2020 Proposed MSGP language is inconsistent with the language of the 2016 Settlement, which conditions eligibility for operators who use RTS *and thereby discharge* PAHs. The 2020 Proposed MSGP simply conditions eligibility on the use of RTS without regard to whether or not PAHs are discharged.
  - EPA has declined to impose water quality-based controls for PAHs. There is no evidence in the history of the MSGP demonstrating that EPA previously believed PAHs warranted monitoring, let alone an effective ban on a specific industrial practice that could involve the release of PAHs. In fact, the lack of monitoring for PAHs over the history of the MSGP is consistent with the fact that the CWA 303(d) state water quality reports rarely identify PAHs as a cause of impairment. The lack of evidence that PAHs cause widespread water quality impacts undermines EPA's basis for seeking to eliminate PAH discharges through a ban on RTS.
  - <u>The MSGP already imposes requirements on discharges to PAH-limited</u> <u>waters</u>. Section 4.2.4.1 of the 2020 Proposed MSGP provides for monitoring of discharges to impaired waters without an EPA-approved or established TMDL, as well as for discharges to impaired waters with an EPA-approved or -established TMDL. This approach represents a more nuanced and appropriate process for monitoring and addressing discharges to PAH-limited waters than the proposed eligibility requirements.
  - EPA has not established a need for water quality-based controls. EPA's only possible legal basis for water quality-based controls for PAHs is establishing requirements based on a "reasonable potential" analysis with respect to attainment of state water quality standards. However, EPA has provided no such explanation, nor any supporting analysis. EPA simply assumes that regulation of RTS is necessary to reduce PAH releases, without any quantitative analysis.
  - <u>The proposed PAH requirements are inconsistent with other NPDES</u> <u>actions</u>. EPA has not taken action to prohibit the use of RTS in any other permit in the NPDES program, which calls into question EPA's claim that use of RTS must be banned in the 2020 Proposed MSGP.
- EPA's proposal raises unprecedented Constitutional and policy concerns.
  - <u>EPA cannot overcome Commerce Clause limitations</u>. The CWA provides no indication that Congress ever intended for EPA to use its authority to ban

products and includes many signs that it did not. Interpreting the CWA to authorize such a radical measure would violate well-established principles of federalism that limit federal power and preserve state power.

- <u>EPA can only pursue a product ban under TSCA and only if it meets all of the TSCA requirements</u>. The CWA does not authorize EPA to ban a product. In fact, EPA only has authority to ban products under the Toxic Substances Control Act ("TSCA"). Moreover, EPA can only implement a ban under TSCA if the ban meets the high legal burden set forth in the statute and regulations thereunder. Here, EPA has not even attempted to meet that burden.
- <u>EPA's cost-impact analysis is deficient</u>. EPA's cost-impact analysis addresses potential costs associated with the use of coal tar-based sealant alternatives and the cost of compliance with proposed COD monitoring and PAH benchmarks. However, it fails entirely to account for the significant economic impacts across the RTS value chain—from coal tar producers, to coal tar refiner and RTS manufacturers, to RTS applicators—which has an estimated annual economic value in excess of \$100 million dollars.
- <u>EPA's proposal is inconsistent with the Regulatory Flexibility Act ("RFA")</u>. This 2020 Proposed MSGP rulemaking will have a "significant economic impact on a substantial number of small entities," thus triggering the RFA. However, EPA took no action to comply with the RFA, even though it could and should have been able to ascertain the significant economic impact on small businesses.

Science-Related Concerns

- <u>RTS is not a significant source of PAHs in the aquatic environment</u>. The proposed eligibility criterion fails to acknowledge and address the weight of current scientific evidence and current best practices in sediment risk assessment.
  - <u>EPA's own CWA guidance acknowledges that, in natural aquatic environments, PAHs strongly adsorb to organic materials and are not bioavailable—and, thus, require specialized risk assessment techniques.</u> None of the literature cited in the Fact Sheet to support the proposed RTS eligibility restriction reflects any of EPA's Toxicity Identification Evaluation (TIE) guidance, guidance on developing Equilibrium Partitioning Sediment Benchmarks (ESBs) for PAHs, or any other method of identifying toxicity or the causes of measured toxicity in sediments in the real world.
  - <u>PAHs in coal-tar sealcoat particles bond particularly strongly and are not</u> <u>bioavailable</u>. EPA's guidance on ESBs and TIEs is grounded in EPA's recognition that, outside the laboratory, exposure to individual PAH compounds is limited by the physical-chemical characteristics of PAHs.
  - <u>Surfaces sealed with RTS do not contribute significant amounts of PAHs to</u> <u>aquatic environments</u>. Many studies show very low levels of PAHs from RTS-sealed surfaces even under unrealistic experimental conditions.

Docket ID No. EPA-HQ-OW-2019-0372 June 1, 2020 Page 6

Rather, the scientific consensus is that the great majority of PAHs in the environment come from atmospheric deposition from combustion sources. Additionally, scientists acknowledge that it is difficult to distinguish PAHs from widely diverse combustion sources, because PAH chemical profiles from those sources are so similar. Further, modeling efforts to apportion PAHs among sources are unreliable, as they rely on a PAH "signature" for RTS that was not generated for that purpose and has never been validated.

- <u>RTS is not a significant source of aquatic or human toxicity</u>. PAHs in the aquatic environment are not particularly toxic to either aquatic life or humans.
  - The studies cited in the Fact Sheet do not rely on EPA's own guidance for assessing the aquatic toxicity of PAHs. Because PAH-containing solids are not very bioavailable or bioaccessible, PAH concentrations have not been found to correlate with observed aquatic toxicity. In recognition of these facts, EPA developed both its ESB and TIE guidance documents so that evaluation of risks for exposures to PAHs in the aquatic environment would more closely reflect their actual toxicity. Environmental field studies of PAHs that employ EPA's recent guidance have not observed them to be toxic. Moreover, the Fact Sheet exclusively cites literature that reflects none of the scientific advances embodied in the ESB and TIE guidance. Rather, the Fact Sheet cites literature that relies on probable effect level values, without any citation to the bioavailability literature.
  - O PAHs' toxicity to humans was widely overestimated until recently- a fact not recognized by the Fact Sheet. The limited bioavailability of PAHs in the environment also complicates the process of assessing potential human health risks from PAH exposures. This complication is exacerbated by the tendency of human health risk assessments of PAHs to rely on outdated EPA guidance documents that were issued more than 25 years ago. The practice of estimating human health risks that could be associated with exposure to PAH-containing substances based on bioassays of animals exposed to substances in forms that do not occur outside the laboratory has been found to be unrepresentative of exposures to PAHs in the environment and to lead to unreliable predictions of toxicity.
- <u>The proposed rule has no rational basis</u>. The weight of the best available science and evidence indicates that PAHs from RTS are not a cause of aquatic toxicity and do not pose risks to humans. Accordingly, the proposed eligibility restriction lacks any rational basis, as it would not reduce risks to human health and the environment—EPA's stated purpose for proposing the restriction.

Industry-Related Concerns

• <u>Alternative products with equivalent performance and cost to RTS are not</u> <u>available</u>. In its 2020 Proposed MSGP Fact Sheet, EPA contends, without supporting facts, that it has identified alternatives that are similar in product performance and cost to RTS. While there are certainly alternatives to RTS, none of them come close to RTS in terms of performance and cost. As documented below, the alternatives do not perform as well as RTS and their lifetime costs are higher.

- <u>RTS performs better than the alternatives</u>. RTS lasts longer and requires less maintenance than RTS alternatives.
  - <u>Asphalt Sealants</u>. Asphalt-based emulsions generally have life cycles of two to three years, whereas RTS will generally last four to six years.
  - <u>Acrylic Sealants</u>. Acrylic coatings (which are often used on tennis courts) are brittle, resulting in the need to resurface courts every few years even though they are not subject to vehicle traffic.
  - <u>Permeable Pavement Products</u>. Particulates infiltrate the pore space of permeable pavement products, leading to declining effectiveness over time and eventual complete clogging. Accordingly, permeable products are usually inappropriate for application on surfaces with vehicle traffic. Additionally, the costs of alternative permeable products at the point of application are often greater than those for RTS products, and the cost of maintenance can be much greater than maintenance of traditional pavements.
- <u>No alternative is cost-competitive</u>. Sealcoating is an integral part of a preventive maintenance program that extends the useful service life of a pavement asset, and it costs considerably less than repaying or other measures that may be needed later to correct chronic or acute pavement problems. A comprehensive assessment of the costs associated with any sealcoating must include the costs incurred over the life cycle of a paved surface, which is where RTS clearly outperforms competitors.
- <u>Independent sources confirm that RTS is the preferred product</u>. The cost and performance qualities discussed above are confirmed by independent sources. Those sources indicate that asphalt life-cycle is two to three years, while that for RTS is four to six years even though the cost of installation is roughly the same.

For all of the reasons summarized above, and laid out in detail in the appendices to these comments, EPA should withdraw the proposed RTS provisions in the 2020 Proposed MSGP. The MSGP should be finalized without those provisions.

Docket ID No. EPA-HQ-OW-2019-0372 June 1, 2020 Page 8

PCTC and the COETF appreciate the opportunity to submit these comments. If you have any questions, or would like any further information regarding the issues raised in these comments, please contact either of us at, for Anne LeHuray, <u>alehuray@pavementcouncil.org</u>; (703) 299-8470 or, for Dave Ailor, <u>dailor@accci.org</u>; (703) 795-3541.

Sincerely,

anne

Anne LeHuray, Ph.D. Executive Director Pavement Coatings Technology Council

Juli

David Ailor ACCCI President On Behalf of the COETF

## **TABLE OF CONTENTS**

### Appendix A Legal and Policy-Related Concerns

I.	EPA Lacks Legal Authority to Limit Permit Eligibility for RTS A1			
	A.	EPA Has No Authority to Ban a Product Under the Clean Water Act A1		
	B.	EPA's Proposal is Inconsistent with its Clean Water Act Authority A2		
	C.	EPA's Proposal is Inconsistent with Its Own Industrial Stormwater Program A		
	D.	EPA Has Not Gone Through the Regulatory Steps Necessary to Establish Technology-Based Effluent Limitations for PAHs		
	E.	EPA N Contro	Aust Operate Within Its CWA Authority to Impose Water Quality-Based	
		1.	The 2020 Proposed MSGP Language is Inconsistent with the 2016 Settlement	
		2.	EPA Has Declined to Impose Water-Quality-Based Controls for PAHs.	
		3.	The MSGP Already Imposes Requirements on Discharges to PAH- Limited Waters	
		4.	EPA Has Not Established a Need for Water Quality-Based Controls A8	
		5.	Proposed PAH Requirements are Inconsistent with Other NPDES Actions	
II.	EPA's Proposed Ban Has Constitutional and Statutory Flaws			
	A.	EPA Cannot Overcome Commerce Clause Limitations A10		
	B.	B. EPA Can Only Pursue a Product Ban Under TSCA and Only If It Meets All of the TSCA Requirements		
III.	EPA's	Cost-I	mpact Analysis is Deficient	
IV.	EPA's	Propos	al is Inconsistent with the Regulatory Flexibility Act	

## Appendix B Science-Related Comments

I.	Refined Coal-Tar Sealcoat (RTS) Is Not a Significant Source of PAHs in the Aquatic Environment, and Its Use Does Not Present Risks to Aquatic Life or Humans. Thus, There Is No Rational Basis for EPA's Proposed RTS Eligibility Restriction				
	A.	Refined Coal Tar Sealcoat Is Not a Significant Source of Biologically Available PAHs in the Aquatic Environment.	B2		

1. EPA's own CWA guidance acknowledges that, in natural aquatic environments, PAHs strongly adsorb to organic materials and are not

		bioavailable—and, thus, require specialized risk assessment techniques		
2. PAHs in coal tar sealcoat particles bond particularly strongly an particularly not bioavailable.				
	3.	Surfaces sealed with RTS do not contribute significant amounts of PAHs to aqueous environments		
<ul> <li>B. Coal Tar Sealcoat Is Not a Significant Source of Aquatic or Human Toxicity</li> <li>1. The studies cited in the Fact Sheet do not rely on EPA's own guidance for assessing the aquatic toxicity of PAHs.</li> </ul>				
				2.
C.	An RTS Ban May Actually Cause Environmental HarmB32			
D.	The Pr	roposed Rule Does Not Have a Rational Basis		

## Appendix C Industry-Related Concerns

I.	Despite EPA's Multiple Assertions that There Are Alternative Products with Similar Performance and Cost to RTS, No Such Alternatives Are AvailableC1			
			1.	Asphalt Sealants
		2.	Acrylic Sealants	.C4
		3.	Permeable Pavements	.C5
	F.	Life-C	ycle Cost-Competitiveness of RTS Is Superior	.C5
	G.	Indepe	ndent Sources Recognize the Superior Value and Performance of RTS	.C7

## Appendix D Kane Driscoll Report

Comments	D2
Summary	D3
Acknowledgement	D4
References	D4

## Appendix E O'Reilly Report

SummaryE1
-----------

Comments	E2
Acknowledgement	E9
References	E9

## Appendix A Legal and Policy-Related Concerns

#### I. EPA Lacks Legal Authority to Limit Permit Eligibility for RTS

#### A. EPA Has No Authority to Ban a Product Under the Clean Water Act.

Part 1.1.8 of the 2020 Proposed MSGP contains an eligibility provision conditioned on the use of refined coal tar-based sealants (RTS). While Part 1.1.8 is not a direct legal ban, the net impact of EPA's action, if finalized, would result in a *de facto* ban on RTS usage. EPA lacks legal authority to ban any product under the Clean Water Act (CWA), as explained more fully below.

Courts commonly find that facially permissive regulations or incentives are coercive in practice.<sup>1</sup> When an incentive amounts to a requirement, it will invoke statutory and Constitutional limits on regulatory authority. Under the CWA, Congress has directed EPA to develop, issue, and enforce *permits*, not *prohibitions*.<sup>2</sup>

Section 301 of the CWA provides in relevant part, "Except in compliance with [the CWA], the discharge of any pollutant is unlawful."<sup>3</sup> The D.C. Circuit has interpreted this language as a limit on EPA's regulatory authority and rejected measures that went beyond the regulation of pollutants.<sup>4</sup> In 1987, the court struck down an "outright ban" on pre-permit construction of new sources.<sup>5</sup> Notably, the court refused to find any implicit Congressional intent to empower EPA to act beyond the narrow confines of its statutory mandate:

[A]lthough Congress full well knows how to confer the power to restrict construction pending issuance of a permit, it chose not to do so here. For example, the [Clean Water] Act includes no requirement that permit issuance or permit review proceedings precede construction, as is the case with the Clean Air Act. Neither does the Clean Water Act confer upon EPA permitting authority over the construction of facilities, as do the Atomic Energy Act and the Federal Power Act. Consequently, this is not a case in which a construction ban is clearly within the agency's jurisdiction under its organic statute. The fact that Congress has vested some agencies with such power demonstrates that when Congress wanted to extend that power, "it knew how to do so and did so expressly."<sup>6</sup>

<sup>5</sup> *NRDC*, 822 F.2d at 129.

<sup>&</sup>lt;sup>1</sup> 567 U.S. 519, 581; *see also S. Dakota v. Dole*, 483 U.S. 203, 211 (1987) ("in some circumstances the financial inducement [] might be so coercive as to pass the point at which 'pressure turns into compulsion."").

<sup>&</sup>lt;sup>2</sup> Defining eligibility for permits in a way that would result in an effective product ban turns this mandate on its head. Adopting permit conditions that prohibit coal tar sealants exceeds EPA's limited authority under the CWA.

<sup>&</sup>lt;sup>3</sup> 33 U.S.C. § 1311(a).

<sup>&</sup>lt;sup>4</sup> See Nat. Res. Def. Council, Inc. v. U.S. EPA, 859 F.2d 156, 169–170 (D.C. Cir. 1988) (citing Nat. Res. Def. Council, Inc. v. U.S. EPA, 822 F.2d 104, 129 (D.C. Cir. 1987). ("EPA's jurisdiction [under the CWA] is *limited* to regulating the discharge of pollutants. . . .") (emphasis added)).

<sup>&</sup>lt;sup>6</sup> *Id*. (citations omitted).

Relying on the same rationale, the D.C. Circuit held in 1988 that the CWA does not empower EPA to regulate point sources themselves. As the court explained, "EPA can properly take only those actions authorized by the CWA—allowing, prohibiting, or conditioning the pollutant discharge... EPA's jurisdiction under the [CWA] is limited to regulating the discharge of pollutants. Thus, just as EPA lacks authority to ban construction of new sources pending permit issuance, so the *agency is powerless to impose permit conditions unrelated to the discharge itself*."<sup>7</sup>

As described in Part II of these comments, PAHs from RTS are not a source of significant toxicity in the aquatic environment. EPA has not quantified the potential PAHs from RTS applications at sites with stormwater associated with industrial activity. And, EPA lacks the authority to proscribe a particular product. While the CWA does authorize EPA to establish water quality-based permit limitations designed to meet water quality standards within a receiving body,<sup>8</sup> it does not authorize the agency to impose past, present or future product-usage permit conditions, especially when it cannot quantify any water quality benefits.<sup>9</sup>

EPA asserts that industrial dischargers are not required to apply for the MSGP, but the fact that there are theoretical NPDES permit alternatives is illusory. Courts commonly find that facially permissive regulations or incentives are in practice coercive. As Chief Justice Roberts noted in *Nat'l Fed'n of Indep. Bus. v. Sebelius*, what appears to be an "inducement" can actually be a "gun to the head" when the consequences of choosing an alternative are dire.<sup>10</sup> When an incentive amounts to a requirement, it will confront statutory and Constitutional limits on regulatory authority.

#### B. EPA's Proposal is Inconsistent with its Clean Water Act Authority.

Congress enacted the CWA "to restore and maintain the chemical, physical, and biological integrity of the Nation's waters." 33 U.S.C. \$1251(a). CWA Section 301(a) prohibits "the discharge of any pollutant" by any person, except as authorized by the Act. 33 U.S.C. \$1311(a). To regulate these discharges, CWA Sections 301 and 304 authorize EPA to establish "effluent limitations," defined as restrictions placed upon pollutants that "are discharged from *point sources* into navigable waters." *Id.* \$\$ 1311, 1314(b), 1362(11) (emphasis added); *see also id.* \$ 1342(a)(1).

CWA Section 402 provides an exception to CWA Section 301's prohibition by allowing certain pollutant discharges to be authorized by an NPDES permit, provided that the discharges meet appropriate "effluent limitations" contained in the permit. 33 U.S.C. § 1342(a). Thus, the CWA, through the NPDES permit program, limits the discharge of pollutants from "point sources"

<sup>&</sup>lt;sup>7</sup> *Id.* at 169-70 (emphasis added).

<sup>&</sup>lt;sup>8</sup> *See* 33 U.S.C. 1342(a) (requiring that discharges meet all applicable requirements of 33 U.S.C. § 1312 authorizing EPA to establish water quality-related effluent limitations).

<sup>&</sup>lt;sup>9</sup> See NRDC, 859 F.2d at 170, n. 8 ("But even considering the whole of EPA's authority under the CWA, the imposition of non-discharge related conditions in new point source permits does not fall within its scope.")

<sup>&</sup>lt;sup>10</sup> 567 U.S. 519, 581 (2012); *see also S. Dakota v. Dole*, 483 U.S. 203, 211 (1987) ("in some circumstances the financial inducement [] might be so coercive as to pass the point at which 'pressure turns into compulsion."").

into waters of the United States, based upon the capabilities of the practices or technologies available to control such discharges. 33 U.S.C. §§ 1311(b)(2), 1314(b), 1316(b)(1)(B).

Congress did not provide EPA with unbridled authority. Rather, the CWA "authorizes the EPA to regulate, through the NPDES permitting system, *only* the discharge of pollutants." *Waterkeeper Alliance, Inc. v. EPA*, 399 F.3d 486, 504 (2d Cir. 2005) (emphasis added). As the D.C. Circuit has explained, "[t]he statute is clear" and contains no language that "undercuts the plain meaning of the statutory text;" EPA may not "meddl[e] inside a facility," because it only has authority over the discharge of pollutants from a point source, and "Congress clearly intended to allow the permittee to choose its own control strategy." *American Iron and Steel Institute v. EPA*, 115 F.3d 979, 996 (D.C. Cir. 1997). EPA "is powerless to impose conditions unrelated to the discharge itself." *N.R.D.C. v. EPA.*, 859 F.2d 156, 170 (D.C. Cir. 1988) (EPA cannot regulate point sources themselves, only the discharge of pollutants); *Service Oil, Inc. v. EPA*, 590 F.3d 545, 551 (8<sup>th</sup> Cir 2009) ("the Clean Water Act gives EPA jurisdiction to regulate... only *actual* discharges—not potential discharges, and certainly not point sources themselves.") (emphasis in original).

In the 2020 Proposed MSGP, EPA attempts to avoid these precedents by characterizing the proposed RTS eligibility restrictions as limitations on discharges from surfaces where industrial activities are located. However, in fact, EPA is limiting the activity of sealing paved surfaces with one particular type of sealer. Further, the clear focus is not on the ongoing pollutant discharges. Rather, EPA is regulating to control possible future pollutant discharges ("surfaces that *will be* initially sealed or re-sealed with coal-tar sealcoat"). As the D.C. Circuit has made clear, EPA cannot regulate an activity or "meddle in the facility;" it can regulate only an existing pollutant discharge from a point source.

#### C. EPA's Proposal is Inconsistent with Its Own Industrial Stormwater Program.

In regulating industrial stormwater through permits, EPA has followed an orderly, structured process, which is guided by the structure of the CWA and EPA's own regulations. EPA's proposal to focus here on the use of RTS on parking lots is inconsistent with that structure and with the information that EPA has collected over the course of implementing the MSGP. In order to illustrate this point, some explanation of that regulatory structure governing industrial stormwater permits is in order.

In 1987, Congress added CWA Section 402(p), which established a phased approach to regulating stormwater discharges, as needed. First, Congress provided in Section 402(p)(1) a blanket exemption for all stormwater discharges, except those requiring NPDES permitting as set forth in Sections 402(p)(2)-(4). In Phase I, Congress required NPDES permits for stormwater discharges from "industrial activities" and certain medium and large municipal separate storm sewer systems (MS4s). 33 U.S.C. § 1342(p)(1)-(4). For Phase II, Congress instructed EPA to study all remaining stormwater discharges to determine the nature of pollutants in those discharges, and establish "procedures and methods to control stormwater discharges to the extent necessary to mitigate impacts on water quality." *Id.* §1342(p)(5). Based on that study, EPA was required to promulgate regulations designating any additional stormwater discharge to be regulated and establishing a "comprehensive program to regulate such designated sources." *Id.* §1342(p)(6). Hence, the 1987 CWA amendments established "an orderly procedure which will enable the major

contributors of pollutants to be addressed first, and all discharges to be ultimately addressed in a manner which will not completely overwhelm EPA's capabilities." 133 Cong. Rec. H168-03 (daily ed. Jan. 8, 1987) (statement of Rep. Roe).

EPA issued its Phase I stormwater regulations in 1990, establishing NPDES permit regulations for stormwater discharges from 11 specific categories of "stormwater associated with industrial activity." *See* 55 Fed. Reg. 47990 (Nov. 16, 1990); 40 C.F.R. §122.26(b)(14)(i)-(xi). In developing the initial MSGP, EPA received massive quantities of industrial stormwater data from over 1,100 industrial group permit applications that it later classified into the initial 26 categories in the 1995 MSGP. EPA issued its Phase II stormwater rule in 1999 after conducting extensive studies of commercial, light industrial, and other sources of stormwater pollutants. In the end, EPA's major focus for the Phase II rule was to expand the existing construction and MS4 permit programs. The only change to its Phase I industrial stormwater permit program was to clarify industrial permitting issues for light industrial sources (so-called "category xi" sources). *See* 64 Fed. Reg. 68,722 (Dec. 8, 1999). Throughout this entire information collection and regulatory analysis processes, RTS was never targeted for possible stormwater regulation.

Moreover, during the Phase I program development, EPA analyzed whether to regulate pollutants associated with parking lots and concluded that parking lots and administrative areas, even at industrial sites, do not meet the definition of "associated with industrial activity." 55 Fed. Reg. at 48010. EPA later confirmed this in its "Stormwater Question & Answer II" document.<sup>11</sup> Additionally, EPA considered but rejected the need to regulate RTS in 2015, in part because parking lots were excluded from the definition of "industrial activity" associated with stormwater discharges.<sup>12</sup> At that time EPA properly questioned whether RTS controls would be effective, given that parking lots would not be affected by the general permit. In the new permit proposal, however, EPA fails to address this issue.

# D. EPA Has Not Gone Through the Regulatory Steps Necessary to Establish Technology-Based Effluent Limitations for PAHs.

As discussed above, EPA has no authority to ban a product under the CWA. Rather than using an eligibility provision to address potential pollutants associated with RTS being used at regulated industrial sites, in areas of industrial activities, EPA should rely upon its authorized powers to control pollutant discharges. The CWA equips EPA with two regulatory mechanisms for controlling pollutant discharges: technology-based limitations (TBELs) and water quality-based effluent limitations (WQBELs). For technology-based controls, the CWA authorizes EPA to promulgate effluent limitations guidelines<sup>13</sup> or other technology-based controls, including non-numeric measures.<sup>14</sup> Historically, EPA has implemented technology-based controls in the MSGP through imposing "best management practices" (BMPs).

<sup>&</sup>lt;sup>11</sup> U.S. EPA, "NPDES Storm Water Program Question and Answer Document Volume 2," EPA B33-F-93-002B (July 1993), *available at* <u>https://www3.epa.gov/npdes/pubs/owm0250.pdf</u>.

 $<sup>^{12}</sup>$  *Id.* at 75.

<sup>&</sup>lt;sup>13</sup> See, e.g., 33 U.S.C. § 1314(b).

 $<sup>^{14}</sup>$  *Id*.

For example, the 2015 MSGP directs permittees subject to TBELs to establish and implement BMPs.<sup>15</sup> With regard to discharges of PAHs from RTS use, EPA has not taken the regulatory steps required to establish TBELs. EPA can establish TBELs for industrial stormwater discharges that come into contact with RTS, but EPA must base the TBELs on the level of control attainable based on currently available technologies.<sup>16</sup>

To establish effluent limitations, EPA must specify the factors to be considered in evaluating available technologies; utilize them to identify the best control technologies, measures, and practices available; and, determine the degree of effluent reduction attainable using the best technology.<sup>17</sup> MSGP permittees then would have flexibility to implement technologies that meet that same level of effluent reduction. Apparently, EPA has not assessed controlling RTS-related pollutants through these standard NPDES processes. Instead, in the 2020 Proposed MSGP, EPA seeks simply to eliminate the use of RTS by conditioning eligibility on use of the product. This approach is improper and unauthorized under the CWA.

Even if EPA did fully develop TBELs to control or eliminate PAHs, it cannot require the use of any specific type of control technology (or, in this case, impose a ban on the use of a particular product).<sup>18</sup> In other words, even if EPA sets TBELs for PAHs in the new MSGP, it cannot ban the use of coal tar sealants. The CWA would still allow the permittee to implement its own control measures to meet any TBEL for PAHs.

For example, one of sealant industry's standard practices is to ensure that coal tar sealants are not applied within 24 hours before a predicted wet weather event. The practice could adequately reduce or eliminate PAHs in an industrial permittee's stormwater discharges, as EPA desires. However, conditioning eligibility for the MSGP on the use of coal tar sealants imposes a specific mandate on a permittee that allows no alternative means of compliance, in contravention of NPDES regulations. While EPA can establish technology-based limitations on discharges of pollutants, it must leave facilities free to choose the means by which they meet such limitations.<sup>19</sup>

As explained earlier, EPA's authority is limited to regulating the discharge of pollutants through point sources; it cannot regulate the activities that generate the pollutants. Here, EPA is attempting to regulate an "activity" (coating asphalt), not pollutants in stormwater discharges associated with industrial activity. Regulating the activity implies that EPA disfavors that activity or that industry, when other sources of PAHs are allowed to be covered by the MSGP. This raises an important concern: if EPA can disfavor an activity, then it may seek to disfavor other activities as well, and ban other products, which goes well beyond its CWA authority.

<sup>&</sup>lt;sup>15</sup> 2015 MSGP, at p. 14.

<sup>&</sup>lt;sup>16</sup> Id.

<sup>&</sup>lt;sup>17</sup> 33 U.S.C. § 1314(b).

<sup>&</sup>lt;sup>18</sup> See NPDES Permit Writers' Manual, at p. 49 (explaining, "The intent of a technology-based effluent limitation is to require a minimum level of treatment for industrial/municipal point sources based on currently available treatment technologies while allowing the discharger to use any available control technique to meet the limitations.")

<sup>&</sup>lt;sup>19</sup> See EPA, NPDES Permit Writers' Manual at 49, available at <u>https://www3.epa.gov/npdes/pubs/chapt\_05.pdf</u>

PCTC/COETF have provided EPA with appropriate TBEL approaches related to RTS usage that are consistent with EPA's TBEL approach throughout the MSGP, including ASTM standards for application or use of RTS.<sup>20</sup> If EPA is intent on addressing RTS in this permit, it should consider those other approaches.

It is worth noting that one of the justifications EPA provides in support of its ban on RTS use does not actually support an eligibility condition or ban. EPA cites its provision on cationic treatment in the Construction General Permit (CGP) as a precedent for its RTS provision here. But, the cationic treatment provision in the CGP is not similar to EPA's proposed eligibility condition here. The cationic treatment provision in the CGP simply provides that the operator must provide notice so that EPA can decide if additional controls or individual permits are needed for the facility. This regulatory approach is very different from the ban on RTS in the 2020 Proposed MSGP. EPA has provided no explanation for why it is treating RTS differently from how it treated cationic treatment in the CGP.

In the 2020 Proposed MSGP, EPA also proposes to require advance notification related to the use of cationic treatment chemicals due to concerns about their impacts on aquatic animals. While we do not support that proposed provision, EPA's disparate treatment of these two possible pollutants is not explained, or justified, demonstrating that the Agency's approach is arbitrary.

#### E. EPA Must Operate Within Its CWA Authority to Impose Water Quality-Based Controls.

# 1. The 2020 Proposed MSGP Language is Inconsistent with the 2016 Settlement.

In the 2016 Settlement, EPA agreed to propose for comment a condition of eligibility providing that "operators who, during their coverage under the next MSGP, will use coal tar sealant to initially seal or to re-seal pavement and thereby discharge PAHs in stormwater are not eligible for coverage under the MSGP and must either eliminate such discharge or apply for an individual permit." The 2020 Proposed MSGP, however, provides that "[t]o be eligible for coverage under this permit, you must not have any stormwater discharges from paved surfaces that will be initially sealed or re-sealed with coal-tar sealcoat where industrial activities are located during your coverage under this permit." The 2020 Proposed MSGP language is inconsistent with the language of the 2016 Settlement, which conditions eligibility for operators who use RTS *and thereby discharge* PAHs. The 2020 Proposed MSGP simply conditions eligibility on the use of RTS without regard to whether or not PAHs are discharged.

The 2020 Proposed MSGP fails to tie the eligibility requirement to actual water quality impacts. Notably, the 2020 Proposed MSGP eligibility language fails to acknowledge that BMPs can effectively reduce discharges of PAHs in stormwater. Effectively banning a class of products because of the potential to lead to discharges that might impair water quality is inconsistent not only with the 2020 Proposed MSGP, but also with this Administration's regulatory policy: "to alleviate unnecessary regulatory burdens."<sup>21</sup> EPA has failed to show that this eligibility condition

<sup>&</sup>lt;sup>20</sup> See infra App. B, Section I, fn.1.

<sup>&</sup>lt;sup>21</sup> E.O. 13777, § 1 (82 Fed. Reg. 12285, Mar. 1, 2017).

provides any water quality benefits, despite significant economic risk to a large number of manufacturers, formulators and applicators, including small businesses.

#### 2. EPA Has Declined to Impose Water-Quality-Based Controls for PAHs.

EPA stated in the 2020 Proposed MSGP Fact Sheet (Fact Sheet) that it agreed to consider all National Research Council (NRC) recommendations in developing the new proposed permit.<sup>22</sup> Regarding PAHs, the NRC recommended that EPA collect PAH discharge data before determining whether PAH monitoring would be warranted. The NRC stated that "additional information and data gathering could help EPA determine if benchmark monitoring is needed for sectors that have the potential to release PAHs."<sup>23</sup> The NRC did not mention RTS specifically, and it certainly did not suggest that RTS should be subject to any special requirements, let alone an effective ban.

The NRC suggested that EPA examine industrial sources associated with "petroleum hydrocarbon exposure"<sup>24</sup> and recommended that EPA explore Chemical Oxygen Demand (COD) or PAH benchmarks. EPA, however, failed to collect information about PAH-limited water bodies or the size of PAH discharges from large industrial processes, let alone the much smaller industrial stormwater sources or RTS stormwater sources.

Given EPA's inaction over the years on PAHs generally, there is no evidence in the history of the MSGP that EPA ever thought that PAHs warranted monitoring, let alone an effective ban on a specific industrial practice that could involve the release of PAHs. In fact, the lack of monitoring for PAHs over the history of the MSGP is consistent with the fact that the CWA 303(d) state water quality reports rarely identify PAHs as a cause of impairment. The lack of evidence that PAHs cause widespread water quality impacts undermines EPA's basis for seeking to limit PAH discharges through an effective ban on RTS.

The MSGP has never required monitoring for PAHs for any sector, even for certain sectors that potentially have much higher PAH releases than could occur from RTS. Rather than focusing on technology-based requirements that can effectively control PAH impacts on water quality, the 2020 Proposed MSGP seeks to categorically prohibit industrial application of a product for which PAH releases are speculative and impacts to water quality are unestablished.

#### 3. The MSGP Already Imposes Requirements on Discharges to PAH-Limited Waters.

The 2020 Proposed MSGP already contains provisions for monitoring discharges to impaired waters, which would include PAH-limited waters. Specifically, Section 4.2.4.1 provides for monitoring of discharges to impaired waters without an EPA-approved or -established TMDL, as well as for discharges to impaired waters with an EPA-approved or -established TMDL. The process outlined in Section 4.2.4.1 establishes a more nuanced and appropriate process for monitoring and addressing discharges to PAH-limited waters than the proposed eligibility requirement, as it addresses (1) PAH impacts to water quality and (2) the discharge of PAHs as

<sup>&</sup>lt;sup>22</sup> 2020 Proposed MSGP Fact Sheet at 3.

<sup>&</sup>lt;sup>23</sup> NRC Study at 33.

<sup>&</sup>lt;sup>24</sup> NRC Study at 33.

opposed to the use of RTS, which may not necessarily result in PAH discharges that impact water quality.

#### 4. EPA Has Not Established a Need for Water Quality-Based Controls.

As discussed above, EPA has only two ways to regulate a pollutant: technology-based controls and water quality-based controls. EPA's only possible legal basis for water quality-based controls for PAHs is establishing requirements based on a "reasonable potential" analysis with respect to attainment of state water quality standards. However, EPA has provided no such explanation and no supporting analysis. *See* 40 CFR § 122.44(d). EPA simply assumes that regulation of RTS is necessary to reduce PAH releases, without any quantitative analysis.

In order to establish WQBELs for a particular pollutant, EPA must evaluate whether a point-source discharge will cause an exceedance of water quality criteria *after* technology-based limits are applied. EPA regulations at 40 CFR §122.44(d) require that all effluents be characterized by the permitting authority to determine the need for WQBELs in the permit. EPA has undertaken no action to identify PAH-limited waterbodies or identify states that have PAH-specific water quality standards. And, the record clearly demonstrates that EPA has not fully considered whether or not technology-based controls can effectively limit discharges of PAHs. Absent further assessment of PAH-limited waterbodies and evaluation of the effectiveness of technology-based requirements for controlling PAH discharges, it is premature for EPA to implement water quality-based controls.

In the 2015 MSGP, EPA stated: "With respect to water quality, in the event that coal tar sealcoat use in industrial areas of a facility covered under the MSGP is found to cause or contribute to an exceedance of a water quality standards, the operator must take additional control measures to control its discharges such that they meet WQS. If it does not, EPA may require the operator to undertake such additional control measures in order to meet the permit's water quality-based effluent limit, or require coverage under an individual permit."<sup>25</sup>

This language appropriately reflects the CWA structure with respect to WQBELs and their application to RTS controls. In the 2020 Proposed MSGP, EPA appears to have ignored its previous analysis regarding water quality-based controls, with no single mention of water quality standards in the Fact Sheet. EPA should ensure that any imposition of water quality-based controls is consistent with the CWA. The simple wisdom of the longstanding MSGP framework used in the 2015 MSGP should govern in the 2020 MSGP.<sup>26</sup>

<sup>&</sup>lt;sup>25</sup> See EPA Response to Comments for 2015 Final MSGP; Docket No. EPA-HQ-OW-2012-0803-0124; Response to Comment ID 0085.001.027 at p. 75, available at https://www.regulations.gov/document?D=EPA-HQ-OW-2012-0803-0135 (last visited May 29, 2020).
<sup>26</sup> The 2015 MSGP and the 2020 Proposed MSGP both contain requirements to address impaired waters, which would include coverage of PAH-impaired waters. EPA provides no explanation as to why this provision is inadequate today to address PAHs.

# 5. Proposed PAH Requirements are Inconsistent with Other NPDES Actions.

To our knowledge, EPA has not taken action to prohibit the use of RTS in any other permit in the NPDES program, which calls into question EPA's claim that use of RTS must be banned in the 2020 Proposed MSGP. The use of RTS is not solely "an industrial activity." As discussed above, there are a number of significant issues with EPA's approach in the 2020 Proposed MSGP to controlling PAH discharges through the MSGP. First, EPA has authority to regulate PAHs, but not the use of RTS. Second, RTS is not the sole source of PAH pollution. Third, PAH requirements are not limited to receiving waters with PAH impairments.

In addition to these fundamental issues with EPA's RTS eligibility criteria and PAH monitoring requirements, EPA has reversed its 2015 position not to exclude RTS from the general permit. In the response to comments on the 2015 MSGP Proposal, EPA stated: "EPA does not have any data on the prevalence of coal tar sealcoat use at facilities covered under the MSGP, nor any data about potential water quality standards exceedances attributable to its use."<sup>27</sup> As a result, EPA rejected an RTS exclusion in the 2015 MSGP permit.

In the 2020 MSGP Proposal, EPA has not presented any data on either topic that it identified in 2015. Specifically EPA has presented no data on the prevalence of coal tar sealcoat use or on potential water quality exceedances related to RTS. EPA has failed to even acknowledge that it had completely reversed its five-year-old determination.

Most significantly, virtually all the documents in the docket now cited by EPA relating to this RTS proposal were available in 2015. EPA fails to identify any new science or any new analysis to support a reversal of its previous determination. EPA jumps from stating that it has no data upon which to draw a conclusion, to concluding that the RTS exclusion is warranted, ignoring the substantial literature pointing to the inadequacy of the underlying science.

As previously discussed in Part I.C, EPA has determined previously that parking lots are excluded from the definition of "industrial activity" associated with stormwater discharges. First, during the Phase I program development, EPA concluded that parking lots and administrative areas, even at industrial sites, do not meet the definition of "associated with industrial activity." 55 Fed. Reg. at 48010. Again, EPA confirmed its position in the "Stormwater Question & Answer II" document. Finally, EPA rejected the need to regulate RTS in 2015 in part because parking lots were excluded from the definition of "industrial activity" associated with stormwater discharges.<sup>28</sup> In the 2020 Proposed MSGP, EPA fails to explain the reversal of its position on this issue.

 <sup>&</sup>lt;sup>27</sup> 2015 EPA MSGP Response to Comments at 75, June 4, 2015 (EPA-HQ-OW-2012-0803-0135)
 <sup>28</sup> *Id.* at 75.

#### **II.** EPA's Proposed Ban Has Constitutional and Statutory Flaws.

#### A. EPA Cannot Overcome Commerce Clause Limitations.

Congress has relied upon its power to regulate commerce in delegating administrative authority under the CWA.<sup>29</sup> That power is limited, however. Particularly where an administrative interpretation "invokes the outer limits of Congress' power," courts will require "a clear indication that Congress intended that result."<sup>30</sup>

*SWANCC v. Army Corps* provides the most relevant example of a court decision protecting state and local authority against unsupported federal encroachment.<sup>31</sup> In *SWANCC*, the Supreme Court struck down CWA-based permit requirements that prohibited local governments from using abandoned gravel pits for solid waste disposal, despite approvals from local and state agencies.<sup>32</sup> In ruling that the Army Corps of Engineers had no statutory basis for refusing to issue a permit, the Court prevented "impingement of the States' traditional and primary power over land and water use" and avoided "significant constitutional and federal questions" about the authority of Congress under the Commerce Clause.<sup>33</sup>

The Court again enforced the limitations of a federal system when it ruled in *U.S. v. Lopez* that Congress did not have the authority to regulate the "mere possession" of firearms without some additional nexus that could substantially affect interstate commerce.<sup>34</sup> In that case, the Court stated: "To uphold the Government's contentions here, we would have to pile inference upon inference in a manner that would bid fair to convert congressional authority under the Commerce Clause to a general police power of the sort retained by the States."<sup>35</sup>

More recently, the Supreme Court ruled that the Commerce Clause could not support health care requirements, emphasizing the value in preserving state authority against administrative overreach: "federalism secures to citizens the liberties that derive from the diffusion of sovereign power. . . . The Framers [] ensured that powers which in the ordinary course of affairs, concern the lives, liberties, and properties of the people were held by governments more local and more accountable than a distant federal bureaucracy."<sup>36</sup>

Interpreting the CWA to authorize an effective ban on a specific product not only "invokes the outer limits" of federal authority under the Commerce Clause, it clearly exceeds them. The CWA provides no indication that Congress ever intended for EPA to use its authority to ban products and includes plenty of indications that it did not. Interpreting the CWA to authorize such

<sup>&</sup>lt;sup>29</sup> See Solid Waste Agency of N. Cook Cty. v. U.S. Army Corps of Engineers, 531 U.S. 159, 172 (2001).

<sup>&</sup>lt;sup>30</sup> *Id.* at 172.

<sup>&</sup>lt;sup>31</sup> *Id*.

<sup>&</sup>lt;sup>32</sup> *Id.* at 165.

<sup>&</sup>lt;sup>33</sup> *Id.* at 161.

<sup>&</sup>lt;sup>34</sup> 514 U.S. 549, 562 (1995).

<sup>&</sup>lt;sup>35</sup> *Id.* at 567.

<sup>&</sup>lt;sup>36</sup> Nat'l Fed'n of Indep. Bus. v. Sebelius, 567 U.S. 519, 536 (2012) (internal citations and quotations omitted).

a radical measure would violate well-established principles of federalism that limit federal power and preserve the authority of states over the "ordinary course of affairs."

# B. EPA Can Only Pursue a Product Ban Under TSCA and Only If It Meets All of the TSCA Requirements.

The CWA does not authorize EPA to ban a product. In fact, EPA only has authority to ban products under the Toxic Substances Control Act ("TSCA"). Moreover, EPA can only implement a ban under TSCA if the ban meets the high legal burden set forth in the statute and regulations thereunder. Here, EPA has not attempted to meet that burden, because it has not attempted to demonstrate that RTS should be banned under TSCA.

As amended in 2016, TSCA establishes a complex and demanding set of procedural requirements and substantive standards that EPA must meet in order to ban a product under the statute. First, EPA must determine that a chemical contained in the product's mixture is a high priority for risk evaluation.<sup>37</sup> Next, EPA must conduct a risk evaluation of that chemical, including scientific peer review, which can be a three to three-and-a-half year process.<sup>38</sup> Then, EPA must determine, based on the risk evaluation, that the chemical poses an unreasonable risk of injury to health or the environment under a condition of use.<sup>39</sup> Finally, EPA must consider the benefits of the product and the economic consequences of potential restrictions, including the availability of effective alternative products.<sup>40</sup> EPA has not taken any of these steps to the extent that would be required to justify a ban under TSCA. In particular, EPA has failed to recognize, much less account for, the impact of the proposal on the sealcoat industry.<sup>41</sup> Moreover, EPA offers no support for its contention that alternative products with equivalent performance and cost are available. In fact, it cannot do so because substantial evidence demonstrates that such alternatives are not available.<sup>42</sup>

It is worth noting that EPA had scheduled a PAH review under the TSCA 2014 Work Plan, but decided to substitute the review of benzo(a)pyrene, an individual PAH, for a study of PAH mixtures. EPA planned to initiate this review as part of the creosote assessment, but never started the creosote evaluation.<sup>43</sup> Instead, EPA moved to assess PAH mixtures as part of an Integrated Risk Information System (IRIS) review, but that was suspended in December 2018.<sup>44</sup> Given the fact that PAHs do not qualify for high priority attention under TSCA, it is unclear how EPA can justify banning PAH discharges from a single industrial application, which is what EPA is proposing in the MSGP. EPA's action is illegal and improper.

<sup>43</sup>https://www.epa.gov/sites/production/files/2015-

01/documents/tsca\_work\_plan\_chemicals\_2014\_update-final.pdf

<sup>&</sup>lt;sup>37</sup> 15 U.S.C. § 2605(b)(1).

<sup>&</sup>lt;sup>38</sup> *Id.* § 2605(b)(2).

<sup>&</sup>lt;sup>39</sup> *Id.* §§ 2605(a), (c).

<sup>&</sup>lt;sup>40</sup> *Id.* §§ 2605(c)(2)(A)-(C).

<sup>&</sup>lt;sup>41</sup> See infra, App. A, Section III.

<sup>&</sup>lt;sup>42</sup> See infra App. C.

<sup>&</sup>lt;sup>44</sup> <u>https://cfpub.epa.gov/ncea/iris2/chemicalLanding.cfm?substance\_nmbr=1033#tab-2</u>

#### III. EPA's Cost-Impact Analysis is Deficient.

EPA's cost-impact analysis addresses potential costs associated with the use of coal tarbased sealcoat alternatives and the cost of compliance with proposed COD monitoring and PAH benchmarks. However, the analysis fails entirely to recognize, much less account for, the economic impacts across the RTS value chain—from coal tar producers, to coal tar refiner and RTS manufacturers—as discussed below.

Six U.S. companies produce crude coal tar at nine metallurgical coke plants throughout the U.S. that use the "byproduct recovery" cokemaking method. These plants heat blended coal in coke chambers or ovens for 12 to 20 hours in the absence of oxygen at temperatures of about 1100 to 1150°C (2000 to 2100°F), evaporating all volatile matter from the coal mass. The recovered volatile matter is condensed and separated into liquid and gaseous fractions, known as crude coal tar and coal gas, respectively. Crude coal tar and coal gas are valuable chemicals, and their recovery is an economic necessity for these plants, accounting for approximately 25 percent of the mass and 35 percent of the value of the coal.

Tar refiners, among others, purchase crude coal tar from coke plants as critical feedstock for their production of refined tar products. One of their refined tar products is RT-12 grade, which is a critical feedstock, base tar, used in producing refined tar pavement sealer.

Finally, manufacturers of coal tar sealer purchase refined tar RT-12 grade from tar refiners and blend it with sand, clay, a proprietary emulsifier, and water to produce RTS. RTS is sold to contractors in the pavement maintenance industry.

Based on market data from 2019, the value chain at risk—from coal tar producers, to coal tar refiners and RTS manufacturers, to RTS applicators—is responsible for an estimated economic value in excess of \$100 million dollars, annually.<sup>45</sup> A number of the entities at risk are small entities, requiring further evaluation pursuant to the Regulatory Flexibility Act, which EPA has not conducted. Moreover, banning a class of products because of the potential to lead to discharges that might impair water quality is inconsistent with this Administration's regulatory policy: "to alleviate unnecessary regulatory burdens." These legal requirements are discussed in detail in the following section.

A number of comments on the 2020 Proposed MSGP have already been posted to the docket. Several of these comments submitted by small entities at risk provide a perspective on the major economic impacts that EPA's proposal would have on the small sealcoating contractors:

• "The EPA, if they effectively ban RTS in the U.S. via the Clean Water Act, will begin the end of the currently thriving asphalt maintenance industry, eliminating the many jobs within and dependent upon it. The much higher cost of having to repave un-sealcoated

<sup>&</sup>lt;sup>45</sup> Notably, sealcoating sales for 50 sealcoat contractors surveyed in 2019 totaled over \$130,000,000. FCP, "The 2019 Sealcoating 50" (June 14, 2019), *available at* <u>https://www.forconstructionpros.com/pavement-maintenance/article/21066876/pavement-magazine-announces-top-50-sealcoating-contractors-for-2019</u>.

asphalt much more often as it deteriorates twice as fast, is a burden that property owners will unnecessarily have to absorb."<sup>46</sup>

- "The proposed change will have a negative impact on my business and employees. It would take more than half of my revenue and would be shortening of the sealcoating season, especially in N.C. where we get longer rainy seasons.... Applicators who often use RTS during sealcoating projects would lose a source for income.... This decrease in sales for all pavement maintenance services would have a catastrophic effect on Carolina Striping Solutions and local businesses across the U.S.<sup>347</sup>
- "This proposal will negatively affect our business and 50+ employees will lose their job."<sup>48</sup>
- "The proposed change will have a serious negative impact on my business and employees because we use RTS exclusively in our application because it lasts longer than other sealers. Other applications must be done multiple times, nearly every year, while we apply every 3-5 years. It is our number one source of income and restrictions would likely cause a negative perception on all sealcoating products which would put our business in danger of failing".<sup>49</sup>
- "The proposed 2020 MSGP will have a negative financial impact on our small business, our employees, our customers, and the overall pavement maintenance industry. In an already frightening economic situation (due to COVID-19), the proposed 2020 MSGP would catastrophically damage the pavement maintenance industry. . . .

The vast majority of our sealcoating customers prefer Coal Tar sealer over other sealer types, because it lasts the longest, and has been proven to provide the BEST results when it comes to preserving their pavement. We have given our customers other sealer options throughout the years, and time and time again, they continue to ask specifically for Coal Tar."<sup>50</sup>

• "Any attempted ban on coal tar pavement sealers would be detrimental to our business and force us to use inferior pavement sealers that don't last through Indiana's rough weather

<sup>&</sup>lt;sup>46</sup>Comment by Decker, Jr., Decker Enterprises, Inc., DBA SealMaster - DelMarVa, Dkt. No. EPA-HQ-OW-2019-0372-0112 (April 29, 2020), *available at* 

https://www.regulations.gov/document?D=EPA-HQ-OW-2019-0372-0112.

<sup>&</sup>lt;sup>47</sup> Comment by Carolina Striping Solutions, Dkt. No. EPA-HQ-OW-2019-0372-0125 (April 30, 2020), *available at* https://www.regulations.gov/document?D=EPA-HQ-OW-2019-0372-0125

<sup>&</sup>lt;sup>48</sup> Comment by John Butler, Pavement Solutions LLC, Dkt. No. EPA-HQ-OW-2019-0372-0101 (April 21, 2020), *available at* https://www.regulations.gov/document?D=EPA-HQ-OW-2019-0372-0101.

<sup>&</sup>lt;sup>49</sup>Comment by John Hartz, Owner, Hartz Sealcoating, Dkt. No. EPA-HQ-OW-2019-0372-0105 (April 29, 2020), *available at* https://www.regulations.gov/document?D=EPA-HQ-OW-2019-0372-0105.

<sup>&</sup>lt;sup>50</sup> Comment by Chelsea Herrick, President, Daniel B. Krieg, Inc., Dkt. No. EPA-HQ-OW-2019-0372-0077 (April 7, 2020), *available at* https://www.regulations.gov/document?D=EPA-HQ-OW-2019-0372-0077.

patterns. Such issues that we would face would be a loss of revenue.... This would also force us to decrease our workforce and put hard working Hoosiers out of a job."<sup>51</sup>

• "The proposed change will have a[] negative impact on our family business and employees. Our business is a seasonal business and I envision a shortening of the seal coat season which will adversely impact on our business profits and decrease the salaries of our employees. A ban on RTS would mean we would have to decrease our employee force, possibly by 50%."<sup>52</sup>

It is very clear from the above comments that there will be a substantial adverse effect on the viability of many applicator firms. These firms are the quintessential small businesses, often family-owned, that are in grave danger of losing the business. Such businesses are more than 85% small business owned and account for thousands of firms nationwide and tens of thousands of employees. It would be unconscionable to destroy these numerous small firms, simply because EPA speculates, without justification, that RTS applications should be terminated without even opining on the expected effect on water quality.

Our Nation is struggling now to recover from what is perhaps the deepest recession since the Great Depression. Manufacturers across the board are moving to respond to the "new normal" by realigning and expanding their supply chains to afford them the greatest possible flexibility at coping with the downturn. Now is perhaps the worst time in a generation for EPA to consider imposing product restrictions on pavement sealer manufacturers and sealcoating contractors on the order of what EPA is proposing in the 2020 MSGP Proposal—restrictions that would limit their raw material options and, in the process, reduce the demand for a valuable product manufactured by their upstream, feedstock providers—the coke producers and tar refiners.

#### IV. EPA's Proposal is Inconsistent with the Regulatory Flexibility Act.

The impacts to individually impacted firms summarized in Section III above strongly suggest that the 2020 MSGP will have a significant economic impact on a substantial number of small entities, thus triggering the Regulatory Flexibility Act (RFA).<sup>53</sup> Yet EPA's *Federal Register* notice fails to address the Agency's RFA obligations.<sup>54</sup> In particular, EPA failed to properly assess the impacts on the RTS industry related to its proposed eligibility provision. In its Cost Impact Analysis, EPA states that the number of facilities affected, average cost per facility, and total cost on all applicable facilities related to the proposed eligibility limitation was "unknown" for each of those categories.<sup>55</sup> This is clearly untrue.

<sup>&</sup>lt;sup>51</sup>Comment by Jay Reece, President, Reece Seal Coating Inc., Dkt. No. EPA-HQ-OW-2019-0372-0109 (April 29, 2020), *available at* https://www.regulations.gov/document?D=EPA-HQ-OW-2019-0372-0109. <sup>52</sup> Comment by John Zarlengo Jr., Vice President of Sales, John Zarlengo Asphalt Paving Company, Dkt.

No. EPA-HQ-OW-2019-0372-0121 (April 30, 2020), *available at* https://www.regulations.gov/document?D=EPA-HQ-OW-2019-0372-0121.

<sup>&</sup>lt;sup>53</sup> See 5 U.S.C. §§ 603-604.

<sup>&</sup>lt;sup>54</sup> 85 Fed. Reg. at 12,294.

<sup>&</sup>lt;sup>55</sup> Cost Impact Analysis at 5.

Previously, EPA has asserted that NPDES permits are not "rules" subject to RFA requirements. However, in promulgating the 2008 Construction Stormwater General Permit, EPA committed to applying the RFA framework and requirements to all NPDES general permits whether or not the Agency considered them "rules" under the APA.<sup>56</sup> EPA must conduct an appropriate and complete RFA analysis for the 2020 Final MSGP, particularly for the RTS industry if it plans to finalize any eligibility restrictions related to RTS.

Further, the President issued Executive Order 13924 on May 19, 2020 focused on regulatory relief to support economic recovery.<sup>57</sup> In it, the President directs all agencies, including EPA, to "identify regulatory standards that may inhibit economic recovery and shall consider taking appropriate action, consistent with applicable law, including by issuing proposed rules as necessary, to temporarily or permanently rescind, modify, waive, or exempt persons or entities from those requirements."<sup>58</sup> This type of regulatory mandate, which has little if any environmental benefit coupled with significant economic impact, is precisely the type of action the President directed agencies to rescind or waive.

<sup>&</sup>lt;sup>56</sup> See 73 Fed. Reg. at 40,342 (July 14, 2008).

<sup>&</sup>lt;sup>57</sup> *See* Executive Order on Regulatory Relief to Support Economic Recovery (May 19, 2020), 85 Fed. Reg. 31353 (May 22, 2020).

<sup>&</sup>lt;sup>58</sup> *Id*. at § 4.

## Appendix B Science-Related Comments

#### I. Refined Coal-Tar Sealcoat (RTS) Is Not a Significant Source of PAHs in the Aquatic Environment, and Its Use Does Not Present Risks to Aquatic Life or Humans. Thus, There Is No Rational Basis for EPA's Proposed RTS Eligibility Restriction.

The Fact Sheet lays out the asserted logic behind EPA's proposed eligibility restriction as to RTS:

- Polycyclic aromatic hydrocarbons (PAHs) are toxic;
- RTS has a high level of PAHs;
- Stormwater causes PAHs to be transported from surfaces sealed with RTS into freshwater; and
- That PAH contribution from RTS is significant, and causes toxicity to aquatic life.

Each of these claims is either false or misleading. As we explain below:

- While some individual PAHs may be toxic in isolation, PAHs occur in the environment in complex mixtures and substances that are largely inert.
- RTS contains a relatively small amount of PAHs.
- Once applied in accordance with industry standards,<sup>59</sup> RTS does not readily yield PAHs, and those that are released soon after application remain bound to organic material in sediments.
- PAHs are also hydrophobic (*i.e.*, virtually insoluble in water). As a result, PAHs from RTS are not a significant source of the PAHs found in aquatic environments.
- PAHs from RTS are not a source of significant toxicity in those environments.

Thus, the proposed eligibility criterion will not produce any meaningful change in the abundance or health of aquatic species in the United States.

The proposed exclusion is based on a collection of claims leading to the assertion that releases of stormwater from RTS-treated surfaces cause aquatic toxicity and pose risk to humans. But, EPA has failed to directly link PAH releases from RTS applications to aquatic toxicity. The reasons for this lack of cause and effect relationship between RTS products and aquatic impacts are numerous and are discussed in detail below.<sup>60</sup>

<sup>&</sup>lt;sup>59</sup> ASTM D3423 requires that RTS not be applied unless there is no rainfall expected within 24 hours. ASTM standard section 2015.

<sup>&</sup>lt;sup>60</sup> It should be noted that in nearly all research conducted on this topic where the study duration exceeds 24 hours, researchers have not taken into account the impact of photolysis, a form of weathering caused by sunlight, on the PAHs contained within RTS. The transformative properties of sunlight and its ability to change the type and quantity of PAHs available at the surface of an RTS-coated parking lot has been thoroughly documented at the chemical level and in the similarity of pyrogenic PAHs from different sources in the environment.

The Administrative Procedure Act requires an agency to show a rational connection between the facts and the agency's conclusion. *FERC v. Electric Power Supply Ass'n*, 136 S. Ct. 760, 782 (2016). The best available science shows that EPA's asserted connections are unfounded. EPA's proposed ban of RTS is therefore irrational and illegal.

# A. Refined Coal Tar Sealcoat Is Not a Significant Source of Biologically Available PAHs in the Aquatic Environment.

# 1. EPA's own CWA guidance acknowledges that, in natural aquatic environments, PAHs strongly adsorb to organic materials and are not bioavailable—and, thus, require specialized risk assessment techniques.

Part 1.1.8 of the Fact Sheet notes that 16 PAHs have been designated as Priority Pollutants, and that "[s]everal PAHs have been shown to be extremely toxic to and bioaccumulate in fish and aquatic invertebrates, and are known or probable human carcinogens."<sup>61</sup> However, EPA's Priority Pollutant List, and much related characterization of PAHs, was developed in the 1970s, when scientific understanding of the environmental chemistry, toxicology, fate, and transport of the PAHs was minimal. In the decades since, our understanding of PAHs in the natural environment has improved greatly. That increased understanding has informed EPA's evaluation of PAHs in other CWA contexts. In particular, EPA guidance from 2003 and 2012 specifically aimed at evaluating PAHs in sediment clearly states—and is premised on—several key facts:

- *Outside laboratory settings, there is almost no exposure to individual PAH compounds.* This is because:
  - In the natural environment, PAHs occur in mixtures containing variable concentrations of PAH and non-PAH compounds.
  - PAHs and PAH-containing materials are hydrophobic. Thus, whether in naturally occurring or anthropogenic materials, PAHs partition strongly to the solid phase, where they have been found to be tightly bound to organic materials in complex mixtures containing dozens or hundreds of PAH and non-PAH compounds. In highly carbonized materials such as soot and coal tar pitch, PAHs can be strongly to irreversibly bound to the solid phase.
  - Reflecting their very low aqueous solubility, PAHs are very rarely detected in aqueous matrices.
- Because of these physical/chemical properties, risk assessments of exposures to PAHs associated with PAH-containing solids demonstrate that these materials are not very bioavailable or bioaccessible.<sup>62</sup>

<sup>&</sup>lt;sup>61</sup> Fact Sheet at 21.

<sup>&</sup>lt;sup>62</sup> See U.S. EPA (2003). Procedures for the Derivation of Equilibrium Partitioning Sediment Benchmarks (ESBs) for the Protection of Benthic Organisms: PAH Mixtures. EPA/600/R-02/013. November 2003; U.S. EPA (2012). Equilibrium Partitioning Sediment Benchmarks (ESBs) for the Protection of Benthic Organisms: Procedures for the Determination of the Freely Dissolved Interstitial Water Concentrations of Nonionic Organics. U.S. EPA/600/R-02/012. December 2012. See also Ruby et al. (2016). Oral Bioavailability, Bioaccessibility, and Dermal Absorption of PAHs from Soil—State of the Science. Environmental Science & Technology, 50(5), 2151-2164. doi:10.1021/acs.est.5b04110.

The virtual insolubility of PAHs is the likely reason PAHs are only rarely identified as a "cause of impairment" (COI) in Clean Water Act (CWA) Section 303(d) lists of impaired and threatened waters.<sup>63</sup> Furthermore, some COI designations based on PAHs are in remote areas, where local sources of anthropomorphic PAHs, much less RTS applications, are few. This further indicates that RTS discharges of PAHs are not significant.<sup>64</sup>

In light of these unique characteristics of PAHs in the environment, the EPA guidance regarding sediment benchmarks recommends an equilibrium partitioning (EqP) approach to developing benchmarks for PAHs in sediments that are protective of benthic organisms. This guidance on Equilibrium Partitioning Sediment Benchmarks (ESBs) also recognizes that bioavailability can vary in different sediments, resulting in variable biological effects concentrations. None of the literature cited in the Fact Sheet to support an eligibility restriction on RTS based on possible PAH releases to water bodies reflects any of EPA's guidance on developing ESB benchmarks for PAHs.

Experience since the 1970s has also led EPA to conclude, in other CWA contexts, that measurement of sediment concentrations of chemicals on the Priority Pollutants List alone is insufficient either to predict the toxicity of sediments or to identify which chemicals are causative when toxicity is observed. The problem is multi-faceted. Toxicity may be:

- related to the presence of unmeasured chemicals;
- correlated with a chemical that is not causative;
- influenced by the relative bioavailability of different sediment constituents; or
- mediated by synergism or antagonism of the chemicals or mixtures present.

To address these problems, EPA has developed guidance on conducting a Toxicity Identification Evaluation (TIE) to "characterize, identify, and confirm the causes of measured toxicity."<sup>65</sup>

Again, none of the literature cited in the Fact Sheet to support the proposed RTS eligibility restriction reflects any of EPA's TIE guidance—nor any other method of identifying toxicity or the causes of measured toxicity in sediments in the real world.

As discussed below, the proposed eligibility criterion is arbitrary and capricious, due to its failure to acknowledge and address the weight of current scientific evidence and current best practices in sediment risk assessment. This failure is particularly glaring given that the Agency's CWA guidance demonstrates that EPA does, in fact, understand the nature of PAHs in the environment and the best practices for assessing related risks.

<sup>&</sup>lt;sup>63</sup> LeHuray, A. (2014). PAHS are Rarely Causes of Impairment in U.S. Clean Water Act Section 303(D) Reports. Presentation at the 35<sup>th</sup> Annual Meeting of the Society of Environmental Toxicology and Chemistry (SETAC) Vancouver, BC November 10, 2014

<sup>&</sup>lt;sup>64</sup> Id.

<sup>&</sup>lt;sup>65</sup> US EPA (2007) Sediment Toxicity Identification Evaluation (TIE): Phases I, II, III Guidance Document. EPA/600/R-07/080. September 2007.

# 2. PAHs in coal tar sealcoat particles bond particularly strongly and are particularly not bioavailable.

EPA's development of guidance on ESBs and TIEs was grounded in EPA's recognition that, outside the laboratory, exposure to individual PAH compounds is limited by the physical-chemical characteristics of PAHs discussed above:

- PAHs occur in mixtures containing variable concentrations of PAH and non-PAH compounds;
- PAHs are hydrophobic compounds that, in highly carbonized materials such as soot and coal tar pitch, can be strongly-to-irreversibly bound to the solid phase; and
- PAHs bound in highly carbonized materials are not very available or accessible to biological systems.

These facts about PAHs generally are equally true of the PAHs in RTS.<sup>66</sup>

The tendency of PAHs to strongly-to-irreversibly bond to highly carbonized particles is illustrated in Figure 1 below:

<sup>&</sup>lt;sup>66</sup> The Fact Sheet errs in asserting that RTS "typically contains 20 to 35% coal tar pitch which is made up of 50% or more PAHs by weight (Mahler, Van Metre, Bashara, Wilson, and Johns, 2005)." Id. at 21. This presentation of PAH content is misleading. RTS is manufactured and applied according to a specific set of materials and methods specified in a series of ASTM International performance standards. The base material used to manufacture sealants is called RT-12, which is a controlled distillation fraction pulled from the coal tar pitch refinery column at a specified temperature and viscosity. ASTM D490-92(2016), Standard Specification for Road Tar, 2016. RT-12 is a UVCB (Chemical Substance of Unknown or Variable Composition, Complex Reaction Products and Biological Materials) that typically contains about 50% PAHs by weight. Undiluted RTS is a physical mixture of RT-12, emulsifier, clay, water, and sand, resulting in 5% to 8% total PAHs by weight in the product delivered to applicators, as described in the ASTM standards (ASTM D4866/D4866M-88(2017)e1, Standard Performance Specification for Coal Tar Pitch Emulsion Pavement Sealer Mix Formulations Containing Mineral Aggregates and Optional Polymeric Admixtures, 2017; ASTM D5727/D5727M-00(2017)e1, Standard Specification for Emulsified Refined Coal Tar (Mineral Colloid Type), 2017; ASTM D6945-03, Standard Specification for Emulsified Refined Coal-Tar (Ready to Use, Commercial Grade), 2013; ASTM D6946-13, Standard Specification for Emulsified Refined Coal-Tar (Driveway Sealer, Ready to Use, Primary Residential Grade), 2013). Before application, the sealant is further diluted with water and additional sand, resulting in a typical total PAH content of 3% to 6% by weight in as-applied sealant. ASTM D3423 / D3423M-84(2015)e1, Standard Practice for Application of Emulsified Coal-Tar Pitch (Mineral Colloid Type), 2015. All these ASTM standards are available at www.astm.org.





That tendency was also documented in a Department of Defense Strategic Environmental Research and Development Program (SERDP) multi-disciplinary study to better understand how the bioavailability of PAHs impacts exposure risks. The study found that:

The source of PAH contamination is the primary factor controlling the portioning behavior of PAHs from soil. PAHs that enter soil as part of a matrix that is rich in black carbon (BC), such as within soot or coal tar pitch, are much less bioavailable than PAHs that are spiked to soil in the laboratory or that enter soil within fuel oil. Mineral characteristics of the soil (e.g., type of content of clay, presence of humic acids) have much less influence on the binding of PAHs.<sup>67</sup>

The implications of relative biological inaccessibility of PAHs in coal tar pitch particles in soil also apply to PAHs contained in sediment particles, as reflected in EPA's ESB guidance for PAHs.<sup>68</sup>

<sup>&</sup>lt;sup>67</sup> Lowney, Y. W. et al. (2017). *PAH Interactions with Soil and Effects on Bioaccessibility and Bioavailability to Humans*. Department of Defense SERDP Project ER-1743.

<sup>&</sup>lt;sup>68</sup> The situation is even more complex when considering materials such as coal tar pitch particles that are present in a soil or sediment matrix that can adsorb PAHs from other sources. Hypothetically, PAH concentrations of pitch particles may increase over time as PAHs from other sources are adsorbed. From the concept of PAH partitioning and sorption, it can be inferred that PAHs adsorbed to the surface of pitch particles during weathering are more readily desorbed than PAHs that are intrinsic to the particles, which remain strongly-to-irreversibly bound. Ruby et al. 2016; Khalil et al. 2006; Xia, H., Gomez-Eyles, J. L., & Ghosh, U. (2016). Effect of PAH source materials and soil components on partitioning and dermal uptake. *Environmental Science & Technology*, *50*(7), 3444-3452. doi:10.1021/acs.est.5b06164.

# **3.** Surfaces sealed with RTS do not contribute significant amounts of PAHs to aqueous environments.

# a) Simulation studies typically focus on unrealistic experimental conditions. Mahler et al. 2005 is also deeply flawed.

The Fact Sheet relies crucially on two studies in which experimenters used hoses to attempt to simulate stormwater runoff from newly sealcoated lots: Mahler et al. 2005 and Rowe & O'Conner 2011. Neither of these studies supports the proposed ineligibility criterion.

We discuss Mahler et al. at great length below, for several reasons. On the one hand, it is a highly influential work, as it gave rise to the entire literature attempting to connect RTS to the presence of PAHs in environmental media. On the other hand, other scientists have been unable to replicate its results, and still other scientists have faulted its logic. Finally, but most importantly, correspondence by the studies' authors released in response to Freedom of Information Act (FOIA) requests reveal that the study violated fundamental principles of scientific integrity and is unreliable.

We then address Rowe & O'Conner, which shows very low levels of PAHs from RTSsealed surfaces, even under unrealistic experimental conditions. Finally, we discuss McIntyre et al. 2016, which explains why, in light of the characteristics of PAHs discussed above, runoff from RTS-sealed surfaces in the real world does not produce PAH contamination of surface waters.

#### (1) Mahler et al. 2005

The authors of the Mahler paper claimed to have shown that RTS "could indeed be the dominant source of PAHs to watersheds with residential and commercial development." The authors based this claim on a forensic analysis. First, they collected samples of suspended solid particles from "simulated runoff" from RTS-sealed parking lots in the Austin, Texas area. Then, they analyzed those particles and plotted the ratios of selected individual PAHs (for example, the concentration of fluoranthene against the concentration of pyrene). They concluded that those ratios were the diagnostic "signature" of RTS. Next, they compared those ratios with ratios derived from 20 samples they claimed were representative of PAH concentrations in Austin-area streams. They concluded that the overlap between the plots of those two ratios showed that RTS runoff "could be dominating PAH loadings" to urban sediments.

However, in a glaring violation of the scientific method, the 20 samples they claimed to represent PAHs in Austin-area streams were not even collected as part of the run-off study. Most did not even come from the Austin area. Rather, the authors (two U.S. Geological Survey scientists) searched their files until they found stream sediments with similar PAH "signatures."

In Figure 2 below, the red squares show the locations where the simulated run-off samples were collected.<sup>69</sup>



Figure 1. Location of parking lots for sampling of simulated runoff, Austin, Texas, 2003.

Figure 2. Location of parking lots for sampling of simulated runoff, Austin, Texas, 2003.

At the bottom edge of Figure 2 is a stream called Williamson Creek. Eight of the 20 samples used to represent Austin's sediments were collected over time from a single Williamson Creek sampling location shown in Figure 3 below.<sup>70,71</sup> As can be seen from Figure 3 below, Williamson Creek is in a different streamshed than the run-off study locations shown in Figure 2 above.

<sup>&</sup>lt;sup>69</sup> Mahler, B. J. et al. (2004 [revised 2007]). Concentrations of polycyclic aromatic hydrocarbons (PAHs) and major and trace elements in simulated rainfall runoff from parking lots, Austin, Texas, 2003 (version 3): U.S. Geological Survey Open-File Report 2004–1208, 87 p. [Online only].

<sup>&</sup>lt;sup>70</sup> Sample locations identified in a letter from William G. Wilber (USGS) to Anne LeHuray (PCTC) dated July 22, 2013.

<sup>&</sup>lt;sup>71</sup> Figure from Mahler et al. 2004 [revised 2007].



Figure 1. Sampling sites and associated watersheds, Austin, Texas.

Figure 3. Sampling sites and associated watersheds, Austin, Texas.

To complete the picture, 12 of the 20 samples used to represent sediment PAHs in Austin streams were collected from three locations in three streams in Fort Worth, about 200 miles from Austin.<sup>72</sup> The Fort Worth locations are shown in Figure 4 below.<sup>73</sup>

<sup>&</sup>lt;sup>72</sup> Letter from Wilber to LeHuray, 2013.

<sup>&</sup>lt;sup>73</sup> Van Metre, P. C., et al. (2003). Occurrence, trends, and sources in particle-associated contaminants in selected streams and lakes in Fort Worth, Texas. USGS Water Investigations Report 03-4169.



Figure 4. Locations of suspended sediment sampling sites and lakes where sediment cores collected, Fort Worth, Texas.

Why did the authors choose those particular locations to derive data supposedly representative of "urban sediment" in Austin? Email correspondence obtained via FOIA reveals that the authors chose the particular stream sediment samples precisely because they matched the claimed PAH signature of RTS and thus "confirmed" the authors' hypothesis. When a colleague asked: "Could you guys fill me in again on what PAH ratios had looked kind of hopeful to differentiate PAH sources?" one of the authors wrote in response:

When the Williamson Creek suspended sediment data was plotted on the same graph, they tended to group with the sealed parking lots as opposed to with the unsealed (asphalt pavement or cement) parking lots. Suspended sediment data from three small urban watersheds in Fort Worth were similar.

The scientific way to test the hypothesis that RTS runoff contributes to PAH concentrations in streams would have been to collect sediments from streams in the vicinity of the parking lots used to generate the simulated runoff—and then to compare them. Instead, however, the USGS scientists found sediment samples that had the matching ratios, and then claimed that these supported their hypothesis. This sort of backward reasoning from conclusions to data is circular logic, not science. It violates basic principles of scientific integrity. Not surprisingly, other scientists with similar expertise have been unable to reproduce the same diagnostic double ratio plots presented in the USGS scientists' article:

In conclusion, we were unable to replicate the computations and identify values from cited sources for a number of the data points represented in Figures 4–6 of Mahler et al. (2005). With regard to the PAH ratio analysis, we could not identify the source of the values presented for stream sediment samples, and the values that we could identify from the City of Austin appear to contradict the interpretation developed by the authors. With regard to the mass balance analysis, we could not identify the source for values from one watershed, the values presented for the other watersheds do not appear to match those from the cited sources, and the previously published values suggest the relative contribution of PAHs from parking lot sources is substantially less than the "majority" source suggested by the authors. Because these uncertainties relate to the two lines of argument indicated to support a conclusion that parking lot sealcoat could be a dominant source of PAHs in urban streams, clarification is important for understanding the strength of the conclusions of this paper.<sup>74</sup>

Scientists who have collected actual stream sediment samples from areas near where Mahler et al.'s simulated runoff samples were collected have found that their PAH ratios did not match. For example, Figure 5 below is drawn using the data from DeMott et al. 2010:<sup>75</sup>

<sup>&</sup>lt;sup>74</sup> DeMott RP, Gauthier TD. 2006. Comment on "Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons." Environ. Sci. Technol. 40(11):3657–3658. *See generally* O'Reilly 2020 (Appendix E) at 3-5.

<sup>&</sup>lt;sup>75</sup> DeMott, R.P. et al. (2010). PAHs in Austin Sediments after a Ban on Pavement Sealers. *Environmental Forensics*, 11:4, 372-382. https://doi.org/10.1080/15275922.2010.526520


Figure 5. PAH fingerprinting shows Austin sediments do not match coal tar signature. CT Park Lot and CT Test Plot data from Mahler et al. 2005. All other data from DeMott et al. 2010.

Scoggins et al. (2007) attempted to relate PAH concentrations in Austin stream sediments to RTS-coated parking lots following the method of Mahler *et al.* (2005),<sup>76</sup> using PAH diagnostic ratio comparisons and also using spatial relationships. They were unsuccessful, in part due to another intrinsic feature of PAHs that undermines the sort of forensics that Mahler *et al.* attempted: PAH ratios in environmental media are as much a function of weathering<sup>77</sup> as they are of source,

 <sup>&</sup>lt;sup>76</sup> Mahler, B. J. et al. (2005). Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons. *Environmental Science & Technology*, 39(15), 5560 - 5566. https://doi.org/10.1021/es0501565

<sup>&</sup>lt;sup>77</sup> Photolysis—transformation by sunlight—is a particularly important weathering process for PAHs. PAHs with four or more rings, such as benzo(a)pyrene, are generally expected to be resistant to biodegradation (Valerio et al., 1990. Intern J Environ Chem 38: 343-9). However, benzo(a)pyrene is susceptible to direct photolysis on soil surfaces exposed to sunlight, with a measured half-life of 0.48 hours in water following exposure to mid-summer sunlight. Half-lives of benzo(a)pyrene adsorbed onto clean glass-fiber filters and two dust loaded filters (0.23 and 0.69 mg/sq cm) and exposed to a UV lamp were 37, 199, and 428 minutes, respectively (Picel et al., 1985 Polynucl Aromat Hydrocarbons. Cooke, Dennis, eds. Battelle Press. 8: 1013-28). The photolysis half-life of benzo(a)pyrene on spruce needle surfaces exposed to full sunlight in Munich, Germany in July 2001 was 33 hours (Niu et al., 2003. Environ Pollut 123: 39-45). Dibenz(a,h)anthracene absorbs strongly at wavelengths >290 nm (Valerio et al., 1990) and is therefore expected to be susceptible to direct photolysis by sunlight. The photolysis of dibenz(a,h)anthracene sorbed to the surface of spruce needles follows first-order kinetics with a half-life of 15 hours under sunlight irradiation(Picel et al., 1985); therefore, photolysis on soil surfaces exposed to sunlight may be an important fate process (Niu et al., 2003).

and PAHs from many diverse sources tend to have increasingly similar diagnostic ratios as they age. The Scroggins et al. report states as follows:

We attempted to identify the sources of PAH in the sediments of our study streams using ratio methods, but we were unsuccessful and found no significant clustering of field data with known source data.... Our inability to associate PAH contamination in our study streams with coal-tar sealant might have been because we analyzed only the 16 EPA priority PAHs in field sediments or because extensive weathering and mixing with other materials occurs as the coal-tar sealant abrades and moves from parking lots to stream systems.

We attempted to explain the magnitude of PAH contamination at the downstream study sites with spatial data. Neither total area of sealed parking lot nor its proximity to sampling locations were significantly correlated with PAH concentrations in the sediments at the downstream sites. A complex mix of age of sealant applied, amount of traffic on lots, local rainfall patterns, flow paths, local stream hydrology, and sediment deposition patterns probably contributes to high variability in the movement of PAHs from parking lots to stream sediments.<sup>78</sup>

The inability to reproduce the relationship between PAHs in sediments in Austin stream beds and in RTS was also reported by Environ (2006).<sup>79</sup>

#### (2) Rowe & O'Conner 2011

In the Rowe and O'Conner study, EPA scientists conducted a study of runoff from paved surfaces (with no traffic) sealed with RTS and asphalt-based sealant (ABS), with an unsealed lot as a control.<sup>80</sup> Runoff from simulated rain was collected over 30 days after sealcoat application and again after six months. The results, depicted in Figure 6 below, show that the samples collected within the first week after application contained the highest—albeit declining— concentrations of Total PAHs. By the 30<sup>th</sup> day of the study, concentrations in runoff appear to have reached a steady state, with approximately the same concentrations for each surface on day 160 as day 30. Total PAH concentrations were highest for the RTS-coated surface, but the highest Total PAH concentration for that surface was less than 500  $\mu$ g/L (about 0.5 ppm), and the highest concentration for benzo(a)pyrene was 2.08  $\mu$ g/L (~2.1 ppb), rapidly dropping to less than 0.1  $\mu$ g/L, which is less than the drinking water standard for benzo(a)pyrene (0.2  $\mu$ g/L, or 0.2 ppb). The

<sup>&</sup>lt;sup>78</sup> Scoggins et al. 2007. Occurrence of polycyclic aromatic hydrocarbons below coal-tar-sealed parking lots and effects on stream benthic macroinvertebrate communities. J North Amer Benthol Soc 26:694–707. Scoggins et al. go on to imply that the failure to reproduce the Mahler et al. finding was probably just a matter of not having the correct data. The possibility that RTS was not the primary source of PAHs in Austin-area streams was not considered as an explanation of the data.

<sup>&</sup>lt;sup>79</sup> Environ (2006). Polycyclic Aromatic Hydrocarbon (PAH) Characteristics for Sediments Collected from Creeks and Streams in Austin, Texas. Report prepared for the Pavement Coatings Technology Center. 63 p. Available at https://www.scribd.com/document/343808345/Polycyclic-Aromatic-Hydrocarbon-PAH-Characteristics-for-Sediments-Collected-from-Creeks-and-Streams-in-Austin-Texas

<sup>&</sup>lt;sup>80</sup> Rowe, AA and O'Connor, TP 2011. Assessment of Water Quality of Runoff from Sealed Asphalt Surfaces. EPA/600/R-10/178

finding—that initial flushing with water of a surface recently coated with a coal tar-based product is rapidly followed by stabilization at concentrations that meet drinking water standards—is consistent with the findings in drinking water pipes lined with coal tar enamel in The Netherlands.<sup>81</sup>



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).

Figure 6. The sum of PAH ( $\Box$ PAH) in runoff for the five sampling events of full-scale study ( $\Box$ PAH=17)

Several other findings from Rowe & O'Conner 2011 are relevant for assessing environmental impacts of RTS. First, the study report indicates that its results reflect only chemical analyses of "dissolved" load in samples of runoff.<sup>82</sup> Second, in the 2020 Proposed MSGP Fact Sheet, EPA requested comment on the correlation between PAHs and chemical oxygen demand (COD; Request for Comment 20). Values for COD and another water quality parameter, total organic carbon (TOC), are described in Rowe & O'Conner 2011 as typical of values found in urban runoff. Comparing the bar chart in Figure 7 below with the total PAH graphs in Figure 6 above, it is evident that, in this study, PAH concentrations attributable to RTS did not correlate with COD. Additionally, RTS is not associated with elevated levels of either COD or TOC.

<sup>&</sup>lt;sup>81</sup> Blokker, E. J. M., et al. (2013). Health Implications of PAH Release from Coated Cast Iron Drinking Water Distribution Systems in the Netherlands. *Environmental Health Perspectives*, 30. doi:http://dx.doi.org/10.1289/ehp.1205220

<sup>&</sup>lt;sup>82</sup> The focus on PAHs in samples of runoff that had been filtered to remove most suspended solids can be inferred from the statement on p. 29 that "[t]his study indicates that there is increased risk in the period immediately after sealant curing when the PAHs may not be associated with sediments."



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

Figure 7. Total organic carbon and chemical oxygen demand concentration in runoff for full-scale study.

#### (3) McIntyre et al. 2016

The Fact Sheet acknowledges that "research studying the particular effects of coal-tar sealcoat in runoff in controlled laboratory tests may overestimate potential adverse effects in the field (Driscoll et al, 2019)."<sup>83</sup> Indeed, the crucial shortcoming of Rowe & O'Conner 2011 is its apparent focus on the possible impacts of sealcoating on unnaturally isolated clean water. The study by McIntyre et al.<sup>84</sup> was expressly designed to study the toxicity of runoff from RTS-coated surfaces before and after the runoff encounters a mix of soil-organic matter materials—as occurs

<sup>&</sup>lt;sup>83</sup> Fact Sheet at 23–24.

<sup>&</sup>lt;sup>84</sup> McIntyre et al. 2016. Soil bioretention protects juvenile salmon and their prey from the toxic impacts of urban stormwater runoff. Chemosphere 132:213–219.

in the real world. The study used an engineered soil mix—called a "bioretention system"—that mimics organic-rich natural soils.

For the McIntyre et al. study, whole runoff samples (that is, waterincluding suspended particulates) were collected. The terms "unfiltered" and "filtered" refer, respectively, to samples that were or were not routed through columns containing the bioretention mix. To illustrate, in the photograph (Figure 8) below,<sup>85</sup> the bottle containing clearer water has not been passed through the bioretention system and is, in the language of the study, "unfiltered," whereas the bottle containing brown water has passed through the soil mix and is, therefore, called "filtered."



Figure 8. Samples of "unfiltered" (clearer liquid) runoff and runoff "filtered" (brownish liquid) through a sand-organic matter bioretention system. Photo from a presentation by J. McIntyre. Used with permission.

Similar to the EPA runoff study, the McIntyre et al. study reported that total PAH concentrations in the runoff decreased over four sampling events, conducted about two hours after the sealcoat was applied and 7, 13, and 207 days after application (see Figure 9 below). The maximum total PAH concentration<sup>86</sup> was about 1,300  $\mu$ g/L (about 1.3 ppm) in unfiltered runoff on Day 0, declining to a plateau at about 100  $\mu$ g/L (about 0.1 ppm) on days 7, 13, and 207. After

<sup>&</sup>lt;sup>85</sup> Figures 8 and 9 are from a presentation by J. McIntyre and are used with her permission.

<sup>&</sup>lt;sup>86</sup> The total concentration of 19 PAHs plus 6 alkyl-PAHs.

filtration in organic-rich simulated soil, however, PAH concentrations were less than  $10 \,\mu g/L (0.01 \, ppm)$  in all samples.

McIntyre et al.'s finding that PAHs in runoff are reduced by virtually 100% upon contact with an organic-rich soil matrix (the study mix was about 60% sand and 40% organic material) is consistent with the strong partitioning of PAHs to the solid phase and virtual insolubility (discussed in Section II.A of this Appendix) that EPA recognized in its guidance on developing ESBs.



Figure 9. Sum of measureds PAH concentrations in unfiltered, filtered (through a simulated soil bioretention system), and control runoff.

In addition to demobilizing the PAHs, filtration through the sand-organic material soil mixture reduced the toxicity of the runoff by over 90%. Toxicity reduction in the real environment comports with EPA's guidance that the causes of sediment toxicity should be identified using TIE methods, and supports the conclusion that toxicity testing of runoff should reflect conditions representative of the real world.<sup>87</sup>

## b) Sediment PAH concentrations have not declined as a result of RTS bans by local and state governments.

(1) Austin, Texas

Austin was the first community to ban RTS, effective January 1, 2006. As noted in the Fact Sheet, a study that compared sediment PAH concentrations in Austin waterways before and

<sup>&</sup>lt;sup>87</sup> Kane Driscoll et al. 2020 (Appendix D to these comments).

after the ban on the use of RTS went into effect found no significant difference.<sup>88</sup> In a 2014 paper, Mahler et al. 2005 reported a decline in the PAH concentrations in Lady Bird Lake in central Austin.<sup>89</sup> Post-publication peer reviewers have, however, questioned the history and statistical analysis onwhich that paper relied, as well as the attribution of the source of PAHs to RTS.<sup>90,91,92</sup>

Importantly, Mahler and Van Metre have consistently failed to note that Lady Bird Lake (formerly known as Town Lake) was constructed in the late 1950s as a source of cooling water for the Holly Street power plant, a fossil fuel-powered electrical generation plant (see Figure 10 below). After a history of fuel oil spills and fires, the City of Austin closed the plant in 2007, the year after the RTS ban went into effect. Possible impacts on PAHs concentrations in lake sediments from elimination of fuel oil spills and fires have never been studied, but they present a much more likely explanation for any decrease of PAH concentrations between 2006 and 2014.

<sup>&</sup>lt;sup>88</sup> DeMott et al. 2010. The sampling events occurred in October 2005 and April 2008.

<sup>&</sup>lt;sup>89</sup> Van Metre, P. and Mahler, B. 2014. PAH concentrations in lake sediment decline following ban on coal tar-based pavement sealants in Austin, Texas. Environ. Sci Technol., 48, pp. 7222-7228.

<sup>&</sup>lt;sup>90</sup> DeMott, R.P.; Gauthier,T.D. (2014) Comment on "PAH Concentrations in Lake Sediment Decline Following Ban on Coal-Tar-Based Pavement Sealants in Austin, Texas." *Environ. Sci. Technol.* 48 (23), pp 14061–14062 http://dx.doi.org/10.1021/es5046088.

<sup>&</sup>lt;sup>91</sup> O'Reilly, K. (2014) Comment on Van Metre and Mahler 2014: "PAH Concentrations in Lake Sediment Decline Following Ban on Coal-Tar-Based Pavement Sealants in Austin, Texas." Available at <u>https://www.scribd.com/document/343814884/O-Reilly-K-2014-Comment-on-Van-Metre-and-Mahler-2014-PAH-Concentrations-in-Lake-Sediment-Decline-Following-Ban-on-Coal-Tar-Based-Pavement-Seala</u>.

<sup>&</sup>lt;sup>92</sup> LeHuray, AP 2016. PPPR of Van Metre and Mahler (2014). https://pubpeer.com/ publications/DEC6835FF61E589EB95C8597944A7F



Source: Austin Chronicle.

Figure 10. Aerial photograph of the Holly Street power plant adjacent to the plant's cooling pond, called Lady Bird Lake (formerly known as Town Lake). City of Austin, Texas.

The City of Austin issued an analysis of PAH concentrations in its monitoring data in 2012.<sup>93</sup> DeMott and Gauthier summarized this analysis as follows:

Researchers from the City of Austin (COA) found no significant difference in  $\Sigma$ PAH16 concentrations in stream sediments collected from 50 of Austin's largest watersheds when grouped by sample date with date ranges of 1996–1999, 2000–2002, 2003–2005, 2006–2008, and 2009–2010. Only when 3-ringed and 4-ringed PAHs were considered separately, were significant decreases reported from the 1996–1999 time frame to more recent time intervals. Thus, any apparent decrease was found to have started years before the ban and there has been no significant change in PAH levels since the ban.<sup>94</sup>

<sup>&</sup>lt;sup>93</sup> Richter, A. Monitoring Polycyclic Aromatic Hydrocarbon Concentrations in Austin, TX, After the Coal Tar Sealant Ban; City of Austin, Watershed Protection Department, Environmental Resource Management Division. SR-12–06. March, 2012.

<sup>&</sup>lt;sup>94</sup> DeMott and Gauthier, 2014. ES&T 48:14061-14062. http://dx.doi.org/10.1021/es5046088

#### (2) District of Columbia

Following Austin's lead, the District of Columbia banned the use of RTS effective July 1, 2009. A study conducted afterward found no difference in sediment PAH concentrations at the sampling locations between 2000 and 2015.<sup>95</sup>

#### c) The scientific consensus is that the great majority of PAHs in the environment come from atmospheric deposition from combustion sources.

Characterizing sources of PAHs in the environment has been an active field of research for many years.<sup>96</sup> There is widespread consensus that, as noted in Appendix E, "combustion processes are responsible for the vast majority of the PAHs that enter the environment."<sup>97</sup>

Efforts to evaluate the contributions of PAH sources—including earlier work by Mahler and Van Metre—have consistently identified atmospheric deposition as a significant source to stormwater, soils, paved areas, and sediments in most urban environments.<sup>98</sup> Specifically, the higher molecular-weight PAHs typical of combustion-derived particulate matter—consistent with motor exhaust, coal combustion products, or wood smoke—have been found to dominate PAH profiles in sediments that are impacted by "urban background" sources.<sup>99</sup> A 2007 EPA review concluded: "Atmospheric deposition of pollutants, including PAHs, is recognized as a significant contributor in many locations to water quality problems. PAH are organic compounds primarily formed from the incomplete combustion of organic materials, such as coal and wood."<sup>100</sup>

A number of studies have demonstrated a link between atmospheric emission sources and PAHs in urban environments. Evaluations of PAH chemistry in sediments from lakes, creeks, and reservoirs from across the United States report temporal links between changes in PAH concentrations and increased automobile use and vehicle emissions.<sup>101</sup> In the upper Midwest, the mass and chemistry of PAHs in lake sediment could be linked to specific atmospheric sources associated with activities such as steel production and motor vehicle use.<sup>102</sup> Automotive emissions have been shown to be a major source of particulate PAHs in aquatic systems in the Los Angeles basin and San Francisco Bay area.<sup>103</sup> The dominance of coal-fired power plants in the eastern

<sup>&</sup>lt;sup>95</sup> Pinkney, A. E. et al. (2019). Trends in Liver and Skin Tumor Prevalence in Brown Bullhead (*Ameiurus nebulosus*) from the Anacostia River, Washington, DC, and Nearby Waters. *Toxicologic Pathology*, 47(2), 174-189. doi:10.1177/0192623318823150.

<sup>&</sup>lt;sup>96</sup> Blumer and Youngblood 1975; Youngblood and Blumer 1975.

<sup>&</sup>lt;sup>97</sup> See also O'Reilly 2020 (Appendix E) at 1-2.

<sup>&</sup>lt;sup>98</sup> Hwang and Foster 2006; Li et al. 2003; Mastral and Callén 2000; Simcik et al. 1999; Stein et al. 2006; Su et al. 2000; Van Metre et al. 2000; U.S. EPA 2007; Yunker et al. 2002.

<sup>&</sup>lt;sup>99</sup> Stout et al. 2004.

<sup>&</sup>lt;sup>100</sup> U.S. EPA 2007.

<sup>&</sup>lt;sup>101</sup> Simcik et al. 1999; Stein et al. 2006; Su et al. 2000; Van Metre et al. 2000.

<sup>&</sup>lt;sup>102</sup> Su et al. 2000; Simcik et al. 1999.

<sup>&</sup>lt;sup>103</sup> Stein et al. 2006; Tsai et al. 2002.

United States and gas-fired power plants in the west is a potential explanation for the regional differences in sediment PAH concentrations.<sup>104</sup>

Two studies found combustion sources to be major contributors of PAHs in urban watersheds. An investigation led by the New York Academy of Sciences (NYAS) found that airbased emissions accounted for about 98% of the PAHs released to the environment in the New York–New Jersey Harbor watershed, with residential wood smoke being the source of about 35% of PAHs in harbor sediments. Only about 1.5% of PAHs were released directly to the land, with the remaining releases linked to water-based sources.<sup>105</sup> PCTC/COETF believe that the PAH emissions estimates and usage assumptions used for some of the sources, including those used for RTS, significantly overestimate the extent of RTS use in the New York–New Jersey harbor drainage area. Even with likely overestimation, however, NYAS concluded that PAHs from RTS contributed less than 1% of the total PAHs in harbor sediments.

Washington State's Department of Ecology (WDOE) conducted a similar study of the sources of PAHs in Puget Sound sediments. The methods used were similar to the New York–New Jersey harbor study. As with NYAS, WDOE found about one third of sediment PAHs were the result of wood smoke, and less than 1% were attributed to RTS.<sup>106</sup>

### d) It is difficult to distinguish the PAH "signature" of RTS from other combustion sources of PAHs in the environment.

Among scientists whose research is focused on source apportionment, it is generally recognized that it is difficult to distinguish PAHs from widely diverse combustion sources (that is, pyrogenic PAHs), because PAH chemical profiles from those sources are so similar. Zou et al. describe the issue this way:

Different PAH sources may exhibit relatively stable and exclusive PAH fingerprints. This is the basis of fingerprints-based PAH source identification. For example, combustion emissions have significantly higher fractions of 4-ring and above PAHs. In contrast, petroleum products (gasoline and diesel) have very high fraction of Nap, minor fractions of 3-ring PAHs, but very low fractions of other PAHs. . . . It seems reasonable to distinguish PAH sources by these differences. However, the picture is getting fuzzier when taking into account a big variety of PAH sources. For example, the PAH fingerprints from low rank coals (lignite A), some of the wood combustion, coal combustion soot (residential), coal-tar, and

<sup>&</sup>lt;sup>104</sup> Mahler et al. 2009.

<sup>&</sup>lt;sup>105</sup> Valle, S., Panero, M. A., and Shor, L. 2007. Pollution Prevention and Management Strategies for Polycyclic Aromatic Hydrocarbons in the New York/New Jersey Harbor. NYAS. 170p.s.

<sup>&</sup>lt;sup>106</sup> WDOE 2011. Control of Toxic Chemicals in Puget Sound: Assessment of Selected Toxic Chemicals in the Puget Sound Basin, 2007-2011. Ecology Publication No. 11-03055.

some of the tunnel air particles are highly similar to each other.... The similarities between multiple PAH sources make the differentiation difficult.<sup>107</sup>

Indeed, one of the flaws in the line of research conducted by Mahler and Van Metre is that they regularly attribute to RTS all environmental PAHs exhibiting the widespread signature of pyrogenic PAHs—particularly weathered pyrogenic PAHs—resulting in the mistaken conclusion that RTS is the source material for a substantial fraction of the pyrogenic PAHs in the environment.<sup>108</sup>

Zou et al. subjected PAHs in sediment collected from the Peoria Pool of the Illinois River to a Baysian chemical mass balance (CMB) analysis, involving PAH chemical signatures ("fingerprints") from 138 different possible sources of PAHs. Their results indicated that PAHs from RTS contributed no more than a few percent of the total.<sup>109</sup>

#### e) Modeling efforts to apportion PAHs among sources are unreliable, as they rely on a PAH "signature" for RTS that was not generated for that purpose and has never been validated.

The difficulty in distinguishing between sources of PAHs based on their chemical signatures or fingerprints is complicated by the fact that many publications on the topic have relied on a "signature" for RTS that was never intended for this purpose and has never been validated. The Fact Sheet frankly acknowledges this problem:

[T]here has been some acknowledgement that the variability of PAH concentrations in different sources is a challenge for all source apportionment models because these models assume PAH source compositions are relatively constant, even though source composition can change between the source and where the concentration measurement is taken (the receptor) (Norris and Henry, 2019). A recent letter to the editor has raised questions on the validity of the source profiles used in some source apportionment studies (O'Reilly and Edwards, 2019). A recent paper noted the challenges with PAH source apportionment to coal-tar sealcoat given the variety of PAH sources in the environment (Zou, Wang, and Christensen, 2015).

As discussed in detail in Appendix E to these comments,<sup>110</sup> receptor models are mathematical procedures for resolving one or more of the following parameters in a mixed chemical system: 1) the number of sources, 2) their chemical characteristics, and 3) the relative contribution of each source in environmental samples. As with use of any mathematical model, understanding the assumptions that underlie the model is critical to interpreting the model results. Key assumptions of least squares-based receptor models, such asEPA's CMB model which was configured by Van Metre and Mahler, are that:

<sup>&</sup>lt;sup>107</sup> Zou, Y. et al. 2015. Problems in the fingerprints based polycyclic aromatic hydrocarbons source apportionment analysis and a practical solution. *Environmental Pollution*, 205, 394-402.: http://dx.doi.org/10.1016/j.envpol.2015.05.029

<sup>&</sup>lt;sup>108</sup> See O'Reilly 2020 (Appendix E) at 5-6.

<sup>&</sup>lt;sup>109</sup> Zou et al. 2015

<sup>&</sup>lt;sup>110</sup> See O'Reilly 2020 (Appendix E) at 5.

- 1. All potential sources have been identified;
- 2. Source profiles are known and stable;
- 3. The number of sources is less than the number of fitting species;
- 4. Source profiles are linearly independent of each other; and
- 5. Measurement uncertainties are random, uncorrelated, and normally distributed.

As stated in the CMB guidance manual,<sup>111</sup> these assumptions are fairly restrictive. While some deviations can be tolerated, they do raise some critical issues that should be considered in interpreting the results. There is an inherent conflict between the first assumption, which seeks to broaden the number of individual source types considered, and the third, which highlights the models' inability to accurately allocate among similar source profiles. Including all important sources is critical, as the model can assign the contribution of missing sources to others used as inputs. The issue of collinearity of compound concentrations raised in the fourth assumption can be a particular challenge with pyrogenic PAHs, because different sources can have generally similar profiles.<sup>112</sup>

The ubiquitous nature of PAHs and the wide range of historical and ongoing combustion sources make it impossible to include all individual sources. As a result, source types are often grouped,<sup>113</sup> although grouping requires selection of a chemical profile that adequately represents the group.<sup>114</sup> PAH profiles are not unique, due to both intersource similarities and intrasource variability. Even at the point of emission or release, PAH chemistry varies with fuel type, oxygen levels, and combustion temperature. Furthermore, seasonal variations in emissions and atmospheric processes challenge the assumption of stability for atmospheric sources.<sup>115</sup> Because both particle-specific PAH ratios and deposition rates differ by particle size, as well as because of photolytic reactions, atmospheric PAH profiles of an individual source shift with distance from an emission source.

Most of the source profiles used by Van Metre and Mahler (2010) and others (Baldwin et al. 2017; Norris and Henry 2019) were initially published in Li et al. (2003) and have not been validated as accurately representing the sources claimed.<sup>116</sup> The purpose of Li's paper was to demonstrate how a receptor model could be applied at a specific site, not to generate profiles that should or could be generally applicable in all locations. Li used an untested multistep approach to combine multiple published data types in an attempt to construct PAH source profiles. Much of the underlying data were not originally published in a way that provided accurate information on PAH profiles. Because the published studies reported analyses of different combinations of PAHs, the data sources for each profile varied between profile and PAHs. Such an approach loses information on the relative concentrations among PAHs that is a critical component of forensic analysis. The median coefficient of variation (standard deviation/mean) of individual PAHs in

<sup>&</sup>lt;sup>111</sup> US EPA 2004. CMB 8.2 users' manual. EPA-452/R-04-011.

<sup>&</sup>lt;sup>112</sup> O'Reilly et al. 2012.

<sup>&</sup>lt;sup>113</sup> E.g., Li et al. 2003.

<sup>&</sup>lt;sup>114</sup> Galarneau 2008.

<sup>&</sup>lt;sup>115</sup> Khairy and Lohmann 2013.

<sup>&</sup>lt;sup>116</sup> O'Reilly et al. 2014, 2015, 2020.

Li's profiles is about 80%, suggesting that the final profiles may not represent any actual source. An Environment and Climate Change Canada researcher questioned the use of such source profiles without sufficient validation.<sup>117</sup> As noted by Li,

[A] definitive signature of a combustion process may not exist due to the complexity of the combustion process. Emissions of PAHs depend on numerous factors which may vary significantly even during a single combustion process. In addition, sampling methods differ, introducing additional differences among published source signatures.

Receptor models assume that source concentration profiles are stable and do not change over time. This is not true with PAHs. "Weathering" is a term used to describe a change in a chemical mixture due to its exposure to the environment. Volatilization, photoreactivity, and biological degradation cause PAHs to change concentrations at different rates, resulting in shifts in the chemical profile.<sup>118</sup> Li's source profiles were generated from the analysis of unweathered samples collected at the point of emission, while the parking-lot dust samples Van Metre and Mahler (2010) used to represent sealer source profiles had been exposed to weathering in the environment for an unknown amount of time. As described in O'Reilly et al. (2012), the use of weathered sealer samples and unweathered emission-source samples as CMB inputs may have skewed the output toward a higher estimated sealer contribution. If the sealer-receptor model input is replaced with the PAH profile of a fresh sealer sample, its source contribution drops to a few percent.<sup>119</sup>

The PAH profile used by Van Metre and Mahler to represent sealer is not unique to weathered sealer and may instead merely represent urban background.<sup>120</sup> The profiles identified as "sealcoat" are also essentially indistinguishable from urban soil, road runoff, or roof dust.<sup>121</sup> Van Metre and Mahler (2003) presented data for seven PAHs from 22 samples collected from an Austin, Texas stream, rooftop runoff, and roadside dust. The median correlation between profiles was r=0.99 and the lowest was r=0.91. Correlations between the nine roof-dust samples and the coal-tar sealcoat profiles used by Van Metre and Mahler (2003) and Norris and Henry (2019) ranged from r=0.93 to 0.99. These findings suggest that a receptor model could not distinguish between the contributions of these sources.<sup>122</sup>

<sup>&</sup>lt;sup>117</sup> Galarneau, E. (2008). Source specificity and atmospheric processing of airborne PAHs: Implications for source apportionment. *Atmospheric Environment*, *42*(35), 8139-8149.

<sup>&</sup>lt;sup>118</sup> See O'Reilly 2020 (Appendix E) at 7.

<sup>&</sup>lt;sup>119</sup> O'Reilly et al. 2014; Zou et al. 2015.

<sup>&</sup>lt;sup>120</sup> O'Reilly 2020 at 7-8.

<sup>&</sup>lt;sup>121</sup> Kay 2003; Selbig 2009; Van Metre and Mahler 2003.

<sup>&</sup>lt;sup>122</sup> The Fact Sheet states that "[t]he Norris and Henry (2019) study alone was not integral to EPA's inclusion of the eligibility requirement on the use of coal-tar sealcoat." Id. at 23. The final MSGP should eliminate any references to the paper, which is an example of regulatory and scientific "hide the ball." The two coauthors were an EPA scientist (Norris) and a professor (Henry). According to a June 10, 2019 email from Rebecca Clausen, EPA, to Anne LeHuray, PCTC, while the two authors have "been collaborating . . . for the past 20 years," the crucial, EPA-funded "Unmix Optimum" model underlying their paper resides with Dr. Henry. As a result, "[t]here is . . . no associated documentation about the format of model input and output that EPA has, or could provide[, and a]ppropriated funds would be required for EPA to contract

#### **B**. Coal Tar Sealcoat Is Not a Significant Source of Aquatic or Human Toxicity.

The Fact Sheet states that "[m]any PAHs can have impacts on humans and the environment. Several PAHs have been shown to be extremely toxic to and bioaccumulate in fish and aquatic invertebrates, and are known or probable human carcinogens."<sup>123</sup> In fact, as explained below, PAHs in the natural environment are not particularly toxic to either aquatic life or humans, and no field study has shown otherwise. Thus, the proposed eligibility criterion would not reduce risks to either.

#### 1. The studies cited in the Fact Sheet do not rely on EPA's own guidance for assessing the aquatic toxicity of PAHs.

#### The physical/chemical properties of PAHs require specialized a) approaches developed by EPA.

As explained in Sections II.A. of this Appendix, PAHs in the environment partition strongly to the solid phase, becoming tightly bound to organic materials in complex mixtures. Because of these physical/chemical properties, PAH-containing solids are not very bioavailable or bioaccessible. As a result, sediment PAH concentrations have not been found to correlate with observed toxicity. In recognition of these facts, EPA developed both its ESB and TIE guidance documents so that evaluation of risks for exposures to PAHs in the aquatic environment would more closely reflect their actual toxicity.

EPA's 2003 ESB guidance recommended a procedure using measurements of sediment concentrations of PAHs and Total Organic Carbon (TOC). EPA's subsequent (2012) ESB guidance referred to this as the "one-carbon model." The one-carbon model assumes a simple binary partitioning factor between aqueous and solid phases, with the effective (i.e., bioavailable) concentration of PAHs being represented by the concentrations estimated to be present in potential pore water (i.e., the water immediately surrounding the solid particles where the balance of the PAHs would be bound). The guidance calculates these pore water concentrations using solidaqueous phase partition coefficients and the organic carbon content of the sediment. Using TOC and PAH concentration data, along with appropriate partitioning factors, the method results in an estimate of freely dissolved-and, presumably, bioavailable-PAH concentrations that can be used to develop an "Interstitial Water Toxic Unit" (IWTU) for the sediment sample.

However, even the one-carbon model has been found to routinely overestimate potential risk. This is because use of TOC concentrations alone results in overestimated pore water PAH concentrations that, in turn, result in overestimated IWTUs. More accurate predictions of freely dissolved PAH concentrations have been found to result from calculations that include additional

with Dr. Henry to develop a version of the code that could be distributed for peer review." This is not consistent with EPA's Information Quality Guidelines, which state: "It is important that analytic results for influential information have a higher degree of transparency regarding (1) the source of the data used, (2) the various assumptions employed, [and] (3) the analytic methods applied . . . ." EPA, Guidelines for Ensuring and Maximizing the Quality, Objectivity, Utility, and Integrity of Information Disseminated by the Environmental Protection Agency, EPA/260R-02-008 (Oct. 2002), § 6.3, pp. 20-21. <sup>123</sup> Fact Sheet at 21.

partitioning to the solid phase related to the presence of other adsorptive materials in the sediment, particularly black carbon and weathered coal tar pitch particles.<sup>124</sup> EPA's 2012 ESB guidance calls this the "two-carbon model." A study of real-world sediments at a former manufactured gas plant (MGP) that calculated concentrations and IWTU values using both the one- and two-carbon methods found, as follows:

Although both the one-phase and two-phase models accurately predicted concentrations of PAHs that were not toxic to aquatic invertebrates, the two-phase model was more often in agreement with results of sediment toxicity tests. While the bioavailability and toxicity of PAHs may vary at other sites, the two-phase model correctly predicted that sediments from these sites with concentrations of total PAHs as high as 52 mg/kg were not toxic to invertebrates.<sup>125</sup>

### b) Properly-conducted field studies of PAHs have not found toxicity.

Consistent with the foregoing, field studies of PAHs in aqueous environments that employ EPA's recent guidance have not observed them to be toxic; indeed, no field study has found otherwise.<sup>126</sup>

The Southern California Coastal Water Research Center (SCCWRC) has conducted studies using TIE and passive sampler (IWTU) methods to identify chemicals in sediments associated with measured toxicity. A study of parking-lot runoff found that every runoff tested had toxic effects, and that chemicals in the dissolved phase were the likely principal cause of toxicity; PAHs were not present in the dissolved phase at concentrations associated with toxicity.<sup>127</sup>

An SCCWRC study of sediments in a California estuary used TIE methods to identify causes of toxicity related to whole sediment and pore water concentrations of a variety of compounds. Toxicity was found to be widespread but highly variable. PAHs were identified in pore waters at concentrations that were thought to be unlikely contributors to the observed toxicity.<sup>128</sup>

<sup>&</sup>lt;sup>124</sup> Khalil, M. F., et al. (2006). Role of Weathered Coal Tar Pitch in the Partitioning of Polycyclic Aromatic Hydrocarbons in Manufactured Gas Plant Site Sediments. *Environmental Science & Technology*, 40(18), 5681-5687. doi:10.1021/es0607032.

<sup>&</sup>lt;sup>125</sup> Kane Driscoll, S. B. et al. (2009). Predicting Sediment Toxicity at Former Manufactured Gas Plants Using Equilibrium Partitioning Benchmarks for PAH Mixtures. *Soil and Sediment Contamination: An International Journal*, *18*(3), 307-319. doi:10.1080/15320380902799508.

<sup>&</sup>lt;sup>126</sup> See Kane Driscoll 2020 (Appendix D) at 1.

<sup>&</sup>lt;sup>127</sup> Greenstein, D. et al. (2004). Toxicity of Parking Lot Runoff After Application of Simulated Rainfall. *Archives of Environmental Contamination and Toxicology*, *47*(2), 199-206. doi:10.1007/s00244-004-3018-0.

<sup>&</sup>lt;sup>128</sup> Greenstein, D. J et al., (2013). The use of sediment toxicity identification evaluation methods to evaluate clean up targets in an urban estuary. *Integrated Environmental Assessment and Management*, doi:10.1002/ieam.1512

For example, Figure 11 below is from a study of sediments at four manufactured gas plant (MGP) sites. The study found that some sediment samples were toxic to the common test species *H. Azteca*, but that the toxicity did not correlate with sediment PAH concentration.<sup>129</sup>



FIGURE 1. CHRONIC TOXICITY TO H. AZTECA (28-DAY) CANNOT BE PREDICTED FROM TOTAL PAH CONCENTRATION IN MGP SEDIMENT

Figure 11. Chronic toxicity to *H. Azteca* (28-day) cannot be predicted from total PAH concentration in MGP sediment.

The U.S. Department of Defense Environmental Security Technology Certification Program (ESTCP) funded a demonstration project to determine whether pore-water PAH concentrations in sediments more accurately reflected observed toxicity than whole-sediment concentrations, and whether EPA's EqP guidance regarding ecological risk might be applied at DoD facilities.<sup>130</sup> Similar to findings in sediments at MGP sites and at a variety of locations studied by the Sediment Contaminant Bioavailability Alliance (SCBA), no correlation was observed between PAH concentrations in sediments collected from the Anacostia River near the Navy Yard and toxicity. Figure 12 below illustrates the Anacostia River data, along with SCBA results from many different locations.

<sup>&</sup>lt;sup>129</sup> Neuhauser, E. et al. (2006). Bioavailability and toxicity of PAHs at MGP sites. Land Contam. Reclam. 14:261-266.

<sup>&</sup>lt;sup>130</sup> Geiger, S. C. (2011). *The Determination of Sediment Polycyclic Aromatic Hydrocarbon Bioavailability Using Direct Pore Water Analysis by Solid-Phase Microextraction*. US Dept. of Defense ESTCP Report ER-200709.



Figure 5-2. Bulk Sediment PAH<sub>16</sub> Compared to *H. azteca* Survival.

Figure 12. Bulk sediment concentration of PAH<sub>16</sub> compared to *H. Azteca* survival in Anacostia River (larger symbols with error bars) and SCBA sediment samples illustrating that PAH concentration is not a predictor of toxicity.

These are just a few examples of the large body of scientific research<sup>131</sup> demonstrating that sediment PAH concentrations are not a predictor of toxicity. Among the insights gained by this research is that, as stated in the ESTCP report of the Anacostia River demonstration project:

[U]se of the TEL [threshold effect level] and PEL [probable effect level] screening values (1.6 and 22.8 mg/kg, respectively) would result in almost all of the PAH16 samples exceeding the screening values, and all of the PAH34 samples exceeding the screening values.<sup>132</sup>

Findings such as these are why EPA and others no longer recommend using TEL and PEL values as generic PAH screening concentrations, instead relying on values generated by EPA's more sophisticated ESB and TIE guidance documents. For this reason, application of EPA's ESB and TIE guidance to regulation of sediment PAHs is not only a more scientifically defensible approach, but also a means of ensuring that EPA focuses resources on problems that actually exist.

## c) The literature cited by the Fact Sheet relies almost exclusively on outdated approaches.

Unfortunately, with one exception, the Fact Sheet exclusively cites literature that reflects none of the scientific advances embodied in the ESB and TIE guidance—guidance which

 <sup>&</sup>lt;sup>131</sup> See also Bryer 2010; Bommarito et al. 2010a, b. See generally Kane Driscoll 2020 (Appendix D) at 2.
<sup>132</sup> See Geiger 2011 at 48

demonstrates that the appropriate measure of PAHs in sediment is freely-dissolved concentrations suitable for calculation of IWTU. Rather, the Fact Sheet cites literature that relies on PEL values, without any citation to the bioavailability literature.<sup>133</sup>

The one exception is Weinstein et al. 2010,<sup>134</sup> which is not a primary source of information about the physico-chemical behavior or risk factors of PAHs, but a screening-level ecological and human health risk assessment. This study compared real world data (measurements of PAHs in sediments collected from 19 stormwater detention ponds in South Carolina) with already available criteria and calculated IWTUs. For the ecological screening assessment sediment, PAH concentrations were evaluated using two methods: (1) comparison of concentrations normalized to 1% TOC with TEL and PEL values, and (2) calculation of ESB values using EPA's one-carbon model (but not the two-carbon model).

Of the 19 ponds evaluated, one was located in a low-density residential area and all four were located in industrial areas containing PAH concentrations calculated to exceed the criterion of >1 IWTUs. (Interestingly, both methods gave similar results.) However, the study did not include an evaluation of the actual ecological health of the detention ponds, which prevents an assessment of whether the screening assessment was an accurate under- or overestimation of actual ecological risks.

In the peer-reviewed literature, there have been few studies that include a quantitative assessment of the health of an ecosystem combined with a study to determine how ecosystem impairment may be associated with the extent and characteristics of PAHs in sediment. Most studies of sediment PAHs compare concentrations with sediment quality thresholds, such as the TEL or PEL consensus standards, rather than with IWTU values.

Studies conducted mostly in the Chesapeake Bay, its tributaries (especially the Anacostia and Potomac Rivers), and in the Great Lakes have focused on bottom-feeding fish, particularly Brown Bullhead catfish. Bottom feeding involves oral and dermal exposure to chemicals in sediments and, conceivably, pore water. Over decades, associations between sediment PAH concentrations and liver or skin tumors in these catfish have been proposed and, at least in the case of skin tumors, invalidated.<sup>135</sup> In a recent update on Brown Bullheads in the Anacostia, a significant decrease in the prevalence of liver tumors was noted between 1996 and 2016. The concentrations of PAHs in sediments at the sampling locations did not change between 2000 and 2015, but PCB and DDT concentrations did decline in fish fillets between 1996 and 2013. The

<sup>&</sup>lt;sup>133</sup> See Fact Sheet at 21 (citing IRIS 2014; NRC 2019; Scoggins et al. 2007; U.S. Department of Health and Human Services 2014).

<sup>&</sup>lt;sup>134</sup> Weinstein, J. E. et al. (2010). Screening-level ecological and human health risk assessment of polycyclic aromatic hydrocarbons in stormwater detention pond sediments of Coastal South Carolina, USA. *Journal of Hazardous Materials*, *178*, 906-916,

<sup>&</sup>lt;sup>135</sup> Pinkney, A. E., et al. (2011). Tumor prevalence and biomarkers of genotoxicity in brown bullhead (*Ameiurus nebulosus*) in Chesapeake Bay tributaries. *Science of the Total Environment*, 248–257. doi:10.1016/j.scitotenv.2011.09.035

authors proposed a complex interaction between PAHs, PCBs, and DDT as a potential explanation of the decline in liver tumor prevalence.<sup>136</sup>

Some researchers have focused instead on *in vitro* studies of cells or cellular extracts exposed to PAHs.<sup>137</sup> Such studies are useful in identifying modes of toxic action. However, they are less useful than studies designed to establish the concentrations of sealant runoff that *could* cause adverse effects to aquatic organisms, or that examine whether sealant runoff *has* caused adverse effects to aquatic organisms in the field.

# 2. The toxicity of PAHs to humans was widely overestimated until recently—a fact not recognized by the Fact Sheet.

The limited bioavailability of PAHs in the environment also complicates the process of assessing potential human health risks from PAH exposures. This complication is exacerbated by the tendency of human health risk assessments of PAHs to rely on one or both of two outdated documents: a provisional PAH guidance issued almost 30 years ago<sup>138</sup> and an oral slope factors for benzo(a)pyrene established by EPA's IRIS program almost 40 years ago.<sup>139</sup>

For example, the Fact Sheet notes that Weinstein 2010 found that four commercial ponds, one low density residential pond, and one golf course pond had sediment PAH concentrations that exceeded preliminary remediation goals (PRGs) promulgated in 2009 by EPA Region IX.<sup>140</sup> Even assuming that PRGs developed using the long-term exposure scenarios mandated by EPA for hazardous waste site risk assessments are an appropriate screening tool for detention pond sediments, the PRGs were developed based on the 1984 IRIS cancer slope factor for benzo(a)pyrene.<sup>141</sup>

However, the 1984 IRIS value for benzo(a)pyrene was superseded by a revised IRIS value finalized in 2017.<sup>142</sup> The 2017 value resulted in a reduction of the cancer slope factor by almost an order of magnitude. For benzo(a)pyrene,which was used by EPA as the index compound for PAHs, the slope factor was reduced from 7.3 mg/kg-day to 1 mg/kg-day. The 2017 IRIS update

<sup>&</sup>lt;sup>136</sup> Pinkney, A. E. et al. (2019).

<sup>&</sup>lt;sup>137</sup> Kienzler et al. (2015); Titaley et al. (2016).

<sup>&</sup>lt;sup>138</sup> U.S. EPA 1993. Provisional Guidance for Quantitative Risk Assessment of Polycyclic Aromatic Hydrocarbons. EPA/600/R-93/089.

<sup>&</sup>lt;sup>139</sup> U.S. EPA/540/1-86/022, NTIS PB86134335 (1984).

<sup>&</sup>lt;sup>140</sup> Fact Sheet at 21.

<sup>&</sup>lt;sup>141</sup> Weinstein 2010.

<sup>&</sup>lt;sup>142</sup> U.S. EPA (2017). Toxicological Review of Benzo(a)Pyrene. EPA/635/R-17/003Fa.

was, therefore, EPA's Agency-wide recognition that PAHs are not as significant a human health risk as had been thought in the early development of quantitative risk estimates.<sup>143,144</sup>

EPA engaged in a years-long effort to revise its 1993 provisional PAH risk assessment guidance,<sup>145</sup> which is based on the concept of relative potency factors (RPFs). In February 2010, EPA submitted a draft RPF guidance document<sup>146</sup> to EPA's Science Advisory Board (SAB) for review. The SAB found that the RPF approach of additive toxicity of individual PAH compounds results in highly misleading estimates of risks associated with exposures to whole PAH mixtures in the real world. In March 2011, the SAB recommended

that EPA consider developing a whole mixtures approach for PAHs. This approach could validate the RPF approach and in the future, could replace the RPF approach. The Agency should set this as a strategic initiative, with a specific timeline and benchmarks, that lays the foundation for an underlying concerted research program.<sup>147</sup>

In response, in May 2011, the Administrator agreed with the SAB's recommendation.<sup>148</sup>

EPA's efforts to set human health benchmarks for PAHs have since shifted to the Toxic Substances Control Act (TSCA) program. In its 2014 update to the TSCA Work Plan,<sup>149</sup> EPA announced that:

EPA believes this chemical [benzo(a)pyrene] and other PAHs should be assessed as a category rather than as individual chemical substances.<sup>150</sup>

<sup>&</sup>lt;sup>143</sup> The 2017 value was derived from a comparison of the tumors induced by coal tar and benzo[a]-pyrene in a two-year bioassay. See Culp, S. et al. 1998. Carcinogenesis 19(1):117–124. The study found that forestomach tumors were induced by benzo(a)pyrene in all three groups of mice. Not only were forestomach tumors not observed in the coal tar bioassays, but the relevance of observations in the rodent forestomach to humans—who do not have forestomach organs – remains a source of uncertainty.

<sup>&</sup>lt;sup>144</sup> U.S. EPA makes available "generic tables" combining the most recent risk-based screening values developed by each regional EPA office. The generic tables includes PAH values updated for the 2017 IRIS assessment of benzo(a)pyrene. Adding together the residential soil screening level (1 in 1 million risk or hazard quotient = 1) for the 17 listed PAHs (including naphthalene), and assuming each compound is present at equal concentrations, yields a screening value of about 33,000 mg/kg (over 3%) total PAHs. <sup>145</sup> U.S. EPA 1993.

<sup>&</sup>lt;sup>146</sup> Development of a Relative Potency Factor (RPF) Approach for Polycyclic Aromatic Hydrocarbon (PAH) Mixtures (February 2010 Draft). EPA/635/R-08/012A.

<sup>&</sup>lt;sup>147</sup> Letter to U.S. EPA Administrator Lisa Jackson from U.S. EPA's SAB, March 17, 2011. EPA-SAB-11-004.

<sup>&</sup>lt;sup>148</sup> Letter from Lisa Jackson to EPA SAB, May 17, 2011.

<sup>&</sup>lt;sup>149</sup> U.S. EPA, TSCA Work Plan for Chemical Assessments: 2014 Update. Office of Pollution Prevention and Toxics, October 2014. The status of this Work Plan and subsequent plans for IRIS assessments is addressed in Appendix A.

<sup>&</sup>lt;sup>150</sup> *Id.* at 7; *see also* Appendix A for more current discussion of EPA's research plans.

EPA is currently using the list of Work Plan chemicals as the principal source of substances to be evaluated pursuant to the revised TSCA legislation. Thus, one can expect that, in the coming decade, EPA will be conducting a risk evaluation of PAHs as a group.

For decades, the U.S. FDA has recognized coal tar as "generally recognized as safe and effective" (GRASE) for use as an over-the-counter topical medication for skin conditions such as dandruff, seborrheic dermatitis, or psoriasis.<sup>151</sup> Because it is used in pharmaceutical applications, coal tar has long been included in FDA's safety surveillance program, known as the FDA Adverse Event Reporting System (FAERS).<sup>152</sup> Thus, human exposure via the skin can be assessed directly. For the years included in the online FAERS database, from 1975 to 2019, there were 74 case reports involving coal tar medicinal products. The reported effects including pruritis (itching, the most common reported effect, 10 of 74), ineffectiveness (7 of 74), hypersensitivity (6 of 74), and a variety of diverse reactions with 5 or fewer reports.

In 2016, the Ministers of Environment and Climate Change Canada and Health Canada published for public comment<sup>153</sup> proposed risk management measures for coal tars and coal tar distillates. The scientific basis for the risk management scope proposed for RTS included calculation of margins of exposure (MOE) for risks potentially related to exposure to PAHs in house dust said to be related to the use of RTS on parking lots using data for residential dust reported by Mahler et al. (2010).<sup>154</sup> The screening assessment reported a lifetime adjusted MOE value of 15,500. By comparison, the United Nations World Health Organization's Joint Expert Committee on Food Additives, the Scientific Committee of the European Food Safety Agency<sup>155</sup> (EFSA), and the UK Committee on Carcinogenicity<sup>156</sup> recommend use of MOE > 10,000 as an indicator of low concern for public health. The EFSA Scientific Committee concluded:

The Scientific Committee is of the view that in general a margin of exposure of 10,000 or higher, if it is based on the  $BMDL_{10}$  from an animal study, and taking into account overall uncertainties in the interpretation, would be of low concern from a public health point of

<sup>&</sup>lt;sup>151</sup> 21 C.F.R. § 358

<sup>&</sup>lt;sup>152</sup> FAERS: <u>https://www.fda.gov/drugs/drug-approvals-and-databases/fda-adverse-event-reporting-system-faers</u>

<sup>&</sup>lt;sup>153</sup> Canada Gazette, Part I, Vol. 150, June 11, 2016. *Risk Management Scope for Coal Tars and their Distillates* based on scientific findings detailed in the document entitled *Draft Screening Assessment, Petroleum Sector Approach: Coal Tars and Their Distillates*. As of this writing (4/27/2020), these documents have not been finalized.

<sup>&</sup>lt;sup>154</sup> Mahler, B. J. et al. (2010). Coal-tar-based parking lot sealcoat: An unrecognized source of PAH to settled house dust. Environ. Sci. Technol. 2010, 44, 894–900. This paper and others related to RTS cited by ECCC in the risk management scope document have been subjected to detailed PPPR.

<sup>&</sup>lt;sup>155</sup> Opinion of the EFSA Scientific Committee (2005) on a request from EFSA related to A Harmonised Approach for Risk Assessment of Substances Which are both Genotoxic and Carcinogenic. doi:10.2903/j.efsa.2005.282.

<sup>&</sup>lt;sup>156</sup> United Kingdom Committee on Carcinogenicity (2012). Committee on Carcinogenicity of Chemicals in Food, Consumer Products and the Environment: Risk Characterization Methods. COC/G 06 – Version 1.0 (2012).

 $https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/315883/Risk_characterisation_methods.pdf$ 

view and might be reasonably considered as a low priority for risk management actions. (EFSA, 2005, p. 20/31).

In summary, the practice of estimating human health risks that could be associated with exposure to PAH-containing substances based on bioassays of animals exposed to substances in forms that do not occur outside the laboratory—that is, exposures to individual PAH compounds—has been found to be unrepresentative of exposures to PAHs in the environment and to lead to unreliable predictions of toxicity. Factors controlling bioavailability variations introduce additional uncertainties. Since publication of the results of bioassays of benzo(a)pyrene and two MGP-type coal tars in the 1990s,<sup>157</sup> the actual risks related to human exposure to PAH-containing materials have been recognized as likely to be considerably lower than if they were calculated using EPA's provisional guidance.<sup>158</sup> This constitutes a perception problem that was addressed for individual PAH compounds via EPA's 2017 IRIS assessment of benzo(a)pyrene,<sup>159</sup> but has yet to be resolved for coal tar and other PAH-containing mixtures.

#### C. An RTS Ban May Actually Cause Environmental Harm

The asserted basis for the proposed RTS eligibility criterion is that it will reduce the discharge of PAHs from RTS-sealed surfaces and thus reduce toxic impacts to receiving waters. As explained above, both elements of that claim are unfounded. But worse, it is likely that an RTS ban will actually cause greater environmental impacts than the status quo. Appendix C of these describes the economic impacts of an RTS ban. The most widely-available substitute for RTS, asphalt-based sealcoat, is more short-lived and requires more frequent application to maintain the integrity of a paved surface. Many pavement owners, faced with this increased cost, will simply stop maintaining their asphalt surfaces and just replace them when required by pavement degradation.

Many facilities use first flush and complete containment zones to capture and/or divert storm water runoff and incidental spillage in "high impact" areas to treatment systems. Adequate maintenance of paved surfaces is critical to these strategies, as it allows for efficient and effective dry cleanup methods to be used in the event of a spill. Reduced pavement maintenance and repair activities would likely result in significant cracking in paved surfaces, as well as voids in and under paved surfaces. This could degrade pollution prevention practices related to storm water collection and/or diversion of runoff to treatment systems or away from "high impact" areas. This could also result in accumulation of sediment, dusts, debris and liquids that can contain pollutants and degrade the overall effectiveness of dry cleanup and/or spill cleanup. Therefore, the RTS ban in the 2020 Proposed MSGP may result in significant adverse impacts and degradation of storm water runoff quality related to other constituents of concern. These other constituents of concern may pose a much greater risk to storm water runoff and/or receiving water quality than those posed by RTS.

<sup>&</sup>lt;sup>157</sup> Culp et al. 1998.

<sup>&</sup>lt;sup>158</sup> U.S. EPA 1993.

<sup>&</sup>lt;sup>159</sup> U.S. EPA 2017.

#### D. The Proposed Rule Does Not Have a Rational Basis.

The foregoing demonstrates that EPA's asserted basis for the proposed rule is unfounded. In the natural environment, PAHs bond tightly to organic materials and are not bioavailable in toxic amounts. This generalization holds true for PAHs from RTS. As a result, storm events do not result in PAHs being transported from RTS-sealed surfaces to the aquatic environment in quantities that are toxicologically significant. Additionally, where bans of RTS have been imposed, levels of PAHs have not declined—or, any declines are more logically associated with other events.

The weight of the best available science and evidence indicates that PAHs from RTS are not a cause of aquatic toxicity and do not pose risks to humans. As a result, the proposed eligibility criterion would not reduce risks.

The APA requires the agency to show a rational connection between the facts and the agency's conclusion. *FERC v. Electric Power Supply Ass'n*, 136 S. Ct. 760, 782 (2016). The best available science shows that EPA's asserted connections are unfounded, and that EPA's proposed ban would not accomplish the agency's goal. The proposed eligibility criterion is, thus, irrational and illegal under the APA.

### Appendix C Industry-Related Concerns

# I. Despite EPA's Multiple Assertions that There Are Alternative Products with Similar Performance and Cost to RTS, No Such Alternatives Are Available.

In its 2020 Proposed MSGP Fact Sheet, EPA contends, without supporting facts, that it has identified alternatives that are similar in product performance and cost to RTS. The agency cites asphalt emulsion sealants and acrylic sealants as examples. It also notes that pervious concrete, permeable asphalt, and paver systems that do not require sealants would reduce discharges, "but may not be appropriate for use with all industrial activities."<sup>160</sup> EPA concludes:

Given the comparable costs among products, EPA assumes that most facilities who intend to use coal-tar sealcoat will be able to find a product alternative at negligible cost difference yet with similar performance.<sup>161</sup>

EPA offers no support for its contentions in the Fact Sheet, nor does it list any references that would support these assertions and assumptions. In fact, there is substantial information in the public domain that should have led EPA to the opposite conclusion.

While there are certainly alternatives to RTS, none of them come close to RTS in terms of performance and cost. As documented below, the alternatives do not perform as well as RTS and their lifetime costs are higher. These are not our observations alone, but reflect the conclusions of many independent parties who have studied the matter.

### A. RTS Performs Better Than the Alternatives.

### 1. Asphalt Sealants

Where RTS is available, the market prefers it over other alternatives. The reason lies in its performance at protecting asphalt from damage related to petroleum chemicals, road salts, and a variety of other chemicals, as well as UV radiation and oxidation. Protection against damage related to petroleum products is an important reason why coal tar-based sealants are specified at civilian and military airports. Because of its superior performance at resisting chemical and environmental insult, RTS has been the preferred pavement maintenance sealant used on pavements in industrial and commercial areas for decades, including parking lots, gas stations, truck and bus terminals, airport aprons, and taxiways. RTS is also used on driveways for protection and to enhance curb appeal.

While asphalt-based emulsions have many of the same beneficial properties as RTS, they lack coal tar emulsion's superior resistance to petroleum, ultraviolet bleaching, and salts. An asphalt emulsion is a mixture of liquid asphalt and water. Manufacturers have started adding special chemicals and pigments to asphalt emulsions to improve their resistance to petroleum products and to enhance other performance characteristics, but they are still more susceptible to

<sup>&</sup>lt;sup>160</sup> Fact Sheet at 24

<sup>&</sup>lt;sup>161</sup> *Id*.

damage caused by petroleum products. Asphalt-based emulsions generally have life spans of two to three years, whereas RTS sealants will generally last four to six years.

## a) From manufacture to application of RTS, every step in the process is governed by performance-based standards.

Beginning in the 1970s, the US Army Corps of Engineers (Corps) undertook investigations of fuel-resistant sealers. Shoenberger (1994) gave an overview of performance issues from the perspective of the military that is still applicable:

Asphalt concrete pavements make up approximately 96% of the surfaced pavements in the United States (Roberts et al. 1991<sup>162</sup>). The majority of parking areas used for low-pressure tire vehicles (automobiles and light trucks) are also paved with asphalt concrete. Since asphalt cement is a petroleum-based product obtained in the distillation of crude oil, it will dissolve or soften when exposed to petroleum-based products. Therefore, asphalt concrete pavements are susceptible to damage from fuel or oil spills or drippage. The damaging materials include gasoline, diesel fuel, hydraulic and brake fluids, aviation fuels, and other petrochemical and synthetic materials. These oils and fluids are required for the operation of vehicles, and the amount of these fluids that falls to the pavement surface from these vehicles depends on the condition of the vehicle and its maintenance operations. ... Fuel spills and drippage result in the softening and leaching away of asphalt binder from the aggregate. This causes pavement failures due to rutting or raveling of the surface aggregate in the spillage areas.

Normally, fuel or solvent spillage is not a problem on roadways. The speed and movement of the vehicle spreads out the spillage over a large area. The spilled material tends to be worn off or it evaporates from the pavement due to traffic and the effects of weather (rain or sunshine). But, areas of slow speed or highly channelized traffic often have sufficient fuel spillage accumulation to cause damage to the asphalt concrete pavement. Parking areas, especially those with constant vehicle turnover, are very susceptible to damage from such spillage. ....

Fuel spillage problems can also be particularly severe for airfield pavements because several types of aircraft engines release the unused portion of fuel remaining in the engine at shutdown, in addition to normal drippage and other losses. These materials will damage the pavement surface almost immediately and even prompt flushing or flooding of the area with water, and ideal evaporation conditions cannot entirely prevent damage. Military installations have all of these problems with fuel spillage plus the possibility of a sabotage scenario. Such a

<sup>&</sup>lt;sup>162</sup> Roberts, F. L., Kandhal, P. S., Brown, E. R., Lee, D.-Y., and Kennedy, T. W. (1991). "Hot mix asphalt material, mixture, design, and construction." Rep., NAPA, Lanham, MD

scenario could involve fuel being intentionally dumped on an airfield pavement in order to interfere with airplane operations.<sup>163</sup>

Among the early findings of the Corps' research was that coal tar-based sealants had superior fuel resistance, but that the products available at the time were inconsistent in performance. The industry responded in 1994 by establishing PCTC as an engineering research and standard-setting program within the Engineering Department at the University of Nevada – Reno (UNR).<sup>164</sup> The program's goal was to research and establish performance-based standards for the manufacture of what the industry now calls refined coal tar-based sealant (RTS). The results of the UNR phase of PCTC's history are reflected in the ASTM standards that cover everything from the production of the refined coal tar base, RT-12, to its application. These standards are:

- ASTM D490-92(2016), Standard Specification for Road Tar;
- ASTM D4866/D4866M-88(2017)e1, Standard Performance Specification for Coal Tar Pitch Emulsion Pavement Sealer Mix Formulations Containing Mineral Aggregates and Optional Polymeric Admixtures;
- ASTM D5727/D5727M-00(2017)e1, Standard Specification for Emulsified Refined Coal Tar (Mineral Colloid Type);
- ASTM D6945-03(2017), Standard Specification for Emulsified Refined Coal-Tar (Ready to Use, Commercial Grade);
- ASTM D6946-13, Standard Specification for Emulsified Refined Coal-Tar (Driveway Sealer, Ready to Use, Primary Residential Grade), 2013); and
- ASTM D3423 / D3423M-84(2015)e1, Standard Practice for Application of Emulsified Coal-Tar Pitch (Mineral Colloid Type).

The ASTM standards are supplemented by PCTC's guides for preparation of performance-based specifications for RTS:<sup>165</sup>

- PCTC Guide Specification-PCTC01: Guide for Preparation of Specifications for the Application of a Refined Coal Tar Emulsion Without Additives Over Asphaltic Pavements
- PCTC Guide Specification-PCTC02: Guide for Preparation of Specifications for the Application of a Refined Coal Tar Emulsion With Additives Over Asphaltic Pavements

RTS manufactured following ASTM's performance-based standards has, for nearly three decades, been consistent and predictable in its resistance to petroleum products and other chemicals and environmental factors that can damage, and shorten the service life of asphalt surfaces. The market still prefers it over the alternatives.

<sup>&</sup>lt;sup>163</sup> Shoenberger, J. (1994). Performance of FuelResistant Sealers for Asphalt Concrete Pavements. *Journal of Materials in Civil Engineering*, 6(1), 137-149. doi:doi:10.1061/(ASCE)0899-1561(1994)6:1(137).

<sup>&</sup>lt;sup>164</sup> PCTC was originally named the Pavement Coatings Technology *Center*. It was renamed as the Pavement Coatings Technology *Council* in 2008 when sponsors of the original Center reorganized PCTC as a 501(c)(6) trade association.

<sup>&</sup>lt;sup>165</sup> Available at http://www.pavementcouncil.org/1520/.

### b) While the performance of some asphalt-based sealants has improved, the variability of such sealants limits consistency.

The performance of some asphalt-based emulsion sealants (ABS) has greatly improved in recent years, although the inherent problem of resistance to petroleum fuels remains less satisfactory. Through research and development into factors such as the composition of asphalts and ingredients to improve asphalt characteristics, companies that make both RTS and ABS have developed ABS that meets the needs of many customers who have less stringent performance requirements. Product consistency, however, is a continuing problem.

To date, performance-based standards have not been developed for ABS because, by both the nature of petroleum and the choices made in petroleum markets and refining processes, the physical properties of asphalt are inconsistent. The asphalt used in the manufacture of ABS is essentially what remains at the end of distillation of heavier crude oils.<sup>166</sup> Crude petroleum extracted from the many different oil fields around the world varies widely from light-to-heavy crude. Over the years, refining processes have evolved to remove increasing amounts of the more-valuable lighter crude components from refining residuals, resulting in asphalts that vary widely in both chemical and physical characteristics. These process changes have exacerbated the inherent variability resulting from the different compositions and characteristics of crude oil extracted from different oil fields around the world.

Changes in the asphalt available to the paving market in recent years have been described, as follows:

North America has experienced (i) significant shifts in the availability of asphalt, (ii) higher costs for the available asphalt but more importantly, (iii) dramatic changes in asphalt quality; all of which, threaten the paving contractor's and roofing manufacturer's long-term ability to provide a high quality product, significantly increases the costs of paving our highways and ultimately, impacts the life cycle of those roofs and highways.<sup>167</sup>

The key to success of a sealant is its performance in protecting underlying asphalt pavements. The reason RTS is the preferred product is because it meets customer performance criteria and does so consistently. Through understanding of the qualities of asphalts available on the market, as well as inclusion of additives in the manufacturing process, some ABS that meets performance criteria is available. But, for reasons beyond the control of the sealant industry, consistency of performance of ABS has been elusive, limiting the ability of ABS sealant manufacturers to consistently provide a high-quality product that meets customer performance expectations.

#### 2. Acrylic Sealants

Acrylic sealants are a specialty product principally used for tennis courts, where they have the advantage of allowing control of the speed of play. As experience has shown on tennis courts,

<sup>&</sup>lt;sup>166</sup> Little-to-no asphalt is produced from light crude oils, such as from some southern US oil fields, or from shale oil.

<sup>&</sup>lt;sup>167</sup> *The Asphalt Challenge*. Engineered Additives LLC. <u>http://engineeredadditives.com/asphaltchallenge.html</u>

however, acrylic coatings are brittle, resulting in the need to resurface courts every few years even though they are not subject to vehicle traffic. Brittleness even more severely limits the useful life of acrylic sealants used on pavements exposed to the heavy load of cars and trucks. Reduced service life only adds to the additional limitation that acrylic sealers are prohibitively expensive for use on large asphalt-paved surfaces. For these reasons, acrylic sealants are not competitive in the pavement maintenance world and are not generally regarded as a viable alternative to RTS.

#### **3.** Permeable Pavements

Permeable pavements are considered a means of ameliorating storm water runoff issues because they are engineered with pore space that allows dissolved and particulate materials washed off by rainfall to permeate the pavement for capture or immobilization by an underlying drainage system or by soils. A standard method of making large concrete or asphalt roadway or parking lot surfaces more porous is to reduce fine particles in the concrete/asphalt mix. Unfortunately, this reduces the load bearing capacity of the pavement. Installation of permeable pavements is also more expensive than traditional pavement, and the pore space available for permeability decreases over time, as the material is compressed or collapsed under the weight of vehicles.

Once such pavements are installed, particulates infiltrate the pore space, leading to declining effectiveness over time and, eventually, complete clogging. In more northerly climates, application of sand and de-icing chemicals can lead to very rapid clogging. To maintain permeability of acrylic pavements, it is necessary to institute a maintenance program involving routine removal of particles from pore space, typically with an industrial vacuum. Without such elaborate and expensive maintenance, infiltration of storm water becomes increasingly inefficient, leading to runoff that is no different than from impervious pavements.

For these reasons alone, permeable products are usually inappropriate for application on surfaces with vehicle traffic. Additionally, the costs of alternative permeable products at the point of application are often greater than those for RTS products, and the cost of maintenance can be much greater than for maintenance of traditional pavements.

#### B. Life-Cycle Cost-Competitiveness of RTS Is Superior

Pavement maintenance programs consist of three different types of operation: preventive maintenance, corrective maintenance, and emergency maintenance. As concluded by the University of Minnesota's Airport Technical Assistance Program (AirTAP) from an assessment of the benefits of a pavement maintenance program:

Preventive maintenance is generally the least expensive type of maintenance, and emergency the most. Emphasizing preventive maintenance will keep pavement in good condition and prolong the time until corrective maintenance is required. A pavement preservation program is designed to preserve a pavement structure, enhance its performance, extend pavement life, and meet user needs. An effective program integrates many preventive maintenance strategies and rehabilitation treatments with the goal of cost-effectively and efficiently enhancing pavement performance...

Pavement preservation has many benefits, the most important of which is preserving a pavement's structural integrity and realizing a substantial maintenance cost savings over the life of the pavement....

To be cost-effective, pavement preventive maintenance treatments should be applied early in the life of a pavement. It is much less expensive to repair a pavement when distresses are just beginning to appear.<sup>168</sup>

Figure 1 below illustrates AirTAP's assessment of the value of a pavement maintenance program.



Figure 1. Preventive maintenance preserves the condition of the pavement and costs significantly less than rehabilitation.

Sealcoating is an integral part of a preventive maintenance program. It extends the useful service life of a pavement asset and costs considerably less than repaying or other measures that may be needed later to correct chronic or acute pavement problems.

A comprehensive assessment of the costs associated with any sealcoating must include the costs incurred over the life cycle of a paved surface. The initial cost is the cost of the sealant being applied to a surface—either RTS or ABS—and the cost of applying it. The cost of application is virtually the same. And, the initial cost of the sealant can be compared in the moment using the price of RTS versus the price of ABS. But, the initial cost does not tell the whole story—the cost over the life cycle of a paved surface must be considered, and this factor favors RTS.

PCTC/COETF estimate that the cost savings of a pavement maintenance program that includes sealcoating results in a 12-year total cost for a commercial installation of about \$0.39/sq. ft. versus an unsealed total cost of \$1.76/sq. ft. This assumes that, after 12 years of use, an unsealed lot would require an overlay with 2" of asphalt.<sup>169</sup> This figure assumes equal longevity for RTS

<sup>&</sup>lt;sup>168</sup> *Pavement preservation: protecting your airport's biggest investment*. AirTAP Briefing Summer 2005. <sup>169</sup> Details of the assumptions used in both commercial and residential examples are illustrated in infographics available at <u>http://www.pavementcouncil.org/education/</u>.

and ABS. When the longer period between needed sealcoat applications of RTS is considered, the 12-year cost of a sealcoat program would be even less than the example calculation.

EPA needs to consider that, in the real world, cost is more than an initial, one-time consideration. The life-cycle cost must be considered. And, when it is, RTS is the clear winner. For this reason, EPA's assumption that,

[g]iven the comparable costs among products, EPA assumes that most facilities who intend to use coal-tar sealcoat will be able to find a product alternative at negligible cost difference yet with similar performance,<sup>170</sup>

is not based in fact.

#### C. Independent Sources Recognize the Superior Value and Performance of RTS

Missouri State University (MSU) studied the relative merits of RTS vs. alternative products in 2015. The MSU Board of Governors, after "much research completed," found asphalt sealant was not recommended due to "cost and longevity." More specifically, MSU found that the asphalt life-cycle was two to three years, while that for RTS was four to six years. And, yet, the cost of installation was almost the same, 0.09/ft<sup>2</sup> for asphalt emulsion and 0.11/ft<sup>2</sup> for RTS.<sup>171</sup> And, according to an article in the Springfield (MO) News-Leader, the University found that asphalt emulsion was "less effective at blocking water and lasts half as long."<sup>172</sup>

Another exhaustive study of sealcoat alternatives was conducted in 2010 by Geoffrey H. Butler, an architect based in Springfield, Missouri. In his white paper, which he provided to the City of Springfield, Mr. Butler explained what he had learned about pavement sealers from designing projects that involved parking lots:

As a developer and property owner, I have experience with both the coal tar sealers and the asphalt based sealers.... The [asphalt based sealer] wore off rather quickly lasting only two years... I have used coal-tar sealants... very successfully. It lasts 4-6 years per application, does not track... and has never re-emulsified.... Coal tar sealant is highly resistant to gas and oil.... The cost to properly repair or replace an asphalt parking lot exceeds the cost to build it in the first place.<sup>173</sup>

These two independent sources confirm the cost and performance qualities discussed above. Many other could be cited. In contrast, EPA provides no explanation or references to support its assertions that cost-effective alternatives are available. In fact, alternatives of similar performance and cost to RTS are not available. EPA's claims to the contrary are simply untrue, and the Agency needs to correct the record.

<sup>&</sup>lt;sup>170</sup> Fact Sheet, p. 24. EPA provides no supporting information for this "assumption."

<sup>&</sup>lt;sup>171</sup> Parking Lot Sealant Summary, Board of Governors Meeting, Missouri State University, February 26, 2015

<sup>&</sup>lt;sup>172</sup> Riley, C. "After trying alternative, MSU resumes use of coal tar sealant," April 3, 2015 Springfield News-Leader, Springfield, Missouri.

<sup>&</sup>lt;sup>173</sup> Geoffrey Butler, AIA, "What I have learned about Coal Tar Sealers," January 14, 2010

### Appendix D Kane Driscoll Report

### Exponent

### EXTERNAL MEMORANDUM

 TO: Anne LeHuray, Pavement Coating Technology Council
FROM: Susan Kane Driscoll, Ph.D., Exponent Inc.
DATE: May 29, 2020
SUBJECT: Responses to EPA Request for Comment 2 National Pollutant Discharge Elimination System (NPDES) Multi-Sector General Permit (MSGP) for Stormwater from Industrial Activities Docket ID: EPA-HQ-OW-2019-0372

These comments are in response to:

Request for Comment 2: EPA requests comment on the following:

• Any studies that provide data on the level of PAHs from coal-tar sealed pavements, the sources of measured PAHs in the aquatic environment, the levels of PAHs in fish and seafood, and associated chemical and biological impacts that may occur via stormwater discharges.

I am an aquatic toxicologist and have been conducting research related to bioavailability and toxicity of petroleum products and polycyclic aromatic hydrocarbons (PAHs) to aquatic organisms for 30 years. I recently performed a comprehensive review of the literature on the potential impacts of refined coal-tar-based sealant (RCTS) runoff to aquatic organisms on behalf of the Pavement Coatings Technology Council (Kane Driscoll et al. 2019). The goal of the review was to evaluate and summarize the strengths and weaknesses of the lines of evidence presented in the scientific literature regarding potential effects. I also provided recommendations for environmentally relevant study designs that could be used to examine cause-effect relationships under field conditions.

Our review of available studies highlights the need for measures of exposure that can be used to predict whether adverse effects are likely to occur in the field. Our review concludes that currently available studies demonstrate that PAHs in RCTS runoff can cause adverse effect under laboratory conditions, but no studies have demonstrated that sealant runoff causes adverse effects to aquatic organisms in the field.

#### **COMMENTS**

The designs of certain studies on the effects of sealcoat runoff on aquatic organisms are not predictive of exposure or effects to aquatic organisms under environmental conditions, where runoff is diluted and constituents interact with organic matter that reduces bioavailability and toxicity.

- Mahler et al. (2015) reported toxic effects (mortality) of simulated sealant runoff to aquatic organisms under laboratory conditions over time. The authors assert that concentrations of PAHs in sealant runoff tested in this laboratory study (median concentration of 16 PAHs of 328  $\mu$ g/L in undiluted runoff; concentrations in diluted runoff were not measured) are approximations of environmental conditions that might occur in urban streams. However, this median concentration is substantially higher than reported median concentrations of 18 PAH compounds (ranging from 0.05 to 5.72  $\mu$ g/L) in runoff from typical urban source areas, including unsealed parking lots, feeder streets, collector streets, arterial streets, rooftops, and strip malls (Selbig et al. 2009). Importantly, although adverse effects were reported for runoff with high concentrations of PAHs collected at the point of sealant application, the authors acknowledge that many factors can alter toxicity in the environment, including sorption of PAHs to dissolved and particulate organic carbon in surface water.
- McIntyre et al. (2016) reported lethal and sublethal effects (e.g., reduced growth, edema) to juvenile and embryo-larval fish from simulated runoff. Concentrations of 25 PAHs in undiluted runoff ranged from 207 to 1,311  $\mu$ g/L. However, treatment of the runoff with a bioretention system consisting of sand, compost, and bark mulch eliminated all lethal effects and nearly reversed all sublethal effects. This study illustrates the potential for environmental factors (e.g., sorption of PAHs by organic material) to reduce toxicity associated with RCTS runoff.
- A study by Bryer et al. (2010) did not show a clear relationship between concentrations of sealant constituents and effects, including abundance and diversity of invertebrates colonizing sediment mixed with dried sealant flakes. The lack of a concentration-effect relationship limits the usefulness of the study in defining causal relationships between sealant runoff and effects, which could be used to assess the probability that sealant runoff causes adverse effects in the environment.
- Two studies by Bommarito et al. (2010a,b) examined effects of dried flakes of sealants on newts and salamanders. No significant effects were reported for survival, and sublethal effects (e.g., growth, swimming duration and distance, mobility, and activity of liver enzymes) did not show clear concentration-response relationships with the reported measures of sealant exposure. In addition, exposure concentrations used in these studies (e.g., maximum total PAH (TPAH) concentrations in sediment >1,000 mg/kg) are higher than typically observed in the environment.

Studies conducted on cell lines or on chemical extracts useful in identifying modes of toxic action, but are not useful for establishing the concentrations of sealant runoff that could cause adverse effects to aquatic organisms or for examining or whether sealant runoff has caused adverse effects to aquatic organisms in the field.

- Kienzler et al. (2015) conducted a study using *in vitro* exposure with an isolated trout cell line to examine potential effects and mode of toxic action, including cytotoxicity and genotoxicity. This *in vitro* study with a trout cell line is useful for demonstrating potential effects and mode of toxic action but lacks the environmental realism of tests with whole organisms and is not useful for defining exposure concentrations that could cause adverse effects in a field setting.
- Titaley et al. (2016) chemically extracted dried flakes of RCTS with the goal of identifying the specific fractions that caused mutagenicity or developmental effects to embryo-larval fish. Chemical extracts containing PAHs were toxic to fish and caused mutagenicity in the bacterial Ames assay. Exposure to chemical extracts is useful for identifying causative agents and mode of toxic action, but the fact that some PAHs are mutagenic and cause developmental effects to embryo-larval fish has been well established. Toxicity tests conducted on chemical extracts are not, however, directly applicable or useful for identifying environmental concentrations that could be used to predict the potential for adverse effects of sealcoat to aquatic organisms in the field.

# Only one study examined whether runoff from sealed parking lots caused adverse effects to aquatic organisms in the field, but the authors were not able to establish that PAHs in sediment were associated with sealant.

• Scoggins et al. (2007) identified a set of streams adjacent parking lots that, based on aerial photography, were thought to have been seal-coated and selected a subset with higher TPAH concentrations downstream (DS, 4–32 mg/kg) than upstream (US, 0.04–3.9 mg/kg) of the seal-coated lots. Habitat quality indices for DS locations were slightly lower (8% on average) than US locations. Authors also reported significant negative relationships between the sum of toxic units (a standard approach for normalizing for differences in the toxicity of individual PAHs) and taxon richness and density. Authors were unable, however, to identify the source of the PAHs using ratio methods or other analyses. In addition, pre-selection of locations with higher sediment PAH concentrations DS versus US may have biased results.

#### **Summary**

• More useful and predictive concentration-response relationships might be established if concentrations in runoff data collected over time are presented as the sum of toxic units for PAHs rather than as TPAH, which does not account for variability in the toxicity of the mixture of individual PAHsover time, and between laboratory and field exposures. Use of US EPA's Equilibrium Partitioning benchmarks (US EPA 2003) to calculate toxic units for PAHs has been shown to be a better predictor of toxicity than screening

level benchmarks based on TPAH (Di Toro et al. 2007), particularly if PAHs are in a form (e.g., soot, particulate) that reduces bioavailability (Kreitinger et al. 2007; Kane Driscoll and Burgess 2007). Considerable research has demonstrated that the toxicity of PAHs varies greatly among environmental samples, and the toxicity of "freely dissolved" or "bioavailable" PAHs is greater than the bioavailability and toxicity of PAHs that are associated with organic matter or other hydrophobic phases (US EPA 2003). Studies that measure freely dissolved PAHs in water and sediment would be a useful next step for assessing potential impacts of runoff from RCTS on aquatic communities (Burgess 2009; US EPA 2017).

- The types of studies and samples collected should be relevant and predictive of exposure to aquatic organisms. For example, simulated runoff samples collected in the laboratory or in the field at the point of application may not represent the concentrations, compositions, or bioavailability of PAHs in downstream bodies of water where aquatic organisms are exposed. Similarly, runoff samples collected less than 24 hours after application may overestimate exposure concentrations and toxicity, since best management practices prohibit application of RCTS within 24 hours of a predicted rain event.
- Field studies that measure concentrations of freely dissolved, individual PAHs (or other hydrophobic organic constituents of RCTS runoff) at the point of exposure to aquatic organisms (e.g., urban or suburban streams) would be more environmentally relevant and useful for developing cause-effect relationships under field conditions.
  - •

### Acknowledgement

These comments were developed with funding from the Pavement Coatings Technology Council.

#### References

Bommarito T, Sparling DW, Halbrook RS. 2010a. Toxicity of coal-tar pavement sealants and ultraviolet radiation to *Ambystoma maculatum*. *Ecotoxicology*. 19:1147–1156.

Bommarito T, Sparling DW, Halbrook RS. 2010b. Toxicity of coal-tar and asphalt sealants to eastern newts, *Notophthalmus viridescens*. *Chemosphere*. 81:187–193.

Bryer PJ, Scoggins M, McClintock NL. 2010. Coal-tar based pavement sealant toxicity to freshwater macroinvertebrates. *Environ. Pollut.* 158:1932–1937.

Burgess, R.M. 2009. Evaluating Ecological Risk to Invertebrate Receptors from

PAHs in Sediments at Hazardous Waste Sites. U.S. Environmental Protection Agency, Ecological Risk Assessment Support Center, Cincinnati, OH. EPA/600/R-06/162.

Di Toro DM, McGrath JA, Stubblefield WA. 2007. Predicting the toxicity of neat and weathered crude oil: Toxic potential and the toxicity of saturated mixtures. Environ. Toxicol. Chem. 26: 24–36.

Kane Driscoll SB, Burgess RM. 2007. An Overview of the Development, Status, and Application of Equilibrium Partitioning Sediment Benchmarks for PAH Mixtures Human Ecolog Risk Assess, 13: 286–301.

Kane Driscoll S, Kulacki K, Marzooghi S. 2019. A Review of the Literature on Potential Effects of Runoff from Refined Coal-Tar–Based Sealant Coating on Aquatic Organisms. *Integr. Environ. Assess. Manag.* 16:17–27. Kienzler A, Mahler BJ, Van Metre PC, Schweigert N, Devaux A, Bony S. 2015. Exposure to runoff from coal-tar-sealed pavement induces genotoxicity and impairment of DNA repair capacity in the RTL-W1 fish liver cell line. *Sci. Total Environ.* 520:73–80.

Kreitinger JP, Neuhauser EF, Doherty FG, Hawthorne SB. 2007. Greatly reduced bioavailability and toxicity of polyclic aromatic hydrocarbons to Hyalella azteca in sediments from manufactured gas plant sites. Environ. Toxicol.Chem. 26: 1146–1157, 2007.

Mahler BJ, Ingersoll CG, Van Metre PC, Kunz JL, Little EE. 2015. Acute toxicity of runoff from sealcoated pavement to *Ceriodaphnia dubia* and *Pimephales promelas*. *Environ. Sci. Technol.* 49:5060–5069.

McIntyre J, Edmunds R, Anulacion B, Davis J, Incardona JP, Stark JD, Scholz N. 2016. Severe coal tar sealcoat runoff toxicity to fish is prevented by bioretention filtration. *Environ. Sci. Technol.* 50:1570–1578.

Scoggins M, McClintock NL, Gosselink L, Bryer P. 2007. Occurrence of polycyclic aromatic hydrocarbons below coal-tar-sealed parking lots and effects on stream benthic macroinvertebrate communities. *J North Am. Benthol. Soc.* 26:694–707.

Selbig WR. 2009. Concentrations of polycyclic aromatic hydrocarbons (PAHs) in urban stormwater, Madison, Wisconsin, 2005–08: U.S. Geological Survey Open-File Report 2009–1077, 46 p.

Titaley IA, Chlebowski A, Truong L, Tanguay RL, Massey Simonicha SL. 2016. Identification and Toxicological Evaluation of Unsubstituted PAHs and Novel PAH Derivatives in Pavement Sealcoat Products. *Environ. Sci. Technol. Lett.* DOI: 10.1021/acs.estlett.6b00116

U.S. EPA. 2003. Procedures for the Derivation of Equilibrium Partitioning Sediment Benchmarks (ESBs) for the Protection of Benthic Organisms: PAH Mixtures. EPA-600-R-02-013. Office of Research and Development. Washington, DC 20460.

U.S. EPA. 2012. Equilibrium Partitioning Sediment Benchmarks (ESBs) for the Protection of Benthic Organisms: Procedures for the Determination of the Freely Dissolved Interstitial Water Concentrations of Nonionic Organics. EPA-600-R-02-012. Office of Research and Development, Washington, DC 20460.

U.S. EPA/SERDP/ESTCP. 2017. Laboratory, Field, and Analytical Procedures for Using Passive Sampling in the Evaluation of Contaminated Sediments: User's Manual. EPA/600/R- 16/357. Office of Research and Development, Washington, DC 20460.
Appendix E O'Reilly Report

# Exponent

To: Anne LeHuray, Pavement Coating Technology Council
FROM: Kirk O'Reilly, Ph.D., J.D., Exponent Inc.
DATE: May 26, 2020
SUBJECT: Comments on: National Pollutant Discharge Elimination System (NPDES) Multi-Sector General Permit (MSGP) for Stormwater from Industrial Activities Docket ID: EPA-HQ-OW-2019-0372

# Summary

These comments are in response to:

Request for Comment 2: EPA requests comment on the following:

- Whether the permit should include an eligibility criterion related to the application of coal-tar sealcoat to paved areas where industrial activities are located.
- Any studies that provide data on the level of PAHs from coal-tar sealed pavements, the sources of measured PAHs in the aquatic environment, the levels of PAHs in fish and seafood, and associated chemical and biological impacts that may occur via stormwater discharges.

I have been conducting research related to the coal-tar sealant issue since 2009, and my comments are based in part on this work.

The information presented in the fact sheet does not accurately describe the scientific consensus on the sources of PAHs in the environment. Many of the papers cited were written by the U.S. Geological Survey's (USGS's) Mahler and Van Metre or their collaborators. These papers ignore most of the body of literature describing environmental sources of PAHs and focus only on the work of the few researchers who support their position concerning the contribution of sealcoats. The fact sheet fails to cite many papers that have raised technical questions about Mahler and Van Metre's research and conclusions. In the field of PAH source evaluations, there is no consensus that coal-tar sealcoat is an important source.

# Comments

#### Combustion processes, not coal-tar sealant, are the primary source of PAHs in stormwater.

Characterizing sources of PAHs in the environment has been an active field of research for many years (Blumer and Youngblood 1975; Youngblood and Blumer 1975). Bibliographies of some review articles have over a 100 references (Lima et al. 2005; Ravindra et al. 2008; Yunker et al. 2002). There is wide spread consensus that most PAHs in sediments can be traced back to combustion related sources. As noted in Lima et al. (2005), "combustion processes are responsible for the vast majority of the PAHs that enter the environment."

Efforts to evaluate the contributions of PAH sources have consistently identified atmospheric deposition as a significant source to stormwater, soils, paved areas, and sediments in most urban environments (Hwang and Foster 2006; Li et al. 2003; Mastral and Callén 2000; Simcik et al. 1999; Stein et al. 2006; Su et al. 2000; Van Metre et al. 2000; U.S. EPA 2007; Yunker et al. 2002). Specifically, the higher-molecular-weight PAHs typical of combustion-derived particulate matter—consistent with motor exhaust, coal combustion products, or wood smoke—have been found to dominate PAH profiles in sediments impacted by "urban background" sources (Stout et al. 2004). An EPA review (U.S. EPA 2007) concluded that "[a]tmospheric deposition of pollutants, including PAHs, is recognized as a significant contributor in many locations to water quality problems. PAHs are organic compounds primarily formed from the incomplete combustion of organic materials, such as coal and wood."

A number of studies have demonstrated a link between atmospheric emission sources and PAHs in urban environments. Evaluations of PAH chemistry in sediments from lakes, creeks, and reservoirs from across the United States report temporal links between changes in PAH concentrations and increased automobile use and vehicle emissions (Simcik et al. 1999; Stein et al. 2006; Su et al. 2000; Van Metre et al. 2000). In the upper Midwest, the mass and chemistry of PAHs in lake sediment could be linked to specific atmospheric sources associated with activities such as steel production and motor vehicle use (Su et al. 2000; Simcik et al. 1999). Automotive emissions have been shown to be a major source of particulate PAHs in aquatic systems in the Los Angeles basin (Stein et al. 2006) and San Francisco Bay Area (Tsai et al. 2002). The dominance of coal-fired power plants in the eastern United States and gas-fired power plants in the west is a potential explanation for the regional differences in sediment PAH concentrations noted by Mahler et al. (2005).

Two studies found combustion sources to be major contributors of PAHs in urban watersheds. An investigation lead by the New York Academy of Sciences (2007) found that air-based emissions accounted for about 98% of the PAHs released to the environment in the New York– New Jersey Harbor watershed, with residential wood smoke being the largest source. Only about 1.5% of PAHs were released directly to the land, with the remaining releases linked to waterbased sources. On the other side of the country, the Washington State Department of Ecology (2011) also found that wood smoke and vehicle emissions accounted for the majority of PAHs released in the Puget Sound watershed.

#### Studies on Stormwater, PAHs, and Coal-tar Sealcoat

Most of the research suggesting that coal-tar sealcoat is an important source of PAHs in stormwater was conducted by USGS's Mahler and Van Metre, their collaborators, or others who

have used their approach. These studies typically ignore most other research on PAH source characterization, including their own prior work, such as Van Metre et al. (2000) and Van Metre and Mahler (2003, 2005).

Published comments highlight some of the technical problems with the work of USGS and its collaborators:

DeMott RP, Gauthier TD. 2006. Comment on "Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons." Environ. Sci. Technol. 40(11):3657–3658.

In conclusion, we were unable to replicate the computations and identify values from cited sources for a number of the data points represented in Figures 4–6 of Mahler et al. (2005). With regard to the PAH ratio analysis, we could not identify the source of the values presented for stream sediment samples, and the values that we could identify from the City of Austin appear to contradict the interpretation developed by the authors. With regard to the mass balance analysis, we could not identify the source for values from one watershed, the values presented for the other watersheds do not appear to match those from the cited sources, and the previously published values suggest the relative contribution of PAHs from parking lot sources is substantially less than the "majority" source suggested by the authors. Because these uncertainties relate to the two lines of argument indicated to support a conclusion that parking lot sealcoat could be a dominant source of PAHs in urban streams, clarification is important for understanding the strength of the conclusions of this paper.

O'Reilly KT, Pietari J, Boehm PD. 2011. Comment on "PAHs Underfoot: Contaminated Dust from Coal-Tar Sealcoated Pavement is Widespread in the U.S." Environ. Sci. Technol. 45(7):3185–3186.

In a series published in Environmental Science and Technology, Mahler, Van Metre, and their colleagues have investigated the role of coal tar-based pavement sealants (CT-sealants) as a source of [PAHs] in urban sediments. Data from single sediment samples from 10 urban lakes were used to support the claim that CT-sealants provided a "substantial contribution" to many urban watersheds. These authors have recently provided sediment data from 40 lakes (Van Metre and Mahler 2010). We applied these data to the forensics analysis methods used in Van Metre et al. 2009. A lack of consistency in the results of the methods used in these two papers calls into question whether this source characterization supports the hypothesis concerning the relative role of CT-sealants.

Sediment PAH source identification has been the subject of significant research, and atmospheric deposition has been recognized as a primary contributor. While challenging conventional wisdom is the essence of science, it is critical to have sufficient and consistent technical support. Mahler and Van Metre have not evaluated the data in a way to test their null hypothesis, so have not eliminated atmospheric deposition as a major source of PAHs.

O'Reilly KT, Pietari J, Boehm PD. 2012. A forensic assessment of coal tar sealants as a source of polycyclic aromatic hydrocarbons in urban sediments. Environ. Forensics 13(2):185–196.

Atmospheric deposition of particles and their subsequent transport by stormwater is a major source of PAHs in urban sediments. Recently, the results of forensic analysis have been used to promote a hypothesis that refined tar-based pavement sealers (RT-sealers) are another significant source. To evaluate this hypothesis, we applied a suite of forensic methods to a wider range of PAH data. Sediments PAH profiles are no more similar to RT-sealers than they are to a number of other environmental inputs. While RT-sealers were not eliminated as a potential source in some locations, forensic methods did not differentiate their contribution from other sources of PAHs, indicating RT-sealers are not a unique or readily quantifiable source of PAHs to the urban environment.

DeMott RP, Gauthier TD. 2014. Comment on "PAH Concentrations in Lake Sediment Decline Following Ban on Coal-Tar-Based Pavement Sealants in Austin, Texas." Environ. Sci. Technol. 48(23):14061–14062.

The timelines from sediment cores reported in the subject paper, other sampling by the City of Austin, and a similar observed pattern of decreasing PAH concentrations beginning in the 1990s at a Texas lake where there was no CTsealer ban, all demonstrate that attributing the entire reported decrease to the ban in Austin is an incomplete explanation of the available data. The similarity of PAH profiles for the lake sediments to non-CT related dust sources also suggests an alternative explanation warranting consideration. The authors have incorporated a degree of conclusiveness to their interpretation of the Austin sealer ban effects that overlooks other available information.

O'Reilly, K. 2014. Comment on "PAH Concentrations in Lake Sediment Decline Following Ban on Coal-Tar-Based Pavement Sealants in Austin, Texas." Submitted to Environ. Sci. Technol. At: <u>https://www.scribd.com/document/343814884/O-Reilly-K-2014-Comment-on-Van-Metre-and-Mahler-2014-PAH-Concentrations-in-Lake-Sediment-Decline-Following-Ban-on-Coal-Tar-Based-Pavement-Seala.</u>

The limited amount of data presented are insufficient to support the conclusions. The results of only two sediment cores that contain pre- and post-ban samples are included. In one case, the range of total PAH concentrations in the two post-ban samples is the same range as that for the last three pre-ban samples. In the other, the supposed decrease in PAH concentration began prior to the sealant ban. Two samples from four other locations were also discussed. The post-ban PAH concentrations were higher at one location and lower at the other three, but a trend cannot be determined with only two data points collected 12 years apart from each location. The paper also fails to mention that many of the sampling locations were adjacent to another source of PAHs, an oil fueled power plant that closed soon after the ban was enacted.

#### **Modeling Studies**

The fact sheet describes the use of source apportionment or receptor models. Receptor models are mathematical procedures for resolving one or more of these parameters in a mixed chemical system: 1) the number of sources, 2) their chemical characteristics, and 3) the relative contribution of each source in environmental samples. As with use of any mathematical model, understanding the assumptions that underlie the model is critical to interpreting the model results in a defensible manner. Key assumptions of least-squares-based receptor models such as the EPA'a chemical mass balance (CMB) used by Van Metre and Mahler are that:

- 6. All potential sources have been identified;
- 7. Source profiles are known and stable;
- 8. The number of sources is less than the number of fitting species;
- 9. Source profiles are linearly independent of each other; and
- 10. Measurement uncertainties are random, uncorrelated, and normally distributed.

As stated in the CMB guidance manual, these assumptions are fairly restrictive (Coulter 2004). While some deviations can be tolerated, they do raise some critical issues that should be considered in interpreting the results. There is an inherent conflict between the first assumption, which seeks to broaden the number of individual source types considered, and the third, which highlights the models' inability to accurately allocate among similar source profiles. Including all important sources is critical as the model can assign the contribution of missing sources to others used as inputs. The issue of collinearity of compound concentrations raised in the fourth assumption can be a particular challenge with pyrogenic PAHs, because different sources can have generally similar profiles (O'Reilly et al. 2012). Sources with similar chemical and physical properties cannot be distinguished from each other by CMB (Coulter 2004).

The ubiquitous nature of PAHs and the wide range of historical and ongoing combustion sources make it impossible to include all individual sources. As a result, source types are often grouped (Li et al. 2003), although grouping requires selection of a chemical profile that adequately represents the group (Galarneau 2008). PAH profiles are not unique, due to both intersource similarities and intrasource variability. Even at the point of emission or release, PAH chemistry varies with fuel type, oxygen levels, and combustion temperature. Furthermore, seasonal variations in emissions and atmospheric processes challenge the assumption of stability for atmospheric sources (Khairy and Lohmann 2013). Because both particle-specific PAH ratios and deposition rates differ by particle size, atmospheric PAH profiles of an individual source shift with distance from an emission source.

Weaknesses in using generic PAH profiles in source modeling has been long understood. In an EPA funded evaluation, Daisey et al. (1986) concluded that published data are not adequate for use in receptor modeling and it is necessary to measure profiles from major *local* sources. The authors noted that much of the data were collected for determining emission rates, not source profiles. Results typically exist only for a limited set of PAH compounds, and these have not been adequately measured in many sources of interest. Other composition variables—e.g., other classes of organics, trace elements, carbon, and particle mass—have rarely been measured simultaneously, which has hindered the development of receptor models for organic species.

Differences in the sampling and analytical methods between studies magnify their incompatibility for subsequent use in receptor models.

Summarizing these challenges, Dr. Elisabeth Galarneau, Environment Canada, developed criteria for the selection of PAH source profiles for use in models such as CMB (Galarneau 2008):

- 1. Potential source emissions data, whether used to identify sources a priori or a posteriori, should be representative of sources emitting to the ambient air under investigation, AND
- 2. The variation in relative species concentrations in the emissions of relevant sources should not render source identification ambiguous, AND
- 3. The uncertainty in measured concentrations in ambient air should not render source identification ambiguous, AND
- 4. All species examined should have similar reactivities with respect to atmospheric oxidants and solar radiation in both the gas and particle phases on the timescales of interest. Assessments of reactivity will be tentative since existing data show inconsistent differences for PAHs on model aerosol substrates and since gas-phase reactivities have not yet been measured for all commonly measured semivolatile PAHs, AND
- 5. All species examined should have similar Henry's law constants if air parcel histories involve time spent over water sufficient for diffusive air-water exchange to occur, AND
- 6. Temperature variations between source and downwind ambient air should be small enough for relative species concentrations to be unaffected by changes in particle/gas partitioning. Alternatively, only species with similar particle/gas partitioning characteristics should be examined if temperature variations are great between sources and downwind air, or only total (gas þ particle) concentrations should be examined, AND
- 7. All species examined should have particle size distributions that are similar enough to negate differences in particle scavenging by precipitation or particle dry deposition, OR
- 8. The source apportionment method used should account for all departures from the preceding guidelines.

These criteria have not been considered in the selection of inputs used in modeling exercises evaluating sealants as a potential PAH source. Most of the source profiles used by Van Metre and Mahler (2010) and others (Baldwin et al. 2017; Norris and Henry 2019) were initially published in Li et al. (2003) and have not been validated as accurately representing the sources claimed (O'Reilly et al. 2014, 2015). The purpose of Li's paper was to demonstrate how a receptor model could be applied at a specific site, not to generate profiles that should or could be generally applicable in all locations. Li used an untested multistep approach to combine multiple published data types in an attempt to construct PAH source profiles. Much of the underlying data was not originally published in a way that provided accurate information on PAH profiles. Because the published studies reported analyses of different combinations of PAHs, the data

sources for each profile varied between profile and PAHs. Such an approach loses information on the relative concentrations among PAHs that is a critical component of forensic analysis. The median coefficient of variation (CV=standard deviation/mean) of individual PAHs in Li's profiles is about 80%, suggesting the final profiles may not represent any actual source. As noted by Li,

[A] definitive signature of a combustion process may not exist due to the complexity of the combustion process. Emissions of PAHs depend on numerous factors which may vary significantly even during a single combustion process. In addition, sampling methods differ, introducing additional differences among published source signatures.

An elevated CV often prevents CMB from finding a solution. To prevent this issue, Li et al. (2003) and authors such as Van Metre and Mahler (2010) artificially set all CVs to 40%. This hides uncertainty in the underlying data and generates invalid model results.

Receptors models assume that source concentration profiles are stable so do not change over time. This is not true with PAHs. "Weathering" is a term used to describe a change in a chemical mixture due to its exposure to the environment. Volatilization, photoreactivity, and biological degradation cause PAHs to change concentrations at different rates, resulting in shifts in the chemical profile. Li's source profiles were generated from the analysis of unweathered samples collected at the point of emission, while Van Metre and Mahler (2010) used weathered parking lot dust samples to represent sealer source profiles. As described in O'Reilly et al. (2012), the use of weathered sealer samples and unweathered emission source samples as CMB inputs may have skewed the output toward a higher estimated sealer contribution. If the sealer receptor model input is replaced with the PAH profile of a fresh sealer sample, its source contribution drops to a few percent (O'Reilly et al. 2014; Zou et al. 2015).

The PAH profile used by Van Metre and Mahler to represent sealer is not unique to weathered sealer and may just represent urban background. The profiles identified as sealcoat are also essentially indistinguishable from urban soil, road runoff, or roof dust (Kay et al. 2003; Selbig 2009; Van Metre and Mahler 2003). Van Metre and Mahler (2003) presented data for seven PAHs from 22 samples collected from an Austin, Texas stream, rooftop runoff, and roadside dust. The median correlation between profiles was r=0.99 and the lowest was r=0.91. Correlations between the nine roof dust samples and the coal-tar sealcoat profiles used by Van Metre and Mahler (2003) and Norris and Henry (2019) ranged from r=0.93 to 0.99. These findings suggest a receptor model could not distinguish between the contributions of these sources.

Papers that highlight problems with the approaches used to model sealant PAH contributions include the following:

O'Reilly KT, Pietari J, Boehm P. 2014. Parsing Pyrogenic PAHs: Forensic Chemistry, Receptor Models, and Source Control Policy. Integr. Environ. Assess. Manag. 10:279–285.

As stated by Van Metre and Mahler (2010), their goal was to test the hypothesis that refined tar (RT) sealer is a major source of PAHs to urban lakes. Without inclusion of a negative control the published results provide little support for the

hypothesis. When using EPA's [CMB] model for hypothesis testing it is critical that the goodness of fit between measured and modeled concentrations be compared for runs with the source of interest and without that source. Comparing the results of the CMB model with results using a multiple lines of evidence forensic approach, we found the CMB output to be inconsistent with either diagnostic ratio analysis or principal component analysis (PCA). The expected relationship between the sealer contribution suggested by CMB and the ratios used to suggest sealer impacts is not observed. PCA was unable to separate weathered RT-sealer sources from the PAH profiles of other environmental particles, and the ranges overlapped between these and some sediment samples. The source profile identified as RT-sealer in the CMB model has not been found to be unique, sealer is not the only source with a similar profile which is a violation of the assumption that source profiles are linearly independent of each other. CMB output includes indicators of whether source collinearity impacts the final results. Care must be taken as the model will estimate source contributions even when it finds sources to be inestimable due to collinearity.

O'Reilly KT, Ahn S, Pietari J, Boehm P. 2015. Use of receptor models to evaluate sources of PAHs in sediments. Polycycl. Aromat. Compd. 35:41–56.

Receptor models can be powerful tools for assessing potential sources of PAHs in sediments. Due to similarities among pyrogenic PAH source profiles, characterizing the contributions of various sources remains a challenge. Application of receptor models requires careful consideration of the underlying assumptions and inherent uncertainties. Evaluation of a recent report using the receptor model CMB on data from stormwater detention pond sediments in Minnesota demonstrates the pitfalls involved in deviating from model assumptions. Model input choices included potential source profiles that are insufficient to explain the variability of the data and exacerbated the lack of independence and collinearity of the data set by excluding data based on a similarity screen. Use of a single receptor model contributed to unsupported conclusions about the identity and contributions of refined tar sealer as a PAH source. When using receptor models to test hypotheses concerning the role of specific sources, the scientific method requires that the null hypothesis be included and that negative controls be applied. Communication of uncertainties associated with model results requires transparency concerning the precision and accuracy of inputs and outputs, as well as conveying an understanding of the range of possible results given choices of input parameters.

O'Reilly KT, Ahn S. 2017. Comment on "Primary sources and toxicity of PAHs in Milwaukeearea streambed sediments." Submitted to Environ. Toxicol. Chem.

The data presented by Baldwin et al. (2017) does not support the paper's conclusions. The evaluation is based on the circular logic of creating a source profile identified as representing RTS to be similar to urban background and then using statistical measurements of that similarity as evidence of causation. By relying on the approach and source profiles of Van Metre et al. (2010), Baldwin et al. obtained a preordained but incorrect outcome. Our evaluation demonstrates that

urban soils have sufficient PAHs to account for those measured in urban sediments and that additional validation of all potential source profiles is necessary if they are to be used to assess local contributions.

O'Reilly KT, Edwards M. 2019. Letter to the Editor: Comment on Norris and Henry (2019), Science of the Total Environment. 135248, ISSN 0048-9697, https://doi.org/10.1016/j.scitotenv.2019.135248.

Accurate and valid source PAH profiles are essential whether they are used as inputs to a mixing model, such CMB, or used to interpret output from an unmixing model like Unmix Optimum (UnmixO). Norris suffers from the same fatal flaws as prior papers (Baldwin et al. 2017; Van Metre and Mahler 2010) that used essentially the same source profiles to evaluate whether refined tar sealers (RTS) are a source of sediment PAHs.

As with all un-mixing results, conclusions about actual source contributions rely on interpretation of the similarity between the modeled composition profiles and actual potential source profiles. It is critical to measure the similarity between a variety of sources, as opposed to only the source(s) of interest, because the correlation values may be similar for multiple sources. Depending on the number of PAHs and the variety of samples included in the analysis, there may not be sufficient information to conclusively distinguish the best matching source. Most of the PAH profiles used in Norris to represent sources are mathematical constructs that have never been validated as accurately representing the sources claimed. The profiles identified as RTS are also essentially indistinguishable from urban soil, road runoff, or roof dust. Due the lack of valid and accurately identified source profiles, Norris and Henry (2019) does not provide new evidence that RTS is an important source of PAHs in urban sediments.

In conclusion, there are numerous technical issues with the PAH source modeling exercises cited in the Proposed 2020 MSGP Fact Sheet. The papers overstate the contribution of sealants and understate uncertainty associated with the modeling results.

## Acknowledgement

These comments were developed with funding from the Pavement Coatings Technology Council.

## References

Baldwin A, Corsi S, Lutz M, Ingersoll C, Dorman R, Magruder C, Magruder M. 2017. Primary sources and toxicity of PAHs in Milwaukee-area streambed sediments. Environ. Toxicol. Chem. 36:1622–1635.

Blumer M, Youngblood WW. 1975. Polycyclic Aromatic Hydrocarbons in Soils and Recent Sediments. Science 188:53–55.

Coulter, CT. 2004. EPA-CMB8.2 Users Manual. EPA-452/R-04-011. U.S. Environmental Protection Agency, Office of Air Quality Planning & Standards, Emissions, Monitoring & Analysis Division, Air Quality Modeling Group. Research Triangle Park, NC. December.

Daisey JM, Cheney JL, Lioy PJ. 1986. Profiles of organic particulate emissions from air pollution sources: status and needs for receptor source apportionment modeling. J. Air Pollut. Control Assoc. 36:17–33.

DeMott RP, Gauthier TD. 2006. Comment on "Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons." Environ. Sci. Technol. 40:3657–3658.

DeMott RP, Gauthier TD. 2014. Comment on "PAH Concentrations in Lake Sediment Decline Following Ban on Coal-Tar-Based Pavement Sealants in Austin, Texas." Environ. Sci. Technol. 48:14061–14062.

Galarneau E. 2008. Source specificity and atmospheric processing of airborne PAHs: Implications for source apportionment. Atmos. Environ. 42:8139–8149.

Hwang HM, Foster GD. 2006. Characterization of polycyclic aromatic hydrocarbons in urban stormwater runoff flowing into the tidal Anacostia River, Washington, DC, USA. Environ. Pollut. 140:416–426.

Kay R, Arnold T, Cannon W, Graham D, Morton E, Bienert R. 2003. Concentrations of Polynuclear Aromatic Hydrocarbons and Inorganic Constituents in Ambient Surface Soils, Chicago, Illinois: 2001-02. USGS, Water-Resources Investigations Report 03-4105.

Khairy M, Lohmann R. 2013. Source apportionment and risk assessment of polycyclic aromatic hydrocarbons in the atmospheric environment of Alexandria, Egypt. Chemosphere 91:895–903.

Li A, Jang J-K, Scheff PA. 2003. Application of EPA CMB8.2 model for source apportionment of sediment PAHs in Lake Calumet, Chicago. Environ. Sci. Technol. 37:2958–2965.

Lima ALC, Farrington JW, Reddy CM. 2005. Combustion-derived polycyclic aromatic hydrocarbons in the environment—A review. Environ. Forensics 6:109–131.

Mahler BJ, Van Metre PC, Bashara TJ, Wilson JT, Johns DA. 2005. Parking lot sealcoat: An unrecognized source of urban polycyclic aromatic hydrocarbons. Environ. Sci. Technol. 39:5560–5566.

Mastral A, Callén M. 2000. A Review on Polycyclic Aromatic Hydrocarbon (PAH) Emissions from Energy Generation. Environ. Sci. Technol. 34:3051–3057.

New York Academy of Sciences. 2007. Pollution Prevention and Management Strategies for Polycyclic Aromatic Hydrocarbons in the New York/New Jersey Harbor.

Norris GA, Henry RC. 2019. Unmix Optimum analysis of PAH sediment sources. Sci. Total Environ. 673:831–838.

O'Reilly, K. 2014. Comment on "PAH Concentrations in Lake Sediment Decline Following Ban on Coal-Tar-Based Pavement Sealants in Austin, Texas." Submitted to Environ. Sci. Technol. At: <u>https://www.scribd.com/document/343814884/O-Reilly-K-2014-Comment-on-Van-Metre-and-Mahler-2014-PAH-Concentrations-in-Lake-Sediment-Decline-Following-Ban-on-Coal-Tar-Based-Pavement-Seala.</u>

O'Reilly KT, Ahn S. 2017. Comment on "Primary sources and toxicity of PAHs in Milwaukeearea streambed sediments." Submitted to Environ. Toxicol. Chem.

O'Reilly KT, Edwards M. 2019. Letter to the Editor: Comment on Norris and Henry (2019). Sci. Total Environ. 135248, ISSN 0048-9697, https://doi.org/10.1016/j.scitotenv.2019.135248.

O'Reilly KT, Pietari J, Boehm PD. 2011. Comment on "PAHs Underfoot: Contaminated Dust from Coal-Tar Sealcoated Pavement is Widespread in the U.S." Environ. Sci. Technol. 45:3185–3186.

O'Reilly KT, Pietari J, Boehm PD. 2012. A forensic assessment of coal tar sealants as a source of polycyclic aromatic hydrocarbons in urban sediments. Environ. Forensics 13:185–196.

O'Reilly KT, Pietari J, Boehm P. 2014. Parsing Pyrogenic PAHs: Forensic Chemistry, Receptor Models, and Source Control Policy. Integr. Environ. Assess. Manag. 10:79–285.

O'Reilly KT, Ahn S, Pietari J, Boehm P. 2015. Use of receptor models to evaluate sources of PAHs in sediments. Polycycl. Aromat. Compd. 35:41–56.

Ravindra K, Sokhi R, Van Grieken R. 2008. Atmospheric polycyclic aromatic hydrocarbons: Source attribution, emission factors and regulation. Atmos. Environ. 42:2895–2921.

Selbig WR. 2009. Concentrations of polycyclic aromatic hydrocarbons (PAHs) in urban stormwater, Madison, Wisconsin, 2005–08: U.S. Geological Survey Open-File Report 2009 1077, 54 pp.

Simcik MF, Eisenreich SJ, Lioy PJ. 1999. Source apportionment and source/sink relationships of PAHs in the coastal atmosphere of Chicago and Lake Michigan. Atmos. Environ. 33:5071–5079.

Stein ED, Tiefenthaler LL, Schiff K. 2006. Watershed-based sources of polycyclic aromatic hydrocarbons in urban storm water. Environ. Toxicol. Chem. 25(2):373–385.

Stout SA, Uhler AD, Emsbo-Mattingly SD. 2004. Comparative evaluation of background anthropogenic hydrocarbons in surficial sediments from nine urban waterways. Environ. Sci. Technol. 38(11):2987–2994.

Su M-C, Christensen ER, Karls JF, Kosuru S, Imamoglu I. 2000. Apportionment of polycyclic aromatic hydrocarbon sources in lower Fox River, USA, sediments by a chemical mass balance model. Environ. Toxicol. Chem. 19:1481–1490.

Tsai P, Hoenicke R, Yee D, Bamford HA, Baker JE. 2002. Atmospheric concentrations and fluxes of organic compounds in the northern San Francisco Estuary. Environ. Sci. Technol. 36:4741–4747.

U.S. EPA. 2007. Survey of New Findings in Scientific Literature Related to Atmospheric Deposition to the Great Waters: Polycyclic Aromatic Hydrocarbons. EPA-452/R-07-011.

Van Metre PC, Mahler BJ. 2003. The contribution of particles washed from rooftops to contaminant loading to urban streams. Chemosphere 52:1727–1741.

Van Metre PC, Mahler BJ. 2005. Trends in Hydrophobic Organic Contaminants in Urban and Reference Lake Sediments across the United States 1970–2001. Environ. Sci. Technol. 239:5567–5574.

Van Metre PC, Mahler BJ. 2010. Contribution of PAHs from coal-tar pavement sealcoat and other sources to 40 U.S. lakes. Sci. Tot. Environ. 409:334–344.

Van Metre PC, Mahler BJ, Furlong ET. 2000. Urban sprawl leaves its PAH signature. Environ. Sci. Technol. 34:4064–4070

Van Metre PC, Mahler BJ, Wilson JT. 2009. PAHs underfoot: Contaminated dust from coal-tar sealcoated pavement is widespread in the United States. Environ. Sci. Technol. 43:20–25.

Washington State Department of Ecology. 2011. Control of Toxic Chemicals in Puget Sound: Assessment of Selected Toxic Chemicals in the Puget Sound Basin 2007–2011. Ecology Publication No. 11-03-055.

Youngblood WW, Blumer M. 1975. Polycyclic aromatic hydrocarbons in the environment: homologous series in soils and recent marine sediments. Geochimica et Cosmochimica Acta 39:1303–1314.

Yunker MB, Macdonald RW, Vingarzan R, Mitchell RH, Goyette D, Sylvestre S. 2002. PAHs in the Fraser River basin: A critical appraisal of PAH ratios as indicators of PAH source and composition. Organ. Geochem. 33:489–515.

Zou Y, Wang L, Christensen E. 2015. Problems in the Fingerprints Based Polycyclic Aromatic Hydrocarbons Source Apportionment Analysis. Environ. Pollut. 205:394–402