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April 13, 2016

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Re: Request for Reconsideration of RFC-14003 under the EPA Information Quality Guidelines/Allegation of Loss of Scientific Integrity

The Pavement Coatings Technology Council (PCTC) represents numerous companies throughout the country that are part of the sealcoat industry. On behalf of PCTC, I write to submit a Request for Reconsideration (RfR) of the Request for Correction (RfC) submitted by PCTC on April 16, 2014, which was designated RFC-14003 by EPA. According to EPA's Information Quality Guidelines (IQGs)[1], those who are dissatisfied with EPA's response[2] to an RfC may file an RfR. PCTC is not only dissatisfied -- it is disappointed that, after almost 2 years, the Agency's response was so unsubstantial. EPA did not deal with the merits of the RfC's arguments. Rather, EPA only made general assertions that it followed appropriate processes and relied upon sufficiently authoritative sources. These assertions cannot, however, rebut the substantive arguments made in the RfC and in this document.

As suggested in the IQGs, this RfR is being submitted within 90 days of EPA's January 19, 2016 response to the RfC. EPA's response to RFC-14003 is included here as Attachment 1. The original RfC is also included as Attachment 2. In that RfC, Exhibit D was a listing of PCTC-

[1] *Guidelines for Ensuring and Maximizing the Quality, Objectivity, Utility, and Integrity of Information Disseminated by EPA*, October, 2002; https://www.epa.gov/sites/production/files/2015-08/documents/epa-info-quality-guidelines.pdf

[2] Beauvais, J., Burke, T.A. (2016). Letter to Anne P. LeHuray responding to Request for Correction (RfC) submitted April 16, 2014, on behalf of the Pavement Coatings Technology Council. 8 pages, Jan. 19. RfC assigned EPA No. RFC 14003. Available at https://www.epa.gov/quality/epa-information-quality-guidelines-requests-correction-and-requests-reconsideration#14003

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sponsored scientific publications on the topic of refined coal tar-based pavement sealant (RTS) in the environment. Attachment 3 is an updated listing of those publications. Attachment 4 is a letter to Alan D. Thornhill of the U.S. Geological Survey (USGS) informing the Survey that Post-Publication Peer Review (PPPR) documents for 16 papers authored by USGS and non-USGS scientists have been posted on the PPPR website, PubPeer.com. Attachments 2, 3, and 4 are incorporated by reference in their entirety into this RfR.

By this document, PCTC also makes an allegation of the loss of scientific integrity. In this case, the information that EPA is disseminating is based on materials that meet the criteria the Committee on Publication Ethics (COPE) has established for retraction of scientific articles; i.e., it relies on publications with "clear evidence that the findings are unreliable." A letter to the editor of *Environmental Science & Technology* that explains this allegation in more detail is included as Attachment 5, and is incorporated by reference into this RfR. Similar letters requesting retraction of the papers relied on by the Agency in preparation of its CADDIS information and Stormwater BMPs could (and will, depending the outcome of USGS deliberations) be prepared for many, if not most, of the RTS papers authored by USGS employees or the scientists who have attempted to build on the USGS work.

An unbiased science review of the body of work by the USGS RTS research team (see PPPRs referred to in Attachment 4) leads to one conclusion -- that the papers constitute advocacy research (LeHuray, 2015). It is the responsibility of the USGS Science Integrity Officer (SIO) to protect the reputation of the Survey as a provider of unbiased and policy neutral information, as well as to ensure the integrity of the scientific record. That official should accordingly withdraw all of the Survey's RTS publications, including the publications relied on by EPA in preparation of its CADDIS example (identified as "EPA Publication #1" in Attachment 2) and its stormwater BMP (identified as "EPA Publication #2" in Attachment 2). Should that happen, scientific integrity considerations mandate that EPA withdraw EPA documents that use the USGS RTS papers for an example of causal analysis and the RTS stormwater BMP.

But even if USGS does not take that step, EPA must withdraw and revise the subject documents for the reasons explained in the RfC and (again) below. Once those documents are withdrawn, PCTC looks forward to working with EPA on developing causal analysis and PAH stormwater BMPs that comport with sound science.

OVERVIEW OF THIS RFR

The RfR is organized according to the Agency's responses to RFC-14003. Agency responses have been categorized as follows:

1. Responses to Concerns about the Quality and Integrity of Scientific Information
   - 1.1 Fitness of the information for its purpose
   - 1.2 Reliance on NRC (2009)
   - 1.3 Use of PAHs as an example

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1.4 Peer Reviewed, Published Critical Commentary Should Trigger Predissemination Review
2. Acknowledgement of Matters of Fact Resulting in Edits to Documents
3. Explanation of EPA Intentions and Definitions
4. Statement Concerning the Absence of Transparency

The RfR concludes with a section on recommended corrective actions.

1. Responses to Concerns about the Quality and Integrity of Scientific Information

1.1: Fitness of the information for its purpose

In its response to RfC-14003, the Agency states that the intended use of CADDIS is to help scientists and engineers in diverse government settings conduct causal assessments of impacts that may be related to substances introduced in aquatic systems. The intended use of stormwater BMPs is "to provide Municipal Separate Storm Sewer System (MS4) operators with examples of practices that can be used to successfully achieve the minimum control measures required under EPA's stormwater regulations."

For reasons that remain unclear, both intended uses result in identification of USGS studies of RTS as examples, even though

- it had been known in the scientific literature available at the time the CADDIS example was prepared that there were no impacts in studied aquatic systems that could be attributed to (or even associated with) RTS (TCEQ, 2003a, 2003b, Scoggins et al., 2007), and
- PCTC is not aware that there is any federal requirement or recommendation that either coal tar-derived materials or PAHs should be considered in MS4 permits.

The Agency cannot assert that a causal analysis of an aquatic system where no impacts related to the example have been demonstrated is fit for purpose. The Agency also cannot assert that an MS4-driven BMP for a class of materials that are not subject to MS4 permit controls is fit for purpose.

1.2: Reliance on NRC (2009)

EPA's response states: "EPA's primary source for the information in the stormwater BMP fact sheet was the National Research Council's report *Urban Stormwater Management in the United States*" (NRC (2009)). This report seems to be, essentially, a literature review without a critical element (systematic review, perhaps) to it. In the report's discussion of PAHs, the New York Academy of Sciences (NYAS) New York-New Jersey Harbor Study (Valle et al., 2007) was not cited. That study included all purported sources of PAHs identified by the NYAS, and the data and findings of the USGS RTS research team was included. PCTC presented evidence at the time the report was written that the NYAS used unjustifiably inflated numbers

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for RTS, but even so, Valle et al. (2007) concluded that only about 0.4% of the PAHs associated with sediments in NY-NJ Harbor were attributable to RTS.

Washington State's Department of Ecology (WaDOE) conducted a study similar to the NY-NJ Harbor study for sediments in Puget Sound (Norton et al., 2011). As in NY, the USGS estimates of RTS contributions of PAHs to sediments were included in the analysis. Again, PCTC believes the values used were too high. Nevertheless, Norton et al. (2011) concluded that between 0.2 and 1.7% of PAHs associated with sediments in Puget Sound were attributable to RTS.

While PCTC has not fully evaluated and does not endorse the source apportionment conclusions reached by either the NYAS or the WaDOE, the values seem more in line with the vast majority of PAH source identification and apportionment literature, including data available in EPA's National Emission Inventory and ATSDR's PAH profile document.

The Agency cannot rely on NRC (2009) as justification that RTS is the source of PAHs for causal analysis and stormwater BMP examples when that report ignored an important contrary conclusion and when more recent researchers who have evaluated PAH source identification and apportionment in sediments have found it to be a minor – if not *de minimis* – source.

1.3: Use of PAHs as an Example

In its response to RfC-14003, the Agency describes CADDIS users as qualified to make decisions about which hypotheses should be included in a causal assessment of impacts on aquatic systems. The Agency then proceeds to provide only one example of a potential PAH source without questioning whether *any* evaluation of PAH sources using CADDIS is a worthwhile exercise.

EPA is a large organization. What is known about PAHs in one part of the organization may not be understood by others within EPA, much less those in other government agencies.

Through long experience at legacy waste sites, EPA's Office of Research & Development has found that PAH concentrations in sediments often do not correlate with toxicity. That office has developed a sophisticated tiered approach to evaluating PAH toxicity in sediments (EPA, 2003; Burgess, 2009) which has been widely demonstrated to be effective (e.g., Geiger, 2011; Kane Driscoll et al., 2010; Neuhauser et al., 2006). That approach, and not CADDIS, should be used in any causal analysis of PAH impacts in aquatic systems.

1.4: Peer Reviewed, Published Critical Commentary Should Trigger Predissemination Review

In its response to RfC-14003, EPA declines to cite peer reviewed publications that are critical of the peer reviewed publications determined to be of "quality, objectivity, and

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transparency … sufficient for their intended uses." The critiques, however, are not trivial, but question the reproducibility of the field-based study, the data used to identify PAH sources, and the verifiability of the modeling results.

The Agency values science with the goal of integrating "the highest quality science into the Agency's policies and decisions.[3]" Peer review, critical commentary and PPPR are recognized as vital elements in ensuring the quality of science. EPA's response to RfC-14003 includes no technical discussion of either the USGS publications or the publications critical of the USGS work. There is no scientific discussion of the process used in finding the USGS publications of sufficient "quality, objectivity and transparency," nor any finding of flaws in the critical publications. The decision to exclude recognition of peer reviewed publications critical of science that is being used as positive examples of EPA-recommended methodologies is at odds with ensuring quality and objectivity. In this particular case, the authors of the studies being used as examples have long resisted transparency.

The Agency cannot fail to recognize that peer reviewed and published critical commentary on a publication warrants recognition and review before disseminating Agency information and communication products based on the critiqued publication.

2. Acknowledgement of Matters of Fact Resulting in Edits to Documents

PCTC acknowledges the edits EPA has made to its information products regarding PAHs. That said, after the edits neither the CADDIS causal analysis example nor the stormwater BMP has improved in conveying to users the context in which PAHs occur and may (or may not) pose undue risks in the environment. Indeed, EPA guidance on determining potential aquatic risks related to PAHs in sediments (EPA, 2003; Burgess, 2009) should be used for this purpose instead of CADDIS, and a PAH stormwater BMP appears to be irrelevant to the MS4 permitting process.

The Agency's attempt to use PAHs as an example of either sediment toxicity or human health impacts is ambitious, but risk assessment of PAHs is a minefield of exceptions and complexities that must be explained. A few of the facts that the Agency should consider for any assessment of PAHs or PAH-containing substances in the environment include:

- Studies attempting to identify and apportion the multiple sources of PAHs in sediments (such as the NY-NJ harbor study and the Puget Sound study) have identified wood-burning fireplaces and stoves as the greatest source of PAHs; RTS has been identified as a minor source in these studies;
- It is EPA's stated policy to assess risks of PAH-containing substances on the basis of available toxicological information about each PAH-containing substance, in preference

[3] https://www.epa.gov/osa

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to using an additive approach that relies on simple addition of toxicity factors of individual PAH compounds relative to the index compound, benzo(a)pyrene (B(a)P);

- EPA's Science Advisory Board has expressed doubts about how well-grounded any additive approach involving PAHs may be in science, as factors such as bioaccessibility, synergism, and antagonism are known to be important, but are not yet fully understood;
- The currently promulgated IRIS document for B(a)P is acknowledged by the Agency to be out dated, with an oral cancer slope factor that is overstated by about an order of magnitude;
- The implications of the designation of coal tar by the US Food and Drug Administration (FDA) as "generally safe and effective for use in over-the-counter" products directly on human skin (21 CFR 358) should be addressed. FDA reviewed its Effects Reporting System for adverse reports in the early 2000s, and found no reason to change its designation of coal tar.
- In the specific case of materials such as RTS, which is a physical mixture containing refined metallurgical grade coal tar, the Agency must explain the RCRA status of coal tar. Coal tar is not a hazardous waste, both by rule (EPA, 1992) and by RCRA criteria (EPA, 2000), and, as such, there are no land disposal restrictions at the federal level;
- The Agency should explain the differences and similarities of coal tar such as found at legacy manufactured gas plant (MGP) waste sites and refined metallurgical grade coal tar such as is used in the manufacture of RTS.

3. Explanation of EPA Intentions and Definitions

PCTC is glad to learn that the Agency is conducting further research with the goal of publishing an independent evaluation of the USGS studies using methods recommended in O'Reilly et al. (2014a). EPA may not be aware that O'Reilly et al. (2014a) was written prior to receiving sufficient data from the USGS (in partial response to a Freedom of Information Act request) to partially reconstruct the USGS inputs and configuration of the Agency's Chemical Mass Balance model. The Agency may want to consider O'Reilly et al. (2015) which took that information into account, as well as the technical memorandum (O'Reilly, 2014b), included in this RfR as Attachment 2, Exhibit E. We would like to advise the Agency that, because USGS has failed to fully comply with its FOIA request, PCTC has brought a lawsuit against the USGS seeking production of the data and documents USGS continues to withhold from PCTC. Based on USGS's descriptions in its Vaughn Index of withheld files, some of the specific data still being withheld includes modeling data. The Agency should also inform the scientists who have been assigned this task about of the comprehensive and detailed PPPRs incorporated into this RfR via Attachment 4.

In its response, the Agency states that it is aware of some recent publications by the USGS RTS research team. PCTC believes that these most recent publications are part of the same advocacy research effort as other publications by the same authors. EPA should consider

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the PPPRs of all the USGS papers, including the most recent ones in its evaluation of the science concerning RTS as well as in updates of current CADDIS and BMP information products.

4. Statement Concerning the Absence of Transparency

The Agency may not have intentionally excluded PCTC from its consideration of using the USGS research team's RTS studies as an example of causal analysis or development of stormwater BMPs. That said, PCTC is not aware that the Agency included any company or industry group in its development processes. PCTC believes this exclusion violates the stated purpose and spirit of the causal analysis effort as well as the stormwater BMP development process.

Recommended Corrective Actions

First, as requested in RfC-14003, EPA should remove the CADDIS example and the stormwater BMP from its websites, immediately, so that appropriate amendments and updates can be implemented with input not only from the USGS, but from industry as well, such as the PCTC.

Second, EPA should draft updated and unbiased summaries regarding the science that now exists with respect to RTS. An important part of that process will be the inclusion of Reference sections and links that allow members of the public to understand the nature and complexities of the scientific debates that are ongoing. The Agency should immediately remove the CADDIS Pavement Sealants and PAHs webpage. Once this is done, the PCTC will gladly participate in any future discussions that the EPA may wish to have geared toward creating an accurate and unbiased webpage that outlines the issues presently being debated regarding RTS. The new webpage must be based on all of the relevant investigations and peer reviewed publications, not just studies generated by the USGS and the City of Austin and the "ban RTS" affinity group that has developed over time.

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PCTC is a 501(c)(6) trade association the members of which are predominantly family-owned small businesses. PCTC members are dismayed that the agenda and advocacy science of a few government employees has gained so much traction within what, to PCTC members, can only be described as behemoth federal agencies such as the USGS and EPA. That said, PCTC is proud to have approached the issues raised by these agencies rationally, using sound science and making the results public for all to see. PCTC requests that EPA also approach the issues raised

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rationally and with transparency. To this end, PCTC welcomes the opportunity to meet with EPA management and its scientists assigned to work on RTS issues.

Please don't hesitate to contact me for additional information,

Very truly yours

Anne P. LeHuray, Ph.D.
Executive Director

cc: Alan D. Thornhill, USGS Science Integrity Officer, athornhill@usgs.gov
William Werkheiser, Acting Deputy Director USGS whwerkhe@usgs.gov
William Guertal, Acting Associate Director Water Resources, USGS wguertal@usgs.gov

List of Attachments

1. Beauvais, J., Burke, T.A. (2016). Letter to Anne P. LeHuray responding to Request for Correction (RfC) submitted April 16, 2014, on behalf of the Pavement Coatings Technology Council. 8 pages, Jan. 19. RfC assigned EPA No. RFC 14003. Available at https://www.epa.gov/quality/epa-information-quality-guidelines-requests-correction-and-requests-reconsideration#14003
2. LeHuray, Anne P. (2014). Request for Correction (RfC) under the EPA Information Quality Guidelines. submitted April 16, 2014, on behalf of the Pavement Coatings Technology Council. 60 pages. RfC assigned EPA No. RFC 14003. Available at https://www.epa.gov/quality/epa-information-quality-guidelines-requests-correction-and-requests-reconsideration#14003. Includes Exhibits A through F, as follows:

   Exhibit A: CADDIS: The Causal Analysis/Diagnosis Decision Information System Volume 2: Sources, Stressors & Responses http://www.epa.gov/caddis/ssr_urb_intro.html

   Exhibit B: EPA Publication #1 - CADDIS Volume 2: Sources, Stressors & Responses: Pavement sealants & PAHs & References for the Urbanization Module http://www.epa.gov/caddis/ssr_urb_wsq4.html & http://www.epa.gov/caddis/ssr_urb_ref.html

   Exhibit C: EPA Publication #2 - STORMWATER BEST MANAGEMENT PRACTICE: Coal-Tar Sealcoat, Polycyclic Aromatic Hydrocarbons, and Stormwater Pollution. EPA 833-F-12-004, November 2012 http://www.epa.gov/npdes/pubs/coaltar.pdf

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Exhibit D: PUBLICATIONS OF SCIENTIFIC STUDIES OF REFINED TAR-BASED PAVEMENT SEALERS (RTS) IN THE ENVIRONMENT SPONSORED BY THE PAVEMENT COATINGS TECHNOLOGY COUNCIL (REV. APRIL 2014)

Exhibit E: TECHNICAL EVALUATION OF VAN METRE AND MAHLER (2010). Prepared by Kirk T. O'Reilly, Ph.D. J.D., Exponent, Inc., March 25, 2014

Exhibit F: Letter from Myron O. Knudson, Director, Superfund Division, EPA Region 6 to Ms. Toby Hammett Futrell, City Manager, City of Austin, TX dated April 17, 2003

3. Publications of Scientific Studies of Tar-Based Sealants in the Environment Sponsored by the Pavement Coatings Technology Council (rev. April 9, 2016)
4. LeHuray, Anne P. (2016). Letter to Alan D. Thornhill, Scientific Integrity Officer, U.S. Geological Survey, dated March 3, 2016.
5. LeHuray, Anne P. (2015). Letter to David L. Sedlak, Editor, *Environmental Science & Technology*, dated November 20, 2015.

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UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
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JAN 19 2016

Dear Dr. LeHuray:

This letter responds to your Request for Correction (RFC) of information pursuant to the *Guidelines for Ensuring and Maximizing the Quality, Objectivity, Utility, and Integrity of Information Disseminated by the Environmental Protection Agency* (EPA Information Quality Guidelines) submitted on behalf of the Pavement Coatings Technology Council (PCTC) and received on April 16, 2014.

In the RFC, you raised a number of issues with respect to the objectivity, transparency, and reproducibility of information included on the EPA webpage titled "EPA CADDIS Volume 2: Sources, Stressors & Response - Pavement sealant & PAHs" (CADDIS webpage) and the November 2012 EPA fact sheet titled "Coal-Tar Sealcoat, Polycyclic Aromatic Hydrocarbons, and Stormwater Pollution" (stormwater BMP fact sheet). You requested that EPA remove both of these publications from its website to make modifications including adding additional information and studies.

The Agency uses a graded approach and well-established Agency policies and procedures as appropriate for each of its information products. The Agency employed an internal review process involving Agency management and a communications product review process to establish the appropriate quality, objectivity, utility, and integrity of these products based on the intended use of the information and the resources available.

The intended use of the website Causal Analysis/Diagnosis Decision Information System, or CADDIS, is to help scientists and engineers in the EPA Regions, states, and tribes conduct causal assessments in aquatic systems. Providing this information assists our state and tribal partners with implementing their local environmental programs. Scientists and engineers are a technical audience that can use their awareness of unique or local inputs and supplement them with specific concerns or directly applicable data as they employ the process and background information. CADDIS instructions recommend that users engage with stakeholders and decision makers and use all relevant information in their scientific evaluation process.

The "Coal-Tar Sealcoat, Polycyclic Aromatic Hydrocarbons, and Stormwater Pollution" fact sheet is one of numerous stormwater BMP fact sheets that EPA has developed and made available on its "Stormwater Menu of BMPs" webpage to provide information on preventing discharges of pollutants into storm sewer systems and the Nation's waters. The intended purpose of the "Stormwater Menu of BMPs" webpage is to provide Municipal Separate Storm Sewer System (MS4) operators with examples of practices that can be used to successfully achieve the minimum control measures required under EPA's stormwater regulations. Each of EPA's stormwater BMP fact sheets is consistent with the EPA Communication Stylebook, which states that "Fact sheets are used to provide information about an issue, project or activity to someone who might have limited knowledge of the subject. They should be limited to one or two pages in length and focus on highlights or the issues of highest importance." The Stylebook further states, "Assume the reader will not have a technical understanding of the issues at hand; write as if presenting a talk to non-experts, not for a journal article."

Regarding your comment on page 3, paragraph 4 of your RFC that coal-tar sealcoat manufacturers and appliers consider the information in the CADDIS webpage and stormwater BMP fact sheet to be influential, EPA disagrees. EPA generally considers influential information for the IQGs to be information disseminated in support of top Agency actions (i.e., rules, substantive notices, policy documents, studies, guidance); information disseminated in support of Economically Significant actions as defined in Executive Order 12866, entitled Regulatory Planning and Review (58 FR 51735, October 4, 1993); or major work products undergoing peer review as called for under the Agency's Peer Review Policy. Neither the CADDIS webpage nor the stormwater BMP fact sheet meet any of these categories, and therefore are not considered to be influential.

EPA also notes that it did not deliberately exclude the PCTC from the development of these products, as you state on page 6, paragraph 3 of your RFC. EPA encourages comments on the information in both products at any time, as described on EPA's stormwater menu of BMPs homepage and on the CADDIS contact page (see http://www.epa.gov/npdes/national-menu-best-management-practices-bmps-stormwater and http://www.epa.gov/caddis/caddis_contact.html).

EPA acknowledges that there are multiple sources of PAHs in the environment, as you noted in your RFC. The CADDIS webpage currently lists sources of PAHs other than coal-tar sealcoats in urban waterways. To clarify the broader scope, the CADDIS webpage will be modified so that the title reads "PAHs" rather than "Pavement Sealants and PAHs." When this edit has been published at the time our pending website migration is complete, you will be notified by the Information Quality Guidelines Processing staff. EPA has also inserted a new opening section of the stormwater BMP fact sheet identifying natural and man-made sources of PAHs, and moved the paragraph describing the concerns related to PAHs into this opening section.

Your RFC raised concerns with the following statement in the stormwater BMP fact sheet: "PAHs are a concern because of their harmful impacts on humans and the environment. They are persistent organic compounds, and several PAHs are known or probable human carcinogens and toxic to aquatic life." One of EPA's responsibilities is to provide information to the regulated community and the public about known human and environmental risks associated with chemicals and compounds, including PAHs.

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EPA acknowledges that not all PAHs have been analyzed for their harmful effects to humans and the environment. Therefore, EPA has retained this statement with one minor modification: "*Many* PAHs are of concern because of their harmful impacts on humans and the environment," [emphasis added]. EPA also added in-text citations to the fact sheet for these statements. The modified fact sheet is posted on EPA's website and is included as an attachment to this letter.

You requested that EPA remove from the stormwater BMP fact sheet a list of states and municipalities that have taken action on coal-tar sealcoat. EPA routinely provides information to the public and interested stakeholders about activities that local governments are taking to address stormwater pollution. Therefore, EPA retains this section as-is.

You also raised concerns with the validity of the U.S. Geological Survey (USGS) studies, as well as a study by Scoggins et al. (2007), that EPA cites on the CADDIS webpage and in the stormwater BMP fact sheet. EPA is aware that there have been on-going requests for correction to the USGS' studies from the PCTC under the USGS' Information Quality Guidelines, and that these requests have been responded to point-by-point in several letters from the USGS. The Agency's evaluation of the USGS studies determined that the quality, objectivity, and transparency is sufficient for their intended uses. EPA is therefore retaining the references to these studies in its publications.

Finally, you requested that EPA consider amending the CADDIS webpage and stormwater BMP fact sheet to include references to several studies funded by the PCTC and listed in Appendix D of the RFC. EPA declines to modify the EPA CADDIS webpage or the stormwater BMP fact sheet to include references to the PCTC-sponsored studies which question the validity of findings by the USGS and others that coal-tar sealcoat is a significant source of PAHs in the environment. As stated earlier, EPA determined the USGS studies cited in the CADDIS webpage and stormwater BMP fact sheet to be sufficient for the intended use. Additionally, EPA has conducted its own research on this topic, and a study that was subject to the Agency's peer and administrative review found that coal-tar sealcoat releases 100 to 1,000 times more PAHs than other types of surfaces (EPA, 2011).

EPA notes that the CADDIS webpage and the stormwater BMP fact sheet were not intended to be a comprehensive source of all literature related to the topic of coal-tar sealcoat and stormwater. EPA's primary source for the information in the stormwater BMP fact sheet was the National Research Council's report *Urban Stormwater Management in the United States*, which is a review of the EPA stormwater permitting program that includes suggestions for improvement (National Research Council, 2009). This report was approved by the Governing Board of the National Research Council, with members from the councils of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine. EPA's stormwater BMP fact sheet went through Agency review appropriate for its intended audience, including various levels of technical and communications review within EPA's Office of Water, and approval by OW management. Further, EPA includes the following caveat on its stormwater menu of BMP homepage: "The BMP examples and references included on these fact sheets are not intended to be comprehensive. Additionally, the list of BMPs is not all-inclusive, and it does not preclude MS4s from using other technically sound practices." EPA has included a reference to this disclaimer on the revised stormwater BMP fact sheet.

As the CADDIS homepage states, CADDIS is "a website developed to help scientists and engineers in the Regions, states, and tribes conduct causal assessments in aquatic systems." The CADDIS webpage on pavement sealants is meant to provide brief background information on a potential issue of concern

in urban aquatic systems. This kind of background information is critical in the initial stages of a causal assessment, where assessors are deciding what hypotheses should be included in the assessment. The CADDIS webpage discusses coal-tar sealcoat and PAHs in the context of multiple environmental stressors associated with urbanization and urban waters. The webpage does not single out coal-tar sealcoats; many other sources and stressors associated with urban development are discussed throughout the Urbanization module. All of the information included throughout CADDIS (including the pavement sealant webpage) was reviewed by a five-person panel of expert external reviewers; these reviewers are listed on the CADDIS site.

EPA's Office of Research and Development (ORD) is conducting further research on this topic, and plans to publish a study evaluating USGS sediment data using a methodology recommended in the O'Reilly *et al.* (2014) article. EPA is also aware that new peer-reviewed research on coal-tar sealcoat, its potential effects on aquatic systems, and the effects of sealcoat bans and their impacts on PAH levels in the environment has been published since the release of these two documents. EPA may update the CADDIS webpage and stormwater BMP fact sheet in the future to include new relevant information.

If you are dissatisfied with this response, you may submit a "Request for Reconsideration" (RFR). EPA requests that any such RFR be submitted within 90 days of the date of this letter. The RFR should reference RFC # 14003. If you choose to submit an RFR, please send a written request to the EPA Information Quality Guidelines Processing Staff via mail (Information Quality Guidelines Processing Staff, Mail Code 2811A, U.S. EPA, 1200 Pennsylvania Ave., N.W., Washington, D.C. 20460); or electronic mail, quality@epa.gov. Additional criteria for information that should be included in the request is listed on the EPA Information Quality Guidelines website: http://www.epa.gov/quality/guidelines-ensuring-and-maximizing-quality-objectivity-utility-and-integrity-information.

Sincerely,

Joel Beauvais
Deputy Assistant Administrator
Office of Water

Thomas A. Burke, PhD, MPH
Deputy Assistant Administrator and EPA Science Advisor
Office of Research and Development

Enclosure

cc: Ann Duncan, P.E.
Chief Information Officer

References

Environmental Protection Agency. (2011). Assessment of Water Quality of Runoff from Sealed Asphalt Surfaces (EPA Publication No. 600/R-10/178). http://nepis.epa.gov/Exe/ZyPDF.cgi?Dockey=P100ECC8.txt

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National Research Council. (2009). *Urban Stormwater Management in the United States.* http://www.epa.gov/npdes/pubs/nrc_stormwaterreport.pdf

O'Reilly, K. T., Pietari, J. and Boehm, P. D. (2014). Parsing pyrogenic polycyclic aromatic hydrocarbons: Forensic chemistry, receptor models, and source control policy. *Integr Environ Assess Manag,* 10: 279-285.

EPA United States Environmental Protection Agency

Stormwater Best Management Practice

Coal-Tar Sealcoat, Polycyclic Aromatic Hydrocarbons, and Stormwater Pollution

NPDES

Minimum Measure

Pollution Prevention/Good Housekeeping

What are the sources of polycyclic aromatic hydrocarbons in the environment?

Polycyclic Aromatic Hydrocarbons (PAHs) are persistent organic compounds. These chemicals come from both natural and man-made sources. PAHs are naturally released in the environment from wildfires, volcanic eruptions, and degradation of biological materials contained in various sediments and fossil fuels (CDC/ATSDR, 1995; White and Lee, 1980). Man-made sources of PAHs in the environment include the incomplete burning of organic materials (e.g. coal, oil, gas, wood, garbage); vehicle exhaust; asphalt; coal-tar and coal-tar based sealcoats; creosote; and cigarette and tobacco smoke (CDC/ATSDR, 1995; CDC, 2009; EPA, 2009; National Research Council, 2009).

Many PAHs are of concern because of their harmful impacts on humans and the environment. They are persistent organic compounds; several PAHs are known or probable human carcinogens and toxic to aquatic life (Integrated Risk Information System (IRIS), 2014; Scoggins, McClintock, Gosselink, and Bryer, 2007).

What Is Coal-Tar Sealcoat?

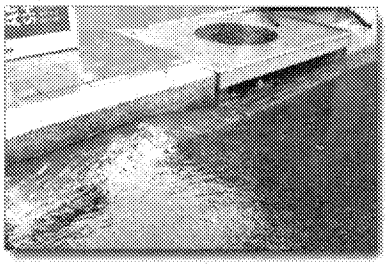
Coal-tar sealcoat is a type of sealant used to maintain and protect driveway and parking lot asphalt pavement. Coal-tar sealcoat typically contains 20 to 35% coal tar pitch, a byproduct of the steel manufacturing industry, which is 50% or more polycyclic aromatic hydrocarbons (PAHs) by weight (Mahler, Van Metre, Bashara, Wilson, and Johns, 2005).

Could Coal-Tar Sealcoat Be a Concern for Stormwater?

Studies found that PAHs are significantly elevated in stormwater flowing from parking lots and other areas where coal-tar sealcoats were used as compared to stormwater flowing from areas not treated with the sealant. For example, one study found the amount of PAHs in stormwater runoff was 65 times higher from parking lots sealed with coal-tar sealant vs. stormwater from unsealed parking lots (Mahler et al., 2005).Another study found that coal-tar sealcoat is the largest source of PAHs to urban lakes (Van Metre and Mahler, 2010). PAHs from coal-tar sealcoat may accumulate in the sediment of stormwater ponds, requiring expensive disposal of the dredged PAH-contaminated sediment (Mahler et al., 2012).

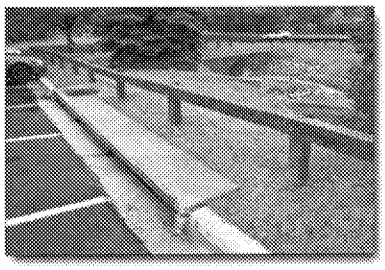
State and Municipality Examples Addressing PAHs from Coal-Tar Sealcoat

Several states and cities have taken action to address PAHs from coal-tar sealcoat. The following are some notable examples:



- The city of Austin, Texas banned the sale and use of coal-tar containing pavement sealants in 2005: http://austintexas.gov/CoalTar
- The District of Columbia banned the sale and use of coal-tar sealcoat in 2009: http://doee.dc.gov/coaltar
- In 2009, Minnesota restricted state agencies from purchasing undiluted coal tar-based sealant and directed its Pollution Control Agency to study the environmental effects of coal tar-based sealants and to develop management guidelines: https://www.pca.state.mn.us/water/restriction-coal-tar-based-sealants
- Washington State banned the sale of coal-tar pavement sealants on January 1, 2012 and banned the use of such sealants after July 1, 2013: https://fortress.wa.gov/ecy/publications/summarypages/1104021.html

Alternatives to Coal-Tar Sealcoat



Pavement options such as pervious concrete, permeable asphalt and paver systems do not require sealants. These types of pavements allow for stormwater to naturally infiltrate, resulting in decreased runoff.

For More Information

For more information you can watch EPA's webinar *Stormwater, Coal-Tar Sealcoat and Polycyclic Aromatic Hydrocarbons* available at: http://www.epa.gov/national-pollutant-discharge-elimination-system-npdes/npdes-training.

For information on assessing the toxicity of PAHs in sediment see: http://nepis.epa.gov/Exe/ZyPURL.cgi?Dockey=30006DOD.txt from EPA's Office of Research and Development.

Additionally, you can visit the USGS webpage on PAHs and coal-tar-based pavement sealcoat: http://lx.usgs.gov/sealcoat.html.

References

CDC. 2009. Polycyclic Aromatic Hydrocarbons (PAHs). http://www.epa.gov/sites/production/files/2014-03/documents/pahs_factsheet_cdc_2013.pdf.

EPA. 2009. Brownfields Profile Glossary. http://ofmpub.epa.gov/sor_internet/registry/termreg/searchandretrieve/glossariesandkeywordlists/search.do?details=&glossaryName=Brownfields%20Profile%20Glossary#.

EPA. 2014. EPA's Integrated Risk Information System (IRIS). http://www.epa.gov/IRIS/.

Mahler, B.J.; Van Metre, P.C.; Bashara, T.J.; Wilson, J.T.; Johns, D.A. 2005. Parking lot sealcoat: An unrecognized source of urban PAHs. *Environ. Sci. Technol.* DOI:10.1021/es0501565.

Mahler, B.J.; Van Metre, P.C.; Crane, J.L.; Watts, A.W.; Scoggins, M.; Williams, E.S. 2012. Coal-tar-based pavement sealcoat and PAHs: implications for the environment, human health, and stormwater management. *Environ. Sci. Technol.* DOI:10.1021/es203699x.

National Research Council. 2009. Urban Stormwater Management in the United States. http://www.epa.gov/sites/production/files/2015-10/documents/nrc_stormwaterreport1.pdf (pre-publication version)

Scoggins, M.; McClintock, N.L.; Gosselink, L.; Bryer, P. 2007. Occurrence of polycyclic aromatic hydrocarbons below coal-tar-sealed parking lots and effects on stream benthic macroinvertebrate communities. *Journal of the North American Benthological Society.* DOI:10.1899/06-109.1

Van Metre, P.C.; Mahler, B.J., Contribution of PAHs from coal-tar pavement sealcoat and other sources to 40 U.S. lakes. *Sci. of the Total Environ., 2010* DOI:10.1016/j.scitotenv.2010.08.014.

White, C. & Lee, M. Identification and geochemical significance of some aromatic components of coal. *Geochimica et Cosmochimica Acta, 1980.* DOI:10.1016/0016-7037(80)90231-8

Fact sheet disclaimer: http://www.epa.gov/national-pollutant-discharge-elimination-system-npdes/national-menu-best-management-practices-bmps#edu

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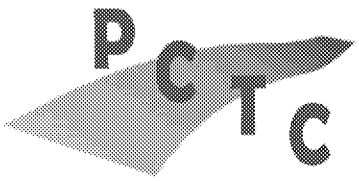

April 16, 2014

Information Quality Guidelines Staff
Ronald Reagan Building
Room M1200
1300 Pennsylvania Ave., N.W.
Washington, DC 20460

Via E-Mail: quality@epa.gov

Re: Request for Correction under the EPA Information Quality Guidelines

Publications: EPA CADDIS Volume 2: Sources, Stressors & Responses – Pavement Sealant & PAHs (EPA Publication #1)
http://www.epa.gov/caddis/ssr_urb_wsq4.html
http://www.epa.gov/caddis/ssr_urb_ref.html

EPA Stormwater Best Management Practice (EPA Publication #2)
"Coal-Tar Sealcoat, Polycyclic Aromatic Hydrocarbons, and Stormwater Pollution"
http://www.epa.gov/npdes/pubs/coaltar.pdf

Dear Sir or Madam:

On behalf of the Pavement Coatings Technology Council (PCTC), which represents numerous companies throughout the country that are part of the sealcoat industry, I write to submit a Request for Correction of information contained within the publications cited above that have been posted by the U.S. Environmental Protection Agency (EPA). This request is made pursuant to the EPA Information Quality Guidelines[1] and the Office of Management and Budget (67 F.R. 8452) in accordance with Section 515 of the Treasury and General Government Appropriations Act for Fiscal Year 2001 (Public Law 106-554).

[1] *Guidelines for Ensuring and Maximizing the Quality, Objectivity, Utility, and Integrity of Information Disseminated by EPA*, October, 2002;
http://www.epa.gov/quality/informationguidelines/documents/EPA_InfoQualityGuidelines.pdf

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INFORMATION REQUIRING CORRECTION – OVERVIEW

As part of a long standing campaign, certain individuals within the United States Geological Survey (USGS) and the City of Austin (COA), Texas, have used their government positions to persuade consumers, legislators, the press and even other government agencies to adopt their goal of banning the use of refined coal tar-based pavement sealants (RTS) across the country. All too often, this campaign has side stepped sound scientific methodology, upon which the vast majority of government scientists pride themselves, and has relied instead on a collection of questionably executed "studies" and press releases in which contrary scientific views are ignored, data are selectively used or withheld without explanation, methodology flaws are overlooked, and perhaps most disturbing, hypotheses are presented as undisputed facts.[2] The EPA has contributed to this flawed process by citing in the documents needing correction several of these studies without first conducting any type of critical review, at least none that has been apparent to the public.

The most obvious flaw in the EPA's approach to the RTS debate is that peer reviewed articles and studies which directly challenge the USGS and COA findings have been ignored. The EPA publications cited above, and examined in greater detail below, are examples of this breakdown in sound scientific methodology. Additionally, the USGS and EPA have been careful to proceed in such a way that neither has been required to hold a single hearing or public comment period that would subject to public scrutiny the USGS' underlying research or decisions made by EPA based on USGS research. In short, USGS strategies to phase out RTS largely have been developed behind closed doors, apparently by a small group of like-minded individuals, who share a common agenda. While such a process may be consistent with "politics as usual," it certainly is not consistent with sound science or government transparency.

Such tactics, instead, are consistent with a flaw that is becoming more and more evident, particularly in the field of government research and policy making. That flaw is known as "White Hat Bias," which has been defined in the scientific literature as "bias leading to the distortion of information in the service of what may be perceived to be righteous ends."[3] The potential for White Hat Bias certainly escalates when "new" discoveries are being pursued that carry with them favorable press attention, monetary grants and notoriety. With respect to the assessment of RTS by the USGS and EPA, the conspicuous absence of any citation of peer reviewed research that happens to be funded by industry should be a clear warning sign that

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[2] Three DQA challenges have been filed against the USGS and its coal tar sealant publications over the past 10 months. The first challenge, filed on May 15, 2013, focused on the USGS' 40 Lakes Paper in which the USGS claimed, mistakenly, that coal tar sealants had been shown to be the primary source of PAH contamination in lakes east of the Continental Divide. The 40 Lakes Paper will be evaluated in greater detail below since it is cited by the EPA. The 2nd DQA challenge, filed on May 31, 2013, focused on the USGS' inappropriate use of catfish tumor photos to frighten the public into considering coal tar sealant bans. The third challenge, filed on September 18, 2013, analyzed in detail a flawed RTS risk assessment conducted by the USGS and a toxicologist from Baylor. The three USGS DQA challenges, which should be reviewed by the EPA before responding to this Request for Correction, may be found at the following USGS websites: http://www.usgs.gov/info_qual/coal_tar_sealants.html and http://www.usgs.gov/info_qual/cancer_risk_coal-tar-sealed_pavement.html

[3] Cope,M and Allison, D, "White Hat Bias: Examples of Its Presence in Obesity research and a Call for Renewed Commitment to Faithfulness in Research Reporting," Int J Obes (Lond); 34 (1): 84-88; January, 2010.; http://www.ncbi.nlm.nih.gov/pmc/articles/PMC2815336/

some sort of bias may be at play. EPA Guidelines, of course, dictate that all reasonable efforts be made to guard against bias in EPA publications. When these efforts fail, corrections must be made. Any failure to take such action will continue to adversely affect those members of the PCTC who distribute or apply RTS because consumers and legislators who are being asked to consider the merits of proposed sealant bans or restrictions are being misled by the EPA publications cited above.

EPA INFORMATION QUALITY GUIDELINES

The OMB Guidelines require that EPA data collection and research activities be "carried out in a consistent, objective, and replicable manner" aimed at ensuring the objectivity, utility, and integrity of information disseminated to the public.[4] To be "objective," information published by the EPA must be presented in an "accurate, clear, complete, and unbiased manner."[5] "Objectivity" also requires that original and supporting data be produced and that sound statistical and research methods be followed.[6]

In developing its own Guidelines, the EPA adopted the quality principles in the Safe Drinking Water Act Amendments ("SDWA") of 1996 which remind staff and scientists that accurate, reliable and unbiased information involves the use of:

> (i) the best available science and supporting studies conducted in accordance with sound and objective scientific practices, including, when available, peer reviewed science and supporting studies.[7]

The EPA Guidelines specifically apply to "information" that the EPA disseminates to the public and generally includes any communication or representation of knowledge, such as facts or data, in any medium or form. Preliminary information disseminated to the public by the EPA is also covered by the Guidelines, as are materials that the EPA posts on its web pages.[8] Thus, the EPA publications for which corrections are being requested above clearly fall within the scope of both the OMB and EPA Information Quality Guidelines.

It should be noted that for influential scientific information (the USGS/EPA assessment of RTS clearly falls into this category), the EPA requires a "higher degree of transparency about data and methods" which "facilitate the reproducibility of such information by qualified third parties."[9] "Reproducibility" means that "independent analysis of the original or supporting data using identical methods would generate similar analytic results." When evaluating environmental issues and risk assessments, the EPA must apply a "weight-of-the-evidence approach that considers all relevant information and its quality, consistent with the level of effort

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[4] *See* USGS Guidelines, Section III; Office of Management and Budget ("OMB") Guidelines, 67 F.R. 8452, 8459 (February 22, 2002); *see also* EPA Information Quality Guidelines, Section 5.1 (October 2002).
[5] *Id.*
[6] USGS Guidelines, 67 F.R. 8452, 8459.
[7] EPA Guidelines, p. 22
[8] *Id.*, p. 15
[9] *Id.*, p. 20

and complexity of detail appropriate to a particular risk assessment."[10] As will be demonstrated below, these standards and guidelines have been overlooked when it comes to RTS.

ASSESSING THE IMPACT OF PAHS ON THE ENVIRONMENT

Many scientists note that contamination of urban lakes and streams by polycyclic aromatic hydrocarbons (PAHs) is widespread in the U.S.[11] The wide spread occurrence of PAHs is not particularly surprising since there is a consensus in the scientific community that PAHs have many potential sources, including vehicle emissions, motor oil, crude oil, power plant emissions, tire particles and industrial releases. Indeed, almost any type of combustion of organic matter will produce PAHs as a by-product, including natural sources such as forest fires and volcanoes. Indeed, PAHs have been around since the beginning of the Earth and are considered possible starting material for the earliest forms of life.[12] Thus, one would expect PAHs to be ubiquitous in our environment and, in fact, they are. Our earliest ancestors generated PAHs. If there was a fire that offered them warmth or light, or cooked their food, PAHs were present. Most of us continue with similar activities today when we grill on the backyard barbecue or throw a few logs into the fireplace.

Given the billions of years that PAHs have been present, one may wonder why the Earth is not overwhelmed by PAHs. The answer is rather basic. PAHs degrade naturally in variety of different ways such as through oxidation, photolysis and biodegradation by microorganisms.[13] PAHs are actually a food source for certain types of organisms. In the atmosphere, it has been reported that benzo(a)pyrene absorbed onto soot is readily photo oxidized, with 60% destroyed within the first 40 minutes of exposure to sunlight.[14] The rate and extent of biodegradation of PAHs in soil are affected by environmental factors such as temperature, pH, oxygen concentration, PAH concentrations and contamination history of soil, soil type, moisture and nutrients. Scientists have observed the half-life of PAHs in soil to range from a few days to several hundred days. In sediment, PAH transformation also takes place, although rates can sometimes be longer than those observed in soil.[15]

According to the Agency for Toxic Substances and Disease Registry (ATSDR), most direct releases of PAHs to the environment are to the atmosphere from both natural and anthropogenic sources, with emissions from human activities predominating.[16] Even with respect to surface waters, most of the PAHs are believed to result from atmospheric deposition.[17] It has been estimated that a total of 11,031 metric tons of PAHs are released to the atmosphere in the United States on an annual basis, with 36% of the total coming from residential heating, 6% from industrial processes, 1% from incineration, 36% from open burning, 1% from power generation,

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[10] *Id.*, p. 21
[11] Van Metre, P. C.; Mahler, B. J., Contribution of PAHs from Coal-Tar Pavement Sealcoat and Other Sources to 40 U.S. Lakes. Sci. of the Total Environ., 2010, v.409, 334-344.
[12] Allamandola, Louis et al. (April 13, 2011); "Cosmic Distribution of Chemical Complexity". *NASA;* http://amesteam.arc.nasa.gov/Research/cosmic.html
[13] ATSDR Toxicological Profile for Polycyclic Aromatic Hydrocarbons, Aug. 1995, pp. 246-54.
[14] *Id.*, pp. 247-48.
[15] *Id.*, pp. 252-54.
[16] *Id.*, p. 230.
[17] *Id.*, p. 235

and 21% from mobile sources.[18] This must be contrasted to the amount of RTS that has been estimated to abrade off of asphalt surfaces in a given year. According to the EPA, this number could be 160 metric tons[19], which we believe to be an elevated estimate, but even still is only 1.5% of the total PAH emissions mentioned above.

Most government agencies, including the EPA, agree that PAHs have a multitude of sources and are ubiquitous, not only throughout the United States, but throughout the world. Seen in the context of the array of possible sources, any attempt to determine the extent to which RTS may have contributed to PAHs found in a given environment, if at all, is bound to require a complex set of analyses that cross over into different scientific disciplines. For example, a scientist with an expertise in field sampling (*e.g.,* water, soil and sediment) may have little expertise at collecting PAH dust samples within homes, and even less expertise when it comes to applying sophisticated "fingerprinting" models to analytical results, such as those used in the specialty discipline of chemical forensics. Given the multidisciplinary areas of expertise involved in evaluating sources of and exposures to PAHs, questions about expertise should be asked. This is particularly true when a small group of government scientists, none with fields of primary expertise involving either PAHs source identification or source apportionment, appear to have influenced and/or published virtually all of the "relevant" articles on RTS. The need for questioning jumps to another level once it becomes evident that this same group has also failed to consider seminal publications in relevant disciplines, industry-sponsored research, and research conducted by the same or sister government agencies that might suggest contrary conclusions. Such circumstances are yet another warning sign that something may be amiss.

As an agency, the EPA is certainly familiar with what goes into a proper scientific evaluation of how certain chemicals, products or industrial processes may impact the environment. Hypotheses must be tested and retested using scientifically and statistically sound sampling techniques. To the extent possible, confounding factors and variables must be controlled, and when that is not possible, study limitations must be clearly expressed in order to avoid misleading policy makers and the public regarding the significance of any initial findings. And of course, critical and independent review is to be encouraged and brought to light, not ignored or suppressed. Many in the scientific community should be involved, not just a few who belong to an affinity group and have a great deal of personal and professional prestige at stake. Proper science and robust public policy demand nothing less.

Yet, when it comes to RTS, the EPA seems to have side stepped its own evaluation criteria. The web publications that are the focus of this Request for Correction make it appear as though the EPA has already determined that RTS adversely impacts human health and the environment and therefore should be banned or voluntarily phased out. As will be demonstrated below, the evidence cited by the EPA in support of this proposition is, at best, preliminary and incomplete, and at worst, a classic example of White Hat Bias. Either way, the EPA publications are misleading and must be corrected. It is ironic that the first EPA web publication addressed in detail below can be found on the EPA's CADDIS website, which emphasizes how important it is

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[18] *Id.*, p. 232

[19] EPA, Assessment of Water Quality of Run-Off From Sealed Asphalt Surfaces, EPA/600/R-10/178, September 2011, p. 4; http://nepis.epa.gov/Exe/ZyPDF.cgi?Dockey=P100ECC8.txt

to consider all relevant evidence when performing an environmental causation analysis.

THE EPA'S CADDIS SYSTEM – GENERAL PRINCIPLES

The Causal Analysis/Diagnosis Decision Information System, or CADDIS, is an EPA initiative developed to help scientists and engineers in the Regions, States, and Tribes conduct causal assessments in aquatic systems. The primary question to be addressed using CADDIS is "How can environmental assessors and managers determine the causes of environmental impairments?" The EPA recognizes the difficulty in answering this question because ecosystems are complex and environmental evidence is diverse.[20] CADDIS users are guided through the evaluation process via a website and guidance documents.

The importance of considering all relevant evidence and weighing it is emphasized throughout the CADDIS website, as demonstrated below:

> **Weighing of evidence**: We believe that all relevant evidence should be considered. Evidence comes from diverse sources of information such as observations at the site, regional monitoring studies, environmental manipulations, laboratory experiments, and general scientific knowledge. Information may come from the literature or may be generated *ad hoc*. Evidence may be generated from information by various methods including interpretation of reported observations, summary statistics, statistical modeling, and mathematical modeling.[21]

Scientists and engineers are further encouraged to consider the manner in which certain types of evidence may not be reliable because of a lack of data, poor quality data, a poorly defined impairment, or multiple causes.[22]

Perhaps most important, the philosophy behind CADDIS acknowledges that decision makers and stakeholders, such as the PCTC, should be viewed as part of the extended team engaged to investigate the cause of aquatic system impairment. As an agency with regulatory responsibilities, the EPA understands that industry provides important insights and often knows about historical impacts that may have become hidden by a changing landscape. Stakeholders also may have collected other types of relevant data over the years. The EPA uses all sorts of information provided by industry on a routine basis to accomplish its regulatory mission, so it is with insight gained from experience that CADDIS instructs the users of its website to "involve your stakeholders and decision-makers often."[23] For reasons that have never been expressed, the PCTC and its members have been excluded by the EPA from the process of evaluating the aquatic impacts of RTS, if any.

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[20] http://www.epa.gov/caddis/si_approach.html
[21] *Id.*
[22] *Id.*
[23] *Id.*

CADDIS - PAVEMENT SEALANTS & PAHs (EPA Publication #1)

Volume 2 of the CADDIS evaluation process deals with Sources, Stressors and Responses. This volume provides background information on many common sources, stressors, and biotic responses in stream ecosystems. According to CADDIS, urbanization is an increasingly pervasive land cover transformation that significantly alters the physical, chemical and biological environment within surface waters. The CADDIS website presents a diagram which identifies a multitude of pathways through which urbanization may affect stream ecosystems. That diagram is attached as Exhibit A.[24]

At the bottom left corner of the diagram is a pathway heading entitled "Water/Sediment Quality," and within it is a sub-heading called "Pavement Sealants." Users of the EPA's CADDIS system are encouraged to click onto the Pavement Sealants subheading in order to obtain more detailed information regarding this specific topic. The link leads to a webpage titled "Pavement Sealants and PAHS." A copy of this webpage is attached as Exhibit B. It is one of the EPA postings that clearly requires correction pursuant to the EPA's Information Quality Guidelines, not to mention the standards of the CADDIS system of causation analysis.

Exhibit B includes a prominently displayed picture of a sealed parking lot. Two seal coat studies are cited immediately below the parking lot photo. The first is a 2005 article authored by USGS scientist Barbara Mahler and others entitled "Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons."[25] This particular article was referenced for its observation (based on a small data set) that PAH concentrations in run-off from several RTS coated parking lots reportedly were 65 times higher than run-off from unsealed parking lots. The second article was published by COA staff scientist Mateo Scoggins and others in 2007 and is titled "Occurrence of Polycyclic Aromatic Hydrocarbons Below Coal Tar Sealed Parking Lots and Effects on Stream Benthic Macroinvertebrate Communities."[26] Although this study was cited for several propositions, it was initially mentioned for its finding that PAH concentrations in stream sediments were significantly higher downstream of coal tar sealed parking lots versus upstream sites. Evidently, Mahler et al. (2005) and Scoggins et al. (2007) were cited together by the EPA to create the mistaken impression that PAHs from RTS sealed parking lots must be to blame for several downstream PAH "hotspots" in COA streams.[27]

Before examining in greater detail below the flaws inherent within Mahler et al. (2005) and Scoggins et al. (2007), it is important to recognize that these two papers are outgrowths of attempts to provide justification for the COA banning the sale and use of RTS within city limits. As will be further explored below, Austin had sought the opinion of and been assured by the Texas Commission on Environmental Quality (TCEQ), the Texas Department of Health (TDH) and the U.S. Environmental Protection Agency (EPA) that pollutants in sediments in Barton Springs were present at such low levels that they posed no threat to either swimmers or the

[24] http://www.epa.gov/caddis/ssr_urb_intro.html
[25] Environ. Sci. Technol., 2005, Vol. 39, pp 5560-5566.
[26] Journal of the North American Benthological Society, 2007, Vol. 26(4): 694-707.
[27] Scoggins et al. (2007) was *not* cited for its finding that, contrary to Mahler et al. (2005), a relationship between the PAH signature of RTS and that of Austin area stream sediments could *not* be identified.

endangered Barton Springs salamander. City staff, however, believed they had identified a threat – PAHs from sealcoated parking lots. The City had engaged various consultants to look at sources of PAHs in soils on a hillside above Barton Springs and, in a series of articles published by the Austin American-Statesman in 2003, the belief of city staff that the consultants were wrong, that the sealcoated parking lot was the source of PAHs.[28] If these news accounts from Austin in the early 2000s are to be believed,[29] banning RTS was a goal long pursued by City staff who apparently had predetermined that RTS should be banned well before the USGS studies were even conducted. Indeed, the desire of COA staff to find a connection between RTS and PAHs in sediment can be seen in the description of the study reported in the newspaper.[30] Needless to say, the potential for research bias under these circumstances was high. By citing and summarizing Mahler et al. (2005) and Scoggins et al. (2007) on the CADDIS website (via Exhibit B) without evaluating the reliability, goals and limitations of these articles, the EPA has done little more than to blindly promote the agenda of certain employees of the USGS and COA to phase out RTS from the marketplace..

In furtherance of the COA's agenda, adopted as its own by the USGS, evidence had to be generated which showed that the above mentioned PAH "hotspots" created unacceptable environmental impacts, either in terms of human health risks or injury to stream biota. Scoggins et al. (2007) was once again cited within Exhibit B to fill this need. According to this study, decreases in downstream macroinvertebrate richness and density could be explained by increased levels of PAHs in sediment, and those PAHs must have come from RTS, at least according to the inferences that are to be drawn from Mahler et al. (2005).[31] The CADDIS posting thereby appears to provide an environmental activist looking to "act locally" with all of the necessary "background" information needed to convince municipalities and governmental entities across the country that RTS should be banned or phased out. The problem with this analysis, however, is that while certain findings from Scoggins et al. (2007) and Mahler et al. (2005) are cited, CADDIS' own criteria as well as subsequent peer reviewed articles that call into question the validity of these findings have been ignored.

DOWNSTREAM IMPACTS UNCONFIRMED

In providing general insights regarding the strength of certain types of evidence, the CADDIS website notes that "if an effect occurs downstream of a source, that is weak supporting evidence for emissions from that source as a cause..."[32] Remarkably, this same type of evidence, characterized by CADDIS as being "weak," is nevertheless offered up in Exhibit B as its primary "proof" that upstream coal tar sealed parking lots must have been causing PAH contamination

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[28] Austin American-Statesman Dec. 31, 2003. *Barton Creek cleanup costs rise: Projected price tag for tending to tainted soil jumps to $1.1 million.* http://www.statesman.com/

[29] *Id.*

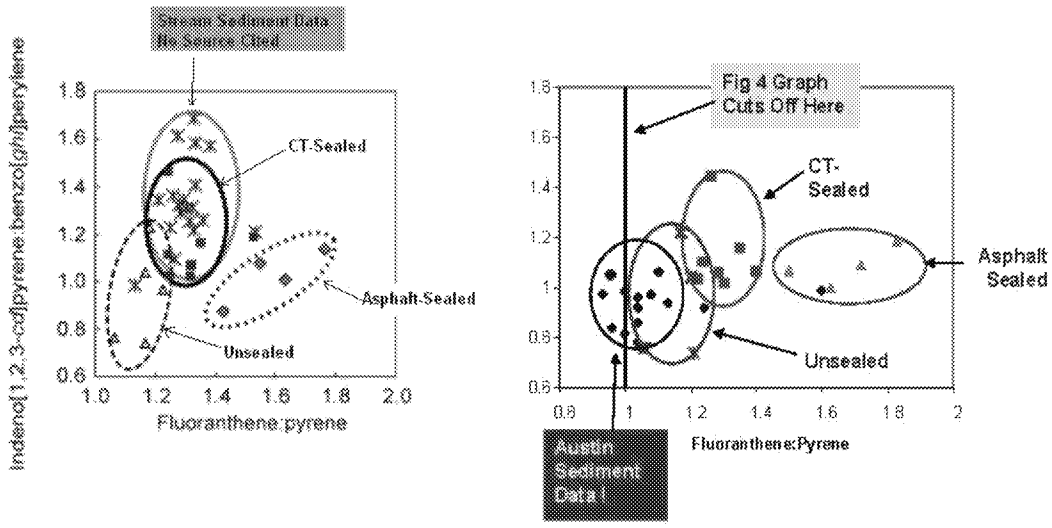
[30] Austin American-Statesman May 5, 2004. *Parking lot toxins lower than reported, agency say.* "The agency [USGS] has been working with Austin water quality officials to test the city's theory that toxic chemicals in sediments in Barton Creek, Barton Springs Pool and other Austin waterways are coming, in large part, from polluted particles that rain washes off parking lots." http://www.statesman.com/

[31] This is a common error of logic. Correlation does not prove causation, The fact that a rooster crows every morning just before sunrise does not mean that the rooster causes the sun to rise.

[32] http://www.epa.gov/caddis/si_approach.html

and biota degradation downstream. As it happens, even Mahler and Scoggins seemed to recognize the flaws in this argument since both attempted to shore up it up by presenting in their articles an environmental forensics technique – PAH double ratio plots - often applied as a first step in attempts to identify sources of PAHs. The double ratio plots, which are not mentioned in Exhibit B but are examined in detail below, failed to demonstrate that RTS from upstream parking lots was, in fact, causing downstream PAH contamination in sediment. Why these results were overlooked in Exhibit B is unknown.

Figure 4 from Mahler et al. (2005; graph on the left, below) seemingly demonstrates that PAH ratios from Austin urban stream sediment closely matched ratios generated by particles washed from parking lots covered with RTS. However, when DeMott and Gauthier (2006[33]; graph on right, below), attempted to reproduce Mahler's graph using actual data from sediments in Austin streams, as provided to them by the COA as the sediment results used in the Mahler et al. (2005) paper, they were unable to either reproduce the graph or find a relationship between RTS and Austin sediment PAHs.



ES&T Article – Figure 4 **Re-Plot with Austin Sediments**

The EPA Guidelines could not be clearer about the importance of the reproducibility of environmental data and findings. In a response to DeMott and Gauthier's comment, Mahler and Van Metre (2006)[34] tried to explain away this inconsistency by asserting that the commenters used different sediment samples. Unfortunately, and contrary to sound scientific procedure, Mahler and Van Metre (2006) did not make the underlying data available, so it was impossible for a long period of time to double check the accuracy of the assertions. Eventually, it became clear that Mahler et al. (2005) did not actually use PAH concentrations from stream sediment in

[33] DeMott, obert P. and Thomas D. Gauthier. "Comment on "Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons"." *Environmental Science & Technology* 40, (2006): 3657-3658

[34] Mahler, B.J., Van Metre, P.C., Bashara, T.J., Wilson, J.T., and Johns, D.A., 2006, *Response to comment on 'parking lot sealcoat: an unrecognized source of urban polycyclic aromatic hydrocarbons'*: Environmental Science and Technology, vol. 40, no. 11, p. 3659-3661. doi: 10.1021/es060585i

the plot, such as was done by DeMott and Gauthier (2006). Instead, Mahler et al. (2005) measured suspended solids in stream water, which were called "suspended sediment." Why Mahler et al. (2005) chose to rely on results of analyses of suspended solids for their fingerprinting and not complete the study design by including results from actual stream sediment is still unknown –and has resulted in confusion- especially since scientists before and after Mahler et al. (2005) have continued to focus their research on sediment.

Just as perplexing, in Mahler et al. (2005) the samples of suspended solids were not collected in the immediate vicinity of the few parking lots that had undergone the wash off testing, nor were the suspended solids all sampled at the same time, the same place or even in the same year. Specifically, four suspended solid samples were collected during the year that immediately preceded the parking lot testing from a creek in Austin having no relationship to the studied parking lots. Twelve other suspended solid samples were collected in yet a different year from three other streams.[35] None of these other three streams was located anywhere near Austin. Instead, they were in central Fort Worth, Tx, which is nearly 200 miles away. Thus, it is clear that none of the suspended solid sampling was spatially or temporally related to the parking lot study in question, which makes the double ratio analysis presented in Mahler et al. (2005) of questionable scientific value.

By the end of 2006, the only published attempt to use an environmental forensics method to compare Austin stream sediment with parking lot PAHs was DeMott and Gauthier's (2006) attempt to reproduce Mahler et al.'s (2005) double ratio plot. As indicated above, DeMott and Gauther's comment (which is mentioned nowhere within Exhibit B) concluded that double ratio plots using actual sediments from Austin failed to demonstrate any relationship between the PAHs in sediment and RTS. Clearly, these findings were inconsistent with the ban on RTS that had already been instituted in Austin. In Scoggins et al. (2007), COA staff also used data collected from Austin stream sediments in double ratio plots to evaluate whether sediment PAHs could be matched to those associated with RTS. The results obtained by Scoggins et al. (2007) were consistent with those found by DeMott and Gauthier (2006). They found "no significant clustering of field data with known source data."[36] Scoggins et al. (2007) then tried to explain why the observed results were inconsistent with what they had hoped to find by speculating as follows:

> 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.[37]

Of course, another possibility not mentioned in Scoggins et al. (2007) is that RTS was not the primary source of PAHs in Austin streams. The failure to find a relationship between

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[35] It was not until the summer of 2013, after PCTC asked for the assistance of the American Chemistry Society (which had published Mahler et al (2005)) that the sources of the 16 "suspended sediment" samples were revealed.
[36] See fn 14, *supra,* p. 702.
[37] *Id.*

the signature of PAHs in RTS and the signature found in sediments reported by Scoggins et al. (2007) confirms the same observation made by DeMott and Gauthier (2006). The failure of Scoggins et al. (2007), and Exhibit B, to acknowledge the possibility that RTS was not the primary source of PAHs found in sediment is problematic and misleading.

The problems and limitations associated with Scoggins et al. (2007) and Mahler et al. (2005) illustrate why data evaluations schemes such as CADDIS include detailed reviews of the scientific bases for making associations between sources and environmental receptors. Are these the types of issues that should have been raised or acknowledged on the CADDIS "Pavement Sealants and PAHS" webpage? Certainly. Indeed, it is difficult to understand how they were omitted. A related question that should be asked is what sort of pre-dissemination review, if any, was conducted by the EPA before Exhibit B was posted? By failing to note the limitations of the Scoggins et al. (2007) and Mahler et al. (2005) articles, policy makers, resource managers, the public and other researchers have been mistakenly led to believe that the science regarding "downstream impacts," as mentioned on the webpage, has been settled when clearly that is not the case.

DATA ON BENTHIC LIFE ARE INCONSISTENT

A second impression created by the CADDIS "Pavement Sealants and PAHS" webpage, and perhaps the most important when it comes to influencing policy makers, is that PAHs from RTS allegedly have been shown to adversely affect biota in nearby streams and bodies of water. Once again, Mahler et al. (2005) and Scoggins et al. (2007) have reported results that in no way prove this hypothesis. Accordingly, the EPA must correct this webpage to reflect the facts and science as they now exist, and not present only those theories that support the agenda-driven "science" of a few.[38]

For example, Scoggins et al. (2007) reports that the COA water body which allegedly had the greatest level of PAH contamination downstream was Barton Creek. A fact not mentioned on the EPA website is that PAHs in Barton Creek sediment also had some of the lowest impacts on biota. Scoggins et al. (2007) failed to find any correlation between the size of sealed parking lots upstream and degradation of downstream biota. Once again, these inconsistencies were attributed to the "complex mix of urban stressors on the benthic communities in these streams."[39] Scoggins et al. (2007) acknowledged that their data set was "very small." Each of these findings cast doubts on the initial impressions generated by Exhibit B, yet no mention of them can be found anywhere within Exhibit B. The public is left to jump to conclusions that are not supported by the data.

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[38] The manner in which Mateo Scoggins and Tom Ennis (both of the COA), Peter Van Metre and Barbara Mahler (both of he USGS), Judy Crane (Minnesota Pollution Control Agency) and Allison Watts (University of New Hampshire) have worked together to pursue their anti-coal tar sealant agenda is spelled out in detail as part of the PCTC's first USGS DQA challenge filed on May 15, 2013 regarding the 40 Lakes Paper and will not be repeated here. See fn 2, *supra*.

[39] Journal of the North American Benthological Society, 2007, Vol. 26(4): p. 704

Scoggins et al. (2007) was not the only study of biota in COA streams. For example, the Texas Commission for Environmental Quality (TCEQ) also sampled Barton Creek sediment a few years earlier in 2003 and 2004 and concluded that with few exceptions, the environmental quality of the creek was better than that expected when compared to typical water bodies in urbanized areas.[40] This was true even though a large parking lot sealed with RTS was identified in the vicinity of the creek and sampling areas and even though a former town gas site is located near Barton Springs Pool. The TCEQ concluded that aquatic life in Barton Creek was not impaired by alleged sediment toxicity associated with PAHs or any other chemical.[41] The Scoggins et al. (2007) study can be best understood as an attempt to ascribe adverse impacts to PAHs derived from RTS in stream and Barton Springs sediment. However, like DeMott and Gauther (2006), Scoggins et al. (2007) was unable to link sediment PAHs to RTS, and further was unable to correlate upstream sealed parking lots with downstream degradation, and did not address previous findings of the minimal to non-existent impacts of PAHs on Barton Creek by the TCEQ.

Another point overlooked by the EPA in its CADDIS "Pavement Sealants and PAHS" webpage is that only two of the seven streams evaluated by Scoggins et al. (2007) had downstream samples that exceeded the Probable Effects Concentration (PEC) of 22.8 mg/kg for PAHs in sediment,[42] and those two streams only barely exceeded this policy driven number which is not applicable when the bioavailability of PAHs in sediment is low – as it is most in most instances. As developed, the PEC of 22.8 mg/kg was intended to apply only to fauna such as worms, slugs and aquatic larvae, and not to fish or humans. PECs are not applicable to non-bottom feeding organisms because PAHs are generally insoluble, do not directly affect water quality and are not associated with exposures in the water column. PAHs sink to the bottom of waterways attached to sediment particles, along with many other substances found in urban and suburban storm water, such as pesticides, herbicides, animal waste, tire particles and soot. Because sediment-bound PAHs are often found to have low bioavailability, the PEC is often overly conservative even for bottom feeding organisms. Humans seldom if ever ingest significant quantities of sediment even when swimming in natural waterways, thus human health risks are miniscule. This fact was taken into account by the Texas Department of Health (TDH) as part of its Barton Springs assessment in 2004, explored in greater detail below.

PAHs are virtually insoluble in water, partitioning strongly into the solid phase where they are tightly bound to organic materials within sediments. The hydrophobic characteristic of PAHs makes them unavailable for exposure to aquatic organisms, and thus there is often no correlation between the measured PAH concentrations in sediments and those concentrations that adversely affect benthic organisms.[43] Thus, predictions of sediment toxicity in the real world cannot be based on measured PAH concentrations. To address the need to be both protective of

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[40] TCEQ, Barton Creek Sediment Toxicity Evaluation to Aquatic Life, July 29, 2004; http://www.tceq.texas.gov/toxicology/barton/BSPFull_PDF.html/at_download/file

[41] *Id.*

[42] Barton Creek had one downstream sample that was measured at 32 mg/kg and Walnut Stream had one that was measured at 30 mg/kg.

[43] Neuhauser, E., J. Kreitinger, D. Nakles, S. Hawthorne, F. Doherty, U. Ghosh, M. Khalil, R. Ghosh, M. Jonker and S. van der Heijden (2006). "Bioavailability and toxicity of PAHs at MGP sites." Land Contamination & Reclamation 14(2): 261 - 266.

the environment and realistic in assessing risks to benthic organisms, EPA developed a tiered approach to evaluating potential sediment toxicity to help policy makers and resource managers expend resources appropriately in managing PAH-containing sediment.[44] The tiered approach involves bioavailability testing of PAHs in the whole sediment (Tier 1), bioavailability testing based on concentrations of PAHs in pore water (Tier 2), and sediment toxicity testing to evaluate the probable risk of adverse effects (Tier 3).

The Department of Defense (DoD) applied EPA's approach to a study of the impact of PAHs on benthic life in the Anacostia River where several military facilities located in the area of Washington D.C. had released large quantities of PAHs into waterways over many decades. The phenomenon the EPA approach was designed to address was observed in the DoD study, in which the PEC value (23 ppm) is actually well below the level at which toxic effects to benthic organisms were observed to occur, as demonstrated in the figure below. [45]

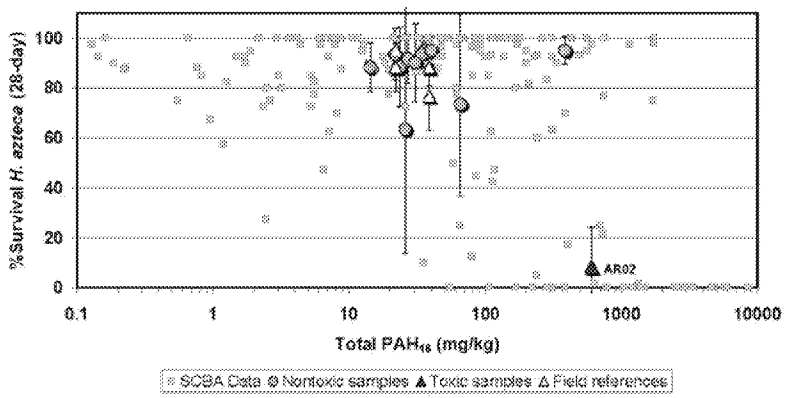

Figure 6-2. Bulk Sediment PAH_{16} Compared to *H. azteca* Survival.

Similar results have been reported by other researchers, such as Neuhauser and colleagues, who also found that standard toxicity testing of benthic organisms showed virtually no toxicity at PAH concentrations much greater than the PEC value of 23 ppm.[46] These results are also consistent with the observation in Scoggins et al. (2007) that Barton Creek, in which one sample had the highest PAHs sediment measurement of 32 ppm, had essentially normal biota. In other words, the alleged environmental concerns generated by the CADDIS "Pavement Sealants and PAHS" webpage, as presently written, have been exaggerated, are speculative, and need to be corrected.

ACTIONS TO BE TAKEN

Presently, it is unknown how the current version of Exhibit B came to be an EPA

[44] Burgess, R. M. Evaluating Ecological Risk to Invertebrate Receptors from PAHs in Sediments at Hazardous Waste Sites (Final Report). U.S. Environmental Protection Agency, Ecological Risk Assessment Support Center, Cincinnati, OH, EPA/600/R-06/162F, 2009.

[45] Geiger, S., AECOM, Final Report - The Determination of Sediment Polycyclic Aromatic Hydrocarbon (PAH) Bioavailability using Direct Pore Water Analysis by Solid-Phase Microextraction (SPME), ESTCP Project ER-200709, Aug. 2010, p. 48 (report may be downloaded at http://www.serdp-estcp.org/Program-Areas/Environmental-Restoration/Risk-Assessment/ER-200709/ER-200709).

[46] See fn 43, *supra*.

publication and disseminated on EPA's website. One possibility is that EPA staff simply relied on the USGS and the COA articles about RTS and their alleged impact on streams and sediment without looking further into the matter. Regardless, Exhibit B represents a clear violation of the EPA's Information Quality Guidelines and of the philosophies behind CADDIS. As such, Exhibit B must be withdrawn pending correction.

If the PCTC had been consulted, as contemplated under the CADDIS guidelines, it is certain that many of the issues raised above would have been brought to the attention of the EPA and presumably would have found their way into a more complete and accurate version of Exhibit B. As demonstrated above, the flaws and limitations of Scoggins et al. (2007) and Mahler et al. (2005) must be addressed along with (1) the ecological assessment conducted by the TCEQ in Barton Springs, (2) the inability to reproduce the information on which the conclusions of Mahler et al.(2005) are based (as reported by DeMott and Gauthier (2006) and confirmed by Scoggins et al. (2007)), and (3) the benthic life toxicity testing performed for the DoD and similar results reported by others. Other more recent studies must also be mentioned. By now, it should be readily apparent that the CADDIS website can no longer cite Scoggins et al. (2007) and Mahler et al. (2005) as though they are the only references worth mentioning on the issue of RTS and its alleged impact on the environment. In order for the CADDIS website to reflect sound science rather than just advocacy, the list of references must be updated to reflect all relevant information, studies and articles. Some of the most recent publications that need to be consulted and cited for their content can be found in Exhibit D, attached.

As things now stand, the EPA should immediately remove the CADDIS Pavement Sealants and PAHs webpage. Once this is done, the PCTC will gladly participate in any future discussions that the EPA may wish to have geared toward creating an accurate and unbiased webpage that outlines the issues presently being debated regarding RTS. The new webpage must be based on all of the relevant investigations and peer reviewed publications, not just studies generated by the USGS and COA and the "ban RTS" affinity group that has developed over time.

EPA STORMWATER BEST MANAGEMENT PRACTICE (EPA Publication #2)

EPA's Stormwater Best Management Practice publication, attached as Exhibit C, has three sections that merit specific attention. The first section is on the left side of page 1 and is entitled "Could Coal-Tar Sealcoat Be of Concern for Stormwater?" It begins with another reference to Mahler et al. (2005) and its assertion that the amount of PAHs found in storm water runoff is higher with RTS sealed parking lots than with unsealed parking lots. Presumably, Mahler et al. (2005) was cited once again to create an impression that RTS must be contaminating nearby streams and ponds. And, once again, there is no mention of the manner in which the findings and methodologies of Mahler et al. (2005) have been questioned in subsequent research and publications.

Even Mahler and co-authors seemed to recognize that neither Mahler et al. (2005) nor Scoggins et al. (2007) had, in fact, demonstrated that PAHs from RTS were a significant source of contamination in stream and pond sediment. Indeed, the inability of DeMott and Gauther (2006) and Scoggins et al. (2007) to reproduce a similarity in PAH ratio signatures between RTS

and actual sediment signatures had not been refuted. Thus, Mahler and several colleagues (including her husband Dr. Peter Van Metre) used additional government funding to research and publish in 2010 an article entitled "Contribution of PAHs from Coal-Tar Pavement Sealcoat and Other Sources to 40 U.S. Lakes."[47] This article (Van Metre and Mahler 2010), is prominently cited in the first section of Exhibit C (left side) for the proposition that "coal tar sealcoat is the largest source of PAHs to 40 urban lakes." As presented, this proposition is uncontested, thereby encouraging readers (including legislators and other government personnel) to infer that this article represents the present state of science on this topic. That is not the case, and any such inference that has been created by the EPA, whether intentional or not, must be corrected. OMB, EPA and CADDIS Guidelines mandate that appropriate action be taken.

VAN METRE & MAHLER (2010) AND SUBSEQUENT CRITIQUES

From the perspective of those who wish to ban RTS, the irreproducibility of the seemingly simple double ratio plot method of source identification must have been disheartening. A different method would be needed if there was going to be any hope of forensically connecting sediment PAHs with RTS. Van Metre and Mahler (2010) attempted to do this by adapting a source identification and apportionment model developed by EPA "to identify sources of inorganic compounds in the atmosphere:" the Chemical Mass Balance (CMB) model.

Publication of Van Metre and Mahler (2010) moved the focus away from Austin, which was understandable in view of the findings published by the TCEQ for Barton Springs and the PAH ratio signature results noted by DeMott and Gauthier (2006) and Scoggins et al. (2007). Use of the CMB model was instead directed toward proving that RTS was the primary source of PAHs in 40 lakes throughout the United States, and in Van Metre and Mahler (2010), they claimed to have achieved this result. An overview of Van Metre and Mahler (2010) allows one to conclude rather quickly that without the new CMB modeling results, there is little to support the USGS' proposition that RTS is the "dominant" or "most substantial" source of PAHs in lake sediment. Thus, if the CMB modeling is in some way flawed, incomplete or inconclusive, then it necessarily follows that the conclusions of Van Metre and Mahler (2010) are also flawed and cannot provide a science-based foundation for any agenda to ban RTS, voluntarily or otherwise.

Several publications challenge the findings of Van Metre and Mahler (2010). The first was a comment on that paper and a companion paper (Van Metre et al., 2009) by O'Reilly et al. (2011).[48] In 2012, a peer-reviewed overview of the body of USGS (Mahler – Van Metre) source identification and apportionment efforts was published by Dr. O'Reilly and others titled "Forensic Assessment of Refined Tar-Based Sealers as a Source of Polycyclic Aromatic Hydrocarbons (PAHs) in Urban Sediments."[49] Of particular significance, O'Reilly et al. (2012) followed CADDIS guidelines by using a "multiple lines of evidence" approach to address the results being claimed by Van Metre and Mahler (2010). According to O'Reilly et al. (2012), while Van Metre and Mahler (2010) identified some similarities between the PAH profiles of

[47] Van Metre, P.C., Mahler, B.J., Science of the Total Environment, Vol. 49, pp. 334-344 (2010).
[48] O'Reilly, K., J. Pietari, and P. Boehm, 2011, Comment on "PAHs Underfoot: Contaminated Dust from Coal-Tar Sealcoated Pavement is Widespread in the U.S.": Environmental Science & Technology, v. 45, p. 3185-3186.
[49] Environmental Forensics, Vol. 13, pp 185-196.

RTS and urban sediments, such profiles are not unique to RTS and likely had been impacted by weathering. It should be noted that O'Reilly et al. (2012) was published six months *before* the EPA posted Exhibit C and the critical comment (O'Reilly et al., 2011) had been published over a year prior to dissemination of Exhibit C.

In another related, peer reviewed article, O'Reilly and coauthors took their analysis further by using Principle Component Analysis (PCA) to evaluate in detail individual sediment sampling locations included in the USGS model. This paper, published online in 2013, is titled "Parsing Pyrogenic Polycyclic Aromatic Hydrocarbons: Forensic Chemistry, Receptor Models, and Source Control Policy."[50] The findings demonstrate that the CMB modeling used in Van Metre and Mahler (2010) fail to support the claim that parking lot sealers are a significant source of PAHs in urban sediments in either Austin, TX or Lake Ballinger, WA. Yet, Exhibit C makes it appear as though the conclusions offered by Van Metre and Mahler (2010) are an unchallenged scientific fact. Obviously, that is not true and Exhibit C accordingly must be removed from the internet and corrected.

Concerns about the misuse of the CMB model (as configured by the USGS) continue, especially since many of the same mistakes are being made by other authors who have adopted what appears to be a USGS approved method. Another government scientist who has worked closely with Drs. Van Metre and Mahler, Dr. Judy Crane of the Minnesota Pollution Control Agency, has stepped into the fray by recently publishing an article that seems to respond directly to the critiques of O'Reilly et al. (2012).[51] While this exchange of ideas is welcomed and fundamental to the progress of science, the manner of the discussion raises several questions. Dr. Crane has crafted her article so that it makes no reference or citation to any of the peer reviewed literature that challenges earlier USGS articles, such as the series of papers published by O'Reilly et al. (2011, 2012, 2014a) or earlier papers that question the purported relationship between RTS and PAHs in urban sediments (DeMott and Gauthier, 2006; DeMott et al., 2010, Scoggins et al., 2007). Yet, Dr. Crane seems to respond to O'Reilly et al. (2012) point by point, especially with respect to his critiques that Van Metre and Mahler (2010) neglected to consider a null hypothesis and multiple lines of evidence. If this reflects an intentional tactic by Dr. Crane to minimize the impact of O'Reilly et al. (2012) and O'Reilly et al. (2014a) by reducing the number of times that these articles are cited, that is a problem and a clear example of White Hat Bias. Additionally, if references to the O'Reilly articles were omitted as part of an effort to prevent others from reading these articles, that is also a problem. Unfortunately, this failure to cite publications with opposing points of view is becoming a regular feature of publications authored by members of the "ban RTS" affinity group.[52]

Regardless, in partial response to a Freedom of Information Act (FOIA) request and in information provided in response to a request for the assistance of the American Chemistry

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[50] Integrated Assessment and Environment Management, 2014a. Vol 10, pp. 279-285.
[51] Crane, J. L., 2013, "Source Apportionment and Distribution of Polycyclic Aromatic Hydrocarbons, Risk Considerations, and Management Implications for Urban Storm Water Pond Sediments in Minnesota, USA;" Arch Environ Contam Toxicol. DOI: 10.1007/s00244-013-9963-8.
[52] Via FOIA responses, PCTC is also in possession of emails that seem to demonstrate a coordination of efforts among USGS, COA and Minnesota government employees to suppress public awareness of industry-funded research. PCTC is investigating how best to make these emails available to the public.

Society's Ethics Committee, the USGS has provided sufficient underlying information to reconstruct the USGS CMB modeling results, which is reflected in an article by O'Reilly and others recently accepted for publication in *Polycyclic Aromatic Compounds* (O'Reilly et al. 2014b). This article uses Dr. Crane's paper as another case study to illustrate the manner in which CMB can be misapplied and lead to erroneous conclusions.[53] O'Reilly et al. (2014b) points out how Dr. Crane makes the same types of mistakes as Drs. Van Metre and Mahler by failing to recognize properly that her results essentially can be recreated even when RTS is removed from the CMB analysis. In scientific terms, all three scientists have failed to prove the null hypothesis which is a fundamental and a necessary step in evaluating any theory, mediating acceptance of the theory by the science community as a whole. In addition, with the information received via the FOIA request, a comprehensive evaluation of the USGS application of the CMB model to PAHs in sediments was made possible, resulting in the *Technical Evaluation of Van Metre and Mahler (2010)* included here as Exhibit E.

CONFUSION REGARDING ENVIRONMENTAL IMPACTS

In the paragraph that follows the reference to Van Metre and Mahler (2010), and after leading the reader to believe that large quantities of PAHs are released into the environment, Exhibit C states that "PAHs are of concern because of their harmful impacts on humans and the environment . . . [S]everal PAHs are known or probable human carcinogens and toxic to aquatic life." Yet, as discussed above, EPA elsewhere recognizes that, in the real world, there is often no correlation between sediment PAH concentrations and toxicity to aquatic organisms, and has developed an approach to determining if sediment toxicity could be of potential concern. Many of the issues regarding the impact of PAHs on aquatic life have already been addressed above and will not be repeated here. Suffice it to say that alleged risks to aquatic life, as claimed in Scoggins et al. (2007) (which is cited in Exhibit C), appear to be exaggerated and, as of yet, still have not been forensically linked to PAHs coming from RTS. Scoggins et al. (2007) is a small study, and inconsistent with earlier findings and conclusions offered by the TCEQ. Further, the Scoggins et al. (2007) study, as well as Exhibit C, does not consider sediment PAH toxicity in the broader context of the extensive science that has been published by others on this topic. As things now stand, the RTS theory being pushed by the USGS is nothing more than a questionable hypothesis being treated as a fact.

As a cautionary note, PCTC recommends that EPA should be careful when reviewing any new claims of environmental impact that might be made by the USGS in the future. The extent to which the USGS has been willing to exaggerate such claims in the past was made clear in a second DQA challenge that was filed by the PCTC on May 31, 2013.[54] The focus of this challenge is the USGS' use of catfish pictures with horrible looking lesions on their lips as evidence of what PAHs and, in theory, RTS can do to fish in streams and rivers. Ironically, it turns out that many of the afflicted catfish that were studied and photographed came from the Anacostia River where, as mentioned above, the DoD had admitted to releasing large quantities of PAHs into the environment. Even more significant, however, is the fact that the lip tumors

[53] O'Reilly, K. T., S. Ahn, J. Pietari, and P. D. Boehm, 2014b, Use of receptor models to evaluate sources of PAHs in sediments: Polycyclic Aromatic Compounds. Awaiting DOI.
[54] *See* fn 2, *supra.*

displayed in photos used by the USGS have *not* been shown to be caused by PAHs or by RTS. One theory presently being considered is that the tumors might instead be related to viruses. Nevertheless, if one goes to the USGS' Coal Tar Sealant "Fact Sheet," the catfish photo can still be seen at page 5, still scaring a trusting public. This illustrates the danger in blindly citing USGS websites and publications, which would seem to be subject to less than rigorous internal review, without first conducting an independent review of the assertions being made. Another way of stating this recommendation is to urge EPA to rely on its own CADDIS guidance in evaluating environmental impacts suggested by any study conducted by any research team.

CONFUSION REGARDING HUMAN IMPACTS

The reference in Exhibit C to "harmful impacts on humans" presumably is both scary and vague. Certainly, the reader is encouraged to jump to the conclusion that RTS can cause cancer since RTS contains PAHs and PAHs "are known or probable human carcinogens."[55] If the goal is to convince the public that RTS use should be banned or discontinued, creating fear is a crucial part of this strategy. As it turns out, this fear has no basis in fact as far as RTS is concerned, and any inference along those lines created by Exhibit C must be clarified and corrected. The purpose of EPA publications is to provide accurate and unbiased information to the public, not to generate unnecessary anxiety.

Referring once again to the much cited Scoggins et al. (2007), it was related earlier that the TCEQ was asked to assess to what extent certain parking lots, covered with RTS, may have impacted nearby biota in and around Barton Springs. The TCEQ repeatedly concluded that no adverse impacts could be found of any significance. As part of that same investigation, the Texas Department of Health (TDH), the ATSDR and EPA Region 6 also became involved to determine if human health was being adversely affected, especially since so many residents of Austin use the Barton Springs pool for recreational swimming, including children of all ages. After conducting an extensive study, as highlighted in the quote below, the TDH and ATSDR concluded that any theoretical cancer concerns raised by the press and environmentalists were insignificant.[56]

> We reviewed the results from water and sediment samples collected by the City of Austin, the United States Geological Survey, the Lower Colorado River Authority, and the Texas Commission on Environmental Quality. We reviewed over 14,500 individual data points, involving approximately 441 analytes, collected over the past 12 years. We screened the contaminants by comparing reported concentrations to health-based screening values and selected twenty-

[55] In the reference section of Exhibit C, readers are encouraged to visit the EPA's Integrated Risk Information System (IRIS) website, presumably to obtain additional information regarding the characterization of B(a)P as a "probable human carcinogen." Presently, as part of the IRIS program, the EPA in Washington D.C. is updating its hazard assessment and toxicological review of B(a)P. Public comments have been solicited. As part of this process, epidemiology and toxicology studies have been reviewed which indicate that the International Agency for Research on Cancer (IARC) needs to revise its PAH hazard assessment to comport with the findings of modern scientific studies rather than centuries old case reports .

[56] ATSDR, Health Consultation Barton Springs Pool, April 18, 2003, p. 1
https://www.google.com/#q=barton+springs+pool+health+consultation

> seven contaminants for further consideration. Of those 27 contaminants, 20 were polycyclic aromatic hydrocarbons (PAHs). The others included arsenic, boron, cadmium, bis(2-ethylhexyl)phthalate, total petroleum hydrocarbons (TPH), thallium, and lead.
>
> * * *
>
> *We did not find any information to support contention that swimming every day in Barton Springs would result in adverse health effects.* Thus, **we have concluded that swimming and playing in Barton Springs Pool poses no apparent public health hazard**. We recommend continued public health education to address any questions that the public may have concerning the risks associated with swimming in the pool. (All emphases in original.)

EPA Region 6 arrived at nearly an identical set of findings which was confirmed in a letter sent by Director Myron Knudson to the City of Austin dated April 17, 2003. His conclusions were:

> 1. The information reviewed does not indicate that people who swim in the Barton Springs Pool would be exposed to levels of contaminants that would be expected to cause adverse effects.
>
> 2. Adverse health outcomes from exposure to soil near the creosote-treated posts used for erosion control near the shallow end of the pool are not likely.
>
> 3. The levels of total petroleum hydrocarbons detected in both the water and sediment in the pool are not expected to result in adverse health outcomes.

A copy of said letter is attached as Exhibit F.

As part of this investigation, the TDH and ATSDR reviewed the literature regarding PAHs and their alleged role in causing cancer in humans. Their findings, quoted below, are clearly inconsistent with the message of fear that has been generated by Exhibit C, as presently drafted. [57]

> Benzo[a]pyrene (BaP) is perhaps the most toxicologically significant PAH and along with several other PAHs has been classified by the EPA as a "probable human carcinogen". This classification is based on animal data where repeated BaP administration in numerous strains of at least four species of rodents and several primates has been associated with increased incidences of total tumors and of tumors at the site of exposure. Human data specifically linking benzo[a]pyrene (BaP), or any of the other PAHs to a carcinogenic effect are lacking. Although lung cancer has been found in humans by exposure to various mixtures of polycyclic aromatic hydrocarbons known to contain BaP including cigarette smoke, roofing tar, and coke oven emissions, it is not possible to conclude from this information that BaP or any other of the PAHs is the responsible agent.

[57] *Id.*, p.7-8.

The TDH and ATSDR also demonstrated through a risk assessment that the greatest theoretical cancer risks would not come from exposure to PAHs in sediment or water, but instead would come from the PAHs that are in our food.[58] These are facts are missing from the EPA publications that are the subject of this Request for Correction. There can be no real dispute that EPA and OMB Guidelines mandate that such information be made available to the public as part of any assessment or summary which raises the issue of RTS and potential human health effects.

In a similar type of analysis, the Food and Drug Administration (FDA) has approved the use of coal tar, which contains high concentrations of PAHs, in therapeutic products and medications that are applied directly to the skin of patients and consumers. In doing so, the FDA considered the long history of coal tar use in this manner, as documented in the scientific literature, and concluded that such exposures do not increase people's risks of cancer.[59] Similarly, there is no evidence that low level or intermittent exposure to coal tar or coal tar pitch, beyond its therapeutic uses, has caused cancer in humans. And perhaps most important for the purpose of this Request for Correction, there is not one peer reviewed study in the world's published medical and scientific literature which has found that RTS specifically causes cancer in humans – not one. This is true despite decades of widespread use. All of this information has been omitted from Exhibit C.

On January 24, 2012, another RTS article written by Drs. Mahler and Van Metre was published. This one was titled "Coal Tar Based Pavement Sealcoat and PAHs: Implications for the Environment, Human Health, and Stormwater Management." It is the last article cited by the EPA in support of Exhibit C. Near the end of this article and in a section captioned "Human-Health Concerns," the authors speculated that "non-dietary ingestion of PAH-contaminated house dust and soil likely are the most important routes of exposure, but a complete human risk analysis is required before the cancer risk associated with ingestion of these media can be quantified." Ten months later just such an analysis was published which will be referred to as the "Williams/USGS Risk Assessment."[60] It would appear that Exhibit C was prepared at roughly the same time, or perhaps a bit earlier, which may explain why the Williams/USGS Risk Assessment is not mentioned within Exhibit C.

A great many flaws were found to exist within the Williams/USGS Risk Assessment which include mistakes and unexplained selective use of data in data collection, sampling techniques, exposure factor calculations, cancer slope factor assumptions, and the interpretation of data. These mistakes led the PCTC to file a third USGS DQA challenge, this one on September 18, 2013. Details will not be repeated here, but can be found at the USGS Quality

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[58] Id., *compare* Appendix D p. 71 (food risks) with Table 4 p. 31 (sediment risks) and Appendix D p. 71 (water risks). Risks associated with an average US diet greatly exceed any alleged risk associated with PAHs in Barton Springs pool.

[59] See Dennis Baker letter, FDA, 2/22/01; http://www.fda.gov/ohrms/dockets/dailys/01/Mar01/030601/pdn0001.pdf

[60] Williams, E.S.; Mahler, B.J.; Van Metre, P.C., Cancer risk from incidental ingestion exposures to PAHs associated with coal-tar-sealed pavement. Environ. Sci. Technol. 2012, v. 47 (2), 1101-1109.

Information website.[61] Suffice it to say that no scientist has yet reproduced the USGS risk assessment findings and conclusions, and it is doubtful that any will do so.[62]

Certainly, all of these facts are the type that should be important to members of the public (including policy makers) who are trying to assess in an even handed manner what type of human health risks, if any, are associated with RTS. Unfortunately, someone at the EPA apparently decided that it was best for the public not to have this information, or in the alternative, was somehow unaware of the many peer reviewed articles and DQA challenges that exist. Either way, Exhibit C must be corrected and amended to include references to these additional resources, articles and facts. To exclude them would be a clear example of White Hat Bias and an ongoing violation of the OMB and EPA Information Quality Guidelines.

WHAT ARE STATES AND MUNICIPALITIES DOING?

The second section within Exhibit C (found on the right side of page 1) that requires correction is the EPA's suggestion that there is a wave of growing support in favor of RTS bans across the country. The first question to be asked is why such information has been included at all. This "scorecard" obviously has nothing to do with science and is an example of advocacy, pure and simple. For this reason alone, this section should be deleted from Exhibit C.

Even if one assumes for the sake of argument only that RTS advocacy somehow is an appropriate activity for the EPA, the next question to be asked is whether this scorecard has been drafted in such a way as to mislead the public into thinking that all "enlightened" governmental entities which have considered the issue have decided to implement bans or restrictions. While this would certainly appear to be intent behind the scorecard, the truth is very much different.

According to the U.S. Census Bureau, there were 36,011 municipalities and townships in the United States in 2007, and of course, 50 states. The only two states that have banned the use of RTS are Minnesota and Washington, and Washington is a state where RTS generally is not used, so a ban there is essentially meaningless. And as for the rest of the 48 states, well over 99.9% of the local governments in these states have no bans or restrictions on the use of CTS. Bills to ban or restrict RTS have been presented in other states, and have been met with defeat, and for good reason. The science does not support the hypotheses and fear being generated by RTS ban advocates.

Quite simply, there is no mandate to ban RTS across the country. Well informed people have decided otherwise. Implying that there is some sort of trend here is inappropriate and another example of White Hat Bias.

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[61] See fn 2, *supra*.

[62] A Comment which questions the conclusions set forth within the Williams/USGS Risk Assessment has also been published, along with a Response from the original authors. *Environ. Sci. Technol.*, **2014**, *48* (1), pp 868–871). Given the format imposed by the Journal, the arguments presented are an abbreviated version of what has been presented in the PCTC DQA challenge filed on September 18, 2013 (see fn 2, supra).

WHAT EFFECT HAVE BANS HAD ON STREAMS?

Although they are in a very small minority, a few places outside of Washington State and Minnesota have implemented RTS bans. As indicated above, the first to do so was Austin, Texas back in 2006. As a result, Austin is an ideal spot to determine if its ban has had any impact on actually reducing the levels of PAHs in sediment.

Such a study was published in a peer reviewed article in 2010 titled, "PAHs in Austin Sediments after a Ban on Pavement Sealers."[63] PAH concentrations were measured in stream sediments collected before and after Austin's municipal ban on the use of coal-tar-based pavement sealers. Samples were collected in October 2005 and again in April, 2008, approximately 2.5 years after the ban. Differences in total PAH concentrations between samples collected before and after the ban showed no net change in PAH levels in Austin stream sediments. Furthermore, evaluation of PAH chemical signatures revealed subtle differences in PAH profiles that appeared to reflect the effects of weathering rather than a change in PAH sources. Indeed, the work by DeMott and colleagues was further evaluated by O'Reilly et al. (2013),[64] with the conclusion that RTS was not an identifiable source of PAHs in Austin sediments before or after the ban.

The point to be made is that Exhibit C clearly is encouraging the public to consider bans or alternatives to RTS in the hope that such actions will somehow improve aquatic systems, but there is no valid evidence at this time which demonstrates that such actions will have any impact at all. Rather than raise this issue in a straight forward manner for scientific discussion, the EPA has ignored it. Any mistaken impressions created in Exhibit C along these lines must be corrected.

LIST OF REFERENCES ARE INADEQUATE

The last section of Exhibit C, on page 2, has a limited and incomplete list of five References, each of which has been analyzed in this Request for Correction of Information and shown to be inaccurate or misleading for a variety of reasons. Thus, Exhibit C as it currently exists must be removed from the EPA website. Suggestions for expanding the list of References are attached as Exhibit D. Additional discussions will be needed to determine how the language of Exhibit C must be changed to properly reflect the status of the science that now exists.

Page 2 of Exhibit C also directs the readers to several websites "For More Information." One such site is a Texas USGS website that was, until recently, called "allthingssealcoat." Quietly, this website name and content has been changed in response to objections made by the PCTC in its first DQA challenge on this issue. The problem is that the website is a far cry from being about "all things sealcoat." In reality, it is a website that does little more than self-promote articles authored by Dr. Mahler and Dr. Van Metre of the USGS. Any publication or DQA

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[63] DeMott, R.P., Gauthier, T.D., Wiersema, J.M. and Crenson, G. (2010). PAHs in Austin Sediments after a Ban on Pavement Sealers. Environmental Forensics, 11:4, 372-382.

[64] O'Reilly, K., Pietari, J. and Boehm, P. (2013). Parsing Pyrogenic PAHs: Forensic Chemistry, Receptor Models, and Source Control Policy. Integrated Environmental Assessment and Management. DOI: [10.1002/ieam.1506

challenge that question the USGS on the issue of RTS has been ignored at this website as though they did not exist, even peer reviewed articles.

Another link included in the "For More Information" section directs the public to the EPA's June 14, 2012 webinar entitled "Stormwater, Coal-Tar Sealcoat and Polycyclic Aromatic Hydrocarbons." It suffers from the same problems as the "allthingssealcoat" website – all information and studies that challenge the agenda to ban RTS are ignored. Specifically, the website provides access to four PowerPoint presentations given by long time core members of the "ban RTS" affinity group: Barbara Mahler, Allison Watts, Judy Crane and Mateo Scoggins. All of them are advocates for RTS bans, without regard to scientific information that contravenes their advocacy position, and have been pursuing this agenda for many years. Flaws in the scientific analyses offered by each webinar presenter have been repeatedly pointed out in numerous peer reviewed articles and in the three DQA challenges filed by the PCTC in 2013. What is missing from the webinar, of course, is any reference to articles such as those published by O'Reilly and DeMott, or to relevant government studies conducted by the TCEQ, ATSDR and Texas Department of Health. Furthermore, it goes without saying that Drs. O'Reilly and DeMott were not invited by the EPA to speak at this webinar (despite each of them having many PowerPoints of their own on these topics), nor was anyone else contacted who might have been able to share the perspective of industry. One perspective was good enough for the EPA – and that perspective did not involve the stakeholder with the most information about the topic.

All Information Quality Guidelines, from the overarching OMB Guidelines to the Agency-specific EPA Guidelines, clearly assert that this type of bias is not to be tolerated. For this reason, any reference by the EPA to the "allthingssealcoat" web site (recently renamed) and the EPA "Stormwater" webinar should be eliminated from any future EPA publication unless references are made to other websites which contain more complete information regarding RTS. For example, if the two links above are not deleted, at least one additional link should be added to EPA publications which direct readers to the PCTC website.[65] The EPA's Information Quality Guidelines as well as its CADDIS require nothing less. The public and other scientists should be allowed to decide for themselves if the arguments and hypotheses offered by the USGS are valid. Self-serving press releases and websites might be a good way for advocates to pursue a certain agenda and obtain a political victory, but it has nothing to do with good science.

CONCLUSION

The outdated and inaccurate RTS summaries offered by the EPA in Exhibits B and C must be corrected, and quickly. Time is of the essence because policy makers and resource managers across the country are being asked to consider this issue, and they likely will continue to rely upon the EPA to provide them with unbiased and accurate summaries, as well as links to the most current peer reviewed studies and information. These responsibilities assumed by the EPA go far beyond mere citation of USGS websites and publications that have been challenged within various peer reviewed articles.

[65] http://www.pavementcouncil.org/scientific-journals

The first step for the EPA is to remove Exhibits B and C from its websites, immediately, so that appropriate amendments and updates can be implemented with input not only from the USGS, but from industry as well, such as the PCTC. The second step is to draft updated and unbiased summaries regarding the science that now exists with respect to RTS. An important part of that process will be the inclusion of Reference sections and links that allow members of the public to understand the nature and complexities of the scientific debates that are ongoing. EPA knows well how to evaluate science information. CADDIS is just one example of well thought out and comprehensive guidance. The guidance discussed above about realistic evaluations of potential risks to aquatic life from PAHs in sediments is another.

As always, the PCTC stands ready to work with the EPA in disseminating to the public accurate, unbiased and scientifically valid information regarding RTS. PCTC remains hopeful that you will reach out and contact us for this purpose.

Respectfully Submitted,

Anne LeHuray
Executive Director PCTC

Exhibit A

CADDIS: The Causal Analysis/Diagnosis Decision Information System

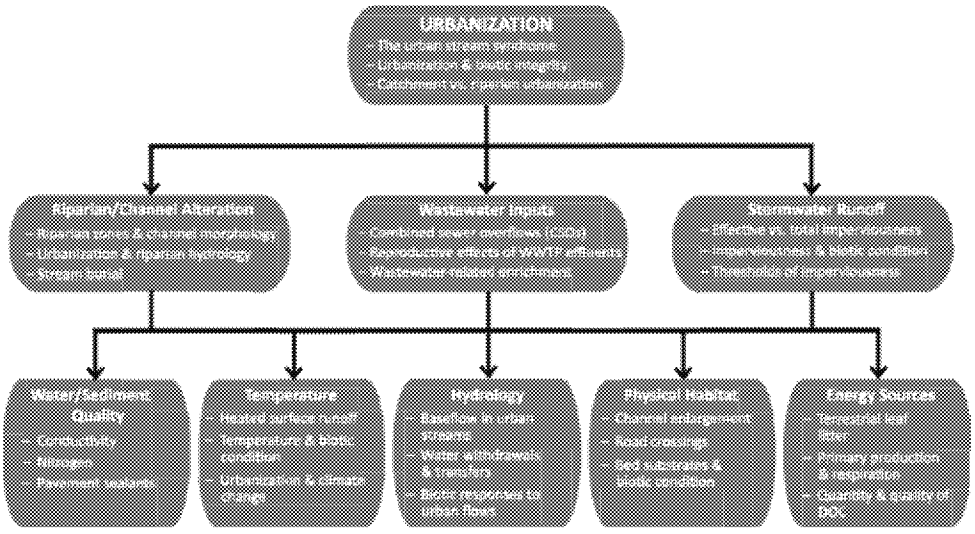
Volume 2: Sources, Stressors & Responses

http://www.epa.gov/caddis/ssr_urb_intro.html

http://www.epa.gov/caddis/ssr_urb_intro.html

EPA

CADDIS Volume 2: Sources, Stressors & Responses



Click on any heading to see more detailed information about that pathway.
Click on subheadings to read more about highlighted topics under each heading.

Urbanization is an increasingly pervasive land cover transformation that significantly alters the physical, chemical and biological environment within surface waters.

The diagram above provides a simple schematic illustrating pathways through which urbanization may affect stream ecosystems. **Riparian/channel alteration**, **wastewater inputs** and **stormwater runoff** associated with urbanization can lead to changes in five general stressor categories: **water/sediment quality**, water **temperature**, **hydrology**, **physical habitat** within the channel, and basic **energy sources** for the stream food web.

This module is organized along these pathways. You can learn more about urban stream sources and stressors by clicking on these headings in the diagram above. You can click on subheadings within each shape to learn about specific topics in greater detail. To return to this organizational diagram from any point in the module, simply click on the Urbanization link in navigation bar (at left) or in the breadcrumbs (at top).

You also can download a **PDF version of the Urbanization module** (44pp, 3MB, About PDF), and view a complete list of references cited in the module.

Last updated on Tuesday, July 31, 2012

Exhibit B
EPA Publication #1

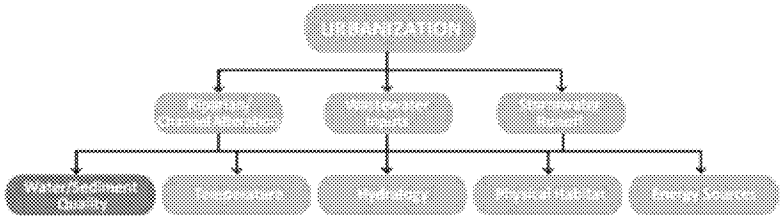
CADDIS Volume 2: Sources, Stressors & Responses

Pavement sealants & PAHs
&
References for the Urbanization Module

http://www.epa.gov/caddis/ssr_urb_wsq4.html
&
http://www.epa.gov/caddis/ssr_urb_ref.html

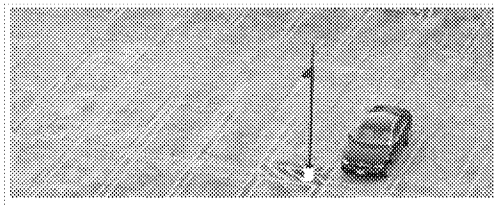
EPA

CADDIS Volume 2: Sources, Stressors & Responses



Pavement sealants & PAHs

Polycyclic aromatic hydrocarbons (PAHs) are common pollutants in urban streams, resulting from numerous transportation-related sources including **oil leakage, vehicle exhaust, tire and brake wear, and pavement erosion**. Many studies have shown that these compounds can **adversely affect stream biota** (e.g., Maltby et al. 1995, Pinkney et al. 2004).



Courtesy of U.S. EPA

Pavement sealants are routinely applied to parking lots and driveways to protect the underlying surfaces, and these sealants can be significant sources of PAHs. For example:

- PAH concentrations were **65 times higher** in runoff from coal-tar seal-coated parking lots versus unsealed parking lots (Mahler et al. 2005).
- PAH concentrations in stream sediments were **3.9 to 32 mg kg^{-1} higher** downstream of coal-tar seal-coated parking lots versus upstream reference sites (Scoggins et al. 2007).

Scoggins et al. (2007) examined the effect of these sealcoats on benthic macroinvertebrate assemblages. They found that:

- Average **macroinvertebrate densities were two times higher** at sites upstream of seal-coated parking lots.
- **Chironomid density decreased** at sites downstream of seal-coated parking lots, whereas **oligochaete density usually increased**.
- **Increases in pool habitat PAH sediment toxicity units** between sites upstream and downstream of seal-coated parking lots explained **decreases in macroinvertebrate richness and density** (Fig 25).

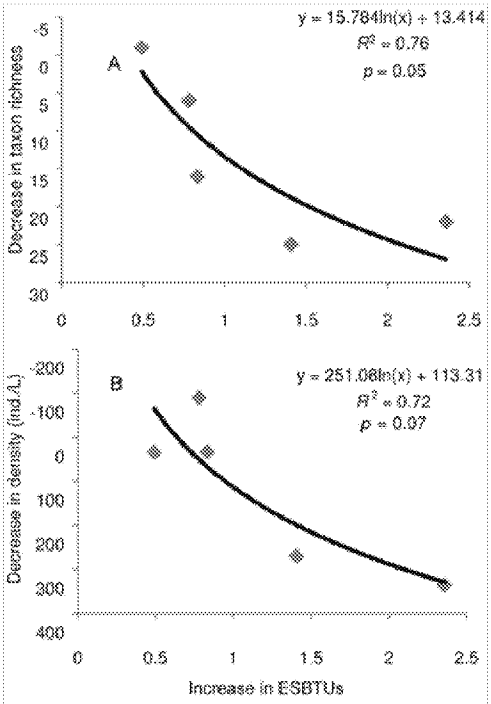


Figure 25. Regression plot of the decrease in (A) macroinvertebrate richness and (B) density between sites upstream and downstream of seal-coated parking lots, as a function of the increase in PAH equilibrium partitioning sediment benchmark toxicity units (ESBTUs) in pool sediments between those sites. ESBTUs were based on 16 EPA priority PAH pollutants; values > 1 suggest toxicity.

From Scoggins M et al. 2007. Occurrence of polycyclic aromatic hydrocarbons below coal-tar-sealed parking lots and effects on stream benthic macroinvertebrate communities. Journal of the North American Benthological Society 26(4):694-707. Reprinted with permission.

Click below for more information on specific topics

Urbanization & connectivity | Nitrogen in urban streams | Pavement sealants

Last updated on Tuesday, July 31, 2012

EPA

CADDIS Volume 2: Sources, Stressors & Responses

References for the Urbanization Module

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Exhibit C
EPA Publication #2

STORMWATER BEST MANAGEMENT PRACTICE:
Coal-Tar Sealcoat, Polycyclic Aromatic Hydrocarbons, and Stormwater Pollution
EPA 833-F-12-004
November 2012

http://www.epa.gov/npdes/pubs/coaltar.pdf

United States Environmental Protection Agency

Stormwater Best Management Practice

Coal-Tar Sealcoat, Polycyclic Aromatic Hydrocarbons, and Stormwater Pollution

Minimum Measure

Pollution Prevention/Good Housekeeping

What Is Coal-Tar Sealcoat?

Coal-tar sealcoat is a type of sealant used to maintain and protect driveway and parking lot asphalt pavement. Coal-tar sealcoat typically contains 20 to 35% coal tar pitch, a byproduct of the steel manufacturing industry, which is 50% or more polycyclic aromatic hydrocarbons (PAHs) by weight.

Could Coal-Tar Sealcoat Be a Concern for Stormwater?

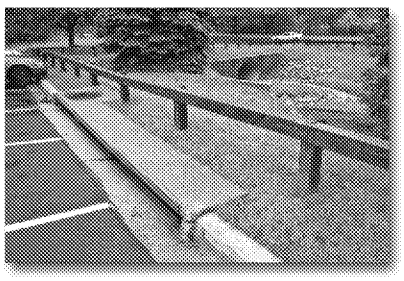
Studies found that PAHs are significantly elevated in stormwater flowing from parking lots and other areas where coal-tar sealcoats were used as compared to stormwater flowing from areas not treated with the sealant. For example, one study found the amount of PAHs in stormwater runoff was 65 times higher from parking lots sealed with coal-tar sealant vs. stormwater from unsealed parking lots. Another study found that coal-tar sealcoat is the largest source of PAHs to 40 urban lakes (Van Metre and Mahler, 2010). PAHs from coal-tar sealcoat may accumulate in the sediment of stormwater ponds, requiring expensive disposal of the dredged PAH-contaminated sediment.

PAHs are of concern because of their harmful impacts on humans and the environment. They are persistent organic compounds, and several PAHs are known or probable human carcinogens and toxic to aquatic life.

What Are States and Municipalities Doing to Address PAHs from Coal-Tar Sealcoat?

Several states and cities have taken action to address PAHs from coal-tar sealcoat. The following are some notable examples:

- The city of Austin, Texas banned the sale and use of coal-tar containing pavement sealants in 2005: http://austintexas.gov/department/coal-tar
- The District of Columbia banned the sale and use of coal-tar sealcoat in 2009: http://green.dc.gov/coaltarban
- In 2009, Minnesota restricted state agencies from purchasing undiluted coal tar-based sealant and directed its Pollution Control Agency to study the environmental effects of coal tar-based sealants and to develop management guidelines: www.pca.state.mn.us/index.php/water/water-types-and-programs/stormwater/municipal-stormwater/restriction-on-coal-tar-based-sealants.html



- Washington State banned the sale of coal-tar pavement sealants on January 1, 2012 and banned the use of such sealants after July 1, 2013: https://fortress.wa.gov/ecy/publications/summarypages/1104021.html

Alternatives to Coal-Tar Sealcoat

Pavement options such as pervious concrete, permeable asphalt and paver systems do not require sealants. These types of pavements allow for stormwater to naturally infiltrate, resulting in decreased runoff.

Office of Water, 4203M
www.epa.gov/npdes/pubs/coal-tar.pdf
www.epa.gov/npdes/stormwater/menuofbmps

EPA 833-F-12-004
November 2012

For More Information

For more information you can watch EPA's webinar *Stormwater, Coal-Tar Sealcoat and Polycyclic Aromatic Hydrocarbons* available at: http://cfpub2.epa.gov/npdes/courseinfo.cfm?program_id=0&outreach_id=645&schedule_id=1169.

For information on assessing the toxicity of PAHs in sediment see: www.epa.gov/nheerl/download_files/publications/PAHESB.pdf from EPA's Office of Research and Development.

Additionally, you can visit the USGS webpage on PAHs and coal-tar-based sealcoat: http://tx.usgs.gov/coring/allthingssealcoat.html.

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EPA's Integrated Risk Information System (IRIS): www.epa.gov/IRIS/

ED_006238_00000702-00056

Exhibit D

PUBLICATIONS OF SCIENTIFIC STUDIES OF REFINED TAR-BASED PAVEMENT SEALERS (RTS) IN THE ENVIRONMENT

SPONSORED BY THE PAVEMENT COATINGS TECHNOLOGY COUNCIL

(REV. APRIL 2014)

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PUBLICATIONS OF SCIENTIFIC STUDIES OF TAR-BASED SEALANTS IN THE ENVIRONMENT SPONSORED BY THE PAVEMENT COATINGS TECHNOLOGY COUNCIL

(REV. APRIL 2014)

Peer Reviewed Papers in Science Journals:

O'Reilly, K., Ahn, S., Pietari, J. and Boehm, P. (2014). Use of Receptor Models to Evaluate Sources of PAHs in Sediments. *Polycyclic Aromatic Compounds*. Awaiting DOI.

O'Reilly, K. T., Pietari, J. and Boehm, P. D. (2014), Parsing pyrogenic polycyclic aromatic hydrocarbons: Forensic chemistry, receptor models, and source control policy. *Integr Environ Assess Manag*, 10:279–285.

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- May 31, 2013: Topic – The USGS is Using Inaccurate and Misleading Photographs of Fish with Skin Tumors as a Scare Tactic to Promote Advocacy Goals
- September 17, 2013: Topic – USGS claims of health risks are based on a "risk assessment" that exaggerates exposure, selects data for inclusion or omission without explanation, fails to consider the many other sources of PAHs, does not use best-available toxicity estimates, and many other flaws of both omission and commission.

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Exhibit E

TECHNICAL EVALUATION OF VAN METRE AND MAHLER (2010)

Prepared by
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March 25, 2014

Exponent

EXTERNAL MEMORANDUM

TO: Anne LeHuray

FROM: Kirk O'Reilly

DATE: March 25, 2014

SUBJECT: Technical Evaluation of Van Metre and Mahler 2010

Exponent has been evaluating the U.S. Geological Survey's (USGS) research concerning the potential role of refined tar sealer (RTS) since 2009. Mahler, Van Metre and their colleagues have published a series of papers promoting "the Mahler hypothesis" that proposes that RTS is a major source of polycyclic aromatic hydrocarbons (PAHs) in urban sediments. As described in O'Reilly et al 2012, there are number of technical issues that raise questions about the conclusions presented in Mahler et al 2005, Van Metre et al 2009, and Van Metre and Mahler 2010. The 2010 paper introduces the application of EPA's chemical mass balance (CMB) model to assess the hypothesis that RTS is the dominant source of PAHs in the sediments of 40 urban lakes. Because the authors described the results of only 4 of about 200 model runs, our initial evaluation of Van Metre and Mahler 2010 was incomplete. In response to a Freedom of Information Act (FOIA) request, the USGS provided sufficient data to recreate these four model runs, but not the complete CMB output files for any of the runs.

The purpose of this memorandum is to discuss a range of technical issues concerning Van Metre and Mahler's application of CMB.

Key findings include:

- CMB can match mixtures of the proposed sources to sediment PAH profiles whether or not a RTS source term is included.
- When the RTS source profile is changed from one based on parking lot dust to another based on the chemical analysis of RTS, CMB eliminates it as a source from most of the sediments samples considered.

The evaluation indicates that the results of CMB do not provide support for the Mahler hypothesis. Because other researchers (Crane 2013; Witter et al 2014) have begun to apply the methods described in Van Metre and Mahler 2010, the USGS should consider retracting the article.

Comments:

1. **The validity of CMB depends on how closely the inputs meet the strict assumptions underlying the model.**

   The following summarizes the assumptions underlying CMB (Coulter 2004) and why Van Metre and Mahler (2010) fails to address them. Some points are discussed in more detail in subsequent sections.

   I. <u>The composition of each source emission profile is consistent over the period model.</u>
      a. No site-specific emission data was used.
      b. The source profiles used were averages of published data. There was no evaluation of how representative they were to actual sources.
      c. The variability among the literature source profiles was taken into consideration.
      d. The composition of the emission sources used are known not be consistent and change to due to fuel, temperature, oxygen availability and other combustion process conditions (Lima et al 2005).

   II. <u>Chemical species do not react with each other or the environment.</u>
      a. PAH react quickly in the atmosphere so emission chemistry does not represent depositional chemistry (Galarneau 2008; Lima et al 2005). This factor was ignored.
      b. Sealers weather resulting in changes in their PAH profile (O'Reilly et al 2012). This was considered.
      c. Together, this results in a greater chance of identifying the sealers as sources.

   III. <u>All sources that contribute significantly to the receptors have been identified and their profile is known.</u>
      a. A limited set of sources was considered. Evaluation of site-specific sources was not conducted.
      b. As noted there is great uncertainty in whether the source profiles used as input represent actual sources.

   IV. <u>The composition of each source is linearly independent or other sources.</u>
      a. The results present indicate a positive relationship between the mass sourced by sealers and the mass sourced by other sources (R^2=0.63). Samples with more sealer also had more other sources.
      b. This is the opposite of the result expected if sealers were actually a source.

V. Measurement uncertainties are random, uncorrelated, and normally distributed.
   a. This assumption could not be met with most of the source data so a generic uncertainty factor of 40% was applied (Li et al 2003).
   b. This value was based typical analytical precision and ignores the variability in the chemical profiles of potential sources.
   c. Profiles based on a limited set of published data are not expected to be random, uncorrelated, or normally distributed.

2. CMB allows calculations of potential relative source contributions only if the actual sources are known.

This statement is based on the simple mathematical concept that one can only calculate an unknown when there are a sufficient numbers of knowns. Without independent verification that the source inputs used are appropriate and sufficient, CMB output cannot be used to verify the contribution of a given source.

3. Discussion of only 4 of 200 model runs provides an incomplete picture the results of the CMB evaluation.

VanMetre and Mahler (2010) states that 200 CMB model runs were conducted, but only four were discussed in detail. While requested, neither the input parameters nor model output of the others 196 runs have been provided. As highlighted in the following comments, model output based on conditions that should have been run by the authors leads to results that are significantly different than those claimed in the article.

Many of the source profiles used by VanMetre and Mahler are from Li et al (2003). As demonstrated by Li and a subsequent paper (Bzdusek et al. 2004), model outputs are highly dependent on model inputs (Figure 3-1). A discussion of receptor modeling requires presentation of the full range of results.

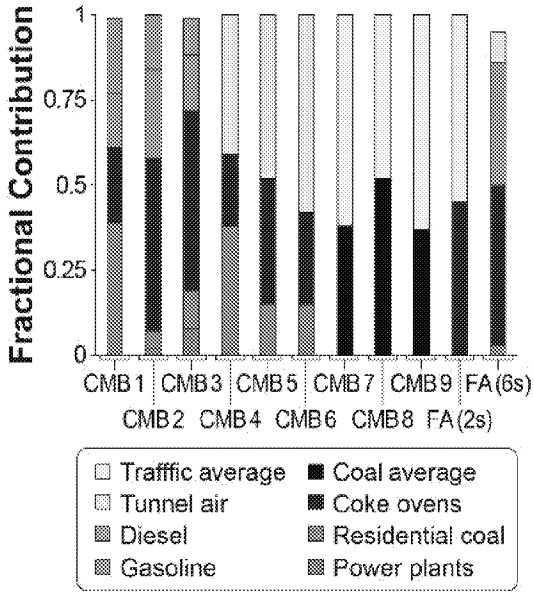

Figure 3-1: Fractional contribution of various PAH source types to sediment of Lake Calumet (Chicago, Illinois) based on nine CMB and two Factor Analysis model runs. Data from Li et al. 2003 and Bzdusek et al. 2004. The range of results highlights the sensitivity of receptor models to the specific inputs.

To resolve this gap, source input profiles used by Van Metre and Mahler were obtained through a FOIA request. Using CMB, we were able to recreate the published Van Metre and Mahler results for the four model runs, A through D, published in 2010. CMB was then rerun either excluding the RTS source or replacing it with an RTS source profile based on another USGS study (Selbig 2009). While Selbig's results were for unfiltered runoff, the low-solubility PAHs should be associated with the particles and thus comparable to the parking dust samples used by Van Metre and Mahler (O'Reilly et al. 2012).

Table 4-1 compares the three model runs. The coefficient of determination, R^2, is the average of the 120 samples. The measured and calculated total PAH concentrations for all 120 samples are shown in Figure 4-1. To compare the goodness of fit between the measured and modeled concentrations, Pearson correlation coefficients, r, were calculated using the results of the 120 sediment samples. The r for each of the three models exceeded 0.998. Note the difference in R^2 between those listed in Table 4-1, which are based on the fitting of individual PAHs within each sample, and r in Figure 4-1 which is fitting the total PAH concentrations.

Table 4-1: Comparison of the average results for three CMB model runs. All input conditions are consistent with Van Metre and Mahler 2010, except for the RTS source profile.

Model Conditions					Average Calculated Source Contribution				
RTS Source Profile	#PAHs	R2	X2	%Mass	RTS	Vehicle	Coal	Oil	Wood
Van Metre 2010	12	0.93	0.94	98.92	61%	26%	6%	1%	6%
Selbig 2009	11	0.92	1.15	103.50	21%	41%	8%	1%	29%
None	12	0.91	1.08	97.10	-	60%	8%	0%	31%

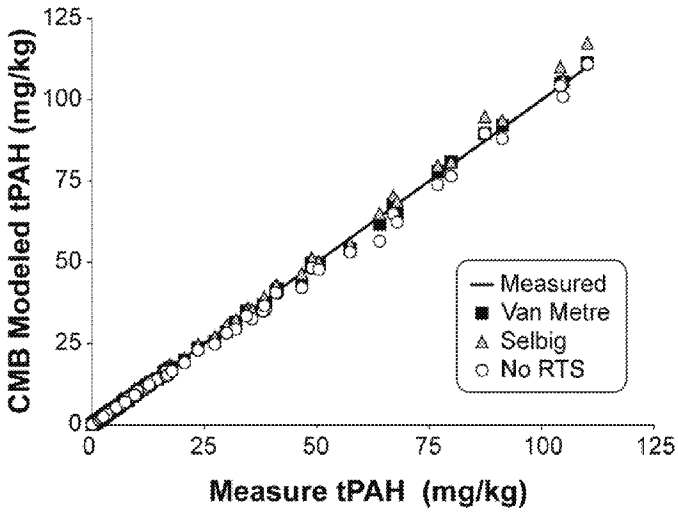

Figure 4-1: Correlations between measured and modeled total PAH concentration, $R^2 > 0.99$, are similar with or with RTS as a CMB source profile.

A collaborator of Van Metre and Mahler used CMB to evaluate sediments collected from storm water detention ponds in Minnesota (Crane 2013) and included model runs with and without RTS as a source input. Crane also found excellent agreement (r>0.99) between CMB model results whether or not RTS was considered a source. While Crane presented statistics that there may be a slightly better fit with the addition of two RTS source profiles than with just two

traffic sources and one wood smoke source, the important finding is that CMB does an excellent job fitting the input source profiles to the sediment source profiles whether or not an RTS related source is included. As the model results are consistent with the null hypothesis, they provide no support for the Mahler hypothesis.

4. **The samples identified as RTS sources are actually dust collected parking lots, which suggest that they are likely a mixture of local sources. CMB indicates minor or no contribution when known RTS sample source profiles are used.**

The RTS profiles used in the 4 CMB model runs were based on the averages of samples collected either from parking lots in Austin, TX (Mahler et al 2005) or lots from 6 cities across the United States (Van Metre et al 2009). The relative contribution of RTS to the PAHs in these samples is unknown, as is the contribution of atmospheric deposition and other urban sources. Unlike the underlying data used to generate the other source profiles, parking lot dust is subject to weathering processes that are similar to those expected for sediment sources (O'Reilly et al 2012). It is interesting to note that while Van Metre and Mahler had source data for fresh RTS and from RTS test plots their use as a source profile was not discussed.

We reran Van Metre and Mahler's Model A replacing the RTS dust samples with average of either the fresh RTS samples or average of the RTS test plot data from Mahler et al 2005. Benzo(e)pyrene was not included as data was not available for the earlier samples. CMB did not identify RTS as a potential source in most of the 120 sediment samples (Table 5-1).

Table 5-1: Summary of results for 3 CMB runs. Consistent with Van Metre's Model A, we first used parking lot dust as the RTS source profile. Based on data from Mahler et al 2005, the second run used the average of 6 samples of fresh RTS and the third, an average of samples collected over 4 weeks from RTS test plots.

	Lot Dust	**Fresh RTS**	**Test Plots**
# of samples with RTS contribution >0%	107	6	15
% RTS contribution			
Min	0.0	0.0	0.0
Max	95	23	33
Mean	46	0.6	1.5
Median	49	0.0	0.0
Average Model Parameters			
R^2	0.93	0.91	0.91
X^2	0.94	1.11	1.10
%Mass	98.9	96.7	96.7

5. **The non-RTS source profiles used by VanMetre and Mahler are mathematical constructs based on the geometric mean of averaged values of PAH ratios taken from 37 articles. The similarity of these constructs to real work sources has not been demonstrated.**

Except for the RTS, Van Metre and Mahler (2010) obtained all source profiles used in the four CMB model runs from the literature. The main source cited, Li et al 2003, also did not measure any actual sources but created profiles based on manipulation of published data from over 20 papers. Much of the data that Li used were not actual sample results, but averages of other data. As noted in the underlying papers cited by Li, the coefficients of variation (CV) or relative standard deviations (SD) of these initial data were high as indicated by a CV>100% or SD>mean. Li did not directly apply the source profiles, but generated geometric means of the ratio between each PAH and benzo(e)pyrene. These ratios were then combined as a PAH profile. In some cases, a partitioning

factor was applied to estimate particle phase concentrations. The number of samples used to generate this average of an average differed between the individual PAHs potentially further skewing the generated profile. The resulting relative standard deviations were so high that Li's initial CMB runs resulted in "inestimable" source contributions for many sediment samples. To resolve this problem, first Li et al. and then Van Metre and Mahler arbitrarily reduced the uncertainty factor used in CMB.

Another problem with the Li data set is that some of the samples were not collected from the environment, but from within emissions pipes. Given the reactivity of PAH in the atmosphere, significant changes in PAH profile would be expected between pre-emission and when associated particles reach sediment.

No one has conducted an evaluation of the relevancy of these calculated profiles to actual sources within the air and watersheds of the lakes studied by VanMetre and Mahler (2010). Without such information, the CMB results have little value.

6. The issue of source collinearity was not adequately addressed.

One of the key assumptions of the CMB model is that source profiles are linearly independent of each other (Coulter 2004). Non-independence or collinearly can be an issue with pyrogenic PAHs due to the similarity of source profiles (O'Reilly et al. 2012). The degree of collinearity depends on the number of source categories, the abundance and variability of fitting species, and the relative contribution of each the source. As conditions vary from sample to sample, it is not possible to state that two or more profiles are overly collinear prior to applying them to a specific sample.

Determining whether collinearly among source profiles impacts model results is an important step in model validation (Watson 2004). CMB output files contain two performance factors that indicate the influence of collinearity. The first is the indicator of whether a source contribution is "estimable." While the model will estimate a source contribution even if a source is determined to be inestimable, it flags each source as either estimable or inestimable. A YES (estimable) indicates that the source contribution estimate combination meets the uncertainty criteria (Coulter 2004). Inestimable sources are caused by excessive similarity or collinearity among the source profiles. The standard errors associated with the estimated contribution of one or more inestimable sources are usually too large to allow an adequate separation of these source contributions to be made. As a means of dealing with inestimable sources is to combine them with other sources, the model suggests estimable liner combinations of inestimable sources. While the combined source results in a fit between sources and samples, it does not

allow differentiation among the contribution estimates of the sources contained in the linear combination.

A second source estimate validation indicator is the Tstat, or ratio of the estimated source contribution to its standard error. A Tstat of greater than two is indicative of a contributing source. A Tstat of less than two suggests the source contribution is lower than the detection limit of the CMB method given the uncertainties associated with the source profile.

A summary of the collinearity indicators from VanMetre and Mahler's Model A is shown in Table 7-1. The RTS profile was not identified as an estimable source any of the 120 sediment samples. While four or five source profiles were provided, no more than two sources were estimable for any of the sediment samples and a majority had no estimable sources. Van Metre's RTS source profile met the Tstat criteria >2.0 for about a third of the samples, and the vehicle exhaust profile met the criteria with the greatest number of sediments samples. To broaden this evaluation, a summary of the collinearity indicators for three other CMB runs discussed in Van Metre and Mahler (2010) are in Table 7-1. The maximum number of samples with RTS as an estimable source was 12. The number of estimable sources identified is consistent with the results presented in Table 2. Van Metre and Mahler's RTS source profile met the Tstat criteria >2.0 for between 44 to 73% of the sediment samples.

CMB's developer admits there are not hard rules concerning how collinearity indicators should be interpreted (Coulter 2004). But the limited number of estimable sources and the few sources meeting the Tstat criteria indicate that the inputs used in this assessment challenge CMB's key assumption that source profiles are linearly independent of each other. Such a finding is not surprising as the chemical similarity of different PAH sources has been identified as an issue which can limit the application of receptor models such as CMB (Galarneau 2008). The problem can be compounded in sediments as weathering that occurs between emission and deposition results in a residual profile of the more stable PAHs (O'Reilly et al. 2012). Without additional consideration of the influence of source collinearity, the results presented in Van Metre and Mahler (2010) are insufficient to support a hypothesis concerning the role of RTS as a PAH source in urban systems.

Table 7-1: Number of estimable sources and Tstat results for Van Metre and Mahler's Model A. The results suggest collinearity among source profiles was not adequately addressed.

Van Metre Model A

Number of samples where source is estimable

	Yes	No
RTS	0	107
Wood	1	64
Coal	13	43
Vehicles	1	107
Fuel oil	36	4

Estimable sources per sample (n=120)

Max	2
0	71
1	47
2	2

Percent of 120 samples Tstat >2.0

	Yes	No
RTS	38%	51%
Wood	2%	52%
Coal	4%	43%
Vehicles	28%	63%
Fuel oil	0%	33%

7. The potential contribution of coal tar from manufactured gas plants (MGPs) was not considered.

Although MGPs have long been known to be an important source of pyrogenic PAHs in the environment (Costa et al 2004) they have been ignored by VanMetre and Mahler. Such plants were typically placed along water bodies and are known sources of sediment contamination. Also, the tars from MGPs were sometimes incorporated into road base, thus spreading the material throughout a region (Hubbard and Draper 1911; Reinke and Glidden 2007) and potentially serving as source of sediment PAHs (Ahrens and Depree 2010).

Review of EPA's list of MGP sites indicates that many of the cities the USGS have been studying have had gas plant operations (EPA 1985). For example, Mahler et al 2005 focused on RTS evaluated suspended solids collected in Austin and Forth Worth, TX but failed to mention that both cities had MGPs. PAH source profiles associated with MGP waste were not included in Van Metre and Mahler's CMB source evaluations.

8. Principal component analysis (PCA) results indicate that the sources used in CMB do not properly represent the actual sources.

PCA is a multivariate approach for evaluating potential sources. When sources and their mixtures are evaluated together, the sources typically appear as end members on a PCA plot. Mixtures should plot within the area bounded by the sources (O'Reilly et al 2014). To evaluate the CMB input and outputs, the 40 lakes data was first run with the proposed source inputs used in Van Meter and Mahler's Model A. Unweathered RTS was also included. As shown in Figure 9-1, few of the sediment samples were within the area bounded by the proposed sources.

Source profiles were then created with combining the sources in the ratio indicated by CMB. These results were then analyzed by PCA. As shown in Figure 9-2, the sediment plots shifted to fit with in the area bounded the modeled identified sources.

These findings highlight two important points. First, they indicate that the source profiles used in CMB do not adequately represent the actual sources as few of the sediment samples plot with the expected area.

Secondly they demonstrate that, while correlation coefficients between the measured and modeled results are high, there are detectable difference in their PAH profiles. While the location of the modeled sediment profiles are consistent with a mixture of the coal, lot dust, and vehicles source profiles indicated by CMB, they do not represent the measured sediment profiles.

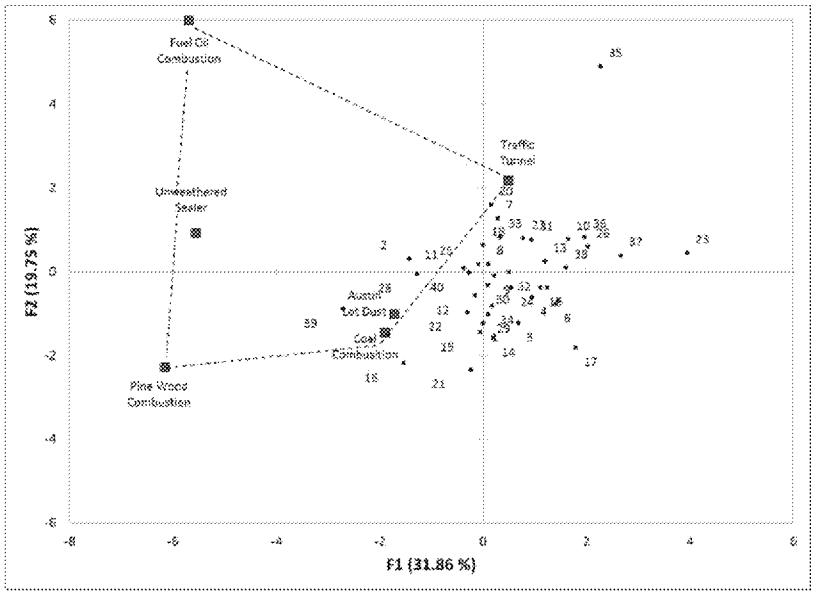

Figure 9-1: PCA of measured sediment profiles and proposed sources.

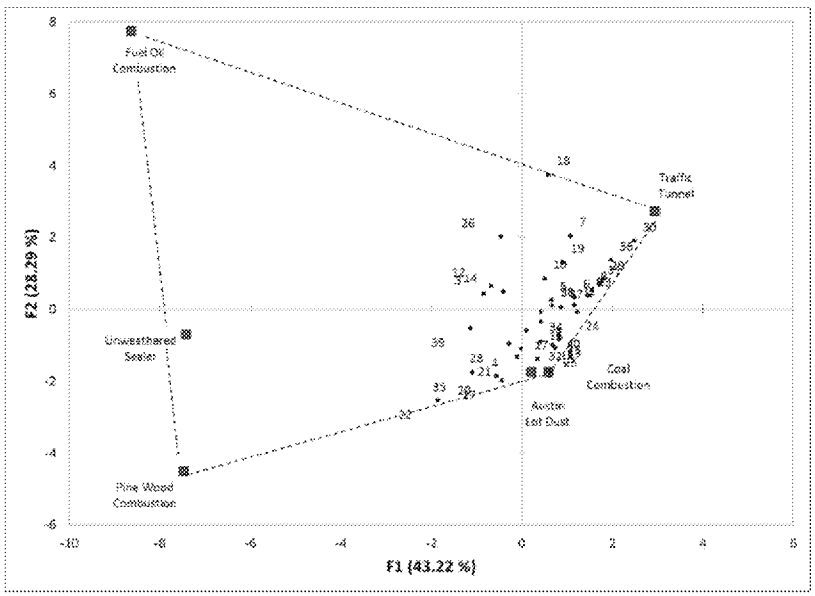

Figure 9-2: PCA of model sediment profiles and proposed sources.

9. Reliance of other researchers on the CMB approach of Van Metre and Mahler (2010) highlights the need for the USGS to acknowledge its limitations

In Crane (2013) and Witter et al (2014), researchers applied Van Metre and Mahler CMB approach with little change to local sediment data sets. Because it has been shown that this approach assigns a RTS contribution to many urban sediments, it is not surprising they obtained similar results. The underlying problems described in this memorandum get lost when researcher defend their results by reliance on what appears to be an USGS approved methodology.

10. The presentation of CMB model results as proven fact in legislative advocacy highlights the need for the USGS to acknowledge the model's limitations

While advocating for a RTS product ban in testimony to Washington State legislators, Van Metre cited the CMB results from a local lake as proof that a problem existed. Similarly, Crane used CMB results to successfully advocate for a product ban in Minnesota. Given the uncertainty in receptor modeling generally, and the weakness in this application, it is critical for agency scientist to accurately describe the meaning of model outputs when they are presented to non-technical policy makers (O'Reilly et al 2013).

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O'Reilly, K., J. Pietari, and P. Boehm. 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, K., J. Pietari, and P. Boehm. 2013. Parsing pyrogenic PAHs: Forensic chemistry, receptor models, and source control policy. Integ Environ Assess and Manage. DOI 10.1002/ieam.1506

O'Reilly, K., S. Ahn, J. Pietari, and P. Boehm. 2014. Use of receptor models to evaluate sources of PAHs in sediment. Polycyclic Aromatic Compounds Journal. Submitted.

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Van Metre, P.C., B.J. Mahler, and J.T. Wilson. 2009. PAHs underfoot: Contaminated dust from coal-tar sealcoated pavement is widespread in the United States. Environ. Sci. Technol. 43(1): 20-25.

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

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Exhibit F

Letter from Myron O. Knudson, Director, Superfund Division, EPA Region 6

to

Ms. Toby Hammett Futrell, City Manager, City of Austin, TX

dated

April 17, 2003

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
REGION 6
1445 ROSS AVENUE, SUITE 1200
DALLAS, TX 75202-2733

April 17, 2003

Ms. Toby Hammett Futrell
City Manager
City of Austin
124 S. Eighth Street, Suite101
Austin, TX 78701

Dear Ms. Futrell:

We have reviewed the Health Consultation prepared by the Texas Department of Health (TDH) and the Agency for Toxic Substances and Disease Registry (ATSDR) for the Barton Springs Pool in Austin, Texas. The U.S. Environmental Protection Agency (EPA) concurs with the overall conclusion of the TDH and the ATSDR that swimming and playing in the Barton Springs Pool do not pose an apparent public health hazard.

We also agree with the specific conclusions and public health action recommendations outlined in the consultation, including:

Conclusions

1. The information reviewed does not indicate that people who swim in the Barton Springs Pool would be exposed to levels of contaminants that would be expected to cause adverse effects.

2. Adverse health outcomes from exposure to soil near the creosote-treated posts used for erosion control near the shallow end of the pool are not likely.

3. The levels of total petroleum hydrocarbons detected in both the water and sediment in the pool are not expected to result in adverse health outcomes.

Public Health Action Recommendations

1. Provide public health education to address any concerns that the public may have concerning the risk associated with swimming in the pool.

2. Investigate the potential for the creosote-treated posts near the shallow end of the pool to serve as a source of polynuclear aromatic hydrocarbons in soil.

3. Continue to monitor the pool for total petroleum hydrocarbons.

ED_006238_00000702-00078

If you have any questions, please contact Jon Rauscher or Don Williams of my staff at (214) 665-8513 or (214) 665-2197, respectively.

Sincerely yours,

Myron O. Knudson
Director
Superfund Division

PCTC
PAVEMENT COATINGS TECHNOLOGY COUNCIL

PUBLICATIONS OF SCIENTIFIC STUDIES OF TAR-BASED SEALANTS IN THE ENVIRONMENT SPONSORED BY THE PAVEMENT COATINGS TECHNOLOGY COUNCIL

(REV. APRIL 9, 2016)

Peer Reviewed Papers in Science Journals:

O'Reilly, K., Ahn, S., Pietari, J. and Boehm, P. (2015). Use of Receptor Models to Evaluate Sources of PAHs in Sediments. *Polycyclic Aromatic Compounds*. 35:1, 41-56.

O'Reilly, K. T., Pietari, J. and Boehm, P. D. (2014), Parsing pyrogenic polycyclic aromatic hydrocarbons: Forensic chemistry, receptor models, and source control policy. *Integr Environ Assess Manag*, 10:279–285.

O'Reilly, K., Pietari, J. and Boehm, P. (2012). A Forensic Assessment of Coal Tar Sealants as a Source of Polycyclic Aromatic Hydrocarbons in Urban Sediments. *Environmental Forensics,* 13:185-196.

DeMott, R.P., Gauthier, T.D., Wiersema, J.M. and Crenson, G. (2010). PAHs in Austin Sediments after a Ban on Pavement Sealers. *Environmental Forensics*, 11:4, 372-382.

Post-Publication Peer Reviews, Letters to the Editor, & Responses Published in Science Journals:

LeHuray, A. (2015) Letter to the Editor in response to Bales. *Integr Environ Assess Manag*. 11(2):185-187. DOI: 10.1002/ieam.1619.

Gauthier, T.D. and DeMott, R.P. (2015). Comment on "Coal-tar pavement sealant use and polycyclic aromatic hydrocarbon contamination in urban stream sediments." *Physical Geography*. 36(1) pp 84-86 http://dx.doi.org/10.1080/02723646.2014.981779

O'Reilly, K. (2015). Letter to the Editor concerning "Source apportionment and distribution of polycyclic aromatic hydrocarbons, risk considerations, and management implications for urban stormwater pond sediments in Minnesota, USA." *Archives of Environmental Contamination and Toxicology*. 68(1) pp 1-3. DOI: 10.1007/s00244-014-0094-7.[1]

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.

[1] Letter to the editor was peer reviewed by the journal.

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O'Reilly, K., Pietari, J. and Boehm, P. (2014). Author's Reply to Van Metre and Mahler's Letter to the Editor on "Parsing pyrogenic polycyclic aromatic hydrocarbons: Forensic chemistry, receptor models, and source control policy." *Integr Environ Assess Manag*. 10(4):489-491. DOI: 10.1002/ieam.1556.

O'Reilly, K., Pietari, J. and Boehm, P. (2014). Author's Reply to Crane's Letter to the Editor on "Parsing pyrogenic polycyclic aromatic hydrocarbons: Forensic chemistry, receptor models, and source control policy." . *Integr Environ Assess Manag*. 10:325–326. DOI:10.1002/ieam.1548

O'Reilly, Kirk (2014). Response to authors' reply on "Coal-tar-based sealcoated pavement: A major PAH source to urban stream sediments" *Environmental Pollution* 191:264-265.

O'Reilly, Kirk (2014). Article Title Misstates the Role of Pavement Sealers. Letter to the Editor of *Environmental Pollution* 191:260-261.

Magee, Brian and Janet Keating-Connolly (2014). Comment on "Cancer Risk from Incidental Ingestion Exposures to PAHs Associated with Coal-Tar-Sealed Pavement". *Environmental Science & Technology*, *48* (1), pp 868–869.

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

DeMott, R.P.; Gauthier,T.D. (2006) Comment on "Parking lot sealcoat: An unrecognized source of urban polycyclic aromatic hydrocarbons." *Environ. Sci. Technol*. 2006, *40*(11), 3657-3658

Post-Publication Peer Review Reports:

Exponent (2015a). Review of: Acute Toxicity of Runoff from Sealcoated Pavement to Ceriodaphnia dubia and Pimephales promelas. Report prepared for the Pavement Coatings Technology Council. Available at http://www.webcitation.org/6fNGp6sm0

Exponent (2015b). Review of: Exposure to Runoff from Coal-Tar-Sealed Pavement Induces Genotoxicity and Impairment of DNA Repair Capacity in the RTL-W1 Fish Liver Cell Line. Prepared for the Pavement Coatings Technology Council. 5p. Available at http://www.webcitation.org/6fSITlyP3

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." Unpublished comment available at http://www.webcitation.org/6fK2uw0Ox

O'Reilly, K. (2014). *Technical Evaluation of Van Metre and Mahler 2010*. Report prepared for PavementCouncil.org by Exponent. Available at http://www.webcitation.org/6fK3aVDrR

Gauthier, T. (2014). *Review of Pavlowsky 2013*. Report prepared for PavementCouncil.org by Environ. Available at http://www.webcitation.org/6flisfTfU Condensed version published as a comment in Physical Geography (Gauthier and DeMott, 2015).

Magee, B. (2014). PAH Vapor Emissions from Coal Tar Pavement Sealers. Technical memorandum prepared by ARCADIS for the Pavement Coatings Technology Council, dated July 30. 5 p. Available at http://www.webcitation.org/6fKFBNdbm

Magee, B. and Keating-Connolly, J. (2013). *Peer Review of Coal-Tar-Sealed Pavement Risk Assessment*. Report prepared for PavementCouncil.org by ARCADIS. Available at http://www.webcitation.org/6fKBho0I6 . Condensed version published as a comment in Environmental Science & Technology (Magee and Keating-Connolly, 2013).

DeMott, Robert, Thomas Gauthier and Michael Masonjones (2013). *Volatilization of PAHs from Coal-Tar-Sealed Parking Lots*. Report prepared for PavementCouncil.org by Environ. Available at http://www.webcitation.org/6fKES4Z5r

Environ International (2010). Review of "Coal-Tar-Based Parking Lot Sealcoat: An Unrecognized Source of PAH to Settled House Dust" by Mahler et al., published in Environmental Science and Technology, January 2010. Report prepared for PavementCouncil.org by Environ. Available at http://www.webcitation.org/6fK1aX2vx

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. Archived at http://www.webcitation.org/6fK0OLSiZ plus appendices archived at http://www.webcitation.org/6fK0ccc2C

Summaries of Post-Publication Peer Reviews Posted for Public Comment

Summaries of scientific reviews commissioned by PCTC have been posted to PubPeer.com, a web site specifically designed for public post-publication peer reviews of papers published in science journals. The links below are to summaries of individual publications.

https://pubpeer.com/publications/62730EDFFC17A5F85CA9EB7FD04C24#fb42729 (Mahler et al. 2005)
https://pubpeer.com/publications/C3ADDD65D7FDDD9D8F3E06EC0B9A2A#fb4273 (Van Metre et al. 2009)
https://pubpeer.com/publications/DEC6835FF61E589EB95C8597944A7F#fb42759 (Van Metre & Mahler 2014)
https://pubpeer.com/publications/F7AA69C873AB96CA862322CF1929BF#fb42838 (Mahler et al. 2010)
https://pubpeer.com/publications/BEE4406AC9EF33CF9E3E6C238F0EDF (Van Metre & Mahler 2010)
https://pubpeer.com/publications/5EBEB3ACD53C7F2FF65624EC6DDA58 (Williams et al., 2013)
https://pubpeer.com/publications/D11E6D8EA68C093ACB155A821E5DFB (Watts et al., 2010)
https://pubpeer.com/publications/1BC1FF805A0E9DE96ADBA73AC443AD#fb43811 (Crane, 2014)
https://pubpeer.com/publications/C95FA81213FD9D30144C36DD6D3DF9#fb44076 (Witter et al., 2014)
https://pubpeer.com/publications/747B19A6260CA08B9CA4908177268A (Scoggins et al., 2007)
https://pubpeer.com/publications/456CA525683D444D8AE75DB9E88554#fb45568 (Van Metre et al., 2012a and 2012b)
https://pubpeer.com/publications/CA5E52B5AD1819E468B800DB24D261 (Mahler et al., 2015)
https://pubpeer.com/publications/EFBBA26FDD35EBF21FC7A96538B03E#fb46601 (Kienzler et al., 2015)

A problem with the DOI is preventing posting of a comment on another paper. When the problem is resolved, the comment will posted on PubPeer. In the meantime, it can be accessed at this link: http://www.pavementcouncil.org/the-study-of-rts-in-springfield-mo-is-critically-flawed-post-publication-peer-review-of-pavlowsky-2013/

Submissions to Government Authorities:

Information Quality Act Request for Correction of Information Under the U.S. Environmental Protection Agency Information Quality Guidelines. Information requiring correction includes a CADDIS web page and a document titled *Stormwater Best Management Practice: Coal-Tar Sealcoat, Polycyclic Aromatic Hydrocarbons, and Stormwater Pollution*. April 16, 2014. Available at http://www.epa.gov/quality/informationguidelines/iqg-list.html

PCTC (2014). The Great Lakes Coal Tar Sealcoat PAH Reduction Project: Comments and Recommendations of the Pavement Coatings Technology Council. Comments submitted to the EPA Great Lakes Program Office and several state agencies located within EPA Region 5. January 21, 2014. Available at http://www.pavementcouncil.org/blog

Information Quality Act Requests for Correction of Information Under the U.S. Geological Survey Information Quality Guidelines, available at http://www.usgs.gov/info_qual/

- May 15, 2013: Topic – There is No Scientific Basis for the USGS to Claim that RTS is a Major Source of PAHs in U.S. Sediments
- May 31, 2013: Topic – The USGS is Using Inaccurate and Misleading Photographs of Fish with Skin Tumors as a Scare Tactic to Promote Advocacy Goals
- September 17, 2013: Topic – USGS claims of health risks are based on a "risk assessment" that exaggerates exposure, selects data for inclusion or omission without explanation, fails to consider the many other sources of PAHs, does not use best-available toxicity estimates, and many other flaws of both omission and commission. Request that the USGS publication regarding the risk assessment (Williams et al., 2012) be retracted.
- October 17, 2014: Topic – Comprehensive Review and Evaluation of the USGS application of EPA's Chemical Mass Balance (CMB) model. Request that the USGS publication (Van Metre and Mahler, 2010) regarding the modeling exercise be retracted.

DeMott, Robert (2004). Review and Evaluation of Coal Tar Emulsion Sealers and Potential Runoff Transport of Polycyclic Aromatic Hydrocarbons. Report prepared for Pavement Coatings Technology Center of the University of Nevada-Reno by Environ, submitted to the City of Austin, TX January 8, 2004. Available at http://www.pavementcouncil.org/scientific-journals.

Articles Published in Magazines for Professionals:

LeHuray, A. (2014). Understanding Sealer Basics. Pavement Maintenance Magazine March 2014 (published online Feb. 25, 2014).

Pietari, J., O'Reilly, K. and Boehm, P. (2010). Polycyclic Aromatic Hydrocarbons in Stormwater and Urban Sediments: A Review. *Stormwater* Magazine. September 2010.

Presentations at Upcoming & Recent Scientific Meetings:

LeHuray, A. (2015). White Hat Bias in the Environmental Sciences. Presentation to be made at the 36th annual meeting of the Society of Environmental Toxicology and Chemistry (SETAC), Salt Lake City November 2015.

Ahn, S. and O'Reilly, K. (2014). The influence of source selection on Chemical Mass Balance modeling results: Implications for source control policy. Presentation at the 35th annual meeting of the Society of Environmental Toxicology and Chemistry (SETAC) Vancouver, BC November 10, 2014.

O'Reilly, K. and Ahn, S. (2014). Mass Balance Modeling of Polycyclic Aromatic Hydrocarbons Sources to Urban Sediments. Presentation at the 35th annual meeting of the Society of Environmental Toxicology and Chemistry (SETAC) Vancouver, BC November 10, 2014.

LeHuray, A. (2014). PAHS are Rarely Causes of Impairment in U.S. Clean Water Act Section 303(D) Reports. Presentation at the 35th annual meeting of the Society of Environmental Toxicology and Chemistry (SETAC) Vancouver, BC November 10, 2014.

LeHuray, A. and Beatty, K. (2014). Key Science Issues to be Considered in the IRIS Hazard Assessment of the Index Compound for the PAHs, Benzo(a)Pyrene. Presentation at the NIOSH 2014 Toxicology and Risk Assessment Conference (TRAC), Cincinnati, OH April 7-10, 2014.

Magee, B. and Keating-Connolly, Janet (2013). Research-Based Input Parameters for Risk Assessment of Coal-Tar-Based Pavement Sealants. Presentation at the 34th annual meeting of the Society of Environmental Toxicology and Chemistry (SETAC) Nashville, TN November 17-21, 2013.

O'Reilly, K., Mudge, S. and Boehm, P. (2013). Receptor Models for PAH Source Characterization: Opportunities and Limitations. Presentation at the 34th annual meeting of the Society of Environmental Toxicology and Chemistry (SETAC) Nashville, TN November 17-21, 2013.

Pietari, J., Ahn, S., O'Reilly, K. and Boehm, P. (2013) Parsing Pyrogenic PAHs—Urban Background or Refined Tar Products? Presentation at the 29th Annual International Conference on Soils, Sediments, Water, and Energy, October 21-24, 2013, Amherst, MA.

O'Reilly, K., Ahn, S., Pietari, J. and Boehm, P. (2013). Use of Receptor Models to Evaluate Sources of PAHs in Sediments. Presentation at the 24th meeting of the International Symposium on Polycyclic Aromatic Compounds (ISPAC 2013) in Corvallis, Oregon USA September 8-12, 2013.

Magee, B. and Keating-Connolly, Janet (2013). Risk Assessment for Coal Tar-Based Pavement Sealants. Presentation at the 24th meeting of the International Symposium on Polycyclic Aromatic Compounds (ISPAC 2013) in Corvallis, Oregon USA September 8-12, 2013.

O'Reilly K, Pietari J and Boehm P. (2012). Use of Alkyl Polycyclic Aromatic Hydrocarbon Data in Evaluating the Contribution of Pavement Sealers to Urban Sediments. Abstract and Platform Presentation at the 2012 annual meeting of the *Society of Environment Toxicology and Chemistry (SETAC).*

Feldpausch, A. and Schoof, R. (2012) Development of a residence-specific, health-based screening criterion for benzo(a)pyrene in indoor dust. Abstract of presentation at the 2012 annual meeting of the *International Society of Exposure Science.*

LeHuray, A.P. (2012). Bans of Pavement Sealers Have Demonstrably Absent Environmental Risk Reduction Benefits but Foreseeable and Knowable Economic Harms. Managing for a Healthy and Sustainable Chesapeake Bay: Human and Ecological Risk: *Joint Meeting of the National Capital Area Chapters of the Society of Environment Toxicology and Chemistry (SETAC) and the Society for Risk Analysis (SRA).* College Park, MD, April 23-24, 2012

O'Reilly, K., Pietari, J. and Boehm, P. (2011). Managing Risks: Will banning pavement sealers have the desired effect? Abstract and Poster Presented at the *32nd Annual Meeting of the Society of Environment Toxicology and Chemistry (SETAC)*, Boston, Nov. 2011.

DeMott, R.P. and Gauthier, T.D. (2011). Use of Mass Balance Bounding Estimates and Sensitivity Analysis to Prioritize PAH Inputs in Urban Systems. Abstract and Poster Presented at the *32nd Annual Meeting of the Society of Environment Toxicology and Chemistry (SETAC)*, Boston, Nov. 2011.

Pietari, J., O'Reilly, K. and Boehm, P. (2011). Environmental Forensics for PAH Source Management: Pavement Sealants and Sediments. Abstract and Poster Presentated at the *Sixth International Conference on Remediation of Contaminated Sediments*, New Orleans, LA Feb. 2011.

O'Reilly, K., Pietari, J. and Boehm, P. (2010). PAHs in Urban Sediments: Forensics Approaches for Assessing the Relative Contribution of Atmospheric Deposition. Abstract and Platform Presentation at the *31st Annual Meeting of the Society of Environment Toxicology and Chemistry (SETAC),* Seattle, Nov. 2010.

Gauthier, T.D. and DeMott, R.P. (2008). Analysis of PAH Concentrations Detected in Austin Texas Stream Sediments Following a Ban on the Use of Coal Tar Sealers. Abstract of Presentation Made at the *29th Annual Meeting of the Society of Environmental Toxicology and Chemistry (SETAC),* Tampa, Nov. 2008.

www.pavementcouncil.org

PCTC

PAVEMENT COATINGS TECHNOLOGY COUNCIL

March 3, 2016

Alan D. Thornhill, Director,
Office of Science Quality and Integrity
U.S. Geological Survey
MS 911 National Center, Reston, VA 20192

Via Email: athornhill@usgs.gov

Subject: Post-Publication Peer Reviews

Dear Dr. Thornhill,

As part of product stewardship programs, it has long been the practice of the private sector to seek expert reviews of scientific publications that touch on products of concern to a company or trade association. Nowadays, such reviews have come to be called post-publication peer reviews (PPPRs), with web sites such as PubPeer and PubMed Commons set up specifically for PPPRs of papers published in science journals. You are, of course, aware that the Pavement Coatings Technology Council (PCTC) has been following standard industry practice by commissioning PPPRs of the work of Drs. Mahler and Van Metre, both employed as research hydrologists by the USGS. PCTC has also commissioned PPPRs of papers by other authors whose work has attempted to build on that of Mahler and Van Metre. You are also aware that PCTC has used some of the PPPRs as the basis of Information Quality Requests for Correction (RfCs) and Requests for Reconsideration (RfRs) filed with the USGS.

In the case of the RfCs and RfRs filed by PCTC, the process has proved torturous, opaque, and glacially slow. Judging from the information available in the Department of Interior's closed scientific integrity case data base, that process seems equally problematic. Yet PCTC believes that if the Office of Science Quality and Integrity (OSQI) were to evaluate the body of work of Drs. Mahler and Van Metre as a whole, OSQI would understand PCTC's contention that the USGS is risking its reputation for scientific integrity and excellence by continuing to defend indefensible advocacy research. From the first paper (Mahler et al., 2005), in which confirmation bias is documented, citation bias is rampant, and white hat bias is proclaimed[1] to more recent

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papers that have involved failure to even recognize prominent and proximate sources of PAHs[2] as well as use of unrepresentative waters and irrelevant species to claim potential ecological harm,[3] the body of work documents a program of advocacy research that is inimical to development of sound public policy. Rather than recite the flaws of individual papers in this letter, the attached tables provide links to summaries of PPPRs of each paper posted on PubPeer. These summaries are public and PCTC welcomes substantive comments.

PCTC would like to point the USGS to its responsibilities as a research institution that encourages its researchers to publish in science journals, as explained in the guidelines of the Committee on Publication Ethics (COPE) included as an attachment to this letter. The guidelines point out that research institutions have a responsibility to ensure the reliability of the published research record, and also suggest that, when a research institution finds a loss of research integrity, it should review all the researchers' publications.

Thank you for your attention to this matter.

Very truly yours

Anne P. LeHuray, Ph.D.
Executive Director

Attachments

cc: William Werkheiser, Acting Deputy Director USGS whwerkhe@usgs.gov
William Guertal, Acting Associate Director Water Resources, USGS wguertal@usgs.gov

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[1] "Previously identified urban sources of PAHs, such as automobile exhaust and atmospheric deposition, have been difficult to control or even quantify because of their nonpoint nature. In contrast, sealed parking lots are point sources, and use of the sealant is voluntary and controllable." Mahler et al. (2005), p. 5565.
[2] The Holly Street Power Plant and its probable influence on sediment PAHs in the adjacent cooling pond (Lady Bird Lake). Van Metre & Mahler (2014).
[3] Mahler et al. (2015), Kienzler et al. (2015).

Table 1
Citations and Links to Post-Publication Peer Reviews of Papers Published by USGS Authors on the Topic of Refined Coal Tar-Based Sealants (RTS)

Publication	Link to PPPR on PubPeer.com
Kienzler, A., Mahler, B. J., Van Metre, P. C., 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. Science of The Total Environment, 520(0), 73-80. doi:http://dx.doi.org/10.1016/j.scitotenv.2015.03.005	https://pubpeer.com/publications/EFBBA26FDD35EBF21FC7A96538B03E#fb46601
Mahler, B. J., Ingersoll, C. G., Van Metre, P. C., Kunz, J. L., & Little, E. E. (2015). Acute Toxicity of Runoff from Sealcoated Pavement to *Ceriodaphnia dubia* and *Pimephales promelas*. Environmental Science & Technology. doi: 10.1021/acs.est.5b00933	https://pubpeer.com/publications/CA5E52B5AD1819E468B800DB24D261
Mahler, B. J., P. Van Metre, T. J. Bashara, J. T. Wilson, and D. A. Johns, 2005, Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons: Environmental Science & Technology, v. 39, p. 5560 – 5566.	https://pubpeer.com/publications/62730EDFFC17A5F85CA9EB7FD04C24#fb42729
Mahler, B. J., Van Metre, P., Wilson, J. T., Musgrove, M., Burbank, T. L., Ennis, T. E., & Bashara, T. J. (2010). Coal-tar-based parking lot sealcoat: an unrecognized source of PAH to settled house dust. Environmental Science & Technology, 44, 894 – 900. doi:10.1021/es902533r	https://pubpeer.com/publications/F7AA69C873AB96CA862322CF1929BF#fb42838

Van Metre, P. C., & Mahler, B. J. (2014). PAH Concentrations in Lake Sediment Decline Following Ban on Coal-Tar-Based Pavement Sealants in Austin, Texas. Environmental Science & Technology. doi:10.1021/es405691q	https://pubpeer.com/publications/DEC6835FF61E589EB95C8597944A7F#fb42759
Van Metre, P. C., Majewski, M. S., Mahler, B. J., Foreman, W. T., Braun, C. L., Wilson, J. T., & Burbank, T. L. (2012b). PAH volatilization following application of coal-tar-based pavement sealant. Atmospheric Environment, 51, 108-115. doi:doi:10.1016/j.atmosenv.2012.01.036	https://pubpeer.com/publications/456CA525683D444D8AE75DB9E88554#fb45568
Van Metre, P., & Mahler, B. J. (2010). Contribution of PAHs from coal–tar pavement sealcoat and other sources to 40 U.S. lakes. Science of the Total Environment, 409, 334 – 344. doi:10.1016/j.scitotenv.2010.08.014	https://pubpeer.com/publications/BEE4406AC9EF33CF9E3E6C238F0EDF
Van Metre, P., Mahler, B., & Wilson, J. (2009). PAHs underfoot: contaminated dust from sealcoated pavements is widespread in the United States. Environ Sci Technol, 43, 20-25.	https://pubpeer.com/publications/C3ADDD65D7FDDD9D8F3E06EC0B9A2A#fb4273
Van Metre, P., Majewski, M. S., Mahler, B., Foreman, W. T., Braun, C. L., Wilson, J. T., & Burbank, T. L. (2012a). Volatilization of polycyclic aromatic hydrocarbons from coal-tar-sealed pavement. Chemosphere, 88(1), 1 – 7. doi:doi:10.1016/j.chemosphere.2011.12.072	https://pubpeer.com/publications/456CA525683D444D8AE75DB9E88554#fb45568
Williams, E. S., Mahler, B. J., & Van Metre, P. (2013). Cancer Risk from Incidental Ingestion Exposures to PAHs Associated with Coal-Tar-Sealed Pavement. Environmental Science & Technology, 47, 1101 – 1109. doi:dx.doi.org/10.1021/es303371t	https://pubpeer.com/publications/5EBEB3ACD53C7F2FF65624EC6DDA58

Table 2
Citations and Links to Post-Publication Peer Reviews of Papers on the Topic of Refined Coal Tar-Based Sealants (RTS) Without USGS Authors

Crane, J. L. (2014a). Source Apportionment and Distribution of Polycyclic Aromatic Hydrocarbons, Risk Considerations, and Management Implications for Urban Stormwater Pond Sediments in Minnesota, USA. Arch Environ Contam Toxicol, 66, 176-200. doi:10.1007/s00244-013-9963-8	https://pubpeer.com/publications/1BC1FF805A0E9DF96ADBA73AC443AD#fb43811
Pavlowsky, R. T. (2013). Coal-tar pavement sealant use and polycyclic aromatic hydrocarbon contamination in urban stream sediments. *Physical Geography, 34*(4-05), 392-415. doi:http://dx.doi.org/10.1080/02723646.2013.848393	http://www.pavementcouncil.org/the-study-of-rts-in-springfield-mo-is-critically-flawed-post-publication-peer-review-of-pavlowsky-2013/
Scoggins, M., McClintock, N. L., Gosselink, L., & Bryer, P. (2007). Occurrence of polycyclic aromatic hydrocarbons below coal-tar-sealed parking lots and effects on stream benthic macroinvertebrate communities. Journal of the North American Benthological Society, 26(4), 694-707. doi:10.1899/06-109.1	https://pubpeer.com/publications/747B19A6260CA08B9CA4908177268A
Watts, A. W., Ballestero, T. P., Roseen, R. M., & Houle, J. P. (2010). Polycyclic Aromatic Hydrocarbons in Stormwater Runoff from Sealcoated Pavements. *Environmental Science & Technology, 44*, 8849 – 8854. doi: 10.1021/es102059r	https://pubpeer.com/publications/D11E6D8EA68C093ACB155A821E5DFB
Witter, A. E., Nguyen, M. H., Baidar, S., & Sak, P. B. (2014). Coal-tar-based sealcoated pavement: A major PAH source to urban stream sediments. *Environmental Pollution, 185*(0), 59-68. doi:http://dx.doi.org/10.1016/j.envpol.2013.10.015	https://pubpeer.com/publications/C95FA81213FD9D30144C36DD6D3DF9#fb44076

COPE COMMITTEE ON PUBLICATION ETHICS

Cooperation between research institutions and journals on research integrity cases: guidance from the Committee on Publication Ethics (COPE)

Summary

Institutions and journals both have important duties relating to research and publication misconduct. Institutions are responsible for the conduct of their researchers and for encouraging a healthy research environment. Journals are responsible for the conduct of their editors, for safeguarding the research record, and for ensuring the reliability of everything they publish. It is therefore important for institutions and journals to communicate and collaborate effectively on cases relating to research integrity. To achieve this, we make the following recommendations.

Institutions should:

- have a research integrity officer (or office) and publish their contact details prominently;
- inform journals about cases of proven misconduct that affect the reliability or attribution of work that they have published;
- respond to journals if they request information about issues, such as disputed authorship, misleading reporting, competing interests, or other factors, including honest errors, that could affect the reliability of published work;
- initiate inquiries into allegations of research misconduct or unacceptable publication practice raised by journals;
- have policies supporting responsible research conduct and systems in place for investigating suspected research misconduct.

Journals should:

- publish the contact details of their editor-in-chief who should act as the point of contact for questions relating to research and publication integrity;
- inform institutions if they suspect misconduct by their researchers, and provide evidence to support these concerns;
- cooperate with investigations and respond to institutions' questions about misconduct allegations;
- be prepared to issue retractions or corrections (according to the COPE guidelines on retractions) when provided with findings of misconduct arising from investigations;
- have policies for responding to institutions and other organizations that investigate cases of research misconduct.

Reference
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COPE COMMITTEE ON PUBLICATION ETHICS

Cooperation between research institutions and journals on research integrity cases: guidance from the Committee on Publication Ethics (COPE)

Introduction

Research institutions (such as universities) and scholarly journals have important duties and common interests in terms of research and publication integrity. Institutions are responsible for the conduct of their researchers and for encouraging a healthy research environment that fosters research integrity. Journals are responsible for the conduct of their editors, for safeguarding the research record, and for ensuring the reliability of everything they publish.

Ensuring research and publication integrity requires institutions and journals to cooperate with each other on all aspects of research and publication integrity. Institutions and journals should also promote best practice among researchers, authors, reviewers, and editors (e.g. by policy development and training). Journals should make efforts to detect misconduct before publication (e.g. by screening for plagiarism). Institutions should investigate possible misconduct and journals should correct or retract findings that are invalid or unreliable (because of misconduct or honest errors) to prevent readers from being misled by them.

COPE (the Committee on Publication Ethics) provides a Forum for its member journals to seek advice from other members on troublesome cases. Based on this experience, we have become aware that editors sometimes face difficulties when trying to work with institutions on cases of possible misconduct.[1,2] From the literature, and discussions with institutions, we are also aware that editors do not always respond appropriately when informed by institutions about research misconduct findings.[3]

Given the importance of journal editors and research institutions collaborating effectively on cases relating to publication ethics and research integrity, COPE has developed the following guidelines in consultation with the individuals and institutions listed at the end of the document.

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Scope

This document focuses on the investigation of possible misconduct. However, this is not to belittle the important roles of prevention, education, etc. outlined above. Ideally, journal and institutional policies should cover all aspects. Journal policies should cover not only their responses to misconduct but also to genuine errors; this is described in more detail in the COPE guidelines on retractions.[4]

Similarly, we recognise that other parties, notably funders, have an important role in fostering research integrity and should be informed about research or publication misconduct relating to research they have funded. These guidelines focus on the roles of institutions and journals but we hope they may help funders to develop their own policies to foster research integrity and responsible conduct of research in collaboration with institutions and journals.

Background principles

The COPE Code of Conduct for Journal Editors (Clause 11.4) notes that, in cases of suspected or alleged research or publication misconduct 'editors should first seek a response from those suspected of misconduct. If they are not satisfied with the response, they should ask the relevant employers, or institution … to investigate'.[5]

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COPE therefore advises that investigations into possible misconduct should generally be undertaken by the researcher's institution and not by editors. If a journal has published unreliable or fraudulent information, the editor has a duty to correct or retract this. However, responsibility for disciplining researchers and ensuring they do not commit further misconduct lies with their institution / employer. Therefore, even when faced with apparently strong evidence of misconduct (e.g. plagiarism or inappropriate image manipulation), and a clear need to correct the published record, editors should liaise with institutions and ensure they are informed.

Journals also need to work with institutions when disputes arise between researchers (e.g. about authorship). As with research misconduct, journals are not in a position to investigate or resolve such disputes, but should refer them to the relevant institution(s).

The following guidelines are therefore based on the principle that institutions have responsibilities for the conduct of their researchers, which include investigating possible misconduct and applying appropriate sanctions, while journals are responsible for what they publish.

While these guidelines encourage exchange of information between institutions and journals regarding cases of possible and proven misconduct, we recognize that full disclosure may sometimes be restricted by considerations of confidentiality (e.g. to protect the identity of a whistleblower), conventions about confidential communication (e.g. peer review comments), and legal considerations.

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Defining misconduct

Several definitions of research misconduct are available and are used by different organizations for various purposes. Difficulties sometimes arise when an institution adopts a narrow or strict definition of serious misconduct which does not include practices that, while falling short of this definition, may nevertheless distort the research record. In such instances an institution may find a researcher not guilty of misconduct yet a journal may consider that a correction or retraction is warranted to safeguard readers (e.g. to alert them to redundant publication).

In these guidelines we do not attempt to define serious or lesser types of misconduct, or so-called 'questionable practices', but we use the term misconduct in its broadest sense to include any practice that may affect the reliability of the research record in terms of findings, conclusions, or attribution.

Recommendations for cooperation between research institutions and journals

1. Points of contact

To facilitate communication, research institutions should designate an individual or office with responsibility for research integrity and dealing with misconduct allegations. Contact details of the research integrity officer(s) should be published prominently on the institution's website. This person (or office) should be free from conflicts of interest in relation to individual cases (i.e. have no involvement with any researcher or project being investigated). If a suitable individual without conflicts of interest cannot be found, it may be necessary to involve an external person in investigations.

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Likewise, journals should publish contact details of their editor(s)-in-chief who should act as the point of contact for questions relating to research and publication integrity. COPE also recommends that journals should appoint an ombudsperson to adjudicate in complaints that cannot be resolved internally or that relate to the conduct of the editor.5

2. Informing each other about cases of research and publication misconduct

If an institution investigates a case of misconduct by one of its researchers and finds misconduct that affects the reliability or attribution of published work (e.g. fabrication or plagiarism), the institution should inform the editor(s) of any journal that has published the affected work. If a case is investigated by another organization (e.g. a national body), the institution should pass on the findings to the relevant journal(s). Institutions should also be prepared to answer editors' questions about any investigation or its findings that are necessary to determine the appropriate response (e.g. retraction or Expression of Concern).

Institutions should also notify journal editors and answer their questions in cases of inappropriate publication practices such as authorship misattribution, redundant publication, duplicate submission, failure to disclose competing interests, or misleading reporting (even if these fall outside the institution's definition of research misconduct). Institutions should also encourage researchers to inform journals about honest errors likely to affect the reliability of published work.

Editors should cooperate with investigations and respond to institutions' questions about misconduct allegations.

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3. Communication between institutions and journals

Institutions should:

- acknowledge receipt of communications from journal editors and respond promptly to allegations of research misconduct;
- inform editors (or respond to enquiries from editors) about on-going investigations into misconduct likely to affect the validity of publications (e.g. to confirm that formal investigations are underway – following initial assessment of the allegation – and state the likely duration) so that editors can issue an Expression of Concern if necessary;
- share the findings of misconduct investigations with journals so that the editor(s) can determine whether retractions or corrections are required;
- ensure that all communications relating to misconduct investigations (such as press briefings and notifications to journals) are clear, accurate and complete.

Both institutions and editors should generally ensure that communications relating to ongoing misconduct investigations are kept confidential between parties; however editors may use an Expression of Concern to inform readers about serious allegations likely to affect the reliability or integrity of a publication.

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Journals should:

- acknowledge receipt of communications from institutions and respond promptly to findings of research misconduct;
- inform institutions about possible misconduct and provide evidence to support these concerns (e.g. analysis of text similarity in cases of suspected plagiarism, or evidence of inappropriate image manipulation);
- investigate allegations of misconduct by researchers acting as peer reviewers for the journal (e.g. that reviewers plagiarized another researcher's work), follow the COPE flowchart on such cases, and liaise with the institution as required;[6]
- follow the COPE guidelines on retractions.[4]

4. Responding to journal concerns about research integrity or publication practices

Institutions should initiate inquiries into allegations of research misconduct or unacceptable publication practice raised by journal editors.

Where possible, journals should provide evidence to support allegations of misconduct or questionable practices (e.g. copies of overlapping publications, evidence of plagiarism). However, editors may be obliged to protect the identity of whistleblowers or of peer reviewers.

Institutions should respond promptly and constructively to editors' requests for clarification of authorship or data ownership. Editors have to rely on the honesty of researchers in declaring their contributions to a project. Journals cannot be expected to adjudicate in authorship disputes and therefore rely on institutions to arbitrate in such matters. Editors should respond to authorship adjudications supplied by institutions and, where necessary, issue corrections (i.e. by adding or removing authors from the by-line of published or submitted articles). Editors should follow the relevant COPE flowcharts in such cases.[7]

5. Cases involving multiple institutions or journals

In collaborative research involving multiple institutions, one institution should be nominated to coordinate investigations and act as the point of contact unless there is an obvious lead institution (e.g. that administers the grant or employs the researchers). Disputes between institutions over authorship or data ownership may require adjudication by an independent arbitrator agreeable to all parties.

Cases of plagiarism, breach of copyright or redundant publication usually involve several journals who should therefore cooperate with each other and share information as required (e.g. about submission dates and copyright transfer agreements) to resolve the issues.

6. Ensuring the reliability of the published research record

If an institutional investigation or disciplinary hearing into research misconduct recommends that a researcher seeks a retraction or correction, the institution should inform the editor(s).

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Similarly, journals should be prepared to issue retractions or corrections when provided with findings of misconduct arising from appropriate investigations.[4]

Publications should be retracted if they prove unreliable (for whatever reason), but if only a small part of the publication is affected (while the majority of findings and conclusions are valid) then a correction should be published.

Expressions of Concern may be published to alert readers to an ongoing investigation into actions likely to affect the reliability of published findings; they should be followed by a retraction, exoneration or correction when the investigation has concluded. Expressions of Concern should not be viewed as 'milder' versions of retractions.

Journals should also be prepared to publish corrections or retractions when honest errors are admitted. Retraction statements should include the reasons for the retraction and should distinguish between cases of misconduct and honest error to encourage researchers to report errors when they occur and ensure no stigma is attached to this.[4]

7. Journal and institutional policies

Institutions should have policies supporting research integrity and good practice (e.g. for authorship), describing research misconduct (e.g. data fabrication and plagiarism) and unacceptable publication practices (e.g. redundant publication, inappropriate authorship, and use of confidential material by reviewers), and how these are handled.[8] Such policies should be publicised and enforced within the institution.

Institutions should encourage researchers to inform journals if errors are discovered in published work.

Journals should have policies about how they handle suspected misconduct and how they respond to institutions and other organizations that investigate cases of research misconduct (e.g. national bodies).

8. Encouraging good practice

Journals should provide clear advice to authors and reviewers and have appropriate policies for editors and staff relating to all aspects of publication ethics.[9,10] Journals should inform authors and readers how they handle cases of suspected research misconduct or unacceptable publication practices.

Institutions should include training in good publication practices as part of their programmes of education in research integrity.

Institutional leaders and journal editors should aim to create research environments that encourage good practice and should lead by example in their own publication practices.

Institutions should ensure that their systems for appointments and assessing research productivity do not create incentives for unacceptable practices, such as redundant publication and guest authorship.

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9. Investigating previous publications

Research and publication misconduct may not be an isolated incident. In many cases, when serious misconduct comes to light, investigation of the researcher's earlier work reveals further problems. Therefore, when a researcher is found to have committed serious misconduct (such as data fabrication, falsification or plagiarism) the institution should review all the individual's publications, including those published before the proven misconduct took place. In such cases, it may be necessary to alert previous employers to enable them to review work carried out by the discredited researcher when working at their institution, to determine the reliability of publications arising from that work (for an example of this see reference[11]).

References

1. Wager E. Coping with scientific misconduct. BMJ 2011;343:d6586
2. Wager E. What do journal editors do when they suspect research misconduct? Medicine & Law 2007;26:535-44
3. Sox HC & Rennie D. Research misconduct, retraction, and cleansing the medical literature: lessons from the Poehlman case. Annals of Internal Medicine 2006;144:609-13
4. Wager E, Barbour V, Yentis S, Kleinert S on behalf of COPE Council. Retractions: Guidance from the Committee on Publication Ethics (COPE). http://publicationethics.org/files/u661/Retractions_COPE_gline_final_3_Sept_09__2_.pdf
5. COPE code of conduct for journal editors. www.publicationethics.org/resources/code-conduct
6. COPE Flowchart: What to do if you suspect a reviewer has stolen an author's idea or data. http://publicationethics.org/files/u2/07_Reviewer_misconduct.pdf
7. COPE Flowcharts: Changes in authorship. http://publicationethics.org/resources/flowcharts
8. Singapore Statement on Research Integrity. www.singaporestatement.org
9. Wager E & Kleinert S (2011) Responsible research publication: international standards for authors. A position statement developed at the 2nd World Conference on Research Integrity, Singapore, July 22-24, 2010. Chapter 50 in: Mayer T & Steneck N (eds) Promoting Research Integrity in a Global Environment. Imperial College Press / World Scientific Publishing, pp 309-16. Also available at: http://publicationethics.org/international-standards-editors-and-authors
10. Kleinert S & Wager E (2011) Responsible research publication: international standards for editors. A position statement developed at the 2nd World Conference on Research Integrity, Singapore, July 22-24, 2010. Chapter 51 in: Mayer T & Steneck N (eds) Promoting Research Integrity in a Global Environment. Imperial College Press / World Scientific Publishing, pp 317-28. Also available at: http://publicationethics.org/international-standards-editors-and-authors
11. Reich ES. Biologist spared jail for grant fraud. Nature News, 28 June 2011. Nature 474, 552(2011); doi 10.1038/474552a

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Further reading

Office of Research Integrity. Handling misconduct. http://ori.dhhs.gov/misconduct/

UK Research Integrity Office. Procedure for the Investigation of Misconduct in Research. www.ukrio.org

Australian Code for the Responsible Conduct of Research. http://www.nhmrc.gov.au/publications/synopses/r39syn.htm

European Science Foundation(ESF)/ All European Academies (ALLEA). The European Code of Conduct for Research Integrity. http://www.esf.org/activities/mo-fora/research-integrity.html

Honesty, Accountability and Trust: Fostering Research Integrity in Canada. The Expert Panel on Research Integrity, 2010. http://www.scienceadvice.ca/uploads/eng/assessments and publications and news releases/research integrity/RI_report.pdf

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*Please note that contributors commented in their individual capacities and therefore this listing does not necessarily indicate that these institutions endorse these guidelines

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PCTC
PAVEMENT COATINGS TECHNOLOGY COUNCIL

November 20, 2015

Dr. David L. Sedlak
Editor, *Environmental Science & Technology*
Malozemoff Chair Professor in Mineral Engineering
University of California, Berkeley
Civil and Environmental Engineering
657 Davis Hall
Berkeley, California 94720

Via email: eic@est.acs.org

Dear Dr. Sedlak,

Re: Request for Retraction

I am the Executive Director of the Pavement Coatings Technology Council (PCTC), a trade association the members of which are involved in manufacturing pavement coatings. With regret, and with full awareness of the gravity of this request, I write to request that you retract:

> Mahler, B. J., P. Van Metre, T. J. Bashara, J. T. Wilson, and D. A. Johns, 2005, Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons: Environmental Science & Technology, v. 39, p. 5560 – 5566 (Mahler *et al.* 2005).[1]

As I show below, there is, in the words of the Committee on Publication Ethics (COPE)'s Retraction Guidelines, "clear evidence that the [paper's] findings are unreliable."

By way of background, Mahler *et al.* (2005) is the first of almost twenty articles published over a decade by Mahler and her colleagues concerning one of the products manufactured by PCTC members, refined coal tar-based pavement sealants (RTS). Over that same period, PCTC has commissioned many scientists, with expertise in a variety of areas, to evaluate these articles. PCTC has asked those evaluators to focus on the science. PCTC has

[1] Included as Attachment A.

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consistently been both surprised and dismayed by the number of times we have been told that the publication under evaluation (i) cannot be reproduced and (ii) is missing some key data, which hinders the evaluation. In hopes that the federal agency involved (the U.S. Geological Survey (USGS)) would "do the right thing" – and follow Federal law – we have pointed out irregularities and requested missing information via attempted scientist-to-scientist communications. As Mahler and colleagues have repeatedly declined to share that data with PCTC, we have been obliged to file Freedom of Information Act (FOIA) requests and Information Quality Act petitions, and even to ask for the assistance of the American Chemistry Society's (ACS) Committee on Ethics. These processes began more than 5 years ago, but have met with little success. Regrettably, it is time to follow another path.

Mahler *et al.* (2005) reports the results of a study of simulated run off from parking lots in Austin, Texas that were coated with a refined coal tar-based pavement sealant (RTS), an asphalt-based pavement sealant (ABS), or not believed to have been coated with a sealant of any variety. In the paper, the connection between polycyclic aromatic hydrocarbons (PAHs) was made by comparison of ratios of PAHs measured in runoff samples to ratios of PAHs in urban stream sediment. The source or identify of the data used to represent urban stream sediment was not provided in Mahler *et al.* (2005). The comparison was illustrated in Figure 4 of Mahler *et al.* (2005) and the method of identifying the source of PAHs was described as follows:

> PAHs comprise a large group of compounds, and PAH assemblage is often used to infer PAH sources (27). Differences in PAH assemblages can be investigated by computing the ratios of selected PAHs (28, 29). The best indicator ratios of coal tar as a PAH source have been identified as fluoranthene:pyrene, indeno[1,2,3-cd]pyrene:benzo[ghi]perylene, and benzo[a]pyrene:benzo[e]pyrene (30, 31). In graphs that combine these ratios, similarities and differences between parking lot and stream samples are apparent (Figure 4): **ratios in the urban stream sediment group match those in runoff from coal-tar-sealed lots more closely than they do those from asphalt-sealed lots and from unsealed lots (asphalt and cement).** We found these ratios were far more effective at distinguishing between the different parking lot samples and stream samples than ratios indicative of combustion versus noncombustion sources, or other approaches such as comparison of parent compound distribution (32). [Mahler *et al.* (2005) p. 5564, emphasis added]

The purported match between PAH ratios observed in RTS and ratios found in unidentified urban stream sediment samples was used, along with some poorly documented PAH load calculations, to make the following claim:

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> We show that a previously unidentified source of urban PAHs, parking lot sealcoat may dominate loading of PAHs to urban water bodies in the United States. [Mahler *et al.* (2005) Abstract, p. 5560]

In a subsequent publication by the same authors (Van Metre *et al.*, 2009), the method of identification of RTS as a source of PAHs used in Mahler *et al.* (2005) was described thus:

> Differences in PAH assemblages can be investigated by computing ratios of selected PAHs; two ratios that have been identified as indicators of coal tar as a PAH source are fluoranthene/ pyrene (F:P) and benzo[a]pyrene/benzo[e]pyrene (A:E) (5,21). These ratios were effective for distinguishing PAH from coal-tar-based sealcoat from other combustion PAH sources in Austin (9). [Van Metre *et al.* (2009); reference (9) is Mahler *et al.* (2005); p. 22]

The data from samples of runoff from RTS sealed parking lots in Austin, TX that had been shown in Mahler *et al.* (2005) to have PAH ratios that matched PAH ratios in the unidentified urban stream sediment samples were then used as a point of comparison with sediment from 9 locales across the United States to posit a regional difference:

> Most central and eastern dust and lake samples plot near each other and near mean values for runoff particles from coal-tar sealcoated pavement in Austin (9),and closer to a coal-tar standard-reference material (SRM) (22) than do Western dust and lake samples. [Van Metre *et al.* (2009) p. 22]

The regional difference was then imputed to different usage patterns of RTS in the eastern and central US compared to the western US.

Publication of Mahler *et al.* (2005) resulted in attempts to reproduce the results. The first attempt grew out of an evaluation of Mahler *et al.* (2005) commissioned by PCTC which resulted in a Comment submitted to and published in Environmental Science & Technology (ES&T) (DeMott & Gauthier, 2006). The Comment made two points:

- 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.

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A second effort at testing the hypothesis that RTS is the dominant source of PAHs in sediment was included in a paper authored by City of Austin, TX, scientists, Scoggins *et al.* (2007). The authors attempted to relate PAH concentrations in Austin stream sediments to RTS-coated parking lots using PAH ratio comparisons as well using spatial relationships. Their findings were 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. [Scoggins *et al.* (2007) p. 702].
>
> 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. [Scoggins *et al.* (2007) p. 705].

A third attempt was part of a PCTC-funded study of PAH concentrations in Austin, TX, area sediments before and after a ban of RTS that went into effect in the City of Austin on January 1, 2006, which resulted in a paper by DeMott *et al.* (2010). Neither the before (2005) nor the after (2008) samples contained PAH ratios similar to those identified as similar to RTS in Mahler *et al.* (2005).

The Mahler *et al.* (2005) hypothesis that RTS is a dominant source of PAHs in urban sediments was rooted in the comparison between PAH ratios in suspended solids in runoff from RTS-sealed parking lots in Austin, TX, and PAH ratios in samples identified as "urban stream sediment," illustrated in Figure 4 of the paper. To try to understand why DeMott & Gauthier (2006) were not able to reproduce the PAH double ratio diagram, and why neither Scoggins *et al.* (2007) nor DeMott *et al.* (2010) were able to identify an RTS signature in stream sediment samples from Austin streams, it is necessary to understand what samples were used as representative of "urban stream sediment" in Mahler *et al.* (2005). Neither that paper nor the Response to Comment (Mahler *et al.*, 2006a) made clear what those data were. As direct communications between PCTC and the USGS had become less than optimal, and the USGS response to PCTC's FOIA request did not shed any light on the identity of the samples, PCTC requested the assistance of ACS as the publisher of Mahler *et al.* (2005), copying USGS management on the correspondence. To its credit, the USGS responded immediately, providing the requested data.[2] PCTC's letter to the ACS and the USGS response are included here as Attachment B.

[2] To be clear, PCTC has an ongoing FOIA lawsuit against the USGS in which the central question concerns additional data related to USGS' RTS research activities that are being withheld. The fact that the USGS has disclosed some data via its FOIA response or by other means does not, in PCTC's view, relieve the USGS of the obligation to disclose all relevant data.

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USGS's response revealed that the data used in Figure 4 of Mahler *et al.* (2005) to represent urban stream sediment consisted of 8 samples of suspended sediment collected over several years from one location in Austin, TX (Williamson Creek) (Mahler *et al.* 2006b), and 12 suspended sediment samples collected from 3 locations in central Fort Worth, TX (Van Metre *et al.* 2003), about 200 miles away. None of these locations or samples are connected spatially, temporally, or hydrologically with the locations used in the Austin parking lot runoff study. The 8 samples from the single Austin location are from a different stream shed than any of the studied runoff locations. No information about potential sources of PAHs from parking lot runoff or other sources is provided for any of these samples.

With the identity of the samples used to represent urban stream sediments known, the question then becomes, why were these samples considered to be representative? The answer comes from Dr. Mahler herself, who wrote the following in an email to Leila Gosselink (an employee of the City of Austin, TX):

> 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 email was received as part of the USGS response to a PCTC FOIA request, and is included as Attachment C to this letter.

It is clear that the samples used to represent urban stream sediment in Mahler *et al.* (2005) were not chosen because they were believed to contain values representative of PAH signatures found in urban stream sediments in Austin or elsewhere. Instead, they were chosen because "they tended to group with [data from] the sealed parking lots." Selective use of data to confirm or generate a preferred hypothesis does not comport with the scientific method.

The evidence that Mahler *et al.* (2005) selectively used data to confirm or generate a preferred hypothesis meets COPE's criteria for "clear evidence that the findings are unreliable." For this reason, Mahler *et al.* (2005) must be retracted.

Thank you for your attention. Please feel free to contact me for additional information.

Very truly yours

Anne P. LeHuray, Ph.D.
Executive Director

Attachments

ED_006238_00000702-00104

cc: Alan D. Thornhill, Director USGS Office of Science Quality & Integrity (athornhill@usgs.gov)
William G. Wilber, USGS NAWQA Program Chief (wgwilber@usgs.gov)

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ATTACHMENT A

Mahler, B. J., P. Van Metre, T. J. Bashara, J. T. Wilson, and D. A. Johns, 2005, Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons: Environmental Science & Technology, v. 39, p. 5560 - 5566.

Environmental Science & Technology

Article

Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons

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Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons

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Polycyclic aromatic hydrocarbons (PAHs) are a ubiquitous contaminant in urban environments. Although numerous sources of PAHs to urban runoff have been identified, their relative importance remains uncertain. We show that a previously unidentified source of urban PAHs, parking lot sealcoat, may dominate loading of PAHs to urban water bodies in the United States. Particles in runoff from parking lots with coal-tar emulsion sealcoat had mean concentrations of PAHs of 3500 mg/kg, 65 times higher than the mean concentration from unsealed asphalt and cement lots. Diagnostic ratios of individual PAHs indicating sources are similar for particles from coal-tar emulsion sealed lots and suspended sediment from four urban streams. Contaminant yields projected to the watershed scale for the four associated watersheds indicate that runoff from sealed parking lots could account for the majority of stream PAH loads.

Introduction

Concentrations of polycyclic aromatic hydrocarbons (PAHs)—a group of widely recognized aquatic contaminants (*1*) comprising numerous carcinogens (*2*)—have been increasing in recent decades in many urban lakes, particularly in areas undergoing rapid urban growth (*3*). PAHs adversely affect mammals (including humans), birds, fish, amphibians, invertebrates, and plants; in the aquatic environment, the effects of PAHs on invertebrates include inhibited reproduction, delayed emergence, sediment avoidance, and mortality, and the effects on fish include fin erosion, liver abnormalities, cataracts, and immune system impairments (*4*). Numerous sources of PAHs to urban runoff have been identified, including automobile exhaust, lubricating oils, gasoline, tire particles, erosion of street material, and atmospheric deposition (*5*–*8*), but uncertainty remains as to their relative importance. Investigations of urban sources of PAHs have thus far overlooked a potentially major source: parking lot sealants, also called "sealcoat". Our objective in this study was to evaluate the contribution of PAHs from sealed parking lots to urban streams.

In the United States and Canada, sealcoat is applied to many parking lots and driveways in an effort to protect the underlying asphalt pavement and enhance appearance. The two primary sealcoat materials on the market are refined coal-tar-pitch-based emulsion and asphalt-based emulsion. Although similar in appearance (glossy black), coal tar and asphalt have different molecular structures stemming from their origins: coal tar is a byproduct of the production of coke from coal, whereas asphalt is derived from the refining of crude petroleum. Coal tar, a known human carcinogen, is 50% or more PAHs by weight (*2*); the predominant constituents of asphalt are bitumens, complex mixtures of hydrocarbons that include asphaltenes, saturates, aromatics, and resins (*9*). Coal-tar-emulsion- and asphalt-emulsion-based sealcoats typically contain 20–35% of the emulsion.

Parking lot sealants are used extensively in the United States and Canada. Although national use figures are not available, the *Blue Book of Building and Construction* (*10*), a directory for the construction industry, lists over 3300 pavement sealant companies in 28 U.S. states. One company advertises the application of 1.7 billion liters to date worldwide (*11*), and another reports having sealed over 33 million square meters (*12*). The City of Austin, population 650000 (2000 census), estimates that about 2.5 million liters of sealcoat is used annually in this city (*13*).

Sealcoat abrades from the parking lot surface relatively rapidly, and reapplication is recommended every two to three years (*14*). In 2003, the City of Austin identified abraded parking lot sealcoat as a possible source of high concentrations of PAHs in streambed sediment (*15*). Here we present evidence suggesting that parking lot sealcoat could indeed be the dominant source of PAHs to watersheds with residential and commercial development.

Experimental Section

Sample Collection. We compared concentrations and yields of particulate PAHs in simulated runoff from parking lots sealed with coal-tar-based sealcoat, from lots sealed with asphalt-based sealcoat, and from unsealed asphalt and cement lots. Thirteen urban parking lots, representing a range of sealant types that are currently in use in Austin, TX, were sampled (Table 1). In addition, four test plots, each about 120 m^2, were sampled. Three of the test plots were sealed just prior to testing, and one was left unsealed (asphalt surface). The test plots are at the Robert Mueller Municipal Airport, Austin, TX, which has been closed since 1999. A full description of the sampling is given in ref *16*. In brief, 50 m^2 areas of each parking lot and the test plots were sprinkled with 2 mm of distilled water (100 L over a 50 m^2 area) to simulate a light rain, and concentrations of PAHs were analyzed in particles filtered from the runoff. The study focused on the particulate fraction, as PAHs in urban runoff, particularly those of higher molecular weight, are mostly associated with particulates (*7*, *17*); for selected samples (test plots and seven parking lots), the dissolved phase also was analyzed. The testing followed a minimum of 5 days with no rainfall. The parking lots were sampled once, and the test plots were sampled three times over a 6 week period. Water was sprayed from a plastic hand-held sprayer at a rate of about 7 L/min from a height of about 0.75 to 1 m. Spill berms were used at the down-slope end of the delineated area to gather water, which was then pumped into high-density polyethylene (HDPE) containers (Figure S1, Supporting Information). Recovery of water and observations about losses of water to wetting and leakage under the berms were noted. The water was returned to the laboratory, poured into a 50 L churn to keep the sample well mixed, and filtered through 0.45 μm pore size PTFE filters. The filters were

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TABLE 1. Sampling Site Characteristics

site name	surface type	date of sealant application	sampling date	study component
$CT_{TP}1$	coal-tar emulsion sealant	Aug 5–6, 2003	8/21/2003	test plot
$CT_{TP}2$	coal-tar emulsion sealant	Aug 5–6, 2003	8/21/2003	test plot
AS_{TP}	asphalt emulsion sealant	Aug 5–6, 2003	8/21/2003	test plot
$UNSAS_{TP}$	unsealed asphalt pavement	Aug 5–6, 2003	8/21/2003	test plot
$CT_{TP}1$	coal-tar emulsion sealant		9/9/2003	test plot
$CT_{TP}2$	coal-tar emulsion sealant		9/9/2003	test plot
AS_{TP}	asphalt emulsion sealant		9/9/2003	test plot
$UNSAS_{TP}$	unsealed asphalt pavement		9/9/2003	test plot
$CT_{TP}1$	coal-tar emulsion sealant		9/26/2003	test plot
$CT_{TP}2$	coal-tar emulsion sealant		9/26/2003	test plot
AS_{TP}	asphalt emulsion sealant		9/26/2003	test plot
$UNSAS_{TP}$	unsealed asphalt pavement		9/26/2003	test plot
$AS_{PL}1$	asphalt emulsion sealant	June 2003	9/7/2003	parking lot
$AS_{PL}2$	asphalt emulsion sealant	June 2003	9/7/2003	parking lot
$AS_{PL}3$	asphalt emulsion sealant	July 2003	9/28/2003	parking lot
$CT_{PL}1$	coal-tar emulsion sealant	March 2003	9/7/2003	parking lot
$CT_{PL}2$	coal-tar emulsion sealant	July 2003	9/28/2003	parking lot
$CT_{PL}3$	coal-tar emulsion sealant	July 2003	9/28/2003	parking lot
$CT_{PL}4$	coal-tar emulsion sealant	July 2003	9/30/2003	parking lot
$CT_{PL}5$	coal-tar emulsion sealant	July 1999	9/30/2003	parking lot
$CT_{PL}6$	coal-tar emulsion sealant	Nov 2000	9/30/2003	parking lot
$UNSAS_{PL}1$	unsealed asphalt pavement		9/8/2003	parking lot
$UNSAS_{PL}2$	unsealed asphalt pavement		9/30/2003	parking lot
$UNSCON_{PL}1$	unsealed concrete pavement		9/8/2003	parking lot
$UNSCON_{PL}2$	unsealed concrete pavement		9/8/2003	parking lot

massaged inside locking bags to remove retained particles, as described in ref *18*, and the recovered particulates were submitted as chilled slurries in clean glass vials to the U.S. Geological Survey National Water Quality Laboratory (NWQL) for analysis. In some cases the filtrate also was shipped, in chilled and clean amber glass bottles, to the NWQL for analysis of dissolved PAH. One or more samples of unfiltered water were collected from the churn for measurement of suspended sediment concentration (SSC), used to determine the mass of sediment recovered during each test. Although the 2 mm of simulated rain was not enough to wash off all of the mobile sediment, the recovered water was visibly clearer toward the end of each application. In samples from the five sites in which SSC was measured in the first 50 L and final 50 L of water, SSC decreased by a mean of 65% (range of 39–84%). We therefore assumed that the tests recovered most of the sediment that would be mobilized from the parking lot surfaces by a rain event, regardless of magnitude. Large, intense storms, however, likely would generate a higher yield of sediment.

The test plot and parking lot scrapings were obtained by scraping a small area (less than 0.25 m^2) with a metal paint scraper. The particulates removed were brushed onto a piece of new cardstock and then into a cleaned glass jar. The paint scraper was cleaned between sites, and a new brush was used at each site. Scrapings were examined by light and electron microscopy (Figure S2, Supporting Information), and submitted to the NWQL for PAH analysis.

Computation of Yields. Losses of water to wetting and losses of water and sediment leaking under the berms were estimated. Recovery of water ranged from 19 to 85 L with a median of 58 L. The lowest recoveries were from flat, unsealed asphalt lots, and the highest recoveries were from sealed lots and cement lots with gentle slopes. On the basis of recoveries and field observations, it was concluded that about 18 L of water was retained on the surface of sealed lots and cement lots and that the remainder of the water loss was a result of leakage past the berms. It was assumed that no yield of particles was associated with the water volume lost to surface wetting and that water leaking past the berms had the same SSC and contaminant levels as recovered water. For unsealed asphalt lots, the loss to wetting was estimated as 36 L for a maximum potential recovery of 64 L. Thus, to estimate the total yield of sediment from each lot, SSC was multiplied by the assumed maximum recovery (82 L for sealed and cement lots and 64 L for unsealed asphalt lots) to account for recovered water and leakage past the berms. Yields of PAH were estimated by multiplying the total yield of sediment times particle concentrations.

Chemical Analysis. Samples were prepared by extracting about 0.5 g dry weight of sample using pressurized solvent extraction at 120 and 200 °C with a mixture of water and isopropyl alcohol. The samples were extracted at each temperature at a pressure of 13800 kPa. Surrogate compounds were added to the sample prior to extraction to verify method recoveries. The extract was cleaned up using polystyrene divinylbenzene and Florisil solid-phase extraction cartridges. The extract was concentrated, solvent exchanged to ethyl acetate, and diluted to 10 mL. An internal standard mixture was added to an aliquot of the extract, and the extract was analyzed by full-scan gas chromatography/mass spectrometry (GC/MS). Difficult sample matrices were diluted before the full-scan analysis, and diluted surrogates were estimated in the samples.

Compound identifications were based on comparison of peak retention times and mass spectra to those of authentic standard compounds for the target compounds. Response factors were calculated for each compound from a set of calibration standards. Quantitation was carried out following the methods of Olson et al. (*19*). For PAHs in the particulate phase, the estimated method reporting limit (MRL) is 5 μg/kg for a 25 g sample. As less than 25 g was extracted, the MRL was raised accordingly, on a sample-by-sample basis. In some cases, MRLs were raised because of background interferences.

Dissolved-phase samples were analyzed following the method described by Fishman and Friedman (*20*), with the

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difference that continuous liquid–liquid extraction was substituted for use of the separatory funnel. In brief, 1 L samples fortified with surrogate compounds were extracted by continuous liquid–liquid extraction for 6 h under acidic and then basic conditions. Internal standards were added and sample extracts concentrated to 1 mL. Samples were analyzed by GC/MS in electron impact mode. Sample identifications were made by matching retention times and mass spectra with those of standard compounds. Quantitation involved use of internal standards and calibration curves generated by standard compounds of known amounts.

Quality control (QC) consisted of environmental and internal laboratory samples. Two duplicate environmental samples for particulate analysis of PAH were collected. For one of the sets of duplicates, ΣPAH differed by 8%; for the second (which had ΣPAH > 4000 mg/kg), ΣPAH differed by 54%. In the equipment blank analyzed for dissolved PAH, three parent PAHs were detected at concentrations about half that of the environmental sample with the lowest concentrations, and less than 1% that of the environmental sample with the highest concentrations.

Laboratory QC samples for particulate PAH analyses consisted of analysis of spiked samples, blanks, and samples of certified reference material (CRM). The median recovery for the six spiked samples was 76%. For the six laboratory blanks, an analyte was detected in 85 of 336 possible cases. The detected concentrations ranged from 0.1% to 3.5% of that in the environmental sample with the lowest concentration for that analyte. For the two analyses of CRM, the recoveries were within the NWQL-established acceptable range for 83% of the cases.

Three commercially available asphalt-based emulsion sealcoat products and six coal-tar-based emulsion sealcoat products were analyzed at DHL Analytical, Round Rock, TX, using EPA method SW 8270 (*21*). In each case, the product sample was taken directly from the container. Concentrations of 16 parent PAHs were determined. The sealants analyzed were not necessarily the same as those applied to the test plots or on the parking lots in use, although there was some overlap (product AS_PA was used on test plot AS_{TP}; product CT_PF was used on test plot $CT_{TP}2$) (Table 2).

Results

Concentrations and yields of total particulate PAH and total dissolved PAH in the runoff and total PAH in the scrapings were computed and compared between parking lot surface types (Table 2). The total particulate PAH (ΣPAH) concentration was computed for each sample as the sum of naphthalene, 2-methylnaphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[*a*]anthracene, chrysene, benzo[*a*]pyrene, and dibenz[*ah*]anthracene, which are the same as those used for the consensus-based sediment quality guidelines of MacDonald et al. (*22*). For unsealed parking lots (asphalt pavement and concrete combined), the mean ΣPAH was 54 mg/kg (range of 7.2–75 mg/kg), more than twice the probable effect concentration sediment quality guideline of 22.8 mg/kg (*22*) (Table 2), and in the range of those found by others in urban and roadway runoff (e.g. refs *23*–*25*). However, the mean ΣPAH concentration from the asphalt-sealed parking lots was more than 10 times higher (mean of 620 mg/kg, range of 250–830 mg/kg) than that from unsealed parking lots, and the mean ΣPAH concentration from the coal-tar-sealed parking lots was 65 times higher (mean of 3500 mg/kg, range of 520–9000 mg/kg) (Table 2; complete concentration data are given in ref *16*). ΣPAH concentrations in runoff from coal-tar-sealed lots were significantly higher than in runoff from other surface types (Kruskal–Wallis test of comparisons, hypothesis of no difference between groups rejected for $p < 0.05$). PAH concentrations from coal-tar-sealed lots also were much higher, in most cases by orders of magnitude, than PAH concentrations in other urban sources such as tire particles, motor oil, and weathered asphalt (Figure 1; note that this figure uses a different summation of PAH). ΣPAH concentrations in runoff from the sealed test plots were generally lower than those from the sealed parking lots, but the difference was not statistically significant, and concentrations from unsealed surfaces, with the exception of one outlier, were similar for test plots and parking lots.

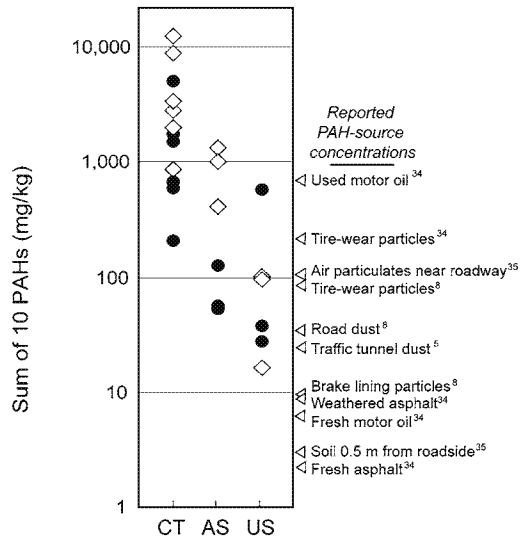

FIGURE 1. Sum of 10 PAHs (fluoranthene, pyrene, benz[*a*]anthracene, benzo[*a*]pyrene, benzo[*e*]pyrene, indeno[1,2,3-*cd*]pyrene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, and benzo[*ghi*]perylene) in particles in runoff from simulated rainfall on coal-tar emulsion sealcoat (CT), asphalt emulsion sealcoat (AS), and unsealed cement and asphalt (US) parking lots (◇) and test plots (●). Parking lots were sampled once, and test plots were sampled three times. Concentrations for other PAH sources reported in the literature also are indicated. These 10 PAHs were summed for this graph to facilitate comparison between experimental and reported concentrations.

Concentrations of ΣPAH in the scrapings ranged from 9500 to 83000 mg/kg for coal-tar-emulsion-sealed surfaces (including test plots) and from 110 to 2000 mg/kg for asphalt-emulsion-sealed surfaces (Table 2). Scrapings of two unsealed asphalt parking lots had ΣPAH concentrations of 7.1 and 20 mg/kg. Scrapings were observed under light and electron microscropy (Figure S2, Supporting Information).

Concentrations of total dissolved PAH (ΣPAH_{diss}, computed as the sum of the same PAHs as in ΣPAH excluding 2-methylnaphthalene; nondetections treated as zeros) for the test plots were about an order of magnitude greater in samples from the coal-tar-sealed test plots than concentrations in samples from the asphalt-sealed test plot, which in turn were about an order of magnitude greater than those from the unsealed test plot (Table 2). Nine of the 16 PAHs analyzed for were detected (complete data are in ref *16*). Higher weight PAHs—benzo[*b*]fluoranthene, benzo[*k*]-fluoranthene, benzo[*a*]pyrene, indenopyrene, benzo[*ghi*]-perylene, benz[*a*]anthracene, and dibenz[*ah*]anthracene—were not detected at laboratory reporting levels ranging from 1.7 to 3.4 μg/L. Four PAHs (acenaphthylene, acenaphthene, chrysene, and fluorene) were detected only in runoff from the coal-tar-sealed test plots; anthracene was detected in runoff from all the sealed test plots but not from the unsealed site. A similar suite of PAHs were detected at those parking lots for which the filtrate was analyzed (Table 2; complete data are in ref *16*).

Concentrations of ΣPAH in the commercially available sealant products and surface scrapings exceeded those of

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TABLE 2. Concentrations of PAH in Washoff Samples, Scrapings, and Unapplied Sealcoat Product[a]

	washoff samples		scrapings	product	
	ΣPAH, mg/kg	ΣPAH_{diss}, μg/L	ΣPAH, mg/kg		ΣPAH(dry), mg/kg
Test Plots					
$CT_{TP}1$, 8/12/03		21	83000	CT_PA	34000
$CT_{TP}1$, 8/21/03	1700	14		CT_PB	113000
$CT_{TP}1$, 9/9/03	530			CT_PC	202000
$CT_{TP}1$, 9/26/03	460	6.9		CT_PD	86000
				CT_PE	49000
$CT_{TP}2$, 8/12/03		11	11000	CT_PF	61000
$CT_{TP}2$, 8/21/03	1200	7.3			
$CT_{TP}2$, 9/9/03	4000			AS_PA	6600
$CT_{TP}2$, 9/26/03	140	3.8		AS_PB	1300
				AS_PC	300
AS_{TP}, 8/12/03		1.3	110		
AS_{TP}, 8/21/03	96	1.2			
AS_{TP}, 9/9/03	40				
AS_{TP}, 9/26/03	28	0.64			
$UNSAS_{TP}$, 8/12/03		0.16			
$UNSAS_{TP}$, 8/21/03	410	0.34			
$UNSAS_{TP}$, 9/9/03	25				
$UNSAS_{TP}$, 9/26/03	14	0.17			
Parking Lots					
$CT_{PL}1$	2000	NA	25000		
$CT_{PL}2$	9000	5.4	15000		
$CT_{PL}3$	2000	7.1	11000		
$CT_{PL}4$	1300	12	9500		
$CT_{PL}5$	520	2.3	9900		
$CT_{PL}6$	5900	16	17000		
$AS_{PL}1$	250	NA	340		
$AS_{PL}2$	830	NA	2000		
$AS_{PL}3$	770	5.1	420		
$UNSCON_{PL}1$	75	NA	NA		
$UNSCON_{PL}2$	69	NA	NA		
$UNSAS_{PL}1$	64	NA	7.1		
$UNSAS_{PL}2$	7.2	0.24	20		

[a]Sums are as defined in the text. NA = not analyzed, CT = coal-tar-based emulsion, AS = asphalt-based emulsion, UNSAS = unsealed asphalt pavement, and UNSCON = unsealed concrete pavement.

the particulates in the washoff. Concentrations of ΣPAH in commercially available coal-tar-based sealcoat products ranged from 3.4 to 20 wt %, compared to 0.03 to 0.66 wt % for asphalt-based sealcoat products analyzed (Table 2; complete data in Table S1, Supporting Information).

Yields of ΣPAH (mass of ΣPAH per unit area of parking lot) computed for the simulated rainfall followed patterns similar to those of concentrations. Complete data for yields can be found in Table S2 in the Supporting Information. As with the concentrations, there was a wide range in the yields for a given surface type, in most cases more than an order of magnitude. The mean yield from coal-tar-sealed lots exceeded that from asphalt-sealed lots by more than a factor of 2, although this difference was not statistically significant (Kruskal–Wallis test of comparison, $p < 0.05$). However, the mean yield from sealed lots (asphalt and coal tar combined) exceeded that from unsealed lots by a factor of 50, and the difference was statistically significant.

Discussion

Runoff from parking lots typically is contaminated with PAHs from leaking motor oil, tire particles, vehicle exhaust, and atmospheric fallout, and it is not surprising that the mean concentration of ΣPAH in particles washed off each of the different surface types exceeded the probable effect sediment quality guideline. However, the large differences between

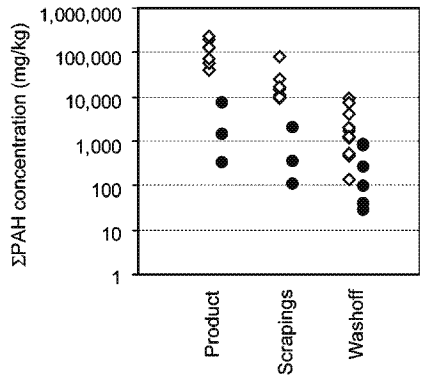

FIGURE 2. Comparison of ΣPAH concentrations in commercially available sealcoat products, scrapings from parking lots, and particles in washoff from parking lots for coal tar (◇) and asphalt (●) based sealants.

concentrations for the different surface types suggest that abraded sealant products are a potentially important (and heretofore unrecognized) contributor to PAH contamination in urban and suburban water bodies.

Comparison of Medium, Aging, and Vehicle Use on Concentrations and Yields. For both coal-tar- and asphalt-emulsion-based sealants, the ΣPAH concentration decreased from the unapplied sealant products to the scrapings to the washoff samples, as did the difference in concentration between the coal-tar-based and asphalt-based sealant samples (Figure 2). The difference in the median ΣPAH concentration between the coal-tar-based and asphalt-based sealants was 70-fold for the products analyzed and decreased to 40-fold for the scrapings and to a factor of about 8 for the washoff samples. Although the chemical changes between the product pre- and postapplication were not the focus of this study, the decrease in ΣPAH concentrations from the scrapings to the washoff particulates and the magnitude of the difference between the coal-tar-sealed lots and the asphalt-sealed lots can be attributed to dilution of abraded particles with less contaminated street dust and the greater abrasion of the asphalt-sealed compared to the coal-tar-sealed surfaces. A simple mass balance, assuming dilution of the coal tar scrapings (median ΣPAH concentration of 13000 mg/kg) by street dust (median ΣPAH concentration of 50 mg/kg) at a proportion of 1 part abraded particles to 7 parts street dust, results in the concentration found in the washoff. If the proportion of abraded particles is increased for the asphalt lots on the basis of the increased yields measured for asphalt-sealed lots (assuming that the greater median particle yield of 320 mg/m^2 from asphalt-sealed lots versus 200 mg/m^2 from coal-tar-sealed lots results from increased abrasion), the concentration found in the washoff from asphalt-sealed lots is well approximated.

The effect of aging of sealants on concentration over the short term (7 weeks) was evident at the test plots (Figure 3a). Overall, the concentration of ΣPAH and ΣPAH_{diss} in the washoff from each test plot decreased over the 7 week period following application. In one instance ($CT_{TP}2$, second sampling of washoff) ΣPAH exceeded that previously sampled, but in all cases the concentration at the end of the period was less than that at the beginning. The PAH assemblage changed over the same period as well, as represented by a comparison of higher molecular weight (MW) to lower MW PAHs. In the particulate samples the ratio of higher MW PAHs (represented by benzo[*a*]pyrene + chrysene) to the lower MW PAHs (represented by fluorene + phenanthrene; these two PAHs were chosen as they were detected in most of the samples) increased at all of the sealed test sites. As the lower MW PAHs are more volatile and soluble than the higher MW PAHs, volatilization and leaching of the lower MW PAHs

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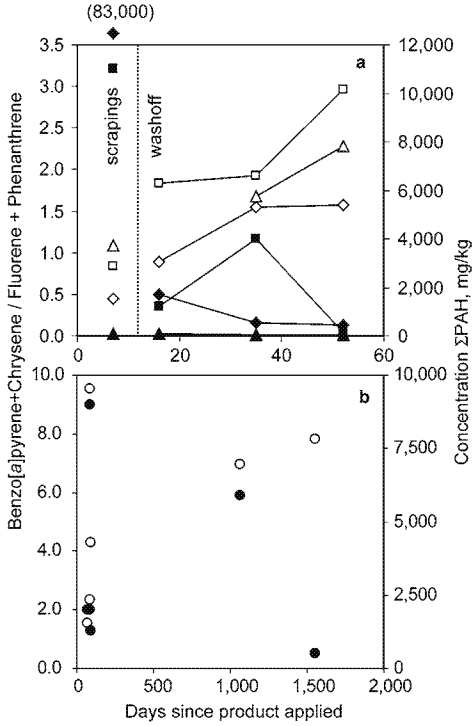

FIGURE 3. ΣPAH concentrations (closed symbols) and PAH ratios of higher and lower molecular weight PAHs (open symbols) (a) in scrapings and particles washed off coal-tar-sealed test plots (□, ◇) and an asphalt-sealed plot (△) and (b) as they relate to the age of coal-tar sealant in samples from parking lots in use.

from the newly applied sealant might be responsible for some of the decrease in concentration. For the parking lots, only coal-tar-sealed lots represented a range of ages, and for these lots there was no relationship between concentration or higher MW to lower MW PAH ratio and age of the sealant (Figure 3b). This might be because the parking lots were each sampled only once; the very wide range in PAH content between products, even those of a similar kind, may mask the effect of aging when time-series data are not available. Although the data are limited, they suggest that lots with older sealant tend to have a higher ratio of higher MW to lower MW PAHs, and that that ratio may reach a plateau after a period of time.

Comparison of the yields from the parking lots to those from the test plots, which receive no vehicle traffic, demonstrates the importance of abrasion of sealcoat by vehicles on ΣPAH yield: the mean ΣPAH yield was 20 and 160 times greater for the coal-tar-sealed and asphalt-sealed parking lots, respectively, than for the analogous test plots. This does not appear to be attributable to use patterns, although traffic counts were not made: the coal-tar-sealed lots are a mix of lots in constant use throughout the day (e.g., shopping center) and those with all-day parking (e.g., office), which are assumed to receive less use than those in constant use; all of the asphalt-sealed lots are all-day parking.

Environmental Implications. Given the extremely elevated concentrations of PAHs in particles washed from sealed parking lots, how important is this contribution to the total mass of PAHs in urban streams? To answer this question, we compared the PAH assemblages and estimated PAH loads associated with particulates in parking lot runoff to those associated with suspended sediment collected during storm flow in four streams: Williamson Creek (Austin, TX) (*18*) and influent streams to Echo Lake, Fosdic Lake, and Lake Como (Fort Worth, TX) (*26*). These four streams are in highly urbanized watersheds (land use for the Austin watershed is about 65% urban, and for the three Fort Worth

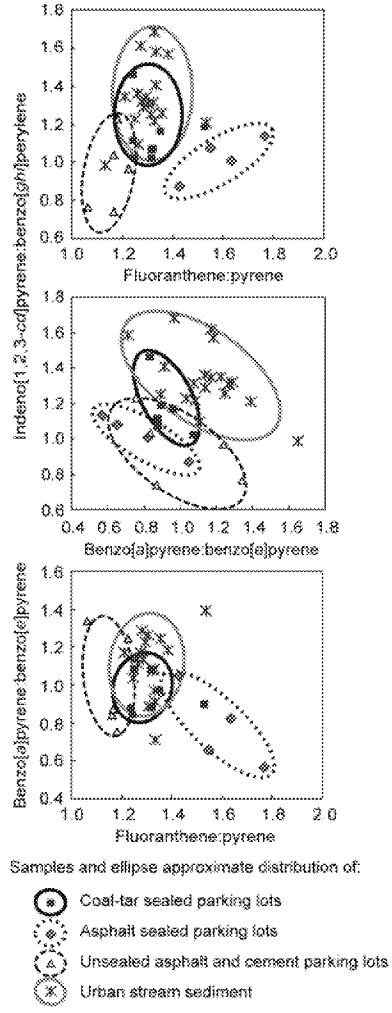

FIGURE 4. Comparison of indicator ratios of PAHs in particles washed from parking lots with coal-tar emulsion sealcoat, asphalt emulsion sealcoat, and unsealed asphalt pavement and concrete pavement, and in suspended sediment collected from four urban streams after storms.

watersheds is more than 90% urban; full land use is given in Table S3, Supporting Information); the streams are ephemeral, and urban runoff is assumed to comprise a large component of storm flow.

PAHs comprise a large group of compounds, and PAH assemblage is often used to infer PAH sources (*27*). Differences in PAH assemblages can be investigated by computing the ratios of selected PAHs (*28, 29*). The best indicator ratios of coal tar as a PAH source have been identified as fluoranthene:pyrene, indeno[1,2,3-*cd*]pyrene:benzo[*ghi*]perylene, and benzo[*a*]pyrene:benzo[*e*]pyrene (*30, 31*). In graphs that combine these ratios, similarities and differences between parking lot and stream samples are apparent (Figure 4): ratios in the urban stream sediment group match those in runoff from coal-tar-sealed lots more closely than they do those from asphalt-sealed lots and from unsealed lots (asphalt and cement). We found these ratios were far more effective at distinguishing between the different parking lot samples and stream samples than ratios indicative of combustion versus noncombustion sources, or other approaches such as comparison of parent compound distribution (*32*). Although alkylated PAH homologues were analyzed (including C1–C5 homologues of the MW 128, 178, 202, 228, and 252 PAHs), their interpretation did not assist in discriminating between PAHs from the different parking lot surfaces.

The relative amount of similarity between groups of samples, as defined by the ratios, was quantified through

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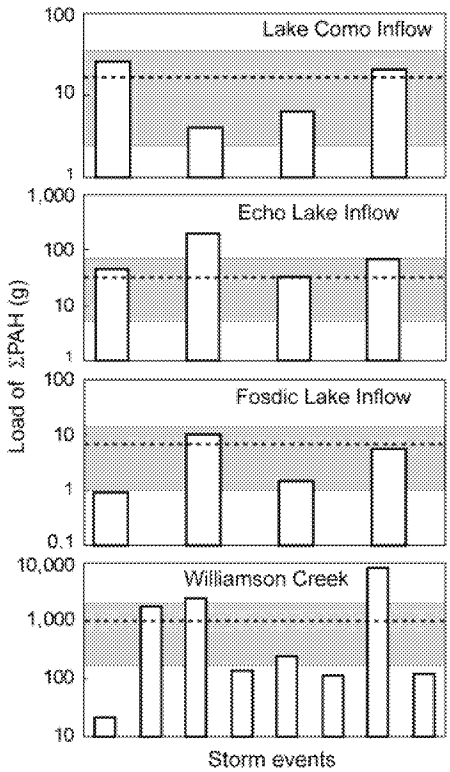

FIGURE 5. Comparison of estimated event loads of ΣPAH from sealed parking lots and measured instream storm-event loads for four urban watersheds. The interquartile range of estimated loads is shown in gray shading, on the basis of 25th and 75th percentile yields computed for sealed parking lots; the mean estimated load is indicated by a dashed line. Measured instream loads for four to eight individual events are shown as bars.

discriminant function analysis. In discriminant function analysis, each significant independent variable adds to discrimination between multiple groups. The three ratios (fluoranthene:pyrene, indeno[1,2,3-*cd*]pyrene:benzo[*ghi*]-perylene, and benzo[*a*]pyrene:benzo[*e*]pyrene) were entered into the analysis as the independent variables, with the different types of samples (coal-tar-emulsion-sealed lots, asphalt-emulsion-sealed lots, unsealed lots, and urban stormflow stream sediments) defining four groups of dependent variables. All three variables were shown to contribute significantly to discrimination between the groups ($p < 0.001$). The distances between the centroids of the groups were determined by computing the squared Mahalanobis distance, which is a measure of the distance between two points in the space defined by two or more correlated variables. The centroid of the group defined by the suspended sediment from urban streams is closest to the centroid of the coal-tar-based sealant group, next closest to that of the unsealed lot group, and farthest from that of the asphalt-based sealant group (squared Mahalanobis distances of 5.7, 13.0, and 25.0, respectively). Thus, on the basis of the three ratios diagnostic of coal-tar sources, the PAH assemblage of the suspended sediment from the urban streams most closely resembles that of the coal-tar-based sealant group, supporting the hypothesis that coal-tar-based sealants are an important source of PAHs in urban streams.

Moving to a mass-balance approach at the watershed scale for each of the four urban watersheds, we compared measured storm-event stream loads of ΣPAH to those estimated to be contributed by sealed parking lots. Digital land-use maps that included parking lots were provided by the Cities of Austin and Fort Worth and were updated using recent aerial photography and site inspections. Sealed and

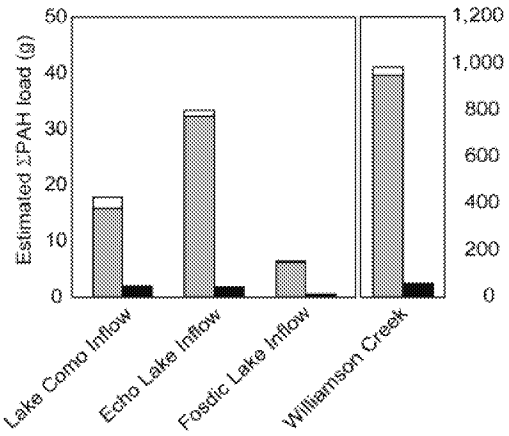

FIGURE 6. Comparison of event loads of ΣPAH for four urban watersheds estimated for parking lots in their current (2004) state (sealed by gray bars and unsealed by white bars) and projected loads if all existing parking lots were unsealed (black bars). Loads were estimated on the basis of the yields from the runoff experiments and the area of parking lots in each watershed.

unsealed lots were identified by site inspection. We computed the hypothetical storm-event load generated by sealed parking lots in each watershed by multiplying the mean yield for sealed parking lots (coal-tar and asphalt emulsion sealcoat combined) determined from the runoff experiments by the sealed parking lot area of each watershed. We assumed that the 2 mm of water applied for the field tests mobilized all available particles, and that all runoff from parking lots entered storm sewers and was delivered to the stream. Although there is substantial variation in event loads for each stream (*18, 26*), for all four watersheds the estimated ΣPAH loads contributed by sealed parking lots are similar in magnitude to measured stream loads, even though sealed parking lots cover only 1−2% of each watershed (Figure 5). These results might explain why an investigation carried out in Marquette, MI, found that runoff from commercial parking lots contributed 64% of the PAH load to the urban watershed studied (*33*).

What would be the effect on PAH loading to these watersheds if parking lots were not sealed? For each of the four watersheds, we compared the ΣPAH load contributed by parking lots (computed on the basis of the aerial extent of unsealed and sealed parking lots) to that obtained by applying the average yield for unsealed lots to all parking lots (Figure 6). We estimate that the ΣPAH load from parking lots in these watersheds would be reduced to 5−11% of the current loading if all lots were unsealed.

With the exception of the sealcoat itself, unsealed parking lots receive PAHs from the same urban sources as do sealed parking lots—e.g., tire particles, leaking motor oil, vehicle exhaust, atmospheric fallout—yet the average yield of PAHs from sealed parking lots is 50 times greater than that from unsealed lots. PAH assemblages and estimated loads further suggest that sealed parking lots could be dominating PAH loading in watersheds with commercial and residential land use. The implications of these results extend beyond Texas to the rest of the United States and Canada, where parking lot sealcoat is used extensively, and to other countries where sealcoat is being introduced. Previously identified urban sources of PAHs, such as automobile exhaust and atmospheric deposition, have been difficult to control or even quantify because of their nonpoint nature. In contrast, sealed parking lots are point sources, and use of the sealant is voluntary and controllable.

Acknowledgments

We thank Robert Eganhouse, E. Terrence Slonecker, and three anonymous reviewers for critical reading of the manuscript.

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This research was carried out as a cooperative project between the U.S. Geological Survey and the City of Austin.

Supporting Information Available

Two figures and three tables. This material is available free of charge via the Internet at http://pubs.acs.org.

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ATTACHMENT B

Letter to Dr. Gregory M. Ferrence from Anne P. LeHuray, dated July 13, 2013

Letter to Anne P. LeHuray from William G. Wilber, dated July 22, 2013.

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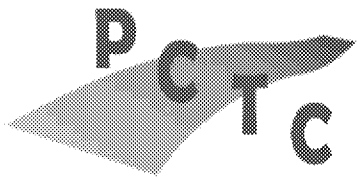

July 8, 2013

Dr. Gregory M. Ferrence
Chairman, American Chemistry Society (ACS) Committee on Ethics
Illinois State University
Via Email: gferren@ilstu.edu

Subject: Request for Assistance in Obtaining Data Collected and Used to Generate Conclusions Published in ACS (and Other) Journals

Dear Dr. Ferrence,

As a scientist and Chair of ACS' Committee on Ethics you well understand the importance of making available data on which conclusions about research results are based. The reasons are clearly explained in the description of the ethical obligation of authors seeking to publish in ACS journals. Every publishing scientist's first ethical obligation is described by ACS as follows:

> An author's central obligation is to present an accurate and complete account of the research performed, absolutely avoiding deception, including the data collected or used, as well as an objective discussion of the significance of the research. Data are defined as information collected or used in generating research conclusions. The research report and the data collected should contain sufficient detail and reference to public sources of information to permit a trained professional to reproduce the experimental observations.

As an ACS member and Executive Director of the Pavement Coatings Technology Council (PCTC), I am asking the Committee's assistance in obtaining data from the United States Geological Survey (USGS) on which conclusions about research results published in ACS journals are based. My request for assistance comes only after numerous efforts to obtain said information, made personally and via a Freedom of Information Act (FOIA) Request, have proven thus far to be unsuccessful.

Information forming the basis of conclusions reached by these USGS scientists concerning refined tar-based pavement sealers (RTS; referred to as "coal tar-based sealants" or a variant containing the words "coal tar" by the USGS) was first formally requested via a FOIA request in April 2010 (USGS 2010-0084). This request, centered on research data and related information, was denied for failure by PCTC to agree to pay associated fees. Essentially the

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same request was made again in April 2011 (USGS 2011-020093) and, in return for over $28,000 in fees, the USGS accepted the request.

In the 2+ years since the "perfected" FOIA request was filed, the USGS has responded with 18 "batches" of materials, including (but not limited to) raw laboratory data packages, thousands of photographs, presentations, copies of published papers with figures, tables and supporting information, advocacy documents, newspaper stories and some tabulated data relating to different pavement sealer research conducted over the past decade. While greatly appreciating the significant volume of FOIA response material received so far, despite repeated follow up requests, the specific data and information underlying several papers published in the ACS Journal of Environmental Science & Technology (ES&T) has still not been produced.

I have been extremely patient in waiting for the USGS to respond by producing the information on which its research conclusions are based. Without this underlying data, the ability to evaluate, understand and reproduce conclusions reached based on the data is significantly impaired. In the meantime, the USGS has been using their research to advocate for bans on RTS while fully aware that sharing of their data has been delayed, if not withheld. A recent newspaper editorial about a long-delayed FOIA response by another federal agency appeared under the headline "Transparency Delayed, Transparency Denied." I couldn't agree more.

With that background, I seek the help of ACS' Committee on Ethics in obtaining the research information detailed in the following paragraphs, organized by ES&T publication.

Mahler, B.J.; Van Metre, P.C.; Bashara, T.J.; Wilson, J.T.; Johns, D.A. Parking lot sealcoat: An unrecognized source of urban polycyclic aromatic hydrocarbons. *Environ. Sci. Technol.* 2005, *39*, 5560–5566.

In 2006 ES&T published a Comment (DeMott and Gauthier, 2006) and Response (Mahler *et al.*, 2006) on the Mahler *et al.* (2005) paper cited above. The Comment focused on two main points:

(1) 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 [*i.e.,* Mahler *et al.* (2005)], and

(2) 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 [*i.e.*, Mahler *et al.* (2005)].

In the Response it was stated that sediment data used to compare polycyclic aromatic hydrocarbon (PAH) double ratio plots by the commenters may not be comparable to the Mahler *et al.* (2005) data however, the authors still did not identify or provide the source of sediment data used in Mahler *et al.* The source of sediment data used in PAH double ratio plots was not identified in Mahler *et al.* (2005), nor was it identified in a companion USGS publication (Mahler *et al.*, 2007).

One PAH double ratio plot showing an apparent overlap of RTS ratios with ratios of PAHs from the unidentified sediment samples has been used by the USGS authors as the basis for conclusions about the purported significant contribution of PAHs to sediment not only in Austin, TX (Mahler *et al.*, 2005) but, in subsequent publications, throughout the United States (see Information Quality Act petition at http://www.usgs.gov/info_qual/documents/Edwards-Wildman-Palmer_PCTC_IQA-Info-Correction-Request051513.pdf).

I ask the Committee's help in identifying the source of sediment sample data used by the USGS to construct the double ratio plots in Figure 4 of Mahler *et al.* (2005).

Mahler, B. J., Van Metre, Peter, Wilson, Jennifer T., Musgrove, Marilyn, Burbank, Teresa L., Ennis, Thomas E. and Bashara, Thomas J. "Coal-tar-based parking lot sealcoat: an unrecognized source of PAH to settled house dust." ***Environ. Sci. Technol.*** **2010, 44: 894 - 900.**

Evaluation and replication of the conclusions reached in Mahler *et al.* (2010) has not been possible due to lack of access to underlying data, including the following.

- Complete tabulated results, including statistical and graphical analysis of data relevant to conclusions reached, for each apartment and parking lot sample collected, including results for all analytes tested (pesticides, flame retardants, PCBs and phthalates, as well as PAHs).,
- For each house dust sample, the mass of dust before and after sieving, the area sampled, and individual PAH concentrations,
- QA/QC data, including samples associated with the 20% of contaminated blank samples,
- Field sampling and collection notes, including equipment calibrations and cleaning procedures, (specifically SOPs for HSV3 operation, sampling and decontamination between samples) ,
- Individual and compiled results of questions asked of households participating in the house dust study and responses given, and

- A means of identifying individual samples and correlating them with specific apartments or parking lots identified in the study and the above-mentioned household questionnaires.

I ask the Committee's help in obtaining the information described above concerning the Mahler *et al.* (2010) publication.

Mahler, Barbara J., Peter Van Metre, Judy Crane, Alison W. Watts, Mateo Scoggins and E. Spencer Williams. "Coal-Tar-Based Pavement Sealcoat and Pahs: Implications for the Environment, Human Health, and Stormwater Management." *Environmental Science & Technology*, 2012.

The ACS-published *Journal of Environmental Science & Technology* has been a repeated venue for publication of sealant research by the USGS-led research team, and in 2012 ES&T published a feature article summarizing the body of the group's pavement sealer research. The USGS-led team has identified RTS as a "dominant" or "significant" contributor of PAHs to sediments and other environmental media using two lines of evidence: the purported overlap of unidentified sediment data with RTS data from a parking lot in Austin, TX, on a PAH double ratio plot (Figure 4 in Mahler *et al.* 2005) and via results of CMB modeling of sediment PAH data from throughout the country (again, for a full explanation, see the Information Quality Act petition at http://www.usgs.gov/info_qual/documents/Edwards-Wildman-Palmer_PCTC_IQA-Info-Correction-Request051513.pdf.). Adaptation of the CMB model to sealer research by the USGS team is described in a paper published in *Science of the Total Environment* (Van Metre & Mahler, 2010). In addition to asking for the assistance of that journal's editor and publisher in obtaining data underlying the CMB model publication, I also ask for your assistance because ES&T has published a number of papers – including the Mahler *et al.* (2012) feature – which rely on the CMB model paper to identify RTS as the source of PAHs in the environment.

In Van Metre and Mahler (2010), it is stated that "The CMB model was run more than 200 times using various combinations of source profiles, fitting parameters (PAHs), estimates of uncertainty, and combinations of lake-sediment samples." Out of the 200 runs, Van Metre and Mahler chose 4 that most closely matched the parameters chosen to represent an undefined "good fit" for the 5 PAH sources chosen as source inputs, and then reported fractional contributions using an average of results of the 4 chosen source input models.

The CMB model is a widely used EPA model, with well-known operating parameters. Researchers who use CMB often publish inputs and outputs of different model runs in support of conclusions reached about particular data sets.

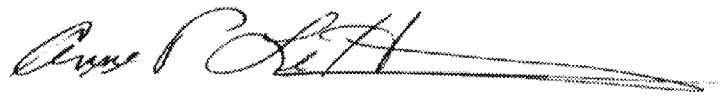
I ask the Committee's help in obtaining the input and output data for the 200 runs mentioned in Van Metre and Mahler (2010), which were referenced and relied upon in the ES&T feature article.

****

Thank you in advance for any assistance you can provide in obtaining the research data critical to evaluating, understanding and reproducing the conclusions published by the USGS-led research team.

Please feel free to contact me at (703) 299-8470 or alehuray@pavementcouncil.org.

Yours truly,



Anne P. LeHuray
ACS Member No. 30043760

cc: Alan D. Thornhill, Director USGS Office of Science Quality & Integrity (athornhill@usgs.gov)
Judy Cearley, USGS FOIA Liaison (jcearley@usgs.gov)
William Wilber, USGS NAWQA Program Chief (wgwilber@usgs.gov)

CITATIONS

DeMott, R.P. and Gauthier, T.D. Comment on "Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons" *Environ. Sci. Technol.* 2006, 40, 3657-3658.

Mahler, B. J., Van Metre, Peter, Wilson, Jennifer T., Musgrove, Marilyn, Burbank, Teresa L., Ennis, Thomas E. and Bashara, Thomas J. "Coal-tar-based parking lot sealcoat: an unrecognized source of PAH to settled house dust." *Environ. Sci. Technol.* 2010, 44: 894 - 900.

Mahler, B.J.; Van Metre P.C., Wilson, J.T. 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, 2004 (revised 2007), 87 p. [Online only]

Mahler, B.J.; Van Metre P.C., Wilson, J.T. Response to Comment on "Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons" *Environ. Sci. Technol.* 2006, 40, 3659-3661.

Mahler, B.J.; Van Metre, P.C.; Bashara, T.J.; Wilson, J.T.; Johns, D.A. Parking lot sealcoat: An unrecognized source of urban polycyclic aromatic hydrocarbons. *Environ. Sci. Technol.* 2005, *39*, 5560–5566.

Mahler, Barbara J., Peter Van Metre, Judy Crane, Alison W . Watts, Mateo Scoggins and E. Spencer Williams. "Coal-Tar-Based Pavement Sealcoat and Pahs: Implications for the Environment, Human Health, and Stormwater Management." *Environmental Science & Technology*, (2012).

Van Metre, Peter and Barbara J. Mahler. "Contribution of Pahs from Coal–Tar Pavement Sealcoat and Other Sources to 40 U.S. Lakes." *Science of the Total Environment* 409, (2010): 334 - 344.

United States Department of the Interior

U.S. GEOLOGICAL SURVEY
Reston, Virginia 20192

In Reply Refer To:
Mail Stop 413

July 22, 2013

Dr. Anne P. LeHuray
Pavement Coatings Sealcoat Council
2308 Mount Vernon Avenue, Suite 134
Alexandria, Virginia 22301

Subject: Re: Request for Assistance in Obtaining Data Collected and Used to Generate Conclusions Published in ACS (and other) Journals

Dear Dr. LeHuray,
Thank you for copying me on your letter to Dr. Gregory Ferrence dated July 8, 2013 requesting assistance in obtaining data and information used in the preparation of three peer-reviewed journal articles authored by Dr. Peter C. Van Metre and Dr. Barbara J. Mahler and published in the journal Environmental Science and Technology. The attached files pertain to the three articles referenced in your letter. This information should be sufficient to evaluate, understand, and reproduce the conclusions published by these authors.

Mahler, B.J., Van Metre, P.C., Bashara, T.J., Wilson, J.T., Johns, D.A., 2005, Parking lot sealcoat: An unrecognized source of urban polycyclic aromatic hydrocarbons, Environmental Science and Technology 39, 5560-5566.

Attachment 1 includes:

- Information to identify the source of sediment sample data used to construct the double ratio plots in Figure 4. These data along with more than 900 other analyses of PAHs performed by the USGS in the past 10 years should be included in the response to the FOIA request that is being processed.

Mahler, B.J., Van Metre, P.C., Wilson, J.T., Musgrove, MaryLynn, Burbank, T.L., Ennis, T.E. and Bashara, T.J., 2010, Coal-tar based parking lot sealcoat: an unrecognized source of PAH to settled house dust, Environmental Science and Technology 44: 894-900.

Attachment 2 includes:

- Tabulated data sufficient to reproduce the experimental observations in Mahler et al, 2010. Data for the other analytes measured (pesticides, flame retardants, PCBs, and phthalates) are not included, as these data and their interpretation were not used in this publication. Individual responses to questions asked of households participating in the house dust study are included (compiled results are provided in the publication).
- Survey results and data for each residence are in a single row.

The field sample equipment used in Mahler et al, 2010 was the Envirometrics Model HVS3 High-Volume Surface Sampler. As noted on p. 895 of the journal article, the sampler was used following the methods recommended by the manufacturer. The manual is available at http://www.envirometrics.com/equip/HVS3manual.pdf. Standard USGS cleaning procedures for equipment used in sediment sampling were used, as documented in the USGS National Field Manual for the Collection of Water-Quality Data, Chapter A8: Bottom-Material Samples (available at http://water.usgs.gov/owq/FieldManual/Chapter8/508Chap8final.pdf). Field sampling and collection notes are being provided to the requestor as part of the FOIA package. They are not included here, however, as they are not necessary to reproduce the experimental observations published, the expressed wish of the requestor.

Mahler, B.J., Van Metre, P.C., Crane, J.L., Watts, A.W., Scoggins, Mateo, and Williams, E.S., 2012, Coal-tar based pavement sealcoat and PAHs: Implications for the environment, human health and stormwater management. Environmental Science and Technology , 46, 6, p 3039-3600.

The original reference for the requested information on the modeling of PAH sources to lakes is Van Metre and Mahler (2010), published in Science of the Total Environment (citation below). Model input data for sources can be obtained from the references in Table 1. All model input data for receptors were provided in Supporting Information Tables S2 and S3 (Van Metre and Mahler, 2010). The rationale for the selection of the four models was provided in Table 3 of the publication and is discussed in the paper. Additionally, the quantitative Chi square, r^2, and percentage of mass estimated, as well as other measures of model "success" such as convergence were provided. As noted in the publication, the four models chosen "are in general agreement with the vast majority of the 200 models tested."

Van Metre, P.C., and Mahler, B.J., 2010, Contribution of PAHs from Coal-Tar Pavement Sealcoat and Other Sources to 40 U.S. Lakes: The Science of the Total Environment, 409, 334-344.

Regards,

William G. Wilber, Ph.D.
Chief, National Water Quality Assessment Program
U.S. Geological Survey

cc:
Peter Van Metre, Hydrologist, USGS Austin, TX

Barbara Mahler, Hydrologist, USGS Austin, TX

Dr. Gregory M. Ferrence, Chairman, American Chemical Society Committee on Ethics

Alan D. Thornhill, Director, USGS Office of Science Quality and Integrity

Judy Cearley, USGS FOIA Liaison

Attachment 1. Selected polycyclic aromatic hydrocarbon compound concentrations in suspended sediment and associated ratios
[Concentrations in ug/kg].

Order of original data	Plot groups	Station ID	Station name	Sampling Date	Sample Time	Fluoranthene	Pyrene	Benzo(e)-pyrene	Benzo(a)-pyrene	Benzo-(g,h,i)-perylene	Indeno-(1,2,3-c,d)-pyrene		F/P	BaP/BeP	IP/BghiP
52	Stream		Williamson Creek GFF	3/17/00	0315	17,391	13,043	9,130	6,522	5,217	8,261		1.33	0.71	1.58
53	Stream		Williamson Creek GFF	5/1/00	0400	10,448	7,980	4,478	3,980	2,985	3,731		1.31	0.89	1.25
54	Stream		Williamson Creek TFE	11/3/00	1410 - 1535	27,000	21,200	12,200	14,300	8,860	14,300		1.27	1.17	1.61
55	Stream		Williamson Creek TFE	5/26/01	1245	12,500	9,360	6,960	6,350	5,930	8,340		1.34	0.91	1.41
56	Stream		Williamson Creek TFE	10/11/01	0600	12,200	8,820	4,940	5,840	4,540	7,130		1.38	1.18	1.57
57	Stream		Williamson Creek TFE	3/19/02	2200	1,720	1,120	496	677	546	660		1.54	1.39	1.21
58	Stream		Williamson Creek TFE	2/20/03	0420	16,300	12,600	5,740	7,030	5,800	7,800		1.29	1.22	1.34
59	Stream		Williamson Creek TFE	3/25/03	1950	15,300	11,500	6,490	6,290	6,180	10,400		1.33	0.97	1.68
60	Stream	08048541	Echo Lake Inflow	5/28/01	0200	801	92	441	<2400	333	<300				
61	Stream	08048541	Echo Lake Inflow	8/17/01	0745	6,310	4,860	2,340	2,920	2,830	3,310		1.36	1.25	1.26
62	Stream	08048541	Echo Lake Inflow	9/20/01	1335	6,050	4,570	2,140	2,300	2,410	3,170		1.32	1.07	1.32
63	Stream	08048541	Echo Lake Inflow	10/11/01	0450	8,130	6,330	2,980	3,380	3,200	4,130		1.28	1.14	1.29
64	Stream	08048620	Fosdic Lake Inflow	8/11/01	1710	1,080	954	436	722	523	515		1.13	1.65	0.98
65	Stream	08048620	Fosdic Lake Inflow	8/18/01	1855	1,130	933	512	600	559	752		1.21	1.17	1.35
66	Stream	08048620	Fosdic Lake Inflow	10/10/01	2000	1,730	1,300	715	775	811	986		1.33	1.08	1.22
67	Stream	08048620	Fosdic Lake Inflow	12/6/01	0108	3,010	2,360	1,330	1,710	1,660	2,220		1.28	1.29	1.32
68	Stream	08047090	Lake Como Inflow	8/30/01	0810	10,400	8,250	4,490	5,120	4,980	6,780		1.26	1.14	1.36
69	Stream	08047090	Lake Como Inflow	9/20/01	1240	9,190	7,360	3,870	4,010	4,640	5,700		1.25	1.04	1.23
70	Stream	08047090	Lake Como Inflow	11/9/01	0915	7,660	6,080	3,160	3,510	4,270	4,680		1.26	1.11	1.10
71	Stream	08047090	Lake Como Inflow	1/23/02	1750	5,630	4,310	2,070	2,620	2,540	3,320		1.31	1.27	1.31

Data for fluoranthene, pyrene, benzo[e]pyrene, and benzo[a]pyrene in Williamson Creek sediments published in USGS Scientific Investigations Report 2006-5262, available at http://pubs.usgs.gov/sir/2006/5262/

Data for fluoranthene, pyrene, and benzo[a]pyrene in Williamson Creek sediments published in USGS Water Resources Investigations Report 03-4169, available at http://pubs.usgs.gov/wri/wri034169/pdf/wri03-4169.pdf

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Attachment 2. Ancillary information and PAH concentrations of settled dust from residences sampled as part of Mahler et al (2010). (Concentrations as ug/kg)

	Sample location	Sealcoat condition	Type of sealcoat	Distance from parking lot to front door (m)	Percent carpeted	Linoleum, tile, or wood floor (Y/N)	Days since floor last cleaned	Wear shoes in the home	Shines	Park bike inside	# smokers	smoker?
1	indoor	n/a	none	4.3	100	N	120	yes	Y	no	0	N
	outdoor	n/a	none	4.3	100	N	120	yes	Y	no	0	N
2	indoor	n/a	none	1.3	100	N	42	yes	Y	no	0	N
	outdoor	n/a	none	1.3	100	N	42	yes	Y	no	0	N
3	indoor	slightly worn	coal tar	8.7	100	N	60	1/2 & 1/2	Y	yes	0	N
	outdoor	slightly worn	coal tar	8.7	100	N	60	1/2 & 1/2	Y	yes	0	N
4	indoor	n/a	none	19.2	0	N	7	yes	Y	yes	2	Y
	outdoor	n/a	none	19.2	0	N	7	yes	Y	yes	2	Y
5	indoor	n/a	none	36	49	N	8	yes	Y	yes	0	N
	outdoor	n/a	none	36	49	N	8	yes	Y	yes	0	N
6	indoor	good	coal tar	0.9	41	N	27	1/2 & 1/2	Y	yes	1	Y
	outdoor	good	coal tar	0.9	41	N	27	1/2 & 1/2	Y	yes	1	Y
7	indoor	several layers, very worn	coal tar	0.8	100	N	9	yes	Y	not answered	0	N
	outdoor	several layers, very worn	coal tar	0.8	100	N	9	yes	Y	not answered	0	N
8	indoor	n/a	none	30	0	Y	10	1/2 & 1/2	Y	no	0	N
	outdoor	n/a	none	30	0	Y	10	1/2 & 1/2	Y	no	0	N
9	indoor	n/a	none	11.8	4	Y	7	no	N	no	0	N
	outdoor	n/a	none	11.8	4	Y	7	no	N	no	0	N
10	indoor	worn	coal tar	14.2	100	N	21	yes	Y	yes	0	N
	outdoor	worn	coal tar	14.2	100	N	21	yes	Y	yes	0	N
11	indoor	n/a	none	7.8	100	N	12	yes	Y	not answered	0	N
	outdoor	n/a	none	7.8	100	N	12	yes	Y	not answered	0	N
12	indoor	n/a	none	8.5	28	N	14	no	N	not answered	0	N
	outdoor	n/a	none	8.5	28	N	14	no	N	not answered	0	N
13	indoor	worn	coal tar	8.1	100	N	14	yes	Y	yes	0	N
	outdoor	worn	coal tar	8.1	100	N	14	yes	Y	yes	0	N
14	indoor	n/a	none	13.4	100	N	30	yes	Y	no	0	N
	outdoor	n/a	none	13.4	100	N	30	yes	Y	no	0	N
15	indoor	older, thick, cracking	coal tar	8.6	0	Y	7	the kitchen	Y	not answered	2	Y
	outdoor	older, thick, cracking	coal tar	8.6	0	Y	7	the kitchen	Y	not answered	2	Y
16	indoor	new & barely worn (1.5 yr old)	asphalt/oil	12.8	100	N	8	no	N	no	0	N
	outdoor	new & barely worn (1.5 yr old)	asphalt/oil	12.8	100	N	8	no	N	no	0	N
17	indoor	not very worn	coal tar	3	100	N	14	no	N	no	0	N
	outdoor	not very worn	coal tar	3	100	N	14	no	N	no	0	N
18	indoor	slightly worn	asphalt	18.5	52	Y	17	yes	Y	no	0	N
	outdoor	slightly worn	asphalt	19.5	52	Y	17	yes	Y	no	0	N
19	indoor	parking spaces	coal tar	20	84	Y	8	no	N	no	0	N
	outdoor	parking spaces	coal tar	20	84	Y	8	no	N	no	0	N
20	indoor	barely worn, newer	coal tar	10.1	100	N	11	1/2 & 1/2	Y	yes	0	N
	outdoor	barely worn, newer	coal tar	10.1	100	N	11	1/2 & 1/2	Y	yes	0	N
21	indoor	brand new; over previous sealcoating	asphalt	12.2	87	Y	10	yes	Y	no	0	N
	outdoor	brand new; over previous sealcoating	asphalt	12.2	87	Y	10	yes	Y	no	0	N
22	indoor	good condition (no cracks)	coal tar	10.7	100	Y	14	no	N	no	0	N
	outdoor	good condition (no cracks)	coal tar	10.7	100	Y	14	no	N	no	0	N
23	indoor	spaces; lots of cracks in pavement	coal tar	13.1	100	N	16	yes	Y	no	0	N
	outdoor	spaces; lots of cracks in pavement	coal tar	13.1	100	N	16	yes	Y	no	0	N

ED_006238_00000702-00125

Attachment 2. Ancillary information and PAH concentrations of settled dust from residences sampled as part of Mahler et al (2010). (Concentrations as ug/kg)

Windows open often	Window level	BBQ grill? Freq of use?	BBQ	Candles/incense	Indoor burning Y or N	# pets	Pet goes outdoor	Typical bike riding locations	Fireplace use	popn (ppl/km2)	multi-family + comm + office + warehouse + streets and roads	desktop computer in same room?	Trips to front door (1-3 scale)	Distance to major road (m)	Anthracene
Never	0	no, but neighbor	Y	no	N	0	N	not answered	no	410	25%	yes	3	71	0
Never	0	no, but neighbor	Y	no	N	0	N	not answered	no	410	25%		3	71	0
Often (weekly)	3	yes/often (1-4x/	Y	no	N	1 dog	Y	not answered	yes	1,581	22%	no	2	562	31
Often (weekly)	3	yes/often (1-4x/	Y	no	N	1 dog	Y	not answered		1,581	22%		2	562	24
Rarely (<1/mo)	1	none/never	N	no	N	0	N	lots, off road	no	2,152	100%	no	1	281	912
Rarely (<1/mo)	1	none/never	N	no	N	0	N	lots, off road	no	2,152	100%		1	281	35,000
Whenever possi	4	yes/Whenever p	Y	no	N	0	N	sidewalks,	no	2,667	50%	no	3	803	136
Whenever possi	4	yes/Whenever p	Y	no	N	0	N	sidewalks,	no	2,667	50%		3	803	214
Often (weekly)	3	none/never	N	not answered		1 cat	N	streets	no	5,548	47%	no	1	16	276
Often (weekly)	3	none/never	N	not answered		1 cat	N	streets	no	5,548	47%		1	16	347
Whenever possi	4	none/never	N	yes, 2x/wk	Y	0	N	lots	no	5,714	48%	no	2	72	148
Whenever possi	4	none/never	N	yes, 2x/wk	Y	0	N	lots	no	5,714	48%		2	72	1,900
Sometimes (mor	2	none/never	N	no	N	1 dog	Y	not answered	no	11,809	73%	no	2	167	2,400
Sometimes (mor	2	none/never	N	no	N	1 dog	Y	not answered	no	11,809	73%		2	167	4,240
Rarely (< 1/mo)	1	none/never	N	Rarely	N	0	N	streets	no	6,590	64%	no	1	204	1,050
Rarely (< 1/mo)	1	none/never	N	Rarely	N	0	N	streets		6,590	64%		1	204	57
Whenever possi	4	none/never	N	no	N	1 dog	Y	not answered	no	5,929	65%	yes	3	456	42
Whenever possi	4	none/never	N	no	N	1 dog	Y	not answered		5,929	65%		3	456	28
Sometimes (mor	2	complex has grill	Y	no	N	0	N	PL	no	4,190	44%	yes	2	208	830
Sometimes (mor	2	complex has grill	Y	no	N	0	N	PL	no	4,190	44%		2	208	23,900
Often (weekly; s	3	none/never	N	some candles	Y	0	N	not answered	no	2,414	38%	no	1	194	0
Often (weekly; s	3	none/never	N	some candles	Y	0	N	not answered	no	2,414	38%		1	194	94
Often (weekly)	3	none/never	N	no	N	visits	N	not answered	no	1,524	84%	no	1	154	202
Often (weekly)	3	none/never	N	no	N	visits	N	not answered	no	1,524	84%		1	154	34
Often (weekly) b	3	none/never	N	yes, candles	Y	2 cats	Y	roads	yes	3,005	56%	no	2	346	662
Often (weekly) b	3	none/never	N	yes, candles	Y	2 cats	Y	roads	yes	3,005	56%		2	346	36,600
Rarely (<1/mo)	1	none/never	N	yes	Y	0	N	not answered	yes	5,171	94%	no	1	362	50
Rarely (<1/mo)	1	none/never	N	yes	Y	0	N	not answered	yes	5,171	94%		1	362	114
Often (weekly)	3	none/never	N	incense	Y	2 cats	N	not answered	no	4,733	80%	no	1	111	114
Often (weekly)	3	none/never	N	incense	Y	2 cats	N	not answered	no	4,733	80%		1	111	2,350
Often (weekly; s	3	none/never	N	some candles	Y	0	N	not answered	no	1,152	55%	no	2	480	0
Often (weekly; s	3	none/never	N	some candles	Y	0	N	not answered	no	1,152	55%		2	480	64
Never	0	none/never	N	yes	Y	0	N	not answered	no	1,010	85%	no	2	214	3,180
Never	0	none/never	N	yes	Y	0	N	not answered	no	1,010	85%		2	214	27,700
Rarely (<1/mo)	1	none/never	N	candles	Y	2 dogs	Y	not answered	no	318	52%	yes	2	266	82
Rarely (<1/mo)	1	none/never	N	candles	Y	2 dogs	Y	not answered	no	318	52%		2	266	50
Whenever possi	4	none/never	N	use an alcohol	Y	0	N	streets	no	1,767	85%	no	2	211	1,050
Whenever possi	4	none/never	N	use an alcohol	Y	0	N	streets	no	1,767	85%		2	211	17,300
Whenever possi	4	none/never	N	yes	Y	1 dog, 1 cat	Y	PL	yes	1,414	91%	no	1	56	1,170
Whenever possi	4	none/never	N	yes	Y	1 dog, 1 cat	Y	PL	yes	1,414	91%		1	56	88,400
Whenever possi	4	yes/often (1-4X/	Y	no	N	0	N	not answered	no	543	87%	no	1	135	350
Whenever possi	4	yes/often (1-4X/	Y	no	N	0	N	not answered	no	543	87%		1	135	79
sliding glass doo	4	none/never	N	no	N	1 dog	Y	not answered	yes	3,152	59%	no	2	802	278
sliding glass doo	4	none/never	N	no	N	1 dog	Y	not answered	yes	3,152	59%		2	802	7,660
Sometimes (mor	2	none/never	N	2x/week	Y	1 dog, 3 cats	Y	not answered	no	4,348	54%	yes	1	738	323
Sometimes (mor	2	none/never	N	2x/week	Y	1 dog, 3 cats	Y	not answered	no	4,348	54%		1	738	13,200

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Attachment 2. Ancillary information and PAH concentrations of settled dust from residences sampled as part of Mahler et al (2010). (Concentrations as ug/kg)

Benzo(a)pyrene	Fluoranthene	Naphthalene	Benz(a)anthracene	Phenanthrene	Pyrene	9H-Fluorene	Acenaphthene	Acenaphthylene	Benzo(b)fluoranthene	Benzo(g,h,i)perylene	Benzo(k)fluoranthene	Chrysene	Dibenz(a,h)anthracene	Indeno(1,2,3-cd)pyrene
249	807	55	169	333	500	0	0	9	476	246	176	318	0	188
64	267	0	0	130	209	0	0	9	137	59	51	121	0	49
56	170	20	47	178	145	0	0	0	137	59	56	106	0	45
139	479	0	186	170	369	0	0	0	292	53	152	369	0	48
7,330	32,700	489	6,240	15,300	25,000	763	778	264	14,700	6,520	5,040	15,200	0	5,330
518,000	3,119,000	8,120	401,000	1,340,000	2,330,000	23,200	29,600	13,400	1,130,000	438,000	404,000	1,180,000	0	369,000
1,360	3,440	0	952	1,350	2,890	0	0	0	2,910	1,150	1,100	1,980	0	1,050
2,970	12,300	55	2,830	3,780	9,840	120	0	41	5,480	2,230	1,620	5,290	0	1,690
2,050	6,430	0	1,860	1,800	5,880	138	0	0	4,000	1,280	1,380	1,090	0	1,380
3,360	7,960	143	2,490	2,370	6,570	94	53	71	8,650	2,320	2,320	5,080	0	2,200
1,210	4,420	138	934	1,220	3,620	73	60	45	2,700	1,220	798	2,000	0	979
26,600	81,100	820	19,600	22,400	64,300	433	257	257	54,000	25,000	21,900	46,700	0	21,800
14,300	44,200	227	14,700	16,200	32,400	1,080	788	322	28,900	12,300	10,400	24,700	0	11,300
36,700	136,000	369	33,100	41,200	69,500	1,350	859	1,330	88,500	22,800	30,400	69,400	0	23,700
12,400	43,700	0	9,420	17,300	34,400	500	0	367	35,000	10,700	8,380	21,100	0	8,520
490	1,620	38	465	556	1,520	27	23	0	1,060	351	343	943	0	395
283	882	44	184	361	599	0	0	17	548	266	225	384	0	204
180	781	16	187	309	595	16	14	5	406	124	174	369	0	108
4,500	13,800	154	4,150	5,630	10,100	274	215	158	8,330	3,230	3,090	8,940	0	3,110
285,000	1,120,000	3,360	289,000	517,000	795,000	18,100	18,800	6,775	582,000	186,000	199,000	548,000	0	174,000
154	411	0	97	0	344	0	0	0	259	204	97	179	0	196
1,050	3,170	66	878	637	2,540	0	38	0	2,280	813	714	1,890	0	702
1,500	5,820	0	1,190	1,940	4,890	0	0	0	2,330	845	949	1,610	0	958
501	1,090	14	348	388	866	17	12	10	733	491	318	568	0	469
8,115	36,050	260	5,390	13,700	26,750	260	223	187	16,050	5,370	4,680	15,900	0	4,700
555,000	2,070,000	5,940	461,000	835,000	1,520,000	14,600	16,100	12,200	1,080,000	448,000	427,000	972,000	0	426,000
226	714	109	203	467	683	0	0	21	477	195	181	445	0	144
1,350	3,480	79	869	1,280	2,840	56	42	43	2,740	729	1,000	2,610	0	882
1,410	2,780	54	3,890	797	2,415	45	0	41	2,730	2,010	1,140	6,870	0	2,050
21,400	195,000	679	19,800	32,100	82,700	671	522	476	49,800	14,600	16,500	44,800	0	13,600
304	888	0	0	252	575	0	0	0	694	314	247	802	230	296
581	2,400	0	398	404	1,480	0	0	27	1,760	627	560	1,380	0	546
24,200	70,700	633	20,800	25,300	55,100	1,310	930	300	38,400	22,200	15,200	38,300	0	18,700
511,000	1,650,000	4,070	407,000	328,000	1,290,000	5,510	1,720	3,490	1,010,000	323,000	326,000	765,000	0	309,000
577	1,000	0	346	550	831	0	0	0	888	415	386	614	0	449
302	861	36	220	198	862	0	0	14	704	252	374	483	0	274
10,800	32,100	487	7,700	11,800	25,200	414	216	233	20,800	10,500	7,070	15,600	0	8,270
395,000	1,420,000	6,100	254,000	288,000	1,090,000	5,040	2,460	3,020	572,000	198,000	213,000	516,000	0	179,000
15,200	39,000	466	13,200	12,400	30,800	446	0	193	25,500	12,300	9,470	20,600	0	10,100
602,500	2,385,000	9,970	616	803,000	1,775,000	23,600	14,435	4,805	1,030,500	373,000	384,000	903,500	0	341,500
3,080	8,565	260	1,460	2,585	5,560	173	0	203	5,265	1,820	2,230	3,690	0	1,825
595	2,610	0	491	650	1,670	0	16	29	1,760	306	560	1,840	0	288
4,040	9,790	212	4,010	1,980	8,250	109	0	48	8,610	4,530	2,730	6,750	0	3,400
131,000	407,000	4,100	134,000	40,900	336,000	1,650	795	1,130	349,000	102,000	107,000	299,000	0	88,000
3,420	9,040	221	3,170	2,480	7,990	113	64	56	5,660	2,360	2,240	5,210	0	2,130
149,000	520,000	3,440	166,000	120,000	454,000	3,840	2,150	2,130	282,000	124,000	105,000	287,000	0	103,000

ED_006238_00000702-00127

ATTACHMENT C

Email to Leila Gosselink from Barbara Mahler, dated July 19, 2004.

Research Associate

Statistics & Information Analysis

Battelle Memorial Institute

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leibranda@battelle.org

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SendTo: leila.gosselink@ci.austin.tx.us

CopyTo: ed.peacock@ci.austin.tx.us,nancy.mcclintock@ci.austin.tx.us,CN=Peter C VanMetre/OU=WRD/OU=USGS/O=DOI@USGS,jenwilso@usgs.gov
BlindCopyTo:
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$Abstract: Leila, My apologies for not responding sooner, we were in the field (Wisconsin) and out of e-mail co
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Leila,

My apologies for not responding sooner, we were in the field (Wisconsin) and out of e-mail contact all of last week, just got in late late late last night.

The following ratios of PAHs were useful in differentiating between the different types of parking lots:

benzo(a)pyrene:benzo(e)pyrene versus
indeno(1,2,3-c,d)pyrene:benzo(g,h,i)perylene
fluoranthene:pyrene versus benzo(a)pyrene:benzo(e)pyrene
phenanthrene:anthracene versus fluoranthene:pyrene

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.

We are very interested to find out how your bed sediment look when graphed this way!

Barbara

****
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****

leila.gosselink@ci.austin.tx.us
07/14/2004 03:56 PM

To: bjmahler@usgs.gov, pcvanmet@usgs.gov
cc: ed.peacock@ci.austin.tx.us, nancy.mcclintock@ci.austin.tx.us
Subject: PAH ratios and fingerprinting

Could you guys fill me in again on what PAH ratios had looked kind of hopeful to differentiate PAH sources (I think even between asphalt and coal tar surfaces). In particular, I was interested in the ratios between the primary PAHs so that I can see if those ratios stick around as they move through the environment (did you all look at the LVSS in Williamson and see if it also looked one or the other way?).

Thanks, Leila

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