

United States Department of the Interior

U.S. GEOLOGICAL SURVEY Reston, Virginia 20192

In Reply Refer To: Mail Stop 150

MAR 1 9 2014

Leonard S. Kurfirst Surdyk and Baker 225 West Washington Street, Suite 2200 Chicago, Illinois 60606

Dear Mr. Kurfirst:

The U.S. Geological Survey (USGS) received the two "Information Quality Act Request for Correction of Information" dated May 15, 2013, and May 31, 2013, which you sent via email on behalf of the Pavement Coating Technology Council (PCTC) regarding USGS publications on the same subject. Information related to the two requests, as well as links to the USGS publications cited in the requests, may be viewed on our web page at: http://www.usgs.gov/info qual/coal tar sealants.html.

The attached response document recaps the PCTC's recommendations for specific corrective action, as well as various comments made in the requests, and provides the USGS response to these specific recommendations and comments. I have reviewed the evaluation of your request and find the USGS responses to be comprehensive in relation to the Information Quality Act (IQA) requirements.

As stated in the attachment, we find the substantive information underlying the publications referred to in the requests to be technically correct, unbiased, and objective and no corrective action will be taken for them. Because news releases are exempt from the IQA process, they are not addressed in the USGS response. The USGS has determined that there was no indication or suggestion that the materials presented at our web site <u>http://tx.usgs.gov/coring/allthingssealcoat.html</u> were intended to represent all relevant scientific literature on the topics presented, but we have changed the URL for this page to <u>http://tx.usgs.gov/sealcoat.html</u> and added the words "USGS Research" to the title for additional clarification of this point.

An appeal process is available if there is any dissatisfaction with our decision regarding the two requests. The procedure for this process is found on the USGS Information Quality Guidelines web site at <u>http://www.usgs.gov/info_qual/</u>.

Sincerely,

Dr. Jerad D. Bales Acting Associate Director for Water

Attachment

USGS Response for two IQA Requests for Information Correction (http://www.usgs.gov/info_qual/coal_tar_sealants.html)

USGS Response to Specific Requested Corrective Action and Comments in the May 15, 2013 -Initial Inquiry:

Requested Corrective Action (page 7): "Since the Information promulgated on the web page titled "Coal Tar Sealant Largest Source of PAHs in Lakes" does not meet USGS or OMB guidelines for information quality, the USGS should remove the aforementioned web page from the NAWQA web site. Likewise, it is also important for the USGS to remove the related press release in its entirety."

USGS Response: The web page <u>http://water.usgs.gov/nawqa/home_maps/sealcoat,html</u> satisfactorily meets the USGS Fundamental Science Practices (FPS) review, approval, and release requirements and as such no corrective action to remove the web page will be taken. "Coal Tar Sealant Largest Source of PAHs in Lakes" <u>http://www.usgs.gov/newsroom/article.asp?ID=2651</u> is the title of a news release. News releases are exempt from USGS Information Quality Guidelines (<u>http://www.usgs.gov/info_qual/</u>), and as such, it will not be removed from the web site or discussed further in this response document.

Requested Corrective Action (page 8): "If the USGS insists on posting and emphasizing for the public some of the findings set forth with the 40 Lakes Paper, then the USGS Guidelines and Manual mandate that it must be done in a way that accurately and objectively reflects the scientific debate that surrounds the issue of coal tar sealants and their hypothetical contribution to sediment contamination. Should the USGS or its scientists choose to go down this route, the undersigned respectfully requests that the PCTC be permitted to provide its input before anything is posted. Hopefully, this type of review will obviate any need for another DQA challenge. The posting of a new web page would also require the issuance of a new press release that properly describes the limited impact that the 40 Lakes Paper has had on this debate."

USGS Response: It is the policy of the USGS that results of its investigations be made available in an impartial and objective manner that will best serve all the public. To ensure this objectivity, the USGS peer review standards require a minimum of two independent scientific reviews for every USGS publication. Research managers and independent scientists assess the author's responses to reviews to ensure those responses are adequate. Approval to publish USGS information products containing new research or interpretive information following peer review is granted by Bureau Approving Officials in the USGS Office of Science Quality and Integrity. When appropriate, USGS scientists may request a courtesy review of material from industry and other outside organizations before public release. But, decisions on the review of material are done on a case-by-case basis. We do not make blanket agreements with individuals or organizations on the review of publications associated with particular topics and external input is carefully reviewed for appropriateness because of the potential for conflict of interest.

Requested Corrective Action (page 8): "Finally, the USGS website "all things sealcoat", at the very least must include appropriate citations to those peer reviewed articles and related publications that have respectfully and scientifically challenged the findings of the USGS "coal tar sealant" articles authored by Drs. Van Metre and Mahler. An alternative solution is to delete the website altogether so

that the public does not mistakenly believe that it reflects an impartial and complete collection of all relevant scientific literature regarding coal tar sealcoating. Clearly, the USGS website has failed in this regard."

USGS Response: "allthingssealcoat" is part of the web address

http://tx.usgs.gov/coring/allthingssealcoat.html. The actual title of the web page is: USGS PAHs and Coal-Tar-Based Pavement Sealcoat. This web page is one of several prepared by the USGS Texas Water Science Center for the purpose of providing stakeholders and the public with background and major findings from USGS projects ongoing in the Center. We do not think there is any indication or suggestion on these web pages that the material presented is intended to represent all scientific literature on the topics presented. However, for additional clarification, the words "USGS Research" have been added to the title, and the URL for this web page has been changed to <u>http://tx.usgs.gov/sealcoat.html</u>.

PCTC Comment (page 2, paragraph 2): <u>http://tx.usgs.gov/coring/allthingssealcoat.html</u> "While the name of the link makes it sound as though it contains "all things" related to seal coating – including all relevant research – a careful review of this USGS sponsored link discloses that it instead consists of little more than a collection of articles primarily written by the same two USGS scientists –Drs. Van Metre and Mahler. They rely heavily on the practice of self-citation and tend to repeat many of their observations and conclusions, sometimes in different journals and publications. The most disturbing fact, however, is that peer reviewed studies which challenge the findings and conclusions reached by Drs. Van Metre and Mahler in the 40 Lakes Paper, and in other similar papers published by them, have simply been ignored by the USGS as though they did not exist. Such an approach reflects a deviation in sound scientific methodology as well as a clear breach of the USGS Information Quality Guidelines summarized below."

USGS Response: As stated above, the words "USGS Research" have been added to the title, and the URL for the web site has been changed to: <u>http://tx.usgs.gov/sealcoat.html</u>. This web page is one of several prepared by the USGS Texas Water Science Center for the purpose of providing stakeholders and the public with background and major findings from USGS projects ongoing in the Center. Drs. Van Metre and Mahler are USGS researchers at the Texas Water Science Center and the commenter is correct that the web page is intended to highlight USGS research on the subject.

With regard to the statement: "peer reviewed studies which challenge the findings and conclusions reached by Drs. Van Metre and Mahler in the 40 Lakes Paper, and in other similar papers published by them, have simply been ignored by the USGS as though they did not exist." Where there is a logical scientific basis for referencing peer-reviewed publications, including PCTC-sponsored publications, the USGS will reference them. However, only two of the 18 citations listed in Exhibit B: "publications of scientific studies of tar-based sealants in the environment sponsored by the Pavement Coatings Technology Council" were published in the peer-reviewed literature (DeMott et al., 2010; O'Reilly et al., 2012) and only one of these (O'Reilly et al. 2012) challenges the findings of the "40 Lakes paper" (Van Metre and Mahler, 2010). Comments in the second PCTC-sponsored journal article (DeMott et al., 2010) are being addressed in a USGS-authored manuscript that is currently in review. Two additional PCTC citations were published as comments to the journal Environmental Science and Technology (DeMott and Gauthier, 2006; O'Reilly et al., 2011). Comments are published by the journal but they are not subject to

scientific peer review. Both of the Comments were responded to by Drs. Van Metre and Mahler and coauthors in the same journal in which the Comments were published (Mahler et al., 2006; Van Metre and Mahler, 2011). USGS supports the use of scientific journals for this type of technical exchange between scientists.

PCTC Comment (page 3, paragraph 3): "Another way to determine if any bias or advocacy exists within the USGS on the issue of coal tar sealants is for the USGS to produce all related data, correspondence and emails concerning its research regarding the 40 Lakes Paper and other similar articles. A FOIA request asking for such materials was sent off two years ago and incredibly, still remains "open" today. As will be demonstrated in greater detail below, the USGS has sought to minimize evidence of advocacy within its ranks by withholding, at least up to now, certain correspondence and email between the USGS staff and other individuals outside the agency who have made it their goal to ban coal tar sealants across the country. These efforts by USGS to withhold certain documents are not only at odds with the above mentioned policies, but also contrary to the need for transparency that is emphasized throughout USGS Guidelines. See e.g., 67 F.R. 8453, 8459."

USGS Response: The aforementioned information was part of a Freedom of Information Act (FOIA) request which is the appropriate mechanism to use to request such information. The following 22 batches of information were sent by express mail on the dates indicated in response to the request; and it is now complete, therefore, no additional action will be taken.

- 1 2 boxes (1 box = 15 in x 12 in x 10 in); 44 pages of emails; 10/11/2011
- 2 CD-ROM; 10 electronic files; 11/3/2011
- 3 CD-ROM; 412 electronic files; 11/18/2011
- 4 CD-ROM; 372 electronic files; 12/29/2011
- 5 CD-ROM; 251 electronic files 12/30/2011
- 6 1 box, and CD-ROM, 583 electronic files, 3/1/2012
- 7 DVD; 160 electronic files, 3/6/2012
- 8 CD-ROM; 345 electronic files, 5/21/2012
- 9 CD-ROM; 81 electronic files; 5/22/2012
- 10 CD-ROM; 81 electronic files 6/4/2012
- 11 5 boxes; 7/27/2012
- 12 CD-ROM; 79 electronic files; 8/10/2012
- 13 4 boxes; 11/1/2012
- 14 5 boxes; 11/2/2012
- 15 1 box; 11/27/2012
- 16 CD-ROM; 2 electronic files; 4/25/2013
- 17 CD-ROM; 7 electronic files and includes 1060 pages of emails 5/31/2013
- 18 CD-ROM; 4 electronic files and includes 653 pages of emails 6/14/2013
- 19 CD-ROM; 5 electronic files and includes 789 pages of emails; 7/8/2013

20 - CD-ROM; 2 electronic files (includes an index with information for approximately 299 files we are withholding in full); 7/25/2013

21 - CD-ROM; 6 electronic files (includes indexes with information for approximately 707 files we are withholding in full); and includes 1731 pages of emails; 7/29/2013

22 - CD-ROM; 3 electronic files (includes an index with information for approximately 3 files we are withholding in full); 8/22/2013

Additional questions related to this FOIA request should be addressed to Mr. Ken Skipper at (303)236-5050 x 230) or kskipper@usgs.gov

PCTC comment (page 4, paragraph 1): "Drs. Van Metre and Mahler claim in their 40 Lakes Paper that contamination of urban lakes and streams by polycyclic aromatic hydrocarbons (hereinafter PAHs) is widespread in the U.S. and has been increasing over the last 40 years. This assertion is not particularly surprising since there is a consensus in the scientific community that PAHs have many potential sources, including vehicle emissions, tire particles, motor oil, crude oil, power plant emissions and industrial releases. Indeed, almost any type of combustion with organic matter will produce PAHs as a by-product, including natural sources such as forest fires and volcanoes down to something as basic as grilling on the backyard barbecue. Thus, one would expect PAHs to be ubiquitous in our environment and, in fact, they are."

USGS Response: Drs. Van Metre and Mahler have noted the many PAHs sources in the urban environment in numerous publications and presentations. For example, 22 urban PAH sources were considered in source-receptor modeling that identified coal-tar-based sealcoat as a major contributor to PAHs in lake sediments (Van Metre and Mahler, 2010). Ubiquity is not synonymous with source strength, however. Lead, for example, occurs naturally and thus is ubiquitous in the environment, and yet a single source, vehicle emissions during use of leaded gasoline, was widely recognized as the dominant source of lead to urban settings (Eisenreich et al., 1986; Callender and Van Metre, 1997). Coal-tar-based sealants stand out relative to all other urban PAH sources (with the exception of other coal-tar-related sources such as coal-gasification waste sites) as having extremely high concentrations of PAHs. PAH concentrations in coal tar sealants tested by the City of Austin averaged 66,000 milligrams per kilogram (mg/kg), about 100–1,000 times higher than PAH concentrations reported for vehicle emissions, tire particles, asphalt sealants, and used motor oil, which range in concentration from about 50 mg/kg to about 500 mg/kg (Mahler et al., 2012).

PCTC comment (page 4, paragraph 2): "Drs. Van Metre and Mahler use a source identification and apportionment method known as the Chemical Mass-Balance Model (hereinafter CMB) to link the PAHs associated with coal tar sealed parking lots to those found in urban lake sediments. Without the CMB modeling results, there is little basis for anyone to conclude that coal tar sealants are the "dominant" or "most substantial" source of PAH contamination in lake sediment. Thus, if the CMB modeling is in some way flawed, incomplete, or inconclusive, then it necessarily follows that the findings published by Drs. Van Metre and Mahler in 40 Lakes Paper must be characterized in the same way. It also follows that the bold and unqualified assertions set forth within the challenged USGS web site and press release would need to be withdrawn or modified significantly to properly reflect the scientific uncertainty that exists."

USGS Response: The USGS application of the CMB model was thoroughly documented in a peerreviewed scientific journal article (Van Metre and Mahler, 2010). The CMB modeling study builds on previous studies that link coal-tar-based sealant to PAH contamination in lakes. For example, direct microscopic identification of coal-tar-pitch particles in pavement dust, street dust, soil, stream sediment, and lake sediment in an urban watershed provided direct evidence of the transport of coal-tar-pitch from sealcoated parking lots to urban lakes, and of the importance of that coal-tar-pitch as a PAH source in the stream and lake sediment (Yang et al., 2010). That study used an approach entirely independent of the CMB model or of any of the other PAH assemblage (fingerprinting) approaches such as PAH ratio analysis. The estimated PAH contribution to Como Lake by Yang et al. (2010) was consistent with the results of the CMB modeling for the same lake (Van Metre and Mahler, 2010). In another study, PAH source-indicator ratio analyses in pavement dust and lake sediment showed greater similarity of the PAH assemblage between lake sediment and dust from coal-tar-sealed pavement in the same watershed than to any other urban PAH sources tested (Van Metre et al., 2009). That same study also showed that PAH concentrations were much lower in two western U.S. lakes and in sealed-pavement dust from their watersheds than in central and eastern U.S. lakes and sealed-pavement dust (~10 fold and ~1,000 fold differences in lake sediment and dust, respectively). These findings confirmed the anecdotal evidence that asphalt-based sealants are commonly used in the western U.S. and added another, geographic, line of evidence of the importance of coal-tar sealant as a PAH source. Other independent researchers of coal-tar sealants and environmental pollution have determined that: coal-tar-sealant is a major cause of PAH contamination in Minnesota stormwater ponds (Crane et al., 2010; Crane et al., 2014); coal-tar sealant is the largest source of PAHs to streams and ponds in Springfield, Mo. (Pavlowsky, 2012); runoff of PAHs from coal-tar-sealcoated parking lots contaminates downstream sediment and stormwater structures (Watts et al., 2010); PAH concentrations in runoff from coal-tar-sealed pavement greatly exceeds that in runoff from unsealed pavement (Selbig et al, 2009); and PAH concentrations in urban streams downstream from inflows from sealed parking lots exceed concentrations upstream from the inflows (adversely affecting ecological communities downstream) (Scoggins et al., 2007). Thus, there are numerous studies that use a wide variety of approaches by several independent research groups that have concluded that coal-tar-based sealants are a major source of PAHs to streams and lakes.

PCTC Comment (page 4, paragraph 4): "First and foremost, even Drs Van Metre and Mahler acknowledge in their 40 Lakes Paper that the alleged accuracy of the CMB model is dependent upon five crucial assumptions, such as chemical species not reacting with each other or the environment. It goes without saying that if any of the five assumptions are incorrect, then the ultimate conclusions offered by Drs. Van Metre and Mahler are anything but certain. The attached PCTC White Paper explains in detail how all five assumptions are likely to be mistaken, some more than others. This is not surprising since the CMB model is an atmospheric source allocation model and generally not used to evaluate sediment. Drs Van Metre and Mahler have done little to demonstrate that their five CMB assumptions in the 40 Lakes Paper are sound. Thus, neither they nor the USGS can proclaim or even suggest that they have in fact proven that "coal tar based pavement sealant is the largest source of polycyclic aromatic hydrocarbons (PAHs) in 40 urban lakes studied by the US Geological Survey" At most, they have presented some data on a hypothesis that requires further testing and analysis."

USGS Response: The peer reviewers and journal editor for the 40 Lake Paper concluded that the work done by Van Metre and Mahler (2010) was a technically sound application of the CMB model and the USGS and the journal approved the manuscript for publication. We refer the PCTC commenters to the CMB manual (Coulter, 2004) which states:

"The six assumptions are fairly restrictive and they will never be totally complied with in actual practice. Fortunately, CMB can tolerate reasonable deviations from these assumptions, though these deviations increase the stated uncertainties of the source contribution estimates (Cheng and Hopke, 1989; Currie et al., 1984; deCesar et al., 1985, 1986; Dzubay et al., 1984; Gordon et al., 1981; Henry, 1982, 1992; Javitz and Watson, 1986; Javitz et al., 1988a, 1988b; Kim and Henry, 1989; Lowenthal et al., 1987, 1988a, 1988b, 1988c, 1992, 1994; Watson, 1979)."

Note: Six assumptions are listed in Coulter (2004), not five. The assumption missing from the PCTC list and Van Metre and Mahler (2010) is "the number of sources or source categories is less than or equal to the number of species". This assumption, although not listed in Van Metre and Mahler (2010), was followed in all of the USGS modeling runs.

Responses to each of the comments in the table on p. 12-13 of exhibit 1 for each the five model assumptions are provided below:

<u>Assumption 1</u>: For a national assessment, the emissions from a potential source are reasonably assumed to be independent of the geographical location of the source. For example, the composition of tire particles in Florida is assumed to be similar to the composition in Wisconsin. We are unclear as to the basis of the criticism of the use of "averages of published data". We also are unclear as to the basis of the comment on "types of emission sources used are known not to be consistent..." If the commenters are referring to emissions in vapor and particles phases, as noted in Van Metre and Mahler (2010), many of the sources were from the compilation of Li et al. (2003), who resolved this problem by converting literature source profiles that contain both gaseous and particulate PAHs to particulate-only profiles. With the exception of fuel-oil combustion, the other profiles used in model runs were from PAHs measured in the particulate phase from wood burning (Schauer et al., 2001), tires and asphalt (Boonyatumanond et al., 2007), and coal-tar-based sealcoat (Mahler et al., 2005; Van Metre et al., 2009).

<u>Assumption 2</u>: PAHs do indeed react in the atmosphere, which is part of the reasoning employed by Li et al. (2003) in converting air emissions to particle-only profiles. The issue of transformation, and in particular in the atmosphere, was explicitly discussed in Van Metre and Mahler (2010) in the third paragraph of the Introduction:

"Source-receptor modeling relies on the assumption that there is no change in PAH profile from source to receptor. Some researchers have rationalized that the assumption is justified for PAHs in lake sediment because PAHs are relatively conservative, the transport pathway is short, burial is rapid, and(or) sediments are anoxic (Christensen et al., 1997; Rachdawong et al., 1998; Li et al., 2003; Bzdusek et al., 2004). More critical approaches include those of Simcik et al. (1999), who reported little change in measured PAH profiles between the atmosphere and surficial lake sediment, and Li et al. (2001), whose modeling results demonstrated that only phenanthrene underwent significant degradation in aerobic sediment and that no degradation occurred in deeper, anoxic sediment layers."

<u>Assumption 3</u>: The comment that the "USGS application considered a limited set of sources assumed to be applicable nation-wide" is false. Twenty-two different PAH sources and profiles were considered in the 40 Lake Paper (Table 1, Van Metre and Mahler, 2010). This is one of the most extensive lists of PAH

sources considered for source-receptor modeling and the first to include coal-tar sealants in this type of modeling. Regarding the contention that the profiles do not represent "actual sources" in the watersheds considered, the USGS responses to Assumptions 1 and 2 apply.

<u>Assumption 4</u>: The model assumption of linear independence of sources does not mean that the mass contribution of each source is independent, but rather that the chemical composition of each source is independent of the other sources (i.e., that the PAH profiles are sufficiently different for the model to distinguish between them). In some situations, it is quite possible, and even logical, that a larger contribution of PAHs from one source (e.g., sealcoat) would coincide with a larger contribution from another source (e.g., vehicles) because both are associated with urbanization. More urbanization logically leads to more of both sources which might result in a positive relationship between them.

<u>Assumption 5</u>: The adoption of a constant 40% uncertainty followed the approach used by Li et al. (2003). The analytical uncertainty for measuring PAHs in sediments by the USGS typically ranges from 10-20%, depending on the PAH. A larger uncertainty was used because the true uncertainty of the potential source profiles is unknown. In response to the final comment: "*Profiles based on a limited set of published data are not expected to be random, uncorrelated, or normally distributed*", the assumption is that "measurement uncertainties" have those properties, not the profiles.

PCTC Comment (page 5, paragraph 2): "Second, Drs. Van Metre and Mahler, along with other scientists, have used other types of chemical fingerprinting methods in the past to compare sealant PAHs with those found in sediment. These other methods relied upon the comparison of various PAH ratios. Details are provided in the attached materials referenced above. Suffice it to say that the USGS adaptation of the CMB model was shown to be inconsistent with all other PAH source apportionment methods, thereby generating further uncertainty regarding the accuracy of the CMB model as utilized by Drs. Van Metre and Mahler. Since reproduction of findings is crucial in order to demonstrate the validity of any hypothesis, these inconsistencies cannot be ignored."

USGS Response: This criticism of Van Metre and Mahler (2010) was made in the Comment to Environmental Science and Technology by O'Reilly et al. (2011) and was addressed in detail in the Response to Comment by Drs. Van Metre and Mahler (Van Metre and Mahler 2011), and the USGS stands by that previous response. That Response states in part:

"O'Reilly et al.'s principal criticism is a supposed lack of a relation between the CT-sealant contributions estimated using the contaminant mass balance model (CMB)³ and PAH ratios for those samples² yet we find a consistent and logical relation. Progressing by quartiles of proportional CTsealant contribution from low to high for the 120 samples from 40 lakes used in ref 3, 16, 44, 70. 94% have fluoranthene:pyrene ratios > 1.15; 16, 41, 60, and 53% of samples have benzo[a]pyrene:benzo-[e]pyrene ratios > 0.87; these ratio thresholds, used in ref 2, were not arbitrary, but rather bounded the samples of sealed parking lot dust from central and eastern U.S. cities where CT-sealant use is common."

PCTC Comment (page 5, paragraph 3): "Third, one of the most basic procedures available for establishing the validity of a hypothesis is to subject the data collected to the null hypothesis. In other words, have Drs. Van Metre and Mahler demonstrated that the same sediment results could not have occurred in the absence of coal tar sealants? Their answer is no. Quite simply, the null hypothesis was not tested by Drs. Van Metre and Mahler as part of their analysis in the 40 Lakes Paper or in any other paper that they have published related to coal tar sealants. Thus, they have not eliminated, for example, atmospheric deposition of PAHs as a primary source of sediment contamination, as opposed to coal tar sealant runoff."

USGS Response: This criticism of Van Metre and Mahler (2010) also was made in the Comment to the Journal, Environmental Science and Technology by Dr. O'Reilly et al. (2011) and was addressed in the Response to Comment by Drs. Van Metre and Mahler (2011). Their Response to Comment specifically addressed the contention that they had "not eliminated...atmospheric deposition of PAHs as a primary source", and the USGS stands by that previous response, which stated:

"O'Reilly et al. contend that we "have not eliminated atmospheric deposition as a major source". In fact, in ref 3 we considered many sources of PAHs with atmospheric pathways, including vehicle emissions, coke-oven emissions and coal, oil, and wood combustion. Vehicle and coal-combustion sources combined accounted for an average of 41% of PAHs in the 40 lakes, slightly less than the contribution from CT-sealants of 50%."

The null hypothesis is tested by the CMB modeling, in the sense of having concluded for some lakes that coal-tar sealants are not a major source of PAHs. Regional differences in PAH sources were clearly indicated by the modeling. The following passage from Van Metre and Mahler(2010) discusses this issue:

Page 340: "High PAH concentrations are not necessarily associated with density of urban land use or proximity to highways. The western urban lakes Tanasbrook Pond (TNB) in Portland, Ore., and Decker Lake (DEK) in Salt Lake City, Utah, have dense urban watersheds (population densities of 844 and 2,100/km², respectively, and a major highway in each) but low Σ PAH concentrations (1.34 and 0.76 mg/kg, respectively; means of the three samples used for modeling). In contrast, the watershed of Lake Anne (ANN) in suburban Washington, D.C., has a population density of 2100/km², similar to that of DEK, but a Σ PAH concentration of 17.0 mg/kg, 22 times greater than that of DEK. The watershed of Palmer Lake (PLM) in suburban Minneapolis, Minn., has a population density of 939/km², similar to that of TNB, but a Σ PAH concentration of 34.1 mg/kg, about 25 times greater than that of TNB. Modeling results indicate that CT sealcoat contributes about 14 and 24 mg/kg of PAH to the sediment of ANN and PLM, respectively, but contributes only 0.80 and 0.26 mg/kg to the sediment of TNB and DEK (Supporting information Table S5). Thus, loading from CT sealcoat appears to be a much stronger driver than urbanization or population density for elevated concentrations of PAHs in urban lake sediment."

The above passage shows that the presence or absence of use of sealants results in a very different level of PAHs in the lakes and that the model is able to handle both situations. Temporal differences in PAH sources further demonstrate that the researchers and the CMB model reached logical conclusions regarding PAH sources in situations where coal-tar sealants were not an important source. The following passage from Van Metre and Mahler (2010) discusses this issue:

Page 342: "The shift in the 1960s and 70s to CT sealcoat as the dominant source of PAHs in seven of the eight lakes analyzed for trends (Fig.5) is in agreement with industry internet sites that report that CT sealcoat use began in the 1950s (Glacier Asphalt. 2010: JDK Pavement Maint. 2010). Large

contributions from coal to the two older urban lakes (HAR and NEW) in the middle of the 20th century were replaced by contributions from CT sealcoat in the 1960s (Fig.5); this is consistent with a report that, in 1940, 55% of American homes were heated with coal and another 23% used wood, but both uses "had virtually disappeared" by 1970 (U.S. Department of Commerce, 1997).

PCTC Comment (page 5, paragraph 4): "Fourth, other scientists at the USGS have conducted similar types of PAH research in the past and have outlined the steps necessary to go from a hypothesis to scientific proof. For example, in 2001-02, the USGS sought to determine the background levels and sources of PAHs in Chicago ambient surface soil. (Kay and others, 2003). In doing so, these USGS scientists went through great lengths to describe how sampling sites were randomly and statistically chosen. In the 40 Lake Paper, no such methodology was implemented. Indeed, only nine lakes were "selected" for the entire western portion of the United States, with merely three being in California, and all of them were located in the far southern part of the state. Just one lake was used to represent all of Oregon and two were somehow selected for Washington. Exactly how these six lakes were "statistically" chosen amongst the thousands of lakes that exist within these states is not described in the 40 Lakes Paper, nor was any effort made to identify what types of lakes were even to be included for consideration. For example, a natural lake can differ significantly from a man-made reservoir and a storm water detention pond in terms of age, size, depth, proximity to roads and function (i.e. recreation/fisheries versus designed trapping/filtering of urban run-off and contaminants). Incredibly, in concluding that "sealcoat has been the primary cause of increases in PAHs since the 1960s Drs. Van Metre and Mahler relied on data from only 8 lakes, two of which were from the Western United States. No one can seriously suggest that this type of analysis is statistically sound. And if it is not, then the limitations of the 40 Lake Paper should have been made public and the need for additional research should have been emphasized, not hidden."

USGS response: The request for correction seems to suggest that a random statistical survey is the only type of valid design for a scientific study. The methodology used in the studies by Kay et al. (2003) and Van Metre and Mahler (2010) are different because they had very different objectives. The objective of Kay and others was to characterize the concentrations of PAH compounds in ambient surface soils in the City of Chicago. A random statistical design was appropriate for this purpose. The lake sampling done by Van Metre and Mahler (2010) is part of the National Water Quality Assessment (NAWQA) Program's approach to assessing long-term changes in the concentrations of hydrophobic contaminants in the Nation's surface waters. The NAWQA Program is accomplished through investigations of some of the largest and most important watersheds and aquifers distributed across the Nation, rather than a random statistical design, because studies of individual watersheds contribute valuable information that assists in better understanding the major factors influencing water quality—a key goal of the NAWQA Program. The use of consistent design, sampling, and laboratory analytical methods in all 40 lakes provides the opportunity to gain national insights through the comparison of findings.

With regard to how the lakes were selected, a description of the lake selection process used by Van Metre and Mahler (2010) is on pages 335-336 of that publication: "Sediment cores were collected from 40 lakes (29 reservoirs and 11 natural lakes) in the United States during 1996 through 2008, age dated and analyzed chemically." Additional details were provided in Supplementary information Table S1 (which is online). "Urban areas for this study were selected on the basis of NAWQA Program study units, metropolitan statistical areas and ecoregions to represent a diversity of ecoregions where a majority of the United States cities and urban populations are located...Eight of the 40 lakes were selected for analysis of historical trends. Six of these lakes one lake in the northern part and one in the southern part of each of the western, central, and eastern regions of the U.S. were selected to represent the effects of rapid urbanization on PAH source loadings regionally, and two lakes were selected to represent the effects of historical changes in PAH source loadings in older urban areas in the north-central and northeastern U.S. Population densities for these lakes range from 223 to about 2900 people/km²."

PCTC Comment (pages 6 and 7, "Bias and Transparency"): A FOIA request related to coal tar sealant research was sent off to USGS on April 15, 2011. . . It should be noted that a virtually identical FOIA request was sent to the Minnesota Pollution Control Agency ("MPCA". This was done because it became apparent several years ago that Drs. Mahler and Van Metre had developed a relationship with a staff member of the MPCA who also shared their interest in banning coal tar sealants. The MPCA emails seem to confirm that a small group of government researchers began to communicate with one another on the issue of coal tar sealants several years ago and quickly began to share behind closed doors a mutual disdain towards anyone who questioned their beliefs. Such conduct, in and of itself, should be a warning flag."

USGS Response: First, as noted above, the FOIA request has been completed; additional questions and issues related to the FOIA should be addressed to Mr. Ken Skipper at (303) 236-5050 (x 230) or kskipper@usgs.gov.

Second, whereas USGS scientists provide scientific data and information to assist those who make management, regulatory and policy decisions, in order to maintain its objectivity, the agency does not make recommendations or support any particular management or regulatory position. The copies of correspondence and other documents authored by Drs. Van Metre and Mahler that were provided under the FOIA request in your March 15, 2013, correspondence to Judy Cearly (Exhibit H) do not express any personal or USGS interest in banning coal tar sealants.

Third, USGS scientists routinely communicate with other scientists that share in their research interests and with managers of other federal, state, and local agencies that are interested in their findings. These types of internal communications are entirely consistent with the mission of the agency, with good science practices, and are generally safeguarded from release (refer to SM 502.5 at http://www.usgs.gov/usgs-manual/500/502-5.html). The publications of USGS scientists follow standard FSP review and approval processes to ensure that findings are presented in an objective and unbiased manner and all of the publications authored by Drs. Van Metre and Mahler meet this standard.

USGS Response to Specific Requested Corrective Action and Comments in the May 31, 2013 Second Inquiry:

Requested Corrective Action (page 7): "Since the fish tumor photograph on the USGS Fact Sheet is inaccurate and misleading, it clearly does not meet USGS or OMB guidelines for information quality and thus must be removed. Likewise, any similar photographs used by USGS scientists as part of presentations to ban or evaluate RTS should also be eliminated from such materials for the same reasons. This includes any presentation to be given in the future by USGS scientists or any USGS

presentation currently available to the public on the internet such as Dr. Mahler's PowerPoint given at the 2010 meeting of the National Water Quality Monitoring Council, posted at

<u>http://acwi.gov/monitoring/conference/2010/C6/C6_Mahler.pdf</u>. Any failure to take such action will adversely affect those members of the PCTC who distribute or apply coal tar sealants since consumers and legislators who are being asked to consider the merits of proposed sealant bans are being misled by these photos."

USGS Response: Slide 4 of the PowerPoint presentation referred to in the request for correction poses the question: "Why be concerned about PAHs?" followed by the statement: "PAHs have adverse health effects for biota and humans." Two photographs follow the statement. The first by Jim Negus shows a healthy bullhead from Tennessee. The second photograph (by Alfred Pinkney) shows a brown bullhead collected from the Anacostia River in Maryland with apparent lip lesions. Whereas, as noted below, high concentrations of PAHs in sediments have been shown to induce these types of tumors in fish, it is not possible to determine the cause of *this* fish's tumors. Other contaminants, environmental factors, and/or biological/viral factors may also cause or contribute to tumor formation in brown bullhead catfish. Rather, the purpose of showing this photograph is to indicate the types of effects hypothesized to be caused by PAH exposure in this fish species. Slides are designed to support an oral presentation, thus must be taken in context. However, to make this clearer to those who only have access to the slides, we will add a caveat to any slides or similar uses of this figure used in future presentations.

PCTC Comment (page 1, "Information Requiring Correction – OVERVIEW"): "As part of an apparent effort to influence and elicit emotional responses from consumers, legislators and the press regarding the environmental impact of refined tar-based pavement sealer (RTS), the USGS and several of its scientists have repeatedly published and disseminated photographs of brown bullhead catfish with horrible looking skin and mouth tumors. A prime example of this strategy can be found within the above mentioned USGS 2011 "Fact Sheet" (2011-3010)."

PCTC Comment (pages 2-3): "To the contrary, U.S. Fish and Wildlife Service researchers, such as A.E. Pickney (referenced by the USGS in the [Fact Sheet] caption), have failed to observe a consistent relationship between PAH sediment contamination and the type of skin tumor portrayed in the photo. Thus, recent research to discover the cause of these catfish tumors has focused instead on other factors such as viruses."

"Perhaps most disturbing, there are biologists within the USGS who know about and have participated in Pickney's quest to find the actual cause of catfish skin tumors. Other USGS scientists while observing the prevalence of certain types of catfish tumors without addressing the issue of causation directly, have noted that "there are probably multiple causal factors for such external tumors". Despite these facts, a third group of USGS scientists, which includes Dr. Barbara Mahler and her husband, Dr. Peter Van Metre, have chosen to use catfish skin tumor pictures as "evidence" of what PAHs and RTS supposedly can do. Thus, the picture remains a part of the USGS Fact Sheet even now... Such actions clearly reflect a breach of USGS Guidelines for science quality and integrity."

USGS Response: The figure and caption referred to is from USGS Fact Sheet 2011-3010 "Coal-Tar Based Pavement Sealcoat, Polycyclic Aromatic Hydrocarbons (PAHs), and Environmental Health." which discusses studies by the USGS that have identified coal-tar-based sealcoat as a major source of

PAH contamination. The target audience for Fact Sheets is the general public, non-core professionals, and core professionals. The typical fact sheet is 4-6 pages and is focused on USGS science. Findings in fact sheets are from previously published studies.

The figure and caption in question are included in a section of the document titled: "What are the concerns? The purpose of the section is to provide the reader with information about the range of potential adverse effects of PAHs to aquatic life and humans. In addition to the potential impacts to Brown Bullheads, the section summarizes several other studies that have shown a relation between coaltar based pavement sealcoat and harmful effects to aquatic communities. The figure in question is from Pinkney et al. (2009), "Tumors in Brown Bullheads in the Chesapeake Bay Watershed: Analysis of Survey Data from 1992 through 2006". Since 1992, the U.S. Fish and Wildlife Service, Chesapeake Bay Field Office has conducted brown bullhead tumor surveys in Chesapeake Bay tributaries to assess the utility of liver and skin tumor prevalence as an environmental indicator.

The request for correction refers to the type of tumor shown in the photograph suggesting that because a more recent publication by Pinkney and others (2013) failed to show a consistent relationship between PAH concentrations in sediment and the incidence of skin tumors in brown bullheads that including a picture of these tumors in this document and in other presentations is misleading. First, the relation between PAH concentrations and the type of tumor is not discussed by the authors of the Fact Sheet. The major point of the figure (and its caption) is that elevated PAH concentrations in sediments are believed (by Pinkney et al.) to be related to the incidence of tumors in brown bullhead catfish in the Anacostia River. There have been numerous investigations of the relation between elevated PAH concentrations and tumor formation in fish that support Pinkney's conclusion. The strongest evidence is for the effects of PAHs and liver tumors [Baumann and Harshbarger (1998); Myers et al (2003), (2008); Vogelbein and Unger 2006; Hawkins et al. (1990); and Schiewe et al. (1991)]. For example, a linkage between PAHs and liver tumors in brown bullheads was established by Baumann and Harshbarger (1998) from surveys conducted in the 1980s and 1990s in the Black River, Ohio, USA. They observed that tumor prevalence increased and decreased consistently with similar changes in sediment PAH concentrations. Further evidence of an association between sediment PAHs and bullhead liver tumors was demonstrated by Pinkney et al. (2004a) in the Anacostia River (Washington, DC), using polycyclic aromatic compound (PAC)-DNA adducts as a biomarker of response to PAH exposure (Reichert et al., 1998). The evidence for a relation between PAHs and skin tumors is not as strong and recent work suggests that other environmental factors may be involved. Skin tumors in brown bullheads have been induced by repeatedly painting the skin with sediment extracts containing high PAH concentrations (Black et al., 1985). Baumann et al. (1996) reported that higher oral and cutaneous tumor prevalence occurred in PAHcontaminated Great Lakes tributaries compared with reference sites. Grizzle et al. (1984) observed an increased prevalence of papillomas in black bullhead (Ameiurus melas) exposed to chlorinated wastewater effluent, and prevalence decreased when chlorination was lowered. Poulet et al. (1994), however, noted the occurrence of orocutaneous tumors in 94 brown bullheads collected from 17 locations (both contaminated and uncontaminated) in New York state did not suggest a strong correlation with exposure to known chemical carcinogens. Pinkney et al. (2011) did not find any relationship between brown bullhead skin tumors and exposure to PAHs or alkylating agents in surveys of several Chesapeake Bay tributaries using DNA adducts as biomarkers. Bunton (2000) concluded that, although skin tumors in brown bullhead are associated with bottom dwelling, feeding, and contact with contaminated sediments,

other (unknown) factors may be involved. Oncogenic viruses, which induce skin tumors in other fish species (Getchell et al., 1998), have never been identified in brown bullhead (Poulet et al., 1993; Poulet and Spitsbergen, 1996). The possibility that other factors may also contribute to skin tumor formation in fish may affect the utility of using skin tumor surveys as an environmental indicator of contaminated bed sediment—a goal of Pinkney's research; but, it does not negate earlier findings showing that high concentrations of PAHs are a potential and important contributor. Nonetheless, the major point of the figure (and its caption) is that elevated PAH concentrations in sediments are believed to be related to the incidence of tumors in brown bullhead catfish. As indicated above, USGS will add a caveat to any slides or similar uses of this figure that we publish or present in the future, and will focus on the proven connection between PAHs and liver tumors in brown bullheads.

PCTC Comment (page 4, paragraph 1): "Another way to determine if any bias or advocacy exists within the USGS on the issue of coal tar sealants is for the USGS to produce all related data, correspondence and emails concerning its research regarding the 40 Lakes Paper and other similar articles. A FOIA request asking for such materials was sent off two years ago and incredibly, still remains "open" today. As will be demonstrated in greater detail below, the USGS has sought to minimize evidence of advocacy within its ranks by withholding, at least up to now, certain correspondence and email between the USGS staff and other individuals outside the agency who have made it their goal to ban coal tar sealants across the country. These efforts by USGS to withhold certain documents are not only at odds with the above mentioned policies, but also contrary to the need for transparency that is emphasized throughout USGS Guidelines and Manual."

USGS Response: This comment is repeated from page 3, paragraph 3 of the May 15, 2013 request. Please refer to the response provided above.

PCTC Comment (page 4, paragraph 3): "Various USGS scientists including Drs. Van Metre and Mahler, claim that contamination of urban lakes and streams by polycyclic aromatic hydrocarbons (hereinafter PAHs) is widespread in the U.S. This assertion is not particularly surprising since there is a consensus in the scientific community that PAHs have many potential sources, including vehicle emissions, tire particles, motor oil, crude oil, power plant emissions and industrial releases. Indeed, almost any type of combustion with organic matter will produce PAHs as a by-product, including natural sources such as forest fires and volcanoes down to something as basic as grilling on the backyard barbecue. Thus, one would expect PAHs to be ubiquitous in our environment and, in fact, they are."

USGS Response: This comment is repeated from page 4, paragraphs 1 and 2 of the May 15, 2013 request. Please refer to the USGS response provided above.

PCTC Comment (page 5, paragraph 1): "When it comes to RTS website postings and presentations, the USGS has disseminated photographs that graphically depict skin catfish tumors. Such an approach is understandable if the intent is to create a visceral reaction as part of an overall plan to scare the public into banning RTS. From a scientific perspective, such an approach is indefensible since the best evidence to date shows no significant relationship between sediment contamination of

any kind and catfish skin tumors. Furthermore, taking it one step further, absolutely no evidence has been cited by the USGS which demonstrates that RTS, as opposed to PAHs in general, has in fact caused any type of fish tumors—liver, skin, or otherwise."

USGS Response: As previously explained, photographs of catfish with tumors have been included in publications and presentations as an example of the harmful effects of high concentrations of PAHs in the environment to aquatic life. As noted earlier, the relation between PAH concentrations and the type of tumor is not discussed by Drs. Van Metre and Mahler. The major point of the figures (and its captions) is that elevated PAH concentrations in sediments are believed to be related to the incidence of tumors in brown bullhead catfish. As stated above, there is strong evidence from multiple scientists showing that high PAH concentrations in sediments contributes to the incidence of fish tumors. PCTC's suggestion that the best evidence to date shows no significant relationship between sediment contamination of any kind and catfish skin tumors is not accurate. As stated in the response to the PCTC comments (pages 1-2), although the evidence for a relation between PAHs and skin tumors is not as strong as for liver tumors, to suggest that there is no evidence for causality is not accurate (e.g., Black et al., 1985, and Baumann et al., 1996). Recent laboratory studies by Pinkney et al. (2013) showed that PAHs and another class of chemicals (alkylating agents such as nitrosamines) caused both liver and skin tumors. Pinkney did not observe a consistent relationship in the Anacostia and Potomac Rivers; but, establishing consistent relationships with individual contaminants in the environment is much more challenging than in the laboratory because of the multiple environmental factors that may be involved-for example, exposure of organisms to both present-use and legacy contaminants and their degradation products many of which we are not capable of measuring in the laboratory. PCTC is correct that showing a relation between PAH concentrations and fish tumors is not the same as showing a direct relation to RTS. However, the findings of Van Metre, Mahler, and others that: 1) RTS is currently one of the largest sources of PAHs to urban streams; and 2) high PAH concentrations in sediment have been associated with harmful effects to aquatic life, including increased incidence of tumor formation in fish, provide a reasoned basis for resource managers to make the linkage and take appropriate action.

PCTC Comment (page 6, paragraph 3): "The MPCA emails seem to confirm that a small group of government researchers began to communicate with one another on the issue of coal tar sealants several years ago and quickly began to share behind closed doors a mutual disdain towards anyone who questioned their beliefs. Such conduct, in and of itself, should be a warning flag."

USGS Response: This comment is repeated from page 7, paragraph 1 of the May 15, 2013 request. Please refer to the USGS response provided above. See PCTC Comment (pages 6 and 7, "Bias and Transparency").

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