Review of
"Coal-Tar-Based Parking Lot
Sealcoat: An Unrecognized Source of
PAH to Settled House Dust" by
Mahler et al., published in
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Review of Mahler et al. (2010) Study Design

The Mahler et al. (2010) study describes an evaluation of polycyclic aromatic hydrocarbons (PAHs) in indoor and outdoor dust collected from apartments and their associated parking lots. Of 23 apartments tested, Mahler et al. (2010) determined that 11 had asphalt parking lots with coal-tar-based sealant and the remaining 12 had concrete or asphalt parking lots that were unsealed or coated with an asphalt-based sealant. Coal-tar-based sealcoated parking lots are referred to as "CT" while non-coal-tar-sealcoated parking lots are referred to as "NCT."

Mahler et al. (2010) found median total PAH concentrations of 4,760 µg/g and 9.0 µg/g in dust collected from CT and NCT parking lots, respectively. Median total PAH concentrations of 129 µg/g and 5.1 µg/g are reported for indoor dust collected from CT and NCT apartments, respectively. The presence or absence of CT parking lots was reported to explain 48 percent of the variance in log-transformed total PAH concentrations in indoor dust. Other factors including land use, frequency of vacuuming, indoor burning, and more also were evaluated. Of these other factors, Mahler et al. (2010) report that only urban land use intensity near the sampled apartment has a significant relationship with total PAH concentrations.

General comments regarding study design are listed here, followed by additional discussion in subsequent sections:

- Lack of precision in selection of sample locations contributes to variability between the sampled areas and consequently, uncertainty regarding external influences when evaluating the results

- Small sample size (especially given lack of precision in sample location selection)

- Particle size fraction evaluated not appropriate for dermal and ingestion exposures

- Dust loading not evaluated

- Incomplete evaluation of independent variables

Analytical Methods

The analytical method used is considered appropriate for analysis of PAH concentrations; although, some analytical difficulties were encountered, preventing quantitation of dibenz[a,h]anthracene in all but one sample. A summary of quality assurance/quality control (QA/QC) data is provided in the Supplementary Information; however, the raw QA/QC data are not presented. This information would be required for a proper evaluation of data quality. For example, the authors report that individual PAH compounds were detected in blank samples more than 20% of the time, but no information is presented to identify which samples were associated with contaminated blank samples.

Sampling Methodology

Mahler et al. (2010) collected 23 indoor and outdoor dust samples between April and July 2008. No detailed information is provided regarding how sample locations were selected other than presence or absence of CT parking lots. Due to a lack of site selection or exclusion criteria other than presence or absence of CT parking lots, other potential factors affecting PAH concentrations in parking lot dust may have been overlooked or unaccounted for. For example, little to no information is presented to support the classification of CT parking lots, parking lot

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selection criteria, or sample location selection criteria – all of which can affect the variability of the data. Site selection appears to have been based solely on "coffee/tea" field screening tests. The "coffee/tea" test is not a standard, validated method so its accuracy in identifying CT parking lots is uncertain.

There is no indication that interviews with apartment maintenance staff and owners and/or review of maintenance records were conducted. Such interviews and records review could not only confirm the use of coal-tar-based sealant, but also provide useful information on lapsed time since sealcoat application, frequency of application, application formulation, and other maintenance history of potential relevance. This information would help to confirm the presence or absence of coal-tar-based sealant as well as optimize uniformity of the sample locations by selecting those with the most similar application and maintenance history.

Also, no criteria are provided for selection of specific sample locations within each parking lot other than avoidance of painted areas and drip lines. The number of days since last rainfall or washing event, traffic and runoff patterns, number of parking lot stalls/cars, and the location of stains, cracks, and debris in each parking lot would help guide when and where to collect samples at each lot. Again, use of this information to inform site selection and timing of sample collection would reduce uncertainties associated with comparability of CT and NCT data. The timing of sample collection with respect to rainfall and washing events is particularly important given the extended, three-month duration of the sample collection period. More precise timing of the sampling event would reduce bias introduced due to sampling during variable weather conditions. For example, little to no rainfall was recorded between February and April of the year the sampling was conducted. This could have resulted in an accumulation of dust at the beginning of the sampling program. In April, heavier and more frequent rainfall was experienced which then decreased in volume and frequency each successive month over the course of the sampling event until its completion in July. Again, depending on the timing of the sample collection, the changing weather could have introduced variability in the dust data.

Similarly, no criteria are provided for selection of apartments other than presence or absence of CT parking lots. Additional criteria such as apartment age, flooring type and age, and period of time occupied by current owner could be used to obtain as uniform a sample population as possible and thereby improve comparability between samples. This is especially useful for small sample sizes where the influence of variable apartment and flooring characteristics as well as influence of previous owners (if newly occupied) will have a greater effect than in larger sample sizes.

The Supplementary Information indicates that the apartment complex build dates range from 1961 to 2007 with a median date of 1978. This indicates that about half of the apartments were more than 30 years old. Although no information is presented to determine the relative age of CT apartments compared to NCT apartments; certain statements in the paper suggest that NCT apartments are much newer. For example, in the second paragraph of the discussion section, the authors explain differences in dust concentrations from parking lots with asphalt-based sealcoat measured in this study with levels detected in a previous study by implying the lots in this study were newer.

> "The difference likely is because the asphalt-based sealcoat on the lots tested by [another study] had been applied over worn coal-tar sealcoat, whereas the asphalt-based sealcoat on the parking lots tested for this study had been applied over new asphalt pavement."

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The presence of new asphalt pavement suggests that the associated apartments are also newer. Also, later in the paper when the authors are discussing potential contributions from coal-tar based flooring adhesives[1], the following statement is made:

> "Of the four other NCT apartments in this study with wood laminate or linoleum flooring, only one was built prior to 2000 and it did not have elevated T-PAH concentrations."

Thus, it appears that the NCT apartments represent newer housing stock compared to CT apartments. To the extent that older apartments (and carpets) reflect longer-term accumulation of PAHs, if for example the apartment is located nearby a heavily travelled roadway, then apartment age may be a significant variable that has not been evaluated.

Field replicate samples were collected at two indoor locations and one outdoor location. These samples were used as measures of sample variability. However, it appears that no field rinsate samples were collected as part of QA/QC procedures. Given the elevated levels of PAHs observed at CT parking lots, it would be useful to evaluate the decontamination process by collecting equipment rinsate samples to verify that the HVS3 was adequately decontaminated between samples. This is a legitimate concern, particularly given the frequency of detecting PAHs in the laboratory method blanks. Further, it is always prudent to first collect samples assumed to have lower levels of contamination and then collect samples assumed to have higher levels of contamination, to minimize cross-contamination of field equipment. Dedicated indoor and outdoor vacuums could be used to first sample CT locations then sample NCT locations, thereby reducing the potential for contamination between CT and NCT locations as well as indoor and outdoor locations. Because a standard operating procedure was not provided in the Supplementary Information, it is not known whether or not measures were employed to reduce cross-contamination of samples. At a minimum, it would be advisable to collect NCT samples first, along with an equipment rinsate sample, followed by collection of CT samples and another equipment rinsate sample to minimize the potential for cross-contamination of samples and determine if cross-contamination was an issue.

The high-efficiency vacuum sampler recommended in ASTM Method D 5438 (2005) was stated to have been utilized according to the manufacturer operation manual (CS_3 Inc. 2004). Mahler et al. (2010) state that a sample was collected from an entry way and adjacent living room floors. In the absence of child residents, sample locations recommended by EPA (2008) for assessing lead in indoor dust include the 1) entryway, 2) bed room, and 3) other room most often occupied by the residents. While Mahler et al. (2010) collected a composite sample, EPA (2008) guidance recommends collection of discrete samples within the targeted areas of the residence so that a weighted average dust concentration can be calculated based on the fraction of the day that the resident spends in each area. In this way, areas with low dust loading are not combined with those with higher dust loading and as discussed below, exposure is related to dust loading.

Dust load is expected to be highest at the entryway and in carpets; however, PAH concentrations are expected to be highest at the entryway and lowest in carpets. Since most time will be spent in the living areas rather than at the entryway, composite samples that combine both areas do not represent average exposure concentrations. The composite

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[1] The use of tar-based flooring adhesives in Germany is reported by Heudorf and Angerer (2001); however, it is not clear that these products were or are available for use in the United States.

concentrations will overestimate the average exposure concentration both due to higher concentration and due to higher loading.

Mahler et al. (2010) note the range in area sampled among apartments (1.6 to 13 square meters indoors, 2.0 to 7.5 square meters outdoors) but the rationale for this variability is not provided. Presumably, the range in sampled area is due to differences in floor type and loading, i.e., larger areas were sampled as necessary to obtain adequate sample size from bare floors or relatively cleaner floors. However, the relative contribution from different areas within apartments could bias the PAH concentrations high or low, depending on the sampled location and loading at that location. Of particular concern is that there may have been consistent over-representation of high concentration dust from the entryway due to heavy dust loads in that area. A discussion of the dust loading levels at sampled locations would be helpful in understanding the concentration data and perhaps explaining the need for the range in sampled area.

Prior to analysis, Mahler et al. (2010) sieved samples to obtain the size fraction less than 500 μm diameter. For the purpose of estimating potential exposures to dusts, EPA (2004, 2008), CS_3 Inc (2004), and ASTM Method D 5438 (2005) recommend obtaining the size fraction that is most likely to adhere to skin surfaces. EPA (2004, 2008) recommends sieving dust samples and analyzing the portion smaller than 250 μm, while CS_3 Inc. (2004) and ASTM (2005) recommend analyzing the size fraction less than 150 μm in diameter. The size fraction obtained by Mahler et al. (2010) does not represent the particle size most likely to adhere to skin surface and may not provide a realistic estimate of exposure material for dermal absorption and ingestion of dust via hand-to-mouth activity. Typically, contaminant concentrations become enriched as particle size decreases (Lewis et al. 1999). Depending on the particle sizes that best represent sloughed parking lot sealant, the influence of particle size in this dataset is uncertain.

Statistical Approach

Given only the PAH analytical data supplied in the Supplementary Information, ENVIRON was able to verify the summary statistics, but could not verify the influence of independent variables reported in the paper. If made available, the additional information obtained from the study participants and characteristics of the sample locations could be used to confirm the significance of parking lot surface type and land use intensity in explaining the variability of PAH concentrations in dust samples, as well as the lack of significance associated with smoking and distance to the nearest roadway.

Additional variables that should be considered but were not reported in Mahler et al. (2010) include apartment and carpet age and degree of sealcoat wear. The age of the apartment and carpet could be important variables explaining differences in indoor dust concentrations. As previously indicated, there is summary information on the apartment age variable provided in the Supplementary Information; however, this variable is not included in Table 1 of the paper, indicating that it was not evaluated as an independent variable.

In contrast, the degree of sealcoat wear is listed in Table 1 of Mahler et al. (2010) as an independent variable potentially related to the levels of PAH detected in indoor dust and parking lot dust samples. However, there is no information presented as to how this wear was evaluated, nor is there any information in the supporting material that summarizes the range of wear levels for the parking lots examined in the study. If parking lot surface type is believed to be a significant factor in explaining indoor and parking lot dust PAH levels, then one might expect that the degree of sealcoat wear should also be a factor.

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Other factors such as the size of the apartment complex or size of the associated parking lot might also be expected to be factor in determining PAH levels in indoor dust, but these data are not presented.

ENVIRON evaluated PAH analytical data available from the Supplementary Information in an attempt to identify patterns in PAHs detected in the CT and NCT samples. No obvious distinction between CT and NCT samples could be discerned from the available data; however, the analytical method used for this study does not allow for a more comprehensive evaluation that could be used to identify unique patterns in the dataset that are specific to the source of the PAHs. From ENVIRON's limited review, observations appear to most closely resemble what would be considered an "urban background" profile as described by Stout et al. (2004).

Metrics for Evaluating Dust Exposure

The variable "days since sampling area last vacuumed" is listed in Table 1 of Mahler et al. (2010) as a variable potentially associated with the level of PAHs in indoor dust. Presumably, this metric is meant to provide a measure of cleanliness in the home. However, a better metric is the actual dust loading in the areas sampled. The information required to calculate dust loadings was collected (i.e. summary information for mass of dust and area sampled is provided); however, these data are not reported and not evaluated.

Both PAH concentration and dust loading from each living area are needed to assess exposures. While PAH concentration is useful in providing the amount of PAH in dust, it does not provide information about the amount of dust that is available on an exposure area or surface. For example, a high concentration of a contaminant in house dust may present a low risk if dust loading is low, or conversely, a low contaminant dust concentration may present a high risk if dust loading is quite high (WTC 2005). Concentration of chemicals in dust, alone, is not adequate for predicting risk.

In most house dust studies involving lead, blood lead levels have been shown to correlate either most closely with lead loading or equally with both lead concentration and lead loading (see review by Adgate et al. 1995). The different results are influenced by a number of factors, including study design, additional sources of lead aside from dust, behavior patterns, bioavailability of the lead in the exposure matrix, and more. For this reason, EPA (2008), ASTM (2005), and CS_3 Inc (2004) recommend evaluating both concentration and loading metrics when evaluating exposures to dust. In this way, the data can be evaluated to obtain the best possible understanding of the chemicals present in dust and potential for exposure of residents.

There also are studies demonstrating that the presence of chemicals in dust have little or no correlation with chemicals measured in humans. For example, Heudorf and Angerer (2001) found no correlation between PAHs measured in house dust and urinary metabolites of PAHs.

University of Michigan researchers studying dioxins in people living in an area contaminated by a manufacturing facility did not find an association between dioxins in house dust / soil and blood dioxin levels (Garabrant et al. 2009). Even though greater concentrations of dioxins were measured in the soil and house dust of homes within the contaminated area compared to a reference area, the primary factors associated with blood dioxin levels were age, history of working at the facility, and fishing and hunting in waterways within the contaminated area (Garabrant et al. 2009). The results of this study demonstrate that many factors must be evaluated to understand exposure to environmental contaminants and no one factor is likely to be responsible for total body burden, given multiple sources and pathways of exposure.

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Similar results were obtained in an exposure study of a population in Calcasieu Parish, Louisiana. The Agency for Toxic Substances and Disease Registry (ATSDR) found elevated blood dioxin levels in older residents only and these levels were found to be associated with historical exposures rather than levels measured in their homes (ATSDR 2006). Although these results relate to dioxins rather than PAHs, they remind us that collection of exposure information is important in helping us to understand potential exposure scenarios. In some cases, measured chemical concentrations in our homes are not always correlated with levels of chemicals measured in our bodies. This is particularly likely for chemicals such as PAHs and dioxins which have other pervasive sources of exposure such as diet.

Given the range in indoor areas sampled by Mahler et al. (2010), it could be assumed that dust loading varied substantially among the residences sampled. Evaluation of loading by residence then may provide insight into potential exposures to PAHs in house dust or potential for tracking parking lot dust into homes. However, the presence of PAHs in indoor dust measured via concentration and/or loading does not necessarily equate to risks to residents.

PAH Toxicity

PAHs are a class of compounds consisting of two or more bonded aromatic rings, excluding those compounds with anything other than hydrogen or carbon atoms. PAHs are formed during incomplete combustion of organic materials such as gas, wood, oil, garbage, cigarettes, and grilled or charbroiled foods. Although there are over 100 PAHs, a subset of 16 are routinely evaluated using standard analytical methods. Of these PAHs, seven have been classified as probable human carcinogens (Group B2) (1993; see EPA's Integrated Risk Information System 2010): benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, dibenz(a,h)anthracene, indeno(1,2,3-cd)pyrene, and chrysene. The seven carcinogenic PAHs are referred to here as cPAHs.

Although some chemical mixtures (e.g., tobacco smoke, chimney soot, others) that include PAHs have been shown to be carcinogenic by inhalation or dermal contact in humans, the relative potency of the individual PAH compounds has not been established in humans. The EPA toxicity assessment for benzo(a)pyrene and the other cPAHs has been based on the results of studies in rodents; however, these chemicals have only been shown to be rodent carcinogens at portal of entry sites such as skin or lung.

To quantify the carcinogenicity of the cPAHs, a relative potency of carcinogenicity was assigned to each cPAH with benzo(a)pyrene used as the reference compound (EPA 1993). Benzo(a)pyrene is the most well-studied of the cPAHs (EPA 1993). The carcinogenic potency of each cPAH was estimated relative to benzo(a)pyrene based primarily on comparison of mouse skin tumor data. While skin tumor data from mice for multiple PAHs may allow a comparison across PAHs, the relevance and predictiveness of this test system for oral cancer risk in humans is questionable. Consequently, there is a high degree of uncertainty associated with cancer risk estimates for the cPAHs.

EPA (1993) recommends the following relative potencies for cPAHs[2]:

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[2] EPA is currently accepting public comments on revised potency factors provided in an external review draft report published in February 2010. This revised guidance includes additional cPAHs and includes revised potency factors for many of the seven currently listed cPAHs; however, the limitations described

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cPAH	Relative potency factors
benzo(a)pyrene	1.0
benzo(a)anthracene	0.1
benzo(b)fluoranthene	0.1
benzo(k)fluoranthene	0.01
dibenz(a,h)anthracene	1
indeno(1,2,3-cd)pyrene	0.1
Chrysene	0.001

Table 2 in Mahler et al. (2010) lists analytical results separately as the sum of total PAHs (16 PAH compounds), benzo(a)pyrene, and sum of cPAHs. However, the cPAH concentrations have not been modified by their relative potencies to benzo(a)pyrene. This means that the cPAH concentrations provided in Table 2 are not equivalencies of benzo(a)pyrene and should not be compared to health- or risk-based values that are based on equivalent concentrations of benzo(a)pyrene. Evaluating the sum of bulk cPAHs without adjustment for relative potency to benzo(a)pyrene results in an assumption that all cPAHs are as potent as benzo(a)pyrene when in fact, that is not the case (EPA 1993).

When evaluating the indoor and outdoor dust concentrations, it is useful to adjust the cPAH concentrations by their relative potencies to better understand the potential risks associated with cPAHs in dust. Concentrations of cPAHs provided by Mahler et al. (2010; see Supplementary Information) were modified by their respective potency factors to obtain total cPAH concentrations presented as a benzo(a)pyrene equivalent concentrations, or benzo(a)pyrene equivalents, for each sample using the following equation:

$$BaPE = \sum_{i=cPAH}^{n=7} C_i \times RPF_i$$

Where:

BaPE =   Concentration of cPAHs as benzo(a)pyrene equivalent

C_i =   Concentration of individual cPAH

RPF_i =   Relative potency factor for each respective cPAH

i =   Each of 7 individual cPAHs

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in the text related to uncertainty associated with extrapolating from rodent portal of entry data to human oral cancer risk have not been addressed by EPA.

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The benzo(a)pyrene equivalent concentrations calculated for samples collected by Mahler et al. (2010) are provided in the following table.

cPAH concentrations as benzo(a)pyrene equivalents (BaPE) in indoor dust.		
Parking Lot Status	Sum of bulk cPAHs (µg/g)	BaPE (µg/g)
CT	22	4.5
CT	94	20
CT	70	15
CT	29	5.7
CT	104	20
CT	8.6	1.7
CT	18	2.3
CT	54	10
CT	30	6.1
CT	47	6.8
CT	156	32
NCT	0.99	0.21
NCT	9.4	1.9
NCT	22	5.0
NCT	3.2	0.75
NCT	8.5	2.0
NCT	12	2.8
NCT	86	17
NCT	0.47	0.085
NCT	1.8	0.36
NCT	1.7	0.31
NCT	2.3	0.63
NCT	1.6	0.34

Regulatory risk assessments rely on the use of benzo(a)pyrene equivalent concentrations to estimate potential risk from exposure to cPAHs. Although it is preferable to evaluate whole mixtures (refined coal-tar-based sealant) as opposed to individual components (cPAHs as benzo(a)pyrene equivalents), the lack of toxicity information specific to whole mixtures led to the development of relative potency methods. This approach does not take into account interactions between individual components of chemical mixtures and also necessitates the use of assumptions about the toxicity of individual compounds relative to a reference compound. In the absence of an alternative method, use of benzo(a)pyrene equivalent concentrations is the accepted method for evaluating risks associated with exposure to cPAHs.

Risk assessment is a tool used to evaluate and manage potential risks from exposure to chemicals. This tool combines assumptions about reasonable maximum exposures of a population (e.g., contact rates, bioaccessibility, duration of exposure, body weight, etc) with measured or modeled data for exposure media (e.g., chemical concentrations in soil, dust, air, water, food, etc) to obtain an estimate of a daily or lifetime intake level for a population. Next,

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this intake level is combined with a quantitative estimate of a chemical's toxicity to obtain an estimated cancer risk or non-cancer hazard. To be protective of more sensitive members of the population, this risk assessment model is intended to overestimate risks rather than underestimate risks.

EPA's National Oil and Hazardous Substances Pollution Contingency Plan guidance (March 8,1990; 40 CFR 300) identifies estimated cancer risks falling between 1×10^{-6} and 1×10^{-4} (or, one additional cancer case per million people and one additional cancer case per ten-thousand people) as within an acceptable risk management range. At sites where cancer risks exceed 1×10^{-4}, a remedial action is considered. In some cases, remedial action may be determined to be unnecessary when risks are slightly greater than 1×10^{-4} or considered necessary when risks are less than 1×10^{-4} (EPA 1991). In this way, risk assessment informs the remedial investigation process and can also be used to evaluate the effectiveness of remediation alternatives if cleanup is warranted.

Risk assessment is not, however, a tool used to predict the incidence of cancer or non-cancer health effects. Exceedance of a risk management guideline, including risk-based screening levels, does not indicate that exposure-related illness will occur. Instead, exceedance of the guidelines indicates that further investigation may be necessary to confirm that appropriate assumptions were incorporated into the risk analysis and/or indicates that cleanup may be recommended.

Comparison to Health-based Standards

As noted by Mahler et al. (2010), there is no regulatory standard for PAHs in indoor or outdoor dust. For lack of a criterion to evaluate the dust data, Mahler et al. (2010) relied on a German Federal Environmental Agency (FEA) value of 10 µg/g for benzo(a)pyrene, established by their Commission for Indoor Air Quality. As discussed below, the FEA value is not a health-based criterion. Consequently, ENVIRON evaluated the applicability of a health-based criterion developed by the World Trade Center (WTC) Indoor Air Task Force Working Group (2003) for cPAHs. The criteria established by the Germany FEA and WTC working group are discussed below.

German FEA Standard

Mahler et al. (2010) rely on a German FEA action level of 10 µg/g for benzo(a)pyrene in household dust. This level was developed in response to concerns regarding coal-tar parquet glues commonly used in homes built in the 1950's and 1960's. In Heudorf and Angerer (2001), 1,213 residents of 511 homes were recruited to evaluate coal-tar in flooring glue. Following analysis of PAH metabolites in urine and benzo(a)pyrene in indoor dust and parquet floor glue, no relationship was observed between levels of PAHs in urine and dust or glue. Also, no difference in PAH levels was observed between homes with and without the suspect parquet flooring. Based on these results, it was not possible for the German FEA to develop an exposure-based limit for PAH contamination in parquet flue and house dust. Heudorf and Angerer (2001) states that the German FEA could not define a threshold limit value below which there would be no risk to residents contacting PAHs in coal-tar-based parquet glue. Instead, the FEA selected the value of 10 µg/g as the maximum limit of benzo(a)pyrene in house dust in an attempt to minimize exposure of residents. Heudorf and Angerer (2001) state this limit applies to benzo(a)pyrene but do not discuss comparison of all cPAHs to this limit. In Mahler et al. (2010), benzo(a)pyrene levels in indoor dust at 4 of 11 CT locations and 1 of 12 NCT locations exceed the German FEA action level of 10 µg/g.

This action level is not a health-based value and is not useful in gaining an understanding of potential health risks associated with exposure to the PAH concentrations measured by Mahler et al. (2010). Based on the results presented by Heudorf and Angerer (2001), exceedance of the FEA value does not provide information about residential exposure or risk levels. For this reason, ENVIRON considered an alternate screening value based on standard risk and exposure assessment assumptions.

World Trade Center Criterion

Multiple federal, state, and local agencies collaborated on development of indoor air and dust screening criteria for chemicals of potential concern, including cPAHs, in an effort to assess environmental health conditions of residences in the vicinity of the collapsed World Trade Center buildings (see WTC 2003). The EPA-led effort resulted in development of peer-reviewed, health-based criteria which were used to support cleanup efforts at residences and other buildings where occupants were assumed to have long-term exposure to pollutants generated during the collapse of the World Trade Center towers.

For development of the cPAH loading criterion for settled dust, the WTC working group relied largely on EPA's *Policy Number 12 on Recommended Revisions to the Standard Operating Procedures (SOPs) for Residential Exposure Assessments* (2001) and EPA's *Risk Assessment Guidance for Superfund Volume 1: Human Health Evaluation Manual, Part E, Supplemental Guidance for Dermal Risk Assessment* (2004). The health-based criterion is based on the toxicity of cPAHs relative to benzo(a)pyrene and assumes exposure via both ingestion and dermal exposure pathways for an individual from age 1 through 31 years.

The WTC criterion for cPAHs was calculated using the following equation,

$$CancerRisk = LADD \times CSF$$

Where,

Cancer risk =   target cancer risk of 1 x 10^{-4}

LADD =   lifetime average daily dose (g/kg-day)

CSF =   cancer slope factor for benzo(a)pyrene (7.3 kg-day/g)

The LADD was calculated as the sum of the potential dose rates (PDRs) for the dermal and ingestion exposure pathways averaged over 70 years. The health-based loading criterion was developed by adjusting the LADD iteratively until the target cancer risk level of 1 x 10^{-4} was reached.

For dermal contact with settled dust, the WTC criterion includes a number of factors to estimate the PDR, including a measure of the contaminant surface load (CSL) and fraction transferred from surface to skin (FTSS), transferrable residue on indoor surfaces (CSL * FTSS), a transfer coefficient (TC), exposure time (ET), and body weight (BW), as shown in the following equation:

$$PDR = \frac{\left(TC \times ET_{hard} \times FTSS_{hard} \times CSL_{hard}\right) + \left(TC \times ET_{soft} \times FTSS_{soft} \times CSL_{soft}\right)}{BW}$$

Where,

PDR =   potential dose rate (g/kg-day)

TC =   transfer coefficient (cm^2/hr)

ET =   exposure time for hard and soft surfaces (hr/d)

FTSS =   fraction transferred from hard or soft surfaces to skin (unitless fraction)

CSL =   contaminant surface load on hard or soft surfaces (g/cm^2)

BW =   body weight (kg)

The transfer coefficient (TC) represents the rate of skin contact with a surface and is based on several assumptions including a contaminant surface load (CSL) value of 50 g/cm^2 for dust loading on typical indoor residential surfaces (obtained from Rodes et al. 2001), as well as skin surface area of 5,000 cm^2 for children and 9,000 cm^2 for adults. Exposure time (ET) was assumed to be 8 hrs/day for carpets (i.e., soft surfaces) and 4 hrs/day for hard floors (i.e., hard surfaces) for children age 0 to 6 years and adults over the age of 18 years. For adolescents age 6 to 18 years, ET was assumed to be lower, 6 hrs/day for carpets and 2 hrs/day for hard surfaces, due to time spent away from home while at school.

The fraction of dust that can be transferred from hard or soft surfaces to skin (FTSS) is based on hand press experiments using lipophilic compounds conducted by Rodes et al. (2001). The values of 0.10 for soft surfaces and 0.50 for hard surfaces from Rodes et al. (2001) were modified to account for body parts that have less intensive contact with indoor surfaces than hands (e.g., arms, legs, face), resulting in FTSS values of 0.05 and 0.25 for soft and hard surfaces, respectively.

The body weight assumed for children (15 kg) and adults (71.8 kg) are based on a compilation of national data provided by EPA (1997).

Once the dermal PDR was calculated, the product then was multiplied by a factor of 0.13 to account for the dermal absorbed fraction of cPAHs.

The WTC criterion also takes into account ingestion of dust via hand-to-mouth contact. Several assumptions for the ingestion PDR are similar to those used to estimate the dermal PDR, as seen in the following equation:

$$PDR = \frac{\left[\left(ET_{hard} \times FTSS_{hard} \times CSL_{hard}\right) + \left(ET_{soft} \times FTSS_{soft} \times CSL_{soft}\right)\right] \times SA \times FQ \times SE}{BW}$$

Where the unique input parameters are,

SA =   skin surface area (cm^2/event)

FQ =   frequency of hand-to-mouth events (events/hr)

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SE =   saliva extraction factor (unitless fraction)

Skin surface area (SA) is based on the area of three fingers only, and was assumed to be 15 cm^2 for children and 45 cm^2 for adults. These values were extrapolated from data provided by EPA (1997). The frequency of hand-to-mouth events (FQ) was extrapolated from a study by Michaud et al. (1994) for four age groups as follows: 1 to 6 yrs, 9.5 events/hr; 7 to 12 yrs, 5 events/hr; 8 to 18 yrs, 2 events/hr; and 19 – 31 yrs, 1 event/hr. A default value of 0.50 from EPA's Office of Pesticide Protection (2001) was selected to represent the fraction of dust transferred from the skin to the mouth (SE).

The values for FTSS for the ingestion exposure pathway were obtained directly from Rodes et al. (2001), 0.10 and 0.50 for soft and hard surfaces, respectively. These values were measured during hand press experiments using lipophilic compounds and dry skin.

One unique aspect of the WTC working group health-based criterion is the assumption that the source of contaminants present as a result of the collapse of the WTC towers is not an infinite source. In other words, it was assumed that regular cleaning of the residences and other occupied buildings would diminish WTC-related contaminants over the assumed 30-year exposure time-frame. Following a review of a number of studies on dissipation of contaminants in indoor dust, it was assumed that the half-life of WTC-related contaminants would be 22 months (resulting in a decay rate constant of 0.38 per year). To account for this, the CSL variable was modified according to:

$$CSL = CSL_{initial} \times e^{-kt}$$

Where "k" is the dissipation rate constant of 0.38 per year and "t" is the time (years) over which the exposure is expected to occur. Assuming a finite source, the WTC criterion for cPAHs is 150 µg/m^2.

EPA was consulted on adjustment of the WTC criterion for cPAHs to eliminate the dissipation rate constant. When assuming an infinite source (i.e., coal-tar sealant is continuously maintained on parking lots throughout exposure duration), the criterion for cPAHs is adjusted downward to 34 µg/m^2 (Maddaloni, personal communication 2010).

Comparison of Dust Data to WTC Criterion

The WTC health-based criterion of 34 µg/m^2 (modified for an infinite source) is considered relevant to residential indoor dust evaluations in other areas because it was intended for residential settings, takes into account both dermal and ingestion exposure pathways, assumes a 30-year exposure time-frame spanning child, adolescent, and adult life stages, is based on standard EPA exposure and risk assessment methodology, utilizes the current recommended cancer slope factor for benzo(a)pyrene, and assumes exposure to all seven cPAHs.

Mahler et al. (2010) indicate that the mass of dust was weighed both before and after sieving but these data were not provided. With dust loading data, a residence-specific dust loading value for cPAHs could be derived according to:

$$\text{Residence} - \text{specific cPAH Loading Level}\left(\frac{\mu g}{m^2}\right) = Loading\left(\frac{g}{m^2}\right) \times Concentration\left(\frac{\mu g}{g}\right)$$

In the absence of the residence-specific dust loading data from Mahler et al. (2010), it can be assumed that the dust loading level in the sampled apartments is 0.5 g/m^2 (WTC 2003). This level of dust loading selected for development of WTC dust screening criteria is consistent with the geometric mean loading of 0.42 g/m^2 for bare floors reported by Adgate et al. (1995), 1.3 g/m^2 for carpets before cleaning and 0.1 g/m^2 for carpets after cleaning reported by Roberts et al. (1999), and range of 0.05 to 7 g/m^2 for bare floors reported by Lioy et al. (2002). A thorough discussion of dust loading levels is provided in the WTC criteria development document (2003).

The WTC criterion of 34 µg/m^2 is derived using the cancer slope factor for benzo(a)pyrene. As discussed previously, when comparing cPAH concentrations to health-based standards, cPAH concentrations must be adjusted by their relative potency factor. Concentrations of cPAHs as benzo(a)pyrene equivalents were used in the equation listed above to calculate residence-specific cPAH loading levels. The cPAH loading levels then can be compared to the WTC criterion of 34 µg/m^2.

cPAH dust loading levels (as benzo(a)pyrene equivalent concentrations) assuming dust loading level of 0.5 g/m^2.		
Parking Lot Status	Sum of cPAH as BaPE (µg/g)	cPAH (as BaPE) Loading (µg/m^2)
CT	4.5	2.3
CT	20	10
CT	15	7.3
CT	5.7	2.8
CT	20	9.9
CT	1.7	0.84
CT	2.3	1.2
CT	10	5.0
CT	6.1	3.0
CT	6.8	3.4
CT	32	16
NCT	0.21	0.10
NCT	1.9	0.93
NCT	5.0	2.5
NCT	0.75	0.38
NCT	2.0	1.0
NCT	2.8	1.4
NCT	17	8.4
NCT	0.085	0.042
NCT	0.36	0.18
NCT	0.31	0.16
NCT	0.63	0.32
NCT	0.34	0.17

As shown above, the maximum cPAH loading level of 16 µg/m^2 is less than half the WTC health-based criterion of 34 µg/m^2. The median cPAH indoor dust loading level for an apartment with a coal-tar sealed parking lot is 3.4 µg/m^2, which is an order of magnitude lower than the WTC criterion. Although indoor dust cPAH concentrations are greater in CT apartments, the

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levels measured by Mahler et al. (2010) are well below health-based standards derived in accordance with WTC methodology.

It should be noted that exceedance of the WTC criterion would not suggest that adverse health effects would be experienced by the resident. Instead, exceedance would indicate that further study of the home may be necessary to better understand PAH sources in the home, exposure pathways, and perhaps biomonitoring to determine whether an exposure is occurring followed by abatement if further investigation indicates that a potential for risk to the resident is apparent.

Mahler et al. (2010) provide a comparison of indoor dust cPAH concentrations from CT sample locations to a concentration of 40 µg/g cPAHs (bulk concentration) provided by Maertens et al. (2008) that is equivalent to a 1×10^{-4} cancer risk level. Maertens et al. (2008) found that ingestion of 0.1 g/day of house dust by children ages 0 to 5 years results in less than 1×10^{-4} cancer risk for cPAH dust concentrations less than 40 µg/g. While seven indoor dust samples exceed this value (six CT locations, one NCT location), it is important to note that the exposure model described by Maertens et al. (2008) is not as sophisticated as that developed for the WTC criterion. For example, Maertens assumes a child will consume 0.1 g of dust per day without considering the dust loading level, the frequency of hand-to-mouth movements, the hand's skin surface area that transfers the dust to the mouth, the amount of dust transferred during the hand-to-mouth movements, or that the 0.1 g/day ingestion rate is based on a combined soil and house dust ingestion rate. In addition, Maertens et al. (2008) do not take into account exposures beyond childhood. Because the WTC criterion is based on a more robust evaluation of exposures from childhood through adulthood, the health-based WTC criterion of 34 µg/m^2 is more appropriate.

Dietary PAH Intakes

Ingestion of PAHs in food and inhalation of PAHs in tobacco smoke, wood smoke, and ambient air are the primary sources of PAH exposure for most people who are not exposed to PAHs in the workplace (ATSDR 1995). The highest levels of PAHs in food are found in foods that are grilled or smoked. On average, the Agency for Toxic Substances and Disease Registry (ATSDR; 1995) estimates that a total daily intake of PAHs includes 0.16 to 1.6 µg from food, 0.207 µg from air, and 0.027 µg from water. The World Health Organization (WHO; 1998) provides a daily intake estimate from food of 0.1 to 8 µg. The WHO (1998) notes that while PAHs may be found on fruits and vegetables due to atmospheric deposition and/or due to food processing such as frying and roasting, the highest levels of PAHs have been found in smoked meat (over 100 µg/kg) and fish (up to 86 µg/kg).

Assuming exposure to cPAHs in dust at the highest detected concentration for a CT location (16 µg/m^2) reported by Mahler et al. (2010), the total daily intake of cPAHs would be 0.28 µg. This intake is based on exposure parameters identical to those used to derive the WTC screening criterion of 34 µg/m^2. This intake for cPAHs not only is shown to be below an acceptable risk management level through comparison with the WTC criterion, but also is consistent with other background exposures via food and air.

Conclusions

ENVIRON performed a technical review of the study, "Coal-Tar-Based Parking Lot Sealcoat: An Unrecognized Source of PAH to Settled House Dust" by Mahler et al. published in *Environmental Science & Technology* (2010). The review was limited to information published in the study itself and Supplementary Information provided by the publisher.

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ENVIRON notes the following points regarding the study by Mahler et al. (2010):

- Short-comings in the study design introduced uncertainty in data quality and in data evaluation, including uncertain identification of coal-tar sealed and non-coal-tar sealed parking lots; absence of characterization of other PAH sources; absence of consideration of ages of apartment complex, parking lot and sealant, and carpeting; collection of composite samples that may not accurately represent exposure potential; and potential for cross-contamination between samples.

- Both concentration and dust loading are important factors in evaluating exposure to chemicals in dust. Mahler et al. (2010) did not evaluate dust loading, which is critical in understanding *how much* dust is available for contact by residents.

- Mahler et al. (2010) did not compare PAH results to a health-based standard to determine the potential risk associated with the levels measured in house dust. Use of the screening level developed for cleanup of residences near the World Trade Center in New York City indicates that cancer-causing PAHs in dust measured by Mahler et al. (2010) are below levels of concern. In fact, the highest level measured by Mahler et al. (2010) in indoor dust is less than half of the World Trade Center screening level, even though PAH concentrations in dust may be overestimated due to the selected sampling method.

- Intake of cancer-causing PAHs in dust occurs every day through the air we breathe and food we eat. The levels measured by Mahler et al. (2010) that could be taken in via house dust are consistent with background intake levels via food, air, and water.

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