

Predictors of Dermal Exposures to Polycyclic Aromatic Compounds Among Hot-Mix Asphalt Paving Workers

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Objectives: The primary objective of this study was to identify the source and work practices that affect dermal exposure to polycyclic aromatic compounds (PACs) among hot-mix asphalt (HMA) paving workers.

Methods: Four workers were recruited from each of three asphalt paving crews (12 workers) and were monitored for three consecutive days over 4 weeks for a total of 12 sampling days per worker (144 worker days). Two sampling weeks were conducted under standard conditions for dermal exposures. The third week included the substitution of biodiesel for diesel oil used to clean tools and equipment and the fourth week included dermal protection through the use of gloves, hat and neck cloth, clean pants, and long-sleeved shirts. Dermal exposure to PACs was quantified using two methods: a passive organic dermal (POD) sampler specifically developed for this study and a sunflower oil hand wash technique. Linear mixed-effects models were used to evaluate predictors of PAC exposures.

Results: Dermal exposures measured under all conditions via POD and hand wash were low with most samples for each analyte being below the limit of the detection with the exception of phenanthrene and pyrene. The geometric mean (GM) concentrations of phenanthrene were 0.69 ng cm⁻² on the polypropylene layer of the POD sampler and 1.37 ng cm⁻² in the hand wash sample. The GM concentrations of pyrene were 0.30 ng cm⁻² on the polypropylene layer of the POD sampler and 0.29 ng cm⁻² in the hand wash sample. Both the biodiesel substitution and dermal protection scenarios were effective in reducing dermal exposures. Based on the results of multivariate linear mixed-effects models, increasing frequency of glove use was associated with significant ($P < 0.0001$) reductions for hand wash and POD phenanthrene and pyrene concentrations; percent reductions ranged from 40 to 90%. Similar reductions in hand wash concentrations of phenanthrene ($P = 0.01$) and pyrene ($P = 0.003$) were observed when biodiesel was substituted for diesel oil as a cleaning agent, although reductions were not significant for the POD sampler data. Although task was not a predictor of dermal exposure, job site characteristics such as HMA application temperature, asphalt grade, and asphalt application rate (tons per hour) were found to significantly affect exposure. Predictive models suggest that the combined effect of substituting biodiesel for diesel oil as a cleaning agent, frequent glove use, and reducing the HMA application temperature from 149°C (300°F) to 127°C (260°F) may reduce dermal exposures by 76–86%, varying by analyte and assessment method.

Conclusions: Promising strategies for reducing dermal exposure to PACs among asphalt paving workers include requiring the use of dermal coverage (e.g. wearing gloves and/or long sleeves), substituting biodiesel for diesel oil as a cleaning agent, and decreasing the HMA application temperature.

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INTRODUCTION

Asphalt (or bitumen) is the residuum produced through the distillation of crude petroleum oil. Asphalt production is driven by performance specifications governed by physical characteristics rather than asphalt chemistry. The chemistry of asphalt is a complex mixture of inorganic and organic compounds including polycyclic aromatic compounds (PACs), which varies based on crude oil source, refinery process, additives, and application practices (Gamble *et al.*, 1999). In the USA, asphalt is primarily used for road paving (87%), an industry that employs ~300 000 workers (NIOSH, 2001).

Asphalt emissions contain a complex mixture of mostly aliphatic organic compounds and a relatively small quantity of PACs to which paving workers are exposed via inhalation and dermal absorption (NIOSH, 2001; McClean *et al.*, 2004a). While epidemiologic studies have described an excess cancer risk among asphalt-exposed workers, limited information about the source of exposure has made it difficult to determine whether the excess cancer risk is attributable to asphalt or co-exposures such as diesel oil, tobacco, and/or coal tar (Boffetta *et al.*, 2003a, 2003b; Burstyn *et al.*, 2007; Schulte, 2007). A semi-quantitative observational method for dermal exposure assessment has been developed and applied to the assessment of dermal exposure to asphalt during road paving (Agostini *et al.*, 2011). Data from dermal exposure measurement surveys (Jongeneelen *et al.*, 1988; Väänänen *et al.*, 2003; McClean *et al.*, 2004b; Väänänen *et al.*, 2005; Fustinoni *et al.*, 2010) provided critical data forming the basis for the dermal exposure assessment method (DREAM) that provided a quantitative estimate of dermal exposure for a large case-control study of asphalt-exposed workers (Olsson *et al.*, 2010).

To inform epidemiological investigations and optimize exposure reduction strategies for the asphalt paving industry, it is necessary to characterize PAC exposures and identify the sources and work practices that affect exposure. During asphalt paving, dermal exposure to PACs may occur through absorption of chemicals in asphalt fumes and vapors, through deposition of chemicals from the air, or from direct contact with the asphalt binder and contaminated tools, equipment, clothing, or other objects. Diesel oil has also been recognized as a source of PAC exposure among paving workers

(Weker *et al.*, 2004) since diesel oil is commonly used as a cleaning agent by spraying or pouring diesel oil on tools and equipment or by dipping tools and equipment into diesel oil.

We assembled a collaborative partnership composed of representatives from government, industry, labor, and academia to address questions about the source and work practices that affect exposure to PACs among asphalt paving workers. Here, we present the results of our effort to quantify dermal exposure to PACs under normal hot-mix asphalt (HMA) paving working conditions as well as under two additional conditions that represent practical and feasible modifications to typical work practices. The first was to substitute biodiesel for the diesel oil that is typically used as a cleaning agent, and the second was to require increased skin coverage through the use of gloves, hat and neck cloth, clean pants, and long-sleeved shirts. An assessment of air exposure to PAC among this population is found elsewhere (Cavallari *et al.*, 2011) as is information on the collaborative partnership and overall study design (Kriech *et al.*, 2011).

METHODS

Study population

The study population included three asphalt paving crews from three construction companies based out of Wisconsin and Indiana, USA. Four volunteers were selected from each crew for a study population of 12 workers. Each crew included a paver operator, screedman, and raker, while the fourth worker varied by crew (foreman, laborer, and shuttle buggy operator). The paver operator primarily sat on top of the paver to control the speed and direction, while the screedman typically stood at the back of the paver to control the depth and width of the asphalt mat. The raker generally used hand tools (e.g. rake, lute, shovel) for detail work around street castings or curbs. The foreman functioned in the screedman capacity for half the time and also performed raking and general foreman duties. The laborer worked away from the paver for over half the time, driving the asphalt emulsion tack truck, and spray painting. The shuttle buggy operator controlled a machine used to remix and store the asphalt during transfer from the truck to the paver. Study protocols were reviewed and approved by National Institute for

Occupational Safety and Health (NIOSH) Human Subjects Review Board and written informed consent was received by each volunteer prior to participation.

Study design

A repeated measures study design was used to monitor dermal exposure to PACs from August through October in 2008 with simultaneous monitoring of the three crews of four workers. Each of the 12 workers was sampled over three consecutive days during four work weeks, resulting in 12 sampling days per worker and a total of 144 worker days. As part of a larger study, each week was designed to evaluate a different exposure scenario: (i) standard operating conditions (baseline scenario), (ii) using powered air purifying respirators (PAPRs) to reduce inhalation exposure (PAPR scenario), (iii) substituting biodiesel for diesel use as a cleaning agent (no diesel scenario), and (iv) providing cotton gloves with latex-coated palm and fingertips, hat and neck cloth, and clean pants, and long-sleeved shirts to reduce dermal exposure (dermal protection scenario). For this analysis, the baseline and PAPR scenarios were combined as both represented standard conditions with regard to dermal exposures. In the 'no diesel' scenario, diesel oil normally used to clean tools and equipment was removed from the site and replaced with a biodiesel product (B-100) comprised 100% mono-alkyl esters of long-chain fatty acids, often produced from vegetable oil or animal fats, and containing no PACs (Bajpai and Tyagi, 2006). Each crew member maintained the same exposure scenario. The order of the week-long exposure scenarios was randomized among the three crews and the crossover design allowed each worker to serve as his own control.

Site conditions and worker observations

Workers were monitored while performing normal paving work that included large primary roads and a parking lot. Highway class pavers with engineering controls (NIOSH, 1997) were used on all but one occasion when a paver equipped with a widener was used to accommodate logistical road issues. Asphalt binder types also varied by crew and project and included the following Superpave performance grades (PG), PG 64-22, PG 58-28, and PG 76-22.

HMA temperatures were monitored at various times throughout the workday. A minimum of six temperature readings were collected daily from the asphalt mat at the back of the screed using an HMA Lab Supply 8" Stainless Steel Dial Stem thermometer, with a 0–400°F range (Catalog #

TM-4500). Measurements of wind speed, air temperature, and humidity using a Kestrel® 4000 Pocket Weather Tracker as well as weather-related comments were collected four times each sampling day. Job specifications including asphalt grade and tonnage use were recorded daily.

At the worksite, workers were unable to wash their hands. No workers appeared to use barrier creams, although suntan lotion use was observed among some users. Workers occasionally used gloves as part of their normal work practice ($n = 32$ worker days); however, during dermal protection scenario week, all workers were required to wear gloves as well as hat and neck cloth, clean pants, and long-sleeved shirts.

Participant characteristics including sex, age, height, weight, and smoking status were obtained at the start of the study. Daily observations of clothing types (length of shirt sleeves, pant description, and glove type) and frequency of use were recorded. Daily observations of downtime (when workers were away from asphalt while eating lunch, waiting for delivery of asphalt, working on equipment, or other activities with no asphalt exposure) were estimated in minutes by on-site personnel based on field records.

Collection of dermal exposure samples

Dermal exposures to PACs were quantified using two sampling technologies: a passive organic dermal (POD) sampler created for this study (Olsen *et al.*, 2011) and a hand wash method previously employed in studies of asphalt workers (Jongeneelen *et al.*, 1988; Väänänen *et al.*, 2005). Both methods were used to collect daily exposure measurements from each worker. The sample preparation process and experimental and instrumental conditions used for both the hand wash and POD samplers are described in detail elsewhere (Kriech *et al.*, 2011).

Hand wash sample collection. Hand wash samples were collected at the beginning and end of each worker's shift using a previously described method (Jongeneelen *et al.*, 1988; Väänänen *et al.*, 2005). Briefly, 3 ml of sunflower oil were added to the palm of one hand and after rubbing the hands together in a washing motion for 1 min, the workers wiped the oil from their hands using a crepe material (DuPont™ Sontara®; creped by Micrex Corporation). Each wipe was extracted with dichloromethane and analyzed by gas chromatography with flame ionization detection (GC/FID) for total organic matter (TOM) and GC with time of flight mass spectrometry (GC/TOFMS) for PAC determinations (Kriech *et al.*, 2011). The presence of nicotine [2-(1-methyl-2-

pyrrolidiny]pyridine] was investigated by GC/TOFMS analysis (Kriech *et al.*, 2011). Nicotine was identified as present when sample concentrations exceeded the practical quantitation limit of 12.5 ng per wash. The surface area of the hand was estimated by first tracing each hand on a sheet of paper. The hand shape was cut and weighed on an analytical balance. Individual area calculations were determined by taking the individual tracing weight divided by the average tracing weight from all participants' hand tracings and multiplying by 820 cm², the average surface area of the hand (Väänänen *et al.*, 2005). Analytical results for hand wash samples were normalized for each individual by dividing by the surface area of the hand.

POD sample collection. POD samplers were designed specifically for this study and are described in detail elsewhere (Olsen *et al.*, 2011). The five-layer sampler was designed to capture the full range of potential HMA emissions as well as other workplace exposures, such as diesel oil. The outer polypropylene layer served as a barrier intended to be analogous to that of human skin. The middle layers included polyurethane foam (PUF) and a C-18 solid-phase extraction (SPE) disk to capture the organic phase that may pass through the outer polypropylene layer. The inner-most layer consisted of activated carbon cloth (ACC) to capture any volatiles that were not retained by the middle layers. An ethylene tetrafluoroethylene (ETFE) layer served to isolate the SPE disk from the ACC layer.

POD samplers were positioned on each worker's wrist/forearm and worn over a full work shift. For the baseline, PAPR, and no diesel exposure scenarios, PODs were positioned on the exterior of the clothing. Over these three exposure scenarios, when participants wore long sleeves, PODs were positioned on the outside of clothing to maintain consistency throughout the scenario and to facilitate comparisons in exposures between scenarios. For the dermal protection exposure scenario, PODs were positioned underneath the long-sleeve cotton shirt to approximate exposure considering dermal protection. PODs were retrieved from workers at the end of the work shift, transferred to a Mylar bag, and stored at -20°C until analyzed.

Once ready for extraction, the five layers of the POD sampler were disassembled. After weighing the polypropylene layer for total particulate matter (TPM), it was extracted with dichloromethane, filtered, and analyzed by high-temperature GC/FID for asphalt binder, GC/FID for TOM, and GC/TOFMS for PAC determinations (Kriech *et al.*, 2011).

Additional analyses were performed on extracts from the PUF/SPE and ACC layers (Kriech *et al.*, 2011). The PUF/SPE extract was analyzed by GC/FID for TOM and GC/TOFMS for PAC determinations. The ETFE/ACC extract was analyzed by GC/FID for TOM and GC/TOFMS for PAC determinations.

Laboratory analysis

A total of 33 individual parent PACs results were obtained using GC/TOFMS. TOM refers to the amount of organics collected on the sampler ranging from C6 to C42 as determined by GC/FID using a modification of USA Environmental Protection Agency Method SW846-8015B (Kriech *et al.*, 2011). Industrial hygiene sample collection and laboratory analysis were performed by Heritage Research Group, Indianapolis, IN, USA, with on-site assistance and oversight during sample collection by a team from NIOSH, Cincinnati, OH, USA.

When the field blanks provided a standard deviation (SD) (calculated when >50% of samples were found to contain quantifiable levels of materials), the limit of detection (LOD) was defined as the method detection limit (MDL) (three times the SD of the field blanks). When no SD could be calculated from the field blanks, a LOD was defined as the instrumental detection limit (IDL). MDLs were calculated for TPM, fluorene, and naphthalene on the polypropylene layer and fluoranthene, fluorene, naphthalene, phenanthrene, and pyrene on the PUF/SPE layers.

In cases where the mean field blank was statistically significantly different than zero, blank correction was performed. Analysis of variance (ANOVA) was performed to determine if there was a difference in blank means by crews. Based on the results of the ANOVA test evaluating the difference in blank values by crew, samples requiring blank correction were subtracted by either the overall mean field blank value or by the mean field blank value for the corresponding crew. When the laboratory reported a value below the LOD, it was used as reported. When no value was reported, the LOD divided by the square root of 2 was substituted.

Data analysis

Distributions of the dermal exposure data were examined using histograms and normal probability plots. Shapiro-Wilks' tests and graphical displays indicated that the dermal data were not normally distributed, and therefore, the data were log-transformed to achieve an approximately normal distribution. Accordingly, geometric means (GMs) and geometric

standard deviations (GSDs) are reported and statistical modeling was performed on the log-transformed data. For summary statistics, only analytes detected in at least 30% of samples were included in tables (with the exception of fluorene and naphthalene, which were included to facilitate comparisons despite low numbers of samples with detectable materials) and multivariate analysis was performed on PAC analytes detected in at least 60% of samples. To examine the relationship between the log-transformed hand wash and POD data, correlation coefficients were calculated using linear mixed-effects models as described by Hamlett *et al.* (2003).

Since multiple dermal samples were collected from each worker, linear mixed-effects models were used to evaluate the determinants of exposure using a compound symmetry covariance matrix structure. The repeated measures design and the use of linear mixed-effects models allows each worker to serve as his own control while evaluating changes between exposure scenarios.

Importantly, for this analysis of dermal exposure, the baseline and PAPR scenarios were combined because the introduction of PAPRs was not intended or expected to appreciably affect measured dermal exposures (confirmed via analyses not shown). Accordingly, our analysis of dermal exposures in this paper compare 2 weeks of baseline conditions, 1 week of biodiesel substitution, and 1 week of dermal reduction.

A priori, we included a four-level categorical variable for task (paver operator, screedman, raker, and other) within each model based on prior studies suggesting the importance of task as predictor of exposure (McClellan *et al.*, 2004b). We also included a dichotomous variable to indicate whether biodiesel was substituted for diesel oil use and a categorical variable for glove use (none, sometimes, or frequent). The glove use variable captured not only the dermal protection exposure scenario when all workers wore gloves but also accounted for glove use during the other exposure scenarios based on-site observation of use and frequency.

Potential predictors of dermal exposures included air temperature, wind speed, relative humidity, crew (categorical), HMA application temperature, hourly work rate (tons of asphalt applied per hour), daily work rate (tons of asphalt applied per day), asphalt grade (categorical variable), minutes sampled, minutes of downtime, smoking status, hand wash nicotine presence, and pre-shift analyte concentration (for hand wash models only). First, each predictor was evaluated in a univariate model. Next, multivariate models were constructed including variables that were marginally

statistically significant ($P < 0.10$) in 50% of univariate models for each analyte. Models were further refined to include variables that were marginally significant ($P < 0.10$) in $\geq 50\%$ of the multivariate models for each analyte. For consistency, final models were constructed to include the same variables for both POD and hand wash methods. The non-linearity of HMA application temperature was evaluated by categorical and squared variables using likelihood ratio tests. All statistical analyses were conducted in SAS statistical software (version 9.2—Cary, NC, USA); statistical significance is reported at the 0.05 level, unless otherwise noted.

RESULTS

The 12 workers who participated in this study were all male, with a mean age of 36 (range of 24–59) years. Six of the workers smoked cigarettes, five were non-smokers, and one worker quit smoking during the study period. One non-smoker chewed tobacco. Nicotine was detected in 49 (35%) of the post-shift hand wash samples. Over the eight sampling weeks, the workers were monitored over a wide range of weather conditions as indicated by the observed air temperature, wind speed, and humidity (Table 1). In general, sampling occurred over the entire work shift with a median time sampled of 652 min or ~ 11 h and a range from 6.5 to 13.6 h. Worksite conditions varied with HMA application temperatures ranging from 121 to 154°C (250–310°F) and a wide range of asphalt use, work rates, and downtime experienced over the 36 sampling days. HMA application temperatures varied by crew, with one crew using a wide range of application temperatures from 127 to 154°C (260–310°F), the second a smaller range on the higher end from 146 to 152°C (295–305°F), and the third using lower application temperatures ranging from 121 to 145°C (250–293°F). Over the 36 sampling

Table 1. Summary of worksite conditions over 36 sampling days.

	Median	Range
Air temperature, °C (°F)	21 (69)	8 (46)–31 (87)
Wind speed, km h ⁻¹ (m h ⁻¹)	6.9 (4.3)	1.0 (0.6)–17 (11)
Humidity, %	59	32–91
Hot-mix temperature, °C (°F)	144 (291)	121(250)–154 (310)
Daily work rate, tons day ⁻¹	1449	418–3300
Hourly work rate, tones h ⁻¹	149	46–289
Time sampled, min	652	387–818
Downtime, min day ⁻¹	120	75–700

Table 2. Summary of pre- and post-shift personal dermal exposure levels from hand wash method over 144 HMA paving worker days across all exposure scenarios.

Analyte	LOD	Pre-shift			Post-shift		
		% Detect	GM (GSD)	Range	% Detect	GM (GSD)	Range
Summary measures ($\mu\text{g cm}^{-2}$)							
Total organic matter	3.54	13	NA ^a	5.26–40.8	54	7.48 (2.4)	5.38–264
Individual PACs ^b (ng cm^{-2})							
Anthracene	0.018	30	NA ^a	0.03–1.0	34	NA ^a	0.04–1.5
Fluoranthene	0.020	32	NA ^a	0.03–0.7	48	NA ^a	0.04–6.2
Fluorene	0.015	25	NA ^a	0.03–3.0	13	NA ^a	0.03–5.0
Naphthalene	0.226	2	NA ^a	0.27–0.67	13	NA ^a	0.23–1.2
Phenanthrene	0.018	91	0.241 (4.4)	0.03–2.0	73	0.285 (7.9)	0.03–6.1
Pyrene	0.020	56	0.063 (4.5)	0.03–2.0	73	0.285 (7.5)	0.03–6.1

^aNA, not applicable, GM and GSD were not calculated for samples with high percentage below the LOD.

^bA total of 33 individual parent PACs results were obtained using GC/TOFMS. PACs investigated, but with >70% below the LOD include: acenaphthene, acenaphthylene, benz[*a*]anthracene, benzo[*a*]pyrene, benzo[*b*]fluoranthene, benzo[*g,h,i*]perylene, 5-methylchrysene, 1-nitropyrene, benzo[*k*]fluoranthene, chrysene, dibenz[*a,h*]anthracene, indeno[1,2,3-*cd*]pyrene, benzo[*j*]fluoranthene, 7,12-dimethylbenz[*a*]anthracene, benzo[*e*]pyrene, 3-methylcholanthrene, dibenz[*a,h*]acridine, dibenz[*a,j*]acridine, 7H-dibenzo[*c,g*]carbazole, dibenzo[*a,e*]fluoranthene, dibenzo[*a,e*]pyrene, benzo[*r,s,t*]pentaphene, dibenzo[*a,h*]pyrene, dibenzo[*a,l*]pyrene, benzo[*b*]naphtho[2,3-*d*]thiophene, cyclopenta[*c,d*]pyrene, and triphenylene.

days, the crews primarily used asphalt grade PG 58-28 (61%) as compared to PG 64-22 (33%) and PG 76-22 (6%). The majority of the work was applying surface course (91%) as compared to base (3%) or binder (or leveling) (6%).

The pre- and post-shift concentrations from the hand wash are summarized in Table 2. The GMs of post-shift concentrations were consistently and significantly ($P < 0.0001$) higher than pre-shift concentrations for each analyte. TOM was detected in 54% of post-shift samples and 13% of pre-shift samples. Of the 33 parent PACs, anthracene, fluoranthene, fluorene, phenanthrene, and pyrene had >30% of the samples above the LOD for post-shift samples. Only phenanthrene and pyrene had >60% of samples above the LOD and were included in additional statistical analyses.

The concentrations from the polypropylene layer of the POD samplers are summarized in Table 3. Due to low detection, analytes within the PUF-SPE and ACC layers were excluded from the table. Of the 33 parent PACs, fluoranthene, phenanthrene, and pyrene had >30% of the samples above the LOD in the polypropylene layer. Similar to hand wash samples, among PACs, only phenanthrene and pyrene within the polypropylene layer had >60% of samples above the LOD and were considered for multivariate analysis.

Figure 1 shows the relationship between phenanthrene and pyrene concentrations in the post-shift hand wash samples and the polypropylene layer of

the POD sampler (phenanthrene in Fig. 1a and pyrene in Fig. 1b). There was a weak linear correlation between the post-shift hand wash and polypropylene layer of the POD for phenanthrene ($r = 0.27$) as well as pyrene ($r = 0.19$). Phenanthrene and pyrene concentrations were moderately correlated within POD samples ($r = 0.64$) and hand wash samples ($r = 0.62$).

For hand wash data, variables that were marginally significant ($P < 0.10$) predictors of phenanthrene or pyrene in univariate analyses included pre-shift concentration, wind speed, crew, glove use frequency, biodiesel use, HMA application temperature, hourly work rate (tons of asphalt per hour), daily work rate (tons of asphalt per day), asphalt grade, and presence of nicotine in hand wash sample. Due to the correlation between hourly and daily work rate, only hourly work rate was retained in multivariate models. Because of the relationship between crew and HMA application temperature with little variation in HMA application temperature by crew, only HMA application temperature was retained in hand wash models.

For POD data (polypropylene layer), variables that were marginally significant ($P < 0.10$) predictors of phenanthrene or pyrene concentration included air temperature, wind speed, crew, glove use frequency, biodiesel use, HMA application temperature, hourly work rate, daily work rate, minutes of downtime, asphalt grade, and hand wash nicotine presence. Like the hand wash models, hourly work

Table 3. Summary of personal dermal exposure levels from POD sampler polypropylene layer over 144 HMA paving worker days across all exposure scenarios.

Analyte	LOD	% Detect	GM (GSD)	Range
Summary measures ($\mu\text{g cm}^{-2}$)				
Total organic matter	6.00	34	NA ^a	6.12–97.4
Total particulate matter	8.04	99	41.17 (2.75)	6.57–597.7
Individual PACs ^b (ng cm^{-2})				
Acenaphthene	0.020	16	NA ^a	0.073–1.99
Fluoranthene	0.034	53	0.12 (5.88)	0.071–14.20
Fluorene	0.73	12	NA ^a	0.787–2.69
Naphthalene	1.16	4	NA ^a	1.26–8.03
Phenanthrene	0.032	92	0.69 (9.31)	0.034–15.76
Pyrene	0.034	73	0.30 (5.59)	0.087–7.67

^aNA, not applicable, GM and GSD were not calculated for samples with high percentage below the LOD.

^bA total of 33 individual parent PACs results were obtained using GC/TOFMS. PACs investigated, but with >70% below the LOD include: acenaphthylene, anthracene, benz[*a*]anthracene, benzo[*a*]pyrene, benzo[*b*]fluoranthene, benzo[*g,h,i*]perylene, 5-methylchrysene, 1-nitropyrene, benzo[*k*]fluoranthene, chrysene, dibenz[*a,h*]anthracene, indeno[1,2,3-*cd*]pyrene, benzo[*j*]fluoranthene, 7,12-dimethylbenz[*a*]anthracene, benzo[*e*]pyrene, 3-methylcholanthrene, dibenz[*a,h*]acridine, dibenz[*a,j*]acridine, 7H-dibenzo[*c,g*]carbazole, dibenzo[*a,e*]fluoranthene, dibenzo[*a,e*]pyrene, benzo[*r,s,t*]pentaphene, dibenzo[*a,h*]pyrene, dibenzo[*a,l*]pyrene, benzo[*b*]naphtho[2,3-*d*]thiophene, cyclopenta[*c,d*]pyrene, and triphenylene.

rate (but not daily work rate) and HMA application temperature (but not crew) was further pursued in multivariate models.

Table 4 shows the parameter estimates, standard errors (SE), and *P*-values for the final post-shift hand wash models. The substitution of biodiesel for diesel oil was associated with a significant decrease in phenanthrene ($P = 0.01$) and pyrene ($P = 0.003$) concentrations as compared to the baseline exposure scenario. Similarly, glove use frequency was significantly associated with both phenanthrene ($P < 0.0001$) and pyrene ($P < 0.0001$) concentrations, such that concentrations decreased with increased glove use (i.e. frequent < sometimes < none of time). HMA application temperature was a significant ($P = 0.01$) predictor of phenanthrene (but not pyrene, $P = 0.15$), with increases in temperature showing increased exposure to phenanthrene. Pyrene and phenanthrene concentrations in hand wash samples were found to be associated with asphalt grade, work rate, nicotine in hand wash, and pre-shift hand wash concentration but typically significant for one analyte or the other. Surprisingly, task was not a significant predictor of either phenanthrene or pyrene in hand wash samples while adjusting for other factors.

Table 4 also shows the parameter estimates, SE, and *P*-values for the final POD models (polypropylene layer), which were consistent with the hand wash results. For instance, increasing frequency of glove use was found to be significantly associated with

reductions in phenanthrene ($P < 0.0001$) and pyrene ($P < 0.0001$) on the polypropylene layer of the POD sampler. A modest reduction in phenanthrene and pyrene was also observed for the biodiesel substitution of diesel oil. However, the observed reduction was not significant. HMA application temperature was a significant predictor of both phenanthrene ($P = 0.02$) and pyrene ($P = 0.03$) with a positive linear association. Phenanthrene concentrations in POD samples were found to be associated with work rate and nicotine in hand wash samples. As was found with the hand wash models, task was not a predictor of phenanthrene or pyrene concentrations for the POD models.

The random effects including the estimated (log scale) variance components for between-worker (σ_{BW}^2) and within-worker (σ_{WW}^2) are presented for phenanthrene and pyrene post-shift hand wash and POD models (Table 4). In each of the models, after adjusting for fixed effects, the σ_{BW}^2 was small (ranging from 0.0 to 0.4) and nearly an order of magnitude lower than the σ_{WW}^2 (ranging from 1.1 to 3.2). This suggests that exposures varied more within-worker from day-to-day than between-worker, which may be a reflection of the study design where each worker was evaluated over multiple exposure scenarios.

To ease interpretation of our findings in Table 4, Table 5 presents the adjusted GM phenanthrene and pyrene concentrations for both sampling methods to show the combined effects of the biodiesel

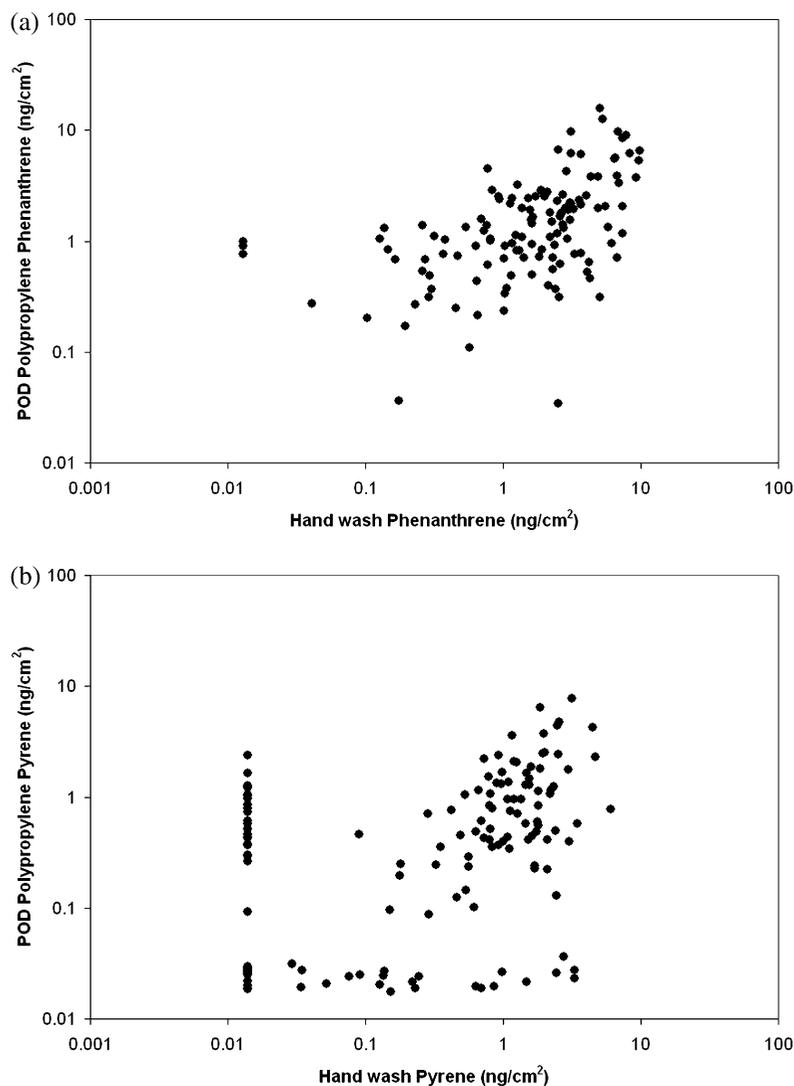


Fig. 1. Correlation between POD polypropylene and hand wash phenanthrene and pyrene concentrations. (a) Phenanthrene hand wash versus POD polypropylene ($r = 0.27$). (b) Pyrene hand wash versus POD polypropylene ($r = 0.19$).

substitution (unrestricted diesel oil use versus biodiesel), glove use (none versus frequent), and HMA application temperature [149°C (300°F) or 127°C (260°F)]. Since the data analysis was conducted using logged data, the GM exposure concentrations were estimated by exponentiating the estimates from the adjusted models. The adjusted GM concentration of phenanthrene in hand wash samples associated with no glove use, unrestricted use of diesel oil, and the higher HMA application temperature of 149°C (3.60 ng cm⁻²) is approximately seven times the adjusted GM concentration of phenanthrene on hand wash associated with glove use (frequent), the

biodiesel substitute, and the lower HMA application temperature of 127°C (0.51 ng cm⁻²). A similar comparison is estimated to yield a 5-fold decrease of pyrene on hand wash, a 4-fold decrease of phenanthrene on the polypropylene layer of the POD sampler, and a 7-fold decrease in pyrene on the polypropylene layer of the POD sampler.

DISCUSSION

Since previous studies of asphalt pavers have demonstrated that the contribution of dermal exposure to the total absorbed dose of PACs may be larger than

Table 4. Determinants of logged dermal exposure (nanograms per square centimeter) among HMA paving workers, based on linear mixed-effects models.

Parameters	Post-shift hand wash				POD ^a			
	Phenanthrene		Pyrene		Phenanthrene		Pyrene	
	β (SE)	<i>P</i> -values	β (SE)	<i>P</i> -values	β (SE)	<i>P</i> -values	β (SE)	<i>P</i> -values
Fixed effects								
Intercept	-2.96 (1.82)	0.14	2.88 (2.79)	0.31	-4.64 (2.63)	0.12	-5.95 (2.51)	0.05
Task		0.18		0.58		<0.0001		0.96
Rakers	Reference		Reference		Reference		Reference	
Screedman	0.14 (0.34)	0.69	0.12 (0.46)	0.81	0.50 (0.12)	0.004	-0.07 (0.64)	0.92
Paver operator	0.67 (0.34)	0.09	0.61 (0.47)	0.23	0.73 (0.14)	0.001	-0.16 (0.65)	0.81
Other	0.68 (0.35)	0.09	0.40 (0.49)	0.43	0.10 (0.15)	0.53	-0.33 (0.66)	0.63
Biodiesel substitution	-0.76 (0.27)	0.01	-1.26 (0.41)	0.003	-0.12 (0.39)	0.75	-0.26 (0.35)	0.45
Glove use		<0.0001		<0.0001		<0.0001		<0.0001
None	Reference		Reference		Reference		Reference	
Sometimes	-0.46 (0.29)	0.13	-0.85 (0.44)	0.07	-0.35 (0.32)	0.30	-0.64 (0.38)	0.11
Frequent	-1.49 (0.24)	<0.0001	-2.33 (0.36)	<0.0001	-2.16 (0.33)	<0.0001	-2.21 (0.32)	<0.0001
Asphalt application temp (per 100°C)	3.26 (1.27)	0.01	-2.77 (1.94)	0.15	4.30 (1.81)	0.02	4.48 (1.75)	0.01
Asphalt Grade		0.01		0.08		0.09		0.45
PG 76-22	Reference		Reference		Reference		Reference	
PG 58-28	-0.21 (0.41)	0.62	1.57 (0.64)	0.03	0.13 (0.65)	0.85	-0.17 (0.54)	0.76
PG 64-22	-1.36 (0.50)	0.02	1.20 (0.77)	0.15	-1.05 (0.76)	0.20	-0.71 (0.66)	0.31
Hourly work rate (tons h ⁻¹)	-0.002 (0.002)	0.29	-0.01 (0.003)	0.04	-0.005 (0.002)	0.03	-0.001 (0.002)	0.64
Nicotine detected in hand wash	0.93 (0.25)	0.0003	1.05 (0.77)	0.005	-0.42 (0.15)	0.01	0.41 (0.38)	0.28
Pre-shift hand wash	0.24 (0.07)	0.002	0.04 (0.10)	0.66	NA ^b		NA ^b	
Random effects								
σ^2 BW ^c	0.1		0.0		0.0		0.4	
σ^2 WW ^c	1.1		2.7		3.2		1.9	

^aPOD, passive organic dermal monitor, polypropylene layer.

^bNA, Pre-shift hand wash concentrations are not applicable to POD samplers.

^cBetween-worker (σ^2 BW) and within-worker (σ^2 WW) variance estimates.

Table 5. Adjusted^a GM exposure levels over different worksite exposure conditions.

Glove use	Diesel oil use	149°C (300°F) Asphalt application temperature				127°C (260°F) Asphalt application temperature			
		Post-shift hand wash		POD ^b		Post-shift hand wash		POD ^b	
		Phenanthrene (ng cm ⁻²)	Pyrene (ng cm ⁻²)	Phenanthrene (ng cm ⁻²)	Pyrene (ng cm ⁻²)	Phenanthrene (ng cm ⁻²)	Pyrene (ng cm ⁻²)	Phenanthrene (ng cm ⁻²)	Pyrene (ng cm ⁻²)
None	Unrestricted	3.60	0.11	2.98	1.73	1.74	0.21	1.14	0.64
None	Biodiesel substitute	1.67	0.03	2.63	1.33	0.81	0.06	1.01	0.49
Frequent	Unrestricted	2.28	0.05	2.10	0.91	1.11	0.09	0.81	0.34
Frequent	Biodiesel substitute	1.06	0.01	1.86	0.70	0.51	0.02	0.71	0.26

^aAll models adjusted for task, asphalt grade, asphalt temperature, hourly work rate, and hand wash nicotine. Hand wash models also adjusted for pre-shift hand wash concentration. Modeled GM results are for screedman, with no nicotine present with median hourly work rate of 149 tons h⁻¹ and median pre-shift hand wash concentrations.

^bPOD, passive organic dermal monitor, polypropylene layer.

inhalation (McClean *et al.*, 2004a; Väänänen *et al.*, 2005; Fustinoni *et al.*, 2010), the primary goal of this analysis was to identify predictors of dermal exposure and evaluate the impact of work practices on dermal exposure among asphalt paving workers. Our findings suggest that substituting biodiesel for diesel oil as a cleaning agent and requiring the use of gloves can significantly reduce the amount of PACs that come into contact with the skin of workers. Additionally, our findings suggest that dermal exposure is positively associated with the HMA application temperature, suggesting that decreasing application temperature would likely decrease dermal exposure.

In the absence of a standardized method, dermal exposure monitoring was performed via two methods, hand wash and POD samplers, in an attempt to thoroughly characterize PAC dermal exposures occurring through multiple routes. We assessed dermal exposure at the hands/wrist location as a correlate of total dermal exposure and to maintain consistency with prior studies of asphalt workers (Jongeneelen *et al.*, 1988; Väänänen *et al.*, 2005, 2006; McClean *et al.*, 2004b; Olsen *et al.*, 2011). However, while dermal exposures on the hand/wrist have been shown to be correlated with exposures at other locations (Fustinoni *et al.*, 2010; Olsen *et al.*, 2011), it is unclear what percentage of total dermal exposure hand/wrist exposures represent. We hypothesized that dermal exposure to PACs may occur when asphalt fumes condense on the skin or from direct contact with asphalt binder, diesel oil, or contaminated tools and equipment. We expected that the hand wash sample concentrations would reflect airborne exposures and transfer from contaminated tools and equipment, while due to the POD location on the wrist/forearm, concentrations are more likely

to reflect primarily airborne exposure levels although transfer from contaminated objects to the outer surface of the POD sampler is still possible. Variations in the source and therefore composition of the POD and hand wash samples can be assessed by looking at changes in the ratio between phenanthrene and pyrene. For the POD sampler using the predicted GMs in Table 5, the phenanthrene/pyrene ratio is 2–3 across the different exposure scenarios, suggesting consistency in exposure composition and source. However, this is not the case for the hand wash samples where the phenanthrene/pyrene ratio varies from 8 to 79 with differences in the ratio observed with glove use, diesel oil use, as well as asphalt application temperature suggesting heterogeneity in exposure composition and therefore source. This adds to support to our hypothesis that the POD samples are reflecting primarily airborne exposures, while the hand wash samples reflect both airborne exposure as well as exposure from transfer which may be from heterogeneous sources including but not limited to liquid diesel oil, lubricants, and asphalt binder. The majority of the analytes were below the LOD for both sampling techniques. Because of this, statistical analysis was only performed on the polypropylene layer of the POD sampler, although combining polypropylene and PUF-SPE layers resulted in similar associations (data not shown).

Within our population, hand wash and POD phenanthrene and pyrene concentrations were more correlated within than between sampling methods. The GM (SD) phenanthrene concentrations were higher [0.69 (9.3) ng cm⁻² for the polypropylene layer of the POD and 1.37 (3.9) ng cm⁻² for the post-shift hand wash] as compared to pyrene [0.30 (5.6) ng cm⁻² for the polypropylene layer of

the POD and 0.29 (7.9) ng cm⁻² for the post-shift hand wash]. In general, in multivariate models, for both POD and hand wash, phenanthrene and pyrene had similar predictors, with the exception of the effect of the biodiesel exposure scenario and hand wash nicotine.

The majority of PAC dermal exposures among this population were similar in magnitude; yet lower than concentrations reported by studies using polypropylene exposure pads, the same material as the first layer of our POD. Compared to the 0.7 ng cm⁻² phenanthrene concentrations, we observed across all exposure scenarios, dermal phenanthrene concentrations were 1.8 ng cm⁻² in Italian pavers (Fustinoni *et al.*, 2010) and 2.6 ng cm⁻² in a Finnish study of pavers applying of stone mastic asphalt (Väänänen *et al.*, 2005). Compared to the current study in which POD pyrene concentrations were 0.3 ng cm⁻² across all exposure scenarios, pyrene concentrations were 0.1 ng cm⁻² among Finnish pavers also exposed to varying amounts of waste material (Väänänen *et al.*, 2006), 0.5 ng cm⁻² in Italian pavers (Fustinoni *et al.*, 2010), 0.6 ng cm⁻² in a Finnish study of pavers applying of stone mastic asphalt (Väänänen *et al.*, 2005), 3.5 ng cm⁻² in US pavers also working with varying amounts of recycled asphalt pavement (RAP) using small pavers without engineering controls (McClellan *et al.*, 2004b), and 12.4 ng cm⁻² among workers in the Netherlands also exposed to coal tar-binding material (Jongeneelen *et al.*, 1988). Likewise, our hand wash phenanthrene and pyrene concentrations of 1.4 and 0.3 ng cm⁻², respectively, were similar to the concentrations of 3.0 and 0.88 ng cm⁻² observed in a study of Finnish pavers using varying amounts of RAP (Väänänen *et al.*, 2005).

Substituting biodiesel for diesel oil resulted in a 53 and 72% reduction in hand wash phenanthrene and pyrene concentrations, yet no significant change in POD phenanthrene and pyrene concentrations. It is a standard work practice in the asphalt paving industry to use diesel oil to rinse down paving equipment, shovels, lutes (or rakes), putty knives, and other tools. Cleaning of tools or equipment occurs when diesel oil is squirted or poured from a plastic jug, sprayed using a hose, or the tools are dipped into a container filled with diesel oil. Additionally, approximately 1–3 gallons of diesel oil is used at the end of the day when the paver is rinsed down by spraying, thus, releasing substantial diesel oil fumes and vapors in the process.

During the biodiesel exposure scenario, the reduction of PACs in hand wash samples, but not in the POD samplers, suggests that the route of dermal

exposure to diesel-derived PAC may be through direct contact. While we also observed a reduction in air PAC exposures with substitution of biodiesel for diesel oil (Cavallari *et al.*, 2011), the reduction in airborne exposures may not have been large enough to produce a statistically significant reduction in the POD polypropylene exposure levels.

In the dermal protection scenario, workers were provided gloves, hat and neck cloth, and clean pants and long-sleeved shirts to reduce dermal exposure. The glove use variable captured the dermal protection exposure scenario (when all workers wore gloves) and also accounted for glove use during the other exposure scenarios. The majority (69%) of frequent glove use occurred during the dermal protection exposure scenario with the remainder (31%) occurring during the baseline exposure scenarios when some workers routinely wore gloves that were not provided as part of this investigation. As compared to baseline, 'frequent' glove use resulted in significant reductions ranging from 29 to 57% in phenanthrene and pyrene. In predictive models for hand wash and POD phenanthrene and pyrene concentrations, we observed an additional statistically significant reduction in exposure with some or frequent time glove use as compared to no glove use. During the dermal protection scenario, PODs were placed under the long sleeve shirts to get a better estimate of actual dermal exposures. Therefore, we expected that the frequent glove use variable would be predictive of lower POD concentrations since workers were provided gloves and long-sleeved shirts during the dermal protection scenario. Since the POD was placed on the wrist/forearm of each participant, the data cannot distinguish whether both or either glove or long sleeve use is responsible for the lowering of PAC exposures observed within this population.

HMA application temperature was a statistically significant predictor of hand wash phenanthrene and POD phenanthrene and pyrene exposures. Mean adjusted models indicate that under baseline conditions, a reduction in HMA application temperature from 149°C (300°F) to 127°C (260°F) may result in a 52–63% reduction in dermal phenanthrene and pyrene levels. This is the first study to demonstrate an association between reductions in dermal exposure to PACs and HMA application temperature. Previously, an evaluation of historical exposures among European asphalt pavers found a similar association between airborne asphalt vapor exposure and HMA application temperature (Burstyn *et al.*, 2000). The lower dermal exposures may be a reflection of the lower concentrations of airborne PACs

during lower HMA application temperatures (Cavallari *et al.*, 2011) resulting in less condensation on skin and tools, equipment, etc. Because of a correlation between crew and HMA application temperature, we could not simultaneously evaluate crew and HMA application temperature effects; however, the positive statistically significant linear association that we observed lends further evidence to a relationship between HMA application temperature and dermal exposures. The association between dermal exposures and HMA application temperature also appears to be independent of the effects of asphalt grade, as the effects of application temperature did not change with removal of asphalt grade from the model (data not shown). We observed a statistically significant decline in dermal pyrene concentrations with an increase in work rate (hourly tons of asphalt applied).

Previous investigations among asphalt pavers found crew to be an important predictor of dermal exposures, most likely serving as a surrogate for other factors including work practices, equipment use, and other production characteristics (McClean *et al.*, 2004b). Likewise, task has also been identified as a determinant of PAC concentration (McClean *et al.*, 2004b), as it serves as an indicator for the degree to which each worker requires actual contact with the asphalt. Yet, we found no association between task and exposure levels, and our models showed little variability between workers, suggesting that task and other differences between workers did not contribute to the variability in dermal exposures. However, given only three workers for each task, the study may have been underpowered to observe differences in exposure by task. (The study was designed to evaluate the effects of the different exposure scenarios not to characterize differences between tasks or crews.) Our finding suggests that smokers have a higher potential for dermal exposure to PACs, as the presence of nicotine in hand wash samples was associated with increased hand wash phenanthrene and pyrene, both of which have been measured in cigarette smoke (Ding *et al.*, 2004). However, dermal exposure to PACs with cigarette smoke exposures warrants additional research as POD samplers showed a statistically significant decline in phenanthrene when nicotine was present in the hand wash and no association with pyrene.

Mean adjusted models predict that with both biodiesel substitution and frequent glove use, 38–88% reductions in phenanthrene and pyrene may be achieved and additionally lowering the HMA application temperature to 127°C (260°F) may further lower phenanthrene levels for a total reduction of 76–86%. However, because the combination of

increased dermal protection and biodiesel substitution along with changes in HMA application temperature were not investigated in this study, additional studies are needed to estimate the effect of the combined exposure scenarios.

The current study was limited to 12 workers over three study crews. While the small sample size may limit the generalizability of the study results, it does not affect the study's internal validity. We attempted to control working conditions during each exposure scenario and across crews, but because this was a real-world experiment and not a controlled lab-based experiment, we did not have perfect compliance. During the dermal protection scenario, gloves may not have been used 100% of the time; despite this, we still observed a reduction in dermal exposures. We also observed a reduction in dermal exposures when biodiesel was introduced. During the baseline scenario, one crew used a biodiesel/diesel blend (20/80), yet reductions in dermal exposure were observed after switching to 100% biodiesel.

CONCLUSIONS

Reductions in dermal exposure to PACs among asphalt paving workers can be achieved by substituting biodiesel for diesel oil as a cleaning agent and requiring increased dermal coverage, such as the use of gloves and/or long sleeves. Decreasing the HMA application temperature may produce additional reductions in dermal exposures to PACs.

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