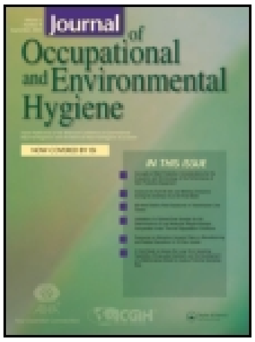

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Dermal Exposure and Urinary 1-Hydroxypyrene among Asphalt Roofing Workers

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The primary objective of this study was to identify significant determinants of dermal exposure to polycyclic aromatic compounds (PACs) among asphalt roofing workers and use urinary 1-hydroxyprene (1-OHP) measurements to evaluate the effect of dermal exposure on total absorbed dose. The study population included 26 asphalt roofing workers who performed three primary tasks: tearing off old roofs (tear-off), putting down new roofs (put-down), and operating the kettle at ground level (kettle). During multiple consecutive work shifts (90 workerdays), dermal patch samples were collected from the underside of each worker's wrists and were analyzed for PACs, pyrene, and benzo(a)pyrene (BAP). During the same work week, urine samples were collected at pre-shift, post-shift, and bedtime each day and were analyzed for 1-OHP (205 urine samples). Linear mixed effects models were used to evaluate the dermal measurements for the purpose of identifying important determinants of exposure, and to evaluate urinary 1-OHP measurements for the purpose of identifying important determinants of total absorbed dose. Dermal exposures to PAC, pyrene, and BAP were found to vary significantly by roofing task (tear-off > put-down > kettle) and by the presence of an old coal tar pitch roof (pitch > no pitch). For each of the three analytes, the adjusted mean dermal exposures associated with tear-off (812 ng PAC/cm^2, 14.9 ng pyrene/cm^2, 4.5 ng BAP/cm^2) were approximately four times higher than exposures associated with operating the kettle (181 ng PAC/cm^2, 4.1 ng pyrene/cm^2, 1.1 ng BAP/cm^2). Exposure to coal tar pitch was associated with a 6-fold increase in PAC exposure ($p = 0.0005$), an 8-fold increase in pyrene exposure ($p < 0.0001$), and a 35-fold increase in BAP exposure ($p < 0.0001$). Similarly, urinary 1-OHP levels were found to be significantly higher on days when an old pitch roof was removed, accounting for a 3.7-fold difference at pre-shift ($p = 0.01$), a 5.0-fold difference at post-shift ($p = 0.004$), and a 7.2-fold difference at bedtime ($p = 0.002$). The pyrene measurements obtained during the work shift were found to be strongly correlated with urinary 1-OHP measurements obtained at the end of that shift ($r = 0.8$, $p < 0.001$) as well as at bedtime ($r = 0.7$, $p < 0.001$). Ultimately, the results of a distributed lag model indicated that dermal exposure during the preceding 40 hours had a statistically significant effect on urinary 1-OHP.

The presence of coal tar pitch was the primary determinant of dermal exposure, particularly for exposure to BAP. However, the task-based differences that were observed while controlling for pitch suggest that exposure to asphalt also contributes to dermal exposures. We found that dermal exposure was a significant determinant of total absorbed dose, suggesting that control strategies aimed at reducing occupational exposure to PACs should include an effort to minimize dermal exposure.

Keywords asphalt, roofing, dermal, biomarkers, exposure

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INTRODUCTION

The U.S. roofing industry comprises employs approximately 200,000 workers who are involved in the removal and application of roofs, a process that can include exposures to asphalt and coal tar pitch.[(1)] Accordingly, roofers are exposed to complex mixtures of polycyclic aromatic compounds (PACs) that include unsubstituted polycyclic aromatic hydrocarbons (PAHs), substituted PAHs, and PAH heterocyclic derivatives.[(2)] The United States National Toxicology Program lists 15 of these compounds as reasonably anticipated to be human carcinogens (NTP, 1998), including benzo(a)pyrene (BAP). In fact, numerous epidemiological studies have described an excess risk of cancer (lung, stomach, bladder, leukemia, and non-melanoma skin cancer) among asphalt-exposed workers; however, the relationship between occupational asphalt exposure and cancer risk remains unclear.[(1,3–5)]

Upon reviewing the available information on human health effects of asphalt exposure, NIOSH concluded that epidemiologic studies of asphalt roofers have generally demonstrated an excess number of lung cancer cases; however, the extent to which the excess cancer risk may be attributed to asphalt exposure was considered to be uncertain in view of the potential confounding by coal tar pitch.[6] Coal tar pitch is a coal-derived product that contains much higher PAC levels than petroleum-derived asphalt and is a well-established carcinogen.[7−9] In the Northeast United States, many roofs on schools, municipal buildings, and commercial structures that were made with coal tar pitch 20 to 40 years ago are still in place. Accordingly, roofers often have exposure to coal tar pitch in the process of removing an old roof so that it can be replaced with a new asphalt roof.

The current study was designed to address questions about the importance of dermal exposure to asphalt and assess the potential impact of exposure to coal tar pitch. Accordingly, dermal exposure measurements and urinary 1-OHP measurements were collected from a population of asphalt roofers over multiple consecutive work shifts. Historically, the assessment of occupational exposure to PAHs has relied primarily on air monitoring; however, there is increasing evidence that dermal contact may be the primary route of PAH exposure in some occupational settings.[10−15] The study was conducted concurrently with researchers from NIOSH who evaluated inhalation exposure, DNA strand breaks, and oxidative damage in the same population of roofers, the results of which are presented separately.[16]

The primary objectives of this study were to:

1) characterize dermal exposure to PACs, pyrene, and BAP over multiple consecutive work shifts;
2) characterize urinary 1-OHP as a measure of total absorbed dose during the same period;
3) identify significant determinants of dermal exposure and total absorbed dose, such as task and coal tar pitch;
4) evaluate the relationship between dermal pyrene exposure and urinary 1-OHP.

METHODS

Study Design

The study population included 26 roofers from four companies who were surveyed from April to September 1998 in Ohio and Kentucky. All participants were male, worked in five different crews, and included 16 smokers and 10 nonsmokers. Written and informed consent was obtained from each participant prior to sampling, and all sampling was conducted in accordance with a standardized human subjects protocol that was approved by the Institutional Review Board at the Harvard School of Public Health.

Each of the five crews consisted of one to eight workers who performed two different jobs: roof workers and kettlemen. The majority of the workers were roof workers (21 workers) who worked primarily on the rooftops while performing two major tasks: tearing off old roofs (tear-off) and putting down new asphalt roofs (put-down). Tear-off was typically performed in the morning followed by put-down in the afternoon; however, during a few days of sampling tear-off and put-down were performed intermittently throughout the day. Additionally, there were a few days when tear-off was not necessary because the building was new such that put-down was the only job conducted. The same roofers who conducted tear-off on a given day also put down new roofs on the same day. They were visibly exposed to large quantities of dust and/or asphalt fume while performing both tasks.

Each crew had one kettleman (five workers) who remained on the ground level while operating the kettle, a device used to heat the asphalt to the desired temperature. Kettlemen had visible exposure to asphalt fume throughout the day, most intensely when adding solid asphalt to the kettle or skimming the molten material. The asphalt types used during this study included Trumbull, Type III Steep ASTM D312-95A (Owens Corning, Toledo OH, USA) and Type IV, 210/225 extra steep roofing asphalt ASTM D312-84 (Sprilast, Arkadelphia AK, USA). In addition to working directly with hot asphalt, the kettlemen regularly cleaned inside and around the dumpsters that contained discarded roof material, a process that often included climbing inside dumpsters to rearrange debris.

Dermal patch samples and urine samples were obtained from all 26 workers over two to five consecutive days. Due to the repeated measures design, a total of 90 "workerdays" were evaluated in the study. Observation forms were used by investigators to record the tasks performed by each worker as well as the characteristics of each jobsite, i.e., weather conditions, asphalt type, and whether the roof being torn off contained coal tar pitch. Questionnaires were used to obtain information about smoking habits, consumption of grilled and fried meats, and personal characteristics.

Though air samples were collected by NIOSH researchers and analyzed for total particulate, benzene-soluble fraction, and PACs, these data have been published previously and did not include an assessment of pyrene in air; accordingly, the air samples are not summarized as part of this assessment of dermal exposure.[16]

Dermal Sampling and Analysis

Dermal patches were attached to the underside of each wrist and generally resulted in the collection of two samples (i.e., left and right wrist) per worker-day; however, whenever possible, a separate set of dermal patches was used during tear-off and put-down to allow for the assessment of task-specific exposures. Of the 71 workerdays among roof workers, 41 workerdays included separate sets of measurements for tear-off (morning) and put-down (afternoon), 15 workerdays included full-shift samples during put-down only, and the remaining 15 workerdays included full-shift samples in which tear-off and put-down were conducted intermittently throughout the day such that task-specific sampling was not possible. All measurements collected from kettlemen represent full-shift exposures.

Complete descriptions of the dermal sampling and analytical methods are provided elsewhere.[17] Briefly, each dermal patch consisted of soft polypropylene filter (Gelman Sciences, 47-mm diameter, 10-μm pore size) embedded in a flexible adhesive bandage that provided an effective surface area of 8.71 cm^2. Following sample collection, the dermal patches were placed in foil-wrapped petri dishes, transported in coolers, and stored at −20°C until analysis. The dermal patch samples were extracted using DMSO and analyzed for polycyclic aromatic compounds (PACs), pyrene, and benzo(a)pyrene (BAP) via high-pressure liquid chromatography (HPLC; Hewlett-Packard Agilent Model#1100).[17]

In cases where the mean field blank amounts were significantly different from zero ($\alpha = 0.05$), the corresponding data were corrected by subtracting the mean field blank amounts from the sample amounts. Method limits of detection (LOD) were estimated for PACs (LOD = 25 ng/cm^2), pyrene (LOD = 2.4 ng/cm^2), and BAP (LOD = 0.5 ng/cm^2) using three times the standard deviation of the field blanks. Using the PAH QTM Standard, approximately 91 percent of the four-ring and larger PAHs were retained in the DMSO layer after partitioning with hexane, with a pooled coefficient of variation of less than one percent.

Urine Sampling and Analysis

Three urine samples (pre-shift, post-shift, and bedtime) were collected from each worker on each day, resulting in the collection of 205 total urine samples (4 to 12 urine samples from each of 26 workers). The first sample was collected at pre-shift on Monday morning and there were approximately 8 hours between each sample. All urine samples were collected in sterilized polypropylene specimen containers, transported in coolers, and then frozen at −20°C until analysis. Urine samples were analyzed for 1-hydroxypyrene (1-OHP) by HPLC, a complete description of which is provided elsewhere.[11]

The method was evaluated by collecting urine from non-exposed individuals (non-smoking graduate students), pooling and splitting it into three one-liter amber glass jars, and spiking one of the jars to 1 ng/ml 1-OHP and another to 10 ng/ml. The third was left unspiked. These control urines were run as regular samples during each batch of worker extractions and analyzed to determine method recovery and precision. The average recovery was 95 and 89 percent for the 1 and 10 ng/ml control urines, respectively, after subtracting the value for the unspiked nonexposed urine. The relative standard deviations were 12 and 14 percent, respectively. All urine samples were analyzed and corrected for creatinine prior to statistical analysis.

Data Analysis

All statistical analyses were conducted using SAS statistical software (SAS Institute, Cary, NC) and significance is reported at the 0.05 level. The dermal exposure data (PAC, pyrene, and BAP) and urinary 1-OHP data were analyzed using descriptive statistics, graphical displays, and linear mixed effects models. Shapiro-Wilks tests and graphical displays indicated that neither the dermal exposure data nor the urinary 1-OHP data were normally distributed; however, a log-transformation of the data did result in approximately normal distributions such that all statistical modeling was conducted using the log-transformed data.

Units for dermal exposure are reported in ng/cm^2. One dermal exposure estimate was calculated for each worker on each sampling day by averaging the left and right wrist measurements. When only one wrist measurement was available, the result from that one sample was used in place of the average. Units for urinary 1-OHP data are reported in μmol/mol creatinine. Since highly dilute or highly concentrated urine samples are generally not suitable for analysis, two urine samples in which the creatinine concentration was either less than 0.3 g/L or greater than 3.0 g/L were excluded from the data analysis.[18]

To evaluate the correlation among PACs, pyrene, and BAP on the dermal patches, traditional methods of estimating correlation coefficients (i.e., Pearson, Spearman) could not be used due to the repeated measures design of the study. Use of these traditional methods would erroneously ignore the number of subjects as the correct sample size while instead using the total number of observations as the incorrect sample size, thereby increasing the degrees of freedom.[19] As an alternative, all correlation coefficients were estimated using linear mixed-effects models as described by Hamlett *et al.* (2003). This approach was also used to evaluate the correlation between pyrene exposure measurements and urinary 1-OHP measurements.

Linear mixed-effects models were also used to analyze the dermal exposure data and evaluate potential determinants of exposure such as task, coal tar pitch, crew, relative humidity, and wind speed. The repeated measures design and use of linear mixed-effects models allowed for evaluation of the fixed effects while estimating between- and within-worker variation. The models used to evaluate dermal exposure can be described as follows:

$$Y_{ijkl} = \ln(X_{ijkl}) = \beta_0 + \beta_{1k}TASK_{ijk} + \beta_{2l}CREW_{ijl} \\ + \beta_3 PITCH_{ij} + \beta_4 RH_{ij} \beta_5 WIND_{ij} + b_i + \varepsilon_{ijkl}$$

where X_{ijkl} represents the exposure level of the i-th worker on the j-th day, and Y_{ijkl} is the natural logarithm of measurement X_{ijkl}. The β's in the model represent the fixed effects for each of the covariates where k={tear-off, put-down, kettle} and l={crews A through E}. Models were fit using a compound symmetry covariance matrix.

A polynomial distributed lag model was used to evaluate the possibility that each urinary 1-OHP measurement is not only influenced by the most recent dermal pyrene exposure, but also by exposures that occurred during previous exposure periods. The appeal of the polynomial distributed lag model is that it is highly flexible and allows for a wide variety of distributed lag effects (increasing, decreasing, or a combination thereof), thus allowing the data to dictate the pattern while reducing the high degree of collinearity that would result from using an unrestricted lag model.[11,20,21]

The distributed lag model required some assumptions regarding dermal exposure that occurred away from work. Though dermal measurements were obtained during the 8-hour work shifts (corresponding to the time periods preceding the post-shift urine samples), exposure measurements were not available during the remaining 16 hours of the day (corresponding to bedtime and pre-shift samples). Since the

TABLE I. Summary Statistics for Dermal Exposure Data

	PAC[A]			Pyrene[B]			Benzo(a)pyrene[C]		
Job/Task	n	GM(GSD) ng/cm²	Range ng/cm²	n	GM(GSD) ng/cm²	Range ng/cm²	n	GM(GSD) ng/cm²	Range ng/cm²
Roof workers[D]	71	898 (4.5)	48–34,014	71	11.0 (5.5)	<2.4–221	71	3.3 (12)	<0.5–59
Tear-off	41	886 (4.6)	49–33,538	41	11.5 (4.8)	<2.4–168	41	4.6 (5.8)	<0.5–84
Put-down	56	344 (3.6)	48–21,437	55	3.8 (5.4)	<2.4–150	54	1.0 (11)	<0.5–59
Kettlemen	19	299 (3.7)	40–4,558	19	4.5 (5.5)	<2.4–34	18	0.9 (18)	<0.5–20

[A] Method limit of detection for PACs: LOD = 25 ng/cm².
[B] Method limit of detection for pyrene: LOD = 2.4 ng/cm².
[C] Method limit of detection for benzo(a)pyrene: LOD = 0.5 ng/cm².
[D] Roof worker data represent full-shift exposures, wheres the tear-off and put-down data represent half-shift exposures.

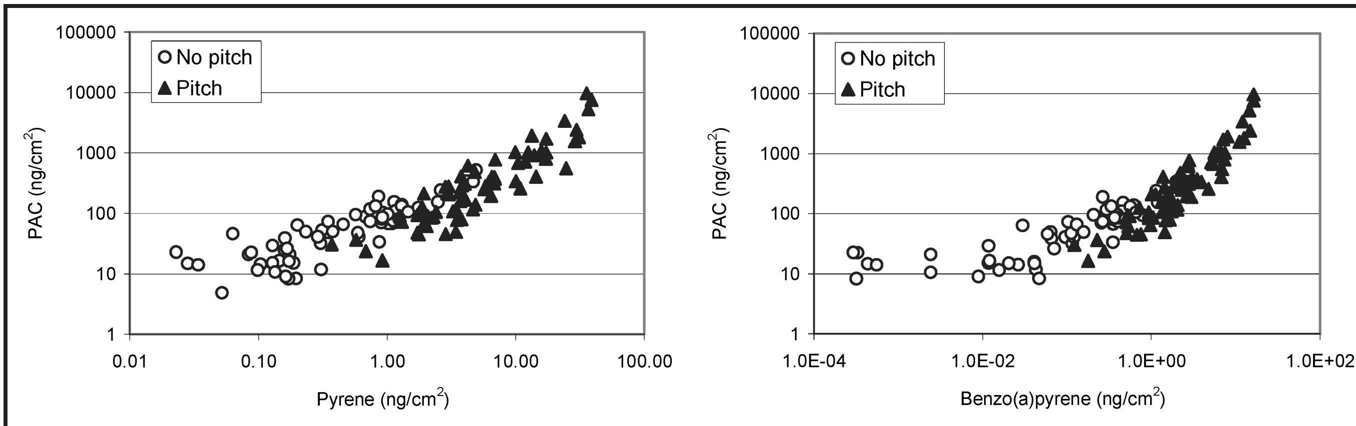
workers did not have occupational exposure during this time, dermal exposures to pyrene were assumed to be zero. Similarly, since the first urine sample was always collected on Monday morning, the same rationale was used to assume zero exposure during the 56 hours prior to collection of the Monday morning pre-shift sample (representing the most recent exposure period and 6 lagged exposure periods during the weekend). Since multiple urine samples were collected from each worker, linear mixed-effects models were ultimately used to evaluate the distributed lag effect of dermal pyrene exposure.

RESULTS

Table I presents the summary statistics for the dermal exposure data, summarizing the PAC, pyrene, and BAP data by job and task. Because the data were not normally distributed, the geometric mean, geometric standard deviation, and range were used to describe the distribution. Dermal exposures among roof workers were significantly higher than among kettlemen for PAC ($p < 0.001$), pyrene ($p = 0.001$), and BAP ($p = 0.004$).

Figure 1 presents the correlation of dermal PAC measurements with pyrene (Figure 1a) and BAP (Figure 1b) measurements obtained from the same worker on the same day. A strong correlation was observed between PAC and pyrene ($r = 0.9$, $p < 0.001$) and between PAC and BAP ($r = 0.8$, $p < 0.001$). Additionally, the data in the figures are stratified according to whether the old roof being removed contained coal tar pitch (i.e., pitch versus no pitch). It is clear from both figures that dermal exposures to PAC, pyrene, and BAP were highest on days when an old pitch roof was removed.

Table II presents the parameter estimates and p-values for all variables in the three models evaluating determinants of dermal exposure to PAC, pyrene, and BAP. The models evaluated the fixed effects of five variables: task, a categorical variable consisting of three levels (tear-off, put-down, kettle); crew, a categorical variable consisting of five levels (crews a through e); pitch, a dichotomous variable indicating whether the old roof being torn off contained coal tar pitch; relative humidity, a continuous variable with values ranging from 65% to 100%; and wind speed, dichotomized as "high wind" and "low wind" using the average wind speed of 7.3 miles per hour as a cut point.



a) PAC vs pyrene (r=0.9, p<0.001)

b) PAC vs BAP (r=0.8, p<0.001)

FIGURE 1. Correlation between dermal PAC with a) pyrene and b) BAP

TABLE II. Results of Final Models Evaluating PAC, Pyrene, and BAP Exposure

	PAC		Pyrene		Benzo(a)pyrene	
Parameters	Estimate (SE)	p-values	Estimate (SE)	p-values	Estimate (SE)	p-values
Fixed effects						
Intercept	5.4 (1.3)		1.2 (1.2)		−3.1 (1.70)	
Task:		0.0008		0.0006		0.0009
Kettleman	−1.5 (0.4)		−1.3 (0.3)		−1.4 (0.40)	
Put-down	−0.7 (0.2)		−0.9 (0.2)		−0.8 (0.30)	
Tear-off	Ref		Ref		Ref	
Pitch:		0.0005		<0.0001		<0.0001
Yes	1.7 (0.3)		2.1 (0.3)		3.6 (0.5)	
No	Ref		Ref		Ref	
Crew:		0.9^{A}		0.06^{B}		0.007
A	0.7 (0.7)		0.7 (0.7)		1.8 (0.9)	
B	0.5 (0..8)		1.1 (0.7)		2.9 (1.0)	
C	0.5 (0.7)		−0.2 (0.7)		0.5 (0.9)	
D	0.2 (0.7)		−0.7(0.7)		0.9 (0.9)	
E	Ref		Ref		Ref	
Wind Speed:						
High	0.03 0.4	0.9^{A}	−0.8 (0.4)	0.03	−1.7 (0.5)	0.001
Low	Ref		Ref		Ref	
Relative Humidity:	2.2 1.5	0.2^{A}	2.9 (1.4)	0.05	6.3 (2.1)	0.003
Random Effects						
σ^2_{Bw} (Full model)^{C}	0.2 (0.1)		0.2 (0.1)		0.2 (0.2)	
σ^2_{ww} (Full model)^{C}	0.9 (0.1)		0.8 (0.1)		1.8 (0.3)	
σ^2_{Bw} (Intercept only)^{D}	0.8 (0.3)		1.4 (0.5)		1.9 (0.9)	
σ^2_{ww} (Intercept only)^{D}	1.4 (0.2)		1.6 (0.2)		3.9 (0.6)	

[A] Not significant in model but retained for consistency.
[B] Marginally significant and retained in model.
[C] Between-worker (σ^2Bw) and within-worker (σ^2ww) variance estimates from full model.
[D] Between-worker (σ^2BW) and within-worker (σ^2ww) variance estimates from intercept-only model.

Task (tear-off > put-down > kettle) and coal tar pitch (pitch > no pitch) were found to be significant determinants of dermal exposure to PAC, pyrene, and BAP. The adjusted mean dermal exposures associated with tear-off (812 ng PAC/cm^2, 14.9 ng pyrene/cm^2, 4.5 ng BAP/cm^2) were approximately four times higher than exposures associated with operating the kettle (181 ng PAC/cm^2, 4.1 ng pyrene/cm^2, 1.1 ng BAP/cm^2). Similarly, dermal exposures were significantly higher on days when the old roof being torn off contained coal tar pitch; however, the magnitude of the effect was different for each analyte such that pitch was associated with a 5.5-fold increase in PAC exposure ($p = 0.0005$), an 8.2-fold increase in pyrene exposure ($p < 0.0001$), and a 35-fold increase in BAP exposure ($p < 0.0001$). Of further interest is the fact that the effect of pitch did not vary by task, i.e., the removal of an old pitch roof not only increased exposures during tear-off but also during put-down and kettle operation.

The variables crew, wind speed, and relative humidity were also found to be significant determinants of dermal exposure to pyrene and BAP but not to PAC; however, since the inclusion of these variables in the PAC model did not change the results for task or pitch, these variables were retained for consistency such that the final PAC model explained 75% of the between-worker variability and 36% of the within-worker variability. Compared to days with low wind speed, high wind speed was associated with lower dermal exposures to pyrene (2.2-fold, $p = 0.03$) and BAP (5.5-fold, $p = 0.001$) while increasing relative humidity was associated with higher levels of dermal exposure to pyrene ($p = 0.05$) and BAP ($p = 0.003$). The final pyrene model explained 86% of the between-worker variability and 50% of the within-worker variability, whereas the final BAP model explained 89% of the between-worker variability and 54% of the within-worker variability.

Table III presents the summary statistics for the pre-shift, post-shift, and bedtime urinary 1-OHP data stratified by job and pitch. Analyzed using a linear mixed effects model, urinary 1-OHP levels were found to be significantly higher on days when an old pitch roof was removed, accounting for a 3.7-fold difference at pre-shift ($p = 0.01$), a 5.0-fold difference at post-shift ($p = 0.004$), and a 7.2-fold difference at bedtime

TABLE III. Summary Statistics for Urinary 1-Hydroxvpyrene (umol/mol Creatinine)

	Start of Week[A]		Pre-Shift[B]		Post-Shift		Bedtime	
Job/Pitch	**n**	**GM(GSD)**	**n**	**GM(GSD)**	**n**	**GM(GSD)**	**n**	**GM(GSD)**
Job								
Roof workers	15	0.4 (2.7)	43	1.0 (3.0)	49	1.3(3.0)	52	1.5 (3.3)
Kettlemen	5	0.2 (3.1)	11	0.5 (3.0)	14	0.6 (3.7)	16	0.6 (3.7)
Coal tar pitch								
Pitch	–	– –	25	2.1 (2.2)	30	2.9 (2.1)	30	3.7 (2.3)
No pitch	20	0.3 (2.8)	29	0.4 (2.0)	33	0.4 (2.3)	38	0.5 (2.0)

[A] Pre-shift samples collected Monday morning following a weekend away from work.
[B] Pre-shift samples collected throughout the workweek (excluding Mondays).

(p = 0.002). Figure 2 presents the correlation between dermal pyrene exposure and urinary 1-OHP measurements obtained at (a) post-shift and (b) bedtime on the same day. The pyrene measurements obtained during the work shift were found to be strongly correlated with urinary 1-OHP measurements obtained at the end of that shift ($r = 0.8$, $p < 0.001$). The same pyrene measurements were also found to be strongly associated with urinary 1-OHP measurements obtained at bedtime (r = = 0.7, $p < 0.001$), approximately eight hours following the end of the work shift. The data shown in Figures 2a and 2b are further stratified by pitch, clearly indicating that the highest pyrene and urinary 1-OHP measurements were obtained on days when an old pitch roof was removed.

Table IV presents the results of the linear mixed-effects model evaluating the distributed lag effects of dermal pyrene exposure, while controlling for age, body mass index, and smoking status. The effect of pyrene exposure was estimated for the most recent exposure period (lag 0, 0 to 8 hours prior to urine collection) as well as each of six preceding exposure periods (lags 1 to 6, 8 to 56 hours prior to urine collection). The total effect of pyrene exposure was calculated by summing the effect estimates for each of the seven exposure periods (lags 0 through 6).

As shown in Table IV, urinary 1-OHP was increased due to dermal exposure that occurred during the preceding 56 hours, though the effect estimates were statistically significant for only the first 40 hours. The largest effect of dermal exposure was observed for the 16- to 24-hour period prior to the collection of each urine sample. Age and smoking status were both found to be positively and significantly associated with urinary 1-OHP, though other factors such as the consumption of grilled food and the use of dandruff shampoo were not significant determinants of urinary 1-OHP and were excluded from the final model.

DISCUSSION

The roofing workers evaluated in this study were exposed to PACs from three sources: asphalt, while tearing off old asphalt roofs and putting down new asphalt roofs; coal tar pitch, while tearing off old coal tar pitch roofs; and cigarette

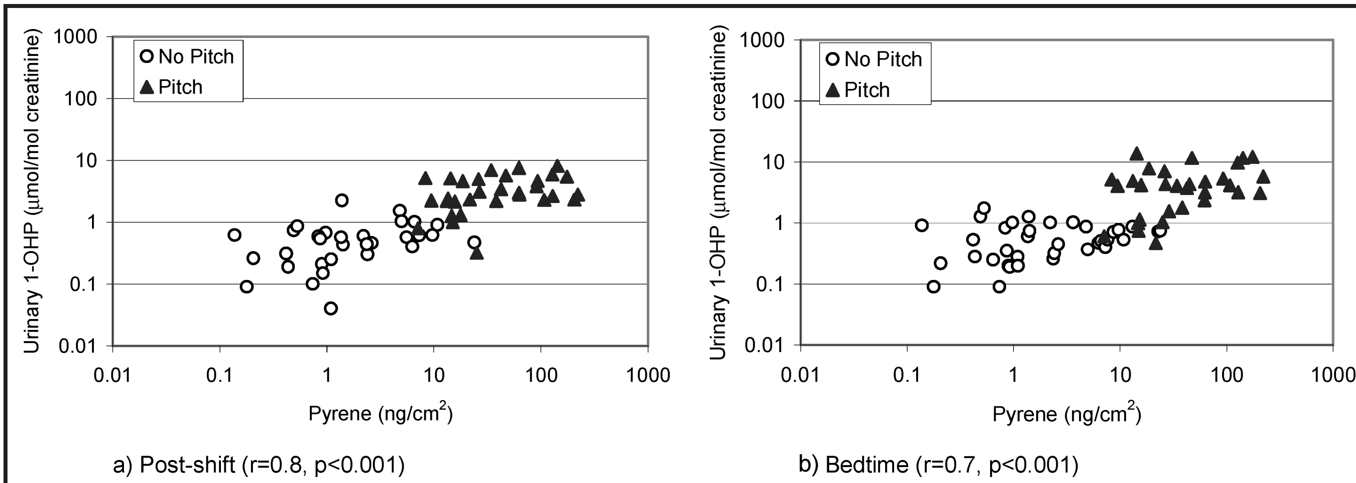


FIGURE 2. Correlation between dermal pyrene exposer and urinary 1-OHP obtained at a) post-shift and b) bedtime

TABLE IV. Results of Distributed Lag Models Evaluating Urinary 1-OHP Among Roofers (ln μmol/mol Creatinine)

Term	Lag (i)	Effect (SE)
Intercept Dermal:		1.5 (0.9)
0–8 hours	0	0.009 (0.002)
8–16 hours	1	0.073 (0.002)
16–24 hours	2	0.014 (0.002)
24–32 hours	3	0.012 (0.002)
32–40 hours	4	0.009 (0.003)
40–48 hours	5	0.005 (0.003)
48–56 hours	6	0.001 (0.003)
Total		0.063 (0.009)
Covariates:		
Age		0.04 (0.01)
BMI		0.01 (0.03)
Smoker		0.5 (0.2)

smoke (16 smokers). Our results indicate that roofing task (tear-off>put-down>kettle) and coal tar pitch (old pitch roof>old asphalt roof) are key factors that influence dermal exposure to PACs, pyrene, and BAP. Our results also indicate that dermal exposure to pyrene is strongly associated with urinary 1-OHP, which provides a useful measure of total absorbed dose. These findings add to the increasing evidence that dermal exposure is an important route of occupational exposure to PACs.[10−14,22,23]

Dermal Assessment

The dermal measurements represent the amount of PAC that was deposited per square centimeter of exposed skin at the wrist. As such, these measurements are not intended to represent total dermal exposure and may not be representative of exposures at other parts of the body. However, measuring dermal exposure at the wrist provides a consistent measure that is useful for evaluating determinants of exposure.

While the approximate 4-fold effect of roofing task was consistent for PAC, pyrene, and BAP, the effect of coal tar pitch was considerably different for each of the three analytes. Most notably, the tear-off of an old coal tar pitch roof accounted for a 35-fold increase in BAP exposure, yielding adjusted mean exposure estimates of 0.4 ng BAP/cm^2 for workerdays without pitch exposure and 12.8 ng BAP/cm^2 for workerdays with pitch exposure. The effect of pitch on BAP exposure was considerably higher than for pyrene (8.2-fold) or PAC (5.5-fold), suggesting that exposure to coal tar pitch may affect exposure to carcinogenic PACs more than other PAC components.

Surprisingly, a similar effect of pitch was observed for all three roofing tasks. We had hypothesized that the presence of an old pitch roof would increase exposures associated with tear-off, but would be less likely to affect exposures associated with put-down or operating the kettle; however, on days when an old pitch roof was removed, the exposures associated with put-down and kettle operation were also elevated. Pitch-related exposures during put-down likely resulted from the presence of pitch dust on the jobsite as well as on clothing, whereas pitch-related exposures among kettlemen likely resulted from the process of cleaning up around the dumpster. It seems likely that clothing contaminated with coal tar pitch could continue to be a source of PAC exposure after leaving the jobsite, potentially resulting in residential exposures.

An evaluation of task-base exposures on non-pitch days provides the best estimate of dermal exposures that are likely to be related to asphalt. On workerdays without pitch exposure, the adjusted mean PAC exposures were 340 ng/cm^2 for tear-off, 161 ng/cm^2 for put-down, and 77 ng/cm^2 for kettlemen. These PAC exposures are in a similar range as our previous assessment of asphalt paving workers, in which dermal PAC exposure among laborers ranged from 123 ng/cm^2 for screedmen on days with low levels of recycled asphalt product (RAP) to 417 ng/cm^2 for rakers on days with high levels of RAP. The dermal exposures were highest for tear-off despite the fact that workers typically covered more of their skin during this task by wearing hats, long-sleeved shirts, gloves, and scarves over the mouth and nose. This protection was not worn during put-down or kettle operation, suggesting that the true differences in task-based exposures may be even larger.

Dermal exposure to pyrene and BAP were also found to vary by relative humidity, wind speed, and crew. Relative humidity was positively associated with dermal exposure, possibly due to the decreased evaporation of perspiration from the skin which could increase the potential for dust to adhere to the skin. Wind speed was negatively associated with exposure, which likely provided natural ventilation to the work area and reduced the potential for contact with airborne dust. Since each crew was measured at different worksites and different days, the variable crew could be serving as a surrogate for any number of factors (e.g., work practice, equipment, unmeasured weather conditions, and/or other production characteristics) that have the actual effect on exposure. Regardless of the true cause, the fact that dermal exposures vary by crew and weather conditions underscores the importance of sampling multiple crews on multiple days when characterizing dermal exposure among roofers.

Urinary 1-OHP

Urinary 1-OHP concentrations were significantly higher following exposure to coal tar pitch, as measured at post-shift, bedtime, and pre-shift the following morning. This is consistent with the finding that pitch exposure resulted in an 8.2-fold increase in dermal exposure to pyrene, the parent compound of which 1-OHP is a metabolite. The dermal pyrene measurements obtained during the work shift were found to be strongly correlated with urinary 1-OHP measurements obtained at the end of that shift (post-shift samples) and approximately eight hours later (bedtime samples). However, such as assessment oversimplifies the relationship between exposure and dose since each urinary 1-OHP measurement is likely influenced by exposure that occurred prior to the most recent 8-hour period. Accordingly, the periods of non-exposure in the 16 hours between post-shift and pre-shift the next day are equally important determinants of dose and should also be taken into account.

Accordingly, the use of polynomial distributed lag models allowed for the analysis of 1-OHP levels, while evaluating the effect of dermal exposure during the preceding 56 hours (divided into seven 8-hour time periods). Because the dermal measurements were only obtained during the 8-hour work shifts, this modeling approach required an assumption that dermal exposure was zero during all 8-hour time periods that were spent away from work. This included the 8-hour period between post-shift and bedtime, the 8-hour period between bedtime and pre-shift (the next day), and all six 8-hour time periods during the preceding weekend.

Table IV shows the effect of dermal exposure to pyrene during each lag period. The results indicate that the preceding 56 hours of dermal exposure had a positive effect on urinary 1-OHP, though the effect estimates were significant for only the most recent exposure period and for the first four lag periods (0 to 40 hours). Interestingly, the most recent exposure period (0 to 8 hours) did not have the largest effect on urinary 1-OHP, a finding that is consistent with previous studies and underscores the importance of evaluating lagged exposures.(11,15) The fact that the largest effect estimates were observed for the first two lag periods (8 to 16 hours and 16 to 24 hours) likely reflects the relatively slow absorption of pyrene by dermal contact.

The linear mixed-effects model used to evaluate the lagged exposures also controlled for age, body mass index, and smoking status. While body mass index was not a significant determinant of urinary 1-OHP, a positive association was found for age while urinary 1-OHP levels among smokers were found to be approximately twice as high as among nonsmokers. The 2-fold effect of smoking was consistent with our previous assessment of urinary 1-OHP among asphalt-paving workers.(11)

CONCLUSIONS

In a recent mortality study, the authors stated the need to control airborne exposures to hazardous substances within the roofing industry.(1) The data presented here suggest that dermal exposure should be controlled as well, particularly during the removal of old coal tar pitch roofs. In the absence of coal tar pitch, asphalt-related dermal exposures were comparable to those observed among asphalt paving workers. Accordingly, both coal tar pitch and asphalt contribute to dermal exposure among roofers. Additionally, dermal exposure was found to be strongly correlated with total absorbed dose, with urinary 1-OHP being influenced by dermal exposure that occurred during the preceding 40 hours. These findings are consistent with a growing body of literature suggesting that dermal contact is an important route of occupational exposure to PAHs.

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