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Inhalation and Dermal Exposure among Asphalt Paving Workers

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The primary objective of this study was to identify determinants of inhalation and dermal exposure to polycyclic aromatic compounds (PACs) among asphalt paving workers. The study population included three groups of highway construction workers: 20 asphalt paving workers, as well as 12 millers and 6 roadside construction workers who did not work with hot-mix asphalt. During multiple consecutive work shifts, personal air samples were collected from each worker's breathing zone using a Teflon filter and cassette holder connected in series with an XAD-2 sorbent tube, while dermal patch samples were collected from the underside of each worker's wrist. All exposure samples were analyzed for PACs, pyrene and benzo[*a*]pyrene. Inhalation and dermal PAC exposures were highest among asphalt paving workers. Among paving workers, inhalation and dermal PAC exposures varied significantly by task, crew, recycled asphalt product (RAP) and work rate (inhalation only). Asphalt mix containing high RAP was associated with a 5-fold increase in inhalation PAC exposures and a 2-fold increase in dermal PAC exposure, compared with low RAP mix. The inhalation PAC exposures were consistent with the workers' proximity to the primary source of asphalt fume (paver operators > screedmen > rakers > roller operators), such that the adjusted mean exposures among paver operators (5.0 μg/m^3, low RAP; 24 μg/m^3, high RAP) were 12 times higher than among roller operators (0.4 μg/m^3, low RAP; 2.0 μg/m^3, high RAP). The dermal PAC exposures were consistent with the degree to which the workers have actual contact with asphalt-contaminated surfaces (rakers > screedmen > paver operators > roller operators), such that the adjusted mean exposures among rakers (175 ng/cm^2, low RAP; 417 ng/cm^2, high RAP) were approximately 6 times higher than among roller operators (27 ng/cm^2, low RAP; 65 ng/cm^2, high RAP). Paving task, RAP content and crew were also found to be significant determinants of inhalation and dermal exposure to pyrene. The effect of RAP content, as well as the fact that exposures were higher among paving workers than among millers and roadside construction workers, suggests that the PAC and pyrene exposures experienced by these paving workers were asphalt-related.

Keywords: asphalt; inhalation; dermal; paving; polycyclic aromatic hydrocarbons

INTRODUCTION

Asphalt (or bitumen) is a dark, semi-solid residual that results from the non-destructive distillation of crude petroleum oil and is widely used as an industrial material (Gamble *et al*., 1999). The annual production of hot-mix asphalt amounts to ~267 million tons in Western Europe and ~440 million tons in the USA (Partanen and Boffetta, 1994), while the road-paving industry employs ~300 000 workers in the United States and accounts for 87% of domestic asphalt production (Asphalt Institute, 1990). The American Conference of Governmental Industrial Hygienists (ACGIH) currently recommends a threshold limit value for asphalt fumes of 0.5 mg/m^3 (benzene-soluble aerosol), while the National Institute for Occupational Safety and Health (NIOSH) recommends an exposure limit of 5 mg/m^3 (total particulate during any 15 min period). The Occupational Safety and Health Administration (OSHA) currently has no standard for exposure to asphalt fumes.

Asphalt contains a complex mixture of polycyclic aromatic hydrocarbons (PAHs), some of which are either known or suspected to be carcinogenic. Because asphalt workers are occupationally exposed

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to PAHs via inhalation and dermal absorption, the carcinogenic potential of asphalt has been under investigation since the 1960s. 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 (Partanen and Boffetta, 1994; Boffetta *et al*., 1997, 2003). However, the relationship between occupational asphalt exposure and cancer risk remains unclear.

Comprehensive reviews of the available literature have been conducted (IARC, 1985; Chiazze *et al*., 1991; Partanen *et al*., 1995; NIOSH, 2000). These reviews were consistent in their evaluation of existing studies, concluding that the lack of data on exposure and potential confounders (such as smoking and exposure to coal tar) were significant limitations that made it impossible to demonstrate a causal association. Specifically, the existing studies were criticized for weak or absent exposure assessments that lacked quantitative measurements of exposure to asphalt or its constituents.

Occupational exposure to PAHs has been shown to be associated with an excess risk of cancer in other populations such as coke oven (Costantino *et al*., 1995), foundry (Andjelkovich *et al*., 1990) and aluminum production workers (Armstrong *et al*., 1994), and it is possible that random misclassification of exposure has limited the ability of previous studies to detect a positive association between occupational asphalt exposure and cancer. Accordingly, the risk of work-related cancer among asphalt workers continues to be an important and controversial issue.

As an alternative to the more common approach of evaluating inhalation exposure to individual PAHs, this study uses a task-based approach to evaluate both inhalation and dermal exposure to polycyclic aromatic compounds (PACs) among asphalt paving workers. The primary objectives of this study were: (i) to determine whether inhalation and dermal exposures to PACs are higher among asphalt-exposed workers than among non-exposed workers; (ii) to identify job factors (i.e. paving task, percent recycled asphalt, etc.) that affect inhalation and/or dermal exposure to PAC; and (iii) to characterize the relationship between inhalation and dermal exposures.

MATERIALS AND METHODS

Study population

The study population included three groups of highway construction workers: 20 asphalt paving workers, as well as 12 millers and 6 roadside construction workers who did not work with hot-mix asphalt. All participants were male, worked for the same company, and lived in the Greater Boston area. Written and informed consent was obtained from each study subject 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.

The paving workers were exposed to hot-mix asphalt while resurfacing roads. At the job-sites, the asphalt was loaded into the front hopper of a paving machine while the screed (attached to the back of the paving machine) was used to adjust the thickness and width of the applied mix. The specific characteristics of the asphalt varied by job and contained recycled asphalt pavement (RAP) in amounts ranging from 0 to 40% of the total mix. An alternative to using 100% virgin mix, RAP refers to milled material that is generated during the grinding of old roads, transported to the asphalt plant and recycled into a new batch of asphalt.

Each of the three paving crews consisted of six to eight workers who performed four different tasks: paver operators, screedmen, rakers and roller operators. The paver operator sat between the hopper and the screed while controlling the path and speed of the paving machine. Two screedmen stood on a platform behind the screed, one controlling the left side and the other controlling the right side. Two to three rakers worked in close proximity to the back of the screed, using rakes and shovels to fill holes and gaps. One or two rolling machines, each with their own operator, were then used to smooth and compact the laid-down asphalt.

The milling workers and roadside construction workers were evaluated because the location and nature of their work is similar to the paving workers, except that neither group typically works with hot-mix asphalt. The milling workers used a large grinding machine and a smaller trimming machine to remove layers of aged asphalt from existing roads in preparation for resurfacing by the paving crews, whereas the roadside construction workers worked with hand tools while repairing guardrails or installing curbs and sidewalks.

Study design

Personal air and dermal patch samples were collected from 38 workers during full work shifts at job-sites located within 1 h of Boston, MA. In May and June 1999, three days of exposure measurements were obtained from 20 pavers. In May and June 2000, multiple days of exposure measurements were obtained from 12 millers and 6 construction workers. Due to the repeated-measures design, the total number of 'worker-days' evaluated in the exposure assessment includes 60 worker-days for pavers, 39 worker-days for millers and 11 worker-days for construction workers.

Each worker-day of sampling included the collection of personal air samples (particulate and vapor), dermal patch samples and meteorological information. Questionnaire information (including demographic, job characteristics and lifestyle) and detailed observations for each worker were also obtained.

Personal air samples. Personal air samples were collected from each worker in accordance with NIOSH Method 5506 (NIOSH, 1998a). The air sampling system consisted of a Teflon filter and cassette holder to collect particulate PACs, an XAD-2 sorbent tube to collect the vapor phase PACs, and a personal air sampling pump operating at 2 l/min. The 37 mm diameter filter (PTFE-laminated with 2 μm pore size) was placed in a cassette and attached to each worker's lapel near the breathing zone, and the sorbent tube containing XAD-2 was attached inline and downstream from each filter cassette. Flow rates were checked before, during and after sample collection using a calibrated rotameter. Opaque filter cassettes and foil-wrapped sorbent tubes were used to prevent sample degradation from sunlight. Samples were transported in coolers and stored at −20°C.

Dermal patch samples. The dermal patch samples were collected from both wrists of each worker. The dermal sampling method was a modification of the method described by Jongeneelen *et al*. (1988) and Van Rooij *et al*. (1993). A soft polypropylene filter (Gelman Sciences, 47 mm diameter, 10 μm pore size) was attached to an exposure pad to create a dermal patch with an effective surface area of 8.71 cm^2. Using an adhesive backing, the patches were attached to the underside of each wrist and resulted in the collection of two samples per worker-day. Following sample collection, the exposure pads were placed in foil-wrapped Petri dishes, transported in coolers, and stored at −20°C.

Laboratory analysis. The analytical method for measuring PACs was developed using a modified version of NIOSH Method 5800 (NIOSH, 1998b). NIOSH Method 5800 was initially developed because the individual components of asphalt-related PACs cannot be easily separated or quantified (NIOSH, 2000). For the PAC analyses in this study, the excitation and emission wavelengths (excitation 270 nm and emission 415 nm) were optimized to target the four-ring and larger PACs, a range that includes carcinogenic compounds such as benzo[*a*]pyrene (BAP), benzo[*a*]anthracene, benzo[*b*]fluoranthene, chrysene and dibenz[*a*,*h*]anthracene.

Particulate and vapor samples were extracted as follows: (i) 4 ml of hexane were added; (ii) samples were sonicated for 1 h; (iii) 2 ml were syringe-filtered and transferred to a clean tube; and (iv) 2 ml DMSO were added for a final extraction volume of 4 ml. Dermal patch samples were: (i) cut with a 33.3 mm punch and each cut out transferred to a labeled culture tube; (ii) 4 ml of DMSO were added; (iii) tubes were capped and sonicated for 1 h; (iv) 2 ml of the extract were transferred to a clean culture tube; and (v) 2 ml of hexane were added for a final extraction volume of 4 ml.

For all air and dermal samples, the extracted mixtures were 'tumbled' overnight, layers were transferred to separate tubes and the DMSO layer was analyzed on the high-pressure liquid chromatograph (HPLC; Hewlett-Packard Agilent Model #1100) for PACs, pyrene and BAP. Known amounts of PAC, pyrene and BAP were diluted to prepare concentrations of 0, 10, 30, 100, 200, 400 and 800 ng/ml. The resulting data produced linear standard curves with intercepts close to 0.0, and *r*-squared values of ∼1 (0.97–0.99).

Estimated as three times the standard deviation of the field blanks, method limits of detection (LOD) were calculated for PACs (LOD_{air} = 0.2 μg/m^3, LOD_{dermal} = 38 ng/cm^2), pyrene (LOD_{air} = 0.01 μg/m^3, LOD_{dermal} = 2.6 ng/cm^2) and BAP (LOD_{air} = 0.01 μg/m^3, LOD_{dermal} = 0.6 ng/cm^2). The total number of field blanks equaled 17% of air samples and 18% of dermal samples. 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.

Data analysis

The air and dermal exposure data were analyzed using descriptive statistics, graphic displays, correlation coefficients and linear mixed-effects models. Shapiro–Wilks' tests and graphic displays indicated that the air and dermal data were not normally distributed; however, a log-transformation of the data did result in an approximately normal distribution. Accordingly, all statistical analyses were conducted using the log-transformed air and dermal data. All statistical analyses were conducted using SAS statistical software, and statistical significance is reported at the 0.05 level.

One total air exposure estimate was calculated for each worker on each sampling day by adding the particulate and vapor measurements. Similarly, 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. Values less than detection limits were included in analyses as one-half the detection limit.

Six individual dermal samples collected from two workers were excluded from the analysis because there was sufficient evidence to suspect that the samples had been contaminated with diesel fuel. Both workers were laborers on the same paving crew

who were observed to be deliberately contaminating the dermal patches with fuel. Furthermore, these six patches were visibly discolored due to saturation, and the resulting PAC measurements were orders of magnitude higher than all other samples.

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 (Hamlett *et al*., 2003). As an alternative, all correlation coefficients were estimated using linear mixed-effects models as described by Hamlett *et al*. (2003).

Linear mixed-effects models were also used to analyze the inhalation and dermal PAC data by job, by paving task, and to evaluate the other potentially important job factors such as RAP, crew, work rate (tons of asphalt applied per hour) and ambient temperature (°F). 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 inhalation and dermal PAC exposure among the asphalt paving workers can be described as follows:

$$\begin{aligned} Y_{ijkl} &= \ln(X_{ijkl}) \\ &= \beta_0 + \beta_{1k}\mathrm{TASK}_{ijk} \\ &\quad + \beta_{2l}\mathrm{CREW}_{ijl} + \beta_3\mathrm{RAP}_{ij} + \beta_4\mathrm{WORKRATE}_{ij} \\ &\quad + \beta_4\mathrm{TEMP}_{ij} + b_i + \varepsilon_{ijkl} \end{aligned}$$

where X_{ijkl} represents the exposure level of the ith paving worker on the jth day, and Y_{ijkl} is the natural logarithm of measurement X_{ijkl}. The βs in the paving model represent the fixed effects for each of the covariates where k = {paver operator, roller operator, raker, screedman} and l = {crew A, crew B, crew C}. Models were fitted using a compound symmetry covariance matrix and different combinations of fixed effects were evaluated using maximum likelihood (ML) estimation (Wolfinger, 1993). The different versions of the models were compared using Akaike's Information Criteria diagnostic values, and the final model was fit using restricted maximum likelihood (REML) estimation.

RESULTS

Table 1 presents the summary statistics for the inhalation and dermal exposure data, summarizing the PAC, pyrene and BAP data for paving workers, milling workers and roadside construction workers. Because the data were not normally distributed, the geometric mean, geometric standard deviation and range were used to describe the distribution of the data. Among all workers, BAP was rarely detected above the detection limits in air (LOD = 0.01 μg/m^3) or dermal samples (LOD = 0.6 ng/cm^2).

In air samples collected from paving workers, PACs were detected above the LOD (0.2 μg/m^3) in 97% of the samples. The partitioning of PACs was such that 65% was detected in the particulate phase (filters) and 35% in the vapor phase (XAD tubes). In the same samples, pyrene was detected above the LOD (0.01 μg/m^3) in 98% of samples, and partitioned

Table 1. Summary statistics for inhalation and dermal exposure data

Analyte/job	Inhalation					Dermal				
	n	Detect (%)	GM (μg/m^3)	GSD (μg/m^3)	Range (μg/m^3)	*n*	Detect (%)	GM (ng/cm^2)	GSD (ng/cm^2)	Range (ng/cm^2)
PAC[a]										
Paving	59	97	4.1	3.1	0.3–40	59	68	89	3.1	46–751
Milling	39	100	1.4	2.6	0.3–6.7	39	26	[d]	[d]	43–757
Roadside construction	11	36	[d]	[d]	0.7–1.5	11	55	45	3.1	45–246
Pyrene[b]										
Paving	59	98	0.18	3.5	0.01–1.7	59	61	3.5	2.7	2.7–25
Milling	39	51	[d]	[d]	0.01–0.05	39	8	[d]	[d]	5.3–7.1
Roadside construction	11	9	[d]	[d]	0.01–0.03	11	0	[d]	[d]	<2.6
Benzo[*a*] Pyrene[c]										
Paving	59	20	[d]	[d]	0.01–0.03	59	12	[d]	[d]	0.8–2.5
Milling	39	44	[d]	[d]	0.01–0.03	39	8	[d]	[d]	0.7–1.2
Roadside construction	11	18	[d]	[d]	0.01–0.03	11	27	[d]	[d]	1.0–1.2

[a]Method limits of detection for PACs: LOD_{air} = 0.2 μg/m^3, LOD_{dermal} = 38 ng/cm^2.
[b]Method limits of detection for Pyrene: LOD_{air} = 0.01 μg/m^3, LOD_{dermal} = 2.6 ng/cm^2.
[c]Method limits of detection for BAP: LOD_{air} = 0.01 μg/m^3, LOD_{dermal} = 0.6 ng/cm^2.
[d]Geometric mean (GM) and geometric standard deviation (GSD) not shown due to low level of detection.

such that 24% was detected in the particulate phase and 76% in the vapor phase. Inhalation exposure to PACs was higher among paving workers than among millers ($P = 0.007$) and roadside construction workers ($P < 0.001$).

In dermal samples collected from paving workers, PACs were detected above the LOD (38 ng/cm^2) in 68% of the samples, and the PAC measurements from left and right wrists were strongly correlated ($r = 0.87$, $P < 0.001$). In addition, the results of a Wilcoxon signed-rank test indicated that dermal exposure to PACs was not significantly different between dominant hand and non-dominant hand ($P = 0.2$). Dermal exposure to PAC was higher among paving workers than among millers ($P < 0.001$) and roadside construction workers ($P = 0.09$).

Figure 1 shows the relationship between PACs and pyrene among paving workers. In air samples (Fig. 1a), there was a strong correlation between PAC and pyrene ($r = 0.87$, $P < 0.001$), whereas Fig. 1b shows a weaker correlation between PAC and pyrene in dermal samples ($r = 0.65$, $p = 0.002$). Figure 2 shows the relationship between inhalation exposure and dermal exposure among paving workers. For exposure to PACs (Fig. 2a), the relationship between inhalation and dermal exposure was different among rakers ($r = 0.15$, $P = 0.5$) than among non-rakers ($r = 0.45$, $P = 0.01$). However, for exposure to pyrene (Fig. 2b), the correlation between inhalation and dermal exposure ($r = 0.59$, $P = 0.006$) was stronger than for PAC exposure and did not vary by task.

Table 2 presents the parameter estimates and *P*-values for all variables in the final paving models evaluating inhalation and dermal exposure to PACs and pyrene. The models evaluated the fixed effects of five variables: task, a categorical variable consisting of four levels (paver operator, roller operator, screedman and laborer); crew, a categorical variable consisting of three levels (crew A, crew B and crew C); RAP, a categorical variable that was dichotomized as 'high RAP' and 'low RAP' using the average RAP of 26% as a cut-off point (using the median RAP of 29% would have resulted in identical 'high RAP' and 'low

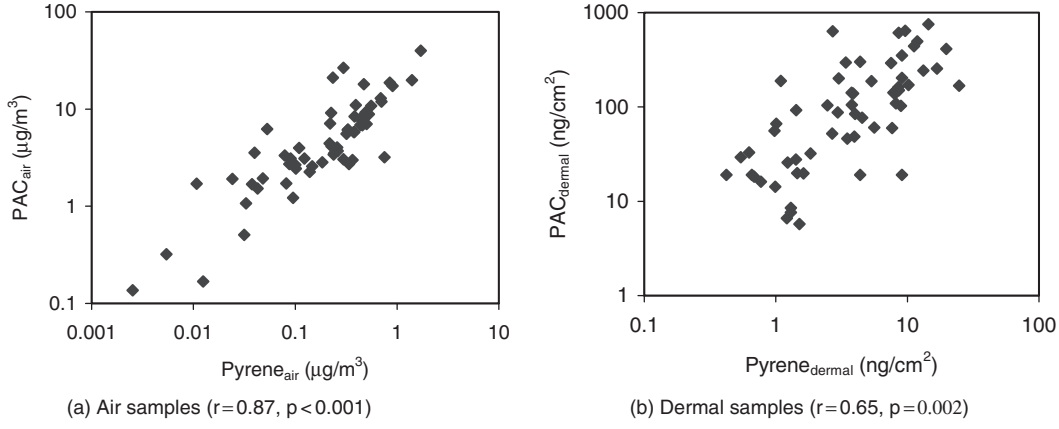

Fig. 1. Correlation between PACs and pyrene among paving workers, evaluated in (a) air samples and (b) dermal samples.

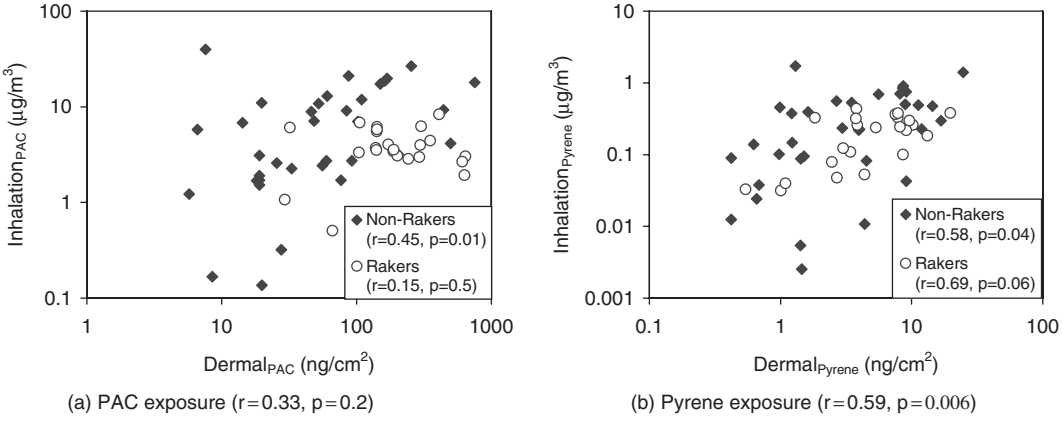

Fig. 2. Correlation between inhalation and dermal measurements among paving workers, evaluated for (a) PAC exposure and (b) pyrene exposure.

Table 2. Results of final models evaluating PAC and pyrene exposure among paving workers

Parameters	PAC exposure models				Pyrene exposure models			
	Inhalation		Dermal		Inhalation		Dermal	
	Parameter Estimates (SE)	*P*-values	Parameter Estimates (SE)	*P*-values	Parameter Estimates (SE)	*P*-values	Parameter Estimates (SE)	*P*-values
Fixed effects								
Intercept	0.09 (0.46)		5.8 (0.63)		−2.0 (0.34)		2.5 (0.63)	
Task		<0.0001		0.003		<0.0001		0.05
Paver operators	0.44 (0.35)		−1.1 (0.52)		0.21 (0.27)		−0.85 (0.45)	
Roller operators	−2.1 (0.33)		−1.5 (0.48)		−2.1 (0.25)		−1.2 (0.42)	
Rakers	−0.70 (0.28)		0.35 (0.40)		−0.78 (0.21)		−0.28 (0.35)	
Screedmen	0.0 (Ref)		0.0 (Ref)		0.0 (Ref)		0.0 (Ref)	
Crew		0.06[a]		0.03		0.06[a]		0.02
A	−0.52 (0.46)		−1.8 (0.64)		−0.82 (0.34)		−2.1 (0.66)	
B	−0.85 (0.36)		−1.5 (0.51)		−0.67 (0.27)		−1.6 (0.49)	
C	0.0 (Ref)		0.0 (Ref)		0.0 (Ref)		0.0 (Ref)	
RAP		0.0005		0.03		<0.0001		0.004
High	1.6 (0.23)		0.87 (0.30)		2.3 (0.17)		1.5	
Low	0.0 (Ref)		0.0 (Ref)		0.0 (Ref)		0.0 (Ref)	
Work rate		0.004		0.2[b]		0.3[b]		0.3[b]
(Tons/h)	0.005 (0.002)		−0.003 (0.002)		0.001 (0.001)		−0.002 (0.002)	
Random effects								
σ^2_{BW} (full model)[c]	0.16 (0.10)		0.36 (0.19)		0.09 (0.05)		0.21 (0.15)	
σ^2_{WW} (full model)[c]	0.23 (0.06)		0.40 (0.09)		0.13 (0.03)		0.50 (0.12)	
σ^2_{BW} (intercept only)[d]	0.73 (0.33)		0.83 (0.32)		0.90 (0.39)		0.24 (0.17)	
σ^2_{WW} (intercept only)[d]	0.63 (0.15)		0.47 (0.11)		0.77 (0.18)		0.74 (0.17)	

[a]Marginally significant and retained in model.
[b]Not significant in model but retained for consistency.
[c]Between-worker (σ^2_{BW}) and within-worker (σ^2_{WW}) variance estimates from full model.
[d]Between-worker (σ^2_{BW}) and within-worker (σ^2_{WW}) variance estimates from intercept-only model.

RAP' categories); work rate, a continuous variable representing the tons of asphalt applied per hour; and ambient temperature, modeled as a continuous variable. Ambient temperature was not found to be a significant determinant of exposure and was excluded from the final models.

For inhalation exposure to PACs among paving workers, the variables task ($P < 0.0001$), RAP ($P = 0.0005$) and work rate ($P = 0.004$) were significant while crew was marginally significant (0.06), such that these variables explained 78% of the between-worker variability and 63% of the within-worker variability. With the exception of work rate, the same variables were found to be significant determinants of inhalation exposure to pyrene, such that the variables in the final pyrene model explained 90% of the between-worker variability and 83% of the within-worker variability. The task-based results for both PACs and pyrene indicated that paver operators were the highest exposed, followed by screedmen, rakers and roller operators. Inhalation exposures to PAC and pyrene were found to be significantly higher when asphalt contained high RAP versus low RAP, and were found to increase as the work rate increased. The variable crew was retained in the model so that the fixed effects of the other job factors could be evaluated while adjusting for crew-based differences.

For dermal exposure to PACs among paving workers, the variables task ($P = 0.003$), RAP ($P = 0.03$) and crew ($P = 0.03$) were significant determinants of exposure, such that these variables explained 57% of the between-worker variability and 15% of the within-worker variability. Task, RAP and crew were also found to be significant determinants of dermal exposure to pyrene, such that the variables in the final pyrene model explained 13% of the between-worker variability and 32% of the within-worker variability. As with the inhalation exposures, dermal exposures to PACs and pyrene were also found to be significantly higher when asphalt contained high RAP versus low RAP. The task-based results for dermal PAC exposure indicated that rakers were the highest exposed, followed by screedmen, paver operators and roller operators. The task-based results for dermal pyrene exposure were slightly different in that the screedmen were the highest exposed, followed by the rakers, paver operators and roller operators.

Table 3. Adjusted mean PAC exposures[a] among paving workers

Task	Inhalation (μg/m^3)		Dermal (ng/cm^2)	
	Low RAP	High RAP	Low RAP	High RAP
Median work rate[b]				
Paver operators	5.0 (2.8–9.1)	24 (13–43)	39 (17–93)	94 (40–221)
Roller operators	0.4 (0.2–0.7)	2.0 (1.2–3.4)	27 (13–59)	65 (30–40)
Rakers	1.6 (1.1–2.4)	7.6 (5–12)	175 (97–316)	417 (231–750)
Screedmen	3.2 (2.0–5.3)	15 (9.4–25)	123 (61–249)	294 (146–593)
Minimum work rate[b]				
Paver operators	3.8 (2–7.17)	18 (9.3–35)	46 (19–112)	109 (43–278)
Roller operators	0.3 (0.2–0.6)	1.5 (0.8–2.7)	32 (14–71)	75 (32–177)
Rakers	1.2 (0.8–1.9)	5.8 (3.5–9.5)	204 (107–388)	485 (241–972)
Screedmen	2.4 (1.4–4.2)	12 (6.6–21)	144 (68–304)	342 (154–757)
Maximum work rate[b]				
Paver operators	11 (5.1–23)	52 (26–103)	26 (9.0–74)	62 (24–160)
Roller operators	0.9 (0.4–1.8)	4.3 (2.3–8.1)	18 (6.7–48)	43 (18–102)
Rakers	3.5 (1.9–6.5)	17 (9.8–28)	115 (49–269)	274 (133–562)
Screedmen	7.0 (3.6–14)	33 (19–60)	81 (32–206)	193 (86–436)

[a]Mean exposure = $e^{[\mu+0.5(\sigma^2_{BW} + \sigma^2_{WW})]}$ where μ is the mean of the logged exposures after adjusting for significant covariates.
[b]Median work rate = 115 tons/h, minimum work rate = 55 tons/h, maximum work rate = 282 tons/h.

Table 3 presents the mean PAC exposure estimates that result from the final models, estimating the inhalation and dermal exposures associated with each paving task while considering the between- and within-worker variability and adjusting for other important determinants of exposure. Since the data analysis was conducted using the normally distributed logged data, the mean exposure concentrations were estimated according to Rappaport *et al.* (1999): $\hat{\mu}_{TASK} = e^{[\hat{\mu}_{TASK} + 0.5(\hat{\sigma}^2_{BW} + \hat{\sigma}^2_{WW})]}$. The task-based inhalation and dermal estimates are stratified by RAP content to demonstrate the extent to which the presence of recycled asphalt increases exposure. Similarly, results are presented at the median (115 tons/h), minimum (55 tons/h) and maximum (282 tons/h) work rate values; however, the effect of work rate was only significant for inhalation exposure to PACs.

DISCUSSION

The primary objective of this exposure assessment was to identify determinants of inhalation and dermal PAC exposure among asphalt paving workers. Because the majority of existing studies focus on inhalation exposure, we collected both dermal and air samples in an effort to obtain a more complete assessment of total exposure. In coke oven workers, there is evidence to suggest that dermal absorption is actually the primary route of exposure to PAHs (Van Rooij *et al.*, 1993). In fact, after evaluating pyrene exposure data (inhalation and dermal) and urinary 1-hydroxypyrene data, Van Rooij *et al.* (1993) estimated that ~75% of the total absorbed dose was attributable to dermal exposure. Accordingly, an assessment of PAH exposure that excludes dermal absorption may underestimate cumulative exposure.

The dermal data are presented in units of ng/cm^2, which represents the average amount of PACs that was deposited per square centimeter of exposed skin (at the wrist) during an 8 h shift. As such, these measurements are not intended to represent total dermal exposure. Also, dermal exposures assessed at the wrist may not be representative of exposures at other parts of the body. For instance, the magnitude of 'hand' exposures would probably be higher than wrists, but the magnitude of 'forearm' or 'neck' exposures would probably be lower than wrists. However, the wrist samples do provide a useful tool for comparing dermal exposures across individual workers and across groups of workers.

Paving task, crew and RAP content were consistently found to be important determinants of exposure in each of the four models evaluating inhalation and dermal exposure to PACs and pyrene. The mean PAC exposure estimates associated with each paving task varied ~12-fold for inhalation exposure and ~6-fold for dermal exposure. The task-based inhalation results for PACs and pyrene were consistent with a task's proximity to the primary source of asphalt fume (paver operators > screedmen > rakers > roller operators), while the task-based dermal results for PACs were consistent with the degree to which each task requires actual contact with asphalt-contaminated surfaces (rakers > screedmen > paver operators > roller operators). The task-based dermal results were similar for pyrene, although exposures among

screedmen were slightly higher than among rakers. In all four models, we found that the lowest exposures were experienced by roller operators, workers who spend the least amount of time near the source and have the least amount of contact with asphalt-contaminated surfaces.

The task-based results for PACs in personal air are considerably higher than those observed by Heikkila *et al*. (2002), a European study in which the arithmetic mean of total PAHs ranged from 2.38 μg/m^3 (roller operators) to 4.28 μg/m^3 (paving operators). The discrepancy is probably due to the fact that Heikkila *et al*. estimated total PAH using the sum of 15 individual unsubstituted PAHs, while the PAC estimates in the present study include all substituted and unsubstituted PACs with at least four rings. Regional differences in asphalt operations may also account for part of the observed differences.

The mean exposure estimates associated with each of the three crews varied ∼2-fold for inhalation exposure (2.2, 3.1 and 5.1 μg/m^3) and ∼6-fold for dermal exposure (36, 49 and 219 ng/cm^2). 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 an actual effect on exposure. Regardless of the true cause, the fact that inhalation and dermal exposures vary by crew suggests that crew is an important factor that should be considered when characterizing PAC exposure among paving workers.

The mean PAC exposure estimates associated with RAP content varied ∼5-fold for inhalation exposure and ∼2-fold for dermal exposure, such that RAP content was a significant determinant of inhalation and dermal exposure. It is unclear whether these increases in exposure actually result from the RAP material or from the higher temperatures required when RAP is added to a new batch of asphalt. The temperature of the asphalt mix was not measured during sampling and therefore could not be evaluated in the exposure models. However, since RAP is specifically a characteristic of the asphalt mix, the fact that RAP content was a significant determinant of exposure in all four exposure models suggests that the PAC and pyrene exposures among paving workers were asphalt-related.

Work rate was calculated as the tons of asphalt applied per hour during a full shift of work in an effort to compare workdays of varying work intensity. Some days were observed to be considerably busier or slower than other days, since the pace of work was often slowed due to the delayed transportation of asphalt mix from the asphalt plant to the jobsite. Increasing work rate was found to increase inhalation exposure to PACs, although work rate was otherwise not a significant determinant of exposure.

The ability to evaluate the effect of ambient temperature on inhalation and dermal exposure was limited by a fairly narrow range of ambient temperature measurements (65–76°F). Although initially included in the exposure models, ambient temperature did not have a significant effect on inhalation or dermal exposures and was excluded from the final models. The temperature of the asphalt would probably have been a more useful measure as a potential determinant of exposure.

Inhalation and dermal exposures to PACs were higher among paving workers than among millers and roadside construction worker. The increased exposure can be attributed to working with hot-mix asphalt, since pavers, millers and roadside construction workers otherwise have a similar potential for exposure to PACs. Millers were probably exposed to PACs during the grinding of old asphalt roads, while roadside construction were probably exposed to PACs in roadside soils while installing curbs, guardrails and sidewalks (Tuhackova *et al*., 2001). However, such exposures may have resulted in under estimating rather than overestimating the portion of exposure that is attributable to working with hot mix asphalt.

The manner in which dermal exposure to asphalt-related PACs occurs is unclear. Dermal exposures may result from direct contact with asphalt-contaminated surfaces, such as hand tools and/or equipment controls, the settling of airborne particles and/or the condensation of vapor. Among paving workers, Burstyn *et al*. (2002) found inconsistent patterns between airborne organic matter and organic matter collected on cotton pads, although it is likely that the sample size ($n = 7$) and the use of different solvents to extract organic matter from dermal pads and filters limited their ability to evaluate the correlation. We found that the correlation between inhalation and dermal PAC exposure was stronger for non-rakers than for rakers, which is probably related to the fact that the rakers have the highest potential for direct contact with asphalt-contaminated surfaces; however, even among non-rakers the correlation was relatively weak ($r = 0.45$, $P = 0.01$). The correlation between inhalation and dermal exposure was stronger for exposure to pyrene ($P = 0.59$, $P = 0.006$), suggesting that dermal exposure may be more likely to be associated with airborne levels for lower molecular weight compounds.

CONCLUSIONS

The task performed by a member of a paving crew was found to be the most important determinant of inhalation and dermal exposure to PACs. The task-based inhalation exposures were consistent with each task's proximity to the primary source of asphalt fume

(the hopper), while the task-based dermal exposures were consistent with the degree to which each task requires actual contact with the asphalt-contaminated surfaces. In addition, inhalation and dermal exposures were significantly higher when the asphalt contained high RAP compared with low RAP, suggesting that the PAC exposures experienced by these paving workers were asphalt related. Similarly, inhalation and dermal PAC exposures were higher among paving workers than among millers and roadside construction workers, further suggesting that the higher PAC exposures were attributable to asphalt.

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