



Statistical Modelling of the Determinants of Historical Exposure to Bitumen and Polycyclic Aromatic Hydrocarbons Among Paving Workers¹

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Introduction: An industrial hygiene database has been constructed for the exposure assessment in a study of cancer risk among asphalt workers.

Aim: To create models of bitumen and polycyclic aromatic hydrocarbons (PAH) exposure intensity among paving workers.

Methods: Individual exposure measurements from pavers ($N = 1581$) were collected from 8 countries. Correlation patterns between exposure measures were examined and factors affecting exposure were identified using statistical modelling.

Results: Inhalable dust appeared to be a good proxy of bitumen fume exposure. Bitumen fume and vapour levels were not correlated. Benzo(a)pyrene level appeared to be a good indicator of PAH exposure. All exposures steadily declined over the last 20 years. Mastic laying, re-paving, surface dressing, oil gravel paving and asphalt temperature were significant determinants of bitumen exposure. Coal tar use dictated PAH exposure levels.

Discussion: Bitumen fume, vapour and PAH have different determinants of exposure. For paving workers, exposure intensity can be assessed on the basis of time period and production characteristics. © 2000 British Occupational Hygiene Society. Published by Elsevier Science Ltd. All rights reserved.

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INTRODUCTION

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The International Agency for Research on Cancer (IARC) is currently engaged in a cohort study of cancer risk among European asphalt workers. The objective of the exposure assessment in the study is to estimate exposures of road paving workers to bitumen and coal tar. An investigation of the potential cancer risk due to bitumen exposure is the primary goal of the study. However, coal tar exposure is an important confounder, since it is 'causally associated with cancer in humans' (IARC, 1985) and has been used in paving processes in the past (Partanen *et al.*, 1994). Because polycyclic aromatic hydrocarbons (PAH) are a 'major component' of coal tar (IARC, 1985), we assumed a

priori that PAH exposure is an appropriate measure of exposure to coal tar.

Previously published inhalable dust, bitumen and PAH exposure data for the asphalt industry have been reviewed (Claydon *et al.*, 1984; Brandt *et al.*, 1985; Darby *et al.*, 1986; Monarca *et al.*, 1987; Ekstrom, 1991; Hansen, 1991; Brandt and Cordingley, 1992; CROW, 1992; Wong *et al.*, 1992; Lien, 1993; Machado *et al.*, 1993; Brandt and DeGroot, 1994; Levin *et al.*, 1994; Greenspan *et al.*, 1995; Hicks, 1995; Kitto *et al.*, 1997). They revealed that in modern road paving workers are typically exposed to 0.1–2 mg m⁻³ of bitumen fume which includes 10–200 ng m⁻³ of benzo(a)pyrene. It was further concluded that these reports were of limited value in assessing historical exposure levels in the industry since they lack comprehensive evaluation of the determinants of exposure in road construction (Burstyn *et al.*, 2000a). However, the published reports could be used to create the following crude grouping of road construction workers into groups with similar exposure (in order of decreasing exposure level to both bitumen and PAHs from left to right):

mastic laying ≥ re-paving > surface dressing >

hot mix paving > cold applications

Substantial non-differential misclassification of exposure can be expected to be present in such a grouping, and the degree of this misclassification would be impossible to estimate. Therefore, given that published reports appear to be insufficient for an accurate exposure assessment in the study, a database of individual exposure measurements in the asphalt industry of participating countries (AWE database) was assembled (Burstyn *et al.*, 2000b). The analysis of the paving workers' exposures collected in the database is presented in this report.

The specific goals of this investigation were: (a) to ascertain appropriate measures of exposure levels for use in the epidemiological study; (b) to identify factors influencing exposure intensity; and (c) to construct statistical models that can be used to infer averages of long-term exposure in the past for a group of road paving workers.

METHODS AND MATERIALS

Description of the AWE database

A detailed description of the Asphalt Worker Exposure (AWE) database is given in the companion paper (Burstyn *et al.*, 2000b). However, key features of the database will also be summarized here. The exposure data collected into the database comprised measurements of exposure levels for a variety of agents among asphalt workers, plus sup-

plementary information. The supplementary information was analogous to collected data from a company questionnaire on production characteristics in companies enrolled in the study. This ensures that AWE data can be linked directly to other data that will be used in the exposure assessment. All available exposure data were collected in eight participating countries ($N = 2007$). The major contributors (70% of samples) were four Scandinavian countries with 35% of samples originating from Norway. Germany contributed 20%. The remaining 10% of samples were approximately equally distributed among France and The Netherlands. A small portion of the data came from Israel. The majority of measurements were personal samples (92%) with the exception of samples collected from German asphalt plants, which were mostly stationary (273/283). Stationary samples were employed to determine vehicle exhaust emissions (CO and NO₂ monitors placed on the machines employed in road paving) as well as to measure bitumen fume/vapour emissions at different positions in asphalt plants and on road paving equipment.

The earliest collected samples originated from the late 1960s, while the majority of samples were collected in the late 1970s and between 1985 and 1997. The data set was sufficiently large to permit statistical modelling of the intensity of exposure to inhalable dust, organic matter in the dust (bitumen fume), organic matter content of gaseous emissions (bitumen vapour) and PAHs, since each agent was measured in 500–1000 samples. The majority of measurements were made among road paving workers ($N = 1581$), and only a few came from asphalt plants (351) and waterproofing or roofing operations (62). The overwhelming majority of measurements have not yet been described in peer-reviewed literature. Measurements were originally collected primarily to test compliance, and for industrial hygiene research. Half of the studies claimed to have used a representative sampling strategy.

In nine data sets included in the AWE database, up to six repeated measurements within individuals were identified. These studies were conducted between 1977 and 1996 in France, The Netherlands, Sweden and Norway. Exposures to dust, bitumen fume, vapour, and PAH in a variety of road paving and asphalt mixing operations were measured repeatedly for individual workers. This was typically achieved by monitoring exposures of a single crew over a number of consecutive days.

Correlation between exposure measures

Prior to statistical modelling, the correlation between different exposure measures has been explored in order to establish which exposure measure would be the best available proxy of agents

Table 1. Definition of company questionnaire-based variables used in statistical modelling

Variable type	Definition	Comments
Dummy	'1' if a positive answer was given to the following question, '0' otherwise	
Tar use	'Recycling tar containing asphalt' or 'Application of coal tar solution' or 'Application of coal tar pitch products' or 'Application of coal tar containing bitumen solution' or 'Application of coal tar emulsion'	In AWE data, five indicators of coal tar use were correlated with primary cause of coal tar use being 'Application of coal tar solution'
Hot mix	'Paving hot mixes'	
Surface dressing	'Surface dressing'	
Oil gravel	'Oil gravel paving'	
Emulsion	'Application of emulsion'	
Other paving	'Other paving'	
Mastic laying	'Mastic paving'	
Recycling operation	Recycling operation; not part of company questionnaire ^a	
Recycle bitumen	'Recycling bitumen binders'	
Recycling tar	'Recycling tar containing asphalt'	
Modified bitumen	Polymer, scrap rubber, fly ash or other modified bitumen	
Categorical		
Country	Country where survey was performed	
Continuous		
Application temperature	Application temperature in °C ^b	
Year	Difference between 1997 and year of the start date of survey	Maximum duration of a survey was one year

^aConsidered to be a potential predictor that can be incorporated into the exposure assessment protocol.

^bMissing values were not uniformly distributed among job titles, therefore they were substituted with job title-specific averages.

considered. Correlation coefficients between inhalable dust, bitumen fume and bitumen vapour exposure levels were computed. A total of 27 different PAHs were determined in some samples in the AWE database. These PAHs were often determined in both fume and vapour, resulting in up to 54 different possible PAH levels for a sample. However, in some samples only benzo(a)pyrene levels were determined. Since we were primarily interested in modelling a proxy for exposure to all PAHs (such as benzo(a)pyrene), the sum of benzo(a)pyrene levels in fume and vapour was used as a measure of benzo(a)pyrene exposure when both were available for a sample. We investigated the correlation between benzo(a)pyrene levels and bitumen fume/vapour levels. In addressing the question of whether benzo(a)pyrene levels are representative of all other PAH levels, we have resorted to principal component analysis. Principal component analysis identified groups of variables that share a common feature. Only those groups of variables which contributed substantially to explaining multiple correlation in the data were examined in detail. Each variable comprising a principal component was judged by the absolute magnitude of its eigen-vector (relative to other variables) in order to see how strongly it was associated with the factor common to all variables in a given principal component.

Building predictive models of exposure

We examined the frequency distributions of exposure levels in order to determine if transformations prior to statistical modelling were warranted. Skew in the distributions signalled that logarithmic transformation was needed to satisfy the assumptions of inferential statistical analysis.

Variables used to represent determinants of exposure were defined on the basis of the company questionnaires. All company questionnaire questions had analogues in the AWE database. Their definitions can be found in Table 1. In addition to these variables, other variables were constructed representing workers, sampling under conditions when highest exposures were anticipated, stationary samples, type of sampling head used, and sorbent type used to collect gas-phase emissions.

Multiple linear models for bitumen fume and bitumen vapour were constructed in the sequential procedure described below. At each step of the model building process, all variables that were not statistically significant (0.10 level) were removed from the models. Newly added variables were kept in the models if they were statistically significant, and if alterations in regression coefficients for other variables appeared to be interpretable. At step 1 all variables listed in Table 1, except for time- and country-related variables, were forced into each model. Patterns of correlation between variables in

Table 1 were explored by computing correlation coefficients, and if a correlation existed ($r > 0.70$), only one of the correlates was used in further modelling. The choice of the correlate was driven by considerations of ease in interpretation, should it be included in the final form of the model. At step 2 a variable representing time trend was added to the models. At step 3 a country variable was added to the models. At step 4 variables representing sampling strategy (worst case versus representative) and sample positioning (area versus personal) were added to the models. At step 5 variables representing sampling methods were introduced into the models. At step 6 country-related variables were removed from the models in order to study their effect on model fit. Negligible change in model fit signalled that country-related variables could be removed without harming the predictive power of the models. The rationale for this was that country-related effects are difficult to interpret and it was considered that small losses in model fit could be compensated for by an increase in generalization of the model. Finally, at step 7 a random worker effect was introduced into the models (Proc Mixed in SAS 6.12, restricted maximum likelihood algorithm, compound symmetry covariance matrix). All effects included at step 6 were treated as fixed effects, and worker identity was introduced as a random variable. This consequently permitted the within- and between-worker components of exposure variability to be quantified. These values are needed for calculation of the maximum likelihood estimates of the average of long-term exposure for a group of individuals.

Models of PAH exposure were constructed by evaluating the statistical significance of significant predictors of total organic matter (that is, bitumen fume and vapour) plus the variable representing tar use. The rationale for this approach was based on the expectation that PAHs originate from both bitumen and coal tar.

The fit of the predictive models was evaluated graphically by plotting observed versus predicted exposure levels. Residuals were analysed by plotting predicted values against residual errors. Statistical analysis was performed using SAS 6.12 (SAS Institute Inc.), data management tasks were performed using Microsoft Access 2.0 (Microsoft Corp.) and graphs were prepared using SigmaPlot version 4.01 (SPSS Inc.).

RESULTS

Exposure levels and correlation between exposure measures

Arithmetic and geometric summary statistics for bitumen fume, bitumen vapour and benzo(a)pyr-

Table 2. Bitumen fume, bitumen vapour and benzo(a)pyrene exposure levels among paving workers

	Non-detectable values			All paving operations ^a			Mastic laying only ^a		
	N	GM	SD	Range	N	GM	SD	Range	
Bitumen fume (mg m^{-3})	184	0.28	13.1	< LOD ^c -260	119	2.29	6.32	0.02-260	
Bitumen vapour (mg m^{-3})	5	1.86	20.0	< LOD-290	35	2.06	3.27	0.23-38.1	
Benzo(a)pyrene (ng m^{-3})	321	8.58	476	< LOD-8000	15 ^d	61.6	2038	< LOD-8000	

^aN: sample size; GM: geometric mean; GSD: geometric standard deviation; AM: arithmetic mean; SD: arithmetic standard deviation.

^bNo units.

^cLOD: method limit of detection.

^dTwo non-detectable values.

ene exposures are summarized in Table 2. It is apparent that the exposures cover a wide range of values and tend to be higher during mastic laying.

There was a high degree of correlation between fume and inhalable dust levels (Fig. 1). This allowed us to construct the following regression model: (mg m^{-3} bitumen fume) = $0.93 \times (\text{mg m}^{-3}$ inhalable dust), which accounted for 97% of variability in bitumen fume levels. The intercept of the above model was not different from zero, and was therefore excluded from the final form of the model. Thus, we have concluded that inhalable dust emitted in road paving consists primarily of organic particulate matter, which we refer to as 'bitumen fume'. In all subsequent analyses, when an inhalable dust level was reported for a sample but the organic matter content of that dust was not determined, we assumed that bitumen fume constituted 93% of inhalable dust and used that value in statistical modelling.

No consistent correlation between bitumen fume and bitumen vapour levels appeared to exist. Its magnitude and statistical significance varied greatly between countries (Table 3). Thus, we concluded that we were justified in creating separate predictive models for bitumen fume and vapour.

Not all PAHs were determined in the 414 samples for which series of PAHs were quantified, so missing values were replaced with averages of reported levels for each PAH. Furthermore, for each PAH a sum of its levels in particle and vapour phase was used as an indicator of 'total' exposure. The first two principal components accounted for 50% of the multiple correlation between PAH levels in fume and vapour. The first principal component appeared to be dominant (it explained 38% of the multiple correlation). None of the PAHs contributed disproportionately to the first two principal components. In effect, when eigenvectors indicated that a set of PAHs was contributing to a principal component (> 0.20), all eigenvectors for the set of 'significant' PAHs were approximately equal (0.20–0.44). The set of PAHs comprising the first principal component consisted of 13 out of 27 measured PAHs. The first two principal components included 18 out of 27 monitored PAHs. Benzo(a)pyrene was an important contributor only to the second principal component and had a high degree of correlation with it ($r = 0.79$, $P < 0.0001$, $N = 414$). Thus, we have concluded that total benzo(a)pyrene is a good proxy of one of the principal sources of PAH exposure. In subsequent statistical modelling we have considered both the first principal component and total benzo(a)pyrene as proxies of PAH exposure. The expression used to calculate the value of the first principal component is given below (all PAH levels were in ng m^{-3}):

$$\begin{aligned} &0.28 \times (\text{acenaphthene}) + 0.27 \times (\text{acenaphthylene}) \\ &+ 0.27 \times (\text{anthanthrene}) + 0.26 \\ &\times (\text{benzo(a)anthracene}) + 0.22 \times (\text{benzo(a)fluorene}) \\ &+ 0.29 \times (\text{benzo(k)fluorathene}) + 0.28 \\ &\times (\text{fluoranthene}) + 0.30 \times (\text{fluorene}) + 0.30 \\ &\times (\text{phenanthrene}) + 0.26 \times (\text{pyrene}) + 0.23 \\ &\times (2\text{-methylnaphthalene}) + 0.29 \\ &\times (1\text{-methylnaphthalene}) + 0.30 \times (\text{biphenyl}) \end{aligned}$$

Benzo(a)pyrene levels had a statistically significant correlation with bitumen fume ($r = 0.42$, $P < 0.0001$, $N = 419$), but bitumen fume exposures explained only 18% (0.42^2) of variability in benzo(a)pyrene levels. This moderate correlation was driven by the high correlation between benzo(a)pyrene and bitumen fume levels during mastic laying (mastic laying: $r = 0.96$, $P < 0.0001$, $N = 12$; other paving: $r = 0.32$, $P < 0.001$, $N = 407$). In general, there was no correlation between benzo(a)pyrene and bitumen vapour exposures ($r = -0.01$, $P < 0.87$, $N = 364$). However, the correlation between mastic vapour and benzo(a)pyrene was very strong ($r = 0.99$, $P < 0.0001$, $N = 10$). Thus, it appeared that factors other than bitumen fume and vapour levels have an important bearing on benzo(a)pyrene exposure levels. The first principal component of PAHs was not correlated with either bitumen fume or vapour.

Thus, on the basis of the correlation patterns presented above, we selected bitumen fume ($N = 1193$), bitumen vapour ($N = 510$), total benzo(a)pyrene ($N = 487$) and the weighted sum of PAH levels comprising the first principal ($N = 414$) as dependent variables in subsequent statistical models.

Statistical distribution of exposure levels and treatment of non-detectable values

All four measures of exposure levels followed skewed distributions, which were normalized by logarithmic transformations (base e). Thus, natural logarithms of exposure levels were used in the subsequent statistical modelling in order to stabilize variance. Values below the limit of detection were set to be equal to that limit. This will result in an upward bias in the absolute levels of bitumen fume and vapour. Nevertheless, this should have negligible effect on the identification of determinants of exposure, since non-detectable values represented only a small proportion of bitumen fume (184/1193) and vapour (5/510) measurements. A large proportion (321/487) of benzo(a)pyrene values appeared to be non-detectable, even if, in order to qualify as a non-detectable value, both fume and

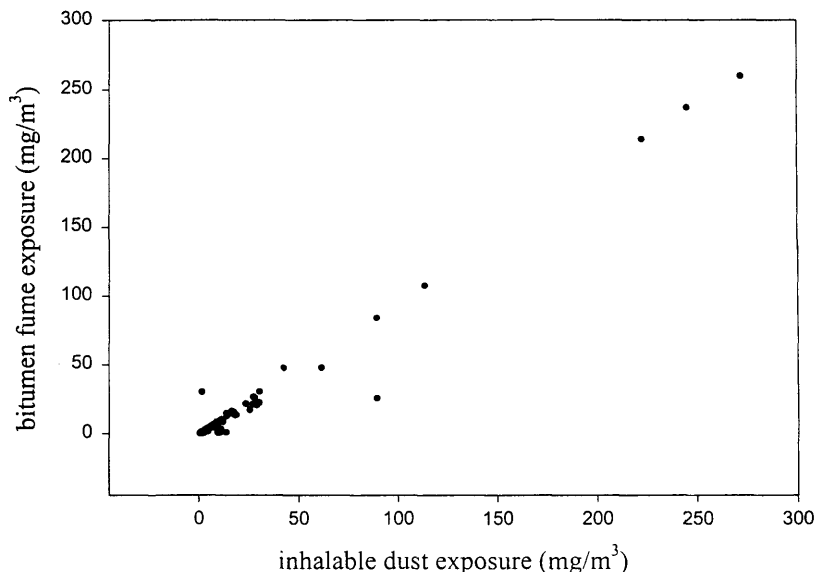


Fig. 1. Correlation of bitumen fume and inhalable dust exposure among asphalt workers (bitumen fume = $0.93 \times$ inhalable dust; $R^2 = 0.97$; $N = 266$).

vapour PAH levels had to be reported as non-detectable. Non-detectable benzo(a)pyrene exposure levels were replaced by survey-specific limits of detection, which ranged from 1 to 50 ng m^{-3} . The use of the detection limit was justified because it was not possible to assess how the limit of detection was calculated in different surveys and we used the reported values as an indicator of the original investigator's best estimate of the measured values.

Predictive models of exposure

Statistical models of exposure to PAHs comprising the first principal component accounted for less than 9% of total variability and revealed no association between production factors and this measure of PAH exposure. On the other hand, statistical models of exposure to bitumen fume, bitumen vapour and benzo(a)pyrene exposure explained 36–43% of total variability and revealed strong associations with various production factors. They are summarized in Table 4. In the table, the reference category (defined by the intercept) depends on what

variables were important in that model, but in all cases hot mix paving is included in the reference category. The models have the same general form:

$$\log_e(\text{exposure}) = \sum_{\text{all } i} \beta_i \times (\text{determinant of exposure } i) + \text{intercept} + \text{worker effect}$$

It is apparent that mastic laying was associated with elevated exposures to all three agents. Recycling operations produced elevated bitumen fume exposures, while below-average bitumen fume exposures resulted from oil gravel paving. Surface dressing, oil gravel paving and elevated application temperatures increased bitumen vapour exposures. There were no differences in the exposures to bitumen fume and bitumen vapour between countries, when adjusted for the effects of other determinants of exposure. The effect of application temperature on bitumen fume exposure in non-mastic applications, adjusted for other predictors of exposure, was the same as in univariate analysis (Fig. 2), but it failed to improve model fit and was thus excluded from the final form of the bitumen fume model. Bitumen vapour and benzo(a)pyrene exposures have declined from the 1970s to the 1990s (11 to 14% per year) more rapidly than bitumen fume exposures (6% per year). These time trends are illustrated in Fig. 3. Since coal tar use is a time-dependent variable, interaction between time trend and tar use was examined in a benzo(a)pyrene model, but it was found to be not statistically significant ($P = 0.29$).

The benzo(a)pyrene model shared most determinants of exposure with the bitumen fume and

Table 3. Summary of the analysis of correlation between bitumen fume and vapour exposure among road paving workers

Country	Number of observations	Pearson r ($P(r = 0)$)
Finland	57	0.03 (0.8)
France	26	-0.05 (0.8)
Germany	116	0.1 (0.3)
Norway	279	0.04 (0.5)
Sweden	23	0.95 (0.0001)
All countries	501	0.01 (0.02)

Table 4. Statistical models of \log_e (mg m^{-3} bitumen fume), \log_e (mg m^{-3} bitumen vapour) and \log_e (ng m^{-3} benzo(a)pyrene)

Variable	Bitumen fume		Bitumen vapour		Benzo(a)pyrene	
	β^a	SE ^a	β	SE	β	SE
Mastic laying	0.88	0.22	0.78	0.47	1.27	0.47
Mastic laying \times worst case (indoors) ^b	1.71	0.29	1.70	0.70	3.07	0.84
Recycling	0.89	0.25		NS	1.51	0.28
Recycling \times worst case ^b	1.67	0.37		NP ^c		NS
Surface dressing		NS ^d	1.88	0.24	0.38	0.20
Oil gravel	-1.51	0.28	0.48	0.47	-0.65	0.38
Tar use		NS		NS	1.68	0.28
Years before 1997	0.062	0.008	0.135	0.034	0.107	0.022
Application temperature in non-mastic paving($^{\circ}\text{C}$)		NS	0.009	0.003		NS
Area sample (yes/no)		NS	2.26	0.37		NS
Sorbent type ^e						
Silica		NA ^f	-2.74	0.24		NS
Charcoal		NA	-0.95	0.37		NS
Sampling head ^g						
37 mm closed face	-1.32	0.15		NA		NS
GGP	1.20	0.16		NA		NP
Intercept	-2.09	0.11	-1.19	0.42	0.91	0.16
Percentage variance explained by fixed effects		41		36		43
Number of observations		1193		510		487
$\text{BW}S_y^2$ ($\text{BR}_{0.95}$) ^h	0.99 (49)	0.20 ⁱ	1.16 (68)	0.31 ⁱ	0.43 (13)	0.28 ⁱ
$\text{ww}S_y^2$ ($\text{wR}_{0.95}$) ^h	1.08 (59)	0.22 ⁱ	1.26 (81)	0.27 ⁱ	1.71 (168)	0.32 ⁱ

^a β : regression coefficient; SE: standard error of regression coefficient.

^bSymbol ' \times ' denotes a multiplicative interaction of variables.

^cNot possible to estimate.

^dVariable is not statistically significant and therefore is not included in the model or did not improve model fit upon inclusion in the model.

^eReference category is XAD2 sorbent.

^fNot applicable.

^gReference category includes: 37 mm open face cassette, 25 mm closed face cassette (Millipore) and PAS6.

^h $\text{BW}S_y^2$: variance of the distribution of logarithmic means of an individual's exposures (between-worker); $\text{ww}S_y^2$: variance of the distribution of logarithmic means of exposure from day to day for an individual (within-worker); $\text{BR}_{0.95}$: ratio of 97.5th to 2.5th percentile of the between-worker distribution of exposures; $\text{wR}_{0.95}$: ratio of 97.5th to 2.5th percentile of the distribution of exposures within a worker.

ⁱStandard deviation of variance components' estimates generated in procedure whereby a model was evaluated 300 times on random subsets of 50% of the data.

vapour models, except with respect to coal tar use. Indeed, when bitumen fume levels and coal tar use were considered as the only predictors of benzo(a)pyrene exposure, the following model resulted, accounting for 32% of variability in benzo(a)pyrene exposure:

$$\log_e (\text{ng m}^{-3} \text{ benzo(a)pyrene}) = 0.10 \times (\text{mg m}^{-3} \text{ total organic fume}) + 2.71 \times (\text{tar use}) + 1.50$$

When tar use is 'yes', the value used is 1; when tar use is 'no' the value used is 0. In the model, tar use *per se* accounted for 20% of variability in benzo(a)pyrene exposure.

Examination of the residuals of the three models revealed no substantial deviations from the assumption of homoscedasticity.

Table 5 presents the estimates of variance components with and without correcting for the significant determinants of exposure (fixed effects). The results presented in Table 5 indicate that predictors

of exposure identified in our models are predominantly associated with between-worker variance components. It would appear that fixed effects explain a greater proportion of between-worker variance in the benzo(a)pyrene model ($\{(2.00 - 0.43)/2.00 = 79\%$) than they do in the bitumen fume ($\{(2.27 - 0.99)/2.27 = 56\%$) and bitumen vapour models ($\{(2.51 - 1.16)/2.51 = 54\%$).

DISCUSSION

Comparison with the literature and interpretation of the observed pattern of determinants of exposure

Organic matter content of inhalable dust emitted during paving. Our finding of a high degree of correlation between inhalable dust and its organic matter content implies that little information is gained about bitumen fume exposure by analysing organic matter content of samples compared to gravimetric analysis. Organic matter content of asphalt dust has been reported to vary considerably, especially under low inhalable dust exposure conditions (Monarca *et*

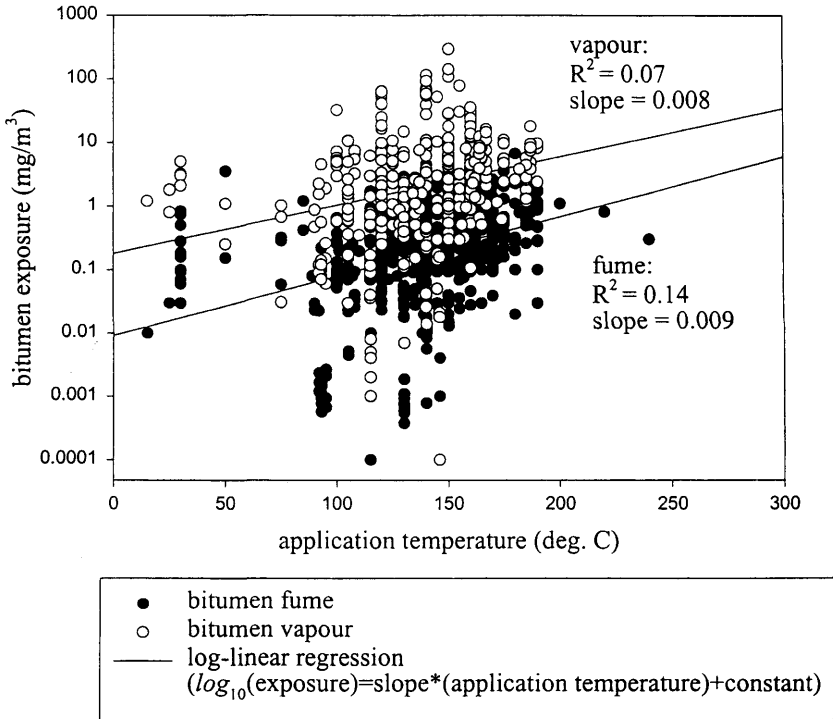


Fig. 2. Influence of application temperature on bitumen fume ($N = 855$) and bitumen vapour ($N = 411$) exposures in non-mastic applications.

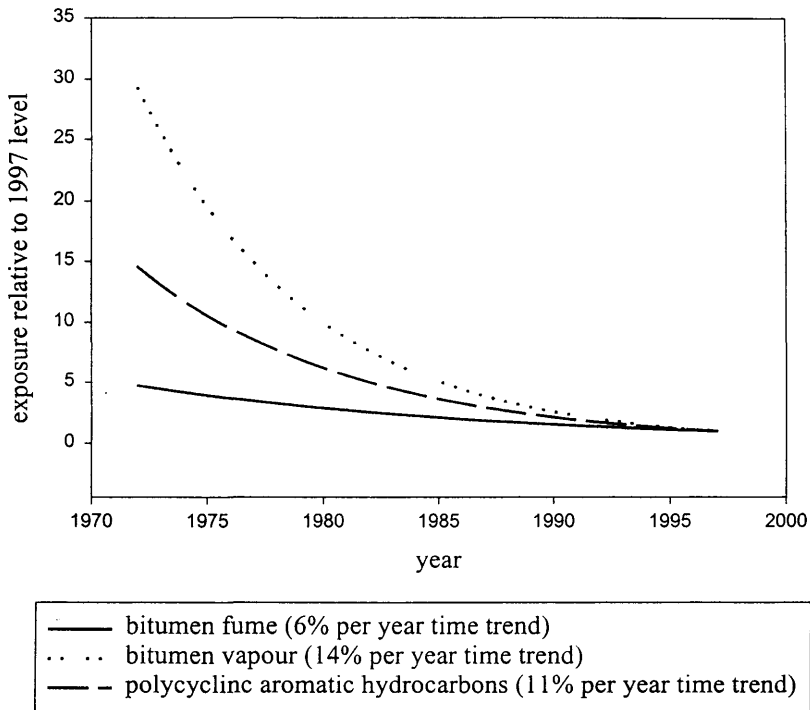


Fig. 3. Predicted time trends in bitumen fume, bitumen vapour and polycyclic aromatic hydrocarbon exposures, adjusted for production characteristics and sampling methods.

Table 5. Influence of fixed effects and random effects on variance components in models of bitumen fume, bitumen vapour and benzo(a)pyrene exposure among paving workers

Model ^a	Dependent variable					
	\log_e (mg m ⁻³ bitumen fume)		\log_e (mg m ⁻³ bitumen vapour)		\log_e (ng m ⁻³ benzo(a)pyrene)	
	BW S_y^2 ^b	ww S_y^2 ^c	BW S_y^2	ww S_y^2	BW S_y^2	ww S_y^2
Worker and fixed effects	0.99	1.08	1.16	1.26	0.43	1.71
Worker	2.27	1.18	2.51	1.30	2.00	1.74
N^d		1193		510		487
k^e		904		346		292

^aAll models include intercept.

^bEstimate of logarithmic between-worker variance.

^cEstimate of logarithmic within-worker variance.

^dNumber of exposure measurements.

^eNumber of workers.

al., 1987; Brandt and Cordingley, 1992; CROW, 1992). It is evident from Fig. 1 that the same pattern holds for our data: there is more variability in organic matter content of particulate emissions when their overall level is low. Removing exposure levels that were above 30 mg m⁻³ from Fig. 1 had the predictable effect of reducing the range of variables and increasing variability in the studied relationship: R^2 decreased to 0.64. The slope of the relationship also decreased from 0.93 to 0.74. Thus, our results are in agreement with previously published reports. However, it is quite possible that the apparent change in organic matter content of inhalable dust emitted in road paving is an artefact of increased uncertainty in quantifying relatively low particulate and organic matter levels. Even if there is a 20% decrease in bitumen fume content from high to low dust operations, the effect is likely to be unnoticeable in a statistical model of bitumen fume, since much stronger predictors of exposure are present in road paving (factors of 2–5). In fact, applying an alternative relationship for translating inhalable dust into bitumen fume levels, assuming increasing content of organic matter with increasing particulate matter, hardly alters the parameters of the bitumen fume exposure model (data not shown). The coefficient most affected was that for time trend, but it was not significantly different from the one presented in this study (95% confidence, two-tailed *t*-test). Thus, statistical models appear to be robust with respect to the above assumptions that could have been made about the relationship between inhalable dust and bitumen fume exposure. Nevertheless, since it has been suggested that organic matter content of inhalable dust is dependent on application temperature and asphalt type (Brandt and Cordingley, 1992), this issue should be investigated further. The efficiency of different solvents and quantification methods may further confound the issue, but it was not possible to explore it using our data.

Sources of PAHs. The PAHs that comprised the first principal component do not vary across different production conditions in road paving. They represent an overall PAH exposure common to all road paving workers. The identity of the underlying source of these PAHs is unclear. The PAHs in the second principal component and benzo(a)pyrene both appear to represent tar use and PAHs in bitumen fume. These two variables are very good predictors of benzo(a)pyrene exposure levels. We were unable to test whether the second principal component is related to changes in bitumen vapour exposure levels, since no bitumen vapour measurements were collected during the use of tar. However, a comparison of the patterns of regression coefficients in bitumen vapour and benzo(a)pyrene models (see the discussion below) indicates that changes in total benzo(a)pyrene exposure may mimic, to some extent, those in bitumen vapour.

Mastic laying. Bitumen fume and vapour exposures arising during mastic laying were 2–13 times higher than those observed in association with hot mix paving. This corroborates findings of other researchers, who have indicated that mastic laying results in much higher bitumen fume exposure levels than other paving activities (Brandt *et al.*, 1985; Hansen, 1991; CROW, 1992). In mastic laying elevated bitumen fume and vapour exposures can be expected, since the source of exposure (that is, the application mixture) is closer to the exposed person's breathing zone owing to the fact that mastic is habitually applied by hand. It has been reported that up to a twofold decrease in inhalable dust exposure levels during paving can be explained by an increase in distance between the exposed person's breathing zone and the source of exposure (Claydon *et al.*, 1984). Furthermore, higher temperatures of mastic mixes can lead to more fuming. In fact, it was not possible to separate the effects of application temperature from those of mastic laying in the models. Differences in composition and physical properties of bitumen used in mastic pav-

ing may also account for this effect. 'Worst case' mastic laying corresponded to applying mastic asphalt in indoor environments (for example a covered parking lot). The apparent fivefold to sixfold difference between indoor and outdoor exposures observed in our models is the same order of magnitude as those reported by Brandt *et al.* (1985). It would also appear that the industrial hygienist who conducted the surveys entered into the AWE database was able to consistently identify indoor mastic laying as a 'worst case' scenario.

The association of mastic laying with a fourfold to eightyfold increase in PAH exposure relative to hot mix paving is rather surprising, since coal tar is not supposed to be used in the formulation of mastic mixes. The explanation for the observed effect may lie with the fact that even though PAH content of mastic emissions may be low, the overall amount of these emissions is so high that it results in PAH exposure comparable to that incurred during the use of coal-tar-containing materials. This hypothesis is supported by: (a) the high correlation between levels of mastic fume or vapour and benzo(a)pyrene exposures reported earlier; and (b) patterns of coefficients in statistical models. However, owing to the controversy about cancer risk in mastic laying and the uncertainty about the contribution of the underlayer with coal tar to PAH exposures during mastic laying (Wong *et al.*, 1992), this issue deserved further attention. If the source of PAHs in mastic laying is the under-layer with coal tar (Wong *et al.*, 1992), then owing to the fact that coal tar is no longer used in paving, we can assume that the high PAH exposures resulting from mastic laying are a thing of the past. However, if the mechanism we have suggested is correct, then high PAH exposures (and the associated health risks) due to mastic laying may still be present, especially in indoor environments.

Re-paving or in situ recycling. Re-paving or *in situ* recycling operations have been associated with elevated bitumen fume and benzo(a)pyrene exposure. This may be due to the fact that coarse particulate is generated during recycling operations in the course of grinding the old asphalt layer. These particles may absorb bitumen fume and vapour generated in heating old asphalt, resulting in elevated bitumen fume rather than vapour exposures. It is not clear as to what defined worst case scenarios in recycling operations. However, there are some indications that the re-paving technique in which old asphalt is heated with a propane burner (hot *in situ* recycling) produced these high exposure situations (CROW, 1992). An alternative to hot *in situ* recycling is milling and recycling at ambient temperatures. Since bitumen vapour was not monitored during these operations, the effect of milling and recycling on bitumen vapour exposure was not possible to estimate. As in the case of mastic laying,

it would appear that industrial hygienists conducting the surveys of recycling operations were able to identify a fivefold increase in exposure as 'worst case'. The association between PAH levels and recycling operations may be due to the correlation between fume and PAH concentrations. It is also possible that the same mechanism that leads to elevated fume levels during recycling operations is also responsible for preferential absorption of heavier PAH, such as benzo(a)pyrene, by particles comprising fume. This would lead to elevated PAH content of fume during recycling/re-paving operations.

Surface dressing and oil gravel paving. On the basis of limited data, CROW (1992) has concluded that surface dressing is associated with elevated fume and PAH exposure relative to hot mix paving. Our results indicate that there is no difference in bitumen fume exposure levels between surface dressing and hot mix paving. It is possible that small differences between these two types of applications do exist, but their magnitude is likely to be small (less than a factor of two), resulting in negligible contrasts in average fume exposure levels. Our observed association between surface dressing and bitumen vapour can be attributed to differences in modes of applying bitumen to road surfaces. In surface dressing, a liquid bitumen solution (cut-back or emulsion) is sprayed onto the road surface, while in other applications (with the exception of oil gravel paving) bitumen is deposited as a part of solid asphalt (a mixture of bitumen and mineral aggregate). It is possible that the spraying of liquid bitumen solution is more conducive to the formation of small droplets and vapour, which are preferentially absorbed by sorbent tubes. It is also likely that bitumen used in surface dressing has a higher proportion of volatile hydrocarbons, originating either from bitumen itself or from the cut-backs which are customarily added to bitumen solutions used in surface dressing. The process of vapour emission during surface dressing is governed by application temperature and vapour pressure of the constituents of the mixture. Given that we cannot expect all surface dressing mixtures to have the same composition, we can expect that application temperature *per se* will not account for the amount of vapour originating in surface dressing. The influence of bitumen composition on the relationship between the amount of bitumen fume emitted and temperature has been reported (Machado *et al.*, 1993).

Oil gravel resulted in bitumen fume exposures, which were lower than those in the reference category (which includes hot mix paving). This was probably the case because fume formation requires bitumen to be heated (as in surface dressing). However, during oil gravel paving, binding material is applied at ambient temperatures, precluding the occurrence of evaporation and condensation pro-

cesses that lead to fume formation. The weak positive association between oil gravel paving and organic vapour levels is probably a result of the application method, similar to surface dressing, in which vapour and small aerosol droplets are generated during spraying of a liquid. It is also likely that volatile organic materials not present in hot mix asphalt or surface dressing mixtures evaporate during oil gravel paving, resulting in elevated vapour exposure.

The observed pattern of relationships between benzo(a)pyrene exposure and these two application methods suggests that it mirrors patterns of organic matter levels in fume and vapour.

Effects of application temperature. The dependence of bitumen fume, vapour and PAH exposure on application temperature has been reported by other investigators (Ekström, 1991; Brandt and Cordingley, 1992; Lien, 1993; Machado *et al.*, 1993; Brandt and DeGroot, 1994; Kitto *et al.*, 1997). The rationale for temperature dependence of the levels of asphalt emissions is that the emissions are formed via evaporation of the organic components of asphalt. This evaporation process is governed, at least in part, by the temperature of a given asphalt mix. In our data, application temperatures were most commonly extracted from production records, but in one survey they were measured at the location of a worker. Separate analysis by the method of assessing application temperature did not yield diverging results. We only observed the effect of application temperature, independent of mastic laying, on bitumen vapour. Despite some authors' claims that application temperature is the primary predictor of bitumen fume and vapour exposure levels (Ekström, 1991; Brandt and Cordingley, 1992; Brandt and DeGroot, 1994), in the AWE data the application temperature alone accounts for 7–14% of variability in bitumen fume and vapour exposure levels (Fig. 2). However, the observed slope of the relationship is of the same order of magnitude as reported in the literature.

There are several possible explanations as to why we did not observe a very strong influence of application temperature on exposures in road paving. First, laboratory studies (Machado *et al.*, 1993; Brandt and DeGroot, 1994; Kitto *et al.*, 1997) do not take into account a multitude of factors that may and probably do influence personal exposure in the workplace. Thus, a variable can be expected to have lower predictive power in the 'real world' than it does under controlled laboratory conditions. Second, the model developed by Brandt and DeGroot (1994) in a set of laboratory experiments has been validated on a very small number of field observations, and thus its generalization remains uncertain. Third, two reports claiming that application temperature has a strong effect on bitumen fume and vapour exposure (Ekström, 1991; Lien,

1993) have not been published in peer-reviewed scientific literature, thus making it difficult to ascertain how the authors arrived at such a conclusion. Incidentally, the data from the last two reports are included in the AWE database and form part of the data set used to build the statistical models presented in this report.

From the point of view of exposure assessment in an epidemiological study, it is quite fortunate that we do not have to rely very heavily on application temperature in making inferences about exposure levels. Our experience has indicated that collecting accurate information about typical application temperatures in the past is difficult, with uncertainty on the order of 10–20°C. Thus, for example, if we had to assign a twofold change in exposure level for every 15°C, as suggested by Ekström (1991), a significant misclassification of exposure would have resulted.

Even though application temperature may not affect the amount of bitumen fume emissions, it may influence their composition (Kitto *et al.*, 1997). Norseth *et al.* (1991) found that health effects of bitumen fume exposure were associated with application temperature. This may indicate that the toxic properties of asphalt emission change with change in application temperature. Thus, one of the principal limitations of the AWE database and our predictive models becomes apparent: they do not take into account either bitumen composition or quality and quantity of non-bitumen components of the binder. Questions about bitumen composition are at present difficult to answer (CROW, 1992; Kitto *et al.*, 1997) since bitumen is defined on the basis of its physical properties rather than its chemical composition. Therefore, the above-mentioned limitation is not an inherent fault of our data, but is merely a reflection of the overall poor current understanding of bitumen chemistry.

Coal tar in paving mixes. The strong relationship between coal tar use and PAH levels observed in our study supports our *a priori* assumption that an assessment of exposure to coal tar can be based on PAH measurements. Our results reveal that in the presence of coal tar, PAH exposures in road paving are elevated by a factor of five (that is, $\exp(1.68)$). However, on the basis of the results of laboratory experiments, PAH content of tar can be assessed to be 100 or even 1000 times higher than that of bitumen (Lindstedt and Sollenberg, 1982; Brandt *et al.*, 1985; Darby *et al.*, 1986; Machado *et al.*, 1993). This apparent difference in the observed effects of coal tar on PAH exposure levels in road paving can be explained by the observation that most benzo(a)pyrene levels were non-detectable, unless coal-tar-containing materials were used in paving. Thus, the absolute magnitude of the observed coal tar effect is dependent on the treatment of non-detectable benzo(a)pyrene levels in statistical analysis. On the

other hand, some doubts have been raised about how representative laboratory-generated emissions are of those encountered in workplace (Niemeier *et al.*, 1988).

Time trend. According to the time trend observed in the AWE data, bitumen fume, bitumen vapour and benzo(a)pyrene exposures have decreased by a factor of 2–3 each decade. The possibility that the time trend in benzo(a)pyrene levels was due to the manner in which we treated non-detectable values was rejected since there was no time trend in the values we used to replace the non-detectable benzo(a)pyrene exposure measurements. We have no technical data that would explain these patterns. The role of other factors that may affect the time trends, such as those related to social and economic conditions, is difficult to ascertain. The use of time trend for which we have no explanation will lead to biased estimates of exposure only if the time trends themselves are estimated with a bias. There is no reason to believe that our time trend estimates for the asphalt industry are biased. For unbiased exposure assessment for epidemiological purposes it is critical to explain *how* exposure levels have changed, not *why* they have done so.

A recent investigation of world-wide time trends in exposure data (Symanski *et al.*, 1998a, b) has indicated that when occupational exposures decrease over time, they generally do so at a rate of 6–17% per year (statistically significant downward trends, $P < 0.05$). Our results are in agreement with these trends. The limitations of the meta-analysis conducted by Symanski *et al.* (1998a) were the inability (a) to obtain individual measurements, and (b) to adjust time trends for the effects of sampling strategy (notably worst case versus representative) and sampling methods. Our estimates of time trends do not suffer from these limitations, since they are based exclusively on individual measurements (corrected for the effect of area sampling) and are adjusted for sampling strategy, analytical methods and production factors. Thus, our results provide additional evidence of the validity of the time trends obtained by Symanski *et al.* (1998a, b).

Variability of bitumen and PAH exposures in paving operations. Estimating an arithmetic mean exposure for a group of workers using multiple linear models presented in this report requires one to have an estimate of within-worker (day-to-day) variability of exposure (Tornero-Velez *et al.*, 1997). Our estimates of within-worker variance presumably underestimate true day-to-day variability in bitumen and PAH exposures. This arises from sampling strategies employed in collecting repeated exposure measurements, in which exposures were monitored over the duration of one project on several consecutive days. Thus, the collected measurements cannot represent a variety of exposure circumstances (that is, projects) that paving workers experience in a

given year. Nevertheless, the within-worker logarithmic variance for outdoor work, such as road paving, was of similar magnitude (1.40) in other data (Kromhout *et al.*, 1993). The reason for this is possibly related to the fact that outdoor work is often not very different from one day to another. Consequently, exposure levels can be expected to be determined to a large extent by environmental factors such as wind and rain, which were not accounted for in either data set.

Our results suggest that the predictors of exposure present in our models are related to differences in exposure between workers. These predictors explain at least half of the variability in exposure concentrations between-workers. It has been suggested that process-related variables influence between-worker variance components (Kromhout *et al.*, 1993). However, the grouping of workers solely on the basis of identified predictor variables will not result in uniformly exposed groups, since mean exposures for each worker in these groups can be expected to vary over a 13–70-fold range (as assessed by $B_{R_{0.95}}$ values).

Methods of measuring bitumen exposure. Area samples of bitumen vapour were 10 times higher than personal ones. A possible explanation for the observed effect is that the stationary samples were placed in areas of high exposure (close to the source, such as the screed of a paving machine) which are avoided by workers. Even though area samples have been generally reported to underestimate personal exposures (Scheeper *et al.*, 1995), the bias in area samples is probably highly dependent on their position. Thus, in estimating an individual's exposure level, stationary sampling should be avoided.

Different sorbents have demonstrated varying affinities to bitumen vapour. It is not clear as to why this is the case. Given that the effect of sorbent choice on the measured bitumen vapour level can be substantial (a factor of 0.06 in the case of silica gel, as compared to XAD2), this issue should be the subject of further research.

The type of sampling head used to collect bitumen fume appears to have had a significant impact on the observed exposure levels. We have chosen the sampling head employed to collect the majority of samples (37 mm open face Millipore sampler) as a reference category, even though we are aware that its collection efficiency deviates significantly from the toxicologically relevant inhalable dust fraction (Kenny *et al.*, 1997). However, these deviations for small particles, such as those that can be expected to form bitumen fume ($< 40 \mu\text{m}$ in aerodynamic diameter), are not dramatic (Kenny *et al.*, 1997). Nevertheless, the GGP sampler, a modified version of the GSP sampler whose collection efficiency better adheres to the inhalable convention (Kenny *et al.*, 1997), would probably have formed a more

appropriate reference category. Unfortunately, the GGP sampler was used to collect only a limited number of samples, all of them from Germany, making the generalization of the effect of the GGP sampler somewhat questionable. Furthermore, the GGP sampler is expected to outperform the 37 mm open-face sampler mostly at low wind speeds (less than 4 m s^{-1}), but at high wind speeds both samplers deviate significantly from collecting inhalable dust (Kenny *et al.*, 1997). According to the AWE database, wind speeds during road paving ranged between 0.5 and 33 m s^{-1} ($N = 303$, $AM = 3.1 \text{ m s}^{-1}$, $GM = 2.1 \text{ m s}^{-1}$, $GSD = 2.3$). Under such variable wind conditions it is very difficult to predict how the collection efficiencies of samplers would be affected. Unfortunately, wind speed was not recorded during sampling with the GGP, making it impossible to correct the observed 'sampler effect' for wind speed. Thus, absolute levels of bitumen fume predicted by our model can underestimate levels of fume available for inhalation by a factor of approximately three. This has important implication for standard setting and risk assessment, indicating that relative performance of different samplers in outdoor environments should be the subject of further research.

In general, characterization of exposure to bitumen has suffered from a lack of standard methods, which have been demonstrated to collect physiologically and toxicologically relevant fractions of bitumen emissions.

Limitations of the predictive models

The AWE data for paving workers are unbalanced. We attempted to compensate for this by choosing the appropriate statistical model (restricted maximum likelihood algorithm) (Searle, 1988). Nevertheless, we cannot alter the nature of the data. Somewhat different subsets of the AWE data were used to construct predictive models for the three agents raising, the possibility that factors which operated in some subsets may not have been present in others. Consequently, the classical problem of unbalanced data is manifested by the fact that some effects cannot be estimated. Thus, the challenge of obtaining a data set which would allow one to conduct a comprehensive evaluation of the determinants of bitumen (fume and vapour) and PAH exposure remains.

There is a noticeable lack of dermal exposure measurements in the AWE database. This reflects the virtual absence of data on dermal exposure in the European asphalt industry, with the exception of one study which measured dermal deposition of PAH during road construction (Jongeneelen *et al.*, 1988). Since the dermal route of bitumen exposure can potentially play a role in increasing the risk of adverse health effects among bitumen-exposed individuals, more research is clearly needed to increase

our understanding of dermal exposure in the asphalt industry.

CONCLUSION

Changes in bitumen fume, bitumen vapour and PAH exposures are each associated with somewhat different factors in road paving. In assessing the intensity of exposure among road paving workers, adjustments for time period and the type of work they have performed must be made. Time trend and a small set of production parameters can be used to explain a substantial portion of variability in exposures among paving workers. Our models, despite their limitations, will help to make the assessment of exposures among road paving workers in a multicentric cohort study a data-driven, reproducible process. The models have also revealed a much more complex pattern of determinants of exposure than could have been ascertained, with any degree of certainty, from the review of published reports alone.

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REFERENCES

- Brandt, H. C. A. and Cordingley, N. (1992) In *Personal Exposures to Bitumen Fumes and PAC During Road Paving and in Laboratory Simulations, 5th Euraspalt Congress*, pp. 166–179. EAPA, The Hague, The Netherlands.
- Brandt, H. C. A. and DeGroot, P. C. (1994) A laboratory rig for studying health aspects related to bitumen fume exposure. *Rassegna del bitume* **23**, 9–15.
- Brandt, H. C. A., DeGroot, P. C., Molyneux, M. K. B. and Tindle, P. E. (1985) Sampling and analysis of bitumen fumes. *Annals of Occupational Hygiene* **29**, 27–80.
- Burstyn, I., Kromhout, H. and Boffetta, P. (2000a) A literature review of levels and determinants of exposure to potential carcinogens and other agents in road construction industry. *American Industrial Hygiene Association Journal*, In press.
- Burstyn, I., Kromhout, H., Cruise, P. J., Brennan, P. *et al.* (2000b) Designing an international industrial hygiene database of exposures among workers in the asphalt industry. *Annals of Occupational Hygiene* **44**, 57–66.
- Claydon, M. F., Eyres, A. R., Guelfo, G., Lentge, H., Mitchell, J. G., Molyneux, M. K. B., Moore, J. P., Rousseaux, G., Sanderson, J. T. and Webb, C. L. F. (1984) Review of Bitumen Fume Exposures and Guidance on Measurement. In *Report no. 6/84*. CONCAWE, The Hague, The Netherlands.
- CROW (1992) Exposure to PAC in Bituminous Road Construction. In *Record 8*. Centrum voor Regelgeving en Onderzoek in de Grond-, Water-, en Wegenbouw en de Verkeerstechniek, Ede, The Netherlands, (in Dutch).
- Darby, F. W., Willis, A. F. and Winchester, R. V. (1986)

- Occupational health hazards from road construction and sealing work. *Annals of Occupational Hygiene* **30**, 445–454.
- Ekström, L. G. (1991) Bitumen fumes and exposure to solvents in connection with the laying of asphalt: Report from exposure measurements carried out in 1990, Johanneshov, Sweden. In *BitTech 625*. Nynas Bitumen AB.
- Greenspan, C. A., Moure-Eraso, R., Wegman, D. H. and Oliver, L. C. (1995) Occupational hygiene characterization of a highway construction project: a pilot study. *Applied Occupational and Environmental Hygiene* **10**, 50–58.
- Hansen, E. S. (1991) Mortality of mastic asphalt workers. *Scandinavian Journal of Work and Environmental Health* **17**, 20–24.
- Hicks, J. B. (1995) Asphalt industry cross-sectional exposure assessment study. *Applied Occupational and Environmental Hygiene* **10**, 840–848.
- IARC (1985) Polynuclear Aromatic Compounds, Part 4, Bitumens, Coal-tars and Derived Products, Shale-oils and Soots. In *IARC Monographs on the Evaluation of Carcinogenic Risks to Humans*, Volume 35. International Agency for Research on Cancer, Lyon.
- Jongeneelen, F. J., Scheepers, P. T. J., Groenendijk, A., Van Aerts, L. A. G. J. M., Anzion, R. B. M., Bos, R. P. and Veenstra, S. J. (1988) Airborne concentrations; skin contamination; and urinary metabolite excretion of polycyclic aromatic hydrocarbons among paving workers exposed to coal tar derived road tars. *American Journal of the Hygiene Association* **49**, 600–607.
- Kenny, L. C., Aitken, R., Chalmers, C., Fabries, J. F., Gonzalez-Fernandez, E., Kromhout, H., Liden, G., Mark, D., Riediger, G. and Prodi, V. (1997) A collaborative European study of personal inhalable aerosol sampler performance. *Annals of Occupational Hygiene* **41**, 135–153.
- Kitto, A. M., Pirbazari, M., Badriyha, B. N., Ravindran, V., Tyner, R. and Synolakis, C. E. (1997) Emissions of volatile and semi-volatile organic compounds and particulate matter from hot asphalts. *Environmental Technology* **18**, 121–138.
- Kromhout, H., Symanski, E. and Rappaport, S. M. (1993) A comprehensive evaluation of within- and between-worker components of occupational exposure to chemical agents. *Annals of Occupational Hygiene* **37**, 253–270.
- Levin, J. O., Andersson, K. and Hallgren, C. (1994) Exposure to low molecular polyamines during road paving. *Annals of Occupational Hygiene* **38**, 257–264.
- Lien, J. (1993) *The Asphalt Project 1991–92: Survey of Occupational Exposures of Asphalt Workers (English summary)*. Statens vegvesen, Vegdirektoratet, Oslo.
- Lindstedt, G. and Sollenberg, J. (1982) Polycyclic aromatic hydrocarbons in the occupational environment: with special reference to benzo(a)pyrene measurements in Swedish industry. *Scandinavian Journal of Work and Environmental Health* **8**, 1–19.
- Machado, M. L., Beatty, P. W., Fetzer, J. C., Glickman, A. H. and McGinnis, E. L. (1993) Evaluation of the relationship between PAH content and mutagenic activity of fumes from roofing and paving asphalts and coal tar pitch. *Fundamentals of Applied Toxicology* **21**, 492–499.
- Monarca, S., Pasquini, R., Scassellati, S. G., Savino, A., Bauleo, F. A. and Angeli, G. (1987) Environmental monitoring of mutagenic/carcinogenic hazards during road paving operations with bitumens. *International Archives of Occupational and Environmental Health* **59**, 393–402.
- Niemeier, R. W., Thayer, P. S., Menzies, K. T., von Thuna, P., Moss, C. E. and Burg, J. (1988) A comparison of skin carcinogenicity of condensed roofing asphalts and coal tar pitch fumes. In *Polynuclear Aromatic Hydrocarbons: A Decade of Progress*, eds M. Cooke and A. J. Dennis, Tenth International Symposium, pp. 609–647. Battelle Press, Columbus, OH.
- Norseth, T., Waage, J. and Dale, I. (1991) Acute effects and exposure to organic compounds in road maintenance workers exposed to asphalt. *American Journal of Industrial Medicine* **20**, 737–744.
- Partanen, T., Heikkilä, P. and Boffetta, P. (1994) Feasibility Phase of a Study on Cancer Risk among Workers in the European Asphalt Industry. In *Internal report 94/005*. International Agency for Research on Cancer, Lyon.
- Scheeper, P., Kromhout, H. and Boleij, J. S. M. (1995) Wood-dust exposure during wood-working processes. *Annals of Occupational Hygiene* **39**, 141–154.
- Searle, S. R. (1988) Mixed models and unbalanced data: wherefrom, whereat and whereto? *Communications in Statistical-Theory Methods* **17**, 935–968.
- Symanski, E., Kupper, L. L., Hertz-Picciotto, I. and Rappaport, S. M. (1998a) Comprehensive evaluation of long term trends in occupational exposure: part 2. Predictive models for declining exposures. *Occupational and Environmental Medicine* **55**, 310–316.
- Symanski, E., Kupper, L. L. and Rappaport, S. M. (1998b) Comprehensive evaluation of long term trends in occupational exposure: part 1. Description of the database. *Occupational and Environmental Medicine* **55**, 300–309.
- Tornero-Velez, R., Symanski, E., Kromhout, H., Yu, R. C. and Rappaport, S. M. (1997) Compliance versus risk in assessing occupational exposures. *Risk Analysis* **17**, 279–292.
- Wong, O., Bailey, W. J. and Amsel, J. (1992) Cancer mortality and incidence in mastic asphalt workers: letter to the editor and author's reply. *Scandinavian Journal of Work and Environmental Health* **18**, 133–141.