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# **COAL-TAR PITCH**

Coal-tar pitch and associated exposures were considered by previous IARC Working Groups in 1984, 1987, and 2005 (IARC, 1985, 1987, 2010). Since that time new data have become available, which have been incorporated in this *Monograph*, and taken into consideration in the present evaluation.

# 1. Exposure Data

# 1.1 Identification of the agent

Studies of coke-oven pitch indicate that coaltar pitch contains: four-membered aromatic hydrocarbon ring systems (e.g. chrysene, fluoranthene, pyrene, triphenylene, naphthacene and benzanthracene); five-membered ring systems (picene, benzo[*a*]pyrene (B[*a*]P) and benzo[*e*] pyrene, benzofluoranthenes and perylene); six-membered ring systems (dibenzopyrenes, dibenzofluoranthenes and benzoperylenes); and seven-ring systems (coronene). Other aromatic chemicals in coal-tar pitch include methylated and polymethylated derivatives, mono- and polyhydroxylated derivatives, and heterocyclic compounds (<u>Betts, 1997</u>).

For a more detailed description of the coaltar distillation process, including coal-tar pitch, the reader is referred to *IARC Monograph* Volume 92 (<u>IARC, 2010</u>), and to the *Monograph* on Occupational Exposures during Coal-tar Distillation in this volume.

# 1.2 Human exposure

### 1.2.1 Occupational exposure

Coal-tar pitch is used in electrode manufacture, roofing and paving.

Studies examining exposures during roofing and paving published since the previous *IARC Monograph* (IARC, 2010) are summarized below.

### (a) Roofing

The exposures associated with roofing are the result of two operations. First, the old roof is removed by cutting, prying and scraping the existing material from the roof, and discarding it. A new roof is then installed by melting solid blocks of coal-tar pitch, pumping or carrying buckets of the molten material to the roof, where layers of roofing felt and liquid coal-tar pitch are spread upon the surface to produce a cover (NIOSH, 2000; IPCS, 2004). Roofers are primarily exposed to PAHs. Other exposures include silica, diesel exhaust, asbestos and organic solvents. Recent studies on airborne concentrations of PAHs measured in roofing are summarized in Table 1.1.

Studies in the USA showed that long after cessation of applying coal-tar pitch on roofs,

Reference Country Year of study	Job/task	No. of subjects	No. of samples	No. of smokers	РАН	Dermal Exposure (ng/cm <sup>2</sup> )		Urinary 1-OH- pyrene (µmol/
						Mean	Range	mol creatinine)
<u>McClean et</u> <u>al. (2007)</u> USA 1998	Tear-off of coal-tar roofs	26						
	Roof workers	21	71	NR	Polycyclic aromatic compound	898	48-30014	
			71		Pyrene	11.0	< 2.4-221	
			71		B[a]P	3.3	< 0.5-59	
	Tear-off	NR	41	NR	Polycyclic aromatic compound	886	49-33538	
			41		Pyrene	11.5	< 2.4-168	
			41		B[a]P	4.6	< 0.5-84	
	Put-down	NR	56	NR	PAC	344	48-21437	
			55		Pyrene	3.8	< 2.4-150	
			54		B[a]P	1.0	< 0.5-59	
	Kettlemen	5	19	NR	Polycyclic aromatic compound	299	40-4558	
			19		Pyrene	4.5	< 2.4-34	
			18		B[a]P	0.9	< 0.5-20	
<u>Toraason et</u> <u>al. (2001)</u> USA 1998	Tear-off of coal-tar roofs							Mean (SD)
	coal-tar tear-off exposure		16		start wk			0.74 (0.86)
			15		end wk			3.55 (2.17)
	asphalt-fume only exposure		6		start wk			0.26 (0.13)
			5		end wk			0.58 (0.29)
	controls		3		start wk			0.08 (0.12)
			3		end wk			0.12 (0.12)

# Table 1.1 Concentrations of PAHs in the ambient air of workers in roofing with coal-tar pitch

B[*a*]P, benzo[*a*]pyrene; NR, not reported; SD, standard deviation; wk, week or weeks

workers can still be exposed to coal tar during tear-off of existing roofs (Toraason *et al.*, 2001; <u>McClean *et al.*, 2007</u>). Working with coal-tar pitch was associated with a sixfold increase in PAH exposure, an eightfold increase in pyrene exposure and a 35-fold increase in benzo[*a*] pyrene exposure. Urinary 1-OH-pyrene concentrations in workers and controls showed a fivefold difference at post-shift and a 7.2-fold difference several hours later, which shows the importance of dermal exposure resulting in a prolonged uptake after the end-of-shift.

# (b) Paving

Roadway paving can be conducted by several methods, including hot-mix laying and chip sealing. In hot-mix laying, the mixture of a binder (coal tar, bitumen or a blended product containing both) and aggregate (stone chips) is spread on the roadway by a paving machine, followed by a roller. In the chip-sealing process (also known as surface dressing), the liquid binder (coal tar, bitumen, or a mixture of the two) is sprayed directly onto the road surface, then the aggregate stone is spread on top and rolled (Darby *et al.*, 1986).

Detailed information on cessation of coaltar use in the European paving industry has been collected in the course of an IARC study on cancer mortality among asphalt workers. Table 1.2 presents the last reported year-of-use of coal tar in paving by any company that participated in the cohort study. The data originated from a company questionnaire and its ensuing evaluation by country-specific experts (Burstyn et al., 2003). A gradient in cessation of use can be seen, with Scandinavian countries ending use earlier than central and southern European countries, such as the Netherlands, France and Germany. However, even within countries, large differences in the use of coal tar have occurred between companies, depending on the supplier of the asphalt mixes and the presence of coke ovens in the neighbourhood. Even after cessation

	2	
Country	Last year of use	
Finland	1965	
Denmark	1974	
Sweden	1974	
Norway	1984	
the Netherlands	1990	
France	1992	
Germany	1995	
From Burstyn <i>et al.</i> (2003)		

# Table 1.2 Cessation of use of coal tar in asphaltpaving (surface dressing)

From Burstyn et al. (2003)

of the use of coal tar, workers in road paving have continued to be exposed to this substance due to the use of recycled coal-tar asphalt in some countries.

# 2. Cancer in Humans

In IARC Monograph Volume 92 (IARC, 2010) it was concluded that there is sufficient evidence in humans for the carcinogenicity of occupational exposures during paving and roofing with coaltar pitch. This was based on studies of pavers and roofers who presumably had been exposed to coal-tar pitch (and often also to bitumen), which suggested increased cancer risks in these occupations. An excess risk for lung cancer was reported in studies of members of a roofer's union in the USA, in analyses of registry-based data on pavers in the United Kingdom and roofers in the USA, and in follow-up studies of cancer incidence among pavers in Finland and the Netherlands (Kennaway & Kennaway, 1947, 1951; Hammond et al., 1976; Milham, 1982; Pukkala, 1995; Swaen & Slangen, 1997; Stern et al., 2000). An increased mortality from urinary bladder, laryngeal or skin (non-melanoma) cancer was observed in one or more of these cohorts, but this finding was not widely supported by other studies. In three casecontrol studies conducted in the USA, a tobaccosmoking-adjusted increase in the risk for lung

cancer was reported among roofers (<u>Schoenberg</u> <u>et al., 1987; Zahm et al., 1989; Morabia et al., 1992</u>) however, none of these increases was statistically significant. A meta-analysis of the case–control studies reported a statistically significant metarelative risk (<u>Partanen & Boffetta, 1994</u>).

Since the previous evaluation (<u>IARC, 2010</u>) a few additional studies have been published with information on paving with coal-tar pitch and associated cancers.

# 2.1 Cohort Studies

Roofing and flooring, and paving of roads involve the use of bitumen and coal-tar pitch. Although coal-tar pitch has been phased out in most countries, some studies published since the previous *IARC Monograph* (IARC, 2010) have included workers in road-paving exposed to both petroleum-based bitumen and coal-tar pitch.

In studies in France, Norway, and Sweden a higher lung cancer mortality or incidence was found among workers first employed while the asphalt mixes still contained some coal tar (before 1970 in France and before 1965 in Norway and Sweden). None of the elevated rates were statistically significant (<u>Bergdahl & Järvholm</u>, 2003; <u>Randem *et al.*, 2003; Stücker *et al.*, 2003). In the French study a higher but statistically non-significant mortality from stomach cancer was found in workers hired before 1970 (<u>Stücker *et al.*, 2003</u>; see Table 2.1, available at <u>http://monographs.iarc.fr/ENG/Monographs/</u> vol100F/100F-12-Table2.1.pdf).</u>

In a study in Finland raod-paving workers were ranked by coal-tar exposure (see Table 2.1 on-line). Relative to those not exposed to coal tar, the risk for lung cancer was 1.49 for workers exposed to very low level of coal tar (assessed semiquantitatively on the basis of score–years) and 10.7 for those who exposed to intermediate levels of coal tar (p for trend = 0.05) (Kauppinen *et al.*, 2003).

Bladder-cancer incidence by estimated average and cumulative benzo[*a*]pyrene exposure levels was evaluated in paving cohorts from Denmark, Finland, Israel, and Norway (see Table 2.1 on-line). An internal comparison showed indications of a trend with average exposure to benzo[*a*]pyrene (*P* for trend not significant) (Burstyn *et al.*, 2007). The overall incidence of bladder cancer in European asphalt workers was similar to that expected (SMR 1.05; Boffetta *et al.*, 2003).

# 3. Cancer in Experimental Animals

Six coal-tar pitches and three extracts of coaltar pitches all produced skin tumours, including carcinomas, when applied to the skin of mice. An extract of roofing-tar pitch had both initiating and promoting activity in separate experiments (IARC, 1985).

# 4. Other Relevant Data

4.1 Mechanistic considerations relevant to the cancer hazards from exposure during roofing and paving with coal-tar pitch

### 4.1.1 Experimental systems

In previous *IARC Monographs* (IARC, 1985, 1987) short-term tests to assess the genotoxicity of coal-tar pitch were reviewed. Coal-tar pitch and roofing-tar emissions were mutagenic in bacteria in the presence of an exogenous metabolic activation system, and in mammalian cells with and without metabolic activation. These agents also induced sister chromatid exchange in Chinese hamster ovary cells and enhanced viral transformation in Syrian hamster embryo cells,

both in the absence and presence of an exogenous metabolic activation system.

Chemical analyses of high-temperature coaltar pitches identified several polycyclic aromatic hydrocarbons that are genotoxic and carcinogenic in experimental studies (IARC, 1985). These include benz[a]anthracene, benzo[b] fluoranthene, benzo[k]fluoranthene, benzo[a] pyrene, dibenz[a,h]anthracene, chrysene, and indeno[1,2,3-cd]pyrene (IARC, 1983, 2010). These polycyclic aromatic hydrocarbons may contribute to the genotoxic and tumorigenic activities of coal-tar pitches.

# 4.1.2 Humans

DNA strand-breaks (measured by single-cell gel electrophoresis; comet assay) and the 8-oxo-deoxyguanosine/deoxyguanosine (8-oxo-dG/dG) ratio (measured by means of HPLC with electrochemical detection) were determined in peripheral blood leukocytes of roofers exposed to dust from coal-tar pitch (coal tar) during removal of existing roofs before applying hot asphalt. When the workers were stratified by 1-hydroxypyrene excretion in the urine, the amount of DNA strand-breaks in their leukocytes increased, and the 8-oxo-dG/dG ratio decreased in a dose-dependent manner (Toraason *et al.*, 2001).

# 4.2 Synthesis

There is strong evidence from experimental data that coal-tar pitch has a genotoxic mechanism of action. There is moderate evidence in humans for a genotoxic mechanism underlying the effects of exposures during roofing and paving with coal-tar pitch, based on one study.

# 5. Evaluation

There is *sufficient evidence* in humans for the carcinogenicity of coal-tar pitch as encountered in paving and roofing. Coal-tar pitch as encountered in paving and roofing causes cancer of the lung.

Also, a positive association has been observed between exposure to coal-tar pitch as encountered in paving and roofing, and cancer of the bladder.

There is *sufficient evidence* in experimental animals for the carcinogenicity of coal-tar pitch.

There is strong evidence from experimental data that coal-tar pitch has a genotoxic mechanism of action. There is moderate evidence in humans for a genotoxic mechanism underlying the effects of exposures during roofing and paving with coal-tar pitch, based on one study.

Coal-tar pitch is *carcinogenic to humans* (Group 1).

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