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Exposure to creosote in the impregnation and handling of impregnated wood

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HEIKKILÄ PR, HÄMEILÄ M, PYY L, RAUNU P. Exposure to creosote in the impregnation and handling of impregnated wood. *Scand J Work Environ Health* 13 (1987) 431–437. The major components of vapors and polycyclic aromatic hydrocarbons in particulate matter were identified and quantified in two creosote impregnation plants and in the handling of treated wood. The vapors were collected on XAD-2 resin (recovery in the range of 82–102 %) and analyzed by gas chromatography. Particulate polycyclic aromatic hydrocarbons were collected on glass fiber filters and analyzed with high-pressure liquid chromatography with a fluorescence detector. The main components of the vapors were naphthalene, methyl naphthalenes, indene, phenol, and its methyl homologues, benzothiophene, diphenyl, acenaphthene and fluorene. The exposure of the workers to vapors varied between 0.1 and 11 mg/m³. The concentrations of particulate polycyclic aromatic hydrocarbons varied between 0.2 and 46 µg/m³. The benzo(a)pyrene concentration was under 0.03 µg/m³, except in manual metal-arc welding and in the boring of railroad ties, where it was 0.24–0.89 µg/m³. In the measurement of creosote vapors, naphthalene could be used as an indicator agent.

Key terms: diphenyl, hydrocarbons, naphthalene, polycyclic aromatic hydrocarbons.

Coal tar creosote is the oldest industrial wood preservative, having been used throughout the world for almost 150 years (23). The preservative value of less-refined coal products was known as early as 1680. The full-cell process was patented by John Bethell in 1828, and the method promoted the usage of coal tar creosote, and, following the development of the empty-cell process, creosote impregnation became profitable by 1920.

Creosote is a brownish-black, oily liquid which is obtained by the fractional distillation of crude coal tars. Coal tars are by-products of the destructive distillation of coal to coke which is used in the manufacture of steel. Creosote is also produced in town-gas processes. The distillation fractions of crude coal tars have been classified into seven fractions on the basis of their boiling ranges (15). Creosote oil may be a blend of various fractions, the boiling range of which is about 200–400°C (19, 23).

Creosotes are mixtures of hundreds of compounds, and, of these, about 100 have been identified and quantified (10, 25). Creosotes have about 20 major constituents with concentrations of more than 1 % (80 % of the total amount of components in an oil). The main components identified are naphthalene and its alkyl homologues. The creosotes used in Finland contain 15–21 % of polycyclic aromatic hydrocarbons (PAH) and 0.03–0.12 % of benzo(a)pyrene (BaP) (10).

The International Agency for Research on Cancer (IARC) has stated that there is sufficient evidence that creosote oils are carcinogenic to experimental animals, but there is limited evidence that creosotes derived from coal tars are carcinogenic in humans (12). When creosote oils were applied to the skin of mice, the mice developed papillomas and carcinomas in many experiments (12, 18, 24). Cutaneous photosensitivity from coal tars has been described by a number of authors (7, 12, 13). Acridine, anthracene, and phenanthrene have been thought to be photosensitizing agents (4). Creosote oils also contain these compounds. Irritation symptoms, pitch warts, skin discoloration, and tearing have been reported to occur among creosote impregnation workers (11, 14). Phenols and some PAH are known to be absorbed through the skin. During the impregnation and handling of impregnated wood, skin contamination is evident when no proper protective clothing is used.

The concentrations of coal tar products in the occupational environment have been measured from samples collected on glass fiber prefilters with silver membrane filters underneath. Particulate matter collected on the glass fiber filters is extracted by cyclohexane or benzene (16, 17). The extracted fraction of particulate matter is called benzene-soluble matter (BSM) or coal-tar pitch volatiles (CTPV), and it is determined by weighing. This method does not identify any constituents of airborne particulate fractions nor sample vapors. Most analyses of PAH in impregnation plants have been limited to the determination of the benzene-soluble matter (21, 22). The method used for this fraction has, however, indicated nonlinearity and a lack of reproducible data when tested with

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creosote (21). Profile analysis of PAH in creosote fumes has only been carried out in two Swedish treating plants (2). The concentrations of creosote vapors have been measured from samples collected in activated charcoal tubes (22) and on XAD-2 resin (2).

In the present study, the major components in creosote vapors have been identified, and the recovery of 12 of them from XAD-2 resin has been accomplished. The concentrations of vapors and PAH in the breathing zone of workers have been measured in creosote impregnation plants and in the handling of impregnated wood.

Materials and methods

Creosote oil was heated to 60°C in a chamber (0.5 m³). Evaporated contaminants were collected simultaneously into an absorption solution (toluene), on silica (SKC 226-48, 150/150 mg), on activated charcoal (SKC 226-01, 50/100 mg) and on XAD-2 resin (SKC 226-30-05, 75/150 mg). The silica was desorbed with diethyl ether, the activated charcoal with carbon disulfide, and the XAD-2 resin with diethyl ether. The samples were analyzed by means of gas chromatography and mass spectrometry.

Recovery of the 12 main components identified in creosote vapors was tested from XAD-2 resin. In a qualitative comparison, the same gas chromatographic peaks could be identified in samples collected with absorption solution, silica, and XAD-2 resin, but in the sample collected on activated charcoal many components were lacking. On the basis of these qualitative results, XAD-2 resin was selected for testing, since personal sampling is difficult with the use of absorption

solution and the humidity in the air may decrease the capacity of silica.

During the study of recovery a glass tube packed with mineral wool was fitted to a XAD-2 tube. The standard mixture in diethyl ether (see table 2 in the Results section) was injected into the mineral wool. The pretube was heated to 80°C, and a total of 5 l of air was sucked through the tubes at a velocity of 0.2 l/min. The adsorption tubes were desorbed with diethyl ether (4 ml) in an ultrasonic bath for 30 min.

The gas chromatographic analyses were performed with a Hewlett-Packard gas chromatograph 5890 with a flame ionization detector. The temperature of the capillary column, 50 m × 0.3 mm filled with polysiloxane phase SE-30, was programmed to proceed from 35 to 270°C at 4°C/min. The compounds were identified by mass spectrometry (Finnigan MAT 8200 and MAT TSQ 4045, electron ionization, mass spectrometry with an INCOS data system). The detection limit for all compounds was approximately 1–5 µg/sample, corresponding to 0.01–0.05 mg/m³ for an air volume of 100 l. A gas chromatogram of a creosote vapor sample collected in an impregnation plant is shown in figure 1. The calculated total concentration of vapors is the sum of concentrations of all the analyzed compounds.

The concentration of particulate PAH was measured from samples collected onto the prewashed (8-h cyclohexane) glass fiber filters at a velocity of 2 l/min. The samples were extracted with cyclohexane (60 ml, 8 h), and the solution was then evaporated to dryness and dissolved ultrasonically in acetonitrile:water (85:15, 1 ml, 4 min). The analysis was completed with reversed-phase high-pressure liquid chromatography and fluorescence detection (Perkin Elmer LS4). The column

Figure 1. A gas chromatogram of creosote vapors collected in the breathing zone of an assistant operator in plant 1. Peak numbers correspond to those in table 1.

was a 100×4.6-mm octadecylsilyl column, and the program was a linear gradient (1 %/min) from 65 % acetonitrile in water (9). A detection limit of 5 pg was achieved for benzo(a)pyrene when the injection volume was 6 µl. This value corresponds to air concentrations of about 8 ng/m³ for a 100-l air sample. The total concentration of PAH is the sum of the concentrations of 11 particulate PAH. Naphthalene and its homologues were not included with the PAH.

Direct exposure to creosote is a consideration within a relatively small population of workers involved in the processing, transporting and installation of treated wood products.

There are seven creosote impregnation plants in Finland. At two of these railroad ties are impregnated, and in the remaining five poles are treated. In impregnation plants about 60–80 persons are exposed to creosote. The impregnation of poles is periodical, depending on exports, but the treatment of railroad ties occupies the workers throughout the year. In the making of switch elements, during the construction and repair of railroads, about 2 000 workers handle timber impregnated with creosote. Railroads are repaired during the summer, but the railroad elements are assembled throughout the year. The rails are welded together by manual metal-arc (MMA) or thermite welding, and about 130 men are employed in the welding of rails in Finland.

The measurements were carried out in two impregnation plants (in one of which railroad sleepers are impregnated, and in the other poles are treated), during the repair of old rails for which molded ties were changed, during the replacement of rails in a railway yard, during the welding of switches, during the making of switch elements in a hall, and during the loading of poles on board ship.

Results

In the chamber study 98 % of the creosote vapors consisted of the agents listed in table 1. The concentrations of compounds 1, 6, 7, 17, 18, 19, 27, and 28 in the samples collected at the workplaces were under the detection limit of the gas chromatographic method (figure 1). The recovery of the 12 main components in creosote vapor from XAD-2 resin varied from 82 to 102 % (table 2).

The total concentrations of creosote vapor and PAH are given in tables 3 and 4. The average concentrations of vapors per workshift varied within the range 0.1–11 mg/m³ during routine operations in the impregnation plants and in the repair and construction of railways. During the cleaning of the creosote warming chamber, carried out annually, and the opening of cylinders, workers were exposed to high peak concentrations.

There were great differences between the exposure levels in the two impregnation plants. These differences can be explained partly by the fact that, in plant 1,

where the cylinder was opened manually, workers remained in proximity of the cylinder for about 30 min/shift. In plant 2, the cylinder was opened automatically, and thus the time spent in the vicinity of the cylinder was restricted to a few minutes. There was also local exhaust suction in plant 2, which was in operation during the loading and unloading of the cylinder.

Naphthalene was the main component of the vapors (figure 2). Its proportion in the total concentration of vapors averaged 52 % in the impregnation plants and 32 % in the handling of treated wood. Naphthalene and the total concentration of vapor showed the linear regression both in impregnation plants ($y = 0.56x - 0.1$) and in handling ($y = 0.32x + 0.1$), the correlation coefficients being 0.99 and 0.97, respectively. The mean concentrations of creosote compounds in the air are

Table 1. Compounds analyzed in the creosote vapors.

Peak ^a	Compound
1	Benzene
2	Toluene
3	Xylenes
4	Trimethylbenzenes
5	Methyl ethyl benzenes
6	Styrene
7	Benzonitrile
8	Phenol
9	Benzofurane
10	Methyl styrenes
11	Indene
12	Cresols
13	Methyl indenenes
14	Xylenols
15	Naphthalene
16	Benzothiophenes
17	Quinoline
18	Isoquinoline
19	Methyl benzonitrile
20	Methyl naphthalenes
21	Diphenyl
22	Dimethyl naphthalenes
23	Ethyl naphthalenes
24	Acenaphthene
25	Dibenzofurane
26	Fluorene
27	Dibenzothiophene
28	Acenaphthylene

^a The numbers correspond to those in figure 1.

Table 2. Recovery of the 12 main components in creosote vapor from XAD-2 resin. The number of samples was five.

Component	Amount injected (µg)	Recovery (%)	
		Mean	SD
Phenol	39.0	98	4
Cresols	34.0	102	3
Xylenols	48.0	100	7
Methyl styrene	28.0	86	11
Indene	18.0	85	5
Naphthalene	51.0	85	9
Diphenyl	11.0	97	2
Dibenzofurane	12.0	98	3
Benzothiophene	17.0	90	5
Quinoline	9.5	90	12
Isoquinoline	8.0	100	18
Fluorene	9.0	82	4

Table 3. Concentrations of vapor and polycyclic aromatic hydrocarbons (PAH) in the creosote impregnation plants.

Measurement site	N ^a	Mean vapor concentration (mg/m ³)	PAH (μg/m ³)	
			Mean ^b	Mean ^c
Plant 1				
Foreman	3	0.9	4.1	0.4
Test borer	1	2.4	11	1.0
Truck driver	3	3.0	11	1.7
Assistant operator	4	9.1	22	2.0
Charworker	3	0.6	3.7	0.3
Operator	3	1.7	24	3.5
Mean	17	3.5	14	1.8
Control room	1	3.5	3	0.8
Opening (10 min)	1	37	106	8.1
Cleaning of chamber	1	71	86	3.4
Tie platers	4	1.7	5.9	0.5
Plating hall	2	2.0	11	0.6
Plant 2				
Test borer	1	1.9	0.9	0.04
Operator	2	1.2	0.5	0.2
Charworker	1	0.5	0.2	0.04
Loader	1	1.0	3.7	0.4
Mean	5	1.3	1.4	0.2
Control room	2	0.8	0.1	0.06
Opening (10 min)	1	35	104	7.5

^a Number of samples.

^b 3–6 aromatic rings.

^c 4–6 aromatic rings.

Table 4. Concentrations of vapor and polycyclic aromatic hydrocarbons (PAH) in the handling of impregnated wood.

Measurement site	N ^a	Mean vapor concentration (mg/m ³)	PAH (μg/m ³)	
			Mean ^b	Mean ^c
Switch element assembly hall				
Element makers				
Doors open	2	4.6	1.3	0.5
Doors shut	2	11	21	1.9
In hall				
Doors open	2	4.7	0.5	0.2
Doors shut	2	9.6	11	0.8
Repairing of railway				
Man on train	1	1	8.1	0.4
Man on bank	1	0.1	2.8	0.3
Spiker	1	0.2	0.8	0.04
Replacement of rails in railway yard				
Thermite welders	2	0.7	5.5	1.3
Element maker	1	3.7	10	0.6
Adjacent to tie stock	1	7.0	46	2.4
MMA ^d welding of switches				
Welders (3 h)	4	0.7	44	21
At 1-m distance	1	0.5		..
Ports				
Stevedores	9	1.0	0.8	0.04

^a Number of samples.

^b 3–6 aromatic rings.

^c 4–6 aromatic rings.

^d MMA = manual metal-arc.

Figure 2. Creosote vapor profiles. (1 = phenol, 2 = methyl styrene, 3 = indene, 4 = benzothiophene, 5 = diphenyl, 6 = acenaphthene, 7 = naphthalene, 8 = methyl naphthalene, 9 = fluorene)

presented in table 5. The concentration range of the main component, naphthalene, was 0.2–5.7 mg/m³ when the higher concentrations during chamber cleaning were excluded.

The main components of the PAH were phenanthrene, anthracene, and pyrene (figure 3). Fluorene was present mainly as vapor. Eleven components were identified

and quantified in the particulate matter. The concentrations of benzo(a)pyrene in the breathing zones of workers varied between ≤0.01 and 0.03 µg/m³ in the treatment plants (table 6). In a hall where switch elements were assembled, the benzo(a)pyrene concentration was 0.02–0.24 µg/m³. The highest level was measured in the breathing zone of the worker respon-

Table 5. The mean concentrations (mg/m³) of creosote vapor components in air. (N = number of samples, MMA = manual metal-arc)

Compound	Impregnation plants			Switch element assembly (N = 8)	MMA welding (N = 3)
	Workers (N = 18)	Openings (N = 2)	Cleaning of chamber (N = 3)		
Toluene	<0.1	1.4	0.4	<0.1	<0.1
Xylenes	0.1	1.2	1.2	<0.1	<0.1
Phenol	0.1	1.1	1.8	<0.1	<0.1
Methyl styrenes	0.2	2.0	1.2	2.7	<0.1
Indene	0.4	2.4	4.2	0.2	<0.1
Cresols	<0.1	0.6	0.2	<0.7	<0.1
Xylenols	0.1	0.6	0.9	0.2	<0.1
Benzothiophene	0.1	1.0	2.8	0.1	<0.1
Isoquinoline	<0.1	0.1	0.4	<0.1	<0.1
Quinoline	<0.1	<0.1	<0.1	0.2	<0.1
Diphenyl	<0.1	0.9	0.9	0.1	0.1
Acenaphthylene	<0.1	<0.1	0.2	<0.1	0.1
Acenaphthene	0.2	0.9	3.0	0.2	0.1
Dibenzothiophene	<0.1	0.1	0.5	<0.1	<0.1
Naphthalene	2.2	16	41	2.6	0.2
Methyl naphthalene	0.3	7.0	6.0	0.8	0.2
Dibenzofurane	<0.1	<0.1	0.7	<0.1	<0.1
Fluorene	<0.1	0.7	2.2	0.1	<0.1
Total	3.7	36	68	7.3	0.7

Figure 3. Polycyclic aromatic hydrocarbon profiles. (1 = fluorene, 2 = phenanthrene, 3 = anthracene, 4 = pyrene, 5 = benzo(a)-fluorene, 6 = chrysene, 7 = benzo(e)pyrene, 8 = benzo(k)fluoranthene, 9 = benzo(a)pyrene, 10 = dibenzo(ah)anthracene, 11 = benzo(ghi)perylene)

Table 6. The mean concentrations ($\mu\text{g}/\text{m}^3$) of polycyclic aromatic hydrocarbons (PAH) in air. (N = number of samples, MMA = manual metal-arc)

Compound	Impregnation plants			Switch element assembly (N = 8)	MMA welding (N = 3)
	Workers (N = 18)	Openings (N = 2)	Cleaning of chamber (N = 3)		
Fluorene	0.9	16	22	0.4	0.1
Phenanthrene	10	61	53	6.5	21
Anthracene	1.0	19	6.0	0.5	1.8
Pyrene	1.1	6.1	3.0	0.6	13
Benzo(a)fluorene	0.31	0.54	0.2	0.06	1.6
Crysene	0.11	0.69	0.2	0.05	2.9
Benzo(e)pyrene	0.03	0.16	0.06	0.07	2.1
Benzo(k)fluoranthene	0.01	0.06	0.02	0.02	0.64
Benzo(a)pyrene	0.01	0.06	0.03	0.04	0.64
Dibenzo(ah)anthracene	<0.01	0.02	<0.03	<0.01	0.15
Benzo(ghi)perylene	<0.01	0.02	<0.03	<0.01	0.16
Total	14	104	85	8.3	44

sible for boring holes in sleepers. During the boring operation, the temperature of the wood may rise above 100°C , as in manual metal-arc welding, for which the highest exposure to benzo(a)pyrene ($0.39\text{--}0.89\ \mu\text{g}/\text{m}^3$) was measured.

Discussion

Creosote vapors and fumes are mixtures of dozens of components. In this study 30 constituents were identified and quantified. Only a few of these agents have been given limit values in workroom air (1). The effects of these components are not well known. Phenol may act as a tumor promoter for PAH (22). A combination of chemicals may have very complex synergistic effects. There is, for example, no evidence that pyrene is carcinogenic, but it has cocarcinogenic effects with PAH (8). Of the vapor phase agents, quinoline is carcinogenic (8).

The majority of the airborne contaminants was in the vapor phase. The proportion of particulate PAH to the total concentration of vapors was below 0.5 % except during manual metal-arc welding, for which it was 3.7 %. The airborne PAH can be both particle-bound and in the vapor phase. Mass equilibrium depends on the boiling point and adsorptive affinity of PAH for the particulate phase. Sampling losses of PAH can occur when air is drawn through filters containing PAH. This effect is of particular importance in the case of compounds which have a notable vapor pressure at room temperature and boiling points below 400°C . The 3- and 4-ring compounds (fluorene, phenanthrene, anthracene) and the 4-ring compound pyrene belong to this group.

Compared to the limit values (1), the concentrations of naphthalenes, indene, and phenols were low, except for short peak values during the opening of cylinders and the cleaning of the warming chamber. In spite of low concentrations, workers complained of symptoms such as respiratory tract and skin irritation (10).

Previously, creosote vapor-phase sampling has not been performed as categorically as it was in this study. Concentrations of xylenes, naphthalene, and its methyl homologues, diphenyl, acenaphthene, anthracene, and phenanthrene have been reported (2, 22). In two Swedish impregnation plants the average concentrations of naphthalene ($2\ \text{mg}/\text{m}^3$) and diphenyl ($0.07\ \text{mg}/\text{m}^3$) were comparable to those of the present findings (20).

Fractions of benzene-soluble matter in coal-tar fumes have been found to contain 20–40 % identified PAH (3, 5). On the basis of these results the concentrations of 40 to $80\ \mu\text{g}/\text{m}^3$ of identified PAH correspond to $0.2\ \text{mg}/\text{m}^3$, which is the threshold limit value of the benzene-soluble fraction (1). The manual metal-arc welders were exposed to PAH levels of $40\ \mu\text{g}/\text{m}^3$, and the assistant operators, the operator in plant 1, and the switch construction personnel to PAH levels of $20\ \mu\text{g}/\text{m}^3$, which is 50 % of the calculated occupational exposure limit. Short peak exposure ($105\ \mu\text{g}/\text{m}^3$) occurred during the unloading and filling of cylinders. The bulk of the inhalation exposure data earlier available on creosote treatment workers was collected by the gravimetric method. At 11 treatment plants the overall average concentration of the benzene-soluble matter was about $0.07\ \text{mg}/\text{m}^3$ (22). If we assume that 20 % of the benzene-soluble fraction consists of identified PAH, the value corresponds to a PAH concentration of $14\ \mu\text{g}/\text{m}^3$.

There is inadequate evidence that 3-ring PAH such as fluorene, phenanthrene, and anthracene are carcinogenic (8). Over 90 % of the particulate PAH consisted of these compounds in our study, except in the manual metal-arc welding process, where 50 % of the particulate PAH consisted of high molecular PAH (4–6 aromatic rings). Benzo(a)pyrene concentrations have generally been used as indicators for PAH. At normal indoor temperatures, benzo(a)pyrene will not evaporate from creosote or treated wood. Only during operations in which the temperature of the creosote or wood rose above 50°C were the benzo(a)pyrene levels found to be elevated. The proportion of

benzo(a)pyrene of the total PAH was 0.04–0.07 % in the impregnation plants, 0.5 % in the switch element hall, and 1.5 % in the manual metal-arc welding of switches. The benzo(a)pyrene concentrations in urban air in Stockholm vary from 0.01 to 0.016 $\mu\text{g}/\text{m}^3$ (6). The benzo(a)pyrene exposure of creosote workers was comparable with these values, provided that the temperature of the creosote or treated wood was not elevated, but exposure to other 4- to 6-ring PAH was higher than that found in urban air.

Factors which exert a greater influence on the vapor and PAH content of the air than the composition of the creosotes used are the work methods, the automation of impregnation, weather conditions, the temperature of the impregnated wood, etc. Due to the linear correlation of naphthalene with the total concentration of vapor, naphthalene could be used as an indicator agent when the exposure of workers to creosote vapors is measured. The concentrations of diphenyl should also be measured.

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