

External and internal exposure to polycyclic aromatic hydrocarbons (PAH) among workers in the production of fire-proof materials – Proposal of a biological monitoring guidance value

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Abstract

In 1999, we introduced the German polycyclic aromatic hydrocarbons (PAH) study. The study was designed as a nation-wide investigation on workers exposed to PAH. One aim of the study was to establish biological monitoring guidance values (BMGVs) for different branches. Here, we report on the production of fire-proof materials. This branch of industry is typically confronted with high exposure to PAH and with PAH-induced occupational (cancer) diseases. One hundred and thirty-five employees participated in the course of seven sampling dates in four different plants in Germany. External exposure was determined by personal air monitoring of the 16 EPA-PAH. Human biological monitoring was accomplished by the determination of 1-hydroxypyrene and monohydroxyphenanthrenes in post-shift spot urine samples.

Concentrations of PAH in the air of the workplaces ranged up to 1102.6 µg/m³. Maximum benzo[a]pyrene concentration was 38.2 µg/m³. The internal PAH exposure of workers was much higher compared with that of the general population. Median concentration for 1-hydroxypyrene was 6.4 µg/g creatinine (maximum 279.6, 90th percentile 23.9 µg/g creatinine) and for the sum of monohydroxyphenanthrene metabolites 13.3 µg/g creatinine (maximum 313.4, 90th percentile 70.8 µg/g creatinine). The following BMGVs for the non-smokers of this branch of industry are proposed: for 1-hydroxypyrene 18 µg/g creatinine and for the sum of hydroxyphenanthrenes 77 µg/g creatinine in urine measured at the end of the shift.

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Introduction

During the production of fire-proof stones and other fire-proof materials, the employees are highly exposed to polycyclic aromatic hydrocarbons (PAH) (Gundel et al., 2000; Marczyński et al., 2005). The main source of

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exposure is coal-tar pitch which is used as binding material.

The carcinogenic activity of many PAH is well known (IARC, International Agency for Research on Cancer; <http://www.iarc.fr>). Occupational exposure to PAH has been associated with cancer of the lung, skin and bladder (Lloyd, 1971; Armstrong et al., 1994; Tremblay et al., 1995; Mastrangelo et al., 1996; Boffetta et al., 1997).

The sole determination of PAH in the air underestimates the actual uptake of PAH because PAH can be absorbed not only by inhalation but also by skin contact. Therefore, it is necessary to establish a human biological monitoring in addition to the determination of PAH in the air in order to assess the actual internal exposure of humans irrespective of the pathway by which the toxicant(s) were taken up.

In 1999, we introduced the German PAH study. The study was designed as a nation-wide investigation on workers exposed to PAH. Determination of external and internal exposure, biomarkers of effect, genotoxic risk assessment of workers and modulation by enzyme polymorphisms are enclosed and some results have been reported elsewhere (Marczynski et al., 2002; Lemm et al., 2004; Marczynski et al., 2005; Mensing et al., 2005; Preuss et al., 2005).

One predominant aim of the study was to establish biological monitoring guidance values (BMGVs) for different branches of industry by using PAH metabolites in order to reduce the carcinogenic risk associated with exposure to PAH at these workplaces. The issue has already been addressed by several institutions; e.g., the American Conference of Governmental Industrial Hygienists (ACGIH) recommends the determination of 1-hydroxypyrene in urine as a biological exposure index (BEI) for assessment of exposure to mixtures containing PAH. 1-Hydroxypyrene was implemented in the threshold limit values (TLV) and BEIs documentation as adopted biological exposure determinant in 2005. However, a BEI value was not evaluated (ACGIH, 2005). The Advisory Committee on Toxic Substances of the British Health and Safety Executive (HSE) has recently evaluated a hygiene-based benchmark guidance value for biological monitoring of PAH exposures by means of 1-hydroxypyrene in urine (HSE, 2003).

Here we report on occupationally PAH-exposed workers in the production of fire-proof materials. The PAH metabolites 1-hydroxypyrene and monohydroxyphenanthrenes were used to assess the internal exposure to PAH.

Materials and methods

Study group

In the course of this nation-wide study, altogether 410 employees from different industrial branches from all over Germany were investigated between 1999 and 2004

concerning their external and internal exposure to PAH at their workplaces.

This paper describes the results concerning the employees from fire-proof materials production plants. The study group consisted of 135 employees (50 non-smokers and 85 smokers) aged from 19 to 62 years (mean: 42 years). All participants gave their informed consent prior to their inclusion into the study. The study was approved by the ethics commission of the Ruhr-University Bochum. Workers participated during seven sampling dates in four different plants. Their working tasks comprised charging, operating and maintaining the mixers, presses, coal-tar saturation plant, electric arc furnaces and product finishing, graphitisation. The main source of exposure to PAH was binding pitch. Almost all of the employees were wearing protective gloves and standard workwear (overalls). In particular cases and at particular workplaces where the dust exposure was high, employees were wearing dust masks. The occupational hygiene was comparable between the different production sites and it can generally be described as normal standard in this branch of industry.

Personal air monitoring of PAH

Particulate-bound PAH and PAH vapours were collected by personal air sampling in the workers' breathing zone by means of glass fibre filters and sorbent tubes filled with XAD-2 (a polystyrene/divinyl benzene-based polymer). The filters and sorbent tubes were stored at -20°C until analysis of the 16 so-called EPA-PAH, namely acenaphthene, acenaphthylene, anthracene, benz[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[ghi]perylene, benzo[a]pyrene, chrysene, dibenz[ah]anthracene, fluoranthene, fluorene, indeno[1,2,3-cd]pyrene, naphthalene, phenanthrene and pyrene. The analysis of the filters and the sorbent tubes was carried out according to the NIOSH method 5506 (NIOSH, 1994). Briefly, the filters were mixed with 5 ml of acetonitrile and shaken for 30 min and then extracted ultrasonically (60 min). Subsequently, the XAD-2 material from sorbent tubes was extracted with acetonitrile (2 ml) and dichloromethane (2 ml). The filter extract and the combined extracts of the XAD-2 material were exposed to a gentle stream of nitrogen to evaporate to dryness. The residues were re-dissolved with acetonitrile, and a 25 μl aliquot was analysed by high-performance liquid chromatography with diode array detection (HPLC/DAD). Monitored wavelengths were 250, 266 and 286 nm. The HPLC/DAD analysis was carried out by an HPLC-System from Merck (Darmstadt, Germany) using a reversed-phase column (LiChrospher PAH 250 \times 3 mm, 5 μm) and a water/acetonitrile (ACN) gradient for separation at 30 $^{\circ}\text{C}$ (0–5 min: 60% ACN; 5–20 min: 60–100% ACN; 20–30 min: 100%

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