



## Emission of polycyclic aromatic hydrocarbons from selected processes in steelworks

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### ABSTRACT

The emission of polycyclic aromatic hydrocarbons from selected processes in steelworks in southern Poland was investigated. Size-segregated samples of air particulate matter (<0.25, 0.25–0.5, 0.5–1.0, 1.0–2.5 and 2.5–10  $\mu\text{m}$ ) were collected at the electric arc furnace and rolling mill. The PAHs were sampled with the personal cascade impactor and identified by HPLC with fluorescence detector. The obtained results showed that collected PAH contents were significantly higher at the electric arc furnace. The highest content of total PAHs (93  $\text{ng m}^{-3}$ ) was present in the smallest particles of 0.5  $\mu\text{m}$  aerodynamic diameter or less, indicating that the ultrafine particles have a high contribution in the overall  $\text{PM}_{2.5}$  fine fraction. Concentrations of Py, CH, BbF, BaP and BghiP came to 76% of total PAHs content in <0.25  $\mu\text{m}$  fraction. The five- and six-ring compounds (BbF, BkF, BaP, DBA, BghiP) with 4-ring chrysene presented typical unimodal size distribution with one predominant peak for this particles' diameter. Phenanthrene and fluoranthene exhibited highest concentrations on coarse particles in the range of 10–2.5  $\mu\text{m}$ , decreasing with decrease of a particle size fraction. Using the toxic equivalent factor (TEF), the mean contributions of the carcinogenic potency of BaP to the air samples collected at the arc furnace and rolling mill (fraction below 0.25  $\mu\text{m}$ ) were determined to be 66.3% and 50.3%, respectively.

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### 1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are contaminants widespread in the environment. Due to the carcinogenicity and mutagenicity of some of these compounds, particularly containing four or more aromatic rings [1–3], they have been included in the European Union priority lists of pollutants [4]. Combustion process of carbonaceous materials at high temperature is one of the major sources of PAHs. Studies concerning their emissions in the process of fossil fuels combustion, organic matters burning, solid waste incineration, wood smoke and motor vehicle sources have been conducted [5–10]. Certain food preparations, such as charcoal grilling, roasting and traditional smoking steps are well known to cause elevated PAHs levels [11]. Once released into the air, gaseous and particle-bound PAHs can be transported over a long distance and transferred to other environmental media such as surface soil and water.

PAHs have been associated with particulate matter (PM), which is a mixture of suspended particles and aerosols, however, three-

to five-ring PAHs have been shown to occur also in the vapor phase [12]. The size of airborne particles determines these parts of the respiratory tract in which the particles are deposited. Currently, there are standards for the mass of particulate matter less than 10  $\mu\text{m}$  in aerodynamic diameter ( $\text{PM}_{10}$ , coarse particles), and less than 2.5  $\mu\text{m}$  ( $\text{PM}_{2.5}$ , fine particles).  $\text{PM}_{10}$  in inhalable fraction are associated with respiratory responses, while  $\text{PM}_{2.5}$  mainly with cardiovascular diseases [13]. PAHs adsorbed onto the respiratory fraction (<5  $\mu\text{m}$ ) of suspended material may penetrate the gas exchange region of the lung causing damage to the alveolar tissues and inducing various health problems [14]. In an urban environment over 80% of particulate matter, in terms of number, is related to fine ( $\text{PM}_{2.5}$ ) and ultrafine ( $\text{PM}_{0.1}$ ) particles, although their total mass is usually insignificant in comparison with the mass of the small number of larger particles [15]. The studies by Li et al. [16] showed that small airborne particles have a high probability to deposit deeper in the respiratory tract. They have also higher burdens of toxins, that adsorbed in the body can result in other health problems than respiratory health effects.

In view of the increasing importance of fine and ultrafine particles for exposure assessment, this study reports the analysis of PAHs in small airborne particles emitted from the selected metallurgical processes. A personal cascade impactor sampler (PCIS) that

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allows separation of airborne particles in several ranges as well as Personal Environmental Monitor (PEM) for measuring PM<sub>10</sub> and PM<sub>2.5</sub> in indoor air were used for the collection of PM.

## 2. Experimental

### 2.1. Sampling sites

The air samples were from a steelworks (in southern Poland) which usually use scrap iron as feedstock. The electric arc furnace was equipped with carbon electrodes to be raised or lowered through the furnace roof. Electric current of the opposite polarity electrodes generates heat between the electrodes and through the scrap. The scrap iron is melted and refined by the electrode and the temperature can reach 1500 °C. The samples were collected close to the electric arc furnace (at 5–6 m distance from a furnace and 1 m from the floor) for 10 h at the flow rate of 9 L min<sup>-1</sup>. About 5400 L of air was passed through four samplers. Ambient-air samples for the particle phase of PAHs were also collected in 4–5 m distance from the rolling mill where bar mills were formed. During sampling process, the plants were operated normally and did not cause any excess variability in emission rate.

### 2.2. The sampling system of PAHs

The particulates were trapped on the 2.5 cm PTFE filters using the personal cascade impactor sampler (PCSI) consisting of four impaction stages. They were separated in the following ranges of particles' aerodynamic diameter: <0.25, 0.25–0.5, 0.5–1.0, 1.0–2.5 and 2.5–10 μm. Particles smaller than 0.25 μm were collected on a 37 mm after-filter made of PTFE (2 μm pore). The air flow was regulated in the range of 5–10 L min<sup>-1</sup> by Leland Legacy pump. Airborne dust particles were also collected using Personal Environmental Monitor (PEM) with 37 mm PTFE filters for PM<sub>10</sub> and PM<sub>2.5</sub> sampling. The gaseous phase was not sampled in the present investigation. After collection, the filters were stored at 0 °C in an ice chest till they were delivered to the laboratory. Subsequently, filters were refrigerated at 4 °C.

### 2.3. Sample extraction and HPLC analysis

The PAHs species determined were the "priority pollutants" identified by the United States Environmental Protection Agency (US EPA), *i.e.*, a 2-ring fluorene (FL), phenanthrene (PH) and anthracene (AN); 3-ring acenaphthene (AC), 4-ring compounds fluoranthene (FLR), pyrene (PY), benzo(*a*)anthracene (BaA) and chrysene (CH), 5-ring benzo(*b*)fluoranthene (BbF), benzo(*k*)fluoranthene (BkF), dibenzo(*ah*)anthracene (DBA) and benzo(*a*)pyrene (BaP) as well as 6-ring benzo(*ghi*)perylene (BghiP) and indeno(*1,2,3-cd*)pyrene (IP).

The PAHs were extracted from the filters with cold dichloromethane (below 5 °C) to prevent losses by volatilization. A 30 min sonic bath with closed vials was used. After extraction, samples were evaporated to dryness under a high purity stream of nitrogen, redissolved in 1 mL of acetonitrile and then analysed according to the method of the National Institute Safety and Health [17]. The standards were prepared by spiking known amounts of the EPA 16PAHs mixture from Supelco onto the filter and the whole analytical procedure including extraction and evaporation step was performed. Therefore, no blank correction was applied.

Separation and identification of PAHs were achieved using HPLC (EliteLaChrom, Merck Hitachi) with fluorescence detection (FL). A reversed phase HPLC column (Pinnacle II PAH, 150 mm × 32 mm, 5 μm particle size) from Restek with a precolumn (20 mm × 3.2 mm) was used for this study. The flow rate was

**Table 1**  
PAH's excitation/emission wavelengths.

PAH	λ <sub>ex</sub> (nm)	λ <sub>em</sub> (nm)
Acenaphthene	280	330
Fluorene	280	330
Phenanthrene	246	370
Anthracene	250	406
Fluoranthene	280	450
Pyrene	270	390
Benzo( <i>a</i> )anthracene	265	380
Chrysene	265	380
Benzo( <i>b</i> )fluoranthene	290	430
Benzo( <i>k</i> )fluoranthene	290	430
Dibenzo( <i>ah</i> )anthracene	290	410
Benzo( <i>a</i> )pyrene	290	430
Benzo( <i>ghi</i> )perylene	290	410
Indeno( <i>1,2,3-cd</i> )pyrene	300	500

0.97 mL min<sup>-1</sup> and the injection volume was 10 μL. Elution was performed with a gradient starting from an eluent composition of 50% acetonitrile in water (up to 5 min). The acetonitrile concentration was increased up to 100% in 25 min, and maintained at 100% for 5 min. The wavelength of the fluorescence detector was altered for each PAH. The excitation/emission wavelengths for each PAH are presented in Table 1. The data was collected and processed with HP ChemStation software.

### 2.4. Quality control and assurance

Procedural blanks were also analysed concurrently with the samples. In blanks none of the detectable PAHs was present. Quantification of PAHs was carried out based on the retention times and peak areas of the calibration standards. At least seven standard concentrations in the range 0.0025–1 mg L<sup>-1</sup> were used in PAHs analysis calibration. Correlation coefficients of the calibration curves were in the range of 0.98–0.99.

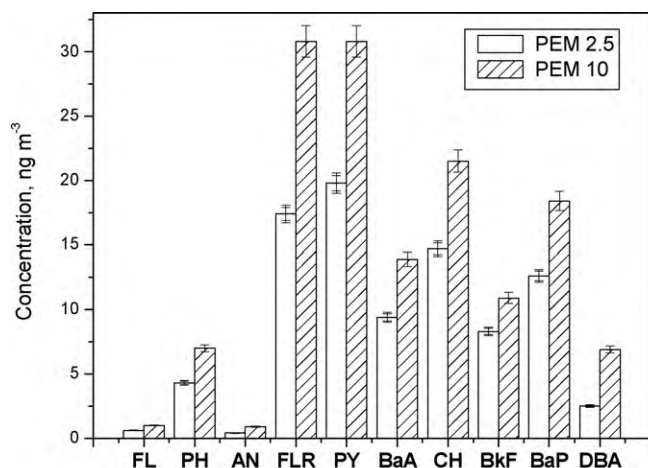
The recovery efficiencies of 14 individual PAHs (EPA 16PAHs mixture excluding the most volatile acenaphthylene and naphthalene) were determined by processing solution with known PAHs concentrations through the same experimental procedures used for the samples. The recovery efficiencies of PAHs, used in this case for the analytical procedure, ranged from 96% to 102%. Mean RSDs of the recovery efficiencies were about 15% (*n* = 4).

The detection limit defined as three times of signal/noise ratio ranged from 0.8 μg L<sup>-1</sup> (phenanthrene, anthracene, pyrene, benzo(*a*)anthracene, chrysene, benzo(*k*)fluoranthene and benzo(*a*)pyrene) to 8.3 μg L<sup>-1</sup> (naphthalene and acenaphthalene) and the limits of quantification were in the range of 2.5–25 μg L<sup>-1</sup>.

## 3. Results and discussion

In the steel and iron industry, PAHs are released from coke manufacturing, sintering, iron making casting, mold poring and cooling as well as steel making [18]. For the same kind of industrial plant, the emission of PAHs is highly dependent on the kind of fuel used in the production process. It was found that the total PAHs emission in the particulate phase (all sizes of particles) increased in the order: coal as a fuel > heating by electric arc > heavy oil as a fuel.

In the presented study the mean content of total PAHs collected at the electric arc furnace during one shift (10 h) was 95.5 ± 7.6 and 135.5 ± 9.1 μg m<sup>-3</sup> in PM<sub>2.5</sub> and PM<sub>10</sub> fractions, respectively. Relative standard deviation (±RSD) was calculated for 4 samples (*n* = 4). The concentrations of 10 individual compounds in these fractions are presented in Fig. 1. The most abundant PAHs in these particles were fluoranthene (FLR) and pyrene (PY), following chrysene (CH), benzo(*a*)pyrene (BaP), benzo(*a*)anthracene (BaA) and



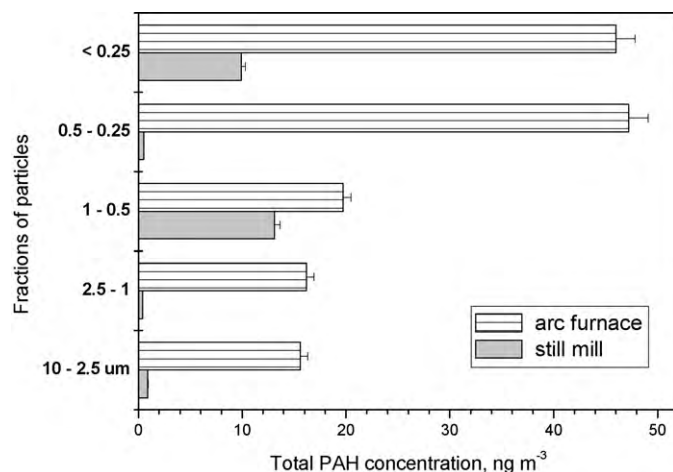
**Fig. 1.** Concentrations of individual PAHs in PM<sub>2.5</sub> and PM<sub>10</sub> fractions (PEM sampler) collected at the electric arc furnace. Bars correspond to mean  $\pm$  RSD ( $n=4$ ).

benzo(*k*)fluoranthene (*BkF*). The sum of FL and PY contributed to 39% and 45.5% of the total PAHs content in the PM<sub>2.5</sub> and PM<sub>10</sub> fractions, respectively. Thus, it appears that PAHs of higher molecular weight are predominant in the electric arc furnace, which corresponds to the results of Yang et al. [19]. The contents of naphthalene (NA), acenaphthene (AC), benzo(*ghi*)perylene (*BghiP*) and indeno(1,2,3-*cd*)pyrene (IP) were below the detection limit of the method. It should be mentioned that NA and AC exist also, and predominantly in the gas phase and the sampling equipment used in this study was inappropriate to collect the most volatile PAHs compounds. In this case, a sampling train to collect suspended airborne particles on a filter and vapor phase compounds on a backup sorbent is recommended. Reported emission of high molecular weight PAHs such as *BghiP* and ID from the steel and iron plants was in the range of 0.34–0.53% (normalized to the sum of PAHs) [18].

Benzo(*a*)pyrene has been used as a good index for the total PAHs carcinogenicity [20]. Recently with the aim of minimizing harmful effects on human health and the environment as a sum of airborne PAHs, the European Union defined a target value for BAP as 1 ng m<sup>-3</sup> for the total content in the PM<sub>10</sub> fraction [21]. The concentrations of BaP in the analysed PM<sub>2.5</sub> and PM<sub>10</sub> samples were found to be 12.6 and 18.4 ng m<sup>-3</sup>, respectively. This indicates that benzo(*a*)pyrene contamination was above the legal limit in EU, which is a worrying factor. Moreover, new findings indicate that DBA has a carcinogenic potency of 10 or more times greater than BaP [22]. Thus, the detection of this compound, despite the lower levels determined (2.5 ng m<sup>-3</sup>) in comparison to BaP (6.9 ng m<sup>-3</sup>), is of special interest.

To determine more in details the size distribution of PAHs in the particles emitted in the steelworks, the personal cascade impactor sampler (PCSI) was used. The particles were separated between the following ranges of aerodynamic diameter: <0.25, 0.25–0.5, 0.5–1.0, 1.0–2.5 and 2.5–10  $\mu\text{m}$ . Distribution of total PAHs content between different sizes of collected particles is presented in Fig. 2. The concentrations of PAHs were much lower for samples collected at the still mill than at the arc furnace.

Considerable amounts of the sum of these compounds at the still mill were found only in 1–0.5  $\mu\text{m}$  (13.1 ng m<sup>-3</sup>) and <0.25  $\mu\text{m}$  (9.9 ng m<sup>-3</sup>) fractions. The highest concentration of PAHs (93 ng m<sup>-3</sup>) was present in the ultrafine particles of 0.5  $\mu\text{m}$  aerodynamic diameter or less, collected at the arc furnace. The obtained results indicate that PAHs present in ultrafine particles have a high contribution in the overall PM<sub>2.5</sub> fraction. It is an interesting issue that at the still mill the highest concentrations of PAHs occurred in the fraction of 1–0.5  $\mu\text{m}$ , which was not observed in similar samples collected at the arc furnace. Probably the differences obtained con-



**Fig. 2.** Distribution of total PAHs content between different size of particles collected in steelworks (PCIS sampler).

cerning PAHs distribution depending on the size of PM were caused by different conditions (e.g. temperatures) of these processes. Fraction PM<sub>2.5</sub> in both processes contained over 80% of PAHs in relation to their content in the PM<sub>10</sub> fraction. Similar values were obtained for samples collected by samplers PEM 2, 5 and PEM 10. The content of PAHs in the fraction PM<sub>2.5</sub> amounts to over 70%. Good compatibility between PEM and PCSI sampling was obtained for PM<sub>2.5</sub> and PM<sub>10</sub> fractions. The total content of PAHs determined using cascade sampler amounts to 128.5  $\pm$  9.6 and 144.3  $\pm$  10.8  $\mu\text{g m}^{-3}$  in PM<sub>2.5</sub> and PM<sub>10</sub> fractions, respectively.

Concentrations of individual PAHs in five particulate fractions collected at the electric arc furnace and rolling mill are presented in Fig. 3. Generally, much higher amounts of individual PAHs were observed once particles were collected at the arc furnace (Fig. 3A). Most targeted PAHs were quantifiable in all size fractions, except for the most volatile compounds such as NA, AC, AN and FL. The five- and six-ring compounds (*BbF*, *BkF*, *BaP*, *DBA* and *BghiP*) and 4-ring chrysene presented typical unimodal size distribution with one predominant peak for fraction lower than 0.25  $\mu\text{m}$ . The peak at this particles size is expected since it is known that PAHs are associated primarily with submicron particles as a result of gas-to-particle transformation after emission [23]. *BaA* was not detected on the smallest aerodynamic diameter particle fraction. Concentrations of *Py*, *CH*, *BbF*, *BaP* and *BghiP* amounts to ca. 76% of total PAHs in that fraction. Phenanthrene and fluoranthene exhibited the highest concentrations on coarse particles in the range of 10–2.5  $\mu\text{m}$ . This concentration decrease with a particle size fraction. Exemplary chromatogram of PAHs emitted with the fraction below 0.25  $\mu\text{m}$  and collected at the electric arc furnace is shown in Fig. 4.

All detected PAHs in samples collected at rolling mill showed predominant distribution in particles of 1–0.5  $\mu\text{m}$  and <0.25  $\mu\text{m}$  size, as it was shown in Fig. 3B. Pyrene is the most abundant compound in these fractions—corresponding to 20% and 22% of  $\Sigma$ PAHs, respectively. The distribution differences of PAHs with respect to the particle size could result from the chemical affinities between the compounds and different size particles, as well as different emission sources and different PAHs reactivity on photooxidation [24]. It is surprising that the fraction of 0.25–0.5  $\mu\text{m}$  diameter for rolling mill sample contains such a small amounts of PAHs comparing to the smaller and larger fractions. At this stage however confirmation and explanation of this situation will require verification with additional studies.

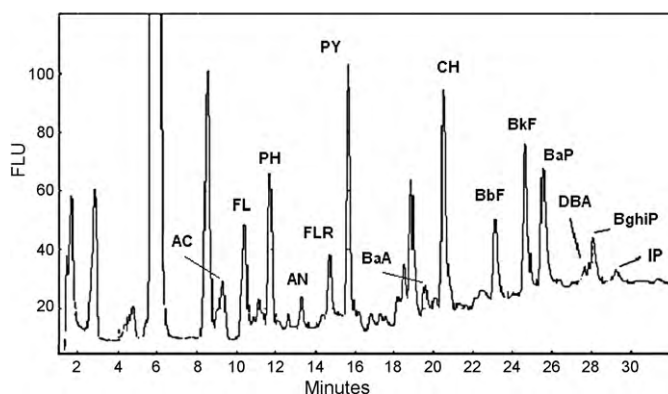
A number of sets for the determination of carcinogenic properties of PAHs mixtures can be found in the literature, although they are all based on a unit value for benzo(*a*)pyrene as the available

**Table 2**  
BaP equivalent concentrations for PAHs in air samples emitted from selected processes in steelwork (fraction <0.25  $\mu\text{m}$ ).

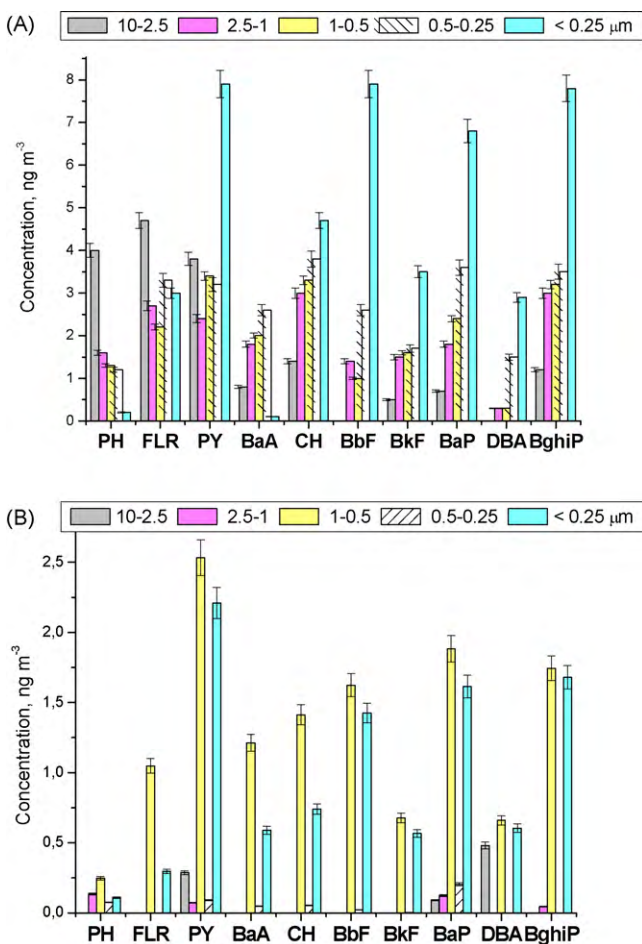
	TEF <sup>a</sup>	Mean concentration ( $\text{ng m}^{-3}$ )		BaP equivalent concentration ( $\text{pg m}^{-3}$ )	
		Arc furnace	Still rolling	Arc furnace	Rolling mill
PH	0.001	0.23	0.109	0.23	0.11
FLR	0.001	3.04	0.296	3.04	0.30
PY	0.001	7.91	2.21	7.91	2.21
BaA	0.1	0.11	0.589	11.0	58.9
CH	0.01	4.68	0.739	46.8	7.40
BbF	0.1	7.92	1.43	79.2	792
BkF	0.1	3.50	0.566	350	56.6
BaP	1	6.82	1.62	6820	1620
DBA	1	2.89	0.603	2890	603
BghiP	0.01	7.81	1.68	78.1	78.1
Sum carcinogenicity				10,286	3219
Contribution of BaP to the total carcinogenic activity (%)				66.3	50.3

<sup>a</sup> Factor by Nisbel and La Goy [25].

toxicology data for this compound are much better than those for any other PAHs [20,25,26]. For the purpose of this paper we used the toxic equivalent factor (TEF), whereby BaP equivalent concentrations for each PAHs are calculated by multiplying the individual PAHs concentration by its corresponding TEF value [25]. Using this approach, the mean contributions of the carcinogenic potency of BaP to the air samples collected in the steelwork (fraction below 0.25  $\mu\text{m}$ ) were determined to be 66.3% (arc furnace) and 50.3% (rolling mill) (Table 2). These results showed the significance of BaP as a surrogate compound for total PAHs in the air. However,



**Fig. 4.** Exemplary HPLC chromatogram of PAHs collected at the electric arc furnace using PCSI sampler (fraction below 0.25  $\mu\text{m}$ ).



**Fig. 3.** Concentrations of individual PAHs in particulate fractions collected at (A) electric arc furnace and (B) rolling mill.

other PAHs also play an important role in the total carcinogenicity of PAHs, such as DBA contributing to 28.1% and 18.7% in samples collected at arc furnace and rolling mill, respectively. The contribution of BbF in the total PAHs carcinogenic activity equals to 24.6% for stilling mill samples.

The results of the presented studies have indicated that similarly to other high temperature processes, PAHs that are part of the emitted particles are mainly present in the ultrafine particles that penetrate the human respiratory tract down to the gas exchange regions.

At this stage however it is recommended to carry out further studies as these particles seem to be the most dangerous to human health.

#### 4. Conclusions

The highest established by EU concentration for BaP, as a target value of  $1 \text{ ng m}^{-3}$  in the atmospheric air, concerns the  $\text{PM}_{10}$  dust fraction.

In case of the working air, Maximum Admissible Concentration (MAC) for BaP established in Poland is much higher and amounts to  $0.002 \text{ mg m}^{-3}$ , for DBA this value equals to  $0.004 \text{ mg m}^{-3}$  and  $0.002 \text{ mg m}^{-3}$  for total PAHs. Considering the working environment these values relate to both the total suspended dust and the gaseous fraction. The results of presented study have shown that ca. 80% of BaP is included in fractions of 1–0.5  $\mu\text{m}$  size. The highest levels of total PAHs concentration were also found in these fractions as well as in the fraction below 0.25  $\mu\text{m}$  diameter. This might suggest that the concentrations of these compounds in the ultrafine particle fraction should be taken into account while establishing

the maximum limit values. Workers exposed to PAHs should be equipped with the personal protective equipment preventing from the infiltration of fine particles to the respiratory tract.

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