

*Original Research*

# Benzo(a)pyrene Emissions in Cities of the Upper Silesia Industrial Area in Southern Poland: 1980-2005

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## Abstract

The aim of our study was to evaluate the levels of benzo(a)pyrene (BaP) in the atmosphere in administrative units of the Upper Silesia Industrial Area, and to analyze their changes in the period 1980-2005.

We calculated the average annual concentrations of benzo(a)pyrene for the administrative units of Katowice, Bytom, Dąbrowa Górnicza, and Ruda Śląska. Join point regression analysis was used to evaluate time trends of BaP pollution levels.

The obtained values of BaP concentrations indicate that the allowable average annual concentration of the compound was significantly exceeded. The highest concentrations of benzo(a)pyrene were recorded in Bytom and Ruda Śląska. Favorable decreasing trends have occurred in all administrative units. The trend persisted until the end of the studied period (excluding Dąbrowa Górnicza). Nevertheless, in 2005 the annual average concentration of BaP still exceeded the Polish norm of 1 ng/m<sup>3</sup>.

**Keywords:** Cities of Silesia agglomeration, level of benzo(a)pyrene, time trends, Poland

## Introduction

The Upper Silesia Industrial Area, especially its central part, is an area of advanced environmental degradation. The pollution levels, which occur in the atmosphere, have been higher here than in any other areas in Poland [1, 2]. Polycyclic aromatic hydrocarbons (PAHs) significantly affect human health. Benzo(a)pyrene (BaP) is considered to be the main representative of these substances and it is proven to be cytotoxic, genotoxic, carcinogenic, teratogenic,

and immunotoxic [3-5]. PAHs are widespread in the environment, so people are commonly exposed. They enter into human organisms by inhalation through the respiratory tract. Most of these compounds are absorbed on powder particles PM<sub>2.5</sub> (particle fraction with aerodynamic diameter less than 2.5 μm), which can easily penetrate lungs [6-8]. The World Health Organization (WHO) states that the most toxic PAHs are benzo(a)pyrene, benzo(k)fluoranthene, benzo(b)fluoranthene, fluoranthene, indeno(1,2,3-c,d) pyrene, and benzo(g,h,i)perylene [9]. In 1992 toxic equivalency factors (TEFs) for particular PAHs were published. Accordingly, BaP was regarded as an index compound,

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while the carcinogenic action of other compounds is calculated in relation to BaP [10]. In Poland, the main health hazard for human health from air pollution occurs in the area of Upper Silesia [11]. According to European Union standards, this area is recognized as ecologically unsafe. The degree of BaP air pollution in the area of Upper Silesia agglomeration has been estimated as very high, which is dangerous for humans [12, 13].

Evaluation of historical trends of BaP emissions in the cities of the Upper Silesia industrial area is useful in showing recent air quality improvement.

Taking the above under consideration, the aim of our study is to evaluate the levels of BaP in the atmosphere in administrative units of the Upper Silesia industrial area, and to analyze their changes in a period of 26 years, i.e. 1980 to 2005.

## Experimental Procedures

The data used in our study was obtained from particular measurement stations located in the Upper Silesia cities. Measurements have been taken in the provincial sanitary and epidemiological station in the city of Katowice, which regularly carries out hygienic evaluation of the environment. The average annual concentration of BaP from 1 January to 31 December in 1980-2005 has been calculated for the administrative units of the Upper Silesia Industrial Area, i.e. Katowice, Bytom, Dąbrowa Górnicza, and Ruda Śląska (Fig. 1).

Between 1980 and 1997, 72 24-hour measurements were taken simultaneously at all measurement stations under random conditions. Suspended powder (fine-grained) was taken using "Staplex" 38-40 m<sup>3</sup>/h suction air

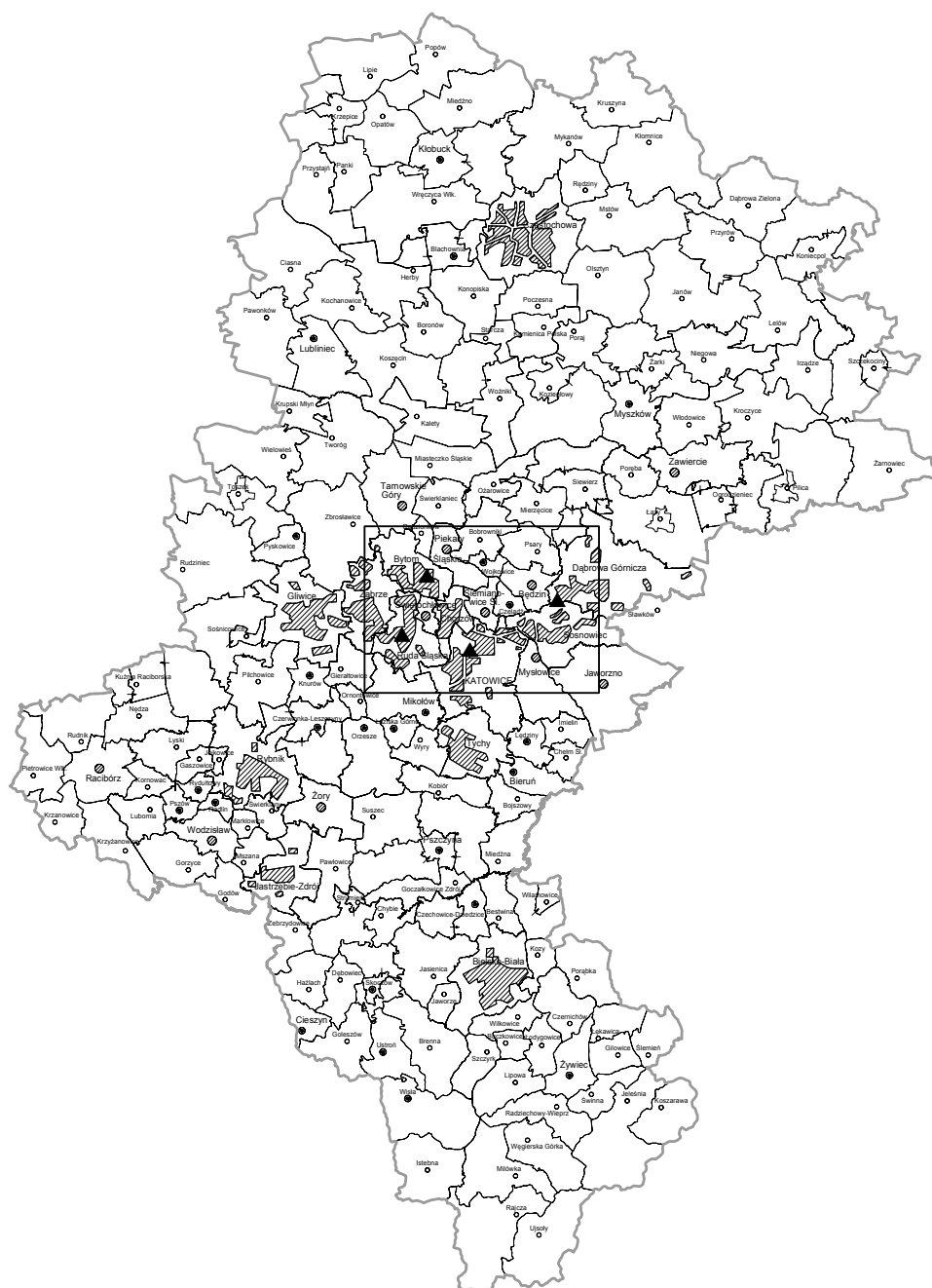


Fig. 1. Administrative units of the Upper Silesia Industrial Area, i.e. Katowice, Bytom, Dąbrowa Górnicza, and Ruda Śląska.

on fiber filters glass type GF/A (Whatman Company) with dimensions 203×254 mm. Pollen count was estimated by weight, based on the difference in mass of the filter before and after sampling. Next, part of the filter was extracted by cyklohexanone in a Soxhelt device. The extract was dissolved in chloroform and submitted to chromatographic analysis in order to estimate the content of BaP using a UV-VIS spectrophotometer in accordance with the norm of the National Institute of Hygiene (NIH) [14]. From 1.1.1998, the samples were taken randomly with a frequency of 10 measurements per month (120 measurements per year.) The suspended powder was measured using a type HVS sampler, equipped with a powder fraction separator with aerodynamic dimension of more than 10 µm, 38-40 m³/h suction air. The powder was kept on the fiber the filters. Suspended powder concentration was estimated by weight and then extracted by cyklohexanone in a Soxhelt device. The extract was estimated by weight as tar substances. Next, chromatographic division was conducted by high-performance liquid chromatography technique (HPLC) in order to estimate content of BaP. The results of BaP concentration measurements were compared with the annual concentration limit of 1 ng/m³.

Throughout the study period (1983-2005) the dust aspirator was calibrated using laboratory calibration of measuring instruments with a current certificate of authentication from the Central Office of Measures.

The measurement stations in these administrative units were located at sites belonging to the Sanitary and Epidemiological Station of Silesia Region. The area is characterized by an average traffic load during the winter, with high pollutant emissions from point sources such as house chimneys and flats, mainly coal-fired.

In administrative units of the Upper Silesia Industrial Area we investigated changes in pollution trends in 1980-2005 using the method of joinpoint regression. This analysis lets us identify, on calendar time scale, the so-called breakthrough points in which the differences in the slope of the linear trend are displayed [15]. The Joinpoint Regression Program chooses best fitted joinpoints where pollution levels change (increase or decrease) in a statistically significant manner. The process of concentration modeling based on yearly BaP concentrations starts with a minimal number of points (zero points means continuous linear trend) and tests if one or more (up to three) points should be added to the model. The final model is the one with the smallest number of points (it can equal 0) in which essential differences of trend occur and the slope of each joinpoint equals the percentage of changes in the annual concentration. Negative and positive values of APC (annual percentage change) show decreasing and increasing trends, respectively. If 95% of APC trust range does not equal 0, we assume that the trend is statistically essential.

The APC is calculated by fitting a least squares regression line to the natural logarithm of the concentrations (BaP) using the calendar year as a regressor variable.

$$y = Ln(\text{concentration})$$

$$x = \text{calendar year}$$

$$y = mx + b$$

$$APC = 100 \times (e^m - 1)$$

(Joinpoint regression program, Version September 2003, National Cancer Institute).

## Results

Based on the analysis performed, the obtained values of the level of BaP in the atmosphere in administrative units of the Upper Silesia Industrial Area in the period 1980-2005 significantly exceeded the Polish norm of annual concentration of 1 ng/m³. The highest concentrations were recorded in the cities of Bytom (1985 – 531.4 ng/m³) and Ruda Śląska (1980 – 366.3 ng/m³) (Table 1).

Table 1. BaP levels (ng/m³) in analyzed cities of Upper Silesia region between 1980 and 2005. Cells with 0.0 indicate missing data.

Year	Katowice	Bytom	Dąbrowa Górnicza	Ruda Śląska
1980	135.5	229.0	174.0	366.3
1981	99.8	180.8	117.0	189.8
1982	110.4	179.6	118.3	148.7
1983	81.5	170.4	99.3	158.7
1984	182.8	313.6	133.5	225.3
1985	189.2	531.4	116.5	212.9
1986	164.4	156.5	107.4	137.0
1987	114	162.0	86.5	159.0
1988	117.8	82.4	110.5	135.8
1989	88.7	164.6	51.5	95.7
1990	125.2	206.0	89.2	68.1
1991	78.6	125.8	49.7	54.7
1992	54.4	86.2	56.4	57.3
1993	67.5	132.7	49.3	79.2
1994	69.1	105.3	35.2	75.6
1995	49.2	76.9	39.5	58.2
1996	53.7	0.0	57.7	43.8
1997	28.3	45.8	20.8	34.0
1998	19.3	23.5	21.7	21.3
1999	15.9	18.6	13.9	17.9
2000	17.0	17.2	5.2	15.8
2001	13.3	13.2	13.5	15.4
2002	11.4	12.8	11.7	13.3
2003	15.5	21.0	23.2	19.0
2004	9.7	11.2	0.0	6.2
2005	7.8	4.1	0.0	8.7

In the 1980s the sanitary conditions in Silesian air were catastrophic because the levels of many pollutants, including BaP, significantly exceeded the norms. Undoubtedly, the method of BaP measurements at that time might have affected the results. It should be stressed that this methodology (UV-VIS spectrophotometer in accordance with the norm of the National Institute of Hygiene (NIH) [14]) was obligatory until 1997. From the beginning of the 1990s a significant decrease in BaP concentration was observed in the area. In the 1990s the range of concentrations was from 13.9 ng/m<sup>3</sup> in Dąbrowa Górnicza (1999) to 206.0 ng/m<sup>3</sup> in Bytom (1990). The levels of BaP in all analyzed cities were over the Polish norm but definitely lower than those in the 1980s. A reduction in the BaP levels in Katowice, Bytom, and Ruda Śląska (Fig. 2) is observed at the end of the study period. In Dąbrowa Górnicza (Fig. 2), however, there has been a significant increase of concentration from 5.2 ng/m<sup>3</sup> in 2000 to 23.2 ng/m<sup>3</sup> in 2003 (2004 and 2005 – no data

available from this location). The highest decrease in the annual BaP concentration over 1980-2005 was found in the area of Ruda Śląska.

Favorable decreasing trends of the BaP level have occurred in all administrative units. In Katowice, the capital of Silesia, in the years 1980-86 an annual increase of 9% per year in the concentration level was recorded, followed by a systematic and statistically significant annual decrease in BaP levels of 14.8% per year. In Ruda Śląska, a very favorable continuous decrease of 13.2% per year has been recorded. In Bytom, an annual decrease of 22% has been observed since the 1990s, whereas it was only 6% in the previous period. Dąbrowa Górnicza is the unit where the change of the trend direction from negative to positive was observed. In the period 1980-96 a statistically significant decreasing trend of 8% was recorded and in the following period (1996-2000) an annual decrease of 35% occurred, after which the annual BaP concentration increased by

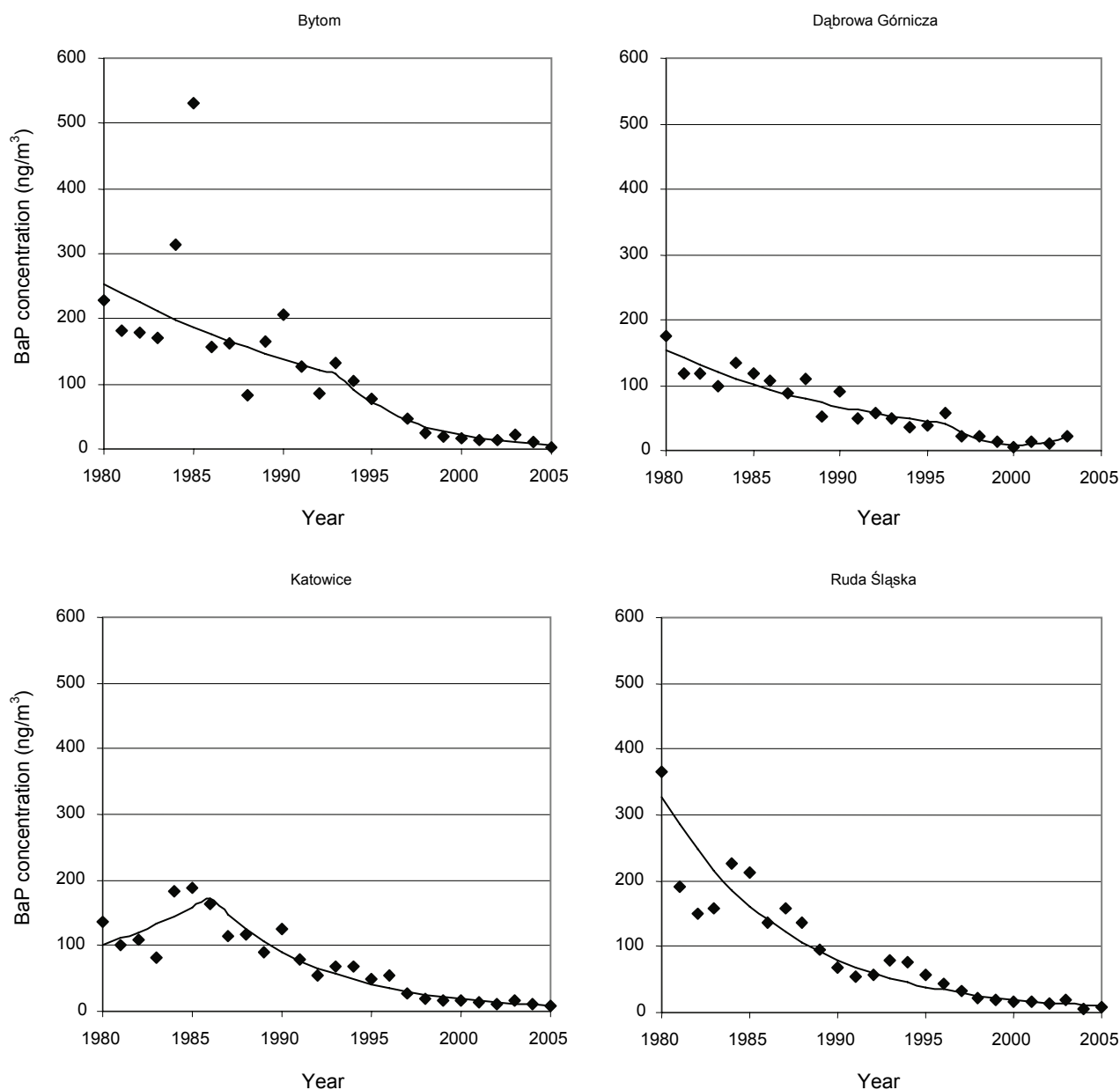


Fig. 2. Variation of BaP (ng/m<sup>3</sup>) pollution levels for Bytom, Dąbrowa Górnicza, Katowice, Ruda Śląska in 1980-2005.

Table 2. Trend statistics of BaP levels in analyzed cities of Upper Silesia region in 1980-2005.

	Trend 1			Trend 2			Trend 3		
	Years	APC	<i>p</i>	Years	APC	<i>p</i>	Years	APC	<i>p</i>
Katowice	1980-1986	9.26	<b>0.164</b>	1986-2005	-14.8	<b>0.00001</b>			
Ruda Śląska	1980-2005	-13.17	<b>0.00001</b>						
Dąbrowa-Górnica	1980-1996	-8.02	<b>0.00001</b>	1996-2000	-35.13	<b>0.013</b>	2000-2003	44.11	<b>0.031</b>
Bytom	1980-1993	-5.92	<b>0.06</b>	1993-2005	-21.67	<b>0.00001</b>			

APC annual percentage change, *p*: significance level

44%. It is undoubtedly a dangerous situation that needs further investigation. At the beginning of the 1990s, a significant decrease in the concentration of atmospheric BaP was observed in the area. The trend persisted until the end of the studied period (excluding Dąbrowa Górnicza city). Nevertheless, in 2005 the annual average concentration of BaP still exceeded the Polish norm of  $1 \text{ ng/m}^3$  (Table 2).

## Discussion

The Upper Silesia Industrial Area occupying the central part of the county of Silesia is the most industrial and most ecologically degraded area of Poland. It is the biggest urban and industrial agglomeration complex in the country, consisting of a number of large cities and industrial areas surrounding them. This influences the landscape and living conditions of inhabitants. The main cause for pollutant emissions here is industry, especially mining and power industries. Heavy industry, largely underdeveloped as well as underinvested, emits enormous amounts of powder and gas into the atmosphere. In view of its environment transformation, the Upper Silesia Industrial Area has been treated as an exceptional district [16, 17]. In the 1990s in Silesia, 55.7 million tons of waste was produced annually, which is about 42% of total national waste. 5.4 k tons was deposited on  $1 \text{ km}^2$ , that is 10 times more than the national average. Additionally, most of the dumping grounds were above-ground and occurred as post mining dumping grounds and metallurgical waste heaps [18]. Moreover, it was the most populated area of Poland with 393 inhabitants per  $\text{km}^2$ .

The introduced analysis in the study, in the period 1980-2005, showed favorable decreasing trends of BaP pollution levels in the areas of Katowice, Bytom, and Ruda Śląska, and a change in the trend from negative to positive in Dąbrowa Górnicza from 2000 (2004 and 2005 – no data available). The major reason for this situation was probably the transformation of the region. For many years Katowice was considered to be a mining and metallurgical industry center. Starting in the 1990s, the production of mines and steelworks was reduced to a large extent and after that more of them were closed down. These changes influenced significantly the reduction of pollution levels in the atmosphere, and led to significant reduction of BaP concentrations. An analogous situation is observed in the areas of

Bytom and Ruda Śląska. The situation in the fourth city is slightly different. In Dąbrowa Górnicza sits the biggest steelworks in Poland – ArcelorMittal Poland, Dąbrowa Górnicza branch and one of the biggest European coke works – Coke Works Friendship. In 1996, the Katowice Special Economic Zone was created. The area of Dąbrowa Górnicza was part of it, and over the years many production and service establishments were created, including Brembo Poland (motorization branch), Ekocem (building branch), Atlas Barbara S.A. (building chemistry), Hobas System Polska (pipe production), and NIKE S.A. (building branch). Such a large number of establishments with different branches may be significant; this may be the main reason for an increase of BaP levels in the area.

In order to verify these assumptions, many measurements should be done in the whole city area and industrial region surrounding it, taking into consideration various elements: summer and winter time (the sources of low emissions from home heating stoves and local coal boiler houses), traffic load on main roads, and weather conditions. These studies have been performed in big city agglomerations in order to estimate people's exposure to pollution [19-22].

Among monitored air pollutants, BaP shows the highest concentration values. It is probably the most hazardous chemical compound to the health of inhabitants of Silesia County. Moreover, in the Silesia area the over-normative average annual values of BaP were recorded also in the 1970s. They were, for example: for Katowice agglomeration –  $118 \text{ ng/m}^3$ , Bytom –  $205 \text{ ng/m}^3$ , Dąbrowa Górnicza  $146 \text{ ng/m}^3$ , and Ruda Śląska –  $272 \text{ ng/m}^3$ . Long-term exposure to BaP is increased by the occurrence of other polycyclic aromatic hydrocarbons (PAH) with similar properties to those of BaP; the acceptable concentration limits for those are not defined [23]. The major anthropogenic emission sources of PAHs are: incomplete burning of mine fuels and wood heating, burning of organic material, waste utilization, and heavy industry connected with coal and oil transformation. Nowadays, the contribution of industry to PAH emissions is systematically decreasing, whereas the main sources of PAHs are toxic fumes and powders coming from low emission sources and fumes from car engines [7, 24, 25]. This is confirmed by results of measurements that show high variations in BaP of concentration in summer as compared to that in the cold seasons [8]. Pollution emitted



by the transportation sector is an increasingly crucial ecological problem in Silesia. The results of a two-year study of PAH pollution status in the air around the region of road infrastructure confirms high health risk for the inhabitants through their exposure to the effects of emissions from substances contained in exhaust gases [7]. High risk occurs especially in the areas directly exposed to traffic emissions, such as trade route canyons, built-up and high-traffic areas, parking lots, and gas stations. It affects mainly cities of Silesia agglomeration [26, 27].

Considering the problem of PAH emissions into the atmosphere (like cigarette fumes), a special source of emission should also be mentioned. Smoking one cigarette brings 20 to 40 ng of BaP into an organism. Smoking 20 cigarettes per day produces about 780 ng of BaP, which means that smokers absorb an additional 16% of BaP from fumes [28]. The amount of global emission of PAHs amounts to about 2 tons per year. It should be remembered that in close spaces the risk of exposure to PAH, by active and passive smokers, is significant [24].

In order to make a comparative analysis of the levels of average annual concentration of BaP in the Silesia area in the period 1999–2005, measurements data from the regions of the so-called Black Triangle area were used. The analysis was included in the reports issued by scientists from three countries (a joint report on air quality in the Tri-border region of the Czech Republic, Poland, and Germany in 1999–2004) [29, 30]. The mentioned area includes: northern Bohemia (Czech Republic), southern (Lower) Saxony (Germany), and the southwestern part of Lower Silesia. The main sources of air pollution in this region are power stations, industrial establishments, social boiler houses, and motor traffic. The data comparison reveals that, in 1999 in the Black Triangle region, values of BaP concentration in the range of 0.20 to 1.56 ng/m<sup>3</sup> were recorded, whereas in the county of Silesia they ranged from 8.8 to 27.6 ng/m<sup>3</sup>. For example, the lowest value of average annual BaP concentration recorded in 1999 and 2004 in Zinnwald was 0.20 ng/m<sup>3</sup>. However, at the same time in Silesia, the lowest concentration was 4.08 ng/m<sup>3</sup>, recorded in Bytom in 2005. The highest value of average annual BaP concentration in Silesia was recorded in 2003 in Dąbrowa Górnicza (23.2 ng/m<sup>3</sup>), whereas in the region of the Black Triangle it was 2.59 ng/m<sup>3</sup> (recorded in 2002 in the city of Jeleniów). As seen in the data, the values of average annual BaP concentration in Silesia agglomeration are significantly higher. Another data comparison is the study of BaP concentration in the industrial and industrial-urban districts in Great Britain in the period 1999–2005 [31]. As seen in the data, in most of the measurement stations in Great Britain the values of average annual BaP concentration did not exceed the value of 1 ng/m<sup>3</sup> approved by European Committee in the “4<sup>th</sup> Daughter Directive” (2004/107/WE) [32].

The highest average annual BaP concentration in that period was recorded in 1999 in Kinlochleven and it was 6.78 ng/m<sup>3</sup>. The sources of BaP in this region were the aluminum recycling establishments. In 2000 the highest average concentration was 2.28 ng/m<sup>3</sup> in Kinlochleven and 1.18 ng/m<sup>3</sup> in Scunthorpe. In Scunthorpe, there is a steelworks

that is the source of pollution emitted into the atmosphere. Also, in the years 2002 and 2003 in Scunthorpe, the acceptable limit was exceeded and the concentration recorded was 1.35 ng/m<sup>3</sup> and 1.26 ng/m<sup>3</sup>, respectively. In 2004 and 2005 there was no record of concentration that exceeded 1 ng/m<sup>3</sup>. The concentration that almost reached that value was recorded in Lisburn in Northern Ireland, where the main origin of pollution was sources of low emission. The values of other measurements are in the range of acceptable values of concentration. In the area of Great Britain, the Expert Panel on Air Quality Standards advised for as low acceptable average annual of BaP as 0.25 ng/m<sup>3</sup> in 2010, in order to lower health risk for the population exposed to BaP [31]. From comparison of the above data with the results of measurements taken in Silesia, it comes out that the values of concentration in the analyzed cities are high in proportion to the values recorded in Great Britain. Based on measurement data from the 1990s, the average area concentration of BaP and PAHs in the atmosphere in different European countries and other regions of the world has been estimated. The above study also takes into consideration the average annual BaP concentration values in the whole country.

It has been estimated that in Europe the lowest average annual BaP concentration occurred in Scandinavia. In Finland it is 0.034 ng/m<sup>3</sup>, in Sweden 0.04 ng/m<sup>3</sup>, and in Norway 0.042 ng/m<sup>3</sup>. Also in Portugal, the recorded average area concentration of BaP is low – 0.067 ng/m<sup>3</sup>. The highest average area concentration of BaP (for a whole country) is recorded in former Czechoslovakia – 2.9 ng/m<sup>3</sup>, in Hungary – 1.9 ng/m<sup>3</sup>, in Germany – 1.5 ng/m<sup>3</sup>, in Poland – 1.4 ng/m<sup>3</sup>, and in Austria – 1.3 ng/m<sup>3</sup> [33]. For example, in 1997 in the Silesia area, the average area concentrations were estimated as: Katowice – 43.3 ng/m<sup>3</sup>, Bytom – 38.9 ng/m<sup>3</sup>, Dąbrowa Górnicza – 26.5 ng/m<sup>3</sup>, and Ruda Śląska – 29.1 ng/m<sup>3</sup>. With these comparisons, it should be stressed that the obligatory regulations about the accepted levels of air pollution do not take into consideration the average area concentration for particular regions. In relation to that, the parameters shown can only be approximately compared with normative values [34]. Recorded in Silesia, the over normative values of BaP concentration, a compound that is thought to be representative of the PAH group are alarming because of the health risk for the population of the area.

In order to improve the atmospheric conditions in the Upper Silesia Industrial Area, first of all the emission needs to be lowered, which means exchanging today's heating methods for pro-ecological ones. Also in industry, old technologies should be replaced by new ones, with less burden on the environment. The significant problem is still motor traffic. Lowering the values of concentration is essential for the sake of the population in that area.

## Conclusions

At the beginning of the 1990s a significant decrease in the concentration of atmospheric benzo(a)pyrene was observed in the agglomeration. The trend persisted until the end of the studied period (excluding Dąbrowa Górnicza city).

Nevertheless, in 2005 the annual average concentration of BaP still exceeded the Polish and European norm of 1 ng/m<sup>3</sup>.

Lower levels of concentrations of BaP is the effect of more frequent use of new technologies and industrial conversion, which has taken place in the area of Silesia during the last 20 years.

Favorable decreasing trends have occurred in all administrative units. Since the mid-1980s, a decrease in BaP annual level of 15% and 13% has been recorded in Katowice and Ruda Śląska, respectively. In Bytom, an annual decrease of 22% has been observed since the 1990s, whereas it was only 6% in the previous period. In Dąbrowa Górnicza a 35% decrease was recorded over the years 1996-2000, but in the following years a 44% increase of annual BaP concentration occurred there.

Evaluation of historical pollution trends in the urbanized and industrialized areas, which include Upper Silesia territory, is essential for long-term air quality analysis.

The data obtained from air monitoring is used, among other things, to translate this knowledge into preventive measures. That is why it is necessary to eliminate or limit the exposure to pollutants of documented pathogenic activity such as BaP.

The abnormalities of BaP concentration levels recorded in the area of Silesia are alarming because of health risks they pose for the inhabitants. We recommend conducting an epidemiological study investigating the impact of BaP on the health of the population.

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