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ASSESSMENT OF INHALATIONAL EXPOSURE OF RESIDENTS OF WROCLAW, KRAKOW AND WARSZAWA TO BENZO[A]PYRENE

OCENA NARAŻENIA INHALACYJNEGO MIESZKAŃCÓW WROCŁAWIA, KRAKOWA ORAZ WARSZAWY NA BENZO(A)PIREN

Abstract: Burning of fossil fuels and biomass, transport and industry are the main sources of PAH (Polycyclic Aromatic Hydrocarbons) in the atmosphere. Benzo[a]pyrene (BaP) is also a compound identified in cigarette smoke. The International Agency for Research on Cancer (IARC) has qualified benzo[a]pyrene for compounds with proven carcinogenic effects on humans. The target value for benzo[a]pyrene, taking into account health protection purposes, is defined in the Regulation of the Minister of the Environment on the levels of certain substances in the air and it is annual average concentration 1 ng/m^3. The aim of the study was to carry out a cancer risk assessment for residents of Wroclaw, Krakow and Warszawa related to inhalation exposure to benzo[a]pyrene in ambient air. The methodology employed by the American Environmental Protection Agency (US EPA) was used in the study. The lifetime exposure of adults and children was assumed. The results of measurements carried out at the air monitoring stations under the State Environmental Monitoring by the Regional Inspectorates for Environmental Protection (RIEP) in Wroclaw, Krakow and Warszawa in the years 2014–2016 were used. The average concentration of BaP in the years 2014–2016 was 3.84, 6.31 and 2.19 ng/m^3 for Wroclaw, Krakow and Warszawa respectively. The calculations show that the highest risk of cancer was obtained for the inhabitants of Krakow: $1.54 \cdot 10^{-5}$ children, $7.52 \cdot 10^{-6}$ women, $6.30 \cdot 10^{-6}$ men. The estimated cancer risk was higher for Krakow residents than for Wroclaw ($1.01 \cdot 10^{-5}$ children, $4.94 \cdot 10^{-6}$ women, $3.82 \cdot 10^{-6}$ men) and Warszawa, where these indicators were the lowest (children: $5.34 \cdot 10^{-6}$, women: $2.61 \cdot 10^{-6}$, men: $2.19 \cdot 10^{-6}$). Children are the group most exposed to the risk of cancer associated with BaP in the air among the examined subpopulations..

Keywords: benzo[a]pyrene, health risk assessment, air pollution

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Introduction

Polycyclic (Polynuclear) aromatic hydrocarbon (PAHs) compounds are a class of complex organic chemicals, which belong to the group of persistent organic pollutants (POPs). Because of their low vapor pressure, some PAHs are present at ambient temperature in air, both as gas and associated with particles. PAHs, such as benzo[a]pyrene (BaP), are almost totally adsorbed on to particles [1]. Benzo[a]pyrene is the most widely investigated PAH as a marker for the carcinogenic risk of PAHs in ambient air [2]. PAHs are characterized by their ability to bioaccumulation, long time of presence in the environment and harmful influence for both, natural environment and human health [3]. PAHs are highly toxic to humans, some compounds are also carcinogenic and mutagenic. Inhalation with the air of PAHs causes the most serious threat for the population's [3, 4]. PAHs compounds are present in the all man's environment components: i.e. water, soil, air and food. These compounds may get into the organism through different paths: dermal contact, digestion, and inhalation [3, 5]. The strong correlation between lung cancer and concentration of the carcinogenic benzo[a]pyrene (BaP) was revealed in the epidemiologic research [3].

Sources of PAHs in the atmosphere are among others: transport, industry, and burning of fossil fuels. Benzo[a]pyrene is also identified in the cigarette smoke [3]. International Agency for Research on Cancer (IARC) has qualified benzo[a]pyrene to compounds for which there is enough evidence to conclude that they can cause cancer in humans [6]. The European directive 2004/107/EC sets a target value (TV) for ambient air concentration of BaP in order to avoid, prevent and reduce harmful effects of this pollutant on human health and the environment as a whole [7]. BaP is measured in PM10 and its target value was set to 1 ng/m^3 as an annual mean. The same level was introduced into Polish legislation. It was estimated [2] that 20 % of the European population is exposed to BaP present in the air in the quantity higher than target value (TV) in the European Union. Only 7 % of the population lives in the areas of estimated permissible BaP concentration equal to 0.12 ng/m^3.

PAHs are formed mainly as a result of pyrolytic processes, especially the incomplete combustion of organic materials, such as coal, wood and petroleum based fuels, often in conditions of local oxygen deficiency [8]. It occurs during industrial and other human activities, such as processing of coal and crude oil, combustion of fossil fuels and including for heating, combustion of refuse, vehicle traffic, cooking and tobacco smoking. BaP is also released naturally from volcanoes and forest fires, but the amounts are very small compared to those released from man-made combustion sources [9]. The commercial, institutional and households fuel combustion sector dominates the emissions of BaP in EU, contributing 71 % of total emissions of this pollutant in 2014. Reported BaP emissions increased by 3 % from 2000 to 2014 [10].

According to the newest Polish emission inventory report published by the National Centre for Emission Management (KOBiZE), total amount of emission of benzo[a]pyrene in Poland was 40.42 Mg in 2015 [9]. The most significant source category is residential sector, mainly household heating systems, which is responsible for 80.11 % of emission. The second large source of national emissions are production processes with coke production as the dominant subsector (18.16 %). Other vehicles and machinery category has the third position and road transport (including passenger cars, light and heavy duty vehicles) had the share of 0.45 % of emission. These values significantly differ from presented in the previous reports, where road transport had about 6 % of emission (e.g. for 2014) [8]. This is the result of the change in the methodology used for estimation of pollutant emissions in the national inventory for mobile sources. The emission has been calculated, for the first time, with the use of software COPERT 4 with use of default transport emission factors. The use of COPERT (Computer programme to calculate emissions from road transport) allows for estimating emissions in accordance with the requirements of international conventions and protocols and EU legislation [9].

In addition other sorts of unorganized emissions, such as stubble burning, burning of plant residues in fields, plots and gardens, incineration of waste and refuse in fires and non-adapted plants lead to deterioration of air quality. PAH emission sources mentioned above are common throughout the country. Because of small height of the emission sources (e.g. house chimney stacks or land surface when burning plant residues in fields and gardens) they lead to high concentrations of PAH on a large area of the country, not only in large cities and agglomerations, but also (and to a considerable extent) in the area of smaller towns and villages with buildings heated individually [8]. In 2015 level of 1 ng/m^3 for annual mean was exceed at 98 % of Polish monitoring stations in all regions of the country [8], so this is a common problem. Higher levels are generally measured in the southern part of Poland. Between 20 % and 24 % of the urban population in the EU countries was exposed to BaP concentrations above 1 ng/m^3 as an annual mean in 2012–2014 [10].

Benzo[a]pyrene concentration data used for analyzes are derived from air quality measurement systems operated by the Regional Inspectorates for Environental Protection. These data series are published on the Polish Air Quality Portal (*http://powietrze.gios.gov.pl*). The annual data series from the period 2011–2016, fulfill the required 33 % coverage time and equal distribution of measurements over the year (tested by the ratio of number of the values from cold to warm season in the range of 0.5–2.0). Averaged annual mean measurement values for the selected urban background stations in three cities (Wroclaw, Krakow and Warszawa – station codes are in the legend of the chart) were analysed as well as the averaged values for all urban stations located in the Polish cities.

Presented indicators are higher than the Target Value set for BaP in the whole of analysed period (Fig. 1). The highest concentrations occurred at the station in Krakow, where they exceeded the concentration levels averaged for Polish cities. This situation occurred in all analysed years, with the exception of 2016. Lower concentrations were recorded in Wroclaw and Warszawa. The data for the latter city indicate a downward trend in BaP concentrations over the period considered, as was the case for Wroclaw, with a slight increase in 2016. In Krakow the tendency was decreasing. It should be noted that the concentration of BaP at this station was in the range of about 5 to 10 times higher than the standard.

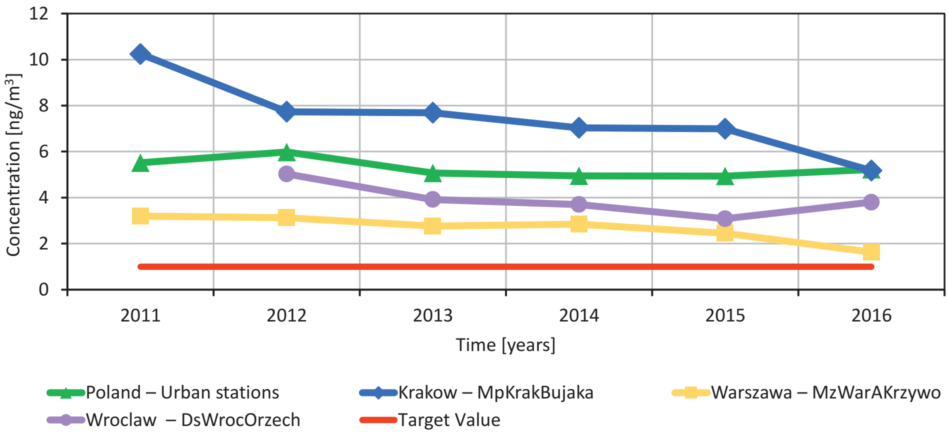

Fig. 1. Changes of annual mean concentration of BaP in selected Polish cities in 2011–2016
Source: own preparation based on SEM (State Environmental Monitoring) data.

Polish cities are characterized by high levels of air pollutants among European cities. Most often mentioned are Krakow and cities of the Silesian agglomeration [11]. Averaged annual mean BaP concentration levels in analysed Polish cities in 2014–2016, measured by State Environmental Monitoring System by the Regional Inspectorates for Environmental Protection (RIEP), are presented in Table 1 [12].

Table 1

Averaged annual mean BaP concentrations in Wroclaw, Krakow and Warszawa

Area	2014	2015	2016
		[ng/m^3]	
Wroclaw agglomeration	3.84	3.57	4.12
		3.84	
Krakow agglomeration	6.89	7.82	4.22
		6.31	
Warszawa agglomeration	2.69	2.22	1.67
		2.19	
Background for Poland [13]		0.55	

Source: SEM data.

Because of the main sources of benzo[a]pyrene, associated with household heating systems, there are very large seasonal differences in the concentration of this pollutant, as evidenced by the example of monthly averages in 2015 (Fig. 2). During warm periods the concentrations are low and similar in the analysed cities. They are significantly higher in the heating season. The differences in the cities included are due

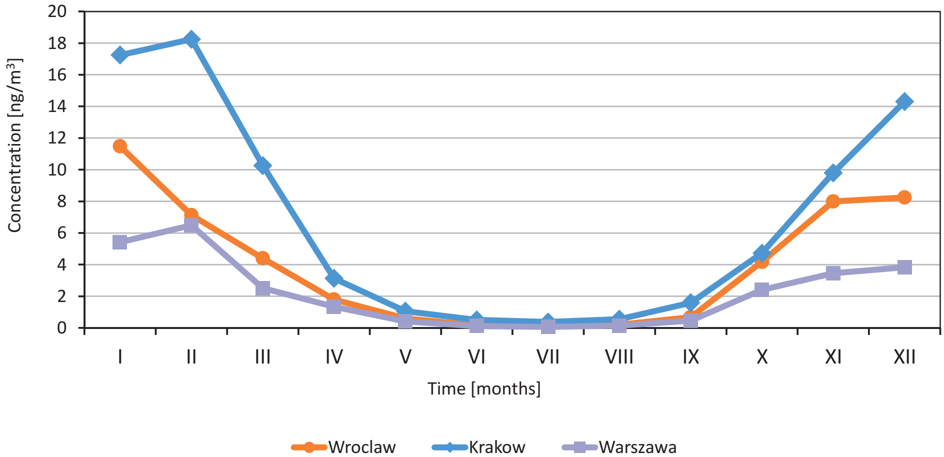

Fig. 2. Variability of monthly average concentrations of BaP in selected cities in 2015
Source: own preparation based on SEM data.

to the influence of the residential heating and the topographic conditions affecting the accumulation of pollutants and possibility of their spread.

Concentrations of BaP determined in the particulate matter, fractions of PM10 and PM2.5, PM1 of selected European cities are presented in table 2 [12]. Basing on the literature only, in case of the Amsterdam city the BaP concentrations in the PM2.5 fraction were not exceeded (according to the target value set for PM10 fraction). In other European cities the exceedance of the target value of 1 ng/m^3 was not reported. Concentration values were between 1.05 and 19.32 ng/m^3.

Table 2

Concentrations of BaP measured in particulate matter in different European cities

Area (country)	Point location	Fraction	BaP [ng/m^3]
Bytom (Poland) – Feb-Mar 2007 [14]	city centre	PM2.5	6.49
		PM10	19.84
Zabrze (Poland) – Oct-Dec 2007 [14]	urban background	PM1	16.08
		PM2.5	19.19
		PM10	19.32
Prague (Czech Republic) – Nov 2002-Jan 2003 [15]	urban background	PM2.5	3.03
		PM10	3.15
Amsterdam (Netherlands) – Jan-Mar 2003 [15]	urban background	PM2.5	0.33
Duisburg (Germany) – Sep-Nov 2002 [15]	urban background	PM2.5	1.05
		PM10	1.10

The aim of the study was to carry out a carcinogenic risk assessment for residents of Wroclaw, Krakow and Warszawa related to inhalation exposure to benzo[a]pyrene in ambient air. Our risk analysis refers to measurement data of the State Environmental Monitoring System by the Regional Inspectorates for Environmental Protection (RIEP) in Wroclaw, Krakow, and Warszawa conducted within the framework of the State Environmental Monitoring (SEM). Risk analysis was carried out with consideration of the lifetime exposure of children and adults (women and men).

Material and methods

In our risk analysis we used the Risk Assessment methodology based on the United States Environmental Protection Agency (US EPA) standards, that includes chronic (lifetime) exposure of the adult and child to benzo[a]pyrene present in the atmospheric air. For the resident scenario, basing on the literature [3, 5] it was assumed that an adult is exposed 24 hours a day for 365 days in the year. For an adult the exposure duration was assumed to be 70 years and for a child 6 years [3, 16, 17].

Estimated daily intake (EDI) of the benzo[a]pyrene during outdoor air inhalation, defining the quantity of the compound penetrating into the body during the day (24 hours) in conversion to 1 kg of the body weight, was calculated using the equation (1).

$$EDI = \frac{C \cdot IR \cdot AF \cdot F \cdot ED}{BW \cdot AT} \tag{1}$$

where: EDI – estimated daily intake [mg/d · kg];
C – mean compound concentration in the air [mg/m^3];
IR – intake rate [m^3/d];
AF – bioavailability factor [unitless];
F – frequency of exposure [d/year];
ED – exposure duration [year];
BW – average body weight [kg]
AT – averaging time [d].

Basing on the literature following values were taken for calculations: average body weight for woman: 65.4 kg, for man: 78.1 kg, for a child: 16 kg. The above values are based on the 50th percentile of the body mass distribution in anthropometric data [3, 17]. As intake rate per day (24 hours) for inhalation it was taken: for an adult: 20 m^3/d, for a child: 10 m^3/d. The other parameters were following: frequency of exposure: 365 days in year, 24 hours per day; bioavailability factor equal to 1. Assuming the same parameters as in the literature [3, 17, 18] let to compare with each other values calculated for Wroclaw agglomeration, Krakow agglomeration and Warszawa agglomeration. The mean BaP concentration was defined as arithmetic mean from the last three years of the SEM monitoring measurements i.e. 2014–2016 [12]. The reference value was the environmental background level for Poland equal to 0.55 ng/m^3. Data were obtained from the *European Commission DG Environment* report [13]. Carcinogenic risk of inhalation exposure to benzo[a]pyrene was calculated using the equation (2).

$$CR = EDI \cdot SF_{\text{BaP}} \tag{2}$$

where: CR – carcinogenic risk;
EDI – estimated daily intake [mg/(d · kg)];
SF_{BaP} – carcinogenic slope factor for BaP.

Carcinogenic slope factor for BaP was taken from the toxicological database of the Office of Environmental Health Hazard Assessment (OEHHA): inhalation exposure $3.9 \cdot 10^0$ (mg/kg · day)$^{-1}$ [19]. Values received in calculation were compared with the acceptable carcinogenic risk level of $1 \cdot 10^{-6}$ [3, 17].

Results and discussion

Using the US EPA Risk Assessment method [16] allows to estimate the existing and predicted health risks at a given concentration of an air pollutant. In the study it was assumed that for carcinogenic risk values resulting from inhalation exposure to BaP in atmospheric air in the Wroclaw, Krakow, and Warszawa affect low emission (households – local boiler rooms), road transport, as well as, unfavourable, from the point of view of pollutants dispersion, landform. Concentration of the benzo[a]pyrene in the given area affects the daily intake value.

In investigated Polish cities measured benzo[a]pyrene concentrations were in range from 2.19 ng/m^3 in Wroclaw to 6.31 ng/m^3 in Krakow. Estimated daily intake (*EDI*) values of BaP during inhalation in investigated Polish cities for men, women and children are given in Table 3.

Table 3

Estimated daily intakes of BaP during outdoor air inhalation in Wroclaw, Krakow and Warszawa

Area	Mean BaP concentration [ng/m^3]	BaP estimated daily intake [mg/(d · kg)]		
		children	adults	
			women	men
Wroclaw agglomeration	3.84	$2.59 \cdot 10^{-6}$	$1.27 \cdot 10^{-6}$	$9.78 \cdot 10^{-7}$
Krakow agglomeration	6.31	$3.94 \cdot 10^{-6}$	$1.93 \cdot 10^{-6}$	$1.62 \cdot 10^{-6}$
Warszawa agglomeration	2.19	$1.37 \cdot 10^{-6}$	$6.70 \cdot 10^{-7}$	$5.61 \cdot 10^{-7}$
Background for Poland [13]	0.55	$3.44 \cdot 10^{-7}$	$1.68 \cdot 10^{-7}$	$1.41 \cdot 10^{-7}$

Basing on our calculations for the investigated cities it can be stated that the highest daily intake of BaP during inhalation received children ($3.94 \cdot 10^{-6}$ mg/(d · kg)), which belong to the most exposed to air pollution subpopulation. The highest BaP daily intake was estimated for Krakow agglomeration inhabitants and the value was about nine times higher than environmental background value for Poland. The second highest daily intake received children and inhabitants of the Wroclaw agglomeration. The influence on air pollutants concentrations have transport and low emission.

Basing on the estimated daily intake (*EDI*) values in Wroclaw, Krakow and Warszawa agglomerations, the carcinogenic risk (*CR*) values were calculated (Table 4). Among investigated subpopulations the highest potential carcinogenic risk (CR) values were assessed for Krakow agglomeration inhabitants.

Table 4

Carcinogenic risk (*CR*) of BaP during outdoor air inhalation in Wroclaw, Krakow and Warszawa

Area	*CR*		
	children	adults	
		women	men
Wroclaw agglomeration	$1.01 \cdot 10^{-5}$	$4.94 \cdot 10^{-6}$	$3.82 \cdot 10^{-6}$
Krakow agglomeration	$1.54 \cdot 10^{-5}$	$7.52 \cdot 10^{-6}$	$6.30 \cdot 10^{-6}$
Warszawa agglomeration	$5.34 \cdot 10^{-6}$	$2.61 \cdot 10^{-6}$	$2.19 \cdot 10^{-6}$
Background for Poland	$1.34 \cdot 10^{-6}$	$6.56 \cdot 10^{-7}$	$5.49 \cdot 10^{-7}$

Risk level considered to be acceptable or insignificant according to the US EPA is equal or less than $1 \cdot 10^{-6}$, whereas unacceptable risk level that absolutely requires protective and remedial actions is equal or higher than $1 \cdot 10^{-3}$ [3, 16, 17]. Mean BaP concentrations in years 2014–2016 for Wroclaw, Krakow and Warszawa were equal to: 3.84; 6.31 and 2.19 ng/m^3, respectively. Our calculations showed that the highest carcinogenic risk (*CR*) values were assessed for Krakow inhabitants: children: $1.54 \cdot 10^{-5}$, women: $7.52 \cdot 10^{-6}$, men: $6.30 \cdot 10^{-6}$. Obtained *CR* values were higher for Krakow inhabitants than for Wroclaw (children: $1.01 \cdot 10^{-5}$, women: $4.94 \cdot 10^{-6}$, men: $3.82 \cdot 10^{-6}$) and Warszawa (children: $5.34 \cdot 10^{-6}$, women: $2.61 \cdot 10^{-6}$, men: $2.19 \cdot 10^{-6}$). Calculated carcinogenic risk values for three agglomerations were also higher than background value for Poland (children: $1.34 \cdot 10^{-6}$, women: $6.56 \cdot 10^{-7}$, men: $5.49 \cdot 10^{-7}$). Carcinogenic risk values obtained for Wroclaw agglomeration were lower than reported in research conducted in years 2008–2010, when potential carcinogenic risk resulting from exposure to benzo[a]pyrene for Wroclaw agglomeration inhabitants was equal to $10.1 \cdot 10^{-6}$ for children, $4.13 \cdot 10^{-6}$ for women, and for men was higher and amounted to: $4.93 \cdot 10^{-6}$ [3]. In research conducted for Wroclaw agglomeration inhabitants in years 2012–2013 carcinogenic risk values during inhalation amounted to: children: $2.69 \cdot 10^{-6}$, women: $1.31 \cdot 10^{-6}$ and men: $1.10 \cdot 10^{-6}$ [17]. Risk values calculated for BaP in Krakow agglomeration in years 2008–2010 were following: children: $15.13 \cdot 10^{-6}$, women: $7.42 \cdot 10^{-6}$, men: $6.21 \cdot 10^{-6}$ and in Warszawa agglomeration: children: $10.1 \cdot 10^{-6}$, women: $4.93 \cdot 10^{-6}$, men: $4.13 \cdot 10^{-6}$ [3]. Mean carcinogenic risk vales for BaP inhalation exposure calculated for Krakow agglomeration inhabitants in years 2007–2016 were following: children $2.00 \cdot 10^{-5}$, infants: $1.44 \cdot 10^{-5}$, women: $9.77 \cdot 10^{-6}$ and men: $8.18 \cdot 10^{-6}$ [18]. Moreover, other exposure pathways (i.e. dermal contact and ingestion of PM) are important factors of the total risk value [20]. Calculations made in our study indicated that in investigated cities air protection programs are required to improve the air quality and for inhabitants' health protection.

Conclusions

Calculated carcinogenic risk values were higher than environmental background level for Poland and comparable with those results that were reported recently in the literature. The most exposed to inhalation carcinogenic risk subpopulation were children due to twice the air exchange in their lungs comparing to adults. Obtained values point the potential carcinogenic risk. Other exposure pathways i.e. dermal contact and ingestion are required to assess the complete impact of air pollutants for human health.

The increase of the pollutant concentrations in the air affect mainly anthropogenic sources, with high contribution of low emission associated with burning of the low quality fuels in households.

Acknowledgement

The publication of paper has been supported by the project "LIFE-APIS/PL – Air Pollution and biometeorological forecast and Information System" LIFE12 ENV/PL/000056 (co-financed by European Union and by NFOSiGW within LIFE+ Program).

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Abstrakt: Źródłami WWA (wielopierścieniowe węglowodory aromatyczne) do atmosfery są m.in. transport, przemysł oraz spalanie paliw kopalnych. Benzo[a]piren jest także związkiem identyfikowanym w dymie papierosowym. Międzynarodowa Agencja Badań nad Rakiem (IARC) zakwalifikowała benzo[a]piren do związków o udowodnionym działaniu rakotwórczym na człowieka. Dopuszczalne stężenie benzo[a]piren ze względu na ochronę zdrowia określono w Rozporządzeniu Ministra Środowiska w sprawie poziomów niektórych substancji w powietrzu i wynosi ono 1 ng/m^3. Celem pracy było przeprowadzenie oceny ryzyka nowotworowego dla mieszkańców Wrocławia, Krakowa i Warszawy związanego z narażeniem inhalacyjnym na benzo[a]piren w powietrzu atmosferycznym. W pracy zastosowano metodykę Amerykańskiej Agencji Ochrony Środowiska (US EPA). W badaniach założono narażenie całożyciowe osoby dorosłej oraz dziecka. W pracy wykorzystano wyniki pomiarów przeprowadzanych w ramach Państwowego Monitoringu Środowiska przez Wojewódzkie Inspektoraty Ochrony Środowiska (WIOŚ) we Wrocławiu, Krakowie oraz w Warszawie w latach 2014–2016. Średnia wartość stężenia B[a]P w latach 2014–2016 wyniosła dla Wrocławia, Krakowa i Warszawy odpowiednio: 3,84, 6,31 i 2,19 ng/m^3. Z przeprowadzonych obliczeń wynika, iż najwyższą wartość ryzyka nowotworowego otrzymano dla mieszkańców Krakowa: dzieci: 1,54 $\cdot$ 10^{-5}, kobiety: 7,52 $\cdot$ 10^{-6}, mężczyźni: 6,30 $\cdot$ 10^{-6}. Wyznaczone wartości ryzyka nowotworowego były wyższe dla mieszkańców Krakowa niż Wrocławia (dzieci: 1,01 $\cdot$ 10^{-5}, kobiety: 4,94 $\cdot$ 10^{-6}, mężczyźni: 3,82 $\cdot$ 10^{-6}) i Warszawy, gdzie wyznaczone wartości ryzyka były niższe (dzieci: 5,34 $\cdot$ 10^{-6}, kobiety: 2,61 $\cdot$ 10^{-6}, mężczyźni: 2,19 $\cdot$ 10^{-6}). Wśród badanych populacji najbardziej narażone na ryzyko nowotworowe związane z obecnością B[a]P w powietrzu są dzieci.

Słowa kluczowe: benzo[a]piren, ocean ryzyka zdrowotnego, zanieczyszczenia powietrza