Original Article

Health Risk Impacts of Exposure to Airborne Metals and Benzo(a)Pyrene during Episodes of High PM_{10} Concentrations in Poland

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Abstract

Objective To check whether health risk impacts of exposure to airborne metals and Benzo(a) Pyrene during episodes of high PM_{10} concentrations lead to an increased number of lung cancer cases in Poland.

Methods In this work, we gathered data from 2002 to 2014 concerning the ambient concentrations of PM_{10} and PM_{10}-bound carcinogenic Benzo(a)pyrene [B(a)P] and As, Cd, Pb, and Ni. With the use of the criterion of the exceedance in the daily PM_{10} mass concentration on at least 50% of all the analyzed stations, the PM_{10} maxima's were selected. Lung cancer occurrences in periods with and without the episodes were further compared.

Results During a 12-year period, 348 large-scale smog episodes occurred in Poland. A total of 307 of these episodes occurred in the winter season, which is characterized by increased emissions from residential heating. The occurrence of episodes significantly (P < 0.05) increased the concentrations of PM_{10}-bound carcinogenic As, Cd, Pb, Ni, and B(a)P. During these events, a significant increase in the overall health risk from those PM_{10}-related compounds was also observed. The highest probability of lung cancer occurrences was found in cities, and the smallest probability was found in the remaining areas outside the cities and agglomerations.

Conclusion The link between PM pollution and cancer risk in Poland is a serious public health threat that needs further investigation.

Key words: Poland; Episodes; Smog; PM_{10}; Metals; B(a)P; Lung cancer; Administrative distribution; Monitoring stations

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INTRODUCTION

In Poland, excessive airborne particulate matter (PM) pollution (often referred to by the mass media as PM smog phenomenon) has been occurring for decades[1]. The problem is so serious that Poland has now become the smog capital of Europe. The main source of Polish smog is the so-called municipal emission, more specifically, burning fossil fuels and biomass for residential heating. In some periods and/or selected locations, traffic emissions can be a significant source of

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smog[2-3]. Unlike the energy sector of most European Union (EU) countries, the Polish energy sector is almost totally dependent on coal, which is the main source of primary PM and precursors of secondary atmospheric particulates[4]. Although Poland has undergone a significant industrial transformation in the last 20 years, with a marked decrease in PM pollution, PM-related smog episodes are still frequent, especially in the southern part of the country[1-2,5]. The phenomenon of PM smog (which can be physically felt as a noticeable burnt odor or manifests as visibility reduction) occurs under unfavourable meteorological conditions, such as atmospheric stagnation, low wind speed, and high relative humidity[6]. In Poland, smog is observed mostly in the winter season under strong ground-based temperature inversions. Despite conditions that favor pollutant dispersion, PM exceedances are sometimes caused by unusually high local emissions, such as those in densely built-up residential areas, where houses are equipped with outdated and inefficient boilers. Smog episodes can vary and include local smog, which covers small-range areas and is often associated with specific emission events[7]; regional smog, which covers medium-sized regions, like industrial districts or cities, and includes instances such as the famous Ruhr District episode in 1962[8] or the London episode in 1952[9]; and also, most interestingly, mesoscale smog, in which the impacted area is extended to metropolitan scale or even covers a whole country[10]. Results of epidemiological and clinical studies indicate that high concentrations of $PM_{2.5}$ and PM_{10} in Poland are correlated with the total mortality rate due to respiratory and blood diseases[11-12]. More than 42,000 premature deaths are attributable to the excessive concentrations of $PM_{2.5}$[13]. Recent findings show that Polish cities struggle with increased incidences of PM-induced lung cancer (from 9.6 to 22.8 cases per 100,000 inhabitants in cities)[14]. So far, there has been no indication of the extent to which PM concentration or its chemical properties [e.g., the content of toxic compounds, some metals, and polycyclic aromatic hydrocarbons (PAHs)] are responsible for the occurrence of lung cancer effects in the population. Although an increase in the frequency of these effects is usually hardly observable, the chronic character of exposure to those pollutants allows us to infer that PM episodes have a significant impact on human health[15]. The extent to which the incidence of smog events, especially on a large scale, increases the probability of negative health outcomes, is currently unclear. Given the lack of a threshold in PM concentration below which no cancer effects will occur, even low concentrations of PM-bound carcinogens may lead to the development of lung cancer. The purpose of this work is to answer to the following question: To what extent does the occurrence of the large-scale PM_{10} smog episodes in Poland elevate the concentrations of PM_{10}-bound ambient carcinogens As, Cd, Ni, Pb, and Benzo(a)pyrene [B(a)P]?

We also aim to check whether these situations lead to an increased number of lung cancer cases in the general population. In this work, we compared the magnitude of inhalation exposure to these pollutants between three types of areas, namely, Polish cities, large urban agglomerations, and the remaining areas.

METHODS

Selection of Episodes

Our analysis was performed based on data concerning PM_{10} and PM_{10}-bound As, Cd, Ni, Pb, and B(a)P concentrations, which were measured across the whole country between 2002 and 2014 under the Polish National Air Monitoring Program. The number of air monitoring stations that provide such measurements is presented in Table S1 (available at www.besjournal.com). The episodes were selected from a series of daily (24 h) PM_{10} concentrations registered within the mentioned period. At present, clearly defined thresholds of PM_{10} daily concentrations that classify a given day as a smog episode are not available[16]. Although a couple of examples are available where these thresholds were defined by appropriate statistical processing of the daily data series[17-18], none of them can be treated as a gold selection standard. Before setting the selection criteria for these episodes, each series of the PM_{10} concentrations from each monitoring site was directed to distribution testing by using the Shapiro-Wilk test (P = 0.05). The assumption was that if the log-normal distribution was prevailing (i.e., more than 50% of all cases), then the median value (or arbitrarily defined percentile) of the annually averaged daily concentrations could be set as a criteria of episode occurrence in a given location[10]. If, however, the distribution was mostly normal (Gaussian), then each PM_{10} concentration that exceeded the value of the arithmetic mean from the annually averaged daily concentrations should be treated as an episode[19]. A summary of the results from this analysis is given in Table 1. In 32% of all the tested cases, the statistical distribution of PM_{10} was log-normal ($P < 0.05$). The lack of a clear dominance of any of the tested distributions did not allow us to use the statistical method as a criterion for episode selection. Therefore, in accordance with the literature[20], the exceedance of the average daily PM_{10} concentration above 50 μg/m^3 (the 24 h threshold value) was set as a criterion for qualifying this as an episode day. We looked for large-scale episodes only. Thus, we assumed that country-level smog occurs when the 50 μg/m^3 concentration was exceeded in at least 50% of all monitoring stations at the same day. On the basis of these criteria, 348 days were selected as large-scale smog events in the 2002-2014 period. This finding means that during the 12-year period, more than 50% of all air monitoring stations in Poland registered a PM_{10} daily concentration of above the permissible level of 50 μg/m^3 over a total of 348 days. The greatest number of such episodes occurred in 2011, with 51 episodes, and the least number of episodes occurred in 2004, with only one episode (Table 1). Assuming that the winter season in Poland lasts from November to March, 307 episodes were assigned to the winter/cold season (the heating season).

In the second part of the analysis, we prepared the database, including the average daily concentrations of PM_{10}-bound As, Cd, Pb, Ni, and B(a)P measured in Poland between 2002-2014. This database was divided into two parts: the first part covers the days of episodes, and the second part covers the rest of the measurement period. In each part of the data, the average daily concentrations were spatially aggregated within agglomerations (cities within provinces with more than 250,000 residents), cities within provinces and outside the agglomerations with a total population of more than 100,000, and areas located within provinces but lying outside the cities and agglomerations (hereafter referred to as remaining areas for more details, see the Supplementary materials available at www.besjournal.com).

Annually averaged concentrations of PM_{10}- bound As, Cd, Ni, Pb, and B(a)P were further compared with the EU air quality standards concerning human health protection, namely, EU Directive 2004/107/EC[22] and air quality recommendations[23-24]. Significant differences

Table 1. Results of Testing the Probability Distribution of PM_{10} Daily (24 h) Concentrations; the Number of Recorded Episodes and the Season of Their Occurrence

Monitoring Period	Total Number of Air Monitoring Stations*	Number of Stations at Which the Daily PM_{10} Data Series Met the Criterion of Normal Distribution	Number of Stations at Which the Daily PM_{10} Data Series Met the Criterion of Log-normal Distribution	Number of Summer/Winter Spisodes Within Calendar Year	Number of PM_{10} Episodes in Each Calendar Year
2002	17	0	9	6/7	13
2003	54	0	22	0/7	7
2004	121	0	44	0/1	1
2005	166	1	68	6/11	19
2006	163	0	59	1/23	24
2007	173	0	64	2/12	14
2008	164	0	56	2/10	12
2009	167	0	60	7/20	26
2010	150	0	35	0/43	43
2011	143	0	30	5/46	51
2012	151	0	14	2/51	53
2013	141	0	52	5/40	45
2014	158	0	53	4/36	40

Note. *All Polish air monitoring stations that provide the measurements of PM_{10} concentration by using automated or manual methods from http://powietrze.gios.gov.pl[21] (Supplementary materials available at www.besjournal.com).

in the mean concentrations of metals and B(a)P between periods with and without the episodes were checked. Given that the averaged multi-annual concentration data tend to follow a normal distribution (Shapiro-Wilk test, $P > 0.05$), the difference between the pair of measurements ($d_i = x_{\text{with E}} - x_{\text{without E}}$) was tested by using Student's *t*-test for dependent samples ($P = 0.05$). This difference was further used to verify the hypothesis that the average for each difference in the studied population is 0 (H_0). The significance value in the Levene's test was greater than our alpha ($P > 0.05$). Thus, we cannot reject the null hypothesis (no difference) for the assumption of the variance homogeneity, which means that concentrations were drawn from populations with the same variance (Tables 3-5).

Analysis of Inhalation Risk

Lung cancer risk that resulted from the exposure to PM_{10}-bound As, Cd, Pb, Ni, and B(a)P was assessed using the deterministic approach[25] according to the US EPA[26] reference methodology. The analysis was performed for periods with and without episodes separately for agglomerations, cities, and remaining areas. The exposure scenario includes the lifetime inhalation exposure of a hypothetical adult Polish resident (without division between male or female).

Estimation of Daily Heavy Metal and B(a)P Intake

The active dose of inhaled carcinogens, namely, metals and B(a)P, was assumed to be equal to the airborne concentration and calculated following Equation 1:

$$E_i = C_i \times IR \quad (1)$$

Where E_i– daily exposure level to i^{th} carcinogen in the adult age group (ng/d), C_i– concentration of i^{th} carcinogen in the air (ng/m^3), *IR*- inhalation rate among adults; the 95th percentile of *IR* for people 31 to 41 years old is 21.4 m^3/day, following US EPA[27].

Cancer Risk

The possibility of additional lung cancer risk was estimated using Incremental Lifetime Cancer Risk (ILCR)[28]. ILCR represents the probability of an individual to develop cancer over his or her lifetime from exposure to PM_{10}-bound metals and B(a)P (for example, 1:100,000 indicates one case of cancer in a population of 100,000). The ILCR value was calculated on the basis of Equation 2, and *CSF* values for carcinogenic pollutants were used in accordance with the Office of Environmental Health Hazard Assessment database (Table 2)[29]. The risk to humans was calculated independently for each pollutant and then summarized. The cumulative lifetime cancer risk as a result of exposure to multiple carcinogens is obtained. This cumulative ICLR was compared with the threshold value. The level of acceptable cancer risk for regulatory purposes is considered within the range of 1×10^{-6} to 1×10^{-4}[30].

$$ILCR = \frac{E_i \times EF \times ED \times (CSF_i)}{BW \times AT} \times cf \quad (2)$$

Where *ILCR*– incremental lifetime cancer risk resulting from a specific dose of carcinogen, E_i– daily exposure level to i^{th} carcinogen in the adult age group (ng/d), CSF_i– slope factor for i^{th} carcinogen (kg × d/mg), *EF*– exposure frequency (day/year)[31], *ED*– exposure duration (year) (human lifespan: 70 years), *AT*– average time for carcinogens AT = 70 (year) x 365 (day/year)[32], *BW*– body weight (70 kg)[27], *cf*– conversion factor (10^{-6}) (ng/mg).

To check whether PM_{10} episodes significantly increase the overall health risk from PM_{10}-related metals and B(a)P, *t*-test for paired data was performed in accordance with the scheme presented in the section 'Selection of Episodes'. This approach involves comparing the *P*-value with the significance level ($P = 0.05$) and rejecting the null hypothesis when the *P*-value is less than the significance level.

RESULTS

PM_{10}-bound As, Cd, Pb, Ni, and B(a)P Concentrations in Poland

Tables 3-5 present the average PM_{10}-bound As, Cd, Pb, Ni, and B(a)P concentrations in Polish cities, agglomerations, and remaining areas divided into

Table 2. Inhalation Unit Risks and Cancer Potency Factors for Risk Analysis[29]

Carcinogen	Inhalation Unit Risk (μg/m^3)$^{-1}$	Inhalation Slope Factor (CSF_i) (mg/kg×d)$^{-1}$
Arsenic	3.3×10^{-3}	1.2×10^{1}
Cadmium	4.2×10^{-3}	1.5×10^{1}
Lead	1.2×10^{-5}	4.2×10^{-2}
Nickel (nickel oxide)	2.6×10^{-4}	9.1×10^{-1}
B(a)P	1.1×10^{-3}	3.9×10^{0}

periods with and without episodes. Table S2 (available at www.besjournal.com) shows the significance of the differences in concentration data between those groups. The mass concentrations of PM_{10}-bound metals indicate that in most cases, their concentrations were within acceptable standards (Tables 3-5), while the B(a)P threshold value (1 ng/m^3[22]) was exceeded in almost every part of the country. The greatest air pollution by B(a)P in periods with and without episodes occurs in the heart of Śląskie Province, specifically in the Rybnicko-Jastrzębska agglomeration (12.3 ng/m^3) (Table 3), which is considered the cradle of Polish mining. In the Łódzka agglomeration, which also lies in the European air pollution hotspot area[33], the B(a)P concentrations were slightly lower (7.9 ng/m^3) than those in the Górnośląska agglomeration, but they also far exceeded the threshold value. In areas located outside the cities and agglomerations (the remaining areas), the highest concentrations of B(a)P were recorded in Łódzka area (9.8 ng/m^3), Krakowska (7.72 ng/m^3), and Śląska (7.92 ng/m^3) (Table 4). Much lower concentrations were found in the northern part of Poland, specifically the areas within Warmińsko-Mazurskie and Podlaskie provinces) (Table 3) with average multi-year concentrations of 0.62 and 1.79 ng/m^3, respectively.

Regardless of the area type, Pb had the highest mass concentrations among metals and was followed by Ni, As, and Cd. The average Pb concentrations in cities, agglomerations, and remaining areas during 2002-2014, including episode occurrence, were 29.4, 32.3, and 23.2 ng/m^3, respectively, and were slightly lower and estimated at 27.6, 30.6, and 22 ng/m^3, respectively, after excluding the episodes. Higher mean concentrations of As, Cd, and Ni in 2002-2014 were also observed in the dataset that includes episodes. The highest concentrations of As and Cd was recorded in the cities– (8.3 ng/m^3-Ds Leg.) and (1.8 ng/m^3 Op.), respectively (Table 5). The highest Ni concentration recorded in the agglomerations was 2.61 ng/m^3 (Table 3). The highest multi-year average concentrations of As and Pb occurred in Legnica with 8.32 and 144 ng/m^3, respectively, which is a direct result of their release from the Legnica copper plant. In terms of Pb concentrations, the Górnośląska agglomeration ranked second among all agglomerations, with an average concentration of 91.4 ng/m^3. The source of metals in the Górnośląska and Rybnicko-Jastrzębska agglomerations, as in the case of Legnica, is the emissions from industrial processes and combustion of fuels in household ovens and car engines[34-36]. Industrialization and metallurgical processes have left a mark on air quality and the destruction of natural resources, especially in the southern regions of the country, which are richly endowed with minerals and coal deposits[37]. Therefore, the difference between the regional levels of metal concentrations generally decreases from the northeast to the southwest of Poland. The spatial distribution of metals (Figure 1) indicates that area type does not greatly influence the concentration levels of PM_{10}-bound As, Cd, Pb, and Ni.

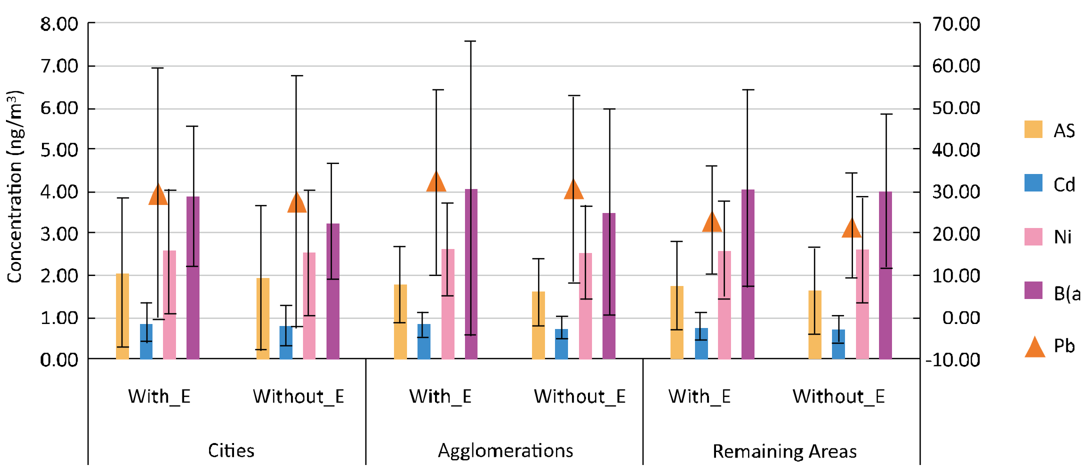

Figure 1. Average PM_{10}-bound metals and B(a)P ambient concentrations in Poland over the 2002-2014 period [including episode (with_E) and without episode occurrence (without_E)]. The bars denote the average mean concentration, while the whiskers denote standard deviation.

Table 3. Average Concentrations (ng/m^3) of As, Cd, Pb, Ni, and B(a)P in 12 Polish Agglomerations in the 2002-2014 Period

Constituents	Agglomerations														Levene's Test		*P*-Value
	Ds	Kp	Lb	Ld	Mp	Mz	Pd	Pm	Sl-G	Sl-RJ	Wp	Zp	Mean	Std.Dev	F	Sig	Paired *t*-test
As WITH_E	2.6	3.3	1.0	2.1	1.8	0.4	0.6	1.4	2.4	2.9	1.8	1.3	1.8	0.9	0.1237	0.7283	0.00042
As WITHOUT_E	2.4	2.9	0.9	1.9	1.7	0.3	0.6	1.3	2.1	2.6	1.7	1.0	1.6	0.8			
Cd WITH_E	0.6	1.1	0.5	0.9	1.4	0.7	0.7	0.4	1.1	1.0	0.7	0.7	0.8	0.3	0.1151	0.7375	0.000792
Cd WITHOUT_E	0.6	1.0	0.4	0.8	1.3	0.7	0.7	0.4	1.0	0.9	0.6	0.6	0.7	0.3			
Pb WITH_E	21.7	45.0	12.5	22.5	45.0	31.9	8.4	15.8	91.4	43.0	21.2	29.9	32.2	22.1	0.0141	0.9065	0.005969
Pb WITHOUT_E	19.9	38.0	12.1	20.0	44.0	31.0	7.7	14.8	90.4	40.0	19.7	29.3	30.7	22.3			
Ni WITH_E	3.6	2.6	1.7	2.3	3.1	5.2	1.2	2.6	2.3	2.0	1.2	3.4	2.6	1.1	0.0002	0.9887	0.003426
Ni WITHOUT_E	3.6	2.6	1.7	2.2	3.0	5.1	1.2	2.7	2.3	1.9	1.2	3.3	2.5	1.1			
BaP WITH_E	3.9	3.4	0.5	7.9	4.3	1.7	2.1	1.8	7.9	12.3	3.2	1.6	4.1	1.5	0.8578	0.3643	0.025991
BaP WITHOUT_E	3.0	3.4	0.5	6.8	2.6	1.4	1.9	1.4	6.2	8.5	3.2	1.6	3.5	2.5			

Note. The significance (*t*-test, $P < 0.05$) and variance homogeneity (Levene's test) between mean concentrations in periods with episodes versus those without episodes are shown. Ds–Wrocławska Kp–Bydgoska, Lb–Lubelska, Ld–Łódzka, Mp–Krakowska, Mz–Warszawska, Pd–Białostocka, Pm–Trójmiejska, Sl-G–Górnośląska, Sl-RJ–Rybnicko-Jastrzębska, Wp–Poznańska, Zp–Szczecińska.

In terms of area type, the PM_{10}-bound metal concentrations change slightly in the following descending order: cities > agglomerations > remaining areas (As); cities > agglomerations > remaining areas (Cd); agglomerations > cities > remaining areas (Pb); and remaining areas > agglomerations > cities (Ni). Unlike the metal concentrations, the mean B(a)P concentrations throughout the 2002-2014 period were significantly higher in remaining areas compared with those in cities or agglomerations (Tables 3-5), which is a direct result of spatial differences in emission from residential combustion sources.

Lung Cancer Risk

Figures 2-4 show a comparison of the inhalation lifetime cancer risk calculated in the scenario included the episode occurrence and assumed their absence according to administrative division. These figures present the spatial distribution of the cumulative lifetime cancer risk among considered cities, agglomerations, and remaining areas. Figure 5 presents the total cancer risk averaged within these differently polluted locations.

The cumulative lung cancer risk, as averaged between cities, agglomerations, and remaining areas in the dataset that includes episodes, was 1.13×10^{-5}, whereas it was 1.04×10^{-5} in the dataset that excludes the influence of the episodes and did not exceed the acceptable risk (1×10^{-4}). Among all the considered cities, the highest cancer risk was found in Częstochowa, Kielce, Legnica, Opole, Wałbrzych, and Zielona Góra both in the periods with and without the episodes. The averaged 'ILCR With_Episodes' value within those locations was 1.8×10^{-5}, while it was one order of magnitude lower and estimated at 1×10^{-6} among the cities lying in the north (i.e., close to the Baltic Sea), such as Elbląg, Olsztyn, and Włocławek. Among agglomerations, the highest risk value was found in Górnośląska and Rybnicko-Jastrzębska areas with 'ILCRWith_Episodes' values equal to 1.74×10^{-5} and 2.15×10^{-5}, respectively. An interesting exception can be found in the remaining areas in Zachodniopomorskie and Mazowieckie provinces (Figure 3), where the number of cancer occurrences in the periods with episodes is lower than that in the periods excluding the episodes. However, this observation is expected because these provinces are among the wealthiest and most economically developed areas of Poland, with the highest GDP per capita and whose commercial energy sector (even in the

Table 4. Average Concentrations (ng/m^3) of As, Cd, Pb, Ni, and B(a)P in the 16 Remaining (other than cities and agglomerations) Areas of Poland in the 2002-2014 Period

Constituents	Remaining Areas																		Levene's Test		P-Value
	Ds	Kp	Lb	Lu	Ld	Mp	Mz	Op	Pk	Pd	Pm	Sl	Sk	Wm	Wp	Zp	Mean	Std Dev	F	Sig	Paired *t*-test
As WITH_E	3.8	1.9	0.4	3.4	2.0	1.5	0.6	2.3	1.4	0.3	1.4	3.0	-	1.1	2.3	0.8	1.7	1.1	0.1237	0.7283	0.001421
As WITHOUT_E	3.6	1.8	0.4	3.3	1.8	1.4	0.6	2.4	1.2	0.3	1.4	2.8	-	1.0	2.1	0.7	1.6	1.0			
Cd WITH_E	1.0	1.0	0.7	0.6	0.6	1.1	0.6	0.5	1.1	-	0.4	1.4	1.3	0.2	0.8	0.5	0.8	0.3	0.1151	0.7375	0.000003
Cd WITHOUT_E	0.9	0.9	0.6	0.6	0.6	1.0	0.5	0.4	1.0	-	0.4	1.3	1.3	0.2	0.8	0.5	0.7	0.3			
Pb WITH_E	40.0	20.7	8.3	24.3	21.0	31.6	20.7	20.6	27.3	13.0	18.4	58.7	-	5.4	20.7	17.4	23.1	13	0.0141	0.9065	0.000054
Pb WITHOUT_E	39.0	20.2	7.8	21.9	19.0	28.8	19.9	19.9	25.4	12.8	17.3	57.0	-	4.9	18.6	16.7	22.0	12.6			
Ni WITH_E	3.8	1.6	2.0	2.8	2.1	3.7	2.4	2.4	1.4	0.5	4.1	5.0	3.8	0.7	3.1	3.4	2.6	1.1	0.0002	0.9887	0.079302
Ni WITHOUT_E	3.9	1.5	2.0	2.8	2.0	3.7	2.4	2.2	1.4	0.5	4.1	3.8	3.3	0.6	3.1	3.3	2.6	1.2			
BaP WITH_E	4.8	3.2	2.6	4.5	9.8	7.7	3.6	4.6	5.5	1.8	5.1	7.9	5.2	0.6	2.5	3.4	4.1	2.3	0.8578	0.3643	0.003419
BaP WITHOUT_E	4.0	3.1	2.4	3.3	7.6	4.2	2.6	2.1	3.6	1.8	3.4	6.9	4.3	2.1	2.5	2.7	4.0	1.8			

Note. The significance (*t*-test, *P* < 0.05) and variance homogeneity (Levene's test) between mean concentrations in periods with episodes versus those without episodes are shown. Ds–Dolnośląska, Kp–Kujawsko-Pomorska, Lb–Lubelska, Lu–Lubuska, Ld–Łódzka, Mp–Krakowska, Mz–Mazowiecka, Op–Opolska, Pk–Podkarpacka, Pd–Podlska, Pm–Pomorska, Sl–Śląska, Sk–Świętokrzyska, Wm–Warmińsko-Mazurska, Wp–Wielkopolska, Zp–Zachodniopomorska. The lack of data '-'.

Table 5. Average (ng/m^3) Concentrations of As, Cd, Pb, Ni, and B(a)P in 18 Polish Cities in the 2002-2014 Period

Constituents	Cities																				Levene's Test		P-Value
	Ds Leg	Ds Walbrz	Kp Toru	Kp Wlod	Lu Gorz	Lu Ziel	Mp Tar	Mz Plock	Mz Radom	Op Opole	Pk Rzesz	Sl Biel	Sl Czesto	Sk Kiel	Wm Elb	Wm Olszty	Wp Kalisz	Zp Kosz	Mean	Std Dev	F	Sig	Paired *t*-test
As WITH_E	8.3	2.2	1.3	1.0	1.4	3.7	1.2	0.7	0.4	2.5	1.5	2.2	3.0	1.9	1.7	1.4	2.2	0.6	2.1	1.8	0.0064	0.9367	0.000108
As WITHOUT_E	8.0	2.2	1.2	0.9	1.4	3.7	1.1	0.7	0.3	2.4	1.3	2.0	2.7	1.6	1.7	1.3	2.0	0.5	1.9	1.7			
Cd WITH_E	1.2	0.8	1.2	1.4	0.5	0.5	1.6	0.8	0.4	1.8	0.8	0.7	1.1	1.3	0.2	0.2	0.6	0.5	0.9	0.5	0.0568	0.8129	0.000549
Cd WITHOUT_E	1.1	0.7	1.2	1.4	0.5	0.4	1.6	0.8	0.4	1.8	0.7	0.6	1.0	1.1	0.2	0.2	0.5	0.4	0.8	0.5			
Pb WITH_E	144.0	25.8	12.4	27.5	24.6	25.7	22.9	27.1	18.7	38.5	23.6	25.4	37.0	34.7	6.9	4.5	15.6	14.4	29.4	30.1	0.0009	0.9760	0.000001
Pb WITHOUT_E	142.0	24.1	11.6	25.7	24.0	24.8	21.8	26.6	16.1	35.8	19.7	22.2	35.0	32.2	6.0	4.0	13.4	13.4	27.7	30.0			
Ni WITH_E	3.3	6.4	1.9	2.7	3.4	2.5	2.1	2.7	1.3	5.9	1.1	1.9	2.5	2.2	1.0	0.7	1.9	3.0	2.6	1.5	0.0176	0.8949	0.021427
Ni WITHOUT_E	2.7	6.4	1.8	2.7	3.3	2.4	2.1	2.7	1.3	5.6	1.1	1.9	2.5	2.1	1.0	0.7	1.8	3.0	2.5	1.5			
BaP WITH_E	6.6	5.0	2.3	2.3	3.9	1.9	3.5	4.7	4.3	7.6	2.7	5.3	3.1	5.8	2.9	1.3	3.5	3.2	3.9	1.7	0.8595	0.3603	0.000958
BaP WITHOUT_E	5.2	4.7	1.9	2.3	2.1	1.6	3.5	4.1	3.8	6.4	2.7	3.1	2.9	4.7	2.9	1.1	3.5	2.4	3.3	1.4			

Note. The significance (*t*-test, *P* < 0.05) and variance homogeneity (Levene's test) between mean concentrations in periods with episodes versus those without episodes are shown. DsLeg–Legnica, DsWalbrz–Wałbrzych, KpToru–Toruń, KpWlocl–Włocławek, LuGorz–Gorzów, LuZiel–Zielona Góra, MpTar–Tarnów, MzPlock–Płock, MzRadom–Radom, OpOpole–Opole, PkRzesz–Rzeszów, SlBiel–Bielsko-Biała, SlCzesto–Częstochowa, SkKiel–Kielce, WmElb–Elbląg, WmOlszty–Olsztyn, WpKalisz–Kalisz, ZpKosz–Koszalin.

rural areas) is based on ecological solutions. Therefore, the rural energy-related PM emissions in these areas is much lower than that in the rest of the country.

When averaging ILCR within all cities, the number of lung cancer cases in those areas was approximately 9% higher than the risk found within the remaining areas (regardless of episode occurrence) (Figure 5). This observation is in good agreement with the popular opinion that urban dwellers are at a higher health risk than citizens living in rural areas or areas located some distance from the agglomerations. The observed dependencies are, however, burdened by fluctuations in the availability of the monitoring data among different provinces and monitoring station types (Table S1, available at www.besjournal.com). In fact, Polish cities and agglomerations are more densely covered by air monitoring stations than other areas within the country.

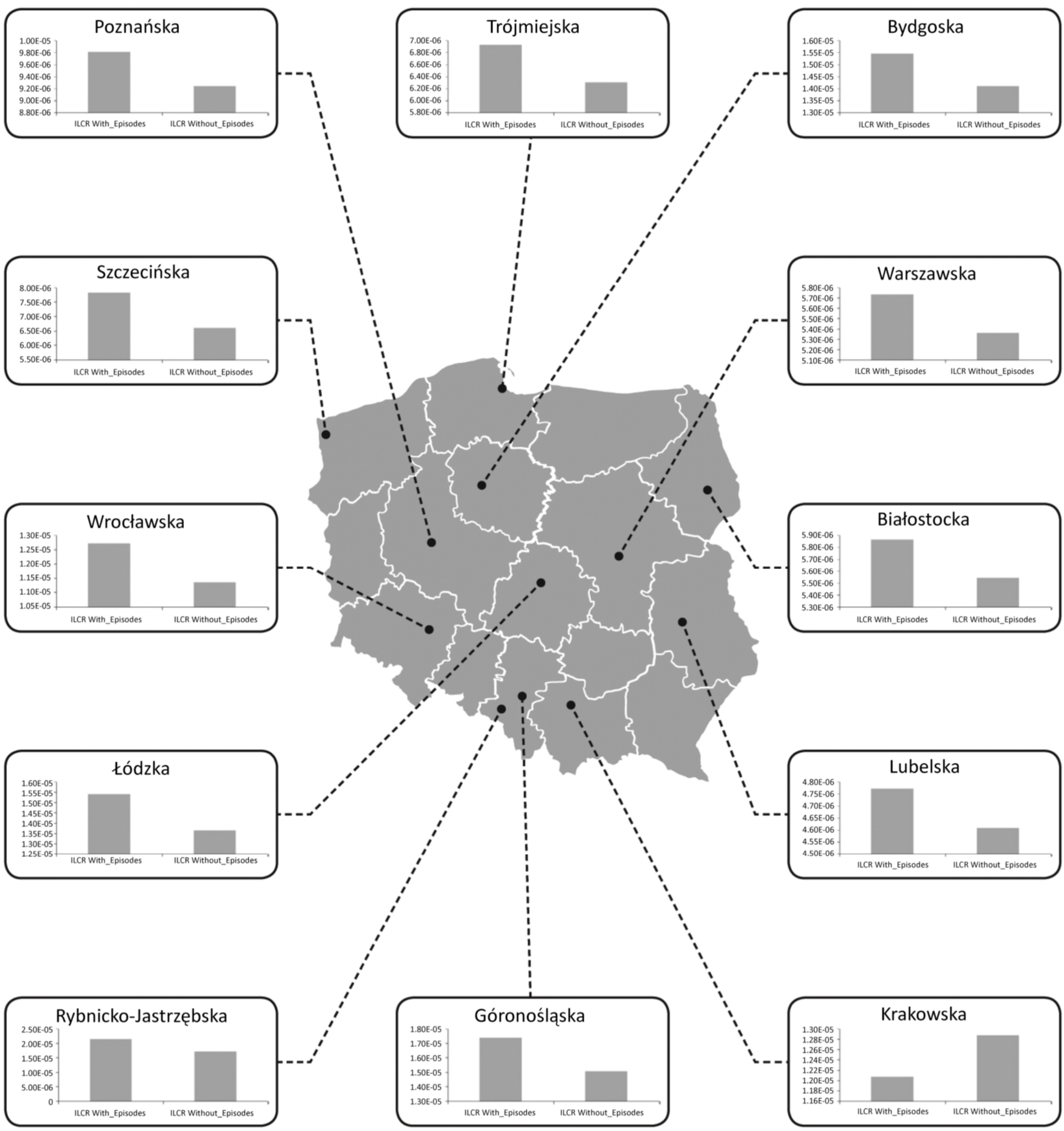

Figure 2. Cumulative lifetime lung cancer risk (ILCR) resulting from exposure to PM-bound As, Cd, Pb, and Ni in Polish agglomerations presented in two datasets, namely, including (ILCR With_Episodes) and excluding PM_{10} episodes (ILCR Without_Episodes). E-05, $\times 10^{-5}$; E-06, $\times 10^{-6}$.

DISCUSSION

Interestingly, while looking at the dataset in Tables 3-5, we can observe that over the past 12 years, metal concentrations were uniformly distributed between agglomerations, cities, and the remaining areas. This situation results from the fact that the energy-related emissions of PM and PM-bound metals in Poland have not undergone any significant changes over the last years. Taking into account the source category, the highest share of metal emission loadings into the atmosphere is attributable to the 1.A.1 Public Power and Industrial Comb sector according to the Selected Nomenclature for Sources of Air Pollution (SNAP97)[38]. The major emission of As, Cd, and Pb originates from 'combustion

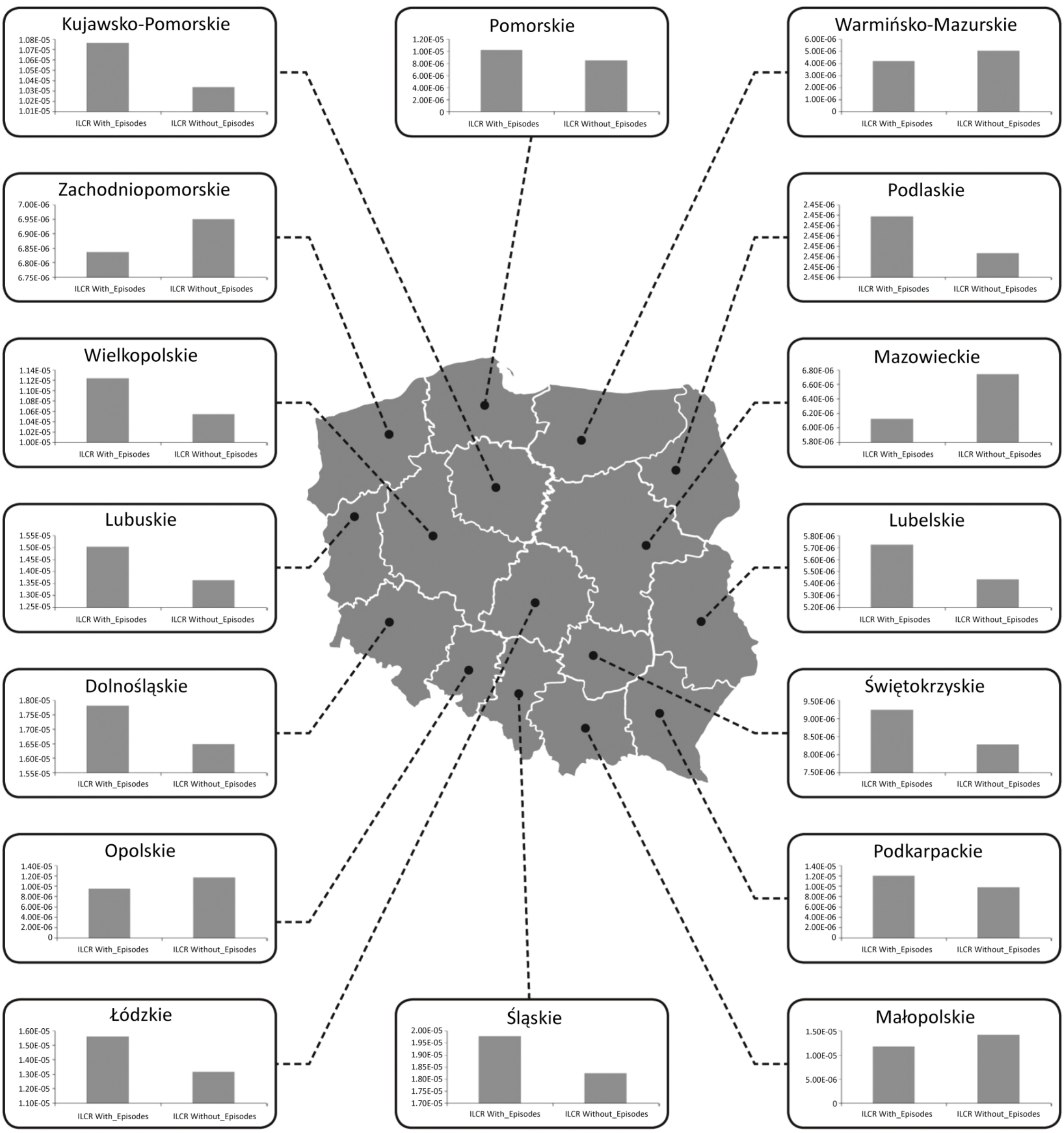

Figure 3. Cumulative lifetime lung cancer risk (ILCR) resulting from exposure to PM-bound As, Cd, Pb, and Ni within Polish provinces but lying outside the agglomerations and cities (referred to as 'remaining areas' in this work) presented in two datasets, namely, including (ILCR With_Episodes) and excluding PM_{10} episodes (ILCR Without_Episodes). E-05, × 10^{-5}; E-06, × 10^{-6}.

in industrial processes' (48%, 56%, and 49% of the total emission rate, respectively), and in the case of Ni, the emissions originate from 'combustion processes outside industry' (54% of the total national emission)[39]. Iron and non-ferrous metallurgy also have a significant share in the amount of the national emission of these pollutants[39-40].

Regardless of location, in almost each case, a statistically significant difference was found in metals and B(a)P concentrations between periods with and without episodes (*t*-test, $P < 0.05$) (Table S2, available at www.besjournal.com). The only exception was the lack of this relevance in the case of Ni concentrations within the remaining areas. The dependencies that we found are not surprising because

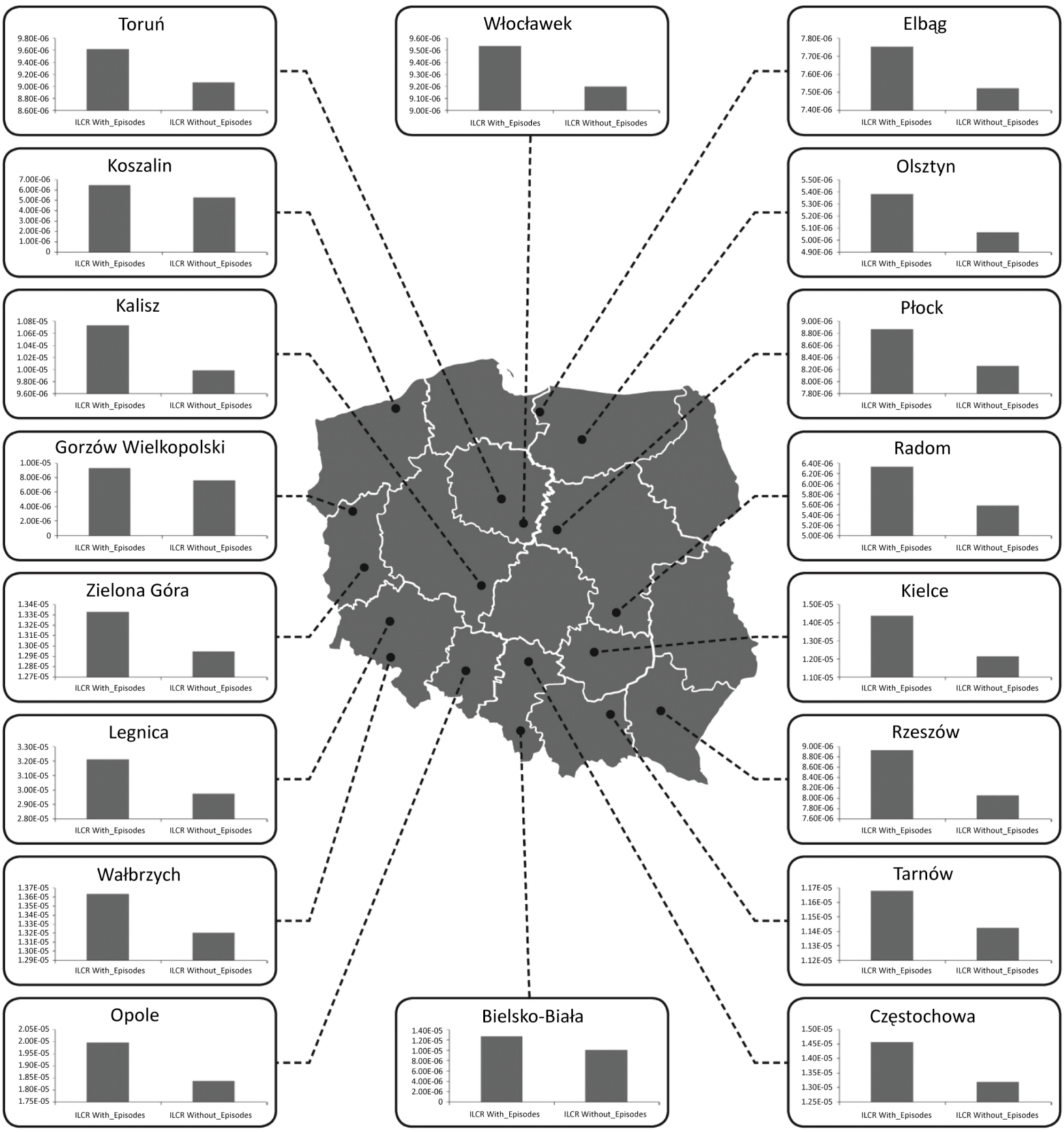

Figure 4. Cumulative lifetime lung cancer risk (ILCR) resulting from exposure to PM-bound As, Cd, Pb, and Ni in Polish cities (with more than 100,000 residents) presented in two datasets, namely, including (ILCR With_Episodes) and excluding PM_{10} episodes (ILCR Without_Episodes). E-05, × 10^{-5}; E-06, × 10^{-6}.

PM_{10} episodes greatly add to the total emission load of PM pollution. In Poland, where the dominant source of PM emission into the atmosphere is burning fossil fuels in the municipal and residential sectors (57% of the PM_{10} fraction is produced by the combustion of solid fuels, mainly lignite and coal[40]), the PM_{10} concentrations are correlated mostly with the concentrations of soot and SO_2. Meanwhile, metals and B(a)P in atmospheric air are primarily associated with fine particles with an aerodynamic diameter of less than 2.5 μm[41]. Stronger or more significant differences in the concentrations of metals and B(a)P between the episode and non-episode periods would be observable for $PM_{2.5}$-bound compounds, especially in the residential areas that tend to be heated with wood and coal, where toxic metals are mainly bound to fine and accumulation particles[42]. Another situation of concern is high-traffic areas, where resuspension of road and soil dust greatly adds to the observed loadings of PM coarse particles[43].

The influence of smog episodes on the levels of PM_{10} and $PM_{2.5}$-bound metals was investigated in the course of studies conducted by[2] at the crossroads and urban background site in Zabrze (in southern Poland). During the episodes, the concentrations of metals associated with PM_{10} increased at the roadside compared with that in the background by approximately 10 times (one order), while $PM_{2.5}$-bound metals showed two to three times elevated concentrations [except Fe (five times)] and Cr (no increase)]. Other studies conducted in Krakow (also in southern Poland) found that during the temperature inversion episodes, the concentrations of PM_{10}-bound B(a)P were 200-400 times higher than the EU target value of 1 ng/m^3[44].

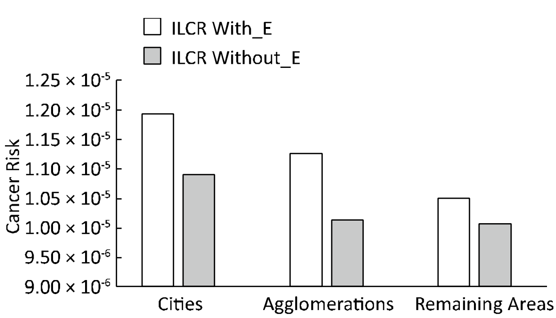

Figure 5. Cumulative lifetime lung cancer risk (ILCR) resulting from exposure to PM-bound As, Cd, Pb, and Ni in Poland presented in two datasets, namely, including (ILCR With_E) and excluding PM_{10} episodes (ILCR Without_E).

This finding proves that air pollution by B(a)P is one of the major environmental problems in Poland and is especially marked in the southern parts of Poland, that is, the extensively industrialized and highly polluted Śląskie coal basin[34-37,45]. Many researchers state that local-scale air pollution episodes are much more evident than large-scale events (e.g., the whole country), where different confounding factors such as meteorological conditions can blur their occurrence[46]. This issue is the reason the Polish Institute for Environmental Protection-National Research Institute suggested that PM episodes should be analyzed at different range levels, from regional to transboundary[18].

Our calculations indicate a significant difference in the total risk between smog and non-smog periods among cities and agglomerations (t-test, $P < 0.05$) (Table S3 available at www.besjournal.com and Figure 5). This observation is in good agreement with the epidemiological data that showed positive associations between urban PM pollution and respiratory outcomes, such as hospital admissions for lower respiratory tract infection, chronic obstructive pulmonary disease, or asthma[47-51]. With regard to the question of the relationship between the number of lung cancer occurrences and the region type, Poland is exposed to concentrations of PM-bound As, Cd, Ni, Pb, and B(a)P that are typically found in the agglomerations and big cities, which are at greater risk compared with the remaining areas (Figure 5). This result is the sum of the risks posed by exposure to all metals and B(a)P. The magnitude of carcinogenic potency factors was higher for Cd and As than that for B(a)P (Table 2), which shifts the risk toward areas that are characterized by the highest concentrations of those elements, that is, cities and agglomerations (Figures 2-3). When looking at the concentration data (Tables 3-5), in the case of B(a)P, the greatest cancer risk will occur in the remaining areas outside the urban sites, whereas in the case of metals, more noticeable effects will occur in cities and agglomerations. Different emission inventories indicate that the less densely populated and less wealthy areas of Poland are more affected by B(a)P than the big cities. Unlike cities or agglomerations that are supplied with heat and hot water by the district heating system, most Polish suburbs and villages are heated by boilers or outdated ovens that emit excessive amounts of air pollutants, including B(a)P[42].

The link between geographical differences in air pollution and mortality/morbidity rates of lung cancer must be sought with extreme caution because the observed dependencies are a cumulative derivative of variances in society, poverty, economy, and national policies, which are often imbalanced between urban and rural areas[52]. Despite good evidence that ambient air pollution increases the risk of lung cancer not only in Poland but also around the world, we must remember that simple risk calculations, such as those presented in this study, cannot be the basis for making an inference about the existence of such a relationship. They only provide an answer to the question concerning the magnitude of exposure to a few atmospheric pollutants.

CONCLUSIONS

A significant contribution of this work is that it investigates and presents the exposure and inhalation risk related to most toxic metals and B(a)P on the basis of existing monitoring data. Moreover, this work determines the extent to which Polish episodes of excessive PM_{10} concentrations add to this exposure. This approach provides an important input to the discussion regarding the additive effect of the smog effect to human exposure, and it helps quantify the spatially diverse estimations of environmentally related cancer. The latest reports of Lung Cancer Research Fund International indicate that lung cancer incidences in Poland at an age-standardized rate per 100,000 (world) averaged for males and females is 38 (WHO Cancer Today Database[53]). This result puts Poland in ninth place among countries in this category and indicates the need for further studies concerning the carcinogenic potential of air pollution, with a special emphasis on the long-term health effects of extremely high PM pollution. Our results clearly demonstrate that the occurrence of elevated PM_{10} concentrations in Poland on a large scale significantly increases the concentration level of carcinogenic compounds such as As, Cd, Ni, Pb, and B(a)P. This finding is a result of the significant environmental impact of PM concentration exceedances, as reflected by an increase in the concentrations of PM-bound metals and B(a)P, which constitute only a trace amount of the total PM_{10} mass. Our results prove that the Polish episodes of high PM_{10} concentrations, although predominantly local, can contribute significantly to country-level PM pollution. This finding also means that both the occurrence of smog episodes and a search for their relationships with health effects should be considered not only on a local scale but also on the country level. However, finding and demonstrating the correlations between local PM episodes and long-term health effects outside the country borders are considerable challenges. Therefore, further research should be directed to elucidate the prospects for attributing lung cancer mortality to the ambient air quality in Poland, with a special emphasis on addressing the health effects of geographical differences in the frequency and magnitude of PM_{10} concentration exceedances.

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REFERENCES

1. Ośródka L, Ośródka K, Święch-Skiba J. Smog zimowy w Górnośląskim Okręgu Przemysłowym jako jeden ze skutków antropogenicznych zmian klimatu lokalnego. Folia Geographica Physica, 1998; 3, 361-9.
2. Pastuszka JS, Rogula-Kozłowska W, Zajusz-Zubek E. Characterization of PM_{10} and $PM_{2.5}$ and associated heavy metals at the crossroads and urban background site in Zabrze, Upper Silesia, Poland, during the smog episodes. Environ Monit Assess, 2010; 168, 613-27.
3. Mira-Salama D, Grüning C, Jensen NR, et al. Source attribution of urban smog episodes caused by coal combustion. Atmos Res, 2008; 88, 294-304.
4. Euracoal statistics, 2016. Coal and lignite production and imports in Europe. European Association for Coal and Lignite aisbl. https://euracoal.eu/info/euracoal-eu-statistics/. [2017-04-01]
5. Ośródka L, Krajny E, Wojtylak M. Analiza epizodów smogowych w sezonie zimowym na Górnym Śląsku. In: Ochrona powietrza w teorii i praktyce (Konieczyński J. Ed), pp. 197-206. Instytut Podstaw Inżynierii Środowiska Polskiej Akademii Nauk, Zabrze, 2006.
6. Majewski G, Rogula-Kozłowska W, Czechowski PO, et al. The impact of selected parameters on visibility: first results from a long-term campaign in Warsaw, Poland. Atmosphere, 2015; 6, 1154-74.
7. Sati AP, Mohan M. Analysis of air pollution during a severe smog episode of November 2012 and the Diwali Festival over Delhi, India. Inter J of Rem Sens, 2014; 35, 6940-54.

8. Wichmann HE. What can we learn today from the Central European smog episode of 1985 (and earlier episodes)? Int J Hyg Environ Health, 2004; 207, 505-20.
9. Bell ML, Davis DL, Fletcher T. A retrospective assessment of mortality from the London smog episode of 1952: the role of influenza and pollution. Environ Health Persp, 2004; 112, 6-8.
10. Vieno M, Heal MR, Twigg MM, et al. The UK particulate matter air pollution episode of March-April 2014: more than Saharan dust. Environ Res Lett, 2016; 11, 059501.
11. Kowalska M, Hubicki L, Zejda JE, et al. Effect of ambient air pollution on daily mortality in Katowice conurbation, Poland. Pol J Environ Stud, 2007; 16, 227-32.
12. Samek L. Overall human mortality and morbidity due to exposure to air pollution. IJOMEH, 2016; 29, 417-26.
13. Valentin Foltescu/EEA. Air quality in Europe Report – 2014 (EEA Report. No 5/2014). European Environment Agency. 2014.
14. Badyda AJ, Grellier J, Dąbrowiecki P. Ambient $PM_{2.5}$ exposure and mortality due to lung cancer and cardiopulmonary diseases in Polish cities. Adv Exp Med Biol, 2017; 944, 9-17.
15. Krzyżanowski M. Wpływ zanieczyszczenia powietrza pyłami na układ krążenia i oddychania (Circulatory and respiratory effects of particulate air pollution). Lekarz Wojskowy (Military Physician), 2016; 1, 17-22.
16. Godłowska J, Tomaszewska AM. Relations between circulation and winter air pollution in Polish urban areas. Arch Environ Prot, 2010; 36, 55-66.
17. Juda-Rezler K, Reizer M, Oudinet JP. Determination and analysis of PM_{10} source apportionment during episodes of air pollution in Central Eastern European urban areas: The case of wintertime 2006. Atmos Environ, 2011; 45, 6557-66.
18. IOŚ-PIB, 2016. Analiza wybranych epizodów wysokich stężeń pyłu PM_{10} z lat 2013-2016. Etap I, Epizody z lat 2013-2014, Warszawa.
19. Katzman TL, Rutter AP, Schauer JJ, et al. $PM_{2.5}$ and PM_{10}-$PM_{2.5}$ compositions during wintertime episodes of elevated PM concentrations across the Midwestern USA. Aero Air Qual Res, 2010; 10, 140-53.
20. Aarnio P, Martikainen J, Hussein T, et al. Analysis and evaluation of selected PM_{10} pollution episodes in the Helsinki Metropolitan Area in 2002. Atmos Environ, 2008; 42, 3992-4005.
21. GIOŚ (Chief Inspectorate of Environmental Protection). The portal on air quality. http://powietrze.gios.gov.pl. [2017-09-4]. (In Polish)
22. EEA Agency. Directive 2004/107/EC of the European Parliament and of the Council of 15 December 2004 relating to arsenic, cadmium, mercury, nickel and polycyclic aromatic hydrocarbons in ambient air. http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32004L0107. [2017-10-11]
23. World Health Organization. Regional Office for Europe. Second Edition. WHO Regional Publications, European Series, No. 91. 2000.
24. World Health Organization. Regional Office for Europe. Air Quality Guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide. Global update (2005) – revised WHO recommendations from 2000 regarding PM_{10}, $PM_{2,5}$, NO_2, SO_2, Ozone. European Series, No. 91, 2005.
25. EnHealth Council. Environmental health risk assessment: guidelines for assessing human health risks from environmental hazards. Canberra, ACT: Dept. of Health and Ageing and EnHealth, 2002.
26. US EPA. (U.S. Environmental Protection Agency). Risk Assessment Guidance for Superfund: Volume I Human Health Evaluation Manual (Part F, Supplemental Guidance for Inhalation Risk Assessment) ('Part F'). Office of Superfund Remediation and Technology Innovation. Washington, DC, 2009.
27. US EPA. (U.S. Environmental Protection Agency). Exposure Factors Handbook: 2011 Edition. Report EPA/600/R-090/052F, Washington, DC, 2011.
28. Liu X, Song Q, Tang Y, et al. Human health risk assessment of heavy metals in soil-vegetable system: a multi-medium analysis. Sci Tot Environ, 2013; 463-464, 530-40.
29. OEHHA. Chemicals database. https://oehha.ca.gov/chemicals. [2017-09-4]
30. Widziewicz K, Loska K. Metal induced inhalation exposure in urban population: A probabilistic approach. Atmos Environ, 2016; 128, 198-207.
31. Chen SC, Liao CM. Health risk assessment on human exposed to environmental polycyclic aromatic hydrocarbons pollution. Sci Tot Environ, 2006; 366, 112-23.
32. RAGS. Risk Assessment Guidance for Superfund Volume I Human Health Evaluation Manual (Part A), EPA/540/1–89/002, Washington, D.C. Office of Emergency and Remedial Response US Environmental Protection Agency, 1989.
33. Kiesewetter G, Schoepp W, Heyes C, et al. Modelling $PM_{2.5}$ impact indicators in Europe: Health effects and legal compliance. Environ Modell Soft, 2015; 74, 201-11.
34. Rogula-Kozłowska W, Kozielska B, Klejnowski K. Concentration, origin and health hazard from fine particle-bound PAH at three characteristic sites in southern Poland. Bull Environ Contamin Toxicol, 2013; 91, 349-55.
35. Rogula-Kozłowska W, Klejnowski K, Rogula-Kopiec P, et al. Spatial and seasonal variability of the mass concentration and chemical composition of $PM_{2.5}$ in Poland. Air Qual Atmos Health, 2014; 7, 41-58.
36. Rogula-Kozłowska W, Majewski G, Czechowski PO. The size distribution and origin of elements bound to ambient particles: a case study of a Polish urban area. Environ Monit Assess, 2015; 187, 240.
37. Rogula-Kozłowska W, Majewski G, Błaszczak B, et al. Origin-oriented elemental profile of fine ambient particulate matter in Central European suburban conditions. Int J Environ Res Public Health, 2016; 13, 715.
38. EEA. EMEP/CORINAIR Emission Inventory Guidebook. 3rd ed. European Environment Agency. Update, 2002. http://www.inemar.eu/xwiki/bin/view/InemarDatiWeb/Classification+of+activities+%28SNAP+97%29. [2017-09-4]
39. IOŚ-PIB, 2016. Assessment of air pollution at the regional background monitoring stations in Poland in 2015 in terms of PM_{10} and $PM_{2.5}$ dust composition and the deposition of heavy

metals and PAHs. Institute of Environmental Protection - National Research Institute (Ocena zanieczyszczenia powietrza na stacjach monitoringu tła regionalnego w Polsce w roku 2015 w zakresie składu pyłu PM_{10} i $PM_{2.5}$ oraz depozycji metali ciężkich i WWA), Instytut Ochrony Środowiska-Państwowy Instytut Badawczy, IOŚ-PIB) Warszawa. http://powietrze.gios.gov.pl/pjp/maps/measuringstation/U. [2017-09-4]

40.GUS, 2016. Central Statistical Office. Environment. Statistical Information and Elaboration. http://stat.gov.pl/files/gfx/portalinformacyjny/pl/defaultaktualnosci/5484/1/17/1/ochrona_srodowiska_2016.pdf. [2017-09-4]

41.Rogula-Kozłowska W. Size-segregated urban particulate matter: mass closure, chemical composition, and primary and secondary matter content. Air Qual Atmos Health, 2016; 9, 533-50.

42.Widziewicz K, Rogula-Kozłowska W, Majewski G. Lung cancer risk associated with exposure to benzo(a)pyrene in Polish agglomerations, cities and other areas. Int J Environ Res, 2018 (In press).

43.Rogula-Kozłowska W. Traffic-generated changes in the chemical characteristics of size-segregated urban aerosols. Bull Environ Contamin Toxicol, 2014; 93, 493-502.

44.Rey M, Astorga C, Cancelinha J, et al. High concentrations of PM_{10} and PAHs during winter smog episodes in Krakow, Poland. European Communities, The 34th International Symposium of Environmental Analytical Chemistry Conference paper. https://www.researchgate.net/publication/236962220_High_Concentrations_of_PM10_and_PAHs_During_Winter_Smog_Episodes_in_Krakow_Poland. [2017-09-4]

45.Kozielska B, Rogula-Kozłowska W. Polycyclic aromatic hydrocarbons in particulate matter in the cities of Upper Silesia. Arch Waste Manage Environ Protect, 2014; 16, 75-84.

46.Rogula-Kozlowska W, Majewski G, Czechowski PO, et al. Analysis of the data set from a two-year observation of the ambient water-soluble ions bound to four particulate matter fractions in an urban background site in southern Poland. Environ Prot Eng, 2017; 43, 137-49.

47.Zhou M, He G, Fan M, et al. Smog episodes, fine particulate pollution and mortality in China. Environ Res, 2015; 136, 396-404.

48.Belleudi V, Faustini A, Stafoggia M, et al. Impact of fine and ultrafine particles on emergency hospital admissions for cardiac and respiratory disease. Epidemiol, 2010; 21, 414-23.

49.Cheng MH, Chen CC, Chiu HF, et al. Fine particulate air pollution and hospital admissions for asthma: a case-crossover study in Taipei. J Toxicol Environ Health, 2014; 77, 1075-83.

50.Tsai SS, Chang CC, Yang CY. Fine particulate air pollution and hospital admissions for chronic obstructive pulmonary disease: a case-crossover study in Taipei. Int J Environ Res Pub Health, 2013; 10, 6015-26.

51.Tsai SS, Chiu HF, Liou SH, et al. Short-term effects of fine particulate air pollution on hospital admissions for respiratory diseases: a case-crossover study in a tropical city. J Toxicol Environ Health, 2014; 77, 1091-101.

52.Wang L, Yu C, Liu Y, et al. Lung Cancer Mortality Trends in China from 1988 to 2013: New Challenges and Opportunities for the Government. Int J Environ Res Public Health, 2016; 13, 1052.

53.WHO cancer today database. http://gco.iarc.fr/today/home. [2017-09-4]

SUPPLEMENTAL FILES

In Poland air pollution assessment is provided in 46 zones. The following zones include:

12 agglomerations: Wrocławska (DsWrocWie; DsWrocWybCon; DsWrocOrzech); Bydgoska (KpBydWarszaw; KpBydPlPozna; KpBydgUjejskiego; KpBydgWPola); Lubelska (LbLublin_Krasn; LbLubObywate; LbLubSliwins); Łódzka (LdLodzCzerni; LdLodzLegion; LdLodzRudzka; LdLodzWIOSARubinst; LdPabiKilins), Krakowska (MpKrakBulwar; MpKrakowWIOSPrad6115; MpKrakBujaka); Warszawska (MzWarAKrzywWSSE; MzWarAlNiepo; MzWarszZelazWSSE; MzWarZeganWSSE; MzWarszBorKomWSSE; MzWarAKrzywo; MzWarTolstoj); Białostocka (PdBialWaszyn); Trójmiejska (Pm.01w.01m; Pm.00.s237m; PmGdaGleboka; PmGdaLecz08m; PmGdyJozBema); Górnośląska (SlDabro1000L; SlKatoKossut; SlZabSkloCur); Rybnicko-Jastrzębska (SlRybniBorki; SlZorySikors); Poznańska (WpPoznanPM10szpital; WpPoznChwial); Szczecińska (ZpSzczecinWSSE; ZpSzczPils02; ZpSzczAndr01)

18 cities with the number of residents above 100 000: Legnica (DsLegAlRzecz); Wałbrzych (DsWalbrzWyso); Toruń (KpToruDziewu; KpTorunSzpMiejski); Włocławek (KpWloclOkrze; KpWloclLady); Gorzów Wielkopolski (LuGorzPilsud; LuGorzKosGdy); Tarnów (MpTarnowWIOSSoli6303; MpTarBitStud); Płock (MzPlockKolegWSSE; MzPlocKroJad); Radom (MzRadomCzWSSE; MzRad25Czerw); Opole (OpOpole246; OpOpoleOsAKr); Rzeszów (PkRzeszWIOSSzop; PkRzeszRejta); Kalisz (WpKaliszPM10; WpKaliSawick); Zielona Góra (LuZielKrotka); Bielsko-Biała (SlBielKossak); Częstochowa (SlCzestoBacz); Kielce (SkKielKusoci; SkKielJagiel); Elbląg (WmElbBazynsk); Olsztyn (WmOlsztyWSSE_Zolnier; WmOlsPuszkin); Koszalin (ZpKoszalinWSSE; ZpKoszSpasow)

16 remaining areas located within provinces but lying outside the agglomerations and cities: Dolnośląskie (DsCzerStraza; DsDzialoszyn; DsJelw05; DsPolKasztan; DsZgorBohGet; DsSzczKopPM; DsOlawZolnAK; DsOsieczow21; DsNowRudSreb; DsSzczaKolej; DsJelGorSoko; DsOlawZolnAK; DsGlogWiStwo); Kujawsko-pomorskie (KpCiechTezni; KpGrudzIkara; KpInowrSolan; KpNaklSkargi; KpWabrzstmob; KpDAFGolub; KpSepolno; KpDAFChelmza; KpDAFRadzyn; KpZielBoryTu; KpGrudSienki; KpKoniczynka; KpTuchPiast); Lubelskie (LbBiaPodOrze; LbChelJagiel; LbKrasKoszar; LbLeczna1000Lecia; LbRadzPodSit; LbRejowiecFabrWIOS; LbZamoHrubie; LbTomaszowLubWIOS); Lubuskie (LuWsKaziWiel; LuZaryWIOS_MAN; LuSulecDudka; LuZarySzyman); Łódzkie (LdKutnoWIOSMWilcza; LdOpocPlKosc; LdPioTrSienk; LdBrzeReform; LdRadomsRoln; LdRawaNiepod; LdSierGrunwa; LdSkiernWIOSMJagiell; LdToMaSwAnto; LdZduWoKrole; LdOpocPlKosc; LdWieluPOW12; LdLowiczSien; LdPioTrKraPr; LdSkierKonop); Małopolskie (MpTrzebiWIOSPils0303; MpSkawOsOgro; MpBochniWSSEKons0105; MpChrzanWSSEGrzy0301; MpNiepo3Maja; MpNoTargWSSESzaf1102; MpNSaczWSSETarn6202; MpProszWIOSKrol1404; MpWadowiWIOSPSka1805; MpBochniWSSEKons0105; MpGorlKrasin; MpTuchChopin; MpNoSaczNadb; MpNSaczWIOSPija6204; MpZakopaSien; MpBochKonfed; MpSuchaBWIOSHand1512; MpTrzebOsZWM; MpBrzeskWIOSWiej0202; MpDabrowWIOSZare0401; MpMiechoWIOSKono0802; MpNowyTaWIOSPows1114; MpOswiecWIOSSnia1302; MpRabkaWIOSChop1113; MpSuchaBWIOSHand1512; MpBukowKolejMOB; MpKetyWyspiaMOB; MpLimanoBoleMOB; MpMysleRynekMOB; MpSlomWolnosMOB; MpSzczawJanaMOB); Mazowieckie (MzOstMazSikorWSSE; MzCiechStrazacka; MzLegZegrzyn; MzNowDwChemWSSE; MzOstrolTargowa; MzOtwockBrzozWSSE; MzPiaseczDworWSSE; MzPruszKraszeWSSE; MzSochPlocWSSE; MzTluszczJKiel MzGranicaKPN; MzMlawOrdona; MzPiasPulask; MzOtwoBrzozo; MzSiedKonars; MzOstroHalle); Opolskie (OpGlubKochan; OpNamys2pyl; OpOlesno3pyl; OpKluczMicki; OpKKozBSmial; OpNysaRodzie; OpZdziePiast); Podkarpackie (PkPrzemWIOSPDom; PkJasloWIOSFlor2; PkMielZaStre; PkNiskoSzkla; PkJarosWIOSJanPawII; PkPrzemyslWIOSMick; PkJasloSikor; PkJarosPruch; PkPrzemGrunw; PkSanoSadowa; PkTarnDabrow; PkDebiGrottg); Podlaskie (PdSuwPulaski); Pomorskie (Pm.06.s712m; Pm.63.s079m; Pm.63.wDSMm; Pm14TCZEw06m; PmWejhPlWejh; PmWladywHallera; Pm.aw07m; PmKosTargo12; PmSlupKniazi; PmSlupOrzesz; PmKwiSportow; PmLebMalcz16; PmLinieKos17; PmMalMicki15; PmGac); Śląskie (SlZywieKoper; SlLublPiasko; SlZawSkloCur; SlRacibRaci_studz; SlCieszCies_dojaz; SlGodGliniki; SlKnurJedNar; SlMyszMiedzi; SlWodziWodz_bogum; SlPszczBoged; SlTarnoLitew; SlGodGliniki); Świętokrzyskie (SkBuskRokosz; SkStaraZlota); Warmińsko-mazurskie (WmDzialdWSSE_Biedraw; WmPuszczaBor; WmGizyckWIOS_Wodoc; WmNiTraugutt; WmIlawAnders); Wielkopolskie (WpKoniWyszyn; WpPilaKusoci; WpLeszno411000; WpGnieznoPM10; WpOstWieWyso; WpGniePaczko; WpLeszKiepur; WpWagrowLipo); Zachodniopomorskie (ZpSwinoujscieWSSE; ZpSzcSzczecinekPSSE; ZpWiduBulRyb; ZpSzczec1Maj; ZpSzcSzczecinek009; ZpMyslZaBram; ZpSzczecPrze).

Table S1. Summary Presenting the Number of Polish Air Monitoring Stations Conducting Measurements of As, Cd, Pb, Ni and B(a)P Concentrations by Type of the Station.

Year	Vovoideship	Urban background					Rural					Sub-urban					Traffic					Industrial				
		As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P
2002							1	1		1	1														1	1
2003		1	3	3	3	1	1	1	1	1	2											1	1	2	1	1
2004		2	6	7	6	2	2	2	2	2	2											1	1	3	1	1
2005		3	10	14	11	3	2	2	2	2	2						1		1	1		1	1	4	1	1
2006		3	12	16	12	3	2	2	2	2	2						1	1		1		1	1	3	1	1
2007		5	14	18	14	5	1	1	1	1	1						1	1	1	1		1	1	3	1	1
2008	DS	5	12	16	12	4	2	2	2	2	2						1	1	1	1		1	1	5	1	1
2009		6	14	14	10	5		1	1	1	1											1	1	5		1
2010		6	6	5	3	6	2	3	2	2	2							1				1	1	5	1	1
2011		6	9	6	6	1	2	1	2	2	1	1	1	1								1	1	4	1	1
2012		8	8	8	8	8	2	2	2	2	2	1	1	1								1	1			1
2013		9	9	9	9	6	2	2	2	2	1		1	1								1				
2014		10	10	10	10	11	1	1	1	1	1															1
2002										1																
2003				1																						
2004				1										2												
2005			1	1	1														1							
2006		2	1	2	1	1								2												
2007		1		1	1	1					1		1	1		1	1	1	1							
2008	KP	9	8	10	9									1		1		1	1	1						
2009		1	1	3	2						1	1	1	1	1			2	2	2						
2010		7	8	9	7			1	1	1	1	1	1	1	1		1	1	1	1						
2011		3	3	3	2		2	2	2	2	1	2	2	2	2	1	1	2	1	2						
2012		4	4	4	3		2	2	2	2	1	2	2	2	2	1	2	2	2	2						
2013		3	3	3	4		2	2	2	2	1	3	3	3	2	1	2	2	2	2						
2014		3	3	3	3	3	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2					
2002									1																	
2003									1	1																
2004			1	2			1	1	1	1																
2005			1	1	1		1	1	1	1																
2006			3										1													
2007																										
2008	LB																									
2009		3		5	4	1				2		2		2												
2010		2	2	2	2	1																				
2011		1	1	1	1																					
2012		1	1	1	1	1																				
2013		2	2	2	2	1																				
2014		2	2	2	2	3					2															
2002																										
2003																										
2004	LU																									
2005																										
2006																										
2007																										

Continued

Year	Vovoideship	Urban background					Rural					Sub-urban					Traffic					Industrial				
		As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P
2008				3																						
2009						1																				
2010			1	3	2	1																				
2011			3	3	3	1																				
2012			4	4	4	1																				
2013			5	5	5	1																				
2014			6	6	6	6																				
2002																										
2003																										
2004				3																						
2005				3	3																					
2006				3	3																					
2007				3	3																					
2008	LD			3	3																					
2009				4	5	1																				
2010				6	6	1																				
2011				5	6																					
2012				13	13	1																				
2013				11	11	1																				
2014				14	14	14																				
2002																										
2003																										
2004				1		1													1					1		
2005																										
2006																								1		
2007		2		2																		1				
2008	MP	9	9	9	9																	1	1		1	
2009		11	10	10	11	1																				
2010		9	10	10	10	1																1	1		1	
2011		5	6	5	5	1																1	1		1	
2012		5	5	5	5	1																1	1		1	
2013		4	4	4	4	1																1	1		1	
2014		5	5	5	5	18																1	1		1	1
2002																									1	
2003			1	3	1																		1	1		
2004		3	3	3	3									1								1	1		1	
2005		2	2	7	2									1								1	1		1	
2006		2	2	4	2	5								1								1	1		1	
2007		3	2	2	2	13								1		1					1	1	1	1	1	
2008	MZ	9	8	11	8	1								1								1	1	1		
2009		3	2	7	2	1								1								1	1	1		
2010		2	2	2	1									1								1	1	1		1
2011		3	2	4	3	1																				
2012		4	4	4	3	1																				
2013		4	4	4	4	1																	1			
2014		4	4	4	4	8										2						1	1			1
2002																										
2003				3																						
2004				4																						
2005	OP		1	1																						
2006			1	1																						
2007																										
2008			3	3	3	1																				

Continued

Year	Vovoideship	Urban background					Rural					Sub-urban					Traffic					Industrial				
		As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P
2009		1	3	2	1	1																				
2010		3	3	3	2	1																				
2011				2		1																				
2012		2	2	2	2	1																				
2013		2	2	2	2	1																				
2014		3	3		3	3																				
2002																										
2003				1																						
2004				1																						
2005			1	1																						
2006			1	1																						
2007		1	1	1	1																					
2008	PD		2	1	1																					
2009			2	2	2																					
2010			1	1	1																					
2011					1																					
2012																										
2013																										
2014		2	2	2	2	2																				
2002		1																								
2003																										
2004																										
2005																										
2006																										
2007			1	1	1	1																				
2008	PK			1		2																				
2009		7	7	7	6	2																				
2010		8	8	8	6	2																				
2011		4	4	4	4	1																				
2012		4	4	4	4	1																				
2013		8	4	4	3	1																				
2014		4	4	4	4	9																				
2002		1																								
2003																										
2004																										
2005																										
2006																										
2007				7		1													1							
2008	PM		8	8	8													1	1	1						
2009		7	9	8	8	2													1							
2010		8	12	10	10	6													1	1						
2011		4	11	9	9	5													1	1	1					
2012		4	12	12	12	6			1	1	1							1			1					
2013		8	7	7	7	7		1	1	1	1										1					
2014		4	10	11	11	10		2	2	2	2							1			1					
2002																										
2003				15																						
2004				13																						
2005	SL																									
2006																										
2007																										
2008																										

Continued

Year	Vovoideship	Urban background					Rural					Sub-urban					Traffic					Industrial				
		As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P
2009		15	15	19	15	1																				
2010		11	11	11	11	1																				
2011		11	10	10	10	1																				
2012		11	10	10	10	1																				
2013		9	8	8	8																					
2014		10	9	9	9	14																				
2002																										
2003																										
2004																										
2005																										
2006			1																							
2007					1																					
2008	SK																									
2009			2	1	2																					
2010		1	1	1	1	1																				
2011		1	1	1	1	1																				
2012		1	1	1	1	1																				
2013		1	1	1	1																					
2014		1	1	1	1	3										1										
2002		2																								
2003																										
2004																										
2005																										
2006																										
2007		2	2	2	2																					
2008	WM		2	2	2	1																				
2009		4	3	3	3	1		1	1	1																
2010		3	4	3	3	1	1		1	1	1															
2011		3	3	3	3	1	1	1	1	1	1															
2012		3	2	2	2	1		1	2	1	1															
2013		4	3	3	3			1	1	1																
2014		3	3	3	3	3	1	1	1	1	1															
2002		3																								
2003																										
2004																										
2005			3	3	3																					
2006		2	4	4		2																				
2007		1	5	6	6	1																				
2008	WP		8	8	8	1																				
2009		3	7	3	7	1																				
2010		5	5	4	4	1																				
2011		3	3	4	3	1																				
2012		4	4	6	4								1		1											
2013		5	5	5	5									1												
2014		5	4	5	5	6								1												
2002				1																						
2003				1																						
2004				1																						
2005				1																						
2006	ZP		1	1																						
2007		1	3	2	2	3	1			1	1							1		1	1					
2008			3	3	3		1											1			1					
2009			2	2	2		1			1								1								

Continued

Year	Vovoideship	Urban background					Rural					Sub-urban					Traffic					Industrial				
		As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P	As	Cd	Pb	Ni	B(a)P
2010		1	3	2	3																					
2011		2	2	3	2	1	1	1		1																
2012	ZP	3	3	3	3	1	1	1	1	1																
2013		3	3	3	3		1	1	1	1																
2014		3	3	3	3	5		1	1	1	1										1					

Note. DS– Dolnośląskie, KP– Kujawsko-pomorskie, LB– Lubelskie, LU– Lubuskie, LD– Łódzkie, MP– Małopolskie, MZ– Mazowieckie, OP– Opolskie, PK– Podkarpackie, PD– Podlskie, PM– Pomorskie, SL– Śląskie, SK– Świętokrzyskie, WM– Warmińsko-mazurskie, WP– Wielkopolskie, ZP– Zachodniopomorskie.

Table S2. Results from *t*–test for Dependent Samples – Concentrations During Episodes vs. Concentrations in the Periods without PM10 Episodes for Agglomerations (a), Cities (c) and Remaining Areas (ra). Marked Differences Are Significant at $P < 0.05000$

Variable	area	*N*	*t*	df	*P*
As	a	12	5.000514	11	**0.00042**
Cd	a	12	4.578443	11	**0.000792**
Pb	a	12	3.396164	11	**0.005969**
Ni	a	12	3.712514	11	**0.003426**
B(a)P	a	12	2.571277	11	**0.025991**
As	ra	16	3.900086	15	**0.001421**
Cd	ra	16	7.197698	15	**0.000003**
Pb	ra	16	5.567338	15	**0.000054**
Ni	ra	16	1.882574	15	0.079302
B(a)P	ra	16	3.471305	15	**0.003419**
As	c	18	5.006867	17	**0.000108**
Cd	c	18	4.242209	17	**0.000549**
Pb	c	18	7.282129	17	**0.000001**
Ni	c	18	2.533450	17	**0.021427**
B(a)P	c	18	3.985114	17	**0.000958**

Table S3. Results from *t*-test for Dependent Samples - ILCR During Episodes *vs.* ILCR in the Periods without PM10 Episodes for Agglomerations (a), Cities (c) and Remaining Areas (ra). Marked Differences Are Significant at $P < 0.05000$

Variable	area	*N*	*t*	df	*P*
ILCR	a	12	3.034650	11	**0.011355**
ILCR	ra	16	1.233310	15	0.236438
ILCR	c	18	5.626131	17	**0.000030**