Letter to Editor

Assessment of Exposure to Traffic-Related Aerosol and to Particle-Associated PAHs in Gliwice, Poland

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Abstract

The results of the pilot study of exposure to airborne particles and polycyclic aromatic hydrocarbons (PAHs) close to a busy street in Gliwice in the spring are presented. Traffic density in the investigated street between 9 a.m. and 6 p.m. was 1400 vehicles per hour. It was found that average daily concentration of PM10 (airborne particles with aerodynamic diameter $\leq 10~\mu m$) increases by 40 $\mu g/m^3$ in the street canyon in relation to locations 100 m from the road, which for inhabitants who live in this street means an increased of risk of respiratory diseases by ten percent. The average concentration of total PAHs near the street was 191.56 ng/m^3 (in the spring and without rain) and was over 1.5 times greater than at the point 100 m from the street, which confirms that exhaust gases emission on busy streets elevates the exposure to total PAHs. However, it does not concern benzo(a)pyrene (BaP), whose main emission sources seem to be industrial and municipal emitters. Exposure to BaP concerns not only the persons who live close to the busy streets, but the greater population of the Gliwice inhabitants. The risk of cancer diseases in the studied area associated with inhalation of aerosol particles containing BaP is 10^{-4} , but persons living in the investigated street have a higher cancer risk of 10^{-3} order.

Keywords: aerosol, traffic, polycyclic aromatic hydrocarbons, benzo(a)pyrene

Introduction

Epidemiological data suggest that an increase of the concentration of airborne particles with aerodynamic diameter $\leq 10~\mu m$ (PM10) by $10~\mu g/m^3$ causes a few-percentage increase of incidences of upper respiratory tract diseases, including asthma. Studies carried out in the last decade in the USA and Western Europe, have shown that exposure to particulate air pollution (despite the significant reduction in the concentration of airborne particles) is still associated with an increase of adverse health effects [1, 2] and mortality [3-5]. Numerous studies indicated that exposure to particulate matter reduces pulmonary function [6] and increases respiratory symp-

toms [7-9], hospital admissions [10, 11] and mortality [12], even at concentrations that currently occur in urban areas [13]. It is especially important that both epidemiological and toxicological studies show the disadvantageous impact of inhalation of airborne particles on the health of men who are exposed to aerosol concentration levels usually considered safe. The results were also confirmed by lack of correlation between a significant improvement of environmental quality and incidents of diseases associated with exposure to airborne particles within the last thirty years in USA and Western Europe. An astonishing lack of decrease in respiratory incidents (including allergies) with a significant decrease of concentration of atmospheric aerosol is also observed in Upper Silesia. This unexpected result can be partially explained by the fact that at the initial period of envi-

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ronmental quality improvement we can usually observe a reduction in coarse dust particles. Such action causes significant reduction of total suspended particles (TSP), but a concentration of fine particles is reduced to a small extent. Not only aerosol concentration, but also aerodynamic diameter of particles is an important parameter as regards the health hazard. Particles of diameter greater than 10 µm are not respirable, so from the health point of view, concentrations of particles less than 10 µm is aerosol mode, which should be controlled. In many countries the concentration of this fraction has already been standardized. In some countries it is planned to introduce a standard for the concentration of airborne particles having an aerodynamic diameter less than 2.5µm (so called PM2.5). It seems that despite observed improvement of air quality, due to a reduction of dust emission from industrial sources, in Europe and in Upper Silesia as well, the real exposure of many people to PM10 particles increased in the last decade together with an avalanche increase of motor transport. Thus it is important to control air quality not only through the existing monitoring system, but also in a vicinity of busy streets. Results of few studies published recently suggest a correlation between dwelling places close to busy streets and upper respiratory tract diseases [14-17]. It seems to be not only a result of increased concentration of PM10, especially fine particles, but also higher toxicity of these airborne particles in a vicinity of busy streets. An aerosol emitted from cars contains several toxic and carcinogenic substances, including polycyclic aromatic hydrocarbons (PAHs), as well as allergens.

Polycyclic aromatic hydrocarbons are generated during the burning of organic substances containing carbon and hydrogen [18]. Mutagenicity and carcinogenicity was found experimentally for many PAHs [19]. Besides, PAHs are the precursors of derivative substances of higher mutagenicity than parent substances [20, 21]. In the atmosphere of urban areas, PAHs of anthropogenic origin dominate; first of all they come from fossil fuels burning [22]. Apart of stationary emission sources, cars play an important role in environment contamination. Indicators of PAH emission from cars depend on many factors like engine type, driving conditions and fuel composition [23, 24].

The main purpose of our pilot study was to assess the elevation of health risk of inhabitants who live near the busy street in Gliwice due to the exposure to PM10 and PAHs. The second work objective was to estimate the role of the busy street as a source of airborne particles and particle-associated PAHs in this town, where during the last hundred years almost all air pollution was caused by industrial emitters. Gliwice is placed in the so-called Upper Silesian Industrial Region – a heavily industrialized part of Poland that remains the most polluted part of Europe. Burning of hard coal in industrial and municipal objects is the main source of air pollutants. In recent years the increasing amount of exhaust gases from cars has appeared.

Methods

The studies were carried out in Gliwice, in the vicinity of the Pszczyńska Street from April to June 2003. The investigation range included:

- measurements of traffic density and structure,
- measurements of PM10 concentration,
- determination of 16 PAH content in aerosol samples.

Measurements of the density and structure of vehicle traffic were performed within 8 days in one-hour intervals for a selected cross-section of the route. The number of light and heavy vehicles moving on the tested road was determined.

The level of PM10 was determined by the gravimetric method using silica filters. Airborne particles were sampled according to the regulations of the PrPN-EN 12341 Draft Standard, at six selected measurement points located in the street canyon at a distance of 100 m form the tested communication route. Measurements were carried out within both 12 and 24 hours. The MVS6D reference aspirators made by Atmoservice, having a certificate for conformity with the CEN EN 12341 Standard, were used for sampling.

The following meteorological parameters: wind speed and direction, air temperature and humidity, were also measured during aerosol sampling. All particular PAHs were determined in the selected samples of airborne particles by capillary gas chromatography using flame ionization detector (FID). Samples were extracted in methyl chloride using an ultrasonic bath, then the extract was filtered and concentrated in helium atmosphere. Concentrated extract was diluted in isopropanol and redistilled water to the required ratio and such prepared samples were extracted to a solid phase. Next, samples were analyzed by the HP5890 Hewlett-Packard gas chromatograph with flame ionization detector. Samples were put into the dispenser of split/splitless type. Evaporator temperature was 260°C and detector temperature was 280°C. The analysis was carried out using a column operating with programmable temperature growth.

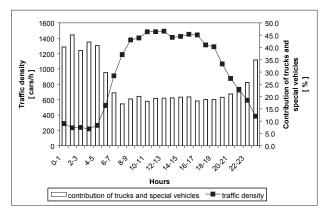


Fig. 1. 24-hour traffic density.

	PM10 concentration [μg/m³]									
Measurement place	24 hours			Day			Night			
	average	δn	N	average	δn	N	average	δn	n	
street canyon	94.0	33.0	16	125.5	47.5	4	69.0	12.4	6	
street canyon - rain	41.9	10.3	6	61.5	9,5	4	40.6	9.2	4	
100 m from the road	54.0	15.9	16	42.3	3.8	4	50.2	14.0	4	

Table 1. Average PM10 (airborne particles with aerodynamic diameter ≤ 10 μm) concentrations in the studied area.

Results and Discussion

Fig. 1 shows a daily profile of vehicular traffic. As can be seen, the traffic density increases rapidly after 8 a.m. from the level about 300 vehicles per hour to over 1,000 vehicles per hour. It should be noted that high traffic density, about 1,400 vehicles per hour, is maintained from 9 a.m. to 6 p.m. Similar heavy traffic has been recorded in large Western European cities [17, 25].

Table 1 contains average values of the PM10 concentrations at the road and at a site 100 m from the road. As can be seen, daily-average concentrations of the PM10 100 m from the road is 54 μg/m³. This value is in agreement with average concentration levels obtained in 1995/ 96 in cities in southern Poland, especially in Upper Silesia [26], where the PM10 concentration in summer varied from 61 µg/m³ in Świętochłowice (center of Upper Silesian Industrial Region - GOP) to 45 μg/m³ in Pszczyna, a city on southern edge of GOP. We can then conclude that earlier processes (i.e. in the 1980s) of rapid decrease of airborne dust concentration in Upper Silesia [27] has been stopped within the last five years. We can make a similar conclusion, when analyzing the measurements of the PM10 concentrations obtained in 2000, in downtown Katowice [28].

In the canyon of Pszczynska street the PM10 concentration is 94.0 μ g/m³, meaning that the impact of this street on the environment causes a relative increase of PM10 particle concentration by 74% in its nearest surroundings. Atmospheric precipitates reduce the PM10 concentration by about 40%, in relation to the level of rainless period, but it does not change the fact that the PM10 daily concentration above 90 μ g/m³, which remains most of the day, is hazardous to the health of inhabitants living in that street. Thus, we can expect about a 10% increase of respiratory diseases in the exposed population. This results from the earlier-discussed epidemiological data, which connects an increase of PM10 concentration by 10% with a few percent increase of respiratory incidents.

The last columns in Table 1 include the 12-hours concentration levels for day and night periods. As can be seen, a concentration 100 m from the road is slightly higher in the night than during the day. Such results are often observed and can be connected with changing meteorological conditions during the night, which are favorable to condensation of pollutants. Besides, increased pollutant emissions

from industrial sources during the night can sometimes be observed. On the contrary, a close road vicinity emission from cars decides about the pollutant concentration level, so as expected, the PM10 concentration near the road is significantly reduced during the night. However, a night reduction in airborne particles emission seems not to be adequate to the five times reduction of traffic intensity in those night hours. Undoubtedly, the reason is the fact that in the night a contribution of trucks to the total car stream increases by over two times.

Figure 2 shows an exemplary PAH chromatogram of airborne particles sampled at Pszczynska Street. The PAHs concentrations are given in Table 2. As it can be seen, average total concentrations of PAHs 100 m from the road is 118.44 ng/m³. This result agrees well with the PAH concentrations recorded in Gliwice in 1992 in the summer season [29], which confirms the hypothesis about stopping a decline in air pollution in Upper Silesia in the second half of the 1990s. An analysis of results presented in Table 2 shows that mass of polycyclic aromatic hydrocarbons contained in the studied aerosol makes 0.28% of the mass of this particulate matter. The average total concentration of PAHs at the road is 191.56 ng/m³, which means a 62% increase in relation to a distant place (background). The increase is significant, but over 10% lower than a relative increase of airborne dust concentration. It also should be noticed that the aerosol in road vicinity contains fewer PAHs in percent than at the reference point. At the road, PAH content is only 0.15% of the PM10 mass. In the rainy season average total PAH concentration was 141.36 ng/ m³, a decrease to 74% of the concentration from rainless period. That much lower decrease in PAH concentration than a decrease of airborne particles concentration (PM10 concentration, as it was given earlier, in a rainy season is only 40% of initial concentration) can be related with more efficient absorption of gaseous PAHs on the particles during rain, which can partly compensate for wet deposition of particles. The above hypothesis, which needs further study, is supported by the fact that airborne particles contained almost two times more PAHs in rainy season (0.29%) than in the days without rain (0.15%).

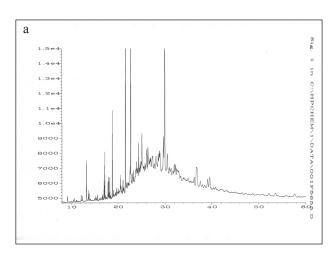
The contribution of each emission source in a recorded PAH concentration level can be assessed on the basis of PAH profiles (that is by an assessment of each

Measurement place	РАН со	PAHs/PM10		
	average	δn	n	[%]
street canyon	191.56	102.8	7	0.15
street canyon – rain	141.36	12.0	4	0.29
100 m from the road	118.44	45.2	4	0.28

Table 2. Average concentration of polycyclic aromatic hydrocarbons (PAHs) contained in airborne particles in the studied area.

component proportion). At present, the car exhaust gases and low emissions (from house heating systems) are indicated as the main sources of PAHs in urban air [30]. A contribution of these two emitters groups in the PAH concentration at Pszczyńska Street can be assessed by analyzing the proportion of IP (indeno[1,2,3-cd]prene) to BNT (benzo[b]naphto[2,1-d] thiophene). If the proportion of IP to BNT is 0.4, then PAHs are coming mainly from cars, when this ratio is 0.9 then recorded PAHs are of municipal origin [30-32]. An IP/BNT quotient calculated from data in Table 1 is on average 0.4, which means that the PAHs contained in airborne dust particles at Pszczyńska Street come mainly from car exhaust gases. However, the IP/BNT quotient does not enable us to discover the influence of industrial sources. Meanwhile, this kind of emission is very important in the studied area due to the coke refinery's vicinity. Coking plants placed in surrounding cities, especially in Zabrze, probably strongly influence PAH content in aerosol particles in Gliwice. Øvrebø et al. [29] and Bodzek et al. [33] have already paid attention to the significance of coking plants in intoxication by PAHs for the population living in Upper Silesia Industrial Zone. In the last decade emissions from this sector have been reduced, but PAH concentrations in a site 100 m away from Pszczynska Street (reference point) which exceed 100 ng/m³ indicates that hydrocarbon emissions from industrial and municipal sources still has an essential contribution to

air pollution in Gliwice. The already discussed fact of higher PAH content in airborne particles in the reference point than near the road also confirms the hypothesis of a significant role of industrial emissions in polluting of air by PAHs. The significance of that emission together with the municipal emission is especially visible outside the road vicinity. During winter season we can expect even more increase in communal emission contribution in air pollution. It should be remembered that burning hard coal is the main source of PAH emissions. It especially concerns BaP emissions. It was calculated that 30% of total BaP emission in the former Federal German Republic in 1981 came from coke burning, 56% from coal burned in heating systems and 13% from motor transport, while from oil heating systems and power industry based on coal burning only below 0.5% [34]. In Polish conditions the hypothesis is supported by studies carried out eleven years ago in Gliwice, which showed the seasonal changeability of PAH concentrations [29]. PAH concentration in September 1992 was 150 ng/m³ (summer average concentration was 60 ng/m³), and in February next year it was even 440 ng/m³, which means a relative increase by 193%. That increase should be connected with PAH emissions from the heating systems based on hard coal burning. Thus, we can expect that winter increase of PAH concentration is over three times higher than a relative increase of concentrations caused by the impact of road.



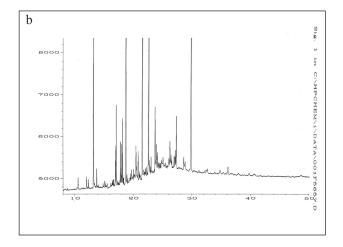


Fig 2. Exemplary chromatogram of polycyclic aromatic hydrocarbons (PAHs) contained in PM10 (airborne particles with aerodynamic diameter \leq 10 μ m). a) in the canyon of Pszczynska Street in Gliwice; b) 100 m from the road.

In Fig. 3 a percentage contribution of each of the PAHs in total PAH concentration at the road and in the reference site has been given. As can be seen, benzo/a/pirene makes 3.1% of total PAH concentrations at road vicinity, which is transformed into a concentration of 4.4 ng/m³ in ambient air, and 4.3% of total PAH concentration 100 m from the road, which gives a concentration of about 5 ng/m³ in air. Thus, the BaP concentration at the road decreases to 1.9 ng/m³ at rainy period. These data indicate that pollutant emissions from cars are not the main source of the BaP emission and support a thesis about the key role of industrial and municipal emitters in the PAH balance in Upper Silesia.

A comparison of literature data with the BaP concentration in this work leads to the conclusion that the concentration of this pollutant in Gliwice was reduced by 4-5 times in relation to values recorded in 1988-89 [33] and two times in relation to concentrations recorded in 1992-93 [29]. Nevertheless, the present BaP concentration in Gliwice can be regarded as high. It is over two times higher than an average BaP concentration recorded in London within years 1991-92 [25]. However, similar concentrations were recorded in 1990 in Bottrop and Castrop-Rauxel-Ickern in Germany [35].

Obtained data enable us to assess the cancer risk for inhabitants at Pszczynska Street in Gliwice due to exposure to BaP. According to the WHO Air Quality Guidelines [34], the risk can be written as follows:

Risk = $4.4 \text{ ngBaP/m}^3 \times 8.7 \times 10^{-5} \text{ m}^3/\text{ngBaP}$, so it is of 10^{-4} order.

Due to approximate BaP concentrations recorded in different sites, the same risk can be attributed to the population living on the area far from the tested street.

It is interesting that the same cancer risk (2.1–4.2 x 10⁻⁴) was also estimated for North Rhine-Westphalia inhabitants living near busy roads [35] as they were exposed to the BaP concentration within the range of 3 to 6 ng/m³.

Besides, inhabitants for Pszczynska Street are exposed to other carcinogenic substances. First of all there are higher total PAH concentrations than in a distant area

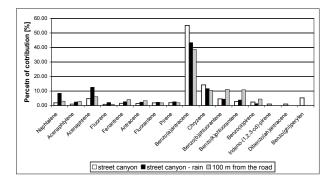


Fig 3. Average contribution of each, single PAH compound in the total PAHs concentration.

from the street. There we can also expect a high sooth concentration and elevated concentration of asbestos [36, 37] as well as cadmium and arsenic. Therefore, we can roughly assess total cancer risk (without a synergism effect), assuming a 70-year exposure, as 10-3. A similar result was achieved by earlier-cited [35]. However, the disease risk of inhabitants at Pszczynska Street results, first of all, from exposure to high concentration of airborne particles. We can expect an increased number of incidents of respiratory diseases among that population. Further studies are needed in that area, which should also include an identification and quantitative assessment of allergic illnesses.

Conclusions

The results show that a process of rapid decrease of atmospheric aerosol concentration in Upper Silesia, which took place in the 1980s and at the beginning of the 1990s has been stopped.

In the canyon of busy Pszczynska Street the daily average of PM10 concentration increases by 40 μ g/m³ in relation to sites 100 m from the road. For inhabitants living at this street, that implies about a 10 percent increase of respiratory incidents.

Total PAH concentration at the road was 191.56 ng/m³ (in spring and without rain) and was over one and half times higher than at the point 100 m away, which confirms the fact that emission of exhaust gases at busy roads increases an exposure to PAHs. However, it does not concern the BaP.

Unlike urban areas in Western Europe, where cars are the most important source of BaP emissions, in the Upper Silesian Region the main role still plays the BaP emission from industry and municipal sources.

BaP exposure embodies a greater population of Gliwice than that living on a busy street and it generates the cancer risk of 10^{-4} order. A total cancer risk for people living at the studied street has been assessed as 10^{-3} .

More precise calculations of cancer risk and its spatial and inter-population distribution require further studies.

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