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Original Research

Environmental Impacts of Long-Term Air Pollution Changes in Kraków, Poland

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Abstract

Annual concentrations of SO_2 and PM have decreased in Kraków since the early 1970s by 70-80%. At present, both pollutants still show the same seasonal pattern as in the 1970s, with maximum in winter and minimum in summer. PM10 is the most abundant air pollutant reaching permanently the levels far behind the allowed values. Since 2001, an increasing concentration trend has been observed, while the concentrations of SO_2 usually stay below the allowed limits. The concentrations of NO_2 and O_3 have been measured since 1992. The allowed values of NO_2 are exceeded only in the high-traffic site (about 10 times per year) while the levels of O_3 are usually within the allowed limits. Studies concerning plant and animal contamination with heavy metals (Cd, Pb, Fe, Zn, Cu) show that cadmium is at present the main danger. In spite of a significant improvement of aerosanitary conditions in Kraków after 1990, negative long-term effects of air pollution on human health can be still observed, especially in children.

Keywords: Kraków, air pollution, health effects, bioindicators

Introduction

After the Second World War, Poland experienced the socialist industrialization that resulted in a significant deterioration of the country's environment. Four regions were classified as ecological disaster zones [1, 2], and Kraków is located in one of them. However, the city is of special cultural and historical importance and since 1978 has been included in the UNESCO List of Natural and Cultural Heritage sites. In 1989, the collapse of communism in Central Europe forced a change of economic systems that have had a profound effect on the environment. In Poland the air pollution emissions decreased significantly due to restructuring of industry, with much smaller production adjusted to the need of the market economy and the introduction of modern technologies. For example, the national emission of SO_2 decreased from 4.3 million tonnes in 1986 [3] to 1.2 million tonnes in 2005 [4]. Consequently, imissions decreased, too, and the air pollution in Kraków has improved similar to the rest of the country, but it is still one of the most polluted Polish cities, with annual emissions of 4.6 thousands tonnes of particulate matter and 6911.7 thousand tonnes of gases in 2006 [4]. The present paper shows the changes in air pollution (SO_2, PM, NO_2, O_3) in Kraków since the late 1960s until 2006, and their various biological impacts that have been studied in Kraków since 1970s. Pollution by particulate matter turned out to be especially harmful for plants, animals and human health, due to the heavy metal content. As the decreasing trends in air pollution have been slowed down or even reversed recently, it is of paramount importance to evaluate the extent of the environmental response to the decrease of air pollution after 1989 and to estimate the environment's vulnerability and inertia in that aspect. Studies concerning the environment

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of Kraków and its surroundings, performed during last over 30 years, allow us to attempt such a task.

The city of Kraków is located in a concave landform that has a large impact on air pollution dispersion conditions (Figs. 1 and 2). The historical city centre is placed in the Vistula river valley bottom (at about 200 m a.s.l.), going from west to east, and also on a limestone tectonic horst (the Wawel hill), emerging from the river valley. Other parts of Kraków occupy both the areas in the river valley with its terraces, and in convex land forms to the south (the Carpathian Foothills, up to 370 m a.s.l.) and to the north (The Kraków -Częstochowa Upland, up to 300 m a.s.l.) of the city centre. The Vistula valley is narrow in the western part of Kraków (about 1 km) and widens to about 10 km in the eastern part. In the western part are several limestone tectonic horsts, reaching about 350 m a.s.l. The city area is not surrounded by the hills only from the east, which makes natural ventilation difficult and favours the occurrence of high air pollution [5]. The geological map of Kraków is available at: http://www.ing.pan.pl/muzeum/5mapage.htm

Fig. 1. Location of Kraków.

Apart from the landform, also the prevailing winds (direction and speed), temperature inversions and urban heat island intensity have a huge impact on air pollution dispersion and concentration. Mean annual wind speed in Kraków is 2 $m \cdot s^{-1}$ and for about 50% of the year the mean daily wind speed does not exceed that value [6, 7], which means a prevalence of poor horizontal ventilation conditions. Atmospheric calms occur on 20-30% of the year, and only on 35% of days does the mean daily wind speed exceed 5 $m \cdot s^{-1}$ [8]. The increase in wind speed does not cause a decrease of air pollution in some parts of the city, e.g. in the southern Podgórze district. Moreover, stronger winds from NE and NW bring air pollution from external, distant sources [9]. Morawska-Horawska [10] investigated vertical distribution of wind directions above Kraków. In winter, when high air pollution often occurs and SW winds prevail, the pollution from the steelwork's high chimneys (about 250 m) is usually taken away from the city while the pollution from the power plant in nearby Skawina (located SW of Kraków) is brought to the city, which was also proved by Niedźwiedź and Olecki [11]. Pollution from the power plant endangers mainly the eastern part of the city.

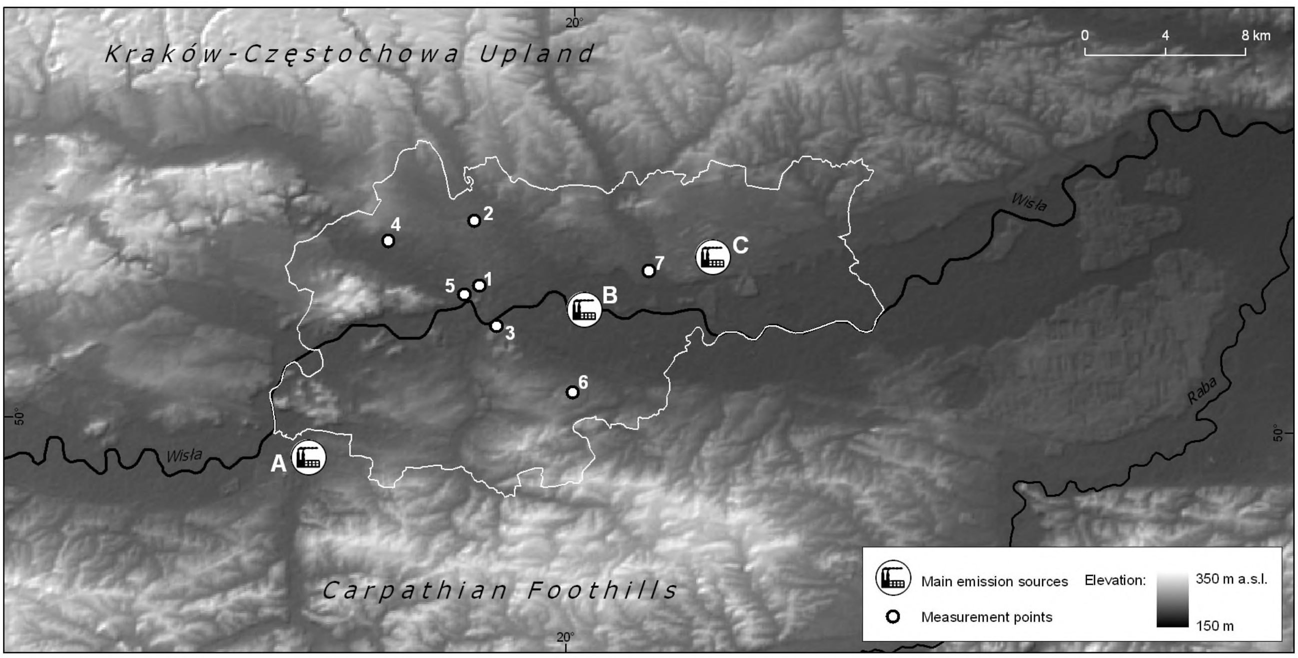

Fig. 2. Relief of Kraków and its environs, the location of air quality monitoring points and main air pollution emission sources. Explanations: white line – administrative borders of Kraków; measurement points of the Voivodship Inspectorate for Environmental Protection in Kraków: 1 – Rynek Główny, 2 – Prądnicka St., 3 – Rynek Podgórski, 4 – Balicka St., 5 – Krasińskiego St., 6 – Prokocim Nowy, 7 – Bulwarowa St.; air pollution emission sources: A – power plant in Skawina, B - Kraków-Łęg Combined Heat and Power Plant, C – steelworks of ArcelorMittal Steel Poland.

In 1970 and 1972, the thermal structure of the lower troposphere (up to 3,000 m above the ground) over Kraków was investigated with airplane measurements. Inversions can occur at any time of the day and usually reach 200 m, but in anticyclonic systems they can extend to 50-800 m; in cyclonic systems 100-600 m. Most chimneys of local heating facilities emit the exhaust gases at a height below 200 m. When combined with frequent temperature inversions it significantly worsened the city's aerosanitary conditions [12]. Sodar measurements performed in Kraków since 1980 by the Institute of Meteorology and Water Management, Kraków Branch, proved that even during 20% of the year, all-day inversions can occur. During the night, in the Vistula River valley, stable atmospheric conditions dominate and threrefore in 86% of cases the temperature inversion was observed. During the remaining 14% of nights, neutral atmosphere conditions were noted. In Kraków, low inversions prevail, i.e. the inversions reach from the ground level up to 150-300 m. Above that, lifted inversions can occur. The low location of the inversion layer is characteristic mainly for the cold half-year, while in summer during the intensive convection conditions, the layer may occur as high as 800 m above the ground [7, 13, 14].

The intensity of the urban heat island, a factor improving air pollution dispersion, was estimated in the 1970s to be 1.2°C on average [15]; the present value is unknown. In 1979, balloon measurements performed up to 500 m above the ground showed that the urban heat island and temperature inversion can occur together and that heat emission from the non-industrial chimneys increases the frequency of low inversions (up to 50 m) above the city [16]. The number of days with fog in the centre of Kraków decreased from about 120 days per year in the 1960s to about 20 in the 1990s [17], which is a factor improving the aerosanitary conditions.

Aerosanitary conditions in Kraków are not favourable because of its specific geographic and meteorological conditions as well as human activities that cause high pollution emissions. Before the Second World War, the area of the city was 47 km^2, and the number of inhabitants amounted to 163,000. Then the city was gradually enlarged to 327 km^2 and at present (2005) has 757,400 inhabitants [18-20]. After the war, Poland was one of the most heavily damaged countries. The destruction in industry amounted to 40 percent of fixed assets and Kraków was one of the very few Polish cities that was not ruined by the Germans. Unfortunately, the national economy could not be rebuilt in the same way as in Western Europe because Poland was incorporated into a group of states whose foreign and domestic policies were dominated by the Soviet Union (USSR). Socialistic industrialization became the utmost goal of the state economic and political activity, in order to achieve the defensive ability demanded by the Cold War [21]. That is why in 1954, a huge steelworks (the Lenin Steelworks; today called ArcelorMittal Steel Poland, Branch Kraków) was completed about 10 km east of Kraków's medieval old town, in Nowa Huta (today a Kraków district). That changed dramatically the spatial development and employment structure of Kraków. The steelworks has been from the beginning the largest source of air pollution in Kraków. In the 1970s, 46% of industrial production in Kraków was generated by the steelworks, which was planned to produce 1.5 million tonnes of steel per year, but in late 1970s produced as much as 7 million tonnes of steel per year [22]. In 1990, the production volume decreased to 1.7 million tonnes of steel per year [23], due to the political and economic system change in Poland; the steelworks began to function according to the free-market economy and had to adjust to the real steel demand instead of producing as much as ordered by the USSR. Another significant source of air pollution in Kraków is the Kraków-Łęg Combined Heat and Power Plant in Nowa Huta that operates on coal and produces electricity and heat for most of the inhabitants of newer districts. In older parts of the city, local heating facilities operating on coal are still serious air pollution sources. Transportation has an increasing role in air pollution origin. In 1972, there were 52,500 cars registered in Kraków, while in 2001 the number reached 253,400 [20, 24]. The transportation area within the city increased from 2974 ha in 1990 to 3281 ha in 2005 [20, 25]. Apart from the air pollution sources within the city, there is a power plant in Skawina (about 5 km from the south-west Kraków's border) and the Upper Silesia industrial region (about 80 km west of Kraków) which deliver long-distance air pollution transported by the prevailing western winds [7, 8].

Table 1. Total emissions (in tonnes per year) of gases and particulate matter from the registered sources of air pollution in Kraków. Since 1992 data for gases without CO_2. Source of data: [20, 26, 27].

Year	Gases	Particulate matter
1970s	850,000	150,000
1992	220,000	30,000
1999	74,300	5,900
2003	61,700	4,800
2004	74,600	5,700

Total emissions (in tonnes per year) of gases and particulate matter from the registered sources of air pollution in Kraków are presented in Table 1. Until 2003 a decrease of both pollutants was observed, while in 2004 emissions increased.

Methods

Study Area

The old town of Kraków has been on the UNESCO List of Natural and Cultural Heritage sites since 1978, which was one of the reasons for intensification of air pollution measurements and environmental impact research in the 1980s. But the first systematic measurements of air pollution were started in 1949-1951 by the National

Table 2. Stations of the automatic air pollution monitoring network in Kraków and the data used in the study.

Station	Location	Characteristics	Period	Pollutants
Rynek Główny Sq.	19°56'12"E 50°03'42"N 210 m a.s.l.	city centre, intensively build-up, pedestrian zone with very limited traffic	1993-2003	SO_2, PM10, NO_2, O_3
Prądnicka St.	19°55'57"E 50°05'27"N 225 m a.s.l.	northern part of the city, dispersed build-up	1994-2006	SO_2, PM10, NO_2, O_3
Rynek Podgórski Sq.	19°56'55"E 50°02'38"N 203 m a.s.l.	southern part of the city, intensive build-up, high traffic site	1993-1999	SO_2, PM10, NO_2
Balicka St.	19°52'23"E 50°04'53"N 218 m a.s.l.	western part of the city, dispersed build-up	1993-1999	SO_2, NO_2, O_3
Krasińskiego St.	19°55'35"E 50°03'27"N 203 m a.s.l.	city centre, urban canyon, high traffic site	1993-2006	SO_2, PM10, NO_2
Prokocim Nowy	20°00'08"E 50°00'53"N 228 m a.s.l.	southern part of the city, dispersed build-up	1993-2002	SO_2, PM10, NO_2, O_3
Bulwarowa St.	20°03'13"E 50°04'10"N 204 m a.s.l.	eastern part of the city, 1 km from the steel works, at the border of a green belt and intensive build-up	*1993-2006	SO_2, PM10, NO_2

* - in the years 1993-97 the station was in two other locations than the present one, but all three are within 1 km of each other

Hydrological and Meteorological Institute (at present the Institute of Meteorology and Water Management); they comprised particulate matter concentration measurements. In 1966, the Voivodship Sanitary-Epidemiological Station in Kraków started to measure particulate matter and gaseous pollution concentration. In 1988, in the Main Market Square (old town centre), automatic measurements of SO_2 and NO_x were started thanks to the cooperation with the city of Göteborg, but they lasted only until 1991. Since 1991, an automatic measurement network has operated in Kraków. It was a donation from the U.S. Environmental Protection Agency for Kraków, as the city was considered a unique and very endangered one. The network is administered by the Voivodship Inspectorate for Environmental Protection in Kraków. Originally it consisted of 7 measurement points, and now it is reduced to 3 points within the city borders [28]. Additionally, since 1997 the measurements of CFCs and SF6 have been realized in Kraków by the Institute of Nuclear Physics, Polish Academy of Sciences [29], and since 1982, the Department of Environmental Physics, University of Science and Technology in Kraków, monitors the concentration and isotope composition of CO_2 in the city [30].

Air Quality Monitoring

The data used in this paper comes from the measurements of the Voivodship Sanitary-Epidemiological Station in Kraków (1968-2001) and the Voivodship Inspectorate for Environmental Protection in Kraków (1992-2006). In the first period, mean data for four districts of the city are used (average from all measurement points in a particular district), while for the second period exact data from the measurement points are presented. The pollutants analyzed are: PM10, SO_2, NO_2, and O_3. The measurement methods and number of measurement points changed with time. In the first period, only SO_2 and suspended particulate matter were measured in 20 samplers placed all over the city (some additional measurements were realized in short-term series and only chosen points). The colorimetric method for SO_2 and reflectometric method for PM were applied. In 1993-2006, the continuous measurements of PM10 (with weight method), SO_2 (with fluorescent method), NO_2 (with chemiluminescent method) and O_3 (with UV method) were realized in automatic stations equipped with instruments produced by Thermo Environmental Instruments, Andersen, FAG, Rupprecht&Patashnick, Environment S.A, ISECO and DIGITEL. Their location and number changed [31]. For the period 1968-2001, the results are shown for the following districts: Śródmieście (city centre), Krowodrza (northern part of the city), Nowa Huta (eastern part of the city), Podgórze (southern part of the city). In 1993-2006, the measurements were realized in the locations presented in Table 2 and Fig. 2. In the following sections the measurement results are compared to limit values which are defined in the ordinance of the Ministry of Environment of the Republic of Poland of 6th June, 2002, published in the Official Journal of Laws, No. 87/796. Only the limit values used at present are recalled; in the past those values were usually higher.

Results

Changes in Air Pollution

Sulphur Dioxide

Mean annual concentrations of SO_2 in the districts of Kraków in 1968-2001 are presented in Fig. 3, while Fig. 4 shows the data for 1993-2006. The allowed limit value of 20 $\mu g{\cdot}m^{-3}$ was exceeded almost in the whole area of the city until 1999, and since 2000 it has been above the allowed level only occasionally and only in the measurement point located in the high traffic site in the city centre (Krasińskiego St.). Mean annual concentrations decreased from about 80-120 $\mu g{\cdot}m^{-3}$ in the mid-1970s to less than 20 $\mu g{\cdot}m^{-3}$ in 2006. A striking feature is the disappearance of huge differences in the concentrations among the different districts. Until the mid-1990s, the highest SO_2 concentrations were noted in the city centre, where many local heating facilities operating on coal were delivering lots of SO_2 and particulate matter to the lower parts of the urban boundary layer. In the years 1968-1973, mean daily concentration of SO_2 reached even 119 $\mu g{\cdot}m^{-3}$ there and in the remaining city area only 53 $\mu g{\cdot}m^{-3}$. Mean monthly concentration in the warm half-year was about 30 $\mu g{\cdot}m^{-3}$ all over the city while in the cold half-year the values reached about 350 $\mu g{\cdot}m^{-3}$ in the city centre and 150 $\mu g{\cdot}m^{-3}$ in the remaining city area [32].

In the period 1993-2006, a clear change in mean monthly concentrations at all stations occurred. Until 1998, in winter the concentrations reached about 100 $\mu g{\cdot}m^{-3}$ in the city centre and around 60 $\mu g{\cdot}m^{-3}$ in other areas. In summer the concentrations all over the city dropped below 20 $\mu g{\cdot}m^{-3}$. Then since 1999, winter concentrations have reached about 40 $\mu g{\cdot}m^{-3}$ and summer ones about 10 $\mu g{\cdot}m^{-3}$. The differences between stations are below 10 $\mu g{\cdot}m^{-3}$. That seasonal pattern seems to be fixed, although during a frosty winter, like in 2005/2006, the concentrations at all stations rise to about 60 $\mu g{\cdot}m^{-3}$.

Daily concentrations of SO_2 available for the years 1993-2006 were used to count the number of days with mean 24-hour concentrations exceeding 125 $\mu g{\cdot}m^{-3}$, which is the allowed value according to health protection standards. The limit should not be exceeded on more than 3 days per year. Such days can occur in Kraków from October to April. (Fig. 5 shows their annual number in particular stations and years). Until 1997, there were about 20 such days per year in the city centre and about 10 in other parts of the city. Since then the daily limit of 125 $\mu g{\cdot}m^{-3}$ is exceeded very rarely, on 1-3 days per year, which is within the allowed value.

Particulate Matter

Figs. 6 and 7 show the changes in mean annual concentrations of the particulate matter in the years 1968-2001 and 1993-2006, respectively. At present, the allowed mean annual value for PM10 is 40 $\mu g{\cdot}m^{-3}$. The general trend of concentration changes shown in Fig. 6 is similar to that of SO_2 (Fig. 3), which is a huge decrease in all districts. In the years 1968-1973, mean daily concentration of PM10 in the city centre reached 180 $\mu g{\cdot}m^{-3}$ and in the remaining city area 109 $\mu g{\cdot}m^{-3}$. Mean monthly concentrations from May to August were about 60 $\mu g{\cdot}m^{-3}$ all over the city while from November to February the values reached about 400-600 $\mu g{\cdot}m^{-3}$ in the city centre and 250 $\mu g{\cdot}m^{-3}$ in the remaining city area [32]. However, in recent years the observed PM10 concentrations and their changes prove that particulate matter is still a huge problem. In the years 1992-2006, the allowed mean annual value was not exceeded only in the year 2000 (Fig. 7). The highest concentrations are always observed in the measurement point located in the high traffic site in the city centre (Krasińskiego St.), reaching 80 $\mu g{\cdot}m^{-3}$, i.e. 200% of the allowed value. Moreover, since 2004 an increasing trend can be observed for concentrations in all measurement points; the allowed value is exceeded everywhere.

Analysis of mean monthly values changes in 1993-2006 proves that until 1998 the seasonal changes in PM10 concentrations were characterized by winter maximum and summer minimum. In winter in the city centre the values were about 120-150 $\mu g{\cdot}m^{-3}$, while in other parts of the city around 75 $\mu g{\cdot}m^{-3}$. From 1999 to 2001, both summer and winter concentrations decreased significantly all over the city, with maximum below 75 $\mu g{\cdot}m^{-3}$. Additionally, the differences between stations became much less than before. However, since 2002 the old seasonal pattern has appeared again, with winter maximum in the city centre of 150 $\mu g{\cdot}m^{-3}$. During the frosty winter 2005/2006 it reached as much as almost 240 $\mu g{\cdot}m^{-3}$.

Daily concentrations show the problem even better. The allowed mean 24-hour concentration is 50 $\mu g{\cdot}m^{-3}$, according to health protection standards. The value should not be exceeded on more than 35 days per year. Fig. 8 shows the number of days when the thresholds of 50, 100 and 200 $\mu g{\cdot}m^{-3}$ are exceeded in particular stations and years. The situation is alarming. The number of days exceeding the allowed value is higher than 35 at all stations in all years, in the city centre it reaches even 250 days per year. At the beginning of the 1990s, the differences between the city centre and other areas were much larger than in 2005 and 2006, because recently the suburbs became more polluted with PM10, while the pollution in the city centre remains all the time at the similar level. Unfortunately, the PM10 concentration quite often exceeds 100 and 200 $\mu g{\cdot}m^{-3}$, especially in the city centre (100 and 20 days per year, respectively). In recent years, the number of days with PM10 concentrations above all the thresholds is increasing at all stations.

The content of heavy metals in PM10 in the years 1992-1999 was studied by Szafraniec and Jędrychowski [33]. Then similar analyses were carried out in 2005 within the Krakow Integrated Project [34]. The most significant changes were observed for lead. The mean concentration for the whole city decreased from 0.133 $\mu g{\cdot}m^{-3}$ in 1992-1994 to 0.079 $\mu g{\cdot}m^{-3}$ in 1997-1999 and in 2005 the value reached 0.093 $\mu g{\cdot}m^{-3}$. Mean concentrations of all heavy metals studied were below the allowed limit values. At present, transportation is the source of PM10 with the heavy metal content slightly higher than PM10 from other sources in Kraków. The chemical analysis of PM10 within the Krakow Integrated Project showed very high levels of benzo(a)pyrene, 50-250 times higher than the 1 ng·m^{-3} limit value. Additionally, Christoph et al. [35] proved that the airborne particulate matter in Kraków contains several times as many PCDD/FS dioxins than the particulate matter in Western Europe.

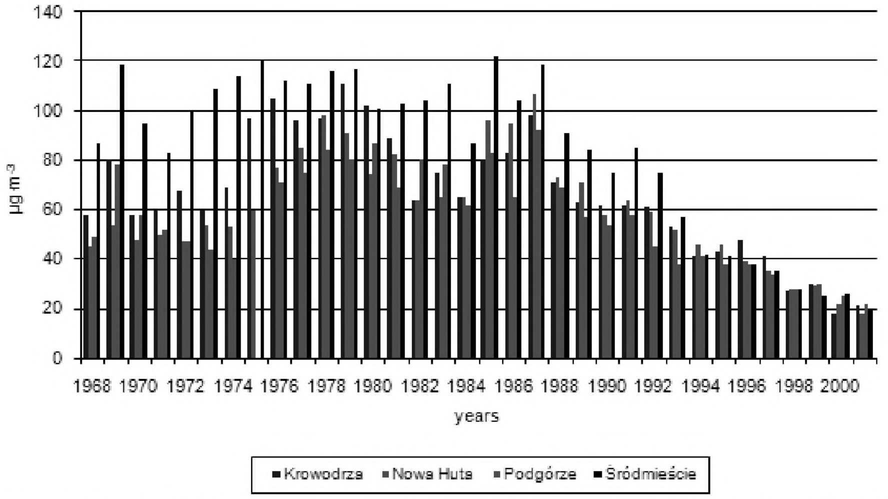

Fig. 3. Mean annual concentrations of SO_2 (in µg·m^{-3}) in the districts of Kraków in 1968-2001. Source of data: [26, 58, 59].

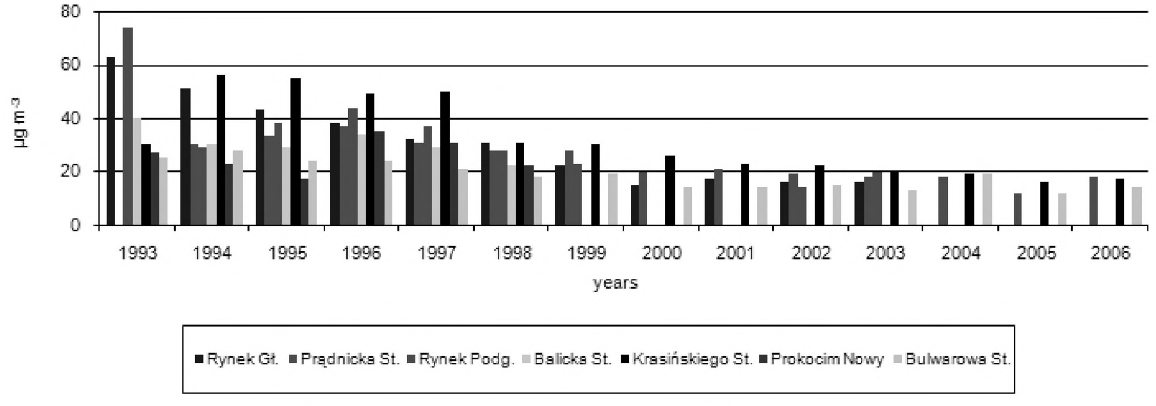

Fig. 4. Mean annual concentrations of SO_2 (in µg·m^{-3}) in measurement points in Kraków in 1993-2006.

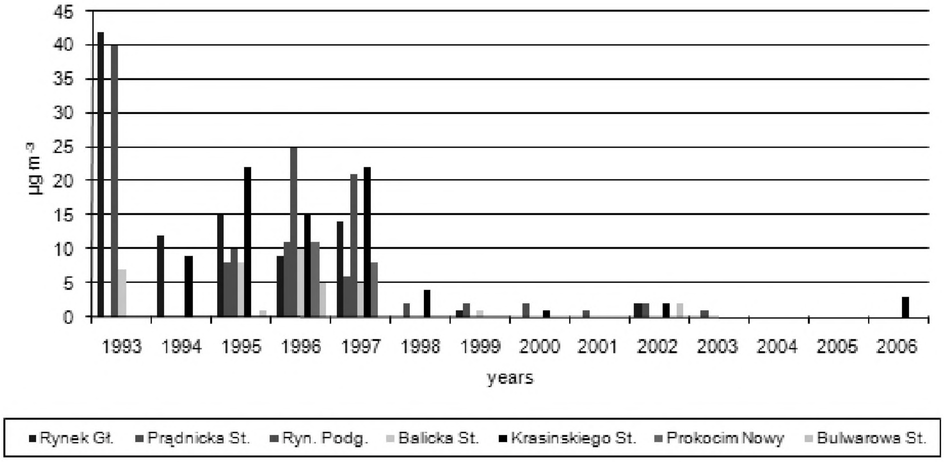

Fig. 5. Annual number of days with 24-hour SO_2 concentrations exceeding 125 µg·m^{-3} in measurement points in Kraków in 1993-2006.

Nitrogen Dioxide

Fig. 9 shows the changes in NO_2 annual concentrations in the period 1992-2006. The allowed value of 40 $\mu g \cdot m^{-3}$ is exceeded every year in the measurement sites with high traffic (Krasińskiego St.), as NO_2 is the pollution delivered mainly by the transportation. The spatial structure of the concentration occurrence seems to be rather stable in the whole period. No clear temporal trends can be observed. Therefore, too high concentrations of NO_2 are still a serious problem like PM10, but unlike in the case of PM10 they occur only in selected parts of the city. It is additionally proved by another index. The allowed hourly concentration on NO_2 is 200 $\mu g \cdot m^{-3}$ according to the health protection standards and it should not be exceeded more than 18 times per year. Such high values were noted in Kraków in the period 1994-2003 only in the city centre, especially at the high traffic site, usually up to 10 times per year, and only in 1997 did the value reach 19 cases (Fig. 10).

Tropospheric Ozone

Tropospheric ozone is a secondary air pollution and its mean annual concentration depends much more on weather conditions (number of hot and sunny days) in a particular year than on emissions of NO_x and organic compounds, which take part in the formation of photochemical smog. In 1999 the mean annual O_3 concentration was about 60 $\mu g \cdot m^{-3}$ and in 2003 it was about 50 $\mu g \cdot m^{-3}$, while in other years of the period 1992-2006 it usually reached 30-40 $\mu g \cdot m^{-3}$, (Fig. 11). The allowed value of O_3, according to the health protection standards, is 8-hour mean concentration of 120 $\mu g \cdot m^{-3}$ and it should not be exceeded on more than 25 days per year. In Kraków the value is exceeded rarely, usually on 15-20 days per year (Fig. 12), which is below the allowed limit. In 2003, there were as many as 47 days with exceeded limit value, but that was due to the extremely hot and sunny summer of that year.

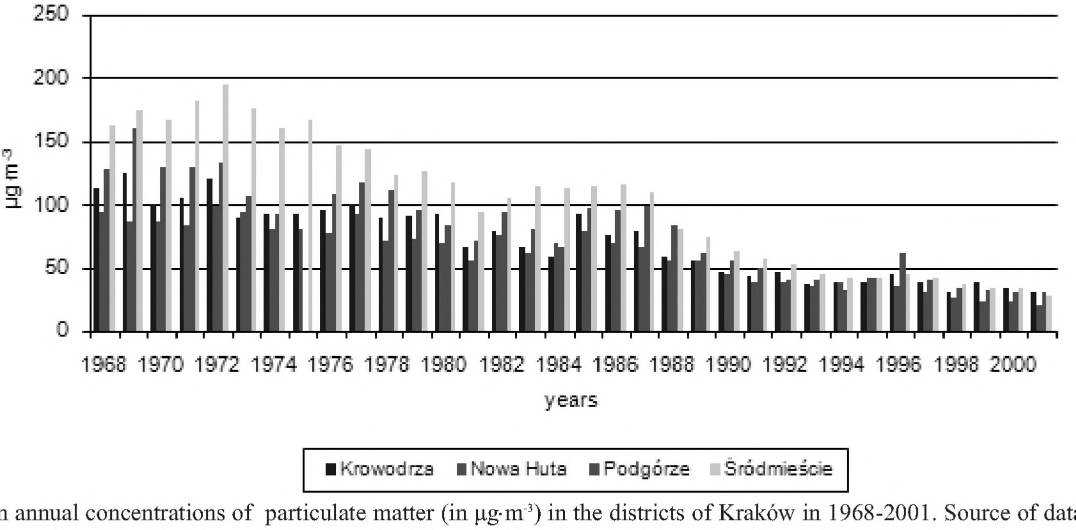

Fig. 6. Mean annual concentrations of particulate matter (in $\mu g \cdot m^{-3}$) in the districts of Kraków in 1968-2001. Source of data: [26, 58, 59].

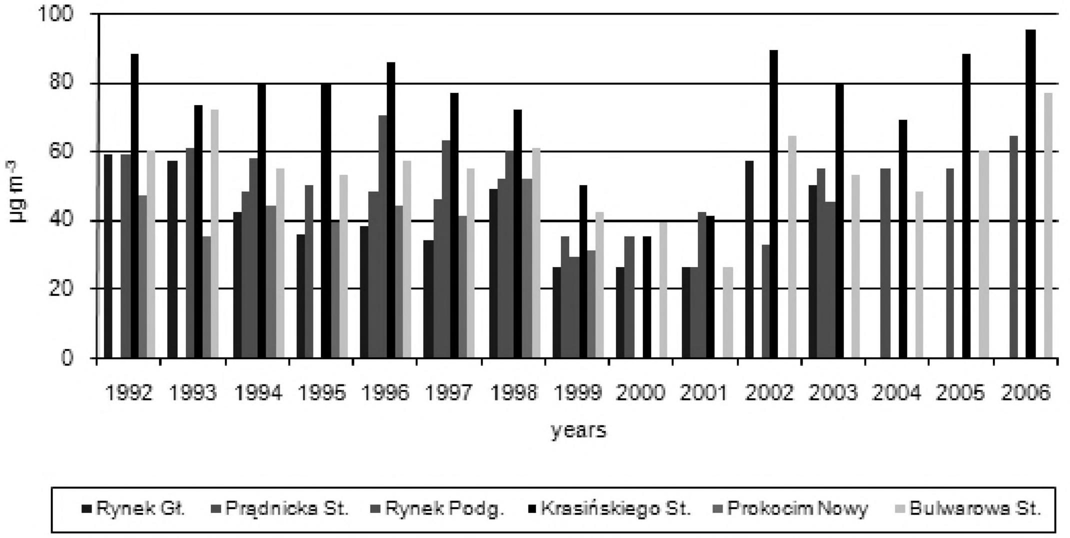

Fig. 7. Mean annual concentrations of PM10 (in $\mu g \cdot m^{-3}$) in measurement points in Kraków in 1993-2006.

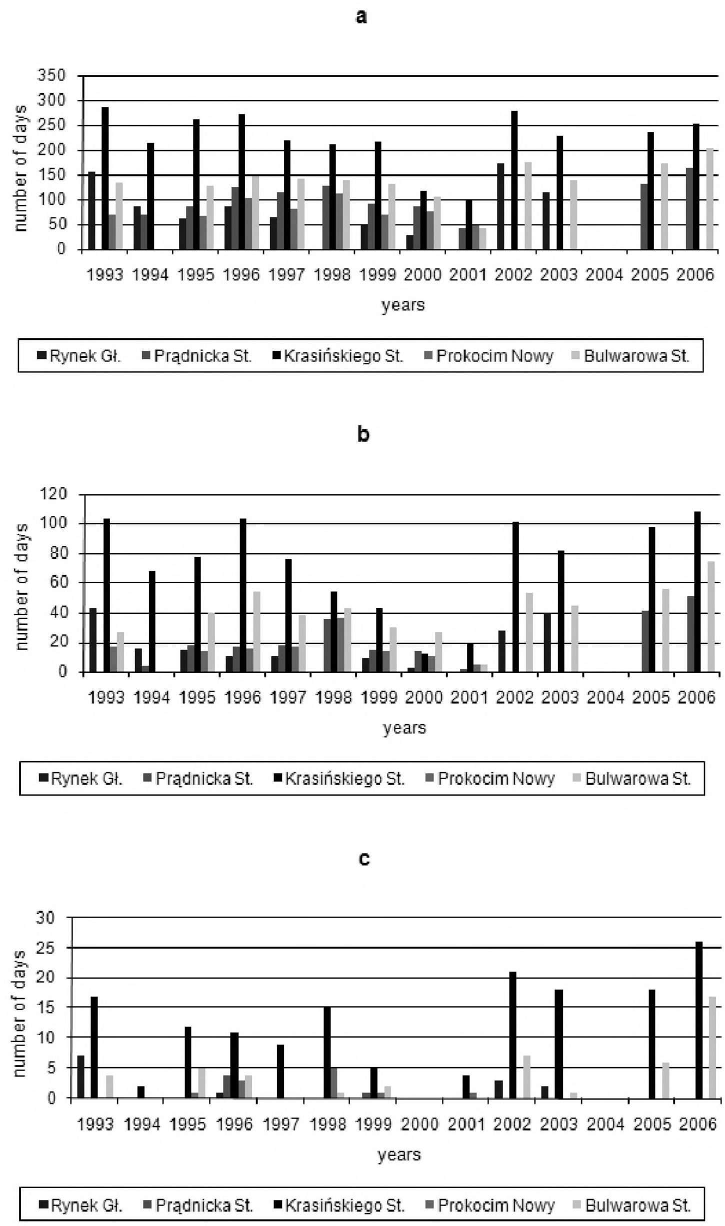

Fig. 8. Annual number of days when the thresholds of PM10 concentration >50 (a), >100 (b) and >200 (c) $\mu g \cdot m^{-3}$ were exceeded in measurement points in Kraków in 1993-2006.

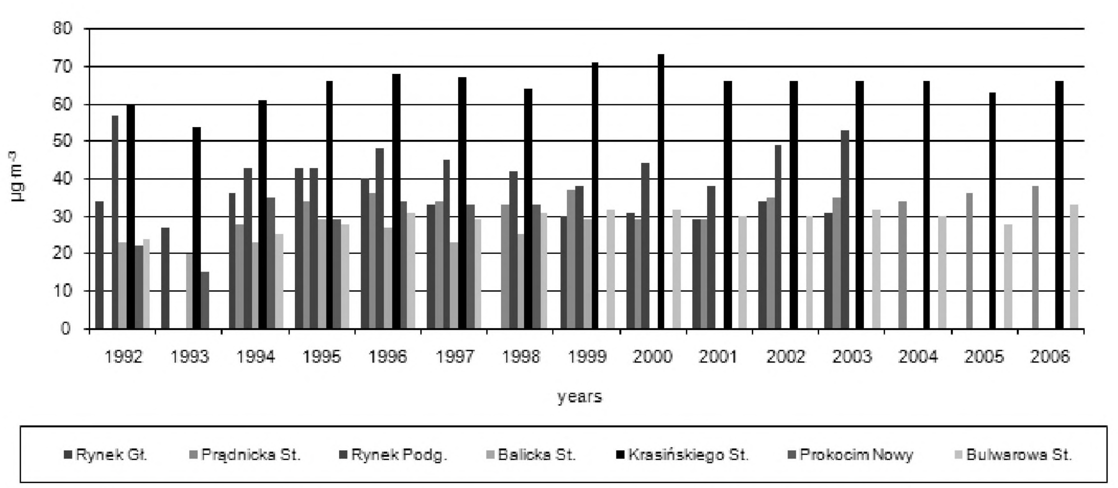

Fig. 9. Mean annual concentrations of NO_2 (in $\mu g \cdot m^{-3}$) in measurement points in Kraków in 1992-2006.

Impact on the Environment

The enormous increase in air pollution in Kraków after the Second World War has had various and profound effects on the environment. The impact on the health of Kraków's inhabitants has been studied since the 1970s. In the Niepołomice Forest, for instance located 10-35 km east of Kraków and covering the area of 11,000 ha, many studies were conducted to detect the air pollution impact on plants and animals. Two next sections present chosen research results showing the changing impact.

Human Health

In Kraków in the 1970s and 1980s, several negative health effects of air pollution were documented. Long exposure to SO_2 and suspended dust caused an increase in asthma cases among children in Kraków in comparison with children from control areas (Nowy Targ and Limanowa, S Poland) [36]. Studies performed in 1968-1981 proved that long exposure to high air pollution decreased respiratory system efficiency and increased the frequency of chronic respiratory system diseases. The death risk due to lung cancer among men increased in that period by 46% [37]. However, as proved by Krzyżanowski and Wojtyniak [32] men who smoke have a higher susceptibility to air pollutants. The studies on asthma among children were continued in the 1990s. In Poland, in the period 1984-1996, hospitalization rates from asthma in children and adolescents below 19 years old increased annually on average by 12.2% in boys and by 10.6% in girls [38]. The study realized in Kraków in 1995 proved that children with asthma resided more frequently in the city areas, with the higher air pollution level (62.5% vs 46.2%) and that the combined effect of air pollution and allergy on the occurrence of asthma in children was multiplicative [39]. In 1995-1997, the survey was continued and proved a strong association between lung growth and allergy in children and outdoor air pollution in the residence area. An increase in the occurrence of allergy, asthma and long-lasting infections of respiratory system was observed among children living in Kraków's districts with high air pollution [37, 40].

In the 1970s and 1980s, the content of oxygen in Kraków's air was permanently as low as 18%. Together with long exposure to high concentrations of CO and CO_2, in many cases it caused oxygen deficit in the heart, brain and kidneys. In result, 50% of deaths were associated with circulatory system diseases, while 20% with cancer. However, in 1980-1988, the share of deaths due to cancer increased by 30%, and one of the main factors was a high and permanent concentration of policyclic aromatic hydrocarbons [36]. The results of the ENHIS project for Kraków showed that in 2001 cardiovascular diseases still remained

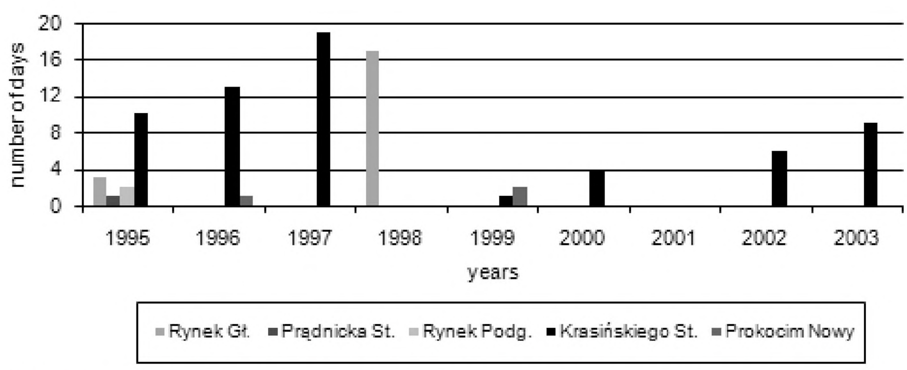

Fig. 10. Annual number of cases when hourly concentration on NO_2 exceeded 200 $\mu g \cdot m^{-3}$ in measurement points in Kraków in 1995-2003.

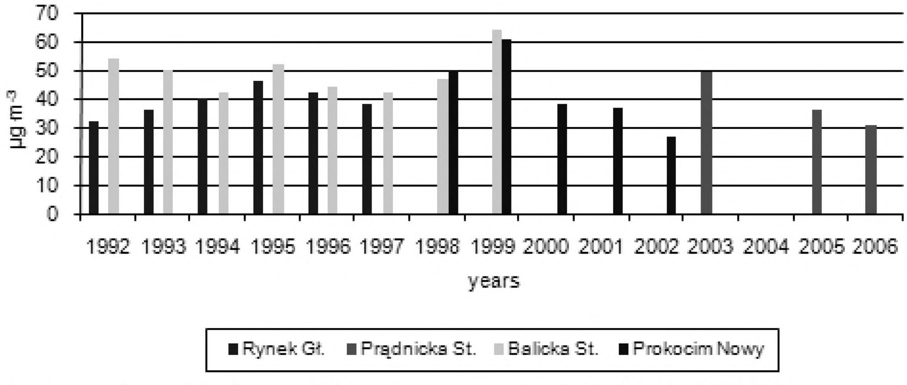

Fig. 11. Mean annual concentrations of O_3 (in $\mu g \cdot m^{-3}$) in measurement points in Kraków in 1992-2006.

Table 3. Concentrations (µg·g^{-1}, dry weight) of lead, cadmium, zinc and iron in the tissues of bank voles from the Niepołomice Forest and Kraków.

Year	Liver				Kidneys			
	Pb	Cd	Zn	Fe	Pb	Cd	Zn	Fe
1985 n=18	11.5	1.9	203	603	25.6	4.0	309	543
1998 n=14	-	1.1	90	638	-	4.2	91	455
2000 n=9	0.4	1.3	88	1080	1.9	7.2	93	674
2002 n=6	0.3	1.3	-	-	0.9	4.7	-	-

Sources: [52, 53, 54, 55]

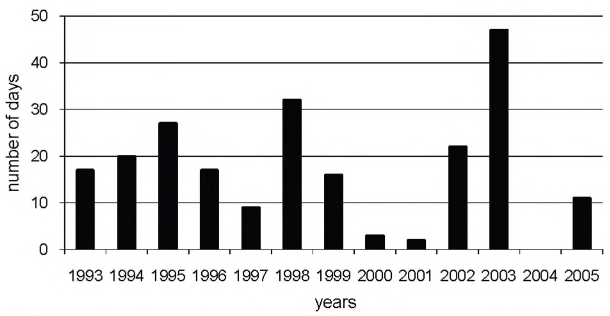

Fig. 12. Annual number of days when 8-hour mean concentration of O_3 exceeded 120 µg·m^{-3} in measurement points in Kraków in 1993-2005.

the most common cause of death (50%), while cancer was responsible for 26% mortality in women and 29% in men. Diseases of the respiratory system caused 3% of all deaths [41].

The health effects of particulate matter are of special significance in new EU member states. Pollution sources which are of medium importance in old EU states, may be of high relevance in the new ones. For instance, Poland alone consumes more coal in the residential sector than the whole EU-15 [34]. As shown above in section "Particulate Matter," PM10 concentrations are the most severe air pollution problem in Kraków at present, likely to increase further. Kraków was one of the cities included in recent international studies about the health effects of PM10. The results of the APHEIS project proved that in cases of short-term exposure, daily means of PM10 below 20 µg·m^{-3} would allow avoid 50 premature deaths including 38 deaths from cardiovascular diseases and a reduction of the long-term air pollution of PM2.5 to a level of 15 µg·m^{-3} would reduce mortality by 636 deaths in one year, about 10% of the annual burden of mortality in Kraków. The ENHIS-1 project results showed that the reduction of the annual average levels of PM10 to 20 µg·m^{-3} would prevent one postneonatal death [41]. The APHEA project was realized in 1991-94 and together with the studies from 1995-99 showed that the increase of PM10 concentration by 50% during 24 hours caused an increase of death risk by 2% on average and 4% in the part of the population 65 years old and more. A similar effect was observed for SO_2 [37, 42]. Within the project APHEA2, realized in 1994-98, the data from Kraków from the period 1990-96 were analyzed. An increase of 10 µg·m^{-3} in daily PM10 increased the daily number of deaths by 0.2% for all ages. The relation was found dependent on NO_2 concentration and climatic zone [43]. In 2004-06, Kraków Integrated Project was realized to investigate indoor and outdoor air pollution impact on health in Kraków. Particles with aerodynamic diameter <2.5 µm (PM2.5) accounted for more than 90% of PM10 mass and the lower the diameter the larger the possible health risk, as already proved in the APHEIS project mentioned above. Long-term exposure to coal-related indoor air pollution is associated with higher prevalence of respiratory symptoms, especially of chronic cough and chronic bronchitis as well as lung functioning. Point estimates for effects from coal-related indoor exposure did not change much when outdoor PM10 level was added to the statistical model [34].

Plants and Animals

Mosses and lichens are the bioindicators used most often to assess air pollution by heavy metals in forests, while vascular plants are used only rarely. In the Niepołomice Forest near Kraków, *Pleurozium schreberi* was used, together with a vascular plant *Moehringia trinervia* (only the aboveground parts were subject to analysis). In both cases, samples were collected and analyzed in the times of huge air pollution emissions, and then again when emissions were significantly lower (*Pleurozium schreberi*: 1975 and 1998, *Moehringia trinervia*: 1984 and 1999). Concentrations of Cd, Pb, Fe, Zn and Cu were determined for *Pleurozium schreberi*, while for *Moehringia trinervia* Cd, Pb, Zn and Cu. The results of the comparison prove that in the moss collected in 1998, the amounts of heavy metals were significantly lower than in 1975. The content of Fe decreased by 52%, Zn by 78%, Cu by 44%, Pb by 86% and Cd by 71%. A similar trend occurred for *Moehringia trinervia* although the changes were much less spectacular; Cd decreased by 32%, Pb by 22%, Zn by 10%, Cu by 22%. As the Niepołomice Forest covers the area of 11,000 ha, the spatial distribution of heavy metal pollution was studied, too. Again, in the case of mosses and lichens the changes are much better pronounced than in the case of *Moehringia trinervia*. The inventories of lichens were made in the Niepołomice Forest in 1960s, 1970s and in 1999-2000. The last one showed the return of several sensitive species of lichens (e.g. *Imshaugia aleurites*) to areas from which they had disappeared. In the case of *Moehringia trinervia*, the spatial pattern of contamination was similar in both study periods, with the highest values in western and northern parts of the forest, closest to Kraków. So the significant reduction in emissions and imission of dust containing heavy metals is well reflected in their concentrations in mosses, but the concentrations in *Moehringia trinervia* do not show such a clear effect. It can be explained by the fact that the Niepołomice Forest receives air pollution from many sources, both distant (Kraków, Silesia Region) and local ones (roads, gas combustion during the exploitation of small rock-oil sites). Another factor may be the recent increase in the acidity of atmospheric precipitation in the discussed area, which activates heavy metals deposited in the soil earlier and allows them to be taken by plants. Even though both the emissions and the concentrations of heavy metals decreased during the last 15 years significantly, the contamination of plants is still above normal levels. In the case of *Pleurozium schreberi*, the comparison of the Niepołomice Forest with the Białowieża National Park (a reference area) shows that the content of Pb, Zn and Cu is twice as high in Niepołomice as in Białowieża, and Cd seven times as high. For *Moehringia trinervia*, the present concentrations of Cd and Zn exceed the values considered as natural for plants, while Cu and Pb do not [44, 45].

In 1998-2001, seven small forest ecosystems in the vicinity of Kraków (up to 35 km from the city centre) were the subject of studies concerning the input of heavy metals to the forest floor. The input was determined by the concentration of these metals in the litter fall, the litter's yearly dynamics and species composition. The dominating species were sycamore, hornbeam and oak, from which sycamore accumulated the highest concentration of metals and oak the lowest. The heavy metals input tended to decrease with increasing distance from Kraków. The annual input of cadmium ranged from 84 to 382 $\mu g \cdot m^{-2}$, lead: 406 to 3717 $\mu g \cdot m^{-2}$, copper: 778 to 2768 $\mu g \cdot m^{-2}$, zinc: 6 to 38 $mg \cdot m^{-2}$, iron: 22 to 263 $mg \cdot m^{-2}$. In spite of a significant decrease in air pollution in Kraków after 1990, the heavy metal input to the forest floor remained the same and in the case of some elements became even higher than in earlier periods [46, 47].

Since 1982, heavy metals content in soils and vegetables grown in Kraków has been monitored. According to Polish legislation, the content of heavy metals in vegetables' dry mass should not exceed 0.4 $mg \cdot kg^{-1}$ for Cd, 2.5 $mg \cdot kg^{-1}$ for Pb and 50 $mg \cdot kg^{-1}$ for Zn. In the 1980s, the soils contained more Cd and Pb than the allowed limit values, while at the beginning of the 1990s the content of all heavy metals was below the allowed limits. However, the vegetables, especially the above-ground parts, still contained too much cadmium, lead and zinc. In the mid-1990s a further decrease in heavy metals' contamination was observed. In the 1980s, the content of Cd in soils was 6 times higher, and that of Cr and Ni 3 times higher than in 1993-95. But still the cadmium concentration was above the norm in some places (0.61-1.34 $mg \cdot kg^{-1}$). The measured amounts of Cd, Pb and Zn were above the allowed limits in vegetables. Results from 2005 proved that the decreasing tendency in vegetables' and soil pollution with heavy metals has been unfortunately reversed in some cases. Mean total concentration of cadmium in soil increased from 1.08 in 1994 to 2.46 in 2005. The Cd content in vegetables reached the values from mid-1980s. Lead concentration in soil has not changed much since the mid-1990s, but it increased in vegetables, although not so dramatically like Cd. Similar tendencies occur for Cu, Fe and Zn. Areas most contaminated with Cd are located in the western part of Kraków, receiving air pollution from both the Upper Silesia region and Kraków industry. Lead is most abundant in the city centre and eastern part of the area, Cu in the southern part and Zn in the southern and western parts, while Fe in the southern and eastern part [48-50].

The study of *Hypogymnia physodes* lichen transplanted from a control area to a few points in Kraków in 2004 and 2005 proved significant contamination with SO_2 as the lichen accumulated the sulphur, especially in winter when the SO_2 concentration in air is the highest. Mean value for winter was about 2000 μg of S per g of dry mass, while in summer it was about 1000 μg of S per g of dry mass [51].

In the years 1985-2002, bank voles (*Clethrionomys glareolus*, Schreber, 1780) from the Niepołomice Forest and Kraków were used as bioindicators of environmental contamination by heavy metals. The accumulation of Cd, Pb, Zn and Fe in animal tissues (liver and kidneys) was determined. Some of the results obtained are shown in Table 3.

Zinc and iron are indispensable to ensure the normal development and function of the organism, while lead and cadmium are toxic elements. Even a relatively low body burden of lead and cadmium can cause histopathological changes in the tissues of rodents permanently exposed to heavy metals [53]. As shown in Table 3, the concentration of lead and zinc showed the decreasing tendency, while cadmium either gradually increased and then decreased (like in kidneys) or first decreased but then increased again (like in livers). The study of Topolska et al. [52] proved that in the Kraków area, the southern zone is the one most contaminated with cadmium, while the eastern one with iron and zinc. The levels of Fe were estimated as non-toxic.

In the years 1990-93 the breeding performance of pied and collared flycatcher populations (*Ficedula hypoleuca* and *Ficedula albicollis*) from the Niepołomice Forest was studied. The concentrations of cadmium (up to 0.35 $\mu g \cdot g^{-1}$ in liver) and lead (up to 1.09 $\mu g \cdot g^{-1}$ in breastbone samples) were higher than in the control area, while the levels of copper, zinc and iron were close to physiological norms [54].

Discussion

After the Second World War, air pollution in Kraków reached huge values due to both large industrial emissions and the location in natural conditions which are very unfavourable for pollution dispersion. After 1989, the aerosanitary conditions were improved significantly compared to earlier periods, but the present tendencies differ for particular pollutants. In spite of increasing traffic, tropospheric ozone is not a relevant air pollution issue in the city, although its levels may be high in the nearby rural areas but there is no measurement network to verify that. NO_2 exceeds the allowed values permanently, but only in the high traffic zones, still it is not transferred into equally often occurrence of high values of O_3. Both NO_2 and O_3 have been measured only since 1992 and both show no clear trends in concentrations in the period 1992-2006. Much more important from the environmental point of view are the changes of SO_2 and PM10 concentrations. The large decrease in their concentrations observed after 1989 has been recently stopped in the case of SO_2 and even reversed in the case of PM10. In both cases winter concentrations are much higher than summer ones, which is the effect of extensive coal usage for heat production. However, SO_2 concentrations exceed the allowed limits only occasionally, while the level of PM10 is permanently too high all winter long. Additionally, transportation recently has become a more important source of particulate matter.

The observed changes in air pollution are transferred into the environmental effects in a complex way. The PM10 emitted in Kraków comes mainly from the steelworks, power plants and private heating facilities operating on coal, but the city receives also large quantities of PM10 from the Upper Silesia industrial region, where many nonferrous smelters and other industrial plants are located. Most particulate matter contain heavy metals, which contaminate soil, affect plants and animals. Soils and plants showed a fast and direct response to the changes in air pollution. As PM10 concentrations were decreasing by the end of the 1990s, the heavy metals content in soils and plants was decreasing, too. Unfortunately, during last few years the opposite tendency occurs. In soils, cadmium content increased dramatically, while the concentration of other heavy metals remained on an almost unchanged level. The sources of Cd are heavy traffic, local industry and coal burning [34]. Cadmium is also delivered to Kraków mainly with particulate matter from the Upper Silesia region transported with prevailing western winds. In the case of plants, increased heavy metals content was observed in vegetables and the input to the forest floor also increased in some cases. Studies on animals do not cover the last few years, but the results obtained up to 2000 show that especially the high concentrations of cadmium in soft tissues are still a serious problem. At present, the heavy metals' content in PM10 in Kraków is below the allowed levels, but the PM10 concentration is increasing every year, which means that the mass of PM10 and also heavy metals entering the area is increasing, too.

According to the CESAR project, realized in the years 1995-96, Poland and Bulgaria were the countries most polluted with PM10 and PM2.5 in Europe, especially in winter [57]. The results of particulate matter measurements in Europe in 2000 additionally proved those findings [60].

Conclusions

In spite of the significant decrease of air pollution in Kraków after 1990, the natural environment of the city and its surroundings is still endangered by that factor, especially by PM10. Industry is the main source of particulate matter emission, but at present also transportation is gaining an increasing role. The PM10 concentrations are still so high that they cause chronic respiratory system diseases, especially in children. The increasing tendency of PM10 concentrations combined with the prevalence of PM2.5 in suspended particulate matter may intensify that problem in the future. In the case of Kraków, the very high concentrations of PM10 are the result of very high emissions but also the city's location in a concave landform which significantly worsens the natural ventilation conditions. The most important environmental secondary effect of air pollution in Kraków is the contamination of soils, plants and animals with heavy metals. The heavy metal burden decreased enormously parallel to air pollution after 1990, but the concentration of cadmium, which is a toxic element, is still too high and likely to increase further. The recently observed changes in air pollution and its environmental impact are the effect of increased traffic in Kraków and emissions from industry, especially energy production. That also includes the local and private heating facilities. Since 1995, a programme of decreasing low-sources' emissions has been realized in Kraków. Until the end of 2007, 18,000 coal stoves and 249 local heating facilities operating on coal have been turned into gas installations. Additionally, 22 renewable energy installations, mainly solar panels, have been established. However, still 12% of flats or houses in the city are heated with coal [61]. Therefore, from the environmental point of view, exchanging the coal-based heat production into other, cleaner alternatives, together with introducing techniques for further decrease of particulate matter emission from industry, seems to be of key importance for the future sustainable development of Kraków.

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