



Article The Impact of Local Anti-Smog Resolution in Cracow (Poland) on the Concentrations of PM10 and BaP Based on the Results of Measurements of the State Environmental Monitoring

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Abstract: As a result of conducted air quality policy, including recent legal regulations (the local antismog resolution), the number of individual solid fuel heating devices in Cracow (Poland) gradually decreased. Reports on air quality in the city indicate that the concentration of pollutants in Cracow's air shows a downward trend. However, a similar tendency in terms of improving air quality is also observed in the entire voivodeship, where, as a result of analogous although less radical measures, the number of individual solid fuel heating devices is also decreasing. The paper discusses the impact of legal regulations in Cracow on the improvement of air quality in the context of changes taking place in nearby cities. Trends in changes in PM10 and BaP (PM10) concentrations are analyzed. The rate of decline of the analyzed pollutants concentrations is estimated with the use of nonparametric linear regression. Analysis showed that the rate of decline in the average annual concentrations of PM10 and BaP (PM10) in Cracow is always higher than for the analyzed cities of the Malopolskie Voivodeship. The difference is more pronounced with regard to the months of the heating season. The rate of changes for the average annual BaP (PM10) concentrations in Cracow, compared to other analyzed cities of the Malopolskie Voivodeship, is more intensive than in the case of PM10 concentrations (1.5 times stronger with regard to the months of the heating season). Since the concentration of BaP (PM10) is a better indicator of the effects of liquidation of high-emission furnaces than the concentration of PM10, it can be concluded that the impact of actions related to the improvement of air quality in Cracow in the context of changes taking place in selected cities of the Malopolskie Voivodeship is more visible.

Keywords: air quality; Cracow; PM10 concentration; BaP concentration; environmental monitoring; anti-smog resolution impact; low emissions

1. Introduction

Air pollution is currently perceived as one of the biggest environmental concerns, next to climate change. The latest World Health Organization (WHO) data on ambient air pollution show a serious problem of air quality faced all over the world [1]. Almost all of the global population (99%) breathe air that exceeds WHO guideline limits, containing high levels of pollutants [2]. Highly urbanized areas are places that suffer in particular from poor air quality. This fact confirms also the European Environment Agency's Report on "Air quality in Europe—2020" [3]. According to this report, concentrations of ambient particulate matter (PM) (which is currently considered to be the best indicator for health effects of ambient air pollution) in the analyzed year continued to exceed the European Union limit values and the more stringent WHO Air Quality Guidelines (WHO AQG) in large parts of Europe. Despite reductions in emissions and ambient concentrations, air quality in Europe remains poor in many areas. This issue also concerns many towns, cities and villages in Poland, especially located in valleys and concave landforms. (e.g., Cracow, referred to in the paper).



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). An important fraction of the exposure to ambient air pollution occurs in cities due to the higher density of human activities and their emissions to the air [4]. Deterioration of air quality is caused not only by the emissions—related to industry, traffic (communication) or the housing sector (domestic fuel combustion for heating purposes)—but also by the blocking of natural ventilation channels, which worsens the dispersion conditions of pollutants already released to the atmosphere. The constantly increasing number of high-rise buildings in cities changes the city wind environment [5]. Urban wind speed shows a downward trend, which aggravates the diffusion of air pollution (also the diffusion of heat, intensifying the phenomenon of urban heat island (UHI)) [5–7].

Poland (Figure 1) is one of the European countries struggling with poor air quality. In the heating season, high concentrations of suspended particulate matter (PM10 and PM2.5), benzo(a)pyrene (BaP), NO_x, SO₂ and other pollutants are recorded in the air. It is widely recognized that the main reason for the poor air quality in Poland is low emissions. The most important problem, in terms of low emissions, is the burning of low-quality coal in local, energy-inefficient heating devices for space heating and domestic hot water production [8]. The percentage share of individual heating devices in the emissions of PM10 and PM2.5 is up to 50% and in the case of benzo(a)pyrene (BaP), it is up to 90% [9].



Figure 1. Location of the study area: Poland, Cracow (Source: own elaboration).

To improve air quality in Poland, various measures are taken at the local and central levels. An example of intensive actions in this field is Cracow, where apart from the implementation of the provisions of the anti-smog resolution for the Malopolskie Voivodeship [10], more radical, local anti-smog resolution for Cracow [11] was adopted. According to this local resolution, since 1 September 2019, it has been prohibited to use coal and wood in boilers, stoves and fireplaces in Cracow (only the use of gaseous fuels or light fuel oil in individual fuel combustion installations is allowed).

Many authors report the effect of air quality control efforts, including PM level changes in urban air [12–14] or going further, describe the consequent health benefits from particulate matter concentrations decreasing after policy implementation [15,16]. The problem of analyzing different policy tools is often raised [17]. Authors describe also the effectiveness of programs supporting the exchange of heating devices [18,19] or long-term trends of air pollutants concentration [20,21]. The paper discusses trends in changes in PM10 and BaP concentrations in the context of a more radical—compared to the rest of the voivodeship—air quality policy in Cracow, including recent legal regulations (the local anti-smog resolution).

2. Study Area

Poland is one of the largest states in Europe (an area of 312.7 thousand km², with a population of 37.7 million). It borders Germany, Russia, Lithuania, Belarus, Ukraine, Czech Republic and Slovakia (Figure 1). From the north, it is washed by the Baltic Sea. The

administrative division of Poland includes 16 voivodeships, one of which is Malopolskie Voivodeship, with Cracow as its biggest city and also the capital of the voivodeship.

Cracow (50°3'41" N, 19°56'18" E) is the second-largest city in Poland, both in terms of population (779,966 people, data from 2020) and area (327 km²) [22]. Until 1795, the city was the formal capital of Poland. It is located in southern Poland (Figure 1), in the central-western part of the Malopolskie Voivodeship. The extent of the city from south to north is 18 km and 31 km from west to east. Cracow, as one of the few large cities in Poland, develops intensively demographically. The city acts as an administrative, cultural, educational, scientific, economic, service and tourist center. After Warsaw (the capital of Poland), Cracow is the second-largest market for modern office space. The city is also the key road and railway hub in Poland.

Cracow is located in a concave landform (Figure 2) at the interface of four geographical regions. The historical city center is placed in the Vistula river valley bottom (at about 200 m a.s.l.), going from west to east [23]. The unique geographical location of Cracow and the associated city climate with the predominance of weak western winds, frequent occurrence of calm weather and temperature inversions cause poor ventilation of the city, which worsens the conditions of the natural environment suffering from emissions related to the housing sector (individual combustion), traffic pollution and industry. An additional factor acting against the air quality in the city is the proximity of the Upper Silesian Industrial District (the main center of coal mining and metallurgy in Poland), and thus the transport of polluted air, gases and dust.



Figure 2. Hipsometric map of Cracow (Source: own elaboration based on https://obserwatorium. um.krakow.pl, accessed on 10 May 2020).

The climatic conditions prevailing in Poland cause that the heating season lasts for more than half of a year, most often from October to April. It slightly differs for different localizations as climatic conditions differ. Table 1 presents the number of heating days per every month, set for Cracow, according to Polish regulations.

Table 1. Number of heating days per month in Cracow.

Month	Ι	II	III	IV	V	VI	VII	VIII	IX	x	XI	XII
Number of days	31	28	31	30	5	0	0	0	5	31	30	31

Generally, thermal conditions in Cracow force intensive heating of buildings up to 60% of winter days, which affects the air quality. During the warm season, episodes of increased air pollution (especially with the ozone) are also observed in the city. It happens usually when the air temperature exceeds 25 °C, which promotes the formation of so-called photochemical smog (caused by emissions mainly related to traffic).

Dangerous (due to aerosanitary conditions) weather situations constitute in Cracow, in total, about 1/4 days a year, and their culmination falls in autumn. In the spring, unfavorable weather conditions (worsening the dispersion of air pollutants) prevail for as much as 65% of days, in summer they constitute 50%, in autumn 72% and winter 30%.

Generally, throughout the year, 54% of the days in Cracow have one or several features of weather that impede the dispersion of air pollutants [24]. Problems with the natural ventilation of the city translate into poor air quality.

Cracow city authorities, aware of problems concerning air quality, make continuous efforts to fight low emissions. A radical reduction of emissions from solid fuel combustion was a necessary condition in Cracow to gradually improve air quality. Since 1995, the Low Emission Reduction Program has been implemented in Cracow, allowing the replacement of coal stoves and local heating facilities operating on coal (usually with gas or district heating). According to the data from the City Hall, 45,649 individual solid fuel heating devices (boilers and stoves) have been closed down since 1995, of which more than half (25,793) closed down in 2012–2019. The year 2017 was record-breaking in this respect when over 6000 furnaces were replaced. The city assumed liquidation of all the remaining furnaces operating on coal until September 2019, as according to the adopted resolution of the Provincial Assembly [11], since 1 September 2019 it has been prohibited to use coal and wood in boilers, stoves and fireplaces in Cracow (only the use of gaseous fuels or light fuel oil in individual fuel combustion installations is allowed). Currently, according to the estimates of the City Hall, around 2000 coal boilers remain in the city. Each of these cases is considered individually due to the variety of problems concerning their replacement.

As a result of conducted air quality policy, including recent legal regulations (the local anti-smog resolution for Cracow), the number of individual solid fuel heating devices in Cracow gradually decreased. Reports on air quality in the city indicate that the concentration of pollutants in Cracow's air shows a downward trend [25]. However, a similar tendency in terms of improving air quality is also observed in the entire Malopolskie Voivodeship, where, as a result of analogous, although less radical measures, the number of individual solid fuel heating devices (especially classless ones) is also decreasing.

The paper discusses the impact of actions implemented to eliminate low emissions in Cracow in the context of changes taking place in the Malopolskie Voivodeship. The comparative method was used for the analysis. To assess trends in changes in PM10 and BaP (PM10) concentrations in Cracow, five cities from the Malopolskie Voivodeship (Figure 3) were selected for the analysis: 1—Bochnia, 2—Tarnow, 3—Gorlice, 4—Nowy Sacz and 5—Zakopane. There are urban background measurement stations operating for a sufficiently long time (minimum from 2011 to 2020) to observe trends in changes in PM10 and BaP (PM10) concentrations.



Figure 3. Malopolskie Voivodeship. Cracow and analyzed cities: 1—Bochnia, 2—Tarnow, 3—Gorlice, 4—Nowy Sacz, 5—Zakopane (Source: own elaboration based on https://www.google.com/maps/, accessed on 10 November 2021).

3. Methodology

The aim of the analysis was to investigate whether the more intensive removal of coal-fired heating devices (boilers, stoves) in Cracow, compared to other analyzed cities of the Malopolskie Voivodeship, resulted in a faster (more visible) improvement of air quality in the city, in terms of the analyzed indicators.

The adopted indicators of air quality changes were the concentration of PM10 dust and the content of benzo(a)pyrene in PM10 dust (BaP (PM10), BaP in PM10). The dominant source of BaP in the air in Poland are fuel combustion processes carried out outside industry and transport, including, first of all, individual heating of buildings (fuel combustion in small heating installations). They are responsible (according to the latest data for 2019) for about 91% of the total PAH emissions (polycyclic aromatic hydrocarbons), which include benzo(a)pyrene (BaP). Household emissions account for the major share of emissions in this category (around 89%) [26,27]. Taking this into account, BaP (PM10) concentrations are the best indicator of the effects of liquidation of high-emission furnaces. Combustion processes outside industry, and thus mainly low emissions, are also largely responsible for the emission of PM10 dust. According to the data of the National Center for Emissions Management [28], the main source of dust emissions into the atmosphere in Poland are fuel combustion processes outside industry and transport. In 2019 (for which data are available), the emissions from these processes accounted for approximately 41% of the total PM10 dust emissions from the country [26,27]. Emissions from the municipal and housing sector, including those related to heating buildings, have the highest share in this category. Therefore, next to BaP (PM10), the PM10 concentration was taken into account as the second-best indicator of the effects of liquidation of high-emission furnaces.

The analysis used verified measurement data available in the database of the Chief Inspectorate for Environmental Protection [29]. These data were collected as part of the State Environmental Monitoring, which is a system for measuring, assessing and forecasting the state of the environment, as well as collecting, processing and disseminating information about the environment. This database contains current measurement data as well as archived measurement data that have been validated based on appropriate procedures. Free viewing and downloading of measurement data from the database is available through the search engine in the Measurement Data Bank on the "Air Quality" Portal. Air quality measurements carried out as a part of the State Environmental Monitoring are performed in accordance with the regulations of the European Union (Directive 2008/50/EC on ambient air quality and cleaner air for Europe) and national regulation (in Poland it is The Environmental Protection Law and the Ordinance of the Minister of the Environment on the assessment of the levels of substances in the air). These regulations allow air quality to be assessed in the EU Member States on the basis of common methods and criteria (including criteria for the location of sampling points, reference measurement methods, air quality assessment procedures or criteria for checking the correctness of data during their aggregation and calculation of statistical parameters).

The analysis used data from urban background measurement stations located in Cracow and selected cities of the Malopolskie Voivodeship (Figure 3, Table 2), operating for a sufficiently long time (minimum from 2011 to 2020) to observe trends in changes in PM10 and BaP (PM10) concentrations.

Nr	City	International Station Code	Station Code	Station Address	WGS84 φ N	WGS84 λ E
1	Cracow	PL0501A	MpKrakBujaka	Kraków, ul. Bujaka	50.010575	19.949189
2	Bochnia	PL0549A	MpBochKonfed	Bochnia, ul. Konfederatów Barskich	49.969017	20.439511
3	Tarnow	PL0502A	MpTarBitStud	Tarnów, ul. Bitwy pod Studziankami	50.020169	21.004167
4	Gorlice	PL0478A	MpGorlKrasin	Gorlice, ul. Krasińskiego	49.658889	21.163336
5	Nowy Sacz	PL0550A	MpNoSaczNadb	Nowy Sącz, ul. Nadbrzeżna	49.619281	20.714403
6	Zakopane	PL0126A	MpZakopaSien	Zakopane, ul. Sienkiewicza	49.293564	19.960083

Table 2. Analyzed urban background measurement stations operating in Cracow and selected cities.

As the impact of individual heating and activities in this area were analyzed, data from industrial and communication stations were not considered. Urban background measurement stations in Cracow and selected cities of the Malopolskie Voivodeship that started operating after 2011 were also not included. The starting point for the analysis was 2011, because according to data, more than half (25,793) of individual solid fuel heating devices (boilers, stoves), which had been closed since 1995, were closed in 2012–2019.

The list of the analyzed urban background measurement stations, along with the codes of the stations and other details, is presented in Table 2.

The analysis was performed for full calendar years between 2011–2020. The average annual and monthly concentrations (based on the 24-h averaging period) of selected pollutants in various locations of the Malopolskie Voivodeship, including Cracow, were compared. The data for Cracow were compared consecutively with the data for each analyzed city of the Malopolskie Voivodeship and with the average value for the analyzed cities of the Malopolskie Voivodeship (except Cracow) calculated as the arithmetic mean. This comparative method was applied to the average annual and monthly concentrations of selected pollutants. The rate of decline of the analyzed pollutants concentrations was investigated.

In order to emphasize the impact of individual heating of buildings, the annual and monthly averages were also analyzed in relation to data only for the months of the heating season, which, considering the climatic conditions prevailing in Poland, lasts for more than half of a year, most often from October to April.

The nonparametric Mann–Kendall test was used to determine trends in the mean annual concentrations of PM10 and BaP (PM10) in the analyzed cities of the Malopolskie Voivodeship.

The statistics of the Mann–Kendall test for monotonic trend were calculated according to [30–32]:

$$S = \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} \operatorname{sgn}(x_j - x_i),$$
(1)

where:

 $sgn(x) = \begin{cases} 1 & gdy \ x > 0 \\ 0 & gdy \ x = 0 \\ -1 & gdy \ x < 0 \end{cases}$ (2)

When H0: S = 0 is true, the mean is E(S) = 0 and variance of S (including the correction term forties) are given by:

$$\operatorname{var}(S) = \frac{n(n-1)(2n+5)}{18} - \frac{\sum_{i=1}^{k} t_i \cdot i(i-1)(2i+5)}{18},$$
(3)

where k is the number of the tied groups in the data set and t_i is the number of data points in the *i*-th tied group. The statistic *S* is approximately normally distributed, with

$$Z = S/\sigma \tag{4}$$

The magnitude of trends was determined using Theil–Sen's slope [33,34]. The non-parametric fitted line was defined as [34]:

$$y = a_{TS}x + b, (5)$$

where a_{TS} is the Theil–Sen slope:

$$a_{TS} = median \frac{y_j - y_i}{x_j - x_i},\tag{6}$$

for all i < j, i = 1, 2, ..., (n-1), j = 2, 3, ..., n, and b is the intercept defined as:

$$b = y_{med} - a_{TS} \cdot x_{med}, \tag{7}$$

where x_{med} and y_{med} are the medians of x and y, respectively.

Regression lines in figures were shown for comparison.

The Mann–Kendall test should be used when no seasonality is observed in data. The occurrence of seasonality means that the data have different distributions for different months of the year. In this study, the monthly averages of both PM10 and BaP (PM10) concentrations are characterized by high seasonality; therefore, the seasonal Mann–Kendall test was used to detect trends in these variables. The seasonal Mann–Kendall test is an extension of the Mann–Kendall test and is described in [35]. When the variable changes seasonally, the seasonal Sen's slope is used to determine the magnitude of trend (linear rate of change). According to [35] the seasonal Sen's slope (Kendall slope) was calculated as:

$$a_{S} = median \frac{x_{ij} - x_{ik}}{j - k},$$
(8)

for all (x_{ij}, x_{ik}) pair i = 1, ..., m, where $i \le k < j \le n_i$, and n_i is the number of known values in the *i*-th season. The seasonal slope estimator is the median of the a_s values.

All statistical calculations were performed using the GNU R software package [36]. For all the tests considered in the paper, the significance level was fixed at 0.05.

4. Results

This chapter presents the results of the analysis of PM10 and BaP in PM10 (BaP (PM10)) concentrations data, measured in 2011–2020 at the urban background measurement stations located in Cracow and selected cities of the Malopolskie Voivodeship. Such an approach allowed for investigating the rate of decline of the analyzed pollutants in Cracow in comparison to other locations of the Malopolskie Voivodeship.

The time series of average annual concentrations of PM10 and BaP (PM10) in Cracow and the analyzed cities of the Malopolskie Voivodeship (Tarnow, Bochnia, Gorlice, Zakopane and Nowy Sacz) were subjected to the Mann–Kendall test, and the results are presented in Figures 4 and 5. Additionally, the variability of the mean value for the analyzed cities of the Malopolskie Voivodeship (excluding Cracow)—calculated as the arithmetic mean of the average annual concentrations of PM10 and BaP (PM10), respectively, in analyzed cities of the Malopolska Voivodeship (excluding Cracow)—was analyzed and presented in Figures 4 and 5.

In order to emphasize the impact of individual heating of buildings, the annual averages were also analyzed in relation to data only for the months of the heating season, which, considering the climatic conditions prevailing in Poland, lasts for more than half of a year, most often from October to April. The time series of average annual concentrations of analyzed pollutants in relation to data only for the months of the heating season (January, February, March, April, October, November and December), and the results of the Mann-Kendall test are presented in Figures 6 and 7.

In Figures 4–7, in addition to the nonparametric line (based on the Theil–Sen coefficient), linear regression is also presented. The aforementioned lines have a similar slope. The regression lines slopes are statistically significant at the significance level of $\alpha = 0.05$.



Figure 4. Comparison of the distributions of average annual concentrations of PM10 in Cracow and the analyzed cities of the Malopolskie Voivodeship in 2011–2020. Significant ($p_{v,MK} < 0.05$) decreasing trends shown using Theil–Sen line with significant slope a_{TS} [µg/m³/year] ($p_{v,TS} < 0.05$). Linear regressions with significant a_{LR} [µg/m³/year] ($p_{v,LR} < 0.05$) are shown for comparison. Malopolskie means the arithmetic mean of the values for individual analyzed cities, excluding Cracow. (Source: own elaboration based on the data from the database of the Chief Inspectorate for Environmental Protection [29]).



Figure 5. Comparison of the distributions of average annual concentrations of BaP (PM10) in Cracow and the analyzed cities of the Malopolskie Voivodeship in 2011–2020. Significant ($p_{v,MK} < 0.05$) decreasing trends shown using Theil–Sen line with significant slope a_{TS} [ng/m³/year] ($p_{v,TS} < 0.05$). Linear regressions with significant a_{LR} [µg/m³/year] ($p_{v,LR} < 0.05$) are shown for comparison. Malopolskie means the arithmetic mean of the values for individual analyzed cities, excluding Cracow. (Source: own elaboration based on the data from the database of the Chief Inspectorate for Environmental Protection [29]).



Figure 6. Comparison of the distributions of average annual concentrations of PM10_{hs} for the heating season in Cracow and the analyzed cities of the Malopolskie Voivodeship in 2011–2020. Significant ($p_{v,MK} < 0.05$) decreasing trends shown using Theil–Sen line with significant slope a_{TS} [µg/m³/year] ($p_{v,TS} < 0.05$). Linear regressions with significant a_{LR} [µg/m³/year] ($p_{v,LR} < 0.05$) are shown for comparison. Malopolskie means the arithmetic mean of the values for individual analyzed cities, excluding Cracow. (Source: own elaboration based on the data from the database of the Chief Inspectorate for Environmental Protection [29]).



Figure 7. Comparison of the distributions of average annual concentrations of BaP_{hs} (PM10) for the heating season in Cracow and the analyzed cities of the Malopolskie Voivodeship in 2011–2020. Significant ($p_{v,MK} < 0.05$) decreasing trends shown using Theil–Sen line with significant slope a_{TS} [ng/m³/year] ($p_{v,TS} < 0.05$). Linear regressions with significant a_{LR} [µg/m³/year] ($p_{v,LR} < 0.05$) are shown for comparison. Malopolskie means the arithmetic mean of the values for individual analyzed cities, excluding Cracow. (Source: own elaboration based on the data from the database of the Chief Inspectorate for Environmental Protection [29]).

Trends in average annual concentrations of PM10 and PM10_{hs} (heating season) in 2011–2020 are presented in Table 3. Trends in average annual concentrations of BaP (PM10) and BaP_{hs} (PM10) (heating season) presents Table 4. Out of the analyzed cases, only in one (Bochnia, BaP (PM10), Table 4) did the Mann–Kendall test not show a trend at the significance level of α = 0.05. In other cases, there are grounds to reject the hypothesis of no trend at the assumed significance level and the $p_{v,MK}$ values of the Mann Kendall test are less than 5%. In most cases, the trends are strong ($p_{v,MK}$ is even lower than 1%) or medium ($p_{v,MK} < 5\%$) (Table 3). Only in Nowy Sacz, for BaP (PM10), the *p*-value of the test is at the borderline of significance ($p_{v,MK}$ is 0.0494). In relation to data only for the months of the heating season (BaP_{hs} (PM10)) the *p*-value of the test for Nowy Sacz is better ($p_{v,MK}$ is 0.0491).

Table 3. Trends in average annual concentrations of PM10 and $PM10_{hs}$ (heating season) in 2011–2020 obtained using the Mann–Kendall (MK) test and the Theil–Sen slope estimator.

NT	City		All Year (PN	A10)	Heating Season (PM10 _{hs})			
Nr	City	Z_{MK}	Significance	a _{TS} [μg/m³/year]	Z_{MK}	Significance	a _{TS} [μg/m ³ /year]	
1	Cracow	-3.04	**	-2.26	-3.22	**	-4.05	
2	Tarnow	-2.24	*	-1.27	-2.86	**	-2.03	
3	Bochnia	-2.78	*	-1.42	-3.04	**	-1.91	
4	Gorlice	-3.22	**	-1.30	-3.22	**	-2.36	
5	Zakopane	-3.66	**	-2.16	-3.58	**	-3.43	
6	Nowy Sacz	-3.04	**	-2.18	-3.22	**	-3.38	
7	Malopolskie Voivodeship	-3.40	**	-1.69	-3.39	**	-2.43	

* if *p*-value $p_{MK} < 0.05$ (medium evidence), ** if *p*-value $p_{MK} < 0.005$ (strong evidence).

NT.	City		All Year (BaP (PM10))	Heating Season (BaP _{hs} (PM10))			
INT	City	Z_{MK}	Significance	a _{TS} [ng/m³/year]	Z_{MK}	Significance	a _{TS} [ng/m ³ /year]	
1	Cracow	-3.79	**	-0.62	-3.58	**	-1.30	
2	Tarnow	-3.06	**	-0.23	-2.68	*	-0.51	
3	Bochnia	-1.71		-0.30	-2.15	*	-0.59	
4	Gorlice	-2.24	*	-0.10	-2.25	*	-0.18	
5	Zakopane	-2.25	*	-0.43	-3.04	**	-1.05	
6	Nowy Sacz	1.89	*	-0.30	-1.98	*	-0.41	
7	Malopolskie Voivodeship	-2.62	*	-0.27	-2.86	**	-0.52	

Table 4. Trends in average annual concentrations of BaP (PM10) and BaP_{hs} (PM10) (heating season) in 2011–2020 obtained using the Mann–Kendall (MK) test and the Theil–Sen slope estimator.

* if *p*-value $p_{MK} < 0.05$ (medium evidence), ** if *p*-value $p_{MK} < 0.005$ (strong evidence).

To determine the magnitude of the confirmed trends, the nonparametric Theil–Sen slope values were calculated. All the Theil–Sen slope a_{TS} are statistically significant at the significance level of $\alpha = 0.05$ (Figures 4–7 and Tables 3 and 4). The Theil–Sen slope a_{TS} in the case of PM10 concentrations is the highest in Cracow ($a_S = -2.26$), but in Nowy Sacz as well as in Zakopane, it is not much lower than in Cracow (Table 3). The comparison of the Theil–Sen slopes values for Cracow and on average for the analyzed cities of the Malopolskie Voivodeship (excluding Cracow) ($a_S = -1.69$) shows that the Theil–Sen slope a_{TS} is higher in Cracow by approximately 25% (Table 3). Compared to Nowy Sacz or Zakopane, the Theil–Sen slope value for Cracow is only 3–4% higher. The analysis of data from the months of the heating season shows a stronger trend in relation to PM10_{hs}. The comparison of the Theil–Sen slopes values for the Malopolskie Voivodeship (excluding Cracow) ($a_S = -4.05$) shows that the Theil–Sen slope a_{TS} is higher in Cracow and on average for the analyzed cities of the Malopolskie Voivodeship (excluding Cracow) ($a_S = -4.05$) shows that the Theil–Sen slope a_{TS} is higher in Cracow and on average for the analyzed cities of the Malopolskie Voivodeship (excluding Cracow) ($a_S = -4.05$) shows that the Theil–Sen slope a_{TS} is higher in Cracow by approximately 40% (Table 3). Compared to Nowy Sacz or Zakopane the Theil–Sen slope value for Cracow is 15–16% higher.

Moreover, in the case of average annual concentrations of BaP (PM10), the Theil–Sen slope a_{TS} is also the highest in Cracow ($a_S = -0.62$) (Table 4), but this time, the difference is more significant compared to other analyzed cities (e.g., 31% higher than in Zakopane, which had the highest Theil–Sen slope a_{TS} from the analyzed cities excluding Cracow). The comparison of the Theil–Sen slope values for Cracow and on average for the analyzed cities of the Malopolskie Voivodeship (excluding Cracow) ($a_S = -0.62$) shows that the Theil–Sen slope a_{TS} is higher in Cracow by approximately 56% (Table 3). The analysis of data from the months of the heating season again shows a stronger trend in relation to BaP_{hs} (PM10). The comparison of the Theil–Sen slopes values for the heating season for Cracow and on average for the analyzed cities of the Malopolskie Voivodeship (excluding Cracow) ($a_S = -1.3$) shows that the Theil–Sen slope a_{TS} is higher in Cracow by approximately 60% (Table 4). Compared to Zakopane (which had the highest Theil–Sen slope a_{TS} from the analyzed cities excluding Cracow) the Theil–Sen slope a_{TS} is higher in Cracow by approximately 60% (Table 4). Compared to Zakopane (which had the highest Theil–Sen slope a_{TS} from the analyzed cities excluding Cracow) the Theil–Sen slope value for Cracow is 19% higher.

The analysis of the rate of changes in the average annual concentrations of PM10 and BaP (PM10) in Cracow and other analyzed locations (Tables 3 and 4) showed that the rate of decline in Cracow is always higher than the rate of decline in each of the analyzed cities, as well as generally for the Malopolskie Voivodeship (represented by the analyzed cities, excluding Cracow). The difference is more pronounced with regard to the months of the heating season (PM10_{hs}, BaP_{hs} (PM10)). The rate of changes for BaP (PM10) concentrations in Cracow, compared to the rate of changes on average in the analyzed cities of the Malopolskie Voivodeship (excluding Cracow), is more intensive than in the case of PM10 concentrations. It is 1.5 times stronger with regard to the months of the heating season, and more than twice when analyzing the whole year data. It was expected, as individual heating of buildings (fuel combustion in small heating installations) is the dominant source of BaP in the air in Poland. In the case of PM10, fuel combustion in small heating installations is the main source of dust emissions, but followed also by industry and transport emissions, especially in big agglomerations such as Cracow. Since the concentration of BaP (PM10) is a better indicator of the effects of liquidation of highemission furnaces than the concentration of PM10, it can be concluded that the impact of actions related to the improvement of air quality in Cracow, in the context of changes taking place in selected cities of the Malopolskie Voivodeship, is more visible.

In the second part of the study, the monthly average concentrations of analyzed pollutants were also investigated. The results are shown in Figures 8 and 9. There is a clear inter-annual variation in the time series of the monthly mean concentrations of PM10 and BaP (PM10). After rejecting the months outside the heating season, the seasonality of the course of monthly mean concentrations of PM10 and BaP (PM10) is still visible. In such a situation, the classical Mann–Kendall test, even with a correction for autocorrelation, may not be effective. Therefore, these ranks were subjected to the seasonal Mann–Kendall test, and the seasonal Sen's slope was used to determine the magnitude of trends.

In 13 out of 14 analyzed cases (Figures 8 and 9 and Table 5), with regard to the months of the heating season, there are grounds to reject the hypothesis about the lack of a trend at the significance level $\alpha = 0.05$ ($p_{v,MK} < 5$). Only in Nowy Sacz, for BaP_m (PM10), $p_{v,MK}$ is 21%, which means no trend in the BaP series. All detected trends are negative.

The highest seasonal Sen's slope value of the monthly average concentration of PM10 variable, for the months of the heating season, is observed in Cracow ($a_{TS} = -3.52$), while in other cities a_{TS} values are much lower (Table 5). The magnitude of the trend on average in the analyzed cities of the Malopolskie Voivodeship (excluding Cracow) is 36% lower ($a_{TS} = -2.26$) than in Cracow.



Figure 8. Distributions of average monthly concentrations of PM10_m in 2011–2020: for all months (blue points) and the heating season (red points). The symbol a_{TS} means seasonality Sen's slope [ng/m³/year], while Z_{MK} and $p_{v,MK}$ are the result of the seasonal Mann–Kendall test. Linear regressions (dotted lines) are shown for comparison. (Source: own elaboration based on the data from the database of the Chief Inspectorate for Environmental Protection [29]).



Figure 9. Distributions of average monthly concentrations of BaP_m (PM10) in 2011–2020: for all months (blue points) and the heating season (red points). The symbol a_{TS} means seasonality Sen's slope [ng/m³/year], while Z_{MK} and $p_{v,MK}$ are the result of the seasonal Mann–Kendall test. Linear regressions (dotted lines) are shown for comparison. (Source: own elaboration based on the data from the database of the Chief Inspectorate for Environmental Protection [29]).

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Nr	Cite		PM10 _m		BaPm			
	City	Z_{MK}	Significance	a _S [μg/m ³ /year]	Z_{MK}	Significance	<i>a_S</i> [ng/m ³ /year]	
1	Cracow	-4.7	**	-3.52	-5.6	**	-0.75	
2	Tarnow	-4.9	**	-1.90	-3.6	**	-0.30	
3	Bochnia	-4.2	**	-1.91	-2.6	*	-0.36	
4	Gorlice	-5.2	**	-1.82	-3.6	**	-0.25	
5	Zakopane	-5.7	**	-2.43	-4.6	**	-0.50	
6	Nowy Sacz	-4.5	**	-2.60	-1.2		-0.18	
7	Malopolskie Voivodeship	-5.1	**	-2.26	-3.2	**	-0.28	

Table 5. Trends in average monthly concentrations of $PM10_m$ and BaP_m (PM10) in 2011–2020 (for the heating season) obtained using the seasonal Sen's slope and the seasonal Mann–Kendall (MK) test.

* if *p*-value $p_{MK} < 0.05$ (medium evidence), ** if *p*-value $p_{MK} < 0.005$ (strong evidence).

The highest seasonal Sen's slope value of the monthly average concentration of BaP (PM10) variable, for the months of the heating season, is observed in Cracow ($a_S = -0.75$). In other analyzed cities a_S values do not exceed -0.5. The magnitude of the trend on average in the analyzed cities of the Malopolskie Voivodeship (excluding Cracow) is 63% lower ($a_S = -0.28$) than in Cracow (Table 5). The rate of changes for monthly averages of BaP (PM10) concentrations in Cracow, in the heating season, compared to the rate of changes on average in the analyzed cities of the Malopolskie Voivodeship (excluding Cracow), is more intensive than in the case of PM10 concentrations (1.75 times stronger). This confirms the results of the analysis so far.

Due to additional factors, especially the differences in weather conditions in particular years, the results of air quality measurements cannot be treated unequivocally. The years 2015, 2018, 2019 and 2020 (according to the thermal classification developed by the Institute of Meteorology and Water Management [37]) have been classified as extremely warm. The average annual temperature recorded at the Cracow Observatory station was 2 °C higher for 2019, and 1.5 °C higher for 2020 than the multi-year average for 1981–2010 [25,38]. Due to the higher than average air temperature in 2015, 2018, 2019 and 2020, the demand for heat for heating buildings was reduced, which resulted in a reduction of low emissions from the municipal and household sector (having a decisive impact on the level of PM10 concentrations in built-up areas), and also reducing dust emissions and dust precursors from heating plants and combined heat and power plants. Therefore, a comparative method was used for the analysis, to eliminate temperature influence as much as possible.

It should also be noted that the data for 2020 is burdened with the impact of the COVID-19 pandemic. In the spring of 2020, in order to prevent the spread of COVID-19 infections in Poland, as in most countries, restrictions were introduced that resulted in the reduction of emissions from certain sectors of the economy (including transport, selected industries) and a potential increase in emissions from municipal and household sources due to the quarantine. Research [39] shows that in large cities of the country, the concentrations of PM10 were reduced in the range from 8.3% to 33.1%.

For a deeper analysis, the results of air quality measurements should also be considered in the context of climatic data, including the intensity of the heating season, the conditions for the dispersion of pollutants or other factors influencing the air quality. The conducted comparative analysis, between cities located relatively close to each other and subject to similar influences of climatic conditions or impacts resulting from the COVID-19 pandemic, is seen as a starting point for further discussions and analyses in that field.

5. Discussion

The problem of air quality in Cracow is inextricably linked with the closest vicinity of the city. This neighborhood consists of 15 municipalities (Figure 10) that also struggle with the problem of air quality: Biskupice, Czernichow, Igolomia—Wawrzenczyce, Kocmyrzow—Luborzyca, Liszki, Michalowice, Mogilany, Niepolomice, Skawina, Swiatniki Gorne, Wieliczka, Wielka Wies, Zabierzow, Zielonki and Koniusza. In these municipalities, the demand for heat is covered by individual heat sources: local and individual boilers and furnaces, where coal (60%) and natural gas (34%) are mainly used. In comparison, in Cracow about 65% of residents are supplied with heat from the municipal heating system (municipal heating network is almost 900 km long). Thanks to the agreement, in areas where there is no municipal network, it will be possible to connect buildings to local networks, supplied with heat from small gas heat sources [40].



Figure 10. Municipalities adjacent to Cracow, which are the closest vicinity of the city. (Source: own elaboration). Legend: 1—Kocmyrzow—Luborzyca, 2—Michalowice, 3—Zielonki, 4—Wielka Wies, 5—Zabierzow, 6—Liszki, 7—Czernichow, 8—Skawina, 9—Mogilany, 10—Swiatniki Gorne, 11—Wieliczka, 12—Biskupice, 13—Niepolomice, 14—Igolomia—Wawrzenczyce, 15—Koniusza.

The municipalities adjacent to Cracow, just like Cracow, started the process of decommissioning coal-fired boilers, but are characterized by a relatively low pace. It is estimated that there are still about 23,000 to be replaced in the municipalities surrounding Cracow. Introduced currently the help of eco-managers resulted in the acceleration of the pace of change and the submission of twice as many applications for co-financing for the replacement of a furnace from the Clean Air Program.

The situation is expected to improve in the near future. According to the Air Protection Program for the Malopolskie Voivodeship, all classless boilers must be replaced by the end of 2022. From January 2023, a fine or a court application will be issued for the use of classless boilers [41].

6. Conclusions

- The implementation of the provisions of the anti-smog resolution for the Malopolskie Voivodeship and the local anti-smog resolution for Cracow results in the general improvement of air quality in Cracow and the Malopolskie Voivodeship in terms of PM10 and BaP (PM10) concentrations.
- The rate of decline in the average annual concentrations of PM10 and BaP (PM10) in Cracow is always higher than the rate of decline in each of the analyzed cities, as well as generally for the Malopolskie Voivodeship (represented by the analyzed cities, excluding Cracow). The difference is more pronounced with regard to the months of the heating season.
- The rate of changes for the average annual BaP (PM10) concentrations in Cracow, compared to the rate of changes on average in the analyzed cities of the Malopolskie Voivodeship (excluding Cracow), is more intensive than in the case of PM10 concentrations. It is 1.5 times stronger with regard to the months of the heating season and more than twice when analyzing the whole year data.
- The highest seasonal Sen's slope value of the monthly average concentration of PM10 and BaP (PM10), for the months of the heating season, is also observed in Cracow. The magnitude of the trend on average in the analyzed cities of the Malopolskie Voivodeship (excluding Cracow) is 36% lower in relation to PM10 and 63% lower in relation to BaP (PM10) than in Cracow.

- The rate of changes for monthly averages of BaP (PM10) concentrations in Cracow in the heating season, compared to the rate of changes on average in the analyzed cities of the Malopolskie Voivodeship (excluding Cracow), is more intensive than in the case of PM10 concentrations (1.75 times stronger).
- Since the concentration of BaP (PM10) is a better indicator of the effects of liquidation
 of high-emission furnaces than the concentration of PM10, it can be concluded that the
 impact of actions related to the improvement of air quality in Cracow, in the context of
 changes taking place in selected cities of the Malopolskie Voivodeship, is more visible.
- Cracow's more radical air quality policy (prohibiting to the use of coal and wood in boilers and fireplaces in Cracow) brings better results than the moderate activities carried out in the Malopolskie Voivodeship.

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