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The Origin of Dust Particles in Atmospheric Air in Krakow (Poland) (Atmospheric Background)

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Abstract: For several decades air pollution in Krakow has been a serious and an unresolved environmental and social problem. The causes of high concentration of particulate matter, such as PM10 and PM2.5 in Krakow are both natural and anthropogenic. Nevertheless, the sources of dust pollution have not been fully determined yet. The main source of dust in Krakow is local emissions, however, particles from adjacent areas might also contribute significantly to the pollution. Transboundary dust should also be taken into account while investigating the problem. The aim of the study is to determine what type of particles are present in the atmospheric air in Krakow and to make an attempt at determining their sources. The analytical method applied in the study was the Scanning Electron Microscopy with Energy Dispersive Spectrometry (SEM-EDS). In addition, the HYSPLIT model was used for data analysis and for determination of particles source areas. The analysis of individual dust particles indicates that they are very diverse in terms of chemical composition and particle size. Moreover, the analysis shows that the particles are of various origins, such as anthropogenic and natural, as well as that some of them are formed in the air by chemical reactions. The analysis of particulate matter demonstrates that the majority of it consists of particles with a diameter of less than 1 µm. The concentration of very fine soot particles (nanoparticles) seems to be the highest, however, spherical aluminosilicate particles such as iron and titanium oxides are also found.

Keywords: air pollution; meteorological parameters; emission; health impact; Poland

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1. Introduction

High concentration of dust particles affects air quality and thus has a significant and direct impact on human health. People living in urban areas are at a greater risk of harmful effects resulting from air pollution. Epidemiological studies have shown adverse associations between exposure to high particulate matter concentration and human health [1–3] as well as they have revealed that there exists a link between the mortality rate increase and presence of smog [4–6]. Although between 2000 and 2010 the PM10 concentration generally decreased in Central Europe due to changes in meteorological conditions such as higher wind speeds or warmer winters, the PM10 concentration in Poland STILL exceeds the standards set by the European Union and World Health Organization (WHO).

Episodes of high concentration of dust pollution are not only present in Krakow. Air pollution is a serious social, economic and environmental problem [7–10] in many cities in Poland as well as in Europe. In Krakow, however, the problem is more noticeable and raises many concerns due to the very high PM2.5 concentration that frequently exceeds norms over selected periods in a calendar year. Particles PM2.5 are particularly dangerous to health of city residents [11] because they have the ability to penetrate and retain in human lungs as well as to enter the blood vessel walls [12–15]. Poor air quality in Krakow remains and poses a serious risk to human health despite many actions taken to reduce the pollution. The presence of heavy metals in the dusts found in Krakow and their negative impact on the health of city inhabitants should also be emphasized [16–18].

The causes of high particulate matter concentration in Krakow are both natural and anthropogenic. Natural factors are primarily associated with the meso- and local microclimatic conditions (i.e., the large number of days with temperature inversion, low average wind speed especially during winter season), whereas anthropogenic factors are mainly associated with emission of pollutants generated by fuel combustion happening in home furnaces, power plants, industry and car engines. In addition, an inflow of solid particles from adjacent areas might also contribute to the problem [19,20].

As previously mentioned, a multitude of factors affecting air quality in Krakow, including unidentified sources of particulate matter, is an important obstacle to improving the aerosanitary conditions in Krakow.

The primary goal of this study is to determine what type of particles are present in the atmospheric air in Krakow. The subject of analysis is the chemical composition, shape and fraction of individual particles. The secondary goal is an attempt to determine how a direction of air masses inflow affects the particles concentration in the air.

2. Materials and Methods

2.1. Study Area

Krakow is a large city with the population of over 750 thousand people. It is situated in the southern part of Poland and is the capital of Lesser Poland Voivodeship (Figure 1). The city is located at the confluence of two large geological structures (with hills up to 100 m in the north and the south). The morphometric diversity of the researched area together with urban development determine local climatic conditions that are characterized by limited ventilation favoring high concentration and low dispersion of dust as well as by the high frequency of temperature inversion i.e., accumulation of dust in boundary layer.

Figure 1. Location of Krakow.

Episodes of the high suspended dust concentration in Krakow are caused by low emission (combustion of solid fuels), a rapid increase of the number of motor vehicles as well as by high emissions from power plants arising from the increased energy consumption. However, air quality in Krakow may also be affected by the inflow of dust from adjacent area as well as by the transboundary transport of particulate matter occurring in favorable circulation conditions and originating in industrial zones located in Germany, Czech Republic, Slovakia and Hungary. The extremely high concentration of suspended dust is frequently recorded during the winter season, which is caused by the combination of both high emissions and weather conditions.

2.2. Data Collection

A dust pollution sampling point was located on the roof of a five-story building (Pedagogical University) at 37 m above ground level. The location of measuring point met all standards pertaining to the distance from a local emission point as well as to the distance from roads and industrial centers. The sampling was carried out between June 2016 and February 2018 over periods characterized by the most coherent conditions that are specific to a particular type of circulation in the atmosphere. Data on dust sources and on-air mass advection sectors was compiled using the HYSPLIT (HYbrid Single Particle Lagrangian Integrated Trajectory) model [21].

The samples consisted of particles collected on polycarbonate filters with pores and with a diameter of 0.4 µm. The material was collected using an aspirator GilAir Plus STP. In order to ensure the representativeness of individual samples, it was necessary to collect at least 500 L of air in conditions characterized by a stable inflow of air masses from a particular direction. An automatic meteorological station provided valid meteorological data including temperature, atmospheric pressure, humidity, wind speed and direction, at the measurement point (Table 1).

Table 1. Meteorological conditions during sampling (cold season/warm season).

Advection Direction	Temperature [°C]	Humidity [%]	Average Wind Speed [m/s]
north	−3/14	59/65	4.1/3.4
east	−4/18	49/66	3.2/2.9
south	2/21	57/74	2.2/2.1
south-west	1/20	59/73	2.9/2.6
west	1/19	80/84	3.6/3.3
north-west	0/18	78/81	3.9/3.5

In total 1526 individual particles were analyzed from twelve collected samples. The chemical composition and a shape of particles were analyzed using the Scanning Electron Microscope (SEM) with energy dispersive spectroscopy (EDS) system. The microscope Hitachi S-4700 allowed for very high magnification (up to 500,000×) which was especially important for investigating a large number of nanoparticles present in the analyzed material.

2.3. Back Trajectory Analysis

Determined backward trajectories of particles were calculated using the HYSPLIT model that traces potential transport pathways of dust pollutants. This model enables a reconstruction of a trajectory of air masses inflow transporting dust particles over the selected area. The model allows both: determining a single trajectory as well as carrying out complex dispersion and deposit simulations using a cloud or particle approximation. Model calculations were performed on historical meteorological data GDAS (Global Data Assimilation System). The back trajectory (24 h back trajectories) analysis was performed on selected days on data collected at 100, 500 and 1000 m above ground level. Model calculations were performed on historical meteorological data from GDAS (Global Data Assimilation System). The input data included meteorological parameters such as wind direction and speed, atmospheric pressure and air temperature.

3. Results

3.1. Typs of Occuring Particles

The chemical composition determined by EDS and single grains analysis as well as a shape of a dust particle were the main criteria used for particles classification. The carried-out analysis along with earlier studies [22–26] identified the following nine types of particles:

- Carbonaceous particles (soot): a single particle of about 50 nm or aggregates made of such several particles. Particles of an elongated, branched shape were also found along with almost isometric aggregates of numerous soot particles.

- Sulfates and chlorides (Na, K, Na-K, Na-K-Ca): were present in a form of irregular particles or polyhedrals and tablet forms. Calcium sulphate (gypsum) as well as other sulphates (containing S $\geq$ 15% by weight) often formed crystal clusters of size larger than 1 µm.
- Carbonates (Mg, Ca): were present in a form of particles of various size. Some of them were smaller than 1 µm.
- Silicates and aluminosilicates (Ca, Ca-K, Ca-Mg-K, Ca-Mg-K-Fe): appeared either as single particles or in a form of aggregates. The aggregates reached the size up to several dozen micrometres. Individual particles either were of irregular shape or they formed plaques. The silicates were classified as particles with Si $\geq$ 2%, while the aluminosilicates contained Al in the range of $\leq$2% to 50% by weight.
- Bioaerosols: particles usually exceeded 1 µm in size. They were particles of biological origin and consisted of plant pollen, plant fragments, insect fragments, fungal spores.
- Iron oxides, i.e., particles with Fe $\geq$ 5% by weight, most often occurred in a form of densely packed aggregates.
- TiO_2 particles: i.e., particles with Ti content of $\geq$5% by weight, formed aggregates of size 100 to 350 nm.
- Aluminum oxides, i.e., particles containing at least 4% of Al by weight, occurred in a form of spherical grains of size ranging from 100 to 300 nm.
- Particles rich in metals, such as for example: Cr, Ni, and in which these metals concentration made up at least $\geq$2% by weight were usually in a form of irregularly shaped grains.

3.2. Chemical Composition, Shape and Size

As previously mentioned, twelve samples were collected in long time period which results in: six samples during the cold weather (from October to March) and six during the warm weather (from April to September). The sampling method consisted in predicting and then monitoring of both circulation and atmospheric processes. In each case, no precipitation was allowed to occur on a particle trajectory path (excluding leaching and wet deposition). In general, the anticyclonic circulation dominated in eleven cases, whereas the cyclonical circulation was observed only once. The most common direction of advection was the south-west sector (four samples). The corresponding samples no. 3 and no. 2 were collected during the air inflow from the east and south. Individual samples were also gathered from the following directions: west (W), north-west (NW) and north (N).

Twenty elements were identified in the analyzed material. The most common element was carbon whose presence was found in 87.31% of all grains. Carbon also had the largest variability in the chemical composition of all tested particles (Figure 2). The carbon distribution analysis showed the clear left-hand asymmetry. Carbon accounted for 95% of all elements in as much as half of all the particles containing this element. The weak right-sided asymmetry was typical of the distribution of sulfur content. In case of elements with occurrence above 10% (i.e., silicon, calcium, aluminum, sodium, potassium, iron, magnesium and oxygen) data showed the right-sided asymmetries of varying intensity. The analysis showed the high concentration—above the 10th percentile—of oxygen, silicon, sodium, calcium and iron in the chemical composition of individual particles. The analysis also demonstrated the trace concentration of cobalt and molybdenum.

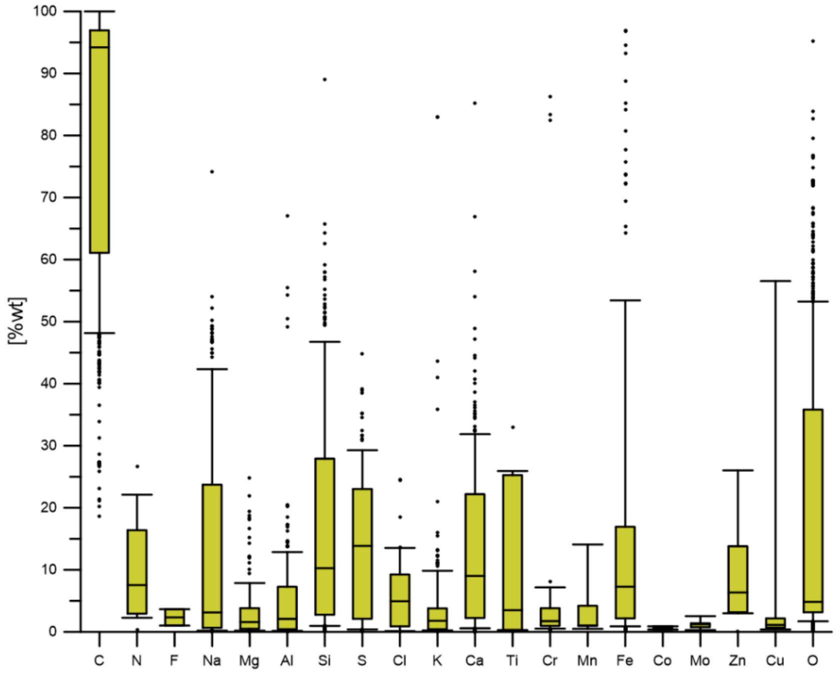

Figure 2. Characteristics of the chemical composition of particulate matter in Krakow.

As previously mentioned, carbon was the dominant element in the composition of dust particles, however, the content of it in the samples varied strongly and depended on a direction of air masses inflow. The lowest content of coal in terms of % by weight in dust particles was observed during the influx of air from the west and accounted for less than 75%. The first quartile of the distribution was 54.3 wt.%, whereas the third quartile reached 57.2 wt.% Moreover, there was a large variation in the concentration of coal in grains during advection from the eastern sector. The highest weight concentration of carbon in the studied PM10 particles occurred during the northern circulation. The first quartile of carbon concentration in the grains during the above-mentioned advection totaled 94.6 wt.%.

The analysis of the occurrence of elements in particulate matter during advection of air masses from selected sectors showed large variability in the frequency and quantity of components in the tested particles. The smallest variation in the chemical composition of particles was present during the northern inflow of air masses. The Figure 3 demonstrates concentration of selected elements (% by weight) in PM10 dust particles in Krakow during advection of air masses in the analyzed sectors 13 elements were found during advection from the above direction. Si proved to have the largest variation in particles weight. The presence of elements such as Cl, Ti, Co and Fe was only marginal. The eastern (Figure 3b) and southern (Figure 3f) circulation demonstrated a relatively small variation in the chemical composition compared with other air mass inflows. Si weight concentration in the particles was slightly elevated during the inflow from the east, whereas the southern advection caused an increase in the Al and Cr concentration. The most diverse chemical composition of the particles occurred during the southwest (Figure 3d) and northwest (Figure 3f) circulation. Among the analyzed particles, the southwest advection caused the increase in concentration of N, Na, Si, S, Ca, Cr, Ti, Zn and O. During advection from the northwest sector, there was an explicit increase in Zn concentration in PM10 particles. The western circulation (Figure 3c) caused the higher weight concentration of Fe among the tested grains.

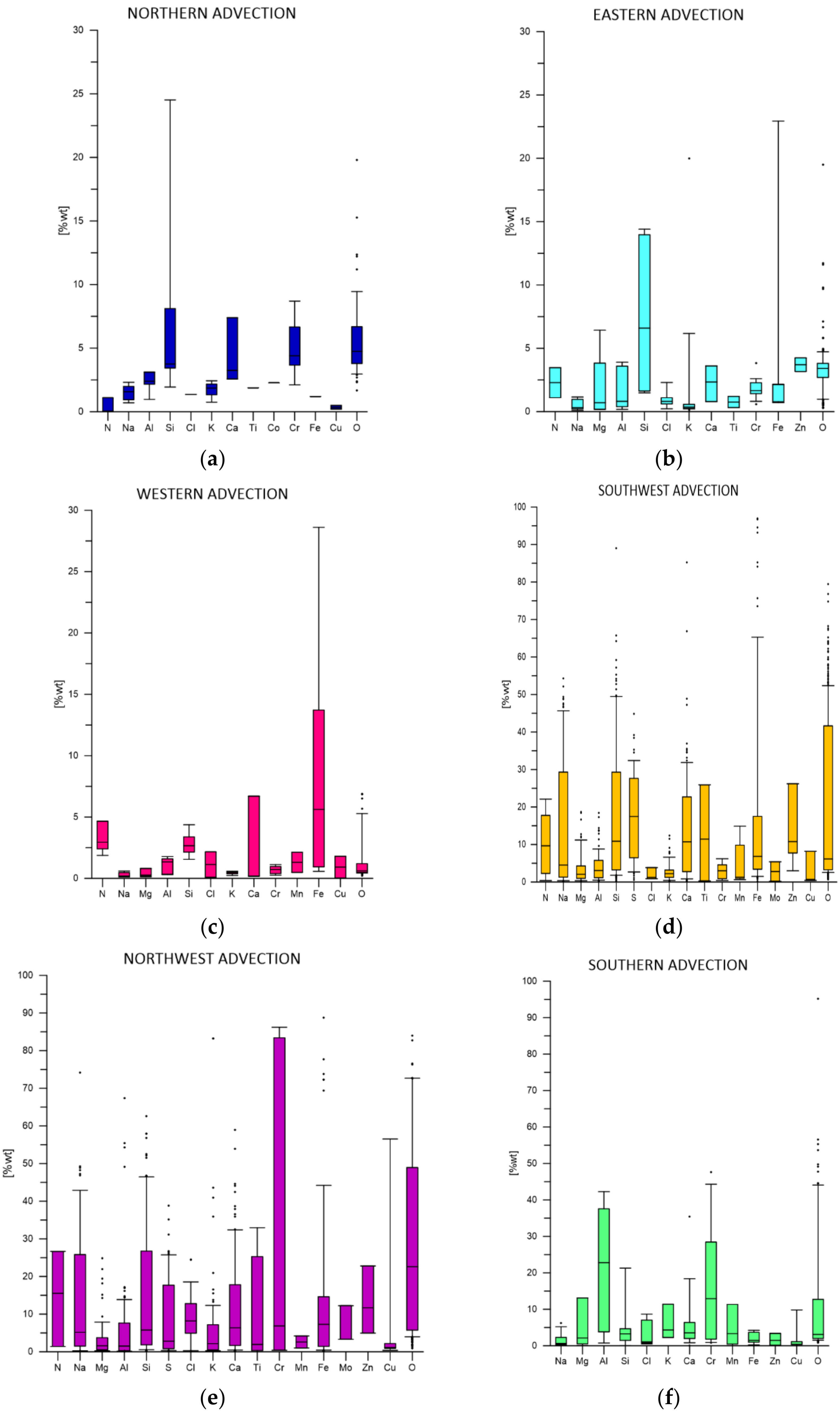

Figure 3. Concentration of individual elements in PM10 in Krakow depending on direction of the inflow of air masses.

A shape of dust particle might also help to determine the possible origin of dust particles. Particles of acicular shape, such as aggregates composed of particles of regular shape, and of spherical habit dominated in the analyzed samples and accounted for 47.3% of all shapes. A particle and plate-like particles made up 30.8% of all tested particles. Regularly shaped spherical particles were the least common.

Directions of air masses inflow and their impact on concentration of elements in PM10 in Krakow. The analysis of dust particles collected on polycarbonate filters showed that the percentage concentration of particles of a specific shape varied depending on a direction of air mass inflow. During advection from the southwest sector acronymic particles constituted the largest share—55.24%—of all tested particles. The share of this type of particles dropped slightly to 54.8% during avection from the eastern sector. Tablet-like and shallow particles dominated during the air masses inflow from the northwest sector and their percentage share was 37.3%. Acaric dust, on the other hand, made up only a small share. Spherical particles occurred most frequently during advection from the northern sector and constituted 27.6%.

Carbon particles were composed either of single grains of size about 50 nm or of aggregates made of several such grain particles (Figure 4). The soot aggregates occasionally took on an elongated and branched shape and their diameter was about 10 µm (Figure 5). The soot particles also formed strongly compacted aggregates. The origin of the analyzed soot particles was difficult to determine due to the fact that their morphology, shape and size varied and depended on the following: a type of fuel burned, the course of the combustion process (temperature, exhaust gas cooling rate) and chemical reactions that took place in the atmosphere. The analysis also showed that the soot aggregates were coated either with secondary organic aerosols of spherical shape and of size up to 500 nm or with irregular and slightly rounded particles (Figure 6).

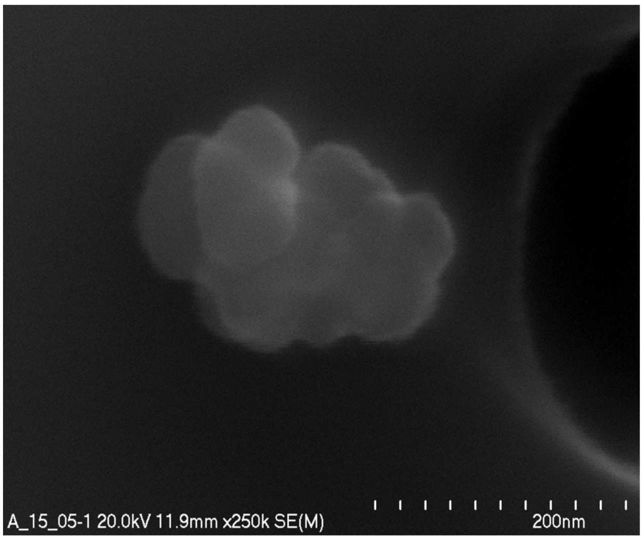

Figure 4. Soot particle aggregate.

Sulphates found in the samples were a relatively large group of particles characterized by irregular or grain-like multi-walled shape (Figure 7). The sulfates relatively often formed crystals in the solution of condensed steam-present on a filter. In addition to the sulphates, chlorides and phosphates were also commonly observed.

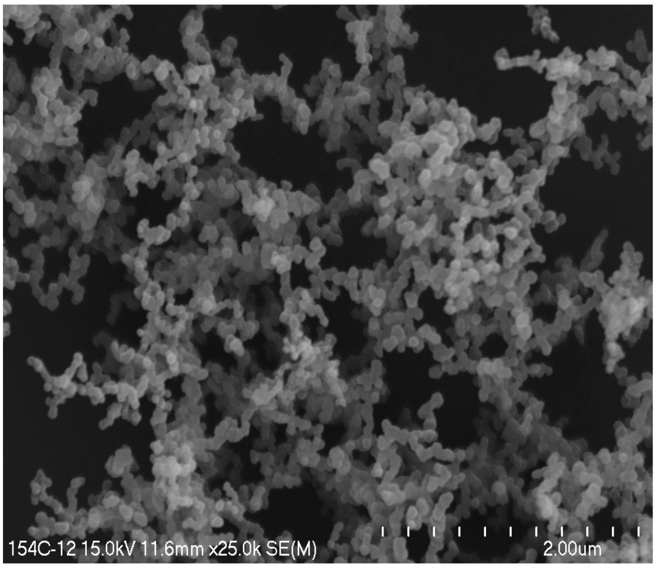

Figure 5. Branched aggregates of soot particles.

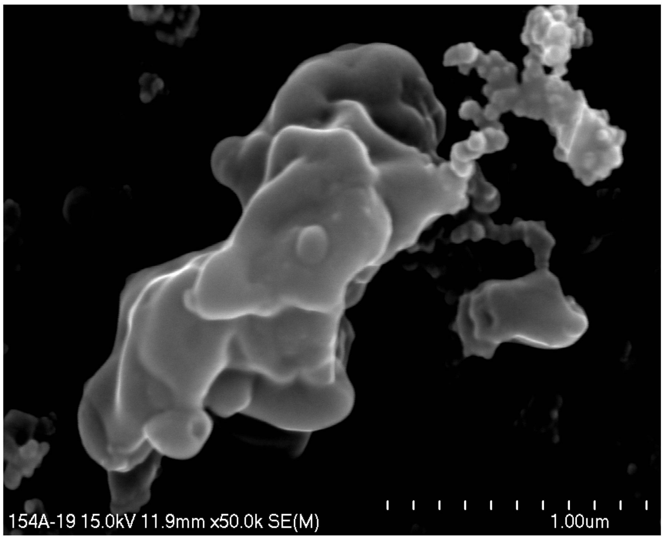

Figure 6. Secondary organic aerosols.

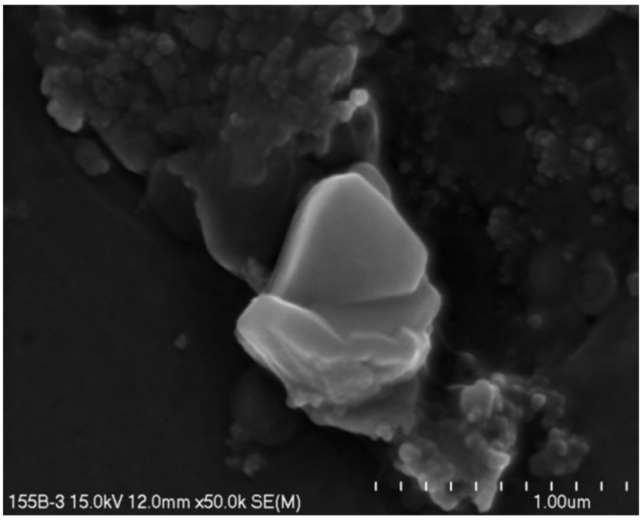

Figure 7. K-Mg and Ca sulfates.

The observed carbonates assumed a form of particles of various size among which particles below 1 µm were found. Mg and Ca carbonates were also identified. The chemical composition of these particles was similar to the stoichiometric formula of dolomite or calcite.

Mineral particles, such as for example silicates and aluminosilicates, occurred either in a form of single grains of irregular shape or in a form of plaques. In addition, aggregates of mineral particles reaching sizes of several dozen micrometers were observed. Particles of this type were dominated by mixed particles whose chemical composition rarely matched the stoichiometric formulas of commonly occurring minerals. Spherical aluminosilicate particles, which were emitted most likely from large power plants, were also observed (Figure 8).

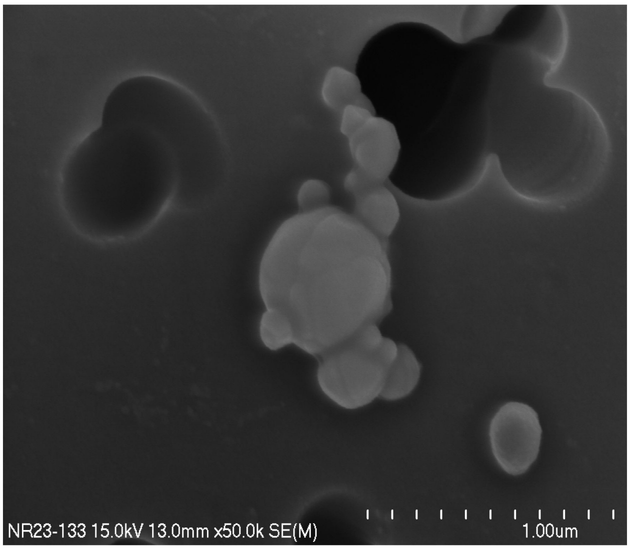

Figure 8. Spherical aluminosilicate particles.

Iron oxides and other metallic particles were most often in a form of densely packed aggregates, or in a form of particles whose size did not exceed 1 µm (Figure 9). The grains in which metals, such as chromium, nickel and zinc, were found usually assumed an irregular or sharp-edged form.

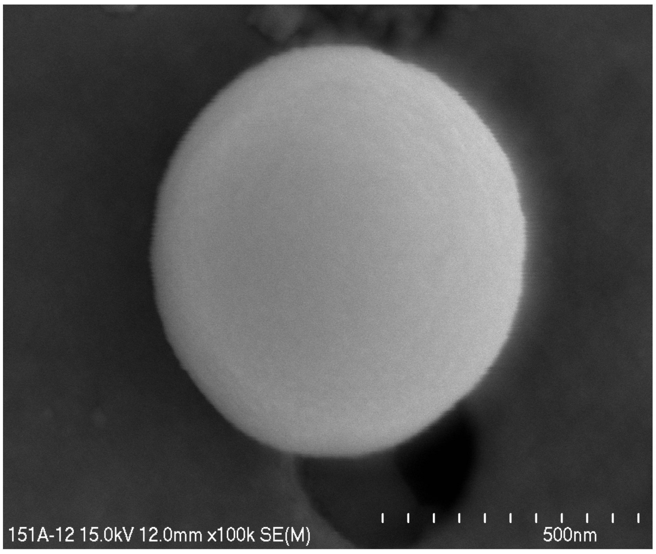

Figure 9. Spherical iron oxide particles.

Titanium oxide particles were also observed in the tested material. They formed very small aggregates of size raging between 100 to 350 nm (Figure 10).

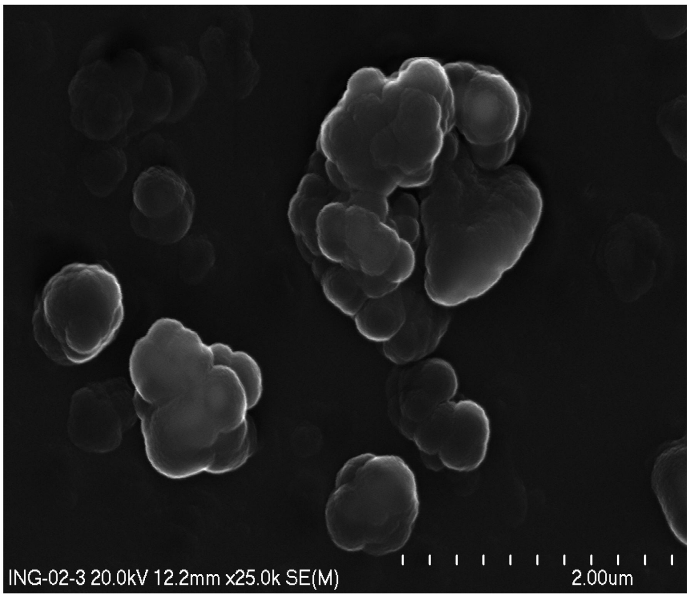

Figure 10. Spherical titanium oxide particles.

Aluminum or zinc oxide particles (Figure 11) occurred sporadically and most often took a form of spherical grains of size ranging from 100 to 300 nm.

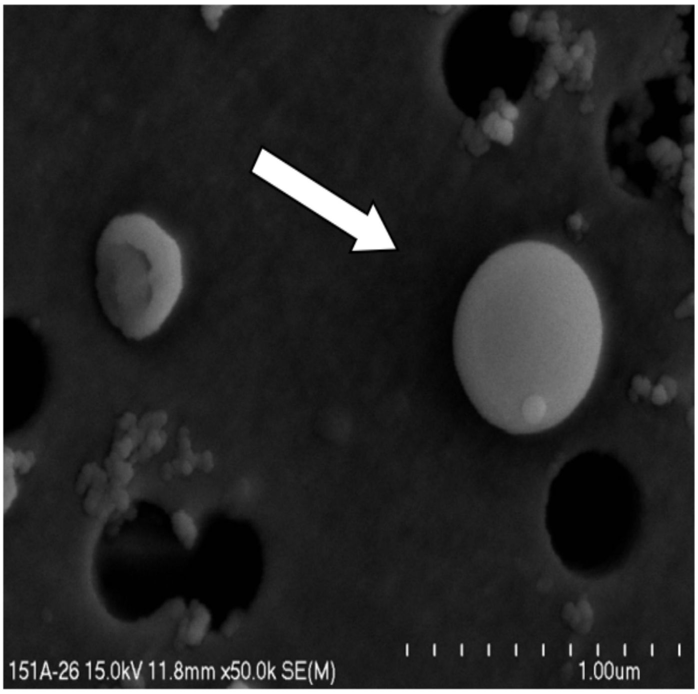

Figure 11. Spherical alumina particle.

Bioaerosols contained a group of particles that included plant pollen, plant fragments (sometimes crushed) or insects. Their size often exceeded 1 µm. The presence of brochosomes (Figure 12) i.e., microscopic hydrophobic granules secreted by insects was observed among the tested aerosols. The occurrence of this type of dust increased particularly in spring.

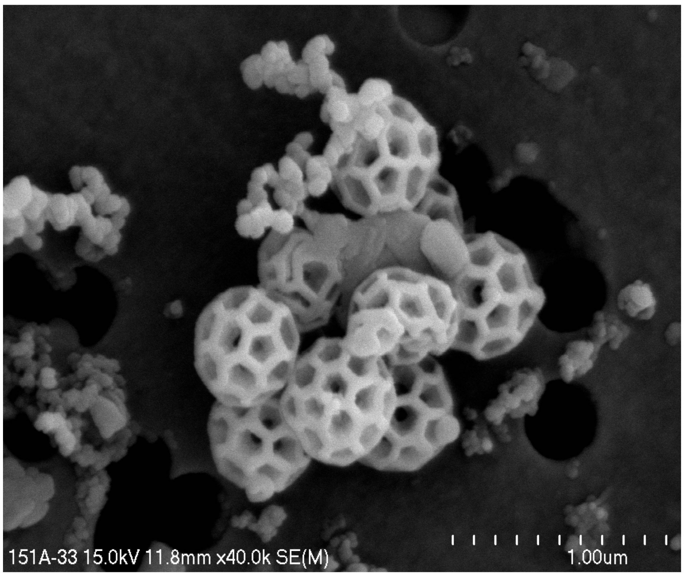

Figure 12. Biological aerosols (brochosomes).

4. Discussion

The diversity of the particles chemical composition and shapes present in the atmospheric air in Krakow is determined primarily by the type of emissions in the city. It is also proved by the studies conducted so far [16,17], where it has been shown that in the dust prevalent mineral phase include quartz and also unburned coal (low emission from household) and gypsum (secondary phase of the anthropogenic origin). Furthermore, probable that some of the dust comes from neighboring areas as well as that they are deposited over Krakow as a result of long-distance transport on days with meteorological conditions favorable for this process.

The analysis of particulate matter in Krakow showed that, in terms of particle size, grains with a diameter smaller than 1 µm dominated. Nanoparticles of size below 100 nm were very often observed in the ultra-fine dust fraction. The similar particle size distribution has also been identified in other studies [27–30]. The most frequently occurring particles were carbon with the soot being the most dominant. As mentioned before the high proportion of these particles is characteristic of urban areas and is commonly associated with dust emissions from coal-fired home furnaces [31–35] and in Krakow case also often burning low-quality coal in coal-fired stoves on area around the city [36].

However, it should be emphasized that soot particles originating from sources such as coal combustion or diesel engines do not differ significantly in morphology and are not distinguishable by their specific chemical composition. Nevertheless, the increase in the percentage share of soot particles in the overall dust concentration in Krakow may be explained by the systematically growing number of vehicles in the city area with a concurrent decrease in the number of solid fuel fired home furnaces. The NO_2 concentration level may be a premise concerning the influence of car emissions on share of soot particles from the combustion of fuels. However, there is no clear correlation in Krakow between the car pollutant levels and the season of the year but greater values of NO_2 is observed in the winter season [37]. This observation supports findings of other studies that have identified traffic emissions as a source of high concentration of dust in other cities around the world [20,38–42].

The particularly high concentration of soot particles was observed in samples collected in the cold half of a year (October–March), which substantiates the previous research findings [43–45]. Other particles that were present in a large number during this period were: secondary aerosols (sulfates associated with the transformation of SO_2 emitted during the combustion of fuels in household furnaces, industry and energy). For the presence of various hydrocarbons (considered i.e., mineral oils and traces of gasoline from combustion motor fuels) in the dusts in Krakow also mentioned in other publications [17].

In the samples collected during the southwestern and western advection, the increased occurrence of particles such as spherical forms of aluminosilicates and iron oxides was observed. This type of dusts most likely come from power plants in Kraków and Skawina (city located around 15 km in south-west direction from Krakow). The influence of the Skawina power plant may be confirmed by the increased content of iron in samples collected during south-western and western advection.

The south-west circulation was distinguishable by the increased concentration of particles containing iron, zinc and titanium. The increased share of these particles during this type of circulation may be associated with emitters located near Budapest on the Slovak-Hungarian border [45]. A similar connection was also observed during the north-west advection during which the collected material was richer in particles containing chromium, zinc, iron and titanium. High concentrations of zinc in the dusts of Krakow are associated with long distance emission from Olkusz area (city located 40 km in north-west direction from Krakow), where a zinc deposit is exploited [17,41,46].

Interestingly, titanium particles were also detected in the sampled air. In years preceding this research, no titanium particles had been found in atmospheric air in Krakow and their current occurrence may be associated with the increased use of titanium compounds in building materials and paints [24].

5. Conclusions

Particulate matter is one of the main pollutants in Europe that pose risk to human health. In the EU-28.80% of urban population is exposed to air pollutants, such as PM10 and PM2.5, which concentration repeatedly exceeded the WHO guidelines. Moreover, it has been established that the long -term exposure to PM2.5 resulted in about 412,000 premature deaths in 2016 in Europe alone [5]. In Poland, the estimated number of premature deaths due to poor air quality is 45,000 annually [36].

The aim of the research was to determine what type of particles are present in the atmospheric air in Krakow (chemical composition, shape and dominate fraction of particles), with particular emphasis on the potential impact of the direction of the inflow of air masses on the diversity of the particles.

Soot particles formed during the combustion of solid fuels in home furnaces, industrial and service plants as well as soot coming from car engines dominated in the analyzed samples.

Among the analyzed particles, the most numerous group (over 70%) consisted of particles with a diameter up to 1 μm. In the ultrafine fraction nanoparticles of size below 100 nm were observed repeatedly. The share of ultrafine dust (over 85%) was particularly significant in samples collected in the cold season (October–March) during north and east advection, characterized by low temperature and low average wind speed. These particles are particularly harmful to human health due to their ability to penetrate and retain in human lungs. It should also be mentioned that the concentration of fine particles present in the air does not translate into a significant weight share (μg) in the unit of air volume (m^3).

Diversity of the chemical composition and occurrence of particles of a specific shape in the tested material depended on a direction of air masses inflow. This variability may be conditioned by the internal structure and intensity of emissions in the city as well as by the potential inflow of pollution from neighboring areas.

In the case of Krakow, the soot particles origin from coal-fired stoves located around of the city are difficult to distinguish from the material emitted in situ. On the other hand, the increased share of particles of different chemical composition during the inflow of air masses from selected directions may most likely indicate the inflow of particles from, among other places, industrialized areas, especially in the west sector of the city.

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Conflicts of Interest: The authors declare no conflict of interest.

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