

MEASUREMENTS OF AEROSOL SIZE DISTRIBUTION IN URBAN
AREAS OF UPPER SILESIA

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e-mail: kskubacz@gig.eu**Keywords:** Roads, ultrafine particles, fine particles, coarse particles, size distribution.

Abstract: Deposition of aerosols in the respiratory tracts depends on their size distribution. Investigation of such distribution has therefore great meaning for appropriate assessment of risk caused by hazardous pollutants that appear in environment both as a result of human activities like industry, emission from motor vehicles, municipal emission due to house furnaces and natural phenomena. The results of screening measurements of size distribution performed in several places located on highly populated areas in Upper Silesia, Poland are described in this paper. The applied equipment makes it possible to cover the range from several nanometers up to 20 μm , practically all particles that belong to the respirable class. Obtained results prove that a lot of particles having a size of the order of submicrons are produced close to high density traffic roads. This concentration strongly changed in time.

INTRODUCTION

Deposition of aerosols in the respiratory tract is mainly controlled by similar phenomena to those occurring during air filtration with fibrous filters. Therefore, diffusion, gravitational sedimentation and inertial impaction should be taken into consideration to explain the major transport mechanisms. Certainly, this process is influenced not only by physical aerosol properties but also by the breathing patterns and flow field in respiratory airways. The total deposition, as reported by Heyder *et al.* [4], increases with increasing breathing rate as a result of inertial impaction, especially for particles greater than 4 μm in diameter and mouth-breathing. Sedimentation has greater significance for aerosols below this diameter and longer residential time in the respiratory system. The third phenomenon, diffusion, is getting the dominant mechanism for particles not greater than 0.1 μm .

Particles, which were not removed from the respiratory tracts by the clearing process, influence people and animals by penetrating into lymphatic pathways and connective tissues. Therefore, investigation of their size distribution is crucial for appropriate assessment of risk caused by hazardous pollutants that appear in environment both as a result of human activities like industry, emission from motor vehicles, municipal emission caused by household furnaces and natural phenomena.

Radon and its progeny can serve as an example of natural hazards. Radon can penetrate from soil into buildings causing serious problems in some countries because of high-

level radiation risk to inhabitants. After decaying of radon such isotopes as polonium-218, lead-214, bismuth-214 and polonium-214 are generated. They are called short-lived radon daughters because their half-time, being less than 30 minutes, is short in relation to half time of radon equal approximately to 3.8 days. At the very beginning these isotopes occur as free atoms, ions or clusters that contain additionally vapors of gases (unattached fraction). Then, within 1–100 s, the radon progeny attach to ambient aerosols forming radioactive aerosols (attached fraction). This process is strongly affected by concentration and size distribution of ambient aerosols [10, 11]. Deposition of radioactive aerosols in the respiratory tracts and their radiotoxicity depends on half-time, aerosol size and total alpha radiation energy released by short-lived radon daughters. During decay of these isotopes beta and gamma radiation occur as well. This radiation however can be neglected because of its considerably lower, in relation to alpha radiation, linear energy transfer to tissue (absorbed energy per unit distance). As a result tissue damage due to alpha radiation is 20 times greater. According to the dosimetric model by Jacobi and Eisfeld [7] the greatest dose related to the unattached fraction of polonium-218 is absorbed in the tracheobronchial region (2–4 bronchi generation). Such isotopes as lead-214 and bismuth-214 occur mainly as attached fraction and affect at most the bronchi generations from 10 to 14. However, the influence of short-lived radon daughters on effective dose is commonly expressed as a combined effect. In the publication of the International Commission on Radiological Protection [6] the conversion coefficient is defined as effective dose per alpha radiation energy released by all these isotopes contained in unit volume and per exposure time. Such coefficient is a measure of hazardous effect of short-lived radon daughters on humans and depends strongly on aerosol size, breathing rate and breathing way. As it was proved, the most hazardous radioactive aerosols have a size

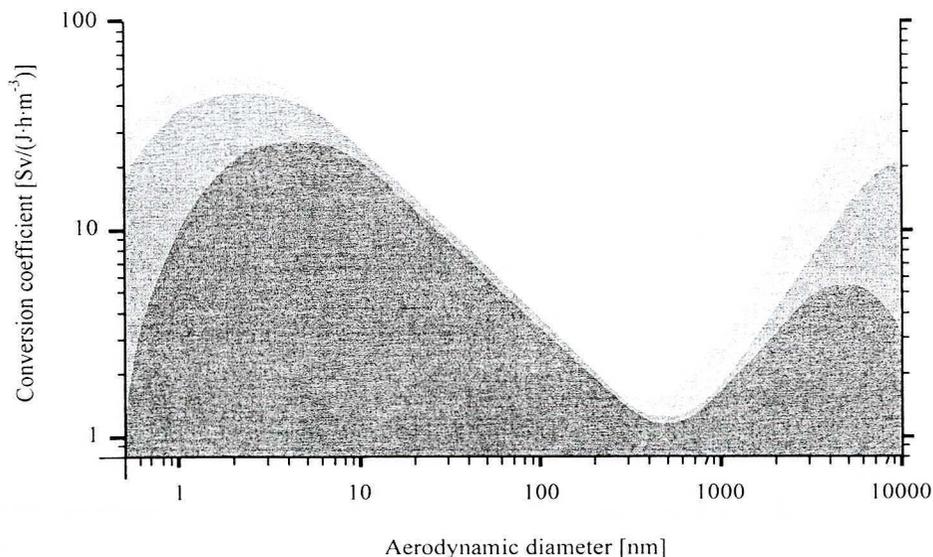


Fig. 1. Dependence of conversion coefficient on particle diameter and breathing pattern: mouth-breathing (1.70 m³/h), mouth-breathing (1.20 m³/h), mouth-breathing (0.75 m³/h), nose-breathing (0.75 m³/h) from top to bottom curve respectively

on the order of nanometers or near to the upper limit of respirable particles equal to 10 μm (Fig. 1). Measurement of radioactive aerosols and their size distribution is based on commonly applied methods, such as impactors and diffusion battery screens that are combined with such radiometric methods as alpha spectroscopy where the alpha radiation is spectrometrically detected to identify isotopes [12].

In the present publication, results of screening measurements of aerosol size distribution are discussed. Three sites were located in an urbanized region of Upper Silesia (Poland) and the last one far from roads and highly populated areas. Applied measuring devices made it possible to investigate the particles size distribution from several nanometers up to 20 μm . Such range covers practically all respirable class of aerosols.

The respirable particles belong to one of the three categories mentioned in the EN-481 standard [14]. According to the definition this category consists of particles that reach the alveoli. Fifty percents of the particles with an aerodynamic diameter of 4 μm belong to the respirable fraction and their dimensions can reach approximately 10 μm . Particles of the thoracic class are able to pass the larynx, and 50% of the in-air-suspended particles with an aerodynamic diameter of 10 μm belong to this class. The largest diameter however can reach approximately 30 μm in this case. The broadest, so-called inhalable class, is composed of particles that can be inhaled by nose or mouth. Particles with aerodynamic diameter greater than 100 μm are not included in the inhalable convention.

Another more detailed classification corresponds to the particles below 10 μm in size (PM10). Particles that are less than 0.1 μm in diameter are commonly defined as ultrafine, these with diameters ranging from 0.1 μm to 2.5 μm as fine, and these from 2.5 μm to 10 μm as coarse [2, 5, 8]. Sometimes an additional definition of "supercoarse particles" is applied, for particles that are greater than 10 μm in size [5].

MEASURING SITES

Measurements were performed in four sites. Two locations were close to busy roads, the third one occurred at a distance of 1.6 km from the road No. 81, but still in the urbanized region of Upper Silesia. The last one was far from highly populated and industrial areas. The detailed description of these measurement points is as follows:

- Paderewski housing estate, Katowice. There are many-storied buildings with asbestos cover. The high-way no A4 runs at a distance of over a dozen meters from this measurements point (N 50° 14' 47", E 19° 01' 43").
- Road No. 81, near Woszycze, a town located between Mikołów and Żory. The measurement point was at a distance of several meters from this road (N 50° 04' 28" E 18° 44' 42").
- Jesionka pond (south of Palowice and west of Mikołów). The location was 1.6 km North-West of the previous measurement point at the road No. 81 (N 50° 04' 57", E 18° 43' 35").
- Hills near Raclawice, a little town at a distance of tens of kilometers north of Krakow. This location was far from roads and densely populated areas (N 50° 11' 28", E 19° 41' 39").

MEASUREMENT METHODS

The SMPS and APS particle spectrometers manufactured by TSI (USA) were applied to perform measurements of aerosol size distributions (Fig. 2). The first one has a measuring range from several nanometers to 1000 nm, and the APS device from 540 nm to about 20000 nm. The SMPS spectrometer consists of the 3080 electrostatic classifier, DMA 3081 or DMA 3085 differential mobility analyzer and WCPC 3785 particle counter. An impactor located at the inlet removes any particles larger than its cut-point size and is used additionally to measure the flow rate. The cut-point size of the chosen impactor (orifice diameter of 0.071 cm) was above 1000 nm.

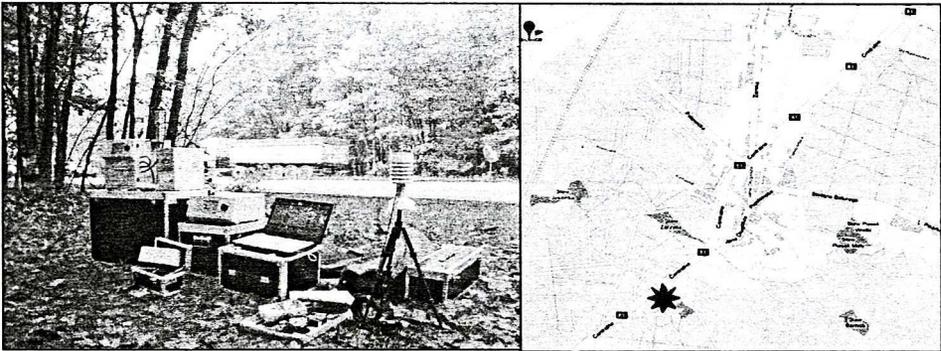


Fig. 2. Measurement site in close proximity to the busy road No. 81 near Woszczyce

According to investigations made by Wiedensohler and Fissan [15], there is no equilibrium of electric charge in ambient air. On the other hand, the measurement relies on separation of particles taking into account their electrical mobility and the non-equilibrium conditions could result in erroneous evaluation of concentration. Therefore, aerosols have to enter a 85Kr Bipolar Charger (Fig. 3), which exposes the aerosol particles to a high concentration of bipolar ions. The particles and ions undergo frequent collisions due to the random thermal motion of the ions and reach a state of equilibrium, in which the particles carry a bipolar charge distribution. Then aerosol particles flow to the Electrostatic Classifier that extracts a known size fraction of particles from the incoming poly-disperse aerosol. A main part of the classifier is the differential mobility analyzer that has an inner cylinder with negative voltage while the outer cylinder is grounded. The electric field causes positively charged particles to be attracted to the negatively charged cylinder and only particles with strictly defined size range are able to reach a small slit located at the bottom of this cylinder and reach the particle counter. This narrow size range depends on electrical mobility of particles, flow rate and Classifier geometry. There are two types of the differential mobility analyzer, the Long DMA (DMA 3081) with measurement range from about 10 nm to 1000 nm and the Nano DMA (DMA 3085) with measurement range from about 2 nm to 150 nm. The voltage of the DMA cylinder can be changed automatically by the condensation particle counter or manually by controls located at the Electrostatic Classifier.

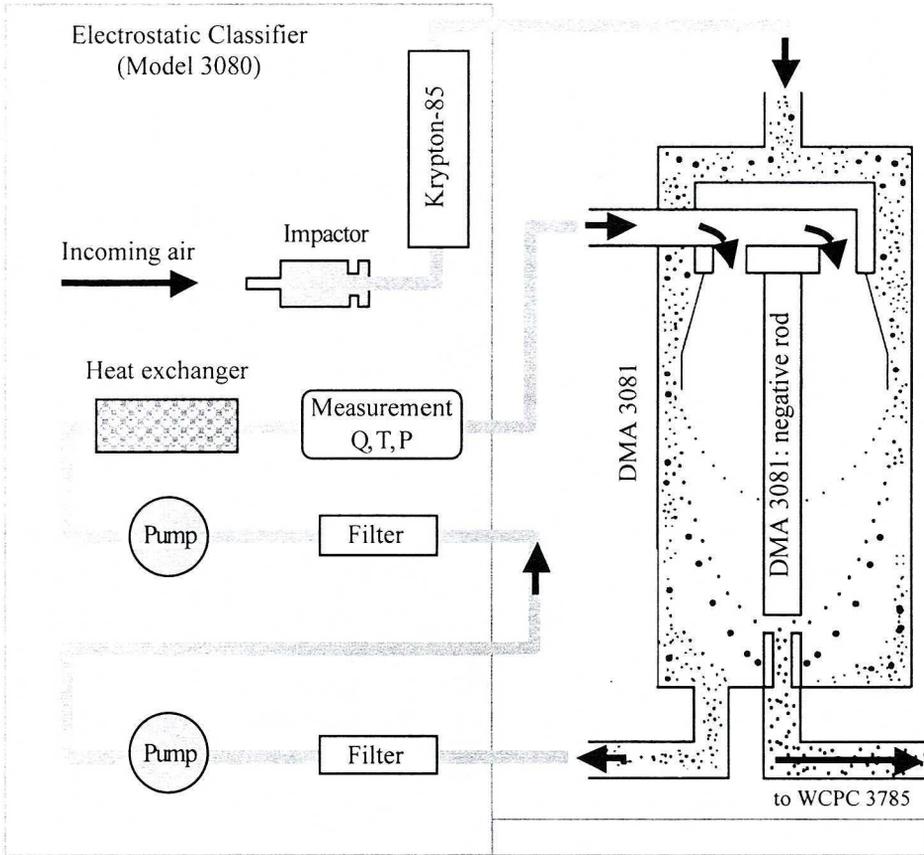


Fig. 3. SMPS spectrometer, classification of submicrometer particles
(Q – flow rate, T –temperature, P – pressure)

It should be mentioned that only a part of incoming sample air (polydisperse flow) is analyzed. The excess air (excess flow) is driven out of DMA, filtered and returned back as so called “sheath flow”. This additional air ensures laminar flow along the cylinder and allows avoiding erroneous size analyzing. The ratio of the sheath flow to polydisperse flow rate on the Electrostatic Classifier is normally set to 10:1. To achieve the best resolution, the sheath flow rate and the excess flow rate must equal each other, and also the polydisperse flow rate and the monodisperse flow rate (air that leaves DMA through the small slit located at the bottom) must also equal each other.

These particles which reach the slit are transferred to a condensation particle counter to determine the particle concentration. Particles are detected optically by laser beam and photodetector that are parts of the particle counter. The detection however is possible when particles have an optically detectable size, typically 2 to 3 micrometers. Therefore, aerosols with fewer diameters have to grow to this size. The mechanism used to grow particles in the particle counter is heterogeneous condensation, whereby particle growth is promoted by the presence of condensation nuclei. This process was supported by distilled water but sometimes butyl alcohol is applied instead. The smallest size of incoming

particles that can be detected this way depends on the ratio of the actual vapor partial pressure and saturation partial pressure. The lower particle detection size, particle diameter at which 50% of particles are detected, is 5 nm for the applied particle counter (3785 WCPC). The upper concentration limit is equal to 20000 particles/cm³ for a single-count mode, when every particle is detected separately, and 10⁷ particles/cm³ for a photometric mode, when the count value, according to special calibration procedure, is calculated based on the concentration of particles measured.

Ultrafine particles are partially caught by each tube which connects spectrometer modules. Therefore, an appropriate correction should be made taking into account the connection length and air flow rate. This correction is possible thanks to the software that governs and supports the measuring process.

The APS spectrometer is a simpler device specifically designed to perform aerodynamic size measurement in real time using low particle accelerations. Aerodynamic diameter is the most important size parameter because it determines a particle's airborne behavior. Time-of-flight particle sizing technology involves measuring the acceleration of aerosol particles in response to the accelerated flow of the sample aerosol through a nozzle. In this case the sheath flow of filtered air is applied as well. This flow is reunited with sample flow at the accelerating orifice nozzle to confine the analyzed particles to the center stream and accelerates the air flow around the particles. As a result small particles, due to lower inertia, reach higher velocity than larger particles. The aerodynamic size of a particle determines its rate of acceleration, with larger particles accelerating more slowly due to increased inertia. As particles exit the nozzle, the time of flight between two laser beams is recorded and converted to aerodynamic diameter using a calibration curve. Information connected to particle detected, is stored to one of 52 channels that the total measurement range is divided by. The measurement range is from 0.5 μm to 20 μm. Smaller particles, 0.3 μm to 0.5 μm, are detected as well but the totalized counts are stored to only one channel.

RESULTS

Chosen statistical characteristics of aerosol size distributions are presented in Tables 1–4. There are count median diameter (M_d), count mean diameter (\bar{d}), aerosol concentration (C), mass median diameter (M_{dm}), mass mean diameter (\bar{d}_m) and aerosol mass concentration (Z). Differences between \bar{d} and \bar{d}_m quantities lie in the weighting mode that is performed in relation to the number of particles or mass of particles:

$$\bar{d} = \frac{\sum_i n_i d_i}{\sum_i n_i} \quad \bar{d}_m = \frac{\sum_i m_i d_i}{\sum_i m_i} \quad (1)$$

where n_i is the number of particles, m_i is mass of particles in the i -th channel with the mid-point diameter of d_i . The channel width depends on the number of channels in the measuring device and its measuring range.

Measurement points in the Paderewski housing estate and at the road No. 81 were located close to places where heavy traffic occurs as opposed to Jesionka pond and hills in the vicinity of Raclawice. There were performed over a dozen of measurements of aerosol size distribution a day. Some statistical characteristics related to these measure-

ments are reported in Tables 1–4 and averaged distributions are illustrated on Figures 4–7. The vertical lines on these plots indicate the measurement range of the SMPS and APS particle spectrometers.

Table 1. Statistical characteristics of aerosol size distribution, the Paderewski housing estate

Quantity	M_d [nm]	\bar{d} [nm]	C [particles/cm ³]	M_{dm} [nm]	\bar{d}_m [nm]	Z [μg/m ³]
SMPS, DMA-3085: 4–100 nm						
Mean	15	28	15297	83	79	0.7
Fluctuation [%]	56	32	91	1	1	58
SMPS, DMA-3081: 102–531 nm						
Mean	161	196	8011	335	337	47.7
Fluctuation [%]	6	4	10	2	1	7
APS: 542–19810 nm						
Mean	626	710	240	965	2660	74.0
Fluctuation [%]	1	1	6	24	18	12
SMPS + APS: 4–19810 nm						
Mean	47	92	23548	626	1740	122.3

Table 2. Statistical characteristics of aerosol size distribution, road No. 81

Quantity	M_d [nm]	\bar{d} [nm]	C [particles/cm ³]	M_{dm} [nm]	\bar{d}_m [nm]	Z [μg/m ³]
SMPS, DMA-3085: 4–100 nm						
Mean	5	12	112266	77	74	1.0
Fluctuation [%]	42	36	81	6	6	29
SMPS, DMA-3081: 102–531 nm						
Mean	172	205	7543	323	335	48.8
Fluctuation [%]	4	3	9	2	1	6
APS: 542–19810 nm						
Mean	583	680	194	965	2706	54.3
Fluctuation [%]	< 1	1	4	39	10	12
SMPS + APS: 4–19810 nm						
Mean	6	25	120002	531	1570	104.0

Table 3. Statistical characteristics of aerosol size distribution, Jesionka pond

Quantity	M_d [nm]	\bar{d} [nm]	C [particles/cm ³]	M_{dm} [nm]	\bar{d}_m [nm]	Z [μg/m ³]
SMPS, DMA-3081: 11–100 nm						
Mean	28	36	8987	69	67	0.6
Fluctuation [%]	6	6	17	1	11	9
SMPS, DMA-3081: 102–531 nm						
Mean	178	217	6709	360	369	56.1
Fluctuation [%]	2	3	1	75		12
APS: 542–19810 nm						
Mean	583	706	141	1114	3106	46.4
Fluctuation [%]	1	< 1	9	20	11	9
SMPS + APS: 4–19810 nm						
Mean	80	123	15837	494	1598	103.1

Table 4. Statistical characteristics of aerosol size distribution, hills in the vicinity of Raclawice

Quantity	M_d [nm]	\bar{d} [nm]	C [particles/cm ³]	M_{dm} [nm]	\bar{d}_m [nm]	Z [μg/m ³]
SMPS, DMA-3085: 4–100 nm						
Mean	54	52	2920	83	83	0.3
Fluctuation [%]	10	10	13	< 1	< 1	3
SMPS, DMA-3081: 102–531 nm						
Mean	262	278	2154	400	402	33.0
Fluctuation [%]	4	3	4	1	< 1	4
APS: 542–19810 nm						
Mean	673	806	201	1486	2732	104.0
Fluctuation [%]	1	3	7	32	33	36
SMPS + APS: 4–19810 nm						
Mean	92	173	5275	1037	2158	137.4

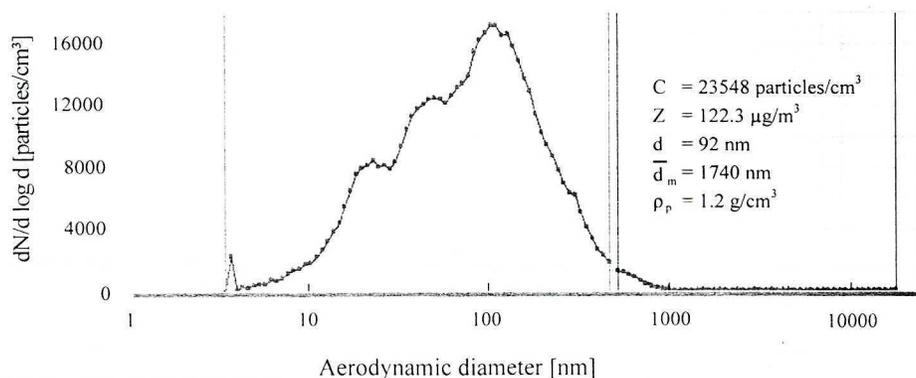


Fig. 4. Averaged aerosol size distribution, the Paderewski housing estate

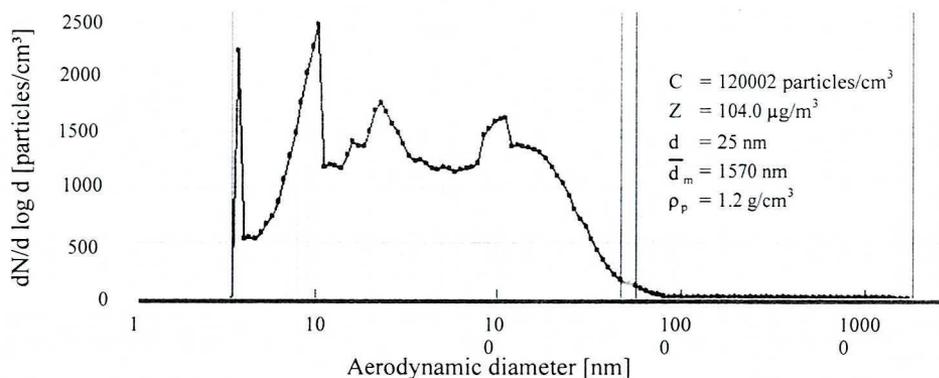


Fig. 5. Averaged aerosol size distribution, road No. 81

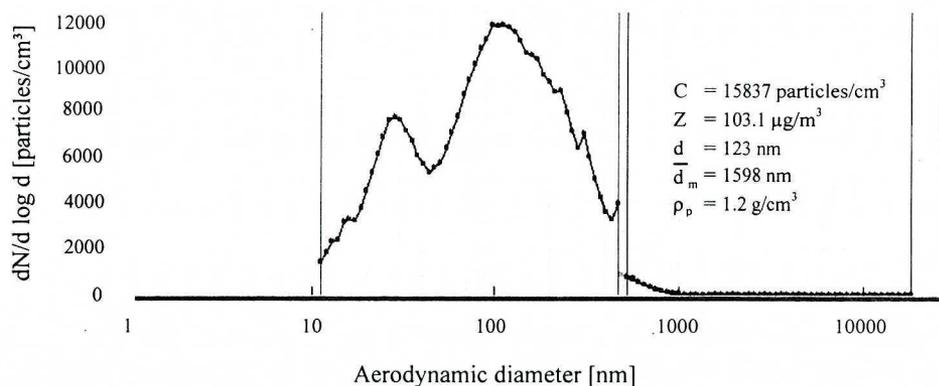


Fig. 6. Averaged aerosol size distribution, Jesionka pond

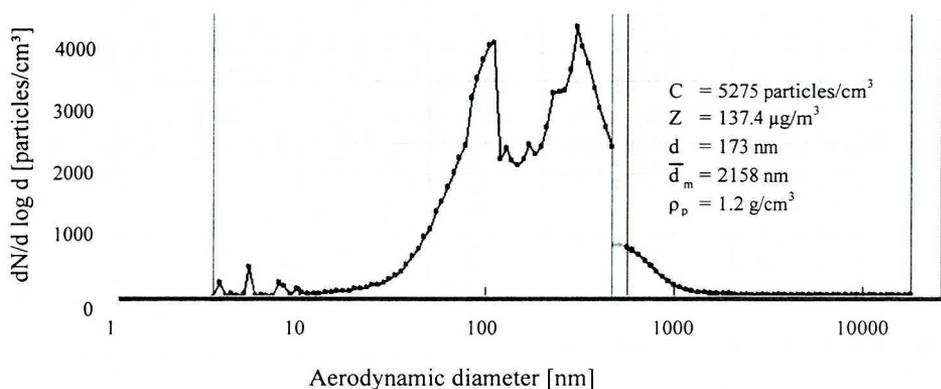


Fig. 7. Averaged aerosol size distribution, hills in the vicinity of Raclawice

Total aerosol mass concentration (liquid and solid aerosols) was equal to $122 \mu\text{g}/\text{m}^3$, $104 \mu\text{g}/\text{m}^3$, $103 \mu\text{g}/\text{m}^3$ and $137 \mu\text{g}/\text{m}^3$ respectively in the Paderewski housing estate, road No. 81, Jesionka pond and hills near to Raclawice. The highest value was measured

therefore far from roads and urbanized areas. It is due to the occurrence of particles greater in size and not by their greater concentration. The count mean diameter and aerosol concentration was for these locations respectively 92 nm, 23548 particles/cm³ and 25 nm, 120002 particles/cm³, and 123 nm, 15837 particles/cm³ and 173 nm, 5275 particles/cm³. Therefore, the aerosol concentration in Woszczyce was 23 times greater than in Raclawice. Simultaneously, the count mean diameter is considerably less in comparison to aerosols which occur in Raclawice and the mass distribution is shifted to lower values. It proves that close to the road mainly ultrafine particles occur.

The measure of fluctuations is a ratio of mean value and standardized deviation (Tabs 1–4). Measurements performed on hills in the vicinity of Raclawice are time-stable. Bigger changes are related only to mass concentration due to fluctuation of concentration of particles with the diameter over 1 µm. On the contrary, close to the road No. 81 strong fluctuations of count diameter and particle concentration correspond to ultrafine particles below 0.1 µm and for diameter over 0.5 µm are less than 5% (Tab. 2). Dynamics of these processes shows that cars produce mainly ultrafine particles and their concentration corresponds to changeable traffic intensity during measurements and a kind of passing vehicles. Such ultrafine particles can be produced by diesel engines as reported by Burtscher [2] and Maricq [8].

There is an interesting problem of risk assessment caused by human activities related to motor traffic. Such evaluation is based often on measurements of PM10 and PM2.5 dust concentration that corresponds to solid particles with diameter up to 10 µm and 2.5 µm respectively. These routine measurements are very important in the existing monitoring system. There is no possibility, however, to measure contribution of both dangerous ultrafine particles and coarse particles with diameter close to 10 µm, and additionally liquid particles are excluded from such measurements. Such procedure can lead to underestimation of hazards especially close to the roads.

Structure of aerosols influences also occurrence of other pollutants in the air. For example, such radionuclides as short-lived radon daughters are caught by ambient aerosols forming radioactive aerosols. Corresponding probability of this process is greater for particles greater in size. On the other hand, contribution of ultrafine particles in the formation of radioactive aerosols increases as their concentration rises. Therefore, there is no simple relationship between radioactive and ambient aerosols. However, in the case of aerosols emitted by cars, the situation is something different. Strong changes of aerosol concentration prove that these hazardous pollutants are generated simply by sources and their interaction with ambient aerosols does not have such critical meaning as for radon daughters.

It is possible in some cases to assess risk connected to interaction of aerosols with different sizes as, for example, for radon daughters (Tab. 5). The recommended conversion coefficient, according to legal requirements [3], corresponds to size of 0.5 µm and nose-breathing rate of 0.75 m³/h. Then its value is equal to 1.1 Sv/(J·h·m⁻³) and mean annual doses can reach 71 µSv (maximal value 178 µSv) assuming 50% of equilibrium and basing on concentration measurements of polonium ²¹⁸Po, 2.62 Bq/m³ (0.024 do 6.59 Bq/m³), performed in open air in Upper Silesia. However, close to busy roads, the aerosol structure can be disturbed and many ultrafine and fine aerosols occur in air resulting in increasing of the conversion coefficient. Taking into account the aerosol size distribution close to the road No. 81 and count mean diameter of 25 nm, the corresponding conver-

sion coefficient is about 10 times greater – $11 \text{ Sv}/(\text{J}\cdot\text{h}\cdot\text{m}^3)$ and the mean annual dose could reach $710 \mu\text{Sv}$ (maximal value $1780 \mu\text{Sv}$) under assumption that count mean sizes of radioactive and ambient aerosols are close to each other. That is slightly less than 1 mSv limit for members of the public [3]. This example shows how risk assessment could change taking into account the aerosol size distribution.

Table 5. Short-lived radon daughters, conversion coefficient and effective doses

Isotope	Conversion coefficient [$\text{Sv}/(\text{J}\cdot\text{h}\cdot\text{m}^3)$]	Annual effective dose [μSv]		
		minimum	maximum	average
^{218}Po	1.1	0.5	178	71
^{218}Po	11.0	5.0	1780	710

Assumptions: equilibrium factor is equal to 50%, annual time to 8760 hours, count mean sizes of radioactive and ambient aerosols are close to each other. The conversion coefficient, $1.1 \text{ Sv}/(\text{J}\cdot\text{h}\cdot\text{m}^3)$ is recommended by legal requirements, and the second one, $11.0 \text{ Sv}/(\text{J}\cdot\text{h}\cdot\text{m}^3)$, corresponds approximately to the count mean size of 25 nm as measured close to the road No. 81 in the vicinity of Woszyczyce; both conversion coefficients were evaluated assuming the nose-breathing rate of $0.75 \text{ m}^3/\text{h}$.

CONCLUSIONS

Performed measurements of aerosol size distributions in Upper Silesia prove that a lot of ultrafine particles can be produced as a result of heavy traffic. Total aerosol mass concentration in the air was comparable for all sites. The highest value was found far from busy roads, in a slightly affected by human activity area close to Raclawice. The aerosol size distribution was very stable there taking into account ultrafine particles. Some changes, however, due to big particles could be observed in relation to both mass mean diameter or mass median and mass concentration. On the contrary, the amount of ultrafine particles, below $0.1 \mu\text{m}$ increased strongly, even by about twenty times, in sites located close to busy roads, while concentrations of larger particles, over $0.5 \mu\text{m}$, were well comparable. Strong fluctuations of count, median diameter and aerosol concentration take place in these locations as well due to ultrafine particles. It proves that ultrafine particles, below $0.1 \mu\text{m}$, occurred mainly close to roads as a result of heavy traffic.

Exposition to the respiratory tracts depends on aerosol penetration ability and then on their size distribution as well. Presently such risk assessment relies on PM_{10} or $\text{PM}_{2.5}$ dust monitoring. The occurrence of big amount of ultrafine particles produced by cars may not result in observable increase of dust concentration and this way the risk can be underestimated. The contribution of ultrafine particles, below $0.1 \mu\text{m}$ to the total mass was in the order of only 1%. Therefore, measurements of size distribution can contribute to appropriate risk assessment. Evaluation of risk caused by short-lived radon daughters can be an example here. The conversion coefficient for particles with diameter of $0.5 \mu\text{m}$ is equal to $1.1 \text{ Sv}/(\text{J}\cdot\text{h}\cdot\text{m}^3)$ but for particles with diameter of 25 nm is ten times greater reaching value of about $11 \text{ Sv}/(\text{J}\cdot\text{h}\cdot\text{m}^3)$ that corresponds to ten times greater dose absorbed by human beings. It is sure that for other pollutants occurring in ground layer of atmospheric air the situation looks like for radon progeny.

There is more and more evidence that some health effects are related to the ultrafine particles because they penetrate cell membranes, blood and brain inducing, for example inheritable mutations [1, 9, 13]. That is the reason why the protection against these particles is getting more and more important.

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POMIARY ROZKŁADÓW ZIARNOWYCH AEROZOLI NA ZURBANIZOWANYCH TERENACH AGLOMERACJI ŚLĄSKIEJ

Depozycją aerozoli w układzie oddechowym zależy od rozkładu ziarnowego aerozoli. Badanie rozkładu ma zatem duże znaczenie dla właściwej oceny zagrożenia spowodowanego przez szkodliwe substancje pojawiające się w środowisku bądź to w następstwie ludzkiej aktywności takiej jak produkcja przemysłowa, komunikacja i paleniska domowe bądź też w wyniku naturalnych procesów. W pracy opisano i oceniono wyniki pilotażowych pomiarów rozkładów ziarnowych aerozoli wykonanych w kilku miejscach zlokalizowanych na terenach zurbanizowanych. Zastosowana aparatura pomiarowa umożliwiła wykonanie badań rozkładów ziarnowych w szerokim zakresie od kilku nanometrów do 20 μm obejmującym praktycznie całą frakcję cząstek respirabilnych. Uzyskane rezultaty wskazują, że w miejscach położonych blisko dróg o dużym natężeniu ruchu pojawiają się duże liczny nanometrycznych aerozoli, których liczba ulega w czasie dużym wahaniom.