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Research paper

Transmission of ultrafine particles through separating systems of dust samplers

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ABSTRACT

In underground mines there is a radiation hazard associated with natural radioactive nuclides. The main sources of radiation exposure in Polish mines are short-lived radon decay products, radioactive mine water containing radium and the sediments precipitated from these waters. For miners, the most common hazard is usually the short-lived radon decay products. Aspirators, equipped with separation systems, are mostly used in order to control this hazard. Inside these aspirators there are meters which measure the radiation emitted by radioactive aerosols collected on the filter. The purpose of these systems is to remove particles from the air stream that do not form the respirable fraction. At the same time, however, a deposition of small-size aerosols takes place in them because of the high values of their corresponding diffusion coefficients. This excludes the possibility of their proper detection by radiation meters. In this paper, the transmission of particles up to 100 nm in size by the separating systems is evaluated. The evaluated transmission ranged from about 60% for sizes of 7 nm, reaching up to 95% at the boundary values of the tested range. The influence of the particle distribution of the aerosols on the radiation calibration coefficients was also investigated in a radon chamber, through their exposure to conditions where the air contained low aerosol concentrations of about 4.0×10^8 particles/m^3 and also when it was nearly 100-times higher. In the first case, the measured sensitivity of the meters was about 20–30% lower, which was probably due to a higher number of small aerosols and, as a result, particle transmission decreased. However, at higher aerosol concentrations, the sensitivity of the meters remained practically the same, regardless of whether the air reached the filters by the separation systems or if they were omitted.

1. Introduction

Among the isotopes naturally occurring in the environment, radon and its short-lived decay products make the largest contribution to the average dose absorbed by humans, reaching up to about 40% percent of the total dose (Jagielak, Biernacka, Henschke, & Sosińska, 1998; UNSCEAR, 2008). In fact, radon itself is not as dangerous as its decay products. However, it is a gas that can be exhaled from soil, rocks or the walls of buildings, initiating the generation of isotopes ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po in the air with a much shorter half-life than the radon itself. Compared to radon, the contribution these isotopes make to the dose absorbed in respiratory tissues is incomparably greater. Under radioactive equilibrium, radon makes up only about 2% of the dose resulting from the exposure to short-lived radon decay products (Jacobi & Eisfeld, 1980).

Short-lived radon progeny are metals that quickly combine with molecules of water vapour and gases to create clusters. Free atoms and the clusters formed comprise the so-called unattached fraction. Measurements indicate that this fraction consists of a clearly separated medium-sized formation of 1.2 nm particles among the whole activity size distribution (Reineking & Porstendörfer, 1990).

If there are ambient aerosols in the air, the particles of the unattached fraction will combine with them, forming the so-called radioactive aerosols (attached fraction) whose particle size distribution depends on the particle size of the ambient aerosols (Porstendörfer, 1994; Skubacz, Wojtecki, & Urban, 2016a). With higher concentrations of ambient aerosols, the contribution of this unattached fraction is significantly reduced (Porstendörfer & Reineking, 1996).

The hazards related to short-lived radon decay products are not only influenced by their concentration but also by the structure of the particle size distribution. Small nanometer-sized radioactive aerosols are many times more dangerous than aerosols measuring several hundred nanometers in size (Heyder, Gebhart, & Stahlhofen, 1980; ICRP, 1994; Zock, Porstendörfer; Reineking, 1996), as particles of such sizes are deposited more efficiently in the respiratory system, and they are also rapidly transported to the central nervous system (Oberdörster et al., 2004).

By recognising the hazards related to short-lived radon progeny in

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the air, in many cases monitoring their concentration has become mandatory. For instance, in the underground mines of Poland, this issue is currently regulated by the Minister of Energy (Regulation, 2016). It is also considered in the Council Directive, 2013/59/EURATOM of 5 December 2013, which lays down the basic safety standards for protection against the dangers arising from exposure to ionising radiation (Council Directive, 2013), and is more stringent in comparison with the previous legislation. The implementation of this new directive will force changes to be implemented in the national Atomic Law Act (2000), especially with respect to radon hazards occurring in underground workplaces (mines and caves) and in buildings. In addition, residential buildings will be adequately protected by the law.

The measure of exposure to short-lived radon decay products is the total energy of alpha radiation that can be released by short-lived radon progeny occurring in a defined unit of volume of air as a result of their complete decay, or the so-called "potential alpha energy concentration" (PAEC). According to the legal requirements in underground mines, systematic control of the PAEC must be carried out. For economic and organisational reasons, radiation detection meters have been developed as additional modules to dust samplers or aspirators, in order to meet the appropriate safety regulations. These modules are called alpha probes, and are located inside separation systems that are made in the form of cyclone units.

The purpose of the cyclone units is to remove larger aerosols from the airstream, which do not belong to the respiratory class. As a result, only the respirable fraction including particles that enter the non-ciliated respiratory tract during breathing (ISO, 1993) is analyzed. However, some of them, especially the small ones with high diffusion coefficients, will be removed from the air stream by their deposition on the inner surfaces of the inlet system before the filter, above which the alpha probes with thermoluminescent detectors are located. These can be defined as losses, because such particles will not be registered by the detection system. In this paper, measurements are presented of the transmission of ultra-fine particles up to 100 nm by the separator systems of two dust samplers – Barbara-3A and AP-2000EX – which are commonly used in underground mines to measure the PAEC. In this case, the transmission is the ratio of the number of particles that are not deposited in the separation system to the number of particles reaching the inlet of the system.

2. Measurement methods

2.1. Dust samplers and alpha probes

Alpha probes (Fig. 1) with thermoluminescent detectors (TLD) are located above the filter that intercepts the respirable particles passing through the cyclone. The TLDs are placed in three sockets and record the radiation emitted by short-lived radon progeny. Each socket contains two TLDs: the first, placed just above the filter, records the alpha, beta and gamma radiation; whereas the second, separated by a thin spacer, can only register the beta and gamma radiation. These TLDs, with a diameter of 6 mm, have a $CaSO_4$:Dy luminophore layer of 0.1 mm. This layer is attached to an aluminium base. The outcomes received as a result of the readouts from these TLDs are proportional to the radiation energy they have absorbed; therefore, they make it possible to evaluate the average PAEC of the short-lived radon progeny over quite a long time period, 8 h (Skubacz & Bywalec, 2003). The lower limit of the detection for that time period is equal to 0.004 μJ/m^3 at a 5% significance level, and is 0.04 μJ/m^3 for a 1 h measurement.

The alpha probes are used in combination with Barbara-3A or AP-2000EX dust samplers (Two-Met, Zgierz). The Barbara-3A is a relatively heavy battery-powered device that should provide continuous operations for at least 8 h. It has intrinsically safe housing, as well as a built-in rotameter that enables control of the air flow during the measurement and its manual correction. The air flow rate is nominally 5 dm^3/min, which ensures the proper separation of the respiratory fraction by the cyclone. The AP 2000EX aspirator is a modern device that is controlled by a microprocessor. The air flow rate can be read on a liquid crystal display and its regulation is possible, in the range 0.7–2.3 dm^3/min. The flow rate of 1.9 dm^3/min, however, ensures the proper separation of the particles travelling through the cyclone.

2.2. Particle spectrometers

The SMPS and APS are complementary spectrometers (TSI and USA) used to determine the distribution of particle concentration in different size ranges. The SMPS spectrometer (Fig. 2), consisting of the Electrostatic Classifier 3080 (EC), the Differential Mobility Analyzer 3081 or 3085 (Long DMA or Nano DMA), and the Condensation Particle Counter 3785 (CPC), was used for the study of the particle size distribution. An impactor of 0.0508 cm in diameter was mounted onto the air inlet of the classifier and the air flowed through the spectrometer with a flow rate of 0.6 dm^3/min (Nano DMA and Long DMA). For such parameters, the measurement sizes ranged from 3 nm to 106 nm for the Nano DMA and from 10 nm to 414 nm for the Long DMA (Table 1). The obtained results were corrected with regard to the deposition of particles inside the measuring system. This correction was performed using internal software based on the current air temperature, atmospheric pressure, and assuming that the lengths of the tube connections between the modules of the spectrometer were in accordance with the manual.

The measuring range in the SMPS spectrometers is divided into slightly over 100 channels. After passing the impactor, the particles are charged in a column containing air ionised by a beta radiation source ^{85}Kr. Subsequently, the particles are separated in the differential analyzer (DMA) by size and are registered by the optical method into the measuring channels of the particle counter (CPC). The measuring channels are referred to as mobility diameters, as the differences in their mobility in the electrostatic field are used during the separation process. The method is described more precisely in a paper by Skubacz (2009).

The APS spectrometer (Fig. 2) operates in the range of aerodynamic diameters ranging from about 0.5 μm to about 20 μm (Table 1). The whole measuring range of the device is divided into 52 channels, and the estimation of particle diameter is based on the particle travel time measured by two laser beams and a comparison of the indications with the calibration curve.

The measuring range of the SMPS spectrometer depends on the parameters that are chosen, and in particular, on the flow rate of the incoming air. Increasing the flow rate shifts the measuring range towards the smaller particles and decreases the range towards the larger sizes. However, because in this case the ultrafine particles were the main area of interest, it was a higher air flow rate through the measurement system, which reduced the losses caused by the diffusion of the particles and shifted the range towards the smaller particles, was chosen. As a result, however, the ranges of the SMPS and APS spectrometers did not coincide: the SMPS had a measurement size range up to 414 nm and the APS from 500 nm.

The aerosol concentration, measured by the SMPS spectrometer, is a function of the mobility diameter while, in the case of the APS spectrometer, it is a function of the aerodynamic diameter. The aerodynamic diameter that corresponds to an ambient particle is defined as the diameter of a spherical particle with a unit density of 1 g/cm^3 and the same settling velocity as the particle. In the study, it was assumed that the dynamic shape factor was equal to 1.0, and the specific density of 1.0 g/cm^3 (water based aerosols) was used to unify the two sets of size distribution measurements. This method is described in detail in an article by Skubacz et al. (2016a).

2.3. Radon chamber

During the tests, the response of the alpha probes was tested by

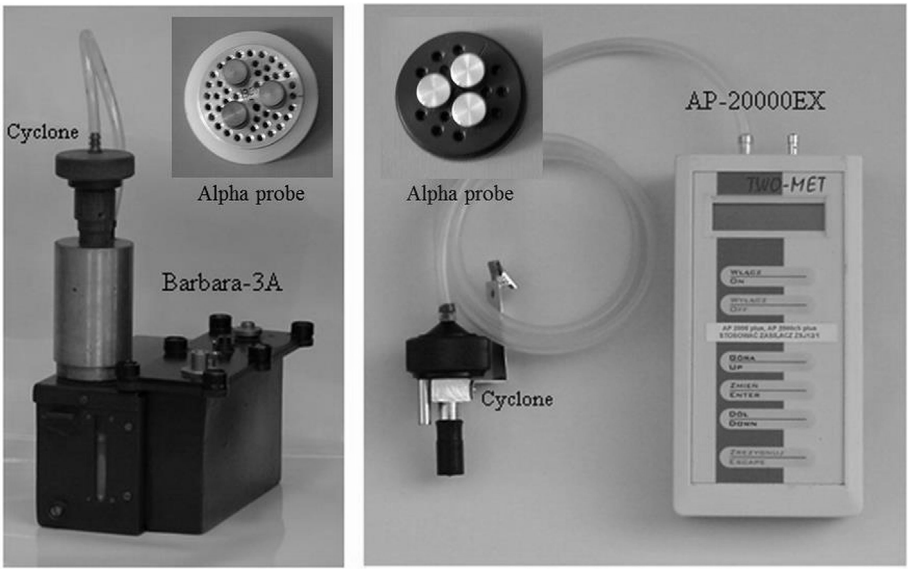

Fig. 1. Barbara-3A dust sampler and AP-2000EX aspirator with cyclones and alpha probes.

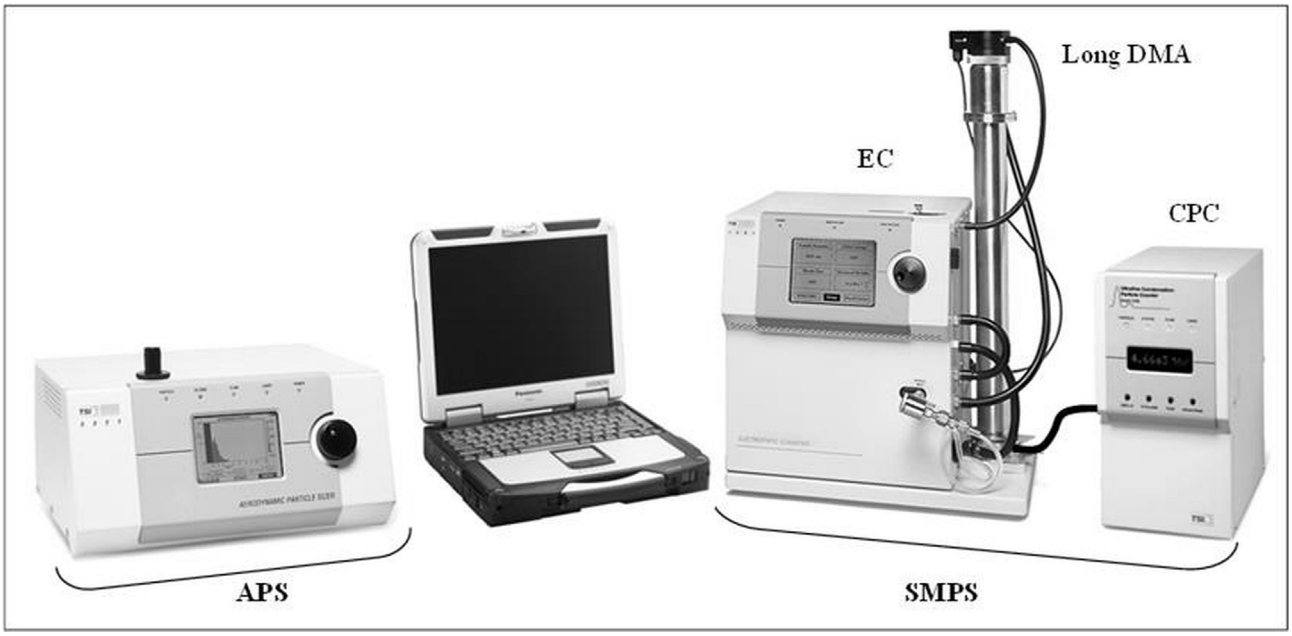

Fig. 2. APS and SMPS particle spectrometers with the Long DMA module.

Table 1
Measurement ranges for the SMPS particle spectrometer.

Particle spectrometer	Lower boundary (nm)	Upper boundary (nm)
SMPS with the Nano DMA module	3.2	105.5
SMPS with the Long DMA module	9.8	414.2
APS	542.0	19810.0

exposing them to both an atmosphere which was depleted and one that was enriched in aerosols. The measurements were made in a radon chamber with a volume of about 17 m^3 and with a radon source placed inside it. Temperatures ranging from −20 °C to +60 °C and a relative humidity from 20 to 90% could be controlled in the chamber. The chamber was additionally equipped with a man lock, material lock and a viewfinder (Fig. 3). The aerosols in the chamber were filtered using a Dwarf-100 × 9 dust collector (Seneya, Finland) or were generated by a Thomex-MB ultrasonic inhaler (Medbryt, Poland) from a 1% NaCl solution.

The reference PAEC inside the chamber was measured according to the method described by Chałupnik, Skubacz and Lebecka (1987). The method is based on a liquid scintillation technique, in which measurements of the alpha and beta particles with an efficiency close to 100% are possible. The quenching effect of a filter in the scintillator can be the most serious problem with this method, but the membrane filter that is used becomes transparent in a toluene based liquid scintillator. During the measurement, the air is pumped through the open-face filter. The filter is then immersed in toluene and its activity is counted during three subsequent time periods, to enable calculations of the radon decay product concentrations as well as the PAEC. The portable Triathler (Hidex Company, Finland) monitor was used to measure the activity of the radon progeny collected on the membrane filter.

Both the aerosol size distribution and the PAEC were controlled

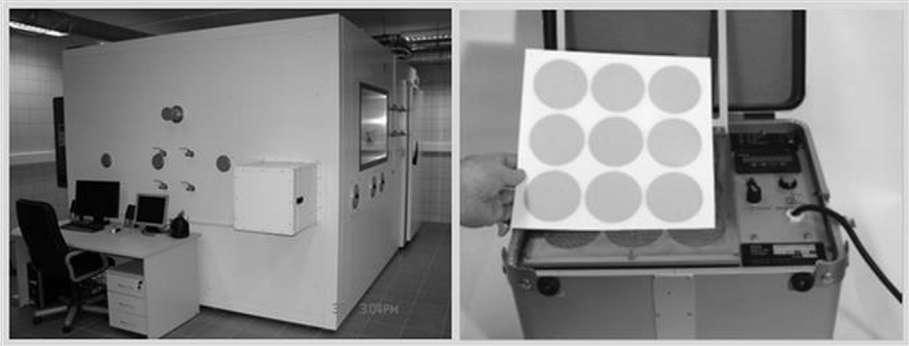

Fig. 3. From the left: Radon chamber and the Dwarf-100 × 9 dust collector (Seneya, Finland).

during the operation of each of the alpha probes. The temperature inside the chamber was in the range of 21–26 °C, while the atmospheric pressure was within 990–1000 mbar and the relative humidity was approximately 40%.

3. Study of the transmission of ultrafine particles by the cyclones

The measurement system used to study the ultra-fine particle transmission through the cyclones is shown in Fig. 4. It was designed to provide an adequate air flow rate through the cyclones of the Barbara-3A dust sampler and the AP-2000EX aspirator, which should be 5 dm^3/min and 1.9 dm^3/min, respectively. These flow rates are higher than that of the air flowing into the SMPS spectrometer. Therefore, an additional pump and a rotameter equipped with a control valve were used to adjust their appropriate values. The rotameter performed only control and stabilisation functions. The actual flow rates in the air stream entering the cyclone and the SMPS spectrometer were measured with a TSI 4140D calibrated flowmeter. After adjustment of the system, so that the cyclone flow rate was 1.9 dm^3/min in the AP-2000EX Two-Met aspirator cyclone or 5.0 dm^3/min for the Barbara-3A dust sampler and 0.6 dm^3/min for the SMPS spectrometer, the values indicated by the rotameter and a built-in flowmeter in the SMPS spectrometer were checked. During the measurement, the TSI 4140D flowmeter was not part of the measuring system, because it could have removed the aerosols. The correct flow was then maintained by adjusting the valve of the rotameter and the valve of the SMPS spectrometer. All of the additional connections in the measuring system were made from antistatic tubing and metal connectors to avoid any losses caused by static electricity.

The aerosol losses caused by diffusion along the distance between the air outlet from the cyclone and the supply to the SMPS spectrometer (Section A-B-C in Fig. 4) were estimated based on the measured length of the connections and the flow rate values in the individual sections. In the examined range, the losses in these additional connections reached a maximum of 20% for the lower measurement limit of 3.22 nm. However, the evaluation of the aerosol transmission of the cyclones was performed in the range from 7 nm to 100 nm. Therefore, the smaller sized particle concentrations were too small and the fluctuations of the concentration were very high to perform statistical analysis of the results. For the aerosols that were 7 nm in size, the losses due to deposition in the additional connections did not exceed 6%, taking into account the formula for the transmission of ultrafine particles through a cylindrical tube (Hinds, 1999).

During the measurements, the concentration of ultrafine particles in the air entering the measuring system and after passing through the cyclones was continuously and simultaneously controlled by two coupled SMPS spectrometers. The concentration of the ultrafine particles in the air flowing into the cyclones is shown in Fig. 5.

A total of several hundred measurements were made for each spectrometric channel. The results of the transmission measurements through the cyclones are shown in Figs. 6 and 7. In both cases, the shape of the curve is relatively similar. In the lowest size ranges, a larger decrease of the transmission was noticeable for the AP-2000EX aspirator than for the Barbara-3A dust sampler, which was probably due to the differences in the flow rates. The flow rate of the aspirator was considerably smaller, although the passage diameters were similar.

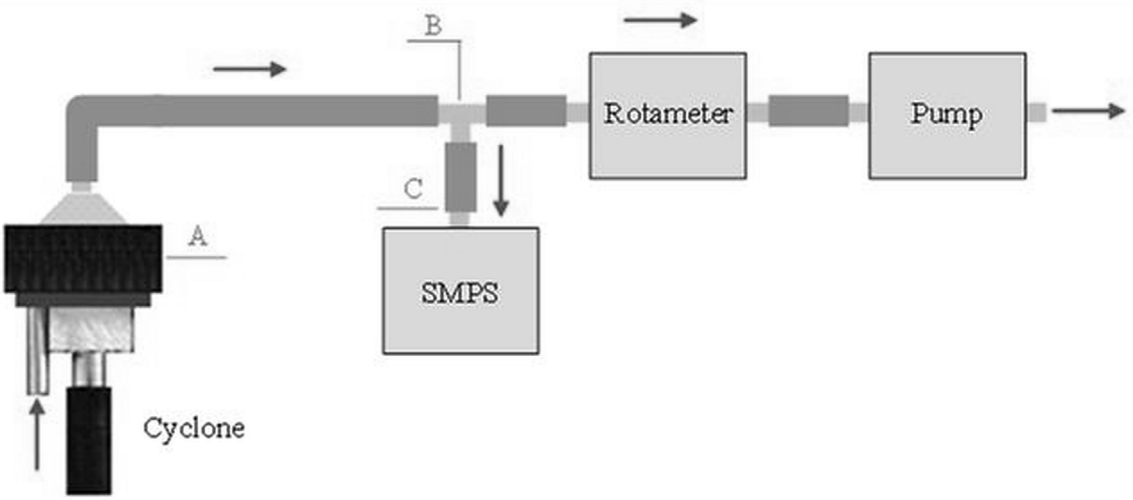

Fig. 4. Measurement of the aerosol transmissions through the cyclones of the AP-2000EX aspirator and Barbara-3A dust sampler using an SMPS spectrometer equipped with a Nano DMA module.

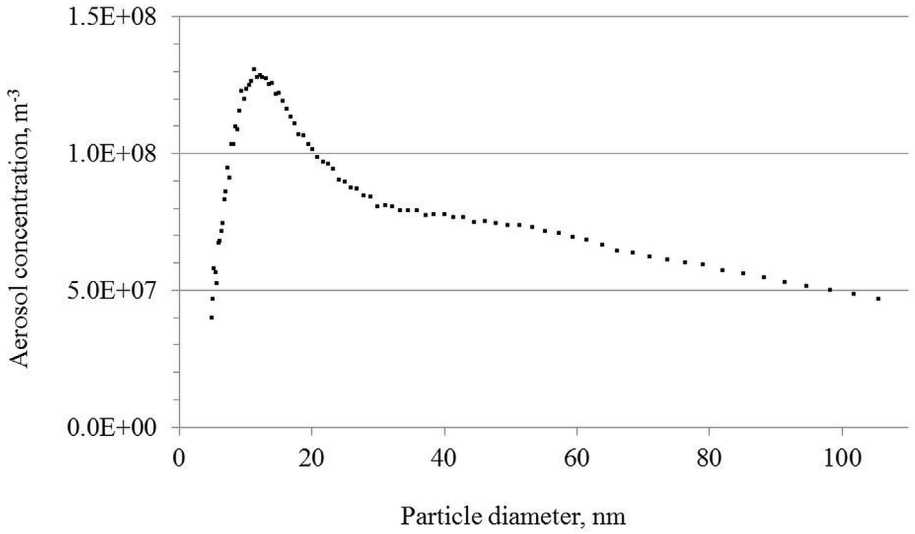

Fig. 5. Concentration of ultrafine particles in the air flowing into the cyclones.

As a result, the velocity of the stream flow through the cyclone of Barbara-3A was higher, which resulted in a reduction in deposition due to diffusion. For the 7 nm size, the particle transmission by the cyclone of the Barbara-3A dust sampler was about 65% and for the AP-2000EX aspirator cyclone it was lower than 60%. For larger particles, the transmission values in both cases were very similar, reaching 93% and close to the upper boundary of the size measurement range.

The uncertainties of the measurements of the transmission coefficients are shown in Figs. 6 and 7 as error bars. The transmission through the cyclones can be approximated by polynomial functions. The correlation coefficients – 0.915 and 0.967 for the Barbara-3A and the AP-2000EX, respectively – indicate a high correlation between these curves and the experimental data.

4. Influence of particle size distributions on the calibration coefficients

The effect of the particle sizes on their transmission value indicates that the particle size distribution can change the value of the calibration coefficients of devices designed to assess the hazards related to short-lived radon progeny. The calibration coefficients are expressed as the number of net pulses received after reading the thermoluminescent detectors that are located inside the alpha probes in relation to the reference PAEC. The values defined in this way are a measure of the sensitivity of the detection system. Measurements of the calibration coefficients for the alpha probes were carried out in two extremely different sets of conditions: when the radon chamber was cleaned of aerosols (Fig. 8, Table 2); and when the aerosols generated by the ultrasonic inhaler were directed to the chamber (Fig. 9, Table 2). Both the AP-2000EX (Probe 1, Probe 2, Probe 3, Probe 4) and the Barbara-3A (Probe 5, Probe 6) were tested inside the radon chamber during aerosol generation with and without the cyclone (calibration coefficients k_o and k_1, respectively), and also in the depleted atmosphere with the cyclone (calibration coefficient k_2). The calibration coefficients were evaluated with uncertainty of 5% at confidence level of 95%. However, in all the experiments, the same alpha probes were subsequently applied to achieve k_o, k_1, and k_2 calibration factors. The relative values of the calibration coefficients are compared in Table 3.

The measurements of the size distribution of the ambient aerosols was performed through the use of SMPS and APS particle spectrometers. The particle size distribution of the radioactive aerosols is dependent on the particle size distribution of the ambient aerosols. It is

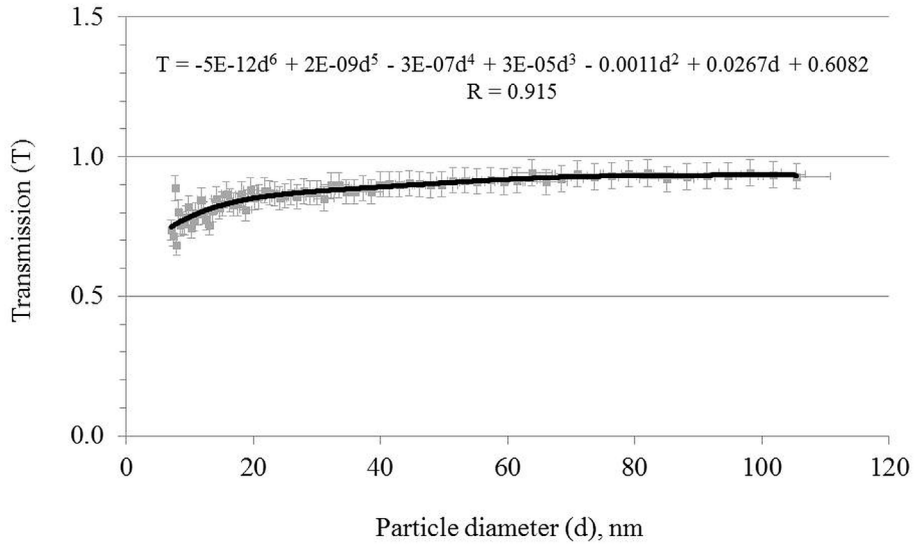

Fig. 6. Aerosol transmission by the cyclone of the Barbara-3A dust sampler.

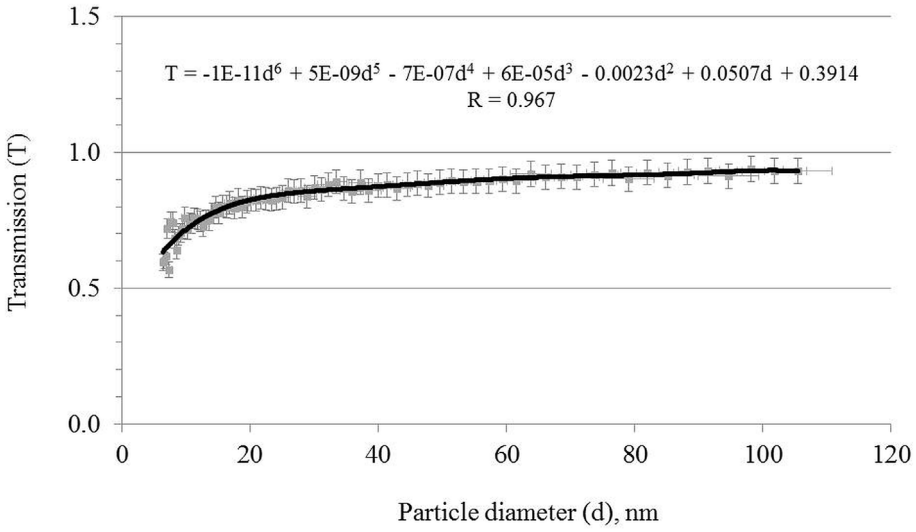

Fig. 7. Aerosol transmission by the cyclone of the AP-2000EX aspirator.

not possible to evaluate the concentration of radioactive aerosols based on the concentration of the ambient aerosols, which is why the distributions are presented in Figs. 8 and 9. However, the distributions defined as the concentrations of the aerosols in a given size range compared to the total concentration are a good representation of the conditions in the ambient air and the mutual relationship between the ambient and radioactive aerosols. The distribution of the radioactive aerosols was calculated according to the method described by Skubacz et al. (2016a).

By comparing the two distributions, it is clearly visible that during aerosol generation, maximum distribution is shifted towards the larger sized aerosols, which, given the transmission estimated for the ultrafine particles, reduces the loss. However, in the aerosol-depleted atmosphere, the contribution of the ultra-fine particles increases considerably: by four times for the environmental aerosols and five times for the radioactive aerosols, at the same time, the average diameters decrease. This is evident in the surface weighted averages that were evaluated by taking into account the weight equal to the surface of the particles for a given size channel divided by the total surface of the particles for the whole measurement range. The surface weighted average diameter was about 340 nm in the depleted atmosphere, while during the generation of the aerosols it was 1500 nm. It can also be observed that the average diameters of the radioactive aerosols were higher when compared to the ambient aerosols. This is due to the mechanism of attachment of the radioactive isotopes to airborne

Table 2
Chosen statistical parameters of aerosols in the depleted atmosphere and during aerosol generation.

Aerosols	Particle concentration	Contribution of ultrafine particles to the total concentration in the range of 3 nm–19810 nm		Particle diameter weight: particle concentration(*)	
		Ambient	Radioactive	Ambient	Radioactive
	(m^{-3})	(%)	(%)	(nm)	(nm)
Depleted atmosphere	4.0×10^8	49	32	123	155
Aerosols generation	322.0×10^8	12	6	274	293

(*) The average diameter was evaluated taking into account weight being equal to the concentration of particles for a channel of a given size divided by the total concentration of particles of the whole measurement range.

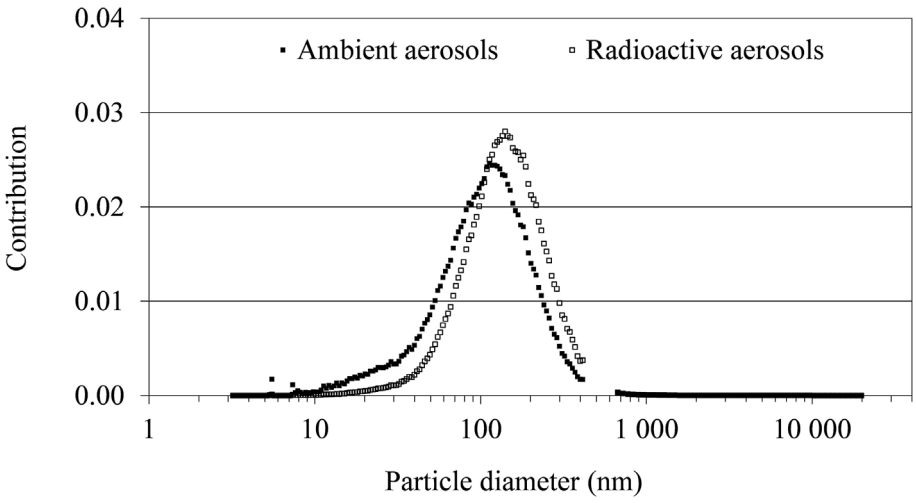

Fig. 8. Particle size distribution in the depleted atmosphere of the radon chamber.

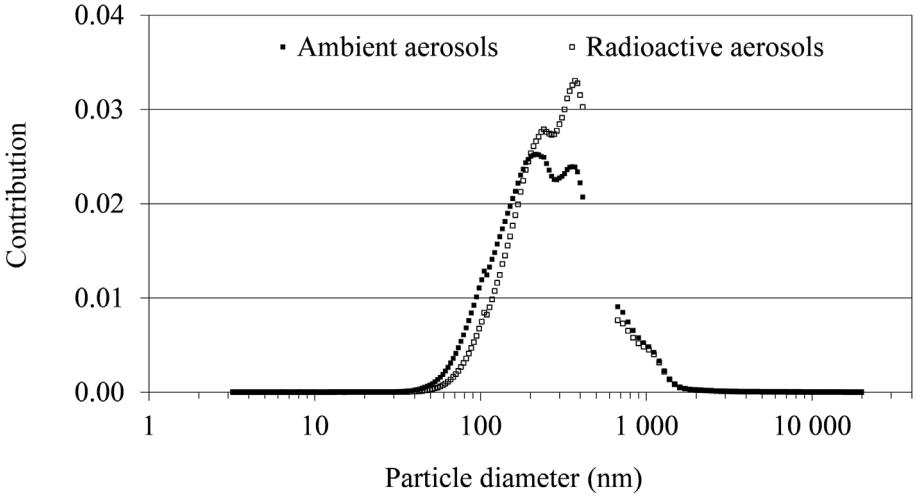

Fig. 9. Particle size distribution during aerosol generation by means of an ultrasonic inhaler.

Table 3
Comparison of the calibration coefficients determined during aerosol generation for alpha probes with and without a cyclone (k_o, k_1 respectively) and aerosol-depleted atmosphere for alpha probes with a cyclone (k_2).

Alpha probe	Cyclone			k_1/k_o	k_2/k_o	Concentration of ambient aerosols (m^{-3})	
	k_o	k_1	k_2			Aerosols generation (k_o, k_1)	Depleted atmosphere (k_2)
Probe 1	No	Yes	Yes	1.04	0.90	4.0×10^{10}	4.8×10^{8}
Probe 2	No	Yes	Yes	0.96	0.79	4.8×10^{10}	4.5×10^{8}
Probe 3	No	Yes	Yes	1.04	0.66	4.2×10^{10}	4.7×10^{8}
Probe 4	No	Yes	Yes	1.02	0.66	2.9×10^{10}	4.6×10^{8}
Probe 5	No	Yes	Yes	0.97	0.74	2.8×10^{10}	3.8×10^{8}
Probe 6	No	Yes	Yes	0.86	0.55	3.1×10^{10}	3.7×10^{8}
Mean				0.98 ± 0.06	0.72 ± 0.11		

particles. The attachment of the radioactive isotopes to ambient aerosols is increased due to an increase in the ambient aerosol concentration and the particle surface (diameter). If the size of the radioactive aerosols increases, it generally means that the number of smaller particles (which is usually higher) did not offset the impact of the less numerous, larger particles on the entire process, resulting in them forming a greater area.

In the situations where the alpha probes were exposed to an aerosol-rich atmosphere (k_o, k_1), there was no clear difference between the configuration in which the cyclone was absent (k_o) and that where the air reached the alpha probe via a cyclone (k_1). The relative ratio of the calibration coefficients was 0.98 ± 0.06. However, this was different in the case where the air reached the alpha probes through the separation system and the aerosol concentration was relatively small (k_2). The average relative decrease in sensitivity was then about 30% (0.72 ± 0.11). With the decreasing concentration of larger environmental aerosols, there was an increase in the concentration of the radioactive aerosols of smaller sizes, including the unattached fraction, formed by particles of an average size of 1.2 nm (Porstendörfer, 1994; Reineking & Porstendörfer, 1990). With such particle size distribution, the particle transmission through the separation system decreased and the sensitivity of the measuring system was reduced.

In the case of the measurements performed in the radon chamber, the aim was only to show that in an atmosphere with a high concentration of small aerosols, regardless of the unattached fraction, the calibration coefficients will differ significantly from those determined for the high concentration of aerosols in which the calibration should be performed. For high concentrations of aerosols, the unattached fraction is of minor importance. It can be assessed based on the equation $414/Z$ [cm^{-3}], as reported by Reineking and Porstendörfer (1990), where Z is the total concentration of aerosols. According to this formula, the unattached fraction is much less than 2% for the high particle concentrations and its impact on the calibration factors can be overlooked.

5. Summary

Measurements of the particle size distribution of aerosols in underground mines (Skubacz, Wojtecki and Urban, 2016a, 2016b) show a significant presence of ultrafine particles, even in the vicinity of the rock mining machines. Their average contribution is around 60% of the total concentration; however, near the places of coal extraction it can reach 90%. This has not been without consequence for the size distribution of the radioactive aerosols containing short-lived radon decay products and the responses of the measuring instruments equipped with separation systems. The losses caused by the separation systems for particles of approximately 7 nm were greater than 40% for the AP-2000Ex aspirator; whereas they were approximately 30% for the Barbara-3A dust sampler. For particles of sizes close to 100 nm, the result was about 5–7% in both cases. This undoubtedly leads to the underestimation of the PAEC value when measurements take place in a low-aerosol concentration atmosphere, because not all the small radioactive aerosols will reach a filter and be detected by the TLDs. Therefore, it seems that the most appropriate procedure is to determine the calibration coefficient of such meters in an atmosphere with a relatively high concentration of aerosols (3–4 × 10^{10} m^{-3}) belonging to the respirable particle class, and to apply corrections if the measurements performed in low aerosol concentrations and their particle size distribution, or at least the concentration, is roughly known, or to perform a cyclone-free measurement in cases where it is reasonable to assume that the atmosphere does not contain significant amounts of non-respirable aerosols consisting of larger particles.

Conflict of interest

None declared

Ethical statement

The research was conducted according to ethical standards.

Funding body

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None.

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