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Research paper

Size distribution of ambient and radioactive aerosols formed by the short-lived radon progeny

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ABSTRACT

The survey of ambient airborne particle size distribution is important when the deposition of radioactive particles is considered in the human lung and the assessment of radiation hazard in occupational exposures or contaminated environments. CLOR (the Central Laboratory for Radiological Protection) in cooperation with CMI (the Central Mining Institute) performed simultaneous measurements of the activity size distribution of radon progeny and ambient aerosols using different types of aerosols. Measurements were performed in a radon chamber with a volume of 17 m^3, where radon was generated by a radium-226 open source, and ambient aerosols by an oil candle, vax candle, and incense sticks. Such measurements were also made in an aerosol depleted atmosphere after cleaning the chamber air by means of a high-efficiency pump and filters. The size distribution of radioactive aerosols containing radon progeny was measured by RPPSS (Radon Progeny Particle Size Spectrometer) with the measuring size ranging from 0.6 nm to approximately 2500 nm. The key parts of this device are the impaction plates and diffusion screen batteries, which collect aerosols of different sizes, and semiconductor alpha detectors which detect the activity of the collected particles. The SMPS (Scanning Mobility Particle Sizer Spectrometer) and APS (Aerodynamic Particle Sizer) were applied to evaluate the size distribution of all aerosols with sizes from approximately 3 nm to 20 µm. Based on the results obtained by these spectrometers, the activity size distributions and related dose conversion factors (DCF) were evaluated both for the exposed workers and the general population.

1. Introduction

According to an evaluation made by the Polish Atomic Agency, each member of the public in Poland received an average dose of 3.56 mSv in 2017, including natural and artificial ionizing radiation sources, also those applied in the medical diagnostics (Annual Report, 2017). The majority of this dose, approximately 34%, is caused by radon and its progeny. Miners are a group of workers that are one of the most exposed to radon hazards. The reason is the increased exhalation of gaseous radon from shredded material, poor ventilation or the ineffective organization of ventilation, a mining system favouring the penetration of air through crushed rock and the transportation of radon and its progeny to mine underground workings. The effects of such exposure have been observed for an extended period of time, and the relationship between exposure to radon and its progeny and an increased incidence of lung cancer has been suspected for almost 100 years (Holaday, 1969).

Radon's progeny such as polonium-218, lead-218, bismuth-214, and polonium-214 provide much more contribution to the dose than radon. They are, in contrast to radon, solid particles, which can be efficiently deposited in the airways. Their half-lives are less than 30 min. After radon decay, they attach very quickly (within much less than 1 s) to trace gases and molecules of water vapour. These clusters have a diameter from 0.5 to 5 nm and are called “unattached fraction”. At the beginning they consist mainly of polonium-218 radionuclide as illustrated in Fig. 1 (Porstendörfer, 1994). These clusters are then combined with ambient aerosols forming so called radioactive aerosols or attached fractions. Usually the sizes of radioactive aerosols are within the range of 50–500 nm. However, when the concentration of ambient aerosols is relatively low, their mean diameter shifts towards the lower sizes.

The dose to the respiratory tracts caused by radon progeny strongly depends on the aerosol size distribution, manner of breathing and breathing rate. Physical deposition processes are associated with mechanisms such as the force of inertia, sedimentation and diffusion. According to Heyder, Gebhart, and Stahlhofen (1980) the diffusion

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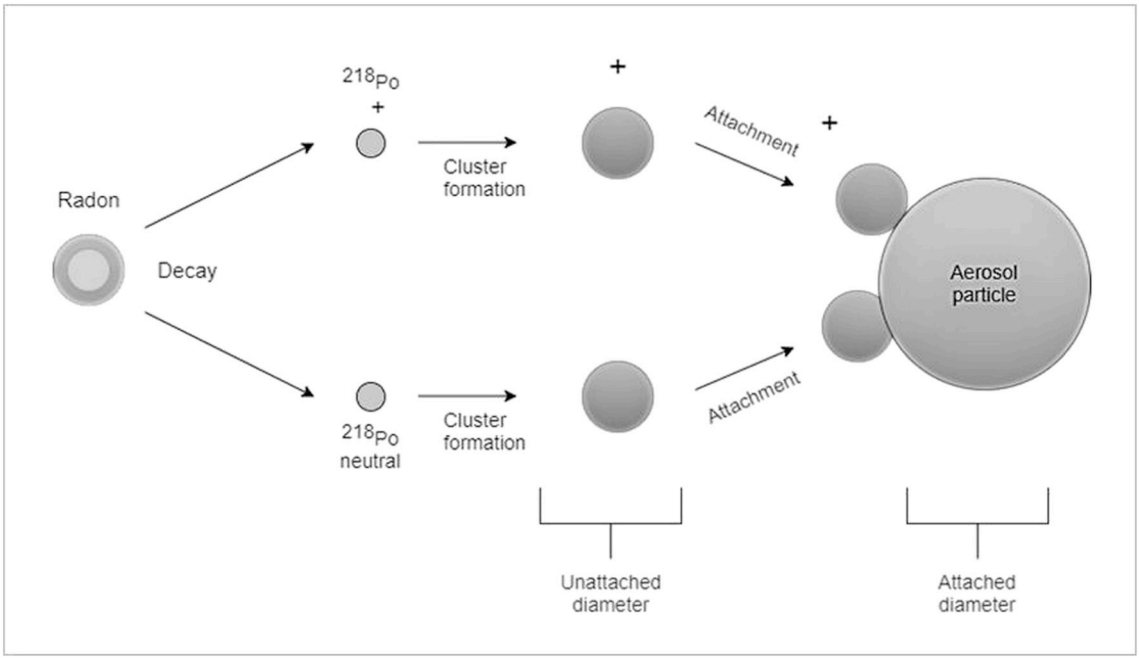

Fig. 1. Basic processes of short-lived radon progeny generation in air defining the “unattached” and “attached” fraction.

mechanism of deposition becomes dominant for particles smaller than 100 nm. Gravitational sedimentation comes to the fore for larger particles up to 4 μm with longer retention time, and the forces of inertia play the most important role for aerosols greater than 4 μm. The dose also depends on the manner of breathing, and is higher for mouth breathing in comparison to nose breathing at the same breathing rate. At the same time higher ventilation rate also results in an increased dose to the respiratory tracts.

Particles with size below 100 nm are called ultrafine particles. They are characterized by a high value of diffusion coefficient and dose conversion factors (DCF). These conversion coefficients are much higher in relation to particles of several hundred nanometers in size. Also the DCFs to coarse fractions of aerosols, defined as a class consisting of particles with sizes from 2.5 μm to 10 μm, reach very high values close to their upper boundary range. However, their concentration in the air is usually not significant because of gravitational sedimentation. The shape and size of the attached fraction strongly depend on the type of the ambient aerosols.

The term “ambient aerosols” used in this article corresponds to all aerosols that occurred inside the radon chamber as a result of their generation and natural processes.

The significant differences in the conversion factors, depending on the aerosol size, makes the determination of the size distribution crucial for the proper estimation of the dose. There are methods to measure such distribution of the radioactive aerosols immediately, but the results are based on at most eight measurements related to different size ranges. On the other hand, there is the possibility to assess the size distribution of the ambient aerosols by using devices that consist of much more size measuring channels and therefore have a much better resolution; they are also much more widespread. However, the problem is achieving the proper transformation from the ambient to radioactive aerosols. Such transformation has been the subject of investigation and the experiments carried out in the radon chamber using different aerosols aimed to compare both results. measurement.

2. Measurement method

2.1. Size distribution of the radioactive aerosols

Measurements techniques for the assessment of the size distribution of radioactive aerosols containing short-lived radon progeny are based on specific units allowing the separation of different fractions of aerosols due to their sizes. The Radon Progeny Particle Size Spectrometer (RPPSS) which was applied has four diffusion screen batteries for the separation of different fractions of finer particles and a three stage impactor for larger particles. Additionally, the open-face filter collects aerosols without any separation. The RPPSS is also equipped with a pump to pass the sampling air through all these units. The configuration of these separation units enables the analysis of the size distribution of aerosols in the range from 0.6 nm up to approximately 2500 nm. Each of them, including the open face filter, is equipped with a semiconductor alpha detector connected to the spectroscopy system. As a result, the alpha radiation occurring during the decay of the radioactive aerosols collected in all of these stages can be recorded, and, as a result, the concentration of the short-lived radon progeny can be evaluated by the RPPSS. Two mathematical algorithms are applied to derive the size distribution and to fit to the obtained measurements results: the Twomey method (Twomey, 1975) and the Expectation Maximisation method (EMax) developed by Maher and Laird (1985). After the deconvolution process, the DCF's coefficients are calculated according to the evaluated size distribution and dosimetric model for the estimation of the dose, recommended by the International Commission on Radiological Protection (ICRP, 1994).

2.2. Size distribution of the ambient aerosols

The size distribution of the ambient aerosols was assessed by three spectrometers to cover the size range from about 3 nm up to 20 μm. The SMPS spectrometer equipped with the Differential Mobility Analyser of Nano type (Nano DMA) was used to separate and record ultrafine particles above 3 nm. The second SMPS spectrometer had Long DMA and the corresponding measuring range was approximately 15–700 nm (Fig. 2). The measurement principles of these spectrometers are similar. The sampling air containing the aerosols is charged and directed to the

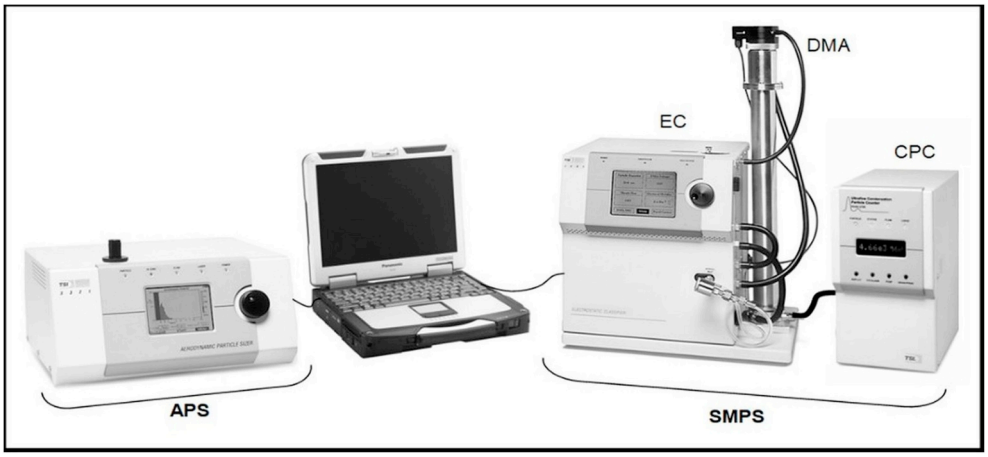

Fig. 2. Aerosol particle sizer (APS) and mobility particle sizer (SMPS).

DMA, which has cylindrical shape. Inside the DMA an electrostatic field is generated and the charged particles transported by the sampling air can be separated according to their mobility and the voltage of the electrode that is located in the centre of the cylinder. The voltage is changed gradually and as a result a further group of particles with specific sizes are separated step by step and recorded by the Condensation Particle Counter (CPC). The assessed distribution is expressed in terms of the mobility diameters.

The Aerodynamic Particle Sizer (APS) is designated to measure the concentration of larger particles in the range from 0.5 μm to 20 μm, expressed in terms of their aerodynamic diameters. In this case, the double laser system makes it possible to assess the flight time of particles and compare the results with the calibration curve. All these spectrometers were made by the TSI company (USA), and have in total approximately 250 size channels.

For fitting the two size distributions related to SMPS and APS spectrometers the nonlinear relationship that links mobility and aerodynamic diameters (Hinds, 1999) can be applied:

$$d_p = d_a\left(\frac{C_C(d_a)}{C_C(d_p)}\right)^{\frac{1}{2}}\left(\frac{\chi\rho_o}{\rho_p}\right)^{\frac{1}{2}} C_C(d) = 1 + \frac{\lambda}{d}\left(2.34 + 1.05e^{-0.39\frac{d}{\lambda}}\right) d = d_a \text{ or } d = d_p \quad (1)$$

There are the following variables: mobility diameter (d_p), aerodynamic diameter (d_a), slip correction factor (C_c), mean free path of the air molecules (λ), aerodynamic shape coefficient (χ), particle density (ρ_p), and unit density (ρ_o). The value of 1 for aerodynamic shape coefficient and 1 g/cm^3 for density was accepted in experiments. That is because no coarse particle were generated inside the chamber and their contribution was negligible. As a result, the entire meaningful size range was covered by the SMPS.

2.3. Assessment of the size distribution of radioactive aerosols based on the size distribution of ambient aerosols

Size distribution of radioactive aerosols can be determined taking into account the attachment rate of the unattached short-lived radon decay progeny (X), attachment coefficient $\beta(d_p)$, and size distribution of the ambient aerosols $Z'(d_p)$ (Porstendörfer, 1994):

$$X = \int_0^\infty \beta(d_p)Z'(d_p)dd_p \quad \beta(d_p) = \frac{2\pi D_f d_p}{\frac{8D_f}{d_p v_f} + \frac{d_p}{2\left(\frac{d_p}{2}+\lambda_f\right)}} \quad (2)$$

The following assumptions related to the particles of the unattached fraction were accepted based on the data recommended by Porstendörfer (1994): mean diffusion coefficient $D_f = 6.8{\cdot}10^{-6}$ m^2 s^{-1}, mean thermal velocity $v_f = 1.72{\cdot}10^2$ m s^{-1} and mean free path $\lambda_f = 4.9{\cdot}10^{-8}$ m.

The probability $P_R(d_{pi})$ of the generation of radioactive aerosols with sizes ranging from d_{pi} to $d_{pi} + \Delta d_{pi}$ was estimated according to the formula by Skubacz, Wojtecki, and Urban (2016):

$$P_R(d_{pi}) \approx \frac{\beta(d_{pi})Z'(d_{pi})}{\sum_{dpi}\beta(d_{pi})Z'(d_{pi})} = \frac{\beta(d_{pi})\frac{\Delta Z(d_{pi})}{\Delta d_{pi}}}{\sum_{dpi}\beta(d_{pi})\frac{\Delta Z(d_{pi})}{\Delta d_{pi}}} \quad (3)$$

where $\Delta Z(d_{pi})$ is the concentration of the ambient aerosols in the measurement size range of Δd_{pi}. The sum should include all possible sizes which have a significant contribution to the generation of radioactive aerosols. The concentration of coarse particles is usually relatively small and their contribution to the generation of the radioactive aerosols can be considered negligible.

2.4. Radon chamber

The radon chamber of 17 m^3 volume was made by the Feutron Company (Germany) and was used to perform research for different aerosols. It is well isolated from the outside atmosphere and equipped with a man and material lock to avoid significant violation of the internal conditions during the exchange of measuring devices and auxiliary equipment (Fig. 3). The temperature and relative humidity can be controlled (−20 − +60 °C; 20–90%.) according to the user defined timing cycle within technically acceptable limits (Skubacz, Chałupnik, Urban, & Wysocka, 2017).

3. Results and their discussion

Measurements were conducted in CMI radon chamber for five different ambient aerosol concentrations made by different types of aerosol sources:

- air in the radon chamber purified with an air filter: depleted concentration of aerosols,
- water aerosols,
- oil candles,
- incense sticks,
- a wax candle.

Some quantities that describe the ambient aerosol distribution in the radon chamber related to these sources are presented in Table 1. The data is based on the results obtained by SMPS and APS spectrometers. In the case of the aerosol-purified atmosphere, the concentration of ambient aerosols was from 2 to 3 orders of magnitude lower compared

Fig. 3. From left: radon chamber; SMPS and APS particle spectrometers (TSI, USA).

to a situation where the additional aerosol sources were applied. Usually, the largest contribution to the total concentration was made by the ultrafine particles, which reached almost 100% when aerosols were generated by oil candles. In this case, the mean diameter of 12 nm was the smallest one. Only in the case of the wax candle source was the situation different and a contribution of 95% to the total concentration was made by the fine particles. A mean particle diameter of 326 nm was the largest observed while the contributions of the coarse and super coarse particles were negligibly small.

The distribution of the ambient aerosols was calculated taking into account their measured number concentrations. It is not possible to evaluate the absolute number concentration of radioactive aerosols. However, based on the aforementioned transformation model, a normalized distribution of such aerosols can be evaluated that gives the probability of their occurrence in a specific size range. That is enough to assess the corresponding dose conversion factor.

Fig. 4 illustrates the distribution of ambient and radioactive aerosols when the oil candles were the aerosol source.

The curve relating to radioactive aerosols is the result of the transformation described above, where the size distributions of the ambient aerosols are the input data. In comparison to the other distributions, the curve is clearly shifted towards smaller particles. Usually the shape of the distribution was similar to the one illustrated in Fig. 5, where the incense sticks were the aerosol source. In comparison to the oil candles, the aerosols generated by the incense sticks were larger and more size scattered because the contribution of the ultrafine aerosols was nearly 100% for the oil candles and 55% for the incense sticks, and the corresponding geometric standard deviations were equal to 2.0 and 3.8, respectively. The aerosol concentration inside the chamber for both cases was about $5 \cdot 10^{-11}$ particles/m^3. Other parameters are given in Table 1. The average diameter of radioactive aerosols is usually larger in comparison to ambient aerosols. This is because the way the radioisotopes are bound to the ambient aerosols depends on the aerosol concentration in the specified size class and their size. The increase in size of radioactive aerosols means that the greater concentration of smaller particles did not compensate the influence on the process of much larger surface of larger particles despite of their relatively smaller concentration.

Fig. 6 shows the distributions of the radioactive aerosols based on the three above described methods when aerosols were generated by incense sticks. The EMax and SMPS + APS based distributions are most similar to each other. This was also the case for other aerosol sources. However, the SMPS + APS based distributions were slightly shifted towards finer particles in relation to the Twomey and EMax methods. This shifting was consistently observed. Excluding the influence of the mathematical methods applied, such regularity can only be caused by a systematic error. The spectrometers were calibrated, but the inflow of sapling air was always measured by a flowmeter and this could be a source of such error. However, the flowmeter was calibrated by an accredited laboratory, and the reported uncertainty was 2% at a 95% confidence level.

The Twomey and EMax algorithms are two different mathematical approaches used to derive size distribution that is as similar as possible to the true one based on the activity results obtained. Therefore, the differences can be caused by the methods applied and the low number of input measurement points, in the case of RPPSS there are 8 measurement points. The other main problems related to achieving the best fitting are related to function extremes (Skubacz, 2017) and that seems to be the case because the shapes are similar. However, the main purpose of the research was to compare the distributions of the radioactive aerosols obtained by different methods and finally assess the corresponding dose conversion factors and this is crucial for proper dose assessment. The dose conversion factors were calculated by taking into account the evaluated distributions and the functional dependence between dose conversion factors and particle size (Porstendörfer, 2001; Zock, Porstendörfer, & Reineking, 1996). The evaluated dose conversion factors based on the RPPSS and (SMPS + APS) results are included in Table 2. They represent both public and occupational exposure at a mouth breathing rate of 0.75 m^3/h and 1.20 m^3/h.

Table 1
Characteristics of the ambient aerosol generated.

Aerosol source	Mean diameter	Total concentration	Contribution to the total concentration			
			Aerosol class (μm)			
			0.004–0.1 (Ultrafine)	0.1–2.5 (Fine)	2.5–10 (Coarse)	10 − 20 (Super coarse)
	(nm)	(m^{-3})	(%)	(%)	(%)	(%)
Purified air	119	6.6E + 08	50	50	5.2E-04	4.2E-06
Water aerosols	87	9.5E + 10	75	25	2.5E-02	4.6E-04
Oil candles	12	5.4E + 11	~100	~0	6.3E-06	1.0E-08
Incense sticks	106	4.1E + 11	55	45	1.1E-03	4.3E-06
Wax candle	326	3.6E + 11	5	95	1.2E-03	2.2E-05

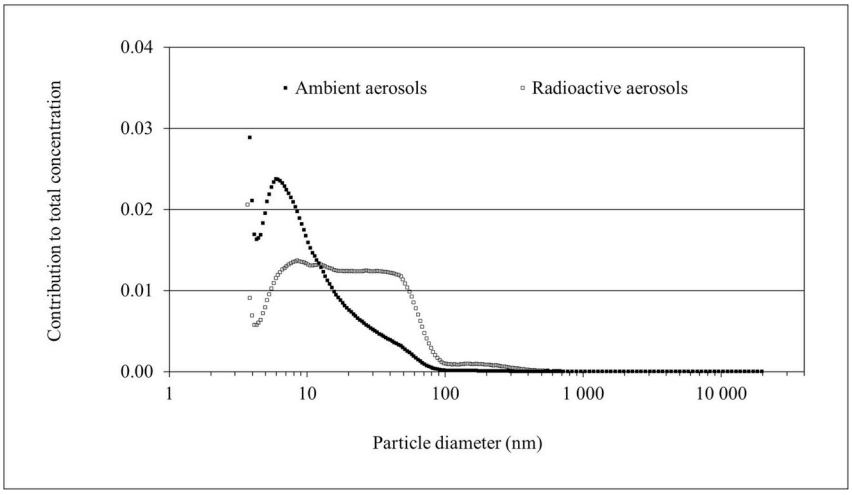

Fig. 4. Size distribution of ambient and radioactive aerosols for oil candles.

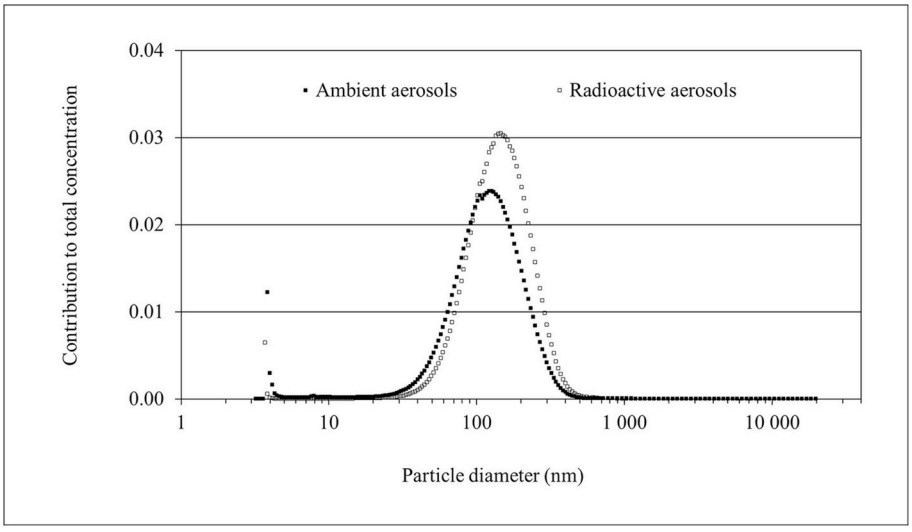

Fig. 5. Size distribution of ambient and radioactive aerosols for incense sticks.

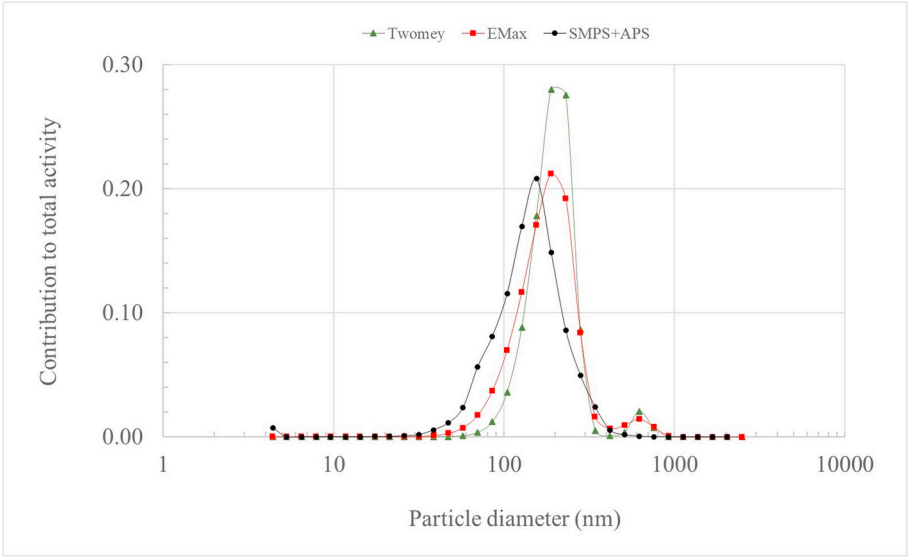

Fig. 6. The distribution of radioactive distribution according to Twomey and EMax method, and based on the data related to the size distribution of ambient aerosols (SMPS + APS) for aerosol generated by the incense sticks.

Table 2
Evaluation of the dose conversion factors for two breathing methods: [1]breathing by mouth at the rate of 0.75 m^3/h and [2]breathing by mouth at the rate of 1.2 m^3/h.

Aerosol source	Radon and its progeny		Dose conversion factor, Sv/(J/m^3h)					
	^{222}Rn	PAEC	Twomey method		EMax method		SMPS + APS	
	Bq/m^3	μJ/m^3	Mouth[1]	Mouth[2]	Mouth[1]	Mouth[2]	Mouth[1]	Mouth[2]
Purified air	1014	3.4	9.7	13.2	10.4	14.8	12.4	13.9
Water aerosols	4000	29.0	1.8	2.2	2.1	2.7	3.5	4.3
Oil candles	12400	50.2	12.0	14.4	12.0	14.6	17.1	19.5
Incense sticks	7760	42.6	1.9	2.3	3.0	3.9	2.7	3.3
Wax candle	7334	29.1	3.5	4.4	5.0	6.4	5.8	6.5
		Average	*5.8*	*7.3*	*6.5*	*8.5*	*8.3*	*9.5*

Potential alpha energy concentration (PAEC) is a measure of exposure to short-lived radon progeny and is equal to the total energy of alpha radiation that can be released by this progeny occurring in a defined unit of volume of air as a result of their complete decay.

Different measuring instruments as well as calculation methods were implemented in order to realise the research aim, yet the calculated values are only slightly different. The average relative differences for the EMax and (SMPS + APS) based methods are in the order of 10–20%, and for the Twomey and (SMPS + APS) methods 20–30%. Nevertheless, it can be noticed that the values obtained by the (SMPS + APS) method are usually higher. This is because these distributions are slightly shifted towards finer particles, for which the dose conversion factors are higher.

4. Conclusions

- There is quite a good agreement between the distributions of radioactive aerosols which were obtained by the Twomey and EMax methods and those which were calculated based on the data obtained by SMPS and APS spectrometers. Importantly, the SMPS and APS based results have much better resolution.
- The SMPS + APS based distributions are slightly shifted towards finer particles in relation to the results obtained by the Twomey and EMax methods. The values of the mean diffusion coefficient, mean thermal velocity and mean free path for the unattached fraction, which were used to perform calculation were checked during experiments. In addition, changes to them, within reasonable limits, does not lead to better compliance of the distributions. The only way to achieve better fitting seems to be a slight change of the attachment coefficient in order to increase the probability of the generation of radioactivity in a given range for bigger particles.
- The calculated dose conversion factors were usually significantly larger than the values recommended in legal regulations, which are equal to 1.4 Sv/(J/m^3 h) and 1.1 Sv/(J/m^3 h) respectively for occupational and public exposure.

Conflicts of interest

None declared.

Ethical statement

Authors state that the research was conducted according to ethical standards.

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