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Unattached fraction of radon progeny in Polish coal mines

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Abstract. The system of the monitoring of the radiation hazard in Polish coal mines is based on the monitoring of the workplaces. This system works since 1989 in all coal mines. It gives a very good basis for further epidemiological investigation and assessment of the health detriment within the population of the mines as a result of the exposure for natural radiation. It is very important problem, due to the fact of the presence in the mines another factors, which probably have a synergetic effects on the respiratory tracts. As the routine instrument, a device called ALFA-31 sampling probe was developed in our laboratory. This device was accomplished to regular dust sampler and simultaneous measurements of dust content and potential alpha energy concentration of radon progeny are obligatory in all underground mines in Poland. But the microcyclone used a separator of the respirable fraction which causes the cut-off of unattached fraction of radon progeny, On the other hand measurements of the unattached fraction of short lived radon progeny play a very important role in the investigations of the adequate dose from this source of radiation hazard. During field experiments the use of the alpha spectroscopy system is necessary, while measurements are done not in the vacuum chambers but under normal pressure. It leads to situation, when particular peaks in alpha spectrum are very wide and interfere with other peaks of another alpha-emitting radionuclides. Such instrumentation was designed and completed, and a survey in several underground mines was performed. The analysis of the obtained results must be done very carefully; in other case it may cause a very big uncertainty of the result. In this paper a new approach to the analysis of the alpha spectra has been described. This approach can be used also in other applications of alpha spectroscopy, in which the analysis of energy of alpha peaks in spectrum is needed. The method of the analysis is based on a non-linear regression. The results of the approximation were tested by the method of non-linear regression and very good fitting have been found. Results of the survey show, that the average ratio of unattached and attached fractions is in Polish coal mines at level 3-5%. But it may cause the significant increase of the dose equivalent, due to our calculations at least 15-20% in comparison with dose equivalent caused by attached fraction.

Introduction

The occurrence of enhanced natural radioactivity in Polish coal mines was discovered in early 60's (Sałdan,1965). Investigations, performed during 70's showed, that underground galleries sources of high gamma radiation were present, as a result of precipitation of deposits of radium and barium sulphates from radium-bearing waters (Tomza and Lebecka, 1981). Further research activities on this field enabled to identify main sources of radiation hazard in underground coal mines (Lebecka et. al., 1985). Results of these investigations showed, that the most important source of ionising radiation in underground galleries are short lived radon progeny. Other sources are radium-bearing waters and radioactive deposits. Enhanced natural radioactivity, especially radon progeny in air (^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po), leads to the increase of radiation exposure for the miners.

Since 1989 monitoring of radiation hazard in Polish coal mines is obligatory. Due to the mining regulations in all underground mines following measurements must be done with the certain frequency:

- concentration of radium isotopes in waters;
- concentration of natural radionuclides in deposits;
- gamma dose rates and gamma doses;
- concentration of potential alpha energy of short lived radon progeny.

Results of the systematic monitoring of the radiation hazard revealed, that radon progeny is the most significant source of doses - more than 90% of cumulative dose equivalent for mines is due to the exposure on radon daughters. But dose equivalent depends also on the aerosol's size distribution and a contribution of the unattached fraction. This fraction consists of very small, charged particles and clusters with the size up to 15nm, showing a detached section in the size distribution curve (Reineking and Porstendörfer, 1986). The theory of the interaction of the aerosols with the respiratory tracts indicates, that unattached fraction is much dangerous as typical aerosols with average diameter 200nm (Birchall et. al. 1994). So, the influence of the unattached fraction on the exposure is much higher than its percentage contribution to activity of radon progeny. Therefore reliable results of measurements of unattached fraction concentration are so important for the evaluation of dose in the mining industry.

In this paper results of measurements of unattached fraction in several Polish coal mines are presented. The assessment of dose equivalent and discussion of errors is also done.

Experimental and methods

The percentage contribution of the unattached fraction (f_p) is a radio of the potential alpha energy of the free fraction to total alpha energy concentration:

$$f_p = \frac{\alpha_1 C_{f1} + \alpha_2 C_{f2} + \alpha_3 C_{f3}}{\alpha_1 C_{o1} + \alpha_2 C_{o2} + \alpha_3 C_{o3}} \qquad (1)$$

$$\alpha_1 = 0.1042 \qquad \alpha_2 = 0.5144 \qquad \alpha_3 = 0.3814$$

where C_{f1}, C_{f2}, C_{f3} are concentrations of unattached fraction of ^{218}Po, ^{214}Pb, ^{214}Bi and C_{o1}, C_{o2}, C_{o3} are total concentrations of above-mentioned isotopes.

Calculations of the concentrations of particular isotopes can be done for instance on the basis of results of measurements done for filters, through which air with aerosol was sucked earlier.

Our counting system consists of two parallel alpha spectrometers for simultaneous measurements and analyses of alpha spectra, emitted by radon progeny, collected on two filters (fig.1. and 2). First filter is the open face one, on which attached and unattached progeny is deposited during sucking of the air. Above second filter a wire screen is placed, therefore only attached fraction could pass the screen and reach the filter. The flow rates through both filters were controlled by two separate flowmeters of the same type.
The pumping time was usually equal 10 minutes, and the flow rate was of about 0.9m^3/h for each filter. At the end of pumping period two semiconductor detectors were placed over filters as soon as possible (fig.2). Later several consecutive measurements of alpha activity collected on the filter were done.
Our considerations were based on the theory of filtration of the air through diffusion screens, described by Cheng and Yeh (1980), Cheng et al. (1980). The screen thickness, screen wire diameter and the solid volume fraction of the screen were chosen for the used air velocity.

Unfortunately, the diffusion screen cuts off also a portion of the attached fraction, what introduces an uncertainty into the equations.

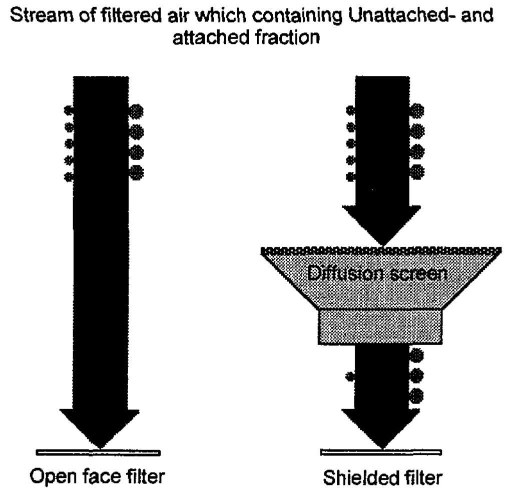

Fig. 1. Pumping of air through the open face - and shielded filter

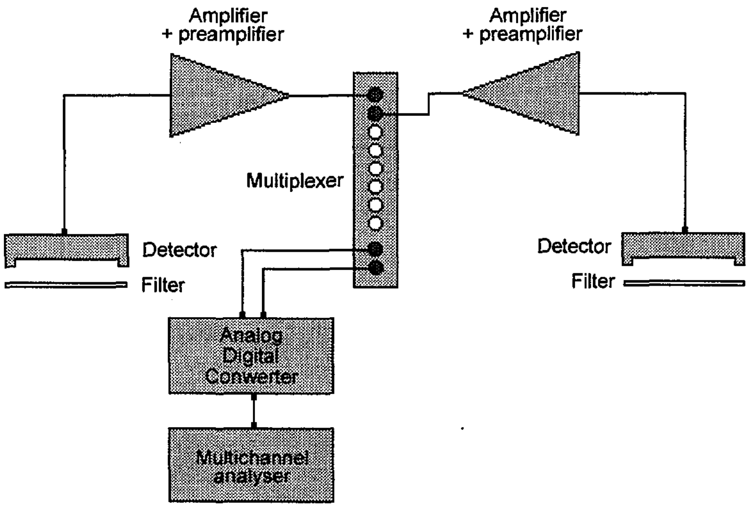

Fig. 2. The schematic picture of the instrumentation for the activity measurements of filters.

Theoretical considerations lead to the solution, connecting results for open face filter (C_{o1}, C_{o2}, C_{o3}) and shielded filter (C_{S1}, C_{S2}, C_{S3}). These results are related due to the following equations:

$$C_{oi} = C_{Si} + \psi C_{fi} + C_{Gi} \quad i = 1,2,3 \text{ for } {}^{218}\text{Po}, {}^{214}\text{Pb i } {}^{214}\text{Bi respectively} \qquad (2)$$

where C_{Oi} are concentrations of the certain radionuclide (i) on the open face filter, C_{Si} are concentrations of the *i-th* radionuclide on the shielded filter; ψC_{fi} *and* C_{Gi} are fraction of the *i-th* radionuclide from the unattached- and attached fraction; caught by the diffusion screen.

Without simplifying of these equations, calculations of the activities are impossible. The solution was proposed by Reineking and Porstendörfer (1990). From experiments with unattached fraction authors drew following conclusions:

- the particle size distributions of the aerosol-attached activities for all short-lived radon daughters are identical within a constant multiplier and the unattached activities of ^{214}Bi (^{214}Po) are negligible.

And than:

$$f_p = \frac{\alpha_1 C_{f1} + \alpha_2 C_{f2}}{\alpha_1 C_{o1} + \alpha_2 C_{o2} + \alpha_3 C_{o3}} \tag{3}$$

where:

$$C_{fi} = \frac{C_{oi} C_{S3} - C_{Si} C_{o3}}{C_{S3} - (1-\psi) C_{o3}} \quad \text{lub gdy} \quad \psi \to 1 \quad C_{fi} = C_{oi} - \frac{C_{o3}}{C_{S3}} C_{Si} \quad i = 1,2 \tag{4}$$

Applying these parameters we were able to calculate parameters of the diffusion screen, with the best properties of cutting off the unattached fraction. The parameters of the used diffusion screen are shown in table 1.

Table 1. Parameters of the diffusion screen

Radius of a screen	Screen thickness	Screen wire radius	Specific density of the screen material	Solid volume fraction
4 cm	59.7 μm	14.7 μm	7.8 g/cm^3	0.282

For the filtration of the aerosols from the air, filters of NUCLEOPORE POLYCARBONATE PC (Costar Corporation, USA) were used. Pore size is equal 0.8μm, while the thickness of the filter was 10μm. These filters ensure better energy resolution of the alpha spectra in comparison with membrane filters FM-1, used by us for routine measurements of dust concentration and potential alpha energy concentration in mines (Pore size 1.5μm and thickness 200μm). Both the energy resolution and the detection efficiency was better by tens of percent for aerosols with diameter of 3μm and for tobacco smoke.

Elements of alpha spectrometers were bought in CANBERRA (USA). It consists of supply unit, two alpha semiconductor detectors, two units with preamplifiers and amplifiers, a multiplexer, and finally, on-board multichannel analyser together with analog digital converter, installed in the notebook.

For the detection of alpha particles two semiconductor detectors ULTRACAM-1700-AM with active surface 1700mm^2 were applied. We had to use such detectors, with the aluminium screen (thickness 0.5μm). These detectors are not sensitive for high moisture and light, moreover, it is possible to clean the surface. Additionally, detectors work not only in the vacuum chamber but also under normal atmospheric pressure and the minimum distance between filter and detector is only 1mm. These features are very important, because as it was above mentioned, that measurements were done in coal mines. The main reason of such choice was that the instrumentation had to be used in very aggressive environment in underground galleries.

Error analysis

The uncertainty of the measurements have been solved accordingly with Currie (Currie, 1969). At first the critical level f_C was calculated:

$$f_c = k_\alpha \sigma_o \tag{5}$$

where σ_o is a standard deviation for $f_p=0$, while k_α is a reduced variable for chosen confidence level α. Due to the previous equations the variance is equal:

$$\sigma_o^2 = \frac{\alpha_1^2\left(C_{S1}^2\sigma^2(C_{o3}) + C_{o1}^2\sigma^2(C_{S3})\right) + \alpha_2^2\left(C_{S2}^2\sigma^2(C_{o3}) + C_{o2}^2\sigma^2(C_{S3})\right)}{\left(\alpha_1 C_{o1} + \alpha_2 C_{o2} + \alpha_3 C_{o3}\right)^2 (C_{S3} - (1-\psi)C_{o3})^2} \tag{6}$$

Later, comparing the result of measurement and the critical level, we were able to calculate the uncertainty of the result:

$$f_p \le f_c \quad \Rightarrow \quad \textit{unattached fraction} \le f_p + t_{1-\gamma} \bullet \sigma(f_p) \tag{7}$$

$$f_p > f_c \quad \Rightarrow \quad f_p - t_{1-\gamma/2} \bullet \sigma(f_p) \le \textit{unattached fraction} \le f_p + t_{1-\gamma/2} \bullet \sigma(f_p) \tag{8}$$

Value $t_{1-\gamma}$ is a reduced variable for the chosen confidence level γ in the one-tiled test, while $t_{1-\gamma/2}$ is a corresponding value for the two-tiled test.

Results

We performed 30 measurements in four underground mines. Three of these coal mines are operating but the fourth one is an abandoned mine, only de-watering system works there. All measurements were done in mines without methane hazard, due to two main reasons. Firstly, in mines with methane hazard the intensity of the ventilation is always higher, therefore radon concentrations are lower than in non-methane mines. On the other hand, in mines with methane hazard, regulations concerning intrinsic safety of applied instrumentation are very rigid, therefore we obtained no permission to do our measurements in such mines.

Sampling sites were chosen thanks to the help of the ventilation services of particular mines. Analysis of the result of obligatory monitoring of potential alpha energy concentration have been done, because we wanted to find places, where enhanced concentration of radon progeny have been measured. Accordingly to the error analysis. Equilibrium-equivalent concentration (EEC) of the total activity of short-lived radon daughters should be higher than 30Bq/m^3. In this case the critical level f_c is better as 2% and we are able to make measurements with the uncertainty of the measured value below few percent.

In several cases, we found the EEC of radon progeny below 30 Bq/m^3, in places, where previous investigations showed much higher values. Such measurements were done mainly in the abandon mine. All such results were not a subject of a further analysis, because the uncertainty exceeded the critical level.

Table 2. Results of measurements of unattached fraction of radon progeny in Polish coal mines.

No.	**Total EEC** C_{oEq} **[Bq/m^3]**	**EEC of unattached fraction** C_{fEq}**[Bq/m^3]**	**Percentage of the unattached fraction** f_p**[%]**	**Critical level** f_c**[%]**	**Environmental parameters** **Humidity/Pressure/Temperature**		
					RH[%]	**P[mbar]**	**t[°C]**
1.	328.5	8.1	**2.5±1.1**	0.7	90	1054	18.8
2.	336.8	10.9	**3.2±1.4**	0.9	77	1046	22.0
3.	526.0	≤6.3	**≤1.2**	0.7	83	1044	21.1
4.	163.0	≤4.2	**≤2.6**	1.8	88	1022	20.5
5.	310.2	≤4.7	**≤1.5**	0.9	96	1031	18.2
6.	330.5	7.6	**2.3±1.2**	0.8	79	1056	22.0
7.	359.9	5.0	**1.4±1.4**	0.9	79	1056	22.0
8.	885.6	≤9.7	**≤1.1**	0.7	87	1071	24.5
9.	290.4	≤5.8	**≤2.0**	1.1	98	1053	21.0
10.	321.6	44.7	**13.9±1.4**	0.8	86	1056	23.0
11.	70.0	9.2	**13.1±1.7**	0.8	89	1049	19.6
12.	1280.1	152.3	**11.9±1.0**	0.6	85	1049	20.8
13.	104.7	≤2.0	**≤1.9**	0.8	91	1022	19.0
14.	440.0	≤8.4	**≤1.9**	1.1	93	1018	20.6
15.	165.1	≤5.1	**≤3.1**	1.5	85	1022	22.7
16.	11.4	≤2.9	**≤25.7**	9.1	51	*awaria*	18.6
17.	15.6	≤0.7	**4.5±5.5**	4.1	51	*awaria*	18.6
18.	91.9	11.1	**12.1±2.1**	1.4	91	1011	18.5
19.	96.5	5.7	**5.9±2.5**	2.0	86	1011	19.2
20.	234.7	20.7	**8.8±1.4**	0.9	86	980	15.0
21.	293.4	5.3	**1.8±1.5**	1.0	89	979	14.3
22.	41.7	9.5	**22.7±2.4**	1.6	90	987	16.6
23.	51.5	6.6	**12.9±2.7**	2.1	92	987	16.6
24.	12.2	≤1.0	**≤8.2**	5.9	83	1022	20.0
25.	9.1	≤2.6	**≤28.1**	21.6	84	1022	17.7
26.	77.4	≤2.2	**≤2.8**	1.8	94	987	16.5
27.	75.9	5.5	**7.3±2.2**	1.7	95	987	16.4
28.	8.5	0.7	**≤8.3**	3.8	46	1006	12.7
29.	10.1	0.9	**8.7±7.8**	5.2	69	1005	13.6
30.	3.8	1.2	**31.2±20.7**	11.2	45	998	13.8

Results of our measurements are shown in table 2. In two out of chosen mines, as sampling site all important places were used, like outflows from longwalls and headings, where enhanced levels of radon progeny have been found earlier. During each measurements, additional parameters were monitored - the barometric pressure, relative humidity and temperature. All these data are also showed in table 2. Analysis of alpha spectra was performed with application of non-linear regression, described in the earlier publication (Skubacz, 1998).

We would like to mention the dependence between the energy resolution of the alpha spectrometry system and a type of aerosols. During calibration of the instrumentation in the radon chamber water aerosols as well as cigarette's smoke aerosols were used. For instance in case of smoke aerosols the *FWHM* was equal 597 keV for alpha peak 6.0 MeV of ^{218}Po and, respectively, 412 keV in case of ^{214}Po and energy 7.7 MeV. When water aerosols were used we measured following values - 396 keV for peak of ^{218}Po and 328 keV for peak of ^{214}Po. Explanation for this phenomenon is that the penetration depth of smaller smoke aerosols is bigger than for big water aerosols. However we took into account the whole energy spectra so there was no difference between efficiency coefficients. This way the analysis of the chosen parts of alpha spectra can introduce some error especially in evaluating of activity concentrations.

Also corrections for the influence of thoron progeny have been done. Into further consideration only the results with critical level better than 2% were taken into account. The confidence level is equal 1σ.

The correlation between concentration of the unattached fraction and concentration of aerosols is known from the literature very well. Unfortunately, we weren't able to measure the concentration of aerosols in underground galleries and the size distribution. Therefore it is very difficult to say, why in certain places the percentage of the unattached fraction was higher as in other sites. We found no correlation of the unattached fraction with dust content nor with moisture. Such measurements we would like to undertake in the future.

Investigations of the unattached fraction, performed by different scientists, were done mainly in uranium mines. Busigin and co-workers (1983) found in two investigated mines, that the maximum contribution of the unattached fraction was 7%, but usually below 2%. Similar results were reported by Porstendörfer (1991) - maximum 7%, an average 3% in a shale mine, while in barite mine contribution of unattached fraction was below 1%. Higher values were found in caves. In Postojna Cave (Slovenia) - maximum contribution of the free fraction was equal 16%, and in average 10%.

In our work, in most sites we measured results below critical level (fig.3). But in 8 cases the contribution of the unattached fraction ranged within 1-10%, in 5 cases within 10-20% and only in one case level 20% was exceeded. The average value was of about 5.2%.

It means, that in the chosen coal mines contribution of the unattached fraction is relatively high. We would like to emphasise, that in two out of 4 mines, concentration of the unattached fraction was measured in all main sites of the ventilation system.

The assessment of the influence of the unattached fraction on the dose equivalents for miners have been done. We applied the method of calculations, described by Zock and co-workers (1996) - we made an assumption that size distribution of the attached fraction is log-normal, with AMD = 200nm and σ = 2.3nm. Such parameters for mines were quoted in publication Birchall et.al. (1994). quoted.

The result of our considerations shows, that the influence of unattached fraction on dose equivalent in Polish coal mines is significant. The average value of the the unattached fraction was calculated as 5.2%, but as an effect of the exposure of the miner, the dose equivalent would increase of about 45% in case of mouth breath with a rate 1.2m^3/h. As a result of the respiration through nose with lower intensity - $0.75m^3/h$ – the dose equivalent would be only 12% higher (see fig.4.).

In one site the concentration of the unattached fraction exceeded 20%. Moreover, the measurement showed rather low uncertainty of that result. Taking into account the same assumptions as above, it leads us to the conclusion, that in that site the dose equivalent is two times higher (100%) in comparison with calculations, made on a basis of the concentration of the attached fraction only. During a closer inspection of the site, performed with employees of ventilation service, we found that the air stream came from outlets of two longwalls at the deeper horizon. The dust content was low and probably it was a reason of such high contribution of the unattached fraction.

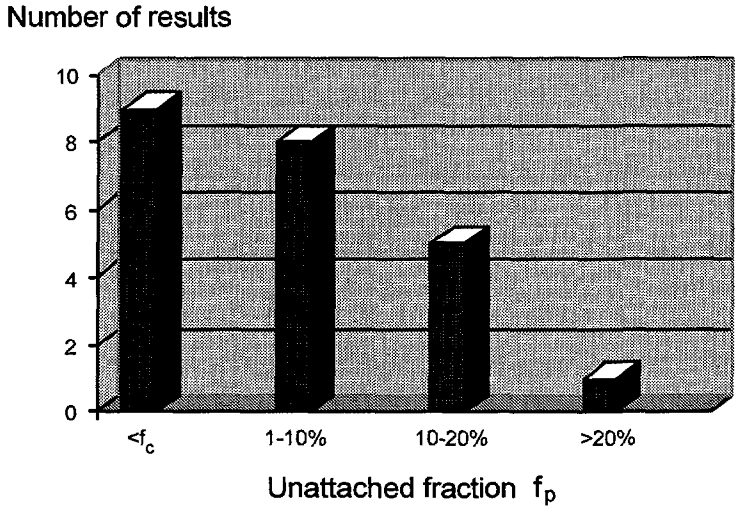

Fig. 3. Distribution of the results of the unattached fraction of radon progeny in coal mines in Poland.

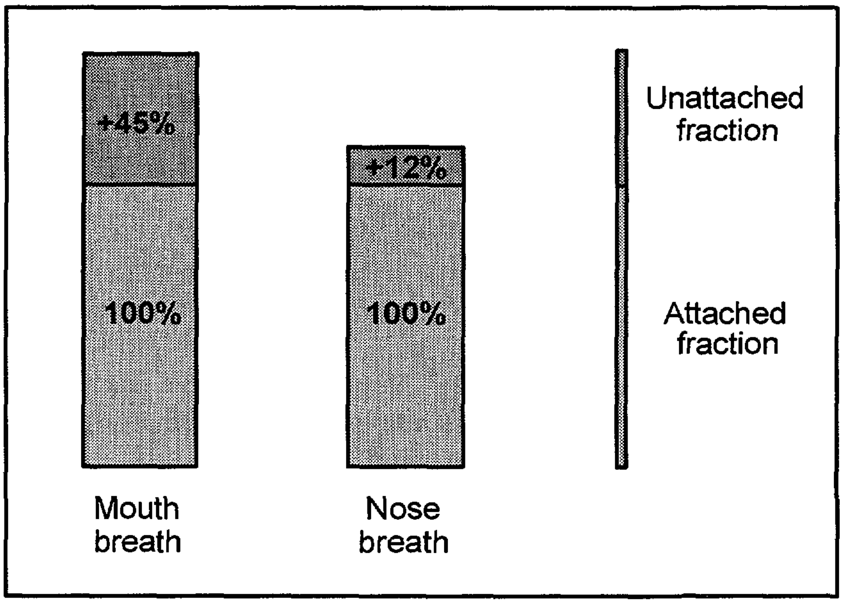

Fig.4. The influence of the unattached fraction (5.2%) on the dose equivalent for the log-normal size distribution of the attached fraction, AMD = 200nm and σ=2.3nm

Finally we would like to point out, that due to our results the concentration of the unattached fraction in chosen coal mines is relatively high. Comparable analysis with measurements in German underground mines (Porstendörfer et. al., 1991) shows, that the average value of the unattached fraction in Polish mines is of about 70% higher. Therefore the dose equivalent for miners seems to be roughly 45% higher as calculated previously, with the assumption that unattached fraction concentration is negligible. Of course, it needs confirmation in the future.

Conclusions

- Investigations of the unattached fraction have been done in four Polish coal mines. In two of these mines measurements were performed at all important workplaces. Collected data show, that the average value of the unattached fraction is of about 5.2%. It would increase the dose equivalent for underground mines – 45% in case of respiration through the mouth but 12% while breathing through the nose. Our campaign was the first one in Poland, therefore we have no possibilities to compare results with other national survey.

- Analysis of uncertainties of measurements of the unattached fraction have been done. Critical level was calculated for the instrumentation, designed and constructed for the survey. Therefore we are able to obtain reliable results for the unattached fraction in cases, when the concentration of the potential alpha energy of the attached fraction is not lower than 30Bq/m^3. But even for much higher PAEC and relatively high contribution of the unattached fraction, critical level is not better than 0.6%. The conclusion can be drawn, that with application of diffusion screen, for all results at level 1-2%, uncertainties are comparable with results.

- The knowledge of the size distribution of the aerosols at the sampling points and in the radon chamber is important. For different AMD the energy resolution in the alpha spectra is different. Typically for smaller particles the penetration depth into the filter media is bigger, therefore the resolution is worse. Our protocol is to analyse whole energy spectrum if we would like to avoid errors due to the different conditions in underground galleries and during calibration.

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