DETERMINATION OF RADON DAUGHTER ACTIVITIES OF DIFFERENT AEROSOL FRACTIONS BY GROSS-α AND GROSS-ß MEASUREMENTS

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Introduction

Inhalation of the short-lived radon decay products, ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po yield the greatest dose contribution to the natural background of the human exposure. One of the dominant parameter related to dose is the distribution of the decay products attached to the different sizes of aerosol particles in the air because the deposition and uptake of the inhaled activities in the lung depend on the particle sizes.

According to the molecular processes in air the daughter element ^{218}Po produced from the ^{222}Rn is a free ion but molecules of water vapour or trace gases coalesce almost immediately around the ion, forming a molecular cluster of 0.5-20 nm in diameter [1]. The ion and the cluster are usually referred to as free, uncombined or unattached decay products. The unattached ^{218}Po is highly mobile and, after 10-100 s, it attaches to an aerosol particle (normal size in the range of 20-500 nm) or plate out on indoor surfaces or transports outdoors with the outgoing air and finally decays into ^{214}Pb. The decay product ^{214}Pb may remain on the aerosol or indoor surface or become unattached as the result of its recoil energy. The behaviour of nuclide ^{214}Pb is similar as of the ^{218}Po one but following the decay the product ^{214}Bi typically remains attached, since the recoil energy from beta decay is not sufficient to promote detachment [2,3].

Our aim was to determine the radioactivities of each short lived radon progeny attached to the aerosol particles with respect to the size distribution in order to improve the assessment of the inhalation dose. Therefore the radioactivities of the samples collected by separation of the aerosol particles with respect to the sizes have been determined. We tried to find any differences in the activities of the individual progenies with respect to the different sized aerosol particles.

Methods

Samples have been provided by an electrical low pressure impactor and for gross-α and gross-β measurements of the aerosol samples ZnS/plastic-type sandwich detectors have been used. The activities of the progenies have been estimated from the gross-type measurements by use a time-dependent model and fitting the model to the experimental data.

The impactor (type ELPI) is useable to real-time monitoring of the distribution of aerosol particles [4,5] in the size range of 30 – 10000 nm with 12 channels, but it is not suitable to collect the unattached progenies. The principle of functioning is based on charging, inertial classification and electrical detection of the aerosol particles. The instrument consists primarily of a corona charger, low pressure impactor (classifying the particles according to their aerodynamic diameter, not charge) and multi-channel electrometer.

In our investigations the aerosol particles were collected from a closed radon chamber in a common laboratory building. The chamber (V = 0,21 m^3) contained a relatively high radon concentration (50-200 $kBq.m^{-3}$) in equilibrium with the short lived daughters generated, without any additional aerosol sources. The chamber-air was pumped by intensity of 0.01 $m^3.min^{-1}$, during 10-15 minutes through the impactor stages and after finish of pumping the radioactivity measurement (CPS) of the collected samples started during 1,5-2 hours by time intervals of 1 or 2 mins. The CPS-values have been provided by ZnS/plastic sandwich detectors with detection efficiencies of 0.05-0.2, depending to the gross-α and -β determinations.

A multiexponential model has been developed for simulation of the measured values and in order to assess the initial activities of the daughters after end of pumping a procedure of parameter estimation has been introduced by the software ModelMaker [6]. The exponential expressions of the model are the following:

$$CPS_{Po-218}(t) = \eta_{Po-218} \cdot A_{Po-218} \cdot e^{-\lambda_{Po-218} \cdot t}$$

$$CPS_{Pb-214}(t) = \eta_{Pb-214} \cdot A_{Pb-214} \cdot e^{-\lambda_{Pb-214} \cdot t} + \eta_{Pb-214} \cdot A_{Po-218} \cdot \lambda_{Pb-214} / \lambda_{Po-218} \cdot \left(e^{-\lambda_{Pb-214} \cdot t} - e^{-\lambda_{Po-218} \cdot t}\right)$$

$$CPS_{Bi-214}(t) = \eta_{Bi-214} \cdot A_{Bi-214} \cdot e^{-\lambda_{Bi-214} \cdot t} + \eta_{Bi-214} \cdot A_{Pb-214} \cdot g_{Pb-214}(\lambda_x) \cdot e^{-\lambda_{Pb-214} \cdot t} + \eta_{Bi-214} \cdot A_{Po-218} \cdot g_{Po-218}(\lambda_x) \cdot e^{-\lambda_{Po-218} \cdot t}$$

$$A_{Po-214}(t) = A_{Bi-214}(t),$$

where:

$CPS_{Po\text{-}218,\ Pb\text{-}214,\ Bi\text{-}214}$: intensity in units of count per second as the result of the radioactive measurements of ^{218}Po, ^{214}Pb and ^{214}Bi, respectively [cps];

$\eta_{Po\text{-}218,\ Pb\text{-}214,\ Bi\text{-}214}$: detection efficiency of activities ^{218}Po, ^{214}Pb and ^{214}Bi, respectively [$cps.Bq^{-1}$];

$A_{Po\text{-}218,\ Pb\text{-}214,\ Bi\text{-}214}$: initial activities of ^{218}Po, ^{214}Pb, ^{214}Bi in the aerosol sample at the end of pumping (at t=0) [Bq];

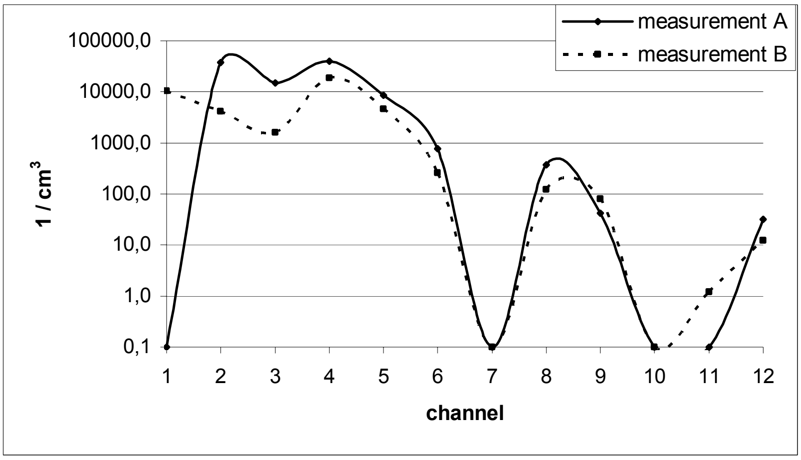
$\lambda_{Po\text{-}218,\ Pb\text{-}214,\ Bi\text{-}214}$: decay constant of ^{218}Po, ^{214}Pb, ^{214}Bi, respectively (s^{-1});

$g_{Pb\text{-}214}(\lambda_x)$, $g_{Po\text{-}218}(\lambda_x)$: constants, containing quotients of decay constants of ^{218}Po, ^{214}Pb, ^{214}Bi.

The time series data of gross-α and gross-ß intensities were fitted simultaneously by least squares method and the initial activities of the daughters were estimated [7].

Results

The air sampling period was reduced to the minimum (10-15 minutes) in order to be able to detect directly the α-activity of ^{218}Po, which has a short half-life of 3.05 minutes. Due to the short sampling time the amount of the collected aerosol particles were quite little so some channel of the impactor did not worked properly. Figure 1 shows the number distribution of the collected aerosol samples as the result two independent measurements. It can be seen that the parallel measurements did not correspond to each others. Nevertheless results of radioactivities could be compared by the ratios of ^{214}Pb / ^{218}Po and ^{214}Bi / ^{218}Po.



Channel	$D_{50\%}$ (nm)
12	7000
11	4300
10	2700
9	1800
8	1100
7	700
6	400
5	300
4	200
3	100
2	60
1	30

Figure 1. Distribution of the number of collected particles and mean grapping diameter of each channels

As examples the time-dependent radioactivities measured in the aerosol samples and the assessed nuclide-specific radioactivities are presented in Figure 2 and Figure 3. The green dots are the measured CPS by the α-detectors [cps], the blue ones by the β-detectors [cps], the green and blue lines are the proper fitted curves and the broken lines are the assessed individual activities of the progenies ^{218}Po, ^{214}Pb and ^{214}Bi [Bq] derived from the measured data and estimated parameters (initial activities).

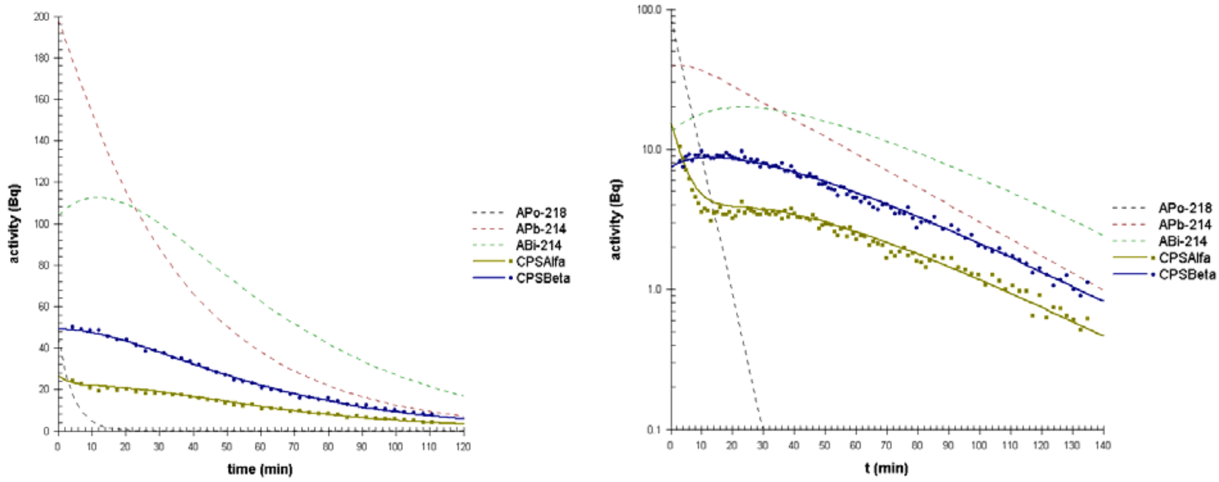
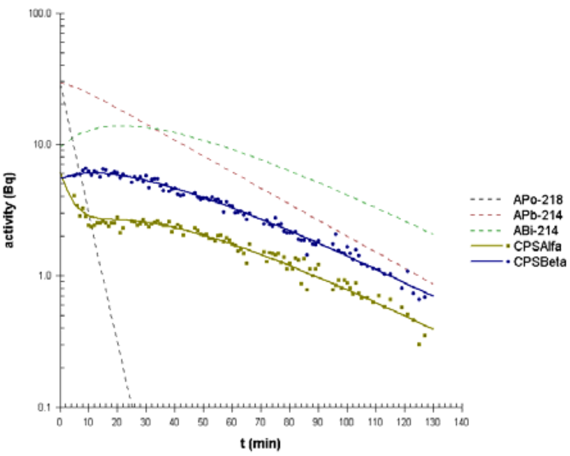


***Figure 2.** Simultaneous fitting of the measured gross-α and gross-β activities (dotts), and the assessed activities of the radon progenies in the **60-100 nm** range aerosol particles as the result of two independent experiments*



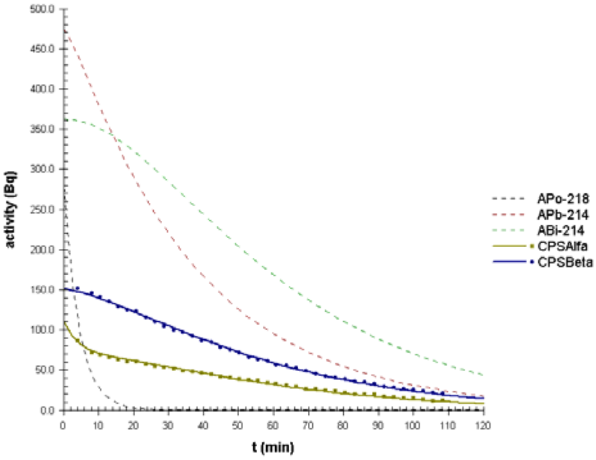


***Figure 3.** Simultaneous fitting of the measured gross-α and gross-β activities (dotts), and the assessed activities of the radon progenies in the **400-700 nm** range aerosol particles as the result of two independent experiments*

In general, standard deviations of the measured data and the assessed initial activities are 10-30 % but in extreme cases much higher. Therefore the ratios of the initial activities produce high ranges.

If we assume that the specific amount of the activities attached to the aerosols are independent on the elements of progenies the simulated activities of the filters during pumping and later on are provided in Figure 4. In addition to the decay processes the simulation has taken into account the intensity of pumping and the dilution of progenies during pumping due to the relatively small volume of the chamber used.

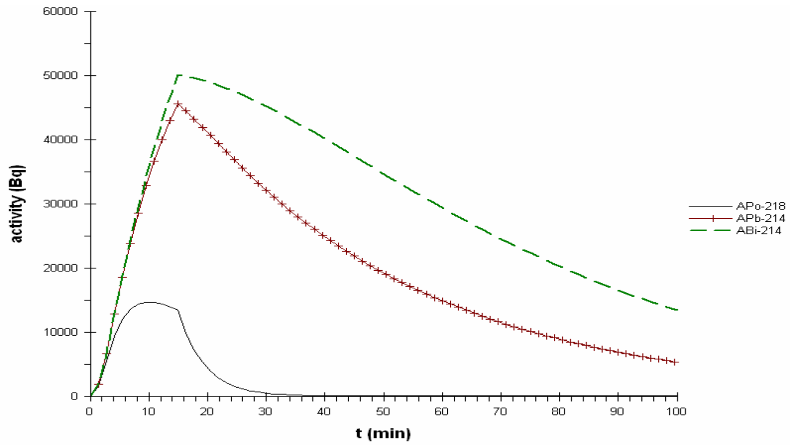


Figure 4. Calculated activities of the progenies attached to aerosol particles (in filters) during pumping (1-15 mins) and after pumping (15-100 mins) by the hypothesis that the attaching is an element-independent process

The analysis of measured and computer simulated results have been focused to the comparison of the initial activities of the radon daughters on the filters. Table 1. shows the ratios of the initial activities of ^{214}Pb / ^{218}Po and ^{214}Bi / ^{218}Po estimated from the experimental data and model fitting with respect to the sizes of aerosol particles. According to the results, in general, the initial activities of the ^{218}Po is higher than the initial activities of the other daughters, in all the ranges of aerosol sizes.

The activity of the daughter ^{214}Po is considered to be the same as the parent ^{214}Bi one, due to the very short lifetime of ^{214}Po.

Table 1. *The ranges of the initial activity-ratios (^{214}Pb / ^{218}Po and ^{214}Bi / ^{218}Po) with respect to the aerosol sizes [number of cases per size ranges: 2-5]*

Size ranges of the aerosol particles (nm)	**Activity ratio of ^{214}Pb / ^{218}Po (measured)**	**Activity ratio of ^{214}Bi / ^{218}Po (measured)**
60-100	0.45 – 4.41	0.15 – 2.30
100-200	0.26 – 2.20	0.03 – 1.14
400-700	1.01 – 1.71	0.33 – 1.30
700 – 1100	0.26 – 2.78	0.11 – 1.88
Ratio derived from element-independent attachment (assessed)	*3.31*	*3.68*

Conclusion

The mean values of the measured initial activity ratios of ^{214}Pb / ^{218}Po and ^{214}Bi / ^{218}Po are less than the ratios derived from model calculations (Table 1 last row), which assume element-independent attachment to particles. It means the ^{218}Po attaches *probably* in a higher amount to the aerosol particles than the other progenies in case of the equilibrium among the progenies in air.

According to our preliminary results a significant tendency between aerosol particle sizes and attachment of different progenies can not be verified.

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