Radon Characteristics Related to Dose for Different Living Places of the Human

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INDRODUCTION

Inhalation of the short-lived radon decay products, ^{218}Po, ^{214}Pb, $^{214}Bi/^{214}Po$, in homes, in the outdoor atmosphere and at work places yield the greatest amount of the natural radiation exposure of the human. In all dosimetric models the dominant parameter related to dose is the activity size distribution of the decay products in the air because the original deposition destination and amount of the inhaled activity deposited in lung depend on the particle size.

For the determination of dose a relation between the potential alpha energy concentration of the radon progeny exposure (C_p t) and dose the so-called dose conversion factor (DCF = Dose/C_p t) is needed. C_p is the potential alpha energy concentration (PAEC) of the short-lived radon decay products, and t is the time of inhalation. In the estimation of the DCF by dose model calculations the activity size distribution in terms of potential alpha energy concentration is an important input parameter. For practical reasons related to measurements the relative activity size distribution should be divided into

- relative size distribution of the unattached radon progeny,
- relative size distribution of the radon progeny aerosol,
- the unattached fraction in terms of potential alpha energy concentration (f_p).

In the following paper, the values of these three quantities, their variation and the main influencing parameters obtained from measurements in indoor air, in the free atmosphere near ground and in the air of different work places is reported. In addition, the variation of the inhalation dose per unit exposure for the different living places of man was estimated, taking into account the progeny characteristics and a modified human repiratory tract model of ICRP 66.

UNATTACHED FRACTION

In air, the radon decay products exist in two forms: (1) as unattached fraction, or (2) attached to the surface of aerosol particles. After their formation the unattached decay products are predominantly positively charged. They form charged and neutral clusters by reactions with water vapour and atmospheric trace gases in the air.

Theoretical considerations show that the dominant parameter which influences the fraction of the unattached radon progeny is the attachment rate $X = \int_0^\infty \beta(d) \frac{\partial Z(d)}{\partial d} \partial d$ to the atmospheric aerosol where the number size distribution is $\frac{\partial Z(d)}{\partial d}$ and the particle concentration is $Z = \int_0^\infty \frac{\partial Z(d)}{\partial d} \partial d$ (1). ß(d) is the attachment coefficient describing the attachment probability to an aerosol particle with the diameter d.

The unattached fraction of the short-lived Radon decay product j with the decay constant λ_j is

$$f_j = \frac{C_j^f}{C_j} = \frac{C_j^f}{\left(C_j^f + C_j^a\right)} \qquad (1)$$

with j = 1: ^{218}Po, j = 2: ^{214}Pb, j = 3: $^{214}Bi/^{214}Po$. C_j^f, C_j^a are the activity concentrations of the unattached and aerosol-attached decay products.

Neglecting the activity losses by plate-out on surfaces and because for almost all environmental air conditions we have λ_j < X (1, 2), the unattached fractions f_j can be written approximately

$$f_j = \frac{\lambda_1}{X}, \qquad f_j = \frac{\lambda_2 \ R_1}{X}, \qquad f_j = \frac{\lambda_2 \ \lambda_3 \ R_1}{X^2}. \qquad (2)$$

With the potential alpha energy concentration $C_p = \sum_{j=1}^{3} k_j\, C_j$ is

$$f_p = \frac{C_p^f}{C_p} \approx \frac{k_1\,\lambda_1 + k_2\,\lambda_2\,R_1}{X} = \frac{k_1\,\lambda_1 + k_2\,\lambda_2\,R_1}{\overline{\beta}\,Z} = \frac{414}{Z\left[cm^3\right]} \tag{3}$$

with k_1 = 0.105, k_2 = 0.516, and the recoil factor R_1 = 0.8, which is the fraction of the desorption from the particle surface after the alpha decay of ^{218}Po. $\overline{\beta} = 1{,}4\mathrm{x}10^{-6}$ cm^3/s is the average attachment coefficient for the atmospheric aerosol obtained from measurements (1).

The results of measurement at different places with different aerosol sources are illustrated in Fig. 1 (3, 4). The measured fp-values as function of the particle concentration Z measured by means of a condensation nuclei counter (CNC) agree well with the values calculated with equation (3) (solid line in Fig. 1).

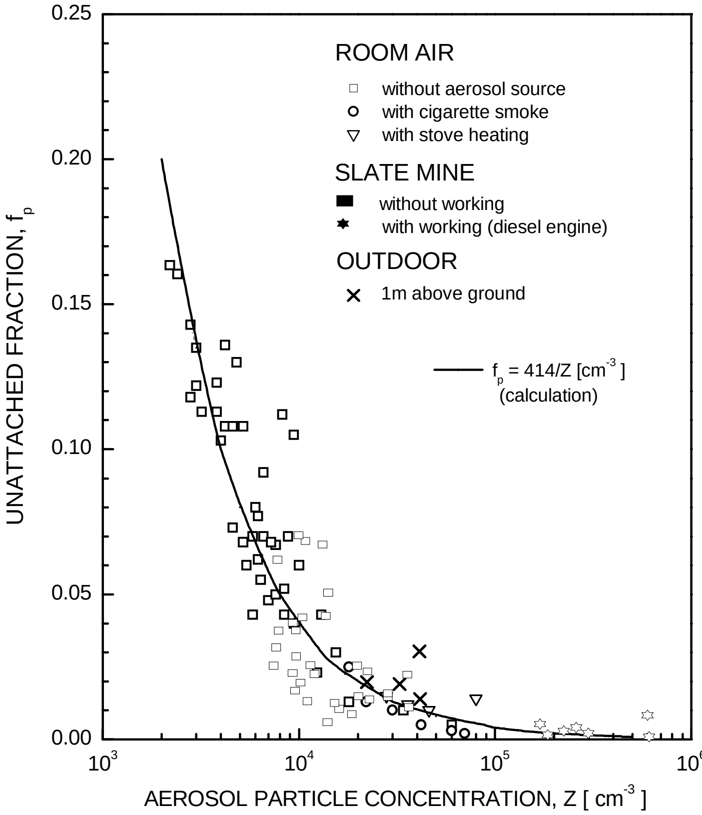

Fig. 1: Unattached fraction of the radon decay products measured at different places as function of the aerosol particle concentration

Many working places have aerosol sources due to human activities and combustion and technical processes with a high particle concentration, Z > 4 x 10^4 particles/cm^3, and therefore f_p-values below 0.01. The f_p-values are higher than 0.1 for places with particle concentrations <4 x 10^3 particles/cm^3. This is the case for example in poorly ventilated rooms (ventilation rate < 0.5 h^{-1}) without additional aerosol sources, rooms with an operating air cleaner, and low ventilated underground caves.

SIZE DISTRIBUTION OF THE PROGENY CLUSTER

The relative activity size distribution of the unattached radon progeny cluster as function of the diameter d depends on the concentration of water vapour, trace gases and the electrical charge distribution of the radionuclide in the atmospheric air. The cluster size is determined via measurement of the diffusion coefficient of the progeny cluster and therefore described with the diffusion equivalent diameter.

The measurement of the size distribution of the unattached decay products cluster (d<10 nm) under realistic environmental conditions is difficult. Only some data of measurements in a few indoor places with higher radon level exist (5). In Table 1 the results of our measurements are summarised.

Table 1: The average values of the relative size distribution of the unattached radon progeny in terms of potential alpha energy concentration (PAEC). The measured values were approximated by a sum of i lognormal distributions, characterised by the activity median diameter (AMD_{ui}), geometric standard deviation (σ_{gui}) and activity fraction (f_{pui}). n = number of measurements; Z = aerosol particle concentration; C_0 = radon concentration; RH = relative humidity.

Place Z [cm^{-3}] RH [%]	n	C_0 [Bq/cm^3]	AMD_{u1} [nm]	Mode 1 σ_{gu1}	f_{pu1}	AMD_{u2} [nm]	Mode 2 σ_{gu2}	f_{pu2}	AMD_{u3} [nm]	Mode 3 σ_{gu3}	f_{pu3}
Dwellings 500 - 3000	26	800 –1500	0.6	1.1	0.13	0.85	1.1	0.35	1.25	1.1	0.52
Water supply station 11000/60%	4	10000	0.60	1.2	0.38	0.80	1.1	0.45	1.3	1.1	0.17
Water supply station 18000/100%	4	490000	0.52	1.1	0.38	0.80	1.2	0.62	-	-	-
Therapy-mine <200 ~98%	5	8000	0.57	1.2	0.39	0.9	1.1	0.61	-	-	-

In most cases the results of the relative activity size distribution measurements were approximated by a multi-modal lognormal distribution:

$$\frac{1}{C_p} \bullet \frac{\partial C_p(d)}{\partial(\log d)} = \sum_i f_{pi} \frac{1}{\log \sigma_{gi} \sqrt{2\pi}} \exp \frac{1}{2} \left(\frac{\log d - \log AMD_i}{\log \sigma_{gi}} \right)^2 \quad (4)$$

Each mode i is characterized by the activity median diameter (AMD_i), the geometric standard deviation (σ_{gi}) and the fraction of the potential alpha energy concentration of the radon decay products (f_{pi}).

In general, the relative size distribution of the unattached cluster in terms of potential alpha energy concentration obtained from measurements under “normal” conditions concerning humidity and radon concentration can be approximated with three lognormal distributions. The AMD-values are 0.60 nm, 0.85 nm and 1.3 nm (Fig. 2). In places with high radon concentration and /or high humidity the fraction with the greatest AMD-value (~ 1.3 nm) was not registered (Fig. 3). An explaination is that almost all of the clusters are neutral in air with such conditions, because we know from chamber studies, that the neutralisation rate increases with higher humidity and radon concentration in air (6).

SIZE DISTRIBUTION OF THE PROGENY AEROSOL

Besides the cluster formation the radon decay products attach to the existing aerosol particles within 1 – 100 seconds, forming the radioactive aerosol of the radon progeny.

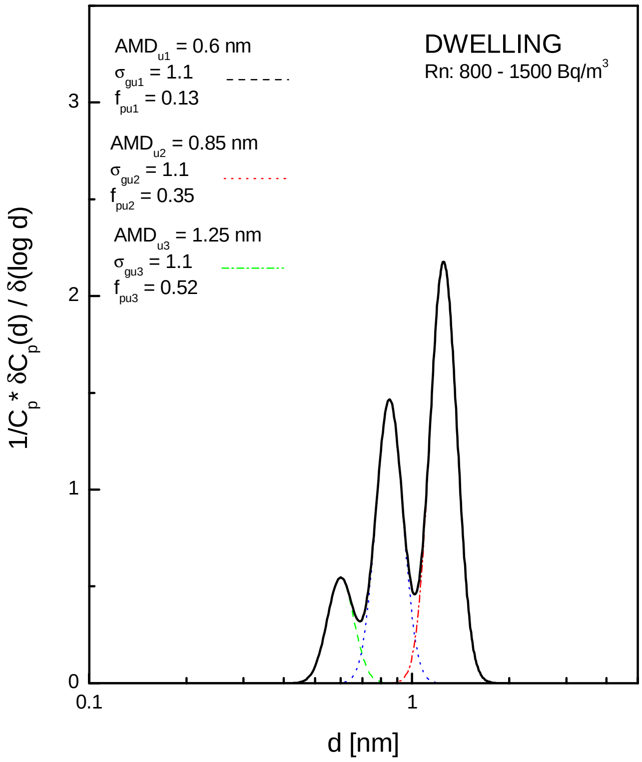

Fig. 2: Relative activity size distribution in terms of potential alpha energy concentration of the unattached radon progeny cluster measured in indoor air

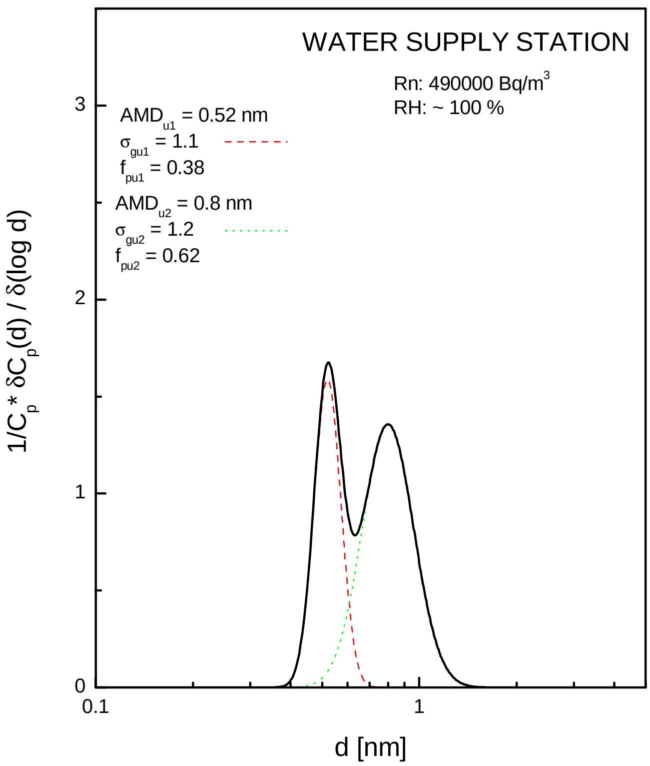

Fig. 3: Relative activity size distribution in terms of potential alpha energy concentration of the unattached radon progeny cluster measured in a water supply station with high radon concentration and high humidity

The activity size distribution $\partial C_j(d)/\partial d$ of the radon decay product j and the number size distribution $\partial Z(d)/\partial d$ size distributions are different (Fig. 4), because the attachment probability ß(d) is a function of the particle diameter d. In spite of the different number size distribution in room air by different aerosol sources, the activity size distribution similar. In all cases, most of the activity is attached on particles of the accumulation size range (100 nm – 1000 nm). The correlation between both size distributions can be expressed by (1)

$$\frac{1}{C_j}\frac{\partial C_j(d)}{\partial d} = \frac{\beta(d)}{X}\frac{\partial Z(d)}{\partial d}, \qquad (5)$$

where C_j is the activity concentration.

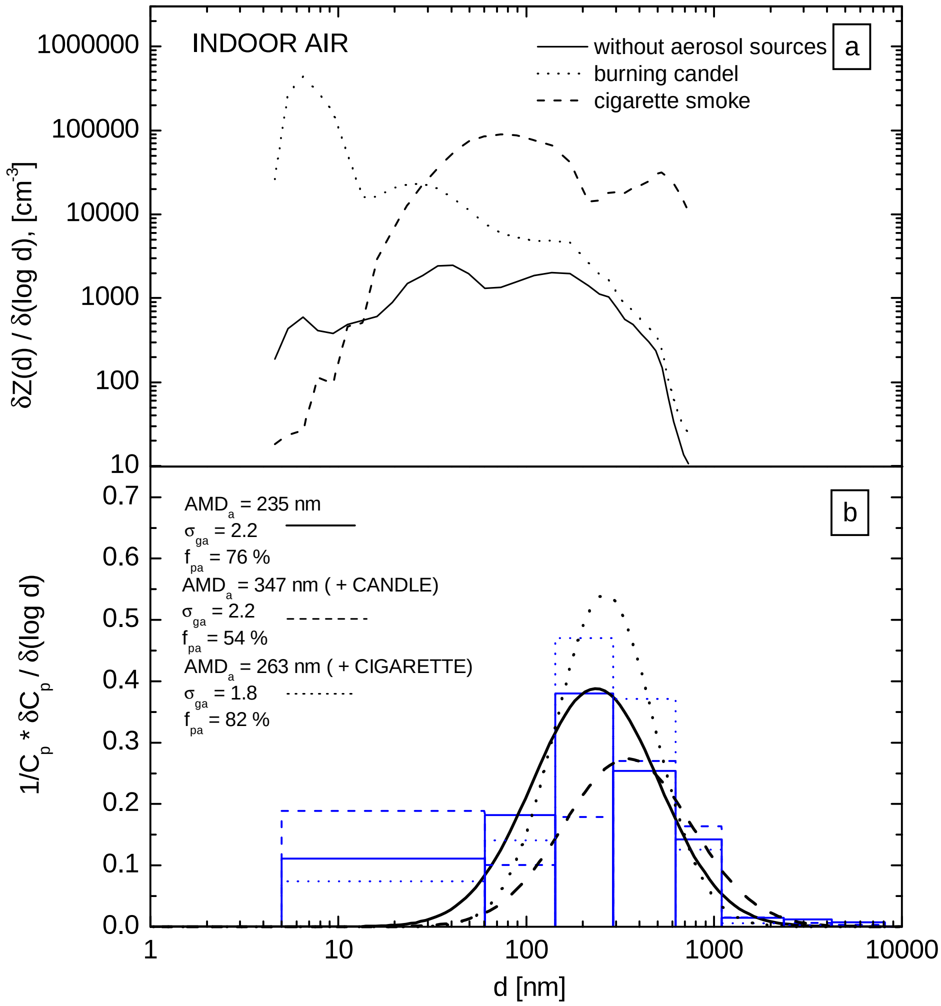

Fig. 4: The number and the activity size distributions obtained by side by side measurements in a room with different aerosol sources

The results of simultaneous measurement of all three short-lived decay products by means of an online alpha cascade impactor show (7), that the differences between the individual size distributions is small (Fig.5). Based on this result the relative aerosol size distribution of PAEC

$$\frac{1}{C_p}\frac{\partial C_p}{\partial d} \approx \frac{1}{C_j}\frac{\partial C_j}{\partial d} \qquad (6)$$

required for dose estimation can be obtained from an aerosol activity size distribution measurement of one of the short-lived decay products.

The results of the activity size distribution measurements carried out at different places in the last years are summarised in Table 2 (8, 9, 10,11); besides the average values the range of the measured values is registered.

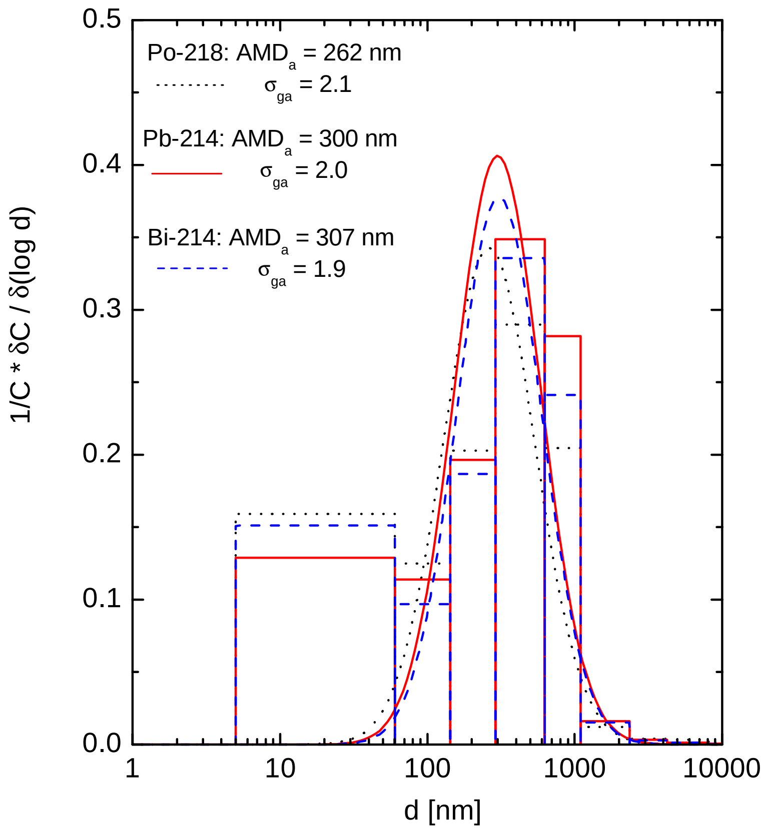

Fig. 5: Activity size distribution of ^{218}Po, ^{214}Pb, and ^{214}Bi measured with the online alpha cascade impactor

Table 2: Parameters of the activity size distribution of the aerosol attached short-lived radon progeny in the air of different places of man: Activity median diameter = AMD; geometric standard deviation = σ_{gi}; fraction of the mode = f_{pi}. The indices i = n, a and c represent the nucleation, the accumulation and the coarse mode, Z = particle concentration

Place/ Z [10^3 cm^{-3}]	Nucleation mode			Accumulation mode			Coarse mode		
	AMD_n [nm]	σ_{gn}	f_{pn}	AMD_a [nm]	σ_{ga}	f_{pa}	AMD_c [nm]	σ_{gc}	f_{pc}
Outdoor air 10 - 70	30-40	1.9-2.2	0.3 (0.2-0.4)	310 (250-450)	2.1 (1.8-3.0)	0.63 (0.4-0.8)	3000 (2000-6000)	1.7 (1.6-2.0)	0,02 (0-0.10)
Dwelling 2 - 500	20-40	1.7-2.1	0.3 (0 – 0.4)	210 (120-350)	2.2 (1.6-3.0)	0.7 (0.6-1)			
Work places 10 - 500	15-40	1.6-2.2	0.3 (0.2-0.5)	300 (150-450)	2.5 (1.8-4.0)	0.55 (0.3-0.8)	5000 (3000-8000)	1.8 (1.1-2.8)	0.15 (0-0.3)

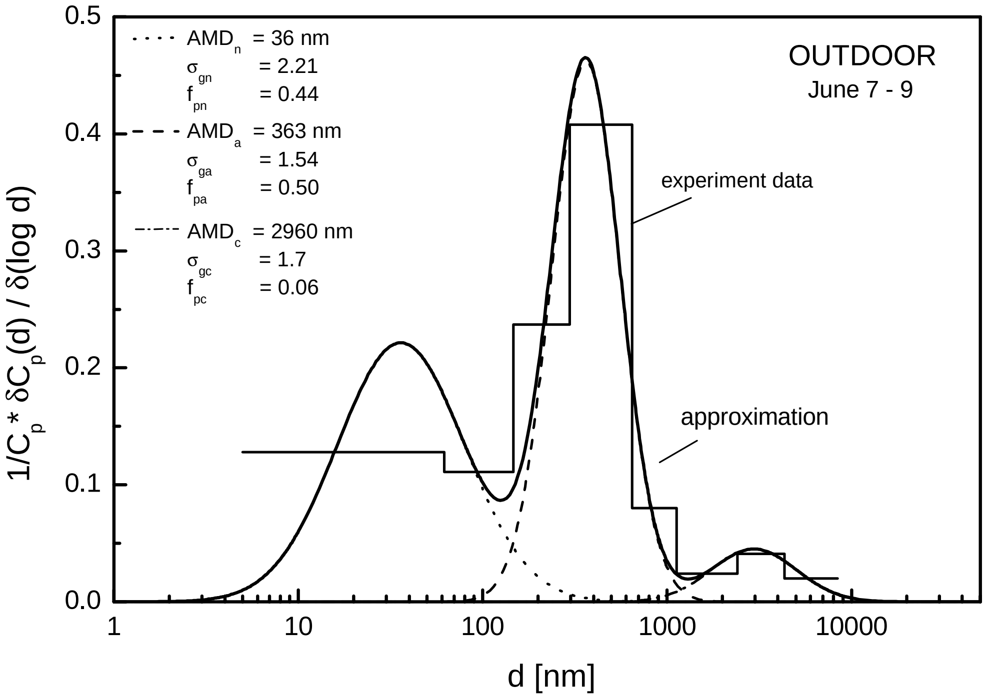

Fig. 6: The average activity size distribution of the radon progeny aerosol in outdoor air for the measurement period June 7 – 9.

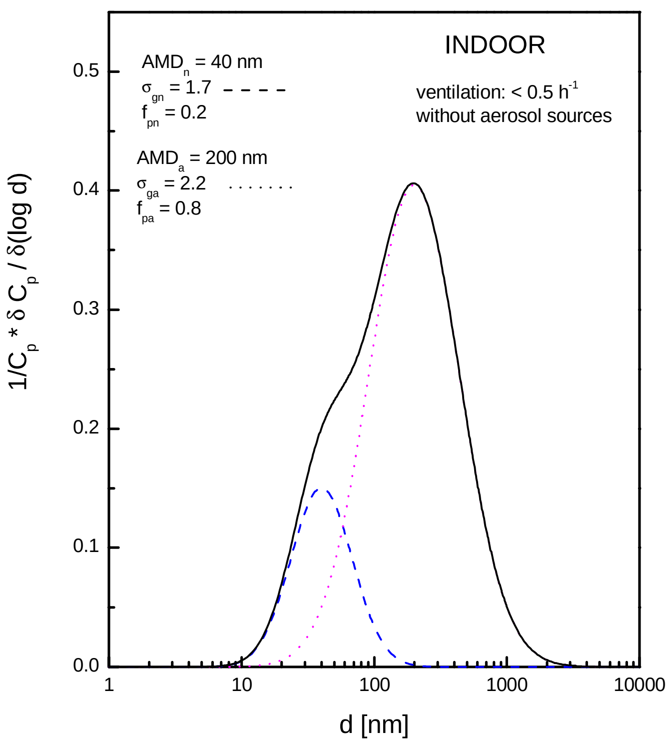

Fig. 7: A typical relative activity size distribution of the potential alpha energy concentration of the radon progeny aerosol in room air

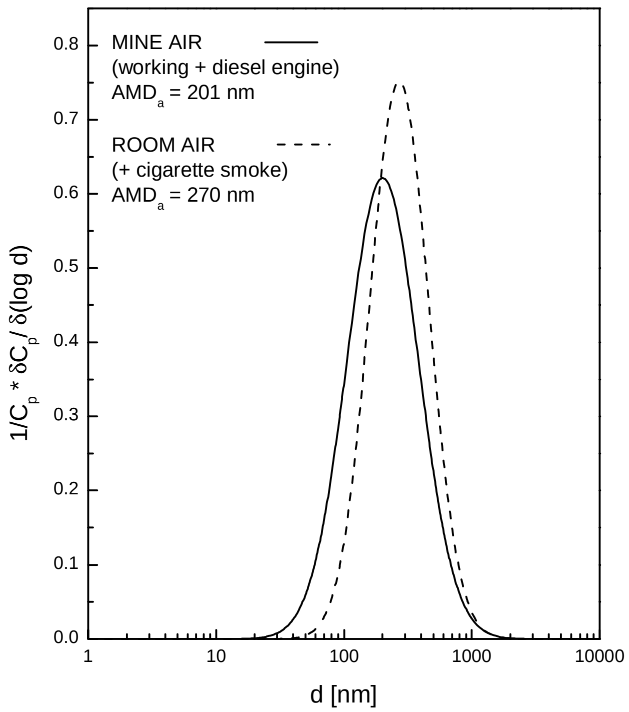

Fig. 8: Relative activity size distribution of the radon progeny aerosol in air containing combustion aerosol from diesel engines and cigarette smoke

In general, the relative activity size distribution of the radon daughter aerosol can be described by three modes, as illustrated by the example for outdoor air (Fig. 6). There is the nucleation mode (AMD_n values: 30 nm - 40 nm), the accumulation mode (AMD_a values: 250 nm – 450 nm), and the coarse mode (AMD_c values: 2000 nm – 6000 nm). The greatest activity fraction of the outdoor air is in the accumulation mode with an average f_{pa} = 0.63. There is a variation of the AMD_i, σ_{gi}, and f_{pi} values (Table 2) obtained from a continuous measurement over three weeks, however the correlation with the weather parameters, aerosol concentration, and differences between day and night hours was not significant (10).

Compared with outdoor air the AMD_a of the accumulation mode in low ventilated rooms (ventilation rate <0.5 h^{-1}) and without additional aerosol sources like cigarette smoking or cooking is shifted to smaller sizes (200 nm) (Fig. 7). In addition, under these conditions the coarse mode of the size distribution is not significant which can be explained by the greater plate-out rate of big aerosol particles on room surfaces.

The results of the measurements carried out at 19 different work places show also activity size distributions with three modes (Table 2). Remarkable is the great variation of the activity fractions of the coarse aerosol particles. The f_{pc}-values vary between 0% and 30%.

In most places with one dominating aerosol source, e.g. cigarette smoking or combustion aerosol by diesel engines, the measured activity size distribution can be approximated by a single lognormal distribution, as is demonstrated by the examples of mine air and cigarette smoking in Fig. 8. The contributions of PAEC in the size ranges of nucleation particles and coarse particles are then generally small.

DOSE CONVERSION FACTORS

The calculation of the dose by inhalation of the radon progeny is based on a lung model with the structure that is related to the recommended ICRP (ICRP 66) respiratory tract model (12, 13). The relative size distribution and the unattached fraction of the radon decay products were the important input quantities for the calculation of the effective dose per unit exposure, the dose conversion factor (DCF).

The results of these calculations show the strong influence of the size distribution and the unattached fraction (f_p) of the radon progeny cluster on dose. Especially important is the f_p-value which shows a large variation for different categories of living places (14).

Table 3: Average dose conversion factor (DCF) for the inhalation of unattached (DCF_u) and aerosol attached (DCF_{ae}) radon decay products in air of human living places arranged according to aerosol conditions. $w_{BB} : w_{bb} : w_{AI} = 0.80 : 0.15 : 0.05$ is the relative cancer sensitivity distribution of the bronchial (w_{BB}), bronchiolar (w_{bb}) and alveolar (w_{AI}) regions of the thoracic lung, v = inhalation rate, Z = particle concentration of the aerosol.

PLACE	PARTICLE CONCENTRATION Z [10^3 cm^{-3}]	NOSE BREATHING v[m^3/h]	DCF_u [mSv/WLM]	DCF_{ae} [mSv/WLM]	DCF = $DCF_u + DCF_{ae}$ [mSv/WLM]
OUTDOOR AIR	20 - 40	1.2	1.5	8.2	9.7
DWELLINGS	5 - 40	0.75	2.4	4.9	7.3
	40 - 500	0.75	0.2	4.0	4.2
WORK PLACES	1 - 10	1.2	7.0	6.0	13.0
	10 - 50	1.2	1.5	5.2	6.7
	50 - 500	1.2	0.5	5.2	5.7
	50 - 500	1.7	0.7	6.5	7.2

Because of the direct correlation between the unattached fraction and the aerosol particle concentration it is reasonable to differentiate the living places of man according to their particle number concentration of the aerosol. Table 3 summarises the DCF-values and the dose fractions of the unattached (DCF_u) and the aerosol attached (DCF_{ae}) radon progeny for outdoor air, for dwellings and for work places. All values are in effective dose. For all places nose breathing was asumed and the relative sensitivity between bronchial (w_{BB}), bronchiolar (w_{bb}) and alveolar interstitial (w_{AI}) regions of the thoracic lung was taken as $w_{BB} : w_{bb} : w_{AI} = 0.80 : 0.15 : 0.05$ (10).

Caused by the different aerosol conditions of the places the range of DCF is mainly determined by the variation of DCF_u, which varies between 0.2 – 7.0 mSv/WLM. The dose conversion factor of the radon progeny aerosol can vary over 4.0 – 8.2 mSv/WLM. At workplaces with low particle concentration ($<10^4$ particles/cm^3) the dose fractions per unit exposure of the radon progeny aerosol and the unattached cluster are about equal.

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