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Assessment of the unattached fraction of indoor radon progeny and its contribution to dose: a pilot study in China

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Abstract

The unattached fraction of radon progeny (f_p) is one of the most important factors for accurate evaluation of the effective dose from a unit of radon exposure, and it may vary greatly in different environments. For precise evaluation of the indoor radon exposure dose and the influence of unattached radon progeny, a pilot survey of f_p in different environments was carried out in China with a portable and integrating monitor. The dose conversion factors for radon progeny are calculated with LUDEP® code, and the dose contributions from the unattached and the attached radon progenies were simultaneously evaluated based on the results of field measurements.

The results show that even though the concentrations of radon progeny vary significantly among different indoor environments, the variations of f_p seem relatively small (9.3–16.9%). The dose contribution from unattached radon progeny is generally larger (30.2–46.2%) in an indoor environment.

(Some figures may appear in colour only in the online journal)

1. Introduction

Radon (^{222}Rn) is a naturally existing radioactive gas, with a half-life of 3.8 days, which can decay into a series of short-lived progeny (^{218}Po, ^{214}Pb and ^{214}Bi). Those progeny could be inhaled into human respiratory system and cause internal exposure. Airborne radon progeny exists in two forms. Those attached to aerosols are called 'attached' radon progeny, with a diameter usually larger than 5 nm. The others are called 'unattached' radon progeny, with a diameter generally less than 5 nm. Due to their small size and high diffusion coefficient, unattached radon progeny could more effectively deposit in the bronchial region of the respiratory system, and consequently give a higher exposure dose per unit of airborne activity than that of the attached particles. Almost all dosimetric models dealing with radon inhalation exposure dose stress the importance of unattached radon progeny [1–3].

The ratio of unattached radon progeny to the total radon progeny is usually called the 'unattached fraction', a term which originated from the work of Chamberlain and Dyson who described the deposition behaviour in a model of the human trachea for unattached radon progeny [4]. More and more attention has been paid to this concept accompanying the development of techniques for the measurement of f_p [5, 6].

For measuring the unattached fraction of radon progeny (f_p), three techniques (the cylindrical tube method, the parallel-plate method and the single wire screen method) have been developed. Of these, the single wire screen method has been widely adopted because of its simplicity [7–9]. In this method, the unattached radon progeny are collected on a single layer wire screen, while attached particles (or total radon progeny) are collected on a back-up filter. The collection efficiency of the screen can be calculated according to the screen collection theory derived by Cheng and Yeh [10]. Then f_p could be calculated from the alpha activities collected by the wire screen as well as the back-up filter.

In different living/working environments, different aerosol conditions and ventilation rates may cause a variation of f_p, and then lead to quite different dose conversion factors (DCFs), which is defined as the dose to the respiratory tract per unit exposure of radon progeny [11]. The International Commission on Radiological Protection (ICRP) in 2009 suggested that 'Dose coefficients will be given for different reference conditions of domestic and occupational exposure, taking into account factors including inhaled aerosol characteristics and between radon and its progeny. Sufficient information will be given to allow specific calculations to be performed in a range of situations' [12]. This point of view has been confirmed again in ICRP Publication 115 [13].

To get a initial assessment of the unattached fraction of radon progeny in different environments in China, a series of field measurements of the f_p and equilibrium-equivalent radon concentration (EEC_{Rn}) in different indoor environments were carried out using a portable and integrating monitor newly developed by ourselves [14]. Initial results for the unattached fraction of radon progeny are shown here and the dose contributions of unattached radon progeny are calculated.

2. Materials and methods

2.1. Measuring device

Field measurement of f_p was carried out using a portable and integrating monitor [14]. The monitor is composed of an unattached radon progeny integrating sampling unit (URPISU), an integrating flow meter and a mini pump (MP-300, SIBATA, Japan). A picture of the system and a cross-section diagram of the URPISU are shown in figure 1. The URPISU is made of stainless steel, with a ⌀20 mm air inlet in the lower part and one outlet at the top. There are several parts in the unit: a 635-mesh wire screen (wire diameter 20 μm, thickness 44 μm, solid volume fraction 0.343) and a piece of circular CR-39 [3] placed on the lower part; four pieces of square CR-39 placed on the periphery of the unit; and a glass microfibre filter placed near the outlet at the top. Ambient air can be sucked into the unit by a pump; the air flow passes the screen first and then passes the filter without being affected.

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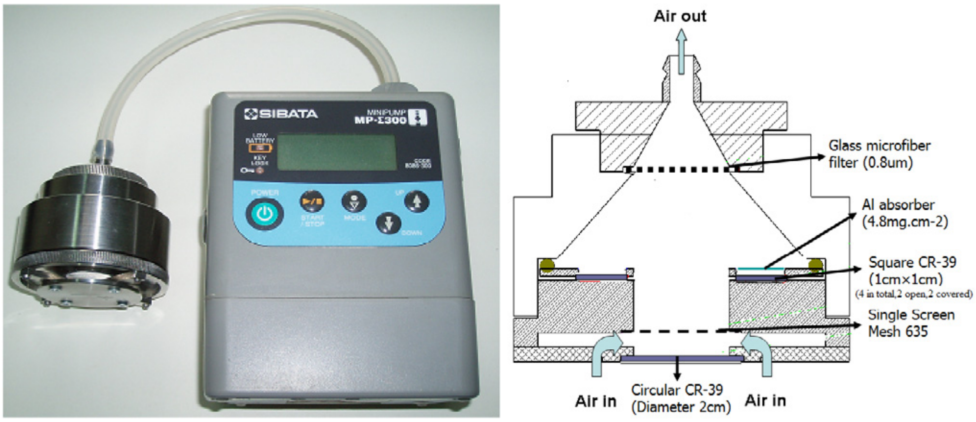

Figure 1. Picture of the system and cross-section diagram of the URPISU.

With a flow rate of 2.92 l min^{-1}, the half cut-off diameter of the 635-mesh wire screen is 5 nm. The unattached radon progeny are collected on the screen and the alpha particles emitted from them are recorded by the circular CR-39. While the attached radon progeny and thoron progeny are collected on the filter, those alpha particles emitted from the surface of the filter can be recorded by the four pieces of square CR-39 (1 cm × 1 cm). In order to discriminate ^{220}Rn progeny from ^{222}Rn progeny, absorbers with area density of 4.8 mg cm^{-2} are located on two pieces of square CR-39s to make only alpha particles with the highest energy (8.78 MeV, emitted from ^{212}Po) impinge on the two pieces of CR-39 while the remaining two pieces of square CR-39 are open.

The equilibrium-equivalent concentrations of both the unattached and attached ^{222}Rn progeny (EEC^{u}_{Rn} and EEC^{a}_{Rn}) as well as the attached ^{220}Rn progeny (EEC_{Tn}) can be simultaneously measured with the monitor. The lower limits of detection for EEC^{u}_{Rn}, EEC^{a}_{Rn} and EEC_{Tn} were estimated to be 0.03 Bq m^{-3}, 0.12 Bq m^{-3} and 0.07 Bq m^{-3}, respectively, for 24 h integrating sampling. Due to the actual field measurement limit, those lower limits of detection were 0.12 Bq m^{-3}, 0.48 Bq m^{-3} and 0.28 Bq m^{-3}, respectively, for 3 h field sampling.

2.2. Dose evaluation

The unattached fraction and the size distribution of radon progeny are the key parameters for determining DCFs for radon exposure dose evaluation. For the size distribution of radon progeny in the indoor environment, the radon progeny present a lognormal distribution with a typical activity median aerodynamic diameter (AMAD) of 200 nm as well as geometric standard deviation (GSD) of 2.2 [15]. In this paper, we tried to evaluate radon lung dose by combining the typical value of indoor radon progeny size distribution and the measurement results of f_p from the field survey. Therefore, the effective dose (E_{dose}) is defined as

$$E_{Dose} = [DCF^{U} f_p + DCF^{A}(1 - f_p)] \times 7000\ h \times EEC_{Rn}$$

where DCF^{U} and DCF^{A} are the dose conversion factors for unattached and attached radon progeny, respectively [nSv (Bq m^{-3} h)$^{-1}$].

In fact, unattached radon progeny also have their own particle size distribution. The typical value is reported to be 0.5 to 3.0 nm, with an averaged AMAD of 0.9 nm and GSD of 1.3 [16].

Table 1. Calculated DCF for unattached and attached radon progeny [nSv (Bq m^{-3} h)$^{-1}$].

	ET region	BB + bb + AI region	Total
Unattached	28.92	99.83	128.75
Attached	1.59	28.78	30.38

The DCF was calculated using the Lung Dose Evaluation Programme (LUDEP® code), which was developed by National Radiological Protection Board (NRPB) based on the ICRP 66 model. It is possible for users to calculate internal exposure from inhaled radioactive nuclides at defined environmentally related coefficients and physiologically related factors [17].

According to the recommendation of National Research Council in its 1991 report, the ratios of radon progeny were set as ^{218}Po:^{214}Pb:^{214}Bi = 1:0.65:0.4 and ^{218}Po:^{214}Pb = 1:0.1 for attached and unattached radon progeny, respectively, in the general indoor environment. The reference breathing rate is 0.78 m^3 h^{-1}, and the rates of absorption of unattached and attached radon progeny into the blood are 1 h and 10 h, respectively [18]. The biokinetic models for ^{218}Po, ^{214}Pb and ^{214}Bi are PO(D), PB(D)S and BI(D). The tissue weighted factor for the extrathoracic (ET) region is selected as 0.025, and the factor of 0.12 for whole lung tissue is evenly allotted into the bronchial region (BB), bronchiolar region (bb) and alveolar–interstitial region (AI). Then the DCFs for both unattached and attached radon progeny to different regions of lung region and total lung can be calculated. The results are listed in table 1.

As shown in table 1, the main doses are distributed in the BB, bb and AI regions for exposure to both attached and unattached radon progeny. Furthermore, DCFs for unattached radon progeny are much bigger than for attached radon progeny. The results of the calculation of DCF for radon progeny are much larger than the figure of 9 nSv (Bq m^{-3} h)$^{-1}$ used nowadays. However, they approximate to the results calculated by James *et al* [19], as pointed out by Marsh *et al* [20]. The slight difference is considered as due to the different particle size distribution and other related parameters adopted for the calculations.

3. Results and discussion

3.1. Field measurement results

To get some knowledge of f_p and EEC_{Rn} as well as EEC_{Tn} in different environments, a series of field surveys were carried out in different indoor environments for both urban and rural areas as well as mines and outdoors in six provinces (Beijing, Shanxi, Guangdong, Guangxi, Yunnan and Xinjiang) in China from April to December 2010. Due to the unattached fraction being influenced by the concentration of aerosol, bedrooms or offices without additional aerosols (like aerosols from smoking, cooking and so on) were chosen for the field survey (and when unoccupied by people). Brick houses in rural areas were chosen for comparison with the urban environment, except those houses in 'YN-GJ' where houses were made of mud and stone.

During indoor measurement, the measurement device was placed in the middle of the room, and sampled for 3 h, and after that it was airproofed with a a plastic bag for 3 days. While outdoors, the device was placed 1 m above the ground or on the top of a building. Three coal mines in Xinjiang Province were chosen for comparison. Those measurements were carried out in the rest time during a working day with the aerator system on for safety. The field measurement results are summarised in table 2.

As shown in table 2, the concentrations of both radon and thoron progeny vary with different indoor or outdoor environments. However, the variations of f_p seem relatively small for both indoor and outdoor environments. A conceivable reason is that our device is based on

Table 2. Results of field measurement for the unattached fraction of radon progeny.

		Sample number	Measurement results[a]		
			EEC_{Rn} (Bq m^{-3})	EEC_{Tn} (Bq m^{-3})	f_p (%)
City	Office	4	15.34 ± 5.15 (10.99–20.19)	1.72 ± 0.82 (0.36–3.01)	10.86 (8.34–15.69)
	Basement	1	224.38 ± 27.95	2.27 ± 0.93	9.27
	Bedroom	2	15.60 ± 3.42 (13.50–17.70)	1.29 ± 0.54 (0.54–1.84)	10.89 (10.28–11.50)
Countryside	North area	2	4.45 ± 1.01 (4.11–4.79)	0.62 ± 0.39 (0.18–1.06)	11.13 (9.94–12.31)
	South area GD-ZQ	4	28.88 ± 11.81 (15.80–45.15)	2.58 ± 0.36 (1.16–4.99)	16.87 (12.50–23.19)
	South area GD-LD	3	49.85 ± 14.49 (39.36–58.31)	2.69 ± 0.49 (1.99–3.80)	10.26 (8.66–11.28)
	South area GX-CW	2	73.66 ± 20.97 (52.28–95.03)	2.62 ± 0.51 (2.56–2.67)	10.72 (10.39–11.30)
	YN-GJ	8	93.59 ± 31.76 (52.67–182.84)	6.18 ± 1.13 (3.77–7.96)	14.99 (8.09–22.02)
Outdoor[b]		4	13.48 ± 5.77 (9.07–31.79)	0.46 ± 0.21 (0.16–0.62)	11.58 (10.57–14.25)
Mine		3	15.08 ± 3.98 (11.54–20.42)	1.12 ± 0.17 (0.93–1.35)	19.60 (18.20–20.60)

[a] In the table the values are the average value with variation range in parentheses.

[b] Measurements were carried out only in the South area. .

integral sampling, and the measurements cannot well reflect the instant fluctuations of aerosol concentration in the f_p values.

The indoor f_p varies from 9.3 to 16.9%, with typical values of 10.9 and 12.8% for urban and rural indoor environments, respectively. The f_p values in indoor environments are quite similar to the results of Canoba's measurements in dwellings without additional aerosol sources in Argentina (f_p: 9%–29%) [21] and also Tokonami's continuous measurement results in dwellings with air circulating systems in Japan (f_p: 11%) [22], a little larger than Reineking's measurement results (f_p: 9%) in Germany [23] and El-Hussein's results for indoor air in the cellar of the Physics Department of El-Minia University, Egypt (f_p: 6 ± 0.5%) [24] as well as Chen's survey results in 14 dwellings in Kaohsiung, Taiwan (f_p average 5.5%, from 2.7 to 13.1%) [25], but much smaller than Huet's 1-year measurement in an old dwelling situated in Brittany (f_p average 31%, from 8 to 67%) [26]. These differences are mainly a result of differing environmental factors, especially the aerosol concentration and ventilation rate. Dwellings in cities usually have more complicated aerosol sources and lower ventilation rates than those in the country, so the typical value for urban indoor environments is smaller than for rural indoor environments.

The outdoor f_p varies from 10.6 to 14.3%, with typical values of 11.6% in the south of China; there seems to nearly be no difference from the indoor results. Those results are quite consistent with Wasiolek's measurements using a single layer screen method in the atmosphere (f_p: 12 ± 4%, from 6 to 22%) in central New Mexico, USA [27], larger than Reineking's ($f_p \sim 2\%$) in Germany [23] and El-Hussein's results in Egypt (f_p: 6.84%) [28]. Owing to the quite different aerosol concentrations in different outdoor environments, the outdoor f_p might show quite large differences.

Our field survey results from the mine (f_p: 19.6%, from 18.2% to 20.6%) are somewhat larger than Stranden's results in mine atmospheres (f_p: 5.9 ± 3.8%) [29] and a little larger than Shimo's survey in tunnel air (f_p: 12%, from 9 to 14%) [30]. The main reason is that our

Table 3. Radon exposure calculated according to field measurement results.

		Effective dose		
		^{222}Rn exposure (mSv year^{-1})	^{220}Rn exposure (mSv year^{-1})	Contribution of unattached radon progeny (%)
City	Office	4.41 (2.97–6.47)	0.48 (0.10–0.84)	34.05 (27.83–44.09)
	Basement	62.04	0.64	30.22
	Bedroom	4.49 (3.83–5.17)	0.36 (0.15–0.52)	34.12 (32.69–35.51)
Countryside	North area	1.29 (1.16–1.42)	0.17 (0.05–0.30)	34.67 (31.87–37.30)
	South area GD-ZQ	9.50 (4.72–16.81)	0.72 (0.32–1.40)	46.24 (37.71–56.13)
	South area GD-LD	14.12 (10.72–16.93)	0.75 (0.55–1.06)	32.64 (28.66–35.02)
	South area GX-CW	21.10 (14.86–27.60)	0.73 (0.72–0.75)	33.72 (32.95–35.06)
	South area YN-GJ	29.56 (14.13–66.61)	1.73 (1.06–2.23)	42.77 (27.17–54.48)
Outdoor		3.94 (2.59–9.88)	0.13 (0.04–0.17)	35.69 (33.37–41.32)
Mine		5.24 (3.09–7.24)	0.31 (0.26–0.38)	50.82 (48.53–52.37)

measurements were carried out during the break time in each coal mine, when the diesel engines were shut down and the aerator systems were still on for safety, so the aerosol concentration in the laneways of the mines was quite low during the sampling time. As shown before, the unattached fraction becomes larger with decrease of the aerosol concentration [11]. So at break times, the unattached fraction of radon progeny in mines is observably that at higher working times, while the aerosol in the laneways mainly comes from the diesel engines.

3.2. Dose evaluation

To understand radon exposure in detail, especially the dose contribution of unattached radon progeny, radon dose evaluation was carried out by adopting the dose evaluation equation introduced above. For thoron exposure, the DCF of 40 nSv [(Bq m^{-3} h)$^{-1}$] given by the UNSCEAR 2000 report was adopted for the calculation. The results are listed in table 3.

As listed in table 2, f_p is generally less than 20% in the places surveyed. However, as shown in table 3, the unattached radon progeny contribute more than 30% of the total radon exposure dose in those places. The reason is that the DCF of unattached radon progeny is nearly four times larger than that of attached radon progeny. The smallest contribution was found in the basement (30.2%), while the largest was observed in the mine (50.8%). No significant difference was found between indoor and outdoor environments or between the houses in the city and those in the countryside, except those in 'GD-ZQ' and 'YN-GJ'. In the mine lane, both radon and thoron concentrations were not quite as high due to the high ventilation rate and possibly low natural nuclides, while the ratio of dose contribution from the unattached radon progeny in the mine lane was the highest, with the largest f_p.

4. Conclusion

The unattached fraction of radon progeny and its size distribution are important factors for accurate evaluation of radon exposure. To precisely evaluate the indoor radon exposure dose and the influence of unattached radon progeny, a pilot survey of f_p in different environments was carried out with a portable and integrating monitor and effective doses were evaluated through a dosimetric method.

Field measurement results show that the concentrations of both radon and thoron progeny largely vary with different indoor and outdoor environments, while the variations in f_p seem relatively small for both indoor and outdoor environments. The indoor f_p varies only from 9.3 to 16.9%, with typical values of 10.9 and 12.8% for urban and rural indoor environments, respectively, where the difference mainly comes from the more complicated aerosol sources and lower ventilation rates in city dwellings than those in the countryside. The outdoor f_p varies from 10.6 to 14.3%, with typical values of 11.6% in the south of China; there seems to be no difference from the indoor results. Due to sampling at break time during a working day, the measurement results in the mine (f_p: 19.6%, from 18.2 to 20.6%) are somewhat larger.

Because the DCF of unattached radon progeny is nearly four times larger than that of attached radon progeny, even though the unattached fraction of radon progeny is generally less than 20% in our living environment (from 9.3 to 16.9%), its dose contribution to the total radon exposure is larger than 30% (from 30.2 to 46.2%).

Actually the unattached fraction of radon progeny changes with time due to the variation in aerosol concentration and ventilation rate in indoor as well as mine environments. Due to the power limit, we can only get an average value over 3 h, which is not so representative of dose evaluation in real environments. A longer time survey or continuous measurement might be needed for more precise dose evaluation and to discover the change in unattached fraction of radon progeny. Considering different measuring methods and devices and also various authors' quite different measurement results, it would be appropriate to conduct an intercomparison of the measuring devices.

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