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Note

Variation of the unattached fraction of radon progeny and its contribution to radon exposure

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Abstract

The unattached fraction of radon progeny is one of the most important factors for radon exposure evaluation through the dosimetric approach. To better understand its level and variation in the real environment, a series of field measurements were carried out indoors and outdoors, and radon equilibrium equivalent concentration was also measured. The dose contribution of unattached radon progeny was evaluated in addition.

The results show that no clear variation trend of the unattached fraction of radon progeny is observed in an indoor or outdoor environment. The average unattached fraction of radon progeny for the indoors and outdoors are $(8.7 \pm 1.6)\%$ and $(9.7 \pm 2.1)\%$, respectively. The dose contribution of unattached radon progeny to total radon exposure is some 38.8% in an indoor environment, suggesting the importance of the evaluation on unattached radon progeny.

Keywords: radon progeny, unattached fraction, EEC, variation, dose evaluation

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

Introduction

Radon (^{222}Rn) is a naturally existing radioactive gas with a half-life of 3.8 d, which can decay into a series of short-life progeny (^{218}Po, ^{214}Pb and ^{214}Bi). This part of the progeny could be inhaled into the human respiratory system and cause internal exposure. The exposure of radon and its short-lived progeny contributes more than half of the natural radiation exposure received by the public [1].

Radon progeny exists in two forms. Those attached to aerosol 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. The unattached fraction of radon progeny, f_p, is usually described as the ratio of the potential alpha energy concentration of unattached radon progeny ($C_{p,u}$) to that of total radon progeny (C_p) [2]. Due to its small size and high diffusion coefficient, the 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 [3, 4]. Compared with the field measurement on radon progeny concentration, the measurement on f_p is rather limited. Marsh and Ishikawa use 8% as the representative value for the dose conversion factor (DCF) calculation [5, 6].

In 2010, the International Commission on Radiological Protection (ICRP) 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.' [7] For precise evaluation of a radon exposure dose in an indoor environment, and the contribution fraction of unattached radon progeny, a pilot survey on f_p was carried out formerly by the author's laboratory, which found that the dose contribution from unattached radon progeny is some 30% or higher than that of total radon exposure [8].

For a better understanding on the level and variation of the unattached fraction of radon progeny, a series of measurements was carried out in both indoor and outdoor environments, and the dose contribution of unattached radon progeny was also evaluated with the dosimetric method on the basis of the field measurement results.

Materials and methods

Measurement of the unattached fraction of radon progeny

For the purpose of dose evaluation, a portable measurement monitor using CR-39[3] as a detector, composed of an unattached radon progeny integrating sampling unit and a digital standard sampling pump (SIBATA Co., Japan) was adopted for the field measurements [9].

The flow rate of the sampling is $2.92\ \mathrm{l} \cdot \mathrm{min}^{-1}$ and sampling time is 24 h. After sampling, the sampling unit is left enclosed for three days to allow the complete decay of thoron progeny. Then the CR-39s are taken out and etched for 8 h at 75 °C in a 7.5 N NaOH solution. The etched tracks are counted under a microscope. The average equilibrium-equivalent concentration of unattached radon progeny ($\mathrm{EEC^u}$) and attached radon progeny ($\mathrm{EEC^a}$) can be calculated as follows.

$$\mathrm{EEC^a} = 0.1627 D_{\mathrm{Rn}}^{\mathrm{a}} \cdot t^{-1} \quad (1)$$

$$\mathrm{EEC^u} = 0.0393 D_{\mathrm{Rn}}^{\mathrm{u}} \cdot t^{-1} \quad (2)$$

where $D_{\mathrm{Rn}}^{\mathrm{a}}$ and $D_{\mathrm{Rn}}^{\mathrm{u}}$ are the net track densities produced by attached and unattached radon progeny (track.cm^{-2}); t is the sampling time in hours; 0.1627 and 0.0393 are from our former work on the development of the monitor [9], which are derived from the relationship

[3] Fukuvi Chemical Industry Co., Ltd, Sanjuhassha-cho, Fukui-city 910-37, Japan.

between the net track density and the concentration of radon progeny based on the sampling flow rate, areas of CR-39s, and total detecting efficiencies. Finally, the f_p can be given from the ratio of EEC^u to ($EEC^u + EEC^a$). The lower level detection limit for EEC^u and EEC^a are 0.03 $Bq \cdot m^{-3}$ and 0.12 $Bq \cdot m^{-3}$, respectively, for 24 h sampling time at a flow rate of 2.92 l min^{-1}.

This integrating measurement was carried out twice each month, one at the start and the other in the middle of every month throughout 2014. The indoor measurement was carried out in a meeting room of a normal office building in Beijing. The outdoor measurement was carried out on the platform at the ground level of the same building. Ambient aerosol concentration, temperature and humidity were also recorded during the measurements. In general, aerosol concentration indoors was between 10 000 and 36 000 cm^{-3} which is measured by a monitor of TSI3781 (TSI, USA). The temperature indoors as well as outdoors during the measurements was 15 °C–28 °C and −2 °C–30 °C, respectively. The humidity indoors and outdoors was 16%RH–76%RH and 26%RH–74%RH, respectively.

To obtain the knowledge of the diurnal variation of the unattached fraction, continuous measurement was performed in the same meeting room in the office building using BWLM-PLUS-2S (Tracerlab Co., German), which collected the unattached radon progeny and radon progeny on a mesh screen and a filter separately. The lower level detection limit for a 60 min cycle is 0.1 Bqm^{-3}.

Calculation of annual effective dose and the dose contribution of unattached radon progeny

Annual effective dose can be calculated by the following equation [8]:

$$E_{\text{Dose}} = [\text{DCF}^{\text{u}} \times f_{\text{p}} + \text{DCF}^{\text{a}} \times (1 - f_{\text{p}})] \times T \times \text{EEC} \tag{3}$$

where, DCF^u and DCF^a are the dose conversion factors for unattached and attached radon progeny, respectively. T is the occupancy time (7000 h indoors and 1700 h outdoors).

Taking the dose conversion factor from the study of Brudecki (94.7 nSv(Bq h $m^{-3})^{-1}$ and 14.1 nSv(Bq h $m^{-3})^{-1}$ for DCF^u and DCF^a, respectively) [10], the annual effective dose of radon progeny and the dose contribution of unattached radon progeny could be calculated as follows:

$$E_{\text{Dose}} = [94.7 \times f_{\text{p}} + 14.1 \times (1 - f_{\text{p}})] \times T \times \text{EEC} \tag{4}$$

$$F_{\text{Dose contribution}} = \frac{E_{\text{unattached radon}}}{E_{\text{Dose}}} = \frac{94.7 \times f_{\text{p}}}{94.7 \times f_{\text{p}} + 14.1 \times (1 - f_{\text{p}})} \tag{5}$$

Results and discussion

The annual variation of f_p in an indoor and outdoor environment

The results of the integrating measurement on EEC and f_p indoors and outdoors in 2014 are shown in tables 1 and 2, respectively.

The EEC of the indoor environment is 6.0 ± 2.5 $Bq \cdot m^{-3}$, which is slightly lower than that of the former result in Beijing [11]. The EEC of the outdoor environment is 6.1 ± 2.0 $Bq \cdot m^{-3}$, which is comparative with former survey results [12].

The average of f_p indoors is 8.7%, and the minimum is 6.3% which appears in June, while the maximum is 12.5% which appears in February. The average value is quite similar to

Table 1. Indoor monthly measurement results of EEC and f_p.

Month	EEC (Bq · m^{-3})	EEC^u (Bq · m^{-3})	f_p (%)
Jan	9.1 ± 0.3	0.77 ± 0.03	8.4 ± 0.5
Feb	3.0 ± 0.1	0.38 ± 0.01	12.5 ± 0.8
Mar	3.4 ± 0.1	0.33 ± 0.01	9.7 ± 0.4
Apr	3.6 ± 0.2	0.34 ± 0.02	9.7 ± 0.5
May	3.0 ± 0.2	0.28 ± 0.01	9.4 ± 0.6
Jun	7.8 ± 0.3	0.50 ± 0.02	6.4 ± 0.3
Jul	6.3 ± 0.3	0.44 ± 0.03	7.1 ± 0.5
Aug	5.2 ± 0.3	0.40 ± 0.01	7.6 ± 0.5
Sep	4.8 ± 0.1	0.47 ± 0.02	9.7 ± 0.5
Oct	6.0 ± 0.2	0.56 ± 0.03	9.4 ± 0.5
Nov	9.6 ± 0.6	0.69 ± 0.03	7.2 ± 0.5
Dec	9.9 ± 0.2	0.77 ± 0.03	7.8 ± 0.5
Mean ± SD	6.0 ± 2.5	0.49 ± 0.16	8.7 ± 1.6

Table 2. Outdoor monthly measurement results of EEC and f_p.

Month	EEC (Bq · m^{-3})	EEC^u(Bq · m^{-3})	f_p (%)
Jan	7.5 ± 0.3	0.72 ± 0.05	9.6 ± 0.6
Feb	7.7 ± 0.3	0.88 ± 0.04	11.5 ± 0.9
Mar	3.4 ± 0.2	0.44 ± 0.02	12.8 ± 1.0
Apr	5.9 ± 0.6	0.48 ± 0.03	8.2 ± 1.0
May	5.0 ± 0.3	0.24 ± 0.03	4.9 ± 0.5
Jun	6.7 ± 0.2	0.79 ± 0.03	11.9 ± 0.5
Jul	3.1 ± 0.2	0.29 ± 0.03	9.4 ± 1.1
Aug	4.8 ± 0.3	0.35 ± 0.03	7.2 ± 0.8
Sep	4.2 ± 0.2	0.46 ± 0.03	10.9 ± 0.8
Oct	7.4 ± 0.2	0.75 ± 0.03	10.2 ± 0.5
Nov	8.8 ± 0.3	0.94 ± 0.03	10.7 ± 0.5
Dec	9.3 ± 0.5	0.81 ± 0.03	8.7 ± 0.5
Mean ± SD	6.1 ± 2.0	0.60 ± 0.23	9.7 ± 2.1

Reineking's measurement results (f_p: 9%) in Germany [13], a little larger than El-Hussein's results in the cellar of the Physics Department of El-Minia University, Egypt (f_p: 6 ± 0.5%) [13] and Chen's survey results in 14 dwellings in Kaohsiung, Taiwan (f_p average 5.5%, from 2.7 to 13.1%) [14], a little smaller than Canoba's measurements in Argentina (f_p: 9%–29%) [15] and Tokonami's results with air circulating systems in Japan (f_p: 11%) [16].

Theoretically f_p is likely to be inversely proportional to aerosol concentration [17], but compared with aerosol concentration measured by TSI3781, f_p has a very weak relationship with it. That demonstrates that f_p is a result of comprehensive environmental factors.

The average outdoors f_p is 9.7%, and the minimum is 4.9% which appears in May, while the maximum is 12.8% which appears in March. This result is a little smaller than our former result outdoors (f_p: 11.58%) [9] as well as Wasiolek's measurements using a single layer screen method (f_p: (12 ± 4)%, from 6% to 22%), and larger than Reineking's (f_p ~ 2%) in Germany [12] and El-Hussein's results in Egypt (f_p: 6.84%) [13].

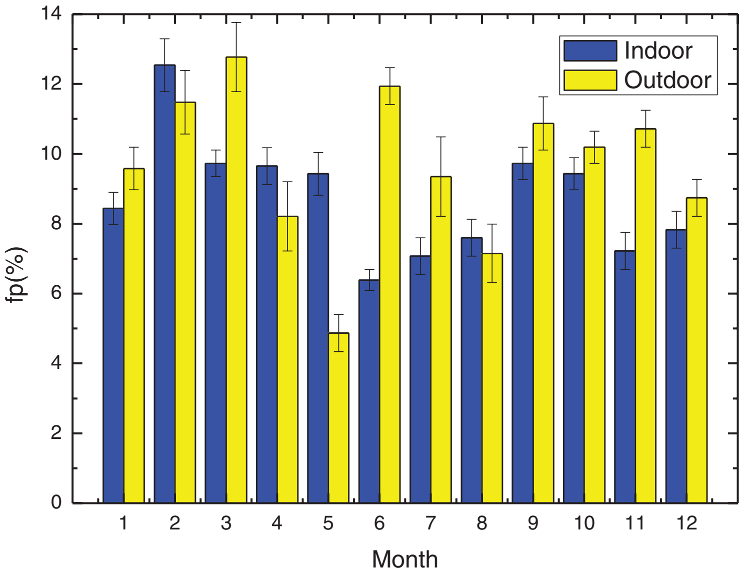

Figure 1. The unattached fraction of radon progeny in an indoor and outdoor environment.

Figure 1 shows only the result of our integrating measurement of f_p to see the general annual variation. Firstly, the average value of f_p outdoors is slightly larger than that indoors, which is much the same as our former survey results [9]. Secondly, f_p looks slightly lower in the summer season (June, July and August). In the North part of China, like Beijing, humidity is higher in the summer, and higher air humidity makes the small size aerosol particles grow larger in size, and consequently it causes a reduction in f_p values. Thirdly, for annual variation in an outdoor environment, several outdoor environmental factors influence f_p values together, and no obvious trend was observed.

The diurnal variation of f_p and EEC in an indoor environment

Continuous measurement of f_p and EEC was carried out in the meeting room using BWLM-PLUS-2S. Results of four individual days which typically represent the diurnal variation are shown in figure 2.

In the four subfigures which clearly show the change of EEC and f_p within 24 h, it is easy to observe a significant diurnal variation of radon progeny concentration, which is higher in the early morning and lower in the late afternoon. That is because radon progeny is mostly influenced by ventilation which is usually higher in the daytime and lower after midnight. Different from EEC, the unattached fraction varies from 3.86% to 14.03% within 24 h, and has no obvious diurnal variation. This can be explained by the fact that the unattached fraction of radon progeny is influenced by more environmental factors, such as aerosol concentration and size distribution as well as ventilation [18], and the level of f_p is a comprehensive result.

The average EEC value of continuous measurement is relatively higher than that of the integrating measurements; that is because they were carried out during a different period. As mentioned before, radon progeny is mostly influenced by ventilation and varies strongly with

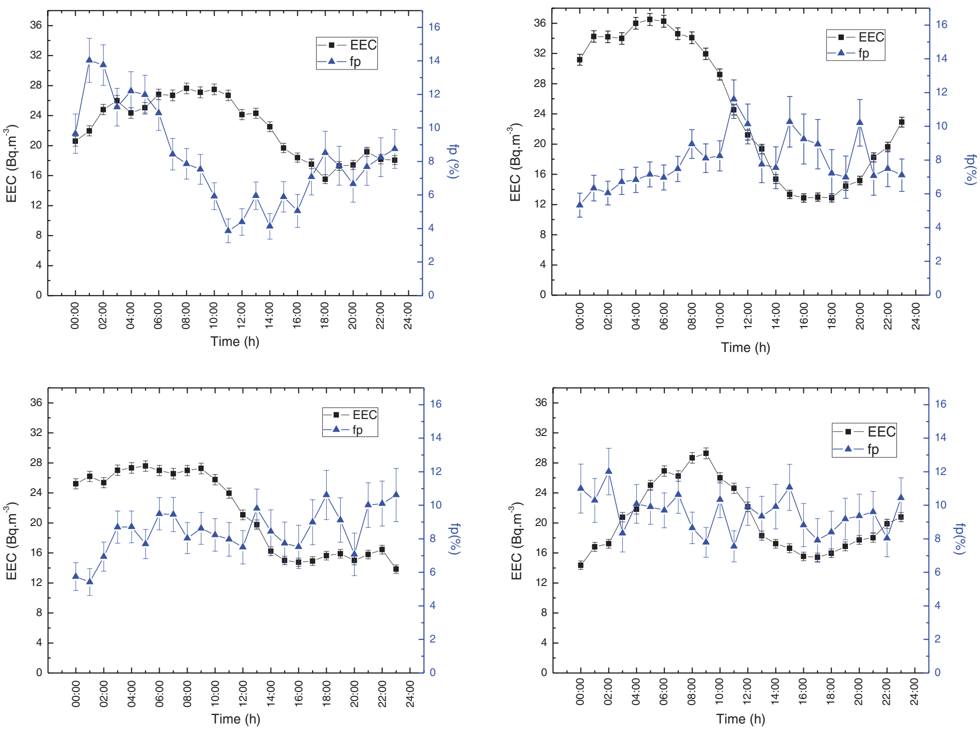

Figure 2. The diurnal variation of f_p and EEC in an indoor environment.

Table 3. Dose contribution of unattached radon progeny.

		Effective dose		
		Unattached radon progeny (mSv yr^{-1})	Total radon progeny (mSv yr^{-1})	Dose contribution of unattached radon progeny (%)
Present study	Indoors	0.328 (0.186–0.510)[a]	0.869 (0.454–1.412)	38.8 (31.5–49.3)
	Outdoors	0.096 (0.039–0.151)	0.229 (0.114–0.340)	41.3 (25.3–50.0)
Guo (2012)	Indoors			34.1 (27.8–44.1)
	Outdoors			35.7 (33.4–41.3)
Brudecki (2014)				31.5
Farkas (2014)				19.7

[a] Range of the value in parentheses.

the weather conditions outdoors, which changes greatly in Beijing. The serious air pollution caused by the stable atmosphere during the period of continuous measurement might be the reason for the relatively high EEC value.

The dose contribution of unattached radon progeny

The annual effective dose and dose contribution of unattached radon progeny are calculated on the basis of our integrated field measurement result and the result is shown in table 3.

The annual effective dose caused by radon progeny in an indoor and outdoor environment is 0.869 mSv and 0.229 mSv, respectively. The dose contribution of unattached radon progeny is 38.8% and 41.3% for indoors and outdoors, respectively, which is quite comparable with Guo's results and Brudecki's result [10], and is higher than what Farkas reported [19].

Conclusion

For a better understanding on the variation of the unattached fraction of radon progeny, a series of field measurements were carried out in both indoor and outdoor environments, and also the dose contribution of unattached radon progeny was evaluated by the dosimetric method.

Results show that the average f_p indoors and outdoors is 8.7% and 9.7%, with a variation of 6.3% ~ 12.5% and 4.9% ~ 12.8%. The dose contribution of unattached radon progeny is 38.8% and 41.3% for indoors and outdoors, respectively.

It can be concluded by the result of our work that even though the level of the unattached fraction of radon progeny is no more that 10% in a normal indoor environment, its dose contribution might be more that 30% of total radon exposure, which cannot be ignored.

More field measurements and dose evaluation of f_p will be needed in future for an improved understanding of the dose and risk implications of exposure to radon and its progeny.

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