## FIELD MEASUREMENT OF THE UNATTACHED FRACTION AND ACTIVITY CONCENTRATION RATIO OF RADON PROGENY IN TYPICAL DWELLINGS

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The unattached fraction  $(f_p)$  and activity concentration ratio of radon progeny  $(C_{218P_0} : C_{214P_b} : C_{214})$  are important for radon exposure dose evaluation. For getting these characteristic parameters in dwellings, a series of field measurement was carried out. For comparison, a semi-continuous measurement was carried out in an office room and outdoors. Results show that the average  $f_p$  is 4.5%  $\pm$  2.2% and 3.8%  $\pm$  1.7% in city dwellings and in rural dwellings, respectively. The average activity concentration ratios are 1:0.94:0.70 for radon progeny and 1:0.07:0.06 for unattached radon progeny in city dwellings, while those in rural dwellings are 1:0.88:0.66 and 1:0.09:0.07. The average values of  $f_p$  are 5.1%  $\pm$  0.9% and 5.4%  $\pm$  3.1% in the office room and in outdoors without significant difference. The average activity concentration ratios are 1:0.88:0.77 for radon progeny and 1:0.11:0.11 for unattached radon progeny in outdoors.

#### INTRODUCTION

Radon is one of the most important contributors to human exposure from natural sources, and its exposure dose mainly comes from short-lived radon progeny ( $^{218}$ Po,  $^{214}$ Pb,  $^{214}$ Bi)( $^{(1)}$ . Short-lived radon progeny usually exists in two forms, the attached form is that radon progeny is attached to the aerosol particles, while the unattached radon progeny remains as atom or clusters( $^{(2)}$ . Due to its small particle size and high diffusion, the unattached radon progeny is easily deposited into the upper respiratory tract, and result in a higher inner exposure than that of attached radon progeny is 7.3–50 times higher than that of attached radon progeny( $^{(4-6)}$ .

The fraction of unattached radon progeny concentration to total radon progeny concentration is usually defined as 'unattached fraction', which can be expressed as either the unattached fraction of specific nuclide activity concentration ( $f_i$ , i = 1,2,3 for <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi, respectively) or the unattached fraction of potential alpha energy concentration ( $f_p$ ). The ratio of <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi activity concentration is usually defined as 'activity concentration ratio', which could be divided into the activity concentration ratio of unattached radon progeny ( $C_{218Po}^{218}$ :  $C_{214Pb}^{214}$ :  $C_{214_{\text{Bi}}}^{u}$ ), the activity concentration ratio of attached radon progeny ( $C_{218_{Po}}^{a}$  :  $C_{214_{Pb}}^{a}$  :  $C_{214_{Bi}}^{a}$ ) and the activity concentration ratio of total radon progeny ( $C_{218_{Po}}$  :  $C_{214_{Pb}}$  :  $C_{214_{Bi}}$ ). The unattached fraction and the activity concentration ratio of radon progeny are important characteristic parameters for radon exposure dose evaluation.

ICRP 115 publication suggested that 'doses from radon and its progeny should be calculated using ICRP biokinetic and dosimetric models', and 'ICRP will provide dose coefficients per unit exposure to radon and its progeny for different reference conditions of domestic and occupational exposure, with specified equilibrium factors and aerosol characteristics'<sup>(7)</sup>. Through reviewing the reference aerosol parameters (unattached fraction as well as size distribution) and activity concentration ratios, ICRP 137 published the dose conversion factors of radon progeny for indoor workplace, mine and tourist cave, but the dose conversion factors for typical dwelling which is needed for public exposure dose evaluation has not been given yet<sup>(8)</sup>.

For radon progeny behaviors study and indoor exposure dose evaluation, quite many field measurements of radon progeny were carried out in dwellings or in other indoor environments<sup>(9–17)</sup>. Previous researches have shown that the unattached fraction

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and activity concentration ratio of different indoor environments are quite variable and affected by many indoor environmental conditions, like ventilation rate and aerosol concentration<sup>(18–25)</sup>. Furthermore, very limited results of the activity concentration ratio of radon progeny are given; measurement results of the activity concentration ratio of unattached radon progeny are even rarer due to its low concentration and sensitivity limitation for field survey<sup>(26–31)</sup>. Actually, the accurate measurement of each of the unattached species of the three radon progenies is difficult but is important for accurate dose evaluation<sup>(32)</sup>.

For the purpose of obtaining the characteristic parameters of radon progeny, especially the unattached fraction and activity concentration ratio of radon progeny in typical dwellings, a series of field measurement was carried out in city and rural areas in China. For comparison, semi-continuous measurement in office room and outdoors is also performed. Interesting results are found and analyzed in detail in this paper, which is quite important for dose evaluation of typical environmental.

#### MATERIALS AND METHODS

#### Instruments and methods

For field measurement, a made-in-order unattached radon progeny measurement device RPM-FF01-2S (Sairatec, China)<sup>(33)</sup> is adopted in this research.

This device has two parts, the total radon progeny measurement part and the unattached radon progeny measurement part. The first part uses 0.8 µm PTFE filter (Millipore, USA) for sampling and 400 mm<sup>2</sup> PIPS detector (SARAD GmbH, Germany) for detection. The other part uses a mesh No.635 wire screen (HeBei NOVA, China) for sampling and 400 mm<sup>2</sup> PIPS detector for detection. The sampling flowrate of both filter and wire screen is 3 1 min<sup>-1</sup>. The screen parameters (wire diameter 18.1 µm, thickness 40.0 µm and solid fraction 0.313) were given by the manufacturer. According to the screen parameters and screen collection efficiency formula<sup>(34, 35)</sup>, the half cut-off diameter (dp50) of wire screen is 4.2 nm. Two independent multichannel analyzers are used for the analysis of alpha spectrum from filter and wire screen separately, and the detection efficiency is between 22 and 23% for both devices.

During measurement, the unattached radon progeny and total radon progeny are collected on wire screen and filter separately, then alpha particles are detected by two PIPS detectors. Due to the diffusivity of unattached radon progeny, it is possible to deposit on the surface of the detector and its holder. This influence can be ignored because the amount of deposition is small.

A revised Wicke method<sup>(36)</sup> is chosen for radon progeny activity concentration measurement, and the total cycle of each measurement is 1 h. The sampling time is from 0 to 30 min, and the measurement time is 0–30 min and 40–60 min separately, with a waiting time of 30–40 min between the sampling period and the measurement period. Two separate alpha spectrums are gotten in each cycle, and the activity concentrations of <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi can be calculated from the net counts of <sup>218</sup>Po-ROIs(N<sub>1</sub>) and in the first spectrum, as well as the net counts of <sup>214</sup>Po-ROIs(N<sub>3</sub>) in the second spectrum; the calculation equations are shown as follows<sup>(36)</sup>.

$$C_{218_{\text{Po}}} = \frac{(0.14709 \cdot N_{\text{I}}) \cdot \text{SL}}{\varepsilon_{\alpha} \cdot F \cdot \varepsilon_{\text{f}} \cdot \text{FT}}$$
(1)

$$C_{214}_{\rm Pb} = \frac{(-0.017750 \cdot N_1 - 0.052133 \cdot N_2 + 0.090427 \cdot N_3) \cdot SL}{\epsilon_{\alpha} \cdot F \cdot \epsilon_{\rm f} \cdot FT}$$
(2)

$$C_{214}_{\text{Bi}} = \frac{(0.0018973 \cdot N_1 + 0.067349 \cdot N_2 - 0.028267 \cdot N_3) \cdot \text{SL}}{\epsilon_{\alpha} \cdot F \cdot \epsilon_{\text{f}} \cdot \text{FT}}$$
(3)

where  $C_{218Po}$ ,  $C_{214Pb}$  and  $C_{214Bi}$  are the activity concentration of <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi separately (Bq·m<sup>-3</sup>).  $\varepsilon_{\alpha}$  is the detection efficiency for alpha particles, *F* is the sampling flowrate (l·min<sup>-1</sup>),  $\varepsilon_{\rm f}$  is the collection efficiency, SL is the loss factor of wire screen and FT is the front to total activity ratio of wire screen. According to the semi-empirical formula from Solomon and Ren<sup>(37)</sup>, SL is 1.04 and FT is 0.84 for our wire screen. The lower-level detection limit of equilibrium equivalent concentration of total radon progeny is nearly 0.1 Bq·m<sup>-3</sup> for this device.

The RPM-FF01-2S unattached radon progeny measurement device is tested in the radon chamber of National Institute of Metrology, China, and comparisons in different environments were carried out in detail with two commercial unattached radon progeny measurement instruments<sup>(38)</sup>.

#### **Field measurements**

For getting the characteristic parameters of radon progeny in real environments, a series of field measurements was carried out during August and September, 2020.

Field survey was carried out in the Lingbao City (110°51'E and 34°32'N), Henan Province, which is located in the central area of China and has typical dwellings both in city area and in surrounding rural area. In city area, typical tall buildings were selected, which are built after the year 2000 and made of

reinforced concrete. In rural area, typical one-story or two-story houses built with red brick materials were surveyed, which are 5-20 km away from the city. All the field measurements are carried out in bedrooms or in living rooms with the open or closed position of doors and windows as found when the location was accessed for sampling, just as it is in real life. Measurement device was put on tables or on beds, 50– 100 cm from floors or from walls. During measurement, cooking and smoking are avoided to eliminate the influence of additional aerosol particles. Most measurements were carried out during the daytime and only a few were carried out after 20 o'clock. Onehour's measurement is carried out in one dwelling and the filter and wire screen are kept at 'clean' to avoid the influence of dust accumulation and radon progeny.

For comparison, field measurements were also carried out in Beijing for more than 20 days, from 11 August to 1 September 2020. A typical office room which is in the first floor of a three-story building and outdoor environment were chosen, and semi-continuous measurements were carried out at the same time using two sets of RPM-FF01-2S monitor. The office room has a size of  $6 \text{ m} \times 3 \text{ m} \times 3 \text{ m}$  and is kept at normal use without smoking and with air-condition off. In outdoors, RPM-FF01-2S is put in a thermometer shelter, which is located in the garden just outside the office building, and the monitor is kept at nearly 1 m above the ground. During field measurement, the temperature changes between 19 and 36°C. and the air humidity changes in the range of 54-95%RH. The measurement cycle of the RPM-FF01-2S monitor is set to 4 h, but sampling and detecting only at the first hour of each cycle and waiting 3 h for the decay of short-lived radon progeny. The filter and the screen are replaced every 1-2 days to avoid dust accumulation.

#### **RESULTS AND DISCUSSIONS**

#### Results of the survey in the city and rural dwellings

During field survey, 1-h measurements were carried out in 31 dwellings in the city and 42 dwellings in the rural area, respectively. However, due to the low concentrations of radon progeny in real dwellings, the activity concentrations of unattached radon progeny in some environments are lower than the lower-level detection limits of RPM-FF01-2S, only 19 effective results with EEC larger than 0.1 Bq·m<sup>-3</sup> in the city dwellings and 25 effective results in the rural dwellings were gotten. Field survey results in the city and rural areas are shown in Tables 1 and 2, where the equilibrium equivalent concentration of total radon progeny (EEC), the EEC of unattached radon progeny (EEC<sup>U</sup>), the unattached fraction of potential alpha energy concentration  $(f_p)$ , the activity concentrations of <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi ( $C_{218p_0}$ ,  $C_{214p_b}$  and  $C_{214_{Bi}}$ , respectively) and the activity concentrations of the unattached <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi ( $C_{218p_0}^{u}$ ,  $C_{214p_b}^{u}$ , and  $C_{214_{Bi}}^{u}$ , respectively) are listed in details.

This survey focuses on the unattached fraction of radon progeny ( $f_p$ ), so the EEC<sup>U</sup> less than the lower-level detection limit is discarded, and its corresponding EEC is also discarded. The average was calculated only from these results. If the real value of EEC<sup>U</sup>, which is below 0.1 Bq·m<sup>-3</sup> would be taken into account, the average EEC<sup>U</sup> would be significantly smaller.

Results show that most unattached radon progeny concentration is quite low. In typical city dwellings, the arithmetic mean of  $EEC^U$  is 0.4 Bq·m<sup>-3</sup> and the standard deviation is 0.2  $Bq{\cdot}m^{-3}.$  The geometric mean of  $EEC^U$  is 0.3  $Bq{\cdot}m^{-3}$  and the geometric standard deviation is 1.8 Bq·m<sup>-3</sup>. The range of EEC<sup>U</sup> is 0.1–0.7 Bq $\cdot$ m<sup>-3</sup>. While in typical rural dwellings, the arithmetic mean of EEC<sup>U</sup> is  $0.5 \text{ Bg} \cdot \text{m}^{-3}$  and the standard deviation is  $0.3 \text{ Bg} \cdot \text{m}^{-3}$ . The geometric mean of  $EEC^U$  is 0.4 Bq m<sup>-3</sup> and the geometric standard deviation is 1.8  $Bq \cdot m^{-3}$ . The range of  $EEC^U$  is 0.1–1.8 Bq·m<sup>-3</sup>. The  $EEC^U$  in rural dwellings is without significant difference with those in city environment. Low concentration led to high uncertainty, and sometimes it was hard to get the accurate result of unattached radon progeny concentration. Although the revised Wicke method has much higher sensitivity than traditional Kerr method and Thomas method<sup>(39-41)</sup>, it is still not easy to accurately measure extremely low unattached radon progeny concentration due to the limitation of sampling flowrate, which is determined by the half cut-off diameter (dp50) and the diameter of wire screen.

In typical city dwellings, the arithmetic mean of EEC is 11.2 Bq·m<sup>-3</sup> and the standard deviation is 7.4 Bq·m<sup>-3</sup>. The geometric mean of EEC is 9.3 Bq·m<sup>-3</sup> and the geometric standard deviation is 1.8 Bq·m<sup>-3</sup>. The range of EEC is 3.1–35.2 Bq·m<sup>-3</sup>. While in typical rural dwellings, the arithmetic mean of EEC is 17.3 Bq·m<sup>-3</sup> and the standard deviation is 17.3 Bq·m<sup>-3</sup>. The geometric mean of EEC is 13.4 Bq·m<sup>-3</sup> and the geometric standard deviation is 1.9 Bq·m<sup>-3</sup>. The average  $f_p$  is 4.5% varies from 1.9 to 7.5% for city dwellings and 3.8% varies from 0.82 to 6.5% for rural dwellings.

The average activity concentration of <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi are 13.7  $\pm$  2.5 Bq·m<sup>-3</sup>, 12.6  $\pm$  3.1 Bq·m<sup>-3</sup> and 8.6  $\pm$  2.1 Bq·m<sup>-3</sup>, while that of unattached <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi are 2.3  $\pm$  0.7 Bq·m<sup>-3</sup>, 0.1  $\pm$  0.2 Bq·m<sup>-3</sup> and 0.1  $\pm$  0.1 Bq·m<sup>-3</sup>, respectively, for city dwellings. However, for rural dwellings, the average activity concentration of <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi are 21.3  $\pm$  2.9 Bq·m<sup>-3</sup>, 18.5  $\pm$  3.5 Bq·m<sup>-3</sup> and

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	$\begin{array}{c} C^{u}_{214\mathrm{Bi}} \\ (\mathrm{Bq}{\cdot}\mathrm{m}^{-3}) \end{array}$	$\begin{array}{c} 0.1\pm0.2\\ 0.1\pm0.2\\ 0.0\pm0.0\\ 0.6\pm0.3\\ 0.5\pm0.3\\ 0.0\pm0.0\\ 0.1\pm0.2\\ 0.1\pm0.2\\ 0.1\pm0.2\\ 0.0\pm0.0\\ 0.0\pm0.1\\ 0.0\pm0.1\\ 0.0\pm0.2\\ 0.0\pm0$	$0.2 \pm 0.2$
	$\begin{array}{c} \mathcal{C}^{u}_{2l4Pb} \\ (Bq{\cdot}m^{-3}) \end{array}$	$\begin{array}{c} 0.2\pm0.3\\ 0.2\pm0.3\\ 0.2\pm0.2\\ 0.1\pm0.4\\ 0.1\pm0.4\\ 0.1\pm0.4\\ 0.1\pm0.4\\ 0.0\pm0.0\\ 0.0\pm0.2\\ 0.1\pm0.4\\ 0.1\pm0.4\\ 0.1\pm0.4\\ 0.1\pm0.4\\ 0.1\pm0.4\\ 0.2\pm0.3\\ 0.2\pm0.3\\ 0.2\pm0.3\\ 0.2\pm0.3\\ 0.2\pm0.3\\ 0.2\pm0.3\\ 0.1\pm0.4\\ 0.0\\ 0.2\pm0.3\\ 0.1\pm0.4\\ 0.0\\ 0.1\pm0.3\\ 0.1\pm$	$0.3 \pm 0.4$
2	$\begin{array}{c} C^{u}_{218Po} \\ (Bq\cdot m^{-3}) \end{array}$	$\begin{array}{c} 1.4\pm 0.5\\ 1.4\pm 0.5\\ 2.4\pm 0.6\\ 1.7\pm 0.5\\ 1.7\pm 0.6\\ 1.7\pm 0.6\\ 1.9\pm 0.6\\ 1.9\pm 0.6\\ 1.9\pm 0.6\\ 1.3\pm 0.3\\ 3.3\pm 0.7\\ 1.3\pm 0.5\\ 1.3\pm 0.5\\ 1.5\pm 0.5\\$	+
	$\begin{array}{c} C_{214{\rm Bi}} \\ ({\rm Bq}{\cdot}{\rm m}^{-3}) \end{array}$	$5.6 \pm 1.3$ $5.6 \pm 2.0$ $5.6 \pm 2.0$ $5.6 \pm 2.0$ $23.7 \pm 5.4$ $4.0 \pm 1.1$ $4.0 \pm 1.2$ $7.8 \pm 1.9$ $11.4 \pm 2.7$ $11.4 \pm 2.7$ $12.8 \pm 2.8$ $8.6 \pm 1.6$ $10.3 \pm 1.8$ $5.0 \pm 1.1$ $7.4 \pm 2.3$ $3.7 \pm 1.1$ $10.1 \pm 1.8$ $5.0 \pm 1.1$ $10.3 \pm 1.8$ $10.5 \pm 2.3$ $3.3 \pm 1.1$ $10.5 \pm 2.8$ $10.5 \pm 2.8$	$14.3 \pm 2.1$
	$\begin{array}{c} C_{214\mathrm{Pb}}\\ \mathrm{(Bq\cdot m^{-3})} \end{array}$	$\begin{array}{c} 7.9\pm1.9\\ 7.3\pm3.5\\ 0.6\pm2.6\\ 8.5.5\pm1.6\\ 7.4\pm1.6\\ 7.4\pm1.6\\ 1.8\pm2.5\\ 1.8\pm2.5\\ 1.4.2\pm4.1\\ 1.7.1\pm4.2\\ 1.3.2\pm2.5\\ 1.5.2\pm2.5\\ 1.5.3\pm2.5\\ 1.5.3\pm2.7\\ 1.5.3\pm2$	$21.3 \pm 3.2$
	$C_{218Po}$ (Bq·m <sup>-3</sup> )	$5.5 \pm 1.2$ $5.0 \pm 2.3$ $6.1 \pm 2.5$ $6.1 \pm 2.5$ $3.5.3 \pm 6.1$ $6.7 \pm 2.1$ $10.0 \pm 1.7$ $6.7 \pm 2.1$ $17.7 \pm 3.4$ $11.7 \pm 2.7$ $17.9 \pm 2.3$ $6.0 \pm 1.3$ $6.0 \pm 1.3$ $17.9 \pm 2.3$ $17.9 \pm 2.2$ $17.9 \pm 2.2$ $17.9 \pm 2.2$ $19.0 \pm 3.5$ $19.0 \pm 3.5$ $22.5 \pm 2.6$ $19.0 \pm 3.5$ $22.5 \pm 2.6$	$28.8 \pm 3.0$
	f p	$\begin{array}{c} 4.8\%\pm2.4\%\\ 2.4\%\pm1.7\%\\ 7.2\%\pm4.1.7\%\\ 1.9\%\pm1.7\%\\ 4.0\%\pm2.9\%\\ 4.5\%\pm2.3\%\\ 3.2\%\pm2.3\%\\ 6.2\%\pm1.9\%\\ 6.2\%\pm1.9\%\\ 6.2\%\pm1.9\%\\ 5.1\%\pm2.0\%\\ 5.1\%\pm2.0\%\\ 5.1\%\pm2.0\%\\ 2.9\%\pm1.2\%\\ 2.7\%\pm1.2\%\\ 2.7\%\pm1.2\%$	$3.1\% \pm 1.2\%$
	EEC (Bq·m <sup>-3</sup> )	$\begin{array}{c} 6.8 \pm 1.1 \\ 6.8 \pm 1.1 \\ 6.2 \pm 2.0 \\ 3.1 \pm 1.6 \\ 3.5.2 \pm 4.9 \\ 5.0 \pm 1.0 \\ 6.4 \pm 1.0 \\ 6.4 \pm 1.0 \\ 6.4 \pm 1.0 \\ 13.5 \pm 2.4 \\ 14.5 \pm 1.6 \\ 14.5 \pm 1.6 \\ 14.5 \pm 1.6 \\ 14.5 \pm 1.6 \\ 11.5 \pm 1.0 \\ 10.5 \pm 2.0 \\ 5.5 \pm 1.0 \\ 10.5 \pm 2.0 \\ 10.5 \pm 2.0 \\ 10.5 \pm 2.0 \\ 15.1 \pm 1.6 \\ 15.1 \pm 1.6 \end{array}$	$19.5 \pm 1.9$
	$EEC^{U}$ (Bq·m <sup>-3</sup> )	$\begin{array}{c} 0.3 \pm 0.2 \\ 0.1 \pm 0.1 \\ 0.2 \pm 0.1 \\ 0.2 \pm 0.1 \\ 0.2 \pm 0.1 \\ 0.1 \pm 0.1 \\ 0.1 \pm 0.1 \\ 0.1 \pm 0.1 \\ 0.2 \pm 0.2 \\ 0.3 \pm 0.2 \\$	$+\!\!+\!\!$
	City dwellings	1 2 c 7 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2	19

Table 1. Field measurement results of radon progeny in typical city dwellings.

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$\begin{array}{c} \mathcal{C}^{u}_{2l4Bi} \\ (Bq \cdot m^{-3}) \end{array}$	$\begin{array}{c} 0.3\pm0.2\\ 0.3\pm0.3\\ 0.0\pm0.2\\ 0.0\pm0.2\\ 0.1\pm0.2\\ 0.1\pm0.2\\ 0.1\pm0.2\\ 0.1\pm0.2\\ 0.0\pm0.2\\ 0.0\pm0.2\\ 0.1\pm0.2\\ 0.1\pm0.2\\ 0.1\pm0.2\\ 0.0\pm0.0\\ 0.0\pm0.0\\ 0.0\pm0.0\\ 0.1\pm0.2\\ 0.2\pm0.2\\ 0.2\pm0$
$C^{\mathrm{u}}_{\mathrm{2l4Pb}}$ (Bq·m <sup>-3</sup> )	$\begin{array}{c} 0.0 \pm 0.0 \\ 0.3 \pm 0.4 \\ 0.2 \pm 0.3 \\ 0.1 \pm 0.2 \\ 0.2 \pm 0.2 \\$
$C^{\mathrm{u}}_{2\mathrm{l8Po}}$ (Bq·m <sup>-3</sup> )	$\begin{array}{c} 4.4\pm1.0\\ 2.5\pm0.7\\ 1.9\pm0.6\\ 3.4\pm0.8\\ 1.1\pm0.5\\ 1.1\pm0.5\\ 1.1\pm0.5\\ 1.1\pm0.5\\ 1.1\pm0.5\\ 3.1\pm0.6\\ 1.1\pm0.5\\ 3.1\pm0.6\\ 1.1\pm0.5\\ 3.1\pm0.8\\ 3.2\pm0.8\\ 3.2\pm0.8\\ 3.2\pm0.8\\ 1.1\pm0.5\\ 1.3\pm0.5\\ 1.3\pm0$
$C_{214\mathrm{Bi}}$ (Bq·m <sup>-3</sup> )	$22.1 \pm 2.5$ $20.7 \pm 2.4$ $15.4 \pm 3.0$ $10.6 \pm 2.7$ $6.1 \pm 1.6$ $6.1 \pm 1.6$ $6.6 \pm 1.2$ $5.2 \pm 1.8$ $10.9 \pm 2.9$ $8.0.9 \pm 1.6$ $7.2 \pm 1.5$ $2.7 \pm 1.7$ $11.1 \pm 2.7$ $11.1 \pm 2.7$ $2.7 \pm 1.3$ $12.1 \pm 2.8$ $2.7 \pm 1.3$ $12.1 \pm 2.8$ $2.3 \pm 1.9$ $15.4 \pm 3.2$ $5.3 \pm 1.9$ $15.4 \pm 2.3$ $15.4 \pm 3.2$ $5.3 \pm 1.9$ $15.4 \pm 3.2$ $15.4 \pm 3.2$ 15.4
$C_{214\mathrm{Pb}}$ (Bq·m <sup>-3</sup> )	$20.6 \pm 3.5$ $18.4 \pm 4.2$ $17.9 \pm 4.2$ $17.9 \pm 2.2$ $10.9 \pm 2.2$ $6.5 \pm 2.5$ $5.1 \pm 1.6$ $7.3 \pm 2.7$ $2.1 \pm 1.6$ $7.3 \pm 2.7$ $2.1 \pm 1.6$ $7.3 \pm 2.7$ $2.1 \pm 1.6$ $7.3 \pm 2.9$ $11.5 \pm 2.4$ $11.5 \pm 2.6$ $12.1 \pm 2.4$ $11.5 \pm 2.6$ $12.3 \pm 3.9$ $25.9 \pm 3.3$ $6.5 \pm 8.1$ $17.5 \pm 4.9$ $17.5 \pm 4.9$ $17.7 \pm 2.8$ $5.8 \pm 1.7$
$C_{218Po}$ (Bq·m <sup>-3</sup> )	$30.3 \pm 3.1$ $24.9 \pm 2.8$ $19.1 \pm 3.5$ $20.3 \pm 3.5$ $8.9 \pm 1.6$ $6.7 \pm 2.1$ $7.3 \pm 1.4$ $4.6 \pm 1.7$ $12.2 \pm 2.8$ $13.2 \pm 1.9$ $14.0 \pm 2.0$ $13.2 \pm 1.9$ $13.2 \pm 1.9$ $17.9 \pm 3.3$ $19.1 \pm 3.5$ $17.9 \pm 3.3$ $17.9 \pm 3.3$ $10.3 \pm 1.7$ $13.5 \pm 2.9$ $10.3 \pm 1.7$ $13.5 \pm 2.9$ $10.1 \pm 3.5$ $10.1 \pm 3.5$ $10.2 \pm 2.6$ $10.3 \pm 1.7$ $12.0 \pm 3.3$ $28.0 \pm 3.3$ $28.0 \pm 3.3$ $28.0 \pm 3.3$ $7.7 \pm 1.4$
$f_{\mathrm{p}}$	$\begin{array}{c} 2.2\%\pm0.9\%\\ 4.3\%\pm1.4\%\\ 5.8\%\pm1.4\%\\ 5.8\%\pm1.9\%\\ 2.9\%\pm1.3\%\\ 4.0\%\pm2.0\%\\ 4.1\%\pm3.0\%\\ 6.5\%\pm3.8\%\\ 5.5\%\pm3.8\%\\ 5.5\%\pm1.6\%\\ 3.7\%\pm1.8\%\\ 3.7\%\pm1.8\%\\ 3.7\%\pm1.4\%\\ 2.9\%\pm1.4\%\\ 2.6\%\pm1.1\%\\ 2.6\%\pm1.2\%\\ 5.5\%\pm1.2\%\\ 0.5\%\\ 6.0\%\pm3.1\%\\ 6.0\%\pm3.1\%\\ 6.0\%\pm3.1\%\\ 0.8\%$
EEC (Bq·m <sup>-3</sup> )	$\begin{array}{c} 22.2\pm2.1\\ 20.0\pm2.0\\ 14.1\pm2.5\\ 15.4\pm2.4\\ 15.4\pm2.4\\ 8.9\pm1.2\\ 5.4\pm1.5\\ 5.4\pm1.5\\ 5.4\pm1.6\\ 15.9\pm2.5\\ 10.6\pm1.4\\ 10.1\pm1.4\\ 10.1\pm1.4\\ 10.1\pm1.4\\ 10.1\pm1.4\\ 10.1\pm1.4\\ 10.1\pm1.3\\ 25.6\pm3.2\\ 19.5\pm2.3\\ 25.6\pm3.2\\ 10.5\pm1.9\\ 10.1\pm2.0\\ 34.4\pm2.8\\ 7.7\pm1.7\\ 7.7\pm1.7$
EEC <sup>U</sup> (Bq·m <sup>-3</sup> )	$\begin{array}{c} 0.4\pm0.\\ 0.7\pm0.3\\ 0.7\pm0.3\\ 0.5\pm0.2\\ 0.5\pm0.1\\ 0.5\pm0.2\\ 0.5\pm0.$
Rural dwellings	2 5 3 3 5 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0

Table 2. Field measurement results of radon progeny in typical rural dwellings.

FIELD MEASUREMENT OF THE UNATTACHED FRACTION

14.7  $\pm$  2.4 Bq·m<sup>-3</sup> and that of unattached <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi are 2.7  $\pm$  0.7 Bq·m<sup>-3</sup>, 0.2  $\pm$  0.3 Bq·m<sup>-3</sup> and 0.2  $\pm$  0.2 Bq·m<sup>-3</sup>, respectively.

It should be noticed that some values of  $C_{214_{\rm Pb}}^{\rm u}$  and  $C_{214_{\rm Bi}}^{\rm u}$  are less than 0. It is reasonable for field measurement, due to the low unattached radon progeny concentration and high measurement uncertainty, as well as the analytical solution of revised Wicke method. These negative values were assumed to be zero for calculation.

# Results of the survey in the office room and outdoors environment

Semi-continuous measurements of the unattached radon progeny concentration in office room and outdoors were carried out in Beijing, and those measurement results are shown in Figure 1.

The unattached fraction  $(f_p)$ , the radon progeny concentration (EEC) and the unattached radon progeny concentration (EEC<sup>U</sup>) of office room and outdoors are demonstrated simultaneously, and the rainfall data from the national meteorological station nearby are also given. Due to the large difference of indoor and outdoor radon progeny concentration, activity concentrations are shown with different coordinate axis.

Results show that the activity concentration of radon progeny and unattached radon progeny in office room are much higher than those in outdoors, but there is no significant difference between the unattached faction of radon progeny in office room and in outdoors.

In office room, the average EEC is  $73.9 \pm 5.9$  Bq·m<sup>-3</sup> with a range of 36.3-151.0 Bq·m<sup>-3</sup>, while the average EECU is 2.8  $\pm$  0.5 Bq·m<sup>-3</sup>with a range of 1.2–5.7 Bq $\cdot$ m<sup>-3</sup>. In outdoors, the average EEC is  $5.7 \pm 0.8$  Bq·m<sup>-3</sup> with a range of 1.3–14.2 Bq·m<sup>-3</sup>. The average EEC<sup>U</sup> is  $0.2 \pm 0.2$  Bq·m<sup>-3</sup> with a range of 0–0.5 Bq·m<sup>-3</sup>. The average  $f_{\rm p}$  is 5.11%  $\pm$  0.86% and varies in the range of 2.1-14.2% in office room. The average  $f_p$  in outdoors is 5.4%  $\pm$  3.1%, nearly in the same level of that in office room, but varying in a rather large range, 0.3-26.7%, compared with these in office room. Lower concentration, higher uncertainty as well as larger aerosol concentration range might be the main reason. It is also found that the  $f_p$  in office room seems to be changed in the same pattern with that in outdoors, which is an interesting founding, and high ventilation may be the reason.

Focus on the daily series variation of EEC and EEC<sup>U</sup> in office room with those in outdoors, a diurnal variation is found for EEC in both office room and in outdoors. Focus on the diurnal variation of EEC in office room, most of the maximum values appear at

12:00–0:00, and the minimum values appear at 0:00– 12:00, the average ratio of maximum to minimum is 1.79. The diurnal variation of EEC in office room is big and affected by human activities. Focus on the diurnal variation of EEC in outdoor, the regularity is obvious. The maximum values appear in 4:00-8:00, and the minimum values appear in 20:00-0:00; the average ratio of maximum to minimum is 2.51. There is no obvious diurnal variation of EEC<sup>U</sup> in outdoor. Furthermore, no diurnal variation of  $f_{\rm p}$  is observed either. For the  $f_p$  in outdoor environment, it increases after precipitation and the highest value of  $f_p$  appears in rainy day. The washout and the high humidity might be the main reason. During rainfall, the attached radon progeny is washed out on the ground, and the new-generated unattached radon progeny from radon gas might keep stable or grow up due to high humidity, which leads to an increase of the unattached fraction.

#### Summary results of different environments

For getting the typical characteristic parameters of radon progeny, especially the unattached fraction and the activity concentration ratio of radon progeny, field survey results and semi-continuous measurement results are summarized in Table 3, in which the unattached fraction of potential alpha energy concentration  $(f_p)$ , the unattached fraction of <sup>218</sup>Po  $(f_1)$  and the activity concentration ratio of radon progeny as well as that of unattached radon progeny are listed up in the table. In the past, most works were focused on EEC in China, only quite limited results of  $f_p$  as well as activity concentration ratio is given in former research. And also the activity concentration ratio of unattached radon progeny is lacking. For comparison, some similar survey results from former research are also listed<sup>(26, 27, 30)</sup>.

Results shown that the  $f_p$ ,  $f_1$  and activity concentration ratios in different environments are quite different. Average value of  $f_p$  and  $f_1$  in city dwellings is higher than these in rural dwellings, and both smaller than these in office room and in outdoors. The typical activity concentration ratio of radon progeny and unattached radon progeny in city dwellings are 1:0.94:0.70 and 1:0.07:0.06, respectively, while these in rural dwellings are 1:0.88:0.66 and 1:0.09:0.07, which is a little different with those in indoor workplace recommended by ICRP 137(8). And the average value of  $f_p$  and  $f_1$  in outdoor environment is 5.4 and 23.8%, with typical activity concentration ratio 1:0.88:0.77 for radon progeny and 1:0.11:0.11 for unattached radon progeny. The average activity concentration ratio in office room is 1:1.18:1.09, which seems unexpected, but it is real for some indoor environment. The similar phenomenon appears in EL-Hussein's and Mostafa et al.'s papers<sup>(27, 31)</sup>. The reason is that unattached fraction of <sup>218</sup>Po is much

Environment	Samples no.	$f_{ m P}$	fı	$C_{218} \frac{P_0}{P_{214}}$ : $C_{214} \frac{P_b}{P_i}$ :	$C^{\mathrm{u}}_{218\mathrm{Po}}$ ; $C^{\mathrm{u}}_{214\mathrm{Pb}}$ ; $C^{\mathrm{u}}_{214\mathrm{Pb}}$ ; $C^{\mathrm{u}}_{214\mathrm{Pb}}$	Remark
City dwelling	19	$4.5\%\pm2.2\%(1.9{-}7.5\%)$	$26.0\% \pm 12.9\% (7.2-46.6\%)$	1:0.94 (0.10-1.85):0.70	1:0.07(0-0.31):0.06	
Rural dwelling	25	$3.8\%\pm1.7\%(0.8{-}6.5\%)$	$17.5\% \pm 6.8\% (5.8 - 35.1\%)$	(c1.1-26.0) 1:0.88 $(0.29-1.67)$ :0.66	(220-0) 1:0.09 (0-0.37):0.07 (0, 0, 25)	
Office room		$5.1\% \pm 0.9\% (2.1 - 14.2\%)$	$28.5\% \pm 3.9\%$ (7.1–81.8%)	(2c.1-2c.0) 1:1.18 $(0.76-2.48)$ :1.09	1:0.15(0-0.37):0.08	124 measurement
Outdoor		$5.4\% \pm 3.1\% \ (0.3-26.7\%)$	$23.8\% \pm 9.0\% \ (0-77.5\%)$	(0.70-2.04) 1:0.88 $(0-2.8)$ :0.77	(220-0) 1:0.11 (0-2.23):0.11 (0, 1, 20)	results
Indoor workplace	I	8%		(0.22-0.04) 1:0.56:0.43 <sup>a</sup>	1:0.1:0	ICRP 137
Closed rooms without additional	10	9.60% (1.60–24.60%)	33.60%	1:0.55:0.47	1:0.09:0.04	1990/Reineking <sup>b</sup>
aerosol particles Closed room without additional	10	6.00% (2.00-8.00%)	14.00% (7.00-20.00%)	1:1.02:0.66	1:0.49:-1.01	1996/A.EL- Hussein <sup>b</sup>
aerosol particles Partly open the window	б	4.27% (2.00–8.10%)	24.33% (15.00–30.00%)	1:0.54:0.50	1:0.15:0.07	1996/A.EL- Hussein <sup>b</sup>
Closed room	6	9.64% (1.30-24.00%)	14.79% (11.76–37.25%)	1:0.72:0.54	1:0.44:-0.08	2012/M.Mohery <sup>b</sup>
Partly open the window	6	5.91% (2.90–12.40%)	15.44% (11.50–36.36%)	1:0.51:0.44	1:0.15:-0.09	2012/M.Mohery <sup>b</sup>
${}^{a}C_{218Po} : C_{214Pb} : C_{21}$ the ICRP 137 report.	$^{4}B_{i} = 1:0.56:0$	0.43 is derived from the referen	${}^{a}C_{218p_0}$ : $C_{214p_b}$ : $C_{214p_b}$ : $C_{214p_b}$ : $C_{214p_b}$ : $C_{14p_b}$ : $C_{214p_b}$ :	$^{[4]}_{Bi} = 1:0.75:0.6, C_{218Po}^{u}: C$	$C_{214Pb}^{u}$ : $C_{214Bi}^{u} = 1:0.1:0 a$	$\inf f_{\rm p} = 8\%$ given in

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Table 3.

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the ICKP 13/ report. <sup>b</sup>The unattached fraction of <sup>218</sup>Po,  $C_{218}P_0$ :  $C_{214}P_b$ :  $C_{214}P_b$ :  $C_{214}P_b$ :  $C_{214}^{u}P_b$ ;  $C_{214}^{u}P_{B_1}$  are recalculated from results table given in papers.

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### FIELD MEASUREMENT OF THE UNATTACHED FRACTION

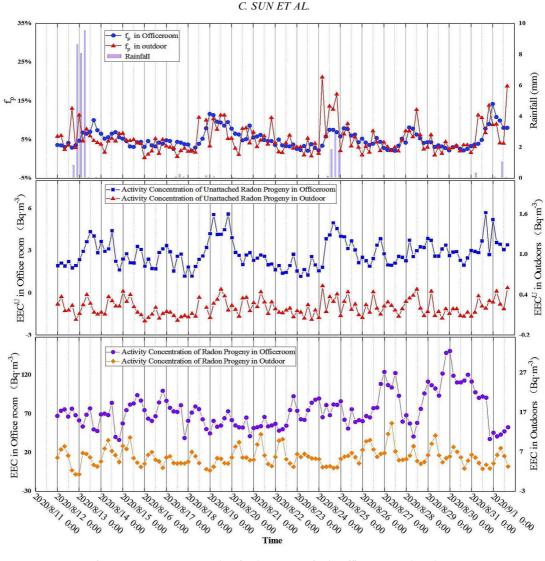


Figure 1. Measurement results of radon progeny in the office room and outdoors.

higher than those of unattached <sup>214</sup>Pb and <sup>214</sup>Bi, and the unattached radon progeny has higher deposition rate than attached radon progeny. Therefore, <sup>218</sup>Po deposited on the room surface will return to the air with <sup>214</sup>Pb due to recoil, which lead to  $C_{214Pb}/C_{218Po}$ and  $C_{214Bi}/C_{218Po}$  higher than 1. And also, this is semicontinuous result in just one office room, which gives us the cue that field survey of different rooms is more important than continuous measurement in one or few rooms for typical characteristic parameters and dose evaluation of radon progeny.

Comparing with former research, the  $f_p$  and  $f_1$  are comparable, but the activity concentration ratios seem a little different. The average values of  $C_{214}$ <sub>Pb</sub>/ $C_{218}$ <sub>Po</sub> and  $C_{214}$ <sub>Bi</sub>/ $C_{218}$ <sub>Po</sub> are higher than

most of those results in Reineking's, Hussein's and Mohery's papers. The average value of  $C_{214}^{u}_{Pb}/C_{218}^{u}_{Po}$ and  $C_{214}^{u}_{Bi}/C_{218}^{u}_{Po}$  seems more physically normal, and  $C_{214}^{u}_{Bi}/C_{218}^{u}_{Po}$  is not zero compared with ICRP 137. Different ventilation rate and different aerosol condition might be the main reason. It is worth noting that Reineking's results were performed in 10 rooms of different houses, Hussein's and Mohery's results were both performed in three rooms and several measurements were carried out in one room at different time. Limited results of different rooms lead to poor statistics. And also, due to the limitation of flowrate and dp50, the measurement sensitivity of the unattached radon progeny measurement device is mainly decided by the measurement methodology and detection efficiency, which is hardly improved and should be paid attention.

#### CONCLUSION

For purpose of getting the typical characteristic parameters of radon progeny, especially the unattached fraction and activity concentration ratio of radon progeny in dwelling, a series of field survey was carried out in typical city and rural dwellings, and semi-continuous measurements were carried out in office room and outdoors for more than 20 days. Nineteen effective results in typical city dwellings and 25 effective results in typical rural dwellings are gotten.

Results show that the  $f_p$ ,  $f_1$  and activity concentration ratios in different environments are quite different. Average value of  $f_p$  is  $4.5\% \pm 2.2\%$  in city dwellings and  $3.8\% \pm 1.7\%$  in rural dwellings, respectively, and in outdoors it is  $5.4\% \pm 3.1\%$  with a large range of 0.3-26.7%. The typical activity concentration ratio of total and unattached radon progeny is 1:0.94:0.70 and 1:0.07:0.06 for city dwellings, while in rural dwellings these are 1:0.88:0.66 and 1:0.09:0.07, and in outdoors these are 1:0.88:0.77 and 1:0.11:0.11, respectively. Those results are a little different with those in indoor workplace recommended by ICRP 137. Those field measurement results will enrich the characteristic parameters data of radon progeny in different environment, which is quite important for dose evaluation of radon exposure in typical environment.

Due to the low concentration of unattached radon progeny, the accurate measurement of each unattached radon daughters' concentration is quite hard for field survey. So more field measurement in different dwellings is important for obtaining the representative characteristic parameters. And also, measurement methods with higher metrological sensitivity are still required in the future.

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