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Field measurement of the ^{218}Po, ^{214}Pb and ^{214}Bi concentrations in typical indoor and outdoor environments in Beijing

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Abstract The activity concentrations of ^{218}Po, ^{214}Pb and ^{214}Bi [i.e. C(^{218}Po), C(^{214}Pb), and C(^{214}Bi)] and the calculated concentration ratios [i.e. 1:C(^{214}Pb)/C(^{218}Po):C(^{214}Bi)/C(^{218}Po)] are necessary for assessing radon and its progenies exposure. In this study, a measurement method of radon progenies concentrations with both high sensitivity and low uncertainty, was developed based on the Kerr method. The field measurement results of radon progeny concentrations and calculated concentration ratios in both typical indoor and outdoor environments in Beijing, China, were reported. The effects of air exchange rate on concentration ratios of radon progenies in indoor environments were discussed.

Keywords Radon progeny · Concentration ratio · EEC · Field measurement · Exposure assessment · Air exchange rate

Introduction

Radon (^{222}Rn) is a natural radioactive gas with a half-life of 3.8 days. Radon decays into a series of short-lived progenies (i.e. ^{218}Po, ^{214}Pb, and ^{214}Bi), which could be inhaled, deposit into lung tissues, and occur inner exposure. It is reported that radon and its progenies contribute more than half of the total natural radiation exposure to human beings [1].

As we know, the activity concentrations (Bq/m^3) of ^{218}Po, ^{214}Pb and ^{214}Bi (i.e. C(^{218}Po), C(^{214}Pb), and C(^{214}Bi)) are necessary for assessing radon and its progenies exposure when calculating the dose conversion factor (DCF), through the lung domestic model [2]. However, in the past decades, due to the limitation of measurement instruments, it was not easy to accurately derive the concentrations of each short-lived radon progeny, as compared with feasibility of radon measurement. Therefore, the radon equilibrium factor and the concentration ratios of radon progenies [i.e. 1:C(^{214}Pb)/C(^{218}Po):C(^{214}Bi)/C(^{218}Po)] were used to describe the disequilibrium relationships between radon and its progenies, which was further used to assess the exposure of radon progenies.

The radon equilibrium factor and the concentration ratios of radon progenies were absolutely different in various environments [3]. Many field measurements were carried out for the radon equilibrium factors, the reference value of which was 0.4, recommended by the United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR) [4]. However, limited results of the concentration ratios of radon progenies were reported, especially after 1980s. In 1967, Haque and Collinson [5] analyzed the concentration ratios of radon progenies under four different ventilation conditions by using a simplified relationship between radon and its progenies. In 1972, Toth et al. [6]. measured the concentration ratios in an enclosed indoor environment from extensive indoor radon progeny surveys. In 1972, Yeates [7] reported 26 measurement results at 14 selected sites from a large-scale survey of indoor environments in the United States. Due to the limited number of results, the reference value of the concentration ratio of radon progenies was not given by

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UNSCEAR yet. Therefore, when assessing the radon exposure through the lung domestic model, studies usually deployed inconsistent and non-referenced radon progeny concentration ratios. Ishikawa et al. [8] used an activity ratio of 0.84:0.45:0.2 for attached radon progeny, and 1:0.1 for the unattached fraction. Wasiolek et al. [9] used an concentration ratio as 1:0.65:0.4 in their work.

So in this study, the radon equivalent equilibrium concentration (EEC) and concentration ratio of ^{218}Po, ^{214}Pb and ^{214}Bi were reported via a series of field measurements in both typical indoor and outdoor environments in Beijing, China from November 2013 to March 2014. Moreover, the radon progeny concentration ratios in both typical indoor and outdoor environments were calculated and reported. The reported concentration ratios of radon progenies via field measurements can give the important clue to radon progeny exposure assessment. Meanwhile, this study also reported the concentration levels of radon EECs and each short-lived radon progeny in both typical indoor and outdoor environments in Beijing.

Experimental

Measurement of radon progenies concentrations

To measure the concentrations of ^{218}Po, ^{214}Pb and ^{214}Bi, Kerr method [10] was used in this study due to its high sensitivity and low uncertainty, especially for ^{218}Po measurement [10, 11]. The whole process can be divided as two parts: (1) sample collection, during which the airborne radon progenies were collected on a filter; and (2) measurement, during which the alpha particles emitted from decayed radon progenies can be counted by an alpha spectrometer. There were three assumptions in the following calculations: (1) the concentrations of radon progenies were stable during the samplings; (2) the influence of environment factors (e.g. temperature, relative humidity, and airborne particle concentration) could be neglected; (3) the alpha self-absorption of filter can be neglected.

Firstly, when radon progenies were collected on the filter, the activity of a radon progeny can be described as:

$$\frac{dN_i(t)}{dt} = C_i f + \lambda_{i-1} N_{i-1}(t) - \lambda_i N_i(t), \tag{1}$$

where $i = 0, 1, 2,$ and 3 stands for ^{222}Rn, RaA (^{218}Po), RaB (^{214}Pb), and RaC (^{214}Bi), respectively, C_i (Bq/m^3) stands for the activity concentration of a radon progeny $i = 1, 2,$ and 3, $N_i(t)$ stands for the number of a radon or radon progeny (i)'s atoms, λ_i stands for the decay constant (s^{-1}) of a radon progeny i, f (L/min) stands for flow rate of the collection pump, t (s) stands for the sampling time. Moreover, the initial conditions of Eq. 1 is N_i $(t = 0) = 0$.

After collection, during the measurement, the activity of a radon progeny on the filter can be described as:

$$\frac{dN_i(T)}{dT} = \lambda_{i-1} N_{i-1}(T) - \lambda_i N_i(T) \tag{2}$$

T (s) stands for the measurement time. The initial conditions of Eq. 2 would be $N_i = 0(T = 0)$.

Then, solve the Eq. 1 and substitute the solutions it into the Eq. 2, we could build a general relationship between M_j and C_i, which is as follows:

$$M_j = a_{j1} C_1 + a_{j2} C_2 + a_{j3} C_3, \tag{3}$$

where M_j is total count during jth time interval. a_{ji} is a constant that is determined by parameters, i.e. λ_i (s^{-1}), T (s), f (L/min), and collection efficiency E. Finally, the concentrations of each radon progeny can be calculated by:

$$\begin{pmatrix} C_1 \\ C_2 \\ C_3 \end{pmatrix} = \begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix}^{-1} \begin{pmatrix} M_1 \\ M_2 \\ M_3 \end{pmatrix}. \tag{4}$$

Within the measurement, radon progenies were collected on a 25 mm diameter glass microfiber (Whatman GF/A, Germany) by a free-oil vacuum pressure pump (VCA5038B, QiHai, China) for 10 min. The collecting efficiency is $\sim 100\%$ with the uncertainty which was lower than 0.01%. The pump flow rate was controlled as 19.54 ± 0.04 L/min and monitored by a flow meter (TSI 4046, Shoreview, MN).

The alpha spectrums of the filters were measured in two time intervals, i.e. from the 2nd to 12th min, and from the 15th to 30th min, after sampling, by a 400 mm^2 Passivized Implanted Planar Silicon (PIPS) detector (BS400-01/03, SARAD, Germany) combined with a portable alpha spectrometer (Spectra 5011, SARAD, Germany). The PIPS detector was placed 2.5 mm higher over the filter. The detection efficiency E of this system was 30.1 ± 0.3%, which was calibrated by a standard ^{241}Am and ^{239}Pu source with the diameter of 20 mm. The counts in the first region of interest (ROI) *ROI1* (i.e. from 3.0 to 6.1 MeV) and the second *ROI2* (i.e. from 6.2 to 7.8 MeV) were recorded, respectively. Take M_1, M_2, and M_3 as the alpha counts in *ROI1* of the first time interval, the alpha counts in *ROI2* of the first time interval, and the alpha counts in *ROI2* of the second time interval, respectively.

The concentrations of ^{218}Po, ^{214}Pb and ^{214}Bi can be calculated using the following equations:

$$C_1 = \frac{1}{fE\eta K_\alpha} \times 0.283 \times 10^{-4} M_1 \tag{5}$$

$$C_2 = \frac{1}{fE\eta K_\alpha} \times (-0.035 \times M_1 - 0.066 \times M_2 + 0.075 \times M_3) \times 10^{-4} \tag{6}$$

$$C_3 = \frac{1}{fE\eta K_\alpha} \times (0.005 \times M_1 + 0.070 \times M_2 - 0.032 \times M_3) \times 10^{-4} \quad (7)$$

where C_i is the concentration of radon progeny (Bq/m^3) and $i = 1, 2, 3$ stands for ^{218}Po, ^{214}Pb and ^{214}Bi, respectively. The f stands for the sampling flow rate (L/min), E is the detection efficiency of the alpha spectrometer, and η is the membrane filtration efficiency ($\eta = 1$), K_α stands for self-absorption correction coefficient for alpha particles ($K_\alpha = 1$).

The standard deviation of each concentration was calculated by the Error Propagation Formula. Considering the uncertainty of the pump velocity ($\sigma_f = 0.04$), the uncertainty of the detection efficiency ($\sigma_E = 0.27\%$), and the uncertainty of measurement counts ($\sigma_{M_i} = \sqrt{M_i}$), the uncertainties of ^{218}Po, ^{214}Pb and ^{214}Bi concentrations could be expressed as follow:

$$\sigma_{C_1} = 0.288 M_1^{1/2} \quad (8)$$

$$\sigma_{C_2} = \left[1.26 \times 10^{-3} M_1 + 4.50 \times 10^{-3} M_2 + 5.86 \times 10^{-3} M_3\right]^{1/2} \quad (9)$$

$$\sigma_{C_3} = \left[3.12 \times 10^{-5} M_1 + 5.09 \times 10^{-3} M_2 + 1.06 \times 10^{-3} M_3\right]^{1/2} \quad (10)$$

The lower detection limit of this measurement system is 0.12 Bq/m^3 for EEC concentration at 95% confidence level.

Field measurements

A total of 30 field samplings in typical indoor environments (i.e. including both offices and dwellings) were conducted under various ventilation conditions in Beijing, China from November 2013 to March 2014. During the field measurement, the temperature, relative humidity and airborne particle number concentrations were monitored. The indoor temperature ranged from 15 to 23 °C, while the relative humidity (RH) was between 12 and 48%. The arithmetic mean of the airborne particle number concentration in indoor environments was $\sim 1.5 \times 10^4$ particles/cm^3.

A total of 50 field samplings in typical outdoor environments were conducted under various weather conditions between November 2013 and March 2014 in Beijing, China. During the sampling period, the outdoor temperature ranged from 2 to 23 °C and the RH ranged from 8 to 80%. The mean airborne particle number concentration in outdoor environments was $\sim 2.5 \times 10^4$ particles/cm^3.

Air exchange rate

The air exchange rate (AER) in this study was determined by the gas tracer method [12]. The background indoor and outdoor CO_2 concentrations was measured firstly by a CO_2 monitor (Tes-1370, TES Taiwan), respectively. After that, certain amount of pure CO_2 was released evenly from a 4 L tank of compressed CO_2 to the room by an experimenter to make the level of CO_2 concentration beyond 1500 ppm. The releasing time length were pre-calculated according to the CO_2 concentration in the tank, the room volume, and the indoor CO_2 background concentration. After releasing, a CO_2 monitor was set in the center of the room under a designated ventilation condition and measures the CO_2 concentration by second for about two hours. The experimenter left the room after setting up the CO_2 monitor and stayed outside during the measurements to avoid the breathing-out CO_2. The measured CO_2 concentrations, the indoor, and outdoor background CO_2 concentrations would be used to calculated the AER.

The AER can be calculated by the following equation:

$$\frac{dC_i(t)}{dt} = \lambda_v C_0(t) - \lambda_v C_i(t), \quad (11)$$

where $C_i(t)$ (ppm) is the indoor concentration of CO_2 at the time of t, $C_0(t)$ (ppm) is the concentration of outdoor CO_2, λ_v is the AER (h^{-1}). Then, the Eq. 11 can be further solved as Eqs. 12 and 13.

$$C_i(t) = (C_i(0) - C_0)\exp(-\lambda_v t) + C_0 \quad (12)$$

$$\lambda_v = -\frac{1}{t} \cdot \ln\left[\frac{C_i(t) - C_0}{C_i(0) - C_0}\right] \quad (13)$$

Finally, AER can be calculated by Eq. 13.

Results and discussion

The ^{218}Po, ^{214}Pb and ^{214}Bi concentrations in typical indoor environments

The frequency distributions of the EEC, ^{218}Po, ^{214}Pb and ^{214}Bi concentrations in a typical indoor environment in Beijing are shown in Fig. 1a–d, respectively. It is shown that, in the typical indoor environment, the EECs ranged from 1.2 to 21.2 Bq/m^3, with a mean value of 6.4 Bq/m^3. The ^{218}Po concentrations fluctuated from 1.5 to 31.5 Bq/m^3, with a mean value of 10.1 Bq/m^3. The ^{214}Pb concentrations varied between 0.7 and 18.5 Bq/m^3 (mean: 5.9 Bq/m^3). The ^{214}Bi concentration changed between 0.8 and 22.6 Bq/m^3, with a mean value of 6.0 Bq/m^3. The mean radon EEC concentration was lower than the results (11.50 ± 6.99 Bq/m^3) in Zhang et al.'s study [13] in Beijing. This might be due to different measurement conditions that Zhang et al.'s study reported the 24-h integrated results under a certain ventilation condition, which was half-open windows at the daytime and closed windows at the nighttime.

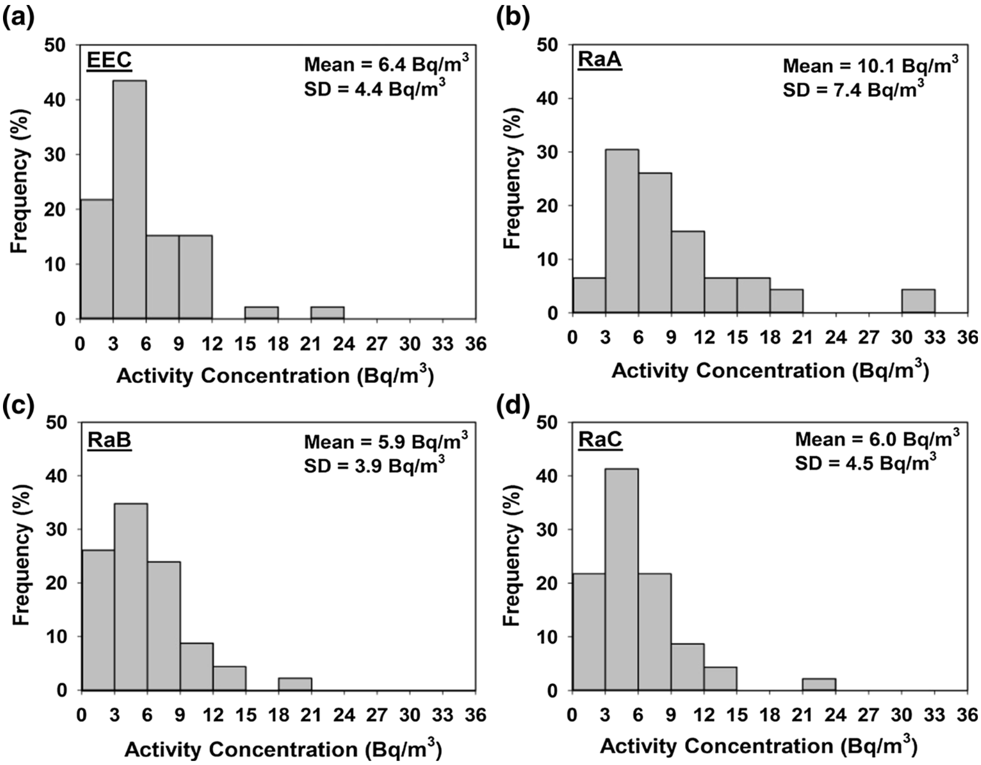

Fig. 1 The frequency distributions of the EEC, RaA, RaB, and RaC concentrations in indoor environments. (The arithmetic mean and standard deviation (SD) of each concentration were identified in the figures)

The ^{218}Po, ^{214}Pb and ^{214}Bi concentrations in typical outdoor environments

The frequency distributions of EEC, ^{218}Po, ^{214}Pb and ^{214}Bi concentrations in the typical outdoor environment in Beijing was shown in Fig. 2a–d, respectively. In the typical outdoor environment, the EEC varied between 1.6 and 12.0 Bq/m^3, with a mean value of 4.0 Bq/m^3. The ^{218}Po concentration fluctuated between 1.7 and 17.2 Bq/m^3, with a mean of 6.1 Bq/m^3. The ^{214}Pb concentration ranged from 0.6 to 13.4 Bq/m^3, with a mean value of 3.0 Bq/m^3. The ^{214}Bi concentration has a range of 1.1–13.1 Bq/m^3, with a mean of 4.5 Bq/m^3.

The concentration ratios of ^{218}Po, ^{214}Pb and ^{214}Bi

In this study, the concentration ratios of ^{218}Po, ^{214}Pb and ^{214}Bi in indoor and outdoor environments were expressed in 1:x:y format, where x and y was C(^{214}Pb)/C(^{218}Po) and C(^{214}Bi)/C(^{218}Po), respectively. The calculated results of radon progeny (i.e. ^{218}Po, ^{214}Pb and ^{214}Bi) concentration ratio in this study are shown in Table 1. Those results were also compared with former researches.

From this study, it is shown that the ratio of radon progeny concentration in indoor and outdoor environments in this study varied in quite a large scale, with the average value of 1:0.59:0.58 in indoor environments and 1:0.50:0.67 in outdoor environments. The indoor average result was lower than Toth's [6] and Yeates et al.'s [7] results, but higher than Haque's [5] and Ishikawa et al's [8] results. The outdoor average result was lower than Yeates et al.'s [7] results. This difference was mainly due to the different environment factors, such as the temperature, relative humidity, and the airborne particle conditions. The influence of various radon progeny concentration ratios on radon dose conversion factor (DCF) can be seen elsewhere [14].

In indoor environments, it is found that the concentrations of ^{218}Po, ^{214}Pb and ^{214}Bi were affected by AERs. So to study if the concentration ratios were also affected by the AERs, the concentrations of EEC, ^{218}Po, ^{214}Pb and ^{214}Bi under various ventilation conditions in a certain room were measured, as well as the AERs in that room [15]. Those results are shown in Table 2.

The results in Table 2 also showed that the ventilation rate had a great influence on the radon progeny concentrations and its ratios. The concentrations of EEC, ^{218}Po, ^{214}Pb and ^{214}Bi obviously increased when the AER decreased. It is because when the AER increased, the air flow could bring into more fresh air and remove the accumulated radon progenies, which would lead to the decrease of the concentrations of RaA, RaB, and RaC, and certainly EEC levels. Moreover, when the AER increased from 0.26 to 3.2/h, the ratio of RaB concentration to RaA definitely increased from 0.54 to 0.96. While the ratio of RaC to RaA increased from 0.43 to 0.83, except the

Fig. 2 The frequency distributions of EEC, RaA, RaB, and RaC concentrations in typical outdoor environments. (The arithmetic mean and standard deviation (SD) of each concentration were identified in the figures)

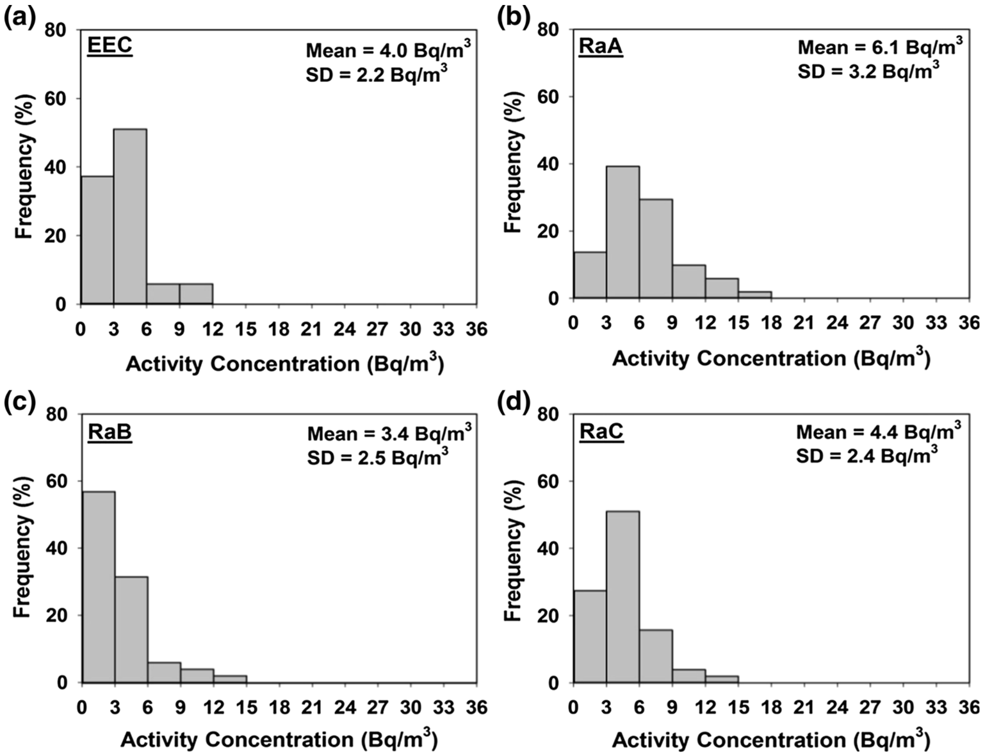

Table 1 The concentration ratios of ^{218}Po, ^{214}Pb and ^{214}Bi in the typical indoor and outdoor environments

Studies	Measurement methods	Environment type	Concentration ratios[a]
Haque and Collinson [5]	–	Dwellings	1:0.56:0.39
Toth [6]	Alpha counter	Dwellings	1:0.86:0.82
Yeates [7]	Gamma radiation measurement	Dwellings	1:0.97 (0.7–1.2):0.68 (0.3–1.1)
		Offices	1:0.88 (0.67–1.1):0.79 (0.43–1.3)
		Outdoor	1:0.83 (0.6–1):0.83 (0.5–1.3)
Ishikawa et al. [8]	–	Dwellings	1:0.54:0.24
Wasiolek et al. [9]	–	Dwellings	1:0.65:0.4
This study	Alpha spectrometer	Indoor	1:0.59 (0.21–1.42):0.58 (0.16–1.16)
		Outdoor	1:0.50 (0.17–1.24):0.67 (0.37–1.65)

[a] The average value and the range are shown separately in outside and inside of bracket

Table 2 The activity concentrations of EEC, ^{218}Po, ^{214}Pb and ^{214}Bi under various ventilation conditions in the same room

Ventilation conditions	AER (h^{-1})	^{218}Po (Bq/m^3)	^{214}Pb (Bq/m^3)	^{214}Bi (Bq/m^3)	EEC (Bq/m^3)	Concentration ratio
Door open, windows open, air conditioner turnoff	3.2	4.2 ± 3.4	4.0 ± 1.3	3.5 ± 1.0	3.8 ± 0.9	1:0.96:0.83
Door close, window(s) open, air conditioner turnoff	2.9	6.8 ± 3.8	5.4 ± 1.5	3.7 ± 1.0	4.9 ± 1.0	1:0.79:0.54
Door close, window(s) close, air conditioner working	0.27	10.3 ± 4.3	6.7 ± 1.7	4.4 ± 1.1	6.2 ± 1.1	1:0.65:0.43
Door close, window(s) close, air conditioner turnoff	0.26	16.1 ± 5.9	8.8 ± 2.2	10.4 ± 1.6	10.2 ± 1.4	1:0.54:0.65

There are four ventilation conditions were considered in this study, i.e. (1) door open, window(s) open, and air conditioner turnoff; (2) door closed, window(s) open and air conditioner turnoff; (3) door closed, window(s) closed, and air conditioner working; (4) door closed, window(s) closed, and air conditioner turnoff, the measured AERs of which were 3.2, 2.9, 0.27, and 0.26/h, respectively. It can be found that the AERs were the least when closed window(s) and door, where a working air-conditioner cannot elevate the AER efficiently in the room. These AER results were similar with Wang et al.'s results [16], among which they measured AERs in 6 dwellings under various ventilation conditions condition of ‘door closed, window(s) closed and air conditioner turnoff’. More work can be adapted into the indoor radon progeny model [17] to numerically simulate the relationship between the activity ratio and AER, which would be helpful to understand the variation of activity ratio.

Conclusions

This study reported the concentration ranges of short-lived radon progenies in typical indoor and outdoor environments in Beijing, China. The results showed that the radon EECs, concentrations of ^{218}Po, ^{214}Pb and ^{214}Bi in typical indoor environments in Beijing were ranged from 1.2 to 21.2, 1.5 to 32.5, 0.7 to 18.5, and 0.8 to 22.6 Bq/m^3, respectively. The EEC, concentrations of ^{218}Po, ^{214}Pb and ^{214}Bi in typical outdoor environments in Beijing were ranged from 1.6 to 12, 1.7 to 17.2, 0.6 to 13.4, and 1.1 to 13.1, respectively.

Moreover, the concentration ratios of radon progenies were also calculated and reported, the mean of which in typical indoor outdoor environments in Beijing were 1:0.59:0.58 and 1:0.50:0.67, respectively. The results of radon progeny concentration ratios varied in quite a large scale, and also the data in indoor environments was positively associated with the air exchange rate.

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