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# INTERCOMPARISON EXPERIMENT AND UNCERTAINTY ANALYSIS OF THREE MEASUREMENT METHODS FOR RADON PROGENY AT NIM

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**An intercomparison experiment of three radon progeny measurement methods, based on alpha spectrometry (Kerr method), alpha–beta spectrometry and liquid scintillation counter (LSC method), was carried out in the standard radon chamber at NIM. Both the consistency and the uncertainty analysis of three different methods were studied in detail. Results show that, at the EEC level of 4000 Bq/m$^3$, the uncertainties of the alpha–beta spectrometry, LSC method and Kerr method were 3.78%, 6.41% and 9.46%, respectively, which are mainly contributed by detection efficiency uncertainty, measuring time uncertainty and counting statistical uncertainty in sequence. The comparison results at different concentration levels show that the measurement values of three methods are consistent within the error range of 3%. The alpha–beta spectrometry can be used as a reference standard method to achieve the simultaneous measurement of RaA, RaB, RaC concentrations and EEC in the radon chamber.**

## INTRODUCTION

Radon is the second cause of lung cancer in the general population after smoking[1, 2], and the dose to lungs is predominantly caused by the deposition of radon progeny in bronchial airways[3, 4]. Accurate measurement of radon progeny concentration is an important way for dose estimation and risk reduction.

To assure the quality and comparability of different methods and instruments for the measurement of radon progeny, intercomparisons among laboratories and instruments are necessary. In the past decades, several intercomparison exercises have been carried out. An international intercalibration and intercomparison programme had been established by Organisation for Economic Cooperation and Development as early as 1983[5], and several intercomparison exercises were carried out in the next few years[6–9]. Later in 2002, four types of radon decay products monitors from three laboratories were intercompared in the radon/aerosol chamber at National Institute of Radiological in Japan[10]. During 2010–2014, three times of intercomparison exercises on radon progeny monitors were organised by the Natural Radiation Division of the National Radiation Protection Institute (NRPI) in Prague[11–13].

In order to set up a national standard of radon and its progeny in China and provide a high-quality calibration system for radon and radon progeny measuring devices by tracing to national standard, National Institute of Metrology (NIM) of China has established a radon chamber with a volume of 12.460 m$^3$[14]. Radon concentration, aerosol size and concentration, and other environmental parameters can be independently regulated to be stable for a long time or varied in a wide range. Control of radon progeny concentration can be realised by automatic regulation of radon concentration as well as aerosol concentration and particle sizes in the chamber. However, a measurement method or device which can be used as a reference to evaluate radon progeny concentration in radon chamber is still unavailable.

In this paper, aiming to explore the difference in performance between different measurement methods for radon progeny, and set up a standard reference, an intercomparison experiment was carried out in the radon chamber at NIM. Both the consistency and the uncertainty analysis of alpha spectrometry (Kerr method), alpha–beta spectrometry and liquid scintillation counter (LSC method) were studied in detail.

## MATERIALS AND METHODS

### Radon progeny measurement methods

Alpha–beta spectrometry is based on simultaneous sampling and measuring for the whole cycle. Counting numbers of beta region, 6.00 MeV alpha

particles and 7.69 MeV alpha particles are $N_1$, $N_2$ and $N_3$, respectively. Considering the collection and decay processes[15], $H_{ij}$ integrated from Formula (1) can be applied to the calculation Formula (2) for radon progeny concentrations. Then pick up detection efficiencies together to a matrix, and the concentration of radon progeny can be calculated by Formula (3):

$$\begin{aligned} H_{11} &= \int_0^T \frac{1-e^{-\lambda_1 \cdot t}}{\lambda_1}\mathrm{d}t = \frac{T}{\lambda_1} + \frac{\exp(-\lambda_1 T)-1}{{\lambda_1}^2} \\ H_{22} &= \int_0^T \frac{1-e^{-\lambda_2 \cdot t}}{\lambda_2}\mathrm{d}t = \frac{T}{\lambda_2} + \frac{\exp(-\lambda_2 T)-1}{{\lambda_2}^2} \\ H_{33} &= \int_0^T \frac{1-e^{-\lambda_3 \cdot t}}{\lambda_3}\mathrm{d}t = \frac{T}{\lambda_3} + \frac{\exp(-\lambda_3 T)-1}{{\lambda_3}^2} \\ H_{12} &= \frac{H_{11}-H_{22}}{\lambda_2-\lambda_1} \\ H_{23} &= \frac{H_{22}-H_{33}}{\lambda_3-\lambda_2} \\ H_{13} &= \frac{H_{12}-H_{23}}{\lambda_3-\lambda_1} \end{aligned} \tag{1}$$

$$\begin{aligned} N_1 &= E_\alpha F \varepsilon_\mathrm{f} H_{11} C_\mathrm{RaA} \\ N_2 &= E_\alpha F \varepsilon_\mathrm{f} (\lambda_2\lambda_3 H_{13} C_\mathrm{RaA} + \lambda_3 H_{23} C_\mathrm{RaB} + H_{33} C_\mathrm{RaC}) \\ N_3 &= E_\beta F \varepsilon_\mathrm{f} [(\lambda_2 H_{12} + \lambda_2\lambda_3 H_{13}) C_\mathrm{RaA} + \\ &\quad (H_{22} + \lambda_3 H_{23}) C_\mathrm{RaB} + H_{33} C_\mathrm{RaC}] \end{aligned} \tag{2}$$

$$\begin{pmatrix} C_\mathrm{RaA} \\ C_\mathrm{RaB} \\ C_\mathrm{RaC} \end{pmatrix} = \frac{1}{q\cdot\varepsilon_\mathrm{F}} \cdot \left[ \begin{pmatrix} 0 & \varepsilon_\beta & \varepsilon_\beta \\ \varepsilon_\alpha & 0 & 0 \\ 0 & 0 & \varepsilon_\alpha \end{pmatrix} \cdot \hat{\mathrm{T}} \right]^{-1} \begin{pmatrix} N_1 \\ N_2 \\ N_3 \end{pmatrix} \tag{3}$$

In which, $q$ is the flowrate, $\varepsilon_\alpha$ and $\varepsilon_\beta$ are detection efficiencies of alpha particles and beta particles, and $\varepsilon_\mathrm{F}$ is the collection efficiency of the filter. $\hat{\mathrm{T}}$ is a matrix only related to collection time and measurement cycle. For one hour cycle, $\hat{\mathrm{T}}$ can be given by the following formula:

$$\hat{\mathrm{T}} = \begin{pmatrix} 880744.61 & 0 & 0 \\ 414695.67 & 4109932.2 & 0 \\ 191763.15 & 1988279.5 & 3583186.5 \end{pmatrix} \tag{4}$$

Formula (5) can be used to calculate the relative uncertainties of radon progeny concentration.

Kerr method is based on grab sampling. The concentration of radon progeny can be calculated by Formula (6) based on Kerr's theory[16]:

$$\begin{pmatrix} e^2_\mathrm{RaA} \\ e^2_\mathrm{RaB} \\ e^2_\mathrm{RaC} \end{pmatrix} = \begin{pmatrix} 0 & 1 & 0 & 1 & 1 & 1 \\ 5.0629 & 0.010312 & 1.3192 & 1.5627 & 5.0629 & 1 \\ 3.1043 & 1.232\times10^{-5} & 7.6087 & 7.6281 & 3.1043 & 1 \end{pmatrix} \begin{pmatrix} e^2_{N_1} \\ e^2_{N_2} \\ e^2_{N_3} \\ e^2_{\varepsilon_\alpha} \\ e^2_{\varepsilon_\beta} \\ e^2_q \\ e^2_{\varepsilon_\mathrm{F}} \\ e^2_T \end{pmatrix} \tag{5}$$

$$\begin{pmatrix} C_\mathrm{RaA} \\ C_\mathrm{RaB} \\ C_\mathrm{RaC} \end{pmatrix} = \frac{1}{q\cdot\varepsilon_\alpha\cdot\varepsilon_\mathrm{F}} \cdot \hat{\mathrm{T}}^{-1} \cdot \begin{pmatrix} N_1 \\ N_2 \\ N_3 \end{pmatrix} \tag{6}$$

where $q$ is the flowrate, $\varepsilon_\alpha$ and $\varepsilon_\mathrm{F}$ are detection efficiency of alpha particles and collection efficiency of the filter. $\hat{\mathrm{T}}$ is the matrix decided by measurement cycle. For 1-h cycle, $\hat{\mathrm{T}}$ is given by Formula (7):

$$\hat{\mathrm{T}} = \begin{pmatrix} 35595.76 & 0 & 0 \\ 7150.797 & 85597.49 & 238456.9 \\ 20185.18 & 172879.5 & 208660.9 \end{pmatrix} \tag{7}$$

Formula (8) can be used to calculate the relative uncertainties of radon progeny concentration:

$$\begin{pmatrix} e^2_\mathrm{RaA} \\ e^2_\mathrm{RaB} \\ e^2_\mathrm{RaC} \end{pmatrix} = \begin{pmatrix} 16.2985 & 0.01409 & 0 & 16.2985 & 1 & 1 \\ 0.24618 & 74.4601 & 173.247 & 16.2985 & 1 & 1 \\ 0.01409 & 133.51 & 58.311 & 16.2985 & 1 & 1 \end{pmatrix} \begin{pmatrix} e^2_{N_1} \\ e^2_{N_2} \\ e^2_{N_3} \\ e^2_{\varepsilon_\alpha} \\ e^2_q \\ e^2_{\varepsilon_\mathrm{F}} \\ e^2_T \end{pmatrix} \tag{8}$$

LSC method is combined with grab sampling and three segments of spectrometry measuring. If counting rates of α and β particles in three measuring segments are $A_1$, $A_2$ and $A_3$, and the flowrate of grab sampling process is $q$, based on the algorithm[17], radon progeny concentration can be calculated by the following formula:

$$\begin{pmatrix} C_\mathrm{RaA} \\ C_\mathrm{RaB} \\ C_\mathrm{RaC} \end{pmatrix} = \frac{1}{q} \cdot \begin{pmatrix} 5.2714 & -2.5585 & 2.4188 \\ -0.4776 & 0.0581 & 0.2029 \\ -0.2609 & 0.4535 & -0.6453 \end{pmatrix} \begin{pmatrix} A_1 \\ A_2 \\ A_3 \end{pmatrix} \tag{9}$$

Statistical uncertainties are shown in the following formula:

$$\begin{pmatrix}\sigma_{\mathrm{RaA}}\\ \sigma_{\mathrm{RaB}}\\ \sigma_{\mathrm{RaC}}\end{pmatrix}=\frac{1}{q}\cdot\begin{pmatrix}27.788\ 6.5460\ 5.8506\\ 0.2281\ 0.0034\ 0.0412\\ 0.0681\ 0.2056\ 0.4165\end{pmatrix}\begin{pmatrix}A_1/C_{\mathrm{RaA}}\\ A_2/C_{\mathrm{RaB}}\\ A_3/C_{\mathrm{RaC}}\end{pmatrix} \quad (10)$$

And standard deviations of RaA, RaB and RaC can be combined with other independent uncertainties.

### Uncertainty of measurement system

For Kerr method and alpha–beta spectrometry, two RPM-SF01 radon progeny monitors (Sairatec, China) were used. Radon progeny can be collected on PTFE membrane filter (Haichengshijie, China) with collection efficiency of 98.6 ± 1.2%. Alpha and beta particles emitted from the filter can be recorded by a 400 $mm^2$ PIPS detector (SARAD, Germany). The detection efficiencies of the alpha–beta monitor were 28.48 ± 0.58% for alpha and 7.60 ± 0.15% for beta and the flowrate was 1.18 ± 0.01 L $min^{-1}$. Alpha detection efficiency of the Kerr monitor was 24.77 ± 0.51% and the flowrate was 2.65 ± 0.06 L $min^{-1}$. All the detection efficiencies were calibrated with electroplated $^{241}Am$ source and $^{90}Sr$ source. The flowrate can be recorded by a mass flowmetre (Honeywell AWN43600, USA) and calibrated by a primary air flow calibrator (Gilian, USA). Both radon progeny concentrations and their uncertainties can be calculated.

For LSC method, nitrocellulose membrane filter with 100% collection efficiency was used for grab sampling. The filter can be dissolved in the scintillation fluid after sampling. Tri-Carb 3100TR liquid scintillation counter (PerkinElmer, USA) was used for counting measurement, and concentrations of RaA, RaB and RaC, together with EEC, can be calculated.

## RESULTS AND DISCUSSION

Intercomparison experiment was carried out in the radon chamber at NIM. The temperature in the radon chamber was set to 25°C, and relative humidity was controlled around 25%. EEC level in the radon chamber was controlled at 4000 Bq $m^{-3}$ and 5000 Bq $m^{-3}$. The measurement results are shown in Figure 1, six groups of data were shown to compare the consistency between different measurement method, and 'NO.' is the number of data group. And also, a set of typical data with uncertainty analysis is shown in Table 1. It can be seen that EEC values of all the three methods reached a good consistency within the relative error range of 3%. It means the basic principles of three methods are consistent and three instruments can verify each other's measurement data on EEC. Uncertainty analysis for both EEC and concentrations of RaA, RaB and RaC was carried out at EEC level of 4000 Bq $m^{-3}$ and 5000 Bq $m^{-3}$, and shown in Table 1. Relative uncertainties of alpha–beta spectrometry, Kerr method and LSC method were 3.78%, 9.46%, 6.41% at 4000 Bq $m^{-3}$, and 3.82%, 10.49%, 5.73% at 5000 Bq $m^{-3}$, respectively.

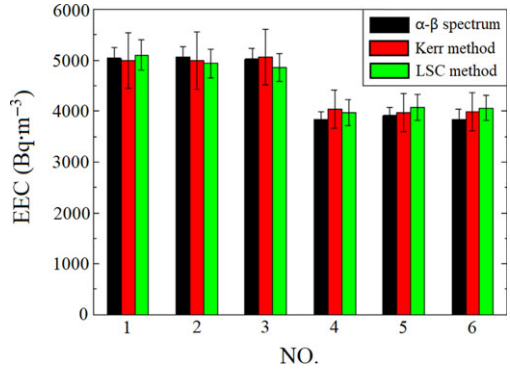


Figure 1. Intercomparison result of EEC values measured by three methods.

For alpha–beta spectrometry, the influence of sampling process has been minimised due to sampling and simultaneous measuring in the whole cycle, so the uncertainty is mainly contributed by detection efficiency of alpha and beta particles. It is interesting that uncertainties of RaB and RaC, whose calculation are involving beta counts, are higher than that of RaA, whose calculation only uses alpha counts of 6.00 MeV. This phenomenon indicates that the use of beta counting numbers may cause additional uncertainty, even if the total uncertainty is still relatively small. For Kerr method, 10 min of grab sampling and relatively short measuring time made counting numbers small and caused increasing of statistical uncertainty, especially for $N_1$ counts, which are main contributor to total uncertainty. As to LSC method, the error of measuring time contributes the most of total uncertainty because of manual operation. It is worth mentioned that the uncertainty of RaA is much higher than those of RaB and RaC. This is probably caused by higher calibration factor (first line of matrix elements in Formula (9)).

It is indicated by the above results that alpha–beta spectrometry performs the best and is suitable to use as the reference standard monitor in follow-up research work.

**Table 1. A set of typical data.**

| Measurement methods | $N_1$ | $N_2$ | $N_3$ | $\varepsilon_\alpha$ | $\varepsilon_\beta$ | $\varepsilon_f$ | $q$ (L/min) | Sampling time (min) | RaA (Bq/m$^3$) | RaB (Bq/m$^3$) | RaC (Bq/m$^3$) | EEC (Bq/m$^3$) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | Relative uncertainties | | | | | | Relative uncertainties | | |
| Alpha–beta spectrometry | 62 621 | 22 492 | 119 783 | 0.285 | 0.076 | 0.986 | 1.18 | 60 | 4701 | 3921 | 3726 | 3914 |
| | 0.40% | 0.67% | 0.29% | 2.03% | 2.03% | 1.22% | 0.74% | 0.17% | 2.57% | 5.51% | 6.89% | 3.78% |
| | 95 649 | 40 984 | 1 970 664 | 0.285 | 0.076 | 0.986 | 1.18 | 60 | 6240 | 4746 | 4966 | 4987 |
| | 0.32% | 0.49% | 0.23% | 2.03% | 2.03% | 1.22% | 0.74% | 0.17% | 2.54% | 6.22% | 6.01% | 3.82% |
| Kerr method | 1892 | 13 437 | 17 988 | 0.248 | — | 0.986 | 2.65 | 10 | 5241 | 3943 | 3655 | 3970 |
| | 2.30% | 0.86% | 0.75% | 2.06% | — | 1.22% | 2.35% | 1.00% | 12.78% | 15.22% | 14.49% | 9.46% |
| | 1422 | 10 190 | 13 390 | 0.248 | — | 0.986 | 2.65 | 10 | 6770 | 4830 | 4899 | 5060 |
| | 2.65% | 0.99% | 0.86% | 2.06% | — | 1.22% | 2.35% | 1.00% | 13.85% | 17.45% | 15.49% | 10.49% |
| LSC method | 24 214 | 20 566 | 17 089 | 1.000 | 1.000 | 0.986 | 3.42 | 10 | 5789 | 3935 | 3524 | 3973 |
| | 0.64% | 0.70% | 0.77% | 0.50% | 0.50% | 1.22% | 0.73% | 2.00% | 32.81% | 3.65% | 10.14% | 6.41% |
| | 30 983 | 26 338 | 21 845 | 1.000 | 1.000 | 0.986 | 3.42 | 10 | 7152 | 5017 | 4604 | 5084 |
| | 0.57% | 0.62% | 0.68% | 0.50% | 0.50% | 1.22% | 0.73% | 2.00% | 30.59% | 3.44% | 8.90% | 5.73% |

[a]All the uncertainties are relative uncertainties.

## CONCLUSIONS

An intercomparison experiment of three radon progeny measurement methods, based on Kerr method, alpha–beta spectrometry and LSC method, was carried out in the standard radon chamber at NIM. At the EEC level of 4000 Bq m$^{-3}$, the uncertainties of alpha–beta spectrometry, LSC method and Kerr method were 3.78%, 6.41% and 9.46%, respectively, which are mainly contributed by detection efficiency uncertainty, measuring time uncertainty and counting statistical uncertainty, respectively. The alpha–beta spectrometry can be used as a reference standard method to achieve the simultaneous measurement of RaA, RaB, RaC concentrations and EEC in the radon chamber, but the accuracy and uncertainty of beta detection efficiency need to be further considered.

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