MEASUREMENT OF THE ²¹²Pb PARTICLE SIZE DISTRIBUTION INDOORS

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A new device has been developed for the measurement of the 212 Pb particle size distribution indoors. This device consists of two wire screens and a back-up filter with a diameter of 2.0 cm. The sampling flow rate is typically 3.0 l min⁻¹. After 3-h sampling time and 6-h waiting time, a CR-39 detector is used for the registration of the alpha particles from the 212 Pb, deposited on the wire screens and the filter, respectively. It appears clear from field measurements that there are no appreciable differences among the particle size distributions from different dwellings within the same location and under the same climate conditions. However, the 212 Pb particle size distributions from the countryside dwellings have different results from those of the city dwellings.

INTRODUCTION

The decay products of radon $(^{222}$ Rn) and thoron $(^{220}$ Rn) gas, once inhaled, may be conducive to well-known health risks⁽¹⁾.

While radon and its progenies have been widely studied since the 1960s, the issue of thoron and its decay products has been of increasing interest only in the past two decades^(2, 3). The ²¹²Pb (also known as ThB) is the most important nuclide among thoron progenies because of its large contribution in the dose⁽⁴⁾.

In particular, the measurement of the size distribution of indoor 212 Pb is very important for the correct evaluation of the dose due to thoron exposure⁽⁵⁻⁸⁾.

In general, there are two types of instruments for the measurement of particle size distribution, such as the low-pressure cascade impactor⁽⁹⁾ and the screen diffusion battery⁽¹⁰⁾. Both types of instruments are too bulky and complex for their use in field measurements.

The goal of the present paper is to develop a portable unit for the measurement of the 212 Pb particle size distribution by using CR-39 detectors.

MATERIALS AND METHODS

Basic theory

Among the different short-lived thoron decay products, 216 Po because of its short half-life (0.15 s.) exists only as an unattached particle, whereas 212 Pb (with an half-life of 10.64 h) is typically attached to aerosols. The 212 Bi has the same size and spatial distribution as 212 Pb⁽⁹⁾. Considering the limited amount of unattached 212 Pb particles and its limited

contribution to the total dose exposure, it can be assumed that all the ²¹²Pb particle aerosols exist as attached particles in the indoor environment.

In particular, for the dose assessment it can be assumed that all the ²¹²Pb are attached to indoor particles.

For the evaluation of the dose due to thoron exposures, both the activity median aerodynamic diameter (AMAD) and geometric standard deviation (GSD) of 212 Pb/ 212 Bi particle size distribution are important input parameters⁽¹¹⁾. To this end, a log–normal distribution can be used to describe aerosol-particle size distribution, when there is no extra-aerosol source in the indoor environment. In this present paper, the attached 212 Pb are described in terms of a log–normal distribution, which is the same as that of the total indoor aerosol particles.

To carry out the field measurement of ²¹²Pb particle size distribution, a new device has been developed, as schematically shown in Figure 1.

This device consists of three holders with a diameter of 2.0 cm, containing, respectively, a 400-mesh screen, a 635-mesh screen and a back-up filter. Indoor aerosol particles are collected at a flow rate of $3.0 \ 1 \ \text{min}^{-1}$. The aerodynamic cut-off diameters, corresponding to particle diameter collected with the efficiency of 50 % (according to the basic theoretical calculation⁽¹²⁾), are 4.32 and 6.88 nm for 400 and 635 mesh, respectively. A sampling time of 3 h (termed T1) has been chosen on the basis of the large differences between the half-life of ²¹²Pb and those of radon decay products, together with the desired low detection limit. The selective measurement of ²¹²Pb has been facilitated by a waiting time of 6 h in order to allow the decay of the radon progenies. Finally, the measurements of the alpha



Figure 1. The diagram of the screen array for ²¹²Pb particle size distribution measurement.

particles, emitted from the 212 Pb, deposited on the wire screens and on the filter, have been carried out by facing them against CR-39 detectors for 3 d.

Calculation methods

The track density of the CR-39 detectors, relative to the 400 mesh, to the 635 mesh and to the back-up filter can be described by the following equations:

$$D_{400} = Q_0 T_1 \times \frac{C_{\text{ThB}} e^{-\lambda_{\text{ThB}} (T_1/2 + T_2)}}{\lambda_{\text{ThB}}} \times \frac{1 - \eta_1}{S}, \quad (1)$$

$$D_{635} = Q_0 T_1 \times \frac{C_{\text{ThB}} e^{-\lambda_{\text{ThB}} (T_1/2 + T_2)}}{\lambda_{\text{ThB}}} \times \frac{1 - \eta_2}{S}, \quad (2)$$

$$D_{\text{filter}} = Q_0 T_1 \times \frac{C_{\text{ThB}} e^{-\lambda_{\text{ThB}} (T_1/2 + T_2)}}{\lambda_{\text{ThB}}} \times \frac{\eta_1 \cdot \eta_2}{S}, \qquad (3)$$

where Q_0 is the flow rate, 3.0 l min⁻¹; λ_{ThB} is the ²¹²Pb decay constant; S is the effective area of CR-39 (3.14 cm²); C_{ThB} is the ²¹²Pb concentration (Bq cm⁻³). The second term above represents the decay loss of the ²¹²Pb during sampling and waiting time; η_1 and η_2 are the penetration rates of a lognormal distribution particle, f(d), passing through wire screen with mesh 400 and 635, respectively.

In order to reconstruct the aerosol particle size distributions, the inversion methods, such as the Twomey method and expectation maximisation

Table 1. Intercomparison experiment of ²¹²Pb size distribution in two indoor environments.

| | CMD ^a (nm) | GSD | AMAD ^b (nm) | AMAD (nm) | GSD |
|--------|--------------------------|------|---------------------------|--------------|-----|
| Site A | 34.5 | 2.39 | 51.8 | 50.0 | 3.1 |
| Site B | 97.1 | 1.76 | 145.5 | 160.0 | 2.2 |

^aAerosol size distribution measured by a SDB (with 1, 2, 4, 8, 16 and 32 layers of mesh 500 screen).

^bCalculation by method as used by Cavallo⁽¹⁵⁾: AMAD=CMD×1.5.

method (EM method)⁽¹³⁾ are usually adopted. In the present paper, a Monte Carlo method has been developed for the evaluation of the screen-array response, which has been termed ThBSDC (ThB size distribution calculation). In practice, η_1 and η_2 are calculated on the basis of the measurements of both the AMAD and GSD of the size distribution over the ranges of 10–500 and 1.1–4.0 nm. Comparing the theoretical results with those measured, the final values of AMAD and GSD can be obtained.

Considering the error of the CR-39 detectors, the variation for random sampling of AMAD and GSD values are designed to be 5 and 0.1 nm, respectively.

RESULTS AND DISCUSSION

Comparison experiment

An experiment has been carried out in two different indoor environments by using a screen diffusion battery $(SDB)^{(14)}$ for comparison. The results of this comparison are shown in Table 1. It appears clear from Table 1 that there is a good agreement between measured values and those calculated.

Field measurement

Field measurements have been carried out in dwellings from different areas. The results from these measurements (reported in Table 2) show a good agreement between measured values and the calculated ones.

From the data in Table 2, it appears clear that there no large difference for ²¹²Pb particle size distributions between different dwellings within the same area and under the same climate conditions. For the field measurements in Yangjiang, the measurements were performed in rainy days at the beginning. By contrast, the last three sets of data have been obtained under good weather conditions. Furthermore, it can also be found that the AMADs of the ²¹²Pb in the countryside or suburb dwellings are smaller than those in cities or downtown.

| Site | House/ground | AMAD (nm) | GSD | Average |
|-------------------------------|--------------|-----------|-----|--------------------------|
| Yangjiang, Guangdong Province | Mud/mud | 40 | 2.7 | AMAD, 63.8 nm: GSD, 2.7 |
| | | 30 | 2.6 | ,,, |
| | | 40 | 1.9 | |
| | Brick/mud | 50 | 3.3 | |
| | | 60 | 2.5 | |
| | Brick/cement | 130 | 2.7 | |
| | Brick/mud | 70 | 3.0 | |
| | Brick/cement | 90 | 2.9 | |
| Datong, Shanxi Province | Cave/mud | 40 | 2.1 | AMAD, 50.0 nm; GSD, 3.1 |
| | Cave/mud | 50 | 3.6 | · · · · |
| | Cave/mud | 60 | 3.6 | |
| Beijing suburb | Brick/mud | 110 | 2.7 | AMAD, 110.0 nm; GSD, 2.5 |
| 5 0 | Brick/mud | 90 | 2.3 | , , , , |
| | Brick/floor | 130 | 2.5 | |
| Beijing downtown | Brick/cement | 160 | 2.2 | AMAD, 155.0 nm; GSD, 2.0 |
| · · · | Brick/cement | 150 | 1.7 | . , , , |

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| Table 2. The measurements of the size usurbution in unreferred muoor chynomicia |
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It should be noticed that the results from field measurements, listed in Table 2, seem smaller than those previously reported. These differences may be due to the use of the low-pressure cascade impactors, characterised by detection limit as large as 80 nm and by the low particle sizes in the countryside.

CONCLUSIONS

A portable and low-cost screen array using a CR-39 detector has been developed for field measurements of the ²¹²Pb particle size distribution.

Pilot measurements have been carried out in different dwellings from different areas. No appreciable differences have been encountered among dwellings from the same area and under the same weather conditions. Relatively small particle sizes have been measured in countryside dwellings, when compared with those from city dwellings. These differences may be due to the better ventilation and to the relatively clean atmospheric conditions of the countryside dwellings.

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