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Controlling ^{212}Bi to ^{212}Pb activity concentration ratio in thoron chambers

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ABSTRACT

It is necessary to establish a reference atmosphere in a thoron chamber containing various ratios of ^{212}Bi to ^{212}Pb activity concentrations ($C(^{212}Bi)/C(^{212}Pb)$) to simulate typical environmental conditions (e.g., indoor or underground atmospheres). In this study, a novel method was developed for establishing and controlling $C(^{212}Bi)/C(^{212}Pb)$ in a thoron chamber system based on an aging chamber and air recirculation loops which alter the ventilation rate. The effects of main factors on the $C(^{212}Bi)/C(^{212}Pb)$ were explored, and a steady-state theoretical model was derived to calculate the ratio. The results show that the $C(^{212}Bi)/C(^{212}Pb)$ inside the chamber is mainly dependent on ventilation rate. Ratios ranging from 0.33 to 0.83 are available under various ventilation. The stability coefficient of the ratios is better than 7%. The experimental results are close to the theoretical calculated results, which indicates that the model can serve as a guideline for the quantitative control of $C(^{212}Bi)/C(^{212}Pb)$.

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1. Introduction

The annual effective dose from inhalation of thoron (^{220}Rn) and thoron progeny (^{212}Pb and ^{212}Bi) contributes on average 9% to that from inhalation of radon and radon progeny (UNSCEAR, 2000). Recently, some publications reported that thoron activity concentrations are higher than radon activity concentrations in high background radiation areas located in China and India (Kudo et al., 2015; Omori et al., 2015, 2016; Yuan et al., 2000; Omori et al., 2016; Bajwa et al., 2015). Special attention should be given to the risk of inhalation of thoron and its decay products in those high background radiation locations rife with thorium-rich soil. Indoor thoron activity concentration is different from measurement positions for its short half-life (55.6 s). Thus, measurement of thoron progeny is necessary for thoron dose assessment (Tokonami et al., 2004). The contribution of ^{212}Bi to equilibrium-equivalent thoron concentration is only about 8%, however, the dose conversion factor of ^{212}Bi is more than two times higher than that of ^{212}Pb in inhalation dose calculation (Li et al., 2008). The combined effect of these factors means that the activity concentrations of the two decay products must be accurately measured in the indoor environment in order to accurately assess their doses.

^{212}Pb has a relatively longer half-life (10.64 h) as compared to ^{212}Bi (60.55 min). The value of $C(^{212}Bi)/C(^{212}Pb)$ is likely equal to 1 when their residence time exceeds 5 h, but ventilation and plate-out of thoron progeny decrease $C(^{212}Bi)/C(^{212}Pb)$. Several indoor environment surveys conducted in different places indicate that $C(^{212}Bi)/C(^{212}Pb)$ differ widely in different environments. According to Reineking et al. (1992), $C(^{212}Bi)/C(^{212}Pb)$ is 0.4 on average and ranges from 0.1 to 0.68 in indoor environments; it is 0.3 on average and ranges from 0.06 to 0.71 outdoors. Stoute et al. (1984) measured this ratio to be 0.61 on average (ranging from 0.21 to 1.17) in a concrete basement, while a value of 0.34 was measured in one-storey house by Zarcone et al. (1986). In order to obtain reliable data on $C(^{212}Bi)/C(^{212}Pb)$ from thoron progeny monitors, it is essential to calibrate the monitors in the thoron chamber containing reference atmospheres with various $C(^{212}Bi)/C(^{212}Pb)$.

The international standard (IEC 61577–4, 2009) does not state specific information about $C(^{212}Bi)/C(^{212}Pb)$; several thoron chambers were established mainly for calibrating thoron monitors, while relatively few measures have been taken to control $C(^{212}Bi)/C(^{212}Pb)$ (Röttger et al., 2009, Buompane et al., 2014, Sorimachi et al., 2014, Gargioni et al., 2003, Kobayashi et al., 2005, Zhao et al., 2010, Möre et al., 1986, Pressyanov et al., 2017). In this work, a method based on different ventilation rates in a thoron chamber system was developed to control various $C(^{212}Bi)/C(^{212}Pb)$.

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2. Materials and methods

2.1. Thoron chamber construction

Fig. 1 presents the thoron chamber system layout diagram consisting of five components: A stainless steel aging chamber, a calibration chamber, solid thoron sources, an aerosol generation and measurement system, and air circuit silicone tubes with an inner diameter of 8 mm. The middle section of the aging chamber is a cylinder (Φ600 × 1200 mm) and both ends are circular truncated cones (the upper base diameter is 25 cm, lower base is 60 cm; slant height is 40 cm) with a total volume of 443 L. The calibration chamber is a rectangular solid (900 × 900 × 1850 mm) with a volume of 1500 L. Two kinds of gas-through solid thoron sources were utilized to produce thoron: One (34 kBq) connected with a pump (VLK, Qihai Ltd., China) to transfer thoron gas into the aging chamber, and the other (30 kBq) employing a fan to transfer thoron gas into the calibration chamber. These sources are made of aged $Th(NO_3)_4$ which was taken as a standard material produced by Amersham (The Radiochemical Centre Ltd., Amersham, Buckinghamshire England) at least 40 years ago. The combined standard uncertainty of the thoron source should be about 3.5%. The emanation coefficient of the two thoron sources is 96.5± 3%, and the thoron yield is constant with a relative standard deviation (RSD) below 2.5% over five years (relative humidity ranges from 50% to 90%, temperature ranges from 5 °C to 38 °C) (Qiu, 2006). Monodispersed aerosol was generated by a condensation monodisperse aerosol generator (CMAG, TSI 3475, USA) utilizing di-2-ethyl hexyl sebacate (DEHS) as the aerosol material. Polydisperse aerosol was generated by burning smokeless incense (HB2060, Gucheng Incense Group Ltd., China) at 1 h intervals in the aging chamber. Aerosol concentration and size distribution were measured at the center of the two chambers with a condensation nucleus counter (CNC 3022, TSI Inc., USA) and electrical low pressure impactor (ELPI, Dekati Ltd., Filand), respectively. An alarm-hygrometer (Model 608-H2, Testo AG, Germany) was utilized to measure air temperature and relative humidity. The sensor is located at the bottom of the thoron chamber.

Aged thoron progeny in the aging chamber was injected into the calibration chamber at a flow rate of v_1 which can be adjusted by the speed pump. Air in the calibration chamber was continuously extracted by a pump (VT 4.4, Becker Ltd., Germany) with a flow rate of v_2, then pumped into the atmosphere or the chamber after being filtered with a high-efficiency particulate-air (HEPA) filter. Filtering the thoron progeny in this manner is similar to natural ventilation, but can be controlled at a comparatively more stable speed. Air in the system can be run in opened or closed mode by adjusting the three-way valve.

2.2. Measuring activity concentrations of thoron progeny

Thoron progeny were sampled by filter membranes (aawp02500, 25 mm diameter, Merck Milipore Ltd., USA). After a sampling period of 10 min, activity in the filter was counted for two counting intervals (2–43 min and 43–120 min) with an Ortec Alpha Ensemble spectrometer (Ortec Plus, Ortec Inc., USA) (Kang et al., 2008). The sampling flow rate was calibrated with a soap film flowmeter (Gilibrator2 model, Sensidyne Inc., USA) with relative standard uncertainty of 1%. Detection efficiency was calibrated by an ^{241}Am standard surface source with relative standard uncertainty of 2%. The combined uncertainty of measurement results including radioactive statistical fluctuation was controlled within 5%. The details of the propagation of the combined uncertainty are presented in Appendix A.

2.3. Theoretical model for $C(^{212}Bi)/C(^{212}Pb)$ calculation

Thoron activity concentration increases for decay of ^{224}Ra in the thoron source, and decreases for decay. Thus the stable thoron activity A_{Tn}^{A} in the aging chamber is given by:

$$A_{Tn}^{A} = \varepsilon \cdot A_{Ra} \cdot \exp\left(- \lambda_{Tn} \frac{V_{in}}{v_1} \right) \tag{1}$$

where λ_{Tn} is the decay constant of thoron, A_{Ra} is the activity of the solid thoron source, ε is the emanation coefficient of the thoron source, and V_{in} is the volume of the tube connecting the thoron source and aging chamber. The effect of ventilation on the thoron activity in the ageing chamber was neglected.

Because the half-life of ^{216}Po is very short and the decay constant of ^{216}Po ($1.66 \times 10^4\ h^{-1}$) is larger than the attachment coefficient (50 ~ 150 h^{-1}) for unattached ^{216}Po atoms to attach to aerosol particles (Porstendörfer, 1984), ^{212}Pb can be seen as decay

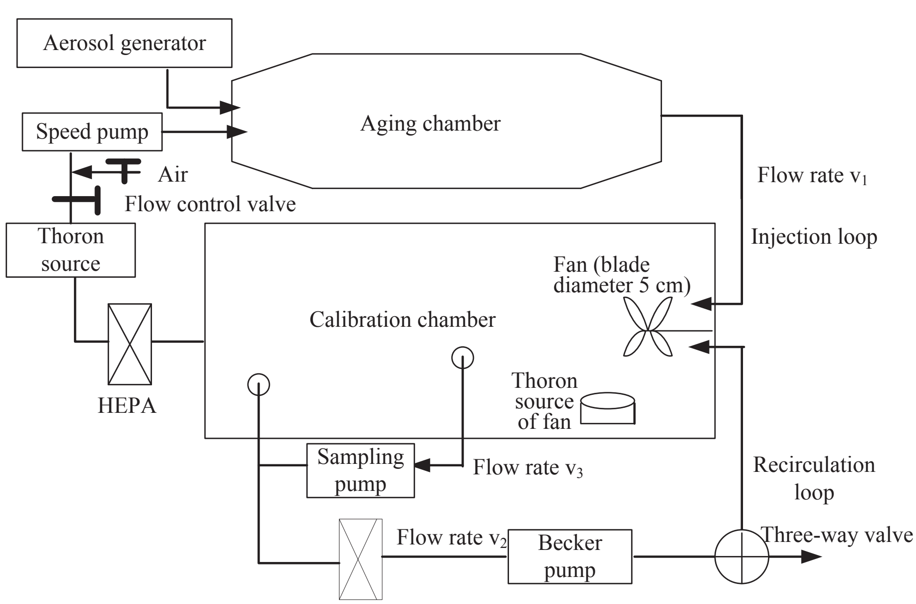

Fig. 1. Schematic diagram of the thoron chamber.

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