



Long-term measurements of unattached radon progeny concentrations using solid-state nuclear track detectors

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ABSTRACT

We described a method for long-term passive measurements of unattached fraction f_p of the potential alpha energy concentration of radon progeny from a set of measured (f_1, f_2, f_3) values, where $f_i = C_i/C_0$ ($i = 1, 2, 3$), and C_0, C_1, C_2 and C_3 were the concentrations of ^{222}Rn , and the airborne concentrations of ^{218}Po , ^{214}Pb and ^{214}Bi , respectively. Jacobi room model parameters were randomly sampled from their lognormal distributions to search for (f'_1, f'_2, f'_3) sufficiently close to (f_1, f_2, f_3) and to determine f_p . There was 99% and 88% chance obtaining an estimated f_p value within 50% and 30% of the true value, respectively.

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1. Introduction

Epidemiological studies have provided reasonably firm estimates of the risk of radon-induced lung cancers. The radon-related absorbed dose in the lung is mainly due to short-lived radon progeny, i.e., ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po , but not to the radon (^{222}Rn) gas itself. Long-term measurements of the concentrations of radon progeny or the equilibrium factor F and the unattached fraction f_p of the potential alpha energy concentration (PAEC), among other information such as the size distribution of radon progeny are needed to accurately assess the health hazards contributed by radon progeny. In particular, there is a potentially large contribution of f_p to the effective dose (or the dose conversion factor DCF). We here take a nominal case where $F = 0.372$ as our example, and perform calculations with our own computer program LUNGDOSE.F90 described by Nikezic and Yu (2001). The DCF will be equal to 13.0 and 18.6 mSv/WLM for $f_p = 0.0343$ and 0.196, respectively. The need for correct f_p values seems pertinent for realistic determination of the effective lung dose.

We denote C_0, C_1, C_2, C_3 and C_4 as the concentrations of ^{222}Rn , and the airborne concentrations of the first (^{218}Po), second (^{214}Pb), third (^{214}Bi) and fourth (^{214}Po) radon progeny, respectively (which are short-lived radon progeny). It is well established that ^{214}Bi and ^{214}Po are always in equilibrium due to the very short half life of ^{214}Po (164 μs), so we will simply use C_3 ($= C_4$) to denote both the airborne concentrations of ^{214}Bi and ^{214}Po . The radon progeny can be further separated into the unattached mode and the attached mode. As such, we also denote C_1^u, C_2^u and C_3^u as

the airborne concentrations of ^{218}Po , ^{214}Pb and ($^{214}\text{Bi}/^{214}\text{Po}$) in the unattached mode, respectively (with $C_3^u = C_4^u$), and C_1^a, C_2^a and C_3^a as the airborne concentrations of ^{218}Po , ^{214}Pb and ($^{214}\text{Bi}/^{214}\text{Po}$) in the attached mode, respectively (with $C_3^a = C_4^a$). Without the superscripts the quantities represent the total concentrations, i.e., in both attached and unattached modes. The equilibrium factor F is defined as

$$F = 0.105f_1 + 0.515f_2 + 0.380f_3 \quad (1)$$

where $f_i = C_i/C_0$ ($i = 1, 2, 3$). The unattached fraction f_p of PAEC is defined as

$$f_p = (0.105f_1^u + 0.515f_2^u + 0.380f_3^u) / (0.105f_1 + 0.515f_2 + 0.380f_3) \quad (2)$$

where $f_i^u = C_i^u/C_0$ ($i = 1, 2, 3$).

A common practice for radon hazard assessment nowadays is to first determine C_0 and then apply an assumed F with a typical value between 0.4 and 0.5. However, in reality, F varies significantly with time and place, and an assumed F cannot reflect the actual conditions (Yu et al., 1996a, b, 1997, 1999; Nikezic and Yu, 2005). This problem cannot be solved through active measurements based on air filtering, since they only give short-term determinations. Methods on long-term measurements of radon progeny concentrations or F using solid-state nuclear track detectors (SSNTDs) have been reviewed and proposed (Amgarou et al., 2003; Nikezic and Yu, 2004; Ng et al., 2007; Yu et al. 2005, 2008). Feasible methods included the “reduced equilibrium factor” (F_{red}) method proposed by Amgarou et al. (2003) and the “proxy equilibrium factor” (F_p) proposed by Nikezic et al. (2004).

On the other hand, however, there were no previous attempts for long-term passive measurements of f_p , except a recent

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preliminary theoretical treatment (Nikezic and Yu, 2010). A common method to measure f_p was to use a single wire screen (see, e.g., James et al., 1972; George, 1972; Raghavaya and Jones, 1974; Stranden and Berteig, 1982; Yu et al., 1996a,b). However, as commented by Tymen et al. (1999), “these methods grab short-duration samples; even if measurements can be automated, assessing long-time exposure can be difficult”. Consequently, Tymen et al. (1999) proposed an active experimental device involving pumps based on an annular diffusion channel, equipped with an LR 115 SSNTD, for estimation of the mean activity concentration of nanometer-sized ^{218}Po over several weeks in indoor atmospheres. However, passive measurements of f_p over a much longer period, say, over a few months or even a year, would be desirable as the results would not be biased due to shorter-term climatic and seasonal changes and would be more representative. In the present work, a method based on solid-state nuclear track detectors (SSNTDs) was proposed for the long-term passive measurements of f_p .

2. Methodology

Our method was based on the Jacobi room model (Jacobi, 1972). In the Jacobi room model, f_i^t and f_i^a ($f_i^t = C_i^t/C_0$; $i = 1, 2, 3$) are the functions of a set of Jacobi room model parameters ($\lambda_v, \lambda_a, \lambda_d^u, \lambda_d^g$) where λ_v is the ventilation rate, λ_a is the aerosol attachment rate, λ_d^u is the deposition rate of unattached progeny and λ_d^g is the deposition rate of attached progeny. The values of f_1, f_2 and f_3 are inter-related with relationships governed by the set of Jacobi room model parameters ($\lambda_v, \lambda_a, \lambda_d^u, \lambda_d^g$). In other words, from a set of ($\lambda_v, \lambda_a, \lambda_d^u, \lambda_d^g$), we could obtain (f_1, f_2, f_3) using the Jacobi room model.

From experimental measurements, a set of (f_1, f_2, f_3) values can be obtained. There can be different methods for long-term measurements of f_1, f_2 and f_3 . We here used the method proposed by Amgarou et al. (2003) as an example. Most SSNTDs can only record alpha particles, so they can give information on f_1 and f_3 (e.g., Amgarou et al. 2003; Nikezic and Yu, 2010) or the sum of ($f_1 + f_3$) (e.g., Nikezic et al. 2004; Yu et al. 2005), but not on f_2 . The “reduced equilibrium factor” F_{red} defined as $F_{red} = 0.105f_1 + 0.380f_3$, as proposed by Amgarou et al. (2003), had a very tight correlation with F . Fig. 1 shows the relationship between F and F_{red} , obtained by sampling the Jacobi room model parameters from lognormal distributions (Yu and Nikezic, 2011), which shows a very tight and linear relationship. The best-fit equation to the relationship now became

$$F = 2.26717 \times F_{red} - 0.03194 \quad (3)$$

As such, once f_1 and f_3 and thus F_{red} were known, F could be determined from Eq. (3), and f_2 could be calculated from the difference between F_{red} and F .

Our objective was to propose a method to make use of the set of experimentally obtained values of (f_1, f_2, f_3) to derive the set of Jacobi room model parameters ($\lambda_v, \lambda_a, \lambda_d^u, \lambda_d^g$), from which f_p (and also F) could be determined. The strategy employed here was to randomly sample the Jacobi room model parameters from their lognormal distributions and then to calculate sets of (f_1', f_2', f_3') values. Yu and Nikezic (2011) pointed out that uniform sampling of Jacobi room model parameters previously employed for computer simulations (e.g., Amgarou et al., 2003, Nikezic et al., 2004, Yu et al., 2005) had discarded the information provided by the best estimated values of these Jacobi room model parameters and had ignored the fact it was unlikely to obtain parameters far away from their best estimated values. Accordingly, Yu and Nikezic (2011) established lognormal distributions for the Jacobi room model parameters, with median values and geometric standard deviations (σ_g) shown in Table 1. These lognormal distributions

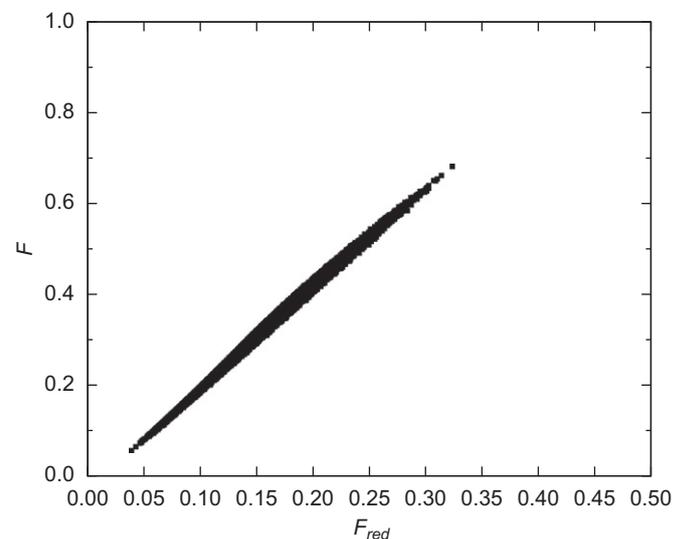


Fig. 1. The relationship between the equilibrium factor F on the reduced equilibrium factor F_{red} , obtained by sampling the Jacobi room model parameters from lognormal distributions (Yu and Nikezic, 2011).

Table 1

Median values and geometric standard deviations (σ_g) for Jacobi room model parameters employed for the computer simulations (Yu and Nikezic, 2011).

| Parameter | Median (s^{-1}) | σ_g |
|---|---------------------|------------|
| Ventilation rate λ_v | 0.55 | 1.3 |
| Aerosol attachment rate λ_a | 50 | 1.8 |
| Deposition rate of unattached progeny λ_d^u | 20 | 1.4 |
| Deposition rate of attached progeny λ_d^g | 0.2 | 1.4 |

generated more realistic distributions for F and f_p (Yu and Nikezic, 2011).

A total of $(2-3) \times 10^5$ sets of Jacobi room model parameters were generated for each set of (f_1, f_2, f_3). In such an approach, it would still be unlikely to be able to generate exactly the same (f_1, f_2, f_3) values as those required, so we allowed some small discrepancies to facilitate the computations. We accepted a set of (f_1^*, f_2^*, f_3^*) values if all $(f_i^{*^{-1}} - f_i^{-1})/f_i^{-1}$ ($i = 1, 2, 3$) were less than 1%. The asterisked values referred to those corresponding to an accepted solution. There could be other forms of acceptance criteria, e.g., $(f_i^* - f_i)/f_i$ could also be an index used in the criterion. While F is a linear combination of f_i , f_p varies approximately inversely with F (e.g., Tymen et al. 1992; Reineking et al. 1994). As such, we hoped $(f_i^{*^{-1}} - f_i^{-1})/f_i^{-1}$ would give better estimations of f_p . The anticipated problem here then was that a set of (f_1, f_2, f_3) values did not necessarily give a unique set of ($\lambda_v^*, \lambda_a^*, \lambda_d^{u*}, \lambda_d^{g*}$) or (f_1^*, f_2^*, f_3^*), and thus a unique value of f_p^* . Without better information, we obtained final values of $\langle f_p^* \rangle$ from all the feasible solutions by calculating the arithmetic means as well as the geometric means. However, our preliminary results showed no significant differences in the accuracy to determine $\langle f_p^* \rangle$ using arithmetic or geometric means, so we restricted ourselves to using arithmetic means in the present work.

To save computation time, if we already achieved 200 sets of (f_1^*, f_2^*, f_3^*), which fulfilled the acceptance criteria for a particular set of (f_1, f_2, f_3), the computation was stopped even if the number $(2-3) \times 10^5$ had not been reached.

3. Verifications using simulated scenarios

To demonstrate the feasibility of the current methodology, we performed verifications of simulated scenarios of f_p by randomly

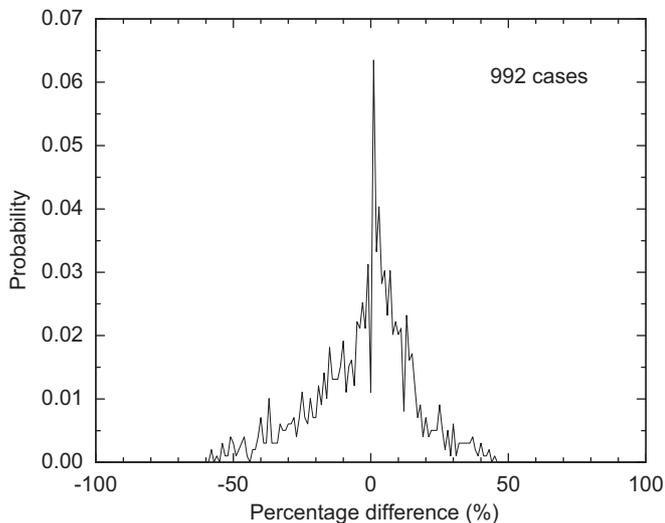


Fig. 2. Probabilities of getting different percentage differences defined by $(f_p - \langle f_p^* \rangle) / f_p$ for 992 solved cases, with $\langle f_p^* \rangle$ calculated using arithmetic means. Here, $\langle f_p^* \rangle$ and f_p^* are directly determined from f_1, f_2 and f_3 generated from the generated Jacobi room model parameters.

sampling the Jacobi room model parameters from their lognormal distributions. A total of 1000 sets of (f_1, f_2, f_3) values were generated, from which f_p^* and $\langle f_p^* \rangle$ were determined. Within the 1000 cases, solutions were found for only 992 cases. For the remaining 8 cases, no (f_1^*, f_2^*, f_3^*) could be found to satisfy the criteria $(f_i^{*-1} - f_i^{-1}) / (f_i^{-1}) < 0.01$ for $i = 1, 2, 3$. These unsolved cases corresponded to more extreme values (i.e., those far away from the median values) in one or more Jacobi room parameters.

The percentage difference was defined by $(f_p - \langle f_p^* \rangle) / f_p$, where $\langle f_p^* \rangle$ and f_p are the values of the unattached fraction estimated using the present methodology and that originally generated by randomly sampling the Jacobi room parameters, respectively. Fig. 2 shows the probabilities of getting different percentage differences for these 992 solved cases out of a total of 1000 cases. The probability of getting percentage differences from -50% to 50% was 0.989, while that from -30% to 30% was 0.884. These were very satisfactory given the simple procedures as well as the relatively relaxed acceptance criterion.

4. Discussion

The present paper developed a method for long-term passive measurements of f_p using SSNTDs. We made use of the lognormal distributions for the Jacobi room model parameters introduced by Yu and Nikezic (2011), which were found to generate more realistic scenarios. With accurately determined (f_1, f_2, f_3) values, there would be about 99% and 88% chances obtaining an estimated f_p value within $\pm 50\%$ and $\pm 30\%$ of the true value, respectively. These were very satisfactory given the simple procedures as well as the relatively relaxed acceptance criterion. It was apparent that the accuracy depended on this criterion.

The current criterion was adopted to provide a reasonable precision, which could be obtained within a reasonable computation time. We could specify a higher precision, say, $(f_i^{*-1} - f_i^{-1}) / (f_i^{-1})$ ($i = 1, 2, 3$) < 0.001 , but we might then have to substantially increase the number of Jacobi room model parameter sets beyond 3×10^5 to obtain solutions, and this would result in a substantial increase in the computation time. Other feasible indices, such as $(f_i^* - f_i) / (f_i)$, could also be explored for the acceptance criteria to give better precision.

It is also remarked here that, for experimental verifications in exposure chambers, the Jacobi room model parameters could be far

away from their realistic values in real-life situations. Under such exposure-chamber conditions, it would be difficult to find solutions unless we could successfully develop some optimization procedures in finding the solutions. Before that, we would have to make sure that the Jacobi room model parameters in the exposure chambers should be realistic in order to make meaningful experimental verifications.

In addition to the determination of f_p for the case of radon, the current method can also be used for simultaneous determinations of F and f_p for thoron, since the same set of Jacobi room model parameters are controlling the concentrations of individual radon and thoron progenies.

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