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LONG-TERM DETERMINATION OF AIRBORNE RADON PROGENY CONCENTRATIONS USING LR 115 DETECTORS AND THE EFFECTS OF THORON

V. W. Y. Choi[1], C. K. M. Ng[1], R. K. K. Lam[1], M. Janik[2], A. Sorimachi[2], C Kranrod[2], D. Nikezic[1,3], S. Tokonami[2] and K. N. Yu[1,*]
[1]Department of Physics and Materials Science, City University of Hong Kong, Tat Chee Avenue, Kowloon Tong, Kowloon, Hong Kong
[2]Environmental Radiation Effects Research Group, National Institute of Radiological Sciences, 4-9-1 Anagawa, Inage-ku, Chiba-shi 263-8555, Japan
[3]Faculty of Science, University of Kragujevac, Kragujevac, Serbia

*Corresponding author: peter.yu@cityu.edu.hk

The 'proxy equilibrium factor' (F_p) method has been developed for long-term determination of airborne radon progeny concentrations using LR 115 solid-state nuclear track detectors. In this paper, the effects of ^{220}Rn on the F_p method have been studied. The correction to the track density was related to a parameter α which was the ratio of the sum of activity concentrations of alpha-particle emitting radionuclides in the ^{220}Rn decay chain to the activity concentration of ^{220}Rn alone. Under commonly encountered circumstances, α could not be smaller than 2. An attempt was made to verify this using the exposure chamber at the National Institute of Radiological Sciences (NIRS), Chiba, Japan. A most interesting observation of $\alpha < 2$ for very high ^{220}Rn concentrations and very low equilibrium factors for ^{220}Rn in the exposure chambers was made. A possible explanation was the substantial deposition of ^{216}Po under the extreme conditions inside the exposure chambers.

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 the radon (^{222}Rn) gas itself. Accordingly, long-term measurements of the concentrations of radon progeny or the equilibrium factor F, among other information such as the size distribution of radon progeny and the unattached fraction f_p of the potential alpha energy concentration, is needed to accurately assess the health hazards contributed by radon progeny.

The equilibrium factor F between radon and its progeny is defined as $F=0.105f_1+0.515f_2+0.380f_3$ where f_i is the ratio of the activity concentration C_i of the i-th radon progeny to the activity concentration C_0 of ^{222}Rn. Here, $i=1$ stands for ^{218}Po, $i=2$ for ^{214}Pb and $i=3$ for ^{214}Bi (or ^{214}Po, because ^{214}Bi and ^{214}Po are in secular equilibrium). The exposure to radon progeny can be expressed in the traditional unit *working level month* and then multiplied by the dose conversion factor by assuming a given aerosol size distribution to give the effective dose.

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[1, 2]. This problem cannot be solved through active measurements based on air filtering, since they only give short-term determinations. Methods based on long-term measurements of radon progeny concentrations or F using solid-sate nuclear track detectors (SSNTDs) have been reviewed[3–5]. Our group has previously proposed a feasible method for long-term measurements of F through the so-called proxy equilibrium factor (F_p)[5, 6].

On the other hand, thoron (^{220}Rn) and its progeny are also present in the ambient environment, which can affect the F_p method. In the present paper, the effects of the presence of ^{220}Rn on the F_p method have been reported.

PROXY EQUILIBRIUM FACTOR (F_P) METHOD

The use of bare LR 115 SSNTD has been proposed to determine the airborne ^{218}Po+^{214}Po concentration and showed that this gave good estimates of F for radon progeny[5, 6]. The LR 115 SSNTD has an upper energy threshold for track formation, which is well below the energy of alpha particles emitted by the radon progeny plateout on the detector, i.e. plateout progeny are not detected by LR 115 SSNTDs.

The responses of the bare LR 115 detector to ^{222}Rn, ^{218}Po and ^{214}Po are expressed by the partial sensitivities ε_i of the detector to these species (i.e. the number of tracks per unit area per unit exposure) with the unit $(m^{-2})/(Bqm^{-3}\ s)$ or just (m). The partial sensitivities ε_i were found to be the same for ^{222}Rn, ^{218}Po and ^{214}Po[5,6]. It is remarked here that the equality of partial sensitivities arises because of the presence of the upper energy threshold for recording alpha-particle tracks in the LR 115 SSNTD. With equal partial sensitivities, the total track density ρ (in track m^{-2}) on the detector due to airborne ^{222}Rn, ^{218}Po and ^{214}Po is given by $\rho = \varepsilon_i(C_0 + C_1 + C_3) \times t$, where t is the exposure time. F_p was defined as[5, 6]

$$F_p = f_1 + f_3 = \frac{\rho}{\varepsilon_i C_0 t} - 1 \qquad (1)$$

By using the Jacobi room model[7], and by randomly varying the associated parameters, viz. ventilation rate λ_v, aerosol attachment rate λ_a, deposition rate of unattached progeny λ_d^u and the deposition rate of attached progeny λ_d^a, in the ranges given in Table 1, F was plotted with F_p as shown in Figure 1 and a good correlation was observed between F and F_p[5].

EFFECTS OF THORON

The presence of ^{220}Rn gas and its alpha-particle emitting progeny in the environment will contribute tracks to the bare LR 115 detector, which will lead to erroneous estimates of F_p and hence F. Corrections should be performed to account for the number of tracks on the detector due to ^{220}Rn and its progeny. If the ^{220}Rn gas has an activity concentration C_{Tn}, and αC_{Tn} is the sum of activity concentrations of alpha-particle emitting radionuclides in the ^{220}Rn decay chain, the previous equation, $\rho = \varepsilon_i(C_0 + C_1 + C_3) \times t$, can be modified to

$$\rho^* = \varepsilon_i(C_0 + C_1 + C_3 + \alpha C_{Tn}) \times t \qquad (2)$$

where ρ^* is the gross track density obtained in the presence of ^{220}Rn and its progeny.

Equation (2) can be rewritten to give a correction in the track density as

$$\rho^* - \rho = \varepsilon_i \alpha C_{Tn} \times t \qquad (3)$$

If α can be determined and when C_{Tn} is measured, F_p can still be obtained from Equations (1) and (3). In an environment with the presence of both ^{222}Rn and ^{220}Rn, their activity concentrations C_0 and C_{Tn} are commonly determined using the 'twin diffusion chamber' method[8−12]. In the following, the value of α has been determined using two different approaches, viz. (1) through the Jacobi room model, and (2) through experimental calibrations, as outlined below.

Table 1. Ranges for the parameters of the Jacobi model employed for the computer simulations in ref. (3) and in the present work.

Parameter	Lower limit	Upper limit
Ventilation rate λ_v	0.2	2.1
Aerosol attachment rate λ_a	5	500
Deposition rate of unattached progeny λ_d^u	5	110
Deposition rate of attached progeny λ_d^a	0.05	1.1

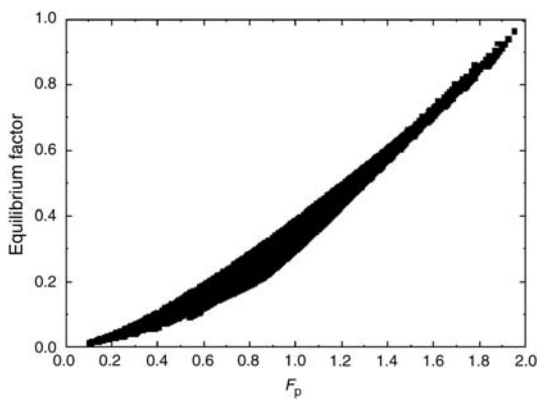

Figure 1. The relationship between the equilibrium factor F and the proxy-equilibrium factor F_p $(= f_1 + f_3)$[5].

Jacobi room model

Again by using the Jacobi room model[7], and by randomly varying λ_v, λ_a, λ_d^u and λ_d^a in the ranges given in Table 1, the distribution of α is found as shown in Figure 2, which gives an average ($\pm$SD)=2.035 $\pm$ 0.020. From Figure 2, it is shown that under commonly encountered situations generated using typical Jacobi room model parameters, α cannot be smaller than 2 and has a very narrow range. These are expected since the first ^{220}Rn progeny, ^{216}Po, has a very short half-life (0.15 s) and is usually deemed to be in radioactive equilibrium with the parent ^{220}Rn gas.

Both ^{220}Rn and ^{216}Po are alpha-particle emitting radionuclides. If the other alpha-emitting progeny of ^{220}Rn have comparatively negligible activity concentrations, which is actually the case, α should be larger than but close to 2. In practice, the assumption of α=2 should be sufficient for correction purposes.

RESULTS

The F_p method and its correction for ^{220}Rn were verified using the exposure chambers at the National

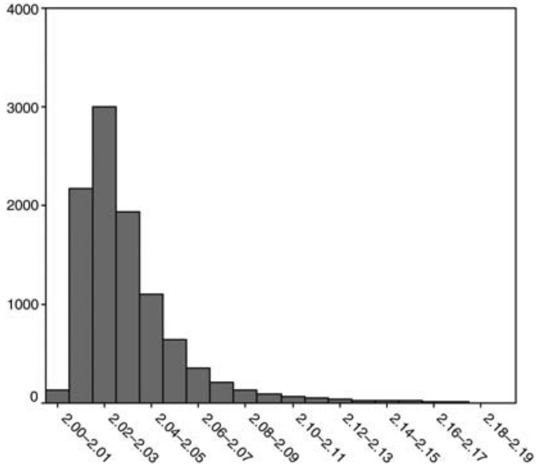

Figure 2. The distribution of α found using the Jacobi room model and by randomly varying λ_v, λ_a, λ_d^u and λ_d^a in the ranges given in Table 1 (total number of histories$=10^4$).

Institute of Radiological Sciences (NIRS), Chiba, Japan. For the verifications, three different radon exposure conditions were used. All involved $C_0=5$ kBq m^{-3}, $t=20$ h, temperature$=20$°C and relatively humidity$=60$ %, with three different F values as 0.073, 0.178 and 0.454. On the other hand, three different thoron exposure conditions were also employed, namely, (1) no thoron exposure, (2) $C_{Tn}=2.2$ kBq m^{-3}, equilibrium factor F_{Tn} for ^{220}Rn$=0.0011$, temperature$=33$°C and relatively humidity$=43$ % and (3) $C_{Tn}=59.5$ kBq m^{-3}, $F_{Tn}=0.0001$, temperature$=32$°C and relatively humidity$=44$ %. Both exposure conditions (2) and (3) corresponded to $t=20$ h. It is remarked that the radon and thoron exposures were separately carried out. As a result, nine combinations of radon and thoron exposures were used, which are summarised in Table 2.

The LR 115 detectors used in the present study were purchased from DOSIRAD, France. After the exposures, the SSNTDs were etched in 10 % aqueous NaOH at 60°C to achieve a removed layer in the range of 5.5–7 μm. The detectors were etched using a magnetic stirrer to provide uniform etching[13], and the removed layer was continually monitored using infrared absorption[14]. The partial sensitivities ϵ_i were calculated from the removed layer x from[2]

$$\varepsilon = -0.00547 + 0.00145x \qquad (4)$$

The measured removed layers, calculated ε_i, measured ρ^* (in the presence of thoron) or ρ (for no thoron), F_p (for no thoron) and F_p^c (corrected F_p using $\alpha=2$ in the presence of thoron), and the derived F values for the LR 115 SSNTDs exposed to different combinations of radon and thoron exposures are shown in Table 3.

Table 2. Codes for different combinations of radon and thoron exposures ($t=20$ h), to be used in Table 3.

Radon	Thoron: Absent	Thoron: 2.2 kBq m^{-3} $F_{Tn}=0.0011$	Thoron: 59.5 kBq m^{-3} $F_{Tn}=0.0001$
5 kBq m^{-3} $F=0.073$	LN	LL	LH
5 kBq m^{-3} $F=0.178$	MN	ML	MH
5 kBq m^{-3} $F=0.454$	HN	HL	HH

Table 3. The values of x, ε_i, ρ^* or ρ, F_p or F_p^c, and F for LR 115 SSNTDs exposed to different combinations of radon and thoron exposures (please refer to Table 2 for the codes).

Code	x (μm)	ε_i (10^{-3} m)	ρ^* or ρ (10^6 m^{-2})	F_p or F_p^c	F
LN	5.54	2.56	1.26	0.37	0.075 ± 0.025
MN	6.76	4.33	2.34	0.50	0.14 ± 0.04
HN	5.78	2.91	2.33	1.22	0.47 ± 0.05
LL	5.96	3.17	2.40	0.22	0.025 ± 0.015
ML	5.76	2.88	2.32	0.36	0.09 ± 0.02
HL	5.65	2.72	2.83	1.01	0.35 ± 0.05
LH	5.57	2.61	18.9	−4.6	–
MH	5.77	2.90	17.0	−8.5	–
HH	6.42	3.84	25.8	−6.1	–

The F values obtained by the F_p method (0.075, 0.14, 0.47) were very close to the actual values (0.073, 0.178, 0.454) for no thoron exposure. For the relatively low ^{220}Rn concentration of 2.2 kBq m^{-3} and a $F_{Tn}=0.0011$, the corrections appeared to be acceptable by taking $\alpha=2$, although the results show signs of over-correction. In contrast, for a very high ^{220}Rn concentration of 59.5 kBq m^{-3} and a very low $F_{Tn}=0.0001$, corrections with $\alpha=2$ were not valid. Feasible values of F_p^c could only be obtained when $\alpha < 2$.

As discussed earlier, in commonly encountered situations, α cannot be smaller than 2. However, under these normal circumstances, F_{Tn} cannot be smaller than 0.01 (also from the Jacobi model). To generate the very low F_{Tn} values of 0.0011 and 0.0001 inside the thoron exposure chambers, extreme values are needed for the parameters in the Jacobi room model. A sample relationship between α and F_{Tn} found using the Jacobi room model and extreme values of the parameters is shown in Figure 3, and it can be seen that $\alpha < 2$ is actually possible for $F_{Tn} < 0.01$. Incidentally, if $\alpha=1.8$ is taken to make corrections for the detectors LL, ML and HL, the F_p^c values will become 0.30, 0.44 and 1.09, respectively,

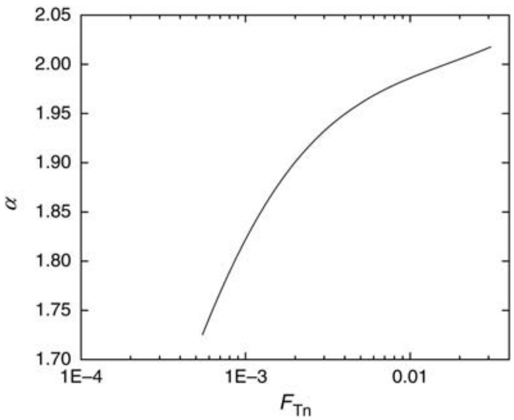

Figure 3. The relationship between α and F_{Tn} found using the Jacobi room model and extreme values of the parameters.

and the derived F values will become 0.050 ± 0.025, 0.10 ± 0.02 and 0.40 ± 0.05, respectively. These are closer to the true values of 0.073, 0.178 and 0.454.

CONCLUSIONS

The proxy equilibrium factor F_p method has previously been proposed for long-term measurements of the equilibrium factor F between radon and its progeny. In the present paper, the effects of ^{220}Rn have been studied on the F_p method. The correction to the track density is related to a parameter α. Under normal situations, α cannot be smaller than 2 and has a very narrow range. This is expected since ^{220}Rn is usually deemed to be in radioactive equilibrium with its first progeny ^{216}Po. In practice, $\alpha=2$ should be sufficient for general correction purposes. However, this assumption does not hold for very high ^{220}Rn concentrations and very low F_{Tn} values, which can be encountered inside exposure chambers. A possible explanation is the substantial deposition of ^{216}Po under these extreme conditions.

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