Investigation of Radon and Thoron Concentrations in a Landmark Skyscraper in Tokyo

Kazumasa Inoue¹, Masahiro Hosoda², Shinji Tokonami³ Tetsuo Ishikawa⁴ and Masahiro Fukushi^{1,*}

¹ Graduate School of Human Health Sciences, Tokyo Metropolitan University
 7-2-10 Higashiogu, Arakawa-ku, Tokyo 116-8551, Japan
 ² Hirosaki University Graduate School of Health Sciences
 66-1 Honcho, Hirosaki, Aomori 036-8564, Japan
 ³ Institute of Radiation Emergency Medicine, Hirosaki University
 66-1 Honcho, Hirosaki, Aomori 036-8564, Japan
 ⁴ Research Center for Radiation Protection, National Institute of Radiological Sciences
 4-9-1 Anagawa, Inage-ku, Chiba 263-8555, Japan

* Correspondence to: Masahiro Fukushi, Ph.D. Tokyo Metropolitan University
7-2-10 Higashiogu, Arakawa-ku, Tokyo 116-8551, Japan Tel: +81-3-3819-1211, Fax: +81-3-3819-1406 E-mail: fukushi@hs.tmu.ac.jp

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ABSTRACT

The temporal variation of the radon concentration, and the radon and thoron concentrations every three months for a year were measured using two types of devices in a landmark skyscraper, the Tokyo Metropolitan Government Daiichi Building. In the measurement of temporal variation of the radon concentration using a pulse type ionization chamber, the average radon concentration was 21 ± 13 Bq m⁻³ (2 - 68 Bq m⁻³). The measured indoor radon concentration had a strong relationship with the operation of the mechanical ventilation system and the activities of the office workers. The radon concentration also increased together with temperature. Other environmental parameters, such as air pressure and relative humidity, were not related to the radon concentration. In the long-term measurements using a passive radon and thoron discriminative monitor, no seasonal variation was observed. The annual average concentrations of radon and thoron were $16 \pm$ 8 Bq m⁻³ and 16 \pm 7 Bq m⁻³, respectively. There was also no relationship between the two concentrations. The annual average effective dose for office workers in this skyscraper was estimated to be 0.08 mSv y⁻¹ for 2000 working hours per year. When considering the indoor radon exposure received from their residential dwellings using the annual mean radon concentration indoors in Japan (15.5 Bq m⁻³), the annual average effective dose was estimated to be 0.37 mSv y⁻¹. This value was 31 % of the worldwide average annual effective dose.

Keywords: Radon concentration; Thoron concentration; Pulse type ionization chamber; Passive monitor; Annual effective dose; Skyscraper

INTRODUCTION

Internal exposure from the radioactive noble gases radon-222 (radon) and radon-220 (thoron) is the second leading cause of lung cancer after smoking in many countries [1]. It has been previously reported that the relative risk of lung cancer increases 16 % per 100 Bq m⁻³ radon concentration [2]. In addition, according to the UNSCEAR [3], the worldwide average annual effective dose due to inhalation of radon, thoron and their decay products is estimated to be 1.2 mSv. Since the worldwide average annual effective dose due to natural radiation is 2.4 mSv, the effect from radon, thoron and their decay products for half of this value.

In dwellings and office buildings, the generation sources of radon and thoron are uranium and radium that are contained within the concrete and floor covers [4]. Especially, in densely populated metropolitan areas of Japan such as Tokyo, the architectural style has been changed from traditional wooden construction to steel construction that uses reinforced concrete containing radon and thoron generation sources. In addition, radon remains at high concentrations in closed environments [1]. Thus, airtight places, such as skyscrapers, and places that lack natural ventilation, such as underground facilities, tend to have increased radon concentration compared to wooden buildings [5-6]. In fact, while the average indoor radon concentration in Japan is 15.5 Bq m⁻³ [7], the mean radon concentration of several skyscrapers was found to be 36.8 Bq m⁻³ [8]. Furthermore, high radon concentrations of 18.9 Bq m⁻³ - 350 Bq m⁻³ have been observed in subway stations that were built using the same ferroconcrete as that in skyscrapers [9-11]. Some studies using recent models of continuous radon monitors have presented the relationship between radon concentration in skyscrapers and underground facilities are lacking. In particular, reports about the radon concentration at higher floors are very valuable.

In past radon measurement studies, thoron was not measured, and its presence was ignored; this is because the measurement of thoron itself is difficult [14]. However, it is necessary to consider the presence of thoron in radon measurement to precisely estimate the radon risk, and the importance of this has come to be recognized in recent reports [14-16]. In the past, radon measurement studies widely utilized passive monitors, and up to 1.8-fold overestimation of the radon concentration was found due to the effect from thoron [17]. To resolve this issue, passive radon and thoron discriminative monitors have been developed by Tokonami [14], and Zhuo et al. [15]. Zhuo et al. reported that the measured thoron concentration at a low-rise apartment house using their monitor was 168±213 Bq m⁻³; this value is high compared to that of radon as described above. Therefore, it may lead to misunderstanding the radon risk in indoor environments with high

thoron concentrations. Although there have been a few reports using passive radon and thoron discriminative monitors, the number of studies is still not enough. Especially, studies that measured the thoron concentration at higher floors are lacking.

In this study, radon and thoron measurements were carried out in a landmark skyscraper located in Tokyo; temporal variation of the radon concentration was obtained using a pulse type ionization chamber, and long-term measurement for one year were done for the radon and thoron concentrations using a passive radon and thoron discriminative monitor. The annual averaged effective dose due to inhalation of radon was also estimated.

MATERIALS AND METHODS

<u>Radon and thoron measurements</u>: The measurements of radon and thoron concentrations were carried out at the Tokyo Metropolitan Government Daiichi Building which has 51 stories above and below ground. The structure height is 243 m; the site area and total floor area are 14,350 m² and 195,567 m², respectively. The windows of the building cannot be opened and closed.

To measure temporal variation of the radon concentration, a pulse type ionization chamber (AlphaGUARD[®] PQ2000 PRO, SAPHYMO GmbH, Germany) was placed on the eleventh floor. The installation location was surrounded by a steel shelf. The pulse type ionization chamber was set to the diffusion mode with a 60 min time interval. Simultaneously, temperature, air pressure and relative humidity were measured. The measurement period was set to six weeks between 6 July and 16 August.

For the long-term radon and thoron measurements, 76 passive radon and thoron discriminative monitors (RADPOT[®]) [15] were placed as shown in Table 1. The RADPOT consists of a radon monitor and a thoron monitor (Figure 1). Each monitor has a different structure; both radon and thoron are detected in the thoron monitor which has four holes. On the other hand, the radon monitor has no holes and it detects only radon because the half-life of thoron is shorter than that of radon. This structural difference produces different ventilation rates, and radon and thoron can be discriminated. The allyl diglycol carbonate, CR-39 is placed inside each monitor to detect alpha rays from radon, thoron and their decay products.

The measurement periods were set to 3 months for a one year period (October – September). Three months after exposure in the monitor, the CR-39 was chemically etched for 6 h in a sodium hydroxide solution at 90 °C. The number of alpha tracks was counted using an optical microscope. The respective lower limits of detection (LLDs) of radon and thoron for this monitor are reported to be 3.5 Bq m⁻³ and 13 Bq m⁻³ for a 90 day period [15-16].

Estimation of annual effective dose: The results obtained from the RADPOTs were used to estimate an annual average effective dose (H) by assuming 8 working hours per day and 250 working days per year in the skyscraper. The estimation method was recommended by the UNSCEAR 2000 report [3].

$$H = F \cdot D \cdot Q \cdot T \qquad (1)$$

Here *F* is equilibrium factor (0.4), *D* is effective dose coefficient (9 × 10⁻⁶ mSv Bq⁻¹ h⁻¹ m⁻³), *Q* is annual mean radon concentration (Bq m⁻³), *T* is working hours (2000 hours). In addition, the annual effective dose in consideration of the exposure dose outside the office was calculated by:

$$H = F \cdot D(Q_1 \cdot T_1 + Q_2 \cdot T_2) \tag{2}$$

where Q_1 is annual average radon concentration in the skyscraper, Q_2 is annual mean radon concentration indoors in Japan (15.5 Bq m⁻³) [7], T_1 is occupancy factor (0.8) × annual working hours (2000 hours), T_2 is occupancy factor (0.8) × [8760 hours per year – annual working hours (2000 hours)]. In this study, to estimate annual average effective dose during working hours in the skyscraper, the annual mean radon concentrations (Q and Q_1) were corrected using the ratio of the mean radon concentration over the whole day to the mean radon concentration during working hours obtained from the pulse type ionization chamber. In addition, the exposure outdoors was not considered because these office workers always used the subway for commuting to and from work.

RESULTS AND DISCUSSION

Figure 2 shows obtained temporal variations of the radon concentration, temperature, air pressure and relative humidity. The average values (range) of the radon concentration, temperature, air pressure and relative humidity were 21 ± 13 Bq m⁻³ (2 - 68 Bq m⁻³), 26.2 ± 1.3 °C (24.5 - 28.0), 998 ± 5 hPa (977 - 1010 hPa), 63 % (56 - 70 %), respectively. The radon concentration rose from midnight to dawn, and then, decreased between dawn and noon. The average radon concentration during working hours (8:00 - 18:00, Monday to Friday), outside working hours (18:00 - 8:00, Monday to Friday), weekends (Saturday and Sunday) and one holiday were 15 ± 10 Bq m⁻³, 18 ± 11 Bq m⁻³ and 30 ± 14 Bq m⁻³, respectively (Figure 3). From these results, the radon concentration had the lowest value during working hours (8:00) and automatically switched off at 18:00. The maximum radon concentration during the measurement period was 68 Bq m⁻³ at 10:00 on 21 July, a holiday, and 63 hours after the air conditioning was stopped. Thus, the radon concentration depended on the operation of the mechanical ventilation system and the activities of the office

workers. The same tendency has also been reported from studies in other countries [5, 13]. In addition, the relationship between the radon concentration and operational status of the air conditioning system that was measured in the same Tokyo metropolitan area has been suggested in a previous report [12]. Regarding the variation of temperature, the temperature increased at weekends because this study was carried out during the summer, and it exhibited the same tendency compared to the radon concentration. However, there was no relationship between radon concentration and other environmental parameters.

The measured annual average radon concentration (range) for the Tokyo Metropolitan Government Daiichi Building was 16 ± 8 Bq m⁻³ (<LLD-55 Bq m⁻³). The reported annual mean radon concentration (range) of another high-rise building located in metropolitan Tokyo was 32 Bq m⁻³ (20 - 66 Bq m⁻³) [8], which was twice the present measured annual average radon concentration. An indoor mean radon concentration in Japan has been reported as 15.5 Bq m⁻³ [7]; this was the same as the presently obtained value.

The quarterly radon concentrations obtained from the RADPOT measurements are shown in Table 2. They were 13 ± 10 Bq m⁻³ (October – December), 16 ± 7 Bq m⁻³ (January – March), 16 ± 9 Bq m⁻³ (April – June), 19 ± 8 Bq m⁻³ (July – September); no seasonal variation was seen. The skyscraper for the present study was not susceptible to changes in outdoor weather because it has been kept highly airtight. Therefore, no seasonal variation was measured, just as in other skyscrapers that have automatically controlled ventilation by air conditioners [12, 16, 18].

The annual average radon concentrations of each floor are shown in Figure 3. In the aboveground floors, the radon concentration showed an upward tendency at the middle-tier level. In a previous study, it has strongly suggested that the radon concentration was correlated with the air ventilation rate [19]. Although the air ventilation rate was not measured in this study, the obtained result might be reflecting the air ventilation rate. On other hand, the radon concentration in the underground floors showed an upward tendency going to the lowest floor. These underground floors were parking areas. Since the air ventilation rate was lower than in the aboveground floors, radon gas might be retained for the underground floors.

The quarterly average thoron concentrations are listed in Table 3. These quarterly average values were calculated, excluding three data that had an issue with the monitor installation places. On the other hand, all measured data were used to obtain the ranges shown in Table 3. In this study, no seasonal variation was exhibited, which was the same finding as for the radon concentration. The annual average thoron concentration in this skyscraper was 16 ± 7 Bq m⁻³ (<LLD - 590 Bq m⁻³), and was lower compared to a previous report (22 ± 18 Bq m⁻³) [14]. Figure 4

shows the annual average thoron concentration of each floor (12 data were not used). No significant trend for the annual average thoron concentration of each floor was exhibited. But, in the first aboveground floor, the maximum measured thoron concentration was 590 Bq m⁻³. The RADPOTs were placed near the wall made of marble and concrete. The thoron concentration depends more significantly on distance from the wall compared to radon [6, 14]. Thus, the obtained data might be affected by the installation place. In this study, the average values have been calculated with the exception of these 12 data.

The ratios of thoron to radon (222 Rn/ 220 Rn) are summarized in Table 4. Annual average 222 Rn/ 220 Rn ratio (range) was 1.4 ± 1.4 (0.2 – 148.0). This value was the same level as that in a previous report for measurements in Japan [16]. The plot of radon versus thoron concentrations is shown in Figure 5. Since the coefficient of correlation was 0.0086, there was no relationship between radon and thoron concentrations. Thus, it was clear that thoron concentrations cannot be simply estimated from the radon concentrations.

The annual average effective dose is shown in Table 5. The annual average effective dose for this skyscraper was 0.08 mSv y⁻¹. The present estimated dose was half the value reported for other skyscrapers in the same metropolitan Tokyo area (0.18 mSv y⁻¹) [12]. In addition, the annual average effective dose that included the dose outside working hours was 0.37 mSv y⁻¹. This value was 82 % of previous reports [16, 20]. In addition, this value was only 31 % of the worldwide average annual effective dose [3].

CONCLUSION

In this study, the measurement of radon and thoron concentrations were carried out in a landmark skyscraper located in Tokyo; the temporal variation of radon was obtained with a pulse type ionization chamber and the long-term measurements of radon and thoron were made with a passive radon and thoron discriminative monitor. In the measurement of temporal variation, the radon concentration was found to depend on operational status of the air conditioners and the activities of the office workers. The radon concentration was correlated with temperature, but not with other environmental parameters. In the long-term measurement using a passive radon and thoron discriminative monitor, the annual average concentrations of radon and thoron were 16 ± 8 Bq m⁻³ and 16 ± 7 Bq m⁻³, respectively. No seasonal variation was exhibited. In addition, there was no relationship between radon and thoron concentrations. The annual average effective dose for this skyscraper was calculated from the measured concentration and was 0.08 mSv y⁻¹. When considering dose outside working hours, the annual average effective dose was estimated to be

 0.37 mSv y^{-1} .

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Figure 1: Diagram of a passive radon and thoron discriminative monitor.



Figure 2

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<u>Figure 2</u>: Temporal variations of radon concentration, temperature, air pressure and relative humidity. The data were measured using a pulse type ionization chamber for 6 weeks with 60 min time interval.





Figure 3: Temporal variations of radon concentrations during weekdays (a), and on weekends and one holiday (b).

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<u>Figure 4</u>: Annual average radon concentrations at each floor. Data were measured using a passive radon and thoron discriminative monitor for every 3 months for a year.

Figure 5 Inoue et al.



<u>Figure 5</u>: Annual average thoron concentrations at each floor. Twelve data were not used for the mean value. Data were measured with radon concentration using a passive radon and thoron discriminative monitor every 3 months for a year.





Figure 6: Correlation between radon and thoron concentrations

Floor level	Number of monitors	
B3F	7	
B2F	7	
B1F	7	
1F	6	
3F	5	
11F	8	
15F	8	
20F	8	
30F	8	
32F	6	
45F	4	

Table 1. The number of passive radon and thoron discriminative monitors installed

Floor	Oct. – Dec.	Jan. – Mar.	Apr. – Jun.	Jul. – Sep.
level	Mean $\pm \sigma$ (range)	Mean $\pm \sigma$ (range)	Mean $\pm \sigma$ (range)	Mean $\pm \sigma$ (range)
B3F	27 ± 16 (12-55)	23 ± 4 (19-31)	23 ± 12 (17-50)	26 ± 10 (13-41)
B2F	20 ± 12 (6-37)	15 ± 5 (6-22)	16 ± 8 (7-31)	14 ± 4 (9-20)
B1F	$9 \pm 4 (< LLD^*-15)$	$9 \pm 6 (< LLD^*-20)$	$13 \pm 9 (< LLD^*-25)$	11 ± 4 (5-17)
1F	$6 \pm 5 (< LLD^*-16)$	$12 \pm 12 (< LLD^*-35)$	$10 \pm 4 \ (5-16)$	$11 \pm 6 (< LLD^*-21)$
3F	12 ± 8 (6-25)	18 ± 7 (10-27)	15 ± 5 (8-20)	23 ± 9 (15-37)
11F	$9 \pm 6 (< LLD^*-17)$	16 ± 7 (12-32)	11 ± 3 (7-15)	23 ± 6 (11-31)
15F	$8 \pm 4 (< LLD^*-17)$	21 ± 4 (16-28)	20 ± 6 (13-31)	22 ± 7 (12-31)
20F	18 ± 7 (7-27)	$20 \pm 4 (12-26)$	18 ± 6 (11-26)	22 ± 4 (18-29)
30F	14 ± 4 (8-18)	15 ± 2 (12-16)	15 ± 5 (9-24)	22 ± 4 (15-27)
32F	13 ± 5 (6-21)	15 ± 4 (9-19)	13 ± 3 (8-17)	16 ± 3 (11-20)
45F	8 ± 3 (5-12)	16 ± 5 (12-23)	11 ± 4 (8-16)	13 ± 2 (11-14)
Mean $\pm \sigma$	$13 \pm 10 (< \text{LLD}^*-55)$	$16 \pm 7 (< LLD^*-35)$	$16 \pm 9 (< LLD^*-50)$	$19 \pm 8 (< LLD^*-41)$

Table 2. Average radon concentrations (Bq m^{-3}) for each floor level

^{*} LLD, lower limit of detection (3.5 Bq m⁻³) [15]

Floor	Oct. – Dec.	Jan. – Mar.	Apr. – Jun.	Jul. – Sep.
level	Mean $\pm \sigma$ (range)	Mean $\pm \sigma$ (range)	Mean $\pm \sigma$ (range)	Mean $\pm \sigma$ (range)
B3F	$22 \pm 17 (< LLD^*-57)$	$17 \pm 7 (< LLD^*-28)$	$30 \pm 22 (< LLD^*-63)$	$15 \pm 18 (< LLD^*-64)$
B2F	$16 \pm 7 (< LLD^*-31)$	$15 \pm 4 (< LLD^*-25)$	$35 \pm 17 (< LLD^*-61)$	$19 \pm 11 (< LLD^*-39)$
B1F	$20 \pm 7 (< LLD^*-51)$	$16 \pm 8 (< LLD^*-31)$	$12 \pm 3 (< LLD^*-15)$	$14 \pm 3 (< LLD^*-21)$
1F	$25 \pm 20^{a} (< LLD^{*}-590)$	< LDD ^{*, a} (< LLD [*] -300)	$17 \pm 6^{a} (14-490)$	$19 \pm 10^{a} (< LLD^{*}-518)$
3F	$15 \pm 2 (< LLD^*-17)$	$13 \pm 2 (< LLD^*-15)$	18 ± 8 (< LLD [*] -31)	$20 \pm 5 (< LLD^*-26)$
11F	$13 \pm 1 (< LLD^*-15)$	$13 \pm 5 (< LLD^*-23)$	$13 \pm 3 (< LLD^*-15)$	$15 \pm 6 (< LLD^*-30)$
15F	$18 \pm 7 (< LLD^*-34)$	$13 \pm 3 (< LLD^*-19)$	$13 \pm 1 (< LLD^*-15)$	$16 \pm 5 (< LLD^*-25)$
20F	$18 \pm 9 (< LLD^*-35)$	$15 \pm 6 (< LLD^*-29)$	$18 \pm 9 (< LLD^*-31)$	$19 \pm 9 (< LLD^*-40)$
30F	$20 \pm 11 (< LLD^*-41)$	$13 \pm 2 (< LLD^*-18)$	$20 \pm 11 (< LLD^*-41)$	$13 \pm 2 (< LLD^*-17)$
32F	$14 \pm 2 (< LLD^*-18)$	$14 \pm 3 (< LLD^*-18)$	$13 \pm 4 (< LLD^*-20)$	$15 \pm 4 (< LLD^*-21)$
45F	$15 \pm 2 (< LLD^*-16)$	$13 \pm 2 (< LLD^*-15)$	$12 \pm 3 (< LLD^*-16)$	$12 \pm 3 (< LLD^*-15)$
Mean $\pm \sigma$	$17 \pm 8^{a} (< LLD^{*}-590)$	$14 \pm 5^{a} (< LLD^{*}-300)$	$16 \pm 9^{a} (< LLD^{*}-490)$	$16 \pm 6^{a} (< LLD^{*}-518)$

Table 3. Quarterly average thoron concentrations (Bq $m^{\mbox{-}3}$) for each floor level

^{*} LLD, lower limit of detection (13 Bq m⁻³) [15]

^a Three data were not used

Floor level	Oct. – Dec.	Jan. – Mar.	Apr. – Jun.	Jul. – Sep.	Annual mean
	Mean $\pm \sigma$ (range)	Mean $\pm \sigma$ (range)	Mean $\pm \sigma$ (range)	Mean $\pm \sigma$ (range)	Mean $\pm \sigma$ (range)
B3F	$1.2 \pm 1.2 \ (0.2-3.8)$	0.8 ± 0.3 (0.4-1.2)	$1.0 \pm 0.6 \ (0.3-1.8)$	$1.0 \pm 0.6 \ (0.4-1.7)$	$1.0 \pm 0.7 \ (0.2 - 3.8)$
B2F	$1.5 \pm 1.7 \ (0.4-5.2)$	1.3 ± 1.3 (0.6-4.2)	$2.4 \pm 1.2 \ (0.7-4.3)$	$1.3 \pm 0.6 (0.7-2.2)$	1.6 ± 1.3 (0.4-5.2)
B1F	$2.5 \pm 1.9 (1.0-6.3)$	$2.7 \pm 2.5 \ (0.7-4.0)$	$1.4 \pm 1.0 \ (0.5-3.3)$	$1.5 \pm 0.6 \ (0.9-2.6)$	$2.0 \pm 1.7 \ (0.5-6.3)$
1F	$3.5 \pm 5.3^{a} (3.7-124.9)$	$0.8 \pm 1.0^{a} (0.4-37.5)$	$1.2 \pm 1.2^{a} (1.1-37.7)$	$1.0 \pm 1.0^{a} (0.6-148.0)$	$1.6 \pm 2.7^{\rm b} \ (0.4-124.9)$
3F	$1.5 \pm 0.7 \ (0.5 - 2.3)$	$0.7 \pm 0.2 \ (0.5 \text{-} 1.0)$	$1.3 \pm 0.8 \ (0.7-2.4)$	$1.0 \pm 1.9 \ (0.4-1.7)$	$1.1 \pm 0.6 \ (0.5-2.4)$
11F	$2.2 \pm 1.4 \ (0.8-3.7)$	$0.9 \pm 0.7 \ (0.4-2.6)$	$1.0 \pm 0.3 \ (0.7-1.1)$	$0.8 \pm 0.8 \ (0.4-2.7)$	$1.2 \pm 1.0 \ (0.4-2.7)$
15F	$3.3 \pm 3.2 \ (0.8-9.7)$	$0.7 \pm 0.2 \ (0.4-1.0)$	$0.7 \pm 0.2 \ (0.4-1.0)$	$0.8 \pm 0.5 \ (0.4-2.1)$	1.3 ± 1.9 (0.4-9.7)
20F	$1.2 \pm 0.7 \ (0.5 - 2.6)$	$0.8 \pm 0.5 \ (0.5 \text{-} 1.7)$	$1.2 \pm 0.8 \ (0.4-2.7)$	$0.9 \pm 0.4 \ (0.4-1.7)$	$1.0 \pm 0.6 \ (0.4-2.7)$
30F	$1.7 \pm 1.2 \ (0.7-3.9)$	$0.9 \pm 0.7 \ (0.5 - 1.3)$	$1.4 \pm 0.9 \ (0.7-3.6)$	$0.6 \pm 0.2 \ (0.5 - 1.0)$	1.1 ± 0.8 (0.5-3.9)
32F	$1.2 \pm 0.5 \ (0.6-2.2)$	$1.0 \pm 0.4 \ (0.7-1.8)$	$1.0 \pm 0.3 \ (0.7-1.5)$	$1.1 \pm 0.5 \ (0.6-1.9)$	1.1 ± 0.4 (0.6-2.2)
45F	$2.0 \pm 0.9 (1.1-3.2)$	$0.8 \pm 0.3 \ (0.6-1.1)$	$1.2 \pm 0.5 \ (0.8-2.0)$	$0.9 \pm 0.3 \ (0.6-1.2)$	$1.2 \pm 0.7 \ (0.6-3.2)$
Mean $\pm \sigma$	$2.1 \pm 2.2^{a} (0.2-124.9)$	$1.1 \pm 1.1^{a} (0.4-37.5)$	$1.3 \pm 0.9^{a} (0.3-37.7)$	$1.0 \pm 0.6^{a} (0.4-148.0)$	$1.4 \pm 1.4^{\rm b} (0.2-148.0)$

Table 4. Average abundance ratios of $^{\rm 220}Rn/^{\rm 222}Rn$ for each floor level

^a Three data were not used

^b Twelve data were not used

Floor level	Annual effective dose	
	(mSv)	
B3F	0.14	
B2F	0.08	
B1F	0.05	
1F	0.05	
3F	0.09	
11F	0.07	
15F	0.09	
20F	0.10	
30F	0.08	
32F	0.07	
45F	0.06	
Mean	0.08	

Table 5. Annual average effective dose for each floor level