

■原著

Measurement of radon and thoron concentrations in the Tokyo Metropolitan University Arakawa Campus building

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Abstract: Smoking and radon inhalation are the primary causes of lung cancer in many countries. The world average annual dose due to radon inhalation is 1.26 mSv y^{-1} , which is more than half of the annual exposure dose from natural radiation sources, 2.40 mSv y^{-1} . In this study, radon and thoron radioactivity concentrations (hereafter referred to simply as concentrations) measurements were carried out in the Tokyo Metropolitan University Arakawa Campus building using a pulse type ionization chamber and passive radon and thoron discriminative monitors. The respective average ($\pm \sigma$) radon concentrations (Bq m^{-3}) for each day of the week from Sunday to Saturday were: 21 ± 7 , 20 ± 7 , 20 ± 8 , 22 ± 6 , 21 ± 7 , 20 ± 6 , 23 ± 7 . On week days, the radon concentration peaked daily at 8:00 am with a value of $25 \pm 6 \text{ Bq m}^{-3}$, it decreased until 7:00 pm reaching a value of $17 \pm 7 \text{ Bq m}^{-3}$, and then, showed a rising trend to the next morning's peak. Radon concentration tended to show a higher value and less fluctuation on weekends. No seasonal change was observed. No correlation was observed between radon concentration and thoron concentration. In Japan, the reported arithmetic average radon concentration indoors is 15.5 Bq m^{-3} and the arithmetic average concentration outdoors is 5.4 Bq m^{-3} . The annual effective dose of radon by inhalation in Japan is 0.64 mSv y^{-1} . The average radon concentration of reinforced concrete buildings tends to be higher, though a radon concentration survey in reinforced concrete buildings in Japan is lacking. Calculated annual average exposure dose in the campus reinforced concrete building was 0.15 mSv y^{-1} . Annual average exposure dose considering an indoor environment other than the Arakawa Campus building was 0.42 mSv y^{-1} .

Key words: Radon concentration, Thoron concentration, Pules type ionization chamber, Passive monitor, Annual effective dose

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Introduction

Inhalation of radon and smoking are the primary causes of lung cancer in many countries¹⁾. The world average annual dose due to radon inhalation is 1.26 mSv y⁻¹, which is more than half of the annual exposure dose from natural radiation source which is 2.40 mSv y^{-1,2)}. The half-life of radon (²²²Rn) is approximately 3.8 days, and it is present in every scene of the living environment. ²²²Rn is a descendant nuclide of the uranium series that is produced from radium (²²⁶Ra) contained in the materials used to form the concrete used in buildings and in soil. Thoron (²²⁰Rn), half-life 55.6 s, is an isotope of radon (²²²Rn) which is a thorium series nuclide generated mainly from materials in the soil. ²²²Rn and ²²⁰Rn are exhaled from the building materials used in walls, floors and ceilings. Both of them are noble gases and they have tendencies to stay in a sealed interior space and to accumulate.

In Japan, the arithmetic mean value of the radon concentration indoors is 15.5 Bq m⁻³ and the average concentration outdoors is 5.4 Bq m^{-3,3)}. Annual effective dose by radon inhalation in Japan is 0.64 mSv y⁻¹ which is 50.8% of the world average value⁴⁾. In general, Japanese houses are predominantly wooden structures, and due to the composition of Japanese soil and bedrock, the annual effective dose by radon inhalation is lower than that in many other countries. However, high-rise buildings are the main structures in metropolitan areas with high population densities and buildings made of reinforced concrete, which is a source of radon and thoron, are more common than conventional wooden buildings. The average radon concentration of reinforced concrete used in buildings has been reported to be as high as 36.8 Bq m^{-3,5)}. In a recent study using active type radon detectors, however, it has found that the radon concentration in reinforced concrete buildings is greatly changed by air conditioning equipment and the its pattern of use⁶⁾, hence there is a need for a survey of radon concentration in reinforced concrete building in Ja-

pan. Although the presence of thoron and its decay products has been ignored because their measurement is not easy, the importance of the measurement has been recognized in order to evaluate the radon risk accurately⁷⁾. Several surveys using radon and thoron discriminative monitors have been reported, but the number of data are not sufficient to assess radon risk.

In this study, exposure dose due to radon inhalation was calculated from measurements of radon and thoron concentrations made using a pulse type ionization chamber and passive radon and thoron discriminative monitors. The measurements were made in the reinforced concrete building of a university branch campus. The building was expected to have a higher value compared to the average indoor radon and thoron concentrations in Japan.

Materials and Methods

Radon and thoron radioactivity concentrations were measured at the Tokyo Metropolitan University Arakawa Campus. The Arakawa Campus is located in Arakawa Ward, Tokyo and has one five-story building with a reinforced concrete structure. The building windows can be opened and closed. Placement of the two kinds of measuring instruments and the measurements were carried out with the consent of the administrator of the Arakawa Campus facility. A pulse type ionization chamber (AlphaGUARD PQ2000 PRO, SAPHHYMO GmbH, Germany) was installed on the building first floor in order to observe the temporal change of the radon concentration. The instrument was operated in the diffusion mode and the radon concentration, temperature, barometric pressure and humidity were measured once every hour. Measurements were carried out for 11 weeks from March 16 (Sunday) beginning at 18:00 until May 29 (Thursday) ending at 12:00 (noon). To measure radon and thoron concentrations throughout the year, passive radon and thoron discriminative monitors (RAD-POT[®]) (Figure 1) were placed at 30

locations on different floor levels (1F to 5F). The numbers of monitors were: 1F, 12; 2F, 6; and 3F, 4F, 5F, 4 on each level. Total measurement period was one year (from October to September) and the average radon and thoron concentrations (Bq cm^{-3}) for three months were measured four times. After being installed for three months, the monitors were collected and chemically etched by sodium hydroxide at 90°C for 6 h. Then the conical etch pits were observed with an optical microscope. The reported lower detection limits (LLDs) for the monitors used in this study were 3.5 Bq m^{-3} and 13.0 Bq m^{-3} for radon and thoron concentrations, respectively. To calculate annual effective dose H_1 (mSv y^{-1}) from radon concentration obtained from passive radon and thoron discriminative monitors, the following equation was used which has been proposed in the UNSCEAR Annual Report 2000 General Assembly Annex B²⁾.

$$H_1 = F \times D \times Q \times T \quad (1)$$

Here F is the equilibrium coefficient of radon (0.4), D is the dose conversion factor ($9 \times 10^{-6} \text{ mSv Bq}^{-1} \text{ h}^{-1} \text{ m}^3$), Q is the average radon concentration in the air, and T is the average time spent in the Tokyo Metropolitan University Arakawa Campus building by workers and students. Based on the method described in the UNSCEAR Annual Report 2000, total time spent in the Arakawa Campus building was estimated as 2,000 h per year, assuming 250 work or class days per year and staying in the building for 8 hours per day. The average air radon concentration was defined as the average radon concentration in air (Bq m^{-3}) during 8 hours in the Tokyo Metropolitan University Arakawa Campus premises from 9:00 to 17:00. Also, the annual effective dose H_2 was calculated considering the exposure dose when not at the Tokyo Metropolitan University Arakawa Campus building using the following formula.

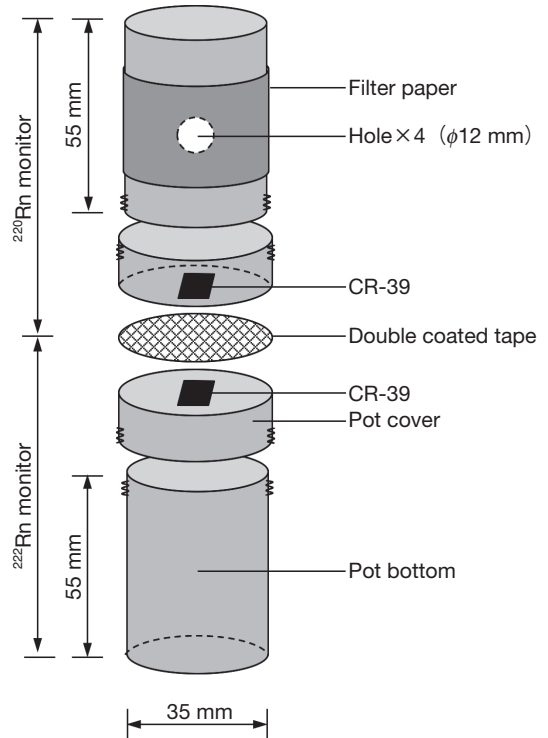


Figure 1 Structure of the passive radon and thoron discriminative monitors (RAD-POT®).

$$H_2 = F \times D(Q_1 \times T_1 + Q_2 \times T_2) \quad (2)$$

Here Q_1 is the average radon concentration in air in the Tokyo Metropolitan University Arakawa Campus building. Q_2 is the average indoor radon concentration in air in Japan (15.5 Bq m^{-3})⁹⁾. T_1 is occupancy factor (0.8) multiplied by the annual average time at the location (2,000 h). T_2 is obtained by subtracting the average time at the location (2,000 h) from the hours in a year (8,760 h) and multiplying by the occupancy factor (0.8).

Results and Discussion

Radon concentration in air was measured in the pulse type ionization chamber for each day of the week and the results are summarized here. The respective average ($\pm \sigma$) radon concentrations (Bq m^{-3}) for each day of the week from Sunday to

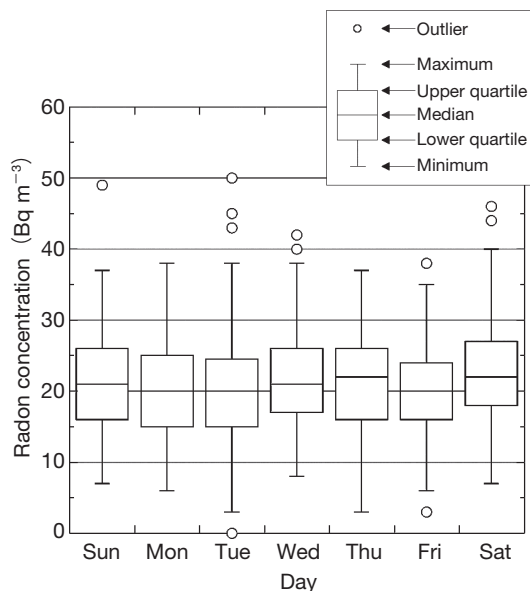


Figure 2 Distribution of radon concentration for each day of the week. Shaded time periods indicate the hours that offices are open and students are attending classes.

Saturday were: 21 ± 7 , 20 ± 7 , 20 ± 8 , 22 ± 6 , 21 ± 7 , 20 ± 6 , 23 ± 7 . **Figure 2** shows the distribution and median value of radon concentrations for each day of the week. The error bars in the graph indicate the maximum and minimum values. The average radon concentration in the measurement period (11 weeks) was 21 ± 7 Bq m⁻³. Although no clear differences between days were observed, the highest average value was observed on Saturday. On the other hand, the lowest average value was observed on Monday morning. This value was 14% lower than the Saturday average radon concentration. As an overall tendency, over the weekend (Friday to Sunday) the radon concentration tended to rise. It is likely because of a decreased indoor air ventilation rate. Opening and closing of doors occurred less often on the weekend because the number of people working and students in the building was reduced and the radon accumulated. Despite being a weekday, Wednesday had a higher average radon concentration for two reasons. The

first is because the number of persons entering and exiting the building was very low from Tuesday afternoon to Wednesday morning due to the workers' and students' schedule. Second, Wednesday is the day of the week on which leaving work on time is recommended for university workers. Therefore, the number of people working in the building during the evening time is less compared to the other weekdays and that led to the rise of radon concentration by the decreased air ventilation rate.

The change per hour of the average value of radon concentration, temperature, pressure and humidity for the five weekdays and two weekend days are shown in **Figure 3** and **Figure 4**. On weekdays, there was a decrease of radon concentration from a peak value of 25 ± 6 Bq m⁻³ observed at 8:00 to the lowest value 17 ± 7 Bq m⁻³ observed at 19:00. After 19:00, the radon concentration had a rising trend until the next morning. On the other hand, radon concentration on the weekend remained high during the day time. These results suggested that the air ventilation rate due to the entrance and exit of people and the operational status of the air conditioning equipment affect radon concentration.

Pressure and humidity showed a similar trend to the radon concentration. However, the trend of temperature was opposite that of radon concentration. From the measurement results of the present study, a certain correlation was observed in the radon concentration and the indoor environment, although the correlation between the indoor factors, such as temperature or humidity, and radon concentration was negative in the prior study⁵⁾. In this study, measurements were carried out in a highly airtight reinforced concrete building, in which the air conditioning management had been strict, in comparison with air conditioning use in a typical house. Therefore, the present results are considered to reflect the operational status of the air conditioning and the indoor ventilation rate.

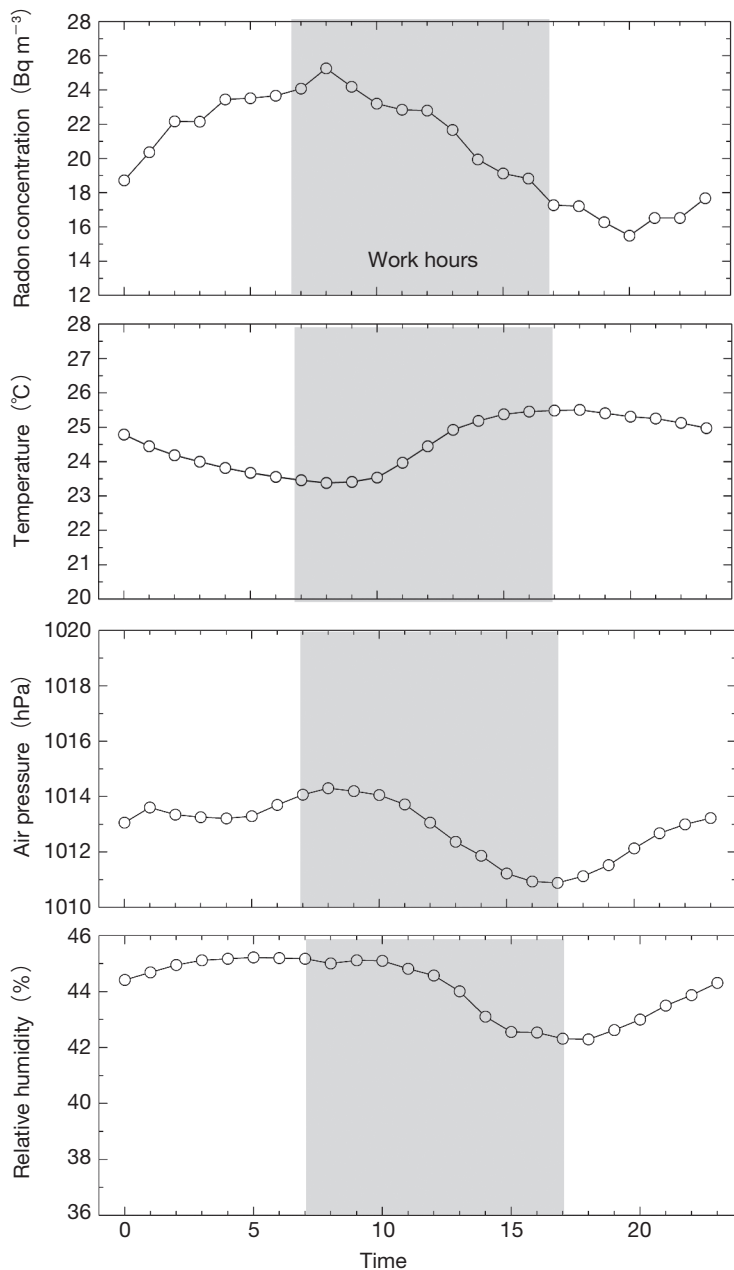


Figure 3 Average value of radon concentration, temperature, pressure and humidity on week days.

The 3-month averages of radon concentration on each floor, which were obtained by the passive radon and thoron discriminative monitors, are listed in **Table 1**. Annual average radon concentration of the entire building was $20 \pm 14 \text{ Bq m}^{-3}$ (3 –

125 Bq m^{-3}), and that value was 130% higher than the indoor average radon concentration in Japan, 15.5 Bq m^{-3} ¹⁰). Compared to a typical house, public facilities including office buildings and university buildings tend to be highly airtight. There

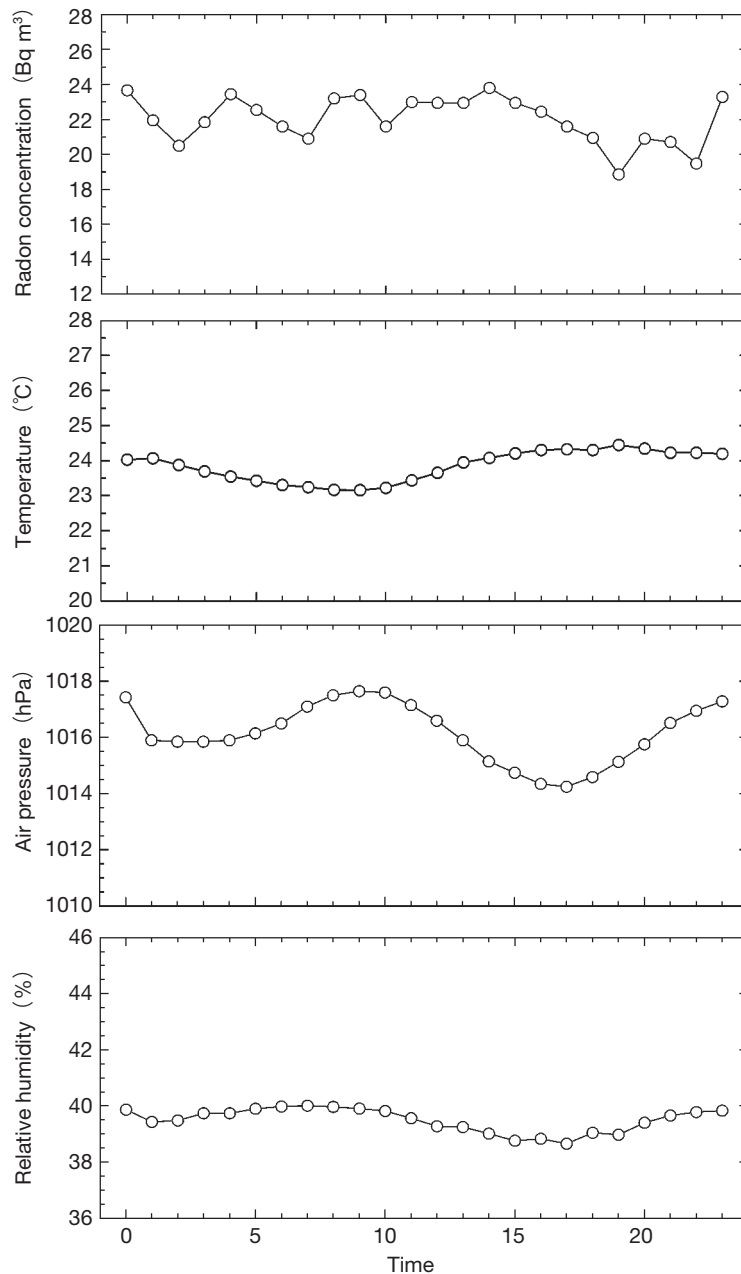


Figure 4 Average value of radon concentration, temperature, pressure and humidity on weekends.

is a tendency that radon concentration is high because in many cases reinforced concrete is used as a primary building material³⁾. The 3-month averages of radon concentration were $23 \pm 22 \text{ Bq m}^{-3}$ (October–December), $17 \pm 13 \text{ Bq m}^{-3}$ (January–

March), $23 \pm 11 \text{ Bq m}^{-3}$ (April–June), $18 \pm 12 \text{ Bq m}^{-3}$ (July–September); and no seasonal variation was observed. Although to the windows of the campus building can be opened and closed, from the viewpoint of security, they are basically

Table 1 Average radon concentrations (Bq m⁻³) for each floor level.

Floor level	Oct.-Dec.	Jan.-Mar.	Apr.-Jun.	Jul.-Sep.
	Ave. ± σ (range)	Ave. ± σ (range)	Ave. ± σ (range)	Ave. ± σ (range)
5F	19 ± 1 (18-20)	14 ± 6 (7-23)	17 ± 4 (10-20)	10 ± 2 (7-13)
4F	25 ± 9 (12-36)	19 ± 15 (8-44)	24 ± 9 (15-29)	18 ± 8 (11-31)
3F	14 ± 6 (5-20)	14 ± 9 (5-28)	21 ± 5 (15-29)	11 ± 5 (4-17)
2F	17 ± 9 (7-31)	12 ± 4 (9-19)	20 ± 9 (7-37)	13 ± 8 (3-22)
1F	30 ± 31 (6-125)	22 ± 17 (3-69)	27 ± 14 (7-52)	25 ± 14 (7-46)
Ave.	23 ± 22 (5-125)	17 ± 13 (3-69)	23 ± 11 (7-52)	18 ± 12 (3-46)

Table 2 Average thoron concentrations (Bq m⁻³) for each floor level.

Floor level	Oct.-Dec.	Jan.-Mar.	Apr.-Jun.	Jul.-Sep.
	Ave. ± σ (range)	Ave. ± σ (range)	Ave. ± σ (range)	Ave. ± σ (range)
5F	17 ± 7 (LLD-29)	ND	20 ± 12 (LLD-40)	12 ± 2 (9-15)
4F	18 ± 9 (LLD-33)	13 ± 3 (9-18)	15 ± 3 (LLD-19)	12 ± 3 (LLD-11)
3F	38 ± 40 (LLD-107)	17 ± 7 (LLD-29)	21 ± 13 (LLD-43)	ND
2F	19 ± 14 (LLD-50)	25 ± 18 (LLD-56)	16 ± 6 (LLD-29)	15 ± 8 (LLD-30)
1F	37 ± 22 (15-74)	14 ± 7 (4-33)	20 ± 10 (LLD-36)	15 ± 8 (7-41)
Ave.	28 ± 24 (LLD-107)	16 ± 10 (LLD-56)	18 ± 9 (LLD-43)	14 ± 7 (LLD-41)

Note: LLD indicates the value was below 13 Bq kg⁻¹. ND indicates not detected.

closed. Therefore, it is reasonable that no seasonal variation was observed when the environment is well managed by the air conditioning system. There was a tendency that radon concentrations measured on the first floor were higher than the other floors. Laboratories occupy most of first floor of the building. Therefore, there is less entering and exiting of doors, and the air ventilation rate is lower compared to other floors leading to the higher radon concentration.

The 3-month averages and the annual averages of thoron concentration on each floor are listed in **Table 2**. Annual average thoron concentration of the entire building was 19 ± 7 Bq m⁻³ (LLD-107 Bq m⁻³) which was lower than previously reported¹¹⁾. The 3-month averages of thoron concentration were 28 ± 24 Bq m⁻³ (October-December), 16 ± 11 Bq m⁻³ (January-March), 18 ± 10 Bq m⁻³ (April-June), 14 ± 7 Bq m⁻³ (July-September); and no seasonal variation was observed, just as for the radon concentration results. It has been reported that there was no effect on the thoron concentration due to seasonal variation⁵⁾. No significant trend for the annual average thoron

concentration on each floor was measured. Thoron concentration up to 107 Bq m⁻³ was observed on the first floor. In this study, the measuring instrument was installed close to a concrete wall due to the matter of space. Thoron concentration measurements are significantly dependent on the distance from the wall compared to radon concentration measurements, and the location of the detector had a bigger effect for the thoron measurements in the present study.

Radon concentrations are plotted against thoron concentrations in **Figure 5**. The correlation coefficient (r) was 0.172. No clear correlation between radon and thoron concentrations was seen. The thoron concentration should not to be estimated simply from the radon concentration as reported in previous studies.

Annual effective dose (H_1) due to radon inhalation in the building of the Tokyo Metropolitan University Arakawa Campus was 0.15 mSv y⁻¹. In a survey that was carried out in a reinforced concrete high-rise building located in the Tokyo metropolitan area, the average radon concentration in the building was 16.0 Bq m⁻³⁶⁾. Annual effective

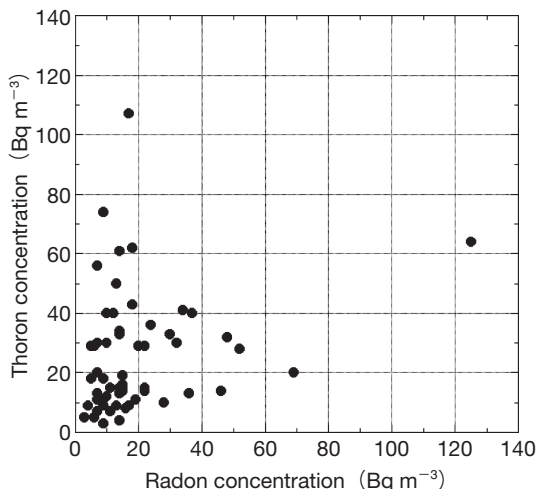


Figure 5 Correlation between radon and thoron concentrations.

dose during working hours was $0.08 \text{ mSv y}^{-1.5}$). The result of the present study was approximately 190% of the previous research value. Annual effective dose (H_2) in the case of considering an indoor environment other than the Arakawa Campus building was 0.42 mSv y^{-1} . This result was 114 % of the annual effective dose 0.37 mSv y^{-1} reported in prior research, and also the value was 33.6% of the dose compared to the global average.

Conclusion

In this study, long term radon and thoron radioactivity concentration measurements were carried out in a reinforced concrete building using the pulse type ionization chamber and passive radon and thoron discriminative monitors. In continuous measurement of radon concentration using the pulse type ionization chamber, the radon concentration varied depending on the air ventilation rate due to the entrance and exit of people and the air conditioning operation status in the building. The average radon and thoron concentrations obtained from the passive radon and thoron discriminative monitors were $20 \pm 14 \text{ Bq m}^{-3}$ and $19 \pm 7 \text{ Bq m}^{-3}$, respectively. No seasonal change and no

correlation between radon and thoron concentrations were observed. Annual exposure dose calculated on the basis of the radon concentrations obtained in the building was 0.15 mSv y^{-1} . Additionally, annual effective dose in the case of considering the indoor environment out of the University was 0.42 mSv y^{-1} .

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