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Estimation of Annual Effective Dose in Namie Town, Fukushima Prefecture due to Inhalation of Radon and Thoron Progeny

Thamaborn Ploykrathok[1†], Koya Ogura[2†], Mayumi Shimizu[1], Masahiro Hosoda[1,2], Yoshitaka Shiroma[3], Hiromi Kudo[2], Yuki Tamakuma[1] and Shinji Tokonami[1*]

[1]Department of Radiation Measurement and Physics Dosimetry, Institute of Radiation Emergency Medicine, Hirosaki University, 66-1 Hon-cho, Aomori 036-8564, Japan
[2]Department of Radiation Science, Graduate School of Health Sciences, Hirosaki University, Hirosaki, Aomori 036- 8564, Japan
[3]University of the Ryukyus, Nishihara, Okinawa 903-0213, Japan
[†]These authors have an equivalent contribution to that of the first author.

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Since Fukushima nuclear accident occurred in 2011, the Japanese government has aimed at reducing the annual effective dose due to high radiation exposure in contaminated areas. Not only the external exposure but also the internal exposure need to be considered for the estimation of the effective dose. In the worldwide average, the effective dose due to radon and thoron by inhalation is estimated to be around 50% from the exposure to natural radiation sources. In the present work, radon, thoron, and thoron progeny measurements have been carried out in 93 dwellings of Namie Town, Fukushima Prefecture. The three-successive-month measurement was conducted for four periods from 2017 to 2019 using a passive type radon-thoron discriminative monitor and a thoron progeny monitor to obtain their activity concentrations and equilibrium equivalent concentration and subsequently to calculate the effective dose. The results showed the method of annual indoor radon activity concentration and thoron progeny concentration to be 31 Bq m^{-3} and 0.7 Bq m^{-3}, respectively. The annual effective doses due to inhalation of radon and thoron progeny using the latest dose conversion factors were estimated to be 1.9 mSv and 0.6 mSv, respectively, and 2.5 mSv in total.

Key words: dose assessment, radon level, thoron progeny, Namie Town

1. Introduction

The Great East Japan Earthquake on 11 March 2011 caused a severe nuclear accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) during which large amounts of radioactive substances were released into the surrounding environment. At the time, all the residents within a 20-km radius from the plant site were forced to evacuate from the affected area. The most contaminated areas, Namie Town, Kawamata Town and Iitate Village[1)], were to the northwest direction from FDNPP. For the present study, the monitoring area was in Namie Town, which is located on the Pacific Ocean coastline of Fukushima Prefecture, and is 4-30 kilometers to the north and northwest of the FDNPP. The town population before the accident was approximately 21,000. On 31 March 2017, the Japanese government partially lifted the evacuation order and then reorganized the evacuation-designated

*Corresponding author: Department of Radiation Physics, Institute of Radiation Emergency Medicine, Hirosaki University, 66-1 Hon-cho, Hirosaki-shi, Aomori 036-8564, Japan
E-mail: tokonami@hirosaki-u.ac.jp

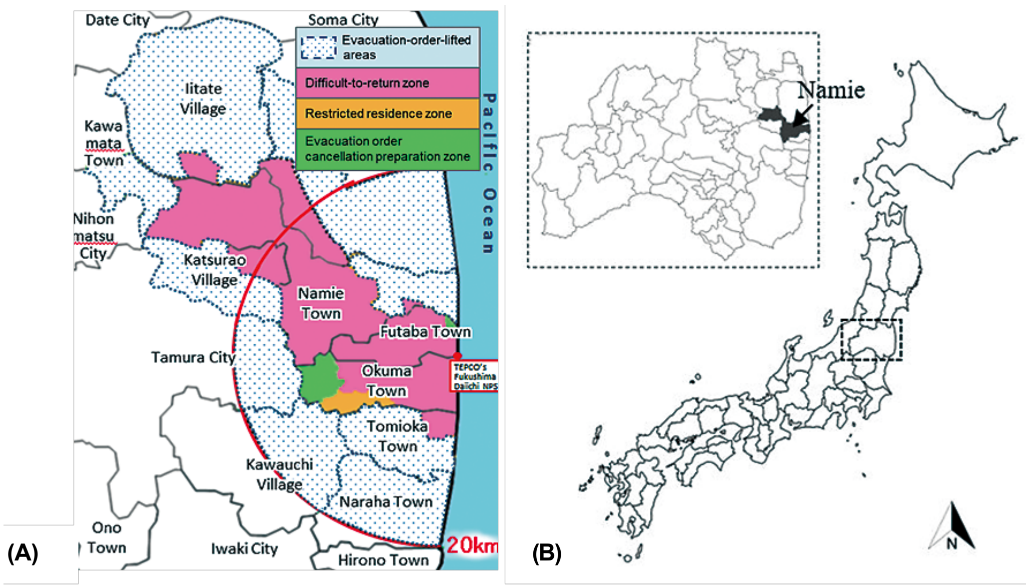


Fig. 1. (A) Officially designed evacuation zones and (B) Location of Namie Town, Fukushima Prefecture, Japan.

zones, which are approximately 2.7% of the entire Fukushima Prefecture area. Some residents returned to the town and have resided there since then[2]. The map in Figure 1(A) shows the officially designed evacuation zones announced by the national government, and taken from the official website of Fukushima Prefecture[3]. Residential restrictions were lifted on the restricted residence zone (annual integrated dose between 20–50 mSv) and the evacuation order-prepared zone (annual integrated dose below 20 mSv), allowing residents to return. Both these zones are in the coastal part of Namie Town and the town's business center and local government offices are within them. Most areas of the town remain in the difficult-to-return zone (annual integrated dose over 50 mSv) with access completely prohibited due to the high residual radiation.

To reconstruct facilities and revitalize the economy in the contaminated areas, the national government aims at a long-term goal to reduce additional annual dose to 1 mSv or less by comprehensive measures. Since the accident, not only decontamination activities, but also radiation monitoring surveys, food safety checks, and risk communication activities, etc., have been conducted. Studies have been carried out for environmental assessments that consider the effects of radiation and radiation risk management through methods such as the car-borne survey[4, 5], food product analysis[6, 7], and radon monitoring[8] in order to collect overall radiation exposure information for residents of Namie Town.

Radon and thoron are considered to be significant contributors to human exposure from natural background radiation sources[9, 10]. Based on their epidemiological studies, the exposure to radon and its progenies is the second-most important cause of lung cancer[11]. Radon and its progenies enter the body through inhalation and deposit on the respiratory epithelium, especially in the bronchi[12]. In 2009, WHO proposed a reference level of 100 Bq m^{-3} to minimize health hazards due to indoor radon exposure. However, if this level cannot be reached under the prevailing country-specific conditions, the chosen reference level should not exceed 300 Bq m^{-3} which represents approximately 10 mSv per year according to calculations by the International Commission on Radiation Protection (ICRP)[13]. A previous survey by Suzuki *et al.*[14] reported that the indoor radon activity concentrations in some Fukushima Prefecture dwellings were >100 Bq m^{-3}, which corresponded to > ~3 mSv. However, a nation-wide survey in Japan by Sanada *et al.*[15] reported the average value of indoor radon concentration as 15.5 Bq m^{-3} and they estimated the average annual effective dose due to radon to be 0.45 mSv[15]. According to the report of the United Nation Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the worldwide average annual human exposure from natural sources is estimated as 2.4 mSv and the effective doses due to inhalation (mainly radon) is approximately 50% of the total effective dose from natural radiation[10]. As the recent study by Omori *et al.*[16] reported, the annual effective dose from natural radiation in Japan is estimated to be 2.2 mSv and the annual effective dose due to inhalation of radon and thoron is estimated to be 0.59 mSv. However, these doses depend on many factors, such as the location, building materials, etc.

Although several nation-wide surveys were carried out before the nuclear accident, those of them were not sufficiently accurate to clarify the natural radiation exposure for a smaller unit, such as a city, towns, or villages. The survey by Tamari *et al.*[17] reported public concerns about radiation exposure after the nuclear accident. There were significant differences between Fukushima and Tokyo residents. The former was more concerned about radiation from the terrestrial environmental, food, and radon than Tokyo residents were. Since the radiation level is a concern in the specific close-by areas affected by the nuclear accident, a radiation survey in a smaller unit is very useful. Such information is valuable to support effective risk communication.

This paper shows the results of radon and thoron progeny monitoring in dwellings of Namie Town that were obtained using a passive radon-thoron discriminative monitor and a thoron progeny monitor to measure the average indoor radon activity concentration and thoron progeny concentration. The paper also estimates the annual effective dose for considering the effects and risks of radiation for health risk assessment.

2. Materials and Methods

2.1. Ethics statement

Approval for this research was obtained from the Ethics Committee of the Graduate School of Health Sciences, Hirosaki University (approval number 2016-061).

2.2. Monitoring site and investigated dwellings

The monitoring area was in Namie Town, Fukushima Prefecture, which is located in eastern Japan. The location map that is shown in Figure 1(B) was made by original maps from D-map.com. The area climate is humid. The average annual temperature is 13.0℃; August is the warmest month with an average temperature of 25.3℃, and January is the coldest, averaging 1.5℃[18]. The average annual precipitation is 1,135 mm; September is the wettest month with an average precipitation of 161 mm, and January is the driest, averaging 47 mm[18].

According to the car-borne survey by Shiroma *et al.*[4] made in June 2017, absorbed dose rate in the air in Namie Town varied from 0.04–11 μGy h^{-1}. These authors estimated the rate varied and from 1–5 μGy h^{-1} in the difficult-to-return zone and from 0.05–1 μGy h^{-1} in the evacuation order-lifted zone[4]. The distribution of absorbed dose rate in the air in 2017 was found to be significantly decreased when compared to the surveys in 2011, 2014 and 2015 by Pornnumpa *et al.*[5] These findings suggest that there were effects from decontamination activities carried out after the FDNPP accident because the values decreased faster than the natural radioactive decay.

The monitored dwellings were selected from among dwellings in the evacuation order-lifted area of Namie Town. Permission to install a monitor was given for a total of 93 houses. The measurements were performed during the period from August 2017 to November 2019. The three-successive-month measurement was conducted for four periods in each dwelling to cover a whole year; the 1st, 2nd, 3rd, and 4th periods were October-December, January-March, April-June and July-September, respectively. In one of the dwellings two monitors were deployed, one each in the living room and the bedroom. Measurements results of the two monitors were combined into one by averaging them. Dwellings made of wood represented 94% of the samples, and the others were steel frame, 5% and reinforced concrete buildings, 1%. Approximately 25% of the dwellings were built over 30 y ago. Most of the monitors were installed in the living room on the first floor, and both radon- thoron discriminating monitors and thoron progeny monitors were placed at the same position.

2.3. Measurement of radon, thoron and thoron progeny concentration using passive monitors.

Passive type radon-thoron discriminative monitors, RADUET (Radosys Ltd., Hungary) were used in the present work; they are known to be suitable for large scale surveys[19]. These devices had two chambers with different diffusion rates; one was a low rate and the other, high. The chambers were made of electro-conductive plastic with an inner volume of 30 cm^2 and each chamber had a CR-39 chip with dimensions of 10×10 mm^2 (BARYOTRAK, Nagase Landauer, Ltd., Japan) at the chamber bottom. The high diffusion rate chamber had six holes in the sidewall and was covered with an electro-conductive sponge that acts as a filter. Therefore, there was a difference in ventilation between the two chambers of the RADUET.

After measurements were completed, the CR-39 chips were taken out of the RADUET and processed at the Institute of Radiation Emergency Medicine (IREM). Chemical etching was performed in 6M NaOH solution at 60℃ for 24 h. After the latent tracks were produced, they were counted at 100×magnification with an optical microscope. To calculate activity concentrations of radon, C_{Rn} (Bq m^{-3}), and thoron, C_{Tn} (Bq m^{-3}), the obtained track densities were used in the following equations:

$$N_L = C_{Rn} \times CF_{L\text{-}Rn} \times T + C_{Tn} \times CF_{L\text{-}Tn} \times T + N_B, \quad (1)$$

$$N_H = C_{Rn} \times CF_{H\text{-}Rn} \times T + C_{Tn} \times CF_{H\text{-}Tn} \times T + N_B, \quad (2)$$

where N_L and N_H are track densities (tracks cm^{-2}) of the low and high diffusion rate chambers of RADUET, respectively, N_B is the background track density (tracks cm^{-2}) of the CR-39 chip, $CF_{L\text{-}Rn}$ and $CF_{H\text{-}Rn}$ are the conversion factors for the low and high diffusion rates of radon exposure (3.03 and 2.94 tracks cm^{-2} kBq^{-1} m^3 h^{-1}, respectively), and $CF_{L\text{-}Tn}$ and $CF_{H\text{-}Tn}$ are the conversion factors for the low and high diffusion rate of thoron exposure (0.03 and 2.08 tracks cm^{-2} kBq^{-1} m^3 h^{-1}, respectively). The conversion factors for radon and thoron were derived from calibration experiments using the radon and thoron calibration chambers at Institute of Radiation Emergency Medicine, Hirosaki University. T is the exposure time (h).

Thoron progeny monitors also used the CR-39 chips. This detector developed by Zhuo and Iida[20] and modified by National Institute of Radiological Sciences, NIRS[21]. The CR-39 chips were mounted on a stainless-steel plate, which was covered with an aluminum-vaporized Mylar film of 71-mm air-equivalent thickness. The total thickness of the film was adjusted to detect only the 8.78-MeV alpha particles emitted from ^{212}Po (thoron progeny). After measurements were completed, the CR-39 chips of the thoron progeny monitors were processed in the same way as the CR-39 chips of the RADUET. The thoron progeny concentration was expressed as equilibrium equivalent thoron concentration, *EETC*. To calculate concentrations, the obtained track densities were substituted into the following equation:

$$N_{TnP} = EETC \times CF_{TnP} \times T + N_B, \qquad (3)$$

where N_{TnP} is the track density (tracks cm^{-2}) of thoron progeny monitors, N_B is the background track density (tracks cm^{-2}) of the CR-39 chip, CF_{TnP} is the conversion factor for the thoron progeny monitor (6.9$\times$10^{-2} tracks cm^{-2} kBq^{-1} m^3 h^{-1}), which was derived from a previous experiment[22], and T is the exposure time (h).

2.4. Evaluation of annual effective doses.

The annual effective doses due to inhalation of radon and thoron were estimated using the radon activity concentrations and *EETCs*. In this estimation, the indoor radon concentrations and *EETC* were obtained by the passive method whereas the outdoor radon concentration was derived from the nation-wide survey by the Japanese Chemical Analysis Center, JCAC[23]. However, the dose due to exposure of thoron outdoors was not included because outdoor thoron progeny concentration was not available. The annual effective doses due to radon (D_{Rn}) and thoron progeny (D_{Tn}) were calculated using the following relation, which was reported by the UNSCEAR:

$$D_{Rn} = [\,(C_{Rn} \times F \times DCF_{RnP} \times OF \times 24 \times 365.25)_{indoor} + (C_{Rn} \times F \times DCF_{RnP} \times OF \times 24 \times 365.25)_{outdoor}\,], \qquad (4)$$

Table 1. Descriptive statistical parameters for indoor radon concentrations and EETCs

	Period[a]	Median (Bq m^{-3})	Range (Bq m^{-3})
Radon	1st	32	6–242
	2nd	28	10–145
	3rd	27	9–137
	4th	31	6–113
	Annual	31	
EETC	1st	0.7	0.2–8
	2nd	0.7	0.1–9
	3rd	0.8	0.2–20
	4th	0.8	0.1–14
	Annual	0.7	

[a] 1st, October-December; 2nd, January-March; 3rd, April-June; 4th, July-September

$$D_{Tn} = EETC_{indoor} \times DCF_{TnP} \times OF \times 24 \times 365.25, \qquad (5)$$

where C_{Rn} is the activity radon concentration (Bq m^{-3}), *EETC* is the equilibrium equivalent thoron concentration (Bq m^{-3}), DCF_{RnP} (1.7$\times$10^{-5} mSv per Bq m^{-3} h) and DCF_{TnP} (1.07$\times$10^{-4} mSv per Bq m^{-3} h) are the radon and thoron dose coefficients, respectively, which are reported in ICRP publication 137[24]; and OF is the occupancy factor (h). The indoor and outdoor occupancy factors for an indoor worker were set as 0.83 and 0.17, respectively. For calculation of equilibrium equivalent radon concentration (EERC), the indoor and outdoor equilibrium factors (F) between radon and its progeny were assumed as 0.4 and 0.6, respectively[25].

The occupancy factors used in the present work were based on answers to a questionnaire of Namie residents, which was conducted during the measurements. The values were estimated from the activity records of the residents living in each dwelling. These records were separated into two categories, time spent within the investigated room and time spent outside the dwelling, and then the average occupancy factors were calculated. The average indoor and outdoor occupancy factors of Namie residents were 0.83 and 0.17, respectively. This suggested the accurate effective doses of Namie residents could be obtained when the actual occupancy factors were used.

3. Results and discussions

3.1. Measurement of radon, thoron, and thoron progeny concentrations in dwellings

The descriptive statistics for indoor radon and thoron progeny concentrations in 93 dwellings of Namie Town are shown in Table 1. The radon concentrations varied from 6–242 Bq m^{-3} and the median in each period were 32, 28, 27 and 31 Bq m^{-3}, respectively. The annual indoor radon concentration was 31 Bq m^{-3}. The thoron progeny

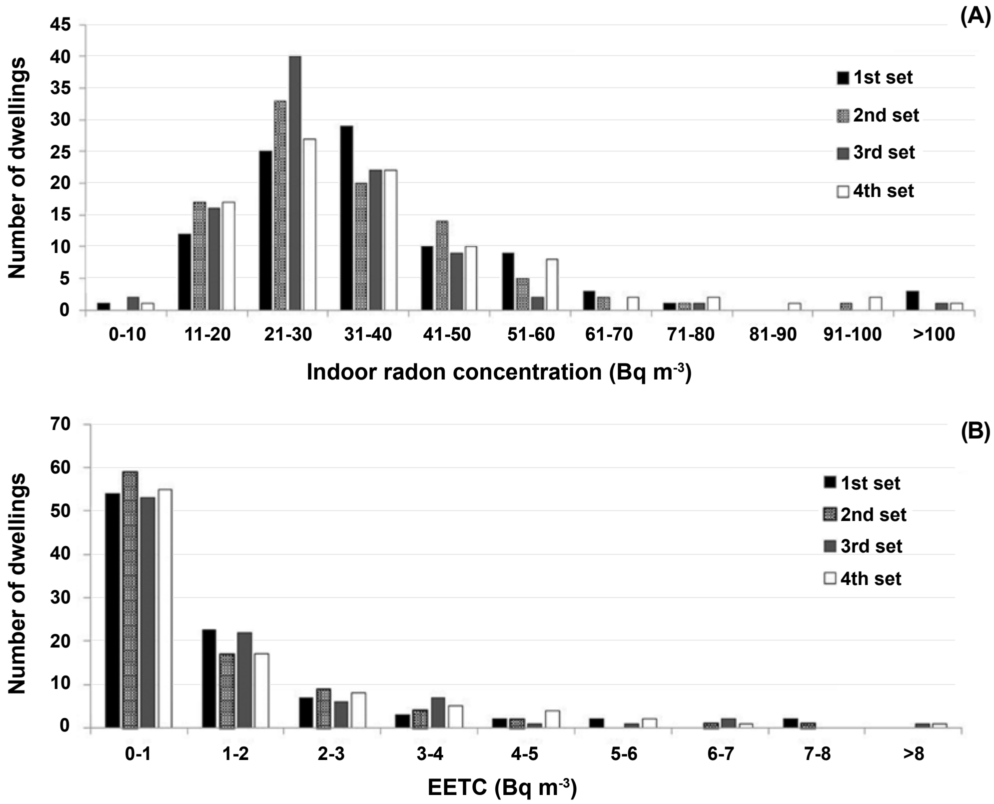
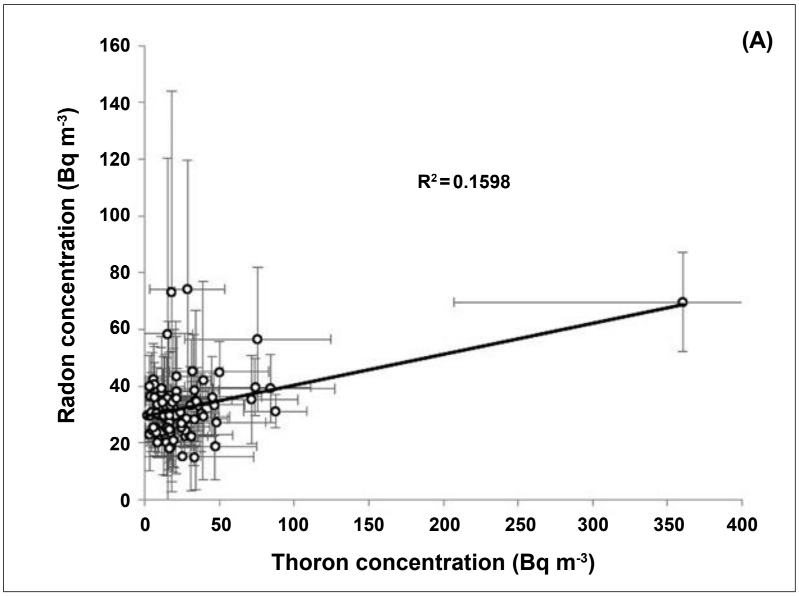


Fig. 2. Frequency distribution of (A) indoor radon concentrations and (B) thoron progeny concentrations, (n = 93).

concentrations varied from 0.1–20 Bq m^{-3} and the median in each period were 0.7, 0.7, 0.8 and 0.8 Bq m^{-3}, respectively. The annual thoron progeny concentration was 0.7 Bq m^{-3}. In the present paper, the annual arithmetic mean with standard deviation is shown for comparison with the previous reports. The annual mean with standard deviation of radon and thoron progeny concentrations were evaluated 34 ± 12 and 1.4 ± 1.6, respectively. The indoor radon concentrations observed in the present work were higher than in previous surveys in Fukushima Prefecture by Fujimoto *et al.*[26] (arithmetic mean of 18.1 Bq m^{-3}) and Suzuki *et al.*[14] (arithmetic mean of 14.3 Bq m^{-3}) as well as in the Tohoku region by Sanada *et al.*[15] (arithmetic mean of 15.5 Bq m^{-3}), while they were lower than the worldwide average reported by WHO (39 Bq m^{-3})[13]. Although the median indoor concentration of the present study did not exceed the reference level, there were significantly different from results of previous studies that were conducted in the similar region. There are many things that can affect radon concentration results, such as different types of monitors, construction materials, ventilation rates, location, and age of dwellings, and seasonal variation. In the present case, the influence of dwelling materials seemed more likely because there were no problems with ventilation and monitoring process. Furthermore, from a gamma-ray spectrum analysis in the surrounding area, uranium and thorium concentrations in soil were found to be not too high (25.8 Bq kg^{-1} and 20.6 Bq kg^{-1}, respectively). It was considered that radon and thoron exhaled from the surrounding soil did not interfere with the measurement. However, information about the building materials for the dwellings of the present study is limited. Therefore, activities must be continued to collect data and raise awareness of factors that affect the results. As for the thoron progeny, the concentration was found lower than in the previous survey by Yamasaki *et al.*[27] (1.5 Bq m^{-3}), but it is considerably higher than the worldwide average of 0.3 Bq m^{-3} [25].

The estimated thoron concentrations varied from 0–87 Bq m^{-3}. The values fluctuated significantly between periods for the same dwelling and between the living room and the bedroom within the same period. As a result, it was not possible to derive the representative value of thoron concentration by the passive type radon-thoron monitors. However, the purpose of thoron measurements is to measure the radon concentration without thoron interference. The results of thoron concentration are only shown here for reference because the concentrations obtained by such monitors strongly



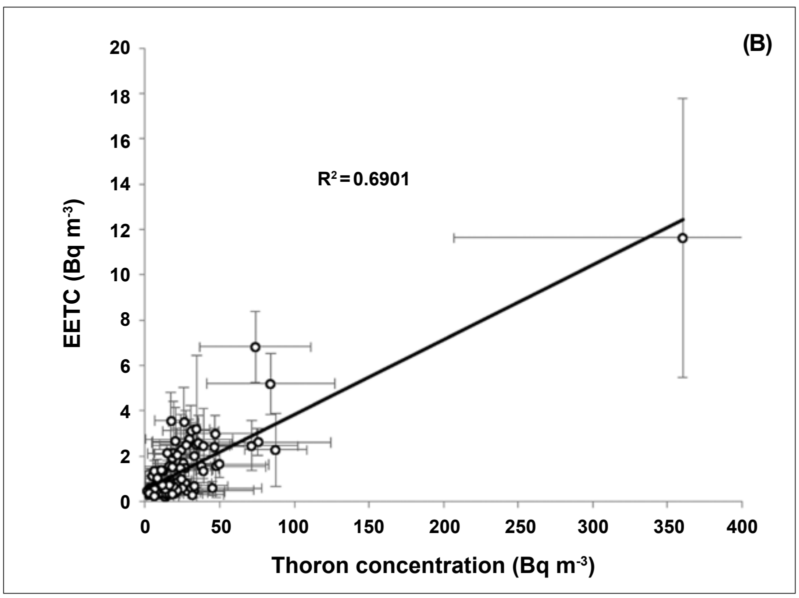


Fig. 3. Correlation of (A) radon concentration and (B) EETC against thoron concentration for each dwelling.

depend on the distance from the wall surface[28, 29]. Thoron can migrate from its source through the atmosphere for a small distance due to its short half-live (55.6 s). Thus, the difference in installation position of the monitor has a significant effect on thoron concentration analysis.

The distributions of radon and thoron progeny concentrations for dwellings are shown in Figure 2. The geometric mean of radon and thoron progeny concentrations are 32 Bq m^{-3} (GSD = 1.4) and 0.9 Bq m^{-3} (GSD = 2.3). The normality tests were performed using the Shapiro-Wilk test. The test of normality shows that the p-value for radon and thoron progeny concentration is 3.7×10^{-8} and 4.2×10^{-14}. The test of lognormality shows that the p-value for radon and thoron progeny concentration is 0.019 and 0.002. The results indicate that both radon and thoron progeny had neither a normal nor a log-normal distribution. However, for both radon and thoron progeny, the similar pattern of frequency distributions was observed in every period.

The relationships between radon concentration and thoron concentration, as well as thoron progeny concentration and thoron concentration, are shown in Figure 3. No correlations were found between radon and thoron progeny or thoron progeny and thoron. These results showed that thoron concentration was independent of other species concentrations. As explained

Table 2. Descriptive statistical parameters for annual effective dose due to radon and thoron progeny

	Median[a] (mSv)	Median[b] (mSv)	Range[c] (mSv)
Radon	1.9	1.0	1.0–4.3
Thoron	0.6	0.2	0.2–9.0
Total	2.5	1.3	1.5–13.0

[a] The median values using the latest dose conversion factor
[b] The median values using the conventional dose conversion factor
[c] The effective dose ranges using the latest dose conversion factor

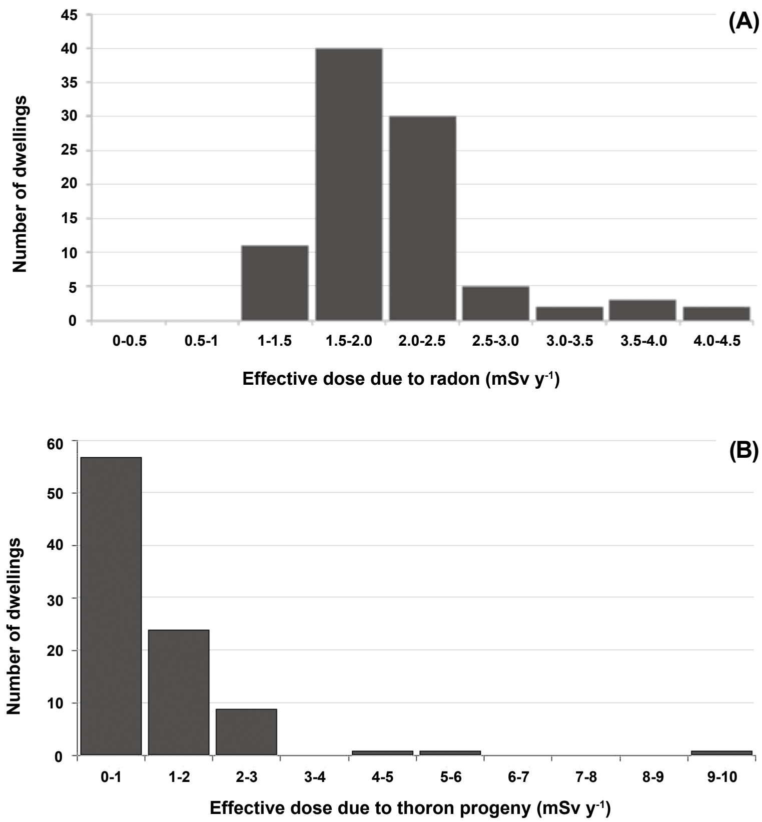


Fig. 4. Frequency distribution of annual effective dose due to (A) radon and (B) thoron progeny, (n = 93).

above, thoron with its rather short half-life could not be distributed uniformly in the investigated rooms, whereas radon and thoron progeny could be considered as homogeneously distributed in the rooms due to their relatively long half-lives. Therefore, the present findings implied that both radon and thoron progeny concentrations could not be predicted from thoron concentrations, as a finding that has been noted in another study as well[21)].

3.2. Estimation of annual effective dose

Radon, thoron and thoron progeny concentrations measured in the 93 dwellings were used to evaluate the annual effective dose. The descriptive statistics for the effective doses due to radon and thoron are shown in Table 2. According to the questionnaire of Namie residents, the indoor and outdoor occupancy factors were used as 0.83 and 0.17, respectively. In order to estimate the effective dose due to radon exposure, the present study used an average outdoor concentration of 6.3 ± 2.6 Bq m^{-3}, which was the outdoor radon concentration in Fukushima Prefecture from the nation-wide survey by Oikawa *et al.*[30)] for calculating the effective dose due to outdoor radon.

The annual effective dose due to radon was estimated to vary from 1.0–4.3 mSv with a median value of 1.9 mSv.

On the other hand, the annual effective dose due to thoron was calculated from the equilibrium equivalent thoron concentration, it was estimated to vary from 0.2–9.0 mSv with a median value of 0.6 mSv. The descriptive of effective doses due to radon and thoron are shown in Figure 4. The total annual effective dose due to radon and thoron varied from 1.5–13.0 mSv, with a median value of 2.5 mSv. A comparison with results from previous studies must be done carefully because the present study used the latest dose conversion factor of ICRP for the annual effective dose estimation. There is approximately two-fold difference between the conventional (9 nSv per Bq m^{-3} h for radon, and 40 nSv per Bq m^{-3} for thoron) and the latest (17 nSv per Bq m^{-3} h for radon, and 107 nSv per Bq m^{-3} for thoron) dose conversion factors. If the conventional conversion factors were applied to radon and thoron concentrations, the annual effective dose due to inhalation of radon and thoron were estimated as 1.0 mSv and 0.2 mSv, respectively. Then, the total annual effective dose was estimated as 1.3 mSv, with the maximum value of 5.5 mSv and the minimum value of 0.7 mSv.

The estimated effective dose due to inhalation of radon was higher than in a previous study by Hosoda *et al.*[8] (0.22 mSv), which was estimated for the temporary houses of evacuees in Fukushima Prefecture, and it was also higher than the Japanese average[16] as well as worldwide annual effective dose[9]. The total annual effective dose due to radon and thoron estimated in the present study was similar to that as previously reported by Yonehara *et al.*[31], who reported the annual effective doses from both radon and thoron ranged from 0.6–2.2 mSv in Japanese traditional houses. However, the median indoor concentration in the present survey did not exceed the internationally recognized reference level in the air of residential dwellings. Furthermore, to provide information for risk communication, studies of the radiation dose have been conducted in areas affected by the FDNPP accident. Kumagai and Tanikawa[32] reported radiation doses of Fukushima residents obtained via the Fukushima Health Management Survey. They reported the average effective dose due to external radiation of Namie residents for the first four months after the FDNPP accident was 0.8 mSv. Hosokawa *et al.*[6] estimated the internal dose of Namie residents due to intake of radiocesium in foodstuff by whole-body counter measurements. As a result, an average committed effective dose due to radiocesium was reported to be approximately 1 μSv. This was considered to be an extremely low risk. On the other hand, the number of food products brought in for detection decreased as this study period progressed, but the number of food products with radioactivity had increased. From the viewpoint of radiation protection, the exposure doses received by Namie residents reported in the present study will be useful for health assessment and risk communication.

4. Conclusions

In this survey, indoor radon and thoron measurements were carried out using passive radon-thoron discriminative monitors and thoron progeny concentration measurements were carried out using thoron progeny monitors for four periods in 93 dwellings in Namie Town, Fukushima Prefecture. The medians of indoor radon and thoron progeny concentrations were obtained as 31 Bq m^{-3} and 0.7 Bq m^{-3}, respectively. The annual effective doses due to the inhalation of radon and thoron progenies were estimated to be 1.9 mSv and 0.6 mSv, respectively, and 2.5 mSv in total. The dwellings had a maximum value of 13.0 mSv and a minimum value of 1.5 mSv. This survey will provide information on natural radiation exposure for Namie residents.

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Conflict of Interest

The authors declare that they have no conflicts of interests.

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