

A comparison of the dose from natural radionuclides and artificial radionuclides after the Fukushima nuclear accident

Masahiro Hosoda¹, Shinji Tokonami^{2*}, Yasutaka Omori^{3,4},
Tetsuo Ishikawa⁵ and Kazuki Iwaoka²

¹Department of Radiological Life Sciences, Division of Medical Life Sciences, Hirosaki University Graduate School of Health Sciences, 66–1 Honcho, Hirosaki, 036–8564, Japan

²Department of Radiation Physics, Hirosaki University, Institute of Radiation Emergency Medicine, 66–1 Honcho, Hirosaki, 036–8564, Japan

³Research Center for Radiation Protection, National Institute of Radiological Sciences, 4–9–1Anagawa, Inage-ku, Chiba-shi, Chiba, 263–8555, Japan

⁴Present address: Department of Radiation Physics and Chemistry, Fukushima Medical University, 1 Hikarigaoka, Fukushima, 960–1295, Japan

⁵Department of Radiation Physics and Chemistry, Fukushima Medical University, 1 Hikarigaoka, Fukushima, 960–1295, Japan

*Corresponding author. Department of Radiation Physics, Hirosaki University, Institute of Radiation Emergency Medicine, 66–1 Honcho, Hirosaki, 036–8564, Japan. Tel and Fax: +81–172–39–5404; Email: tokonami@hirosaki-u.ac.jp

Received August 27, 2015; Revised December 1, 2015; Accepted December 4, 2015

ABSTRACT

Due to the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, the evacuees from Namie Town still cannot reside in the town, and some continue to live in temporary housing units. In this study, the radon activity concentrations were measured at temporary housing facilities, apartments and detached houses in Fukushima Prefecture in order to estimate the annual internal exposure dose of residents. A passive radon–thoron monitor (using a CR-39) and a pulse-type ionization chamber were used to evaluate the radon activity concentration. The average radon activity concentrations at temporary housing units, including a medical clinic, apartments and detached houses, were 5, 7 and 9 Bq m⁻³, respectively. Assuming the residents lived in these facilities for one year, the average annual effective doses due to indoor radon in each housing type were evaluated as 0.18, 0.22 and 0.29 mSv, respectively. The average effective doses to all residents in Fukushima Prefecture due to natural and artificial sources were estimated using the results of the indoor radon measurements and published data. The average effective dose due to natural sources for the evacuees from Namie Town was estimated to be 1.9 mSv. In comparison, for the first year after the FDNPP accident, the average effective dose for the evacuees due to artificial sources from the accident was 5.0 mSv. Although residents' internal and external exposures due to natural radionuclides cannot be avoided, it might be possible to lower external exposure due to the artificial radionuclides by changing some behaviors of residents.

KEYWORDS: radon, Fukushima, internal dose, external dose, natural source, artificial source

INTRODUCTION

Large amounts of artificial radionuclides were released by the accident at TEPCO's Fukushima Daiichi Nuclear Power Plant (FDNPP) in March 2011, and areas were heavily contaminated in the northwest direction from the plant site; these areas included Namie Town, Iitate Village and Kawamata Town [1, 2]. All residents of Namie Town were evacuated to other locations, such as Fukushima City and Nihonmatsu City, as a result (Fig. 1A), and none have been able to reside in the

town since then. Some of the evacuees continue to live in temporary housing units, which are prefabricated buildings (Fig. 1B). Environmental radiation monitoring and internal dose estimation were carried out in Namie Town by the authors immediately after the FDNPP accident [3]. Consequently, an agreement to provide reconstruction support, including carrying out environmental assessment, proposing decontamination measures, and educating the evacuees and Namie Town workers about radiation and decontamination measures was

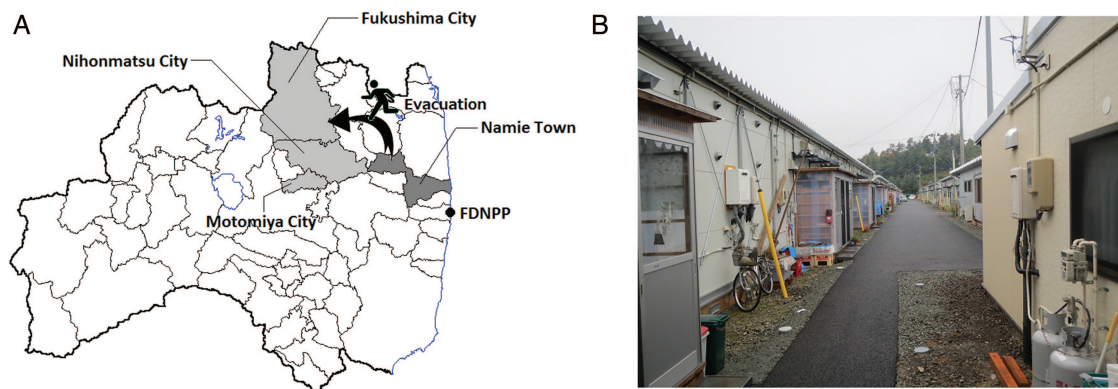


Fig. 1. (A) Map of Fukushima Prefecture. Residents of Namie Town were evacuated to other locations. (B) Photo showing the outside appearance of temporary housing units.

reached between Namie Town and Hirosaki University on 29 September 2011 [3].

As well, whole-body counter (WBC) examinations to estimate internal exposure to radiocesium have been carried out by Fukushima Prefecture and Namie Town, beginning several months after the accident [4]. Thousands of WBC examinations have also been carried out within Fukushima Prefecture [5–7]. Many institutions measure radiocesium contamination of food using high-purity germanium detectors and NaI(Tl) scintillation counters [8, 9]. External dose estimations for children and pregnant woman have been carried out using radio photoluminescence dosimeters or optically stimulated luminescence dosimeters [10]. The goal of these examinations has been to estimate the additional doses and to evaluate the effects on the residents caused by the artificial radionuclides released from the reactor buildings.

A fact that is often ignored by the general public (or unknown to them) is that people were exposed to natural radiation sources such as radon, cosmic-rays and terrestrial gamma-rays before the FDNPP, and that such exposure continues on a daily basis. According to a report by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the effective doses from radon (^{222}Rn) and thoron (^{220}Rn) and their decay products comprise ~50% of the world's average effective dose [11]. Radon and thoron, which are radioactive noble gases, are normally generated by radioactive decay from radium (^{226}Ra and ^{224}Ra) in soil, rocks and building materials. Radon inhalation is believed to increase the risk of lung cancer and is second only to tobacco smoking as a risk factor [12]. It has been previously reported by Darby *et al.* [13] that the relative risk of lung cancer increases 16% per 100 Bq m^{-3} indoor radon activity concentration. Recently, the WHO [12] proposed a reference level for indoor radon gas ranging from 100 to 300 Bq m^{-3} .

UNSCEAR has reported the estimated effective doses for residents in each city of Fukushima Prefecture [14]. An annual effective dose of 2.1 mSv was used as the Japanese average value from natural radiation in this report. It is well known, however, that natural radiation doses depend on characteristics of the location, such as the types of bedrock, building material types, and other factors. Indoor radon activity concentration $>100 \text{ Bq m}^{-3}$ (which corresponds to ~3 mSv) as an annual effective dose was found in a house in Fukushima

Prefecture [15]. Furthermore, indoor radon measurements in the temporary housing facilities for evacuees have not been carried out or reported until now. It can be easily imagined that evacuees want any available information about radiation exposure by the various radiation sources. Information about natural radiation doses should be the baseline for the public when considering the effects of artificial radiation. However, the evacuees who are living in temporary housing do not know the natural radiation doses around their houses. In the present study, therefore, the indoor radon activity concentration was measured at temporary housing facilities (these included units for living and a medical clinic), apartments and detached houses in Fukushima Prefecture in order to estimate the annual internal exposure dose. Moreover, the estimated dose by indoor radon was compared with the first-year internal and external doses from the artificial radionuclides released by the FDNPP accident as estimated using results published in the literature. The average effective doses caused by natural and artificial radiation were compared to gain some perspective on the overall exposure of the evacuees from Namie Town in relation to that of residents of the main cities such as Fukushima City and Nihonmatsu City. This comparison will give useful information for considering the effects of artificial radiation on the residents.

MATERIALS AND METHODS

Measurement of indoor radon activity concentration using an active method

A pulse-type ionization chamber (AlphaGUARD PQ2000PRO, Saphymo GmbH, Germany) was used to evaluate the daily variation of indoor radon activity concentration. The AlphaGUARD was placed on a desk in the Tsushima Clinic (located within a temporary housing facility in Nihonmatsu City). The radon activity concentration was measured during the period from October 2012 to January 2013. The diffusion measurement mode was used with a time interval of 60 min. This AlphaGUARD was calibrated in the radon calibration chamber at the National Institute of Radiological Sciences (NIRS), Japan. The reference value of the radon activity concentration in the radon chamber at NIRS is based on the measurement value obtained by the standard ionization chamber calibrated at the Physikalisch Technische Bundesanstalt (PTB), Germany [16]. The temperature

and relative humidity in the radon calibration chamber can be controlled, respectively, in the ranges of 5–30°C with an error of 0.5°C, and 30–90% with an error of 3% [17, 18]. The calibration conditions of the AlphaGUARD in the radon calibration chamber at NIRS were as follows. The average radon activity concentration, temperature and relative humidity were 9460 Bq m^{-3} , 21.0°C and 59.9%, respectively. In this calibration experiment, the calibration factor of the AlphaGUARD used was found to be 0.995. After calibration, the AlphaGUARD was put into a stainless steel tank for estimation of background concentration using nitrogen gas, and that was determined to be 4.7 Bq m^{-3} .

Measurement of indoor radon activity concentration using a passive method

A passive type radon–thoron discriminative monitor called a RADUET (Radosys Ltd, Hungary, Fig. 2) [19] using a solid-state track detector (CR-39; Radosys Ltd) was used to evaluate the average radon activity concentration. Radon (^{222}Rn , half-life: 3.82 d) in air could penetrate into the chamber through an invisible air gap between its lid and bottom by molecular diffusion. Since this air gap functioned as a high diffusion barrier, hardly any thoron could enter

the chamber with such a small pathway due to its very short half-life (55.4 s). In order to detect thoron, six holes of 6 mm diameter were opened in the chamber side wall and covered with an electroconductive sponge. In this way, thoron could easily go into the chamber, and radon and thoron could be discriminated by the monitor; however, the electroconductive sponge prevented radon and thoron progeny from easily going into the chamber. Usually, the low and the high air-exchange-rate chambers are set as a pair at the measurement place.

Since higher indoor radon activity concentrations are generally observed in winter, the measurements were made during the period from October 2012 to January 2013, and monitors were placed in 50 residences in Fukushima City, Nihonmatsu City and Motomiya City. Among them, 25 residences were temporary housing units (including the Tsushima Clinic), 14 were apartments and 11 were detached houses. Almost all of the RADUETs were set in the living room on the first floor. After exposing each monitor for three months, the CR-39 was chemically etched for 6 h in a 6.25 M NaOH solution at 90°C. The number of alpha tracks was counted using the automatic track reading system (Radosys Ltd.). Average radon and thoron activity concentrations (X_{Rn} and X_{Th}) were calculated using Eqs (1) and (2) [19]. The radon activity concentrations obtained were considered to include the contribution by thoron in the measurement place. Thus,

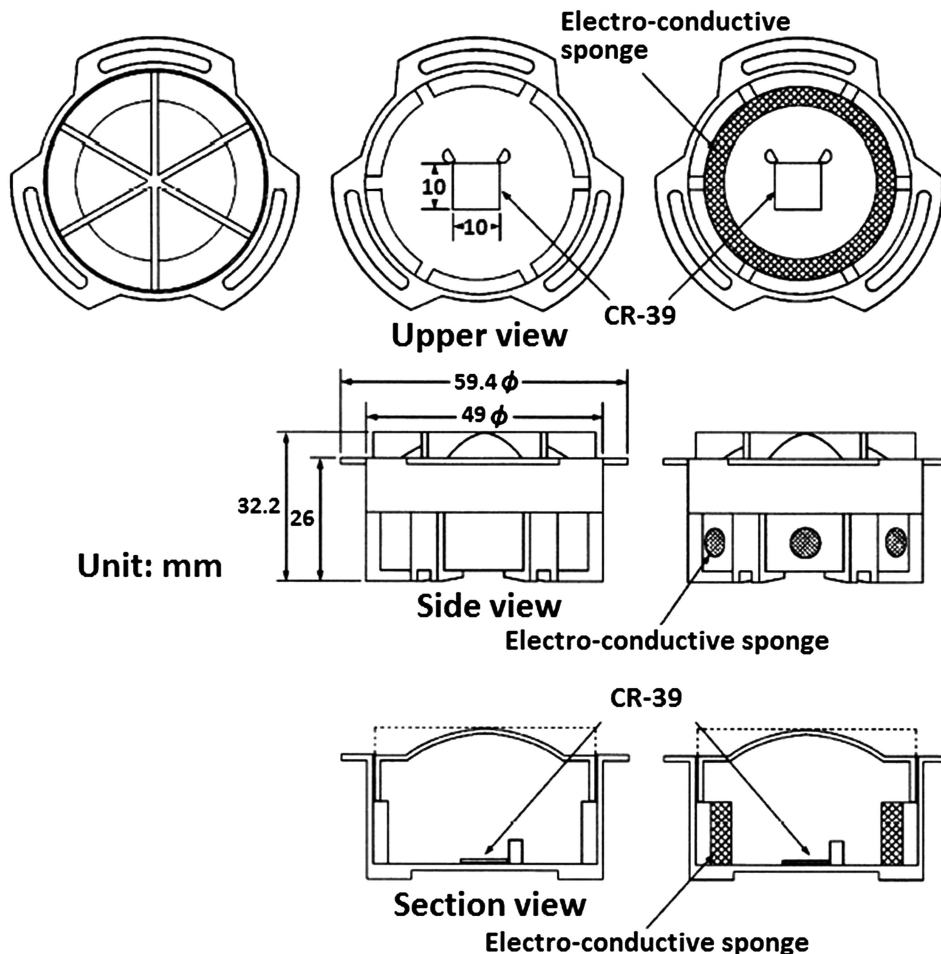


Fig. 2. Schematic drawings of the passive type radon and thoron discriminative detector (RADUET) [19].

the obtained radon activity concentrations were not influenced by thoron in the measurement place.

$$N_L = X_{Rn} \cdot CF_{Rn1} \cdot T + X_{Tn} \cdot CF_{Tn1} \cdot T + B \quad (1)$$

$$N_H = X_{Rn} \cdot CF_{Rn2} \cdot T + X_{Tn} \cdot CF_{Tn2} \cdot T + B \quad (2)$$

Here, N_L and N_H are alpha track densities (tracks cm^{-2}) for the low and the high air-exchange-rate chambers, respectively. These values were obtained by using the automatic track reading system. X_{Rn} and X_{Tn} are the respective mean activity concentrations (Bq m^{-3}) of radon and thoron during the exposure period in this study. CF_{Rn1} and CF_{Tn1} are the respective conversion factors ($\text{cm}^{-2} \text{ kBq}^{-1} \text{ m}^3 \text{ h}^{-1}$) from alpha track densities to radon and thoron activity concentrations for the low air-exchange-rate chamber. CF_{Rn2} and CF_{Tn2} are the respective conversion factors ($\text{cm}^{-2} \text{ kBq}^{-1} \text{ m}^3 \text{ h}^{-1}$) from alpha track densities to radon and thoron activity concentrations for the high air-exchange-rate chamber. The respective conversion factors were evaluated from the calibration experiments in the radon and thoron calibration chambers at NIRS. T is the exposure time (h), and B is the background alpha track density (tracks cm^{-2}) on the CR-39 detector.

RESULTS AND DISCUSSION

Radon activity concentrations by AlphaGUARD and RADUET

The temporal variation of indoor radon activity concentration measured in the clinic using the AlphaGUARD is shown in Fig. 3A. The maximum value of 41 Bq m^{-3} was observed at 8:00 (JST) on 19 November 2012. The arithmetic mean and the standard deviation for the observed results were evaluated as 15 and 7 Bq m^{-3} , respectively. Also for the clinic, the mean daily variation of indoor radon activity concentrations for each measurement month are shown in Fig. 3B. The data exhibit the general pattern reported in numerous studies [20–22]. The maximum and minimum values of each measurement month were observed in the morning (around 9:00 JST) and in the afternoon (around 15:00 JST), respectively. It is well-known that the daily variation of indoor radon activity concentration depends on the human behavior. Usually, the reception hours of the clinic are from 9:00 (JST) to 16:00 (JST). Many patients come and go frequently from the entrance of the clinic during these hours. Moreover, the clinic staff members and salespersons are using the back entrance. Thus, indoor radon activity concentration might be decreased during the reception hours due to the human behavior. On the other hand, radon which was exhaled from the soil and walls might begin to accumulate in the room after the staff working hours.

Radon activity concentrations for temporary housing units, including the clinic, apartments and detached houses, are summarized in Table 1A. The arithmetic mean and the standard deviation of the indoor radon activity concentrations obtained by the RADUETs at the temporary housing units, apartments and detached houses were 5 ± 3 , 7 ± 3 and $9 \pm 8 \text{ Bq m}^{-3}$, respectively. These values included data for the lower limit of detection (referred to as not detected, ND) of radon activity concentration, which was evaluated to be $\sim 3 \text{ Bq m}^{-3}$. The difference in indoor radon concentration due to the type of housing was not statistically significant. The maximum value and the standard uncertainty of $28 \pm 2 \text{ Bq m}^{-3}$ was observed for one detached house. According to Suzuki *et al.* [15], the arithmetic mean of the indoor radon activity concentrations in Fukushima Prefecture

dwelling after adjusting for seasonal fluctuation was $\sim 10 \text{ Bq m}^{-3}$. In the present study, the numbers (percentages) of ND data for radon activity concentration for each house type were: temporary housing units, seven (28%); apartments, one (7%); and detached houses, three (27%). According to Sanada *et al.* [23], although they had only six data, indoor radon activity concentrations in the prefabricated houses were lower than those in other houses. Moreover, they observed a lower seasonal variation of indoor radon activity concentrations in the prefabricated houses compared with in other houses. In addition, the radon exhalation rate from building materials is well-known as an important influential parameter affecting the indoor radon activity concentration [24]. Decorated gypsum boards are used as wall materials for most of the temporary housing units. According to previous reports [25–28], radon exhalation rates from the gypsum boards were relatively lower than those from other building materials. Although there is no information about the kind of building materials for the detached houses, the results obtained in this study suggest that some of the detached houses are possibly using building materials which include low level ^{226}Ra activity. Radon exhalation rates from building materials of the temporary housing units and the detached houses might be lower than those from the apartments. Furthermore, the average values of indoor radon activity concentrations obtained in this study were at a similar level to the reported average value of outdoor radon activity concentration in Fukushima Prefecture: 6.3 Bq m^{-3} [29]. Outdoor radon gases might enter the temporary housing units due to their low air-tightness. Based on the surface geological map of Fukushima Prefecture [30], loam, granite and black schists are distributed around Nihonmatsu City. The radon exhalation rate from the soil surface, which is one of the influential parameters for outdoor and indoor radon activity concentrations, depends on the surface geology [31, 32]. Although the radon exhalation rates were not measured around the measurement points of the indoor radon activity concentrations, the entry of radon exhaled from the soil surface into the residences cannot be completely ruled out.

On the other hand, the ranges of indoor thoron activity concentrations obtained by the RADUETs at the temporary housing units, apartments and detached houses were ND–16, ND–35 and ND–39 Bq m^{-3} , respectively. The lower limit of detection for thoron activity concentration was reached for 28 residences (56%). The lower limit of detection for thoron activity concentration depends on the radon activity concentration at the measurement place, and the lower limit of detection values in the present study ranged from 3 to 26 Bq m^{-3} . Thus, the arithmetic means and standard deviations at each house type were not summarized in Table 1. It is well known that the indoor thoron activity concentration depends on the distance from the wall surface [33, 34]. Since the RADUETs were installed at different distances from the wall surface in each location, the annual effective dose for thoron at each residence was not estimated in this study.

Annual effective doses for radon were evaluated using the following equation, which was reported by UNSCEAR [35]:

$$E = \{(Q \cdot F \cdot T \cdot K)_{\text{indoor}} + (Q \cdot F \cdot T \cdot K)_{\text{outdoor}}\}. \quad (3)$$

Here, Q is the radon activity concentration (Bq m^{-3}), F is an equilibrium factor (indoors, 0.4; outdoors, 0.6), K is the dose coefficient ($9 \times 10^{-6} \text{ mSv/Bq h m}^{-3}$) [11] and T is the occupancy time (h). The indoor and outdoor occupancy factors for adults were,

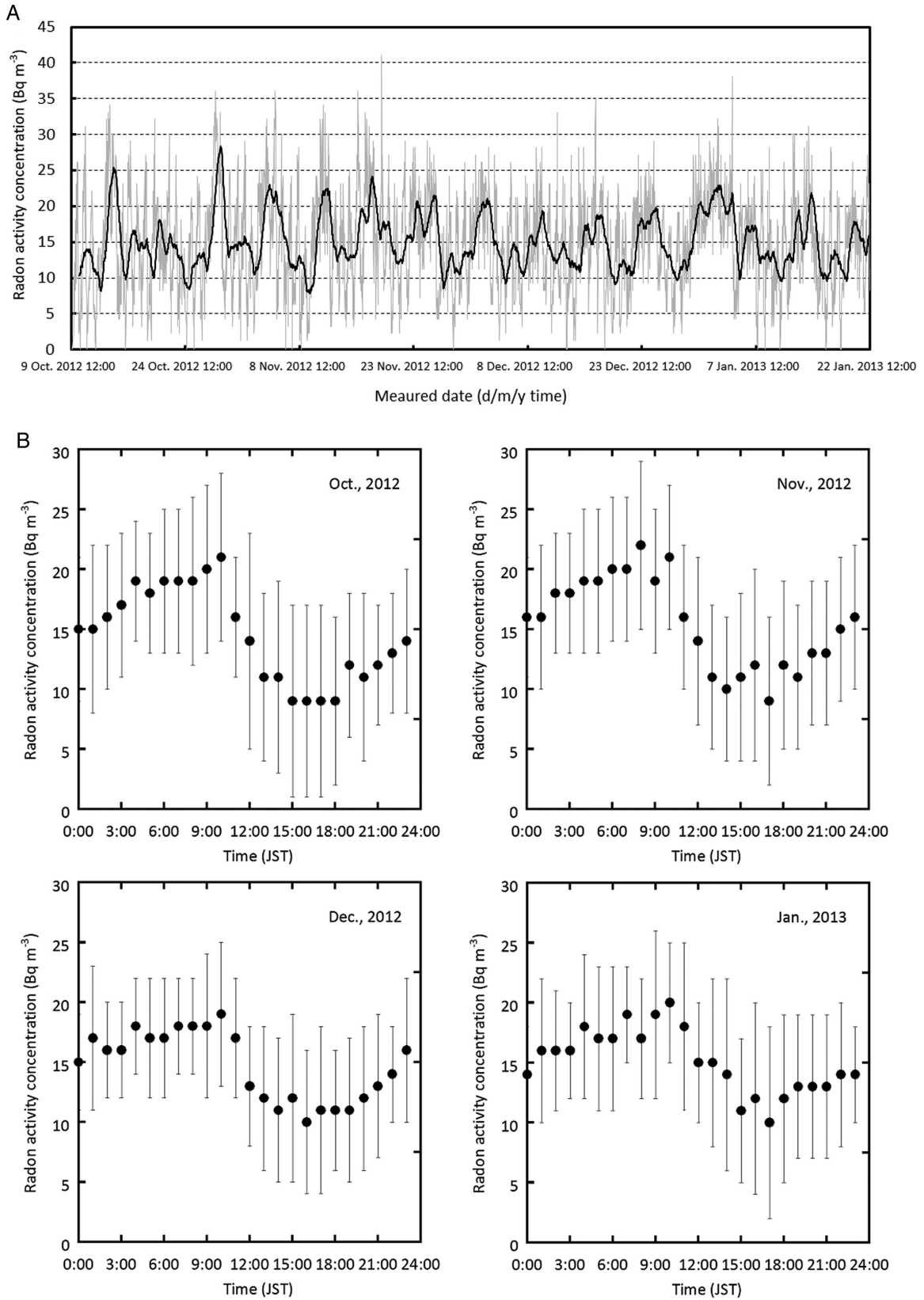


Fig. 3. (A) Temporal variation of indoor radon activity concentration, measured using the AlphaGUARD placed in the medical clinic within a temporary housing facility. Black line shows the moving average of 24 h. (B) The daily variation of indoor radon activity concentration in the clinic in October, November, and December 2012, and January 2013.

Table 1. Indoor radon activity concentrations by RADUET (A) and annual effective doses by inhaled radon (B) for temporary housing units and Tsushima Clinic, apartments and detached houses

(A) Type of residence	Number of residences	Radon activity concentration ^a (Bq m ⁻³)	Maximum (Bq m ⁻³)	Minimum (Bq m ⁻³)
Temporary housing unit and Tsushima Clinic	25	5 ± 3	11	<3 (ND)
Apartment	14	7 ± 3	10	<3 (ND)
Detached house	11	9 ± 8	28	<3 (ND)
(B) Type of residence	Number of residences	Annual effective dose ^a (mSv)	Maximum ^b (mSv)	Minimum ^c (mSv)
Temporary housing unit and Tsushima Clinic	25	0.18 ± 0.07	0.36	<0.12 (ND)
Apartment	14	0.22 ± 0.07	0.33	<0.12 (ND)
Detached house	11	0.29 ± 0.24	0.83	<0.12 (ND)

^aThese values are indicated as average ± standard deviation (1σ). ^bThese values were based on the maximum measured values. ^cThese values were based on the minimum measured values.

respectively, set as 0.9 and 0.1 for an indoor worker or a pensioner, according to the report by UNSCEAR [14]. The outdoor radon activity concentration in Fukushima Prefecture has been reported as 6.3 Bq m⁻³ [29]. The indoor radon activity concentrations are usually higher than the outdoor values. Although some of the indoor radon activity concentrations that were obtained in this study were lower than the average value for the outdoor one, the published average value of outdoor radon activity concentration in Fukushima Prefecture was used for the dose estimation. The seasonal correction factors for indoor radon activity concentration have been reported by researchers for European countries [36–40]. The average values of the reported seasonal correction factors from October to January were widely distributed from 0.80 to 0.95. However, no factors for Japanese houses have been published. Thus, dose estimation in this study was carried out without considering any seasonal correction factor for Japanese houses.

The annual effective doses by inhaled radon in temporary housing facilities, apartments and detached houses are summarized in Table 1B. The annual effective dose due to indoor radon varied from <0.12 (ND) to 0.83 mSv, and the average with standard deviation was estimated to be 0.22 ± 0.12 mSv. Moreover, the averages of the annual effective dose for each type of residence were 0.18, 0.22 and 0.29 mSv, respectively. The average values of indoor radon activity concentrations for wooden, concrete and prefabricated houses in winter were read from Fig. 4 in reference 23, and these values were respectively about 1.3, 1.2 and 1.1 times higher than those for all seasons. Thus, the estimated effective doses in this study might be overestimated by about 10–30%. However, these doses are all lower than the reported Japanese average: 0.45 mSv (15.5 Bq m⁻³) [23].

A comparison of the effective dose by indoor radon and the effective dose caused by the FDNPP accident

The nationwide absorbed dose rates in the air in Japan were measured by NIRS from 1967 to 1977 in a nationwide external dose estimation project [41]. The average of the absorbed dose rate in air for

Fukushima Prefecture was reported as 53 nGy h⁻¹ ($n = 28$) in this project, and this value was similar to the nationwide average [42], which was determined as 50 nGy h⁻¹, also before the accident. The dose rate in Namie Town had the highest value (77 nGy h⁻¹) in Fukushima Prefecture due to the distribution of weathered granite as bedrock [41]. On the other hand, the minimum value in Fukushima Prefecture was reported as 34 nGy h⁻¹ at Sukagawa City [41], and that was 44% of the value at Namie Town. The dose rates at Fukushima City and Nihonmatsu City were reported as 45 nGy h⁻¹ and 48 nGy h⁻¹, respectively [41]. Thus, the external dose due to natural radionuclides has decreased by ~40% for Namie Town evacuees who have been living in Fukushima City and Nihonmatsu City in the present study.

The annual external dose due to natural radionuclides such as ²³⁸U and ²³²Th series and ⁴⁰K was estimated using the average of the absorbed dose rate in air in Fukushima Prefecture (53 nGy h⁻¹) multiplied by the dose conversion factor and occupancy time. The dose conversion factor for converting from absorbed dose to effective dose was reported as 0.748 Sv Gy⁻¹ by Moriuchi *et al.* [43]. The occupancy time was set as 8760 h (24 h × 365 d) in this estimation. It is necessary to consider the indoor to outdoor dose rate ratio to estimate the external dose in houses based on an outdoor dose rate. A few papers have reported the ratios of indoor to outdoor dose rates for Japanese buildings [44–46]. Abe *et al.* [44] reported that the indoor dose rates were similar to the outdoor dose rates for wooden dwellings. Matsuda *et al.* [45] reported that the mean ratios (standard deviation) of indoor/outdoor dose rates for reinforced concrete dwellings, fireproof wooden dwellings and light gauge steel dwellings were 0.95 (0.15), 0.77 (0.10) and 0.72 (0.13), respectively. Iyogi *et al.* [46] reported that the mean ratios for stucco-covered walls and other walls were 1.6 ± 0.3 and 1.4 ± 0.27, respectively. Moreover, the activity concentrations of natural radionuclides (which are the sources of the indoor absorbed dose rates in air) depend on the building materials [24]. These facts suggest that it is difficult to set one value for the indoor to outdoor dose rate ratio. Thus, in this study, an external

dose estimation such as in the authors' previous studies [47, 48] was made, excluding the occupancy factor and the indoor to outdoor dose rate ratio due to building materials. The average and the range of annual external doses due to natural radionuclides in Fukushima Prefecture were estimated to be 0.35 and 0.22–0.50 mSv, respectively. The average and the range of annual effective dose due to indoor radon in this study were also estimated to be 0.21 and ND–0.89 mSv, respectively. Fujimoto and O'Brien [49] calculated the population-weighted average of the ambient dose equivalent rate due to secondary cosmic-rays in Fukushima Prefecture as 0.33 mSv. Also, the Japanese average of the committed effective dose due to natural radionuclides as a result of the intake of dietary foods has been evaluated as 0.98 mSv [50]. In particular, 0.73 mSv of this total was estimated to come from ^{210}Po , a radionuclide which occurs naturally in marine products. No data for the dose from intake of dietary foods in Fukushima Prefecture have been published. Thus, the Japanese average of the committed effective dose was used for the dose estimation. The total estimated annual effective dose (the average value from natural radionuclides) in Fukushima Prefecture in this study was 1.9 mSv, and this value was not the result of the FDNPP accident. According to the report by the Nuclear Safety Research Association [51], the annual effective dose from natural radiation in Japan has been estimated to be 2.1 mSv. The estimated value of 1.9 mSv in this study and the Japanese average are similar (~ 2 mSv), and the slight difference is attributed to uncertainties in the dose estimations.

On the other hand, some papers have been published on the thyroid equivalent dose of Fukushima Prefecture residents and short-term visitors [4, 52, 53]. Tokonami *et al.* [52] reported that the maximum thyroid dose due to inhalation of ^{131}I for the evacuees from Namie Town and Minami-soma City was 33 mSv among 62 persons from 0 to 83 years old. Matsuda *et al.* [53] measured ^{131}I activity concentrations for evacuees and short-term visitors to Fukushima Prefecture using WBC measurements, and the maximum thyroid dose was calculated to be 20 mSv. Kamada *et al.* [54] collected urine samples from residents in Kawamata Town and Iitate Village and found that the thyroid doses for the residents were from 27 to 66 mSv. The present authors [4] estimated that the maximum internal exposure of the thyroid to ^{131}I on the basis of ^{134}Cs accumulated in the body, as measured by the WBC, was 18 mSv. Kim *et al.* [55] estimated that the maximum thyroid dose to 1080 children residing in Fukushima Prefecture was 58 mSv. The results of a screening examination carried out by the Nuclear Emergency Response Local Headquarters were used for their estimation. The maximum thyroid doses ranged from 18 to 66 mSv. Since these values were given as equivalent dose, it is necessary to convert these to effective dose to allow comparison with other exposures; this was done using the tissue-weighted factor ($=0.04$) reported by ICRP Publication 103 [56]. Effective doses were calculated to be 0.7–2.6 mSv.

A program by Fukushima Prefecture authorities estimated the internal dose due to intake of radiocesium using WBC measurements for more than 150 000 residents (as of September 2013), and most of the estimated doses (99.98%) were <1 mSv in terms of the committed effective dose [57]. Tsubokura *et al.* [6] estimated internal dose using the WBC for more than 9000 residents in Minami-soma City, and the maximum committed effective dose was estimated to be ~ 1.1 mSv. Harada *et al.* [8] estimated annual effective dose due to dietary intake of radiocesium, and their maximum estimated value in Fukushima Prefecture

was ~ 0.10 mSv. This estimation assumed that the daily intake of radiocesium (^{134}Cs and ^{137}Cs) was constant throughout the year.

If the average of the effective dose in Fukushima Prefecture from natural radionuclides due to intake of dietary foods was assumed to be the same as the Japanese average of 0.98 mSv, the effective dose due to internal exposure for the evacuees was evaluated to be 1.2 mSv, considering the average effective dose of 0.22 mSv from radon as obtained in this study. Thus, the maximum effective dose for ^{131}I was about two times higher than that for dietary foods and radon. On the other hand, the maximum committed effective dose for radiocesium was observed as a similar value to the maximum effective dose for dietary foods and radon.

The external doses for the first four months after the accident (11 March to 11 July 2011) for 421 394 residents in Fukushima Prefecture were estimated by the Fukushima Health Management Survey, and the effective dose distributions for the residents were: <1 mSv, 62.0%; <2 mSv, 94.0%; <3 mSv, 99.4%; and the mean and maximum values were 0.8 and 25 mSv, respectively [58]. Thus, the external dose to almost all residents was estimated at <3 mSv, which was about six times higher than that for the maximum external dose due to natural sources in Fukushima Prefecture.

According to the UNSCEAR 2013 report [14], the estimated settlement average effective dose to adults evacuated from Namie Town for the first year was 5.0 mSv. Although the dose estimation methods of UNSCEAR and this study were different, those average effective doses were used in the following comparison to gain some perspective on the overall exposure of the evacuees. The total average effective dose (including natural sources) to residents for the first year was found to be ~ 6.9 mSv, and 28% of the total effective dose came from natural sources. Moreover, the contribution of the effective dose from radon was $\sim 3\%$ of the total exposure. However, the most recent nationwide indoor radon survey [15] found the indoor radon activity concentration in Fukushima Prefecture exceeded 100 Bq m^{-3} . In this case, the annual effective dose caused by inhaled radon corresponded to >3 mSv.

The radon activity concentrations obtained in this study did not exceed the reference level reported by the WHO. These results suggest that action to decrease the amount of radon in the temporary housing facilities is not necessary. Although it was difficult to estimate the uncertainty of the effective doses due to natural and artificial radionuclides, the maximum effective doses for inhaled ^{131}I and ingested radiocesium for the first year were about four times higher than that for inhaled radon. However, this difference would be much decreased two years after the FDNPP accident. Thus, the dose contribution from natural sources will increase due to the almost constant natural radiation dose. That is, residents' annual internal and external exposure due to natural radionuclides (estimated as 1.9 mSv in this study) cannot be avoided. On the other hand, it might be possible to reduce the external exposure due to the artificial radionuclides by changing some behaviors of residents, for instance by using information about the spatial distribution of levels of potential external exposure, external exposure could be reduced by either avoiding certain areas or minimizing time spent in those areas.

ACKNOWLEDGEMENTS

The authors would like to thank Dr. Shunji Sekine and Mr. Norio Konno at the Tsushima Clinic for their kind assistance in carrying out this study.

FUNDING

This work was partly supported by a Hirosaki University Grant for Exploratory Research by Young Scientists (2012). Funding to pay the Open Access publication charges for this article was provided by an individual research grant from Hirosaki University to Prof. Shinji Tokonami.

REFERENCES

- Hosoda M, Tokonami S, Sorimachi A, et al. The time variation of dose rate artificially increased by the Fukushima nuclear crisis. *Sci Rep* 2011;1:87.
- Hosoda M, Tokonami S, Tazoe H, et al. Activity concentrations of environmental samples collected in Fukushima Prefecture immediately after the Fukushima nuclear accident. *Sci Rep* 2013;3:2283.
- Tokonami S, Hosoda M, Iwaoka K, et al. Outline of the recovery support project for the great east Japan earthquake to Namie Town, Fukushima Prefecture. *Jpn J Health Phys* 2015;50:11–9 (in Japanese).
- Hosoda M, Tokonami S, Akiba S, et al. Estimation of internal exposure of the thyroid to ^{131}I on the basis of ^{134}Cs accumulated in the body among evacuees of the Fukushima Daiichi Nuclear Power Station accident. *Environ Int* 2013;61:73–6.
- Momose T, Takada C, Nakagawa T, et al. Whole-body counting of Fukushima residents after the TEPCO Fukushima Daiichi Nuclear Power Station accident. *Proceedings of the 1st NIRS Symposium on Reconstruction of Early Internal Dose in the TEPCO Fukushima Daiichi Nuclear Power Station Accident* 2012. NIRS-M-252, 67–82.
- Tsubokura M, Gilmour S, Takahashi K, et al. Internal radiation exposure after the Fukushima Nuclear Power Plant disaster. *JAMA* 2012;308:669–700.
- Hayano RS, Tsubokura M, Miyazaki M, et al. Internal radiocesium contamination of adults and children in Fukushima 7 to 20 months after the Fukushima NPP accident as measured by extensive whole-body-counter surveys. *Proc Jpn Acad Ser B Phys Biol Sci* 2013;89:157–63.
- Harada K, Fujii Y, Adachi A, et al. Dietary intake of radiocesium in adult residents in Fukushima Prefecture and neighboring regions after the Fukushima Nuclear Power Plant accident: 24-h food-duplicate survey in December 2011. *Environ Sci Technol* 2013;47:2520–6.
- Nihei N, Tanoi K, Nakanishi T. Inspections of radiocesium concentration levels in rice from Fukushima Prefecture after the Fukushima Dai-ichi Nuclear Power Plant accident. *Sci Rep* 2015;5:8653.
- Ishikawa T. A brief review of dose estimation studies conducted after the Fukushima Daiichi Nuclear Power Plant accident. *Radiat Emerg Med* 2014;3:21–7.
- United Nations Scientific Committee on the Effects of Atomic Radiation. *Sources and Effects of Ionizing Radiation. UNSCEAR 2008 Report, Volume I. Report to the General Assembly with Scientific Annexes*. http://www.unscear.org/docs/reports/2008/09-86753_Report_2008_Annex_B.pdf (23 October 2015, date last accessed).
- World Health Organization. WHO Handbook on Indoor Radon – A Public Health Perspective, Geneva, Switzerland. http://whqlibdoc.who.int/publications/2009/9789241547673_eng.pdf (23 October 2015, date last accessed).
- Darby S, Hill D, Auvinen A, et al. Radon in homes and risk of lung cancer: collaborative analysis of individual data from 13 European case-control studies. *BMJ* 2005;330:223–6.
- United Nations Scientific Committee on the Effects of Atomic Radiation. *Sources, Effects and Risks of Ionizing Radiation. UNSCEAR 2013 Report, Volume I. Report to the General Assembly with Scientific Annexes A: Levels and Effects of Radiation Exposure Due to the Nuclear Accident after the 2011 Great East-Japan Earthquake and Tsunami*. http://www.unscear.org/docs/reports/2013/13-85418_Report_2013_Annex_A.pdf (23 October 2015, date last accessed).
- Suzuki G, Yamaguchi I, Ogata H, et al. A nation-wide survey on indoor radon from 2007 to 2010 in Japan. *J Radiat Res* 2010;51:683–9.
- Ishimori Y, Ishikawa T, Tokonami S. Radon intercomparison experiment at PTB in Germany. *Jpn J Health Phys* 2005;39:263–7.
- Ichitsubo H, Yamada Y, Shimo M, et al. Development of a radon-aerosol chamber at NIRS-general design and aerosol performance. *J Aerosol Sci* 2004;35:217–32.
- Tokonami S. Thoron in the environment and its related issues. *Indian J Phys* 2009;83:777–85.
- Tokonami S, Takahashi H, Kobayashi Y, et al. Up-to-date radon-thoron discriminative detector for a large scale survey. *Rev Sci Instrum* 2005;76:113505.
- Sorimachi A, Kranrod C, Tokonami S, et al. Anomalously high radon concentrations in a dwelling in Okinawa, Japan. *Radioisotopes* 2009;58:807–13.
- Inoue K, Hosoda M, Tokonami S, et al. Investigation of radon and thoron concentrations in a landmark skyscraper in Tokyo. *J Radioanal Nucl Chem* 2013;298:2009–15.
- Porstendörfer J, Butterweck G, Reineking A. Daily variation of the radon concentration indoors and outdoors and the influence of meteoroidal parameters. *Health Phys* 1994;67:283–7.
- Sanada T, Fujimoto K, Miyano K, et al. Measurement of nationwide indoor Rn concentration in Japan. *J Environ Radioact* 1999;45:129–37.
- Iwaoka K, Hosoda M, Tabe H, et al. Activity concentration of natural radionuclides and radon and thoron exhalation rates in rocks used as decorative wall coverings in Japan. *Health Phys* 2013;104:41–50.
- Keller G, Hoffmann B, Feigenspan T. Radon permeability and radon exhalation of building materials. *Sci Total Environ* 2001;272:85–9.
- Ujić P, Čeliković I, Kandić A, et al. Internal exposure from building materials exhaling ^{222}Rn and ^{220}Rn as compared to external exposure due to their natural radioactivity content. *Appl Radiat Isot* 2010;68:201–6.
- Chen J, Rahman NM, Atiya IA. Radon exhalation from building materials for decorative use. *J Environ Radioact* 2010;101:317–22.

28. Saad AF, Al-Awami HH, Hussein NA. Radon exhalation from building materials used in Libya. *Radiat Phys Chem* 2014;101:15–9.
29. Oikawa S, Kanno N, Sanada T, et al. A nationwide survey of indoor radon concentration in Japan. *J Environ Radioact* 2006;87:239–45.
30. National Land Information Division, National Planning and Regional Policy Bureau, Ministry of Land, Infrastructure, Transport and Tourism. Tokyo, Japan. <http://nrb-www.mlit.go.jp/kokjo/tochimizu/F2/MAP/207010.pdf>. (23 October 2015, date last accessed).
31. Hosoda M, Shimo M, Sugino M, et al. *In situ* measurements of radon and thoron exhalation rates and their geological interpretation. *Jpn J Health Phys* 2004;39:206–14 (in Japanese with English abstract).
32. Hosoda M, Shimo M, Sugino M, et al. Radon and thoron exhalation rate map in Japan. In: Paschoa AS, Steinhäusler F (eds). *The Natural Radiation Environment, AIP Conference Proceedings 1034*. New York: American Institute of Physics, 2008, 177–80.
33. Doi M, Fujimoto K, Kobayashi S, et al. Spatial distribution of thoron and radon concentrations in the indoor air of a traditional Japanese wooden house. *Health Phys* 1994;66:43–9.
34. Tokonami S. Why is ^{220}Rn (thoron) measurement important? *Radiat Prot Dosimetry* 2010;141:335–9.
35. United Nations Scientific Committee on the Effects of Atomic Radiation. *UNSCEAR 2000 Report to the General Assembly with Scientific Annexes: United Nations*. <http://www.unscear.org/docs/reports/annexb.pdf> (23 October 2015, data last accessed).
36. Pine J, Fearn T, Darby SC, et al. Seasonal correction factors for indoor radon measurements in the United Kingdom. *Radiat Prot Dosimetry* 1995;58:127–32.
37. Baysson H, Billon S, Laurier D, et al. Seasonal correction factors for estimating radon exposure in dwellings in France. *Radiat Prot Dosimetry* 2003;104:245–52.
38. Burke Ó, Long S, Murphy P, et al. Estimation of seasonal correction factors through Fourier decomposition analysis—a new model for indoor radon levels in Irish homes. *J Radiol Prot* 2010;30:433–43.
39. Burke Ó, Murphy P. Regional variation of seasonal correction factors for indoor radon levels. *Radiat Meas* 2011;46:1168–72.
40. Kozak K, Mazur J, Kozłowska B, et al. Correction factors for determination of annual average radon concentration in dwellings of Poland resulting from seasonal variability of indoor radon. *Appl Radiat Isot* 2011;69:1459–65.
41. Abe S. Efforts to obtain Japanese profile of ambient natural radiation exposure. *Jpn J Health Phys* 1982;17:169–93 (in Japanese).
42. Furukawa M, Shingaki R. Terrestrial gamma radiation dose rate in Japan estimated before the 2011 Great East Japan Earthquake. *Radiat Emer Med* 2012;1:11–6.
43. Moriuchi S, Tsutsumi M, Saito K. Examination on conversion factors to estimate effective dose equivalent from absorbed dose in air for natural gamma radiations. *Jpn J Health Phys* 1990;25:121–8 (in Japanese).
44. Abe S, Fujimoto K, Fujitaka K. Relationship between indoor and outdoor gamma ray exposure in wooden houses. *Radiat Prot Dosimetry* 1984;7:267–9.
45. Matsuda H, Fukaya M, Minato S. Measurements of indoor and outdoor natural radiation exposure rates in model houses. *Jpn J Health Phys* 1990;25:385–90 (in Japanese).
46. Iyogi T, Ueda S, Hisamatsu S, et al. Environmental gamma-ray dose rate in Aomori prefecture, Japan. *Health Phys* 2002;82:521–6.
47. Hosoda M, Tokonami S, Omori Y, et al. Estimation of external dose by car-borne survey in Kerala, India. *PLoS ONE* 2015;10:e0124433.
48. Inoue K, Hosoda M, Fukushi M, et al. Absorbed dose rate in air in metropolitan Tokyo before the Fukushima Daiichi Nuclear Power Plant accident. *Radiat Prot Dosimetry* 2015;167:231–4.
49. Fujimoto K, O'Brien K. Estimation of dose due to cosmic rays in Japan. *Jpn J Health Phys* 2002;37:325–34 (in Japanese).
50. Ota T, Sanada T, Kashiwara Y, et al. Evaluation for committed effective dose due to dietary foods by the intake for Japanese adults. *Jpn J Health Phys* 2009;44:80–8.
51. Nuclear Safety Research Association. *Radiation Dose Received in Daily Life*. Tokyo: Nuclear Safety Research Association, 2011.
52. Tokonami S, Hosoda M, Akiba S, et al. Thyroid doses for evacuees from the Fukushima nuclear accident. *Sci Rep* 2012;2:507.
53. Matsuda N, Kumagai A, Ohtsuru A, et al. Assessments of internal exposure doses in Fukushima by a whole body counter within one month after the Fukushima Power Plant accident. *Radiat Res* 2013;179:663–8.
54. Kamada N, Saito O, Endo S, et al. Radiation doses among residents living 37 km northwest of the Fukushima Dai-ichi Nuclear Power Plant. *J Environ Radioact* 2012;110:84–9.
55. Kim E, Kurihara O, Tani K, et al. (16 May 2015) Intake ratio of ^{131}I to ^{137}Cs derived from thyroid and whole-body doses to Fukushima residents. *Radiat Prot Dosimetry*, .
56. International Commission on Radiological Protection. *The 2007 Recommendations of the International Commission on Radiological Protection. ICRP Publication 103*. Oxford, UK: Elsevier, 2008.
57. Miyazaki M, Ohtsuru A, Ishikawa T. An overview of internal dose estimation using whole-body counters in Fukushima Prefecture. *Fukushima J Med Sci* 2014;60:1–6.
58. Ishikawa T, Yasumura S, Ozasa K, et al. The Fukushima Health Management Survey: estimation of external doses to residents in Fukushima Prefecture. *Sci Rep* 2015;5:12712.