Regular Article

Characteristics of the Environmental Radon and Thoron in Minamidaito-jima, a Comparatively High Background Radiation Island in Japan

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General survey for the environmental radon and thoron has been performed in Minamidaito-jima, Okinawa prefecture, southwestern part of Japan. *In situ* measurements of the radon and thoron exhalation rates were conducted at a total of 7 points using an accumulation technique with a ZnS(Ag) scintillation detector. The medians of radon and thoron exhalation rates were estimated to be 31 mBq m^{-2} s^{-1} and 3,970 mBq m^{-2} s^{-1}, respectively. The atmospheric radon concentrations were measured at 2 points with an ionization chamber, and the maximum concentration was estimated to be 94 Bq m^{-3}. For the surface soil samples collected at the measurement points of the exhalation rates, arithmetic means for ^{238}U and ^{232}Th series concentration were estimated to be approximately 145 Bq kg^{-1} dry and 101 Bq kg^{-1} dry, respectively. These results indicated that the environmental radon and thoron levels in Minamidaito-jima are fairly higher than those of the average of Japan. And it is strongly suggested that the exhalation rates and the concentrations are enhanced mainly due to the surface soil with the relatively high radioactivity.

Key words: radon and thoron exhalation rates, *in situ* measurement, atmospheric radon concentration, ^{238}U and ^{232}Th series concentrations, Minamidaito-jima, Okinawa, Japan

1. Introduction

Relatively high background radiation levels are recognized in the several places of Okinawa prefecture, subtropical region of southwest Japan (Fig. 1). For instance, fairly high atmospheric radon concentrations were measured in Okinawa-jima and Miyako-jima[1-3]. Also in these islands, radon (^{222}Rn) and thoron (^{220}Rn) exhalation rates from the soil surface are approximately two times higher than those of the mainland of Japan[4-6]. In addition, the dose rate in air due to terrestrial gamma radiation in Miyako-jima and Daito Islands is generally higher than that of the average of all over Japan[7-9]. And then, these studies strongly suggest that the dark

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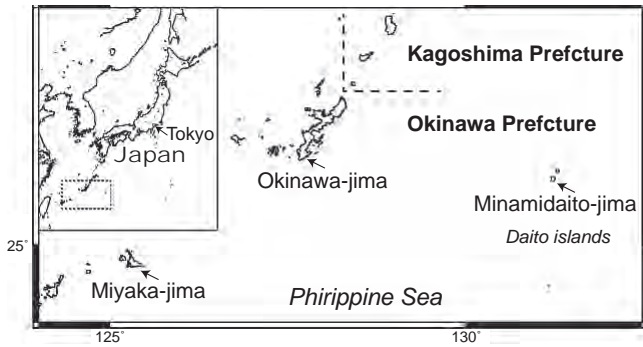E-mail: k128606@eve.u-ryukyu.ac.jp

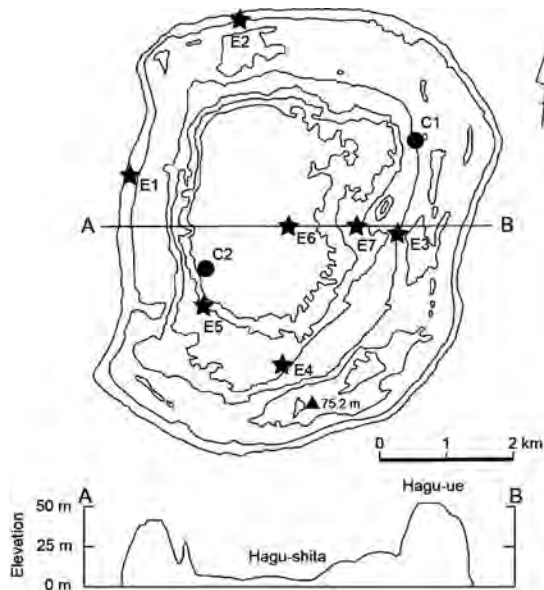**Fig. 1.**　Map showing the location of Minamidaito-jima.

Fig. 2.　Topography of Minamidaito-jima.　★ : measurement point of radon and thoron exhalation rates and sampling point of soil.
● : measurement point of outdoor radon concentration.

Fig. 3.　Schematic diagram of the simultaneous *in situ* measurement system for radon and thoron exhalation rates from the ground surface[22].

Table 1.　Radon and thoron exhalation rates from the soil surface. Locations of the measurement points are shown in Figure 2

Point	Exhalation rate ± S.D. (mBq m^{-2} s^{-1})	
	Radon	Thron
E 1	9 ± 2	3,970 ± 723
E 2	29 ± 5	2,570 ± 485
E 3	137 ± 14	5,530 ± 988
E 4	*ND* (< 8)	3,200 ± 594
E 5	33 ± 5	3,440 ± 634
E 6	24 ± 4	4,510 ± 816
E 7	33 ± 5	4,650 ± 840
Arithmetic mean ± S.D.	44 ± 46	3,980 ± 927

red soil, so-called Shimajiri-maji in Okinawan word, widely distributed in the prefecture is an enhancer for the background radiation level[2, 5, 6, 8, 9]. However, the environmental radon and thoron studies were not enough about in the prefecture.

　In this study, as a part of the above researches to estimate the detail for the background radiation level in Okinawa prefecture, *in situ* measurements of radon and thoron exhalation rates from the soil surface and atmospheric radon concentration, and analyses of natural radionuclides for the soils were carried out intensively in Minamidaito-jima located 360 km east off Okinawa-jima (Fig. 1). Minamidaito-jima is a coral island, uplifted atoll, with 30.6 km^2 in area and the maximum altitude of 75.2 m. The island is topographically divided into a doughnut-shaped upland and a central round basin (Fig. 2). The upland and the basin are called Hagu-ue and Hagu-shita in localism, respectively. Emerged age of the atoll is
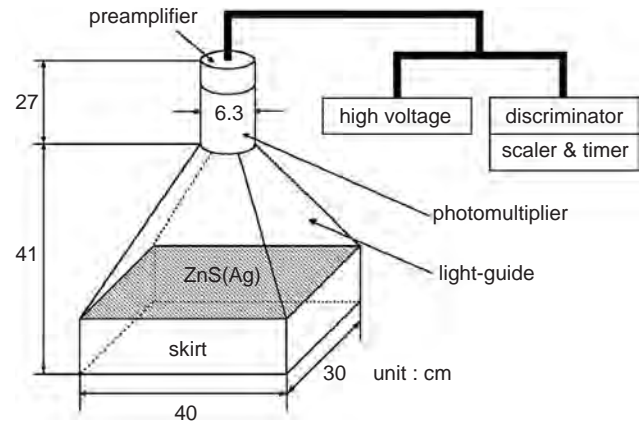
presumed to be 1.2-1.6 million years ago[10]. In the process of and after the emergence, the island was covered with eolian dust from the Eurasian high background radiation area mainly before the Last Glacial Age[8, 9, 11, 12]. The eolian dust is considered the main material of Shimajiri-maji[12, 13] with thickness of 1-2 m for Hagu-ue and 2-4 m for Hagu-shira[14].

2. Method

2.1. In situ Measurements of Radon and Thoron Exhalation Rates from Soil Surface

Radon and thoron exhalation rates were measured with an *in situ* type instrument (Fig. 3)[15, 16]. Because the detail of instrument has been described previously[17-22], an outline is provided here. The instrument is composed of accumulation chamber (skirt section) for covers the ground, scintillation detector with aluminized Mylar sheet (a pattern of slanted lines section in Figure 3), light guide, photomultiplier tube, pulse counting part, scaler and timer. The area of large acrylic board coated with a ZnS(Ag) scintillator is 1,200 cm^2. The ZnS(Ag)

scintillator is connected to a photomultiplier tube with 6.3 cm diameter by a tapering light guide. The accumulation chamber (13 L volume) collects radon and thoron gases exhaled from the ground surface. In this study, the count rate was recorded every 30 second during 30 minutes from setting the instrument on the ground.

The thoron exhalation rate E_T was obtained by the following formula;

$$E_T = (N_{10} - N_b)\, CF_T$$

where N_{10} is the count rate (cpm) after 10 minutes from measurement start, N_b is the count rate of background, and CF_T is the conversion factor for thoron (18.1 ± 3.2 mBq m^{-2} s^{-1} cpm^{-1}) [17-22].

Because thoron and its decay product (^{216}Po) reach equilibrium after 7-8 minutes, the count rate for the thoron exhalation rate is assumed to be N_{10}. On the other hand, the count rate after 30 minutes from measurement start (N_{30}) is used for radon exhalation rate, because the count rate of radon and its decay products (^{218}Po and ^{214}Po) continue gradually increase subsequent after 10 minutes.

The radon exhalation rate E_R was obtained by the following formula;

$$E_R = (N_{30} - N_{10})\, CF_R$$

where N_{30} is the count rate after 30 minutes from measurement start (cpm), and CF_R is the conversion factor for radon (0.521 ± 0.040 mBq m^{-2} s^{-1} cpm^{-1}) [17-22]. The measurement errors for radon and thoron have been estimated to be 19% and 6% [20, 22], respectively.

The detection limits of radon and thoron exhalation rates were calculated by the following formulas [17, 18];

Tn: $\sqrt{N_b} \times CF_T$ mBq m^{-2} s^{-1}

Rn: $\sqrt{\dfrac{2}{3} N_b + N_{10}} \times CF_R$ mBq m^{-2} s^{-1}

The *in situ* measurements were carried out at a total of 7 points covered with Shimajiri-maji (Fig. 2, E1-E7) on 17 and 18 July 2010 during clear or cloudy conditions. To avoid the artificial influence on the exhalation rates, flat and non-vegetation soil grounds beside farmland were selected as the measurement points.

2.2. Analyses for ^{238}U and ^{232}Th Series and ^{40}K Concentrations of Soil Sample

To assess the sources of radon and thoron, Shimajiri-maji samples were taken from the same points for the measurements of the exhalation rates (Fig. 2, E1-E7). Analyses for ^{238}U series, ^{232}Th series and ^{40}K concentrations in the soil samples were performed with a high-purity germanium semiconductor detector with 80,000 s measurement time as follows;

The soil samples were enclosed in cylindrical polypropylene container with ϕ48 mm $\times$ H55 mm after

drying for 24 h at 105 °C. To reach radioactive equilibrium between ^{226}Ra and ^{222}Rn, the air-tight container was retained for 40 days.

The photo peaks of ^{214}Pb at 352 keV and ^{214}Bi at 609 keV, were used for the estimation of the ^{238}U series (^{226}Ra) concentration, and the photo peak of ^{228}Ac at 911 keV was used for the estimation of the ^{232}Th series (^{228}Ra) concentration. The ^{40}K concentration was also determined on the basis of its single photo peak at 1,461 keV.

2.3. Short-term Measurement of Atmospheric Radon Concentration

The measurements were performed every one hours at 2 sites under the eaves of private houses beside the farmlands, C1 and C2 (Fig. 2). The equipment with an ionization chamber (SAPHYMO, Alpha Guard) set the height from ground surface within 1 m in diffusion mode that the design optimized for fast passive diffusion. The measurement range and error were 2 - 2,000,000 Bq m^{-3} and about 3%, respectively. The measurement periods were 16: 00 JST on 16 July to 19: 00 JST on 18 July 2010 (53 h) for C1, and 13: 00 JST on 16 July to 9: 00 JST on 19 July 2010 (69 h) for C2.

3. Results and Discussion

3.1. Radon and Thoron Exhalation Rates

The exhalation rates at 7 points (Fig. 2, E1-E7) are listed in Table 1. The arithmetic mean $\pm$ S.D., range and median of the radon exhalation rates excepted ND (E4), were calculated to be 44 ± 46 mBq m^{-2} s^{-1}, 9-137 mBq m^{-2} s^{-1} and 31 mBq m^{-2} s^{-1}, respectively. The detection limit (ND value) for E4 was calculated to be 8 mBq m^{-2} s^{-1}. Also for the thoron, the rates were calculated to be $3,980 \pm 927$ Bq m^{-2} s^{-1}, 2,570-5,530 mBq m^{-2} s^{-1} and 3,970 mBq m^{-2} s^{-1}, respectively.
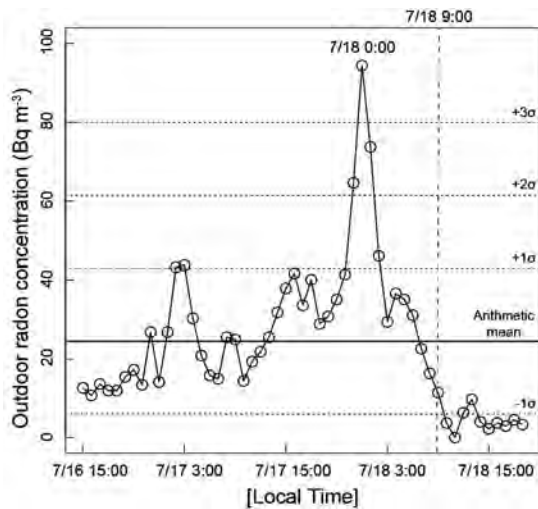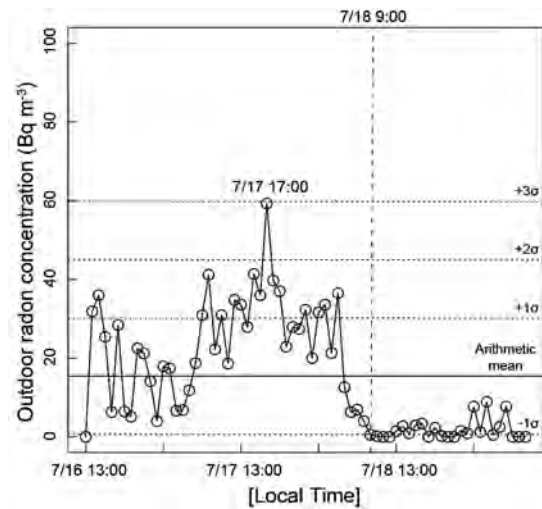
The arithmetic mean for radon exhalation rate observed in Minamidaito-jima is 2 times higher than that of Okinawa-jima (22 mBq m^{-2} s^{-1}, 27 data) [6]. Also the arithmetic mean is 5 times higher than that of the mainland of Japan (8.6 mBq m^{-2} s^{-1}, 111 data) [4]. The arithmetic mean for thoron is 2 times higher than that of Okinawa-jima (2,100 mBq m^{-2} s^{-1}, 30 data) [6]. Also the arithmetic mean is 5 times higher than that of the mainland of Japan (800 mBq m^{-2} s^{-1}, 111 data) [4]. From the above, it is considered that Minamidaito-jima is an area of especially high radon and thoron exhalation rates in Japan.

3.2. ^{238}U and ^{232}Th Series and ^{40}K Concentrations of Soil Sample

The the concentrations of surface soils, Shimajiri-maji, collected at 7 points (Fig. 2, E1-E7) are listed in Table 2. The arithmetic mean $\pm$ S.D., range and median of ^{238}U

Table 2. ^{238}U series, ^{232}Th series and ^{40}K concentrations for soil samples. Locations of the sampling points are shown in Figure 2

Point	Concentration (Bq kg^{-1} dry)		
	^{238}U series	^{232}Th series	^{40}K
E 1	79 ± 1	107 ± 1	585 ± 5
E 2	92 ± 1	114 ± 1	645 ± 6
E 3	190 ± 1	106 ± 1	587 ± 5
E 4	254 ± 1	94 ± 1	597 ± 6
E 5	107 ± 1	91 ± 1	530 ± 5
E 6	141 ± 1	60 ± 1	613 ± 6
E 7	153 ± 1	136 ± 1	546 ± 5
Arithmetic mean ± SD	145 ± 61	101 ± 24	586 ± 5

Fig. 4. Variation of outdoor radon concentration observed at C1 (Fig. 1).

Fig. 5. Variation of outdoor radon concentration observed at C2 (Fig. 1).

series concentrations were estimated to be 145 ± 61 Bq kg^{-1} dry, 79-254 Bq kg^{-1} dry and 141 Bq kg^{-1} dry, respectively. For ^{232}Th series, the concentrations were also estimated to be 101 ± 24 Bq kg^{-1} dry, 60-136 Bq kg^{-1} dry and 106 Bq kg^{-1} dry, respectively. And for ^{40}K, the concentrations were estimated to be 586 ± 39 Bq kg^{-1} dry, 530-645 Bq kg^{-1} dry and 587 Bq kg^{-1}dry, respectively.

In this study, the data were not enough to examine the relationships between exhalation rates and nuclide concentrations. On the other hand, the other studies suggested that there are the weak correlations between the radon and thoron exhalation rates and the ^{238}U and ^{232}Th series concentrations, respectively[6, 18]. The arithmetic means of ^{238}U and ^{232}Th series concentrations of surface soils collected at a total of 11 points in Okinawa-jima were estimated to be about 67 Bq kg^{-1} dry and 55 Bq kg^{-1} dry, respectively[23]. Therefore, it is considered that the concentrations of Shimajiri-maji in Minamidaito-jima are about two times higher than that of the soils in Okinawa-jima. Also the results suggest that the high radon and thoron exhalation rates observed in Minamidaito-jima are derived from relatively high ^{238}U and ^{232}Th series concentrations of the soils.

3.3. Atmospheric Radon Concentration

The variations of atmospheric radon concentrations at C1 and C2 are shown in Figs. 4 and 5, respectively. The arithmetic mean ± S.D. and range of the concentrations at C1 were estimated to be 25 ± 19 Bq m^{-3} and ND-94 Bq m^{-3}, respectively. The maximum value was observed at 0: 00 JST on 18 July, and then the concentration rapidly decreased (Fig. 4). At C2, the arithmetic mean ± S.D. and range of the concentrations were estimated to be 15 ± 15 Bq m^{-3} and ND-59 Bq m^{-3}, respectively. The maximum value was observed at 17: 00 JST on 17 July (Fig. 5).

The nationwide arithmetic mean for the outdoor radon concentration in Japan was estimated to be 6.1 Bq m^{-3} [24]. Though there are some difficulties to compare the data mainly for the time length reason, it is considered that the means of atmospheric radon concentrations observed in Minamidaito-jima are about 3-4 times higher than that for the nationwide survey.

Minamidaito-jima is an isolated island in Philippine Sea (Fig. 1). In general, the concentration of ^{226}Ra, parent nuclide of radon, in the surface seawater is extremely lower than those of rocks and soils[25, 26], and the atmospheric radon concentration on the ocean is very
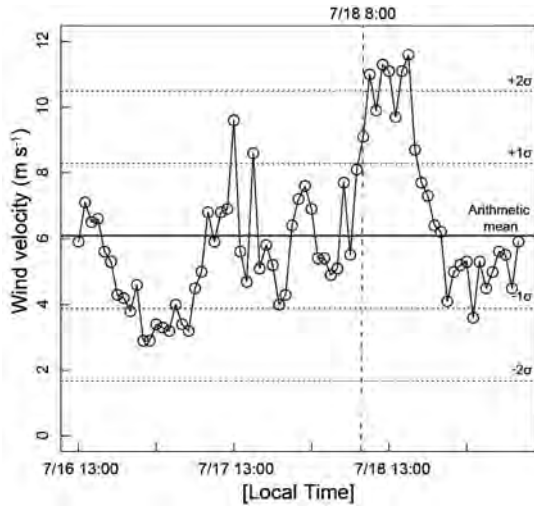
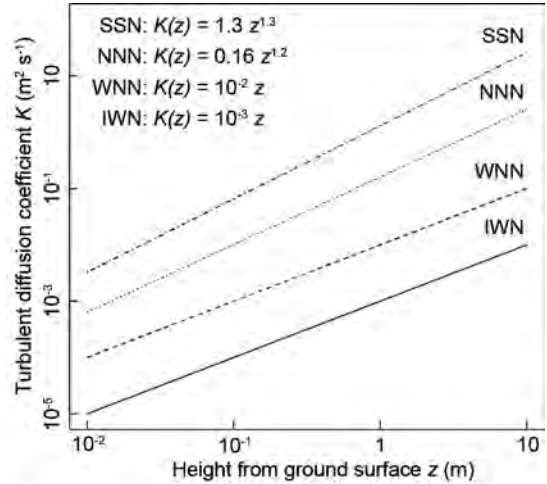**Fig. 6.** Variation of the wind velocity during the measurement of atmospheric radon concentration[29].

Fig. 7. Vertical profiles of the turbulent diffusion coefficient in the atmosphere based on Jacobi and Andre (1963)[30].

low, e.g. 0.039 – 0.048 Bq m[-3] on the Antarctic Ocean[27]. In addition, [238]U series concentration of coral reef limestone in Okinawa prefecture are vary low[8, 23], roughly < 10 Bq kg[-1]. Therefore, it is considered that the main source of the relatively high atmospheric radon concentration are soils in Minamidaito-jima, Shimajiri-maji, and that the basin area, Hagu-shita (Fig. 2), may act as a reservoir of atmospheric radon.

In general, the time-varying for the atmospheric radon concentration in the study area can be explained by the daily change of atmospheric condition. For instance, because of the radon exhaled from soil surface to the upper layer by growth of the atmospheric convection, the radon concentration near the ground surface decreases during the daytime. On the other hand, the concentration increases during the nighttime, because the radon collects in the lower atmosphere by the growth of atmospheric inversion layer[28]. And it is considered that the wind strengthened from 8:00 JST on 18 July (Fig. 6) caused the relatively low concentration started about 9:00 JST on 18 July (Figs. 4 and 5). Namely, the air with extremely low radon concentration flown into Minamidaito-jima from the ocean area is a plausible reason for the decreasing of radon concentration during the measurements at least.

3.4. Relationship between Atmospheric Radon Concentration and Radon Exhalation Rate

To estimate the atmospheric radon concentration was calculated by a method based on Jacobi and Andre (1963)[30]. If the horizontal coordinates x and y at an height z from the ground level (z = 0), the radon (Rn) and thoron (Tn) and these decay products concentration at a points in the atmosphere is given by the following general equation;

$$\frac{\partial n_i}{\partial t} = \nabla \cdot (K \nabla n_i) - u \cdot \nabla n_i + v_i \frac{\partial n_i}{\partial z} + \lambda_{i-1} n_{i-1} - (\lambda_i + \Lambda_i) n_i \quad (1)$$

where, i is the index number in the decay chain (Rn, Tn: i = 1; [218]Po, [216]Po: i = 2; ···), n_i is the concentration of the ith nuclide, K is the turbulent diffusion coefficient, $u_{(x, y, z)}$ is the components of the mean wind velocity, v_i is the mean sedimentation velocity of the ith nuclide (Rn, Tn: v_1 = 0), λ_i is the radioactive decay constant of the ith nuclide and Λ is the mean removal rate of the ith nuclide, caused by washout and rainout (Rn, Tn: Λ_1 = 0).

Therefore, the radon concentration in the atmosphere (n) is given by the following equation;

$$\frac{\partial n}{\partial t} = \nabla \cdot (K \nabla n) - u \cdot \nabla n - \lambda n \quad (1)'$$

where, λ is radioactive decay constant of radon.

The equilibrium profile for steady-state conditions at constant radon exhalation rate is obtained from (1) if $\partial n / \partial t$ = 0. To solve (1)' the following additional assumptions are substituted;
1. $\partial n / \partial x = \partial n / \partial y = 0$.
2. $u_z = 0$.

Those assume that the vertical turbulence profile and the radon exhalation rate is the constant at each place, And the vertical wind velocity is small compared with the velocity of vertical turbulent diffusion.

With these assumptions the equilibrium vertical profile of radon can be obtained from the following differential equation;

$$\frac{d}{dz}\left(K \frac{dn}{dz}\right) - \lambda n = 0 \quad (2)$$

To solve (2), the following boundary conditions were introduced:

Table 3. Estimated and measured atmospheric radon concentration on Minamidaito-jima

	Atmospheric radon concentration (Bq m^{-3})
SSN	0.2
NNN	1.5
WNN	14
IWN	143
C1	25 (ND-94)
C2	15 (ND-59)

1. $\int_0^\infty \lambda n\,dz = E.$
2. $n\,(z \to \infty) \to 0.$

where E is the measured median radon exhalation rate from the ground surface in the Minamidaito-jima (0.031 Bq m^{-2} s^{-1}), The condition 1 means that the whole activity of radon in the vertical atmospheric column is equal to the measured exhalation rate. The condition 2 assumes the result of radioactive decay.

Accordingly, (2) can be changed the following equation:

$$\frac{dn}{dz} = \frac{E}{K} \quad (3)$$

where, Jacobi and Andre (1963) presented the four characteristics K profiles that dependent on the height from ground surface (z) according to the variations of vertical wind velocity and the stability of atmosphere[30]. In this study, the K profiles on the each case (SSN, NNN, WNN and IWN) are approximated the exponent function $K(z)$ between 1 cm to 100 m (Fig. 7).

From the above, (3) represent the following formula;

$$n = E \int \frac{1}{K(z)} \, dz \quad (3)'$$

In addition, the calculated height was assumed to be 10^{-2} m − 1 m. The measurements of atmospheric radon concentration were performed at the height from ground surface within 1 m. In the height under few mm, because the molecular viscosity is predominant over the turbulent diffusion, the lower limit is decided to be 10^{-2} m.

The results of the calculations were shown in Table 3. The range of the estimated atmospheric radon concentrations was calculated to be 0.2 Bq m^{-3} − 143 Bq m^{-3}. On the other hand, the ranges of the measured concentrations were determined to be ND − 94 Bq m^{-3}. The estimated concentrations were likely to consist with the measured concentrations. In particularly, the estimated concentration for WNN (14 Bq m^{-3}), the case of the weak vertical mixing condition throughout the ground layer[30], was reasonable compared with the arithmetic means of measured concentrations (25 Bq m^{-3} for C1 and 15 Bq m^{-3} for C2). In addition, the estimated

concentration for IWN (143 Bq m^{-3}), the very weak mixing condition assumed strong inversion near ground[30], suffices for the maximum of the measured concentrations (94 Bq m^{-3} for C1 and 59 Bq m^{-3} for C2). In other words, the results strongly suggest that the atmospheric radon concentration in Minamidaito-jima is enhanced due to the relatively high radon exhalation rate from soil surface.

4. Conclusion

General survey for the environmental radon and thoron has been performed in Minamidaito-jima, a subtropical island of southwest Japan. The results indicated that the radon and thoron exhalation rates from the soil surface and the atmospheric radon concentration in the island are specifically higher than those of the average of Japan. And it was considered that the cause of the high exhalation rates and concentration is a particular soil mainly derived from eolian dust, Shimajiri-maji, widely distributed in the island.

Acknowledgment

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