Radon and thoron exhalation rates and their some correlating factors

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Abstract. Radon and thoron exhalation rate was measured in 23 sites of the capital and 9 prefectures. The various exhalation rates were obtained in the sites and the different among measured values was interpreted by difference of geology. The attention was paid to the moisture content and the weather element in measurement of the exhalation rate in a laboratory measurement. The radon and thoron exhalation rate differed greatly due to the measurement sites. Good positive correlation (Correlation coefficient: 0.78) was accepted between the radon and the thoron exhalation rates. It was suggested that another side can be presumed by measuring one side by making use of the regression. Radon and thoron exhalation rate according to geology increased in proportion to a sedimentary rock, granite, *shirasu*, andesite, basalt, and *Kanto loam*. The geology was, therefore, considered to be an important element to explain the difference among exhalation rates. It was suggested that the SiO_2 content in the earth layer was a good index in order to presume an exhalation rate of radon and thoron emanated from the earth surface. Radon exhalation rate decreased linearly with the increase in moisture content. Thoron exhalation rate showed the same tendency as the UNSCEAR report.

1. Introduction

The world average value of the annual effective dose by natural radiation is about 2.4 mSv [1]. The effective dose from radon, thoron and their decay products occupies about 50% of the whole. The urainium-238 and thorium-232 from which radon and thoron generate exist mainly among soil and rock and they decay at each existing position in the soil and rock. Radon and thoron emanate mainly from the earth surface through the gap in soil to the atmosphere. Radon gas enters into the house from various gaps in wall and open window or door. There are many houses where radon concentration is very high in Europe, America and other regions and countries, and these several governments have regulated radon concentration in dwellings, whereas Japan is in the starting stage of examination of the radon action level. [2]. It is, therefore, important to know the radon and thoron exhalation and also geology, and these information are useful for presumption of a high radon concentration area.

Radon and thoron exhalation rate is influenced of environmental factors, such as weather element, water content, and geology. The difference of geology distribution and soil element in the crustal layer might affect the exhalation rate, therefore *in-situ* measurement was performed in the work. Moreover, the dependency of exhalation rate to water content of soil was examined using soil sampled at *in-situ* measurement in the laboratory measurement.

2. Material and Method

2-1 Outline of measuring instrument of exhalation rate

The *in-situ* measuring instrument for radon and thoron (MSZ) developed by Shimo *et al.* [3, 4] for the purpose of estimating radon and thoron exhalation rates were used in this study. The instrument is constructed by the large acrylics board of about 1200 cm^2 with ZnS (Ag) scintillator and the skirt section (volume 13 liter) for saving up radon and thoron gases which dissipate from the earth surface. A counting efficiency of the instrument is 19.2±1.0%. Each counting time was 30 seconds, and it continuously continued for 30 minutes. The thoron and radon exhalation rates were calculated by count rates of 10 minutes and 30 minutes after measurement start, respectively. The conversion factors for concentration from count rate for thoron exhalation rate and radon exhalation rate are 18.1±3.2 and 0.521±0.040 in unit of $\mathrm{mBq \cdot m^{-2} \cdot s^{-1} \cdot cpm^{-1}}$, respectively.

2-2 Investigation Region

Measurement was performed in a total of 23 sites of the capital and 9 prefectures. The number of measurement was two or more times in almost measurements. However, there were several sites where count was less than the detection limits. The geology of each measurement site is respectively basalt, andesite, granite, sedimentary rock, *Kanto loam* and *shirasu*, and it is weathering soil except the last two soils.

2-3 Measurement of moisture content

The experimental procedure was performed as follows; Weathering granite soil was put into the 275×210×100mm container and the probe of an aqua meter□UIZ-ECH10 UIZIN size 150×32 mm) was installed in the center of the soil sample. The adjustment of the amount of moisture was performed by spraying water on the soil surface with a spray. After that, the exhalation rate measuring device was put on the soil. Measurement time was 30 minutes like *in-situ* measurement. The radon and thoron exhalation rate E was computed from the following equation 2-1 [5],

$$E = A_{\mathrm{Ra}} f \rho \left(\lambda_{\mathrm{Rn}} D_e \right)^{1/2} \left[\frac{\varepsilon_a}{\left(\varepsilon_a + \kappa \varepsilon_w + \rho K \right)} \right]^{1/2} \qquad \text{2-1}$$

where A_{Ra} was ^{226}Ra content of soil, f emanation coefficient, ρ soil density, ε_a air filled porosity, ε_w water filled porosity, K sorbed geseous partition coefficient, κ aqueous geseous partition coefficient, D_e effective diffusivity, λ_{Rn} decay constant (^{222}Rn), and λ_{Rn} decay constant (^{220}Rn). The calculated value E was computed with ^{226}Ra content of soil measured by γ-ray spectrometry.

3. Result and Discussions

3-1 Radon and thoron exhalation rate by Area

The measurement result of radon and thoron exhalation rate is shown in Fig.3-1. The arithmetic average value of the radon and thoron exhalation rates by all 49 data were obtained to be 7.6±2.6 mBq $\cdot m^{-2}\cdot s^{-1}$, 692±156 mBq$\cdot m^{-2}\cdot s^{-1}$, respectively. The maximum values of the radon and thoron exhalation rate were 33.7±4.8 mBq$\cdot m^{-2}\cdot s^{-1}$ and 3705±84 mBq$\cdot m^{-2}\cdot s^{-1}$, respectively, which were measured in Minamiaiki-mura, Nagano (ER9) where is the sedimentary rock area. Radon exhalation rates of Chiba-shi, Chiba (ER4:*kanto loam*), Tsuruga-shi, Fukui (ER11:granite), and Fujimi-mura (ER1:basalt), Gumma (basalt) were below the detection limits (ND). These sites also showed the minimum value of the thoron exhalation rate, which were 110±43.4, 77.8±39.3, and 14±10, respectively, in unit of mBq$\cdot m^{-2}\cdot s^{-1}$. When the maximum value (ER9) and the minimum value (ER1) of the radon and thoron exhalation rate were compared, ratios of maximum to minimum were taken to be 37 times and 265 times, respectively.

On the other hand, UNSCEAR 2000 report shows that the world averages of the radon and thoron exhalation rates are 26.2 mBq$\cdot m^{-2}\cdot s^{-1}$ and 1000 mBq$\cdot m^{-2}\cdot s^{-1}$,respectively [1]. The obtained value in the survey was smaller than the value of UNSCEAR; radon and thoron exhalation rates were respectively about 26% and 70% of values shown in UNSCEAR report.

The average value of outdoor radon concentration in Japan is 6.2 Bq$\cdot m^{-3}$, whereas a world average is 10 Bq$\cdot m^{-3}$. This fact suggests that the radon exhalation rate in Japan is relatively small in comparison to a world average.

3-2 Correlation between the radon exhalation rate and the thoron exhalation rate

The correlation between radon exhalation rate and thoron exhalation rate was indicated in Fig.3-2 and plotted values were arithmetic means of each measurement location. This figure shows the positive correlation between the radon exhalation rate and thoron exhalation rate with correlation coefficient of 0.78 and we deduce the following formula:

$$Tn = 110Rn^{0.85} \qquad \cdots \quad 3\text{-}1$$

It was admitted that the thoron exhalation rate was about 100 times of the radon exhalation rate. This formula, furthermore, means that another side can be presumed by measuring one side.

3-3 Geology and radon and thoron exhalation rate

The relation between geology and the exhalation rate is shown in Fig.3-3. And the figure indicates that the exhalation rate has an increasing tendency in order of basalt, andesite, and granite. An igneous rock is classified into 4 groups of super basic rock (under 45%), basic rock (basalt etc.; 45-52%), neutral rock (andesite etc.; 52-66%), and acidic rock (granite etc.; over 66%) according to weight percent of SiO_2 content shown in bracket [6]. The top layer of *Kanto loam* originates in basalt, and its SiO_2 concentration is 32.31-40.22% [7]. The SiO_2 content of *shirasu* which originates in liparite is 65-73% [8]. The correlation between the exhalation rate and the SiO_2 concentration in rock which was shown in reference [9] was shown in Fig.3-4. Exhalation rates from *shirasu* and *Kanto* *loam* were mutually compared and the followings was clear; both radon and thoron exhalation rates from *shirasu* of high SiO_2 concentration were bigger than those from *Kanto loam* of low SiO_2 concentration. Furthermore, it was cleared that a strong positive correlation was observed between the radon and thoron exhalation rate and the SiO_2 concentration, and that the correlation coefficient of radon and thoron exhalation rates were 0.89 and 0.90, respectively. The data also showed that radon and thoron exhalation rate increased in proportion to the SiO_2 concentration in the soil. The author, therefore, considered that the SiO_2 concentration in the soil and rock is able to be used as an effective index for searching big and/or small exhalation rate area.

3-4 Moisture content and radon and thoron exhalation rate

The relation between the radon exhalation rate and the moisture in soil was examined using sampled soil in the laboratory measurement. The result was shown in Fig.3-5. The difference between measured value and calculated one was observed in the range of low moisture content. In the moisture content of about 8-16%, the decreasing inclination of calculated value was roughly similar to that of measured value. Over the range over 16%, the measured value decreased linearly in inverse proportion to the increase of moisture content whereas calculated one was little change. The calculated value was 8.4 time of the measured maximum value at moisture content of 23%. The authors think that this cause is to be assigned the reference value [1] adopted in the calculation except radium concentration which was measured. The authors also consider that more detailed examination can be performed by substituting the measured value of each factor of a soil sampled. Moreover, the value obtained in the survey showed good coincidence with Koarashi *et al.* [10], who obtained absolute exhalation rate and its variation characteristics in sandy soil. This result shows that a problem has moisture content in a calculation value at 6% over. The radium concentration of the soil adopted by Koarashi et al. was 26.27 $Bq\cdot kg^{-1}$ and it was 50% smaller than that (57.0 $Bq\cdot kg^{-1}$) of the weathering granite soil used by this study. Although radium content was about 2 times, the radon exhalation rate was almost equal. This fact shows that the exhalation rate is greatly influenced not only moisture content but many other factors.

For the moisture dependency of thoron exhalation rate, both calculated and measured values were shown in Fig.3-6. The measured value was 3.8 time of the calculated value at moisture content of 0.8%. These values were almost consistent together at about 5%. Over the range over 5%, the measured value decreased linearly in inverse proportion to the increase of moisture content whereas calculated one was little change.

In another case, Tozawa [11] reported that the maximum value of thoron exhalation rate appeared at moisture content of 4%. As above mentioned, the authors results showed that the thoron exhalation rate decreased rapidly when the moisture content increased over 10%. On the other hand, the calculated exhalation rate was different from the two measurements, and the discrepancy between measured and calculated values must be explained in the following work.

4. Conclusion

Radon and thoron exhalation rate was measured in 23 sites of the capital and 9 prefectures. The various exhalation rates were obtained in the sites and the different among measured values was interpreted by difference of geology. The attention was paid to the moisture content and the weather element in measurement of the exhalation rate in a laboratory measurement. Consequently, the following knowledge was acquired.

(1) The arithmetic average values of the radon and thoron exhalation rate were 7.6±2.6 and 692±156 in the unit of $mBq \cdot m^{-2} \cdot s^{-1}$.The radon and thoron exhalation rate differed greatly due to the measurement sites. When maximum value was compared with the minimum value, the ratio of maximum to minimum radon exhalation rate was to be 37 times and the ratio of thoron exhalation rate to be 265 times.

(2) Good positive correlation (Correlation coefficient: 0.78) was accepted between the radon and the thoron exhalation rates. It was suggested that another side can be presumed by measuring one side by making use of the regression.

Radon and thoron exhalation rate according to geology increased in proportion to a sedimentary rock, granite, *shirasu*, andesite, basalt, and *Kanto loam*.The geology was, therefore, considered to be an important element to explain the difference among exhalation rates.

(3) The strong positive correlation was accepted between the radon and thoron exhalation rate and the SiO_2 concentration estimated from reference. Each correlation coefficient was calculated to be 0.89 and 0.90, respectively. It was suggested that the SiO_2 content in the earth layer was a good index in order to presume an exhalation rate of radon and thoron emanated from the earth surface.

(4) The relation between exhalation rate and moisture content was obtained in the laboratory measurement using weathering granite soil. Radon exhalation rate decreased linearly with the increase in moisture content. Thoron exhalation rate showed the same tendency as the UNSCEAR report.

(5) Measured exhalation rate was compared with the calculated one. These values differed each other in the low moisture content, and the difference between the two values was several times.

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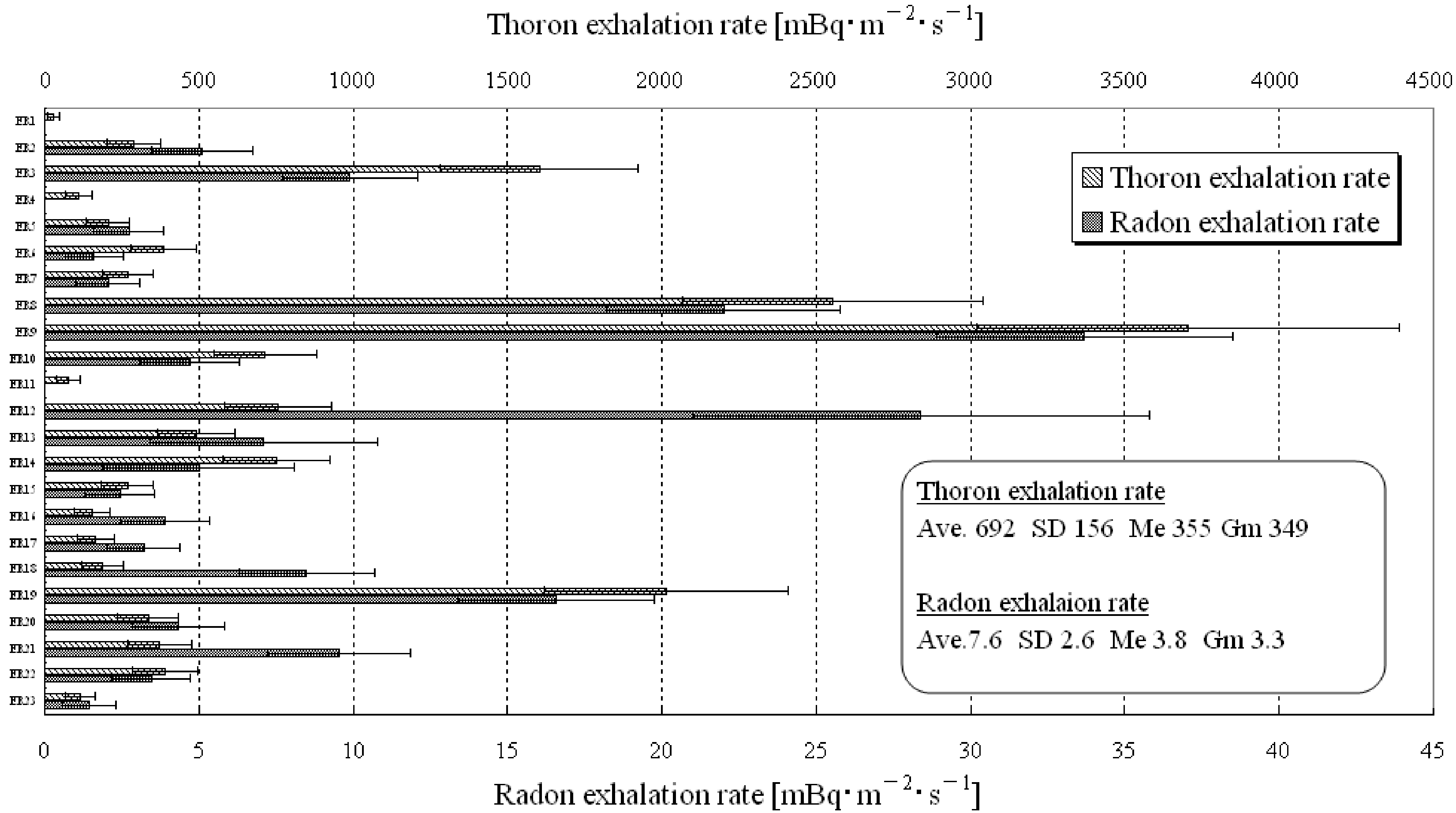
Table 2□1 Investigation area and its geology

Prefecture	City	Sample Number	Number of measurements	Geology
Gumma	Fujimi Village	ER1	4	Basalt
Ibaraki	Tsukuba City	ER2	3	Granite
	Yasato Town	ER3	2	Granite
Chiba	Chiba City	ER4	22	*Kanto loam*
Tokyo	Katsushika-ku	ER5	4	*Kanto loam*
Nagano	Yamanouchi Town	ER6	1	Andesite
	Komoro City	ER7	2	Basalt
	Saku Town	ER8	1	Sedimentary rock
	Minamiaiki Village	ER9	1	Sedimentary rock
	Minamimaki Village	ER10	2	Andesite
Fukui	Tsuruga City	ER11	5	Granite
Toyama	Jouhana Town	ER12	1	Sedimentary rock
	Himi City	ER13	1	Sedimentary rock
	Asahi Town	ER14	1	Sedimentary rock
Kumamoto	Ubuyama Village	ER15	2	Basalt
	Aso Town	ER16	3	Andesite
	Oguni Town	ER17	2	Basalt
	Ohtsu Town	ER18	1	Basalt
Miyazaki	Miyazaki City	ER19	3	Sedimentary rock
	Miyakonojo City	ER20	2	*Shirasu*
Kagoshima	Kagoshima City	ER21	2	*Shirasu*
	Yoshida Town	ER22	2	*Shirasu*
	Sakurajima Town	ER23	5	Basalt


Thoron exhalation rate [mBq･m^−2･s^−1]
0 500 1000 1500 2000 2500 3000 3500 4000 4500
Thoron exhalation rate
Radon exhalation rate
Thoron exhalation rate
Ave. 692 SD 156 Me 355 Gm 349
Radon exhalaion rate
Ave.7.6 SD 2.6 Me 3.8 Gm 3.3
0 5 10 15 20 25 30 35 40 45
Radon exhalation rate [mBq･m^−2･s^−1]

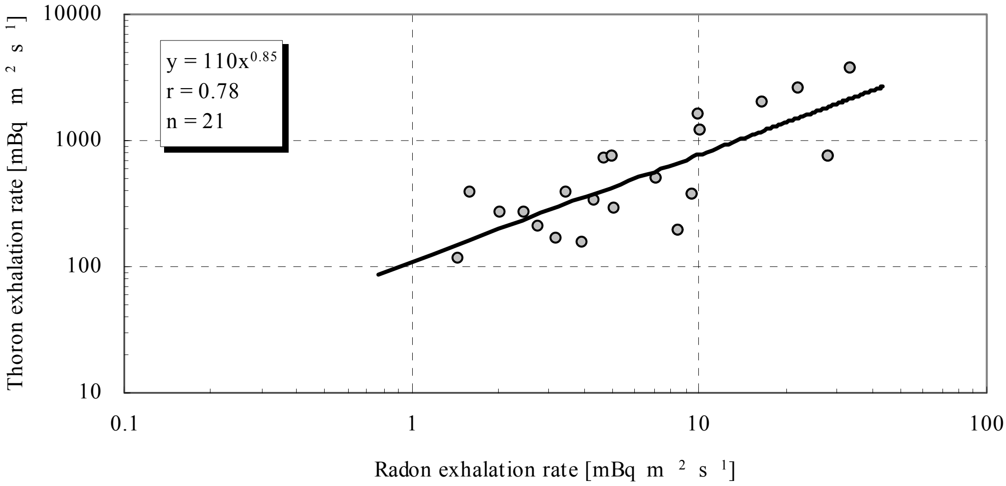


Fig.3□2 Relation between radon exhalation rate and thoron exhalation rate. (Average)

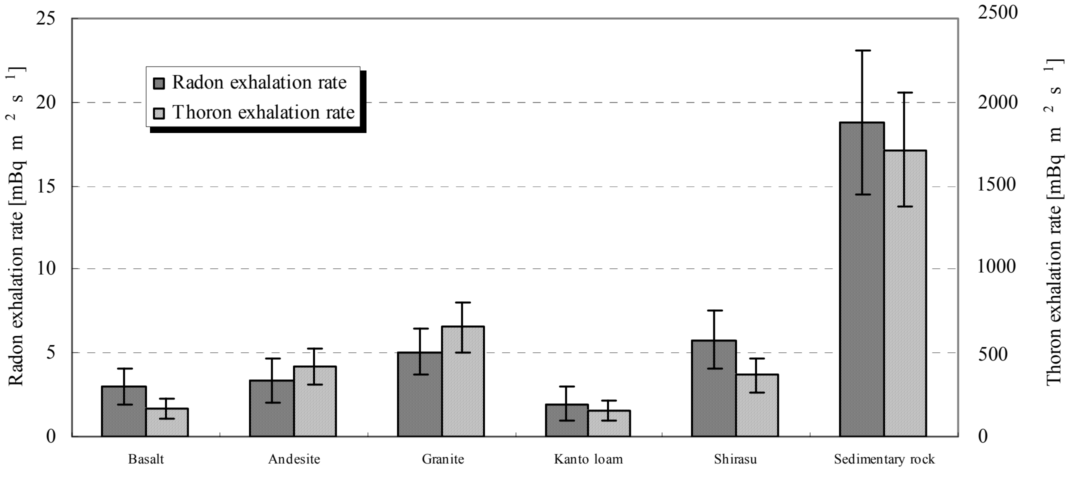


Fig.3□3 Relation between geology and radon and thoron exhalation rates.

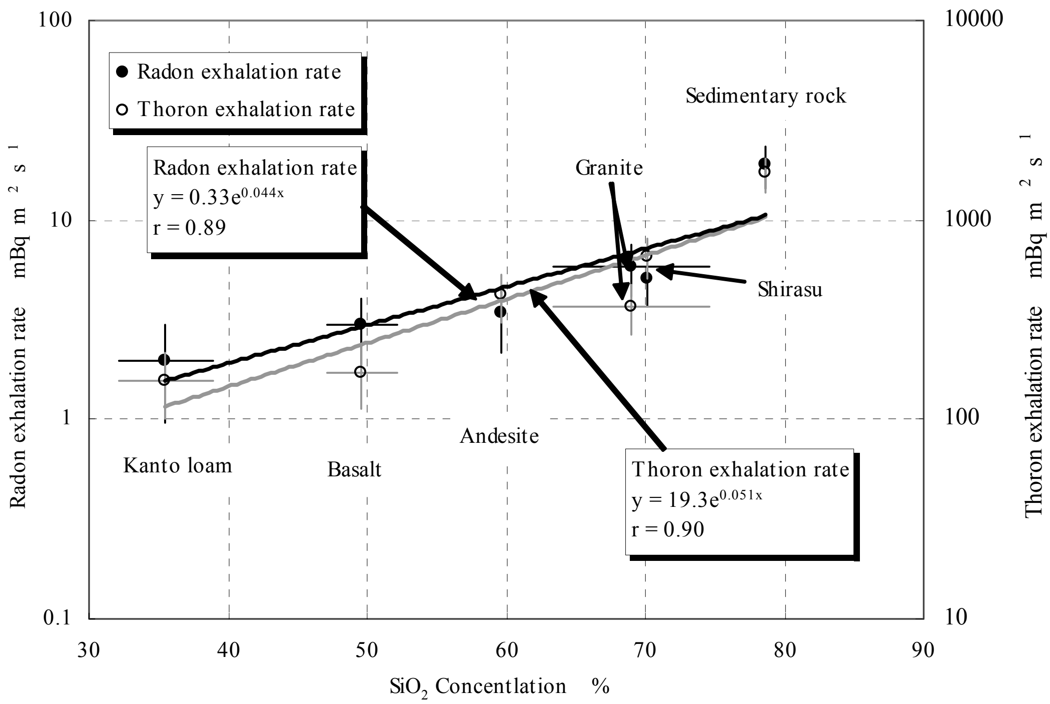


Fig.3□4 Relation between soil component and radon and thoron exhalation rate.

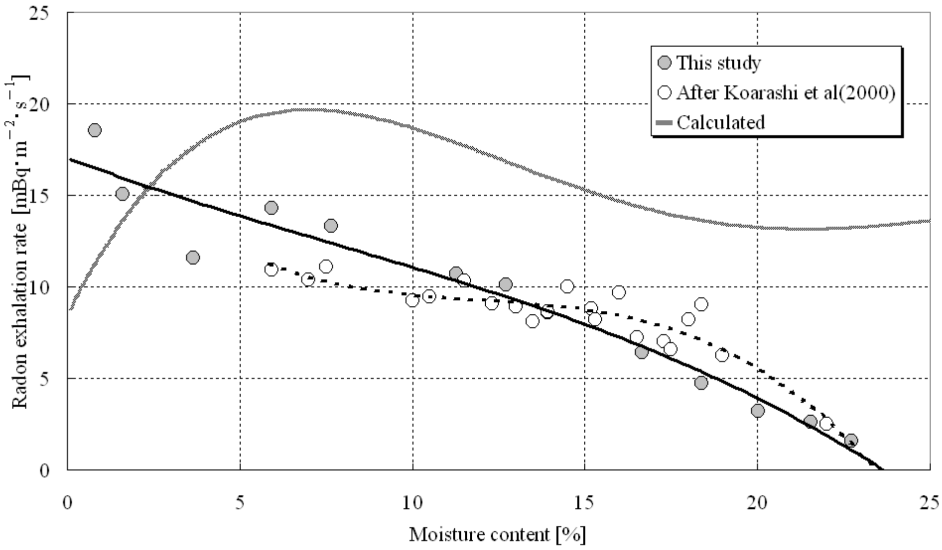


Fig.3□5 Comparison of calculation and survey .(Radon exhalation rate)

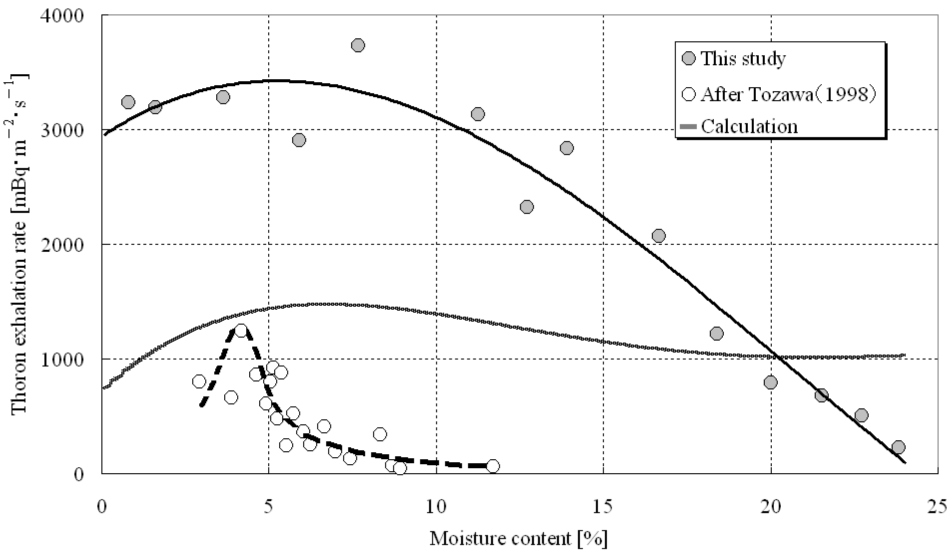


Fig.3□6 Comparison of calculation and survey .(Thoron exhalation rate)