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ORIGINAL

Investigation on Thoron Progeny and Radon Progeny Concentrations in Living Environment and an Estimation of Their Effective Dose to the Public

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This study was conducted to investigate the concentrations of thoron (^{220}Rn) progeny and radon (^{222}Rn) progeny and to compare the radiation exposure due to them in different kinds of dwellings. The results indicated that thoron progeny concentrations indoors might strongly depend on the kinds of building material used in each dwelling, whereas the radon progeny concentration did not depend on the kinds of building material. The average thoron progeny concentration in the traditional Japanese dwellings with soil wall was 3.22 Bqm^{-3}, and the radon progeny concentration in the dwellings was 8.35 Bqm^{-3}. The annual effective dose equivalent to thoron progeny was 1.19 mSv, which was higher than that of radon progeny (0.82 mSv) in the same dwellings assuming an occupancy factor of 1.

KEY WORDS: thoron progeny concentration, radon progeny concentration, dwellings, building materials, annual effective dose equivalent, exposure rate, ^{232}Th content, ^{238}U content, public

I INTRODUCTION

Particular attention has been drawn to the indoor radon (^{222}Rn) problem in recent years since ^{222}Rn and its progeny indoors contribute the largest dose of exposure to the general public in living environment (UNSCEAR 1982[1]; 1988[2]). Concentrations of ^{222}Rn and its progeny in air have been studied in dwellings as well as mines, because these concentrations are important for radiation exposure, especially in the living environment. In contrast, there are few studies of thoron (^{220}Rn) and its progeny in the living environment (HULTQWIST[3] and POHL[4]). The reason for this situation was perhaps that it had been thought little thoron gas could diffuse into a room before decaying because of short half-life of ^{220}Rn (55.6 s), and therefore radiation exposure from the inhalation of thoron and its progeny was negligible compared with radon and its progeny. However, it was suggested recently that the exposure due to ^{220}Rn and its progeny should not be neglected (STEINHÄUSLER 1975[5]; KELLER 1984[6]; SCHERY 1985[7]; KATASE 1988[8]), and more research on thoron and its progeny's behavior in the living environment is urgently needed.

The purpose of this study was to make a primary

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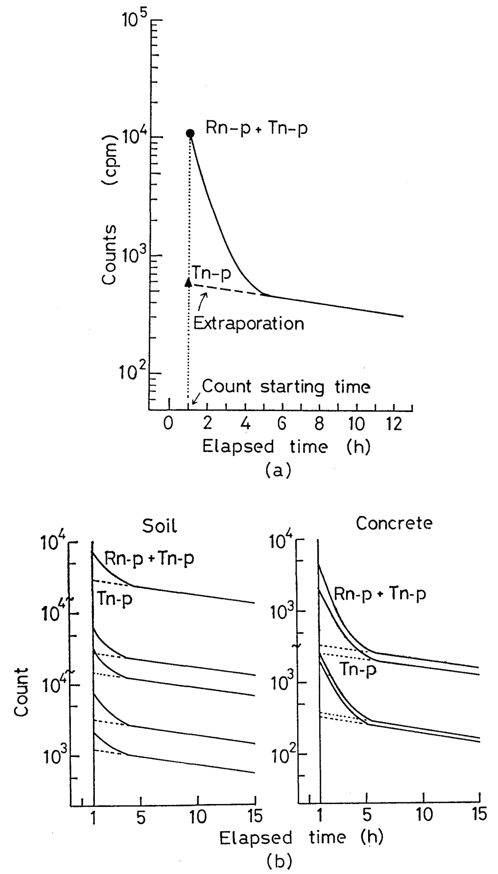

Fig. 1(a) Decay curve of Tn and Rn progeny on the sampling filter, Tn-p: Tn progeny and Rn-p: Rn progeny.
(b) Practical decay curve of Tn and Rn progeny in the dwellings of soil and concrete.

investigation of the different concentration levels of thoron progeny and radon progeny in dwellings with different kinds of building materials, and also to evaluate the effective dose of each.

II MEASURING METHODS

To estimate thoron progeny and radon progeny concentrations, a thoron progeny and radon progeny monitor (TRP monitor) was used throughout the investigation. Thoron progeny and radon progeny were deposited on the fixed filter of the monitor during filtration. After sampling thoron progeny and radon progeny, alpha particles emitted from thoron progeny and radon progeny on the filter were immediately counted with a ZnS(Ag) scintillation counter positioned in front of the filter (SHIMO *et al.*, 1984[9]). Throughout the investigation, the cellulose nitrate membrane filter of 47 mm in diameter and 0.8 μm in pore size (ADVANTEC, Toyo Roshi Ltd., Japan) was used, the flow rate was 17.5 *l*/min, the sampling time was 58 min, and the counting time was also 58 min. A piece of filter sampled was counted at least every 12 h to construct a decay curve. Radon progeny decays faster than thoron progeny because of the differences in half-life (the half-life of radon progeny: ^{218}Po, 3.04 min, ^{214}Po, 26.8 min, ^{214}Bi, 19.9 min; and the half-life of thoron progeny: ^{212}Pb, 10.6 h; ^{212}Bi, 60.0 min). The decay curve is shown in **Fig. 1**(a). The count of thoron progeny immediately after sampling (▲ in **Fig. 1**(a); EV) can be derived by extrapolating the decay curve. If we assume the radioactive equilibrium among thoron progeny, which is thought to actually be a non-equilibrium mixture, the "Equilibrium Assumed Thoron Progeny (EAT) concentration" could be defined and calculated from EV.

After the manner of the EAT concentration, the "Equilibrium Assumed Radon Progeny (EAR) concentration" can be defined under the assumption of radioactive equilibrium among radon progeny, which is also a non-equilibrium mixture. The EAR concentration is the same value as the "apparent radon daughter concentration" proposed for estimating the effective dose due to radon progeny by SHIMO in his former paper.[10] He showed close agreement between the equilibrium equivalent radon concentration and the apparent radon progeny concentration. In addition, the EAR concentration is obtained by applying the one-count method to the operation of the monitor, whereas the equilibrium equivalent radon concentration is taken by the three-count method or by multiplying the radon concentration by the equilibrium factor, therefore the EAR concentration is simple to calculate and the concentration error is relatively small. In this study, the EAR concentration was calculated by subtracting the EV count (▲ in **Fig. 1**(a)) from that of thoron progeny plus radon progeny (symbol ●).

Decay curves were made in each measurement and one of the curves of the investigation is shown in **Fig. 1**(b). The measurement continued for at least 4 days at each location. The position of the apparatus in dwellings during measurement

Table 1 Dwelling classification of our investigation.

Building materials	Proportion in Japan	Number of dwellings investigated
Soil	73%	10
Concrete	15%	15
N.B.M.*	12%	7

*New building materials.

was about 2 m from a wall.

Another continuous measuring apparatus (MGR monitor) (NISHIMURA 1988) was used for evaluating the diurnal variations of both thoron progeny and radon progeny concentrations in a laboratory of Nagoya University throughout the year. This MGR monitor has a turntable and disc-shaped filter holders are supplied on the table from a holder case. Just after sampling air with the first filter for 7 min, the filter holder moves sequentially from a sampling position to two counting positions. A ZnS(Ag) scintillation counter located at each counting position detects the alpha particles emitted from the filter surface. The first counter detects alpha particles from radon progeny and thoron progeny just after sampling, and the second counter takes alpha particles emitted from thoron progeny three hours after sampling. In the investigation, the MGR monitor was practiced by the one-count method, and EAT and EAR concentrations were calculated from the two counts obtained with counters, respectively.

For an advanced analysis of thoron progeny and radon progeny concentrations in dwellings, the contents of ^{232}Th and ^{238}U in building materials were evaluated from gamma counts obtained with a 3″ø×3″ NaI(T1) scintillation spectrometer (MINATO 1978[12]).

III CLASSIFICATION OF BUILDING MATERIALS

According to UNSCEAR 1988, 23% of radon sources indoor are from building materials, and 46% is due to the diffusion and convection from the floor of a room. In fact, this is only the general conclusion on radon sources indoors and the value here is the average value worldwide. Considering some concrete problems on indoor air pollution, the main points are the contents of uranium and thorium in building materials and the exhalation rates of radon and thoron from the surface of interior wall of a building (IKEDA, 1991[13]).

Buildings or dwellings are generally classified by construction style and building materials. In Japan, buildings can be classified by frame construction into timber frame, concrete frame and steel frame, and the concrete frame is classified by reinforced concrete and concrete block. Dwellings can also be classified by building materials into conventional construction and pre-fabricated construction. For the close relation of building materials to thoron concentrations (YONEHARA and AOYAMA 1991[14]; IIDA *et al.*, 1990[15]), buildings researched in this study were classified only by the interior wall materials. The number of dwelling investigated in this study is listed in **Table 1**, and the table also shows the percentage of each kind of building on the basis of national statistics. One of the major characteristics of dwellings in Japan is that about 73% of all the dwellings are wooden framed houses of which the interior walls are soil.[16] A wooden-framed soil-wall house (hereafter, a dwelling made of soil) is the traditional style of building construction in Japan.

The indoor surveys of 32 houses were performed around Nagoya city which is located in the center of the main island of Japan. Ten dwellings are made of soil, 15 dwellings of reinforced concrete, and 7 dwellings of plaster board; board materials and others except soil and concrete we refer to as new building materials.

IV RESULTS AND DISCUSSION

The investigation of thoron progeny and radon progeny concentrations in dwellings which were made of different kinds of building materials was carried out from December 1988 to the end of 1991. During the 3 years, 32 dwellings were measured in all. In most of the investigated dwellings, one or two rooms were measured and some of the dwellings were measured many times throughout the year to show the seasonal variation of thoron progeny and radon progeny concentrations. The diurnal variation was also investigated only in our laboratory, which is a concrete building.

1. Contents of ^{232}Th and ^{238}U in different kinds of building materials

A fraction of the thoron and radon activities

Table 2 Contents of ^{232}Th and ^{238}U in different building materials.

Building materials	^{232}Th (ppm)	^{238}U (ppm)
Soil	8.64±0.15	2.52±0.31
Concrete	8.86±0.19	2.67±0.28
N.B.M.*	5.30±0.11	2.38±0.16

*New building materials.

produced by the decay of ^{232}Th and ^{228}U in building materials enters buildings by diffusion, and this phenomenon is recognized to be the main source of thoron and radon in the traditional Japanese dwellings of timber frame and soil wall (YONEHARA and AOYAMA, 1991[14]); IIDA *et al.*, 1990[15]). The results are shown in **Table 2**. From this table, it is clear that the contents of ^{238}U in different kinds of building materials were nearly the same, but the contents of ^{232}Th in concrete and soil were significantly higher than those in the new building materials.

In addition, the gamma exposure rate in the dwellings was derived from the contents of ^{232}Th and ^{238}U with conversion factors (BECK, 1972[17]), *i.e.* 0.31 μR/ppm for ^{232}Th and 0.62 μR/ppm for ^{238}U, respectively. Then the annual effective doses from ^{232}Th series and ^{238}U series (in mSv) are estimated from **Table 2** as follows: ^{232}Th: 0.15 (for soil), 0.15 (for concrete), and 0.091 (for new building materials); and ^{238}U: 0.084 (for soil), 0.091 (for concrete) and 0.077 (for new building materials) under the assumptions of 1 μR/h = 8.77 nGy/h and 1 Sv = 0.7 Gy.

2. Thoron progeny and radon progeny concentrations in dwellings with different building materials

As mentioned above, **Fig. 1**(b) is an example of decay figures obtained from the measurements in soil and concrete dwellings, respectively. It is clear from the figure that thoron progeny concentrations in the dwellings made of soil were higher than those of the dwellings made of concrete.

The estimated average thoron progeny and radon progeny concentrations in dwellings made of different kinds of building materials are shown on **Table 3**. From this table, it can be seen that the dwellings made of soil and concrete had about the same radon progeny concentration, which was slightly higher than that of the dwellings made of new building materials. As for the concentrations of thoron progeny, the dwellings made of soil had the highest level, 3.23±2.23 Bqm^{-3}, this value was nearly 5 times higher than that of dwellings made of concrete (0.60±0.24 Bqm^{-3}) which was the lowest thoron progeny concentrations in the three kinds of building materials.

Concentrations of thoron progeny or radon progeny are generally dependent on the exhalation rates of thoron or radon gases diffusing from the surfaces of building materials into a room, and ventilation condition of a room, etc. Thoron or radon gases enter a room mainly by crossing the interface of building materials. When ^{224}Ra or ^{226}Ra in building material decay, the resulting ^{220}Rn or ^{222}Rn atoms must first escape from the grain of building material to the pores filled with air which eventually diffuses through these pores to enter a room from the surface of the interior wall. The diffusion process is mathematically described by the definition of an effective diffusion coefficient that includes an allowance for the convoluted path. The exhalation rate can be expressed as (STRANDEN, 1980[18]):

$$E=(C\cdot\varrho\cdot\eta)\sqrt{K\cdot\lambda}\, tgh(d/2\sqrt{\lambda/K}) \quad (1)$$

where E is the exhalation rate of thoron or radon

Table 3 Thoron progeny and radon progeny concentrations in dwellings of different building materials (Bqm^{-3}).

Building materials	Number of dwellings	Concentration	
		Thoron progeny	Radon progeny
Soil	10	3.22±2.23**	8.35±5.08
Concrete	15	0.60±0.24	8.69±3.47
N.B.M.*	7	1.52±0.13	7.02±3.00

* New building materials.
** Mean±SD.
Period of investigation: December 1988 to the end of 1991.

Table 4 Data of soil and building materials for the radon and thoron exhalation.

Parameter	Nuclei	Soil	Building material
C[Bqkg^{-1}]	^{226}Ra	40	60
	^{228}Th	40	50
ρ[kgm^{-3}]		2	1.5
η[—]	^{222}Rn	0.1	0.05
	^{220}Rn	0.05	0.01
K[m^{2}s^{-1}]		5×10^{-6}	5×10^{-7}
E[Bq(m$^2\cdot$s)$^{-1}$]	^{222}Rn	0.02	5×10^{-4}
	^{220}Rn	1	5×10^{-2}

in Bqm^{-2}s^{-1}; C is the content of ^{232}Th or ^{226}Ra in building materials (Bqkg^{-1}); ϱ is the density of building material (kgm^{-3}); η is the emanation factor, *i.e.* the fraction of thoron or radon entering the interstitial volume, and it is strongly related to the porosity of the building material; λ is the decay constant of thoron or radon (s^{-1}); K is the effective diffusion coefficient (m^{2}s^{-1}), and d is the thickness of the wall (m).

The parameters are summarized from some of the literature by Porstendörfer[19] and are shown in **Table 4**. In addition, the effective porosity P is shown as $P=K/D$ where D is the diffusion coefficient, and the diffusion length R is defined as $R=\sqrt{K/\lambda}$.[19] It is clear from **Table 4** that the effective diffusion coefficient K of soil is about 10 times larger than that of building materials, whereas there is no difference in the number of nuclei between thoron and radon. The activity mass concentrations of ^{228}Th and ^{226}Ra are equal, as shown in the table, and a slight difference appears between the contents in soil and in building materials. As is shown in **Table 2**, the content of ^{232}Th is about 4 times higher than that of ^{238}U. If we multiply the contents of ^{232}Th and ^{238}U obtained in our investigation by the decay constants (^{232}Th: 1.4×10^{10} y and ^{238}U: 4.468×10^{9} y) of these nuclei, respectively, we can get the activity mass concentrations of ^{232}Th and ^{238}U; these concentrations are almost equal to those of ^{228}Th and ^{226}Ra shown in **Table 4**. From these observations, we can conclude that the thoron exhalation rate is much larger than the radon exhalation rate and that the exhalation rates of thoron and radon for soil are pretty larger than those for building materials. And we can guess that those differences are caused by the different diffusion lengths of thoron and radon in materials.

Table 5 Ratios of Tn progeny and Rn progeny concentrations in air to ^{238}U and ^{232}Tn contents in building materials in Bqm^{-3}/ppm.

Material	Tn progeny	Rn progeny
Soil	0.37	3.31
Concrete	0.068	3.25
N.B.M.	0.29	2.95

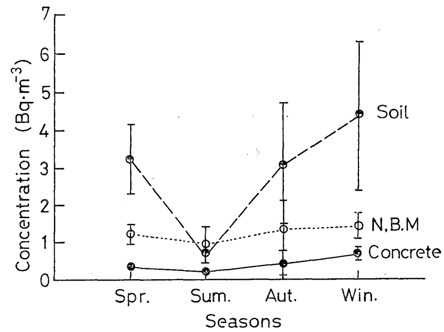

Fig. 2 Seasonal variation of Tn-progeny concentration in different dwellings; N.B.M.: New Building Materials.

To clarify the above, the ratios of thoron progeny and radon progeny concentrations indoors to ^{232}Th and ^{238}U contents in wall materials are shown in **Table 5**. These ratios are generally useful to compare thoron progeny and/or radon progeny concentrations in dwellings made of different materials with each other. The ratios of thoron progeny concentration to ^{232}Th content were the same in values obtained in dwellings made of soil and new building material, and these values were significantly higher than the value obtained in dwellings made of concrete, whereas the ratios of radon progeny concentration to ^{238}U content obtained in three cases of building materials were nearly the same. The ratios of thoron progeny concentration to ^{232}Th content were about one-tenth or less compared to the radon progeny concentration to ^{238}U content. The ratios of thoron progeny concentrations to ^{232}Th content may change under different room conditions, *e.g.* ventilation rate, the wall-surface coating and so on. Therefore, rather more detailed work on this topic is needed in the future.

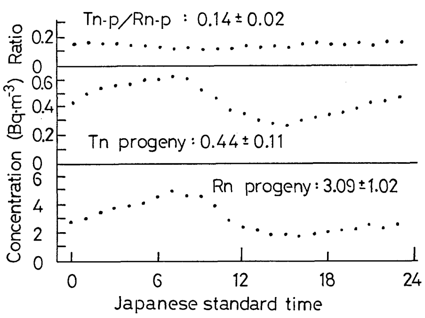

Fig. 3 Diurnal variation of Tn and Rn progeny concentrations in a concrete dwelling.

3. Variation of thoron progeny and radon progeny concentrations

Seasonal variation of thoron progeny was measured in one room of each kind of building material, *i.e.* soil, concrete and new building materials, throughout the year, and their results are shown in **Fig. 2**. In the dwellings made of soil, the thoron progeny concentration was higher in winter and lower in summer, whereas for dwellings made of concrete or new building materials, thoron progeny concentrations showed no significant seasonal variation. This study covered only 32 dwellings located in Nagoya city, and the real situations of seasonal variation of indoor thoron progeny and radon progeny concentrations are still to be explored.

Figure 3 shows diurnal variations of both thoron progeny and radon progeny concentrations in a dwelling made of concrete. The measurement was carried out from October 1990 to February 1991. The average concentrations of thoron progeny and radon progeny in the period were 0.44 ± 0.11 Bqm^{-3} and 3.09 ± 1.02 Bqm^{-3}, respectively, and the ratio of thoron progeny concentration to radon progeny concentration during this period showed a constant value of approximately 0.14 ± 0.06. The result also suggested that the concentration of thoron progeny indoors might vary analogously with the diurnal radon progeny concentration.

V DOSE EVALUATION OF THORON PROGENY AND RADON PROGENY

When atoms of thoron progeny or radon progeny are inhaled, a major fraction of the atoms is removed from the air stream by impact or diffusion processes and retained on the interior surfaces of the respiratory organ. Those atoms will decay on the surfaces of the airways, applying the major fraction of the radiation dose to the target cells associated with the alpha particles emitted by ^{212}Bi (6.05 and 6.09 MeV) and ^{212}Po (8.78 MeV) for the ^{220}Rn series, and ^{218}Po (6.00 MeV) and ^{214}Po (7.69 MeV) for the ^{222}Rn series.

The effective doses due to thoron progeny or radon progeny are calculated from the equilibrium equivalent concentration of thoron or radon (EEC of Tn or of Rn) and the conversion factor, which is related to the breathing rate and the occupancy factor. In this study, our main interests for dose are to know the difference between doses in different building materials, and to clarify the difference of contribution between thoron progeny and radon progeny. We, therefore, used the EAT and the EAR concentrations described in the former session instead of the EECs of Tn and Rn, and adopted the following conversion factor; If the mean breathing rate is taken as 0.8 m^3h^{-1} (referring to the report of UNSCEAR 1988[2)]) and the occupancy factor is assumed to be 1 (*i.e.* spending 24 h indoors), the conversion factors from EECs of thoron and radon into the annual effective doses are 0.37 mSv $(Bqm^{-3})^{-1}$ and 0.098 mSv $(Bqm^{-3})^{-1}$, respectively. Consequently, the annual effective dose to the inhabitants due to thoron progeny and radon progeny in dwellings made of different kinds of building materials are evaluated as shown in **Table 6** together with the external effective dose obtained in the former session. In the Japanese traditional style dwell-

Table 6 Annual effective dose in mSv.

Material	Internal			External			Total
	^{220}Rn progeny	^{222}Rn progeny	Subtotal	^{232}Th	^{238}U	Subtotal	
Soil	1. 19	0. 82	2. 0	0. 15	0. 084	0. 23	2. 23
Concrete	0. 22	0. 85	1. 1	0. 15	0. 091	0. 24	1. 34
N.B.M.	0. 56	0. 69	1. 2	0. 091	0. 077	0. 17	1. 37

ings of which walls were made of soil, the average contribution of thoron progeny to the annual effective dose was 1.19 mSv, which was slightly higher than that of radon progeny (0.82 mSv). On the other hand, in the dwellings made of concrete and new building materials, the contributions of thoron progeny to the annual effective doses were 0.22 mSv and 0.56 mSv, respectively, which were lower than those of radon progeny. It was reported by UNSCEAR 1988[2] that the worldwide average value of the annual effective dose due to radon and radon progeny was 1.10 mSv, and that due to thoron and thoron progeny was 0.16 mSv. The occupancy factor indoors is 0.8 and outdoors is 0.2. The results of our investigation, even though a different occupancy factor was applied, show that in some dwellings, the radiation exposure due to thoron progeny may be possibly high, or even higher than that of radon progeny.

On the other side, the annual effective doses due to gamma radiation emitted from ^{232}Th and ^{238}U in building materials (0.15–0.09 mSv for ^{232}Th and 0.08–0.09 mSv for ^{238}U) were nearly equal to those shown in the report of UNSCEAR 1988.[2]

The results of this investigation indicate that the annual effective doses due to thoron progeny and radon progeny in dwellings made of soil, especially in Japanese traditional houses, were nearly double the average value shown in the report of UNSCEAR 1988,[2] and those values in dwellings made of other materials were almost the same. These results suggested that the influence of thoron progeny could not be neglected in some kinds of dwellings in Japan, indicating that the effective doses shown in this study tended to be overestimated because a large occupancy factor was assumed in the estimation.

VI CONCLUSION

Throughout the 3-year investigation, thoron progeny and radon progeny concentrations were surveyed in dwellings made of different building materials. It was suggested that thoron progeny concentrations were strongly affected by the type of building material used, whereas the contents of ^{232}Th in these different kinds of building materials were nearly equal. On the other hand, the respective radon progeny concentrations and ^{238}U contents were almost the same in different kinds of building materials. The traditional Japanese dwellings with soil wall showed the highest thoron progeny concentration (3.22 Bqm^{-3}) and the annual effective dose due to thoron progeny was 1.19 mSv, which was higher than that due to radon progeny (0.82 mSv). These results suggested that the radiation exposure due to inhalation of thoron progeny could not be neglected in some dwellings.

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