Journal of Nuclear Science and Technology

ISSN: 0022-3131 (Print) 1881-1248 (Online) Journal homepage: https://www.tandfonline.com/loi/tnst20

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To cite this article: Qiuju GUO , Takao IIDA , Katsumi OKAMOTO & Tadashi YAMASAKI (1995) Measurements of Thoron Concentration by Passive Cup Method and Its Application to Dose, Assessment, Journal of Nuclear Science and Technology, 32:8, 794-803, DOI: 10.1080/18811248.1995.9731775

To link to this article: https://doi.org/10.1080/18811248.1995.9731775

Journal of NUCLEAR SCIENCE and TECHNOLOGY, 32[8], pp. 794~803 (August 1995).

Measurements of Thoron Concentration by Passive Cup Method and Its Application to Dose Assessment

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(Received September 13, 1994), (Revised February 24, 1995)

A new type of passive integrating cup monitors was developed by using a 50 mm radius stainless steel hemisphere to measured indoor radon (^{222}Rn) and thoron (^{220}Rn) concentrations. By placing a pair of cup monitors with an air exchange opening of diameter 5 mm and four openings of diameter 20 mm at a 20 cm distance from wall during about three months, the concentrations of both gases could be assessed from the α track densities on the cellulose nitrate (CN) films. The ^{222}Rn and ^{220}Rn concentrations were surveyed with the cup monitors in the different types of dwellings around Nagoya in Japan over three years. The ^{220}Rn concentrations were rather high in the dwellings with soil wall, and the mean ^{220}Rn concentration was 160 ± 12 Bq·m^{-3}. The ^{220}Rn exhalation rate from wall and the ^{220}Rn diffusion were evaluated from the distribution of ^{220}Rn concentrations in the dwellings. The results of the surveys have also clarified the relationship between the ^{220}Rn concentrations at a 20 cm distance from wall and the ^{220}Rn progeny concentrations measured in the same dwellings. Then, the annual mean effective dose equivalent due to ^{220}Rn progeny was expected to be 0.67 mSv·yr^{-1} in the dwellings with soil wall.

***KEYWORDS:* radon 222, radon 220, radon 220 progeny, radioactivity, passive cup monitor, cellulose nitrate film, calibration, exhalation rate, diffusion coefficients, effective dose equivalents**

I. INTRODUCTION

The passive cup method with solid state nuclear track detectors (SSNTD) has been the main method of indoor radon (^{222}Rn) measurements[1]~[3]. In indoor ^{222}Rn surveys in Japan, quite high ^{222}Rn concentrations were sometimes observed by passive methods[4][5] in Japanese traditional dwellings made of timber frame and soil wall. However, the ^{222}Rn progeny concentrations measured by active methods in the same dwellings were not so high but thoron (^{220}Rn) progeny concentrations were rather high[6]. Moreover, some studies [7]~[10] have made it clear that high ^{220}Rn concentrations were observed near the soil walls in the traditional Japanese dwellings. Since there are the dwellings with soil walls in all over the world, it is important to evaluate the annual effective dose equivalent dose due to ^{220}Rn and its progeny.

Passive ^{220}Rn integrating monitors that are handy, low-priced, and useful for long-term measurements, are suitable for large scale survey of the dwellings with high ^{220}Rn concentration. On the basis of passive ^{222}Rn cup monitor[11] with cellulose nitrate (CN) film (Kodak-Pathé, LR115 type2), we have studied and developed a new type of passive integrat-

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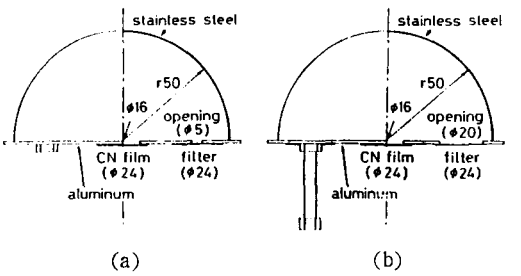
ing ^{222}Rn and ^{220}Rn cup monitors. Indoor ^{222}Rn and ^{220}Rn concentrations can be measured at the same time with the monitors. Another type of passive ^{220}Rn monitor has been developed and reported by Doi *et al.*[9][10][12][13] recently. The monitor has a structure of two 37.5 mm and 60 mm radius hemispheric chambers with polycarbonate films. The separation of ^{222}Rn and ^{220}Rn concentrations is performed by using two step diffusion method and electrochemical etching.

The present paper describes the construction and the characteristics of the new type of passive cup monitors and some small surveys of mean ^{222}Rn and ^{220}Rn concentrations indoors around Nagoya in Japan over three years. Moreover, we propose a simple assessment method of dose due to exposure of ^{220}Rn progeny indoors.

II. Construction of ^{222}Rn and ^{220}Rn Cup Monitors

We have developed several types of cup monitors for measuring indoor ^{222}Rn concentrations[11]. On the basis of the study, a pair of passive integrating ^{222}Rn and ^{220}Rn cup monitors was developed newly. The cross sections of the monitors are shown in **Fig. 1**. Both ^{222}Rn and ^{220}Rn cup monitors have been constructed by mounting a stainless steel hemisphere with radius 50 mm on an aluminum plate bottom, that has a 16 mm inner diameter circular window with a CN film in the center. The ^{222}Rn and ^{220}Rn monitors have different opening diameters on the bottom. A membrane filter (ADVANTEC TOYO, CN, pore size 0.8 μm) was positioned on each of the openings, through it ^{222}Rn or ^{220}Rn gases in the monitor can be exchanged by diffusion naturally. For the ^{222}Rn cup monitor, the exchange rate must be high enough to measure ^{222}Rn concentration and on the other hand must be as small as possible to minimize the influence of ^{220}Rn. For ^{220}Rn measurements, the opening area of ^{220}Rn cup monitor should be as large as possible to make the exchange rate high. By considering the exchange rate and the concentration ratios of ^{222}Rn and ^{220}Rn inside to outside the monitors (see Sec. III-1.), the ^{222}Rn and ^{220}Rn monitors were determined to have a 5 mm exchange opening and four exchange openings of diameter 20 mm, respectively.



(a) (b)

Fig. 1 Passive integrating ^{222}Rn and ^{220}Rn cup monitors (a) for ^{222}Rn measurement and (b) for ^{220}Rn measurement

To avoid the influence of various environmental factors[11] on CN film and to take into account the etchable tracks energy, a 6 μm thick Mylar was fixed inside the CN film. The CN film is etched for 165 min at 60°C in 2.5 N NaOH solution after exposure. The etched tracks are counted using image processing method[14], and the counting area of CN film is 1.0 cm^2.

Both ^{222}Rn and ^{220}Rn concentrations can be measured by putting a pair of the monitors at the same place during a sufficient period of time. Then, the obtained etched-track densities, N_{Rn} and N_{Tn} [tracks·cm^{-2}], on the CN films of ^{222}Rn and ^{220}Rn monitors are represented by the following equations,

$$N_{Rn}=Q_{Rn}CF_{RnA}T+Q_{Tn}CF_{TnA}T+B, \quad (1)$$

$$N_{Tn}=Q_{Rn}CF_{RnB}T+Q_{Tn}CF_{TnB}T+B, \quad (2)$$

where Q_{Rn} and Q_{Tn} are the mean concentrations of ^{222}Rn and ^{220}Rn in Bq·m^{-3}, CF_{RnA} and CF_{TnA} are the ^{222}Rn and ^{220}Rn calibration factors of the ^{222}Rn cup monitor in tracks·cm^{-2} (Bq·m^{-3}·h)$^{-1}$, CF_{RnB} and CF_{TnB} are the ^{222}Rn and ^{220}Rn calibration factors of the ^{220}Rn cup monitor in tracks·cm^{-2}(Bq·m^{-3}·h)$^{-1}$, T is exposure time in h, and B is the background track density of CN film in tracks·cm^{-2}.

Since the ^{222}Rn monitor has the ^{222}Rn or ^{220}Rn gas exchange rate that is small enough to ignore the influence of ^{220}Rn and high enough to measure ^{222}Rn concentration, we could assume that $CF_{RnA} \gg CF_{TnA}$ and $CF_{RnA} = CF_{RnB}$. Moreover, CF_{Rn} and CF_{Tn} are used

for CF_{RnA} and CF_{TnB} hereafter. Consequently, the following equations can be obtained from Eqs. (1) and (2):

$$Q_{Rn}=\frac{N_{Rn}-B}{CF_{Rn}T}, \quad (3)$$

$$Q_{Tn}=\frac{N_{Tn}-N_{Rn}}{CF_{Tn}T}. \quad (4)$$

Therefore, ^{222}Rn and ^{220}Rn concentrations can be obtained from the etched-track densities on the CN films if the calibration factors CF_{Rn} and CF_{Tn} are known.

III. Characteristics of ^{222}Rn and ^{220}Rn Monitors

1. Concentration Ratios of ^{222}Rn and ^{220}Rn Inside to Outside Monitor

Radon-222 or ^{220}Rn gas in the monitor is exchanged naturally by diffusion through the membrane filter. The variation of the concentration of ^{222}Rn or ^{220}Rn inside the monitor can be described by the following differential Equation[15]:

$$\frac{dn_{in}}{dt}=-\lambda n_{in}-\gamma(n_{in}-n_{out}), \quad (5)$$

where n_{in} and n_{out} are the number concentrations of ^{222}Rn or ^{220}Rn gas inside and outside the monitor in m^{-3}, λ is the decay rate of ^{222}Rn or ^{220}Rn in h^{-1} and γ is the exchange rate in h^{-1}. The decay rates of ^{222}Rn and ^{220}Rn are 0.00755 and 45 h^{-1}, respectively. The exchange rate of ^{222}Rn or ^{220}Rn in Eq. (5) is represented by

$$\gamma=\frac{D_f A}{\delta V}, \quad (6)$$

where D_f is the diffusion coefficient of ^{222}Rn or ^{220}Rn in the filter in $m^2 \cdot h^{-1}$, A and δ are its area in m^2 and thickness in m, and V is the interior volume of the monitor in m^3. The D_f/δ value of the membrane filter had been estimated to be 2.09 $m \cdot h^{-1}$ by Iida *et al.*[16]. It is considered that the diffusion coefficient of ^{220}Rn is the same as that of ^{222}Rn.

The mean ^{222}Rn or ^{220}Rn concentration inside the monitor could be calculated by integrating the solution of Eq. (5). If exposure time is very long, ^{222}Rn or ^{220}Rn concentration ratio inside to outside monitor can be expressed by the following equation,

$$R=\frac{\bar{Q}_{in}}{\bar{Q}_{out}}=\frac{\gamma}{\lambda+\gamma}, \quad (7)$$

where $\bar{Q}_{in}$ and $\bar{Q}_{out}$ are the mean ^{222}Rn or ^{220}Rn concentrations inside and outside the monitors in $Bq \cdot m^{-3}$.

Table 1 shows the ^{222}Rn or ^{220}Rn exchange rates of both monitors and ratios of ^{222}Rn and ^{220}Rn concentrations inside to outside the monitors. These values were calculated from Eqs. (6) and (7). For the ^{222}Rn monitor, ^{220}Rn concentration ratio of 0.0035 is very low enough to neglect the influence of ^{220}Rn, and ^{222}Rn concentration ratio decreases only by approximately 4.6%. Then, the ^{222}Rn concentration obtained with the ^{222}Rn monitor is nearly the same of that with the ^{220}Rn monitor. On the other hand, the ^{220}Rn concentration ratio inside to outside the ^{220}Rn monitor is only 0.1822, although the opening area of ^{220}Rn cup monitor was made as large as possible.

2. Calibration of ^{222}Rn Cup Monitors

Radon-222 and ^{220}Rn exposures measured with the monitors are evaluated from the track densities on CN films as expressed in Eqs. (3) and (4). Therefore, it is necessary to investigate the relationship between track densities and ^{222}Rn and ^{220}Rn exposures in order to determine the calibration factors of the two monitors.

Table 1 Exchange rates and ratios of ^{222}Rn and ^{220}Rn concentrations inside to outside the monitors

Type of monitor	Opening		Exchange rate (h^{-1})	Concentration ratio	
	Diameter(mm)	Number		^{222}Rn	^{220}Rn
^{222}Rn	5.0	1	0.1566	0.954	0.0035
^{220}Rn	20.0	4	10.024	0.999	0.1822

Calibrations of the ^{222}Rn monitors were performed by setting ten cup monitors into a 9L stainless steel chamber that was connected with a 1.5 L cylindrical ionization chamber containing high ^{222}Rn concentration and a pump to a circle. The ^{222}Rn was introduced from the ionization chamber into the 9 L chamber by circulation with the pump. The ^{222}Rn cup monitors were exposed in the 9 L chamber for 3 d. The ^{222}Rn concentration at the beginning of exposure was estimated from the current measured with the ionization chamber after circulating. The ^{222}Rn calibration experiments were carried out four times. The mean CF_{Rn} was derived to be $(2.15\pm0.19)\times10^{-3}$ tracks·cm^{-2} (Bq·m^{-3}·h)$^{-1}$. Calibration factor of the ^{222}Rn monitor has also been calculated by random walk method*[17]. The result was 2.08×10^{-3} tracks·cm^{-2} (Bq·m^{-3}·h)$^{-1}$ and agrees well with the experimental value.

3. Calibration of ^{220}Rn Cup Monitors

Calibration of the ^{220}Rn monitors was quite difficult because of the short half-life (55.6 s) of ^{220}Rn. Five pairs of ^{222}Rn and ^{220}Rn monitors were set in a closed vessel that has a diameter of 30 cm and a height of 80 cm. A ^{220}Rn source of thorium powder was put on the bottom of the 60 L cylindrical vessel. In order to make the ^{220}Rn concentration in the vessel as uniform as possible, a small mixing fan has been run weakly during the exposure. The uniformity of the ^{220}Rn concentration in the vessel was confirmed experimentally by using a scintillation cell.

The ^{222}Rn and ^{220}Rn cup monitors were hung along the inner wall at the positions of 45 and 30 cm from the bottom of the 60 L vessel for 7 to 15 d. The aluminum bottom plates of the monitors faced inside. The ^{220}Rn concentrations in the vessel were measured two or three times every day by taking the air at the same height as ^{220}Rn monitors into evacuated scintillation cell. The results of five calibration experiments are shown in **Fig. 2**. The mean CF_{Tn} was $(5.06\pm0.89)\times10^{-4}$ tracks·cm^{-2}(Bq·m^{-3}·h)$^{-1}$. The relatively large uncertainty is mainly due to the difficulty of the ^{220}Rn concentration measurements. Calibration factor of the ^{220}Rn monitor has also been

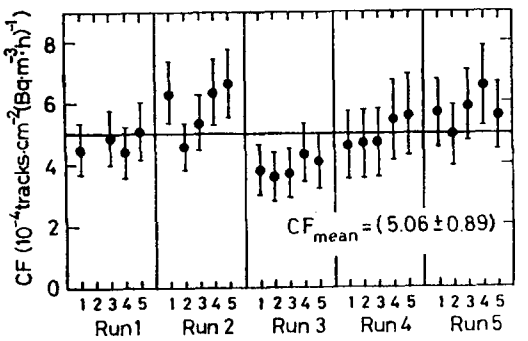


Fig. 2 Experimental results for calibration factor of ^{220}Rn cup monitor

calculated by random walk method[17], considering the ^{220}Rn concentration ratio inside to outside the ^{220}Rn monitor of 0.1822. Then, we assumed that ^{220}Rn is distributed uniformly in the cup monitor. The theoretical value of 4.91×10^{-4} tracks·cm^{-2}(Bq·m^{-3}·h)$^{-1}$ was in agreement with the experimental results.

4. Detection Limits

The background track densities were evaluated by placing both ten cup monitors with CN films and ten CN films in a ^{222}Rn-free vessel[11]. From the experiments, the background track density was found to be

$$B=(B_0\pm\Delta B_0)+(0.0037\pm0.0009)T, \quad (8)$$

where the first term is the inherent background track density, the second term may be the increasing rate of background track density due to α particles from the inner stainless steel wall of the cup monitor, and T is the exposure time in h.

Using the values of the uncertainty in background track density, the counting area, and the calibration factor, the lower detection limit defined by Currie[18] is found to be 6.4 Bq·m^{-3} for an exposure time of 90 d.

* The calibration factor was evaluated as follows. First, the suspension rate of ^{218}Po in the cup monitor and the deposition distributions of ^{218}Po and ^{214}Po on the internal surface of the cup monitor were calculated by molecular diffusion model. Next, the probability of etched-tracks formation of α-particles emitted from ^{222}Rn, ^{218}Po and ^{214}Po atoms were estimated by Monte Carlo simulation. Then, ^{222}Rn is distributed uniformly in cup monitor.

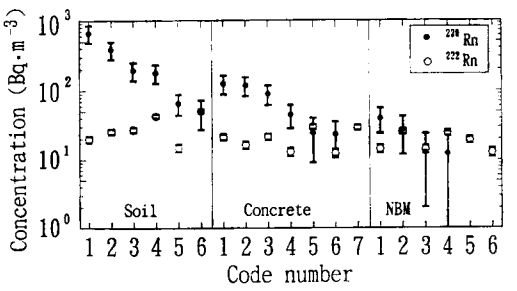
As seen from Eq. (4), the detection limit of the ^{220}Rn cup monitor depends on the uncertainty of the ^{222}Rn concentration instead of the background track density. When we assume that the ^{222}Rn concentration is 10 Bq·m^{-3}, the lower detection limit of the ^{220}Rn monitor is calculated to be 31 Bq·m^{-3} for 90d exposure.

IV. Practical Measurements with the Passive Method

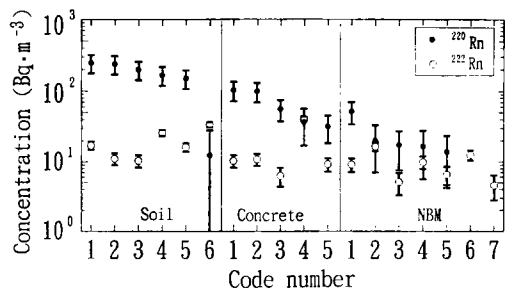
1. Indoor ^{222}Rn and ^{220}Rn Concentrations

Small scale surveys of ^{222}Rn and ^{220}Rn concentrations indoors were performed with the passive integrating cup monitors. Walls of dwellings were considered to be the main source of ^{220}Rn indoors. Therefore, according to the building material walls, dwellings were classified into three types: soil, concrete and new building material (NBM), such as gypsum board and heat insulating material. Most of traditional Japanese dwellings were made of timber frame and soil wall. The concentrations of ^{222}Rn and ^{220}Rn at a distance of 20 cm from wall in different types of dwellings were measured 10 times from Feb. 1991 to Jan. 1994 at intervals of 3 months. During the period, 27 dwellings were surveyed around Nagoya in Japan: 12 of them were made of soil, 7 of them were concrete and the other 7 were NBM.

Two examples of the surveys of indoor ^{222}Rn and ^{220}Rn concentrations from 29 Oct. 1991 to 30 Jan. 1992 and 8 Jun. to 2 Sep. 1992 are shown in **Fig. 3**. The code numbers of cup monitors are written down in order of ^{220}Rn concentration according to dwelling types. Moreover, the results of mean ^{222}Rn and ^{220}Rn concentrations classified by building materials are represented in **Fig. 4**. As shown in Figs. 3 and 4, mean ^{220}Rn concentrations of the dwellings with soil walls were the highest compared with dwellings made of other building materials. On the other hand, mean ^{220}Rn concentrations of NBM dwellings

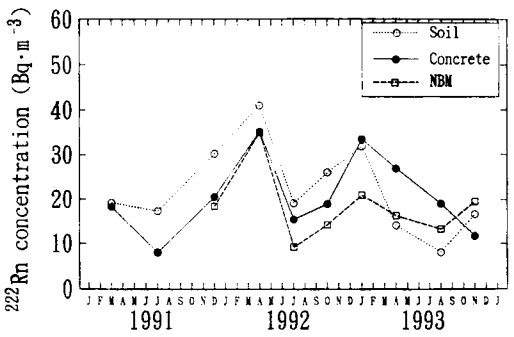


(a)

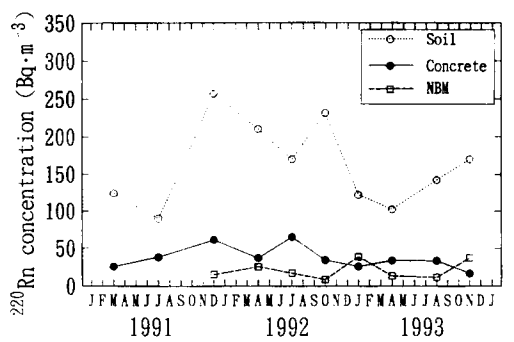


(b)

Fig. 3 Results of surveys (a) from 29 Oct. 1991 to 30 Jan. 1992 and (b) 8 Jun. to 2 Sep. 1992: (●) ^{220}Rn concentration, (○) ^{222}Rn concentration



(a)



(b)

Fig. 4 Mean concentrations of (a) ^{222}Rn and (b) ^{220}Rn classified by building materials

were the lowest, and in some dwellings the ^{220}Rn concentration were below the lower detection limit. On the contrary, no significant difference was found in ^{222}Rn concentrations in different types of building materials. The results indicate that mean ^{222}Rn concentrations in all types of dwellings and mean ^{220}Rn concentrations in the dwellings with soil wall show a clear seasonal variation of a summer minimum and a winter maximum. This trend might depent upon the influence of ventilation indoors.

Indoor ^{222}Rn and ^{220}Rn concentrations are generally dependent on many factors, such as exhalation from wall, the ventilation condition of dwelling and the contents of ^{226}Ra and ^{224}Ra in the building materials. As the half-life time of ^{220}Rn is rather short, exhalation may become the most important factor. The porosity of a soil wall can be assumed to be higher than other building materials and it facilitates ^{220}Rn diffusion. This may be the reason for the high ^{220}Rn concentration in soil wall dwellings.

2. Distribution of ^{220}Rn Concentrations

As the shortage of the half-life of ^{220}Rn, it is assumed that the distribution of ^{220}Rn may be quite different to that of ^{222}Rn. We investigated the dependence of ^{220}Rn concentration on the distance from the wall in a dwelling with soil wall. Six pairs of ^{222}Rn and ^{220}Rn monitors were set at distances of 10, 20, 40, 60, 80, and 100 cm from soil wall for 3 months. The result of the measurements from 31 Oct. 1991 to 28 Jan. 1992 is shown in **Fig. 5**.

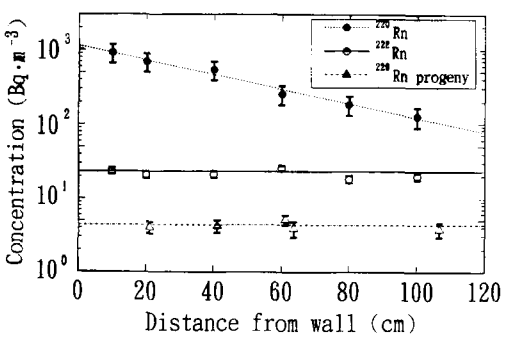


Fig. 5 Indoor ^{220}Rn, ^{222}Rn and ^{220}Rn progeny concentrations as a function of distance from soil wall (29 Oct. 1991 to 30 Jan. 1992)

This figure shows that the ^{222}Rn concentration was independent of the distance from soil wall, but the ^{220}Rn concentration reduced exponentially with as a function of the distance from the wall. This suggests that it is necessary to keep the distance from the soil wall constant when we measure indoor ^{220}Rn concentration.

During the period of ^{222}Rn and ^{220}Rn measurements with cup monitors, the distribution of ^{220}Rn progeny concentration was also measured at the different distance from soil wall by an active filter method[6]. As shown in Fig. 5, ^{220}Rn progeny concentration was nearly independent of the distance from soil wall. The uniformity of ^{220}Rn progeny concentration in dwelling was also confirmed by a model calculation[19].

3. Diffusion and Exhalation of ^{220}Rn

In the same dwelling with soil wall, the measurements of ^{220}Rn concentrations by using six pairs of cup monitors have been continued at intervals of 3 months. The results are shown in **Fig. 6**. The ^{220}Rn concentrations in all seasons decreased exponentially with the distance from wall.

In order to evaluate the distribution of indoor ^{220}Rn concentrations quantitatively, effective diffusion coefficient of ^{220}Rn in dwelling and exhalation rate of ^{220}Rn from wall were evaluated as follows. If the wall was considered to be a infinite plane, ^{220}Rn concentration at a distance of X m from wall could be expressed by the following equation[20]:

$$Q(X)=\frac{E_{Tn}}{\sqrt{\lambda_{Tn}D}}\exp(-\sqrt{\lambda_{Tn}/D})X, \quad (9)$$

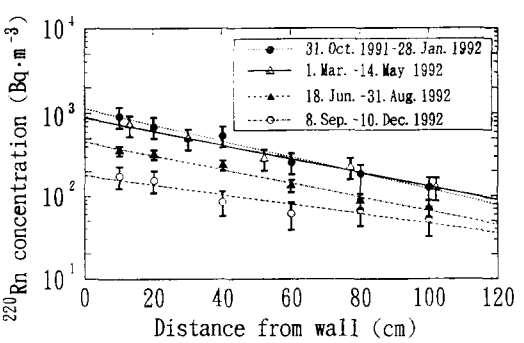


Fig. 6 Distributions of ^{220}Rn concentrations indoors

where $Q(X)$ is the ^{220}Rn concentration at a distance of X m from wall in Bq·m^{-3}, E_{Tn} the ^{220}Rn exhalation rate from wall in Bq·m^{-2}·s^{-1}, λ_{Tn} the ^{220}Rn decay rate in h^{-1}, D the effective diffusion coefficient of ^{220}Rn in air in m^2·s^{-1}, and X the distance from wall in m.

Using the least squares method, the values of E_{Tn} and D have been determined in order to fit the observed distribution of ^{220}Rn concentrations shown in Fig. 6 into Eq. (9). The calculated values of E_{Tn} and D with standard deviations are shown in **Table 2** as the results of dwelling A. In Table 2, the results obtained in dwellings B and C with soil wall and dwelling D made of NBM are also listed. The effective ^{220}Rn diffusion coefficient is generally higher in summer and lower in winter. The change in season may be explained by the change of ventilation indoors. The higher ventilation rate in summer may make ^{220}Rn gas diffusion faster, but in winter ^{220}Rn can not diffuse far from wall before decaying because of lower ventilation rate. As seen from Figs. 4 and 6, the ^{220}Rn concentration in room air is not insensitive to usually used ventilation rate even though the half-life of ^{220}Rn is short.

For the exhalation rate of ^{220}Rn in dwelling A with soil wall, E_{Tn} is higher in winter and lower in summer. The result in dwelling B shows no seasonal variation. As ^{220}Rn gas is exhaled from the top few cm of wall[21], it is expected that the nature of the surface layer has a great influence on the ^{220}Rn exhalation rate. The water content in building materials in the main factor having effects upon ^{220}Rn exhalation from wall. In winter, the reason of the high ^{220}Rn exhalation rate in dwelling A may be due to the low water content of soil wall.

Exhalation rate of ^{220}Rn from soil wall in Table 2 shows more than ten times as much as that from brick and heavy concrete[22], 0.01 to 0.11 Bq·m^{-2}·s^{-1}. Therefore, it can be concluded that high ^{220}Rn concentration in the dwelling with soil wall arises from the high ^{220}Rn exhalation rate from wall.

V. Dose Assessment Method

In this Chapter, we will estimate indoor ^{220}Rn progeny concentration from the ^{220}Rn concentration measured with passive cup monitors at a distance of 20 cm from wall, and evaluate annual effective dose equivalent due to exposure of ^{220}Rn and ^{220}Rn progeny indoors.

1. Estimation of ^{220}Rn Exhalation Rate from ^{220}Rn Concentrations

Distribution of ^{220}Rn concentrations in dwellings is affected by effective diffusion coefficient of ^{220}Rn and exhalation rate of ^{220}Rn from wall. Equation (9) can be rewritten as follows:

$$E_{Tn}=Q(X)\sqrt{\lambda_{Tn}D}\exp(\sqrt{\lambda_{Tn}/D})X. \quad (10)$$

As shown in **Table 2**, effective diffusion coefficient ranges from 1.2 to 7.4×10^{-3} m^2·s^{-1}. If we measured ^{220}Rn concentrations at a distance

Table 2 Effective ^{220}Rn diffusion coefficient in dwelling and ^{220}Rn exhalation rate from wall estimated from distributions of ^{220}Rn concentrations indoors

Dwelling	Measurement period	D(10^{-3} m^2·s^{-1})	E(Bq·m^{-2}·s^{-1})
A	31 Oct. 91-28 Jan. 92	2.5±0.8	6.4±1.7
	1 Mar. 92-14 May 92	3.4±1.5	5.8±1.7
	18 Jun. 92-31 Aug. 92	6.3±3.9	1.6±0.6
	8 Sep. 92-10 Dec. 92	3.3±0.8	3.0±0.5
B	8 Jun. 92- 7 Sep. 92	7.4±4.5	2.5±0.9
	8 Sep. 92- 7 Dec. 92	3.6±0.9	2.2±0.4
	29 Jun. 93-23 Sep. 93	2.7±0.9	2.8±0.7
	7 Oct. 93- 5 Jan. 94	1.2±0.5	2.7±0.9
C	11 Oct. 93- 1 Jan. 94	3.5±2.3	1.1±0.5
D	8 Oct. 93-10 Jan. 94	1.5±0.8	0.8±0.4

of 20 cm* from wall, $Q(0.2)$, exhalation rate of ^{220}Rn from wall can be calculated to be in ranging from 0.0074 $Q(0.2)$ to 0.0124 $Q(0.2)$ from Eq. (10). Thus, when effective diffusion coefficient varies by a factor 7, ^{220}Rn exhalation rate could be estimated by uncertainty of only ±25%. Therefore, Eq. (10) can approximate with a following equation,

$$E_{Tn}=0.0099\ Q(0.2). \quad (11)$$

As described above, it is possible to estimate ^{220}Rn exhalation rate from wall by using the ^{220}Rn concentration measured with passive cup monitors at a distance of 20 cm from wall.

2. Relationship between ^{220}Rn and ^{220}Rn Progeny Concentrations

Since ^{212}Pb (ThB) has relatively long half-life of 10.6 h in ^{220}Rn progeny, ^{212}Pb is the most important isotope controlling the radiation exposure due to ^{220}Rn progeny. The ^{212}Pb concentration in dwelling is expressed as follows[15]:

$$C_B=\frac{\lambda_a\lambda_B N_B S/V+\lambda_v C_{BO}}{\lambda_B+\lambda_v+\lambda_d}, \quad (12)$$

where N_B is defined by

$$N_B=E_{Tn}[\lambda_{Tn}(\lambda_B+\lambda_a+\sqrt{\lambda_{Tn}(\lambda_B+\lambda_a)})], \quad (13)$$

C_B is the indoor ^{212}Pb concentration in Bq·m^{-3}, λ_a the attached coefficient of free nuclei to aerosol particles in s^{-1}, λ_B the decay rate of ^{212}Pb in s^{-1}, S the surface area of wall in m^2 from which ^{220}Rn exhales, V the volume of the room in m^3, λ_v the ventilation rate of the room air in s^{-1}, C_{BO} outdoor ^{212}Pb concentration in Bq·m^{-3}, and λ_d the deposition rate of attached ^{212}Pb atoms in s^{-1}. As seen from Eq. (12), ^{212}Pb concentration varies mainly in dependence on λ_v and λ_d[19].

Here, we assume that the ratio of surface area exhaling ^{220}Rn to volume of the room is about 1/3 and does not vary on dwellings. The values of the another parameters are assumed to be; $\lambda_a=0.0139$ s^{-1}[23], $\lambda_v=1.39\times10^{-4}$ s^{-1}[24] and $\lambda_d=5.6\times10^{-5}$ s^{-1}[23]. Moreover, ^{220}Rn progeny concentration is equal to about 1.1 times ^{212}Pb, if ^{212}Pb and ^{212}Bi are in equilibrium. Consequently, indoor ^{220}Rn progeny concentration could be expressed approximately to be proportional to ^{220}Rn exhalation rate as follows:

$$C_{TP}=1.3\ E_{Tn}, \quad (14)$$

where C_{TP} is the ^{220}Rn progeny concentration in Bq·m^{-3}. By substituting E_{Tn} of Eq. (11), Eq. (14) can be written as follows:

$$C_{TP}=0.013\ Q(0.2). \quad (15)$$

Equation (15) indicates that it is possible to estimate indoor ^{220}Rn progeny concentration from the ^{220}Rn concentration measured with passive cup monitors at a distance of 20 cm from wall.

3. Comparison of Measured ^{220}Rn and ^{220}Rn Progeny Concentrations

The measurement of ^{220}Rn progeny concentrations was also one of the subjects in our survey. In some dwellings, ^{220}Rn progeny concentrations were measured by an active filter method[6] and with an integrating ^{220}Rn progeny monitor[25] during the surveys of ^{220}Rn concentrations with passive cup monitors. **Figure 7** shows the relationship between measured ^{220}Rn and ^{220}Rn progeny concentrations. In Fig. 7, the broken line is the lower detection limit of ^{220}Rn concentration measured with a pair of cup monitors. The correlation coefficient between ^{220}Rn and ^{220}Rn progeny concentrations was 0.79. This result was expected as described in Sec. IV-2. From the value of the regression coefficient in Fig. 7, relationship between ^{220}Rn progeny and ^{220}Rn concentrations is given by

$$C_{TPm}=0.015\ Q(0.2)_m, \quad (16)$$

where C_{TPm} and $Q(0.2)_m$ are the measured concentrations of ^{220}Rn progeny and ^{220}Rn, respectively. The relation of Eq. (16) gives fairly good agreement with the theoretical relation in Eq. (15).

* If ^{220}Rn concentration could be measured with same accuracy at every distance, the most suitable distance is 50 cm from wall. The range of ^{220}Rn exhalation rate deduced from Eq. (10) decreases with increasing distance below 50 cm. On the other hand, the accuracy of the measured ^{220}Rn concentration becomes better with decreasing distance. Then, we selected the distance of 20 cm, considering the range, the accuracy and the ease of setting the monitors.

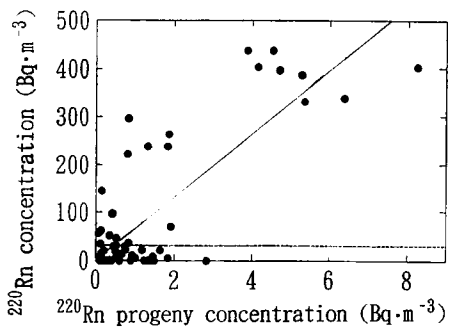


Fig. 7 Relationship between measured ^{220}Rn and ^{220}Rn progeny concentrations

4. Estimation of Annual Effective Dose Equivalent

Annual effective dose equivalents due to ^{222}Rn and ^{220}Rn and their progeny were estimated by using the measured values of ten times small scale surveys. The results classified by building materials are shown in **Table 3**. The effective dose equivalent factors of ^{222}Rn and ^{220}Rn gases are given to be 1.8 and 0.96 μSv·yr^{-1}(Bq·m^{-3})$^{-1}$[26], respectively. The dose factors of ^{222}Rn and ^{220}Rn progeny are also estimated to be 70 and 280 μSv·yr^{-1} per Bq·m^{-3}(EEC : equilibrium equivalent concentration) using a mean breathing rate of 0.8 m^3·h^{-1} indoors and an average occupancy factor of 0.8 indoors[26]. For calculating ^{222}Rn progeny concentrations, an equilibrium factor between ^{222}Rn and its progeny was assumed to be 0.4 indoors. On the other hand, ^{220}Rn progeny concentration were calculated by using the relationship between ^{220}Rn and its progeny concentrations shown in Fig. 7. For reference, annual outdoor dose equivalents are also shown in Table 3.

UNSCEAR 1988 Report[26] gives the typical annual effective dose equivalents of 1.10 mSv·yr^{-1} for ^{222}Rn and its progeny and 0.16 mSv·yr^{-1} for ^{220}Rn and its progeny. As shown in Table 3, the estimated dose equivalents for ^{222}Rn and its progeny were similar in all dwellings built with different building materials, and a little lower than the value of UNSCEAR. The annual effective dose equivalents due to ^{220}Rn in Table 3 were estimated by using the concentrations measured at a 20 cm distance from wall. Since the ^{220}Rn concentration decreases exponentially with the distance, the dose equivalent can be ignored at a distance of more than 100 cm from wall. The annual effective dose equivalents due to ^{220}Rn progeny in the dwellings of concrete and NBM were very low, and nearly the same level as 0.16 mSv·yr^{-1}. However, in the

Table 3 Estimation of annual effective dose equivalents due to ^{222}Rn and ^{220}Rn and their progeny

Building materials	^{222}Rn		^{222}Rn progeny	
	Concentration (Bq·m^{-3})	H_{eff} (μSv·yr^{-1})	Concentration (Bq·m^{-3})	H_{eff} (μSv·yr^{-1})
Soil	19.1± 0.9	34.4± 1.6	7.64±0.36	530±30
Concrete	23.3± 1.5	41.9± 2.7	9.32±0.60	650±40
NBM	17.5± 1.2	31.5± 2.1	7.00±0.48	490±30
Outdoor			4.1†	70
Building Materials	^{220}Rn††		^{220}Rn progeny	
	Concentration	H_{eff}	Concentration	H_{eff}
Soil	159.7±12.4	153.3±11.9	2.40±0.19	670±50
Concrete	41.9± 3.3	40.2± 3.2	0.63±0.05	180±10
NBM	22.9± 3.0	22.0± 2.9	0.34±0.05	100±10
Outdoor			0.1†	9

† Typical outdoor concentrations in Nagoya[27].
†† The H_{eff} values due to ^{220}Rn are estimated by using the concentrations measured at a 20 cm distance from wall. The H_{eff} at a distance of more than 100 cm from wall can be ignored, since ^{220}Rn concentration decreases exponentially with the distance.

dwellings with soil wall, the estimation shows that the average dose equivalent due to ^{220}Rn progeny was calculated to be 0.67 mSv·yr^{-1} and may be equal to that due to ^{222}Rn and its progeny. It may sometimes leads to doses high enough to cause concern for human health.

VI. CONCLUSIONS

A pair of passive cup monitors with a different air exchange openings was developed for measuring simultaneously ^{222}Rn and ^{220}Rn concentrations. Experimental calibration factors of the ^{222}Rn and ^{220}Rn monitors were $(2.15\pm0.19)\times10^{-3}$ and $(5.06\pm0.89)\times10^{-4}$ tracks ·cm^{-2}(Bq·m^{-3}·h)$^{-1}$ and agree well with the calculated values. The three years surveys proved that the handy, low-priced passive cup monitors are useful for long-term measurements and suitable for measuring ^{222}Rn and ^{220}Rn concentrations in various living environments.

Indoor ^{220}Rn concentrations were very high in traditional Japanese dwellings with soil wall. The ^{220}Rn concentration decreases exponentially with the distance from wall. The effective diffusion coefficient of ^{220}Rn in dwelling and the exhalation rate of ^{220}Rn from wall were evaluated from the distribution of the ^{220}Rn concentrations. Then, indoor ^{220}Rn progeny concentration could be estimated from the ^{220}Rn concentration at a 20 cm distance from wall. From the results of the surveys, the average annual effective dose equivalent due to ^{220}Rn progeny was expected to be 0.67 mSv·yr^{-1} in the traditional Japanese dwellings.

For the affection by relatively high background track density of CN film, the lower detection limit was not low enough in some cases. At the present, the authors have been studying the characteristics of passive cup monitors with CR-39 detectors and surveying indoor ^{222}Rn and ^{220}Rn concentrations by using the monitors.

ACKNOWLEDGMENT

The authors would like to appreciate Prof. R.J. de Meijer and Dr. E.R. van der Graaf of Kernfysisch Versneller Instituut, Rijksuniversiteit Groningen, the Netherlands, for their kind support in carrying out the ^{220}Rn calibration experiment.

—REFERENCES—

(1) ALTER, H. W., FLEISCHER, R. L.: *Health Phys.*, **40**, 693 (1981).
(2) URBAN, M., PIESCH, E.: *Radiat. Prot. Dosim.*, **1**, 97 (1981).
(3) BARTLETT, D. T., *et al.*: *ibid.*, **17**, 139 (1986).
(4) YONEHARA, H., *et al.*: *ACS Symp. Ser.*, **331**, p. 172 (1987), Washington, DC.
(5) KOBAYASHI, S., *et al.*: *Proc. 3rd Int. Symp. on Advanced Nucl. Energy Res., Mito, Japan*, p. 63 (1991).
(6) GUO, Q., *et al.*: *Radiat. Prot. Dosim.*, **45**, 357 (1992).
(7) IIDA, T., SONE, K., IKEBE, Y.: *Proc. 25th Annu. Meetings of Japan Health Phys. Soc.*, p. 63 (1990), [in Japanese].
(8) YONEHARA, H., AOYAMA, T.: *Proc. 26th Annu. Meetings of Japan Health Phys. Soc.*, p. 16 (1991), [in Japanese].
(9) DOI, M., KOBAYASHI, S., FUJIMOTO, K.: *Radiat. Prot. Dosim.*, **45**, 425 (1992).
(10) DOI, M., KOBAYASHI, S.: *Health Phys.*, **66**, 43 (1994).
(11) IIDA, T., *et al.*: *ibid.*, **69**[3], 1 (1995).
(12) DOI, M., KOBAYASHI, S.: *ibid.*, **66**, 274 (1994).
(13) DOI, M., KOBAYASHI, S.: *Hoken Butsuri (J. Japan Health Phys. Soc.)*, **29**, 155 (1994).
(14) NURISHI, R., *et al.*: *Nihon-Genshiryoku-Gakkai Shi (J. At. Energy Soc. Japan)*, **36**, 133 (1994), [in Japanese].
(15) WARD, W. J., *et al.*: *Rev. Sci. Instrum.*, **58**, 1440 (1977).
(16) IIDA, T., *et al.*: *Health Phys.*, **54**, 139 (1988).
(17) NURISHI, R.: Master's Thesis, Nagoya Univ., 1994, [in Japanese].
(18) CURRIE, L. A.: *Anal. Chem.*, **40**, 586 (1968).
(19) YAMASAKI, T., GUO, Q., IIDA, T.: *Radiat. Prot. Dosim.*, **59**, 135 (1995).
(20) KATASE, A., *et al.*: *Health Phys.*, **54**, 283 (1988).
(21) SCHERY, S. D.: *J. Air Waste Manage. Assoc.*, **40**, 493 (1990).
(22) FOLKERTS, K. H., KELLER, G., MUTH, H.: *Radiat. Prot. Dosim.*, **9**, 27 (1984).
(23) PORSTENDÖRFER, J.: *ibid.*, **7**, 107 (1984).
(24) MATSUO, Y., *et al.*: *"Kenchiku to Kisho (Building and Weather)"*, p. 89 (1986), Asakura, [in Japanese].
(25) YAMASAKI, T., IIDA, T.: *Health Phys.*, **68**, 840 (1995).
(26) UNSCEAR: *"Sources, Effects and Risks of Ionizing Radiation"*, p. 95, 102, 108 (1988), New York, UN.
(27) YAMASAKI, T., *et al.*: *Hoken Butsuri (J. Japan Health Phys. Soc.)*, (in printing), [in Japanese].