SOME PROBLEMS ON THE MEASUREMENT OF ^{222}Rn CONCENTRATIONS BY PASSIVE CUP METHOD

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Abstract—**A passive cup monitor has been constructed by using a 50 mm radius stainless steel hemisphere. The conductive housing has reduced the scatter in track densities. In dwellings with high ^{220}Rn concentrations, it is particularly necessary to measure ^{222}Rn concentration with a monitor having a radon exchange rate less than 0.1 h^{-1}.**

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Key words: ^{222}Rn; ^{220}Rn; radon; detector, nuclear-track

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INTRODUCTION

PASSIVE ^{222}RN monitors that are handy, low-priced, and useful for long-term measurements are suitable for measuring mean ^{222}Rn concentration levels in various living environments. The passive cup method with solid state nuclear track detectors (SSNTD) has been the main method of indoor ^{222}Rn measurement (Alter and Fleischer 1981; Urban and Piesch 1981; Bartlett et al. 1986). In indoor ^{222}Rn surveys in Japan, quite high ^{222}Rn concentrations were sometimes observed by passive methods (Yonehara et al. 1987; Kobayashi et al. 1991) in Japanese traditional dwellings made of wood and soil walls. High ^{220}Rn progeny concentrations have been often observed in such dwellings (Guo et al. 1992). These results suggest that some of the high ^{222}Rn concentrations measured by passive methods might be influenced by high ^{220}Rn exhalation from soil walls.

The authors have developed several types of cup monitors for measuring indoor ^{222}Rn concentrations. The present paper describes some problems with measurements of ^{222}Rn concentrations by the passive cup method, and the characteristics and the performances of the cup monitors designed to minimize these problems.

PASSIVE CUP METHOD

Principle and some problems

The relationship between ^{222}Rn exposure and track density on a SSNTD is given by

$$Q = \frac{N - BA}{CF\,AT} \tag{1}$$

where Q is the mean ^{222}Rn concentration in Bq m^{-3}, N is the number of tracks counted in field A, B is the background track density in cm^{-2}, A is counting area in cm^2, CF is the calibration factor in tracks cm^{-2}(Bq m^{-3} $h)^{-1}$, and T is exposure time in h.

The track density due to alpha particles and the background track density on SSNTDs are believed to follow the Poisson distribution. However, the uncertainty of the cup method was always greater than could be accounted for by Poisson counting statistics, even if the cup was exposed to high ^{222}Rn concentrations under controlled conditions (Nelson 1987; Pearson et al. 1992). The sources of error have been considered to be associated with the etching process and track counting.

In order to evaluate ^{222}Rn concentrations with the passive cup monitor, we need to consider the following factors with regard to SSNTD: (1) unchanged quality for a long time; (2) reproducibility of the etching process and; (3) low background track density, and, moreover, with regard to measurement by the cup method; (4) exposure time; (5) effect of environmental factors such as UV light, temperature, and humidity; (6) shape and material of cup; (7) alpha particles emitted from the inner wall of cup; and (8) the influence of ^{220}Rn. In this study, we used cellulose nitrate (CN) film‡ for SSNTD that is easy to handle and has a relatively low background track density.

^{222}Rn cup monitor

We have developed several types of passive cup monitors. One was a sealed polyethylene cup of low alpha emitting material. However, this cup monitor did not yield a good linear relationship between track density and ^{222}Rn exposure. Electrostatic fields within the monitors might affect the plate-out distribution on ^{222}Rn progeny (Nelson 1987), because more than 80% of ^{218}Po atoms have a positive charge when formed from ^{222}Rn (Leung and Phillips 1987). According to measurements with a surface potential meter, most of the polyethylene

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‡ LR115 type2, Kodak-Pathé, 8-26 rue Villiot 75594, Paris Cedex, 12, France.

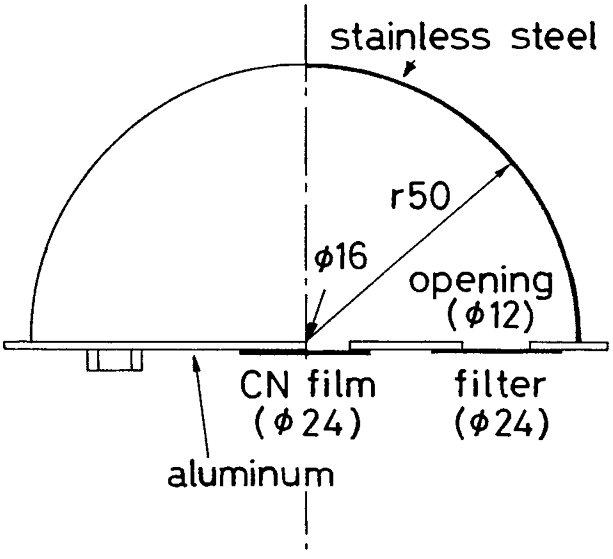

Fig. 1. Cross-section of hemispherical-shaped ^{222}Rn cup monitor.

cups were somewhat negatively charged. We have electrified negatively the inner bottom wall of cups by rubbing with cloth. The track densities obtained with the cups that were set in air with high ^{222}Rn concentrations increased in proportion to the surface potential. These results show that the variation in track densities may be mainly due to the effect of electrostatic fields within the monitors.

It is advisable that the cup wall material be conductive and low alpha emitting. Stainless steel has the lowest alpha emitting rate in metals (Al-Bataina and Janecke 1987). A hemispherical shape is suitable for a cup monitor to arrange the incident alpha particles on the SSNTD in approximately the same energy as the ^{222}Rn progeny deposited on the inner wall of the monitor, since the number of detectable tracks on CN film depends mainly on the incident alpha energy (Jönnson 1981). An air layer of 43–66 mm is required to reduce the 7.69 MeV alpha particles emitted from ^{214}Po atoms to the energy of 0.5–4.2 MeV that produces etchable tracks on CN film (Nurishi et al. 1994).

In view of these considerations, the passive ^{222}Rn cup monitor was constructed by mounting a stainless steel hemisphere with a 50 mm radius on an aluminum plate with a 16 mm inner diameter circular window and a CN film in the center. Offset from the center is a 12 mm diameter circular opening in front of which a membrane filter[§] (Pore size 0.8 μm) is positioned. The cross section of the passive monitor is shown in Fig. 1. The 12 mm diameter was determined by considering the ^{222}Rn exchange rate as follows. ^{222}Rn atoms in the monitor are exchanged by diffusion through the membrane filter. The ^{222}Rn exchange rate (Iida et al. 1988) of the monitor is represented by

$$\gamma = \frac{DA}{\delta V}, \tag{2}$$

where γ is the ^{222}Rn exchange rate in h^{-1}, D is the diffusion constant of ^{222}Rn in the filter in $m^2\ h^{-1}$, 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/δ value of the membrane filter was estimated to be 2.09 m h^{-1} by Iida et al. (1988). When the window diameter is 12 mm, the ^{222}Rn exchange rate γ is derived to be 0.90 h^{-1} by eqn (2). It is considered that the exchange rate for ^{220}Rn is the same as that for ^{222}Rn. When the radon concentration outside the cup monitor n_{out} is constant, the inside concentration n_{in} is given by

$$n_{\text{in}} = \frac{\gamma}{\lambda + \gamma}\{1 - \exp[-(\lambda + \gamma)t]\}n_{out}, \tag{3}$$

where λ is the radon decay rate in h^{-1}. Since the decay rates of ^{222}Rn and ^{220}Rn are 0.00755 h^{-1} and 45 h^{-1}, respectively, the ^{222}Rn and ^{220}Rn concentrations inside the monitor approach about 99% and 2.0% of the outside concentrations after enough time has elapsed. Therefore, one could ignore the influence of ^{220}Rn on the measurement of ^{222}Rn concentrations except when extremely high ^{220}Rn concentrations are encountered.

The CN film was etched for 165 min at 60°C in 2.5N NaOH solution. To reduce the error associated with the etching process, we have always etched test CN films with a piece of CN film exposed to 4.2 MeV alpha particles, the critical energy to produce an etchable track on CN film (Nurishi et al. 1994). The etched tracks are counted in an area of 1 cm^2 by using a microscope and image processing method.

Characteristics

In order to measure ^{222}Rn exposure with the cup monitor, it is necessary to evaluate the calibration factor and the background track density in eqn (1). Calibrations were performed by setting ten cup monitors in a 9 L stainless steel chamber connected with a 1.5 L cylindrical ionization chamber containing high ^{222}Rn concentration. The ^{222}Rn was introduced from the ionization chamber into the 9 L chamber by circulation with a pump. The monitors inside the 9 L chamber were exposed for 3 d, and the ^{222}Rn concentration at the beginning of exposure was measured with the ionization chamber. The mean calibration factor was derived to be $CF = (2.42 \pm 0.08) \times 10^{-3}$ [tracks cm^{-2}(Bq m^{-3} h)$^{-1}$].

Background track densities were evaluated as follows. Ten cup monitors with CN films and ten CN bare films were placed in a ^{222}Rn-free vessel three times for 71, 108, and 83 d. From these experiments, the background track density was found to be

$$B = (B_0 \pm \Delta B_0) + (0.0037 \pm 0.0009)T, \tag{4}$$

[§] Cellulose nitrate membrane filter, ADVANTEC TOYO, 1-5-10, Kotobuki, Daito-ku, Tokyo 111, Japan.

where the first term is the inherent background track density including etching and counting processes for each CN film, and the second term may be the increasing rate of background track density due to alpha 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 detection limit defined by Currie (1968) is found to be 6.4 Bq m^{-3} for an exposure time of 90 d.

PRACTICAL APPLICATION

Mean ^{222}Rn concentrations in different types of dwellings were measured to test the performance of the ^{222}Rn cup monitor for 3-mo intervals from May 1989 to February 1990. The results of the test survey from November 1989 to February 1990 are shown in Fig. 2. The locations of the survey were 17 dwellings made of wood and soil wall, 11 dwellings made of concrete, and 3 dwellings made of new building materials (NBM) such as plaster board. One monitor was set in each dwelling except for a concrete laboratory, in which five monitors were set at the same place to study the variations. The code numbers of cup monitors in Fig. 2 are written down in order of ^{222}Rn concentration according to dwelling types. Average ^{222}Rn concentration levels of two other surveys in summer and autumn were about 2/3 of the winter survey shown in Fig. 2.

To confirm the reliability of the data obtained with cup monitors, an electrostatic integrating ^{222}Rn monitor (EIRM) was placed in each of three house types. The EIRM used was not affected very much by ^{220}Rn (see Appendix). The mean ^{222}Rn concentrations measured with five cup monitors of code number 7–11 in the concrete laboratory building and a cup monitor of code number 1 in the NBM house agree well with the EIRM results. However, the mean ^{222}Rn level obtained with a cup monitor of code number 1 in the wooden house was about 2 times that obtained with the EIRM. A similar tendency was also seen in both summer and autumn surveys. Very high ^{220}Rn progeny concentrations of 3.0–6.3 Bq m^{-3} have been observed in the wooden house (Guo et al. 1992). These results suggest that, in a wooden house, the measurement of ^{222}Rn concentration with the cup monitor may be affected by only about 2% of ^{220}Rn concentration outside of the monitor.

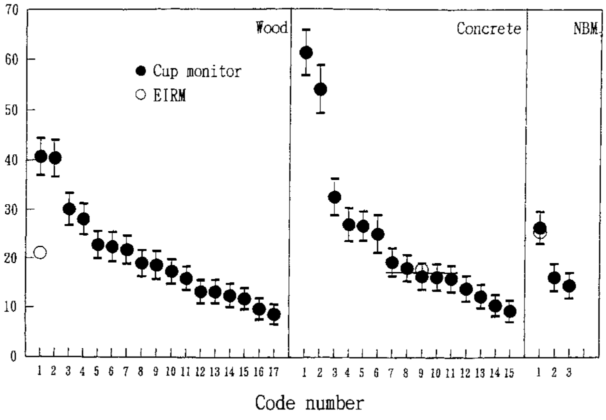

Fig. 2. The results of the test survey from November 1989 to February 1990.

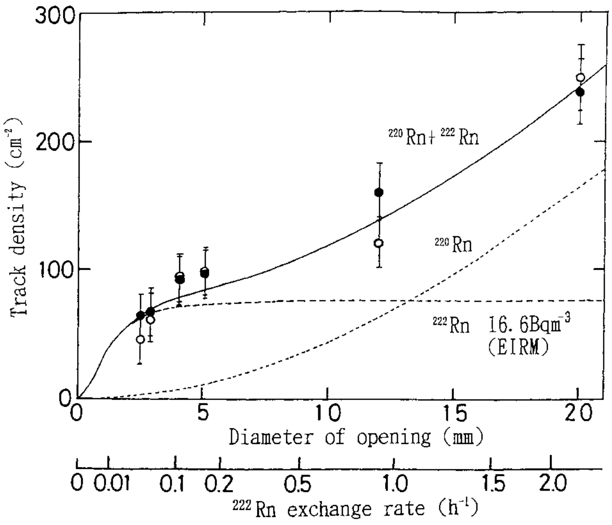

Fig. 3. The influence of ^{220}Rn exhaling from a soil wall on ^{222}Rn concentration measurements. The dashed and dotted lines represent the expected track densities due to ^{222}Rn and ^{220}Rn, respectively. The solid line is the sum.

To verify this supposition, the following measurements were carried out in the wooden house. We made 6 types of cup monitors with different opening diameters of 2.6, 3.0, 4.1, 5.1, 12, and 20 mm by twos. From eqn (2), the radon exchange rates of these monitors are estimated to be 0.042, 0.057, 0.105, 0.163, 0.90, and 2.51 h^{-1}, respectively. The cup monitors and an EIRM were exposed at a distance of 20 cm from the soil wall for 1 mo. The result is illustrated in Fig. 3. It is clear that track density depends on the opening diameter. The dashed line represents the expected track density with a cup monitor that was calculated, based on the ^{222}Rn concentration measured with the EIRM. On the other hand, the dotted line shows the track density due to ^{220}Rn that can be calculated from assuming that the difference between the track density measured with cup monitors of opening diameter 20 mm and expected track density resulting from ^{222}Rn concentration is caused by ^{220}Rn. The sum of the dashed and dotted lines is shown by a solid line in Fig. 3. The observed track densities from cup monitors agree well with the solid line. From the measurement, the mean ^{220}Rn concentration at a distance of 20 cm from the soil wall was estimated to be more than 1,000 Bq m^{-3} by using the radon exchange rate and a calculated cup monitor calibration factor for ^{220}Rn.

CONCLUSIONS

A passive cup monitor is a simple and handy method for measuring ^{222}Rn concentration. A passive monitor has some unclear factors. In the present study, some problems with the cup method were clarified. It is necessary that the cup wall material be conductive and low alpha emitting. A 50 mm radius hemispherical shape is suitable for a cup monitor. The ^{222}Rn concentrations in various living environments differ with the location and season. It is important that the observed track density is not affected by various environmental factors, especially ^{220}Rn. In the dwellings like typical Japanese houses that sometimes have high ^{220}Rn concentrations, care is required to measure ^{222}Rn concentration by the passive cup method. In those dwelling a cup monitor with a radon exchange rate less than 0.1 h^{-1} should be used or both ^{222}Rn and ^{220}Rn concentrations should be elevated at the same time by using two monitors with different radon exchange rates.

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Appendix

Electrostatic integrating ^{222}Rn monitor

Fig. A1 shows the EIRM (GS-201B[||]) which has almost the same construction as that developed by Iida et al. (1988). The EIRM has a volume of about 2.26 L. ^{222}Rn atoms are exchanged by diffusion through a membrane filter of 30 mm diameter positioned at the bottom. ^{222}Rn decays by emission of an alpha particle to ^{218}Po, and most of ^{218}Po atoms are positively charged. The positive ^{218}Po ions are collected electrostatically on the electrode. The 6.0 MeV alpha particles emitted from ^{218}Po atoms can be detected with the CN film set in the electrode. On the other hand, the 7.69 MeV alpha particles from ^{214}Po atoms do not produce etchable tracks on CN film according to the structure of the collecting electrode. The inside of the EIRM was dehumidified with a P_2O_5 drying agent, since the electrostatic collection of ^{218}Po^{+} atoms depends on the humidity in air.

The measured radon exchange rate of the EIRM was 0.67 h^{-1}. Therefore, the mean ^{222}Rn and ^{220}Rn concentrations inside the monitor are expected to be about 99% and 1.5% of the outside concentrations. Thus, ^{222}Rn concentration could be measured correctly. On the other hand, only 1.5% of the outside ^{220}Rn atoms could pass through the filter. Moreover, more than four fifths of the atoms decay during diffusion at a distance of 45 mm from the filter to the mesh because the diffusion coefficient of radon atom in air is about 1×10^{-5} m^2 s^{-1} (Chamberlain 1991). Positive ^{220}Rn progeny ions produced only in the collecting volume over the mesh are collected on the electrode. In the ^{220}Rn progeny, the only 6.05 MeV alpha particles from ^{212}Bi can be detected with CN film in the electrode. ^{212}Bi atoms emit alpha particles in 36% of decays. Therefore, the EIRM are not affected very much by ^{220}Rn.

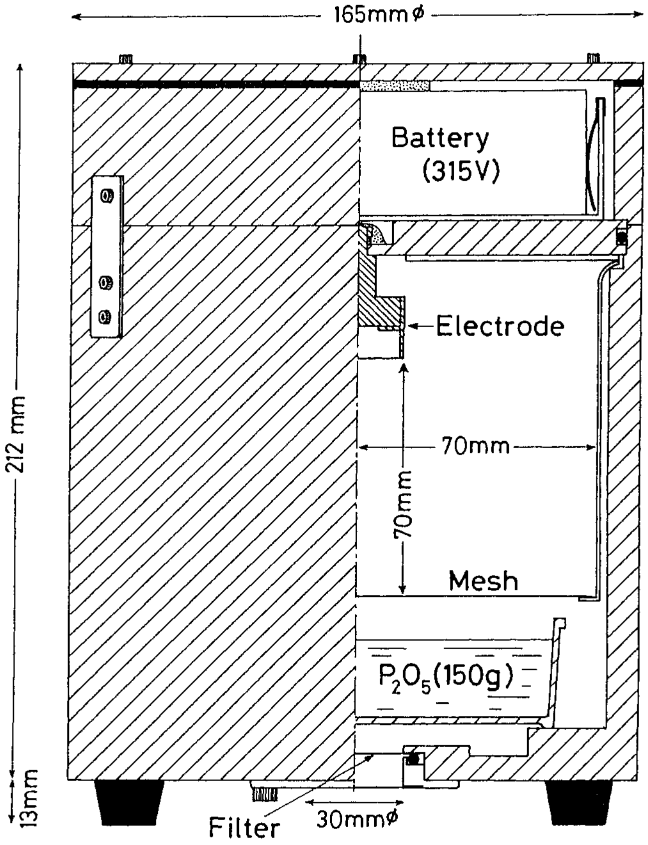

Fig. A1. Electrostatic integrating ^{222}Rn monitor.

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[||] GS-201B, Aloka Co., LTD., 6-22-1, Mure, Mitaka-shi 181, Japan.