Original

Radon Measurement Using a Liquid Scintillation Spectrometer

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ABSTRACT

A convenient radon detecting device for the purpose of estimating natural radiation exposure is described. The α radioactivity of radon gas adsorbed in fine active carbon particles exposed to air is measured with a liquid scintillation spectrometer (Packard-PICO-RAD system). Its detection limits are 2 mBq/ℓ in air and 0.5 Bq/ℓ in water with an accuracy of about 10%. Radon concentrations at Misasa hot springs in Tottori prefecture were measured using this method. They were 0.16~7.7 Bq/ℓ in a bath room and 0.057~0.36 Bq/ℓ outdoors. Radon concentrations of the hot springs were 82~1,700 Bq/ℓ.

KEYWORDS

Radon, Liquid scintillation spectrometer, Packard-PICO-RAD, Misasa hot springs, in air

1. Introduction

The annual exposure dose due to natural radiation is around 2.4 mSv (240 mrem), varying with the location of residence and the environment. The exposure in the environment is predominantly due to the respiration of radon, followed by cosmic and terrestial γ-ray radiation and the intake of natural radionuclides.[1] Recently worldwide attention has focused on exposure due to natural radioactivities,[2] especially radon and its daughters (including thoron).[1),3),4] Detections methods of radon are classified into sampling devices and durations, detectors and analyses, etc. For sampling devices, there are several kinds of filter trapping methods,[5),6] electrostatic trapping ones, active carbon trapping ones,[7] etc.. For detection instruments, there are liquid scintillation counters[8)-12] and nuclear track detectors,[13),14] etc. These radon measurement methods are applied to the investigation of hot spring radiation exposure as well as earthquake forecasting.[15)-17] The behavior of radon and its decay products have also been widely studied. However, many radon researchers in Japan are working almost individually, since the measurement method

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is not yet standardized at present.

We have adopted a simple and highly sensitive radon measurement method, named the PICO-RAD system, using active carbon adsorption vials manufactured by Packard Co. an liquid scintillation spectrometer. We have measured the radon concentration in air and water at the well-known Misasa hot spring area in Tottori prefecture.

2. Methods

2.1 PICO-RAD System

The PICO-RAD radon detector is a vial containing an active carbon adsorber and has the following characteristics.

(1) It is exposed to air for 12~96 hrs.
(2) The treatment of the exposed vial is simple.
(3) Humidity does no influence the calculating procedure.
(4) It is light and of small size.

The liquid scintillation counting procedure should be begun after complete radiation equilibrium. The radon survey of the hot springs in the Misasa area using this system is as follows.

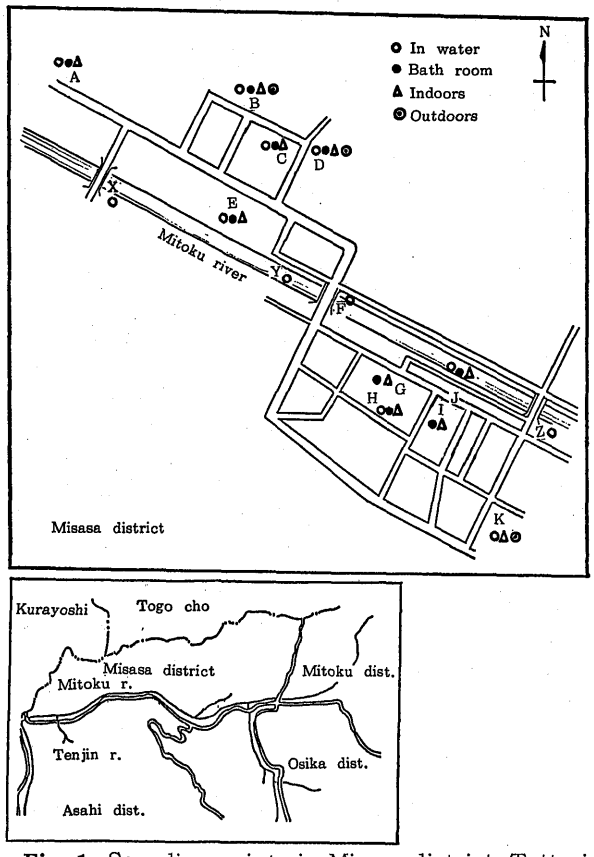

Fig. 1 Sampling points in Misasa district, Tottori

2.2 Sampling

Hot springs gush out at Misasa and Yamada along both banks of the Mitoku stream, the branch of Tenjin river flowing to the north in the middle of the Tottori prefecture. Daily gushing amounts to about 3,000 m^3 and its temperature is 36～85℃. It contains Rn, Na and Cl or Rn, Na, Cl and HCO.[2] We sampled the hot spring water along both banks of the Mitoku stream. **Fig. 1** shows the spots where radon was determined.

2.3 Radon measurement

Radon measurements were done in air (bath rooms, indoors, outdoors). (1) The lid of the PICO-RAD vial was removed for 24 hours for radon adsoption, then 10 mℓ of scintillation solution (INSTA-FLUOR) was poured into each vial. These vials were brought to our laboratory and the radon concentration was measured with a liquid scintillation spectrometer (TRI-CARB 2250 CA Type). (2) Ten milliliters of sampled water was poured into a 20 mℓ glass vial containing 10 mℓ of scintillator solution. After shaking for 30 seconds, the radon concentration was measured with a liquid scintillation spectrometer.

3. Results and Discussion

3.1 Elution and disintegration of radon

The variations in radon concentration in air and in the hot springs are shown in **Fig. 2.** The counting rate of radon in air increased to the maximum value 20 hours after scintillator pouring and decreased with the same half-life as Rn, while the counting rate of radon in water reached a maximum 5 hours after scintillator pouring and decreased with a slightly longer half-life. This difference may be due to the mixing of some radioactive nuclides other than radon.

Fig. 3. shows the variation in radon concentration immediately after pouring scintillator into the vial. The radon concentration maintained a constant value form 20 hours to 180 hours (8 days). From these results, it is concluded that the measurement should

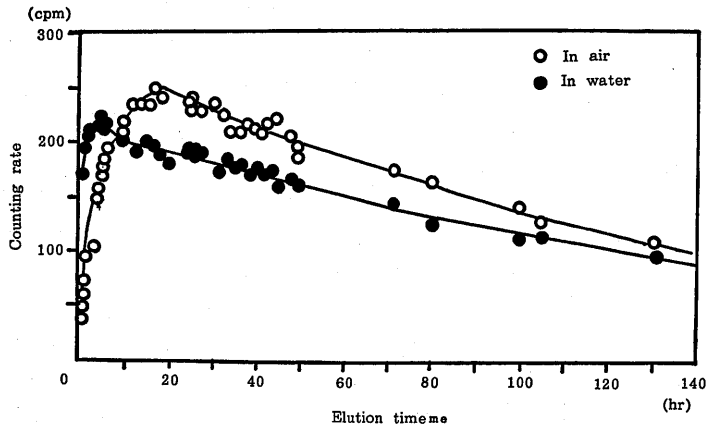

Fig. 2 The Variation of counting rate after liquid scintillator is poured into a vial of the PICO-RAD system

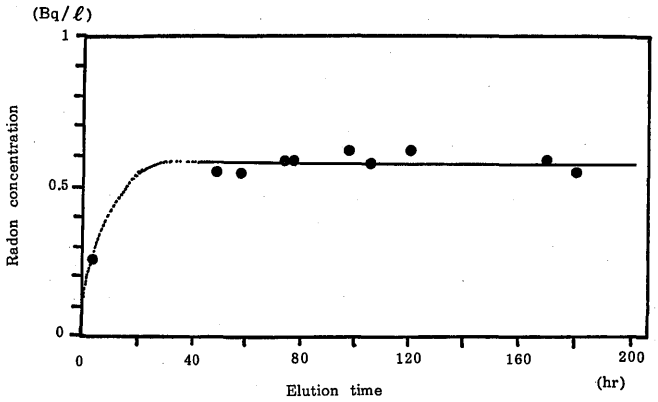

Fig. 3 The variation in radon concentration after liquid scintillator is poured into a vial of the PICO-RAD system

be made 20 hours after scintillator pouring and not later than 8 days.

3.2 Characteristics of radon measurement using the PICO-RAD system

The measurement of radon concentration is made after radiation equilibrium. Accumulated counts due to α-radioactivity of Rn, ^{218}Po (RaA) and ^{214}Po (RaC'), and β-radioactivity of ^{214}Pb (RaB) and ^{214}Bi (RaC) are determined and the concentration of ^{222}Rn in one liter of drinking water is calculated from the these counts. The detection limit was determined from twice the standard deviation of the counts of the PICO-RAD detector of which the lid was put on immediately after scintillator (INSTA-FLUOR) addition without exposure to air. Ten minutes of counting gave 37 mBq/ℓ.

From this value, the detection limit of radon in air was determined to be 3 mBq/ℓ. The relative standard deviation (accuracy) was estimated to be about 10%, which was derived from the counted value of the same sample replicated several times.

This method has the following shortcomings.

(1) Since the sample water is poured directly into the PICO-RAD counting vial, other α-emitting nuclides in the uranium series could be also counted.

(2) Since solvent extraction is used, radionuclides in the thorium series having the same chemical forms could be counted together.

(3) Since the half-life of Rn is short (3.8 days), the counting period after sampling is limited.

3.3 Comparison of measurements of radon in water

Table 1 shows the comparison between the PICO-RAD system by the Packard Co. and the toluene method as well as OPTI-FLUOR method which we formerly used. Since the procedure of sampling and treatment were about the same, the usefulness of the PICO-RAD system was clearly examined.

The toluene method[8),10)] is as follows: since toluene easily dissolves Rn and is a good

Table 1 The comparison among measurements of radon concentration in water

Measurement	Sample Volume (mℓ)	Liquid Scintillator (mℓ)	Detection limit (Bq/ℓ)	Accuracy (%)
Toluene method	500	25	0.090	10.0
OPTI-FLUOR method	10	10	1.530	10.0
PICO-RAD system	10	10	0.549	11.4

scintillator, of 25 mℓ toluene is added to a 500 mℓ sample of water in a 1 liter Erlenmeyer flask. After 5 minutes of strong shaking, it remains still for 10 minutes. The formed toluene layer is transferred to a scintillation vial for counting. Background counts, radioactivity decaying, extraction condition (temperature etc.) and the amount of sample water, etc.[8] are compensated for.

The OPTI-FLUOR method is as follows: the OPTI-FLUOR solution is added to the sample water in the flask. After 30 seconds of strong shaking, radon is extracted. The other procedures are the same as those of the toluene method.

The detection limit of radon concentration in water determined by the PICO-RAD system is 0.5 Bq/ℓ and seems to be sufficient for radon measurement in water, though it is a little higher than that by the toluene method. The detection limit of the OPTI-FLUOR method is higher than the other two methods. However, it has some advantages, i. e. the assay procedure is simple and easy, and glass vials can be used. The accuracies for the three methods were all nearly 10%, being defined as the be relative standard deviations derived from 10 countings of the same sample. **Fig. 4** shows the comparison of radon concentration in water in the Misasa area using toluene method, OPTI-FLUOR method and PICO-RAD system. The radon concentration in the hot springs, which we measured by the PICO-RAD system this time, was in the range of 82～1,700 Bq/ℓ. This is nearly the same level as those which we measured by other two methods in 1989.

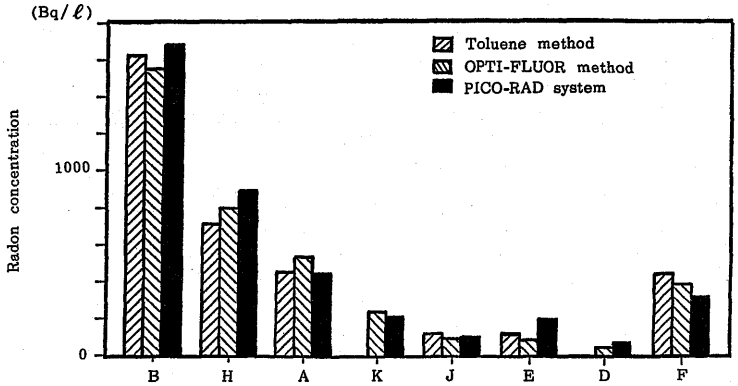

Fig. 4 The comparison of radon concentration measurements. B, H, A..... show the sampling points.

3.4 Radon concentration in water and air in the Misasa area

Radon concentration in water and air inthe Misasa area is summarized in **Fig. 5**. Radon concentration in the hot springs was in the range of 82~1,700 Bq/ℓ, being nearly

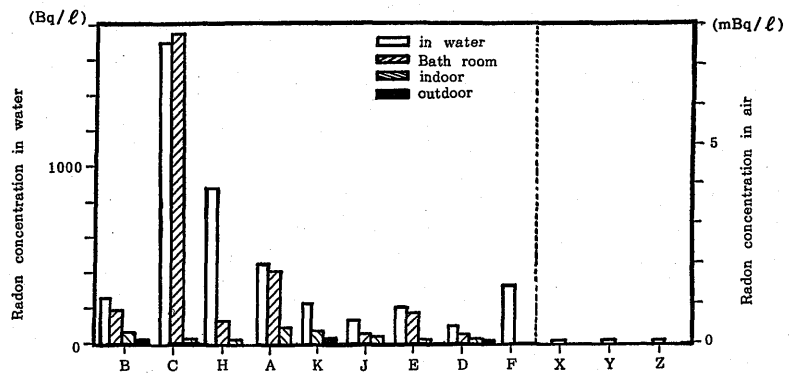

Fig. 5 Radon concentration in the Misasa.
B, C, H, ... show the sampling points.

the same range as the reference.[2] Radon concentration in the river was in the range 13~18 Bq/ℓ. Radon concentration in air was in the range of 0.16~7.7 Bq/ℓ in the bath rooms, 0.057~0.36 Bq/ℓ indoors, 0.05~0.12 Bq/ℓ outdoors. These seem to be somewhat higher in comparison to the reference,[2] i.e. 0.019~0.13 Bq/ℓ indoors and 0.007~0.063 Bq/ℓ outdoors. In the hotels where the hot water is supplied from the outside, the radon concentration in the water is high. In the hotels where the radon concentration in hot spring is high, the radon concentration in air of the bath rooms is also high. However, at point C in **Fig. 1**, the radon concentration in air of bath rooms is low, though radon concentration in hot spring is high. This seems to be due to the good ventiliation and high ceiling. But the features of hotels at points B and C are different, though they both have fountain heads. The radon concentration varies with the environment such as place, time, height from the surface of the ground, and also with the arrangement of the hot spring in the hotel which includes ventilation of the bath rooms and dilution of the hot spring with tap water.

3.5 Radon concentration in the control area

Indoor and outdoor radon concentrations in the areas around Higashi-Osaka city (Osaka prefecture, Wakayama prefecture and Hyogo prefecture) were measured. The outdoor radon concentration was near the detection limit, while the indoor radon concentration was generally higher.

These values are consistent with the values reported by UNSCEAR in 1988,[3] i. e., 3 mBq/ℓ in air and 1 Bq/ℓ in water. Our radon data in concrete houses seem to be higher than those in wooden houses, though the number of samples was seven and the radon concentration did show some variation. Our recent data derived using the PICO-RAD system showed nearly the same level as those of the above reports or references. However,

we have not made a comparison with a standard sample yet. Hereafter, standardization should be indispensable.

4. Summary

Radon concentration in air and water at the Misasa hot spring area in Tottori prefecture was investigated using the PICO-RAD in the system made by Packard Co. as follows.

1) Radon concentration in bath rooms at Misasa area was 0.16～7.7 Bq/ℓ, while outdoors it was 0.05～0.12 Bq/ℓ, and indoors it was 0.057～0.36 Bq/ℓ. These values are 5～30 times the values derived in control areas such as Higashi-Osaka city.

2) Radon concentration in hot springs varied in the range of 82～1,700 Bq/ℓ, while that in the Mitoku river water was 13～18 Bq/ℓ.

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