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Estimation of External and Internal Doses Resulting from the Use of Artificial Radon Spa Sources

Masahiro Hosoda[1], Tetsuo Ishikawa[2], Yumi Yasuoka[3], Sarata K. Sahoo[4], Shin-ichi Katoh[5], Masayuki Ogawa[5], Susumu Ogashiwa[5], Kazuki Iwaoka[5] and Shinji Tokonami[6]*

[1]*Hirosaki University Graduate School of Health Sciences, 66-1 Hon-cho, Hirosaki, Aomori, Japan*
[2]*Fukushima Medical University, 1 Hikarigaoka, Fukushima, Japan*
[3]*Kobe Pharmaceutical University, 4-19-1 Motoyamakitamachi, Higashinada-ku, Kobe, Hyogo, Japan*
[4]*National Institute of Radiological Sciences, 4-9-1 Anagawa, Inage-ku, Chiba, Japan*
[5]*Chuoh College of Medical Technology, 3-5-12 Tateishi, Katsushika-ku, Tokyo, Japan*
[6]*Hirosaki University, Institute of Radiation Emergency Medicine, 66-1 Hon-cho, Hirosaki, Aomori, Japan*

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In this study, four types of readily available artificial radon spa sources, sold as ceramic materials, were studied and dose estimations of the ^{222}Rn emitted from them were made. When people use these sources in bathtub water for bathing, the equivalent dose received by the gonads has been estimated to be 0.40-0.63 nSv for each bathing time (30 min). When each source was put into tap water, the ^{222}Rn concentrations each emitted ranged from 0.5 to 0.7 Bq L^{-1}. If a person drinks 500 mL of water with a ^{222}Rn concentration of 18 Bq L^{-1} every day, the person's maximum annual effective dose is estimated to be 12 nSv.

Key words: artificial radon spa sources, external dose, internal dose, effective dose

1. Introduction

Recently, many goods containing a large amount of radium (^{226}Ra in the ^{238}U-series and ^{228}Ra in the ^{232}Th-series) are marketed as expensive health-enhancing products. Since bathing in radon spas, which are one kind of hot springs, is popular among the general public in Japan, many people have become interested in artificial radon spa sources for their home baths. These sources are used by putting them into bathwater. Ordinary bathwater is said to become radon spa water by radon released from the sources. Some of these sources have high concentrations of ^{226}Ra and ^{228}Ra. Radon is generated from the radium-containing goods by radioactive decay. Radon is a radioactive noble gas that is taken into the body by breathing and it is well-known as a hazardous material that can cause lung cancer through internal exposure.

According to the International Commission on Radiological Protection (ICRP) Publication 103[1], radiation safety management for radon should be dealt with separately from other sources. The World Health Organization (WHO) has recommended the reference level of indoor radon concentration as 100-300 Bq m^{-3} in its handbook[2]. On the other hand, since the drinking water is the most important food, thus, radon in drinking water is important as well as indoor radon from the view point of human health[3, 4]. The U. S. Environmental Protection Agency (EPA) has proposed that the maximum

*Shinji Tokonami: Hirosaki University, Institute of Radiation Emergency Medicine, 66-1 Hon-cho, Hirosaki, Aomori, Japan
E-mail: tokonami@hirosaki-u.ac.jp

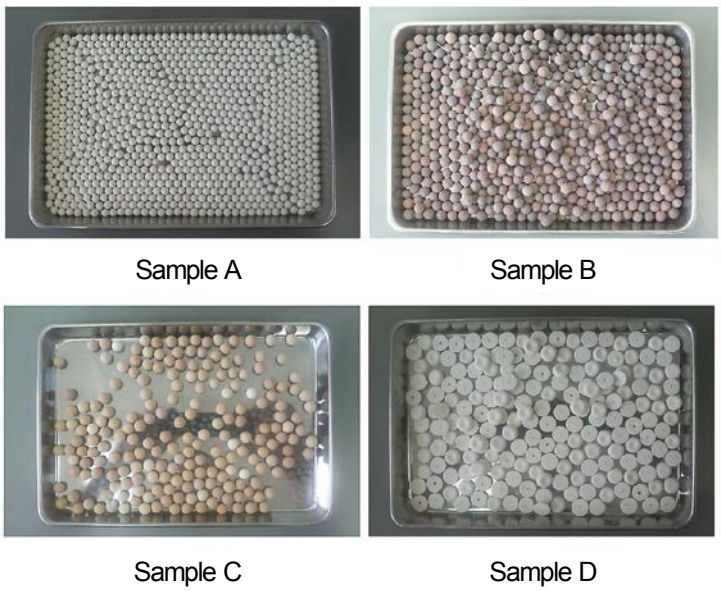

Fig. 1. Different types of materials used for the artificial radon spa source.

contaminant level for ^{222}Rn in drinking water should be 11.1 Bq L^{-1} [5]. WHO[6] and the European commission[7] were recommended that the reference level for ^{222}Rn in drinking water for public water is 100 Bq L^{-1}.

The ICRP Publication 115 has also dealt with lung cancer risk from radon[8]. These facts aside, discussion on the regulation of indoor radon has not progressed in Japan due to the bigger concern about the effects of the Fukushima nuclear accident. On the other hand, it has been found that some products such as cosmetics, personal ornaments and ceramic tile included large amounts of radionuclides, and radiation measurements of these products for the estimation of external dose were reported[9-12]. However, external and internal dose estimations from using artificial radon spa sources have not been published.

Previously, the authors made a radiological characterization of artificial radon spa sources[13]. In the present study, the authors estimated the external and internal doses from four artificial radon spa sources. The external dose was estimated from gamma-rays emitted from the sources when used in the bath. The internal dose was estimated when consuming drinking water containing radon (^{222}Rn) that had been released by the sources. The safety of using these radon spa products at home was discussed based on the obtained dose estimations.

2. Materials and Methods

2.1. Source samples

Figure 1 shows photos of the four source samples which were used for external and internal dose estimation in this study. In the authors' previous study[13], absorbed dose rates in air and α - and β -ray count rates were measured for each sample at a distance of 1 cm from the sample surface using a NaI(Tl) scintillation survey meter (TCS-171, Aloka Co., Ltd, Japan), a GM survey meter (TGS-133, Aloka Co., Ltd, Japan) and a ZnS(Ag) survey meter (TCS-222, Aloka Co., Ltd, Japan), respectively. ^{226}Ra concentrations of four samples were analyzed using a high purity germanium detector. Sample A had the highest values among them. ^{226}Ra concentrations, surface count rates for α - and β -rays, and absorbed dose rates in air of the four sources, Samples A, B, C and D, were 2.9-3.5 kBq kg^{-1} dry, 0.4-1.0 kcpm, 6.8-9.9 kcpm, and 1.7-2.7 μGy h^{-1}, respectively[13].

2.2. Measurement of the concentration in water of ^{222}Rn released from the artificial radon spa sources

Usually, in Japan, the temperature of bathtub water is about 40°C. In this experiment, a thermostat chamber was filled with 15 L of tap water. For the preliminary experiment in which Sample A was measured, the temperature of the thermostat chamber was set to 40°C. However, by the next day the volume of water in the thermostat chamber had decreased drastically due to evaporation. Then, the temperature of the thermostat chamber was lowered to 25°C for comparison with the preliminary result at 40°C. Significant differences in these results were not observed. Thus, the temperature was adjusted from 25 to 30°C. Since the thermostat chamber represented a home bathtub, it was not covered in this study. Although there would be some decrease in the water-amount by evaporation, it was ignored. A

specific weighed amount of each sample was put into a bag having gas permeability and then the bags were separately placed in the water-filled chamber to allow radon to be released. The weighed amounts were: Sample A, 500 g; Sample B, 781 g; Sample C, 572 g; and Sample D, 245 g. Every day at the same time, 100 mL of tap water was withdrawn from the thermostat chamber using an injection syringe. Then, the water sample in the injection syringe was slowly put into a bottle (AquaKIT, Genitron Instruments, Germany), to prevent the generation of bubbles, and the bottle containing the water sample was put into the ^{222}Rn concentration measurement system. ^{222}Rn in the water sample was ejected to the gas phase in this system by a bubbling method using an accessory pump (AlphaPUMP, Genitron Instruments). ^{222}Rn concentration in the gas phase was measured for more than 30 min at one minute intervals using a pulse type ionization chamber (AlphaGUARD, Genitron Instruments)[14]. Measurement of background ^{222}Rn concentration was carried out before the water samples were injected by the same procedure as stated above. The detection limit by this method was estimated as 0.8 Bq L^{-1}.

3. Results and Discussion

3.1. Estimation of the external dose from using artificial radon spa sources

Absorbed doses (Gy) to the gonads (D_t) by using each artificial radon spa source were calculated by the following equation (1).

$$D_t = \dot{D}_a \cdot T \cdot e^{\mu_a (L_1+L_3) - \mu_w L_2} \cdot \left(\frac{L_1+L_3}{L_2} \right)^2 \cdot \frac{(\mu_{en}/\rho)_t}{(\mu_{en}/\rho)_a} \qquad (1)$$

Here, $\dot{D}_a$ is absorbed dose rate in air (Gy h^{-1}) from the source sample surface, T is bathing time (30 min), L_1 is distance between the sample surface and detector surface (0.01 m), L_2 is distance between the sample surface and gonads (0.50 m), L_3 is distance between the probe surface of the survey meter and the effective center of detector (0.02 m)[15], μ is the linear attenuation coefficient (m^{-1}, a: air, w: water). The value of $(e^{\mu_a (L_1+L_3) - \mu_w L_2})$ depends on the gamma-ray energies[16]. According to the previous report by authors[13], the ranges of concentrations of ^{238}U (^{226}Ra), ^{232}Th (^{228}Ra), and ^{40}K in the samples were 2.8-3.5, 15-28 and 1.2-3.3 kBq kg^{-1}, respectively. No dose rate conversion factors for the ^{238}U and ^{232}Th series and for ^{40}K for artificial materials have been published. Such factors have been reported by UNSCEAR2008[17] for soil, and in this case the value for ^{232}Th is higher than those for ^{238}U and ^{40}K. Thus, to ensure a conservative external dose estimation, a maximum value for $(e^{\mu_a (L_1+L_3) - \mu_w L_2})$ of 0.12 at gamma-ray energies of 2.61 MeV for ^{208}Tl was used. The ratio of the mass energy absorbed coefficient of tissue and air $((\mu_{en}/\rho)_t/(\mu_{en}/\rho)_a)$ is almost constant (1.0-1.1) at gamma-ray energies up to 3.0 MeV[16], and it was set as 1.1. When a person uses these radon spa sources in the bathtub water for a 30 min bathing period, the absorbed dose received by the gonads is estimated to be 0.40-0.63 nGy for each bathing time. On the other hand, when the radiation weighting factor of gamma-ray is set as 1.0 according to the ICRP Publication 103[1], the equivalent dose to the gonads is estimated to be 0.40-0.63 nSv for each bathing time. Thus, this result suggests that the health effects to the gonads from the examined artificial radon spa sources are negligible.

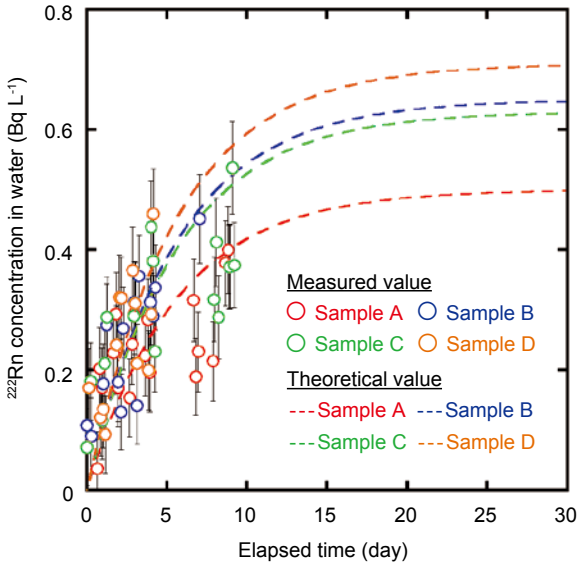

Fig. 2. Time-dependent change of ^{222}Rn concentration in water emitted from each sample.

3.2. Concentration in water of ^{222}Rn released from the artificial radon spa sources

According to the Japanese Hot Springs Law, a spring containing more than 74 Bq L^{-1} of ^{222}Rn is regarded as a radon hot spring[18]. The time variation of ^{222}Rn concentration in water is shown in Figure 2. Although the ^{222}Rn concentrations for every water sample of all four examined sources are below the detection limits, it is clear that the ^{222}Rn concentration in water increases with elapsed time. ^{222}Rn concentrations at the radioactive-equilibrium condition (A_e) were calculated for every water sample using equation (2).

$$A_t = A_e (1 - e^{-\lambda t}) \qquad (2)$$

Here, A_t is the concentration of ^{222}Rn measured at time t (Bq m^{-3}), λ is the decay constant of ^{222}Rn, and t is the accumulation time for sampling (s). ^{222}Rn concentrations at the radioactive-equilibrium condition in this experiment were estimated to be from 0.5 to 0.7 Bq L^{-1} (Figure 2). This result suggests that the ^{222}Rn concentrations in water

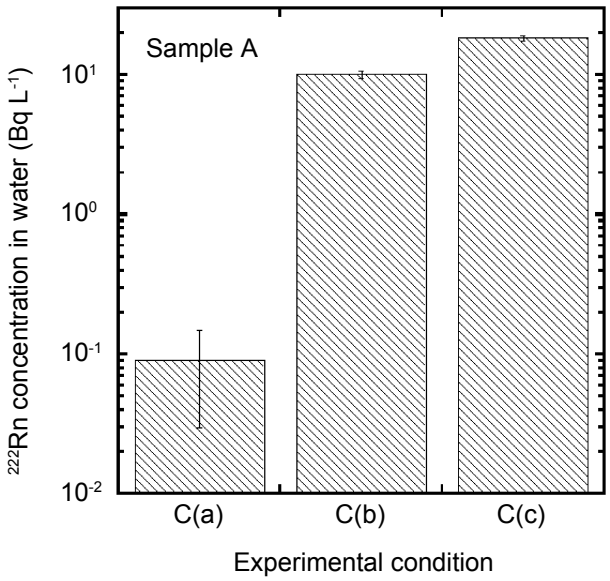

Fig. 3. The ^{222}Rn concentration in water for each set of experimental conditions. C(a): Background (without sample). C(b): 500 g of Sample A + tap water added to the half volume of the bottle. C(c): 500 g of Sample A + tap water added to the full volume of the bottle. [Error bars are shown as 1 σ.]

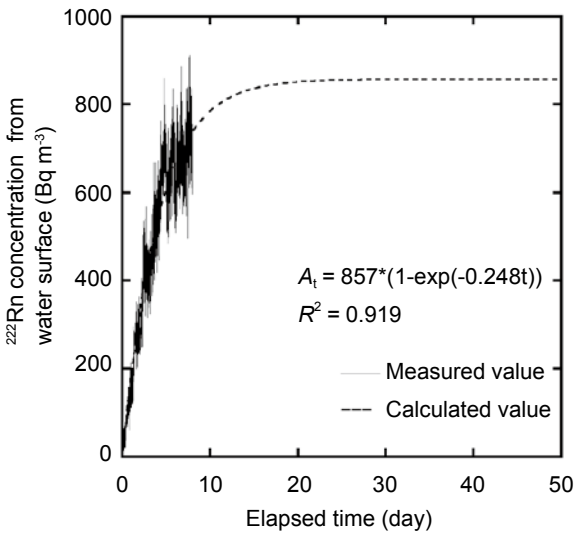

Fig. 4. Time-dependent change of ^{222}Rn concentration released from the water surface.

when using these artificial radon spa sources are much lower than the value for designation as a radon hot spring as stated in the Japanese Hot Springs Law.

Since the present study assumed the thermostat chamber as a bathtub, the chamber was not covered. For that reason, the possible exhalation of ^{222}Rn from the water surface to the air can be considered to occur even though ^{222}Rn is generated in the water. So consideration was given to the amount of exhaled ^{222}Rn from water to the air. Sample A which had the highest ^{226}Ra concentration (3.5 kBq kg^{-1})[13] was used for this. Three sets of experimental conditions were used as follows. (a) A cylindrical bottle (volume 500 mL; open surface area of 4.7×10^{-3} m^2) was completely filled with tap water as a blank sample. (b) 500 g of Sample A was put into a cylindrical bottle (500 mL; 4.7×10^{-3} m^2) and then tap water was added to half-fill the bottle to allow exhalation of ^{222}Rn from the water surface to the gas phase. (c) 500 g of Sample A was put into a cylindrical bottle (500 mL; 4.7×10^{-3} m^2) and then tap water was added to completely fill the bottle; this did not give any exhalation space for ^{222}Rn in the bottle. A cap of the bottle was closed at every experimental condition. All the water was taken from each experimental bottle after 8 days, and put into separate AquaKIT measurement bottles. Then, the ^{222}Rn concentration in the water was measured using the AlphaGUARD. These results are shown in Figure 3. ^{222}Rn concentrations in water at the three sets of conditions were evaluated. Values for C(a), C(b) and C(c) are 0.1 Bq L^{-1}, 10 Bq L^{-1}, and 18 Bq L^{-1}, respectively. Although the ^{222}Rn concentration in water for C(c) is the highest, this value does not exceed the designation value for the radon hot spring. Thus, these findings suggest that the artificial radon spa sources are not able to turn home baths into a radon hot spring.

^{222}Rn concentration released into the gas phase inside the bottle for C(b) was measured at every 10 min using the AlphaGUARD. It is well known that radon exhalation rates from material surfaces increases with sampling flow rates by the *pumping effect*[19]. The mechanism of the pumping effect is as follows: as the sampling flow rate becomes large, the pressure in the bottle becomes increasingly lower than the outside pressure, allowing radon to be more easily exhaled from the water surface into the gas phase inside the bottle, thus increasing radon exhalation rates. Thus, the sampling flow rate of AlphaPUMP was adjusted as 0.1 L min^{-1}. The time variation of ^{222}Rn concentration released into the gas phase inside the bottle is shown in Figure 4. The radon concentrations in water $A_{t,w}$ at time t were calculated using Nernst-Noyes-Whitney equation (Eq. 3)[20].

$$\frac{dA_{t,w}}{dt} = kS\,(A_{s,w} - A_{t,w}) \qquad (3)$$

Here, $\frac{dA_{t,w}}{dt}$ is rate of dissolution, k is a constant, S is surface area of artificial radon spa source, $A_{s,w}$ is ^{222}Rn concentration in water at the saturated state. Moreover, the ^{222}Rn concentrations in air $A_{t,a}$ at time t were calculated by equation (4) which used a transfer factor (TF) from the water phase to the air phase.

$$A_{t,a} = TF \cdot A_{t,w} = TF \cdot A_{s,w}\,(1 - e^{-kSt}) = A_{s,a}(1 - e^{-kSt}) \qquad (4)$$

Here, $A_{s,a}$ is the ^{222}Rn concentration at the saturated state released from the water surface, and this value was evaluated to be 857 Bq m^{-3} using the KaleidaGraph (Synergy software, U.S.A.). Even if radon is generated from these samples in water, it is continuously exhaled from the water surface to the gas phase. Exhalation rate of ^{222}Rn from the water surface for C(b) was calculated by equation (5)[21].

$$E = A_{s,w} \cdot v \quad (5)$$

Here, E is exhalation rate of ^{222}Rn (Bq m^{-2} s^{-1}), $A_{s,w}$ is ^{222}Rn concentration in water (Bq m^{-3}) at the saturated state, and v is diffusion velocity in water (m s^{-1}). The ^{222}Rn concentration in water at the saturated state was estimated to be 7.3×10^4 Bq m^{-3} by equation (4). The diffusion velocity in water reported by Shimo and Ishii[21], which is 1.4×10^{-6} (m s^{-1}), was used for this calculation. Exhalation rate of ^{222}Rn was obtained as 102 mBq m^{-2} s^{-1}, and that was approximately seven times higher than the value (14 mBq m^{-2} s^{-1}) for a static water surface reported by Shimo and Ishii[21]. The surface area of the thermostat chamber as a substitute for a bathtub was 0.203 m^2 and it is much higher than the surface areas (4.7×10^{-3} m^2) of the cylindrical experimental bottles used. Thus, it might be that a higher exhaled ^{222}Rn concentration is observed from the water surface of the thermostat chamber.

3.3. ^{222}Rn emanation coefficient of the artificial radon spa sources

^{222}Rn concentrations in bath water showed similar values to that of tap water even though ^{226}Ra concentrations in these sources were approximately 100 times higher than the values in natural soils[22]. One of the main factors influencing radon generation from the artificial radon spa sources is the ^{222}Rn emanation coefficient. ^{222}Rn emanation coefficients of these samples were reported to range from 0.001 to 0.01[13]. ^{222}Rn emanation coefficients of weathered granite soil samples and building materials which were measured using the same method were previously reported by the authors as: soil samples, 0.3-0.4[23] and building materials 0.002-0.2[24]. Thus, ^{222}Rn emanation coefficients of artificial radon spa sources are about one-tenth those of the natural samples. A few studies have been reported on the ^{222}Rn emanation coefficient of samples sintered at high temperatures[25, 26]. According to these reports, the pore volume of the sintered sample becomes small and that means the emanation space for ^{222}Rn is small. Thus, the radon emanation coefficient will become low according to the increase of sample sintering temperature. An electron microscope image is shown in Figure 5. The pore space of Sample A, a ceramic material, is small. Accordingly, it is suggested that the ^{222}Rn exhalation to the water was suppressed.

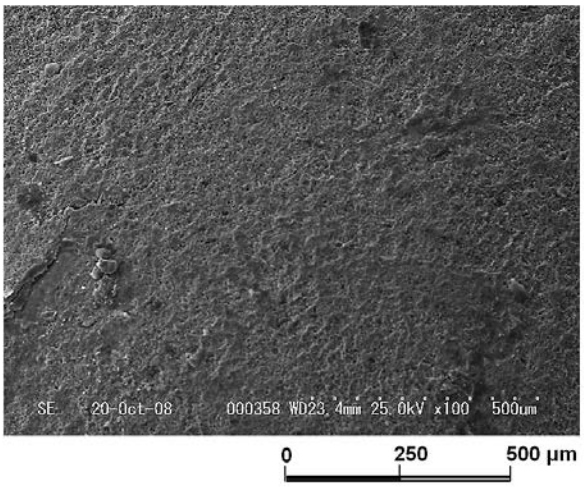

Fig. 5. Electron microscope image of the Sample A surface.

3.4. Internal dose estimation by inhalation of ^{222}Rn from bathtub water

^{222}Rn concentration exhaled from the water surface to the air at the saturated state was estimated as 857 Bq m^{-3} as shown in Figure 4. The transfer factor of ^{222}Rn from water to the atmosphere was estimated to be 10^{-2} by equation (4) in this study, and that was 100 times higher than the value (10^{-4}) reported by UNSCEAR[27]. In order to mainly breathe in exhaled ^{222}Rn near the water surface during the bathing time, it seems that the dose by inhalation must be higher than the dose outside the bathtub. The internal dose from inhalation of exhaled ^{222}Rn from the water surface was calculated by equation (5)[28].

$$D = K \cdot T \cdot F \cdot Q \quad (5)$$

Here, D is effective dose (mSv) during the bathing time due to inhaled ^{222}Rn which is exhaled from bath water to the air, K is dose conversion factor (9×10^{-6} mSv (Bq h m^{-3})$^{-1}$)[16], T is bathing time (h) and it was assumed as 30 min, and F is equilibrium factor. According to the UNSCEAR report, the representative value of F is 0.4[28]. However, the equilibrium factor of 0.33 which was measured at a radon hot spring in Gifu Prefecture, Japan was used for the estimation[29]. Q is ^{222}Rn concentration (Bq m^{-3}) in the atmosphere. In this study, this value was set as 857 Bq m^{-3} which was shown in Figure 4 as the ^{222}Rn concentration after 30 days. If the assumed bathing time is 30 min, the estimated effective dose is 1.3 µSv for each bathing time.

According to the manufacturers' manuals for these sources, it is necessary to put them into the bath water a few hours before bathing. From the present evaluation, the concentration of ^{222}Rn released from the water surface after a few hours is about 20 Bq m^{-3} as shown in Figure4, and the effective dose for this ^{222}Rn concentration is

obtained as 0.03 μSv for each bathing time. Although a straight comparison is not possible since the experimental conditions differ from actual hot spring conditions, the effective dose in the radon hot springs in Gifu Prefecture which has a ^{222}Rn concentration in water of 260 Bq L^{-1} was estimated by Shimo *et al.*[30] as 0.046 μSv. The transfer factor from water to air was considered as 10^{-4} in their estimation. If the transfer factor was the same as the value (10^{-2}) in this study, the estimated effective dose might be higher.

3.5. Internal dose estimation of ^{222}Rn by intake of water sample

The internal dose from daily ^{222}Rn intake was calculated by equation (6)[31] for the case that a person drinks 500 mL of water with a ^{222}Rn concentration Q_W of 18 Bq L^{-1}.

$$D_W = K_W \cdot W \cdot Q_W \quad (6)$$

Here, K_W is dose conversion factor (3.5×10^{-6} mSv Bq^{-1}) and W is water intake per year (182.5 L y^{-1}). The annual effective dose to intake of water is estimated to be 12 μSv.

4. Conclusions

In this study, the external and internal doses from four commercially available artificial radon spa sources were estimated, and the safety of using these products at home was assessed on the basis of these dose estimations. Equivalent dose to the gonads was estimated to be 0.40-0.63 nSv for each bathing time. Moreover, the annual effective dose was estimated to be 12 μSv, even if a person drank 500 mL of water with a ^{222}Rn concentration of 18 Bq L^{-1} every day. These results suggest that the health effects to the users from the assessed artificial radon spa sources are negligible. Furthermore, although the measured ^{226}Ra concentrations in four samples were high as previously reported[13], the ^{222}Rn concentrations in bathtub water did not exceed the designated value for the radon hot spring. Thus, this result suggests that none of the artificial radon spa sources assessed would be effective in turning a home bath into a radon hot spring. However, as we reported previously[13], it is noted that the annual dose could be significant if people stay very close to these samples every day. Recently, many consumer products which include naturally occurring radioactive materials (NORM) are available to buy on the internet. Thus, the measurements of these samples might be needed for dose estimation to the users.

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Disclosure

All the authors report no conflicts of interest.

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