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Note

Experimental Demonstration for Measurement of Radon in Water by a Liquid Scintillation Counter Using Large Vials

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Exposure to radon occurs not only by inhaling radon but also by drinking water. When using the direct method, the radon concentration in water is measured with a conventional liquid scintillation counter (LSC), which can measure samples contained in vials up to 20 ml. The purpose of our research is to develop effective measures for lowering the minimum detectable radon concentration. For the present study, the radon concentration in water was determined using a high efficiency LSC, which can measure larger than usual samples and hence detect lower minimum radon concentrations. In this case, it is a problem that only a few institutes are equipped with the high efficiency LSC. Moreover, the samples are not designated as articles prohibited for mailing by the Japanese Postal Law if they have been prepared. But, by using a mineral oil scintillator with an ignition point higher than 30℃, the samples can be transported to measuring institute without being designated as articles prohibited for mailing by the Japanese Postal Law. We examined the conditions for that. The results suggest that radon leakage from samples contained in Teflon LSC 100 ml vials is negligible for 6 days after sampling. When samples can be mailed to an institute equipped with a counting system capable of measuring the 100 ml vials, results of the samples can be obtained at lower minimum detectable radon concentrations.

Key words: radon concentration in water, high efficiency, liquid scintillation counter (LSC), minimum detectable radon concentration, 100 ml vial, radon leak

1. Introduction

Radon (^{222}Rn) in air and water is the main source of natural radiation received by the general public[1]. The U. S. Environmental Protection Agency (EPA)[2] has proposed that the maximum contaminant level for radon in drinking water should be 11.1 Bq l^{-1}.

The Pharmaceutical Society of Japan published the Methods of Analysis in Health Science[3] which established the method for measuring radon concentration in drinking water with a liquid scintillation counter (LSC) using an extraction method, based on earlier studies[4-6].

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However, more sophisticated technology is required to carry out the extraction method, and prevent radon from leaking from the water sample into the air. The EPA introduced the direct method using just the alpha ray range of the LSC[2, 7]. Tanaka *et al.*[8] demonstrated the direct method using a wider energy range of the LSC (hereafter called direct method). When the conventional LSC was used, however, the minimum detection limits of the extraction method were lower than the direct method[8].

Liquid scintillation analysis of low level radon in water by alpha-beta discrimination of the LSC has been studied order to lower the minimum detection limit. According to the "Handbook of Radioactivity Analysis[9]", calibrations with appropriate quench corrections are necessary, although they do not provide accurate results for variably quenched samples with variable radionuclide compositions.

The use of the high efficiency LSC (e.g. low-background type of LB-LSC series manufactured by Hitachi-Aloka Medical, Japan) has a counting system capable of measuring samples in large sample vials (100 ml vials), as well as in the conventional sized 20 ml vials. When applying the direct method, measurements by the high efficiency LSC using larger than usual samples in the 100 ml vials are effective for reducing the minimum detection limits[8]. To ensure an effective reduction in the minimum detection limits, this method is both simple and practical.

The samples are not designated as an article prohibited for mailing by Japanese Postal Law when they are prepared using a mineral oil scintillator (high-efficiency mineral oil scintillator, cat. no.6NE9579, PerkinElmer Life and Analytical Sciences, U.S.A.) for the direct method since the ignition point of the mineral oil scintillator is higher than 30°C. In this paper, the 100 ml vials were checked as to whether or not they would leak radon if they were sent through the mail. If samples can be mailed, the counting system capable of measuring 100 ml vials at one place can be widely used by researchers at other institutions.

In this paper, radon concentration in groundwater (test sample) was measured using the direct method reported by Tanaka *et al.*[8]. The "standard radon concentration" was determined by the conventional LSC, using the conversion factor for ^{226}Ra standard[10]. Samples contained in 100 ml vials with the same known radon concentration levels were sent to an institute where the high efficiency LSC was available for use. The samples were measured with the high efficiency LSC and the results were examined to evaluate whether the radon concentration level was kept in the samples.

2. Method

Groundwater taken from a well in Kobe City (34° 44' N and 135° 17' E) was used for the tests. The depth of the well, groundwater temperature and pH were 80 m, 19 ºC and pH 6.8, respectively. The radon concentration of this ground water is around 230 Bq l^{-1}, and was a value over the alternative maximum contaminant level (148 Bq l^{-1}) proposed by the EPA[2]; therefore this level was high enough to check for leaks of radon.

The test samples were prepared at Kobe Pharmaceutical University (KPU) as described next. 50 ml of the mineral oil scintillator was added to a Teflon LSC 100 ml vial (Hitachi-Aloka Medical) and then 50 ml of the groundwater was gently added to the bottom of the LSC vial using a 50 ml syringe. The oil-in-water emulsion[11] was prevented by the addition of 0.5 g of NaCl to the mineral oil scintillator before the groundwater was added. To prepare a sample for measuring the background, 50 ml of distilled water was added to the LSC vial instead of the groundwater. A cap was then tightened onto the vial. The vial was then shaken for 30 s. Hereafter, this test sample was called T_{100}. The number of T_{100} samples was 7.

The set of T_{100} samples was put in a PVDF bag and the bag was sealed. This set was sent from KPU to Mie Prefecture Health and Environment Research Institute (MPHERI), where a high efficiency LSC (Model LB-5; Hitachi-Aloka Medical) was available. The T_{100} samples were measured with the LB-5.

The standard samples (S_{20}) were prepared using 10 ml of the mineral oil scintillator, which was added to a glass LSC 20 ml vial (PerkinElmer Life and Analytical Sciences). Then 10 ml of the groundwater was gently added to the bottom of the LSC vial using a 10 ml syringe. 0.1 g of NaCl was added to the mineral oil scintillator before the groundwater was added to prevent the oil-in water emulsion. The samples were shaken for 30 s. The number of S_{20} samples was 6.

The sample set of S_{20} was put in the dark and allowed to stand for at least 4 h before measurement. The S_{20} samples were measured first with the 2300TR (PerkinElmer Life and Analytical Sciences) and this value was called "the standard radon concentration". The S_{20} samples were sent from KPU to MPHERI where they were measured with the LB-5. The S_{20} samples were then sent back to KPU and they were measured again with the 2300TR until about 10 days after their preparation.

When measuring the samples with the LSC, the integral counts (0-2000 keV) were estimated using three wide energy ranges (for example 50-2000, 75-2000 and 100-2000 keV)[8, 9]. The integral counting rate of 0-2000 keV (N_0 (cps)) was close to the disintegration rate (dps)[10, 12-13]. Radon concentration in water C_R (Bq l^{-1}) can be calculated using Equation (1):

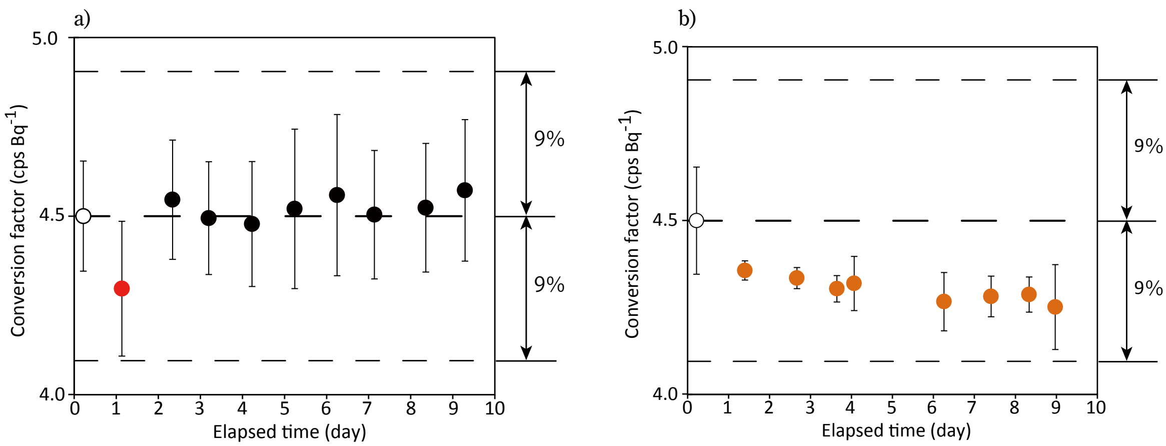

Fig. 1. Conversion factor variation compared with the standard value.
a) Measuring S_{20} sample set with 2300TR and LB-5
b) Measuring T_{100} sample set with LB-5
○ Measuring the standard radon concentration with 2300TR (f = 4.5 cps Bq^{-1})
● (red) Measuring S_{20} with LB-5
● (black) Measuring S_{20} with 2300TR
● (orange) Measuring T_{100} with LB-5

$$C_R = (N_{S0} - N_{B0}) \times \exp\left(\frac{0.693 \times t_E}{T}\right) \times \frac{1}{f} \times \frac{1}{V} \qquad (1)$$

where N_{S0} (cps) is the integral counting rate of the sample, N_{B0} (cps) is the integral counting rate of the background sample, T (s) is the half-life of radon (3.3 × 10^5 s), t_E (s) is the elapsed time it takes between the commencement of water sampling and the midpoint of the counting time. V (l) is the water volume and f (cps Bq^{-1}) is the conversion factor.

To obtain "the standard radon concentration" (this was the first measurement of the S_{20} samples with the 2300TR), the applied f was 4.5 cps Bq^{-1} (standard value: open circle in Figures 1a and 1b). This conversion factor (4.5 cps Bq^{-1}), which was obtained using the mineral oil scintillator with the 2300TR at KPU, was determined by the comparison measurements using the ^{226}Ra standard sample and radon water samples[8].

3. Results and discussion

Figure 1 shows the results of the variation in f in Equation (1). According to the EPA[14], the proposed acceptance limits for triplicate analyses at the 99% confidence intervals are 9%. Figure 1a shows the variation of f in the S_{20} set, compared with the standard value. When measuring the 20 ml vial sample with the LB-5 (closed red circle in Figure 1a) and 2300TR (closed black circle in Figure 1a), all results were within the acceptable range (± 9%). The conversion factor using S_{20} with the LB-5 was determined as 4.5 cps Bq^{-1}.

Figure 1b shows the variation of f in T_{100} with LB-5 (closed orange circle), by comparing it with the standard value (open circle). All results were within the acceptable range (± 9%). When measuring the 100 ml vial sample with the LB-5, the conversion factor for the use of LB-5 was determined as 4.5 cps Bq^{-1}. Radon leakage from the Teflon LSC 100 ml vials was still negligible for 6 days (1.5 times the half-life of radon) after sampling.

A further important point is the minimum detectable radon concentration (MDC_{radon}) of this method, which was given in Equation (2)[15, 16] by Tanaka *et al.*[8]

$$MDC_{radon} = \frac{\dfrac{2.71}{t_c} + 4.65\sqrt{\dfrac{N_B}{t_c}}}{f \times V} \qquad (2)$$

where N_B (cps) is the counting rate of the background sample, t_c (s) is the counting time, V (l) is the water volume, and f (cps Bq^{-1}) is the conversion factor in Equation (1). The results of the minimum detectable radon concentrations differ depending on the location, as the counting rates of the background are different in each place. When the conventional LSC (t_c = 3600 s) was used, the minimum detectable radon concentrations of the extraction method and the direct method were 0.03-0.04 Bq l^{-1} and 0.5-1 Bq l^{-1}, respectively[8, 10]. The high efficiency LSC (t_c = 3600 s) has a counting system capable of measuring samples in 100 ml vials as well as the conventional 20 ml vials. When using the direct method, the measurement of the high efficiency LSC using 100 ml vials can be effective for lowering the minimum detectable radon concentrations (0.07 Bq l^{-1})[8] .

When the samples can be mailed to an institute equipped with LB-5, the results of radon detection for the samples can be obtained at lower minimum detectable radon concentrations.

4. Conclusion

When determining the amount of exposure due to inhalation and drinking of radon from water, the radon concentration in water is measured using the direct method with a conventional LSC that can measure samples contained in up to 20 ml vials. The purpose of our research is to determine whether effective measures can be developed for lowering the minimum detectable radon concentration. In this study, the direct method was used to determine radon concentration in water samples with the 2300TR (using glass LSC 20 ml vials) and LB-5 (using glass LSC 20 ml vials and Teflon LSC 100 ml vials). The conversion factor of the direct method used was 4.5 cps Bq^{-1}, which was obtained using the mineral oil scintillator. This material is not designated as prohibited for mailing by Japanese Postal Law, because the ignition point of the mineral oil scintillator is higher than 30 ˚C. The results obtained in the present study suggested that radon leakage was still negligible for 6 days (1.5 times the half-life of radon) after sampling when samples were kept in the 100 ml vials. It was judged possible to mail samples to an institute equipped with a counting system capable of measuring 100 ml vials and, thus, make the system available to researchers at other institutes. The method is an effective way to lower the minimum detectable radon concentrations (0.07 Bq l^{-1}).

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