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原　　著

Comparison of Simplified Liquid Scintillation Counter (Triathler) with Conventional Liquid Scintillation Counter in the Measurement of Radon Concentration in Water

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水中ラドン濃度測定における通常型液体シンチレーションカウンタと簡易型測定器（Triathler）との比較

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要　　旨

水中ラドン濃度測定について，現場での測定が可能なトライアスラー（簡易液体シンチレーションカウンタ）と，汎用の液体シンチレーションカウンタとを比較した．トライアスラーにおいても，鉱泉分析法指針に示された方法より簡便な測定法の直接測定法で，測定が可能であることがわかった．3チャンネル（50-1,016 ch, 75-1,016 ch, 100-1,016 ch）で測定し，最小二乗法により0-1,016 chの値を求めた．計数効率は470%であった．汎用の液体シンチレーションカウンタの測定値との誤差は5%以下で，定量限界は約7 Bq/kg（60分測定）であった．トライアスラーを用いた直接法による水中ラドン濃度の測定法は，鉱泉分析法指針に示された方法の代替として有効である．

キーワード：ラドン，水，液体シンチレーションカウンタ，トライアスラー，校正，鉱泉分析法指針

Abstract

We showed that Triathler can be used for measuring radon concentration of around 74 Bq/kg in water as a direct method. Triathler is cheaper than a conventional liquid

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scintillation counter (LSC), and can be brought to a sampling site. The direct method is easier than the extraction method required by the Guidelines of Analytical Method of Mineral Springs of Japan (GAMM). The optimized window values of 3 efficiency tracing points were determined at 50–1,016 ch, 75–1,016 ch and 100–1,016 ch. The counting rate of 0–1,016 ch was calculated by the three counting rates in the optimized window values using the least-square method. The counting efficiency was 470%. We obtained a set of 5 samples (radon concentration : over 60 Bq/kg) for each sampling point and applied a measuring time of 60 min. Under these conditions, the difference between Triathler and LSC was lower than 5%, and the quantification limit of Triathler was approximately 7 Bq/kg. This method is available as an alternative of the method required by the GAMM.

Key words : Radon, Water, Liquid scintillation, Triathler, Calibration, Guidelines of Analytical Method of Mineral Springs of Japan

1. Introduction

The liquid scintillation method is commonly used for measuring radon (^{222}Rn) concentration in water, and is recommended in the Guidelines of Analytical Method of Mineral Springs of Japan (GAMM) (Ministry of the Environment of Japan, 2002). According to the GAMM, a spring with radon concentration of more than 74 Bq/kg is regarded as a mineral spring. There are many varieties of mineral springs in Japan. The GAMM requires that radon concentration should be measured by either the IM-fontactoscope method or the liquid scintillation method (Ministry of the Environment of Japan, 2002 ; Horiuchi and Murakami, 1977 ; Horiuchi, 1978). However, the IM-fontactoscope is no longer produced. In addition, we found that the radon-in-water concentrations measured with the IM-fontactoscope method deviated from those measured with other methods (Japan Health Physics Society, 2004 ; Ishikawa *et al.,* 2003, 2004). Recently, the Hot Spring Law (Onsen-ho, Government of Japan, 1984) was revised in October 2007. The revised law requires that the components of mineral springs should be measured every 10 years. The radon-in-water concentrations will have to be measured using the liquid scintillation method.

The GAMM adopts the “extraction method” which involves extracting radon from a water sample to toluene scintillator in a bottle, and transferring the scintillator to a vial, then measuring the vial with a conventional liquid scintillation counter (LSC). The toluene scintillator is described in detail in Section 2.1. In this paper, “E-sample” indicates the sample prepared by the extraction method using a toluene scintillator. The U.S. Environmental Protection Agency (U.S. EPA, 1998 ; American Society for Testing and Materials, 1999 ; American Public Health Association, American Water Works Association and Water Environment Federation, 1998) adopts the “direct method” which involves adding a water sample to a water immiscible scintillator in a vial, then extracting radon from the water to the scintillator in the vial and measuring the vial with LSC. For the measurement of radon concentration in mineral spring must be done and meet the GAMM (the radon concentration 74 Bq/kg), Yasuoka *et al.* (2004) proposed that the “direct method” is preferable than the extraction method. While the counting rates of the direct method are reduced by one thirty as compared with the extraction method, the direct method is simpler than the extraction method. In addition, the direct method makes it easy to prevent radon from escaping from a sample. In this paper,

"D-sample" and "M-sample" indicate the samples for the direct method using toluene scintillator and high efficiency mineral oil scintillator® (PerkinElmer Life and Analytical Sciences, Inc., cat. no. 6NE9571), respectively.

The liquid scintillation method has been performed with LSC, which is used in a laboratory only. Recently, a portable and simplified liquid scintillation counter, Triathler (Hidex Ltd. in Finland) is commercially available (Haaslahti *et al.,* 2000). It is cheaper than LSC, and can be brought to a sampling site. If it can be used for the in-situ measurement of radon concentration in water, it will be very useful for the measurement that should be conducted according to the GAMM. However, it is not clear how radon concentration in water is measured with Triathler without ^{226}Ra standard sources. It is difficult to use ^{226}Ra standard sources due to Japanese government regulations.

For the measurement of radon concentration in water, the direct method using LSC already has been alternative for the extraction method required by the GAMM (Yasuoka *et al.,* 2004). The calibration factor of LSC for the direct method was obtained by using a radium standard (Yasuoka *et al.,* 2005). The purpose of this study is to demonstrate the direct method with Triathler can be used as an alternative of the direct method with LSC. We will describe how Triathler can be used for the measurement of radon concentration in water by the direct method. First, a counting rate of a scintillator including high radon concentration is measured with Triathler using the direct method, and it is compared with that measured with LSC using the direct method to setting desired measuring conditions. Second, the counting rate measured with Triathler using the direct method is compared with that measured with LSC and Triathler for the extraction method. Third, counting rates of scintillator including the various values of radon concentration are measured with Triathler and LSC.

2. Materials and Methods

2.1 Sample preparation

Three kinds of samples (E-sample, D-sample and M-sample) were prepared as follows. The data are from one representative of five samples in each group. Much information about the E-sample (Ministry of the Environment, Japan, 2002 ; Horiuchi and Murakami, 1977 ; Horiuchi, 1978), the D-sample (Japan Health Physics Society, 2004 ; Yasuoka *et al.,* 2004) and M-sample (U.S. EPA, 1998 ; Cook *et al.,* 2003) has been published concerning the proper method for sampling water for radon measurement. Groundwater taken from a well in Kobe Pharmaceutical University was used for the experiments. The depth of well, groundwater temperature and pH were 80 m, 18.5℃ and pH 6.8, respectively.

For the E-sample, several tens mL of toluene scintillator was mixed with about 1 L groundwater in a bottle by shaking the bottle for 2 minutes. Radon was transferred into the toluene scintillator by shaking, i.e., radon was extracted from the groundwater. The scintillator containing radon was transferred to a flask for a uniform radon concentration. Twenty mL of the scintillator containing radon was transferred into each vial. This method is referred to as the extraction method in this paper. Toluene scintillator contains 2,5-diphenyloxazole (4 g/L) and 1,4-bis [2-(5-phenyloxazolyl)] benzene (0.1 g/L) in toluene.

The D-sample and M-sample were prepared as follows : groundwater containing radon was transferred to a flask for a uniform radon concentration. For the D-sample, toluene scintillator (10 mL) was mixed with 10 mL groundwater in a vial by shaking the vial for 30 seconds. For the M-sample, high efficiency mineral oil scintillator® (10 mL) was mixed with 10 mL groundwater in a vial by shaking the vial for 30 seconds (U.S. EPA, 1998 ; American Society for Testing and Materials, 1999 ; American Public Health Association, American Water Works Association and Water Environment Federation, 1998). Both of them are referred to as the direct methods in this paper.

The counting rates of the D-sample and the E-sample, which were measured with Triathler, were compared with that measured with LSC to setting desired measuring conditions. Several tens mL of scintillator (toluene scintillator or high efficiency mineral oil scintillator) was mixed with groundwater (about 1 L) in a bottle by shaking the bottle for 2 minutes. The scintillator containing radon was transferred to a flask. To prepare the D-sample, 10 mL of radon-free water and 10 mL of the scintillator containing radon were mixed in a vial by shaking the vial for 30 seconds. To prepare the E-sample, 10 mL of scintillator (radon-free) and 10 mL of the scintillator containing radon were mixed in a vial by shaking the vial for 30 seconds.

2.2 LSC and Triathler

Triathler has only one photomultiplier tube, while LSC has two photomultiplier tubes which help differentiate background signals from true nuclear decay events in the scintillation vial, using coincidence counting. Also, the shield of Triathler is thinner than that of LSC. The high background count rate mainly occurs by thermal and electronic background noise in the 0–10 keV region (L'Annunziata, 2003). An optional software of Triathler enables the separation of alpha/beta counting (Haaslahti *et al.,* 2000 ; Horiuchi *et al.,* 2006). In the present study, however, we used Triathler without alpha/beta separation software.

2.3 Measurement of radon concentration with LSC and Triathler

After waiting for more than 4 hours, the radon concentrations of samples were measured. Subscripts of L and T show that the samples were measured with LSC (PerkinElmer Life and Analytical Sciences, Inc., Tri-Carb 2300TR) and Triathler (Type 425–034), respectively. For example, the D_L means that D-sample was measured with LSC. The measuring time was 10 minutes (unless otherwise stated), except for the D_T. The measuring time for the D_T was 60 minutes. Five sets of samples were prepared for each measurement method. The mean and standard deviation were calculated from the measurements of five samples.

A background sample for the E-samples was 20 mL of the toluene scintillator. A background sample for the D-samples was 10 mL of the toluene scintillator and 10 mL of radon-free water. A background sample for the M-samples was 10 mL of high efficiency mineral oil scintillator and 10 mL of radon-free water. All background samples were measured for 60 minutes with LSC and Triathler. The standard deviations of the background samples were calculated based on standard error (Eq. 1) (Japan Radioisotope Association, 2001),

$$\sigma=\sqrt{\frac{m}{t}} \quad \cdots\cdots(1)$$

where m is the counting rate of background (cpm), t is the measuring time (min) and σ is the standard deviation of the background.

When the D_L and E_L-samples were measured with LSC, the optimized window values have 3 efficiency tracing points : 50–2,000 keV, 75–2,000 keV and 100–2,000 keV. The counting rate of 0–2,000 keV is calculated by the three counting rates of the optimized window values using the least-square method (Japan Health Physics Society, 2004 ; Yasuoka *et al.,* 2004 ; Noguchi, 1975a, 1975b). In the case of LSC which we used, the counting efficiency was proven to be 470% according to our comparison experiments with a ^{226}Ra standard (Yasuoka *et al.,* 2005), while the counting efficiency is theoretically 500%. In the case of Triathler, the optimized window values of 3 efficiency tracing points are discussed in Section 3.1.

2.4 Radon concentration

Using the toluene scintillator, the radon concentration (Bq/kg) in water is calculated by the Noguchi-Saito formula on the gas-liquid distribution described by Saito (Japan Health Physics Society, 2004 ; Saito and Takata, 1992 ; Saito *et al.,* 1993 ; Saito and Takata, 1994 ; Saito, 1997). The equilibrium distribution between toluene scintillator, water, and air was given by this calculation. Unless otherwise noted, the radon concentration and the counting rates were indicated considering the decay correction for the elapsed days (between the time of shaking the sample and the midpoint of counting period for the sample).

PerkinElmer Life and Analytical Sciences, Inc. (2009, pers. com.) shows radon in water was measured with PerkinElme Tri-Carb LSC in the optimized region from 200 keV to 600 keV, using the M_L-sample. For this sample, 10 mL high efficiency mineral oil scintillator was mixed with 10 mL groundwater in a vial by shaking the vial for 30 seconds. In addition, PerkinElmer Life and Analytical Sciences, Inc. suggested that the radon concentration (Bq/L) in the groundwater would be calculated using Eq. (2) following the standard method 7500-Rn (American Public Health Association, American Water Works Association and Water Environment Federation, 1998).

$$C_R=netcps\times\exp\left(\frac{0.693\times T}{3.824}\right)\times\frac{1}{3.2\times0.01} \quad \cdots\cdots(2)$$

where *netcps* (cps) is the net count per second (i.e., after subtraction of background), T is the length of elapsed days, 3.824 is the half life of radon in days, the groundwater volume is 0.01 L, the calibration constant is 3.2 cps/Bq and C_R is radon concentration (Bq/L) in the groundwater.

2.5 Measuring various radon concentration in water

According to Ministry of the Environment of Japan (2002), the spring water with a radon concentration of more than 74 Bq/kg is regarded as “mineral spring”, one of the varieties of mineral springs. The net counting rates (within a range of 0–2,000 keV) corresponding to a radon concentration of about 74 Bq/kg are about 5,900 cpm (E-sample) and 190 cpm (D-sample). The E-sample and D-sample were allowed to stand for some days and then measured again

the samples of varied radon concentration were measured with LSC and Triathler. For a single sample, the counting rate measured with LSC was compared with the counting rate measured with Triathler. The radon concentration and the counting rates were calculated considering the decay correction for the elapsed days (between the midpoint of counting period for the sample measured with LSC and that measured with Triathler).

Using the 5 levels of radon concentration in water (from 60 Bq/kg to 130 Bq/kg), the radon concentration was measured with the direct method (U.S. EPA, 1998 ; Cook *et al.,* 2003 ; Japan Health Physics Society, 2004 ; Yasuoka *et al.,* 2004) and the extraction method (Ministry of the Environment of Japan, 2002 ; Horiuchi and Murakami, 1977 ; Horiuchi, 1978). D-sample was measured with LSC (D_L) and Triathler (D_T), M_L-sample was measured with LSC, and E-sample was measured with LSC (E_L) and Triathler (E_T).

While Triathler is a portable device and can be brought to a sampling site, the LSC can not be moved from a laboratory. For this reason, the samples sometimes need to be sent to a laboratory by refrigerated (5℃) cargo. In this case, we investigated how long the samples were stored at room temperature before starting the measurements. D_L- and M_L-samples were prepared by the groundwater, and the samples set aside in the dark for 4 hours. At starting time, the counting rates of the D_L- and M_L-samples were about 5,500 cpm and 4,000 cpm, respectively. These samples were refrigerated (5℃) for 18 hours, and were repeatedly measured with LSC for 30 hours.

3. Results and Discussion

3.1 Determining optimized window values of Triathler

The samples were measured with Triathler by setting ^{32}P key. Figure 1 shows the pulse height spectra of radon and its decay products of D_T-sample measured with Triathler. The optimized window values of 3 efficiency tracing points were determined at 50–1,016 ch, 75–1,016 ch and 100–1,016 ch. The 3 efficiency tracing points are available due to exception of the 0–10 keV region, in which the high background count rate mainly occurs. The starting channels (50, 75 and 100 ch) could be determined in the beta area of Fig. 1 (see Appendix I). There were noise counts over 1,016 ch. The counting rate of 0–1,016 keV is calculated by the three counting rates in the optimized window values using the least-square method. Under these conditions the samples were measured with Triathler. Table 1 shows D and E-samples were measured with LSC (D_L and E_L) and Triathler (D_T and E_T). There were no significant differences (the significance prob-

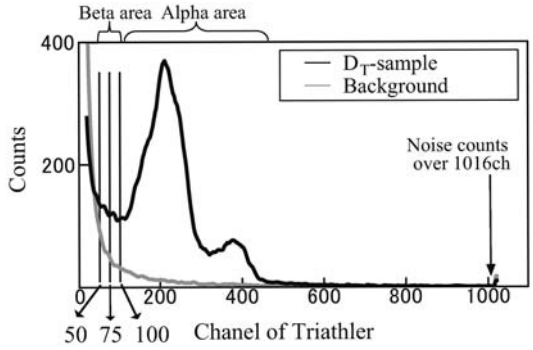

Fig. 1 Pulse height spectrum of radon and its decay products measured D_T-sample and background sample with Triathler. The optimized window values of 3 efficiency tracing points were determined 50–1,016 ch, 75–1,016 ch and 100–1,016 ch.
D_T : The direct method performed with Triathler using toluene scintillator.

ability (P values) ranged from 0.6 to 0.9) between E_L and others (E_T, D_L, and D_T). If the net counting rate of 0–2,000 keV (LSC) is over 7,000 cpm, the results between the D_L-sample and the others (D_T, E_L and E_T-samples) were same. In the case of Triathler, the counting efficiency was estimated to be 470%.

3.2 Measuring various radon concentration in water

For a single sample, the counting rate measured with LSC was compared with the counting rate measured with Triathler. Figure 2a shows the relationship between the E_L net counting rates of 0–2,000 keV and the E_T net counting rates of 0–1,016 ch when the E-sample was measured. Figure 2b shows the relationship between the D_L net counting rates of 0–2,000 keV and the D_T net counting rates when the D-sample was measured. If a single sample is measured with some LSCs, the acceptance limits of differences are±5% (U.S. EPA, 1998). D_L (or E_L) was defined as a standard. Hereafter the difference (*Difference %*) between D_L (or E_L) and D_T (or M_L or E_T) means the following Eq. (3) :

Table 1　Results of measuring E-sample and D-sample with conventional liquid scintillation counter (LSC) and Triathler

	D_L	D_T	E_L	E_T
Background	18	141	27	258
Average±standard deviation	7,586±443	7,592±462	7,476±218	7,484±175
Significance probability D_L	—	0.87	0.63	0.64
Significance probability E_L	—	—	—	0.89

D_L : The direct method performed with LSC using toluene scintillator.
D_T : The direct method performed with Triathler using toluene scintillator.
E_L : The extraction method performed with LSC using toluene scintillator.
E_T : The e extraction method performed with Triathler using toluene scintillator.

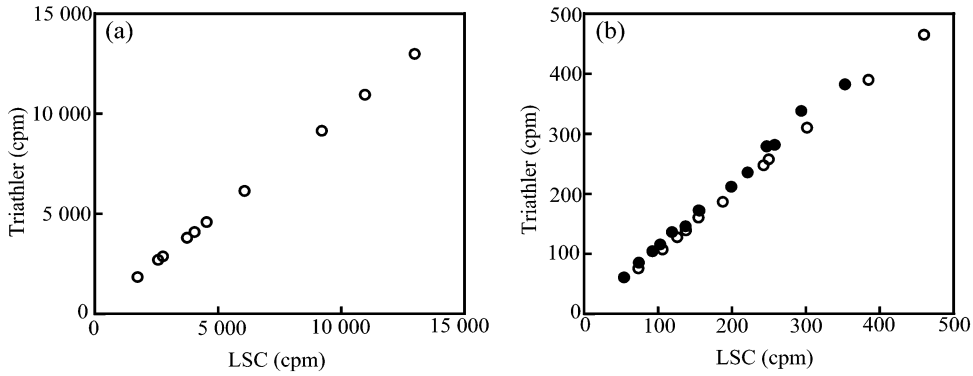

Fig. 2　The integral counting rate of conventional liquid scintillation counter (LSC) versus Triathler.
a) Extraction method
○ : the differences of the counting rate of Triathler were less than 5%.
b) Direction method
○ : the differences of the counting rate of Triathler were less than 5%.
● : the differences of the counting rate of Triathler were more than 5%

$$Difference\ \% = \frac{D_T - D_L}{D_L} \times 100 \quad \cdots\cdots(3)$$

The differences between E_L and E_T were within±5%. However the some differences between D_L and D_T were over±5%. As a result, the method, which E-sample is measured with Triathler instead of LSC, is fully capable of measuring the criteria of mineral spring (more than 74 Bq/kg).

D-sample was measured with LSC and Triathler, and M_L-sample was measured with LSC, using the 5 levels of radon concentration in water (from 60 Bq/kg to 130 Bq/kg). Table 2 shows radon concentrations, the differences in the radon concentration between D_L-samples and others (D_T-sample, M_L-samples, E_L-sample and E_T-sample), and the measuring times. In our experiment, the differences between D_L-sample and others (D_T-sample, E_L-sample and E_T-sample) were within±6% (≤±2.5%, ≤±5.7% and ≤±4.9%, respectively). Additionally,

Table 2　Result of the measurement.

Method	Direct method			Extraction method	
Liquid Scintillator	Toluene scintillator		High efficiency mineral oil	Toluene scintillator	
Equipment	LSC	Triathler	LSC	LSC	Triathler
Background counting rate (cpm)	18	141	5.5	27	258
Radon concentration	D_L (Bq/kg) Standard M.T.	D_T (Bq/kg) Difference % M.T.	M_L (Bq/L) Difference % M.T.	E_L (Bq/kg) Difference % M.T.	E_T (Bq/kg) Difference % M.T.
	58.3±1.7 — 30 min	59.0±1.1 1.7% 60 min	62.8±0.9 7.3% 30 min	60.0±0.6 2.8% 10 min	60.5±0.9 3.7% 10 min
	66.6±1.9 — 30 min	65.5±3.0 −1.7% 60 min	71.7±2.3 7.0% 30 min	70.4±1.2 5.7% 10 min	69.9±1.3 4.9% 10 min
	74.5±1.1 — 30 min	73.7±0.2 −1.1% 60 min	80.1±0.3 7.5% 30 min	76.3±0.7 2.4% 10 min	74.8±0.4 0.3% 10 min
	118.2±3.4 — 10 min	116.8±1.3 −1.2% 60 min	123.6±2.8 5.5% 10 min	119.5±1.9 1.1% 10 min	118.4±2.5 0.2% 10 min
	127.9±2.8 — 10 min	124.7±2.2 −2.5% 60 min	128.6±3.7 0.5% 10 min	125.9±1.5 −1.5% 10 min	123.8±1.2 −3.2% 10 min

D_L : Direct method performed with LSC using toluene scintillator.
D_T : Direct method performed with Triathler using toluene scintillator.
M_L : Direct method performed with LSC using high efficiency mineral oil
M.T. : Measuring time
LSC : Conventional liquid scintillation counter

the differences between D_L-sample and M_L-sample were also within＋7.5%.

The quantification limit of the counting rate (Q cpm) is defined in terms of equation (4),

$$Q = 10\sigma \quad \cdots\cdots(4)$$

where σ is the standard deviation of the background. The background for D_T (147 cpm) was much higher than the background for D_L (18 cpm) and M_L (4.6 cpm). If the measuring times of D_T, D_L, and M_L were 60 min., 10 min., and 10 min., the quantification limits of radon concentrations for D_T, D_L, and M_L-samples were approximately 7 Bq/kg, 5 Bq/kg and 4 Bq/L, respectively.

After the samples were refrigerated (5℃) for more than 6 hours, the radon concentrations of M_L-samples showed the level at starting time, and the radon concentrations of D_L-samples ranged from 95 to 98% of the level at starting time.

4. Conclusions

We showed that Triathler with the extraction method can be used for measuring water with a radon concentration of around 74 Bq/kg, which is the criterion of “mineral spring”. We also showed that Triathler with the direct method can be used for measuring of radon concentration in groundwater. The optimized window values of 3 efficiency tracing points were determined at 50–1,016 ch, 75–1,016 ch and 100–1,016 ch. The counting rate of 0–1,016 keV is calculated by the three counting rates in the optimized window values using the least-square method. The counting efficiency was estimated to be 470%. If we obtain a set of 5 samples (radon concentration : over 60 Bq/kg) for each sampling point and apply a measuring time of 60 min., the differences between Triathler and LSC were lower than 5% (＜2.5% in our experiment), and the quantification limit was approximately 7 Bq/kg. This method is available as an alternative of the GAMM.

Acknowledgments

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Appendix I

Figure A1 shows the pulse height spectra of radon and its decay products for D_T-sample measured with two Triathlers (Type 425–034). There were individual differences in the spectrum. In this paper, we used Triathler with the spectrum I. The optimized window values of 3 efficiency tracing points from both of Triathlers were determined at 50–1,016 ch, 75–1,016 ch and 100–1,016 ch. The counting rate of 0–1,016 keV is calculated by the three counting rates in the optimized window values using the least-square method. The counting rates of E_L-sample (about 13,000 cpm) were higher than the counting rates of E_T-sample (spectrum I : 1.6±0.9%, spectrum II 2.1±0.8%). If a single sample is measured some LSCs, the acceptance limit of error is±5% (U.S. EPA, 1998). The differences between E_T-sample and E_L-sample were within±5%. The counting rates of D_L-sample (about 7,000 cpm) were higher than the counting rates of D_T-sample (spectrum I : 1.6±1.3%, spectrum II 2.8±1.4%). The variation of the pulse height spectrum has the possible effect on the counting rate.

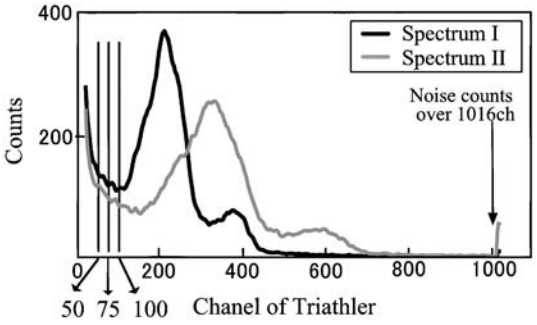

Fig. A1　The pulse height spectra I and II of radon and its decay products for D_T-sample. The spectra were measured with two Triathlers (year purchased : 2005 (spectrum I) and 2007 (spectrum II)), whose models were same (Type 425–034). In this paper, we used Triathler with the spectrum I.