

# DETERMINATION OF ATMOSPHERIC RADIOACTIVITY USING A MEMBRANE FILTER AND LIQUID SCINTILLATION SPECTROMETRY

YOSHIO HOMMA, YUKO MURASE and KEIKO HANDA

Kyoritsu College of Pharmacy, 1-5-30, Shibakoen, Minato-ku, Tokyo 105 Japan

**ABSTRACT.** We describe a simple and reliable procedure to determine  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$  in air using a membrane filter and a liquid scintillation spectrometer. We dissolved a 0.65- $\mu\text{m}$ -pore cellulose nitrate membrane filter, on which the short-lived  $^{222}\text{Rn}$  daughters were collected in a liquid scintillator; we measured total  $\alpha$  and  $\beta$  activity with 100% counting efficiency by a modified integral counting method. Based on the obtained effective half-life (*ca.* 38 min) of the total  $\alpha$  and  $\beta$  activity, we determined the corresponding activity at the end of sampling. The sensitivity of the method is  $2.06 \times 10^{-3}$  Bq liter $^{-1}$  for each nuclide, which is adequate for monitoring the levels of these nuclides in homes. We present preliminary local measurements of indoor  $^{222}\text{Rn}$  daughters.

## INTRODUCTION

The hazards to health and the environment of airborne daughters of naturally occurring  $^{222}\text{Rn}$  have long been recognized. Therefore, it is important to determine widely and routinely the concentration of  $^{222}\text{Rn}$  and its decay products in air. A common method to determine the  $^{222}\text{Rn}$  decay products in air is to filter a large volume of air and to assay the radioactivity left on the filter with an alpha counter, such as an ionization chamber or a ZnS(Ag) scintillation counter. Instead of using the air filter/ $\alpha$  counter technique, we can now substitute an  $\alpha$  spectrometer. However, this method usually measures only  $\alpha$ -emitting daughters (*i.e.*,  $^{218}\text{Po}$ ,  $^{214}\text{Po}$ ) with counting efficiency <50%.

The problem of sample self-absorption and the memory effect of the detection caused by the deposition of short-lived  $^{222}\text{Rn}$  daughters can be troublesome. Here we report a relatively simple and quick method for determining airborne  $^{222}\text{Rn}$  daughters using a membrane filter and a liquid scintillation spectrometer. We dissolved directly in the liquid scintillator a 0.65- $\mu\text{m}$ -pore cellulose nitrate membrane filter, on which the short-lived  $^{222}\text{Rn}$  daughters had been collected. Based on the analysis of the buildup and decay curve from the initially isolated daughter nuclides, which corresponds to  $t_{1/2} = 38.3$  min, we determined the absolute disintegration rate of alpha- and beta-emitting  $^{222}\text{Rn}$  daughters by a modified integral counting method (MICM) of liquid scintillation counting (LSC). Our results indicate that this method is suitable for the measurement of environmental  $^{222}\text{Rn}$  products, because a large volume of air can be determined using a commercially available LS spectrometer.

## METHODS

We collected the short-lived  $^{222}\text{Rn}$  daughters on a 0.65- $\mu\text{m}$ -pore cellulose nitrate membrane filter (Advantec Toyo Co., Ltd., Tokyo, Japan) using a high-volume air-sampling pump. Air was sampled typically for 40 min at  $\sim 5$  liter  $\text{min}^{-1}$ . Then, the sample filter, on which short-lived  $^{222}\text{Rn}$  daughters collected, was placed in a standard counting vial with a few drops of water and 2 ml of 2-methoxymethanol and stirred mechanically for 2 min. During this time, the filter was dissolved completely. Then, 19 ml of liquid scintillator solution was added to a total volume of 21-ml solution. The liquid scintillator used in this work is an aqueous counting scintillant, ACSII® (Amersham Corp., Chicago, Illinois, USA).

Pulse-height distributions were measured using an Aloka LS spectrometer, Model LSC-3500 (Aloka Co., Ltd., Tokyo, Japan). To obtain the disintegration rates of  $^{222}\text{Rn}$  daughters, we modified the integral counting method (Horrocks 1964; Goldstein 1965; Homma & Murakami 1977) with the

LS spectrometer to take into account the detection threshold of LS system. The sample filter was also measured with an intrinsic Ge detector (Princeton Gamma-Tech) coupled to a 4096-channel analyzer (Canberra Series 35).

## RESULTS

Figure 1A shows the pulse-height spectrum for the sample measured with a 1024 channel analyzer of an Aloka LSC-3500 LS spectrometer over the energy region ~1–700 keV; the  $\alpha$  peaks of  $^{218}\text{Po}$  and  $^{214}\text{Po}$  were superimposed on the  $\beta$  continuum of the daughters,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ . Figure 1B represents the determination of the sample by an MICM. The absolute disintegration rate of the sample can be determined more accurately than the conventional integral counting method (CICM) by extrapolating the integral pulse-height spectrum to the detection threshold. Details of the MICM are reported elsewhere (Homma, Murase & Handa, ms.).

Figure 2 shows a typical plot of the total activity of Rn daughters as a function of time elapsed after the sample was prepared for counting. The decay curve of the total activity has an effective half-life of 38.3 min between 20 and 70 min after collection. The observed decay patterns of the samples point clearly to the absence of any significant adsorption of  $^{222}\text{Rn}$  on the sample filter; the collection of  $^{222}\text{Rn}$  on the filter would yield an increased effective half-life for the filter activity. If we assume that equal activities of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$  are present at the end of the sampling (*i.e.*,  $A^0 = N_A\lambda_A = N_B\lambda_B = N_C\lambda_C = N_{\dot{C}}\lambda_{\dot{C}}$ ), and that the parent,  $^{222}\text{Rn}$ , is removed from the filter as described above, the activity of individual daughter products is then given by

$$A_A = A^0 e^{-\lambda_A t} \quad (1)$$

$$A_B = A^0 e^{-\lambda_B t} + A^0 \lambda_B \left[ \frac{e^{-\lambda_A t}}{\lambda_B - \lambda_A} + \frac{e^{-\lambda_B t}}{\lambda_A - \lambda_B} \right] \quad (2)$$

$$A_C = A^0 e^{-\lambda_C t} + A^0 \lambda_C \left[ \frac{e^{-\lambda_B t}}{\lambda_C - \lambda_B} + \frac{e^{-\lambda_C t}}{\lambda_B - \lambda_C} \right] + A^0 \lambda_B \lambda_C \left( \frac{e^{-\lambda_A t}}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)} + \frac{e^{-\lambda_B t}}{(\lambda_A - \lambda_B)(\lambda_C - \lambda_B)} + \frac{e^{-\lambda_C t}}{(\lambda_A - \lambda_C)(\lambda_B - \lambda_C)} \right) \quad (3)$$

where  $N_0$  is the number of atoms at the end of sampling,  $N$  is the number of atoms at time,  $t$ ,  $A^0$  is the activity of nuclide at the end of the sampling,  $A$  is the activity of nuclide at time,  $t$ ,  $\lambda$  is the decay constant of nuclide, and suffixes A, B, C and  $\dot{C}$  refer to  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ , respectively. We assumed that the activity of  $^{214}\text{Po}$  equals that of  $^{214}\text{Bi}$ , *i.e.*,  $A_{\dot{C}} = A_C$ , because we regard the transition of  $^{214}\text{Bi}$  (19.8 min) to  $^{210}\text{Pb}$  (22.3 yr), *via*  $^{214}\text{Po}$  (164  $\mu\text{sec}$ ), as a single event involving simultaneous  $\alpha$  and  $\beta$  emission.

Figure 2 also shows the activity of separate daughter products and the consequent growth and decay of the total  $\alpha$  and  $\beta$  activity calculated from the equations described above. The total  $\alpha$  and  $\beta$  activity has an effective half-life of 38.7 min between 20 and 70 min, which seems to be characteristic of an initial collection of nearly equal activities of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ .

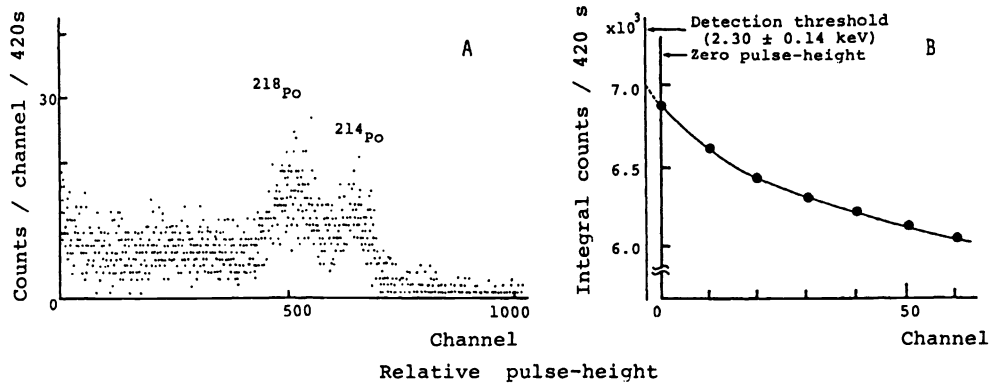


Fig. 1. Determination of the absolute disintegration rate of the sample by the MICM. A. Differential pulse-height spectrum; B. Integral pulse-height spectrum.

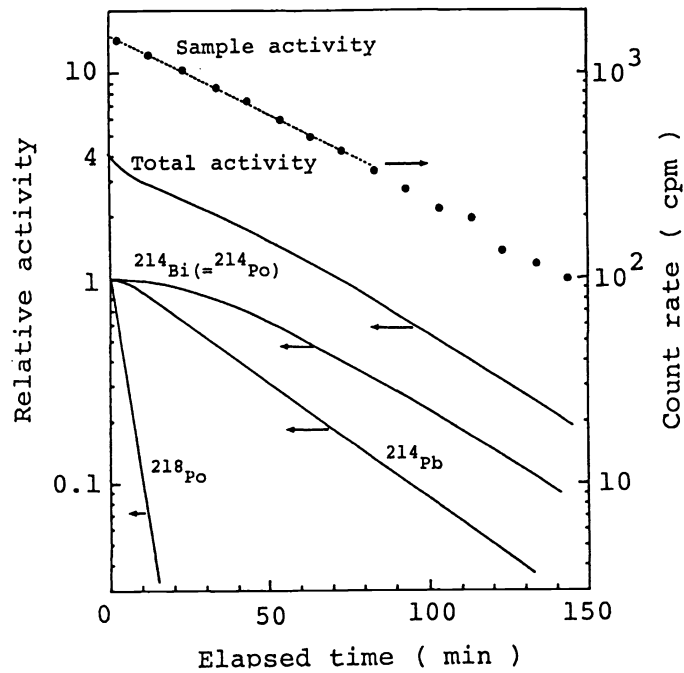


Fig. 2. Comparison of a decay curve of sample activity and the calculated activity of separate  $^{222}\text{Rn}$  daughters and the consequent growth and decay of the total  $\alpha$  and  $\beta$  activity. • = sample activity; — = calculated activity.

Because of the resemblance in the decay curve (Fig. 2), and because two  $\alpha$  peaks and the  $\beta$  continuum have nearly equal count rates (Fig. 1A), it is reasonable to assume that sample radioactivities are due to  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ . Radioactive equilibrium appears to exist among its daughters;  $\gamma$ -ray spectra of the sample filter measured with a Ge-spectrometer indicate that the growth and decay of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  are identical with those obtained by LSC.

TABLE 1. Concentration of  $^{222}\text{Rn}$  Daughters at Kyoritsu College of Pharmacy

Sampling locations		Concentration ( $\times 10 \text{ Bq m}^{-3}$ )	Sampling locations		Concentration ( $\times 10 \text{ Bq m}^{-3}$ )
Warehouse	1	$4.5 \pm 0.1$	Office	1	$1.2 \pm 0.1$
	2	$2.1 \pm 0.1$		2	$5.6 \pm 0.1$
	3	$9.9 \pm 0.2$		3	$1.6 \pm 0.1$
	4	$2.1 \pm 0.1$		4	$2.2 \pm 0.1$
	5	$24.7 \pm 0.3$		5	$2.1 \pm 0.1$
	6	$18.5 \pm 0.2$		6	$1.7 \pm 0.1$
	7	$15.2 \pm 0.2$		7	$0.9 \pm 0.1$
	8	$1.1 \pm 0.1$	Library		$6.3 \pm 0.1$
Laboratory	1	$4.0 \pm 0.1$	Library stock room		$4.3 \pm 0.1$
	2	$8.2 \pm 0.2$	Lecture room 1		$3.5 \pm 0.1$
	3	$0.7 \pm 0.1$	2		$7.9 \pm 0.2$
	4	$1.0 \pm 0.1$	3		$6.6 \pm 0.1$
	5	$2.3 \pm 0.1$	Gymnasium 1		$8.2 \pm 0.2$
	6	$2.5 \pm 0.1$	2		$3.0 \pm 0.1$
	7	$1.2 \pm 0.1$	Auditorium		$9.2 \pm 0.2$
	8	$0.6 \pm 0.1$	Platform 1		$0.8 \pm 0.1$
	9	$2.5 \pm 0.1$	2		$1.4 \pm 0.1$

Measurements of indoor  $^{222}\text{Rn}$  daughters in and around our university were carried out by MICM; Table 1 lists the results. If “ $f$ ”, the fraction of  $^{222}\text{Rn}$  daughters that is free and not combined with particulate matter, is known accurately, the measured daughter activities may be converted to  $^{222}\text{Rn}$  activity in air. However, a wide variety of  $f$  values have been reported. Some investigators claimed that, even with its short half-life, the first  $^{222}\text{Rn}$  daughter,  $^{218}\text{Po}$  ( $t_{1/2} = 3.05 \text{ min}$ ), and the subsequent daughters are not always in equilibrium with the parent,  $^{222}\text{Rn}$ . Thus, this method is useful only for approximate environmental measurements of  $^{222}\text{Rn}$  in air. The sensitivity of the method for 220-liter samples is  $2.06 \times 10^{-3} \text{ liter}^{-1}$  for each nuclide, with 10% fractional error at 95% confidence.

As previously described (Murase, Homma & Takiue 1989),  $^{222}\text{Rn}$  and its  $\alpha$ -emitting daughters produce air luminescence in the gas above the liquid scintillator in the sample vial, yielding counts that cause errors in quantitative LS measurement. However, the sample vial was almost full of liquid scintillator solution, and  $^{222}\text{Rn}$  was not collected in this study. Hence, we neglected the error due to air luminescence counts.

Interference can occur from the long-lived thoron ( $^{220}\text{Rn}$ ) daughters. However,  $^{220}\text{Rn}$  ( $t_{1/2} = 55.6 \text{ sec}$ ) and its first daughter,  $^{216}\text{Po}$  ( $t_{1/2} = 0.145 \text{ sec}$ ), decay substantially in a few minutes and the activity due to the Th series then increases slowly as the product  $^{212}\text{Pb}$  increases with a half-life of 10.64 h. Thus, these interferences can be corrected by recounting the sample after a delay of a few hours. The method has the following advantages: simple and reliable sample preparation; 100% counting efficiency (for  $\alpha$  particles and also for  $\beta$  particles from  $^{222}\text{Rn}$  daughters); low detection limit ( $2.06 \times 10^{-3} \text{ Bq liter}^{-1}$ ) for each nuclide; no sample self-absorption associated with deposited samples; and freedom from memory effect of the detector caused by the deposition of short-lived  $^{222}\text{Rn}$  daughters.

## REFERENCES

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