*Journal of the Radiation Protection*
*Vol. 4, No. 1, July, 1979*

**<Original>**

# The Measurement of Airborne Radon Daughter Concentrations in the Atmosphere

**Chung Woo Ha, Jai Ki Lee, Philip S. Moon**

*Korea Atomic Energy Research Institute, Seoul, Korea*

**Chong Chul Yook**

*Dept. of Nuclear Engineering Han Yang University, Seoul, Korea*

**Abstract**

A simple method for determining the airborne concentration of radon daughter products has been developed, which is based on gross alpha counting of the air filter collections at several time intervals after completion of air sampling. The concentration of each nuclide is then obtained from an equation involving the alpha disintegrations, the sampling time, and the known numerical coefficients. The state of radioactive disequilibrium is also investigated. The atmosphere sampled in the TRIGA Mark-III reactor room was largely in disequilibrium. The extent of radioactive disequilibrium between radon daughter products seems likely depend on sampling times associated with turbulence conditions. The data obtained here will certainly provide useful information on the evaluation of internal exposure and calibration of effluent monitoring instruments.

## 1. Introduction

The commonly used methods for monitoring particulate radioactivity in the air depend on sampling, using suction of air through a suitably selected filter medium. Activity collected on the filter is measured by a suitable detector. Usually a continuous air-monitoring instrument is used to have a continuous check on the rate of change of airborne radioactivity. If the activity at the place of moitoring includs short-lived activity, a few hours' delay is given to measure the long- lived activity. An alarm incorporated in an air monitor is used to give warning of incidents of high activity.

The air monitoring systems based on the above techniques are in common use in most nuclear installations. However, these systems generally do not provide a true guidance on the inhalation exposure of workers without due consideration to factors such as the size distribution, the nature of contamination in working areas by detailed studies of the type of radionuclides likely to be release in the air, the specific radioactivity of individual particles, and the environmental factors such as humidity, temperature, air-circulation pattern, background radiation level, etc.

In view of the great importance of the assessment of inhalation exposure, it is necessary to establish a fundamental criteria

of the natural radioactivity of Radon/Thoron and it's daughter products in interpretation of data. However, to date, there are no studies to provide evidence for the state of equilibria among the radon daughters in the atmosphere of both TRIGA Mark-III reactor hall and open air. Even if the evidence did become available, it would still be necessary to give some thought to the radioactive equilibrium in order to find the parameters in which the (MPC)a is best expressed, and hence to find what quantities the health physicist should measure when assessing the hazard from fission products released into air. Although this work was motivated by the desire to have accurate data when analyzing airborne radioactivity in the TRIGA Mark-III reactor hall, other applications include laboratory studies of the physics of the radon daughter aerosols and studies of the behavior of the radon daughters in standard air-sampling techniques for where evaluation of the state of equilibrium is important. This paper is the direct outgrowth of this need. It does not introduce a new-method of sampling, but rather a method of interpreting daughter products activities in a filter sample with improved accuracy. This method presented here also provides estimates of error in the nuclide concentrations as well as useful information for estimating a priori the accuracy of the nuclide concentration determinations as a function of sampling time and counting times. The data obtained here will certainly provide useful information on the evaluation of internal exposure and calibration of effluent monitoring instruments.

## 2. Decay Properties of Radon and Thoron

The sequence of successive decay products of radon and its daughters is shown in Figure 1. Since alpha particles are the only radiation which will be considered, the nuclides which decay by alpha emission are of primary interest. Only 0.02% of the decays of Radium A result in the emission of $\beta$ particles so that Radium A may be reasonably considered as a pure alpha particle emitter. Although Radium C is not an alpha emitter, it effectively behaves as one, since it primarily decays to Radium C′ which then instantantaneously emits alpha particles. Radium B, although not an alpha emitter, decays to Radium C, and therefore contributes to the appearance of alpha particles from Radium C.

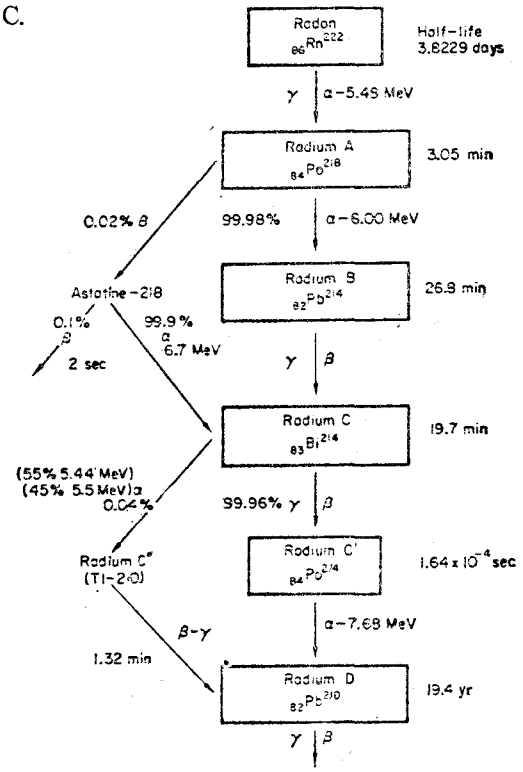



**Fig. 1.** Radon decay chart

Figure 2 shows the decay scheme of Thoron and its daughters. The short half-life of Thorium A makes it impossible to differentiate from Thorium B in this analysis regimen; therefore, collection of Thorium B

atoms really means collection of both Thorium A and Thorium B. Thorium B is not an alpha emitter, but it is important because it decays relatively slowly to Thorium C. Thorium C is effectively an alpha emitter in that 33.7% decays by alpha emission and 66.3% decays by beta emission to Thorium C′ which instantaneously emits an alpha particle. Thorium D is stable and need not be considered.

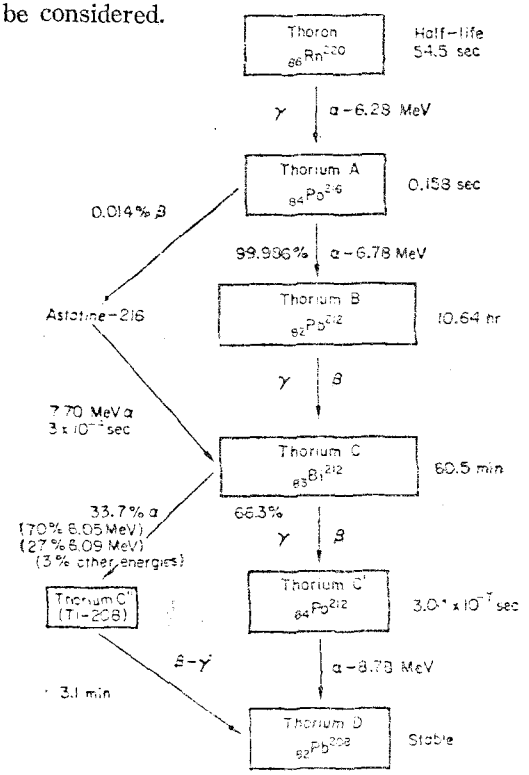



**Fig. 2.** Thoron decay chart

### 3. Mathematical Formulation

The mathematical formulation relevant to these studies are available in literatures[1] and will be described only briefly. Since the half-lives of first three daughter products are small compared to that of radon, radon daughter activity can be approximately described as

$$N_i = N_0 \sum_{i=0}^{n} R_i e^{-\lambda_i t} \quad \cdots\cdots (1)$$

where

$$R_i = \frac{\lambda_i}{\lambda_n} \prod_{j=1, j\neq i}^{n} \frac{\lambda_i}{(\lambda_j - \lambda_i)}$$

Here $t$ is the build-up time (or sampling time) in minutes and $N_0$ is the intial activity of radon. If $t$ is specifically considered to refer to the time at the end of sample collection, the $N_i$ values given in these equations describe the numbers of atoms of each of the isotopes on the sample at the end of collection. $\bar{N}_i$ is used to refer to these total sample quantities.

To differentiate the sampling period from the decay and observation period a new time variable, $\theta$, is defined with $\theta=0$ at the end of collection. Therefore, $N_j(t)=\bar{N}_j$=the number of of atoms present at $\theta=0$, Thus in the $\theta$ coordinate system, $N_j(0)=\bar{N}_j$

The activity on the filter sample for a collection time $J$ at time $\theta$ after end of collection can be calculated using the following equations;

$$M(i) = F\left\{ \frac{1}{\lambda_i} - \sum_{k=1}^{i} \frac{e^{-\lambda_i J}}{\lambda_k} \cdot \frac{\prod_{n=1}^{i-1} \lambda_n}{\prod_{n=1}^{i} (\lambda_n - \lambda_k)} \right\} \quad \cdots (2)$$

Where $M(i)$ is the $i$-th daughter of a parent which is collected at the rate of $F$ atoms per unit time ($F$ involves concentration, flow-rate and collection efficiency). We will have RaA, RaB, and RaC from direct collection of the three isotopes as well as ingrowth due to parents. Integration of the equation between times $\alpha$ and $\beta$ after end of collection gives disintegration rates, $D(\alpha, \beta)$ for that period. Thus, with the aid of computer, the variation of the decay curves of RaA, RaB, RaC and total activities can be studied as a function of the parameters, sample collection time ($J$), and time interval

after end of sample collection ($\theta$).

The general equations giving the nuclide activity concentrations are of the form

$$Q_i=\frac{1}{VE}\sum_j K_{ij}D_j\ {}_pC_i/l(i=1,6,j=1,6)\cdots\ (3)$$

Where $V$ is the the flow rate in $ft^3$/min. and $E$ is the detector efficiency in cpm/dpm. The coefficient $K_{ij}$ depends on the values of the sample collection time($J$) and the time interval after end of sample collection ($\theta$). $D_j$, which is total alpha counts observed by detector during counting period $\alpha$ to $\beta$, is described by the following equations.

$$D_i(\alpha_i,\beta_i)=\sum_j H_{ji}N_j^0(i=1,6,j=1,6)\cdots\ (4)$$

Equation (4) is an equation in two independent variables with 6 unknowns. The equation could be solved using simplified simultaneous equation method of Tsivoglou et al.[2] which is commonly employed, but a better solution is to perform a weighted least-squares regression analysis[3] on an entire set of various count determinations.

Thus the regression analysis is applied on Eq. (4), described more briefly as $D_i=f(\alpha_i,\beta_i)$, with six unknowns, $\overline{N}_j^0$ $j=1,6$ and the fit is computed with a set of $n$ data points.

A weighting factor, $\omega_i=1$/variance of $D_i$ must be considered with each data determination since, although the statistical errors of the data points are normally distributed as required in regression analysis, the variance of these errors are unique and different.[3]

Therefore, the system which is to be minimized is;

$$\sum_{i=1}^{n}\{\omega_i[f(\alpha_i,\beta_i)-D_i]^2\}\cdots\cdots\cdots\cdots\cdots\cdots\ (5)$$

The minimization is accomplished by taking the partial derivatives with respect to the 6-parameters, yielding 6 equations in 6 unknowns summed over the entire set of data points. This system of equation may be described in matrix form as;

$$[\mathbf{D}]=[\mathbf{H}][\mathbf{N}]\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\ (6)$$

where $[H]$ is a matrix having elements;

$$\mathbf{H}_{jk}'=\sum_i \omega_i h_{ji}h_{kj}'\ (j=1,6,\ k=1,6)$$

with the $h$ values given by;

$$\left.\begin{aligned}
h_{1i}&=-\left(1+\frac{R_2}{\lambda_1}\right)(e^{-\lambda_1\beta_i}-e^{-\lambda_1\alpha_i}-\left(\frac{R_2}{\lambda_2}\right)\\
&\quad(e^{-\lambda_3\beta_i}-e^{-\lambda_2\alpha_i})+\left(\frac{R_1+R_2}{\lambda_3}\right)\\
&\quad(e^{-\lambda_3\beta_i}-e^{-\lambda_3\alpha_i})\\
h_{2i}&=-\frac{R_3}{\lambda_2}(e^{-\lambda_2\beta_i}-e^{-\lambda_2\alpha_i})+\frac{R_3}{\lambda_3}\\
&\quad(e^{-\lambda_3\beta_i}-e^{-\lambda_3\alpha_i})\\
h_{3i}&=-(e^{-\lambda_3\beta_i}-e^{-\lambda_3\alpha_i})\\
h_{4i}&=\frac{R_4}{\lambda_4}(e^{-\lambda_4\beta_i}-e^{-\lambda_4\alpha_i})-\frac{R_4}{\lambda_5}\\
&\quad(e^{-\lambda_5\beta_i}-e^{-\lambda_5\alpha_i})\\
h_{5i}&=-(e^{-\lambda_5\beta_i}-e^{-\lambda_5\alpha_i})
\end{aligned}\right\}\cdots\ (7)$$

and with

$$\mathbf{N}=\begin{pmatrix}N_1^0\\N_2^0\\N_3^0\\N_4^0\\N_5^0\end{pmatrix}$$

Solving this system, equation (4) by Gauss-Jordan method, values for $N_j^0$ are obtained.

On substituting these values into the resulting equation from combination of Eq. (3) with Eq. (4) coefficient $K_{ij}$ can be obtained,

Each of the values of the parameters $N_j^0$ has a statistical error associated with it. An estimate of the standard deviations, $A_j$ of the $i$-th value is given by;

$$A_j=S\sqrt{\sum_{i=0}^{k-1}\sum_{j=0}^{k-1}x_{ij}\ x_{mi}\ C_{im}}\cdots\cdots\cdots\cdots\cdots\cdots\ (8)$$

or in matrix notation

$$A_j=S\sqrt{I_{jj}}$$

where $I_{jj}$ is the $j$-th diagonal element of the matrix [I] which is the inverse of the matrix [H] in equation (5) and $S^2$ is defined by

$$S^2=\frac{\sum_{i=1}^{n}\omega_i[f(\alpha_i,\beta_i)-D_i]^2}{n-2} \quad \cdots\cdots\cdots\cdots (9)$$

In order to increase the accuracy of the computed quantities of Radon/Thoron daughters on each sample, the value of each calculated $N_i^0$ was compared to its standard error. If its value was less than twice its standard error, it was assumed to be effectively absent from the sample, and the analysis was performed again with one less unknown.

## 4. Selection of Sampling and Counting Times

It is evident that the following variables affect accuracy; sampling times, counting times, nuclide ratios, counter efficiency and sampling flow rate. Since it was desired to select operating conditions for high accuracy, a computer program* has been developed from which concentration, standard deviation equations, and coefficients, $K_{ij}$ could be obtained for different sampling and counting times. The program yielded equations (6) and (7) with numerical coefficients depending on the sampling and counting times. These programs are written in the FORTRAN IV for use on the CYBER 73-18 computer. The sampling times chosen for investigation were, 5, 10, 20, 30, 40, 50 and 60 min. For each of these times, a number of different counting time interval sets were investigated, with the start of the counting period set at 0.5 min. after the end of sampling and the finishing time set at 20 min. or 40 min. after the end of sampling. To allow time for recording alpha counts and resetting the scaler, 0.5 min. or 1 min. was allowed between the time interval counts.

Twenty time interval sets ending at 20 min. and at 40 min., respectively, were investigated for each of the seven sampling times.

*The computer program used in this investigation is available from the authors upon request.

## 5. Experimental

In our experiments, collection rates were 1 ft³/min. obtained using the low volume air sampler (Bendix, Co.) and milipore filter papers (HAWP 04700) of dimensions, 47 mm diameter. Collection times varied from 5 to 60 min. The samples were counted after a delay of 0.5 min. for successive intervals of 0.5 min. and 1 min. respectively. The sample collections were made in the open atmosphere and the room of TRIGA Mark-III reactor.

The counting of the samples was done using a 100 mm$\phi$ ZnS(Ag) detector. The large size of the crystal gave good efficiency for the high energy alpha particles. The detector was calibrated using standard uranium source certified by Japan Radioisotope Association (JRIA). The counting efficiency of the system which was deduced from the source-detector geometry and also by use of a calibrated uranium alpha source was found to be 5%.

## 6. Results and Discussion

Equilibrium ratio of radon daugters determined for several hundred samples by the method outlined in this paper are given in Table 1 for the assumed conditions of continuous and instantaneous emission of radon from the soil and for secular equilibrium in the atmosphere. Figure 3 gives decay curves

of RaA, RaB, and RaC or the filter samples, respectively, for different sample collection times and counting intervals. As explained, such curves can be analyzed for RaA, RaB, and RaC using a weighted least square method. Such a method has been presented in detail by Raabe[4] for alpha decay curves. Figure 4 gives the alpha decay on filter sample from $^{222}Rn$ daughters as a function of time resulting from the regression analysis.

It can be found from Fig. 3 that the higher accuracy can be possible, particulary for RaA, as the sampling time increases and the starting time of counting after the end of sampling is shortened. Therefore, longer sampling times are desirable for better estimates of the working level. In retrospect, it appears that it is also important to count well at times sufficiently long after collection so that RaA has decayed to better-analyze for RaB and RaC.

It can also be noted that the atmosphere sampled in the reactor room are largely in disquilibrium and the extent of radioactive disequilibrium between radon daughter products seems likely to have a decreasing tendancy with shortening the sampling time. It should be mentioned that the average activity of RaA as well as of RaB, RaD and RaC′ of $2.9 \times 10^{-7}$ $\mu$Ci/cc is much higher than the

**Table 1.** Equilibrium ratio of radon daughters in the TRIGA Mark-III reactor room.

| Sample No. | Sampling Time | Counting Time interval | Calculated Equilibrium Ratio | Concentration pCi/ | | |
|---|---|---|---|---|---|---|
| | | | | RaA | RaB | RaC |
| 1—1 | 5 min | 30 sec | 1 : 0.98 : 0.71 | 289. | 282. | 205. |
| 1—2 | 10 min | 30 sec | 1 : 1.11 : 0.72 | 329. | 364. | 236. |
| 1—3 | 20 min | 30 sec | 1 : 0.68 : 0.39 | 559. | 383. | 219. |
| 1—4 | 30 min | 30 sec | — | | unstable | |
| 1—5 | 40 min | 30 sec | 1 : 0.38 : 0.19 | 1121. | 431. | 208. |
| 1—6 | 50 min | 30 sec | 1 : 0.42 : 0.17 | 1119. | 470. | 186. |
| 1—7 | 60 min | 30 sec | 1 : 3.26 : 3.42 | 131. | 427. | 448. |
| 1—8 | 5 min | 60 sec | 1 : 1.15 : 1.37 | 78. | 90. | 107. |
| 1—9 | 10 min | 60 sec | 1 : 0.86 : 0.70 | 167. | 143. | 117. |
| 1—10 | 20 min | 60 sec | — | | unstable | |
| 1—11 | 30 min | 60 sec | 1 : 0.70 : 0.48 | 260. | 252. | 172. |
| 1—12 | 40 min | 60 sec | 1 : 1.18 : 0.97 | 247. | 291. | 239. |
| 1—13 | 50 min | 60 sec | 1 : 0.73 : 0.61 | 440. | 319. | 267. |
| 1—14 | 60 min | 60 sec | 1 : 0.93 : 0.73 | 442. | 410. | 321. |

internationally accepted maximum level of activity of $^{222}Rn$ in radioactive equilibrium with its daughter products for continuous exposure of the general public. Tóth[5] has recently reported similar levels in unventilated dwellings in Hungary.

The origin of the fluctuations in the concentrations of the daughter products in the reactor room will be a topic for further investigation. However, it may be noted that a sudden change in the RaA concentration usually corresponds to an opposite change in the atmosphere conditions, suggesting a possible dependence of the $^{222}Rn$ emanation rate on the atmospheric pressure.

It is felt that the technique is simple and

Fig. 3-A.

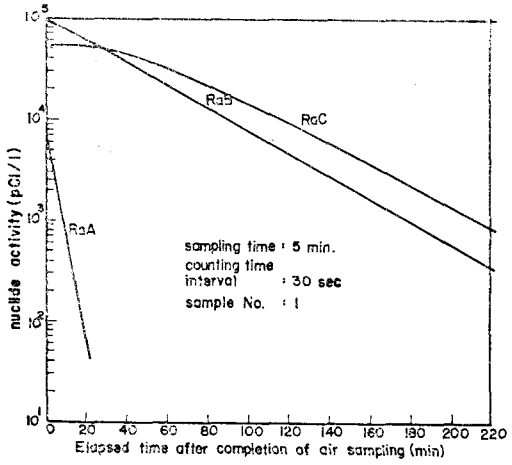



Fig. 3-B.

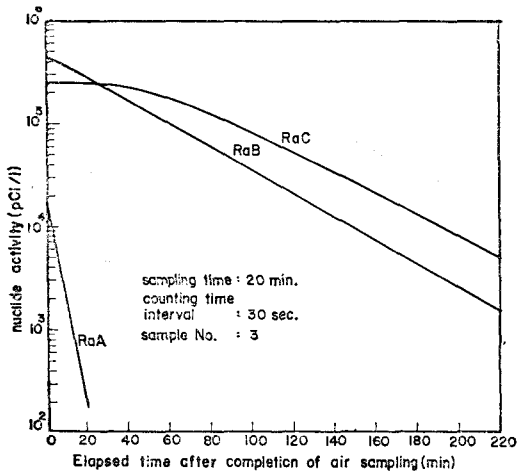



Fig. 3-C.

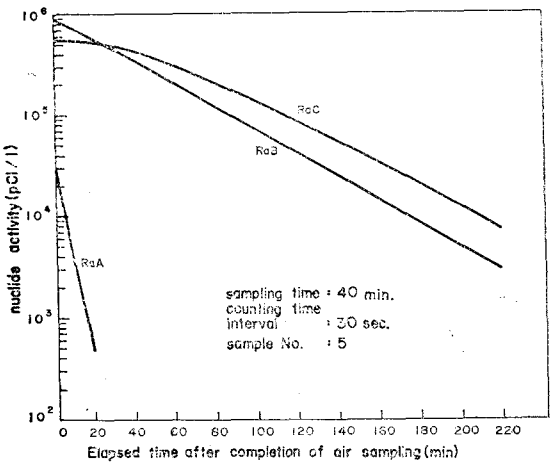



Fig. 3-D.

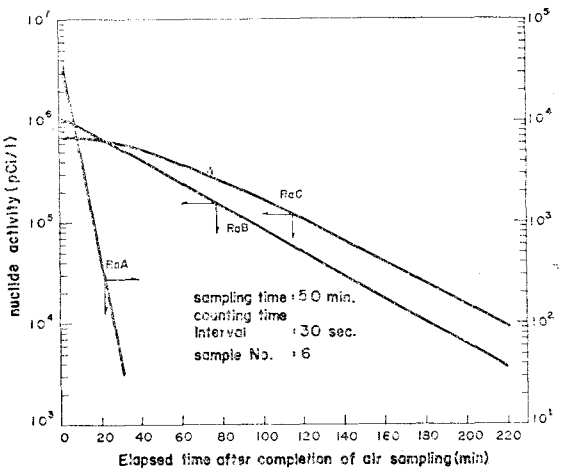



Fig. 3-E.

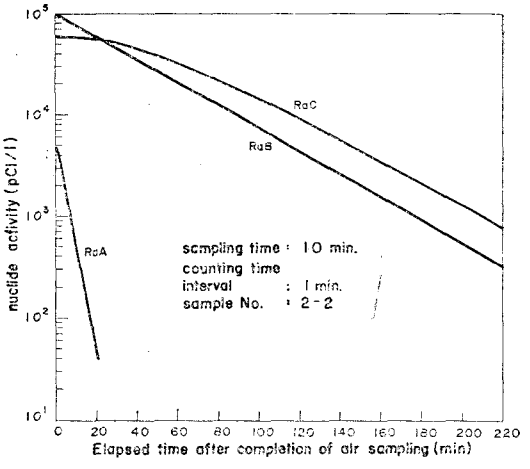



Fig. 3-F.

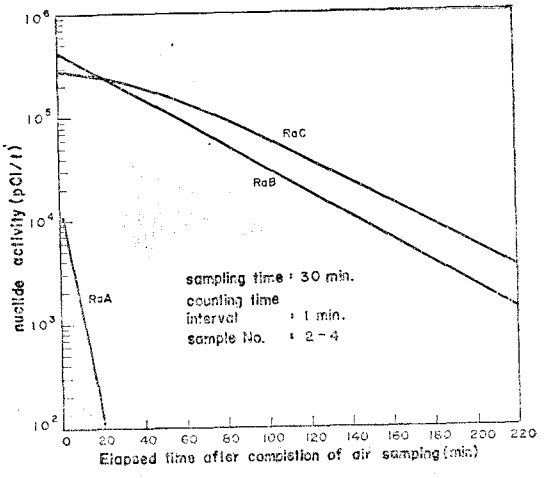



**Fig. 3.** Decay curves of RaA, RaB and RaC on the filter sample for different sample collection times.

Fig. 3-G.

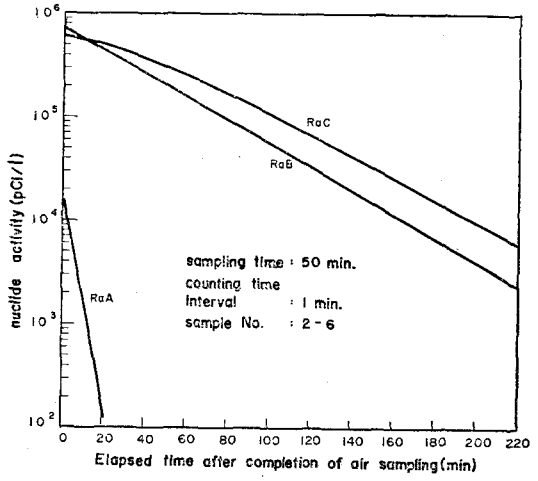
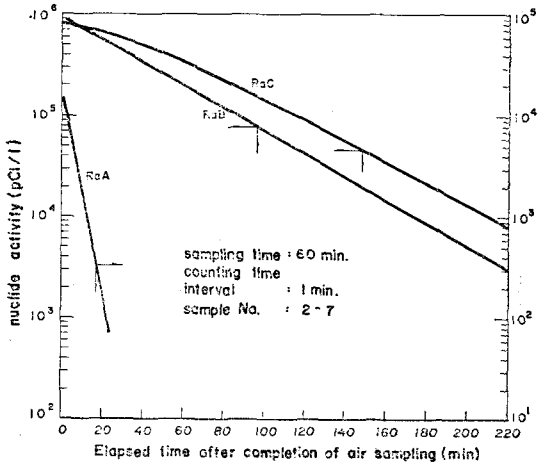



Fig. 3-H.



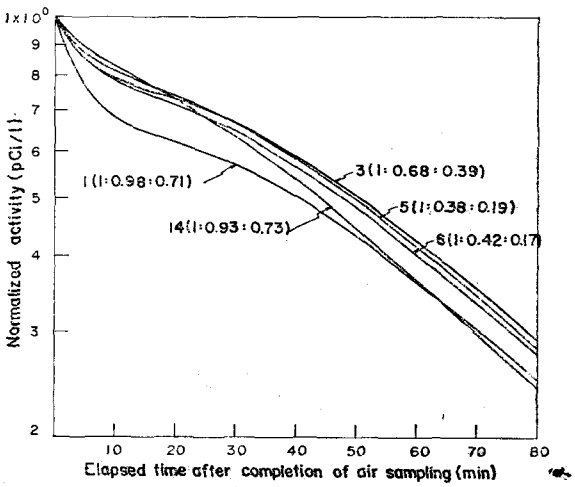




**Fig. 4.** Observed alpha decay curves of Radon daughter products on filter sample with different disequilibrium for different sampling times.

will be very useful in relating turbulence condition to disequilibrium of radon daughters.

## 7. Conclusion

The result for measurement of radon daughter activity concentration will certainly provide useful information on the evaluation of internal exposure and calibration of effluent monitoring instruments. The state of radioactive disequilibrium of short-lived radon daughters can be estimated in the open atmosphere with improved accuracy.

## Acknowledgement

The authors express deeply their appreciation to Mr. Yong Dai Han for his enthusiastic assistance in sampling and counting the nmerous samples.

## References

1. B. Hultgvist, Studies on Naturally Occurring Ionizing Radiations, Almqvist and Wiksells, A. B., Stockholm (1956).
2. E.C. Tsivoglou, H.E. Ayer, and D.A. Holaday: Nucleonics, **11**(9), 40 (1953).
3. R.G.D. Steel and J.H. Torrie, Principles and Procedures of Statistics, *pp.180-181*, McGraw-Hill, New York (1960).
4. O.G. Raabe and M.E. Wrenn, Health phys., **17**, 593 (1969).
5. A. Tôth, Health Phys., **23**, 281 (1972).

## 대기중 라돈 붕괴생성물의 공기중 방사능 농도의 측정

하정우, 이재기, 문석형
한국원자력연구소 방사선관리실

육 종 철
한양대학교 원자력공학과

## 개 요

공기시료채집 종료후 공기여과지에 채집된 시료중 방사능을 일정한 시간구간을 두어 계측함으로써 얻은 붕괴곡선을 이론적 방법에 의하여 분석할 수 있는 간단한 방법을 개발하였으며, 이 방법을 이용하여 라돈 붕괴생성물 각각의 공기중 방사능 농도를 결정하였다. 라돈 붕괴생성물 각 핵종의 방사능 농도는 알파붕괴, 시료채집시간, 그리고 수치계수의 함수로 표시된 방정식으로 부터 얻었다. 그리고 대기중 라돈 붕괴생성물 개개의 방사평형상태도 또한 조사하였다. TRIGA Mark-III 원자로실내에서 채집한 공기시료는 상당히 비평형상태에 있었다. 라돈 붕괴생성물들 간의 방사성 불평형의 정도는 공기와류조건과 관련된 공기시료 채집시간에 따라 상당히 달라지는 것같았다.

본 연구 결과에서 얻은 자료는 인체 내부방사선 피폭선량평가와 기체 방사성 물질 방출감시기 교정에 유용한 기초자료가될 것이 확실하다.