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Measurement of Radon Daughters in Airborne Dust

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—ABSTRACT—

A simple method has been established for determining RaA, RaB and RaC concentrations in airborne dust. This is to evaluate the concentration from measurement of total alpha activities in three selected-time intervals after an air sample is taken from the membrane filter paper (mean pore size: 0.8μm). As a preliminary trial, a time-variation of the concentrations has been determined using the single-filter method at the KAERI site (N. Lat. 37°38′ and E. Long 127°15′), Seoul, Korea.

It appears that there is a large variation of the concentrations depending on the sampling time. Generally the highest value was observed in the morning that may coincide with the highest density of atmosphere in a day while the lowest value was obtained around fourteen o'clock.

1. Introduction

The presence of the radon daughters in air makes it difficult to evaluate the artificial airborne radioactive concontration in the working environment of nuclear facilities.

Various methods have been suggested for determining the radon daughters' concentration. Among them, the single-filter method has been widely used because of simplicity [1)-4)]. This is to determine the concentration from measurements of total alpha activities in three selected-time intervals after air sample is taken.

The purpose of this study was to investigate a time-variation of radon daughters in airborne dust at the KAERI site (N. Lat. 37°38′ and E. Long. 127°15′) using the single-filter method. An air sample was taken from the membrane filter paper with mean pore size of 0.8 μm. Subsequent total alpha activities were measured by a 2π-internal proportional counter in three selected-time intervals, and the concentration was theoretically calculated.

2. Theoretical Background

^{222}Rn, a naturally occurring member of uranium ecay series, is daughter product of ^{226}Ra and ends with the stable isotope, i.e., ^{206}Pb. Usually it exists as an inert gas together with its daughters in the surrounding atmosphere. The characteristics of radon and its daughters is described in Table 1. [5), 6)] It is well known that ^{226}Ra has a half life (1620 years) much longer than its successive dau-

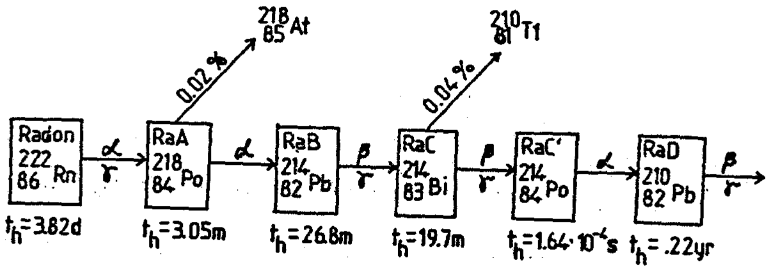

Fig. 1. Radon Decay Series

ghters (see Fig. 1). This makes it to be a fixed-rate source of ^{222}Rn whose half life is 3.825 days.

Since ^{222}Rn is the inert gas, it passes freely through the filter paper and thus does not retain on it. The chemically active daughters, i.e., RaA, RaB, Rac, RaC′, however, tend to adhere easily to the airborne dust in the atmosphere, so they can be readily sampled by the filtering method. RaC′ is assumed to be RaC because it has a very short half life of 160 μsec compared to that of its precursor. Since the half life of RaD, i.e., 22 years, it is assumed to be of semi-stable isotope for simplicity of calculation. In many cases, this simplification is the usual way.

Prior to a derivation of the relevant equation, some nomenclatures to be used are described as follows:

N_i=The number of atoms of the i-th isotope on the filter paper.

N_i^0=The number of the i-th isotope on the filter paper at the end of sampling.

T=The duration of sampling (min).

Q_i=The atmospheric concentration of the i-th isotope (atoms/l).

V=The sampling flow rate (l/min).

λ_i=The disintegration constant of the i-th isotope (min^{-1}).

$A(\theta)$=The total alpha counting rate at θ minutes after the end of sampling.

θ=The time elapsed after the end of sampling (min).

$A(\theta_1, \theta_2)$=The number of counts from θ_1 to θ_2 minutes.

C_i=The atmospheric concentration of the i-th isotope (pCi/l).

E=The counting efficiency of radiation.

The build-up equation during sampling may be derived by considering the deposition, decay and growth from the parent for each isotope. The general form is

$$\frac{dN_i}{dt}=Q_iV+\lambda_{i-1}N_{i-1}-\lambda_iN_i\ (i=1,2,3) \quad (1)$$

Table 1. Characteristics of Radon and It's Daughters

Common name or symbol	Nuclide	Principal radiation (MeV) Alpha	Beta	Gamma (Av.)	Half life	Disintegration constant (min^{-1})	remarks
Radon	^{222}Rn	5.49		0.510	3.825 d	1.258×10^{-4}	Inert gas
Radium A	^{218}Po	6.00			3.05 m	2.273×10^{-1}	Chemically active
Radium B	^{214}Pb		0.67 1.03	0.295	26.8 m	2.586×10^{-2}	〃
Radim C	^{214}Bi		1.01, 1.51 3.26	1.050	19.7 m	3.519×10^{-2}	〃
Radium C′	^{214}Po	7.69			1.64×10^{-4}s	2.536×10^{5}	〃
Radium D	^{210}Pb		0.015 0.061		22 yr	5.994×10^{-8}	〃

Table 2. Daily Variation of Radon Daughters' Concentration*

Date	Radon daughters' concentration 10^{-2} pCi/l				Remarks
	RaA	RaB	RaC	Total	
11:00, Aug. 6	8.33	21.54	25.38	55.25	Cloudy
〃, Aug. 7	12.99	37.73	31.17	81.88	Cloudy
〃, Aug. 9	15.84	25.25	15.52	56.61	Cloudy after rain
〃, Aug. 10	12.24	21.28	20.25	53.78	Clear
〃, Aug. 11	32.99	35.54	17.82	86.35	Cloudy
〃, Aug. 12	74.80	46.20	24.37	145.37	Rainy
〃, Aug. 13	54.22	61.69	78.87	194.78	Rainy
〃, Aug. 14	6.64	22.23	42.66	71.53	Cloudy after rain
〃, Aug. 16	38.81	35.65	23.49	97.89	Clear
〃, Aug. 17	21.92	15.24	20.56	57.71	Clear
〃, Aug. 18	40.28	27.34	24.90	92.53	Clear
〃, Aug. 20	13.54	11.20	13.13	37.87	Cloudy
〃, Aug. 21	46.13	25.25	17.23	88.62	Cloudy
〃, Aug. 23	4.55	5.43	9.67	19.66	Clear after rain
〃, Aug. 24	25.00	25.94	9.57	60.52	Cloudy
〃, Aug. 25	26.59	20.59	13.44	61.33	Cloudy
〃, Aug. 26	22.79	25.38	12.81	60.98	Clear
〃, Aug. 27	12.43	23.12	22.99	58.54	Rain
〃, Aug. 28	35.14	29.11	14.86	79.10	Cloudy
〃, Avg. 31	81.00	31.44	16.30	128.74	Clear
〃, Sept. 1	88.34	52.96	34.14	175.44	Clear
〃, Sept. 2	103.65	77.02	40.31	220.98	Cloudy
〃, Sept. 3	17.84	53.42	45.69	116.95	Clear
Average	34.64	31.77	25.00	91.41	

*Measured from August 6 to September 3, 1976 at KAERI(N. Lat. 37°38′, E. Long. 127°15′), Seoul, Korea.

Here i=1,2 and 3 represent RaA, RaB and RaC respectively.

The decay of activity after the end of sampling is developed by taking account of the decay and the growth from the parent for each particular isotope. The decay equation of the i-th isotope after the end of sampling is then

$$\frac{dN_i}{d\theta}=\lambda_{i-1}N_{i-1}-\lambda_i N_i \quad (i=1,2,3) \qquad (2)$$

Integrating Eqs. (1) and (2), Ni and N_i^0 can be easily obtained[7]. The alpha counting rate $\dot{A}(\theta)$ at time θ after the end of sampling is directly proportional to the sum of the activity for RaA and RaC, viz.

$$\dot{A}(\theta)=E(\lambda_1 N_1+\lambda_3 N_3) \qquad (3)$$

The alpha counts from θ_1 to θ_2 will become

$$A(\theta_1,\theta_2)=\int_{\theta_1}^{\theta_2}\dot{A}(\theta)\,d\theta \qquad (4)$$

The solution of Eq. (4) is then

$$A(\theta_1,\theta_2)=E\{-1.0235N_1^0X-(4.2596N_1^0+3.7747N_2^0)Y+(3.2832N_1^0+2.7748N_2^0-N_3^0)Z\} \qquad (5)$$

with $X=e^{-\lambda_1\theta_2}-e^{-\lambda_1\theta_1}$

$Y=e^{-\lambda_2\theta_2}-e^{-\lambda_2\theta_1}$

$$Z=e^{-\lambda_3\theta_2}-e^{-\lambda_3\theta_1}$$

For the time intervals (2, 5), (6, 20) and (21, 30) min, Eq. (5) will be

$$A(2,5)=E(0.31656N_1^0+0.00853N_2^0+0.09337N_3^0) \quad (6)$$

$$A(6,20)=E(0.32490N_1^0+0.10799N_2^0+0.31493N_3^0) \quad (7)$$

$$A(21,30)=E(0.09576N_1^0+0.09564N_2^0+0.12965N_3^0) \quad (8)$$

Table 3. Hourly Variation of Radon Daughters' Concentration*

O'clock	Radon daughters' concentration 10^{-2}pci/l			
	RaA	RaB	RaC	Total
10:00	34.96	5.75	2.2	42.91
11:00	27.55	11.90	0.39	39.84
12:00	2.30	7.31	3.07	12.68
13:00	—	—	—	—
14:00	14.51	6.36	4.84	25.71
15:00	13.35	4.80	2.47	20.62
16:00	23.45	2.26	3.04	28.75
17:00	3.46	6.46	1.34	11.26
Average	17.09	6.41	2.47	25.97

*Measured on Sep. 13, 1976 at KAERI (N. Lat. 37°38', E. Long. 127°15'), Seoul, Korea.
-No reliable data were obtained.

Table 4. Hourly Variation of Radon Daughters' Concentration*

O'clock	Radon daughters' concentration 10^{-2}pci/l			
	RaA	RaB	RaC	Total
10:00	38.35	50.61	38.63	127.59
11:00	52.58	41.28	43.44	137.30
12:00	19.03	11.48	10.35	40.86
13:00	41.28	19.92	5.54	66.74
14:00	31.36	16.24	2.51	50.11
15:00	—	—	—	—
16:00	36.90	13.53	1.20	51.63
17:00	28.04	11.90	3.71	43.65
Average	35.35	23.55	15.04	73.94

*Measured on Oct 4, 1976 at KAERI (N. Lat. 37°38', E. Long. 127°15'), Seoul, Korea.
-No reliable data were obtained.

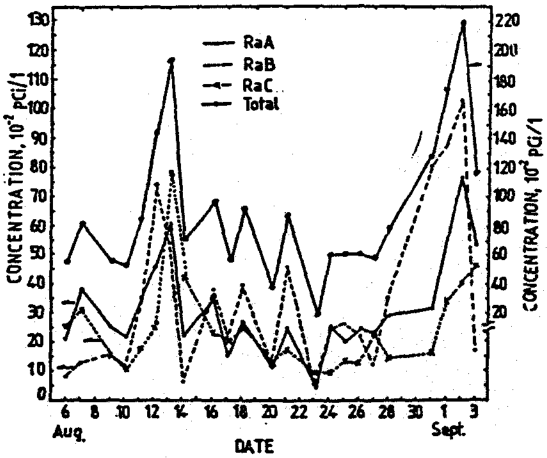

Fig 2. Daily Variation of Radon Daughterts' Concentration

For T=5 min, N_i^0 is

$$N_1^0=29.1860C_1V \quad (9)$$

$$N_2^0=18.7601C_1V+402.5764C_2V \quad (10)$$

$$N_3^0=0.8508C_1V+25.0853C_2V+289.2849C_3V \quad (11)$$

Having thus found the N_i^0 valus, we may now substitute in Eqs. (6), (7) and (8) to obtain the corresponding concentrations.

$$C_1=\frac{1}{VE}\{0.1689A(2,5)-0.0820A(6,20)+0.07753A(21,30)\} \quad (12)$$

$$C_2=\frac{1}{VE}\{0.00122A(2,5)-0.02057A(6,20)+0.04909A(21,30)\} \quad (13)$$

Table 5. Hourly Variation of Radon Daughters' Concentration*

O'clock	Radon daughters' concentration 10^{-2}pCi/l			
	RaA	RaB	RaC	Total
10:00	—	—	—	
11:00	16.39	17.45	7.66	41.50
12:00	1.84	10.38	13.00	25.22
13:00	—	—	—	
14:00	—	—	—	
15:00	—	—	—	
Average	9.12	13.92	10.33	33.37

*Measured on Oct. 11, 1976 at KAERI (N. Lat. 37°38', E. Long. 127°15'), Seoul, Korea.
-No reliable data were obtained.

Table 6. Hourly Variation of Radon Daughters' Concentration*

O'clock	Radon daughters' concentration 10^{-2}pCi/l			
	RaA	RaB	RaC	Total
10:00	28.93	50.99	35.63	114.55
11:00	—	—	—	
12:00	6.07	37.93	59.72	103.72
13:00	26.06	37.93	59.72	119.43
14:00	25.74	23.03	37.68	86.45
15:00	20.13	29.38	22.95	72.46
16:00	11.69	22.39	18.05	52.40
17:00	8.02	6.92	10.49	25.43
Average	18.95	30.53	33.55	82.06

*Measured on Oct. 18, 1976 at KAERI (N. Lat. 37°38', E. Long. 127°15'), Seoul, Korea.
-No reliable data were obtained.

Table 7. Hourly Variation of Radon Daughters' Concentration*

O'clock	Radon daughters' concentration 10^{-2}pCi/l			
	RaA	RaB	RaC	Total
10:00	2.33	12.96	6.5	21.79
11:00	—	—	—	
12:00	—	—	—	
13:00	—	—	—	
14:00	21.01	13.53	1.66	36.20
15:00	14.41	13.45	5.01	32.87
16:00	—	—	—	
17:00	21.29	9.57	3.32	34.18
Average	14.76	12.38	4.12	31.26

*Measured on Oct. 25, 1976 at KAERI (N. Lat. 37°38', E. Long. 127°15'), Seoul, Korea.
-No reliable data were obtained.

$$C_3=\frac{1}{VE}\Big\{-0.02252A(2,5)+0.03318A(6,20)-0.03771A(21,30)\Big\} \quad (14)$$

The inherent counting efficiency for alpha particle was theoretically calculated to be 96%[8].

Little data are known for the filtering efficiency. On this study it is therefore

Table 8. Hourly Variation of Radon Daughters' Concentration*

O'clock	Radon daughters' concentration 10^{-2}pCi/l			
	RaA	RaB	RaC	Total
10:00	21.79	7.73	23.06	52.58
11:00	21.97	19.49	33.34	74.80
12:00	23.48	16.85	41.14	81.47
13:00	43.83	12.75	13.17	69.75
14:00	—	—	—	
15:00	—	—	—	
Average	27.77	14.21	27.68	69.66

*Measured on Nov. 1, 1976 at KAERI (N. Lat. 37°38', E. Long. 127°15'), Seoul, Korea.
-No reliable data were obtained.

assumed to be 100%, but is expected to be reconsidered when the reliable data are available.

3. Experimental

The head of air sampler (Bendix, Low Volume Air Sampler) was placed at a height of 170 cm above the ground level of the KAERI site (N. Lat. 37°38' and E. Long 127°15'). The airborne dust samples were

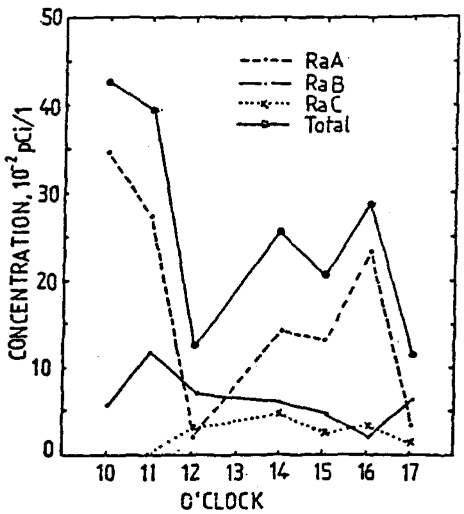

Fig. 3. Hourly Variation of Radon Daughters' Concentration (Measured on Sept. 13, 1976)

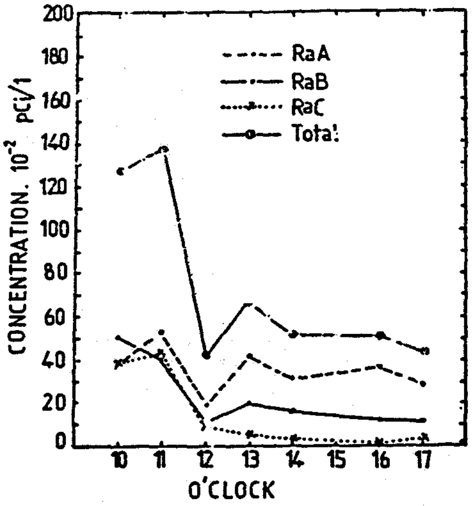

Fig. 4. Hourly Variation of Radon Daughters' Concentration (Measured on Oct. 4, 1976)

taken from a membrane filter paper of 25 mm in diameter and 0.8 μm in pore size.

The sampling time chosen for investigation was five minutes[9]. The total alpha activity was then measured using a 2π-internal proportional counter (Aloka, Model TDC-5) in three selected-time intervals, i.e., 2-5, 6-20 and 21-30 minutes, respectively.

4. Results and Discussion

In Table 2 and Fig. 2, the concentrations of radon daughters were measured at 11 o'clock every day for four weeks from August 6 through September 3, 1976. The standard deviation in terms of relative error associated with the numerical values given in Table 2 is ±24.7% to ±92.5% depending on counting results. Usually the uncertainty in RaA values was comparatively larger than that in both RaB and RaC, frequently leading to physically unacceptable results, say negative values. Comparatively large concentration was observed around August 13 and September 2. No clear account for this reason can be made yet. The minimum total activity was obtained on August 23 whereas the maximum value was on September 2. The latter is about ten times as high as the former. This may be a strong indication that the concentration of radon daughters is closely related to the meteoro-

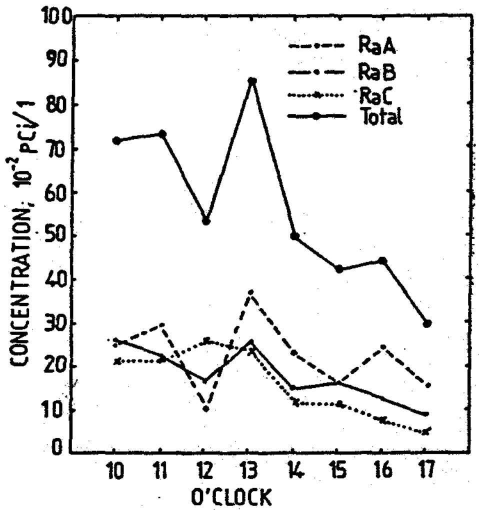

Fig. 5. Hourly Variation of Radon Daughters' Concentration (Averaged from measurements on every Monday from Sept. 13 to Nov. 1, 1976)

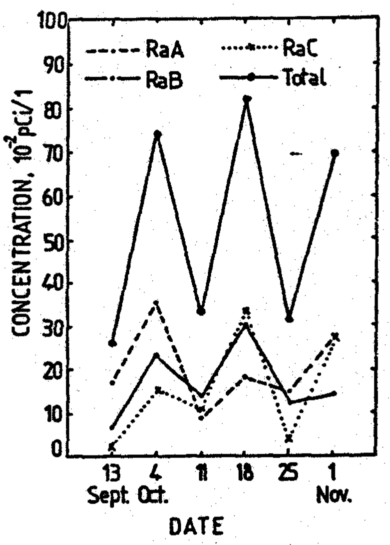

Fig. 6. Weekly Variation of Radon Daughters' Concentration

Table 9. Hourly Variation of Radon Daughters' Concentration*

O'clock	Radon daughters' concentration 10^{-2} pCi/l						
	RaA		RaB		RaC		Total
	Average	Max.	Average	Max.	Average	Max.	Average
10 : 00	25.07(5)	38.35	25.61(5)	50.99	21.20(5)	38.63	71.88(5)
11 : 00	29.62(4)	52.58	22.53(4)	41.28	21.21(4)	43.44	73.36(4)
12 : 00	10.54(5)	23.48	16.79(5)	37.93	25.46(5)	59.72	52.79(5)
13 : 00	37.06(3)	43.83	25.24(3)	43.05	23.01(3)	50.32	85.31(3)
14 : 00	23.16(4)	31.36	14.79(4)	23.03	11.67(4)	37.68	49.62(4)
15 : 00	15.96(3)	20.13	15.88(3)	29.38	10.14(3)	22.95	41.98(3)
16 : 00	24.01(3)	36.90	12.73(3)	22.39	7.43(3)	18.05	44.26(3)
17 : 00	15.20(4)	28.04	8.71(4)	11.9	4.72(4)	10.49	28.63(4)
Average	22.58(31)		17.78(31)		15.61(31)		55.98(31)

*Measured on every Monday from Sept. 13, 1976 to Nov. 1, 1976 at KAERI(N. Lat. 37°38′, E. Long 127°15′), Seoul, Korea.
( ) Number of measurements during six weeks.

Table 10. Weekly Variation of Radon Daughters' Concentration*

Date	Radon daughters' concentration 10^{-2} pCi/l						
	RaA		RaB		RaC		Total
	Average	Max.	Average	Max.	Average	Max.	Average
Sept. 13, 1976	17.09(7)	34.96	6.41(7)	11.90	2.47(7)	4.84	25.97(7)
Oct. 4, 1976	35.35(7)	52.58	23.55(7)	50.61	15.04(7)	43.44	73.94(7)
Oct. 11, 1976	9.12(2)	16.39	13.92(2)	17.45	10.33(2)	13.00	33.37(2)
Oct. 18, 1976	17.95(7)	27.93	30.53(7)	50.99	33.55(7)	59.72	82.06(7)
Oct. 25, 1976	14.76(4)	21.29	12.38(4)	13.53	4.12(4)	6.5	31.26(4)
Nov. 1, 1976	27.77(4)	43.83	14.21(4)	19.49	27.68(4)	33.34	69.66(4)
Average	20.34(31)		16.83(31)		15.53(31)		52.71(31)

*Measured from 10 a.m. to 5 p.m. at KAERI(N. Lat. 37°38′, E. Long. 127°15′), Seoul, Korea.
( ) Number of measurements a day.

logical condition.

An hourly variation of radon daughters' concentration was measured between 10 a.m. and 5 p.m., and every Monday from September 13 to November 1, 1976. The results are presented in Tables 3 to 8. Figs. 3 and 4 show the concentration values obtained on September 13 and October 4, respectively. In general, the highest value is observed in the morning that may coincide with the highest density of atmosphere in a daytime with a few exception. On the contrary, the lowest value is observed around fourteen o'clock.

Table 9 and Fig. 5 show an hourly variation of the daughters' concentration averaged from data in Tables 3 to 8. The daily averaged value is listed in the last row of this table. As might be expected, the parent concentration is larger than that of its daughters.

Table 10 and Fig. 6 indicate a weekly variation of the radon daughters' concentration measured on every Monday from September 13 through November 1, 1976. A sawtooth pattern in the concentration is noted, but it is rather too early to draw up any conclusion from the limited experimental data. Although the data obtained in this study are rather rough, these are reasonably agreeable with those by others[10), 11)].

5. Conclusion

An experimental determination of radon daughters' concentration in air has been performed using a single-filter method at the KAERI site.

A conclusion drawn up in this study is as follows:

1) A variation of radon daughters' concentration is closely related to meteorological conditions.
2) The parent concentration is comparatively higher than that of its daughters.
3) The highest concentration is normally observed in the morning whereas the lowest value is around fourteen.

6. References

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—抄　錄—

空氣浮遊塵內의 Radon 崩壞生成物의 濃度測定

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韓國核燃料開發公團

槪　要

空氣浮遊塵內에 存在하는 라돈 崩壞生成物 即 RaA, RaB 및 RaC의 簡單한 濃度測定法을 確立하였다. 이것은 單一集塵裝置로 membrane 濾過紙(平均 pore size =0.8 μm)에 採取한 試料의 全알파 放射能을 時差別로 測定하므로서 濃度를 決定하는 方法이다. 一聯의 豫備實驗으로써 이 方法을 利用하여 韓國原子力硏究所 構內에서 라돈 崩壞生成物의 時間的 濃度變化를 測定하였다.

이들 濃度의 時間的 變化는 甚하였는데 大槪의 境遇 最高値는 大氣의 密度가 比較的 높은 아침에 나타났고 最低値는 其 反對인 午後에 나타났다.