Measurement of
Radon Progeny Concentrations
in Air by Alpha-Particle Spectrometry

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ALPHA-PARTICLE SPECTROMETRY

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ABSTRACT

A technique is presented for measuring air concentrations of the short-lived progeny of radon-222 by the use of alpha-particle spectrometry. In this technique, the concentration of RaA, RaB, and RaC are calculated from one integral count of the RaA and two integral counts of the RaC' alpha-particle activity collected on a filter with an air sampling device. The influence of air sampling and counting intervals of time on the accuracy of the calculated concentrations is discussed in the report. A computer program is presented for use with this technique. It is written in the BASIC language. The program will calculate the air concentrations of RaA, RaB, and RaC, and will estimate the accuracy in these calculated concentrations.

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Introduction

The concentrations of short-lived progeny of radon attached to dust particles in the air can be measured by taking three counts of alpha-particle activity collected on a filter with an air sampling device. In a technique reported by Tsivoglou et al.[1], the concentrations of RaA, RaB, and RaC in air are calculated from three count rates of the combined alpha-particle activity of RaA and RaC' at 5, 15, and 30 min after the collection of an air sample and in modification of this technique reported by Thomas[2], they are calculated from three integral counts of the combined alpha-particle activity of RaA and RaC' from 2 to 5, 6 to 20, and 21 to 30 min after the termination of the air sample collection. An advantage of these two techniques is that a rather simple counter operated with air in the small gap between the detector and filter can be used, since the 6.0 and 7.7 MeV alpha-particle activity from RaA and RaC', respectively, does not have to be resolved; but these techniques do not offer the accuracy, especially in the calculated concentration of RaA, provided by a spectrometry technique reported by Martz et al.[3] In this spectrometry technique, the concentrations of the short-lived radon progeny are calculated from one count rate of the RaA and two count rates of the RaC' alpha-particle activity on the filter at 5 min and at 5 and 30 min, respectively, after the collection of the air sample.

In measurements using the alpha-particle spectrometry technique, the filter could be counted in a vacuum with a silicon surface-barrier detector, but to insert the filter either an elaborate counter with an air interlock would be required or the counter would have to be cycled

from the vacuum to atmospheric pressures. Because of potential breakdown of diode detectors at about 0.1 Torr, the bias voltage on the detector should also be cycled making the latter method of inserting the filter both complicated and time consuming. A counter which is more convenient for routine use with the spectrometry technique and a modification of this technique which improves the accuracy and sensitivity of radon daughter measurements have been developed at this laboratory.[4,5]

In this counter, helium is flowed between a diode detector and filter which are separated by a distance of about 0.5 cm. By using helium to replace the air in the small gap between the detector and filter, the counter can be operated at atmospheric pressures with little loss in resolution and considerable gain in filter handling simplicity compared to measurements in a vacuum.

In the modified spectrometry technique used with the counter, the concentrations of RaA, RaB, and RaC are determined from one integral count of the RaA and two integral counts of the RaC' alpha-particle activity collected on the filter. A computer program has been written in the BASIC language for use with this technique. This program will calculate the concentrations of RaA, RaB, and RaC in air and will estimate the accuracy in these calculated concentrations for the air sampling rate, the air sampling time, the counting times, and the detection efficiency of the counter used in counting the activity collected on the air filter. The matrix equations used in the computer program are discussed below. Sampling times and counting times are recommended which give a good overall accuracy in the calculated concentrations for a wide range of RaB/RaA and RaC/RaA activity ratios.

The computer program and an example of the printout of this program are

given in Appendices A and B, respectively.

Theory of the Modified Spectrometry Technique

In developing equations descriptive of the time dependence on the

collection and counting of radon progeny on a filter, the alpha particle

decay of RaC' occurs almost instantenously after the decay of RaC and

can be attributed to RaC.

The set of differential equations describing the rate at which radon

progeny atoms are collected on a filter with an air sampling device is

$$\frac{dn_i(t)}{dt} = q_i v + \lambda_{i-1} n_{i-1}(t) - \lambda_i n_i(t) \tag{1}$$

where
$$i = \begin{cases} 1 \text{ for RaA} \\ 2 \text{ for RaB} \\ 3 \text{ for RaC} \end{cases}$$

n_i = number of the ith type of atom on filter

λ_i = decay constant of ith type of atom in min.$^{-1}$

q_i = air concentration of ith type in atoms/liter

v = air sampling rate in liters/min

These linear differential equations are of the form

$$\frac{dy}{dx} + ay = f(x) \tag{2}$$

with a general solution

$$y = e^{-ax} \left[y_0 + \int f(x) e^{ax} dx \right] . \tag{3}$$

where y_0 is a constant of integration. In the solutions to equations(1), the constants of integration are evaluated using the initial conditions that each $n_i(t)$ is equal to zero at the start of the sampling interval of time, i.e. $n_i(t) = 0$ at $t = 0$. For a sampling interval of time t', ie. $t = t'$, measured in minutes, the number of radon progeny atoms which collect on the filter is

$$\text{RaA:} \quad n_1(t') = q_1 v \left[\frac{1 - e^{-\lambda_1 t'}}{\lambda_1} \right] \tag{4}$$

$$\text{RaB:} \quad n_2(t') = q_1 v \left[\frac{1 - e^{-\lambda_2 t'}}{\lambda_2} + \frac{e^{-\lambda_2 t'} - e^{-\lambda_1 t'}}{\lambda_2 - \lambda_1} \right] + q_2 v \left[\frac{1 - e^{-\lambda_2 t'}}{\lambda_2} \right] \tag{5}$$

$$\text{RaC:} \quad n_3(t') = q_1 v \left[\frac{1 - e^{-\lambda_3 t'}}{\lambda_3} + \frac{e^{-\lambda_3 t'} - e^{-\lambda_2 t'}}{\lambda_3 - \lambda_2} - \frac{2 e^{-\lambda_3 t'}}{(\lambda_3 - \lambda_2)(\lambda_3 - \lambda_1)} \right.$$

$$\left. + \frac{2 e^{-\lambda_2 t'}}{(\lambda_3 - \lambda_2)(\lambda_2 - \lambda_1)} - \frac{2 e^{-\lambda_1 t'}}{(\lambda_3 - \lambda_1)(\lambda_2 - \lambda_1)} \right]$$

$$+ q_2 v \left[\frac{1 - e^{-\lambda_3 t'}}{\lambda_3} + \frac{e^{-\lambda_3 t'} - e^{-\lambda_2 t'}}{\lambda_3 - \lambda_2} \right] + q_3 v \left[\frac{1 - e^{-\lambda_3 t'}}{\lambda_3} \right] \tag{6}$$

Equations (4), and (5), and (6) can be written matrix from as

$$N = v K Q \tag{7}$$

or

$$
\begin{vmatrix} n_1(t') \\ n_2(t') \\ n_3(t') \end{vmatrix} = v \begin{vmatrix} k_{11}(t') & k_{12}(t') & k_{13}(t') \\ k_{21}(t') & k_{22}(t') & k_{23}(t') \\ k_{31}(t') & k_{32}(t') & K_{33}(t') \end{vmatrix} \begin{vmatrix} q_1 \\ q_2 \\ q_3 \end{vmatrix} \tag{8}
$$

where capital letters are used in equation (7) to represent the matrices shown in equation (8). The elements of the matrix K are

$$k_{11}(t') = (1 - e^{-\lambda_1 t'})/\lambda_1$$

$$k_{12}(t') = 0$$

$$k_{13}(t') = 0$$

$$k_{21}(t') = (1 - e^{-\lambda_2 t'})/\lambda_2 + (e^{-\lambda_2 t'} - e^{-\lambda_1 t'})/(\lambda_2 - \lambda_1)$$

$$k_{22}(t') = (1 - e^{-\lambda_2 t'})/\lambda_2$$

$$k_{23}(t') = 0 \tag{9}$$

$$k_{31}(t') = (1 - e^{-\lambda_3 t'})/\lambda_3 + (e^{-\lambda_3 t'} - e^{-\lambda_2 t'})/(\lambda_3 - \lambda_2)$$

$$- \lambda_2 e^{-\lambda_3 t'}/(\lambda_3 - \lambda_2)(\lambda_3 - \lambda_1) + \lambda_2 e^{-\lambda_2 t'}(\lambda_3 - \lambda_2)(\lambda_2 - \lambda_1)$$

$$- \lambda_2 e^{-\lambda_1 t'}/(\lambda_3 - \lambda_1)(\lambda_2 - \lambda_1)$$

$$k_{32}(t') = (1 - e^{-\lambda_3 t'})/\lambda_3 + (e^{-\lambda_3 t'} - e^{-\lambda_2 t'})/(\lambda_3 - \lambda_2)$$

$$k_{33}(t') = (1 - e^{-\lambda_3 t'})/\lambda_3.$$

By letting v equal zero in equation (1), another set of differential equations is obtained which describe the decay of radon progeny atoms on

the filter once sample collection is terminated. These linear differential equations are also of the form given by equation (2) with a solution of the form specified by equation (3). In evaluating the constants of integration in the solutions of these equations, the time t is measured from the start of the counting interval of time, ie. t = 0 at the end of the sampling interval of time, and the number of radon progeny atoms on the filter at the start of the counting interval of time $n_i(t)$ is set equal to the number of atoms on the filter at the end of the sampling interval of time $n_i(t')$, ie. $n_i(t) = n_i(t')$ at t = 0.

At a time t measured in minutes from the start of the counting interval of time, the number of each type of radon progeny atoms on the filter is

RaA:
$$n_1(t) = n_1(t') \left[e^{-\lambda_1 t} \right] \tag{10}$$

RaB:
$$n_2(t) = n_1(t') \left[\frac{\lambda_1 e^{-\lambda_1 t}}{\lambda_2 - \lambda_1} - \frac{\lambda_1 e^{-\lambda_2 t}}{\lambda_2 - \lambda_1} \right] + n_2(t') \left[e^{-\lambda_2 t} \right] \tag{11}$$

RaC:
$$n_3(t) = n_1(t') \left[\frac{\lambda_1 \lambda_2 e^{-\lambda_1 t}}{(\lambda_3 - \lambda_1)(\lambda_2 - \lambda_1)} - \frac{\lambda_1 \lambda_2 e^{-\lambda_2 t}}{(\lambda_3 - \lambda_2)(\lambda_2 - \lambda_1)} \right.$$
$$\left. + \frac{\lambda_1 \lambda_2 e^{-\lambda_3 t}}{(\lambda_3 - \lambda_2)(\lambda_3 - \lambda_1)} \right] + n_2(t') \left[\frac{\lambda_2 e^{-\lambda_2 t}}{\lambda_3 - \lambda_2} - \frac{\lambda_2 e^{-\lambda_3 t}}{\lambda_3 - \lambda_2} \right]$$
$$+ n_3(t') \left[e^{-\lambda_3 t} \right] \tag{12}$$

Equations (10) and (12) can be used to relate an integral count c_j of alpha-particle activity on the filter in a counting interval of time designated as t_j to the number of atoms of each type of radon progeny on the filter at the termination of the sample collection by the relation

$$c_j(t_j) = g \left[\sum_i \int_{t_{js}}^{t_{je}} \lambda_i n_i(t) \, dt \right] \tag{13}$$

where t_{js} and t_{je} are the times at the start and end of the counting interval t_j, respectively, and g is the detection efficiency or the geometry factor of the counter. The sum in this relationship is over $i = 1$ and 3 for integral counts of the combined RaA and RaC activity, $i = 1$ for integral counts of only the RaA activity, and $i = 3$ for integral counts of only the RaC activity. Substitution of equations (10) and (12) into equation (13) and integration with respect to time an equation of the type

$$c_j (t_j) = g \left[l_{j1}(t_j)n_1(t') + l_{j2}(t_j)n_2(t') + l_{j3}(t_j)n_3(t') \right] \tag{14}$$

since the $n_i(t')$'s are independent of t. Three integral counts of the alpha-particle activity on the filter can, of course, be related to the number of radon progeny atoms of each type collected on the filter at the end of the sampling period or at the start of the counting period by a matrix of the form

$$
\begin{vmatrix} c_1(t_1) \\ \\ c_2(t_2) \\ \\ \\ c_3(t_3) \end{vmatrix} = g \begin{vmatrix} l_{11}(t_1) & l_{12}(t_1) & l_{13}(t_1) \\ \\ l_{21}(t_2) & l_{22}(t_2) & l_{23}(t_2) \\ \\ \\ l_{31}(t_3) & l_{32}(t_3) & l_{33}(t_3) \end{vmatrix} \begin{vmatrix} n_1(t') \\ \\ n_2(t') \\ \\ \\ n_3(t') \end{vmatrix}
\tag{15}
$$

or

$$
C = g \, L \, N \tag{16}
$$

where capital letters, as before, are used in equation (16) to represent the matrices shown in equation (15).

In this modification of the spectrometry technique, the three integral counts of alpha-particle activity are related to the number of atoms of each type of radon progeny on the filter at the start of the counting period by the equations:

$$
\text{RaA in } t_1: \quad c_1 = g \int_{t_{1s}}^{t_{1e}} \lambda_1 n_1(t) \, dt
$$

$$
\text{RaC in } t_2: \quad c_2 = g \int_{t_{2s}}^{t_{2e}} \lambda_3 n_3(t) \, dt
$$

and

$$
\text{RaC in } T_3: \quad c_3 = g \int_{t_{3s}}^{t_{3e}} \lambda_3 n_3(t) \, dt. \tag{17}
$$

The two time intervals t_1 and t_2 are selected to be equal in the measurements of activity on the filter, but this in not necessary. Elements of the matrix L for this technique obtained by substitution

of equations (17) and (12) into equations (17), integration with

respect to t, and comparison with equation (14) have the following forms:

$$l_{11}(t_1) = e^{-\lambda_1 t_{1s}} - e^{-\lambda_1 t_{1e}}$$

$$l_{12}(t_1) = 0$$

$$l_{13}(t_1) = 0$$

$$l_{21}(t_2) = \frac{\lambda_2 \lambda_3}{(\lambda_3 - \lambda_1)(\lambda_2 - \lambda_1)} (e^{-\lambda_1 t_{2s}} - e^{-\lambda_1 t_{2e}})$$

$$- \frac{\lambda_1 \lambda_2}{(\lambda_3 - \lambda_2)(\lambda_2 - \lambda_1)} (e^{-\lambda_2 t_{2s}} - e^{-\lambda_3 t_{2e}})$$

$$+ \frac{\lambda_1 \lambda_2}{(\lambda_3 - \lambda_2)(\lambda_3 - \lambda_1)} (e^{-\lambda_3 t_{2s}} - e^{-\lambda_3 t_{2e}})$$

$$l_{22}(t_2) = \frac{\lambda_3}{\lambda_3 - \lambda_2} (e^{-\lambda_2 t_{2s}} - e^{-\lambda_2 t_{2e}}) - \frac{\lambda_2}{\lambda_3 - \lambda_2} (e^{-\lambda_3 t_{2s}} - e^{-\lambda_3 t_{2e}})$$

$$l_{23}(t_2) = e^{-\lambda_3 t_{2s}} - e^{-\lambda_3 t_{2e}}$$

$$I_{31}(t_3) = \frac{\lambda_2 \lambda_3}{(\lambda_3 - \lambda_1)(\lambda_2 - \lambda_1)} (e^{-\lambda_1 t_{3s}} - e^{-\lambda_1 t_{3e}}) \tag{18}$$

$$- \frac{\lambda_1 \lambda_3}{(\lambda_3 - \lambda_2)(\lambda_2 - \lambda_1)} (e^{-\lambda_2 t_{3s}} - e^{-\lambda_2 t_{3e}})$$

$$+ \frac{\lambda_1 \lambda_2}{(\lambda_3 - \lambda_2)(\lambda_3 - \lambda_1)} (e^{-\lambda_3 t_{3s}} - e^{-\lambda_3 t_{3e}})$$

$$I_{32}(t_3) = \frac{\lambda_3}{\lambda_3 - \lambda_2} (e^{-\lambda_2 t_{3s}} - e^{-\lambda_2 t_{3e}}) - \frac{\lambda_2}{\lambda_3 - \lambda_2} (e^{-\lambda_3 t_{3s}} - e^{-\lambda_3 t_{3e}})$$

$$I_{33}(t_3) = e^{-\lambda_3 t_{3s}} - e^{-\lambda_3 t_{3e}}.$$

Equations (7) and (16) can be combined to obtain the matrix relation

$$C = gv \, L \, K \, Q \tag{19}$$

and inverted to give

$$Q = \frac{1}{gv} K^{-1} L^{-1} C = \frac{1}{gv} M \, C \tag{20}$$

which yields a set of equations relating the three integral counts of activity on the filter to the air concentration of the radon progeny. The equations of this set have the following forms:

RaA: $\quad q_1 = \frac{1}{gv} \left[m_{11}(t',t_1)c_1 + m_{12}(t',t_2)c_2 + m_{13}(t',t_3)c_3 \right]$

RaB: $q_2 = \left[\dfrac{1}{gv} m_{21}(t',t_1)c_1 + m_{22}(t',t_2)d_2 + m_{23}(t',t_3)c_3 \right]$ (21)

RaC: $q_3 = \left[\dfrac{1}{gv} m_{31}(t',t_1)c_1 + m_{32}(t',t_2)c_2 + m_{33}(t',t_3)c_3 \right]$

where the coefficients m_{ij} are the elements of the matrix M obtained by the relation

$$M = K^{-1} L^{-1}.$$ (22)

In the above relation, the matrices K^{-1} and L^{-1} are the inverse matrices of K and L, respectively.

Mathematical expressions can be obtained for m_{ij} from the equations given in the paper, but these are very complicated functions of the sampling and counting times. In our use of these equations, the coefficients m_{ij} which are elements of the matrix M are obtained by first calculating the magnitudes of the matrix elements of K and L given in equations (9) and (18) for the sampling times and counting times used in the measurements. Next, the matrices are inverted to obtain the magnitudes of the matrix elements of K^{-1} and L^{-1} which are then multiplied together to yield the matrix element m_{ij} of M. A computer program, RPCON4, has been written to perform these matrix operations, to calculate the air concentrations of the radon progeny, and to estimate the accuracy of the calculated concentrations. In this program, the accuracy of the calculated concentrations of the radon progeny are estimated by the equation

$$s^2_{q_i} = \frac{1}{(gv)^2} \left[\sum_{j=1}^{3} m^2_{ij} s^2_{c_j} + \left(\sum_{j=1}^{3} m^2_{ij} c^2_j \right) \left(\frac{s^2_v}{v^2} + \frac{s^2_g}{g^2} \right) \right]$$ (23)

where s_{q_i} = uncertainity in the calculated concentrations with
      $i = 1$ for RaA, 2 for RaB, and 3 for RaC,

  s_{c_j} = statistical uncertainity of one standard deviation in
      a count of the activity with $j = 1, 2,$ and 3,

  s_v = systematic uncertainity in the air sampling rate,

and s_g = systematic uncertainity in the detection efficiency of
      the counter.

A copy of this program and an example of the printout of the program are

given in Appendices A and B, respectively, of this report.

Discussion

The systematic uncertainities in the air sampling rate and the

detection efficiency, which must be estimated by the user of the pro-

gram, are difficult to evalute, but with care are usually in the range

of 2 to 5 percent. A statistical uncertainty of one standard deviation

in each of the three counts is used automatically in the program for

estimating the accuracy in the calculated concentrations. The statis-

tical uncertainties of the counts and, therefore, the accuracy in the

calculated concentrations are influenced by the sampling rate, the sam-

pling time, the detection efficiency of the counter, and the counting

times. Of these factors, the influence of the sampling and counting

times on the accuracy in the calculated concentrations, are the most

difficult to evaluate. In evaluating the influence of the sampling

and counting times, only the statistical uncertainties need be con-

sidered because the systematic uncertainties of the measurements are

not influenced by these factors. A starting time of 2 min for the

first counting interval was selected because this was the shortest

practicable time for transferring the filter with the very short 3.05

min half-life RaA activity from the air sampling device to the counter,
and an ending time of 30 min was selected because a counting period of
this length has become standard for techniques of this type.[1,2,3]
For sampling times of 5 to 15 min, one RaA counting interval from 2 to
12 min and two RaC' counting intervals from 2 to 12 and 15 to 30 min
after the termination of the air sample collection were found to give
a good overall accuracy for a wide range of RaB/RaA and RaC/RaA activity
ratios. Sampling intervals of time greater than 15 min were also
investigated, but were not found to offer any great improvement in the
accuracy of the technique. Coefficients m_{ij} for use in hand calculations
of the air concentrations of the radon progeny in air by equations (21)
and the accuracy in the calculated concentrations by equations (23)
are given in Table C1 of Appendix C. Three sets are given in this
appendix: one for a sampling time of 5 min, one for a sampling time
of 10 min, and another for a sampling time of 15 min. All of these
sets are for a RaA counting time of 2 to 12 min and RaC' counting
times of 2 to 12 and 15 to 30 min. In Table C2 of Appendix C, a
comparison is made of the estimated accuracy in the calculated con-
centrations for each of the above air sampling intervals of time.

In a similar modification of the spectrometry technique, Jonassen
and Hayes[6] recommended one RaA counting interval from 120 to 320 sec
(2 to 5.33 min) and two RaC' counting intervals from 120 to 320 sec
(2 to 5.33 min) and 1200 to 1600 sec (20 to 26.67 min). A comparison
of the accuracy of the modified spectrometry technique for these
counting times and those recommended for general use in this paper

are given in Table 1 for a 10 min sampling interval of time and for
several different RaB/RaA and RaC/RaA activity ratios. In this table,
the comparison is made in terms of fractional standard deviations with
units of $(gvr)^{-1/2}$ where r is the activity of RaA in pCi per liter of
air, v is the sampling rate in liters per min of air, and g is the de-
tection efficiency. A similar comparison of the accuracy of the mod-
ified spectrometry technique with that offered by other counting tech-
niques has been given by Jonassen and Hayes.

The fractional standard deviations of Table 1 give an estimate of
the overall accuracy of the calculated concentrations only for small
values of r where counting statistics are poor and dominate. For larger
values of r where counting statistics are good, the systematic uncer-
tainties in the air sampling rate and detection efficiency must, of
course, be considered in estimating the accuracy of the calculated
concentrations. It can be seen from the table that the counting intervals
of 2 to 12 and 15 to 30 min yield a greater accuracy that the counting
intervals of 120 to 320 sec (2 to 5.33 min) 1200 to 1600 sec (20 to
26.67 min).

By use of the shorter counting intervals, Jonassen and Hayes were,
however, able to suggest the use of two additional counts of the RaC'
activity from 480 to 680 sec (8 to 11.33 min) and 1800 to 2200 sec (30
to 36.67 Min) to improve the accuracy in the calculated concentrations
of RaB and RaC. These two additional counts of the RaC' activity and
the count of the RaA activity from 120 to 320 sec (2 to 5.33 min) are
used to calculate a second set of concentrations of RaB and RaC. By
averaging the two sets of calculated concentrations for RaB and RaC,
their standard deviations are slightly improved. The computer

Table 1. Comparison of the accuracy of the modified spectrometry technique for a sampling interval of 10 minutes and (A) two counting periods from 2 to 12 and 15 to 30 minutes and (B) two counting intervals from 120 to 320 and 1200 to 1600 seconds after the termination of the air sample collection. The fractional standard deviations in the table are given in terms of $(gvr)^{-1/2}$ where g is the detection effeciency, v is the air sampling rate in liter per minute, and r is the activity of RaA in pCi per liter of air.

RaB/RaA and RaC/RaA Activity Ratios	(A)			(B)		
	$\dfrac{S_{q_1}}{q_1}$	$\dfrac{S_{q_2}}{q_2}$	$\dfrac{S_{q_3}}{q_3}$	$\dfrac{S_{q_1}}{q_2}$	$\dfrac{S_{q_2}}{q_2}$	$\dfrac{S_{q_3}}{q_3}$
1 and 1	0.213	0.259	0.187	0.277	0.311	0.228
0.6 and 0.4	0.213	0.313	0.322	0.277	0.374	0.387
0.4 and 0.2	0.213	0.368	0.505	0.277	0.455	0.586

program, RPCON4, can be used with three different counting intervals, and therefore, can be used to analyze the second set of counting data on a filter, as well as the first set of counting data, in this method of improving the accuracy of the RaB and RaC concentrations.

With the two counting intervals of 2 to 12 and 15 to 30 min recommended for general use, it is possible to measure equilibrium concentrations of 1, 1, and 1 pCi/l of RaA, RaB, and RaC with relative standard deviations of about 15, 18, and 13 percent, respectively. This assumes an air sampling time of 10 min, an air sampling rate of 10 l/min, and a counter efficiency of 0.20. For these same typical sampling and counting conditions, disequilibrium concentrations of 1, 0.4, and 0.2 pCi/l of RaA, RaB, and RaC can be measured with relative standard deviations of about 15, 26, and 36 percent, respectively. If the air sampling time is increased to 15 min and the air sampling rate to 17 l/min, the disequilibrium concentrations of 1, 0.4, and 0.2 pCi/l of RaA, RaB, and RaC can be measured with relative standard deviations of about 11, 18, and 26 percent, respectively, and the equilibrium concentrations of 1, 1, and 1 pCi/l of RaA, RaB, and RaC can be measured with relative standard deviations of about 11, 12, and 10 percent, respectively.

The counter developed by Perdue et al.[4] has been used with this technique in studying the effects of sealants on radon emanation from concrete,[7] in making measurements of radon daughter concentrations in homes in the Grand Junction area of Colorado,[8] and in calibrations of radon daughter dosimeters.[9] The computer program, RPCON4, has been of great help in analyzing the data in these studies.

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APPENDIX A

A listing and summary of the computer program, RPCON4, is provided in this appendix. The computer program can be summarized as follows:

Line Numbers of Program	Function
100- 130	Dimension Statements
140- 520	Variable Input Data Supplied by User
530- 580	Fixed Input Data
590- 830	Calculation of Matrix Elements of K given by equations (9) in text
890	Inversion of the Matrix K
900-1220	Calculation of Matrix Elements of L given by equation (18) in text
1230	Inversion of the Matrix L
1240	Calculation of Matrix Elements of M by equation (22) of text.
1250-1260	Calculation of Matrix Elements of Q by equation (20) of text.
1270-1350	Estimation of the Accuracy of the Calculated Concentrations by equations (23) of text and calculation of concentrations in terms of activity
1360-1380	Calculation of Activity Ratios of the concentrations
1400-1500	Printout of Data

```
100 DIM A$(3), A(3), C(3), D(3), E(3), E1(6), E2(6), E3(6), F(3,2)
110 DIM G1(6), G2(6), H2(6), H3(6), I1(6), I2(6), I3(6), J(3), K(3,3)
120 DIM L(3,3), M(3,3), P(3), Q(3), R(3,3), S(3,3), T(6), U(3)
130 DIM V(3), Y(3), Z(3)
140 PRINT "SAMPLE DESCRIPTION";
150 INPUT A$
160 PRINT
170 PRINT "THE AIR SAMPLING TIME IN MIN IS ";
180 INPUT T
190 PRINT
200 PRINT "THE AIR SAMPLING RATE IN LITERS/MIN AND THE"
210 PRINT "       UNCERTAINTY IN THIS QUANTITY IN PERCENT"
220 PRINT "       ARE";
230 INPUT V, X
240 PRINT
250 PRINT "THE GEOMETRY FACTOR OF THE DETECTOR USED TO"
260 PRINT "       COUNT THE SAMPLE AND THE UNCERTAINTY IN"
270 PRINT "       THIS QUANTITY IN PERCENT ARE";
280 INPUT G, Y
290 PRINT
300 PRINT "INTEGRAL COUNTS OF THE INDICATED RADON DAUGHTER"
310 PRINT "ACTIVITY IN THE TIME INTERVALS I1, I2, AND I3:"
320 PRINT "       RA-A IN I1 =";
330 INPUT C(1)
340 PRINT "       RA-C IN I2 =";
350 INPUT C(2)
360 PRINT "       RA-C IN I3 =";
370 INPUT C(3)
380 FOR I = 1 TO 3
390 Z(I) = SQR(C(I))/C(I)
400 NEXT I
410 PRINT
420 PRINT "STARTING TIMES TS AND ENDING TIMES TE IN MIN"
430 PRINT "OF THE INDICATED COUNTING TIME INTERVALS ARE:"
440 PRINT "       TS, TE OF I1 =";
```

```
450 INPUT I(1), I(2)
460 PRINT "        IS, IE OF I2 =";
470 INPUT I(3), I(4)
480 PRINT "        IS, IE OF I3 =";
490 INPUT I(5), I(6)
500 PRINT
510 PRINT
520 PRINT
530 A$(1) = "RA-A"
540 A$(2) = "RA-B"
550 A$(3) = "RA-C"
560 D(1) = 2.272E-1
570 D(2) = 2.586E-2
580 D(3) = 3.518E-2
590 F(2,1) = D(2) - D(1)
600 F(3,1) = D(3) - D(1)
610 F(3,2) = D(3) - D(2)
620 IF (D(1)*I) < 50 GO TO 650
630 E1 = 0
640 GO TO 660
650 E1 = EXP(-D(1)*I)
660 IF (D(2)*I) < 50 GO TO 690
670 E2 = 0
680 GO TO 700
690 E2 = EXP(-D(2)*I)
700 IF (D(3)*I) < 50 GO TO 730
710 E3 = 0
720 GO TO 740
730 E3 = EXP(-D(3)*I)
740 G1 = (1 - E1)/D(1)
750 H1 = (1 - E2)/D(2)
760 H2 = (E2 - E1)/F(2,1)
770 I1 = (1 - E3)/D(3)
780 I2 = (E2 - E3)/F(3,2)
790 I3 = (D(2) * (((E2 - E3)/F(3,2)) - ((E1 - E3)/F(3,1))))/F(2,1)
```

```
800  K(1,1) = G1
810  K(1,2) = 0
820  K(1,3) = 0
830  K(2,1) = H1 + H2
840  K(2,2) = H1
850  K(2,3) = 0
860  K(3,1) = I1 - I2 + I3
870  K(3,2) = I1 - I2
880  K(3,3) = I1
890  MAT S = INV (K)
900  F2 = D(2) * D(3)
910  F3 = D(1) * D(2) * D(3)
920  FOR I = 1 TO 6
930  IF (D(1)*I(I)) < 50 GO TO 960
940  E1(I) = 0
950  GO TO 970
960  E1(I) = EXP(-D(1)*I(I))
970  IF (D(2)*I(I)) < 50 GO TO 1000
980  E2(I) = 0
990  GO TO 1010
1000 E2(I) = EXP(-D(2)*I(I))
1010 IF (D(3)*I(I)) < 50 GO TO 1040
1020 E3(I) = 0
1030 GO TO 1050
1040 E3(I) = EXP(-D(3)*I(I))
1050 G1(I) = E1(I)
1060 G3(I) = E3(I)
1070 H2(I) = (D(3)*E2(I))/F(3,2)
1080 H3(I) = (D(2)*E3(I))/F(3,2)
1090 I1(I) = (D(2)*D(3)*E1(I))/(F(3,1)*F(2,1))
1100 I2(I) = (D(1)*D(3)*E2(I))/(F(3,2)*F(2,1))
1110 I3(I) = (D(1)*D(2)*E3(I))/(F(3,2)*F(3,1))
1120 ON I GO TO 1220, 1130, 1220, 1180, 1220, 1180
1130 J = I/2
1140 L(J,1) = G1(I-1) -G1(I)
```

```
1150 L(J,2) = 0
1160 L(J,3) = 0
1170 GO TO 1220
1180 J = I/2
1190 L(J,1) = I1(I-1) -I1(I) -I2(I-1) +I2(I) +I3(I-1) -I3(I)
1200 L(J,2) = H2(I-1) -H2(I) -H3(I-1) +H3(I)
1210 L(J,3) = G3(I-1) -G3(I)
1220 NEXT I
1230 MAT R = INV (L)
1240 MAT M = S*R
1250 MAT Y = M*C
1260 MAT Q = (1/(G*V))*Y
1270 FOR I = 1 TO 3
1280 J(I) = ((Z(I)*C(I))/(V*G))†2
1290 NEXT I
1300 FOR I = 1 TO 3
1310 U(I) = SQR((M(I,1)†2)*J(1) + (M(I,2)†2)*J(2) + (M(I,3)†2)*J(3))
1320 V(I) = (U(I)*100)/Q(I)
1330 P(I) = SQR((V(I)†2) + (X†2) + (Y†2))
1340 A(I) = (Q(I) * Q(I))/2.22
1350 NEXT I
1360 E(1) = 1.00
1370 E(2) = (Q(2)*G(2))/(Q(1)*G(1))
1380 E(3) = (Q(3)*G(3))/(Q(1)*G(1))
1390 :                'LLLL          ##.##†††††      ##.##†††††     +-###.#%      ##.##
1400 PRINT "CONCENTRATIONS OF RADON DAUGHTER RADIONUCLIDES IN AIR"
1410 PRINT
1420 PRINT "      RADON DAUGHTER";"         CONCENTRATION IN AIR";
1430 PRINT "              FRACTION OF"
1440 PRINT "      RADIONUCLIDE";"    ATOMS/LITER";"  PCI/LITER";
1450 PRINT "   SIGMA";"       EQUIL. CONC."
1460 PRINT
1470 FOR I = 1 TO 3
1480 PRINT USING 1390, A$(I), Q(I), A(I), P(I), E(I)
1490 NEXT I
1500 END
```

APPENDIX B

An example of the printout of the computer program, RPCON4, is provided in this appendix.

SAMPLE DESCRIPTION ? "EXAMPLE"

THE AIR SAMPLING TIME IN MIN IS ? 10

THE AIR SAMPLING RATE IN LITERS/MIN AND THE
    UNCERTAINITY IN THIS QUANTITY IN PERCENT
    ARE ? 10, 2

THE GEOMETRY FACTOR OF THE DETECTOR USED TO
    COUNT THE SAMPLE AND THE UNCERTAINITY IN
    THIS QUANTITY IN PERCENT ARE ? 0.25, 5

INTEGRAL COUNTS OF THE INDICATED RADON DAUGHTER
ACTIVITY IN THE TIME INTERVALS T1, T2, AND T3:
    RA-A IN T1 = ? 55
    RA-C IN T2 = ? 254
    RA-C IN T3 = ? 371

STARTING TIMES TS AND ENDING TIMES TE IN MIN
OF THE INDICATED COUNTING TIME INTERVALS ARE:
    TS, TE OF T1 = ? 2, 12
    TS, TE OF T2 = ? 2, 12
    TS, TE OF T3 = ? 15, 30

CONCENTRATIONS OF RADON DAUGHTER RADIONUCLIDES IN AIR

RADON DAUGHTER RADIONUCLIDE	CONCENTRATION IN AIR ATOMS/LITER	PCI/LITER	SIGMA	FRACTION OF EQUIL. CONC.
RA-A	9.79E+00	1.00E+00	+- 14.5%	1.00
RA-B	5.15E+01	6.00E-01	+- 20.2%	0.60
RA-C	2.52E+01	4.00E-01	+- 21.4%	0.40

APPENDIX C

Matrix elements m_{ij} of the matrix M are given in Table C1 of this appendix. These can be used in hand calculations of the concentrations of RaA, RaB, and RaC and of the accuracy of the calculated concentrations by equations (21) and (23), respectively. The matrix elements are for one count of RaA activity, c_1, on the filter from 2 to 12 min and two counts of RaC' activity, c_2 and c_3, on the filter from 2 to 12 and 15 to 30 min, respectively, after the termination of the air sampling. Three sets of matrix elements are given in the table: a first set for a 5 min air sampling interval, a second set for a 10 min sampling interval, and a third set for 15 min sampling interval. Estimates of the accuracy in the calculated concentrations for each of these sampling times and for several different RaB/RaA and RaC/RaA activity ratios are given in Table C2. The fractional standard deviations of Table C2 are in terms of $(gvr)^{-1/2}$ where g is the detection efficiency, v is the sampling rate in 1/min and r is the RaA activity in the air in pCi/l.

Table C1. Matrix elements m_{ij} of M needed to calculate the concentra-
tions of RaA, RaB, and RaC and to estimate the accuracy of
the calculated concentrations by hand using equations (21)
and (23). These are for one count of RaA activity, c_1, from
2 to 12 minutes and two counts of RaC' activity, c_2 and c_3,
from 2 to 12 and 15 to 30 minutes, respectively, after the
termination of the air sampling. The air sampling interval
of time are given in the table.

Matrix Elements m_{ij} of M	Air Sampling Intervals of Time		
	5 min	10 min	15 min
m_{11}	0.5878	0.4449	0.4127
m_{12}	0	0	0
m_{13}	0	0	0
m_{21}	-0.6254	-0.4819	-0.4514
m_{22}	-1.7172	-0.9140	-0.6478
m_{23}	1.9622	1.0445	0.7402
m_{31}	0.05138	0.05120	0.05003
m_{32}	1.2147	0.7158	0.5547
m_{33}	-0.4865	-0.3276	-0.2792

Table C2. Accuracy of the modified spectrometry technique for sampling time intervals of 5, 10, and 15 minutes. The counting times are 2 to 12 minutes for RaA activity and 2 to 12 and 15 to 30 minutes for RaC' activity on the filter after the termination of the air sampling. Fractional standard deviations given in the table are in terms of $(gvr)^{-1/2}$ where g is the detection efficiency, v is the air sampling rate in liter per minute, and r is the activity of RaA in pCi per liter.

RaB/RaA and RaC/RaA Activity Ratios	Sampling Time Interval								
	5 min.			10 min.			15 min.		
	$\dfrac{s_{q_1}}{q_1}$	$\dfrac{s_{q_2}}{q_2}$	$\dfrac{s_{q_3}}{q_3}$	$\dfrac{s_{q_1}}{q_1}$	$\dfrac{s_{q_2}}{q_2}$	$\dfrac{s_{q_3}}{q_3}$	$\dfrac{s_{q_1}}{q_1}$	$\dfrac{s_{q_2}}{q_2}$	$\dfrac{s_{q_3}}{q_3}$
1 and 1	0.245	0.349	0.220	0.213	0.259	0.187	0.204	0.222	0.180
0.6 and 0.4	0.245	0.409	0.380	0.213	0.313	0.322	0.204	0.267	0.318
0.4 and 0.2	0.245	0.484	0.580	0.213	0.368	0.505	0.204	0.324	0.488