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MEASUREMENT OF AIRBORNE CONCENTRATIONS OF RADON-220 DAUGHTER PRODUCTS BY ALPHA-PARTICLE SPECTROMETRY

G. D. Kerr, M. T. Ryan, and P. T. Perdue

Health and Safety Research Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

DISCLAIMER

MEASUREMENT OF AIRBORNE CONCENTRATIONS OF RADON-220 DAUGHTER PRODUCTS BY ALPHA-PARTICLE SPECTROMETRY*

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ABSTRACT

The decay of naturally occurring uranium-238 and thorium-232 produces radon-222 and radon-220 isotopes which can escape into the atmosphere. If these radon gases become concentrated in air, their daughter products may present an inhalation hazard to man. The airborne concentrations of radon-222 can usually be measured very accurately in the presence of normal airborne concentrations of radon-220 and its daughters.

In contrast, the measurements of the airborne concentrations of radon-220 daughters are usually complicated by the presence of radon-222 and its daughters even at normally occurring airborne concentrations. The complications involved in these measurements can be overcome in most situations by using an alpha particle spectrometer to distinguish the activity of radon-222 daughters from that due to radon-220 daughters collected on a filter.

A practical spectrometer for field measurements of alpha particle activity on a filter is discussed. Also discussed are methods that can be used to make measurements of either radon-220 or radon-222 daughter concentrations in air, or simultaneous measurements of the airborne daughter concentrations of both radon isotopes.

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INTRODUCTION

Over a period of about five years we have made extensive measurements of radon-222 daughter concentrations in air using a modification of an alpha particle spectrometry technique suggested by Martz et al. (1). In our radon-222 daughter measurements, the concentrations of RaA, RaB, and RaC in air are calculated from one count of the 6.00 MeV alpha particle activity of RaA and two counts of the 7.69 MeV alpha particle activity of RaC' collected on a filter. Various aspects of these measurements and some results have been discussed elsewhere (2-5).

Recently we have used a modification of an alpha particle spectrometry method suggested by Duggan (6) to measure radon-220 daughter concentrations in air as well. It is impossible to resolve the activity on the filter due to the 6.05 and 6.09 MeV alpha particles from ThC and the 6.00 MeV alpha particles from RaA, but the activity can be partitioned between RaA and ThC using the 8.78 MeV alpha particle activity from ThC'. In our radon-220 daughter measurements, the concentrations of ThB and ThC in air are calculated from two counts of the 8.78 MeV alpha particle activity of ThC' collected on a filter.

METHOD OF MEASUREMENTS

Our alpha particle spectrometer shown in Fig. 1 utilizes a modified gas flow proportional counter to house a 450 mm^2 silicon diode (3). Normally this type of detector is used in a vacuum, rather than air, to improve the resolution of the measurements. The need for vacuum equipment in our measurements is eliminated by flowing helium through the chamber at atmospheric pressure. This provides considerable gain in resolution compared to counts of filter activity made in air and eliminates filter handling complications encountered when using a vacuum (2). It is not necessary, for example, to cycle the bias voltage on the diode because there are no changes in pressure. The resolution of our spectrometer for an air sample collected at a flow velocity of about 50 cm per sec with a membrane (Metricel, Gelman GN-6) and glass-fiber (Acropor, Gelman An-450) filter having a median pore size of 0.45 μ are compared with a more porous filter (Whatman grade 4) in Fig. 2. Either the membrane or the glass-fiber filter with a median pore size of 0.45 to 0.80 μ allows easy resolution of the 6.00 and 7.69 MeV alpha particles emitted by RaA and RaC', respectively. At flow velocities of up to 100 cm per sec, these filters are normally more than 99% efficient in collecting aerosols (7).

Figure 3 shows several counts of RaA, RaC', ThC, and ThC' alpha particle activity collected on a filter. Instead of count rates used by Martz et al. (1) and Duggan (6), we use counting intervals of 10 min or more to improve the accuracy and sensitivity of the measurements. Our counting intervals of time are always measured from the termination of air sample collection. Air sampling periods of 5 to 15 min are normally used in our measurements followed by a 2 to 12 min count of the 6.00, 7.69, and 8.78 MeV alpha particle activity from RaA, RaC', and ThC', respectively. Note that the 2 to 12 min count of the 6.00 MeV alpha particle activity from RaA in Fig. 3 contains a small contribution from the 6.05 and 6.09 MeV alpha particles from ThC. Alpha particles having these energies are emitted in about 36% of the ThC decays, while 8.78 MeV alpha particles are emitted in the remaining decays. Thus, the 8.78 MeV alpha particle count from 2 to 12 min can be multiplied by 36/64 or 0.56 to obtain the ThC contribution to the 2 to 12 min count of RaA activity on the filter.

A second RaC' count from 15 to 30 min plus the first 2 to 12 min counts of the RaA and the RaC' alpha particle activity on the filter and a computer program written in BASIC are used to obtain the airborne concentrations of the radon-222 daughters. This program, designated as RPCON4, will calculate the concentrations of RaA, RaB, and RaC in air and will estimate the accuracy in these calculated concentrations for variable air sampling rate, air sampling time, counting time, and detection efficiency of the alpha particle spectrometer used to measure the activity on the filter (4). A modification of this program, designated as THOR2, is used to obtain the airborne ThB and ThC daughter concentration of radon-220 from the first 2 to 12 min count of the 8.78 MeV alpha particle activity of ThC' plus a second ThC' count of 10 min or more starting at least 200 min after the termination of the air sample collection. The concentration of ThB daughters in air is determined primarily from the second count of ThC' activity on the filter, while the concentration of ThC daughters in air is determined primarily from the first count of ThC' activity on the filter. These two counts are, however, very complex mathematical functions of the air sampling time, the counting times, and the activity ratio of ThC:ThB daughters in air.

METHOD OF ANALYSIS

The THOR2 computer program is used to calculate the concentrations of ThB and ThC in air and to estimate the accuracy in these calculated concentrations as follows. First, the two counts, c_1 and c_2, of the 8.78 MeV alpha particle activity are related to the number of ThB and ThC atoms, n_1 and n_2 respectively, on a filter at the end of a sampling period by the matrix equation

$$C = 0.64 \; g \; L \; N \tag{1}$$

or

$$\begin{vmatrix} c_1 \\ \\ c_2 \end{vmatrix} = 0.64\ g \begin{vmatrix} l_{11} & l_{12} \\ \\ l_{21} & l_{22} \end{vmatrix} \begin{vmatrix} n_1 \\ \\ n_2 \end{vmatrix} , \qquad (2)$$

where capital letters are used in Eq. (1) to represent the matrices shown in Eq. (2). The elements l_{ij} of the matrix L are

$$l_{11} = \lambda_2(e^{-\lambda_1 t_{1s}} - e^{-\lambda_1 t_{1e}})/(\lambda_2 - \lambda_1) - \lambda_1(e^{-\lambda_2 t_{1s}} - e^{-\lambda_2 t_{1e}})/(\lambda_2 - \lambda_1) ,$$

$$l_{12} = e^{-\lambda_2 t_{1s}} - e^{-\lambda_2 t_{1e}} ,$$

$$l_{21} = \lambda_2(e^{-\lambda_1 t_{2s}} - e^{-\lambda_1 t_{2e}})/(\lambda_2 - \lambda_1) - \lambda_1(e^{-\lambda_2 t_{2s}} - e^{-\lambda_2 t_{2e}})/(\lambda_2 - \lambda_1) ,$$

$$l_{22} = e^{-\lambda_2 t_{2s}} - e^{-\lambda_2 t_{2e}} , \qquad (3)$$

where t_{1s} and t_{1e} are the starting and ending times of the first counting interval, t_{2s} and t_{2e} are the starting and ending times of the second counting interval, and λ_1 and λ_2 are the decay constants of ThB and ThC, respectively. In Eqs. (1) or (2), the factor 0.64 is the number of 8.78 MeV alpha particles emitted in the decay chain of either a ThB or ThC atom, and the factor g is the detection efficiency or geometry factor of the spectrometer used in measuring the 8.78 MeV alpha particle activity on a filter.

Next, a relationship between the number of ThB and ThC atoms on a filter at the end of a sampling period of time, t, and the concentrations of ThB and ThC atoms in air, q_1 and q_2, respectively, is established by the matrix equation

$$N = v\ K\ Q \tag{4}$$

or

$$\begin{vmatrix} n_1 \\ \\ n_2 \end{vmatrix} = v \begin{vmatrix} k_{11} & k_{12} \\ \\ k_{21} & k_{22} \end{vmatrix} \begin{vmatrix} q_1 \\ \\ q_2 \end{vmatrix} , \tag{5}$$

where

$$k_{11} = (1 - e^{-\lambda_1 t})/\lambda_1 ,$$

$$k_{12} = 0 ,$$

$$k_{21} = (1 - e^{-\lambda_2 t})/\lambda_2 + (e^{-\lambda_2 t} - e^{-\lambda_1 t})/(\lambda_2 - \lambda_1) ,$$

$$k_{22} = (1 - e^{-\lambda_2 t})/\lambda_2 , \tag{6}$$

and the factor v is the air sampling rate used in the measurements. Equations (1) and (4) can be combined to obtain

$$C = 0.64\ gv\ L\ K\ Q \tag{7}$$

and inverted to yield

$$Q = \frac{1}{0.64\ gv}\ K^{-1}\ L^{-1}\ C = \frac{1}{0.64\ gv}\ M\ C . \tag{8}$$

The computer program THOR2 calculates the matrices K and L, inverts these to obtain the matrices K^{-1} and L^{-1}, and multiplies these together to obtain the matrix M for the sampling and counting times of the measurements.

Finally, the elements m_{ij} of the matrix M are used to calculate the concentrations of ThB and ThC in air by Eq. (8) and to estimate the accuracy in these calculated concentrations by

$$\frac{s_{q_i}^{\;2}}{q_i^{\;2}} = \frac{\sum_j m_{ij}^{\;2} s_{c_j}^{\;2}}{\left(\sum_j m_{ij} c_j\right)^2} + \frac{s_v^{\;2}}{v^2} + \frac{s_g^{\;2}}{g^2} \quad , \tag{9}$$

where s_{c_j} = the standard deviation of the two counts c_1 and c_2,

s_v = the standard deviation of the air sampling rate,

s_g = the standard deviation of the detection efficiency,

s_{q_i} = the standard deviation of the calculated concentrations q_1 and q_2 of ThB and ThC, respectively.

The THOR2 program, which is available on request, automatically uses standard deviations of $(c_1)^{1/2}$ and $(c_2)^{1/2}$ for the two counts of the 8.78 MeV alpha particle activity on the filter. Table 1 shows an example of the data input and output of this program.

DISCUSSION

An example of the accuracy and sensitivity of our radon-220 daughter measurements in an area with a ventilation rate of about two air changes per hour is given in Table 2. These data are for a spectrometer with a detection efficiency of 0.25 and an air sampling rate of 12 l/min. The air sampling time is 15 min followed by 2 to 12 min and 200 to 220 min counts of the 8.78 MeV alpha particle activity from ThC' on the filter. Estimates in Table 2 of the standard deviation in the calculated concentrations of ThB and ThC in air are based only on the uncertainties in the two counts of the ThC' activity on the filter. These uncertainties dominate the accuracy of the measurements at very low concentrations of ThB and ThC in air, while the accuracy of the measurements at very high concentrations is determined mainly by the uncertainties in the air sampling rate and detection efficiency of the spectrometer. The uncertainties in the air sampling rate and the detection efficiency are difficult to evaluate but with care are usually in the range of 2 to 5%. While our measurements provide information only on the ThB and ThC concentrations in air, they can be used in some instances to obtain additional information on the ventilation rate and thereby the airborne concentrations of ThA and radon-220 (6).

The methods discussed here have been used to make simultaneous measurements of the airborne concentrations of radon-222 and radon-220 daughters in areas contaminated with thorium-232 and in several buildings contaminated with raffinates from the processing of thorium ore. Recently, airborne concentrations of radon-219 daughters as high as, or higher than, the normally more abundant airborne concentrations of radon-220 and radon-222 have been measured in a building contaminated with raffinates from uranium ore processing. A method developed for the simultaneous measurement of the airborne daughter concentrations of all three radon isotopes will be discussed elsewhere (8).

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Table 1. Example of data input and output of THOR2 computer program

```
THOR2


SAMPLE DESCRIPTION ?  EXAMPLE

THE AIR SAMPLING TIME IN MIN IS ?  15

THE AIR SAMPLING RATE IN LITERS/MIN AND THE
     UNCERTAINTY IN THIS QUANTITY IN PERCENT
     ARE ?  12, 2

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

COUNTS OF 8.8 MEV ALPHA ACTIVITY FROM TH-C'
     IN THE TIME INTERVALS T1 AND T2:
     COUNTS IN T1= ?  262
     COUNTS IN T2= ?  1048

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= ?  200, 220


     CONCENTRATIONS OF THORON DAUGHTER RADIONUCLIDES IN AIR


     THORON DAUGHTER        CONCENTRATION IN AIR
     RADIONUCLIDE     ATOMS/LITER    PCI/LITER        SIGMA

         TH-B         2.08E+03       1.02E+00    +-    6.3%
         TH-C         5.88E+01       3.03E-01    +-   11.6%
```

Table 2. Example of the accuracy and sensitivity of radon-220 measurements in an area with a ventilation rate of about two air changes per hour[a]

Radon-220 daughter concentrations in air in pCi l^{-1}		Standard deviation in radon-220 daughter measurements in percent[b]	
ThB	ThC	ThB	ThC
10.0	3.0	1.0	3.2
3.0	1.0	1.9	5.9
1.0	0.3	3.3	10.2
0.3	0.1	6.0	18.5
0.1	0.03	10.3	32.6
0.03	0.01	19.0	57.3

[a]These data are for a spectrometer with a detection efficiency of 0.25 and an air sampling rate of 12 liters per minute. The air sampling time is 15 min followed by a 2 to 12 min and a 200 to 220 min count of the 8.78 MeV alpha particle activity from ThC' on the filter.

[b]These standard deviations are based only on the standard deviations in the counts of alpha particle activity on the filter.

LIST OF FIGURES

PHOTO 2910-72

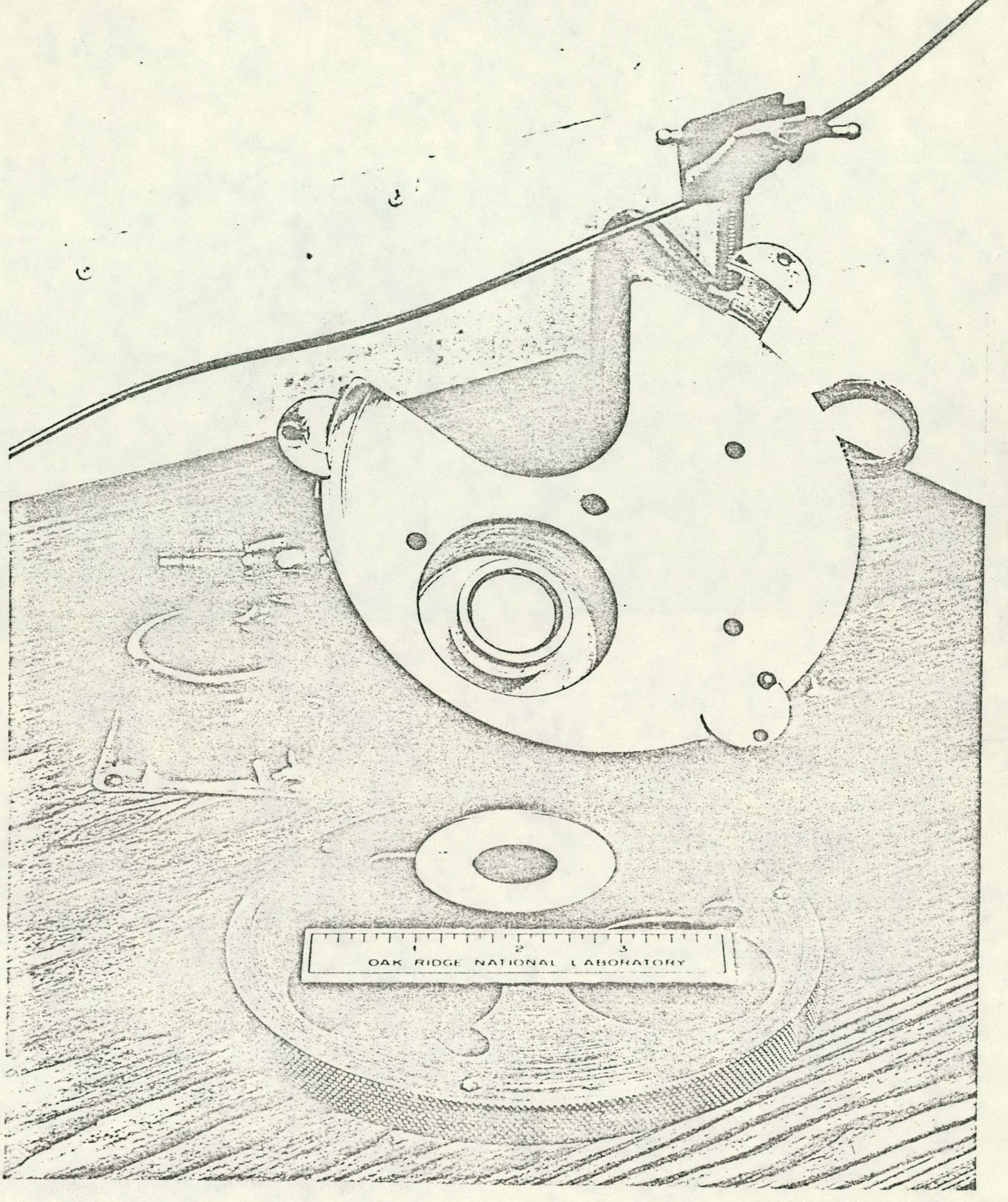

Fig. 1

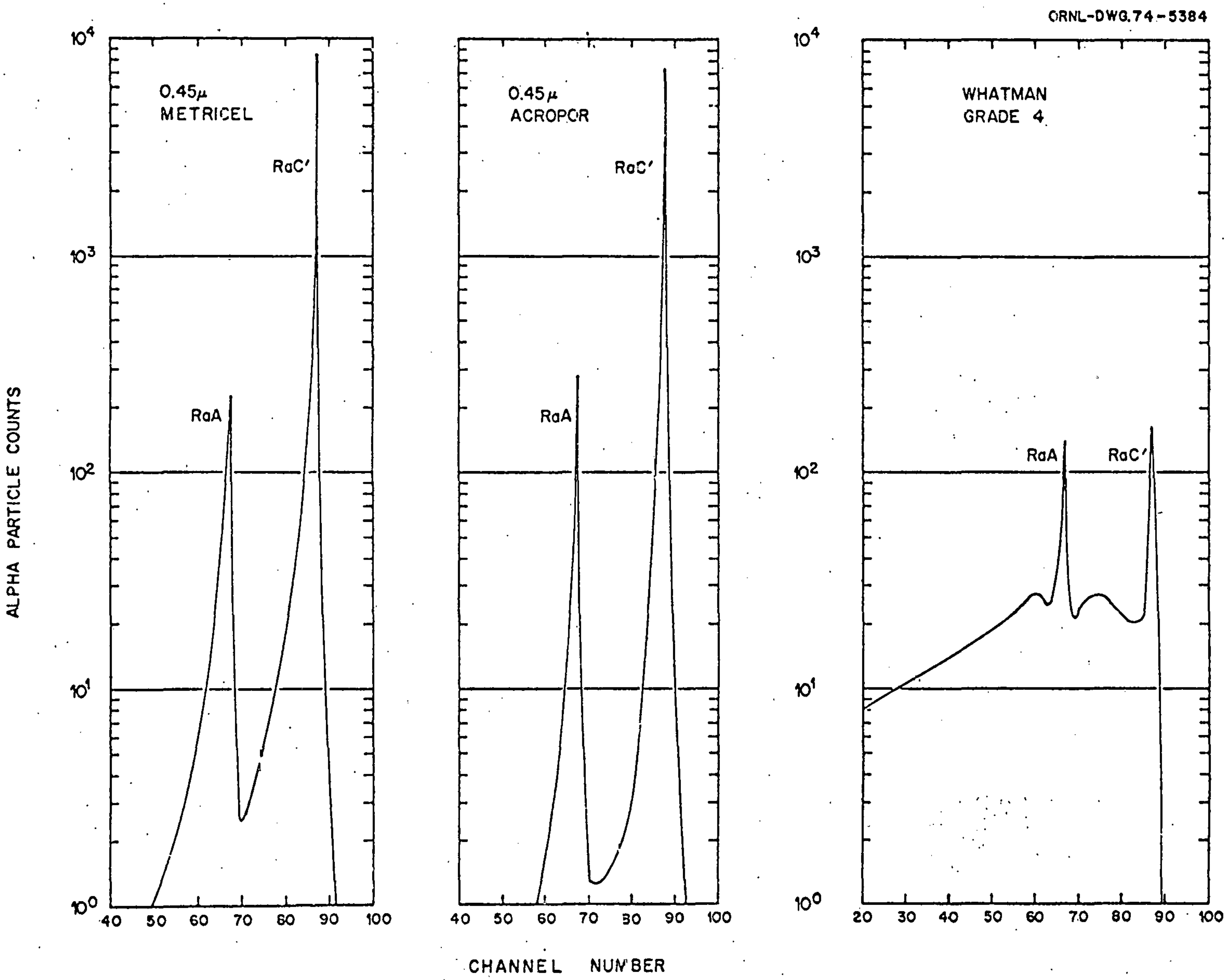
ORNL-DWG.74-5384
ALPHA PARTICLE COUNTS
0.45μ
METRICEL
RaC′
RaA
0.45μ
ACROPOR
RaC′
RaA
WHATMAN
GRADE 4
RaA
RaC′
10⁰
10¹
10²
10³
10⁴
40 50 60 70 80 90 100
40 50 60 70 80 90 100
20 30 40 50 60 70 80 90 100
CHANNEL NUMBER

Fig. 2

ORNL-DWG 77-21199

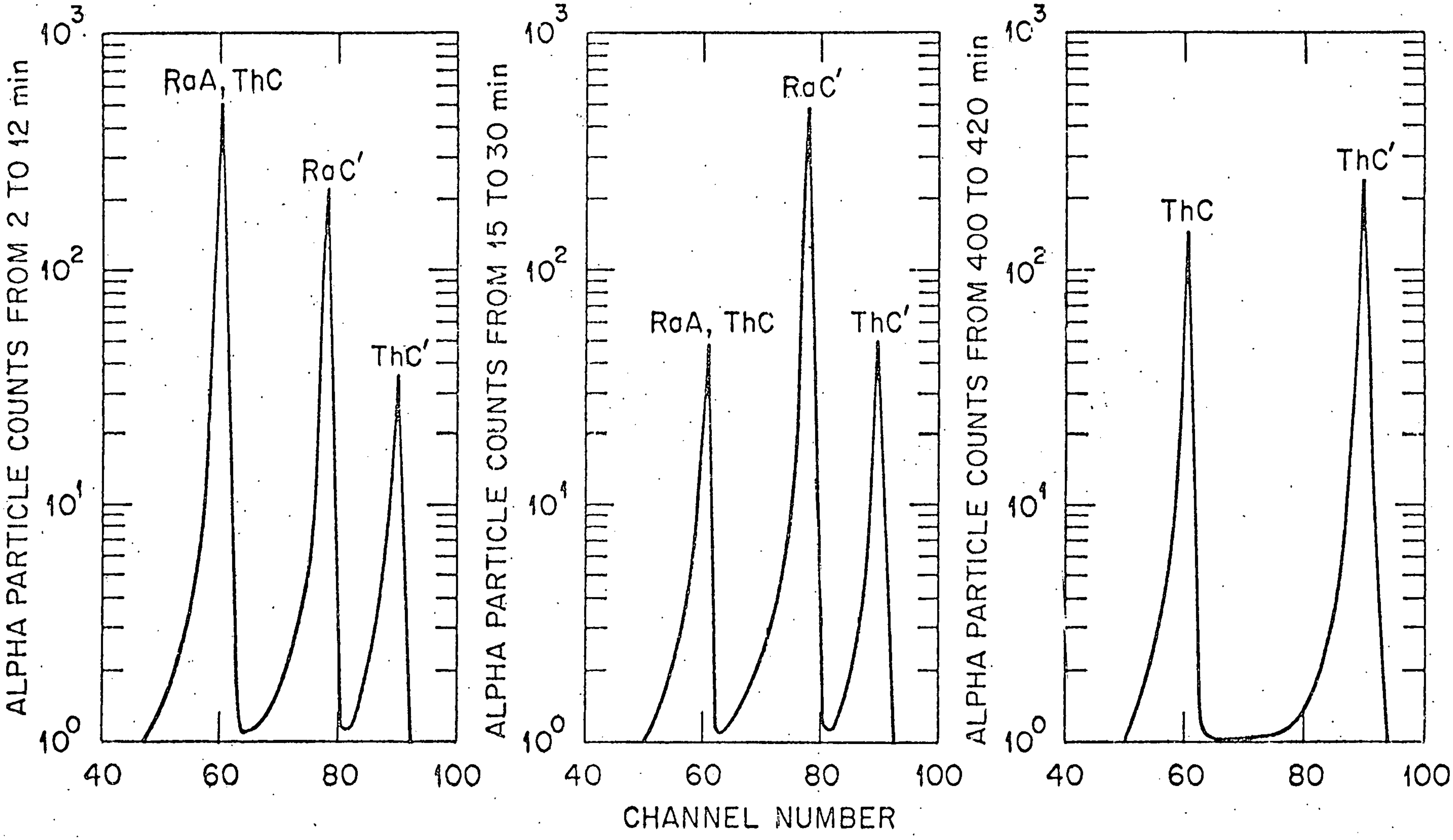

Fig. 3