

ANALYSIS OF ATMOSPHERIC CONCENTRATIONS OF RaA, RaB AND RaC BY ALPHA SPECTROSCOPY*

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Abstract—A new method is presented for determining the airborne concentrations of RaA, RaB and RaC in atmospheres contaminated with radon-222. The method employs alpha spectroscopy to measure the count rates of RaA and RaC' present on a membrane filter sample at two post-sampling times. The individual air concentrations and the statistical variances associated with each then may be calculated from the equations given. Theoretical and experimental comparisons are presented which indicate the improved accuracy of the spectroscopic method over methods previously available.

1. INTRODUCTION

THE IMMEDIATE source of airborne radioactivity in uranium mines is radon gas, although, due to a combination of physical and biological factors, radon itself does not contribute significantly to the hazardous radiation dose. Radon-222, a naturally occurring member of the uranium decay series, is present throughout the biosphere in small concentrations due to the wide distribution of trace quantities of natural uranium. A large fraction of the radon that is formed within crystalline rocks decays harmlessly before it can escape by diffusion processes. However, once radon has reached a crystal boundary it may migrate more readily along interstitial spaces until it is finally released to the atmosphere. In porous rock or soil, radon can move rapidly by diffusion or may be carried by ground water to the surface.

In poorly ventilated mines containing even low grade uranium deposits, radon concentrations can reach relatively high values. Measurements in working uranium mines over a period of several years⁽¹⁾ have indicated that maximum concentrations in poorly ventilated areas can exceed 25,000 pCi/l., although dilution ventilation techniques properly employed in recent years have reduced the average concentration in most working areas to a few hundred pCi/l.

The relatively short half-lives of the first four radon decay products are less than, or at most comparable to, typical ventilation turn-over times for mine air. Hence there is sufficient time for the radioactive build-up of significant concentrations of these daughter products in an atmosphere where radon is present. Conversely, the long half-life of RaD prevents this isotope and all subsequent progeny from achieving an appreciable build-up in mine atmospheres.

Field methods, based on gross alpha counting of filter samples, have been developed for determining the amount of radon daughter activity present in radon contaminated air.^(2,3) A method due to Tsiroglou *et al.*,⁽⁴⁾ has been used to estimate the individual air concentrations of the three short lived radon daughters, RaA, RaB and RaC. This paper discusses a new method for determining the individual concentrations of the above radon daughters in air, based on the use of alpha spectroscopy. Conventional air sampling techniques employing membrane or glass fiber filters are assumed. A necessary requirement is the retention of the alpha activity on a thin surface layer of the filter for adequate resolution of the RaA and RaC' alpha peaks.

2. THEORETICAL CONSIDERATIONS

During sampling at a flow rate V , the radon daughter atoms collected on a membrane filter

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obey time relations obtained from the following set of differential equations:

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

where

N_i = number of atoms of the i^{th} isotope on the filter

λ_i = decay constant of the i^{th} isotope in min^{-1}

Q_i = air concentration of the i^{th} isotope in atoms/l.

V = sampling flow rate in l./min

1-RaA

i = 2-RaB

3-RaC

The solution of this set of differential equations is straightforward, and when the integration constants are evaluated in terms of the initial condition that at $t = 0$, $N_i = 0$, leads to the following equations which hold only during the sampling period.

$$\text{(RaA)} \quad N_1 = \frac{Q_1V}{\lambda_1} (1 - e^{-\lambda_1 t}) \quad (1)$$

$$\begin{aligned} \text{(RaB)} \quad N_2 = & \frac{(Q_1 + Q_2)V}{\lambda_2} (1 - e^{-\lambda_2 t}) \\ & + \frac{Q_1V}{\lambda_2 - \lambda_1} (e^{-\lambda_2 t} - e^{-\lambda_1 t}) \quad (2) \end{aligned}$$

$$\begin{aligned} \text{(RaC)} \quad N_3 = & \frac{(Q_1 + Q_2 + Q_3)V}{\lambda_3} (1 - e^{-\lambda_3 t}) \\ & + \frac{(Q_1 + Q_2)V}{\lambda_3 - \lambda_2} (e^{-\lambda_3 t} - e^{-\lambda_2 t}) \\ & + \frac{Q_1\lambda_2V}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} (e^{-\lambda_3 t} - e^{-\lambda_1 t}) \\ & + \frac{Q_1\lambda_2V}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} (e^{-\lambda_3 t} - e^{-\lambda_2 t}). \quad (3) \end{aligned}$$

If the atmosphere is sampled for a time period T , the numbers of atoms for each daughter at the termination of sampling, $N_1(T)$, $N_2(T)$ and $N_3(T)$, are given by equations (1), (2) and (3) with t replaced by T .

For the time period after the sampling is terminated, a new time coordinate is defined with $t = 0$ at the end of sampling. The set of differential equations obeyed during the post sampling period are similar to the previous set,

except that $V = 0$. The solutions, when the integration constants are evaluated in terms of $N_i = N_i(T)$ at $t = 0$, become for the post sampling period:

$$\begin{aligned} \text{(RaA)} \quad N_1 = & N_1(T)e^{-\lambda_1 t} = \frac{Q_1V}{\lambda_1} \\ & \times (1 - e^{-\lambda_1 T})e^{-\lambda_1 t} \quad (4) \end{aligned}$$

$$\begin{aligned} \text{(RaB)} \quad N_2 = & \frac{\lambda_1 N_1(T)}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \\ & + N_2(T)e^{-\lambda_2 t} \quad (5) \end{aligned}$$

$$\begin{aligned} \text{(RaC)} \quad N_3 = & \frac{\lambda_1 \lambda_2 N_1(T)}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} \\ & \times (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \\ & - \left[\frac{\lambda_1 \lambda_2 N_1(T)}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} - \frac{\lambda_2 N_2(T)}{(\lambda_3 - \lambda_2)} \right] \\ & \times (e^{-\lambda_2 t} - e^{-\lambda_3 t}) + N_3(T)e^{-\lambda_3 t} \quad (6) \end{aligned}$$

Since the half-life of RaC' is quite short (1.60×10^{-4} sec), the decay of each RaC atom is followed practically instantaneously by the alpha decay of RaC', hence for the mathematical development the RaC' alpha emission may be treated as if it originated in the decay of RaC. The alpha activities of the two alpha emitting radon daughters, RaA and RaC', are obtained by multiplying the appropriate N_i by λ_i , treating RaC as if it was responsible for the RaC' alpha emission.

In order to determine the separate air concentrations of the radon daughters originally present in the air from which the sample was collected, the alpha activity present on the filter at definite post sampling times must be determined. The Tsivoglou method measures the gross alpha activity at three different times, usually five, fifteen and thirty minutes after the termination of sampling, and solves for the three unknowns Q_1 , Q_2 and Q_3 by matrix algebra. The spectroscopic method uses an alpha spectrometer to determine the RaA and RaC' activities separately at two post sampling times, and solves for the original air concentrations by matrix algebra involving only two equations and two unknowns.

3. SPECTROSCOPIC METHOD

The alpha activities at the two counting times, t_1 , and t_2 , are related to the original air

concentrations by the following equations:

$$\text{Activity of RaA at time } t_1 = A_1(t_1) = \lambda_1 N_1(t_1) \quad (7)$$

$$\text{Activity of RaC' at time } t_1 = A_3(t_1) = \lambda_3 N_3(t_1) \quad (8)$$

$$\text{Activity of RaC' at time } t_2 = A_3(t_2) = \lambda_3 N_3(t_2) \quad (9)$$

It is desired not only to determine the air concentrations of each daughter in the sampled atmosphere, but to provide an estimate of the standard deviation associated with these quantities. The primary sources of error involved in the exact determination of Q_1 , Q_2 and Q_3 are the variances associated with the alpha counting statistics, the flow rate and the counting yield. The component of variance attributed to counting yield contains geometrical contributions due to the sample filter and detector arrangement, as well as statistical errors involved in instrument reading and calibration. Each of these experimentally determined quantities and their associated standard deviations are written as:

$$\text{Alpha count rate} = C_i(t) \pm \alpha_i(t)$$

$$\text{Flow rate} = V \pm v$$

$$\text{Detector efficiency} = Y \pm y$$

$$\text{Alpha activity} = \frac{C_i(t_i) + \alpha_i(t_i)}{Y \pm y}$$

Using this notation we can rewrite equation (7) replacing $N_1(t_1)$ by equation (4), to give:

$$\begin{aligned} \frac{A_1(t_1)}{V} &= \frac{C_1(t_1) \pm \alpha_1(t_1)}{(Y \pm y)(V \pm v)} \\ &= Q_1(1 - e^{-\lambda_1 T})e^{-\lambda_1 t_1} \quad (10) \end{aligned}$$

Solving for Q_1 and its associated standard deviation σ_1 , combining the standard deviations by regular statistical procedures, gives

$$\begin{aligned} Q_1 \pm \sigma_1 &= \frac{C_1(t_1)e^{\lambda_1 t_1}}{YV(1 - e^{-\lambda_1 T})} \\ &\pm \frac{C_1(t_1)e^{\lambda_1 t_1} \left[\frac{\alpha_1^2(t_1)}{C_1^2(t_1)} + \frac{y^2}{Y^2} + \frac{v^2}{V^2} \right]^{1/2}}{YV(1 - e^{-\lambda_1 T})} \quad (11) \end{aligned}$$

Once $Q_1 \pm \sigma_1$ has been determined, a set of two algebraic equations in the two remaining

unknown Q_2 and Q_3 can be written from equations (8) and (9) with the appropriate replacements from equations (1), (2), (3), (5) and (6) giving:

$$\begin{aligned} \frac{C_3(t_1) \pm \alpha_3(t_1)}{(Y \pm y)(V \pm v)} - K_1(Q_1 \pm \sigma_1) &= K_3 Q_2 \\ &+ K_5 Q_3 \quad (12) \end{aligned}$$

$$\begin{aligned} \frac{C_3(t_2) \pm \alpha_3(t_2)}{(Y \pm y)(V \pm v)} - K_2(Q_1 \pm \sigma_1) &= K_4 Q_2 \\ &+ K_6 Q_3 \quad (13) \end{aligned}$$

where the constants K_i result from grouping the coefficients belonging to the individual Q_i . Solving equations (12) and (13) by matrix algebra, again combining the standard deviations by the statistical methods appropriate for simultaneous linear equations, gives

$$\begin{aligned} Q_2 \pm \sigma_2 &= \frac{K_6 \left[\frac{C_3(t_1)}{YV} - K_1 Q_1 \right] - K_5 \left[\frac{C_3(t_2)}{YV} - K_2 Q_1 \right]}{K_3 K_6 - K_4 K_5} \\ &\pm \left\{ \frac{K_6^2 \left[\frac{C_3^2(t_1)}{Y^2 V^2} \left(\frac{\alpha_3^2(t_1)}{C_3^2(t_1)} + \frac{y^2}{Y^2} + \frac{v^2}{V^2} \right) \right. \right.}{+ K_1^2 \sigma_1^2} + K_5^2 \left[\frac{C_3^2(t_2)}{Y^2 V^2} \left(\frac{\alpha_3^2(t_2)}{C_3^2(t_2)} \right. \right. \\ &\left. \left. + \frac{y^2}{Y^2} + \frac{v^2}{V^2} \right) + K_2^2 \sigma_1^2 \right] \right\}^{1/2} \\ &\pm \frac{K_3 K_6 - K_4 K_5}{K_3 K_6 - K_4 K_5} \quad (14) \end{aligned}$$

$$\begin{aligned} Q_3 \pm \sigma_3 &= \frac{K_3 \left[\frac{C_3(t_2)}{YV} - K_2 Q_1 \right] - K_4 \left[\frac{C_3(t_1)}{YV} - K_1 Q_1 \right]}{K_3 K_6 - K_4 K_5} \\ &\pm \left\{ \frac{K_3^2 \left[\frac{C_3^2(t_2)}{Y^2 V^2} \left(\frac{\alpha_3^2(t_2)}{C_3^2(t_2)} + \frac{y^2}{Y^2} + \frac{v^2}{V^2} \right) \right. \right.}{+ K_2^2 \sigma_1^2} + K_4^2 \left[\frac{C_3^2(t_1)}{Y^2 V^2} \left(\frac{\alpha_3^2(t_1)}{C_3^2(t_1)} \right. \right. \\ &\left. \left. + \frac{y^2}{Y^2} + \frac{v^2}{V^2} \right) + K_1^2 \sigma_1^2 \right] \right\}^{1/2} \\ &\pm \frac{K_3 K_6 - K_4 K_5}{K_3 K_6 - K_4 K_5} \quad (15) \end{aligned}$$

The constants K_i in the above equations are given by

$$K_1 = \frac{\lambda_2 \lambda_3 (1 - e^{-\lambda_1 T}) (e^{-\lambda_1 t_1} - e^{-\lambda_3 t_1})}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}$$

$$\left[\frac{\lambda_2 \lambda_3 (1 - e^{-\lambda_1 T})}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} - \frac{\lambda_3 (1 - e^{-\lambda_2 T})}{(\lambda_3 - \lambda_2)} \right. \\ \left. + \frac{\lambda_2 \lambda_3 (e^{-\lambda_1 T} - e^{-\lambda_2 T})}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} \right] (e^{-\lambda_2 t_1} - e^{-\lambda_3 t_1}) \\ + e^{-\lambda_3 t_1} \left[1 - e^{-\lambda_3 T} + \frac{\lambda_3}{\lambda_3 - \lambda_2} (e^{-\lambda_3 T} - e^{-\lambda_2 T}) \right. \\ \left. + \frac{\lambda_2 \lambda_3 (e^{-\lambda_3 T} - e^{-\lambda_1 T})}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} + \frac{\lambda_2 \lambda_3 (e^{-\lambda_2 T} - e^{-\lambda_3 T})}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} \right] \\ K_2 = \text{same as } K_1 \text{ except } t_1 \text{ replaced by } t_2 \\ K_3 = \frac{\lambda_3 (1 - e^{-\lambda_2 T}) (e^{-\lambda_2 t_1} - e^{-\lambda_3 t_1})}{(\lambda_3 - \lambda_2)} \\ + (1 - e^{-\lambda_3 T}) e^{-\lambda_3 t_1} \\ + \frac{\lambda_3}{\lambda_3 - \lambda_2} (e^{-\lambda_3 T} - e^{-\lambda_2 T}) e^{-\lambda_3 t_1}$$

$K_4 = \text{same as } K_3 \text{ except } t_1 \text{ replaced by } t_2$

$$K_5 = (1 - e^{-\lambda_2 T}) e^{-\lambda_2 t_1}$$

$K_6 = \text{same as } K_5 \text{ except } t_1 \text{ replaced by } t_2$

4. MODIFIED TSIVOGLLOU METHOD

The Tsivoglou method for determining the separate radon daughter air concentrations, Q_1 , Q_2 and Q_3 , requires three measurements of the gross alpha activity on the filter samples at three post-sampling times. In the original development, the three counting times selected were 5, 15 and 30 min after the termination of sampling, and the published method included only the constants for these times.

To be completely general, the method can be modified to account for other counting times, but a reasonable spacing of the counting times is required for accuracy. A modified Tsivoglou method is presented here which allows for variation in the counting times and includes a theoretical development of the statistics involved.

The gross alpha activity of the filter sample includes the alpha counts from both RaA and RaC' and, for the three counting times t_1 , t_2 and t_3 , is given by:

$$\frac{C(t_1) \pm \alpha(t_1)}{(Y \pm y)(V \pm v)} = K_7 Q_1 + K_8 Q_2 + K_9 Q_3 \quad (16)$$

$$\frac{C(t_2) \pm \alpha(t_2)}{(Y \pm y)(V \pm v)} = K_{10} Q_1 + K_{11} Q_2 + K_{12} Q_3 \quad (17)$$

$$\frac{C(t_3) \pm \alpha(t_3)}{(Y \pm y)(V \pm v)} = K_{13} Q_1 + K_{14} Q_2 + K_{15} Q_3 \quad (18)$$

where

$$K_7 = K_1 + (1 - e^{-\lambda_1 T}) e^{-\lambda_1 t_1}$$

$$K_8 = K_3$$

$$K_9 = K_5$$

$$K_{10} = K_7, \text{ with } t_1 \text{ replaced by } t_2$$

$$K_{11} = K_3, \text{ with } t_1 \text{ replaced by } t_2$$

$$K_{12} = K_5, \text{ with } t_1 \text{ replaced by } t_2$$

$$K_{13} = K_7, \text{ with } t_1 \text{ replaced by } t_3$$

$$K_{14} = K_3, \text{ with } t_1 \text{ replaced by } t_3$$

$$K_{15} = K_5, \text{ with } t_1 \text{ replaced by } t_3$$

The solution of these three simultaneous algebraic equations provides the daughter concentrations and the standard errors associated with each. These solutions are:

$$Q_1 \pm \sigma_1 = \frac{J_1 C(t_1) + J_2 C(t_2) + J_3 C(t_3)}{YV} \\ \pm \frac{(J_1^2 D_1 + J_2^2 D_2 + J_3^2 D_3)^{1/2}}{YV} \quad (19)$$

$$Q_2 \pm \sigma_2 = \frac{J_4 C(t_1) + J_5 C(t_2) + J_6 C(t_3)}{YV} \\ \pm \frac{(J_4^2 D_1 + J_5^2 D_2 + J_6^2 D_3)^{1/2}}{YV} \quad (20)$$

$$Q_3 \pm \sigma_3 = \frac{J_7 C(t_1) + J_8 C(t_2) + J_9 C(t_3)}{YV} \\ \pm \frac{(J_7^2 D_1 + J_8^2 D_2 + J_9^2 D_3)^{1/2}}{YV} \quad (21)$$

where $J_1 = (K_{11}K_{15} - K_{12}K_{14})/M$

$$J_2 = (K_9K_{14} - K_8K_{15})/M$$

$$J_3 = (K_8K_{12} - K_9K_{11})/M$$

$$J_4 = (K_{12}K_{13} - K_{10}K_{15})/M$$

$$J_5 = (K_7K_{15} - K_9K_{13})/M$$

$$J_6 = (K_9K_{10} - K_7K_{12})/M$$

$$J_7 = (K_{10}K_{14} - K_{11}K_{13})/M$$

$$J_8 = (K_8K_{13} - K_7K_{14})/M$$

$$J_9 = (K_7K_{11} - K_8K_{10})/M$$

$$D_1 = C^2(t_1) \left[\frac{\alpha^2(t_1)}{C^2(t_1)} + \frac{y^2}{Y^2} + \frac{v^2}{V^2} \right]$$

$$D_2 = C^2(t_2) \left[\frac{\alpha^2(t_2)}{C^2(t_2)} + \frac{y^2}{Y^2} + \frac{v^2}{V^2} \right]$$

$$D_3 = C^2(t_3) \left[\frac{\alpha^2(t_3)}{C^2(t_3)} + \frac{y^2}{Y^2} + \frac{v^2}{V^2} \right]$$

$$M = K_7K_{11}K_{15} + K_8K_{12}K_{13} + K_9K_{10}K_{14} \\ - K_9K_{11}K_{13} - K_8K_{10}K_{15} - K_7K_{12}K_{14}$$

In the special case where the sampling time was 5 min and the count rates have been determined by counts at 5, 15 and 30 min, equations (19), (20) and (21) become:

$$Q_1 \pm \sigma_2 = \frac{5.808C_5 - 13.16C_{15} + 8.306C_{30}}{YV} \\ \pm \frac{(33.73D_1 + 173.2D_2 + 68.99D_3)^{1/2}}{YV}$$

$$Q_2 \pm \sigma_2 = \frac{-3.843C_5 - 12.74C_{15} + 30.85C_{30}}{YV} \\ \pm \frac{(14.77D_1 + 162.3D_2 + 951.5D_3)^{1/2}}{YV}$$

$$Q_3 \pm \sigma_3 = \frac{-1.856C_5 + 25.305C_{15} - 20.58C_{30}}{YV} \\ \pm \frac{(3.445D_1 + 640.1D_2 + 423.7D_3)^{1/2}}{YV}$$

where C_5 , C_{15} and C_{30} are the gross alpha count rates at 5, 15 and 30 min.

5. THEORETICAL COMPARISON OF THE TWO METHODS

The two methods of obtaining the separate radon daughter concentrations in the sampled

atmosphere have been compared theoretically. The exact decay curve exhibited by a 5 min filter sample from an equilibrium atmosphere containing 100 pCi/l. each of the radon daughters was obtained by computer programming equations (1) through (6). With the theoretical individual and gross alpha count rates known, the statistical accuracy of the two methods were compared at selected counting times. The counting times for this comparison were 5, 15 and 30 min for the Tsivoglou method and 5 and 30 min for the spectroscopic method.

The selected post sampling counting times drastically influence the accuracy of the analysis. Procedural aspects usually dictate the initial counting time, however, a minimum delay between the end of sampling and the first count is important since the accuracy for the estimated RaA concentration relies heavily on an early count. A delay of several minutes between the first count and succeeding counts is also necessary for optimum results. The above mentioned counting times are near optimum choices for both methods from practical and statistical considerations.

If zero variance is assumed in the flow rate and detector efficiencies, a comparison of the accuracy provided by the two methods can be made directly. Under these conditions, the standard deviations associated with each daughter concentration obtained by the Tsivoglou method depends directly on the square root of the count rate. The count rate is proportional to the product of the atmospheric concentrations, the flow rate, and the counting yield. If the latter two are fixed, the standard deviation depends only on the square root of the atmospheric concentrations. Figure 1 shows the dependence of the percent standard deviation of arbitrary, but representative, choices of a flow rate equal to 10 l./min and a counting yield of 0.2, for an atmosphere in radioactive equilibrium.

The standard deviations obtained by the spectroscopic method with zero variances for the flow rate and detector efficiency again depend on the square root of the atmospheric concentration, but the accuracy available by this method is improved over that available from the Tsivoglou method. The percent standard deviation for the spectroscopic method

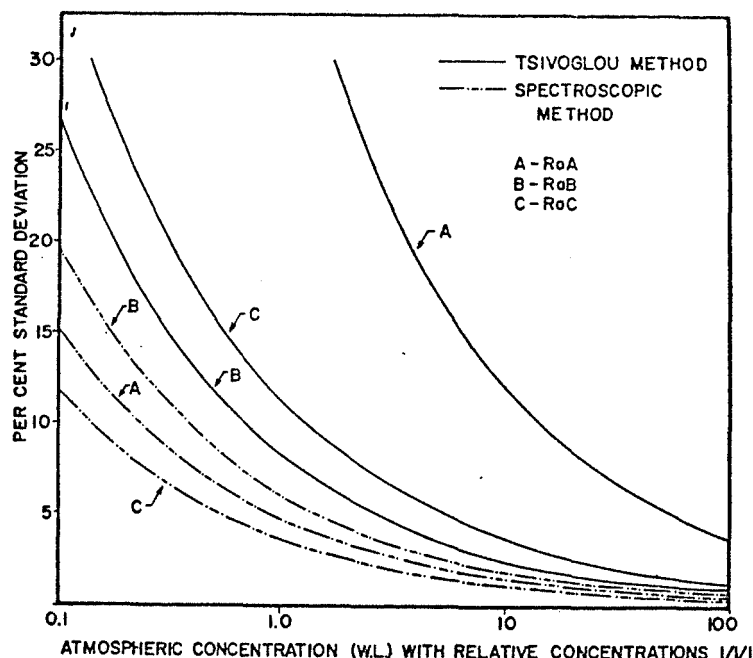


FIG. 1. The minimum percent standard deviations associated with the individual estimates of the radon daughter concentrations in air for filter samples analyzed by the Tsivoglou and Spectroscopic methods. The percent standard deviations shown are for counting statistics only when one minute counts are made at 5, 15 and 30 min. post sampling with a detector of 20% counting yield. An air sample of 50 l. was assumed in both cases from an atmosphere of radon daughters in radioactive equilibrium at the listed concentration Working Level Units.

also is presented in Fig. 1. It should be noted that the percent standard deviations depend upon the relative concentrations of the radon daughters as well as the atmospheric concentration of the particular isotope of interest. An exception is the determination of RaA by the spectroscopic method, which depends only upon the RaA atmospheric concentration.

The contribution to the total percent standard deviation from the flow rate and counting yield standard deviations is dependent upon the relative concentrations of the radon daughters, however independent of the absolute atmospheric concentration. These contributions, for two different relative concentrations, are tabulated in Table 1 as the percent contribution to the total standard deviation associated with each daughter concentration estimate, for various values of the standard deviation of either the flow rate or the counting yield. To arrive at the total standard deviation, the percent standard

deviations must be combined as the square root of the sum of squares.

6. EXPERIMENTAL COMPARISON OF THE TSIVOGLOU AND SPECTROSCOPIC METHODS

To compare the two methods experimentally, air samples were collected over a twelve hour period in the main ventilation exhaust tunnel of the Cotter Corporation's Schwartzwalder Mine near Golden, Colorado. The exhaust tunnel was selected as a sampling site because it integrated the exhaust air from all of the mine areas and, as such, was subject to smaller fluctuations than any other single mine area. The air contained dust and blasting smoke in suspension, and the air velocity was a constant 25 m/min. Fifteen filter samples were collected approximately evenly spaced over a twelve hour period.

Table 1. Percent standard deviation contribution to the total standard deviation of the estimated radon daughter concentrations for various percent standard deviations in flow rate or counting yield. Values for two relative concentrations are given

Flow rate or counting yield standard deviation (%)	Relative concentrations RaA/RaB/RaC	Percent standard deviation contribution to total standard deviation					
		Spectroscopic method			Tsivoglou method		
		RaA	RaB	RaC	RaA	RaB	RaC
1	1/1/1	1.0	2.7	1.7	17.9	3.6	5.11
	1/0.6/0.2	1.0	1.9	2.8	7.4	2.7	10.7
3	1/1/1	3.0	7.9	5.2	53.7	10.7	15.4
	1/0.6/0.2	3.0	5.7	8.4	22.1	8.0	32.1
5	1/1/1	5.0	13.2	8.7	89.6	17.9	25.6
	1/0.6/0.2	5.0	9.6	14.0	36.8	13.4	53.5
10	1/1/1	10.0	26.4	17.4	179.1	35.8	51.1
	1/0.6/0.2	10.0	23.4	28.1	73.6	26.8	106.9

The samples were analyzed both for gross alpha activity and for the individual RaA and RaC' alpha activities at the same time, using a solid state alpha spectrometer and multichannel analyzer system. The counting yield and the sampling flow rate with their associated standard deviations were experimentally determined and found to be 0.217 ± 0.0043 c/d and 4.35 ± 0.13 l./min., respectively.

The individual radon daughter concentrations were computed separately by both methods and the results are presented in Table 2.

Table 3 gives the mean individual radon daughter concentrations for the fifteen samples, the mean theoretical standard deviations and the mean experimental standard deviations obtained by the two methods.

7. DISCUSSION

It is apparent that the spectroscopic method gave more reliable estimates of the individual radon daughter concentrations, particularly for the short-lived RaA component. It appears also that the experimentally determined standard

Table 2. Results from the analysis of membrane filter samples collected in a uranium mine. All samples were collected within a 12 hr period

Number	Estimated atmospheric concentrations (pCi/l.)							
	Tsivoglou method				Spectroscopic method			
	RaA	RaB	RaC	Relative concentrations	RaA	RaB	RaC	Relative concentrations
1	1566	725	343	1.00/0.46/0.22	1350	696	391	1.00/0.52/0.29
2	993	604	628	1.00/0.61/0.63	1360	627	510	1.00/0.46/0.38
3	628	856	713	1.00/1.36/1.13	1540	858	538	1.00/0.56/0.35
4	1242	817	704	1.00/0.66/0.57	1505	832	611	1.00/0.55/0.41
5	1006	751	616	1.00/0.75/0.61	1311	758	518	1.00/0.58/0.40
6	1581	984	627	1.00/0.62/0.40	1632	917	598	1.00/0.60/0.37
7	1720	748	385	1.00/0.44/0.22	1337	713	448	1.00/0.53/0.34
8	1391	758	460	1.00/0.55/0.33	1311	740	464	1.00/0.56/0.35
9	1356	787	491	1.00/0.58/0.36	1375	761	480	1.00/0.55/0.35
10	1433	796	464	1.00/0.56/0.32	1388	772	466	1.00/0.56/0.34
11	1200	713	469	1.00/0.59/0.39	1323	708	425	1.00/0.53/0.32
12	1822	628	478	1.00/0.35/0.26	1378	618	535	1.00/0.45/0.39
13	1655	757	252	1.00/0.46/0.15	1163	701	368	1.00/0.60/0.32
14	518	645	701	1.00/1.25/1.35	1391	707	454	1.00/0.51/0.33
15	718	685	650	1.00/0.95/0.91	1379	708	467	1.00/0.51/0.34

Table 3. Comparison of experimental and theoretical statistical analyses by the Tsivoglou and spectroscopic methods for the fifteen samples listed in Table 2

Isotope	Tsivoglou method			Spectroscopic method		
	Average estimated concentration	Experimental standard deviation	Theoretical standard deviation	Average estimated concentration	Experimental standard deviation	Theoretical standard deviation
RaA	1321	28.6	35.0	1382	7.8	4.0
RaB	750	12.6	10.9	744	12.0	8.5
RaC	532	26.8	24.0	484	14.1	11.0

deviations associated with each estimate agree quite well with the theoretical standard deviations derived for the two methods. The wide range in relative concentrations given by the Tsivoglou method was due largely to the poor estimate of the RaA concentration produced by this method. This contrasted sharply with the small variation in the relative concentrations given by the spectroscopic method.

In the published literature the relative radon daughter concentrations obtained by the Tsivoglou method seldom include statistical estimates of the uncertainties present. The analysis presented here would indicate that many of the relative concentrations so determined are unreliable. The disadvantage of the spectroscopic method of analysis is that it requires more expensive and elaborate instrumentation which precludes its use for normal field measurements in the present form.

8. SUMMARY AND CONCLUSIONS

A method for determining radon daughter concentrations in air has been developed which provides reliable estimates of the individual daughter components. The method includes the theoretical standard deviations associated with each daughter concentration estimate, and it has been demonstrated by repetitive sampling that the theoretical estimates agree with experi-

ment. A modification of the original Tsivoglou method has been developed which gives the theoretical standard deviations in the estimates provided by this technique, and data comparing the two methods is supplied. It is concluded that the spectroscopic method is superior to the Tsivoglou method for determining absolute and relative concentrations.

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