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OPTIMIZING MEASUREMENT SENSITIVITY TO FACILITATE MONITORING ENVIRONMENTAL LEVELS OF Rn-DAUGHTER CONCENTRATIONS†

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Summary

In the measurement of environmental levels of radioactivity, the primary problem is the accumulation of a statistically meaningful number of counts within a reasonable period of time. In the case of measurements of airborne ^{222}Rn-daughter concentrations, the problem is further complicated by the particularly short half-life, 3.05 minutes, of RaA (Po^{218}). Since three Rn-daughters — RaA, RaB (Pb^{214}) and RaC (Bi^{214}) — are of interest, the equations interrelating these Rn-daughter concentrations were derived from the laws of radioactive-series decay. These equations, although straightforward, are cumbersome to solve. To facilitate the efficient use of these equations, a computer program has been written which permits the calculation of Rn-daughter concentrations or expected counts for a given set of measurement parameters (flow rate and detector efficiencies). A subroutine then calculates the optimum pumping and counting times required to provide the number of counts necessary for acceptable statistics at environmental levels of ^{222}Rn-daughter concentrations. This subroutine contains a set of parameters, flow rate and efficiencies, that are fixed using realistic restrictions. The use of these optimized pumping and counting times results in maximum measurement sensitivity under realistic constraints.

Introduction

In previous papers[1,2,3] we described the principles of ^{222}Rn-daughter determination from observed alpha and beta counts and the instrumentation based on these principles. The lowest detectable concentrations with these early instruments were around 1 pCi/liter. For environmental applications an instrument with a detection limit of about 0.1 pCi/liter corresponding to about 10^{-3} Working Level (WL) in equilibrium seemed desirable. At such low concentrations the number of Rn-daughter atoms per liter of air is very small (e.g., 0.1 pCi/liter in equilibrium corresponds to about 1 RaA-atom/liter, 9 RaB-atoms/liter and 6 RaC-atoms/liter of air). This implies that the number of observable disintegrations is also drastically reduced, and an optimization of the measurement sensitivity under existing mechanical, physical and practical constraints becomes mandatory.

Optimizing the Measurement Sensitivity

First, we state the primary constraints:

1) The measurement of the ^{222}Rn-daughter concentrations should be completed in a reasonably short time.

2) The precision of the measurement of 10^{-3} WL under equilibrium conditions should be about 33% relative standard deviation (RSD) or lower. In this case RSD = (standard deviation/10^{-3} WL).

With these two constraints given, one can begin to develop the equations which relate the given ^{222}Rn-daughter concentrations to the observed number of counts. We will make the principle clear by considering the simplest case, RaA, and will then describe the computational procedures for the more complicated cases of RaB and RaC.

The observed alpha counts, A, from RaA are related to the airborne concentrations of RaA, N_A (in atoms/liter) by the following equation:

$$A = \frac{E_A V N_A}{\lambda_A}\left(1-\exp^{-\lambda_A t_B}\right)\exp^{-\lambda_A d}\left(1-\exp^{-\lambda_A t_D}\right) \quad (1)$$

where:

E_A = detection efficiency for RaA alpha particles

V = flow rate (liters/min)

λ_A = decay constant for RaA (min^{-1})

t_B = buildup or pumping time (min)

d = delay time (min)*

t_D = decay or counting time (min).

* This delay is required in order to provide time for the transport assembly to move the filter membrane from the pumping position to the counting position.

It follows immediately from (1) that d should be as small as possible.

The operating characteristics of the filter membrane transport system required that d = 13 sec (0.2167 min). It is also clear that V should be as large as possible. The need to conserve a sharp alpha spectrum, and limitation of the size of the air sampling port by the dimensions of the available alpha detector, forced the limitation of flow rate to about 40 liters/min. We found experimentally that penetration of the aerosol particles into the filter was not significant for this flow rate and that the deformation of the filter membrane by the force of the air stream was tolerable. An enlarged air sampling port size (the diameter of the air sampling port is 30 mm) would have allowed us to use a higher flow rate, but the corresponding increase in counts would have been insignificant since the diameter of active area of the detector was fixed at 30 mm. Since the instrument will use ac power, the power consumption of the motor

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drive for the pump was not a limiting consideration. With the size of the air sampling port and the detector fixed we proceed to investigate the values of the

buildup factor $\left(1 - \exp^{-\lambda_A t_B}\right)$

and the decay factor $\left(1 - \exp^{-\lambda_A t_D}\right)$

for RaA. We calculated the number of alpha counts from 0.1 pCi/L of RaA using preliminary values for t_A and t_B ranging from four to ten minutes. From the obtained number of counts we calculated the relative standard deviations for N_A. Initially we tried $t_B \neq t_D$, but it became clear that $t_B = t_D$, for $t_B + t_D = t$, maximized the product of buildup and decay factor. This can be shown easily in the following manner:

The inequality with $\epsilon > 0$

$$\left(1 - \exp^{[-\lambda_A t_B]}\right)^2 > \left(1 - \exp^{-\lambda_A[t_B + \epsilon]}\right)\left(1 - \exp^{-\lambda_A[t_B - \epsilon]}\right)$$

yields (2)

$$1 < \cosh(\lambda_A \epsilon) \text{ for all } \epsilon > 0$$

which is certainly true.

From the values for the relative standard deviation (RSD) for N_A and (2) we concluded that $t_B = t_D = 3$ min would give us an adequate number of counts. We found RSD ≅ 30.5% for a RaA concentration of 0.1 pCi/liter (E_A = 38.54%, V = 38.7 liters/min). The inequality (2) suggested to us that what was true for RaA should be true for RaB and RaC. Since the equations corresponding to (1) for these other nuclides are much more complicated, we used the computer program described in the next section to obtain the number of beta counts from RaB and RaC and the number of alpha counts from RaC'.

Explanation of Computer Program to Calculate Expected Number of Observed Counts for Various Counting and Pumping Times

In a previous paper[3] on the calibration theory for a Remote Working Level Monitor, the basic equations showing the relationship between the ^{222}Rn-daughter concentrations and the theoretical counts are developed. Further refinements to these equations are made in the calibration program which takes into account the overlap of the RaC' spectrum into the RaA spectrum. This program interrelates the observed [RaA, Ra(B+C) and RaC'] counts to the ^{222}Rn-daughter concentrations. The coefficients used in this program were derived using a counting time and a pumping time of 2.0 min. Additional mathematical manipulation is necessary to generate a program that will relate observed counts for assumed ^{222}Rn-daughter concentrations to a wide range of pumping and counting times.

If we rewrite Eq. (1) into the form:

$$A = (E_A V N_A) C_1 \qquad (3)$$

then C_1 is a numerical coefficient for a fixed pumping time (t_B) and a fixed counting time (t_D) given by

$$C_1 = \frac{\left(1 - \exp^{-\lambda_A t_B}\right) \exp^{-\lambda_A d} \left(1 - \exp^{-\lambda_A t_D}\right)}{\lambda_A}$$

It becomes obvious that new coefficients are required for each change of pumping or counting times. This requires excessive mathematical manipulations even for a programmable calculator. Therefore, a subroutine was written to handle these equations for all three ^{222}Rn-daughter concentrations. This subroutine calculates the nine required coefficients (C_1–C_9) for the set of equations listed below which relate the Rn-daughter concentrations (in atoms/liter) to the observed counts corrected for the overlap of the RaA and RaC' spectrum.

$$\begin{aligned} A &= C_1 E_A V N_A \\ B + C &= (C_2 E_B + C_3 E_C) V N_A \\ &\quad + (C_4 E_B + C_5 E_C) V N_B \\ &\quad + C_6 E_C V N_C \\ C' &= (C_7 N_A + C_8 N_B + C_9 N_C) E_A V \end{aligned} \qquad (4)$$

where:

N_B = airborne concentrations of RaB (in atoms/liter)

N_C = airborne concentrations of RaC (in atoms/liter)

E_B = RaB efficiency

E_C = RaC efficiency.

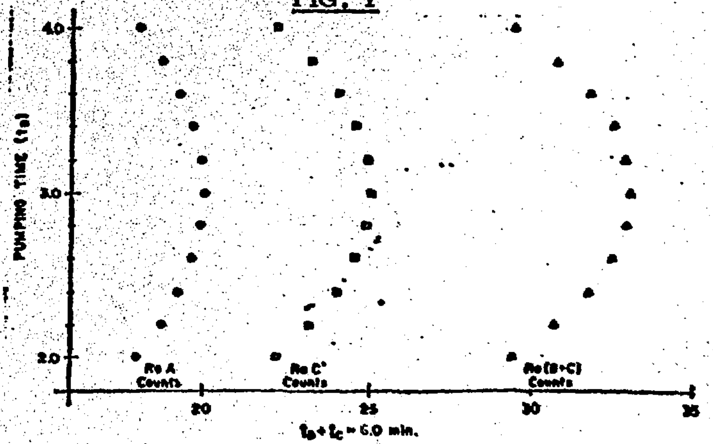
As shown in Eq. (4), the ^{222}Rn-daughter concentrations of interest are assumed and fixed in the program (N_A = 0.977, N_B = 8.585 and N_C = 6.31 atoms/liter). A realistic flow rate is also fixed in the program (V) as well as the alpha efficiency and the two beta efficiencies (E_A, E_B and E_C, respectively).

The program calculates these counts [A, (B+C) and C'] for each requested pumping and counting time. The program asks for pumping time limits and then for counting time limits. It then asks for the iteration time interval. After receipt of this input data it then makes the calculations and prints out the pumping time, counting time, the RaA counts in channel A, the Ra(B+C) counts in channel B, the RaC' counts in the C channel and the Working Level. An example is listed below:

Pumping Time	Counting Time	RaA Counts	Ra(B+C) Counts	RaC' Counts	WL
3.0	2.8	19	31	23	0.00098
3.0	3.0	20	33	25	0.00098
3.0	3.2	21	36	26	0.00098

As discussed earlier, it was shown that for RaA the counts were maximized by fixing $t_B = t_D$ for any given sum of pumping and counting times. The results of the computer program agreed with these findings for the RaA counts and gave similar results for Ra(B+C) and RaC' counts as indicated in Fig. 1.

FIG. 1



This plot of the computer output data also shows the symmetry observed between the pumping and counting times. This implies that the same counts will be generated for specific fixed measuring times ($t_B + t_D$), when the counting time and pumping time are interchanged [i.e., ($t_B + t_D$) = 6 = (2.0 + 4.0) or (4.0 + 2.0)]. This information would be very useful when power is limited, such as with a battery-powered instrument, since the pump motor is the major power drain of the system. It should be pointed out, however, that unequal pumping and counting times will not produce the maximum count.

For specific design criteria, such as battery-powered operation, it may be desirable to compromise the time parameters. The differences in counts at low Working Levels are small for short measurement periods; and, since a short pumping time may be desirable, a small increase in measurement time would compensate for the loss in the optimum ratio. For example:

Pumping Time	Counting Time	RaA Counts	Ra(B+C) Counts	RaC' Counts	WL
2.0	5.0	21	37	28	0.98×10^{-3}

The next question to be addressed is whether the predicted counts are adequate to yield the desired precision at these environmental levels? Based on the calculation of the relative standard deviations (RSD) of the predicted counts, adequate precision is obtained at environmental levels as shown in the following table.

Relationship of Counts to RSD at Specific Concentrations

RSD (%)	Rn-daughter concentrations		Counts	Background Counts
30.5	RaA	0.100 pCi/L	20	0
72.5	RaB	0.096 pCi/L	33	15*
26.7	RaC	0.101 pCi/L	25	0
30.8	WL	0.983×10^{-3}	-	-

* This background count corresponds to a gamma background of approximately 5 μR/hr.

Conclusion

We have described a procedure for the design of a ^{222}Rn-daughter monitor for environmental concentrations of 0.1 pCi/liter or higher. We found that these concentrations could be measured with adequate precision in a gamma background of about 10 μR/hr within approximately 6 min in a fully automatic manner. We have also shown that the program discussed is very useful in a variety of designs and that no one set of design parameters will be the optimum for every conceivable instrument. A device designed according to these principles[1,2,3] is presently being used by the United States Environmental Protection Agency to monitor dwellings in Florida.

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