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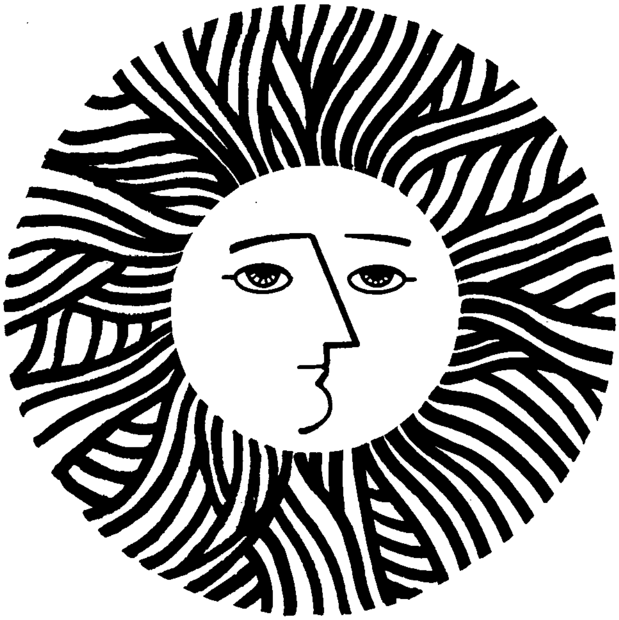

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ABSTRACT

A technique for measuring ^{222}Rn-progeny concentrations in air, involving counting alpha decays for three intervals on a filter through which air has been drawn, is optimized for measuring low concentrations indoors. Counting intervals are selected to minimize a linear combination of the minimum measurable concentrations -- concentrations at which the relative standard deviation in the measurement due to counting statistics is 20%. The effects on the MMCs of varying total measurement time, sampling and delay times, and of radon progeny activity ratios are considered. Previous work on this technique focussed on measurements in uranium mines where concentrations of radon progeny are typically much higher than in residences. By extending the total measurement time from 35 to 60 minutes, the MMCs are reduced by factors of 3, 7, and 4 for ^{218}Po, ^{214}Pb, and ^{214}Bi, respectively, thereby permitting precise measurement of indoor concentrations down to the order of one pCi/l.

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INTRODUCTION

A widely used technique for measuring the concentrations of ^{222}Rn progeny (^{218}Po, ^{214}Pb, and ^{214}Bi) in air involves drawing air through a filter, then counting the total alpha activity on the filter for three specified time intervals after sampling. The concentrations of the radon decay products are calculated by taking linear combinations of the three count totals, with the coefficients obtained by solving the Bateman equations. (Fu-Chia and Chia-Yong provide general equations for the counts resulting from the alpha or beta decay of radon progeny collected on a filter (Fu78).)

Tsivoglou _et al_. first proposed such a technique for measuring radon progeny concentrations in mine atmospheres; they used a ratemeter to determine alpha activity 5, 15, and 30 minutes after sampling (Ts53). Integrating devices, i.e. counters, subsequently became sufficiently portable for use in the field and, because they yield better precision, replaced ratemeters for measuring radon progeny activity on filters. Thomas optimized the count-interval timing to minimize measurement precision for a total measurement time of 35 minutes; he recommended counting from 2 to 5, 6 to 20, and 21 to 30 minutes after a five-minute sampling period (Th72). Busigin and Phillips showed that delays of greater than one minute between counting intervals yield somewhat better precision (Bu80). In optimizing measurement precision they also considered uncertainties due to variations in the rate at which radon progeny are collected on a filter. They suggested counting from 2 to 5, 7 to 15, and 25 to 30 minutes after a five-minute sampling period.

In each of these three papers, the authors discuss application of the measurement technique only in uranium mines, where radon progeny concentrations of interest are in the range of ten to several hundred pCi/l. In recent years a number of researchers have studied radon and radon progeny in residences, where the concentrations of interest commonly range from less than one to ten pCi/l. (See, for example, the recent special issue of Health Physics on indoor radon.) The precision of the total-alpha three-count method is not adequate for measuring low concentrations of radon decay products indoors when the total measurement time is limited to 35 minutes. For example, assuming a sampling rate of 10 liters per minute, a counting efficiency of 0.4, and progeny activity ratios of 0.6 for ^{214}Pb:^{218}Po and 0.4 for ^{214}Bi:^{218}Po, the concentrations at which the relative standard deviation in the measurement is 20% due to counting statistics alone are 7.7, 1.6, and 2.5 pCi/l for ^{218}Po, ^{214}Pb, and ^{214}Bi, respectively (Na81).

Although measurement precision can be improved by increasing the sampling flow rate, high flow rates can perturb the environment being measured. An alternative approach to improving measurement precision involves extending the total measurement time beyond 35 minutes. In addition, in many cases it may be practical to begin counting one minute after the end of sampling, rather than two, thereby improving the measurement precision for ^{218}Po. In this note, I discuss the effects of total measurement time, sampling time, delay time, and radon progeny activity ratios on measurement precision using the total-alpha three-count technique. I show that extending the total measurement time to 60 minutes, with a one-minute delay between the end of sampling and the beginning of the first counting interval, leads to an improvement in measurement sensitivity by factors of 3, 7, and 4 for ^{218}Po, ^{214}Pb, and ^{214}Bi, respectively, relative to using a 35-minute measurement time with a two-minute delay.

OPTIMIZATION PROCEDURE

The basis for optimizing count-interval timing is the minimum measurable concentration (MMC) which I define to be the concentration at which the relative standard deviation (RSD) in the measurement due to counting statistics is 20%, assuming the product of detector efficiency and sampling flow rate to be 1.0 liters per minute. To compute the MMCs I define the following symbols:

C is a 1 X 3 matrix where C_i is the number of alpha counts detected in the ith counting interval;

I is a 1 X 3 matrix where I_j is the activity concentration of the jth radon decay product (^{218}Po, ^{214}Pb, ^{214}Bi, respectively);

M is a 1 X 3 matrix where M_j is the minimum measurable concentration of the jth radon decay product;

M_p is the minimum measurable concentration of the potential alpha energy concentration (PAEC) (working level) of radon progeny;

p is the PAEC of radon progeny;

R is a 1 X 3 matrix of activity ratios where $R_j = I_j/I_1$;

$X = (0.00103, 0.00507, 0.00373)$ ($WL \cdot l \cdot pCi^{-1}$), so that

$$p = \sum_{j=1}^{3} X_j I_j$$

V is the sampling flow rate; and

η is the detection efficiency for alpha decays.

The count matrix can be expressed as a function of the activity matrix:

$$C_i = \eta V \sum_{j=1}^{3} H_{ij} I_j, \tag{1}$$

where H is a 3 X 3 matrix whose elements depend upon the sample and count-interval timing (see Appendix). This equation can be inverted to obtain

$$I_j = \frac{1}{\eta V} \sum_{i=1}^{3} K_{ji} C_i \tag{2}$$

where $K = H^{-1}$.

Likewise, the PAEC can be computed from the count matrix as

$$p = \frac{1}{\eta V} \sum_{i=1}^{3} L_i C_i, \tag{3a}$$

where

$$L_i = \sum_{j=1}^{3} X_j K_{ji}. \tag{3b}$$

The minimum measurable concentrations can then be computed, using standard propagation-of-error formulae (Be69), for specified timing conditions and activity ratio matrix, R;

$$M_j = \frac{A}{R_j}\left[\sum_{i=1}^{3} K_{ji}{}^{2}\left(\sum_{g=1}^{3} H_{ig} R_g\right)\right], \text{ and} \tag{4}$$

$$M_p = \frac{A}{\left[\sum_{i=1}^{3} X_i R_i\right]}\left[\sum_{i=1}^{3} L_i^{2}\left(\sum_{g=1}^{3} H_{ig} R_g\right)\right] \tag{5}$$

where, by definition,

$$A = \frac{1}{(\mathrm{RSD})^2 \eta V} = 25. \tag{6}$$

To choose among different count-interval timing sets, a single optimization parameter must be selected. In previous work, ^{218}Po measurement precision was used as the optimization parameter (Th72, Bu80). In addition to doing calculations following this example, I used two other optimization parameters, $M_a{}^*$ and $M_b{}^*$, where

$$M_a^* = 1/3\ (M_1 + M_2/R_2 + M_3/R_3), \text{ and} \tag{7a}$$

$$M_b^* = 1/3\ (M_1 + M_2 + M_3). \tag{7b}$$

For each of the three cases the results were quite similar; for all results reported in this note M_a^* was used as the optimization parameter.

Optimizations were performed for specified values of the sampling time, the start of the first counting interval, the total measurement time, and the activity ratio matrix. The four intermediate times (end of first count interval, beginning and end of second count interval, and beginning of third count interval) were varied using a gradient search technique (Be69) from an arbitrary initial value to obtain the optimal values (i.e., those that result in the minimum value of M_a^*). Near the minimum of M_a^*, small changes in the timing have little effect on the MMCs, so, to make the timing easy to use in practice, integral-minute time sets in the vicinity of the optimal time set were evaluated. The integral-minute time set with the lowest value of M_a^* is taken as the recommended timing.

All of the optimization results reported in this note were computed with an activity ratio matrix of (1, 0.5, 0.4), representative of ratios found inside houses with relatively low air-exchange rates (i.e., less than 0.5 air changes per hour.) The optimal timing is quite insensitive to the activity ratio matrix; only for extreme disequilibrium does the optimal timing change significantly.

Throughout this note, I use the notation (t_s, t_{1a}-t_{1b}, t_{2a}-t_{2b}, t_{3a}-t_{3b}) to specify measurement timing, where t_s is the sampling time, and t_{ia} and t_{ib} give the start and end times, respectively, of the ith counting interval. Each of these times is referenced to the beginning of sampling, so t_{3b} is the total measurement time and t_{1a}-t_s is the delay time.

RESULTS AND DISCUSSION

The effect on measurement precision of varying the total measurement time for the total-alpha three-count technique was examined by computing optimized MMCs for total measurement times between 30 and 80 minutes, with a sample time of five minutes, delay times of one and two minutes, and an activity ratio matrix of (1, 0.5, 0.4). The results of this analysis are plotted in Figure 1 along with points indicating the MMCs for the timing recommended by Thomas (Th72). The most important feature of this figure is the steepness of the curves, particularly for total measurement times in the vicinity of 35 minutes. This result indicates that modest increases in the total measurement time lead to substantial improvements in measurement precision. To select the best total measurement time, one must balance the goal of improved measurement precision with any of a number of factors which favor rapid measurements such as desire for quick results, high repetition rate, and low measurement cost. For many applications in residences a total measurement time of 60 minutes provides a reasonable compromise between these goals.

Other aspects of the results presented in Figure 1 are worth mentioning. The measurement precision of the total-alpha three-count technique is much poorer for ^{218}Po than for either ^{214}Pb or ^{214}Bi over the entire range of total measurement times considered. It is, perhaps, a surprising result that measurement precision for ^{218}Po, with a 3-minute half-life, continues to improve as measurement times are extended beyond one hour; in essence this occurs because the contribution to counts detected during the first counting interval from sampling ^{214}Pb and ^{214}Bi in air is more precisely known if longer measurement times are used, hence the counts detected due to the collection of ^{218}Po are more precisely known. Figure 1 also shows that the differences in measurement precision between the timing recommended by Thomas and the optimized timing for a 35-minute measurement time and a two-minute delay are small. Finally, PAEC is seen to be measured far more precisely than any of the individual decay-product concentrations; this result is due to a cancellation of errors, as noted by Busigin and Phillips (Bu80).

The effect of varying the sampling time on measurement precision is shown in Figure 2. For a total measurement time of 60 minutes the MMC for ^{218}Po has a minimum at a sampling time of six minutes, and over the range of 3.7 to 9.5 minutes varies only between 11 and 12 pCi/l. For the other two radon decay products longer sampling times result in better measurement precision over the entire range analyzed (2 to 14 minutes). An implicit assumption, however, in the total-alpha three-count technique is that the concentrations of radon progeny in air remain constant throughout the sampling period. This assumption will be better satisfied for shorter sampling times. It therefore seems reasonable to continue to use a five-minute sampling time as recommended by

Thomas (Th72).

The effect of varying the delay time between the end of sampling and the beginning of the first count interval is most significant for the measurement precision of ^{218}Po, as one might expect given its short half-life (Figure 3). Clearly, minimizing this delay time is desirable. Thomas concluded that, for measurements in a mine, two minutes was a reasonable minimum. For measurements in houses, the sampling site and counting site are commonly in close proximity making a one-minute delay time practical.

A summary of measurement precision and calculation constants for the timing recommended by Thomas and for optimized 60-minute measurement periods with one- and two-minute delays is presented in Table 1. The radon progeny concentrations are calculated from equations (2) and (3a) and standard deviations in the measurements due to counting statistics are

$$\sigma_{I_i} = \frac{1}{\eta V}\left(\sum_{j=1}^{3} K_{ij}^{2} C_j\right)^{1/2} \text{, and} \tag{8a}$$

$$\sigma_P = \frac{1}{\eta V}\left(\sum_{j=1}^{3} L_j^{2} C_j\right)^{1/2} . \tag{8b}$$

Table 1 presents the MDCs for each of these techniques for three activity-ratio conditions. A high degree of disequilibrium is seen to improve the measurement precision for ^{218}Po and reduce that for ^{214}Bi; conversely, complete equilibrium results in poor precision for ^{218}Po and improved precision for ^{214}Bi. The measurement precision for ^{214}Pb is seen to be only slightly affected by the activity ratios.

This result can be seen more clearly in Figures 4 and 5 where lines of equal MMCs are plotted in a triangular space whose points represent all sustainable radon progeny activity ratios. Figure 4 is plotted for the timing recommended by Thomas while Figure 5 is computed for the timing recommended in this work for a 60-minute period with a one-minute delay. Comparing these figures we see that the MMCs are improved roughly by factors of 3 for ^{218}Po, 7 for ^{214}Pb and 4 for ^{214}Bi for all activity ratios. Most of the measurements of indoor radon progeny concentrations made by researchers at Lawrence Berkeley Laboratory lie below but near the diagonal line that reflects the condition $R_2 = R_3$ (see, for example, Re81). In this region, only the measurement precision for ^{218}Po depends significantly on activity ratios.

Busigin and Phillips showed that fluctuations in the rate of collection of radon progeny on a filter substantially increase measurement uncertainty, particularly for ^{218}Po (Bu80). They assumed a normal distribution and characterized the fluctuations by specifying a relative standard deviation (RSD) for pump speed and concentration variations. They suggest that this contribution to uncertainty be included in optimizing any radon progeny measurement technique. Close examination of Table 1 in their paper shows, however, that including this contribution in the optimization has only a modest impact on measurement precision -- insufficient, in my judgement to mandate its inclusion, particularly since the variations are at best difficult to quantify. (In their Table 1, the differences in RSD for ^{218}Po are less than 3% between using timing #11, optimized for no fluctuation in collection rate and timing #18, their suggested procedure, optimized for 3% RSD in pump speed). In Table 2 I present RSDs of progeny concentrations calculated for timing sets recommended by Thomas, by Busigin and Phillips, and in this work. The results are presented for three different sets of radon progeny concentrations and for RSDs in the rate of collection of radon progeny on the filter of 0% and 5%. While Busigin and Phillip's timing shows the greatest insensitivity to collection rate fluctuations (as measured by the fractional increase in RSD of the measured progeny concentrations between $\Delta I = 0\%$ and $\Delta I = 5\%$), the 60-minute measurement procedure has substantially lower RSDs for all conditions.

SUMMARY

The total-alpha three-count technique, as developed for use in mines, has inadequate precision for many applications studying radon progeny indoors. By extending the total measurement time from 35 to 60 minutes the measurement precision is improved substantially, thereby allowing radon concentrations typically found in residences to be measured with moderate precision and modest sampling flow rate and detector efficiency requirements. The timing sequences optimized for a 60-minute measurement period with a five-minute sampling period and one- and two-minute delays are (5, 6-9, 12-29, 40-60) and (5, 7-10, 13-30, 42-60) respectively.

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APPENDIX: CALCULATING THE H-MATRIX ELEMENTS

For a timing sequence specified by (t_o, t_{1a}-t_{1b}, t_{2a}-t_{2b}, t_{3a}-t_{3b}), the H-matrix elements are given by

$$H_{i1} = G_{11}(t_{ib}) - G_{11}(t_{ia}) + G_{31}(t_{ib}) - G_{31}(t_{ia}) \qquad i=1,3 \qquad \text{(A1a)}$$

and

$$H_{ij} = G_{3j}(t_{ib}) - G_{3j}(t_{ia}) \qquad i=1,3; j=2,3. \qquad \text{(A1b)}$$

The functions $G_{ij}(t)$ give the number of decays during the interval t_o to t of the ith decay product due to the collection on the filter of one pCi of the jth decay product:

$$G_{11}(t) = \frac{2.22}{\lambda_1{}^2} r_1 s_1(t), \tag{A2a}$$

$$G_{31}(t) = \frac{2.22}{\lambda_1}\left[\frac{f_{21}f_{31}}{\lambda_1} r_1 s_1(t) + \frac{f_{12}f_{32}}{\lambda_2} r_2 s_2(t) + \frac{f_{13}f_{23}}{\lambda_3} r_3 s_3(t)\right], \tag{A2b}$$

$$G_{32}(t) = \frac{2.22}{\lambda_2}\left[\frac{f_{32}}{\lambda_2} r_2 s_2(t) + \frac{f_{23}}{\lambda_3} r_3 s_3(t)\right], \tag{A2c}$$

and

$$G_{33}(t) = \frac{2.22}{\lambda_3^2} r_3 s_3(t). \tag{A2d}$$

In these equations λ_j is the decay constant of the jth decay product ($\lambda_1 = 0.227$ min^{-1}, $\lambda_2 = 0.0259$ min^{-1}, $\lambda_3 = 0.0352$ min^{-1}); and

$$f_{ij} = \frac{\lambda_i}{\lambda_i - \lambda_j}, \tag{A3a}$$

$$r_i = 1 - e^{-\lambda_i t_o} \text{ , and} \tag{A3b}$$

$$s_i(t) = 1 - e^{-\lambda_i (t-t_o)} . \tag{A3c}$$

Measurement Timing (min) [reference]	Calculation Matrix Elements					Minimum Measurable Concentrations				
	i	K_{11}	K_{12} ($pCi \cdot dis^{-1} \cdot min^{-1}$)	K_{13}	L_i ($WL \cdot dis^{-1} \cdot min^{-1}$)	$R_2:R_3$	^{218}Po (pCi/l)	^{214}Pb (pCi/l)	^{214}Bi (pCi/l)	PAEC (WL)
(5, 7-10, 11-25, 26-35) [Th72]	1	0.16894	-0.08200	0.07753	9.616×10^{-5}	1.0:1.0	68.7	6.7	7.8	0.0102
	2	0.00122	-0.02057	0.04909	-6.499×10^{-5}	0.5:0.4	35.1	6.5	9.4	0.0097
	3	-0.02253	0.03318	-0.03771	18.81×10^{-5}	0.3:0.1	19.1	5.5	19.2	0.0088
(5, 7-10, 13-30, 42-60) [present work]	1	0.13121	-0.04844	0.03294	4.863×10^{-5}	1.0:1.0	32.0	1.0	2.1	0.0014
	2	-0.01237	-0.00264	0.01609	0.222×10^{-5}	0.5:0.4	16.7	1.0	2.6	0.0014
	3	-0.00638	0.01756	-0.01500	5.955×10^{-5}	0.3:0.1	9.4	1.0	5.4	0.0014
(5, 6-9, 12-29, 40-60) [present work]	1	0.10458	-0.03868	0.02371	3.745×10^{-5}	1.0:1.0	20.9	0.9	1.9	0.0012
	2	-0.00904	-0.00374	0.01485	0.420×10^{-5}	0.5:0.4	11.2	0.9	2.3	0.0012
	3	-0.00655	0.1689	-0.01258	5.279×10^{-5}	0.3:0.1	6.5	0.9	4.8	0.0012

Table 1. Calculation constants and minimum measurable concentrations of radon progeny for the total-alpha three-count technique. The three time sets listed are the 35-minute measurement timing recommended by Thomas (Th72), and 60-minute measurement times with two-minute and one-minute delays recommended in this work. Equations (2), (3) and (8) in the text give the relationships between the counts observed, the calculation constants, and the progeny concentrations and measurement uncertainties. The minimum measurable concentration is defined as the level at which the relative standard deviation in the measurement due to counting statistics is 20% assuming the product of flow rate and detector efficiency to be 1.0 liters per minute.

Measurement Timing (minutes) [reference]	Radon Daughter Concentrations (pCi/l) ^{218}Po:^{214}Pb:^{214}Bi	Relative Standard Deviation in Measured Concentration					
			ΔI = 0%			ΔI = 5%	
		^{218}Po	^{214}Pb	^{214}Bi	^{218}Po	^{214}Pb	^{214}Bi
5, 7-10, 11-25, 26-35	5.0:1.5:0.5	0.20	0.19	0.62	0.24	0.23	0.76
[Th72]	4.0:2.0:1.6	0.29	0.18	0.24	0.38	0.24	0.34
	2.0:2.0:2.0	0.58	0.18	0.19	0.80	0.25	0.28
5, 7-10, 12-20, 30-35	5.0:1.5:0.5	0.20	0.20	0.62	0.23	0.23	0.72
[Bu80]	4.0:2.0:1.6	0.30	0.18	0.24	0.36	0.22	0.30
	2.0:2.0:2.0	0.59	0.19	0.19	0.74	0.23	0.25
5, 6-9, 12-29, 40-60	5.0:1.5:0.5	0.11	0.08	0.30	0.14	0.11	0.42
[present work]	4.0:2.0:1.6	0.17	0.07	0.12	0.22	0.10	0.18
	2.0:2.0:2.0	0.32	0.07	0.09	0.44	0.10	0.15

Table 2. Relative standard deviations in measured radon progeny concentrations using three different count-interval time sets. ΔI is the relative standard deviation in the rate at which decay products are collected on a filter. The flow rate and detector efficiency are assumed to be 10 l/min. and 0.4, respectively, typical values for measurements indoors.

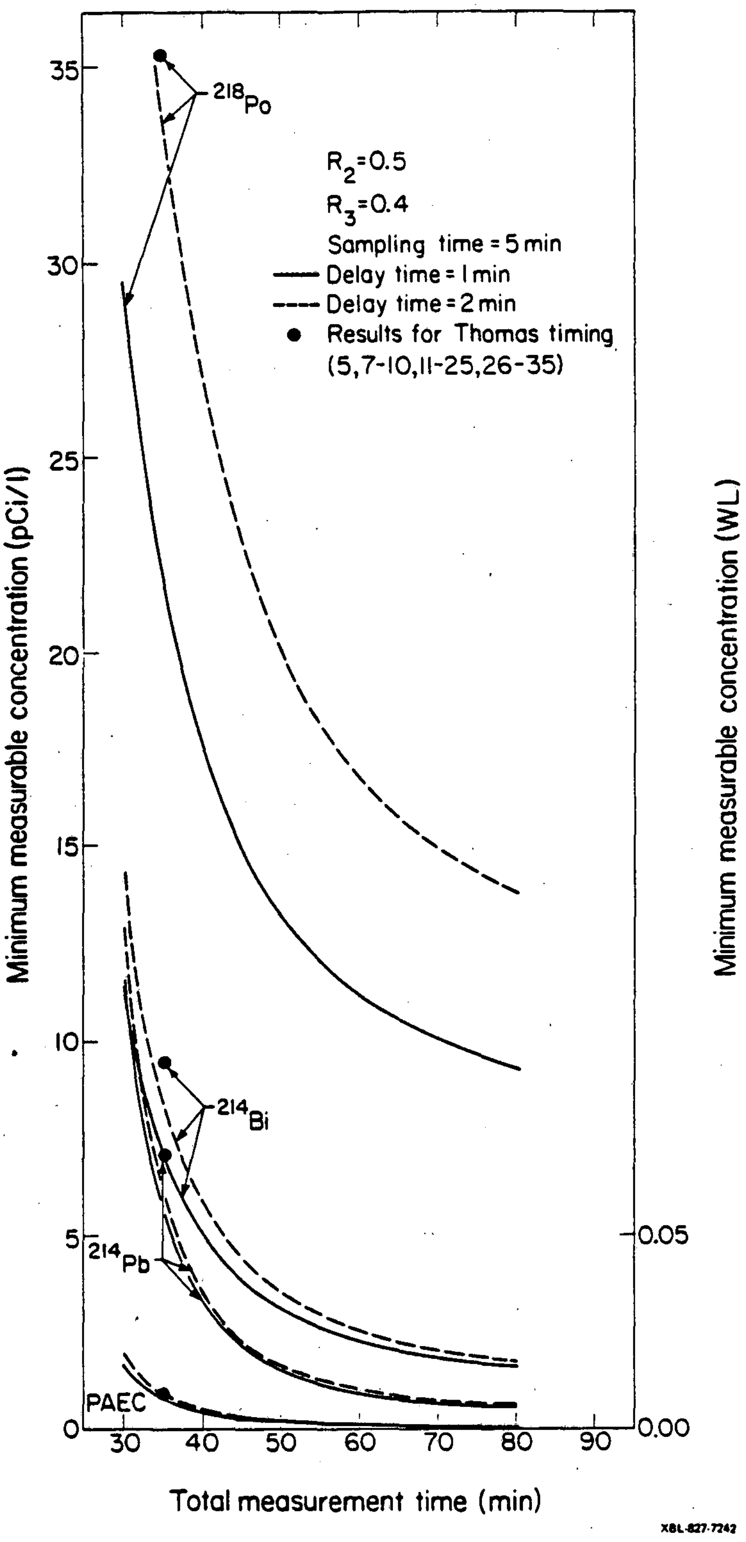

Figure 1. Optimized minimum measurable concentrations of radon progeny for the total-alpha three-count technique as a function of total measurement time.

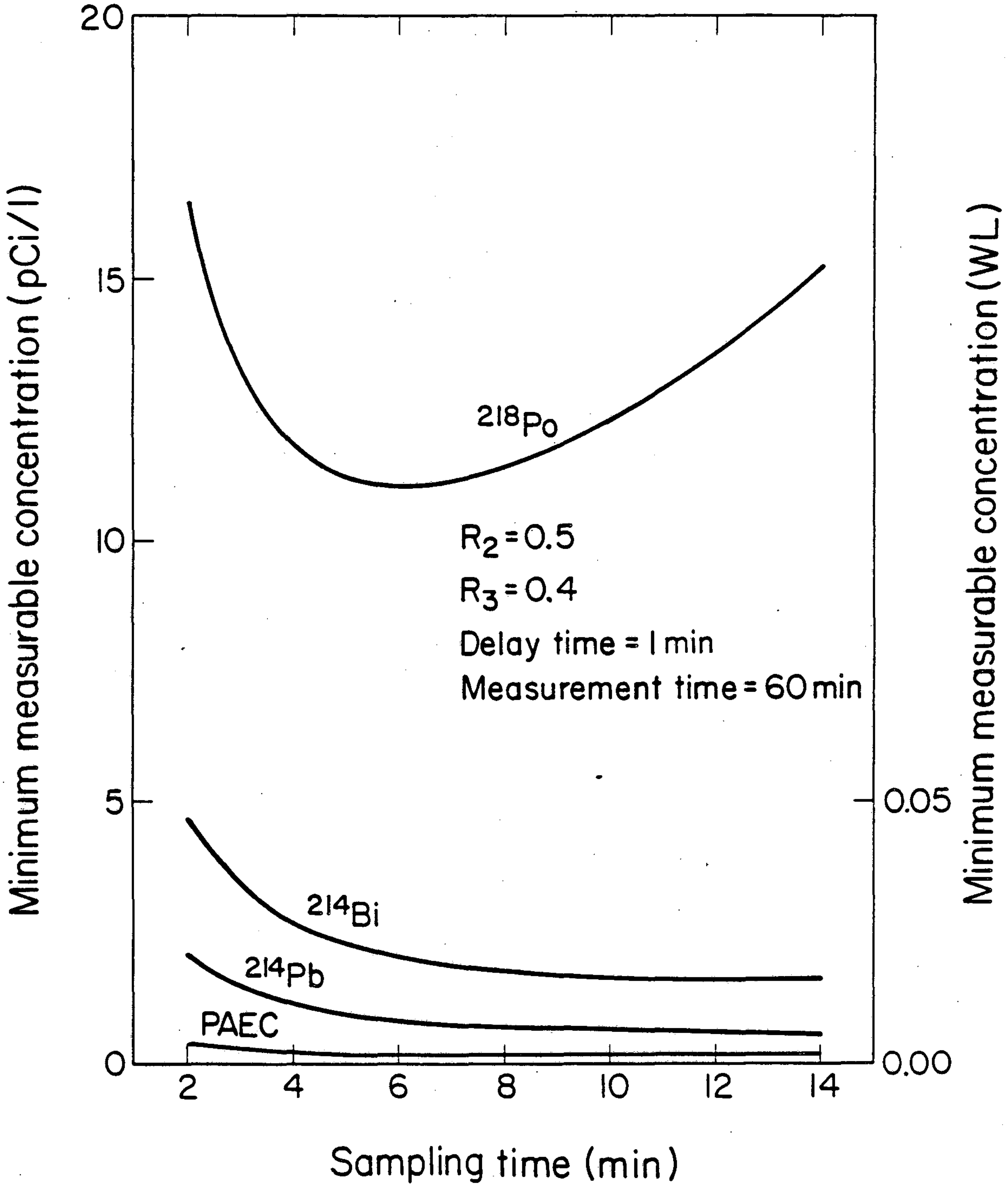

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Figure 2. Optimized minimum measurable concentrations of radon progeny as a function of sampling time.

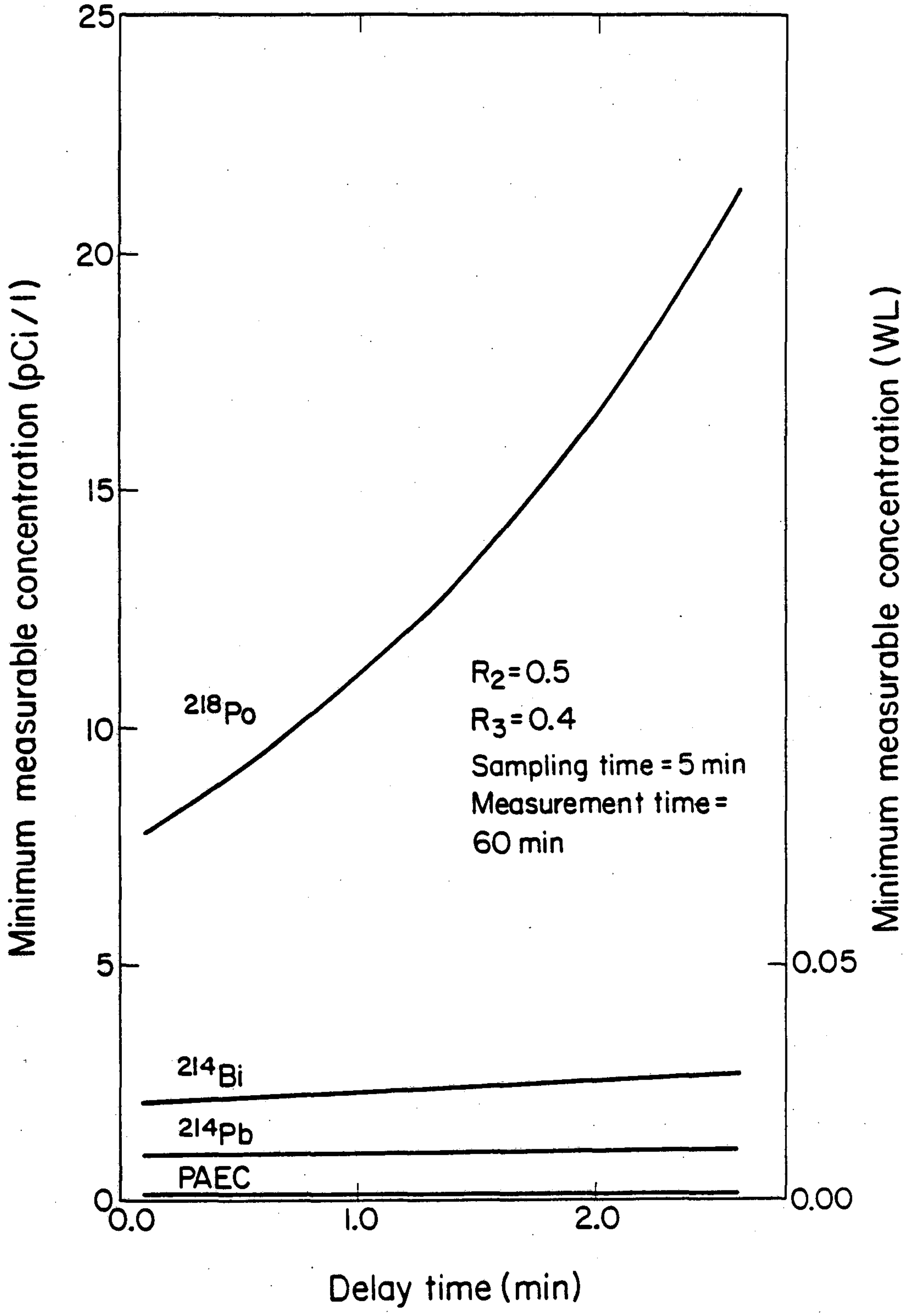

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Figure 3. Optimized minimum measurable concentrations of radon progeny as a function of delay time between the end of sampling and the beginning of first counting period.

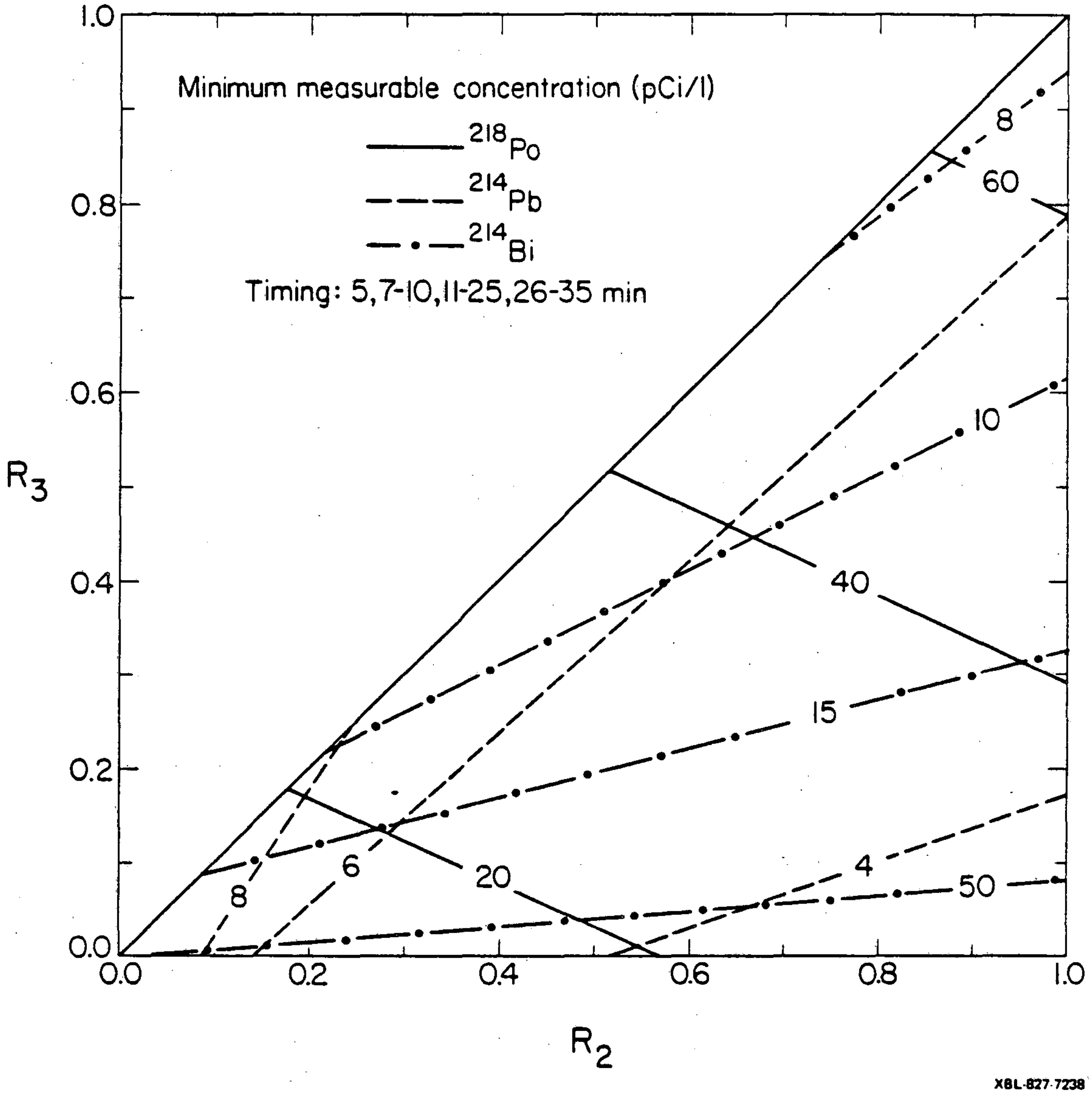

Figure 4. Isopleths of minimum measurable concentrations of radon progeny as functions of activity ratios for timing recommended by Thomas (Th72). R_2 and R_3 are the activity ratios of ^{214}Pb and ^{214}Bi, respectively, to ^{218}Po.

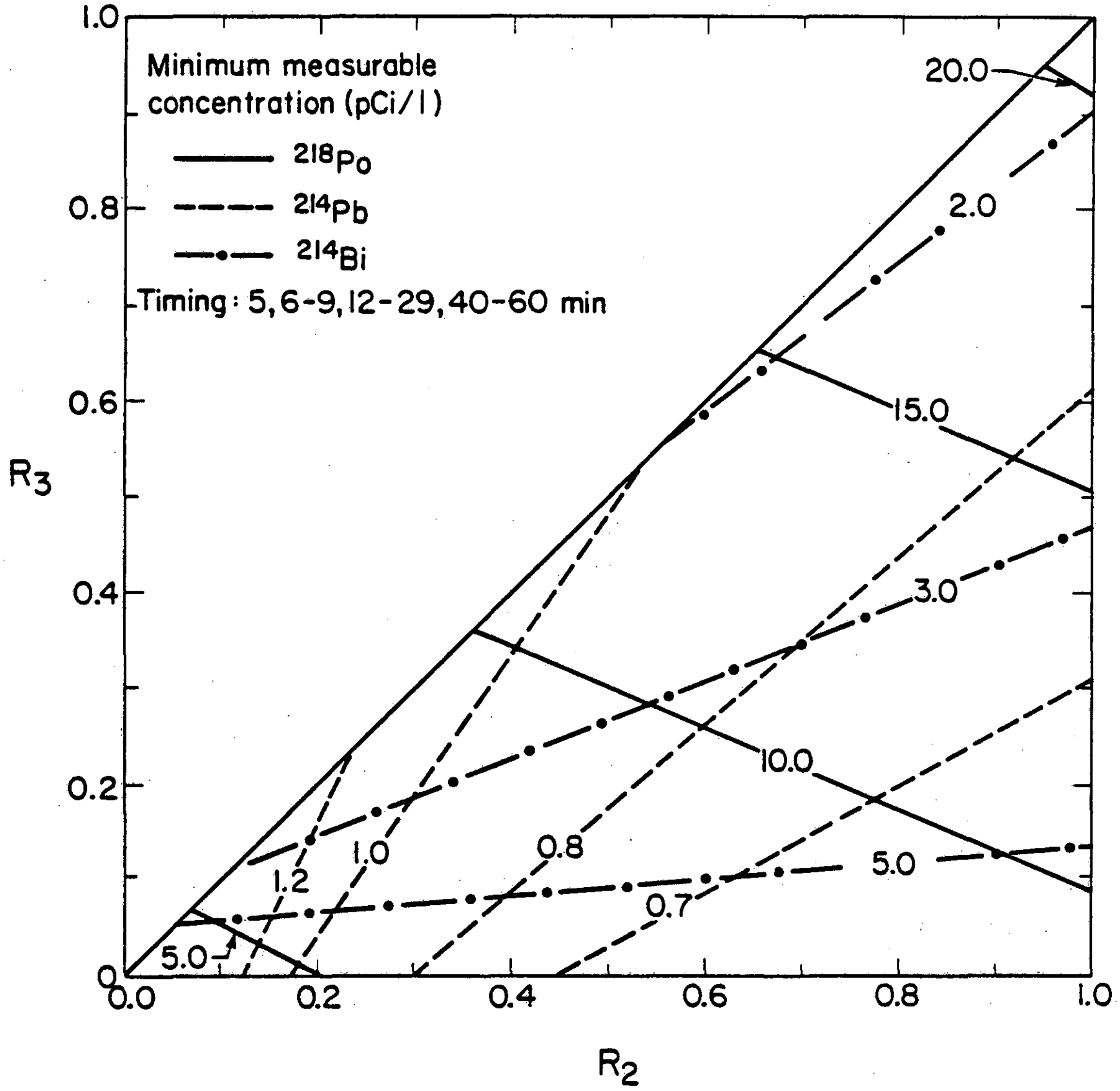

Figure 5. Isopleths of minimum measurable concentrations of radon progeny as functions of activity ratios for optimized 60-minute total measurement time with a one-minute delay between the end of sampling and the beginning of the first counting interval.

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