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### Author

Nazaroff, W.W.

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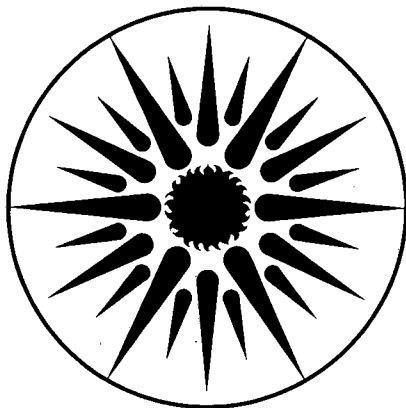
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IN RESIDENCES

W.W. Nazaroff

January 1983

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W.W. Nazaroff

Building Ventilation and Indoor Air Quality Program  
Lawrence Berkeley Laboratory  
University of California  
Berkeley, California 94720

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ABSTRACT

A technique for measuring  $^{222}\text{Rn}$ -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 typically found indoors. Sampling and counting intervals are selected to minimize a linear combination of the minimum measurable concentrations (MMCs) -- 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. The effect on measurement precision of variations in the rate of collection of radon progeny on the filter is also evaluated. Previous work on this technique has often focussed on measurements in uranium mines where concentrations of radon progeny are typically much higher than in residences. With sequential sampling and counting intervals, extending the total measurement time from 35 to 60 minutes reduces the MMCs by factors of 3, 7, and 4 for  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Bi}$ , respectively, thereby permitting precise measurement of indoor concentrations down to the order of one pCi/l. By employing an instrument that counts while sampling, the MMCs can be further reduced by factors of 5.5 for  $^{218}\text{Po}$ , 2 for  $^{214}\text{Pb}$  and 2.5 for  $^{214}\text{Bi}$ , again for a 60-minute measurement period.

## INTRODUCTION

A widely used technique for measuring the concentrations of  $^{222}\text{Rn}$  progeny ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Bi}$ ) 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 (Fu78, for example).

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 special issue of Health Physics on indoor radon, scheduled for publication in early 1983.) The precision of the total-alpha three-count method with sequential sampling and counting intervals is not adequate for measuring low concentrations of radon decay products indoors when the total measurement time is limited to 35 minutes. For example, using the timing recommended by Thomas and assuming a sampling rate of 10 liters per minute, a counting efficiency of 0.4, and progeny activity ratios of 0.6 for  $^{214}\text{Pb}:$  $^{218}\text{Po}$  and 0.4 for  $^{214}\text{Bi}:$  $^{218}\text{Po}$ , 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  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Bi}$ , respectively (Na81).

Although measurement precision can be improved by increasing the sampling flow rate, high rates of air movement may perturb the environment being measured (see, for example, Ho79). An alternative approach to improving measurement precision involves extending the total measurement time beyond 35 minutes. Also it is practical in many cases to begin counting one minute after the end of sampling, rather than two, thereby improving the measurement precision for  $^{218}\text{Po}$ .

Yet another approach to improving measurement precision is to overlap the sampling and counting intervals. To do so requires a sampling head which has a detector facing the front surface of the filter. James and Strong reported on the development of such an instrument for use in mines (Ja73), and Cliff demonstrated that by counting during sampling

one could achieve significantly better precision, particularly for  $^{218}\text{Po}$ , than results from the timing recommended by Thomas (C178). It is my perception that in spite of this finding most workers continue to use sequential sampling and counting intervals. This may be due to the unavailability of commercial instruments. Whether the complex sampling-head geometry may lead to substantial errors from non-uniform collection of radon progeny on the filter, or from their deposition on other sampling-head surfaces has not been fully addressed in the literature. Such potential errors are neglected in the analysis presented here.

Raabe and Wrenn proposed the use of regression analysis with total-alpha detection and many counting intervals to improve measurement precision (Ra69). Again this approach is not widely used, perhaps because of the complexity of data analysis: data from the three-count technique can be easily analyzed with a hand-held programmable calculator, while the regression analysis is easily accomplished only with the assistance of a microcomputer.

The use of alpha spectroscopy to separately count the decays from  $^{218}\text{Po}$  and  $^{214}\text{Po}$  during two count intervals has been widely investigated (Ma69, Jo74, Tr79, Na81). For fixed air-flow rates during sampling, this approach offers improvements in precision over the total alpha techniques. Again, though, this approach does not appear to be widely used.

Groer and his colleagues have developed a instrument which uses alpha spectroscopy and gross beta measurement during a single counting interval after sampling to measure concentrations of the three radon progeny (Gr73, Ke78). The advantage of this instrument is that precise



measurements can be made rapidly; its primary disadvantages are its complexity which results in high cost, and its bulk.

Optimizations described in this paper focus on the use of total-alpha three-count techniques for measuring radon progeny concentrations indoors. Both sequential and overlapped sampling and counting intervals are analyzed. For sequential intervals 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  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Bi}$ , respectively, relative to using a 35-minute measurement time with a two-minute delay. For overlapped sampling and counting intervals I show that one can achieve modest improvements in precision by relaxing some of the constraints Cliff applied in his analysis. I find that measurement sensitivity using overlapped intervals and 60-minute total measurement time is improved by factors of 5.5, 2 and 2.5 for  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ , respectively, relative to using sequential intervals and a 60-minute total measurement time.

#### 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  $i$ th counting interval;

$I$  is a  $1 \times 3$  matrix where  $I_j$  is the activity concentration of the  $j$ th radon decay product ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ , respectively);

$M$  is a  $1 \times 3$  matrix where  $M_j$  is the minimum measurable concentration of the  $j$ th 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  $1 \times 3$  matrix of activity ratios where  $R_j = I_j/I_1$ ;

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

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

$V$  is the sampling flow rate; and

$\eta$  is the average detection efficiency for  $^{218}\text{Po}$  and  $^{214}\text{Po}$  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  $3 \times 3$  matrix whose elements depend upon the sample and count-interval timing (see Appendix). This equation can be inverted to obtain

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

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

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

$$P = \frac{1}{\eta V} \sum_{i=1}^3 L_i C_i, \quad (3a)$$

where

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

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

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

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

where, by definition,

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

To choose among different count-interval timing sets, a single optimization parameter must be selected. In previous work,  $^{218}\text{Po}$  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

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

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

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.

For sequential sampling and counting intervals, 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 vector. 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 easier 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. For overlapped sampling and counting intervals the same general procedure was followed, however the beginning of the first count interval was fixed at zero and the sampling time was allowed to vary.

All of the optimization results reported in this note were computed with an activity ratio vector 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 vector; only for extreme disequilibrium does the optimal timing change significantly.

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

## RESULTS AND DISCUSSION

The effect on measurement precision of varying the total measurement time for the total-alpha three-count technique with sequential intervals 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 vector 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  $^{218}\text{Po}$  than for either  $^{214}\text{Pb}$  or  $^{214}\text{Bi}$  over the entire range of total measurement times considered. It is, perhaps, a surprising result that measurement precision for  $^{218}\text{Po}$ , 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  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  is more precisely known if longer measurement times are used, hence the counts detected due to the collection of  $^{218}\text{Po}$  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 products concentrations; this result is due to a

cancellation of errors, as noted by Busigin and Phillips (Bu80).

Results of a similar analysis for overlapped intervals are presented in Figure 2. As Cliff concluded, using overlapped intervals is seen to substantially improve measurement precision for  $^{218}\text{Po}$ . While these curves are less steep than those in Figure 1, the fractional improvements in increasing measurement time from 35 to 60 minutes are comparable. We also see in Figure 2 that while time sets recommended by Cliff can be substantially improved for measurement times of less than 40 minutes, the time sets he proposed for total times greater than 40 minutes are near optimal. The constraints Cliff imposed which were relaxed here are (1) fully coincident sampling period and first counting interval and (2) fixed one-minute delays between counting periods. Better precision is obtained by allowing the end of the sampling period to differ from the end of the first counting interval and by using delays between counting intervals of greater than 1 minute.

The effect of varying the sampling time on measurement precision for sequential intervals is shown in Figure 3. For a total measurement time of 60 minutes the MMC for  $^{218}\text{Po}$  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 for sequential intervals to use a

five-minute sampling time as recommended by Thomas (Th72). With overlapped intervals constraining the sampling time to 5 minutes results in a substantial loss of precision for each isotope; for example the minimum measurable concentrations for the timing (5, 0-7, 9-27, 38-60) and  $R_2:R_3 = 0.5:0.4$  are 3.4, 0.9 and 1.7 pCi/l compared to 2.0, 0.4, and 0.9 pCi/l for  $^{218}\text{Po}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Pb}$ , respectively, using the optimal timing of (16, 0-11, 17-34, 43-60).

For sequential intervals 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  $^{218}\text{Po}$ , 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 six time sets is presented in Table 1. The first three sets are constrained by the requirement of sequential sampling and counting periods. They are the timing recommended by Thomas and the optimized 60-minute measurement periods with one- and two-minute delays. The second three sets use overlapped sampling and counting periods. They are the 57-minute measurement timing proposed by Cliff and optimized 45- and 60-minute measurement periods. Using the constants in this table the radon progeny concentrations are calculated from equations (2) and (3a) and standard deviations in the measurements due to counting statistics are



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

$$\sigma_p = \frac{1}{nV} \left( \sum_{j=1}^3 L_j^2 C_j \right)^{1/2}. \quad (8b)$$

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

This result can be seen more clearly in Figures 5-7 where lines of equal MMCs are plotted in a triangular space whose points represent all sustainable radon progeny activity ratios. Figure 5 is plotted for the timing recommended by Thomas, Figure 6 is computed for the timing recommended in this work for a 60-minute period with a one-minute delay, and Figure 7 is based on a 60-minute period with overlapped sampling and counting intervals. Comparing these figures we see that (1) by extending the total measurement time from 35 to 60 minutes the MMCs are improved roughly by factors of 3 for  $^{218}\text{Po}$ , 7 for  $^{214}\text{Pb}$  and 4 for  $^{214}\text{Bi}$  for all activity ratios; and (2) by using overlapped sampling and counting intervals, the MMCs can be further improved by factors of 5.5 for  $^{218}\text{Po}$ , 2 for  $^{214}\text{Pb}$  and 2.5 for  $^{214}\text{Bi}$ . 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, Re83). In this region, only the measurement precision for  $^{218}\text{Po}$  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  $^{218}\text{Po}$  (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  $^{218}\text{Po}$  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 for radon progeny concentrations calculated for timing sets recommended by Thomas, by Busigin and Phillips, by Cliff, 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 with sequential intervals has substantially lower RSDs for all conditions. With  $\Delta I = 0\%$  the time sets utilizing overlapped intervals show substantial further improvement, particularly for  $^{218}\text{Po}$ . However,

with  $\Delta I = 5\%$  the differences in RSD for  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  between the 60-minute periods using sequential and overlapped intervals are small.

#### 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. Further improvements can be achieved by employing an instrument that permits simultaneous sampling and counting. In this case, the timing sequences optimized for 45- and 60-minute total measurement times are (12, 0-9, 13-27, 34-45) and (16, 0-11, 17-34, 43-60).

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APPENDIX: CALCULATING THE H-MATRIX ELEMENTS

For a timing sequence specified by  $(t_0, t_{1a}-t_{1b}, t_{2a}-t_{2b}, t_{3a}-t_{3b})$ , the H-matrix elements are given by

$$H_{i1} = \frac{\eta_1}{\eta} [G_{11}(t_{ib}) - G_{11}(t_{ia})] + \frac{\eta_3}{\eta} [G_{31}(t_{ib}) - G_{31}(t_{ia})] \quad i=1-3, \quad (A1a)$$

and

$$H_{ij} = \frac{\eta_3}{\eta} [G_{3j}(t_{ib}) - G_{3j}(t_{ia})], \quad i=1-3; j=2,3 \quad (A1b)$$

where  $\eta_1$  is the detection efficiency for  $^{218}\text{Po}$  decays,  $\eta_3$  is the detection efficiency for  $^{214}\text{Po}$  decays and  $\eta = (\eta_1 + \eta_3)/2$ . For the purposes of optimization I assume that  $\eta_1 = \eta_3$ . The functions  $G_{ij}(t)$  give the number of decays on the filter during the interval 0 to  $t$  of the  $i$ th decay product due to the sampling of one pCi/l of the  $j$ th decay product at a rate of one l/min:

For  $t \leq t_0$ ,

$$G_{11}(t) = \frac{2.22}{\lambda_1} \left( t - \frac{r_1(t)}{\lambda_1} \right), \quad (A2a)$$

$$G_{31}(t) = \frac{2.22}{\lambda_1} \left( t - \frac{f_{21}f_{31}}{\lambda_1} r_1(t) - \frac{f_{12}f_{32}}{\lambda_2} r_2(t) - \frac{f_{13}f_{23}}{\lambda_3} r_3(t) \right), \quad (A2b)$$



$$G_{32}(t) = \frac{2.22}{\lambda_2} \left( t - \frac{f_{32}}{\lambda_2} r_2(t) - \frac{f_{23}}{\lambda_3} r_3(t) \right), \quad (\text{A2c})$$

$$\text{and } G_{33}(t) = \frac{2.22}{\lambda_3} \left( t - \frac{r_3(t)}{\lambda_3} \right); \quad (\text{A2d})$$

for  $t \geq t_0$

$$G_{11}(t) = \frac{2.22}{\lambda_1} \left( t_0 - \frac{r_1(t_0)}{\lambda_1} s_1(t-t_0) \right), \quad (\text{A3a})$$

$$G_{31}(t) = \frac{2.22}{\lambda_1} \left( t_0 - \frac{f_{21}f_{31}}{\lambda_1} r_1(t_0) s_1(t-t_0) \right.$$

$$\left. - \frac{f_{12}f_{32}}{\lambda_2} r_2(t_0) s_2(t-t_0) - \frac{f_{13}f_{23}}{\lambda_3} r_3(t_0) s_3(t-t_0) \right), \quad (\text{A3b})$$

$$G_{32}(t) = \frac{2.22}{\lambda_2} \left( t_0 - \frac{f_{32}}{\lambda_2} r_2(t_0) s_2(t-t_0) - \frac{f_{23}}{\lambda_3} r_3(t_0) s_3(t-t_0) \right), \quad (\text{A3c})$$

and

$$G_{33}(t) = \frac{2.22}{\lambda_3} \left( t_0 - \frac{r_3(t_0)}{\lambda_3} s_3(t-t_0) \right). \quad (\text{A3d})$$

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

$$f_{ij} = \frac{\lambda_i}{\lambda_i - \lambda_j}, \quad (\text{A4a})$$

$$r_i(t) = 1 - e^{-\lambda_i t}, \text{ and} \quad (\text{A4b})$$

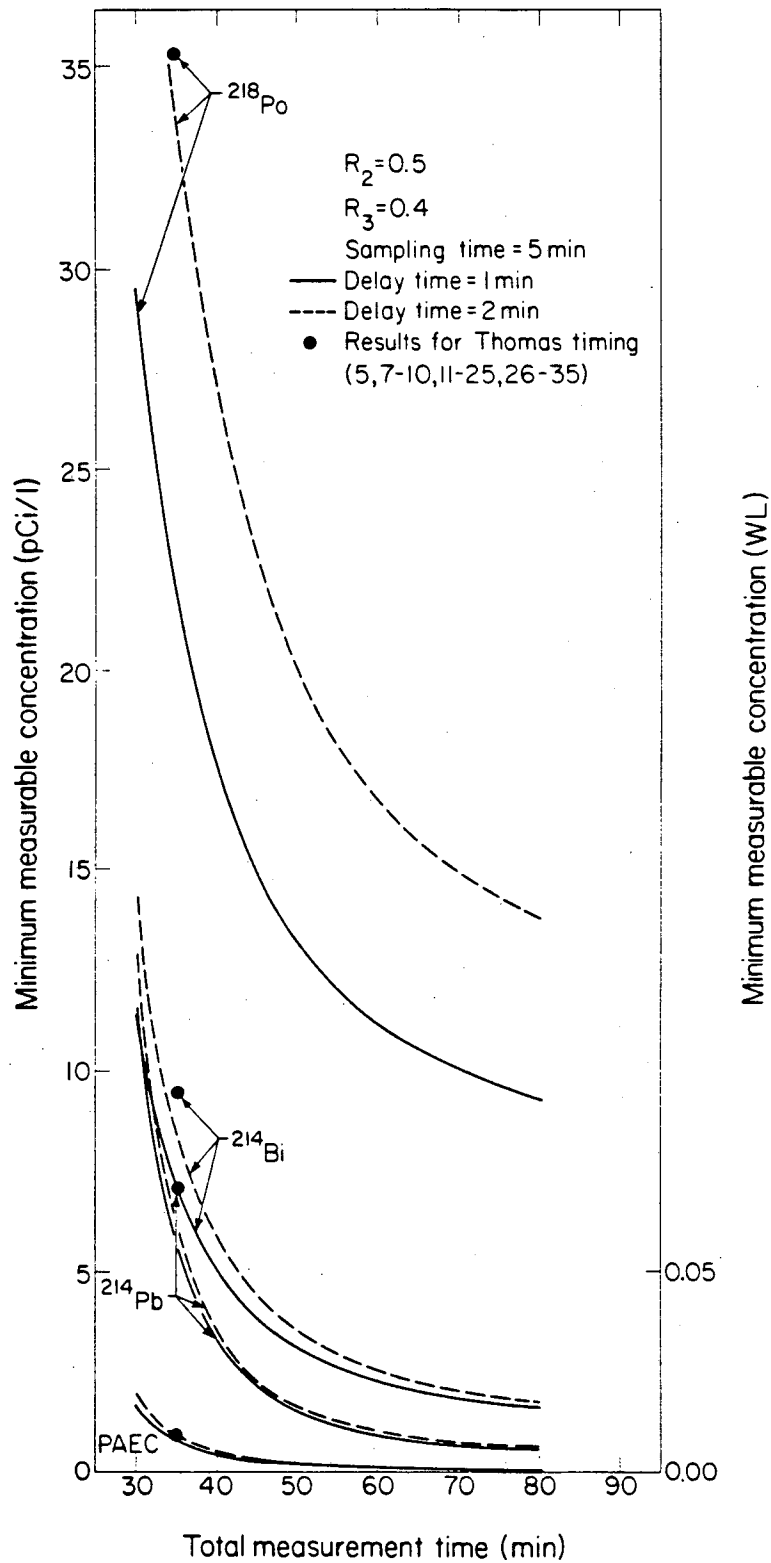
$$s_i(t-t_0) = e^{-\lambda_i(t-t_0)}. \quad (\text{A4c})$$

Table 1. Calculation constants and minimum measurable concentrations of radon progeny for the total-alpha three-count technique. The first three time sets listed are for systems which count after sampling is complete, while the second three are for systems in which the sampling and counting interval may be overlapped. 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 (min) [reference]	Calculation Matrix Elements					Minimum Measurable Concentrations				
	1	$K_{11}$	$K_{12}$ (pCi·dis <sup>-1</sup> ·min <sup>-1</sup> )	$K_{13}$	$L_1$ (WL·l·dis <sup>-1</sup> ·min <sup>-1</sup> )	$R_2:R_3$	<sup>218</sup> Po (pCi/l)	<sup>214</sup> Pb (pCi/l)	<sup>214</sup> Bi (pCi/l)	PAEC (WL)
(5, 7-10, 11-25, 26-35) [Th72]	1	0.16894	-0.08200	0.07753	$9.616 \times 10^{-5}$	1.0:1.0	68.7	6.7	7.8	0.0102
	2	0.00122	-0.02057	0.04909	$-6.499 \times 10^{-5}$	0.5:0.4	35.1	6.5	9.4	0.0097
	3	-0.02253	0.03318	-0.03771	$18.81 \times 10^{-5}$	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	$4.863 \times 10^{-5}$	1.0:1.0	32.0	1.0	2.1	0.0014
	2	-0.01237	-0.00264	0.01609	$0.222 \times 10^{-5}$	0.5:0.4	16.7	1.0	2.6	0.0014
	3	-0.00638	0.01756	-0.01500	$5.955 \times 10^{-5}$	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	$3.745 \times 10^{-5}$	1.0:1.0	20.9	0.9	1.9	0.0012
	2	-0.00904	-0.00374	0.01485	$0.420 \times 10^{-5}$	0.5:0.4	11.2	0.9	2.3	0.0012
	3	-0.00655	0.01689	-0.01258	$5.279 \times 10^{-5}$	0.3:0.1	6.5	0.9	4.8	0.0012
(15, 0-15, 16-36, 37-57) [C178]	1	0.012820	-0.011318	0.007904	$0.480 \times 10^{-5}$	1.0:1.0	4.3	0.5	0.9	0.0005
	2	-0.000414	-0.002552	0.005852	$-0.160 \times 10^{-5}$	0.5:0.4	2.2	0.5	1.2	0.0005
	3	-0.001691	0.006162	-0.004981	$1.923 \times 10^{-5}$	0.3:0.1	1.3	0.5	2.4	0.0004
(12, 0-9, 13-27, 34-45) [present work]	1	0.027917	-0.014191	0.011231	$1.287 \times 10^{-5}$	1.0:1.0	5.4	1.3	1.4	0.0014
	2	0.000722	-0.005384	0.013020	$-0.669 \times 10^{-5}$	0.5:0.4	3.0	1.2	1.7	0.0013
	3	-0.005241	0.009443	-0.008368	$4.637 \times 10^{-5}$	0.3:0.1	1.9	1.1	3.7	0.0012
(16, 0-11, 17-34, 43-60) [present work]	1	0.018855	-0.009364	0.006747	$0.612 \times 10^{-5}$	1.0:1.0	3.6	0.4	0.7	0.0004
	2	-0.000871	-0.002045	0.005873	$0.117 \times 10^{-5}$	0.5:0.4	2.0	0.4	0.9	0.0004
	3	-0.002383	0.005681	-0.004638	$1.943 \times 10^{-5}$	0.3:0.1	1.2	0.4	1.9	0.0004

Table 2. Relative standard deviations in measured radon progeny concentrations using six different count-interval time sets.  $\Delta I$  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.

Measurement Timing (minutes) [reference]	Radon Daughter Concentrations (pCi/l) $^{218}\text{Po}$ : $^{214}\text{Pb}$ : $^{214}\text{Bi}$	Relative Standard Deviation in Measured Concentration					
		$\Delta I = 0\%$			$\Delta I = 5\%$		
		$^{218}\text{Po}$	$^{214}\text{Pb}$	$^{214}\text{Bi}$	$^{218}\text{Po}$	$^{214}\text{Pb}$	$^{214}\text{Bi}$
5, 7-10, 11-25, 26-35 [Th72]	5.0:1.5:0.5	0.20	0.19	0.62	0.24	0.23	0.76
	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 [Bu80]	5.0:1.5:0.5	0.20	0.20	0.62	0.23	0.23	0.72
	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 [present work]	5.0:1.5:0.5	0.11	0.08	0.30	0.14	0.11	0.42
	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
15, 0-15, 16-36, 37-57 [Cl78]	5.0:1.5:0.5	0.05	0.05	0.22	0.11	0.11	0.44
	4.0:2.0:1.6	0.08	0.05	0.09	0.17	0.12	0.20
	2.0:2.0:2.0	0.15	0.05	0.07	0.35	0.12	0.17
12, 0-9, 13-27, 34-45 [present work]	5.0:1.5:0.5	0.06	0.09	0.28	0.11	0.13	0.44
	4.0:2.0:1.6	0.09	0.08	0.11	0.15	0.14	0.19
	2.0:2.0:2.0	0.17	0.08	0.08	0.30	0.15	0.17
16, 0-11, 17-34, 43-60 [present work]	5.0:1.5:0.5	0.05	0.05	0.20	0.10	0.10	0.38
	4.0:2.0:1.6	0.07	0.05	0.08	0.14	0.10	0.17
	2.0:2.0:2.0	0.14	0.05	0.06	0.28	0.11	0.15



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Figure 1 Optimized minimum measurable concentrations of radon progeny for the total-alpha three-count technique as a function of total measurement time, with the constraint that counting may not begin until sampling is complete. Sampling time is fixed at 5 minutes.

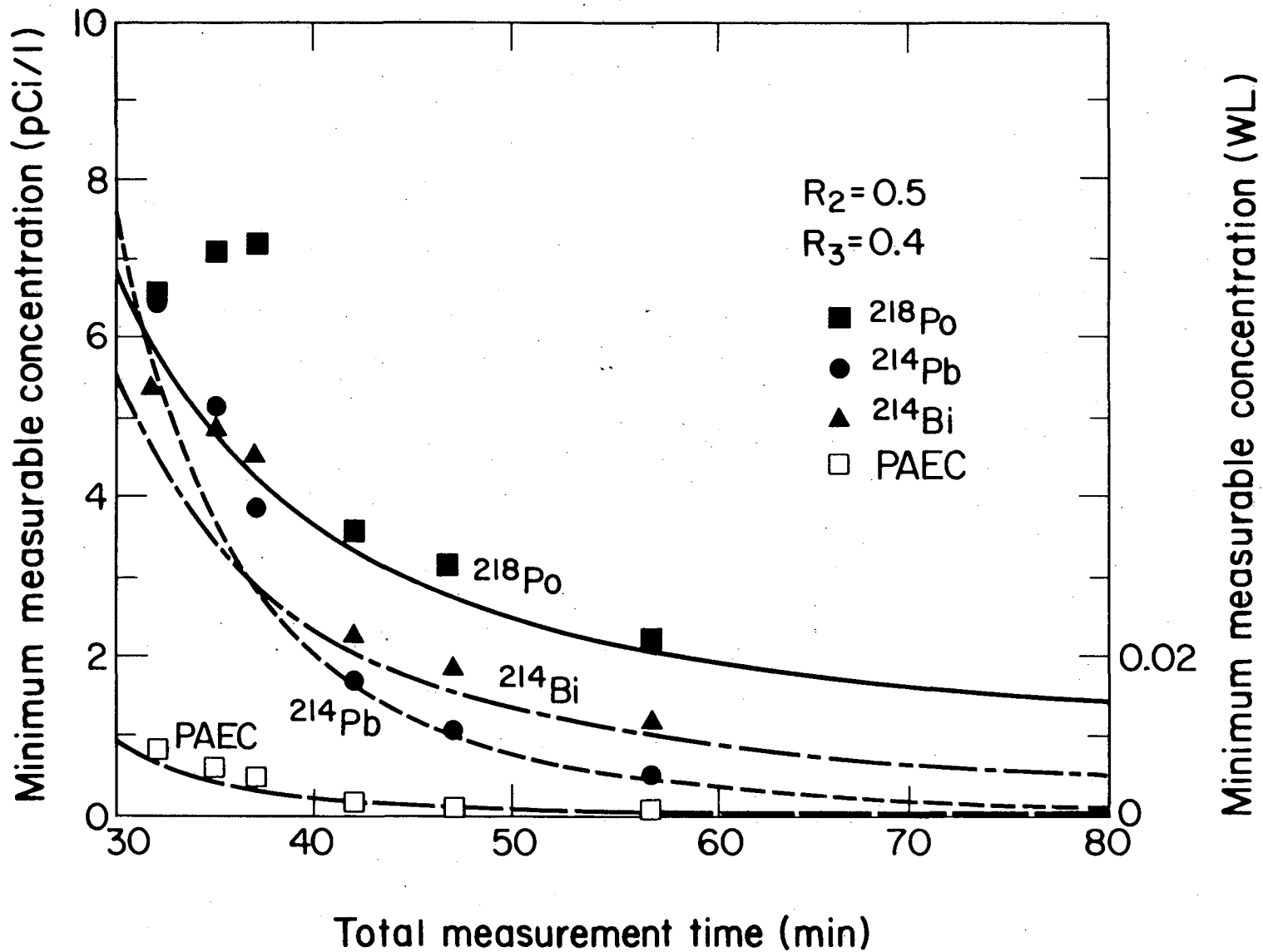
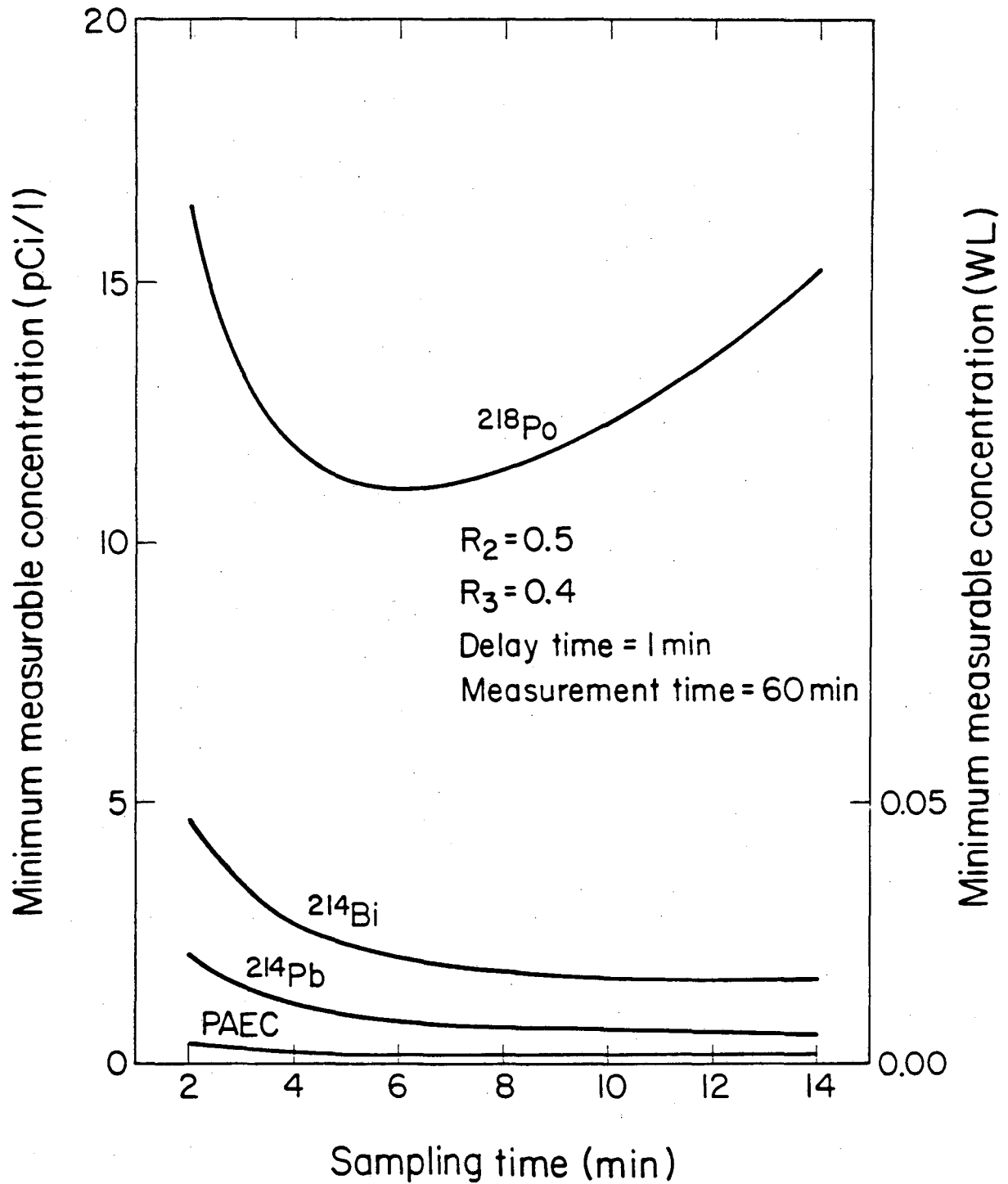


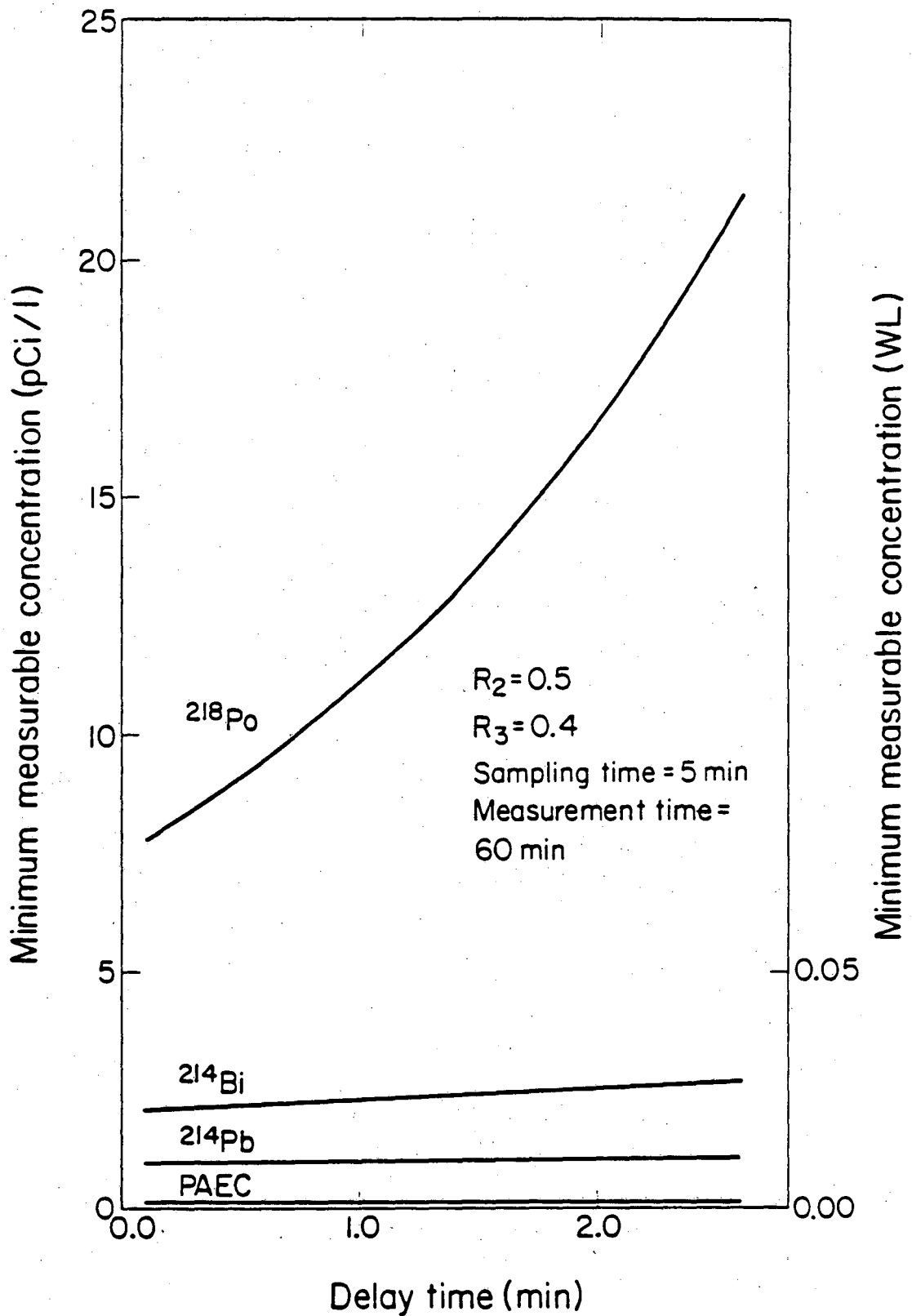
Figure 2 Optimized minimum measurable concentrations of radon progeny for the total-alpha three-count technique with counting permitted during sampling, and sampling time allowed to vary. The discrete points reflect the precision of using the time sets proposed by Cliff (C178).

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Figure 3 Optimized minimum measurable concentrations of radon progeny as a function of sampling time for sequential sampling and counting intervals.



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Figure 4 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.



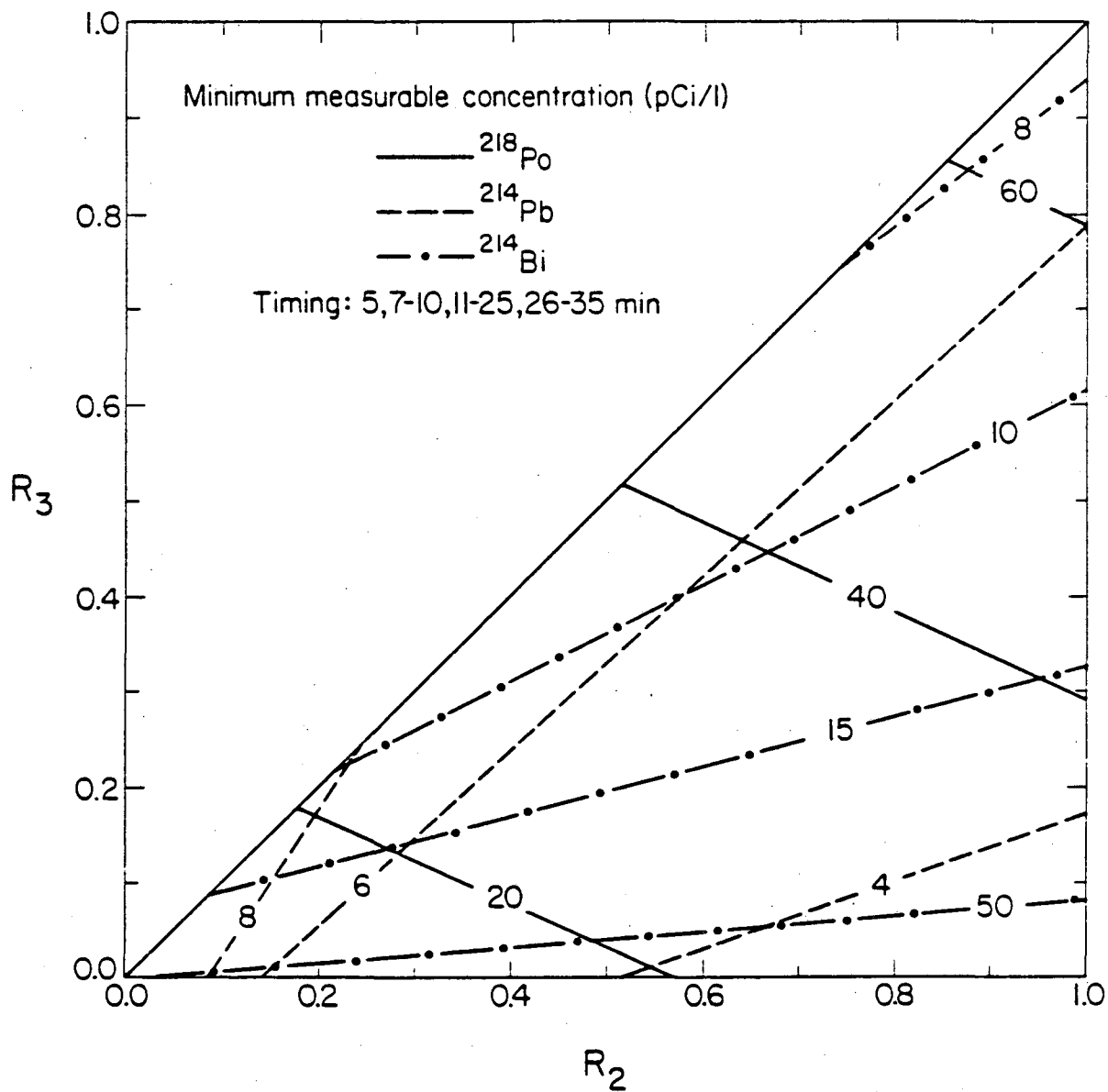
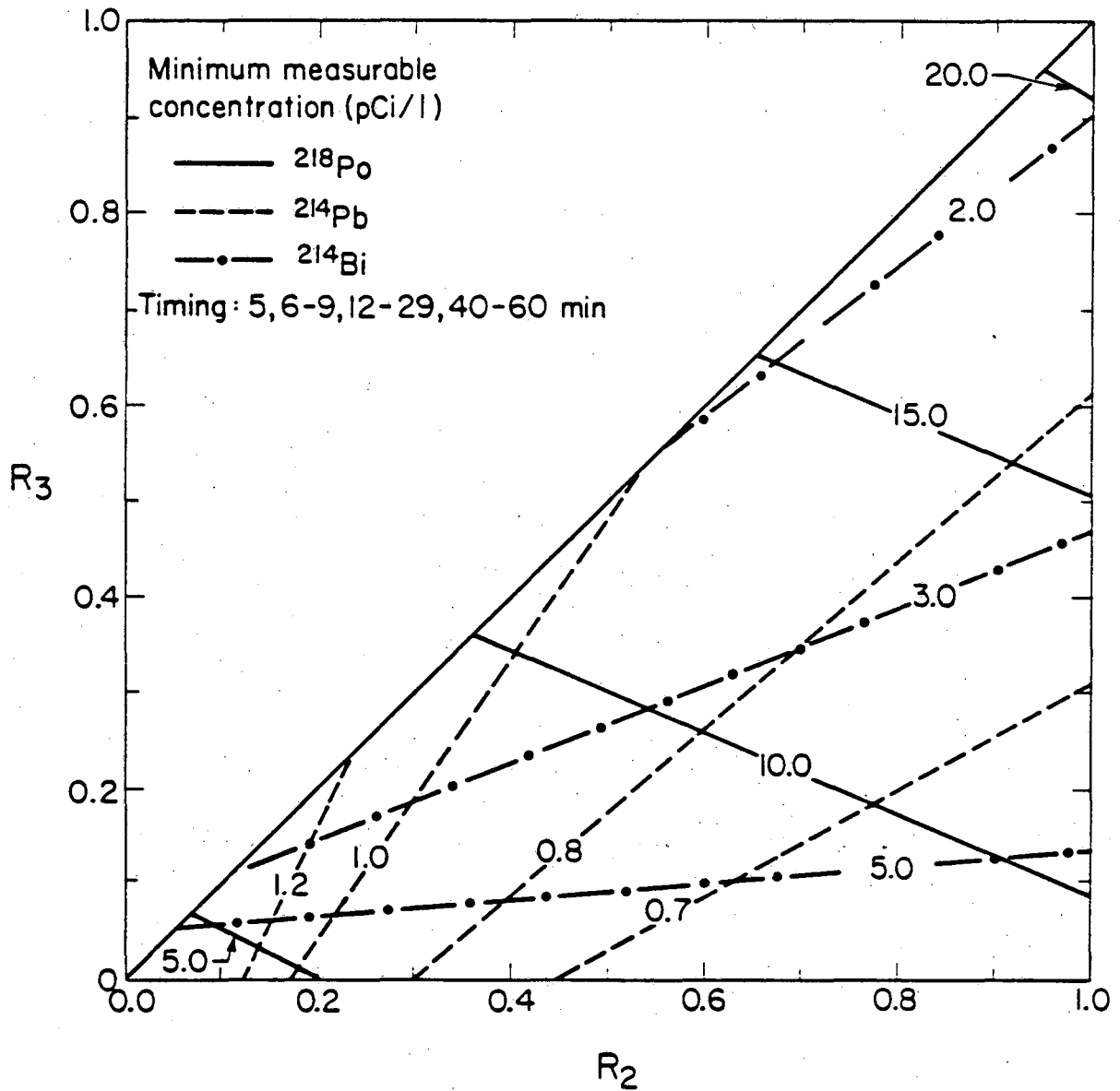


Figure 5 Isonpleths of minimum measurable concentrations of radon progeny as functions of activity ratios for timing recommended by Thomas (Th72).  $R_2$  and  $R_3$  are the activity ratios of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  respectively to  $^{218}\text{Po}$ .



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Figure 6 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.

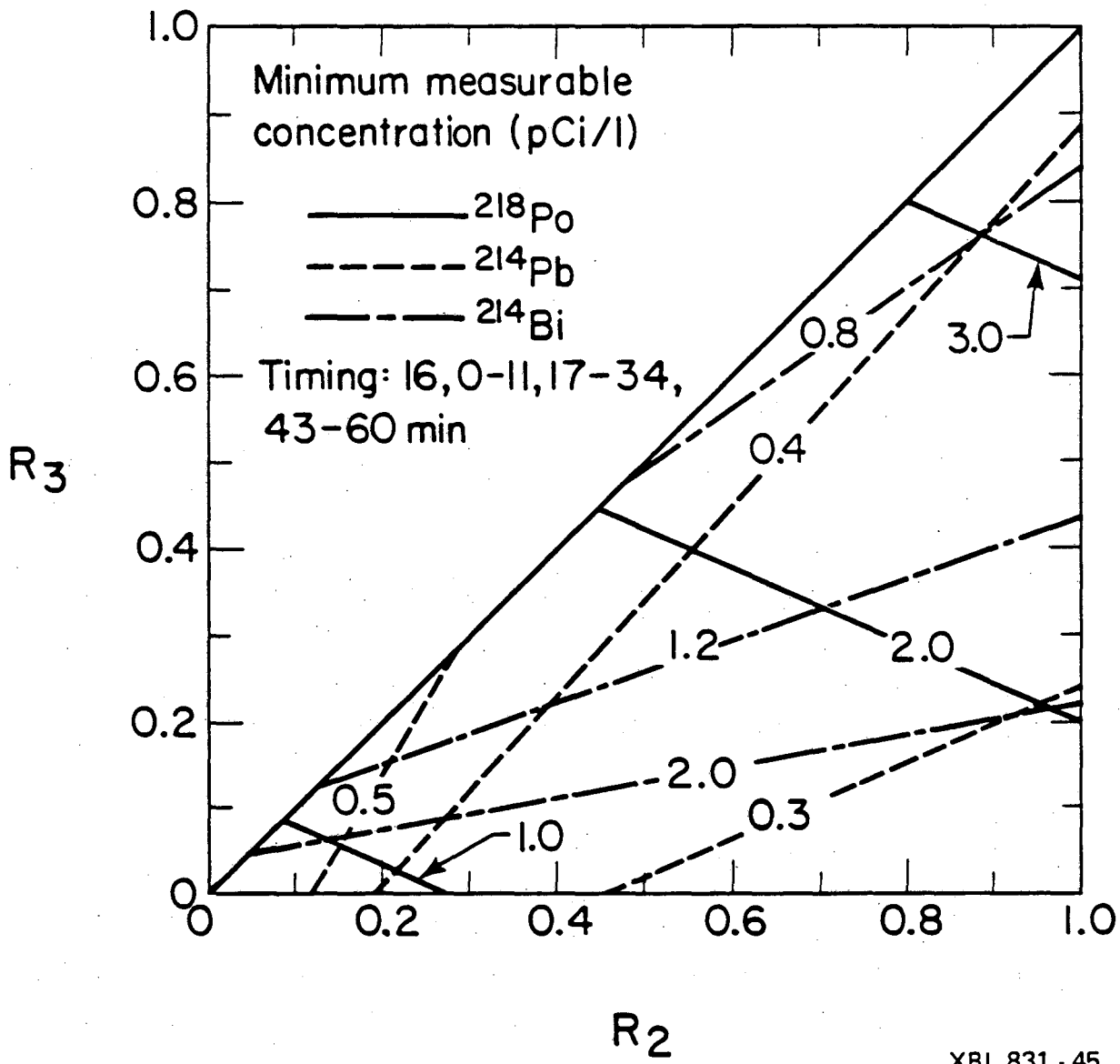


Figure 7 Isopleths of minimum measurable concentrations of radon progeny as functions of activity ratios for optimized 60-minute total measurement time in which the counting may be done while sampling.

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TECHNICAL INFORMATION DEPARTMENT  
LAWRENCE BERKELEY LABORATORY  
UNIVERSITY OF CALIFORNIA  
BERKELEY, CALIFORNIA 94720