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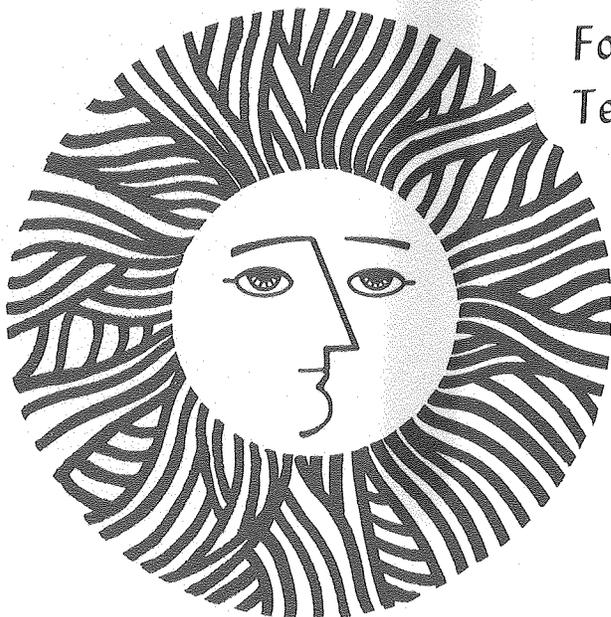
A RAPID SPECTROSCOPIC TECHNIQUE FOR DETERMINING
THE POTENTIAL ALPHA ENERGY CONCENTRATION OF
RADON DECAY PRODUCTS

K.L. Revzan and W.W. Nazaroff

July 1981

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OF RADON DECAY PRODUCTS

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Abstract

We consider the application of alpha spectroscopy to the rapid determination of the potential alpha energy concentration (PAEC) of radon decay products indoors. Two count totals are obtained after a single counting period. The PAEC is then estimated by a linear combination of the count totals, the two coefficients being determined by analysis of the dependence of the statistical and procedural errors on the equilibrium conditions and the sampling, delay, and counting times. For a total measurement time of 11 min, the procedural error is unlikely to exceed 20% for equilibrium conditions commonly found indoors; the statistical error is less than 20% at a PAEC of 0.005 WL, assuming a product of detector efficiency and flow rate of at least 1.0 l/min. An analysis is made of techniques based on a total alpha count, and the results are compared with those obtained with the rapid spectroscopic technique; the latter is clearly preferable when the measurement time does not exceed 15 min.

Keywords: alpha spectroscopy, indoor air pollution, instrumentation, measurement methods, radon daughters.

INTRODUCTION

The potential alpha energy concentration (PAEC) is the usual measure of the health hazards of airborne decay products ("daughters") of ^{222}Rn ("radon"). Determination of the PAEC requires the measurement of the airborne concentrations of the three decay products ^{218}Po , ^{214}Pb , and ^{214}Bi , which may be carried out by collection of a filtered air sample and subsequent counting of the activity present on the filter. Because ^{218}Po and the fourth decay product, ^{214}Po , emit alpha particles of different energies, and because the decay of the latter may be taken as occurring simultaneously with that of ^{214}Bi , alpha spectroscopy provides, for each counting period, two count totals that may be used to determine the concentrations of the three decay products of interest. Two counting periods are then necessary for a complete determination of the PAEC. The alternatives to alpha spectroscopy are a total alpha measurement with three counting intervals and use of alpha and beta spectroscopy with a single counting interval. All of these techniques require either a long measurement time or a complex instrument, either of which may be disadvantageous under given circumstances. It is common, therefore, to estimate the PAEC on the basis of an incomplete measurement.

The predominant estimation technique, that of Kusnetz (Ku56), calls for the counting of the total alpha activity on a filter during a single interval, which begins some time after the end of sampling. The sampling, delay, and counting intervals are at the disposal of the experimenter, subject to such limits on total measurement time as he may choose to impose. The resulting count total is multiplied by a conversion factor, which depends on the three intervals, to arrive at the

estimated PAEC. Since the count total and the actual PAEC are different linear combinations of the concentrations of the three radon daughters, there is an inherent "procedural" error in the estimation, except at equilibrium conditions for which the linear combinations differ by a constant. By adjusting the measurement intervals to the equilibrium conditions of interest, an optimum value of the conversion factor can, in principle, be found.

Satisfactory results using approximations such as the Kusnetz method require a fairly long total measurement time, even if the range of equilibrium conditions is restricted. If sensitivity is to be maximized, the portion of the measurement time taken up by the delay between sampling and counting must be minimized. A short delay time does not allow ^{218}Po to decay significantly before counting begins and, therefore, leads to an exaggeration of the contribution of this daughter to the estimated PAEC, increasing the procedural error. Where high sensitivity is not required, the delay time may be increased to the point that this source of error becomes relatively unimportant; in this case the Kusnetz method will prove satisfactory.

The low activity levels generally encountered indoors require the highest possible sensitivity consistent with an acceptably short measurement time and a satisfactorily low procedural error over a broad range of equilibrium conditions. It is in the satisfaction of these often conflicting requirements that a rapid spectroscopic technique offers significant advantages over total alpha methods.

As we have seen, alpha spectroscopy requires two counting periods for complete determination of the PAEC. From a single period we obtain the concentration of ^{218}Po and a count total which depends on this

concentration and those of the two succeeding daughters. It has been demonstrated by Schiager (Sc77) that, under certain circumstances, a weighted combination of these two totals may be used to estimate the PAEC; this method will be shown to be equivalent to the assumption of a relationship among the actual concentrations of the three daughters. If equilibrium conditions are such that this assumption holds approximately, an accurate estimate will be obtained. The method of estimating PAEC using two count totals obtained after a single counting interval will be referred to as the rapid spectroscopic technique.

In the following section, we derive the equations showing the dependence of the procedural and statistical errors of the rapid spectroscopic technique on the three measurement intervals and the daughter equilibrium ratios. Subsequently, a simple model of daughter behavior is used to predict the equilibrium ratios that may be expected to arise under conditions prevailing indoors; the values obtained are used to optimize the technique for these conditions. In the same section, we show that data obtained in one house using the complete (two counting interval) spectroscopic technique generally falls in the range predicted by the model and, further, that the rapid technique provides a good estimate of the PAEC. Finally, we compare the rapid technique with total alpha methods. The former will be seen to be more accurate, particularly when the measurement time is limited. The knowledge of the concentration of ^{218}Po provided by the rapid spectroscopic technique is an added advantage when there is an interest in prevailing equilibrium conditions.

THEORY OF THE RAPID SPECTROSCOPIC TECHNIQUE

The alpha spectrum of radon daughters has two peaks, arising from the 6.0 and 7.7 MeV decays of ^{218}Po and ^{214}Po , respectively. Assuming the upper portion of the spectrum to be divided into two channels, corresponding to the two peaks, energy degradation due to instrumental imperfections produces a broadening of the peaks that is sufficient to produce a small overlap of the upper peak into the lower channel, typical values being less than 5%. When two counting intervals are used, alpha spectroscopy produces four count totals, of which only three are needed to determine the PAEC, making the remaining value available for the calculation of the overlap factor (Na80). This factor may be determined to a sufficient degree of accuracy for a given instrument by taking the average of the results of a number of measurements.

Given a knowledge of the overlap and the background level, the corrected count totals obtained in the two channels are:

$$N_1 = \bar{N}_1 - \theta \bar{N}_2; \quad N_2 = (1 + \theta) \bar{N}_2, \quad (1)$$

where the subscripts 1 and 2 refer to the lower and upper channels, respectively, \bar{N}_n is the actual number of counts in the nth channel, corrected only for background, and θ is the overlap factor, i.e., the fraction of ^{214}Po counts appearing in the lower channel. The relationship among the corrected counts, the timing intervals, and the airborne activity concentrations may then be written:

$$N_n = \sum_{m=1}^3 \eta f a_{nm} I_m \quad [n = 1,2] \quad (2)$$

where I_m is the activity concentration of the mth decay product, η is

the efficiency of the detector, and f is the flow rate of the sampling pump. The half-lives of the first three daughters are 3.05 min, 26.8 min, and 19.7 min, respectively. The decay of the fourth product, because of its very short half-life (0.1 msec), is treated as if it were simultaneous with that of the third. We assume that the efficiency of detection is independent of energy. Let

$$S_i = 1 - \exp(-\lambda_i T_s)$$

$$D_i = \exp(-\lambda_i T_d)$$

$$C_i = 1 - \exp(-\lambda_i T_c) \quad (3)$$

where λ_i is the decay constant of the i th decay product (i.e., $0.693 T_i^{-1}$, where T_i is the i th half-life), and T_s , T_d , and T_c are, respectively, the sampling, delay, and counting times. Then it can be shown that

$$a_{11} = k S_1 D_1 C_1 / \lambda_1^2; \quad a_{12} = a_{13} = 0$$

$$a_{21} = k \left\{ \lambda_2 \lambda_3 S_1 D_1 C_1 / [\lambda_1^2 (\lambda_2 - \lambda_1) (\lambda_3 - \lambda_1)] \right.$$

$$\left. + \lambda_1 \lambda_3 S_2 D_2 C_2 / [\lambda_1 \lambda_2 (\lambda_1 - \lambda_2) (\lambda_3 - \lambda_2)] \right\}$$

$$+ \lambda_1 \lambda_2 S_3 D_3 C_3 / [\lambda_3 \lambda_1 (\lambda_1 - \lambda_3) (\lambda_2 - \lambda_3)] \Big\}$$

$$a_{22} = k \left\{ \lambda_3 S_2 D_2 C_2 / [\lambda_2^2 (\lambda_3 - \lambda_2)] + \lambda_2 S_3 D_3 C_3 / [\lambda_3 \lambda_2 (\lambda_2 - \lambda_3)] \right\}$$

$$a_{23} = k S_3 D_3 C_3 / \lambda_3^2 \quad (4)$$

where k is a conversion factor determined by the choice of units. (If I_n is taken in pCi/l, all λ_n in min^{-1} , and f in 1/min, then $k = 2.22 \text{ min}^{-1} \cdot \text{pCi}^{-1}$, and the units of a_{nm} are min/pCi .)

Since the rapid technique provides a determination of I_1 , it is convenient to write (2) in the form:

$$N_1 = \eta f a_{11} I_1 \quad (5)$$

$$N_2 = \eta f (a_{21} + a_{22} R_2 + a_{23} R_3) I_1, \quad (6)$$

where

$$R_2 = I_2/I_1; \quad R_3 = I_3/I_1 \quad (7)$$

Similarly, we may write the PAEC as:

$$W = (c_1 + c_2 R_2 + c_3 R_3) I_1 \quad (8)$$

where, if W is to be in WL, $c_1 = 0.00104$, $c_2 = 0.00507$, and $c_3 = 0.00373$, assuming I_1 to be in pCi/l.

It is clear that R_2 and R_3 , which are necessary to a determination of W , cannot be found from (6). If, however, a theoretical or empirical relationship between the two ratios could be established, this relationship and equations (5) and (6) would be sufficient to determine W . Suppose now that the probability of occurrence of various values of the two ratios were known. It would then be possible to define a curve that passed through the region of high probability in R_2 - R_3 space. If we were then to estimate W using this relationship, we would find the estimate to be correct in situations where the actual values of R_2 and R_3 fell on the curve, while in other situations there would be a procedural error that increases with the distance of R_2 and R_3 from the curve. If the points of high probability clustered, it would be possible to estimate W in such a way that a low procedural error becomes highly likely.

The optimal curve of zero procedural error depends on the conditions under which the estimation technique is to be used. The simplest of all choices, and one that will be found to be satisfactory in practice, is a straight line; this choice, which is implicit in the work of Schiager (Sc77), leads to an estimate of the PAEC that is a linear combination of the two count totals obtained from a single counting period. Let us, then, assume the following relationship between R_2 and R_3 :

$$R_3 = m(0) R_2 + b(0). \quad (9)$$

The arguments of the slope, m , and the y -intercept, b , indicate that actual values of R_2 and R_3 on this line yield an estimate of W that is free of procedural error. Eliminating I_1 , R_2 , and R_3 with (5), (6), and (8) gives the estimated PAEC, which we shall call w , i.e.,

$$w = (A N_1 + B N_2) / (\eta f), \quad (10)$$

where

$$B = (c_2 + m(0) c_3) / (a_{22} + m(0) a_{23}) \quad (11)$$

and

$$A = [c_1 + b(0) c_3 - (a_{21} + b(0) a_{23}) B] / a_{11} \quad (12)$$

The choice of estimation coefficients, A and B, depends on the choice of the slope and intercept of the line of zero procedural error and on the three timing intervals that determine the coefficients a_{nm} . We might, alternatively, have begun with (10), with A and B unknown, and demonstrated that the procedural error vanishes when R_2 and R_3 satisfy (9); $m(0)$ and $b(0)$ may be expressed in terms of A and B as follows:

$$m(0) = (c_2 - a_{22} B) / (a_{23} B - c_3) \quad (13)$$

$$b(0) = (c_1 - a_{21} B - a_{11} A) / (a_{23} B - c_3) \quad (14)$$

Let the procedural error be designated e_p , i.e.,

$$e_p = (w - W) / W; \quad (15)$$

then $w = W(1 + e_p)$ and, from (8) and (10),

$$(AN_1 + BN_2) / (\eta f) = (1 + e_p) (c_1 + c_2 R_2 + c_3 R_3) I_1. \quad (16)$$

Using (5), (6), and (16) to solve for R_3 , we find a linear relationship

of the same sort as (9), i.e.,

$$R_3 = m(e_p) R_2 + b(e_p) \quad (17)$$

where

$$m(e_p) = [c_2(1 + e_p) - B a_{22}] / [B a_{23} - (1 + e_p)c_3] \quad (18)$$

$$b(e_p) = [c_1(1 + e_p) - A a_{11} - B a_{21}] / [B a_{23} - (1 + e_p)c_3]. \quad (19)$$

The curves of constant procedural error are lines whose slopes and intercepts are given by (18) and (19), respectively.

We shall be interested in determining the region of the positive R_2 - R_3 space for which the procedural error is less than some selected value, ϵ . This "region of ϵ error" is simply that part of the space which lies between the line defined by $m(\epsilon)$ and $b(\epsilon)$ and that defined by $m(-\epsilon)$ and $b(-\epsilon)$. From (18) we see that the slopes cannot be chosen independently, depending as they do upon a single parameter; similarly, fixing one intercept determines the other. Ideally, we would calculate the area of the positive R_2 - R_3 plane for which $|e_p| < \epsilon$ and maximize it with respect to the three timing intervals, assuming a constrained total measurement time. The two estimation coefficients would then be determined.

Not all regions of the R_2 - R_3 plane are of equal importance, however, so that a determination of the maximum area lying within the limits on procedural error might well produce an estimation technique that is valid for daughter equilibrium ratios that rarely, if ever, occur in practice. If we had sufficient experience on which to base an estimate

of the probability of occurrence of given values of R_2 and R_3 , we could determine the probability within any region of ϵ error and, at least in principle, analytically determine the optimum region for a given ϵ . Sufficient data for such a procedure do not exist at present, and, in selecting regions of interest in the R_2 - R_3 plane, we are compelled to rely on theory, which is in an embryonic state, and on such limited data as are available. A further problem is raised by the interrelationship between the procedural and statistical errors, which may necessitate a compromise in the selection of the timing intervals.

It is simplest to determine the effect of changes in the timing intervals on the procedural error by finding the regions of ϵ error for a number of values of ϵ and for a number of choices of timing periods. We assume, for convenience, that the line of zero procedural error passes through the origin; lines whose intercepts with the R_3 -axis are greater than 0 are of little interest to us, since normally $R_3 < R_2$, while lines that intersect the R_2 -axis at points distant from the origin are likely to pass through regions of lesser interest than that chosen. By taking a few examples, one can demonstrate that, for a given absolute procedural error, ϵ , the slopes $m(\epsilon)$ and $m(-\epsilon)$ increase and decrease, respectively, with increasing ϵ , as do the intercepts $b(\epsilon)$ and $b(-\epsilon)$. For any ϵ , and for any chosen slope, $m(0)$, the area of the region of ϵ error is largest when the sampling and counting times are equal, although the effect of a departure from this condition is quite small.

Increasing the delay time increases the area of the region of ϵ error somewhat. When the total measurement time is short, however, the effect of a change in T_d is small. When, for example, $T_s + T_d + T_c = 11$ min, $T_s = T_c$, and $m(0) = 0.75$, a delay time of 1 min gives, for $\epsilon = 0.1$

(i.e., a procedural error of 10%): $m(\leftarrow) = 0.94$, $b(\leftarrow) = 0.025$, $m(-\leftarrow) = 0.59$, and $b(-\leftarrow) = -0.021$. When the delay time is increased to 2 min, all other factors remaining the same, we find $m(\leftarrow) = 0.95$, $b(\leftarrow) = 0.026$, $m(-\leftarrow) = 0.58$, and $b(-\leftarrow) = -0.022$. The effect may, perhaps, be made clearer by considering the change in the procedural error under some likely equilibrium conditions distant enough from the line of zero procedural error to be illustrative. If the actual equilibrium ratios are $R_2 = R_3 = 0.5$, delay times of 1 and 2 min give, under the above conditions, procedural errors of 9.3% and 8.1%, respectively. When measurement times are longer, the effect of comparable changes in T_d becomes smaller.

The percentage statistical error, e_s , in the estimation of the PAEC is, from (10):

$$e_s = \frac{\sqrt{A^2 N_1 + B^2 N_2}}{A N_1 + B N_2}$$

$$= \frac{\sqrt{A^2 a_{11} + b^2 (a_{21} + a_{22} R_2 + a_{23} R_3)}}{[A a_{11} + B (a_{21} + a_{22} R_2 + a_{23} R_3)] \sqrt{\eta f I_1}} \quad (20)$$

The statistical error decreases with increasing N_1 and N_2 . A simple calculation shows that both counts are maximum when, for a fixed total measurement time, $T_s = T_c$. Since this condition on T_s and T_c also maximizes the region of \leftarrow error, we may assume, throughout what follows, that the sampling and counting times are equal.

The effect on the statistical error of changes in the delay time is seen most easily by examining Fig. 1. All of the coefficients, a_{mn} ,

decrease monotonically with T_d , once again assuming a fixed measurement time, so that both count totals are greatest when T_d is a minimum. Hence the statistical error, in contrast to the procedural, decreases with increasing T_d . As an example, let us again take $\epsilon = 0.1$, $m(0) = 0.75$, $b(0) = 0$, $T_s = T_c = 5$ min, and select values of R_2 and R_3 on the line of zero procedural error, say $R_2 = 0.5$, $R_3 = 0.375$. From (20) we find that $e_s \sqrt{\eta f I_1}$ is 0.179 when T_d is 1 min, and 0.199 when T_d is 2 min, the activity concentration being taken in pCi/l. When high sensitivity is a consideration, this difference is more significant than the relatively minor increases in procedural error resulting from the same change in T_d . Accordingly, at short measurement times, the delay time should be reduced to the practical minimum whenever conditions are such that the statistical error is likely to be significant. Under other circumstances, the choice of delay time may be made on the basis of appropriate calculations.

CHOICE OF ESTIMATION COEFFICIENTS

A simple model of the behavior of radioactive decay products (Ja72;Po78) provides the following description of the relationship among the activities of airborne concentrations of the members of the radon decay chain: It is assumed that the decay products are distributed bimodally in size, those in the smaller mode being referred to as "free" or "unattached" and those in the larger as "attached," the attachment being to atmospheric particulates. Attachment takes place at a rate λ_a , which is assumed not to vary with the product being considered. Activity deposits upon surfaces at rates λ_d^f for the free mode and λ_d^a for the attached, the rates again being identical for the three decay products

of interest. Upon decaying, molecules leave the surfaces of the particles to which they are attached at a rate $r_n \lambda_n$, where λ_n is the decay constant of the nth series member and r_n represents the fraction of the attached molecules that recoil. It is not difficult to show that, in any interior whose ventilation rate is λ_v , the steady-state activity concentration of the nth member of the chain is

$$I_n^f = \frac{\lambda_{n-1} I_{n-1}^f + \lambda_{n-1} r_{n-1} I_{n-1}^a}{\lambda_n + \lambda_v + \lambda_d^f + \lambda_a} \quad (21)$$

for the free mode, and

$$I_n^a = \frac{\lambda_{n-1} (1 - r_{n-1}) I_{n-1}^a + \lambda_a I_n^f}{\lambda_n + \lambda_v + \lambda_d^a} \quad (22)$$

for the attached. Here I_0 is the activity concentration of radon and I_n that of the nth decay product. The ventilation rate and indoor levels are assumed to be such as to make the influence of exterior concentrations of radon and its decay products negligible, the usual situation.

If we again treat the decay of the fourth product as if it were simultaneous with that of the third, we may solve (21) and (22) for the daughter equilibrium ratios R_2 and R_3 . If R_2 and R_3 are then calculated over ranges of values of the several rates that are appropriate to the conditions of interest, we can identify the region of R_2 - R_3 space for which, based on this model, the rapid estimation procedure is required to be reasonably accurate. Optimization of the procedure may follow.

We consider attachment rates, λ_a , from 2.5 to 250 hr^{-1} (Kr79), the middle of this range being the rate corresponding to a particle concen-

tration of $\sim 10^4 \text{ cm}^{-3}$, which is typical of the indoor environment (Si78). On the basis of the work of Sehmel (Se71) which, although more appropriate to mines than to houses, seems to provide the best available model, we consider the deposition rate of the attached fraction to be $\leq 0.1 \text{ hr}^{-1}$, which is sufficiently small in comparison with the other rates as to have a negligible influence on the concentrations. The deposition rate of the unattached fraction may be as much as three orders of magnitude greater than that of the attached fraction because of the increasing effect of molecular diffusion as the size of the particle diminishes (Se71); we therefore consider values of λ_d^f ranging from 1 to 100 hr^{-1} . The recoil fraction, r_1 , of ^{218}Po will be taken as 0.83 (Me76). Those of the beta-emitters ^{214}Pb and ^{214}Bi will be considered negligible, i.e., $r_2 = r_3 = 0$. The ventilation rates of interest here are those typical of closed residences. Although these are usually 1 hr^{-1} or less, we have, for the sake of completeness, considered ventilation rates from 0.5 to 3.0 hr^{-1} .

The ratios R_2 and R_3 predicted by the model under the above assumptions are shown in Fig. 2, 3, and 4, where we have superimposed lines of constant procedural error, as explained below. It is noteworthy that ratios in the lower right hand side of the figures do not appear. (Indeed, they do not appear even when the range of the rates is greatly extended.) Measurements made in a tightly constructed house at ventilation rates ranging from 0.07 hr^{-1} to 0.8 hr^{-1} produce similar results, as shown in Fig. 5 where, again, lines of constant procedural error have been superimposed (Na81).* The area of the R_2 - R_3 plane for which an accurate estimation of the PAEC is most desirable is made quite clear by

the figures.

The curves of constant procedural error introduced in the previous section are straight lines, the slopes of which increase with the value of the error. Because daughter equilibrium ratios near the line defined by $R_3 = R_2$ are likely to occur, and because ratios for which $R_3 > R_2$ are unlikely, it is desirable to choose some maximum allowable procedural error, say ϵ , and to bring the line of constant error ϵ into coincidence with the line defined by $R_3 = R_2$. (This is a change from our previous convention of taking the line of zero procedural error as passing through the origin.) If ϵ is well chosen, the estimate of the PAEC will ordinarily be within ϵ of the actual value. Taking $m(\epsilon) = 1$ in (18) and $b(\epsilon) = 0$ in (19) yields the estimation coefficients, i.e.,

$$B = \frac{(1 + \epsilon) (c_2 + c_3)}{a_{22} + a_{23}} \quad (23)$$

$$A = \frac{(1 + \epsilon) c_1 - a_{21} B}{a_{11}} \quad (24)$$

To determine these coefficients, it is necessary to choose ϵ and the three timing intervals, T_s , T_d , and T_c . We shall assume that $T_s = T_c$ and that $T_d = 1$ min, which is a practical lower limit. The accuracy of the estimation for a given set of parameters may be most usefully expressed in terms of some measure of the equilibrium conditions for which $|e_p|$ is less than ϵ , and in terms of some characteristic statistical error. An indication of the region of the R_2 - R_3 plane for which $-\epsilon$

*Most of the points on this figure for which $R_3 > R_2$ may be accounted for by the relatively large experimental errors resulting from the low activities obtaining.

$\leq e_p \leq \epsilon$ is given by the slope of the line of constant procedural error $-\epsilon$, i.e., $m(-\epsilon)$. The intercept of this line is always negative and always decreases with the slope, so that the area of the plane bounded by the lines of $e_p = \pm\epsilon$ increases as the slope decreases. The characteristic statistical error will be found from (20), with $R_2 = 0.5$ and $R_3 = m(0) R_2 + b(0)$, i.e., with the equilibrium ratios on the line of zero procedural error. Table 1 provides values of the estimation coefficients A and B (in WL·l/min), the slopes and intercepts $m(0)$, $b(0)$, $m(-\epsilon)$, and $b(-\epsilon)$, and the characteristic statistical error e_s , for several combinations of ϵ , T_s , and T_d .

A choice of $\epsilon = 20\%$ includes the region of greatest physical interest with some margin for the occurrence of unusual values of the model parameters. Under these assumptions, the coefficients A and B are 7.25×10^{-5} WL·l/min and 1.93×10^{-4} WL·l/min, respectively. The lines of constant procedural error are depicted in Fig. 2-4, where they are superimposed upon the daughter equilibrium ratios obtained from the model, and in Fig. 5, where they are superimposed upon experimental data. In most cases of interest, the procedural error is not greater than 20%. The parameters were chosen so that the overestimate could not exceed 20%; under no circumstance does the underestimate exceed 50%. Lines of constant random error at a product of flow rate, detector efficiency, and ^{218}Po activity concentration of 1 pCi·l/min are shown in Fig. 6. From this figure one can determine the largest value of the PAEC for which a given statistical error occurs. At $e_s = 20\%$ this value is ~0.005 WL.

A test of the rapid spectroscopic estimation technique was made during the course of an experiment in which the attachment rate, ini-

tially quite low, gradually increased and subsequently declined somewhat. Under these circumstances, both R_2 and R_3 should be low initially and should, in turn, rise and subsequently decline. This prediction is supported by the data, which yield appropriate values of the estimated PAEC. The results are shown in Table 2, which uses count totals corrected for overlap (the average value of θ being 0.021) and a product of detector efficiency and flow rate of 4.04 l/min. The actual PAEC is here taken to be the value obtained through the use of the two-count-interval spectroscopic technique. The difference between the results produced by the two techniques does not exceed 19%. When the correction for overlap is omitted for the sake of simplicity, the procedural error changes by approximately 10% in the direction of overestimation; in this case the largest error is 20%.

An examination of Fig. 2-4 reveals that, because of our adjustment of the parameters of the approximation technique, the values of R_2 and R_3 predicted by the model, even when the ventilation rate is as high as 3 hr^{-1} , fit nicely into the region for which $|e_p| \leq 20\%$, as do the data of Fig. 5. Points for which $R_3 \ll R_2$, those for which the technique is least accurate, are unlikely to occur unless the deposition or attachment rates of ^{214}Pb were to be greatly different from those of ^{214}Bi , which is unlikely.

COMPARISON WITH OTHER METHODS

In the Kusnetz method (Ku56), one simply multiplies the total number of alpha counts obtained during a given period by a conversion factor, which depends on the sampling, delay, and counting intervals, to obtain the estimated PAEC. For a sufficiently long delay time, the distinct linear combinations of I_1 , I_2 , and I_3 that determine the count total, N , and the PAEC, W , come to resemble each other sufficiently that a numerical factor may be used to convert N to W with a procedural error that remains quite small whatever the prevailing equilibrium conditions. Rolle (Ro72) has shortened the delay time considerably and demonstrated that the method is satisfactory when actual concentrations are similar to those obtained from a model that may be applicable to conditions obtaining in mines. It is possible to compare the total alpha and spectroscopic methods using the approach of the previous section.

Let us write N in the following form (cf. (5) and (6)):

$$N = \eta f [a_{11} + a_{21} + a_{22}R_2 + a_{23}R_3] I_1 \quad (25)$$

Then, if we approximate W by

$$w = \gamma N / (\eta f) \quad (26)$$

where our conversion factor, γ , is related to that of Kusnetz, F , by the expression

$$\gamma = \frac{1}{T_s T_c F}, \quad (27)$$

we find that the analysis leads, for a given procedural error e_p , to the

familiar relationship

$$R_3 = m(e_p)R_2 + b(e_p) \quad (28)$$

where now

$$m(e_p) = \frac{c_2(1 + e_p) - \gamma a_{22}}{\gamma a_{23} - (1 + e_p) c_3} \quad (29)$$

and

$$b(e_p) = \frac{c_1(1 + e_p) - \gamma(a_{11} + a_{21})}{\gamma a_{23} - (1 + e_p) c_3} \quad (30)$$

Comparing these equations with (18) and (19), we find that, in contrast with the spectroscopic method, the total alpha estimation does not permit the independent establishment of the slope and intercept of a line of given procedural error. In attempting to optimize the total alpha method, a choice of intercept near the origin, which avoids the placement of the line of zero error outside the usual limits of R_2 and R_3 , may lead to a slope which places the entire line outside the region of interest. For short measurement times, a compromise must be made, by trial-and-error, between the demands of slope and intercept.

If the measurement time is long, however, these limitations become insignificant. In a typical application of the Kusnetz method, we take $T_s = T_c = 10$ min and $T_d = 40$ min to obtain the results of Table 3, in which $m(0)$ and $b(0)$ are, respectively, the slope and intercept of the line of zero procedural error, and $m(\pm 1)$ and $b(\pm 1)$ define the boundaries of the 10% region. Given a satisfactory choice of the conversion factor, almost all of the usual equilibrium conditions lie within the

boundaries; Fig. 7 illustrates the situation when $\gamma = 1.60 \times 10^{-4}$ WL·l/min. The sensitivity of the results to the choice of conversion factor is apparent from the table; the analysis given here provides a basis for refinement of the technique.

When the total measurement time is relatively short, as in the Rolle method, the total alpha method requires a relatively long delay time between sampling and counting to avoid the overrepresentation of ^{218}Po in the estimation of the PAEC. The rapid spectroscopic method permits the reduction of the delay time to the minimum allowed by the instrument employed, with consequent sharp reduction in the statistical error and with relatively small sacrifice in procedural accuracy. We consider the total alpha method with timing intervals suggested by the literature (Ro72) and with conversion factors chosen to bring the line of 20% procedural error as closely as possible into coincidence with the line defined by $R_3 = R_2$. (This choice of conversion factor offers some improvement in procedural accuracy over those previously used.) The slopes and intercepts at $e_p = 0, \pm 20\%$ and the statistical error at $R_2 = 0.5$ and $R_3 = m(0) R_2 + b(0)$ are given in Table 4 for measurement times of 16, 23, and 31 min.

For comparison, similar information for the spectroscopic method is given in Table 5, where here $m(.2) = 1$ and $b(.2) = 0$ in all cases. Statistical errors of the two methods are effectively identical when the delay time is that of Rolle. The improvement in statistical accuracy of the spectroscopic method when the delay time is reduced is clear from the tables. At a measurement time of 23 min, the improvement is insignificant and is offset by the loss of procedural accuracy. At 16 min, however, a 50% improvement in statistical accuracy is obtained with a

loss of procedural accuracy which is, for most purposes, much less important. On the other hand, table 4 shows that reduction of the delay time of Rolle leads to the movement of the intercept of the line of 20% error away from the origin, reducing the area of the 20% region significantly. In contrast to the slope, the intercept is insensitive to changes in conversion factor, so that efforts to improve the accuracy of the Rolle method by adjustment of γ are ineffective. Because of the small effect on procedural accuracy of reducing the delay time, the measurement time used with the spectroscopic method is limited only by the sensitivity required; as indicated above, a measurement time of 11 min gives satisfactory results when the desired sensitivity is 0.005 WL, assuming a product of detector efficiency and flow rate of at least 1 l/min.

SUMMARY AND CONCLUSIONS

We have described a technique for the rapid estimation of the potential alpha energy concentration (PAEC) of radon decay products; the technique uses alpha spectroscopy to obtain two count totals after a single counting interval. The availability of the two totals makes possible an estimate of the PAEC that is superior to that of the single-count method in both precision and accuracy, particularly when the total measurement time is short.

One count total represents the decay of sampled ^{218}Po and the other that of ^{214}Po , which arises from sampled ^{218}Po , ^{214}Pb , and ^{214}Bi . From these, the PAEC can be estimated if a relationship can be assumed between the concentrations of the latter two products. Such an assumption is equivalent to the selection of a curve of zero procedural error

in the space of the daughter equilibrium ratios R_2 and R_3 . For values of R_2 and R_3 not lying on this curve, the estimation of the PAEC will result in a procedural error, e_p .

If a linear relationship between the two ratios is assumed, the estimate of the PAEC becomes a linear combination of the two count totals. We may select, based upon consideration of a simple model of radon decay product behavior, the slope and y-intercept of the line of any desired procedural error. Based on these choices, and on the choices of timing intervals, which depend principally on the reduction of the statistical error to an acceptably low level, we are able to select a pair of estimation coefficients.

Placement of the line of 20% procedural error on the line defined by $R_3 = R_2$ and the use of the rapid procedure with sampling and counting intervals of 5 min, separated by a decay interval of 1 min, lead to the following estimate of the PAEC:

$$w = (7.25 \times 10^{-5} N_1 + 1.93 \times 10^{-4} N_2) / (\eta f) \quad \text{WL}, \quad (31)$$

where η is the detector efficiency and f is the flow rate in 1/min. Under these circumstances, the line of zero procedural error is defined by

$$R_3 = 0.65 R_2 - 0.042. \quad (32)$$

The absolute procedural error is then less than 20% for most equilibrium ratios of interest; for any physical ratios we have $0.2 \geq e_p \geq -0.5$. At a product of detector efficiency and flow rate of 1 1/min, and concentrations of ^{218}Po , ^{214}Pb , and ^{214}Bi of 1.0, 0.4, and 0.3

pCi/l, respectively (0.004 WL), the statistical error is less than 20%.

The procedural and random errors resulting from this estimate, as well as those resulting from other choices of counting intervals and parameters, can be compared with estimates obtained from a single count registering the decays of both ^{218}Po and ^{214}Po . For total measurement times less than 15 min, the spectroscopic technique is clearly superior in the size of the area of the R_2 - R_3 plane for which the estimate may be expected to result in a procedural error below a fixed limit, or in the smallness of the statistical error, or in both of these. The minimum measurement time for which precise results may be obtained is smaller for the spectroscopic technique, principally because it allows the decay time to be reduced to the minimum allowed by the instruments. The spectroscopic technique may be recommended, therefore, whenever both short measurement times and high sensitivity are needed.

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Table 1. Rapid spectroscopic estimation technique: parameters, slopes and intercepts of lines of constant procedural error, and statistical error for various timing intervals and maximum procedural errors. The statistical error is calculated for R_2 and R_3 on the line of zero procedural error with $R_2 = 0.5$.

T_s (min)	T_d (min)	T_c (min)	ϵ	A (10^{-5} WL·l/min)	B (10^{-4} WL·l/min)	$m(0)$	$b(0)$	$m(-\epsilon)$	$b(-\epsilon)$	$e_s \sqrt{I_{inf}}$ ($\sqrt{pCi/min}$)
5	1	5	0.05	6.35	1.69	0.89	-0.013	0.78	-0.026	0.18
			0.10	6.62	1.77	0.79	-0.024	0.62	-0.045	0.19
			0.15	6.94	1.85	0.71	-0.034	0.49	-0.060	0.19
			0.20	7.25	1.93	0.65	-0.042	0.39	-0.073	0.20
			0.25	7.52	2.01	0.59	-0.049	0.30	-0.083	0.21
7.5	1	7.5	0.05	4.10	0.76	0.87	-0.015	0.75	-0.029	0.13
			0.10	4.32	0.80	0.76	-0.028	0.57	-0.051	0.13
			0.15	4.50	0.84	0.67	-0.038	0.43	-0.068	0.13
			0.20	4.68	0.87	0.60	-0.047	0.31	-0.081	0.14
			0.25	4.91	0.91	0.53	-0.055	0.22	-0.091	0.15
10	1	10	0.05	3.24	0.44	0.85	0.018	0.72	-0.033	0.10
			0.10	3.38	0.46	0.73	-0.032	0.51	-0.057	0.10
			0.15	3.51	0.48	0.63	-0.044	0.36	-0.076	0.11
			0.20	3.69	0.50	0.55	-0.054	0.24	-0.090	0.11
			0.25	3.82	0.52	0.48	-0.062	0.14	-0.101	0.11

Note: All headings are defined in the text.

Table 2. Results of a test of the rapid spectroscopic technique: an experimental comparison of the rapid spectroscopic technique (yielding an estimated PAEC, w) and a complete daughter measurement (yielding equilibrium ratios, R_2 and R_3 , and the "actual" PAEC, W); e_p is the percentage difference between w and W .

R_2	R_3	W (WL)	w (WL)	e_p (%)
0.27	0.23	0.051	0.057	11.8
0.29	0.14	0.082	0.079	-3.7
0.70	0.45	0.151	0.157	4.0
0.68	0.58	0.161	0.174	8.1
0.73	0.61	0.178	0.199	11.8
0.76	0.60	0.167	0.183	9.6
0.75	0.57	0.156	0.165	5.8
0.72	0.59	0.146	0.160	9.6
0.69	0.62	0.132	0.149	12.8
0.70	0.61	0.125	0.141	12.8
0.59	0.57	0.095	0.113	18.9
0.49	0.41	0.079	0.088	11.3

Note: All headings are defined in the text.

Table 3. The Kusnetz method: Conversion factors and slopes and intercepts of the boundaries of the region of 10% procedural error; $T_a = T_c = 10$ min, $T_d = 40$ min.

y (10^{-4} WL·l/min)	F (WL·l·min)	m(.1)	b(.1)	m(0)	b(0)	m(-.1)	b(-.1)
1.55	143	2.12	-0.35	0.87	-0.33	0.28	-0.31
1.56	142	2.24	-0.36	0.92	-0.33	0.31	-0.31
1.57	141	2.36	-0.36	0.97	-0.33	0.34	-0.31
1.58	141	2.50	-0.36	1.03	-0.33	0.37	-0.32
1.59	140	2.63	-0.37	1.09	-0.33	0.41	-0.32
1.60	139	2.78	-0.37	1.15	-0.33	0.44	-0.32
1.61	138	2.93	-0.37	1.21	-0.33	0.48	-0.32
1.62	137	3.10	-0.37	1.28	-0.33	0.51	-0.32
1.63	136	3.28	-0.38	1.35	-0.34	0.55	-0.32
1.64	135	3.47	-0.38	1.42	-0.34	0.59	-0.32
1.65	135	3.67	-0.39	1.49	-0.34	0.63	-0.32

Note: All headings are defined in the text.

Table 4. The Rolfe method: Slopes and intercepts of the boundaries of the region of 20% procedural error for various sampling, delay, and counting times; statistical errors at $R_2 = 0.5$ on the line of zero procedural error.

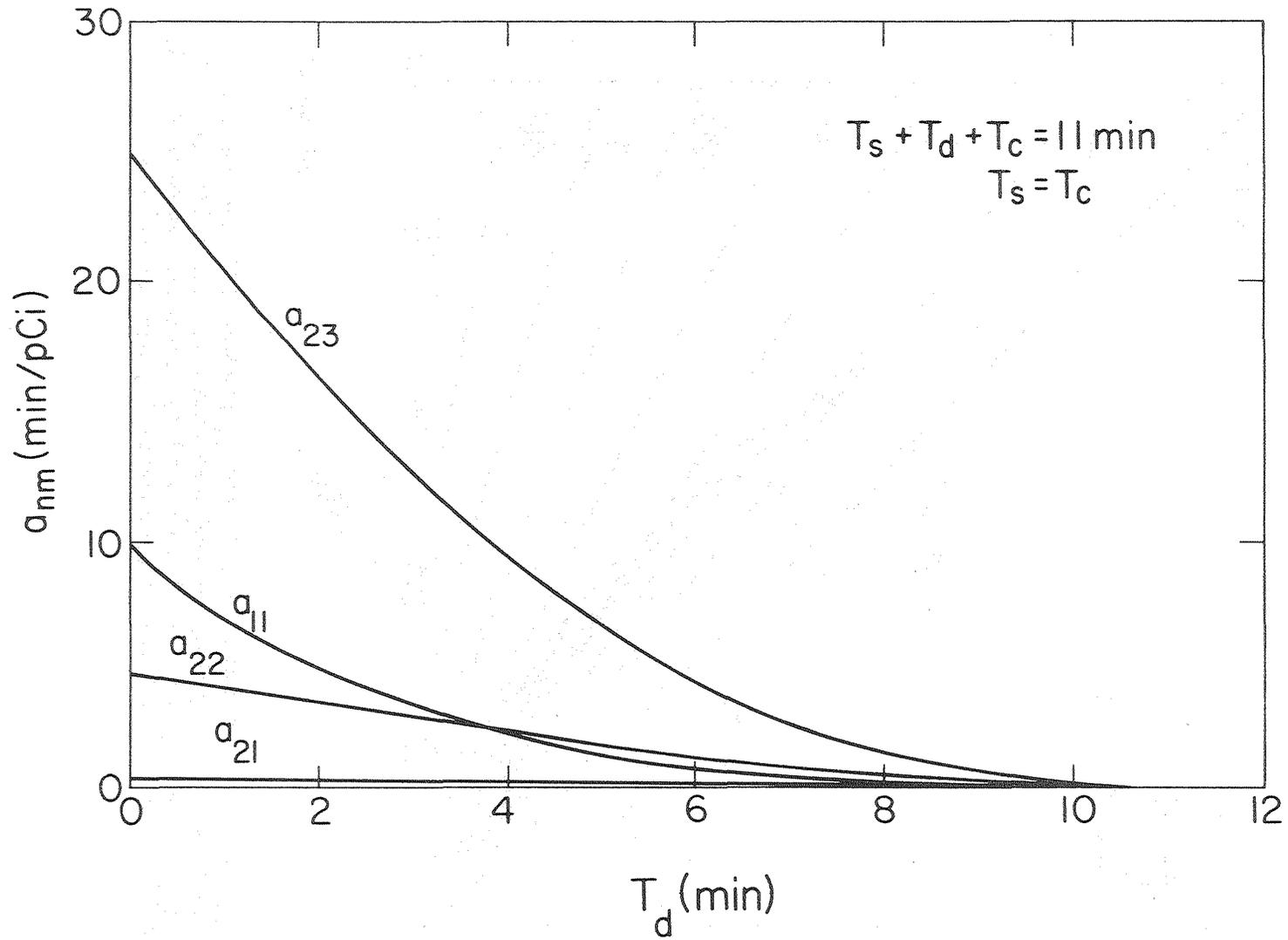
T_s (min)	T_d (min)	T_c (min)	y ($\times 10^{-4}$ WL·l/min)	$m(.2)$	$b(.2)$	$m(0)$	$b(0)$	$m(-.2)$	$b(-.2)$	$e_s \sqrt{I/nf}$ ($\sqrt{\mu\text{Ci/min}}$)
7.5	1	7.5	0.87	1.01	-0.251	0.60	-0.251	0.32	-0.260	0.12
7	2	7	1.00	1.01	-0.190	0.59	-0.206	0.30	-0.217	0.13
6.5	3	6.5	1.17	0.99	-0.135	0.57	-0.161	0.28	-0.179	0.15
6	4	6	1.37	1.01	-0.084	0.57	-0.121	0.27	-0.145	0.16
5.5	5	5.5	1.64	1.00	-0.040	0.55	-0.085	0.24	-0.117	0.18
5	6	5	1.99	1.21	0.026	0.65	-0.041	0.31	-0.084	0.20
11	1	11	4.16	1.00	-0.071	0.52	-0.113	0.21	-0.141	0.09
10	3	10	5.07	1.00	0.029	0.49	-0.037	0.17	-0.080	0.10
15	1	15	2.32	1.01	0.068	0.42	-0.016	0.07	-0.068	0.07

Note: All headings are defined in the text.

Table 5. The rapid spectroscopic technique: Slopes and intercepts of the boundaries of the region of 20% procedural error for various sampling, delay, and counting times; statistical errors at $R_2 = 0.5$ on the line of zero procedural error.

T_s (min)	T_d (min)	T_c (min)	$m(0)$	$b(0)$	$m(-.2)$	$b(-.2)$	$e_s \sqrt{I_1 n f}$ ($\sqrt{\text{pCi/min}}$)
7.5	1	7.5	0.60	-0.047	0.31	-0.081	0.13
5	6	5	0.54	-0.054	0.23	-0.092	0.20
11	1	11	0.52	-0.056	0.21	-0.093	0.09
10	3	10	0.49	-0.060	0.16	-0.099	0.10
15	1	15	0.42	-0.069	0.07	-0.110	0.07

Note: All headings are defined in the text.



XBL 811-73

Figure 1. Coefficients a_{nm} plotted against decay time T_d with fixed total measurement time of 11 min.

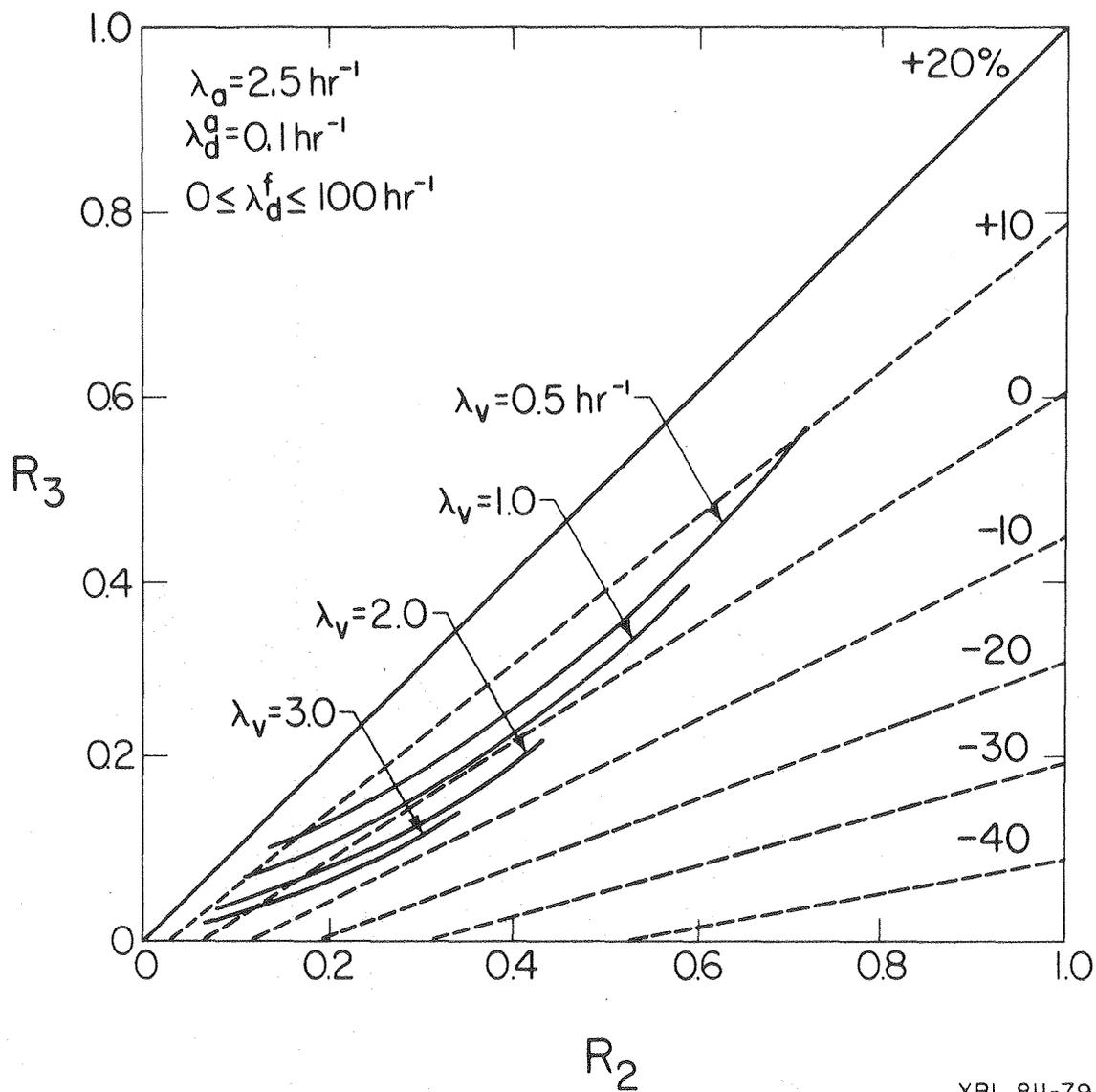


Figure 2. Equilibrium ratios R_2 and R_3 predicted by the radon decay product model with values of the parameters as indicated; lines of constant procedural error of the rapid spectroscopic technique, optimized for an 11 min total measurement time, are superimposed.

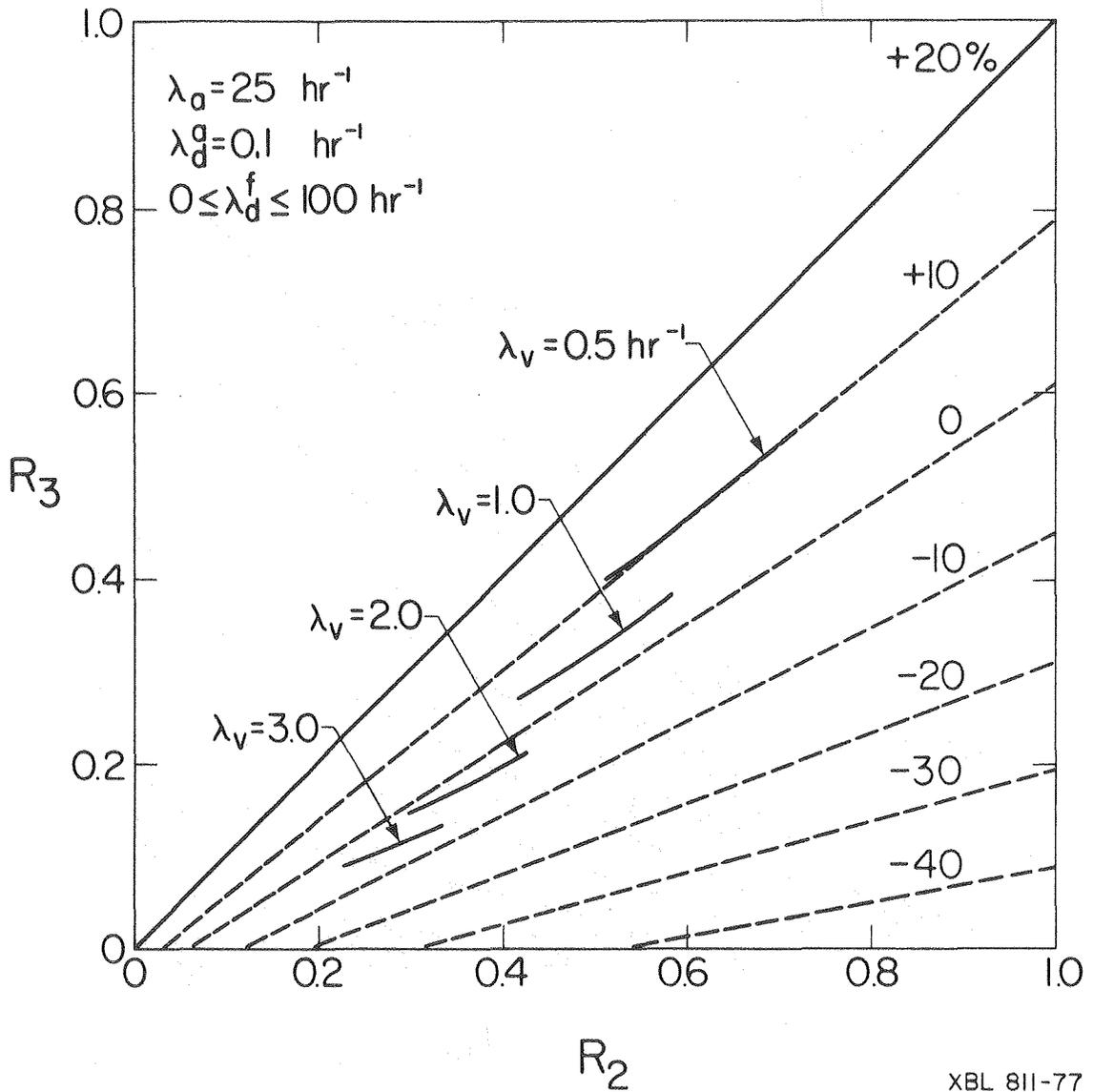


Figure 3. Equilibrium ratios R_2 and R_3 predicted by the radon decay product model with values of the parameters as indicated; lines of constant procedural error of the rapid spectroscopic technique, optimized for an 11 min total measurement time, are superimposed.

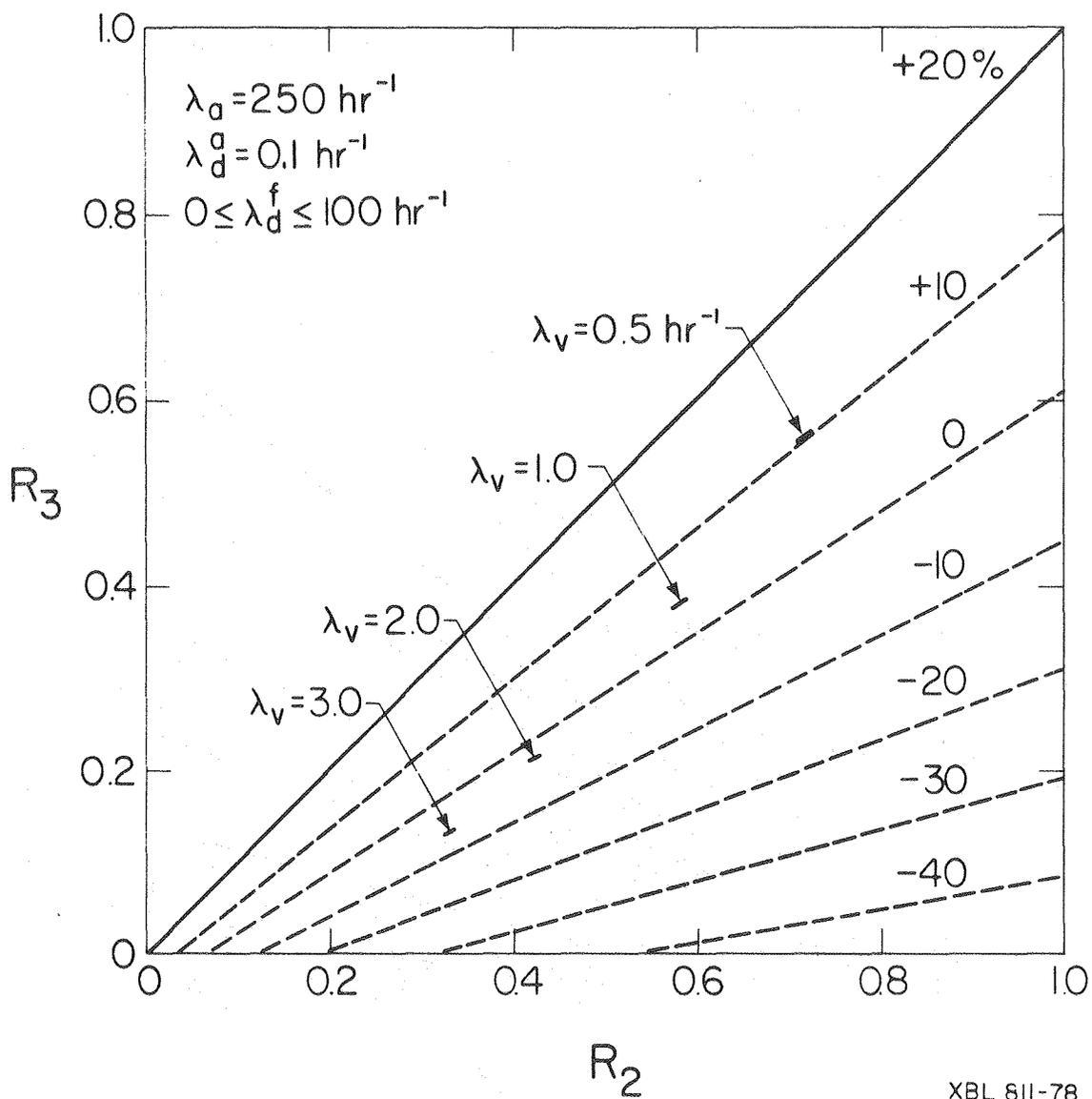
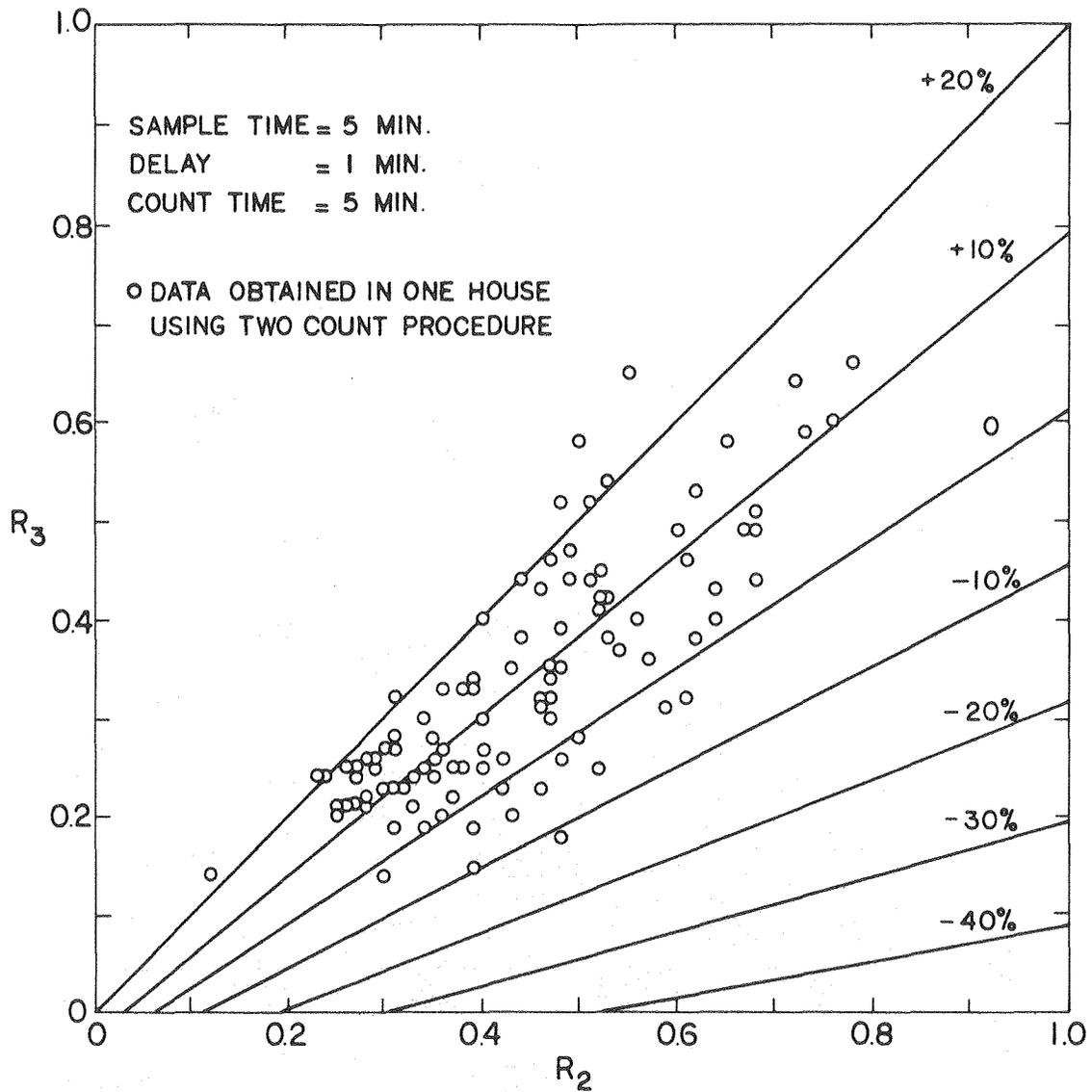


Figure 4. Equilibrium ratios R_2 and R_3 predicted by the radon decay product model with values of the parameters as indicated; lines of constant procedural error of the rapid spectroscopic technique, optimized for an 11 min total measurement time, are superimposed.



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Figure 5. Data from a tightly constructed home, with lines of constant procedural error as in 3, 4, and 5.

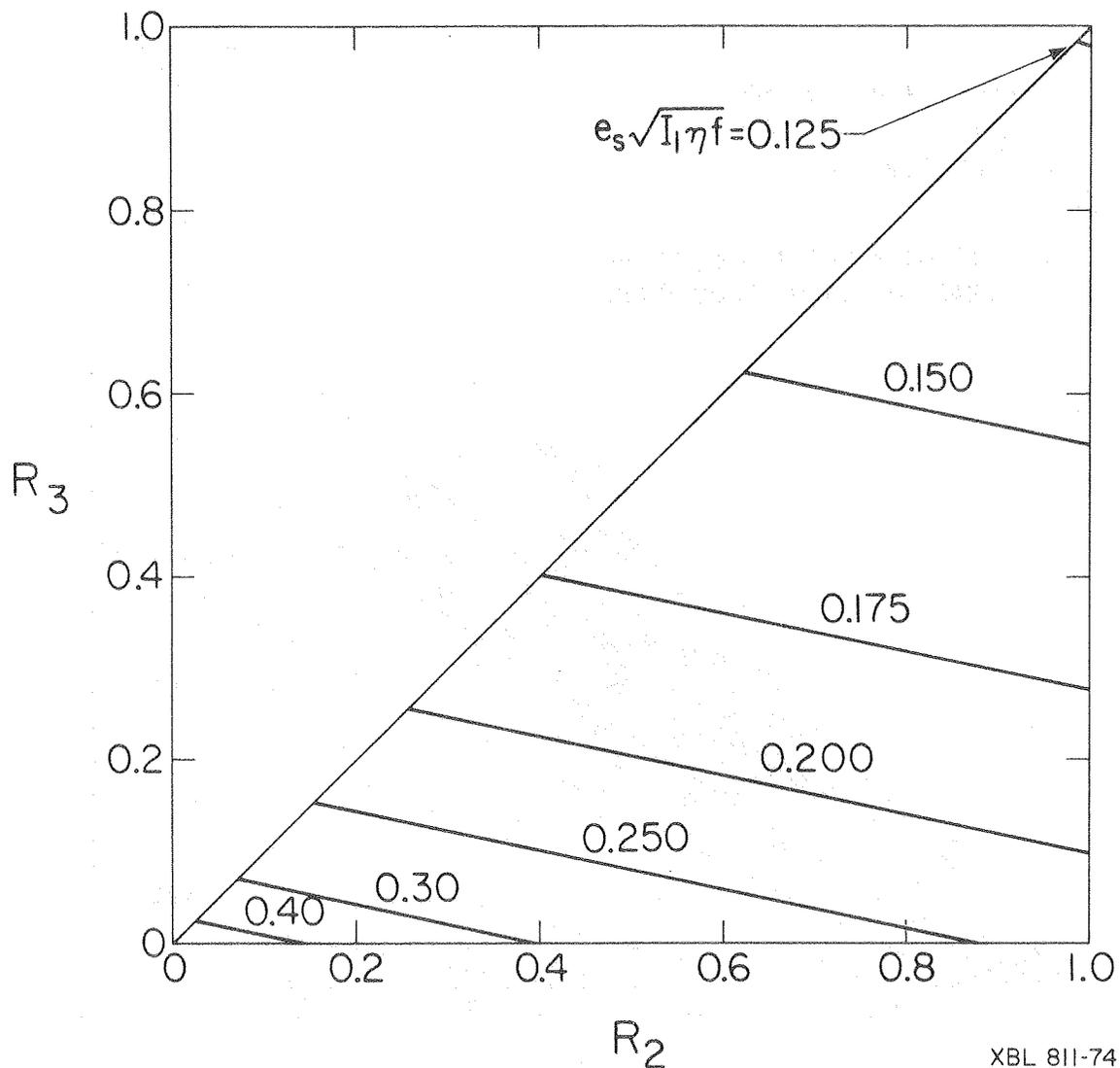
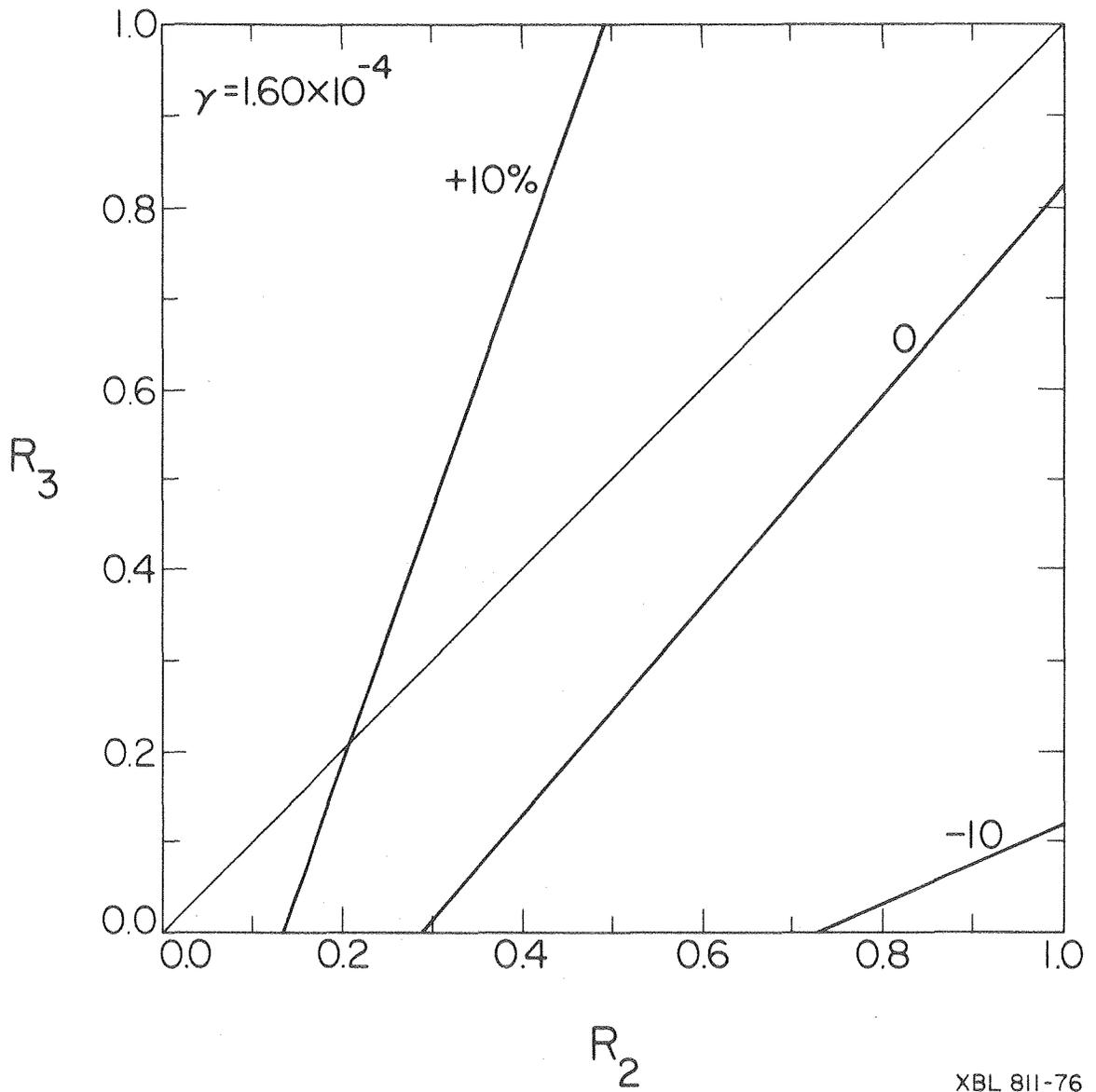


Figure 6. Lines of constant statistical error of the rapid spectroscopic technique, optimized for an 11 min total measurement time, plotted against the equilibrium ratios R_2 and R_3 . The error is normalized to a product of ^{218}Po concentration (I_1), detector efficiency (η), and flow rate (f) of 1 pCi/min. Under this condition the statistical error is always $\leq 20\%$ at a PAEC of .0052 WL.



XBL 811-76

Figure 7. The Kusnetz method: lines of constant procedural error for the conversion factor $\gamma = 1.60 \times 10^{-4}$ WL¹/min.

