LBL-10769
CONF-810153--1

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

ENERGY & ENVIRONMENT DIVISION

MASTER

Presented at the Second Special Symposium on Natural Radiation Environment, Bombay, India, January 19-23, 1981

ALPHA-SPECTROSCOPIC TECHNIQUES FOR FIELD MEASUREMENT OF RADON DAUGHTERS

W.W. Nazaroff, A.V. Nero, and K.L. Revzan

June 1981

Prepared for the U.S. Department of Energy under Contract W-7405-ENG-48

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Presented at the
Second Special Symposium on
Natural Radiation Environment
Bombay, India
January 19-23, 1981

LBL-10769
EEB-Vent 80-21

ALPHA SPECTROSCOPIC TECHNIQUES FOR FIELD MEASUREMENT OF RADON DAUGHTERS

W.W. Nazaroff, A.V. Nero, and K.L. Revzan

Building Ventilation and Indoor Air Quality Program
Energy and Environment Division
Lawrence Berkeley Laboratory
University of California
Berkeley, CA 94720

June 1981

DISCLAIMER

This work was supported by the Assistant Secretary for Conservation and Renewable Energy, Office of Buildings and Community Systems, Buildings Division, and by the Director, Office of Energy Research, Office of Health and Environmental Research, Human Health and Assessments Division and Pollutant Characterization and Safety Research Division of the U.S. Department of Energy under Contract No. W-7405-ENG-48.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ABSTRACT

Alpha spectroscopic techniques have not often been used to measure radon daughter concentrations in field studies because the equipment required is bulky and expensive. With advances in integrated circuit technology, less expensive, portable instruments have been developed that now make it possible to use these techniques to measure the low concentrations of radon daughters typically found indoors. One of two procedures may be used, two-count or single-count, depending on whether precision of measurement or rapidity of measurement is the more important objective. We have analyzed the effectiveness of both procedures. The results show that the two-count method permits individual radon daughter concentrations to be measured at the low level of one picocurie per liter with a relative standard deviation of about 20% within a total measurement time of 40 minutes (assuming the product of detector efficiency and sampling flow rate is one liter per minute.) With the single-count procedure, using the same assumption, potential alpha energy concentration can be estimated within a total measurement time of 11 minutes with less than 20% relative procedural error for common equilibrium conditions; the relative standard deviation is about 20% at 0.005 working level. These findings are compared with results obtained by other commonly used procedures.

Keywords: radon, daughter products, measuring methods, alpha spectroscopy

INTRODUCTION

Measuring low concentrations of radon daughters in buildings and in outdoor air requires techniques that are more sensitive than those generally applied in mines. At present, most field measurements of radon-daughter concentrations are made by measuring total alpha decays on a filter through which air has been drawn. Techniques based on alpha spectroscopy, which discriminate between the alpha particles from ^{218}Po and ^{214}Po, although more sensitive, have in most cases been restricted to laboratory studies because the bulky and expensive equipment they require made field applications impractical. Recent advances in integrated circuit technology, however, have led to the development of easily portable instruments for measuring radon daughters by alpha spectroscopy.[1,2] In order to realize the full potential of such instruments, one must carefully select the duration of and delay time between sampling and counting interval(s), and the data analysis procedure.

In measuring radon daughters one usually wants to achieve both measurement detail, often meaning high precision, and short measurement time -- objectives which, in practice, may conflict. Depending on experimental goals, however, one or the other objective may be emphasized. In ordinary field surveys, for example, detail can often be sacrificed in favor of shortening measurement time. On the other hand, when studying the behavior of radon daughters under various environmental conditions, precise measurement of individual daughters assumes priority. In this paper we examine two alpha spectroscopic procedures; one emphasizes the goal of high precision, and the other that of short measurement time.

PRECISE MEASUREMENT: TWO-COUNT INTERVAL-PROCEDURE

In the most general situation, the airborne activity concentrations of radon daughters are regarded as independent variables. To determine daughter concentrations without any prior assumptions about their relative activities, one must measure three independent parameters (i.e., decay counts) of the filter. In alpha spectroscopy, the counts from ^{218}Po and ^{214}Po can be independently registered; thus two counting intervals provide more than enough information for calculating the individual daughter concentrations. In the procedure described here, the

fourth count is used to compute the fraction, Θ, of the ^{214}Po counts that appear in the ^{218}Po channel because of energy lost as the alpha travels from the decay site to the detector.

The notation for the four count totals resulting from a measurement is: low-energy (^{218}Po) peak, first interval total = D_1, second interval total = D_4; high-energy (^{214}Po) peak, first interval total = D_2, second interval total = D_3. (All of these values are assumed to be corrected for background.)

The energy overlap factor, Θ, is then given by

$$\Theta = (D_4 - s\, D_1) \,/\, (D_3 - s\, D_2), \tag{1}$$

where

$$s = (e^{-\lambda_1 t_y} - e^{-\lambda_1 t_2}) \,/\, (e^{-\lambda_1 t_x} - e^{-\lambda_1 t_1}). \tag{2}$$

Here, λ_1 is the decay constant for ^{218}Po (0.227 min^{-1}), t_y and t_2 are the respective beginning and end times of the second counting interval, and t_x and t_1 are the respective beginning and end times of the first counting interval. (All times are in units of minutes and are measured from the beginning of sampling.)

From the definition of Θ and the count totals, D_1 through D_4, one can obtain the elements of the "corrected" count vector, $\underline{N}$, as

$$N_1 = D_1 - \Theta\, D_2, \quad N_2 = D_2\,(1 + \Theta), \quad \text{and} \quad N_3 = D_3\,(1 + \Theta). \tag{3}$$

It can be shown that for any given sampling and counting intervals, the elements of the vector $\underline{N}$ are linear combinations of the concentrations of the radon daughters in the sampled air. (See, for example, ref. 2.) For alpha spectroscopy, assuming equal detection efficiencies for the two alpha particles, this relation can be written in the form:

$$\underline{N} = (H \cdot \underline{I})\, \eta f, \tag{4}$$

where η is the detection efficiency, f is the sampling flow rate (1/m), and $\underline{I}$ is a 3-element vector (I_1,I_2, I_3) where the subscripts 1,2, and 3 refer to the first three radon daughters, ^{218}Po, ^{214}Pb, and ^{214}Bi, res-

pectively.* The 3 x 3 matrix, H, is obtained by solving the differential equations for radon daughter activity during and after sampling, and then integrating over the counting intervals used.[2,3] Equation (4) is inverted to obtain

$$\underline{I} = (K \cdot \underline{N}) / \eta f, \tag{5}$$

where $K = H^{-1}$. The values of s and of the matrix elements, k_{ij}, for three timing schemes are presented in Table 1. Once these values are obtained, the data for the two-count-interval alpha spectroscopic procedure can be analyzed in the field with a hand-held calculator.

To optimize the precision obtained with the two-count-interval technique, we define the minimum measurable concentration, MMC, for individual radon daughters; the MMC is the concentration in air of a radon daughter such that the standard deviation in the measurement, associated only with the random nature of radioactive decay, is 20% of the sampled concentration. The standard deviation is computed by standard propagation of error formulae[4] under the approximation that the number of counts observed, in either peak and for any time interval, is an independent, Gaussian-distributed variable. The MMC's are inversely proportional to the product of the detection efficiency and the sampling flow rate which, for this discussion, is assumed to be equal to one liter per minute. Finally, the closed-form solutions for the MMC's depend on the relative activities of the daughters, specified by two values, R_2 and R_3, which represent the ratio of activity concentrations of ^{214}Pb and ^{214}Bi to ^{218}Po, respectively.

We define a figure of merit for daughter measurement techniques as a linear combination of the MMC's, labeled MMCS:

$$MMCS = (1/3) \sum_{i=1}^{3} b_i \, MMC_i, \tag{6}$$

where

$$b_i = 100 \, (0.00103 + 0.00507 \, R_2 + 0.00373 \, R_3) / R_i, \tag{7}$$

---

*Because of its short half-life, the fourth daughter, ^{214}Po, is assumed to decay simultaneously with its predecessor.

and $R_1 = 1$. The product b_i MMC_i equals 100 W, where W is the potential alpha energy concentration (PAEC) resulting from daughter activities $I_1 = MMC_i/R_i$, $I_2 = I_1R_2$, and $I_3 = I_1R_3$. Thus each MMC has a "health effect" weighting factor so that a lower MMCS implies a more precise measurement of individual daughter concentrations at a given PAEC.

The MMCS was evaluated for fixed sampling and measurement times and over a range of equilibrium conditions.[2] In general, the best results are obtained by minimizing the delay between sampling and the first counting interval; for our manually-operated instrument, one minute was selected as a reasonable minimum. To find the minimum value of the MMCS, a computer program was written to search a two-dimensional time space that specifies when the first counting interval ends and when the second begins. Figure 1 shows a plot of the MMC's and the MMCS for optimized timing schemes as a function of total measurement time for the two-count-interval procedure. Although these results were obtained for a single sampling time (10 minutes) and for a single equilibrium condition (R_2:R_3 = 0.6;0.4), they are typical. For total measurement times below 35 minutes, the curves slope sharply downward, indicating that greater precision is associated with longer measurement time; above 35 minutes the gains are less pronounced. The figure also indicates that even with modest flow rates and detection efficiencies (again, the product is assumed to be one 1/m), a measurement time of 40 minutes allows the individual daughters to be measured at 1 pCi/l with a relative standard deviation of about 20%.

The MMCS can be evaluated for any completely determined measurement technique.[2] Table 2 compares the precision obtained from the two-count-interval alpha spectroscopic technique with that obtained from other techniques. As is evident, the procedure presented is more precise than other procedures commonly used.

RAPID ESTIMATION: SINGLE-COUNT-INTERVAL PROCEDURE

When short measurement time is the objective, as in field surveys, it is possible to use a single counting interval immediately after sampling. In this method, the ^{218}Po concentration is measured directly

from the total count in the low-energy peak. The PAEC is estimated from a linear combination of the counts in the two peaks. Thus, we obtain

$$I_1 = k_{11} N_1 / \eta f, \tag{8}$$

and

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

where w is the estimated PAEC, in units of working level, N_1 and N_2 are the counts due to ^{218}Po and ^{214}Po decays, respectively, and A and B are constants. The use of this technique for estimating PAEC introduces a "procedural" error resulting from incomplete knowledge of the equilibrium conditions. This error can be understood by realizing that there are many values of the PAEC which, depending on the equilibrium ratios, R_2 and R_3, will yield a given combination of counts, N_1 and N_2.

Figure 2 presents a map of the procedural error, for all equilibrium conditions, based on a sampling time of 5 minutes and a counting time of 5 minutes after a one-minute delay. The values of A and B and 7.25×10^{-5} and 1.93×10^{-4} WL·l/m, respectively, while k_{11} is equal to 0.0634 pCi/minute. Also plotted here are data obtained in one house where the two-count-interval procedure was used.[5] Out of approximately 100 data points, about 50% lie within the contours for 10% procedural error, and only the data points for which I_3 is greater than I_2 lie outside the 20% contours. The random errors encountered in estimating PAEC by this procedure can be similarly plotted, given the activity of one of the daughters.[6] For the data plotted in Figure 2, assuming ^{218}Po concentration of 1 pCi/l, and an ηf product of 1 l/m, the random error would, in all cases, be less than 20%.

To test this procedure, a series of samples taken in a room-sized test chamber were analyzed by both the single-count-interval and the two-count-interval procedures. The results of this test are presented in Table 3. In all cases, the disagreement is less than 20%.

The rapid spectroscopic procedure for estimating PAEC offers advantages over the single-count, total alpha methods commonly used. The Kusnetz method[7], while introducing a smaller procedural error, typically requires about 45 minutes for a single measurement. The Rolle method[8]

is not well-suited to measurement times below 16 minutes and, even if this total time is used, a 6-minute delay between 5-minute sample and counting periods is required for maximum precision. The long delay results in a greater random error than is obtained using the alpha spectroscopic procedure with a 16-minute measurement time. Finally, the rapid spectroscopic procedure allows us to obtain an indication of the radon daughter equilibrium by comparing the independently measured ^{218}Po concentration and the estimated PAEC.

CONCLUSIONS

The alpha spectroscopic procedures presented can be used effectively for measuring indoor concentrations of radon daughters in the field. The two-count-interval technique permits more precise measurement of individual radon daughter concentrations indoors than other procedures. For field surveys, where rapid PAEC determination is often the goal, the single-count-interval procedure presented here requires less measurement time than the total alpha procedures commonly used.

REFERENCES

1. Carson, D.W. (1979). Canalph-3: A portable three-channel alpha spectrometer for measuring the daughter products of radon and thoron, Report of Canada Centre for Mineral and Energy Technology.
2. Nazaroff, W.W. (1980). A residential radon daughter monitor based on alpha spectroscopy, M. Eng. thesis, University of California, Berkeley, CA (Lawrence Berkeley Laboratory Report LBL-10768).
3. Evans, R.D. (1969). Engineers' guide to elementary behavior of radon daughters, Health Phys., 17:229-252.
4. Young, H.D. (1962). Statistical treatment of experimental data, New York, McGraw-Hill.
5. Nazaroff, W.W., Boegel, M.L., Hollowell, C.D., and Roseme, G.D. (1981). The use of mechanical ventilation with heat recovery for controlling radon and radon-daughter concentrations in residences, Lawrence Berkeley Laboratory Report LBL-10222, to be published in Atmos. Env.
6. Revzan, K.L. and Nazaroff, W.W. (1981). A rapid spectroscopic technique for determining the potential alpha energy concentration of radon decay products, Lawrence Berkeley Laboratory Report LBL-11854, to be submitted to Health Phys.
7. Kusnetz, H.L. (1956). Radon daughters in mine atmospheres: A field method for determining concentrations, Ind. Hyg. Quart., 17:85-88.
8. Rolle, R. (1972). Rapid working level monitoring, Health Phys., 22:233-238.

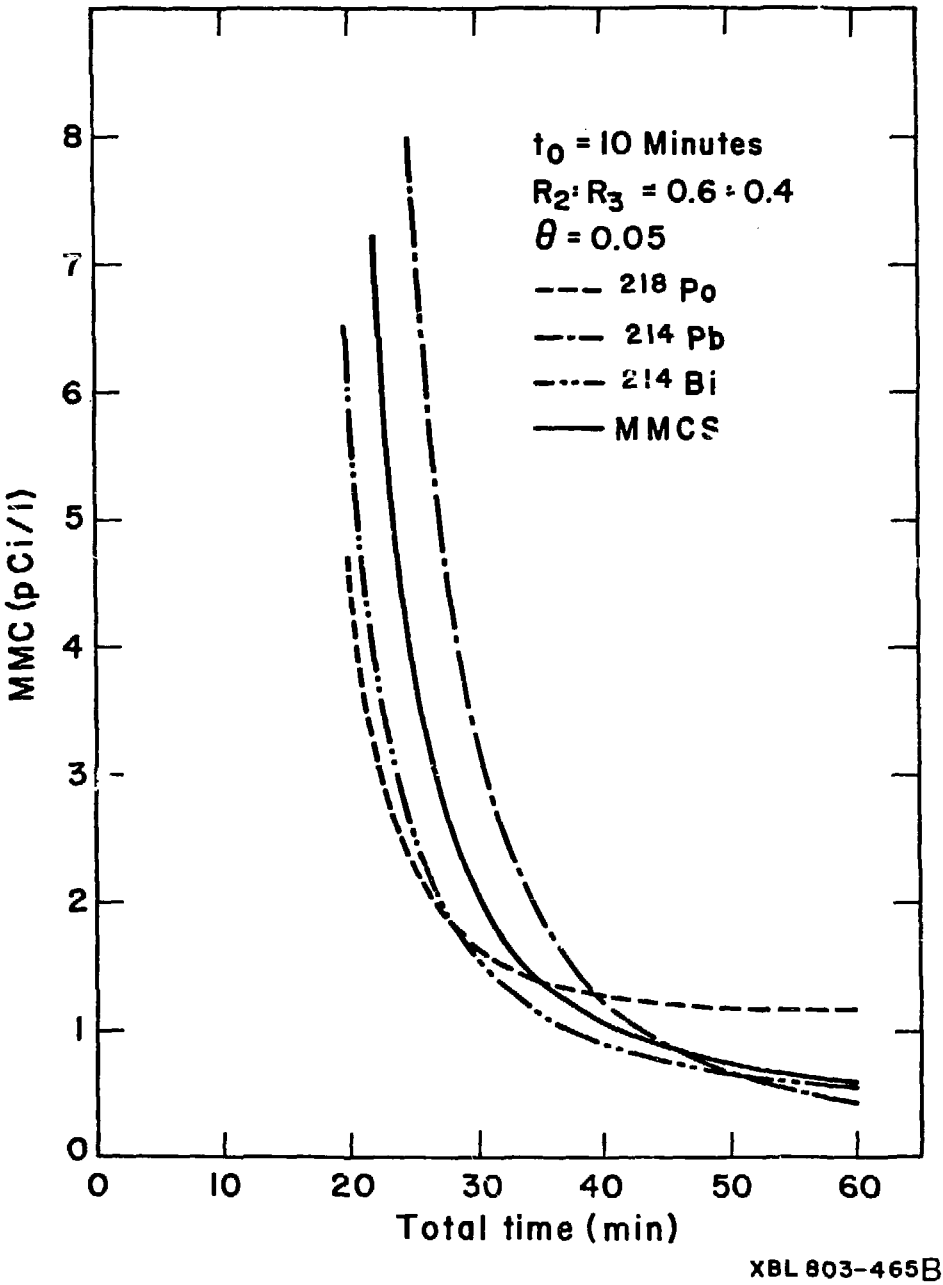

FIGURE 1. MINIMUM MEASURABLE CONCENTRATIONS OF RADON DAUGHTERS AS A FUNCTION OF TOTAL MEASUREMENT TIME FOR OPTIMIZED TWO-COUNT ALPHA-SPECTROSCOPIC TECHNIQUE.

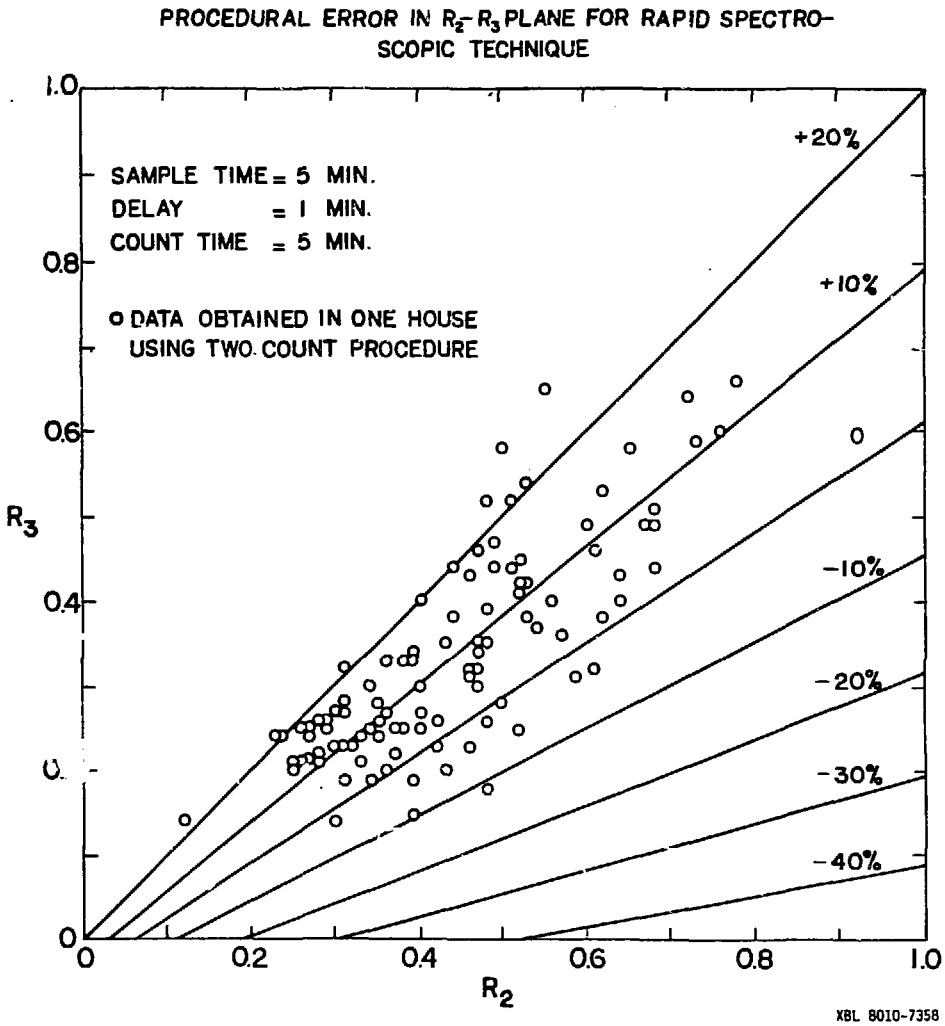

FIGURE 2. PROCEDURAL ERROR IN RAPID (SINGLE-COUNT) SPECTROSCOPIC TECHNIQUE. CONTOURS ARE LABELED WITH THE PERCENT OVERESTIMATE (+) OR UNDERESTIMATE (-) OF PAEC ASSOCIATED WITH THE UNCERTAIN EQUILIBRIUM CONDITIONS. ALSO PLOTTED ARE DATA FROM ONE RESEARCH HOUSE OBTAINED BY THE TWO-COUNT-INTERVAL PROCEDURE.

TABLE 1. CONSTANTS USED IN TWO-COUNT ALPHA-SPECTROSCOPIC PROCEDURE FOR DETERMINING RADON DAUGHTER CONCENTRATIONS

T_0	T_X-T_1 (MINUTES)	T_Y-T_2	K_{11}*	K_{21}	K_{22} (pCi/MIN)	K_{23}	K_{31}	K_{32}	K_{33}	S
5	6-15	24-40	0.0494	-0.00600	-0.0150	0.0178	0.00075	0.0174	-0.00502	0.0187
10	11-19	27-40	0.0389	-0.00476	-0.0104	0.0122	0.00064	0.0116	-0.00430	0.0300
10	11-21	31-50	0.0363	-0.00451	-0.00634	0.00779	0.00064	0.00902	-0.00302	0.0117

*$K_{12} = K_{13} = 0$

TABLE 2. COMPARISON OF TECHNIQUES FOR MEASURING RADON DAUGHTER CONCENTRATIONS

METHOD	T_0	T_X-T_1	T_Y-T_2	T_Z-T_3	MMC_1 [a]	MMC_2 [a]	MMC_3 [a]	MMCS [a]
	(MINUTES)				(pCi/L.)			
THOMAS-TSIVOGLOU [b]	5	7-10	11-25	26-35	30.9	6.5	9.9	11.3
CLIFF-JAMES-STRONG [c]	10	0-10	11-26	27-42	3.2	1.8	2.5	2.2
EWLM [d]	3	3.2-6.2			2.9	4.2	2.6	3.1
ALPHA SPECTROSCOPIC [e]	5	6-15	24-40		1.4	1.7	1.0	1.2
	10	11-19	27-40		1.2	1.3	0.9	1.0
	10	11-21	31-50		1.1	0.7	0.6	0.7

[a] VALUES COMPUTED FOR EQUILIBRIUM RATIOS R_2:R_3 = 0.6:0.4.

[b] THOMAS, J.W. (1972). HEALTH PHYS. 23:783-789.

[c] CLIFF, K.D. (1978). PHYS. MED. BIOL. 23:55-65.

[d] OVERLAP θ = 0.24, WITH RELATIVE SD OF 0.12, KEEFE, D.J., ET. AL. (1973) THE ENVIRONMENTAL WORKING LEVEL MONITOR. ARGONNE NAT. LAB. REPORT NO. P7628C.

[e] OVERLAP θ = 0.05.

TABLE 3. RESULTS OF A TEST OF THE RAPID SPECTROSCOPIC PROCEDURE

PAEC[a] (WL)	R_2[a]	R_3[a]	W[b] (WL)	$\frac{W-PAEC}{PAEC}$
0.051	0.27	0.23	0.057	+0.118
0.082	0.29	0.14	0.079	-0.037
0.152	0.70	0.45	0.157	+0.040
0.161	0.68	0.58	0.174	+0.081
0.178	0.73	0.61	0.199	+0.118
0.167	0.76	0.60	0.133	+0.096
0.156	0.75	0.57	0.165	+0.058
0.146	0.72	0.59	0.160	+0.096
0.132	0.69	0.62	0.149	+0.128
0.125	0.70	0.61	0.141	+0.128
0.095	0.59	0.57	0.113	+0.189
0.079	0.49	0.41	0.088	+0.113

[a] MEASURED BY TWO-COUNT-INTERVAL PROCEDURE.

[b] OBTAINED FROM 5-MIN. SAMPLE, 5-MIN COUNT WITH ONE-MIN. DELAY.

$A=7.25 \times 10^{-5}$, $B=1.93 \times 10^{-4}$