

VARIATION WITH EQUILIBRIUM CONDITION OF THE CALIBRATION FACTORS FOR CONTINUOUS RADON PROGENY MONITORS

Phillip H. Jenkins
Bowser-Morner, Inc.
Dayton, OH

ABSTRACT

The calibration factor for a continuous monitor that uses gross alpha-particle counting for measuring radon progeny concentration theoretically should vary as a function of the relative concentrations of the radon progeny; therefore, an inherent error is introduced by using a constant calibration factor, regardless of the equilibrium condition, as is the practice for most models of radon progeny monitor. An equation was derived describing the calibration factor as a function of the concentrations of the radon progeny and was used to calculate values of the calibration factor over the entire theoretical range of relative concentrations of radon progeny that comprise a constant collective radon progeny concentration. The results of these calculations showed that the calibration factor varies by approximately 10% over the entire theoretical range of relative concentrations of radon progeny. The range of relative concentrations that typically exist in indoor air is a small subset of the entire range that is theoretically possible; therefore, the calibration factor varies, in practice, due to this effect by less than 10% and the inherent error introduced by the detection and analysis method is not serious. Loss of radon progeny, particularly ^{218}Po , due to plateout on surfaces of the monitor before the progeny are collected on the filter has the effect of lowering the value of the calibration factor. This effect can be significant at conditions of low equilibrium when the fraction of radon progeny that are unattached to aerosols in the air is increased and plateout is more likely to occur. Two types of continuous radon progeny monitor, Alpha Nuclear, Model 100 CIRAS and Thomson & Nielsen, Model TN-WL-02, were exposed in Bowser-Morner's radon chamber under different conditions of radon progeny equilibrium. A calibration factor was derived for each monitor from the data for each exposure. The results indicated that the calibration factor for the Alpha Nuclear monitor varied from about 8 cph/mWL at low equilibrium conditions to about 16 cph/mWL at high equilibrium conditions, and that over the same range of equilibrium conditions the calibration factor for the Thomson & Nielsen monitor varied from about 3 to 9 cph/mWL. This variation in the calibration factor as a function of equilibrium condition is much more serious than the inherent error in the detection and analysis method. These results indicate that measurements of radon progeny concentration could be in error by as much as a factor of two or more if the equilibrium condition at the time of the measurement is significantly different from the equilibrium condition at which the monitor was calibrated.

INTRODUCTION

The basis for the detection and analysis for most models of continuous monitors used for the measurement of radon progeny concentration is gross alpha-particle counting of radon progeny collected on a filter. Typically, a gross-alpha count is made over a period of time, such as one hour, and a calibration factor is applied that converts the net count rate to the radon progeny concentration. With this method alone, it is not possible to determine the relative concentrations of the radon progeny, or the equilibrium condition. Even if one can assume that the efficiency for the collection of radon progeny is constant, the calibration factor for this type of monitor theoretically should vary as a function of the relative concentrations of the radon progeny; therefore, an inherent error is introduced by using a constant calibration factor regardless of the equilibrium condition.

Another factor that affects the value of the calibration factor is loss of radon progeny due to plateout on surfaces of the monitor before the progeny are collected on the filter. If the fraction of each of the radon progeny lost due to plateout were constant regardless of the equilibrium condition, then the value of the collection efficiency and the value of the counting efficiency could be combined into one constant term. However, plateout is more likely

to occur with radon progeny that are not attached to aerosols in the air, and the fraction of radon progeny, particularly ^{218}Po , that is unattached increases as the equilibrium ratio decreases. Therefore, there should be a greater loss of radon progeny, particularly of ^{218}Po , resulting in a lower calibration factor, at a low equilibrium condition than at a high equilibrium condition.

In order to gain an understanding of the magnitude of the variations in the calibration factor with the relative concentrations of radon progeny, two approaches were used; one based on theory and the other based on measurements made in a radon chamber. An equation was developed describing the calibration factor of a continuous radon progeny monitor, using gross-alpha counting, as a function of the individual concentrations of the radon progeny, and the variation in the calibration factor with different relative radon progeny concentrations was investigated. The effect on this theoretical variation of different amounts of loss of ^{218}Po due to plateout was also considered. The calibration factors for two models of continuous radon progeny monitor were measured by exposing the monitors in a radon chamber at different conditions of radon progeny equilibrium.

THEORETICAL EQUATION FOR THE CALIBRATION FACTOR

The first step in deriving an equation describing the calibration factor for a radon progeny monitor is to develop expressions describing the activities of the two alpha-emitting radon progeny, ^{218}Po and ^{214}Po , collected on the filter. It is assumed that the flow rate of the pump and the individual concentrations of the radon progeny are constant during the measurement, and that the efficiency for collection of the radon progeny from the air is 100%. A further assumption is that after ^{218}Po and ^{214}Pb are collected, their subsequent decay products are also collected on the filter. The activities of ^{218}Po and ^{214}Po on the filter are described by the following equations:

$$A_1 = F C_1 [1 - \exp(-\lambda_1 t)] / \lambda_1 \quad (1)$$

$$A_4 = F C_1 \lambda_2 \lambda_3 \left\{ [1 - \exp(-\lambda_1 t)] / [\lambda_1 (\lambda_2 - \lambda_1) (\lambda_3 - \lambda_1)] + [1 - \exp(-\lambda_2 t)] / [\lambda_2 (\lambda_1 - \lambda_2) (\lambda_3 - \lambda_2)] + [1 - \exp(-\lambda_3 t)] / [\lambda_3 (\lambda_1 - \lambda_3) (\lambda_2 - \lambda_3)] \right\} + F C_2 \lambda_3 \left\{ [1 - \exp(-\lambda_2 t)] / [\lambda_2 (\lambda_3 - \lambda_2)] + [1 - \exp(-\lambda_3 t)] / [\lambda_3 (\lambda_2 - \lambda_3)] \right\} + F C_3 [1 - \exp(-\lambda_3 t)] / \lambda_3 \quad (2)$$

where

- A_1 & A_4 = Activity of ^{218}Po and ^{214}Po , respectively, on the filter (pCi);
- F = Flow rate (L/min);
- $C_1, C_2, \text{ \& } C_3$ = Concentrations of ^{218}Po , ^{214}Pb & ^{214}Bi , respectively, in air (pCi/L);
- $\lambda_1, \lambda_2, \text{ \& } \lambda_3$ = Decay constants of ^{218}Po , ^{214}Pb & ^{214}Bi , respectively (min^{-1}); and
- t = Sampling time (min).

A further assumption inherent to eqns (1) and (2) is that, because of its extremely short half-life, ^{214}Po is always in secular equilibrium with ^{214}Bi , and that the alpha-particle decay of each ^{214}Po nucleus occurs simultaneously with the beta-particle decay of its parent ^{214}Bi nucleus. This assumption is frequently used to simplify the mathematics related to the decay and ingrowth of radon progeny, and its use introduces an insignificant amount of error.

The exponential terms in eqns (1) and (2) indicate the exponential growth of activity on the sample filter during the first few hours of sampling. These exponential terms become small when the sampling time exceeds four hours. Under the condition of steady state the exponential terms can be replaced with zeroes, and eqns (1) and (2) become:

$$A_1 = F C_1 / \lambda_1 \quad (3)$$

$$A_4 = F C_1 \lambda_2 \lambda_3 \left\{ 1 / [\lambda_1 (\lambda_2 - \lambda_1) (\lambda_3 - \lambda_1)] + 1 / [\lambda_2 (\lambda_1 - \lambda_2) (\lambda_3 - \lambda_2)] + \right. \\ \left. 1 / [\lambda_3 (\lambda_1 - \lambda_3) (\lambda_2 - \lambda_3)] \right\} + \\ F C_2 \lambda_3 \left\{ 1 / [\lambda_2 (\lambda_3 - \lambda_2)] + 1 / [\lambda_3 (\lambda_2 - \lambda_3)] \right\} + F C_3 / \lambda_3 \quad (4)$$

These equations can be simplified by first defining terms consisting only of the decay constants and then substituting the constant terms into eqns (3) and (4).

$$K_1 = 1 / \lambda_1 \quad (5)$$

$$K_2 = \lambda_2 \lambda_3 \left\{ 1 / [\lambda_1 (\lambda_2 - \lambda_1) (\lambda_3 - \lambda_1)] + [1 / [\lambda_2 (\lambda_1 - \lambda_2) (\lambda_3 - \lambda_2)] + \right. \\ \left. [1 / [\lambda_3 (\lambda_1 - \lambda_3) (\lambda_2 - \lambda_3)]] \right\} \quad (6)$$

$$K_3 = \lambda_3 \left\{ [1 / [\lambda_2 (\lambda_3 - \lambda_2)]] + [1 / [\lambda_3 (\lambda_2 - \lambda_3)]] \right\} \quad (7)$$

$$K_4 = 1 / \lambda_3 \quad (8)$$

$$A_1 = F K_1 C_1 \quad (9)$$

$$A_4 = F (K_2 C_1 + K_3 C_2 + K_4 C_3) \quad (10)$$

Note that in eqn (10) the terms containing K_2 and K_3 describe the activity of ^{214}Bi on the filter due to the collection of ^{218}Po and ^{214}Pb , respectively, from the air. It can be shown algebraically that K_1 and K_2 are equal. This makes sense physically, because at steady state the portion of the activity of ^{214}Po on the filter due to the collection of ^{218}Po from the air is equal to the activity of ^{218}Po on the filter. The values of the half-lives ($t_{1/2}$) of ^{218}Po , ^{214}Pb , and ^{214}Bi are 3.05 min, 26.8 min, and 19.7 min, respectively. The value for each decay constant is calculated from the relationship that $\lambda = \ln(2)/t_{1/2}$, and the values of K_1 through K_4 are calculated to be 4.40 min., 4.40 min, 38.7 min, and 28.4 min, respectively.

Assuming that a single value can be used for the counting efficiency for the detection of alpha particles emitted by ^{218}Po and ^{214}Po , the observed count rate is described by the following equation:

$$R = 133.2 E (A_1 + A_4) \quad (11)$$

where

- R = Count rate (cph);
- 133.2 = Conversion factor, 2.22 dpm/pCi x 60 min/h (dph/pCi); and
- E = Counting efficiency (c/d).

E in eqn (11) may also contain a constant factor for the efficiency of collection of radon progeny from the air. Substituting the values of A_1 and A_4 from eqns (9) and (10),

$$R = 133.2 E F (K_1 C_1 + K_2 C_1 + K_3 C_2 + K_4 C_3) \quad (12)$$

or

$$R = 133.2 E F (8.8 C_1 + 38.7 C_2 + 28.4 C_3) \quad (13)$$

The collective radon progeny concentration is expressed as a function of the individual concentrations of the radon progeny in the following equation:

$$RPC = 1.05 C_1 + 5.08 C_2 + 3.73 C_3 \quad (14)$$

where

RPC = Radon progeny concentration (mWL)

The calibration factor is the ratio of the net count rate, R, to the collective radon progeny concentration, RPC, as expressed in the following equation:

$$CF = 133.2 E F (8.8 C_1 + 38.7 C_2 + 28.4 C_3) / (1.05 C_1 + 5.08 C_2 + 3.73 C_3) \quad (15)$$

THEORETICAL VARIATION IN CALIBRATION FACTOR WITH NO PLATEOUT LOSSES

The exact magnitude of the calibration factor is not important here, only the manner in which it varies as a function of the relative concentrations of the radon progeny. In order to calculate values of the calibration factor, a value of 0.125 L/min was used for the flow rate, F, and a value of 0.143 c/d was used for the counting efficiency. A set of values for the concentrations of the radon progeny were chosen to span the entire range that is theoretically possible. A value of 1 WL for the collective radon progeny concentration was used in the calculations. The exact value of the collective radon progeny concentration is unimportant, as long as it is constant.

The concentration of each of the radon progeny, ^{218}Po , ^{214}Pb , and ^{214}Bi , in pCi/L that alone is equivalent to 1 WL was determined from eqn (14) by setting RPC to 1000 mWL and solving for C_1 , C_2 , and C_3 , assuming in each case that the concentrations of the other two decay products is zero. These concentrations of ^{218}Po , ^{214}Pb , and ^{214}Bi were determined to be 952 pCi/L, 197 pCi/L, and 268 pCi/L, respectively. Values of the calibration factor were calculated for every combination of C_1 , C_2 , C_3 , in increments of 0.1 WL, that sum to 1 WL. The values of C_1 , C_2 , C_3 were converted to the unit of pCi/L before being used in eqn (15). The calculated values of the calibration factor are listed in Table 1 and shown graphically in Fig. 1. Note that for constant values of the concentration of ^{218}Po , the calibration factor changes only slightly with changes in the relative concentrations of ^{214}Pb and ^{214}Bi .

The graph in Fig. 1 shows the values of the calibration factor as a triangular surface. The corner of the surface toward the upper left represents the condition where only ^{218}Po is present, the corner toward the lower right where only ^{214}Pb is present, and the corner toward the upper right where only ^{214}Bi is present. In the area of the surface away from the edges all three of the radon progeny contribute to the collective concentration. From the values of CF in Table 1, and from the surface in Fig. 1, it is seen that the calibration factor varies slightly with changes in the relative concentrations of the radon progeny. The largest value occurs when only ^{218}Po is present, and the smallest value occurs when only ^{214}Pb is present. The largest value is 10.1% greater than the smallest value.

The surface shown in Fig. 1 represents all possible combinations of the three radon progeny that contribute to a constant collective radon progeny concentration. Combinations of the relative concentrations represented by portions of the surface near the edges of the triangle are typically not found in an indoor environment. Therefore, the variation in the calibration factor in practical situations is likely to be considerably less than 10%. This indicates that the inherent error associated with using gross-alpha counting for the continuous measurement of radon progeny concentration is not seriously large.

Table 1. Values of Calibration Factor, CF, for Various Relative Values of Radon Progeny Concentrations

<u>Radon Progeny Conc. (WL)</u>			<u>CF (cph/mWL)</u>	<u>Radon Progeny Conc. (WL)</u>			<u>CF (cph/mWL)</u>
<u>^{218}Po</u>	<u>^{214}Pb</u>	<u>^{214}Bi</u>		<u>^{218}Po</u>	<u>^{214}Pb</u>	<u>^{214}Bi</u>	
1.0	0.0	0.0	19.96	0.3	0.2	0.5	18.68
0.9	0.1	0.0	19.77	0.3	0.1	0.6	18.68
0.9	0.0	0.1	19.77	0.3	0.0	0.7	18.69
0.8	0.2	0.0	19.59	0.2	0.8	0.0	18.49
0.8	0.1	0.1	19.59	0.2	0.7	0.1	18.49
0.8	0.0	0.2	19.59	0.2	0.6	0.2	18.49
0.7	0.3	0.0	19.41	0.2	0.5	0.3	18.49
0.7	0.2	0.1	19.41	0.2	0.4	0.4	18.50
0.7	0.1	0.2	19.41	0.2	0.3	0.5	18.50
0.7	0.0	0.3	19.41	0.2	0.2	0.6	18.50
0.6	0.4	0.0	19.22	0.2	0.1	0.7	18.50
0.6	0.3	0.1	19.22	0.2	0.0	0.8	18.50
0.6	0.2	0.2	19.23	0.1	0.9	0.0	18.30
0.6	0.1	0.3	19.23	0.1	0.8	0.1	18.31
0.6	0.0	0.4	19.23	0.1	0.7	0.2	18.31
0.5	0.5	0.0	19.04	0.1	0.6	0.3	18.31
0.5	0.4	0.1	19.04	0.1	0.5	0.4	18.31
0.5	0.3	0.2	19.04	0.1	0.4	0.5	18.32
0.5	0.2	0.3	19.04	0.1	0.3	0.6	18.32
0.5	0.1	0.4	19.05	0.1	0.2	0.7	18.32
0.5	0.0	0.5	19.05	0.1	0.1	0.8	18.32
0.4	0.6	0.0	18.86	0.1	0.0	0.9	18.32
0.4	0.5	0.1	18.86	0.0	1.0	0.0	18.12
0.4	0.4	0.2	18.86	0.0	0.9	0.1	18.12
0.4	0.3	0.3	18.86	0.0	0.8	0.2	18.13
0.4	0.2	0.4	18.86	0.0	0.7	0.3	18.13
0.4	0.1	0.5	18.87	0.0	0.6	0.4	18.13
0.4	0.0	0.6	18.87	0.0	0.5	0.5	18.13
0.3	0.7	0.0	18.67	0.0	0.4	0.6	18.13
0.3	0.6	0.1	18.67	0.0	0.3	0.7	18.14
0.3	0.5	0.2	18.68	0.0	0.2	0.8	18.14
0.3	0.4	0.3	18.68	0.0	0.1	0.9	18.14
0.3	0.3	0.4	18.68	0.0	0.0	1.0	18.14

EFFECT OF LOSS OF ^{218}Po DUE TO PLATEOUT

When the concentration of aerosols in the air is low, the percentage of radon progeny that are not attached to particles is increased. This is particularly true for ^{218}Po whose half-life is so short that it may decay before it contacts a particle on which to attach. In this condition, the radon progeny tend to migrate and attach to surfaces, such as walls, floors, ceilings, curtains, etc., thus depleting the collective radon progeny concentration in the air. The relative concentration of ^{218}Po in the air is increased; therefore, the region of the surface in Fig. 1 that is typical of this condition is nearer the corner in the upper left.

Continuous radon progeny monitors by necessity must pump air through some type of sampling head and through a filter which is in close proximity to a detector. Some monitors are designed so that the air must pass through a small opening in the case and into the interior of the monitor before reaching the filter. When unattached radon progeny enter the interior of the monitor, there is a high probability that they will attach to some surface of the monitor itself before they reach the filter where they should be collected and detected. This loss due to plateout is more likely to occur with ^{218}Po than with the other two radon progeny, simply because its unattached fraction is likely to be larger than those of the other two radon progeny.

The calculation of the set of calibration factors plotted in Fig. 1 was repeated with the assumption that 10% of the ^{218}Po in the air is lost to plateout. This set of calibration factors is shown graphically in Fig. 2. In this case, the 10.1% variation of the calibration factor present in Fig. 1 is almost entirely eliminated. This set of data indicates that if it were possible to design a monitor such that 10% of the ^{218}Po were consistently lost to plateout, the calibration factor would be nearly constant over the entire range of relative concentrations of the radon progeny.

The calculation of the calibration factors was repeated twice more, assuming that 20% and 30% of the ^{218}Po in the air is lost to plateout. These sets of calibration factors are shown graphically in Fig. 3 and Fig. 4. In progressing from Fig. 1 through Fig. 4, the effect on the surface representing the calibration factor values is that the upper left corner of the surface is lowered in increments of 10%. It is reasonable to expect that if the radon progeny are in a high equilibrium condition, represented by a point of the surface somewhere in the vicinity of the middle of the triangle, then there would be little loss of ^{218}Po due to plateout and the surface shown in Fig. 1 would be appropriate. If the aerosol concentration in the air is lowered, producing a lower equilibrium condition, represented by a location on the surface that is closer to the upper left corner, then the loss of ^{218}Po due to plateout is increased and a surface such as the ones shown in Fig. 2 through Fig. 4 would be appropriate depending on the magnitude of the equilibrium condition and the amount of loss due to plateout. In the case of 30% loss of ^{218}Po , the variation across the entire surface shown in Fig. 4 is still only about 20%.

MEASUREMENTS OF CALIBRATION FACTORS

Measurements of calibration factors were made under different conditions of radon progeny equilibrium using two types of continuous radon progeny monitors; Alpha Nuclear, Model 100 CIRAS and Thomson & Nielsen, Model TN-WL-02. The monitors were exposed in Bowser-Morner's radon chamber for a minimum of 24 hours after allowing four hours to achieve a steady-state condition. Both monitors were capable of printing or storing gross alpha-particle counting data on an hourly basis. The background count rate for each monitor was determined by exposing it to an atmosphere of nitrogen for a minimum of 24 hours, after allowing four hours to achieve a steady-state condition.

During each exposure, several grab sample measurements of the radon progeny concentration were made using the modified Tsivoglou method (Thomas 1972). Bowser-Morner's use of this method has been verified over a range of equilibrium conditions through intercomparisons exercises at the Department of Energy's Environmental Measurements Laboratory (EML). Each grab sample measurement was paired with a monitor's net count for the hour in which the grab sample was made. A least squares procedure was applied to the paired data to select the value of the calibration factor that produced the best fit of the monitor's hourly data to the grab sampling data. This value of the calibration factor was applied to all of the hourly measurements from the monitor to produce hourly average radon progeny concentrations. The hourly measurements of radon progeny concentration were averaged to produce the radon progeny concentration for the entire exposure period. Hourly radon measurements were made during the same time period using a continuous monitoring system consisting of three detectors. This system is calibrated by comparisons with grab scintillation cells, which in turn are verified through intercomparisons with EML and with the radon laboratories of the U. S. Environmental Protection Agency in Montgomery, Alabama and Las Vegas, Nevada. The average radon progeny concentration and the average radon concentration were used to determine the equilibrium ratio for the exposure period.

This procedure was used to determine the calibration factor for the two monitors at several conditions of radon progeny equilibrium. The equilibrium condition was held as constant as possible during each exposure, and was changed between exposures by varying the concentration of aerosols in the chamber air. The results of these measurements are presented in Table 2 and shown graphically in Fig. 5. The results indicated that the calibration factor for the Alpha Nuclear monitor varied from about 8 cph/mWL at an equilibrium of 14.5% to about 16 cph/mWL at an equilibrium condition of 45.0%. Over the same range of equilibrium conditions, the calibration factor for the Thomson & Nielsen monitor varied from about 3 cph/mWL to about 9 cph/mWL.

These results indicate a much greater variation in the calibration factor than predicted by the theoretical investigation discussed above. This implies that, at a very low equilibrium condition, the loss of ^{218}Po due to plateout is much greater than 30%. Perhaps also at a very low equilibrium condition there is loss of unattached ^{214}Pb and/or ^{214}Bi .

Table 2. Measured Values for the Calibration Factors of Two Types of Monitor

<u>Equilibrium (%)</u>	<u>Calibration Factor (cph/mWL)</u>	
	<u>Alpha Nuclear</u>	<u>Thomson & Nielsen</u>
14.5	7.86	3.17
18.0	12.05	5.11
35.0	15.84	8.89
43.0	16.41	8.61
45.0	16.55	--

CONCLUSIONS

The inherent error associated with using gross alpha-particle counting as the basis for a continuous radon progeny monitor was shown to be less than 10% and is not a serious concern. The variation of the calibration factor for a continuous radon progeny monitor was shown theoretically to vary by no more than 20% if the loss of ^{218}Po due to plateout is no more than 30%. Measurements of the calibration factors for two types of continuous radon progeny monitor, however, indicated that the calibration factor can vary by a factor or two or more depending on the equilibrium condition. More measurements should be made to verify these conclusions, since a small number of measurements of the calibration factor for each monitor was made. Also, this study should be expanded to include equilibrium conditions greater than 45%, additional models of continuous radon progeny monitor, and more than one instrument of each type. If the results of this study are typical, however, then a measurement using a continuous radon progeny monitor could be in error by as much as a factor of two or more if the equilibrium condition at the time of the measurement is significantly different from the equilibrium condition at which the monitor was calibrated.

REFERENCE

Thomas, J. W., "Measurement of Radon Daughters in Air," *Health Physics*; 23:783, 1972.

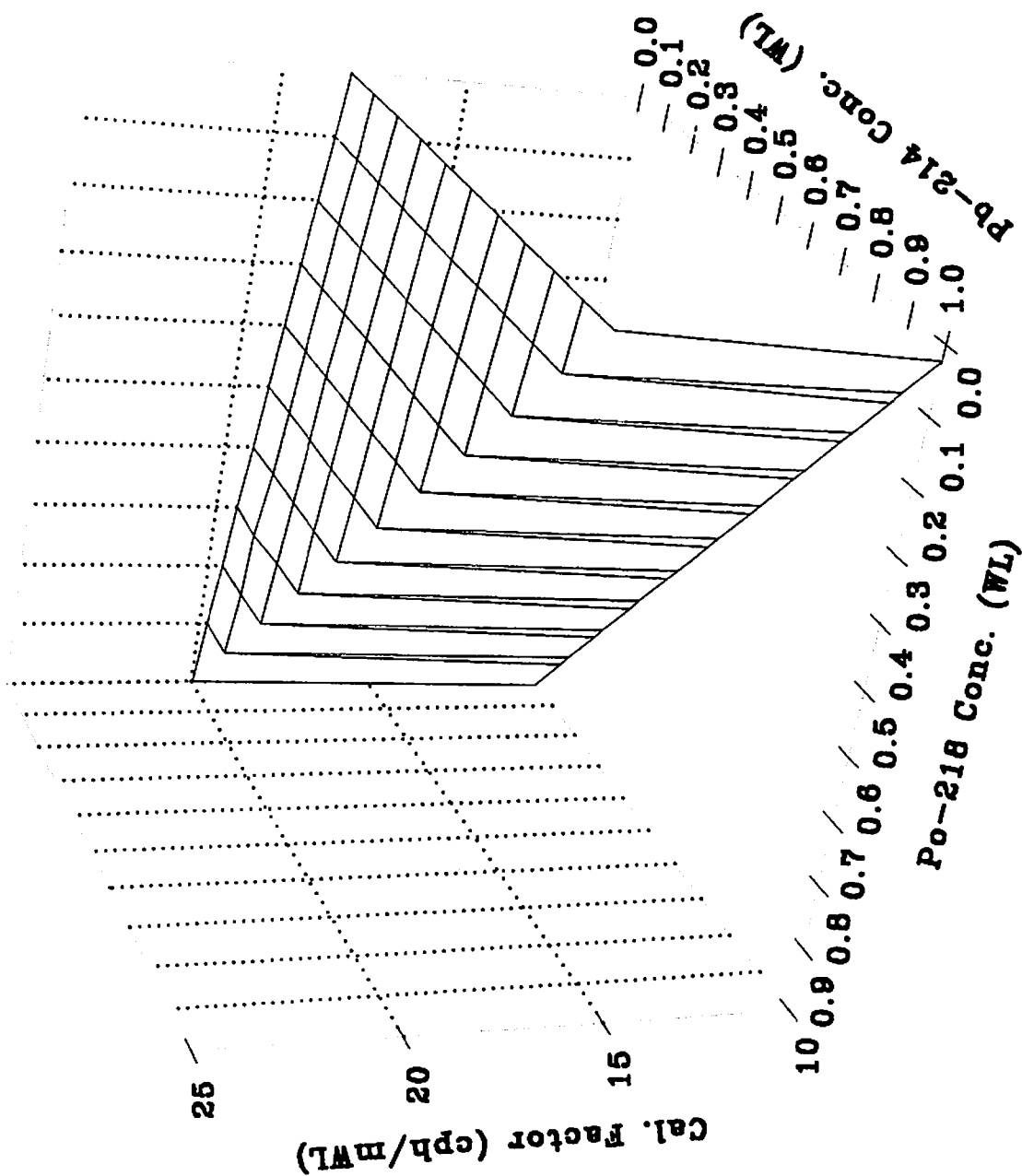


Fig. 1. Variation of CF with Relative Progeny Concentrations

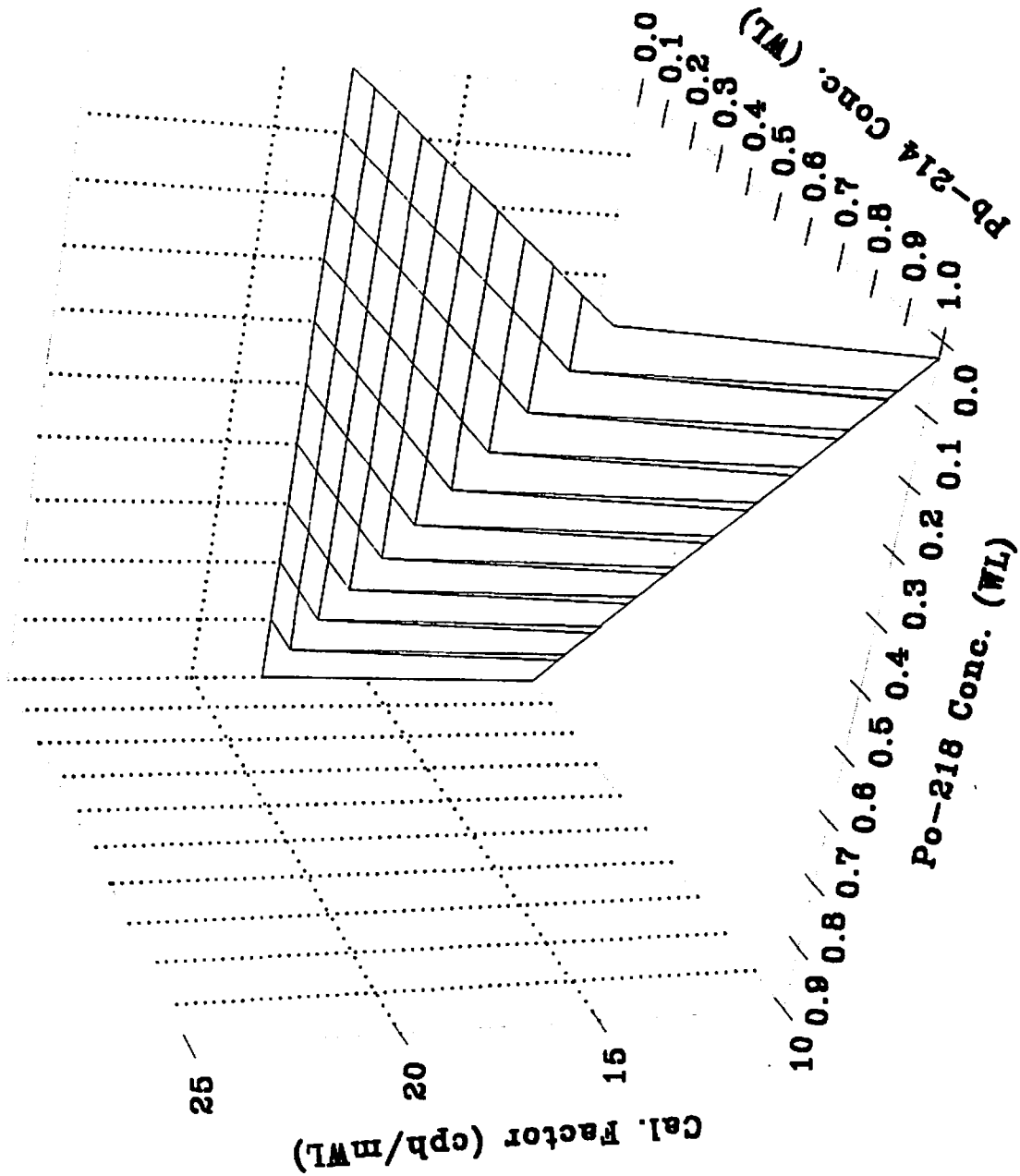


Fig. 2. Variation of CF - 10% Loss of Po-218 to Plateout

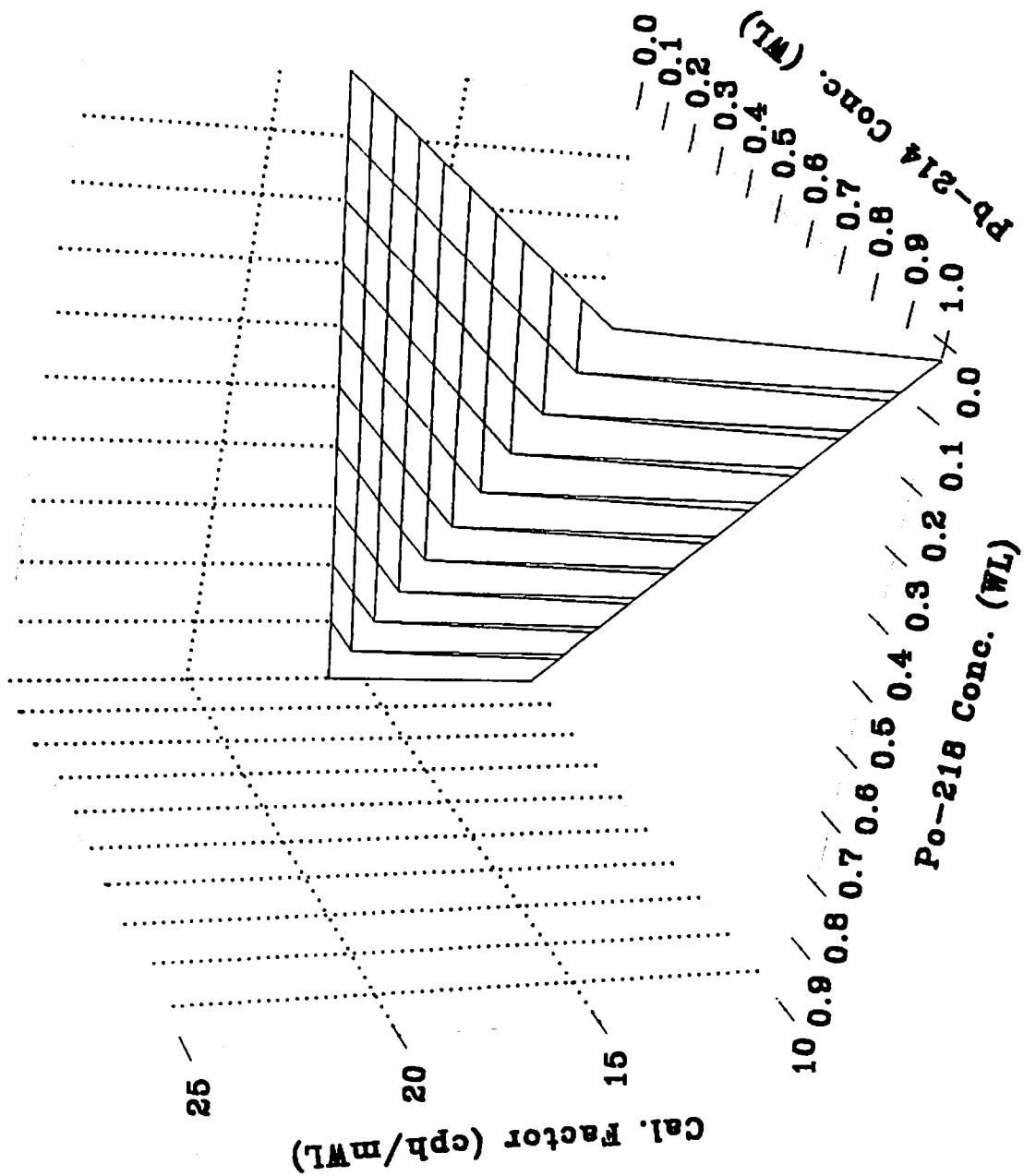


Fig. 3. Variation of CF - 20% Loss of Po-218 to Plateout

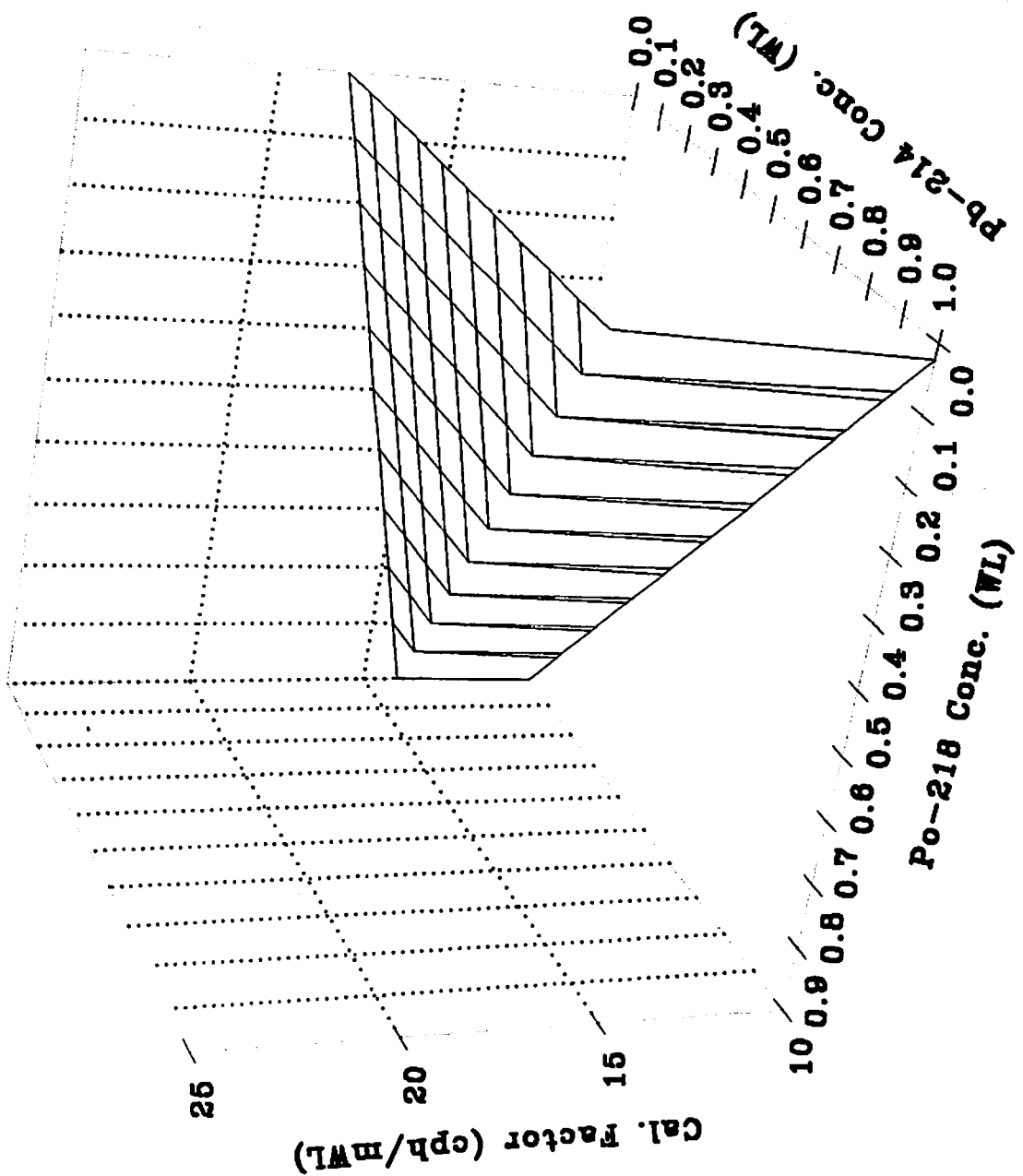


Fig. 4. Variation of CF - 30% Loss of Po-218 to Plateout

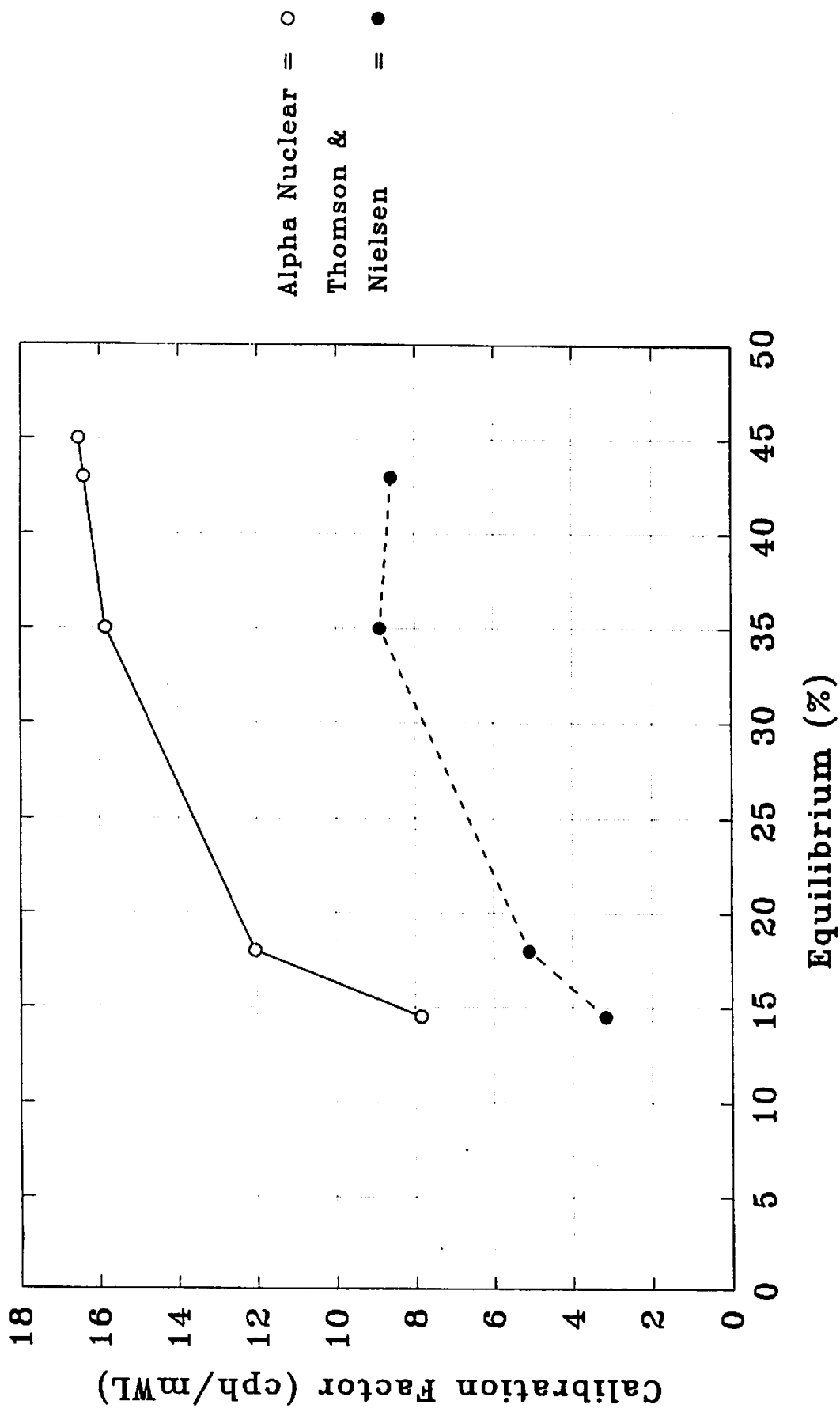


Fig. 5. Measured Values of Calibration Factors