

# Lawrence Berkeley National Laboratory

## Recent Work

### Title

Statistical Uncertainties Associated with Multiplexed Sampling with a Continuous Radon Monitor

### Permalink

<https://escholarship.org/uc/item/1pk652bq>

### Journal

Health physics, 64(3)

### Authors

Modera, M.P.  
Bonnefous, Y.

### Publication Date

1991-06-01



# Lawrence Berkeley Laboratory

UNIVERSITY OF CALIFORNIA

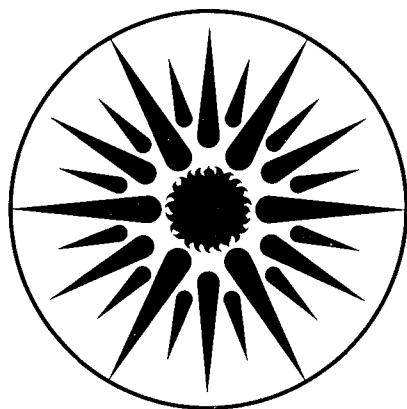
## APPLIED SCIENCE DIVISION

Submitted to Health Physics

### Radon Concentration Measurement of Multiplexed Samples with a Continuous Radon Monitor

Y. Bonnefous and M.P. Modera

June 1991



APPLIED SCIENCE  
DIVISION

1 LOAN COPY 1  
1 Circulates 1  
1 for 4 weeks 1 Bldg. 50 Library.  
Copy 2

LBL-30742

## **DISCLAIMER**

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

To be submitted to Health & Physics.

**Radon Concentration Measurement of Multiplexed  
Samples with a Continuous Radon Monitor**

Yves Bonnefous\* and Mark P. Modera  
Indoor Environment Program  
Lawrence Berkeley Laboratory  
Berkeley, California

\* Visiting Researcher from Ecole Nationale  
de Travaux Publics de l'Etat, Lyon,  
France.

---

This work was supported by the Director, Office of Energy Research, Office of Health and Environmental Research of the U.S. Department of Energy under Contract DE-AC03-76SF00098. Support for Yves Bonnefous's research participation was granted by ENTPE of Lyon, France.

**Radon Concentration Measurement of Multiplexed  
Samples with a Continuous Radon Monitor**

Yves Bonnefous<sup>\*</sup> and Mark P. Modera

Indoor Environment Program

Lawrence Berkeley Laboratory

Berkeley, California

June 1991

**ABSTRACT**

A Continuous Radon Monitor (CRM) is a flow-through device that counts alpha emissions from a gas sample as it passes through a scintillation cell. To improve its ability to track time-varying radon concentrations, a forward-marching data analysis procedure that takes into account the production, deposition and decay of radon daughters as a function of time had been previously developed by Busigin et. al. This paper examines some of the constraints associated with applying a CRM and the forward-marching analysis procedure to the problem of temporally varying radon concentrations. It utilizes Monte-Carlo simulations, together with limited experimental data, to perform: 1) an examination of the potential bias of the forward-marching analysis technique, 2) an examination of a simplified methodology for evaluating concentration-measurement uncertainty for known radon concentration histories, and 3) an analysis of an example application of the simplified uncertainty analysis to the multiplexing of soil-gas samples. The results of these analyses indicate: 1) that the forward-marching analysis technique is relatively unbiased, at least at concentrations above 4 pCi/l for 2-3 minute timesteps, and 2) that a simplified uncertainty analysis procedure based upon Poisson statistics and no error correlation between intervals provides generally acceptable estimates of measurement uncertainty.

---

<sup>\*</sup> Visiting Researcher from Ecole Nationale de Travaux Publics de l'Etat, Lyon, France.

## INTRODUCTION

Radon concentration monitoring in buildings is typically performed to check for elevated indoor radon concentrations, or to evaluate the effectiveness of radon mitigation work. Thus, most measurements are performed using relatively inexpensive integrating alpha-emission detectors. On the other hand, many research situations, in buildings and elsewhere, involve measurements of temporally-varying radon concentrations, for which more precise, and significantly more expensive, monitoring equipment is required. One apparatus that has been developed to meet these needs is a Continuous Radon Monitor (CRM), a flow-through device that counts alpha emissions from a sample as it passes through a scintillation cell (Thomas 1979). The major advantage of this device is its ability to track temporal variations in radon concentration. However, due to accumulation of radon daughters and their subsequent decays within the scintillation cell, the temporal resolution of a CRM is limited. To significantly reduce this limitation, a forward-marching data analysis procedure that takes into account the production, deposition and decay of radon daughters as a function of time was developed (Busigin 1979).

One current application of CRMs has been in experiments designed to monitor, understand, and model radon transport in and around buildings. As buildings have significant spatial as well as temporal variations in radon concentrations, detailed experiments require precise spatial and temporal resolution, pushing the limits either of existing research budgets or existing measurement equipment. Two examples of situations involving spatial variability in radon concentration are: 1) characterization of radon transport within a multizone building, and 2) characterization of soil-gas radon concentration fields. In both cases, the spatial variability could be accounted for either by increasing the number of CRMs, or by multiplexing the various monitoring points through the same CRM. The first of these options often proves to be prohibitively expensive, whereas the latter situation translates to a CRM time-resolution problem. It is the latter solution which forms the subject-matter of this paper.

This paper examines the constraints associated with applying a single CRM to the problem of monitoring spatially and temporally varying radon concentrations, including: 1) an examination of the stability and practicality of a previously developed temporal analysis technique (Busigin 1979), 2) an examination of a simplified methodology for evaluating concentration-measurement uncertainty for CRMs, and 3) an example application of the simplified uncertainty analysis to the multiplexing of soil-gas samples.

## UNCERTAINTY ISSUES

Radon concentration measurements, or any measurements that rely on the detection of radioactive decays to quantify the number of atoms present, are subject to the standard statistical uncertainties associated with any radioactive decay process. In general, the number of radioactive decays from a given number of radioactive atoms of a given specie (e.g., radon) in any time interval can be represented by a binomial distribution (Goldanskii 1962), whereas for a sufficiently large number of disintegrations within an interval the binomial distribution can be approximated by a Poisson distribution (Goldanskii 1962). Situations for which a Poisson distribution is valid have the distinct advantage that the uncertainty is simply equal to the square root of the mean number of disintegrations.

When using a continuous radon monitor to determine radon concentration, additional uncertainties and potential biases are introduced by the accumulation and disintegration of radon daughters ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ ,  $^{214}\text{Po}$ ) within the sample volume. As mentioned above, due to the long times intervals (2-3 hours) required to reach radioactive equilibrium between radon and its daughters, techniques that use forward-marching relationships to take into account the accumulation and decay of  $^{218}\text{Po}$  (half-life of 3.11 min),  $^{214}\text{Pb}$  (half-life of 26.8 min),  $^{214}\text{Bi}$  (half-life of 19.9 min), and  $^{214}\text{Po}$  (half-life of 164  $\mu\text{s}$ ) have been developed (Busigin 1979)\*. However, as a CRM does not utilize alpha spectroscopy, it is unable to isolate the contributions of individual species decays to the total number of decays detected in a given interval, and the forward-marching analysis algorithm therefore assumes that the partition is equal to the mean value (i.e.,

uses mean value relationships deterministically to determine the partition among the species). Moreover, the background disintegrations of deposited radon daughters and their associated statistical uncertainties reduces the signal-to-noise ratio, thereby increasing the uncertainty in the measured radon concentration<sup>\*\*</sup>. For these reasons, the uncertainties associated with applying the forward-marching analysis technique need to be quantified as a function of the variables characterizing a time-series of input radon concentrations. This paper focuses on the statistical uncertainties associated time-varying radon concentration estimates based upon measurements made with a CRM.

In addition to the statistical uncertainties discussed above, dynamic CRM measurements also have the potential for being biased by changes in sample radon concentration during a counting interval (Busigin 1979). This effect can be treated as an uncertainty whenever the radon concentration is varying in a random manner, however it could also show up as a systematic error, for example as a phase shift in periodic concentration variations. The magnitude of the potential bias or scatter due to time-varying radon concentrations can be reduced (or the frequency response of the measurement system increased) by reducing the analysis time, however such reductions in analysis time can create additional statistical uncertainties. A similar issue that must be considered with dynamic CRM measurements is the treatment of mixing time within the cell, an effect which can be seen as a special case of the time-varying concentration problem. Finally, the idea of multiplexing a single CRM has associated with it the additional issue of optimizing the uncertainties in a measurement series (or cycle). Based upon the problem of signal-to-noise ratios that vary depending upon the previous concentration history in the CRM, the choice of an appropriate sampling order, and the development of a full-cycle uncertainty analysis are required. The goal of this paper is to derive a workable procedure for estimating statistical uncertainties that could be incorporated into an experimental-design

\* There are additional daughters that result in an  $\alpha$  decay, however the first in this chain is  $^{210}\text{Pb}$ , which has a half-life of 22.3 years. Because of the long half-life by  $^{210}\text{Pb}$ , the  $\alpha$  decays which are eventually produced slowly increase with the use of the scintillation cell and are treated as a background count rate for the cell. The importance of the statistical uncertainty added due to these disintegrations depends on the total radon exposure of the cell, and the level of concentration being measured.

\*\* If temporal resolution on the order of ten minutes is adequate, a more recent CRM design could prove useful. This device monitors only the concentration of  $^{218}\text{Po}$  (which attains equilibrium in about 10 mins) via  $\alpha$  spectroscopy with a solid-state detector (Watnick 1986). Although it is not discussed in detail in this paper, this device could prove to be appropriate for multiplexed concentration measurements, the apparent trade-off being a somewhat lower collection efficiency, and therefore higher statistical uncertainty.



framework that takes into account the various sources of uncertainty and potential bias.

## STATISTICAL UNCERTAINTY FOR A CRM

As discussed above, the statistical uncertainty associated with a radon concentration measurement made with a CRM is a function of the concentration level, the duration of the counting interval, and the concentration history of that CRM. To include each of these effects, we will start with the equation derived by Busigin (Busigin et. al. 1979) for the average concentration for counting interval  $j$ :

$$C_{Rn,j} = \frac{Y_j}{0.037 V a \xi \tau} - h_1(1 - \frac{1}{a})C_{Rn,j-1} - \dots - h_i(1 - \frac{1}{a})C_{Rn,j-i} - \dots \quad (1)$$

where:

$C_{Rn,j}$  is the predicted average concentration in counting interval  $j$  [Bq/m<sup>3</sup>],

$Y_j$  is the number of alpha disintegrations in counting interval  $j$  [-],

$V$  is the volume of the scintillation cell [m<sup>3</sup>],

$a$  is a constant that depends on the length of the counting interval, the half-lives of <sup>222</sup>Rn, <sup>218</sup>Po, <sup>214</sup>Pb, and <sup>214</sup>Bi, and the scintillation-cell alpha-capture efficiencies for <sup>222</sup>Rn, <sup>218</sup>Po, and <sup>214</sup>Po [-],

$\xi$  is the scintillation-cell alpha-capture efficiency for <sup>222</sup>Rn [-],

$\tau$  is the length of the counting interval [s],

$h_i$  are constants that depend on the number of counting intervals back in time,  $i$ , the length of the counting interval, and the half-lives of <sup>222</sup>Rn, <sup>218</sup>Po, <sup>214</sup>Pb, and <sup>214</sup>Bi[-], and

$i$  is the index of the number of counting intervals back in time [-].

Given Equation 1, for the first measurement following a fresh-air flush period long enough to essentially eliminate previously-deposited radon-daughter decays, the variance

in the estimated radon concentration would be:

$$\sigma_{C_{m,1}}^2 = \frac{\sigma_{Y_1}^2}{(0.037 a V \xi \tau)^2} \quad (2)$$

However, the estimation of the variance even for this first period in a measurement series (Equation 2) is not as straightforward as it seems. The variance in the number of disintegrations,  $\sigma_{Y_1}^2$ , cannot be determined directly from a Poisson distribution, as the total number of disintegrations does not follow such a distribution due to the non-equilibrium nature of radon daughter generation and decays.

Using Equation 1 to estimate the uncertainty in the second point in a series of measurements results in:

$$\sigma_{C_{m,2}}^2 = \frac{\sigma_{Y_1}^2}{(0.037 a V \xi \tau)^2} + h_1^2 \left(1 - \frac{1}{a}\right)^2 \sigma_{C_{m,1}}^2 - \frac{2 h_1 \left(1 - \frac{1}{a}\right)}{(0.037 a V \xi \tau)} \sigma_{Y_2, C_{m,1}} \quad (3)$$

Compared to Equation 2, Equation 3 requires an additional piece of information, namely the correlation between the observed number of counts in period 2 and the estimated concentration in period 1. As one proceeds through the measurement series beyond period 2, this correlation issue is compounded.

## UNCERTAINTY ESTIMATION BY SIMULATION

To obtain a trustworthy analysis of the statistical uncertainty in any radon concentration measurement made with a CRM requires some means of dealing with: 1) the non-Poisson distribution of disintegrations under non-equilibrium conditions, and 2) the correlation between concentration estimates for different time periods. The means that was chosen to estimate of the significance of these two effects was a Monte-Carlo simulation of the entire decay-chain process within a flow-through scintillation cell. This was accomplished by simulating the random variation in  $^{222}\text{Rn}$  decays within the cell over very short time intervals (5 s) using a binomial distribution. The simulated number of decays is then assumed to be equal the number of atoms of the first radon daughter,  $^{218}\text{Po}$ , produced, whose decays are subsequently assumed to follow an independent binomial distribution with the appropriate half-life. It is thus assumed that all of the

$^{218}\text{Po}$  created is deposited within the scintillation cell, an assumption that can easily be modified, but which does not seem unreasonable based upon laboratory measurements of radon and radon-daughter counting efficiencies\*. Repeating the binomial-law decay/production process for all subsequent radon daughters,  $^{218}\text{Pb}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Bi}[-]$ , the simulation tracks all alpha decays from each species within the radon decay chain.\*\* The use of a 5-second time step assures that the decay of any particular species could be predicted by a single binomial distribution for each time step, and that the new species produced could be introduced at the beginning or end of each time interval with little or no effect on the simulation results. The latter issue was tested, the result being that the number of counts in any 3-min time interval was not biased by more than 0.5% when comparing beginning and end introduction of new species. The simulation results presented are based on assuming that the decays occur at the end of each interval. A schematic for this program is shown in Figure 1 (from Bonnefous 1989).

The simulation program developed could thus be used directly to estimate the uncertainty and bias in any radon concentration determined with Equation 1 for any given time history of radon concentrations passing through a scintillation cell. Figure 2 presents the average concentrations determined with Equation 1 using a three-minute counting interval for 1000 Monte Carlo simulations. Comparison of the average predictions with the input concentrations for the up-down staircase in Figure 2 suggests that Equation 1 is a relatively unbiased predictor of the radon concentration. The predictions in Figure 2 are biased at the 95% confidence level, however the magnitude of the bias is less than or equal to 1.6%↓. The concentration levels employed for this test are typical of those that might be found in soil gas (or in a house with unusually elevated concentrations). Figure 3 presents the coefficients of variation of the predicted concentrations in Figure 2, which appears to follow the expected trend of lowest uncertainty at the highest concentration, and highest uncertainty at the low concentration following a period of high concentration. The former would be expected simply upon the reduction in statistical uncertainty

\* CRMs were calibrated with a flow rate of 200 cc/min, which resulted in a radon counting efficiency of  $0.73 \pm 0.05$ , and an average radon daughter counting efficiency of  $0.82 \pm 0.03$ .

\*\* Due to its short half life (164  $\mu\text{s}$ ), the decay of  $^{214}\text{Po}$  was assumed to coincide with its production.

↓ Based on the size and the lack of correlation between the two biases, it can be shown that the observed bias in the analysis procedure (Equation 1) is not the result of assuming that the decays occur at the beginning of each 5 second interval.

associated with an increased number of decays, and the latter would be expected based upon the reduction in signal to noise due the radon daughters deposited in the cell during the high-concentration period. However, although the uncertainty estimates in Figure 3 appear to be adequate for most scientific error analyses, they suffer from the severe handicaps of being neither generalizable, nor sufficiently easy to generate (The computer time required to generate the data in Figures 2 and 3 is not justifiable under most circumstances).

As a first step in searching for a simplified, easy-to-compute estimate of the uncertainty in radon concentration estimates, the significance of the non-Poisson nature of non-equilibrium decays and the potential correlation between the errors in different counting intervals, were examined. To do so, estimates of standard deviations in predicted radon concentration were computed using an extension of Equation 3 based upon the assumption that there is no correlation between the number of counts in any given interval and the concentrations in the previous intervals:

$$\sigma_{C_{m,j}}^2 = \frac{\sigma_{Y_j}^2}{(0.037 a V \xi \tau)^2} + h_1^2 \left(1 - \frac{1}{a}\right)^2 \sigma_{C_{m,j-1}}^2 + \dots - h_i^2 \left(1 - \frac{1}{a}\right)^2 C_{Rn,j-i}^2 + \dots \quad (4)$$

By further assuming that the counts in any interval follow a Poisson distribution, the uncertainty in any concentration estimate could be determined with Equation 4 based simply on the number of observed counts in each interval. These uncertainty estimates are compared with the coefficients of variation obtained directly from the simulations in Figure 4. From a practical experimental-design point of view, the simplified uncertainty estimates in Figure 4 appear to be surprisingly good predictors of the actual (simulation-based) measurement uncertainties. The simplified uncertainty estimates are negatively biased on average, the size of the negative bias being largest at the beginning of the measurement series.

To understand the sources of the observed errors in the simplified estimates in Figure 4, the effect of non-Poisson counting statistics can be isolated from the effect of ignoring correlations between counting intervals. This is accomplished by using the observed variances of the counts in each interval rather than the variances predicted by a Poisson distribution in Equation 4. The observed variance in simulated counts, as well as the

variance predicted by Poisson statistics are quantified in Figure 5 for the concentration history shown in Figure 2. As expected, the deviation between the actual (simulated) variance and the Poisson-based variance tends to decrease over the time period after any change in concentration, and decreases quickly after radon injection is ceased, at which point the observed decays are associated with the decay series of  $^{214}\text{Po}$ , which is at this point close to equilibrium. Substituting the simulation-based count variances into Equation 4, the new estimates of the uncertainty in predicted radon concentrations are compared with the observed values and Poisson-based estimates in Figure 6.

The results in Figure 6 indicate that much of the difference between the original estimates and the observed uncertainties can be explained by the non-Poisson statistics of the counts in any interval, which results in turn from the non-equilibrium nature of the radon decay-chain disintegrations. On the other hand, the uncertainty estimates based on the observed number of counts seem to be consistently positively biased relative to the expected uncertainty, indicating the relative significance of correlations between measurements made in sequential intervals. The direction of that bias can be theoretically confirmed with a single-decay-product, two-timestep example (see Appendix).

Based upon the small magnitude of the differences between the observed and estimated uncertainties in Figures 4 and 6, we would like to be able to generalize the applicability of the simplified uncertainty estimates. To do so requires an examination of the impacts of: 1) changes in the concentration level, 2) the length of counting intervals, and 3) the shape of the imposed pattern of actual radon concentrations. Figures 7 and 8 contain comparisons (analogous to Figure 4) of estimated and observed uncertainties in radon concentrations determined with Equation 1 for radon concentration histories symmetric to that in Figure 2. Figures 7 and 8 differ from Figure 4 simply in that the concentration levels were respectively one and two orders of magnitude smaller than that used for Figure 2. Table 1 summarizes the results of the simulation-based analysis of the performance of the simplified uncertainty analysis estimations based upon Poisson statistics and no correlation, and includes no-correlation based estimates for comparison purposes.

**Table 1: Summary of Uncertainty Estimates for Radon-Concentration  
Estimates Based on Equation 1.**

<b>Max Conc. in Stair [pCi/l]</b>	<b>Average Coefficient of Variation [%]</b>	<b>Uncertainty Ratio ** (Mean) [-]</b>	<b>Uncertainty Ratio ** (Std. Dev.) [-]</b>	<b>Uncertainty Ratio ** (Std. Err.) [-]</b>	<b>Uncertainty Ratio↓ (Mean) [-]</b>
900	6.4	.95	.06	.02	1.05
90	21.	.94	.06	.01	1.09
9	65.	.96	.06	.02	1.06

\* Based on 1000 simulations.

\*\* Based on Poisson statistics and no correlation between intervals, compared to uncertainty obtained directly from 1000 simulation results.

↓ Based on actual statistics of counts and no correlation between intervals, compared to uncertainty obtained directly from 1000 simulation results.

As suggested by Figures 4, 7, and 8 as well as Table 1, the performance of the simplified uncertainty analysis is relatively unaffected by the radon concentration level. The most simplified uncertainty estimates (based on Poisson statistics and no correlation between successive counting intervals) are on average lower than that observed directly from the simulations, however this negative bias is consistent across two orders of magnitude in concentration, and is on the order of 5%.

An analysis of the bias of Equation 1 concentration estimates was also performed. Unlike the simplified uncertainty estimate analysis, it was found that as the magnitude of the input radon concentration was decreased, the magnitude of the bias between Equation 1 estimates and the true input concentrations seemed to increase. Figure 9 illustrates the tracking ability (i.e., apparent bias) of the forward-marching analysis procedure in Equation 1 for the different concentration levels. The average biases for the three-step

analyses were 1.01, 0.99 and 0.97 for 917, 90 and 9 pCi/l peak concentrations, respectively. A more in-depth analysis of these results showed that the increase in magnitude of the concentration-estimate bias stemmed from larger biases in the simulation-based distributions of observed decays. Both these effects were found to decrease as the number of simulations at lower concentrations was increased. Before applying the results of this paper to low-concentration (<10 pCi/l) time-series measurements, it would be prudent to perform additional simulations at low concentrations (however longer periods of constant concentration and longer analysis-time intervals would also be necessary to provide acceptable uncertainties at these concentrations).

## LABORATORY EXPERIMENTATION

The performance of the forward-marching analysis procedure (Equation 1) was also briefly examined in the laboratory. The experiment performed involved the use of a radon source, a fresh-air source, and mass flow controllers to create an input concentration profile equivalent to that shown in Figure 2. The details of the experimental procedure are included in Bonnefous 1989, however the key differences between the simulation profile and the experimentally created profile are 1) there is a finite mixing time ( $\approx 1$  min) associated with the cell volume during concentration changes, 2) the CRM used had experimentally-determined counting efficiencies of 0.73 ( $\pm 0.05$ ) for radon and 0.82 ( $\pm 0.03$ ) for radon daughters, compared to unity counting efficiencies in the simulations, and 3) there are additional uncertainties introduced by the instrumentation.

The results of the laboratory experiment are shown in Figure 10, which indicates good agreement between the input concentration and the estimates based on Equation 1. The coefficients of variation of the concentration estimates for the last four concentration estimates in each of the three steps are 6.5%, 5.9%, and 11.1%.<sup>\*</sup> These results are consistent with the simulation-based uncertainties presented in Figure 4, suggesting that the simulations can provide reasonable estimates of measurement uncertainty.

<sup>\*</sup> The first concentration estimate after a change in concentration was excluded to avoid mixing biases.

## PRELIMINARY FIELD APPLICATION

Both to test the performance of the simplified uncertainty estimation procedure for a different input radon concentration history (shorter steps and a longer test), as well as to utilize the procedure in its first practical application, the problem of analyzing multiplexed soil-gas samples from the soil surrounding a specially designed test structure was examined (Fisk et. al. 1989). This examination is based on field data from a thirteen-sample measurement series, during which each probe was sampled for 8 minutes. Based upon analysis with Equation 1 of the monitored history of counted decays, a concentration history was constructed (indicated by the solid line in Figure 11). This concentration history was then input to the simulation program (Figure 1), and run through a 500-simulation series. The simulation outputs were then analyzed on a two-minute time interval to check for any bias in Equation 1, and to evaluate the performance of Equation 4 (with and without Poisson statistics assumption). The mean predictions based on Equation 1 are presented in Figure 11, whereas the observed coefficients of variation, as well as the two estimates, are presented in Figure 12. Figure 11 suggests that there is little or no bias in the concentration estimates for this measurement series. Figure 12 is consistent with Figure 6, indicating that the Poisson-based no-correlation estimates generally underpredict the uncertainties, whereas the no-correlation estimates consistently overpredict the uncertainties. These results are summarized in Table 2, which is surprisingly consistent with Table 1, considering that the analysis time-step is one-third shorter, the time between concentration changes almost 50% shorter, the concentration level at least three times higher, and the concentration profile considerably different, compared to the three-step concentration histories used for Table 1. Both the Poisson-statistics/no-correlation underestimation of uncertainty, and the overprediction of no-correlation estimates are indistinguishable from the results in Table 1.



Table 2: Summary of Uncertainty Estimates for Radon-Concentration Estimates Based on Equation 1 for Soil-Gas Application.					
Input Conc.	Average Coefficient of Variation <sup>*</sup> [%]	Uncertainty Ratio <sup>**</sup> (Mean) [-]	Uncertainty Ratio <sup>**</sup> (Std. Dev.) [-]	Uncertainty Ratio <sup>**</sup> (Std. Err.) [-]	Uncertainty Ratio <sup>↓</sup> (Mean) [-]
13-Step Stair (Fig. 7)	5.9	.95	.04	.006	1.06
<p><sup>*</sup> Based on 500 simulations.</p> <p><sup>**</sup> Based on Poisson statistics and no correlation between intervals, compared to uncertainty obtained directly from 500 simulation results.</p> <p><sup>↓</sup> Based on actual statistics of counts and no correlation between intervals, compared to uncertainty obtained directly from 500 simulation results.</p>					

## DISCUSSION

The simulations performed in this study suggest that a simplified uncertainty determination procedure based upon Poisson statistics and no correlation between successive counting intervals can provide reasonably reliable estimates of the uncertainty associated with estimating the radon concentration history from the observed decay history with a previously derived forward-marching analysis procedure (Equation 1). However, as a closed-form mathematical expression for determining the limitations of the uncertainty estimation procedure was not derived, the evidence is principally empirical. As with any empirical relationship, it is critical to understand that relationship and to be able to define its limitations. The data presented in this paper, combined with some practical limitations and a general appreciation of the processes involved, can be used to help us

understand those limitations.

On a fundamental level, the performance of the simplified uncertainty estimates made with Equation 4 stems from two counteracting effects: 1) uncertainty overprediction due to ignoring error correlations between intervals, and 2) uncertainty underprediction due to assuming that the observed counts in each interval follow a Poisson distribution. Considering these two facts, one might like to understand how the significant independent variables likely to be associated with different applications can affect the two counteracting uncertainty biases. The first observation to be made relative to this point is that our simulations have spanned almost three orders of magnitude in concentration, and have included changes in analysis as well and concentration-variation intervals, yet they do not indicate any significant changes in the overall performance of the simplified estimates (see Tables 1 and 2). Moreover, the coefficient of variation of the estimated to simulated uncertainty ratio is on the order of 5%, suggesting that the performance of the uncertainty estimates did not change dramatically over the course of the experiments simulated, the implication being that experimental designs based upon the uncertainty estimate are not likely to be far from optimal.

To go beyond the integrated (overall) performance results for the simulated concentration histories, a rigorous analysis of the uncertainty estimation procedure would need to consider the separate effects of a number of experimental-design variables. Some of the variables to be included might be: 1) the magnitude of the concentrations being monitored, 2) the number of decays within an analysis time-interval (i.e., the product of concentration and deposited daughter level times time-interval length), 3) the degree of radon-daughter deposition within the cell, 4) the direction of the concentration variations between time intervals (i.e., the sign of the derivative of the concentrations being introduced into the scintillation cell), and 5) the time interval over which input concentrations are varied (i.e., the magnitude of the concentration derivative). These latter two variables essentially define the shape of the input radon concentration profile. Some limited examinations of these variables were conducted, the results of which are summarized below.

Concerning the first experimental-design variable, our simulations have shown that the impact of concentration level on the combined effect of Poisson-statistics and no-correlation assumptions is small (see Table 1). Separate analyses of the impact of concentration level on each assumption were also performed, and showed that neither was systematically impacted by concentration level.

To address the impact of the absolute number of decays in an interval (the second experimental-design variable), data from the simulations, which cover almost three orders of magnitude in decays per interval (4-2500), was used. A scatter plot, based on these simulations, of the no-correlation/true-uncertainty ratio versus counts per interval indicates no particular trend. Similarly, a scatter plot of the effect of using the Poisson assumption (obtained by subtracting the no-correlation estimates from the Poisson-statistics/no-correlation estimates) also doesn't show a trend with the number of counts per interval.

Relative to the degree of radon-daughter deposition within the scintillation cell (the third experimental-design variable), the impact of the no-correlation assumption is expected to be smallest when the cell is not charged with daughters. Scatter plots of all the data from the three-step simulations and the soil-gas simulations indicated the expected impact of radon-count ratio (the ratio of radon decays to total decays during a period) on the no-correlation assumption, namely that lower radon-count ratios are correlated with higher no-correlation overpredictions. These plots also showed a much less obvious impact of radon-count ratio on the Poisson-statistics assumption, in this case with the opposite sign. Based upon these results, it seems that the radon to total decay ratio could possibly be used to predict the performance of the uncertainty estimates over the course of an experiment, however a more rigorous analysis of this sort was deemed inappropriate at this point.

Concerning the impacts of the shape of the input concentration profile on the performance of the simplified uncertainty estimates (experimental-design variables 4 and 5), several trends could be observed in the simulation results, however none of these trends were deemed significant enough to merit in-depth examination. It should be noted however that only two different time periods between concentration changes and two

analysis-time intervals were simulated.

## CONCLUSIONS

Two main conclusions can be drawn from the research presented in this paper. The first conclusion is that a previously-derived forward-marching algorithm can provide relatively unbiased radon concentration estimates from the time history of observed decays in a flow-through scintillation-cell device. The second conclusion is that the simplified uncertainty analysis algorithm examined in this paper can provide accurate estimates of the uncertainty of radon concentrations determined with the forward-marching algorithm.

Based on Monte-Carlo simulation results for three-step radon concentration histories and a soil-gas sampling application, it seems that the forward-marching analysis algorithm can provide relatively unbiased concentration estimates (< 3.5%) over a wide range of radon concentrations (45-2800 pCi/l). An increased bias in concentration estimates (< 7%) was however observed at low concentrations (4.5 to 9 pCi/l). This increased bias could be explained somewhat by the observed bias in simulation samples at low concentrations (these biases decreased as the number of simulations was increased), however additional analyses would be needed to fully understand the performance of the algorithm for such low-concentration/short-timestep analyses (i.e., low-event timesteps). On the other hand, from the practical point of view, a somewhat larger bias at low concentrations is not especially disturbing, particularly in light of the fact that the uncertainty at those concentrations and timesteps is on the order of 40-80%, which would make such measurements almost universally unacceptable.

Concerning the second conclusion, the Monte-Carlo simulations were also used to verify the performance of a simplified algorithm for analyzing the uncertainty of radon concentrations obtained with the forward-marching analysis algorithm. For each of the four sets of simulations, the simplified uncertainty algorithm estimated the average uncertainty to within 6% of the true value. It was also found that the performance of the simplified algorithm did not change significantly over a wide range of radon

concentrations (4.5-2800 pCi/l). The success of this algorithm, which assumes a Poisson distribution for observed decays, and that the errors are uncorrelated between adjacent timesteps, seems to derive from the the fact that the effects of these two assumptions tend to cancel. The Poisson assumption was found empirically to consistently underpredict the actual variance in observed decays during a timestep, whereas the assumption of no correlation consistently overpredicted the variance in concentration estimates (an analytical justification of this result is derived in the appendix). As a result of these analyses, and the fact that the performance of the uncertainty estimation algorithm did not change dramatically with the independent variables associated with experimental design, it seems that the simplified statistical tool developed could be applied to experimental design for many applications of CRMs (including frequency-response analysis), however caution should be exercised at concentration levels, analysis timesteps, or concentration profiles, significantly different from those examined in this paper.

### **Acknowledgements**

This work was supported by the Director, Office of Energy Research, Office of Health and Environmental Research of the U.S. Department of Energy under Contract DE-AC03-76SF00098. Support for Yves Bonnefous's research participation was granted by ENTPE of Lyon, France.

We are thankful to Francis Allard of INSA de Lyon (France), Gerard Guarracino of ENTPE (France), William Nazaroff and Ann Kalinowski of UC Berkeley, and Lev Ruzer of LBL for their comments on the report. We also appreciate the assistance of Darryl Dickerhoff and Brian Smith of LBL in computer programming and laboratory experimentation.

## REFERENCES

- Y. Bonnefous, "Le radon dans l'habitat mesure en continu de la concentration en radon", Diplome d'Etudes Approfondies, Presente a l'I.N.S.A. de Lyon, Laboratoire d'accueil: L.A.S.H. - Ecole Nationale des Travaux Publics, Vaux-en-Velin, France, September 1989.
- A. Busigin, A.W. Van Der Vooren and C.R. Phillips, "Interpretation of the response of continuous radon monitors to transient radon concentrations", *Health Physics* 37 (November), pp. 659-667, 1979.
- W.J. Fisk, S. Flexser, A.J. Gadgil, H.-Y. Holman, M.P. Modera, T.N. Narasimhan, T. Nuzum, K.L. Revzan, R.G. Sextro, A.R. Smith, Y.W. Tsang, and H.A. Wollenberg "Monitoring and Modeling for Radon Entry Into Basements: A Status Report for the Small Structures Project", September 1989, Lawrence Berkeley Laboratory Report, LBL-27692.
- Goldanskii, *Counting Statistics of Nuclear Particles*, International Monograph Series, Hindustan Publishing Corporation INDIA-6, 1962.
- J.W. Thomas and R.J. Countess, "Continuous radon monitor", *Health Physics* 36 (June), pp. 734-738, 1979.
- S. Watnick, N. Latner and R.T. Graveson, "A  $^{222}\text{Rn}$  monitor using  $\alpha$  spectroscopy", *Health Physics* 50(5), pp. 645-646, 1986.

## APPENDIX: Simplified Analysis of Covariance Impacts on Uncertainty Estimates

The following is a two-time-step analysis with Equation 1 of a single-daughter decay series.

For a two-timestep analysis, Equation 1 can be reexpressed in more compact notation as:

$$C_{Rn,2} = A Y_2 - B C_{Rn,1} \quad (\text{A.1})$$

where:

$A$  is a positive constant equal to  $\frac{1}{0.037 V a \xi \tau}$ , and

$B$  is a positive constant equal to  $h_1(1 - \frac{1}{a})$

However, the number of decays in the second interval,  $Y_2$ , is equal to the sum of the radon decays in that interval and the decays of radon daughters deposited in that interval and in earlier intervals. For simplicity, we will only use one previous time interval for our analysis, including the effects of earlier intervals into a parameter denoting the fraction of deposited daughters associated with the concentration in that interval. The results would be qualitatively similar for a complete analysis, and needlessly more complex for the qualitative arguments being presented herein. Thus, the number of decays in the second interval,  $Y_2$ , can be expressed as:

$$Y_2 = Z_2 + D E C_{Rn,1} \quad (\text{A.2})$$

where:

$Z_2$  is the number of radon decays in interval 2 plus the radon daughter decays in interval 2 due to radon decays in interval 2,

$D$  is a random variable quantifying the number of decays in period 2 of radon daughters deposited on the cell walls at the end of period 1, and

$E$  is the ratio of deposited radon daughters at the end of period 1 to the average radon concentration in period 1.

If we now substitute Equation A.2 into Equation A.1, we obtain:

$$C_{Rn,2} = A Z_2 + (D E A - B) C_{Rn,1} \quad (\text{A.3})$$

Remembering that  $D$  is a random variable, the variance of  $C_{Rn,2}$  based upon Equation A.3 can be shown to be:

$$\sigma_{C_{m,2}}^2 = A^2 \sigma_{Z_2}^2 + A^2 E^2 \bar{C}_{Rn,1}^2 \sigma_D^2 + (\bar{D} E A - B)^2 \sigma_{C_{m,1}}^2 + A^2 E^2 \sigma_D^2 \sigma_{C_{m,1}}^2 \quad (\text{A.4})$$

where:

$\bar{C}_{Rn,1}$  is the expected value of the concentration in interval 1, and

$\bar{D}$  is the expected value of the number of decays in period 2 of radon daughters deposited on the cell walls at the end of period 1.

If, on the other hand, if the variance of  $C_{Rn,2}$  were calculated based on Equation 1 assuming that  $Y_2$  were independent of  $C_{Rn,1}$ , the computed variance would be:

$$\sigma_{C_{m,2}}^2 = A^2 \sigma_{Y_1}^2 + B^2 \sigma_{C_{m,1}}^2 \quad (A.5)$$

If we now compute the variance of  $Y_2$  based on Equation A.2:

$$\sigma_{Y_1}^2 = \sigma_{Z_1}^2 + E^2 \bar{C}_{Rn,1}^2 \sigma_D^2 + \bar{D}^2 E^2 \sigma_{C_{m,1}}^2 + E^2 \sigma_D^2 \sigma_{C_{m,1}}^2 \quad (A.6)$$

the variance that would be observed based upon the assumption of no correlation is:

$$\sigma_{C_{m,2}}^2 = A^2 \sigma_{Z_1}^2 + A^2 E^2 \bar{C}_{Rn,1}^2 \sigma_D^2 + (\bar{D}^2 E^2 A^2 + B^2) \sigma_{C_{m,1}}^2 + A^2 E^2 \sigma_D^2 \sigma_{C_{m,1}}^2 \quad (A.7)$$

Remembering that  $A$ ,  $B$ , and  $\bar{D}$ , are all positive, the simple algebraic inequality:

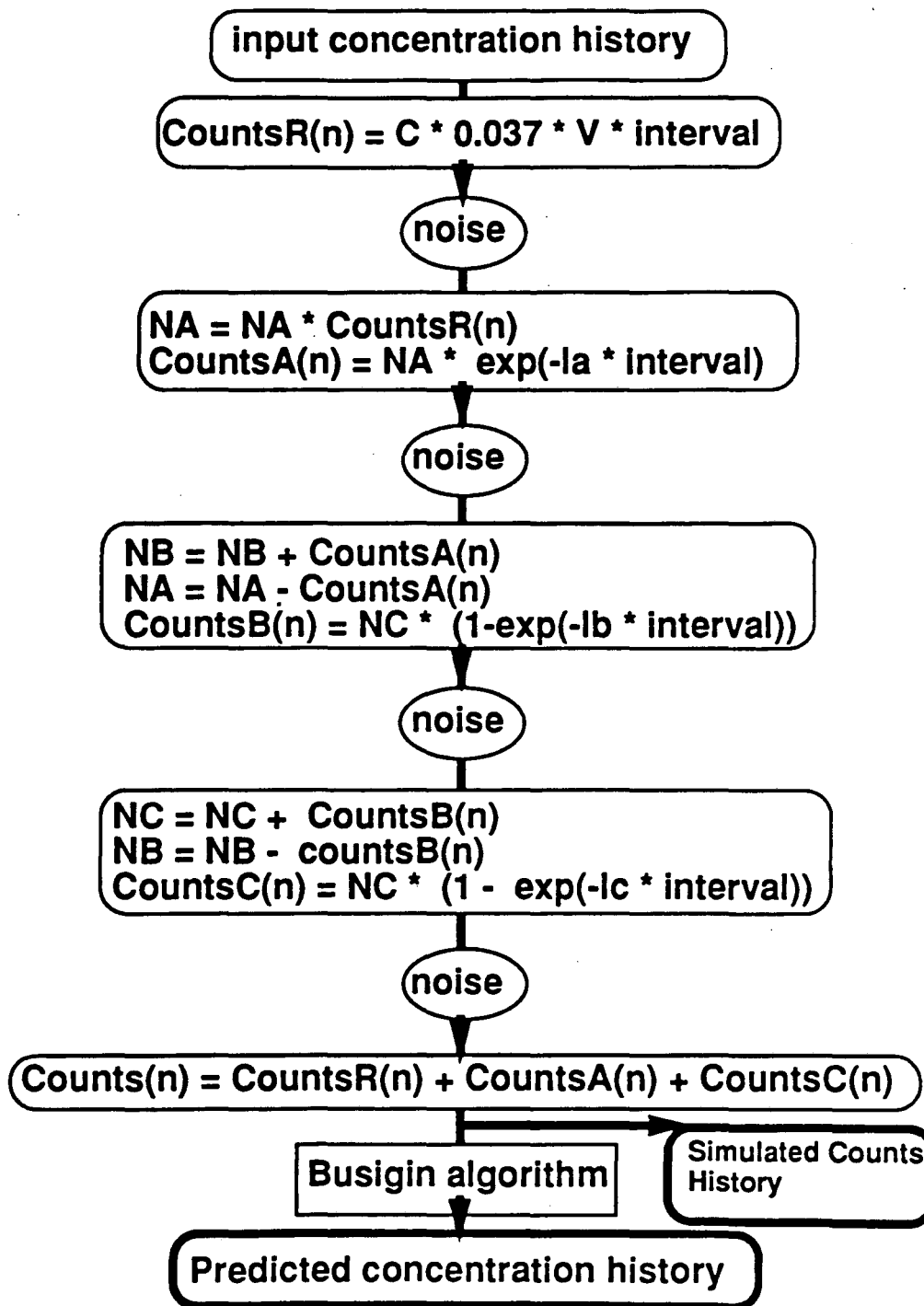
$$(\bar{D}^2 E^2 A^2 + B^2) > (\bar{D} E A - B)^2 \quad (A.8)$$

it is clear that the independence-based variance estimates will always be larger than the true variance. More specifically, the ratio of the no-correlation variance estimate to the true variance can be expressed as:

$$\frac{\sigma_{nocor}^2}{\sigma_{true}^2} = 1 + 2 \bar{D} E A B \frac{\sigma_{C_{m,1}}^2}{\sigma_{true}^2} \quad (A.9)$$

The relationship in Equation A.9 was found to be consistent with the true and no-correlation estimates of the radon concentration variances obtained from the simulations of the soil-gas sampling problem examined in the body of the paper.





Note : Noise is generated from a Binomial distribution

Figure 1. Flow chart for Monte-Carlo simulation program used to estimate distributions of alpha-decay histories based upon a known input radon concentration history.

### Predicted Radon concentration

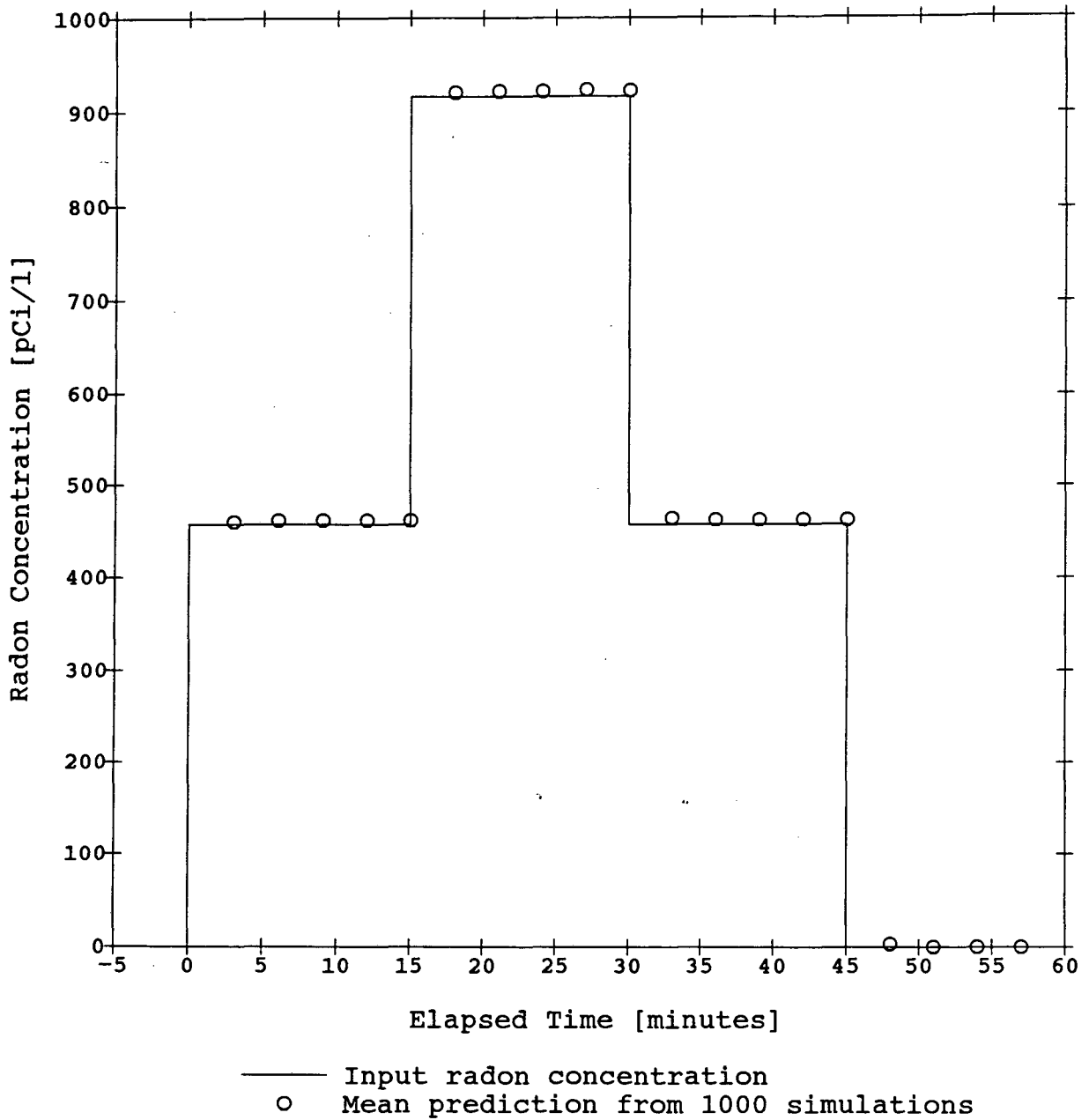


Figure 2. Radon concentration history input to decay simulation program, and radon concentration estimates obtained with forward-marching analysis procedure (Equation 1) applied on a three-minute time interval to the simulated decays from 1000 simulations.

### Coefficient of Variation of Radon Concentration

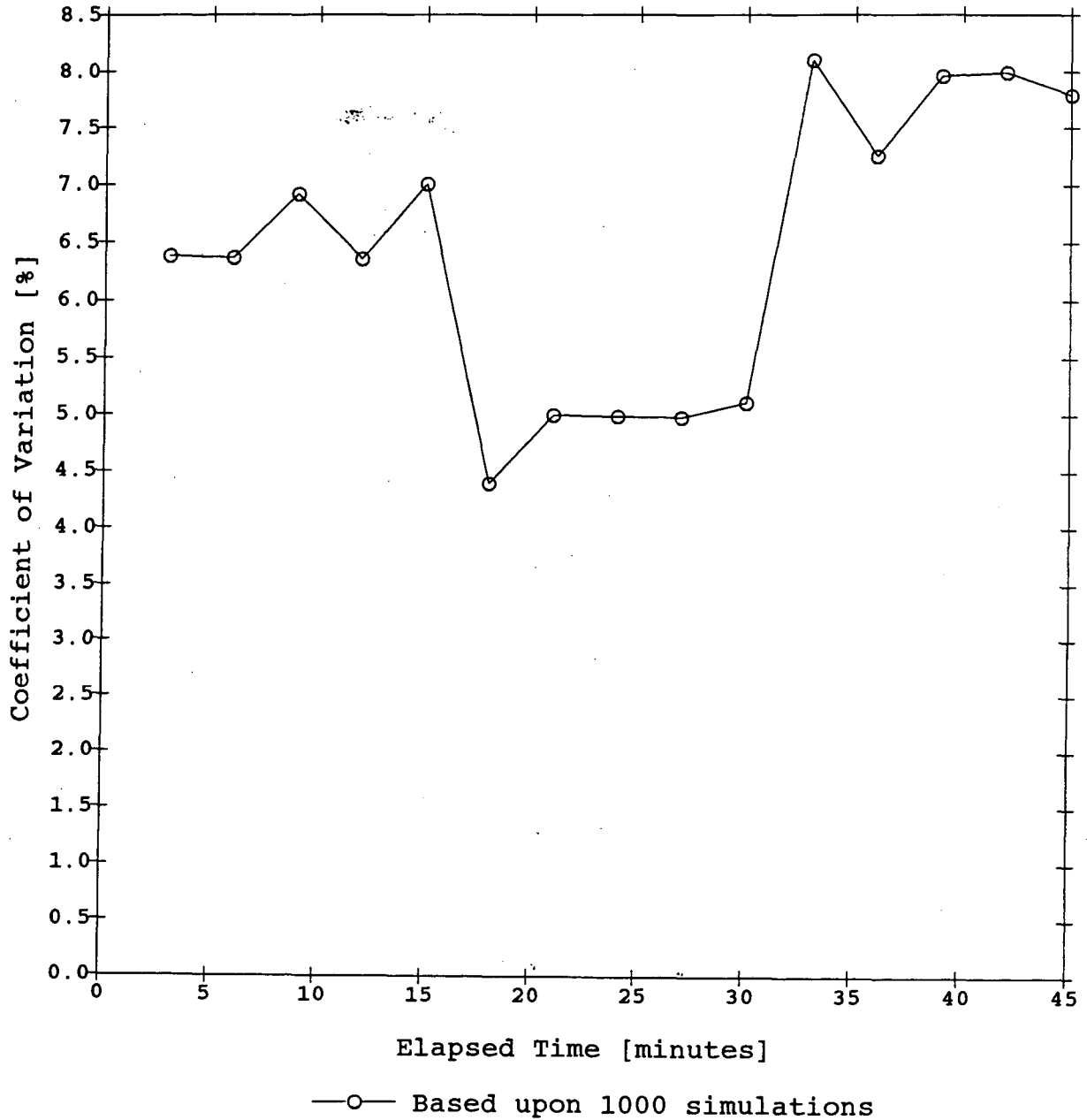


Figure 3. Coefficient of variation of the radon concentration estimates obtained with the forward-marching analysis procedure (Equation 1) applied on a three-minute time interval to the simulated decays from 1000 simulations on the input concentration history depicted in Figure 2.

### Coefficient of Variation of Radon Concentration

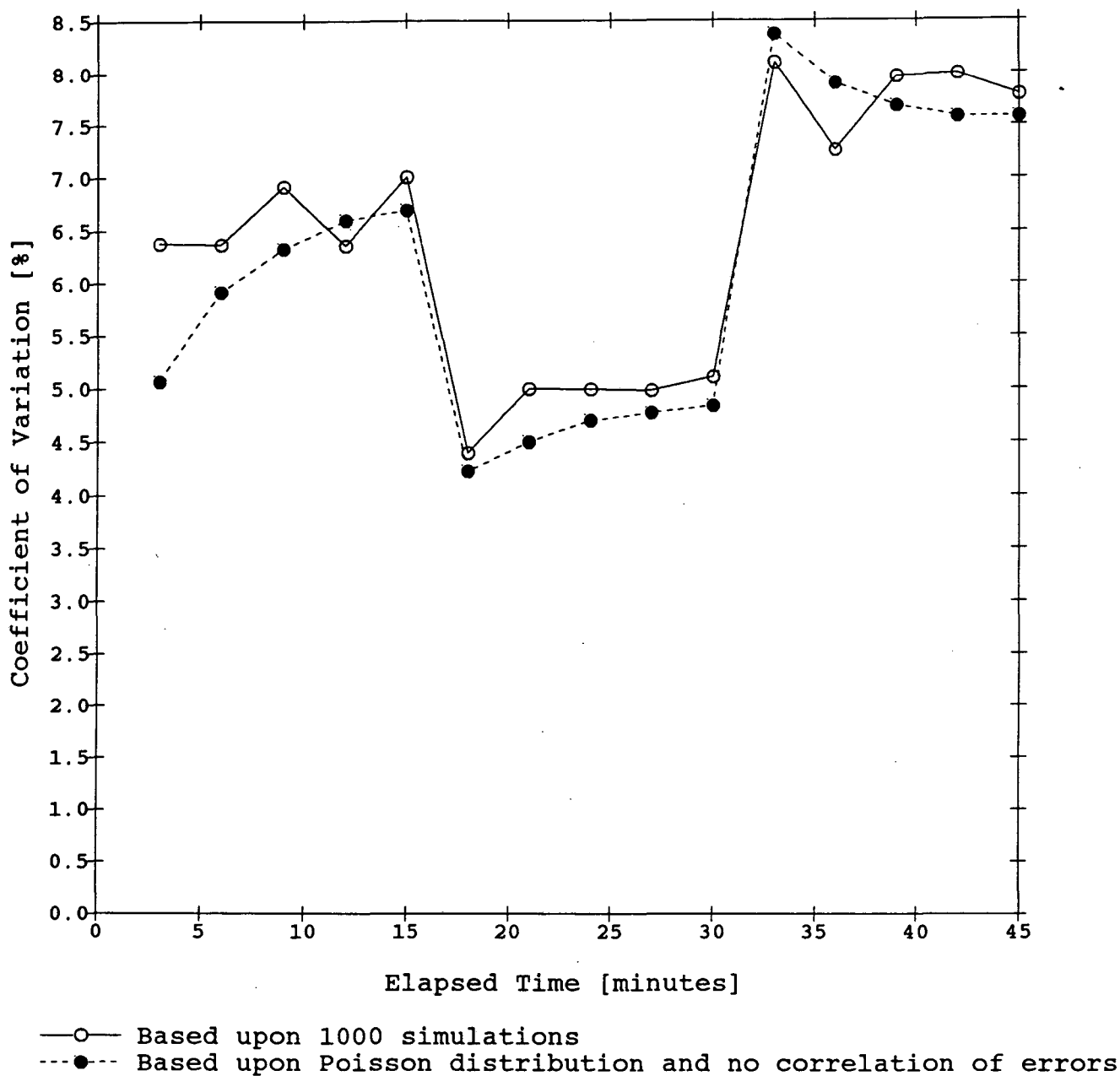


Figure 4. Coefficient of variation of the radon concentration estimates obtained from the simulated data (same as Figure 3), and coefficient of variation estimates based on Equation 4 (i.e., no correlation between intervals) using Poisson distributions for decay variances.

### Standard Deviation of Alpha Disintegrations

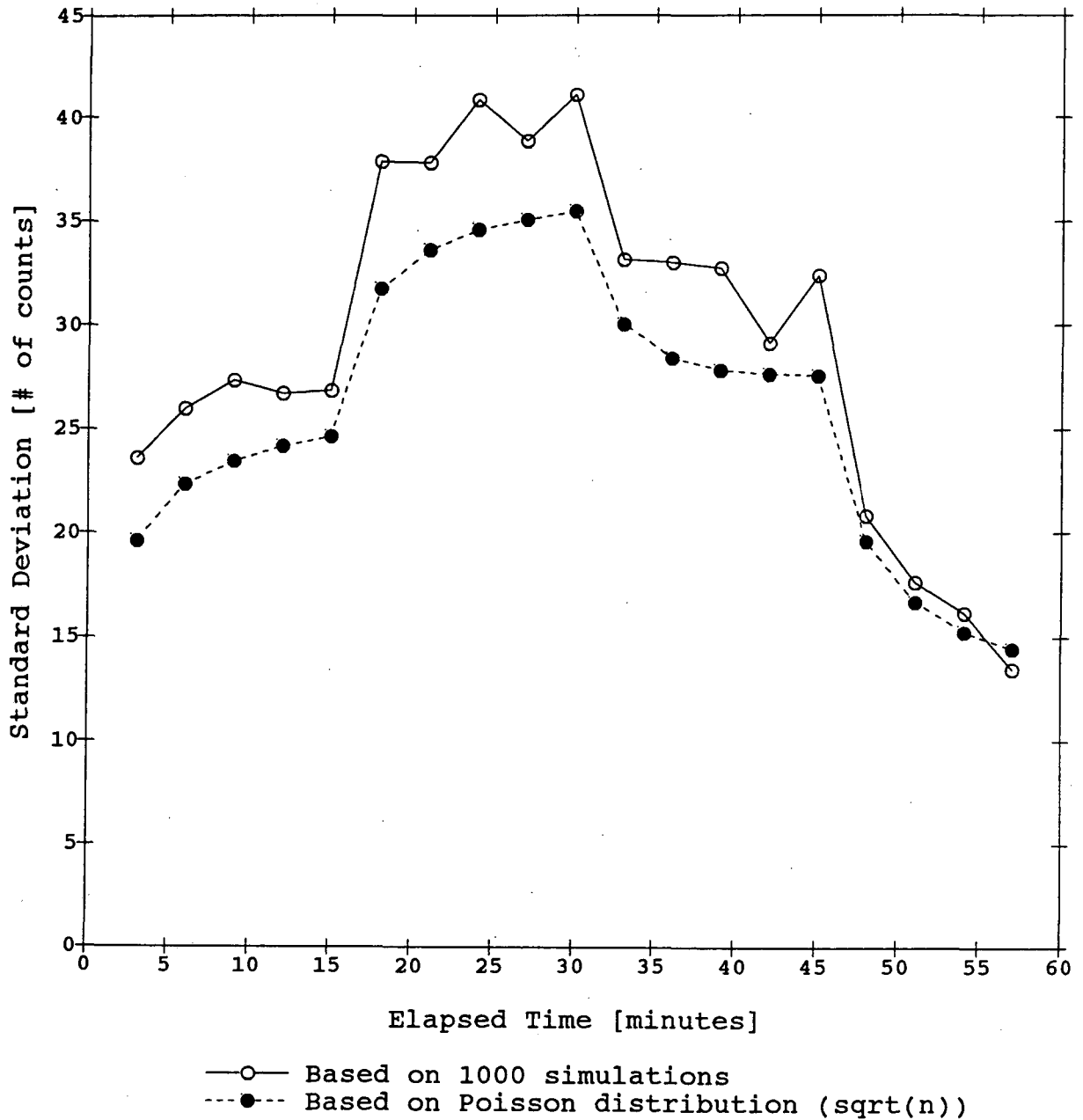
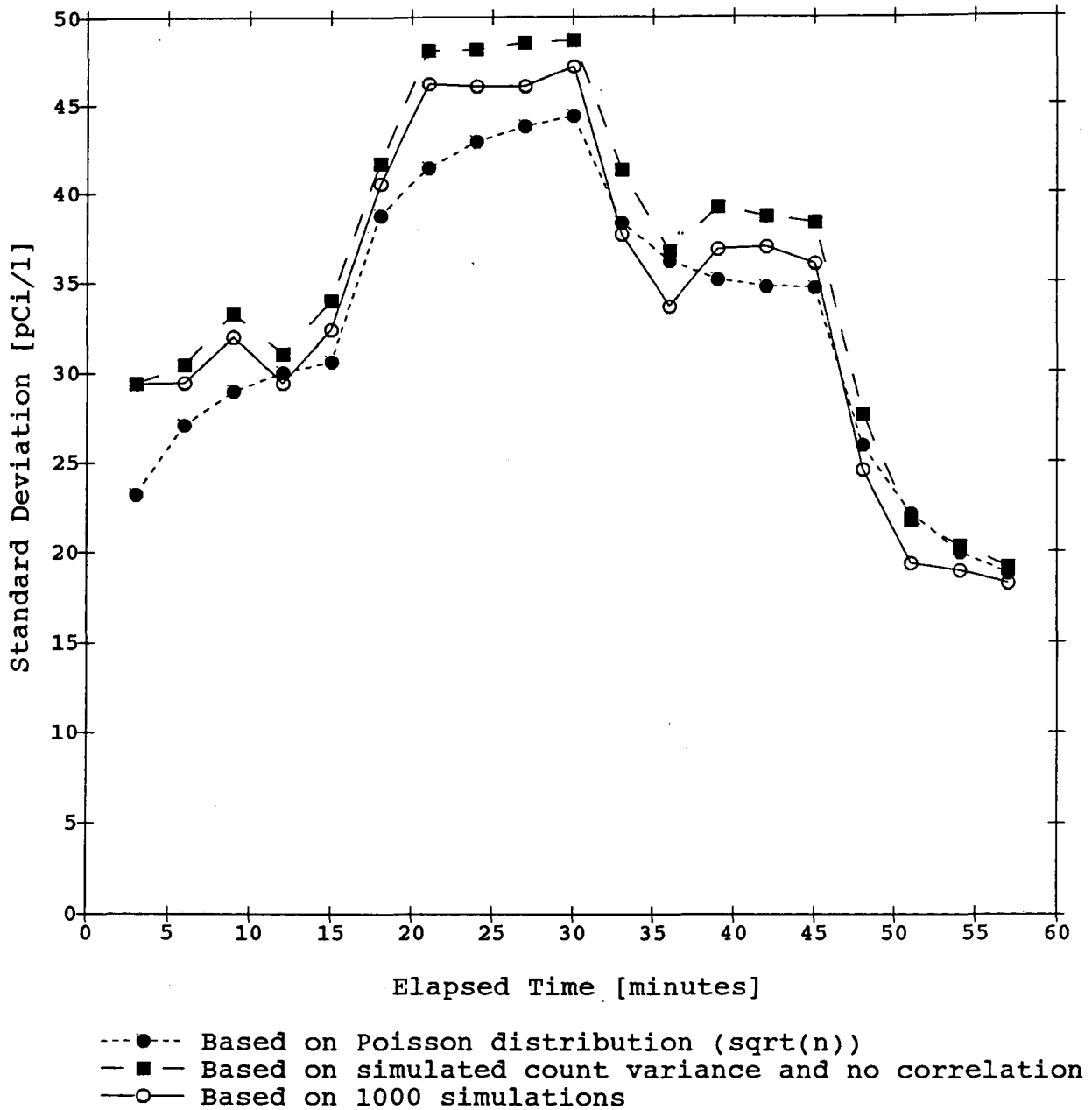


Figure 5. Standard deviation of the number of detected decays obtained with the forward-marching analysis procedure (see Figure 2), and standard deviation of the number of detected decays predicted by assuming decays to be Poisson distributed.

### Standard Deviation for Radon Concentration



**Figure 6.** Absolute standard deviation of the radon concentration estimates obtained from the simulated data (Figures 2 and 3), estimates based on Equation 4 with Poisson distributions for decay variances (Figure 4), and estimates based on Equation 4 with simulated decay variances (data from Figure 5).

## Coefficient of Variation of Radon Concentration

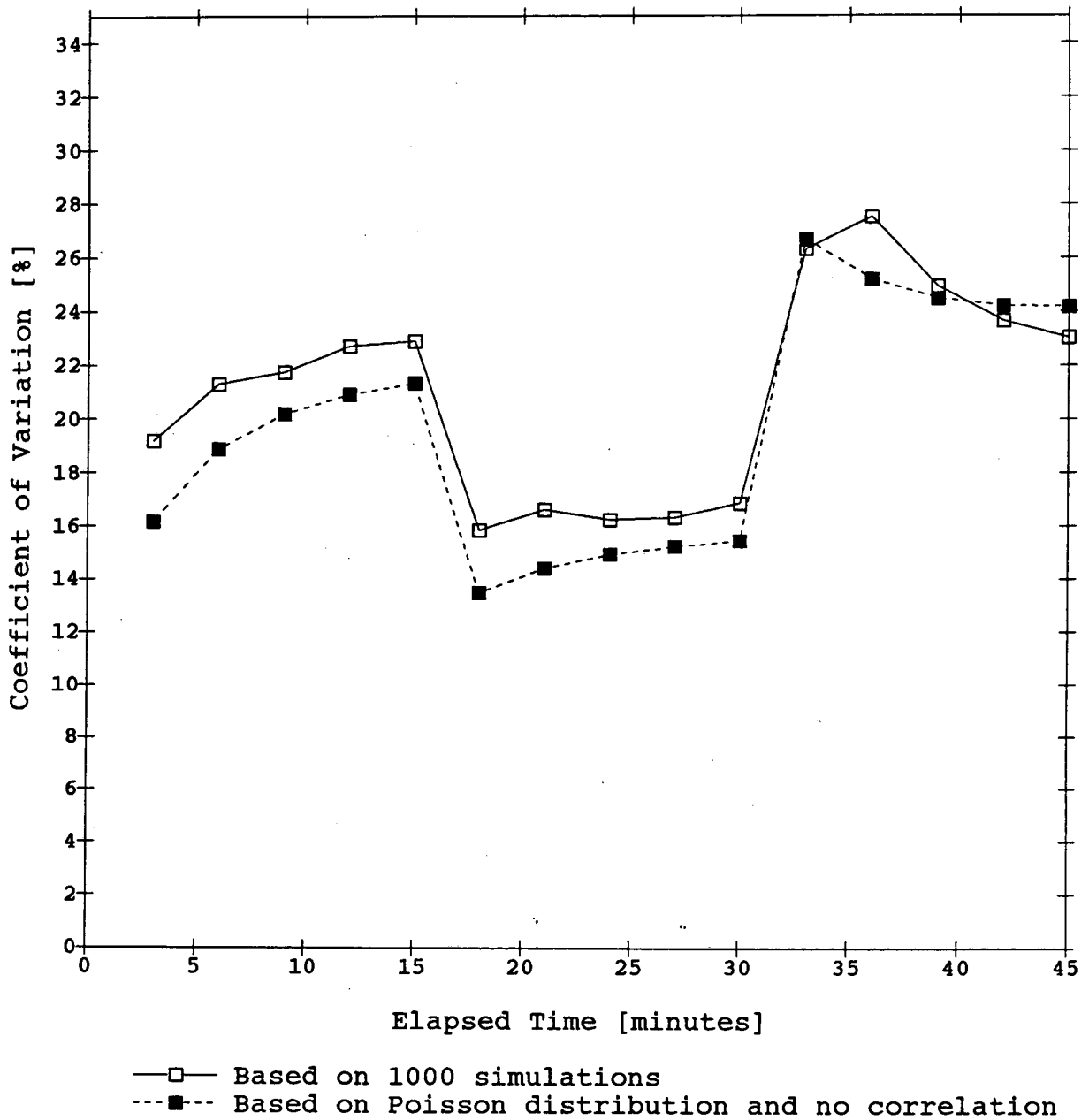


Figure 7. Coefficient of variation of the radon concentration estimates obtained from the simulated data (same input profile as Figure 2, except one order of magnitude lower concentration levels), and coefficient of variation estimates based on Equation 4 (i.e., no correlation between intervals) using Poisson distributions for decay variances.

### Coefficient of Variation of Radon Concentration

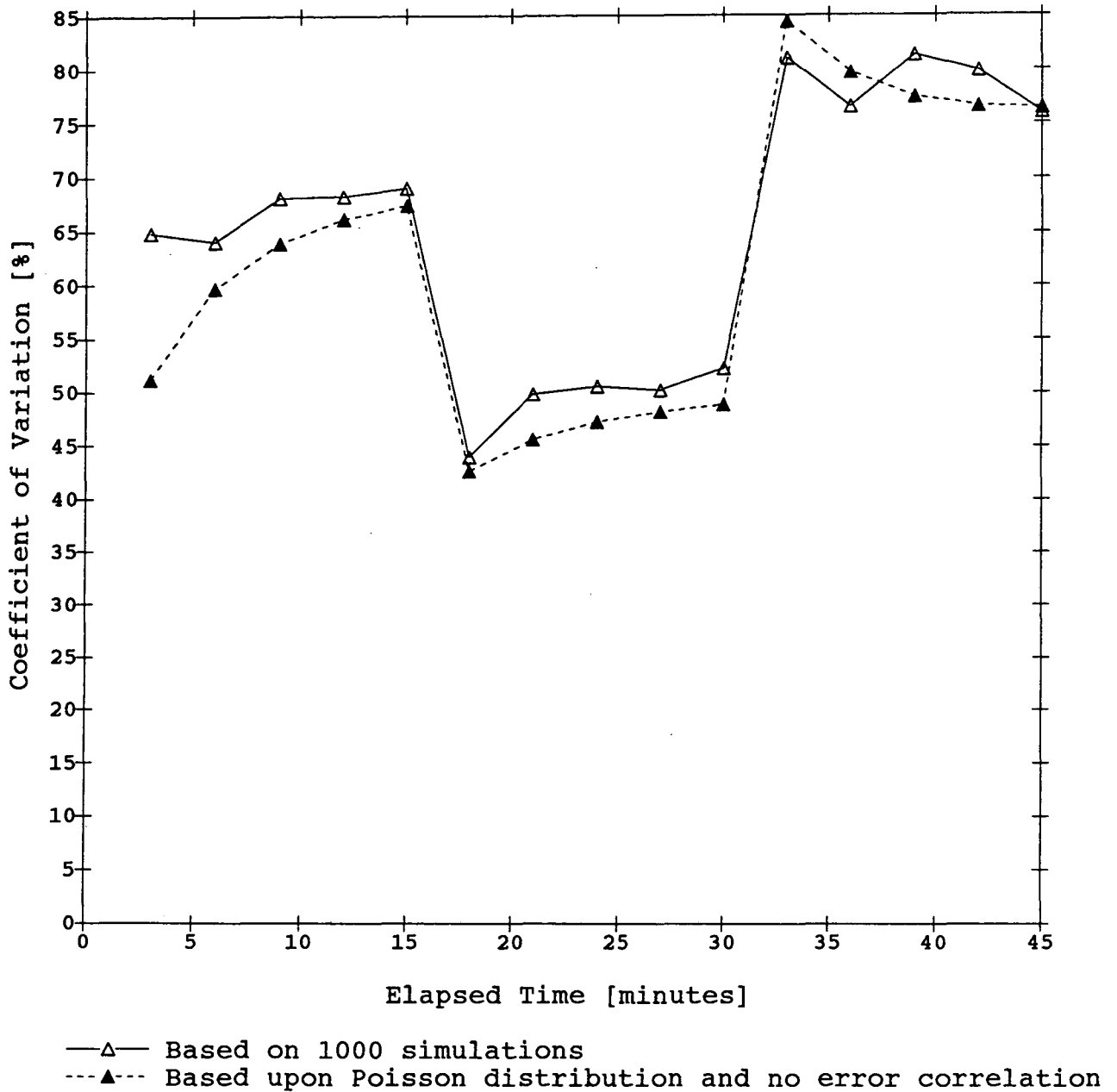


Figure 8. Coefficient of variation of the radon concentration estimates obtained from the simulated data (same input profile as Figure 2, except two orders of magnitude lower concentration levels), and coefficient of variation estimates based on Equation 4 (i.e., no correlation between intervals) using Poisson distributions for decay variances.



## Radon Concentration Bias for Forward-Marching Algorithm

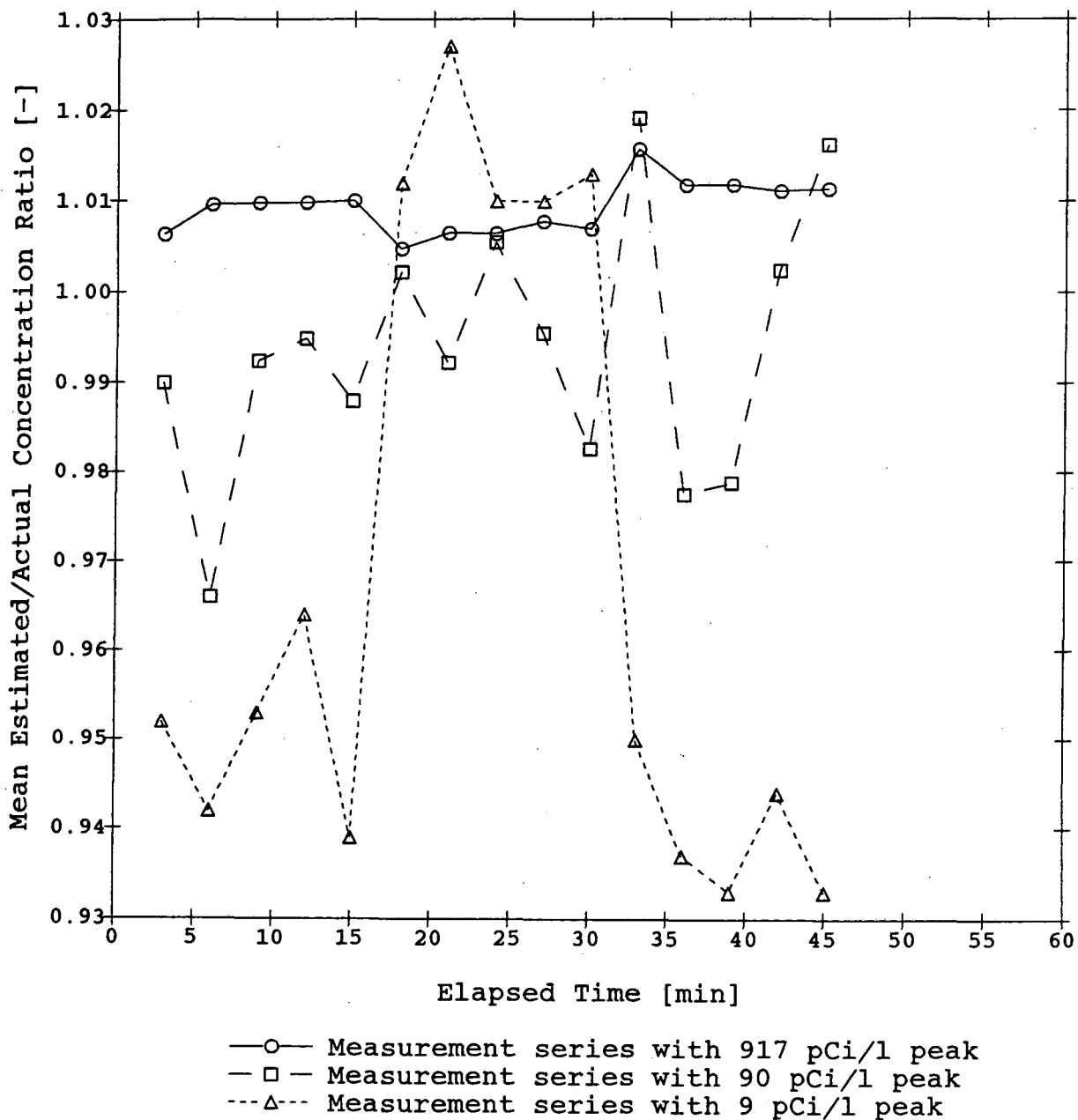


Figure 9. Bias of concentration estimates based on forward-marching analysis procedure (Equation 1) for three-step input concentration histories similar to that shown in Figure 2. Quoted biases are for peak concentration levels of 900 pCi/l, 90 pCi/l, and 9 pCi/l.

# Laboratory Test of Radon Concentration Tracking

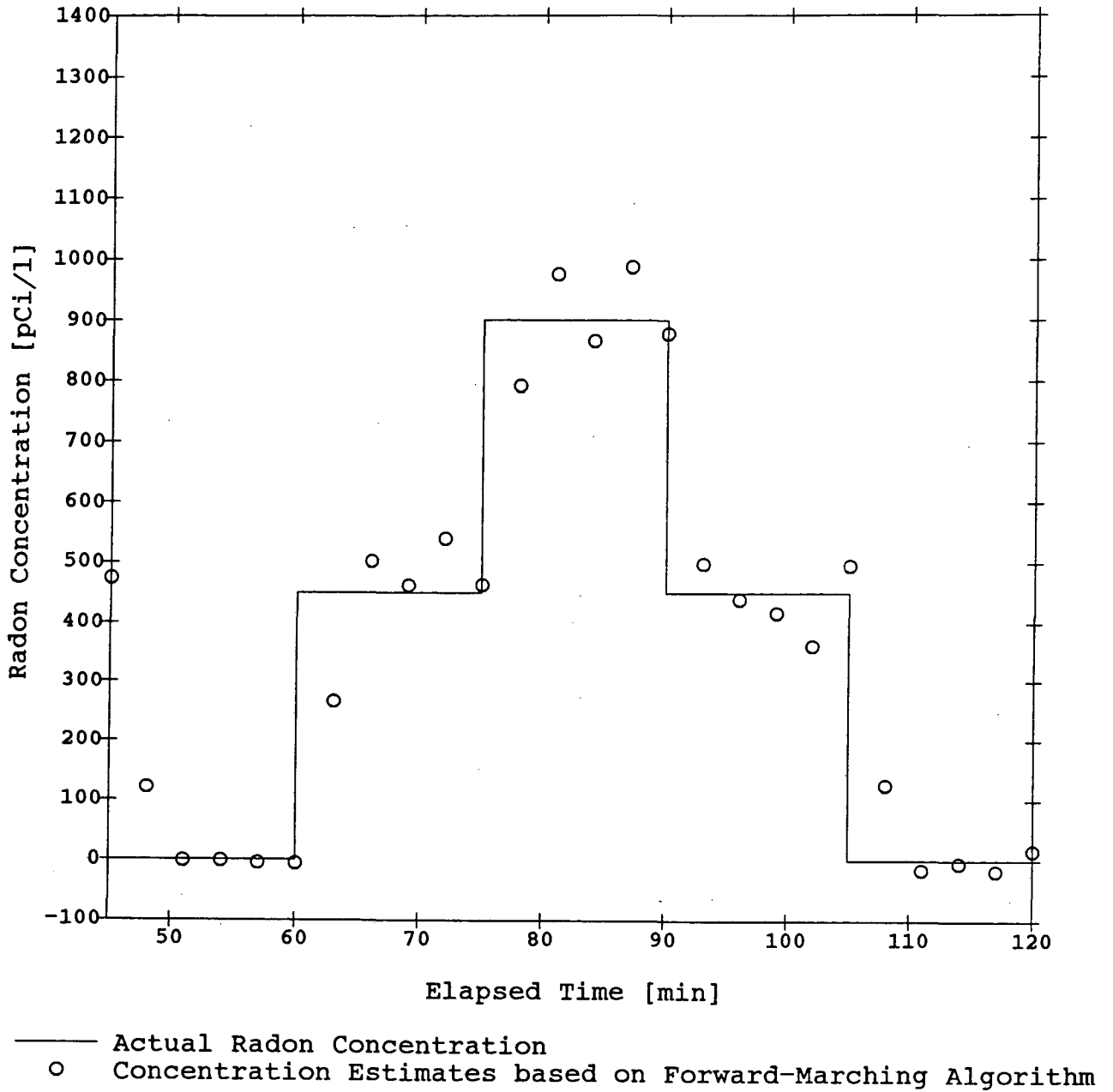


Figure 10. Radon concentration history input to laboratory Continuous Radon Monitor, and radon concentration estimates obtained with forward-marching analysis procedure (Equation 1) applied on a three-minute time interval to the measured decays.

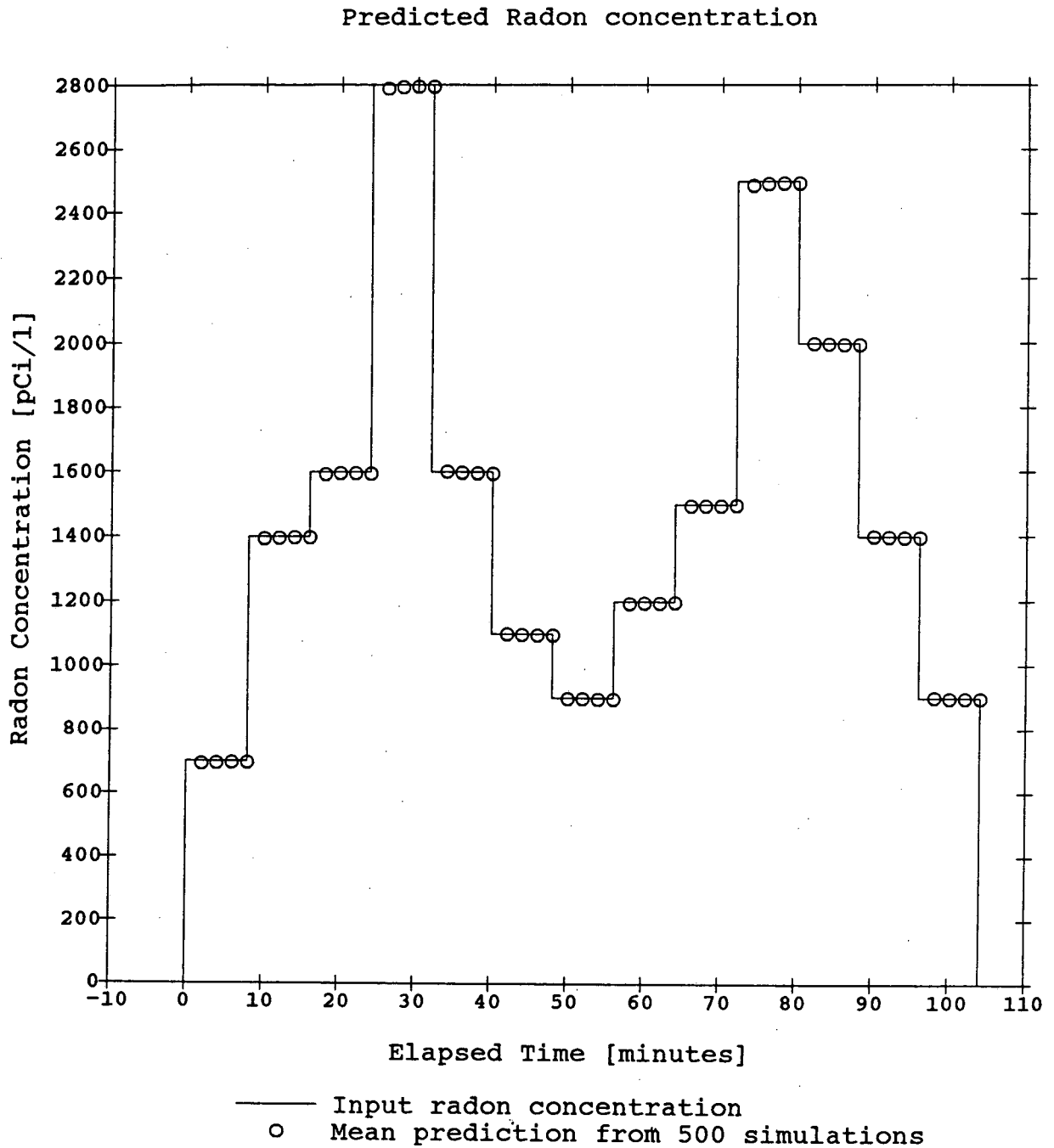


Figure 11. Radon concentration history input to decay simulation program, and radon concentration estimates obtained with forward-marching analysis procedure (Equation 1) applied on a two-minute time interval to the simulated decays from 500 simulations.

## Standard Deviation for Radon Concentration

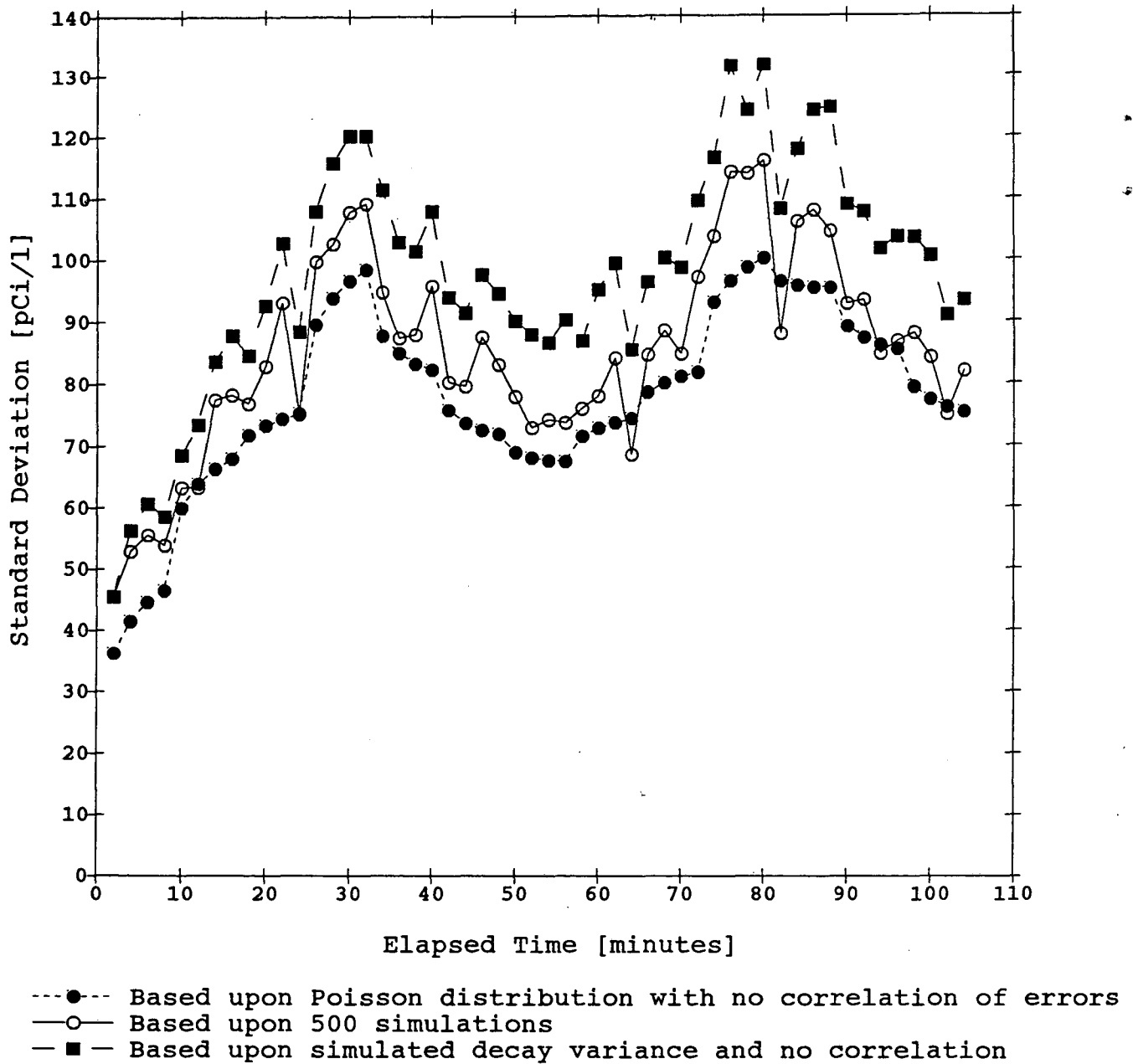


Figure 12. Coefficient of variation of the radon concentration estimates obtained from the simulated data for the profile in Figure 11, and coefficient of variation estimates based on Equation 4 (i.e., no correlation between intervals) using Poisson distributions for decay variances.

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