ESTIMATING STRENGTHS OF INDIVIDUAL RADIOISOTOPES IN A MULTIPLE-ISOTOPE SOURCE

The Imprecision of the Estimates Is Partitioned Into Poisson, Sampling, and Mechanical Variations

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The imprecision of the Estimates Is Partitioned Into Poisson, Sampling, and Mechanical Variations

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by

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ABSTRACT

In work related to radiation shielding, the use of radioisotope techniques, and activation analysis, an experimenter must often analyze counting data where counts are caused by the natural background and by the decay of more than one radioisotope. In this report a procedure is developed for estimating the strength of each isotope at different times from several decaying radioactive samples of a single multiple-isotope source. In addition, the procedure provides a method for placing confidence limits on the strengths and a method for partitioning the imprecision of estimating the strengths into three principal causes: Poisson variation, sampling error, and residual error (called mechanical error).

An operational FORTRAN I1-D computer program, SAND, implements the procedure. The procedure and program were tested by using fictitious data with known properties as inputs. The results of the simulation were in reasonable agreement with the theoretical values.
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INTRODUCTION

In various nuclear physics laboratories there exist dilute solutions of mixtures of a few radioactive isotopes for which an estimate is desired of the expected rate of radioactive decay* in counts per minute per given aliquot quantity of the solution at some given historic instant of time.** Usually it is desired to break this total estimate into the parts associated with each isotope.

To accomplish this, it is customary to take several aliquot samples and observe counts from each of these during measured short-time periods at several different ages (lengths of time from the desired historic instant \( t_0 \) *** to the beginning of the measured period). Because many materials in nature, and particularly in a nuclear physics laboratory, are experiencing some radioactivity, the observing device (Geiger counter) will count slowly, even with no sample, thus, it is necessary to record counts of this background during relatively long, measured time periods near the time when the counter is to be used for sample counting. The purpose of this background count is to correct the observed counts to obtain apparent sample counts.

Given all the foregoing data, the following questions arise:

1. At the desired historic instant, designated \( t_0 \), what is the estimate of decay rate?

2. At \( t_0 \) what is the breakdown of the total decay rate into the various isotope sources?

The laboratory data used to make the foregoing estimates will be subject to the following three sources of imprecision:

a. Sampling error; that is, inability to place exactly the same amount of material in exactly the same position on the watch glass each time in each finally prepared sample ready for counting and recounting.

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* The terms "rate of decay" and "strength at some instant of time" are used interchangeably throughout this report.


*** The reader is referred to the Glossary of Symbols on a foldout page at the back of this report.
b. Poisson distributed variation; that is, the fact of nature that a multiplicity of samples of exactly the same size and strength (same expected counts per minute) will show variation among themselves in the actual number of radioactive events in a given minute. The nature of this Poisson fluctuation is known.

c. All other uncorrected sources of imprecision lumped under the arbitrary heading "mechanical variation." These sources may include slight positioning variation, extraneous counting errors other than from background, temperature variation of mechanical setup, slight gain or loss of material in "dried" sample between successive observations, and inexactness of dead-time, \( r \), correction used.

Because of a, b, and c the estimates 1 and 2 will not be exact. This raises the further questions:

3. How imprecise is the estimate of total decay rate at \( t^*_0 \)?

4. How imprecise is the estimate of each isotope decay rate at \( t^*_0 \)?

An incidental result of attempting to supply an answer to 3 will be an approximate answer to

5. How much of the imprecision in estimating decay rate at \( t^*_0 \) is due to:
(a) sampling error, (b) Poisson error, and (c) mechanical error?

It is the purpose of this report to develop a computational procedure for approximating answers to 1, 2, 3, 4, and 5. In short, these five questions constitute the problem to be solved in this report.

Briefly, the computational procedure will consist mainly of a sequence of parallel-weighted linear-least-squares curve fits. On each iteration each datum is weighted inversely proportional to its current estimated variance. The repetitions are terminated when the last two current estimates of the mechanical error standard deviation differ by less than some arbitrarily small value. The sampling error standard deviation is estimated by an additional iterative procedure using the foregoing results.

It is assumed that the number of isotopes in the solution and their decay constants are known; that is, the procedure does not estimate the number of isotopes in the solution, nor does it estimate the decay constants since better methods exist for this purpose.
DESCRIPTION OF MATHEMATICAL CONCEPTS

General

Several aliquot samples (total of I samples) are taken from a liquid solution of radioactive material, placed on I watch glasses, and allowed to evaporate. Then each sample is observed several times by means of a detector (J, times for the ith sample, i = 1, 2, ..., I) in a fixed counting system with fixed geometry. The number of counts recorded during a period of duration Tij minutes will be designated Cij for the ith sample and its ith observation (i = 1, 2, ..., I; j = 1, 2, ..., J,). The age of the sample in minutes, reckoned from t0, an arbitrary historic instant of interest when t = 0, to the beginning of the period of duration Tij, will be designated tij.

The assumed known number of radioactive isotopes in the source solution is L. The known decay constant in reciprocal minutes from the ith isotope is designated λj (j = 1, 2, ..., L).

Because the detector records counts from background radioactivity in addition to those strictly from the sample under observation, an effort is made to correct for this by observing background with the same instrument at a time near tij. The length of this observation and the number of counts observed are, respectively, T* and C*. In practice "near" may mean merely in the same working day. Thus, a single background reading may be used to correct several observations and give rise to T* and C* combinations which are numerically identical for various i and j combinations.

In the sampling process an effort is made to place the same amount of material on each watch glass in the same relative position. There is a slight variation in this process, and the effective quantity will be assumed to be a normally distributed random variable with mean Q and standard deviation σs so that σs may be construed as a proportional standard deviation of error. For example, σs = 0.01 would indicate a sampling error standard deviation of 1% of the expected effective sample size.

A period of time elapses between successive observations of the same sample, a period in which laboratory conditions change. An effort is made to make all observations with exactly the same mechanical and electrical arrangement. Nevertheless, there may be a slight variation in this process which affects the actual Cij recorded. If one could imagine a fictitious sample (sample number h) which would experience exactly the same number of radioactive events every minute, regardless of age, then the successive observations Ch1, Ch2, Ch3, ... with fixed Th and no background noise will be assumed to be items of a normally distributed random variable with mean M and standard deviation Mσm. Now σm may be construed as a proportional
error due to all causes (called mechanical) other than sampling error and errors that result from the Poisson phenomenon (assumed independent of mechanical errors).

If one now imagines an infinite number of samples of the same solution of radioactive material and an infinite number of observations of each, all between \( t^*_{\infty} \) and \( t^*_{\infty} + T \), for fixed \( T \), then the background-corrected counts per minute would constitute a two-dimensional array of data entries, designated here as \( \eta_{ij} \), infinite in length and infinite in width. The row (sample) mean of such a fictitious array of data will be designated \( \mu_{ij}(0,T) \) counts per minute. The portion of this due to the \( \ell \)th isotope will be designated \( \mu_{ij}(0,T) \), where

\[
\mu_{ij}(0,T) = \sum_{\ell=1}^{L} \mu_{ij}(0,T)
\]

If the average of the infinity of the \( \mu_{ij}(0,T) \)'s is designated as \( \mu_{0}(0,T) \), then question 1 of the Introduction asks for \( \mu^0_{ij} \), an estimate of \( \mu^0_{ij} \), where zero bias error is assumed and

\[
\mu^0_{ij} = \lim_{T \to 0} \mu_{ij}(0,T)
\]

Similarly, question 2 asks for the \( \mu^0_{ij} \)'s, which are estimates of the \( \mu^0_{ij} \)'s, where

\[
\mu^0_{ij} = \lim_{T \to 0} \mu_{ij}(0,T)
\]

The mean of the \( i \)th sample infinite row "at \( t = 0 \)" was designated as \( \mu_{i}(0,T) \), where it is supposed that

\[
\mu_{i}(0,T) = \lim_{j_i \to \infty} \frac{1}{j_i} \sum_{j=1}^{j_i} \eta_{ij}
\]

and that

\[
\mu_{ij}(0,T) = \lim_{|i| \to \infty} \frac{1}{|i|} \sum_{i=1}^{i} \mu_{ij}(0,T)
\]

By an observation "at \( t = 0 \)" is meant an observation of small duration containing \( t = 0 \) such that the expected counting rate at \( t = 0 \) is equal to the average expected counting rate during the time interval \( T \). Recall that this is merely a fiction inserted to clarify concepts.
Briefly, the problem is to obtain estimates of $\mu_1^0, \mu_2^0, \ldots, \mu_L^0, \sigma_0$, and $\sigma_m$; to compute confidence intervals expressing the imprecision in knowledge of the values of $\mu_1^0, \mu_2^0, \ldots, \mu_L^0$, and $\mu_0^0$; and finally to separate the imprecision in estimating $\mu_0^0$ into the three sources: (a) sampling error, (b) Poisson variation, and (c) mechanical variation. To perform these estimates, it will be necessary to start the computation with $\tilde{\sigma}_{m_0}, \lambda_1^0, t_j^0, T_j^0, C_i^0, t_{ij}^0, T_{ij}^0,$ and $C_{ij}^0$, where $\ell = 1, 2, \ldots, L; j = 1, 2, \ldots, J_i; i = 1, 2, \ldots, I$. It should be carefully noted that this whole report is concerned solely with imprecision and not laboratory mean bias over the testing period. Any such bias in $\mu_0^0$ will be carried over into $\tilde{\mu}_0^0$, and as such will remain unknown in this computation.

To reach a "solution" to the problem, least-squares estimates of $\mu_0^0, \ldots, \mu_L^0$ for each sample will be found by using weighted linear regression. For a minimum-variance estimate, the weight used for each datum should be inversely proportional to the variance in knowledge (ignorance) of the "true" (biased by the particular sample bias) value represented by the datum. Given a particular sample, this variance for each datum is the sum of those caused by Poisson phenomenon and mechanical variation. (The last two statements taken together contain a "right inexactness stemming from a small interdependency between those elements of data which were computed using a common background reading.)

Ideally the weight of a datum should be based upon the true variance for that datum; however, only an estimate of the variance is available. The curve-fitting method depends upon the weights, which in turn depend upon the fitted parameters. Thus, it is necessary to iterate on weights and fitted parameters simultaneously for all samples to convergence of the estimated mechanical error. This leads to a stable $\tilde{\sigma}_{m_i}$ and I stable-weighted least-squares curve fits.

Preliminary Theoretical Discussion

In the foregoing, the word "expected" has been used in its technical sense and as such does not mean "anticipated." For example, consider an infinitely large quantity of radioactive solution and the division of a large portion of this with no sampling or mechanical error into a large number $I$ of exactly equal-sized subquantities. Now imagine $I$ background-free detector setups with absolutely no variation between the setups. At the time $t_i^0$, initiate simultaneously the $I$ countings (one subquantity for each counter) and terminate them all simultaneously $T$ minutes later. This gives $I$ integer counts $C_i$ (for each counter) and terminate them all simultaneously $T$ minutes later. This gives $I$ integer counts $C_i$ (for each counter) and terminate them all simultaneously $T$ minutes later. This gives $I$ integer counts $C_i$. Let

\[ c = \frac{1}{I} \sum_{i=1}^{I} c_i. \]
If $I$ is allowed to increase indefinitely, $C$ will approach a stable value known as the expected value of $C$. Restated

$$E(C) = \lim_{I \to \infty} \bar{C}$$

Thus, the expected value of a random variable is merely the average of the numerical outcomes of an infinite number of trials. The infinite number of outcomes is the population associated with the random variable. A few outcomes are a sample of the population. In general the individual $C_i$'s will show considerable variation among themselves. This variation of the population of counts has been called the Poisson phenomenon. Indeed, the a priori probability that one particular counter will count exactly $h$ radioactive events during the period $t_0$ to $t_0 + T$ is given by the Poisson formula

$$P(C_i = h) = \frac{e^{-E(C)}(E(C))^h}{h!} \quad \text{for } h = 0, 1, 2, \ldots$$

Now if the variance of $C$ is defined to be

$$\sigma_C^2 = E\{[C - E(C)]^2\}$$

it will be found that

$$\sigma_C^2 = E(C)$$

or equivalently that the standard deviation of counts

$$\sigma_C = \sqrt{E(C)}$$

which is a very interesting property of a Poisson distributed random variable such as $C$. Restated

$$\sigma_C^2 = \lim_{I \to \infty} \frac{1}{I} \sum_{i=1}^{I} [C_i - E(C)]^2$$

Because counts per minute for no detector deadtime, $\tau = 0$, is $N = C/T$, $T$ merely plays the role of a scale factor with the following consequences:
\[
E(N) = E \left( \frac{C}{T} \right) = \frac{1}{T} E(C)
\]

\[
\sigma_N^2 = E \left\{ \left( \frac{C}{T} - E \left( \frac{C}{T} \right) \right)^2 \right\} = \frac{1}{T^2} E \left\{ (C - E(C))^2 \right\}
\]

\[
= \frac{\sigma_C^2}{T^2} = \frac{E(C)}{T^2} = \frac{E(N)}{T}
\]

From above
\[
\sigma_N = \frac{1}{T} \sigma_C = \frac{1}{T} \sqrt{E(C)} = \sqrt{\frac{E(N)}{T}}
\]

Of course in a real-world laboratory it is not possible to perform the idealized experiment of this section because there may be only one detector. In such a case several samples may be observed at different ages, or each sample may be reobserved at different ages. For a fixed period length of \(T\) minutes the expected counts will decrease or decay with age. Fortunately, the physical law governing this decay of expectation is known. If at time \(t_0\) the instantaneous expected background-free counts per minute are

\[
\mu_C = \sum_{\xi=1}^{L} \mu_C
\]

then at any future instant \(t\)

\[
\mu_C'(t) = \sum_{\xi=1}^{L} \mu_C e^{-\lambda_{\xi} t}
\]

and over a period from \(t\) to \(t + T\) the expected counts, assuming \(r = 0\) (perfect detector), are

\[
E(C) = \left[ \int_{t}^{t+T} \mu_C'(t) \, dt \right] + E(C^*)
\]

where \(C^*\) is the actual counts from background during \(T\).

Equivalently, the expected counts per minute solely from sample are
\[
E(\eta) = \frac{1}{T} \int_t^{t+T} \mu_r(t) \, dt = \sum_{\ell=1}^{L} \mu^0_{\ell} e^{-\lambda_{\ell} T} \left( \frac{1 - e^{-\lambda_{\ell} T}}{\lambda_{\ell} T} \right)
\]

Letting
\[
X(t, T; \lambda_{\ell}) = e^{-\lambda_{\ell} t} \left( \frac{1 - e^{-\lambda_{\ell} T}}{\lambda_{\ell} T} \right)
\]
then
\[
E(\eta) = \sum_{\ell=1}^{L} \mu^0_{\ell} X(t, T; \lambda_{\ell})
\]

For the \(j^{th}\) observation \((j = 1, 2, \ldots, J_j)\) of the \(i^{th}\) sample
\[
E(\eta_{ji}) = \sum_{\ell=1}^{L} \mu^0_{\ell} X(t_{ji}, T_{ji}; \lambda_{\ell})
\]
and
\[
\eta_{ji} = \sum_{\ell=1}^{L} \mu^0_{\ell} X(t_{ji}, T_{ji}; \lambda_{\ell}) + \epsilon_{ji}
\]

where \(\epsilon_{ji}\) is a random error consisting of the algebraic sum of the random Poisson sample error (reckoned from expected counts per minute), the random Poisson background error (reckoned from expected background counts per minute), and the random mechanical error.

Thus the estimates of
\[
E(\eta_{ji}) = \sum_{\ell=1}^{L} \mu^0_{\ell} X_{\eta_{ji}}
\]
can be obtained from a weighted least-squares fit of \(L + 1\) columns of data (each \(J_j\) long). The column headings are \(\eta_{ji}, X_{1i}, X_{2i}, \ldots, X_{Li}\) and the corresponding entries are \(\eta_{ji}, X(t_{ji}, T_{ji}; \lambda_{\ell}) = X_{\eta_{ji}}\), where \(j = 1, 2, \ldots, J_j\) and \(\ell = 1, 2, \ldots, L\). Each of the \(J_j\) lines of data will be weighted at each iteration inversely proportional to the last iterated estimate of \(E(\epsilon_{ji}^2)\). After studying the next section, the careful reader will note that the foregoing sentence is very slightly inexact. Because a single background reading may be used for more than one combination of \(i\) and \(j\), the next section deliberately modifies the method of weighting so as to reckon each background count once in the whole data-weighting computation system.
Clearly, one will never be able to perform enough countings to display the whole population of outcomes, but must be content to perform several tests, giving merely a sample of several numerical valued outcomes from the population. The average of the sample of outcomes is called a sample mean and as such constitutes an estimate of the population mean. For example

\[
\bar{C} = \overline{E(C)}
\]

In general this is the best available estimate without further testing. Nonetheless, it may be a poor estimate. It is customary to quantify the poorness or goodness of the estimate by computing a confidence interval at some expressed level of confidence for the quantity estimated. A 0.95 interval for \( E(C) \) is a range of possible values with specified lower and upper end points where the interval has the following interpretation: “The true value of \( E(C) \) is not known; however we are 95% sure that whatever its value may be it does lie in this range.” Thus, as the precision of the estimate increases, the confidence interval decreases in length. In this report answers to questions 3 and 4 of the Introduction are expressed as 0.95 confidence intervals.

**PROBLEM SOLUTION**

The broad plan for solution will consist of four main steps:

1. All \( \mu_{0} \) of the \( \mu_{i}^{0} \) are computed. Each of these computations uses the \( J_i \) observations associated with the particular sample; \( \sigma_{m0} \), the historic estimate of proportional mechanical error standard deviation with \( f_0 \) associated degrees of freedom; and a least-square fit to the \( J_i \) weighted points to arrive at the estimates of

\[
\mu_{i}^{0} = \sum_{k=1}^{L} \mu_{k,i}^{0}
\]

where it is assumed that

\[
\mu_{k,i}^{0} = \lim_{T \to 0} \mu_{k,i}^{0}(0,T) \quad \text{and} \quad \mu_{i}^{0} = \lim_{T \to 0} \mu_{i}^{0}(0,T)
\]

Estimates of \( \sigma_{m} \) based upon the sum of historic and current degrees of freedom are used in the computation for weighting. To obtain \( \sigma_{m} \) all \( I \) samples are used. Thus, all \( I \) least-square fits are performed in parallel using the results of the \( k^{th} \)
iteration to obtain all the $\tilde{\mu}_{ik}^o$ ($I$ times $L$ of these) and $\tilde{\sigma}_{mk}$ to be used in the $(k + 1)^{th}$ iteration. The magnitude of $\sigma_s$, the proportional sampling error standard deviation, in no way influences these computations because each fit is for a fixed single sample.

2. The $I$ final $\tilde{\mu}_{1}^o$'s, $\tilde{\sigma}_o$, the historic estimate of $\sigma_s$ with $d$ associated degrees of freedom; and $S^o_{\mu_{1}}$, the estimated variance of $\mu_{1}^o$, are used to compute a new $\tilde{\sigma}_s$ by iteration.

3. The statistics from the foregoing final iterations are used for computing confidence intervals, and for partitioning total uncertainty in $\tilde{\mu}_o$ into its three basic components.

4. The final $\tilde{\sigma}_m$ and $\tilde{\sigma}_s$, based on both current and past data, together with their respective associated degrees of freedom, are retained as inputs to future data analysis.

**Estimation of $\mu_{1}^o$ and $\sigma_m$**

The linear mathematical model is

$$\mu_{j}(t_{ij}, T_{ij}) = \sum_{q=1}^{L} \mu_{qj}^o X_{qij}$$

where the transformed independent variables are

$$X_{qij} = e^{-\lambda_{q} t_{ij}} \left( 1 - e^{-\lambda_{q} T_{ij}} \right)$$

with the corresponding linear statistical model

$$\eta_{ij} = \sum_{q=1}^{L} \mu_{qj}^o X_{qij} + e_{ij}$$

where the transformed dependent variables are

$$\eta_{ij} = \frac{C_{ij}}{T_{ij} \left( 1 - \tau \frac{C_{ij}}{T_{ij}} \right)} - \frac{C_{ij}^o}{T_{ij}}$$

and $e_{ij}$ is the $j^{th}$ deviation for the $i^{th}$ sample.
The strength of each isotope for the \(i^{th}\) sample is estimated by the weighted least-squares method, where each weight is inversely proportional to the variance of the datum. Since the variances are unknown, they must be estimated from past and current data. This leads to a nonlinear problem which is solved iteratively by starting with some initial estimates for the weights.

Using estimated weights, for each \(i\) at each iteration, minimize

\[
\sum_{j=1}^{J_i} w_{ijk} \left[ \eta_{ij} - \tilde{\mu}_{ik} (t_{ij}, T_{ij}) \right]^2 = \sum_{j=1}^{J_i} w_{ijk} \left( \eta_{ij} - \sum_{k=1}^{L} \tilde{\mu}_{ik}^0 X_{g_{ij}} \right)^2
\]

with respect to \(\tilde{\mu}_{ik}^0\) to obtain estimates \(\tilde{\mu}_{ik}^0\) by solving the resulting \(L\) simultaneous linear equations.

The estimation of weights at each iteration requires the partitioning of the estimated variance into its components. As a consequence, a new estimate of the sampling error \(\sigma_s\) is obtained.

An estimate of the total variance due to both mechanical and Poisson sources for \(j^{th}\) observation of the \(i^{th}\) sample is the usual

\[
\frac{S_{ik}^2}{w_{ijk}} = \frac{1}{J_i - L} \sum_{j=1}^{J_i} w_{ijk} \left( \eta_{ij} - \sum_{k=1}^{L} \tilde{\mu}_{ik}^0 X_{g_{ij}} \right)^2
\]

There are two causes for each deviate

\[
\eta_{ij} - \tilde{\mu}_{ik} (t_{ij}, T_{ij})
\]

namely, the error variance stemming from Poisson phenomena estimated to be

\[
\frac{N_{ij}^* + \tilde{\mu}_{ik} (t_{ij}, T_{ij})}{T_{ij}} + \frac{N_{ij}^*}{T_{ij}}
\]

plus the error variance stemming from mechanical causes estimated to be

\[
\left[ \tilde{\sigma}_{mk} \tilde{\mu}_{ik} (t_{ij}, T_{ij}) \right]^2
\]

A point by point partitioning of each deviate into amounts ascribable to mechanical and to Poisson sources is performed to obtain a point estimate of the current sampling error \(\sigma_m^*\). These point estimates are averaged over
all current observations to get $\tilde{\sigma}_{m'k}^2$, which, together with the historic estimate $\tilde{\sigma}_{ma}^2$, are averaged to provide an updated historic estimate for the next iteration. The foregoing procedures, as well as those for estimating the weights and source strengths, are summarized in the following simultaneous equations:

For $k = 0, 1, 2, \ldots$, last

$$w_{ijk+1} = \frac{K_{ij}}{N_{ij} + \frac{N_i^*}{T_{ij}} + (\tilde{\sigma}_{ma} \eta_{ij})^2}$$

$$= \frac{K_{ik+1}}{N_{ij} + \frac{\tilde{\mu}_{.k} (t_{ij}, T_{ij})}{T_{ij}} + \frac{N_i^*}{T_{ij}} + \left(\frac{\tilde{\sigma}_{mk} \tilde{\mu}_{.k} (t_{ij}, T_{ij})}{T_{ij}}\right)^2}$$

for $k > 0$

where $K_{ik}$ is consistent with

$$\sum_{i=1}^{J_i} w_{ijk+1} = 1$$

In matrix notation, for $h = 1, 2, \ldots, L$,

$$(\tilde{\mu}_{ik+1})_{L \times 1} = \left(\sum_{i=1}^{J_i} w_{ijk+1} X_{hi} \chi_{ij}\right)^{-1} \left(\sum_{i=1}^{J_i} w_{ijk+1} X_{hi} \eta_{ij}\right)_{L \times 1}$$

$$= \frac{1}{(J_i - L)} \left\{ \frac{J_i}{J_i - L} \sum_{i=1}^{J_i} w_{ijk+1} \left[ \tilde{\eta}_{ij} - \tilde{\mu}_{ik+1} (t_{ij}, T_{ij}) \right]^2 \right\}$$

where

$$\tilde{\sigma}_{m'k+1}^2 = \sum_{i=1}^{1} (J_i - L) \left\{ \frac{J_i}{J_i - L} \sum_{i=1}^{J_i} w_{ijk+1} \left[ \tilde{\eta}_{ij} - \tilde{\mu}_{ik+1} (t_{ij}, T_{ij}) \right]^2 \right\}$$

$$\Omega = \frac{\left[ \tilde{\sigma}_{mk} \tilde{\mu}_{ik+1} (t_{ij}, T_{ij}) \right]^2}{\left[ \tilde{\sigma}_{mk} \tilde{\mu}_{ik+1} (t_{ij}, T_{ij}) \right]^2 + \frac{\tilde{\mu}_{ik+1} (t_{ij}, T_{ij}) + N_i^*}{T_{ij}} + \frac{N_i^*}{T_{ij} U_{ij}}}$$

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\[ \bar{\sigma}_{mk+1}^2 = \frac{f_0 \bar{\sigma}_{m0}^2 + f_1 \bar{\sigma}_{m'k+1}^2}{f_0 + f_1} \]  

(4)

where \( \bar{\sigma}_{m0} \) is the historic estimate of the proportional mechanical error based on \( f_0 \) degrees of freedom, and

\[ f_1 = \sum_{i=1}^{1} (J_i - L) \]

is the additional degrees of freedom available from the current data.

The introduction of the integers \( U_{ij} \) associated with background reading requires explanation. A single background reading may at times be used to correct more than one observation of the same sample or even of different samples. The product of the number of samples for which a background reading has been used to compute \( \eta_{ij} \)'s times the number of observations of the \( i^{th} \) sample for which this particular background reading has been used is designated \( U_{ij} \). It is introduced into the iteration merely to assure that the variance due to a single background reading gets counted only once in the averaging process over all observations for all samples.

Each deviate was divided by \( \mu_{ik}(t_{ij}, T_{ij}) \) to assure that \( \bar{\sigma}_m \) will be on a proportional mechanical error basis, as defined. The last multiplier in Equation 3 partitions each squared deviate into the fraction blamed on mechanical error. The sum of the \( J_i - L \) (degrees of freedom associated with \( i^{th} \) sample) appearing in the denominator and the multipliers \( J_i - L \) in the numerator average \( \bar{\sigma}_m^2 \) over all the total

\[ \sum_{i=1}^{1} (J_i - L) \]

degrees of freedom available.

Iteration is terminated when two successive \( \bar{\sigma}_m \) differ by less than an arbitrarily small amount, \( \delta \), say 0.00005 (0.005%); that is,

\[ |\bar{\sigma}_{m'k+1} - \bar{\sigma}_{m'k}| < \delta \]

At the end of the least-square fitting process, in addition to the final \( \bar{\sigma}_m \), there are available for each sample the final \( L + 1 \) estimates contained in

\[ \bar{\mu}_i^q = \sum_{k=1}^{L} \bar{\mu}_{kj}^q \]
These and final inverse matrices with associated $S_1^2$ from the least-square fitting process are used in establishing confidence intervals and in estimating $\sigma^2$.

**Estimation of $\mu^o_i$ and $\sigma^2$**

The $l$ samples have been used to obtain the final estimates $\mu^o_1$, $\mu^o_2$, ..., $\mu^o_l$, together with the $l$-associated final $L$ by $L$ inverse matrices and $S_j^2$ used for the least-squares fits.

In using these it will be assumed erroneously that the estimates $\tilde{\mu}^o_1$, $\tilde{\mu}^o_2$, ..., $\tilde{\mu}^o_l$ are independent. The slight dependency that may exist between two particular $\tilde{\mu}^o_i$'s stems solely from the use of some of the same background readings for the computation of each of the two $\tilde{\mu}^o_i$'s. This dependency is negligible in general because almost invariably $N_{ij} < N_{lj}$. To consider the dependency would unduly complicate the computation.

From the last iteration of the preceding section

$$S_{\mu^o_i}^2 = A_i S_i^2$$

where $A_i$ is the sum of all the $L$ elements of the last inverse matrix for sample $i$.

To obtain $\mu^o_i$ and $\tilde{\sigma}_i^2$, weights $v_1$, $v_2$, ..., $v_l$ will be iterated with the $l$-fixed $S_{\mu^o_i}^2$ and the $l$-fixed $\mu^o_i$ as inputs to obtain a stable estimate of $\tilde{\sigma}_i^2$. The iteration algorithm to accomplish this consists of the successive use of the following simultaneous equations. For $k = 1, 2, 3, \ldots, \text{last}$

$$v_{ik} = \frac{k_1}{(\tilde{\mu}_i - \tilde{\sigma}^2_{i0})^2 + S_{\mu^o_i}^2} \quad \text{for } k = 1$$

$$= \frac{k_k}{(\tilde{\mu}_i - \tilde{\sigma}^2_{ik-1})^2 + S_{\mu^o_i}^2} \quad \text{for } k > 1$$

where $k_k$ is consistent with

$$\sum_{k=1}^{l} v_{ik} = 1$$
\[ \tilde{\mu}_{.,k}^0 = \sum_{i=1}^{1} v_{ik} \tilde{\mu}_{i}^0 \]  

(6)

\[ \tilde{\sigma}_{k}^2 = \frac{1}{1 - 1} \sum_{i=1}^{1} v_{ik} \left( \frac{\tilde{\mu}_{i}^0 - \tilde{\mu}_{.,k}^0}{\tilde{\mu}_{.,k}^0} \right)^2 \left( \frac{(\tilde{\mu}_{.,k}^0 \tilde{\sigma}_{k-1}^2) + S_{so}^2}{\tilde{\mu}_{.,k}^0} \right) \]  

(7)

\[ \tilde{\sigma}_{k}^2 = \frac{(1 - 1) \tilde{\sigma}_{k-1}^2 + d \tilde{\sigma}_{so}^2}{d + 1 - 1} \]  

(8)

where \( \tilde{\sigma}_{so} \) is the historic estimate of proportional sampling error based on \( d \) degrees of freedom. The historic estimate \( \sigma_{so} \) stems from previous analyses of all pertinent counting data from a laboratory before the currently being analyzed.

**Estimation of \( \mu_{k}^0 \).**

Inputs to the estimation of \( \mu_{k}^0 \):

1. \( v_1, v_2, ..., v_l \) from the last iteration of the preceding section.

2. The \( \tilde{\mu}_{k}^0 \) (L of these) from the final iteration of the section entitled "Estimation of \( \mu_{k}^0 \) and \( \sigma_{m}^0 \)."

Each of the \( \tilde{\mu}_{k}^0 \) will be computed as a simple partitioning of the \( \tilde{\mu}_{.,k}^0 \) so that

\[ \tilde{\mu}_{.,k}^0 = \sum_{k=1}^{L} \tilde{\mu}_{k}^0 . \]

as follows:

\[ \tilde{\mu}_{k}^0 = \sum_{i=1}^{1} v_i \tilde{\mu}_{k,i}^0 \]

This partitioning is based not on statistical grounds, but on the physical fact that all samples of equilibrium solutions of mixtures contain the same ratio of ingredients, to a precision on the order of the reciprocal of the square root of the number of molecules involved in the ingredient sample.
Confidence Intervals for $\mu_0$ and $\mu_2^0$.

Let $t^*$ be a value computed such that

$$ \int_{-t^*}^{t^*} f(t) \, dt = 0.95 $$

where $f(t)$ is the Student's $t$ density function for $t$ with $l - 1$ degrees of freedom. Then the 0.95 confidence interval for $\mu_0$ extends over the interval

$$ \mu_0^0 \pm t^* \sqrt{\frac{\sum_{i=1}^{l} v_i (\mu_1^0 - \mu_0^0)^2}{l - 1}} $$

and for $\mu_2^0$ over the interval

$$ \mu_2^0 \pm t^* \sqrt{\frac{\sum_{i=1}^{l} v_i (\mu_1^0 - \mu_2^0)^2}{l - 1}} $$

Partitioning $\tilde{\sigma}_{\mu_0}^2$

The estimated variance in estimating $\mu_0$ is

$$ \tilde{\sigma}_{\mu_0}^2 = \frac{1}{l - 1} \sum_{i=1}^{l} v_i (\mu_i^0 - \mu_0^0) $$

which has three components that are assumed independent. These components are errors attributed to sampling, mechanical difficulties, and Poisson phenomena, and may be expressed as

$$ \tilde{\sigma}_{\mu_0}^2 = \left( \tilde{\sigma}_{\mu_0}^2 \right)_s + \left( \tilde{\sigma}_{\mu_0}^2 \right)_m + \left( \tilde{\sigma}_{\mu_0}^2 \right)_p $$

The computation of these three components will now be discussed. A crude estimate of
where $A_i$ is the sum of all the $L^2$ elements in the last inverse matrix for sample $i$ and

$$S_i^2 = \frac{1}{J_i - 1} \sum_{i=1}^{J_i} w_{ij} \left[ \eta_{ij} - \bar{\mu}_i(t_{ij}, T_{ij}) \right]^2$$

To partition $S_i^2$ into its Poisson and mechanical components and simultaneously assure that the sum of the parts equals the whole, it is necessary to partition the individual squared deviates. The point estimate for the $j^{th}$ observation on the $i^{th}$ sample for the variation due to the Poisson phenomenon is

$$a_{ij} = \frac{\bar{\mu}_i(t_{ij}, T_{ij}) + N_{ij}^*}{T_{ij}} + \frac{N_{ij}^*}{U_{ij} T_{ij}}$$

and for the variation due to mechanical causes is

$$\beta_{ij} = \left[ \bar{\sigma}_m \bar{\mu}_i(t_{ij}, T_{ij}) \right]^2$$

then

$$\left( \tilde{\sigma}_{\mu_i}^2 \right)_p = \frac{A_i}{J_i - L} \sum_{i=1}^{J_i} w_{ij} \left[ \eta_{ij} - \bar{\mu}_i(t_{ij}, T_{ij}) \right]^2 \frac{a_{ij}}{a_{ij} + \beta_{ij}}$$

$$\left( \tilde{\sigma}_{\mu_i}^2 \right)_m = \frac{A_i}{J_i - L} \sum_{i=1}^{J_i} w_{ij} \left[ \eta_{ij} - \bar{\mu}_i(t_{ij}, T_{ij}) \right]^2 \frac{\beta_{ij}}{a_{ij} + \beta_{ij}}$$

which guarantees that

$$S_{\mu_i}^2 = \left( \tilde{\sigma}_{\mu_i}^2 \right)_m + \left( \tilde{\sigma}_{\mu_i}^2 \right)_p$$
Hence, define:

\[ a_{..} = \sum_{i=1}^{1} v_i^2 \left( \frac{\tilde{\sigma}_{..}^2}{\tilde{\mu}_{..}} \right) \]

\[ \beta_{..} = \sum_{i=1}^{1} v_i^2 \left( \frac{\tilde{\sigma}_{..}^2}{\tilde{\mu}_{..}} \right) \]

where \( a_{..} \) and \( \beta_{..} \) are crude estimates of \( \left( \frac{\tilde{\sigma}_{..}^2}{\tilde{\mu}_{..}} \right)_p \) and \( \left( \frac{\tilde{\sigma}_{..}^2}{\tilde{\mu}_{..}} \right)_m \), respectively.

There is no guarantee that \( a_{..} + \beta_{..} \) may not exceed \( \tilde{\sigma}_{..}^2 \), because through the vagaries of chance the \( I \) values of \( \tilde{\mu}_{..} \) may be unusually close together.

Let

\[ \gamma_{..} = (\tilde{\mu}_{..} \tilde{\sigma}_{..})^2 \sum_{i=1}^{1} v_i^2 \]

Finally

\[ \left( \frac{\tilde{\sigma}_{..}^2}{\tilde{\mu}_{..}} \right)_p = \frac{a_{..} + \beta_{..} + \gamma_{..}}{\tilde{\sigma}_{..}^2} \tilde{\sigma}_{..}^2 \] (9)

\[ \left( \frac{\tilde{\sigma}_{..}^2}{\tilde{\mu}_{..}} \right)_m = \frac{\beta_{..}}{a_{..} + \beta_{..} + \gamma_{..}} \tilde{\sigma}_{..}^2 \] (10)

\[ \left( \frac{\tilde{\sigma}_{..}^2}{\tilde{\mu}_{..}} \right)_s = \frac{\gamma_{..}}{a_{..} + \beta_{..} + \gamma_{..}} \tilde{\sigma}_{..}^2 \] (11)

Summary

The Problem Solution section may be summarized briefly in the following manner:

1. Transform the raw data as specified in the section entitled, "Estimation of \( \mu^0_k \) and \( \sigma_m \)."
2. For each i compute a least-square fit of the form

\[ \tilde{\mu}_i (t_{ij}, T_{ij}) = \tilde{\mu}_{i2} X_{2ij} + \ldots + \tilde{\mu}_{iL} X_{lij} \]

3. From these I results estimate \( \sigma_m^2 \) by using Equation 3.

4. Use Equation 4 to compute \( \tilde{\sigma}_{m2} \) and then an array of new weights \( w_{ij} \).

5. Continue this 2-3-4 iterative sequence until

\[ \left| \tilde{\sigma}_{m^k} - \tilde{\sigma}_{m^{k-1}} \right| < 5 \times 10^{-5} \]

6. From the last iteration obtain \( \tilde{\sigma}_m \), \( \tilde{\sigma}_m \), \( f_0 + f_1 \), all \( \tilde{\mu}_o \), all \( \tilde{\mu}_s \), and all \( S_{oo} \).

7. Using the iterative scheme given by Equations 5 through 8, obtain \( \tilde{\mu}_o \), \( \tilde{\sigma}_s \), and \( d + 1 - 1 \) on convergence; that is, such that

\[ \left| \tilde{\sigma}_{s^k} - \tilde{\sigma}_{s^{k-1}} \right| < 5 \times 10^{-5} \]

8. Using the weights, \( w_i \)'s, of the last iteration in step 7, compute all \( \tilde{\mu}_o \) with 0.95 confidence limits on \( \tilde{\mu}_o \) and each \( \tilde{\mu}_o \).

9. Use Equations 9, 10, 11 to partition the estimated variance of \( \tilde{\mu}_o \) into the components attributed to Poisson, mechanical, and sampling causes.

**DISCUSSION OF MATHEMATICAL MODEL**

The procedure outlined in this report estimates, simultaneously, total random-and-systematic error, its Poisson component, its sampling component, and its residual component, called mechanical error (a presumed composite from all causes other than sampling variation and Poisson phenomenon). This analytical procedure is expressed in terms of the error components at time \( t_0 \), that is, age zero. The error under discussion is error in estimating corrected system counts per minute for an exact intended aliquot.
In principle, this unified simultaneous approach should be superior to separate estimations of the three foregoing components of error. In passing, it should be noted clearly that this report is concerned with an analysis, not a calibration; that is, laboratory biases will never be brought to light by this procedure alone. The analysis may, however, be used for the calibration of a laboratory when laboratory data from a known standard solution mixture are available.

Because the procedure developed in this report is an initial effort, it is not the last word. An effort was made to obtain either unbiased estimates, maximum likelihood estimates, or both, so that the results could be claimed to be "best" in some sense. Unfortunately, such an effort did not seem feasible at all points of the development. The slight statistical imperfections that have crept into the work will now be discussed.

The mathematical model assumes that the standard deviation in counts per minute for the random error from all causes other than sampling and Poisson phenomenon is proportional strictly to expected corrected counts per minute regardless of age, isotope solution component strengths, or length of observing period. Perhaps this is not true in practice. All other unaccounted-for errors are encompassed by $\sigma_m$, so that $(\mu^0 \sigma_m)^2$ plays the role of a residual. Nevertheless, the unitless proportionality constant $\sigma_m$ is very meaningful for the physicist. The statistician will note that $\sigma_m$ is the coefficient of variation for "other causes," assumed, perhaps erroneously, to be constant.

On the other hand, the model $\mu^0 \sigma_s$ for standard deviation of sampling random error should be almost true in fact.

In computing the variance of a datum, expressed as counts-per-minute squared, heavy use has been made of the fact that for a Poisson density of counts, the mean and variance are equal. Statisticians generally accept this "fact," but the physicist will note immediately that the units are wrong; counts cannot equal the square of the number of counts. To make physical sense, the statisticians' equation needs a unit, $u^*$, with dimension reciprocal-counts so that

$$\mu = u^* \sigma^2$$

which translated says, "for the population of a fixed Poisson process, the positive square root of the numerical value of the mean expressed as a pure number is the same as the standard deviation of the population expressed as a pure number." The unit $u^*$ has been deliberately left out of the Problem Solution section. To check on unit consistency of the equations, $u^*$ will need to be introduced.
The Student's \( t \) density has been freely used in computing confidence intervals, even though it is realized that the uncertainty in estimating the various \( \mu \)'s is not strictly represented by a normal distribution, since some of this uncertainty stems from nonnormal Poisson processes. It is believed that use of the Student's \( t \) does not result in very serious errors. This belief is based upon the rapid convergence usually encountered in applying the central limit theorem and upon the fact that the \( \mu \)'s, \( \mu_i \)'s, and particularly the \( \sum_{i=1}^{J} \mu_i \) are sufficiently large to nearly normalize the uncertainty.

Criticism could be directed at the following type of sequential steps which were employed freely:

1. \( a_{a+b}^2 = a_a^2 + a_b^2 \)

2. \( \tilde{a}_{a+b}^2 = \tilde{a}_a^2 + \tilde{a}_b^2 \)

In the first place, 1 is not true unless the random variables \( a \) and \( b \) are independent of each other. Where 1 was used the two random variables involved were only very nearly independent. The step 1 to 2 is not valid in general unless the quantities in 2 are unbiased.

Additional criticism might be directed at partitioning for \( \tilde{a}_m \) by ratios rather than differences; partitioning \( \tilde{a}_s \) is, of course, open to the same criticism. This method of partitioning was employed to avoid negative \( \tilde{a}_m^2 \) and negative \( \tilde{a}_s^2 \), since such results are both impossible and meaningless in practice and cause insurmountable difficulties in using weights based on estimated variances. The extent of possible bias introduced by this method of partitioning is unknown. An adequate treatment of estimated weights or of partitioning variances into their various components by the ratio method is not given in the literature, and investigation of these subjects would constitute a larger study.

In spite of the foregoing reservations it is believed that this combined unified approach will prove to be a significant improvement over piecemeal analyses, and a vast improvement over assuming that all variation is Poisson. Use of the combined approach will certainly enable a laboratory to keep records of its \( \tilde{a}_s \) and \( \tilde{a}_m \) estimates with the hope that these estimates will drift lower with the age of the laboratory as techniques improve. Certainly these estimates will point to the area where the most improvement is to be made. Should \( a_s \) and \( a_m \) regress significantly with time, their regression estimates would be the ones to use in the weighting process.
It is noted that under many sets of conditions even the best estimate has 
an appreciable chance of missing considerably. For the problem of this report, 
the following will tend to improve the estimate of expected decay rate at some 
historic time of interest:

1. Decrease of sampling error.
2. Sampling replication.
3. Decrease of other errors.
4. Replicating and lengthening periods of observing samples.

Most notable is item 2. Presuming reasonable efforts in areas 1 and 3 have 
been made, but an appreciable sampling error persists, the greatest gain per 
unit of effort likely can be accomplished by increasing the number of samples 
of the isotope solution. Simply replicating the number of observations of a 
given sample will not average out sampling fluctuation. When records of \( \tilde{\alpha}_m \) 
and \( \tilde{\alpha}_q \) are in hand it will be possible to use them to design a schedule for 
minimum-variance estimate within a fixed total cost.

**SIMULATION**

A FORTRAN program was written to implement the procedure given in 
the Problem Solution section. It is presented and discussed in the Appendix. 
How well does this procedure work? In order to obtain a preliminary answer 
to this, it is desirable to operate the program using input data from a decay 
phenomenon with known parameters. To accomplish this it was necessary to 
develop a data generator.

**Data Generator**

The known constants and parameters required for generating data are 
\( \lambda_R, L, \mu^0_R, \alpha_s, \sigma_m, I, J_i, T_{ij}, T^*_i \), and \( B_{ij} \) (expected background counts per 
minute).

The objective is to simulate \( I \) samples and create \( J_i \) observations (\( i = 1, 
2, ..., I \)) at different ages of the samples, assuming a constant parameter for 
the Poisson-distributed background, and also to create an independent set of 
background observations. This is accomplished in the following steps.

**Step 1:** I random numbers are drawn from a normal population with 
mean zero and standard deviation \( \sigma_s \). Name these \( r_1, r_2, ..., r_I \). For each \( i \) 
compute:
Step 2: For each of the $I$ samples draw $J_i$ random numbers from a normal population with mean zero and standard deviation $\sigma_m$. Name these $q_{i1}, q_{i2}, \ldots, q_{ij}$. Compute \( \sum_{i=1}^{I} J_i \) quantities

\[
(1 + q_{ij}) \left[ \mu_{i1}^{o} \left( \frac{1 - e^{-\lambda_1 T_{ij}}}{\lambda_1 T_{ij}} \right) e^{-\lambda_1 t_{ij}} + \ldots + \mu_{iL_i}^{o} \left( \frac{1 - e^{-\lambda_L T_{ij}}}{\lambda_L T_{ij}} \right) e^{-\lambda_L t_{ij}} \right]
\]

Step 3: With each of the $T_{ij}$ ($\xi_{ij} + B_{ij}$) as the Poisson parameter, select at random one count and name it $C_{ij}$. Select at random one count from each Poisson density with parameter $B_{ij} T_{ij}$ and name it $C^*_i$.

Fifteen sets of data were generated. For each set the input constants and parameters were

- $\lambda_1 = 0.30/\text{min}$
- $\lambda_2 = 0.15/\text{min}$
- $\lambda_3 = 0.05/\text{min}$
- $\mu_1^{o} = 8,000 \, \text{cpm}$
- $\mu_2^{o} = 1,500 \, \text{cpm}$
- $\mu_3^{o} = 500 \, \text{cpm}$
- $\sigma_s = 0.05$
- $\sigma_m = 0.08$
- $I = 5$
- $J_i = 10$
- $t_{ij} = (0, 2, 4, 6, 8, 15, 25, 35, 45, 55) \, \text{minutes every } i$
- $T_{ij} = (1, 1, 1, 1, 5, 5, 5, 5, 5) \, \text{minutes every } i$
- $T_{ij}^* = 10 \, \text{minutes every } i \text{ and every } j$
\[ B_{ij} = 10 \text{ cpm every } i \text{ and every } j \]
\[ U_{ij} = 1 \text{ every } i \text{ and every } j \]

The output \( C_{ij} \) and \( C^*_{ij} \) were analyzed with the computer program SAND (Appendix), as explained in the next subsection.

**Results**

The 15 sets of \( C_{ij} \) and \( C^*_{ij} \) together with the information

\[ (\lambda_1, \lambda_2, \lambda_3) = (0.30, 0.15, 0.05)/\text{min} \]
\[ t_{ij} = (0, 2, 4, 6, 8, 15, 25, 35, 45, 55) \text{ minutes} \]
\[ T_{ij} = (1, 1, 1, 1, 5, 5, 5, 5, 5) \text{ minutes} \]
\[ T^*_{ij} = 10 \text{ minutes} \]
\[ \tau = 0 \text{ minute} \]

were inputted to the computer program in succession, thereby accumulating historic \( \sigma_m \) and \( \sigma_s \) with associated degrees of freedom after each set for use in the next set. Set 1 was rerun after set 15 and then set 2 was rerun. These runs are designated 1' and 2' in Tables 1 through 6, and the slight differences from 1 and 2 noted stem from the change in historic \( \sigma_m \) and \( \sigma_s \). The historic estimates of percentage errors are graphed in Figure 1.

![Figure 1. Estimated sampling and mechanical errors versus computer run number.](image-url)
The total uncertainty in \( \mu_0 \), expressed as a variance appears in the last column of Table 1. The other columns contain the analysis of the total uncertainty into sampling, mechanical, and Poisson sources.

The 17 estimates of \( \mu_0^0, \mu_1^0, \mu_2^0, \) and \( \mu_3^0 \), with 0.95 confidence intervals are contained in Table 2. It will be recalled that \( \mu_0^0 = 10,000, \mu_1^0 = 8,000, \mu_2^0 = 1,500, \) and \( \mu_3^0 = 500 \) cpm.

The running estimates (current and historical) of \( \sigma_m \) and \( \sigma_s \) are contained in Table 3. The degrees of freedom for 1' and 2' are admittedly wrong since the old data of 1 and 2 were used, giving really no new degrees of freedom.

In detail Table 4 shows the estimates of \( \mu_0^0, \mu_1^0, \mu_2^0, \) and \( \mu_3^0 \) from each of the 75 samples. The reader will note the entries are wilder than those of Table 2, which showed the more stable weighted averages of five samples.

Uncertainty in estimating \( \mu^0 \) from a single sample expressed as total variance is analyzed into mechanical and Poisson sources in Table 5 for each of the 75 samples.

For each of the 75 samples Table 6 displays \( S_i^2 \) and the product \( S_i^2 \) times the elements of the symmetric inverse matrix under the headings of variances and covariances.

### Table 1. Analysis of Variance of Mean Count Rate at Time t = 0 (All Isotopes)

<table>
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<tr>
<th>Run</th>
<th>Sampling</th>
<th>Mechanical</th>
<th>Poisson</th>
<th>Total</th>
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<td>14726</td>
<td>275432</td>
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<tr>
<td>2'</td>
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<td>204693</td>
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Table 2. Mean Decay Rate at Time $t = 0$
and 0.95 Confidence Limits

<table>
<thead>
<tr>
<th>Run</th>
<th>All Isotopes (cpm)</th>
<th>First Isotope (cpm)</th>
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<tr>
<td></td>
<td>Lower Limit</td>
<td>Mean $\mu_0$</td>
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<td>9277.1</td>
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D.F. = Degrees of freedom associated with updated historic estimate.

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(All Isotopes – Fixed Sample)

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Table 6. Variances and Covariances of Fitted Parameters and Weighted Variance of Errors

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Discussion of Results

Fifteen runs on different simulated samples of data with parameters
$$\mu_\circ = \mu_1^\circ + \mu_2^\circ + \mu_3^\circ$$

have been analyzed by means of the FORTRAN program. For each run 0.95 confidence intervals for \(\mu_1^\circ\), \(\mu_2^\circ\), and \(\mu_3^\circ\) were printed as outputs (see Table 2). If the procedure were perfect, in the long run 5% of the intervals would fail to bracket (contain) the parameter represented. Five percent of fifteen is 0.75. Thus a priori the expected number of misses per parameter is 0.75 and for all four parameters three misses. Actually, two misses were experienced, \(\mu_2^\circ\) at run 1 and \(\mu_2^\circ\) at run 9. If the procedure is imperfect with reference to confidence intervals, a large number of additional runs would be needed to “prove” it statistically.

The data generator introduced mechanical error with parameter
$$\sigma_m = 0.08$$

From the 15 runs with their available 525 degrees of freedom for estimating \(\sigma_m\), the final estimate was:
$$\tilde{\sigma}_m = 0.0771$$

Is the procedure biased downward? The difference -0.0029 is not statistically significant at the 0.95 level. Thus more runs would be needed to prove that the procedure is imperfect for estimating \(\sigma_m\). Even if the procedure is imperfect, the bias appears to be small. The authors anticipated a small upward bias rather than the downward bias actually observed, because the variance of the sum of the mechanical and Poisson components should be slightly more than the sum of the two components, due to a small positive covariance. This covariance stems from the slight tendency for large Poisson errors to be associated with large mechanical error since \(\sigma_m\) is on a percentage basis. The procedure ignores this tendency by partitioning as though the two sources were independent. The procedure should have a tendency to lead to a divisor too small in the proportioning for \(\tilde{\sigma}_m\) and thus give a \(\tilde{\sigma}_m\) too large.

The data generator introduced sampling error with parameter
$$\sigma_s = 0.05$$
From the 15 runs with their available 60 degrees of freedom for estimating \( \sigma_s \), the final estimate \( \tilde{\sigma}_s = 0.0654 \) misses the parametric value by a disquieting amount. While the physicist may not be concerned with a bias of 1-1/2%, this is statistically significant at the 0.99 level, from which it is concluded that the procedure estimates \( \sigma_s \) with an upward bias. Why? Some of it can stem from the fact that the procedure treats sampling, Poisson, and mechanical error sources as independent, whereas because of the percentage concept there are actually three small positive covariances between these three sources. In the proportioning for \( \tilde{\sigma}_s \), these were not included in the divisor – which should result in a tendency to bias \( \tilde{\sigma}_s \) upward.

It is noted that \( \sigma_s \) is estimated at the last step of the computation. It is therefore suspected that the accumulated effects of all the slight imperfections of the procedure finally appear in \( \tilde{\sigma}_s \). Two of the approximations suspected of contributing a large part of the imperfections are (1) the use of estimated variances instead of true variances in the weighting and (2) the inefficient estimation of Poisson parameters by the partitioning of each regression error. (A discussion of the efficiency of estimating Poisson parameters can be found in another report.*

Imprecise as the procedure may be it is believed to be a much better approach than the customary assumption that all error is Poisson distributed. It is hoped that physicists will use this procedure and that statisticians will attempt to improve it.

FINDINGS

A procedure implemented as the SAND computer program is now available for use by nuclear physicists. Given a multiple isotope source and a series of observations on some samples of the source, this FORTRAN computer program will estimate the strength of each isotope; place confidence limits thereon; break the uncertainty into Poisson error, sampling error, and residual (called mechanical) error; supply an estimate of the standard deviation of sampling error; and supply an estimate of the standard deviation of mechanical error.

CONCLUSIONS

The procedure developed in this report constitutes a considerable improvement over the customary technique, whereby overoptimistic confidence intervals have been based on only Poisson variation. Even though the procedure contains statistical imperfections that would require considerably more research to remove, it does a reasonably good job of breaking total variation into its three sources.

RECOMMENDATION

It is suggested that nuclear physicists use the procedure described in this report for the statistical analysis of counting data.
Appendix

SAND, A FORTRAN II-D COMPUTER PROGRAM FOR THE
STATISTICAL ANALYSIS OF NUCLEAR DATA

The procedure described within the body of this report is implemented
by the computer program SAND on an IBM 1620 with an IBM 1311 Disk Drive.
A minimum 1620 (20K memory) with no special features but a disk is
required. The programming language is the disk version of FORTRAN II;
however, the disk requirement may be replaced by magnetic tape by changing
the FETCH and RECORD statements in the following subroutines: FANDW,
INPUT2, and BCKGND.

All input to the program is from the card reader, and all output is to the
card punch for off-line listing on an IBM 407. The card I/O statements may
be replaced by tape I/O through a simple sifting program.

The program for the IBM 1620 is limited to a maximum of 25 samples
per run.

The format of the input for each run, the card deck setup (Figure 2), a
sample input with corresponding output (Figure 3), and the listing of the
FORTRAN II-D source program (Figure 4) follow.

FORMAT OF INPUT FOR EACH RUN

Card 1. Title card; FORMAT (80H

Card 2. I, L, τ, τ₀, oₘ₀, oₜ₀, d, oₘᵣ; FORMAT (215, 7E10.0)

where I = numbers of samples (I < 25)
L = number of isotopes (L < 5)
τ = detector deadtime in min/count
τ₀ = historic time at t = 0 (minutes)
oₘ₀ = historic estimate of mechanical error (percent)
oₜ₀ = degrees of freedom associated with oₘ₀. (If f₀ = 0, then
this is a starting estimate used in solving the nonlinear
equation in oₘᵣ.)
\[ \tilde{\sigma}_{so} = \text{historic estimate of sampling error (percent)} \]

\[ d = \text{degrees of freedom associated with } \tilde{\sigma}_{so}. \quad (\text{If } d = 0, \text{ then this is a starting estimate used in solving the nonlinear equation in } \tilde{\sigma}_{so}) \]

\[ \tilde{\sigma}_{mr} = \text{partially updated } \tilde{\sigma}_{mo} \text{ used in restarting program after interruption (normally blank)} \]

Card 3. \( \lambda_1, \lambda_2, ..., \lambda_L \); FORMAT (5E15.8)

where \( \lambda_0 \) = decay constant for isotope \( \ell \) (\( \ell = 1, 2, ..., L \leq 5 \)) in reciprocal minutes

Card 4. \( J_1, J_2, ..., J_i \); FORMAT (1515)

where \( J_i \) = number of observations for sample \( i \) (\( i = 1, 2, ..., I \leq 25 \))

Observation Cards. \( T_{ij}, C_{ij}, T_{ij}^*, C_{ij}^*, t_{ij} \); FORMAT (5E15.8)

where the subscript \( j \) varies the most rapid; \( j = 1, 2, 3, ..., J_i \) for \( i = 1, 2, 3, ..., I \)

Figure 2. Deck setup for each run.
Figure 3. Sample input and output.

Sample Input

```
ZZJOB
ZXXFOSSAND
*LOCALSAND, INPUT1, INPUT2, BCKGND, LS0G, VIESLE, SANDS, MEC1, SAMPL1, SAMPL2,
*LOCALCMPTS, LIMITS, STUDNT, SPANDC, SANDS1, INPUTC
0 SIMULATED DATA FOR RUN NUMBER 6
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**Figure 3. Continued.**

ZZZZ END OF JOB

CONTINUED
Sample Output

1 STATISTICAL ANALYSIS OF NUCLFAR DATA (SAND)

SIMULATED DATA FOR RUN NUMBER 6
NUMBER OF SAMPLES 5
NUMBER OF ISOTOPES 3
HISTORIC TIME AT T=0 0.00 MIN
DETECTOR DEADTIME 0.00 MIN/COUNT
DECAY CONSTANTS *30 1/MIN
*15
*05

OBSERVATIONS PER SAMPLE
10
10
10
10
10
10

HISTORIC MECHANICAL ERROR 6.74 PERCENT
DEGREES OF FREEDOM 175
HISTORIC SAMPLING ERROR 6.60 PERCENT
DEGREES OF FREEDOM 20
TOLERANCE ON ERROR 0.05 PERCENT
MAXIMUM ITERATIONS ALLOWED 25

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Figure 3. Continued.

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0 MEAN DECAY RATE AT TIME T=0
(ALL ISOTOPES)
ANALYSIS OF VARIANCE

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0 DECAY RATE (COUNTS/MIN) AT TIME T=0

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0 UPDATED HISTORIC MECHANICAL ERROR 6.92 PERCENT
DEGREES OF FREEDOM 210

0 UPDATED HISTORIC SAMPLING ERROR 6.29 PERCENT
DEGREES OF FREEDOM 24

CONTINUED
Figure 3. Continued.

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MEAN DECAY RATE AT TIME T=0
(ALL ISOTOPES - FIXED SAMPLE)
ANALYSIS OF VARIANCE

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<td>153900</td>
<td>36837</td>
<td>190739</td>
</tr>
</tbody>
</table>

NOTE: The slight discrepancies between the values in this sample output and those given in Tables 1-5 are the result of using historic mechanical and sampling errors truncated to three digits as inputs.
Figure 4. Program listing.

<table>
<thead>
<tr>
<th>ZZJOR</th>
<th>SAND</th>
</tr>
</thead>
<tbody>
<tr>
<td>ZZDUP</td>
<td></td>
</tr>
<tr>
<td>*DELETE</td>
<td></td>
</tr>
<tr>
<td>ZZFOR</td>
<td></td>
</tr>
<tr>
<td>*DISKSAND</td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>ANALYSIS OF COUNTING DATA FROM SAMPLES OF RADIOACTIVE MATERIALS</td>
</tr>
<tr>
<td></td>
<td>W. L. WILCOXSON</td>
</tr>
<tr>
<td>C</td>
<td>*</td>
</tr>
</tbody>
</table>

GLOSSTARY OF SYMBOLS

- AMDA(L) = DECAY CONSTANT (1/MINUTES) FOR THE L'ITH ISOTOPE
- AVMECH = ABSOLUTE MECHANICAL VARIANCE
- AVMIJ = FRACTION OF SQUARED ERROR AT POINT IJ ATTRIBUTED TO MECHANICAL CAUSES
- AVMI(J) = ABSOLUTE VARIANCE DUE TO MECHANICAL CAUSES
- AVPM = ABSOLUTE VARIANCE DUE TO POISSON AND MECHANICAL
- AVPIJ = FRACTION OF SQUARED ERROR AT POINT IJ ATTRIBUTED TO POISSON PHENOMENA
- AVP(I) = ABSOLUTE VARIANCE DUE TO POISSON PROCESSES
- AVPOSN = ABSOLUTE VARIANCE DUE TO POISSON PROCESS
- AVSAMP = ABSOLUTE VARIANCE DUE TO SAMPLING
- A(L1+1L2) = ELEMENT OF LEAST SQUARES MATRIX
- C = COUNTS OBSERVED DURING PERIOD T
- CSTAR = BACKGROUND COUNTS OBSERVED DURING PERIOD TSTAR
- DELTA = MAXIMUM ABSOLUTE RELATIVE ERROR ALLOWED
- DF = DEGREES OF FREEDOM
- DFMECH = DEGREES OF FREEDOM FOR HISTORIC MECHANICAL ERROR
Figure 4. Continued.

C DESAMP = DEGREES OF FREEDOM FOR HISTORIC SAMPLING ERROR
C EMECH = MECHANICAL ERROR (PERCENT)
C EMU0T = SUM(EMU(I,L)*X(L),L=1,LMAX),
C SUBSCRIPTS I, J, AND K ARE IMPLIED.
C EMU0 = LAST SUM(V(I)*SEMU(I),I=1,IMAX)
C EMU(I,L) = EXPECTED COUNTS PER MINUTE AT TIME=TO ATOMIC
C SAMPLE I IN SAMPLE L. (FITTED PARAMETER)
C EN = (C/T)/[(1.-TAU*(C/T))
C ENSTA = CSSTAR/TSSTAR
C ESAMP = SAMPLING ERROR (PERCENT)
C I = SAMPLE INDEX
C IJ = SINGLE SUBSCRIPT EQUIVALENT TO DOUBLE SCRIPT I,J.
C IMAX = NO. SAMPLES OF RADIOACTIVE MATERIAL
C IF = 1 FOR K=1, 2 FOR K GREATER THAN 1
C ISAMP = 1 IF IMAX=1, 2 IF IMAX IS GREATER THAN 1
C J = OBSERVATION INDEX
C JMAX I = JMAX(I) FOR SOME I
C JMAX(I) = NO. OF OBSERVATIONS OF THE I TH SAMPLE
C K = ITERATION INDEX
C KMAX = MAXIMUM NUMBER OF ITERATIONS ALLOWED
C L = ISOTOPE INDEX
C L1*L2 = DUMMY INDEXES
C LMAX = NO. OF ISOTOPES IN THE SOURCE SOLUTION
C LMAX1 = LMAX+1
C N = 1 OR 2, A CONTROL VARIABLE
C S2 = TOTAL VARIANCE DUE TO MECHANICAL AND POISSON
C SEMU(I) = SUM(EMU(I,L)*L=1,LMAX)

CONTINUED
Figure 4. Continued.

C STUT = STUDENT'S T FOR IMAX-1 DEGREES OF FREEDOM
0055
C TO GIVE 95 PERCENT CONFIDENCE LIMITS.
0056
C SUMPX = SUM(A(L1*L2)+L1=1,LMAX),L2=1,LMAX)
0057
C T = LENGTH OF TIME (MINUTES) FOR OBSERVATION OF SAMPLE
0058
C TAU = DETECTOR DEADTIME (MINUTES)
0059
C TEMP,TEMP1... = TEMPORARY STORAGE
0060
C TIME = TIME AT BEGINNING OF TIME INTERVAL T
0061
C TO = HISTORIC TIME REFERENCE
0062
C TSTAR = LENGTH OF BACKGROUND OBSERVATION (MINUTES)
0063
C U = 'NUMBER' OF TIMES BACKGROUND OBSERVATION IS USED
0064
C VEMU = VARIANCE OF FMU
0065
C VMCH = MECHANICAL COMPONENT OF VARIANCE (PROPORTIONAL)
0066
C VMCHH = HISTORIC MECHANICAL VARIANCE
0067
C VMCHW = CURRENT VALUE OF VMCH USED FOR WEIGHTING
0068
C VSAMP = SAMPLING COMPONENT OF VARIANCE (PROPORTIONAL)
0069
C VSAMPH = HISTORIC SAMPLING VARIANCE
0070
C VSEMU(I) = VARIANCE OF SEMU(I)
0071
C W = WEIGHT TO DETERMINE MECHANICAL COMPONENT
0072
C X(I) = INDEPENDENT VARIABLE IN THE LEAST SQUARES FIT
0073
C X(LMAX) = DEPENDENT VARIABLE IN THE LEAST SQUARES FIT
0074
C SENSE SWITCH 1 FOR MAJOR STATISTICS OUTPUT
0075
C SENSE SWITCH 2 ON FOR OUTPUT OF TRANSFORMED DATA
0076
C SENSE SWITCH 3 ON FOR OUTPUT OF WEIGHTS
0077
C CONTINUED
Figure 4. Continued.

C SENSE SWITCH 4 ON IF DIFFERENT BACKGROUND FOR EACH OBSERVATION 0082
C SORT RESULTANT OUTPUT ON CC 2 AND LIST ON 407 0083
C * 0084
C * 0085
C DEFINE DISK(20,2000) 0086
C DIMENSION X(5),A(5,6),EMU(25,5),VSEM(25),VSEM(25),JMAX(25) 0087
1 AMADA(5),V(25),AVM(25),AVP(25) 0088
COMMON X,W,AVM1J,AVPIJ,ENSTAR,ENJ,TIME,CSTAR,TSTAR,C,T, 0089
1 A,EMU,SEMUD,SEMUD,JMAX,AMADA,V,AVX,AVP, 0090
2 AVMECH,AVPAM,AVPOSN,AVSAMP,DELTAL,DG,FMECH,EMUDT,EMU0,ESAMP,I,IJ, 0091
3 IMAX,IR,VSAMP,JMAX,K,KMAX,LMAX,LMAX1,S2,SUMMX,TAU,T0,VEU0, 0092
4 VMECH,VMECHH,VSAMP 0093
COMMON VMECHH,DFMECH,VSAMPh,DFSAMP,SUMDF,ESAMPS,EMECHS 0094
FMECH=0, 0095
DFMCH=0, 0096
ESAMP=0, 0097
DFSAMP=0, 0098
10 CALL INPUT() 0099
CALL INPUT 0100
CALL INPUT2 0101
CALL BCKGND 0102
C BEGIN ITERATIONS FOR VARIANCE COMPONENT DUE TO MECHANICAL CAUSES 0103
K=1 0104
12 IJ=1 0105
VMECH=0, 0106
I=1 0107
CONTINUED
Figure 4. Continued.

14 CALL LSOG
   CALL MISSLF(N)
   GO TO(20,10),N
20 CALL SANDS
   CALL SANDS1
   I=I+1
   IF(IMAX-I),25,14,14
25 CALL MECH(N)
   GO TO(40,50),N
40 K=K+1
   IF(KMAX-K),42,12,12
42 PUNCH 45
45 FORMAT(20H, *CHECK CONVERGENCE)
50 GO TO(60,55),ISAMP
55 CALL SAMPL1
   CALL SAMPL2
   CALL CMPNTS
60 CALL LIMITS(STUDENT(DF))
   GO TO(10,65),ISAMP
65 CALL SPANDC
   GO TO 10
C THE FOLLOWING ARE DUMMY STATEMENTS TO FORCE LOADING
C OF INLINE SUBROUTINES WITH MAIN PROGRAM.
70 CALL FANDW(EXPF(SORTF(ABSF(DF))))
   CALL EXIT
END

CONTINUED
SUBROUTINE FANDW(N)
C
5
10
20
30

W, L, WILCOXON
DIMENSION X(6), A(5, 6), EMU(25), SEMU(25), VSEMU(25), JMAX(25),
AMDA(5), V(25), AVM(25), AVP(25)
COMMON X, W, AVMI, AVPIJ, ENSTAR, ENU, TME, CSTAR, TSTAR, C, T,
A, EMU, SEMU, VSEMU, JMAX, AMDA, V, AVM, AVP,
2 AVMECH, AVPAM, AVPSON, AVSAMP, DFLTA, DF, EMCH, EMUDT, EMUO, ESAMP, I, IJ,
3 IMAX, IR, ISAMP, JMAX, K, KMAX, LMAX, LMAX1, 52, SUMMX, TAU, TC, VEMUC,
4 VMFCH, VMFCH, VSAMP
COMMON VMCHH, DFMECH, VSAMP, DFSAMP, SUMDF, ESAMPS, EMFCS
DIMENSION VECTOR(17)
EQUIVALENCE (VECTOR, T)
FETCH(IJ) VECTOR
GO TO (70, 10, 10) N
10 EMUDT=.9
DO 20 L=1, LMAX
20 EMUDT=EMUDT+EMU(I, L)*X(L)
GO TO (71, 30, 70) N
30 AVPIJ=(EMUDT+ENSTAR)/T+ENSTAR/(T+TSTAR)
AVMIJ=VMCHH*EMUDT**2
W=1/(AVMIJ+(EMUDT+ENSTAR)/T+ENSTAR/TSTAR)
IJ=IJ-1
IF(SENSF SWITCH 3)40, 60

Figure 4. Continued.
Figure 4. Continued.

40 PUNCH 50,K,IJ,AVPIJ,AVM2J,W
50 FORMAT(2H 2,i3,i5,3E15.8)
60 RECORD(IJ) VECTOR
70 RETURN
   END

ZDPOS
ZZJOB
ZDUP
*DELETINPUTO
ZZFOR
*LDISKINPUTO

SUBROUTINE INPUTO
  COMMON X,AVMJ,AVPJ,ENSTAR,EN,U,TIME,CSTAR,TSTAR,K,T,
  1 AMDA,EMU,AVM,VA,AVM,AVP,
  2 AVMECH,AVPAM,AVPSON,AVSAMP,DELTA,DF,EMECH,EMU0,ESAMP,I,IK,
  3 IMAX,IR,ISAMP,JMAXI,K,KMAX,LMAX,LMAX1,S2,SMUX,TAU,T0,VEMU0,
  4 VMCEM,VMCECM,VMCECH,VMCECM,VSAMP,VSAMP,SUMDF,ESAMP,FMECHS
  EQUIVALENCE(X(1),EM), (X(2),DFM), (X(3),ES), (X(4),DFS), (X(5),EMSTR)  
  READ 20
  PUNCH 10
  10 FORMAT(45H1) STATISTICAL ANALYSIS OF NUCLEAR DATA (SAND)/
  PUNCH 20
  20 FORMAT(80H

CONTINUED
Figure 4. Continued.

30 FORMAT(215,7F10.0)
   PUNCH 40,IMAX,LMAX,TO,TAU
40 FORMAT(19H NUMBER OF SAMPLES,I14/20H NUMBER OF ISOTOPES,I13/
   1 22H HISTORIC TIME AT T=0,
   2 F11.2,4H MIN/19H DFTFCTOR DEADTIME,F14.2,10H MIN/COUNT)
   READ 50,(AMDA(L),L=1,LMAX)
50 FORMAT(5E15.8)
   PUNCH 60,(AMDA(L),L=1,LMAX)
60 FORMAT(17H DECAY CONSTANTS,F16.2,6H 1/MIN/(17X,F16.2))
   READ 70,(JMAX(I),I=1,IMAX)
70 FORMAT(1515)
   PUNCH 80,(JMAX(I),I=1,IMAX)
80 FORMAT(25H OBSERVATIONS PER SAMPLE,F18/(25X,F18))
   RETURN
END

ZJJOBI
ZJDU
*DELETEINPUT
ZJFOR
*LDISK

SUBROUTINE INPUT1
C
C CONTINUATION OF INPUT ROUTINE
DIMENSION X(6),A(5,6),EMU(25,5),SEM(25),VSEM(25),JMAX(25),
1 AMDA(5),V(25),AVM(25),AVP(25)
COMMON X,W,AVM,AVP,ENSTAR,FN,TIME,CSTAR,TSTAR,C,T,
1 A,EMU,SEMU,VSEM,MAX,AMDA,V,AVM,AVP,
2 AVMECH,AVPAM,AVPOSN,AVSAMP,DELTA,DF,EMECH,EMUDT,EMU,ESAMP,I,II,

CONTINUED
Figure 4. Continued.

```plaintext
3 IMAX, IR, ISAMP, JMAXI, K, KMAX, LMAX, LMAXI, S2, SUMX, TAU, T0, VEMUO*
4 VMECH, VMECHW, VSAMP
COMMON VMECHH, DFMECH, VSAMPH, DFSAMP, SUMDF, ESAMPS, EMECHS
EQUIVALENCE (X(1)*EM), (X(2)*DFM), (X(3)*ES), (X(4)*DFS), (X(5)*EMSTRT)
LMAX1=LMAX+1
IF (FM+DFM) 93, 93, 92
92 DFMECH=DFM
  VMECHH=(0.01*EM)**2
  VMECHW=(0.01*EMSTRT)**2
  EMECH=EM
  GO TO 94
93 VMECHW=(0.01*EMECH)**2
  VMECHH=VMECHW
94 IF (VMECHW) 95, 95, 96
95 VMECHW=0.01
96 VMECH=VMECHW
  IF (ES+DFS) 110, 110, 100
100 DFSAMP=DFS
    ESAMP=ES
110 ESAMPS=ESAMP
    VSAMP=(0.01*ESAMP)**2
    VSAMPH=VSAMP
    IF (VSAMP) 120, 120, 130
120 VSAMP=0.01
130 IR=1
    SUMDF=0.
    DO 140 I=1, IMAX
       JMAXI=JMAX(I)
       DF=JMAXI-LMAX
       CONTINDED
```
Figure 4. Continued.

140 SUMDF=SUMDF+DF
       PUNCH 150,EMECH,DFMECH,ESAMP,DFSAMP
150 FORMAT(27H HISTORIC MECHANICAL ERROR,F6.2,8H PERCENT/
1 7X,18DEGREES OF FREEDOM,18/
2 25H HISTORIC SAMPLING ERROR,F8.2,8H PERCENT/
3 7X,18DEGREES OF FRFFDOM,18)
       KMAX=25
       DELTA=0.005
       PUNCH 160,DFLTA,KMAX
160 FORMAT(20H TOLERANCE ON ERROR,F13.3,8H PERCENT/
1 28H MAXIMUM ITERATIONS ALLOCATED,15)
       RETURN
       END

ZZJOB
ZZDUP
*DELETEINPUT2
ZZFOR
*LDISK

SUBROUTINE INPUT2

C W. L. WILCOXON

C CONTINUATION OF INPUT ROUTINE

DIMENSION X(6),A(5,6),EMU(25,5),SEM(25),VEMU(25),JMAX(25),
1 AMDA(5),V(25),AVM(25),AVP(25)

COMMON X,W,AVMJ,AVPJJ,ENSTAR,EN,TIME,CSTAR,TSTAR,C,T,
1 A,EMU,FSMU,JSMU,JMAX,AMGA,V,AVM,AVP,
2 AVMECH,AVPAM,AVPOSN,AVSAMP,DELTA,DF,EMECH,EMUDT,EMU0,FSAMP,1,1J,
3 IMAX,IR,ISAMP,JMAX,I,K,KMAX,LMAX,LMAX1,S2,SUMX,TAU,T0,VEMU0,

CONTINUED
**Figure 4. Continued.**

```
4 VMECH,VMECHW,VSAMP
COMMON VMECHH,DFMECH,VSAMPH,DFSAMP,SUMDF,ESAMPS,EMECHS
DIMENSION VECTOR(17)
EQUIVALENCE VECTOR,T
I=1
DO 60 I=1,IMAX
JMAXI=JMAX(I)
DO 60 J=1,JMAXI
READ 10,T,C,TSTAR,CSTAR,TIME
10 FORMAT(5E15,8)
TIME=TIME-TC
C TRANSFORM DEPENDENT VARIABLE AND CORRECT FOR DETECTOR DEADTIME
EN=(C/T)/(1.-TAU*(C/T))
FNSTAR=CSTAR/TSTAR
C CORRECT DEPENDENT VARIABLE FOR BACKGROUND
X(LMAXI)=EN-ENSTAR
C TRANSFORM INDEPENDENT VARIABLE AND CORRECT FOR DECAY
DO 30 L=1,LMAX
X(L)=1.
IF(AMDA(L)>30,30,20)
20 X(L)=EXP(-AMDA(L)*TIME)*(1.-EXP(-AMDA(L)*TIME))/(AMDA(L)*TIME)
30 CONTINUE
U=0.
IF(SENSF SWITCH 4)40,50
40 U=1.
C COMPUTE WEIGHT FOR FIRST ITERATION
50 AVPIJ=EN/T*ENSTAR/TSTAR
AVMIJ=VMECHW*X(LMAXI)**2
W=1./(AVPIJ+AVMIJ)
```

CONTINUED
Figure 4. Continued.

10 IJR=1
IJM=0
DO 130 II=1,I*MAX
IJ0=IJM+1
IJM=IJ0+JMAX(II)-1
DO 130 J1=IJ0,IJM
FFETCH(IJR)VECTOR
IJR=IJR-1
IJ=IJ0
C COMPUTE NUMBER OF TIMES BACKGROUND IJR IS USED FOR SAMPLE II
DO 50 K1=IJ0,IJM
FETCH(IJ)VCTR
IFI(ENSTAR-FNSTR)50,20,50
20 IF(IJ-1-IJR)30,40,40
C U IS ALREADY COMPUTED FOR THIS BACKGROUND READING
30 U=U1
GO TO 100
40 U=U+1
50 CONTINUE
C COMPUTE NUMBER OF SAMPLES FOR WHICH BACKGROUND IJR IS USED
IJ=1
IJNXT=1
COUNT=1
DO 90 I=1,I*MAX
JMAX=JMAX(I)
IJNXT=IJNXT+JMAX
IF(I-I1)60,90,60
CONTINUED
60 DO 70 J=1,JMAXI
   FETCH(IJ)VCVR
   IF(ENSTAR-ENSTR)70,80,70
70 CONTINUE
   GO TO 90
   80 COUNT=COUNT+1.
90 IJ=IJNXT
C UPDATE U AND RF-RECORD RECORD
   U=U*COUNT
   100 IF(SFNSF SWITCH 2)110,130
   110 PUNCH 120, IJR, T, CSTAR, TIME, IJR, U, EN, ENSTAR, X(LMAX), W,
       1 IJR, (X(L), L=1,LMAX)
   120 FORMAT(2H1*13,5E15.8/2H2*13,5E15.8/2H3*13,5E15.8)
   130 RECORD(IJR)VECTOR
   140 RETURN
END
ZZJOB
LSOG
ZZDUP
*DELETLSOG
ZZFOR
*LDISK
SUBROUTINE LSOG
C W, L, WILCOXON
C GENERATE LEAST SQUARES MATRIX
   DIMENSION X(5), A(5,5), EMU(25,5), SEMU(25), VSEMU(25), JMAX(25),
   1 AMDA(5), V(25), AVM(25), AVP(25)
   COMMON X,W, AVMIJ, AVPIJ, ENSTAR, EN, U, TIME, CSTAR, TSTAR, CS
   CONTINUED
Figure 4. Continued.

1 A*EMU+SEMU*VSEMU*JMAX*AMDA*V+AVM*AVP,
2 AVMECH+AVPAM+AVPOSN+AVSAMP+DELTA+DF*EMECH+EMUDT*EMUO*ESAMP*I*IJ,
3 JMAX*IR*ISAMP+JMAX*K*KMAX*LMAX*LMAX1*S2*SUMM*X*TAU*T0*VEMUO,
4 VMECH+VMECHW+VSAMP
COMMON VMECHH+DFMFCH+VSAMPH+DFSAMP+SUMDF+ESAMPS+EMECHS
DO 10 L=1,LMAX
DO 10 L1=1,LMAX1
10 A(L,L1)=0.
JMAX=JMAX(I)
DO 20 J=1,JMAX1
CALL FANDW(IR)
DO 20 L=1,LMAX
DO 20 L1=1,LMAX1
20 A(L,L1)=A(L,L1)+W*X(L)*X(L1)
RETURN
END

ZZJOB
ZDUP
*DELETMISSILE
ZZFOR
*LDISK
SUBROUTINE MISSLE(N)
C
W* L* WILCOXSON
C
MATRIX INVERSION AND SOLUTION OF SIMULTANEOUS LINEAR EQUATIONS
DIMENSION X(6),*(5,6),EMU(25),5,SEMU(25),VSEMU(25),JMAX(25),
1 AMDA(5),V(25),AVM(25),AVP(25)
COMMON X,W*AVM1J,AUPIJ,ENSTAR,EN..U,TIMF,CSTAR,TSTAR,C,T,

CONTINUED
Figure 4. Continued.

1 A,EMU,SEMU,VSEMU,JMAX,AMDA,V,AVM,AVP,
2 AVMECH,AVPAM,AVPOSN,AVSAMP,DELTA,DF,FMECH,EMU,T,EMU,ESAMP,I,IJ,
3 IMAX,IR,ISAMP,JMAX,K,KMAX,LMAX,LMAX1,SZ,SUMX,TAU,T,T,VECMU,
4 VMECH,VMECHW,VSAMP
COMMON VMECHH,DFMECH,VSAMPH,DFSAMP,SUMDF,ESAMPS,EMECS
N=1
DO 70 L=1,LMAX
TEMP=A(L,L)
A(L,L)=0.
IF(TEMP)30,10,30
10 N=2
TYPE 20
20 FORMAT(34HZERO ELEMENT ON DIAGONAL OF MATRIX/14HCASE ABANDONED)
RETURN
30 DO 40 L1=1,LMAX1
40 A(L,L1)=A(L,L1)/TEMP
DO 70 L2=1,LMAX
IF(L2-L)50,70,50
50 TEMP=A(L2,L)
A(L2,L)=0.
DO 60 L1=1,LMAX1
60 A(L2,L1)=A(L2,L1)-TEMP*A(L,L1)
70 CONTINUE
RETURN
END
CONTINUED
SUBROUTINE SANDS
C
C SAVE PARAMETERS AND SUM MATRIX ELEMENTS
C COMPUTE TOTAL VARIANCE S2 FOR SAMPLE I
C PARTITION TOTAL INTO MECHANICAL AND POISSON COMPONENTS
C
DIMENSION X(16),A(5,6),EMU(25,5),SEMU(25),VSEMU(25),JMAX(25),
1 AMDA(5),V(25),AVM(25),AVP(25)
COMMON X,W,AVMIJ,AVPIJ,ENSTAR,ENSTAR,TIME,CSTAR,TSTAR,CST,
1 A,EMU,SEMU,VSEMU,JMAX,AMDA,V,AVM,AVP,
2 AVMECH,AVPAM,AVPOSN,AVSAMP,DELTA,DF,FMCH,EMUO,ESAMP,I,IJ,
3 IMAX,IR,ISAMP,JMAXI,K,KMAX,LMAX,LMAX1,S2,SUMX,TAU,T0,VEMUO,
4 VMCH,VMCHW,VSAMP
COMMON VMCHH,DFMECH,VSAMP,DFSAMP,SUMDF,ESAMPS,EMECHS
DO 10 L=1,LMAX
10 EMU(I,L)=A(L,LMAX1)
SUMO=0.
SUMP=0.
SUMM=0.
SUMW=0.
SUMV=0.
IJ=IJ-JMAX
CONTINUED
Figure 4. Continued.

DF=JMAXI-LMAX
EJMAXI=JMAXI
DO 20 J=1,JMAXI
CALL FANDW(3)
TEMP=W*(X(LMAXI)-EMUDT)**2
TEMP1=TEMP/(AVMIJ+AVP1J)
SUMVM=SUMVM+AVMIJ*TEMP1/EMUDT**2
SUMP=SUMP+AVP1J*SUMP1
SUMM=SUMM+AVMIJ*TEMP1
SUMSQ=SUMSQ+TEMP
20 SUMW=SUMW+W
S2=SUMSQ/(DF*SUMW)
VMech=VMech+EJMAXI*SUMVM/SUMW
SUMX=0
SEMU(I)=0
DO 30 L=1,LMAX
SEMU(I)=SEMU(I)+EMU(I,L)
DO 30 L=1,LMAX
A(L,L1)=SUMW*A(L,L1)
30 SUMMX=SUMMX+A(L,L1)
VSEMU(I)=S2*SUMMX
AVP(I)=SUMP*SUMMX/(DF*SUMW)
AVM(I)=SUMM*SUMMX/(DF*SUMW)
RETURN
END

CONTINUED
Figure 4. Continued.

**ZZJOB**

**ZGDUP**

*DELETSANDS1*

**ZZFOR**

*LDISKSANDS1*

**SUBROUTINE SANDS1**

**C**

**CONTINUATION OF SANDS SURROUNTE**

**C**

*W. WILCOXON*

**DIMENSION X(6),A(5,6),EMU(25,5),SEMU(25),VSEMUI(25),JMAX(25),**

**1 A(MAX,5),V(25),AVM(25),AVP(25)**


**1 A*,EMU*,SEM*,V*,MAX*,A*,AVM*,AVP**


**4 VM*,VMECH*,VSA**

**COMMON VMECH*,DFMECH*,VSA*,DFSA*,SUMDF*,ESAMP*,S*MECH**

**IF(SENSE SWITCH 1)40 C*90**

**40 DO 50 L=L,M**

**50 PUNCH 66*,I*,K*,A(L,L1)*L1=1*L1**

**60 FORMAT(2H 4,13.5,5E14.8)**

**PUNCH 70*,I*,K*,EMU(I,L)*L=1*LMAX**

**70 FORMAT(2H 5,13.5,5F14.8)**

**PUNCH 80*,I*,K*,SEMUI(I,L)*L*AVM(I)*AVP(I)*S2**

**80 FORMAT(2H 6,13.5,5E14.8)**

**90 RETURN**

**END**

**CONTINUED**
ZZJOB
ZZDUP
*DELETMECH
ZZFORD
*LDISK

SUBROUTINE MECH(N)
W. L. WILCOXON

COMPUTE MECHANICAL COMPONENT OF ERRORS
DIMENSION X(6),A(5,6),EMU(25,5),VSEMU(25),JMAX(25),
1 AMDA(5),V(25),AVM(25),AVP(25)
COMMOT X(5),AVMJ,AVPI,ENSTAR,EMU,TIME,CSTAR,TSTAR,C,T,
1 A*EMU,SEMUEMU,JMAX,AMDA,V,AVM,AVP,
2 AVMECH,AVPM,AVPSN,AVSAMP,DFLT,DF*FMECH,FMUO,TFM,ESAMP,1,1J,
3 IMAX,IR,ISAMP,JMAK,1,K,KMAX,LMAX,LMAX,5,M2,SUMM,TAU,TC,VEMU0,
4 VMECH,VMECHV,VEAM

COMMON VMECHH,DFMECH,VSAMP,DFSAMP,SMDF,ESAMPS,EMECHS
N=1
VMECH=VMECH/SUMDF
EMECH=EMECH
EMECH=100.*SORTF(VMECH)
GO TO(20,40),1R
20 PUNCH 30
30 FORMAT(1HO,14X,16HMECHANICAL ERROR/,
1 32H ITERATION CURRENT HISTORIC)
IR=2
40 VMECH=(DFMECH*VMECHH+SUMDF*VMECH)/(DFMECH+SUMDF)
EMECHS=100.*SORTF(VMECH)
PUNCH 50,K,EMECH,EMFCHS

CONTINUED
Figure 4. Continued.

50 FORMAT(15,F15.2,F10.2)
10 IF(ABSF(EMECH-EMECHO)-DELTA)90,90,100
90 N=2
100 VMECHW=VMECH
RETURN
END

ZZJOB
SAMPL1
ZZDUP
DELETESAMPL1
ZZFOR
LDISK

SUBROUTINE SAMPL1

C W. L. WILCOXSON
C CALCULATE AVERAGE DEFAY RATE AT TIME TO
DIMENSION X(6)*A(5,6)*EMU(25,5)*SEMU(25,5),VSFMU(25,5),JMAX(25),
1 AMDA(5,5),V(25,5),AVM(25,5),AVP(25)
COMMON X,W,AW,AVI,AVPI,ENSTAR,FMU,TIME,CSTAR,TSTAR,C,T,
1 A,SMU,SEMU,VEMU,JMAX,AMDA,V,AVM,AVP,
2 AVMECH,AV,AVP,AVPOSN,AVSAMP,DELTA,DF,EMECH,EMUDT,FMU0,FSAMP,1,1,
3 IMAX,IR,ISAMP,JMAXI,K,KMAX,LMAX,LMAX1,S2,SUMX,TAU,T0,VEMU0
4 VMECH,VMECHI,VSAMP
COMMON VMECHH,DFMECH,VSAMPH,DFSAMP,VSAMPH,ESAMP,H,EMECHS
C CALCULATE INITIAL ESTIMATE OF MEAN
SUMV=N
SUM=0
DO 10 I=1,IMAX
TEMP1=1/((SMU(I)**2)*VSAMP+VSFMU(I))
SUMV=SUMV+TEMP1
10 CONTINUE
10 SUM=SUM+TEMP1*SEMU(I)
   EMUN=SUM/SUMV
C ITERATE TO FIND ESTIMATE OF MEAN BASED ON T TORIC SAMPLING ERROR
   DO 30 K=1,KMAX
   TEMP2=FMUN
   SUMV=0*
   SUM=0*
   TEMP=VSAMPLE*EMUN**2
   DO 20 I=1,IMAX
      TEMP1=1./(TEMP+VSEMU(I))
      SUMV=SUMV+TEMP1
20 SUM=SUM+TEMP1*SFMU(I)
   FMUN=SUM/SUMV
   IF(ABSF((EMUN-TEMP2)/EMUN)-0.00005)<0.01 40,40,30
30 CONTINUE
40 PUNCH 50
50 FORMAT(1H0,15X,14HSAMPLING ERROR/
             1 32H ITERATION CURRENT HISTORIC)
   RETURN
END
ZZJOB      SAMPL2
ZZDUP
*DELETESAMPL2
ZZFOR
*LDISK
   SUBROUTINE SAMPL2
C W. L. WILCOXSON
CONTINUED
Figure 4. Continued.

C BEGIN ITERATIONS FOR SAMPLING ERROR
C AND AVERAGE COUNTS/MINUTE AT TIME TO
DIMENSION X(6), A(5, 6), EMU(25, 5), VEMU(25), JMAX(25),
1 AMDA(5), VM(25), AV(25)
COMMON X, W, AVMIJ, AVPIJ, ENSTAR, DU, TIME, CSTAR, TSTAR, C, T,
1 A, EMU, VEMU, JMAX, AMA, VM, AV, AP, VM, EMU
2 AVMECH, AVPAM, AVPOSN, AVSAM, DELTA, EMU, EMECH, EMU, EMU, EMU, EMU, EMU
3 IMAX, IR, ISAMP, JMAX, K, KMAX, LMAX, S, SUMX, TAU, T0, VEMU
4 VMECH, VMECHV, VSAMP
COMMON VMMECH, DMECH, VSA, DSA, SAMP, SUMDF, ESAMPS, EMECHS
EI=IMAX
DEF=IMAX-1
ESAMP=100.*SORTF(VSAM)
DO 70 K=1, KMAX
AVSAM=VSAMP*EMU**2
ESAMPO=FSAMP
C COMPUTE AVERAGE DECAY RATE AT TIME TO
SUM=0.
SUMV=0.
DO 10 I=1, IMAX
TEMP1=1./(AVSAM+VEMU(I))
VI(I)=TEMP1
SUMV=SUMV+TEMP1
10 SUM=SUM+TEMP1*SEMU(I)
EMU0=SUM/SUMV
C CALCULATE ABSOLUTE SAMPLING VARIANCE
SUM50=0.
SUMSAM=0.
Figure 4. Continued.

DO 20 I=1,IMAX
TEMP1=V(I)
V(I)=V(I)/SUMV
TEMP2=V(I)*(SEMU(I)-EMUO)**2
SUMSAM=SUMSAM+AVSAMP*TEMP1*TEMP2
20 SUMSO=SUMSO+TEMP2
VEMU=SUMSO/DF
AVSAMP=SUMSAM*EI/DF
VSAMP=AVSAMP/EMUO**2
ESAMP=100.*SORTF(VSAMP)
VSAMP=(DFSAMP*VSAMPH+DC*VSAMP)/(DFSAMP+DF)
ESAMPS=100.*SORTF(VSAMP)
PUNCH 30,K,ESAMP,ESAMPS
30 FORMAT(15,F15.2,F10.2)
IF(SENSE SWITCH 1)40,60,60
40 PUNCH 50,K,EMUC,EMUC,AVSAMP,(K,I,V(I),I=I,IMAX)
50 FORMAT(12H 8,13,3F15.8/(2H 9,13,15,E15.8))
60 IF(ABS(DOMESAMP-ESAMP0)-DELTAS*9.970,70
70 CONTINUE
80 PUNCH 80
CONTINU
80 FORMAT(20H**CHECK CONVERGENCE)
90 RETURN
END

CONTINUED
Figure 4. Continued.

2ZJOB
2ZDUP
*DELETCMPNTS
2ZFOR
*LDISK

SUBROUTINE CMPNTS

M. L. WILCOXSON

C PARTITION TOTAL VARIANCE INTO ITS THREE COMPONENTS

DIMENSION X(6),A(5,6),EMU(25,5),SFMU(25),VSEMU(25),JMAX(25),
1 AMDA(5),V(25),AVM(25),AVP(25)

COMMON X,W,AVM1J,AVPIJ,ENSTAR,EU,TIME,CSTAR,TSTAR,C,T
1 A,EMU,SFMU,VSEMU,JMAX,AMDA,V,AVM,AVP,
2 AVMECH,AVPM,AVPOSN,AVSAMP,DFLTA,DF,FMECH,EMU,ESAMP,I,JI
3 IMAX,IR,ISAMP,JMAX1,K,LMAX1,LMAX2,SMUX,TAU,TO,EMU,
4 VMECH,VMECH1,VSAMP,

COMMON VMECHH,DFMECH,VSAMPH,DFSAMP,SUMDF,ESAMPS,EMECHS

SUMV2=0.
ALPHA=0.
BETA=0.

DO 10 I=1,IMAX

TEMP=V(I)**2
SUMV2=SUMV2+TEMP

ALPHA=ALPHA+TEMP*AVP(I)

10 BETA=BETA+TEMP*AVM(I)

GAMMA=SUMV2*VSAMP*EMU**2

SUM=ALPHA+BETA+GAMMA

AVPOSN=(ALPHA/SUM)*EMU
Figure 4. Continued.

AVMECH = (BETA/SUM) * VEMU
AVSAM P = (GAMMA/SUM) * VEMU
PUNCH 20
20 FORMAT (29H0 MEAN DECAY RATE AT TIME T=0/
1 9X, 14H ALL ISOTOPES) / 6X, 2D ANALYSIS OF VARIANCE
PUNCH 30, AVSAMP, AVMECH, AVPOSN, VEMU
30 FORMAT (30H COMPONENT VARIANCE/)
1 30H COMPONENT VARIANCE/
2 30H -------------------------------/
3 13H SAMPLING * F17*0/13H MECHANICAL * F17*0/13H POISSON
4 F17*0/30H -----------------------------/ 13H TOTAL * F17*0)
RETURN
END

ZZJOB
ZZDUP
*DELET LIMITS
ZZFOR
*LDISK

SUBROUTINE LIMITS(STUT)
C
W. I. MILCOXON
C
OUT PUT MEANS AND 0.95 CONFIDENCE LIMITS ON MEANS
DIMENSION X(5), AJX(6), EMU(25), SEMU(25), VSEM(25), JMAX(25),
1 AMN(5), VM(25), AVP(25)
COMM ON XW, AVMIJ, AVPIJ, ENSTAR, ENU, TIME, CSTAR, ISTAR, C, T,
1 A, EMU, SEMU, VSEM, JMAX, AMN, VM, AVM, AVP,
2 AVMECH, AVPAM, AVPOSN, AVSAM, DELTA, OF, EMFCH, EMUDT, EMUQ, ESAMP, I, J
3 IMAX, IR, ISAMP, JMAX, K, KMAX, LMAX, LMAX1, S2, SUMX, TAU, TO, VEMU,

CONTINUED
Figure 4. Continued.

```
4 VMECH, VMECHW, VSAMP
   COMMON VMECHH, DFMECH, VSAMPH, DFSAMP, SUMDF, FSAMPS, EMECHS
   GO TO (2,3), ISAMP
2 VEMU0=VSEMU11
   EMUC=SEMU11
3 TEMP=STUT*SORTF(VEMU0)
   TEMPI=EMU0-TEMP
   TEMPS=FMUC+TEMP
   PUNCH 4, TEMPI, EMU0, TEMPS
4 FORMAT(1HO, 9X), 35HDECAY RATE (COUNTS/MIN) AT TIME T=0/
   1 9H ISOTOPE, 5X, 10H, 95 L, C, L, 5X, 9HMEAN RATE, 6X, 10H, 95 U, C, L/
   2 7H ALL, 3F15, 1)
   IF(LMAX-1), 11, 5
C COMPUTE MEAN AND VARIANCE FOR EACH ISOTOPE
5 DO 10 L=1, LMAX
   GO TO (6,7), LMAX
6 AVG=FMU11, L
   VAR=52*A(L,L)
   GO TO 9
7 AVG=0
   SUMS0=0
   DO 8 I=1, IMAX
      AVG=AVG+V(I)*EMU(I,L)
   8 SUMS0=SUMS0+V(I)*EMU(I,L)**2
   VAR=(SUMS0-AVG**2)/DF
C CALCULATE CONFIDENCE LIMITS
9 TEMP=STUT*SORTF(VAR)
   TEMPI=AVG-TEMP
   TEMPS=AVG+TEMP
```

CONTINUED
10 PUNCH 12,L,TEMPI,AVG,TEMP2
12 FORMAT(4X,I2,1X,3F15.1)
11 DFMECH=DFMECH+SUMDF
FMECH=FMECHS
DF=IMAX-1
DFSAMP=DFSAMP+DF
ESAMP=ESAMPS
PUNCH 20,DFMECH,ESAMP,DFSAMP
20 FORMAT(35H0 UPDATED HISTORIC MECHANICAL ERROR,F6.2,8H PERCENT/
1 15X,18DEGREES OF FREEDOM,18/
2 33H UPDATED HISTORIC SAMPLING ERROR,F8.2,8H PERCENT/
3 15X,18DEGREES OF FREEDOM,18)
RETURN
END
ZZJOB
ZZDUP
*DELETPANDC
ZZFOR
*LDISKPANDC
SUBROUTINE SPANDC
C
C
C
C
COMMON X,W,AVMIJ,AVPIJ,ENSTAR,ENU,TIME,CSTAR,TSTAR,C,T,
1 AVMECH,AVPAM,AVPOSN,AVSAM,DFLTA,DF,ESEC,FMUT,EMUG,ESAMP,I,IJ,
Figure 4. Continued.

3 IMAX, IR, ISAMP, JMAX, J, KMAX, LMAX, LMXX, TAU, TO, VEMUC
4 VMECH, VMECHW, VSAMP
COMMON VMECH, DFMECH, VSAMP, DFSAMP, SUMDF, ESAMPS, EMFCHS
PUNCH 10, (L, L=1, 5)
10 FORMAT (1HO, 35X, 7HISOTOPE/15X, 3HALL, 5112/
18H SAMPLE, 21X, 40HMEAN DECAY RATE (COUNTS/MIN) AT TIME T=0)
DO 20 I=1, IMAX
20 PUNCH 30, I, SEMU(I), (EMU(I, L), L=1, LMAX)
30 FORMAT (16, 2X, 6F12.1)
PUNCH 40
40 FORMAT (1HC, 25X, 27HMEAN DECAY RATE AT TIME T=0/
125X, 29H(ALL ISOTOPES - FIXED SAMPLE)/
230X, 20HANALYSIS OF VARIANCE/
38H SAMPLE, 3X, 7HWEIGHTS, 6X, 10HMECHANICAL, 3X, 7HPoisson, 4X, 5HTOTAL)
DO 50 I=1, IMAX
50 PUNCH 60, I, V(I), AVM(I), AVP(I), VSFMU(I)
60 FORMAT (16, F11.4, 5X, 3F11.0)
RETURN
END

ZZJOB
ZZDUP
*DELTSTUDNT
ZZFOR
*LDISK
FUNCTION STUDNT(DF)
C W. L. WILCOXON
C CALCULATE STUDENTS T AT 0.95 LEVEL

CONTINUED
Figure 4. Continued.

```
STUDENT=12.706
IF(DF-2)>1,3
1 STUDENT=4.393
2 RETURN
3 STUDENT=(1.96*DF+0.60033+0.95910/DF)/(DF-0.20259+0.11588/DF)
RETURN
END
ZZZZ EAD OF JOB
```
### GLOSSARY OF SYMBOLS

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_i$</td>
<td>Sum of all the $L^2$ elements of the last inverse matrix for sample $i$ in the iterated least-squares procedure.</td>
</tr>
<tr>
<td>$B_{ij}$</td>
<td>Expected value of the background for the $j^{th}$ observation on the $i^{th}$ sample.</td>
</tr>
<tr>
<td>$C, C_{ij}$</td>
<td>Counts observed during period $T, T_{ij}$.</td>
</tr>
<tr>
<td>$C^<em>, C_{ij}^</em>$</td>
<td>Counts of background activity observed during period $T^<em>, T_{ij}^</em>$.</td>
</tr>
<tr>
<td>Cov $(a, b)$</td>
<td>Estimated covariance of $a$ and $b$.</td>
</tr>
<tr>
<td>$d$</td>
<td>Number of degrees of freedom associated with the historic estimate of the sampling error, $a_{so}$.</td>
</tr>
<tr>
<td>$E$</td>
<td>The expected value or arithmetic average value operator, such that in the discrete case $E(y) = \lim_{n \to \infty} \frac{1}{n} \sum_{h=1}^{n} Y_h$, where $y$ is any random variable and $Y_1, Y_2, Y_3, ...$ are values that $y$ takes on in successive occurrences.</td>
</tr>
<tr>
<td>$f_0$</td>
<td>Number of degrees of freedom associated with the historic estimate of the mechanical error, $\sigma_{mo}$.</td>
</tr>
<tr>
<td>$f_1$</td>
<td>Number of degrees of freedom associated with the current estimate of the mechanical error, $\sigma_{mo}^*$.</td>
</tr>
<tr>
<td>$f(t)$</td>
<td>Student's $t$ probability density function.</td>
</tr>
<tr>
<td>$h$</td>
<td>An integer; $h = 0, 1, 2, ...$</td>
</tr>
<tr>
<td>$l$</td>
<td>Number of aliquot samples of radioactive solution.</td>
</tr>
<tr>
<td>$i$</td>
<td>Index to label sample; $i = 1, 2, 3, ..., I$.</td>
</tr>
<tr>
<td>$J_i$</td>
<td>Number of observations of the $i^{th}$ sample.</td>
</tr>
<tr>
<td>$j$</td>
<td>Index to label observation; $j = 1, 2, 3, ..., J_i$.</td>
</tr>
<tr>
<td>$K_{ij}$</td>
<td>A normalizing constant for the $i^{th}$ sample on the $k^{th}$ iteration in the computation of the weights $w_{ijk}$ so that $\sum_{j=1}^{J_i} w_{ijk} = 1$.</td>
</tr>
<tr>
<td>$k$</td>
<td>Iteration index used in least-squares curve-fitting procedure; also in estimating $a_s$.</td>
</tr>
<tr>
<td>$L$</td>
<td>Number of radioactive isotopes in the source solution.</td>
</tr>
<tr>
<td>$\xi$</td>
<td>Index to label isotope; $\xi = 1, 2, 3, ..., L$.</td>
</tr>
<tr>
<td>$M$</td>
<td>The mean of a normally distributed random variable. Used for illustrative purposes only when discussing the mechanical error.</td>
</tr>
<tr>
<td>$N, N_{ij}$</td>
<td>Counts per minute from both sample and background corrected for deadtime of the detector.</td>
</tr>
<tr>
<td>$N^<em>, N_{ij}^</em>$</td>
<td>Apparent counts per minute from background activity.</td>
</tr>
<tr>
<td>$P$</td>
<td>Probability.</td>
</tr>
<tr>
<td>$Q$</td>
<td>The mean of a normally distributed random variable. Used for illustrative purposes only when discussing the sampling error.</td>
</tr>
<tr>
<td>$q_{ij}$</td>
<td>A normally distributed random variable with mean zero and standard deviation $\sigma_m$ labeled for the $j^{th}$ observation of the $i^{th}$ sample used in the simulation.</td>
</tr>
</tbody>
</table>
Number of observations of the $i^{th}$ sample.

Index to label observation; $j = 1, 2, 3, ..., J_i$.

A normalizing constant for the $i^{th}$ sample on the $k^{th}$ iteration in the computation of the weights $w_{ijk}$ so that

$$\sum_{j=1}^{J_i} w_{ijk} = 1$$

Iteration index used in least-squares curve-fitting procedure; also in estimating $\sigma_s$.

Number of radioactive isotopes in the source solution.

Index to label isotope; $\ell = 1, 2, 3, ..., L$.

The mean of a normally distributed random variable. Used for illustrative purposes only when discussing the sampling error.

Counts per minute from both sample and background corrected for deadtime of the detector.

$$N = \frac{C}{T} \left( \frac{1}{1 - \tau \frac{C}{T}} \right)$$

Apparent counts per minute from background activity.

$$N^* = \frac{C^*}{T^*}$$

Probability.

The mean of a normally distributed random variable. Used for illustrative purposes only when discussing the sampling error.

A normally distributed random variable with mean zero and standard deviation $\sigma_m$ labeled for the $i^{th}$ observation of the $i^{th}$ sample used in the simulation.

$\mu_{ij}$

A normally distributed random variable with mean zero and standard deviation $\sigma_m$ labeled for the $i^{th}$ sample used in the simulation.

$S_{ik}^2$

For the $k^{th}$ iteration the weighted sum of squares of deviations of the observed counts per minute for the $i^{th}$ sample from the corresponding fitted curve divided by the degrees of freedom:

$$S_{ik}^2 = \frac{1}{J_i - L} \sum_{j=1}^{J_i} w_{ijk} \left( n_{ij} - \tilde{\mu}_{ij}(t_{ij}, T_{ij}) \right)^2$$

$S_{ij}^2 = \frac{1}{L} \sum_{i=1}^{L} w_{ijk}^2 \left( \tilde{\mu}_{ij}(t_{ij}, T_{ij}) \right)^2$

Time in minutes expended in observing a sample of radioactive solution.

Time in minutes expended in observing the background.

Age of sample in minutes at the beginning of the observation period $T$ or $T_{ij}$, where $t$ and $t_j$ are reckoned from a historic instant of time $t_0$, when $t = 0$.

A historic instant of time when the age of all samples used in a particular experiment are reckoned to be zero minutes old, that is, $t = 0$.

Variable defined by Student’s $t$ density function.

If $a$ is the number of samples for which a particular background reading was used in the computations, and $b$ is the number of times it was used just for the $i^{th}$ sample, then $U_{ij} = a b$.

$w_{ijk}$

Weight for the $i^{th}$ observation of the $i^{th}$ sample on the $k^{th}$ iteration.
GLOSSARY OF SYMBOLS (Continued)

\( X(t, T; \lambda_q) \)  Independent variable in the iterated linear least-squares curve fit (corrected for decay).
\[
X(t, T; \lambda_q) = e^{-\lambda_q t} \left( \frac{1 - e^{-\lambda_q T}}{\lambda_q T} \right)
\]

\( x_{ij} \)  Same as \( X(t_{ij}, T_{ij}; \lambda_q) \).

\( a_{ij} \)  Individual point estimate of the variation due to the Poisson phenomenon for the \( j \)th observation of the \( i \)th sample.
\[ a_{..} = \text{Crude estimate of } \left( \frac{\sigma_{..}^2}{\mu_{..}} \right) \]

\( b_{ij} \)  Individual point estimate of the variation due to mechanical causes for the \( j \)th observation of the \( i \)th sample.
\[ b_{..} = \text{Crude estimate of } \left( \frac{\sigma_{..}^2}{\mu_{..}} \right) \]

\( c_{ij} \)  Crude estimate of \( \left( \frac{\sigma_{..}^2}{\mu_{..}} \right) \).

\( e_{ij} \)  Random error on the \( j \)th observation of the \( i \)th sample.
\[ e_{..} = \text{Random error on the } \left( \frac{\sigma_{..}^2}{\mu_{..}} \right) \]

\( \eta \)  \( \eta_i = M - N^* \) and \( \eta_{ij} = N_{ij} - N_{ij}^* \)

\( \kappa_k \)  A normalizing constant in the estimation of the \( \mu_{..}^o \)'s such that
\[ \sum_{i=1}^{L} \kappa_k = 1 \]

\( \lambda_q \)  A decay constant in reciprocal minutes for the \( q \)th isotope, \( q = 1, 2, \ldots, L \). It is assumed known and free from error in this report.

\( \mu_{ij}^o(t) \)  Expected instantaneous counts per minute from isotope \( L \) of sample \( i \) at age \( t \), corrected for background and deadtime of the detector.
\[ \mu_{ij}^o(t) = \mu_{ij}^o e^{-\lambda_q t} \]

\( \mu_{ij}(t) \)  Expected value of \( \mu_{ij}^o(t) \); that is,
\[ \mu_{ij}(t) = \lim_{t \to \infty} \frac{1}{t} \sum_{i=1}^{L} \mu_{ij}^o(t) \]

\( \mu_{ij'}(t) \)  Same as \( \mu_{ij}(t) \) except for all isotopes:
\[ \mu_{ij'}(t) = \sum_{\xi=1}^{L} \mu_{ij}^\xi(t) \]

\( \mu_{ij}(t, T) \)  Expected counts per minute for a period \( T \) from isotope \( L \) of sample \( i \) at age \( t \), corrected for background and deadtime of the detector.
\[ \mu_{ij}(t, T) = \sum_{\xi=1}^{L} \mu_{ij}^\xi(t, T) \]

\( \mu_{ij}(t, T) \)  Same as \( \mu_{ij}(t, T) \) except for all isotopes in the sample; that is,
\[ \mu_{ij}(t, T) = \sum_{\xi=1}^{L} \mu_{ij}^\xi(t, T) \]

\( \mu_{ij}(t, T) \)  The population mean of \( \mu_{ij}(t, T) \); that is,
\[ \mu_{ij}(t, T) = \lim_{t \to \infty} \frac{1}{t} \sum_{i=1}^{L} \mu_{ij}(t, T) \]

\( \mu_{ij}(t, T) \)  The population mean of \( \mu_{ij}(t, T) \); that is,
\[ \mu_{ij}(t, T) = \sum_{\xi=1}^{L} \mu_{ij}(t, T) \]
The population mean of \( \mu_j(t,T) \); that is, \( \bar{\mu}_j(t,T) = \lim_{N \to \infty} \frac{1}{N} \sum_{i=1}^{N} \mu_i(t,T) \)

The population mean of \( \nu_j(t,T) \); that is, \( \bar{\nu}_j(t,T) = \lim_{N \to \infty} \frac{1}{N} \sum_{i=1}^{N} \nu_i(t,T) \)

The population mean of \( \mu(t,T) \); that is, \( \bar{\mu}(t,T) = \lim_{N \to \infty} \frac{1}{N} \sum_{i=1}^{N} \mu_i(t,T) \)

The population mean of \( \nu(t,T) \); that is, \( \bar{\nu}(t,T) = \lim_{N \to \infty} \frac{1}{N} \sum_{i=1}^{N} \nu_i(t,T) \)

The population mean of \( \lambda(t,T) \); that is, \( \bar{\lambda}(t,T) = \lim_{N \to \infty} \frac{1}{N} \sum_{i=1}^{N} \lambda_i(t,T) \)

The population mean of \( \beta(t,T) \); that is, \( \bar{\beta}(t,T) = \lim_{N \to \infty} \frac{1}{N} \sum_{i=1}^{N} \beta_i(t,T) \)

The population mean of \( \kappa(t,T) \); that is, \( \bar{\kappa}(t,T) = \lim_{N \to \infty} \frac{1}{N} \sum_{i=1}^{N} \kappa_i(t,T) \)

The population mean of \( \theta(t,T) \); that is, \( \bar{\theta}(t,T) = \lim_{N \to \infty} \frac{1}{N} \sum_{i=1}^{N} \theta_i(t,T) \)

Continued
GLOSSARY OF SYMBOLS (Continued)

\( \tilde{\sigma}_{mk} \) An estimate of \( \sigma_m \) on the \( k \)th iteration of the least-squares process. \( \tilde{\sigma}_{m0} \) is a historical estimate required for starting the iterated least-squares process.

\( \tilde{\sigma}_{m0} \) The appropriate historic estimate of the proportional mechanical error based on \( f_o \) degrees of freedom.

\( \sigma_{m'} \) The current proportional mechanical error based on \( \tilde{\sigma}_{m0} \).

\( \tilde{\sigma}_{mk} \) An estimate of \( \sigma_{m'} \) on the \( k \)th iteration.

\( \sigma_s \) Proportional sampling error.

\( \tilde{\sigma}_{sk} \) An estimate of \( \sigma_s \) on the \( k \)th iteration of the least-squares process.

\( \tilde{\sigma}_{s0} \) The appropriate historic estimate of the proportional sampling error based on \( d \) degrees of freedom.

\( \sigma_{s'} \) The current proportional sampling error based on \( \tilde{\sigma}_{s0} \).

\( \tilde{\sigma}_{sk} \) An estimate of \( \sigma_{s'} \) on the \( k \)th iteration.

\( \tilde{\sigma}_{\mu^2} \) Total estimated variance of \( \mu \).

\( \tilde{\sigma}_{\mu^2} = (\tilde{\sigma}_{\mu^2})_m + (\tilde{\sigma}_{\mu^2})_p + (\tilde{\sigma}_{\mu^2})_s \)

\( (\tilde{\sigma}_{\mu^2})_m \) Estimated variance of \( \mu \) due to mechanical effects.

\( (\tilde{\sigma}_{\mu^2})_p \) Estimated variance of \( \mu \) due to Poisson causes.

\( (\tilde{\sigma}_{\mu^2})_s \) Estimated variance of \( \mu \) due to sampling effects.

\( \tau \) Deadtime of the detector in minutes.

When used as a superscript, a quantity essentially at time \( t \):

\[ \bar{\mu}_{21}^t = E \left[ \lim_{T \to 0} \frac{1}{T} \int_0^T du \right] \]

When used as a subscript, for that particular subset usage is usually different.

A bar placed over any symbol indicates an arithmetic average of the number of items; for example:

\[ \overline{\alpha} = \frac{1}{n} \sum_{i=1}^{n} \alpha_i \]

A tilde placed over any symbol is an estimate of the true value. 

\( \tilde{\mu}_{21} \) is an estimate of \( \mu_{21} \).
An estimate of $\sigma_m$ on the $k^{th}$ iteration of the least-squares process, $\sigma_{mo}$ is a historical estimate required for starting the iterated least-squares process. The appropriate historic estimate of the proportional mechanical error based on $\sigma_m$. The current proportional mechanical error based on $\sigma_{mo}$. An estimate of $\sigma_m$ on the $k^{th}$ iteration. Proportional sampling error. An estimate of $\sigma_p$ on the $k^{th}$ iteration of the least-squares process. The appropriate historic estimate of the proportional sampling error based on $\sigma_{po}$. The current proportional sampling error based on $\sigma_{po}$. An estimate of $\sigma_p$ on the $k^{th}$ iteration. Total estimated variance of $\mu$. $\bar{\sigma}^2_{\mu} = (\bar{\sigma}^2_{\mu_m})_m + (\bar{\sigma}^2_{\mu_p})_p + (\bar{\sigma}^2_{\mu_s})_s$ Estimated variance of $\mu$ due to mechanical effects. Estimated variance of $\mu$ due to Poisson causes. Estimated variance of $\mu$ due to sampling effects. Deadtime of the detector in minutes. When used as a superscript, it indicates the quantity essentially at time $t = 0$; for example, $\mu^o_{ Edi} = \lim_{T \to 0}\frac{1}{T}\int_0^T v(t, T) \, dt$ When used as a subscript, it acts as a placeholder for that particular subscript. Interpretation of usage is usually different in each case. A bar placed over any symbol indicates an arithmetic average or the mean of a finite number of items; for example, $\bar{Y} = \frac{1}{n} \sum_{h=1}^{n} Y_h$ A tilde placed over any quantity indicates it is an estimate of the true quantity; for example, $\bar{\mu}^o_{ Edi}$ is an estimate of $\mu^o_{ Edi}$. 81
In work related to radiation shielding, the use of radioisotope techniques, and activation analysis, an experimenter must often analyze counting data where counts are caused by the natural background and by the decay of more than one radioisotope. In this report a procedure is developed for estimating the strength of each isotope at different times from several decaying radioactive samples of a single multiple-isotope source. In addition, the procedure provides a method for partitioning the imprecision of estimating the strengths into three principal causes: Poisson variation, sampling error, and residual error (called mechanical error).

An operational FORTRAN II-D computer program, SAND, implements the procedure. The procedure and program were tested by using fictitious data with known properties as inputs. The results of the simulation were in reasonable agreement with the theoretical values.
ESTIMATING STRENGTHS OF INDIVIDUAL RADIOISOTOPES IN A MULTIPLE-ISOTOPE SOURCE

The Imprecision of the Estimates is Partitioned Into Poisson, Sampling, and Mechanical Variations

In work related to radiation shielding, the use of radioisotope techniques, and activation analysis, an experimenter must often analyze counting data where counts are caused by the natural background and by the decay of more than one radioisotope. In this report a procedure is developed for estimating the strength of each isotope at different times from several decaying radioactive samples of a single multiple-isotope source. In addition, the procedure provides a method for placing confidence limits on the strengths and a method for partitioning the imprecision of estimating the strengths into three principal causes: Poisson variation, sampling error, and residual error (called mechanical error).

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