



**DISCRETE COUNTING OF SHORT LIVED ISOTOPES WITH LOW
BACKGROUND DETECTORS**

THESIS

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THESIS

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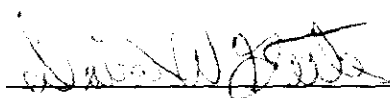
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Abstract

Radiation counting experiments are usually used to quantify activities of materials that are long-lived with respect to the count durations. Counts obtained include detections of background radiation. The usual statistical analysis (HPS 13.31) for estimating the activity and its uncertainty (mean and standard deviation) seriously overestimate the uncertainty when the activity and background are very low. Strom and McClellan [2001] reviewed this difficulty. We consider the case of short-lived nuclides for which the objective is to quantify the number of atoms, n , that were present in a sample when it was drawn, rather than the activity, which is changing during the measurement. Mathews and Gerts [2008] analyzed this case and developed formulas from which the probability distribution, $P(n \mid \text{counts, experiment parameters, background information})$, can be computed. They used this to develop experiment design processes that minimize the smallest detectable quantity of material, thus maximizing sensitivity for the detection problem. Here, their distribution is used to establish the mean quantity, $\langle n \rangle$, and the equal-tails confidence interval (CI) for any specified confidence level, in order to determine the precision of the measurement, defined as the width of CI divided by the mean. An experiment *quantifies* n if the

precision is better than a specified precision tolerance. A quantity is *quantifiable* (by a specified experiment design) if the expectation that a measurement would quantify it exceeds a specified expectation tolerance. (These definitions are intended to be analogues of *quantification limit* for activity in the long-lived case.) Methods and software are developed for determining the minimum quantifiable quantity (MQQ) for a given experiment design, and for adjusting the count duration to achieve the lowest MQQ, hence maximizing the sensitivity for the quantification problem. Plots of MQQ vs. count duration support tradeoff decisions. Monte Carlo methods have been used to validate this analysis and experiment design software.

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David E. Mather

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List of Acronyms

ANSI	American National Standards Institute
cdf	Cumulative Density Function (continuous)
	Cumulative Distribution Function (discrete)
EDC	Experiment Design Code
ERW	Expected Relative Width
HPS	Health Physics Society
LMQQ	Lowest Minimum Quantifiable Quantity
MQQ	Minimum Quantifiable Quantity
pdf	Probability Density Function (continuous)
	Probability Distribution Function (discrete)
PoQ	Probability of Quantification

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DISCRETE COUNTING OF SHORT LIVED ISOTOPES WITH LOW BACKGROUND DETECTORS

1. Introduction

We consider the problem of low-level radiation counting with very low backgrounds. The objective is to develop statistical analyses and experiment design methodologies that maximize sensitivity of such experiments. Mathews and Gerts (2008) treat the detection problem: detecting the presence of any quantity of a nuclide that is short-lived compared to the count duration. Their derivation of the various conditional probabilities that are needed here is reviewed in sections 2.1 through 2.4 so that this document may stand alone. They developed experiment design methods that provide the count duration (for an otherwise fully-specified counting measurement) that maximizes sensitivity and the critical number of counts needed to declare detection at a specified confidence level. Here, we extend their work to treat the quantification problem and develop experiment design software that finds the count duration that minimizes the quantity required to ensure a sufficiently precise measurement or

that provides a tradeoff curve of minimum quantifiable quantity vs. count duration for an otherwise fully-specified counting measurement.

1.1 Motivation

Accurate measurement of small quantities of radioisotopes is a necessity for several Department of Defense applications. As progress is made in developing detectors capable of counting decays while registering very little background, the minimum radioisotope population required to quantify the measurement within a reasonable window of uncertainty has decreased. However, contemporary analysis methods rely on normal distributions for analyzing these measurements, and have limited sensitivity in situations where the total number of counts does not meet the Gaussian assumptions (Mathews, 2007). Contemporary methods also fail to apply when the duration of the measurement is on the order of magnitude or greater than the half life of the isotope being measured (Knoll, 2004). Under these conditions, it is possible that the Gaussian assumptions will result in reporting unphysical negative sample sizes. Preliminary indications from this work suggest that these restrictions decrease detector sensitivity by an order of magnitude. In order to best use low background detectors on short lived isotopes, modern data analysis and acquisition methodology must incorporate exact statistics rather than Gaussian assumptions.

1.2 Theory

The standard methodology and analysis for the measurement of a radioactive source was first introduced by Lloyd Currie in 1968. Prior to his paper, there was enough inconsistency in the definition of “detection limit” to cause a large amount of disagreement amongst scientists. Currie described an analysis method assuming that radioactive decay and background were governed by Poisson counting statistics with normally-distributed error. The method involved pairing sample measurements with a “blank” background measurement and subtracting out the background.

The method of pairing blank and sample measurements is still in practice today. Under normal circumstances, the radioisotope being measured is of significant enough quantity and the measurement time short enough compared to the half life of the isotope that the activity can be assumed to be constant and the expected foreground distribution to be an adequate approximation to the normal (Gaussian) distribution. For example, the analysis presented in the American National Standards Institute standard in conjunction with the Health Physics Society, HPS 13.30 (Heid 1996) assumes a Gaussian estimate to both the foreground and background distribution by pairing a sample count with a blank count and subtracting the blank. Given this method, it is possible for zero or even negative counts to satisfy the decision criterion when a small number of atoms exist in the sample. This result is unrealistic because it is impossible to

have a sample containing a negative number of isotopes. The ANSI method also does not sufficiently account for uncertainty in the measurement of the mean background count rate in the case of very low background. This is because a single short measurement in low background results in a broad, normal distribution of the count rate.

Recently, this standard has been challenged in the case of low level counting situations (Strom and MacLellan, 2001) in favor of Poisson distributed foreground and background. This reduces the possibility of achieving negative counts, but it maintains the assumption that the counting time is at least an order of magnitude less than the half life of the isotope being measured. To remove this restriction, this paper demonstrates the use of the binomial distribution to construct the likelihood function for the foreground counts.

The background count is also a source of uncertainty. Although the Poisson distribution may be assumed, the mean count rate will be estimated based on a single or series of discrete background counts and is thus subject to its own variation that must be addressed. The Poisson distribution is the probability of registering b counts given a known mean background rate. These conditional probabilities may be reversed by applying Bayes' Rule (Black and Thompson, 2001). In this manner a function for the actual background rate can be generated by taking a measurement of the background.

1.3 Classic Experiment Design

Consider a sample of a short lived radioisotope with an initial unknown quantity of n atoms and a known decay constant λ obtained at time t_0 . A delay t_d occurs between the collection and the start of measurement. The measurement begins at time t_1 and ends at time t_2 for a total measurement time of t_c . The detector records c total counts that contain an unknown number of k foreground and b background counts. The mean background counts μ in t_c is inferred from a background measurement of duration t_B containing B counts.

The number of counts will give rise to a probability distribution function for n . A confidence interval will be constructed on the cumulative density function for n , as shown in Figure 1.

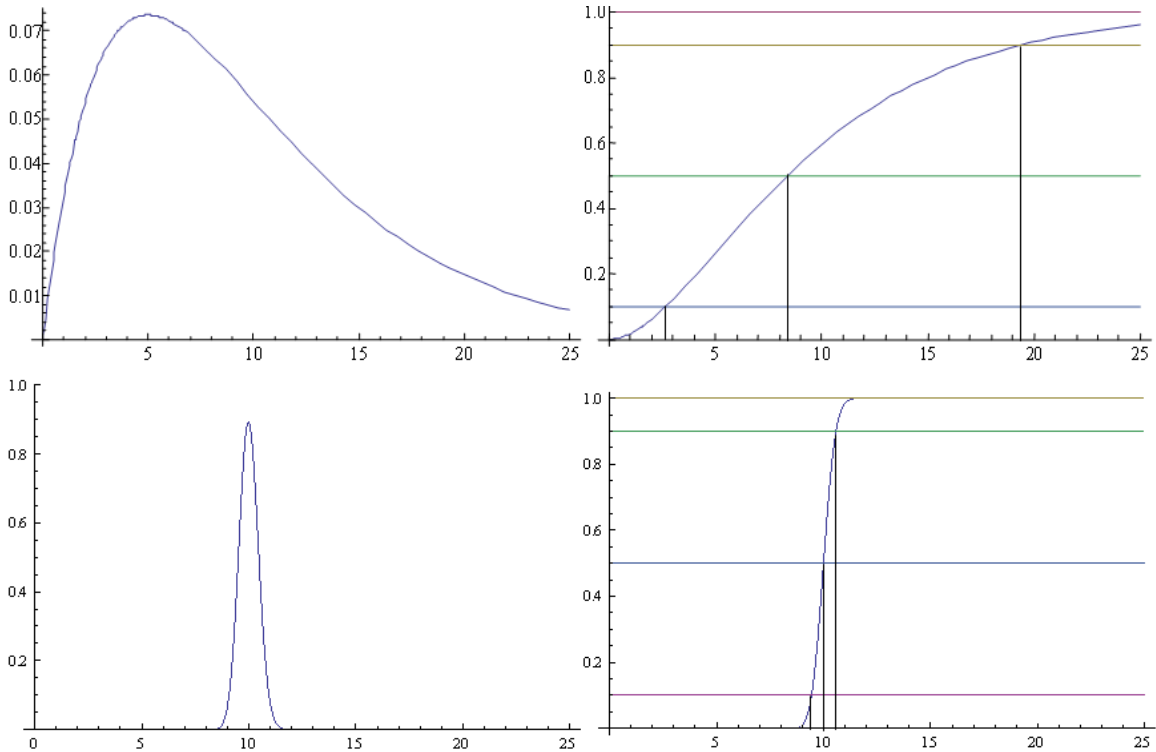


Figure 1 - Two PDFs and CDFs with 80% confidence intervals and medians

A measurement will be considered quantified if the distribution satisfies narrowness requirements as defined by the relative width: the ratio of the width of the confidence interval with the expectation value for n . This is a departure from the practice presented in HPS 13.30 which simply builds a confidence interval from Gaussian assumptions. For any experimental scenario, there is a lowest value for n that can be expected to be quantified in a desirable fraction of attempts. This value is the Minimum Quantifiable Quantity or MQQ. Altering the conditions of the experiment will affect the MQQ. The lowest possible value of the MQQ under conditions optimized for sensitivity is the Lowest Minimum Quantifiable Quantity, or LMQQ.

1.4 Statement of the Problem

In this thesis, I describe a method for the analysis of experimental results to construct a probability distribution function for n and a confidence interval to describe the narrowness of the function. I also describe a method for calculating the expectation of quantification given n and the known experimental conditions. Furthermore, I describe software that uses these methods to find the MQQ and calculate it as a function of the count duration. This data is used to find the LMQQ as well as to describe tradeoff choices in experiment design. Finally, I describe software used to simulate the experiment as well as verify and analyze

the discrete method. This method is general and can be applied to any short-lived radioactive isotope.

1.5 Scope

This research investigated the application and performance of an experiment optimization method using discrete statistics. The method is explored for strengths and weaknesses and is compared to the ANSI standard HPS 13-30.

1.6 Assumptions and Limitations

The computational method described assumes that the only sources of counts include the sample material, where the number of atoms that decay is governed by the Binomial distribution with each atom being treated as a Bernoulli trial, and a constant-mean Poisson distributed background. These two sources are statistically independent because there is no causal relationship between the two sources, and the mean count rate is assumed to be so low that dead time is negligible in the detector. This project is limited to the calculation and modeling of the counts registered from a small quantity of a short lived radioisotope being measured in a low background detector.

1.7 Approach

This thesis demonstrates the mathematical basis for the statistical analysis of the problem in section 1.3. This methodology is programmed into a numerical method using the FORTRAN language. This method is used both to analyze data from experiments and to evaluate and optimize experiment design for maximum sensitivity. The code is verified with a separate verification tool. This tool is then used to explore the discrete method by simulating the experiment in a range of scenarios designed to test the failure modes of the application. These data are used to describe a technique for setting up an experiment and analyzing data from a low background detector and to compare its efficiency and accuracy to current classical methods.

2. Governing Equations and Statistical Relations

Accounting for each source of measurement event is crucial to effectively modeling the discrete nature of recorded counts in a small sample and low background scenario. In this investigation, only the foreground measurement of actual decays, and background from random events unrelated to the experiment are included. Given the very low number of counts expected in the experiment, it is also reasonable to assume that any dead time in the detector will have a negligible impact on the outcome. Thus, dead time can be ignored, allowing the foreground and background counts to be treated as statistically independent and the inputs used to model those events to be treated independently.

The following derivation of a pdf for n given the experimental data is paraphrased from Mathews (2007).

2.1 Binomial Distributed Foreground

The probability of collecting foreground counts is the product of the probability that an atom will decay during measurement and the likelihood that the detector will actually register that decay. The probability that an atom of a given species will decay is given by equation (1)

$$p_{decay}(t) = 1 - e^{-\lambda t}, \quad (1)$$

where λ is the species' decay constant and t is the amount of time given for decay. To account for a delay between the collection or generation of the sample and the start of measurement, the probability that an atom will decay during the counting must include not just the probability that it will decay during the measurement time, but also the probability that it will not decay during the delay:

$$p_{decay}(t_d, t_c) = e^{-\lambda t_d} (1 - e^{-\lambda t_c}). \quad (2)$$

The probability that a foreground count will actually be registered is the product of the probability of decay and the efficiency of the detector. The efficiency, ϵ , of a detector is the fraction of decays in the sample that are detected and can be greatly affected by the geometry and scale of the detector as well as being influenced by environmental factors such as temperature. For the purposes of this investigation, efficiency will be fixed and is assumed to be known with sufficient precision to neglect uncertainty, setting the probability of detection at

$$p_{detect} = p_{decay} \epsilon. \quad (3)$$

Because each decay and background count are considered separate and independent, whether an atom decays and is counted can be treated as a Bernoulli trial and the number of counts k registered from n atoms is governed by the binomial distribution

$$P(k | n, p_{\text{detect}}) = \binom{n}{k} p_{\text{detect}}^k (1 - p_{\text{detect}})^{n-k} . \quad (4)$$

This formula is ideal for analysis of small samples because the probability distribution is restricted to foreground decays with a value of zero to n , whereas the Poisson approximation is biased high in the case of low values for n and the Gaussian approximation allows for unrealistic possibilities for negative sample size, especially given long counting times.

2.2 Poisson Distributed Background

Background counts are defined as rare random events unrelated to the sample that cause counts in the detector. It is impossible to completely eliminate the background; however methods such as measurements in highly-shielded clean rooms and coincidence counting have succeeded in significantly decreasing background. Background is also assumed to be purely random and not driven by constant sources elsewhere in the laboratory. Given these assumptions, background counts can be characterized by the Poisson process. The distribution of the number of background counts b measured in a set time window with an expectation value μ is given by

$$P(b | \mu) = \frac{\mu^b e^{-\mu}}{b!} . \quad (5)$$

Because μ cannot be known, it must be measured experimentally. To do that, a background measurement must be run separate from the sample

measurement. In order to gain as much information as possible about μ , it is preferable to make the background measurement much longer than the sample measurement. In reality, a background count may be affected by the time of day, time of year, and various activities such as other experiments in the vicinity of the detector. For the sake of simplicity, the mean background rate is treated as stable. Given a stable mean background rate, the preferred blank measurement time is long compared to the sample measurement. Increasing the blank measurement time increases the amount of information the researcher has regarding the mean background count rate. This narrows the distribution of probable values of μ .

The functional variation of μ is calculated from the background data. The value a is defined to be the ratio between the foreground and background measurement times, where

$$a = \frac{t_c}{t_B} . \tag{6}$$

Uncertainty in a can be neglected by assuming that the count times are precisely known. Because the background is governed by the Poisson process, the expected mean counts in the sample and background counts are related by the same time ratio

$$\tilde{\mu} = \mu / a , \tag{7}$$

where $\tilde{\mu}$ is the expectation value for the background count. The probability density function governing $\tilde{\mu}$, given a and the number of background counts B , is transformed to μ by

$$f(\mu | a, B) d\mu = f(\tilde{\mu} | a, B) d\tilde{\mu}, \quad (8)$$

where

$$\frac{d\tilde{\mu}}{d\mu} = \frac{1}{a}. \quad (9)$$

The Poisson distribution only describes the probability of B counts given the expectation value, not the other way around. Bayes' Theorem can be used to swap the conditional probabilities.

$$f(\tilde{\mu} | B) = \frac{P_{prior}(B)P(B | \tilde{\mu})}{\int_0^{\infty} P_{prior}(B)P(B | \tilde{\mu}') d\tilde{\mu}'} \quad (10)$$

In order to avoid bias in the calculation, the $P_{prior}(B)$ distribution is defined to be flat and uninformative (Mathews, 2007). This means that the constant probabilities for the background priors cancel. The integral of the function remaining in the denominator from zero to infinity equals 1, yielding the equation for $\tilde{\mu}$:

$$f(\tilde{\mu} | a, B) = \frac{\tilde{\mu}^B e^{-\tilde{\mu}}}{B!}. \quad (11)$$

Transforming $\tilde{\mu}$ to μ , by using equation (7) yields the conditional probability density function for μ :

$$f(\mu | a, B) = \frac{1}{a} \frac{\mu^B e^{-\mu/a}}{B!} = \frac{\mu^B e^{-\mu/a}}{a^{B+1} B!}. \quad (12)$$

The probability distribution function for recording b background counts during the sample measurement given background data a and B is the product of the probability of b counts given expectation value μ and the functional distribution of μ given the background data integrated over all possible values of μ :

$$P(b | a, B) = \int_0^\infty P(b | \mu) f(\mu | a, B) d\mu = \left(\frac{a}{1+a} \right)^b \left(\frac{1}{1+a} \right)^{1+B} \binom{B+b}{b}. \quad (13)$$

This distribution, based on Poisson statistics, is discrete while allowing for a real mean count rate with uncertainty depending on how much information has been gathered on the background. Furthermore, it has no negative tails that would be included in a normal approximation to the background.

2.3 Total Counts

The analysis thus far is capable of calculating the probability of recording c counts given n atoms. All other experimental constants such as detector efficiency or delay time will be treated as implicit unless explicitly required by the analysis. Thus, the probability of recording c counts given n is

$$P(c | n) = \sum_{k=0}^c P(k | n, p_{\text{detect}}) P(b = c - k | a, B). \quad (14)$$

However, the purpose of the experiment is to calculate a distribution function for the number of atoms in the sample at $t=0$ given that c counts have been measured. Again, by using Bayes' Theorem and an uninformative prior regarding the distribution of n , the conditional probabilities for $P(c | n)$ can be swapped. Holding all other variables constant, the probability of having n atoms given c counts is given by

$$P(n | c) = \frac{P(c | n)}{\sum_{n'=0}^{\infty} P(c | n')} . \quad (15)$$

The sum in the denominator cannot be carried out to infinity, however, it can be carried to sufficiently high values for n' that the value of the sum will fall within a reasonable tolerance of the true answer.

2.4 Acceptable Uncertainty

In order to successfully declare a measurement as quantified, the researcher must construct a confidence interval over the pdf for n with a confidence level $1-\alpha_c$, where α_c is defined by the researcher. It is possible to construct confidence bounds in place of intervals should a lower or upper limit be desired, but this research is limited to intervals. This confidence interval can be constructed such that it is symmetric about the point estimator in n or that it has equal probability tails. In this case, the equal probability tails is preferred

because it is possible to construct a symmetric confidence interval that contains negative numbers.

The width of this confidence interval must be compared to a point estimator that is chosen between the mean, the median and the mode. In this case, the mode is rejected because the distributions are not always symmetric. The median is the preferred point estimator because it can be calculated at the same time as the equal tailed probability distribution and, because the distributions are well behaved, is very similar to the mean. However, in this research the mean is the point estimator used because at the time of its initial coding, it worked well in the construction of the program regardless of its computational cost. It is calculated using a trimmed mean where computation is ceased when a satisfactory fraction of the distribution function is covered.

Quantification can be declared once the ratio of the width of the confidence interval to the value of the point estimator is less than θ , a value defined by the researcher. In order for an initial number of atoms to be quantifiable, then there must be an expectation that quantification will be declared $1-\alpha_q$ of trials, where α_q is defined by the researcher.

2.5 Optimizing Δt_c for Greatest Sensitivity

For each experiment, there exists a value for n that is the Minimum Quantifiable Quantity. Optimizing for greatest sensitivity is then a matter of adjusting parameters such that the defined confidence levels are met exactly or

exceeded for the lowest possible MQQ. The value for n that corresponds to the MQQ at maximum sensitivity is the Lowest Minimum Quantifiable Quantity (LMQQ).

While factors such as the efficiency of the detector and the mean background level are important, the only parameter that the experimenter can reasonably change is the count duration. The quality of the results of a low count experiment changes as the time window changes. This is because time has differing impacts on the number of foreground and background counts as expressed in equation (14). The contribution by the foreground from an increase in count duration in the first few half lives is great because the probability of an atom decaying increases rapidly during that time. However, the marginal utility of additional counting time decreases as the atoms in the sample decay away. In contrast, the background rate stays constant in time and the contribution of the background to the total counts will continue to increase linearly with time. This means that for each value of n , there is an optimal measurement time.

3. Computer Code Implementation

The primary function of the experiment design code is to optimize the detection sensitivity for the Lowest Minimum Quantifiable Quantity and to calculate the ideal time of measurement given the defined experimental constraints. It also contains a function intended for data analysis that outputs a probability distribution function for values of n given experimental data from a measurement. Finally, it is designed to output the minimum quantifiable quantity as a function of the conditions of the experiment.

3.1 Implementation of Discrete Statistics

The implementation of discrete statistics can be straightforward or problematic depending on which of the calculations the code is executing. The data analysis routine is a simple calculation to determine a probability distribution function for n given c and the other known data. This analysis benefits greatly from the discrete nature of the exact statistics by not being restricted to the assumptions made by a Gaussian calculation. The second calculation, finding the LMQQ and optimizing the counting time, is made much more complicated by the implementation of discrete statistics. These statistics introduce discontinuities in the results of the calculation brought on by the integer nature of the non-continuous functions.

In this investigation, the primary source of discontinuity comes from the method used to define a confidence interval for the purposes of calculating the Probability of Quantification (PoQ) of the measured data. The PoQ is a value determined from a known n and calculated over all c in the equation:

$$P_{\text{Quantify}}(n) = \sum_{c=0}^{\infty} P(c|n)q(c), \quad (16)$$

where

$$q(c) = \begin{cases} 1 & c \Rightarrow \text{quantification} \\ 0 & \text{else} \end{cases}. \quad (17)$$

Calculating the probability of quantification for the MQQ may appear to be the simplest method when attempting to determine optimal measurement time for a high sensitivity count. However, because the function is not continuous both in time and counts, it becomes broad, jumpy, and is difficult to maximize in conditions at or near the LMQQ due to the combined fact that the PoQ always has a positive slope with respect to counting time (with exception to the location of the jumps) and that the location of the jumps is dependant on the method used to define the confidence interval. The code avoids these difficulties by interpolating between values of n' on its probability density function and measuring the relative width of the confidence interval in terms of real numbers.

Figure 1 shows the probability of quantifying a measurement given known information about the experiment based on a known initial quantity of atoms. Each of the family of “lines” displayed corresponds to the probability of

achieving at least a minimum number of counts that will be quantified in the time given. Depending on the method of interpolation, counts above this minimum are nearly always quantified. All of these lines increase and decrease in probability. It is possible for these lines to stay at or near 1 for a significant number of half lives before their probability of being accepted drops off. A more detailed discussion of this can be found in Chapter 4.

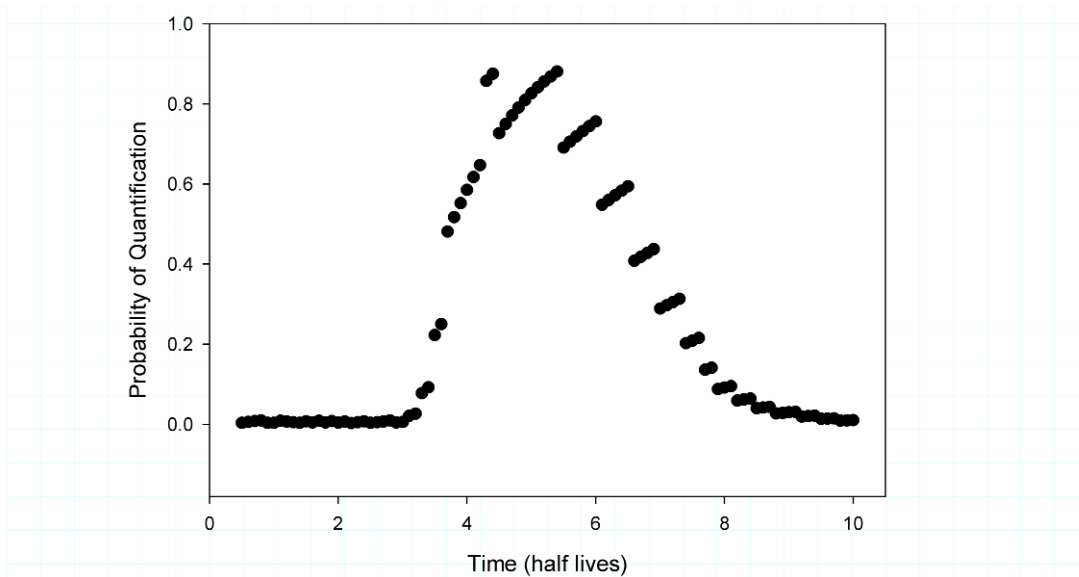


Figure 2 - Probability of Quantification at LMQQ¹

$$\begin{array}{llll} \epsilon = 1 & t_B = 20 & \alpha_c = .2 & \theta = .2 \\ t_d = 0 & B = 1 & \alpha_w = .2 & \end{array}$$

¹ All data presented in Chapter 3 are calculated using the same experimental variables.

3.2 Implementation of Non-Discrete Measure of Confidence

In order to alleviate the problems introduced by the development of confidence intervals on the discrete probability distribution function for $P(n|c)$, the code interpolates to determine a pseudo-interval that is continuous in n . The expectation value of a pdf is a real number, and once an interpolation method is used, the width of the confidence interval itself is also a real number. The ratio of the two is the relative width w_r of the interval where

$$w_r(c) = \frac{CI_{hi} - CI_{low}}{\langle P(n|c) \rangle}. \quad (18)$$

Because any calculated width varies as a function of c , it can be treated in the same way as the probability of acceptance, yielding the Expected Relative Width (ERW);

$$ERW(n, data) = \sum_{c=0}^{\infty} P(c|n) w_r(c) \quad (19)$$

which is a function of n and requiring the other data from the experiment, but having the advantage of being much better behaved than its discrete counterpart, thanks to the fact that it experiences no discontinuities.

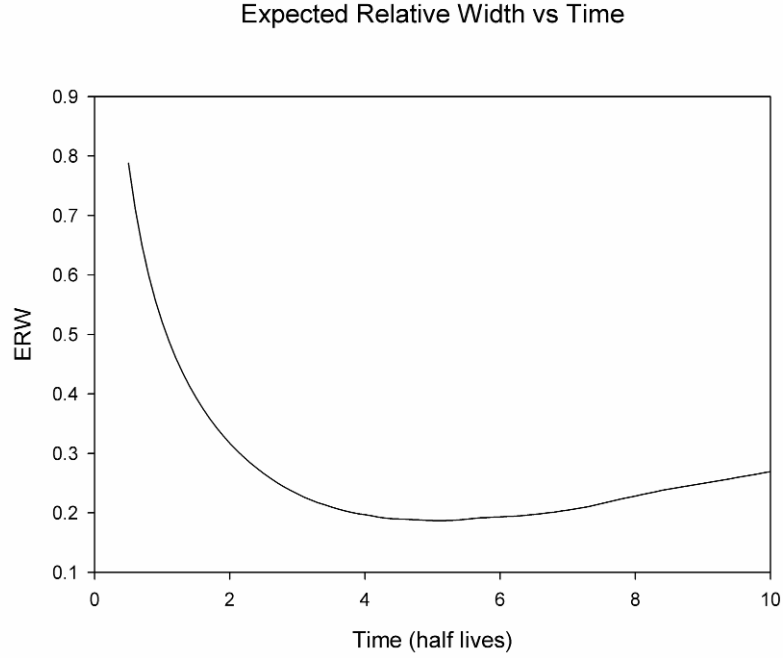


Figure 3 - Expected Relative Width at LMQQ

The interpolation method used to define the confidence interval must be chosen with some care. The cumulative density function is a monotonic, non-decreasing function, and any data calculated by an interpolation scheme should reflect that. The goal in selecting an interpolation method is to yield a continuous function for $w(n|c, t_c)$ that is also continuous in the first derivative with one local minimum to promote ease in minimizing it. Without interpolation, any function for relative width will be discontinuous. Linear interpolation yields a function for the expected relative width that is continuous, but not in the first derivative, presenting a minimization problem with multiple local minima and an increased degree of uncertainty in the final result. Cubic

spline interpolation is continuous in both the first and second derivative but risks losing the monotonic nature of the cumulative distribution function. However, an algorithm presented by Fritsch and Carlson (1980) successfully uses the cubic basis functions in a monotone piecewise interpolation that is continuous in the first derivative and generates a function for the expected relative width that contains only one local minimum. This is the default method that is used by the program.

To interpolate using the monotone piecewise method, the algorithm first inputs the two points of interest and their adjacent outside points. It defines the two variables α and β as the ratios of the endpoint derivatives to the slope of the secant line, Δk . Fritsch and Carlson determined the region of monotonicity to be the shaded region in Figure 4.

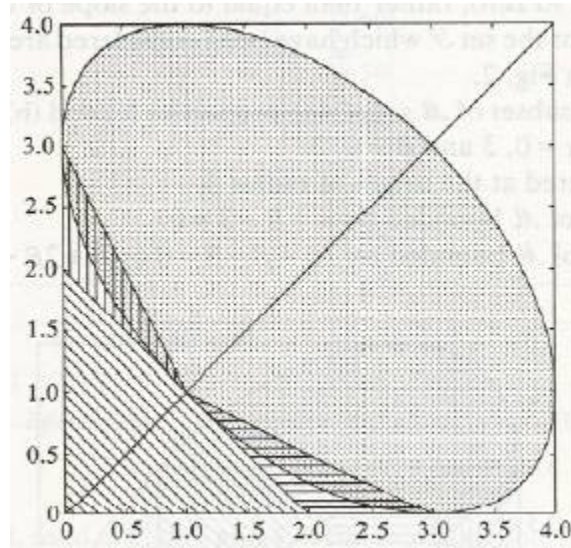


Figure 4 - The monotonicity region from Fritsch and Carlson.

If α and β fall outside the monotone region, then a correction value τ is defined for each point,

$$\tau_k = 3(\alpha_k^2 + \beta_k^2)^{-1/2}, \quad (20)$$

and their derivatives are adjusted accordingly:

$$m_k = \tau_k \alpha_k \Delta k. \quad (21)$$

.It does not yield a perfectly well behaved function, which would preferably be continuous in each derivative, but the results are the most stable of the methods described above.

Figures 5 and 6 illustrate the effects that the different interpolations methods have on the probability of quantification and expected relative width given the same experimental data. In nearly all situations, the PoQ is higher with a narrower ERW. Figure 5 also demonstrates the desirability of the cubic monotone interpolation over the linear interpolation when it comes to searching for an absolute minimum for the ERW.

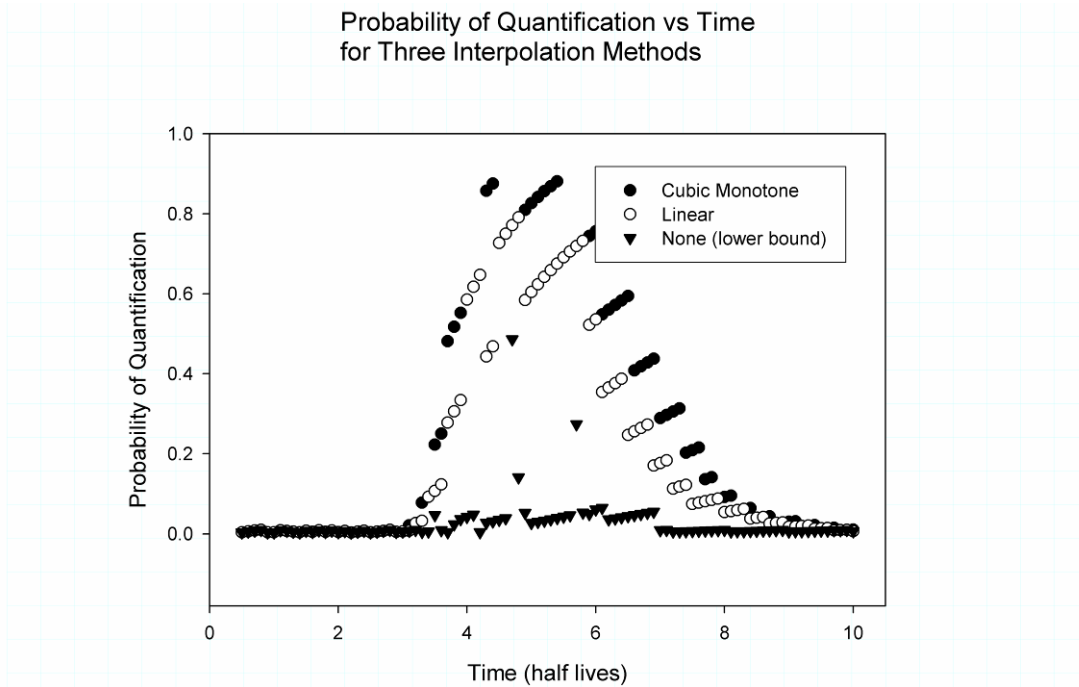


Figure 5 - Effect of interpolation on Probability of Quantification

Expected Relative Width vs Time
for Three Interpolation Methods

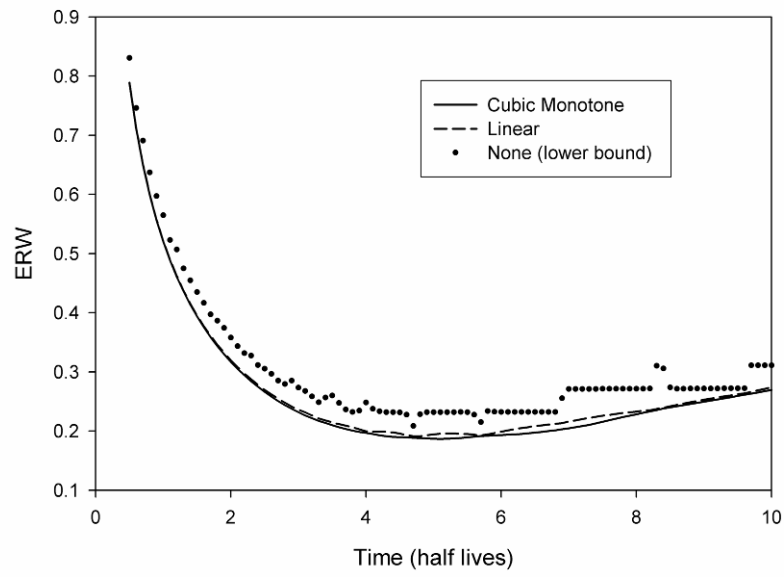


Figure 6 - Effect of Interpolation on Expected Relative Width

Expected Relative Width vs Time
for Three Interpolation Methods

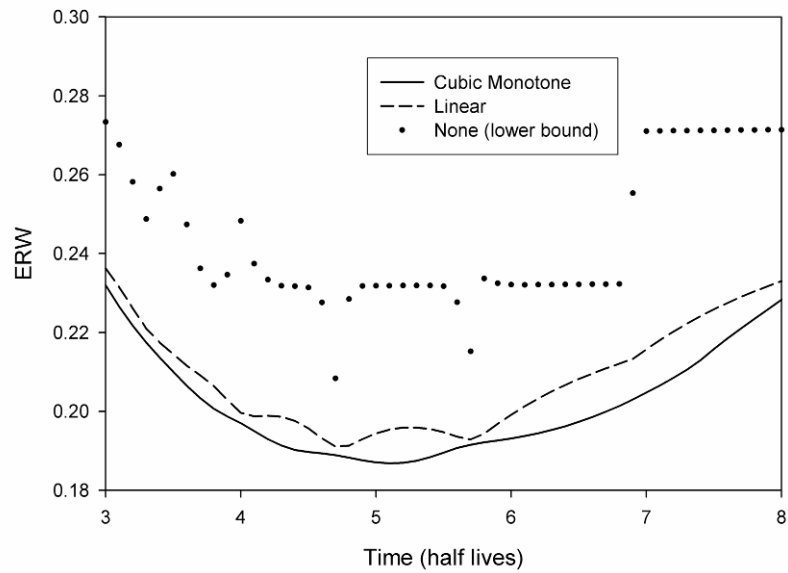


Figure 7 - Effect of interpolation on ERW near lower bound

3.3 Algorithm of the LMQQ Code

This Experiment Design Code is designed to run on a set of default parameters for decision criteria and interpolation, but also allows the user to stipulate specific methods. The algorithms for the three primary functions differ slightly from one another. The experiment optimization code reads in a user generated input file. In order to avoid calculating individual choose results for binomial foreground probabilities, the code initializes a 1000 by 1000 array of choose results based on the following relation:

$$\binom{n}{k} = \binom{n}{k-1} \frac{n-k+1}{k}. \quad (22)$$

The code then enters a loop to determine the LMQQ. It begins by taking steps in time by half lives starting with 1. For each step it initializes the probability functions for foreground and background detection. It then calculates the Expected Relative Widths with respect to n starting from $n=1$ until it finds a corresponding ERW that is less than or equal to θ . Each time step yields a different value for n . Once the lowest n is determined, the code uses the bisection method in time to find the optimal counting time for that value of n . It then calculates the Probability of Acceptance given n and t_c . If the PoQ is greater than $1-\alpha_a$ then it checks the ERW and PoQ at optimal time for $n-1$, otherwise, it begins increasing n until a satisfactory PoQ is achieved.

The other two routines are based on functionality already built into the optimization code. The Minimum Quantifiable Quantity function works just like the optimization function except that time is no longer variable. The data analysis routine uses the formulas presented in Chapter 2. The code inputs the experimentally known variables and outputs a probability distribution function for n , as well as whether or not the function is sufficient to declare a measurement. These routines are the source of data for the experiment simulation and verification routine.

3.4 Generation of Binomial and Poisson Distributed Random Data

To test this program it is useful to be able to generate realistic data from known experimental conditions to input into the analysis code. The number of counts during sample measurement that can be attributed to foreground must follow a binomial distribution. The data for the background count and the background counts registered during sample measurement must follow a Poisson distribution. These numbers can then be fed into the Experiment Design Code for analysis.

The function that generates random binomial data inputs the number of atoms, n that are in the sample, the delay time t_d , and the measurement time t_c . All times in this calculation are measured in half lives of the species being measured. For each atom in the sample, a random number between zero and one R is sampled from a uniform distribution by the standard random number

generator for the Intel FORTRAN compiler for Microsoft Visual Studio. The

time that each atom decays D_i is then equal to

$$D_i = \frac{-\ln(1 - R_i)}{\ln(2)}. \quad (23)$$

If D_i is greater the delay time and less than the sum of the delay and measurement time then the quantity of foreground counts is incremented by one.

The background count generation function comes from Donald Knuth's *Seminumerical Algorithms*, 1969. It inputs the mean background count rate μ , and the measurement time t_c . A target number L is defined as

$$L = e^{-\mu t}. \quad (24)$$

A counting variable p is set to 1. The function then enters a loop. It generates a random number R , and redefines p as

$$p = p * R. \quad (25)$$

If p is greater than L , then the number of background counts is incremented by one and the loop continues, otherwise the total background count is reported.

All data for testing can be rapidly and repeatedly generated in this fashion. The simulation package is then capable of generating scores of sets of realistic data, while being able to judge the results from the EDC against the known sample size and background count rate.

4. Testing and Verification

True validation is extremely difficult given current experimental capabilities because it is not only difficult to generate a sample that contains precisely n atoms, but also to do so for a range of species under multiple experimental conditions. However, the analysis algorithms may be verified using existing mathematics software coupled with a very simple random data generation simulation. The simulation is given n , μ and the experimental data provided to the Experiment Design Code in order to produce data for the EDC to analyze and determine its effectiveness. This code was used both to verify expected error rates given the inputs provided by the user, as well as to compare the performance of the discrete method against the HPS 13.30 standard. The verification method presented here demonstrates a ground up capability based approach that begins by verifying individual subroutines in the Experiment Design Code, followed by a systems level investigation intended to measure the overall performance of the capabilities described in Chapter 3.

4.1 Piecewise Subroutine Verification

Most subroutine verification was accomplished using Microsoft Excel, SigmaPlot or Mathematica. The initialization of the choose array was output to a file and compared against the choose function in Mathematica and agreed to a minimum of 10 digits of precision. The probability distributions for detection

and background counts are also compared to Mathematica calculations with similar results. The monotone cubic interpolation function is compared against a range of monotone increasing functions. The output values of function agree with the array values provided, and the points between maintain a visually pleasing and monotone increasing trend, as shown in Figure 6.

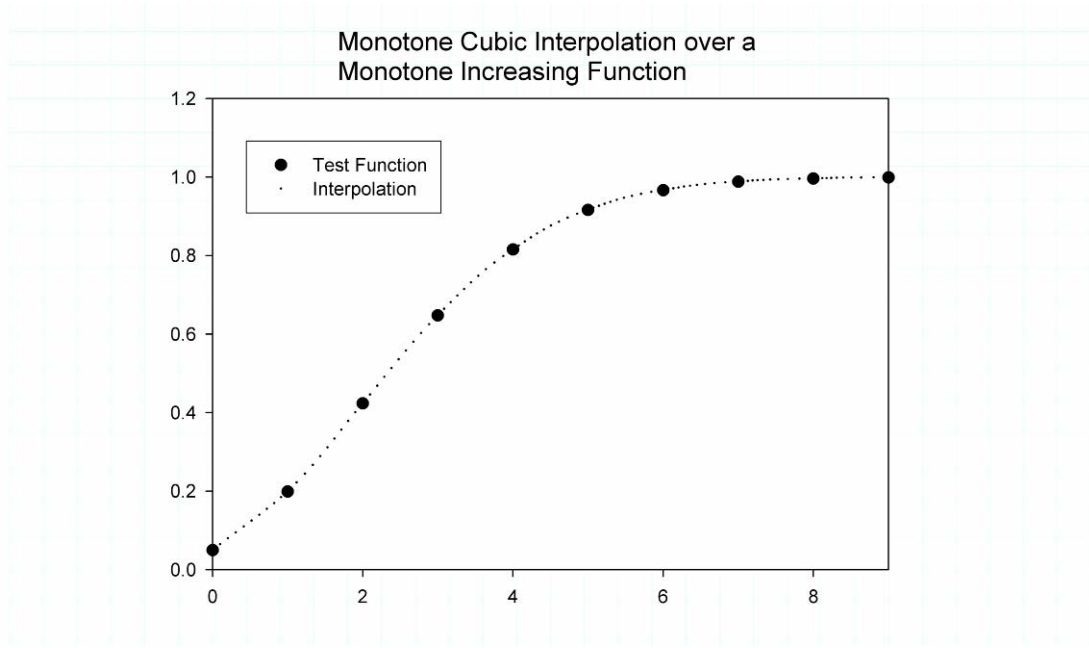


Figure 8 - Verification of Monotone Cubic Interpolation

4.2 Verification of the Experiment Simulator

Systems level verification of the Experiment Design Code requires that the simulation program create foreground and background data that are binomial and Poisson distributed respectively. In order to do this, data sets of 1000 random points were generated under fixed conditions. Those points are then gathered into a histogram and compared to the expected values as described by

the probability density function for the distribution under those circumstances. The statistics package associated with the Sigma Plot (v9.0) scientific graphing software is then used to perform a paired t-test to determine goodness of fit. The paired t-test was chosen because it was the only paired data test available in a standard installation of this software. For each test, the t value is below 1×10^{-3} with orders of magnitude reaching below 10^{-6} in areas where the distributions are narrow. The corresponding P values are reported from .998 to 1. Figure 9 demonstrates a handful of the data sets generated as part of this analysis.

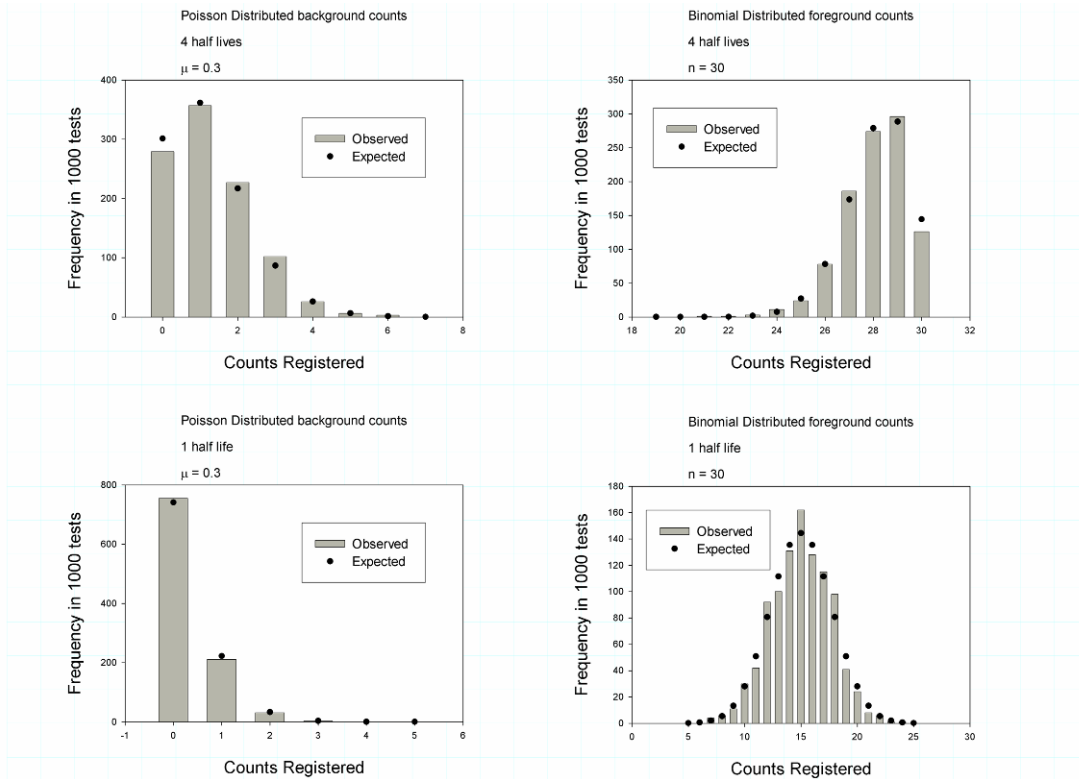


Figure 9 - Verification of Poisson and Binomial data generation

4.3 Confidence Interval Generation

The confidence interval generation routine requires, by its nature, three separate methods of verification. This is because the confidence interval itself has real boundaries that were interpolated between points on a discrete probability density function. The first two tests are related in that they both involve discrete measurements. The first is a simple check to determine if the sum of the probabilities inside the confidence interval is less than or equal to $1-\alpha$. The second checked that the sum of those same points with the two bordering points outside the confidence interval is greater than $1-\alpha$. These tests are intended to ensure that the interpolation routine is being called for the correct locations in the pdf.

The data for both tests are generated by the experiment simulation using random counts for the sample and background measurements and a range of confidence levels. The total failure rate of the first discrete test is about 25.3%. That value is relatively insensitive to confidence level, rising to 28.8% at $\alpha=0.3$ and decreasing to 21.2% at $\alpha=.01$. The failure rate of the second test is 0% in all cases. These levels of failure are almost always to be expected given the conservative manner in which the confidence intervals are being defined. Constructing the confidence interval using the cumulative distribution function allows for the possibility that the confidence bounds will be located between points that constitute a large jump in the probability distribution function.

Figure 10 demonstrates the confidence interval constructed on a sample function of n that fails the first test. The calculated confidence bounds are approximately 35.6 and 40.3. The sum of the probabilities between these bounds is approximately 80.1, a number that fails the first $1-\alpha$ test.

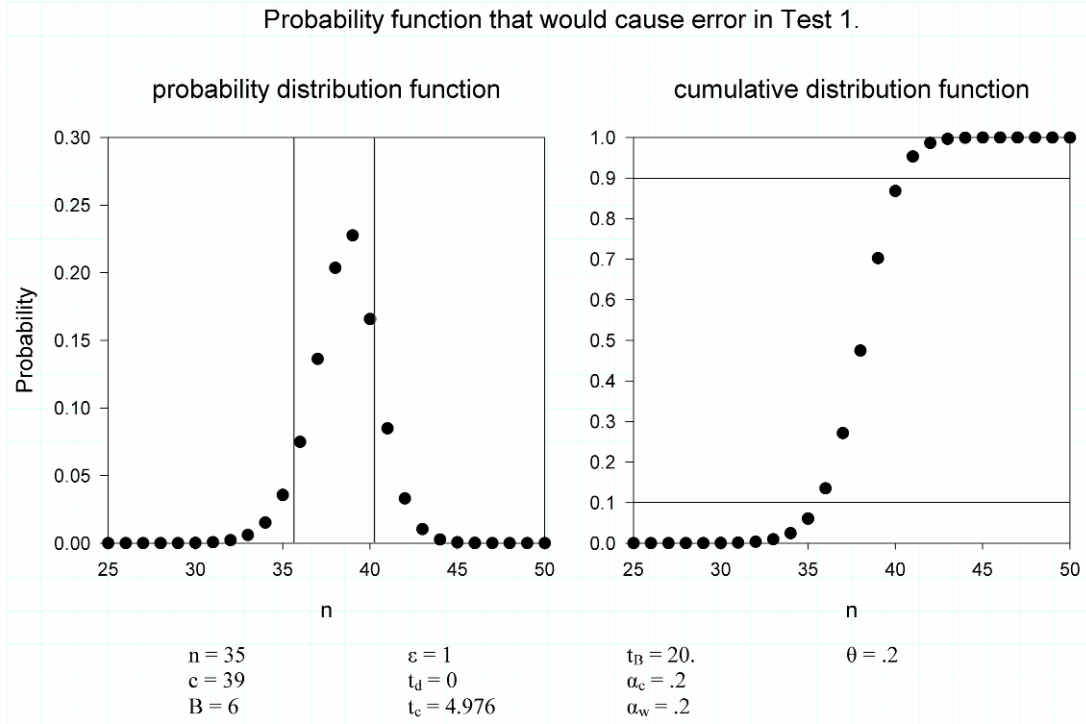


Figure 10 - Confidence interval location in $P(n)$ that fails the $1-\alpha$ test.

A twenty-five percent rate of error may seem alarming, but given where it occurs, it can easily be ignored. Errors tend to occur in situations of relatively extreme high and low values for n where quantification is not in question. Also, the fact that the second test always pass demonstrates that the confidence intervals err on the side of conservatism.

The third verification method for the confidence interval is intended to ensure that the original value for n actually falls within the confidence interval at a rate at or above $1-\alpha$. This has to be done with care because the experimental method and the outcome of the pdf calculation for n are sensitive to the background method. Therefore, several tests are presented which treat the background input differently. They include a Poisson distributed variable b , background counts that are fixed at the most probable level given the mean count rate, as well as b counts that were fixed with a high and low bias. As expected, the unbiased test shows a higher than average success rate than the other three tests. Its success rate is also within a percentage point of $1-\alpha$. The box and whisker plots in Figure 11 show the variation of the results seen in this test. Each data point used to construct this plot is an average of 100 trials. Each of the box and whiskers themselves are calculated from 100 such points. In the plot, it appears the confidence intervals are too narrow. It is possible that a margin of 1 or 2 should be added to the calculated LMQQ to account for failures of the confidence interval to contain n at LMQQ.

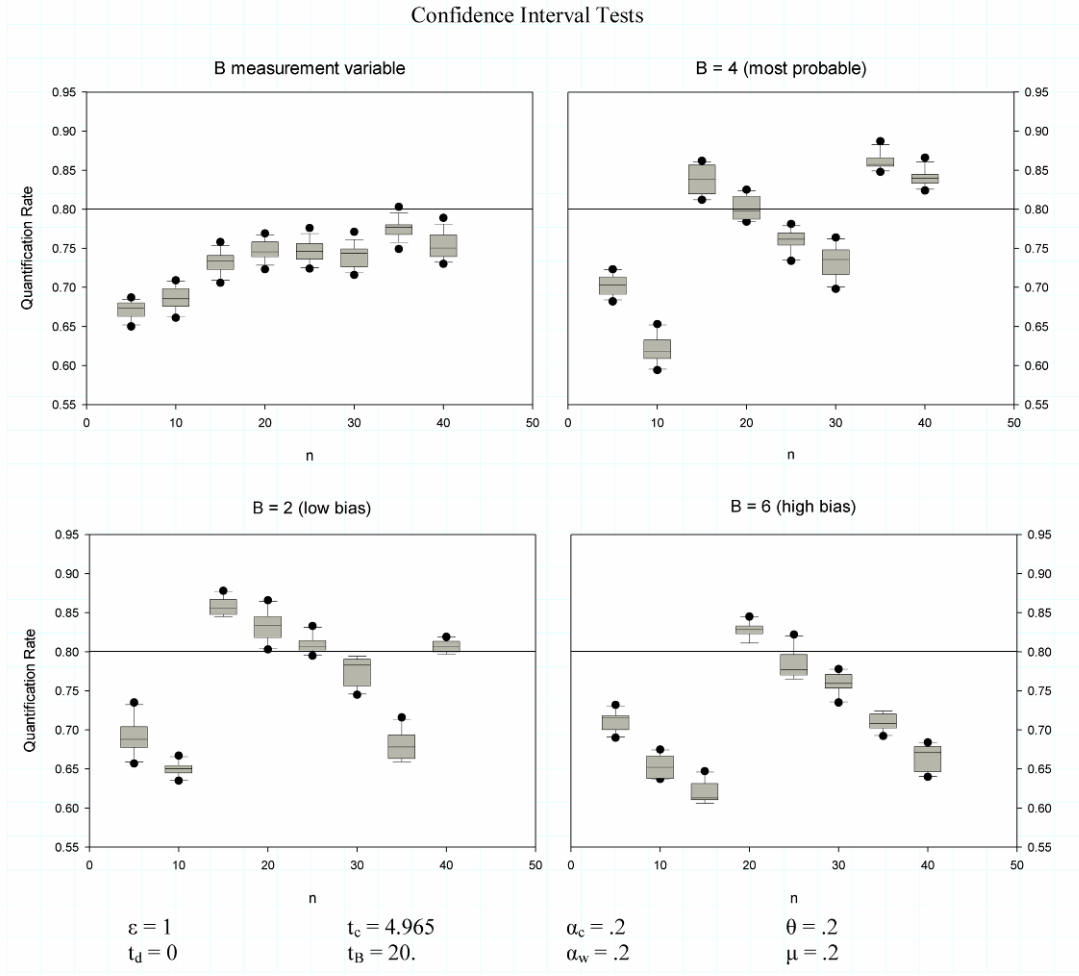


Figure 11 - Verification of confidence interval success, MQQ = 19

There is a noticeable downward trend in the fixed background scenarios. Performing these same tests in detail for much higher values of n was avoided in the interest of computer time. However, a limited extension of n to 100 was performed for a tenth of the trials with a fixed high probability background count. This test indicated that the jumps in the data continue with local negative slope, but an overall positive trend similar to that shown in the plot with variable background.

It should be noted that the results of each fixed background test exceed confidence requirements at the value of n for which the Experiment Design Code optimized the counting time (the counting time during the variable background test is fixed, thus introducing additional error and lowering the values for that plot). This is encouraging because it suggests that the basic experimental method is sound. However, bands of insensitivity where n is higher than the LMQQ are not acceptable. This can occur in the regions in Figure 11 where the quantification distributions fall below $1-\alpha_c$.

4.4 User Input Requirements vs. Simulated Outcomes

Given that the individual building blocks are sound, it is possible to perform a series of systems level tests on the experiment design code. These tests are intended to verify three functions. First, the code should successfully quantify the data of a fraction of trials equal to or greater than $1-\alpha_q$. The confidence intervals should also contain the test quantity of atoms with a rate equal to or greater than $1-\alpha_q$. Finally, the LMQQ reported by the EDC should be the actual LMQQ at the proper time reported.

The first two requirements may be verified simultaneously. As the experiment simulation feeds the analysis routine randomly generated counts, the fraction of trials that produced confidence intervals that were too wide to be quantified was tallied. At the same time, the boundaries of the confidence intervals of accepted trials were recorded and compared to the sample size to

determine how often those intervals did not include n . After over a quarter of a million trials over a range of quantification probabilities and sample sizes, the fraction of quantified samples is never less than $1-\alpha_q$, provided that the sample contains at least the minimum quantifiable quantity. Quantification levels rise quickly to 100% as n increases.

The tests to quantify error are run with 10,000 sample batches. For each sample, if a confidence interval is accepted, then the high and low boundaries are recorded. The histograms displayed in Figures 11 and 12 show how these data sets are distributed. The sample size in these figures is abnormally high compared to the LMQQ of 19 in order to eliminate the impact of non acceptance to the magnitude of the data in the graphs. However, the proportions of the data distributions are very insensitive to sample size. In each test case, the empirical coverage corresponds closely with the calculated theoretical coverage.

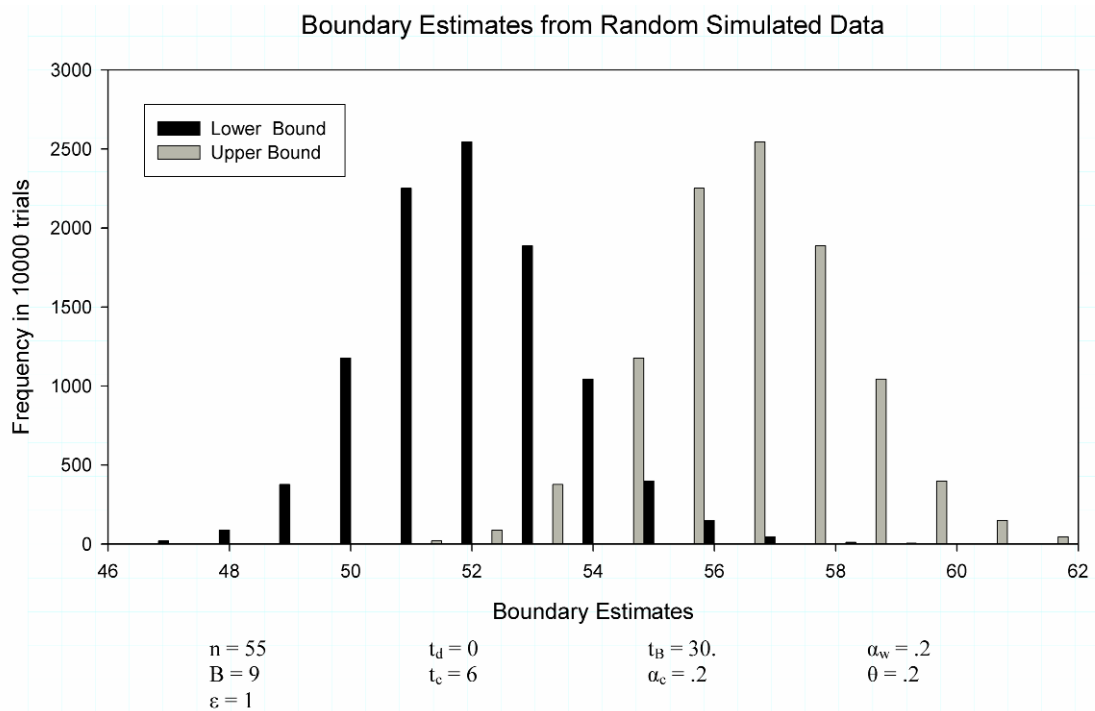


Figure 12 - Confidence interval boundary estimates; B is fixed

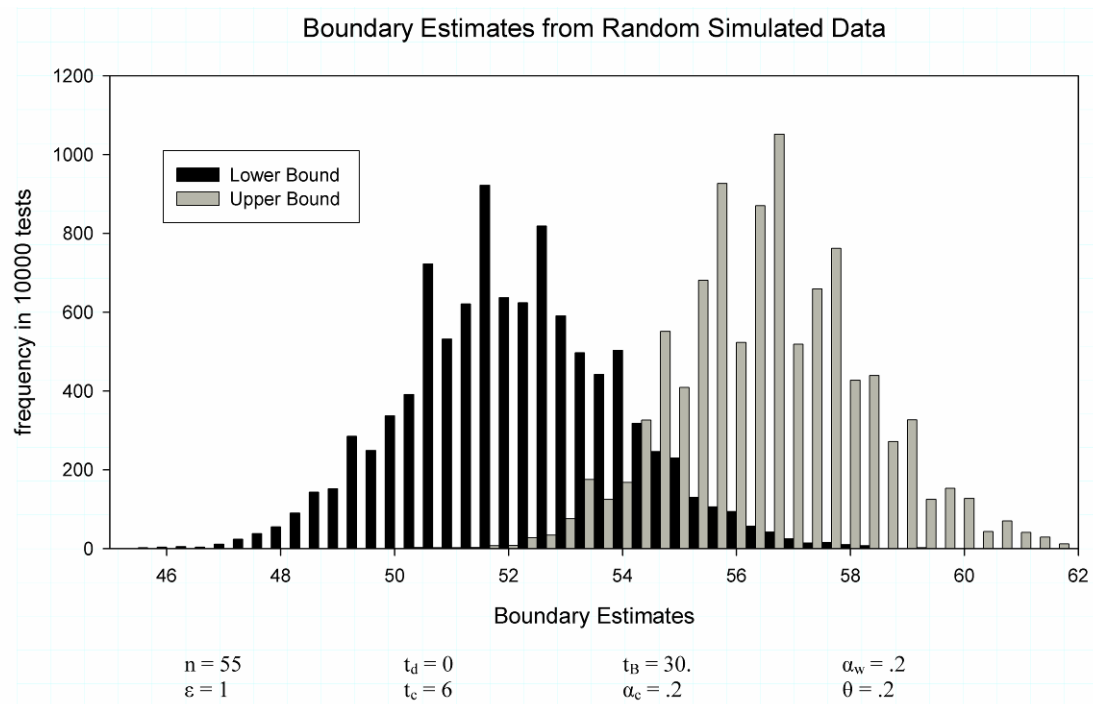


Figure 13 - Confidence interval boundary estimates; B is variable

Error in these tests occurs where the lower boundary is above the sample size of 55 atoms or the upper boundary is below it. In all cases the error is within 2 percentile points of α_c . It is important to note that a bias in the background measurement did not significantly increase the acceptance error. However, it did bias the expectation value by as much as 8% of $\langle n \rangle$ by using bias limits that covered 80% of the possible values of B.

This same experimental scenario is also tested to determine the LMQQ. The first test is a simple check to determine the number of trials that declare a valid confidence interval given n and t_c . The value of n is increased through the predicted LMQQ. The expectation is that the first value of n that demonstrates an acceptable success rate would be the LMQQ and all subsequent values of n would succeed at the same rate or higher. The first chart in Figure 14 contradicts that expectation. The success rate does increase to $1-\alpha$, but does not reach the desired level until $n = 20$, instead of the desired LMQQ of 19. That, by itself is acceptable, but greater values of n actually decrease the success rate in 10,000 tests. Extended testing with higher values for n demonstrated a tendency similar to the one shown in Figure 11. Success rates fluctuate around $1-\alpha$ with a general increasing trend. This is possibly due to sample populations that are likely to produce measurement counts that will force the errors in confidence interval generation discussed above.

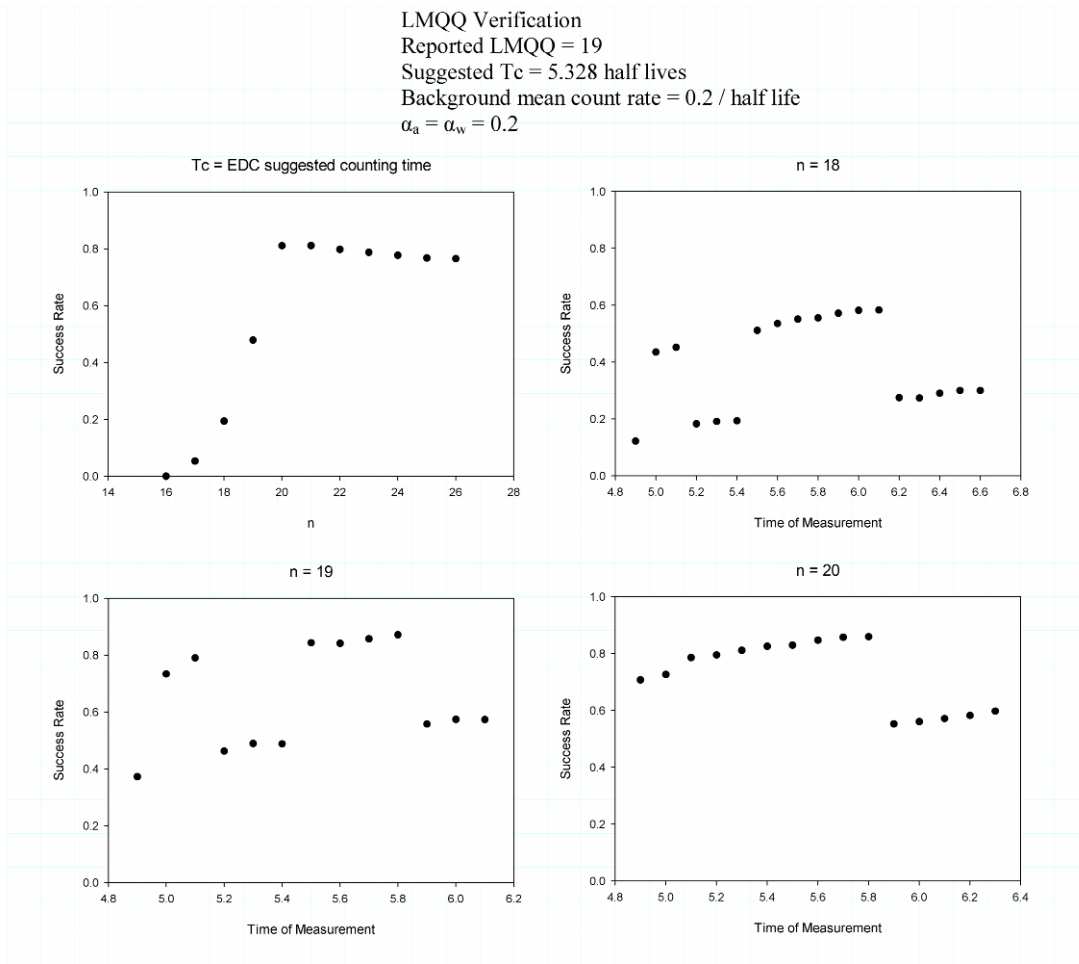


Figure 14 - LMQQ Verification for n and t_c

Figure 14 also demonstrates where errors generated by the confidence interval generation subroutine manifest themselves in time. The three charts measured in time all display discontinuities in the success rate. It should be noted that the data sets presented are calculated with no bias in the background measurement; however biased measurements return similar results with discontinuities in separate locations. Variable background measurements yield smoother results with slightly more error and variation in the outcomes.

4.5 Design Code vs. HPS 13.30

The American National Standards Institute and the Health Physics Society have maintained an analysis method since 1996 that is based on Gaussian error analysis of the measurements. Its standard of measurement is also in terms of mass or activity instead of number. Equation (26) shows the formula to determine the activity in the sample.

$$A = \left[\frac{c}{t_c} - \frac{B}{t_B} \right] / K \quad (26)$$

K is a calibration constant that takes efficiency and process losses into account. In order to account for the appreciable decrease in quantity of a short lived nucleus during measurement time, HPS13-30 translates the actual counting time to an “effective counting interval” in the activity calculation:

$$t_G = e^{-\lambda t_d} (1 - e^{-\lambda t_c}) / \lambda. \quad (27)$$

A confidence interval in count rates is determined by estimating the deviation of the Gaussian distribution by taking the square root of the variance.

$$\sigma = \sqrt{\frac{c}{t_c^2} + \frac{B}{t_B^2}} \quad (28)$$

The σ_α corresponding with α_c defined in the experiment design is found by solving for a value of x on the error function for the normal distribution such that

$$\sigma_\alpha = \sigma x = \frac{1 - \alpha_c}{2}. \quad (29)$$

The final confidence interval equals $A \pm \sigma_\alpha$.

Because this analysis method calculates an activity instead of a number, that activity is translated into a rough number using the following activity relation:

$$n = \frac{A}{\lambda}. \quad (30)$$

The HPS standard is compared to the method presented in this thesis using the same data simulation code described above. Given identical confidences, both methods should converge at a success rate of $1-\alpha_a$ at their point of maximum sensitivity. Figure 15 demonstrates one such test where both methods are attempted from simulated data where the time of measurement is calculated for maximum sensitivity for the experiment design code. The value of n is then increased until the amount of error reaches $1-\alpha_a$. The EDC reaches the desired quantification rate of 80% more than a full order of magnitude prior to the HPS method. However, at values of n higher than 50, calculation times to simulate 10,000 random measurements, the time to calculate confidence intervals increase rapidly. This introduces an interesting engineering problem about where the EDC method should hand off responsibility for analysis to the HPS method.

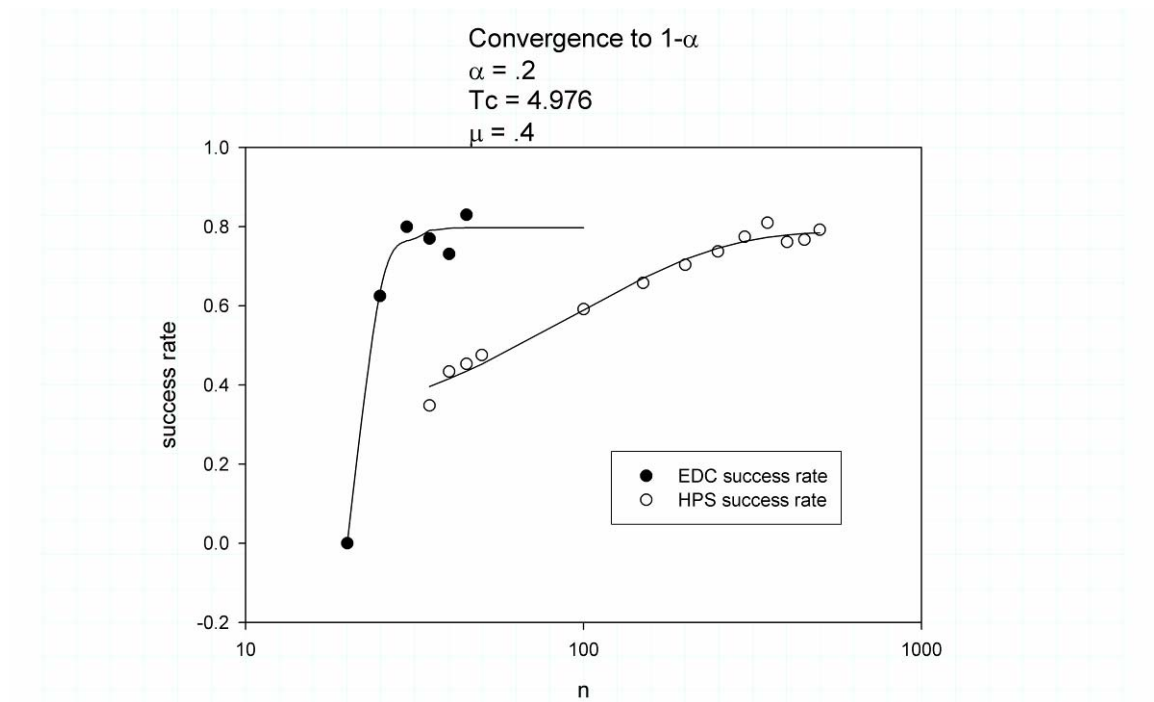


Figure 15 - Comparative convergence to $1-\alpha$ for 2 methods

5. Use of the Experiment Design Code

The experiment design scheme that optimizes sensitivity for a given measurement is not necessarily the best one to satisfy the purposes of the user. There are times when expediency may be more important than optimal sensitivity. For instance, if the researcher is attempting to measure from a large number of samples to find one that has a species presence higher than a threshold value, then processing these samples quickly to maximize throughput and minimize delay time is more important than maximum sensitivity. There may also be times where the physical demands of the detector or schedule constraints place an upper limit to the count duration. The experiment design code allows users to determine the tradeoffs between the demands of the experiment and the amount of time that may be saved by sacrificing some on those requirements. Given a lower bound on the expected relative width and desired probability of acceptance, the code can output a minimum detectable quantity to describe a function of sensitivity under current conditions.

5.1 Inherent Flexibility in Time

When graphed against time, the minimum expected relative width for the LMQQ must fall below θ . The probability of acceptance must also lie above the line defined by $(1-\alpha_a)$. While it may be desirable to maintain the optimal

counting time to maximize robustness, the measurement time can easily be decreased to the first of those limits without sacrificing any of the specified experimental limits. There will be an increase in error, but not more than is acceptable. This method can be used under any circumstance but is also the one least likely to generate a large reduction in measurement time.

5.2 Flexibility in Sensitivity

Increasing the acceptable level of error by increasing any of the three measurements of acceptability will increase flexibility in time by lowering the ERW curves and raising the PoQ curves. Additionally, the user may declare a minimum quantity of interest, resulting in a similar trend in ERW and PoQ curves.

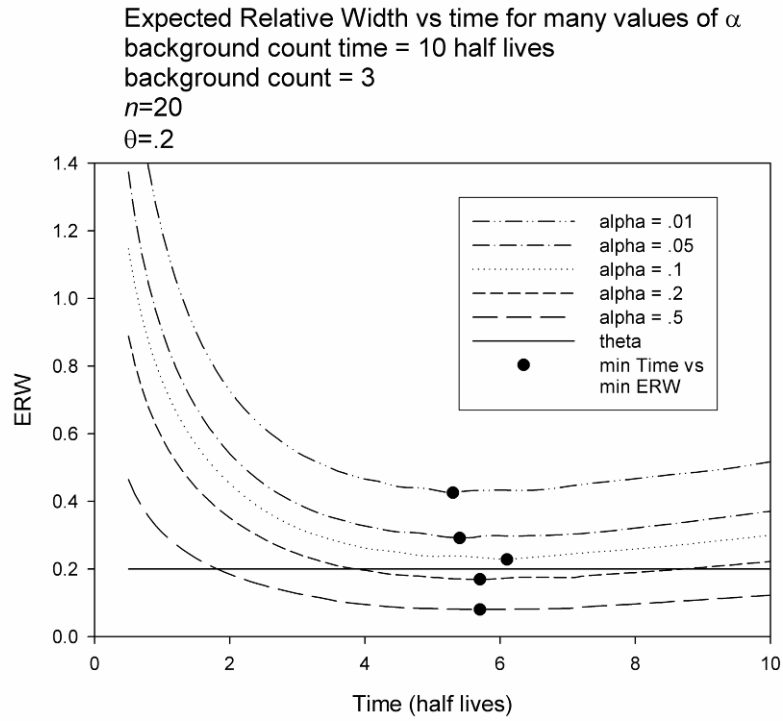


Figure 16 - Effect of flexibility in α_w on count time requirement.

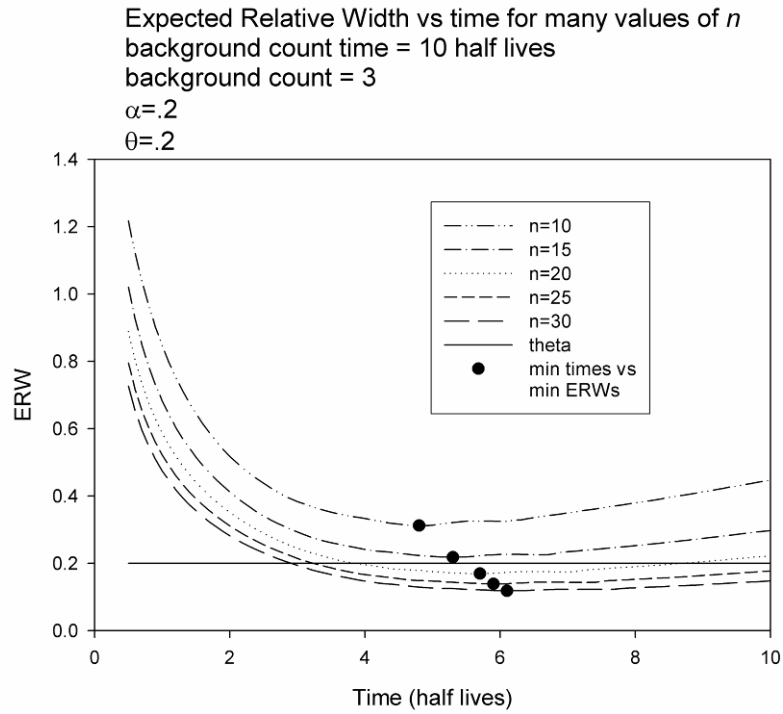


Figure 17 - Effect of flexibility in MQQ on time requirement.

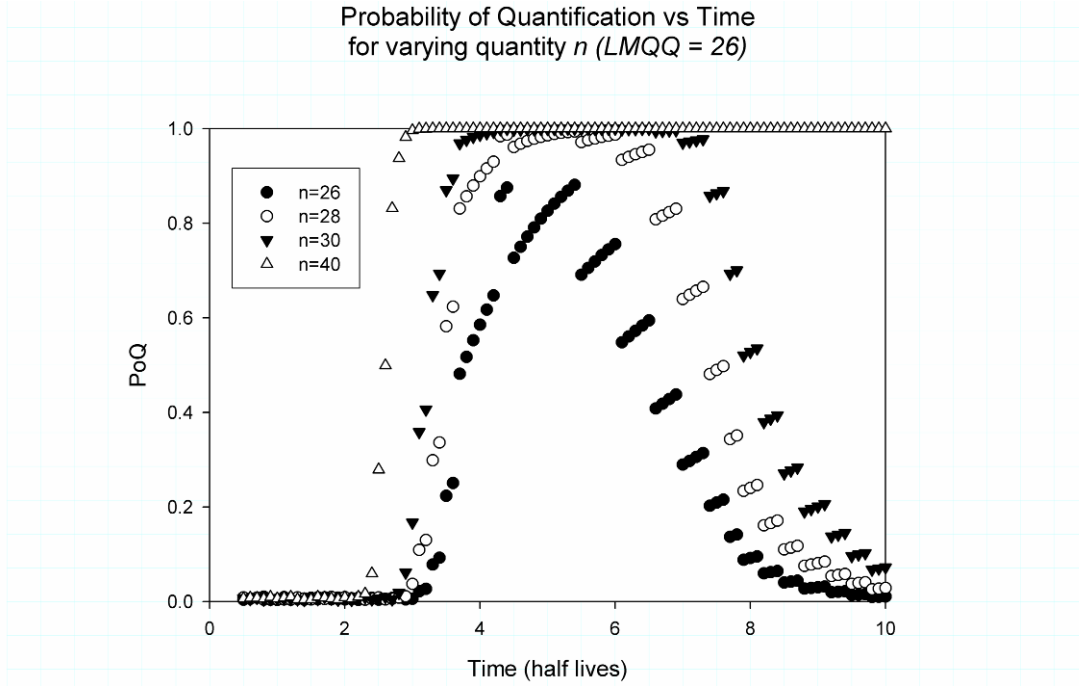


Figure 18 - Flexibility in time for increasing values of n

5.3 Precision in Knowledge of the Background

Additional precision, and thus flexibility, can be gained through an increased certainty in the mean background count rate. Assuming the background is constant, this can be accomplished by simply making a longer measurement prior to the actual experiment. Keeping all other factors constant, a longer background measurement can lower error by as much as an order of magnitude. Figure 19 shows the fraction of trials where n was quantified but fell outside the confidence interval. Increased knowledge in the background decreases the probability of this kind of error.

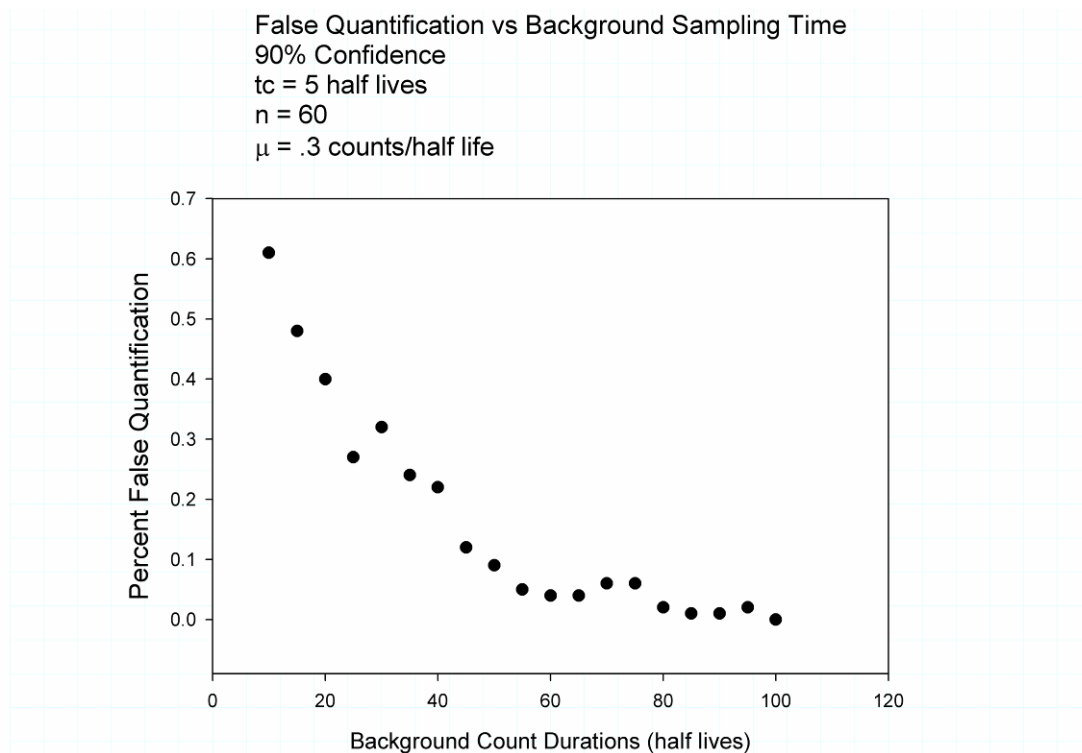


Figure 19 - False Quantification vs. Background Sampling Time

5.4 Minimum Quantifiable Quantity

If the experimental setup and the counting times are fixed, the code may also be used to determine the MQQ. This is also an effective method of adjusting a variable that might be constrained by reality to tune an experiment to an acceptable level of sensitivity. The following graphs detail the importance of short delay times and optimal measurement times in a situation where the experimental procedure may be inflexible.

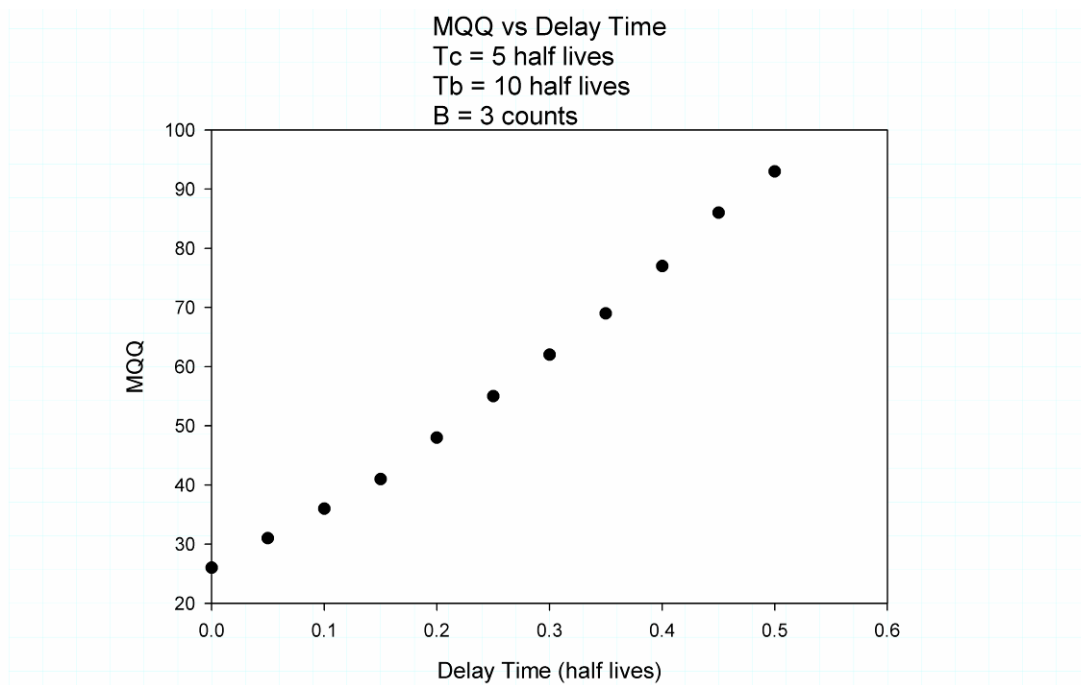


Figure 20 - MQQ vs. Delay Time

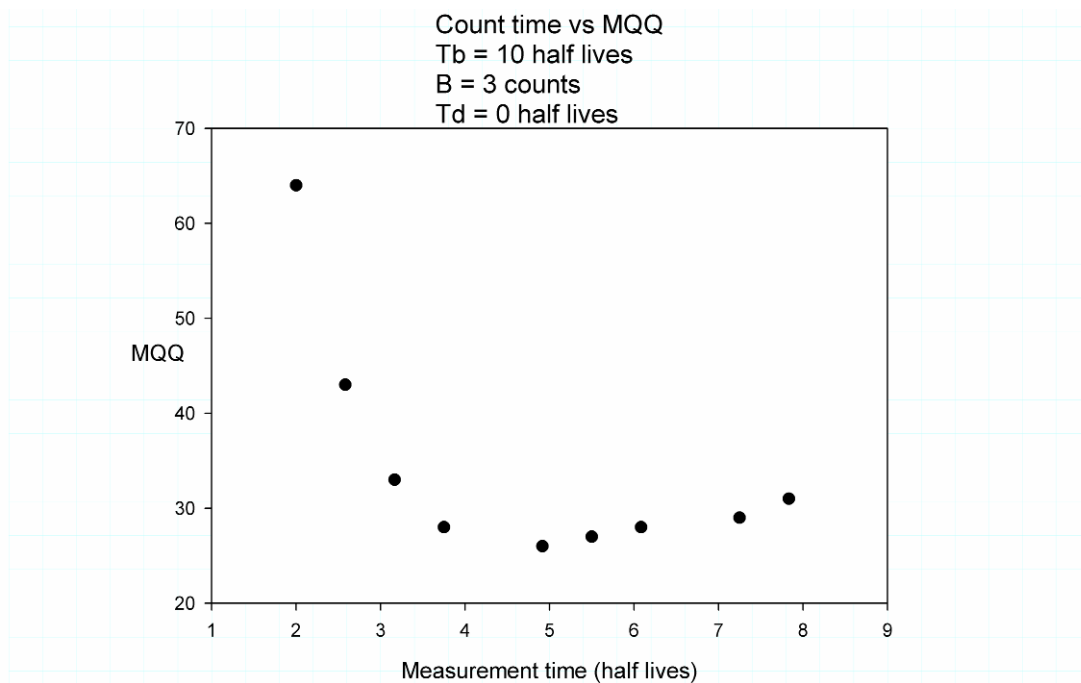


Figure 21 - MQQ vs. Measurement Time

6. Conclusions and Recommendations

Given the data from the simulation, it is clear that the experiment design method presented is capable of very sensitive quantification measurements and that its mathematical basis is sound. This body of research will form the basis for future work that will make this method fully applicable. Advancements should be made in several areas in order for the experiment optimization code to be fully applicable towards modern low count problems. As of this writing, the tool is capable of optimizing measurement times for very small numbers of atoms, analyzing the data from such measurements, and calculating the nominal sensitivity of a given setup provided that conditions meet the assumptions listed in Section 1.6. Removing these restrictions requires work in a handful of primary areas of research.

6.1 Improvements in Confidence Interval Generation

The confidence interval generation method presented in this paper is limited in that it is prone to large jumps in error as the probability distribution function changes over integer values. The generation of the relative expected width has decreased much of this problem in the initial calculation, but it does not eliminate the issue. This analysis method may always be hampered by these discontinuities. If this is true then the level of uncertainty generated by

confidence interval error needs to be characterized. However, logic that produces confidence intervals with smooth error characteristics would be preferable.

6.2 Background Characterization

The background count will never be a perfect Poisson distribution. Driving forces such as nearby sources and laboratory activities can never be perfectly shielded against. The background will also never remain constant, if for no other reason than the fact that cosmic events are out of our control. Continued work should be done in characterizing the possible distributions of background counts and how those differences will affect the outcome of the calculations presented in this paper. Furthermore, procedures that take into account the possibility of a fluctuating mean background count should be explored. These procedures may include fluctuations based on activities that occur on a regional, daily, or even seasonal basis.

6.3 Application of Real Time and Species Variables

The code currently calculates time in half-lives of a non-specific species. In order for this code to be more applicable to the laboratory, it should include a library of radioactive species and their half lives in order to translate these calculations into real measurement times.

6.4 Development of a Graphical User Interface

Running this code by means of namelist input and output files is slow and cumbersome. A GUI designed specifically for the lab would be able to input data from the user quickly and with fewer errors than a text based file. Furthermore, the output would be much more customizable with a graphic interface as opposed to the current text based version.

6.5 Analysis Hand off to Gaussian and Zone of Poor Sensitivity

Gaussian analysis assumptions are acceptable for situations where an order of magnitude greater sensitivity is unnecessary. There comes a point where the sample size is great enough that the analysis should revert back to classical methods. Additionally, exact statistics calculations become very taxing on computer resources well prior to the point where the Gaussian method is accurate. If the EDC is not sufficiently optimized, a zone of poor sensitivity will exist where the sample size is too high to efficiently calculate using exact methods, but too low for precision using Gaussian methods.

6.6 Code Optimizations for High Values of n

The current code contains optimizations that account for some aspects of the calculation. For instance, all necessary values of the binomial coefficient are initialized recursively at the beginning of the program. Also, the probability of

decay detection and background counting for a given t_d and t_c are calculated only once for a given time step. However, run times are very lengthy for any calculation where the expected number of atoms is very high. For instance, this can occur when counts are high, detector efficiency is low, the experimenter demands a particularly high degree of confidence, or the time of measurement is significantly less than the duration of the species' half life. At the least, this code could be rebuilt such that separate calculations could be run in parallel on multiple processors.

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14. ABSTRACT We consider radiation counting experiments used to measure quantities of materials that are short-lived with respect to the count durations. The HPS 13.31 statistical analysis seriously overestimates the uncertainty when the quantity and background are very low. We consider the case in which the objective is to quantify the number of atoms, n, that were present in a sample when it was drawn. Mathews and Gerts [JRNC, 2008] analyzed this case and developed formulas for the probability distribution of n, in order to develop experiment design processes that minimize the smallest detectable quantity of material, thus maximizing sensitivity for the detection problem. We extend their effort to the quantification problem: designing such experiments to find the count duration that yields the lowest possible minimum quantifiable quantity, given the other measurement parameters. A value of n is quantifiable if the precision of the measurement, defined as the width of the confidence interval for n divided by the mean value of n is sufficiently likely to be better than a specified precision tolerance. Our analysis and methods are confirmed by Monte Carlo simulation.					
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