EXPERIMENTAL VERIFICATION OF VARIANCE RESULTS FOR THE SECOND AND THIRD REDUCED FACTORIAL SAMPLE MOMENTS IN NEUTRON MULTIPLICITY COUNTING

Tom Burr [1,*] & Mark Smith-Nelson[2]
[1] Statistical Sciences, Los Alamos National Laboratory, Los Alamos, NM, USA
[2] Advanced Nuclear Technology, Los Alamos National Laboratory, Los Alamos, NM, USA
*Email: tburr@lanl.gov

ABSTRACT
Neutron multiplicity counting is an established method to estimate the spontaneous fission rate in an item. The probability distribution describing the number of neutrons detected in a short time window is typically summarized by its first few moments. For the random error contribution to assay uncertainty, it is useful to evaluate the variances of the second and third reduced factorial sample moments of the probability distribution, because these moments can be related to the spontaneous fission rate. Therefore, a previous paper derived exact expressions for the variances and covariances of the second and third reduced sample moments for nonoverlapping (and overlapping) counting gates, and compared them to the corresponding variances in simulated data. This paper analyzes real data to show that a bootstrap method and the analytical variance expressions give the same results.

Keywords: *bootstrap; sample moments*; *neutron multiplicity counting.*

1. INTRODUCTION

Neutron multiplicity counting is a method to estimate the spontaneous fission rate in an item that includes other neutron sources [1,2]. The probability distribution describing the number of neutrons detected in a short time window (256μs for example) is typically summarized by its first few moments. By making assumptions regarding neutron emission and detection in the item, results are available that relate the sample moments to features of the item, including the spontaneous fission rate [1,2].

Reference [1] derived exact expressions for the variances and covariances of the second and third reduced sample moments for either randomly-triggered or signal-triggered nonoverlapping (and overlapping) counting gates, and compared them to the corresponding variances in simulated data. In addition, a bootstrap method was presented as an alternate but effective way to estimate these variances. The purpose of this paper is to real data to demonstrate agreement between variances of the sample moments as estimated by the bootstrap method and as estimated by the analytical expressions. Because the analytical expressions were derived for any stationary-in-time underlying probability distribution describing the number of neutrons detected in a short time window, they should apply to the real data described in Section 4, provided there is no source or detector drift that would lead to non stationariy.

2. BACKGROUND

The signature to estimate Pu that is exploited in neutron multiplicity counting is the process of spontaneous fission. Spontaneous fission primarily from Pu^{240} leads to multiple (0 to 8 or more) neutrons emitted and detected per fission. Total neutron counting counts all detected neutrons; coincidence counting counts neutron pairs that arrive close in time (within the time of a coincidence gate); multiplicity counting tallies all possible counts of neutrons (0, 1, 2, 3, …,) that are emitted, detected, and counted within each gate, and then typically uses the second and third moments to estimate item properties, as described below.

3. SAMPLE MOMENTS

Modern neutron multiplicity detectors typically open $n = 10^5$ to 10^7 gates each of length approximately 256μs and record the number of detected neutrons in each gate. The gates can be opened on the basis of a detected neutron, randomly, or on a fixed frequent time schedule. This paper considers non-overlapping gates opened on a fixed time schedule. For example, if the gate length is 256μs and the count time is 300 seconds, then approximately 1.2×10^6 gates are opened during the assay.

An integer-valued count, x_i, is observed in each of n non-overlapping random counting windows, denoted $x_1, x_2, \ldots, x_n$. Denote the largest observed count as L and the Feynman frequencies of each possible count as $f_0, f_1, \ldots, f_L$, where $\mathrm{f_j} = \frac{1}{n}\sum_{i=1}^{n} I(x_i = j)$, with the indicator function $I(\,)$ equal to 1 if its argument is true and 0 otherwise.

The *r*th sample moment is defined as $m'_r = \frac{1}{n}\sum_{i=1}^{n} x_i^r = \sum_{j=1}^{L} j^r f_j$. In the limit as $n \to \infty$, the *r*th sample moment converges to the *r*th population moment (by the strong law of large numbers). The *r*th sample factorial moment is $m'_{[r]} = \frac{1}{n}\sum_{i=1}^{n} x_i(x_i - 1)...(x_i - r + 1) = \sum_{j=1}^{L} j(j-1)...(j-r+1) f_j$, and the reduced sample factorial moment is $M_r = \frac{m'_{[r]}}{\text{r!}}$ and it can be shown that $\text{var}(m'_r) = (1/n)(\mu'_{2r} - (\mu'_r)^2)$.

The results below are expressed using Greek symbols such as μ'_r when population moments are involved. In practice, population moments are estimated using the corresponding sample moments. So, for example, the estimate of μ'_r, denoted $\hat{\mu}'_r$, is given by $\hat{\mu}'_r = m'_r = \frac{1}{n}\sum_{i=1}^{n} x_i^r = \sum_{j=1}^{M} j^r f_j$. Therefore, to implement the results below, compute m'_r for *r* = 1, 2, …, 6, and use $\hat{\mu}'_r = m'_r$ in place of μ'_r.

Note that m'_1 is often denoted as $\overline{x}$ and that the maximum likelihood version of the sample variance, $s^2 = \frac{1}{n}\sum_{i=1}^{n}(x_i - \overline{x})^2$ estimates the population variance $\mu'_2 - (\mu'_1)^2$, which is the population second central moment (see definitions below). The unbiased version of the sample variance is $\frac{1}{n-1}\sum_{i=1}^{n}(x_i - \overline{x})^2$, but in our case, *n* is very large so that distinction will not be important, and for consistency throughout, we divide by *n* rather than by *n* -1.

Denote the first three reduced factorial moments as $M_1 = m'_1,\ M_2 = (1/2)(m'_2 - m'_1),\ \text{and}\ M_3 = (1/6)m'_3 - (1/2)m'_2 + (1/3)m'_1,$ respectively.

For randomly-triggered gates, define "reduced moments" $N_1 = M_1,\ N_2 = M_2 - (1/2)M_1^2,\ N_3 = M_3 - M_1 M_2 + (1/3)M_1^3$. The variance and covariance calculations in [1] were for these reduced moments (and for alternate reduced moments for signal-triggered gates). The expressions used below for variances and covariances of N_1, N_2, and N_3 are lengthy and given in [1], This paper extracts variance results from [1] for the first three reduced factorial moments M_1, M_2, and M_3, giving:

$$\sigma^2_{M_2} = (1/n)\{\mu'_2 - (\mu'_1)^2\} \qquad (1),$$

$$\sigma^2_{M_2} = (1/4n)[\{\mu'_4 - (\mu'_2)^2\} + \{\mu'_2 - (\mu'_1)^2\} - 2\{\mu'_3 - \mu'_2\mu'_1\}] \quad (2),$$

and

$$\sigma^2_{M_3} = (1/36n)[\{\mu'_6 - (\mu'_3)^2\} + 9\{\mu'_4 - (\mu'_2)^2\} + 4\{\mu'_2 - (\mu'_1)^2\} - 6\{\mu'_5 - \mu'_3\mu'_2\} + 4\{\mu'_4 - \mu'_3\mu'_1\} - 12\{\mu'_3 - \mu'_2\mu'_1\}] \quad (3).$$

As an alternative to Eqs. (1-3), a simple bootstrap strategy to estimate $\sigma^2_{M_1}$, $\sigma^2_{M_2}$, and $\sigma^2_{M_3}$ (or to estimate $\sigma^2_{N_1}$, $\sigma^2_{N_2}$, and $\sigma^2_{N_3}$) is described in [1]. In the bootstrap strategy, the *n* gates are sampled with replacement for bootstrap sample 1, 2, …, n_{boot}. For each bootstrap sample, the reduced factorial moments M_1, M_2, and M_3 are calculated, and their variances can then be calculated across bootstrap samples.

4. NUMERICAL EXAMPLES ON REAL NEUTRON MULTIPLICITY DATA

Using two real data collections, Tables 1 and 2 present comparisons of Eqs. (1-3) to the bootstrap-based-estimate of $\sigma_{M_1}, \sigma_{M_2}, \text{and}\ \sigma_{M_3}$.

Table 1. Data set 1, n = 10^5 gates. Estimates of $\sigma_{M_1}, \sigma_{M_2}, \text{and } \sigma_{M_3}$.

Stddev Estimates	$\hat{\sigma}_{M_1}$	$\hat{\sigma}_{M_2}$	$\hat{\sigma}_{M_3}$
Eqs 1-3	0.0027	0.0027	0.0019
Bootstrap	0.0027	0.0027	0.0019

The relative standard deviations are approximately 1%, 2.7%, and 7.5% for M_1, M_2, and M_3, respectively.

Table 2. Data set 2, n=10^4 gates. Estimates of $\sigma_{M_1}, \sigma_{M_2}, \text{and } \sigma_{M_3}$.

Stddev Estimates	$\hat{\sigma}_{M_1}$	$\hat{\sigma}_{M_2}$	$\hat{\sigma}_{M_3}$
Eqs 1-3	0.0086	0.0085	0.006
Bootstrap	0.0086	0.0085	0.006

The relative standard deviations are approximately 3.3%, 8.5%, and 23.9% for M_1, M_2, and M_3, respectively, which are approximately $\sqrt{10}$ times the relative standard deviations from Example 1, reflecting the reduction from $n = 10^5$ gates to $n = 10^4$ gates.

Notice that the bootstrap and Eqs. (1-3) give the same results to the number of digits displayed. Most modern multiplicity counting detectors open $n = 10^5$ to 10^7 time gates, so the fact that the bootstrap and analytical approaches exhibit close agreement with only $n = 10^4$ gates and $n = 10^5$ gates is encouraging.

As explained in the Appendix, the covariances cov(M_1, M_2), cov(M_1, M_3), and cov(M_2, M_3) are also important, for example, in using measured M_1, M_2, and M_3 to estimate the spontaneous fission rate. For examples 1 and 2, both the bootstrap-based estimates and corresponding analytical expressions give 0.76, 0.44, and 0.82 for corr(M_1, M_2), corr(M_1, M_3), and corr(M_2, M_3). The analytical expressions are based on repeated use of the fact that $\text{cov}(m'_q, m'_r) = (1/n)\{\mu'_{q+r} - \mu'_q \mu'_r\}$.

Tables 3 and 4 are the same as Tables 1 and 2, respectively, but are for $\hat{\sigma}_{N_1}$, $\hat{\sigma}_{N_2}$, and $\hat{\sigma}_{N_3}$. More digits are displayed in Tables 3 and 4 than in Tables 1 and 2 simply for more complete comparison, because the two correlation estimates (bootstrap and analytical) such as corr(N_1, N_3) did not agree quite as well as for corr(M_1, M_3). However, the agreement is still excellent.

Table 3. Data set 1, n = 10^5 gates. Estimates of $\sigma_{N_1}, \sigma_{N_2}, \text{and } \sigma_{N_3}$.

Stddev Estimates	$\hat{\sigma}_{N_1}$	$\hat{\sigma}_{N_2}$	$\hat{\sigma}_{N_3}$
Eqs 1-3	0.00272	0.001747	0.000992
Bootstrap	0.00271	0.001748	0.000991

The relative standard deviations are approximately 0.4%, 7%, and 61% for N_1, N_2, and N_3, respectively.

Table 4. Data set 2, n=10^4 gates. Estimates of $\sigma_{N_1}, \sigma_{N_2}, \text{and } \sigma_{N_3}$.

Stddev Estimates	$\hat{\sigma}_{N_1}$	$\hat{\sigma}_{N_2}$	$\hat{\sigma}_{N_3}$
Eqs 1-3	0.00858	0.00549	0.00313
Bootstrap	0.00857	0.00552	0.00317

The relative standard deviations are approximately 1%, 23%, and 197% for N_1, N_2, and N_3, respectively, which are approximately $\sqrt{10}$ times the relative standard deviations from Example 1, reflecting the reduction from $n = 10^5$ gates to $n = 10^4$ gates.

For example 1 the bootstrap-based estimates of corr(N_1, N_2), corr(N_1, N_3), and corr(N_2, N_3) are 0.11, 0.02, and 0.26, respectively. The corresponding analytical expressions are also 0.11, 0.02, and 0.26, respectively. For example

2, the bootstrap-based estimates of corr(N_1, N_2), corr(N_1, N_3), and corr(N_2, N_3) are 0.11, 0.03, and 0.26. The corresponding analytical expressions are 0.11, 0.02, and 0.26.

5. SUMMARY

This paper uses two real data sets to extend reference [1] by comparing analytical variance expressions to bootstrap alternatives for the reduced factorial sample moments M_1, M_2, and M_3. For the example considered, using either $n = 10^4$ or $n = 10^5$ gates resulted in excellent agreement between the variance expressions in Eqs. 1-3 and the bootstrap alternatives. In addition, there was excellent agreement in corr(M_1, M_2), corr(M_1, M_3), and corr(M_2, M_3) as estimated by the bootstrap and corresponding analytical expressions. The same excellent agreement between bootstrap-based and analytical expressions was observed for the reduced moments N_1, N_2, and N_3. This excellent agreement provides confirmation of the analytial expressions and illustrates that for the experimental setup for the two real data sets, the probability distribution describing the number of neutrons detected in a short time window is stationary over time.

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APPENDIX. THE BASICS OF NEUTRON MULTIPLICITY COUNTING

This appendix provides additional context and background for neutron multiplicity counting to infer the spontaneous fission rate in a neutron-emitting sample.

One complication in neutron multiplicity counting is that other nuclear reactions generate neutrons in the sample, such as those arising from the production of α particle interactions with low density material such as oxygen, water, fluorine, etc. (denoted (α,n) neutrons). Detected neutrons are therefore a combination of those produced by spontaneous and induced fission and other nuclear reactions. Relatively low energy neutrons are typically detected by capture in He3 tubes in a "well counter," following several energy-loss collisions. The "neutron die-away time" is the time from neutron birth to capture in the He^3 tubes, and is typically much greater than the time scale over which all neutrons from a single nuclear reaction are emitted.

One advantage of neutron multiplicity counting is that neutrons easily penetrate dense samples, so that neutrons from the entire volume of the sample can be detected. However, low-density material such as hydrogeneous material or neutron absorbers such as boron can degrade performance, unless suitable adjustments can be made. A key fact is that it is possible to estimate the mass of Pu240 that would give the same neutron count distribution as that obtained from all the even isotopes (Pu238, 240, 242) in the actual sample [2].

The "point model" [2] is the main model describing neutron emission and detection. The point model assumptions lead to expressions that relate the first three factorial moments of the neutron source distribution to the spontaneous fission rate, *SF*, the neutron leakage multiplication, *M* (which quantifies the rate that neutrons are lost to absorption), and the ratio, α, of (α,n) neutrons to spontaneous fission neutrons. Therefore, using the first three sample moments, it is possible to solve for three unknown sample properties [2]. Typically, the three properties solved for are *SF*, *M*, and α. The efficiency ε is often measured using known sources; in special cases such as waste drums, *M* is known fairly accurately so instead, ε can be estimated. Using three equations to solve for three unknowns leaves no degrees of freedom for model checking, so ideally there will be auxiliary information to justify the model. It is possible to convert an estimate of *SF* to an estimate of Pu mass if isotopic information is available for the sample. Here, we only consider estimation of *SF*, *M*, and α.

Many samples contain Pu isotopes that can be induced by incident (α,n) neutrons and spontaneous fission neutrons to fission, leading to neutron chains or bursts of relatively large numbers of neutrons (5 to 30 neutrons in some chains). The presence of neutron chains due to induced fission impacts the distribution of the number of detected neutrons in counting gates [2-4]. However, the variance results in [1] that we verified in Section 4 are valid whether induced fission occurs or not.

For sample/detector combinations that approximately meet the assumptions, the assay is bias free and precision is determined by calculations such as those shown in [1-3]. Perhaps surprisingly, in this case, there is no need for calibration using representative standards. A practical goal for assay precision is 1% relative standard deviation in 1000 second measurements, with the limiting factor in reaching this goal is the relatively poor relative standard deviation (RSD) of the third moment. Realistically, all items will depart in some ways from the point model assumptions, leading to item-specific biases, and calibration (often using calorimetry to produce working standards) can be used to empirically correct the assay results, particularly for impure samples leading to altered ε. Items such as moist or impure Pu oxide, oxidized Pu metal, and some categories of scrap and waste have presented challenges. Assay biases depend on the extent of departure from the point model and can range from negligible to more than 10% RSD [2]. To summarize this paragraph, the extent to which the sample/detector combination obeys the "point model" assumptions impacts the total measurement error in estimating the SF rate, but in nearly all cases, for the random error contribution, it is useful to evaluate the variances of the second and third reduced sample moments as defined below.

The exact variance results provided in [1] could be used in place of approximate variance results used, for example, [3] to estimate the random error variance, of the effective Pu240 mass. The approximate results in [3] used moments up to the fifth moment; the exact results in [1] included all moments up to the sixth moment. In the common case of using three equations to solve for any subset of (*SF*, *M*, and α), a weighted least squares solution can then use an exact weight covariance matrix W based on the variances $\sigma^2_{N_1}, \sigma^2_{N_2}$, and $\sigma^2_{N_3}$ and covariances cov(N_1, N_2), cov(N_1, N_3),and cov(N_2, N_3) which are provided in [1], or on $\sigma^2_{M_1}$, $\sigma^2_{M_2}$, and $\sigma^2_{M_3}$ and the covariances cov(M_1, M_2), cov(M_1, M_3),and cov(M_2, M_3). Most approaches use the first three reduced factorial sample moments, defined in the main text.

Details such as detector die-away time and dead time must be considered, but these variances and covariances given below provide for efficient estimates of (*SF*, *M*, and α) and for estimates of the variances and covariances of these estimates. In addition, these exact results provide a way to quantify the random error variance of the assay as a function of the counting time.