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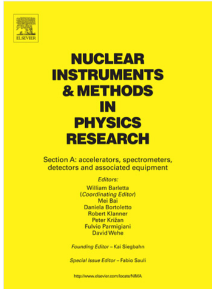
To be published in: Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment

DOI to publisher's version: 10.1016/j.nima.2017.08.036



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PII: S0168-9002(17)30922-1
DOI: http://dx.doi.org/10.1016/j.nima.2017.08.036
Reference: NIMA 60055

To appear in: *Nuclear Inst. and Methods in Physics Research, A*

Received date : 27 April 2017
Revised date : 3 August 2017
Accepted date : 21 August 2017

Please cite this article as: C. Dubi, S. Croft, A. Favalli, A. Ocherashvili, B. Pedersen, Estimating the mass variance in neutron multiplicity counting—A comparison of approaches, *Nuclear Inst. and Methods in Physics Research, A* (2017), http://dx.doi.org/10.1016/j.nima.2017.08.036

Estimating the mass variance in neutron multiplicity counting - A comparison of approaches

C. Dubi[1], S. Croft[2], A. Favalli[3], A. Ocherashvili[1], and B. Pedersen[4]

[1]Nuclear Research Center Negev (NRCN), POB 9001, Beer Sheva, Israel
[2]Oak Ridge National Laboratory,P.O.B 2008, Oak Ridge, TN 47831-6166 USA
[3]Los Alamos National Laboratory, P.O.B 1663, Los Alamos, NM 87545 USA
[4] Nuclear Security Unit, institute of transuranium elements, Via E. Fermi 2749, Ispra, Italy.

August 3, 2017

Abstract

In the standard practice of neutron multiplicity counting , the first three sampled factorial moments of the event triggered neutron count distribution are used to quantify the three main neutron source terms: the spontaneous fissile material effective mass, the relative (α, n) production and the induced fission source responsible for multiplication.
This study compares three methods to quantify the statistical uncertainty of the estimated mass: the bootstrap method, propagation of variance through moments, and statistical analysis of cycle data method. Each of the three methods was implemented on a set of four different NMC measurements, held at the JRC- laboratory in Ispra, Italy, sampling four different Pu samples in a standard Plutonium Scrap Multiplicity Counter (PSMC) well counter.

Keywords: Neutron Multiplicity Counting, Passive neutron interrogation, Uncertainty Quantification.

1 Introduction

In the standard practice of Neutron Multiplicity Counting (NMC), the first three sampled factorial moments of the event triggered neutron count distribution are used in an inversion model to extract the spontaneous fission rate, the (α, n) rate and the multiplication of the item. A significant advantage of NMC over other nondestructive assay methods is the

relative transparency of structural materials to neutrons, making it a useful method when sampling impure, poorly characterized items.
As in any experimental method, uncertainty estimation is an inherent part of the measurement, and no result is complete without it. Yet, at present, there is no comprehensive guide regarding how to estimate the uncertainty of the measured mass using NMC.
Typically, "uncertainties" can be divide into three categories: uncertainties in the physical parameters (such as detection efficiency, the prompt fission multiplicity distributions etc.), systematic errors (due to model assumptions- such as the single energy point model and neglecting the delayed neutrons - or due to numeric methods) and the statistical uncertainty due to the random nature of neutron counting (and fractionally larger when sampling the higher moments).
From an operational point of view, understanding the statistical error has high importance for two main reasons: first, sampling high moments of the count distribution is vulnerable to a large statistical uncertainty. Second, out of all the uncertainty factors mentioned , the statistical uncertainty is the only one the user can control by extending the duration of the measurement.
The objective of the present study is to perform a comparison between three methods for estimating the statistical uncertainty of the estimated mass: the bootstrap method, propagation of variance through moments and statistical analysis of cycle data.
The comparison was done experimentally. Each of the three methods was implemented on a set of four NMC measurements, held at the JRC- laboratory in Ispra, Italy, sampling four different Pu samples in a Plutonium Scrap Multiplicity Counter (PSMC) well counter [1]. In order to create a reference value, the measurement was repeated for a sufficient number of times (30-90), and the statistical spread of the repetitions was used as the reference value.
The paper is arranged in the following manner: section 2 gives the necessary background on NMC and give an overview of the paper. Section 3 describes the different methods used to estimate the statistical uncertainty. Section 4 describes the experimental setting and introduce and explain the reference values used for comparison. Section 5 describes the experimental results, and section 6 concludes.

2 Neutron Multiplicity counting

2.1 Neutron Multiplicity Counting and the SVM method

Most spontaneous fissile materials emit neutrons in a known rate (per mass unit). Thus, in a system with a known detection efficiency, the mass of the spontaneous fissile material is propositional to the average count rate of the spontaneous fission neutrons in a known proportion. However, such simple consideration only provides a partial solution, since the count rate of the neutron detections is highly influenced by two additional neutron sources: (α, n) reactions in sample impurities, and induced fissions (typically in the odd Plutonium isotopes). Moreover, since the detection system is often based on ^3He proportional counters imbedded in a moderating medium, variations in the energy spectrum between the different neutron sources have a negligible effect on the counter efficiency or the die away time, and the neutrons can not be distinguished through energetic considerations.
On the other hand, since the three sources have a different statistical nature, the contribution of each source can be quantified by measuring higher moments of the count distribution.

Such general considerations are referred to as Neutron Multiplicity Counting (NMC) or Time Interval Analysis (TIA).
Most spontaneous fissile materials emit neutrons in a known rate (per mass unit). Thus, in a system with a known detection efficiency, the mass of the spontaneous fissile material is propositional to the average count rate of the spontaneous fission neutrons in a known proportion. However, such simple consideration only provides a partial solution, because the count rate of the neutron detections is highly influenced by two additional neutron sources: (α, n) reactions in sample impurities, and induced fissions (typically in the odd isotopes). Moreover, since the detection system is often based on ^3He proportional counters imbedded in a moderating medium, and all neutron sources have (more or less) the same energetic spectrum, the neutrons can not be distinguished through energetic considerations. On the other hand, because the three sources have a different statistical nature, the contribution of each source can be quantified by measuring higher moments of the count distribution. Such general considerations are referred to as NMC or time interval analysis.
The shift register method is routinely used in NMC [2], where the so called Singles, Doubles and Triples rate are used to quantify the three neutron sources. Other methods include the Random Trigger Interval (RTI) method [3] and the Skewness-Variance-Mean (SVM) method [4]. Because all methods, eventually, sample the first the moments of the count distribution (although through different random variables), all methods are mathematically equivalent [1] [5].
Since the outline of the present study is estimating the statistical uncertainty in the observables - and the final mass result -our choice is the SVM method, where the sampled quantities are very simple: the first three central moments of the number of detections in consecutive (fixed) gates.
In more detail, the SVM method is implemented in the following manner: the measurement (of duration of T_{tot}) is divided into N consecutive gates of duration T (where T is typically on the order of the system neutron die away time, and $N = T_{tot}/T >> 1$). Denoting the number of neutron detections in the k_{th} gate ($1 \leq k \leq N$) by X_k, the sample mean is given by $\widehat{E}(X) = \frac{1}{N}\sum_{k=1}^{N} X_k$, sample variance is evaluated through $\widehat{Var}(X) = \frac{1}{N-1}\sum_{k=1}^{N}(X_k - E(X))^2$ and the skewness by $\widehat{Sk}(X) = \frac{1}{N-1}\sum_{k=1}^{N}(X_k - E(X))^3$[2]. Once the sampling is done, the generalized factorial neutron multiplicity moments- defined as the factorial moments of the number of neutron emmited in an entire fission chain starting with

[1]The term "mathematically equivalent" refers to the fact that all methods share the same physical interpretation and model assumptions (and the same statistical convergence rate). But how the information is obtained may differ: different hardware, overlapping vs. non overlapping gates, different accidental estimations, different dead time formulation etc. The expectation for all methods will be the same even though the uncertainty might not.

[2]We use the notations $\widehat{E}(X), \widehat{Var}(X)' \widehat{Sk}(X)$ rather that $E(X), Var(X), Sk(X)$ to distinct between the sampled moments, and the theoretical moments, as would be sampled in a infinite measurement

a single source event- are related to the sampled moments by [4]:

$$\begin{aligned} D_{G,1} &= \frac{\widehat{E}(X)}{SP_dT} \quad (2.1)\\ D_{G,2} &= \frac{\left(\widehat{Var}(X) - \widehat{E}(X)\right)}{SP_d^2(e^{-\lambda T} - 1 + \lambda T)/\lambda} \\ D_{G,3} &= \frac{\left(\widehat{Sk}(X) - 3\widehat{Var}(X) + 2\widehat{E}(X)\right)}{SP_d^3(e^{-2\lambda T} + e^{-\lambda T} - 3 + 2\lambda T)/2\lambda}. \end{aligned}$$

where P_d is the detection efficiency (the probability that an emergent neutron will be detected), S is the source rate- the number of source events (spontaneous fissions or (α, n)) per time unit and λ is the reciprocal of the detector system die away time, adopting the exponential model.
Finally, the generalized factorial moments are used to quantify the different neutrons sources through the so called "Bohnel Method" [14, 15], describing the generalized factorial moments in term of the following parameters:

1. The spontaneous fission fraction U: the fraction of the source that is due to spontaneous fissions only[3]

2. The leakage multiplication factor M_L: the neutron leakage multiplication factor, defined as the product between the total multiplication and the probability of neutron leakage [16].

3. $D_{sf,n}, D_{if,n}$ The n_{th} factorial moments of the neutron emission distribution in a spontaneous/induced fission (respectively)

Denoting by $D_{G,\ell} = D_{G,1}(U, M_L)$ the ℓ_{th} factorial moment of the distribution of the number of neutron emitted in an entire fission ignited by a single spontaneous source event, explicit formulas for $D_{G,\ell}, (\ell = 1, 2, 3)$ in the prompt, point kinetics approximation are given by:

$$\begin{aligned} D_{G,1}(U, M_L) &= (U(D_{sf,1} - 1) + 1)M_L \quad (2.2)\\ D_{G,2}(U, M_L) &= M_L^2\left(UD_{sf,2} + \frac{M_L - 1}{D_{if,1} - 1}(U(D_{sf,1} - 1) + 1)D_{if,2}\right) \\ D_{G,3}(U, M_L) &= M_L^3(UD_{sf,3} + \frac{M_L - 1}{D_{if,1} - 1}(3UD_{sf,2}U_{if,2} \\ &+ D_{if,3}(U(D_{sf,1} - 1) + 1)) + 3\left(\frac{M_L - 1}{D_{if,1} - 1}\right)^2 D_{if,2}^2(U(D_{sf,1} - 1) + 1)) \end{aligned}$$

Equation 2.1 and 2.2 form a set of three (non linear) equations with three unknowns. Once the set of equations is solved, the mass is proportional to $S \times U$, and the proportion coefficient is the reciprocal of the spontaneous fission rate (per gram). When measuring Pu samples, the spontaneous fission rate is approximately 473.5 fissions per gram per second [2].

[3]if we denote by S_f the spontaneous fission rate, and by S_α the (α, n) rate, then $S = S_f + S_{alpha}$ and $U = S_f/(S_f + S_\alpha)$.

2.2 Aim and Motivation

In recent years, the use of NMC methods has seen constant growth, becoming a standard tool in safety, safeguards and facility operations. Thus, the need for a full uncertainty quantification is becoming more important. In response, we see growing interest, both academic and practical, in uncertainty quantification in NMC [17].
As stated, in the present study, we will restrict our discussions to the third factor only: statistical uncertainty. Quantification of the statistical uncertainty of the measurement variance in NMC, naturally, has been studied before, and there are several publications regarding both the estimation ([6],[7],[10], to state a few) and the optimization ([11],[12]) of the measurement variance. The outline of the present study is to perform a comparative research between three of the both basic and widely used methods for estimating the statistical uncertainty in the measurements. With the end user of NMC at mind, we applied the following principles through the study:

1. All procedures and methods were described in full detail.
2. Comparison was carried out not only in terms of the observables (the measured moments), but also - and more importantly- in terms of the final outcome of the measurement- the mass.
3. The different methods were compared not only between themselves- which will only indicate if the methods agree or not- but also with a reference value (which will be explained in section 4.2). This allows us to determine through quantification which method performs best.

3 Estimating the statistical uncertainty in NMC

In this section, a description of each of the methods studied is presented.
Before introducing the methods, we start with a remark. In the first two methods described, estimating the statistical uncertainty in the measured mass is done in two steps: The first is to estimate the uncertainty of the experimental observables (here $E(X)$, $Var(X)$ and $Sk(X)$). Then we must run a sensitivity analysis, in order to understand how the error in each observable propagates onto the final result of the estimated mass. In section 3.2 we give a mathematical formalism connecting the uncertainty in the measured values and the statistical uncertainty of the mass.

3.1 Description of methods

3.1.1 Method I: Statistical Analysis of Cycle Data (SACD)

The first method described is the most basic one. By definition, the best estimation for the statistical uncertainty in any sampled value is to repeat the experiment enough times, and directly sample the width of the distribution. From a practical point of view, however, this may be prohibitive: the uncertainty estimation often needs to be obtained through a single measurement. On the other hand, the measurement can always be treated as a "composition" of any number of sub-measurements. Thus, we may estimate the statistical uncertainty in the following manner: The total measurement will be broken into n sub- measurements (with, typically, $n \geq 10$), each of duration T/n. Then, for each sub-measurement, the first three central moments are evaluated. For a measurement of duration T/n, the standard deviation of the j_{th} moment, can be estimated through the standard deviation between the different sub-measurement. Finally, the standard deviation for the full measurement is estimated by division byx $\sqrt{n}$.

Theoretically, in each measurement we may also compute the mass, and then estimate the standard deviation of the mass directly. This, however is somewhat problematic because it requires each sub-measurement to yield a meaningful result (that is , significantly larger than the detection limit). Looking at equations 2.1, we see that the variance must be larger than the mean, and the skewness must be larger than the variance added to twice the mean (in terms of the multiplicity method, this is just to state that the doubles and triples must have a positive rate). If the sub-measurement is too short, the statistical uncertainty will cause a deviation from these conditions, resulting with an estimated multiplication factor M_L smaller than one, or a negative sampled (α, n) rate- which are nonphysical values. In terms of the mass, this will create a very large error, which is totally incompatible with the statistical error of the full measurement. This approach, in fact, was also tested when working on the present study, and as we will demonstrate in section 5, results are less favorable.

3.1.2 Method II: Propagation of Variance through Moments (PVM)

As a general fact, if X is a random variable, then the variance of $g(X)$ for a general function[4] g is given by $E\left[(g(X) - E(g(X)))^2\right]$. In particular, the variance of the variance and the variance of the skewness may be evaluated by:

$$Var(Var(X)) = E\left[\{(X - E[X])^2 - E\left[(X - E(X))^2\right]\}^2\right] \tag{3.3}$$

$$Var(Sk(X)) = E\left[\{(X - E[X])^3 - E\left[(X - E(X))^3\right]\}^2\right] \tag{3.4}$$

.

Using equations 3.3 and 3.4 we can explicitly write down the variance of the variance as an algebraic combination of the first four moments, and the variance of the skewness as an algebraic combination of the first six moments (see appendix A). Since the first six moment may be sampled from the data, this will result with an approximation of the standard deviation for both the variance and the mean.

One last remark: This study uses the SVM method. Thus, the moments sampled are the central moments. Ref [5] established that all moments based NMC methods are equivalent, and have the same statistical convergence rate. Appendix A gives explicit formulas for the variance of the second and third factorial moments in terms of the first six moments[5]

3.1.3 Method III: The Bootstrap method (BS)

The bootstrap method is a standard re-sampling method, aimed to estimate the statistical uncertainty when sampling from a large population [18]. The bootstrap method was adopted

[4]From a theoretical point of view, the function g must satisfy certain mathematical conditions, such as Lebesque integrability. Practically, all the conditions are met in the present context, and the formulas are fully applicable

[5]The PVM methods works here because the gates are not overlapping and we are using the simple count distribution. When using the event triggered gate "$(R + A)$" in the multiplicity method [2], it will however bring some potential complications.

to NMC in [6, 19], and was carefully studied in [13]. A full description of the method is beyond the scope of this paper, but the general idea is to create the so called "bootstrap distribution" by a random re-shuffling process of the data.
The "shuffling" process is done by breaking the total measurement into very short gates (typically, in the order of 1 sec.), and then the gates are randomly chosen to create a new "pseudo-measurement". This shuffling procedure is repeated for a large number of times (typically 300 repetitions) and then, the standard deviation is estimated as the standard deviation of the bootstrap distribution.
As stated, out of the three methods introduced in this paper, the bootstrap method is the only one in which we will estimate the uncertainty in the mass directly [13]. The implementation of the method in the present study used 2 second gates, and the reshuffling procedure was repeated 300 times (see [13] for the exact reshuffling procedure).

3.2 Input/Output Error propagation

Our next step is to understand how the statistical uncertainty of the sampled moments will effect the uncertainty in the mass. In the remainder of this section, we follow the methodology introduced in [7], added for sake of completeness (see also [8]). According to the current common practice, we adopt the point model equations.
Combining equation 2.1 and 2.2 will result with an explicit set of equations of the form:

$$\begin{aligned} E &= F_1(S, U, M_L) \\ Var &= F_2(S, U, M_L) \\ Sk &= F_3(S, U, M_L) \end{aligned}$$

Therefore, using the first order Taylor series to estimate the uncertainty, we may write:

$$\begin{aligned} \Delta_E &= \left|\frac{\partial F_1}{\partial S}\right| \Delta_S + \left|\frac{\partial F_1}{\partial U}\right| \Delta_U + \left|\frac{\partial F_1}{\partial M_L}\right| \Delta_{M_L} \\ \Delta_{Var} &= \left|\frac{\partial F_2}{\partial S}\right| \Delta_S + \left|\frac{\partial F_2}{\partial U}\right| \Delta_U + \left|\frac{\partial F_2}{\partial M_L}\right| \Delta_{M_L} \\ \Delta_{Sk} &= \left|\frac{\partial F_3}{\partial S}\right| \Delta_S + \left|\frac{\partial F_3}{\partial U}\right| \Delta_U + \left|\frac{\partial F_3}{\partial M_L}\right| \Delta_{M_L} \end{aligned} \tag{3.5}$$

or $\begin{pmatrix} \Delta_E \\ \Delta_{Var} \\ \Delta_{Sk} \end{pmatrix} = \mathcal{D} \begin{pmatrix} \Delta_S \\ \Delta_U \\ \Delta_{M_L} \end{pmatrix}$, where $\mathcal{D} = \begin{pmatrix} \frac{\partial F_1}{\partial S} & \frac{\partial F_1}{\partial U} & \frac{\partial F_1}{\partial M_L} \\ \frac{\partial F_2}{\partial S} & \frac{\partial F_2}{\partial U} & \frac{\partial F_2}{\partial M_L} \\ \frac{\partial F_3}{\partial S} & \frac{\partial F_3}{\partial U} & \frac{\partial F_3}{\partial M_L} \end{pmatrix}$.
However, in the present context, we are interested in the opposite direction: we use the sampled values of σ_E, σ_{Var} and σ_{Sk} as estimators for the uncertainties Δ_E, Δ_{Var} and Δ_{Sk}, and Δ_S, Δ_U and Δ_{M_L} are computed through

$$\begin{pmatrix} \Delta_S \\ \Delta_U \\ \Delta_{M_L} \end{pmatrix} = \mathcal{D}^{-1} \begin{pmatrix} \sigma_E \\ \sigma_{Var} \\ \sigma_{Sk} \end{pmatrix} \tag{3.6}$$

Before we continue, some clarification is required regarding formula 3.6. Equation 3.6 *is not* a formula for variance of the computed values, but rather a geometric approximation

of the uncertainty. Assume the statistical uncertainty of E, Var and Sk are σ_E, σ_{Var} and σ_{Sk}. This means that the "true" values of the point (E, Var, Sk) in the domain $A = [E-\sigma_E, E+\sigma_E] \times [Var-\sigma_{Var}, Var+\sigma_{Var}] \times [Sk-\sigma_{Sk}, Sk+\sigma_{Sk}]$. If we assume that the uncertainties are small enough (with respect to sampled values), equation 3.6 describes the image of A in the (S, U, M_L) plane. Thus, Δ_S and Δ_U (as defined in equation 3.6) serves as an estimate of the statistical uncertainty (this sort of geometric approach is often used, see [7, 8, 9]).
Finally, since the mass is estimated through $mass = \frac{S \times U}{473.5}$, we have that[6]:

$$\Delta_{mass} = \frac{U \times \Delta_S + S \times \Delta_U}{473.5} \tag{3.7}$$

4 Experimental setting

4.1 General

To compare methods, all three were implemented on a set of four ^{240}Pu scrap metal samples. All measurements were taken at the Joint Research Center (JRC) laboratory, Ispra, Italy. Measurements were taken using standard PSMC [1] neutron coincidence counter (reported detection efficiency of 50%, detector die-away time 50 μs, calibrated using a ^{252}Cf source). All measurements were analyzed with a gate width of 150 μs (three die away times). A detailed description of the measurements is given in the table below:

Table 1: Description of the Pu samples and measurement times.

Sample	Pu mass [g]	^{240}Pu effective [wt %]	^{240}Pu effective mass [g]	Measurement Time[h]
CBNM61	6.6	24.9	1.6434	6.6
CBNM70	6.6	18.0	1.1880	5.1
CBNM84	6.6	14.1	0.9306	1.8
CBNM93	6.6	6.3	0.4092	16.8

All the measurements were fairly long (in terms of a NMC measurement), so that we can create a reference to which the different methods can be compared. The characteristics of the reference value will be described in the next section.

4.2 Construction of the reference value

To create a point of reference, each of the measurements was broken into sub-measurements of duration T_s, with T_s ranging between 3 to 10 minutes. Meaning, we have divided the total measurement into smaller segments and considered the count separately: for each sub-interval, all first three central moments were evaluated, and the mass was computed. This procedure results in a distribution of all three central moments and the mass. The number of samples for each distribution, of course, depended on both the duration of the total measurement and T_s. For instance, in measurement CBNM93, we took $T_s = 10min$,

[6]Once again, equation 3.7 does not compute the standard deviation of the mass, but rather estimates the propagation in the the confidence interval geometrically from the (U, S, M_L) plane on to the mass

resulting in a total of 96 samples. Then the variance of each of the four distributions sampled was used as the "reference variance".
For instance, figure 2 shows the sampled distribution of the mean, variance, skewness and mass in the measurement of sample CBNM93.

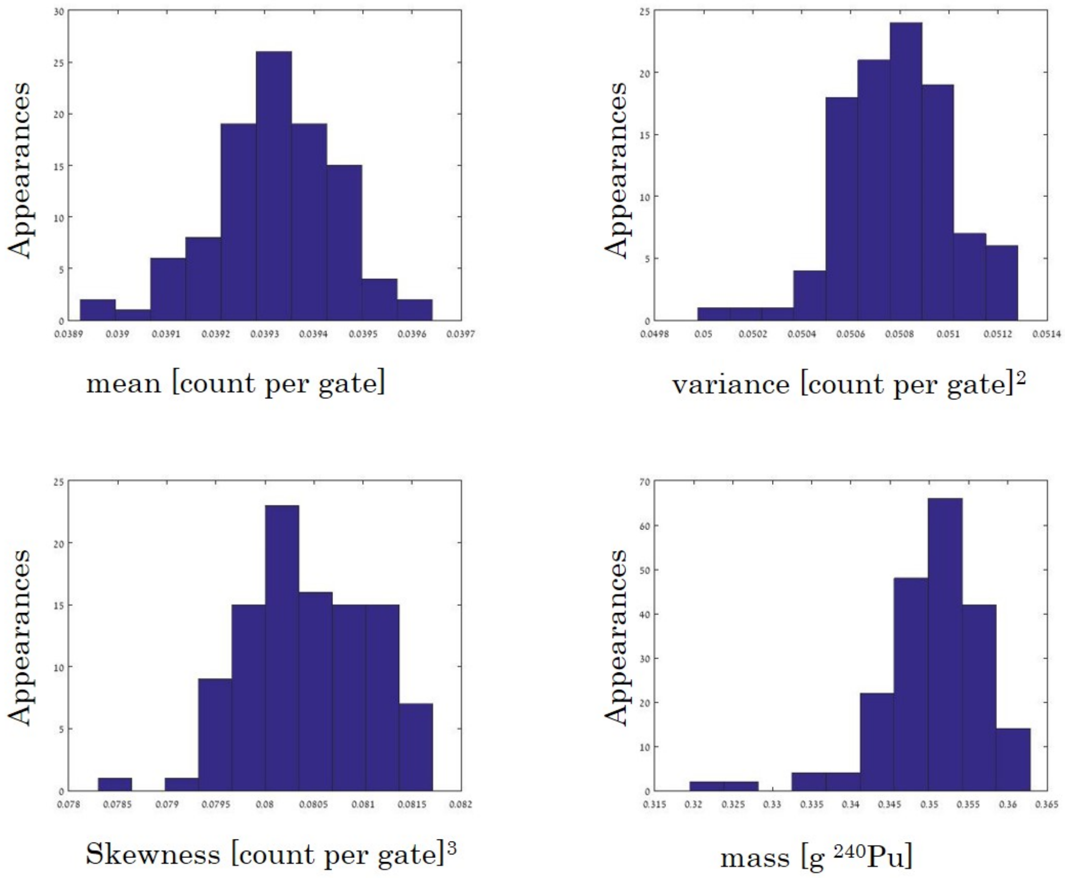

Figure 1: The sampled distribution (on intervals of duration $T_S = 10$ minutes) for the mean, variance, skewness and mass in sample CBNM93.

The construction of the reference value is, in theory, exactly what we have described in method (I). However, because we did not divide in the square root of the number of repetitions, the variance measured is the statistical uncertainty of a single measurement of duration T_s. Therefore, and this is a very important point, from each measurement **only a single sub-interval of duration T_s was analyzed through all three methods (the same sub interval)**. This captures the usual situation, namely that an uncertainty estimate is needed for a single period.

Table 2 below shows the duration T_s and the number of repetitions for each sample:

Table 2: Measurement duration T_s and the number of repetitions for each sample.

Sample	CBNM61	CBNM70	CBNM84	CBNM93
$T_s[min]$	10	10	3	10
number of repetitions	41	32	39	101

To summarize this section, we give three concluding remarks:

1. The reference value for the variance is, in a sense, the best estimate possible. It is explicitly the statistical variance (of both the moments and the mass) of a NMC measurement.

2. The estimated uncertainties evaluated through the different methods does not "see" the entire measurement, only the sub-interval of duration T_s analyzed.

3. The uncertainty on the reference value is not always negligible. Assuming that the sampled variance has a normal distribution, we can approximate the 1σ uncertainty with $1/\sqrt{2(N-1)}$, where N is the number of sub measurements. This gives an 11% error bar for CBNM61, 13% for CBNM 70, 12% for CBNM84 and 7% for CBNM93.

5 Experimental results

5.1 Full implementation on all samples

In this section, the results of a full implementation of all three methods are given. Before the results are described, one technical remark: As described earlier, a full implementation of the method requires a numeric evaluation of the partial derivatives of $F_i(S,U,M_L),(i=1,2,3)$. Theoretically, this can be done be computing the partial derivatives symbolically, and then inserting the computed values of S,U and M_L. To simplify the computation, we computed the derivatives numerically, by taking an incrementation of 0.01% to each of the values, and used the central approximation for the derivative :

$$\frac{df}{dx}|_{x_0} \approx \frac{f(x_0+\delta_x)-f(x_0-\delta_x)}{2\delta_x}$$

The full results, in terms of the statistical uncertainty on each sampled moment (and the mass) are presented in tables 3-6 below:

Table 3: Results for sample CBNM 61

Method	σ_E	σ_{Var}	σ_{Sk}	$\Delta_{mass}[g]$
SACD (I)	2.59×10^{-4}	4.69×10^{-4}	1.5×10^{-3}	0.0122
PVM (II)	2.62×10^{-4}	4.89×10^{-4}	1.5×10^{-3}	0.0180
BS (III)	2.17×10^{-4}	4.23×10^{-4}	1.57×10^{-3}	0.0175
reference	3.04×10^{-4}	5.36×10^{-4}	1.4×10^{-3}	0.0182

Table 4: Results for sample CBNM 70

Method	σ_E	σ_{Var}	σ_{Sk}	$\Delta_{mass}[g]$
SACD (I)	1.85×10^{-4}	3.49×10^{-4}	1.2×10^{-3}	0.0135
PVM (II)	2.19×10^{-4}	4.00×10^{-4}	1.3×10^{-3}	0.0134
BS (III)	2.3×10^{-4}	4.27×10^{-4}	1.2×10^{-3}	0.0138
reference	2.86×10^{-4}	5.45×10^{-4}	1.81×10^{-3}	0.0121

Table 5: Results for sample CBNM 83

Method	σ_E	σ_{Var}	σ_{Sk}	$\Delta_{mass}[g]$
SACD (I)	2.82×10^{-4}	5.11×10^{-4}	1.6×10^{-3}	0.0180
PVM (II)	2.41×10^{-4}	4.69×10^{-4}	1.6×10^{-3}	0.0191
BS (III)	2.36×10^{-4}	4.74×10^{-4}	1.6×10^{-3}	0.0223
reference	2.77×10^{-4}	4.93×10^{-4}	1.46×10^{-3}	0.0201

Table 6: Results for sample CBNM 93

Method	σ_E	σ_{Var}	σ_{Sk}	$\Delta_{mass}[g]$
SACD (I)	1.11×10^{-4}	2.19×10^{-4}	6.52×10^{-4}	0.0060
PVM (II)	0.92×10^{-4}	1.73×10^{-4}	5.74×10^{-4}	0.0061
BS (III)	1.08×10^{-4}	1.95×10^{-4}	6.04×10^{-4}	0.0058
reference	1.29×10^{-4}	0.22×10^{-4}	0.62×10^{-4}	0.0065

The results, in terms of the estimated relative error of the mass for all four samples (and all three methods) are given in table 7 below:

Table 7: relative error of the mass estimation in all three methods

Sample Sample	Reference (repetition)	Method I (SACD)	Method II (PVM)	Method III (BS)
CBNM 61	1%	0.6%	0.9%	0.9%
CBNM 70	1%	0.5%	1.1%	1.1%
CBNM 84	2.6%	2.4%	2.5%	2.8%
CBNM 93	1.9%	1.7%	1.7%	1.7%

From table 7, methods (II) and (III) have similar results, with a maximal error of about 20% in σ_{mass} compared to the reference value (translating into 0.2% in terms of the relative error). Since the uncertainty of the reference value is roughly 10% (see final remark of section 4.2), it is safe to state both methods performed fairly well.

The estimates obtained by the first method are slightly biased, with a maximal error of about 50% (translating to 0.5% in mass). It is worth mentioning that all the estimates obtained using the first method (SACD) are under estimates (with respect to the reference value).

Finally, we have repeated method (I), but now with computing the mass for each sub-measurement, and directly estimating the uncertainty of the mass. For samples CBNM 93 and CBNM 61, results were very similar (0.9% 1.3% respectively). for sample CBNM 70 the uncertainty was estimated by 1.7%, which is considerably larger than the reference value- although all values of M_L were larger than 1. For sample CBNM 84, about 30% of the sub-interval resulted with a value of $M_L < 1$, and the uncertainty estimation was approximately 5%, almost twice as low as the reference value.

5.2 Statistical uncertainty for longer measurements

Although the results so far demonstrate equivalent results between all three methods and the reference values, they all share one short coming: the total measurement time for which the results were presented were in the range of 3-10 minutes- all well below the typical operational measurements. In the relevant masses, a typical measurement would be around 0.5

hours. However, in a 30 min. measurement, we wold need a minimal measurement of 16 hours to create a reference value (estimated with the classic number of 30 samples). Looking at table 2, the only measurement that was long enough was for CBNM93.

Therfore, to estimate the methods in a period of 30 min., a different approach was taken. In general, we expect the the statistical error, as a function of the measurement time T, would be proportional to $1/\sqrt{T}$. We have repeated the statistical uncertainty analysis for all four samples in all three methods, for 20 min. and 30. min. measurements. The results are presented in figures 2 - 5.

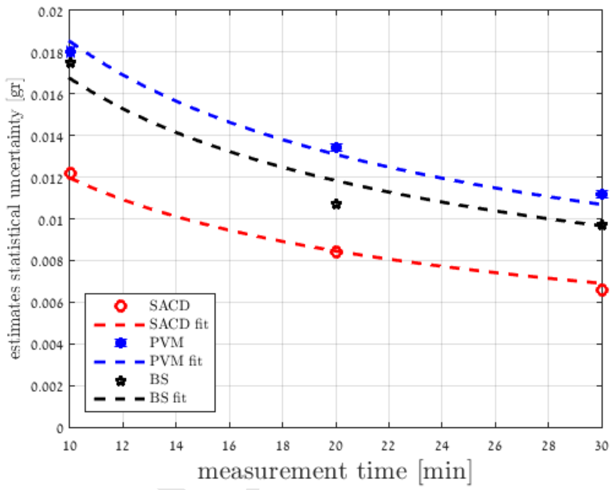

Figure 2: Estimated statistical uncertainty for sample CBNM61.

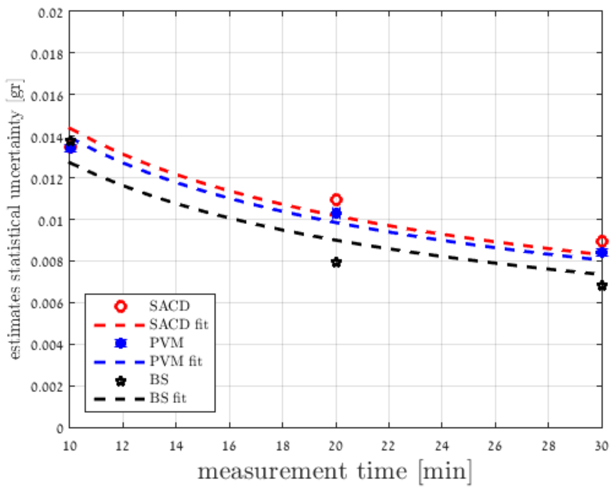

Figure 3: Estimated statistical uncertainty for sample CBNM70.

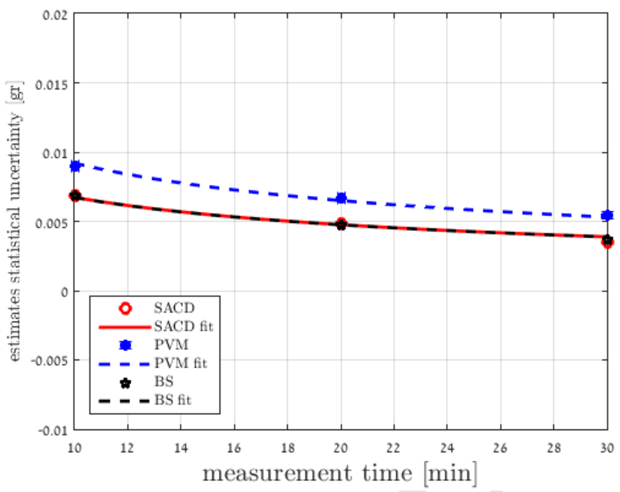

Figure 4: Estimated statistical uncertainty for sample CBNM84.

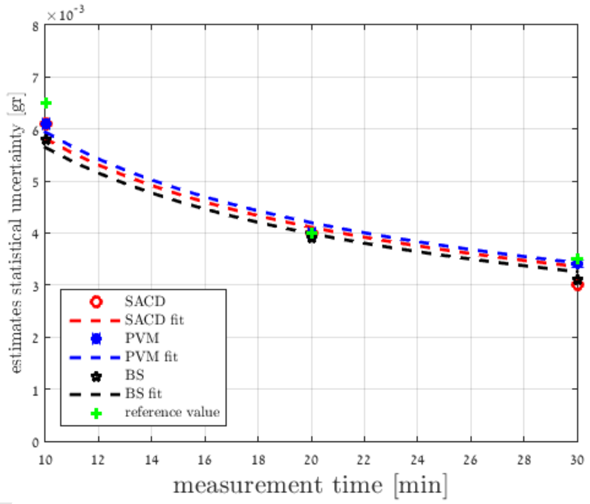

Figure 5: Estimated statistical uncertainty for sample CBNM93.

As we can see, the fit to a $1/\sqrt{T}$ functional form is well within a 10% error, and the discrepancy between the three methods is as in the previous section: all less the 0.4% in the sample mass.

6 Summary and concluding remarks

A comparison among three approaches for estimating the statistical uncertainty in NMC was introduced: The bootstrap method, propagation of variance through moments and statistical analysis of cycle data.

The purpose of the study is to define and validate simple schemes, which, we believe, may serve as guidelines for estimating the statistical uncertainty in the NMC measurement. For that purpose, once the methods were presented, we compared the results using a set of 4 different Pu samples (all with the same geometry an total mass of Pu, differing only in the isotopic composition), measured on a PSMC well counter at the JRC laboratory, in Ispra, Italy. The assay was based on inverting point model equations. The similarity of the measurement items geometry minimizes model bias across the data set, while the differences between the samples are sufficient to allow practical conclusions. The outcome of each method was compared to a reference value, obtained by a brute force measurement of the statistical uncertainty.

As a general note, looking at table 7, we see that all three methods produce good results. The last two resulted in a maximal error of about 20% in σ_{mass} (translating into 0.2% in terms of the relative error), while the uncertainty on the refernce value was about 10% .The maximal error in the SACD is 50% in σ_{mass} (translating to 0.5% in the relative error). . Therefore, the results presented here suggest methods (II) and (III) as operational methods. From a technical view point, all three methods are very simple to implement, and do not demand considerable computation resources. Still, we add a remark on implementation of the bootstrap method: As defined in [13] and [6], implementation of the bootstrap method requires random shuffling with replacement of the original data. As an alternative, we can compute the central moments (or Singles, Doubles and Triples rate), and then shuffle the moments. Computing the moments of the count rate distribution is by far the "bottle neck" from a run time point of view, typically taking of the order of seconds (in the present study, about 10-20 sec. for all samples). While this is acceptable in terms of a single measurement, when repeated 300 times to build the bootstrap distribution, this might accumulate to a long run time. Because in the second method the moments are only computed once (and the run time of the shuffle is negligible), the second method reduces the run time dramatically.

Acknowledgements

This work was sponsored in part by the U.S. Department of Energy (DOE), National Nuclear Security Administration (NNSA), Office of Nonproliferation Research and Development (NA-22).

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A Appendix A: Explicit formulas for the standard deviation of the sampled variance and skewness

In the following section we denote by M_j the j_{th} moment of a distribution:

$$M_j = E(X^j) = \sum_{n=1}^{\infty} p_n n^j$$

Using the above notation, equation 3.3 can be written as:

$$Var(Var(X)) = M_4 - 4M_1M_3 + 8M_1^2M_2 - 4M_1^4 - M_2^2$$

and equation 3.4 as

$$Var(Sk(X)) = M6 - 6M_5M_1 + 15M_4M_1^2 - 20M_3M_1^3 + 15M_2M_1^4 - 5M_1^6 - (M_3 - 3M_2M_1 + 2M_1^3)^2$$

Equivalent terms for the factorial moments are given:

$$Var(X(X-1)) = M_4 - M_1^2 + 2M_1M_2 + (1 + M_1)(M_2 - 2M_3)$$

$$Var(X(X-1)(X-2)) = M_6 - 6M_5 + 13M_4 - (2M_1 - 4M_2 + M_3)^2 - 12M_3 + 4M_2$$

From a sampling point of view, denoting by n_j the number of detection in the j gate, M_j can be sampled in two distinct manners: First, if we denote by f_n the number of gates for which $n_j = n$ detections, then $p_n \equiv f_n / \sum_k f_k$, and then M_j is sampled by

$$M_j \equiv \sum_{n=0}^{\infty} n^j \times \frac{f_n}{\sum_k f_k}$$

This form makes it clear the p_n is not a theoretical probability distribution but rather an experimentally based quantity.
Another option, since $N >> 1$ (here N is the total number of gates), we can estimate M_j via

$$M_j \equiv \frac{1}{N} \sum_{k=1}^{N} n_k^j$$