

Neutron Multiplicity Counting for Future Verification Missions: *Bias When the Sample Configuration Remains Unknown*

Malte Göttsche¹, Gerald Kirchner¹

¹University of Hamburg
Centre for Science and Peace Research (ZNF)
Beim Schlump 83
20144 Hamburg
Germany

Abstract. Passive neutron multiplicity counting to determine plutonium mass is used nowadays *inter alia* in Safeguards applications. As opposed to total neutron rate counting, it can determine plutonium mass also in oxides and samples with induced fission processes if the isotopic composition is known. Neutron multiplicity counting may be helpful for other missions. These may include CBRN response related to nuclear trafficking, and verification of nuclear material, including the nuclear fuel cycle and military stocks under potential future regimes. Sometimes, the exact sample configuration may remain unknown. Despite the technique's clear advantages, limitations require further study. Besides the influence of possible shielding between sample and detector, the assay results are dependent on the configuration, in particular geometry, of the fissile material. Assay bias for highly multiplicative samples has already been studied to some extent which lead to the introduction of corrections to the point model which is usually used to calculate the fissile mass. This paper presents MCNPX-PoliMi simulation results of different plutonium sample configurations to critically evaluate eventual bias occurring with the point model. In addition, correction methods will be evaluated.

1. Introduction

Neutron multiplicity counting is a technique that is frequently used for Safeguards purposes. When the isotopic composition of plutonium is known, it measures the plutonium mass. It performs very well in environments where the detector has been fully characterized and where the configuration of plutonium samples is largely known (in particular the geometry and density of the sample and the shielding between sample and detector) so that the detector can be calibrated with samples that have a similar configuration as the measured sample. This is usually the case for Safeguards applications.

In cases where the sample configuration remains, however, largely unknown, potential bias in the plutonium mass assessment should be studied. This could be the case for CBRN response related to nuclear trafficking, and verification of military nuclear material, including warheads, as part of a future arms control verification regime or a Fissile Material (Cutoff) Treaty. In the case of CBRN response, assessments of suspicious items must be performed quickly, which precludes the possibility of a detailed analysis of the item configuration. For military material and warhead verification, the exact configuration of the item will most likely not be known to an inspector, due to the sensitive nature of this information [1].

In this paper, plutonium mass bias will be evaluated for samples with a high neutron multiplication, for example weapons-grade plutonium samples of high mass. It shows how bias can be removed when the sample configuration is roughly known with the help of Monte Carlo simulations, which is, however, not possible in scenarios where knowledge on the configuration remains unknown. In such scenarios, an understanding of the mass bias is

important to understand the uncertainty of the information and correspondingly the confidence that can be gained from such a measurement. This paper is based on simulations with the MCNPX-PoliMi code [2].

2. Methods for bias studies

Neutron multiplicity counting measures the plutonium mass by counting the neutrons that are detected within a defined gate length after a neutron trigger. The measured quantities are the Singles, Doubles and Triples rates S , D and T , which are the neutron detection rate and the rates of coincidences between two and three correlated neutrons. Using S , D and T , this method allows a discrimination of neutrons from spontaneous fission, induced fission and (α, n) reactions in oxides. It is based on a derivation by Böhnel [3] which inter alia makes the assumption that the induced fission rate is constant regardless of where the original spontaneous fission occurred (“point model”). The neutron multiplicity counting output is the neutron multiplication M (a measure to describe the amount of induced fission neutrons), α (the (α, n) to spontaneous fission ratio) and m (the plutonium fissile mass which is obtained from measuring the amount of spontaneous fission). To be able to deduce M , α and m , the detector efficiency ε must be known as well as the doubles and triples gate fractions f_d and f_t , which describe the ratio of Doubles and Triples that are detected within the finite gate length compared to those that remain undetected (as correlated neutrons may arrive after the gate has been closed). In addition, nuclear multiplicity data is used, for example the moments of the induced fission multiplicity distribution ν_i (for details see [4]). In the case of simulations, a Monte Carlo code such as MCNPX-PoliMi produces S , D and T (“simulation”). Then, M , α and m can be calculated (“multiplicity analysis”).

The overall bias that may occur in neutron multiplicity counting can be separated into two components: Bias related to the transport of neutrons from the sample to the detector and their detection on the one hand and bias related to the neutron production of the fissile sample on the other hand. Examples of the former include shielding between sample and detector which influences the detection efficiency by capturing neutrons or changing the neutron energy distribution [5]. The latter refers to processes that occur directly in the fissile material. In the following sections, bias related to the neutron production and interactions within the fissile material are addressed. In this section, some methods that will be used in the subsequent sections to study bias are introduced.

2.1. *Detector*

To understand specific effects, they must be studied in isolation from the others. To exclude shielding and detection issues, ε , f_d and f_t may not vary. Furthermore, neutron reflection from the detector to the sample should be minimized.

This is achieved by simulating an “idealistic detector”. We assume a hollow sphere consisting of He-3 at a density of 10 g/cm^3 . The inner and outer radii are set to almost infinity (20 m and 40 m). He-3 both moderates the neutrons and subsequently captures them. Because of the high density and the large volume, the detector has an efficiency $\varepsilon = 1$. All neutrons are captured in the detector and all capture events are detection events. This has been proven by MCNPX-PoliMi simulations which show that no neutrons escape the detector in the outward direction. The gate fractions f_d and f_t both equal unity when the simulated gate length is

sufficiently long to count all coincidences. Reflection of neutrons from the detector back to the sample is minimized due to the large detector cavity.

2.2. *Consistent nuclear multiplicity data*

The nuclear multiplicity data required as input for the multiplicity analysis must be carefully chosen to ensure that it corresponds to the MCNPX-PoliMi simulation so that no systematic uncertainty is introduced: Usually, the induced fission moments are treated as constants in neutron multiplicity counting. The default values are those of Pu-239 at 2 MeV, irrespective of the actual neutron energy spectrum in the sample [6]. As the induced fission moments depend on the energy of the incoming neutron [7], the average moments should, however, be deduced from the fission neutron spectrum. The average induced fission moments are calculated according to

$$\overline{\nu_{ij}} = \int \nu_{ij}(E) \cdot W(E) \cdot w_{\sigma}(E) dE$$

This is a weighted average of the j -th factorial moment of the induced fission multiplicity distribution $\nu_{ij}(E)$ over two parameters: First, the fission neutron spectrum $W(E)$, corresponding to the fraction of neutrons in the sample with the energy E and second, the probability of these neutrons to induce fissions $w_{\sigma}(E)$, the fission cross-section normalized to unity. The result is $\nu_{i1} = 3.209$; $\nu_{i2} = 8.571$; $\nu_{i3} = 18.740$. In this calculation, it has been assumed that no neutron moderation takes place in the sample and that induced fission only occurs in Pu-239. These assumptions are reasonable but introduce a small systematic uncertainty.

2.3. *Simulation uncertainties*

As long as all parameters remain identical in the analysis and the simulations, their choice does not introduce any bias.¹ This leaves statistical uncertainties from the simulation and the small systematic uncertainty from the multiplicity data. To test this for the idealistic detector, the nuclear multiplicity data and the simulation uncertainties, a solid sphere of 20 g plutonium (94% Pu-239, 6% Pu-240, $\rho = 19.8 \text{ g/cm}^3$) placed in the centre of the idealistic detector has been simulated (simulated measurement time 3600 s). The comparison between the real data and the multiplicity analysis output is found in Table 1. Statistical uncertainties are given. As a metal was simulated, the multiplicity analysis correctly determined $\alpha = 0$. The deviations between real values and multiplicity analysis remain acceptably small; the deviation for the multiplication lies slightly outside the statistical uncertainty which can be attributed to the multiplicity data uncertainty. The systematic multiplicity uncertainty is small and for our purposes negligible. The data does not indicate additional systematic uncertainties. Without adapting the usually used neutron multiplicity data, the fissile mass is overestimated by around 3%, as simulations show.

3. **Bias from highly multiplicative samples**

As a first bias study, unreflected fissile material in a configuration of high multiplication is investigated. Simulations of solid spheres (configuration as above, but varying masses and correspondingly radii) and hollow spheres (same configuration as above, but inner radius of 3.5 cm and varying masses and correspondingly outer radii) were performed with the multiplicity analysis, the results are shown in Table 2. In all simulations, $\alpha \cong 0$.

¹ Once a simulation is compared to a measurement, however, the uncertainty must be included.

	Multiplication	Fissile mass
True values	1.0996	20.00 g
Multiplicity analysis	1.0988 ± 0.0004	20.02 ± 0.09 g
Deviation	-0.07 ± 0.04 %	0.10 ± 0.45 %

Table 1: Comparison of the multiplicity analysis to the true data for a 20 g weapon-grade plutonium metal solid sphere

	M_{true}	m_{true} [g]	M_{mult}	m_{mult} [g]	ΔM [%]	Δm [%]
Solid spheres	1.1887	100	1.1841 ± 0.0004	95.66 ± 0.44	-0.39 ± 0.03	-4.3 ± 0.5
	1.3997	500	1.4235 ± 0.0005	469.4 ± 2.4	1.70 ± 0.03	-6.1 ± 0.6
	1.5873	1000	1.6343 ± 0.0006	889.9 ± 5.0	2.96 ± 0.04	-11.0 ± 0.6
Hollow spheres	1.1575	1000	1.1571 ± 0.0004	1001.0 ± 4.1	-0.03 ± 0.03	0.1 ± 0.4
	1.5955	4000	1.6115 ± 0.0006	3851 ± 20	1.00 ± 0.04	-3.7 ± 0.5
	1.9577	6000	1.9966 ± 0.0006	5541 ± 32	1.99 ± 0.03	-7.7 ± 0.6

Table 2: Multiplicity analysis results of MCNPX-PoliMi simulations, the true simulated values and the deviations of the simulated from the true values

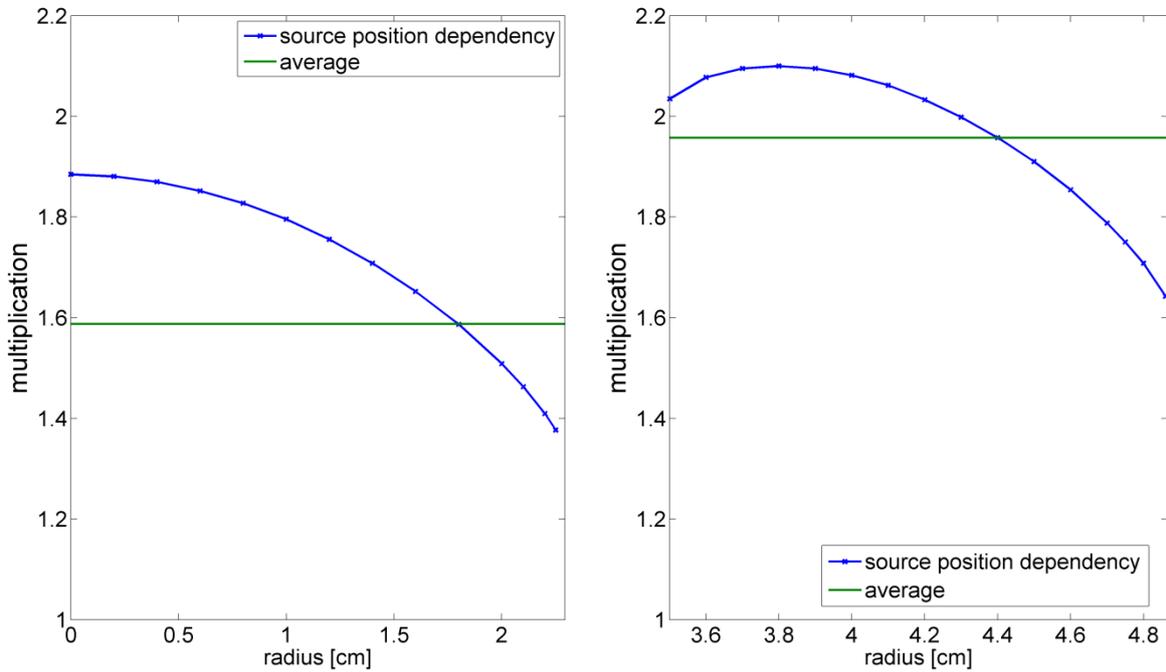


Fig. 1: Multiplication as a function of spontaneous fission source radius (blue) and the average multiplication from spontaneous fission over the full fissile material volume. Left is the 1000 g solid sphere, right is the 6000 g hollow sphere case.

In both simulation series, the multiplication increases with sample mass as is expected. For the hollow spheres, the same multiplication values require larger masses than for the solid spheres, which is also expected. For small multiplications, the multiplicity analysis results agree with the true values either within their statistical uncertainties or are slightly larger (see also Table 1), which can be explained by the uncertainties on the multiplicity data as discussed above. However, as mass and multiplication become larger, the multiplication is overestimated by as much as 3% and the mass underestimated by as much as 11%. Both deviations cannot be explained by the statistical uncertainties and increase with increasing mass/multiplication. This behavior alludes to a systematic error.

3.1. *Spatially variant multiplication*

To identify the systematic error, this section takes a closer look at the physics in the fissile material. The 1000 g solid sphere and 6000 g hollow sphere are analyzed in more detail as both have a fairly high mass. Two different geometries are chosen to find similarities or differences in the related physics.

We look at the multiplication process of spontaneous fission neutrons emitted at specific locations within the sample $M(\vec{r})$. For the discussed samples, we have simulated spontaneous fission from sphere surfaces of different radii that are located within the spheres, see Fig. 1. Each data point corresponds to one MCNPX-PoliMi simulation. This clearly demonstrates that multiplication depends on the position from which a spontaneous fission neutron was started to initiate the multiplication process, contrary to the “point model” assumption. From the spatially dependent multiplication $M(\vec{r})$, the sample's average multiplication can be calculated by appropriate weighing

$$\langle M \rangle = \frac{1}{V} \int M(\vec{r}) dV$$

3.2. *Existing correction model*

There has been some success in applying correction factors to the multiplicity analysis that depend on the measured multiplication: Krick et al. [8] ran MCNPX simulations with plutonium cylinders of different height/diameter ratios and masses. For each simulation, the multiplicity analysis was performed with results deviating from the true values. Then, for each simulation, separate S, D and T correction factors were empirically determined for all simulations to get the correct values. It was found that the correction factors can be given as a function of multiplication, independent of fissile mass. As these correction factors are determined empirically without a full theoretical understanding, it is difficult to generalize this approach, despite its specific success.

Croft et al. [9] proposed another model coupled with the desired physical understanding: In multiplicity analysis equations, the multiplication appears up to the fifth order ($n = 5$). According to the “point model”, assuming constant multiplication at all spontaneous fission locations, $M^n = \langle M \rangle^n$. The physically correct equation would however be $M^n = \langle M^n \rangle$, and $\langle M^n \rangle = \langle M \rangle^n$ is indeed only true when the assumption of constant multiplication is correct, which is a questionable assumption in the presented cases. Therefore, Croft et al. propose correction factors [9]

$$g_n = \frac{\langle M^n \rangle}{\langle M \rangle^n}, \quad n = 2 \dots 5$$

They are calculated from

$$\langle M^n \rangle = \frac{1}{V} \int M^n(\vec{r}) dV$$

We have solved the adapted multiplicity equations for M , α and m . Then, we successfully tested the result by taking measurement data of plutonium samples with little multiplication and setting $g_n = 1$. The result is identical to the multiplicity analysis based on the “point model”.

3.3. Correction procedure and results

To calculate $\langle M^n \rangle$, Croft et al. [9] propose to use a polynomial fit for the simulated data points (spontaneous fission surfaces) in order to integrate $M^n(\vec{r})$. We have tested this approach and find that a polynomial fit is very volatile: For an acceptable fit, we find that the fit function must be a third order polynomial; in this case, however, slight changes to the data points can have dramatic effects on the integral of the polynomial fit. Therefore, we interpolate between the individual data points using a linear fit. The results of the integration are found in Table 3. The fitting process naturally introduces an uncertainty which in this case is estimated to be less than 2% in all cases as the data points appear to be sufficiently close and the function is expected to be smooth without outliers.

With these results, a corrected multiplicity analysis can be performed. The final results are given in Table 4. The uncertainties are statistical uncertainties as above. All values lie slightly outside their statistical uncertainty, but are a significant improvement compared to Table 2. The deviations are small enough to be reasonably explained by the interpolation uncertainty and the multiplicity data uncertainty. The improved methodology reduces mass deviations of -11.0% and -7.7% to 2.3% and 1.2%. This is an indication that spatially variant multiplication is indeed the main reason for the high deviations of the point model analysis.

	1000 g solid sphere	6000 g hollow sphere
$\langle M^1 \rangle$	1.5878	1.9566
g_2	1.0068	1.0048
g_3	1.0215	1.0142
g_4	1.0441	1.0280
g_5	1.0751	1.0461

Table 3: Multiplication and correction factors determined from integrating the interpolated data.

	M_{corr}	ΔM	m_{corr}	Δm
1000 g solid sphere	1.5902 ± 0.0005	0.2%	1023 ± 5	2.3%
6000 g hollow sphere	1.9627 ± 0.0006	0.3%	6074 ± 33	1.2%

Table 4: Results of the corrected multiplicity analysis and the deviations to M_{true} and m_{true}

Our approach solves the issue when it is possible to simulate samples of similar configurations to obtain correction factors that can be applied for specific measurements. However, the problem remains for scenarios where knowledge on the sample configuration is scarce. Comparing the solid and hollow spheres in Table 2, one can see that the deviation is neither a simple function of multiplication (as assumed by Krick et al. [8]), nor mass (which Croft et al. propose [9]). It depends on the specific function $M(\vec{r})$, as documented in Fig. 1. Considering the cases of CBRN response and military materials and warhead verification, without having any information on configurations, bias can be expected to be large. As information will remain incomplete due to the protection of sensitive information, a robust uncertainty assessment is strongly advised.

4. Conclusions

Neutron multiplicity counting is routinely used in Safeguards. There are further potential applications, including CBRN response as well as verification of military materials and warheads as part of a future arms control verification regime or a Fissile Material (Cutoff) Treaty. Measured plutonium masses are significantly underestimated when the measured samples are highly multiplicative. This is due to the neutron production by induced fission within the material. Further systematic uncertainties arise from the transport of neutrons from the sample to the detector and their detection.

This paper has demonstrated that the reason of this bias is the “point model assumption” which is seriously violated for highly multiplicative samples: Our solution to the corrected multiplicity analysis equations removes the bias.

To remove the bias in Safeguards applications, Monte Carlo simulations must be used to obtaining correction factors. This is possible for many Safeguards applications, but not for CBRN response and military material and warhead verification. In these cases, having a very rough knowledge on item configuration may still help to gain some understanding on plausible correction factors. It remains crucial to include this bias in an overall uncertainty assessment for verification scenarios to determine the amount of confidence that can be gained from the measurement.

5. Acknowledgements

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