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Spatial Multiplication Model as an alternative to the Point Model in Neutron Multiplicity Counting

Danielle K. Hauck, Vlad Henzl

LA-UR-#####

EXECUTIVE SUMMARY

The point model is commonly used in neutron multiplicity counting to relate the correlated neutron detection rates (singles, doubles, triples) to item properties (mass, (α,n) reaction rate and neutron multiplication). The point model assumes that the probability that a neutron will induce fission is a constant across the physical extent of the item. However, in reality, neutrons near the center of an item have a greater probability of inducing fission than items near the edges. As a result, the neutron multiplication has a spatial distribution.

The singles rate is a function of the average neutron multiplication and is minimally affected by the spatial distribution. However, the higher order detection rates (doubles, triples, etc.) are functions of the neutron multiplication raised to an integer power, which is estimated in the point model by raising the average neutron multiplication to a power. However, due to the spatial variation, the average value of the neutron multiplication raised to a power is **not** equal to the average of the neutron multiplication distribution raised to a power.

The approach described here assumes that the neutron multiplication has a specified spatial distribution. The spatial distribution is raised to integer powers then averaged. The resulting values are placed into the existing point model equations in place of the traditional neutron multiplication terms. The spatial multiplication model is derived here for the singles, doubles, triples, quadruples and pentuplets.

Two parameters are required to describe the spatial distributions explored in this paper: M_L (the average leakage multiplication which is traditionally used in the point model) and M_R , a new parameter used to describe the radial extent of the curve in question. One way of implementing the spatial multiplication model is to use the singles, doubles, triples and quadruples values to solve for 4 unknowns: spontaneous fission rate, (α,n) reaction rate, average neutron multiplication (M_L) and the spatial multiplication extent (M_R). This approach resulted in improvements on the order of a factor of 1/3 better accuracy in the assayed mass of Pu-240 for leakage multiplications up to 2.5. The degree of improvement was determined from a series of simulations of Pu metal items with a range of masses, densities and shapes.

It is also possible to predict the spatial extent of the leakage multiplication curve (M_R) based on the physical relationship between M_R and the average leakage multiplication (M_L). In this paper this approach is referred to as the known M_R spatial multiplication model. The higher the leakage multiplication, the more “curvy” the multiplication spatial distribution, and the larger is M_R . The relationship between M_R and M_L was quantified from the same series of simulations that was used for testing. The spatial model was then applied using the singles, doubles and triples rates to find the three

traditional unknowns (mass, (α,n) reaction rate and neutron multiplication). Figure 1 shows the improvement in the assayed mass value up to an average leakage multiplication of 2.5 by using the known M_R spatial multiplication model.

Figure 1. Accuracy in assayed Pu-240 mass from the known M_R spatial multiplication model compared to accuracy from the point model. The spatial extent parameter (M_R) was treated as a known value determined from correlation with the average leakage multiplication. The errors in both models increase for average leakage multiplications greater than 2.5.

For the results shown in Figure 1, the M_R vs M_L curve was based on the first five detected multiplicities (singles, doubles, triples, quadruples and pentuplets) in a series of simulations of Pu metal cylinders. The results were best when the relationship between M_R and M_L was found from as many detected multiplicity rates as possible (up to pentuplets in this paper). While the results are highly promising, there are several issues that will be further evaluated in future work:

1. Test the model for items that have (α,n) reactions (Note that the model works well for Pu metal items in which the alpha ratio is treated as an unknown).
2. Determine if the shape of the multiplication curve is dependent on the neutron detector (it is expected not to be).
3. Improve the M_R vs M_L correlation by including higher order terms from sextuplets or hexuplets (important for correcting large highly multiplying items).
4. Use simulations to gain an improved description of the actual spatial multiplication curve in different items.
5. Further explore the analogy with reactor physics to extend and improve the multiplication model.

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1. INTRODUCTION

The point model is commonly used in neutron multiplicity counting to relate the correlated neutron detection rates (singles, doubles, and triples) to item properties (spontaneous fission rate, (α, n) reaction rate and neutron multiplication). In conventional safeguards measurements the spontaneous fission rate is used to deduce the mass of the material of interest, usually plutonium.

The point model has several associated assumptions, including that the probability that a neutron will induce fission is a constant across the physical extent of the item. Another way of stating this assumption is that the neutron multiplication is constant across the item. However, in reality, neutrons born near the center of an item have a greater probability of inducing fission than neutrons born near the edges. As a result, the neutron multiplication has a spatial distribution.

According to the point model, the singles (d_1), doubles (d_2), triples (d_3), quadruples (d_4) and pentuplets (d_5) are functions of the neutron leakage multiplication raised to various integer powers (M_L^n) where n ranges from 1 to 9. In the conventional point model, the average neutron multiplication is raised to integer powers ($\langle M_L \rangle^n$). A spatial distribution of neutron multiplication can be incorporated into the point model equations by reinterpreting the M_L^n terms as the average of the spatial distribution raised to the power n ($\langle M_L^n(r) \rangle$), where r is used to represent spatial location in the item.

As shown in Figure 2, using the powers of the average neutron multiplication underestimates the true neutron multiplication raised to a power. The result in practical terms is that the point model correctly predicts this singles rate, but underestimates the doubles and triples rates as well as the higher order multiplicity rates. The amount by which the doubles and triples are underestimated is directly related to the neutron multiplication of the item. The higher order detection rates (e.g. triples) are more severely underestimated than lower order detection rates (e.g. doubles).

Figure 2. The effects of the spatial distribution of the neutron multiplication on the average value of the neutron multiplication raised to a power.

In neutron multiplicity counting the point model equations are inverted to find the spontaneous fission rate, (α, n) reaction rate and the neutron multiplication. Since the equations underestimate the doubles and triples rates, the result of inversion is an inflated (α, n) reaction rate to explain the fact that the

singles rate is inflated compared to the doubles and triples, and an underestimated mass and neutron multiplication to explain the fact that the doubles are inflated with respect to the triples [1].

The approach used here is to assume a spatial distribution of the neutron multiplication and to replace the $\langle M_L \rangle^n$ terms in the point model (the neutron multiplication average raised to a power) with $\langle M_L^n \rangle$ terms which represent the spatial distribution of neutron multiplication raised to a power then averaged. Previous work recognized this effect of the spatial distribution of the neutron multiplication on the analysis results [2]. The approach taken in this previous work was to use rigorous simulations to determine weights on the $\langle M_L \rangle^n$ terms in the point model. Here, we take a more analytical approach of identifying the shape of the multiplication curve.

As described below, the model is tested using simulated data with dead-time free, infinite gate detection rates. In other words, the singles, doubles, triples, rates etc. are free of any effects of gating or dead time. This was done on purpose to study the effects of the analysis model without the additional challenges of determining the correct detection rates. Dead time and gating are being treated in different portions of work, and will be integrated in future work.

2. THE POINT MODEL AND SPATIAL MULTIPLICATION MODEL EQUATIONS

This section describes how the point model equations can be reinterpreted to take into account the spatial distribution of the neutron multiplication in the item.

The point model equations are derived and extended to pentuplets in a separate paper [3]. The same notation is used here. According to the point model, the exact equations for the detection multiplicity rates as a function of item parameters are:

$$d_m = \varepsilon^m (S_\alpha \overline{\mathcal{V}_{\alpha m}} + S_f \overline{\mathcal{V}_{sm}}) \quad \text{Eq. 1}$$

where ε is the detection efficiency, S_α is the rate at which (α, n) reactions are occurring in the item, and S_f is the rate of spontaneous fission. The leakage multiplicity rates, or the average number of multiplets of order m leaked from fission chains induced by (α, n) neutrons and spontaneous fission neutrons, are given by $\overline{\mathcal{V}_{\alpha m}}$ and $\overline{\mathcal{V}_{sm}}$, respectively. For multiplets of order one the notation is simplified: $\overline{\mathcal{V}_{\alpha 1}} \equiv \overline{\mathcal{V}_\alpha}$ and $\overline{\mathcal{V}_{s1}} \equiv \overline{\mathcal{V}_s}$. The leakage multiplicity rates for singles, doubles, triples, quadruples and pentuplets for point model assumptions are given by:

$$\overline{\mathcal{V}_\alpha} = M_L \quad \text{Eq. 2}$$

$$\overline{\mathcal{V}_s} = \bar{v}_s M_L \quad \text{Eq. 3}$$

$$\overline{\mathcal{V}_{\alpha 2}} = \bar{v}_{l2} p M_T M_L^2 \quad \text{Eq. 4}$$

$$\overline{\mathcal{V}_{s2}} = (\bar{v}_{s2} + \bar{v}_s p M_T \bar{v}_{l2}) M_L^2 \quad \text{Eq. 5}$$

$$\overline{\mathcal{V}_{\alpha 3}} = (\bar{v}_{3l} p M_T + 2 \bar{v}_{l2}^2 p^2 M_T^2) M_L^3 \quad \text{Eq. 6}$$

$$\overline{\mathcal{V}_{s3}} = (\bar{v}_{s3} + (2 \bar{v}_{s2} \bar{v}_{2l} + \bar{v}_s \bar{v}_{3l}) p M_T + 2 \bar{v}_s \bar{v}_{l2}^2 p^2 M_T^2) M_L^3 \quad \text{Eq. 7}$$

$$\overline{\mathcal{V}_{\alpha 4}} = (5 \bar{v}_{l2}^3 p^3 M_T^3 + 5 \bar{v}_{l2} \bar{v}_{l3} p^2 M_T^2 + \bar{v}_{l4} p M_T) M_L^4 \quad \text{Eq. 8}$$

$$\overline{\mathcal{V}_{s4}} = (\bar{v}_{s4} + (\bar{v}_s \bar{v}_{I4} + 2\bar{v}_{s2} \bar{v}_{I3} + 3\bar{v}_{s3} \bar{v}_{I2})pM_T + (5\bar{v}_{s2} v_{I2}^2 + 5\bar{v}_s \bar{v}_{I2} \bar{v}_{I3})p^2 M_T^2 + 5\bar{v}_s v_{I2}^3 p^3 M_T^3)M_L^4 \quad \text{Eq. 9}$$

$$\overline{\mathcal{V}_{\alpha 5}} = (14v_{I2}^4 p^4 M_T^4 + 21v_{I2}^2 \bar{v}_{I3} p^3 M_T^3 + (6\bar{v}_{I2} \bar{v}_{I4} + 3v_{I3}^2)p^2 M_T^2 + \bar{v}_{I5} p M_T)M_L^5 \quad \text{Eq. 10}$$

$$\begin{aligned} \overline{\mathcal{V}_{s5}} = & (+\bar{v}_{s5} + (\bar{v}_s \bar{v}_{I5} + 2\bar{v}_{s2} \bar{v}_{I4} + 3\bar{v}_{s3} \bar{v}_{I3} + 4\bar{v}_{s4} \bar{v}_{I2})pM_T \\ & + (6\bar{v}_s \bar{v}_{I2} \bar{v}_{I4} + 3\bar{v}_s v_{I3}^2 + 12\bar{v}_{s2} \bar{v}_{I2} \bar{v}_{I3} + 9\bar{v}_{s3} v_{I2}^2)p^2 M_T^2 + (21\bar{v}_s v_{I2}^2 \bar{v}_{I3} + 14\bar{v}_{s2} v_{I2}^3)p^3 M_T^3 \\ & + 14\bar{v}_s v_{I2}^4 p^4 M_T^4)M_L^5 \end{aligned} \quad \text{Eq. 11}$$

Where the spontaneous fission and induced fission emission probabilities ($\bar{v}_{In}, \bar{v}_{sn}$) are given as reduced factorial moments. Note that these are conventionally given as the standard factorial moments. The equations are modified appropriately to account for difference in definition.

The total multiplication, given by

$$M_T = \frac{1}{1 - p\bar{v}_I} \quad \text{Eq. 12}$$

is the total number of neutrons emitted in the item over all generations in a fission chain, resulting from a single source neutron. The leakage multiplication, given by

$$M_L = (1 - p)M_T = \frac{1 - p}{1 - p\bar{v}_I} \quad \text{Eq. 13}$$

is the average number of neutrons leaked from the item per source neutron and its resulting fission chain.

The term pM_T can be written instead as $pM_T = (M_L - 1)/(\bar{v}_I - 1)$ to retrieve the familiar form of the point model equations. By redistributing the leakage multiplication term (M_L), The leakage multiplicity rates can also be rewritten as

$$\overline{\mathcal{V}_\alpha} = M_L \quad \text{Eq. 14}$$

$$\overline{\mathcal{V}_s} = \bar{v}_s M_L \quad \text{Eq. 15}$$

$$\overline{\mathcal{V}_{\alpha 2}} = \frac{\bar{v}_{I2}}{(\bar{v}_I - 1)} [M_L^3 - M_L^2] \quad \text{Eq. 16}$$

$$\overline{\mathcal{V}_{s2}} = \bar{v}_{s2} M_L^2 + \frac{\bar{v}_s \bar{v}_{I2}}{(\bar{v}_I - 1)} [M_L^3 - M_L^2] \quad \text{Eq. 17}$$

$$\overline{\mathcal{V}_{\alpha 3}} = \frac{\bar{v}_{I3}}{(\bar{v}_I - 1)} [M_L^4 - M_L^3] + 3 \frac{\bar{v}_{I2}^2}{(\bar{v}_I - 1)^2} [M_L^5 - 2M_L^4 + M_L^3] \quad \text{Eq. 18}$$

$$\overline{\mathcal{V}_{s3}} = \bar{v}_{s3} M_L^3 + \frac{(3\bar{v}_{s2} \bar{v}_{I2} + \bar{v}_s \bar{v}_{I3})}{(\bar{v}_I - 1)} [M_L^4 - M_L^3] + 3 \frac{\bar{v}_s \bar{v}_{I2}^2}{(\bar{v}_I - 1)^2} [M_L^5 - 2M_L^4 + M_L^3] \quad \text{Eq. 19}$$

$$\overline{\mathcal{V}_{\alpha 4}} = \frac{\bar{v}_{I4}}{(\bar{v}_I - 1)} [M_L^5 - M_L^4] + 10 \frac{\bar{v}_{I2} \bar{v}_{I3}}{(\bar{v}_I - 1)^2} [M_L^6 - 2M_L^5 + M_L^4] + 15 \frac{v_{I2}^3}{(\bar{v}_I - 1)^3} [M_L^7 - 3M_L^6 + 3M_L^5 - M_L^4] \quad \text{Eq. 20}$$

$$\begin{aligned}\overline{\mathcal{V}_{s4}} = & \bar{v}_{s4}M_L^4 + \frac{(\bar{v}_s\bar{v}_{I4}+2\bar{v}_{s2}\bar{v}_{I3}+3\bar{v}_{s3}\bar{v}_{I2})}{(\bar{v}_I-1)}[M_L^5 - M_L^4] + \frac{(5\bar{v}_{s2}\bar{v}_{I2}^2+5\bar{v}_s\bar{v}_{I2}\bar{v}_{I3})}{(\bar{v}_I-1)^2}[M_L^6 - 2M_L^5 + M_L^4] \\ & + 5\frac{\bar{v}_s\bar{v}_{I2}^3}{(\bar{v}_I-1)^3}[M_L^7 - 3M_L^6 + 3M_L^5 - M_L^4]\end{aligned}$$

Eq. 21

$$\begin{aligned}\overline{\mathcal{V}_{\alpha 5}} = & \frac{\bar{v}_{I5}}{(\bar{v}_I-1)}[M_L^6 - M_L^5] + \frac{(6\bar{v}_{I2}\bar{v}_{I4}+3\bar{v}_{I3}^2)}{(\bar{v}_I-1)^2}[M_L^7 - 2M_L^6 + M_L^5] + 21\frac{\bar{v}_{I2}^2\bar{v}_{I3}}{(\bar{v}_I-1)^3}[M_L^8 - 3M_L^7 + 3M_L^6 - M_L^5] \\ & + 14\frac{\bar{v}_{I2}^4}{(\bar{v}_I-1)^4}[M_L^9 - 4M_L^8 + 6M_L^7 - 4M_L^6 + M_L^5]\end{aligned}$$

Eq. 22

$$\begin{aligned}\overline{\mathcal{V}_{s5}} = & \bar{v}_{s5}M_L^5 + \frac{(\bar{v}_s\bar{v}_{I5}+2\bar{v}_{s2}\bar{v}_{I4}+3\bar{v}_{s3}\bar{v}_{I3}+4\bar{v}_{s4}\bar{v}_{I2})}{(\bar{v}_I-1)}[M_L^6 - M_L^5] \\ & + \frac{(6\bar{v}_s\bar{v}_{I2}\bar{v}_{I4}+3\bar{v}_s\bar{v}_{I3}^2+12\bar{v}_{s2}\bar{v}_{I2}\bar{v}_{I3}+9\bar{v}_{s3}\bar{v}_{I2}^2)}{(\bar{v}_I-1)^2}[M_L^7 - 2M_L^6 + M_L^5] \\ & + \frac{(21\bar{v}_s\bar{v}_{I2}^2\bar{v}_{I3}+14\bar{v}_{s2}\bar{v}_{I2}^3)}{(\bar{v}_I-1)^3}[M_L^8 - 3M_L^7 + 3M_L^6 - M_L^5] \\ & + 14\frac{\bar{v}_s\bar{v}_{I2}^4}{(\bar{v}_I-1)^4}[M_L^9 - 4M_L^8 + 6M_L^7 - 4M_L^6 + M_L^5]\end{aligned}$$

Eq. 23

The basic concept of the spatial multiplication model is to assume a spatial distribution of the leakage multiplication ($M_L(r)$). The spatial distribution is raised to the n th power. The new distribution is then averaged and used in equations 14-23 in place of the average M_L raised to a power as in the point model.

3. SIMULATIONS FOR TESTING THE SPATIAL MULTIPLICATION MODEL

Several simulations were performed for understanding and evaluation the spatial multiplication model. All simulations are for Pu metal cylinders in the Active Well Coincidence Counter (AWCC) [4]. In all cases the isotopic content of the Pu metal cylinders were 10% (atomic) Pu-240 with the remainder made up of Pu-239. The MCNPx output files provide for each simulation:

- the total number of spontaneous fissions (N_{SF}),
- the total number of neutrons originating from spontaneous fission ($N_{SFneutrons}$),
- the reduced factorial moments for the number of correlated neutrons emitted from each spontaneous fission ($\bar{v}_s, \bar{v}_{s2}, \bar{v}_{s3}, \dots$),
- the total number of induced fissions (N_{IF}),
- the total number of neutrons originated from induced fission ($N_{IFneutrons}$),
- the reduced factorial moments for the number of correlated neutron emitted from each induced fission ($\bar{v}_I, \bar{v}_{I2}, \bar{v}_{I3}, \dots$),
- the total number of neutron detections ($N_{detected\ neutrons}$), and
- the reduced factorial moments for the number of correlated neutrons that are detected per each spontaneous fission ($\bar{v}_{Det}, \bar{v}_{Det2}, \bar{v}_{Det3}, \dots$).

Important item parameters for each simulation can then be determined from these output values. The total multiplication (M_T) is determined from the ratio of the total number of neutrons created to the number of spontaneous fission neutrons as follows:

$$M_T = \frac{N_{SFneutrons} + N_{IFneutrons}}{N_{SFneutrons}} \quad \text{Eq. 24}$$

The probability of induced fission (p) is then found by rearranging Eq. 12 and using the simulation values for M_T and $\bar{\nu}_I$:

$$p = \frac{1}{\bar{\nu}_I} \left(1 - \frac{1}{M_T} \right) \quad \text{Eq. 25}$$

The leakage multiplication (M_L) is then determined from Eq. 13 using the values of M_T and p obtained from the simulation. The efficiency is found from the ratio of neutrons that are detected to those that leaked:

$$\varepsilon = \frac{N_{detected\ neutrons}}{(1 - p)(N_{SFneutrons} + N_{IFneutrons})} \quad \text{Eq. 26}$$

The singles rate can be determined for the simulation using either the spontaneous fission emission multiplicities and leakage multiplication, or the detection emission multiplicities per spontaneous fission.

$$d_1 = \varepsilon S_f \bar{\nu}_s M_L = \bar{\nu}_{Det} S_f \quad \text{Eq. 27}$$

The spontaneous fission rate (S_f) is determined from the mass of the simulated Pu, the isotopic content and the specific spontaneous fission rate for Pu-240 of 473.5 spontaneous fissions/sec/gram. The higher order detection rates can similarly be determined from:

$$d_n = \bar{\nu}_{Detn} S_f \quad \text{Eq. 28}$$

For all simulations described in this paper the efficiency is treated as a known value as determined from the simulation results.

3a. Simulations for quantifying improvement over the point model

A series of simulations were performed for evaluating the accuracy in the point model results and accuracy achieved with variations on the spatial multiplication model. A range of Pu masses and densities were simulated as shown in Table 1. For each selected scenario listed in Table 1, a total of 11 simulation cases were ran with varying height to radius ratios as shown in Table 2.

The M_L , ε , d_1 , d_2 , d_3 , d_4 , and d_5 were determined for each simulation. Figure 3 shows how the average leakage multiplication varied with weight and shape of the Pu metal cylinder. Square cylinders with height equal to twice the radius had the largest leakage multiplication. The efficiency and induced fission emission probabilities varied slightly with mass and shape of the Pu cylinders due to small

changes in the energy distribution of neutrons in the item. The spontaneous fission emission probabilities varied only by small statistical deviations between different simulated cases.

Table 1. Simulated Pu masses and densities for evaluating analysis model accuracy.

Nominal Mass of Pu metal (g)	Density	True mass of Pu-240
250	density normal for Pu metal	25.09
500	density half that for Pu metal	50.19
1000	density normal for Pu metal	100.38
2000	density twice that for Pu metal	200.75
4000	density normal for Pu metal	401.51

Table 2. Height to radius ratios for each simulation case which were run for each scenario listed in Table 1.

Case ID	H/R ratio		Case ID	H/R ratio		Case ID	H/R ratio
Case 1	0.004		Case 4	0.5		Case 10	4
Case 7	0.2		Case 3	1		Case 11	5
Case 6	0.25		Case 8	2		Case 2	78.1
Case 5	0.33		Case 9	3

Figure 3. The average leakage multiplication as a function of mass and shape of Pu metal cylinders.

To evaluate the accuracy of the point model and the spatial multiplication model, the detected multiplicity rates determined from the simulation (d_1 , d_2 , d_3 and in some cases d_4) were treated as knowns and used to invert the model equations. The model results for the Pu-240 mass, leakage multiplication (ML) and alpha ratio (known to be zero for each of the simulated cases) were compared to the “true” values obtained from the simulations results. In all cases the efficiency was treated as a known value as was obtained from the simulation results. The spontaneous fission emission probabilities used in the evaluations were based on an average of the simulations results. For each simulation scenario, the induced fission emission probabilities were determined from the average simulation values for Case 7 through Case 11. Cases 1 and 2 are extreme geometries (a disk and a tube respectively) and were not included in the determination of the average induced fission emission probabilities.

3b. Simulations for understanding the shape of the multiplication curve

A series of different potential curves were tested for describing the spatial distribution of the leakage multiplication and improving upon point model results. However, to gain a preliminary understanding of the shape of the leakage multiplication in an actual item, a series of simulations were performed. This section describes the simulations that were performed to understand the shape of the multiplication curve and introduces the results.

Simulations of a 4kg square right cylinder were performed to measure the multiplication as a function of position in the item. The simulations included a series of 50 samples made up of cylinders and annuli inside the item which underwent spontaneous fission. For each sampling the remainder of the item was made up of the same Pu metal composition but without evaluating spontaneous fissions. Annuli with each combination of height and radius were simulated.

Figure 4. Positions of individual annuli of Pu-240 that were simulated to obtain the shape of the neutron multiplication distribute within a Pu metal cylinder.

The results are shown in Figure 5 with the curve fit $\phi = A J_0(v_0 r / \tilde{R}) \cos(\pi z / \tilde{H})$ where $A = 2.313$, $\tilde{H} = 19.398$ and $\tilde{R} = 5.1$. The shape of the curve fit is based on the shape of neutron flux in a bare, homogenous reactor core. The spatial distribution of the leakage multiplication may differ from the expected distribution due to reflection of neutrons back into the item. The flux profile is accurate for items in a vacuum for which there would be zero reflection. The AWCC was simulated with a cadmium liner on the inner surface of polyethylene which is intended to prevent reflection. However, such protection is not perfect.

Figure 5. The spatial distribution of the neutron leakage multiplication with in a 4kg square right Pu metal cylinder.

4. ERRORS IN THE POINT MODEL

The point model results for each of the simulated scenarios and cases are given in Appendix 1. The singles, doubles and triples rate, as well as the efficiency were treated as known values to find the mass, alpha ratio and average leakage multiplication.

In general, the mass is underestimated and the alpha ratio and leakage multiplication are over estimated. This trend is consistent with previous results and is understood to result from the spatial distribution of the neutron multiplication within the item. [5] As shown in Figure 6, the error is strongly dependent on the leakage multiplication, with some small dependence on the mass and density of the item.

As a result of small deviations between the emission probabilities used in the analysis and those obtained from the simulations, there is a tendency for the mass error to be positive for the C1 and C2 simulations. Another reason for positive errors may be small statistical errors in the simulated values of the singles, doubles and triples.

5. FUNCTIONS FOR DESCRIBING THE SPATIAL DISTRIBUTION OF NEUTRON MULTIPLICITY

The functions that were tested for describing the spatial distribution of neutron multiplicity were purposely treated as functions of the radius only. In a cylinder, reactor physics (discussed more below) predicts that the multiplicity will have a cosine distribution in the vertical direction and a Bessel function distribution in the horizontal direction. However, the inclusion of two different spatial distributions depending on direction would necessitate the introduction of two additional unknowns.

Figure 6. Negative bias in the Assayed Mass determined by the Point Model

Instead, it was assumed that the items had the same spatial distribution shape and extent of curvature (M_R) in both the horizontal direction and vertical direction. As a result, the spatial multiplication model introduces only one additional unknown (M_R). This assumption is similar to assuming that the item is spherical. It is possible to extend the approach presented in this paper to include anisotropic spatial distribution and two M_R values for different directions. In that case, either the analysis would need to include pentuplets rates, or additional evaluations for how the M_R values relate either to each other or to the average leakage multiplication (M_L).

5a. Cosine Function

The cosine function is a convenient function to try for representing the spatial distribution of neutron multiplicity because it is possible to analytically solve for the average of the distribution after it has been raised to a power. In addition, the cosine function is a reasonable fit based on a previous study [5] that found the leakage multiplication as a function of position in the item. It is easy to show that the results roughly fit a cosine distribution. Therefore, for testing purposes it was assumed that the neutron multiplicity has a radial distribution

$$M_L(r) = M_A \cos(ar) \quad \text{Eq. 29}$$

where M_A is the amplitude and $M_R = aR$ is the period of the cosine curve at the radial extent ($r = R$) of the item. In general, the average value of the cosine spatial distribution is given by

$$\langle M_L^n \rangle = \frac{1}{R} \int_0^R \cos^n(ar) dr \quad \text{Eq. 30}$$

Using the general formula for integrals of $\cos^n(ar)$:

$$\int \cos^n(ar)dr = \frac{1}{na} \sin(ar) \cos^{n-1}(ar) + \frac{n-1}{n} \int \cos^{n-2}(ar)dr \quad \text{Eq. 31}$$

then the average value of each power of the neutron multiplication distribution is given by

$$\langle M_L \rangle = \frac{M_A}{M_R} \sin(M_R) \quad \text{Eq. 32}$$

$$\langle M_L^2 \rangle = M_A^2 \left(\frac{1}{2M_R} \sin(M_R) \cos(M_R) + \frac{1}{2} \right) \quad \text{Eq. 33}$$

$$\langle M_L^3 \rangle = M_A^3 \left(\frac{1}{3M_R} \sin(M_R) \cos^2(M_R) + \frac{2}{3M_R} \sin(M_R) \right) \quad \text{Eq. 34}$$

$$\langle M_L^4 \rangle = M_A^4 \left(\frac{1}{4M_R} \sin(M_R) \cos^3(M_R) + \frac{3}{8M_R} \sin(M_R) \cos(M_R) + \frac{3}{8} \right) \quad \text{Eq. 35}$$

$$\langle M_L^5 \rangle = M_A^5 \left(\frac{1}{5M_R} \sin(M_R) \cos^4(M_R) + \frac{4}{15M_R} \sin(M_R) \cos^2(M_R) + \frac{8}{15M_R} \sin(M_R) \right) \quad \text{Eq. 36}$$

$$\langle M_L^6 \rangle = M_A^6 \left(\frac{1}{6M_R} \sin(M_R) \cos^5(M_R) + \frac{5}{24M_R} \sin(M_R) \cos^3(M_R) + \frac{5}{16M_R} \sin(M_R) \cos(M_R) + \frac{5}{16} \right) \quad \text{Eq. 37}$$

$$\langle M_L^7 \rangle = M_A^7 \left(\frac{1}{7M_R} \sin(M_R) \cos^6(M_R) + \frac{6}{35M_R} \sin(M_R) \cos^4(M_R) + \frac{8}{35M_R} \sin(M_R) \cos^2(M_R) + \frac{16}{35M_R} \sin(M_R) \right) \quad \text{Eq. 38}$$

The spatial multiplication model with a cosine functional dependence is achieved by substituting the values for $\langle M_L^n \rangle$ given in Eq. 32-38 into the point model equations (Eq. 14-23) in place of M_L^n .

The results from the Cosine based spatial multiplication model are provided in Appendix 2. The first four detection rates as obtained from simulations were treated as known values along with the efficiency. The spatial multiplication model was analytically inverted to find the mass of Pu-240, the alpha ratio, the average leakage multiplication ($\langle M_L \rangle$) and the spatial extent of the multiplication curve (M_R). The mass of Pu-240, alpha ratio and average leakage multiplication determined from the assay were then compared to the known values from the simulation results.

5b. Bessel Function

Based on analogy with reactor physics (described below), another spatial distribution to try is the Bessel function. The Bessel function of the first order is given by

$$J_0(r) = \sum_{m=0}^{\infty} \frac{(-1)^m}{m! \Gamma(m+1)} \left(\frac{r}{2} \right)^{2m} \quad \text{Eq. 39}$$

Where $\Gamma(z)$ is the gamma function. The Bessel function $J_0(r)$ is an even function that looks similar to a cosine function except that the amplitude decays as r increases. Based on the analogy with reactor physics, it is assumed that the neutron multiplicity has a radial distribution given by

$$M_L(r) = M_A J_0(r) - 1 \quad \text{Eq. 40}$$

It is not possible to analytically find the average values of the Bessel distribution raised to a power, as was done for the cosine distribution above. As a result, the average values for the neutron multiplication curve raised to each power were found from analytical integration.

The results from the Bessel function based spatial multiplication model are also provided in Appendix 2. A repeat of the assay used for the cosine distribution was performed to find the mass of Pu-240, the alpha ratio, the average leakage multiplication ($\langle M_L \rangle$) and the spatial extent of the multiplication curve (M_R) when assuming a Bessel distribution. The mass of Pu-240, alpha ratio and average leakage multiplication were then compared to the known values from the simulation results.

5c. Other functions

In addition to the cosine curve and Bessel function, the following functions were tested:

$$M_L(r) = M_A \cos^2(r) \quad \text{Eq. 41}$$

$$M_L(r) = M_A (1 - r^2) \quad \text{Eq. 42}$$

$$M_L(r) = M_A (1 - r^4) \quad \text{Eq. 43}$$

$$M_L(r) = M_A (1 - r^6) \quad \text{Eq. 44}$$

The results from the spatial multiplication model using each of these functional forms to describe the neutron multiplication are also provided in Appendix 2. An identical analysis was performed using the first four detection rates and efficiency as known values to find the mass of Pu-240, the alpha ratio, the average leakage multiplication ($\langle M_L \rangle$) and the spatial extent of the multiplication curve (M_R). Analytical integration was used to find the average of each distribution raised to a power (identical to the procedure used to find the Bessel function results).

5d. Relationship to Reactor Physics

The equations for the neutron multiplication can be directly related to equations commonly used in reactor physics. In reactor physics, the multiplication factor k is given by

$$k = \eta f P_{NL} \quad \text{Eq. 45}$$

where the parameter $\eta = \bar{\nu}_l P_f$ is the average number of neutrons created from induced fission per each neutron absorbed in the reactor fuel, P_f is the conditional probability that if a neutron is absorbed in fuel it will induce a fission reaction, the parameter $f = P_{AF}$ is the conditional probability that if a neutron is

absorbed it will be absorbed in the fuel and P_{NL} is the nonleakage probability, or the probability that a neutron will not leak out of the system before absorption.

In neutron multiplicity counting it is commonly assumed that a neutron either leaks from the item or induces fission. In other words, it is assumed that the probability of non-leakage is equal to the probability of inducing fission, or that $P_{NL} = p$. In addition, within the assumption of neutron multiplicity counting the conditional probabilities P_f and P_{AF} are equal to unity: $P_f = P_{AF} = 1$. As a result, the multiplication factor is given by

$$k = \bar{\nu}_I p \quad \text{Eq. 46}$$

which is a formula often quoted in neutron multiplicity counting. In reactor physics the non-leakage probability is given by

$$P_{NL} = \frac{1}{1 + L^2 B_g^2} \quad \text{Eq. 47}$$

where $L = \sqrt{D/\Sigma_a}$ is the neutron diffusion length where D is the diffusion coefficient and Σ_a is the macroscopic absorption cross-section. The parameter B_g^2 is called the geometric buckling in reactor physics. Together, the term $L^2 B_g^2$ provides a measure of how much neutron leakage occurs as a result of diffusion out of the edges of the item. The neutron multiplicity equation for the total multiplication (M_T) can be rearranged to find an expression for the probability of induced fission:

$$p = \frac{M_T - 1}{\bar{\nu}_I M_T} \quad \text{Eq. 48}$$

Setting Equations 47 and 48 equal to each other reveals a relationship between the neutron leakage and the total multiplication:

$$L^2 B_g^2 = \frac{\bar{\nu}_I M_T}{(M_T - 1)} - 1 \quad \text{Eq. 49}$$

The flux profile for a number of homogenous reactor shapes can be solved analytically. [6] Some such shapes and their corresponding flux profiles in a vacuum are shown in Figure 6. Each of the shapes in Figure 6 also has an analytically solvable geometric buckling deduced directly from the flux profile.

It is also possible to relate the flux profile to either the total multiplication or the leakage multiplication parameters utilized in neutron multiplicity counting. The induced fission rate in the item can be found from reactor physics principles as:

$$\dot{S}_I = \phi N \sigma_f \quad \text{Eq. 50}$$

Where $\dot{S}_I$ is the induced fission rate in the item, N is the number density (cm^{-3}) of atoms that undergo induced fission (in this case Pu-239) and σ_f is the microscopic cross section (cm^2) for induced fission. The accent on the induced fission term is used to indicate a per volume parameter. Therefore $\dot{S}_I$ has units of fissions/ cm^3/s .

Figure 6. Analytically derived flux profiles for homogenous reactors in a vacuum.

It is also possible to write the induced fission rate in terms of the leakage multiplication starting from the definition of total multiplication. Usually total multiplication is a summed value over the entire item. In the following treatment the total multiplication is treated as a function of the position in the item, indicated by radius (r) and height (h). The total multiplication is the number of neutrons produced from both spontaneous and induced fission per each spontaneous fission neutron:

$$M_T(r, h) = \frac{\bar{\nu}_S \dot{S}_f(r, h) + \bar{\nu}_I \dot{S}_I(r, h)}{\bar{\nu}_S \dot{S}_f(r, h)} \quad \text{Eq. 51}$$

This can be rearranged to find that

$$\dot{S}_I = \frac{\bar{\nu}_S}{\bar{\nu}_I} \dot{S}_f (M_T - 1) \quad \text{Eq. 52}$$

By setting equations 50 and 52 equal to each other the following equations for the flux can be found:

$$\phi = \frac{\bar{\nu}_S}{\bar{\nu}_I} \frac{S_f}{N \sigma_f} (M_T - 1) = \frac{\bar{\nu}_S}{(\bar{\nu}_I - 1)} \frac{S_f}{N \sigma_f} (M_L - 1) \quad \text{Eq. 53}$$

As a result, it is clear that the leakage multiplication minus unity should follow the flux profiles described in Figure 6. This lends physical legitimacy to using the Bessel curve to describe the spatial distribution of leakage multiplication within cylindrical items. However, it is understood in reactor physics that the analytical results for the curve shapes are not good within several mean free path lengths of neutrons from the edge of the item. Therefore, it is legitimate to experiment with curves other than the Bessel function to find the best experimental results.

5f. Summary of Results for the four parameter spatial multiplication model

The results for the four parameter spatial multiplication model with various spatial distribution functions are given in Appendix 2. Selected results are shown in Figure 7.

Figure 7. Results from the 4 Parameter Spatial Multiplication Model for various selected Curve shapes

6. THREE PARAMETER KNOWN M_R SPATIAL MULTIPLICATION MODEL

The spatial extent (M_R) of the leakage multiplication curve tends to increase with the average leakage multiplication (M_L). For small leakage multiplications, the spatial distribution of the leakage multiplication is nearly flat across the item. However, for large average leakage multiplications the leakage multiplication at the center of the item may be much larger than the leakage multiplication at the edge of the item.

The concept of the three parameter known M_R spatial multiplication model is to identify a relationship between M_R and M_L . This way, the spatial multiplication model can be inverted with three detection rates: singles, doubles and triples.

The relationship between M_R and M_L was found from the same simulations that were used for testing the accuracy of the model results. However, all simulations results were treated as known values and the value of M_R was adjusted to find the best fit between the “true” detection rates from the simulation results and the predicted detection rates based on the spatial model, and the “true” alpha ratio, mass and average leakage multiplication. The best-fit M_R was then plotted against the average leakage multiplication for each simulated scenario and case.

Different relationships between M_R and M_L were found depending on the shape of the assumed neutron multiplication curve and the number of detection rates that were used to find the best fit. The results are shown in Figure 8 from three selected data sets: (1) an assumed Bessel function distribution to the leakage multiplication with a best fit based on the singles, doubles and triples detection rates, (2) an assume cosine multiplication distribution with a best fit based on the singles, doubles, triples and

quadruples detection rates and (4) an assumed Bessel function multiplication distribution with a best fit based in singles, doubles, triple, quadruples and pentuplets detection rates.

Figure 8. The relationship between M_R and M_L . Results are shown for (1) an assumed Bessel function multiplication distribution with a best fit based on the singles, doubles and triples rates, (2) an assume cosine multiplication distribution with a best fit based on the singles, doubles, triples and quadruples rates and (3) an assume Bessel function multiplication distribution with a best fit based in singles, doubles, triple, quadruples and pentuplets.

Curves were fit to the M_R vs M_L plots to quantify the relationship between the two parameters. The curve fits used were of the form:

$$M_R = \frac{1}{a} \text{acosh} \left(\frac{M_L - x}{A} \right) \quad \text{Eq. 54}$$

The parameters from each curve fit are given in Table 3.

Table 3. Fit parameters for the relationship between M_R and M_L

Shape	a	A	x
Bess (S,D,T)	2.1	0.22900	0.92100
Cos (S,D,T,Q)	4.0	0.05897	1.04103
Bess (S,D,T,Q,P)	2.3	0.21026	0.88974

The spatial multiplication model was tested again for the cosine curve and the Bessel function curve using three detection rates and a “known” M_R based on the curves shown in Figure 8. The results have a tendency to overestimate the mass value, indicating that M_R may be being overestimated as well. As shown in Figure 9, the M_R curve based on more detection rates (including quadruples and pentuplets) results in better mass values, especially for high leakage multiplications, then finding MR based on singles, doubles and triples alone. As shown in Appendix 1, the quadruples and pentuplets rates are very high for the large items with high leakage multiplications. Therefore, it may be reasonable that the

higher order detection rates need to be taken into account when quantifying the relationship between M_R and M_L .

Also as shown in Figure 9, even the best correction to the point model still starts to significantly over estimate the mass value for leakage multiplications above about 2.5. The spatial multiplication model offers a substantial improvement for leakage multiplications below 2.5. Leakage multiplications greater than 2.5 are rare in the current applications of neutron multiplicity counting. However, it may be possible to improve the analysis for higher multiplying systems through a better quantification of the relationship between M_R and M_L , potentially by accounting for sextuplets and hexuplets.

Figure 9. Results for the three parameter known M_R spatial multiplication model.

7. FUTURE WORK

Additional work which could improve this analysis and determine its true limits of reliability include:

Presence of (α,n) ratio – all of the results presented here are for Pu metals. However, the (α,n) ratio was treated as an unknown in the models and it is expected that the models will also work well when a source of (α,n) neutrons is present (such as in Pu oxides). It is also important to test the relationship between M_R and M_L for items which have a source of (α,n) neutrons to make sure that the extent parameter (M_R) is still mostly a function of M_L .

Dependence on neutron detector – all the simulations presented here were done for the active well coincidence counter (AWCC). The spatial multiplication model described here is a function of the item rather than the detector. We expect to achieve good results with any counter, but this should be confirmed.

Improved M_R vs M_L correlation by including higher order terms – It may be possible to obtain a more reliable M_R vs M_L correlation for highly multiplying items by including higher order terms (hextuplets, sextuplets, etc.). For low multiplying items, the higher order detection rates are very small and therefore have small impact on the predicted value of M_R .

Improved description of the spatial multiplication curve – It may be possible to use simulations to better understand the shape of the leakage multiplication distribution within the item. The spatial multiplication model will have improved results if an improved description of the curve can be obtained. The spatial model could be extended to include different spatial distributions depending on orientation in the item.

Extend the analogy with reactor physics to improve results – It is known in reactor physics that the neutron current can be obtained from the spatial gradient of the neutron flux. In neutron multiplicity counting, the neutron current at the edge of the item is directly related to the neutron detection rates. It may be possible to extend the spatial multiplication model to account for principals that are already used in reactor physics.

Work is ongoing at LANL in a related neutron multiplicity project. The additional work will be undertaken as part of the new project as appropriate and in line with that project's goals.

8. REFERENCES

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APPENDIX 1. SIMULATIONS RESULTS

Simulation	e	D1	D2	D3	D4	D5
Pu 250g C1	0.321047	8423	2565	473	73	13
Pu 250g C7	0.311373	9133	3762	1446	673	363
Pu 250g C6	0.311158	9240	3942	1621	818	479
Pu 250g C5	0.311056	9389	4199	1883	1048	683
Pu 250g C4	0.311201	9605	4583	2305	1455	1064
Pu 250g C3	0.312694	9940	5188	3036	2267	1977
Pu 250g C8	0.315554	10117	5462	3350	2602	2299
Pu 250g C9	0.317496	10112	5392	3231	2456	2144
Pu 250g C10	0.318621	10058	5246	3023	2204	1849
Pu 250g C11	0.319340	9995	5100	2827	1972	1578
Pu 250g C2	0.305743	8558	3022	864	280	107
Pu 500g .5xp C1	0.320420	17098	5466	1154	230	54
Pu 500g .5xr C7	0.312852	17870	6836	2254	869	388
Pu 500g .5xr C6	0.312549	18016	7072	2458	1012	483
Pu 500g .5xr C5	0.312274	18214	7396	2753	1232	640
Pu 500g .5xr C4	0.312277	18511	7882	3217	1602	920
Pu 500g .5xr C3	0.313185	18962	8613	3956	2261	1497
Pu 500g .5xr C8	0.315284	19202	8935	4265	2540	1745
Pu 500g .5xr C9	0.316737	19201	8858	4144	2405	1607
Pu 500g .5xr C10	0.317565	19131	8688	3941	2210	1436
Pu 500g .5xr C11	0.317967	19037	8502	3738	2025	1265
Pu 500g .5xr C2	0.305400	17110	6055	1741	570	219
Pu 1kg Case 1	0.319288	35256	12437	3346	975	332
Pu 1kg Case 7	0.310190	39900	21278	12867	9830	8595
Pu 1kg Case 6	0.309821	40787	23204	15594	13348	13031
Pu 1kg Case 5	0.309507	42048	26100	20091	19821	22344
Pu 1kg Case 4	0.309526	43994	30973	28765	34416	46900
Pu 1kg Case 3	0.311436	47175	39828	47638	73552	128856
Pu 1kg Case 8	0.315631	48728	43997	57507	97324	187275
Pu 1kg Case 9	0.318269	48433	42443	53111	86092	159326
Pu 1kg Case 10	0.319805	47738	39892	46664	70573	121704

Pu 1kg Case 11	0.320625	47002	37468	40956	57760	92947
Pu 1kg Case 2	0.308504	38141	18166	9136	5728	4098
Pu 4kg Case 1	0.317551	74687	31493	12313	5748	3077
Pu 4kg Case 7	0.305598	105269	121734	219542	512181	1362405
Pu 4kg Case 6	0.304940	113389	160016	370747	1114353	3829952
Pu 4kg Case 5	0.304296	127145	242451	806951	3474790	16985073
Pu 4kg Case 4	0.304190	155307	492330	2925047	22390776	194388752
Pu 4kg Case 3	0.307443	232648	1936967	32074520	673667193	15886309243
Pu 4kg Case 8	0.315076	280381	3504077	87851507	2779866475	98491429096
Pu 4kg Case 9	0.319730	252630	2441863	47099897	1149362934	31402561512
Pu 4kg Case 10	0.322316	221904	1564429	21795434	387172942	7782593811
Pu 4kg Case 11	0.323727	198965	1070304	11198097	149683436	2255189753
Pu 4kg Case 2	0.312524	98588	92896	127137	223408	444942
Pu 2kg 2xr C1	0.315211	164771	92799	59943	48529	44616
Pu 2kg 2xr C7	0.309173	190030	163735	199468	312492	556827
Pu 2kg 2xr C6	0.308309	198964	194920	282546	527899	1120283
Pu 2kg 2xr C5	0.307457	212886	250903	462764	1103207	2992224
Pu 2kg 2xr C4	0.307024	237420	373504	985220	3356634	12963800
Pu 2kg 2xr C3	0.309173	286376	721129	3282262	19186879	126756253
Pu 2kg 2xr C8	0.314814	310207	932949	5166919	36671097	294277519
Pu 2kg 2xr C9	0.318269	299785	816471	4046330	25743152	185342556
Pu 2kg 2xr C10	0.320105	284795	676287	2869695	15652184	96391620
Pu 2kg 2xr C11	0.320970	271268	566599	2072737	9766044	52085543
Pu 2kg 2xr C2	0.311499	197240	187495	260262	464523	941589

APPENDIX 2. POINT MODEL ASSAY RESULTS

Simulation	True Leakage Multiplication	Assayed Mass	Deviation from True Mass	Assayed Alpha Ratio	Assayed Average Leakage Multiplication	Deviation from true Average Leakage Multiplication
Pu 250g C1	1.02503	25.19	0.40%	-0.0023	1.02350	-0.15%
Pu 250g C7	1.14601	25.01	-0.32%	0.0043	1.14511	-0.08%
Pu 250g C6	1.16023	24.95	-0.56%	0.0060	1.15985	-0.03%
Pu 250g C5	1.17938	24.89	-0.80%	0.0081	1.17975	0.03%
Pu 250g C4	1.20586	24.76	-1.32%	0.0120	1.20784	0.16%
Pu 250g C3	1.24199	24.48	-2.43%	0.0206	1.24733	0.43%
Pu 250g C8	1.25268	24.50	-2.35%	0.0199	1.25822	0.44%
Pu 250g C9	1.24441	24.51	-2.31%	0.0194	1.24975	0.43%
Pu 250g C10	1.23340	24.58	-2.03%	0.0174	1.23778	0.36%
Pu 250g C11	1.22292	24.64	-1.79%	0.0153	1.22658	0.30%
Pu 250g C2	1.09359	25.13	0.16%	-0.0001	1.09230	-0.12%
Pu 500g .5xp C1	1.04248	50.36	0.34%	-0.0040	1.04469	0.21%
Pu 500g .5xr C7	1.11590	50.32	0.26%	-0.0027	1.11754	0.15%
Pu 500g .5xr C6	1.12606	50.32	0.26%	-0.0027	1.12772	0.15%
Pu 500g .5xr C5	1.13948	50.24	0.10%	-0.0014	1.14151	0.18%
Pu 500g .5xr C4	1.15805	50.08	-0.22%	0.0010	1.16095	0.25%
Pu 500g .5xr C3	1.18280	49.83	-0.71%	0.0049	1.18719	0.37%
Pu 500g .5xr C8	1.18979	49.74	-0.89%	0.0062	1.19483	0.42%
Pu 500g .5xr C9	1.18426	49.83	-0.71%	0.0047	1.18888	0.39%
Pu 500g .5xr C10	1.17688	49.91	-0.55%	0.0036	1.18099	0.35%
Pu 500g .5xr C11	1.16963	49.94	-0.50%	0.0030	1.17350	0.33%
Pu 500g .5xr C2	1.09452	50.28	0.18%	-0.0031	1.09747	0.27%
Pu 1kg Case 1	1.07856	101.04	0.66%	-0.0035	1.07527	-0.31%
Pu 1kg Case 7	1.25642	98.64	-1.73%	0.0154	1.25919	0.22%
Pu 1kg Case 6	1.28589	97.80	-2.57%	0.0220	1.29142	0.43%
Pu 1kg Case 5	1.32697	96.72	-3.64%	0.0306	1.33621	0.70%
Pu 1kg Case 4	1.38832	94.84	-5.52%	0.0462	1.40442	1.16%
Pu 1kg Case 3	1.47956	92.05	-8.30%	0.0705	1.50724	1.87%
Pu 1kg Case 8	1.50796	90.63	-9.71%	0.0833	1.54178	2.24%

Pu 1kg Case 9	1.48641	91.29	-9.05%	0.0768	1.51781	2.11%
Pu 1kg Case 10	1.45805	92.23	-8.12%	0.0681	1.48565	1.89%
Pu 1kg Case 11	1.43189	93.22	-7.13%	0.0592	1.45567	1.66%
Pu 1kg Case 2	1.20759	99.21	-1.16%	0.0096	1.21013	0.21%
Pu 4kg Case 1	1.14871	201.88	0.56%	-0.0021	1.14474	-0.35%
Pu 4kg Case 7	1.68238	175.95	-12.36%	0.1114	1.72710	2.66%
Pu 4kg Case 6	1.81607	166.24	-17.19%	0.1642	1.88375	3.73%
Pu 4kg Case 5	2.04070	150.38	-25.09%	0.2660	2.15187	5.45%
Pu 4kg Case 4	2.49357	121.64	-39.41%	0.5226	2.70282	8.39%
Pu 4kg Case 3	3.69581	77.30	-61.50%	1.3003	4.17265	12.90%
Pu 4kg Case 8	4.34618	64.69	-67.78%	1.7117	4.97394	14.44%
Pu 4kg Case 9	3.85902	72.74	-63.77%	1.4265	4.38936	13.74%
Pu 4kg Case 10	3.36246	83.86	-58.23%	1.1260	3.78609	12.60%
Pu 4kg Case 11	3.00174	98.29	-51.04%	0.8369	3.33754	11.19%
Pu 4kg Case 2	1.54070	185.21	-7.74%	0.0644	1.56899	1.84%
Pu 2kg 2xr C1	1.27648	399.80	-0.43%	0.0060	1.27427	-0.17%
Pu 2kg 2xr C7	1.50092	376.44	-6.24%	0.0537	1.51930	1.22%
Pu 2kg 2xr C6	1.57589	366.73	-8.66%	0.0755	1.60418	1.80%
Pu 2kg 2xr C5	1.69083	350.88	-12.61%	0.1141	1.73659	2.71%
Pu 2kg 2xr C4	1.88835	323.74	-19.37%	0.1898	1.96831	4.23%
Pu 2kg 2xr C3	2.26190	280.01	-30.26%	0.3440	2.41324	6.69%
Pu 2kg 2xr C8	2.40621	264.94	-34.01%	0.4075	2.59082	7.67%
Pu 2kg 2xr C9	2.30013	273.80	-31.81%	0.3671	2.46738	7.27%
Pu 2kg 2xr C10	2.17258	286.45	-28.66%	0.3148	2.31622	6.61%
Pu 2kg 2xr C11	2.06381	299.47	-25.41%	0.2660	2.18571	5.91%
Pu 2kg 2xr C2	1.54622	368.22	-8.29%	0.0688	1.57748	2.02%

APPENDIX 3. FOUR PARAMETER SPATIAL MULTIPLICATION MODEL RESULTS

In the following column headers, “Cos” indicates an assumed spatial distribution of $M_L(r) = M_A \cos(ar)$, “Bess” indicates a spatial distribution of $M_L(r) = M_A J_0(r) - 1$. “Cos^2” indicates $M_L(r) = M_A \cos^2(r)$, and “1-r^n” indicates a spatial distribution of $M_L(r) = M_A (1 - r^n)$.

Sim ID	Cos	Bess	Cos^2	1-r^2	1-r^4	1-r^6
Pu 250g C1	0.44%	0.44%	0.44%	0.44%	0.44%	0.44%
Pu 250g C7	0.68%	0.68%	0.68%	0.68%	0.76%	0.84%
Pu 250g C6	0.84%	0.84%	0.84%	0.88%	1.00%	1.08%
Pu 250g C5	0.92%	0.92%	0.88%	0.92%	1.08%	1.24%
Pu 250g C4	0.80%	0.80%	0.80%	0.80%	1.04%	1.28%
Pu 250g C3	0.88%	0.88%	0.88%	0.92%	1.43%	1.99%
Pu 250g C8	0.12%	0.12%	0.12%	0.12%	0.44%	0.72%
Pu 250g C9	0.40%	0.40%	0.40%	0.40%	0.76%	1.12%
Pu 250g C10	0.48%	0.48%	0.48%	0.48%	0.80%	1.12%
Pu 250g C11	0.36%	0.36%	0.32%	0.36%	0.60%	0.80%
Pu 250g C2	0.52%	0.52%	0.52%	0.52%	0.52%	0.52%
Pu 500g .5xp C1	1.12%	1.12%	1.12%		1.12%	1.12%
Pu 500g .5xr C7	0.54%	0.54%	0.54%		0.54%	0.54%
Pu 500g .5xr C6	0.42%	0.42%	0.42%		0.42%	0.42%
Pu 500g .5xr C5	0.10%	0.06%	0.10%		0.10%	0.10%
Pu 500g .5xr C4	0.02%	0.02%	0.02%		0.04%	0.04%
Pu 500g .5xr C3	0.22%	0.22%	0.22%		0.28%	0.34%
Pu 500g .5xr C8	0.10%	0.10%	0.10%		0.16%	0.22%
Pu 500g .5xr C9	-0.02%	-0.02%	-0.02%		0.02%	0.04%
Pu 500g .5xr C10	0.04%	0.04%	0.04%		0.08%	0.10%
Pu 500g .5xr C11	0.04%	0.04%	0.04%		0.08%	0.10%
Pu 500g .5xr C2	0.48%	0.48%	0.48%		0.48%	0.48%
Pu 1kg Case 1	0.66%	0.69%	0.69%	0.69%	0.69%	0.69%
Pu 1kg Case 7	0.48%	0.49%	0.48%	0.49%	0.74%	0.98%
Pu 1kg Case 6	0.39%	0.40%	0.39%	0.40%	0.81%	1.23%
Pu 1kg Case 5	0.10%	0.11%	0.09%	0.12%	0.72%	1.38%
Pu 1kg Case 4	-0.70%	-0.70%	-0.72%	-0.68%	0.23%	1.35%
Pu 1kg Case 3	-2.37%	-2.37%	-2.40%	-2.35%	-1.06%	0.67%
Pu 1kg Case 8	-2.80%	-2.80%	-2.84%	-2.77%	-1.05%	1.64%

Pu 1kg Case 9	-2.29%	-2.28%	-2.32%	-2.25%	-0.57%	2.04%
Pu 1kg Case 10	-2.05%	-2.05%	-2.08%	-2.02%	-0.65%	1.27%
Pu 1kg Case 11	-1.72%	-1.71%	-1.74%	-1.69%	-0.58%	0.86%
Pu 1kg Case 2	0.37%	0.37%	0.37%	0.37%	0.51%	0.64%
Pu 4kg Case 1	0.83%	0.83%	0.83%	0.83%	0.84%	0.85%
Pu 4kg Case 7	-3.79%	-3.79%	-3.85%	-3.74%	-1.41%	2.88%
Pu 4kg Case 6	-5.96%	-5.96%	-6.05%	-5.87%	-1.99%	17.01%
Pu 4kg Case 5	-12.10%	-12.11%	-12.21%	-12.00%	-7.35%	10.70%
Pu 4kg Case 4	-23.80%	-23.81%	-23.93%	-23.67%	-18.27%	0.60%
Pu 4kg Case 3	-49.95%	-49.96%	-49.99%	-49.91%	-48.10%	-46.08%
Pu 4kg Case 8	-58.77%	-58.77%	-58.79%	-58.75%	-57.74%	-56.79%
Pu 4kg Case 9	-53.85%	-53.85%	-53.88%	-53.82%	-52.50%	-51.16%
Pu 4kg Case 10	-43.82%	-43.83%	-43.89%	-43.74%	-40.67%	-36.29%
Pu 4kg Case 11	-37.16%	-37.17%	-37.24%	-37.08%	-33.84%	-28.73%
Pu 4kg Case 2	-2.52%	-2.52%	-2.54%	-2.50%	-1.55%	-0.43%
Pu 2kg 2xr C1	0.86%	0.86%	0.86%	0.87%	0.97%	1.05%
Pu 2kg 2xr C7	-1.02%	-1.02%	-1.04%	-1.00%	-0.02%	1.16%
Pu 2kg 2xr C6	-2.25%	-2.25%	-2.28%	-2.22%	-0.84%	1.04%
Pu 2kg 2xr C5	-4.19%	-4.19%	-4.24%	-4.14%	-1.91%	2.02%
Pu 2kg 2xr C4	-9.02%	-8.89%	-8.97%	-8.82%	-5.70%	1.73%
Pu 2kg 2xr C3	-18.71%	-18.59%	-18.66%	-18.51%	-15.37%	-9.34%
Pu 2kg 2xr C8	-22.42%	-22.31%	-22.38%	-22.24%	-19.31%	-14.35%
Pu 2kg 2xr C9	-20.12%	-20.01%	-20.08%	-19.93%	-16.78%	-10.82%
Pu 2kg 2xr C10	-17.44%	-17.32%	-17.39%	-17.24%	-14.14%	-8.00%
Pu 2kg 2xr C11	-14.31%	-14.19%	-14.27%	-14.11%	-10.87%	-3.64%
Pu 2kg 2xr C2	-2.69%	-2.68%	-2.71%	-2.66%	-1.58%	-0.23%

APPENDIX 4. KNOWN MR SPATIAL MULTIPLICATION MODEL RESULTS

In the following column, “Cos” indicates an assumed spatial distribution of $M_L(r) = M_A \cos(ar)$, and “Bess” indicates a spatial distribution of $M_L(r) = M_A J_0(r) - 1$. The values in parentheses (S,D,T, etc.) indicate how many multiplicities rates were used to find the relationship between M_R and M_L where S is singles, D is doubles, T is triples, Q is quintuplets and P is pentuplets.

Sim ID	Bess (S,D,T)	Cos (S,D,T,Q)	Bess (S,D,T,Q,P)
Pu 250g C1	0.40%	0.40%	0.40%
Pu 250g C7	-0.32%	-0.08%	-0.16%
Pu 250g C6	-0.52%	-0.16%	-0.24%
Pu 250g C5	-0.72%	-0.16%	-0.32%
Pu 250g C4	-1.00%	-0.24%	-0.44%
Pu 250g C3	-1.55%	-0.68%	-0.96%
Pu 250g C8	-1.32%	-0.40%	-0.68%
Pu 250g C9	-1.39%	-0.52%	-0.80%
Pu 250g C10	-1.32%	-0.48%	-0.72%
Pu 250g C11	-1.24%	-0.40%	-0.64%
Pu 250g C2	0.16%	0.16%	0.16%
Pu 500g .5xp C1	0.34%	0.34%	0.34%
Pu 500g .5xr C7	0.26%	0.30%	0.30%
Pu 500g .5xr C6	0.26%	0.36%	0.34%
Pu 500g .5xr C5	0.10%	0.32%	0.26%
Pu 500g .5xr C4	-0.20%	0.20%	0.10%
Pu 500g .5xr C3	-0.55%	0.04%	-0.12%
Pu 500g .5xr C8	-0.69%	-0.02%	-0.20%
Pu 500g .5xr C9	-0.55%	0.08%	-0.08%
Pu 500g .5xr C10	-0.46%	0.12%	-0.04%
Pu 500g .5xr C11	-0.44%	0.08%	-0.06%
Pu 500g .5xr C2	0.18%	0.18%	0.18%
Pu 1kg Case 1	0.66%	0.66%	0.66%
Pu 1kg Case 7	-0.67%	0.25%	-0.04%
Pu 1kg Case 6	-0.91%	0.05%	-0.28%
Pu 1kg Case 5	-1.03%	-0.08%	-0.44%
Pu 1kg Case 4	-1.23%	-0.42%	-0.79%
Pu 1kg Case 3	-1.16%	-0.72%	-1.10%
Pu 1kg Case 8	-1.57%	-1.28%	-1.65%

Pu 1kg Case 9	-1.62%	-1.23%	-1.61%
Pu 1kg Case 10	-1.61%	-1.08%	-1.46%
Pu 1kg Case 11	-1.47%	-0.82%	-1.21%
Pu 1kg Case 2	-0.80%	-0.06%	-0.26%
Pu 4kg Case 1	0.56%	0.80%	0.78%
Pu 4kg Case 7	1.73%	1.06%	0.82%
Pu 4kg Case 6	2.07%	0.55%	0.30%
Pu 4kg Case 5	3.26%	0.34%	-0.03%
Pu 4kg Case 4	8.27%	2.86%	1.79%
Pu 4kg Case 3	43.33%	32.59%	28.15%
Pu 4kg Case 8	71.49%	58.06%	51.51%
Pu 4kg Case 9	49.75%	38.32%	33.33%
Pu 4kg Case 10	30.19%	20.82%	17.40%
Pu 4kg Case 11	20.06%	12.24%	9.88%
Pu 4kg Case 2	1.31%	1.47%	1.18%
Pu 2kg 2xr C1	0.91%	1.86%	1.55%
Pu 2kg 2xr C7	1.33%	1.72%	1.34%
Pu 2kg 2xr C6	1.50%	1.48%	1.13%
Pu 2kg 2xr C5	1.79%	1.07%	0.74%
Pu 2kg 2xr C4	2.76%	0.78%	0.41%
Pu 2kg 2xr C3	7.40%	3.20%	2.45%
Pu 2kg 2xr C8	9.99%	5.00%	3.99%
Pu 2kg 2xr C9	7.66%	3.23%	2.41%
Pu 2kg 2xr C10	5.40%	1.68%	1.04%
Pu 2kg 2xr C11	4.09%	1.00%	0.49%
Pu 2kg 2xr C2	1.01%	1.12%	0.76%