

LA-UR-17-20319 (Accepted Manuscript)

# Extension of the Dytlewski-style dead time correction formalism for neutron multiplicity counting to any order

Croft, Stephen Favalli, Andrea

Provided by the author(s) and the Los Alamos National Laboratory (2017-09-20).

**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.2017.06.032

Permalink to record: http://permalink.lanl.gov/object/view?what=info:lanl-repo/lareport/LA-UR-17-20319

#### **Disclaimer:**

Approved for public release. Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.



### **Accepted Manuscript**

Extension of the Dytlewski-style dead time correction formalism for neutron multiplicity counting to any order

Stephen Croft, Andrea Favalli



 PII:
 S0168-9002(17)30686-1

 DOI:
 http://dx.doi.org/10.1016/j.nima.2017.06.032

 Reference:
 NIMA 59923

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

Received date :23 February 2017Revised date :8 June 2017Accepted date :20 June 2017

Please cite this article as: S. Croft, A. Favalli, Extension of the Dytlewski-style dead time correction formalism for neutron multiplicity counting to any order, *Nuclear Inst. and Methods in Physics Research, A* (2017), http://dx.doi.org/10.1016/j.nima.2017.06.032

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

### Extension of the Dytlewski-Style Dead Time Correction Formalism for Neutron Multiplicity Counting To Any Order

Stephen Croft<sup>1</sup>, Andrea Favalli<sup>2</sup>

crofts@ornl.gov, afavalli@lanl.gov

<sup>1</sup>Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831-6166 USA

<sup>2</sup>Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM 87545 USA

### Abstract

Neutron multiplicity counting using shift-register calculus is an established technique in the science of international nuclear safeguards for the identification, verification, and assay of special nuclear materials. Typically passive counting is used for Pu and mixed Pu-U items and active methods are used for U materials. Three measured counting rates, singles, doubles and triples are measured and, in combination with a simple analytical point-model, are used to calculate characteristics of the measurement item in terms of known detector and nuclear parameters. However, the measurement problem usually involves more than three quantities of interest, but even in cases where the next higher order count rate, quads, is statistically viable, it is not quantitatively applied because corrections for dead time losses are currently not available in the predominant analysis paradigm. In this work we overcome this limitation by extending the commonly used dead time correction method, developed by Dytlewski, to quads. We also give results for pents, which may be of interest for certain special investigations. Extension to still higher orders, may be accomplished by inspection based on the sequence presented. We discuss the foundations of the Dytlewski method, give limiting cases, and highlight the opportunities and implications that these new results expose. In particular there exist a number of ways in which the new results may be combined with other approaches to extract the correlated rates, and this leads to various practical implementations.

Keywords: neutron coincidence counting; neutron multiplicity counting; dead time corrections.

#### Introduction

Time correlation analysis (TCA) of detected-neutron pulse trains using various time gating methods is the basis of fission rate determination used for the detection, verification and assay of certain special nuclear materials (SNMs). For quantitative analysis three things are needed: a way to extract average correlation counting rates from the registered pulse train, an understanding of how to either include or correct for rate loss effects, and a way to interpret the rates in terms of the physical properties of interest of the item. In the following we shall address all three of these aspects with a focus on a scheme for making practical dead time corrections.

How to extract multiplet information from the pulse train using either Signal Trigger Inspection (STI) histograms, Random Trigger Inspection (RTI) histograms, or MIXed coincidence gating expressions that make use of the difference histogram is summarized in [1-3]. Of these three gating schemes the MIXed has been by far the mostly widely used in multiplicity shift register counting for the past three decades in the field of applied nuclear safeguards. In multiplicity counting using shift register logic it is traditional to use MIXed expressions up to triples. Here we show explicitly how to use all three gating approaches to extract dead time corrected (DTC) correlated rates up to fifth order. Extension to higher order rates is also made clear through the recent extension of the point-model equations up to any order [7,36]. Preliminary expressions for the DTC in the MIXed scheme have already been reported [14,15] but not the derivation or related discussion. Because there is formal equivalence between the information that can be extracted from the pulse train using the various autocorrelation analysis methods, STI, RTI, or MIXed, one is free to use which ever scheme gives the best statistical precision and least dead time bias. We note, for example, that there is a close connection

between the RTI approach for extracting doubles and the Feynman-Y statistic [3,11]. When using random (with respect to the pulse train) triggering fast accidental sampling (FAS) [4] at a high clock rate (greater than the incident recorded event rate), which leads to overlapping gates (distinct to the original Feynman sampling which uses contiguous gates; although the expectation values are the same) is always preferred on grounds of precision and is to be recommended when room background is not changing.

Widely used expressions for dead time corrected (DTC) singles, doubles and triples rates from multiplicity shift register (MSR) based passive neutron multiplicity counting (PNMC) are available in [5-6]. We refer to this approach as the Dytlewski dead time correction method. However, in practice, in applying the generic Dytlewski method, there are variants which differ in detail. We shall discuss some of these variants so that ambiguity of use might be avoided. We explain the assumptions behind Dytlewski's method and also extend the discussion provided by Dytlewski in several other important ways, in particular, to higher order rates, beyond triples. In particular we give explicit expressions for how to compute the dead time correction coefficients needed to extract correlated rates up to pents.

### **Dytlewski's Original Dead Time Correction Treatment**

In traditional signal (neutron event) triggered neutron multiplicity analysis the number of occurrences of multiplicity *i* in the signal-triggered Reals plus Accidentals (R+A) and randomly triggered or Accidentals (A) inspection intervals are recorded [3]. Denoting these two histograms by  $N_i$  and  $B_i$  and the data acquisition time by *t*, the apparent or measured singles,  $S_m$ , doubles,  $D_m$ , and triples,  $T_m$ , rates are calculated as follows [5-6]:

(1)

$$S_m = \frac{1}{t} \cdot \sum_{i=0}^{\infty} N_i = \frac{1}{t} \cdot \sum_{i=0}^{\infty} B_i$$

These two expressions for the measured singles rate follow from the fact that the singles rate is also the time averaged trigger rate which is the rate of inspection interval (coincidence gate) openings. These equations therefore also embody the expectation that, for internally consistent (conforming to the mathematical assumptions) data, the number of times each of the two coincidence intervals are inspected during the data acquisition period is equal to the total number of neutron events electronically recorded by the MSR. A data consistency check is performed to ensure that this is the case, within one or two recorded events, consistent with the internal timing of the multiplicity shift register modules, and provided this check is satisfied, the singles rate is then conventionally calculated from the  $B_i$  histogram. Corresponding expressions for the doubles and triples are:

$$D_m = \frac{1}{t} \cdot \sum_{i=1}^{\infty} i \cdot (N_i - B_i)$$
<sup>(2)</sup>

$$T_m = \frac{1}{t} \cdot \sum_{i=2}^{\infty} \frac{i \cdot (i-1)}{2} \cdot (N_i - B_i) - A_m \cdot \frac{D_m}{S_m}$$
(3)

where  $(N_i - B_i)$  is the signal triggered difference-histogram, and  $A_m$  is the measured accidentals coincidence rate for doubles counting, derived from the random triggered A-histogram, and is given by:

$$A_m = \frac{1}{t} \cdot \sum_{i=1}^{\infty} i \cdot B_i \tag{4}$$

Because the RTI histogram is conventionally being sampled at the incoming event rate  $S_m$ , and because on the average one would expect to find the number of pulses in the gate to be equal to  $S_m \cdot T_g$ , where  $T_g$  is the duration of the gate, we may also calculate the expectation value of the accidental coincidence doubles rate from:

(5)

$$A_c = S_m \cdot \left( S_m \cdot T_g \right)$$

For conforming data we expect  $A_m$  and  $A_c$  to be numerically equal within the limits of counting precision. Typically, a software check is made to confirm this is true. Failure of this test is taken as an indication of either a fault in the data acquisition system, or more usually, an indication that the counting rate is not steady throughout the assay period, for example because neutron emitting sources were in movement near to the instrument during data collection, or because the natural background changed. Provided one has an independent means for believing the experimental conditions are stable (the quiescent conditions of the measurement such as room background are unchanging), then  $A_c$  is usually the more precise predictor of the accidentals rate. It is therefore tempting, and potentially beneficial, to adopt the calculated accidentals rate where ever possible. Assuming  $A_m$  and  $A_c$  to be numerically interchangeable, therefore, we can write a variation to the Dytlewski recipe for computing the measured triples rate from the histogram data as follows:

$$T_m = \frac{1}{t} \cdot \sum_{i=2}^{\infty} \frac{i \cdot (i-1)}{2} \cdot (N_i - B_i) - S_m \cdot D_m \cdot T_g$$

For numerical work this should give similar (if not exactly) the same results, but the important thing to note is that the results obtained using this expression, which differs from that given by Dytlewski [see following chapters], should be of comparable or better quality. That is to say, any numerical differences should be inconsequential in the context of the statistical power of the experimental data and therefore unimportant from a practical standpoint (although from a purely numerical and code implementation perspective any differences will be genuine).

(6)

When used to test for variation in the ambient background during the assay, agreement between  $A_m$  and  $A_c$ , although only as good as the combined precision, can non-the-less be quite sensitive, because  $A_m$  tracks with the incoming pulse train commensurate with the time resolution governed by the action of the shift-register. On the other hand  $A_c$  is calculated using the average singles counting rate formed over the entire duration of the assay and so can not follow a changing rate. Thus any difference between  $A_m$  and  $A_c$  provides strong evidence that the rate was not constant.

In place of  $A_c$  one may also adopt the accidentals rate  $A_F$  derived from FAS, as alluded to earlier. This is derived from the accidentals histogram formed by sampling the pulse train at a high clock frequency (e.g. at 50 MHz, an order of magnitude greater than the highest anticipated neutron count rate). The high sampling rate ensures that the expectation values of moments calculated from the histogram are obtained with a precision which is limited only by the inherent information content present on the pulse train. FAS also tracks any changes in the counting rate in near real time according to the workings of the shift register and so comparing  $A_F$  with  $A_m$  is a futile exercise in this case, because the self-consistency check will never fail. But comparing

 $A_F$  with  $A_c$  is a valid test. If this test passes the added benefit of using FAS is that the accidental histogram can be used to obtain more precise results that the traditional event triggered scheme for all orders of correlated rate – not just doubles. The only change to the algorithm is trivial, being one of accidental histogram normalization [1]. The formulation is then executed in terms of the histograms expressed as relative frequency distributions (see Appendix I).

According to the work of Dytlewski and co-workers [5-6], under certain simplifying assumptions [8], which are discussed in the Assumptions Section, the corresponding dead time corrected doubles and triples, rates  $D_c$ , and  $T_c$ , may be calculated from the multiplicity histograms as follows, in terms of an effective fixed-value extending dead time, d, per event, for the system:

$$D_c = CF_S \cdot \frac{1}{t} \cdot \sum_{i=1}^{\infty} \alpha_i \cdot (N_i - B_i)$$
(7)

$$T_c = CF_S \cdot \left[\frac{1}{t} \cdot \sum_{i=2}^{\infty} \beta_i \cdot (N_i - B_i) - \left(\frac{1}{t} \cdot \sum_{i=1}^{\infty} \alpha_i \cdot B_i\right) \cdot \frac{D_c}{S_c}\right]$$
(8)

The  $\alpha_i$  and  $\beta_i$  factors may be thought of as dead time corrected versions of the reduced factorial moment weight factors. They are functions of the dead time parameter, d, [5] and are dependent on the assumptions of the particular dead time model assumed [5,8], as well as on the operational gate width setting. Prescriptions for computing them are given in [8], in the limit  $d \rightarrow 0$   $\alpha_i \rightarrow 1$ and  $\beta_i \rightarrow \frac{i(i-1)}{2}$  such that the dead time corrected expressions collapse to the previously stated dead time free expressions. Here  $CF_S$  is the trigger rate (or singles rate) dead time correction. Expressions for the  $\alpha_i$  and  $\beta_i$  factors were given by Dytlewski in [5]. We discuss them in greater detail in Appendix II. The concept of a fixed extending dead time, adopted by Dytlweski, is an

idealization in practice where a distribution of values is more realistic. Therefore, when applying Dytlewski DTC method, there is an additional assumption that an "effective" DT value is adequate over the dynamic range of interest and degree of correlation on the pulse train.

Dylewski does not, however, address the singles rate loss correction explicitly within the framework of the specific dead time model he applies. Instead he assumes, without making any justification in reference to the model, a simple first order form, namely:

$$CF_S \approx e^{d \cdot S_m}$$
 (9)

Equations (9), (7) and (8) are the basis of the original Dytlewski prescription for calculating dead time corrected singles, doubles and triples rates from multiplicity shift register algorithms. Later a self-consistent DTC for singles was developed by Croft et al [10].

#### **Dytlewski's Dead Time Model Assumptions**

Dytlewski claims that *exact* dead time correction formulae are derived for the first and second factorial moments of the measured multiplicity distribution. It is important to keep in mind however that his results are based on several important assumptions [5] that deserve discussion:

1. The detector system is assumed to behave in accordance with the ideal one-channel paralyzable dead time model with a fixed dead time (DT) parameter, d. In this mathematical model each neutron detected prevents another from being registered for a time period d. If another neutron does interact in the sensitive detector volume within the

dead period then, it is not counted, but extends the dead time by a further period d. In reality, however, the DT per pulse may vary depending on details of the signal formation process and the system may comprise multiple elements. The prevalent systems used for international safeguards today are multi-channel devices comprising several groups of <sup>3</sup>He filled proportional counters, with each group connected to an amplifier/discriminator, and the (finite width) logic pulses from each amplifier/discriminator being electronically summed to create a single pulse train. The summation may be direct or through a multi-input derandomizer board in which events are read into a buffer to avoid pile up. Further, the effective dead time per interaction depends on the point of creation and orientation of the proton and triton reaction products with respect to the anode wire.

- 2. The item being measured is a steady state (on the average) emitter. That is to say, over the period of observation, we are not concerned with appreciable radioactive decay, or pulsed interrogation, or rotation of an item inside a measurement cavity etc., so that the average properties of the pulse train is constant in time.
- 3. The dead time is small compared to the predelay and much smaller that the gate width, used in the shift register analysis; the shift register analysis being one form of time correlation analysis.
- 4. Dytlewski makes use of the earlier result of Vincent [8] for the expression of P(k, n) the probability that k dead time losses occur in a group of n true events. The expression for P(k, n) are based on the assumption that a homogeneous (random) Poission counting process is at work over the duration of coincidence gate intervals,  $T_g$ . This is a simplification because it does not account for the time correlation that exists between events originating from a common fission chain and which interact as a cluster spread out

in time commensurate with the lifetime of neutrons in the moderated detector assembly. In principle this limitation can be overcome by formulating the dead time affected detection process within the frame work of the prompt induced-fission point neutron model using a generalized homogeneous Poisson distribution [16-18]. DTC implementations based on these formalisms is not currently used by the safeguards community, for example, it is not available in the predominantly used INCC software [9] adopted for use by the International Atomic Energy Agency (IAEA). An implicit consequence of Dytlewski's use of Vincent's P(k, n) expressions is that the DTC's applied to both the (R+A)- and A-histograms, and hence also the difference histogram R=[(R+A)-A], are the same. In reality one expects the dead time losses in the (R+A)gate to be higher because it is opened close in time to the triggering event meaning that there is a greater chance of there being a correlated events early in the (R+A)-gates, and therefore violating the assumption that the true arrival time of events within the gate is random with a constant probability density. Another consequence of this assumption is that, within the Dytlewski framework, there is no mention of the system die-away time (the 1/e-time response of neutron detector) only of the dead time and the gate width. In practice the gate width is usually chosen to be commensurate (within a factor of two in either direction) with the die-away time in order to achieve near optimal counting precision. Thus in practice the gate width is usually indicative of the die-away time of the system.

5. The P(k,n) expressions of Vincent [8] assume that the coincident gate is free when it is opened. In other words it is assumed that the gate is not blocked by the dead time from a previous event, or sequence of events, at the start of the inspection. Because of this

assumption P(n, n) = 0, i.e. at least one event (the first event) is always detected from the group, even if all the rest are lost to dead time [see Appendix II].

- 6. It is assumed that the highest populated histogram bin with index  $n_{max}$  corresponds to the largest true value of the group size *n* present. There is no way of experimentally knowing if the highest recorded event was actually due to an even higher multiplicity event that suffered DT losses or not. From a pragmatic perspective, however, and because of the steep drop of the histogram occupancy with *n*, this assumption seems reasonable.
- 7. Dytlewski's  $\alpha$  and  $\beta$  coefficients, which appear in the expressions for the first and second factorial moments, contain terms involving  $1/[1-(n-1)\phi]^n$  where  $\phi = d/T_g$  is the ratio of the DT to the gate width and n is the bin number, n = 0 to  $n_{max}$ . To be defined  $(n_{max} - 1)\phi < 1$  and when  $n_{max}$  large, and  $\phi$  is finite to avoid the coefficients getting excessively large, such that a single high multiplicity outlier can grossly distort the measured rate, ideally one would prefer  $(n_{max} - 1)\phi \ll 1$ . Because the Dytlewski DTC method is approximate, resting on the assumption of random pulse arrival across the gate, this condition is also necessary to limit the DT corrections to "modest values", where an approximate treatment likely to remain acceptable in terms of providing a correction which is fit for the intended purpose (and which must be judged experimentally). This provides a driver for minimizing detector system dead time at the design stage, for example by distributing the efficiency through many amplifier/discriminator units and summing the pulse train going into the high-speed MSR using a derandomizer circuit, particularly when systems of low die-away are involved

and the product of the mean number of counts recorded in a gate with the dead time is significant compared to unity (i.e. is not <<0).

8. The rate loss correction to the trigger rate can be approximated in an empirical *ad hoc* way. This assumption is required because the original Dytlewski scheme does not explicitly consider the trigger (singles rate) dead time correction in terms of the model he develops. A self-consistent singles DTC was only developed subsequently [10] by our group. It should be noted however that the self-consistent Dytlewski-Croft singles DTC scheme does not reproduce the well-known analytical result for a random neutron source, (because of the assumption of a flat arrival distribution across the gate) although it is a close approximation for all practical purposes. The same can be said of the original *ad hoc* treatment of Dytlewski.

### Variations in how Dytlewski's Dead Time Correction Might be Applied in Practice

In practice, the application of the basic Dytlewski DTC method lends itself to multiple reasonable variations. We identify some of these here. Our intention is simply to note that when it comes to implementing the basic concept different workers may take slightly different approaches and these can give rise to slightly different numerical performance. It is therefore important when reporting results which may be being used by others to benchmark calculations or to validate software to be clear on exactly what equations are being applied, even though the different choices may all be acceptable from a practical applications perspective.

- 1. In place of  $e^{d \cdot S_m}$  the result for a random Poisson source  $e^{d \cdot S_c}$  is used instead for the trigger rate correction factor  $CF_S$
- Comparison of (8) and (3) implies that the dead time corrected Accidentals rate is given

by 
$$A_c = S_c^2 \cdot T_g = \left(CF_S \cdot \frac{1}{t} \cdot \sum_{i=0}^{\infty} B_i\right)^2 \cdot T_g = CF_S \cdot \frac{1}{t} \cdot \sum_{i=1}^{\infty} \alpha_i \cdot B_i$$
 which requires for

internal consistency of the theory,  $CF_S = \frac{1}{S_m \cdot T_g} \cdot \frac{\sum_{i=1}^{\infty} \alpha_i \cdot B_i}{\sum_{i=0}^{\infty} B_i} = \frac{\sum_{i=1}^{\infty} \alpha_i \cdot B_i}{\sum_{i=1}^{\infty} i \cdot B_i}$ . These are the Croft-

Dytlewski equations [10]. For conforming data, that is for data which passes the calculated equals the measured Accidentals test, either form may be used. For simulated pulse-train data subject to end effect errors (because equilibrium does not exist in the modelling space), the second of the two alternatives is more immune to propagating residual bias. The form of the Croft-Dytlewski Singles rate DTC factor is perhaps more obvious to understand when one recognizes that the average measured Singles rate may also be obtained from the expectation value of the mean number of counts per unit time observed in the A-gate. That is  $S_m = \frac{\langle i \rangle}{T_q} = \frac{1}{T_q} \sum_{i=0}^{\infty} i \left( \frac{B_i}{\sum_{i=0}^{\infty} B_i} \right)$ , where  $\langle i \rangle$  denotes the mean value of *i* formed over the normalized experimental frequency distribution  $\frac{B_i}{\sum_{i=1}^{\infty} B_i}$ . This is the form we'd use with FAS. By extension  $S_c = \frac{\langle \alpha_i \rangle}{T_a}$ , which leads to the result quoted for the Singles dead time correction, namely  $CF_S = S_c/S_m = \langle \alpha_i \rangle / \langle i \rangle = \sum_{i=1}^{\infty} \alpha_i \cdot B_i / \sum_{i=1}^{\infty} i \cdot B_i.$ 

3. For <sup>252</sup>Cf one would expect, for a given detector, the rate ratios formed from background corrected rates,  $D_c/S_c$  and  $T_c/D_c$  to be constant and characteristic of the <sup>252</sup>Cf fissioning system. In an entirely arbitrary attempt to improve matters some implementations have provisions to implement a Dytlewski correction with greater flexibility factors of the

form exp ( $c_D$ . d) applied to the expression for the corrected Doubles rate of the Dytlewski expressions and of the form exp ( $c_T$ . d) applied to the Triples expression [19]. Alternatively the chosen forms are of the type  $(1 + c_D$ . d) and  $(1 + c_T$ . d), respectively, . This introduces two additional parameters. A common constraint is to set  $c_D = c_T$  which seems to be a "good choice" based on practical experience and also has the advantage of reducing the number of additional parameters to be determined by one. However, there is no theoretical basis for these additional factors and many expert practitioners (including us) recommend setting  $c_D = c_T = 0$ . Furthermore, just because one can improve the quality of residuals to  $D_c/S_c$  and  $T_c/D_c$  data for a set of <sup>252</sup>Cf calibration data has not been shown, to our knowledge, to improve the quality of DTCs of multiplying items of interest. This is in fact a difficult thing to do because for real Pu items the true rates are unknown. It would be interesting to exam such questions using fruitful Monte Carlo modelling.

4. Multiplicity shift register data acquisition modules often have hardware/firmware capability to acquire traditional Neutron Coincidence Counting (NCC) rates directly. The Totals and Reals rates being analogous to Singles and Doubles, but derived from hardware logic and not the histograms. Furthermore depending on what is known about the item and or depending on the precision obtained on the Triples rate, perhaps only the Singles and Doubles will be used in the analysis subsequently selected, or perhaps a Doubles only calibration will be chosen. In such cases there is an established tradition for how DTCs are made. The most commonly used empirical approach is as follows:  $S_c = S_m \cdot \exp[S_m \cdot (a + b \cdot S_m)/4]$  with  $D_c = D_m \cdot \exp[S_m \cdot (a + b \cdot S_m)]$ , where a

and b are empirical constant determined during system characterization. In terms of an effective dead time parameter, for a pulse train which is not highly correlated, these approximations may be approximated by [11]:  $S_c = S_m \cdot \exp[S_m \cdot d \cdot (1 + S_m \cdot d)]$  with  $D_c = D_m \cdot \exp \left[4 \cdot S_m \cdot d \cdot (1 + S_m \cdot d)\right]$ . The expression for Singles gives the correct limiting behavior for a purely random pulse train which is  $S_c = S_m \cdot \exp[S_c \cdot d]$  and which has already been given as an alternative approximation for  $CF_S = S_c/S_m$ . Yet another option is to use corrections of the form  $S_c = S_m \cdot \exp[A \cdot S_c]$  with  $D_c = D_m \cdot$  $\exp[B \cdot S_c]$ , where A and B are referred to as the singles and doubles dead time coefficients respectively and are treated as empirical constants [12]. Normally these are determined conveniently using the twin <sup>252</sup>Cf-source method (although this usually is no more than a two rate method). The ratio B/A is not constrained but is allowed to be determined freely by the data. Although the ratio is often numerically close to 4, itcan differ from this theoretical value by a statistically significant amount – perhaps in a band  $\pm 10\%$ . For consistency between traditional NCC algorithms and Passive Neutron Multiplicity Counting (PNMC) algorithms there is a case for using these empirical forms of dead time corrections, developed for Totals and Reals, for singles and doubles, invoking Dytlewski's expression only for the Triples rate with an ad hoc value of d, specific for the triples.

5. Yet another option exists in the same spirit as the above. The Matthes and Haas results has recently been reworked by us into a simple analytical form for easy implementation [13]. This has the form (written in the forward or predictive style):

$$S_m = S_c \cdot \left(1 - \frac{D_c/f_d}{S_c} \cdot \frac{d}{\tau}\right) \cdot exp\left[-S_c \cdot d \cdot \left(1 - \frac{1}{2} \cdot \frac{D_c/f_d}{S_c} \cdot \frac{d}{\tau}\right) + 0\left(\left[\frac{d}{\tau}\right]^4\right)\right]$$
(9)

$$D_m = D_C \cdot \left(1 - 2.S_c \cdot d \cdot \left[1 - \frac{D_c/f_d}{S_c} \cdot \frac{d}{\tau} \cdot \left(1 + \frac{1}{2} \cdot \frac{\theta_2}{\theta_1}\right)\right]\right) \cdot exp\left[-2.S_c \cdot d \cdot \left(1 - \frac{D_c/f_d}{S_c} \cdot \frac{d}{\tau}\right) + 0\left(\left[\frac{d}{\tau}\right]^4\right)\right]$$
(10)

Where, O is the usual mathematical notation for the order of magnitude error introduced by the approximation, d is the dead time parameter for the system and  $\tau$  is the 1/e dieaway time of the system, and for a system with an exponential capture time distribution:

$$f_{d} = e^{-T_{p}/\tau} \cdot \left(1 - e^{-T_{g}/\tau}\right)$$
(11)

$$\theta_1 = \tau \cdot e^{-T_p/\tau} \cdot \left(1 - e^{-T_g/\tau}\right) = \tau \cdot f_d \tag{12}$$

$$\theta_2 = \tau \cdot \frac{e^{-2 \cdot T_p / \tau} \cdot \left(1 - e^{-2 \cdot T_g / \tau}\right)}{2} \tag{13}$$

where  $T_p$  and  $T_g$  are the pre-delay and coincidence gate width settings respectively, used in the shift register analysis. From these relationships, we find by algebraic rearrangement the following equality for the case that the detector (rather than the item kinetics) dominates the capture time distribution and exhibits a single exponential die-away profile:

$$1 + \frac{1}{2} \cdot \frac{\theta_2}{\theta_1} = 1 + \frac{f_d + 2 \cdot e^{-(T_p + T_g)/\tau}}{4}$$
(14)

Again the idea here would be to use these simple analytical results for the dead time corrected singles and doubles rates, invoking the Dytlewski expression only for the triples rate.

There is also an important additional twist. For each of the variations described, which are to do with the evaluation of intermediate steps, one may use either of the two forms listed for the dead

time corrected triples rate, that is either Eqn (6) or (8), which would introduce additional numerical differences unless the fully internally self-consistent Croft-Dytlewski intermediate results are used.

From this discussion it should be clear that when a dead time correction is being described it is necessary to give a complete description of the actual dead time correction equations being used. If not, then there is sufficient ambiguity that exact numerically agreement may not be obtained by another (independent) worker trying to reproduce or replicate how the dead time correction was estimated and applied.

#### Extracting the Effective Dead Time Parameter: A Proposed New Experimental Method

In principle many methods might be acceptable for estimating the dead time including a first principles based assessment of the signal formation and processing chain. However, treating the dead time as an effective parameter to be estimated from detector characterization measurements is likely to remain a common practice. Data taken to exercise a system before it enters service can be used for this purpose. Also, basing the dead time on direct measurements may partially compensate for any mismatch between the way the actual system behaves and the idealizations supporting the theory. Such methods could include twin sources <sup>252</sup>Cf; <sup>252</sup>Cf +AmLi series; <sup>252</sup>Cf series; AmLi histogram analysis; time interval analysis; pulse injection methods etc. Here we propose a potentially simple way to estimate the dead time parameter experimentally.

The self-consistent Dytlewski-Croft-Favalli histogram formulated dead time correction method can be used to analyze the A-histogram collected using a pseudo-random neutron source for mean and reduced factorial moments. By adjusting *d* to correct the measured distribution back to the characteristics of the input Poisson distribution we have a way to determine the effective dead time parameter of measurement system using a single AmLi sealed radionuclide neutron source (as well as, if available, AmF or AmB ( $\alpha$ ,n)-neutron sources, while AmBe ( $\alpha$ ,n)-neutron source is not good to our aim due to (n,2n) correlation in Be[34])) [35], for example, rather than by the traditional means commonly used by the safeguards community which requires a set of <sup>252</sup>Cf sources spanning a broad dynamic range of counting rate and is based on achieving  $D_c/S_c$ and  $T_c/D_c$  ratios which are independent of source strength. Note, the Poisson distribution possesses the following interesting property that we can utilize for doing this. The p<sup>th</sup> reduced factorial moment of a Poisson distribution is given by:

$$\frac{i(i-1)(i-2)\dots(i-[p-1])}{p!} = \frac{\lambda^p}{p!}$$
(15)

Thus, experimentally, our choice of d may be based on the requirement that the ratio of twice the second factorial moment to the square of the first factorial moment should equal unity for a random neutron source. Algebraically, within Dytlewski framework, this procedure for estimating the dead time d may be expressed as follows:

pick d such that  $\frac{2 \cdot \sum_{i=2}^{\infty} \beta_i \cdot B_i}{\sum_{i=1}^{\infty} \alpha_i \cdot B_i} = 1$ 

(16)

This is a new result for how the (gate width setting dependent) effective dead time parameter for use specifically with the Dytlewski-Croft-Favalli MSR dead time correction method [14,15] might be experimentally determined. If the acquired data supports the extraction of higher order moments with sufficiently high precision, one could include those also, and thus estimate *d* from an over determined set of relations. The utility of this proposed new method has yet to be tested experimentally.

#### **Extension to Quads and Pents**

The beauty of Dytlewski's work [5] is that it provides a straightforward prescription for how to calculate the dead time corrected reduced factorial moments directly from the observed item specific multiplying shift register (MSR) histograms as simple vector operations. Expressions for the  $\alpha$ - and  $\beta$ -vectors appearing in the expressions for doubles and triples are given in [5]. For routine use these need to be calculated once and for all for a given detection system because they are part of the systems characteristic parameter set, like neutron detection efficiency and die-away time, which should not ordinarily change. To compute the quads rate from mixed STI and RTI histograms we require the extension to the third reduced factorial moment in order to obtain a similar prescription. That is, we seek the general expression for  $\gamma_i$  in the following action on the measured multiplicity histogram  $H'_i$ :

(16)

$$M_{3} = \sum_{i=1}^{\infty} \frac{i(i-1)(i-2)}{6} \cdot H_{i} = \sum_{i=3}^{\infty} \gamma_{i} \cdot H_{i}'$$

The quads rate is then derived from the recorded multiplicity histograms as follows:

$$Q_{c} = C_{S} \cdot \frac{1}{t} \cdot \sum_{i=3}^{\infty} \gamma_{i} \cdot (N_{i} - B_{i}) - \left(\frac{1}{T_{g}} \cdot \frac{\sum_{i=2}^{\infty} \beta_{i} B_{i}}{\sum_{i=0}^{\infty} \beta_{i}}\right) \cdot \left(D_{c} \cdot T_{g}\right) - S_{c} \cdot \left(T_{c} \cdot T_{g}\right)$$
(17)

Extending the method to the Pents rate follows a similar path requiring general expressions for  $\eta_i$ in the relation for the fourth reduced factorial moment.

$$M_4 = \sum_{i=1}^{\infty} \frac{i(i-1)(i-2)(i-3)}{24} \cdot H_i = \sum_{i=4}^{\infty} \eta_i \cdot H_i'$$
(18)

Again using mixed STI and RTI histogram expressions, the pents rate is derived from the recorded multiplicity histograms as follows:

$$P_{c} = C_{S} \cdot \frac{1}{t} \cdot \sum_{i=4}^{\infty} \eta_{i} \cdot (N_{i} - B_{i}) - \left(\frac{1}{T_{g}} \cdot \frac{\sum_{i=3}^{\infty} \gamma_{i} B_{i}}{\sum_{i=0}^{\infty} B_{i}}\right) \cdot \left(D_{c} \cdot T_{g}\right) - \left(\frac{1}{T_{g}} \cdot \frac{\sum_{i=2}^{\infty} \beta_{i} B_{i}}{\sum_{i=0}^{\infty} B_{i}}\right) \cdot \left(T_{c} \cdot T_{g}\right) - S_{c} \cdot \left(Q_{c} \cdot T_{g}\right)$$
(19)

where we have used the result:

$$S_c = \left(\frac{1}{T_g} \cdot \frac{\sum_{i=1}^{\infty} \alpha_i B_i}{\sum_{i=0}^{\infty} B_i}\right) \tag{20}$$

which simply expresses that the dead time corrected mean number of events recorded in the accidentals gate per unit time defines the dead time corrected singles rate. Deriving mixed expression for the higher order correlated rates follows the same simple pattern.

Note, as previously highlighted, within the approximations and limitations of the Dytlewski dead time correction model the same manipulations are applied to the (R+A)-histogram as to the A-histogram. Thus, our extended Dytlewski approach also gives us a way to compute dead time corrected correlated rates based on STI-only (with RTI single DTC), and RTI-only data as well as the MIXed STI/RTI histogram expressions. For the purposes of illustration in this section we elect to use one form of the mixed expressions. We discuss all three rate extraction options in Appendix I [see also Ref. 22, 23].

The problem of dead time correction now becomes one of computing the  $\gamma_i$  and  $\eta_i$  functions. This has been reported but without justification or discussion in [14] and [15] respectively. In the Appendix II we give the results along for how to compute them and also approximate forms. When the random triggered inspection histogram alone is being used to compute pents we need dead time coefficients to extract the fifth reduced factorial moment. This leads to the introduction of what we call the  $\xi_i$  coefficients and we give a full description of these also.

In Appendix II we provide the mathematical foundation of the complete Dytlewski-style approach. The transformation matrix between the observed and DTC multiplicity histograms is developed. Expressions for the coefficients are obtained. The first few are written out explicitly and provide the starting point for iterative numerical evaluation. For a given measurement

system the coefficients only need to be calculated once. This is the first comprehensive mathematical discussion of the Dytlewski-Croft-Favalli DTC formalism.

Appendix I we give expressions for how to extract the DTC factorial moment multiplet rates from the acquired histograms. We provide STI, RTI and MIXed results. Traditionally only the MIXed results have been used by the international safeguards community and only up to third order. Extraction of the DTC rates closes the loop on how to then exploit the point-model equations which are to be solved in order to make a quantitative assay of an item. For completeness we work in terms of normalized histograms so that the fast accidental sampling scheme (which we always favor) is naturally included.

### Conclusions

The pioneering work of Böhnel [32], and also of Hage and his colleagues [33], especially during the early 1980's time period paved the way for the practical implementation of three parameter multiplicity counting analysis based on time correlation counting. From a practical stand point the application of fourth and higher order correlations [7] in neutron counting has been hampered by the lack of a convenient and reliable rate loss compensation scheme. In this work we have summarized the current state of the practice in an accessible way and extended a popular dead time correction technique to fourth and fifth order. The generalization to all orders is also made clear. In particular we have provided various ways to extract higher order multiplet rates, a self-

consistent dead time correction scheme for singles, and the necessary closed form interpretation model equations for the factorial moments. A companion paper on experimental demonstration of the main theoretical results presented here is planned.

The principle limitations of high order multiplicity counting are associated with counting precision, accuracy of the rate loss treatment, and validity of the point model assumptions. Statistical viability taken together with the requirements of the basic theoretical model demands the use of a detector with a high and flat (in energy and space) efficiency, a detector with short and nearly exponential die-away profile, a detector with as little system dead time as practical, and conforming items (for example dry plutonium dioxide powders decoupled from their surrounding). These are not trivial requirements and may result in quadruplet and pentuplet analysis to be useful only under special conditions. While the detector design will play a significant role in the feasibility of using, we have identified options for signal extraction and correction. Breaking free of the constraints of the point model will be necessary. A point model based Monte Carlo pulse train simulation which by definition includes accidentals but not the full transport details of the problem is probably adequate to explore the influence of dead time parametrically, including sampling from a distribution of dead times rather which is a refinement over the assumption of a fixed value, but it is apparent even so that long runs are needed to achieve adequate precision for the higher orders – hence the attraction of minimizing dead time through design so that a simple algebraic approach is satisfactory. Monte Carlo methods as a replacement for the point model equations seems impractical in the very near term for routine use or for real time applications, although detailed look-up tables may be an approach to study. In some applications analytical extensions to the point model might be possible and useful

depending what is known or can be safely assumed about the item. But we expect that deterministic codes, which are far faster than Monte Carlo methods, may be able to plug the gap by adapting and extending algorithms developed reactor noise analysis.

Our objective here has been to identify and develop a pathway for the exploration of higher order multiplets to solve practical non-proliferation and nuclear safeguards measurement problems. The practical benefits to other areas, for example, benchmark quality sub-criticality experimental work aimed at determining basic nuclear data, remains to be assessed and sits on the cutting edge of research in this area.

This work also suggests several paths of future research. In the Dytlewski formulation of multiplet dead time corrections the gate width enters through the parameter  $\phi = \frac{d}{T_g}$ . As far as we are aware this implied dependence has not been systematically studied experimentally and needs to be done. Dytlewski's work [5] is based on Vincent's expressions for P(k, n), the probability that *k* events will be lost to dead time when the true size of the group is *n* [8]. It is an interesting proposition to replace this foundation by Vincent's later expressions [16] in which he extended his earlier analysis, which assumed a Poisson arrival time distribution over the gate, to explicitly include time correlation present on the pulse train. Not discussed in the present paper are the potential benefits of using cross-correlation in addition to autocorrelation TCA for both dead time correction and precision. Preliminary work suggests this will also be a fruitful avenue to explore.

### ACKNOWLEDGEMENTS

This work was sponsored by the U.S. Department of Energy (DOE), National Nuclear Security Administration (NNSA), Office of Nonproliferation Research and Development (NA-22). We thank Dr Peter Santi and Dr Bill Geist for encouraging and supporting this work.

### References

[1] S. Croft, D. Henzlova, and D.K. Hauck, Extraction of correlated count rates using various gate generation techniques: Part I Theory, Nuclear Instruments and Methods in Phys. Res. A691 (2012)152-158.

[2] D. Henzlova, S. Croft, H.O. Menlove, and M.T. Swinhoe, Extraction of correlated count rates using various gate generation techniques: Part II Experiment, Nuclear Instruments and Methods in Phys. Res. **A691** (2012) 159.

[3] S.Croft, A.Favalli, D.Hauck, D.Henzlova, P.Santi, Feynman Variance-to-mean in the context of passive neutron coincidence counting, Nuclear Instruments and Methods, A686 (2012), 136-144.

[4] J.E. Stewart, S.C. Bourret, M.S. Krick, M.R. Sweet. T.K. Li and A. Gorobets, New shift-registers electronics for improved precision of neutron coincidence and multiplicity assays of plutonium and uranium mass, Los Alamos National Laboratory report LA-UR-99-4927(1999). Also Proc. 6<sup>th</sup> International Meeting on Facility Operations – Safeguards Interface, sponsored by the American Nuclear Society Isotopes and Radiation Division, Jackson Hole, Wyoming, USA, September 20-24 (1999).

[5] N. Dylewski, Dead-time corrections for multiplicity counters, Nucl Instr. and Meth. A305 (1991) 492-494.

[6] N. Dytlewski, M.S. Krick and N. Ensslin, Measurement variances in thermal neutron coincidence counting, Nucl. Instr. and Meth. A327 (1993)469-479.

[7] A.Favalli, S.Croft, P.Santi, Point model equations for neutron correlation counting: extension of Böhnel's equations to any order, Nucl. Instr. and Meth. **A795**(2015)370-375.

[8] C.H. Vincent, Type II dead-time losses between detections occurring at random times within a gate-open time, Nucl. Instr. and Meth. in Phys. Res. **A269** (1988) 335-337.

[9] M.S. Krick, W.C. Harker, P.M. Rinard, T.R. Wenz, W. Lewis, P. Pham and P. De Ridder The IAEA Neutron Coincidence Counting (INCC) and the Deming least-squares fitting program Los Alamos national Laboratory report LA-UR-98-2378. Also Proc. 39<sup>th</sup> Annual INMM Meeting, Naples, FL, USA, July 26-30 (1998).

[10] S. Croft, L.G. Evans, A. Favalli, D. Hauck, D. Henzlova, P. Santi, and M.T. Swinhoe, A

New Alternative Singles Rate Dead Time Correction Algorithm for use in Neutron Multiplicity Analysis, Nucl. and Instrum. Meth. in Phys. Res. **A681** (2012)16-20.

[11] S.Croft, D.Henzlova, D.K.Hauck, A.Favalli, P.A.Santi, The Feynman-Y-statistics in relation to shift-register neutron coincidence counting: precision and dead time, 53<sup>rd</sup> INMM Annual Meeting (2012), Orlando, USA.

[12] S. Croft and A.R. Yates, The characterization of NMAS-12; a passive neutron coincidence counter for use at Dounreay, 21st Annual ESARDA (European Safeguards Research and Development Association) Symposium on Safeguards and Nuclear Material Management, Seville, Spain, 4-6 May 1999. ESARDA 29 EUR 18963 EN(1999) 515-523.

[13] S. Croft, L.G. Evans, A. Favalli, D.K. Hauck, D. Henzlova, and P. Santi, Revisiting the form of dead time corrections for neutron coincidence counting, Radiation Measurements, vol.47, issue 7 (2012), pages 467-474.

[14] S. Croft and A. Favalli, Dead Time Corrections for Neutron Multiplicity Counting, Letter to the Editor, Nucl. and Instrum. Meth. in Phys. Res. A686 (2012).

[15] S.Croft and A.Favalli, Higher Order Dead Time Corrections for Neutron Multiplicity Counting, ESARDA bulletin 47, 2012

[16] C.H.Vincent, Analysis of neutron multiplicity measurements with allowance for dead-time losses between time-correlated detections, Nucl. Instr. and Meth. in Phys. Res. A317 (1992) 281-296.

[17] W. Hage and D.M. Cifarelli, Correlation analysis with neutron count distribution for a paralyzing dead-time counter for the assay of spontaneous fissioning material, Nucl. Sci. & Engin. 112(1992)136-158.

[18] D.K. Hauck, S. Croft, L.G. Evans, A. Favalli, P. Santi, J. Dowel, Study of a Theoretical Model for the Measured Gate Moments Resulting from Correlated Detection Events and an Extending Dead Time, Nuclear Instruments and Methods in Physics Research Section A, Vol.719, 2013, pages. 57-69.

[19] NDA2000 Technical Reference Manual (2007), Canberra Industries, 800 Research Parkway, Meriden, CT 06450, US. [20] D. Henzlova, H. O. Menlove, S. Croft, A. Favalli, and P. Santi, The Impact of Gate Width Setting and Gate Utilization Factors on Plutonium Assay in passive correlated neutron counting, Nucl Instrum and Meths in Phys Res A 797 (2015) 144-152.

[21] S. Croft, P. Blanc, and N. Menaa, Precision of the accidentals rate in neutron coincidence counting – 10475, Proc. WM2010 CD ROM, Waste Management Symposium March 7-11, 2010, Improving the Future by Dealing with the Past, Phoenix, Arizona, USA.

[22] S.Croft, A.Favalli, D.Henzlova, Extracting Neutron Multiplet Rates from Normalized Multiplicity Shift Register Histograms, Institute of Nuclear Material Management, Annual Meeting 58<sup>th</sup>(2015),CA, US.

[23] S.Croft, T.Culter, A.Favalli, W.Geist, V.Henzal, D.Henzlova, K.Koehler, B.Parker, P.Santi, Exstracting Gate Utilization Factors, Institute of Nuclear Material Management, Annual Meeting 59<sup>th</sup>(2016),GA, US.

[24] S.Croft, D.Henzlova, A.Favalli, D.Hauck, P.A.Santi, The optimum choice of gate width for neutron coincidence counting, Nuclear Instruments and Methods A, vol.764 (2014), pages 322-329. Note: In the printed (journal) version there is an obvious labelling error in Fig.2 and Fig.3 because as explained in the text only source Cf3b was used with the ENMC. This was corrected for the on-line version.

[25] L. C-A. Bourva and S. Croft, Monte Carlo calculations of the neutron coincidence gate utilisation factor for passive neutron coincidence counting. Nuclear Instruments and Methods in Physics Research A 431 (1999) 485-508.

[26] S. Croft and L. C-A. Bourva, The measurement of passive neutron multiplicity counter gate utilisation factors and comparisons with theory. Nuclear Instruments and Methods in Physics Research A 453 (2000) 553-568.

[27] S Croft, RD McElroy and S Kane, Coincidence gate utilization factors for neutron correlation counters with up to three components in the die-away profile. At ICEM07: The 11<sup>th</sup> International conference on Environmental Remediation and radioactive Waste management, September 2-6 (2007), Bruges, Belgium. Paper ID 7173.

[28] S. Croft, M. Schear, and L.G. Evans, Analytical expressions for the gate utilization factors of passive multiplicity counters including signal build-up, Proceedings of the 51<sup>st</sup> Annual Meeting of the Institute of Nuclear Materials Management, July 11-15, 2010, Baltimore, MD, USA. CD ROM (2010). Also available at <u>www.inmm.org</u>.

[29] S. Croft, R.D. McElroy, S. Philips, M.F. Villani, and L.G. Evans, Dead Time Behaviors in Passive Neutron Correlation Counting, Waste Management Symposia, WM'07 February 25 – March 1 (2007), Tucson, Arizona, USA. Paper 7258.

[30] N. Ensslin, W.C. Harker, M.S. Krick, D.G. Langner, M.M. Pickrell, and J.E. Stewart, Application guide to neutron multiplicity counting, Los Alamos National Laboratory Report LA-UR-98-4090 (1998).

[31] I. Pázsit, A. Enqvist and L. Pál, A note on the multiplicity expressions in nuclear safeguards, Nucl Instrum and Meth in PR A603(2009)541-544.

[32] K.Bohnel, the effect of multiplication on the quantitative determination of spontaneously fissioning isotopes by neutron correlations analysis, Nuclear Science and Engineering, 90, number 1 (1985) 75-82.

[33] W.Hage, D.M.Cifarelli, On the factorial moments of the neutron multiplicity distribution of fission cascades, Nucl Instrum and Meth in PR A236 (1985) 165-177.

[34] S. Croft, P.B. Sharp, The measurement and calculation of the correlated neutron production rate from 241AmO2Be( $\alpha$ , n) sources, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, Volume 354, Issue 2, 1995, Pages 458-463, ISSN 0168-9002, http://dx.doi.org/10.1016/0168-9002(94)01051-X.

[35] S.Croft, S.Cleveland, A.Favalli, R.D.McElroy Jr, A.T.Simone, Estimating the effective system dead time parameter for correletated neutron counting, (2017) in Nuclear Instruments and Methods A https://doi.org/10.1016/j.nima.2017.04.042.

[36] S. Croft, A. Favalli, Incorporating delayed neutrons into the point-model equations routinely used for neutron coincidence counting in nuclear safeguards, Annals of Nuclear Energy, Volume 99, January 2017, Pages 36-39, ISSN 0306-4549, https://doi.org/10.1016/j.anucene.2016.08.012

## Appendix I: Extracting Neutron Multiplet Rates from the Normalized Multiplicity Shift Register Histograms

For completeness, in this Appendix, we summarize the expressions that may be used to extract the dead time corrected multiplet rates from the normalized multiplicity shift register histograms. Using normalized histograms allows the fast accidental sampling strategy to be incorporated from the onset [21]. Some of these results have been previously reported in the proceedings of the Annual Meeting of International Nuclear Materials Management [22, 23].

The familiar SDTQP rates are related to the more fundamental multiplet rates  $R_1 R_2 R_3 R_4 R_5$ through extraction-method dependent gate utilization factors, where  $R_1 R_2 R_3 R_4 R_5$  are the reduced factorial moments on the pulse train in the limit of infinite coincidence gate width (and zero predelay).

The STI histogram is naturally normalized to the total number of neutron event triggers recorded during the assay. We have:

$$\sum_{i=0}^{\infty} N_i = N_{TOT} = S_m t \tag{A1}$$

The RTI histogram may be normalized differently, for instance in the case of FAS we have:

$$\sum_{i=0}^{\infty} B_i = N_{FAS} = \omega t \tag{A2}$$

where  $N_{FAS}$  is the number of periodic clock requests at a frequency  $\omega$  to inspect the contents of the A-gate. Both MIXed and RTI-only implementations when applied to MSR histograms benefit from the use of fast accidental sampling to improve the overall precision. FAS is always

favored when the counting conditions are stable as it leads to the best overall precision in the extracted rates over the entire dynamic range. Because of the different sampling schemes it is therefore convenient to work in terms of the corresponding normalized frequency distributions. The normalized histograms are defined as follows:

$$n_{i} = \frac{N_{i}}{N_{TOT}}$$
(A4)  

$$b_{i} = \frac{B_{i}}{N_{FAS}}$$
(A5)  

$$r_{i} = n_{i} - b_{i}$$
(A6)

We define the following operations to compute dead time corrected reduced factorial moments over these three distributions in the spirit of the Dytlewski dead time correction model:

$$m_{y0} = \sum_{i=0}^{\infty} y_i = 1$$
(A7)  

$$m_{y1} = \sum_{i=1}^{\infty} \alpha_i \cdot y_i$$
(A8)  

$$m_{y2} = \sum_{i=2}^{\infty} \beta_i \cdot y_i$$
(A9)  

$$m_{y3} = \sum_{i=3}^{\infty} \gamma_i \cdot y_i$$
(A10)  

$$m_{y4} = \sum_{i=4}^{\infty} \eta_i \cdot y_i$$
(A11)  

$$m_{y5} = \sum_{i=5}^{\infty} \xi_i \cdot y_i$$
(A12)

where *y* stands for *n*, *b* or *r*.

In practice summations can start at i = 0 since the definition of the multipliers ( $\alpha$  through  $\xi$ ) ensure zero contribution outside the stated range. The upper range of the summation ends at the last value of *i* that is populated.

The STI and MIXed Gate Utilization Factors (GUFs) for Doubles through pents,  $f_2$  through  $f_5$ , (also denoted by  $f_d$  through  $f_p$ ) represent the fraction of the ideal multiplet rate that falls within the finite gating structure of the MSR. Thus, taking quads as an example we can write  $Q = f_4 R_4$ . Note  $f_1 \equiv 1$  which reflects the fact that the measured Singles counting rate is unaffected by the choice of either the predelay and coincidence gate width. In the case of RTI TCA the

corresponding GUFs are denoted by  $w_2$  through  $w_5$  (again  $w_2 \equiv 1$ ) and these in turn again represent the fraction of the ideal multiplet rates collected by the finite coincidence gate width [1,2].

GUFs may be estimated in various ways [23-28]. They are usually treated as fixed values, characteristic of the detector system, and are dealt with through detector characterization and calibration.

Next we give the forms of the RTI, STI and MIXed multiplet expressions in terms of the ideal reduced factorial multiplets. After that we provide expressions in terms of the histogram (dead time corrected) reduced factorial moments which are better suited practical calculations.

#### **RTI only Expressions**

The dead time corrected reduced factorial multiplet rates extracted solely from the normalized RTI histogram under the current scheme are therefore given by:

$$R_1 = S_c = \frac{m_{b1}}{T_g} \tag{A13}$$

$$R_{2} = \frac{D_{c}}{f_{d}} = \frac{1}{w_{2}} \left[ \frac{m_{b2}}{T_{g}} - \frac{1}{2} R_{1} \left( R_{1} T_{g} \right) \right]$$
(A14)

$$R_3 = \frac{T_c}{f_t} = \frac{1}{w_3} \left[ \frac{m_{b3}}{T_g} - R_1 \left( w_2 R_2 T_g \right) - \frac{1}{6} R_1 \left( R_1 T_g \right)^2 \right]$$
(A15)

$$R_{4} = \frac{Q_{c}}{f_{q}} = \frac{1}{w_{4}} \left[ \frac{m_{b4}}{T_{g}} - R_{1} (w_{3}R_{3}T_{g}) - \frac{1}{2} (w_{2}R_{2}) (w_{2}R_{2}T_{g}) - \frac{1}{2} R_{1} (R_{1}T_{g}) (w_{2}R_{2}T_{g}) - \frac{1}{2} R_{1} (R_{1}T_{g})^{3} \right]$$
(A16)

$$R_{5} = \frac{P_{c}}{f_{p}} = \frac{1}{w_{5}} \left[ \frac{m_{b5}}{T_{g}} - R_{1} (w_{4}R_{4}T_{g}) - (w_{2}R_{2})(w_{3}R_{3}T_{g}) - \frac{1}{2}R_{1} (w_{2}R_{2}T_{g})^{2} - \frac{1}{2}R_{1} (R_{1}T_{g})(w_{3}R_{3}T_{g}) - \frac{1}{6}R_{1} (R_{1}T_{g})^{2} (w_{2}R_{2}T_{g}) - \frac{1}{120}R_{1} (R_{1}T_{g})^{4} \right]$$
(A17)

#### **MIXed Expressions**

Historically shift-registers were conceived to count correlated pairs (the trigger plus the number of events falling in the associated gate). In the absence of dead time and other non-ideal behaviors the difference between the (R+A)- and A-registers per unit time gave the pairs (also known as the Reals, coincidence or Doubles) rate directly. With the advent of MSR modules the convention of working with differences, in this case the difference between the STI and RTI histograms persisted. The dead time corrected reduced factorial multiplet rates extracted from a combination of the normalized STI and RTI histograms, under the current dead time model are given, in a convenient and instructive form by [1,2, 22,29]. These extend the conventional multiplicity counting expressions for the first three moments [30,31] commonly used for applications.

Singlets:

$$R_{1} = S_{c} = C_{S} \left( \frac{N_{TOT}}{t} \right) = \left[ \frac{\sum_{i=1}^{\infty} \alpha_{i} \cdot b_{i}}{\sum_{i=1}^{\infty} i \cdot b_{i}} \right] \left( \frac{\sum_{i=0}^{\infty} N_{i}}{t} \right)$$
(A18)  
Doublets:  
$$R_{2} = \frac{D_{c}}{f_{d}} = \frac{1}{f_{2}} R_{1} m_{r1} = \frac{1}{f_{2}} \left[ R_{1} m_{n1} - R_{1} \left( R_{1} T_{g} \right) \right]$$
(A19)

The form on the far right hand side, which makes use of the calculated Accidentals Doubles rate, may be more precise, for data collected under steady ambient background rate conditions, when the RTI histogram is acquired using event triggering. On the other hand, when fast accidentals

sampling is used the result based on the difference expression is generally favored because it gives greater precision.

Triplets to Pentuplets:  

$$R_{3} = \frac{T_{c}}{f_{t}} = \frac{1}{f_{3}} \Big[ R_{1} m_{r2} - R_{1} \Big( f_{2} R_{2} T_{g} \Big) \Big]$$
(A20)  

$$R_{4} = \frac{Q_{c}}{f_{q}} = \frac{1}{f_{4}} \Big[ R_{1} m_{r3} - \frac{m_{b2}}{T_{g}} \Big( f_{2} R_{2} T_{g} \Big) - R_{1} \Big( f_{3} R_{3} T_{g} \Big) \Big]$$
(A21)  

$$R_{5} = \frac{P_{c}}{f_{p}} = \frac{1}{f_{5}} \Big[ R_{1} m_{r4} - \frac{m_{b3}}{T_{g}} \Big( f_{2} R_{2} T_{g} \Big) - \frac{m_{b2}}{T_{g}} \Big( f_{3} R_{3} T_{g} \Big) - R_{1} \Big( f_{4} R_{4} T_{g} \Big) \Big]$$
(A22)

### **STI only Expressions**

The dead time corrected reduced factorial multiplet rates extracted from the normalized STI histogram, BUT with the singles dead time correction that is used for the accidentals histogram, under the current scheme are given by:

$$R_1 = S_c = C_S \left(\frac{N_{TOT}}{t}\right) = \left[\frac{\sum_{i=1}^{\infty} \alpha_i \cdot b_i}{\sum_{i=1}^{\infty} i \cdot b_i}\right] \left(\frac{\sum_{i=0}^{\infty} N_i}{t}\right)$$
(A23)

$$R_2 = \frac{D_c}{f_d} = \frac{1}{f_2} \left[ R_1 m_{n1} - R_1 (R_1 T_g) \right]$$
(A24)

$$R_{3} = \frac{T_{c}}{f_{t}} = \frac{1}{f_{3}} \Big[ R_{1} m_{n2} - (f_{2} + w_{2}) R_{1} \big( R_{2} T_{g} \big) - \frac{1}{2} R_{1} \big( R_{1} T_{g} \big)^{2} \Big]$$
(A25)

$$R_{4} = \frac{Q_{c}}{f_{q}} = \frac{1}{f_{4}} \Big[ R_{1}m_{n3} - (f_{3} + w_{3})R_{1}(R_{3}T_{g}) - f_{2}w_{2}R_{2}(R_{2}T_{g}) - \frac{1}{2}(f_{2} + 2w_{2})R_{1}(R_{1}T_{g})(R_{2}T_{g}) - \frac{1}{6}R_{1}(R_{1}T_{g})^{3} \Big]$$
(A26)

$$R_{5} = \frac{P_{c}}{f_{p}} = \frac{1}{f_{5}} \Big[ R_{1}m_{n4} - (f_{4} + w_{4})R_{1}(R_{4}T_{g}) - (f_{3}w_{2} + f_{2}w_{3})R_{2}(R_{3}T_{g}) - \frac{1}{2}(f_{3} + 2w_{3})R_{1}(R_{1}T_{g})(R_{3}T_{g}) - \frac{w_{2}}{2}(2f_{2} + w_{2})R_{1}(R_{2}T_{g})^{2} - \frac{1}{6}(f_{2} + 3w_{2})R_{1}(R_{1}T_{g})^{2}(R_{2}T_{g}) - \frac{1}{24}R_{1}(R_{1}T_{g})^{4} \Big]$$
(A27)

Note these expressions are more complicated to apply because they involve both kinds of GUFs (both  $f_{\mu}$  and  $w_{\mu}$ ) and so, for a given application, introduce an extra set of experimental parameters that need to be established ahead of time. Note too that because the trigger neutron forms one part of the time correlation multiplet, the order of the action on the histogram is one less than the order of the multiplet. That is  $R_5$ , for example, involves terms up to  $m_{n4}$ . In comparison, in the RTI case, terms up to  $m_{b5}$  are involved.

#### **RTI** only Expressions (for practical implementation)

We shall write the expressions for SDTQP suitable for practical use when FAS is employed. We are working in terms of SDTQP because this is the established tradition within the safeguards community. We use the notation:  $x_n = \frac{w_n}{f_n}$ . Taking quads as an example, we therefore understand that:  $x_4QT_g = w_4\left(\frac{Q}{f_4}\right)T_g = w_4R_4T_g$ , which, being the product of a rate and a time is just a number with units of quads, with the presence of the RTI GUF multiplier ( $w_4$ ) accounting for finite gate width of the randomly placed gate on the pulse train.

$$ST_g = m_{b1} \tag{A28}$$

$$x_2 DT_g = m_{b2} - \frac{m_{b1}^2}{2} \tag{A29}$$

$$x_3 T T_g = m_{b3} - m_{b2} m_{b1} + \frac{m_{b1}^3}{3}$$
(A30)

$$x_4 Q T_g = m_{b4} - m_{b3} m_{b1} - m_{b2} \left(\frac{m_{b2}}{2} - m_{b1}^2\right) - \frac{m_{b1}^4}{4}$$
(A31)

$$x_5 PT_g = m_{b5} - m_{b4}m_{b1} - (m_{b3} - m_{b2}m_{b1})(m_{b2} - m_{b1}^2) + \frac{m_{b1}^5}{5}$$
(A32)

### MIXed Expressions (for practical implementation)

Convenient SDTQP rates are straightforward to derive by successive substitution into the academic forms. Thus:

$$S = \frac{m_{b1}}{T_g}$$
(A33)  

$$D = \frac{m_{b1}}{T_g} m_{r1}$$
(A34)  

$$T = \frac{m_{b1}}{T_g} [m_{r2} - m_{b1}m_{r1}]$$
(A35)  

$$Q = \frac{m_{b1}}{T_g} [m_{r3} - m_{b2}m_{r1} - m_{b1}(m_{r2} - m_{b1}m_{r1})]$$
(A36)  

$$P = \frac{m_{b1}}{T_g} [m_{r4} - m_{2}m_{r4} - m_{b2}(m_{r2} - m_{b1}m_{r1})]$$
(A36)

$$P = \frac{m_{b1}}{T_g} \left[ m_{r4} - m_3 m_{r1} - m_{b2} (m_{r2} - m_{b1} m_{r1}) - m_{b1} (m_{r3} - m_{b2} m_{r1} - m_{b1} (m_{r2} - m_{b1} m_{r1})) \right]$$
(A37)

### STI only Expressions (practical considerations)

Let us first consider the special case where there are no dead time losses.

$$S = \left(\frac{\sum_{i=0}^{\infty} N_i}{t}\right) = \frac{N_{TOT}}{t}$$
(A38)

$$D = Sm_{n1} - S(ST_g) \tag{A39}$$

$$T = Sm_{n2} - S(1 + x_2)(DT_g) - \frac{1}{2}S(ST_g)^2$$
(A40)

$$Q = Sm_{n3} - S(1 + x_3)(TT_g) - Dx_2(DT_g) - S\left(\frac{1}{2} + x_2\right)(DT_g)(ST_g) - \frac{1}{6}S(ST_g)^3$$
(A41)

$$P = Sm_{n4} - S(1 + x_4)(QT_g) - D(x_2 + x_3)(TT_g) - S\left(\frac{1}{2} + x_3\right)(TT_g)(ST_g) - S\left(x_2 + \frac{x_2^2}{2}\right)(DT_g)^2 - S\left(\frac{1}{6} + \frac{x_2}{2}\right)(DT_g)(ST_g)^2 - \frac{1}{24}S(ST_g)^4$$
(A42)

We immediately see that there is no Dytlewski-style way to estimate the dead time corrected singles rate solely from the STI-histogram. This is because  $S = N_{TOT}/t$  is the observed trigger rate and missing triggers (opening of the coincidence gate) due to dead time losses cannot be accounted. There are just fewer gate opening but we don't know how many fewer. Our earlier results for the dead time corrected singles rate made use of the RTI-histogram. And in a fully self-consistent scheme we would expect the dead time corrected Accidentals doubles rate to agree with the value calculated using the dead time corrected Singles rate,  $D_C = S_C^2 T_g$ .

A second serious drawback is evident when one considers how the GUFs come into play. Take the STI-only expression for triples which we may re-write as follows:

$$T = S\left[m_{n2} - (1 + x_2)m_{n1}(ST_g) + \left(\frac{1}{2} + x_2\right)(ST_g)^2\right]$$
(A43)

We observe that the factor  $x_2$  is embedded in the expression and there is no way to eliminate it using information from lower order terms. It therefore has to be treated as a well-known parameter determined independently of the (item specific) assay data. The same kind of dilemma exists in the expression for Q and also P. This reflects the fact the event triggered STI histogram captures a combination of genuine short term correlations and chance coincidences with respect to the trigger events and, within the STI-only formalism, it is not possible to get at the chance coincidence fraction in isolation experimentally. To account for the chance events one must make use of the theoretical relationships governing the structure of the data which imports the gate factor ratios.

For these two reasons (the lack of a self-consistent trigger rate dead time compensation scheme, and the manifestation of the two types of gate utilization factors in the multiplet-rate expressions) the STI-only expressions do not readily lend themselves to practical implementation.

#### **Appendix II: Mathematical Foundations**

The average measured multiplicity histogram, represented in matrix notation by the column vector  $\mathbf{H}'$  with elements  $H'_i$  corresponding to the number of times that multiplicity i was observed, where i = 0 to  $i_{max}$  and  $i_{max} < instrument limit$ , is taken as an approximation to the expectation value of the true underlying multiplicity histogram, denoted by the column vector,  $\mathbf{H}$ , by the equation:

$$H' = ZH \tag{B1}$$

where Z is a square matrix of dead time loss probabilities. Note we have taken care to qualify our description of this equation in terms of average, or expectation, quantities with the understanding that actual measured data will be subject to statistical variation. Experimental data are usually broken into a series of shorter acquisition cycles, each of viable precision, so that a statistical analysis can be performed to extract the uncertainty and covariance structure in the derived rates. The 'best' assay is based on the analysis of the average (or summed) histogram, with the cycle data being used solely to quantify the uncertainty.

For illustration, in the case in which the highest multiplicity  $i_{max} = 5$ , relation (B1) may be written explicitly as follows:

$$\begin{bmatrix} H'_{0} \\ H'_{1} \\ H'_{2} \\ H'_{3} \\ H'_{4} \\ H'_{5} \end{bmatrix} = \begin{bmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & P(1,2) & P(2,3) & P(3,4) & P(4,5) \\ 0 & 0 & P(0,2) & P(1,3) & P(2,4) & P(3,5) \\ 0 & 0 & 0 & P(0,3) & P(1,4) & P(2,5) \\ 0 & 0 & 0 & 0 & P(0,4) & P(1,5) \\ 0 & 0 & 0 & 0 & 0 & P(0,5) \end{bmatrix} \begin{bmatrix} H_{0} \\ H_{1} \\ H_{2} \\ H_{3} \\ H_{4} \\ H_{5} \end{bmatrix}$$
(B2)

where P(k, n) is the probability that k dead time losses will occur in a group of n true events.

We see immediately that the Z matrix a special kind of square matrix known as an upper triangular matrix. Triangular matrices are easier to invert than general invertible square matrices

because one can solve them by successive back substitution, in the case of an upper triangular matrix starting with the highest order term,  $H_5 = H'_5/P(0,5)$ . The determinant is just the product of diagonal entries.

Several interesting aspects of this relation deserve comment. Because it is assumed that the coincidence gate is unblocked when it is opened, the first event in of a group is always detected. Algebraically this may be expressed as follows:

$$P(n,n) = 0, n > 0$$
 (B3)

Consequently, under this scheme of dead time loss, occurrences of true high order multiplicity, can never get completely downgraded to measured multiplicity of order 0 (zero). Additionally signal triggered order zero events always get recorded with perfect fidelity, P(0,0) = 1 and P(0,1) = 1. Taken together this is the explanation for why:

$$H_0' = H_0 \tag{B4}$$

The P(k, n) are true probabilities which necessarily satisfy the following normalization condition:

$$\sum_{k=0}^{n} P(k,n) = \sum_{k=0}^{n-1} P(k,n) = 1$$
(B5)

where the second form follows because P(n, n) = 0 as discussed.

This normalization condition expresses the fact that, when true high multiplicity events suffer dead time losses, the measured histogram gets shifted downwards to lower values, but the overall number of records in the dead time affected histogram remains the same. That is, according to this model, the histogram changes shape but the area is unchanged. Mathematically this is expressed by the following equality that holds between the zeroth moment of the (dead time

corrected) true multiplicity histogram and the (dead time affected) measured multiplicity histogram:

$$M_0 = \sum_{i=0}^{\infty} N_i = \sum_{i=0}^{\infty} N'_i$$
 (B6)

To put this in another way, the number of records (the number of times the coincidence gate was inspected) is set externally by the number of gate triggers (e.g. event or clock). The effect of dead time losses is simply to redistribute the tally pattern to lower bins. In the case of neutron-event triggering, the trigger rate also requires dead time correction as discussed in the main text.

Formally then, the expectation value of the dead time corrected multiplicity histogram may be obtained from the measured histogram by the solution of our earlier equation, namely:

$$H = Z^{-1}ZH = Z^{-1}H'$$
 (B7)

This direct method is suitable for the correction of low rate, low multiplicity data. For example, for in-burst correction of fission-neutron losses recorded from a weak fission source, an example being measurements used to generate the basic prompt neutron multiplicity distribution. Having generated the dead time corrected histogram it can be manipulated in whatever way is required, for instance to generate reduced factorial moments of any order. However, according to Dytlewski [5] this direct approach fails (presumably for typical thermal well safeguards counters and including the type he cites in his paper, 77.4 ns dead time and coincidence gate width of 16  $\mu$ s) when the maximum multiplicity exceeds about 20. The reason given is that the dead time correction factors comprising  $\mathbf{Z}'$  and which are functions of the P(k, n) cannot be computed with adequate accuracy for the inversion to remain numerically robust. Despite this apparent impasse, what Dytlewski [5] then showed was that the first and second reduced factorial moments could none-the-less be computed accurately using the  $\alpha_i$  and  $\beta_i$  coefficients. We shall discuss this observation in greater detail below. We also extend Dytlewski's idea of using a simple matrix operation to extract reduced factorial moments to order three, four and five. This allows us to go beyond triples enabling us to also calculate quads and pents for all three gating structures (STI, RTI and MIXed) [see Appendix I]. For each order we present the results explicitly although a generalized notation would be more compact. We do this because the explicit expressions are more convenient for visualizing the trends and also for software coding and code checking. We also provide the general form of the dead time correction matrix coefficients for any order.

Taken together, the self-consistent dead time correction of the singles rate, the Dytlewski assumptions, and the generalized extension in matrix form, constitute what is referred to as the Dytlewski-Croft-Favalli (DCF) dead time correction method.

The first reduced factorial moment of the dead time corrected histogram is evaluated as follows:

$$M_1 = \sum_{i=0}^{\infty} iH_i$$
 (B8)  
which in matrix notation becomes:

$$\mathbf{M}_1 = \mathbf{C}_1 \mathbf{Z}^{-1} \mathbf{H}' = \boldsymbol{\alpha} \mathbf{H}' \tag{B9}$$

where  $C_1$  is the row matrix with elements which are the weighting coefficients to generate the first reduced factorial moment, that is:

$$\boldsymbol{C_1} = \begin{bmatrix} 0 & 1 & 2 & 3 & \dots & i & \dots & \infty \end{bmatrix}$$
(B10)

and  $\boldsymbol{\alpha}$  is the row vector  $\boldsymbol{C}_1 \boldsymbol{Z}^{-1}$  with elements  $\alpha_i$ , i = 0 to  $\infty$  ( $i_{max}$  in practice).

Expressions for the second and higher reduced factorial moments follow with obvious extension of the above notation. For the second reduced factorial moment we have:

$$M_2 = \sum_{i=0}^{\infty} \frac{i(i-1)}{2} H_i$$
(B11)

which in matrix notation becomes:

$$\boldsymbol{M}_2 = \boldsymbol{C}_2 \boldsymbol{Z}^{-1} \boldsymbol{H}' = \boldsymbol{\beta} \boldsymbol{H}' \tag{B12}$$

where  $C_2$  is the row matrix with elements which are the weighting coefficients to generate the second reduced factorial moment, that is:

$$C_2 = \begin{bmatrix} 0 & 0 & 1 & 3 & \dots & \frac{i(i-1)}{2} & \dots & \infty \end{bmatrix}$$
 (B13)

and  $\boldsymbol{\beta}$  is the row vector  $C_2 Z^{-1}$  with elements  $\beta_i$ , i = 0 to  $\infty$  ( $i_{max}$  in practice) as before, and is to be understood in all similar cases.

For the third reduced factorial moment we have:

$$M_3 = \sum_{i=0}^{\infty} \frac{i(i-1)(i-2)}{6} H_i$$
(B14)

which in matrix notation becomes:

$$M_3 = C_3 Z^{-1} H' = \gamma H' \tag{B15}$$

where  $C_3$  is the row matrix with elements which are the weighting coefficients to generate the third reduced factorial moment, that is:

$$C_3 = \begin{bmatrix} 0 & 0 & 0 & 1 & \dots & \frac{i(i-1)(i-2)}{6} & \dots & \infty \end{bmatrix}$$
 (B16)

and  $\boldsymbol{\gamma}$  is the row vector  $\boldsymbol{C}_{3}\boldsymbol{Z}^{-1}$  with elements  $\gamma_{i}$ , i = 0 to  $\infty$ .

For the forth reduced factorial moment we have:

$$M_4 = \sum_{i=0}^{\infty} \frac{i(i-1)(i-2)(i-3)}{24} H_i$$
 (B17)  
which in matrix notation becomes:

$$M_4 = C_4 Z^{-1} H' = \eta H' \tag{B18}$$

where  $C_4$  is the row matrix with elements which are the weighting coefficients to generate the third reduced factorial moment, that is:

$$\boldsymbol{C_4} = \begin{bmatrix} 0 & 0 & 0 & 1 & \dots & \frac{i(i-1)(i-2)(i-3)}{24} & \dots & \infty \end{bmatrix}$$
(B19)

and  $\boldsymbol{\eta}$  is the row vector  $\boldsymbol{C}_{4}\boldsymbol{Z}^{-1}$  with elements  $\eta_{i}$ , i = 0 to  $\infty$ .

For the fifth reduced factorial moment we have:

$$M_5 = \sum_{i=0}^{\infty} \frac{i(i-1)(i-2)(i-3)(i-4)}{120} H_i$$
(B20)

which in matrix notation becomes:

$$M_5 = C_5 Z^{-1} H' = \xi H' \tag{B21}$$

where  $C_5$  is the row matrix with elements which are the weighting coefficients to generate the third reduced factorial moment, that is:

 $\boldsymbol{C}_{5} = \begin{bmatrix} 0 & 0 & 0 & 0 & 1 & \dots & \frac{i(i-1)(i-2)(i-3)(i-4)}{120} & \dots & \infty \end{bmatrix}$ (B22)

and  $\boldsymbol{\xi}$  is the row vector  $\boldsymbol{C}_{5}\boldsymbol{Z}^{-1}$  with elements  $\boldsymbol{\xi}_{i}, i = 0$  to  $\infty$ .

Note, as we shall see explicitly again later, to calculate a given order multiplet rate from the STI or MIXed, requires the factorial moment of one less order, because the trigger neutron provides one of the time-correlated events. In contrast, to calculate a given multiplet rate from the RTI histogram alone, however, requires the factorial moment matrix operation of the same order. Thus, to compute Single, Doubles, Triples, Quads, Pents (SDTQP), from the STI or MIXed expressions requires only the  $\alpha_i$ ,  $\beta_i$ ,  $\gamma_i$ , and  $\eta_i$  functions. But to compute pents solely from the RTI histogram also requires knowledge of the  $\xi_i$  functions. That is to say, to compute quads from the RTI histogram requires that we have a dead time correction scheme up to pents (in the STI and MIXed schemes). We could proceed along similar lines to even higher order rates which would call for relations for even higher order reduced factorial moments, but, because for all cases of practical safeguards interest in quantitative neutron multiplicity counting quads is already at the limit of statistical viability, we have elected to stop our explicit illustration at pents here.

Dytlewski [5] gives expressions for the  $\alpha_i$  and  $\beta_i$  functions but not for  $\gamma_i$ ,  $\eta_i$  or  $\xi_i$ , because he only considers SDT using mixed expressions. In formulating the expressions for  $\alpha_i$  and  $\beta_i$  Dytlewski adopted the expressions for P(k, n) from Vincent [8] based on the assumptions outlined in the main text. The form of P(k, n) is thus:

$$P(k,n) = \frac{(n-1)!}{(n-1-k)!} \sum_{j=0}^{k} \frac{(-1)^{k-j}}{j!(k-j)!} [1 - (n-1-j)\phi]^n$$
(B23)

For the case  $i_{max} = n = 5$  used in our explicitly worked example above, we find:

$$P(0,2) = (1 - \phi)^2$$
$$P(1,2) = 1 - (1 - \phi)^2$$

$$P(0,3) = (1 - 2\phi)^3$$
$$P(1,3) = 2(1 - \phi)^3 - 2(1 - 2\phi)^3$$
$$P(2,3) = 1 - 2(1 - \phi)^3 + (1 - 2\phi)^3$$

$$P(0,4) = (1 - 3\phi)^4$$

$$P(1,4) = 3(1 - 2\phi)^4 - 3(1 - 3\phi)^4$$

$$P(2,4) = 3(1 - \phi)^4 - 6(1 - 2\phi)^4 + 3(1 - 3\phi)^4$$

$$P(3,4) = 1 - 3(1 - \phi)^4 + 3(1 - 2\phi)^4 - (1 - 3\phi)^4$$

$$P(0,5) = (1 - 4\phi)^5$$

$$P(1,5) = 4(1 - 3\phi)^5 - 4(1 - 4\phi)^5$$

$$P(2,5) = 6(1 - 2\phi)^5 - 12(1 - 3\phi)^5 + 6(1 - 4\phi)^5$$

$$P(3,5) = 4(1 - \phi)^5 - 12(1 - 2\phi)^5 + 12(1 - 3\phi)^5 - 4(1 - 4\phi)^5$$

$$P(4,5) = 1 - 4(1 - \phi)^5 + 6(1 - 2\phi)^5 - 4(1 - 3\phi)^5 + (1 - 4\phi)^5$$

We see that the P(k, n) coefficients become challenging to evaluate accurately by direct numerical methods for large n because the terms become large and fluctuate in sign.

In addition to the general expressions, we shall now list the leading six elements of the  $\alpha$ ,  $\beta$ ,  $\gamma$ ,  $\eta$  and  $\xi$  matrices. These are both pedagogically instructive and also helpful in checking any software implementation of the general expressions. We also give general limiting approximations for the case where the dead time is vanishingly small.

 $\alpha_n$  functions

 $(\alpha_1 - 1) = 0$ 

and for  $n \ge 2$ 

$$(\alpha_n - 1) = \sum_{k=0}^{n-2} \binom{n-1}{k+1} \frac{(k+1)^k \phi^k}{[1 - (k+1)\phi]^{k+2}}$$

(B24)

In factorial notation the Binomial coefficient is given by:

$$\binom{n-1}{k+1} = \frac{(n-1)!}{(k+1)!(n-2-k)!}$$
 (B25)

with the usual convention that 0! = 1.

The leading six terms are:

$$\begin{aligned} \alpha_0 &= 0\\ \alpha_1 &= 1\\ \alpha_2 &= 1 + \frac{1}{(1-\phi)^2}\\ \alpha_3 &= 1 + \frac{2}{(1-\phi)^2} + \frac{2\phi}{(1-2\phi)^3}\\ \alpha_4 &= 1 + \frac{3}{(1-\phi)^2} + \frac{6\phi}{(1-2\phi)^3} + \frac{9\phi^2}{(1-3\phi)^4}\\ \alpha_5 &= 1 + \frac{4}{(1-\phi)^2} + \frac{12\phi}{(1-2\phi)^3} + \frac{36\phi^2}{(1-3\phi)^4} + \frac{64\phi^3}{(1-4\phi)^5} \end{aligned}$$

To first order in  $\phi$  in the limit  $\phi \rightarrow 0$  we have:

 $\alpha_n \approx n(1+1(n-1)\phi + \cdots)$ 

To illustrate the use of this limiting result, let us suppose that the pulse train is only slightly perturbed so that we may approximate the accidentals histogram, to first order, by the unperturbed Poisson count distribution with mean  $\langle n \rangle = \lambda$ . The average singles rate is therefore, to some rough approximation, given by the dead time corrected average of events falling in the accidentals gate per unit time:

(B26)

$$S_c = \frac{\langle \alpha_n \rangle}{T_g} \approx \frac{\langle n \rangle + \langle n(n-1) \rangle \phi}{T_g} + \dots \sim \frac{\lambda + \lambda^2 \phi}{T_g} \sim S_m (1 + S_m d)$$
(B27)

where we have used the definition  $\phi = d/T_g$  and the relation that for a Poisson distribution the second factorial moment is equal to the square of the mean. Thus we see, in this limiting case and subject to this approximate logic that the singles dead time correction behaves as expected. The singles dead time correction factor being of the order of  $(1 + S_m d)$ .

 $\beta_n$  functions

$$\beta_2 = (\alpha_2 - 1) = \frac{1}{(1 - \phi)^2}$$

and for  $n \geq 3$ 

$$\beta_n = (\alpha_n - 1) + \sum_{k=0}^{n-3} \binom{n-1}{k+2} \frac{(k+1)(k+2)^k \phi^k}{[1-(k+2)\phi]^{k+3}}$$
(B28)

In factorial notation the Binomial coefficient is given by:

$$\binom{n-1}{k+2} = \frac{(n-1)!}{(k+2)!(n-3-k)!}$$

(B29)

with the usual convention that 0! = 1.

The leading six terms are:

$$\beta_0 = 0$$
  

$$\beta_1 = 0$$
  

$$\beta_2 = \frac{1}{(1-\phi)^2}$$
  

$$\beta_3 = \frac{2}{(1-\phi)^2} + \frac{1+2\phi}{(1-2\phi)^3}$$
  

$$\beta_4 = \frac{3}{(1-\phi)^2} + \frac{3+6\phi}{(1-2\phi)^3} + \frac{6\phi+9\phi^2}{(1-3\phi)^4}$$
  

$$\beta_5 = \frac{4}{(1-\phi)^2} + \frac{6+12\phi}{(1-2\phi)^3} + \frac{24\phi+36\phi^2}{(1-3\phi)^4} + \frac{48\phi^2+64\phi^3}{(1-4\phi)^5}$$

To first order in  $\phi$  in the limit  $\phi \rightarrow 0$  we have:

$$\beta_n \approx \frac{n(n-1)}{2} (1 + 2(n-1)\phi + \cdots)$$
 (B30)

 $\gamma_n$  functions

$$\gamma_3 = \beta_3 - (\alpha_3 - 1) = \frac{1}{(1 - 2\phi)^3}$$

and for  $n \ge 4$ 

$$\gamma_n = [\beta_n - (\alpha_n - 1)] + \sum_{k=0}^{n-4} \binom{n-1}{k+3}^{\frac{1}{2}(k+1)(k+2)(k+3)^k \phi^k}_{[1-(k+3)\phi]^{k+4}}$$
(B31)

In factorial notation the Binomial coefficient is given as:

$$\binom{n-1}{k+3} = \frac{(n-1)!}{(k+3)!(n-4-k)!}$$
(B32)

with the usual convention that 0! = 1.

The leading six terms are:

$$\gamma_0 = 0$$
  

$$\gamma_1 = 0$$
  

$$\gamma_2 = 0$$
  

$$\gamma_3 = \frac{1}{(1 - 2\phi)^3}$$

$$\gamma_4 = \frac{3}{(1-2\phi)^3} + \frac{1+6\phi}{(1-3\phi)^4}$$
$$\gamma_5 = \frac{6}{(1-2\phi)^3} + \frac{4+24\phi}{(1-3\phi)^4} + \frac{12\phi+48\phi^2}{(1-4\phi)^5}$$

To first order in  $\phi$  in the limit  $\phi \to 0$  we have:

$$\gamma_n \approx \frac{n(n-1)(n-2)}{6} (1 + 3(n-1)\phi + \cdots)$$
 (B33)

 $\eta_n$  functions

$$\eta_4 = \gamma_4 - [\beta_4 - (\alpha_4 - 1)] = \frac{1}{(1 - 3\phi)^4}$$

and for  $n \ge 5$ 

$$\eta_n = \{\gamma_n - [\beta_n - (\alpha_n - 1)]\} + \sum_{k=0}^{n-5} \binom{n-1}{k+4} \frac{\frac{1}{6}(k+1)(k+2)(k+3)(k+4)^k \phi^k}{[1 - (k+4)\phi]^{k+5}}$$
(B34)

In factorial notation the Binomial coefficient is given as:

$$\binom{n-1}{k+4} = \frac{(n-1)!}{(k+4)!(n-5-k)!}$$
(B35)

with the usual convention that 0! = 1.

The leading six terms are:

$$\eta_{0} = 0$$
  

$$\eta_{1} = 0$$
  

$$\eta_{2} = 0$$
  

$$\eta_{3} = 0$$
  

$$\eta_{4} = \frac{1}{(1 - 3\phi)^{4}}$$
  

$$\eta_{5} = \frac{4}{(1 - 3\phi)^{4}} + \frac{1 + 12\phi}{(1 - 4\phi)^{5}}$$

To first order in  $\phi$  in the limit  $\phi \to 0$  we have:

$$\eta_n \approx \frac{n(n-1)(n-2)(n-3)}{24} (1 + 4(n-1)\phi + \cdots)$$
 (B36)

 $\xi_n$  functions

$$\xi_5 = \eta_5 - \{\gamma_5 - [\beta_5 - (\alpha_5 - 1)]\} = \frac{1}{(1 - 4\phi)^5}$$

and for  $n \ge 6$ 

$$\xi_n = (\eta_n - \{\gamma_n - [\beta_n - (\alpha_n - 1)]\}) + \sum_{k=0}^{n-6} \binom{n-1}{k+5} \frac{\frac{1}{24}(k+1)(k+2)(k+3)(k+4)(k+5)^k \phi^k}{[1-(k+5)\phi]^{k+6}}$$
(B37)

In factorial notation the Binomial coefficient is given as:

$$\binom{n-1}{k+5} = \frac{(n-1)!}{(k+5)!(n-6-k)!}$$

with the usual convention that 0! = 1.

The leading six terms are:

$$\xi_0 = 0$$
  

$$\xi_1 = 0$$
  

$$\xi_2 = 0$$
  

$$\xi_3 = 0$$
  

$$\xi_4 = 0$$
  

$$\xi_5 = \frac{1}{(1 - 4\phi)^5}$$

To first order in  $\phi$  in the limit  $\phi \to 0$  we have:

$$\xi_n \approx \frac{n(n-1)(n-2)(n-3)(n-4)}{120} \left(1 + 5(n-1)\phi + \cdots\right)$$
(B39)

In the general case we can write:

$$\pi_{n}^{(m)} = \left(\pi_{n}^{(m-1)} - \left(\pi_{n}^{(m-2)} - \left(\pi_{n}^{(m-3)} - \dots \left(\pi_{n}^{(1)} - 1\right)\right)\right)\right) + \sum_{k=0}^{n-m-1} \binom{n-1}{k+m} \frac{\frac{1}{(m-1)!} (k+1)(k+2)\dots (k+m)^{k} \phi^{k}}{\left[1 - (k+m)\phi\right]^{k+m+1}}$$
(B40)

where  $\alpha_n, \beta_n...$  are generalized as  $\pi_n^{(m)}$ , and  $\pi_n^{(1)} = \alpha_n, \pi_n^{(2)} = \beta_n ...$ 

The first order expansions are only useful for cases in which the histograms extend only to low multiplicity and when the dead time is also small compared to the gate width. The optimum gate width for shift-register multiplicity counting is usually set close to about 1.26 times the system die-away time so as to achieve near optimum precision [20]. The applicability of the first order expansion is potentially very restrictive on the value of  $\phi$ . For instance, for a MSR which score the histograms over the range n = 0 to 511, the in the case of  $\gamma_{511}$  we require  $3 \cdot 510 \cdot \phi =$  $1530 \cdot \phi \ll 1$ . Suppose d = 20 ns and  $T_g = 20$  µs and so  $\phi = 0.001$ . In such an extreme case the first order expansion would certainly NOT be suitable to cover the full depth of the instrumentally available histogram. But, if the highest populated histogram is far below the instrumental maximum, then for this illustrative example there may be a domain where the first order approximations might work to an acceptable degree of accuracy. A higher order Taylor polynomial might provide an adequate approximation for all practical applications of interest provided the measurements remain within certain boundaries e.g.  $n \le 511$  and  $\phi \le 0.01$ . We have not looked at this further however because implementation of the full expressions is straightforward and there seems to be no compelling reason to introduce an approximate approach with penalizing qualifications.