Non-Poisson Counting

Suppose an aliquot of a laboratory sample is being analyzed for a radionuclide and the determinative step of the analysis is a radioassay performed using a radiation counter. The aliquot initially contains N atoms of the analyte, and each of these atoms will produce some nonnegative number of counts C_i during the assay. (N might be very large.) Assume the analyte's decay chain includes one or more short-lived states and that the atom emits radiation of some type when decaying from each state. So, each C_i may be 0, 1, or greater than 1.

Assuming the C_i are independent and have the same mean $E(C_i) = \mu_C$ and variance $V(C_i) = \sigma_C^2$, the index of dispersion*, or Fano factor, for the total number of counts produced by the N atoms is

$$J = \frac{V\left(\sum_{i=1}^N C_i\right)}{E\left(\sum_{i=1}^N C_i\right)} = \frac{\sum_{i=1}^N V(C_i)}{\sum_{i=1}^N E(C_i)} = \frac{N\sigma_C^2}{N\mu_C} = \frac{\sigma_C^2}{\mu_C} \tag{1}$$

So, the index of dispersion J is the same regardless of whether we consider the total counts obtained from all N atoms of the analyte or just the counts produced by a single atom.

Question: What is the index of dispersion J for the number of counts C produced by one hypothetical atom of analyte in the source?

Solution: We need expressions for the mean $E(C)$ and the variance $V(C)$, and both of these can be found by conditioning on the history of the atom H.

$$E(C) = E\big(E(C \mid H)\big) \quad \text{and} \quad V(C) = V\big(E(C \mid H)\big) + E\big(V(C \mid H)\big) \tag{2}$$

For a particular history h,

$$E(C \mid H = h) = \sum_{r \in A_h} \varepsilon_r \quad \text{and} \quad V(C \mid H = h) = \sum_{r \in A_h} \varepsilon_r (1 - \varepsilon_r) \tag{3}$$

where A_h denotes the set of detectable radiations emitted by the atom in history h, and ε_r denotes the instrument's counting efficiency for radiation r.† So, the mean $E(C)$ is given by

$$E(C) = E(E(C \mid H)) = \sum_h \Pr[H = h] E(C \mid H = h) = \sum_h \Pr[H = h] \sum_{r \in A_h} \varepsilon_r \tag{4}$$

where the outer sum is over all possible histories of the atom h, or all histories that produce detectable radiation. The variance $V(C)$ is found as follows:

$$\begin{aligned}
V(C) \quad &= V\big(E(C \mid H)\big) + E\big(V(C \mid H)\big) \\
&= E\big(E(C \mid H)^2\big) - E\big(E(C \mid H)\big)^2 + E\big(V(C \mid H)\big) \\
&= E\big(E(C \mid H)^2 + V(C \mid H)\big) - E(C)^2 \\
&= \sum_h \Pr[H = h]\big(E(C \mid H = h)^2 + V(C \mid H = h)\big) - E(C)^2 \\
&= \sum_h \Pr[H = h]\left(\left(\sum_{r \in A_h} \varepsilon_r\right)^2 + \sum_{r \in A_h} \varepsilon_r (1 - \varepsilon_r)\right) - E(C)^2 \\
&= \sum_h \Pr[H = h]\left(\left(\sum_{r \in A_h} \varepsilon_r\right)^2 - \sum_{r \in A_h} \varepsilon_r^2\right) + \sum_h \Pr[H = h] \sum_{r \in A_h} \varepsilon_r - E(C)^2 \\
&= \sum_h \Pr[H = h]\left(\left(\sum_{r \in A_h} \varepsilon_r\right)^2 - \sum_{r \in A_h} \varepsilon_r^2\right) + E(C) - E(C)^2
\end{aligned} \tag{5}$$

So, J is given by

* The index of dispersion is defined as the ratio of the variance to the mean. For Poisson counting, $J = 1$.

† The symbol $\in$ denotes set membership. So, the sum is over all radiations r contained in the set A_h, which means all radiations emitted in history h.

$$J = \frac{V(C)}{E(C)} = 1 + \frac{S}{E(C)} - E(C) \tag{6}$$

where

$$S = \sum_h \Pr[H = h] \left(\left(\sum_{r \in A_h} \varepsilon_r \right)^2 - \sum_{r \in A_h} \varepsilon_r^2 \right) \quad \text{and} \quad E(C) = \sum_h \Pr[H = h] \sum_{r \in A_h} \varepsilon_r \tag{7}$$

In many cases $E(C)$ is very small, because C is almost always zero. In these cases we can use

$$J = 1 + \frac{S}{E(C)} \tag{6′}$$

Simplifying assumption: Assume the efficiency of the radiation counter is either ε or 0 for each radiation emitted by the decaying atom. Let R denote the number of detectable radiations emitted by the atom (a random variable). For a particular history of the atom, h, let R_h denote the number of detectable radiations emitted in that history (a number). In this case,

$$E(C) = \varepsilon \sum_h \Pr[H = h] R_h = \varepsilon \, E(R) \tag{8}$$

$$S = \varepsilon^2 \sum_h \Pr[H = h] \, (R_h^2 - R_h) = \varepsilon^2 E\big(R^2 - R\big) \tag{9}$$

where

$$E(R) = \sum_h \Pr[H = h] R_h \quad \text{and} \quad E(R^2 - R) = \sum_h \Pr[H = h](R_h^2 - R_h) \tag{10}$$

Notice that $E(R^2 - R) = 0$ unless there are histories h for which $R_h > 1$.

Equation 6 then becomes

$$J = 1 + \varepsilon \left(\frac{E(R^2 - R)}{E(R)} - E(R) \right) \tag{11}$$

and equation 6′ becomes

$$J = 1 + \varepsilon \frac{E(R^2 - R)}{E(R)} \tag{11′}$$

When equation 11′ is valid, it can be used to obtain bounds for the value of J. It is easy to see that

$$R^2 \le R_{\max} R \tag{12}$$

where $R_{\max}$ denotes the maximum possible value of R, or the maximum number of detectable radiations a decaying atom of the analyte can emit. Therefore,

$$E(R^2 - R) \le E(R_{\max} R - R) = (R_{\max} - 1) \, E(R) \tag{13}$$

Equations 11′ and 13 together imply

$$1 \le J \le 1 + \varepsilon (R_{\max} - 1) \tag{14}$$

Example 1: When analyzing a sample for ^{226}Ra by counting emanated ^{222}Rn in a Lucas cell, where $R_{\max} = 3$ and $\varepsilon \approx 0.75$, inequality 14 implies $1 \le J \le 1 + 0.75(3 - 1) = 2.5$.

Example 2: When analyzing a sample for ^{234}Th by beta-counting, where $R_{\max} = 2$ and $\varepsilon \approx 0.5$, inequality 14 implies $1 \le J \le 1 + 0.5(2 - 1) = 1.5$.

Example 3: Consider the ^{226}Ra analysis again. A slightly simplified decay chain for ^{226}Ra is

$$^{226}\text{Ra} \rightarrow {}^{222}\text{Rn} \rightarrow {}^{218}\text{Po} \rightarrow {}^{214}\text{Pb} \rightarrow {}^{214}\text{Bi} \rightarrow {}^{214}\text{Po} \rightarrow {}^{210}\text{Pb}$$

Although ^{210}Pb is not stable, it is relatively long-lived, and we can consider it to be essentially stable when calculating J. Number these states sequentially from 0 to 6. The history of a ^{226}Ra atom in the sample aliquot may now be defined by:

(a) the state, F, of the atom at the time when the Lucas cell is filled;
(b) whether the atom is recovered and captured in the Lucas cell ($Y = 1$ or 0);
(c) the state, B, of the atom at the beginning of the counting measurement; and
(d) the state, T, of the atom at the end of the counting measurement.

We assume that $Y = 0$ unless $F = 1$. I.e., an atom can be recovered only if it happens to be in the ^{222}Rn state when the Lucas cell is filled. Let

t_{I} = time allowed for ingrowth of ^{222}Rn from ^{226}Ra (ending when the Lucas cell is filled);
t_{D} = time from filling of the Lucas cell till counting begins;
t_{S} = count time; and
ε = counting efficiency for alpha-particles.

Then

$$E(R^n) = \sum_{i=0}^{6}\sum_{j=i}^{6}\sum_{k=j}^{6} \Pr[Y=1, F=i, B=j, T=k]\, R_{j,k}^n\,, \quad \text{for } n = 1 \text{ or } 2, \tag{15}$$

where $R_{j,k}$ denotes the number of alpha-particles emitted as an atom decays from state j to state k. We can omit histories where $Y \equiv 0$ (e.g., when $i \neq 1$) or where $R_{j,k} = 0$ (e.g., when $j = 6$ or $k = j$). So,

$$E(R^n) = \sum_{j=1}^{5}\sum_{k=j+1}^{6} \Pr[Y=1, F=1, B=j, T=k]\, R_{j,k}^n \tag{16}$$

and we can calculate the probability of each remaining history as follows:

$$\begin{aligned} \Pr[Y=1, F=1, B=j, T=k] &= \Pr[F=1]\Pr[Y=1 \mid F=1]\Pr[B=j \mid F=1]\Pr[T=k \mid B=j] \\ &= P_{0,1}(t_{\mathrm{I}})\Pr[Y=1 \mid F=1]\, P_{1,j}(t_{\mathrm{D}})\, P_{j,k}(t_{\mathrm{S}}) \end{aligned} \tag{17}$$

where $P_{i,j}(t)$ denotes the function that gives the probability that an atom initially in state i will be in state j after time t has elapsed. So,

$$E(R^n) = P_{0,1}(t_{\mathrm{I}})\Pr[Y=1 \mid F=1] \sum_{j=1}^{5} P_{1,j}(t_{\mathrm{D}}) \sum_{k=j+1}^{6} P_{j,k}(t_{\mathrm{S}})\, R_{j,k}^n \tag{18}$$

In theory,

$$P_{i,j}(t) = \lambda_i \lambda_{i+1} \cdots \lambda_{j-1} \sum_{k=i}^{j} \frac{\mathrm{e}^{-\lambda_k t}}{\prod\limits_{\substack{p=i \\ p \neq k}}^{j} (\lambda_p - \lambda_k)}\,, \quad \text{for } i \leq j, \tag{19}$$

where λ_i is the decay constant for state i, although other formulations may be better for accurate calculations. Note that $P_{0,1}(t_{\mathrm{I}})$ is the probability that an atom of ^{226}Ra (state 0) will be an atom of ^{222}Rn (state 1) after time t_{I} has elapsed, and the long half-life of ^{226}Ra makes this is a very small probability. So, $E(R)$ is very small, and we can estimate J by

$$J = 1 + \varepsilon\,\frac{E(R^2 - R)}{E(R)} = 1 + \varepsilon\,\frac{\sum\limits_{j=1}^{5} P_{1,j}(t_{\mathrm{D}}) \sum\limits_{k=j+1}^{6} P_{j,k}(t_{\mathrm{S}})\,(R_{j,k}^2 - R_{j,k})}{\sum\limits_{j=1}^{5} P_{1,j}(t_{\mathrm{D}}) \sum\limits_{k=j+1}^{6} P_{j,k}(t_{\mathrm{S}})\, R_{j,k}} \tag{20}$$

The following table shows the values of $R_{j,k}$ to be used in equation 20.

$\boldsymbol{R_{j,k}}$

j \ ***k***	**2** ^{218}**Po**	**3** ^{214}**Pb**	**4** ^{214}**Bi**	**5** ^{214}**Po**	**6** ^{210}**Pb**
1 ^{222}**Rn**	1	2	2	2	3
2 ^{218}**Po**	0	1	1	1	2
3 ^{214}**Pb**		0	0	0	1
4 ^{214}**Bi**			0	0	1
5 ^{214}**Po**				0	1

Note that if $j > 2$ or $k < j + 2$, then $R_{j,k}^2 = R_{j,k}$ because $R_{j,k} = 0$ or 1. So,

$$J = 1 + \varepsilon \frac{\sum_{j=1}^{2} P_{1,j}(t_\mathrm{D}) \sum_{k=j+2}^{6} P_{j,k}(t_\mathrm{S})(R_{j,k}^2 - R_{j,k})}{\sum_{j=1}^{5} P_{1,j}(t_\mathrm{D}) \sum_{k=j+1}^{6} P_{j,k}(t_\mathrm{S}) R_{j,k}} \tag{21}$$

Equation 21 still needs simplification to be practical for implementation at a typical lab. With this goal in mind, if we assume t_D is long enough for the radon progeny to reach equilibrium, we can estimate

$$\begin{aligned}\frac{E(R^2 - R)}{E(R)} &= \lim_{t_\mathrm{D}\to\infty} \frac{\sum_{j=1}^{2} P_{1,j}(t_\mathrm{D}) \sum_{k=j+2}^{6} P_{j,k}(t_\mathrm{S})(R_{j,k}^2 - R_{j,k})}{\sum_{j=1}^{5} P_{1,j}(t_\mathrm{D}) \sum_{k=j+1}^{6} P_{j,k}(t_\mathrm{S}) R_{j,k}} = \lim_{t_\mathrm{D}\to\infty} \frac{\sum_{j=1}^{2} \mathrm{e}^{\lambda_1 t_\mathrm{D}} P_{1,j}(t_\mathrm{D}) \sum_{k=j+2}^{6} P_{j,k}(t_\mathrm{S})(R_{j,k}^2 - R_{j,k})}{\sum_{j=1}^{5} \mathrm{e}^{\lambda_1 t_\mathrm{D}} P_{1,j}(t_\mathrm{D}) \sum_{k=j+1}^{6} P_{j,k}(t_\mathrm{S}) R_{j,k}} \\ &= \frac{\sum_{j=1}^{2} \frac{\lambda_1 \lambda_2 \cdots \lambda_{j-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)\cdots(\lambda_j - \lambda_1)} \sum_{k=j+2}^{6} P_{j,k}(t_\mathrm{S})(R_{j,k}^2 - R_{j,k})}{\sum_{j=1}^{5} \frac{\lambda_1 \lambda_2 \cdots \lambda_{j-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)\cdots(\lambda_j - \lambda_1)} \sum_{k=j+1}^{6} P_{j,k}(t_\mathrm{S}) R_{j,k}}\end{aligned} \tag{22}$$

With a reliable algorithm (e.g., Siewers) for $P_{j,k}(t_\mathrm{S})$, this ratio can be calculated accurately. Since the ratio is a function of t_S but not t_D, its values can also be tabulated easily.

t_S / min	$E(R^2 - R) / E(R)$	t_S / min	$E(R^2 - R) / E(R)$
5	0.269	400	1.760
10	0.413	500	1.808
15	0.502	600	1.840
20	0.566	700	1.863
30	0.665	800	1.880
60	0.907	900	1.893
90	1.107	1000	1.904
120	1.264	2000	1.952
150	1.385	3000	1.968
180	1.476	4000	1.975
210	1.547	5000	1.980
240	1.601	6000	1.983
300	1.680	∞	1.994

The limit as $t_\mathrm{S} \to \infty$ is based on the fact that

$$\lim_{t_\mathrm{S}\to\infty} P_{j,k}(t_\mathrm{S}) = \begin{cases} 0 & \text{if } k < 6, \\ 1 & \text{if } k = 6. \end{cases} \tag{23}$$

If we define the cell *calibration factor*, CF, to be the ratio of the expected counts to the expected ^{222}Rn disintegrations in the cell, and if we continue to assume equilibrium of radon and progeny, then

$$CF = \lim_{t_D \to \infty} \frac{\varepsilon \times E(R)}{e^{-\lambda_1 t_D}(1 - e^{-\lambda_1 t_S})} = \frac{\varepsilon}{1 - e^{-\lambda_1 t_S}} \sum_{j=1}^{5} \frac{\lambda_1 \lambda_2 \cdots \lambda_{j-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)\cdots(\lambda_j - \lambda_1)} \sum_{k=j+1}^{6} P_{j,k}(t_S)\, R_{j,k} \tag{24}$$

Although the value of CF here appears to depend on the count time t_S, the assumption of equilibrium means that it does not. The value depends only on the efficiency and on the ratio of the total alpha activity to the ^{222}Rn activity, which remains constant at equilibrium. So, we can take the limit as $t_S \to \infty$ to get

$$CF = \varepsilon \sum_{j=1}^{5} \frac{\lambda_1 \lambda_2 \cdots \lambda_{j-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)\cdots(\lambda_j - \lambda_1)} R_{j,6} = 3.0097\,\varepsilon \tag{25}$$

which implies $\varepsilon = CF\,/\,3.0097$. Alternatively, we can consider an infinitesimal count time $t_S \to 0$ and apply L'Hôpital's Rule to equation 24. The derivatives of $P_{j,k}(t)$ can be calculated using the Maclaurin series:

$$P_{j,k}(t) = \lambda_j \lambda_{j+1} \cdots \lambda_{k-1} \sum_{n=0}^{\infty} \frac{t^{k-j+n}}{(k-j+n)!} h_n(-\lambda_j, -\lambda_{j+1}, \ldots, -\lambda_k) \tag{26}$$

where $h_n(x_1, x_2, \ldots, x_m)$ denotes a *complete homogeneous symmetric polynomial* of degree n. The derivatives at $t = 0$ are given by:

$$P_{j,k}^{(n)}(0) = \begin{cases} 0 & \text{if } n < k - j \\ \lambda_j \lambda_{j+1} \cdots \lambda_{k-1} \times h_{n-k+j}(-\lambda_j, -\lambda_{j+1}, \ldots, -\lambda_k) & \text{if } n \ge k - j \end{cases} \tag{27}$$

In particular, $P'_{j,j+1}(0) = \lambda_j$ and $P'_{j,k}(0) = 0$ if $k > j + 1$. So,

$$\begin{aligned} CF &= \varepsilon \, \frac{\lambda_1 R_{1,2} + \dfrac{\lambda_1}{\lambda_2 - \lambda_1} \lambda_2 R_{2,3} + \dfrac{\lambda_1 \lambda_2 \lambda_3 \lambda_4}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)(\lambda_4 - \lambda_1)(\lambda_5 - \lambda_1)} \lambda_5 R_{5,6}}{\lambda_1} \\ &= \varepsilon \left(1 + \frac{\lambda_2}{\lambda_2 - \lambda_1} + \frac{\lambda_2 \lambda_3 \lambda_4 \lambda_5}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)(\lambda_4 - \lambda_1)(\lambda_5 - \lambda_1)} \right) \\ &= 3.0097\,\varepsilon \end{aligned} \tag{28}$$

> **NOTE**: The factor 3.0097 is the ratio of the total alpha activity to the ^{222}Rn activity at equilibrium, and equation 28 shows that it equals the sum of the equilibrium activity ratios for ^{222}Rn, ^{218}Po, and ^{214}Po.

We also have the mathematically less-than-obvious fact that for any count time t_S,

$$\sum_{j=1}^{5} \frac{\lambda_1 \lambda_2 \cdots \lambda_{j-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)\cdots(\lambda_j - \lambda_1)} \sum_{k=j+1}^{6} P_{j,k}(t_S)\, R_{j,k} = 3.0097\left(1 - e^{-\lambda_1 t_S}\right) \tag{29}$$

which makes $E(R^2 - R)/E(R)$ slightly easier to calculate.

$$\frac{E(R^2 - R)}{E(R)} = \frac{\displaystyle\sum_{j=1}^{2} \frac{\lambda_1 \lambda_2 \cdots \lambda_{j-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)\cdots(\lambda_j - \lambda_1)} \sum_{k=j+2}^{6} P_{j,k}(t_S)(R_{j,k}^2 - R_{j,k})}{3.0097\left(1 - e^{-\lambda_1 t_S}\right)} \tag{30}$$

When we use the actual values of $R_{j,k}$, we see that

$$\frac{E(R^2 - R)}{E(R)} = \frac{2P_{1,3}(t_S) + 2P_{1,4}(t_S) + 2P_{1,5}(t_S) + 6P_{1,6}(t_S) + \dfrac{2\lambda_1}{\lambda_2 - \lambda_1} P_{2,6}(t_S)}{3.0097\left(1 - e^{-\lambda_1 t_S}\right)} \tag{31}$$

To obtain an equation that is more easily implemented in software or a spreadsheet, expand the function $P_{j,k}(t_S)$ in the numerator of equation 30 and combine terms that have the same exponential factors.

$$\frac{E(R^2-R)}{E(R)} = \frac{\displaystyle\sum_{j=1}^{2}\frac{\lambda_1\lambda_2\cdots\lambda_{j-1}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)\cdots(\lambda_j-\lambda_1)}\sum_{k=j+2}^{6}\lambda_j\lambda_{j+1}\cdots\lambda_{k-1}(R_{j,k}^2-R_{j,k})\sum_{i=j}^{k}\frac{\mathrm{e}^{-\lambda_i t_\mathrm{S}}}{\prod_{\substack{p=j\\p\neq i}}^{k}(\lambda_p-\lambda_i)}}{3.0097\left(1-\mathrm{e}^{-\lambda_1 t_\mathrm{S}}\right)}$$

$$= \frac{\sum_{i=1}^{6}a_i\mathrm{e}^{-\lambda_i t_\mathrm{S}}}{1-\mathrm{e}^{-\lambda_1 t_\mathrm{S}}} = \frac{a_6+\sum_{i=1}^{5}a_i\mathrm{e}^{-\lambda_i t_\mathrm{S}}}{1-\mathrm{e}^{-\lambda_1 t_\mathrm{S}}} \tag{32}$$

where

$$a_i = \frac{1}{\alpha_\mathrm{eq}}\sum_{j=1}^{\min(i,2)}\sum_{k=\max(i,j+2)}^{6}\frac{\lambda_1\lambda_2\cdots\lambda_{k-1}(R_{j,k}^2-R_{j,k})}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)\cdots(\lambda_j-\lambda_1)\prod_{\substack{p=j\\p\neq i}}^{k}(\lambda_p-\lambda_i)} \tag{33}$$

and where α_eq is the equilibrium alpha activity ratio:

$$\alpha_\mathrm{eq} = 1+\frac{\lambda_2}{\lambda_2-\lambda_1}+\frac{\lambda_2\lambda_3\lambda_4\lambda_5}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)(\lambda_4-\lambda_1)(\lambda_5-\lambda_1)} = 3.0097 \tag{34}$$

Note: If we try a similar trick on the denominator of equation 22, expanding $P_{j,k}(t_\mathrm{S})$ and combining exponential terms, equation 29 shows that we get

$$\sum_{j=1}^{\min(i,5)}\sum_{k=\max(i,j+1)}^{6}\frac{\lambda_1\lambda_2\cdots\lambda_{k-1}R_{j,k}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)\cdots(\lambda_j-\lambda_1)\prod_{\substack{p=j\\p\neq i}}^{k}(\lambda_p-\lambda_i)} = \begin{cases}-\alpha_\mathrm{eq} & \text{if } i=1,\\ \alpha_\mathrm{eq} & \text{if } i=6,\\ 0 & \text{otherwise.}\end{cases}$$

If we define $M = (E(R^2-R)/E(R))\,/\,\alpha_\mathrm{eq}$, then we have

$$J = 1+CF\times M \qquad \text{where} \qquad M = \frac{c_6+\sum_{i=1}^{5}c_i\mathrm{e}^{-\lambda_i t_\mathrm{S}}}{1-\mathrm{e}^{-\lambda_1 t_\mathrm{S}}} \tag{35}$$

and where $c_i = a_i\,/\,\alpha_\mathrm{eq}$. The coefficients c_i are listed below.

$c_1 = -0.666\,536\,563\,852$	$c_4 = -0.004\,843\,321\,446\,08$
$c_2 = 0.000\,126\,146\,641\,28$	$c_5 \approx 0$
$c_3 = 0.008\,737\,065\,085\,14$	$c_6 = 0.\,662\,516\,673\,571$

Notice that $M \to c_6$ as $t_\mathrm{S} \to \infty$. So, an upper bound for J is $1 + CF \times c_6$, and since, $CF \le 3.0097$, J never exceeds 2.994. (Note: Although one might expect the maximum value to be exactly 3, it is slightly less than 3, because ^{222}Rn atoms that decay to ^{218}Po before counting starts will generate fewer than 3 counts.)

Examining the coefficients c_i, we see that the short half-life ($162.3\ \mu\mathrm{s}$) of ^{214}Po makes the value of c_5 so small that we can neglect the corresponding term $c_5\mathrm{e}^{-\lambda_5 t_\mathrm{S}}$ in the equation for M. The short half-life also makes the exponential factor $\mathrm{e}^{-\lambda_5 t_\mathrm{S}}$ tiny for any feasible count time t_S. So, we end up with the following equations, which are easily implemented in an electronic spreadsheet.

$$J = 1+CF\times M \qquad \text{where} \qquad M \approx \frac{c_6+\sum_{i=1}^{4}c_i\mathrm{e}^{-\lambda_i t_\mathrm{S}}}{1-\mathrm{e}^{-\lambda_1 t_\mathrm{S}}} \tag{36}$$

For tiny values of t_S, equation 36 in practice may generate severe rounding errors. An application of L'Hôpital's Rule to equation 31, using the Maclaurin series to differentiate $P_{j,k}(t)$, gives the limit:

$$\lim_{t_\mathrm{S}\to 0}\frac{E(R^2-R)}{E(R)} = 0 \tag{37}$$

So, $M \to 0$ and $J \to 1$ as $t_\mathrm{S} \to 0$.

A double application of L'Hôpital's Rule to M / t_S, still using equation 31 for $E(R^2 - R) / E(R)$, leads to the following limit:

$$\lim_{t_\mathrm{S}\to 0} \frac{M}{t_\mathrm{S}} = \lim_{t_\mathrm{S}\to 0} \frac{E(R^2 - R)/E(R)}{t_\mathrm{S}\alpha_\mathrm{eq}} = \frac{2P''_{1,3}(0)}{2\lambda_1\alpha_\mathrm{eq}^2} = \frac{\lambda_2}{\alpha_\mathrm{eq}^2} \approx 0.02492\ \mathrm{min}^{-1} \tag{40}$$

The time t_S must be no more than a few seconds to make this limit useful for approximating M. The same value for the limit can be found by differentiating equation 31 and applying L'Hôpital's Rule once, although that approach may require a little more work.

Example 4: Consider the ^{234}Th analysis again. A simplified decay chain for ^{234}Th is

$$^{234}\mathrm{Th} \to {}^{234\mathrm{m}}\mathrm{Pa} \to {}^{234}\mathrm{U}$$

where the 0.16 % branch to ^{234}Pa has been ignored. The half-life of ^{234}U is so long that we can consider it to be essentially stable. If we apply all the same tricks as in example 3 to beta-counting ^{234}Th and $^{234\mathrm{m}}$Pa in equilibrium, we get

$$\begin{aligned} \frac{E(R^2 - R)}{E(R)} &= \frac{\displaystyle\sum_{j=0}^{0} \frac{\lambda_0\lambda_1\cdots\lambda_{j-1}}{(\lambda_1-\lambda_0)(\lambda_2-\lambda_0)\cdots(\lambda_j-\lambda_0)} \sum_{k=j+2}^{2} P_{j,k}(t_\mathrm{S})(R_{j,k}^2 - R_{j,k})}{\beta_\mathrm{eq}\left(1-\mathrm{e}^{-\lambda_0 t_\mathrm{S}}\right)} \\ &= \frac{P_{0,2}(t_\mathrm{S})(R_{0,2}^2 - R_{0,2})}{\beta_\mathrm{eq}\left(1-\mathrm{e}^{-\lambda_0 t_\mathrm{S}}\right)} \\ & \frac{2\lambda_0\lambda_1}{\beta_\mathrm{eq}\left(1-\mathrm{e}^{-\lambda_0 t_\mathrm{S}}\right)} \sum_{i=0}^{2} \frac{\mathrm{e}^{-\lambda_i t_\mathrm{S}}}{\prod_{\substack{p=0 \\ p\neq i}}^{2} (\lambda_p - \lambda_i)} \end{aligned} \tag{41}$$

where $\lambda_0 = \lambda(^{234}\mathrm{Th})$, $\lambda_1 = \lambda(^{234\mathrm{m}}\mathrm{Pa})$, $\lambda_2 = \lambda(^{234}\mathrm{U}) \approx 0$, and where β_eq is the ratio of the total beta activity to the ^{234}Th activity at equilibrium:

$$\beta_\mathrm{eq} = 1 + \frac{\lambda_1}{\lambda_1 - \lambda_0} = \frac{2\lambda_1 - \lambda_0}{\lambda_1 - \lambda_0} \tag{42}$$

Algebraic manipulation produces the following:

$$\frac{E(R^2 - R)}{E(R)} = \frac{2}{\beta_\mathrm{eq}(\lambda_1 - \lambda_0)}\left(\lambda_1 - \lambda_0 \frac{1-\mathrm{e}^{-\lambda_1 t_\mathrm{S}}}{1-\mathrm{e}^{-\lambda_0 t_\mathrm{S}}}\right) = \frac{2}{2\lambda_1 - \lambda_0}\left(\lambda_1 - \lambda_0 \frac{1-\mathrm{e}^{-\lambda_1 t_\mathrm{S}}}{1-\mathrm{e}^{-\lambda_0 t_\mathrm{S}}}\right) \tag{43}$$

If the beta-particle counting efficiency is ε, then

$$J = 1 + \varepsilon\frac{E(R^2 - R)}{E(R)} = 1 + \frac{2\varepsilon}{2\lambda_1 - \lambda_0}\left(\lambda_1 - \lambda_0 \frac{1-\mathrm{e}^{-\lambda_1 t_\mathrm{S}}}{1-\mathrm{e}^{-\lambda_0 t_\mathrm{S}}}\right) \tag{44}$$