MULTI-CHANNEL STATISTICAL ANALYSIS OF LOW-LEVEL RADIOACTIVITY IN THE PRESENCE OF BACKGROUND COUNTS

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ABSTRACT

This paper presents a multi-channel extension to the Poisson-based Bayesian statistical analysis of the net count rate in a sample. The derivation gives the net count rate probability density distribution in analytical form. In addition, it presents a Normal approximation that is appropriate when the number of counts is large. The new analysis can exhibit a lower false positive probability than methods that consider the counts for each peak separately or sum the counts for all the expected peaks associated with a radioactive substance.

INTRODUCTION

Little (1982), followed by Prosper (1988), Loredo (1990), Fröhner (1997), Groer (2002), and Loparco and Mazziotta (2011), introduced an exact Bayesian analysis for the point and interval estimation of the background and net count rates. Those papers assumed that the net count rate is positive and therefore did not address the decision rule.

The current paper presents an extension of the Bayesian method to obtain the multi-channel analysis of spectra and provides a multi-channel decision rule.

One of the goals of radiation counting statistics is to make inferences about the presence or absence of radioactive substances. The usual and simplest way to deal with spectra is to define a region of interest and sum all the counts in this region. In this manner, the analyst can use one of the single channel decision rules described by Strom and MacLellan 2001. Alternatively, each peak is analyzed separately, and if the number of counts exceeds the decision rule for any of them, the spectrum is examined to check if other expected peaks are present. Ideally, a decision rule should consider all the peaks simultaneously and should account for the way the net counts are distributed between channels. Taking into consideration the distribution of the counts between channels forms the basis of the methodology presented in this paper.

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METHODS

Exact Bayesian analysis

The derivation follows the notation of Little (1982), in which roman letters represent measurements and greek letters are true values, but is based on the equations of Jaynes (1976) and Fröhner (1997).

The blank and sample measurements in each channel \( i \) are described by their respective Poisson distributions, eqns (1) and (2).

\[
P(x_{i1} \mid \mu_i s_i) = \frac{e^{-\mu_i s_i} (\mu_i s_i)^{y_{i1}}}{x_{i1}!} \tag{1}
\]

\[
P(y_{i1} \mid \omega_i t_i) = \frac{e^{-\omega_i t_i} (\omega_i t_i)^{y_{i1}}}{y_{i1}!} \tag{2}
\]

In these equations, \( x_{i1} \) represents the number of background counts recorded in channel \( i \) for a count time \( s_i \), and \( y_{i1} \) is the number of sample counts recorded in channel \( i \) for a count time \( t_i \). The background count rate in channel \( i \) is given by \( \mu_i \) and the sample count rate is given by \( \omega_i \).

Fröhner (1997) showed that the posterior calculated with Bayes’ theorem, eqn (3), is a gamma distribution when a scale invariant prior distribution \( P(\mu_i \mid x_{i0}, s_0) = \mu_i^{-1} \) is selected.

\[
P(\mu_i \mid x_{i0}, y_{i1}, s_0, s_i) = \frac{P(x_{i1} \mid \mu_i s_i)P(\mu_i \mid x_{i0}, s_0)}{\int_0^\infty P(x_{i1}, s_i \mid \mu_i)P(\mu_i \mid x_{i0}, s_0) d\mu_i} \tag{3}
\]

Furthermore, chain application of Bayes’ theorem with a gamma distribution prior and a new set of measurements, also gives a gamma distribution posterior. The gamma distribution is the conjugate prior to the Poisson distribution. This motivates the use of gamma functions for the prior probability distribution of the background and sample, eqns (4) and (5). The prior used by Fröhner (1997) is obtained by setting \( x_{i0} = 0 \) and taking the limit \( s_0 \to 0 \). The uniform prior used by Little (1982) is obtained by setting \( x_{i0} = 1 \) and \( s_0 = 0 \).

\[
P(\mu_i \mid x_{i0}, s_0) = \frac{e^{-\mu_i s_0} s_0^{x_{i0}} \mu_i^{y_{i0} - 1}}{\Gamma(x_{i0})} \tag{4}
\]

\[
P(\omega_i \mid y_{i0}, t_0) = \frac{e^{-\omega_i t_0} t_0^{y_{i0}} \omega_i^{y_{i0} - 1}}{\Gamma(y_{i0})} \tag{5}
\]
Having defined $x_i = x_{0i} + x_{ii}$ as the sum of prior and measured counts for the background measurement in channel $i$, $s = s_0 + s_1$, the sum of prior and measured count time for the background measurement, $y_i = y_{0i} + y_{ii}$, the sum of prior and measured counts for the sample measurement in channel $i$, $t = t_0 + t_1$, the sum of prior and measured count time for the sample measurement, the posteriors are a pair of gamma distributions, $P(\mu_i \mid x_i, s)$ and $P(\omega_i \mid y_i, t)$.

Jaynes (1976) gave a Bayesian equation for the probability that $\omega_i$, the sample count rate in a single channel $i$, is greater than the background $\mu_i$. The solution to this equation gives a decision rule that does not rely on the null hypothesis; therefore it is a departure from traditional decision rules (Strom and MacLellan 2001).

$$P(\omega_i > \mu_i) = \int_0^\infty P(\mu_i \mid x_i, s) \left\{ \int_0^\infty P(\omega_i \mid y_i, t) d\omega_i \right\} d\mu_i$$ (6)

Jaynes’ equation can be rewritten without loss of generality by assuming a net count rate $\Delta$ added to the background count rate $\mu_i$.

$$\omega_i = \mu_i + \Delta$$ (7)

$$P(\omega_i > \mu_i) = \int_0^\infty P(\mu_i \mid x_i, s) \left\{ \int_0^\infty P(\mu_i + \Delta \mid y_i, t) d\Delta \right\} d\mu_i$$ (8)

The background $\mu_i$ can take any value between zero and infinity. Since $\omega_i$ can also take any value between zero and infinity, then $\Delta$ can therefore take any value between $-\infty$ and $+\infty$.

Little (1982) and Fröhner (1997) assumed that $\Delta$ is positive and used a binomial expansion to factor the parameters $\mu_i$ and $\Delta$. A general solution that assumes no prior knowledge about the presence of net activity in the sample and that covers the range $-\infty < \Delta < \infty$ can be obtained by solving the problem in two parts: when $\omega_i > \mu_i$, $\Delta = \omega_i - \mu_i$. When the converse is true, $\Delta' = \mu_i - \omega_i = -\Delta$, and the equations are modified accordingly. Both partial solutions are combined to obtain the solution over the full range of values for $\Delta$.

$$P(\omega_i > \mu_i) = \int_0^\infty P(\mu_i \mid x_i, y_i, s, t) d\mu_i \times \int_0^\infty P(\Delta \mid x_i, y_i, s, t) d\Delta$$ (9)

The integral over $\mu_i$ is absorbed in the normalization of the function of $\Delta$ and the result gives a general expression for the Bayesian decision rule: the probability that the sample contains net activity is simply the integral of the probability distribution for the net count rate over all positive values of $\Delta$. Calculating the probability distribution
\( P(\Delta) \) gives both the decision rule and the point and interval estimates\(^\dagger\).

\[
P(\omega_t > \mu_t) = P(\Delta > 0) = \int_0^\infty P(\Delta | x_i, y_i, s, t) d\Delta
\]  

(10)

Up to now, the derivation has considered a single channel at a time. The multi-channel solution requires considering \( N \) channels simultaneously. In the multi-channel case, the true values \( \mu_t \) for the background count rate are independent. In contrast, the true value of the count rate in the sample \( \omega_t \) is proportional to a single value of the net count rate \( \Delta \) for all the channels, as shown in eqn (11).

\[
\omega_t = \mu_t + \Delta \cdot p_i
\]  

(11)

The parameter \( p_i \) is the probability of detecting a net count in channel \( i \) when the radioactive substance is present. It can be determined empirically by measurement, or it can be calculated from the multiplicity (or yield) of the peak \( m_i \) and the absolute peak efficiency of the detector \( \varepsilon_i \), as in eqn (12). Thus the method is applicable to arbitrary gamma emission spectra.

\[
p_i = \frac{\varepsilon_i \cdot m_i}{\sum_{i=1}^N \varepsilon_i \cdot m_i}
\]  

(12)

The derivation considers the solution for \( \Delta > 0 \) first. The required probability distributions for \( \mu_t \) and \( \Delta \) is calculated from the joint probability of observing \( x_i \) and \( y_i \) counts, given \( \mu_t \) and \( \Delta \), and Bayes’ theorem:

\[
P(\mu_t, \Delta | x_{i0}, x_i, y_{i0}, y_i) = \frac{P(x_{i0}, y_{i0} | \mu_t, \Delta)P(\mu_t, \Delta | x_{i0}, y_{i0})}{\int_0^\infty \int_0^\infty P(x_{i0}, y_{i0} | \mu_t, \Delta)P(\mu_t, \Delta | x_{i0}, y_{i0}) d\mu_t d\Delta}
\]  

(13)

Replacing the Poisson and gamma distributions for \( \mu_t \) and \( \omega_t \) in eqn (13), the joint probability distribution for \( \mu_t \) and \( \Delta \), given the data \( x_{i0}, y_{i0}, x_i, y_i \) is given in eqn (14). The term in the denominator of eqn (13) is not calculated since the normalization of eqn (14) makes the double integral equal to one.

\[
P(\mu_t, \Delta | x_i, y_i) \propto \prod_{i=1}^N e^{-\mu_t \cdot s_i} \mu_t^{y_i - 1} e^{-\mu_t \cdot \Delta \cdot p_i \cdot y_i} (\mu_t + \Delta \cdot p_i)^{y_i - 1}
\]  

(14)

Expanding \((\mu_t + \Delta \cdot p_i)^{y_i - 1}\) using the binomial distribution.

\(^\dagger\) To verify this assertion, eqn (6) was solved analytically and it gave the same result as the solution based on the net count rate, eqn (10).
where the binomial coefficient is
\[
\binom{a}{b} = \frac{a!}{(a-b)!b!}
\]  

The probability distribution for \( \Delta \) is obtained by integration over all the \( \mu_i \) variables, as nuisance parameters (Box and Tiao 1973).

The probability distribution for \( \mu_i \) is obtained by integration over \( \Delta \) and \( \mu_j \), where \( j \neq i \). The integrals over cancel \( \mu_j \) in the normalization factor therefore the true value of \( \mu_i \) depends solely on the measurements in channel \( i \).

The integrals in eqns (18) and (19) are of the form
\[
\int_0^\infty e^{-\Delta} \Delta^z d\Delta = t^{(z+1)}z!
\]  

After recasting the summation to run from \( z_i = 1 \ldots y_i \), and using the equivalence between \( \Gamma(z) = (z-1)! \) when \( z \) is an integer, the net count rate distribution for \( \Delta \) is given by eqn (21). The expression inside the square bracket is a gamma function, the same that was used as a prior for \( \mu_i \) and \( \omega_j \), although in this equation it is function of the net counts \( z_i \).

In this form, the term inside the curly bracket can be approximated by the positive part of a Normal distribution (Fig. 1). The result is a probability distribution proportional to the product of Normal distributions. This approximation is discussed in the next section.
The final form of the Poisson-based solution needs to be expressed as a single summation of gamma functions that can be normalized when integrated over all values of \( \Delta \). Eqn (21) is written as the product of polynomials in \( \Delta \).

\[
P(\Delta | x_i, y_i) \propto \prod_{i=1}^{N} e^{-P_i \Delta} \sum_{z_i=1}^{V_i} C_i(z_i) \cdot \Delta^{z_i-1}
\]

where

\[
C_i(z_i) = \left( \frac{x_i + y_i - z_i - 1}{y_i - z_i} \right) \times \frac{s^{y_i} t^{x_i}}{(s + t)^{x_i + y_i - z_i}} \times \frac{\Delta^{z_i-1}}{(z_i - 1)!}
\]

To obtain the coefficients for each power of \( \Delta \), eqn (22) is written in matrix notation:

\[
P(\Delta | x_i, y_i) \propto \begin{pmatrix} f_1 & f_2 & \cdots & f_y \end{pmatrix} \begin{pmatrix} 1 \\ \Delta \\ \vdots \\ \Delta^{y-1} \end{pmatrix} e^{-\Delta \cdot t}
\]
\[
\begin{pmatrix}
  f_1 \\
  f_2 \\
  \vdots \\
  f_y
\end{pmatrix} =
\begin{pmatrix}
  C_N(1) & 0 & 0 & 0 \\
  C_N(2) & C_N(1) & 0 & 0 \\
  \vdots & \vdots & \vdots & \ddots \\
  C_N(3) & C_N(2) & C_N(1) & 0 \\
\end{pmatrix}
\begin{pmatrix}
  f_1 \\
  f_2 \\
  \vdots \\
  f_y
\end{pmatrix}
\begin{pmatrix}
  C_1(1) \\
  C_2(1) \\
  \vdots \\
  C_y(1)
\end{pmatrix}
\] (25)

Where the vector \( f_z \) contains the coefficients of the polynomial in \( \Delta \) and the coefficients \( C_1(z_1), C_2(z_2), \ldots, C_N(z_N) \) are the polynomial coefficients for each of the channels from 1 to \( N \). The matrices for each channel are square, of size \( y \times y \), where \( y = \sum_{i=1}^{N} y_i \). Note that these matrices of coefficients are sparse. If channel \( i \) has only \( y_i \) counts, only the first \( y_i \) values of the vector or matrix will be filled with coefficients. The generating function for the matrix corresponding to channel \( i \) is

\[
a_i(\text{row}, \text{col}) = C_i(\text{row})
\]

\[
a_i(\text{row} + (\text{col} - 1), \text{col}) = C_i(\text{row})
\] (26)

for \( \text{row} = 1 \) to \( y_i \), \( \text{col} = 1 \) to \( \sum_{i=1}^{N} y_i \).

The resulting solution is a polynomial in powers of \( \Delta \), where \( z = \sum_{i=1}^{N} z_i \) is the total number of net counts for all channels.

\[
P(\Delta | x_i, y_i) \propto e^{-\Delta} \sum_{z=1}^{y} f_z \cdot \Delta^{-1}
\] (27)

This polynomial can be written as the normalized sum of gamma functions

\[
P(\Delta | x_i, y_i) = \sum_{z=1}^{y} g_z \cdot \left[ \frac{e^{-\Delta t} \Delta^{-1}}{(z-1)!} \right]
\] (28)

\[
g_z = \frac{f_z \cdot (z-1)! t^{-z}}{\sum_{z=1}^{y} f_z \cdot (z-1)! t^{-z}}
\] (29)

Each coefficient \( g_z \) in the summation in eqn (28) represents the probability that a total of \( z \) net counts will be observed. These coefficients are proportional to the probability that \( z_i \) counts will be observed in channel \( i \), given the measurements \( x_i, y_i \) (Fröhner 1997). By inspection of eqns (23) and (29), it is apparent that the coefficients \( g_z \) include the following factors

\[
g_z \propto p_1^{(z_1-1)} p_2^{(z_2-1)} \ldots p_N^{(z_N-1)} \frac{(z-1)!}{(z_1-1)!(z_2-1)\ldots(z_N-1)!}
\] (30)
This is the multinomial function and it gives its name to this solution. It should be noted that the probability density at $\Delta = 0$ is given by the gamma function for $z = 1$. The value $z = 0$ does not contribute to the net count rate probability distribution and is not included in the summation (Fröhner 1997).

Eqns (28) and (29) are the solution for $\Delta > 0$. The solution for $\Delta < 0$ is obtained by swapping the $x$ and $y$ variables and the $s$ and $t$ variables. This gives a new set of coefficients $C'_i(z_i)$.

$$C'_i(z_i) = \left( x_i + y_i - z_i - 1 \right) \times \frac{s^y t^z}{(s + t)^{y_i + z_i}} \times \frac{p_{i}^{z_i - 1}}{(z_i - 1)!} \quad (31)$$

These coefficients, entered into matrices, are used to calculate the coefficients $f'_z$. The final solution for positive and negative values of $\Delta$ is given by the coefficients $g_z$ and $g'_z$, and gamma functions $P(\Delta, z, t)$.

$$P(\Delta \mid x_i, y_i) = \sum_{z=1}^{z} g'_z \cdot P(-\Delta \mid z, t) + \sum_{z=1}^{z} g_z \cdot P(\Delta \mid z, t) \quad (32)$$

$$P(\Delta \mid z, t) = \frac{e^{-\Delta t \Delta^{-1}}}{(z - 1)!} \quad (33)$$

$$g_z = \frac{f'_z \cdot (z - 1)! t^{-z}}{\sum_{z'=1}^{z} f'_{z'} \cdot (z'-1)! t^{-z'} + \sum_{z''=1}^{z} f''_{z'} \cdot (z''-1)! t^{-z''}} \quad (34)$$

When the net count rate is known to be positive (the probability that net activity is present in the sample is one), eqns (28) and (29) should be used instead of eqns (32), (33) and (34). An Excel spreadsheet that contains Visual Basic macros implementing these equations is available from the author‡.

**Normal approximation**

Eqn (21) can be approximated by a product of Normal distributions.

$$P(\Delta \mid x_i, y_i) \propto \prod_{i=1}^{N} \frac{1}{\sqrt{2\pi v_i}} \exp \left( -\frac{(p_i \cdot \Delta - r_i)^2}{2v_i} \right) \quad (35)$$

$$r_i = \frac{y_i}{t} - \frac{x_i}{s} \quad (36)$$

‡ [www.i-s-r.ca/statistics.html](http://www.i-s-r.ca/statistics.html)
Eqn (35) can be recast as the product of Normal distributions with individual means \( r_i \cdot p_i^{-1} \) and variances \( v_i \cdot p_i^{-2} \).

\[
P(\Delta | x_i, y_i) \propto \exp \left( - \sum_{i=1}^{N} \frac{(\Delta - r_i \cdot p_i^{-1})^2}{2v_i \cdot p_i^{-2}} \right)
\]  

Expanding the term in the exponent

\[
\sum_{i=1}^{N} \frac{(\Delta - r_i \cdot p_i^{-1})^2}{2v_i \cdot p_i^{-2}} = \frac{1}{2} \left[ \Delta^2 \sum_{i=1}^{N} \frac{1}{v_i \cdot p_i^{-2}} - 2\Delta \sum_{i=1}^{N} \frac{r_i \cdot p_i^{-1}}{v_i \cdot p_i^{-2}} + \sum_{i=1}^{N} \frac{r_i^2 \cdot p_i^{-2}}{v_i \cdot p_i^{-2}} \right]
\] 

Dividing by the coefficient of \( \Delta \), gives a Normal distribution, eqn (42), with mean and variance given by eqns (40) and (41).

\[
r = \frac{\sum_{i=1}^{N} r_i \cdot p_i}{\sum_{i=1}^{N} \frac{p_i^2}{v_i}}
\]  

\[
v = \frac{1}{\sum_{i=1}^{N} \frac{p_i^2}{v_i}}
\]

\[
P(\Delta | x_i, y_i) = \frac{1}{\sqrt{2\pi v}} \exp \left( -\frac{(\Delta - r)^2}{2v} \right)
\]  

When the net count rate probability distribution spans the full range \(-\infty < \Delta < \infty\), the mean and variance for the net count rate distribution are given by the net count rate parameters calculated from the measurements.

\[
\bar{\Delta} = r
\]  

\[
\sigma_{\Delta}^2 = v
\]

If the sample is known to contain net activity, the tail of the Normal distribution for \( \Delta < 0 \) is truncated and the resulting distribution re-normalized (Little 1982). The mean and standard deviation of the net count rate distribution are then given by

\[
\bar{\Delta} = r + \sqrt{v} \cdot \frac{g_0}{1 - p_o}
\]
\[
\sigma_\Delta^2 = v - (\Delta - r) \cdot \Delta 
\]
\[
p_0 = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{-\frac{r^2}{2\sigma^2}} d\tau 
\]
\[
g_0 = \frac{1}{\sqrt{2\pi}} \exp \left( -\frac{r^2}{2\sigma^2} \right) 
\]

The solution given by eqns (40) and (41) is similar to the equations that Fröhner (1997) and Heisel et al. (2009) obtained.

The net count rate can be converted to activity by dividing by the efficiency factor \( \varepsilon \) for the channels included in the calculation.

\[
\varepsilon = \sum_{i=1}^{N} \varepsilon_i \cdot m_i 
\]

Net Activity [Bq kg\(^{-1}\)] = \( \Delta \) \( \frac{\varepsilon}{\varepsilon} \) (50)

False positive rate

It seems plausible that the multi-channel methods proposed here should have a lower false positive rate than the single channel methods. To verify this, it is necessary to derive a multi-channel extension to the equation for the probability of false positive introduced by Strom and MacLellan (2001).

\[
P(y > DR \mid \mu_t) = \sum_{x_1=0}^{\infty} \sum_{x_2=0}^{\infty} \ldots \sum_{x_N=0}^{\infty} \prod_{j=1}^{N} P(x_i \mid \mu_t) 
\]

\[
\times \left[ \sum_{x_1=0}^{\infty} \sum_{x_2=0}^{\infty} \ldots \sum_{x_N=0}^{\infty} \text{if DR is true add} \left\{ \prod_{i=1}^{N} P(y_i \mid \mu_t) \right\} \right] 
\]

\[
P(x_i \mid \mu_t) = \frac{e^{-\mu_t} (\mu_t)^x}{x!} 
\]

\[
x = \sum_{i=1}^{N} x_i 
\]

\[
y = \sum_{i=1}^{N} y_i 
\]

As in Strom and MacLellan’s equation, the first set of summations represents the probability of observing the given background counts, and the second set of summations represents the probability of observing background counts that exceed the decision rule. DR represents the decision rule for accepting the presence of net activity.
For illustration purposes, the example assumes two channels \((N = 2)\), having the same background \(\mu_1 = \mu_2\) and the same probability of detection in each channel, \(p_1 = 0.5\) and \(p_2 = 0.5\). To allow comparison of Fig. 2 to Strom and MacLellan’s curves, the total background when both channels are summed is \(\mu = \sum_{i=1}^{N} \mu_i\) and is plotted on the horizontal axis. The vertical axis shows the probability of false positive, when measuring the background twice (once for the background \(x_i\), once for the unknown sample \(y_i\)).

![Graph showing false positive rate as a function of background level](image)

**Fig. 2. False positive rate as a function of background level**

The curve labeled “Currie” implements the traditional decision rule (DR) based on eqn (55) (Currie 1984), and calculates the false positive rate with the multi-channel extension of Strom and MacLellan’s equation, eqn (51). If \((y \geq x + L_c)\), then the content of the curly bracket is added to sum; if it is less, it is not added. The dotted curve is identical to the curve published in Strom and MacLellan (2001).

\[ L_c = 1.645\sqrt{2x} \]

\[ (55) \]

The other curves use the Bayesian decision rule (DR), were the probability that the sample contains net activity above background is calculated from the measurements \(x_i\) and \(y_i\). If \(\int_0^{\infty} P(\Delta|x_i,y_i) d\Delta > 0.95\), then the content of the bracket is added to the sum; if it is less, then it is not.
The curve labeled “Normal (sum channels)” implements the single channel Normal decision rule (DR) for the sum of the counts in all channels. This curve is identical to the curve obtained by Strom and MacLellan (2001) for the decision rule of Altshuler and Pasternak 1963.

The false positive rate rolls-off for low background values ($\mu t < 1$) because the Poisson distribution peaks at $x = 0$ when the background values are in that range. It is not possible to devise a decision rule that performs equally well for all values of $\mu t < 1$, given that the most likely measurement is $x = 0$. The Bayesian approach looks at the problem from a different perspective. It does not start from a known value of $\mu t$, but estimates a possible range of low background count rate values compatible with a given measurement.

The curve labeled “Normal (either channel)” implements the single channel Normal decision rule (DR) for each channel individually. If the decision rule is true in either channel, the radioactive substance is detected. As can be expected, the false positive rate is twice as much as when the counts are summed.

The curve labeled “Multi-Normal” implements the Multi-Normal decision rule (DR) proposed here. This curve shows a lower false positive rate when the background is low ($\mu t < 1$), but gives the expected false positive rate ($\alpha$) when the background is high.

In summary, Fig. 2 shows that the Multi-Normal decision rule gives a false positive rate that is lower or equal to the single channel decision rule.

**RESULTS**

**Numerical example**

The multi-channel analysis methodology is first demonstrated with a numerical example involving the detection of $^{60}$Co activity in a generic spectroscopy system.

In this example, the radiation counter detects gamma rays from $^{60}$Co disintegration at 1.173 MeV and 1.333 MeV. It is assumed that the counter has two counting channels centered on each peak, such that photons from the 1.173 MeV peak are registered in channel 1, and those from the 1.333 MeV peak are registered in channel 2. Both gammas are emitted in cascade with essentially the same probability (99.91% and 100%). Assuming the efficiency of the detector is identical at both energies, the probability of registering a photon from $^{60}$Co in either channel is the same.

Two sets of analysis data were created, as shown in Table 1. The first corresponds to a false positive and the second to a sample that contains $^{60}$Co. A scale invariant prior is assumed in both cases.
Table 1. Prior and measurement data for the background and two sample analyses

<table>
<thead>
<tr>
<th>Peak</th>
<th>Background Counts</th>
<th>Background Time (ks)</th>
<th>Sample (analysis 1) Counts</th>
<th>Sample (analysis 1) Time (ks)</th>
<th>Sample (analysis 2) Counts</th>
<th>Sample (analysis 2) Time (ks)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.173 MeV (channel 1)</td>
<td>(x_0 = 0) (s_0 = 0)</td>
<td>(y_{01} = 0) (t_0 = 0)</td>
<td>(y_{01} = 0) (t_0 = 0)</td>
<td>(y_{01} = 0) (t_0 = 0)</td>
<td>(y_{01} = 0) (t_0 = 0)</td>
<td>(y_{01} = 0) (t_0 = 0)</td>
</tr>
<tr>
<td></td>
<td>(x_{11} = 3) (s_1 = 2.0)</td>
<td>(y_{11} = 3) (t_1 = 2.0)</td>
<td>(y_{11} = 7) (t_1 = 2.0)</td>
<td>(y_{11} = 7) (t_1 = 2.0)</td>
<td>(y_{11} = 7) (t_1 = 2.0)</td>
<td>(y_{11} = 7) (t_1 = 2.0)</td>
</tr>
<tr>
<td>1.333 MeV (channel 2)</td>
<td>(x_{02} = 0) (s_0 = 0)</td>
<td>(y_{02} = 0) (t_0 = 0)</td>
<td>(y_{02} = 0) (t_0 = 0)</td>
<td>(y_{02} = 0) (t_0 = 0)</td>
<td>(y_{02} = 0) (t_0 = 0)</td>
<td>(y_{02} = 0) (t_0 = 0)</td>
</tr>
<tr>
<td></td>
<td>(x_{12} = 6) (s_1 = 2.0)</td>
<td>(y_{12} = 14) (t_1 = 2.0)</td>
<td>(y_{12} = 10) (t_1 = 2.0)</td>
<td>(y_{12} = 10) (t_1 = 2.0)</td>
<td>(y_{12} = 10) (t_1 = 2.0)</td>
<td>(y_{12} = 10) (t_1 = 2.0)</td>
</tr>
<tr>
<td>Summed</td>
<td>(x_0 = 0) (s_0 = 0)</td>
<td>(y_0 = 0) (t_0 = 0)</td>
<td>(y_0 = 0) (t_0 = 0)</td>
<td>(y_0 = 0) (t_0 = 0)</td>
<td>(y_0 = 0) (t_0 = 0)</td>
<td>(y_0 = 0) (t_0 = 0)</td>
</tr>
<tr>
<td></td>
<td>(x_1 = 9) (s_1 = 2.0)</td>
<td>(y_1 = 17) (t_1 = 2.0)</td>
<td>(y_1 = 17) (t_1 = 2.0)</td>
<td>(y_1 = 17) (t_1 = 2.0)</td>
<td>(y_1 = 17) (t_1 = 2.0)</td>
<td>(y_1 = 17) (t_1 = 2.0)</td>
</tr>
</tbody>
</table>

The data from Table 1 is first analyzed using a conventional method. The decision level according to NUREG CR-4007 (Currie 1984) and N13.30 (Health Physics Society 1996) is given by eqn (55) and the data from Table 1, expressed in terms of net counts, is presented in Table 2.

Table 2. NUREG CR-2007 and N13.30 detection values for two analyses of \(^{60}\)Co

<table>
<thead>
<tr>
<th>Peak</th>
<th>Background Counts</th>
<th>Decision level (L_C)</th>
<th>Sample net counts</th>
<th>detected</th>
<th>Sample net counts</th>
<th>detected</th>
</tr>
</thead>
<tbody>
<tr>
<td>channel 1</td>
<td>3</td>
<td>4</td>
<td>0</td>
<td>no</td>
<td>4</td>
<td>yes</td>
</tr>
<tr>
<td>channel 2</td>
<td>6</td>
<td>6</td>
<td>8</td>
<td>yes</td>
<td>4</td>
<td>no</td>
</tr>
<tr>
<td>summed</td>
<td>9</td>
<td>7</td>
<td>8</td>
<td>yes</td>
<td>8</td>
<td>yes</td>
</tr>
</tbody>
</table>

As shown in Table 2, when each channel is considered separately, the data from analysis 1, channel 1 does not exceed the decision level, while the data from channel 2 does. The data from analysis 2 exceeds the decision level for channel 1, while it is does not for channel 2. The summed counts exceed the decision level for both analyses.

Next, the data from Table 1 is processed using the multi-channel methods proposed here. The first analysis of the sample gives zero net counts in channel 1 and eight net counts in channel 2. This result is unlikely since only the peak at 1.333 MeV is present. This is reflected in the point and interval estimates presented in Table 3. The probability of net activity due to \(^{60}\)Co is about 81% for the Multinomial and Multi-Normal methods and the mean is about 2 counts per kilosecond (CPKS). The probability interval for the true value of the net count rate spans the range between –2.3 to 6.6 counts per kilosecond. Fig 3 shows the net count rate probability density distribution obtained with the Multinomial and Multi-Normal for analysis 1.
Table 3. Probability of net activity and point and interval estimates (CKPS) of the net count rate probability distribution for two analyses of the sample

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Analysis 1</th>
<th>Analysis 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Probability of net activity Multinomial</td>
<td>81.4%</td>
<td>95.5%</td>
</tr>
<tr>
<td>Probability of net activity Multi-Normal</td>
<td>80.5%</td>
<td>94.7%</td>
</tr>
<tr>
<td>Probability of net activity Poisson</td>
<td></td>
<td>94.6%</td>
</tr>
<tr>
<td>Probability of net activity Normal</td>
<td></td>
<td>94.2%</td>
</tr>
<tr>
<td>Mean – standard deviation Multinomial</td>
<td>1.96 ± 2.25</td>
<td>3.85 ± 2.35</td>
</tr>
<tr>
<td>Mean – standard deviation Multi-Normal</td>
<td>1.85 ± 2.15</td>
<td>4.00 ± 2.48</td>
</tr>
<tr>
<td>Mean – standard deviation Poisson</td>
<td></td>
<td>4.00 ± 2.55</td>
</tr>
<tr>
<td>Mean – standard deviation Normal</td>
<td></td>
<td>4.00 ± 2.55</td>
</tr>
<tr>
<td>95% probability interval Multinomial</td>
<td>–2.28 : +6.59</td>
<td>–0.71 : +8.58</td>
</tr>
<tr>
<td>95% probability interval Multi-Normal</td>
<td>–2.36 : +6.06</td>
<td>–0.86 : +8.86</td>
</tr>
<tr>
<td>95% probability interval Poisson</td>
<td>–0.97 : +9.09</td>
<td></td>
</tr>
<tr>
<td>95% probability interval Normal</td>
<td>–1.00 : +9.00</td>
<td></td>
</tr>
<tr>
<td>Median Multinomial</td>
<td>1.81</td>
<td>3.77</td>
</tr>
<tr>
<td>Median Multi-Normal</td>
<td>1.85</td>
<td>4.00</td>
</tr>
<tr>
<td>Median Poisson</td>
<td></td>
<td>3.95</td>
</tr>
<tr>
<td>Median Normal</td>
<td></td>
<td>4.00</td>
</tr>
</tbody>
</table>

Fig. 3. Multinomial, Multi-Normal and Normal approximations for Analysis 1 (background \( x_1 = 3, x_2 = 6 \), sample \( y_1 = 3 \) and \( y_2 = 14 \), \( p_1 = 0.5 \) and \( p_2 = 0.5 \))
The second analysis yields four net counts in channel 1 and four in channel 2 (Table 1). This is more in line with the expectation that both peaks would be detected when $^{60}$Co is present in the sample. In Table 3, the probability of net activity due to $^{60}$Co is about 95% for the Multinomial and Multi-Normal methods and the mean is about 4 counts per kilosecond. The probability interval covers from –0.7 to 8.6 counts per kilosecond. Fig. 4 shows the net count rate probability density distributions for analysis 2.

Summing the counts in the two channels, as in the single channel method (Little 1982), gives the same result for both analyses: a probability of net activity of about 95% and a mean of about 4 counts per kilosecond (Table 3). The probability interval ranges from –1 to 9 counts per kilosecond. The curve for the Normal solution is shown in both Figs. 3 and 4. This result for the sum is expected since both analyses give eight net counts above background.

**Experimental measurements**

The second example involves experimental measurements carried out by the Environmental Laboratory of Hydro-Québec, located near the Gentilly-2 nuclear generating station. In the spring of 2011, two samples of maple syrup, collected from different local farms, were flagged by the Canberra peak analysis software for potentially containing $^{131}$I. The station was operating normally at the time, and there are no other likely sources of $^{131}$I contamination in the vicinity.
The first sample of maple syrup was collected at a local farm on 4 April 2011, twenty four days after the Fukushima accident. The sample was placed in a 1 L marinelli on 6 April 2011 and was counted with a high purity germanium detector for 60 kilosecond.

The data for this measurement (analysis 8492) are summarized in Table 4. A scale invariant prior is assumed and the data is analyzed using the Bayesian methods presented here. The Multinomial and Poisson equations are not used because the number of counts is too large. The results are presented in Table 5, and the probability density curves are shown in Fig. 5. The curve “Normal 364 keV channel” corresponds to the analysis of the 364.5 keV peak, using the single channel method looking at one peak individually. The curve “Normal sum of channels” corresponds to the single channel analysis of the counts summed over the four peaks. The curve “Multi-Normal” analyzed the four peaks simultaneously, using eqns (43) and (44). In the Multi-Normal case, the probability that the sample contains $^{131}$I is significant (99.3%) and matches the result of considering the 364.5 keV peak alone (99.6%). When the four peaks are summed, the background increases and the probability that net activity is present is less significant (43.1%).

**Table 4.** Counting data for two analyses of a sample counted for 60 ks

<table>
<thead>
<tr>
<th>Peak (keV)</th>
<th>$p_i$</th>
<th>Analysis 8492</th>
<th>Analysis 8531</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Det 3 – 06 April 2011</td>
<td>Det 1 – 11 April 2011</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Background</td>
<td>Sample</td>
</tr>
<tr>
<td>284.30</td>
<td>0.068</td>
<td>1166</td>
<td>1083</td>
</tr>
<tr>
<td>364.49</td>
<td>0.860</td>
<td>823</td>
<td>936</td>
</tr>
<tr>
<td>636.97</td>
<td>0.059</td>
<td>420</td>
<td>399</td>
</tr>
<tr>
<td>722.89</td>
<td>0.013</td>
<td>370</td>
<td>348</td>
</tr>
<tr>
<td>Summed</td>
<td>1.000</td>
<td>2779</td>
<td>2766</td>
</tr>
</tbody>
</table>

**Table 5.** Probability of $^{131}$I net activity and point and interval estimates (Bq kg$^{-1}$) of the probability distribution for the net activity for two analyses of the sample

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Analysis 8492</th>
<th>Analysis 8531</th>
</tr>
</thead>
<tbody>
<tr>
<td>Probability of net activity Multi-Normal</td>
<td>99.3%</td>
<td>100%</td>
</tr>
<tr>
<td>Probability of net activity Normal (sum of channels)</td>
<td>43.1%</td>
<td>100%</td>
</tr>
<tr>
<td>Probability of net activity Normal (364 keV channel)</td>
<td>99.6%</td>
<td>100%</td>
</tr>
<tr>
<td>Mean – standard deviation Multi-Normal</td>
<td>0.070 ± 0.028</td>
<td>0.040 ± 0.025</td>
</tr>
<tr>
<td>Mean – standard deviation Normal (sum of channels)</td>
<td>–0.008 ± 0.044</td>
<td>0.035 ± 0.028</td>
</tr>
<tr>
<td>Mean – standard deviation Normal (364 keV channel)</td>
<td>0.077 ± 0.029</td>
<td>0.042 ± 0.026</td>
</tr>
<tr>
<td>95% probability interval Multi-Normal</td>
<td>0.014 : 0.125</td>
<td>0.0 : 0.086</td>
</tr>
</tbody>
</table>
The same sample was counted again, on 11 April 2011 (analysis 8531) and the counting data is presented in Table 4. The statistical analysis of the second measurement can take two forms, depending on the information available to the analyst.

If the analyst knows that the first measurement had confirmed the presence of net activity due to $^{131}\text{I}$, it would be inappropriate to assume that $\Delta$ can take negative values. The probability that the sample contains net activity is exactly one and the mean and variance of the net count rate distribution are given by eqns (45) and (46). The results of the second analysis are shown in Table 5 and Fig. 6. The analysis of the second measurement shows that the $^{131}\text{I}$ activity disappears very quickly with a half-life of 8 days and does not constitute a health risk.
If, on the other hand, the sample had not been previously analyzed, it would be appropriate to assume that \( \Delta \) can take negative and positive values, as in eqns (43) and (44). The quantities obtained under this assumption are shown in Table 5 and Fig. 7. This second analysis gives a lower probability (80.5\%) that the sample contains net activity.

Fig. 6. Maple Syrup, Analysis 8531 on 11 April 2011 for known net activity
Fig. 7. Maple Syrup, Analysis 8531 on 11 April 2011 for unknown net activity

A second sample of maple syrup that is not analyzed here shows the peaks of $^{131}\text{I}$ more clearly and is not as interesting from a statistical analysis point of view.

**DISCUSSION**

As Fig 1 shows, when the count time for the background and the sample are identical, the Multinomial distribution is almost perfectly symmetric and is well approximated by the Multi-Normal distribution. For most applications, it is recommended to use the Multi-Normal approximation since it is easier to calculate.

Taking into consideration the spectral information improves the results obtained for $^{60}\text{Co}$ analyses 1 and 2. As Table 3 shows, when the spectrum does not match the expected profile, the probability that net activity is present is reduced. When the spectrum profile matches the expected probabilities, the probability of net activity (indicating the presence of a radioactive substance) matches the values obtained with the single channel analysis. It can be concluded that the single channel analysis implicitly assumes that the measured net count rates in each channel are distributed according to the expected probabilities.

In Fig. 3, the mode of the Multinomial distribution lies below the mode of the Multi-Normal distribution. Nevertheless, as Table 3 shows, the mean of the Multinomial distribution is slightly higher than the mean of the Multi-Normal distribution because the Multinomial distribution has an asymmetric shape and has an unusually large tail for
large values of the net count rate. For example, at a net count rate of 8 counts per kiloseconds, the probability density of the Multi-Normal is 0.003 and the probability density of the Multinomial is 0.008. This difference is enough to raise the mean of the Multinomial distribution.

Finally, Table 5 shows that the $^{131}$I analysis of an environmental sample using the proposed multi-channel method gives results that perform as well as methods that consider each peak separately, and yet has a lower false positive rate. It performs better than methods that sum the counts in all the peaks.

As Fig 7 shows, the proposed Bayesian methods can incorporate prior knowledge regarding the presence of a radioactive substance. The point and interval estimates for the residual activity present in the sample analyzed on 11 April 2011 are constrained to positive values since $^{131}$I had been reliably detected in the same sample on 06 April 2011.

CONCLUSION

An extension of the Bayesian derivation by Little (1982) to multi-channel analysis gives a net count rate probability distribution that takes into account the expected spectral shape in each channel of the spectrum. This can reduce the probability of false positives. A Poisson-based derivation is presented for a case where the number counts is small. When the number of counts is large, the Poisson-based solution is well approximated by a Normal equation and this approximation was also derived.

The net count rate probability distribution can be used to calculate the probability that an unknown sample contains a radioactive substance. It also allows the calculation of point and interval estimates of the net count rate: mean, standard deviation, median, and percentiles. Finally, it can take into account information about the presence of net activity in a sample by limiting the probability distribution to positive values of the net count rate.

The Multinomial and Multi-Normal methods are not expected to perform better than single channel methods when the total number of net counts is less than the number of channels. The multi-channel methods work best when there are enough counts to fill-in the spectrum.

The multi-channel method cannot presently deal with peak interference problems. The required extension of the multi-channel method would calculate the net count rate probability distribution for a radionuclide when several radioactive substances contribute to the counts in a given peak. The model for this problem is the object of ongoing research.
Acknowledgement

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REFERENCES


