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Empirical Confirmation of the Critical Level for Zero and Near Zero Background Measurements

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ABSTRACT

The alpha spectroscopy system of the Lawrence Livermore National Laboratory, Hazards Control Department evaluates electroplated samples, typically urine and feces, for alpha emitting radionuclides. Most of the samples processed by the alpha spectroscopy system are evaluated for Plutonium-239 (Pu-239), an important radionuclide used in research. This paper evaluates the Pu-239 background response of the Lawrence Livermore National Laboratory Hazards Control Department's alpha spectroscopy system. Background measurements of the alpha spectroscopy system have been studied to determine an appropriate method for establishing the *a postori* critical level for detection of plutonium alpha activity. Several methods of establishing the 95% confidence interval for over 4,900 background measurements were evaluated. Two methods appear to provide reasonable results so as to assure an appropriate 95% confidence interval. This report provides the results of this evaluation and the comparison of the various methods tested to establish an empirical evaluation of the critical level using a commercially available analysis program**.**

Introduction

Low-level radiation counting systems, such as those used in alpha spectrometry, present unique problems in determining whether a measurement is truly positive. Typically, when a measurement results in less than 20 counts, the count data are considered to follow Poisson distribution characteristics. When a measurement exhibits more than 20, the Poisson characteristics of the count data can be represented by Gaussian distribution characteristics (Cember85). As defined by ANSI 13.30, the determination of the *a postori* decision level (a.k.a. critical level) is based on the assumption that the count data behaves according to a Gaussian statistical distribution.

The 95% critical level (i.e., the level at which 95% of the background counts will register as 'background') for a known Gaussian distribution is defined as:

$$L_c = \overline{X} + 1.645 \times \sqrt{\sigma_B^2}$$

where: L_c is the 95% critical level;
$\overline{X}$ is the mean background;
σ_B^2 is the variance associated with the background distribution and;
1.645 is the one-sided 95^{th} percentile z-value for a normal distribution.

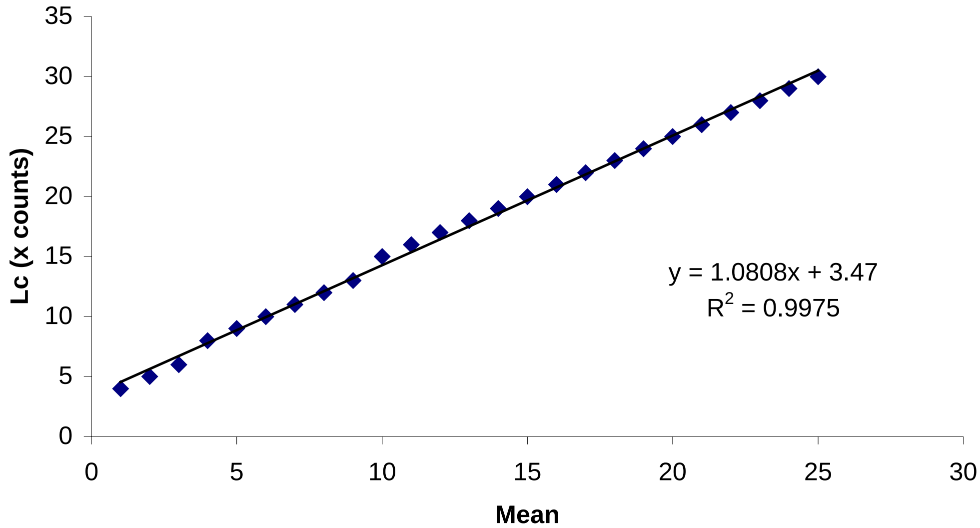
Nuclear count data are usually represented Poisson characteristics. In the Poisson distribution, the value of σ_B^2 is considered to be equal to $\overline{X}$. This concept is used in nuclear count statistics when evaluating the critical level using a single background count. A Poisson distribution 95% critical level can be evaluated by directly using the Poisson distribution as defined by:

$$P(x, \lambda) = \frac{e^{-\lambda} \lambda^x}{x!}$$

where: x is the measured count; ;
λ is the true mean.

If the Poisson distribution is used to evaluate the 95 percentile critical level for a range of means (λ), an empirical function can be derived to describe the behavior of the critical level as a function of the 'mean' background count. This evaluation and empirical function relationship is illustrated in Fig. 1.

Fig 1. The 95% confidence level Lc as a function of the Poisson mean.



Difficulties can arise with the both the Gaussian and Poisson methods for establishing the 95% critical level when the variance and background counts respectively equal zero. The Poisson function is undefined when the mean count is zero. Likewise, when there is only a single background measurement used to establish the critical level, the estimate of σ_B^2 will be zero when the background count equals zero.

In 1968, Currie proposed a method for low-count rate and zero count data. This method involved a factor of 2.71, which assures a detection limit (L_d) of approximately 3 counts when the count approaches zero. However, in Currie's derivation, the addition of this factor was only applied to the minimum detectable activity (a.k.a decision level), not the critical level. In an attempt to compensate for low count rate and zero count data an alternative method for computing the critical level is applied by the commercial software currently used at Lawrence Livermore National Laboratory. This method adds 2.71 to the Currie computed critical level, such that the critical level is calculated as:

$$L_c = Counts_{Bkg} + 1.645 \times \sqrt{2 \times Counts_{Bkg}^2} + 2.71$$

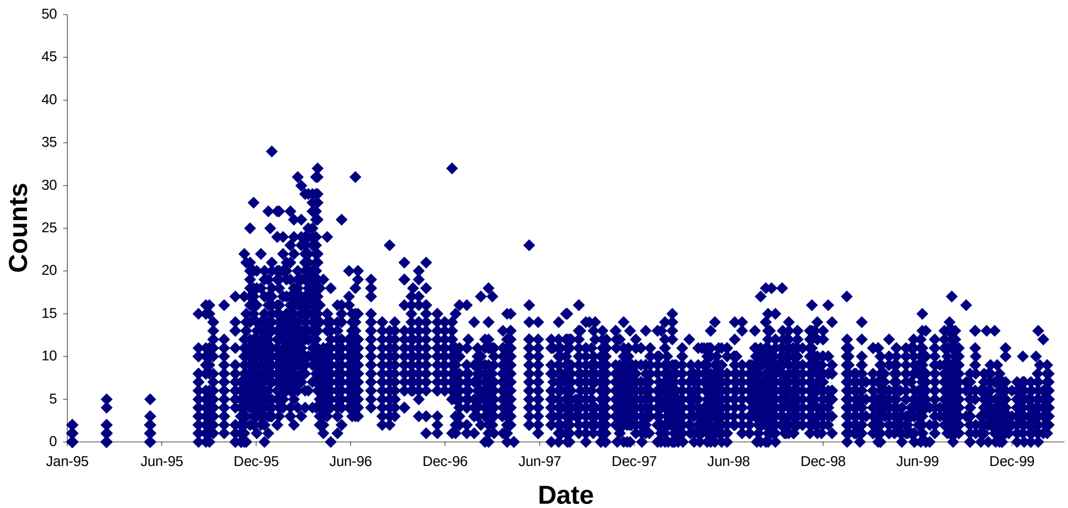
The purpose of this study is to evaluate the critical level method applied at LLNL and to determine if alternate methods of establishing the critical level should be used.

Methods

The alpha spectrometry system at LLNL contains 34 Canberra Alpha Spectrometers (Model 7401) controlled by Canberra Alpha Management System (AMS) software. Each detector is calibrated at least once a month. Background measurements are performed periodically. From January 1995 to February 2000 there have been a total of 4,967 background measurements, divided among the 34 spectrometers with each measurement lasting 2.5 days. From January 1995 to August 27, 1999, background measurements were performed using empty chambers. After August 27, 1999, background measurements were performed with a blank electro-deposition planchet loaded in the normal sample counting position. After January 10, 1997 to the present, the alpha chambers, when not under vacuum, are purged with dry nitrogen to maintain a low background. Prior to the institution of dry nitrogen purge, several mechanical and maintenance problems were known to exist with the alpha spectroscopy system. Detector changes in recent years have been minimal, however prior to January 1997, detectors often failed and required changing or repair.

Background count data for the Pu-239 region of interest was obtained from the computer records maintained by the AMS. The entire data set was sorted according to date and the data for each measurement was segregated. A graphical evaluation of the time-sorted data was then performed to observe anomalies in the data. A large degree of variability was observed prior to January 10, 1997 due to the mechanical and maintenance problems in the 1995 to 1997 time period (Fig. 2).

Fig. 2. Alpha background counts for the LLNL alpha spectroscopy system.



Only data post January 10, 1997 were evaluated in this study. Further analysis of the background count data was performed by segregating the data according to the time period of events associated with modification of the alpha spectrometry system as previously described. The number of background measurements performed since January 10, 1997 to February 2000 was 3,287 for the 36 spectrometers. A subset analysis was performed for background measurements made from January 1997 to August 27, 1999 and August 27, 1999 to February 2000.The mean, standard deviation, and median of the background count data for this period was determined for each subset of measurements. A probability distribution was also generated with overlays of the expected Poisson and Gaussian using the evaluated mean and standard deviation.

Reagent and urine blank count data for the Pu-239 region was also evaluated. Between the period of January 10, 1997 and February 2000 there were 191 reagent blanks and 191 urine blanks counted on the alpha spectroscopy system. Each data set was graphically evaluated to determine any anomalies. The means, standard deviations, and medians were determined for the reagent and urine blank count data. Probability density distributions were also generated with overlays of the expected Poisson and Gaussian using the evaluated means and standard deviations.

Between January 1997 and March 2000, there were 2,068 routine samples counted. These samples were process-plated sample data from workers at LLNL. Sample counts and the most recent background count data were evaluated to determine the number of samples that would be above the 95% critical level. Net sample counts were obtained by subtracting the most recently acquired 2.5-day background count from the gross sample count.

Since some of the samples could be from personnel with low-level or previous exposure to plutonium, the sample data was prescreened for elevated count results. The sample data was first sorted lowest to highest number of gross counts. Based on an analysis of the background, reagent blank, and urine blanks it was determined that background and blank data rarely exceeded 19 counts since January 1997. Therefore only sample counts with a gross count of less than 19 counts, a total of 1785 samples, were used for this study. Both background and sample count data with a gross count that was less than 19 counts were used to evaluate four critical level computational methods. The methods used to evaluate the critical levels are mathematically described below.

Evaluation of the Currie "well known blank" method:

This method is applicable when there is a significant amount of count time and counts in the background. The number of counts in the background must be sufficient enough to have a variance that can approximate a Gaussian distribution. The formulation for this critical level calculation is:

$$L_c = 1.645 \times \sqrt{B}$$

where B is the background counts for a well-known measurement. For this evaluation, the well-known background was taken as the average background value for the different subsets of data.

Evaluation of the Currie "paired observations" method:

When using paired observations, such as a background count and a sample count, where one count is added to or subtracted from another count (e.g., a background count is subtracted from a gross count), then the following critical level formulation from Currie was used:

$$L_c = 1.64 \times \sqrt{2 \times B}$$

Evaluation of the Poisson distribution 95th percentile critical level method:

The relationship between the Poisson mean and the critical level as previously demonstrated was used to establish the Poison critical level at various count rates (see Fig. 1). The calculated critical level was rounded to preserve the discrete characteristics of the Poisson distribution. The relationship used for the background count (mean) to the critical level was:

$$L_c = 1.0808 \times B + 3.47$$

Note: in contrast to the pure Poisson function a zero background count using this functional relationship generates a critical level of 3 counts.

Evaluation of the Canberra Alpha Measurement System method:

When there are equivalent background and sample measurement times, the default critical level calculation as applied by the Canberra Alpha Measurement System used at LLNL reduces to a basic 'count' form of:

$$L_c = 2.325 \times \sqrt{B} + 2.71$$

For each of the applicable critical level methods within each subset, the percent of sample counts above or below the critical level were then tallied. These tallies were used to determine if the typical background count and critical level method appear to represent a true 95th percentile level.

An alternate method of determining the critical level is to evaluate the Poisson or Gaussian distribution of a given 'historical' population set of background measurements. The Poisson or Gaussian cumulative probability distribution was computed using fifty of the most recent background measurements prior to the measurement of interest. The measurement of interest was then evaluated to determine if it the counts exceeded the one-sided confidence level (95% for this study) for the distribution of the 50 background measurements. Since there was a significant change in the method of background collection in August 1999 (i.e., use of a steel plate while accumulating background), this method was evaluated for two separate populations of background measurements, the composite of background measurements since January 10, 1997 as well as for background measurements collected after August 27, 1999. The percent of the time that the (background) count of interest exceeded the 95th percentile based on the previous 50 background measurements was then tallied. Ideally, this sliding average method should demonstrate a 5% false positive indication. This technique was also applied to the background data set for each spectrometer.

Finally, reagent and urine blank counts were evaluated against the 95th percentile using a Normal and Poisson distribution based on the most recent 50 background measurement values obtained for the spectrometer used to measure the reagent blank or urine blank sample. Only samples since August 27, 1999 were used for this evaluation.

Results

The results of the means, medians, and standard deviations for the background and low background sample count (i.e., less than 19 counts) data are provided in Table 1. The mean counts for populations and counters ranged from 2.56 to 8.08 over the 2.5-day count period. The mean background count for all counters between January 1997 and August 27, 1999 tended to have higher background counts than during the period after August 27, 1999. Reagent blank, urine blank, and sample count data tend to be consistent with background data since August 1999.

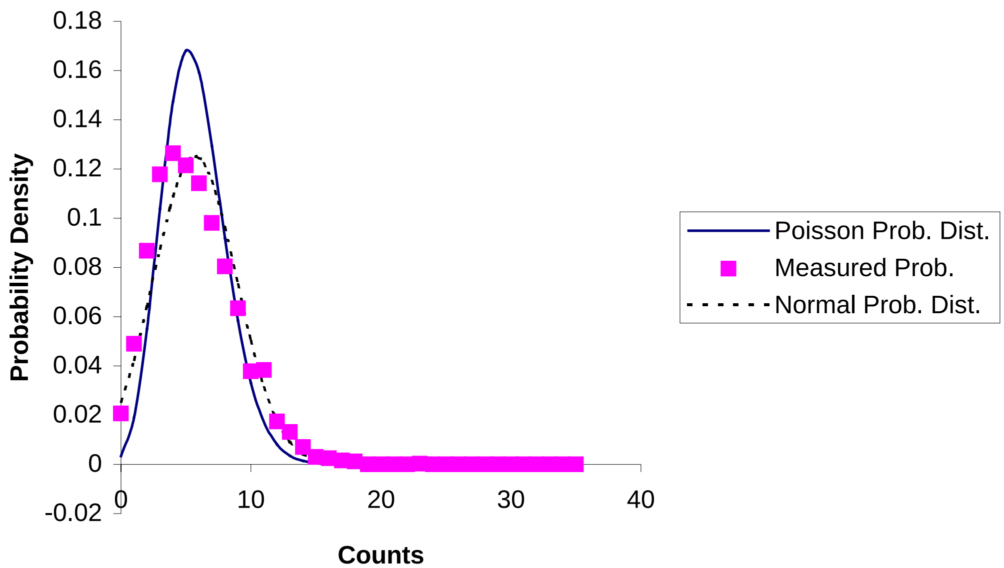
Table 1. Alpha counter background measurement data.

Data Set	N	Mean	Median	Std. Dev. (1s)
All Counters, All Measurements (Jan 1997 – Feb 2000)	3285	5.69	5	3.19
All Counters, All Measurements (Jan 1997 - Aug 27, 1999)	2824	5.93	6	3.22
All Counters, All Measurements (Aug 27, 1999 – Feb 2000)	461	4.15	4	2.44
Reagent Blanks (all counters)	180	3.93	4	2.84
Urine Blanks (all counters)	190	4.38	4	3.56
Sample Counts by Spectrometer[1]	1785	4.38	4	3.23
1	88	6.05	6	2.43
2	86	6.84	7.5	3.00
3	90	5.02	4	2.86
4	89	6.02	6	3.01
5	88	4.18	4	4.18
6	88	6.55	6	2.89
7	86	5.97	5	3.16
8	86	6.23	6	2.94
9	97	4.66	5	2.38
10	100	7.7	8	3.62
11	99	5.19	5	2.80
12	101	6.11	6	3.07
13	100	3.13	3	2.29
14	98	4.21	4	2.43
15	99	6.38	6	2.71
16	101	5.54	5	2.96
17	88	6.82	7	2.96
18	89	7.43	7	3.15
19	90	8.08	8	3.29
20	90	6.14	6	3.24
21	89	4.01	3	2.47
22	89	5.31	5	2.75
23	90	6.47	6	2.88
24	88	5.76	6	2.36
25	88	4.30	4	2.75
26	89	6.80	7	3.35
27	89	7.20	7	2.96
28	92	6.12	6	3.14
29	92	5.71	6	2.52
30	92	6.26	6	3.03
31	92	7.49	7	3.48
32	91	7.02	6	3.41
33	83	3.86	3	2.79
34	91	5.12	4	3.69
35	88	2.52	2	1.63
36	89	2.56	2	2.08

[1] Gross counts from personnel sample count data where the gross count was less then 19 counts.

The probability density distribution of the data from January 1997 to February 2000 demonstrates a skewed distribution with tailing as the background counts increase (Fig. 3). Neither the Poisson nor the Gaussian distributions tend to adequately represent the data distribution.

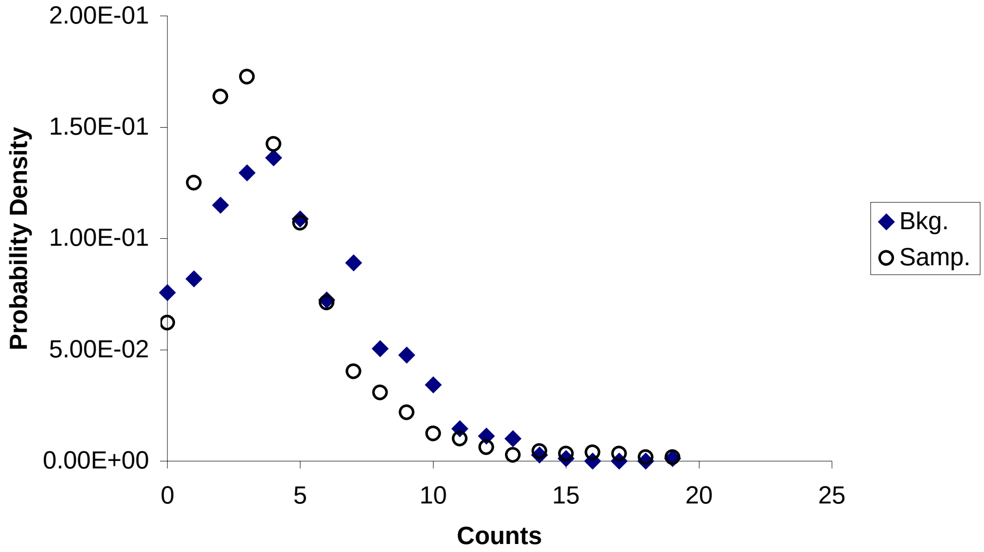
Fig. 3. Pu Background Probability Density Distribution for all measurements between Jan 1997 and Feb 2000.



Note: The Poisson distribution is a discrete distribution but is provided as a continuous function for visual comparison with the data.

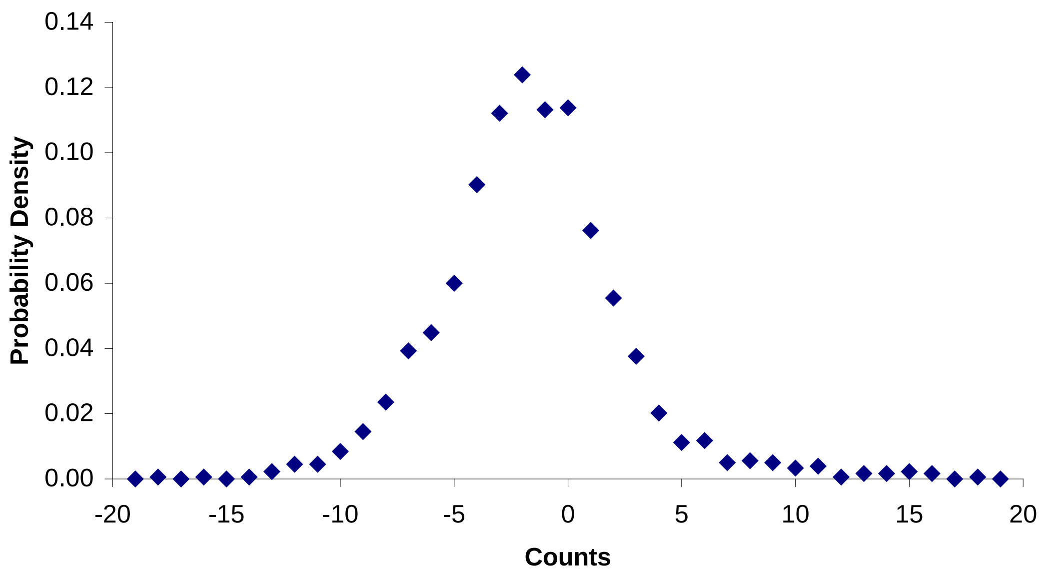
Fig. 4 illustrates the observed difference in the background and sample distributions for the sample population.

Fig. 4. Personnel gross sample count and associated background count distributions.



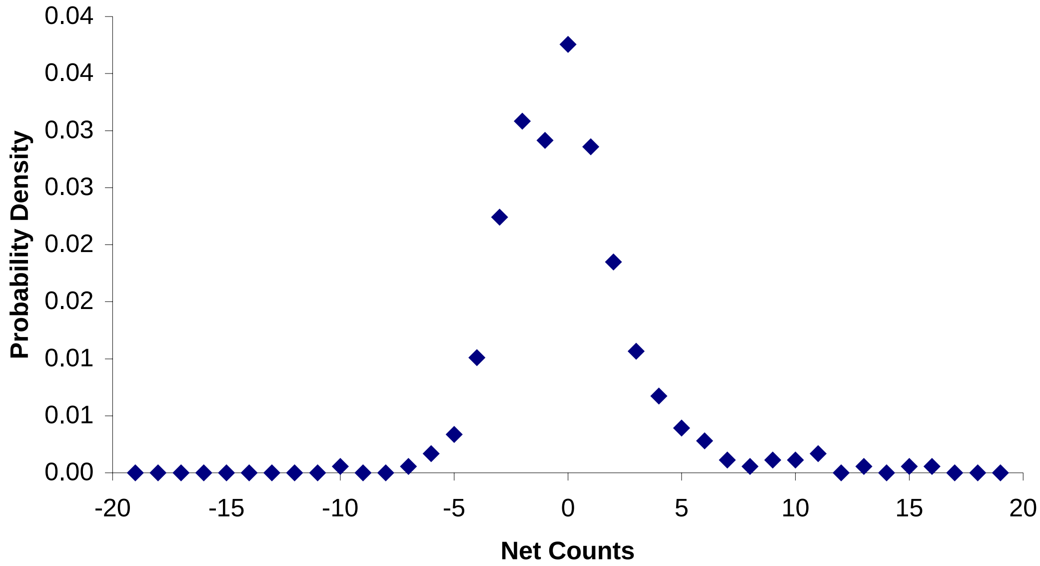
When the net sample population was evaluated, the distribution tended to have a slight negative bias as shown in Fig. 5.

Fig. 5. Net sample count distribution January 1997 – February 2000.



However, when the net sample distribution is evaluated for samples analyzed after Aug 27, 1999, when blank planchets were instituted for background counts, the negative bias in the net sample population is no longer present as seen in Fig 6.

Fig. 6. Net sample probability distribution for samples analyzed after August 1999.



When the background and sample count data were ranked from lowest to highest value, the count at which 95% of the count data were observed to be below that value are provided in Table 2.

Table 2. Observed 95th percentiles for background and sample data sets.

Population	N	One sided 95th Percentile (counts in 216000 s)
Background – all counters (Jan 1997 – Feb 2000)	3285	12
Background – all counters (Aug 27, 1999 – Feb 2000)	461	9
Reagent Blanks (all counters)	180	10
Urine Blanks (all counters)	190	11
Sample Background Counts	382	11
Sample Gross Counts	382	10
Sample Net Counts	382	5
Sample Net Counts (Aug 27, 1999 – Feb 2000)	382	5

The calculated 95th percentile (one-sided) for the three methods of calculating the critical level using the average counts form the data sets (background, reagent blank, urine blank, and sample background populations) are summarized in Table 3.

Table 3. Computed critical levels (net counts in 216000 s) for the evaluated populations sets using population averages.

Population	Currie CL	Poisson CL	Canberra CL
Background – all counters (Jan 1997 – Feb 2000)	6	10	8
Background – all counters (Aug 27, 1999 – Feb 2000)	5	8	7
Reagent Blanks	5	8	7
Urine Blanks	5	8	8
Personnel Sample Background Counts	5	9	8

Note: The Currie 'well known' critical level is not evaluated since the average background count was far below 20 counts.

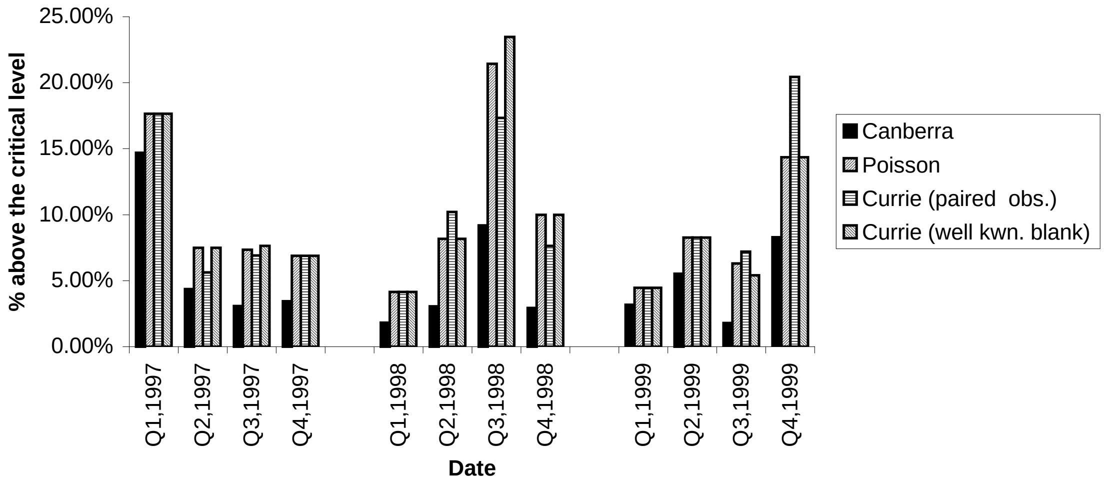
When the critical level is computed for each sample using the most recent chamber background count, the number of sample results above the critical level for each of the three methods is provided in Table 4.

Table 4. The percent of false positives observed for 1,782 samples using the most recent chamber background and various methods of calculating the 95th percentile critical level.

	% above the L_c by method			
Time Period	**Canberra**	**Poisson**	**Currie (well known blank)**	**Currie (paired samples)**
All Data	4.20%	8.18%	8.24%	8.80%
1997	4.78%	7.85%	7.85%	6.48%
1998	3.19%	8.24%	8.51%	7.45%
1999	5.05%	8.75%	8.59%	10.77%
2000	4.79%	6.16%	6.16%	12.33%

When the sample data is further evaluated by quarter, the number of sample results above the critical level for each of the three methods becomes more variable as illustrated in Fig. 7.

Fig. 7. Percent of sample measurements that are above the critical level by quarter and year.



When the current background count was evaluated against the critical level that was established using Normal and Poisson distributions of the 50 previous background measurements, the percent of measurements that exceeded the 95th percentile critical level can be found in Table 5.

Table 5. Percent of background counts that exceed the 95th percentile level for the Normal and Poisson distributions of the previous 50 background measurements.

Population	Normal	Poisson
All Counters combined (Jan 1997 – Feb 2000)	6.8%	12.5%
All Counters combined (Jan 1997 - Aug 27, 1999)	6.9%	13%
All Counters combined (Aug 27, 1999 – Feb 2000)	6.1%	8.9%
Spectrometer Number		
1	2.2%	4.5%
2	2.3%	3.4%
3	2.2%	3.3%
4	4.4%	5.6%
5	2.2%	2.2%
6	2.2%	4.5%
7	3.4%	3.4%
8	4.3%	4.3%
9	2.0%	2.0%
10	2.0%	6.9%
11	5.9%	9.9%
12	3.9%	5.8%
13	5.9%	6.9%
14	7.0%	8.0%
15	3.0%	4.0%
16	8.7%	13.6%
17	0.0%	3.3%
18	3.9%	3.9%
19	4.3%	8.5%
20	1.1%	4.3%
21	5.4%	9.7%
22	0.0%	2.2%
23	2.1%	5.3%
24	1.1%	1.1%
25	1.1%	2.2%
26	1.1%	3.3%
27	4.3%	4.3%
28	4.2%	7.4%
29	4.2%	6.3%
30	7.4%	9.5%
31	1.1%	3.2%
32	6.4%	8.5%
33	18.2%	18.2%
34	9.8%	12.2%
35	2.6%	2.6%
36	2.6%	5.1%
Average of all Detectors	3.93%	5.79%

Table 6 shows the results of reagent and urine blank counts that were evaluated against the 95th percentile critical level using a Normal and Poisson distribution of the 50 background measurements most recently collected prior to the sample measurement.

Table 6. Percent of background measurements that exceed the 95th percentile level for the Normal and Poisson distributions of the previous 50 background measurements.

Sample Type	Normal	Poisson
Reagent Blank	4.17%	4.17%
Urine Blank	5.88%	5.88%

Discussion

The alpha spectroscopy system at LLNL uses a consistent method to analyze alpha spectra. The analysis method includes the determination of the critical level and the detection level for each count. This uniformity in analysis assures consistency and provides for a practical implementation of procedures. The purpose of this study was to evaluate empirical data to verify or establish an appropriate method for the determination of the 95th percentile critical level when there is zero or near zero events.

Background and sample data from the LLNL alpha spectroscopy system were analyzed to determine the appropriate 95th percentile critical level for the plutonium region of interest. Over 3200 background measurements and 1782 sample measurements collected over approximately a three-year period were used in the evaluation. Each measurement consisted of 2.5 days worth of counting in 36 alpha chambers. Reagent blanks as well as urine blanks were also evaluated.

The percent of measurements above or below the critical levels using the four different calculation methods were tallied. Since all four methods are theoretically based on the 95th percent confidence interval for background, critical levels should demonstrate a 5% rate of positive (falsely positive) measurements. More than three years of data consisting of 1782 sample measurements demonstrates that the Poisson and Currie methods are continuously above the 5% false positive rate. On a quarterly basis, the Currie ‘well known’ background method for the calculation of the critical level demonstrated a false positive rate greater than 5% in six out of twelve quarters. The Poisson and the Currie ‘paired observation’ based critical levels demonstrated a false positive rate greater than 5% in 10 of the twelve quarters. In contrast, the Canberra critical level calculation method exceeded the 5% false positive rate 4 out of 12 quarters.

A bias in the background prior to August 27, 1999 has been observed, such that background was over-subtracted by approximately 2 counts during this time period (Fig. 5). The observation of effect with such a low background count rate requires the collection of a large number of background measurements. Since each background measurement takes 2.5 days, the accumulation of an adequate background measurement population can take years, especially if the facility needs to process personnel samples at the same time. At this point in time, there is a reasonable indication that the bias observed prior to August 27, 1999 has been eliminated by the use of a steel planchet.

When the background count is zero, the critical level for the Currie methods is undefined (Currie68). In the entire population of 3,285 background measurements, 2% of the background measurements were zero. Likewise, among the sample population 2% of the backgrounds used to evaluate the critical level for sample measurements were also zero. Since August 27, 1999 when ‘empty chamber’ backgrounds were no longer used, approximately 5% of the backgrounds used for sample data analysis were equal to zero.

Samples collected in the last quarter of 1999 and in the early portions of the year 2000 (Table 4 and Fig. 7), indicate that the Canberra method for the calculation of the critical level is an appropriate method when using a single background count for establishing the critical level. Measurements from the early portions of the year 2000 indicate a 4.79% false positive rate for the Canberra method in contrast to all of the other methods which indicate false positive rates of 12%. In the last quarter of 1999, after the new background method was applied, the Canberra method demonstrated an 8.3% false positive rate while the other methods ranged from 14% to 20 % false positive rates.

According to theory, the best method for evaluating the critical level would be to use a moving average distribution background method. The 95th percentile for a Normal and Poisson distribution was determined using 50 background measurements that immediately preceded the measurement being evaluated. For the LLNL system, 50 background measurements for a particular detector can represent approximately one year’s worth of background data collection. The data in this study indicate that the Normal distribution trends to be an appropriate method for establishing the critical level (see Table 6) when considering the entire data set. However, if the historical background for an individual spectrometer is used to evaluate whether a particular count exceeds the 95th percentile then, on the average, a Poisson distribution of the 50 previous background counts generated a more appropriate critical level. Urine and Reagent blank data (since August 1999) that was evaluated against a critical level that was generated by the preceding 50-counter/chamber backgrounds demonstrated a 4.17% and 5.88% ‘false positive’ rate for the Normal and Poisson distributions, respectively.

Conclusions

The use of a database of previous background measurements provides a technically sound method for establishing the critical level of a stable counting system. Evaluations using reagent and urine blank data demonstrate that a critical level based on the 50-preceeding background measurements will result in the appropriate false positive level. Either a Gaussian or Poison distribution can be applied to the database for the determination of the critical level. However, use of a database of previous background counts and the application of a Poisson or Normal distribution to this database in order to determine the critical level is currently not available within the alpha spectroscopy software used at LLNL. Likewise, a significant amount of time must pass before there are enough background measurements to establish an adequate database to be used for critical level determination. Finally, changes to a system can alter the response of the system and make previous databases invalid thereby requiring the establishment of a new database of measurements.

The existing LLNL alpha spectroscopy system requires a technically sound method for computing the critical level using a single background measurement. Therefore, in the absence of a database of background measurements, the empirical data tend to support the use of the following Canberra formulation for establishing the critical level:

$$L_c = 2.325 \times \sqrt{B} + 2.71$$

This method for the determining the critical level is a hybrid of the Currie "paired observation" critical level and the Currie detection limit (L_D), whereby a factor of 2.71 is added to the detection limit to compensate for zero background situations.

The 3,285-background count probability distribution observed in this study tended to demonstrate non-normal characteristics. Neither the Poisson nor the Normal distribution appeared to completely describe the observed distribution (Fig 3) of background. The evaluation of the four critical level methods tends to support this conclusion. For example, the number of false positives tends to be excessive when using either of the Currie methods, which are based on the Gaussian distribution.

The effect of a background bias does not currently seem to have a large influence on the conclusions regarding how the critical level should be computed. However only 461 background measurements have been collected since this bias was eliminated and conclusions must be guarded.

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