



**MIRION**  
TECHNOLOGIES

# Detection Limit Concepts According to Currie

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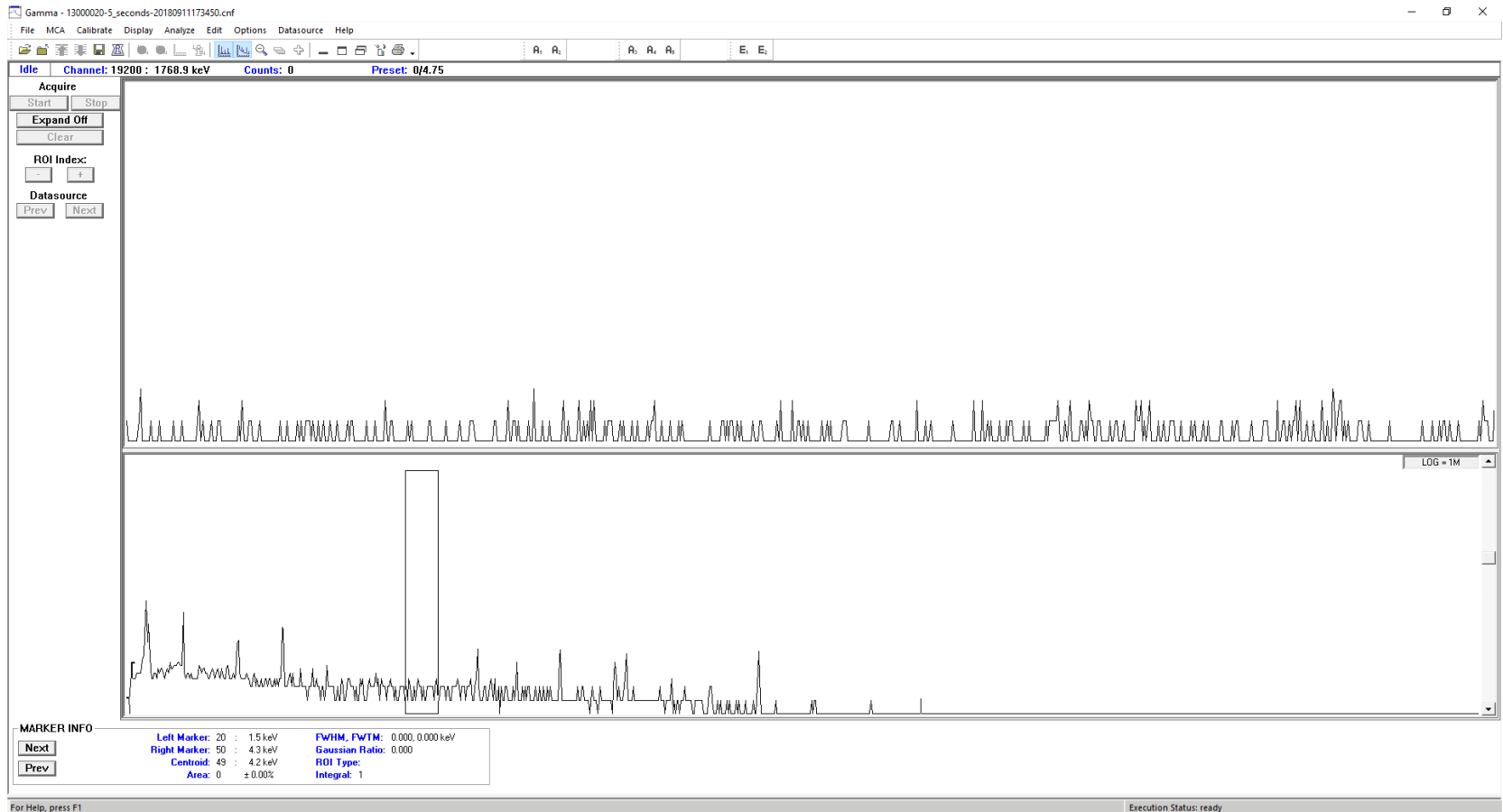


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Lloyd A. Currie introduces three concepts: critical level, detection limit, and determination limit in his landmark article “Limits for Qualitative Detection and Quantitative Determination: Application to Radiochemistry” from 1968. The article is one of the most often cited and discussed publications in radiochemistry.

This webinar will explain these three concepts and how Currie intended for them to be used. They are the foundations that later developments in detection limits are building on which makes them still relevant over 50 years after they were introduced. Examples will be given from gamma spectroscopy and it will be shown how these are implemented in Genie™ 2000 Spectroscopy Software.

# Is there something there?



# Introduction

- Radioactive decay is a random process
- Our measurement equipment is not perfect
- We can determine if a signal (nuclide) is present
- We cannot determine that a nuclide is not present
  - There is always a possibility that there are some very small amount present
- If a nuclide is not determined to present we should be able to give an upper limit on the amount present
  - The nuclide is not seen, therefore the activity needs to be below a value
- We should also be able to determine the performance of our measurement equipment
  - With this measurement setup I can expect that I will find nuclide activities that are above a value

# The Currie method

- In 1968 Lloyd A. Currie published a set of definitions of limiting levels for determining if a signal is present and equations for calculating these limits
- Before this publication there had been many different definitions and ways of calculating limits
  - A standardization was needed
- It became the standard for reporting in a lot of industries
  - It's still used today
-

# Currie's three limiting levels

- $L_C$  - *Critical level* - the net signal level (instrument response) above which an *observed* signal may be reliably recognized as “detected”
- $L_D$  - *Detection limit* - The “true” net signal level which may be *a priori* expected to lead to a detection
- $L_Q$  - *Quantification limit* - The level at which the measurement precision will be satisfactory for quantitative determination
- Currie also defines an *upper limit* for a signal that has not been detected.
- *A priori* vs *A posteriori*
  - *A priori* – relating to or derived by reasoning from self-evident propositions
  - *A posteriori* - relating to or derived by reasoning from observed facts

# Definitions

- Blank
  - $\mu_b$  - limiting (“true”) mean
  - $B$  – Observed value
  - $\sigma_B$  - Standard deviation
- Gross
  - $\mu_{S+B}$  - limiting mean
  - $S + B$  – Observed value
  - $\sigma_{S+B}$  - Standard deviation
- Net Signal
  - $\mu_S$  - limiting mean
  - $S = (S + B) - B$
  - $\sigma_S = \sqrt{\sigma_{S+B}^2 + \sigma_B^2}$
- The blank is defined as the signal resulting from a sample which is identical, in principle, to the sample of interest, except that the substance sought is absent. The blank thus includes the effects of interfering species

# Two fundamental aspects of detection

- 1 – Given an observed signal (net) signal,  $S$ , one must decide whether a *real* signal has been detected. i.e if  $\mu_S > 0$
- 2 – Given a measurement process, one must estimate the minimum *true* signal,  $\mu_S$ , which may be expected to yield a sufficiently large observed signal  $S$  that it will be detected.
- The first aspect relates to the *a posteriori* binary (qualitative) decision based upon the observation  $S$  and a definite criterion for detection.
  - Following the decision one should establish either an upper limit or a confidence interval
- The second aspect relates to the *a priori estimate* of the detection capabilities of a given measurement process



# The signal

- We are usually interested in activities or activity concentration of nuclides in our sample at some point in time
- However the detector measures the energy deposited by radiation in the active volume of the detector
- We determine the number of counts in peaks (this is our signal) and apply a correction factor to get the nuclide activity (efficiency calibration, decay correction, etc)
- First we have to look at how we determine the signal and the uncertainty in the signal

# Counts above continuum

- The peak area ( $S$ ) is calculated as

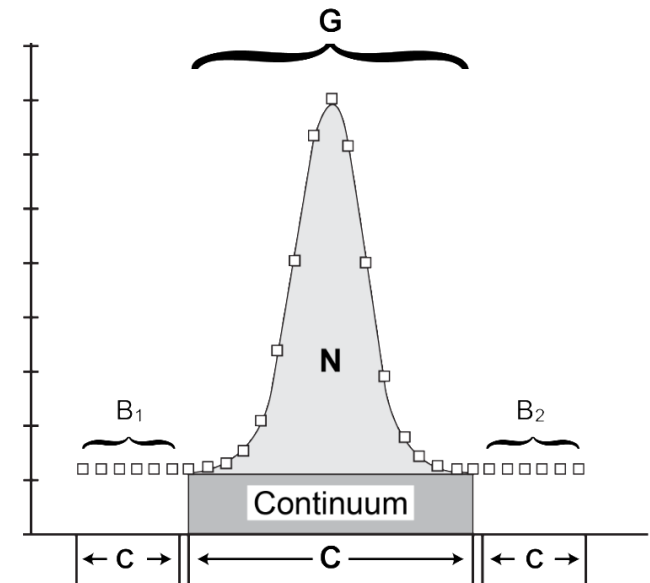
$$S = G - C$$

- Where  $G$  is the total number of counts in the ROI,  $C$  is the counts in the continuum
- Applying the uncertainty propagation formula

$$\sigma_S = \sqrt{\sigma_G^2 + \sigma_C^2}$$

- The uncertainty of the total number of counts is

$$\sigma_G = \sqrt{G}, \sigma_G^2 = G$$



# Linear continuum

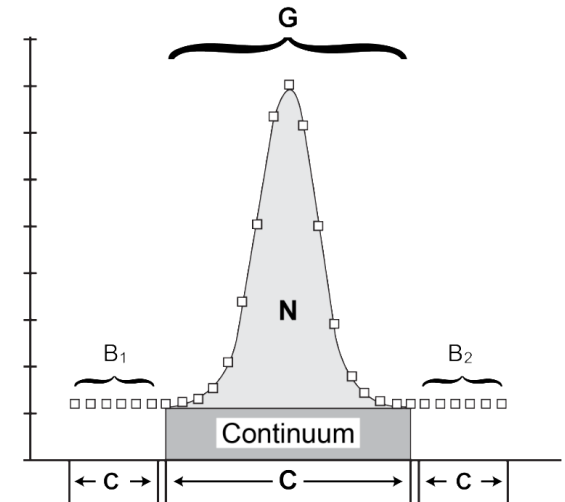
- The linear continuum assumes that the continuum can be described by a straight line from the left side of the ROI to the right side of the ROI

$$C = \left(\frac{N}{2n}\right) (B_1 + B_2)$$

- Where  $N$  is the number of channels in the ROI,  $n$  is the number of channels on the side of the ROI and  $B_1$  and  $B_2$  are the number of counts on the sides of the ROI

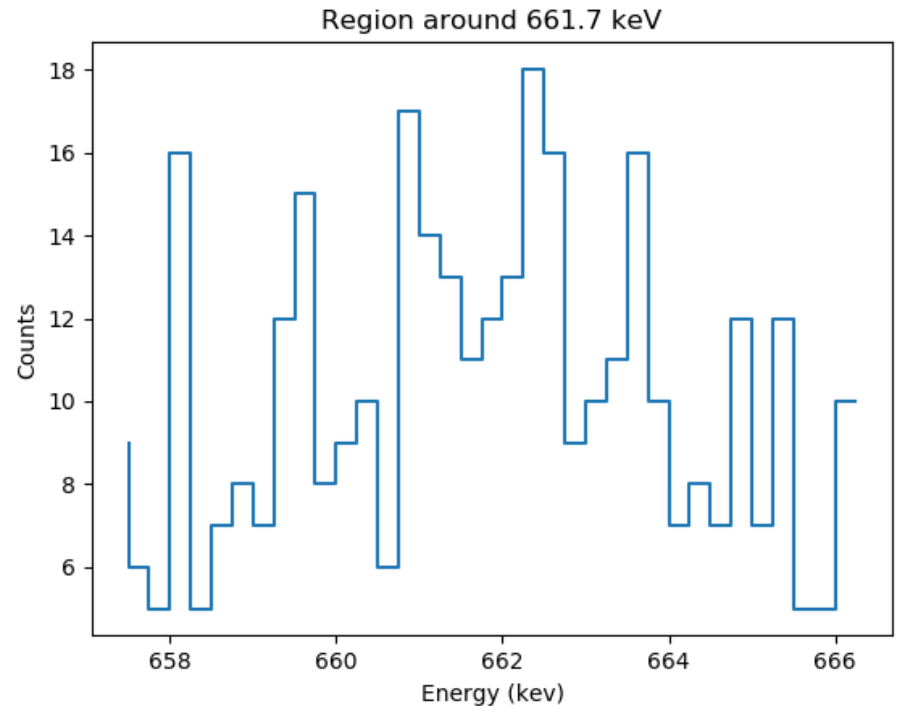
- Applying the uncertainty propagation formula
- $$\sigma_C^2 = \left(\frac{N}{2n}\right)^2 \sigma_{B_1}^2 + \left(\frac{N}{2n}\right)^2 \sigma_{B_2}^2 = \left(\frac{N}{2n}\right)^2 (B_1 + B_2)$$

$$\sigma_s = \sqrt{G + \left(\frac{N}{2n}\right)^2 (B_1 + B_2)}$$



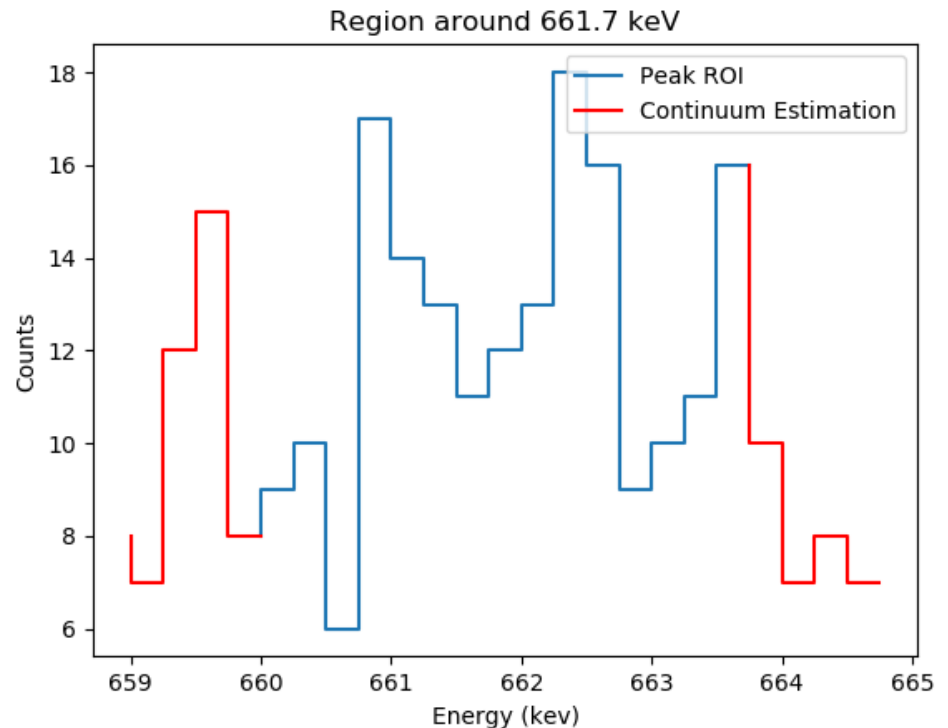
# The signal

- Assume that we want to know if Cs-137 is present in the sample
- Is there a peak present at 661.7 keV?
- We can calculate the size of the signal at 661.7 keV



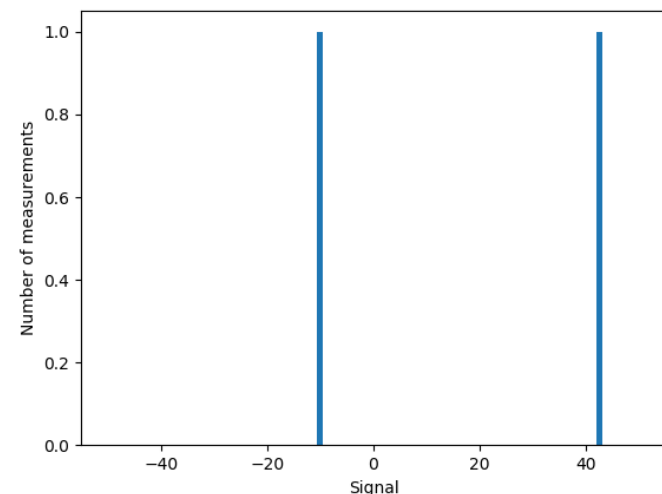
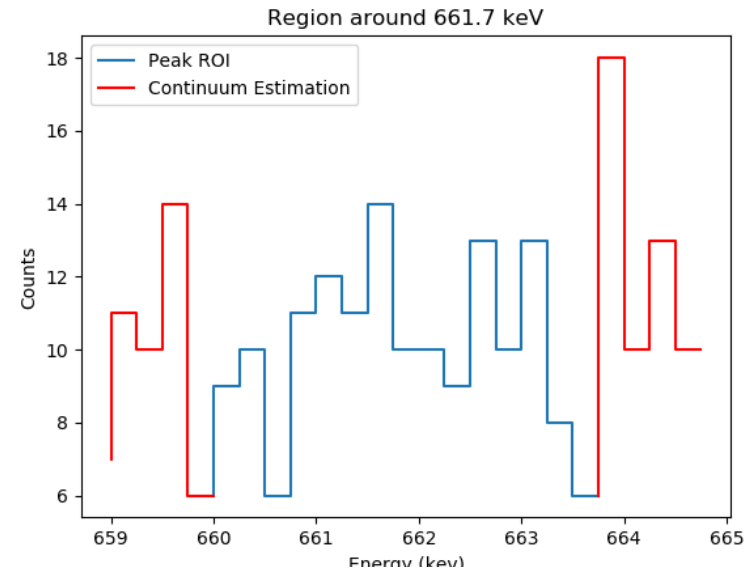
# Calculating the signal

- If we calculate the peak area using the linear continuum we get
- $S = 42$
- $\sigma_S = 22.18$  (52.8%)
- But is this a real signal or just statistical fluctuations of the continuum
- Let's repeat the measurement



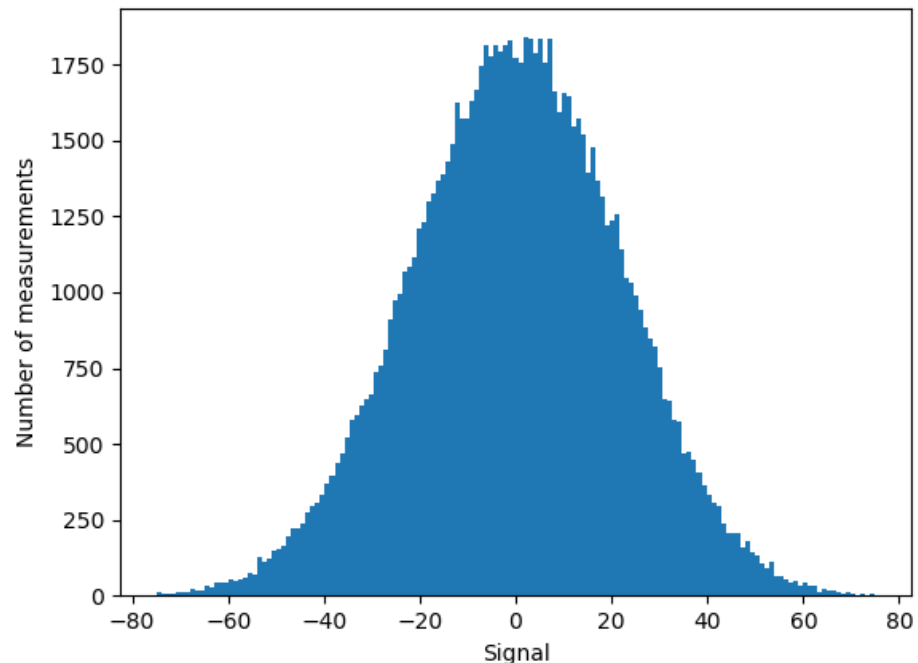
# 2<sup>nd</sup> measurement

- The second time we get a negative S
- $S = -10$
- $\sigma_S = 22.76$  (227%)
- We can make a histogram of the distributions of the signal from the measurements
- Let's do this many times



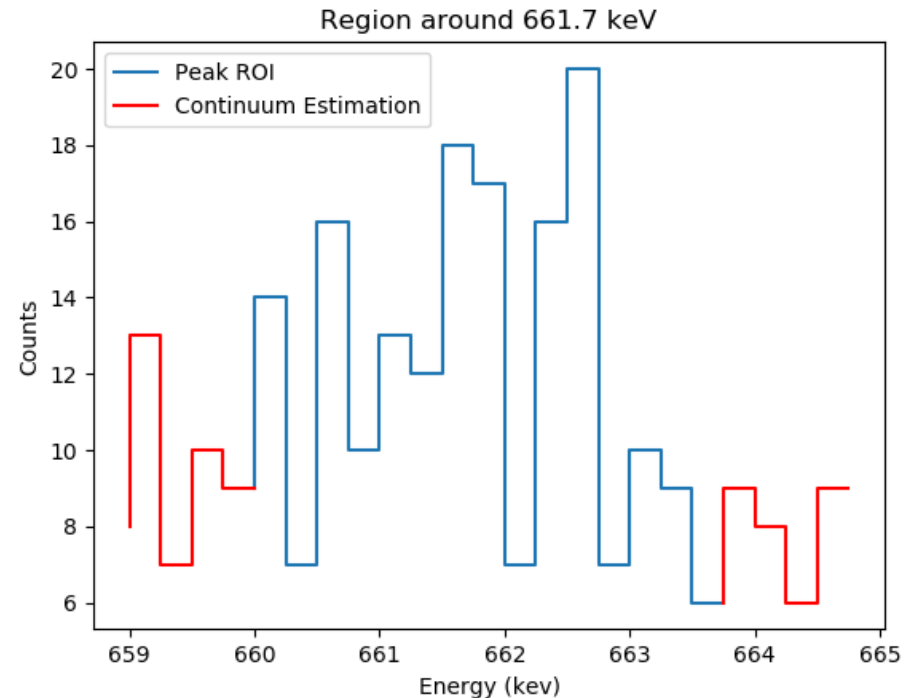
# 100000 measurements

- The calculated signal ranges from -80 to 80 counts
- The most probable value is to get a signal that is close to 0
- The probability of calculating a signal goes down the further from 0 the signal gets.
- The standard deviation is 21.9 and the 95<sup>th</sup> percentile is 36.



# Small real signal

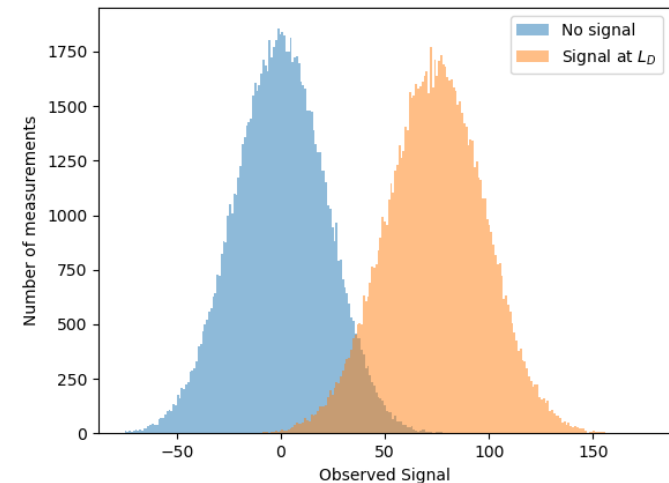
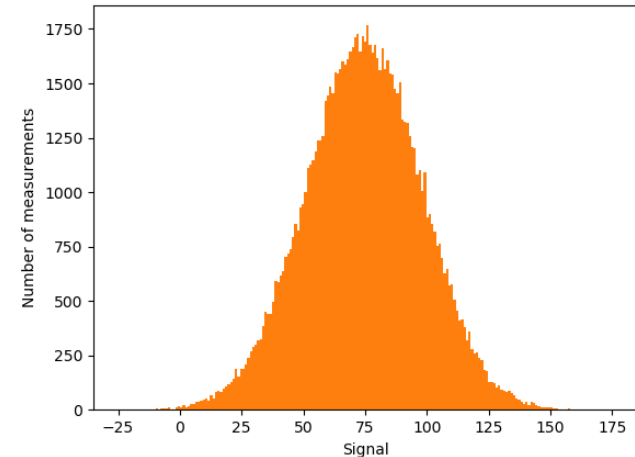
- If we add a sample containing a small amount of Cs-137 and make a measurement
- We can calculate the size of the signal
- $S = 59$
- $\sigma_s = 21.6$  (36.6%)





# 100000 measurements of the sample

- The calculated signal ranges from 0 to 150
- It's not very different from the distribution when there was no real signal
- It is shifted by 75 counts (true signal)
- The width is slightly larger
- If we measure an unknown sample and calculate the peak area to be 35 counts. Is it a real signal or just fluctuations from the continuum?



# Is the signal real?

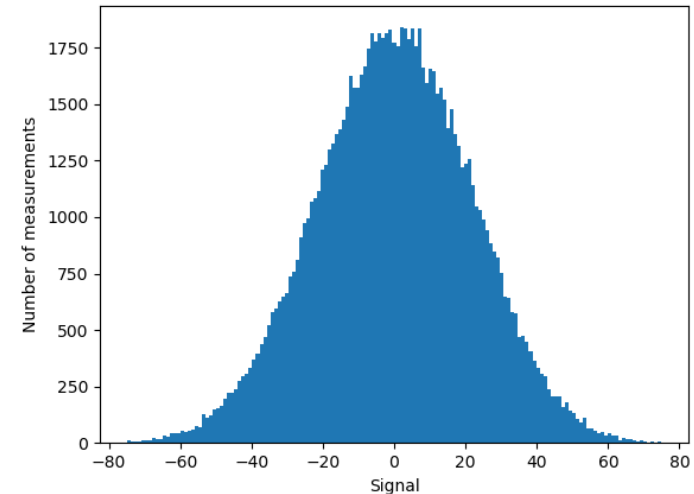
- There are two hypotheses, the signal is from the blank or the signal is real
  - We need to test which is most likely
- It is possible to make two errors
  - 1) deciding that the signal is present when it is not ( $\alpha$ : false positive)
  - 2) deciding that the signal is not present when it is ( $\beta$ : false negative)
- We need to decide how often we can tolerate making each of these errors

# Is the signal real?

- Remember the calculation of the observed signal from the 100 000 measurements.
- The critical level  $L_c$  is the size of the signal where  $1 - \alpha$  gives the correct answer. I.e. Not detected

$$L_c = k_\alpha \sigma_0$$

- The critical level only depends on the uncertainty of the observed signal when the substance is not present
- It will always be in counts



# The *a priori* detection limit

- The detection limit  $L_D$  is defined so that the probability distributions of outcomes (when the true signal is  $L_D$ ) intersects  $L_C$  such that the fraction  $1 - \beta$  will correspond to the correct decision, “detected”

$$L_D = L_C + k_\beta \sigma_D$$

- This means that the  $L_D$  is the smallest true signal possible that the measurement system will detect with  $1 - \beta$  probability while having a  $\alpha$  probability of incorrectly determine that a true signal of 0 is determined to be detected

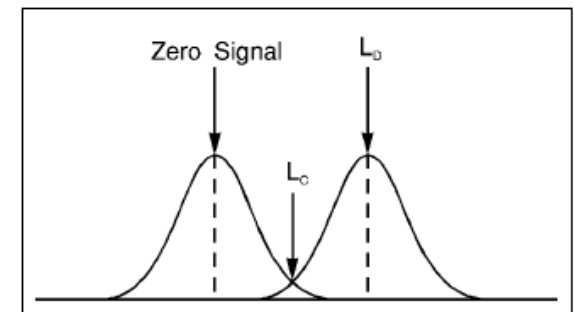
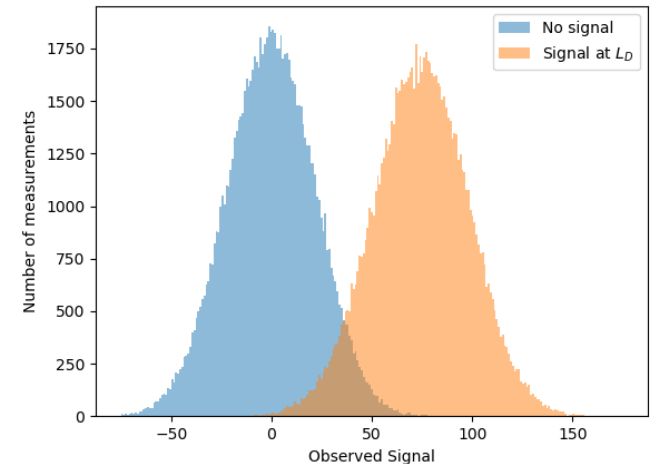


Figure 8 Illustrating the LC and LD Concepts

# Upper limit

- If the observed signal is less than the critical level the decision “not detected” should be reported.
- This doesn't necessarily mean that the signal is not present
  - It means that the observed signal was not large enough that we with  $1 - \alpha$  probability could say that the signal was not from the blank

- An upper limit should be reported

$$S + k'_\gamma \sigma_S$$

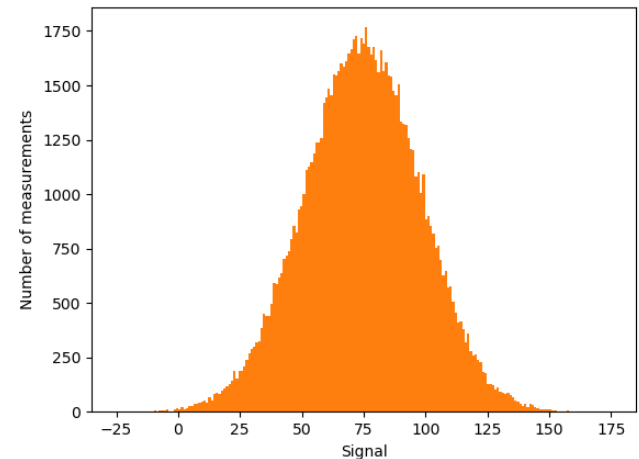
- Where  $k'_\gamma$  is the one sided confidence level

# Confidence interval

- If the observed signal is larger than the critical level the decision “detected” should be reported.
- An interval may be stated for the signal based on the confidence level  $1 - \gamma$

$$S \pm k_{\gamma} \sigma_S$$

- This means that with  $1 - \gamma$  certainty the true signal is within the confidence interval



# Quantitative Analysis

- It may not be satisfactory to have a decision that the signal is present or not with an upper limit or with a wide confidence interval.
- It may be necessary to know the size of the signal that will give a relative standard deviation that is smaller than a value  $q$
- We can therefore define the Determination Limit  $L_Q$  for which the uncertainty is smaller than  $q$

$$L_Q = k_q \sigma_Q$$

- Where  $1/k_q$  is the required relative standard deviation



# Questions?



# How can we apply this to Gamma spectrometry

# $L_C$ - Critical level

- The critical level depends on only on the uncertainty of the 0-signal ( $\sigma_0$ )

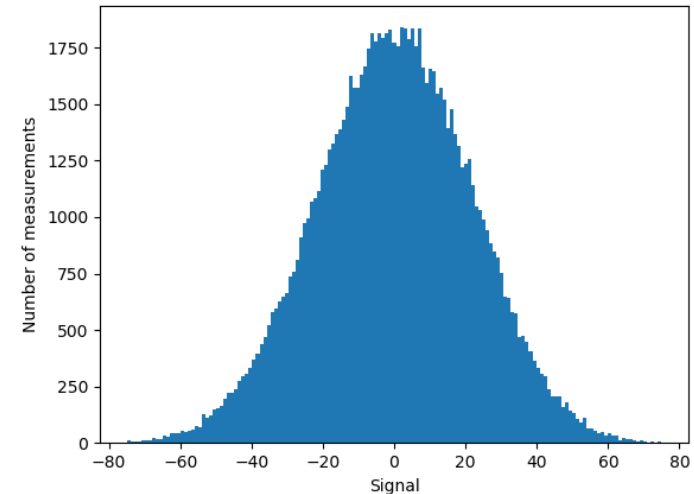
- The variance of the net signal is
$$\sigma_S^2 = \sigma_{S+B}^2 + \sigma_B^2 = (S + B) + \sigma_B^2$$

- But for the 0-signal  $S = 0$ 
$$\sigma_0^2 = B + \sigma_B^2$$

- Now we can write the expression for the critical level

$$L_C = k_\alpha \sigma_0 = k_\alpha \sqrt{B + \sigma_B^2}$$

- This depends on the size and the uncertainty of the background signal.



# $L_D$ - Detection limit

- The  $L_D$  is defined as  

$$L_D = L_C + k_\beta \sigma_D = k_\alpha \sigma_0 + k_\beta \sigma_D$$
- We need to know what  $\sigma_D$ , the variance when  $S = L_D$ , is  

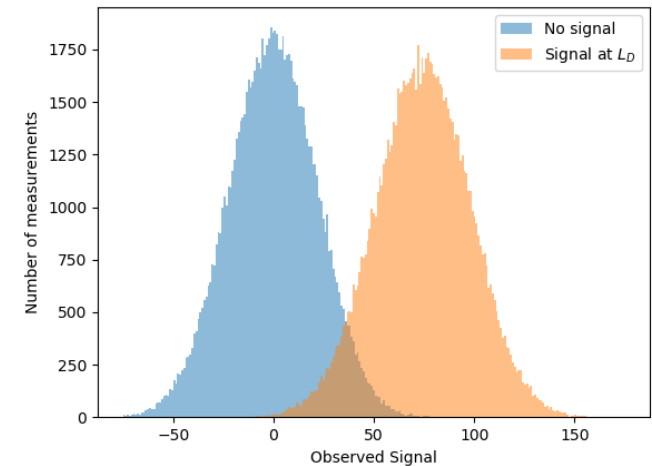
$$\sigma_D^2 = L_D + \sigma_0^2$$

- So we can write

$$L_D = L_C + k_\beta \sqrt{(L_D + \sigma_0^2)}$$

- Using that  $\sigma_0 = \frac{L_C}{k_\alpha}$  we can write a general expression for  $L_D$

$$L_D = L_C + \frac{k_\beta^2}{2} \left( 1 + \sqrt{1 + \frac{4L_C}{k_\beta^2} + \frac{4L_C^2}{k_\alpha^2 k_\beta^2}} \right)$$



# $L_D$ - Detection limit

- A common case is when  $k_\alpha = k_\beta = k$  then the formula reduces to

$$L_D = k^2 + 2L_C = k^2 + 2k\sigma_0$$

- Note the  $k^2$  term which means that there is a minimum value that the  $L_D$  can have even if  $\sigma_0$  is 0.

# $L_Q$ - Quantification limit

- We want to be able to determine the activity of a nuclide with 10% relative standard deviation ( $k_q = 10$ )

$$L_Q = k_q \sqrt{L_Q + \sigma_0^2}$$

- Which can be solved for  $L_Q$

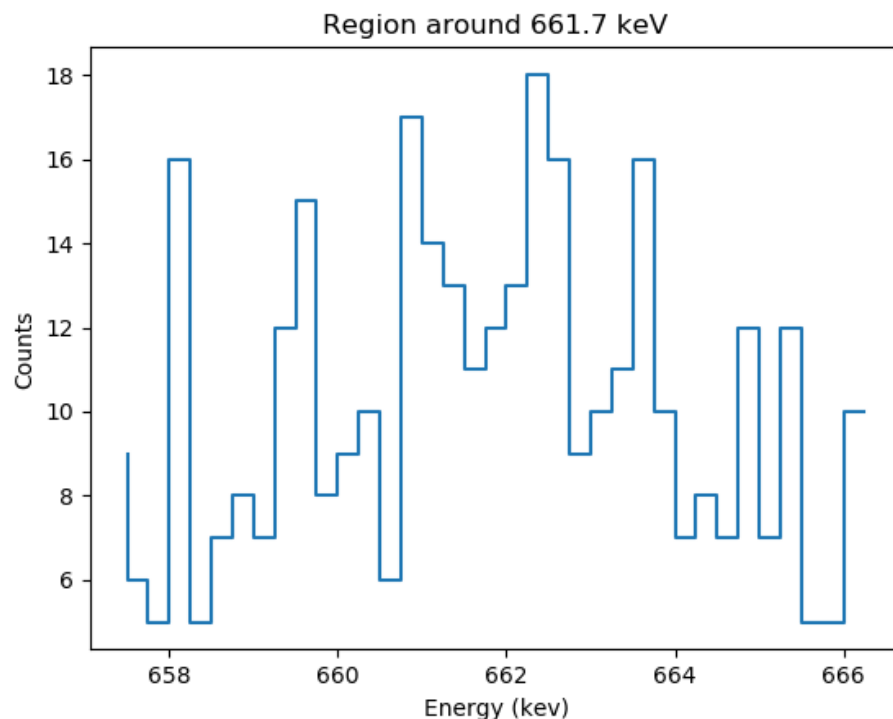
$$L_Q = \frac{k_Q^2}{2} \left( 1 + \sqrt{1 + \frac{4\sigma_0^2}{k_Q^2}} \right)$$

# Calculation of the critical level in Genie

- The calculation of the critical level in Genie depends on if the peak is found or not
  - For peaks that are not found the continuum (which we call  $C$ ) are simply the counts in the region of interest where we are looking for the signal.
  - If a peak has been found the continuum is estimated from the channels immediately to the left and right of the region of interest. (Using the continuum model chosen during peak analysis)
- The region of interest is determined during the peak area analysis for peaks that has been found but what about the region of interest for peaks that were not found

# The ROI of unidentified peaks

- How do we determine the size of the region of interest if we are looking for a signal at 661.7 keV (Cs-137)?

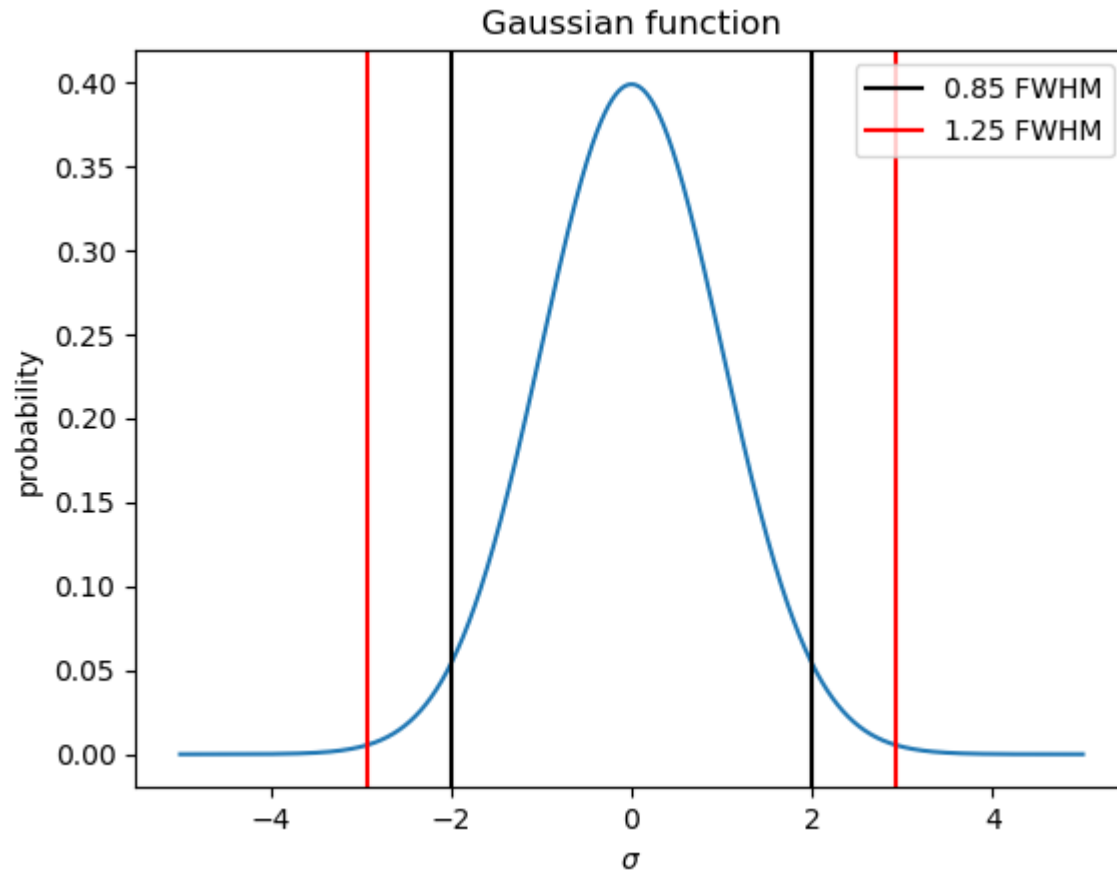


# The ROI of unidentified peaks

- From the shape calibration we know the expected FWHM at the energy of interest
- If we are assuming that we have Gaussian shaped peaks we can relate the FWHM to the width of the Gaussian  $FWHM = 2.335\sigma$
- This means that if we use an ROI that is  $\pm 1FWHM$  we will use a width of  $2.335\sigma$  or the ROI will cover about 98% of the expected counts from a peak at that location
- Common values for the ROI width are
  - $0.85 FWHM = 1.985\sigma$  which will cover about 95% of the expected counts
  - $1.25 FWHM = 2.919\sigma$  which will cover about 99.6% of the expected peak counts for a peak in that region
- A note of caution, if the region is too small there could be counts from the peak outside the region and this will lead to incorrect results



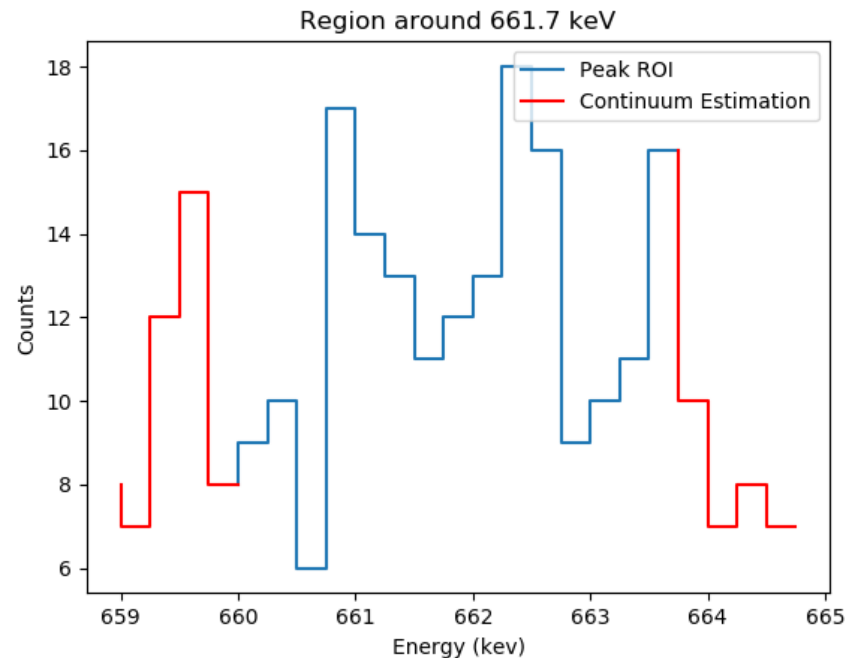
# Peak coverage



# Example

- In this case there is no peak detected
- The continuum counts are just the sum of the counts in the ROI
$$C = \sum y_i = 193$$
- If we assume a 5% false positive rate then  $k = 1.645$  and  $L_C$  becomes

$$\begin{aligned} L_C &= 1.645 \sqrt{C + \sigma_C^2} \\ &= 1.645 \sqrt{2C} = 32.3 \end{aligned}$$



# What if we have detected a peak

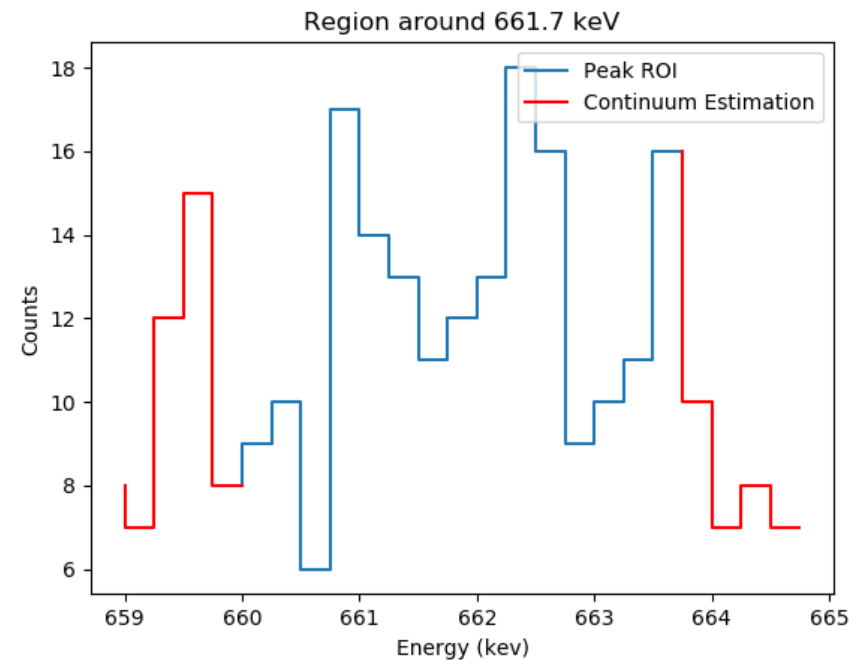
- Now we have to estimate the continuum from the region next to the ROI.

$$C = \left(\frac{N}{2n}\right) (B_1 + B_2)$$

$$\sigma_C^2 = \left(\frac{N}{2n}\right)^2 (B_1 + B_2)$$

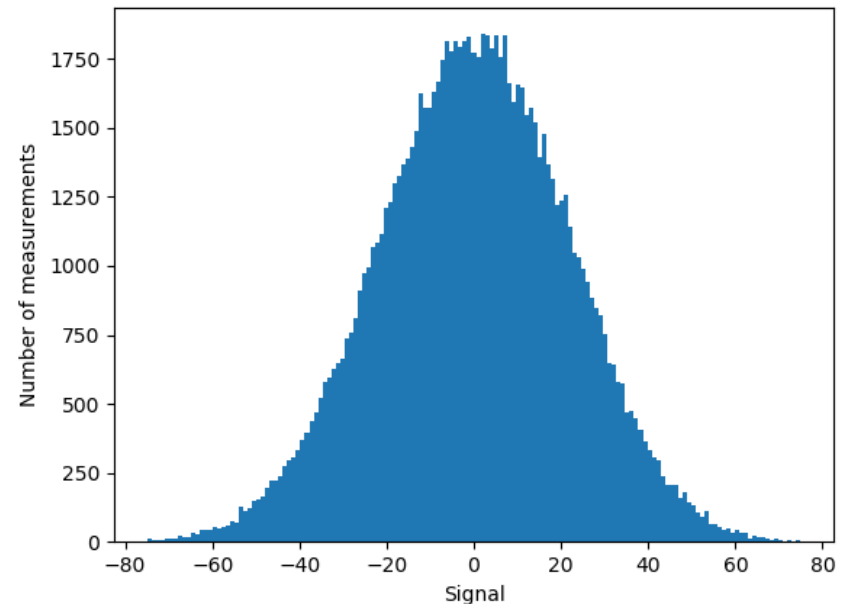
- So in this case the  $L_C$  becomes

$$\begin{aligned} L_C &= 1.645 \sqrt{C + \sigma_C^2} \\ &= 1.645 \sqrt{\left(\left(\frac{N}{2n}\right) + \left(\frac{N}{2n}\right)^2\right) (B_1 + B_2)} \\ &= 34.9 \end{aligned}$$



# Example

- Both method gives answers that are close to each other
- 32.3 and 34.9 are good estimates of the 95<sup>th</sup> percentile of the distribution

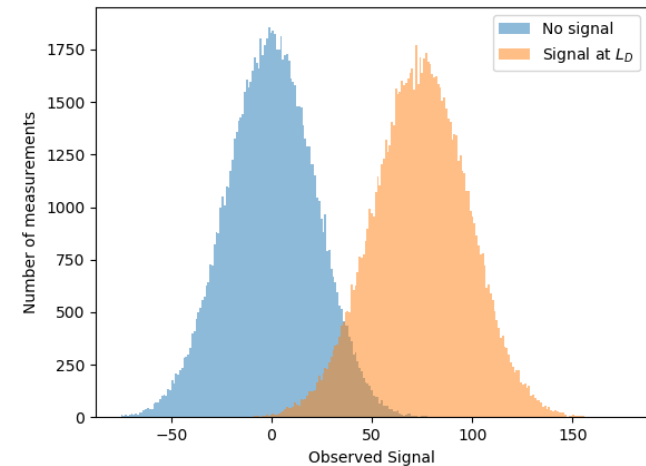
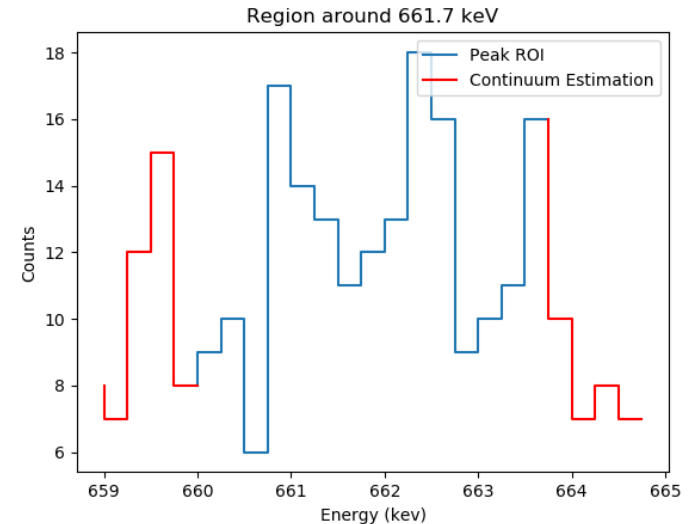


# $L_D$ - Detection limit

- If we want to calculate the  $L_D$  for the same measurement, and we can accept 5% false positive rate and 5% false negative rate we get

$$L_D = k^2 + 2L_C = k^2 + 2k\sigma_0 = \\ \approx 2.71 + 3.29\sqrt{2C} = 2.71 + 4.65\sqrt{C}$$

- $L_D$  increases with the  $\sqrt{C}$
- Estimate the continuum from the counts in the ROI  $L_D = 67.3$
- Estimate the continuum from the channels next to the ROI  $L_D = 77.8$



# Limits for physical quantities

- The decision level needs to only be known in counts
  - The signal that is compared to the decision level is measured in counts
- For the detection limit it is often desired to convert to activity
  - It is useful to know what level of activity that would with 95% confidence be detected with the measurement procedure
- This conversion is typically done with a calibration factor relating to the detector response

$$MDA = \frac{L_D}{\varepsilon T I K} = \frac{2.71 + 3.29\sqrt{2C}}{\varepsilon T I K} = \frac{2.71 + 4.65\sqrt{C}}{\varepsilon T I K}$$

- The continuum  $C$  increases linearly with time so the MDA decreases as  $\frac{1}{\sqrt{T}}$
- To get a reduction in MDA by a factor of 2 the measurement time has to increase by a factor of 4

# Summary of Currie method

- $L_C$  - the net signal level (instrument response) above which an *observed* signal may be reliably recognized as “detected”
- $L_D$  - The “true” net signal level which may be *a priori* expected to lead to a detection
- The first quantity should be used to determine if a real signal is present or not
- The second is an *a priori* detection limit and it will tell you how large a real signal needs to be so that the observed signal will be classified as detected with  $1 - \beta$  probability while a 0-signal will only result in detected with  $\alpha$  probability.
  - It should NOT be used to determine if a signal is present or not!
- If a signal is determined to be present the value of the signal plus a confidence interval should be reported
- If a signal is not determined to be present an upper limit should be reported

# How to apply the critical level test in Genie

- The critical level test is selected in the Peak area calculation step
- Check the box 95% critical level test
- This means that the only value of  $\alpha$  that Genie supports is 5%
- In practice this is the only value that people are using

Sum / Non-Linear LSQ Fit Setup

Peak Area Region  
Start channel: 1  
Stop channel: 4096

Continuum: 4 Channels  
☒ Channels ☐ FWHM

Continuum function: Step

Residual Search  
☐ Perform Search Threshold: 4.00  
Minimum separation (FWHM): 1.00

ROI Limits Determination  
Max. Num. FWHMs between peaks: 4.00  
Max. Num. FWHMs for left limit: 4.00  
Max. Num. FWHMs for right limit: 4.00  
☐ Use Fixed ROI Limits

☒ 95% Critical level test  
☐ Use fixed FWHM  
☒ Use fixed tail parameter  
☐ Fit singlets  
☐ Display ROIs  
☐ Reject zero area peaks  
☐ Use DOECAP rules

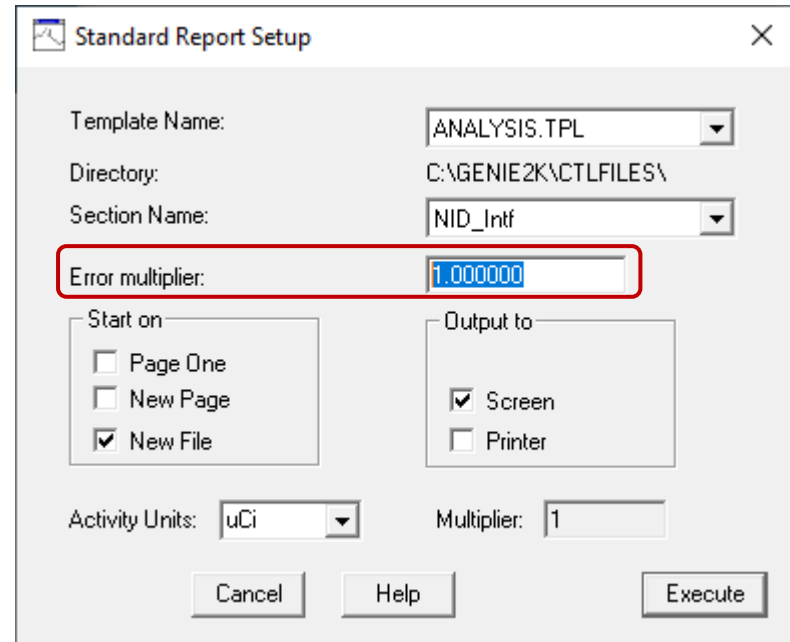
☐ Generate Report

Cancel Help Execute



# How to report a detected signal with a confidence limit

- The NID with interference correction calculates the activity or activity concentration and the  $1\sigma$  uncertainty
- A different confidence limit can be selected in the report set up
  - The error multiplier corresponds to the k value of the confidence interval
  - Remember to use the two-sided interval



The image shows a 'Standard Report Setup' dialog box. It contains several fields and checkboxes. The 'Error multiplier' field is highlighted with a red rectangle and contains the value '1.000000'. Other fields include 'Template Name' (ANALYSIS.TPL), 'Directory' (C:\GENIE2K\CTLFILERS\), 'Section Name' (NID\_Intf), 'Activity Units' (uCi), and 'Multiplier' (1). There are also checkboxes for 'Start on' (Page One, New Page, New File) and 'Output to' (Screen, Printer).

Field	Value
Template Name	ANALYSIS.TPL
Directory	C:\GENIE2K\CTLFILERS\
Section Name	NID_Intf
Error multiplier	1.000000
Start on	<input type="checkbox"/> Page One <input type="checkbox"/> New Page <input checked="" type="checkbox"/> New File
Output to	<input checked="" type="checkbox"/> Screen <input type="checkbox"/> Printer
Activity Units	uCi
Multiplier	1

Buttons: Cancel, Help, Execute

# Example report

```
*****
*****  I N T E R F E R E N C E   C O R R E C T E D   R E P O R T   *****
*****
```

Nuclide Name	Nuclide Id Confidence	Wt mean Activity (uCi/unit)	Wt mean Activity Uncertainty
K-40	0.998	1.8197726E-01	8.1553133E-03
CO-57	0.999	2.9000187E-02	1.0330334E-03
CO-60	1.000	9.8212689E-02	1.1601595E-03
SE-75	0.340	9.7695389E-04	6.8992749E-04
? KR-85	0.999	8.5342083E+00	2.3190632E-01
? SR-85	0.992	7.9866521E-02	2.1705369E-03
Y-88	0.998	2.9403418E-01	4.8703472E-03
CD-109	0.996	3.5504082E-01	2.2888346E-02
SN-113	0.941	6.5487795E-02	1.9268007E-03
CS-134	0.709	7.8966031E-03	3.1692063E-04
CS-137	1.000	4.2519774E-02	1.2482395E-03
CE-139	0.998	1.8339178E-02	7.3207327E-04
HG-203	0.987	6.0442489E-02	2.2938002E-03
PB-212	0.399	6.2898411E-03	7.1053160E-04
BI-214	0.555	9.1663692E-03	5.0186517E-04
PB-214	0.484	5.8486760E-03	6.7147578E-04

? = Nuclide is part of an undetermined solution

X = Nuclide rejected by the interference analysis

@ = Nuclide contains energy lines not used in Weighted Mean Activity

Errors quoted at 1.000 sigma

# How to calculate the $L_D$

- The  $L_D$  is calculated under detection limits -> Currie MDA
  - It is always calculated in units of activity or activity concentration
- Editable parameters are
  - Confidence factor
  - The ROI width
  - Variable MDA constants
- Recommendation: Change the ROI width from the defaults
- $L_D = k^2 + 2L_C = k^2 + 2k\sigma_0 = C_0 + C_1\sigma_0$

The screenshot shows the 'Minimum Detectable Activity (MDA) Setup' dialog box. It contains the following fields and controls:

- Confidence Factor:** A text box with '5.00' and a '%' symbol.
- Perform variable ROI widths:** A checked checkbox. To its right is 'ROI Width: +/-' followed by a text box with '1.25' and 'FWHM'.
- Use variable MDA constants:** An unchecked checkbox. Below it are 'Add Constant:' with a text box containing '2.71' and 'Multiplier:' with a text box containing '3.29'.
- Cascade correction:** A section containing:
  - Coincidence library:** A text box with 'C:\GENIE2K\CAMFILES\coi\_lib.clb' and a 'Select...' button.
  - Geometry Composer file:** A text box and a 'Select...' button.
  - Perform cascade correction:** An unchecked checkbox.
- Generate Report:** A checked checkbox.
- Buttons:** 'Cancel', 'Help', and 'Execute' at the bottom.

# Example report

- Genie calculates the  $L_D$  (MDA) for every line and uses the lowest value for as Nuclide  $L_D$
- For non-identified nuclides the Activity column is the observed signal recalculated into activity or activity concentration
- The upper limit would be the activity +k times the activity uncertainty

```
*****
*****          N U C L I D E   M D A   R E P O R T          *****
*****
```

Detector Name:  
Sample Geometry: @25 cm  
Sample Title: GENIE-PC Spectrum No. 1  
Nuclide Library Used: C:\GENIE2K\CAMFILES\STDLIB.NLB

	Nuclide Name	Energy (keV)	Yield (%)	Line MDA (uCi/unit)	Nuclide MDA (uCi/unit)	Activity (uCi/unit)
+	K-40	1460.81*	10.67	2.18558E-02	2.186E-02	1.81977E-01
	SC-46	889.25	99.98	2.01045E-03	2.010E-03	6.58658E-04
		1120.51	99.99	2.20785E-03		2.99193E-03
+	CO-57	122.06*	85.51	1.82702E-03	1.827E-03	2.91508E-02
		136.48*	10.60	8.35294E-03		3.33071E-02
+	CO-60	1173.22*	100.00	3.13130E-03	1.228E-03	9.93055E-02
		1332.49*	100.00	1.22829E-03		9.71557E-02
+	SE-75	96.73	3.41	2.84636E-02	1.333E-03	-3.59854E-03
		121.11*	16.70	1.18514E-02		1.89094E-01
		136.00*	59.20	1.89474E-03		7.55523E-03
		198.60	1.45	5.50959E-02		2.82458E-02
		264.65	59.80	1.33279E-03		-2.98804E-04
		279.53*	25.20	6.72441E-03		9.62423E-02
		303.91	1.32	6.19906E-02		1.95332E-02
		400.65	11.40	8.82940E-03		-4.98404E-03
+	KR-85	513.99*	0.43	3.98442E-01	3.984E-01	8.53421E+00
	@ KR-85M	151.18	75.30	1.00000E+20	1.000E+20	1.00000E+20
	@	304.87	14.00	1.00000E+20		1.00000E+20
+	SR-85	513.99*	99.27	3.72878E-03	3.729E-03	7.98665E-02
+	Y-88	898.02*	93.40	4.66434E-03	2.334E-03	2.94791E-01
		1836.01*	99.38	2.33399E-03		2.92102E-01
+	CD-109	88.03*	3.72	3.77177E-02	3.772E-02	3.73414E-01
+	SN-113	255.12	1.93	4.43331E-02	2.709E-03	7.22656E-02
		391.69*	64.90	2.70938E-03		6.54878E-02
+	CS-134	475.35	1.46	5.44269E-02	1.004E-03	1.93325E-02
		563.23	8.38	1.05376E-02		4.26918E-03
		569.32	15.43	5.89546E-03		7.19708E-03
		604.70*	97.60	1.00352E-03		6.86604E-03

# Two most important points

- The critical level should be used to determine if a signal is real or not
  - Detect or not detect
- The detection limit is a performance measure of the detection system
  - How large does a signal have to be for me to detect it with 95% probability
  - Should not be used to determine if a signal is real or not.



# Questions?



# Thank you!