Implementing ISO11929 at our laboratories

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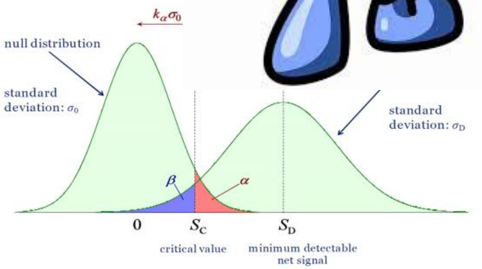


Contents

- Why ISO 11929 ?
- Refreshing decision threshold and detection limit (Currie approach)
- What is ISO 11929 all about ?
  - Characteristic limits
  - Limits of the confidence interval
- How should we report our results according to ISO 11929 ?
- How will we implement this in our laboratories

Why implementing ISO11929 ?

- Many ISO norms involving radioactivity measurements (in use in our labs) are refering to ISO11929 (see list in following slides)
- Authorities (i.e. follow European legislation and guidelines → refering also to ISO11929 so they expect us to work according to this norm
- Some fundamental reasons:
  - MDA value according to Currie is generaly underestimating
  - Measurements close to zero with large uncertainty include negative activity values (which is physically not possible)

ISO norms related tot ISO11929

RADIOLOGICAL PROTECTION (not up to date)

•ISO 11665-1:2012 (Measurement of radioactivity in the environment – Air: radon-222 – Part 1: Origins of radon and its short-lived decay products and associated measurement methods).

•ISO 11665-4:2012 (Measurement of radioactivity in the environment – Air: radon-222 – Part 4: Integrated measurement method for determining average activity concentration using passive sampling and delayed analysis).

•ISO 11665-5:2012 (Measurement of radioactivity in the environment – Air: radon-222 – Part 5: Continuous measurement method of the activity concentration).

•ISO 11665-6:2012 (Measurement of radioactivity in the environment – Air: radon-222 – Part 6: Spot measurement method of the activity concentration).

•ISO 18589-1:2005 (Measurement of radioactivity in the environment – Soil – Part 1: General guidelines and definitions.

•ISO 18589-3:2007 (Measurement of radioactivity in the environment – Soil – Part 3: Measurement of gamma-emitting radionuclides).

•ISO 18589-4:2009 (Measurement of radioactivity in the environment – Soil – Part 4: Measurement of plutonium isotopes (plutonium 238 and plutonium 239 + 240) by alpha spectrometry).

•ISO 18589-5:2009 (Measurement of radioactivity in the environment – Soil – Part 5: Measurement of strontium 90).

•ISO 18589-6:2009 (Measurement of radioactivity in the environment – Soil – Part 6: Measurement of gross alpha and gross beta activities).

•ISO 18589-7:2013 (Measurement of radioactivity in the environment – Soil – Part 7: In situ measurement of gamma-emitting radionuclides).

•ISO 28218:2010 (Radiation protection – Performance criteria for radiobioassay).

ISO norms related tot ISO11929

ISO TC 85 SC 3 (NUCLEAR FUEL CYCLE) (not up to date)

•ISO 11483:2005 (Nuclear fuel technology – Preparation of plutonium sources and determination of 238Pu/239Pu isotope ratio by alpha spectrometry).

•ISO 21847-1:2007 (Nuclear fuel technology – Alpha spectrometry – Part 1: Determination of neptunium in uranium and its compounds).

•ISO 21847-2:2007 (Nuclear fuel technology – Alpha spectrometry – Part 2: Determination of plutonium in uranium and its compounds).

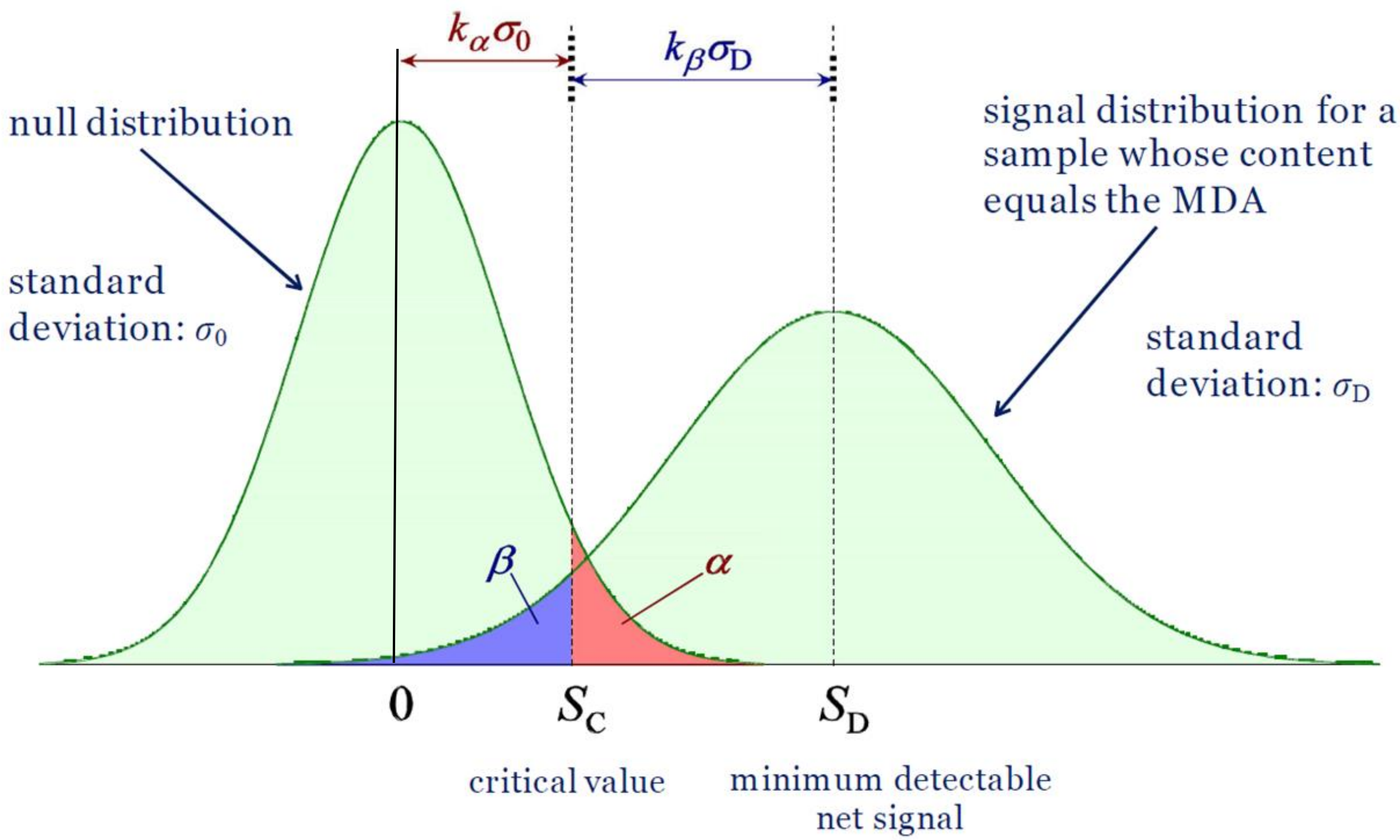
•ISO 21847-3:2007 (Nuclear fuel technology – Alpha spectrometry – Part 3: Determination of uranium 232 in uranium and its compounds).

ISO norms related tot ISO11929

ISO TC147 SC3 (RADIOACTIVITY) (not up to date)

- ISO 9696:2007 (Water quality – Measurement of gross alpha activity in non-saline water – Thick source method).
- ISO 9697:2008 (Water quality – Measurement of gross beta activity in non-saline water – Thick source method).
- ISO 9698:2010 (Water quality – Determination of tritium activity concentration – Liquid scintillation counting method).
- ISO 10703:2007 (Water quality – Determination of the activity concentration of radionuclides – Method by high resolution gamma-ray spectrometry).
- ISO 10704:2009 (Water quality – Measurement of gross alpha and gross beta activity in non-saline water – Thin source deposit method).
- ISO 11704:2010 (Water quality – Measurement of gross alpha and beta activity concentration in non-saline water – Liquid scintillation counting method).
- ISO 13160:2012 (Water quality – Strontium 90 and strontium 89 – Test methods using liquid scintillation counting or proportional counting).
- ISO 13161:2011 (Water quality – Measurement of polonium 210 activity concentration in water by alpha spectrometry).
- ISO 13162:2011 (Water quality – Determination of carbon 14 activity – Liquid scintillation counting method).
- ISO 13163:2013 (Water quality – Lead-210 – Test method using liquid scintillation counting).
- ISO 13164-1:2013 (Water quality – Radon-222 – Part 1: General principles).
- ISO 13164-2:2013 (Water quality – Radon-222 – Part 2: Test method using gamma-ray spectrometry.
- ISO 13164-3:2013 (Water quality – Radon-222 – Part 3: Test method using emanometry).
- ISO 13165-1:2013 (Water quality – Radium-226 – Part 1: Test method using liquid scintillation counting).
- ISO 13165-2:2014 (Water quality – Radium-226 – Part 2: Test method using emanometry).
- ISO 13166:2014 (Water quality – Uranium isotopes – Test method using alpha-spectrometry).

Decision threshold and detection limit
Currie approach



Definitions

- The detection limit L_D is the smallest value of the measurand above the decision level for which the wrong assumption that the physical effect is absent does not exceed the specified probability β
- The quantity L_D is used to find out whether a measurement procedure is suitable for the measurement purpose e.g. in compairing with a specified reference value L_R

Decision threshold and detection limit
Currie approach

- A count exceeding S_C is detected
- A count below S_C is not detected
- But there is a probability α of a false decision that a signal has been detected while there was no signal

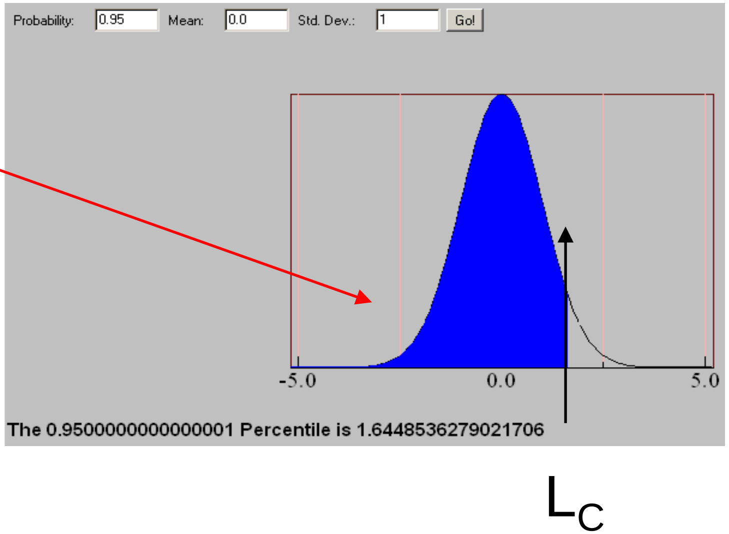
  FALSE POSITIVE
- A count equal to S_D has a probability β of deciding that there is no signal while there was:

  FALSE NEGATIVE
- “The detection limit S_D is the smallest value of the measurand above the decision level for which the wrong assumption that the physical effect is absent does not exceed the specified probability β”

Decision threshold and detection limit
Currie approach

- General case: $k_{1-\alpha} = 1.645$ ($\alpha = 0.05$)

$$L_C = 1.645\,\sigma_0$$



- Simple counting

$$L_C = 1.645\sqrt{2B}$$

- Simple peak analysis (gamma spectrometry, n and m number of channels in ROIs)

$$L_C = 1.645\sqrt{B\left[1+\frac{n}{2m}\right]}$$

Decision threshold and detection limit
Currie approach

$$\sigma^2(N) = \sigma^2(G) + \sigma^2(B)$$

$$\sigma^2(N) = \sigma^2(N + B) + \sigma^2(B)$$

$$\sigma^2(0) = \sigma^2(0 + B) + \sigma^2(B)$$

$$\sigma^2(0) = 2\sigma^2(B)$$

$$\boxed{\sigma(0) = \sqrt{2B}}$$

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Decision threshold and detection limit Currie approach

- general case:

$$L_D = 2.71 + 3.29\,\sigma_0$$

- Simple counting

$$L_D = 2.71 + 3.29\sqrt{2B}$$

- Peak integration

$$L_D = 2.71 + 3.29\sqrt{B\left[1 + \frac{n}{2m}\right]}$$

Lloyd A. Currie: “Limits for Qualitative Detection and Quantitative Determination”
Anal. Chem. Vol.40, No.3, 1968

Decision threshold and detection limit
Currie approach

$$L_D = 2.71 + 3.29\,\sigma_0$$

Counts

$$MDA = \frac{2.71 + 3.29\,\sigma_0}{\varepsilon\, I\, t}$$

Activity

But these parameters are not free of uncertainty

Lloyd A. Currie: "Limits for Qualitative Detection and Quantitative Determination"
Anal. Chem. Vol.40, No.3, 1968

What is ISO 11929 all about ? decision threshold & detection limit

- A procedure for the estimation of characteristic limits:

  Decision threshold y^* and **detection limit** $y^\#$

- Basis(1) *ISO/IEC Guide 98-3, GUM*
  - y primary measurement result ($y < 0$ *no problem*)
  - $u(y)$ primary standard uncertainty
  - Model of measurand $Y = G(X_1, X_2, X_3, X_4, \ldots . X_m)$
- Basis(2) *Bayesian statistics*
  - $\tilde{y}$ true value of measurand (**y ≥ 0;** ***non-negative measurand***)
  - $\hat{y}$ best estimate (non negative)
  - $\tilde{u}(\tilde{y})$ standard uncertainty of the true value
  - $u(\hat{y})$ standard uncertainty associated with $\hat{y}$

Decision treshold & detection limit
According to ISO11929

- Decision threshold:

$$y^* = k_{1-\alpha}\tilde{u}(0)$$

Standard uncertainty when zero activity

- Detection limit:

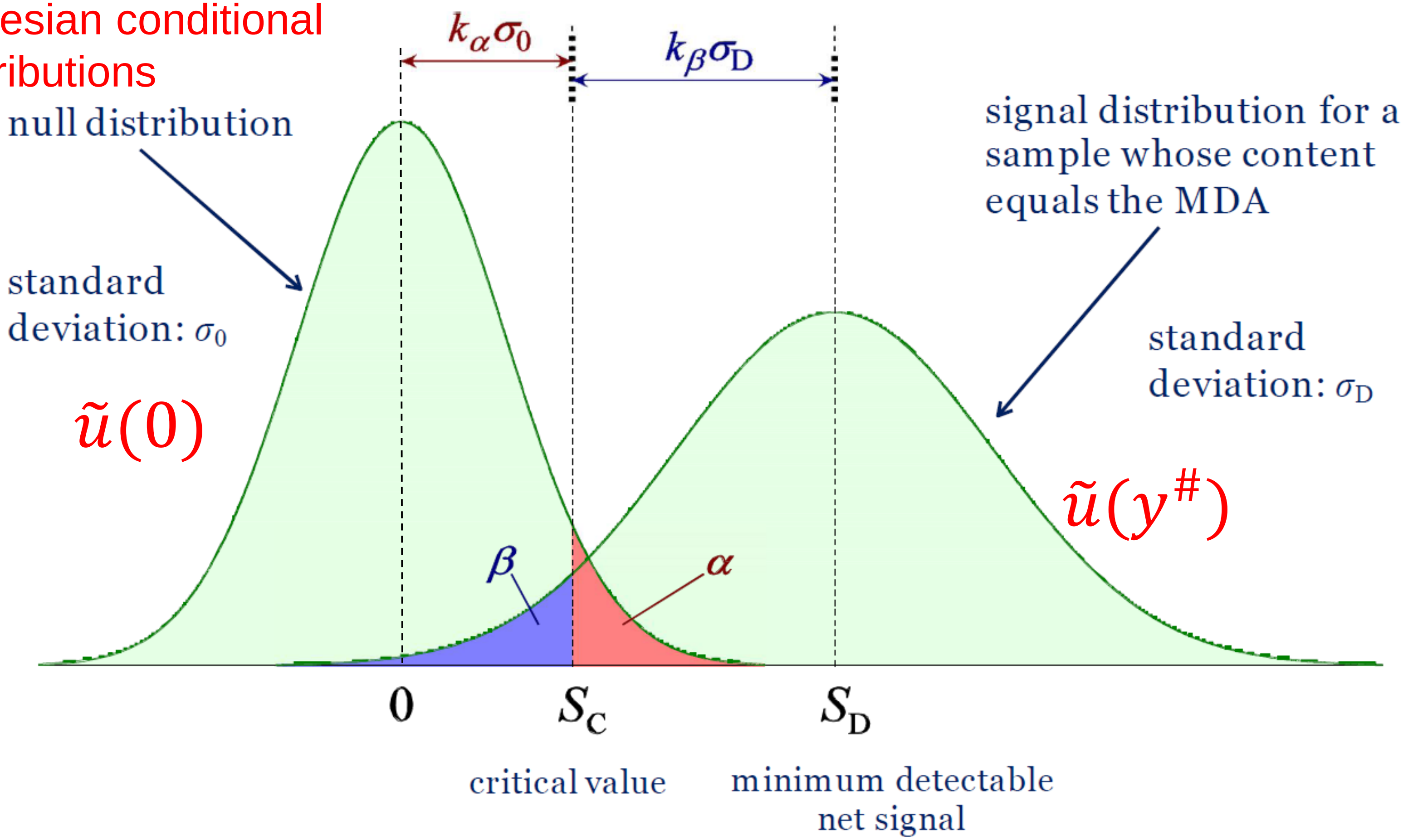
$$y^\# = y^* + k_{1-\beta}\tilde{u}(y^\#)$$

Standard uncertainty when activity = $y^\#$

Implicit equation

What is ISO 11929 all about ?
decision treshold & detection limit

Bayesian conditional distributions



Decision threshold Detection Limit

How does this relates to the Currie formula ?

- Currie formula (counts) is obtained considering a simplified model (linear model -> $\tilde{u}^2(\tilde{y}) = c_0 + c_1\tilde{y}$)
- Currie formula (activity, MDA) is obtained by assuming uncertainty for the conversion factor w, (u(w)).

$$A = Nw$$

This is detection efficiency and some other conversion parameters (mass, volume…)

$$u(A) = Nw\sqrt{\frac{u(N)^2}{N^2} + \frac{u(w)^2}{w^2}}$$

How does this relates to the Currie formula ?

- Decision level (*activity or activity concentration*)

$$L_C = kw\sqrt{\frac{b}{t_s} + \frac{b}{t_0}}$$

OK

- Detection limit (*activity or activity concentration*)

$$L_d = \frac{2L_C + \frac{k^2 w}{t_s}}{1 - k^2 u(w)_{rel}{}^2}$$

This is the difference →

These simplified formulae are the result of the selection $k_{(1-\alpha)} = k_{(1-\beta)} = k$
$u^2(y) = c + y$

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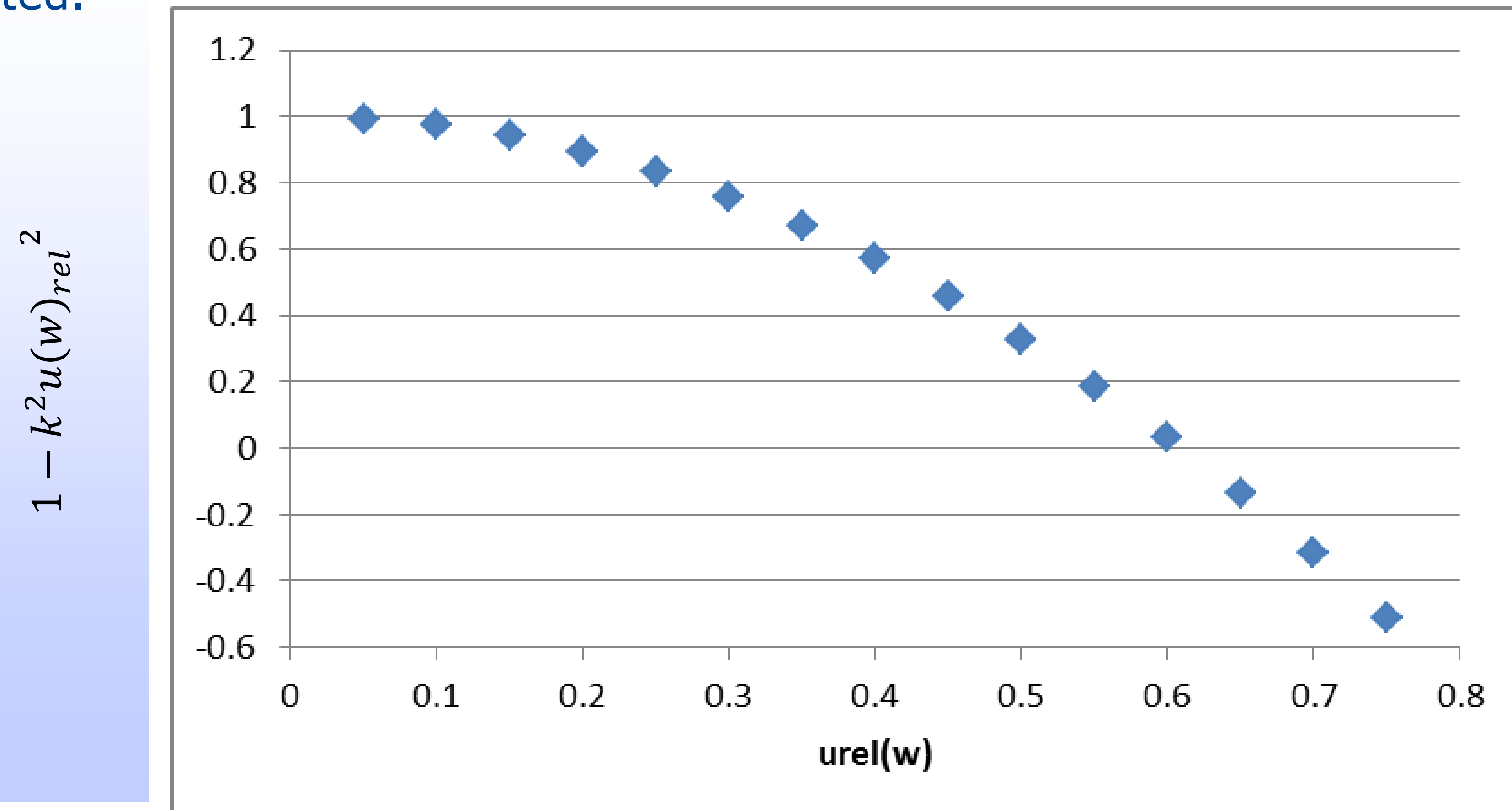
How does this relates to the Currie formula ?

For small relative uncertainty of *w* the correction factor is almost equal to 1 (but for large values may be negative!, no detection limit can be computed.



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How does this relates to the Currie formula ?

- MDA is not obtained by taking DL (counts) and scale this to activity
  - Uncertainty on the scaling factor is involved, but when this factor is small, MDA is almost DL (activity)

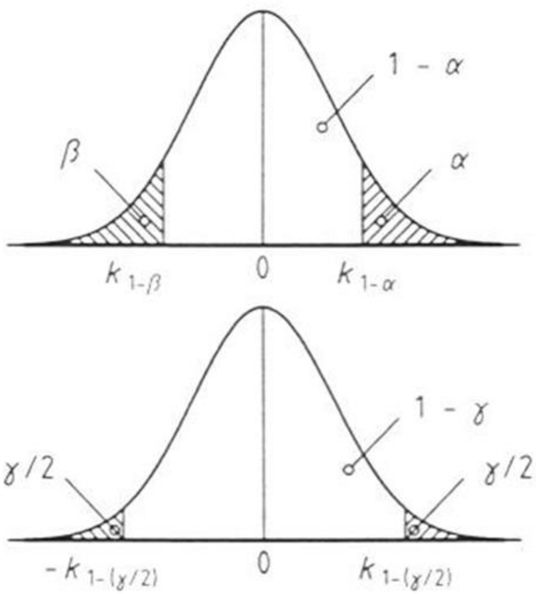
- Pitfall: simplified models following ISO 11929 may still fail to produce acceptable results
  - in low count rate applications (alpha spec ?)
  - $\sqrt{n+1}$ is to be considered for small n
  - If background counts do not follow poisson statistics

    (e.g. due to other variations involved)

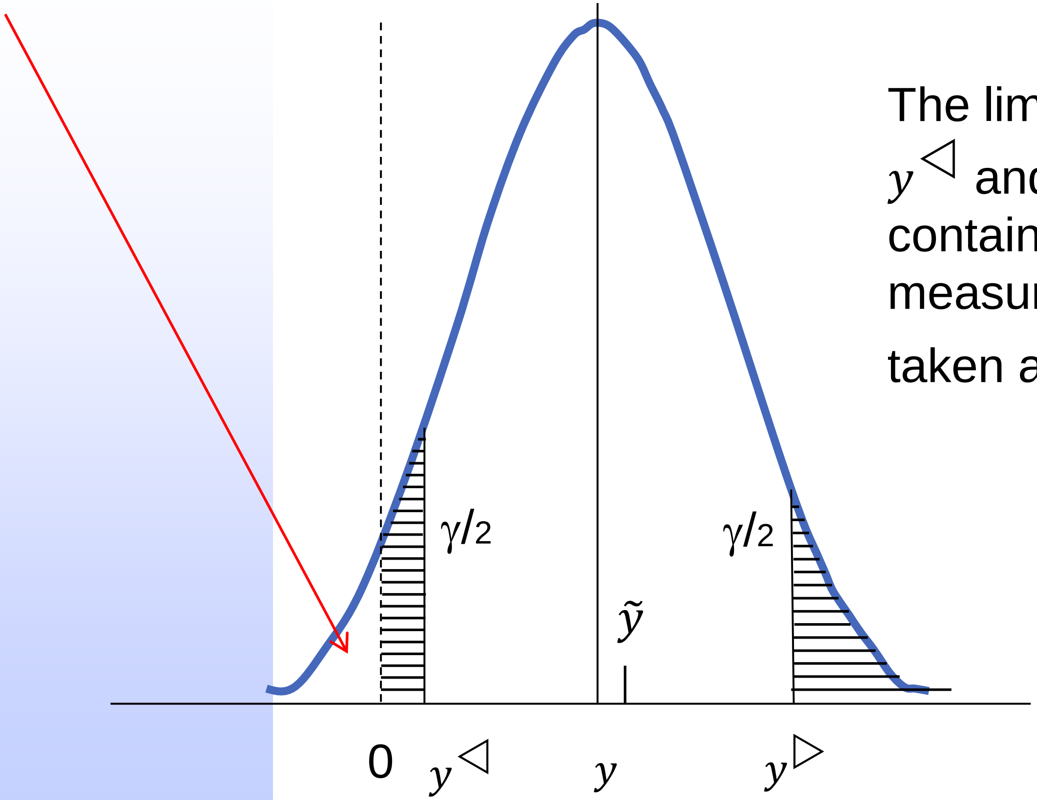
How does this relates to the Currie formula ?

Table 2 — Values $k_{1-\alpha}$, $k_{1-\beta}$, $k_{1-(\gamma/2)}$ as a function of the error probabilities α and β and of the confidence level $1-\gamma$ (quantiles of normal distribution)

Error of probability α or β	Confidence level $1-\gamma$	$k_{1-\alpha}$, $k_{1-\beta}$, $k_{1-(\gamma/2)}$
0,1586	0,682	1,000
0,1000	0,800	1,282
0,0500	0,900	1,645
0,0250	0,950	1,960
0,0228	0,955	2,000
0,0100	0,980	2,326
0,0050	0,990	2,576
0,0014	0,997	3,000
0,0010	0,998	3,090



The limits of the confidence interval take into account the fact that the measurand is non-negative although *y* can be negative.



The limits of the confidence interval $y^{\triangleleft}$ and $y^{\triangleright}$ define a confidence interval containing the true value $\tilde{y}$ of the measurand with a specified probability taken as $1 - \gamma$

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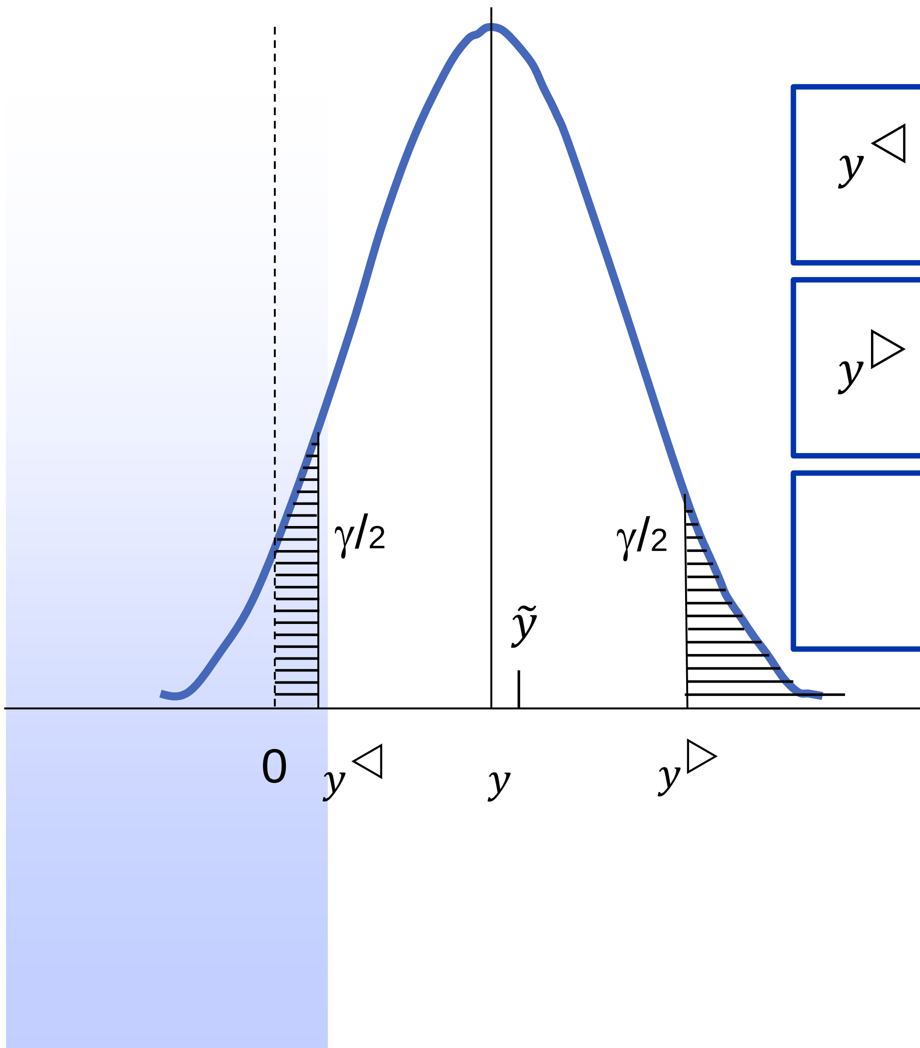
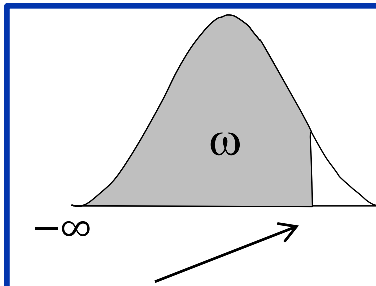
What is ISO 11929 all about ?
Best estimate
Limits of the confidence interval



$$y^{\triangleleft} = y - k_p\, u(y) \qquad p = \omega.(1 - \frac{\gamma}{2})$$

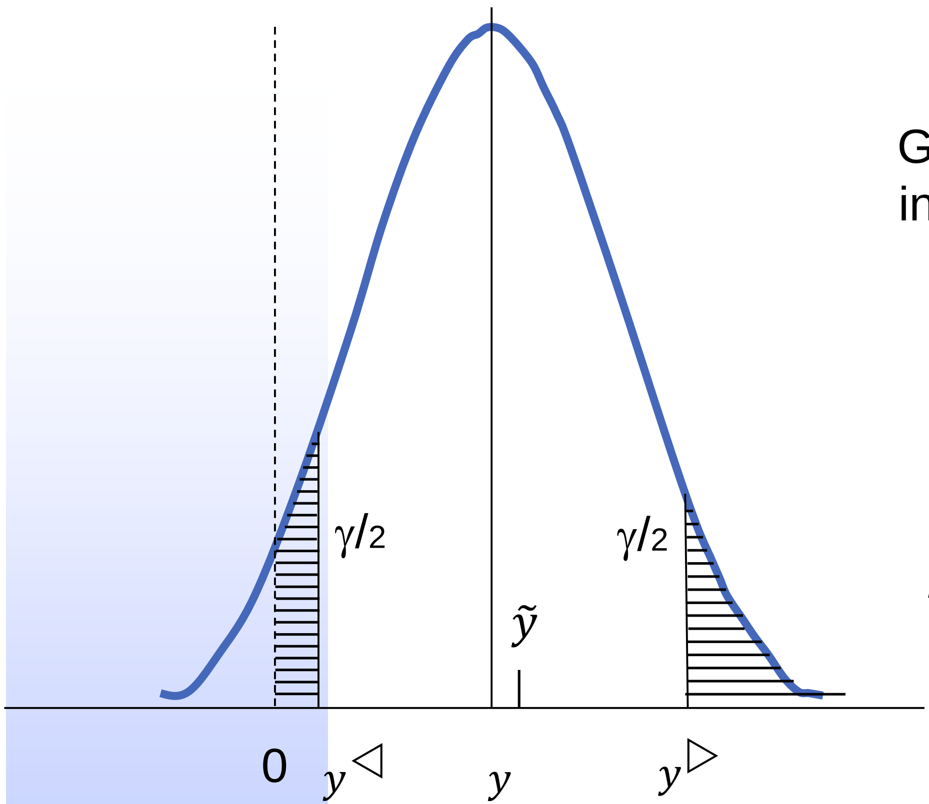
$$y^{\triangleright} = y + k_q\, u(y) \qquad q = 1 - \frac{\omega\gamma}{2}$$

$$\omega = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\frac{y}{u(y)}} exp\left(-\frac{\nu^2}{2}\right) d\nu$$



$$\frac{y}{u(y)} > 4 \rightarrow \omega \approx 1$$

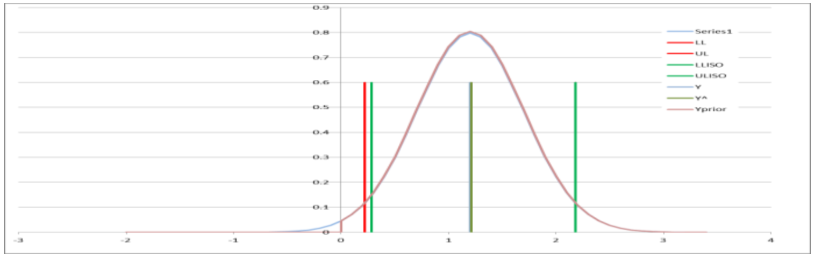
What is ISO 11929 all about ?
Best estimate
Limits of the confidence interval

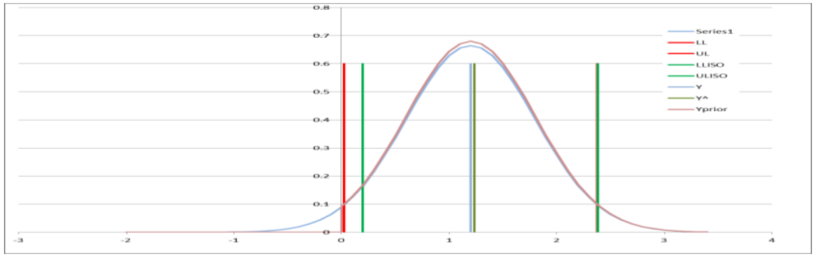


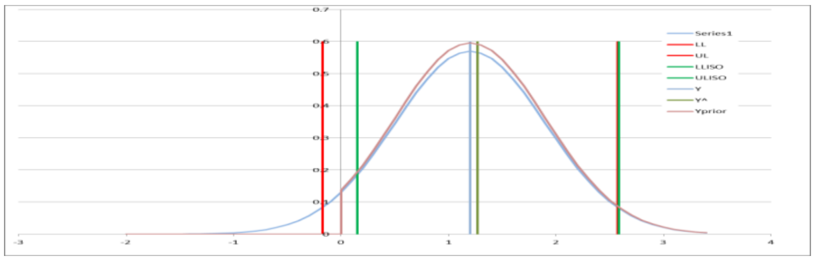
Generally the limits of the confidence interval are not symmetrically around

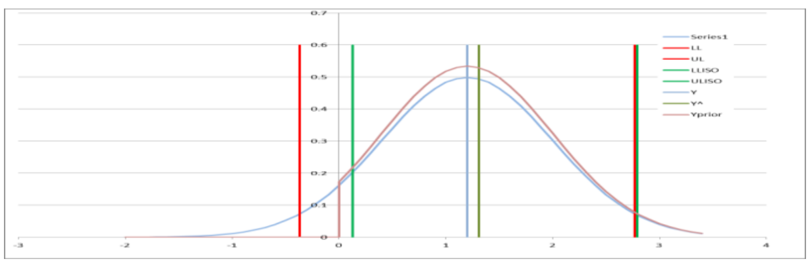
y or $\tilde{y}$

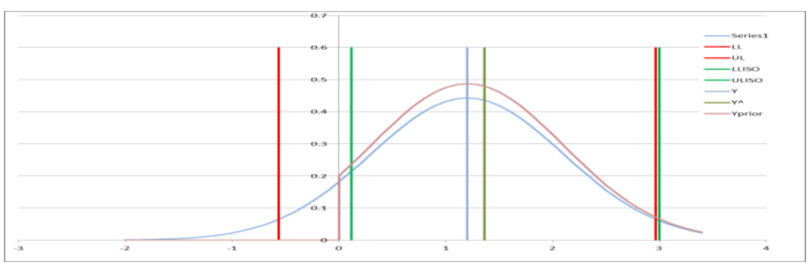
$$\tilde{y} = y + \frac{u(y)\exp\left(-\frac{y^2}{2\,u^2(y)}\right)}{\omega\sqrt{2\pi}}$$

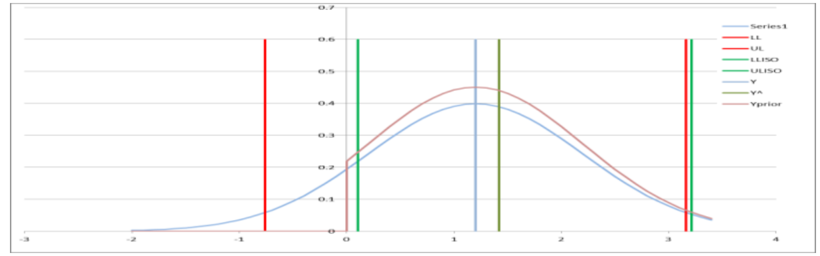


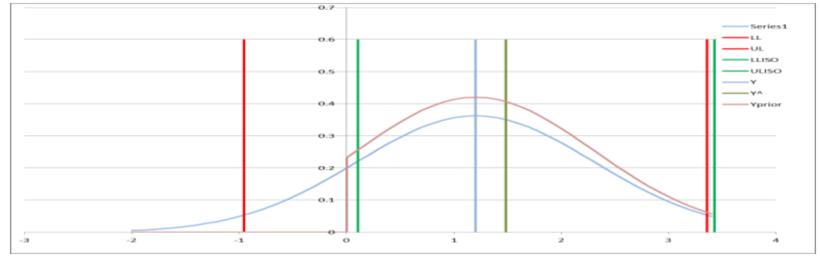












Examples of confidence limits and best estimate conversion for different u(y)/y values

These are easily computed using predifined functions in Excel

What is ISO 11929 all about ?
Best estimate
Limits of the confidence interval

γ	Risk of exceeding quoted confidence limits	None	Set by user
ω	Required to calculate p and q	$\Phi\left[\frac{y}{u(y)}\right]$	Use $norm.s.dist\left(\left[\frac{y}{u(y)}\right], true\right)$
p	Required to calculate k_p	$\omega.\left(1-\left[\frac{\gamma}{2}\right]\right)$	
q	Required to calculate k_q	$1-\left[\frac{\omega.\gamma}{2}\right]$	
k_p	Coverage factor for lower confidence limit	Complex	Use $norm.s.inv(p)$
k_q	Coverage factor for lower confidence limit	Complex	Use $norm.s.inv(q)$
$y^{\triangleleft}$	Lower confidence limit	$k_p.u(y)$	
$y^{\triangleright}$	Upper confidence limit	$k_q.u(y)$	
$\hat{y}$	Best estimate of y when $\frac{y}{u(y)} < 4$	$y+\frac{u(y).e^{\left\{\frac{-y^2}{\left[2.(u(y))^2\right]}\right\}}}{\omega.\sqrt{2.\pi}}$	
$u(\hat{y})$	Best estimate of $u(\hat{y})$ when $\frac{y}{u(y)} <$ 4	$\sqrt{u^2(y)-(\hat{y}-y).\hat{y}}$	

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What is wrong with my *old* expanded uncertainty ?

- Expanded uncertainty for activity can yield a confidence interval including negative values
  - This is corrected by ISO 11929
  - But it requires at least 3 numbers to be specified (limits of confidence and best estimate)
  - When relative uncertainty is small
    - $\rightarrow \tilde{y} \approx y$
    - $\rightarrow \mathrm{u}(\tilde{y}) \approx u(y)$

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How to report ?

Condition	Report	Comments
$y < y^*$	$< y^*$	The effect is not detected. Qualify this information with: *'This is the decision threshold for ^{m}A in this analysis; ^{m}A has not been detected in this analysis.'*
$y^* < y < y^\#$	$< y^\#$	The effect is detected, but not quantifiable. Qualify this information with: *'This is the detection limit for ^{m}A in this analysis, and is approximately twice the decision threshold; it is possible that ^{m}A has been detected, but is not quantifiable in this analysis.'*
$y^\# < y < 4.u(y)$	$\hat{y} \pm k.u(\hat{y})$	A best estimate of the result may be reported. This information ***may*** be qualified with: *'^{m}A has been identified and quantified in this analysis, although the result is close to the detection limit, $y^\#$, which is reflected in the relatively large uncertainty.'*
$4.u(y) < y$	$y \pm k.u(y)$	The result may be unambiguously reported and no additional qualification is needed. It ***may*** be instructive for the user if this statement is made: *'^{m}A has been unambiguously identified and quantified in this analysis, where the detection limit for this analysis is $y^\#$'*

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NPL

Conclusion

- ISO 11929 allows also to deal with the special situations
- Pitfalls can be associated with the validity of the models used
- Currie approach is a simplified model compatible with ISO11929 if uncertainty on conversion factor (to go from counts to activity) is also considered
  - Breaks down at low count rates (as before)
- When not detected: report detection limit (as proof of what the method can measure)
- Reporting according to ISO11929: in case of negative values in confidence interval or important relative uncertainty:
  - Report limits of confidence interval (2 numbers)
  - Best estimate
  - Primary results ?

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Some further comments

For large values of relative systematic uncertainty we find that the ISO 11929 method tends to overestimate the MDA, and for very large values it fails to return a finite or physically meaningful value at all. Furthermore, for small values of the systematic uncertainty, the correction provided by the ISO 11929 MDA compared to the traditional Currie MDA is very small, and for applications in this regime adoption of the ISO approach may not provide significant benefits.

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Minimum detectable activity, systematic uncertainties, and the ISO 11929 standard

J. M. Kirkpatrick · R. Venkataraman · B. M. Young

How to implement ISO11929 in our labs

- Spreadsheet reporting ⟶ change formulea
- Commercial software
  - Genie 2K gammaspec:→ ISO11929 is included (CAMparameters)
- Home made softwares
  - Change formulae
- At very low counts -> choise of any alpha & beta not free!
- Reporting according to ISO11929: in case of negative values in confidence interval or important relative uncertainty:
  - Many reporting tools are not ready yet for this situation

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