Uncertainty calculations (GUMUF and MC), and calculation of critical limits

Henrik Ramebäck
Swedish Defence Research Agency,
FOI, Umeå, Sweden

Invited lecture at NKS GammaSkills 2023

© Swedish Defence Research Agency

Covered during the training day

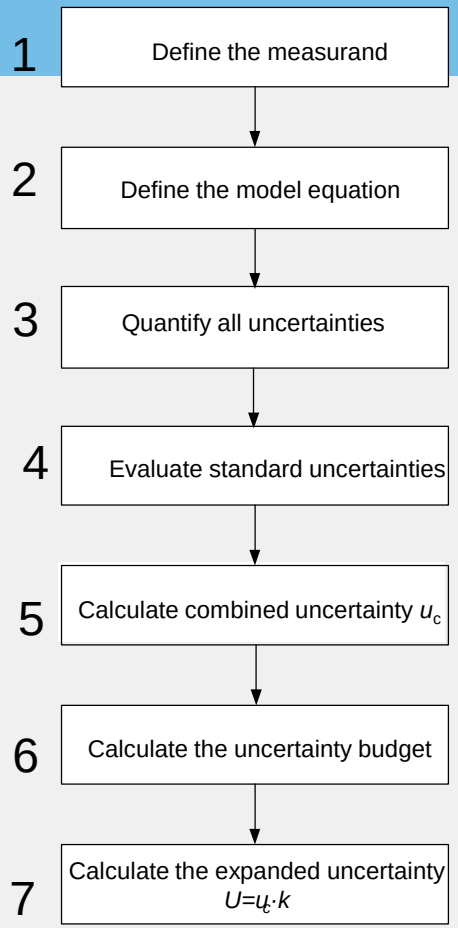
GUMUF *vs.* MC

© Swedish Defence Research Agency

The GUM method:

A tool for easy uncertainty propagation is beneficial!
Suggestion: Kragten spreadsheet
(or a dedicated software…)

J. Kragten, 1995, Chemom. Intell. Lab. Syst., 28, 89



+8: Reporting the measurement result

© Swedish Defence Research Agency

The GUM Uncertainty Framework (uncertainty propagation)

Holds when:

- The model equation is linear (or the uncertainties of non-linear input quantities are ’small enough’)
- The probability distributions of the input quantities are normal

➔The GUMUF gives (almost) exact results
but can otherwise often give results that are fit-for-purpose for most applications…

© Swedish Defence Research Agency

The GUM Uncertainty Framework (uncertainty propagation)

Does not hold when:

- The model equation is non-linear
- The probability distributions of the input quantities are not normal and have a significant contribution to the uncertainty

  ~~➔The GUMUF gives (almost) exact results but can otherwise often give results that are fit-for-purpose for most applications~~…

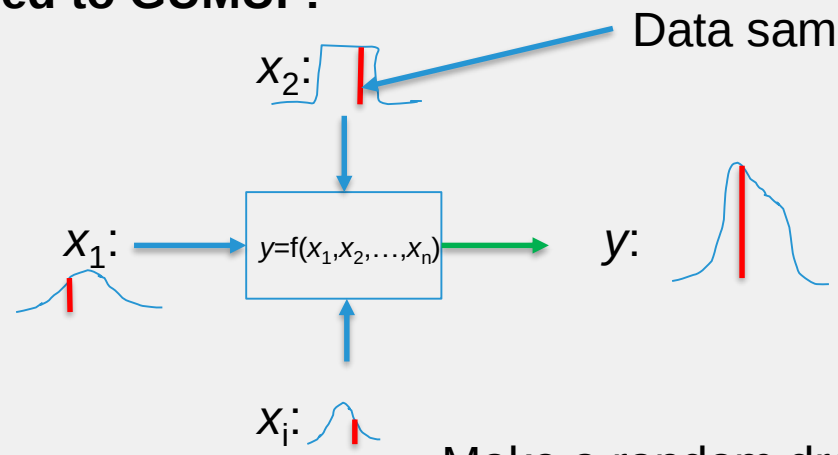
- ➔Propagate distributions (MC)
- Might have implications on mean value and percentiles (Remember: decisions!)

© Swedish Defence Research Agency

MC calculation: The principle

Accually more intuitive compared to GUMUF!

Propagation of distributions:



Data is sampled randomly from the PDFs of each input quantity
➔Many data needed to ensure sampling from the tails of *e.g.* Gaussian PDF (where the probability is low)

Make a random draw from each distribution and calculate the result
Repeat this *n* times
➔Calculate the resulting pdf

© Swedish Defence Research Agency

Distributions

- Here we will consider draws from two distributions:

  -Normal
  -Rectangular

- Draws from other distributions, see JCGM 101:2008

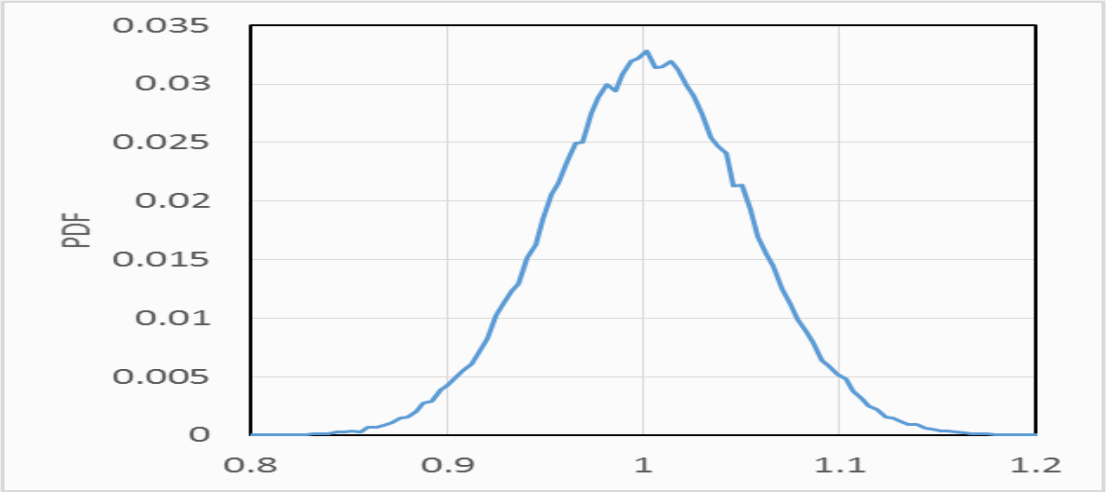
© Swedish Defence Research Agency

Data sampling: Normal distribution

Normal distribution:

- Random samples from the distribution :

- Excel: *x*=NORM.INV(*prob; mean; std unc*)
  prob=RAND()
  mean=1
  std unc=0.05
- *n*=100 000:

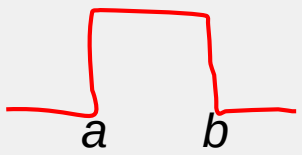


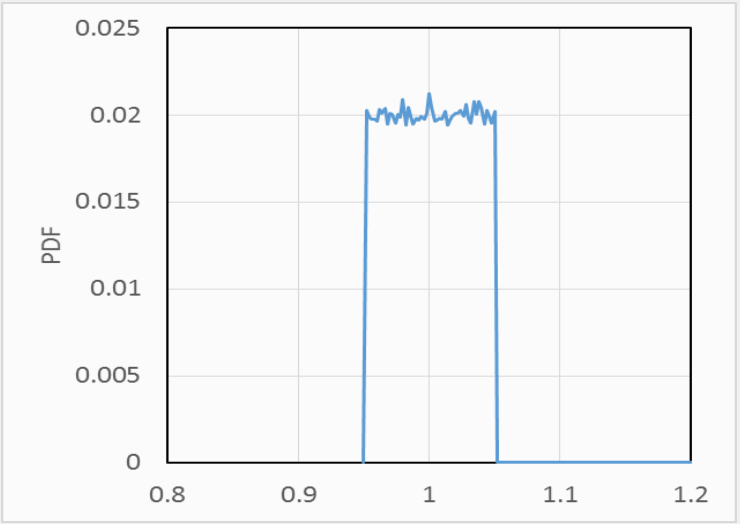
© Swedish Defence Research Agency

Data sampling: Rectangular distribution

Rectangular distribution:

- Random samples from the distribution:
- Excel: $x=a+(b-a)\cdot RAND()$
  $a=0.95$
  $b=1.05$
- $n=100\ 000$:





© Swedish Defence Research Agency

Data analysis in Excel

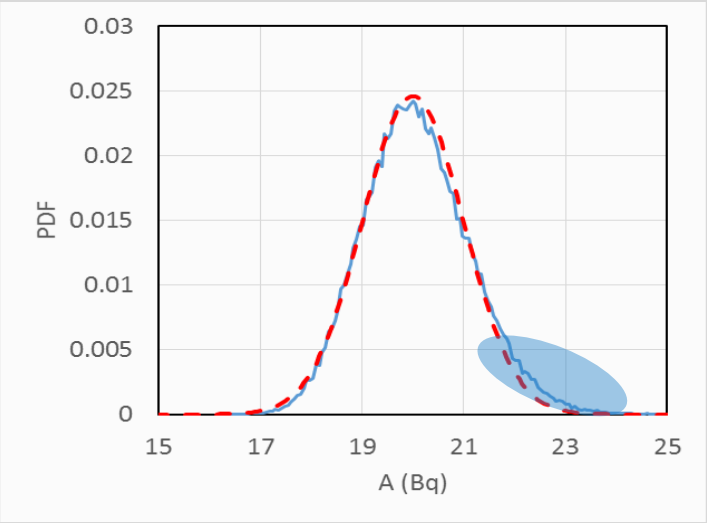
- Analyse the measurand with respect to the mean, standard deviation and confidence interval (CI):
  -AVERAGE(*range*); *range*: the sample size (*n*)
  -STDEV(*range*)
  CI:
  -PERCENTILE(*range*; *prob.*) gives CL for a given *probability*
  Ex: For 95% CI calculate
  PERCENTILE(*range*; 0.025) and
  PERCENTILE(*range*; 0.975)
  ➔Probabalistic symmetric: Instead of ±*a*, stated as (+*b*; -*c*). If linear (enough) *a*≈*b*≈*c*

© Swedish Defence Research Agency

Example: Efficiency transfer

- Uncertainties, norm.dist., k=1:
  -N_{net}: 1%
  -I_γ: 0.1%
  -t_m constant
  -ε: 1%
  - m_{sample}: 0.1%
  -k_{ET}: 5%

$$A = \frac{N_{net}}{t_m \cdot \varepsilon \cdot k_{ET} \cdot I_\gamma \cdot m_{sample}} \cdot (k_{TCS})$$



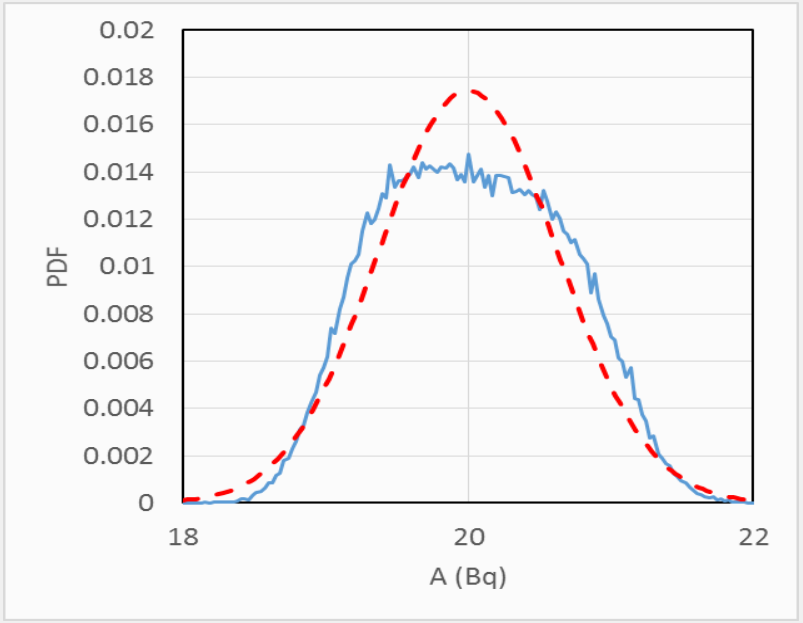
Dashed red: Uncertainty propagation

© Swedish Defence Research Agency

Example: Efficiency transfer

- Uncertainties, norm.dist., k=1:
  - N_{net}: 1%
  - I_γ: 0.1%
  - t_m constant
  - ε: 1%
  - m_{sample}: 0.1%
  - k_{ET}: 5% rect. dist. (half-width)

$$A = \frac{N_{net}}{t_m \cdot \varepsilon \cdot k_{ET} \cdot I_\gamma \cdot m_{sample}} \cdot (k_{TCS})$$



Dashed red: Uncertainty propagation

© Swedish Defence Research Agency

Critical limits:
Decision threshold and detection limit

Why determine decision thresholds and detection limits?

- At the end of the day our measurement results are to be used as a part in a decision making process.
- Is a measurement method fit-for-purpose*, i.e.* can we use it for its intended use?
- What if the detection limit is above *e.g.* a regulatory limit or too close to it?
  ➔The method will not be fit-for-purpose!
- We might then need to modify the method if possible (other geometry, count longer, measure a larger sample,…), or use another method:
  -Gamma spectrometry vs. alpha spectrometry vs.
  mass spectrometry for measurement of *e.g.* ^{241}Am

© Swedish Defence Research Agency

ISO 11929-1:2019

Decision threshold, *y:**
"*value of the estimator of the measurand, which, when exceeded by the result of an actual measurement using a given measurement procedure of a measurand quantifying a physical effect, is used to decide that the physical effect is present*

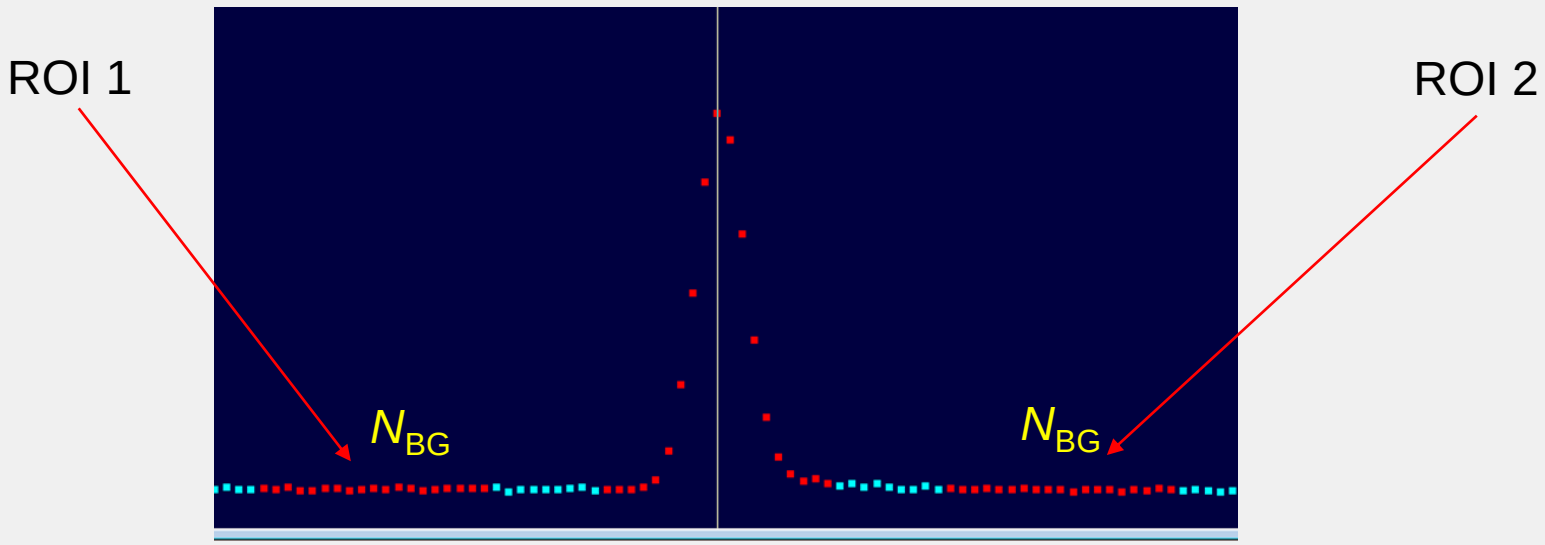
***Detection limit, y#*:**
"smallest true value of the measurand which ensures a specified probability of being detectable by the measurement procedure

Observe terminology:
Detection limit, MDA,…, sensitivity

© Swedish Defence Research Agency

The single ROI based approach

- Here we consider the gross number of counts in a region of interest, ROI, with the same width as the peak and close in energy to the peak
- Regions both above and below the peak may be considered if there is not too much of a step between the high- and low energy side



© Swedish Defence Research Agency

The single ROI based approach

- We will consider the paired observation case
- Applies when $t_{sample}=t_{BG}$
- Or in gamma spectrometry when the ROI of the background estimation has the same width as the peak (same number of channels)

© Swedish Defence Research Agency

ISO 11929-1:2019

The estimate *y* of the measurand *Y* is a function of several input quantities:

$$y = f(x_1, x_2, \ldots, x_n)$$

A model equation for radiation measurements:

$$y = w \cdot (N_G - N_{BG})$$

(*w* is the conversion factor for calculating *e.g.* an activity per mass unit from, here, a number of observed counts in the sample and background measurements)
Uncertainty propagation applying the GUMUF gives:

$$u(y)^2 = w^2 \cdot (N_G + N_{BG}) + (N_G - N_{BG})^2 \cdot u(w)^2$$

© Swedish Defence Research Agency

ISO 11929-1:2019

…: $$u(y) = \sqrt{wy + 2N_{BG}w^2 + y^2\frac{u(w)^2}{w^2}}$$

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

Detection limit: $y^\# = y^* + k_{1-\beta}\,u(y^\#)$

Set $k_{1-\alpha}=k_{1-\beta}=k$

y=0: $y^* = kw\sqrt{2N_{BG}}$

…$y=y^\#$: $y^\# = w\dfrac{k^2 + 2k\sqrt{2N_{BG}}}{1 - k^2u(w)^2_{rel}}$

Currie, or the signal domain (Paired observation)

ISO 11929:2019 considering the uncertainty of the conversion factor w ➔Activity domain

© Swedish Defence Research Agency

Decision threshold, y*

The decision threshold is a quantile of the net background distribution!
α is the risk of a false positive detection (area under the distribution above the critical limit, not including the limit!).

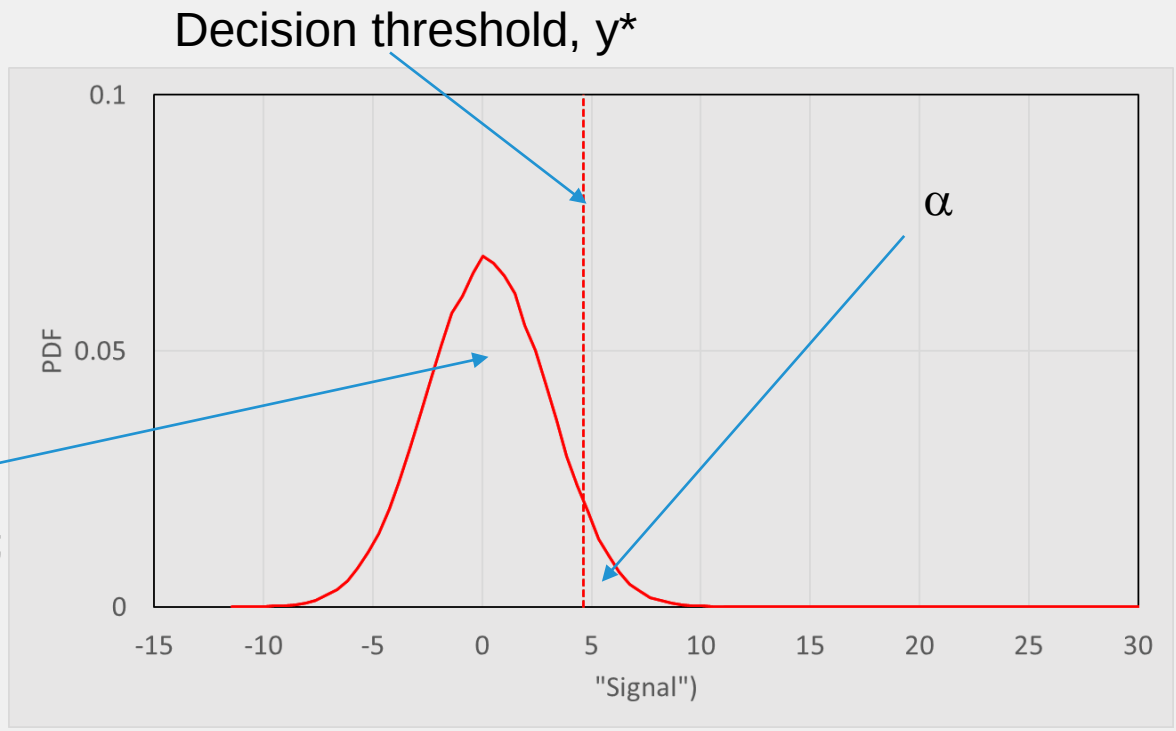
$$y^* = k_{1-\alpha} u(0)$$

Distribution of net signal of the background (no analyte present; paired observations)
(N_{net}=N_{BG}-N'_{BG})

$$u(0) = \sqrt{2N_{BG}}$$

α=5%➔$k_{1-\alpha}$=1.645➔

$$y^* = k_{1-\alpha}\sqrt{2N_{BG}} = 2.326\sqrt{N_{BG}}$$



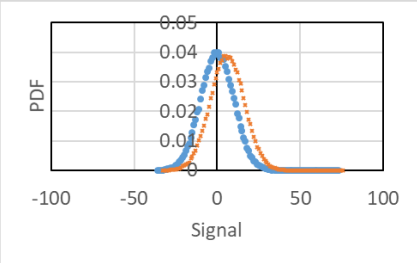
© Swedish Defence Research Agency

Detection limit, $y^{\#}$

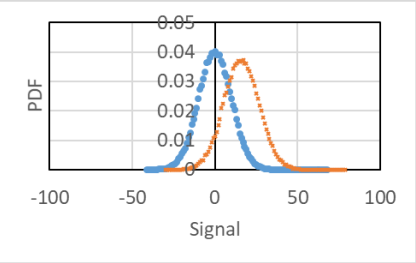
- Decide that something is not present when it is
  ➔Error of the second kind (β)
- Normally we accept a 5% risk that we decide that something is not present although it is (at the detection limit).

$$\beta = P\left(y \leq y^{*} \middle| \tilde{y} = y^{\#}\right)$$

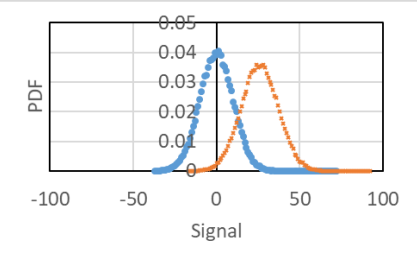
- **Hypothesis test:**
  We want to make the decision if, here, the net signal is statistically different compared to the net background signal for given risks.



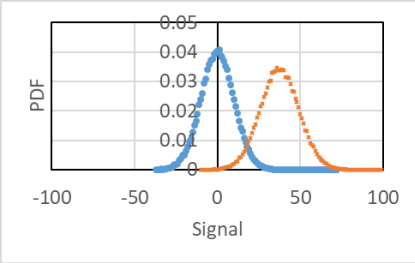
Net signal=5



Net signal=15



Net signal=25



Net signal=L_d=36.5

© Swedish Defence Research Agency

Detection limit

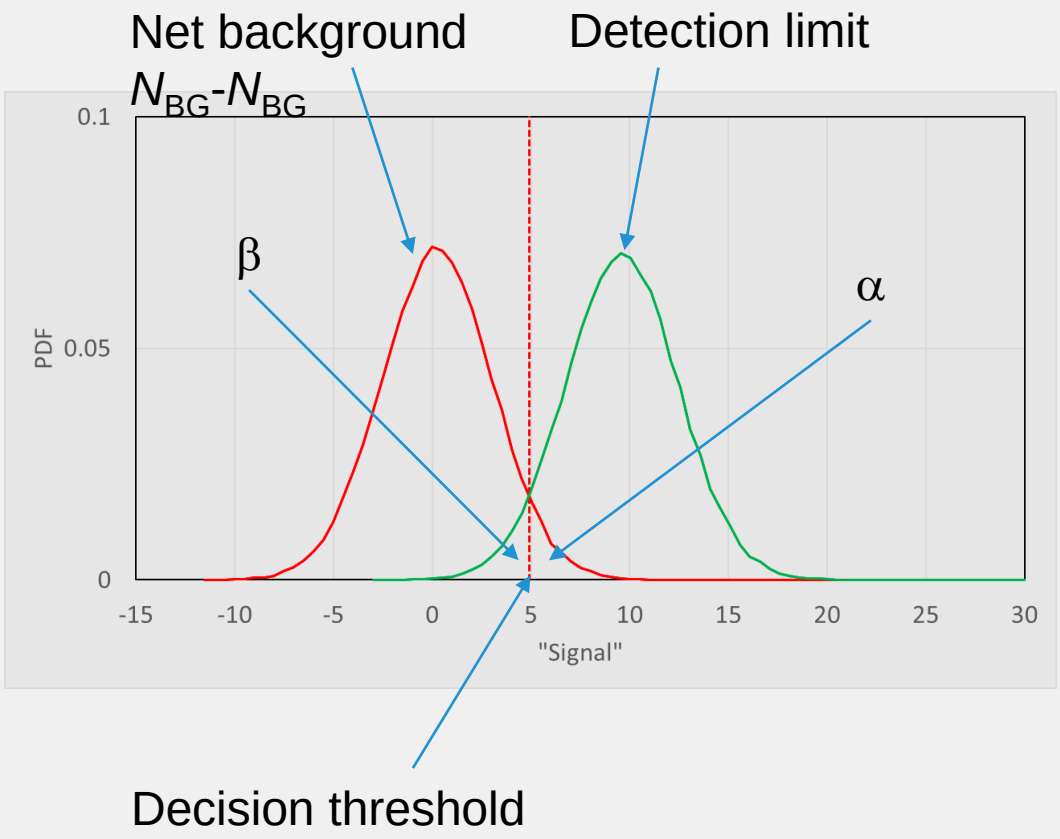
Is the net signal statistically different compared to the net background signal for the *a priori* risks α and β?

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

-Simple case assuming $u_0 \approx u_{y\#}$:
$y^{\#}=2y^{*}$ (detection limit is twice the decision threshold)
(This applies relatively often in gamma ray spectrometry.)

Currie: $y^{\#}=2.71+2y^{*}$ vs. $y^{\#}=2y^{*}$:

	Error if $y^{\#}=2y^{*}$
N_{BG}=30:	1.11
N_{BG}=100:	1.06
N_{BG}=300:	1.03
N_{BG}=1000:	1.02
N_{BG}=3000:	1.01



© Swedish Defence Research Agency

The signal domain vs. the activity domain

- Currie (1968) considered when a *signal* can be considered to be above a critical limit, *e.g.* $y>y^{\#}$ ($N>L_d$)
- What if the uncertainty in the conversion factor, w, transforming a signal to the unit of the measurand is large?

$$\text{Signal} \overset{w}{\rightarrow} \text{Activity}$$

© Swedish Defence Research Agency

The conversion factor, *w*

- Factor to convert an observed instrumental signal/response, *e.g.* counts, to the unit of our measurand (*e.g.* activity concentration [Bq/kg])
- In gamma ray spectrometry:

$$w = \frac{1}{\varepsilon \cdot k_{ET} \cdot t_m \cdot I_\gamma \cdot m}$$

Efficiency; ET correction; measurement time; photon emission probability; mass of sample

(The conversion factor may, of course, also include other correction factors like k_{TCS}, decay correction,…)

© Swedish Defence Research Agency

MDA_{Currie} vs. $MDA_{ISO:11929}$

- *MDA* calculated according to Currie only consider the dispersion in the observed background signal:

$$y^{\#} = MDA_{Currie} = w\left[k^2 + 2k\sqrt{2N_{BG}}\right]$$

- *MDA* calculated according to ISO 11929 consider also the dispersion in all other input quantities

$$y^{\#} = MDA_{ISO11929} = w\frac{k^2 + 2k\sqrt{2N_{BG}}}{1 - k^2 u(w)_{rel}^2}$$

© Swedish Defence Research Agency

The conversion factor and ISO 11929

- When calculating the detection limit in the unit of the measurand, the uncertainty of the conversion factor should be considered
- Implications when the relative uncertainty in *w* is ’large’ (denominator in the equation below becomes <0)

$$MDA_{ISO11929} = \frac{MDA_{"Currie"}}{1 - k_{1-\beta}^2 u_{rel}(w)^2}$$

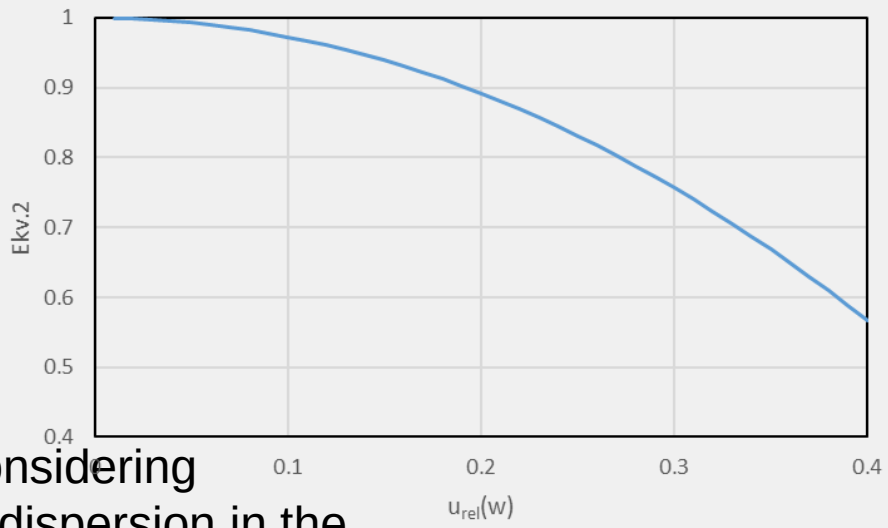
© Swedish Defence Research Agency

The conversion factor and ISO 11929

- For small uncertainties in the conversion factor (<5%, *k*=1) this will only be of marginal consequence, but for larger ones it has to be considered.

’Currie limit’ (in activity) vs. ISO 11929:

$MDA_{Currie}/MDA_{ISO11929}$



’Currie limit’: *MDA* based on Currie not considering the uncertainty in *w*, *i.e.* only considering dispersion in the number of counts (from counting statistics)

© Swedish Defence Research Agency

The conversion factor and ISO 11929

- **When do we have large uncertainies in the conversion factor?**

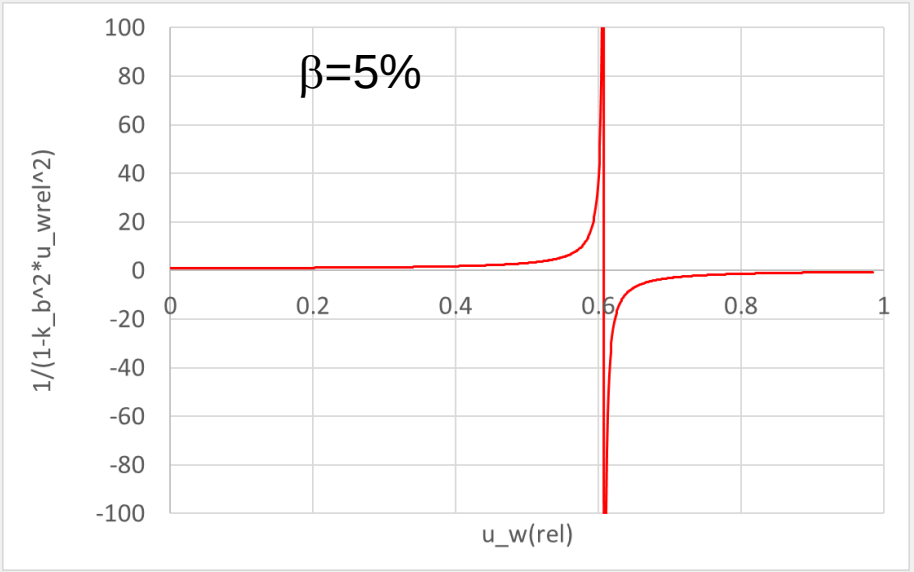
-*In situ* gamma ray spectrometry measurements
-Laboratory measurements in the low energy region (matrix effect) and correction(s) using ET (no Cutshall correction) for matrices, like sediments, with ’unknown’ composition
-…
➔Large uncertainty in k_{ET}

© Swedish Defence Research Agency

The conversion factor and ISO 11929

- For very large uncertainties in the conversion factor the denominator eventually change sign and becomes negative. For β=5% this happens when $u_{w.rel}$≈60%.
- This results in negative detecion limits for $u_{w.rel}$>60% (and ∞ and –∞ when $u_{w.rel}$≈60%)
- Why?

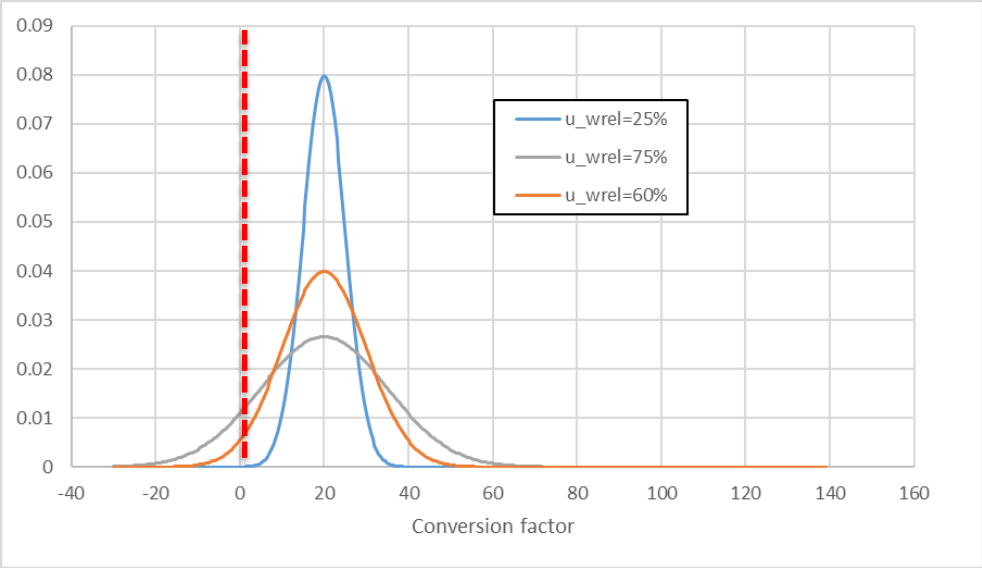
$$MDA_{ISO11929} = \frac{MDA_{Currie}}{1 - k_{1-\beta}^2 u_{rel}(w)^2}$$

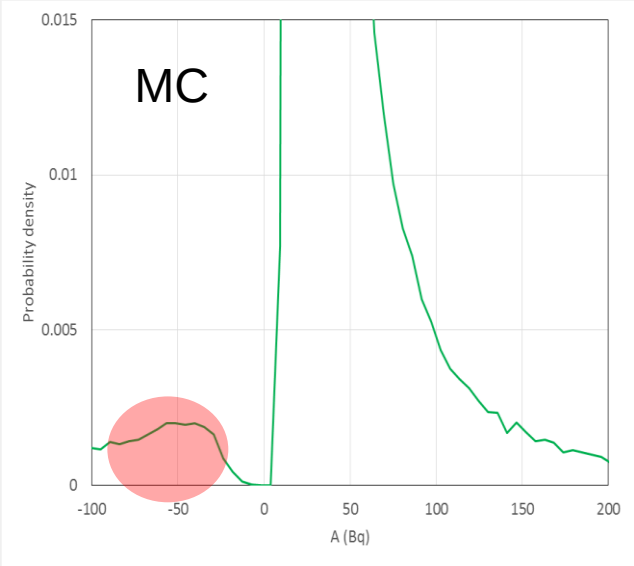


© Swedish Defence Research Agency

Why?

- When the relative uncertainty of the conversion factor becomes large, a large part of its lower tail will be on the negative side.
- This will result in a fraction of the activity distribution ending up as negative activities. (Activity is a non-negative quantity!)





$u(w)_{rel}$=62%

© Swedish Defence Research Agency

The conversion factor and ISO 11929

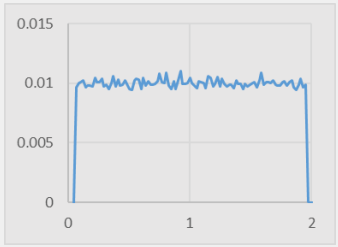
- For *large* uncertainties in *w* and modelling with a normal distribution there will be a large probability for negative *w*
- Activity is a non-negative measurand:
  The input quantities for calculating *w* could not be negative➔Non-physical
  (k_{ET}>0; m>0; t_m>0; I_γ>0; ε>0)
- Choose to model, here k_{ET}, with a distribution not resulting in negative values:
  -Rectangular; Triangular;…
  ➔MC-methods and restricting *w* to only positive values (see ISO 11929-2:2019 …Part 2: Advanced applications)

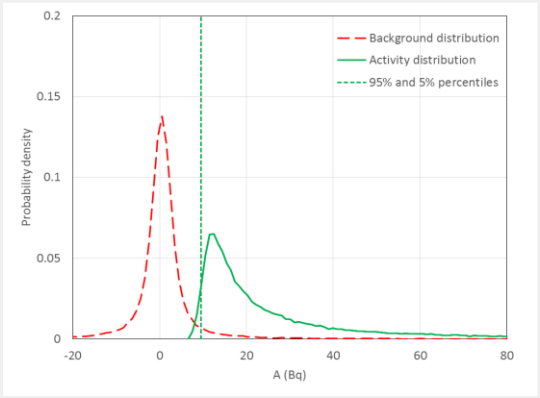
© Swedish Defence Research Agency

The conversion factor and ISO 11929

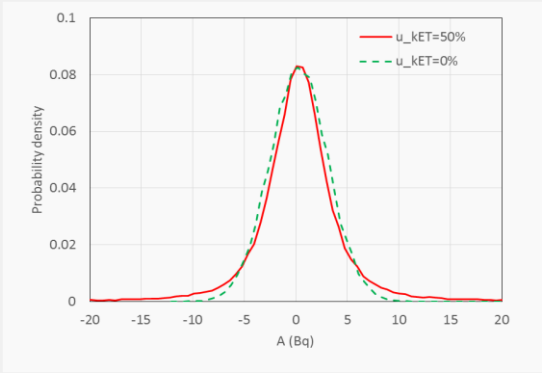
Two observations:

- MC calculation of critical limits:
  Restrict *w* to only positive values. Here we model k_{ET} with a wide rectangular distribution (*a*=0.01; b=1.99)





- GUMUF results in no influence of *u*(*w*) on the decision threshold, but using MC we can see an influence in the tails



© Swedish Defence Research Agency

Relevant literature

- L.A. Currie, *Limits for qualitative detection and quantitative determination. Application to radiochemistry*, Anal. Chem., 40, 586, 1968
- J.C. Lochamy, *The minimum-detectable-activity concept*, NBS SP456, 1976
  (consider when $t_{sample} \neq t_{BG}$)
- ISO 11929-1:2019:
  Determination of the characteristic limits (decision threshold, detection limit and limits of the coverage interval) for measurements of ionizing radiation – Fundamentals and application – Part 1: Elementary applications
- ISO 11929-2:2019:
  Determination of the characteristics limits (decision threshold, detection limit and limits of the coverage interval) for measurements of ionizing radiation – Fundamentals and application – Part 2: Advanced applications

© Swedish Defence Research Agency

Summary

- MC is a better choice for uncertainty calculations when the criteria for GUMUF do not hold
- Critical limits are important since it tells if a method would be fit-for-purpose
- The ISO 11929:2019 requires that the uncertainty of the conversion factor is considered when determining crtitical limits
- The ISO 11929:2019 will be fit-for-purpose for most laboratory applications
- Very large uncertainties in the conversion factor will eventually result in negative detection limits
- This might be solved using MCM and modelling the input quantities considering only positive values. Again, *A* is a non-negative quantity

© Swedish Defence Research Agency

Thank you!

henrik.ramebeck@foi.se

© Swedish Defence Research Agency