Recent work on detection limits in gamma-ray spectrometry

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Outline

- Why is critical limits, *e.g.* detection limits, important?
- I: ”Optimization” of filling height
- II: Critical limits: GUMUF vs. MC
- III: A MC method for low-level measurements: comparison of MC with the expressions given by Currie
- Summary

Why is critical limits important?

- Decide if an analyte is present, or not, in a particular ’sample’ (sample in a wide definition…) based on given inputs (α, β, modelling of input quantities)
- This decision should be on a statistical basis, and not *ad hoc*, which might result in wrong decisions:
  Sometimes used for further calculations/estimations➔error in these calculations/estimations!
- Sometimes a common misunderstanding: ’*Detection limit is about measuring ’as low as possible’!’*
  It is not! It is a performance characteristic of a fixed measurement method, although it might be a $f(t,x)$, and a part in the process to decide the presence or not of a specific analyte

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Part I. Optimal filling height

- *Ramebäck and Vidmar: JRNC, 326, 343, 2020*
- Is it always favorable to measure as large samples as possible in gamma ray spectrometry?
- Or might it be counter-productive with respect to MDA?
- **Hypothesis:** For any fix container diameter there might be an optimum amount of sample (sample height) in order to reach the lowest MDA for some specific sample types if such a sample has a high degree of attenuation in combination with a content of *e.g.* natural radioactivity generating a background to the sample spectrum

…Optimal filling height

- *Sample types*:
  -Acified MQ-water samples (density: 1.0 g/cm^3)
  -Zircon sand (high density: 2.7 g/cm^3): activities from the natural decay series
- *Container*:
  -Fixed diameter: ~70 mm
  -Filling height: 10-60 mm (10 mm increment)

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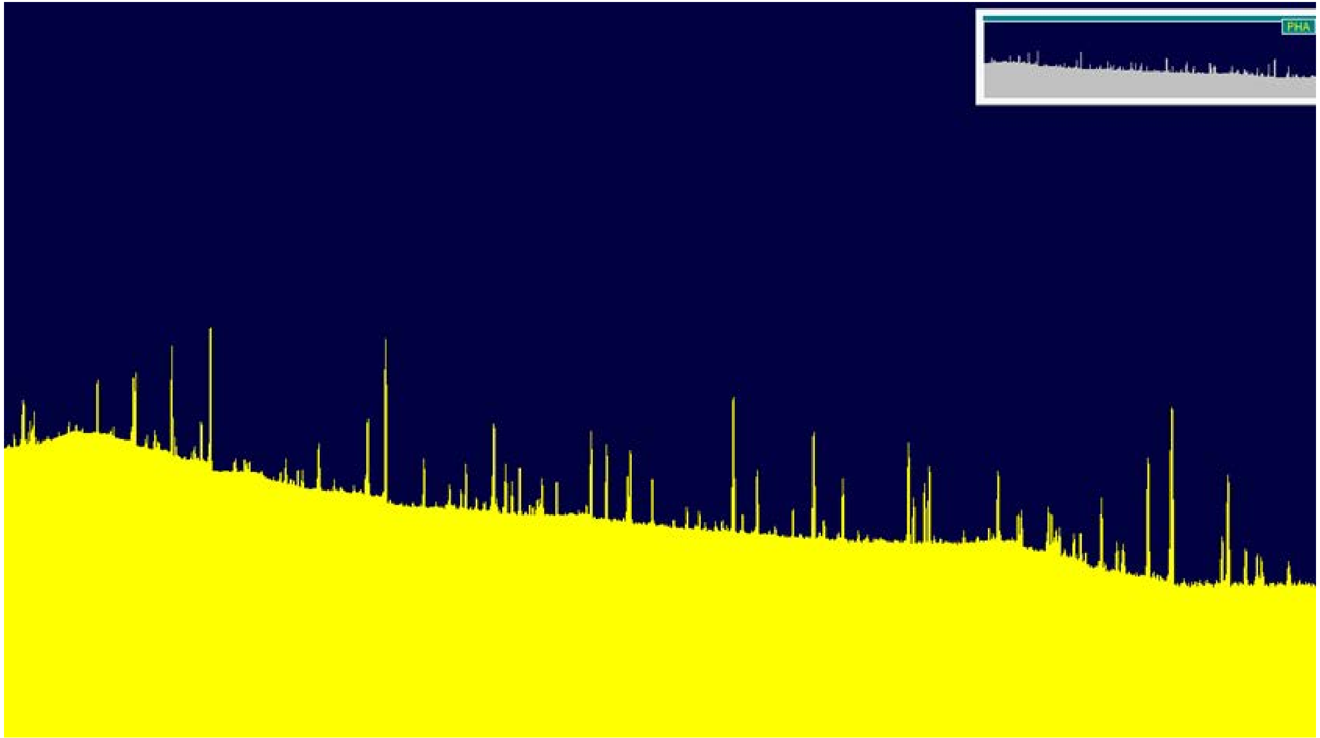
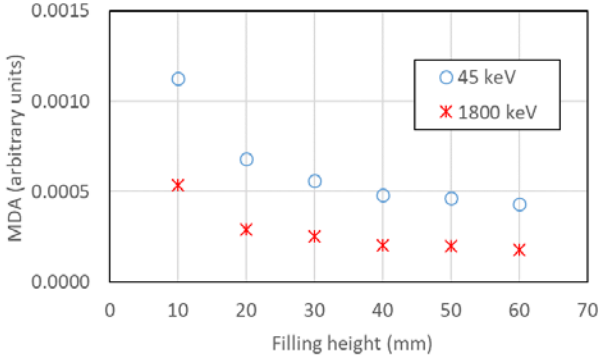
Background: 22 h, zircon sand, 60 mm filling height.



…Optimal filling height: Results

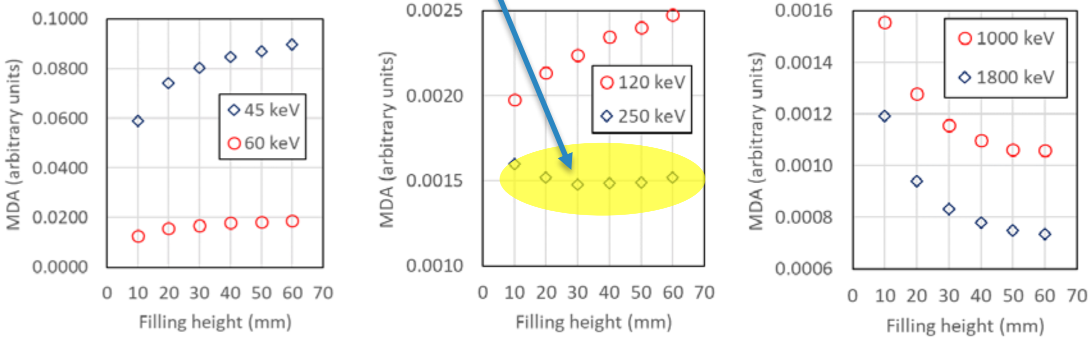
-Water sample:



No limitation in the amount of sample!

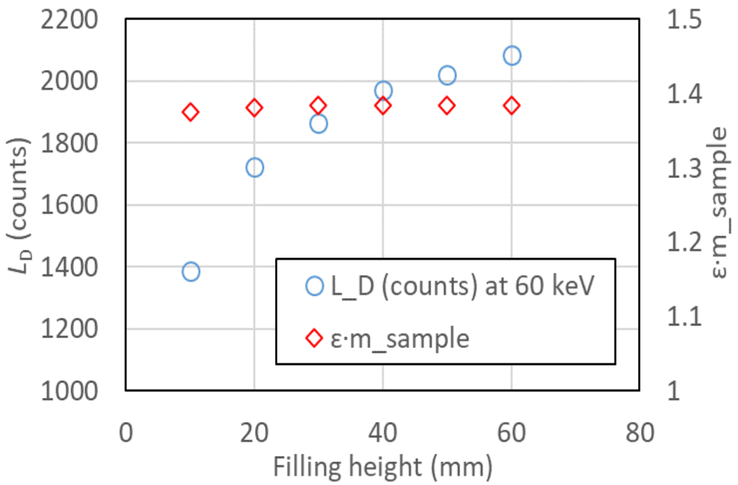
-Zircon sand:

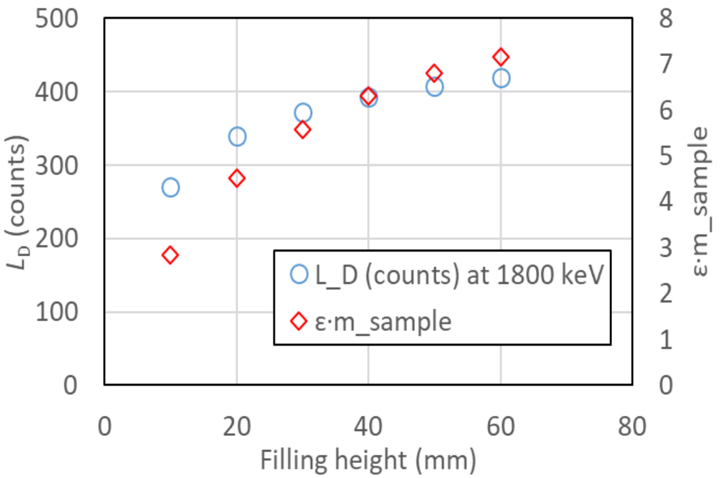
There is accually an optimum, in this case at about H=30 mm for *Eγ* of about 250 keV resulting in a minimum in MDA!



…Optimal filling height: results

- There is a 'competition' between the increase in background due to a higher amount of sample and the increase in $\varepsilon \cdot a$ (efficiency times amount of sample)





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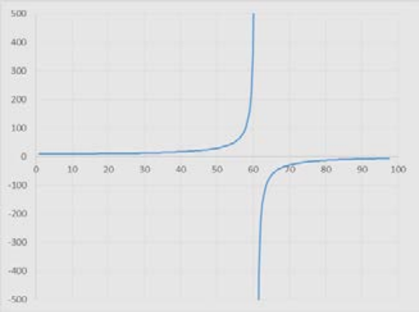
Part II. GUMUF vs. MC for calculation of critical limits

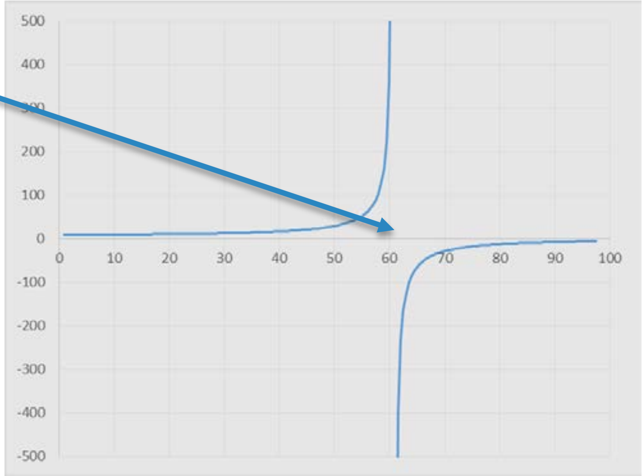
- *Ramebäck, Persson, Ekberg, Lindgren, Bruggeman: ARI, 156, 2020 (108949)*
- Using the GUMUF for calculation of detection limits (ISO 11929:2019) result in a singularity for high values of the uncertainty in the conversion factor for calculation of MDA from an instrumental signal. For β=5% this occurs at u_{relw}≈61%.
- *When such high uncertainties in a conversion factor?*



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

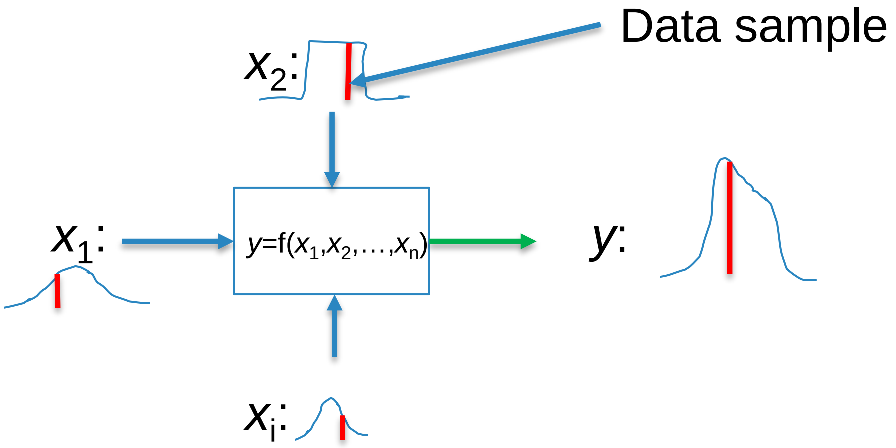
Monte Carlo for critical limits

- ISO 11929:
  Calculations of critical limits should take all uncertainties into account, not only the uncertainty in instrumental signals:
  We go from the '*signal domain*' to the '*activity domain*'. This conversion has uncertainties
- **Consequence:** a singularity when β=5% at about 61% uncertainty in the conversion factor ($w=1/(t \cdot I_g \cdot \varepsilon \cdot \boldsymbol{k_{ET}})$ applying the GUMUF
- Around (below) u_{wrel}=61% MDA will become eternity, and (above) –eternity, thereafter negative MDA!
- **Reason:** a large uncertainty in, here k_{ET}, results in 'sampling' negative values since a normal distribution is assumed in GUMUF
- **Solution:** restrict k_{ET} to only positive values (a negative efficiency is non-physical)



MC calculation: Principle

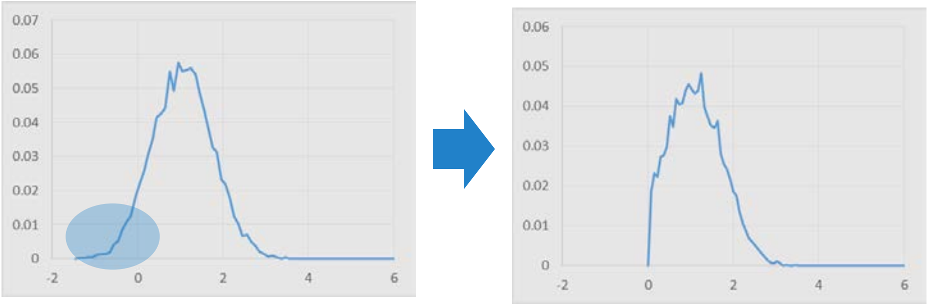
Propagation of distributions (*cf.* propagation of uncertainties):



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

A solution: Truncate the PDF of *e.g.* k_{ET}

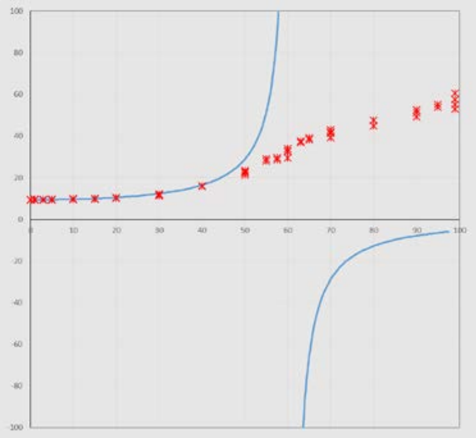
Truncate the PDF for the efficiency ’just above’ zero→not sampling negative values…
Justification: We can not have negative efficiencies (it would imply photon emissions from the detector)



Here, a truncated normal distribution is shown.
It was also modelled as rectangular, triangular and lognormal distributions

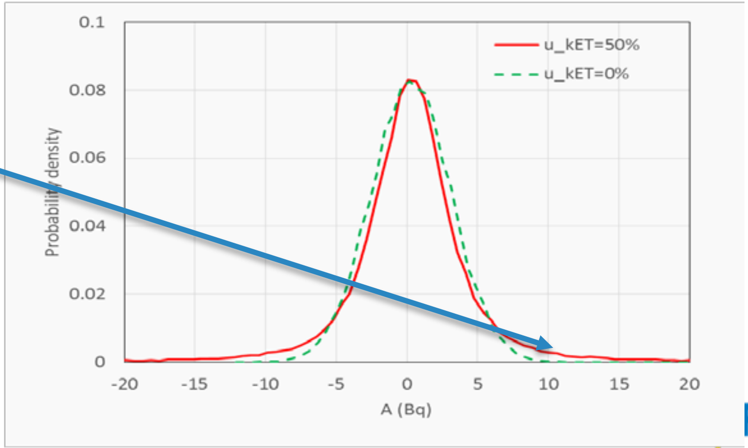
Result

Again, using GUMUF results in *e.g.* negative MDA when the relative uncertainty of the conversion factor is too high.



Another result: Using GUMUF the uncertainty in the conversion factor will not have an effect on the decision threshold *y** although a transformation from the signal domain to the activity domain is done.

With MC we see that it has an effect (it has an effect of the PDF of the 'zero' in the activity domain and will therefore have an influence on the (1-α)-percentile:



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

Part III: Low-level measurements

- *Rameback, Persson, Ekberg, Vesterlund, Bruggeman: ARI, 178, 2021 (109959)*
- How well do expressions for calculations of critical limits work for cases of low-level measurements (few events)?
- Most (all?) ’equations’ assumes normal distribution
- Here, MC calculations were done for a Poission distributed parameter
- Results here given for comparison with Currie’s expressions (the article includes some more expressions)

Low-level measurements

- ISO 11929 and Currie:
  - $\alpha = P\left(N > L_c \middle| \tilde{N} = 0\right)$, *i.e.* detected when $N > L_c$
- ISO 11929:
  - $\beta = P\left(N < L_c \middle| \tilde{N} = L_d\right)$
- Currie:
  - $\beta = P\left(N \leq L_c \middle| \tilde{N} = L_d\right)$
- ISO 11929: L_c not included in either false positives or false negatives!

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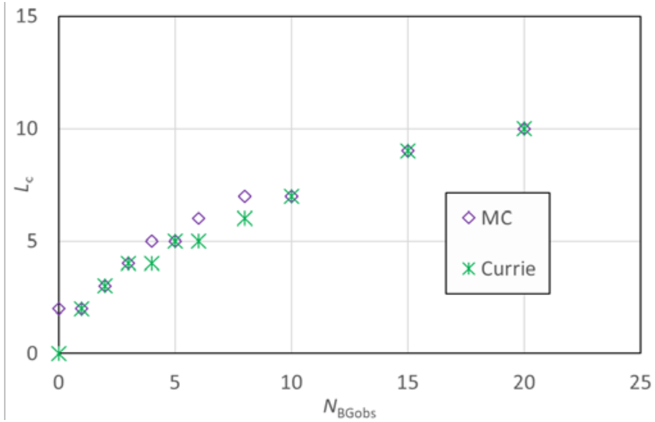
Low-level measurements

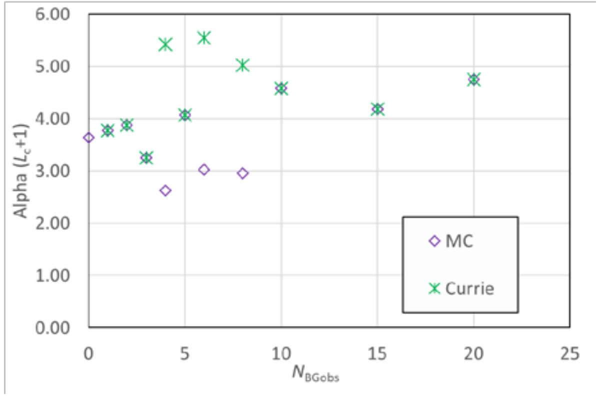
- In (at least) one publication the definition of a false positive was given as
  -$\alpha = P\left(N \geq L_c \middle| \widetilde{N} = 0\right)$, *i.e.* detected when $N \geq L_c$
  This resulted in a too high false positive rate, and the conclusion was that Currie's equation do not give proper results for L_c.
- Observe: In the publication another definition of 'detected' as compared to Currie was used (**≥ *vs.* >**)!
- There will be an impact on the calculated false positive rate if L_c is included or not in α!

Low-level measurements

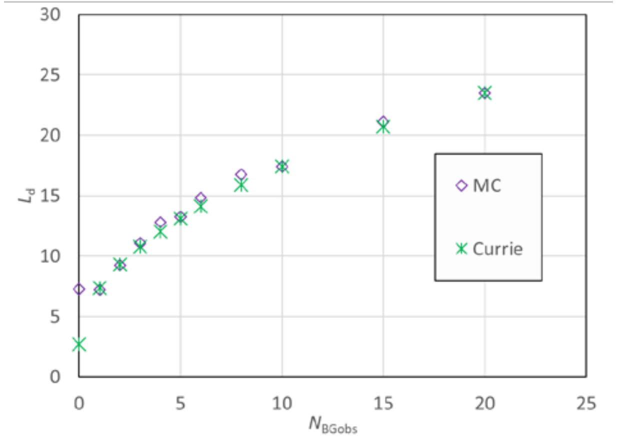
- MC calculations were used for sampling from the Poisson distribution (Binomial distribution for a low probaility and large number of tries➔Poisson distribution)
- α set to a maximum of 5%
- β set to 5% (the parameter λ is in the Poisson distribution a continous variable)

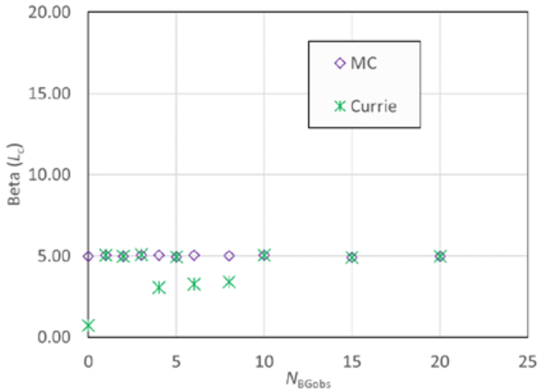
-Decisions threshold:





-Detection limit:





Summary

- For some sample matrices and in some energy region there exist a filling height resulting in a lowest MDA
- Negative detection limits when GUMUF is applied can be avoided using MC calculation in combination with restricting the input quantities for calculation of the conversion factor *w* to only positive values (negative ones is non-physical: *t* positive; I_γ positive; ε positive; k_{ET} positive;…)
- Critical limits in low-level measurements as calculated using expressions given by Currie results in good agreement compared to a MC method also for a few events in a background