LSC 2017

Advances in Liquid Scintillation Spectrometry

With a little bit of effort, manufacturers could make better LS counters, even suitable for radionuclide metrology!

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Background

During an international specialists' meeting on LS techniques in radionuclide metrology*, the attendee reached the conclusion that there is no longer a commercial LS counter available on the market which could fulfill the requirements of a metrological instrument for the application of the CIEMAT/NIST efficiency-tracing method

- Two main reasons:
  - The international market of LS counters is small (about 200 counters per year ?) and companies making good instruments (Beckman, Wallac,) disappeared during the last twenty years. There is no more real concurrence on the market, so no strong motivation to drive the improvement of the quality of the counters

  - Most of the LS counters are oriented towards the measurement of low activities, where some features (e.g. linearity) are not so critical and where the measurement uncertainty requirement is not so strict

* Liquid Scintillation Working Group of the International Committee for Radionuclide Metrology

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What is metrology?

Traditional definition (BIPM):

The science of measurement, embracing both experimental and theoretical determinations at any level of uncertainty in any field of science and technology.

In LSC, this supposes:

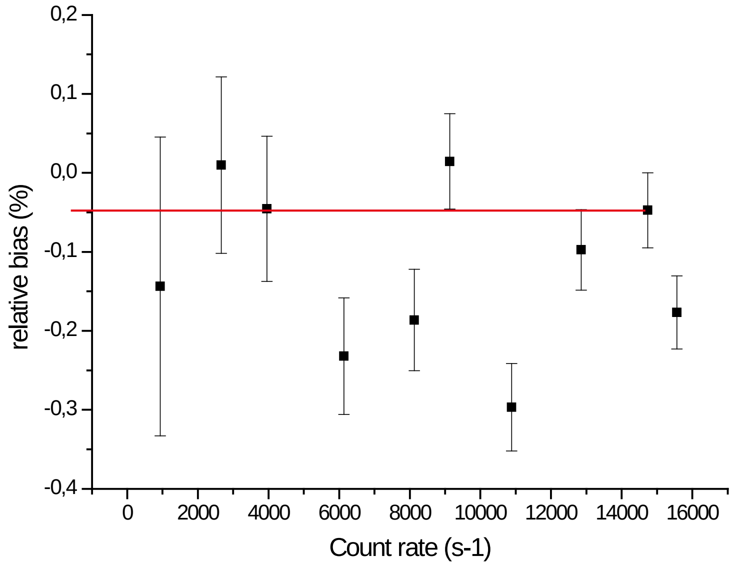
- The use of a ”good” instrument, where the variability of the measured quantity is under statistical control (i.e. all variability effects are identified and accounted for by calculable uncertainties)
- A sound determination of the measurement uncertainties
- The use of a procedure to determine the detection efficiency (quenching curve, internal standardization, tracer method (CIEMAT/NIST,…)

This latter point is outside of the scope of this talk and we will focus on the two first points

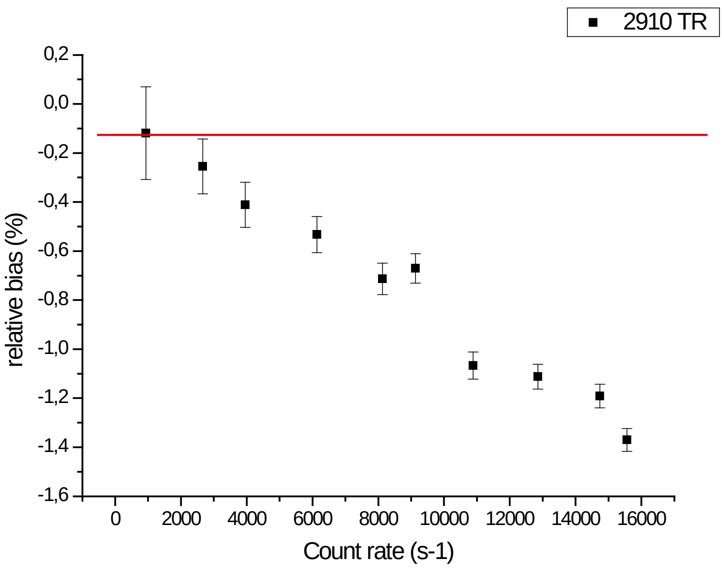
Observed drawbacks of commercial LS counters

- Linearity problems
- Quenching index measurement
- Negative effects of the active guard
- Differential non-linearity
- Unknown dead-time system
- Unknown PMT's threshold levels
- Long-term drift of the counter
- Poor accuracy of the starting time of the measurement
- No traceability of the reference clock of the system
- Black box effects (for those who want to make a sound uncertainty budget and thus to understand how the system is working)

Linearity (BIPM counters)



Beckman LS6000 TA

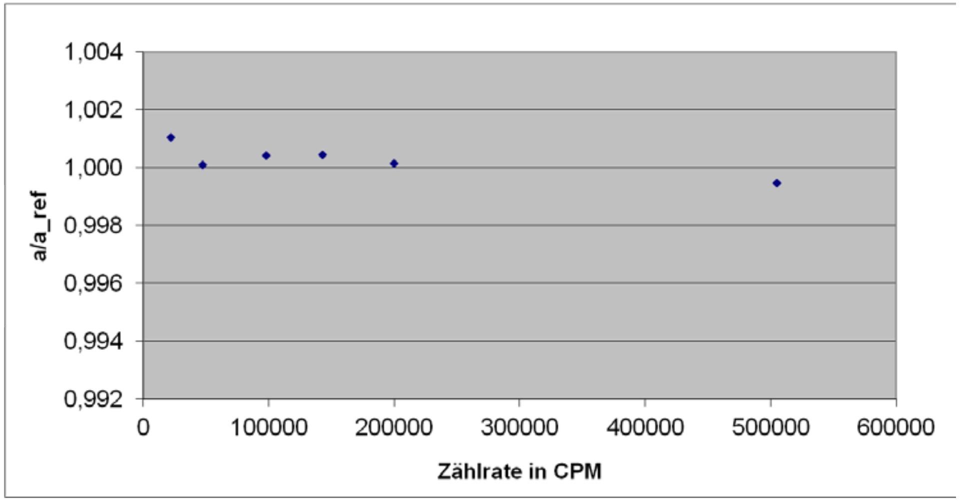


TriCarb 2910 TR

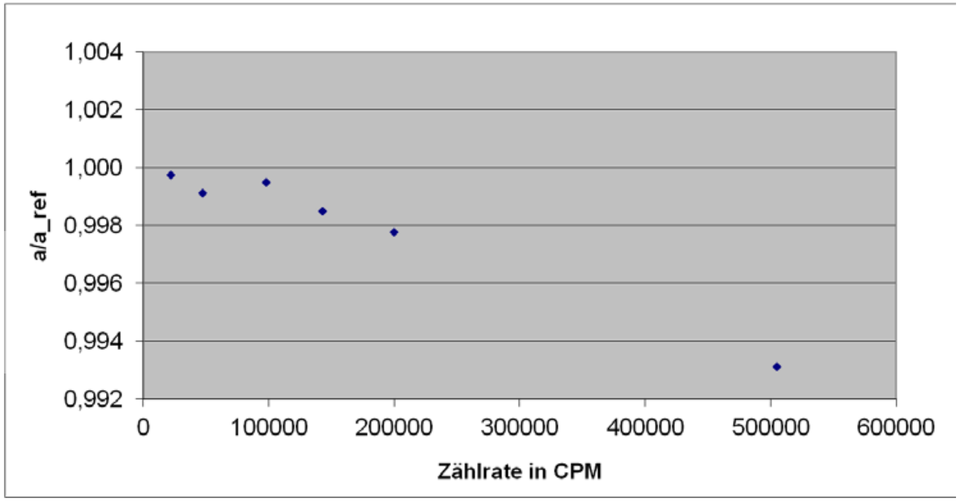
The venerable Beckman counter (more than 20 years old) has a reliable behavior…

Examples counters at PTB

TriCarb 2800 TR (2006)



TriCarb 2810 TR (2009)



Example of Hidex SL 300 counter*

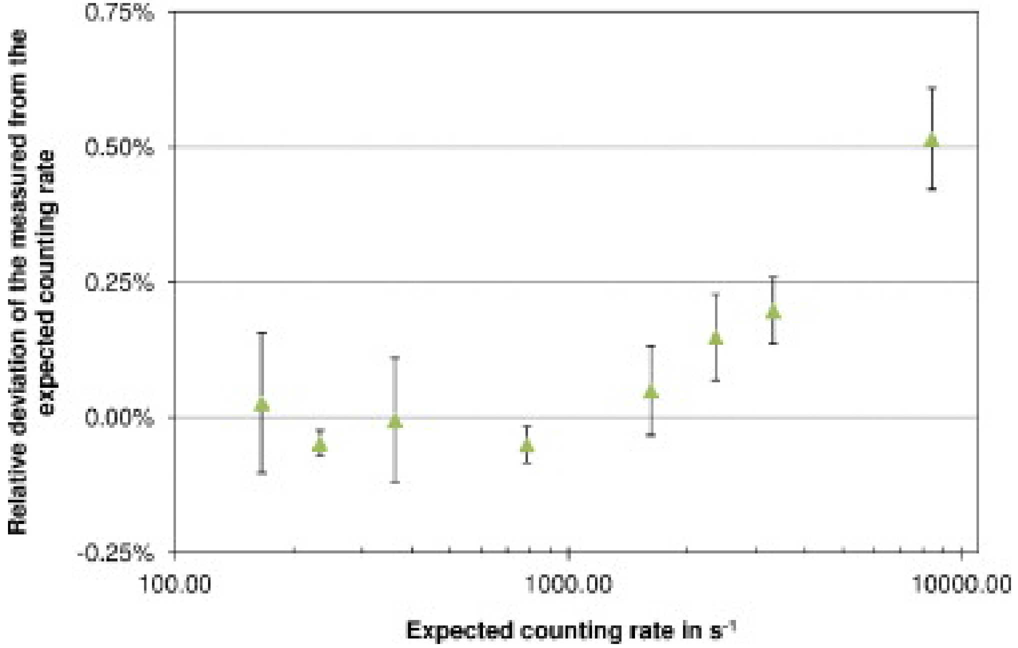


Fig. 1. System linearity check—deviations of the measured counting rates from the expected ones in the measurement of 241Am. The bars indicate the standard deviation of three or more repetition measurements.

* Karsten Kossert, Marco Capogni, Ole J. Nähle, **Bilateral comparison between PTB and ENEA to check the performance of a commercial TDCR system for activity measurements,** Applied Radiation and Isotopes, Volume 93, 2014, 38–44,

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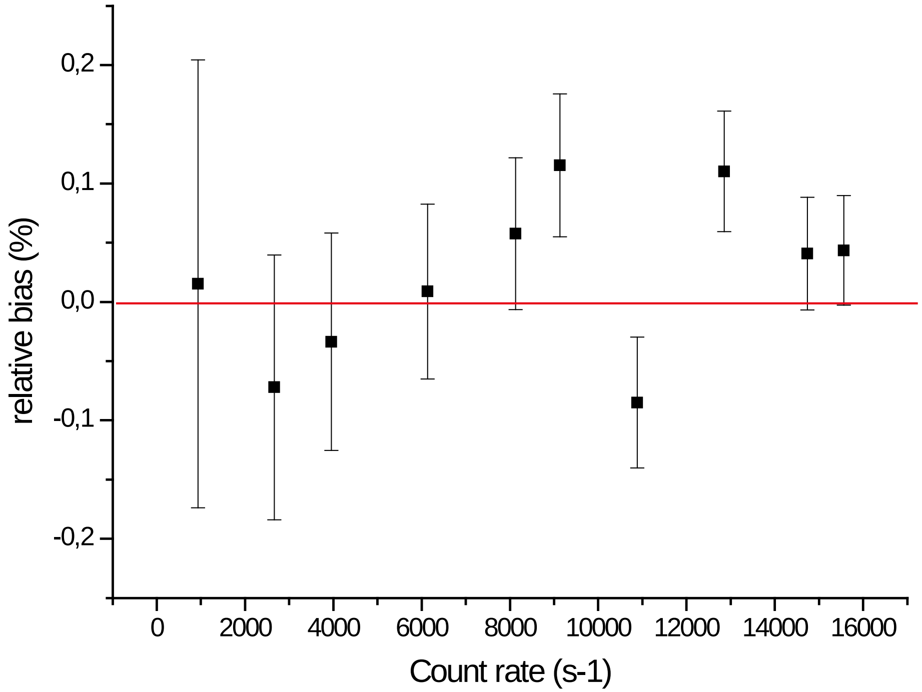
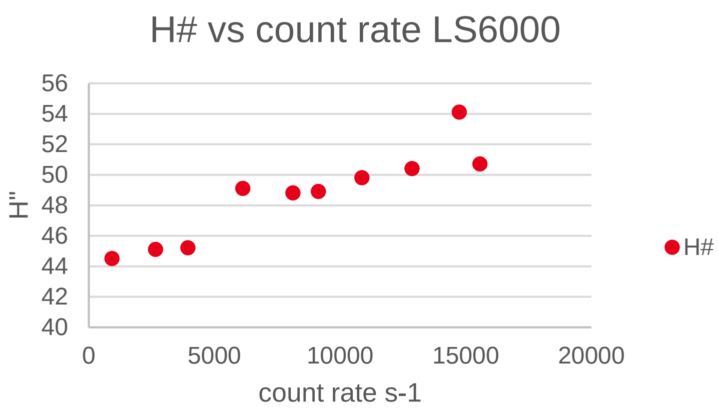
LNHB RCTD1 counter (with MAC3 unit)

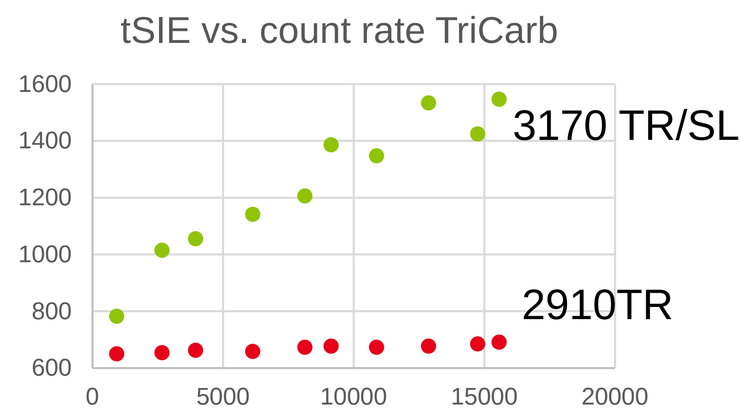


So, with a good design, it is possible to make a linear LS counter!

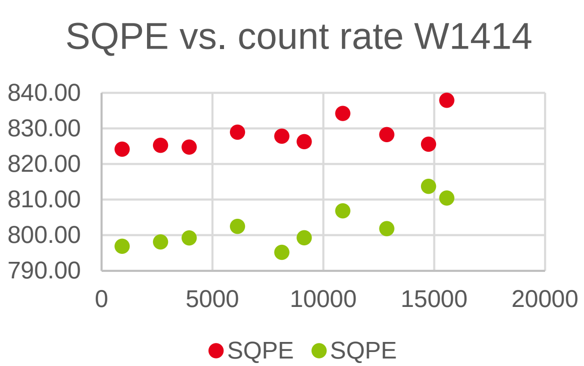
Quenching index ($^{90}Sr/Y$ sources with increasing activity but same quenching)



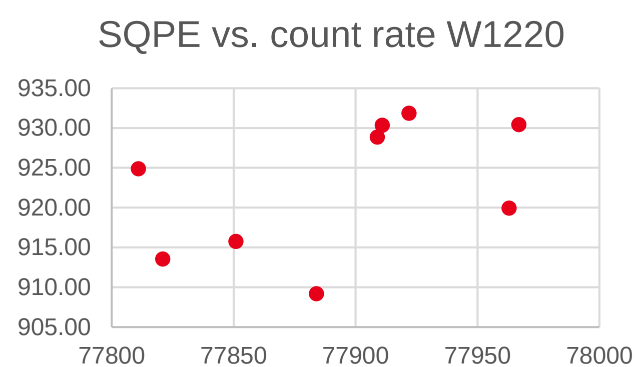
Beckman



Perkin Elmer TriCarb



Wallac (2 identical (?) counters)



Wallac Quantulus

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Active guard

Real anti-coincidence system with plastic or liquid scintillators:

- The active guard is inside the lead-shielding
- Photon emission from the source decreases the detection efficiency (γ,X, bremsstrahlung)
- This causes a bias in the measurement of some radionuclides (e.g. β^+)
- This could be overcome if the guard signals can be identified (original Quantulus) or if the guard can be disconnected

Pseudo-active guard with a BGO crystal

- High sensitivity to γ (a γ peak is even visible in the spectrum)
- Impossible to calculate the detection efficiency
- High bias for quenching index determination
- Quenching index dependent of temperature

Personal view:
No serious metrological measurement can be achieved with LS counters with BGO

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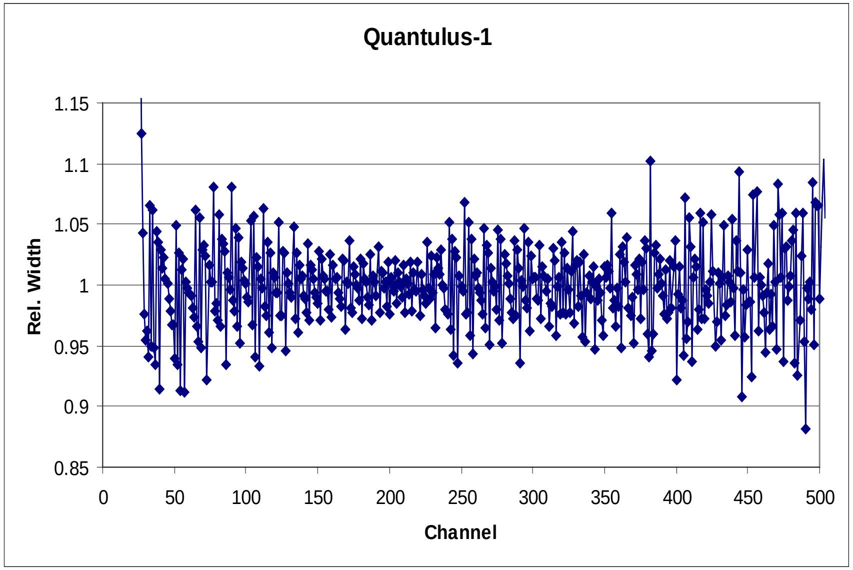
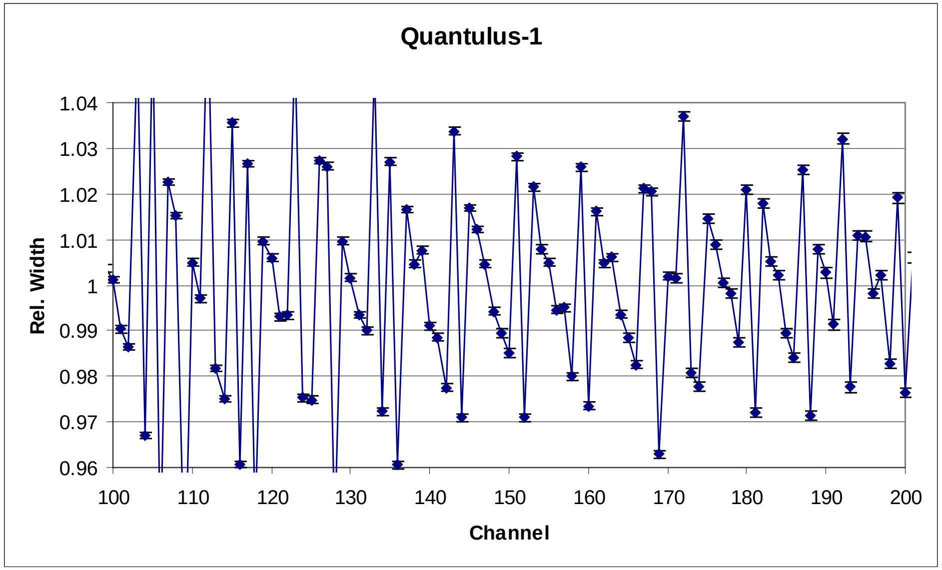
Differential non-linearity (DNL)

Imperfection of the ADC → non uniform width of LSC channels

No problem for the total count, but distortion of the shape of the spectrum

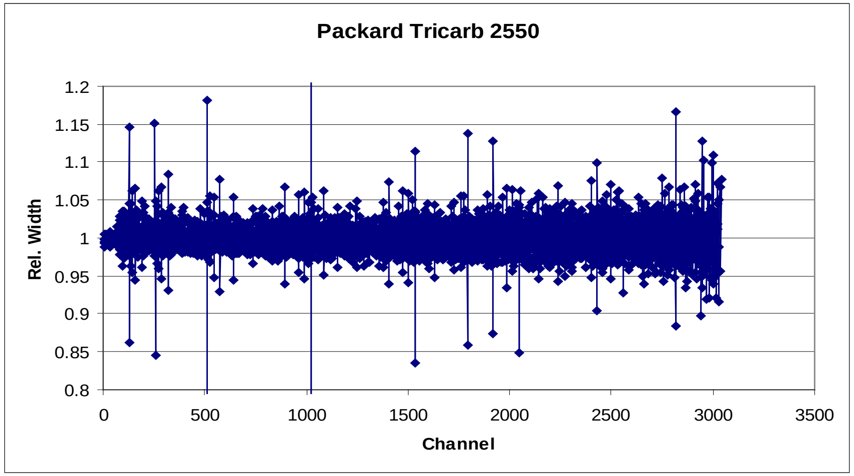


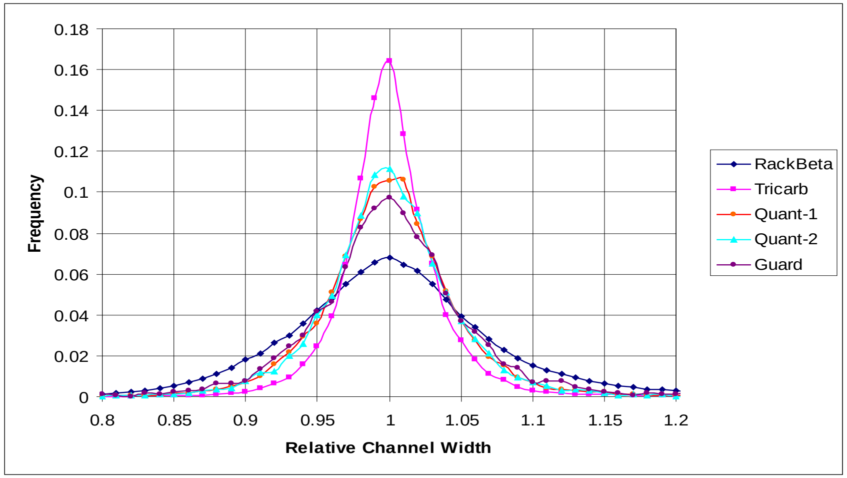


Example of Quantulus (original) counter

Differential non-linearity (continued)

Other examples. Is the DNL of new counters lower?





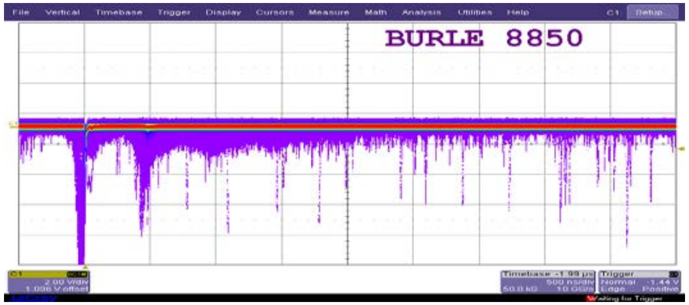
Observed in all counters, but can be compensated by software

Thanks to L. Tsankov, Department of Nuclear Engineering, University of Sofia

K. Mitev, G. Gerganov, T. Boshkova, L. Tsankov, S. Georgiev, I. Dimitrova, “Development of numerical algorithms for comparison of liquid scintillation spectra for determination of the activity of beta- emitting nuclides in the presence of other nuclides in the LS sample”, Final report on contract No 220000003 between Kozlodui Nuclear Power Plant and Sofia University “St. Kliment Ohridski”, Sofia, 2012 (in Bulgarian).

Dead-time system

- In LSC, the detected pulses can be followed by after-pulses (delayed fluorescence, after-pulses of the PMT's…)
- After-pulses can last up to a few tens of µs
- Self-correlation of the output of a PMT



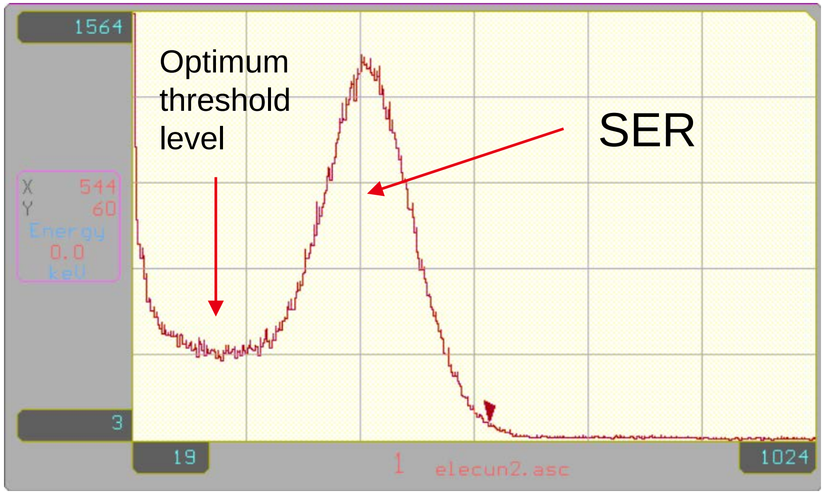
- It is essential to impose a dead-time to avoid the effects of after-pulses
- An extending-type dead-time system offers the best protection against after-pulses; its duration can be accurately measured using a live-time clock
- For the measurement of some radionuclides (e.g. ^{222}Rn, with short half-life daughters) it is necessary to know the duration and the type of dead-time to account for counting losses

<u>But:</u>

- Commercial LS counters are black boxes (no information on the dead-time circuit)
- Some features (e.g. delay before burst) complicates the behavior of the counter
- Unfortunately dead-time properties can only be known by reverse engineering (e.g. measurement of ^{222}Rn source)

PMT threshold levels

- The CIEMAT/NIST efficiency tracing method supposes that the PMTs can detect single photons (hypothesis of the model)
- This supposes an adjustment of the thresholds in the valley before the single electron response (SER)

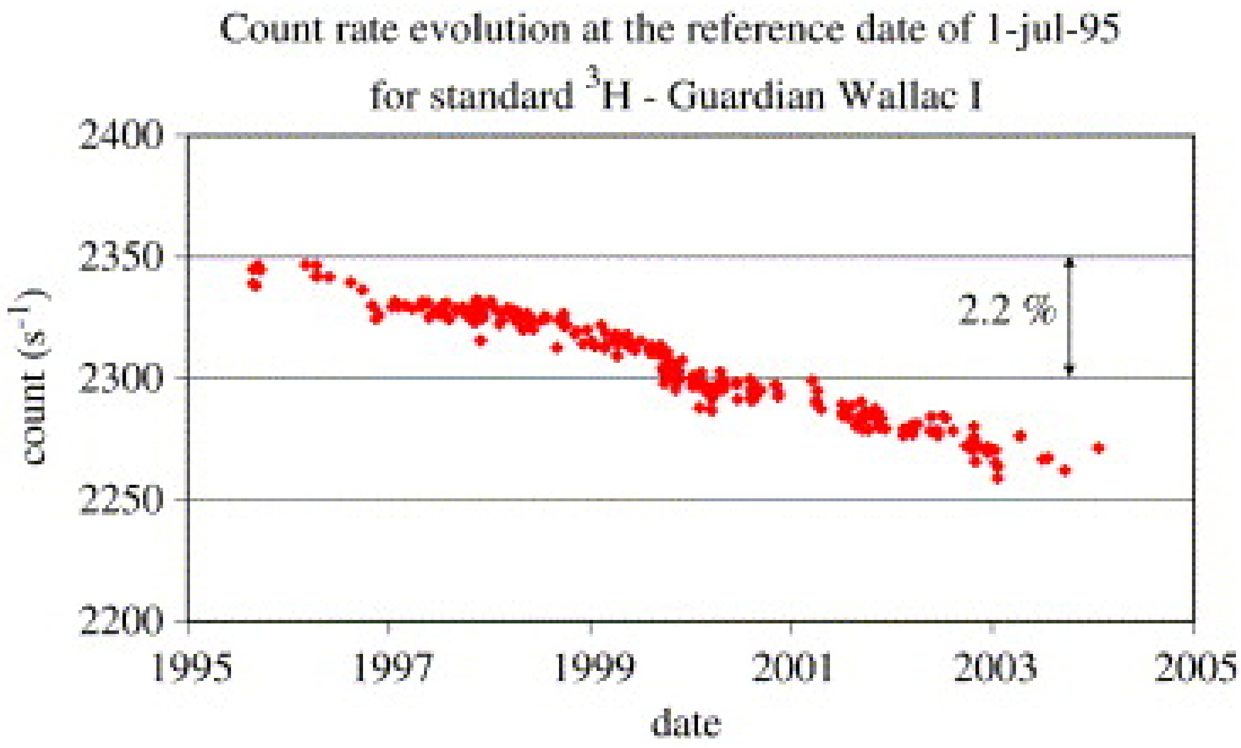


But: to my knowledge, no commercial LS counter gives information on the threshold adjustment with reference to the SER position

There is some evidence that this threshold (or the gain of the PMTs) changes with time…

Long-term stability of the counter

Example of Wallac 1414, but similar behavior has been observed with other counters



F. Jaubert, I. Tartès, P. Cassette, **Quality control of liquid scintillation counters,** Applied Radiation and Isotopes, Volume 64, Issues 10–11, 2006, 1163–1170

Uncertainty in the measurement starting time

- The activity of a radionuclide must be given for a reference date and time
- This time must be traceable to UTC (or to local time traceable to UTC)
- In some counters the measurement starting time is only known with a resolution of one minute
- This can cause a significant bias in the measurement of short half-life radionuclides

Example of measurement bias *vs.* bias on the measurement time

Radionuclide	Half-life	Relative bias for a timing bias of:			
		5 s	15 s	30 s	45 s
^{18}F	1,8288 h	0.05%	0.16%	0.32%	0.47%
^{11}C	20,361 min	0.28%	0.85%	1.7%	2.5%
^{15}O	2,041 min	2.8%	8.1%	15.6%	22.5%

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Traceability of the counter internal clock

- The output of an LSC counter is a counting rate (unit: s^{-1})
- The s is derived from an internal clock (generally a quartz oscillator)
- The bias (or uncertainty) of the frequency of this clock directly influences the uncertainty of the measured counting rate
- Even if most quartz oscillators can be considered as stable and accurate, there is a need for formal traceability of the frequency of this oscillator
- This can be easily achieved by providing access to the frequency of this clock (or multiple or division of this frequency)

Improvements for metrological applications

Key points for the improvement of metrological properties:

- Accuracy and resolution of the measurement date and time reported by the counter; traceability to UTC
- Accuracy of the live-time determination
- Linearity of the counter
- Compensation of the differential non-linearity
- Determination and control of the threshold of the PMTs discriminator levels
- Optimization of the dead-time unit
- Definition of a robust quenching level indicator
- Traceability of the reference clock used to define the measurement live-time
- Possibility to disconnect the active guard (or any background reduction system)
- Possibility to record and save the spectrum obtained with the external source to determine the quenching index (to give the possibility to check its value)

Conclusions

Even if "hardcore metrology" use of LS counters is marginal within the LS community:

- Instrument manufacturers must not underestimate the influence of the National Metrology Institutes (NMI) on all the laboratories using LS techniques, especially when these laboratories are accredited

- Most of the problems observed in actual commercial counters can be solved using simple and cheap means, by following our suggested improvements

- The metrological community is always available to help the manufacturers to improve their instruments (if necessary with confidential agreements)

- Many NMIs will be forced to replace their existing LSC counters in the near future…

So, do not miss this opportunity to sell good instruments!

Acknowledgments

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Henri Becquerel

σας ευχαριστώ

gracias

благодаря

ngiyabonga

Dankie

obrigado

grazie

köszönöm

THANK YOU

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Merci

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