Safeguards: Modelling of the Detection and Characterization of Nuclear Materials

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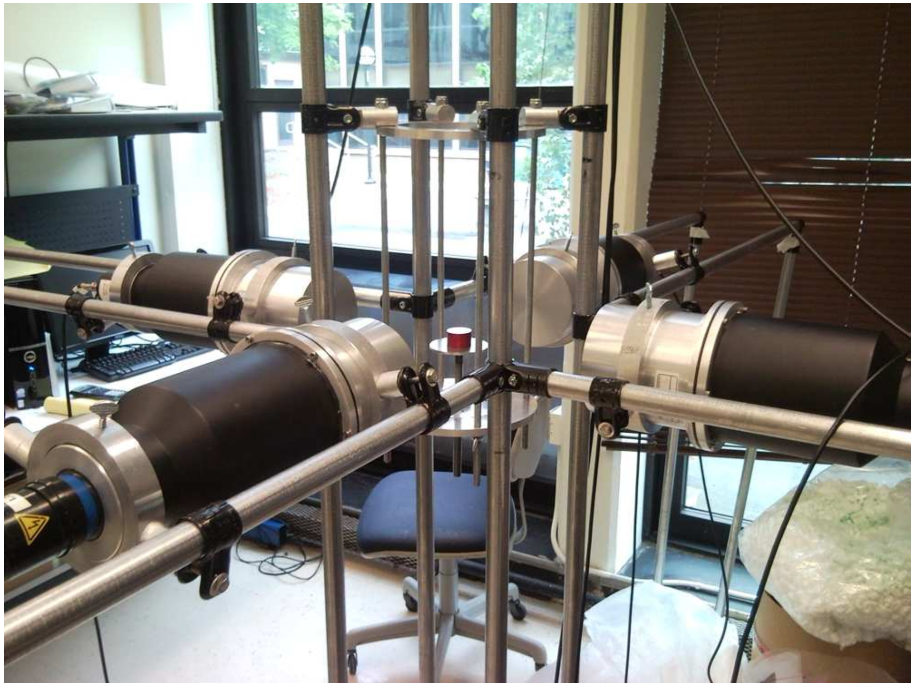


Corrigenda

Status of PAPER IV should now read *Nuclear Instruments & Methods A*, **615**, 62–69 (2010).

Status of PAPER VII should now read *accepted for publication in Nuclear Instruments & Methods A*, (2010).
DOI: 10.1016/j.nima.2010.02.119

Part 1, Particle Number Distribution

This thesis is an introduction to and a summary of the work published in the following papers:

- ✔ PAPER I

  A. Enqvist, I. Pázsit and S.A. Pozzi, "The Number Distribution of Neutrons and Gamma Photons Generated in a Multiplying Sample"
  Nuclear Instruments & Methods A, **566**, 598–608 (2006).
- ✔ PAPER II

  A. Enqvist, I. Pázsit and S.A. Pozzi, "The Detection Statistics of Neutrons and Photons Emitted from a Fissile Sample"
  Nuclear Instruments & Methods A, **607**, 451–457 (2009).

Part 2, Multiplicity Theory

- ✔ PAPER III

  I. Pázsit, A. Enqvist and L. Pál, "A note on the multiplicity expressions in nuclear safeguards."
  Nuclear Instruments & Methods A, **603**, 541–544 (2009).

- ✔ PAPER IV

  A. Enqvist, I. Pázsit and S. Avdic, "Sample Characterization Using Both Neutron and Gamma Multiplicities"
  Nuclear Instruments & Methods A, **615**, 62–69 (2010).

- ✔ PAPER V

  S. Avdic, A. Enqvist and I. Pázsit, "Unfolding sample parameters from neutron and gamma multiplicities using artificial neural networks."
  ESARDA Bulletin, **43**, 21–29 (2009). Invited paper.

Part 3-4, Scintillation Detectors & Correlation Measurements

- ✔ **Paper VI**

  S.A. Pozzi, M. Flaska, A. Enqvist and I. Pázsit, "Monte Carlo and Analytical Models of Neutron Detection with Organic Scintillation Detectors."
  Nuclear Instruments & Methods A, **582**, 629–637 (2007).
- ✔ **Paper VII**

  A. Enqvist and I. Pázsit, "Calculation of the light pulse distributions induced by fast neutrons in organic scintillation detectors."
  Accepted for publication in Nuclear Instruments & Methods A.
- ✔ **Paper VIII**

  A. Enqvist, M. Flaska and S.A. Pozzi, "Measurement of Neutron/gamma-ray Cross-Correlation Functions for the Identification of Nuclear Materials"
  Nuclear Instruments & Methods A, **595**, 426–430 (2008).
- ✔ **Paper IX**

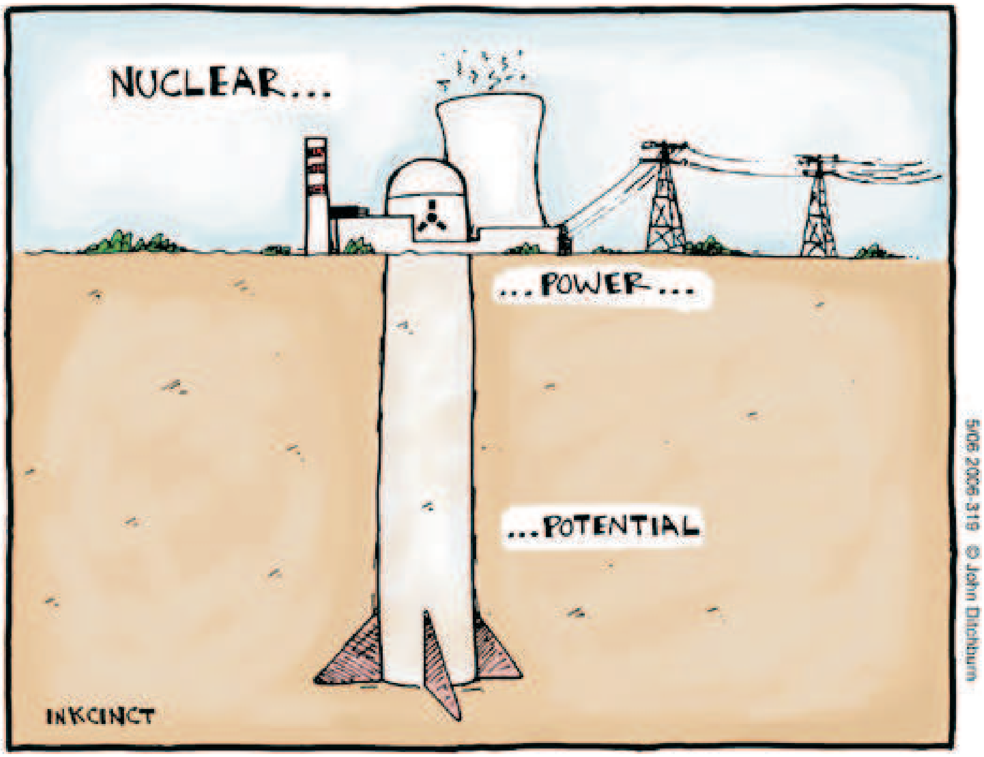
  A. Enqvist, M. Flaska and S.A. Pozzi, "Initial Evaluation for a Combined Neutron and Gamma-ray Multiplicity Counter"
  Submitted to Nuclear Instruments & Methods A.

Safeguards

- ✔ Safeguards is a very broad area ranging from nuclear physics to political regulations.
- ✔ Three main tools are used for describing the physics of nuclear materials:
  - ✘ Analytical models.
  - ✘ Simulations.
  - ✘ Experiments.
- ✔ All of which have strengths and weaknesses.
- ✔ The work presented in the thesis is mostly based on analytical methods for describing the physical processes governing the multiplication and detection of neutrons and gamma rays. The results have often been compared to simulations, and experiments in form of measurements have also been performed to gain additional insight.

Potential Perils?

The motivation of the work is the double-edged sword that nuclear materials presents: It is a way to provide carbon emission-free energy, while at the same time producing waste which needs careful management.



The Task

- ✔ The subject of the thesis is the non-intrusive investigation of nuclear materials.
- ✔ Detection, identification and quantification of nuclear materials from the properties of the detected radiation.

Branching Processes and Fissile Material

In fissile material the life of neutrons is a branching process in which neutrons can generate additional neutrons and reactions.

- ✔ Correlated events.
- ✔ Microscopic physics generating macroscopic effects.

Probability Distributions

- ✔ Neutrons in a fissile sample can undergo a number of different processes.
  - ✘ Fission.
  - ✘ Capture.
  - ✘ (n, xn)-reactions.
  - ✘ Escape the sample.
- ✔ Using the associated reaction probabilities, which depend on sample characteristics, probability balance equations can be formulated.
- ✔ Generating functions and master equations then provide the tools needed to describe the behaviour of particles in the sample.
- ✔ The goal: the probability $p(n)$ of generating n particles when starting with a single particle or a source event such as spontaneous fission with the emission of several neutrons.

The Theory

$$p_1(n) = (1-\mathrm{p})\delta_{n,1} + \mathrm{p}\sum_{k=1}^{\infty} p_i(k) \sum_{\{n_1+n_2+\ldots+n_k=n\}} \prod_{i=1}^{k} p_1(n_i) \quad . \tag{1}$$

converted using generating functions to simple master equations:

$$\begin{aligned} h(z) &= (1-\mathrm{p})z + \mathrm{p}q_i[h(z)], & (2)\\ H(z) &= q_{sf}[h(z)]. & (3) \end{aligned}$$

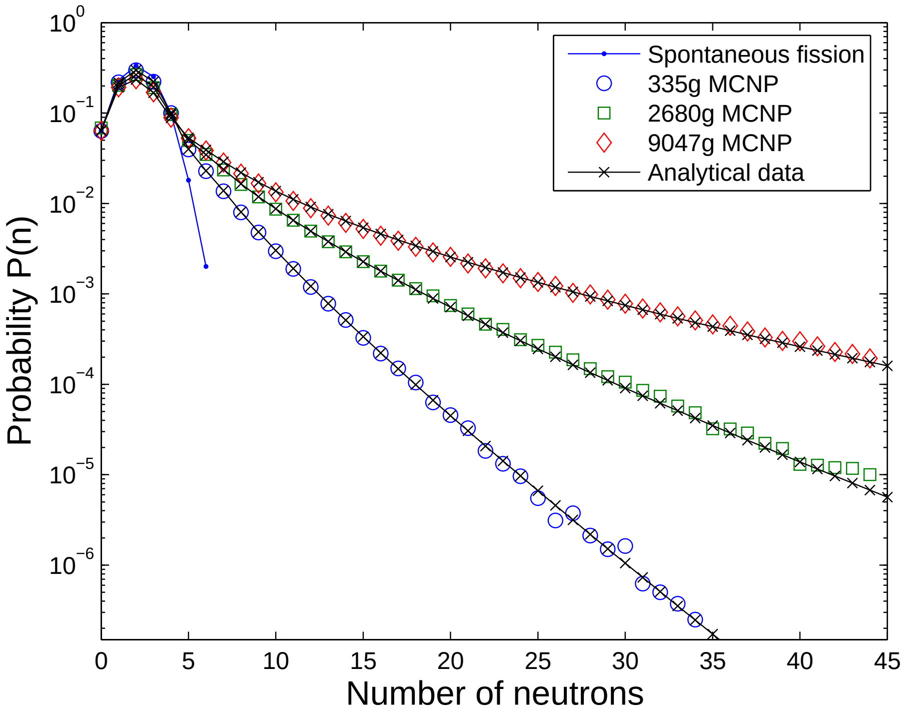
giving probability terms such as:

$$P(2) = \frac{1}{2}\left(\frac{1-p}{1-p\overline{\nu_f}}\right)^2 \left[\overline{\nu_s(\nu_s-1)} + \frac{p}{1-p\overline{\nu_f}}\overline{\nu_s}\,\overline{\nu_f(\nu_f-1)}\right]. \tag{4}$$

through:

$$p_1(n) = \frac{1}{n!}\frac{d^n h(z)}{dz^n}\bigg|_{z=0} \quad \text{and} \quad P(n) = \frac{1}{n!}\frac{d^n H(z)}{dz^n}\bigg|_{z=0}. \tag{5}$$

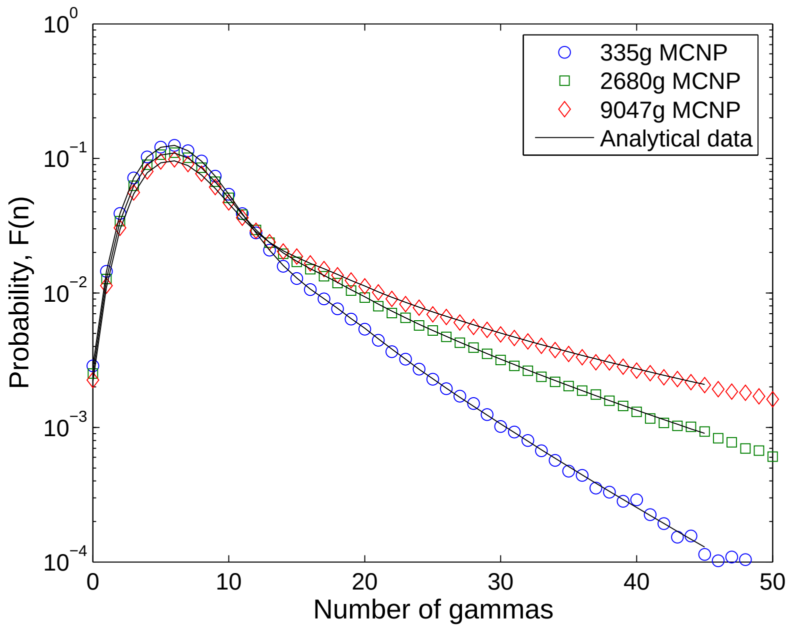
Neutrons



- ✔ Probabilities of having n neutrons generated when starting with a source event, as calculated from expressions derived from master equations.
- ✔ Dependence on mass shown for 20 wt% ^{240}Pu and 80 wt% ^{239}Pu metallic samples.
- ✔ Change compared to non-multiplying case.
- ✔ Excellent agreement with MCNP-PoliMi.

Gamma Rays

Gamma rays have no self multiplication, instead they are generated as a by-product of the branching process of neutrons.



- ✔ Dependence on mass shown.
- ✔ Wider distribution compared to neutrons.
- ✔ Contains the information needed for calculating the moments.

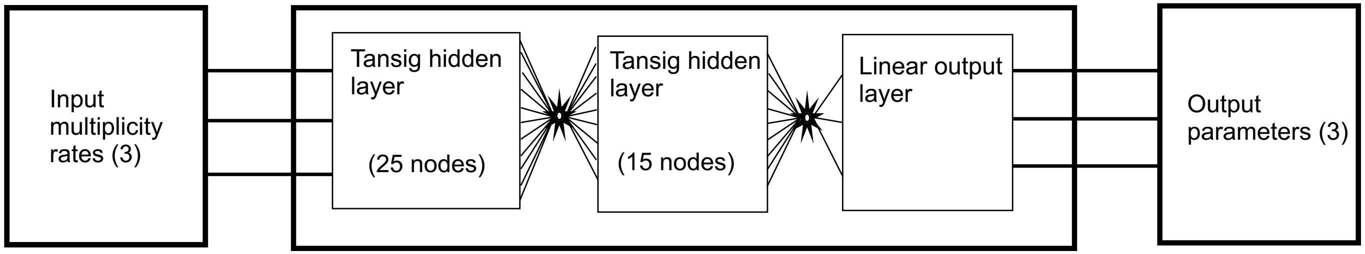
The models were extended to also include absorption and detection.

Multiplicity Counting

- ✔ Neutron multiplicity counting is used extensively for materials control and accountability (MC&A).
- ✔ The system has four parameters: neutron leakage multiplication, alpha ratio, fission rate and neutron detection efficiency.
- ✔ Three multiplicity rates are normally measured: singles, doubles and triples.
- ✔ The systems are usually based on ^{3}He-detectors, which are sensitive to slow neutrons.
- ✔ The analysis is based on the same type of master equations previously mentioned.
- ✔ Use of scintillation detectors opens of possibilities of detecting not only neutrons.

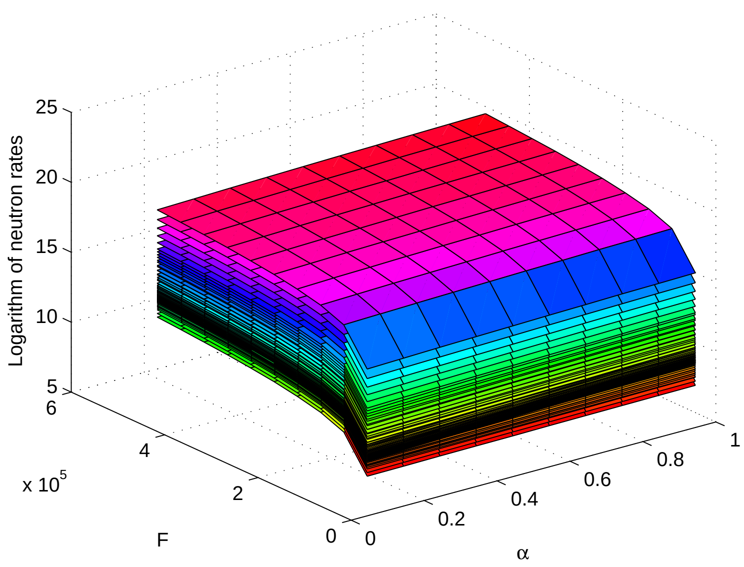
Extensions and Neural Networks

- ✔ The formalism was extended to take into account not only gamma rays, but also mixed particle multiples such as $nn\gamma$, $n\gamma$.
- ✔ The generation of neutrons and gamma rays shows interdependence.
- ✔ The analysis grows more and more complicated which motivated the usage of artificial neural networks for inverting the solutions (unfolding).
- ✔ Parameter unfolding using the information in an over-determined system.



Method Validation

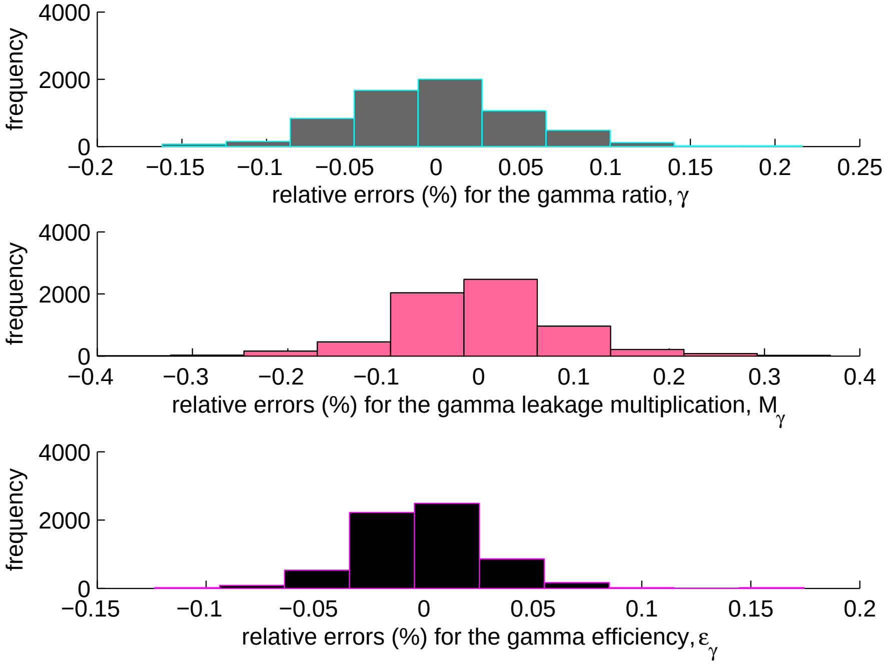
	fission rate (F)	α	p
max. abs. rel. error (%)	0.0001	0.0021	0.0001
mean error of training (%)	-1.98e-9	-4.14e-7	4.96e-9
standard deviation of training (%)	7.56e-6	2.11e-4	3.24e-5
mean error of test data (%)	1.28e-6	-7.15e-6	2.56e-6
standard deviation of test data (%)	9.34e-6	1.42e-4	3.29e-5



- ✔ The neural network was validated using neutron multiplicities.
- ✔ The accuracy of the unfolded parameters, especially the fission rate (F), which is directly linked to the sample mass, is very encouraging.

ANN Results

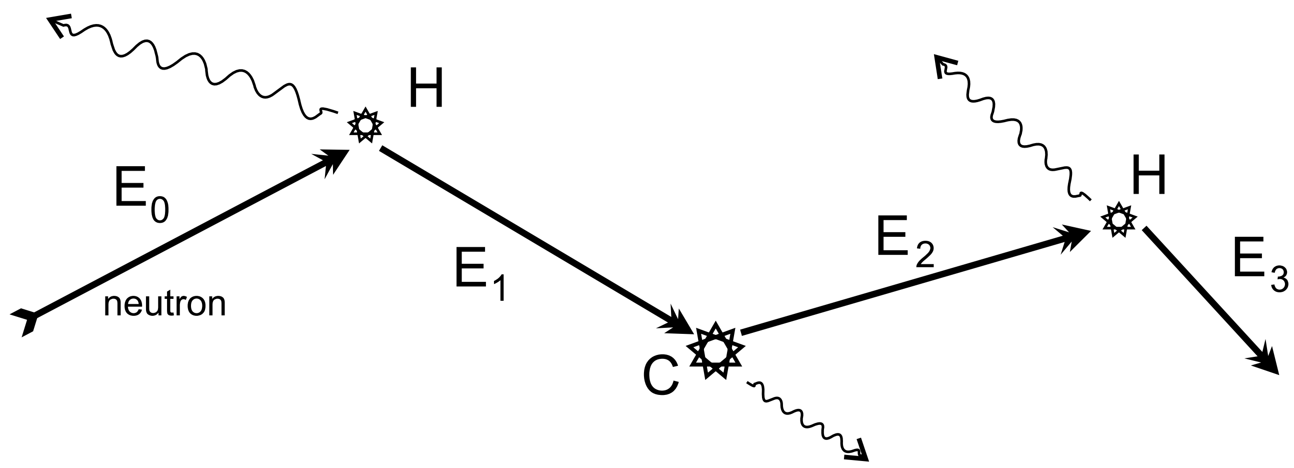
- ✔ The multiples up to third order give a total of 9 measurables.
- ✔ Additional parameters such as gamma ray detection efficiency, gamma ratio and gamma leakage multiplicity, needs to be unfolded.



- ✔ Sensitivity analysis performed by adding noise to the input data.
- ✔ Omission of input parameters showed which measurables are most important and which could be omitted in a measurement due to redundancy.

Neutron Scattering

- ✔ The light pulses generated in a scintillation detector by fast neutrons, depend on the scattering history.
- ✔ Energy is transferred in the collisions and transformed into light depending on the nuclei involved in the scattering.
- ✔ The order and type of nuclei are very important to the generated light pulse.



Convolution Integrals

New types of mathematical tools needed. It is not a branching process but rather interdependent processes:

$$f_{HCH}(L,E_0) = \int_0^{L-l_1}\int_0^{L} f_H\left[L-(l_1+l_2), E_0-(T_h(l_1)+T_c(l_2))\right]\times$$

$$f_C\left[l_2, E_0-T_h(l_1)\right] f_H[l_1,E_0]\times W\left[E_0-T_h(l_1)-T_c(l_2)-T_h(L-(l_1+l_2))\right] dl_1 dl_2.$$

using:

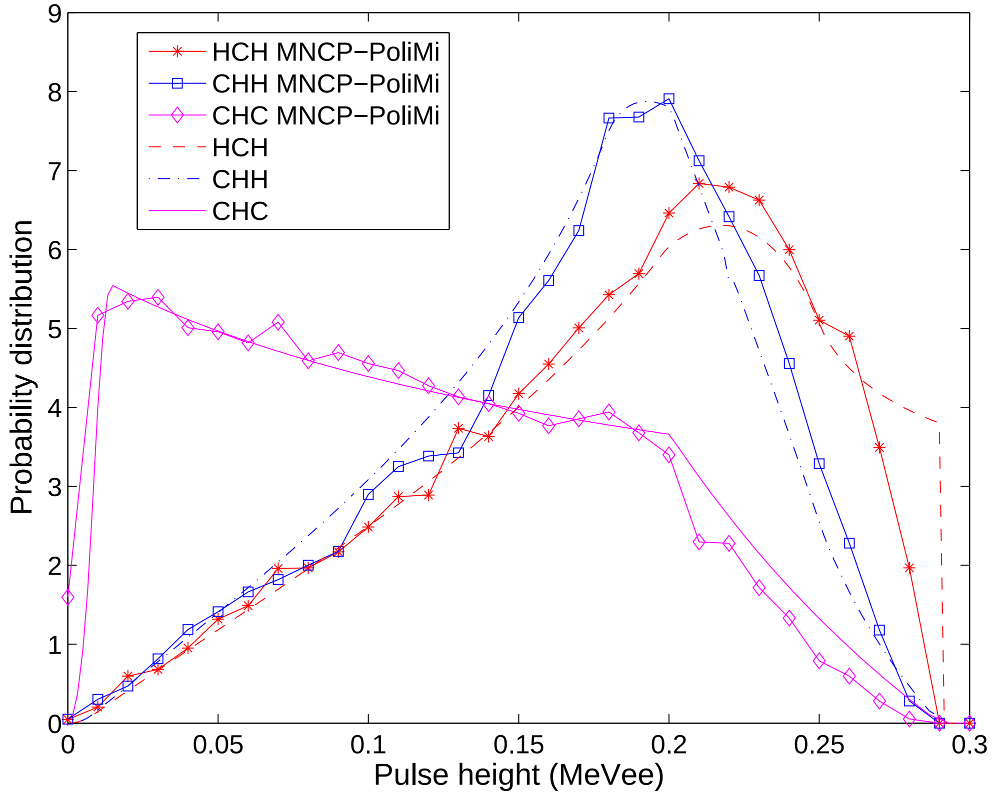
$$f_{1h}(L,E_0) = \frac{1}{E_0\sqrt{b^2+4aL}},$$

$$f_{1c}(L,E_0) = \frac{\theta(L_{max,c}-L)}{c\,(1-\alpha)\,E_0}.$$

$$T_h(L) = \frac{\sqrt{b^2+4aL}-b}{2a} \quad , \quad T_c(L) = L/c$$

Collision Histories

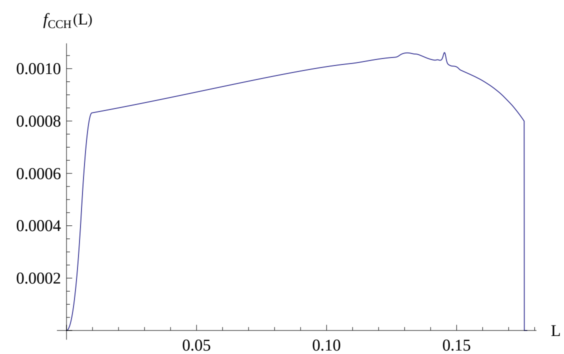
The light pulse amplitude distribution was calculated for individual collisions histories for mono-energetic neutrons of 1.5 MeV.



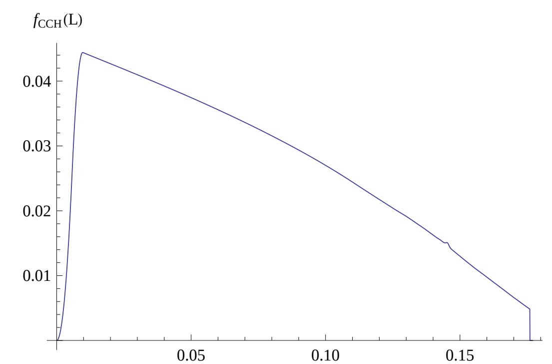
- ✔ Even for collisions histories with the same type of nuclei, the light pulse distribution is very different.
- ✔ Comparisons with MCNP-PoliMi show good agreement.
- ✔ The results from individual collisions histories can be used to understand the shape of the full light pulse distribution.

Size Dependence

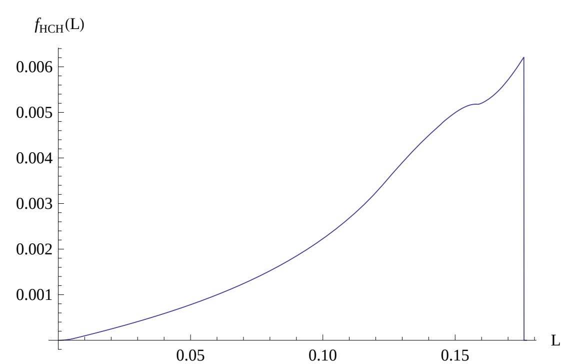
The differences depending on detector size and collision history are accurately highlighted.



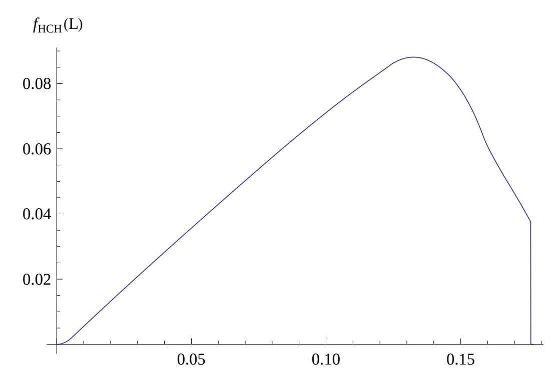
(a) CCH, 1 cm.



(b) CCH, 10 cm.



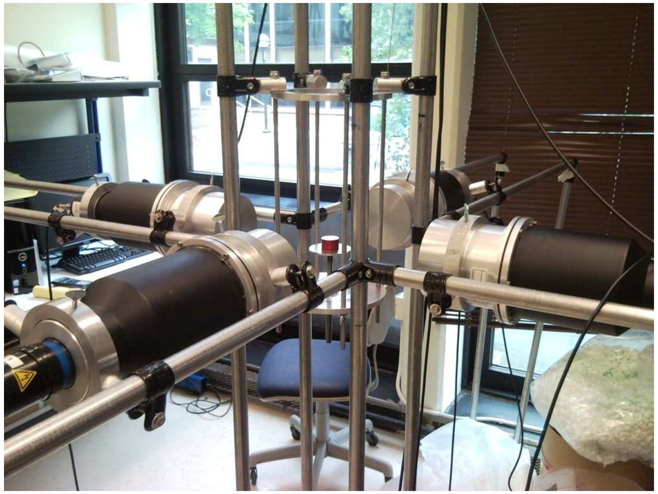
(c) HCH, 1 cm.

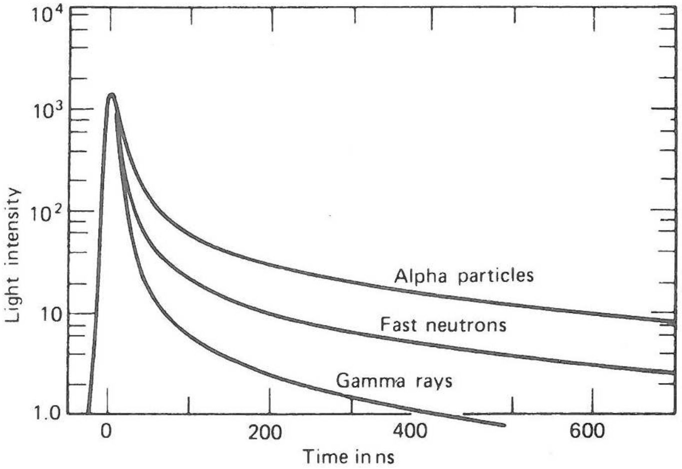


(d) HCH, 10 cm.

Event Correlations

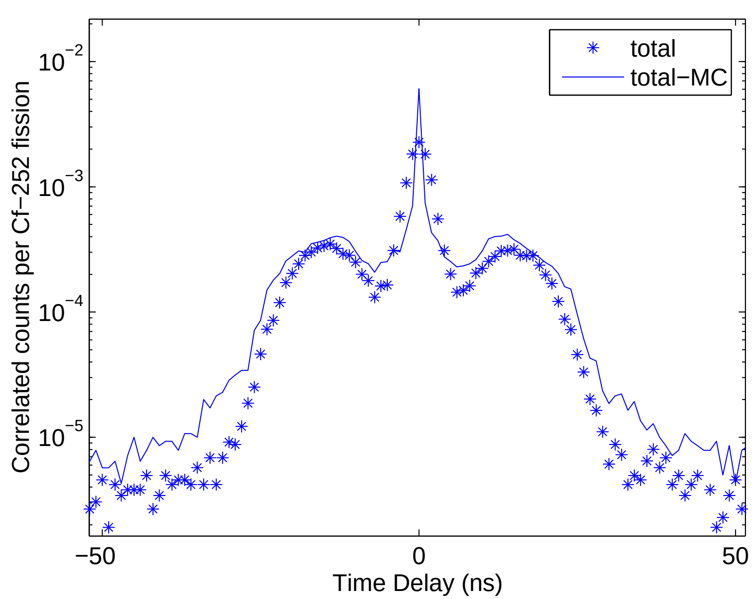
- ✔ Fissions generate a number of neutrons and gamma rays in each event.
- ✔ Simultaneously detecting multiple particles is therefore a good indicator of fissile material.
- ✔ Correlation measurements could also give additional information about sample parameters in a similar way to multiplicity measurements.
- ✔ Detecting both types of radiation has good benefits.

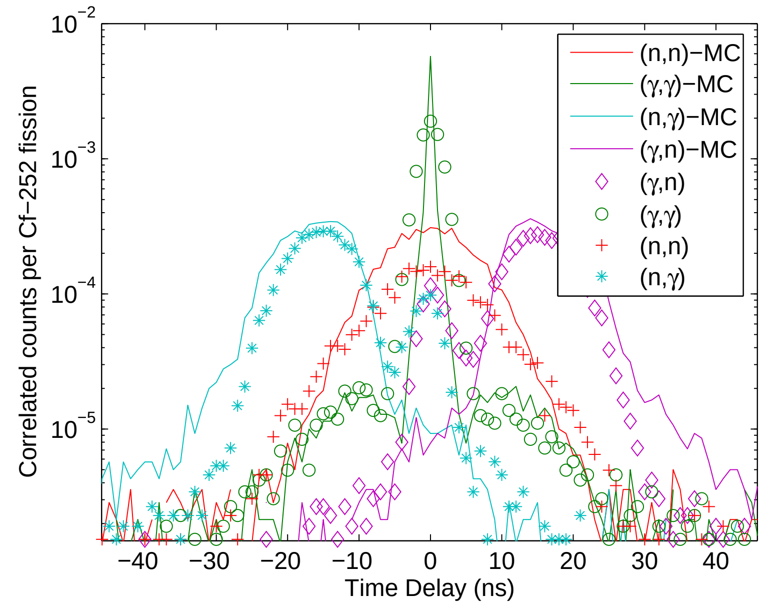




Cross-Correlations

- ✔ Cross-correlation (CC) measurements utilize two correlated pulses to give a sample-geometry signature.
- ✔ It requires very fast detectors and systems, due to the inherent speed of the gamma rays and neutrons.
- ✔ The accuracy of the CC functions depends on the pulse shape discrimination (PSD) and other considerations.



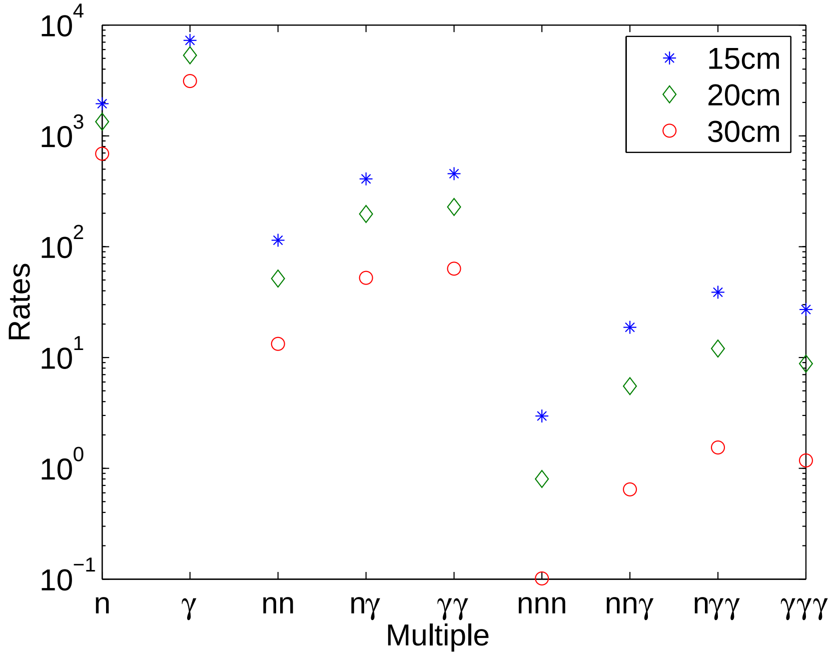


Mixed Multiplicity Counting

✔ As seen from multiplicity theory, successful detection of multiples of both neutron and photons might be used to improve pure neutron multiplicity counting.

+ No moderation needed, more measurables.

– Low detection efficiency, requires PSD or two types of detectors.



Conclusions

- ✔ The work has shown the importance of using multiple approaches: models, simulations and experiments all have strengths and weaknesses.
- ✔ Simple physical processes such as neutron scattering gives very complex analyzes and behaviours in for example scintillation detectors.
- ✔ Performing measurements have allowed for a possibility to determine which ideas are applicable, and which are subject to technical limitations.
- ✔ Algorithms and tools of mathematical physics used here occur not only in the area of safeguards but also other fields such as reactor physics.