

# Neutron Multiplicity Simulations: Geant4 versus MCNPX-PoliMi

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**ABSTRACT.** Safeguards applications, control of fissile materials and warhead verification all rely or will likely rely to a certain extent on neutron multiplicity measurements. To improve, develop, and evaluate measurement techniques, it is important to have reliable simulation tools. Beyond the technical properties of a tool, it is also beneficial if it were open source Software. This allows for easier access, but also for increased participation in development. A neutron multiplicity simulation routine based on Geant4, an open source Monte Carlo framework, has been developed over recent years. It includes appropriate particle sources including a special treatment for  $(\alpha, n)$  reactions, particle transport in the problem and analysis of the resulting pulsetrain data. To examine the capabilities of the Geant4 tool and the validity of the results, we compare them with MCNPX-PoliMi simulations in order to discuss the benefits and limitations of the Geant4 tool.

## 1 Introduction

A neutron multiplicity measurement is a non-destructive assay method carried out mainly to estimate plutonium mass of various samples.<sup>1</sup> As a nuclear safeguards technique, it is currently used to verify plutonium balances in the fuel cycle. It is, however, also relevant for disarmament verification application, e.g. as part of a verification regime to verify existing military stockpiles, or to authenticate nuclear warheads by determining plutonium content in inspected items. If the plutonium mass of a warhead is considered sensitive information, the method can be coupled with an information barrier.<sup>2</sup>

Neutron multiplicity counting research, e.g. understanding its capabilities under specific item configurations and further circumstances, and corresponding detector development rely to a large extent on simulations. Especially for the use in nuclear safeguards and disarmament verification, simulations can help to check a larger variety of detector, item and shielding setups, including possible cheating scenarios. Simulations allow for

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the development of measurements of samples that can only rarely be measured, for example kilogram quantities of plutonium.

This paper shows results for a first comparison of experimental results to those calculated with MCNPX-PoliMi<sup>3</sup> and a newly developed tool, the Open Neutron Multiplicity Simulation. The next section contains a description of the new tool, followed by a section to describe the measurement setup. Then, we present results of the simulations, followed by a short discussion.

## 2 Open Neutron Multiplicity Simulation

The Open Neutron Multiplicity Simulation (ONMS) software, developed in recent years, is an application based on the Geant4 framework.<sup>4</sup> Besides ensuring that ONMS's results are in sufficiently accurate agreement with experimental results, its development has an additional goal: The software and all its components should be open source. This would allow everyone to access the software, and also view the application's source code. ONMS is published under the GNU General Public License and available on GitHub, a platform to share and collaboratively improve open source software: <http://github.com/nuclearfreesoftware/onms>. It is possible for everyone interested to suggest changes and improvements to the code.

Such an open source approach has several advantages for the use of software in nuclear arms control. Currently used software is often proprietary (no available source code), and in many cases subject to export controls. This can limit cooperation among different states, as not all actors may have access to a specific tool. Source code access allows everyone involved to search for possible errors, but also for intentionally placed cheating mechanisms. It enables independent and critical reviews of research and supports a common understanding of a treaty verification tool's capabilities and limitations. As such, open source software could help increase trust and confidence when software results are crucial for important decisions made by states and international organizations.

The development and usage of applications for arms control purposes is often limited to small expert groups. The open source approach could help increasing participation. Such participation not only includes the possibility for everyone to use various tools, but also the possibility to generate interest among experts and specialists beyond the arms control community. As an example, it might be easier obtaining help from computer science specialists for various aspects of code development, or even from hackers ("white hat", i.e. ethical hackers) discovering possible vulnerabilities.

To carry out successful simulations of neutron multiplicity measurements, ONMS must be able to fulfill the following four main requirements:

- appropriate definition of spontaneous fission neutron sources, including correct multiplicity distributions for relevant isotopes
- neutron sources for neutrons from  $(\alpha, n)$  reactions
- neutron transport through user defined geometries, in particular including thermal neutron scattering, which is especially relevant for scattering in polyethylene
- routine to derive the measured multiplicity distribution and its moments (singles/doubles/triples) from the detected pulsetrain

All functions have been implemented in ONMS. The source definition is included in two special libraries. The user can specify a decaying material, for which the `ONMSMaterialDecay` class calculates spontaneous fission and  $(\alpha, n)$  activity. Data for decay half-life are taken from Geant4 (`G4ENSDFSTATE`), spontaneous fission branching ratios were manually added based on the most recent issues in the Journal *Nuclear Data Sheets*.

The yield from  $(\alpha, n)$  has to be calculated by summing the following equation over all possible  $\alpha$  decay energies  $E_\alpha$ , weighted by the respective decay intensities.

$$Y(E_\alpha) = \sum_i a_i \int_0^{E_\alpha} \frac{\sigma_i(E')}{S(E')} \cdot dE' \quad (1)$$

In the equation,  $a_i$  denotes the concentration of isotope  $i$  in which  $(\alpha, n)$  reactions occur,  $\sigma_i(E)$  the respective cross section and  $S(E)$  the stopping power of the source material mixture. For the stopping power, empirical data from (Ziegler 1977)<sup>5</sup> are used, for the  $(\alpha, n)$  reaction cross sections the evaluated data set JENDL/AN 2005 was used. The latter has been put together by members of the Charged Particle Nuclear Data Working Group, which was established under the Japanese Nuclear Data Committee at the Japan Atomic Energy Research Institute and includes 17 low-Z elements.<sup>6</sup> The data set also includes partial cross sections for reactions where the residual nucleus is left in an excited state. These were used to calculate the emitted neutron energy spectrum. To make the data readable for ONMS, the class `ONMSAlphaData` was developed.

As it is based on Geant4, ONMS uses a different Monte Carlo particle transport routine than most other nuclear engineering tools, which nearly all rely on MCNP (Monte Carlo N-Particle).<sup>7</sup> Important for the Monte Carlo transport is the availability of adequate cross sections. Geant4 itself includes point wise cross sections for neutron reactions of elements up to  $Z = 92$ , but none beyond. Instead of relying on the Geant4 data sets, ONMS uses data sets provided by the International Atomic Energy Agency (IAEA) and a group at the Centro de Investigaciones Energeticas, Medioambientales y Tecnologicas,

Spain.<sup>8</sup> These cross sections are based on common evaluations (ENDF-VII, JEFF 3.1) and include transuranic isotopes. For the simulations presented in the following, the ENDF-VII data sets are used. For neutron transport in the thermal regime, neutrons not only interact with individual nuclei, but also with larger molecular structures. These reactions can be treated by Geant4 similar to the  $S(\alpha, \beta)$  card in MCNP, using a slightly modified physics list. ONMS provides such a modified physics list; the thermal scattering cross sections are also part of the Geant4 data set.

The last of the four requirements listed above is a routine to derive multiplicity distributions and the singles, doubles and triples rates from the simulated detection events. ONMS includes a special library that implements an approach similar to electronic shift registers. Various steps of the software development have been presented at previous INMM conferences,<sup>9</sup> additional validation of the software has been carried out recently using data from all four phases of the ESARDA Neutron Multiplicity Benchmark exercise.<sup>10</sup>

### 3 Description of the Measurements

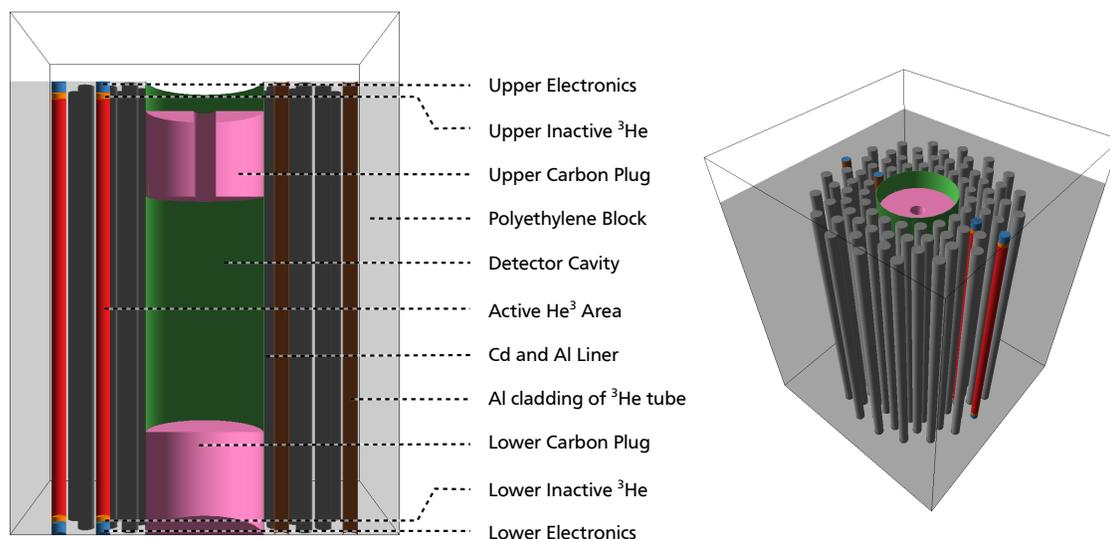


Figure 1: Geant4 rendering of Plutonium Scrap Multiplicity Counter.

In 2012, measurements of several plutonium samples were carried out at the Joint Research Centre, Ispra, Italy. For these measurements, a Plutonium Scratch Multiplicity Counter was used. A rendering of the detector can be seen in Figure 1. The detector

efficiency is 54.6% for plutonium oxides and 56.1% for metals, the difference originating from different neutron spectra. The detector was modeled according to publicly available descriptions. Detailed specifications can be found for example in the GitHub repository of ONMS, which include the model of the detector as a file in the Geometry Description Markup Language format. The detector uses 80  $^3\text{He}$  tubes embedded in a polyethylene block. The height of the block is 70.1 cm, it has a square base cross section with a width of 66.1 cm.

We present simulations for eight of the measured samples. Mass and plutonium isotopic composition of the samples are shown in Table 1. The samples include both plutonium metals and oxides, the implementation of  $(\alpha, n)$  neutron sources being relevant for the latter. In the metal samples, the plutonium is in the form of PuNiCu alloys. The samples are enclosed in small aluminum cylinders, which in turn are enclosed in stainless steel containers of cylindrical shape with a thicker top (PERLA Container). All oxide samples exist in powder form, and are enclosed in a simple aluminum cylinder. The isotopic compositions in the table were taken from characterizations carried out by the Joint Research Centre in Ispra when the samples were packaged. The results have been recorded using the Canberra JSR-14 shift register and were analyzed using the IAEA Neutron Coincidence Counting (INCC) code. At the count rates of the presented samples, dead time is negligible.

Table 1: Details of simulated and measured samples. PM1, PM2 and PM3 contain plutonium, copper, and nickel, the other samples are in the form of plutonium dioxide powder.

Sample	Pu Mass (g)	Density N (g/cm <sup>3</sup> )	Isotopic vector (wt %)					
			<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>242</sup> Pu	<sup>241</sup> Am
PM1	12.5	20.06	0.004	95.42	4.529	0.032	0.015	0.245
PM2	18.8	19.78	0.004	95.493	4.455	0.033	0.015	0.235
PM3	18.9	22.43	0.025	91.358	8.468	0.102	0.047	0.895
PuO2-10	1.987	4.243	0.058	86.082	13.27	0.321	0.27	1.483
PuO2-20	4.983	5.32	0.058	86.082	13.27	0.321	0.27	1.483
PuO2-21	9.967	5.676	0.058	86.082	13.27	0.321	0.27	1.483
PuO2-22	19.919	5.671	0.058	86.082	13.27	0.321	0.27	1.483
PuO2-23	20.566	5.166	0.084	70.906	26.856	0.691	1.463	4.878

## 4 Simulation and Results

The simulations for the previously described samples were carried out in different steps. Full results for singles, doubles and triples have been calculated on different machines. ONMS calculations have been carried out on Della, a high-performance cluster at

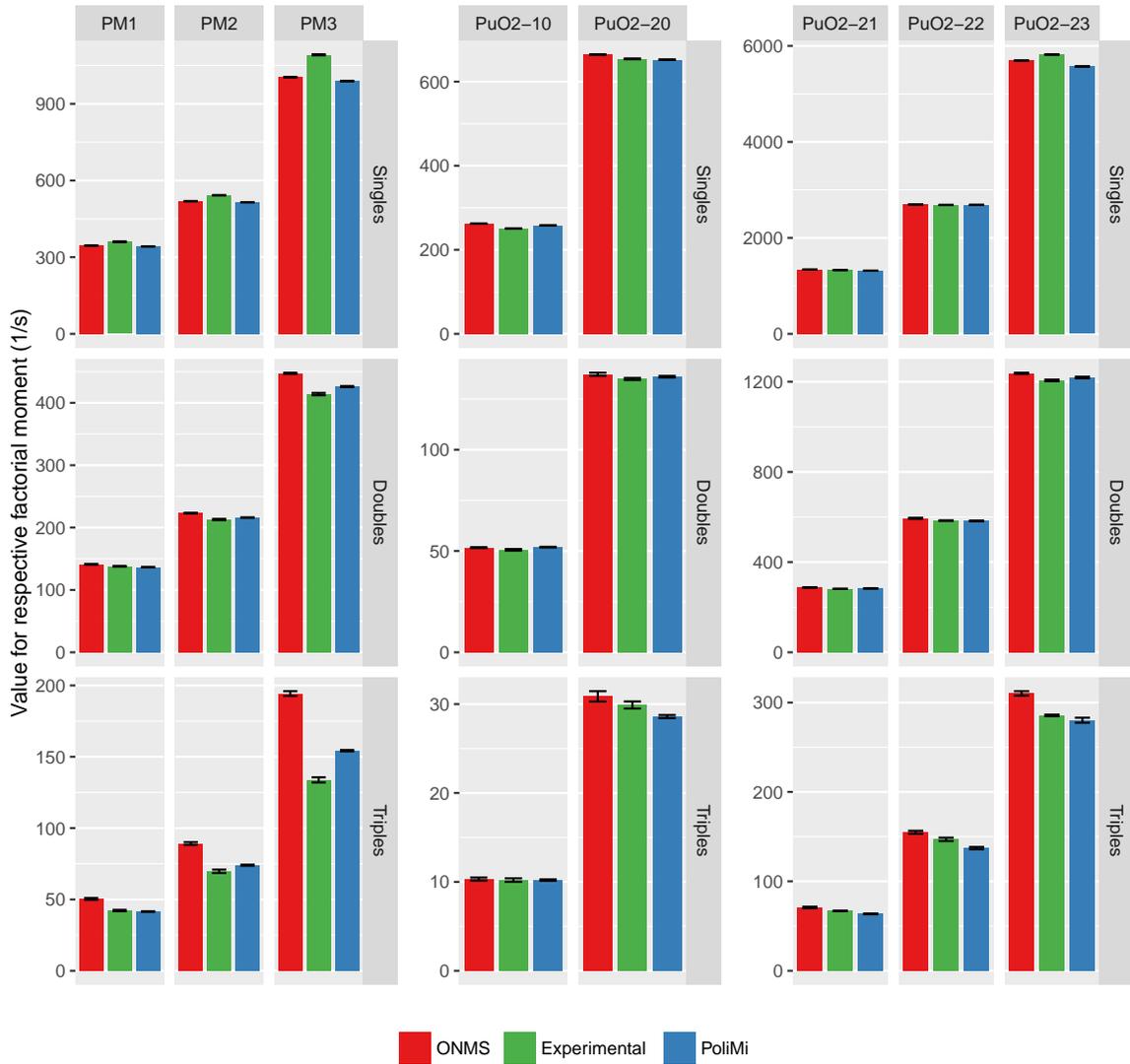


Figure 2: Results of simulations (ONMS and MCNPX PoliMi) and measurements (Experimental) for singles, doubles and triples for the different samples.

Table 2: Computation time required for simulation of 1000 s measurements for different samples. All values are given in minutes.

Sample	PM1	PM2	PM3	PuO2-10	PuO2-20	PuO2-21	PuO2-22	PuO2-23
PoliMi	11.20	16.98	32.33	8.81	21.95	44.50	91.14	189.21
ONMS	241.0	328.8	615.8	199.0	420.8	790.2	1533	3180

Princeton University. In each case and for each sample, ten separate simulations have been carried out. Resulting values are averages of these separate simulations, given uncertainties are calculated based on the standard deviation of these results. The results for MCNPX-PoliMi were calculated at the University of Hamburg Physnet cluster and have been published previously.<sup>11</sup> After having obtained the neutron pulsetrains from MCNPX-PoliMi, a Matlab script was used for the subsequent analysis.

The results of the calculation are shown in figure 2. Clearly, singles are well reproduced for all samples by both codes. This is not only an indication of code functionality, but also of proper modeling of the samples. Overall, doubles and triples are also reproduced well. For the large metallic sample (PM3), both applications overestimate the triples rate, ONMS by about 35%, MCNPX-PoliMi by 10%.

To compare the computation time of both applications, a single simulation run for each sample was carried out. These calculations were carried out on the same computer to allow for better comparison. The machine had a single CPU core (Intel Core 2 6400) with 2.13 GHz and 2 GB of memory. Results of the calculations are shown in Table 2, which shows the actual used computation time. The results show that PoliMi is faster by a factor of 15 to 20 compared to ONMS, due to three reasons:

First, to be able to use ONMS on larger machines, calculations typically have been divided into small steps, which are calculated separately to efficiently use multiple cores. For all given samples, the 1000 s measurements were divided into 200 five second steps. For each step, full initialization including the loading of cross-sections was carried out, generating an overhead. Tests show that this overhead accounts for about 10% of computation time, at least for the discussed samples.

The other two reasons are related to the neutron cross-section treatment of Geant4 (not changed by ONMS), where large amounts of time are spent for Doppler broadening of resonances and for the treatment of thermalized neutrons. These issues were identified by running ONMS using the `valgrind`, a dynamic code analysis tool. It was run in the “callgrind” mode, where it logs every call to defined functions and methods, producing a callgraph which also shows information on the most often called functions. For this calculation, a shorter simulation (only 90 s) measurement was used.

The results can be visualized in different ways, one option is a map of all subsequent calls. Figure 3 shows a subset of such a map, in this case for the `G4ParticleHPThermalScattering::ApplyYourself` function which is responsible for the treatment of thermalized neutron scattering and accounts in total for 35% of all calls. The figure shows large areas (call numbers) for functions like `malloc` (memory allocation). It seems that these are called continuously during execution, it might be possible to optimize the library by carrying out some of these calls in an initialization step only.

Not shown in the figure are calls for neutron cross-sections at higher energies. For these, the method `G4Nucleus::GetThermalNucleus` accounts for nearly 38% of the total call number. It is responsible, among other things, for Doppler broadening of the neutron interaction cross-sections depending on a material's temperature. Here, a possible optimization could be to use cross-section data sets that are pre-processed to include such broadening for a specific temperature prior to start simulations. Such improvements, as well as changes with regard to thermal neutron scattering, would require changes in the functionality of Geant4, the underlying Monte Carlo of ONMS.

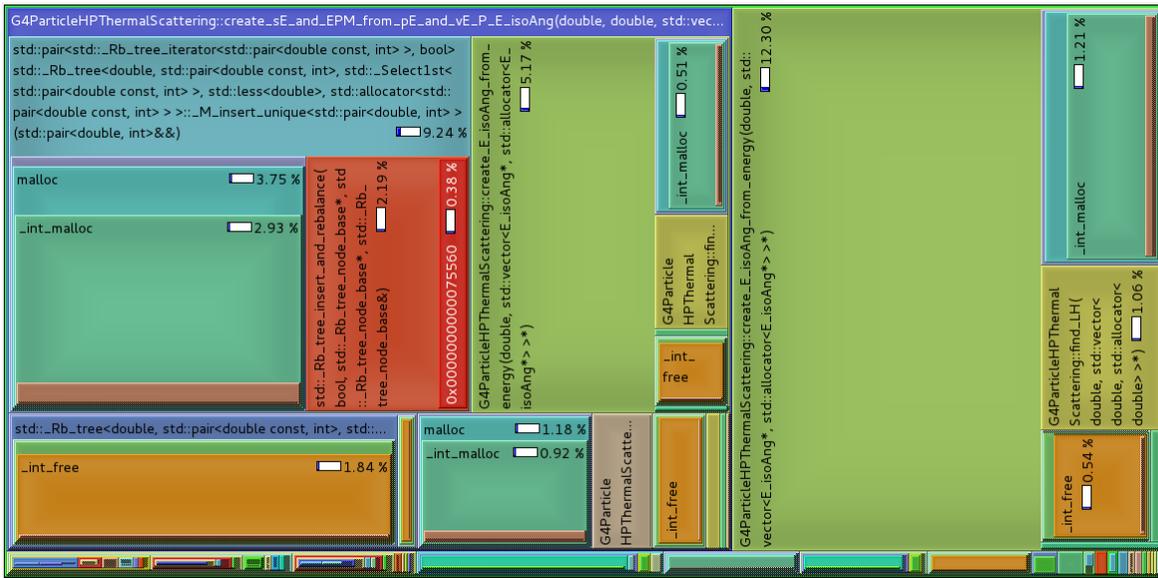


Figure 3: Subsection of Callee Map for `G4ParticleHPThermalScattering`, created with `KCacheGrind` based on `valgrind` results. The area represents about 40% of the total call number of an ONMS run.

## 5 Conclusion

The article shows results for simulations of neutron multiplicity measurements, comparing MCNPX-PoliMi and a newly developed tool, ONMS. The results of both tools show good agreement with measurements for the discussed samples. The simulations based on ONMS take significantly more computational time compared to MCNPX-PoliMi. Analyzing the reasons for the higher resource requirements, possibilities for improvement could be found with regard to the treatment of thermal neutrons and Doppler broadening of neutron cross sections. In return, ONMS is advantageous as it is available as open source software, hence everyone can use the software freely. It is

also possible for every user or developer to analyze the source code. The open source aspect can be beneficial to improve trust, transparency and confidence in the results, and is therefore particularly relevant for the cooperative development of arms control verification tools.

## Notes

<sup>1</sup>A detailed description of the method itself can be found for example in N. Ensslin et al. *Application Guide to Neutron Multiplicity Counting*. LA-13422-M. Los Alamos National Laboratories, 1998.

<sup>2</sup>An overview of different measurement methods relevant for information barriers are presented in M. Götttsche and G. Kirchner. “Measurement Techniques for Warhead Authentication with Attributes: Advantages and Limitations”. In: *Science & Global Security* 22.2 (2014). DOI: 10.1080/08929882.2014.918805.

<sup>3</sup>S. Pozzi et al. “MCNPX-PoliMi for nuclear nonproliferation applications”. In: *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 694.0 (2012), pp. 119–125. ISSN: 0168-9002. DOI: 10.1016/j.nima.2012.07.040.

<sup>4</sup>S. Agostinelli et al. “Geant4 - a simulation toolkit”. In: *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 506.3 (3 2003), pp. 250–303. ISSN: 0168-9002. DOI: 10.1016/S0168-9002(03)01368-8.

<sup>5</sup>J. F. Ziegler. *Helium: stopping powers and ranges in all elemental matter*. Vol. 4. New York: Pergamon Press, 1977.

<sup>6</sup>T. Murata, H. Matsunobu, and K. Shibata. *Evaluation of the (alpha, xn) Reaction Data for JENDL/AN-2005*. JAEA-Research 2006-052. Japan Atomic Energy Agency, 2006.

<sup>7</sup>An example for an export-controlled application, source code access is only granted after request.

<sup>8</sup>E. Mendoza, D. Cano-Ott, C. Guerrero, and R. Capote, IAEA technical report INDC(NDS)-0612, Vienna, 2012. Data available online at <http://www-nds.iaea.org/geant4>. E. Mendoza, D. Cano-Ott, T. Koi and C. Guerrero, IEEE Trans. Nucl. Science 61 (2014) 2357.

<sup>9</sup>M. Kütt. “Simulation of Neutron Multiplicity Measurements using Geant4”. In: *55th Annual Meeting of the Institute for Nuclear Materials Management, Atlanta, GA*. 2014, M. Kütt. “Geometry Independent (alpha, n) Rate Estimation for Neutron Multiplicity Measurement Simulations with Geant4”. In: *56th Annual Meeting of the Institute for Nuclear Materials Management, Indian Wells, CA*. 2015.

<sup>10</sup>P. Peerani and M. Swinhoe. “ESARDA Multiplicity Benchmark Exercise - Final Report”. In: *ESARDA Bulletin* 34 (2006), pp. 2–32, P. Peerani et al. “ESARDA Multiplicity Benchmark Exercise - Phases III and IV”. In: *ESARDA Bulletin* 42 (2009), pp. 2–25.

<sup>11</sup>M. Götttsche. “Reducing Neutron Multiplicity Counting Bias for Plutonium Warhead Authentication”. Dissertation. Universität Hamburg, 2015.