INTRODUCTION OF PLUTONIUM SYSTEMS TO THE NUCLEAR CRITICALITY SLIDE RULE

M. Duluc
Institut de Radioprotection et de Sûreté Nucléaire (IRSN)
B.P. 17, 92262 Fontenay-aux-Roses Cedex, France
matthieu.duluc@irsn.fr

D. Heinrichs, S. Kim
Lawrence Livermore National Laboratory (LLNL)
7000 East Avenue, Livermore, California 94551, United States
heinrichs1@llnl.gov
kim53@llnl.gov

T. Miller, C. Celik, C. Hopper*
Oak Ridge National Laboratory (ORNL)
P. O. Box 2008, MS-6170, Oak Ridge, Tennessee 37831, United States
millertm@ornl.gov
celikc@ornl.gov
hoppercm@comcast.net

A. Brown, C. Wilson
Atomic Weapons Establishment (AWE)
Aldermaston, Reading, RG7 4PR, United Kingdom
Alex.Brown@awe.co.uk
Chris.Wilson@awe.co.uk

M. Troisne
Millennium
16 avenue du Québec – SILIC 628, 91945 Courtabœuf, France
marc.troisne-millennium@irsn.fr

ABSTRACT

AWE (UK), IRSN (France), LLNL (USA) and ORNL (USA) began a long term collaboration effort in 2015 to update the nuclear criticality Slide Rule for the emergency response to a nuclear criticality accident. This document, published almost 20 years ago, gives order of magnitude estimates of key parameters, such as number of fissions and doses (neutron/gamma), useful for emergency response teams and public authorities. This paper presents, firstly, the motivation and the long term objectives for this update, then introduces the initial critical configuration, and finally presents the preliminary results obtained with modern 3D radiation transport codes and nuclear data for new configurations including plutonium systems.

Key Words: **Nuclear criticality accident, Slide rule, Plutonium systems.**

---

* Retired

1. INTRODUCTION

In 1997 Oak Ridge National Laboratory published the report “An Updated Nuclear Criticality Slide Rule” [1-2] as a tool for emergency response to a nuclear criticality accident. A similar document was produced by the Institut de Radioprotection et de Sûreté Nucléaire in 2000 [3]. According to [1], this kind of document “permits continued updating of information during the evolution of emergency response, including exposure information about accident victims, estimates of potential exposures to emergency response re-entry personnel, estimates of future radiation field magnitudes, and number of fissions (fission yield) estimate” without precisely knowing the initial conditions leading to the criticality accident.

This document gives order of magnitude estimates of key parameters, useful for emergency response teams and public authorities. In practice, the “Slide Rule” provides estimates of the following information based upon variable times and distances from the accident:

- the magnitude of the number of fissions based on personnel or field radiation measurements,
- prompt neutron- and gamma-dose at variable unshielded distances from the accident,
- the skyshine component of the prompt dose,
- time-integrated total radiation dose estimates,
- accumulated one-minute gamma radiation dose as a function of time after the accident, and
- dose-reduction factors for variable thicknesses of steel, concrete, and water.

The 1997 Slide Rule provides estimates for five unreflected spherical uranium systems that give general characteristics of operations typical of facilities licensed by the US Nuclear Regulatory Commission. An example of this Slide Rule is presented in Figure 1.

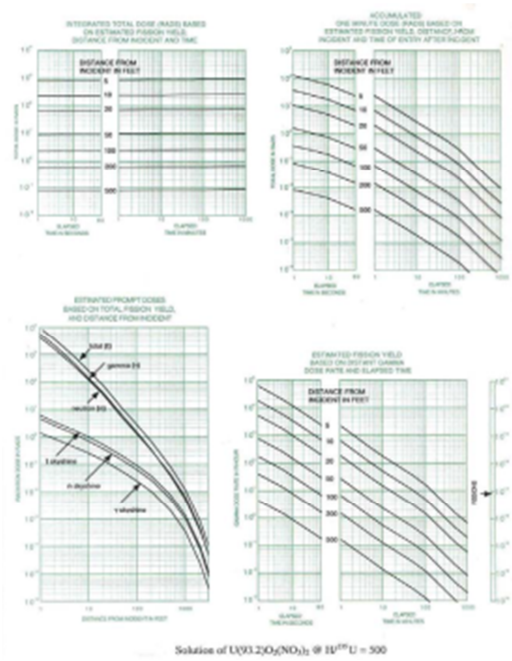

Figure 1. “1997 Slide Rule” for the uranyl nitrate solution.

Several laboratories have determined the need to review, update, and expand the contents of the 1997 Slide Rule. In particular, the only conversion factors used to provide doses (Henderson flux-to-dose factors) are outdated, so additional conversion factors are used. Also, new configurations need to be added, especially plutonium systems, for facilities that operate for the U. S. Department of Energy.

2. LONG TERM OBJECTIVES OF THIS UPDATE

A long term collaboration effort between AWE (UK), IRSN (France), LLNL (USA) and ORNL (USA) in the framework of the U.S DOE Nuclear Criticality Safety Program [4], began in order to update the Slide Rule with modern tools and add new configurations, taking into account the experience of several laboratories in using the 1997 Slide Rule. As a result, the complete work is envisioned to spread over many years and to be divided into five steps:

1. Redo with modern radiation transport tools and nuclear data libraries, for the same configurations and assumptions, the calculations performed initially for the 1997 estimation of the doses;
2. Perform additional calculations to improve the quality/quantity of information given to the user of the Slide Rule in order to not only give a value but also the possible variations and the area of applicability of this value. These additional calculations might include:
   a. new configurations (impact of the geometry and composition of the source, new fissile media including plutonium systems, multiple layers of shielding, etc.),
   b. new flux-to-dose conversion factors (for dosimetry, radiological protection and instrumentation purposes),
   c. impact of parameters on the result (sensitivity/uncertainty studies, such as thickness and composition of the ground, humidity and density of the air, etc.);
3. Review and improve the section regarding the estimation of the number of fissions;
4. Add other sections to the document like a section regarding actions to stop an on-going criticality accident (for example, standards with neutron poison);
5. Based on the previous work, the final task will be the development of a Slide Rule "application" for a handheld device (e.g. smartphone).

At the end, this work should improve the expertise for the real time response to a criticality accident in order to minimize the consequences of such an accident. In addition, this work will provide the opportunity to suggest experiments allowing the complete or partial validation of the tool results (benchmarking effort).

Another consequence of this collaborative effort might be the creation of “computer benchmarks” in order to test and validate the various variance reduction methods and to establish best practices when dealing with this kind of problem.

The first step was to redo with modern radiation transport tools, for the same configurations and assumptions, the calculations performed initially for the 1997 estimation of the doses. This effort was presented in a previous paper [5]. This present paper introduces the initial critical configuration, then presents and discusses preliminary results for new configurations (step 2) including plutonium systems, calculated by modern 3D radiation transport codes, MCNP, SCALE and COG.

3. "STEP 2" CONFIGURATION OVERVIEW

3.1. Critical systems (source)

For step 1, the initial sources of the 1997 Slide Rule were used. These are the following critical uranium systems:

- Unreflected sphere of 4.95 wt% enriched aqueous uranyl fluoride,
- Unreflected sphere of damp 5 wt% enriched uranium dioxide,
- Unreflected sphere of 93.2 wt% enriched uranyl nitrate,
- Unreflected sphere of 93.2 wt% enriched uranium sphere,
- Unreflected sphere of damp 93.2 wt% enriched uranium oxide.

For step 2 a new fissile media, plutonium at various moderation ratios (H/Pu), was used. This media is a "notional" media only composed of ^{239}Pu metal homogeneously mixed with water. For a given moderation ratio, this media minimizes the critical size of the source compared to other plutonium material (oxide, fluoride, etc.). Five moderation ratios (H/Pu) are considered: 0, 10, 100, 900 and 2000. These values roughly cover the range of representative's sizes (sphere radius) and neutron/gamma leakage and spectra for this plutonium media. Three "configurations" were considered:

- a bare critical sphere (same geometry as the initial (step 1) configuration),
- a bare critical cylinder with three different height-to-diameter ratios (0.5, 1 or 2),
- a sphere directly surrounded by a steel reflector (reflector thickness varies from 0.1 cm to 20 cm, which modifies the critical radius of the plutonium sphere).

3.2. Model description for step 2

The geometry for step 2 is derived from the 1997 Slide Rule model. It consisted of a simple 2-D air-over-ground configuration with the source located at the radial center of a right-circular cylinder. The radius and the height of the air cylinder was 1530 m. The center of the critical assemblies (spheres or cylinders) were all 1 m above the ground. The ground is now modelled as a 50 cm layer of concrete (the initial configuration and the step 1 configuration considered 1 ft (30.48 cm) of concrete). Additional calculations not shown here demonstrated that this modification has no impact on dose results for a ground made of concrete.

Figures 2 and 3 present the model for these initial calculations. For more clarity, all the information needed to calculate the step 1 and step 2 configurations were written in specific documents using a "benchmark format" [6] [7].

Neutron and gamma doses were calculated as a function of distance from 0.3 m to 1200 m. The distance between the source and the detector is measured from the external surface of the source (either plutonium for bare geometry or steel when the reflector is considered) to the center of the detector. The detector used for step 2 was chosen to be a cylindrical shell with a square cross-section of 5 cm x 5 cm to take advantage of the symmetry of the problem. The center of the detector is also at a height of 1 m above the ground.

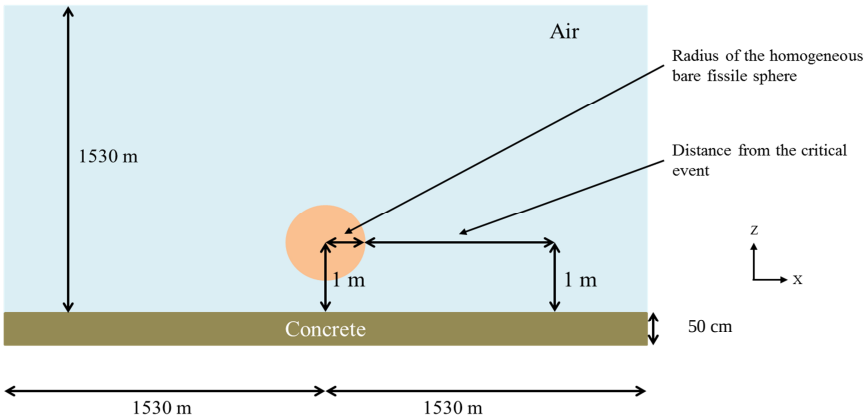

Figure 2. X-Z elevation view of the initial configuration.

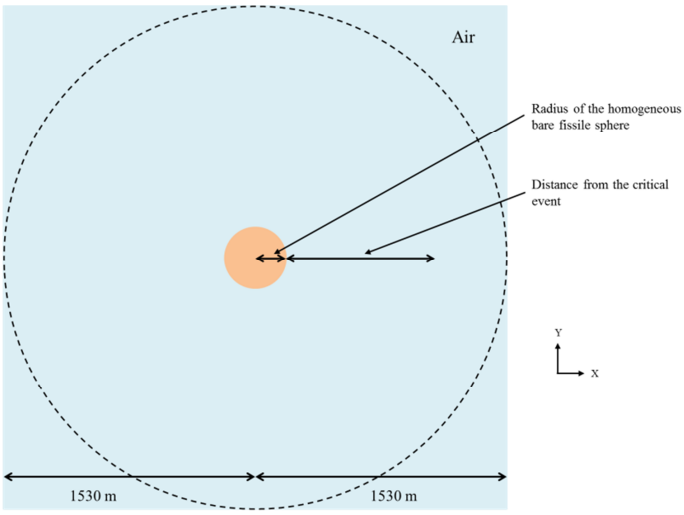

Figure 3. X-Y plan view of the initial configuration.

4. RESULTS AND DISCUSSION

This section presents and discusses the preliminary simulation results for the plutonium cases described in the previous section. As specified in reference [7], 690 results are needed to cover all the cases for step 2. The laboratories used various codes and methods, presented hereafter. Every laboratory has used the flux-to-dose conversion factors provided in the ANSI/HPS N13.3 standard [8] to compare their results in this study. This standard is specifically dedicated to the dosimetry of a criticality accident and deals with deterministic consequences of a criticality accident. Other conversion factors will be used in the future to compare the results obtained. In addition, some laboratories have performed plutonium calculations with the Henderson flux-to-dose conversion factors to compare not only the dose given by uranium systems (step 1) and plutonium systems (step 2) but also the two flux-to-dose conversion factors (ANSI/HPS N13.3 and Henderson) applied to pluto-

nium systems. Based on results from step 1, good agreement was observed between laboratories and the original Slide Rule on prompt doses (neutron and gamma). On the contrary, some discrepancies were found between laboratories and with the original Slide Rule on delayed gamma doses. Based on these observations, only prompt doses are considered in this paper whereas the complete analysis of discrepancies between laboratories for delayed gamma needs to be completed before performing additional calculations of this kind.

4.1 Codes and methods used

4.1.1 MCNP

MCNP6.1 [9] was used with the continuous energy ENDF/B-VII.1 cross section library. An F4 tally (i.e. track length estimate of cell flux) was used. A two steps method was used to generate the source term and simulate the doses. The first step is a static calculation (KCODE mode) to determine the distribution of fission neutron production inside the plutonium sphere. Many possibilities were considered but all the MCNP results presented were obtained with 20 meshes (SMESH for a sphere) or 20x20 meshes (CMESH for a cylinder), having the same volume, in which the fission reaction rate were tallied. The second step used the results of the first step to describe a fixed source (SDEF mode) of fission neutrons. A Watt spectrum was used for the energy distribution. The prompt gamma and neutron doses were determined in the same calculation, the gammas being produced by the neutron interactions inside the plutonium sphere. Indeed, in the second step, the fission neutron production is turned off (treated as absorption) but the gammas are produced (NONU = 0). Weight windows, in space and energy, were used. To generate the weight windows and the biased source for the Monte Carlo fixed source calculation, ADVANTG, a 3D deterministic code [10], was used, in order to obtain the adjoint flux. Additional information regarding this kind of calculations with MCNP might be found in references [11] and [12].

4.1.2 SCALE

SCALE 6.2.1 [13] was used with the ENDF/B-VII.1 cross section data. The libraries used with KENO and MAVRIC/Monaco both used a continuous energy representation of the cross sections. The CAAS analysis capability, coupling KENO and MAVRIC/Monaco, was used for this analysis, which is nearly an identical two-step methodology used for the MCNP analysis. First, KENO was run with a Cartesian mesh tally of the fission neutron production, which captured the asymmetry due to the ground 1 m below the center of each fissile assembly. Then MAVRIC/Monaco used the KENO tally as a fixed source, generated variance reduction parameters, and simulated the prompt doses. Region tallies were used in the model to calculate doses at the desired distances by introducing cells that were cylindrical shells in the actual problem geometry, as was done with MCNP. For the prompt dose calculations, total nu-bar was used just like the MCNP calculations.

4.1.2 COG

COG 11.2 [14] [15] was used with ENDF/B-VII.1 cross-section data. COG is a general purpose, multi-particle, high-fidelity Monte Carlo code developed by Lawrence Livermore National Laboratory (LLNL). It provides accurate simulation results for complex 3-D shielding, criticality safety,

and activation problems. A direct one-step criticality/detector calculation method was applied for all prompt neutron and gamma ray dose calculations. Each neutron and gamma particle is tracked from its birth in fission within the fissile volume to its absorption in the system or score at the detectors at various distances in one single, massively parallel, COG supercomputer run with no variance reduction biasing applied.

4.2 Results

Personal absorbed doses presented in this paper are given in Gray for criticality accidents that generate 10^{17} fissions. The following sections will present the results for the bare plutonium sphere (4.2.1) with additional comparisons with step 1 uranium systems and flux-to-dose conversion factors, the bare plutonium cylinder (4.2.2), and the plutonium sphere surrounded by a steel reflector (4.2.3).

4.2.1 Bare Plutonium sphere

The figures 4 and 5 show the neutron and gamma dose for the bare spherical plutonium system calculated with MCNP with the ANSI/HPS N13.3 flux-to-dose conversion factors. For a given distance, the neutron dose is higher when the moderation ratio (H/Pu) is low. The difference between the extreme cases goes from a factor 13 for short distances to a factor 4.5 for long distances. Regarding prompt gamma doses, the lowest dose is obtained for the metal plutonium system whereas the highest dose is obtained for an intermediate moderation ratio. At short distances, the difference between the extreme cases is a factor 5 on prompt gamma doses. After 500 meters, all the gamma curves become closer with a maximum ratio of 1.5. All these observations can be roughly explained by the various auto-absorption inside the sphere and by the fact that neutrons are produced only in the plutonium sphere whereas prompt gammas are produced not only in the plutonium sphere but also by neutron with (n, gamma) reactions within the surrounding environment.

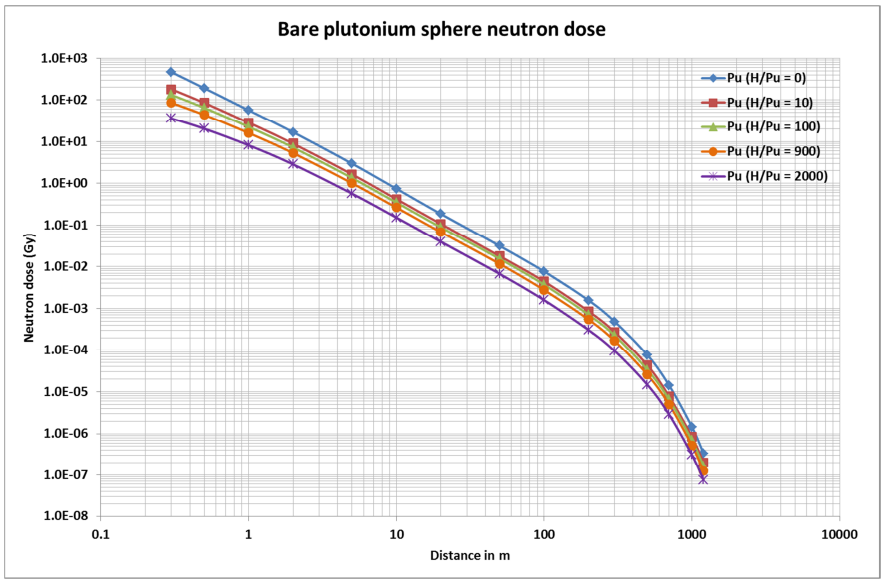

Figure 4. Neutron doses for bare plutonium sphere.

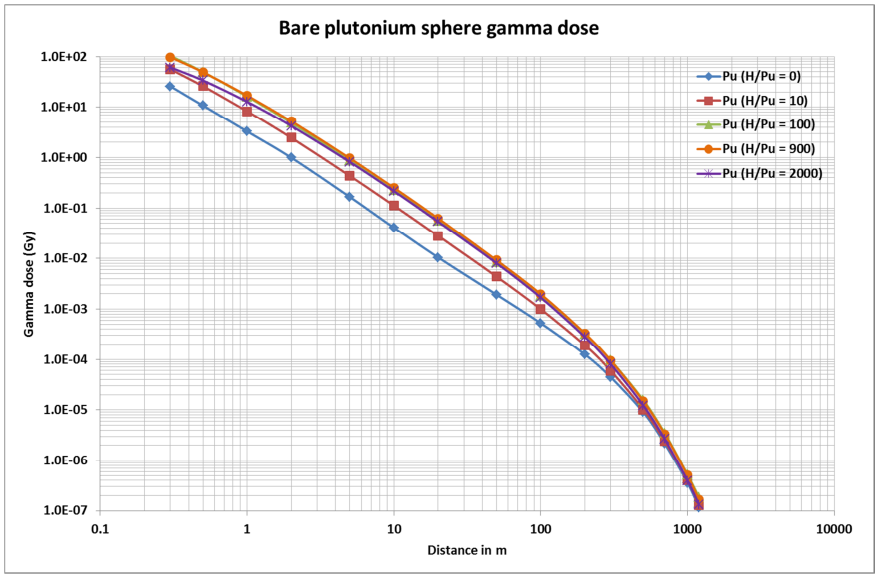

Figure 5. Prompt gamma doses for bare plutonium sphere.

The figure 6 shows the neutron/gamma dose ratio for the bare spherical plutonium system. For all the moderation ratios, the neutron/gamma dose ratio is relatively constant from 0.3 cm up to 20 meters. Beyond 20 meters, the ratio constantly decreases for metal plutonium whereas it continues to increase then decreases after 500 meters for other configurations.

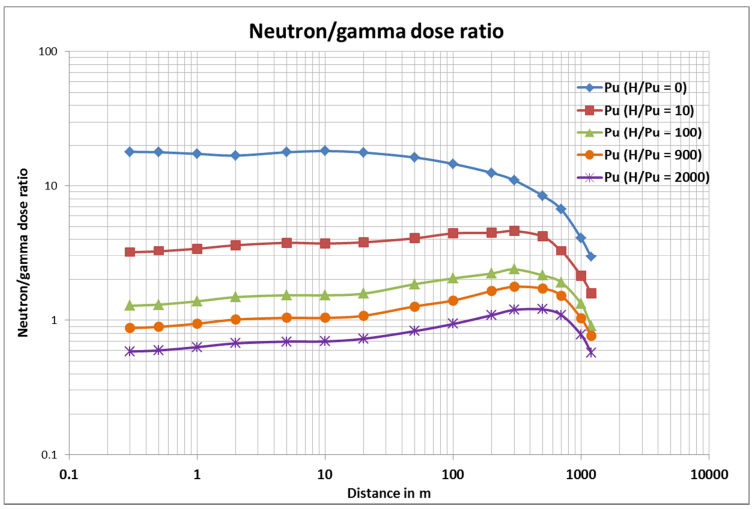

Figure 6. Neutron/gamma dose ratio for bare plutonium sphere.

The figure 7 presents the code to code comparison of neutron and gamma doses for the bare plutonium sphere. Good agreement is observed between codes with discrepancies generally lower than 5 %. At long distances, the discrepancies between codes are higher (without any trend) but with higher uncertainties. Generally, the relative error (1 σ) on the code results is lower than 1 % until

500 meters and lower than 5 % at long distances. For prompt gamma doses, a small discrepancy between codes is perceptible and might be due to the different treatment of bremsstrahlung by these codes. MCNP's thick target bremsstrahlung model accounts for the electromagnetic cascade of gammas and electrons that produce many low energy bremsstrahlung gammas, and allows users to not perform electron transport for geometries with thick shielding materials. All of SCALE's fixed-source radiation transport codes use gamma production data based on ENDF, which does not include this sort of bremsstrahlung. When this model is turned off (PHYS:P j 1), the MCNP and SCALE results are statistically the same. Regarding COG, whenever an electron-producing photon reaction occurs, COG checks whether the reaction occurred in a region enabled by the user for electron transport. If not (the case here), then the electron energy is immediately deposited. All these observations regarding code to code comparison could be applied to the other kind of calculations performed in this paper, including figures 12 and 13 of appendix A.

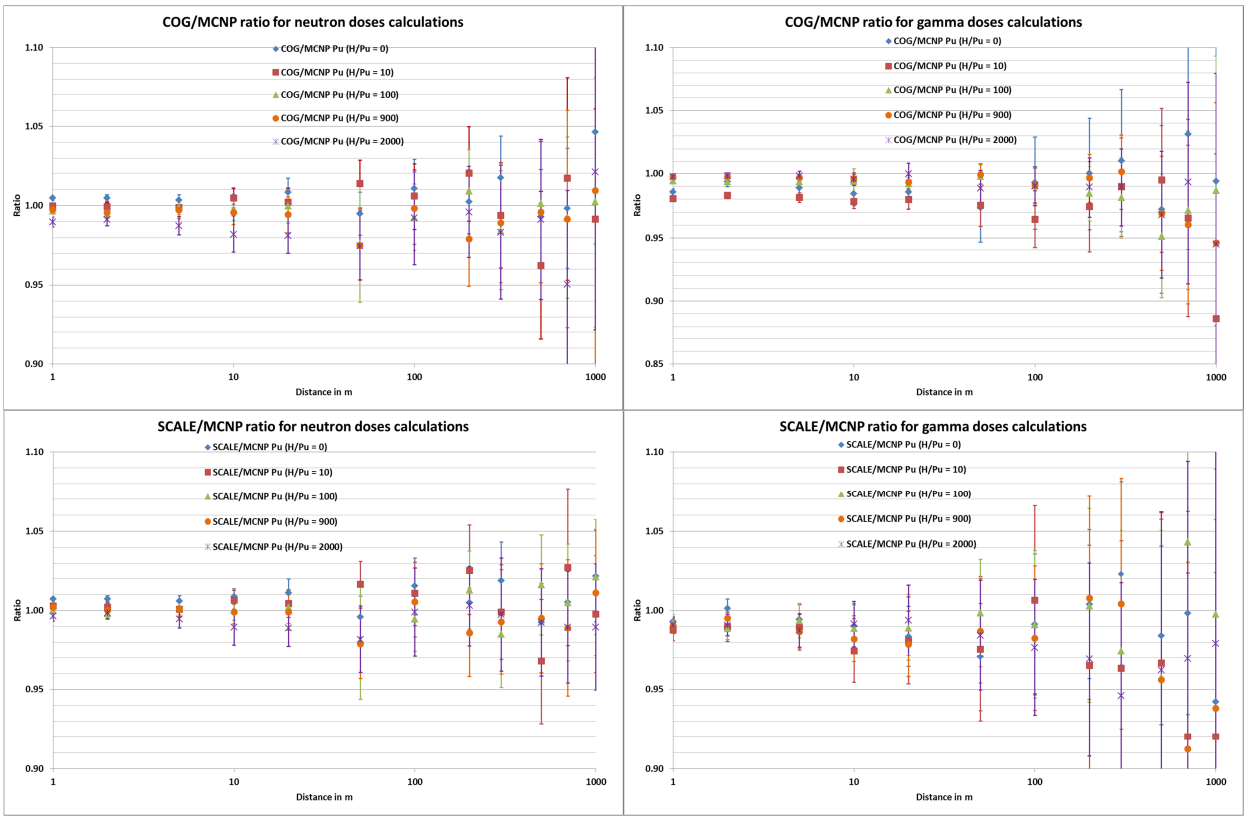

Figure 7. Code to code comparison for the bare plutonium sphere.

The figure 8 presents the ratio between neutron and gamma doses calculated with MCNP for bare plutonium system with the Henderson flux-to-dose conversion factor (used in the original slide rule) and the ANSI/HPS N13.3 flux-to-dose conversion factor. It can be seen that the ANSI/HPS N13.3 flux-to-dose conversion factor is more penalizing than the Henderson flux-to-dose conversion factor (at least 20 % for neutron with an increase when the distance increases and about 10 % for gamma independently of the distance). Indeed, the ANSI/HPS N13.3 flux-to-dose conversion factor is al-

ways larger than the Henderson flux-to-dose conversion factor with an increasing difference for intermediate and low neutron energies.

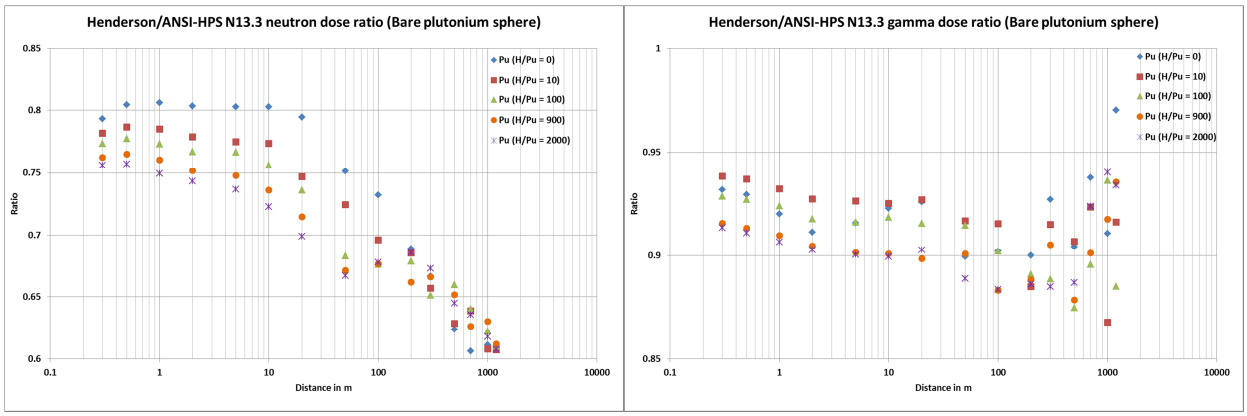

Figure 8. Henderson/ANSI-HPS N13.3 dose ratios for bare plutonium sphere (MCNP).

The figure 9 presents the neutron and gamma doses for the uranium systems (from the original slide rule) and the plutonium system (from step 2) calculated with MCNP with the Henderson flux-to-dose conversion factor. These figures show that there are no major differences, in terms of trend, between plutonium and uranium systems. The extreme cases (max/min) for doses are:

- for neutron, plutonium metal and moderated plutonium (H/Pu = 2000),
- for prompt gamma, intermediate plutonium system (H/Pu=100 or 900) and uranium metal.

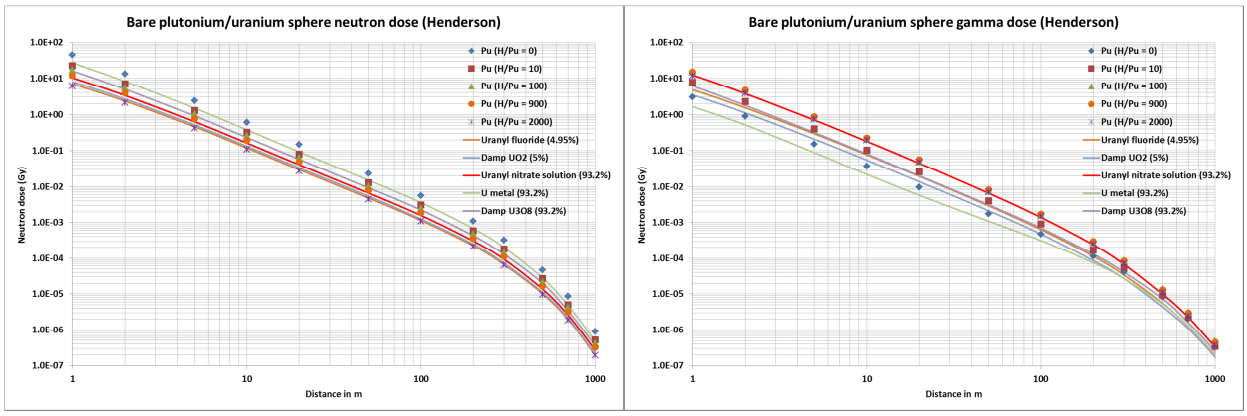

Figure 9. Bare sphere doses using Henderson flux-to-dose conversion factors (MCNP).

It can be seen than, on average for the Henderson flux-to-dose conversion factors, the plutonium metal configuration generates doses 70 % higher than the uranium metal configuration (for both neutrons and prompt gamma). This kind of result shows the interest to update the original slide rule with plutonium systems.

4.2.2 Bare Plutonium cylinder

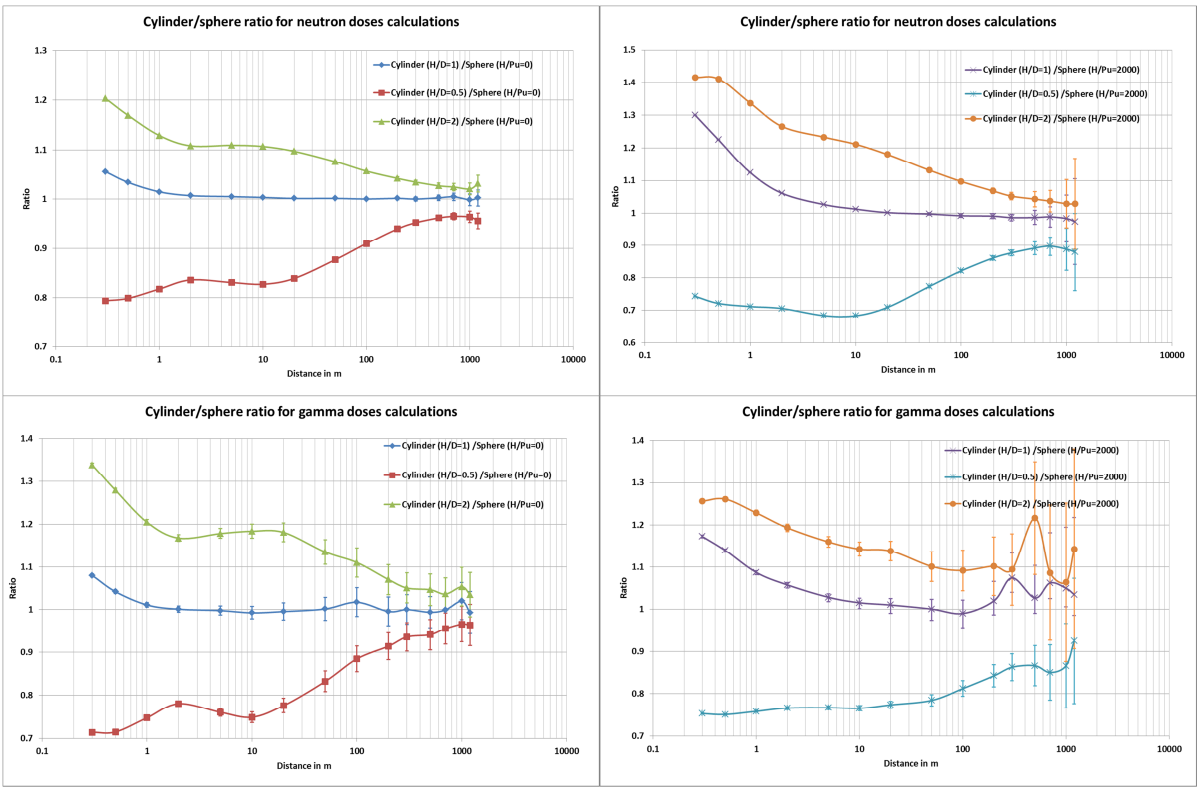

Figure 10. Bare Cylinder/sphere dose ratios (SCALE).

The figure 10 shows, for neutron and prompt gamma, the ratio between the doses calculated with the critical bare plutonium cylinders (with three different height-to-diameter ratios (0.5, 1 or 2)) and with the critical bare plutonium sphere. The results are shown for two moderation ratios, H/Pu=0 and H/Pu=2000. The discrepancies between the three cylinders and the sphere are within 30 % and decrease with the increasing distance. The solid angle of the various geometries explains the behavior of the ratio. The ratio tends to approach one for long distances but is not completely reached for gamma dose for H/Pu=2000 and a cylinder with a height-to-diameter ratio of 0.5.

4.2.3 Plutonium sphere surrounded by a steel reflector

The figure 11 shows, for neutron and prompt gamma, the ratio between the doses calculated with steel reflected plutonium sphere (with various thicknesses) and with bare plutonium sphere. The results are shown for two moderation ratios, H/Pu=0 and H/Pu=2000.

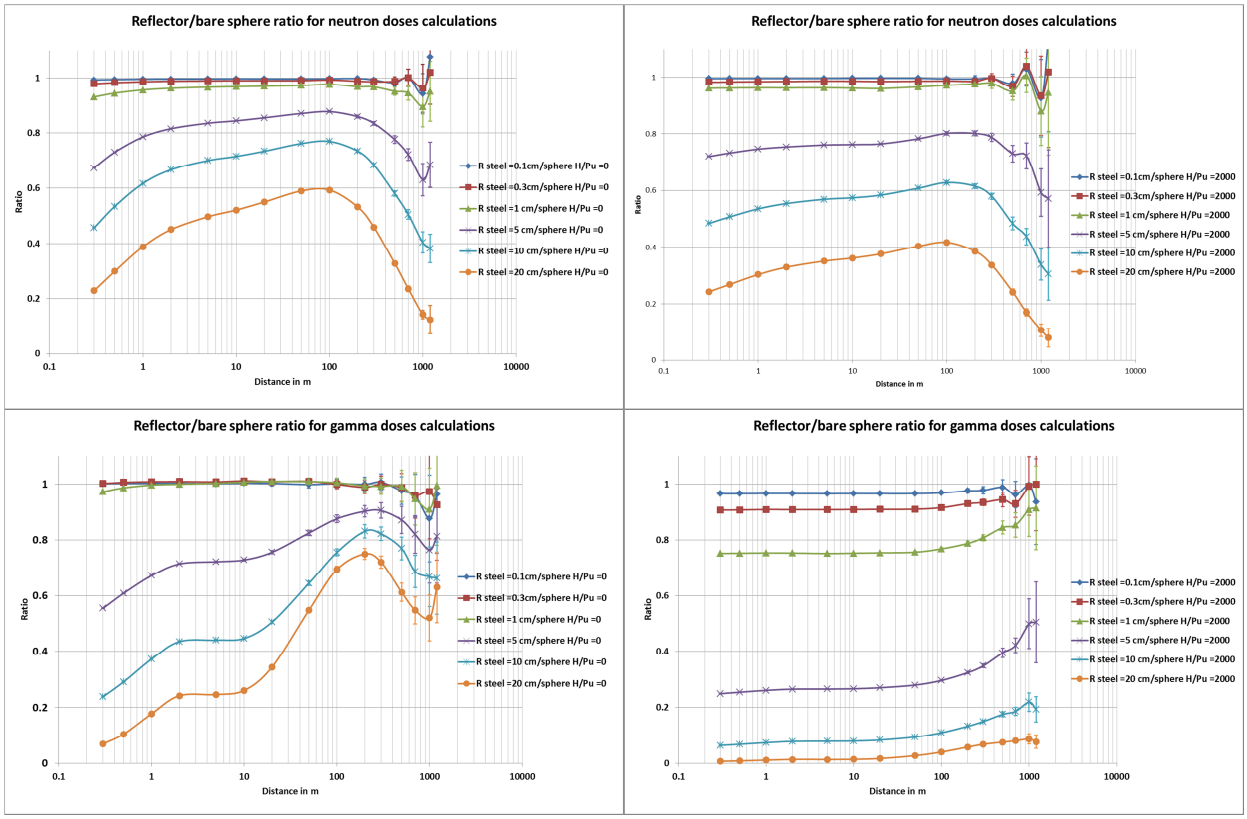

Figure 11. Reflected/bare sphere dose ratios (COG).

For these reflector and fissile material configurations, the bare sphere is the most penalizing configuration. The decrease of the size of the critical plutonium sphere (implying less absorption inside the plutonium sphere) does not compensate for the attenuation of the dose due to the reflector. The attenuation effect is not constant with the distance and depends on the moderation ratio and the type of incident particle (neutron / gamma). The steel reflector is particularly efficient against the gamma dose for the moderated plutonium (H/Pu=2000).

5. CONCLUSIONS

AWE (UK), IRSN (France), LLNL (USA) and ORNL (USA) began a long term collaboration effort in 2015 to update the nuclear criticality Slide Rule for the emergency response to a nuclear criticality accident. This document gives order of magnitude estimates of key parameters, useful for emergency response teams and public authorities. The second step of this update introduces plutonium systems, new geometries, and new flux-to-dose conversion factors to the nuclear criticality slide rule. Regarding the calculation of the prompt doses, the results from the modern tool used for this update are consistent with small discrepancies on prompt gamma due to the different codes gamma transport treatment of bremsstrahlung. The addition of plutonium systems results does not modify the trend of the dose curves compared to the initial uranium systems. The more recent flux-to-dose conversion factors are more penalizing than the initial ones, in particular for neutrons at long dis-

tances. The considered bare cylinders have an impact on the doses up to 30% compared to the bare sphere but approach, more or less quickly, to the sphere dose for long distances. The steel reflector deeply modifies doses and the effect depends on several parameters (distance, moderation ratio, type of radiation). It is then difficult to attribute one reduction factor value to a given thickness of steel. In addition to the finalization of the calculations and the complete analysis of the results, effort will be devoted on the calculation of delayed gamma doses and other tasks like the impact of multiple layers of shielding, impact of the thickness and the composition of the surrounding environment (ground, humidity of the air, etc.).

In parallel to this Slide Rule update effort (useful to limit the radiological consequences if a criticality accident should occur), this collaborative effort might be a good opportunity to create "computer benchmarks" in order to test and validate the various variance reduction methods and to establish best practices when dealing with this kind of problem (in particular fission source calculation). Finally, this work will provide the opportunity to suggest experiments allowing the validation of the tool results (benchmarking effort). Indeed, the ICSBEP handbook for this subject is only composed of seven benchmarks, with only one case representative of a criticality accident [16].

ACKNOWLEDGMENTS

This work was performed under the auspices of the U.S. Department of Energy by Oak Ridge National Laboratory and Lawrence Livermore National Laboratory under contracts DE-AC05-00OR22725 and DE-AC52-07NA27344, respectively, and was funded by the U.S. Department of Energy Nuclear Criticality Safety Program and by IRSN. IRSN authors would like to thank Sylvain BEYTOUT, from MILLENNIUM, for the preliminary calculations he did for this update.

REFERENCES

[1] B. L. Broadhead et al., "An Updated Nuclear Criticality Slide Rule: Technical Basis," Report NUREG/CR-6504 Vol. 1 ORNL/TM-13322/V1, (1997).
[2] C. M. Hopper, and B. L. Broadhead, "An Updated Nuclear Criticality Slide Rule: Functional Slide Rule," Report NUREG/CR-6504, Vol. 2 ORNL/TM-13322/V2, (1998).
[3] Fiches Accident-Type, IPSN internal report, (2000).
[4] NCSP website, http://ncsp.llnl.gov/
[5] M. Duluc et al., "Update of the Nuclear Criticality Slide Rule for the Emergency Response to a Nuclear Criticality Accident," ICRS 13 RPSD 2016 conference, October 3-6, 2016 Paris, France (2016).
[6] "Update of the nuclear criticality Slide Rule calculations - initial configuration," SR-U-UNREFLECTED-GROUND-001, IRSN internal report (2015).
[7] Update of the nuclear criticality slide rule calculations – plutonium configurations, SR-Pu-UNREFLECTED-GROUND-001 and SR-Pu-STEEL-GROUND-001, IRSN internal report (2017).
[8] "Dosimetry for Criticality Accidents," ANSI/HPS N13.3-2013 (2013).
[9] J. T. Goorley et al., "Initial MCNP6 release overview – MCNP6 version 1.0," LA-UR-13-22934, (2013).

[10] S. W. Mosher et al., "ADVANTG—An Automated Variance Reduction Parameter Generator," ORNL/TM-2013/416 Rev. 1 (2015).
[11] B. C. Kiedrowski, "MCNP6 for criticality accident alarm systems – A Primer," LA-UR-12-25545 (2012).
[12] T. M. Miller and D. E. Peplow, "Guide to performing computational analysis of criticality accident alarm systems," ORNL/TM-2013/211 (2013).
[13] B. T. Rearden and M. A. Jessee, eds., "SCALE Code System," Oak Ridge National Laboratory report ORNL/TM-2005/39, Version 6.2. Available from Radiation Safety Information Computational Center as CCC-834, (2016).
[14] "COG: A Multiparticle Monte Carlo Code Transport Code, Version 11.1," CCC-829, Released by Radiation Safety Information Computational Center (2015).
[15] R. M. Buck et al., "COG11.1 Description, New Features, and Development Activities," pp. 910-922, International Conference on Nuclear Criticality Safety (ICNC 2015), Charlotte, NC, September 13-17, (2015).
[16] T. M. Miller and al., "Neutron Activation Foil and thermoluminescent dosimeter responses to a bare pulse of the CEA Valduc SILENE critical assembly," ALARM-TRAN-AIR-SHIELD-001, International Criticality Safety Benchmark Evaluation Project, OECD/NEA/ICSBEP (2015).

APPENDIX A. ADDITIONAL CODE TO CODE COMPARISONS

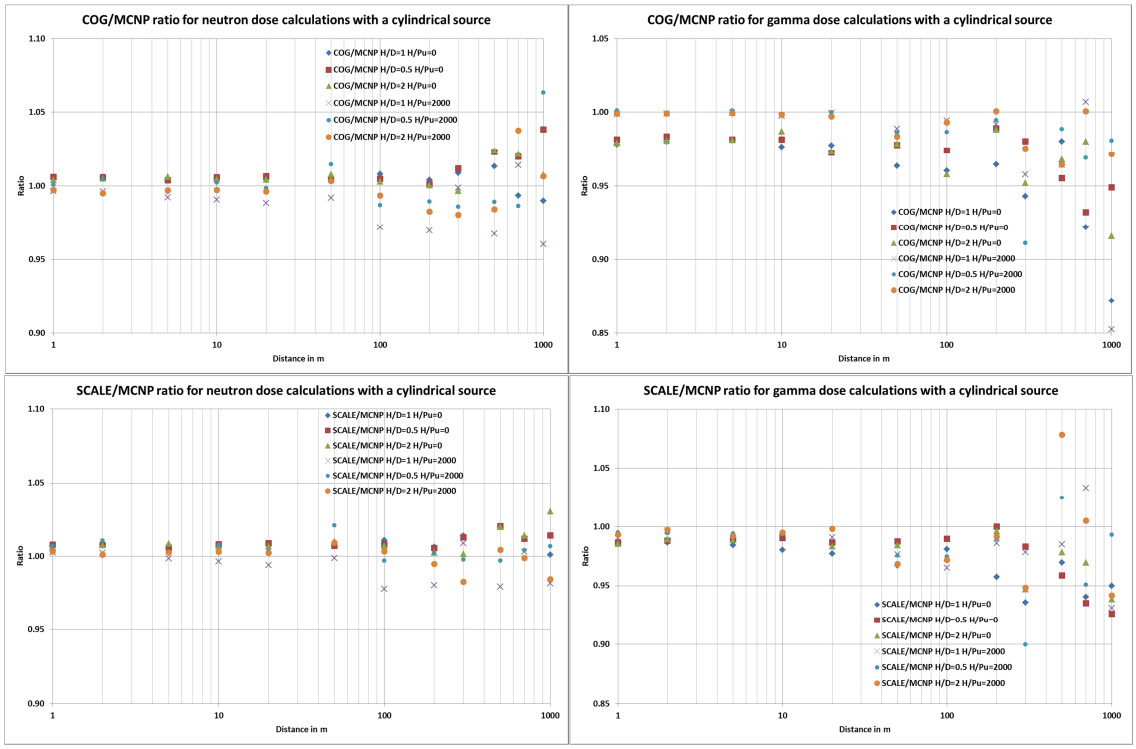

Figure 12. Code to code comparison for the bare plutonium cylinders.

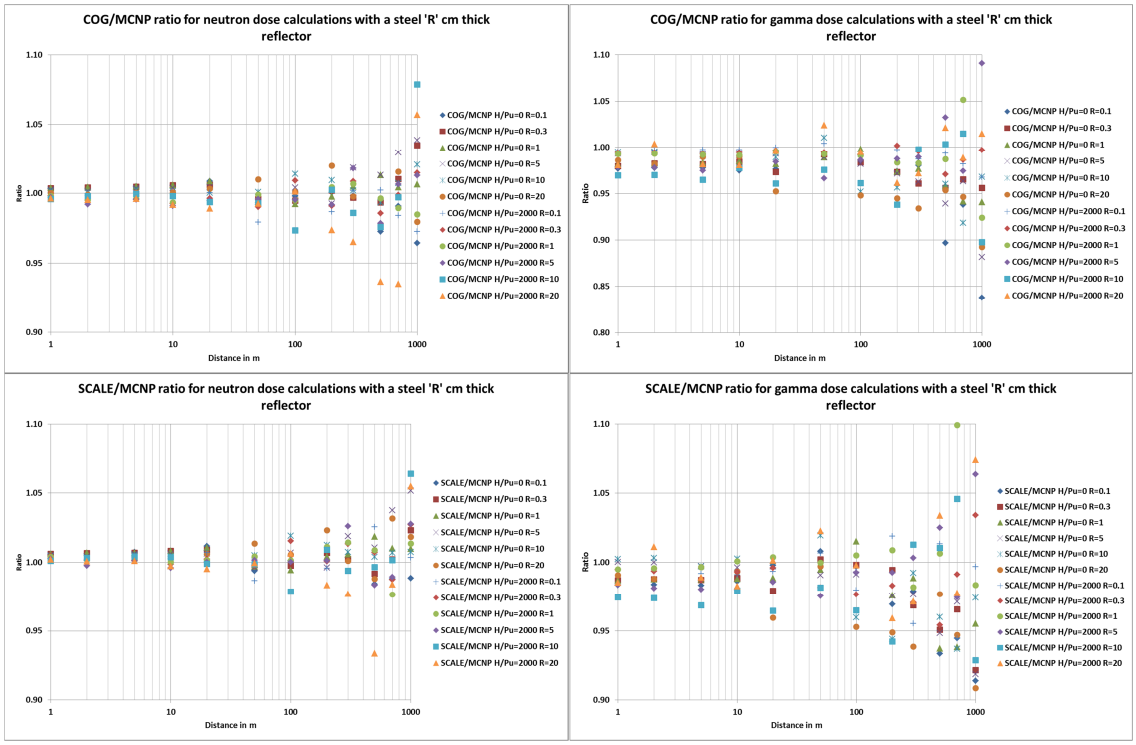

Figure 13. Code to code comparison for the plutonium spheres surrounded by a steel reflector.