Calculations Versus Measurements for Residual Dose Rates from SNS Spent Structures

I.I. Popova*, F.X. Gallmeier, S. Trotter, M. Dayton

ORNL, PO box 2008, MS 6466, Oak Ridge, TN 37831-6474, US
** corresponding author email: popovai@ornl.gov; phone: +1-865-898-4380; fax:+1-865-574-4140*

***ABSTRACT**: Residual dose rate measurements were conducted on target vessel #13 and proton beam window #5 after extraction from their service locations. These measurements are used to verify calculation methods of radionuclide inventory assessment that are typically performed for nuclear waste characterization and transportation of these structures. Neutronics analyses for predicting residual dose rates are carried out using the transport code MCNPX and the transmutation code CINDER90. For transport analyses a complex and rigorous geometry model of the structures and their surroundings are applied. The neutronics analyses are carried out using the Bertini and CEM high energy physics models for simulating particles interactions above the table-based cross section range. Obtained calculational results are analyzed and compared to the measured dose rates and overall show good agreement within 25%, which shows applicability of the methods used in analyses.*

Keywords: Accelerator Facilities, Particle Transport, Monte Carlo Codes, Dose Rates, Radioactive Waste

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1. INTRODUCTION

The Spallation Neutron Source (SNS) in Oak Ridge, Tennessee [1], is an accelerator driven neutron scattering facility for materials research. Presently SNS is capable to operate at 1.4 MW proton beam power incident on a mercury target with a proton beam energy of 1 GeV and in a pulse structure of 60 Hz repetition rate. SNS consists of accelerator facility, target facility, and a world-class suite of neutron scattering instruments. The SNS target is liquid mercury, which is housed in a double walled steel vessel. The proton beam window (PBW) establishes the boundary between the accelerator and the target, through which the full beam passes before it hits the target.

Being exposed to a high-radiation environment, both target vessel and PBW reach their end-of-life because of radiation induced material damage and pulse-induced erosion. The maximum allowable peak damage of steel and Inconel structures, which are materials for target vessel and PBW module, is 12 dpa [2]. To ensure safe operation, they are scheduled for periodic replacement, which takes place during facility shutdown for maintenance. Spent components are dismounted, stored for cooling down, and finally transported off-site for permanent storage. The target vessel is exchanged about two - three times per year and the PBW is exchanged every two - three years.

Each spent structure that leaves the SNS site requires supporting documentation with radionuclide inventory and dose rate prediction for the time of the transportation. Neutronics analyses are performed, assuming realistic irradiation history and decay scenario to ensure that the container/package, housing the structure, is compliant with the waste management regulations. These analyses require usage of a complex and rigorous geometry of the spent components and their surroundings, application of particle transport and transmutation codes, and involves data analysis [3].

To validate these analyses and show applicability of codes and methods, an effort to perform measurements of dose rates from spent structures during their cool-down time has taken place. Measurements are done for the spent target vessel #13 and the spent PBW module #5. Neutronics analyses are carried out to calculate residual dose rates induced from each component for the time during measurements. To estimate how sensitive calculations are to initial assumptions, analyses for the spent target vessel are done using different high-energy physics models and introducing some fluctuation in the geometry.

The calculational results for both components are analyzed and compared to measured residual dose rates.

2. METHODS FOR NEUTRONIC ANALYSES

Full analyses for the spent component radionuclide inventory and dose rates are performed for each off-site shipment, containing target vessel or PBW module, with realistic irradiation history and for the time of the shipment. Because these analyses are occurring on regular basis, to facilitate the process, an automated script system that runs the analyses and generates the report, for both target vessel and PBW, was developed.

Three-dimensional radiation transport calculations with the state-of-the-art Monte-Carlo code MCNPX Version 2.7.0 [4] and the latest as-built target station model including the PBW module (Figure 1) are performed to simulate the radiation environment. Specifically, isotope production rates due to spallation reactions and the below-20-MeV neutron fluxes in the 63 group CINDER90 group structure are calculated for target facility components. ENDF/B7.0 is used for table based cross sections. The calculations are performed twice with two different high energy physics models - Bertini and CEM.

Simulated isotope reaction rates and neutron fluxes are extracted from the transport calculation output and are fed into the CINDER90 transmutation code [5, 6] using the standardized ACTIVATION_SCRIPT [7] to calculate the radionuclide inventory of the components. In order to obtain local distributions of radionuclide inventory and subsequent decay source terms, the components are subdivided into small pieces (cells). The target vessel model is represented by 140 cells and the PBW model by 116 cells. Realistic irradiation history and decay time, reconstructed from archived measured accumulated energy of SNS operations, are used in the analyses.

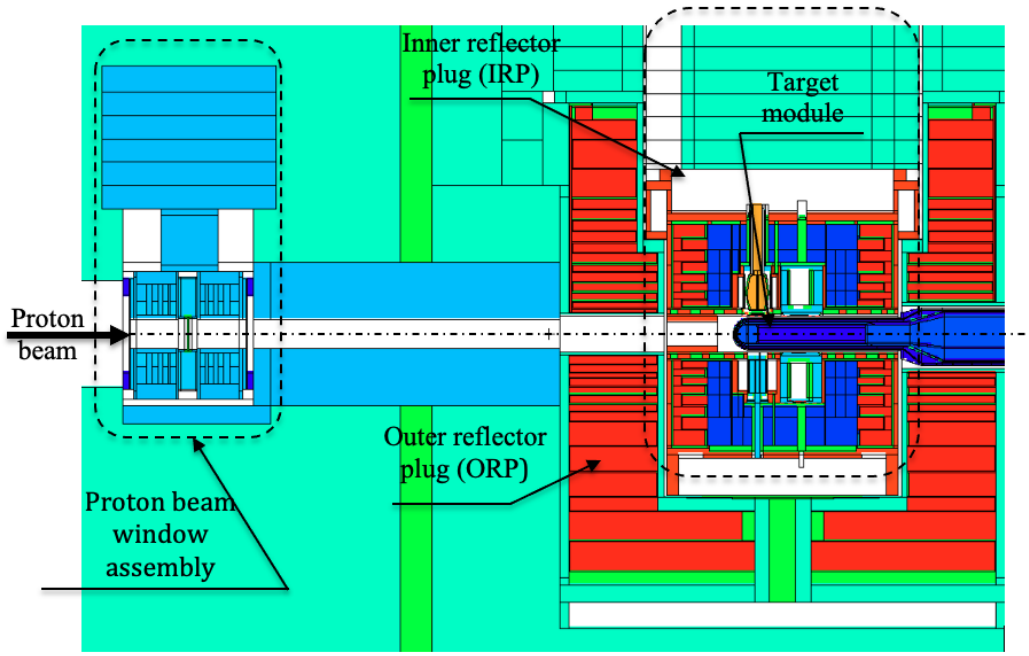

Figure 1. MCNPX model of target station, including PBW module, vertical cross section through the beam center line.

Decay gamma sources for a defined history of build-up and decay are extracted from each cell and compiled to a source term in MCNPX by running the GAMMA_SOURCE_SCRIPT [8]. The decay gamma source is prepared for the time when measurements of the dose rates were performed.

Then the decay gamma source terms are utilized in photon transport calculations for both target vessel and PBW module. To simulate the measurements, cubes of $2x2x2cm^3$ at the detector positions are defined around the components at the locations of the measurements. For the residual dose rate scoring in the detector volume, a track-length flux tally, is applied. Dose rates are obtained by folding fluxes with SNS specific flux to dose conversion coefficients [9].

3. GEOMETRY

For residual dose rate assessment, the models of the target vessel and PBW module are extracted from as-built target station model and are used for the transport analyses. Below is a description of these components.

3.1 Target vessel

The high-energy and high-intensity proton pulses produced by the SNS accelerator strike the target vessel, which is a container for the target material - liquid mercury. The target vessel is located in the middle of the target monolith and heavily shielded. The target vessel material is stainless steel 316L.

When the target vessel is ready for replacement, the mercury and cooling water are drained out. However, we assume, there is still 200 g of activated mercury dispersed in the target vessel. This and next assumption is based on operational experience [10]. In addition, we assume that 10% of the mercury radionuclide inventory (other than mercury, gold and noble gas isotopes) deposited on mercury exposed steel piping uniformly and it is 4.16% (surface fraction of the target vessel relatively the whole loop). For the analyses, this was accounted for by leaving the mercury inside the target vessel at reduced density – 1.253e-4g/cm^3.

The calculational model for the target vessel and detector positions are shown in Figure 2a and 2b, which also shows horizontal and vertical cross sections through the beam center line. The stainless steel vessel is represented by the blue color, dispersed mercury and plated isotopes are represented by the green color, and surrounding the target vessel is air represented by the red color. Detectors are positioned at 0 (surface), 30, and 100 cm distance from the target vessel surfaces along the wide and the narrow sides of the target vessel and under the target nose. Because of the uncertainty of detector position, additional set of analyses are performed with detectors shifted 3 cm away from the component surface.

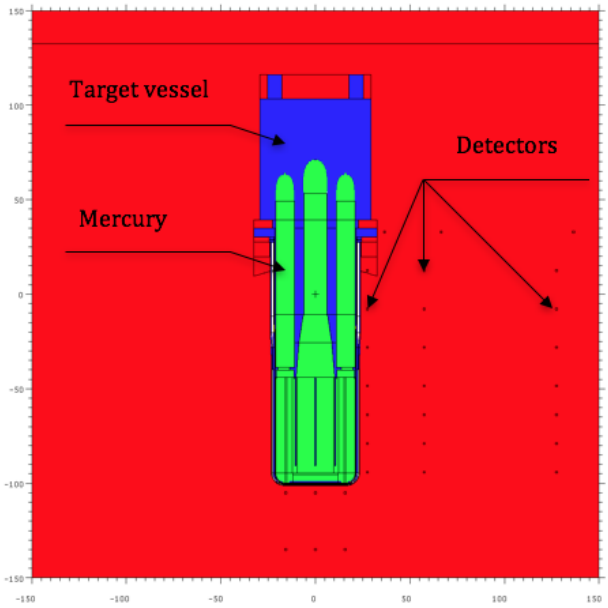

Figure 2a. MCNPX model of target vessel module, horizontal cross section through the beam center line.

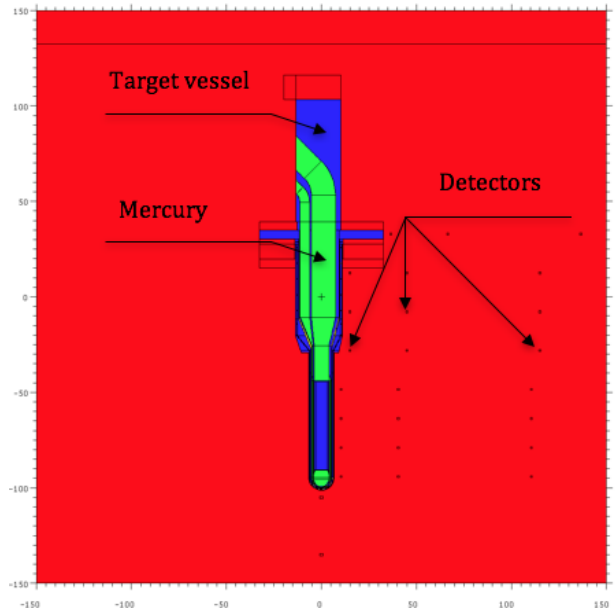

Figure 2b. MCNPX model of target vessel module, vertical cross section through the beam center line.

To analyze the impact of mercury residue inside the target vessel on the calculated dose rates two more geometry modifications are prepared. In one set the mercury is modeled as a puddle on the bottom of the target nose (measurements are conducted with the target nose pointing down), which actually corresponds to the observations during target vessel replacements. In another set, the mercury is completely omitted in the analyses. All the reference detector locations simulated along with the 3 cm shift from the surface for both modified geometries

3.2 PBW

The PBW module is mounted in the target monolith, approximately 2.3 meters upstream from the target. Prior to intercepting the target, the full proton beam passes through the window and about 3% of protons [7] undergo spallation reactions in the Inconel and water of the PBW. Therefore, the PBW receives a significant radiation exposure, leading to high activation in the materials.

The PBW module is comprised of an Inconel-718 window encased in a stainless steel-316 holder and water-cooled collimator assembly. The Inconel window is water cooled and the model consists of 2 layers – 0.1778-cm-thick upstream layer and 0.1524-cm-thick downstream layer. The holder is attached to a steel shield plug. The module weighs 3140 kg. Water lines for cooling the assembly exit the shield plug on top and are shear-cut during PBW module removal activities. These lines along with the interior of the water jacket are dried to a dew point of –35 F with compressed air prior to removing the PBW module for disposal.

The calculational model for the PBW module and detector positions is shown in Figure 3a and 3b. Detectors are positioned at 30-cm-distance from PBW surfaces. In this figure green represents steel, blue represents the mating surfaces of an inflatable seal, and red represents air surrounding the PBW module.

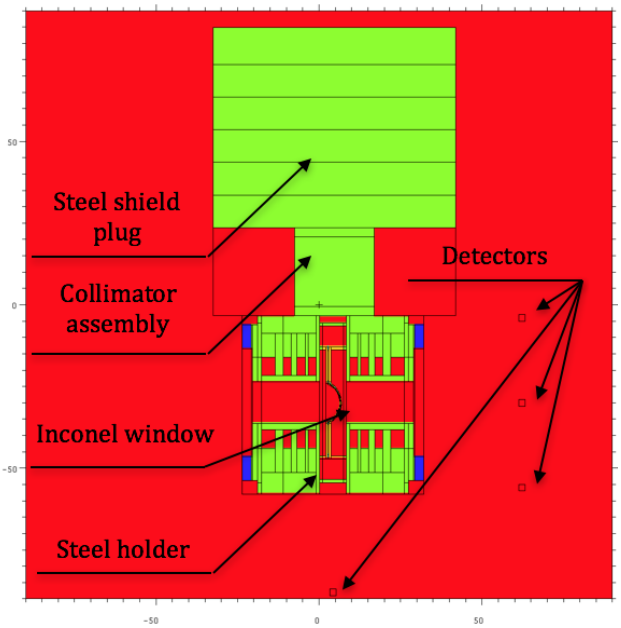

Figure 3a. Calculational model of PBW module, vertical cross section through the beam center line parallel to the beam direction

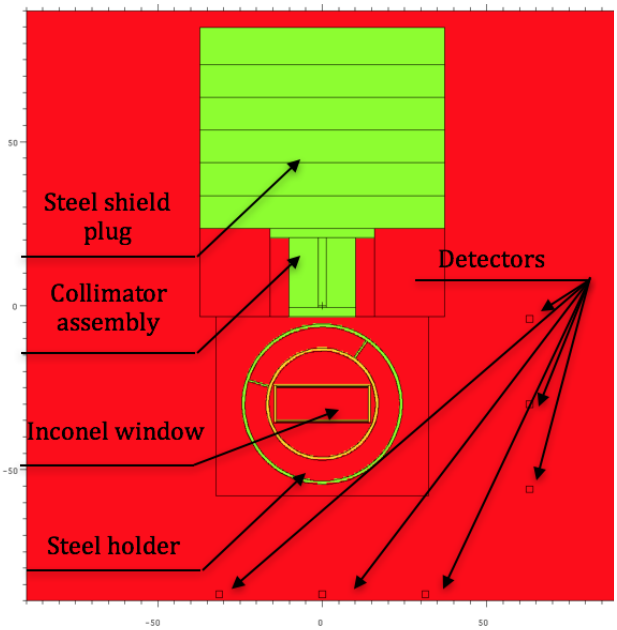

Figure 3b. Calculational model of PBW module, vertical cross section through the beam center line perpendicular to the beam direction.

4. MEASUREMENTS

A specialized high-range radiation detection instrument Ludlum Model 9-7 with 9-7-BH Detector (Figure 4) is used for measurements of the residual dose rates. According to the specification, the detector range is 0.1 – 199.9 Sv/h with a resolution of 0.1 Sv/h and 10% linearity. The active detector volume (ion chamber) is 7cm^3, 2.54-cm-diameter and 1.64-cm-thickness. There is an acrylic cap, 0.9-cm-thick lid a front of the ion camera, which shifts its positioning to about 1 cm from the detector's edge. Modeling the active detector volume directly on the surface and 3-cm-away from the spent structure will bracket the actual ion chamber positioning and covers the uncertainty in detector positioning.

The detector calibration is performed at ORNL with Cs-137 source with maximum dose of 0.63 kR/h. A certificate of calibration is issued in February 2016.

Both spent target vessel and spent PBW modules are highly irradiated, and during the measurements are located in the target service bay. The target vessel in the service bay is handled by a crane to place the target in the measurement position. Suspending the target vessel from the crane allows for a perfectly aligned target with the nose facing down. The remote handling instrumentation is used to position detectors near both spent components. The positioning accuracy is about 1 cm.

Figure 4. Radiation detection instrumentation Ludlum Model 9-7.

Measurements are performed locating the detecting instrument at the component surface, 30-cm, and 100-cm from the spent target vessel surfaces at numerous locations. The detector placement around target vessel #13 30-cm from the surfaces near the wide and narrow side of the target and under the target nose (detectors 17, 18, 19) is shown

in Figure 5. Placement on the surface and at 100-cm from the surface is at the same axial locations but just at decreased/increased distance from the component surface.

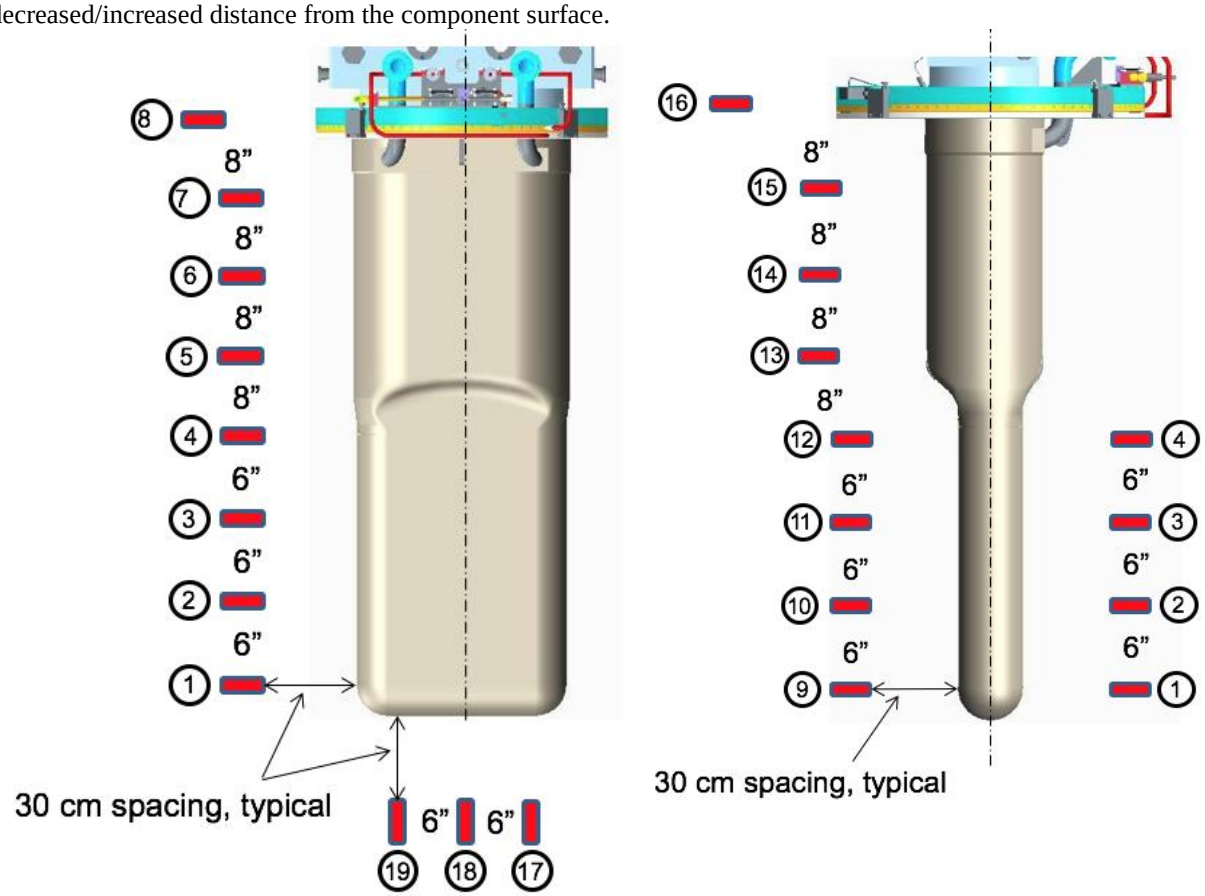

Figure 5. Detector positioning at 30-cm from the surfaces for target vessel #13.

For the PBW module, dose rate measurements are only performed at 30-cm from the surfaces. The detector placement around PBW module #5 is shown in Figure 6.

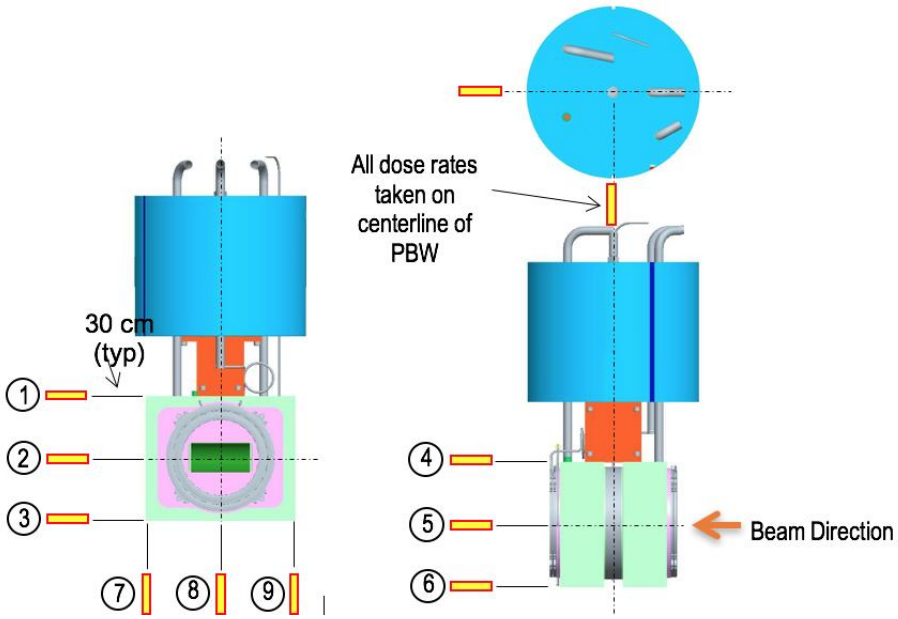

Figure 6. Detector positioning at 30-cm from the surfaces for PBW #5.

5. RESULTS AND DISCUSSIONS

Analyses for residual dose rates are performed for the time of dose rates measurements, which is 106 days cooling down for the target #13 and 80 days cooling down for the PBW module #5 after beam termination on target.

5.1 Target vessel

The service lifetime [7] for target vessel #13 is approximately 0.50 years in which it accumulated slightly more than 2588.55 MWh proton beam energy at 1 GeV proton energy.

The target vessel residual dose rates analyses are performed using two high energy physics models to obtain the isotope production rates and fluxes under 20 MeV neutron energy. Residues of mercury at 200 g, which is left over after the drainage from the target vessel, are modeled in two ways – dispersed inside the target vessel volume, and as a puddle at of the target nose. For each set of measurements, detector at the surface, 30 cm and 100 cm from the spent target vessel surfaces, analyses are done for 4 geometry modifications:

- Detectors are located as described above, mercury is dispersed inside the vessel;
- All the detectors are shifted 3 cm away from the surface, mercury is dispersed inside the vessel;
- All the detectors are shifted 3 cm away from the surface, mercury modeled as puddle at the target vessel nose;
- All the detectors are shifted 3 cm away from the surface, no mercury in the target vessel.

Eight analyses for each detector position are performed. The standard deviation (Monte Carlo uncertainty) for most of the simulated dose rate values was less than 1%, except a few locations, where the standard deviation was slightly higher but lower that 4%. Ratio of the calculated dose rate over measured dose rate is presented in Figures 7, 8 and 9 for detectors location at the surface, 30 cm from the surface, and 100 cm from the surface of the target vessel, respectively. For the measurements performed on the target vessel surface the values for detector positions 1,2, 3 and 18 are above the detector's range.

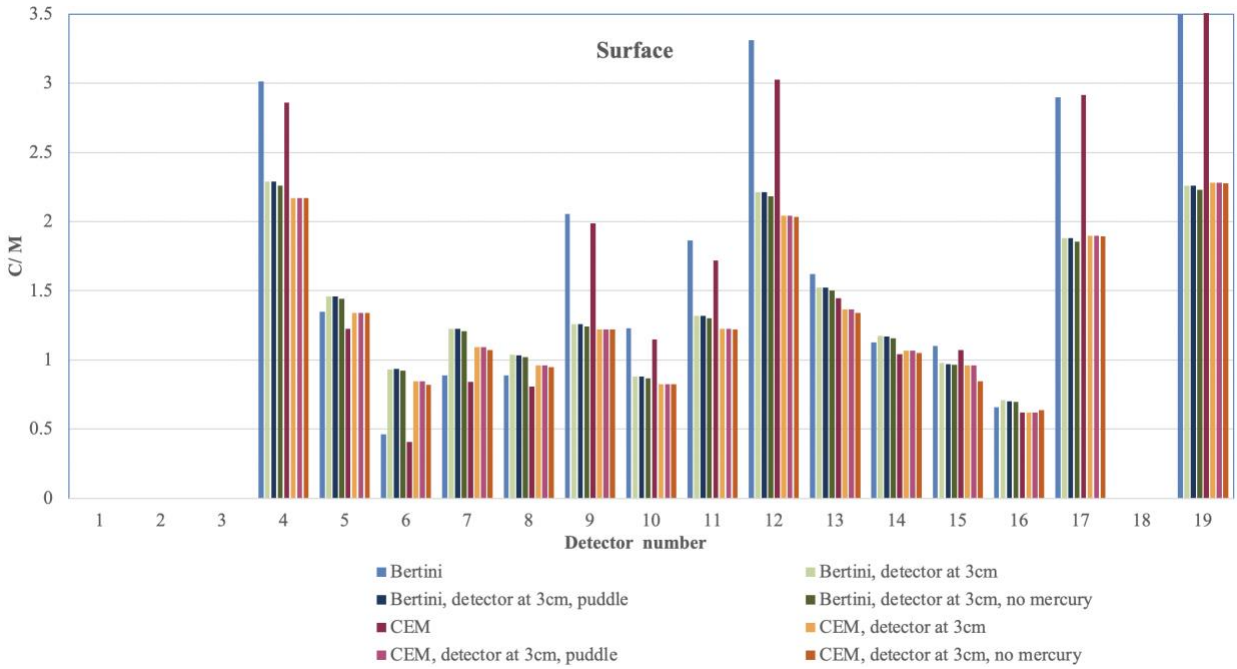

Figure 7. Ratio of the calculated dose rate over measured dose rate (C/M), detectors at the surface and 3 cm from the surface.

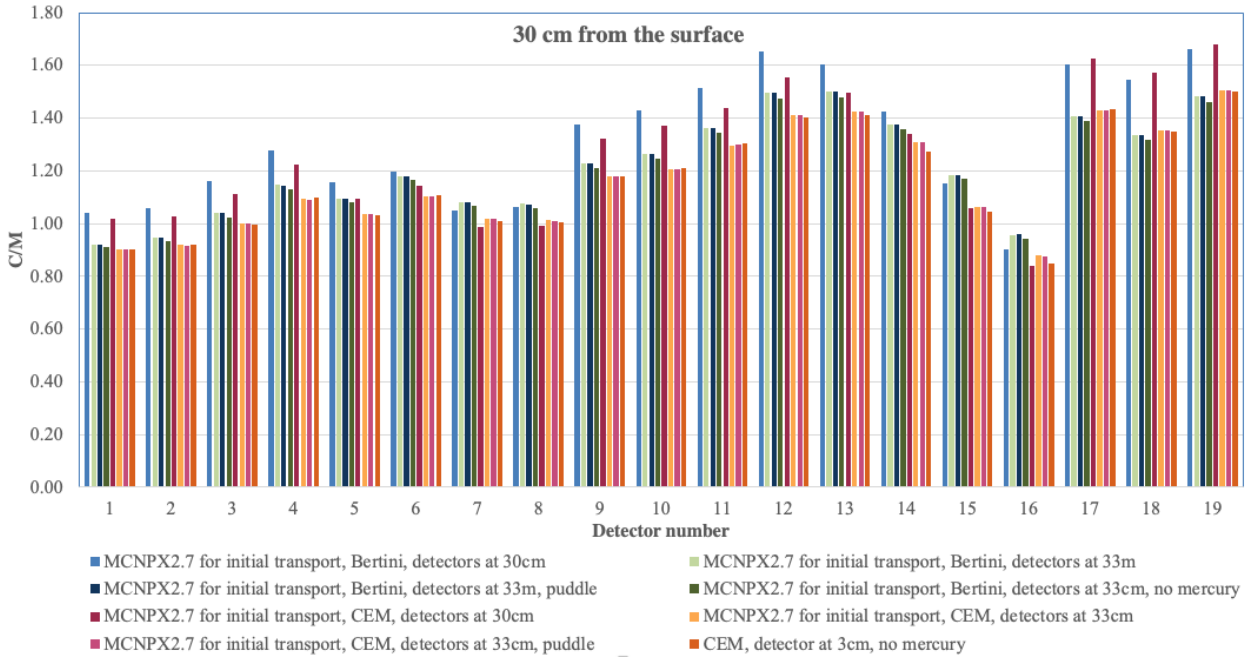

Figure 8. Ratio of the calculated dose rate over measured dose rate (C/M), detectors at 30 cm from the surface and 33 cm from the surface.

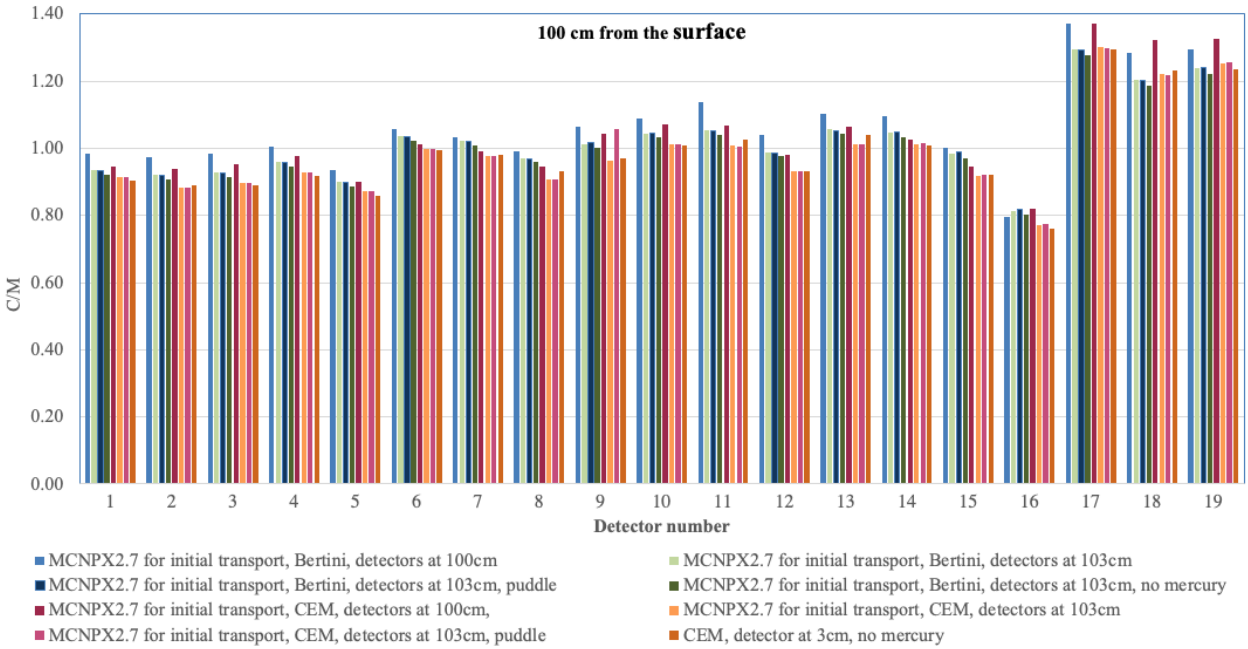

Figure 9 Ratio of the calculated dose rate over measured dose rate (C/M), detectors at 100 cm from the surface and 103 cm from the surface.

Obviously the farther the detector was located from the surface the better the consistency between measured and calculated dose rates, which is explained by uncertainty in the detector positioning. From all three plots, it is clear that a 3 cm shift in the detector position impacts the dose rates. Reasons for this are that the detector body is located approximately 1-cm from the detector cap, the effect of the detector body has not been included, and the detector placement has uncertainty of about 1 cm. For the detector location at the surface this impact is between 10% and 27%, for 30 cm from the surface it is between 4% and 13% and for 100 cm from the surface is between 2% and 6%. Modeling mercury as a puddle versus dispersed mercury or omitted mercury does not impact the analyses, the dose rate difference is less than 1%. Using the Bertini model versus the CEM model to obtain activation reaction rates changes dose rates between 2% and 10%. Along the target vessel dose rate calculated with CEM-based source terms are lower compered to analyses with Bertini-based source terms, the dose rate under the target nose is related in opposite way.

The comparison of measured dose rates versus calculated on the surface is more sensitive to detector positioning uncertainty. This sensitivity should result in a larger scatter of the data, which we see on Figure. 7. Comparison for detectors positioned at 30-cm from the surface and at 100-cm from the surface shows that measured dose rates versus calculated dose rates are within 25% except a few locations, where this difference is up to 50%.

The comparison indicates that the presence / distribution of the mercury really doesn't make much difference. The variation caused by moving the mercury around (or removing it) is significantly less than the overall variance associated with the Monte Carlo statistical uncertainty alone. By contrast, the choice of high-energy physics model for obtaining reaction rates (CEM versus Bertini) is significant, as is the precise detector position. In both of those cases, the difference between the two calculations is much larger than what can be attributed to statistical variation. Results comparisons show that the overall best match is MCNPX2.7 for initial transport, "CEM, detectors at 33cm, no mercury".

5.2 PBW

The PBW#5 module had a service lifetime of approximately 2.34 years in which it accumulated about 12796 MWh proton beam energy at 1GeV proton energy.

Analyses for the dose rates are performed only for one detector positioning. The respective analyses for the PBW module are performed using MCNPX version 2.7 with the Bertini model for simulating the high-energy particles. The standard deviation for most of the calculated dose rate values was less than 3%.

Calculated and measured dose rates (Sv/h*10^6) and ratio of the calculated dose rate over measured dose rate (C/M) are presented in Figure 10. Most of the calculated results are quite consistent with measured data, within 20% and generally higher than measured data. There are two detector locations number 1 and 6, where the calculated dose rates are differing from measured dose rates by about 40%. This detector position is at the edge of the steel holder and moving it up or down may make a large difference in addition to the sensitivity to detector distance to the PBW module surface.

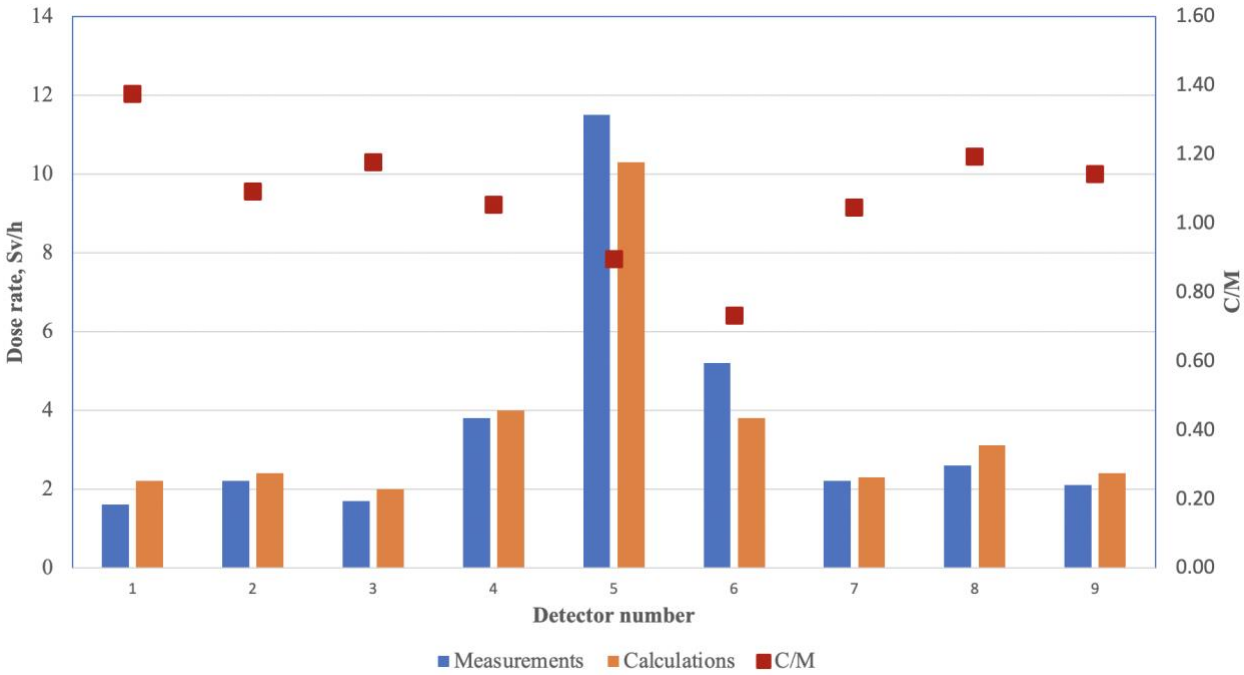

Figure 10 Calculated and measured dose rates (Sv/h*10^6) and ratio of the calculated dose rate over measured dose rate (C/M).

6. CONCLUSIONS

Measurement of residual dose rates from target vessel #13 and PBW module #5 are done and compared to calculated dose rates. For target vessel four geometry modifications are introduced. Reaction rates, which are used for each set of analyses, are calculated using Bertini and CEM high energy physics models.

The measured and calculated contact dose rates from the target vessel show large variations and, as it was shown above, is very sensitive to placement of the detectors. Overall for rest of the locations agreement is good, the difference is less that 50%. Using Bertini versus CEM for obtaining reaction rates shows difference in the dose rates between 2%

and 10%. Due to uncertainty in the detectors measurements and placement it is not possible to judge, which model works better.

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