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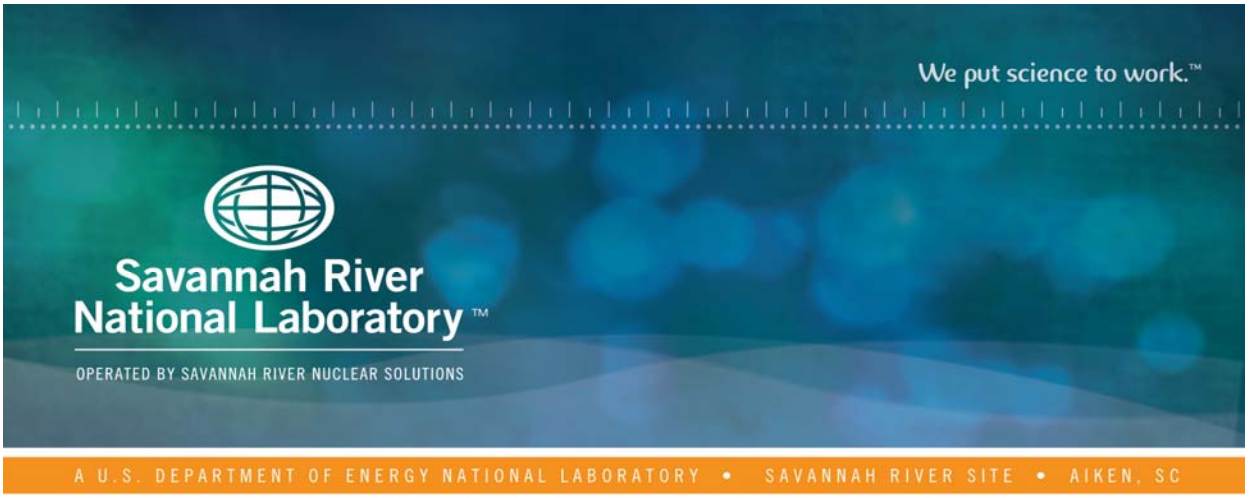

Gamma-Ray Imaging Assay of Cells 3-5 of the East Cell Line in the 235-F Plutonium Fuel Form Facility

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Savannah River National Laboratory™
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EXECUTIVE SUMMARY

In August and September, 2016, scientists from the Savannah River National Laboratory (SRNL) took a series of gamma-ray imaging measurements through the cell windows in front of Cells 3-5 on the east line of the Plutonium Fuel Form (PuFF) Facility using an electrically cooled, high-purity germanium detector. A Germanium Gamma Ray Imager (GeGI) was utilized since it allowed for the location from which the radiation was being emitted to be identified by incoming gamma-ray energy. This measurement technique provided a tool which allowed for the relative concentration of Pu-238 to be mapped throughout each cell. The mapping and new assay data were then used to update the model used in an assay discussed in a 2014 report (SRNL-STI-2014-00629) and to calculate a more accurate value for the holdup in each of the cells [1]. Note that the mapping and new assay data did not replace the previous assay data in the model. Rather, the mapping and new assay data provided additional details on source distribution, which supplemented the previous assay data.

The cell interiors were assayed using two different modes, Compton (a wide angle, less sensitive counting mode) and pinhole (a narrow field of view but lower efficiency counting mode). Compton mode allowed for more of the cell to be quantified in an individual measurement but the resolution of the image created was not as fine as the pinhole method. Data analysis was performed using three gamma-rays emitted by Pu-238 (99.85 keV, 152.7 keV, and 766.4 keV) providing three independent estimates of the relative quantities of Pu-238 holdup in each of the cells. However, the pinhole method was only valid for the 99.85 keV and 152.7 keV peak energies. There weren't enough counts from the 766.4 keV gamma ray to provide statistically meaningful data to image the distribution of the Pu-238 hold-up in the cells. Therefore, a combination of the two modes was used to create the best image possible.

Two main source adjustments were made to the model from 2014. These were adjustments to the amount of Pu-238 on the HEPA filter for each cell and to the amount on the walls of each cell. The model from the 2014 report was rerun based on the new source distribution and it was shown that the concentration on the walls and HEPA filters would not have been quantifiable by the detectors used in the measurements made for the 2014 report. Therefore, the calculated concentration from the GeGI results was combined with the 2014 results to determine the best estimate of Pu-238 holdup in Cells 3-5.

The results of the assay measurements, along with the results from the assays reported in 2014 and in an earlier report from 2006 [3] are found in the table that follows. All uncertainties in this table (as well as

the rest of the report) are given as 1σ. The total holdup in the interior of Cells 3-5 was determined to be **22.5** ± **1.7** grams Pu-238, as compared to the reported 2014 result of **20.8** ± **1.5** grams Pu-238. The difference is due to the added holdup measured on the HEPA filter and walls of the cells.

	Current Assay Mass (g)	**2014 Assay Mass (g)**	**2006 Scoping Assay Mass (g)**
Cell 3 Interior	3.0 ± 0.5 (16%)	2.5 ± 0.4 (16%)	2.17 ± 1.45 (73%)
Cell 4 Interior	10.4 ± 0.7 (7%)	9.6 ± 0.6 (7%)	9.82 ± 7.27 (73%)
Cell 5 Interior	9.1 ± 0.5 (6%)	8.7 ± 0.5 (6%)	4.58 ± 3.25 (73%)
Total Cells 3-5 Interior	22.5 ± 1.7 (8%)	20.8 ± 1.5 (8%)	16.57 ± 8 (50%)

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LIST OF ABBREVIATIONS

Bq	Bequerel (decays per second)
cm	centimeter
D&D	Decontamination and Decommissioning
FWHM	Full-Width at Half-Maximum
HEPA	High-Efficiency Particulate Air (filter)
HPGe	High-Purity Germanium
GeGI	Germanium Gamma-Ray Imager
keV	kilo-electron-Volt
LaBr	Lanthanum Bromide
LANL	Los Alamos National Laboratory
LN	Liquid Nitrogen
MCA	Multi-channel Analyzer
MCNP	Monte-Carlo n-Particle Transport Code
MCNP5	Monte-Carlo n-Particle Transport Code, Version 5
MLD	Minimum Level of Detection
mm	millimeter
NDA	Non-Destructive Assay
PuFF	Plutonium Fuel Form
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
SS	Stainless Steel
WBS	Work Breakdown Structure

1.0 Introduction

The Plutonium Fuel Form (PuFF) facility is located in Building 235-F near the geographic center of the Savannah River Site. The facility was used to produce iridium-encapsulated Pu-238 spheres and pellets for use as radioisotope thermal generators, primarily for the space program. The facility is divided between two cell lines, the East Cell Line used to process the powdered Pu-238 oxide raw material into fuel forms and the West Cell Line used to encapsulate the fuel forms in iridium. Between 1978 and 1984, the PuFF facility processed approximately 165 kilograms of Pu-238. In 1984, the facility was placed in "enhanced readiness mode". During this time, the inert argon atmosphere in the East Cell Line was not maintained. The purpose of the inert argon atmosphere was to prevent corrosion from the high alpha activity of Pu-238. As a result, corrosion soon made the East Cell Line inoperable, particularly the aluminum remote manipulators. The facility has not been decontaminated since the intent was to continue operations and, after the failure of the manipulators, much of the cell interiors are inaccessible [2].

Since the PuFF facility was no longer in an operable state, it was targeted for Decontamination and Decommissioning (D&D). Therefore, a rigorous estimate of the holdup in the facility needed to be determined before the D&D process could move forward. An initial scoping in-situ gamma-ray assay was performed in the PuFF facility in 2006 [3]. The current official estimate of Pu-238 holdup in the facility is based upon these measurements. Using this holdup estimate as a source term, SRS has performed a risk analysis that indicated a seismic event that induces a full-facility fire in 235-F could lead to a 12,100 rem dose to a co-located worker [4]. Based on this risk assessment, SRS is taking steps to decontaminate the facility [5]. One of the first steps taken, as described in the 2014 report, was to improve upon the quality of the in-situ gamma-ray assay data. Carts and collimators were specially designed and used to again survey the equipment in PuFF facility. A LaBr detector was used, along with appropriate analysis software, to measure holdup from underneath the East Cell Line. While the previous scoping work consisted of 32 measurements [3], the 2014 series of assay measurements included nearly 400 measurements, with most of the increase occurring on the East Cell Line [1]. However, these measurements were only made below the cells. With the later removal of two of the windows from each cell; it then became plausible to assay the cells from the window the operators previously used to see into the cells. This measurement direction allowed for the walls and HEPA filters to be measured, something not possible in the 2014 study. This most recent set of measurements in 2016 results in images indicating the locations of the Pu-238 in Cells 3-5 to support the results reached in and compensate for the

limitations seen in the 2014 study. Data analysis for the current set of measurements was conducted with greater rigor. A computer modeling program, Monte Carlo n-Particle 6.1 (MCNP), was used to evaluate a variety of possible physical distributions for the Pu-238 source term in each cell and estimate cross-talk between neighboring cells. This report describes the Cells 3-5 holdup measurements and subsequent data analysis. It is in direct support of the following Work Breakdown Structure (WBS) elements as defined by the Deactivation Project Plan for the 235-F PuFF Facility [5]:

- 01.29.24.01.09.05, Develop Method/Design for Enhanced Characterization, Cells 3-9
- 01.29.24.01.15.04, Perform Enhanced Characterization of Cells 3-5

In accordance with the Deactivation Project Plan, the above WBS elements provide for "enhanced characterization" of Cells 3-5 prior to intrusive deactivation activities such as material removal and decontamination. This report documents one component (exterior measurements coupled with MCNP modeling) of the planned initial, enhanced characterization of Cells 3-5.

2.0 Data Collection

In August and September 2016, scientists from the Savannah River National Laboratory (SRNL) took a series of gamma-ray imaging measurements through the cell windows in front of Cells 3-5 on the east line of the PuFF facility using a Germanium Gamma Ray Imager (GeGI), which is an electrically cooled, high-purity germanium detector. The GeGI was utilized since it allowed for the location from which the radiation was being emitted to be identified by incoming gamma-ray energy. The software for the GeGI was provided by the manufacturer and ran on a supplied tablet PC. Another specialized cart was fabricated for this work to hold the detector in a horizontal orientation, allowing for the detector to be moved inside of the window frame (the outer windows were removed by the project team) to minimize the distance to the inside of the cell. The cart also allowed for the detector system to be pitched vertically and laterally to allow for different areas to be in the field of view of the GeGI when needed. A camera located on the top of the GeGI captured an image of what the GeGI was measuring to align the spectral results with the visual image. The GeGI was wrapped in plastic to prevent it from becoming contaminated when it was moved inside of the window frame. A lead collimator was used for all data acquisitions during the pinhole mode but not the Compton mode. The lead collimator was designed by the vendor (PHDS Co.) who developed the GeGI so that it would correctly interface with the detector system. Multiple Compton (wide angle) and pinhole (narrow field of view) measurements were taken of each cell and were used to validate the results obtained in 2014 as well as determine the concentration in

the HEPA filters and on the walls of the cells, which were not measurable above the minimum level of detection (MLD) in previous assays.

2.1 Confirmation of 2014 Study and HEPA Filter Measurements

In August and September 2016, the GeGI was used to determine the location of holdup inside of Cells 3-5. To do this, a combination of wide angle Compton images and more focused pinhole images were used to determine the intensity of the gamma rays which originated from different locations in the cell. A wide angle image was taken at first and then the areas that yielded a higher intensity than their surroundings were measured again in the pinhole mode for greater accuracy. In total, 9 overnight collections were made to characterize the cells. The goals of these measurements were two-fold: to determine if the distribution of the Pu-238 holdup on the floor of the cell agreed with the 2014 study results, and determine whether there was holdup above the floor which could not be measured accurately by the below-cell 2014 assays. The Compton images produced by the GeGI software for the 3 cells follow.

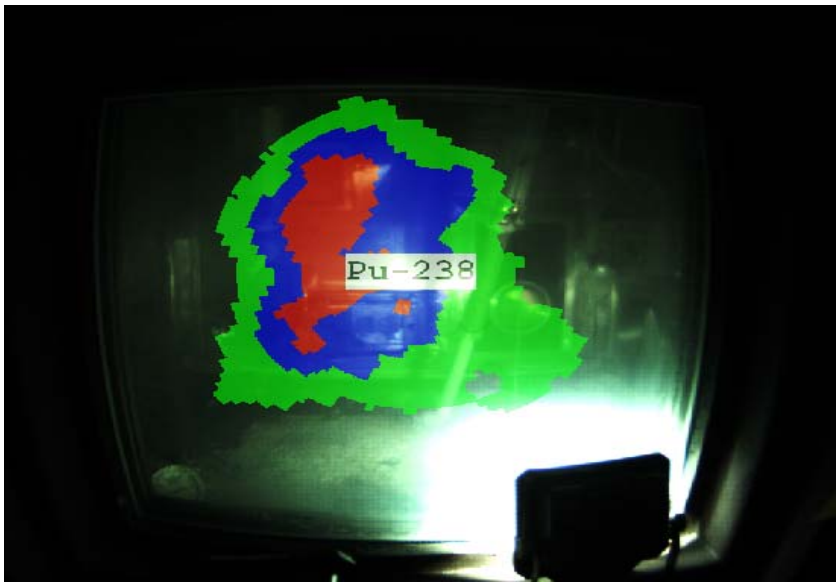

Figure 1: Compton Image of Cell 3

Figure 2: Compton Image of Cell 4

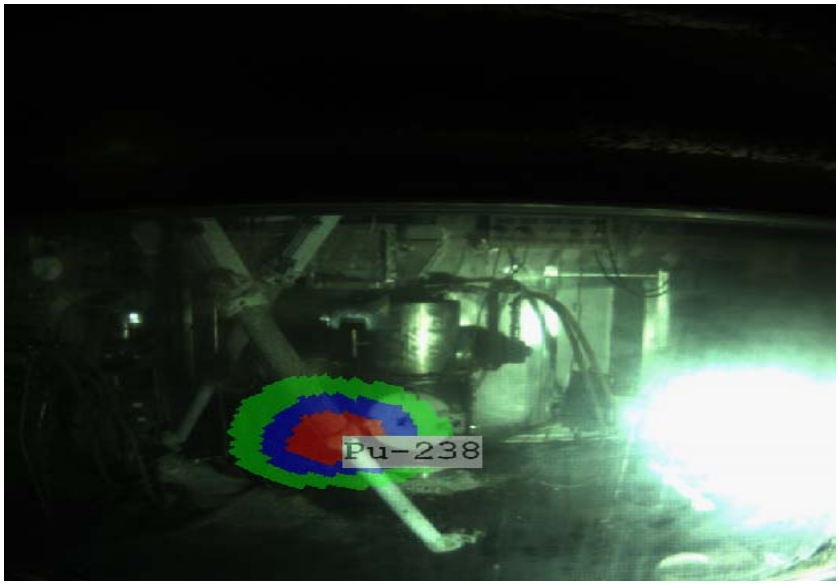

Figure 3: Compton Image of Cell 5

These images helped determine where the pinhole images should be taken for the cells. Two pinhole images were taken of each cell aimed at the more intense areas. These images are shown in Figures 4 - 9. In order to get a composite image of the entire cell from the different areas of the cell measured, combined images were creating using computer code written for that purpose. To create the combined pinhole images, the individual pinhole images from the GeGI were taken and projected onto the cell. The dimensions of the cell were obtained from engineering drawings. The location of the GeGI relative to the

cell was measured by the scientist. The dimensions of the pinhole and detector are taken from the GeGI specifications. Using a computer language, Python, each pixel in the image was projected through the pinhole onto one of the surfaces of the cell (floor or one of the three walls). The resulting composite images with relative gamma-ray intensity maps are shown in Figures 10 – 12. The color scale for the combined pinhole images is the same as for the individual pinhole images .

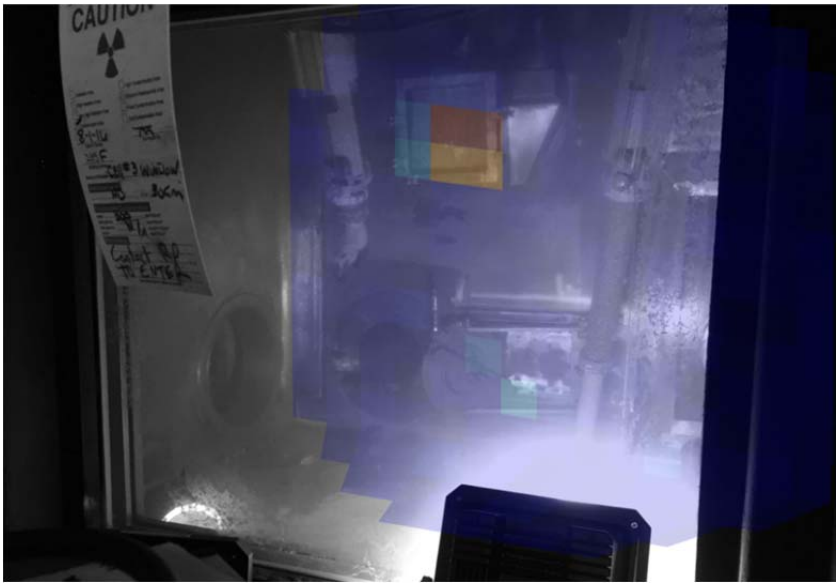

Figure 4: First Pinhole Image of Cell 3

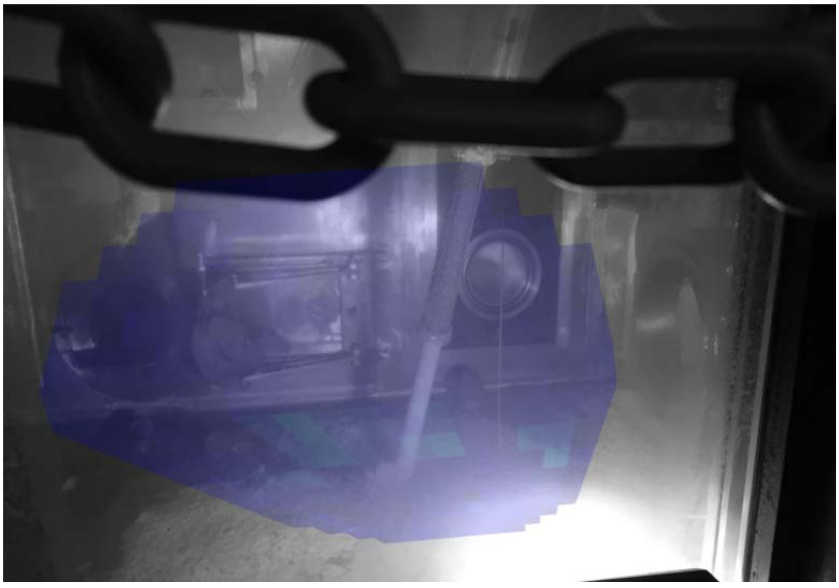

Figure 5: Second Pinhole Image of Cell 3

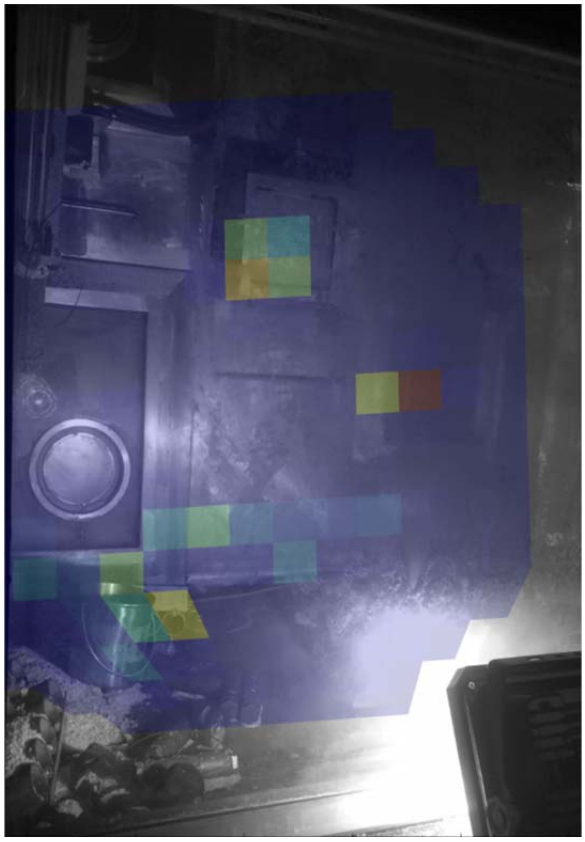

Figure 6: First Pinhole Image of Cell 4

Figure 7: Second Pinhole Image of Cell 4

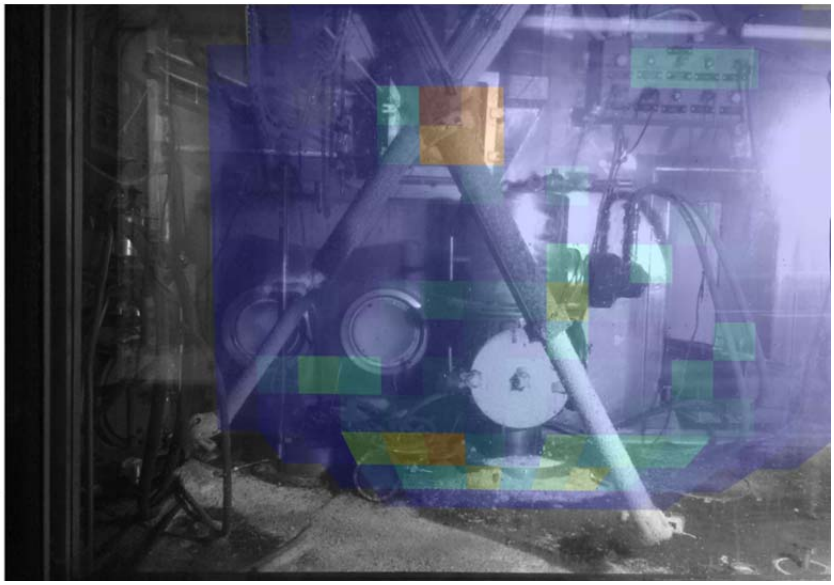

Figure 8: First Pinhole Image of Cell 5

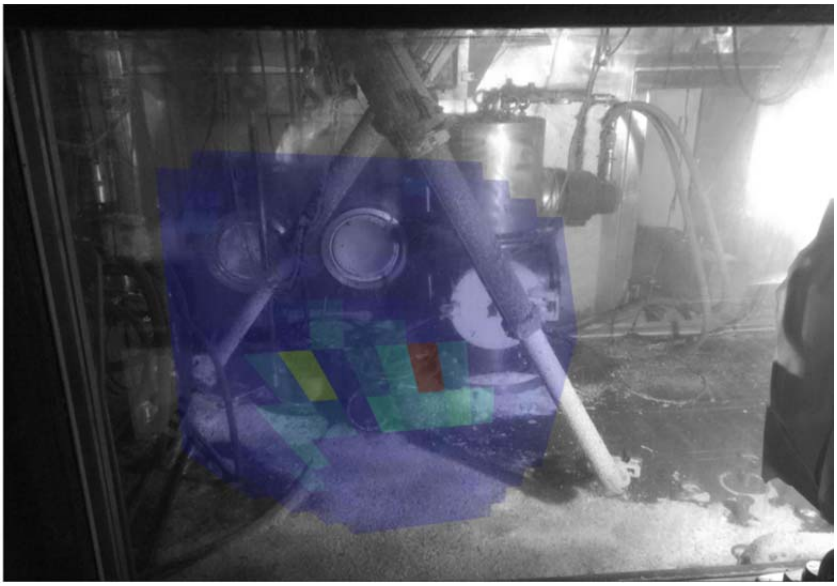

Figure 9: Second Pinhole Image of Cell 5

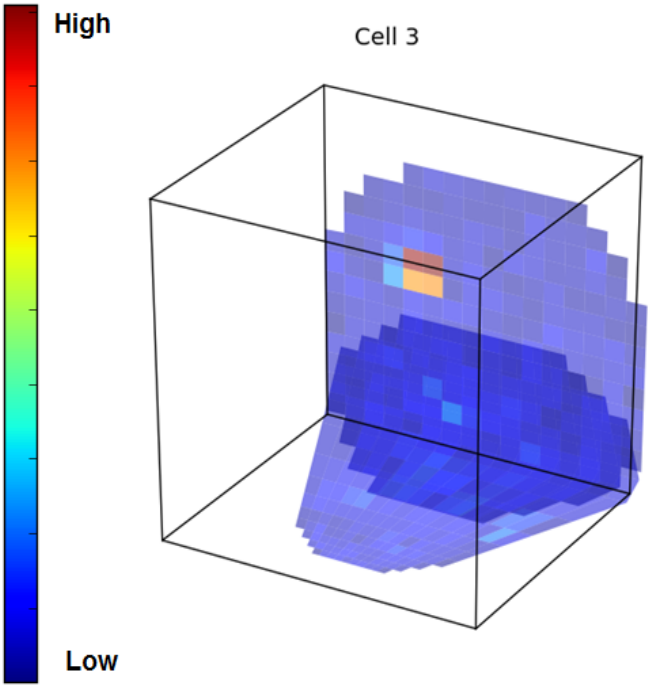

Figure 10: Combined Pinhole Results for Relative Gamma Intensity from GeGI for Cell 3

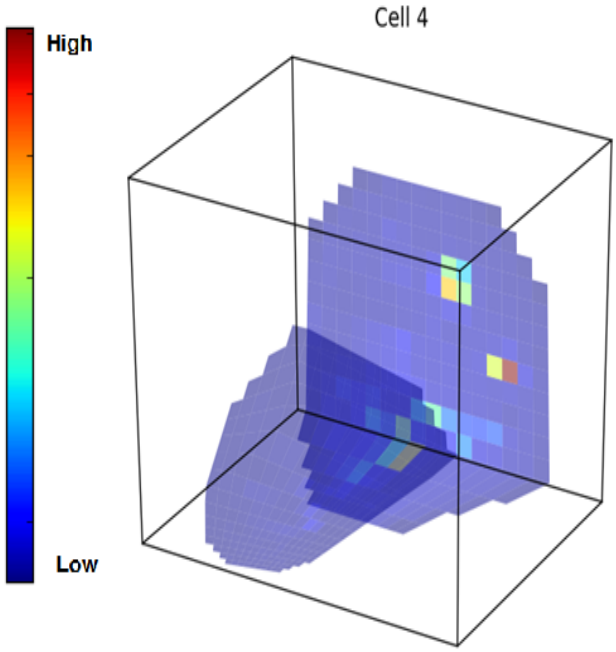

Figure 11: Combined Pinhole Results for Relative Gamma Intensity from GeGI for Cell 4

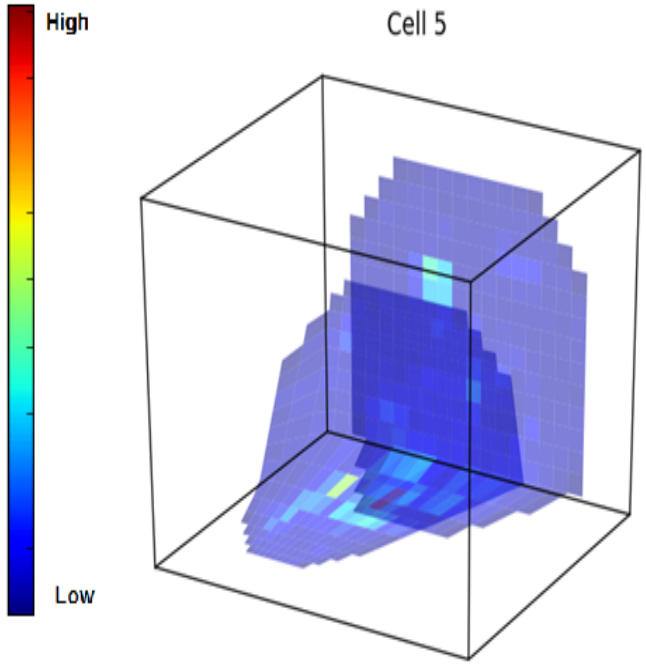

Figure 12: Combined Pinhole Results for Relative Gamma Intensity from GeGI for Cell 5

The numerical results from all 9 images (1 Compton and 2 pinhole per cell for all 3 cells) were compiled and used to calculate the relative concentration for each area in each cell. The areas with the largest intensity on the floor of the cell from the GeGI measurements correlated with the 2014 study and the only area of significant contribution not on the floor was from the HEPA in each cell. While the walls did not contain activities above the MLD, it was assumed that the MLD concentration determined for the walls was present on all locations on the back and side walls to be conservative in our estimations. The relative intensities are shown pictorially in Figure 13. The relative intensities of Pu-238 are given in Tables 1-3.

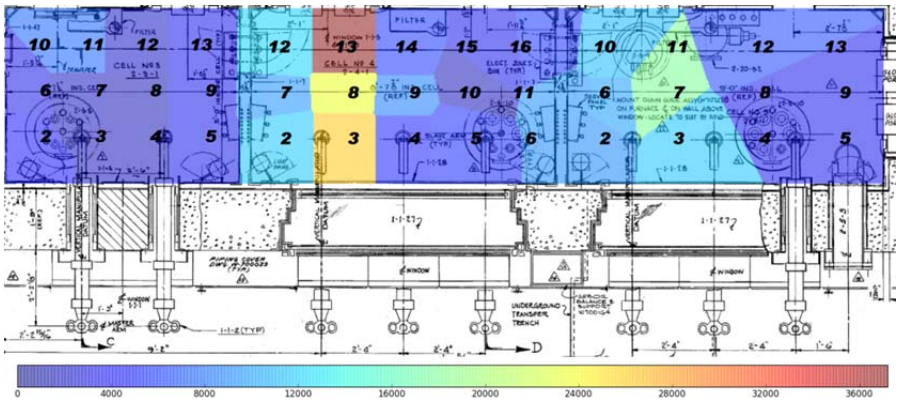

Figure 13: Relative Pu-238 Concentrations Based on GeGI results and the 2014 study

Table 1: Relative Intensity of GeGI Results Based on Location in the Cell for Cell 3

Floor Location	% of Counts
2	7.7%
3	4.9%
4	3.2%
5	6.4%
6	6.3%
7	3.9%
8	3.0%
9	6.5%
10	10.3%
11	14.2%
12	4.0%
13	6.6%
HEPA	22.9%
Total	100.0%

Table 2: Relative Intensity of GeGI Results Based on Location in the Cell for Cell 4

Floor Location	% of Counts
2	6.8%
3	15.0%
4	2.9%
5	3.0%
6	3.2%
7	5.5%
8	13.7%
9	5.0%
10	1.0%
11	2.2%
12	7.1%
13	21.5%
14	3.4%
15	2.0%
16	2.7%
HEPA	4.9%

Total	100.0%

In Cell 4, one other location displayed sufficient gamma-ray emissions to also warrant consideration in the total distribution in the cell. The Cell 4 wing cabinet emissions were strong enough to make an impact on the GeGI holdup results but would not have significantly impacted the below-cell gamma detectors used in the 2014 study. The Pu-238 content in the wing cabinets was large enough that it represented 6.4% of the Pu-238 signal measured by the GeGI for the content in Cell 4. The wing cabinets are areas, separate from the cells, where work could be done to assist the corresponding cell. For the current work, wing cabinet emissions were not used in the total calculation of the holdup inside of the cell, since the emissions were from sources in the cabinet and not the cell itself.

Table 3: Relative Intensity of GeGI Results Based on Location in the Cell for Cell 5

Floor Location	% of Counts
2	9.0%
3	11.3%
4	3.5%
5	3.5%
6	9.5%
7	17.4%
8	4.9%
9	3.5%
10	10.9%
11	14.1%
12	6.3%
13	4.9%
HEPA	0.8%
High Panel*	0.4%
Total	100.0%

*Not used in overall calculation of cell distribution

The Cell 5 wing cabinet was never placed into service and was never connected to Cell 5 which is why the GeGI did not detect any gamma rays with energies characteristic of Pu-238 coming from the pass

through between the wing cabinet and the cell. The high panel in Cell 5 had a contribution of only 0.4% , which did not significantly change the overall result. However, it was included for posterity.

The MCNP models used in 2014 were repeated using this new distribution to determine the effect of the HEPA holdup on the overall number of counts seen by the detectors below the cells. The addition of the HEPA caused less than a 1% difference in Cells 4 and 5. The impact for Cell 3 was a larger, less than 3%, difference. This is because its holdup was a larger percentage of the total. This modeling shows that the geometric attenuation due to the location of the HEPA far above the floor made the HEPA effectively hidden from the view of the below-cell detector configuration used in 2014. This same effect was also seen for the MLD level of concentration assumed to be on the walls. The MLD on the walls was determined by taking a representative sample of the wall using GeGI and using that value to assume all the walls had the same amount of Pu-238 on them. This is quite conservative since the area selected was the location with the largest holdup on the wall. To account for this new holdup due to the HEPA and walls, the results from the 2014 assay were adjusted to include this holdup based on the ratio between the floor, HEPA and wall values. The errors were also adjusted accordingly.

The results from the GeGI data collections support the 2014 measurements presented in SRNL-STI-2014-00629 with respect to the floor holdup. This study confirms the validity of those measurements and allows the current measurement team to confidently use those results to determine the holdup reported in the current report.

3.0 Assay Results

The mass of Pu-238 for each cell is given in the following table.

Table 4: Current Assay Results Compared with the 2014 Study and the 2006 Scoping Measurements

	Current Assay Mass (g)	2014 Assay Mass (g)	2006 Scoping Assay Mass (g)
Cell 3 Interior	3.0 ± 0.5 (16%)	2.5 ± 0.4 (16%)	2.17 ± 1.45 (73%)
Cell 4 Interior	10.4 ± 0.7 (7%)	9.6 ± 0.6 (7%)	9.82 ± 7.27 (73%)
Cell 5 Interior	9.1 ± 0.5 (6%)	8.7 ± 0.5 (6%)	4.58 ± 3.25 (73%)
Total Cells 3-5 Interior	22.5 ± 1.7 (8%)	20.8 ± 1.5 (8%)	16.57 ± 8 (50%)

With the inclusion of the HEPA and wall concentrations, the holdup increased by 8.17% from the 2014 measurements. This was done by summing the total measured previously in 2014 with the ratios determined for the HEPA and walls using the GeGI. The percentage of the total holdup in the cell is also included to help the reader ascertain the concentration in each section of the cell. Using the values reported in 2014 as well as the GeGI results, the percentage of the concentration in each cell is estimated to be distributed as:

Table 5: Distribution of Pu-238 in Cells 3-5

	Cell 3 (g Pu-238)	**% of Total**	**Cell 4 (g Pu-238)**	**% of Total**	**Cell 5 (g Pu-238)**	**% of Total**
Total on floor	2.2	72.3%	9.3	89.3%	8.5	93.1%
Total on HEPA	0.6	21.5%	0.5	4.6%	0.1	0.8%
Max on walls	0.2	6.2%	0.6	6.1%	0.5	6.1%

As expected, Cell 3 displayed lower holdup inventory than the other two cells measured. This expectation was based on the operation of the PuFF facility and the work performed in each of the individual cells. Therefore, a higher percentage of the holdup is in the HEPA filter compared to the rest of the cell it occupies as compared to Cells 4 and 5.

4.0 References

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