Contract No:

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LDRD-2019-00144
LDRD External Report Summary
SRNL-MS-2019-00185

Title: Large Area Airborne Contamination Monitoring

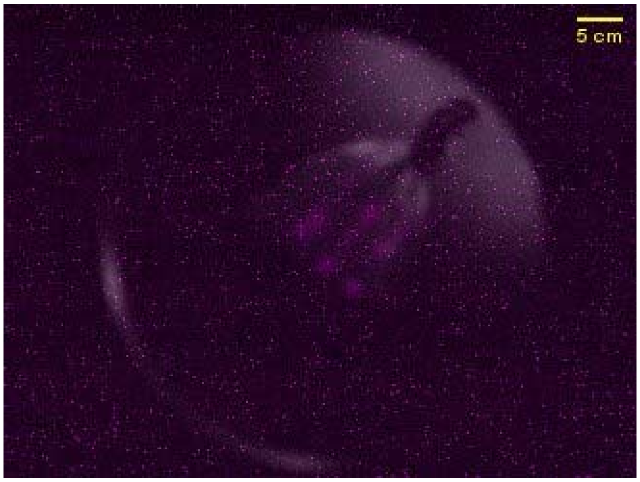

Figure 1 Image of a 3x3 array of Pu-238 particles buried in folds of a section of HEPA filter.

Current D&D operations at Hanford have demonstrated a flaw in the current state of the art capability of defining airborne contamination boundaries. Airborne particulate emissions of Pu-239 from CM2H operations on the Hanford Pu Finishing Plant were detected well beyond areas controlled for airborne plutonium, putting numerous workers at risk for radiological assimilations. Forty-two Hanford workers had positive bioassay results for plutonium uptake. Work areas determined to be free of airborne contamination as measured by state-of-the-art continuous alpha monitoring (CAM) systems were later discovered not to be. Airborne contamination was scattered to a such degree that CAM systems were surveying insufficient volumes of air to accurately predict where respiratory protection was required. A $19,000 state of the art portable CAM system used at both Hanford and at SRS screens air for alpha activity at the rate of 1 CFM. This research project will explore the feasibility of using commercially available air purifying systems to screen significantly larger volumes of air for airborne actinide contamination. Commercial household HEPA based air purifying units screen large volumes of air, a simple $300 unit being looked at for this study could screen a volume of air that would require over 300 of the $19,000 CAMs. The goal of the project was to determine the feasibility of building a detection system capable of screening the air purifiers pre-filters and HEPA filters for any actinide bearing hot particles that have been captured. The simplest technologies available to Field-Radiological-Protection personnel are hand-held alpha field survey meters. With the size and the corrugated structure of these HEPA filters, hand screening is not practical. This project researched alternatives capable of automating this function.

Two potential technologies were examined. One technology evaluated was the measurement of x-ray emissions from the actinides, taking advantage of the higher emission rates of the L x-rays compared to the gamma emissions of isotopes such as Pu-238 or Pu-239. High resolution to low resolution systems were evaluated. Branching ratio data on the actinides in the literature is incomplete, and a study was initiated to measure branching ratios of the various L emissions of a number of the plutonium isotopes. A Monte Carlo simulation was also run to calculate the effect the attenuating HEPA media would have on x-ray measurements on spectrometers centered in each quadrant above the surface of the HEPA. The second technology evaluated was measuring the light from a scintillating media applied to the surface of the HEPA filter using a ThorLabs Scientific-Grade Digital Camera Model 8051M. A light tight enclosure capable of holding the 2’x2’x1” HEPA filter was fabricated. Numerous scintillation media were evaluated for both the sensitivity and for resolution of measuring alpha activity. Figure 1 shows images of a 4”x4” section of HEPA filter with a 3 x 3 grid of Pu-238 contamination loaded inside the crevices of the HEPA.

Awards and Recognition

1 presentation on aspects of this research given at a conference, with 2 more planned

Intellectual Property Review

This report has been reviewed by SRNL Legal Counsel for intellectual property considerations and is approved to be publicly published in its current form.

SRNL Legal Signature

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Signature **Date**

LDRD-2019-00144
LDRD Report
SRNL-MS-2019-00185

Title: Large Area Contamination Monitoring

Project Team: D. P. DiPrete (Primary), K. M. Fenker, T. S. Whiteside, A Brand, A. Jolie

Thrust Area: ES

Project Start Date: October 1, 2019
Project End Date: September 30, 2019

Current D&D operations at Hanford have demonstrated a flaw in the current state of the art capability of defining airborne contamination boundaries. Rapid analysis of large volumes of air to more accurately measure airborne contamination could reduce personnel risk and speed decontamination and decommissioning activities across the DOE complex and beyond.. This work meshes with the DOE-EM mission to safely conduct radiological clean-up operations.

This project's overall goal was to study the feasibility of rapidly determining airborne alpha contamination using inexpensive high-volume household air purifiers. As opposed to conventional continuos alpha monitors which deposit alpha activity on simple filter papers, these air-purifiers deposit activity on large complex HEPA filter surfaces. This project has explored the feasibility of using various detectors and scintillators to rapidly analyze these large, complex samples for alpha contamination. This included using a CCD-based camera to detect scintillations caused by alpha-particles interacting with ZnS(Ag) plates, ZnS(Cu) powder, and multiple organic scintillators. It also included evaluating multiple X-ray spectrometers to examine the sensitivity of these instruments with this complex sample type. A study of the distribution of radon and its progeny onto these media was conducted. This work outlined an approach for analyzing CCD-based image data into actionable information.

FY2019 Objectives

- Develop a cost-effective method to screen large volumes of air for actinide airborne contamination.
- Evaluate activated carbon and HEPA filters for radon removal from air volume samples
- Determine how quickly and easily various detection systems can determine the presence of actinide contamination on the filters
- Evaluate X-ray spectrometers (NaI and SrI2) and compare to U-LEGE
- Evaluated sensitivities of scintillation systems measured by light vs what conventional solid state alpha spectrometers and ZnS gross alpha scalars can do
- Gather data for which system or systems will work best in an array to rapidly analyze filters
- Determine depth particles penetrate the HEPA filter – We did not deploy the Air Purifiers into airborne-contaminated SRS facilities so we couldn't satisfy this objective

Introduction

Decontamination and decommissioning (D&D) activities at the Hanford Pu Finishing Plant were significantly set back by the inability of the contractor to protect the workforce when faced with an unusual dispersion of airborne activity combined with the environmental conditions found at the Hanford Site. The Department of Energy reached out to SRNL as well as across the complex for help with the airborne monitoring effort, but technologies weren't available to significantly bolster the efforts that were currently underway. The event was controlled eventually by numerous applications of chemical fixative on the source term, but all D&D activities were suspended for a significant period of time. Providing solutions to such situations is a perfect fit for SRNL, DOE's EM National Laboratory. The situation at Hanford will likely have repercussions across the DOE complex in other situations where DOE faces D&D challenges. Finding enhancements to technologies in the toolbox that the DOE community currently has for airborne monitoring in challenging situations will augment SRNL's reputation as a National Laboratory capable of providing technical solutions to DOEs radiological clean-up efforts. Challenges facing Hanford in D&Ding its plutonium legacy facilities will be challenges SRS will share as it tackles the D&D challenges of its F-Area Plutonium legacy facilities. Upon successful completion of this scoping effort, additional funding opportunities will be sought from funding sources such as the Hanford Grand Challenges program, or DOE programs such as NNSA NA-22 or 241. .

Approach

At the project onset, it was determined the most promising approach to rapidly screen large surface area samples for alpha activity would be to contact the surface of the media with a scintillator but provide an offset from the light collection and the actual scintillator. So rather than pursue the approach of using photomultiplier tubes or multiple avalanche photodiodes, a ThorLabs Scientific-Grade Digital Camera Model 8051Mwas procured and evaluated. In the early stages of the project, a small light tight box to enclose the camera and a scintillating sample was put together. The system was first evaluated with scintillating paint to ensure functionality.

Plates of ZnS phosphor, bound onto a plastic light pipe backing were procured. Such plates are what are connected to photomultiplier tubes in the SRS RPD hand-held alpha survey meters. High activity plates of Pu-238 (~1E7 dpm) were prepared in the Nuclear Measurements radiochemistry labs and sealed in contact with a piece of ZnS(Ag) plate. The Pu-238 activated ZnS(Ag) plates were then measured in the light tight box to successfully establish proof-of-principle.

A sensitivity study was then conducted to establish whether this technology was practical. A series of stainless steel planchets ranging from 1E2 to 1E5 dpm were prepared. Each planchet was sealed with a piece of ZnS plate in contact and measured with the light tight box. Algorithms were generated to sum multiple exposures with the camera to increase the sensitivity of the system. Numerous improvements with the light tight box were carried out before it could successfully measure the 1E2 dpm sample.

The design of a light-tight container that could hold both a HEPA filter in a drawer slide and the mounted digital camera was contracted out to EES to be constructed. Construction of the light-tight container took roughly 5 months, and numerous additions were made over the next couple of months to make the box light tight. The box was then evaluated using the standards prepared initially for the small protype box. Once sensitivity was established, experiments were conducted to evaluate other scintillator systems.

The ZnS(Ag) plates were evaluated against ZnS(Cu) powder, Perkin-Elmer Enhance, and Utlima Gold AB liquid scintillation cocktail. ZnS(Cu) is a similar scintillator to what was on the plates. It has a Cu activator, not a Ag activator, so it emits light at a slightly different wavelength. However, this system is a powder, not a plate, so it can be applied to uneven surfaces and be in direct contact with the alpha emitters, as opposed to the ZnS(Ag) plates, which would be separated from the alpha activity by some distance on complex surface structures such as a HEPA filter. That distance thru air would heavily attenuate alpha emissions prior to reaching the scintillator. A third scintillating media studied was Perkin Elmer Enhance. Perkin Elmer Enhance is a spray-on scintillator used in the medical research community to study the behaviors of biological systems labeled with various beta emitters. The fluors in this system are POPOP and PPO, the classic organic scintillators of historical liquid scintillation cocktails. The final scintillation system studied was the environmentally friendly liquid scintillation cocktail Perkin Elmer Ultima Gold AB used heavily by the Nuclear Measurements Group's radiochemistry team. Three different activities of Pu-238, 1E5, 1E4, and 1E3 dpm were prepared for each scintillation system and were evaluated by the camera system.

Of all the scintillation systems studied, only the Perkin Elmer Enhance system completely failed. A final study was undertaken where 4"x4" portions of the Air Purifying Units HEPA filters were cut out. For each section, KimWipes were compressed into small balls and were each spiked with Pu-238 at ~1E5 dpm. The Pu-238 was dried onto the KimWipe balls, and the balls were inserted into the folds of the HEPA filter in a 3 by 3 pattern. One section had a ZnS(Ag) plate affixed to the surface. One section was dusted with ZnS(Cu) powder. One section has Ultima Gold AB Liquid scintillation cocktail applied to each area containing a KimWipe.

Concurrent to the scintillation studies a feasibility study was conducted looking at the potential for x-ray emissions for the detection of alpha activity. A couple of configurations were considered. A single x-ray spectrometer fixed above the HEPA filter in the manner of the camera was deemed not practical. The solid angle would be so large at a distance sufficient to measure the entire filter by one spectrometer would be too insensitive for any practical measurements. To load a device with a large number of spectrometers closely coupled with each section of the HEPA would be cost prohibitive. One detector put into a robotic systems where it is used to scan across the entire surface area of the HEPA filter would be too time intensive. A compromise system was envisioned where one spectrometer was centered over each quadrant so having 4 spectrometers in total. A Monte Carlo N-Particle mathematical model was built to study the counting efficiencies of such a system, and how those efficiencies changed for a particle buried in different areas of the quadrant. Several detection systems were looked at, a thin windowed SrI detector, a thin windowed NaI detector, and a high resolution HPGe thin windowed planar spectrometer. In addition to the x-ray detector study, a series of plates of various Plutonium Isotopes were prepared and analyzed in an attempt to add to the lack of knowledge in the available Nuclear Datasets on actinide x-ray emissions, namely those for Pu-238, Pu-239 and Pu-240.

The final area of study was a short look on where Radon and it's progeny were depositing in the high volume Whirlpool Whispure Air Purifiers purchased for this project. The Whirlpool Whispure Air Purifiers have a HEPA that is particularly suitable for this project, being flat not round, relatively thin at 1", and not too large at 2"x2". In addition to the HEPA particle filters the Air Purifiers have a thin activated charcoal mesh designed to remove household odors. A short study was conducted seeing where alpha activity showed up in this system when the air purifiers were run in non-radiological areas. The alpha activity was measured using a hand-held ARW alpha spectrometer. A HPGe spectrometer was also deployed in an attempt to get some radiological isotope identification as well.

An air purifying system was delivered to 235-F in the Spring, but while the facility personnel were willing, it became too much of a hurdle with Area RPD management to get the device deployed in an airborne area to generate a real world sample for analysis.

Results/Discussion

Results of the Radon Study indicated Radon or more likely its progeny deposited on the activated charcoal odor screen, not the HEPA filter itself. These measurements were made using the ARW alpha spectrometer. Identification of the isotopes captured using a HPGe detector were unsuccessful due to the high ambient background of the natural decay products that the attempt was being made to measure. An algorithm was developed to address Radon rejection by decaying measured light over a number of pictures summed in time to see if excess light would remain. This algorithm is addressed in reference SRNL-STI-2019-00599.

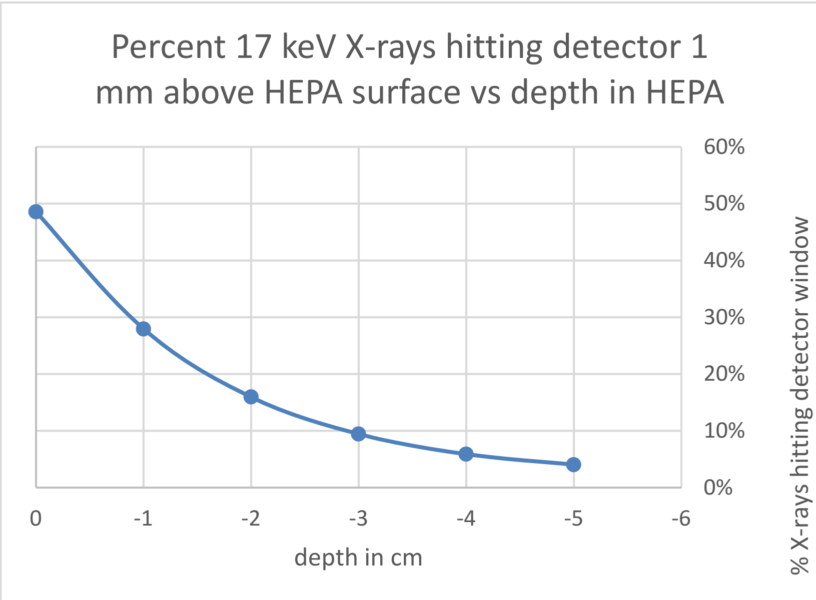

Figure 2 MCNP Model of x-rays Reaching Detector Window from HEPA Surface

The x-ray studies generated less promise than the scintillation studies. The MCNP analysis looked at the percentage of 17 keV x-rays that would reach a spectrometer set 1 mm above the HEPA, looking at only ¼ of the entire HEPA filter. The model showed that even with 4 detectors in place in a system, the counting efficiencies would preclude this approach leading to a rapid, highly sensitive alpha analysis of a HEPA filter system. More than 4 detectors would lead to a cost prohibitive system. The results of the MCNP model are provided in Figure 2. The x-ray studies did provide some nuclear data on x-ray emissions of a number of actinides that may be publishable and are still being evaluated and will be addressed further in SRNL-STI-2019-00598

The scintillation system did show promise for making this a real-world system, and although we made many measurements there is still some substantial work remaining to bring this to fruition. Figure 3 has a picture of the HEPA assay device prototype fabricated for this project.

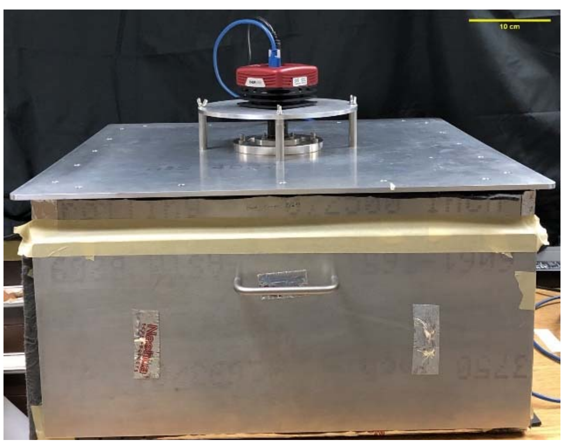

Figure 3 Light tight box with camera and lens mounted on top. Box is sized to be able to image area of 19 in x 19 in. Max distance between camera lens and sample is 7 in.

We were able to image Pu-238 at various activities (e7-e3 dpm) using a solid sheet of ZnS(Ag) scintillator and the CCD-based digital camera. These images can distinguish the location of the Pu-238 on the substrate where they are deposited. Figure 4 provides an example of some of the sensitivity measurements where an array of plutonium plates from 1E5 to 1E2 dpm were placed inside the assay system and measured.

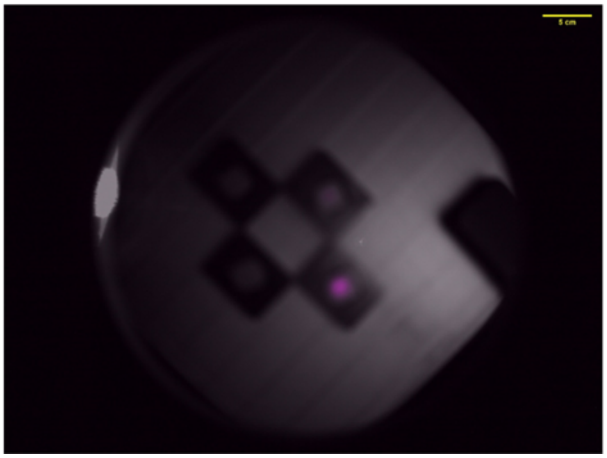

Figure 4. Solid Zn(S) on top of plates on top of filter, activities range from 1E5, 1E4, 1E3, 1E2 dpm (counter-clockwise from bottom right), 1x1 binning. Scintillation shown in magenta.

To increase sensitivity of the system, an algorithm was developed to sum a number of exposures. To acquire images, a ThorLabs Scientific-Grade Digital Camera Model 8051M was used, with a Kowa LM5JC10M Lens. Unfortunately, this lens has a 2/3" optical format while the camera has a 4/3" optical format, this causes vignetting, which is when a dark ring appears around the borders of the image in Figure 4. While not ideal, this did provide a constant "no-information" background against which to compare the dark current. To connect the lens to the camera the installed IR filter on the camera was removed. This camera has a peak quantum efficiency of 51% near 450 nm. This corresponds well with the Zn(S) emission spectra, which also peaks near 450 nm.

The camera was set up by using methods commonly used for astrophotography (ref stark-labs-gain-offset). First the black-level offset was set by taking a bias frame (the lens cap was in place and the exposure time was 0 ms) and then finding the minimum value of the 16-bit greyscale value of all the pixels in the image (0 is black and 65,535 is white). The black-level offset was changed to ensure the minimum value in this image was greater than 100 and less than 1000. For this camera it works out be a black-level offset value of 135. The appropriate gain for this camera by taking a picture of the room ceiling and examining the maximum value in the image. If it was well below 65k, the gain was increased. If it was at 65k, the gain was decreased. For this camera, at all bin-levels except 1x1 a gain setting of 0 caused a "bunch-up" of pixels in the histogram at 65k. This "bunch-up" is where the pixels are hitting full-well capacity (maximum number of electrons stored). When the camera's binning was set to 1 by 1, the gain needed to be increased to 35 to cause the pixels to hit full-well capacity.

With the camera's gain and black-level set up the sensitivity of the camera was evaluated through the binning process. When pixels are binned, their values are combined. For example in 2x2 binning, 2 vertical and 2 horizontal adjacent pixels are combined into 1 pixel. In 3x3 binning 3 vertical and 3 horizontal pixels are combined, etc. This camera has binning levels from 1 to 10 in both the horizontal and vertical axis. The vertical axis is binned in hardware and the horizontal axis is binned in software. For simplicity's sake, when we binned the camera, we changed both the horizontal and vertical bins by the same amount. The science images acquired in this work were taken at 5x5 binning. Using these techniques an idea of the sensitivity of the system and the number of time intervals binned is provided in figure 5.

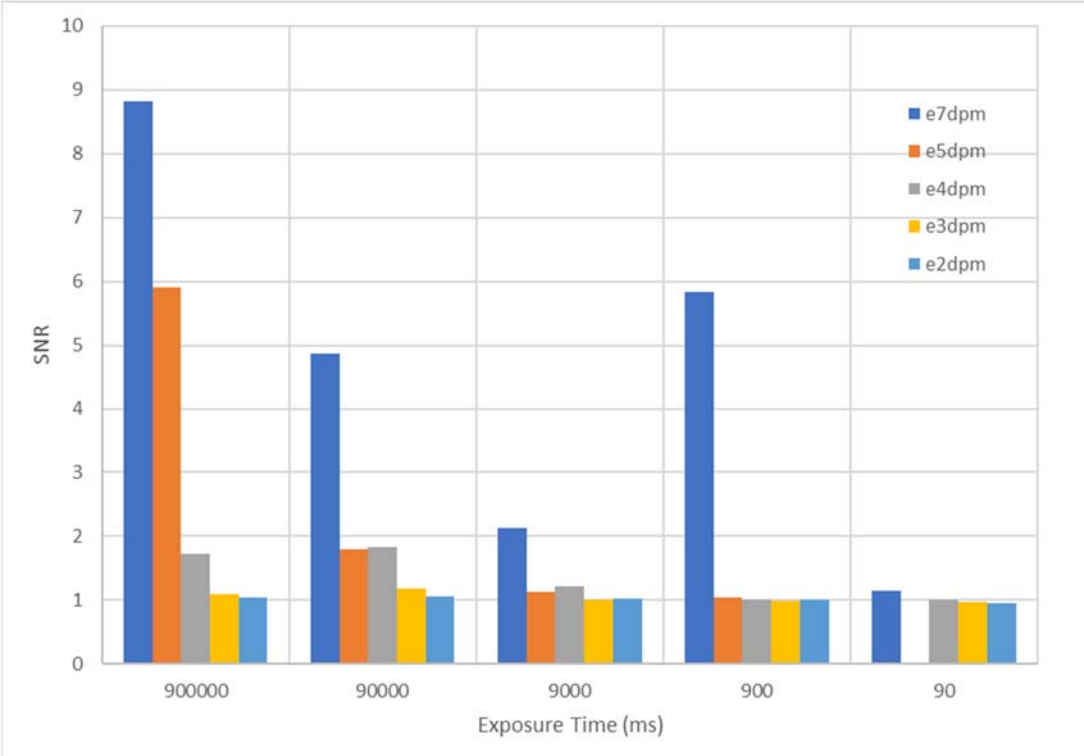

Figure 5. Signal to Noise Ratio (SNR) compared to sub-Exposure Time

Different scintillating materials were evaluated. Solid sheets of ZnS(Ag) were the brightest. This may be because the plastic coating on the back acted as a light pipe (See figure 6). It is not clear if the activity imaging is being washed out from the light pipe properties of the plastic coating.

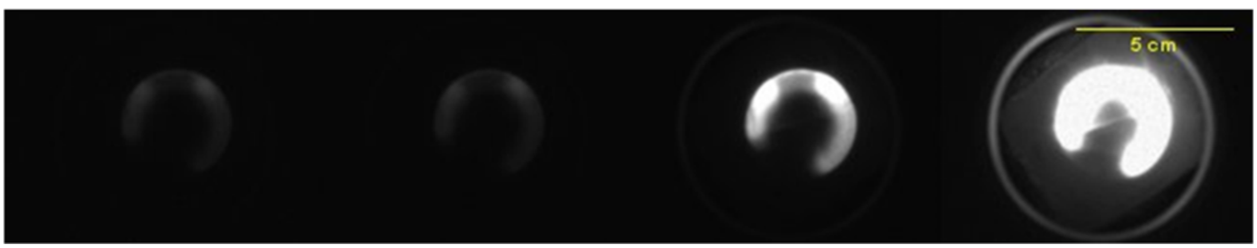

Figure 6. Pu-238 with 1E7 dpm underneath Zn(S) scintillator at sub-exposure times of 900, 9000, 90000, and 900000 ms.

Powdered ZnS(Cu) was also evaluated and by using it, were able to possibly locate large particles of Pu-238 at specific locations on the substrate. See Figure 7.

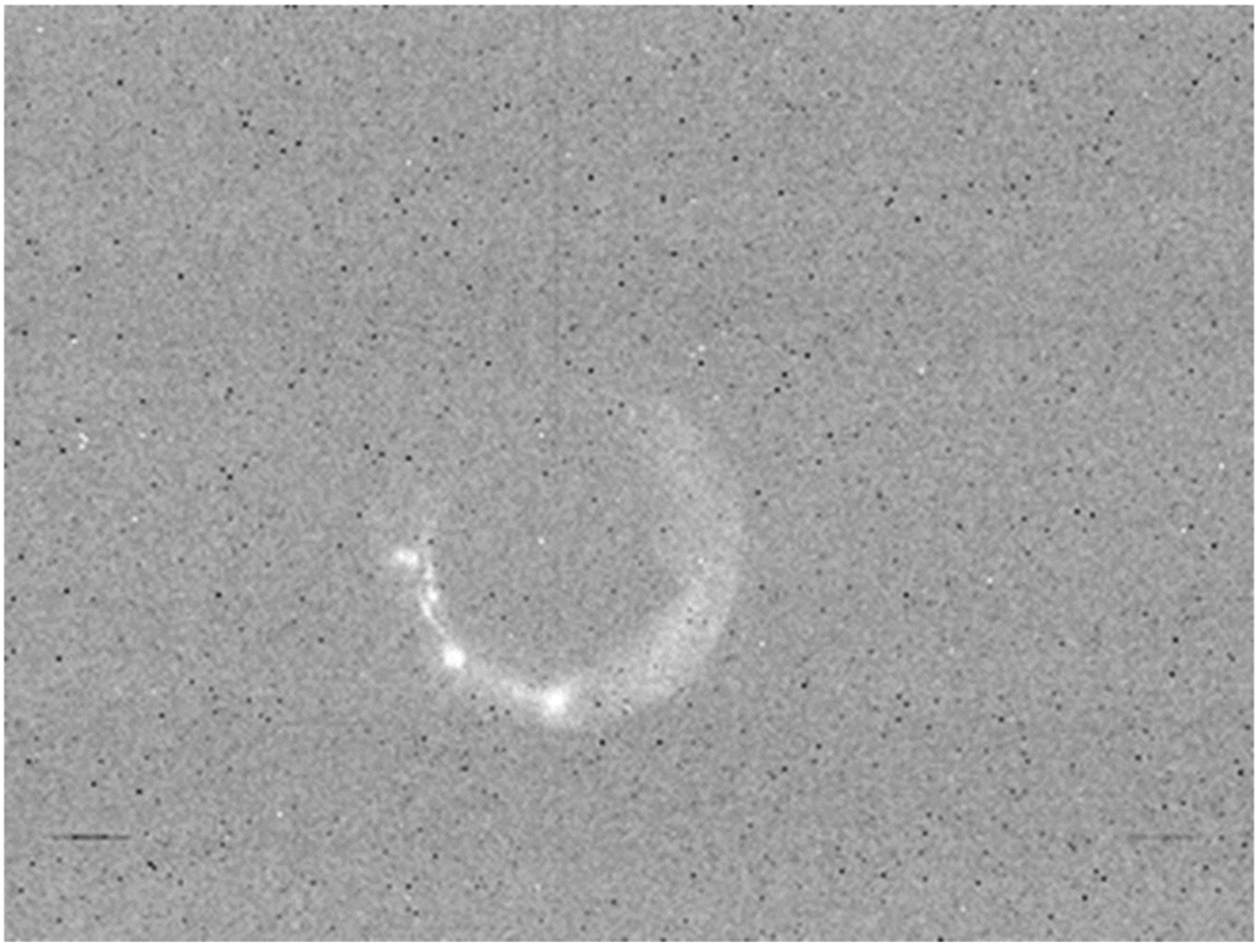

Figure 7. Powdered Zn(S) over 106 dpm Plutonium-238 plate, 60 second sub-exposure time, 10x10 binning

PerkinElmer's Ultima-Gold Liquid Scintillation Cocktail (LSC) was also reactive enough to show activity. With the LSC and with solid ZnS, Pu-238 deposited within the actual HEPA filters was imaged (see Figures 8, 9, and 10). With an improved powder deposition technique, the ZnS powder would also probably work to image alpha emitters deposited on a filter.

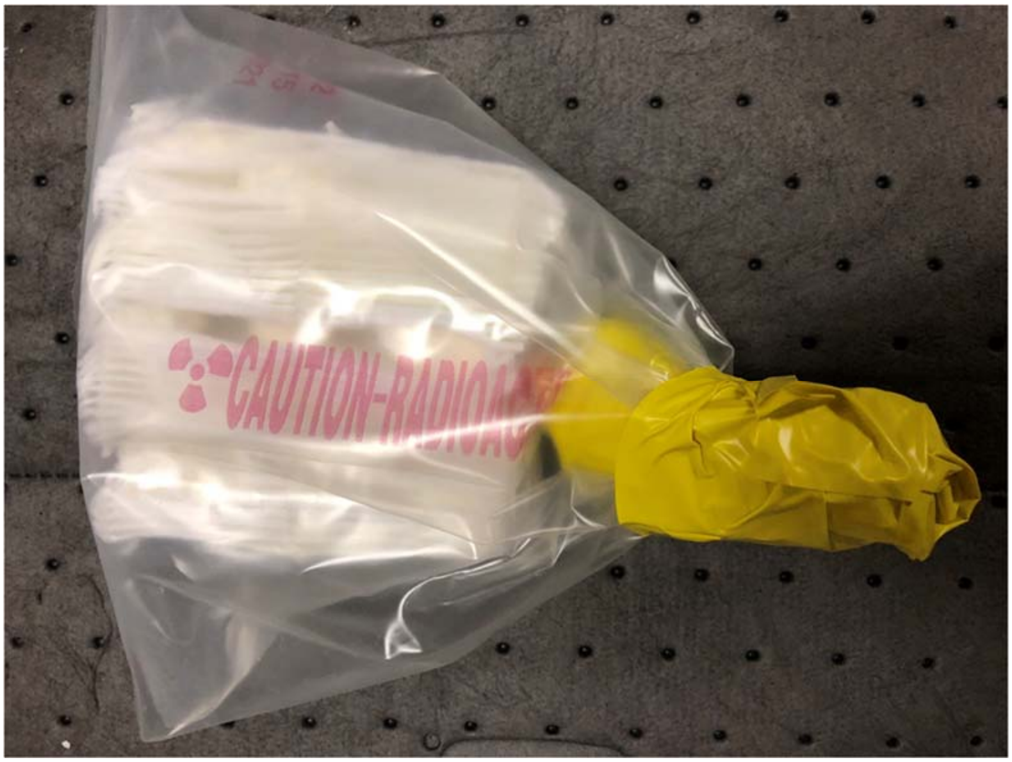

Figure 8. Picture of filter containing Pu-238 and LSC cocktail.

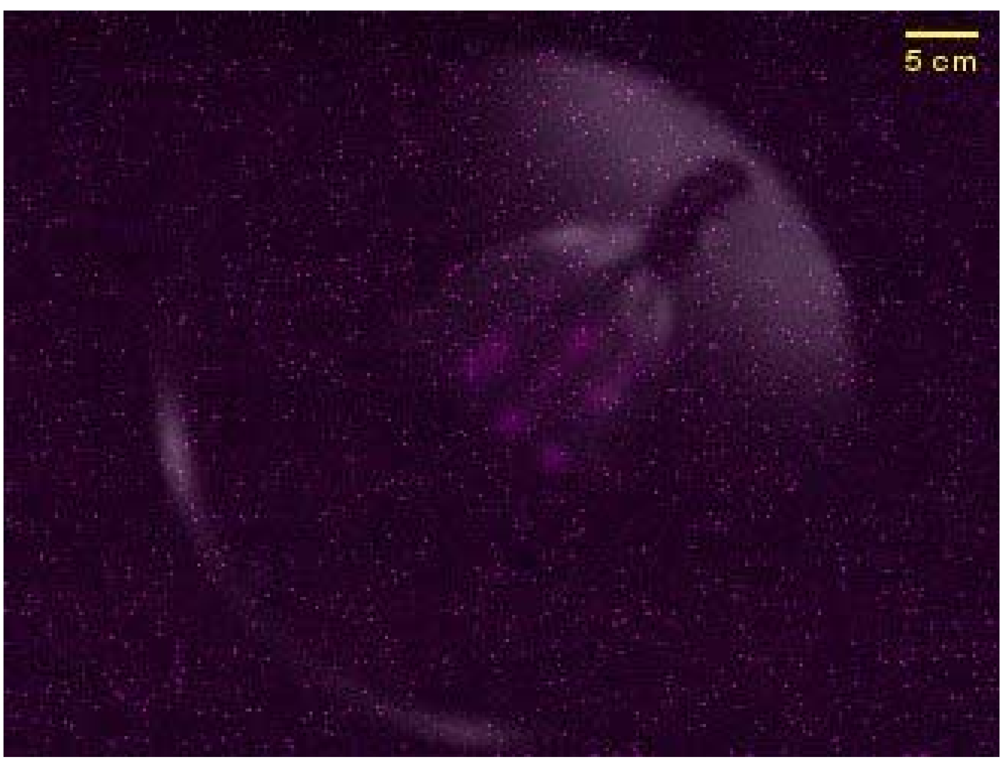

Figure 9. LSC cocktail on top of 106 dpm Plutonium in 9 locations in filter, 60 second sub-exposure time, 10x10 binning

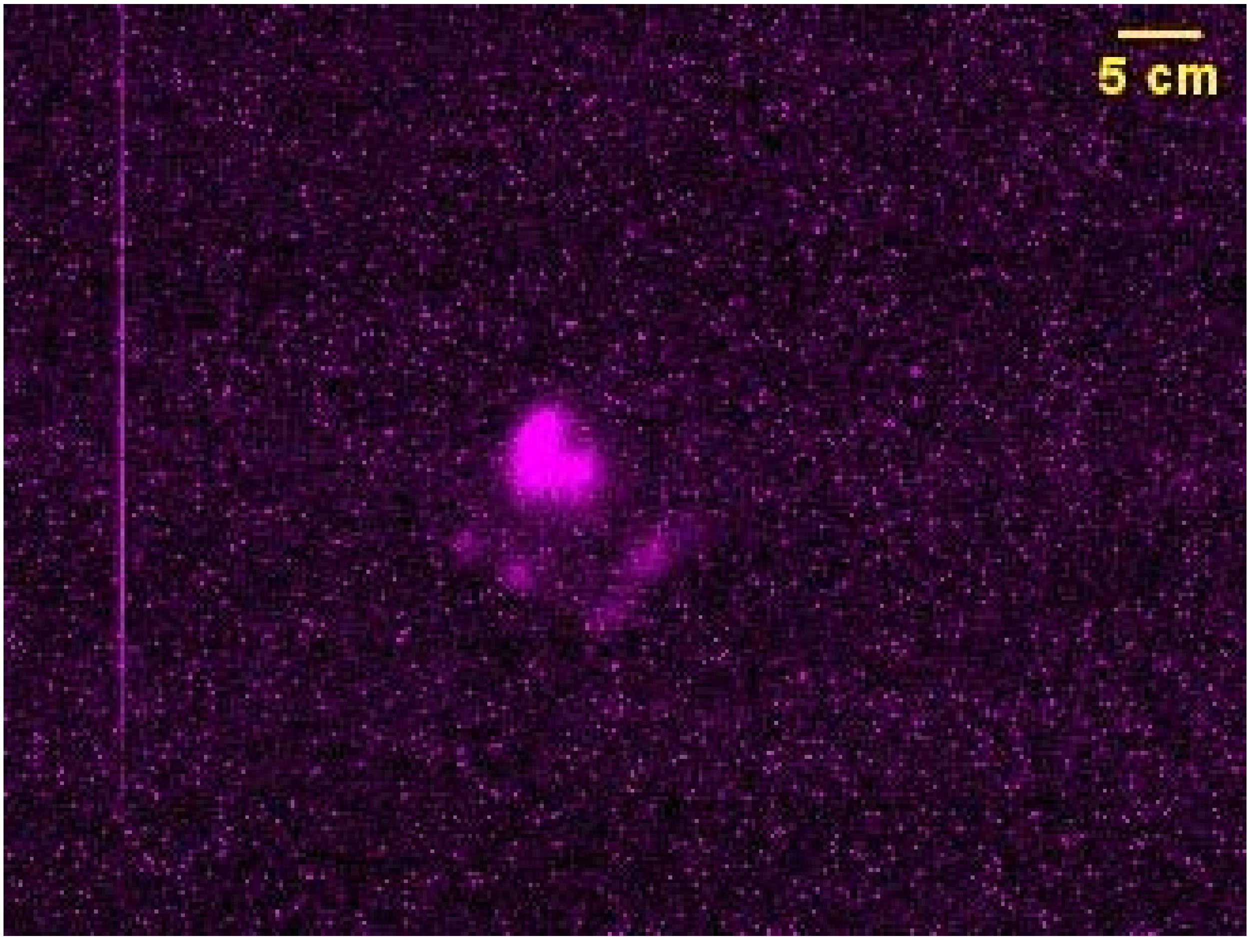

Figure 10. Solid Zn(S) on top of 106 dpm Plutonium in 9 locations in filter, 60 second sub-exposure time, 10x10 binning.

While imaging contamination on the filters was achieved, there is more work needed to be done to improve the resolution and sensitivity of the imaging. It is suspected that while they are the brightest, the ZnS(Ag) sheets are losing resolution because the plastic sheet substrate is acting as a light pipe. Perhaps a ZnS(Ag) laid down on a fractured substrate would provide improved resolution. The most inexpensive of these scintillators was the liquid scintillation cocktail. Work needs to be done to evaluate the most efficient way to apply the cocktail, and how much needs to be applied to work. Several different activated ZnS compounds exist, a study of the various scintillators to see which shows the most promise. Finally the system needs to be applied to some real world samples taken in contamination areas to access the effectiveness of this system.

FY2019 Accomplishments

- Built an alpha imager which can be used for this project or others

- Determined sheet of ZnS or LSC spray will work to image contamination on and within air filter, and with improvements to the technique, ZnS powder could also be used.
- We can directly see where hotspots of Pu are located on media
- Light-tight media holder created and method to acquire good photos developed
- Evaluated distribution of Radon and Progeny on this Air Purifier systems
- Acquired nuclear data on x-ray emissions of a number of actinides which could be of value to the Nuclear Data literature

Future Directions

- Study current scintillation agents properties further to determine if sensitivity limits can be extended
- Study more scintillating agents to refine properties of imager
- Study scintillation agent delivery systems
- Deploy air purifiers in- and collect filters from- an airborne radiation contaminated area
- Evaluate results of assay system vs dissection of real world air filters to see where contamination located
- Develop algorithm to turn image data into quantifiable "action" data: How much activity is on the filter, when is alpha (not Radon) contamination found.
- Use technique developed to precisely locate deposition of alpha emitting material on samples, for QA/QC purposes.

FY 2019 Publications/Presentations

1. 2nd International Conference on Radioanalytical and Nuclear Chemistry (RANC 2019)
2. SRNL-STI-2019-00596 Imaging plutonium activity with a CCD-based camera
3. SRNL-STI-2019-00597 Scintillator evaluation using a CCD-based camera
4. SRNL-STI-2019-00598 Comparison of X-ray emissions of Pu-238 samples
5. SRNL-STI-2019-00599 Theoretical approach to Large Area Airborne Contamination Monitoring using a CCD-based camera

FY 2020 Publications/Presentations

1. SERMACS2019 - Large Area Airborne Contamination Monitoring, Savannah, GA 10/20
2. Seminar at UNC-Charlotte - The Savannah River Site: Nuclear Measurements around the DOE Complex, 11/25

References

1. Gain/Offset: http://www.stark-labs.com/help/blog/files/GainAndOffset.php
2. Camera Specs: Thor-labs manual
3. SNR and exposure time: http://dslr-astrophotography.com/long-exposures-multiple-shorter-exposures/\
4. SNR and future work: http://www.stark-labs.com/craig/resources/Articles-&-Reviews/SNR-Part-1.pdf

5. Science Images: http://spiff.rit.edu/classes/phys445/lectures/readout/readout.html
6. Science Images: http://spiff.rit.edu/classes/phys445/lectures/median/median.html
7. https://www.sciencedirect.com/science/article/pii/S0168900218318199?via%3Dihub
8. https://www.nature.com/articles/s41598-018-21500-z
9. https://www.sciencedirect.com/science/article/pii/S1350448718301641#fig2
10. https://www.ncbi.nlm.nih.gov/pmc/articles/PMC6346089/

Acronyms

DOE-EM	Department of Energy Environmental Management
DOE-NNSA	Department of Energy National Nuclear Security Administration
SRS	Savannah River Site
SRNL	Savannah River National Laboratory

Intellectual Property

None

Total Number of Post-Doctoral Researchers

None

Total Number of Interns or Summer Students

1 – Adam Judy