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# RASA Field Performance at US International Monitoring Stations

**Douglas Hunter**

**Michael Maxwell**

SEPTEMBER 2021

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# **RASA Field Performance at US International Monitoring Stations**

Douglas Hunter  
Michael Maxwell

March 2021

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## **EXECUTIVE SUMMARY**

This report attempts to collate, analyze, and summarize RASA performance over time at each of 11 US operated IMS stations. The performance over time was compared to routine maintenance operations.

The data, in general, were summarized into a set analyzed almost exclusively in Excel. The data set allows for further probing into RASA performance over time. The data set would permit data to be routinely added for continued analysis.

In general, all RASA systems ran within the CTBTO mandated the MDA requirements over their current lifetime of operations. The largest impacts to performance appeared to arise from variabilities of Germanium gamma detectors as they were replaced.

It is noted that the air volumes of samples were not provided and had to be assumed to be constant over the period analyzed.

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## LIST OF ABBREVIATIONS

ARR	Automated Radionuclide Reports
CTBTO	Comprehensive Test Ban Treaty Organization
DTRA	Defense Threat Reduction Agency
FWHM	Full Width at Half Maximum Statistical Function
GDMS	General Dynamics Mission Systems
HPGe	High Purity Germanium
keV	Kiloelectron Volts
IMS	International Monitoring Stations
$\mu\text{Bq}/\text{scm}$	MicroBecquerel per standard cubic meter
$\text{mBq}/\text{scm}$	MilliBecquerel per standard cubic meter.
MDA	Minimum Detectable Activity
PHD	Pulse Height Determination
NORM	Naturally Occurring Radioactive Material
PHD	Pulse Height Determination
RASA	Radionuclide Aerosol Sampler Analyzer
ROI	Region of Interest
scm	Standard cubic meter
SOH	Status of Health
SRNL	Savannah River National Laboratory



## 1.0 Introduction

This report summarizes RASA performance at 11 US operated International Monitoring Stations (IMS). The focus of the study was to correlate maintenance activities impacts over extended duration of a RASA's operation. Maintenance activities were supplied by General Dynamics. The data were retrieved from the CTBTO database.

As per the operation manual (CTBT/WGB/TL-11,17/18), minimum requirements for particulate monitoring, requirements specify a baseline sensitivity of 10 to 30  $\mu\text{Bq}/\text{scm}$  for Ba-140 (the upper limit is intended for high background areas). The required assessment of gamma ray spectrometry systems is outlined elsewhere (Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization. Certification of Radionuclide Laboratories. CTBT/PTS/INF.96/Rev.9, 1 August 2012.). Calculation of Minimum Detectable Activities requirements were taken from this manual.

This report will attempt to collate, analyze, and summarize a large volume of data. In order to make the study more useful, a separate set of spreadsheets has been generated that will permit further investigation of the underlying data. These spreadsheets include raw spectrometry data, spectrometry efficiency curve fits, spectrometry resolution curve fits, and the MDA analyses. These spreadsheets will allow DTRA or others to continue monitoring the MDA improvements/degradations over time as equipment is repaired or renovated. In general, the spreadsheets were prepared primarily in Excel. Spectra were analyzed in PeakEasy v4.96 and efficiency curves were calculated in Origin, all on a PC platform.

SRNL, as an outsider, approached this task as "blindly" as possible such that a unique take on data might occur. The received CTBTO manual (Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization. Certification of Radionuclide Laboratories. CTBT/PTS/INF.96/Rev.9, 1 August 2012) only addressed the Ba-140 MDA requirement. Initial work included all 4 gamma energies but was subsequently trimmed to the transition at 537 keV. (in concurrence with Operational Manual for Radionuclide Monitoring and the International Exchange of Radionuclide Laboratories, CTBT/WGB/TL-11,17/18/Rev.6, 6 March 2019.)

An approach delineating performance between two components was adopted. First, static system detection performance (represented by BlankPHD files) was analyzed. The data give insight into RASA performance as a machine with respect to detector sensitivity for a given background environment; however, these data were collected at best once per quarter. Second, dynamic system performance (represented by SamplePHD files) was analyzed, giving insight into detector sensitivity and background environment, but also varying environmental radiation collections.

## 2.0 Results and Discussion

### 2.1 Data Analysis files

In general, an Excel file was built around the data. One Excel file was created for each of 11 locations (see Table 1). The tabs within each file track the results discussed below. Maintenance information was supplied by General Dynamics (RASA Station Upgrade

Timeline\_GDMS\_rev2). These data were overlaid on pulse height data to infer impacts of maintenance/updates to performance over time.

## 2.2 Blank Determinations

From a functionality perspective, the first step was to look at Blank Pulse Height Data (BlankPHD), which provides pulse height data (PHD) acquired by counting an unexposed filter paper. All BlankPHD data files from all US operated IMS stations were downloaded and assessed.

These data represent inherent background noise within the RASA system as well as environmental background activity from the environment. Total counts per spectrum normalized for count time (blank cpm) was assessed over time. These data were overlaid on dates of service/maintenance. The stations and time periods covered are listed in Table 1.

**Table 1. List of Sites Analyzed**

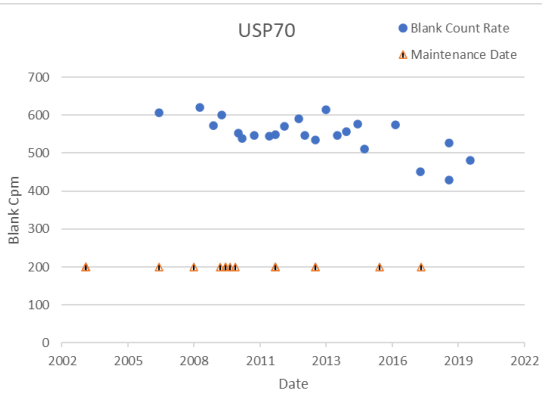
Station	Designation	Time Period
USP70	Sacramento, CA	2006-Present
USP71	Sand Point, AK	2006-Present
USP72	Melbourne, FL	2004-Present
USP73	Palmer Station, Antarctica	2005-Present
USP74	Ashland, KS	2003-Present
USP75	Charlottesville, VA	2003-Present
USP76	Salchaket, AK	2006-Present
USP77	Wake Island, West Pacific Ocean	2007-Present
USP78	Midway Island, North Pacific Ocean	2009-Present
USP79	Oahu, HI	2005-Present
USP80	Guam, Western Pacific Ocean	2007-Present

## 2.3 Detector Total Count Rates

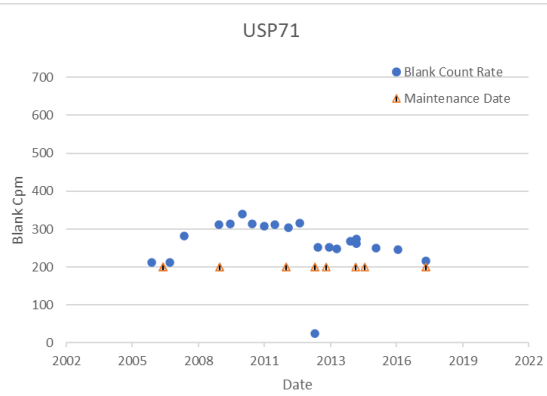
Representation of data is presented as control charts to allow an operator to look for variations which may not currently be listed in the maintenance records. Additional statistical models could be employed but may be challenging to adopt given current variables and unknowns. The current data are organized in a manner that would permit such analysis to occur. This allows comparison of total count rates between stations as well as within stations over time. Excursions over 700 cpm

were clear indicators that maintenance was required due to overlap of maintenance activities during these periods. Plots of total count rates vs time for each location are summarized in Figure 1 below. (Note: The triangles on the plots indicate maintenance dates. The vertical position of the maintenance triangle means nothing.)

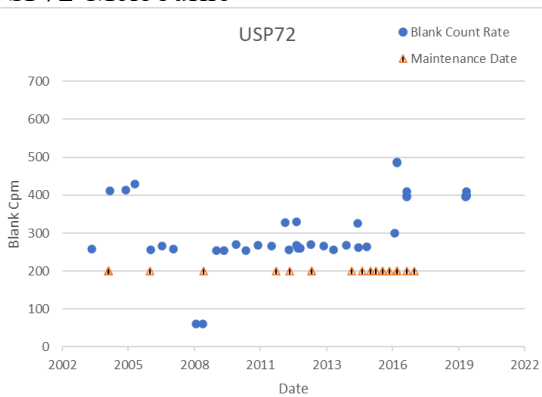
USP70-Sacramento



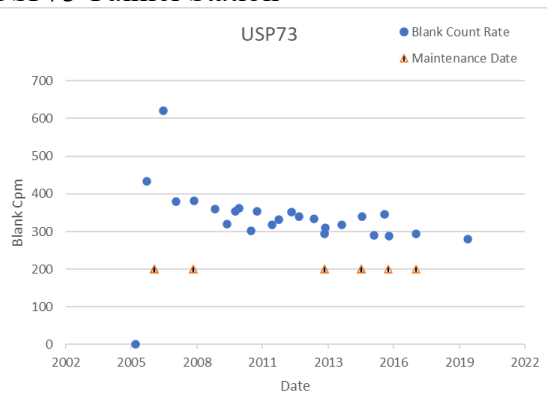
USP71-Sand Point



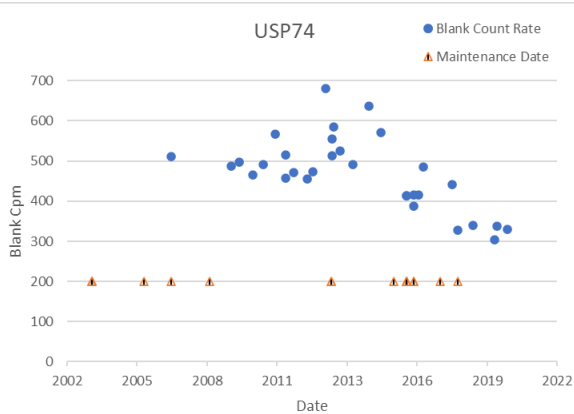
USP72-Melbourne



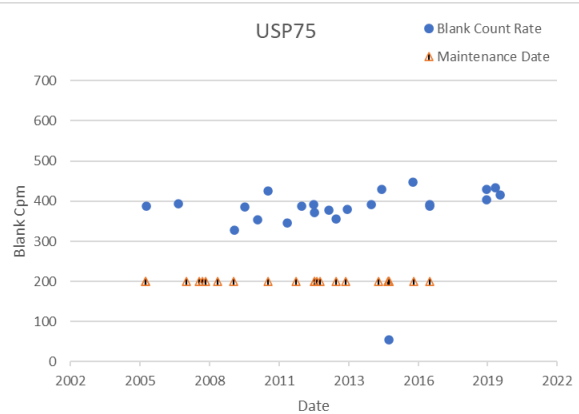
USP73-Palmer Station



USP74-Ashland



USP75-Charlottesville



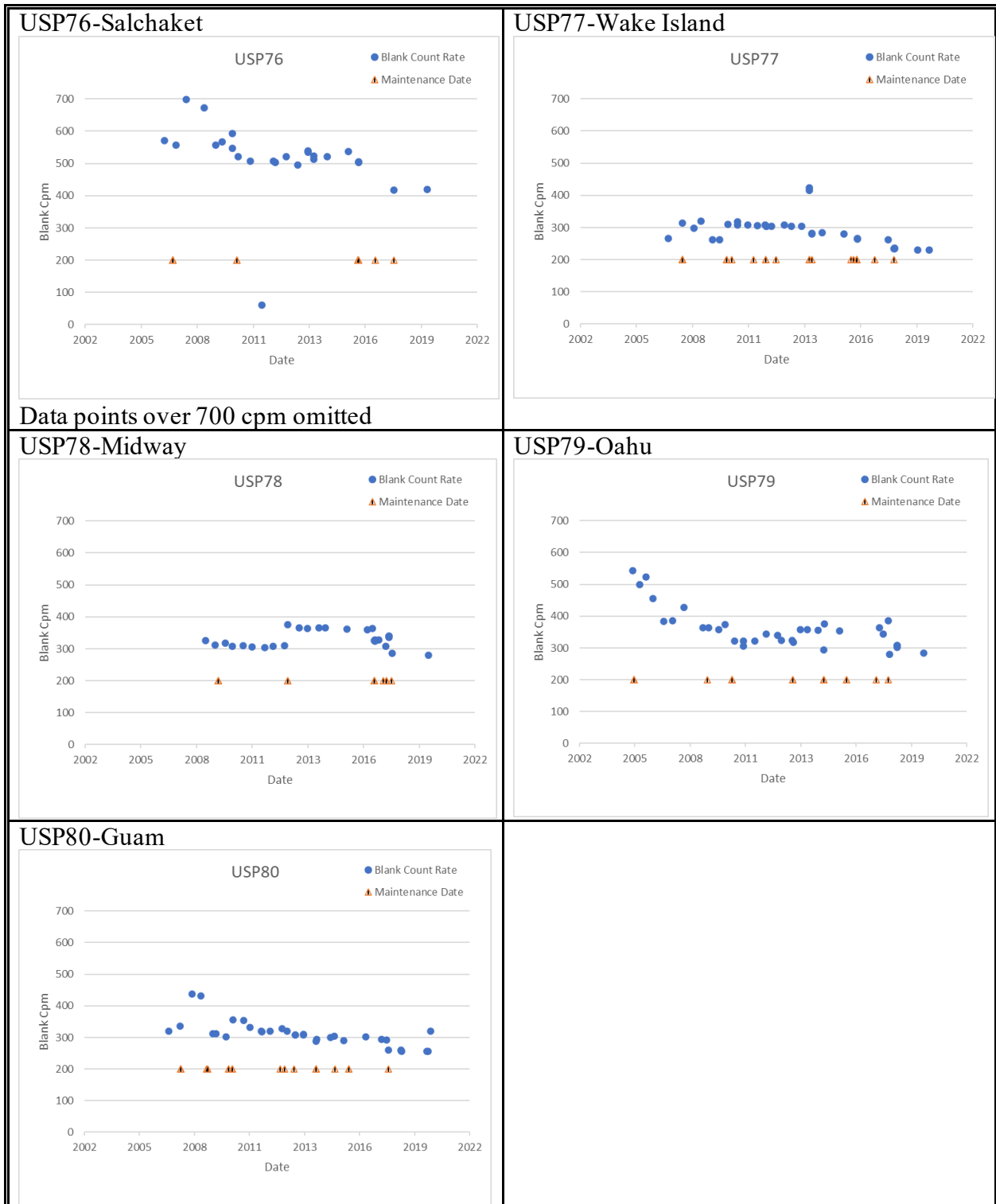


Figure 1. Detector total count rates over operation time (●). Maintenance is logged as well (Δ).

## 2.4 Total Spectral Count Rate Discussion

Inspection of the charts indicates that RASA units exhibit a spread of blank count rates from 200-700 cpm in typical operation. In some instances (i.e., Oahu) The blank background has decreased during operation. Most have been consistent, and none have demonstrated major increases (outside of “step-function” changes correlated with maintenance.)

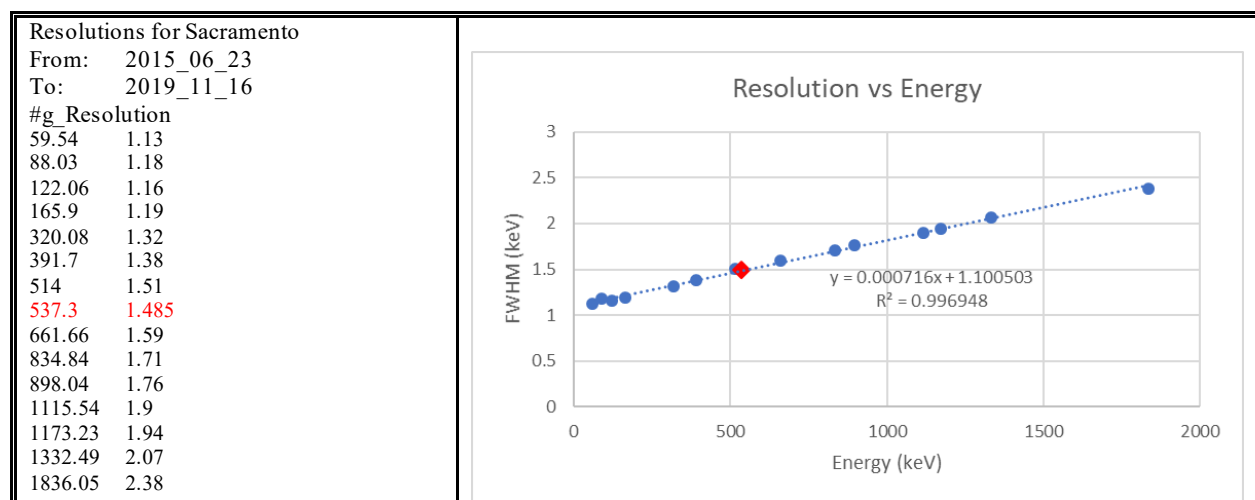
It appears that stations could be clumped as to their class of count rates. Three stations fall in the 200-300 cpm class for the preponderance of their operation, five stations fall within 300-400 cpm and three stations fall within 400-700 cpm bins. This almost “Gaussian” distribution suggests that the range of background counts for RASA, whether attributable to inherent background of its location, human maintenance and operation, or within the design of the system, merely represents the range of performance in operation. These authors would expect trends to certain locations correlated with natural background radiation variations. Such correlations were not attempted in this investigation.

## 2.5 Minimum Detectable Activity (MDA)

Calculation of the MDA can be broken down to discreet calculational steps, providing insight into the impact of each of those steps on the final MDA determination. These steps include resolution, efficiency, and final MDA determination. The model for the MDA calculation adopted in this investigation is summarized in Appendix A.

## 2.6 Detector Resolution.

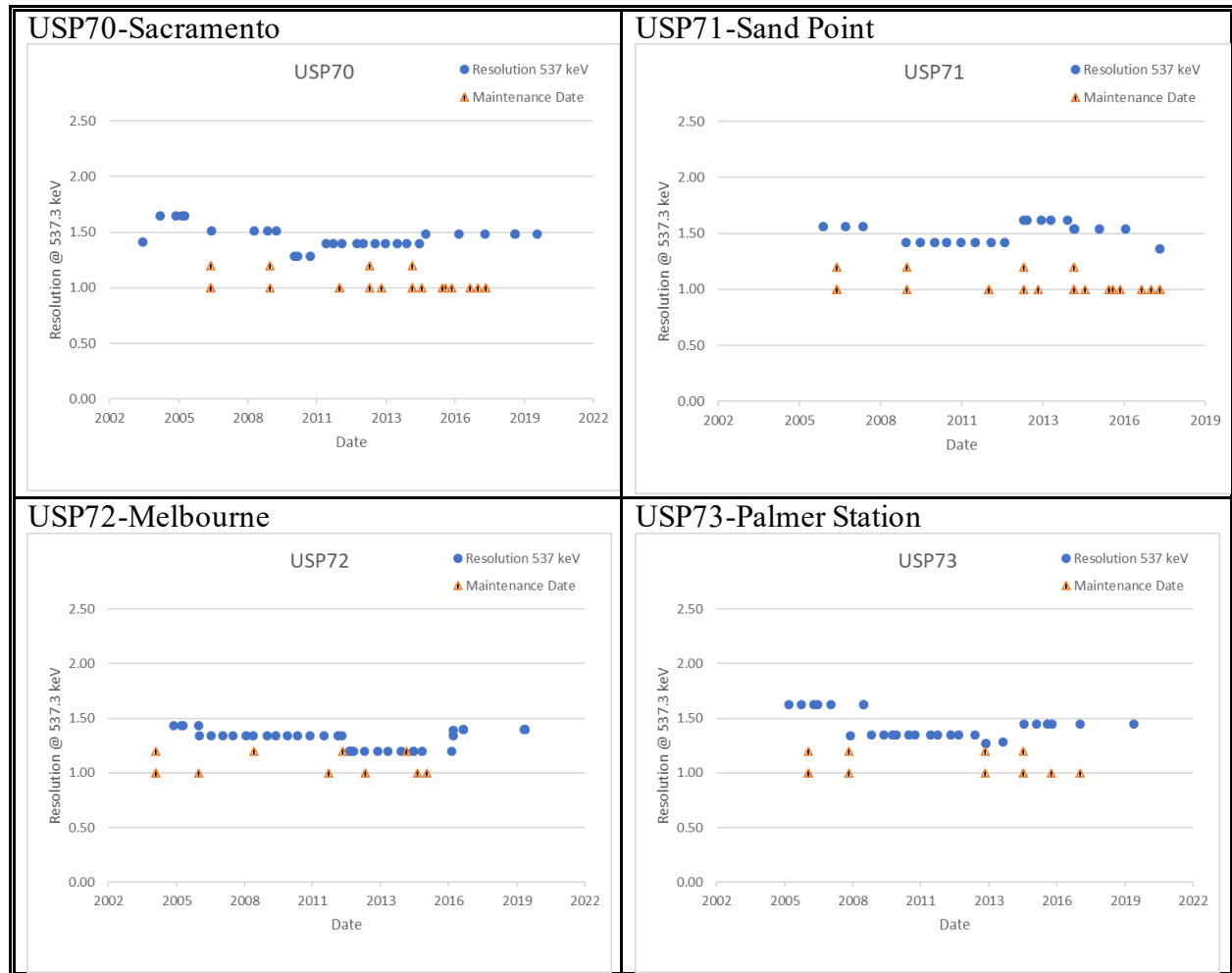
The resolution of the detector will define the region of interest (ROI) channels in the gamma spectrum. The FWHM results of a multi-element standard are included in each BlankPHD file. These are plotted and a linear fit is used to interpolate to the assumed resolution of Ba-140 at 537.3 keV. An example is illustrated below in Figure 2.



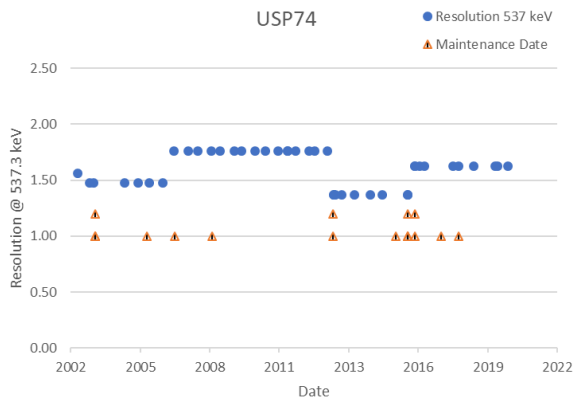
**Figure 2. Determination of Ba-140 resolution at 537.3 keV from BlankPHD file header information**

As per procedure (CTBT/WGB/TL-11,17/18), the ROI is defined +/-1.25 FWHM ( $3\sigma$ ) on either side of the hypothetical peak centroid at 537.3 keV). The square root of the sum of the counts within this ROI yields the standard deviation of the background (Equation 4, Appendix A). This loads into the MDA calculation. From a control chart perspective, a smaller value for the resolution is desirable.

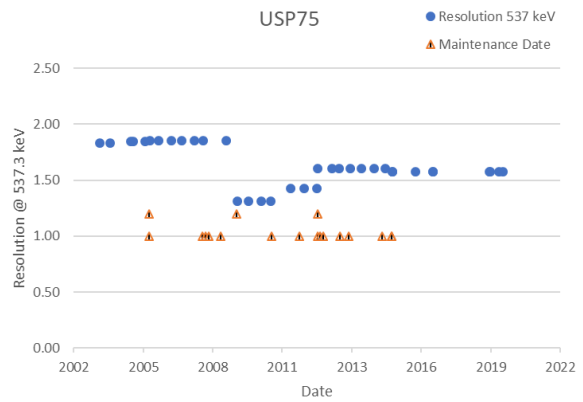
Plots of resolution vs time for each location are summarized in Figure 3 below. It is important to recognize that “background” impacts the MDA (Appendix A, Equation 2) as the square root, whereas detector efficiency impacts the MDA linearly.



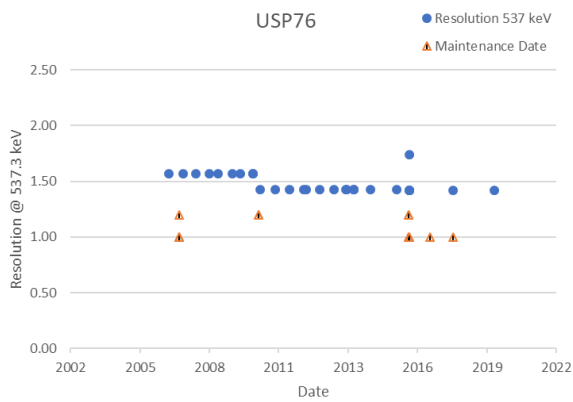
USP74-Ashland



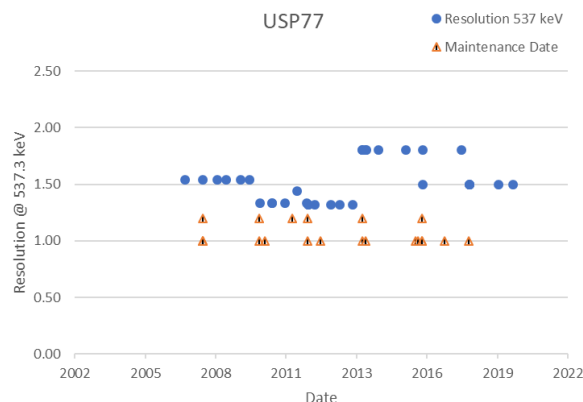
USP75- Charlottesville



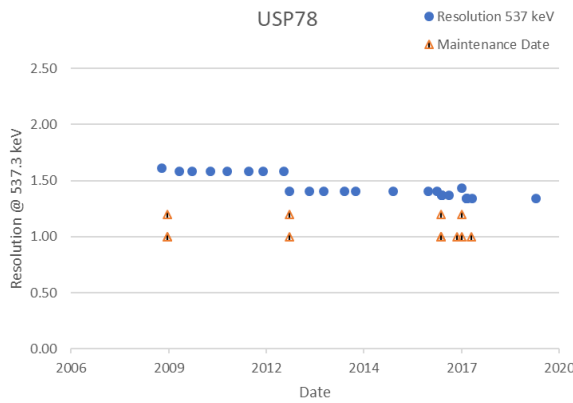
USP76-Salchaket



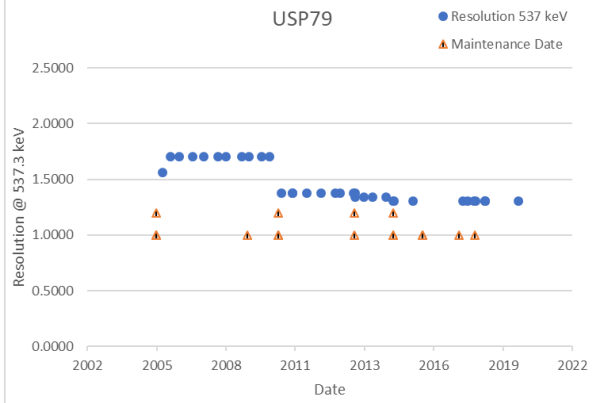
USP77-Wake Island



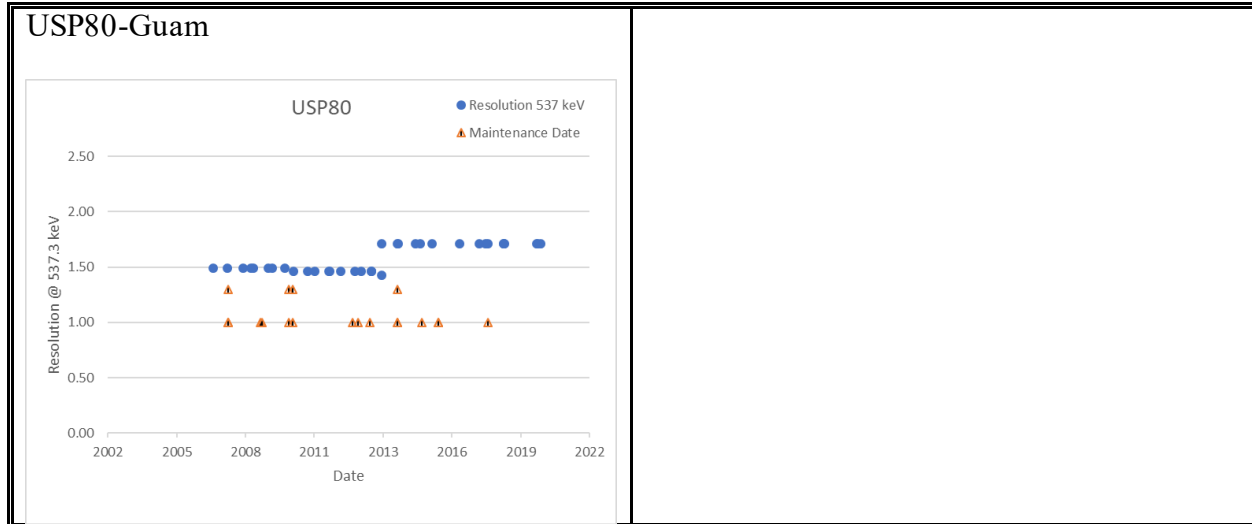
USP78-Midway



USP79-Oahu







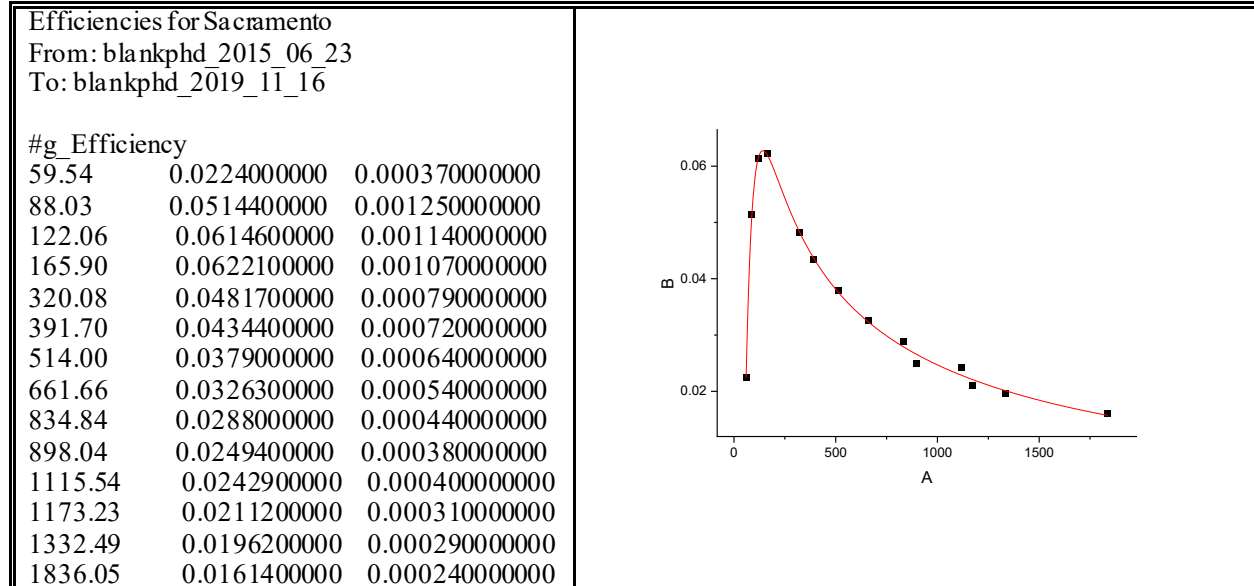
**Figure 3. Resolution of HPGe detectors over time (●). Maintenance is logged as well (Δ). The elevated Δ's are indicative specifically for detector replacement.**

## 2.7 Discussion on Resolution

Resolution measurements are typically only made when there is a maintenance requirement. There are no data representing resolution degradation as the HPGe detectors age, which is not of concern, except that more frequent measurements could provide status of health (SOH) on resolution degradation as the detectors age. Instances were found where the same resolution data were used for 5+ years since resolution in the spectra header data only changes when changing detectors. This fact is pointed out merely as an observation, not as a recommendation. It may be possible to track resolution from daily sample data (i.e., the positron emission peak at 511 keV could be used; some analysts will use Radon daughters). All these solutions suffer from varying intensities over time. Overall, resolution variations are not impacting the MDA significantly due to the square root of the ROI relationship mentioned above.

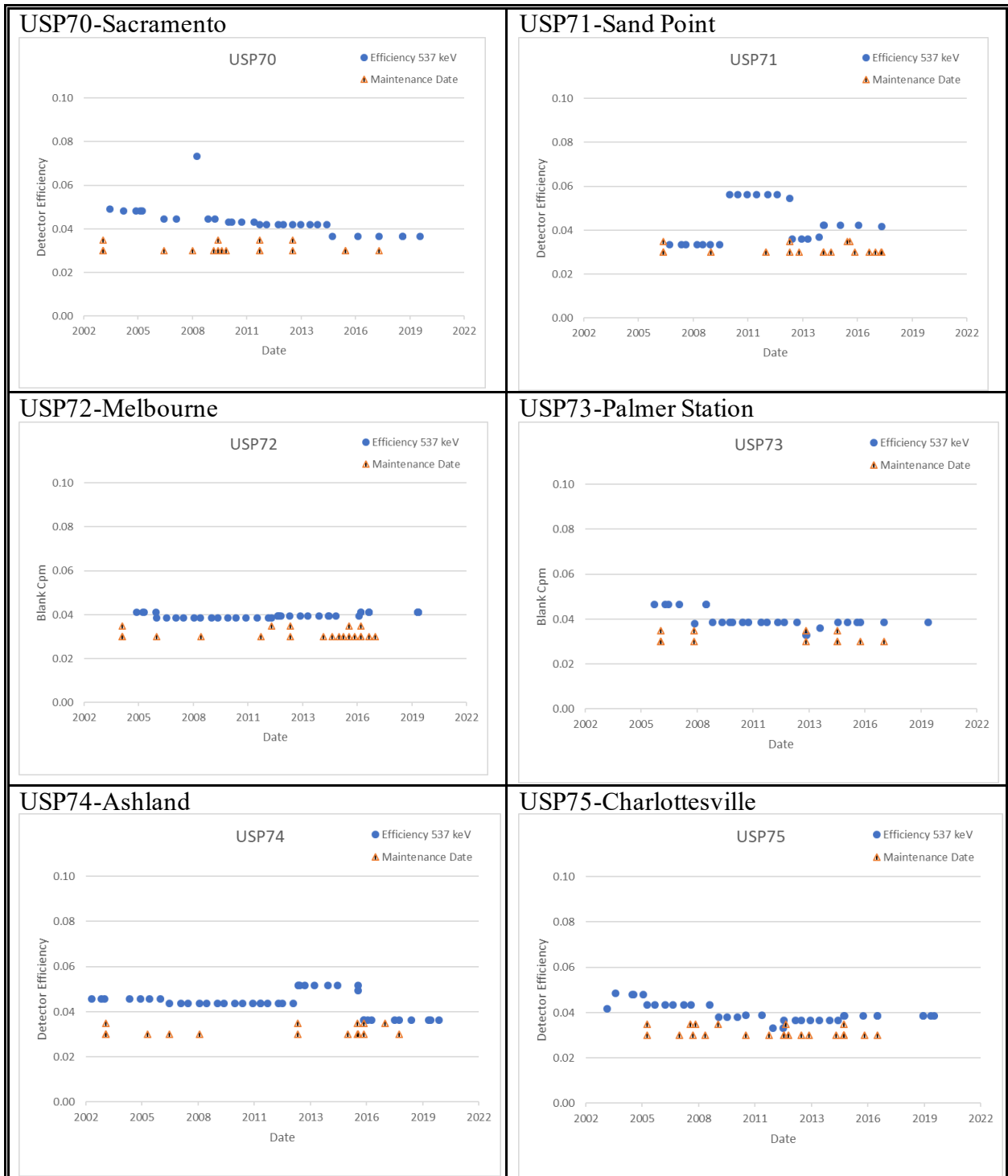
## 2.8 Detector Efficiency

Efficiency data was pulled from the BlankPHD file headers for each location over time. Exponential polynomial fits were applied to interpolate the detector efficiencies at the Ba-140 peak energy, 537.3 keV. The fits were performed in OriginPro 2020. Simpler fit models could be applied within Excel if requested. Uncertainties of fits can be estimated to be less than 5% at the Ba-140 peak energy, 537.3 keV. Figure 4 below illustrates the basic step to assess the detector efficiency at 537.3 keV.



**Figure 4. Curve fitting to assess detector efficiency at the Ba-140 peak, 537.3 keV**

Efficiency charts are illustrative on efficiency's impact on overall sensitivity of the RASA System (Figure 5). As new detectors were installed in during maintenance, detector efficiencies could vary dramatically. Case in point, USP71 -Sand Point demonstrates a striking efficiency jump with detector maintenance and the impact to the MDA is impacted inversely (i.e., increase in Efficiency, decrease in MDA/scm).





**Figure 5. Detector efficiency over time (●). Maintenance dates are logged as well (Δ). Multiple maintenance activities on the same date are indicated by offset triangles**

## 2.9 Discussion on Efficiency

The overall improvement in efficiency over time is clearly observable and correlates with an improvement in the MDA. The improvement relevant since the MDA is inversely proportional to detector efficiency. Prescreening of detector performance before installation can direct insight into impact on the RASA MDA performance. It is also clear that efficiency data is not routinely evaluated since that data is not required by IMS and IDC operational manuals.

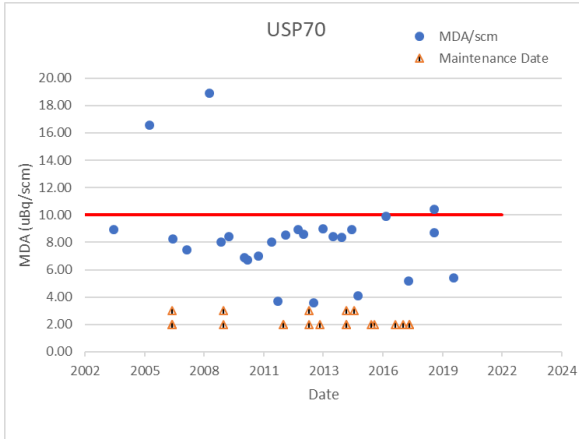
## 2.10 Minimum Detectable Activity (MDA)

The MDA are plotted as MDA/scm in  $\mu\text{Bq}/\text{scm}$  (Figure 6). The calculation assumes a 24-hour collection with a flow of 1000 scm/hour for 24 hours. Measured flow rates were not supplied. MDA/scm is therefore directly inversely proportional to the flow. If the performance as assessed by control charts is just meeting requirements, a lower than assumed flow could knock the analysis out of specification. If an uncertainty in flow could be established, then an uncertainty could be assigned to whether requirements are being achieved or not.

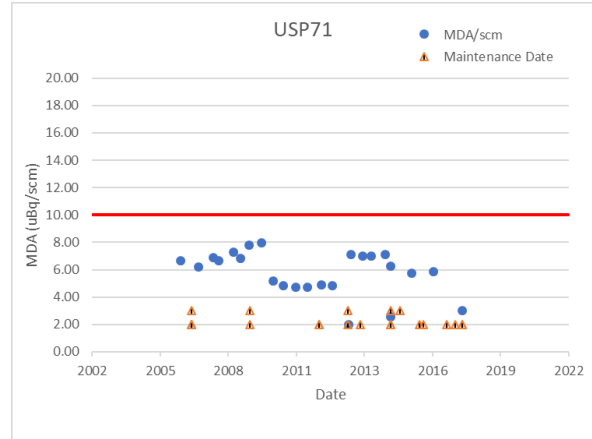
What may appear as anomalously low MDA/scm values are attributable to longer collection times (3-day count times versus 1-day count times). The data are retained for completeness of BlankPHD data. The low MDA/scm must be expressed as the equivalent of a 1-day count.

In general, RASA systems performed consistently over time with the largest drift in performance appearing to be correlated with detector maintenance where efficiency is a dominant variable.

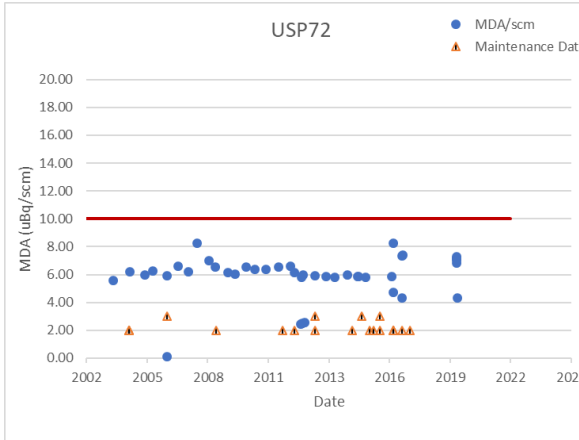
USP70-Sacramento



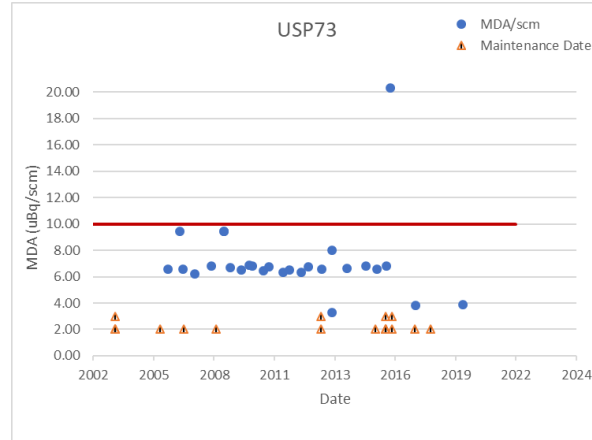
USP71-Sand Point



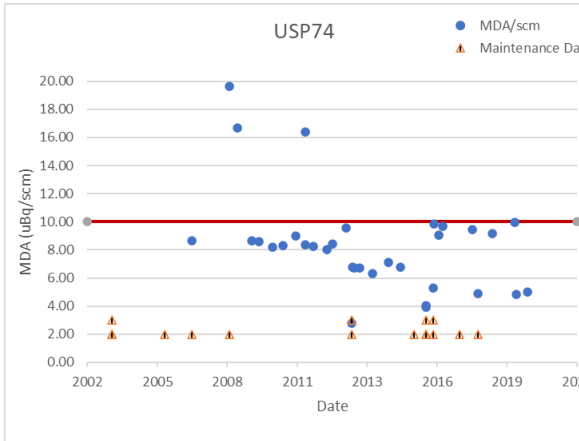
USP72-Melbourne



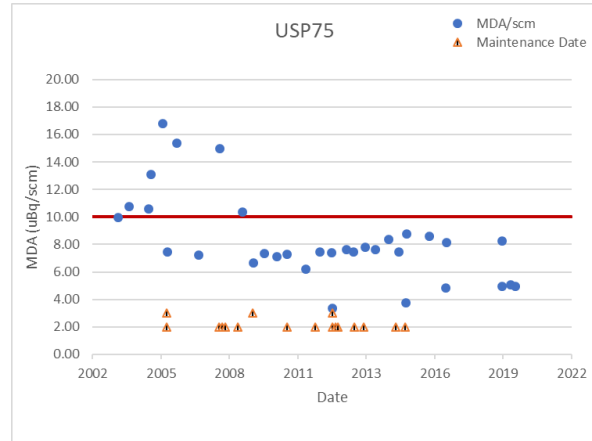
USP73-Palmer Station

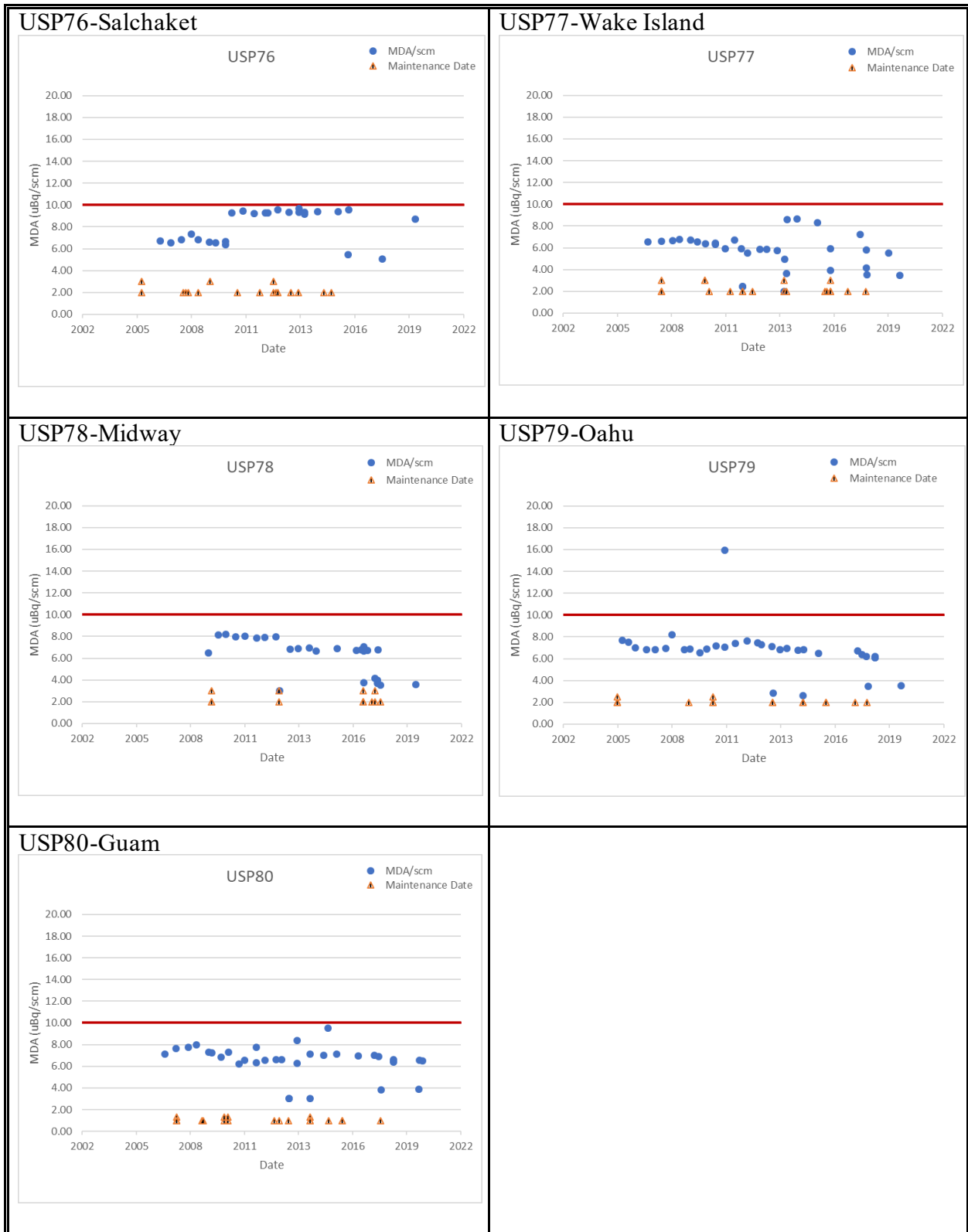


USP74-Ashland



USP75-Charlottesville





**Figure 6. The MDA (●) in (µBq/scm) is plotted over time. Maintenance dates are logged as well (Δ). The horizontal red line defines the lowest MDA required (10 µBq/scm).**

## 2.11 MDA Discussion

In general, the sites appear to have remained relatively constant in performance over time. In a few cases (i.e., USP78), some seem to have improved and that appears to be driven by a new detector with improved efficiency being installed during maintenance.

GDMS noted the lead plug improved the Ba-140 MDA at each RASA station. This analysis compared blanks prior to and after the lead plug installation in 2017 to remove impacts from other maintenance activities wherever possible. However, that improvement is not readily apparent relative to overall system performance when assessing overall count rates with respect to total counts. Less impact if any impact is observed with respect to an improved MDA. These are observational comments and do not imply that addition is not useful. However, variation in detectors performance can have an equal or greater impact and perhaps this observation lends to the discussion of cost benefit for specifying the highest efficiency possible during the detector purchase process. As stated, a direct inverse relationship exists between efficiency and the MDA.

## 2.12 MDA Impact with Respect to Flow

Blanks are collected without flow and represent sensitivity relative to the system and its environment. Sensitivities were collected as per an idealized flow of 24,000 scm of air sampled over 24 hours. Samples have flow data but also have naturally occurring radioactive material (NORM) collected from the air. The impact of flow is a direct impact to the MDA calculation (example; double the flow would lower the MDA by a factor of two). Unfortunately, analysis of gamma spectra of samples would also include an increased NORM and impedes interpretation of only flow impacts. The general path adopted here was to take one site (USP70, Sacramento) and retrieve SamplePHD sample files roughly a week prior and a week after blanks were measured. This approach allowed an initial look in variability of flow bracketing the time of blank collection.

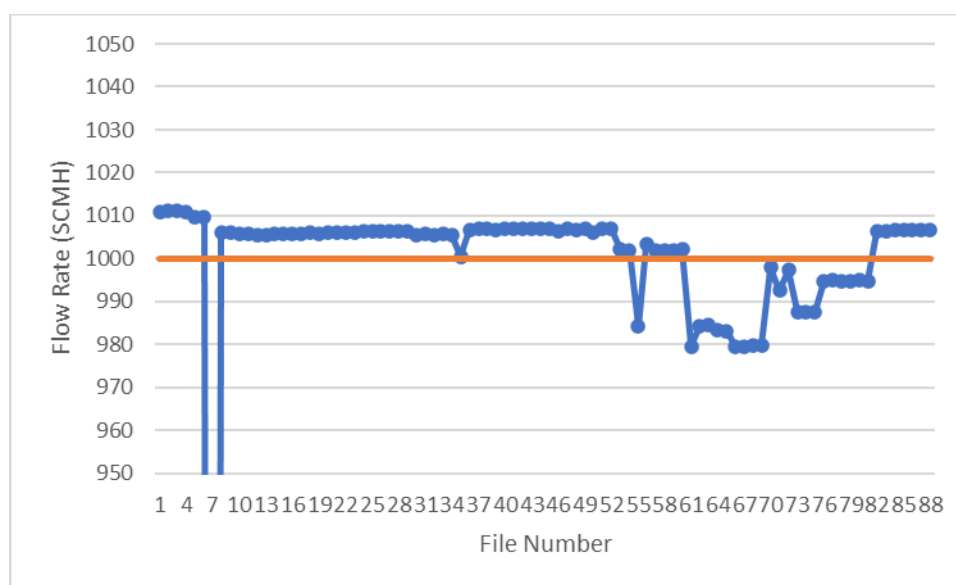
Automated Radionuclide Reports (ARRs) were not provided to SRNL. To speed the analysis, process code was written to extract flow, collection time and other pertinent information from sample files as shown below. Files were manually downloaded. If a complete data set of flows were requested from SRNL, a special request should be made to acquire the data which could then be quickly analyzed for detailed flow variances over time. In the output example below (Figure 7), it is also important to note that flow is extracted from the sample file and flow rate is then calculated from the collection time which helps to normalize against total flow that arose from variable run times.



File	type	Loc	Collstrt	CollStop	dys	scm	scm/min)	AcqStrt	Real(s)	Live(S)
sphdf20130603.txt	SAMPLEPHD	USP70	6/3/2013 21:39	6/4/2013 21:37	0.99816	24217.68	16.84884	6/5/2013 21:39	84308.34	83956.02
sphdf20130604.txt	SAMPLEPHD	USP70	6/4/2013 21:39	6/5/2013 21:37	0.998137	24223.34	16.85317	6/6/2013 21:39	84308.32	83965.22
sphdf20130605.txt	SAMPLEPHD	USP70	6/5/2013 21:39	6/6/2013 21:37	0.998148	24223.74	16.85325	6/7/2013 21:39	84308.22	83987.8
sphdf20130606.txt	SAMPLEPHD	USP70	6/6/2013 21:39	6/7/2013 21:37	0.99816	24216.95	16.84833	6/8/2013 21:39	84308.24	82901.44
sphdf20130607.txt	SAMPLEPHD	USP70	6/7/2013 21:39	6/8/2013 21:37	0.998137	24189.44	16.82958	6/9/2013 21:39	813.92	787.2
sphdf20130609.txt	SAMPLEPHD	USP70	6/9/2013 21:39	6/10/2013 21:05	0.975833	23648.54	16.82931	6/11/2013 23:03	11689.4	10390.38
sphdf20130615.txt	SAMPLEPHD	USP70	6/15/2013 20:35	6/22/2013 23:37	7.126262	25861.43	2.520161	6/23/2013 23:39	77108.6	76759.66
sphdf20130622.txt	SAMPLEPHD	USP70	6/22/2013 23:39	6/23/2013 23:37	0.998113	24102.2	16.76928	6/24/2013 21:39	84306.16	83958
sphdf20130623.txt	SAMPLEPHD	USP70	6/23/2013 23:39	6/24/2013 21:37	0.914838	22088.14	16.76689	6/25/2013 21:39	84306.94	83988.1
sphdf20130624.txt	SAMPLEPHD	USP70	6/24/2013 21:39	6/25/2013 21:37	0.998137	24096.06	16.76462	6/26/2013 21:39	84310.02	83965.58

**Figure 7. Example table of flows extracted from samples files selected before and after a blank run on 2013/06/15 at USP 70**

Approximately, 90 SamplePHD files were downloaded spanning 10 blank measurements made between 2013 and 2019. The flows are tabulated in Figure 8 below in a fashion that demonstrates generally variations in flow rate over a 6-year time span.

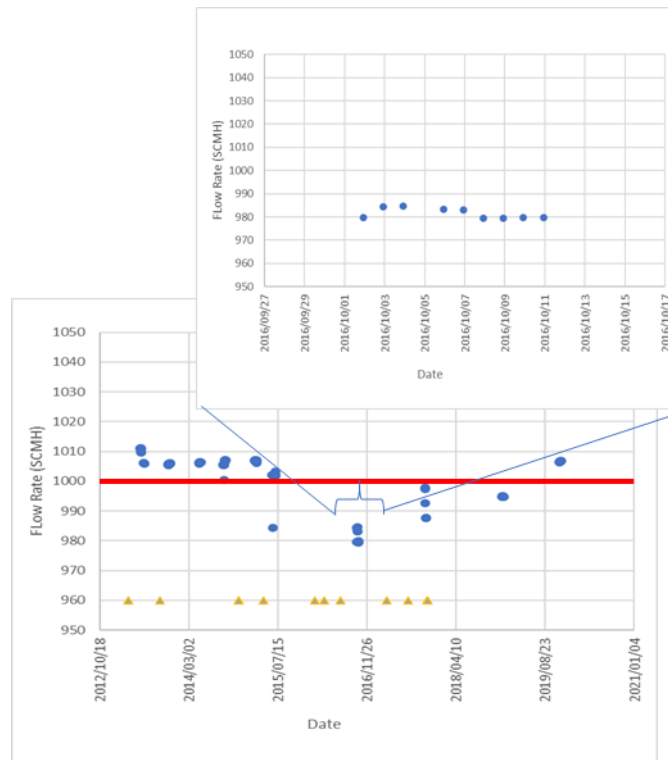


**Figure 8. Linear representation of flow rates prior and post blank acquisition 2013-2019. The red line represents the nominal flow rate of 1000 SCMH (24,000 scm per 24 hour).**

### 2.13 MDA Impact by Flow Discussion

During the 6-year interval and for the limited SamplePHD files selected from this one site, flow rates ran about 1% above the nominal and dropped 2% below the nominal when “issues” were occurring. For this one site example, the decrease below nominal in flow rates would not have impacted meeting the MDA requirements. The same data from Figure 8 are re-plotted with respect to acquisition dates below in Figure 9. Maintenance activities have also been added. An insert is added to show details around one blank collection. A special request could be made to CTBTO for

all sample files from all sites. This large collection of spectra could be used to generate a full data set on RASA flow, energy dependent MDA of daily operation for all sites. Alternately the data could also be collected from the ARRs.



**Figure 9. Temporal representation of flow rates prior and post blank acquisition 2013-2019. The red line represents the nominal flow rate of 1000SCMH (24,000 scm per 24 hour). Maintenance dates are logged as well ( $\Delta$ ).**

## 2.14 Overall Discussion

The variation in data observed for total counts appears to be greater point-to-point than does the MDA data. We have discussed that daily variations in background (i.e., perhaps Radon), have a diminished impact on MDA since signal to noise only grows as the square root of the counts. Furthermore, the representative gamma energy for Ba-140 (537.3 keV) is mostly outside of the highest impact background energies due to Radon daughter products.

### 3.0 Conclusions

SRNL has delved into performance versus maintenance data correlations for US RASA systems. This provided SRNL an opportunity to learn the manners/conventions of RASA operation as per CTBTO conventions. Data was mostly manually distilled to take the time to review the data. It was curious that there were numerous occasions where Be-7 was observable on blanks. Distilled data was worked in Excel Spreadsheets readable by GD and others. The effort was guided by how MDAs are calculated to ascertain impacts of maintenance on performance.

Contributions to overall MDA calculations (see equation 1, Appendix A) should be considered in planning MDA improvement strategies:

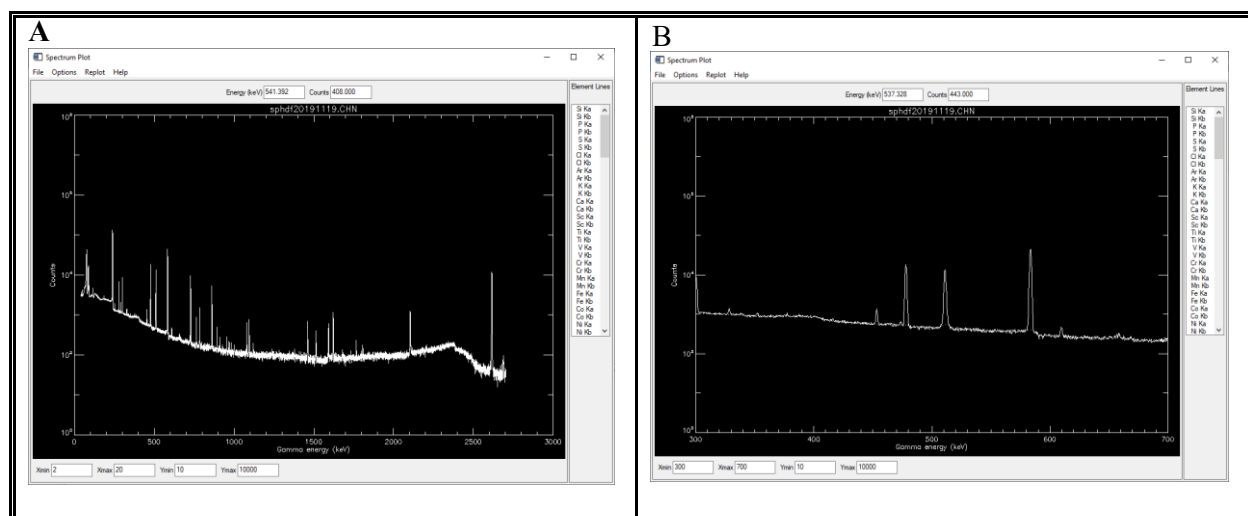
- Largest variable is clearly efficiency of Ge detectors as they are swapped-out; efficiency is directly (inversely) proportional to MDA
- Improved shielding had small/variable impact due to a square root dependence on MDA
- Flow is also directly (inversely) proportional to MDA when MDA is expressed as a Specific Activity

At this first level of inspection, it is difficult to advise specific actions for improved RASA operations. In general, RASA systems are performing within requirements. RASA performance grading appears to also be location specific bringing additional parameters such as location backgrounds, environmental conditions, and accessibility. Final interpretation of data is best done by GDMS. The structure of the data base was designed to be extensible and a working tool to gain insights into performance drifts over time.

### 4.0 Recommendations, Path Forward or Future Work

Most of this work was performed manually. This was necessary for investigators without a longstanding knowledge of RASA operation to “look” at data to gain insight into historical performance. Based on this manual approach, approaches for automated data analysis have been formed.

The SRNL team has extensive experience in synchrotron x-ray analysis software applications that can be readily applied to gamma data. The power in these tools is the ability to process many spectra quickly and pull pertinent data into large arrays for further data analysis. Indications of these abilities is the example in Figure 7 to pull flow data directly from SamplePHD files. A further example is shown below in Figure 10, where SamplePHD data can now be pulled into an existing tool package. This opens the possibility of batch peak fitting as well as batch background fitting. This would permit quick extraction of relevant data from SamplePHD files into large arrays to investigate correlations and trends over time. Furthermore, it may be possible to track persistent versus variable background variations permitting an enhanced ability to resolve instrument performance degradation faster from daily data. The example of background determination determined by Kajfosz, J. and Kwiatek, W.M. (1987), has proven a superior approach for dilute X-ray fluorescence spectra as well as for astronomical applications for cosmic backgrounds. In layman’s terms, this technique defines a “spirograph” wheel diameter that is run underneath the spectrum delineating gamma peaks from general Compton scattering.



**Figure 10. SamplePHD data pulled into existing analysis packages. Data can be read and batch process (A) as well as peak fitting and background estimation (B)**

With automation (batch processing), work could continue into MDA of sample (SamplePHD) files and impacts of variables such as flow and background activity (dynamic versus static systems discussed earlier). The foundations of automatic file distillation are mostly in place (determination of efficiency, resolution, etc.). Such automation would allow rapid distillation of results and more time data mining. If a special request were made to CTBTO, all sample files from all sites could be requested to generate full data set on RASA flow, energy dependent MDA of daily operation for all sites. Assessments on relative magnitudes could be made on variables from static versus dynamic components of the MDA to guide weighted methods for improvements.

It appears to these authors that interpretation of CTBTO guidelines is left primarily to analysts. Such automation should therefore be designed with flexibility to extract fundamental information from gamma spectra (SamplePHD files). As an example of discussion of the existing analysis in this report, it was pointed out that resolution measurements on blanks are typically only made when there is a maintenance requirement. No data representing resolution degradation as the HPGe detectors age exists. This fact is not of particular concern except that more frequent measurements could provide status of health (SOH) on resolution degradation as the detectors age. Instances where the same resolution data is used for 5+ years. We pointed out that resolution could be tracked via daily sample data. We proposed the omnipresent positron emission peak at 511 keV could be used whereas an attending analyst said he used a radon daughter line at 583 keV. Such flexibility for fundamental data extraction should be anticipated in generating an automated tool of value for GDMS monitoring RASA functionality as well as analysts.

It is our opinion that fundamental information would be defined as parameters leading to the calculation of the MDA (Appendix 1). Furthermore, the parameter space would need to be extended to permit automated extraction for the MDA at all energies within the gamma spectrum. Choosing an MDA focused on the Ba-140 energy is useful as a treaty guideline, but ultimately analysts and RASA performance monitoring would benefit from the ability to selectively monitor

those fundamental parameters across the gamma spectrum. It is also clear that flow performance (embedded within Sample PHD files) is a necessity for monitoring RASA performance as well as the determination of specific activity for any radioactive element.

## 5.0 References

Kajfosz, J. and Kwiatek, W.M. (1987) "Non-polynomial approximation of background in x-ray spectra." Nucl. Instrum. Methods B22, 78-81.

Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization. Certification of Radionuclide Laboratories. CTBT/PTS/INF.96/Rev.9, 1 August 2012.

Operational Manual for Radionuclide Monitoring and the International Exchange of Radionuclide Laboratories, CTBT/WGB/TL-11,17/18/Rev.6, 6 March 2019.

## Appendix A. Constants and Calculations

The nuclear data parameters are:

Radionuclide: Ba-140

Half-life (days): 12.75

Key line energy (keV): 537.3

Emission probability (%): 24.4

The calculation of an *MDA* (Bq) for gamma ray spectrometry systems:

$$MDA = \frac{L_D}{T \cdot \varepsilon_\gamma \cdot S_\gamma \cdot P_\gamma \cdot K_C} \quad (1)$$

Detection limit  $L_D$ :

$$L_D = 2.71 + 4.65\sqrt{\mu_B} \quad (2)$$

Standard deviation of the background,  $\sqrt{\mu_B}$ :

$$\mu_B = \sum^{ROI} Counts_i \quad (3)$$

Decay correction during acquisition time,  $K_C$ :

$$K_C = \frac{1 - e^{-\lambda_i t_C}}{\lambda_i t_C} \quad (4)$$

Where:

$L_D$  lower limit of detection at the 95% confidence level

$\sqrt{\mu_B}$  standard deviation of the background at the energy of interest (ROI is defined as  $\pm 1.25$  FWHM ( $\pm 3\sigma$ ) on either side of the hypothetical peak centroid)

$T$  acquisition live time (s)

$\varepsilon_\gamma$  attenuation corrected efficiency (counts per gamma) at the energy of interest

$S_\gamma$  correction for true coincidence summing; Since the Ba-140 decay energy at 537.3 is a single,  $S_\gamma$  was taken to equal one

$K_C$  decay correction during acquisition time

$\lambda_i$  decay constant for the isotope  $i$  ( $s^{-1}$ )

$t_C$  clock real time between start and end of acquisition (s)

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Bach, Christopher W [christopher.w.bach1.ctr@mail.mil](mailto:christopher.w.bach1.ctr@mail.mil)  
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Maxwell, Michael [mike.maxwell@srnl.doe.gov](mailto:mike.maxwell@srnl.doe.gov)  
Nuessle, Patterson [patterson.nuessle@srnl.doe.gov](mailto:patterson.nuessle@srnl.doe.gov)  
Ostrowski, Shaugn E [shaugn.e.ostrowski.civ@mail.mil](mailto:shaugn.e.ostrowski.civ@mail.mil)  
Williams, David G [david.g.williams36.civ@mail.mil](mailto:david.g.williams36.civ@mail.mil)  
Wright, Matthew S [matt.wright@gd-ms.com](mailto:matt.wright@gd-ms.com)