Total Gamma-ray Counting in Support of Arms Control

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ABSTRACT

The verification of the absence of special nuclear material (SNM) may be a key requirement in potential future warhead treaty verification. An absence verification system could use detailed gamma-ray spectral features to discriminate between benign radioactive sources, such as depleted uranium (DU), and special nuclear materials, such as plutonium (Pu) or highly enriched uranium (HEU), but that approach could reveal more information than the host may be willing to allow. Previous work has suggested that the ratio of total gamma-ray counts using a hand-held sodium iodide detector acquired with and without a thin tungsten attenuator can differentiate DU from HEU and Pu for the limited range of geometries considered, which included engineered shielding. Current work is expanding that study to include ratios obtained with different detector materials (polyvinyl toluene, Geiger-Mueller tube), varying attenuator types and thicknesses, and lower energy discriminator thresholds to find an ideal procedure with maximum discriminating power without using spectral analysis. The approach works well for bare materials but as with all passive measurements can be defeated by adequate shielding materials. The shielding materials may consist of container walls or engineered shielding inside those walls. Transmission measurements through the outer container will also be conducted to determine the impact of potential shielding hidden within the container. The transmission measurements will be performed off-axis to avoid passing through the material being surveyed. The transmission measurements will produce a sensitivity threshold that can be combined with the total gamma-ray counting results to give confidence that if SNM in a certain amount were present, it would be detected, and the lack of gamma-ray detection is due to the absence of the material and not due to additional shielding. The modeling study is being verified through laboratory benchmark measurements.

INTRODUCTION

The ability to confirm the absence of special nuclear material (SNM) is a valuable tool for potential future warhead treaty verification. The work presented here is exploring methods for performing these measurements using gamma-ray counters while protecting potentially sensitive information. As a result, methods that rely on gamma-ray spectrometry or on neutron measurements are not considered. Instead, total counts in a detector, counts above a select few energy thresholds, and gamma dose are being examined.

Previous work by the authors indicated that SNM could be distinguished from depleted uranium (DU) using only total gamma counts from a handheld sodium iodide (NaI) detector with and without a thin but high stopping-power attenuator (tungsten). An example of the notional setup is shown in Figure 1. The previous work was based on modeling and looked at a limited set of geometries, attenuators, detectors, and analysis techniques. This study is expanding on that work by increasing the number of geometries considered, attenuator thicknesses and materials, detector types, and measurement approaches. The goal is to develop a measurement approach that can identify the presence of SNM while adequately distinguish SNM from benign materials across a wide range of material types and geometries.

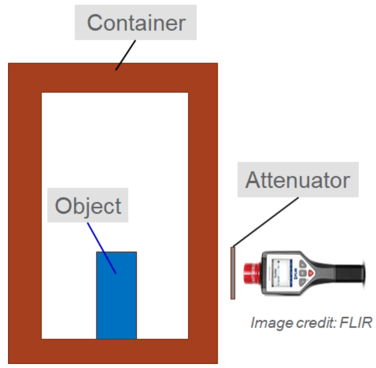

Figure 1. A notional measurement for the Simple Source Separator method

The high-level questions we seek to address in this work are:

- What is a sound measurement approach for absence measurements?
- Can this approach be modified to support attribute measurements?
- What is the minimum mass that can be observed and under what conditions?
- How much shielding can be tolerated?
- What enrichment can we differentiate from HEU (90+%)?
- Can the approach be practically implemented during an inspection?

These questions will be addressed using a combination of modeling and selected laboratory measurements as will be described in this paper.

APPROACH

"Simple" is a complicated word to define for any measurement approach. For arms control, any verification approach that would be part of a treaty must be agreed upon by both sides and should be straightforward to conduct. Complicated pieces of equipment or procedures will require complicated processes to confirm that they perform as intended, are safe and do not reveal any sensitive information. If a procedure can be designed that uses simple (possibly analog) detectors and measurement procedures while still having a good separation between different material types, then it may be more likely to be adopted into a treaty.

The methods explored in this project leverage differences in spectra of gamma-rays from different materials to confirm the absence of special nuclear materials. The measurement approach must be able to detect the presence of gamma-emitting material, and if such material is present, discriminate special nuclear material from other materials that are not accountable. The special nuclear material of interest is plutonium and highly enriched uranium (HEU), with enrichments greater than 90%. The primary unaccountable material of concern is depleted uranium (DU), as other benign gamma sources can be addressed in different ways.

Four different measurement approaches for utilizing the differences in the spectra with hand-held detectors have been considered. The initial approach measured total count rates with and without an attenuator. The combination of the ratio of the rates and the unattenuated rate enabled discrimination of HEU and Pu from DU. A second approach is to use counts rates for different lower-limit discriminator thresholds. A lower-limit discriminator would be easy to implement in hardware without enabling the recording of spectra. A third approach is to compare dose rate and count rate, or analogously total energy deposited per count. The fourth approach is to look at count rates using two different types of detectors which are selected to have different energy responses. Each of these approaches is "simple" to implement, protects sensitive information by limiting the nature of information recorded, and has the potential to leverage differences in gamma spectra to perform the detection and the SNM/DU discrimination required.

For the approach looking at counts above a lower-level threshold, it is assumed that gamma-ray counting for a few different energy thresholds is acceptable. The Trusted Radiation Identification System (TRIS) [1], a NaI-based gamma-ray spectroscopy templating system, uses over a dozen energy bins to form the template. The TRIS measurement process handles the information in these bins as sensitive information, which suggests that 12 energy bins is beyond the upper limit of what is acceptable. Current focus for this work with this approach is three sets of counts, a total count rate and two count rates with different thresholds.

A demonstration of the potential separation power of the approach using measurements with and without an attenuator is shown in Figure 2. In this measurement approach, a handheld instrument, such as an Identifinder-R400 [2] could make a 2.5 minute measurement with and without a thin sheet of highly attenuating material, such as lead or tungsten. By comparing the ratio of the two measurements against the unattenuated rate, a large separation is observed between materials like plutonium and HEU from other materials like DU and low enriched uranium (LEU). Figure 2 shows the results of a simulation of this type of measurement for a variety of materials, masses, and geometries. The vertical/diagonal lines correspond to hollow sphere geometries, while the horizontal lines correspond to solid geometries of spheres and cylinders. More details are covered in the next section.

MODELING STUDY FOR SIMPLE SOURCE SEPARATOR ALGORITHM

The Gamma Detector Response and Analysis Software (GADRAS) [3] was used to generate simulated detector responses for a variety of attenuated and unattenuated measurements. The materials considered in this study were all GADRAS default materials: DU (0.2% U-235), LEU (3.3% U-235), HEU (90% U-235), and Pu (α-phase, 4.5% Pu-240). The masses for each material ranged from 100 g to one IAEA significant quantity (75 kg for DU and LEU, 25 kg for HEU, 8 kg for Pu) [4] in 15 geometric steps. The geometries were chosen to span a wide range of possibilities for distributing the mass: solid sphere, right circular cylinder twice as long as its diameter (viewed both from the side and end-on), and hollow spheres with inner radii of 5, 10, and 15 cm. The thicknesses and sizes of the geometries were then calculated from the desired mass and known densities of the materials.

The simulated results presented in Figure 2 were generated using the default handheld Identifinder-R500-NaI with the crystal dimensions modified to match the Identifinder R400 NG that was used in the previous study (3.5 cm diameter, 5.1 cm thick). Two measurements were modeled: one bare detector measurement and a second with a 2 mm thick lead sheet in front of the detector to attenuate the emissions from the variety of materials. Natural background was injected into the spectra and subtracted off as would be the case for a real measurement. Various methods to plot the results to provide material discrimination were studied, but the displayed plot showing the ratio of the unattenuated to attenuated counts versus the unattenuated rate showed the most promise. The unattenuated counts helped to separate out fast and slow counting geometries while the ratio had some sensitivity to the average energy of the emitted gamma rays.

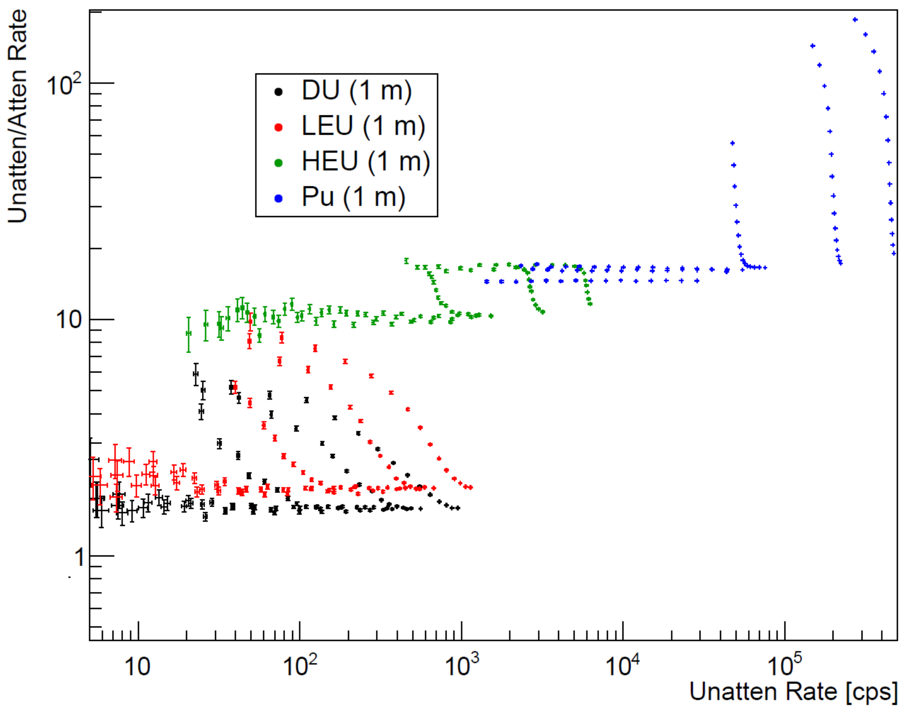

Figure 2. Simulated material discrimination with a handheld NaI detector with and without 2 mm of lead

A similar plot was made using the GADRAS default Detective-EX200, an approximately 50% relative efficiency high purity germanium (HPGe) detector. Because the method was looking at total counts, the superior energy resolution of the HPGe detector did not play a role, and the overall results also indicated good separation between Pu/HEU and DU/LEU.

Aluminum and iron were also studied as attenuator materials using GADRAS to investigate the effects of changing the atomic number significantly while maintaining the same areal density. The separation between materials was not as good. The higher stopping power of lead tends to have more differentiation between the higher and lower energy gamma rays present in the various materials.

While plots like Figure 2 are useful for giving a qualitative picture of the effectiveness of the method, a quantitative metric is desirable. To this end, confusion matrices were studied. A confusion matrix depicts the frequency at which a data point may be misclassified due to statistical similarity to other data points. An example of this approach is shown in Figure 3. The uncertainty is determined by adding in quadrature the statistical uncertainties of the 2.5-minute counts (including background measurements) and an estimated modeling uncertainty of 10% (discussed later). If both the ratio of unattenuated-to-attenuated counts and the absolute count rate for two different geometries agreed within on standard deviation, then the pair of geometries were plotted in the confusion matrix.

In Figure 3, various geometries and masses are plotted for a single material on each vertical or horizontal axis. The white and green squares are to help guide the eye for groups that are the same geometry. The geometries start with spheres and move on to the hollow spheres of increasing internal radius, then the cylinders viewed from the side and cylinders as viewed end on. For each geometry, data is listed in order of increasing mass. A detailed version of the upper left part of Figure 3 is presented on the right side of Figure 3, showing the confusion between DU and LEU geometries. For example, the DU and LEU spheres have similar responses for similar masses (upper right green box of the right figure), which is expected from Figure 2. Next, the DU spheres are being confused with the LEU cylinders viewed end on (upper left white square). These results make sense physically as a cylinder viewed end on is relatively similar to a sphere, geometrically. Other regions show less straightforward overlaps in responses. Importantly, the Pu/LEU, HEU/DU, and Pu/DU confusion matrices are almost completely empty indicating there is very little statistical overlap between those responses. Less than 2% of the total number of geometries had statistical overlap. This allows a quick comparison of the performance for different detectors and materials. For example, the same NaI detector but using 2 mm of iron had 4.1% statistical overlap and using 8 mm of aluminum had 4.3% overlap.

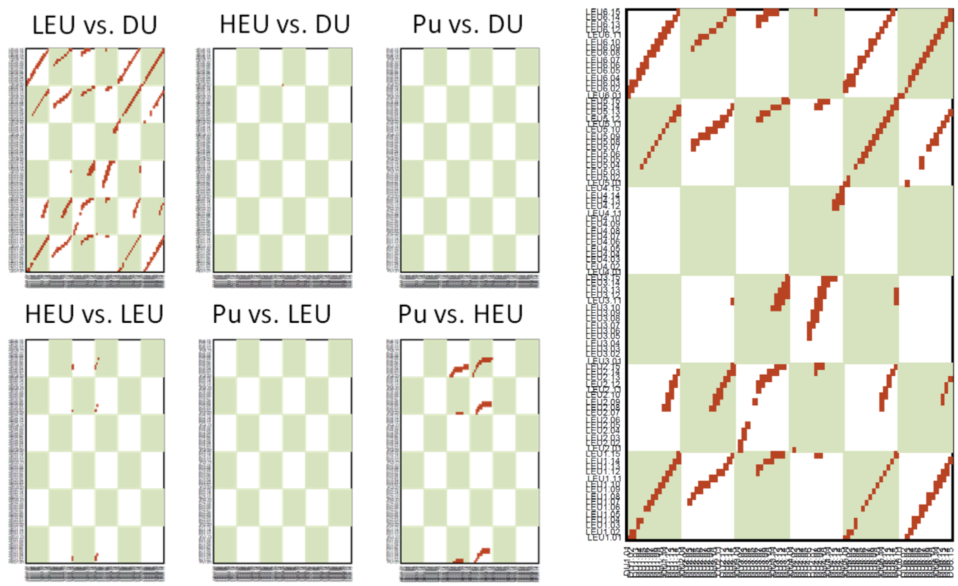

Figure 3. Confusion matrices for the NaI case with 2 mm of lead. Left) All unique combinations of the 4 materials. Right) Detail for the LEU vs. DU case

A study on the approach utilizing count rates with different lower-limit discriminators was conducted. For this study, the original geometries described above were further attenuated using 7 different thicknesses of tungsten, DU, lead, and high-density polyethylene (HDPE), resulting in 8700 combinations, to study possible impacts of shielding. An example of the results are shown in Figure 4. A neighborhood component analysis algorithm [5] was applied to this expanded set, and the top two performing thresholds were then ratioed to the total counts and plotted against each other. In general, the LEU and DU objects formed a channel between the HEU and Pu objects. The separation is sufficient that two polynomial discrimination curves could be used to separate many of the HEU/Pu geometries from the DU/LEU ones. Not surprisingly, significant overlap occurred with DU attenuated materials and bare DU objects.

An example of the approach looking at dose and count rates is shown in Figure 5. The GADRAS calculated gamma dose is divided by the total gamma-ray emission rate (leakage) and plotted against the total gamma-ray emission rate. The Pu and HEU geometries mostly separate out nicely from the DU and LEU geometries. The effects of model uncertainty need to be incorporated but prior experience with the original approach (unattenuated/attenuated) indicates that most geometries will still be distinguishable given the large amount of separation.

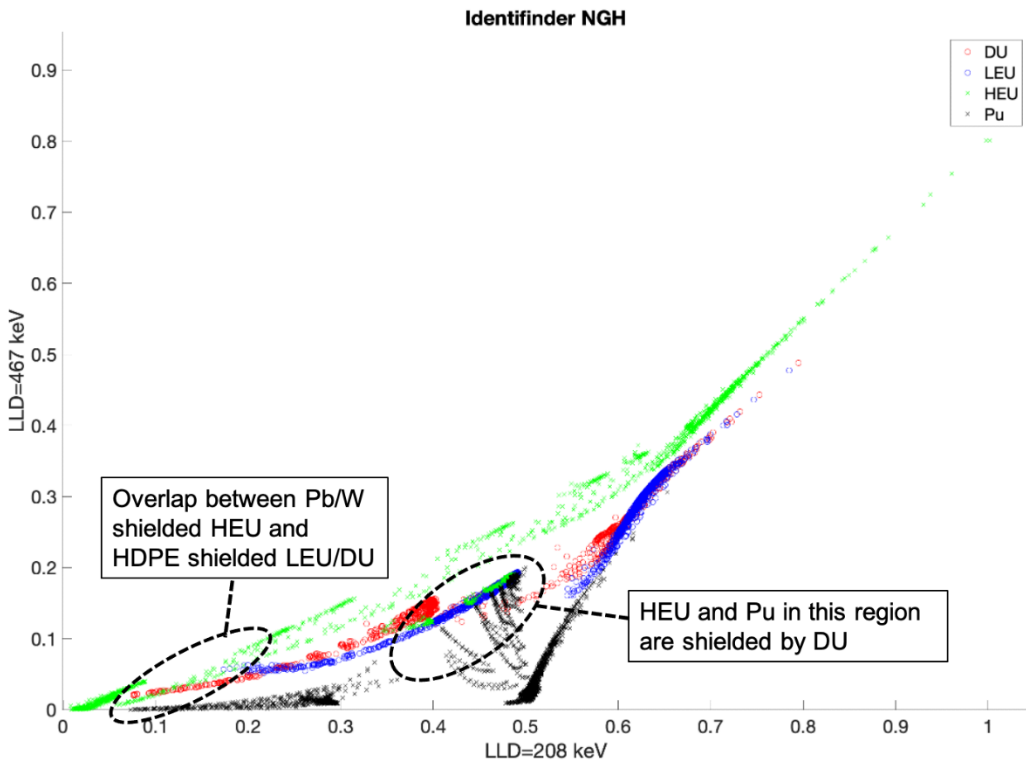

Figure 4. Discrimination of materials using ratios of counts above a threshold for the Identifinder NGH. The axes are the ratio of counts above 208 keV to total counts vs. counts above 467 keV to total counts. Results are shown for objects of various shapes, shieldings and masses.

The use of two different detectors was also investigated using the same geometries as utilized for the analysis of Figure 2. The two different detectors were a NaI detector and a polyvinyl toluene (PVT) detector as they have very different response to gamma rays as a function of energy. However, the combination of those two detectors still had overlapping geometries for 3.8% of the cases (compared to 1.8% using the attenuator). While the Pu had good separation, the HEU was not very well separated for the lighter mass cases. This can also be understood since lead attenuator has significantly more stopping power than either detector material and will therefore have a better ability to separate spectra based on the gamma-ray energies.

MEASUREMENTS

Benchmarking

Laboratory measurements were performed to determine the fidelity of the findings from the GADRAS simulations. A set of measurements was conducted using laboratory check sources. Measurements with SNM sources are planned for the near future. The laboratory check sources enable a simple comparison of detector response to the source and attenuators across a wide range of relevant gamma-ray energies.

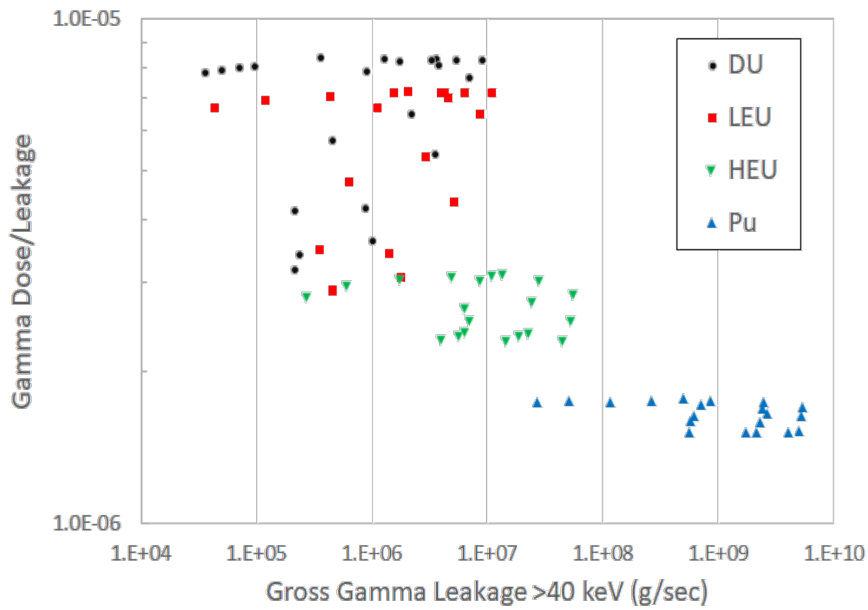

Figure 5. Plot of the ratio of the gamma dose to the total gamma-ray emission rate (leakage) versus the total gamma-ray emission rate.

The detectors that were used for benchmarking in the laboratory were: a FLIR Identifinder (NaI), an Ortec IDM-200 (50% relative efficiency HPGe), and a Geiger-Mueller counter (Ludlum 44-9 probe). These systems span a range of energy resolution and efficiency. The check sources chosen were ^{241}Am, ^{133}Ba, ^{137}Cs and ^{60}Co to span a range from 60 keV up to 1.3 MeV. Three attenuating materials were chosen: aluminum, steel, and lead. The thicknesses were chosen to span a range of areal densities from 2.4 g/cm^2 (about 2 mm of lead) to 14.4 g/cm^2 (5.08 cm of Al, 2.54 cm of steel, 1.27 cm of lead). The sources were placed 50 cm from the front face of the detector, and spectra (or counts) were collected for 300 seconds. Backgrounds with the different attenuator configurations were also collected. There was some reduction of the background counts when an attenuator was placed close to the detectors, as expected. The detector responses for all sources and all attenuators and were simulated in GADRAS and compared with measured values. Figure 6 shows the comparison of modeled and measured spectra for the bare NaI and HPGe detector. Table 1 shows the results of a comparison between attenuated and bare count rates for a set of thin and thick attenuators. If the obvious outlier (^{133}Ba response for thick lead) is ignored, the average difference between measurement and simulation is 10%.

Transmission

For absence measurements, an observation of low count rates could be due to either a lack of gamma-emitting material or to shielding within the container attenuating the gammas. Transmission measurements of the container are a likely means to address potential shielding. A transmission measurement would consist of two count rate measurements, one with and one without the container separating the source and the detector. The source likely should be collimated to reduce scattering contributions. A mCi-level ^{137}Cs source is a good candidate source.

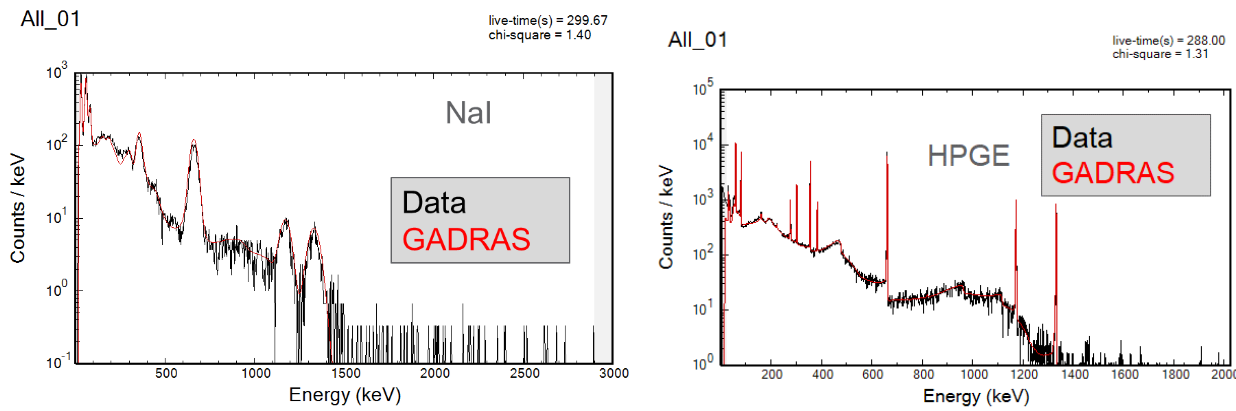

Figure 6. Left) Comparison of data and GADRAS simulation for the bare NaI detector and the four check sources. Right) The same plot but for the HPGe detector

Table 1. Comparison of results for single source NaI measurements with GADRAS simulations

Attenuator	Source	Meas. [cps]	GADRAS [cps]	Meas/ GADRAS
None	All	235.1	233.1	1.01
Al 2.54 cm	All	167.3	174.4	0.96
Fe 2.54 cm	All	85.1	90.6	0.94
Pb 0.21 cm	All	105	119.6	0.88
Pb 1.27 cm	All	37.6	41.6	0.90
None	Ba-133	94.4	82.2	1.15
Al 2.54 cm	Ba-133	58.9	54.7	1.08
Fe 2.54 cm	Ba-133	18.7	18.1	1.03
Pb 0.21 cm	Ba-133	25	25.4	0.98
Pb 1.27 cm	Ba-133	2.9	0.83	3.49
None	Cs-137	78	80.4	0.97
Al 2.54 cm	Cs-137	75.4	78.9	0.96
Fe 2.54 cm	Cs-137	44.5	49.2	0.90
Pb 0.21 cm	Cs-137	57.4	66.6	0.86
Pb 1.27 cm	Cs-137	21.2	22.8	0.93
None	Co-60	26.9	29.2	0.92
Al 2.54 cm	Co-60	26.6	30.3	0.88
Fe 2.54 cm	Co-60	18	23.5	0.77
Pb 0.21 cm	Co-60	23.1	27.9	0.83
Pb 1.27 cm	Co-60	12.3	18	0.68
None	Am-241	43.2	41.1	1.05

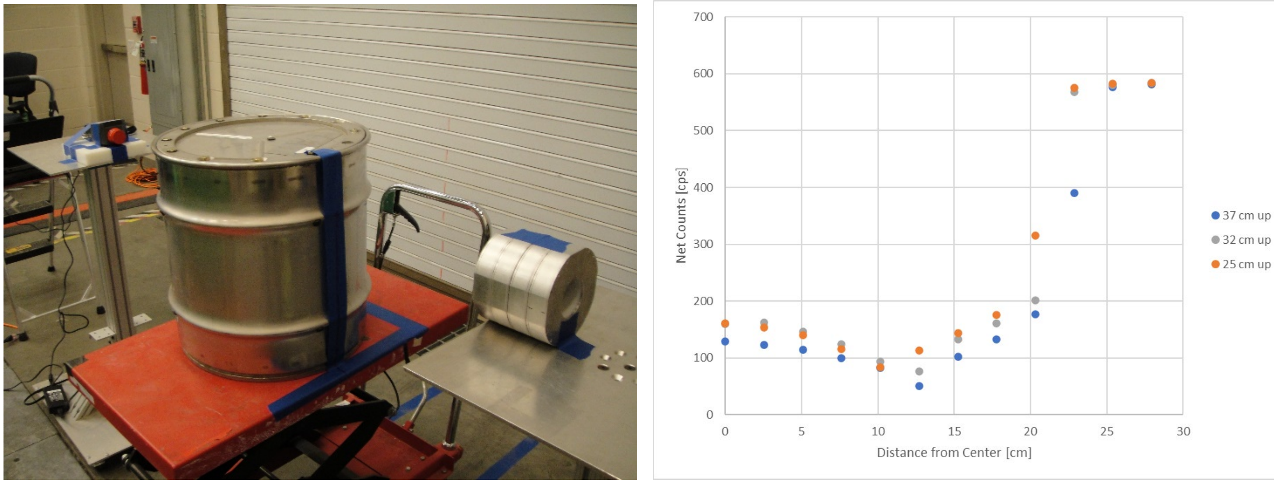

Figure 7. Left) Photograph of the transmission measurement experimental setup. Right) The net count rates observed at various horizontal and vertical offsets.

To study transmission measurements, a count/rate scan along various locations of an empty AT-400R container were conducted. A mCi-level ^{137}Cs source and an NaI-based Identifinder detector were used to collect five-minutes of data at each position. Figure 7 shows a photograph of the transmission measurement experimental setup on the left and a plot of the measurement results on the right. The measurement shows clear sensitivity to the basic internal structure of the container which was empty except for a thin, cylindrical container. If there were significant additional attenuation present, a decrease in counts would be observed. Given the known geometry and source strength, an approximate average areal density can be determined (assuming the materials are known). Importantly, the measurement gives a lower limit of material masses that the separator algorithm should be able to detect. By combining a transmission measurement with the paired attenuation measurement, either the absence of material or the presence of a certain amount of material can be determined.

CONCLUSIONS

The experimental and modeling effort to examine options for a simple source separator has made much progress over the past year. The findings of the original study that this work was based on were confirmed and various measurement approaches such as attenuator thickness and type as well as analysis techniques have been examined. The immediate plans are to continue to confirm the modeling results with more measurements. While confidence in the modeling has been strengthened through comparison with detailed measurements of both attenuation and transmission cases, the method will ultimately be tested against a variety of SNM types and configurations in the laboratory. These measurements will allow the team to address the high-level questions listed earlier in this paper. Knowing where the algorithm performs well and where it cannot confirm absence of special nuclear materials will be important for determining how this approach can be applied in a potential future arms control agreement.

ACKNOWLEDGMENTS

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