An Integrated Approach to Precision Neutron Measurements in a Dynamic Environment

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Abstract

In a nuclear facility, precise neutron measurements may need to be taken for the purpose of quantifying the mass of safeguards-relevant nuclear material. However, this material is often located in an environment with substantial and fluctuating background noise due to other nuclear material located within the same process area. To control background, the nuclear material of interest is often transported from a working area to a dedicated shielded measurement area. This transportation of the safeguards-relevant nuclear material introduces new pathways for the material to be diverted by a would-be proliferator or stolen by a thief. Likewise, it increases the time required to take these critical measurements. This work aims to remove the transportation step from the process and enable the use of high precision non-destructive assay instruments in an environment with a dynamic background signal. A methodology is demonstrated which uses dispersed neutron detectors in a mock glovebox environment to quantify a dynamic background count rate and remove it from a high-level neutron coincidence counter measurement. This methodology builds upon previous efforts to develop an algorithm for localizing and quantifying sources within this mock glovebox environment by integrating said algorithm with a background change detection algorithm, a background signal inference algorithm, and a mass prediction algorithm to form a single methodology for making near real time nuclear material accountancy measurements. The effectiveness of this methodology is quantified experimentally by measuring the mass of a Cf-252 source in an environment where several other Cf-252 sources are moving throughout the room during the measurement. Over the course of a two-hour measurement, sources were moved within the gloveboxes to create ten distinct background configurations. Analysis of the resulting data yielded a 55.4% average error in the mass measurement when our methodology is not applied, and application of the methodology reduced the average error to -2.6%. This methodology serves as a proof of concept that precise non-destructive assay measurements can be taken inline at a nuclear process facility, thus reducing possible pathways for the diversion or theft of safeguards-relevant nuclear material in addition to potentially enabling critical measurements to be taken more quickly.

1. Introduction

Any quantity of nuclear material must be accurately quantified for safety, security, and safeguards concerns. Thus, Nuclear Material Control and Accountability (NMC&A) is an essential operational aspect for any facility working with nuclear materials such as uranium and plutonium. Quantification of uranium and plutonium can be done using neutron coincidence counting. One detector capable of taking coincidence counting measurements for plutonium is a High-Level

Neutron Coincidence Counter (HLNCC). An HLNCC consists of 18 ^{3}He tubes embedded in a cylindrical polyethylene body surrounding a cadmium lined cavity [1]. The HLNCC can quantify plutonium by measuring neutrons emitted from a source in the ^{3}He tubes. The neutron counts acquired from a source can be compared to various known relationships to determine the mass of nuclear material within the cavity. For such measurements to be accurate, a known background neutron count rate must be established. In process facilities that work with nuclear material, material is often being moved, measured, or processed while mass measurements are being performed. Material that is actively being handled can cause varying background count rates at the fixed location of the coincidence counter, making accurate assay measurements difficult and complicating an NMC&A system. As a result, material processing either needs to be halted for a measurement to be taken, or the material needs to be transported to a location with a consistent neutron background count rate to be assayed. The transportation of nuclear material before it has been adequately assayed can cause additional material movements which take time and resources to complete.

In this paper, we report on the development of a proof-of-concept technique for conducting accurate assay measurements of a neutron emitting source using an HLNCC with a varying background count rate. For this proof-of-concept technique, an HLNCC located in a testbed facility shown in Figure 1. This testbed consists of an HLNCC and four mock gloveboxes, with a ^{3}He detector at each corner of each glovebox.

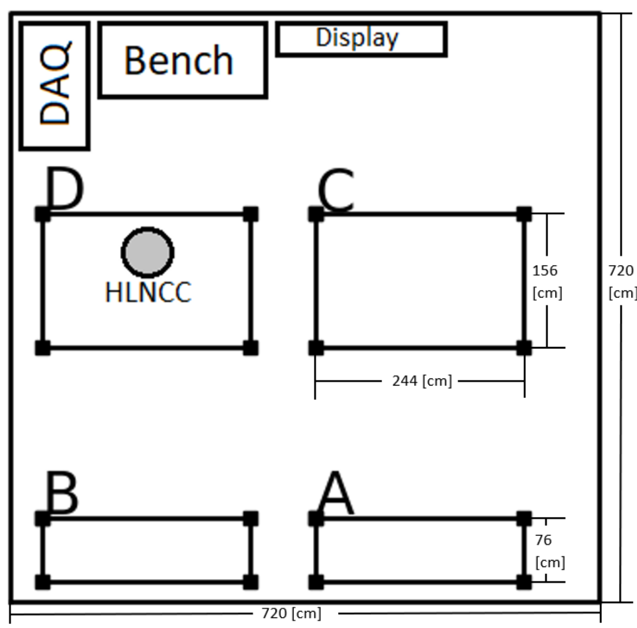

Figure 1: Layout of the testbed facility showing the HLNCC, four mock gloveboxes, and sixteen ^{3}He tubes which are indicated by the black squares at the corners of the gloveboxes. Likewise, some additional features of the testbed such as the data acquisition system (DAQ), a bench, and a display screen for data, are included at the top of the testbed. This figure is a modification of a figure that initially appeared in [2].

The HLNCC takes a measurement of a ^{252}Cf source over a period of approximately two hours, during which up to four other ^{252}Cf sources are moved around in four mock gloveboxes in the testbed facility to create several different background signals over the course of the assay measurement. Several algorithms, including an in-situ source localization algorithm, a background count rate prediction algorithm, and a background count rate change detection algorithm are integrated and are used to analyze the HLNCC count rate data to produce data that subtracts out the background count rate. This background-subtracted data can then be used to make accurate assay measurements. This technique was developed at Los Alamos National Laboratory (LANL) as part of the Dynamic Material Control (DYMAC) project. This work is a continuation of ongoing efforts at LANL, which were presented at the INMM/ESARDA joint annual meeting in 2021 [2,3]. Related work supporting the DYMAC project including work on optimizing neutron well counters, benchmarking algorithms in an NMC&A digital twin, and optimizing neutron instrument settings based on a list-mode analysis technique are being presented on ta the 2023 INMM/ESARDA joint annual meeting [4,5,6].

2. Methods

2.1 Process flow

The flow of data for this assay technique is shown in Figure 2. Data from the 16 glovebox detectors and 16 ^{3}He detectors in the HLNCC are recorded into a csv file by the DAQ every second. Only 16 of the 18 ^{3}He detectors in the HLNCC are used due to data acquisition limitations. The data is then read by a change detection algorithm, which calculates when the background count rate is stable or dynamic, as well as indicating if the background count rate has changed from one stable value to another stable value. During periods of a stable background count rate, the source localization and characterization algorithm uses the data from the sixteen glovebox detectors to locate sources throughout the gloveboxes and determine what their source strength is. Then, the background subtraction algorithm predicts what the background count rate for the HLNCC is based on the source location and strength information. The background count rate prediction is subtracted from the real time data to produce a background free dataset, which is used in part to determine the mass of the material being assayed. Note that so far this methodology has only been tested on data after it was acquired; it has not been tested live while data is actively being acquired.

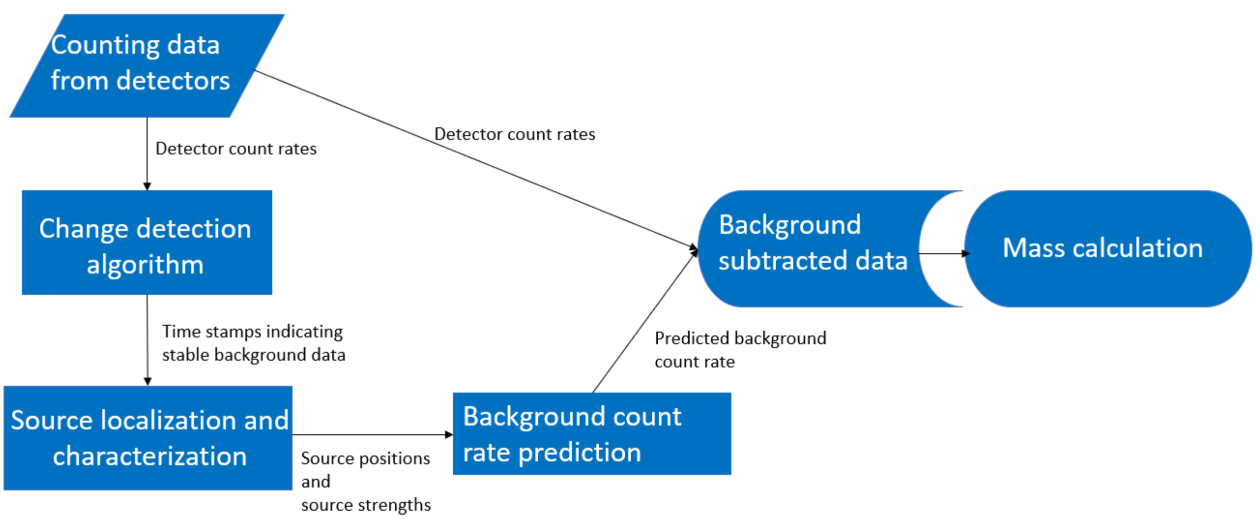

Figure 2: Overview of the process flow for the background subtraction technique described in this paper.

2.2 Background change detection algorithm

The background subtraction methodology is only valid during periods where the influence on the HLNCC count rate from external sources is relatively stable as a function of time. If a source is actively being moved about the room, this technique is not currently advanced enough to rapidly locate and characterize a source to accurately predict its background influence on the HLNCC measurement. Therefore, the change detection algorithm's function is to determine periods of stability and instability in the data. Knowing periods of stability and instability allows us to both determine when the background subtraction technique can be applied to the HLNCC counting data and provides a trigger for re-setting the source localization and characterization algorithm so that new sources can be accurately characterized.

The background subtraction algorithm examines second-by-second counting data from the sixteen ^{3}He tubes located at the corners of the gloveboxes. The basis of this algorithm utilizes the scipy module's independent t test function (scipy.stats.ttest_ind) in Python to compare p values calculated by this function against a user defined alpha value to determine if the counting data is considered stable.

The algorithm considers three possible states for the data: stable, unstable, and tentative stability. For this paper, stability is established in the count rate if, for all 32 detectors, the p value resulting from an independent t test comparing the most recent 12 seconds of counting data from a given detector and the previous 30 seconds before that is larger than the user defined alpha value. Once this initial criteria for stability have been met, the most recent 12 seconds are continuously compared to all of the data since a period of stability was established. Stable periods of data can use this technique to make background corrections to the HLNCC count rate. There is no special significance for using 12 seconds in this methodology; the value merely provided an adequately large dataset such that a mean value could be established with limited uncertainty for purposes of conducting the t tests.

Data is considered unstable if the p value resulting from a t test on the counting data of any of the 32 detectors is less than the user defined alpha value. When this occurs, the null hypothesis that there is no statistical change in the data is rejected, and a claim is made that the data is unstable. Unstable data cannot be used with this technique as a predicted background count rate cannot be adequately determined if it is rapidly fluctuating due to active movement of the sources external to the HLNCC. When the state of the system switches from stable to unstable, a change in the state of the system is claimed.

Data is considered tentatively stable immediately after a change is detected. In this state, the data counting data from the sixteen glovebox detectors is continually checked to see if a stable state arises. Initially in this state, the most recent two seconds of data are compared to the previous three seconds via an independent t test. This latter set of three data points will be referred to as the baseline set. If the p value resulting from the t test is greater than a user defined alpha value, the second most recent data point is added to the baseline data set, a new data point is read in, and the process repeats by comparing the most recent two datapoints to the baseline. Once at least 20 data points are present in the baseline, the process changes to compare the most recent 12 data points to all the data points since tentative stability was established. Once the size of the baseline data set reaches 30, indicating that 30 consecutive data points have been considered tentatively stable, the data in the baseline is relabeled as stable. If the p value is ever less than the alpha value, the data is considered unstable, and the tentative stability process is restarted using the five most recent data points supplied to the algorithm. It should be noted that the alpha value used to determine changes from stability to instability is different than the alpha value used to change from tentatively stable to stable. The alpha value used to change from stable to unstable is set to be very small to minimize false positive errors; this criterion did not need to be as strict when re-establishing stability in the data as there is potential for larger standard deviations due to smaller data sets being compared.

An example of the application of this algorithm is displayed in Figure 3. Figure 3 displays the count rate data from a ^{3}He detector linked to data acquisition channel 09 over roughly a seven-minute period. The count rate rapidly increases between 11:36:00 and 11:37:00 and rapidly decreases between 11:41:00 and 11:42:00; this counting data corresponds to a physical situation where a source was brought close to the detector for a period of approximately five minutes and then moved further away. The highlighted data in red that spans from one of the red lines to one of the green lines indicates that the algorithm has flagged that portion of the data as unstable, while all other periods of data are deemed to be stable. The yellow shaded region corresponds with logbook data of when the source handler indicated a period of stability based on clock times observed before and after moving the source.

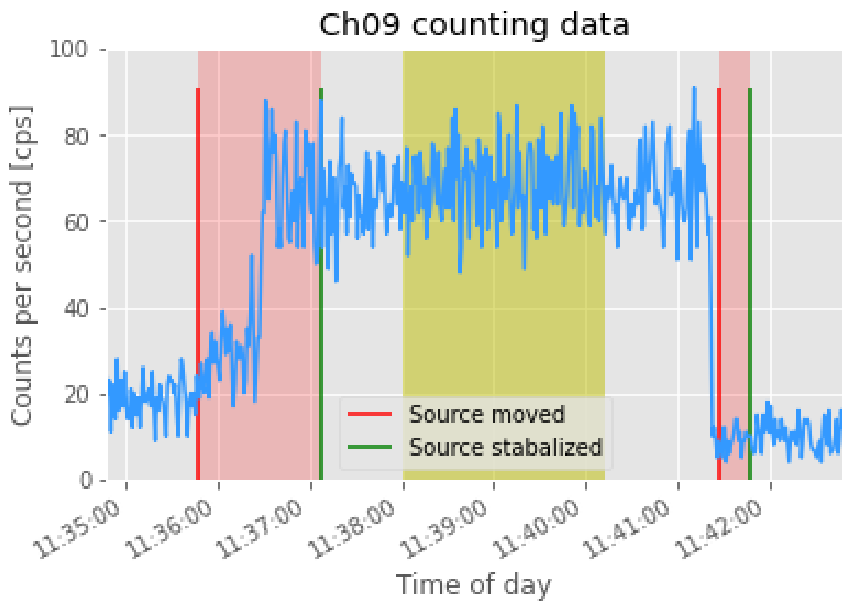

Figure 3: Example of the change detection algorithm applied to counting data from the ^{3}He detector connected to data acquisition channel 09 of 32.

2.3 In-situ source localization and characterization algorithm

The purpose of the in-situ source localization and assay algorithm is to use real-time counting data from the 16 ^{3}He detectors located at the corners of the gloveboxes to determine the position and strength of sources located in the gloveboxes. At the time this work was conducted, this algorithm was limited to locating sources within the four gloveboxes and was only able to produce localization data for one source per glovebox. To do this, the larger localization and characterization algorithm is comprised of four distinct components. Initially, the algorithm localizes a single source within one of the four zones based on template matching to normalized experimental data. Using this positional data, a prediction is made for the normalized count rate in each of the sixteen glovebox detectors. Then, this process is repeated by making independent predictions in each zone and using superposition to predict what the count rate in each detector should be based on a combination of these independent sources; this process is repeated until the solution converges to a match of the experimental data. Finally, a threshold criterion is imposed to eliminate very weak, non-physical sources which occasionally appear as part of the algorithms results. Figure 4 visualizes a sample result produced by this algorithm for four sources. The relative size and brightness of the source indicates their relative source strength, while the contours provide an illustration of relative predicted count rates, with higher count rates being expected in the lighter contours. Reference 2 talks more extensively about this algorithm.

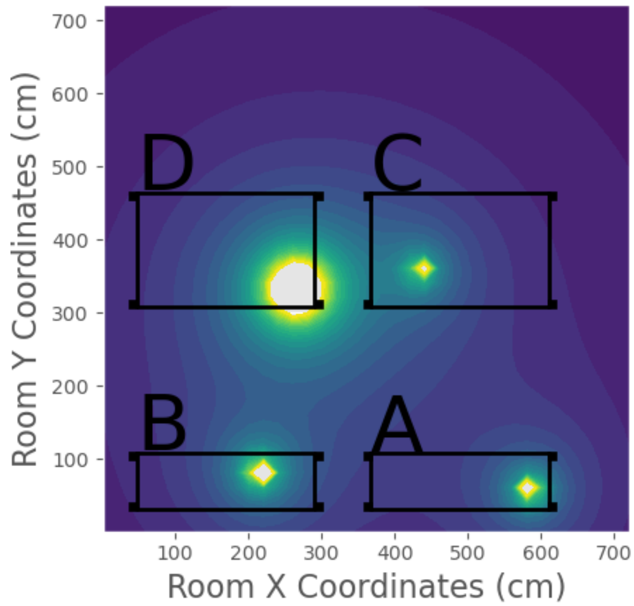

Figure 4: Contour plot visualization of an example result from the source localization and characterization algorithm. Brighter contours indicate higher expected neutron count rates.

2.4 Background count rate prediction

Eight measurements were taken at varying locations in the four gloveboxes with ^{252}Cf sources of varying strength. The resulting data was curve fit to produce a mathematical relationship relating the distance from a background source to the HLNCC and the efficiency with which the HLNCC registers counts from the source. This relationship is graphed in Figure 5 and the corresponding equation between distance and HLNCC detection efficiency is listed in Equation 1.

$$HLNCC\ Efficiency = 3.7495 * distance\ to\ source^{-1.5504} \quad (1)$$

Once the background sources are located and characterized, the background prediction algorithm uses the location information to determine the distance between each source and the center of the HLNCC. These distances are then supplied to the mathematical relationship for background prediction to determine an HLNCC efficiency value for counting data being detected from each source. Finally, each efficiency value is multiplied by the appropriate source strength and summed to find the total predicted influence on the HLNCC count rate.

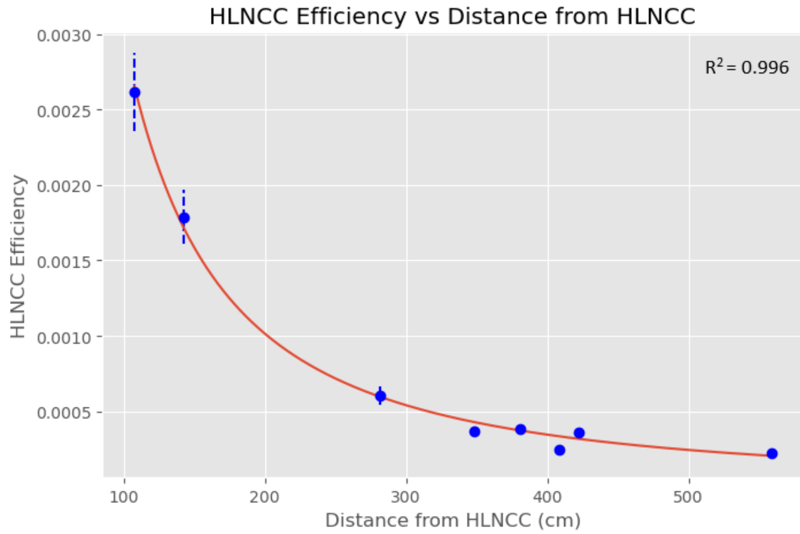

Figure 5: Plot displaying empirical relationship between the efficiency at which the HLNCC registers counts from an external background source and the distance of that source from the HLNCC. Error bars display 2σ error.

2.5 Background count rate subtraction and mass calculation

Once the predicted background count rate of the HLNCC measurement resulting from external neutron sources is known, this value is subtracted from the sum of the averages of each of the 16 HLNCC detectors for a given period of stability. The result of this calculation is the background subtracted average singles count rate of the HLNCC. Using this value, the mass of the ^{252}Cf source could be calculated using the relationship in Equation 2.

$$mass\ [g] = 2.31614 * 10^{-12} * count\ rate\ [\frac{n}{s}] \tag{2}$$

This empirical relationship was found by conducting neutron singles measurements of six previously assayed ^{252}Cf sources in the HLNCC and finding a line of best fit between the singles count rates and the corresponding known masses.

3. Results

The background subtraction proof-of-concept technique is tested by applying the technique to data from an approximately two-hour measurement. For the two-hour measurement, a ^{252}Cf of known activity is placed in the HLNCC for the entirety of the measurement. Then, four other ^{252}Cf sources are sporadically positioned throughout the four gloveboxes to create 13 distinct stable background configurations (i.e. no sources are moving) and 12 unstable background configurations (i.e. sources are actively being moved). Note that for several of the distinct configurations, several sources were placed near each other within the same glovebox to mimic a larger, single source in that glovebox. The goal is to use the technique described above to accurately identify the 13 distinct configurations and make a mass measurement during those periods of stability while also identifying the 12 periods of instability and refraining from using those periods to make a calculation as the HLNCC background count rate is changing to rapidly for this technique in its current state to accurately subtract out the background influence.

The performance of this technique relied in part on the user defined alpha values for detecting changes from periods of stability to instability, which will be referred to as alpha 1, and changes from instability back to stability, which will be referred to as alpha 2. The acquired data was analyzed using a variety of alpha 1 values ranging from 1.0E-12 to 1.0E-7 and alpha 2 values ranging from 0.005 to 0.05. Over this range of values, the technique described above predicted 15 periods of stability over the specified time range and 14 periods of instability in 13 of the 21 different combinations tested. Eight combinations resulted in 16 periods of stability with 15 periods of instability. In each case, all 13 periods of stability were correctly identified; the identified stable periods beyond 13 occurred during periods the source handler had manually indicated as being unstable. The additional periods of stability were less than a minute in length. Because all 32 detectors are currently used to detect changes, even if their count rates are on the order of a few counts per second, a very small increase or decrease caused by a variety of events such as cosmic radiation or human interference may lead to changes in count rate stability that are not reflected in the configurations set by the operator.

For each test case, the technique produces a background corrected mass estimate and an uncorrected mass estimate with no adjustment to the HLNCC count rate for each period of stable data. Examples from a test case with an alpha 1 value of 1E-8 and an alpha 2 value of 0.05 are plotted in Figure 6 for the uncorrected mass estimates and Figure 7 for the corrected mass estimates.

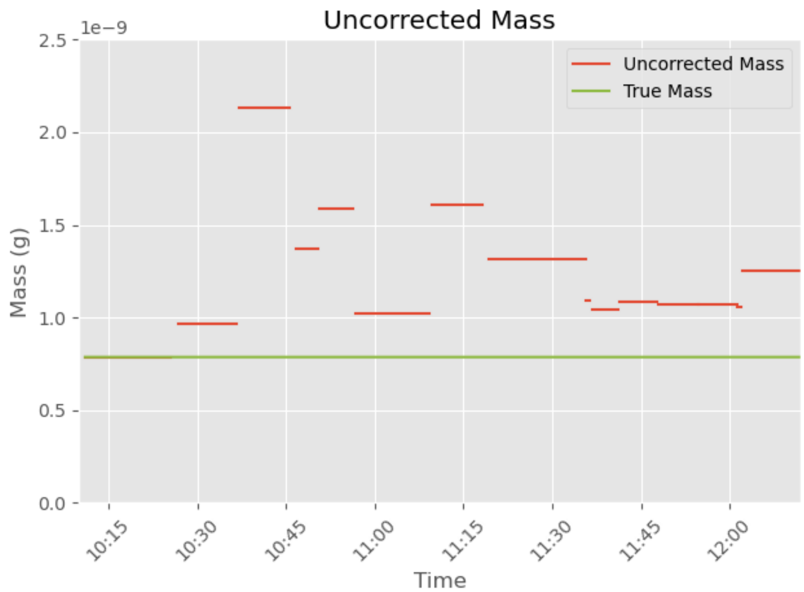

Figure 6: Resulting uncorrected mass predictions of a ^{252}Cf source over the course of a roughly two-hour measurement with thirteen distinct background source configurations.

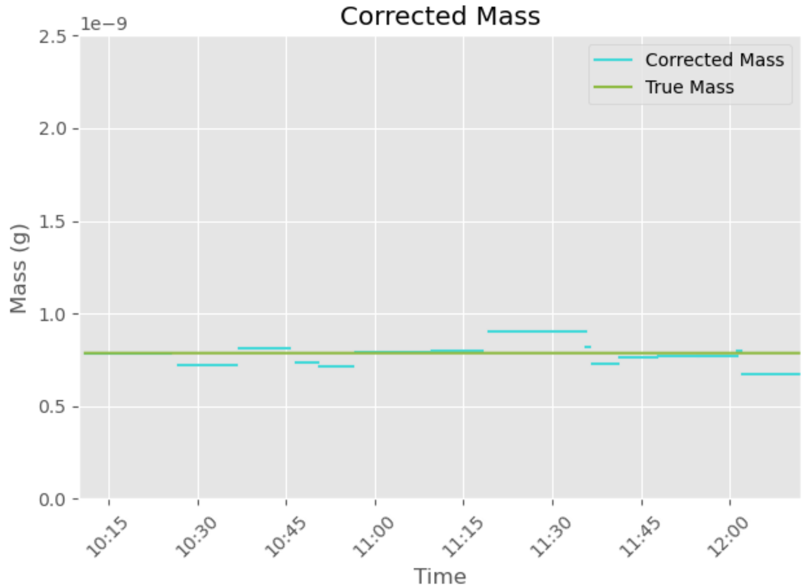

Figure 7: Resulting corrected mass predictions of a ^{252}Cf source over the course of a roughly two-hour measurement with thirteen distinct background source configurations.

In Figure 6, the uncorrected mass is an overprediction in every case as the HLNCC count rate is elevated due to neutrons being counted from external sources. Figure 7 shows significantly improved mass estimates when the background signal is subtracted out, although the technique performance of the technique varied across the different configurations. To help quantify the performance of this technique, the error in mass between the prediction and the true mass of the sample was calculated for each stable period in the data, and then these errors were averaged. Across the different combinations of alpha 1 and alpha 2 values tested, the average error in the uncorrected mass estimate ranged from 53.6% to 56.6%, while the average error in the background corrected measurements ranged from -3.6% to -1.8%. Averaging the results of all cases produced an average error in mass estimate of 55.4% using uncorrected neutron data and an average error in mass estimate -2.6% using the background corrected neutron data. The technique performed best in the case where alpha 1 was set to 1E-08 and alpha 2 was set to 0.05, which resulted in an uncorrected average error in mass of 56.5% and a corrected average error in mass of -1.8%.

4. Conclusion

The methodology presented in this paper describes a technique for conducting background subtraction from HLNCC measurements in an environment with a dynamic neutron background by identifying stable regions of neutron counting data, localizing and characterizing any sources external to the HLNCC, and predicting a background count rate which is subtracted from the HLNCC counting data. The modified counting data following background subtraction allows for accurately assaying the mass of the nuclear material inside the HLNCC. The background subtraction technique successfully serves as a proof-of-concept technique for taking nuclear material measurements with a fluctuating background signal successfully. By quantifying the background influence of external sources and subtracting this influence from an HLNCC measurement, a significant improvement is seen in the accuracy of the HLNCC measurements. The presented background subtraction technique demonstrates how NMC&A measurements may be taken in a facility that handles nuclear materials without the need to transport that material to a specified measurement location, thus reducing possible diversion pathways. Current work is investigating improving the background influence prediction relationship to be a function of both an x and y distance from the HLNCC to better account for varying geometry such as walls in the testbed facility. Before this technique can be applied to an actual process facility, future work will need to investigate how multiple sources within a single glovebox or dispersed sources can be accounted for, the inclusion of gamma detectors to utilize multiple possible nuclear material signatures, and how to better account for error propagation across the multiple connected components of this technique.

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