Improving the Confidence Associated with Passive Total Neutron Counting in the Nuclear Weapon Disarmament Verification Process

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Abstract:

Passive total neutron counting is an important tool in the nuclear weapon disarmament monitoring and verification process proposed by the International Partnership for Nuclear Disarmament Verification (IPNDV). In the process, neutron measurements of given treaty accountable items (TAIs) are repeated multiple times in different locations and environments, and the measured neutron count rates are expected to remain unchanged. However, neutrons are heavily scattered in the environment, and the change in location or geometry of the environment can produce varying results in neutron measurements which can deteriorate the confidence of passive total neutron counting. In this paper, we have studied different kinds of neutron detection instruments and methods in various environments to determine the effects of the environment on passive total neutron counting and to develop recommendations and procedures to minimize and take these environmental factors into account. As a result, the moderated ^{3}*He proportional counter was the most promising type of instrument in terms of how the change in an environment impacted the measured neutron count rates. However, even with the* ^{3}*He counter, the environmental influence increased rapidly with the source-to-detector distance (SDD). For example, with one-meter, two-meter, and three-meter SDDs, the maximum difference in count rates between outdoor and indoor measurements was 10.06(7) percent, 18.6(3) percent, and 28.1(5) percent, respectively. To mitigate the impact of the measurement environment, we propose to use a bare* ^{252}*Cf reference source measured in the same geometries as the nuclear TAIs to estimate the influence of the environment on the measured neutron count rates. Using this technique in the same conditions as above produces indoor predictions that differ by 2.55(3) percent with a one-meter SDD, 0.334(5) percent with two-meter SDD and 1.93(4) percent with a three-meter SDD from the actual indoor measurements.*

Keywords: nuclear disarmament; IPNDV; neutron counting; neutron scattering

1. Introduction

Since 2014, International Partnership for Nuclear Disarmament Verification (IPNDV) has been identifying challenges associated with nuclear disarmament verification and developing potential procedures and technologies to address them [1]. Among other things, IPNDV has developed a 14-step nuclear weapon dismantlement framework [2]. Monitoring and verification technologies associated to different steps have also been studied in more detail [3]. Declarations, inspections, the chain of custody, and various passive radiation measurements and information barriers play a key role throughout the disarmament process [4], [5]. Information barriers are employed to prevent the release of classified information while allowing meaningful conclusions [6]. Note that IPNDV considers disarmament verification activities from the viewpoints of both the host (the owner of the nuclear explosive devices (NED)) and the inspecting party.

This article primarily concentrates on passive total neutron counting, which is used to confirm the presence of neutron-emitting special nuclear material (SNM) in the treaty accountable items (TAIs). In this article TAI refers both to complete NED and/or dismantled SNM from the NED. In passive total neutron counting, the neutron count rate is measured at a certain distance using a neutron detector, and the neutron rate is at some certain level proportional to the total mass of the SNM in the TAI. During the disarmament verification process, the TAI may be measured multiple times in different locations and environments, and the neutron count rate should remain unchanged throughout the process. A dramatic drop in the count rate could indicate reduction of the mass of the SNM during the process, which can deteriorate confidence in the disarmament verification process.

Neutron counting is prone to environmental influence, as neutrons are easily scattered and reflected by the environment, including floor, walls, air, etc. [7], [8]. The neutron count rate acquired in passive total neutron counting is determined by neutrons not only coming directly from the source but also through scattering processes. Consequently, changes in the environment can change the portion of scattered neutrons. From the confidence point of view, it is important to comprehend how much the environment can influence the measured neutron count rate.

In this paper, we have studied how the environment influences the neutron count rates of different kinds of neutron detection instruments and methods, including moderated ^{3}He proportional counter, ^{6}LiF/ZnS(Ag) scintillator-based detector, as well as LaBr$_3$ and NaI(Tl) scintillators that detect neutron induced high-energy gamma-rays. The moderated ^{3}He proportional counter can be considered the gold standard for neutron detection. However, due to the global shortage of ^{3}He isotope [9], it was important to study how the alternative neutron detection technologies perform in passive total neutron counting. In this study, we have also developed and tested an additional monitoring procedure to mitigate the impact of environmental factors on the confidence associated with neutron counting and the overall monitoring and verification process.

2. Materials and Methods

Experimental work was conducted at the Radiation Metrology Laboratory (RML) at the Finnish Radiation and Nuclear Safety Authority (STUK). RML is responsible for maintaining national measurement standards of ionizing radiation in Finland and provides calibration services for various companies and institutions.

2.1 Experimental Setup

Due to the limited measurement time available in the RML, different types of commercially available neutron detectors were irradiated simultaneously with an industrial neutron source, and the total neutron count rates were measured as a function of source-to-detector distance (SDD) in three different environments: 1) outdoors, 2) the neutron source placed approximately in the middle of the RML's calibration hall (subsequently referred to as Indoor I) and 3) the neutron source placed one meter away from the concrete wall at the back of the calibration hall (subsequently referred to as Indoor II). The outdoor measurements were conducted in the asphalt parking plot belonging to STUK. The calibration hall is 16 meters in length, 5.5 meters wide, and 5 meters in height and has approximately one-meter-thick concrete walls. The measurement setups are depicted in Figures 1, 2 and 3.

Figure 1: The setup depicting indoor measurements (Indoor II setup) with a moderated ^{252}Cf source placed on the tripod one meter away from the concrete wall at the back of the calibration hall.

In the indoor measurements, detectors were placed side by side on the electronically movable table (height of one meter), and the neutron source was placed approximately 1.1 meters above the ground on the tripod. In the outdoor measurements, detectors were placed on the stationary table (height of 0.8 meters), and the neutron source (placed

Figure 2: The setup depicting outdoor measurements. Moderated ^{252}Cf source was placed on the table trolley and the source-to-detector distance was measured with a laser range finder.

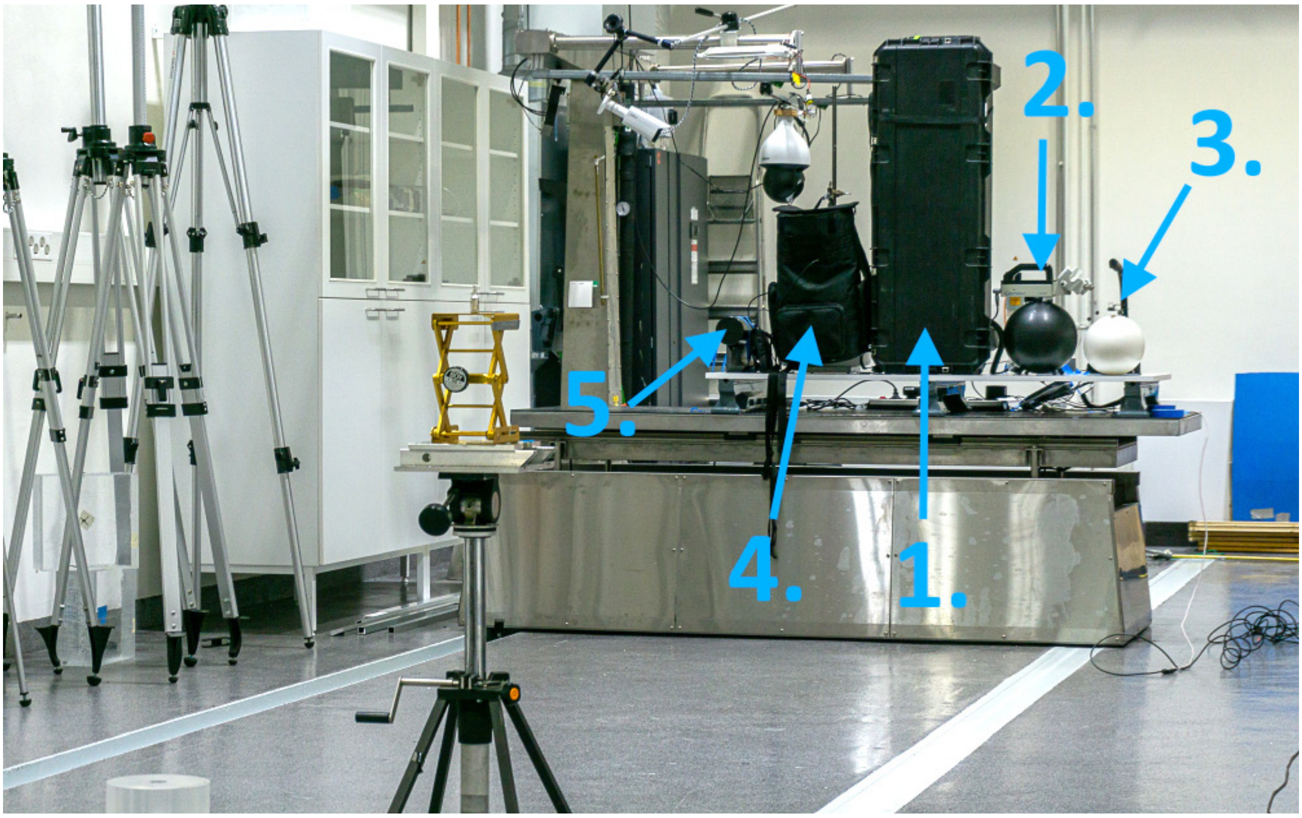

Figure 3: The setup depicting indoor measurements (Indoor I setup) with bare ^{252}Cf source placed on the tripod in the middle of the calibration hall. Detectors numbered in the figure are: 1) Environics RanidPort, 2) Berthold LB 6411, 3) Mirion SN-D-2, 4) Environics RanidPro200 ^{6}LiF/ZnS(Ag) neutron plate, 5) Environics RanidPro200 LaBr$_3$.

approximately 0.9 meters above the ground) was moved with a table trolley. Neutron count rates with bare and moderated source were measured in the SDD range of 1 to 7 meters with one-meter intervals. Based on the IPNDV verification process, the neutron detector cannot be placed in contact with a bare or containerized NED due to safety reasons. Therefore, a minimum distance of one meter, also discussed by the experts representing the nuclear-weapon states in the IPNDV meetings, was selected for the present study. The maximum distance of seven meters was due to calibration hall dimensions preventing us from operating the movable table beyond seven meters from the source (Indoor I measurements).

The SDDs were measured from the centre of the neutron source to the front face of the central detector (NaI(Tl)), and the distances to other detectors were calculated with the Pythagorean theorem by also knowing the distances between the central detector and the other detectors. With each SDD, data was collected for several minutes to ensure enough data was gathered to limit statistical error. Background measurements without the neutron source were performed before and after each of the three series of measurements, and the background count rate of the latter measurement was subtracted from the measured neutron count rates.

A neutron-emitting ^{252}Cf isotope was used as the source of neutrons in this study. The neutron energy spectrum of ^{252}Cf-spontaneous fission [10] with an average neutron energy of 2 MeV is very similar to special nuclear materials (SNM), such as ^{239}Pu [11] and ^{235}U [11] used in nuclear weapons. The ^{252}Cf used in this study had a calculated activity of 179.5 MBq (nominal activity of 500 MBq with a reference date of Nov-15-2016) emitting 2.15x10^{7} neutrons per second. The source (made by QSA Global Inc.) was a cylindrical sealed capsule with a single stainless-steel encapsulation surrounded by a custom aluminium shield.

The measurements were performed with a bare and moderated ^{252}Cf source. Moderation reduces the speed of fast neutrons and increases the portion of slower (thermal) neutrons, making them more susceptible to neutron capture. In the nuclear disarmament verification process, the exact geometry of the TAI is not known, e.g., the nuclear material can be shielded, moderated, or bare. Therefore, it was important to study whether moderation enhances or diminishes the influence of the environment on the results of passive total neutron counting. We used a partially hollow cylinder of polymethyl methacrylate (PMMA) to moderate the ^{252}Cf source. The moderation was 9 cm thick from the sides, 8 cm thick from the bottom, and 6.8 cm thick from the top.

2.2 Neutron Detectors

The detectors used in this study are depicted in Figure 3. Detector in the centre (numbered 1 in the figure) is the

RanidPort Mobile, a radiation portal monitor made by Environics (a subsidiary of Bertin Technologies). RanidPort features a four-liter volume 4"x4"x16" NaI(Tl) scintillator and is capable of indirectly detecting neutrons through high-energy gamma radiation using the energy gate of 3.5 to 8.5 MeV [12]. The natural gamma-ray background above 3.5 MeV is reasonably low (see Section 3). In the present experimental conditions, additional high-energy gamma-ray signals are caused by neutron capture reactions occurring in the detector and the environment, and there are also prompt fission gamma rays being emitted directly from the source. The biggest advantage of using an indirect neutron detection method based on high-energy gamma-ray signals is that conventional 0 – 3 MeV gamma-ray spectroscopy can be performed simultaneously with the same detector. In the disarmament verification process, conventional gamma-ray spectroscopy performed behind the information barrier using a medium energy resolution detector can provide useful information from the TAI [13].

The second detector was the Berthold LB 6411, which is used as the reference neutron ambient equivalent dose meter in the RML. Berthold LB 6411 is a ^{3}He and methane gas-filled proportional counter with wide neutron energy measuring range from thermal to 20 MeV. The gas-filled counter tube inside the detector is surrounded by a 250 mm polyethylene moderator sphere moderating fast neutrons to lower energies.

The third detector was the Mirion SN-D-2 neutron dose probe (also moderated ^{3}He gas-filled proportional counter) connected to the Canberra Colibri TTC survey meter. The data from the SN-D-2 probe was not utilized in this study but was used for internal purposes at STUK to compare the neutron count rates between Berthold LB 6411 and SN-D-2.

The fourth detector was the ^{6}LiF/ZnS(Ag) neutron detector plate made by Symetrica and is part of the RanidPro200 backpack-operated radionuclide identification device made by Environics. The plate is composed of two ^{6}LiF/ZnS(Ag) neutron screens coupled to a wavelength-shifting plastic (polyvinyl toluene) and not containing any moderation other than the external plastic shell and the nylon backpack fabric surrounding the plate.

The fifth detector was the 2"x2" $LaBr_3$ scintillator which is also part of the Environics RanidPro200 backpack. The $LaBr_3$ detector is capable of indirect detection of neutrons through high-energy gamma radiation (energy gate of 3.5 MeV to 5.7 MeV) as the NaI(Tl) detector. In this study, the $LaBr_3$ detector was taken out from the backpack and placed on the measuring tables.

3. Results

Figure 4 shows the background subtracted data sets including estimations for uncertainties caused by the statistics and distance, measured in Indoor I, Indoor II, and Outdoor measurement setups. The uncertainties for the distance were calculated using the standard method of error propagation. The background count rates in Outdoor measurements were 0.007(3) cps for the ^{3}He proportional counter, 0.14(2) cps for the $LaBr_3$ scintillator, 0.23(2) cps for ^{6}LiF/ZnS(Ag) scintillator and 13.4(2) for the NaI(Tl) scintillator. Inside the calibration hall, the background rates were somewhat higher. In the Figure 4, the neutron count rate has been plotted as a function of source-to-detector distance (SDD) for each detector using moderated and bare ^{252}Cf source. For each data set in Figure 4, we have fitted an inverse power function (equation (1)) using the least-squares fitting weighted with the absolute statistical uncertainty of each data point. The fitting was performed using the curve_fit function of Python's SciPy-library [14].

$$R = \frac{A}{r^{x}} + B, \tag{1}$$

where r is the SDD and A, B, and x are parameters of the inverse power function. Similar function with parameter x fixed to two was used in the ref [15]. Note that the parameter $A + B$ equals the count rate at 1-meter SDD. Parameter B is needed to improve the performance of the fitting function. Without scattering, the intensity of radiation is inversely proportional to the square of the SDD, and the parameter B would be equal to zero. However, in the case of neutron radiation, the neutron scattering will cause the intensity to diverge from the inverse square law and result in a lower exponent than two and non-zero values of B.

The parameters gained by fitting equation (1) to data sets are shown in Table 1. As depicted, none of the data sets comply with the inverse square law, but all result in a lower exponent than two. The relative percentage differences (RPD) of A, B, and x parameters when comparing the indoor measurements to the outdoor measurements are also listed in Table 1. For the ^{3}He proportional counter, the parameters A, B, and x change the least between the measurement setups. Thus, from the confidence point of view, results obtained by the ^{3}He proportional counter are the least influenced by the environment and can be considered the most promising type of neutron detector compared to the other detectors tested in this study. Note from Figure 4 that the neutron count rates measured with the ^{3}He proportional counter in the Indoor I configuration at longer SDDs (> 5 m) are comparable to the rates in the Indoor II configuration with both the bare and the moderated source. This is probably caused by the neutrons that are backscattered from the back wall to the detectors. At seven meters SDD, the back wall was approximately two meters from the detectors, and thus with longer SDDs, the

Configuration	^{3}He counter			^{6}LiF/ZnS(Ag)			LaBr$_3$			NaI(Tl)		
	A	x	B	A	x	B	A	x	B	A	x	B
	%[a]	%[a]	%[a]	%[a]	%[a]	%[a]	%[a]	%[a]	%[a]	%[a]	%[a]	%[a]
Indoor I, bare ^{252}Cf	**140.1 (6)**	**1.61 (2)**	**0.9 (2)**	**793.5 (12)**	**0.465 (1)**	[b]	**58.6 (12)**	**0.56 (3)**	**-7.6 (15)**	**4263 (4)**	**1.156 (3)**	**300 (3)**
	0.448 (3)	7.02 (6)	220 (50)	107.9 (3)	55.0 (4)	[b]	44 (1)	66 (4)	2000 (800)	56.81 (8)	28.02 (8)	3800 (400)
Indoor II, bare ^{252}Cf	**147.5 (4)**	**1.50 (1)**	**-0.4 (2)**	**1307.2 (9)**	**0.5848 (6)**	[b]	**88.2 (9)**	**0.64 (2)**	**-10.5 (11)**	**5722 (4)**	**0.947 (2)**	**-65 (4)**
	5.73 (3)	13.6 (1)	50 (21)	242.5 (6)	43.4 (3)	[b]	116 (2)	60 (2)	3000 (1100)	110.5 (2)	42.5 (1)	710 (80)
Outdoor, bare ^{252}Cf	**139.5 (6)**	**1.73 (1)**	**-0.8 (2)**	**381.7 (9)**	**1.033 (6)**	**-23.3 (7)**	**40.8 (4)**	**1.62 (3)**	**-0.33 (11)**	**2718 (3)**	**1.647 (3)**	**-8.1 (7)**
	[c]	[c]	[c]	[c]	[c]	[c]	[c]	[c]	[c]	[c]	[c]	[c]
Indoor I, mod ^{252}Cf	**56.3 (3)**	**1.66 (2)**	**0.4 (1)**	**2909 (2)**	**1.120 (2)**	**27 (2)**	**109.5 (4)**	**1.13 (2)**	**0.3 (4)**	**6776 (4)**	**1.418 (2)**	**232 (2)**
	4.44 (4)	4.72 (5)	320 (120)	3.741 (4)	20.47 (5)	141 (8)	2.17 (2)	32.5 (5)	130 (140)	45.68 (5)	5.217 (9)	410 (7)
Indoor II, mod ^{252}Cf	**58.7 (2)**	**1.56 (2)**	**-0.1 (1)**	**3948 (3)**	**0.963 (2)**	**-178 (3)**	**144.8 (5)**	**1.02 (2)**	**-5.6 (6)**	**8137 (5)**	**1.095 (2)**	**-223 (4)**
	9.03 (7)	10.7 (2)	80 (200)	40.78 (5)	31.59 (8)	170 (3)	35.0 (3)	39.0 (6)	500 (100)	74.95 (7)	26.86 (5)	199 (5)
Outdoor, mod ^{252}Cf	**53.9 (4)**	**1.74 (2)**	**-0.2 (1)**	**2804 (3)**	**1.408 (3)**	**-66 (1)**	**107.2 (7)**	**1.67 (2)**	**-0.9 (2)**	**4651 (4)**	**1.50 (1)**	**-74.7 (11)**
	[c]	[c]	[c]	[c]	[c]	[c]	[c]	[c]	[c]	[c]	[c]	[c]

[a] Relative percentage difference (RPD). [b] The fitting was done by setting the parameter B fixed and equal to zero. [c] Reference environment for RPD.

Table 1: Parameters A, B and x obtained by fitting equation (1) to the data sets. The relative percentage difference (RPD) in fit parameters A, B and x are shown for each indoor data set when the outdoor measurement setup was used as the reference environment. Uncertainties of the fitted parameters and RPD values are given in parenthesis.

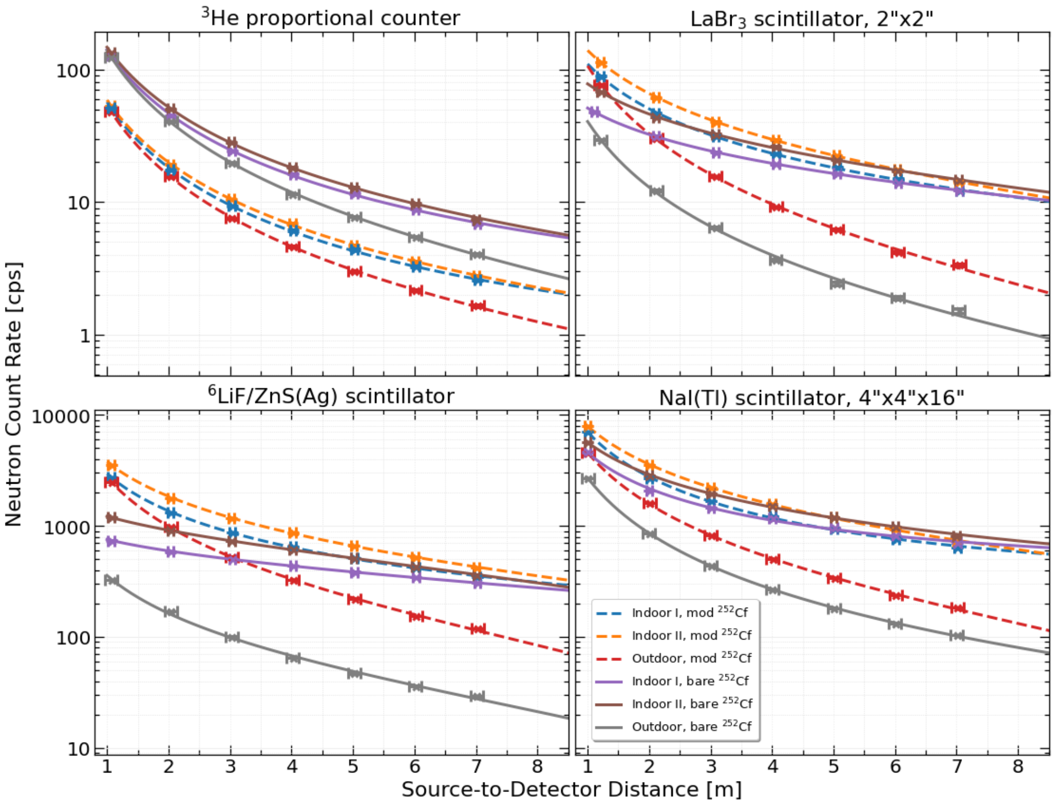

Figure 4: Neutron count rate vs. source-to-detector distance for various neutron detectors using the bare and moderated ^{252}Cf source in three different measurement setups. Equation (1) was fitted for the data points and the resulting parameters are given in Table 1. The uncertainties for neutron count rates and SDDs are marked for each data point.

Indoor I setup was somewhat similar in geometry to the Indoor II setup.

For the other detectors evaluated, the influence of the environment on neutron count rates was more dramatic. The worst performer seemed to be the ^{6}LiF/ZnS(Ag) scintillator-based neutron detector, as it had the largest difference in parameters A, B, and x values between configurations. Also, the indirect high-energy gamma-ray-based neutron detectors did not perform as consistently as the ^{3}He proportional counter, where a significant environmental sensitivity even at 1-meter SDD is visible $(A + B)$. In all measurement setups, the highest count rates were associated to NaI(Tl) and ^{6}LiF/ZnS(Ag) detectors.

As can be seen from Figure 4, the moderation had a substantial impact on the neutron count rates measured with all detector types. For instance, with the ^{3}He proportional counter, the measured count rates were reduced by almost half when the source was moderated. For the other detectors tested, the moderation impacted oppositely: the count rates were higher when the source was moderated. Consequently, it is relevant for the inspectors monitoring the nuclear disarmament to know if the shielding or moderation i.e., the surrounding material of the TAI has changed in between neutron measurements.

4. Discussion

Although the moderated ^{3}He proportional counter was the most reliable neutron detector tested, the measured neutron count rates of the ^{3}He proportional counter still varied significantly in various environments. Figure 5 shows the relative percentage differences in the neutron count rates vs. the source-to-detector distance when comparing outdoor measurements to indoor measurements (circles). As depicted, the RPD increases rapidly when the distance increases. In Outdoor/Indoor II data set, the difference is 18.6(3) percent with distance of two meters but increases to 28.1(5) percent with three meters. Therefore, the neutron detector should be placed as close as possible to the object which is to be verified. Also, for simplicity, a fixed distance and preferably a similar type of detection instrument should be used in every neutron measurement performed during the disarmament verification process.

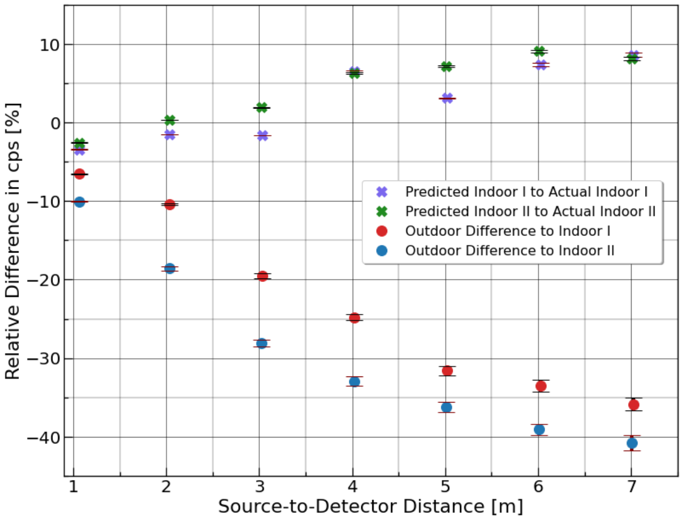

Figure 5: Relative percentage difference in neutron count rate of ^{3}He proportional counter vs. source-to-detector distance. In the figure, circles represent how much the measured neutron count rates of the outdoor measurements differ from the indoor measurements, and crosses show how much the predicted neutron count rates differ from the actual measured indoor values. Uncertainties are included for each data point.

Notice that even with the SDD of 1.07 meters, there would still be a 10.06(7) percent difference in the neutron count rates between the Outdoor and Indoor II measurement configurations. Assuming that the neutron count rate correlates with the SNM mass of the TAI, then the 10.06(7) percent reduction in the Outdoor count rate would indicate that a significant amount of SNM has gone missing between the Indoor II and Outdoor measurements. As shown earlier, if the SDDs get longer the differences in count rates become larger, which can further deteriorate the confidence in the disarmament verification process.

To mitigate the environmental impact on the confidence of neutron measurements in the nuclear disarmament verification process, we propose adding a ^{252}Cf reference source measurements to it. In the modified process, the reference source is always measured in the same geometries as the TAIs. This approach is based on the assumption that the TAI and ^{252}Cf count rates have similar behaviour as a function of measuring environment. If the assumption is valid then the TAI and ^{252}Cf measurement data can be used to predict the TAI count rates (R) in new environments, see equation (2).

$$\frac{R_{252\mathrm{Cf},\,\mathrm{location}\ 2}}{R_{252\mathrm{Cf},\,\mathrm{location}\ 1}} = \frac{R_{\mathrm{predicted\ TAI},\,\mathrm{location}\ 2}}{R_{\mathrm{TAI},\,\mathrm{location}\ 1}}. \qquad (2)$$

The performance of the method is examined in Figure 5, where we assume that the moderated ^{252}Cf source is a sensitive TAI. In the figure, crosses show how much the predicted (using equation (2)) indoor neutron count rates differ from the actual measured values. As an example, using one-meter (1.07 m exactly), two-meter (2.03 m exactly) and three-meter (3.03 m exactly) SDDs, the predicted and measured values of Indoor II differ by 2.55(3) percent, 0.334(5) percent, and 1.93(4) percent, respectively. These values demonstrate the potential associated with the proposed technique.

The introduced ^{252}Cf reference source can also be applied for other purposes. For example, it can be employed to test and calibrate the performance of the host-provided neutron detection instruments. By comparing data from new and older measurements performed in similar conditions as well after calculating the required decay corrections one can make conclusions about the device performance and make the necessary corrections if needed. By applying the lessons learned from this study such instrument testing/calibration should be performed in as an open environment as possible using a short (preferably constant) SDD. Between inspections, the host could store ^{252}Cf reference source in a sealed container.

5. Conclusions

In this paper, we show that all the tested neutron detection instruments and methods are sensitive to source shielding/moderation. Therefore, changes on those should be communicated to the inspecting party. The selection of an optimal neutron detector for verification use is important. Neutron monitoring simplifies if only one type of instruments is used. In the neutron measurements as short measurement distances as possible should be employed to minimize the influence of the environment on the measurement results (using a constant measurement distance would facilitate the analysis work even further). Testing a host-provided instrument with a well-calibrated ^{252}Cf reference source in an open environment and with a short measurement distance adds confidence to the overall process (instrument performance check). Between inspections, the host could store such reference sources in a sealed container.

The study also shows that the confidence provided by the neutron measurements can be significantly enhanced by performing measurements with the bare ^{252}Cf reference source in the same environments and geometries as the actual measurements of the TAIs. In the future, one could continue the studies of this approach by using more realistic TAI surrogates. Note that the neutron count rates recorded from the TAIs in different environments are probably classified information, but their comparisons may not be. Therefore, part of the data analysis may have to be conducted behind the information barrier.

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