Detection and Localization of Weapons Grade Plutonium with an Array of NaI Detectors

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Abstract An array of scintillators is a multipurpose sensitive detection system of photons and neutrons while also providing directional sensitivity. Several simulations were performed with Geant4 software to optimize the design of such an array for neutrons. A crucial design criterion was an excellent detection and localization capability, and the system must be practical for operational field work. A large array, containing four 4 L NaI detectors, can fulfil these criteria. Such an array, clad with 3 - 5 cm of PVC, has a neutron counting efficiency which is of the same order of magnitude as the state-of-the art commercial neutron detectors based on helium tubes or plastic (LiF/ZnS). For a count time of 12 minutes, the array can detect 4 kg of weapons grade plutonium at a distance of 50 m. The detection time can be essentially shortened down to a few minutes by increasing the thickness of the PVC up to 10 cm.

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Keywords gamma radiation, neutrons, detection, locationing, nuclear weapons, CBRNE, national defence, research, comprehensive defence approach

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Asekelpoisen plutoniumin havainnointi ja paikantaminen NaI matriisi-ilmaisimella

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Tiivistelmä Tuikeaineisiin perustuva matriisi-ilmaisin on monikäyttöinen fotonien ja neutronien havaitsemisjärjestelmä, joka myös tuottaa säteilylähteen suuntatiedon. Useita simulointeja tehtiin Geant4-ohjelmistolla tarkoituksena optimoida tällaisen ilmaisimen rakenne neutroneille. Ratkaisevina suunnittelukriteereinä olivat erinomainen havaitsemis- ja paikannustehokkuus ja soveltuvuus kenttätyöhön. Suuri matriisi, joka sisältää neljä neljälitraista NaI-detektoria, täyttää nämä kriteerit. PVC-muovilla (3 - 5 cm) päällystetyn matriisin neutronien havaitsemistehokkuus on samaa suuruusluokkaa kuin parhaissa kaupallisissa heliumputkissa tai muovipohjaisissa ilmaisimissa (LiF/ZnS). Matriisi voi havaita 4 kg aselaatuista plutoniumia 50 metrin päästä 12 minuutissa. Havaitsemisaikaa voidaan olennaisesti lyhentää muutamaan minuuttiin lisäämällä PVC:n paksuutta (10 cm).

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Asiasanat gammasäteily, neutronit, havaitseminen, paikannus, ydinaseet, CBRNE, maanpuolustus, tutkimus, kokonaismaanpuolustus

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Detektion och lokalisation av vapenbart plutonium med scintillationsdetektorer i form av en matris

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Referat

Scintillationsdetektorer i form av en matris är ett mångsidigt och känsligt detektionssystem för fotoner och neutroner samtidigt som den ger riktningskänslighet. Flera simuleringar utfördes med Geant4 för att optimera utformningen av en sådan matris för neutroner. Ett avgörande designkriterium var en utmärkt detektions- och lokalisationsförmåga, och systemet måste vara praktiskt för operativt fältarbete. Ett stort system, som innehåller fyra NaI-detektorer (4 liter), kan uppfylla dessa kriterier. En sådan matris, klädd med 3-5 cm PVC, har en neutronräkningseffektivitet som är av samma storleksordning som toppmoderna kommersiella neutrondetektorer baserade på heliumrör eller plast (LiF/ZnS). På en räknetid av 12 minuter kan systemet upptäcka 4 kg vapenbart plutonium på ett avstånd av 50 m. Upptäckningstiden kan i princip förkortas till några minuter genom att öka tjockleken på PVC upp till 10 cm.

Klausul

Den här publikation är en del i genomförandet av forskningsfinansiering av Försvarets vetenskapliga delegation. (www.defmin.fi/matine) De som producerar informationen ansvarar för innehållet i publikationen. Textinnehållet återspeglar inte nödvändigtvis statsrådets ståndpunkt.

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Contents

1 Introduction

Detection of neutron sources is important for national security. Most development efforts in this regard have thus far been focussed on portal monitoring, a context in which the source-detector distance is short, typically 2 m (GAO 2011). Other national security applications would benefit from a detection system that can detect and localize a neutron source from much longer distances extending to 100 m or beyond. In principle this is possible for plutonium and other neutron-emitting sources because they produce penetrating neutron radiation that can be difficult to conceal by shielding. Detection of uranium from long distances is much more difficult because of the low emission rate of photons and neutrons.

Several large-area arrays have been designed for long-range neutron detection (Marseden 2012, Maurer 2012). Most often, such systems are based on moderated helium tubes, boron-lined proportional counters, or other designs (LiF). A moderator-free detector can be made directionally sensitive for thermal neutrons through shielding and collimators. Peurrung et al. (1999) have constructed such a detection system using an array of 23 He-3 tubes positioned side by side to constitute an area of 1 m^2. In front of the detector, a hexagonal aluminium grid (honeycomb) coated with B-10 absorbs thermal and epithermal neutrons. Test measurements showed that a Cf-252 source emitting 1.2×10^8 n/s could be detected within seconds at a source-detector distance of 100 m, and the authors concluded that a moderated neutron source emitting 3×10^5 n/s could be detected at distances over 50 m for counting times of 1000 s (c.f. Section 2 for the source term of a nuclear weapon).

Studies conducted in Sweden (De Geer 1990) indicated that a nuclear weapon containing plutonium in the core and depleted uranium in the tamper could be detected with a passive neutron detection system at a distance of 120 m whereas a gamma spectrometer, utilising U-238 photons at 1001 keV, could detect the weapon at a distance of 15 m, assuming a 15 min counting time for both measurements. However, a shield (borated polyethylene) around a plutonium pit may prevent neutron detection, and lead or other heavy metals may hinder detection of emitted photons.

A detection distance of 100 m for plutonium seems to be realistic as was verified in 1989 on the Black Sea by the Americans and the Russians who carried out joint stand-off measurements on an actual warhead. For a short review, see Appendix 1.

The present study focuses on one single multi-purpose detection system that has the following properties:

- High detection efficiency for gamma and neutron radiation
- Gamma spectrometric capability (nuclide identification)
- Robustness and reliability
- Source localization capability
- Low cost

An array of NaI detectors could fulfil all the requirements. An NaI detector is a common work horse used widely in environmental monitoring and nuclear security. Previously it has been demonstrated that an efficient portal monitor can be built from a 4L NaI detector clad in PVC (Holm 2013). The energy region 10 - 3000 keV is used for gamma spectrometry and the energy region 3000 – 10000 keV for neutron counting. The chlorine atoms in the PVC convert thermal neutrons to a cascade of high-energy photons. In principle, the array is scalable and can be made as large as is needed for a specific application. The present study deals with neutron detection and source directional estimation of a 2x2 NaI array. The photon response of such an array is analyzed elsewhere (Toivonen 2024).

2 Neutron Emission Rate of a Nuclear Weapon Containing Plutonium

A notional nuclear weapon model was published in 1990 by US and Russian scientists (Fetter 1990). This has been recently revisited by a German group using Monte Carlo techniques (Kütt 2021).

Nuclear weapons emit gamma and neutron radiation. It is of particular interest to understand the emission rate of neutrons from a warhead containing plutonium. The relevant Fetter weapon model represents 4 kg of weapons-grade material having 93.3 % of Pu-239 (Table 1) in the form of a shell having a diameter of 10 cm. The plutonium is in the centre surrounded by a beryllium reflector and tungsten or uranium tamper. Conventional HMX explosive comprises the next shell, and the whole structure is encapsulated in an aluminium case.

There are two mechanisms producing initial neutrons in the nuclear material: spontaneous fission and alpha decay producing neutrons through (α, n) reaction in low-Z materials. The notional weapon model as described by Fetter contains oxygen impurities and therefore some neutrons are produced by alpha particle reactions. However, this mechanism is of secondary importance. Omitting this neutron production mechanism yields, for a pit of 4 kg weapons-grade plutonium, a neutron emission rate of 249,100 neutrons/s by radioactive decay (Kütt 2021).

The neutron production rate from the plutonium pit is not the same as the number of neutrons emitted by the weapon system as a whole. The number of neutrons is increased in the pit and the reflector, but the tamper, explosives and aluminium case absorb neutrons. Based on data in Table 1, the surface emission rate of the notional war head is 199,400 neutrons/s (Kütt 2021).

Table 1. Neutron emission rate in nuclear material and isotope composition of a notional nuclear weapon (Kütt 2021, Fetter 1991).

	T1/2 [years]	Neutron emission rate [neutrons/g/s]	Composition (Fetter model) [%]
^{238} Pu	87.7	2.63×10^{3}	0.005
^{239} Pu	2.441×10^{4}	0.0152	93.3
^{240} Pu	6561	1031	6.0
^{241} Pu	14.329	1.7233×10^{-3}	0.44
^{242} Pu	3.75×10^{5}	1722	0.015
^{241}Am	432.6	1.47	
Other/oxygen		(α, n) reaction	0.2

3 NaI Array and Symmetry Model

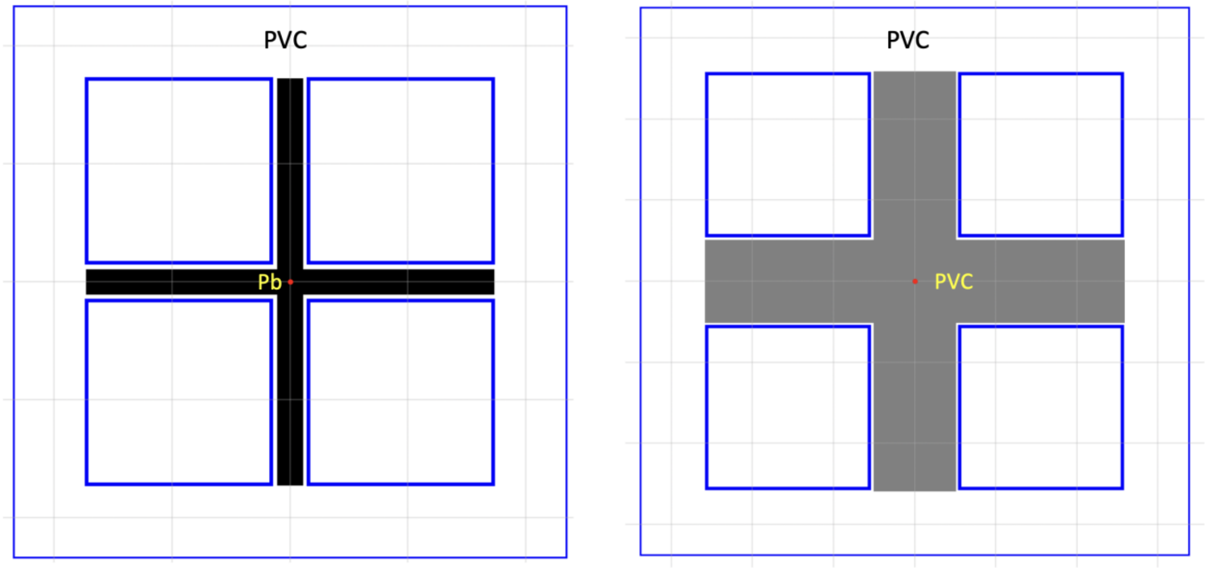
A symmetrical array of NaI detectors is not only a sensitive detection instrument for gamma emitters but it is also an excellent tool for source directional estimation. We have developed a simple and robust model, known as *Symmetry Model* (Toivonen 2024), for the estimation of the azimuth (2D array) and the elevation angle (3D array) of a source. The performance capability of the model has been verified with Monte Carlo simulations and experimental studies. The azimuth of a source emitting low-energy photons can be estimated with an accuracy of 4 - 6 degrees and in special cases, without any systematic error, with a precision of about one degree, depending on statistical variations.

The *Symmetry Model* works for any radiation depositing energy symmetrically all over the array. The model works not only for the primary incident radiation when the particles travel on a straight line from the source to the detector, but it also works with the scattered radiation, assuming the environment is symmetrical relative to the array. The present paper is intended to provide evidence for the capability of the NaI array and the *Symmetry Model* to detect and localize neutron sources.

A directionally sensitive array, based on four NaI detectors with volume of 4 L, is described in Figure 1. The detectors are separated with lead plates (1 cm thick) which attenuate photons travelling through one detector to another. This design improves directional accuracy of photon measurements and removes many coincidence events which are not useful for source localization. Lead plates are not needed for the neutron detection, and in fact, they impair neutron detection efforts, and they should be replaced with PVC, when the focus is on neutron detections.

For neutron detection, the whole NaI array is covered with PVC (2 – 5 cm). PVC contains hydrogen and chlorine atoms. Hydrogen is an efficient moderator of neutrons and chlorine captures thermal neutrons and creates a cascade of high-energy photons. Gamma cascades, with total absorbed energy above 3000 keV in the array, are a neutron signature.

Figure 1. Arrays of 4 NaI detectors with dimensions of 10x10x40 cm^3. Left: multipurpose design for photon and neutron detection. Right: optimized design for neutron detection. In both cases the whole array is covered with PVC.



4 Efficiency and sensitivity

The intrinsic efficiency of the array at the distance *r* is defined as

$$\varepsilon_i(r) = \frac{A\varepsilon(r)}{4\pi r^2} \tag{1}$$

where $\varepsilon(r)$ is the counting efficiency at the distance *r* and *A* is the cross-sectional area of the array. Simulations performed in vacuum provide the true intrinsic efficiency. The response curve should be a straight line as a function of the source-detector-distance (*r* >> 5 m). At the face of the array the area of the NaI detectors is 0.08 m^2. However, the total area of the array, including the PVC cover, is 0.11 m^2. In the calculation below a value of 0.1 m^2 is used for the array cross section.

The sensitivity of the instrument can be defined in an alternative way by using the reference neutron flux *nv* defined as

$$nv = \frac{N_0}{4\pi r^2} \tag{2}$$

where N_0 is the neutron emission rate of the source (neutrons/s). Then the sensitivity of the instrument is

$$N_s = \frac{c}{nv} \tag{3}$$

where c is the detected neutron count rate (cps). The unit of N_S is cm^2 (counts.cm^2/neutrons).

5 Simulations

All simulations were conducted using Geant4 (Agostinelli 2003) and the SWORD interface. All materials were defined in terms of isotopic abundance as opposed to elemental composition. All simulations were conducted using the QGSP_BIC_HP reference physics list using the Quark Gluon String model and the Binary Cascade model with high precision neutrons. QGSP_BIC_HP is identical to QGSP_BIC except that neutrons of 20 MeV and lower use the High Precision neutron models and cross sections to describe elastic and inelastic scattering, capture and fission. Low energy electromagnetic physics were replaced with Penelope.

Radioactive decay was enabled. Cf-252 was employed as a surrogate for WGPu neutrons. This isotope is frequently encountered in this role and its suitability for this purpose has been confirmed (Taggart 2020). The source activity was 100 MBq; therefore, its neutron emission rate was 1.156E7 n/s. The counting time varied between 5 - 150 s depending on the source-detector distance. In simulations for long distances 12 NaI arrays were deployed at 30 degrees intervals. This arrangement provided good counting statistics at all distances (uncertainty below one percentage).

The direct point source simulations for given source-array distances were conducted in a world space consisting of two opposing hemispherical volumes, the upper being 1000 m in radius and comprised of air (1.29 kg m^{-3}) and the lower comprised of land or water with the same radius. Both detector and source were positioned at 1 m above the interface of the two hemispheres. The simulated land environment consisted of silicon dioxide (silica, SiO_2). The sea water salinity was 3.5 % (1.9 % and 1.08 % for Cl and Na, respectively).

6 Counting Time for Source Detection

Signal to noise ratio is a good indicator of the detection capability of the array. A common acceptance criterion, for example in gamma spectrometric analyses, for the existence of the signal (*S*) in the presence of the background (*B*) is the "3 sigma" rule:

$$\frac{S}{\sqrt{B}} > 3 \tag{4}$$

where the factor 3 refers to a false positive probability of 1:1000 (Gaussian distribution; the exact abscissa is 3.09).

For a source emitting N_0 particles per second, Equation (4) provides the minimum data acquisition time t required for a statistically significant observation:

$$t = \frac{9B}{[N_0 \varepsilon(r)]^2} \tag{5}$$

For a Cf-252 source, $\varepsilon(r)$ is calculated with Monte Carlo simulations. The data are fitted to a power function $y = C/r^n$ in two parts.

In the environment there is no natural isotope emitting photons above 3000 keV. The highest significant photon energy is 2614 keV (Tl-208, radon decay product). However, cosmic radiation contains some high-energy particles which contribute to the background at energies above 3000 keV.

The most important particles of cosmic origin are neutrons and muons. The neutron flux is of the order of 40 – 200 neutrons /(m^2s) (Kouzes 2008). Muons, heavy particles travelling in a straight line, arrive to the Earth with an intensity of about 1 muon/(cm^2min), i.e. 170 muons/(m^2s) (Jovanovic 2018). Most of these events can be eliminated with energy discrimination (a muon travelling 1 cm in NaI deposits energy of about 8 MeV).

Cosmic primary and secondary particles interact in the atmosphere, but they also interact with high-Z solid materials especially, producing additional particles. This may create radiation, known as “ship effect”, which increases neutron flux near large structures such as a ship at sea (Koutzes 2007).

The background count rate *B* can only be estimated experimentally. A value of 1.6 cps for one 4L NaI has been reported (Holm 2013). We may then assume for an array of four such detectors that $B \cong 6$. This can also be justified with the following reasoning. The cross-sectional area of the array is about 0.1 m^2. Then the number of cosmic neutrons passing the array is about 10 n/s, assuming a flux of 100 n/(m^2s). The muons will further increase the count rate, but the intrinsic efficiency is by no means near 100 % and most of them can be discriminated by their high energy (>> 10 MeV). Therefore, the estimated background seems to be realistic.

7 Capability of an NaI Array to Detect and Localize a Neutron Source

7.1 Detection Efficiency

The following analysis aims at understanding the efficiency of the NaI array – when varying the amount of PVC in the system - to detect neutrons in different environments.

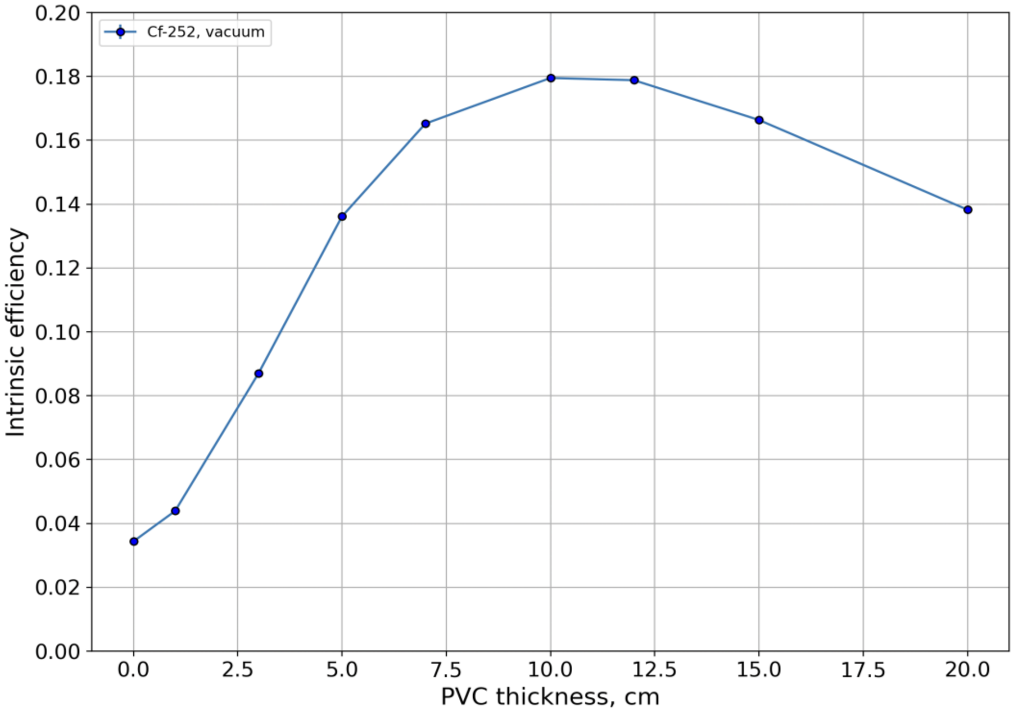
7.1.1 Intrinsic efficiency

The PVC thickness has an impact on the counting efficiency of the NaI array. The hydrogen atoms in PVC moderate neutrons effectively and then the thermal neutrons generate a cascade of high-energy photons in reactions with chlorine atoms. However, PVC also absorbs neutrons. There is, therefore, an optimum thickness of the PVC layer for photon production. Figure 2 shows that this thickness is about 10 cm.

A PVC thickness of 10 cm would make the array heavy and cumbersome for operational usage as a gamma spectrometer. Therefore, the multipurpose photon/neutron design of the NaI array has only 3 cm PVC. This layer improves intrinsic neutron efficiency from 3.4 % to 8.7 % (Figure 2).

The design of an operational instrument should allow for the adding of supplementary layers of PVC around the array when the focus of the measurements is neutron sources rather than gamma emitters. On the other hand, for the most sensitive neutron detection the design should be flexible enough to allow removal of the Pb slabs from the centre and replace them with PVC.

Figure 2. Intrinsic efficiency of the NaI array as function of the PVC thickness around the array for neutrons from Cf-252 source.



Thermal neutrons

The intrinsic efficiency of the array for *thermal neutrons* was studied in vacuum simulations by exposing the array to an incident beam of 0.025 eV neutrons. The intrinsic efficiency, when using PVC cladding of 3 cm, was 0.69 but was reduced to 0.56 for a PVC cladding of 5 cm. The reason for this observation is that PVC is not needed for neutron thermalization and therefore many interactions take place at the outer surface of the PVC, with the photons originating from chlorine having a smaller probability of hitting the NaI scintillators.

7.1.2 Impact of environment on counting efficiency

In Figure 3 the intrinsic counting efficiency of the NaI array for fast neutrons emitted by Cf-252 is compared with the true counting efficiency for neutrons in the environment. The true counting efficiency of the array is essentially higher than the intrinsic efficiency. It is about 20 % up to a source-array distance of 10 m and then it increases, the reason being neutron interactions with the atoms of the environment which then have an impact on the slope of the efficiency curve as a function of distance. Neutron scattering from the ground creates epithermal and thermal neutrons and they provide most of the counts in the array above 3000 keV. A detailed analysis of the impact of the environment and PVC cover is given in Table 2 at a source-array distance of 20 m. Air and ground amplify the signal by a factor of 7.5 – 9.5 relative to vacuum.

Figure 3. Relative counting efficiency of the NaI array with 3 cm PVC cover for neutrons emitted by Cf-252 as a function of distance. The lower curve, simulated in vacuum, is the true intrinsic efficiency for the fast neutrons emitted by Cf-252 (the response should be a straight line; the data point at 2 m should be ignored because the beam is not parallel at short distances). The upper curve, simulated in sand/air environment, contains the amplification provided by the scattered neutrons which have lost most of their energy in the environment.

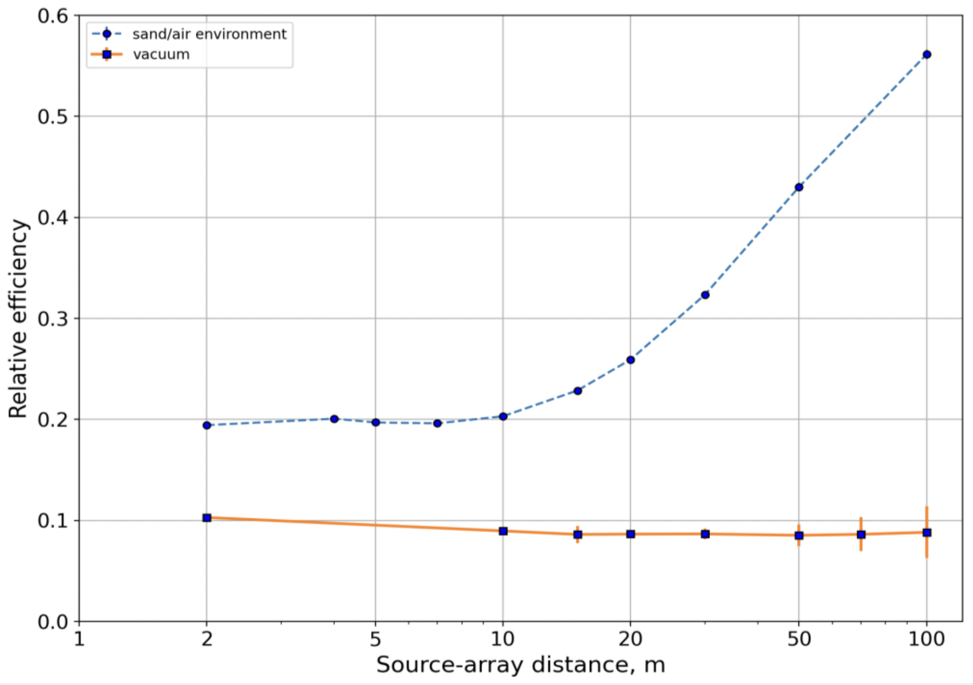


Table 2. Total counts in a detection system of 12 NaI arrays, each array consisting of 4 NaI detectors, from a Cf-252 source emitting 11,560,000 n/s (counting time 50 s, energy interval > 3000 keV, source-detector distance 20 m). The figures in brackets are normalized counts relative to a bare array in the vacuum. *Singlet = one energy deposition event within the array (no coincidence between voxels)

Environment	PVC	Singlets*	All counts	Fraction of singlets
Vacuum	None	2 126 (1.0)	4 746 (1.0)	0.45
Vacuum	3 cm	5 573 (2.6)	11 929 (2.5)	0.47
Air/Ground	None	11 941 (5.6)	19 955 (4.2)	0.60
Air/Ground	3 cm	20 297 (9.5)	35 756 (7.5)	0.57

Sea water is a very different environment to neutrons than land. The Cf-252 neutrons produce high-energy photons directly in the interactions with chlorine atoms in the salinized water. However, hydrogen is a strong neutron attenuator reducing the neutron flux arriving to the array. These two phenomena are competitors for the counting efficiency. Appendix 2 shows the efficiency comparison at sea and on land. Up to 20 m the efficiencies are very near each other but then the land environment provides a larger signal to the array. The efficiency ratio is 1.7 at the distance of 100 m.

The count rate of the array is a complex function of the environment. “Salt amplification” of the sea water was studied by comparing Cf-252 neutron count rates in the sea and lake environments. In the sea environment the count rate (> 3000 keV) was about 10 % higher at distances between 10 – 100 m (Figure B in Appendix 3), and the ratio seems not to be constant as a function of distance. However, the hydrogen peak (2224 keV) is attenuated in sea water environment as compared with lake water. The chlorine atoms in the sea water remove neutrons thus reducing 2224 keV photons of hydrogen directly and later indirectly because of reduced neutron flux hitting the PVC cover in the array (Figure C in Appendix 3).

A more striking impact of the environment is shown in Appendix 4. In a concrete tunnel the count rate is increased by a factor of ten at a source-array distance of 10 m. The signal amplification is much less at shorter and larger distances.

7.1.3 Detection efficiency as a function of distance

The capability of the NaI array to detect neutrons is shown Figure 4. The shape of the efficiency curve has two components. At short distances (< 12 m) the response follows the $1/r^2$ law but at longer distances the slope is essentially smaller (1.5).

Figures 4 and 5 give a benchmark comparison of the performance of the NaI array. The array is almost ten times more sensitive than a commercial portable SPIR-Pack detection system for gamma and neutron radiation [Toivonen 2023]. The sensitivity is of the same order of magnitude as in helium tubes and plastic detectors (Figure 5). For portal monitors, the ANSI standard sets the sensitivity requirement to 2.5 cps/ng at 2 m for Cf-252. This figure is 1.4 cps/ng for the NaI array (5 + 1 cm PVC). Obviously, if the array would be used for portal monitoring the 4 L detectors should be replaced with 8 L NaI detectors, or alternatively, a detection unit would consist of two arrays with 4 L detectors.

The minimum data acquisition time as a function of the source-array distance is given in Table 4 showing that a warhead emitting 200,000 n/s can be detected in 12 min or 1.9 hours at distances of 50 and 100 m, respectively.

Appendix 5 shows an efficiency comparison between all energy deposition events and singlets. Efficiency based on all events, including coincidences, at high energies (> 3000 keV) is about twice larger as the efficiency based on singlets. This phenomenon is also seen in Table 2. Both ways of analysis are useful; the detection sensitivity depends on the local background count rate.

The neutron counting efficiency is doubled by increasing the thickness of PVC from 3 cm to 5 cm and replacing the central Pb with 1 cm PVC Figure 4). The detection times would then also be improved by a factor 2, assuming that also the background count rate is doubled (Equation (5)).

Figure 4. Efficiency of the NaI array with PVC cover for detection of a Cf-252 source as a function of the source-detector distance. The NaI data, including coincidences, were fitted in two parts with a power function, which gives different slopes at short and long source-detector distances. For comparison, one measured result was added, a single 4 L NaI detector (Holm 2013) and one simulation result referring to an array having 5 cm PVC in the centre. The lowest curve refers to a commercial backpack detector (AmBe data from literature, (Toivonen 2023).

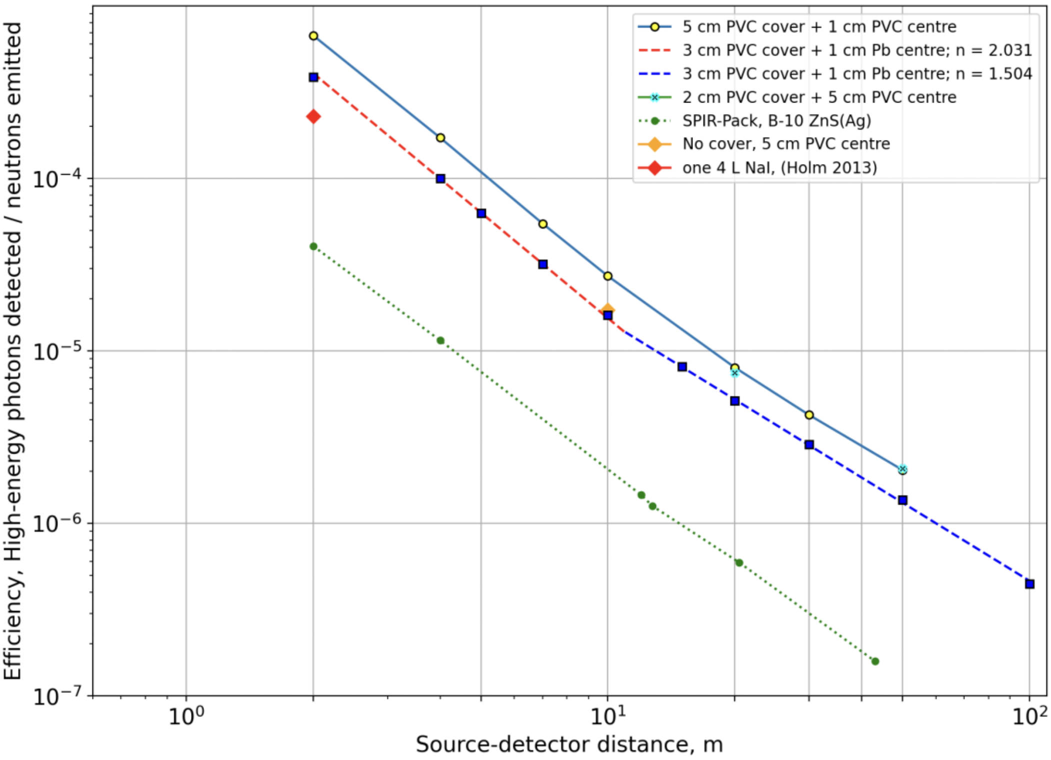


Figure 5. Sensitivity comparison (cps/nv @2m): NaI array performance versus commercial state-of-the-art neutron counters based on He-3 or plastic with Li/ZnS(Ag).[1]

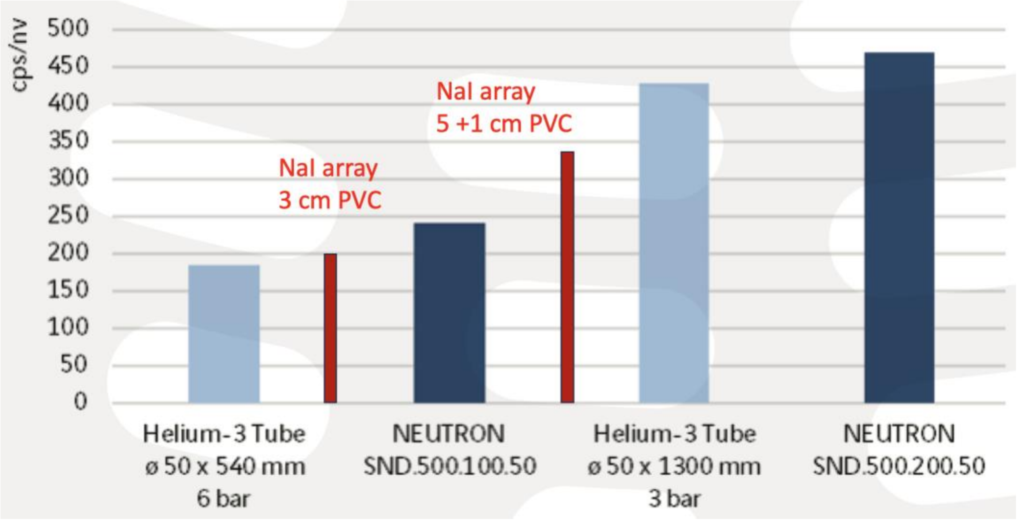


Table 3. Minimum counting time of the NaI arrays with 3 cm PVC cover to detect a Cf-252 source (all counts, including coincidences).

Distance [m]	Source = 200,000 n/s Counting time [s]	Source = 1,000,000 n/s Counting time [s]
10	5.2	0.21
30	165	6.6
50	720	28.8
100	6 765	270

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[1] Nuviatech Instrument. https://www.nuviatech-instruments.com/wp-content/uploads/sites/3/2017/12/NVG-375016-Fichesx4-NEUTRON-Aout2019-V4-2-1.pdf

7.2 Directional Sensitivity

The 2x2 NaI array provides source direction not only for photons but also for neutrons. The directional estimate can be obtained from the scattered particles. Table 4 shows the azimuth estimation results for the Cf-252 source as a function of source-array distance.

Table 4. Estimated azimuth of Cs-252 source by the NaI array. The analysis is based on singlets in the array. The true azimuth was 0 degree; source term 11,560,000 n/s.

Distance [m]	Estimated azimuth 10 - 3000 keV [deg]	Estimated azimuth 3000 - 10000 keV [deg]	Counting time [s]
5	1.6 ± 1.6	8.2 ± 3.7	5
10	0.7 ± 0.8	0.9 ± 2.8	50
20	2.1 ± 1.8	14.9 ± 5.8	50
50	0.3 ± 5.6	37.8 ± 14.4	50
100	3.9 ± 7.0	58.9 ± 15.1	250

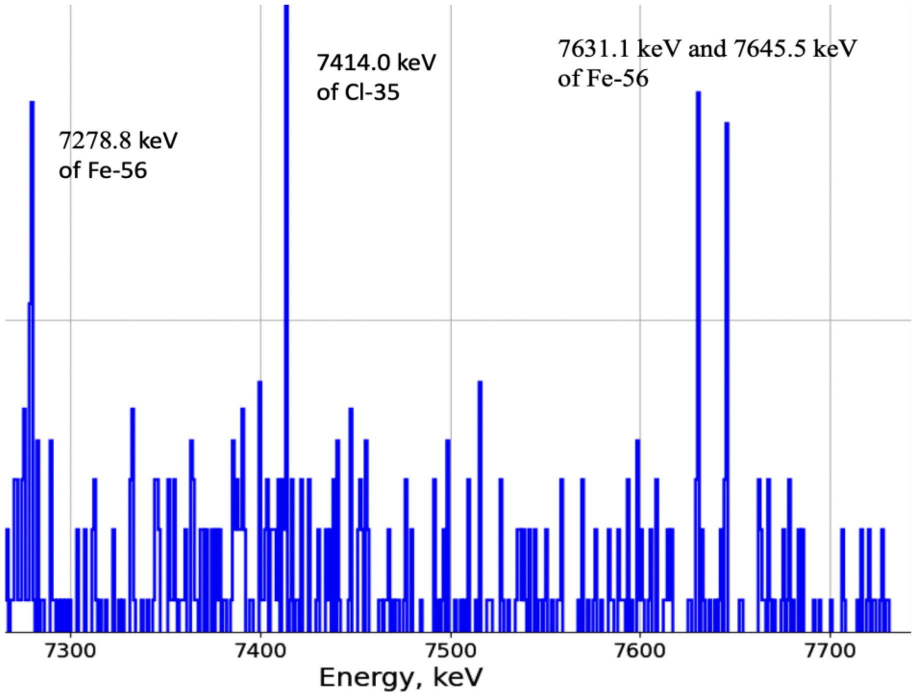
The energy region 10 – 3000 keV is better for the source localization than the higher energy region. A source emitting 1.156E7 n/s can be located with a precision of ± 7.0 degrees in 250 s which means that it is also detected. A nominal warhead (0.2E6 n/s) would then be located and detected in 14450 s (250 s multiplied with the source term ratio). However, the directional estimation was performed without any background contribution which has an impairing effect on the statistics.

8 Discussion

We have designed a multipurpose NaI array for photon and neutron detection, including directional sensitivity. The array has four large NaI detectors with total volume of 16 litres. The detection system, including PVC converters, is heavy (100 kg). However, the system is scalable, and a smaller backpack version is a straightforward application. A prototype, based on 1.5”x1.5” cylindrical NaI detectors, has already been built (Ihantola 2023, Toivonen 2024).

For optimized photon and neutron detection capability, one single large NaI detector would be an excellent instrument. Namely, the larger the detector, the more full-energy deposition events occur. However, such a system has severe operational issues. For example, a 16 L NaI scintillator would have a weight more than 50 kg and its operation would be difficult (mobility, temperature stabilization) and it does not have directional source localization capability. An array of smaller detectors, when operated by list-mode data acquisition, provides the same detection sensitivity. The coincidence events in the different detectors can be summed to one event. Then the array works as if it would be one integrated scintillator. The elements of the array are standard industrial components, and the complete directional array can be assembled from parts that are not heavier than 20 kg. In fact, an operational instrument should be modular which can be rapidly built from separate parts, including scintillators and embedded photomultipliers and auxiliary parts, such as Pb shields and PVC moderators.

Figure 6. Chlorine and iron peaks simulated from a Cf-252 source by the NaI array when the array is installed near a large iron slab.



When the NaI array is installed on an environment containing heavy elements, iron in particular, the neutron count rate is further enhanced. We made simulation experiments putting a large amount of iron in the vicinity of the NaI array, and consequently, the count rate was doubled (Figure 6). This observation can be interpreted from two perspectives. The count rate from a neutron source is increased but the background count rate will go up, too. The latter phenomenon is known as the “ship effect” (cosmic neutrons and other particles induce high-energy photons in heavy materials). The presence of iron is obviously very important in military applications. A more detailed analysis requires further studies.

The NaI array has neutron detection sensitivity comparable to commercial state-of-the-art neutron counters. It has two additional advantages: the array is a very sensitive gamma spectrometer, and it has directional sensitivity for gamma and neutron radiation. Its main application is not being a portal monitor but a search instrument in field operations or covering a large area for the detection of photon and neutron sources in or near a critical infrastructure of a state.

There are scientific and technical detection possibilities which could further improve neutron detection capability, including directional sensitivity. The individual elements of the array could be optimized for neutron interactions. Instead of NaI scintillators, boron-lined tubes (or helium tubes, if available) could be chosen for the array elements. Plastic detectors are ideal array materials. They contain hydrogen and are therefore excellent moderators. Special materials, containing isotopes with large neutron cross-sections such as Li-6, B-10, Cd-157, must then be included in the design to induce thermal neutron reactions within the array.

The present study shows that it is possible to build a large NaI detection array which can detect a nuclear warhead from a distance of 50 m (12 min), and in principle an operational system can be built for improved response having a shorter detection time at much longer distances (> 100 m).

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Appendix 1: Black Sea Experiment

The Black Sea experiment refers to Soviet and American efforts to detect a nuclear warhead from a distance of about 100 m. The following brief review is based on the papers of Fetter and Hippel 1990, Belyaev 1990 and Cochran 2011.

The experiment was conducted under the auspices of the Soviet Academy of Sciences and the Natural Resources Defence Council (NRDC) in 1989. *Slava*, a flag ship of the Soviet Black Sea Fleet, was anchored offshore at Yalta. For the joint experiments, a cruise-missile launcher was installed unshielded above the deck of *Slava*.

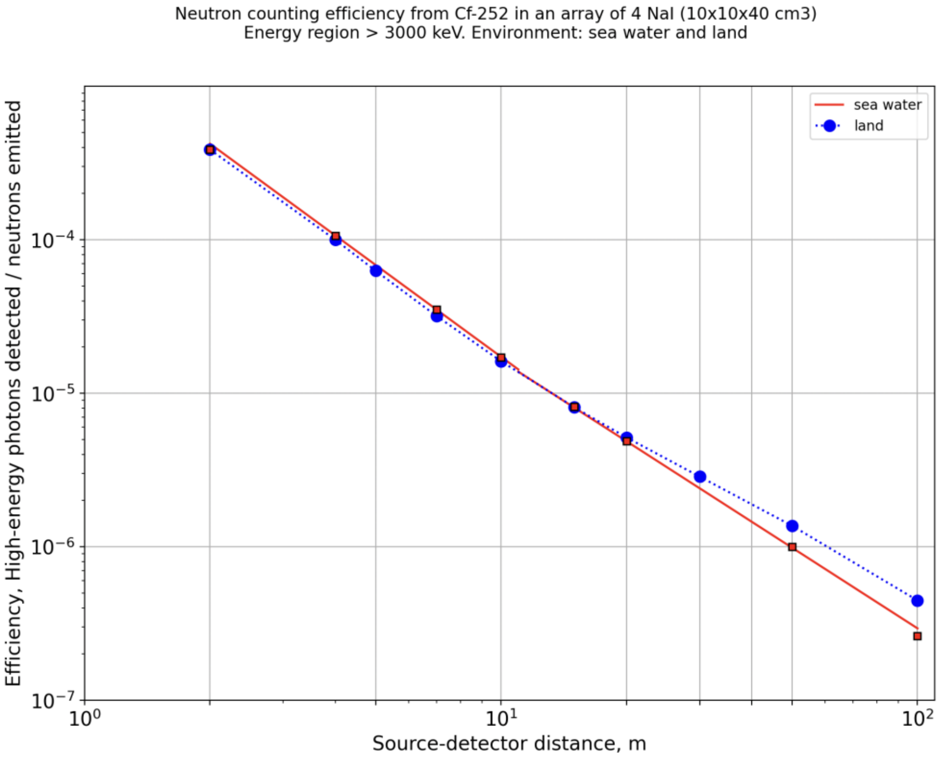
A high-resolution gamma spectrometer was used by NRDC. The measurements were conducted near the warhead (70 cm) in 10 min data acquisition. The gamma spectra, when the background had been subtracted, revealed the presence of U-235 (186 keV), Pu-239 (375 and 414 keV) and Am-241 (722 keV). In addition, the detection of Tl-208 at 2614 keV showed that the detected uranium is recycled material; Tl-208 is a decay product of U-232 which is an artificial isotope formed in the neutron flux of a reactor.

The weak U-238 peak at 1001 keV indicated that the uranium in the warhead was highly enriched. Another conclusion from the gamma spectrometric studies was that there was no heavy-metal shielding around the warhead. These structures would shield the low-energy photons from U-235 and Pu-239, and then no peak could be detected in the measurements.

The Russians made experiments at long source-detector distances using several different gamma spectrometers. The largest spectrometer was an array having a cross-sectional area of 0.25 m^2 installed on a ship passing along the side of the cruiser. None of the gamma spectrometric measurements were successful. However, the Russians claimed that in earlier experiments they had been able to detect the warhead at a distance of 13 m. The Russian neutron measurements revealed the presence of the nuclear material. A set of large He-3 proportional neutron counters was used, having the total cross-sectional area of 2.5 m^2. There were two such systems mounted in helicopters which were either hoovering above *Slava* or flew past the ship. These helicopter systems detected the plutonium at about 70 m, and the Russians said that the detection capability could reach 100 – 150 meters.

Appendix 2: Efficiency Comparison of NaI Array at Sea and on Land

Figure A. Efficiency at sea and on land of the NaI array with 3 cm PVC cover for detection of a Cf-252 source. All energy deposition events are considered, including coincidences.



Appendix 3: Comparison of neutron counts in seawater and lake water environments

Figure B. Ratio of all counts in NaI array one meter above the sea water level and lake water level. Energy region > 3000 keV, 2-sigma uncertainty.

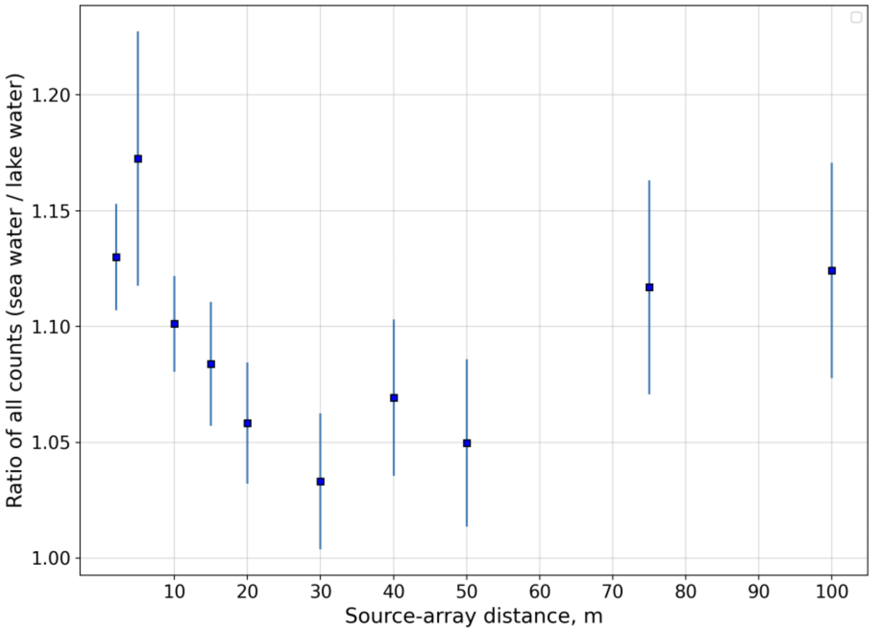
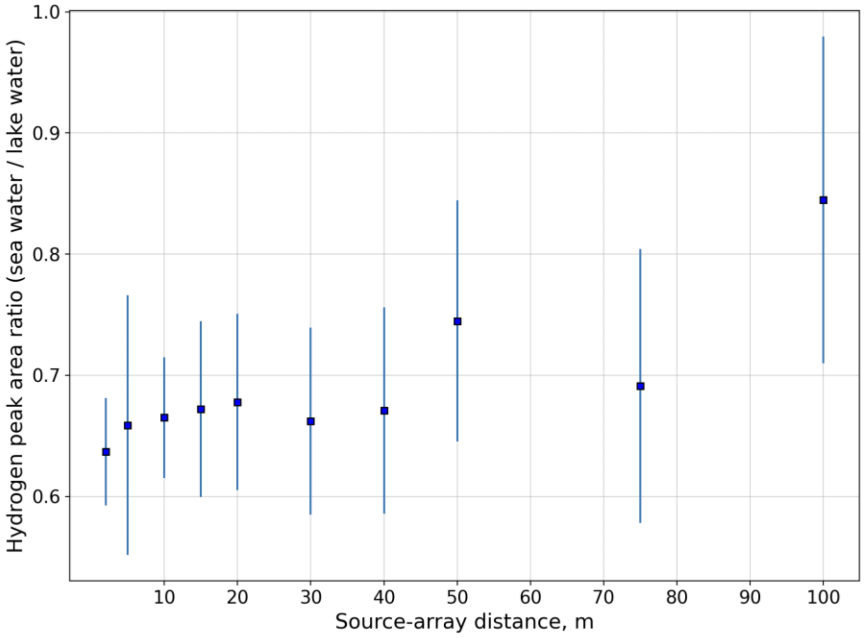
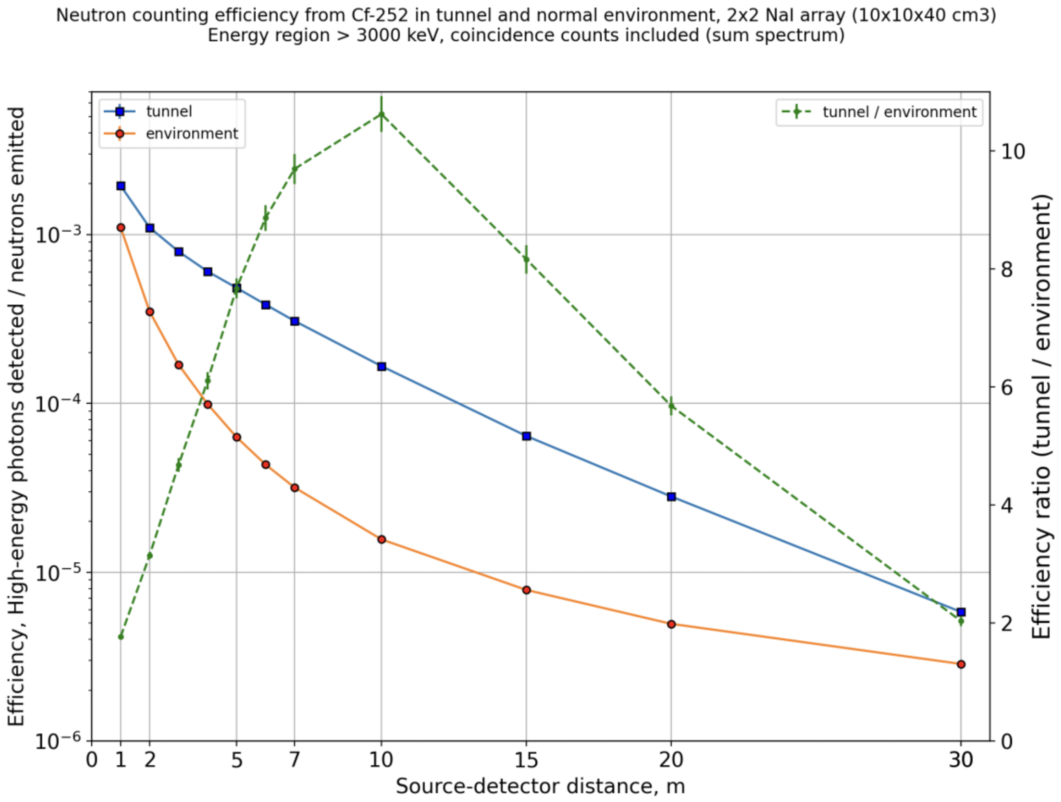


Figure C. Thermal neutron induced hydrogen peak area ratio (2224 keV) in NaI array one meter above the sea water level and lake water level, 2-sigma uncertainty.



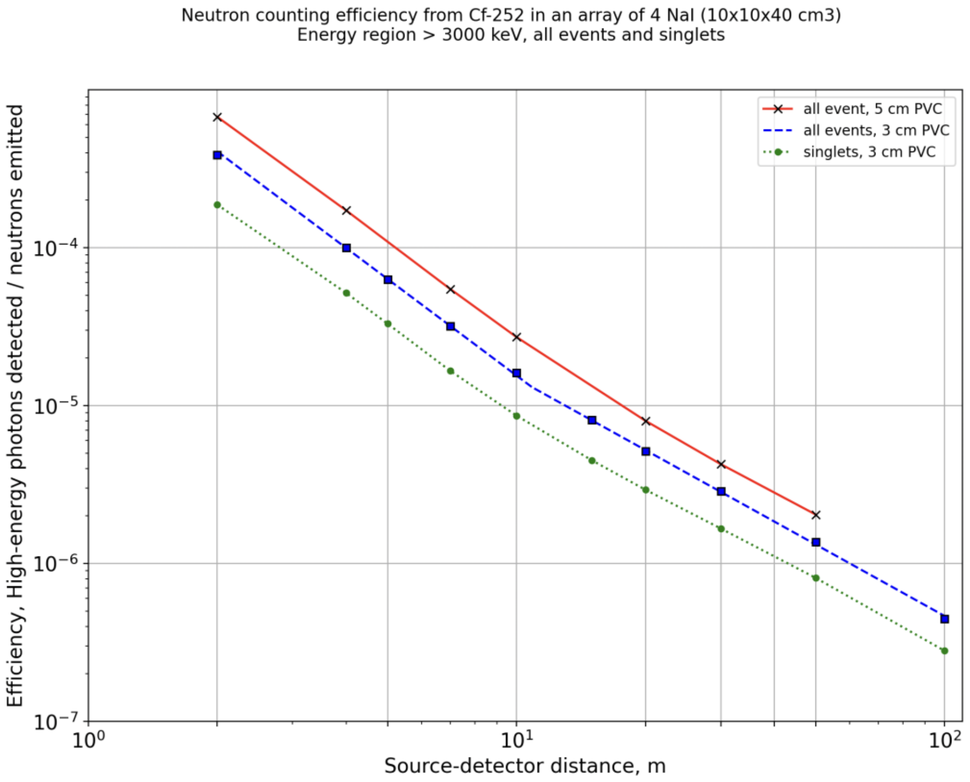
Appendix 4: Increased Neutron Signal by Concrete

Figure D. Comparison of Cf-252 neutron counting efficiencies of the NaI array with 3 cm PVC cover: long concrete tunnel having sand floor versus environment. The width and height of the tunnel were 4 m, and the concrete thickness was 1 m.



Appendix 5: Efficiency Comparison of NaI Array - All Energy Deposition Events versus Singlets

Figure E. Efficiency of the NaI array for detection of a Cf-252 source in land environment. The array was covered with PVC and there was 1 cm Pb in the centre.



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