GeCo - A gamma-ray spectroscopy system for evaluation of coincidence methods in radionuclide monitoring

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Abstract

Radionuclide monitoring is a proven means of non-intrusive verification of the nuclear test ban treaty. In addition to that, the potential use of radionuclide monitoring spans beyond the detection of nuclear test explosions, since radionuclides can also be released and detected from operations of nuclear fuel cycle facilities, such as the reactor operation and nuclear reprocessing of plutonium production.

In this work, we consider the use of coincidence and anticoincidence techniques as a means to increase the sensitivity in radionuclide monitoring, in terms of improved minimum detectable amount for radionuclides of interest to filter stations used in radionuclide monitoring. In particular, a multi-detector setup is currently being prepared for the evaluation of the technique, and to provide validation data for a coincidence detector simulation codes.

In this presentation, we will describe the multi-detector setup assembled for enabling the evaluation of various types of spectrometry, including 1) gamma-gamma coincidence (from dual detectors and up to five High Purity Germanium (HPGe) detectors), 2) anticoincidence using BGO active shield with single HPGe detector, as well as use of multiple detectors in add-back mode, i.e. simply using the combined detector volume for increased efficiency of single gamma rays. We will present the results of measurements of a calibration sample, and provide a discussion on the advantages and disadvantages of the tested techniques in the context of radionuclide monitoring.

1. Introduction

Any use of nuclear explosive devices is prohibited by the Comprehensive Nuclear-Test-Ban Treaty (CTBT) that has been signed by 187 states. Although the treaty is not yet in effect, the International Monitoring System (IMS) is being prepared for verification of the treaty through the use of a global network of remote monitoring stations. A key in this network is the 80 radioactive particle monitoring stations, where airborne particles are collected on air filters that are subsequently examined with gamma-ray spectrometry [1]. In parallel, radionuclide monitoring is performed by several national networks of monitoring stations, which have objectives within e.g. nuclear safety.

In nuclear explosions, a vast number of fission and activation products are created. Several hundred of these are gamma emitters with sufficient yield and half-life to allow for detection after atmospheric transport across considerable distances [2]. However, any radionuclide monitoring network is by economic necessity sparse, leading to a risk that radioactive releases from a treaty violation go undetected due to dilution and decay during atmospheric transport. By improving the ability to detect nuclides in a radioactive release plume, we aim to improve the ability to localize the source of the release, to determine the release time, and to understand the nature of the source, whether a treaty violation or not.

Coincidence methods in gamma-ray spectrometry have previously been employed in radionuclide monitoring to improve the sensitivity to nuclides of interest [3,4]. In the last decade, dual detector systems [5], [6] and anti-Compton shielded [7] detectors have been employed for coincidence spectrometry. In this work, we describe an experimental setup recently assembled in Uppsala University, the recently assembled Germanium detectors in Coincidence (GeCo), to explore the use of multiple detection elements to enhance sensitivity. The setup will be used to build competence in coincidence spectrometry, to provide validation data for simulation models used for the design of the next-generation detector system, and to explore the benefits of the different coincidence modes and analysis techniques for the sensitivity to key nuclides in filter samples. In this paper, the detector system is described and data are analyzed according to the different coincidence modes demonstrated.

2. Coincidence spectrometry

The GeCo was built to explore the use of coincidence gamma-ray spectrometry to enhance the sensitivity to key nuclides in the verification of the CTBT. The GeCo setup may be used for coincidence or anticoincidence measurements, or (due to having a large number of detection elements) combinations of the two methods may also be employed.

2.1 Coincidence technique

When recording gamma-rays with a system of multiple detection elements, events can be discriminated before analysis based on whether there are coincident events in multiple detection

elements, or if not (i.e. anticoincidence). Coincidence measurements are powerful to selectively interrogate radionuclides that emit multiple gamma-rays in cascade, and thereby reduce the everpresent background from other nuclides. By reducing the detector's sensitivity to background, a small peak of interest is more likely to significantly exceed the noise, which leads to an improved detection limit. Any radionuclides with a decay followed by multiple gamma transitions from the same nucleus, such as Cs-134 (see decay scheme in Figure 1), may be measured with the use of coincidence spectrometry. The gamma-ray emissions of such cascades happen nearly simultaneously and have typical separations in the picosecond timescale.

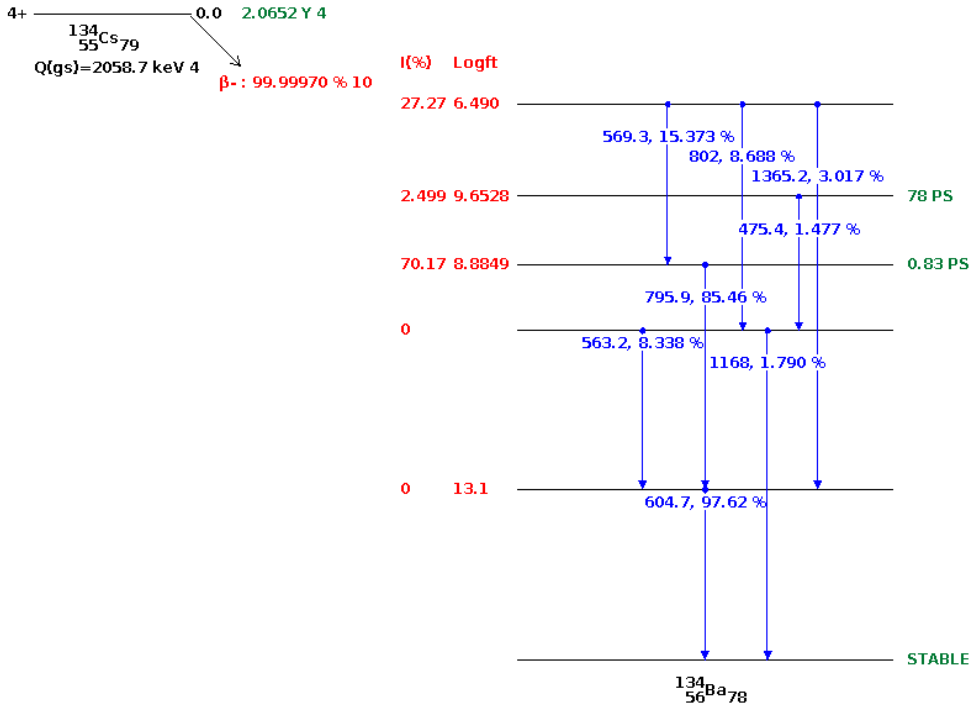

Figure 1. The decay scheme of Cs-134. Upon the beta decay to Ba-134, deexcitation of the daughter nucleus involves multiple gamma emissions in less than a nanosecond. Image from NUDAT3 [8].

When analyzing events that are in coincidence, further selection based on energy is an option, which can be used to optimize the sensitivity to a particular nuclide of interest. In this work we will demonstrate spectra analyzed in two manners; 1) with total spectrum coincidence, meaning that all events that have a coincident event in another detector are counted, and 2) gated coincidence, where events are counted only if a coincident event of neighbor detector element falls within a narrow peak energy region of a particular gamma transition of a selected nuclide. In the result section below we compare the analysis of Cs-134 in these two ways, and with the use of conventional single gamma-ray spectrometry. In this demonstration, we obtain a gated coincidence spectrum by using a gate on the 605 keV peak in one of the neighbor detectors. As seen in Figure 1, the 605 keV gamma ray is in coincidence with multiple other emissions, the strongest intensity of which is the 796 keV gamma ray.

2.1 Anticoincidence technique

The use of anticoincidence is also possible with GeCo. It is planned for with the use of a BGO Compton shield surrounding one of the HPGe detectors. This BGO shield can be employed to discriminate gamma-rays that scatter in the HPGe detector and escape after incomplete energy deposition, generating a continuum background in the target HPGe. Such escaping gamma-rays are shielded and detected by the active BGO, and the corresponding event in the HPGe is discriminated. This method is often employed to increase sensitivity to nuclides with lower gamma-ray energy than the dominant background nuclides, by reducing their corresponding continuum background that otherwise interferes with the peak of interest [9]. This method is equally applicable also to radionuclides with a simple decay scheme with a single gamma-ray. Note that in this work, we don't present results from anticoincidence measurements, but such tests are planned for future work.

3. The GeCo setup

3.1 Sample holder

The GeCo measures the gamma-ray activity from a cylindrically shaped (of type P60) can containing the air filter sample. The air filter processing and can are identical to the procedures used in the radionuclide monitoring stations of the national Swedish network operated by FOI. The sample is placed in a custom-made 3D-printed sample holder, see photos in Figure 2. The sample holder is located in the center of GeCo's 5 HPGe detectors as illustrated by Figure 3.

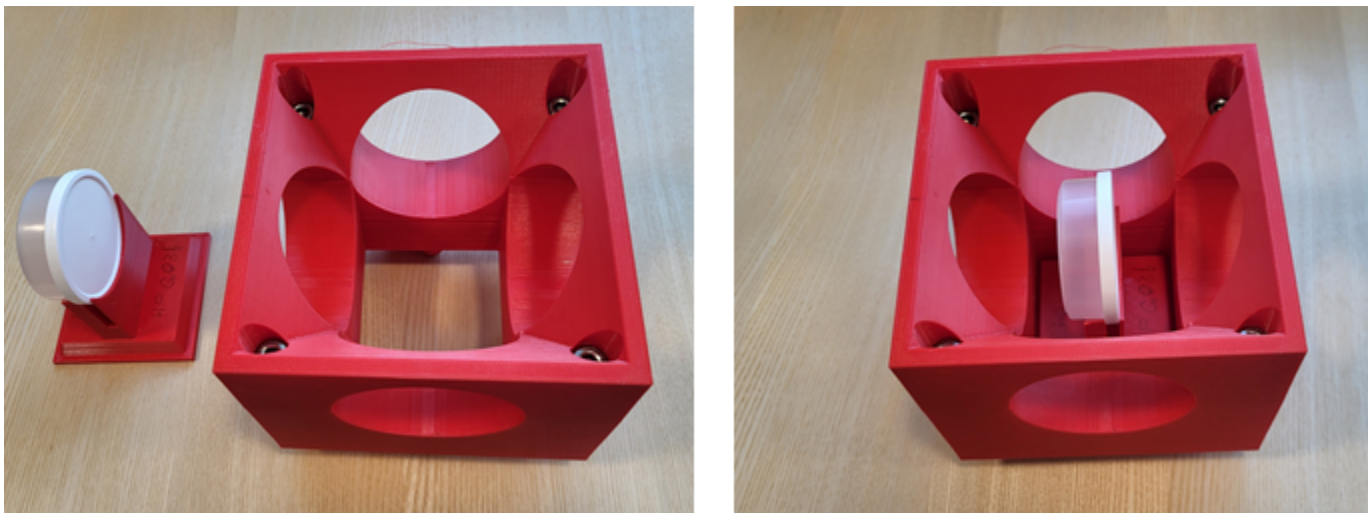

Figure 2. The 3D-printed sample holder (red), with an empty can for demonstration. For a simple change of the sample, once the detector setup is fully assembled, a module for fixation of the sample can be extruded from beneath the sample holder. Four rotating locking hooks hold the extractable module in place once inserted. In addition, the sample holder serves to secure the four horizontal HPGe detectors in a reproducible position. Left: Sample extracted. Right: Sample inserted and locked in place.

3.2 Detectors

GeCo uses five n-type HPGe detectors, one of which employs active shielding of six BGO scintillators. Of the five HPGe detectors, two are facing the end caps of the cylindrical sample (detectors 3 and 4). These two detectors cover the largest solid angle of the respective detectors and are thus exposed to the highest count rates. Three other detectors are facing the cylindrical mantle area of the sample; two of them are placed horizontally (detectors 1 and 2) and one vertically (detector 5) above the sample. All horizontally oriented HPGe detectors are fixed in position by the custom-made sample holder. These four detectors are of the same model, Canberra GR1020. The vertically oriented HPGe detector is a Canberra EGC-55, which is held in place above the sample by a custom-made framework with four legs, each positioned outside one of the corners of the sample holder in the assembled GeCo.

The vertically placed HPGe detector has six BGO detectors placed around it, which can be used for anti-coincidence shielding. The BGO detector is supported by the same framework as the vertical HPGe detector. The GeCo layout is shown in Figure 3, with CAD-generated images and a photo of the assembled setup.

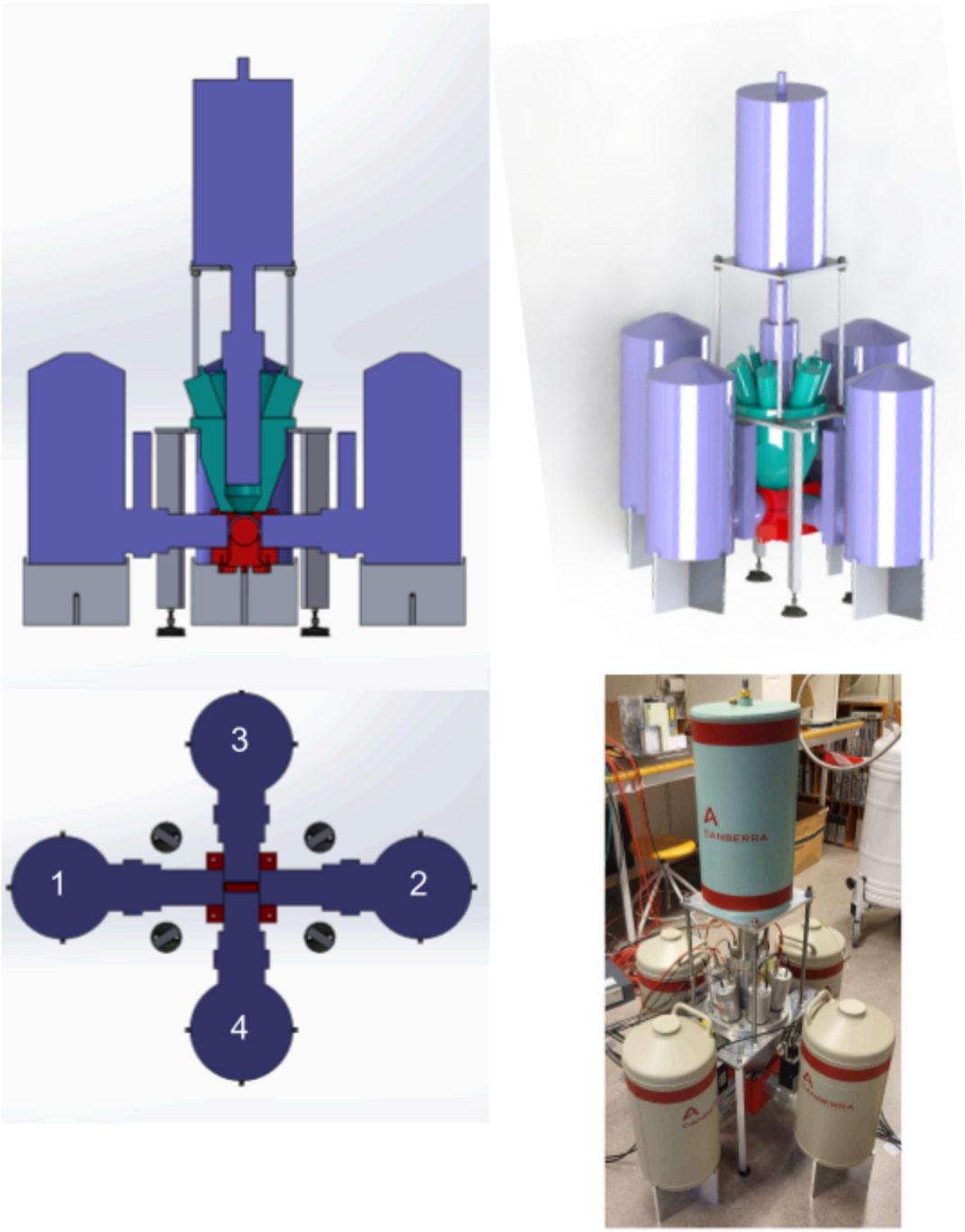

Figure 3. The germanium detectors with cooling (blue). The sample holder (red). The BGO detectors (green). Upper left: Vertical 2D slice of CAD model. Upper right; 3D rendering of CAD model. Lower left: Horizontal 2D slice of CAD model. Lower right: A photograph of the mounted detectors.

3.2 Data collection and format

The signals from all detectors are collected using an 8-channel Caen V1782 DAQ system. To enable the use of in total of 11 detection elements with an 8-channel DAQ, the BGO detectors are summed using a fan-in NIM module and read out in the remaining channels, after assigning individual channels to each HPGe detector.

The raw data of the CAEN Compass DAQ system is stored in ROOT Trees in ROOT files (but can optionally be stored as CSV files). Each event is stored as a leaf with the properties board, channel, energy, timestamp, and an error flag. The energy is the uncalibrated energy (in channels), and the timestamp contains the time of the recorded event in picoseconds.

This data format gives a great versatility to explore (in the offline analysis) various coincidence modes, allowing for the comparison of different analysis routines, including coincidence, anticoincidence, and single-gamma spectrometry, all with the same data collection.

4. Experiment

3.1 Calibration sample

In this paper, we demonstrate results from data recorded with a calibration sample, consisting of an air filter with artificially added radionuclides, as detailed in Table 1. The sample is contained in a P60 container. It can be noted that many of the radionuclides are of very weak activity, due to aging of the calibration sample. However, the activity of e.g. Cs-134 is still detectable and presents coincident emissions of gamma rays.

Table1. Table of nuclide activities in calibration sample, at the time of the measurement.

Nuclide	**Activity [Bq]**
Am-241	1189
Cd-109	27
Co-57	0.011
Cs-134	34
Cs-137	249
Mn-54	0.110
Zn-65	0.024

5. Results

The data is sorted according to their timestamps. Two consecutive events are considered coincident if the time difference of their timestamps is within 400 ns. This is much larger than the time separation between the gamma-ray emissions, shown in Figure 1, but the setting is governed mostly by the charge carrier collection in the HPGe, which has a spread in the order of 100 ns. The 2D coincidence spectrum of HPGe3 and HPGe4 is shown in Figure 4. Note that the coincident gamma energies of Cs-134 are prominent (horizontal and vertical lines) around 563, 569, 605, and 796 keV. The diagonal lines mostly correspond to the interaction of one single gamma ray in GeCo, where it Compton scatters in one detector and then deposits the rest of its energy in the other detector. In this manner, a low level of background is caused also by nuclides with a single gamma-ray emission, even in the highly selective coincidence analysis, such as the gamma rays of 1461 keV of the naturally occurring K-40.

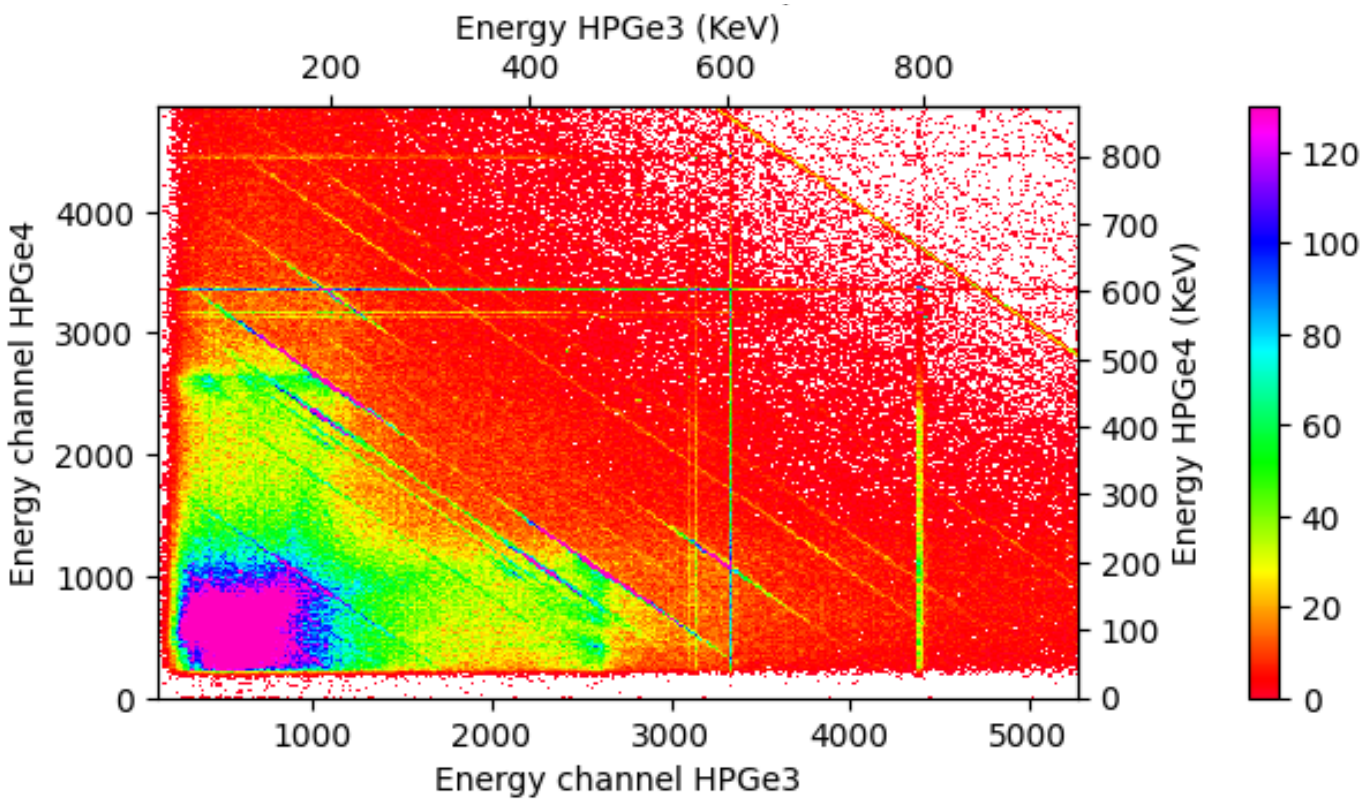

Figure 4. The 2D coincidence spectrum of HPGe3 and HPGe4 is shown in Figure 2. Note that the coincident gamma energies of Cs-134 are prominent (horizontal and vertical lines) around 563, 569, 605, and 796 keV. The diagonal lines correspond to the interaction of one gamma which Compton scatters in one detector and then deposits the rest of its energy in the other detector.

One of the benefits of the coincidence measurement technique is that it reduces the background of the measurement, as is demonstrated in Figure 5. The figure compares a spectrum taken with HPGe3 without any coincidence limitations, with a coincidence spectrum where we demand a coincidence event in HPGe4 (of any energy), and in addition a coincidence spectrum with

energy gating on the 605 keV gamma in the HPGe4 detector. In Table 2, the signal-to-background is calculated for the 796-keV full energy peak for the three compared options.

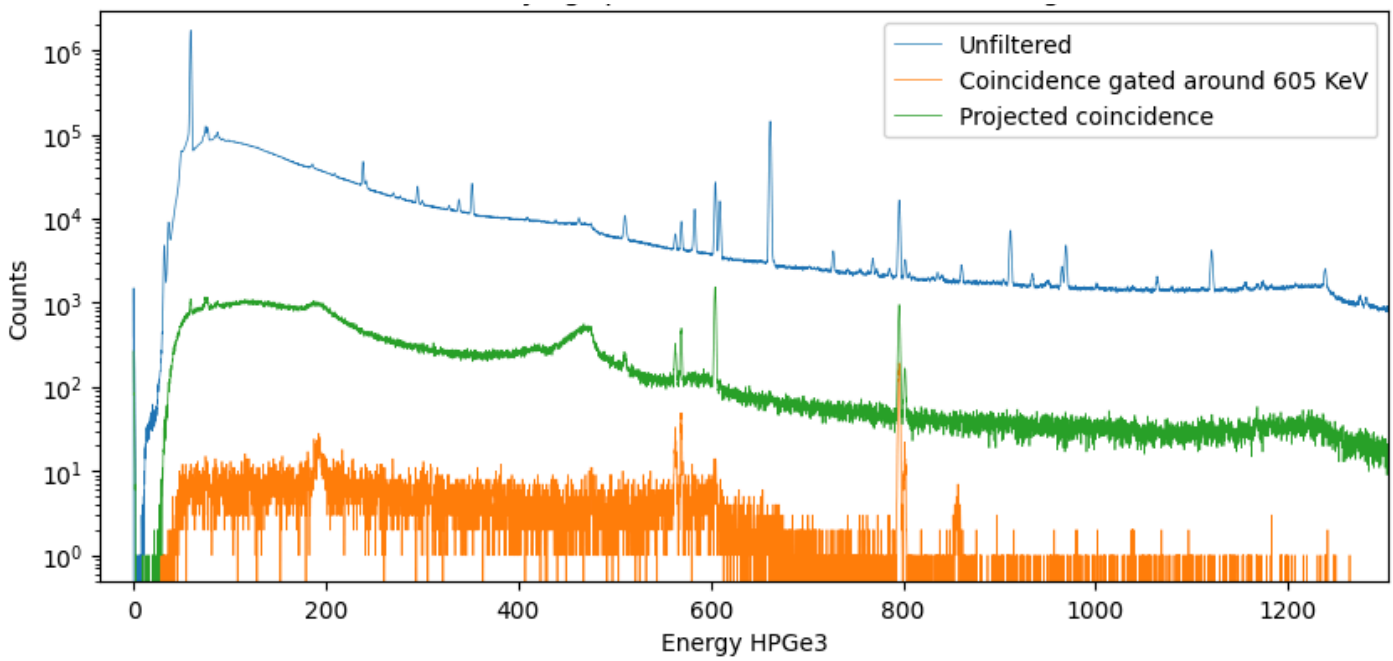

Figure 5. Three PHS spectra of HPGe3. The upper blue line corresponds to the traditional PHS of the detector, without any coincidence limitations. The middle green line corresponds to the coincidence spectrum of HPGe3 where we demand a coincident event in HPGe4 (of any energy). The lower orange curve corresponds to the coincidence spectrum of HPGe3 and HPGe4 where we demand a 605 keV gamma in the HPGe4 detector.

Table 2. Comparison of the signal-to-background of the 796 photo peak of the spectra in Figure 3. In Column 1, the analysis method is given. In Column 3, the number of counts is given, after the background has been subtracted. In Column 4, the estimated background is given. In column 5, the uncertainty of the number of counts is calculated. In column 6, the Signal-to-Background is calculated.

Analysis method	**Energy**	**Counts**	**Background**	**σ_{Counts}**	**S/B**
No Coincidence (blue)	796	178 965	81 345	584	2.2
Coincidence (green)	796	10 626	1 773	119	9.06
Gated Coincidence (orange)	796	2 152	11	46.6	195

6. Discussion and Outlook

A coincidence gamma-ray spectrometry system, the GeCo, has been assembled at Uppsala University and the first experimental tests have been performed. The detector was demonstrated to work in coincidence mode with the use of up to five HPGe detectors, providing ten pairs of detectors for coincidence events of two-fold multiplicity. One of the HPGe detectors has an active shield of BGO scintillators, providing the additional possibility to evaluate anticoincidence detection to reduce background from Compton continuum events.

While the powerful reduction of background can be noted in Figure 5, the technique also reduces the peak efficiency. Therefore, a goal of future design and optimization of dedicated coincidence setups for radionuclide monitoring is to obtain a good compromise between low background and high efficiency. The results from GeCo will provide realistic training and validation data for modeling signal and background intensities in coincidence spectra.

The data collected in the first measurement campaign is now undergoing analysis to perform efficiency calibration for single and coincidence detection modes of key nuclides, and in addition, the background in respective peak regions will be assessed in respective modes. Efficiency and background count rates provided, the Minimum Detectability Activity (MDA) will be evaluated for key CTBT nuclides, and for the respective coincidence modes that GeCo enables, so the techniques can be compared.

Further work is planned on estimating the minimum detectable activity for comparison with conventional single gamma-ray spectrometry. Also, validation of a Geant4-based simulation framework will be performed. This simulation framework will form the basis for further development of radionuclide monitoring detectors using the coincidence technique. It is planned that based on the learning outcomes from the use of the GeCo detector, a next-generation detection system will be designed and optimized. Several multi-element detector types are currently under consideration for the next-generation setup; including segmented dual, well, and clover detectors. Evaluation of such setups for coincidence detection of airborne radionuclides relevant to CTBT verification will be performed by simulation studies, aiming to perform a selection based on performance, and subsequently a detailed detector design.

In the final stage of the project, a coincidence detector system dedicated to radionuclide monitoring will be assembled and tried in long-term operation. The aim is to form an experience basis to at the end of the project allow for informed judgment about the potential of coincidence detection systems to further the capability of radionuclide monitoring for treaty verification.

7. References

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