

HEALTH RESEARCH ABSTRACT SUBMISSIONS

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Title of Research *	: The Use of Gamma Spectroscopy to Conduct Rapid Urine Radionuclide Screens in the Event of a Radiological Emergency
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Introduction & Purpose *

In the event of a radiological emergency, laboratories will need to have the capabilities to determine radionuclide exposure concentrations rapidly to assist public health officials and clinical providers in providing effective treatment. At the moment, few state laboratories are able to analyze large numbers of biological samples within a relatively short period of time due to lack of resources and guidance. Most laboratories are not equipped with highly efficient instrumentation, have limited experience working with the sample matrix medium, and are encountering dwindling numbers of skilled personnel to conduct these types of analyses. During an event of this magnitude, there is a high likelihood that state and national public health laboratories will be overwhelmed with the demands of testing both biological and environmental samples. Rapid screening techniques will be crucial for quantifying radionuclides so treatment can be prioritized for those with detectable radiological exposures¹.

The DOE/NRC Interagency Working Group on Radiological Dispersal Devices has identified radioactive materials of greatest concern likely to be used in radioactive dispersal devices or "dirty bombs"². The Centers for Disease Control and Detection (CDC) have developed analytical methods for three gamma-emitting radioactive isotopes – Cobalt-57 (57Co), Cobalt-60 (60Co), and Cesium-137 (137Cs)³. We examined only 60Co and 137Cs. 60Co (T1/2 = 5.271 years) is a commonly used commercial source of gamma radiation, emitting 1.173 and 1.332 MeV⁴. 137Cs (T1/2 = 30 years) is an important fission product due to its high fission yield and long half-life emitting 0.6616 MeV⁴. 137Cs can be found in consumer products, industrial equipment, and to a lesser degree in medical devices, nuclear reactors, and hospital waste⁴. After exposure to cobalt and cesium via inhalation, ingestion or dermal contact, they're absorbed in the blood leaving the body slowly through urine^{3,4}.

Many state public health laboratories have access to High Purity Germanium (HpGe) gamma detectors ranging between 10 to 30 % efficiency. Very few laboratories have 70% efficient detectors. Shorter sample count times have been observed with greater efficiency ratings for detectors. Along with the efficiency of the detector, the density of the matrix and spatial orientation of the sample counting container to the detector would need to be considered as they can have an effect on the detection capability and instrument calibration.

The existing methods currently used to analyze radionuclides in urine, for instance, are limited due to low throughput, high volume requirements for samples, and lack of guidelines for detection limits of radionuclides². In response to this limited capacity of laboratories to handle the level of population monitoring necessary to measure radiologic internal

contamination, the CDC developed the Urine Radionuclide Screen method². This technique allows a laboratory to “screen” samples containing as little as 50 milliliters of urine taken at a “point-in-time” to simultaneously identify and quantify more than 10 high-priority radionuclides within 24 hours². But, CDC’s aim for laboratories is to detect as low as a dose of 1 ALI (Annual Limit of Intake)⁷. Sparse guidance is found in previous studies that will aid laboratories to translate units of radioactive activity, i.e. from curies to ALIs.

Experimental Design *

To meet the guidelines of the CDC’s Urine Radionuclide Screen method⁴, a place-holding apparatus was developed to hold multiple urine containers. Instead of placing the sample containers directly on top of the gamma detector, they stand upright around it so they’re positioned closer to the detector. A maximum of six 50-mL vials can be held by the apparatus. Laboratory testing of environmental samples have usually been processed at a rate of one vial per run. This apparatus permits the analysis of several samples in a given run.

A HPGe gamma detector Model GC1819 (Canberra Industries, Inc., Meriden, CT, <http://www.canberra>) with a relative efficiency of 18% was used for the analysis. The geometry is a closed-end coaxial with a diameter of 5.5 mm, length of 40 mm, active column of 74.4 mL, and a distance of 5 mm from the window. The energy resolution specifications are 1.9 keV Full Width at Half Maximum (FWHM) at a gamma energy level of 1.33 MeV and 1.0 keV FWHM at 122 keV. A room temperature within 73°F ±3 was maintained. All analyses were performed using GammaVision-32® Version 6 software.

A gamma spectroscopy library file needed to be first constructed incorporating certain isotopes of greatest concern assembled by the CDC³ and their energy levels. Six 50 mL blank sample vials (total volume of 0.3L) were then placed in the apparatus around the detector surrounded by a lead shield. The analysis of the blank tubes was used to develop a background subtraction file of natural peaks that are observed for correction of future sample spectrums. Six 50 mL vials were prepared for calibration samples by spiking a known 10-isotope mixture into de-ionized distilled water. These 10 isotopes in the mixture were ²⁴¹Am, ¹⁰⁹Cd, ⁵⁷Co, ¹³⁹Ce, ¹¹³Sn, ¹³⁴Cs, ¹³⁷Cs, ⁵⁴Mn, ⁸⁸Y, and ⁶⁵Zn obtained from Eckert & Ziegler Analytics (Atlanta, GA, <http://www.analytisc.com>). The isotopes’ energy, abundance, known activity, and reference activity dates were retrieved from the Eckert & Ziegler Analytics’ calibration certificates. The activity peaks (minimum of 10,000 counts) of these isotopes were used to calibrate the energy and efficiency of the detector for this new protocol.

In addition, 6 blank samples of 50 mL de-ionized distilled water were prepared for the purpose of determining detection limits. The set of six blank samples were placed in the apparatus and counted at different count times ranging from 30 minutes to 17 hours. The volume used to calculate the Minimum Detectable Activity (MDA) was based only on 50 mL to represent one positive tube from the 6 tubes to provide the lowest possible intervention limit. Another set of analyses was conducted consisting of one calibration spiked sample and 5 blanks counted at various times. This would represent a positive urine sample for radiological activity in the entire composite of 6 tubes for the purpose of comparing minimum detection capabilities and intervention cutoff values of a 6 tube screening composite (Figure 1). If activity levels exceed our cutoff points, individual samples would be analyzed separately. There were also three sets of 50-mL vials spiked with low, medium, and high activity levels for ⁶⁰Co and ¹³⁷Cs obtained from Eckert and Ziegler Analytics. The low, medium and high levels were based on values of effective doses acquired from CDC’s Urine Radionuclide Screen method⁴ and detection limits from our detector. Each sample was analyzed and compared to test the detection limits and accuracy of the calibration of the detector.

According to the CDC’s Urine Gamma Emitting Isotopes HPGe Laboratory Protocol⁴, the minimum intervention levels from inhalation for urinary excretion concentrations for a sample volume of 50 mL obtained on a given day corresponding to an Effective Dose of 0.1 Sievert (Sv) were determined for ⁶⁰Co and ¹³⁷Cs. The effective dose refers to doses occurring from external and internal exposures⁸. Most radionuclides are particulates grouped as Type F, Type M, or Type S absorption

type8. Type F has fast dissolution and a high level of absorption to blood whereas Type S has slow dissolution and low level of absorption to blood8. Type M has intermediate rates of dissolution and an intermediate level of absorption to blood8. The most conservative approach was to use the CDC values for an effective dose of 0.1 Sv with a Type S absorption type4 as the concentration we would set as the intervention cutoff level. Based on our calculations, it was 0.0339 microcuries/L For 60Co and 0.00119 microcuries/L for 137Cs.

Results *

Minimum detection limits (MDL) were determined for 14 radioisotopes (Table 1). Six samples were filled with de-ionized water and considered to be blanks and counted for 30 minutes and as long as 17 hours.

Once our calibration, background correction file, and MDLs were established, preliminary studies were conducted. One calibration standard sample was added to the 5 blank sample batch and prepared as discussed previously. At a count time of 30 minutes, the agreement of the level of activity detected compared to the standard known concentration was greater than 10% for some of the isotopes (Table 2). On the other hand, a count time of 60 minutes and 17 hours produced an agreement within 10% compared with the standard known activity for all the isotopes. A count time of 30 minutes was therefore eliminated as a possibility for a shorter screen count time.

The maximum capacity for urine gamma spectroscopy analyses is shown in Table 3. The current protocol count time to analyze a urine sample requires 17 hours with a volume of at least 200 mL. Using this new protocol, a maximum number of samples can be analyzed with a smaller volume of 50 mL. During a radiological emergency, the laboratory is in continuous operation. The maximum throughput is 28 urine samples at a count time of 6 hours. By decreasing the count time to 1 hour, the number of samples that can be analyzed is close to 70. Implementing the Rapid Urine Radionuclide Screen, 1008 samples can be examined. This is 6 times the number of samples that can be tested with a 1-hour count time.

Additional validation studies were performed with 137Cs and 60Co isotopes spiked at 3 different activity levels. A low activity sample was set below the minimum detectable amount for each isotope at a 1 hour count time. The middle level was designated as the urinary excretion concentration limit for an effective dose of 0.1 Sievert with a Type S absorption. The high level was the limit based on a urinary excretion concentration limit for an effective dose of 0.5 Sievert with a Type S absorption. For the low level, the samples were detected below the minimum detectable amount (Table 4). For the middle level, the accuracy at a 1-hour count time for 137Cs and 60Co was 124 and 105%, respectively. For the high activity level, the accuracy for the 6 hour count was improved to 127.6% for 137Cs and 99.5% for 60Co.

Conclusions *

For a 50 milliliter urine sample, the quantification of 10 radionuclides using a custom-made matrix was compared and detected at recovery rates within $\pm 10\%$ from the known spiked sample with a count time as low as 60 minutes. Those spiked samples of isotopes with 137Cs and 60Co prepared with lower activity indicate a shorter count time can be used but the accuracy at a count time of 60 minutes is not as strong versus a count time of 6 hours. These preliminary results demonstrate that rapid urine radionuclide screening and measurements can be performed with shorter count times and small sample volumes, but further studies of count times between 1 and 6 hours need to be conducted. Additional validation studies are needed to continue to optimize and evaluate the CDC's Rapid Urine Radionuclide Screening protocol.

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