EVALUATION OF LABORATORY DETECTION SYSTEMS FOR FISSION PRODUCT DETECTION

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ABSTRACT

To effectively monitor the Comprehensive Nuclear-Test-Ban Treaty (CTBT), an International Monitoring System (IMS) is being constructed which will encircle the Earth with explosion detection technology [1]. Radionuclide measurements will be carried out for aerosols and for radioactive xenon isotopes. The radionuclide portion of the IMS requires a group of laboratories to perform quality assurance measurements on the radionuclide stations and to perform confirmatory analyses of samples of interest. The goal of the re-analysis is greater confidence in the detection of fission products, which are potential proof of a CTBT violation. The selection of laboratory technology for this mission can critically affect the confidence level attainable in a fixed measurement time.

Short lived fission product spectra have been created to evaluate several instrumentation approaches for laboratory measurements of the Comprehensive Nuclear Test Ban Treaty. Modest germanium detectors of several types and in shields of varying background have been used to acquire daily spectra from a hypothetical CTBT detection scenario: A group of sources was designed to simulate a CTBT sample acquired at continental distance from a small, atmospheric explosion. These spectra have been analyzed to determine what additional confidence may be expected from confirmatory laboratory measurements.

The performance of the diverse group of detectors was estimated in several ways and as a function of time. In addition, the number of isotopes detected was tabulated for certain detectors as a function of source strength. These performance estimations should provide a clear indication of the trade-off between cost and measurement quality.

Key Words: Radionuclide, Aerosol, Fission, Isotope, Atmospheric

OBJECTIVE

While chemical dissolution of the filters may be an optional approach to laboratory analysis, high-sensitivity gamma-ray spectroscopy will be the best choice for sample re-analysis because it is relatively inexpensive, non-destructive, and the most easily adapted to diverse samples. While bBeta-gamma coincidence is probably the best choice for xenon isotopes, options in high-resolution gamma-ray spectroscopy with germanium detectors for aerosol samples are considered in this work.

The simulation scenario selected has been based upon experience gathered from actual atmospheric detonations measured at PNNL during the 1960's and 1970's. Although PNNL routinely monitored the atmosphere for nuclear detonations [2], the explosions listed in Table 1 were chosen because of the interesting down-wind distances (~10,000 km) and the range of explosion sizes (~20 kilotons).

Predicting probable downwind travel time of an atmospheric plume is not within the scope of this work. The average downwind travel time reported for the detonations shown above was chosen for the pre-sampling decay (12 days). An additional 3 days of decay in the CTBT station was added and finally an additional 3 days of sample-shipment time was allowed. The total pre-measurement decay time for this scenario is therefore 18 days.

The sample fissions reported in Table 1 were determined from 1-week samples acquired with high-volume displacement pumps and 1-week measurements on high-efficiency, multi-parameter sodium iodide counting systems. In order to adapt this for use in the CTBT measurement regime, the fissions/sample were adjusted to reflect the daily volume collected (20,000 m^3) by the Radionuclide Aerosol Sampler/Analyzer (RASA) [3]. Sample splitting is expected to occur before measurement of a sample at a laboratory, but since the degree of splitting has not been determined at the time of this writing, no sample splitting has been assumed.

Table 1.Historical Explosion Data and Equivalent RASA Sample

Explosion Date	Putative Yield	Sample fissions	RASA fissions
10-16-64	20 kt	7.54×10^9	4.43×10^{10}
5-14-65	>20 kt	3.60×10^9	3.35×10^9
10-27-66	<20 kt	7.67×10^{10}	7.14×10^{10}
12-24-67	15-20 kt	1.10×10^{10}	1.02×10^{10}
11-18-71	20 kt	1.47×10^9	2.45×10^9
1-7-72	<20 kt	2.03×10^9	3.38×10^9
1-23-76	<20 kt	2.57×10^8	4.28×10^8
9-17-77	15 kt	6.05×10^9	1.01×10^{10}
3-15-78	<20 kt	3.33×10^{10}	5.55×10^{10}
12-14-78	<20 kt	3.30×10^9	5.50×10^9
10-16-80	<20 kt	3.62×10^8	6.08×10^8

Quartz ampoules containing 1.2 micrograms of uranium solution were exposed to a thermal neutron flux at the Washington State University reactor to produce a total of 1.58×10^9 fissions per ampoule. Samples of this solution were stippled onto media representing two basic expected sample configurations: the cylindrical geometry of the RASA and the essentially point-like geometry of compressed filters.

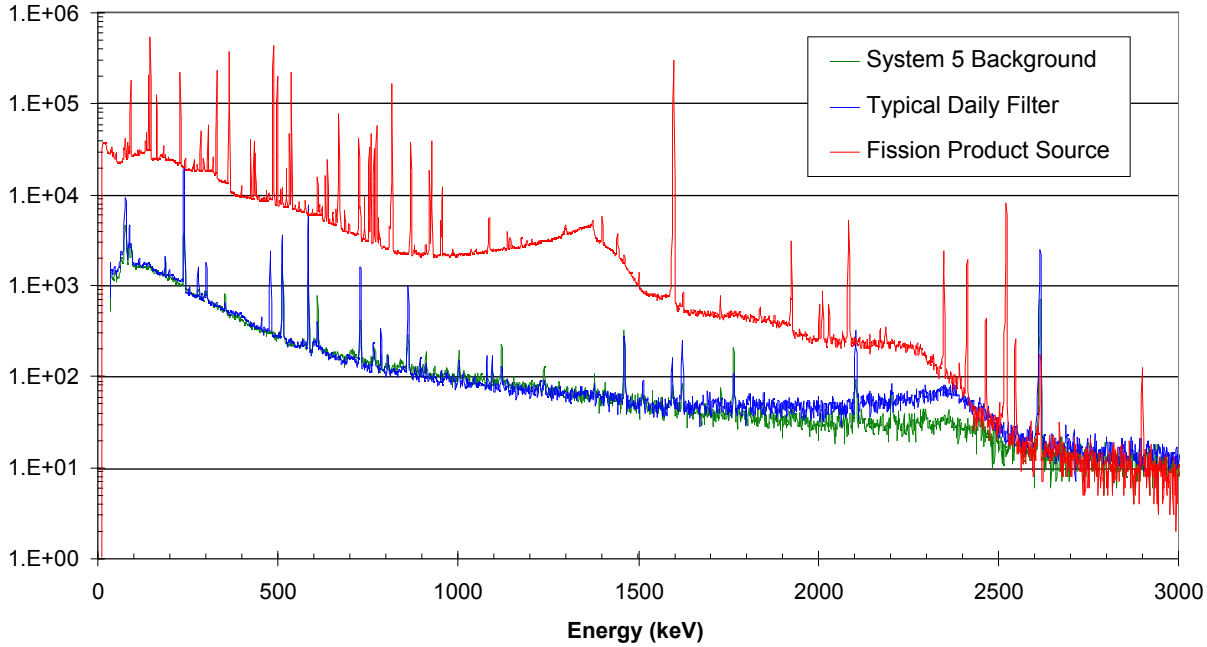

Figure 1. Fission Product Spectra (System 5)

The relatively challenging scenario selected for the experiment is a small detonation at a great distance. The CTBT laboratories will only need to fulfill basic requirements through certified procedures, and may therefore have different measurement approaches. The approaches selected for this experiment were selected to typify the CTBT lab approaches and to determine their expected performance.

The procedure expected of a CTBT lab will primarily consist of gamma-ray spectroscopy of samples already measured at a radionuclide station. Regular quality checks of the radionuclide stations are expected to constitute the majority of analyses performed. However, the ultimate mission of the laboratory is to enhance confidence in radionuclide results in the case of a suspected weapon violation. Thus laboratory samples will be mostly filters with natural isotopes only.

For these reasons, the laboratory detection procedure must exceed the sensitivity of the radionuclide stations by approximately an order of magnitude. This can be achieved by a combination of longer counting times, larger detectors, and lower backgrounds. Low background detectors are an excellent approach since the radon daughter activity in the filter will be much lower after a few days of decay provided by the sample transport time, provided the sample has been properly handled. Longer counting time is facilitated by allowing up to one week for the results of the lab measurement.

Several detection approaches have been evaluated, and are shown in Table 2. These systems are all in regular use at PNNL either in a typical counting-room or within a RASA. The MDA listed for these systems is derived from a 24-hour system background blank.

Table 2. Evaluated detection systems

No.	Relative Eff. HPGE Type	Shielding	MDA(Bq) ^{140}Ba	FWHM
1	90% P type	10-cm Pb, Passive	0.122	1.9 keV
2	36% P type	10-cm Pb, Passive	0.804	2.2 keV
3	47% N type	10-cm Pb, Passive	0.705	2.1 keV
4	41% P type	10-cm Pb, Passive	0.511	1.8 keV
5	29% P type	15-cm Pb, Active	0.044	2.8 keV

System 1 is a mechanically cooled P-type detector of 90% relative efficiency. This detector is similar to the detectors specified for use in the RASA. In this experiment, it is encased in a 10-cm thick lead cave with an inner copper lining. The source material was stippled onto a filter similar to a RASA filter and counted in a RASA geometry, i.e. wrapped around the detector. The detector resolution was measured to be 1.9 keV FWHM at 1332 keV.

System 2 is a 36% relative efficiency P-type detector in a 10-cm thick lead shield with an inner iron lining. The source was created in a point-like geometry and measured at a distance of 10 cm from the detector face, to minimize summing effects. System 2 has a measured resolution of 2.2 keV FWHM at 1332 keV.

System 3 is a 47% relative efficiency N-type detector in a shield similar to System 2 with a point-like source at 10 cm. System 3 has a thin Be entrance window and a resolution of 2.1 keV FWHM at 1332 keV.

System 4 is a commercial extra low-background detector encased in a low-background commercial shield. The detector is a 41% relative efficiency P-type detector and has a measured resolution of 1.8 keV at 1332 keV. The shield is a large diameter, 10 cm lead shell lined with copper. The point-like source was positioned 10 cm from the detector face.

System 5 is a custom built 29% relative efficiency P-type detector [4]. The construction materials have all been selected for low background. The shield is 15 cm of lead. The inner 5-cm is 160-year-old lead recovered from a sunken freighter. A 10-cm, active cosmic anti-coincidence system surrounds the entire shield. System 5 has a measured resolution of 2.8 keV FWHM at 1332 keV. The point-like source prepared for this system was placed at a distance of 1.9 cm from the side of the detector, making the geometry factor similar to System 1.

To fairly evaluate the systems under consideration, it may seem necessary to mathematically add in the signals expected from the measurement of atmospheric filters decayed to appropriate age. Figure 1 shows the comparison of a 14-day-old fission product spectrum to appropriately decayed high-volume aerosol filters. Besides the ^{208}Tl peak at 2614.47 keV, there is little impact. In actual fact, the fission product signal could be substantially smaller before radon daughters become important.

Automated gamma-ray analysis using RAYGUN [5] was chosen as the ultimate comparison of the detection approaches, and the results are tabulated in Table 3. All uncertainties, upper-limit values, and net peak areas determined by RAYGUN are based on a background continuum that RAYGUN computes for all regions of any given data spectrum. The RAYGUN data reduction code currently uses an algorithm to compute upper-limit values that is based on the branching fraction, the detection efficiency at the given energy and the computed background continuum in the region of the gamma-ray used to quantify a particular radionuclide. The reported upper-limit corresponds to a 95% Confidence Interval for the Minimum Detectable Amount (MDA) when using an integrating window that is ±1.4 FWHM around the centroid. (An integrating region-of-interest that is 1.4 FWHM-units wide is optimal for MDA calculations in cases where the presence of the radionuclide is low enough to be in question. A wider integrating

window is used by RAYGUN when a peak is definitely present and net counts are required to compute a quantitative activity level.)

Table 3. Spectral Analysis Results

	Fission Product	System 1		System 2		System 3		System 4		System 5	
	Isotopes	(Bq)	% Error	(Bq)	% Error	(Bq)	% Error	(Bq)	% Error	(Bq)	% Error
1	Cerium-141	230	1.0%	200	1.0%	250	1.0%	200	1.2%	250	1.2%
2	Barium-140	420	2.8%	350	3.3%	430	5.6%	400	2.1%	330	11.5%
3	Lanthanum-140	460	3.9%	420	2.3%	480	0.7%	460	0.7%	300	10.8%
4	Zirconium-95	42	3.1%	41	1.4%	36	6.6%	69	1.8%	52	5.1%
5	Molybdenum-99	40	9.9%	51	6.5%	58	14.7%	54	4.4%	65	21.3%
6	Ruthenium-103	81	1.0%	71	1.0%	77	1.1%	75	1.4%	57	1.2%
7	Neodymium147	110	6.2%	110	2.8%	110	3.2%	110	2.8%	56	28.5%
8	Iodine-131	130	2.5%	81	3.1%	84	2.7%	90	1.2%	85	6.8%
9	Tellurium-132	53	1.4%	61	1.0%	58	2.2%	59	1.4%	72	3.5%
10	Cerium-144	38	2.1%	24	3.7%	45	2.8%	32	7.9%	43	4.3%
11	Iodine-133	< 3.1		< 6.1		< 17		< 43		5.9	60.1%
12	Antimony-126	< 0.28		0.19	21.1%	< 1.3		< 2.0		0.4	74.9%
13	Antimony-127	2.3	9.8%	4.1	16.4%	3.7	9.4%	4	17.7%	< 2.7	
14	Cesium-137	1.2	5.8%	1.3	9.0%	< 0.84		< 1.4		0.5	41.2%
15	Cadmium-115	< 0.81		< 0.96		< 2.9		< 4.6		< 1.9	
16	Cesium-136	< 0.28		< 0.41		< 1.3		< 2.0		< 0.56	
17	Rhodium-105	< 1.7		< 2.0		< 5.8		< 11		8.0	36.7%
18	Silver-111	< 4.3		< 5.1		< 14		< 26		< 9.3	
19	Tellurium-129m	15	11.8%	< 30		< 84		< 130		< 39	
20	Zirconium-97	< 2.8		< 8.1		< 11		< 32		< 9.3	
21	Europium-156	< 4.1		< 6.1		< 14		< 33		< 7.0	
22	Ruthenium-106	22000	38.8%	25000	29.5%	11000	67.0%	150000	20.9%	19000	17.0%
23	Technetium-99m	< 0.62		-0.81		< 2.0		< 3.5		< 1.3	
24	Niobium-95	16	1.1%	13	1.3%	13	1.7%	15	2.3%	13	1.9%
25	Europium-155	< 0.90		< 1.2		< 3.0		< 4.6		< 2.8	
26	Antimony-125	< 1.2		< 2.0		< 5.0		< 9.8		< 2.8	
27	Yttrium-91	110	17.4%	< 111		< 290		< 500		< 150	
28	Tin-125	< 4.8		< 6.6		< 18		< 36		< 8.5	
29	Cerium-143	5.8	3.8%	5.57	4.0%	5.9	5.3%	5	16.5%	5.5	17.7%
30	Tellurium-131m	< 1.6		< 2.5		< 7.7		< 13		< 3.6	
31	Samarium-153	0.84		< 1.5		< 2.8		< 4.1		2.9	29.1%
32	Promethium-151	< 4.1		< 5.6		< 16		< 29		< 8.9	
33	Palladium-112	< 7.8		< 14		< 40		< 65		< 19	
34	Terbium-161	140	18.6%	< 220		110	7.0%	17	22.1%	310	4.5%
35	Cadmium-115m	< 19		< 46		< 98		< 160		< 69	
36	Silver-112	< 7.8		< 14		< 40		< 65		< 19	
37	Tin-123	< 32		< 54		< 130		< 220		< 74	
38	Strontium-89	< 150		220	68.4%	< 770		< 1100		210	71.9%
39	Cesium –134	< 0.44		< 0.71		< 2.4		< 3.2		< 0.88	
40	Palladium-111	< 4.3		< 5.1		< 14		< 26		< 9.3	
41	Strontium-91	< 1.4		2.0	30.0%	< 6.9		< 11.3		2.3	25.4%
42	Yttrium-93	51	3.9%	46	4.8%	53	6.1%	59	14.3%	52	12.4%
43	Palladium-109	83	5.3%	< 13		34	13.9%	< 45		< 27	
44	Tellurium-127m	<2700		< 3000		< 210		< 270		< 370	
45	Tellurium-127	< 47		< 71		< 210		< 360		< 113	
46	Antimony-128	< 0.62		< 1.3		< 3.0		< 6.0		< 1.7	
47	Iodine-130	< 0.51		< 1.0		< 2.7		< 4.9		< 1.5	
48	Iodine-135	< 4.6		< 5.6		< 13		< 32		< 7.8	
49	Promethium-149	< 6.3		< 8.1		20	27.5%	< 40		< 13	
50	Samarium-156	< 3.7		< 6.1		< 13		< 26		< 8.4	
51	Europium-157	< 6.7		< 10		< 29		< 50		< 15	
52	Iodine-132	250	5.1%	300	10.5%	370	4.6%	380	3.5%	250	3.5%

While the absolute efficiency of the counting geometries was not known exactly (it isn't needed for detection), variations in the loss of iodine isotopes in sample preparation may still be noted. Iodine isotopes represent a special challenge in this type of sample simulation. Since 1/3 to 1/2 of iodine in the atmosphere

from an explosion will be in a form suitable for filtration trapping, the iodine in the irradiated quartz ampoule was clearly at least twice what should be expected relative to, say, ^{99}Mo. On the other hand, after the sample has been diluted and stippled onto a substrate, then dried under heat lamps in a hood, the volatile iodine isotopes may be reduced.

CONCLUSIONS AND RECOMMENDATIONS

The data in Table 3 may be manipulated in several ways to evaluate the different detection approaches. The most trivial would be to count the number of detected isotopes for each system. Additional isotopes, particularly if in reasonable ratios, would constitute enhanced confidence that a marginal IMS radionuclide detection is defensible. One may require, however, that only the higher quality isotopes should be used for this determination. Table 4 shows the cumulative number of isotopes for each system with error bars at or below a range of threshold values.

The comparison between the systems is approximately a tie between System 1 and System 4 when very restrictive limits are placed on isotopic detections. Where any detection is counted, System 5 is superior, and System 4 is worst. However, it is clear that this measure does not strongly differentiate the systems.

A Figure of Merit (FOM) is desired which more directly captures the enhanced confidence that the laboratory measurement provides. The first concept that was considered by the authors was to estimate the average confidence in a hit by a given system, then multiply by the number of isotopes detected by that system.

Assuming that one could construct the confidence level of a detection from a RAYGUN output, however, it is obvious that the result of this exercise would be simply the sum of the confidence of each isotope. Thus, one need only construct a relative measure of confidence for each isotope, sum all for a given system, then compare. The same thresholds may be applied to judge the systems only on the best isotopes, all isotopes, or any threshold between.

A candidate for detection confidence may be simply formed by inverting the magnitude of the error bar. Thus, an isotope with a 50% uncertainty would have a confidence FOM of 2, while an isotope detected with a 1% uncertainty would have a confidence FOM of 100. Table 5 shows this FOM computed for the same thresholds shown in Table 4.

This measure shows System 4 to be superior regardless of how restrictive the acceptance criteria. This information can be interpreted in several ways and there are several implications. Firstly, modestly lowered background reduction with a modestly sized detector are superior to a larger (x2) detector in a simple shield or an elaborate shield but smaller (x0.7) detector.

The failure of System 5 to exceed the other systems is due to the fact that the sample-associated background dominates the spectrum. In fact, even the radon daughters in the filter were not important. So with the low background masked by the source, the only remaining important feature of the detector was its small size. This shows that a lower MDA for ^{140}Ba computed from system background may be a deceptive measure of system value.

The recommendation that may be drawn from this experiment is that there is more than one way to adequately configure laboratory hardware to perform a confirmatory measurement on a CTBT aerosol sample. The best results for samples with significant fission-product content will come from simultaneously reducing background and increasing detector size. Ultra-low-background detectors will not have an impact on this problem. Extremely large detectors will also not improve the detection greatly.

In fact, it is the interference of the different fission products that limits the confidence FOM. In this light, it would seem that greater selectivity might be the way to increase the FOM, if increases were desired. Improved resolution or coincidence techniques could be employed to increase selectivity, but the immediate advantage would only be to shorten the time needed to reach the required sensitivity, and thus increase the throughput of the laboratory.

On the other hand, large or ultra-low background detectors (Systems 1 and 5) can fill an important role for samples that have no significant fission product content, which have several days of radon-daughter decay, and which have been handled in a manner that does not add contamination. In this case, the ^{140}Ba MDA listing in Table 2 is more indicative of system performance. It should be remembered that the vast majority of samples are expected to be of this type.

The final conclusion is all CTBT parties would be best served by well equipped laboratories which can have procedures in place for both low and high activity samples.

Table 4. System Comparison by Isotopes Detected

Max Error %	System 1	System 2	System 3	System 4	System 5
2%	4	5	4	6	3
3%	8	9	8	9	3
5%	11	12	9	11	7
10%	17	15	15	12	9
20%	20	16	17	15	14
30%	20	19	18	17	18
50%	21	19	18	17	20
100%	21	20	19	17	23

Table 5. System Comparison by Confidence FOM

Max Error %	System 1	System 2	System 3	System 4	System 5
2%	362	448	393	508	219
3%	486	528	511	635	219
5%	596	663	564	686	322
10%	688	689	657	699	356
20%	707	705	671	717	394
30%	707	717	675	727	409
50%	710	717	675	727	415
100%	710	718	676	727	419

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