EVALUATION OF FISSION PRODUCT ISOTOPES FOR FIELD OR LABORATORY DETECTION

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ABSTRACT

An International Monitoring System (IMS) is being created to monitor the Comprehensive Nuclear-Test-Ban Treaty (CTBT). Radionuclide aerosols will be monitored to provide positive proof of an atmospheric explosion. In addition, a group of laboratories will perform quality assurance and confirmatory analyses of samples of interest. The field and laboratory systems will perform gamma-ray spectrometric analysis of air filters. While laboratories may undertake additional analysis such as chemical separation and beta counting, the scope of this work is to make evaluations with respect only to gamma-ray spectrometry.

Short lived fission product spectra have been acquired on gamma-ray spectrometers appropriate for both field and laboratory measurements of compliance with the Comprehensive Nuclear Test Ban Treaty. These spectra simulate a reasonable CTBT sample indicative of a small atmospheric explosion occurring at continental distance from a monitoring station. The radionuclides can be ranked by the measurement uncertainty, signal-to-noise ratio, and other means. Additional isotopes may be ranked by using simulated spectra from individual isotopes in a variational method. The results of this ranking are shown and could contribute to the formation of a list of reasonable isotopes to trigger a CTBT bulletin.

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

OBJECTIVE

This work seeks to experimentally establish a ranked list of isotopes observable in a challenging scenario: a 1 kt explosion in the atmosphere at about 10,000 km upwind.

The function of the IDC requires a library of radionuclides for automated aerosol spectral analysis, and a library of relevant radionuclides is needed to screen spectra into a multi-level categorization scheme. In particular, a list of fission products typical of an atmospheric explosion is required to highlight events of increased importance. The categorization scheme requires detection of one relevant fission-product isotope for Level 4 designation, and two relevant fission-product isotopes for Level 5 designation. Samples that generate Level 5 spectra may be sent to a laboratory for confirmatory analysis. The case for xenon isotopes is substantially different and will not be discussed here.

RESEARCH ACCOMPLISHED

A given radionuclide should rank highly if it has several favorable criteria. These include:

- Good $T_{1/2}$ or product of another isotope with good $T_{1/2}$
- Good decay characteristics
- Good branching ratio
- Good gamma-ray energy
- No or few interferences with natural isotopes
- No or few interferences with calibration isotopes
- No or few interferences among fission products

Most of these criteria lend themselves well to simple calculation. Even the influence of the natural background may be trivially calculated by computing minimum detectable concentrations based upon a reasonable atmospheric sample [1]. Unfortunately, potential interferences with other fission products are not so easily resolved.

Computation methods do exist for synthetically creating spectra [2], and this would allow for the cross interferences among fission isotopes. However, to provide a close simulation of reality we would first need to correct the fission product lists for differing deposition rates of chemical species in the atmosphere and other effects. Given that this data is unknown and potentially varies in specific regions, detonation environments, and times of year, the simplest approach is to physically create the sources in a rapid reactor irradiation. The spectra that are produced may contain certain anomalies that can be ignored:

- Relatively high 1001 keV (^{234m}Pa) and 185 keV (^{235}U)
- Minimal loss of iodine isotopes

To experimentally take all the above parameters (except fractionation) into consideration, we created a fission product source experimentally by irradiating ampoules containing an enriched-uranium solution. Quartz ampoules containing 1.2 micrograms of uranium solution were exposed to a thermal neutron flux at the University of Washington reactor to produce a total of 1.58×10^9 fissions per ampoule. The liquid was calibrated for fission product content, and a small quantity was introduced as a source into a detector of very low background. The liquid was diluted and a sample representing a simulated atmospheric explosion detection filter was created. The activity was selected using data from historic atmospheric sampling at PNNL [3], specifically 11 samples where an atmospheric detonation at Lop Nor was detected in Richland, WA (yield: ~20 kt, range: ~10,000 km, travel time: ~12 days). Variation between reactor fission product yields and weapon fission product yields was ignored, since the results are essentially qualitative.

Table 1. Original Fission Product Library for Automated Analysis

Be-7	Cd-115	Cs-134
K-40	Cd-115m	Cs-136
Sr-89	Sn-123	Cs-137
Sr-91	Sn-125	Ba-140
Y-91	Sb-125	La-140
Y-93	Sb-126	Ce-141
Zr-95	Sb-127	Ce-143
Zr-97	Sb-128	Ce-144
Nb-95	Te-127	Nd-147
Nb-97	Te-132	Pm-149
Mo-99	Te-127m	Pm-151
Tc-99m	Te-129m	Sm-153
Ru-103	Te-131m	Sm-156
Rh-105	I-130	Eu-155
Rh-106	I-131	Eu-157
Pd-109	I-132	Tb-161
Ag-111	I-133	
Ag-112	I-135	

The number of sample fission atoms was 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 3.8×10^8 fissions to reflect the daily volume collected (20,000 m3) by the Radionuclide Aerosol Sampler/Analyzer (RASA) [4]. 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.

The spectrum obtained from this sample was evaluated using an automated analysis code, RAYGUN [5], which has many functional similarities to the code used by the IDC and treaty signatories. The library used for the automated RAYGUN analysis was reasonably comprehensive (see Table 1), containing 53 isotopes and 273 gamma-rays.

This library was derived from fission product libraries previously considered for a relevant radionuclide list [6], with several additions and deletions. The nuclear decay information that makes up this library (gamma-ray energies, half life, and branching ratios) was compiled from various sources and adjusted to maximize the likelihood of detection.

Simulated 14-day Detection Results

In order to gain some idea of the effectiveness of various isotopes in detection, selected isotopes were ranked using several methods. The aggregate ranking is shown in Table 2. This ranking also includes isotopes that were below the level of detection. This allows us to determine which isotopes would be the next likely list entries.

Direct spectral comparison of the 14-day-old fission spectra and a typical daily filter on a RASA in Richland, WA indicated that the normal atmospheric isotopes and background signals (radon daughters, K-40, Be-7) did not play a significant role. Therefore, no daily filter spectral data were added in.

The appearance of Cs-137 on this list is slightly surprising, given the long half-life. Since Cs-137 may be re-suspended from old weapons debris or from Chernobyl fallout, this isotope will not add great confidence if it is one of only two detected isotopes.

Since only particulate forms of iodine isotopes are expected to be captured by aerosol filters, the technique used to create the test sample probably contained artificially elevated iodine levels. For this reason, iodine isotopes were deleted from this list. This is a judgement based on the fact that the chemical form of iodine may vary and the measured quantity will be highly dependent on the percentage of iodine in particulate form. It could be argued that this deletion represents a lost detection capability, and certain iodine isotopes may need to be added to the list, although the evidence above indicates that it is not likely needed to detect an atmospheric explosion. It was also noted that the many lines of iodine isotopes could cause difficulty with the data reduction algorithms.

Effects of Decay Time

One might argue that the list derived above will only be applicable at the 14-day decay time, since the growth and decay of isotopes may greatly adjust the relative worth of an isotope. For this reason, spectral data from the same sample at a short decay time (2 days) was also analyzed in the same way. This spectrum is not a logical extension of the original scenario: it has 2-days worth of decay but 14-days worth of dilution. It is intended only to show the effects of decay on the table generated from the 14-day measurement.

Table 2. Ranking of 14-day-old sample of fission products.

Ranking	Radionuclide/Pair	T 1/2	Energy (keV)	Comments
1	Ce-141	32.5 d	145.4	
2	Ba-140/La-140	12.7 d	537.3/487.0	
3	Mo-99	2.75 d	140.5/739.6	140.5 most intense, but low energy attenuates
4	Zr-95/Nb-95	64 d	756.7/765.8	
5	Ce-144	285 d	133.5	
6	Nd-147	11 d	531	Interference on all peaks
7	Sm-153	1.9 d	103.2	
8	Pd-112/Ag-112	0.88 d	617.5	
9	Cs-137	11000 d	661.7	
10	Sr-89	50.5 d	910.3	Te-131m interferes with the only peak
11	Cs-136	13.2 d	1048.1	Not seen in spectrum
12	Cs-134	752 d	604.7/795.8	Not seen in spectrum
13	Zr-97/Nb-97	0.70 d	743.4/657.9	Not seen in spectrum
14	Tb-161	6.9 d	57.1	Not seen in spectrum
15	Eu-156	15.2 d	1230.7	Not seen in spectrum
16	Pd-111/Ag-111	7.5 d	342.1	Not seen in spectrum
17	Cd-115	2.2 d	336.2	Not seen in spectrum
18	Cd-115m	45 d	933.8	Not seen in spectrum

Note: Radionuclides 1-10 are ranked according to number of net counts in the preferred peak and/or the statistical uncertainty assigned by the reduction software (based on counts in peak, interference, attenuation, etc.) Radionuclides 11-18 were either not visible in the spectrum or were not clearly identified by the reduction software. Synthetic spectra [5] were added until the software identified the radionuclide, in which case ranking is by the activity level required to achieve software identification.

The two-day ranking is so similar to the 14-day list, it is clear that decay times between 2 and 14 days are not greatly significant.

Table 3. Ranking of 2-day-old sample of fission products.

Ranking	Radionuclide/Pair	T 1/2	Energy (keV)	Comments
1	Mo-99	2.75 d	140.5/739.6	140.5 is most intense, but low energy attenuates
2	Ba-140/La-140	12.7 d	537.3/487.0	
3	Zr-97/Nb-97	0.70 d	743.4/657.9	
4	Ce-141	32.5 d	145.4	
5	Zr-95/Nb-95	64 d	756.7/765.8	
6	Tb-161	6.9 d	57.1	
7	Sm-153	1.9 d	103.2	
8	Ce-144	285 d	133.5	
9	Nd-147	11 d	531	Interference on all peaks
10	Pd-112/Ag-112	0.88 d	617.5	
11	Sr-89	50.5 d	910.3	Te-131m interferes with the only peak
12	Cs-134	752 d	604.7/795.8	Not seen in spectrum
13	Eu-156	15.2 d	1230.7	Not seen in spectrum
14	Cs-136	13.2 d	1048.1	Not seen in spectrum
15	Cs-137	11000 d	661.7	Not seen in spectrum
16	Pd-111/Ag-111	7.5 d	342.1	Not seen in spectrum
17	Cd-115m	45 d	933.8	Not seen in spectrum
18	Cd-115	2.2 d	336.2	Not seen in spectrum

Note: Radionuclides 1-11 are ranked according to number of net counts in the preferred peak and/or the statistical uncertainty assigned by the reduction software (based on counts in peak, interference, attenuation, etc.) Radionuclides 12-18 were either not visible in the spectrum or were not clearly identified by the reduction software. Synthetic spectra [5] were added until the software identified the radionuclide, in which case ranking is by the activity level required to achieve software identification.

CONCLUSIONS AND RECOMMENDATIONS

While a human analyst could have adjusted the data analysis to glean more from these spectra, the list of automatically detected radionuclides in Table 2 represents the easily detectable isotopes in the spectra. This list should therefore be adequate for normal event screening into a multi-level scheme. Given that the current concept of the screening system contains only 1 or 2 fission products corresponding to Level 4 or Level 5, a list of the 10 most likely isotopes should be adequate under all circumstances. With the exception of iodine isotopes, the detection of other fission products not on this list (for instance from Table 1 but not Table 2) without some or all of the isotopes in Table 2 is highly unlikely, and may constitute a pathological sample requiring special consideration.

For example, elevated Be-7 is likely due to unusually high cosmic-ray spallation in the atmosphere. Cosmic ray neutrons can spoof the detection of Mo-99, due to neutron reactions in the germanium crystal. On the other hand,
the high volatility of iodine may allow the observation of only iodine isotopes from a leaky underground test. This scenario argues for the addition of iodine isotopes as appropriate to Table 2.

There is little recurring cost in maintaining a large library for automated data processing. One possible pitfall is that radon daughter gamma-ray lines may be at similar energies and if efficiency data are in error, an isotope may be erroneously detected. Otherwise, there is no cost in allowing States Parties subscribers to create custom screening algorithms based on custom data flags developed for the IDC.

Although this simulated 1 kt detection is statistically unquestionable, it should be noted that the only intra-element isotopic ratio that can be formed of these detected isotopes is Ce-141/Ce-144, which is not the

most desirable. This general result supports the conclusions previously established concerning the difficulty in broadly applying fission product ratios [7].

Table 4. Total Fission Product vs. Total Activation Product Activity in ORIGEN Calculations in Underground, Underwater, and Atmospheric Activation Scenarios.

	3 days	10 days	30 days
U-235 FP	1.3E+17	3.5E+16	1.0E+16
UG AP	1.2E+16	3.7E+14	2.1E+14
UW AP	2.1E+15	1.3E+14	1.0E+14
ATM AP	3.8E+12	3.4E+12	2.6E+12

Note: All values in Bq.

Concerning activation products: since a reasonable 1 kt detection at 10,000 km after 14 days leaves a large signal, there is no need to use activation products in the Relevant Radionuclide List. Table 4 compares gross activity from several activation scenarios with fission products [8]. It could be argued that these isotopes should be in the automated data processing library, thereby allowing data-product subscribers to flag these isotopes. However, while this may be useful for States Parties purposes, it is not necessary for detection of a test ban treaty violation.

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