PNNL Review of Proposed Relevant Radionuclide List

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PNNL Review of Proposed Relevant Radionuclide List

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Introduction

A list of fission products and activation products has been proposed for possible adoption as an official table of relevant isotopes for CTBT use. It is our understanding that the purpose of this list is to discriminate Level 4 spectra from Level 5 spectra in the decision logic diagram. The current understanding is that a single short-lived, relevant isotope that is atypical for a station would cause a spectrum to be marked as Level 4. A second relevant isotope would cause a spectrum to be marked as Level 5, which would perhaps require a sample to undergo additional laboratory confirmation measurements.

The list consists of a very comprehensive set of fission products and activation products. We have examined the list for accuracy and have also flagged potential problems with members of the list. In our opinion, several of these isotopes have serious problems and many have no practical chance of ever being the first or second detected isotopes. We are not arguing whether or not these isotopes might be seen in a large atmospheric test. On the other hand, there may be no harm associated with having a long list.

The issue of activation products is different. Some activation products are indicative of the soil or rock composition in the vicinity of an explosion. Others may only be dependent on materials in the weapon or in the support structures. We don't think that a great deal of analysis of these isotopes by the CTBTO should be encouraged. In any case, if particulate activation products are in the atmosphere, fission products should be even more prevalent, thus removing the need for an activation list component.

In order to inject some reality into the discussion, we point out that for 11 historical low-yield Chinese atmospheric tests detected at PNNL (~20 kt, range ~10,000 km, travel time ~12 days), a RASA would have collected an average of 6×10^9 fission atoms on a filter. Since the detection threshold for the RASA (and other CTBT RN aerosol systems) is about 10^6 fissions, we claim that fission products alone will allow very high-confidence detection of atmospheric detonations, and that the major fission products may well cover up the minor fission products.

Table 1. Low-yield Chinese Weapon Tests Detected in Richland, WA. RASA Fissions are computed by applying the ratio of actual air volumes to the daily RASA air volume.

Date	Putative Yield	Measured Fissions	RASA Fissions
10/16/64	20 kt	7.54E+09	4.43E+10
5/14/65	>20 kt	3.60E+09	3.35E+09
10/27/66	<20 kt	7.67E+10	7.14E+10
12/24/67	15-20 kt	1.10E+10	1.02E+10
11/18/71	20 kt	1.47E+09	2.45E+09
1/7/72	<20 kt	2.03E+09	3.38E+09
1/23/76	<20 kt	2.57E+08	4.28E+08
9/17/77	15 kt	6.05E+09	1.01E+10
3/15/78	<20 kt	3.33E+10	5.55E+10
12/14/78	<20 kt	3.30E+09	5.50E+09
10/16/80	<20 kt	3.62E+08	6.08E+08

Explanation of the Comments

Table 3a and 3b provided by Dr. Steve Lewis have been augmented in order to provide simple, isotope-by-isotope comments and are located at the bottom of this paper. The new columns are:

Nuclide Half Life	A half life <1d means that after just a few days travel time, this isotope will be relatively unimportant.
Nuclide Production Yield	Especially low yields are reported in percentages. Anything low compared to 6% will not have a good chance to be the first or second isotope.
Nuclide γ Energy or Abundance	Low gamma energy or low gamma abundance makes an isotope more vulnerable to background.
Gamma Interferences	A gamma ray in close proximity to a natural isotope or other fission product will either be unusable or require special rehabilitation.

In addition, a column entitled 'Comments and Suggestions' reports a variety of problems or alternate selected gamma rays. For instance, certain isotopes are actually measured using daughter isotopes' gamma ray lines. Other isotopes are typically used as calibration standards and would be frequently detected as shine-through in the RASA and other CTBT RN aerosol systems. Table 4 is a direct comparison between the lists which comprise Table 3 and PrepCom Task Leader Paper TL 2/10.

An Experiment

In order to gain some idea of the effectiveness of various isotopes in detection, we created a fission product source by irradiating a uranium bearing liquid in a reactor. 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 spectrum obtained from a 1 day count of the source at age 14 days was normalized to represent the result of the collection of 3.8×10^8 fissions (1 kt explosion 10,000 km upwind). An automated analysis of the spectrum using the informal PNNL CTBT isotope library was performed. The resulting detected isotopes have been sorted in Table 2.

Table 2. Sorted isotopes in a 14-day-old fission product spectrum. These isotopes are sorted by concentration and by error in the concentration (a measure of confidence). The library used did not contain all the isotopes in the DeGeer lists. Minimum Detectable Concentrations (MDC's) for unobserved isotopes are also shown in the Sorted by Concentration columns.

Sorted by Error				Sorted by Concentration			
Rank	Isotope	Error	Bq/SCM	Rank	Isotope	Bq/SCM	Error
1	Ce141	1.03	1.02E-02	1	Ba140	1.43E-02	10.99
2	Te132	1.03	2.99E-03	2	La140	1.27E-02	9.01
3	Ru103	1.04	2.36E-03	3	I132	1.06E-02	3.5
4	Ce144	1.94	1.75E-03	4	Ce141	1.02E-02	1.03
5	I132	3.5	1.06E-02	5	Nd147	4.24E-03	23.23
6	Sb127	5.63	1.81E-04	6	I131	3.47E-03	9.59
7	Mo 99	6.84	1.47E-03	7	Te132	2.99E-03	1.03
8	Zr 95	8.66	2.16E-03	8	Ru103	2.36E-03	1.04
9	La140	9.01	1.27E-02	9	Zr 95	2.16E-03	8.66
10	I131	9.59	3.47E-03	10	Ce144	1.75E-03	1.94
11	Ba140	10.99	1.43E-02	11	Mo 99	1.47E-03	6.84
12	Cs137	13.79	2.04E-05	12	Sb127	1.81E-04	5.63
13	Cs136	15.06	2.65E-05	13	Cs136	2.65E-05	15.06
14	Co 60	18.07	1.53E-05	14	Cs137	2.04E-05	13.79
15	Nd147	23.23	4.24E-03	15	Co 60	1.53E-05	18.07

MDC's for unobserved isotopes in library

	Isotope	Bq/SCM	Error
N/A	Ce143	1.89E-04	100
N/A	Ag111	1.52E-04	100
N/A	I133	1.05E-04	100
N/A	K 40	6.09E-05	100
N/A	Zr 97	5.41E-05	100
N/A	Cd115	3.76E-05	100
N/A	Ag110	1.18E-05	100
N/A	Cs134	1.03E-05	100
N/A	Na 24	1.73E-06	100

Note: The computation of error includes both statistical (counting) errors and other sources of error.

Conclusions

Our conclusion is that in a near worst case scenario where the plume avoids detection for 2 weeks and only one station intercepts, the signal should still be incontrovertible. A list of relevant nuclides for the purpose of screening events could be very short with little fear of missing any real event. Our recommendation is to eliminate activation products, consider elimination of certain problem fission products, and resist adding new fission products except for good reasons. In addition, we recommend that the gamma ray energies should either be deleted or at least have the energies rounded to the nearest keV. Finally, we recommend the adoption of a single nuclear data source for half-lives, gamma-ray energies, and branching ratios.

Table 3a. Proposed Radionuclide List: Fission products.

Fission Product Nuclide	Half-life	Primary γ energy in keV*	Primary γ intensity in %	Nuclide Half Life	Nuclide Prod'n Yield	Nuclide γ Energy or Abund	γ Interference	Comments & Suggestions
Strontium-91	9.63	h	1024.3	33.4	< 1 d			Also 556 keV (61%)
Yttrium-91 •	58.51	d	1204.8	0.30		< 1 %		
Yttrium-93	10.18	h	266.9	7.32	< 1 d			
Zirconium-95 •	64.02	d	756.7	54.5				
Niobium-95 •	34.975	d	765.8	99.8			768 (Bi-214)	
Zirconium-97 •	16.91	h	743.4	93.1	< 1 d		Sb-128	
Molybdenum-99 •	65.94	h	140.5 D	89.4			Tc-99m	
Technetium-99m	6.01	h	140.5	89.1	< 1 d		Mo-99	
Ruthenium-103 •	39.26	d	497.1	90.9				
Rhodium-105 •	35.36	h	319.1	19.2				
Ruthenium-106 •	373.59	d	621.9 D	9.93				γ is from Rh-106 daughter
Palladium-109	13.712	h	88.0	3.61	< 1 d	Low E, < 5%		
Silver-111 •	7.45	d	342.1	6.7				
Palladium-112	21.03	h	617.5 D	43	< 1 d	< 0.017		
Cadmium-115m	44.6	d	933.8	2.0		< 0.017	< 5 %	934 (Bi-214)
Cadmium-115 •	53.46	h	336.2	45.9		< 0.017		339 (Ac-228)
Tin-123	129.2	d	1088.6	0.6		< 0.017	< 1 %	
Tin-125 •	9.64	d	1067.1	10				
Antimony-125 •	2.7582	y	427.9	29.6				
Antimony-126 •	12.46	d	695.0	99.6			Te-129m	Also 666 keV (99.6%)
Antimony-127 •	3.85	d	685.7	36.8				
Tellurium-127m	109	d	57.6	0.50		Low E, < 1%		
Tellurium-127	9.35	h	418.0	0.99	< 1 d	< 1 %	415	
Antimony-128	9.01	h	743.2	100	< 1 d		Zr-97	Also 754 keV (100%)
Tellurium-129m •	33.6	d	695.9	3.19		< 5 %	Sb-126	
Iodine-130	12.36	h	536.1	99	< 1 d			Also 668 keV (97%) Protected isotope
Tellurium-131m	30	h	773.7	49.9			Te-132	
Iodine-131 •	8.02070	d	364.5	81.7				
Tellurium-132 •	3.204	d	772.6 D	75.6			Te-131m	Also 668 keV (99%)
Iodine-133 •	20.8	h	529.9	87	< 1 d		Nd-147	
Iodine-135	6.57	h	1260.4	28.9	< 1 d			Also 1132 keV (23%)
Cesium-136 •	13.16	d	1048.1	80				Also 818 keV (100%) Protected isotope
Cesium-137 •	30.07	y	661.7 D	85.1				
Barium-140 •	12.752	d	537.3	24.39				
Lanthanum-140 •	1.6781	d	1596.2	95.4				
Cerium-141 •	32.501	d	145.4	48.2				
Cerium-143 •	33.039	h	293.3	42.8			295 (Pb-214)	
Cerium-144 •	284.893	d	133.5	11.09				
Neodymium-147 •	10.98	d	531.0	13.1			I-133	
Promethium-149	53.08	h	286.0	3.1		< 5 %	288 (Bi-212)	
Promethium-151	28.40	h	340.1	22.5			339 (Ac-228)	
Samarium-153	46.27	h	103.2	31.4				
Europium-155 •	4.7611	y	105.3	21.2				
Samarium-156	9.4	h	203.8	20.8	< 1 d	< 0.017		
Europium-156 •	15.19	d	1153.7	6.8			1155 (Bi-214)	Also 1231 keV (89%)
Europium-157	15.18	h	370.5	11	< 1 d	< 0.007		Also 413 keV (19%)

* D - denotes γ is from a Daughter product.

Table 3b Proposed Radionuclide List: Activation products.

Non-fission Product Nuclide	Half-life	Primary γ energy in keV	Primary γ intensity in %	Nuclide Half Life	Nuclide γ Energy or Abund	γ Interference	Comments & Suggestions
Sodium-24 ●	14.959	h	1368.6	100	< 1 d		
Potassium-42	12.36	h	1524.7	18.1	< 1 d		
Scandium-46	83.79	d	889.3	100			
Scandium-47	3.3492	d	159.4	67.9			
Chromium-51 ●	27.702	d	320.1	10			
Manganese-54 ●	312.12	d	834.8	100			
Cobalt-57 ●	271.79	d	122.1	85.6			Energy Calib. Std.
Cobalt-58 ●	70.82	d	810.8	99			
Iron-59	44.503	d	1099.2	56.5		1094 (Ti-208)	
Cobalt-60 ●	1925.1	d	1332.5	100			Energy Calib. Std.
Copper-64	2.7	h	1345.8	0.47	< 1 d	< 1 %	1347 (Ti-208 sum peak)
Zinc-65 ●	244.26	d	1115.5	50.6			
Zinc-69m	13.76	h	438.6	94.8	< 1 d		
Gallium-72	14.1	h	834.1	95.6	< 1 d		
Arsenic-74	17.77	d	595.8	59.4			
Arsenic-76	1.0778	d	559.1	45			
Rubidium-84	32.77	d	881.6	69.0			
Rubidium-86	18.631	d	1076.7	8.64		1079 (Bi-214)	
Yttrium-88 ●	106.65	d	1836.1	99.2			Energy Calib. Std.
Zirconium-89	78.41	h	909.0	99.9		911 (Ac-228)	
Rhodium-102	207	d	475.1	47			
Silver-106m	8.28	d	717.2	28.9			
Silver-108m	418	y	722.9	90.8		722	
Silver-110m	249.79	d	657.8	94			
Antimony-120	5.76	d	1171.7			< 2 %	1172 (Co-60)
Antimony-122 ●	2.7238	d	564.2	69.3			
Antimony-124 ●	60.20	d	602.7	97.8			
Cesium-132	6.479	d	667.7	97.5			
Barium-133 ●	10.52	y	356.0	62.05			
Cesium-134	2.0648	y	604.7	97.6			
Europium-152m	9.3116	h	841.6	14.6	< 1 d		
Europium-152 ●	13.537	y	1408	20.9		1408 (Bi-214)	
Thulium-168	93.1	d	816.0	50.0			
Thulium-170	128.6	d	84.2	3.26		Low E < 5 %	
Tungsten-187	23.72	h	685.7	27.3	< 1 d		
Iridium-190	11.78	d	186.7	52.4		186 (Ra-226, U-235)	
Iridium-192 ●	73.827	d	316.5	82.81			
Gold-196	6.183	d	355.7	86.9		295 (Pb-214)	
Gold-196m	9.7	h	147.8	42.5	< 1 d		
Gold-198 ●	2.69517	d	411.8	96			
Lead-203	51.873	h	279.2	81		277 (Ti-208)	
Radium-224	3.66	d	241.0	4.1		< 5 %	238 (Pb-212)
Uranium-237 ●	6.75	d	208.0	21.2			
Neptunium-239 ●	2.3565	d	277.6	14.38		277 (Ti-208)	
Americium-241 ●	432.2	y	59.5	35.9		Low E	Energy Calib. Std.

Table 4. Comparison of Proposed Relevant Radionuclide List and Task Leader Paper TL-2/10

DeGeer/Lewis Fission List

PrepCom Task Leader Paper TL-2/10,

Table 1

				Differences (%)			
		Half Life		Energy		Branching	
		Half Life		Energy		Branching	
Strontium-91	9.63 h	1024.3	33.40%	11.3 h	264.4	56.1%	
* Yttrium-91	58.51 d	1204.8	0.30%	38.8 h	239.1	1.4%	0.00%
Yttrium-93	10.18 h	266.9	7.32%	9.63 h	555.6	63.0%	
* Zirconium-95	64.02 d	756.7	54.50%	2.71 h	1383.9	90.0%	0.79%
* Niobium-95	34.975 d	765.8	99.80%	3.54 h	934.5	13.9%	0.00%
* Zirconium-97	16.91 h	743.4	93.10%	10.1 h	267.0	7.0%	0.00%
* Molybdenum-99	65.94 h	140.5	89.40%	64.02 d	756.7	54.5%	0.00%
Technetium-99m	6.01 h	140.5	89.10%	3.61 d	235.7	24.1%	0.00%
* Ruthenium-103	39.26 d	497.1	90.90%	34.97 d	765.8	99.8%	0.01%
* Rhodium-105	35.36 h	319.1	19.20%	23.4 h	778.2	96.9%	0.06%
* Ruthenium-106	373.59 d	621.9 D	9.93%	16.9 h	743.4	92.5%	0.00%
Palladium-109	13.712 h	88	3.61%	2.7476 d	739.6	12.1%	0.00%
* Silver-111	7.45 d	342.1	6.70%	6.01 h	140.5	89.1%	0.00%
Palladium-112	21.03 h	617.5 D	43.00%	39.27 d	497.1	91.0%	-0.03%
Cadmium-115m	44.6 d	933.8	2.00%	35.4 h	318.9	19.1%	-0.11%
* Cadmium-115	53.46 h	336.2	45.90%	4.44 h	724.5	49.0%	0.03%
Tin-123	129.2 d	1088.6	0.60%	1.020 y	622.1	9.9%	-0.03%
* Tin-125	9.64 d	1067.1	10.00%	13.5 h	88.03	3.6%	-0.28%
* Antimony-125	2.7582 y	427.9	29.60%	7.47 d	342.1	6.7%	0.00%
* Antimony-126	12.46 d	695	99.60%	21.04 h	617.4	43.0%	0.00%
* Antimony-127	3.85 d	685.7	36.80%	5.37 h	298.6	10.0%	0.02%
Tellurium-127m	109 d	57.6	0.50%	44.6 d	933.8	2.0%	0.00%
Tellurium-127	9.35 h	418	0.99%	2.228 d	336.2	45.9%	0.00%
Antimony-128	9.01 h	743.2	100.00%	129.2 d	1088.6	0.6%	0.00%
* Tellurium-129m	33.6 d	695.9	3.19%	9.63 d	1067.1	9.0%	0.10%
Iodine-130	12.36 h	536.1	99.00%	2.758 y	427.9	29.4%	0.01%
Tellurium-131m	30 h	773.7	49.90%	12.4 d	695.0	99.7%	0.48%
* Iodine-131	8.0207 d	364.5	81.70%	3.84 d	685.7	35.3%	0.26%
* Tellurium-132	3.204 d	772.6 D	75.60%	9.01 h	754.0	100.0%	-1.45%
* Iodine-133	20.8 h	529.9	87.00%	4.4 h	812.6	43.5%	0.00%
* Detected by Swedish Surveillance Network				33.6 d	459.6	4.5%	0.00%
Using different gamma ray and branching ratio				1.25 d	773.7	38.1%	0.00%
Using an isotope unique to the listing				11.9 d	184.0	1.7%	0.00%
				8.040 d	364.5	81.2%	-0.24%
				3.204 d	667.7	101.3%	0.00%
				20.8 h	529.9	87.0%	0.00%

Bold face if over 0.5% difference.

Table 4. (Continued) Comparison of Proposed Relevant Radionuclide List and Task Leader Paper TL-2/10
DeGeer/Lewis Fission List

Table 1

	6.57 h	1260.4	28.90%		6.57 h	1260.4	28.9%		Differences (%)
Iodine-135	13.16 d	1048.1	80.00%	Cesium-136	13.16 d	818.5	99.7%	0.00%	
* Cesium-136	30.07 y	661.7 D	85.10%	Cesium-137	30.17 y	661.7	85.2%	-0.33%	0.00%
* Cesium-137	12.752 d	537.3	24.39%	Barium-140	12.75 d	537.3	24.4%	0.02%	-0.04%
* Barium-140	1.6781 d	1596.2	95.40%	Lanthanum-140	1.678 d	1596.2	95.4%	0.01%	0.00%
* Lanthanum-140	32.501 d	145.4	48.20%	Lanthanum-141	3.9 h	1354.5	1.6%	0.00%	0.00%
* Cerium-141	33.039 h	293.3	42.80%	Cerium-141	32.5 d	145.4	48.2%	-0.25%	0.00%
* Cerium-143	284.893 d	133.5	11.09%	Cerium-143	1.38 d	293.3	42.8%	0.10%	0.00%
* Cerium-144	10.98 d	531	13.10%	Cerium-144	284.6 d	133.5	11.1%	0.00%	-0.09%
* Neodymium-147	53.08 h	286	3.10%	Praseodymium-145	6.0 h	748.2	40.0%	0.00%	
Promethium-149	28.4 h	340.1	22.50%	Neodymium-147	10.98 d	91.1	27.9%	-0.02%	0.00%
Promethium-151	46.27 h	103.2	31.40%	Promethium-149	2.212 d	286.0	3.1%	0.03%	0.00%
* Samarium-153	4.7611 y	105.3	21.20%	Promethium-150	2.7 h	333.9	74.0%	-0.06%	0.00%
* Samarium-155	9.4 h	203.8	20.80%	Promethium-151	1.183 d	340.1	22.5%	1.70%	0.00%
* Samarium-156	15.19 d	1153.7	6.80%	Samarium-153	1.929 d	103.2	31.4%	0.00%	0.00%
* Europium-157	15.18 h	370.5	11.00%	Europium-155	4.68 y	86.5	32.8%	-0.07%	
				Samarium-156	9.4 h	811.8	9.7%	0.33%	
				Europium-156	15.2 d	87.6	24.0%		
				Europium-157	15.13 h	413.0	23.0%		
				Gadolinium-159	18.6 h	363.6	11.0%		
				Terbium-161	6.88 d	74.5	12.2%		
				Xenon-133	5.24 d	81.0	36.6%		
				Xenon-133m	2.19 d	232.9	10.3%		
				Cesium-134	2.065 y	604.7	97.6%		
				Xenon-135	9.1	249.8	90.5%		

* Detected by Swedish Surveillance Network

Using different gamma ray and branching ratio

Using an isotope unique to the listing

Bold face if over 0.5% difference.