NRL

National Radiation Laboratory

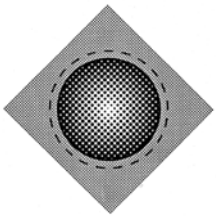
The CTBT verification significance of particulate radionuclides detected by the International Monitoring System

K M Matthews

2005/1
NRL Report

The CTBT verification significance of particulate radionuclides detected by the International Monitoring System

K M Matthews



NRL

National Radiation Laboratory

National Radiation Laboratory
Ministry of Health
PO Box 25099, Christchurch, New Zealand

ISSN 1174-6483 (Print)
ISSN 1179-2817 (Online)

National Radiation Laboratory Web site: http://www.nrl.moh.govt.nz

Contents

Abstract

The role of radionuclide monitoring in verification of Comprehensive Nuclear-Test-Ban Treaty (CTBT) verification and the associated concept of Relevant Radionuclides are reviewed. Three groups of radionuclides produced in nuclear weapon detonations are identified: fuel-, fission- and activation-products. The relative likelihoods of detection, by the International Monitoring System (IMS), of radionuclides within each group are assessed in order to provide a means of assisting National Data Centres (NDC) in their verification efforts. Accordingly, a distinction is made between CTBT verification "relevance" and "significance". Detection Probability Indices (DPI) for all relevant fission-products are calculated on the basis of fission yield, gamma emission intensity, decay rate and detector efficiency. Significant fission-products are defined as those within the group of 10 nuclides with highest DPI values in the time period of 3 – 20 days after detonation of weapons involving 6 different fission scenarios: normal or high-energy fission of ^{235}U, ^{238}U, ^{239}Pu. Relative activation- and fuel-product significance ratings are assessed by comparison with historical precedents, particularly in relation to fission-product detections. It is concluded that the significant fuel-products are ^{237}U, ^{239}Np and ^{241}Am; and significant fission products are ^{95}Nb, ^{95}Zr, ^{97}Zr, ^{99}Mo, ^{103}Ru, ^{105}Rh, ^{115}Cd, ^{126}Sb, ^{127}Sb, ^{131}I, ^{131m}Te, ^{132}Te, ^{133}I, ^{136}Cs, ^{140}Ba, ^{140}La, ^{141}Ce, ^{143}Ce, ^{144}Ce, and ^{147}Nd; while there are no significant activation products. This selection is validated by comparison with historical monitoring records; and these nuclides proposed to be the primary nuclides necessary for NDC screening of detections reported by the IMS.

1 Introduction

Radioactivity monitoring for the purposes of Comprehensive Nuclear-Test-Ban Treaty (CTBT) compliance verification comprises particulate and noble-gas monitoring intended primarily for detection of above- and below-ground weapon tests, respectively. This report focuses on particulate monitoring and considers the relative significance of the various radionuclides with a view to formulating operating procedures for National Data Centres (NDC).

Treaty verification is uniquely concerned with detection and identification of suspicious events. Historically, national radioactivity monitoring programmes maintained by some States have had additional functions, particularly concerning investigation of the nature of weapons. The radionuclides released during a nuclear detonation provide information on weapon design but this is not the primary concern of the Treaty compliance-verification effort. Certainly, after detection and identification of a clandestine nuclear weapon test detailed scientific analysis would be likely to follow, at least in some States, in order to gain information on the perpetrator's weapon programme but this is beyond the scope of verification. The possible significance of radionuclides in weapon design studies are not considered here.

1.1 Background information

Compliance verification monitoring for the CTBT involves four technologies: seismic, hydro-acoustic and infrasound (collectively termed the "seismo-acoustic" technologies) and radioactivity monitoring (the "radionuclide" technology). The seismo-acoustic technologies are intended to provide real-time monitoring in the underground, underwater, and atmospheric environments and to give accurate location details. In contrast, the radionuclide technology would have a relatively delayed response because detection is dependent on atmospheric transport of radioactive materials and their subsequent collection and analysis, but it is the only technology capable of providing forensic proof of the "nuclear" nature of any suspicious event. The four technologies thus work in concert; with the seismo-acoustic group providing immediate warning and location but without unambiguous identification, and the radionuclide monitoring providing forensic proof if key radionuclides are detected. Although radionuclide detection cannot be assured, the network has been designed to provide a minimum 90% probability of detecting any aboveground nuclear-weapon detonation with an explosive yield equivalent of 1 kiloton TNT, within 10 days. This design was based on requirements for particulate monitoring, where there was already much global experience in monitoring airborne radioactive materials, but it was recognised that evasive testing scenarios might involve underground tests which would not be detectable by this means unless there was significant venting. The noble-gas monitoring capability was accordingly added to the network, with equipment to be installed at half of the 80 particulate monitoring stations to provide the possibility of detection of vented xenon isotopes in the atmosphere.

Prime responsibility for CTBT verification monitoring rests with the Treaty's State Parties whose NDCs provide national advice concerning detection of possible breaches. The verification system includes the International Monitoring System (IMS) which comprises some 196 globally distributed monitoring stations - 50 seismic, 11 hydroacoustic, 55 infrasound, and 80 radionuclide stations – plus a further 120 auxiliary seismic stations under national control. Data from the IMS stations are transmitted to the International Data Centre (IDC) in Vienna where processing and interactive review functions are performed and a range of "products" prepared for distribution to each NDC which requests them. The IDC is precluded from offering opinions or judgements as to the possible significance of any detected event because this is the domain of the State Parties themselves. It is therefore important that NDCs have procedures in place for interpretation of IDC products such that they are able to advise their National Authorities in a reliably consistent manner. This report provides information to support procedures applied to interpretation of IDC products pertaining to particulate radionuclide monitoring.

1.2 Particulate radionuclide analysis and the IDC

The atmosphere contains a range of naturally occurring radionuclides arising from the emanation of radon gas from the soil and interactions of cosmic radiation with elements in the environment. Any non-natural, or "anthropogenic", radioactive materials released to the atmosphere must be detected and identified within this "background" of natural radioactivity. For particulate radionuclide monitoring this is achieved by high-resolution gamma-spectroscopic analysis of materials collected from the atmosphere. The IMS involves 24-hour sample collection by filtration of air at flow-rates of 500 m^3/h or more; a 24-hour "decay" period during which the air filter is set aside to allow short-lived natural radionuclides, which might otherwise render anthropogenic radionuclides also present on the filter undetectable, to undergo their normal process of radioactive decay; followed by a 24-hour analysis during which the gamma spectrum of the filter is acquired. All of this takes place at the monitoring station before the spectral data are transmitted via satellite to the IDC. The IDC receives a number of spectra from each station every day – an initial quality-control check spectrum to ensure that the equipment is working properly, followed by 11 "preliminary" sample spectra at two-hour intervals leading up to the final "full-sample" spectrum after 24 hours of analysis time. The quality control spectrum is used for various automatic checks, and a selection of preliminary sample spectra (particularly the 4-hour and 10-hour spectra) is processed automatically for use in diagnostic routines associated with identification of certain radionuclides (notably ^{99m}Tc), but most attention is focused on the full-sample spectrum which is processed automatically and reviewed interactively. Because of their volume, spectral data must be processed automatically by special software but, as with all automated routines, this unavoidably introduces the possibility of errors – through failure to detect radionuclides which are present, false reporting of some which are not present, or misidentifying detected radionuclides. Interactive review of the full-sample processing results is intended to correct any such errors. After processing and review, the IDC provides two separate products to requesting NDCs: the Automated Radionuclide Report (ARR) and the Reviewed Radionuclide Report (RRR). The ARR becomes available almost immediately after receipt of the raw spectral data at the IDC – processing is completed within seconds and the ARR despatched

automatically to subscribers and made available on the IDC secure website. Although results contained in the ARR should only be regarded as provisional, its distribution provides possible early warnings. For the RRR, IDC operating practice requires its distribution within 24 hours of data receipt, although before Treaty entry into force (EIF) the IDC operates for normal business hours five days per week, so the RRR may be delayed for up to 3 days.

1.3 Radionuclide screening

There are many radionuclides present in the environment which might be detected on any day at any station. Naturally occurring and gamma-emitting radionuclides present in the atmosphere include members of the natural uranium and thorium decay series arising from decay of ^{222}Rn and ^{220}Rn to ^{214}Pb, ^{212}Pb, ^{210}Pb, ^{214}Bi, ^{212}Bi, and ^{208}Tl; terrestrial radionuclides contained in resuspended dust including other ^{238}U and ^{232}Th series nuclides together with ^{40}K and other primordial nuclides; and cosmogenic nuclides such as ^{7}Be, ^{22}Na and ^{24}Na. Particularly problematic from a Treaty-verification perspective is ^{212}Pb which, together with its radioactive progeny, produces complex gamma spectra and does not fully decay away during the 24-hour decay period (half-life 10.6 hours). In gamma spectra from normal IMS air filters the IDC has found that, on average, 90% of the spectral lines recorded are associated with ^{212}Pb and its progeny.

Against this natural background, treaty verification requires that any anthropogenic radionuclides present in the atmosphere must also be detected and identified. There are many possible such anthropogenic radionuclides arising from a number of sources: hospitals and medical research facilities where radiopharmaceuticals are used, dispensed, or disposed of; university or other research institutions where radioactive tracers are used; industries where radioactive materials are commonly used and might be released to the environment accidentally; nuclear power reactors which release noble-gases; nuclear fuel reprocessing plants; the legacy from historic nuclear weapon tests which distributed fallout globally; contaminated sites, particularly in the former USSR; and the Chernobyl nuclear reactor accident which distributed ^{137}Cs and other products throughout the Northern Hemisphere, with this ^{137}Cs still being detected almost daily at some stations (notably station SEP63 in Sweden) through resuspension. In addition to the natural background radioactivity there is, therefore, also an anthropogenic background, and IMS experience already indicates that radionuclides from some of these sources are detected on a daily basis.

In addition to these natural and anthropogenic backgrounds, the detector systems themselves provide additional radionuclide detections through neutron-activation reactions of their component elements with neutrons derived from cosmic radiation. One of these activation products in particular, ^{75m}Ge, is difficult to distinguish from the fission-product ^{99m}Tc.

Nuclear weapons themselves all include a component of nuclear fission in their operating design and this produces a wide range of anthropogenic fission products which are radioactive. In addition, an intense neutron flux induces radioactivity in materials by nuclear excitation, producing neutron-activation products. Furthermore,

the range of fission and activation products varies with weapon type, design and detonation location.

The application of atmospheric radioactivity monitoring in CTBT verification has three roles: detection, identification, and location of nuclear weapon tests. As indicated earlier, location is more the province of the seismo-acoustic technologies; but the detection and identification roles largely rest on the abilities of the radionuclide technology to detect and identify key radionuclides within the complex natural and anthropogenic background radioactivity fields which contribute to every analysis performed. The concept of "screening" of detections in terms of their relevance to CTBT verification is therefore of paramount importance. To this end, the concept of "relevant radionuclides" was introduced as a basis for such screening.

1.4 Relevant radionuclides

Lars-Erik De Geer conducted a thorough review[1] of all possible radionuclides produced in nuclear weapon detonations and defined a list of "Relevant Radionuclides" which the Treaty State Parties accepted as being the working basis of radionuclide event reports for the IDC. The review considered three general groups of radionuclides classified according to their origin as: nuclear-fission products; neutron-activation products; and nuclides arising as weapon fuel residues or activation products, termed here "fuel products". A subset of relevant radionuclides, from the 3500 radioactive nuclides known altogether, was created by applying criteria as follows.

1. **Activity considerations**

   The nuclide half-life must be between 6 hours and 1000 years. Shorter half-lives would result in the nuclide decaying away before analysis, while longer half-lives would mean the rate of decay was too low for there to be a reasonable chance of detection (decay rate, or "activity", is inversely proportional to half-life).

2. **Detectability considerations**

   The nuclide must emit gamma radiation (to enable detection by the IMS gamma-spectroscopic systems); with a primary gamma energy greater than 50 keV (the high-resolution detectors employed in the IMS are relatively insensitive to lower-energy gamma radiation); and with a primary gamma intensity (fraction of decay events which produce the gamma radiation) greater than 0.1%.

3. **Production considerations**

   Fission-product yields from at least one scenario involving ^{235}U, ^{238}U or ^{239}Pu in either simple fission or thermonuclear devices must be greater than 0.1%. Although fission products are obviously of prime interest in compliance verification, neutron-activation products were included because many are produced and some have been detected historically. Requirements for

inclusion of products arising from non-fuel materials were that the production cross-section must be greater than 0.1 barn; environmental target elements must have natural abundances greater than 0.1%; and the product of cross-section and abundance must be greater than 100.

Additionally, some fuel-products were included in the list, along with products arising from diagnostic neutron-flux detectors in the devices themselves, but gases were excluded from this list pertaining to particulates (although xenon isotopes are relevant according to the above criteria).

Based on these requirements, a relevant nuclides list was compiled with 92 radionuclides: 4 fuel products, 47 fission products, and 41 neutron-activation products as listed in Table 1.

Application of the "relevant nuclides list" in IDC operations is highlighted by the five-level spectrum categorisation scheme which is applied as an event screening mechanism. Full-sample spectra interactively reviewed by the IDC are assigned a "categorisation level" according to their content, as follows:

Level 1: Spectrum contains only naturally occurring radionuclides, at normal atmospheric concentrations.

Level 2: Spectrum contains naturally occurring radionuclides but with one or more of them being at abnormal concentrations; or the spectrum contains anthropogenic radionuclides which are not on the relevant nuclides list.

Level 3: Spectrum contains one or more relevant anthropogenic nuclides which are commonly detected at the station and are at normal atmospheric concentrations for that station.

Level 4: Spectrum contains one relevant anthropogenic nuclide which either is not commonly detected at the station, or is commonly detected but is now at an abnormally high atmospheric concentration.

Level 5: Spectrum contains more than one relevant anthropogenic nuclide, including at least one relevant fission product.

The concept of nuclide relevance thus plays a key role in IDC operations and in event screening mechanisms applied there.

1.5 Relevance versus Significance

The relevant nuclide list is deliberately conservative in that it includes a wide range of nuclides which could conceivably be of nuclear-weapon origin. In compiling the list, however, no consideration was given to the relative likelihood of detection of the chosen nuclides. The list was intended only to sort the "possible" nuclides from the "impossibles", with no ranking according to importance, and it is applied at the IDC as if all relevant radionuclides have equal probability of detection, which clearly cannot be the case in reality.

The complexity of gamma spectra and the statistical fluctuations inherent in radioactivity counting data result in uncertainties in analysis results, particularly where small spectral structures are interpreted as being "real" and associated with anthropogenic radionuclides. The 92 relevant nuclides listed in Table 1 are associated with their 92 principle gamma emissions plus secondary emissions which collectively represent several hundred possible gamma-spectral lines. If a spectral structure falls within an energy band of typically ±0.5 keV width surrounding one of these energies it may be identified as a relevant nuclide. There is thus a considerable chance of a relevant-nuclide association being declared spuriously, and an unavoidable risk that a false alert may arise. The IDC naturally assumes a conservative approach such that a spectral structure may be associated with an anthropogenic nuclide fitting its energy profile unless there are sound objective reasons for rejecting such identity. The IDC thus deliberately "errs on the safe side" rather than risk failure to notify NDCs of relevant-nuclide detections. For the NDCs, this requires daily scrutiny of reports of relevant-nuclide detections which might, or might not, be real. As IMS/IDC experience develops, and better spectral analysis and nuclide identification routines become available, the incidence of such false alarms should decrease, but NDCs will always face the decision as to whether or not the detection of any particular nuclide(s) indicates a Treaty breach.

A system of prioritising radionuclides according to their verification significance would therefore be helpful because whether or not detection of relevant radionuclides indicates occurrence of a suspicious event can best be judged on the basis of which particular nuclides have been detected. Clearly, not all of the 92 nuclides on the relevant list can have equal probabilities of detection – and nuclides with relatively low probability would not be detected in the absence of those with higher detection probability.

This report therefore addresses the issue of nuclide "significance", or the relative likelihood of detection, with the aim of providing an additional screening method which an NDC could apply to screen out unimportant or spurious events from those reported according to nuclide relevance only. A list of "Significant Radionuclides" is derived as a subset of the relevant radionuclides. This secondary classification of radionuclides is based mainly on the concept of "Detection Probability Indices" as described in succeeding Sections.

All CTBT-relevant radionuclides were considered in the present study, with grouping according to three general types: fuel-, fission- and activation-products.

Table 1: The list of CTBT relevant particulate radionuclides[1].

Fission products	Activation products	Fuel products
Strontium-91	Sodium-24	Radium-224
Yttrium-91	Potassium-42	Uranium-237
Yttrium-93	Scandium-46	Neptunium-239
Zirconium-95	Scandium-47	Americium-241
Niobium-95	Chromium-51	
Zirconium-97	Manganese-54	
Molybdenum-99	Cobalt-57	
Technetium-99m	Cobalt-58	
Ruthenium-103	Iron-59	
Rhodium-105	Cobalt-60	
Ruthenium-106	Copper-64	
Palladium-109	Zinc-65	
Silver-111	Zinc-69m	
Palladium-112	Gallium-72	
Cadmium-115m	Arsenic-74	
Cadmium-115	Arsenic-76	
Tin-123	Rubidium-84	
Tin-125	Rubidium-86	
Antimony-125	Yttrium-88	
Tellurium-125m	Zirconium-89	
Antimony-126	Rhodium-102	
Antimony-127	Silver-106m	
Tellurium-127m	Silver-108m	
Tellurium-127	Silver-110m	
Antimony-128	Antimony-120	
Tellurium-129m	Antimony-122	
Iodine-130	Antimony-124	
Tellurium-131m	Caesium-132	
Iodine-131	Barium-133	
Tellurium-132	Caesium-134	
Iodine-133	Europium-152m	
Iodine-135	Europium-152	
Caesium-136	Thulium-168	
Caesium-137	Thulium-170	
Barium-140	Tungsten-187	
Lanthanum-140	Iridium-190	
Cerium-141	Iridium-192	
Cerium-143	Gold-196	
Cerium-144	Gold-196m	
Neodymium-147	Gold-198	
Promethium-149	Lead-203	
Promethium-151		
Samarium-153		
Europium-155		
Samarium-156		
Europium-156		
Europium-157		

2 Fuel products

Radioactive debris arising from the nuclear fuel itself are of major significance in Treaty verification because they have the potential to be released with high levels of radioactivity. Weapon fuels may contain the following[1]:

Tritium	^{3}H
Lithium deuteride	^{6}LiD, ^{7}LiD
Weapons-grade uranium	^{234}U (<1%), ^{235}U (90%), ^{238}U
Depleted uranium	^{235}U, ^{238}U (>99%)
Weapons-grade plutonium	^{239}Pu (93.5%), ^{240}Pu (6%), ^{241}Pu (0.5%)
Uranium-233 (unlikely fuel)	^{233}U and ^{232}U

Decay products of the above, present in the fuel to a degree depending on its age, include: ^{241}Am, ^{236}U, ^{237}Np, ^{233}U, ^{229}Th, and ^{224}Ra. Some original "unburned" fuel residue or its decay products may be released into the atmosphere during detonation. Of these residues, the relevant-nuclide list includes ^{241}Am and ^{224}Ra, with the latter being associated only with weapons based on ^{233}U.

During thermonuclear detonation, the intense neutron flux causes multiple neutron-capture reactions, but the resulting high-mass products decay rapidly. For ^{238}U activation, the products of highest abundance[1] are ^{239}U, ^{240}U and ^{241}U which respectively decay to ^{239}Pu (through ^{239}Np), ^{240}Pu and ^{241}Pu. High-energy neutrons (up to 14.1 MeV) cause (n, 2n) reactions which, with ^{238}U, produce ^{237}U in high abundance and which decays (half-life 6.7d) to ^{237}Np. Fission weapons do not produce sufficiently high neutron flux for multiple capture to occur, but one-step capture produces ^{239}U, which decays through ^{239}Np to ^{239}Pu. Plutonium in the core is also activated to ^{240}Pu and ^{241}Pu, which decay respectively to ^{236}U and ^{241}Am. Of these reaction products, the relevant-nuclide list includes ^{239}Np and ^{237}U as being detectable by the IMS (the $^{239}Np/^{237}U$ ratio for thermonuclear and fission devices is about 1 and above 30, respectively[1]).

The fuel-products therefore contribute ^{241}Am, ^{224}Ra, ^{239}Np and ^{237}U to the list of relevant radionuclides (Table 1). Of these, ^{224}Ra is of low significance because construction of weapons fuelled by ^{233}U is unlikely – it is considered by De Geer[1] to be "an impractical weapons fuel". The remaining three nuclides were detected[2] as prominent features of gamma spectra acquired through analysis of particulates collected after Chinese aboveground weapon tests of 1975 - 1977, and so are regarded here as "significant" for Treaty verification.

Conclusion: significant fuel-product radionuclides are: **^{241}Am**, **^{239}Np** and **^{237}U**.

3 Fission products

All nuclear weapon types, whether pure "fission" or "fusion", include a component of nuclear fission in their mode of operation. Radioactive fission products are therefore of prime importance in detecting such devices after detonation. The list of relevant nuclides includes 47 such products. The detectability of a fission product depends on the factors described below.

3.1 Production yield

Fission yield is recorded as the percentage of fission events which produce the given nuclide. Fission-yield data provided in the De Geer report[1] for relevant radionuclides are listed in Table 2 for six weapon fission scenarios: normal ^{235}U, ^{239}Pu or ^{238}U fission ($^{235}U_f$, $^{239}Pu_f$, $^{238}U_f$ respectively), and fission in thermonuclear weapons where high-energy neutrons are involved ($^{235}U_{HE}$, $^{238}U_{HE}$, $^{239}Pu_{HE}$ respectively). The relevant-nuclide list included nuclides for which at least one of the fission yields was greater than or equal to 0.1%.

The higher the level of production of a fission product, and hence the larger the amount released to the atmosphere, the greater its chance of detection (within the limitations caused by other factors below).

3.2 Gamma emission characteristics

The gamma emission characteristics of any nuclide obviously influence its detectability by the gamma-spectrometric systems employed by the IMS. The three factors of prime importance in this context are: (i) the specific activity (rate of decay of a unit mass) of the nuclide, (ii) the energy of the principal gamma radiation, and (iii) its emission intensity (percentage of nuclear decay events which produce the gamma radiation).

Specific activity is a measure of the nuclear decay rate. The faster the decay, the more radiation is produced, so the greater the chance of detection. As determined by the basic law of decay, the "decay constant", λ, is inversely proportional to half-life ($\lambda = Ln2/T_{1/2}$). The likelihood of detection in the present context is therefore also inversely proportional to half-life.

The energy of the gamma radiation is linked to detectability through the detection-efficiency characteristics of the HpGe detectors employed in the IMS systems, as described in Fig. 1 which shows efficiency curves constructed using the efficiency calibration equations provided with every RRR from each of the 36 stations in operation in June 2005, based on the energies of the primary gamma lines for relevant radionuclides as given in Table 2. The detectability of any radionuclide is proportional to the detection efficiency applying at the energy of its primary gamma emission. Gamma emission intensity also obviously contributes to detection likelihood – the higher the intensity, the greater the chance of detection.

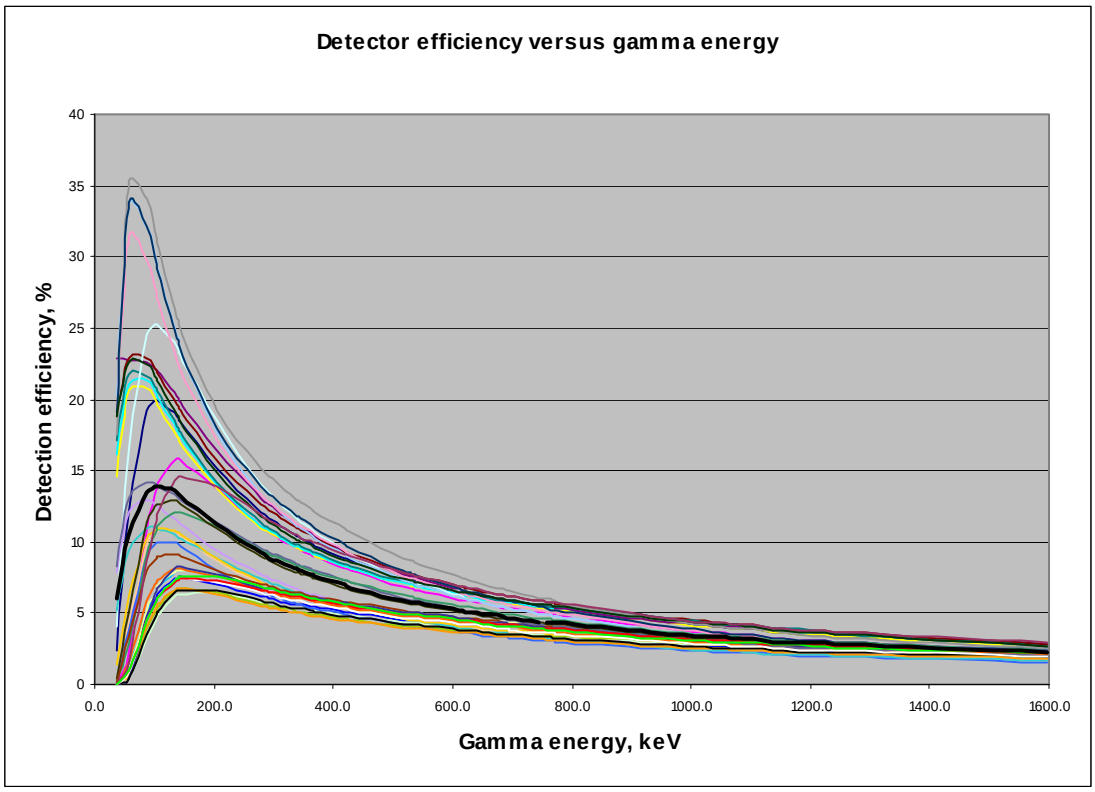
Based on the gamma emission characteristics, detection likelihood is thus proportional to detector efficiency and gamma intensity, and inversely proportional to half-life.

Table 2: Data for CTBT-relevant fission products[1]: half-life, primary gamma energy and intensity, and fission yields arising from normal and high-energy fission.

Fission product	Half-life, d	γ energy keV	intensity %	Fission yield, %					
				$^{235}U_f$	$^{238}U_f$	$^{239}Pu_f$	$^{235}U_{HE}$	$^{238}U_{HE}$	$^{239}Pu_{HE}$
^{91}Sr	0.40	1024.3	33.4	5.73	4.04	2.51	4.81	3.87	2.22
^{91}Y	58.51	1204.8	0.3	5.73	4.04	2.52	4.82	3.87	2.24
^{93}Y	0.42	266.9	7.3	6.25	4.91	3.82	5.19	4.53	3.22
^{95}Zr	64.02	756.7	54.5	6.43	5.14	4.67	5.17	4.89	3.92
^{95}Nb	34.98	765.8	99.8	6.43	5.14	4.67	5.18	4.89	3.93
^{97}Zr	0.70	743.4	93.1	6.00	5.56	5.27	5.14	5.28	4.40
^{99}Mo	2.75	140.5*	89.4	5.94	6.17	5.98	5.14	5.71	4.75
^{99m}Tc	0.25	140.5	89.1	5.23	5.43	5.26	4.52	5.02	4.18
^{103}Ru	39.26	497.1	90.9	3.24	6.28	6.83	3.21	4.62	5.21
^{105}Rh	1.47	319.1	19.2	1.20	4.05	5.36	1.87	3.22	4.27
^{106}Ru	373.59	621.9	9.9	0.53	2.49	4.36	1.61	2.45	3.54
^{109}Pd	0.57	88.0	3.6	0.08	0.25	1.04	1.17	1.12	2.48
^{111}Ag	7.45	342.1	6.7	0.04	0.07	0.36	1.08	0.99	1.54
^{112}Pd	0.88	617.5*	43.0	0.04	0.06	0.19	1.08	1.03	1.38
^{115m}Cd	44.60	933.8	2.0	0.00	0.00	0.01	0.46	0.07	0.12
^{115}Cd	2.23	336.2	45.9	0.03	0.03	0.07	0.64	0.79	1.16
^{123}Sn	129.20	1088.6	0.6	0.00	0.00	0.02	0.06	0.01	0.27
^{125}Sn	9.64	1067.1	10.0	0.04	0.03	0.12	0.92	0.64	1.18
^{125}Sb	1007.40	427.9	29.6	0.07	0.05	0.18	1.46	1.20	1.95
^{125m}Te	57.40	109.3	0.3	0.02	0.01	0.04	0.33	0.27	0.44
^{126}Sb	12.46	695.0	99.6	0.01	0.01	0.04	0.34	0.19	0.62
^{127}Sb	3.85	685.7	36.8	0.31	0.14	0.50	2.16	1.49	2.14
^{127m}Te	109.00	57.6	0.5	0.05	0.02	0.08	0.36	0.24	0.39
^{127}Te	0.39	418.0	1.0	0.31	0.14	0.50	2.17	1.49	2.19
^{128}Sb	0.38	743.2	100.0	0.01	0.00	0.04	0.42	0.14	0.68
^{129m}Te	33.60	695.9	3.2	0.14	0.17	0.24	0.71	0.36	0.93
^{130}I	0.52	536.1	99.0	0.00	0.00	0.00	0.03	0.00	0.13
^{131m}Te	1.25	773.7	49.9	0.43	0.26	0.92	1.34	0.42	1.88
^{131}I	8.02	364.5	81.7	3.22	3.29	3.88	4.10	3.99	4.35
^{132}Te	3.20	772.6*	75.6	4.66	5.13	5.15	4.09	4.65	3.30
^{133}I	0.87	529.9	87.0	6.72	6.76	6.91	5.36	6.00	4.48
^{135}I	0.27	1260.4	28.9	6.30	6.94	6.08	4.22	5.50	3.96
^{136}Cs	13.16	1048.1	80.0	0.01	0.00	0.12	0.23	0.02	0.75
^{137}Cs	10983.1	661.6*	85.1	6.22	6.05	6.58	4.93	5.15	4.45
^{140}Ba	12.75	537.3	24.4	5.98	5.82	5.32	4.50	4.61	3.70
^{140}La	1.68	1596.2	95.4	5.98	5.82	5.33	4.53	4.61	3.84
^{141}Ce	32.50	145.4	48.2	5.95	5.34	5.15	4.49	4.38	3.56
^{143}Ce	1.38	293.3	42.8	5.73	4.62	4.34	3.82	3.91	2.80
^{144}Ce	284.89	133.5	11.1	5.27	4.55	3.67	3.17	3.72	3.96
^{147}Nd	10.98	531.0	13.1	2.14	2.59	1.99	1.62	2.09	1.71
^{149}Pm	2.21	286.0	3.1	1.04	1.63	1.24	0.81	1.46	1.06
^{151}Pm	1.18	340.1	22.5	0.41	0.80	0.78	0.36	0.80	0.73
^{153}Sm	1.93	103.2	31.4	0.17	0.41	0.43	0.20	0.39	0.46
^{155}Eu	1738.99	105.3	21.2	0.04	0.14	0.21	0.08	0.16	0.23
^{156}Sm	0.39	203.8	20.8	0.02	0.08	0.15	0.05	0.11	0.17
^{156}Eu	15.19	1153.7	6.8	0.02	0.08	0.15	0.06	0.11	0.21
^{157}Eu	0.63	370.5	11.0	0.01	0.04	0.11	0.04	0.08	0.11

*: radiation from short-lived decay product

Fig. 1: Efficiency calibration curves for all 35 detectors employed at operational IMS stations, as at June 2005. The central heavy dark line represents the mean.



3.3 Detection Probability Index

As discussed above, the likelihood of detection of fission-product radionuclide F is proportional to the following factors:

Fission yield (%): F_y

Gamma intensity (%) for principal line of energy e: $F_{e,i}$

Detector efficiency (%) at gamma energy e: E_e

Decay rate (inversely proportional to half-life, $T_{1/2,F}$) $1/T_{1/2,F}$

A measure of the relative probability of detection can be obtained by multiplying these factors to give a "Detection Probability Index", DPI, which can be used as the basis for comparing radionuclide detection likelihoods:

$$(DPI)_F = \frac{F_y.F_{e,i}.E_e}{T_{1/2,F}}$$

Detection probability varies with time, as the radionuclide decays, so DPI at time t is as follows:

$$(DPI)_{F,t} = \frac{D_t.F_v.F_{e,i}.E_e}{T_{1/2,F}} \qquad \textbf{Equation 1}$$

where D_t is the activity fraction remaining at time t, as calculated from the decay equation $A/A_o = e^{-\lambda t}$.

The DPI values were calculated for all relevant fission products, at times 1, 3, 5, 10, 20, 100, and 300 days after production, using the mean network detector efficiency curve (Fig.1), as a means of comparing their relative likelihoods of detection. The DPI is, by definition, intended for comparison of relative values only – it is a simple means of comparison of radionuclides in terms of their detectability by the IMS. The absolute values have no meaning. The time-frame of primary interest is 3 – 20 days as this is the most likely period during which fission products would be detected by the IMS, and the 100 and 300 day periods were considered only for long-term comparisons with historical data.

DPI values were calculated for all CTBT-relevant nuclides according to the above equation for $(DPI)_{F,t}$, for t = 1, 3, 5, 10, 20, 100, 300 d, and results for each time were ordered by descending DPI value so that nuclides with the highest relative detection likelihood would be shown at the top of the list. This was repeated for each of the fission scenarios. Results are shown in Figs 2 – 7.

A comparison of DPI values for each fission type is provided in Table 3 for the 3-day time-band (3 days after production). Here DPI values are also shown normalised to a value of 100 for the nuclide with the highest value (^{99}Mo in all cases). For the case of ^{235}U fission, for example, the 10th nuclide in the order, ^{140}Ba, has a normalised DPI value of only 5, meaning that ^{99}Mo would be 20 times more likely to be detected than ^{140}Ba. The relativity between detection likelihoods is depicted graphically in Figs 8 and 9, where it can be seen that relative values span six orders of magnitude.

It is inconceivable that fission products near the bottom of the order could be detected in the absence of those near the top if their source was a nuclear weapon detonation. The concept of "significance" here is related to this ranking. Nuclides near the top of the ranking order would have greater significance for verification purposes than those near the bottom, even though all are CTBT-relevant.

Nuclide significance obviously varies with time as the effects of variations in relative decay rate become apparent. This is illustrated in Figs 10 and 11 which respectively show trends in relative significance over 1-20 and 1-300 day periods for ^{235}U and ^{239}Pu fission, where relative significance is defined as the fractional DPI value (the DPI value expressed as a fraction of the total of all DPI values for the time-band). As can be seen in Figs 10 and 11, the longer-lived nuclides such as ^{137}Cs do not become significant until after considerable delays by which time the short-lived nuclides, which initially had greatest significance, have decayed away.

It is suggested that NDCs might apply this concept of significance in their verification considerations by focussing attention on those nuclides with most significance, ignoring those of lower significance. Nuclides of lower ranking might obviously be of interest following any Treaty violation, but they would not be detected in the

absence of those with higher rankings. In this context the following definition is applied.

> A **significant** fission-product radionuclide is one which appears within the group of 10 with highest DPI values at intervals of either 3, 5, 10, or 20 days after production in any of six processes: ^{235}U, ^{239}Pu or ^{238}U fission; ^{235}U, ^{239}Pu or ^{238}U high-energy fission.

The list of significant nuclides was compiled from results shown in Figs 2 - 7, as summarised in Table 4 below.

Clearly this definition and the calculation of DPI values provide relative information only on the fission-products most likely to be detected following any nuclear detonation. Inclusion of all the fission-products with highest DPI values in a "significant nuclides list", as shown in Table 4, would be sufficient for detecting and identifying a real event. It is not necessary that all have to be detected, or even that they must be detected in the order shown, for a real event to be indicated. The variable nature of weapon design, variations in detector characteristics, and uncertainties over environmental fractionation, make it impossible to predict accurately the relative amounts of fission products formed, and the DPI method is intended only to provide a means of distinguishing those most likely to be detected from the others, enabling a focus in the verification-monitoring effort.

3.3.1 Extraneous factors

The DPI model, as described by Equation 1, highlights major differences in nuclide detection likelihoods but is clearly simplistic. There are additional factors which could be incorporated into the model but its original purpose should remain in focus – namely that it was intended to determine the level of difference between nuclides in terms of their detectability and to highlight major differences. For the record, however, some of these additional factors and their possible effects are described below.

(a) **Environmental fractionation.** Fractionation through which nuclides may exhibit different chemical and physical behaviours in the atmosphere and possibly leading to selective dispersion or deposition, was not considered in this model. Over the short time-span of up to 20 days, any such effects would be likely to be small compared to differences in DPI values.

(b) **Collection efficiency.** It was assumed that all particulate forms of radioactivity would have equal probability of entrapment on the air filters.

(c) **Detector efficiency variations.** Variations in detector efficiency from the mean efficiency-energy relationship used here would influence relative DPI values. Recalculation of values using the extreme detector performances (the uppermost and lowest curves in Fig.1, referring to stations NZP46 and JPP38, respectively) showed, however, that the effects are minor compared to variations between DPI values. Slight changes in rankings of nuclides may occur, but without alteration of the above choice of significant nuclides.

(d) **Incorporation of MDA.** The minimum detectable activity (MDA) varies with energy due to the variation of the spectral background-continuum count-rate with energy. The background-continuum is due to the influence of cosmic background radiation and Compton events associated with detected radionuclides, mainly ^{212}Pb and its decay products. Background spectra generally display a continuum-curve shape similar to the energy-efficiency curve for the detector and, to a first and simplistic approximation, the continuum count-rate is proportional to detector efficiency (with discrepancy at low energies due to shielding). Background count rate is therefore a function of Ø.E, where Ø is the gamma radiation flux impinging on the detector (more or less uniform over the energy range under consideration here), and E is detector efficiency. The MDA at any point in the spectrum is proportional to √(background), and therefore proportional to √E. As detection probability is inversely proportional to MDA, a $\sqrt{E_e}$ term could have been included in the denominator of Equation 1, having the effect of reducing the numerator E_e term to $\sqrt{E_e}$. The effect of this modification to Equation 1 was tested by recalculating all DPI values incorporating the $\sqrt{E_e}$ term in Equation 1. It was found, as in (c) above that the effect was to slightly change the DPI rankings of some nuclides at some time intervals. The only change it introduced to the choice of the top-10 nuclides was that ^{131m}Te entered the significant category at the 3-day time interval (only) for high-energy ^{239}Pu fission. Because of the gross simplification involved, it was not considered worthwhile including the $\sqrt{E_e}$ term permanently in the model but in the interests of conservative reporting, ^{131m}Te was included in the list of significant nuclides (Table 4) even though its significance may be questionable (it has not been detected previously in nuclear-test monitoring[1]).

(e) **Effects of station background. ^{212}Pb.** There is a wide variability between atmospheric ^{212}Pb concentrations recorded at IMS stations, as exemplified by the pair of stations AUP06 and AUP08 which display up to three orders of magnitude between their respectively high and low ^{212}Pb levels. The background count-rate levels, and hence MDA values, would vary between these stations accordingly. The ^{212}Pb family includes nuclides with principal gamma energies across the IMS range (up to 2.6 MeV ^{208}Tl), however, with the result that the background Compton continuum associated with these nuclides is affected more or less uniformly across the spectral range. The relativity between nuclide detection likelihoods is therefore preserved.

Table 3: Relevant fission products ordered by DPI, 3 days after production, for each fission scenario (Nmlsd = normalised to ^{99}Mo)

Normal 235U fission			Normal 239Pu fission			Normal 238U fission			High-energy 235U fission			High-energy 239Pu fission			High-energy 238U fission		
Nuclide order	DPI: 3 days		Nuclide order	DPI: 3 days		Nuclide order	DPI: 3 days		Nuclide order	DPI: 3 days		Nuclide order	DPI: 3 days		Nuclide order	DPI: 3 days	
	Value	Nmlsd		Value	Nmlsd		Value	Nmlsd		Value	Nmlsd		Value	Nmlsd		Value	Nmlsd
Mo-99	1211.35	100	Mo-99	1219.51	100	Mo-99	1258.26	100	Mo-99	1048.21	100	Mo-99	968.68	100	Mo-99	1164.45	100
I-133	357.42	30	I-133	367.52	30	I-133	359.54	29	I-133	285.08	27	I-131	262.61	27	I-133	319.12	27
Ce-143	351.28	29	Te-132	272.57	22	Ce-143	283.23	23	I-131	247.52	24	I-133	238.28	25	Te-132	246.11	21
Te-132	246.64	20	Ce-143	266.07	22	Te-132	271.51	22	Ce-143	234.19	22	Te-132	174.66	18	I-131	240.88	21
La-140	219.63	18	I-131	234.24	19	La-140	213.75	17	Te-132	216.47	21	Ce-143	171.66	18	Ce-143	239.71	21
I-131	194.39	16	La-140	195.75	16	I-131	198.62	16	La-140	166.37	16	La-140	141.03	15	La-140	169.31	15
Zr-97	181.77	15	Zr-97	159.66	13	Zr-97	168.44	13	Zr-97	155.72	15	Zr-97	133.30	14	Zr-97	159.96	14
Ce-141	109.35	9	**Rh-105**	143.47	12	Rh-105	108.41	9	Ce-141	82.51	8	Rh-105	114.29	12	Rh-105	86.19	7
Nb-95	74.64	6	Ce-141	94.64	8	Ce-141	98.14	8	Nb-95	60.13	6	**Cd-115**	76.41	8	Ce-141	80.49	7
Ba-140	56.17	5	**Ru-103**	91.94	8	Ru-103	84.53	7	**Sb-127**	57.15	5	Ru-103	70.13	7	Ru-103	62.19	5
Ru-103	43.61	4	Nb-95	54.21	4	Nb-95	59.66	5	Rh-105	50.05	5	Ce-141	65.42	7	Nb-95	56.76	5
Rh-105	32.12	3	Ba-140	49.97	4	Ba-140	54.67	4	Te-131m	43.49	4	Te-131m	61.02	6	Cd-115	52.04	4
Zr-95	23.16	2	Sm-153	33.19	3	Sm-153	31.64	3	Ru-103	43.21	4	Sb-127	56.62	6	Ba-140	43.30	4
Te-131m	13.96	1	Te-131m	29.86	2	Pm-151	21.11	2	Ba-140	42.27	4	Nb-95	45.62	5	Sb-127	39.42	3
Sm-153	13.12	1	Pm-151	20.59	2	Zr-95	18.52	1	Cd-115	42.16	4	Sm-153	35.50	4	Sm-153	30.10	3
Nd-147	12.32	1	Zr-95	16.82	1	Nd-147	14.91	1	Pd-112	25.59	2	Ba-140	34.75	4	Pd-112	24.40	2
Pm-151	10.82	1	Sb-127	13.23	1	Te-131m	8.44	1	Zr-95	18.62	2	Pd-112	32.69	3	Pm-151	21.11	2
Sr-91	8.84	1	Nd-147	11.45	1	Pm-149	8.10	1	Sm-153	15.44	1	Sb-126	19.71	2	Zr-95	17.62	2
Sb-127	8.20	1	Pm-149	6.16	1	Tc-99m	6.32	1	Sb-126	10.81	1	Pm-151	19.27	2	Te-131m	13.63	1
Y-93	7.33	1	Tc-99m	6.13	1	Sr-91	6.23	0	Pm-151	9.50	1	Zr-95	14.12	1	Nd-147	12.03	1
Tc-99m	6.09	1	Cd-115	4.61	0	Y-93	5.76	0	Nd-147	9.32	1	Cs-136	12.73	1	Pm-149	7.26	1
Pm-149	5.17	0	Pd-112	4.50	0	Sb-127	3.70	0	Sr-91	7.42	1	Nd-147	9.84	1	Sb-126	6.04	1
Ce-144	2.74	0	Y-93	4.48	0	Ce-144	2.36	0	Y-93	6.09	1	Ag-111	8.41	1	Sr-91	5.97	1
Cd-115	1.98	0	Sr-91	3.87	0	Cd-115	1.98	0	Ag-111	5.90	1	Pd-109	5.53	1	Tc-99m	5.85	1
Pd-112	0.95	0	Pd-109	2.32	0	Pd-112	1.42	0	Tc-99m	5.26	1	Pm-149	5.27	1	Ag-111	5.41	0
I-135	0.84	0	Cs-136	2.04	0	I-135	0.93	0	Pm-149	4.03	0	Tc-99m	4.87	1	Y-93	5.31	0
Sb-126	0.32	0	Ag-111	1.97	0	Pd-109	0.56	0	Cs-136	3.90	0	Y-93	3.78	0	Pd-109	2.50	0
Cs-137	0.24	0	Ce-144	1.90	0	Ag-111	0.38	0	Pd-109	2.61	0	Sr-91	3.42	0	Ce-144	1.93	0
Ag-111	0.22	0	Sb-126	1.27	0	Ru-106	0.34	0	Sn-125	2.47	0	Sn-125	3.17	0	Sn-125	1.72	0
Pd-109	0.18	0	I-135	0.81	0	Sb-126	0.32	0	Sb-128	1.94	0	Sb-128	3.15	0	I-135	0.73	0
Cs-136	0.17	0	Ru-106	0.59	0	Sm-156	0.23	0	Ce-144	1.65	0	I-130	2.55	0	Sb-128	0.65	0
Sn-125	0.11	0	Eu-157	0.53	0	Cs-137	0.23	0	I-130	0.59	0	Ce-144	2.06	0	Eu-157	0.38	0
Y-91	0.08	0	Sm-156	0.44	0	Eu-157	0.19	0	I-135	0.56	0	Eu-157	0.53	0	Cs-136	0.34	0
Ru-106	0.07	0	Sn-125	0.32	0	Eu-156	0.09	0	Te-129m	0.30	0	I-135	0.53	0	Ru-106	0.33	0
Te-129m	0.06	0	Cs-137	0.25	0	Sn-125	0.08	0	Sb-125	0.29	0	Sm-156	0.49	0	Sm-156	0.32	0
Sm-156	0.06	0	Sb-128	0.19	0	Te-129m	0.07	0	Te-125m	0.23	0	Ru-106	0.48	0	Sb-125	0.24	0
Eu-157	0.05	0	Eu-156	0.18	0	Y-91	0.06	0	Ru-106	0.22	0	Sb-125	0.39	0	Cs-137	0.20	0
Sb-128	0.05	0	Te-129m	0.10	0	Eu-155	0.02	0	Eu-157	0.19	0	Te-129m	0.39	0	Te-125m	0.19	0
Te-127	0.03	0	Te-127	0.04	0	Te-127	0.01	0	Cs-137	0.19	0	Te-125m	0.30	0	Te-129m	0.15	0
Eu-156	0.02	0	Sb-125	0.04	0	Sb-125	0.01	0	Te-127	0.19	0	Eu-156	0.25	0	Eu-156	0.13	0
Sb-125	0.01	0	Y-91	0.04	0	Te-125m	0.01	0	Sm-156	0.15	0	Te-127	0.19	0	Te-127	0.13	0
Te-125m	0.01	0	Eu-155	0.04	0	Te-127m	0.00	0	Cd-115m	0.07	0	Cs-137	0.17	0	Y-91	0.05	0
Eu-155	0.01	0	Te-125m	0.03	0	Cd-115m	0.00	0	Eu-156	0.07	0	Eu-155	0.04	0	Eu-155	0.03	0
Te-127m	0.00	0	Te-127m	0.00	0	Sn-123	0.00	0	Y-91	0.07	0	Y-91	0.03	0	Te-127m	0.01	0
Sn-123	0.00	0	Cd-115m	0.00	0	Sb-128	0.00	0	Te-127m	0.02	0	Cd-115m	0.02	0	Cd-115m	0.01	0
Cd-115m	0.00	0	Sn-123	0.00	0	I-130	0.00	0	Eu-155	0.01	0	Te-127m	0.02	0	Sn-123	0.00	0
I-130	0.00	0	I-130	0.00	0	Cs-136	0.00	0	Sn-123	0.00	0	Sn-123	0.00	0	I-130	0.00	0

Table 4: Significant fission-products defined for each fission scenario, where ● indicates the nuclide is relevant to that type of fission event.

Nuclide	^{235}U fission	^{239}Pu fission	^{235}U H-E f	^{239}Pu H-E f	^{238}U fission	^{238}U H-E f
Mo-99	●	●	●	●	●	●
I-133	●	●	●	●	●	●
Ce-143	●	●	●	●	●	●
Te-132	●	●	●	●	●	●
La-140	●	●	●	●	●	●
I-131	●	●	●	●	●	●
Zr-97	●	●	●	●	●	●
Ce-141	●	●	●	●	●	●
Nb-95	●	●	●	●	●	●
Ba-140	●	●	●	●	●	●
Ru-103	●	●	●	●	●	●
Zr-95	●	●	●	●	●	●
Nd-147	●	●	●		●	●
Ce-144	●	●			●	
Rh-105		●		●	●	●
Sb-126			●	●		●
Sb-127			●	●		●
Cd-115				●		
Cs-136				●		
Te-131m*				●		

* see Section 3.3.1 d.

There are thus 20 significant fission-products altogether, with a subset of 12 which have highest DPI values irrespective of weapon type: ^{99}Mo, ^{133}I, ^{143}Ce, ^{132}Te, ^{140}La, ^{131}I, ^{97}Zr, ^{141}Ce, ^{95}Nb, ^{140}Ba, ^{103}Ru and ^{95}Zr.

Conclusion: significant fission-products are **^{99}Mo, ^{133}I, ^{143}Ce, ^{132}Te, ^{140}La, ^{131}I, ^{97}Zr, ^{141}Ce, ^{95}Nb, ^{140}Ba, ^{103}Ru, ^{95}Zr** although the set could be expanded conservatively to include **^{136}Cs, ^{115}Cd, ^{127}Sb, ^{126}Sb, ^{105}Rh, ^{144}Ce, ^{147}Nd and ^{131m}Te**.

The validation of this selection of nuclides is considered in Section 5.

Fig. 2: Rankings of Detection Probability Indices (DPI) for the case of normal ^{235}**U** fission in time-bands of 1, 3, 5, 10, 20, 100, 300 days, ordered by decreasing DPI value in each column. The "top 10" fission products in each time-band are highlighted, and time-bands of primary interest are highlighted in green. Individual nuclides are linked to indicate changes in their relative detection likelihood due to decay. Nuclides in the "top-10" group in the 3 – 20 day period are highlighted in red.

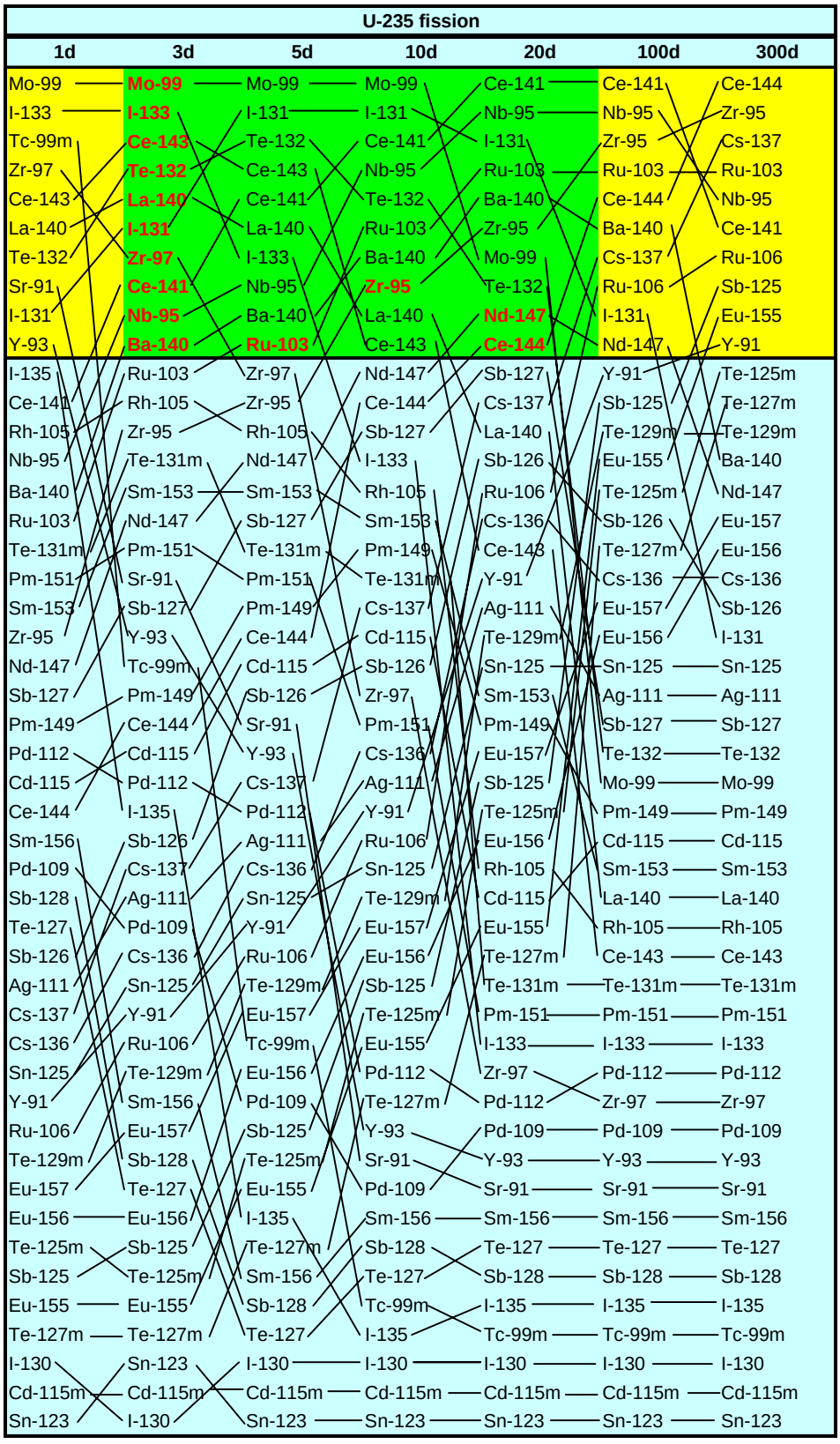


Fig. 3: Rankings of Detection Probability Indices (DPI) for the case of high-energy ^{235}**U** fission in time-bands of 1, 3, 5, 10, 20, 100, 300 days, ordered by decreasing DPI value in each column. The "top 10" fission products in each time-band are highlighted, and time-bands of primary interest are highlighted in green. Individual nuclides are linked to indicate changes in their relative detection likelihood due to decay. Nuclides in the "top-10" group in the 3 – 20 day period are highlighted in red.

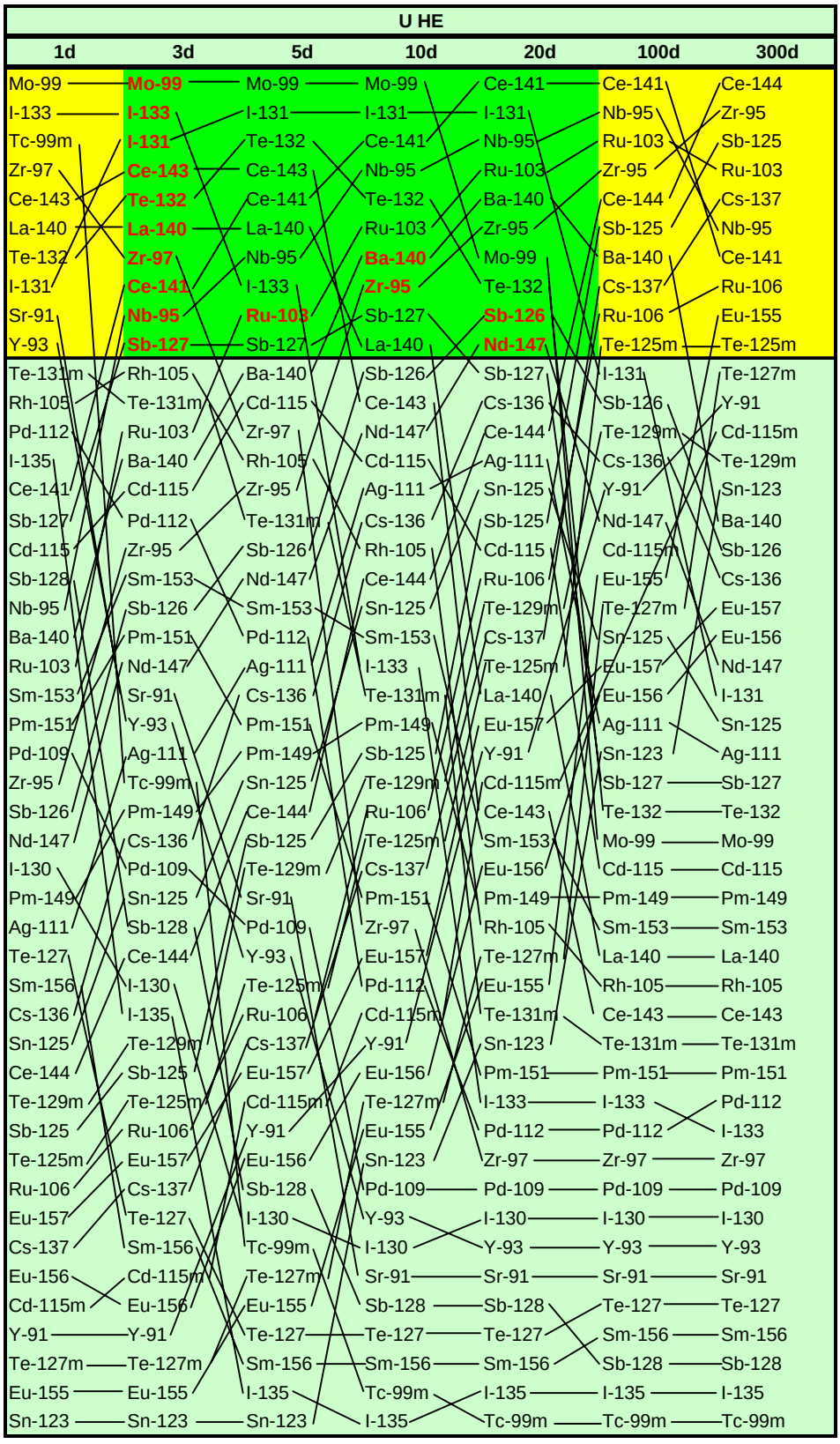


Fig. 4: Rankings of Detection Probability Indices (DPI) for the case of normal ^{239}**Pu** fission in time-bands of 1, 3, 5, 10, 20, 100, 300 days, ordered by decreasing DPI value in each column. The "top 10" fission products in each time-band are highlighted, and time-bands of primary interest are highlighted in green. Individual nuclides are linked to indicate changes in their relative detection likelihood due to decay. Nuclides in the "top-10" group in the 3 – 20 day period are highlighted in red.

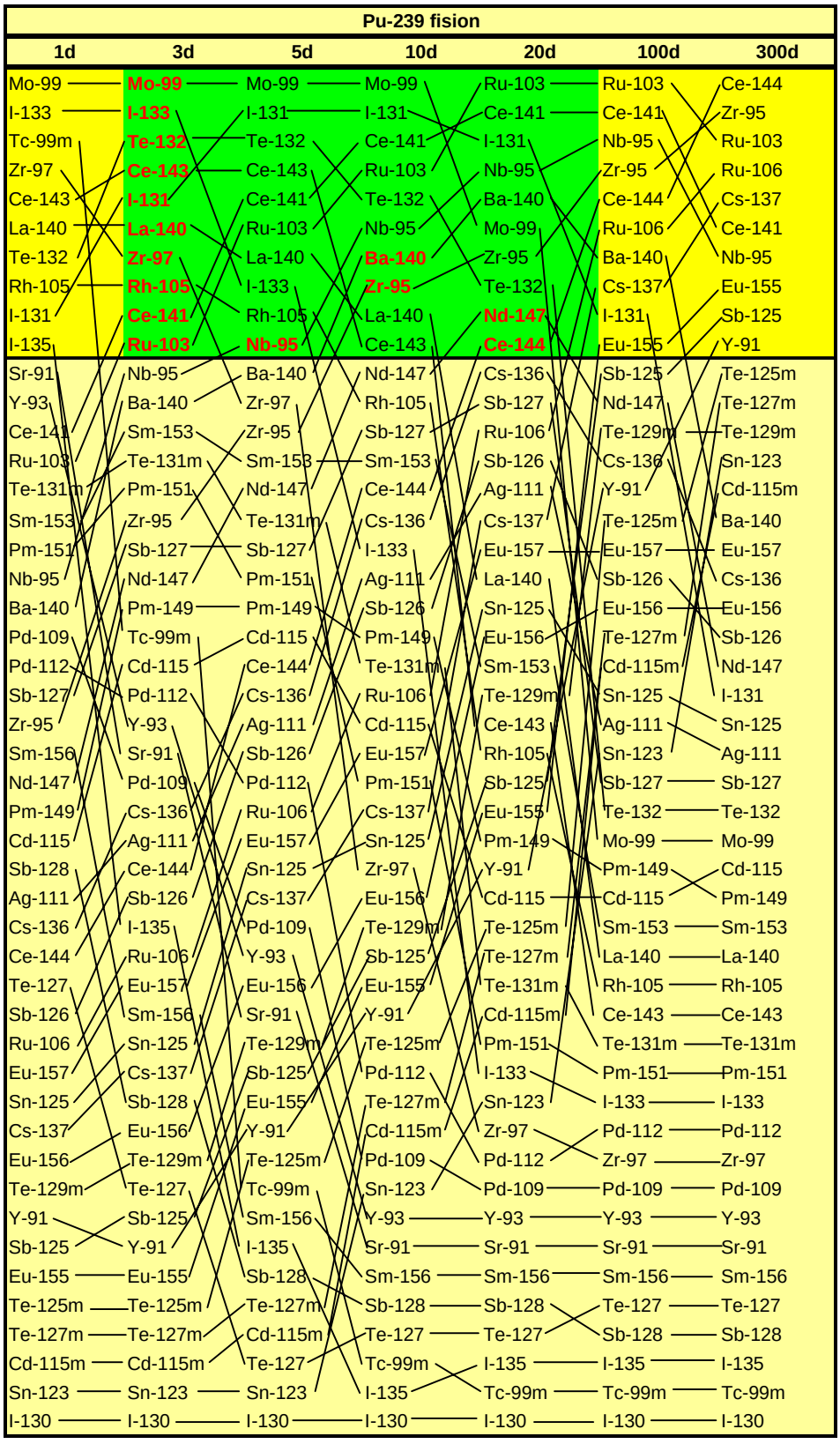


Fig. 5: Rankings of Detection Probability Indices (DPI) for the case of high-energy ^{239}**Pu** fission in time-bands of 1, 3, 5, 10, 20, 100, 300 days, ordered by decreasing DPI value in each column. The "top 10" fission products in each time-band are highlighted, and time-bands of primary interest are highlighted in green. Individual nuclides are linked to indicate changes in their relative detection likelihood due to decay. Nuclides in the "top-10" group in the 3 – 20 day period are highlighted in red.

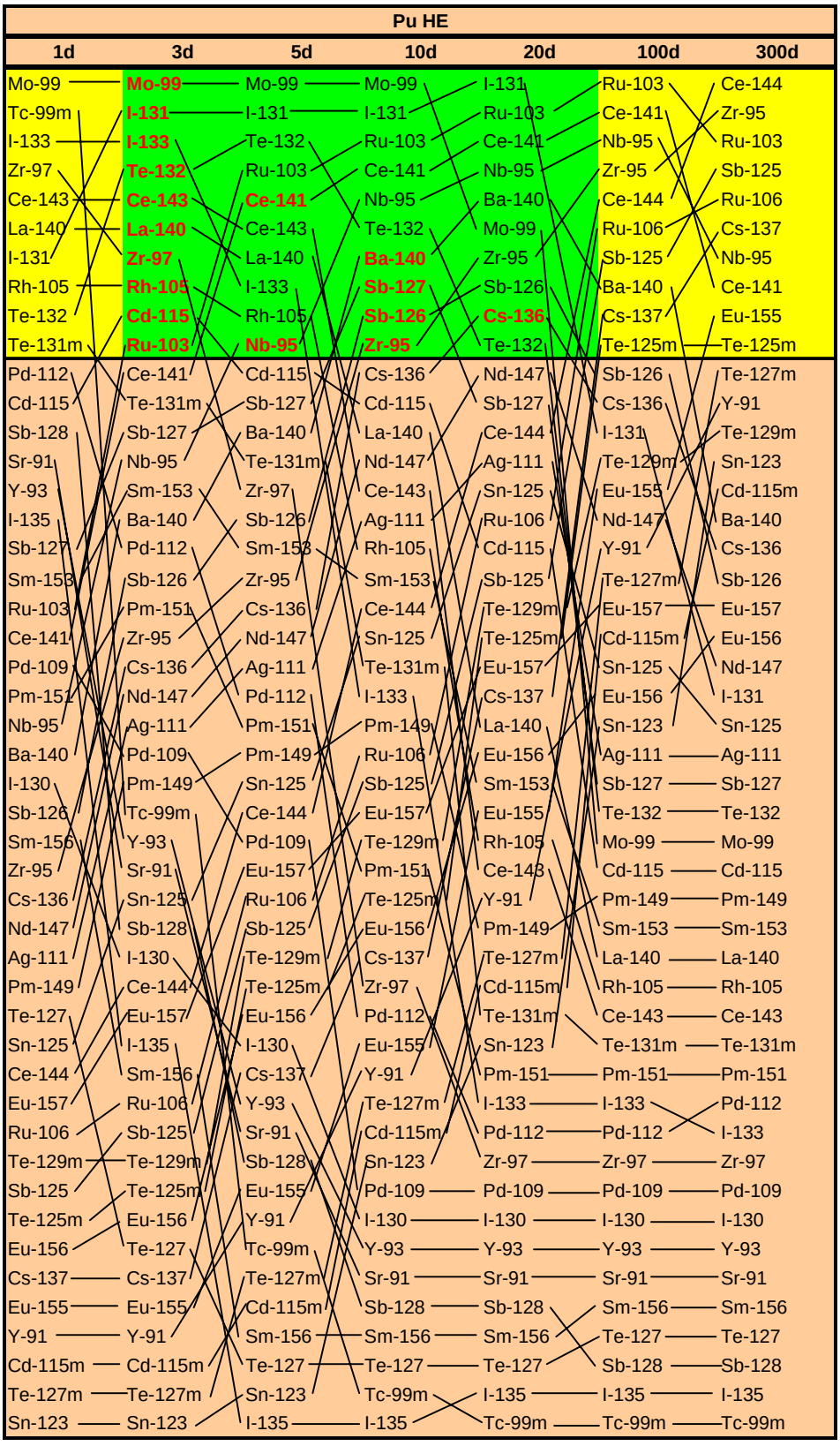


Fig. 6: Rankings of Detection Probability Indices (DPI) for the case of normal ^{238}**U** fission in time-bands of 1, 3, 5, 10, 20, 100, 300 days, ordered by decreasing DPI value in each column. The "top 10" fission products in each time-band are highlighted, and time-bands of primary interest are highlighted in green. Individual nuclides are linked to indicate changes in their relative detection likelihood due to decay. Nuclides in the "top-10" group in the 3 – 20 day period are highlighted in red.

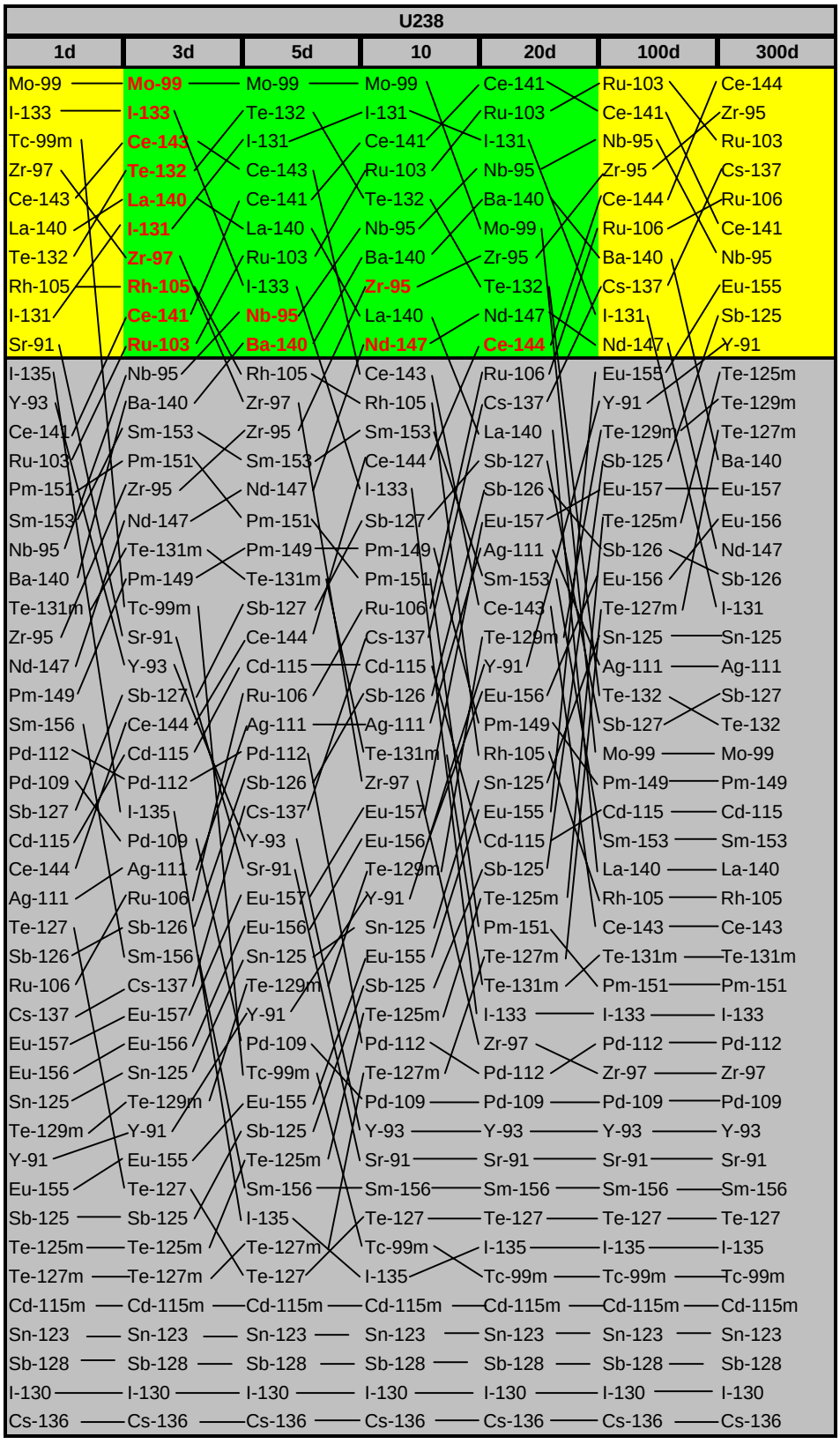


Fig. 7: Rankings of Detection Probability Indices (DPI) for the case of high-energy ^{238}**U** fission in time-bands of 1, 3, 5, 10, 20, 100, 300 days, ordered by decreasing DPI value in each column. The "top 10" fission products in each time-band are highlighted, and time-bands of primary interest are highlighted in green. Individual nuclides are linked to indicate changes in their relative detection likelihood due to decay. Nuclides in the "top-10" group in the 3 – 20 day period are highlighted in red.

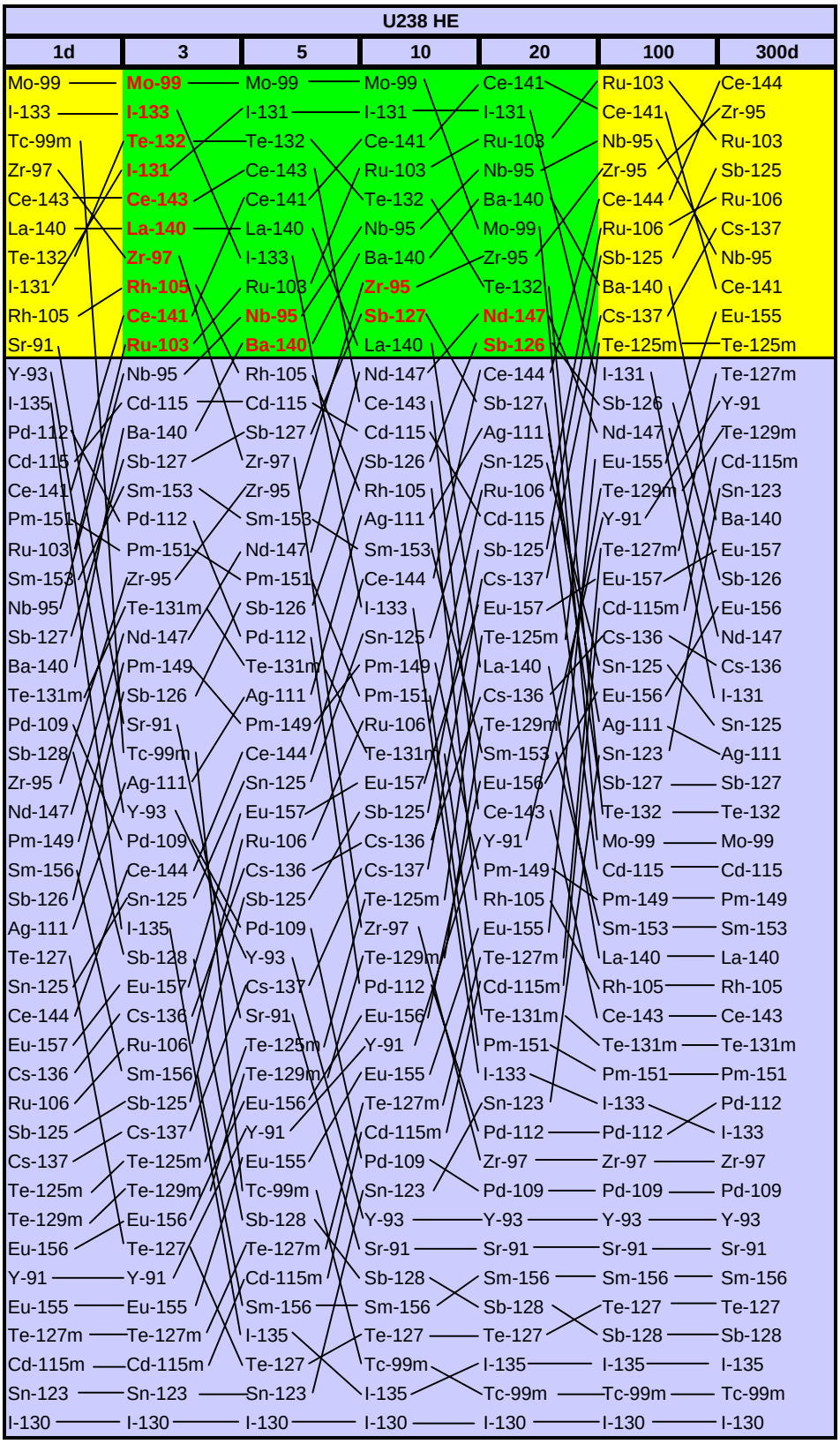
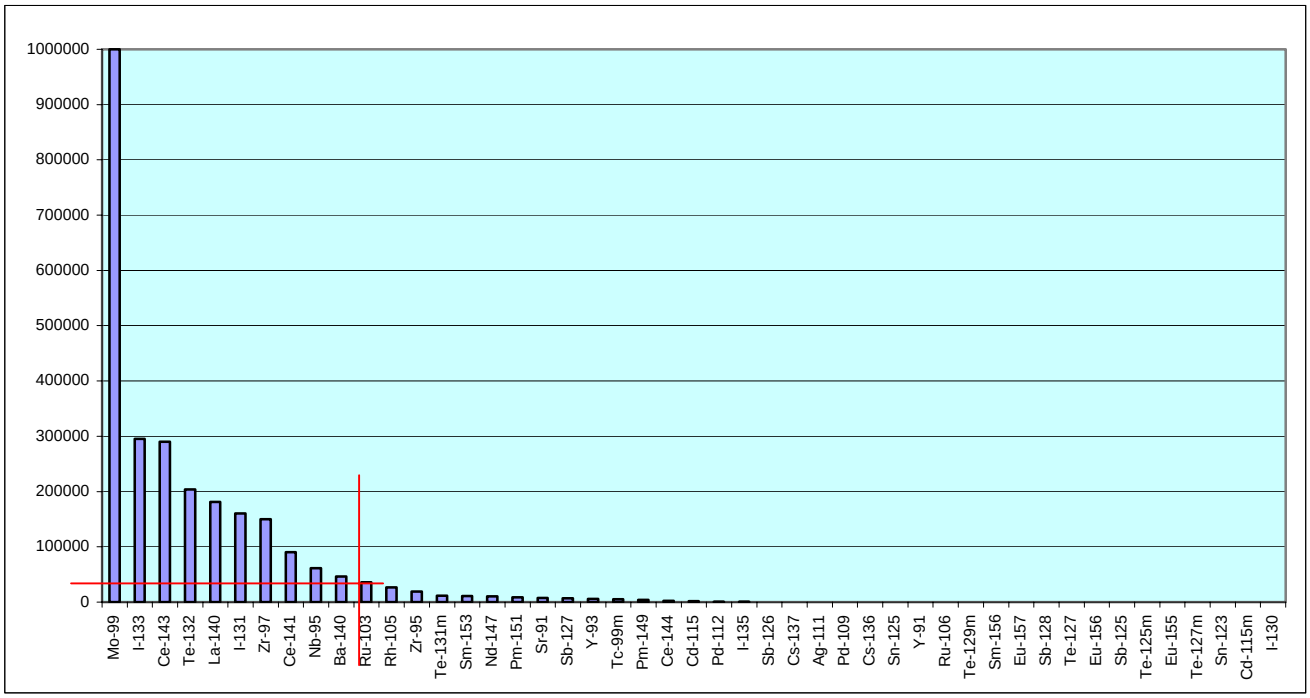


Fig. 8: Graphical representation of the scale of relative detection likelihoods represented by scaled DPI values (y axes) for fission products from normal ^{235}U fission, 3 days after production, depicted with a linearly (upper) and logarithmically (lower). The "top 10" nuclides are indicated by the red lines.



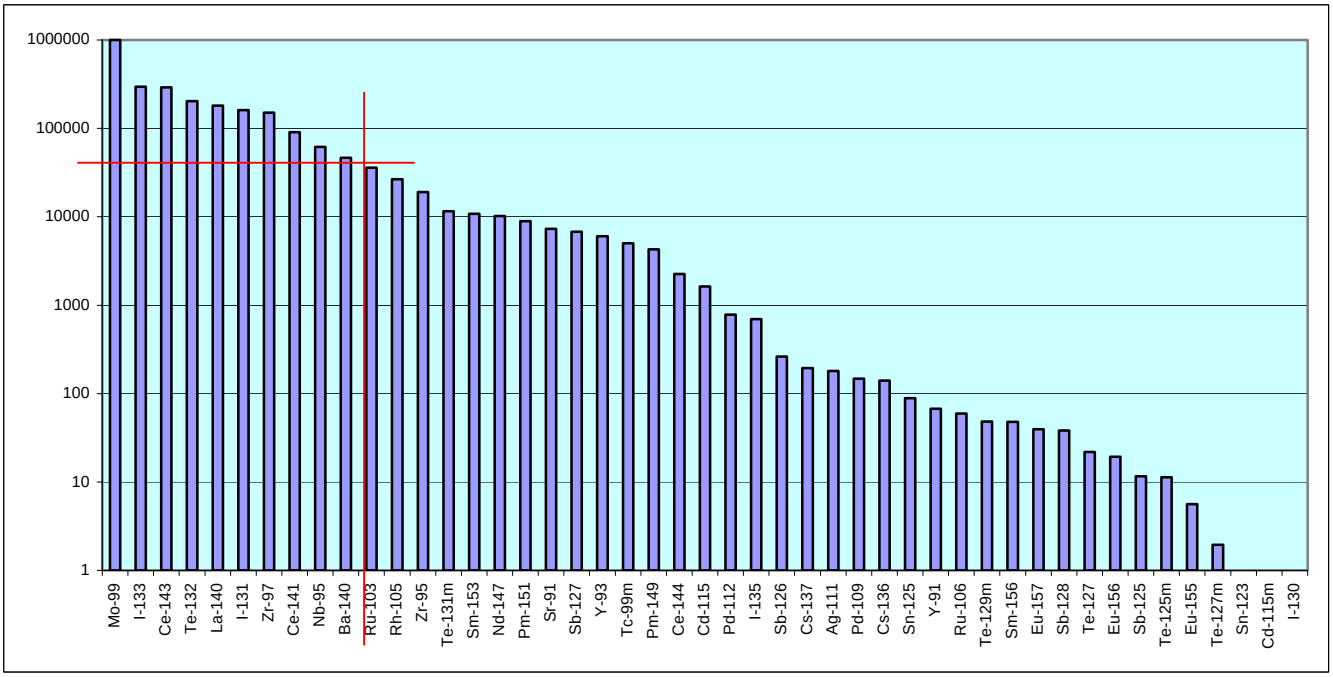
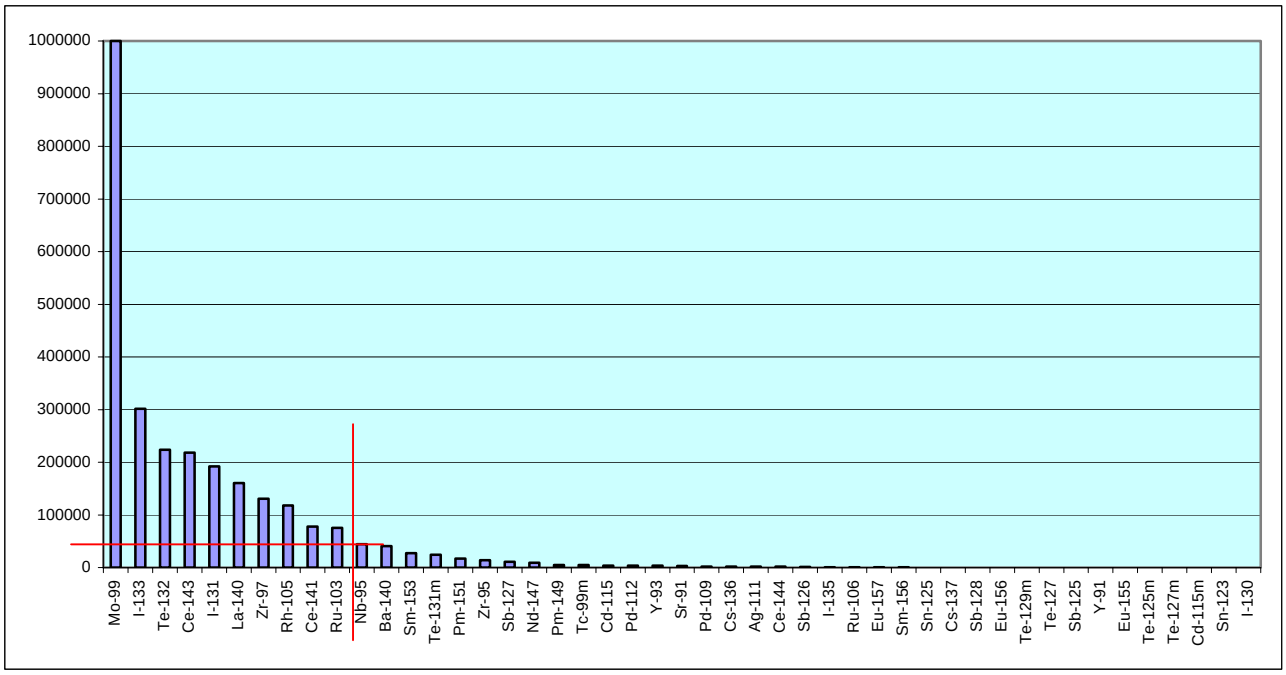


Fig. 9: Graphical representation of the scale of relative detection likelihoods represented by scaled DPI values (y axes) for fission products from normal ^{239}Pu fission, 3 days after production, depicted with a linearly (upper) and logarithmically (lower). The "top 10" nuclides are indicated by the red lines.



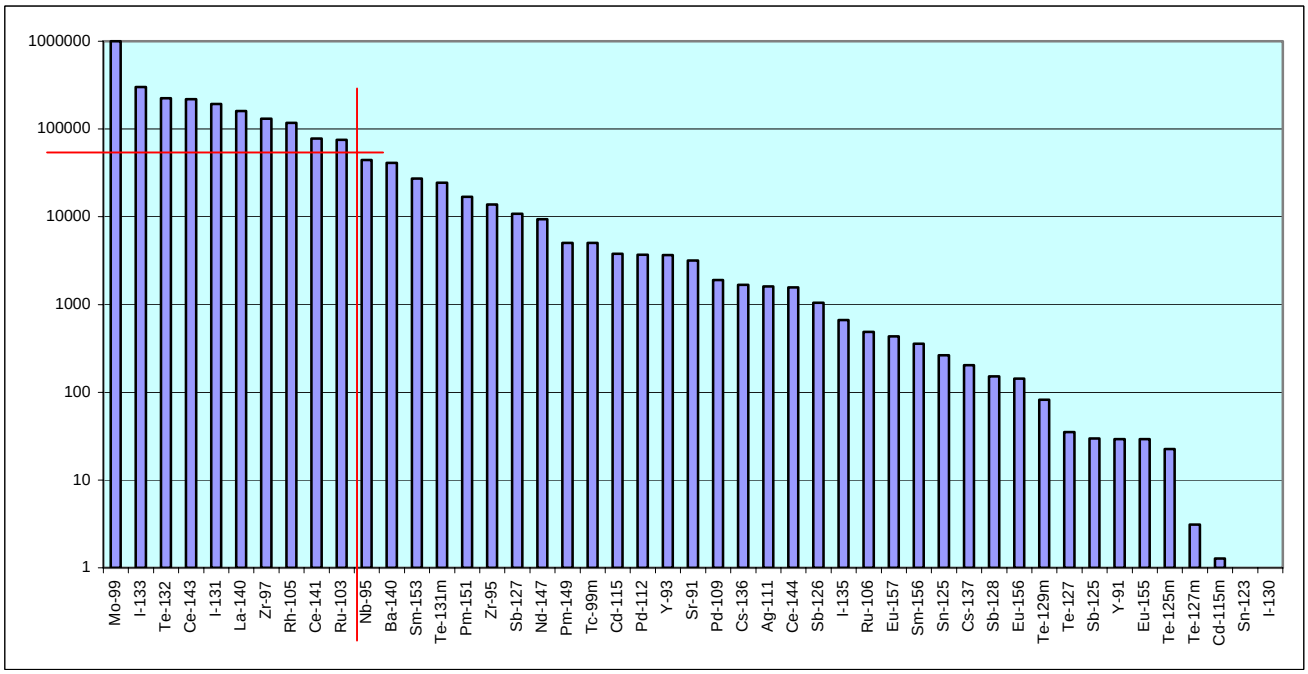
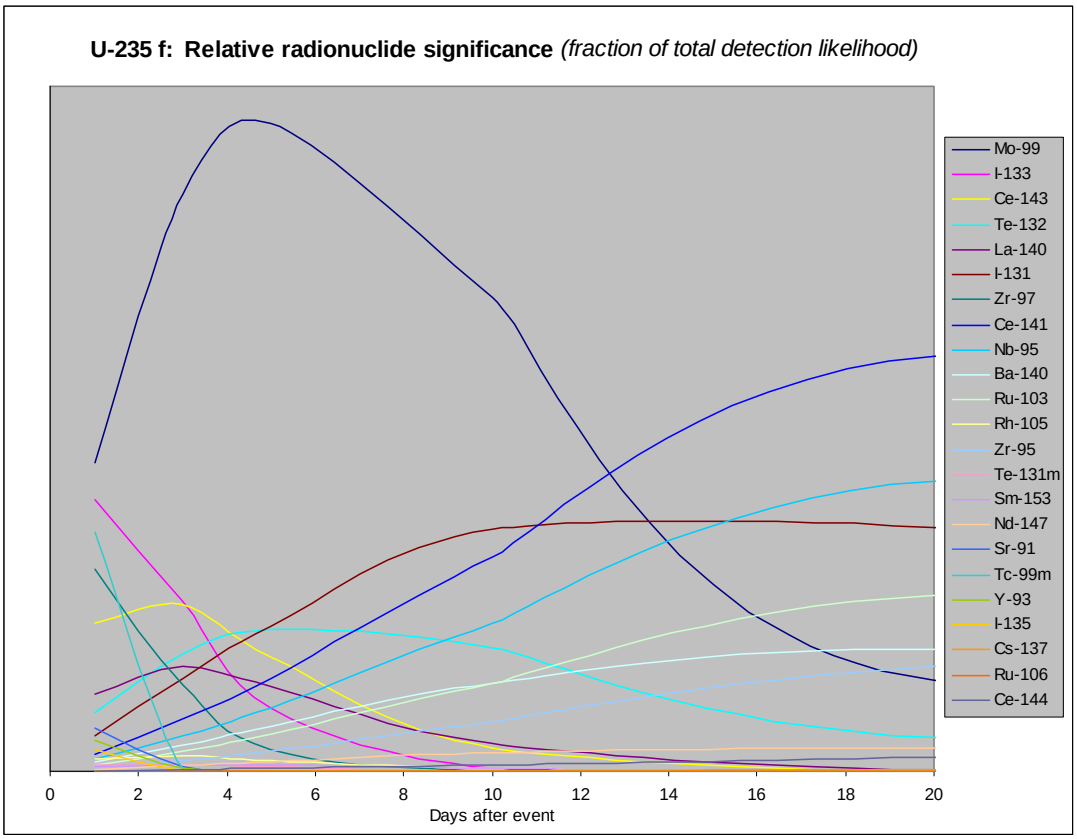


Fig. 10: The variation of ^{235}U fission-product significance over the periods of 20 and 300 days after production.



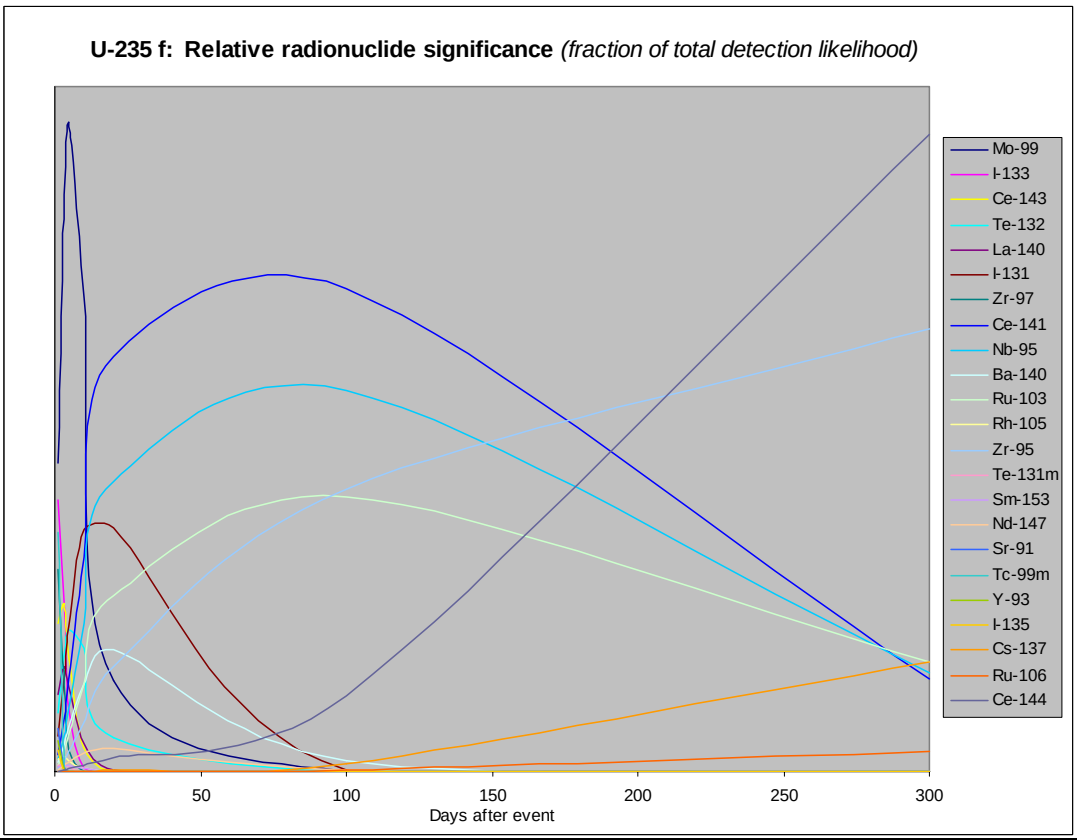
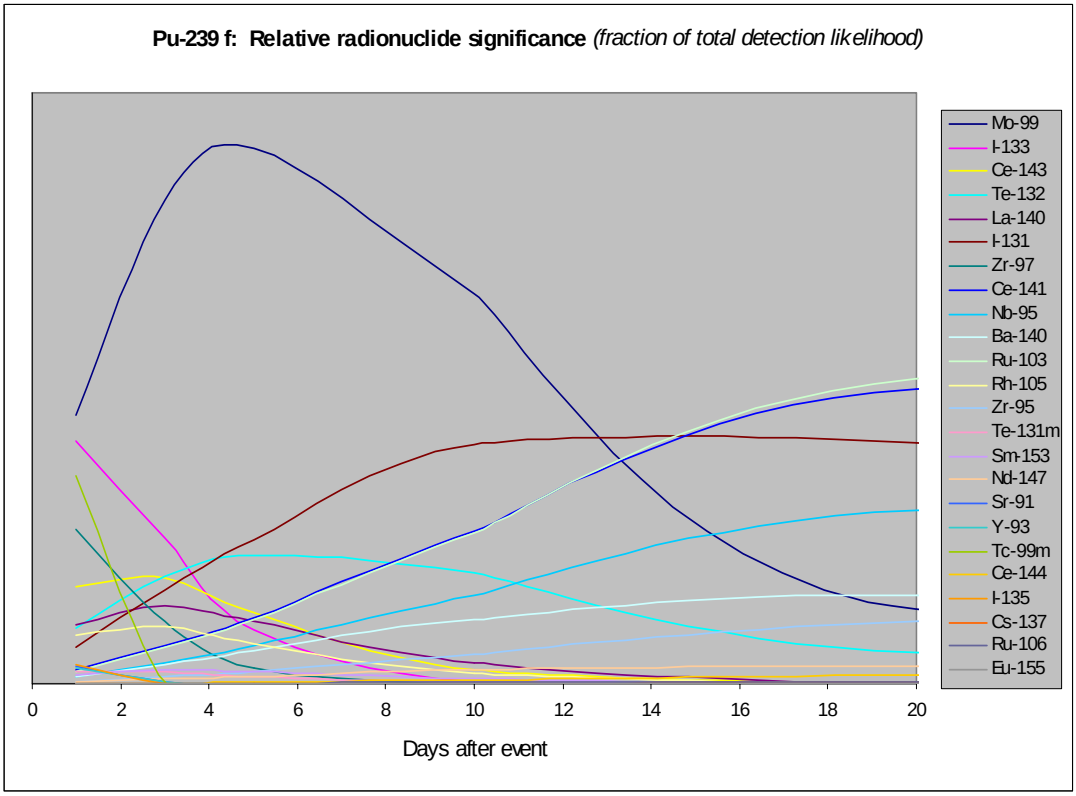
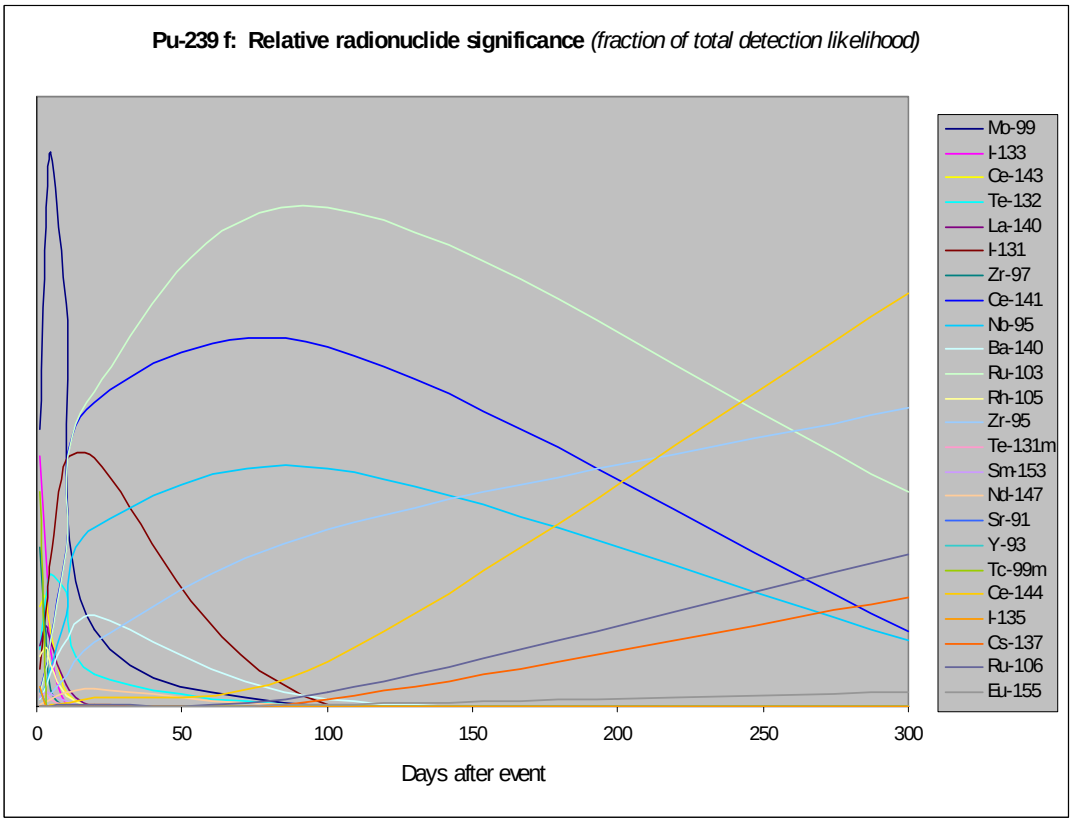


Fig. 11: The variation of ^{239}Pu fission-product significance over the periods of 20 and 300 days after production.





4 Neutron-activation products

Neutron-activation products arise through interaction of neutrons produced during nuclear fission with elements involved in the construction of the device, its fuel, and in the environment surrounding the detonation point. Their detection following any nuclear detonation is therefore undoubtedly of major scientific interest in providing vital clues to the composition, design and performance of the device, and any scientific study of weapons would be incomplete without analysis of activation products. This does not, however, necessarily imply "significance" in the present context of CTBT compliance verification where their likelihood of detection relative to the fission products themselves should be considered.

Following the analogy of fission products described in Section 3.3, the likelihood of detection of an activation product is proportional to the amount of target element present, the isotopic abundance of the target isotope of that element, the cross-section for the neutron reaction, the gamma intensity of the primary emission from the product and the detector efficiency at that energy, and inversely proportional to product half-life, with a correction for decay during the period of interest. These proportionalities translate into the following relationship.

DPI for activation-product A at time t, $\mathbf{(DPI)_{A,t}} = \dfrac{\mathbf{D_t.M_T.A_T.A_c.A_{e,i}.E_e}}{\mathbf{T_{1/2,A}}}$

where M_T = amount of target element (relative mass, moles, or ppm)
A_T = abundance of target isotope, %
A_c = production factor (cross-section), barns
D_t = product decay factor
$A_{e,i}$ = Gamma intensity of principal gamma emission, %
E_e = Detector efficiency at principal gamma energy, %
$T_{1/2,A}$ = Half-life of activation product A.

The application of the DPI concept to activation products is, however, not straightforward because the composition of devices is variable and the M_T values to be used in the above relationship are never known. Indeed, it is the provision of information on relative amounts of materials that makes neutron-activation so useful in elucidating device composition. While the unknown nature of device composition severely limits the applicability of the DPI concept here, some broad generalisations can be made.

Firstly, it is known[1] that normal fission weapons do not produce sufficiently high neutron flux for significant multiple neutron capture to occur, and one-step (n, γ) neutron capture only is involved. Thermonuclear reactions, in contrast, produce very-high-energy neutrons (up to 14 MeV) which can cause (n, 2n) reactions together with (n, p) and (n, α) reactions.

Secondly, construction components (which do not include fuel) may never be known exactly, but there are some obvious ones including[1]: steel, aluminium, brass, plastics, titanium, beryllium, and high explosives. These together provide the following

elements to be activated: hydrogen, beryllium, carbon, nitrogen, oxygen, aluminium, chlorine, titanium, chromium, manganese, iron, cobalt, nickel, copper and zinc. There is also a possibility of the presence of lead (shielding and tamper materials), tungsten (tamper material), boron (neutron shielding), gallium (plutonium phase-stabilising agent), and antimony (to strengthen lead); together with very small quantities of gold, silver and caesium in electronic components[1].

Thirdly, activation reactions are expected to take place in three distinct regions: within the above device components themselves, within materials surrounding the device itself (in an underground scenario), and within the general environment of soil, water, or air. Within the latter two categories, outside the device itself, a softer neutron flux is expected so most reactions there would be of the (n, γ) type. In addition, there may be components deliberately included in the weapon which, when activated, provide information on performance – these "tracers" are not considered in detail here because they are not associated with the weapons themselves and their production is unpredictable and incidental to that of the primary weapon products. These radionuclides were included in the relevant nuclides list simply because they had been detected historically.

These three sets of factors allow at least some application of the DPI concept to activation products. With environmental materials, for example, device composition is not a factor and only the natural abundance of elements is required in place of M_T in the above equation.

Relevant activation products are listed in Table 5, together with details of their production. Four activation products arise principally from interaction with environmental materials sodium, potassium and europium which pertain to soil, rock, or seawater environments; 24 are produced from reactions of materials contained in the device itself; and 13 are primarily produced as performance-monitoring tracers; and some are produced from both device and environmental materials, notably ^{24}Na, ^{54}Mn and ^{59}Fe.

Table 5: Neutron-activation products included in the relevant radionuclides list (excluding fuel-products mentioned in Section 2).

Product	Half-life d	Main reaction	Target	Isotopic A. %	X, barns	E, keV	I %
Principally involving activation of environmental materials							
^{24}Na	0.62	(n, γ)*	^{23}Na	100	0.40	1368.6	100
^{42}K	0.52	(n, γ)	^{41}K	6.73	1.46	1524.7	18.1
^{152m}Eu	0.39	(n, γ)	^{151}Eu	47.8	5300	841.6	14.6
^{152}Eu	66932	(n, γ)	^{151}Eu	47.8	2800	1408.8	20.9
Principally involving activation of device materials							
^{46}Sc	83.79	(n, p)	^{46}Ti	8.25	0.27	889.3	100
^{47}Sc	3.35	(n, p)	^{47}Ti	7.44	0.14	159.4	67.9
^{51}Cr	27.70	(n, γ)*	^{50}Cr	4.35	7.80	320.1	10
^{54}Mn	312.12	(n,p)	^{54}Fe	5.85	0.5	834.8	100
^{58}Co	70.82	(n, p)	^{58}Ni	68.08	0.6	810.8	99
^{59}Fe	44.50	(n, γ)*	^{58}Fe	0.28	1.7	1099.2	56.5
^{60}Co	1925.1	(n, γ)	^{59}Co	100	74	1332.5	100
^{64}Cu	0.53	(n, γ)	^{63}Cu	69.17	4.97	1345.8	0.47
^{65}Zn	244.26	(n, γ)	^{64}Zn	48.6	1.45	1114.5	50.6
^{69m}Zn	0.57	(n, γ)	^{68}Zn	18.8	3.0	438.6	94.8
^{72}Ga	0.59	(n, γ)*	^{71}Ga	39.89	31.2	834.1	95.6
^{106m}Ag	8.28	(n, 2n)	^{107}Ag	51.84	0.7	717.2	28.9
^{108m}Ag	152674	(n, γ)	^{107}Ag	51.84	1.2	722.9	90.8
^{110m}Ag	249.8	(n, γ)	^{109}Ag	48.16	72.3	657.8	94
^{120}Sb	5.76	(n, 2n)	^{121}Sb	57.21	0.6	1171.7	100
^{122}Sb	2.72	(n, γ)*	^{121}Sb	57.21	200	564.2	69.3
^{124}Sb	60.20	(n, γ)	^{123}Sb	42.79	125	602.7	97.8
^{132}Cs	6.48	(n, 2n)	^{133}Cs	100	1.30	667.7	97.5
^{134}Cs	754.2	(n, γ)	^{133}Cs	100	437	604.7	97.6
^{187}W	0.99	(n, γ)	^{186}W	28.43	485	685.7	27.3
^{196}Au	6.18	(n, 2n)	^{197}Au	100	2	355.7	86.9
^{196m}Au	0.39	(n, 2n)	^{197}Au	100	0.14	147.8	42.5
^{198}Au	2.70	(n, γ)	^{197}Au	100	1550	411.8	96
^{203}Pb	2.16	(n, 2n)	^{204}Pb	1.4	1.9	279.2	81
Tracer materials							
^{57}Co	271.79						
^{74}As	17.77						
^{76}As	1.08						
^{84}Rb	32.77						
^{86}Rb	18.63						
^{88}Y	106.65						
^{89}Zr	3.27						
^{102}Rh	207						
^{133}Ba	3842						
^{168}Tm	93.1						
^{170}Tm	128.6						
^{190}Ir	11.78						
^{192}Ir	73.83						

Main reaction: * implies there are other significant reactions, depending on device type, but the one shown is considered to be the main one in the present context.
Isotopic A = isotopic abundance of target isotope, %
X = cross-section or resonance integral of principal reaction, barns
E = primary gamma energy of activation product, keV
I = intensity of primary gamma, %.
All data are from reference 1.

4.1 Environmental materials

DPI values calculated for nuclides arising from environmental activation are shown in Table 6, where it can be seen that ^{24}Na is the most likely activation product to be detected soon after detonation.

Table 6: Relative significance ratings for activation products produced through reactions of environmental constituents

Activation product	DPI values after elapsed times			Reaction type
	3 days	10 days	20 days	
^{24}Na	1384.4	0.6	0.0	(n, γ)
^{152m}Eu	35.9	0.0	0.0	(n, γ)
^{42}K	29.3	0.0	0.0	(n, γ)
^{59}Fe	7.6	6.8	5.9	(n, γ)
^{152}Eu	0.3	0.3	0.3	(n, γ)
^{54}Mn	20.9	20.6	20.2	(n, p)

The activation products referred to in Table 6 pertain to detonation scenarios involving detonation in or near the ground or seawater. The primary purpose of the particulate radionuclide monitoring network is the detection of above-ground explosions, however, and the target nuclides are not present in the atmosphere. De Geer[1] states, for detonations in the atmosphere, "No CTBT relevant nuclide is expected to be produced by neutron activation of the air constituents". The only generalisation that can be made here is that for aboveground detonations there will be no significant activation products, but where the fireball may have contacted the Earth's surface ^{24}Na would be the most likely activation product to be detected in the short term, with ^{54}Mn and ^{59}Fe becoming more significant as time progresses.

4.2 Device materials

Device composition (which excludes fuel in the present context) may not be known, but some generalisations seem possible. It would be reasonable to expect, for example, that (1) there would be much more steel present than gold and silver; (2) the steel might be of high quality, perhaps even of the "stainless" variety and that its composition in terms of iron, chromium, nickel and manganese might approximate that of stainless steel; (3) electronic components, which contain gold, silver, caesium and copper, would be present in amounts which are very small compared to the steel; (4) the lead content is unlikely to be higher than the steel in terms of mass, and its antimony content could be of the order of 0.5%. Even with these generalisations it is not possible to objectively apply the DPI concept in the absence of relative mass data.

DPI values were calculated without inclusion of the relative mass term to at least show the propensity of some elements for activation. Results are shown in Table 7, and indicate that gold, antimony and tungsten are strongly activated in (n, γ) reactions while chromium, zinc, copper, iron and silver are not. This does not, however, necessarily mean gold activation products would be more detectable than the others

because there is likely to be relatively little gold in the device compared to copper, for example. To convert the data in Table 7 to true DPI values, each figure would have to be multiplied by the relative mass of the respective target element.

If a gold:iron mass ratio of 1:10000 were assumed, with the iron being in the form of stainless steel, with an equivalent amount of lead present, with silver and caesium being present in similar amounts to the gold, and with the other target elements being present in intermediate amounts, then the relative detection probability orders would be as shown in Table 8. The history of nuclide detection is also shown in Table 8 where nuclides coloured red have been detected following nuclear tests in the past[1]. The clustering of previously detected nuclides near the top of the DPI order provides some support for the mass ratios assumed here, although the results should obviously be interpreted with caution. Concerning which activation products from device components would be most likely to be detected in the 3 – 20 day timeframe, the results presented here indicate the following:

Without consideration of relative masses, ^{198}Au, ^{122}Sb and ^{187}W would be the most likely nuclides detected from normal fission devices, with ^{196}Au being added for thermonuclear devices. Applying the relative mass ratios used here, ^{187}W, ^{122}Sb, ^{124}Sb, ^{198}Au, ^{60}Co, ^{51}Cr, ^{65}Zn, and ^{54}Mn would be the most likely choices.

Table 7: DPI values (rounded) for neutron activation products arising from device components, calculated without inclusion of the relative mass term.

Product	Target	DPI (3 days)	DPI (10 days)	DPI (20days)
(n, γ) reactions				
^{198}Au	^{197}Au	17900000	3000000	230000
^{122}Sb	^{121}Sb	760000	130000	10000
^{187}W	^{186}W	220000	2000	2
^{124}Sb	^{123}Sb	44000	41000	37000
^{134}Cs	^{133}Cs	30000	30000	30000
^{72}Ga	^{71}Ga	24000	6	<1
^{110m}Ag	^{109}Ag	6000	6000	6000
^{69m}Zn	^{68}Zn	2000	<1	<1
^{60}Co	^{59}Co	1000	1000	1000
^{51}Cr	^{50}Cr	100	80	60
^{65}Zn	^{64}Zn	50	50	50
^{64}Cu	^{63}Cu	20	<1	<1
^{59}Fe	^{58}Fe	2	2	1
^{108m}Ag	^{107}Ag	<1	<1	<1
(n, p) reactions				
^{58}Co	^{58}Ni	230	210	200
^{46}Sc	^{46}Ti	160	150	140
^{47}Sc	^{47}Ti	140	30	4
^{69m}Zn	^{69}Ga	60	<1	<1
^{59}Fe	^{59}Co	20	16	14
^{54}Mn	^{54}Fe	4	4	4
^{64}Cu	^{64}Zn	1	<1	<1
^{60}Co	^{60}Ni	<1	<1	<1
(n, α) reactions				
^{24}Na	^{27}Al	200	<1	<1
^{51}Cr	^{54}Fe	2	1	1
^{60}Co	^{63}Cu	<1	<1	<1
^{59}Fe	^{62}Ni	<1	<1	<1
(n, 2n) reactions				
^{196}Au	^{197}Au	16000	7000	2000
^{132}Cs	^{133}Cs	7000	3000	1000
^{122}Sb	^{123}Sb	4000	700	50
^{120}Sb	^{121}Sb	1200	500	200
^{106m}Ag	^{107}Ag	500	250	100
^{203}Pb	^{204}Pb	350	40	2
^{58}Co	^{59}Co	140	130	120
^{54}Mn	^{55}Mn	100	100	100
^{51}Cr	^{52}Cr	70	60	50
^{65}Zn	^{66}Zn	50	50	45
^{69m}Zn	^{70}Zn	10	<1	<1
^{64}Cu	^{65}Cu	1	<1	<1

Table 8: Detection probability orders (decreasing downward) for activation products 3, 10 and 20 days after production, based on NPI values calculated with the relative mass assumptions described in the text. Nuclides in red have been detected historically[1].

3 days	10 days	20 days
(n, γ) reactions		
^{187}W	^{122}Sb	^{124}Sb
^{122}Sb	^{198}Au	^{122}Sb
^{72}Ga	^{124}Sb	^{198}Au
^{198}Au	^{187}W	^{60}Co
^{69m}Zn	^{60}Co	^{51}Cr
^{124}Sb	^{51}Cr	^{65}Zn
^{60}Co	^{65}Zn	^{134}Cs
^{51}Cr	^{134}Cs	^{59}Fe
^{65}Zn	^{59}Fe	^{110m}Ag
^{64}Cu	^{72}Ga	^{187}W
^{134}Cs	^{110m}Ag	^{108m}Ag
59Fe	^{69m}Zn	^{72}Ga
^{110m}Ag	^{64}Cu	^{69m}Zn
^{108m}Ag	^{108m}Ag	^{64}Cu
(n, p) reactions		
^{58}Co	^{58}Co	^{58}Co
^{46}Sc	^{46}Sc	^{46}Sc
^{47}Sc	^{54}Mn	^{54}Mn
^{69m}Zn	^{47}Sc	^{47}Sc
^{54}Mn	^{59}Fe	^{59}Fe
^{59}Fe	^{60}Co	^{60}Co
^{64}Cu	^{69m}Zn	^{69m}Zn
^{60}Co	^{64}Cu	^{64}Cu
(n, α) reactions		
^{24}Na	^{51}Cr	^{51}Cr
^{51}Cr	^{60}Co	^{60}Co
^{60}Co	^{59}Fe	^{59}Fe
^{59}Fe	^{24}Na	^{24}Na
(n, 2n) reactions		
^{203}Pb	^{203}Pb	^{51}Cr
^{122}Sb	^{51}Cr	^{65}Zn
^{51}Cr	^{65}Zn	^{58}Co
^{65}Zn	^{122}Sb	^{54}Mn
^{120}Sb	^{58}Co	^{203}Pb
^{58}Co	^{120}Sb	^{120}Sb
^{69m}Zn	^{54}Mn	^{122}Sb
^{54}Mn	^{196}Au	^{196}Au
^{196}Au	^{132}Cs	^{132}Cs
^{132}Cs	^{106m}Ag	^{106m}Ag
^{64}Cu	^{69m}Zn	^{69m}Zn
^{106m}Ag	^{64}Cu	^{64}Cu

4.3 Comparison with fission products

While consideration of which activation products might be most detectable after a nuclear weapon detonation is scientifically interesting, and there is no doubt that much attention would be focussed on activation products following such an event, the important issue in the present context is whether or not activation products are significant in terms of detection of an event. To answer this their relative detectability compared to fission products must be considered. As fission is necessary to produce the neutron flux required for activation reactions, there cannot be activation products in the absence of fission products. Intuitively then, it would be expected that fission products alone might be sufficient as indicators of an event having taken place. Whether or not this is so can only be determined empirically through historical data pertaining to real events and, as indicated below (Section 5), there has not been much historical opportunity for this.

Recent re-analysis[5] of 1980 Swedish monitoring data[3] (see Section 5.2) provided information on the relativity between fission- and activation-products following an above-ground event where the fireball evidently did not contact the ground, so no activation-products from reactions with soil and rock (environmental) materials would be expected. The relativity between fission- and activation-products was assessed by comparing spectral peak areas obtained by analysis of the raw data at NRL[5]. Results are shown in Table 9.

As can be seen in Table 9, the only activation products detected were as follows:

^{51}Cr	possible contribution to 319 keV peak
^{54}Mn	305 counts
^{58}Co	697 counts (questionable peak identity)
^{60}Co	453 counts (one gamma line only detected)
^{88}Y	244 counts
^{124}Sb	359 counts
^{198}Au	667 counts

The activation-products detected fit well with the predictions made in Section 4.2 above, but their peak areas are orders of magnitude lower than those attributable to the most significant fission-products, which had peak areas in the range 671 – 159000 counts and with the "top 10" being above 7000 counts.

This relativity between fission- and activation-products is supported by the 1976 Swedish spectrum referred to in Section 5.1 (Fig. 12) where although fission-products are obviously visible in the gamma spectrum, activation-products are not. Nor do activation-products contribute significantly to the 1980 Swedish spectrum (Fig.13).

Conclusion: Some predictions may be possible concerning the relative likelihood of detection of activation-products. Compared to fission-products, however, activation-products products are not significant for CTBT verification purposes within the context of atmospheric particulate monitoring.

Table 9: Detections of significant fission-products and relevant activation-products in the laboratory proficiency-test spectrum[6].

Spectral peak areas (net counts for principal gamma emissions)			
Relevant activation-products		**Significant fission-products**	
Nuclide	**Counts**	**Nuclide**	**Counts**
^{24}Na	Not detected	^{131}I	159000
^{42}K	Not detected	^{141}Ce	156000
^{46}Sc	Not detected	^{103}Ru	134000
^{47}Sc	Not detected	^{140}La	99300
^{51}Cr	Not detected*	^{140}Ba	55200
^{54}Mn	305	^{95}Zr	35600
^{57}Co	Not detected	^{132}Te	22000
^{58}Co	697**	^{99}Mo	16800
^{59}Fe	Not detected	^{147}Nd	11900
^{60}Co	453 (at 1173 keV)	^{95}Nb	7230
^{64}Cu	Not detected	^{144}Ce	5780
^{65}Zn	Not detected	^{127}Sb	4950
^{69m}Zn	Not detected	^{106}Ru	1030
^{72}Ga	Not detected	^{143}Ce	778
^{74}As	Not detected	^{126}Sb	671
^{76}As	Not detected		
^{84}Rb	Not detected		
^{86}Rb	Not detected		
^{88}Y	244		
^{89}Zr	Not detected		
^{102}Rh	Not detected		
^{106m}Ag	Not detected		
^{108m}Ag	Not detected		
^{110m}Ag	Not detected		
^{120}Sb	Not detected		
^{122}Sb	Not detected		
^{124}Sb	359		
^{132}Cs	Not detected		
^{133}Ba	Not detected		
^{134}Cs	Not detected		
^{152m}Eu	Not detected		
^{168}Tm	Not detected		
^{170}Tm	Not detected		
^{187}W	Not detected		
^{190}Ir	Not detected		
^{192}Ir	Not detected		
^{196}Au	Not detected		
^{196m}Au	Not detected		
^{198}Au	667		
^{203}Pb	Not detected		

* There was a peak at 319.2 keV (^{51}Cr principal line is at 320.1 keV) with area 2640 counts, which might have been partially attributable to ^{51}Cr.

** Questionable peak identity

5 Validation of significance rankings

Any system for prioritising nuclides should be validated by comparison with experimental data. There are only limited historical opportunities for such validation because the requirement would be for above-ground weapons tests to have occurred at a time when high-resolution gamma spectrometers were available for use in IMS-type monitoring systems. These conditions occurred jointly only in the late period of Chinese above-ground tests during 1975-1980 when high-resolution Ge(Li) detectors had just become available. Validation of the choice of significant fission-products was sought by consideration of four sets of data arising from the Swedish monitoring programme[2,3], a programme maintained in Finland[4], and results obtained in New Zealand from monitoring in the South Pacific[5]. Each of these is discussed below.

5.1 Swedish 1976 spectrum

An atmospheric particulate sample was collected on 26 November 1976, following an aboveground nuclear test by China on 17 November. The sample was analysed 13 days after the event. The spectrum[2] is illustrated in Fig. 12, while detections of nuclides are summarised in Table 10. As can be seen in Fig.12, virtually every spectral peak is associated with the defined Significant Nuclides.

The "top 10" fission-products detected were, in decreasing order of detection likelihood: ^{99}Mo, ^{143}Ce, ^{105}Rh, ^{132}Te, ^{131}I, ^{141}Ce, ^{103}Ru, ^{140}Ba, ^{115}Cd and ^{127}Sb. All of these nuclides are included in the list of significant nuclides described in Section 3.3 (Table 4). The results shown in Table 10 include ^{95}Zr, ^{147}Nd, ^{144}Ce and ^{136}Cs, and these are also classed here as being significant nuclides. Moreover, the Swedish results include ^{105}Rh and ^{115}Cd, which are considered here as significant nuclides pertaining to thermonuclear (high-energy neutron) devices, and these too are included in the list of significant nuclides. These Swedish results thus confirm that the list of significant nuclides would have been sufficient for detection, identification and even classification of this event. The significant fuel-products, ^{237}U and ^{239}Np are also clearly evident in the spectrum (Table 10, Fig. 12). It is also interesting to note that activation-products, which have very low detection likelihood values compared to the significant fission products (^{58}Co, ^{54}Mn, ^{88}Y, ^{57}Co, ^{65}Zn and ^{60}Co have values of 0.01% or less of that of ^{99}Mo, Table 10), are barely, if at all, visible in the spectrum (Fig. 12) – reinforcing the view that activation-products are not significant relative to fission-products.

5.2 Swedish 1980 spectrum

A recent proficiency test[3] for radionuclide laboratories supporting the CTBT verification effort was also used to test the significant-nuclides list, and as an indicator of the relative significance of fission- and activation-products. The proficiency test was based on another high-resolution spectrum obtained during monitoring of Chinese weapon tests. The device was thermonuclear with a yield of up to 1 Mt, detonated above-ground on 16 October 1980. The sample was collected at an altitude of 14 km on 27 October 1980 and analysed 16 days after the event. The raw spectral data were re-analysed by various laboratories, including the National Radiation Laboratory (NRL) in 2003[5]. The most significant fission-products detected were as shown in Table 11 where it can be seen that the top-10 fission products are all represented in the proposed significant-nuclides list (Table 4). The spectrum itself is shown in Fig. 13, where the significant nuclides are highlighted. It is clear that by far the majority of spectral peaks are associated with fission-products defined here as being significant (^{132}I is included because it is the decay product of ^{132}Te and the two are detected together).

Table 10: Nuclides detected by Sweden following a Chinese nuclear test in 1976. Published results[3] were in terms of number of atoms detected. For fission- and activation-products these were converted here to detection likelihoods (activity x intensity x efficiency) relative to ^{99}Mo. Nuclides classed here as being significant for CTBT verification purposes are highlighted in **red**. Activation products are shown in blue.

Nuclide	Relative No. atoms	Half-life d	Relative activity	Gamma intensity %	Detector efficiency %	Detection likelihood	Rel to Mo-99
Np-239	41.400	2.4	17.62				
U-237	31.000	6.8	4.59				
Mo-99	9.400	2.8	3.38	89.4	13.4	4038.6	100.00
Ce-143	6.160	1.4	4.46	42.8	8.9	1706.1	42.24
Rh-105	5.560	1.5	3.71	19.2	8.4	599.9	14.86
Te-132	5.850	3.3	1.80	75.6	4.3	583.8	14.46
I-131	5.830	8.0	0.73	81.7	7.7	455.0	11.27
Ce-141	7.360	32.4	0.23	48.2	13.2	144.6	3.58
Ru-103	8.700	39.6	0.22	90.9	6.1	122.4	3.03
Ba-140	7.100	12.8	0.56	24.4	5.8	78.3	1.94
Cd-115	0.409	2.2	0.18	45.9	8.1	68.8	1.70
Sb-127	1.040	3.9	0.27	36.8	4.8	46.9	1.16
Zr-95	7.980	65.5	0.12	54.5	4.4	29.0	0.72
Nd-147	3.450	11.0	0.31	13.1	5.8	24.0	0.59
Ag-111	0.516	7.5	0.07	6.7	8.0	3.7	0.09
Ce-144	6.430	284.0	0.02	11.1	13.6	3.4	0.08
Sn-125	0.217	9.7	0.02	10.0	3.2	0.7	0.02
Cs-136	0.029	13.0	0.00	80.0	3.3	0.6	0.01
Sb-126	0.015	12.4	0.00	99.6	4.7	0.6	0.01
Co-58	0.085	71.3	0.00	99.0	4.1	0.5	0.01
Ru-106	3.340	367.0	0.01	9.9	5.2	0.5	0.01
Te-129m	0.829	33.5	0.02	3.2	4.7	0.4	0.01
Cs-137	9.310	11020.0	0.00	85.1	4.9	0.4	0.01
Sb-125	0.749	997.0	0.00	29.6	6.9	0.2	0.00
Eu-156	0.094	15.2	0.01	6.8	3.0	0.1	0.00
Y-91	7.170	58.5	0.12	0.3	2.9	0.1	0.00
Mn-54	0.051	312.0	0.00	100.0	4.0	0.1	0.00
Y-88	0.035	107.0	0.00	99.2	2.0	0.1	0.00
Eu-155	0.216	1810.0	0.00	21.2	13.9	0.0	0.00
Co-57	0.003	270.0	0.00	99.0	13.8	0.0	0.00
Zn-65	0.007	244.0	0.00	50.6	3.1	0.0	0.00
Co-60	0.007	1920.0	0.00	100.0	2.6	0.0	0.00

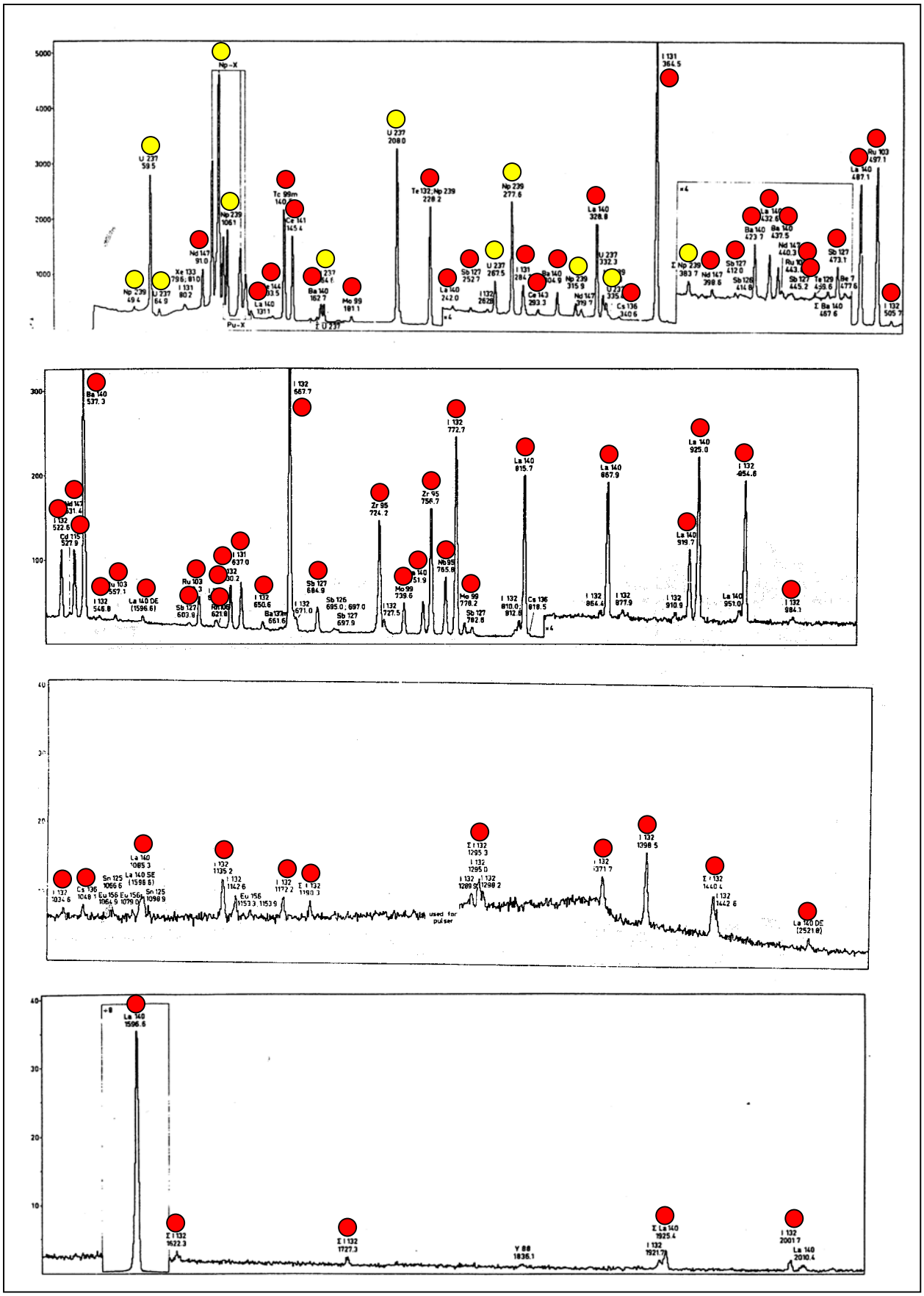


Fig. 12: Spectrum[2] of debris a 1976 Chinese test 13 days after detonation, in intervals of roughly 0-500, 500-1000, 100-1500, 1500-2000 keV. Significant fission-products are indicated by ● , and fuel-products by ● . (Note: ^{132}I is the short-lived decay product of ^{132}Te).

Table 11: The "top 10" fission-product detections in the NRL re-analysis of Swedish spectral data from 1980. Red colour indicates inclusion in the proposed significant nuclides list.

Fission-product	Peak area (net counts)
^{131}I	159000
^{141}Ce	156000
^{103}Ru	134000
^{140}La	99300
^{140}Ba	55200
^{95}Zr	35600
^{132}Te	22000
^{99}Mo	16800
^{147}Nd	11900
^{95}Nb	7230

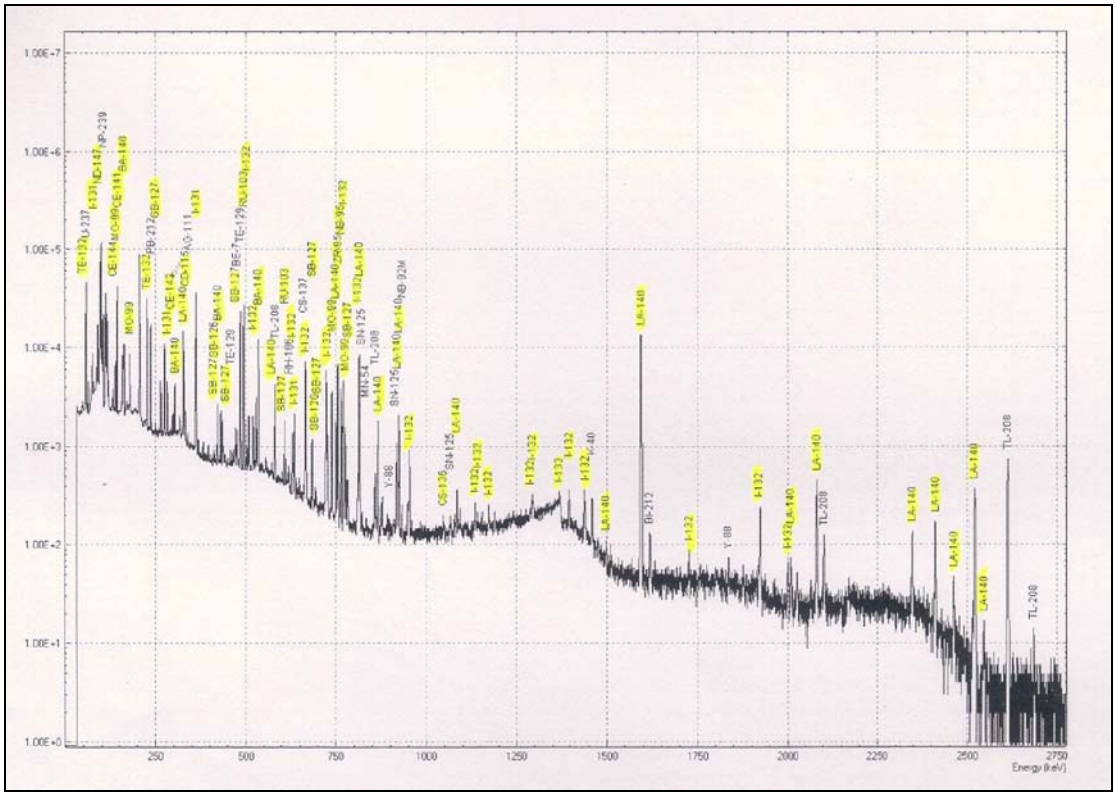


Fig. 13: Spectrum from a 1980 Chinese weapon test[3]. Significant nuclides defined in the present report are highlighted yellow.

5.3 Finnish monitoring

Environmental monitoring conducted by Finland[4] during the period 1977 to 1980 focussed on the fission-products ^{99}Mo, ^{131}I, ^{132}Te, ^{140}Ba, ^{147}Nd, ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{125}Sb, ^{137}Cs, ^{141}Ce, ^{144}Ce, ^{155}Eu most of which are classed here as significant.

5.4 NRL monitoring, 1966

Further validation of the significant nuclides list was provided by an incident in 1966 when a French nuclear test conducted at Mururoa resulted in detection of fresh fission products in Fiji[6]. The nuclides were detected by low-volume air sampling with analysis by low-resolution gamma spectroscopy. The spectrum is shown in Fig. 14. The detected nuclides were: ^{141}Ce, ^{99}Mo, ^{132}Te, ^{131}I, ^{103}Ru, ^{95}Zr, ^{140}Ba – all included in the proposed significant-nuclides list.

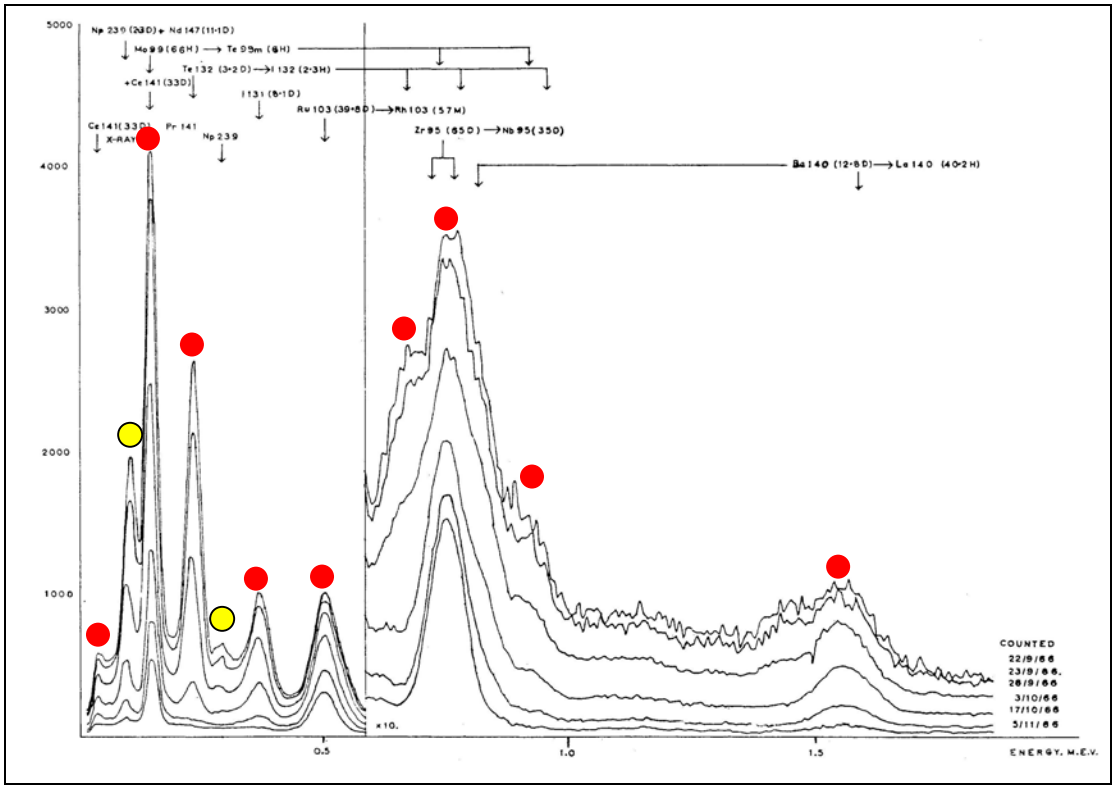


Fig 14: Low-resolution spectra acquired over success days in Fiji following a 1966 French test[6]. Structures due to significant fission-products are indicated by ● , and fuel-products by ○.

Conclusion: It is clear that the recommended list of significant radionuclides does indeed represent those which are most likely to be detected following an aboveground nuclear detonation, and that their choice is consistent with those focussed on in other national monitoring programmes.

6 Conclusion

Nuclear weapon detonations result in three general groups of products for use in detection of the event and in proving its nuclear origin. These are fuel-products resulting from fuel debris and its reactions; fission-products resulting from the fission reactions themselves, and activation-products from reactions of the neutrons produced during fission with device and environment components. The relevant-nuclides list is a compilation containing all radionuclides from within these groups which could conceivably be detected by the IMS, without consideration of their relative likelihoods of detection.

Relative detection likelihoods of the relevant fission-products can be judged on the basis of their Detection Probability Indices which incorporate factors pertaining to production, gamma emission, and detector efficiency. This allows the compilation of a list of "significant" particulate fission-products, which are those most likely to be detected in the atmosphere after an aboveground nuclear weapon detonation.

Compared to fission-products, activation-products are not significant for weapon detection in the CTBT-verification context, although they would be of scientific interest in elucidating device design. In contrast, the fuel-products included in the relevant-nuclides list are mostly considered to be significant in this context.

These considerations lead to the compilation of the following list of CTBT-significant radionuclides, for application during the period up to 20 days after detonation.

Fuel-products:	^{241}Am, ^{239}Np, ^{237}U
Fission-products:	^{99}Mo, ^{133}I, ^{143}Ce, ^{132}Te, ^{140}La, ^{131}I, ^{97}Zr, ^{141}Ce, ^{95}Nb, ^{140}Ba, ^{103}Ru, ^{95}Zr, ^{136}Cs, ^{115}Cd, ^{127}Sb, ^{126}Sb, ^{105}Rh, ^{144}Ce, ^{147}Nd, ^{131m}Te
Activation-products:	None

Within the period up to 10 days after production, ^{99}Mo is by far the most predominant fission-product likely to be detected. Because of this, special attention should be paid to its detection even if this occurs in the absence of other significant radionuclides, particularly if the ^{99}Mo is detected at trace levels such that other significant nuclides might have been present but below their limits of detectability. A corollary to this is that IDC analysis procedures should be developed to give as much confidence as possible in distinguishing between ^{99m}Tc and ^{75m}Ge, and between supported and unsupported ^{99m}Tc.

The above list of Significant Radionuclides is proposed for application by NDCs in screening IMS particulate monitoring results, with sufficient confidence for the detection and identification of any above-ground nuclear weapon detonation (within the limitations of the IMS itself).

References

1 De Geer, L E. *CTBT Relevant Radionuclides*. Technical Report PTS/IDC-1999/02, April 1999. CTBTO, Vienna. Also: *Comprehensive Nuclear-test-Ban Treaty: relevant radionuclides*, Kerntechnik 66 (2001) 3, 113-120.

2 De Geer L E, Arntsing R, Vintersved I, Sisefsky J, Jabobsson S, Engström J-A. *Particulate Radioactivity, Mainly from nuclear explosions, in air and precipitation in Sweden mid-year 1975 to mid-year 1977*. FOA rapport C 40089-T2(A1), November 1978. Försvarets Forskningsanstalt, Sweden.

3 *Proficiency Test for Radionuclide Laboratories Supporting the Network of IMS Radionuclide Stations (PTE2003)*. Technical Report CTBT/PTS/TR/2004-2, August 2004, CTBTO, Vienna.

4 *Studies on Environmental Radioactivity in Finland*. Annual Reports STL-A26 (1976-1977), STL-A32 (1978), STL-A34 (1979), STL-A38 (1980), Institute of Radiation Protection, Helsinki.

5 Hermanspahn, N. Results of NRL analysis of PTE2003 spectrum – Pers. Comm.

6 *Environmental Radioactivity in New Zealand*. Report NRL-F23, Dept of Health, 1966.