T3.2-O2 More sensitive measurements that radionuclide laboratories can do for special studies

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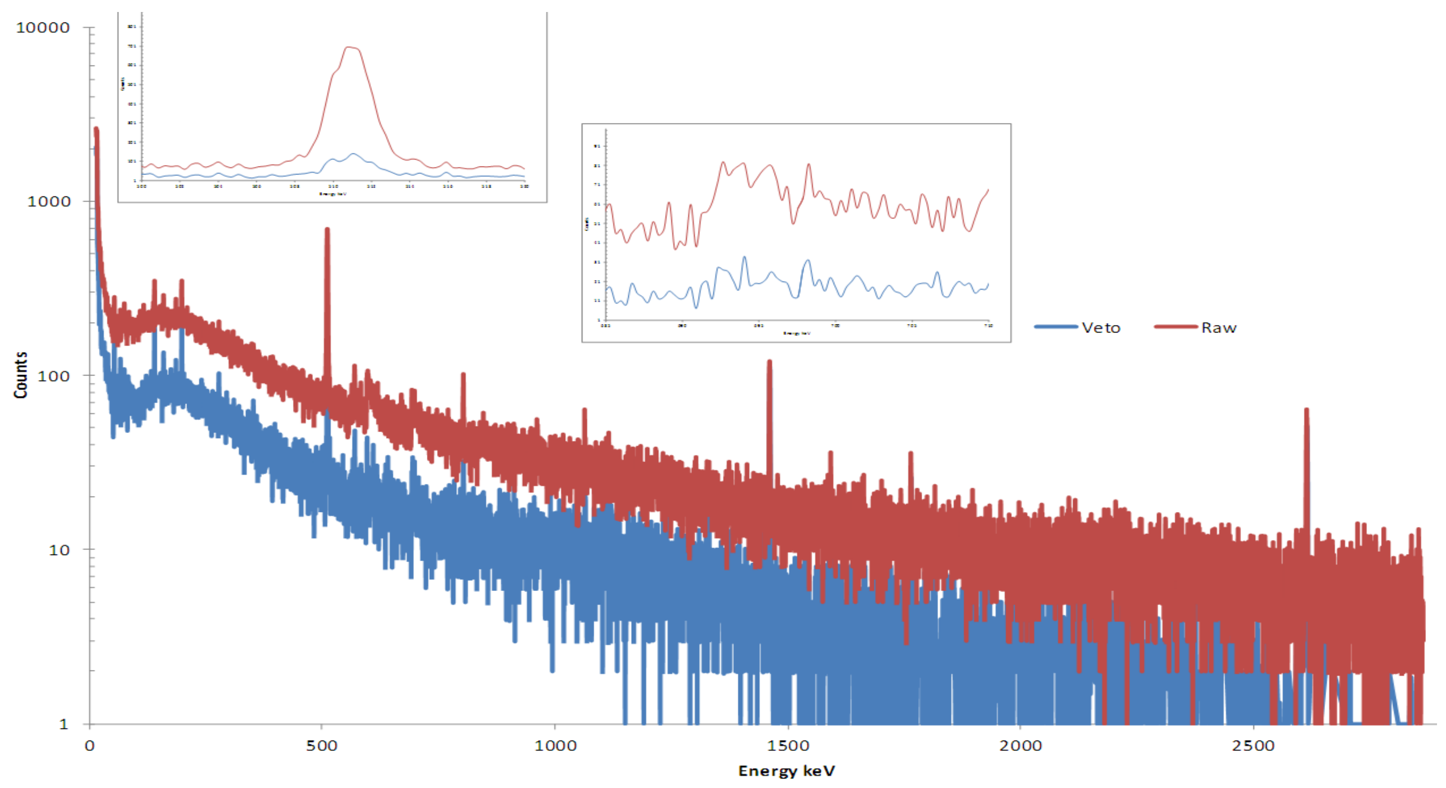
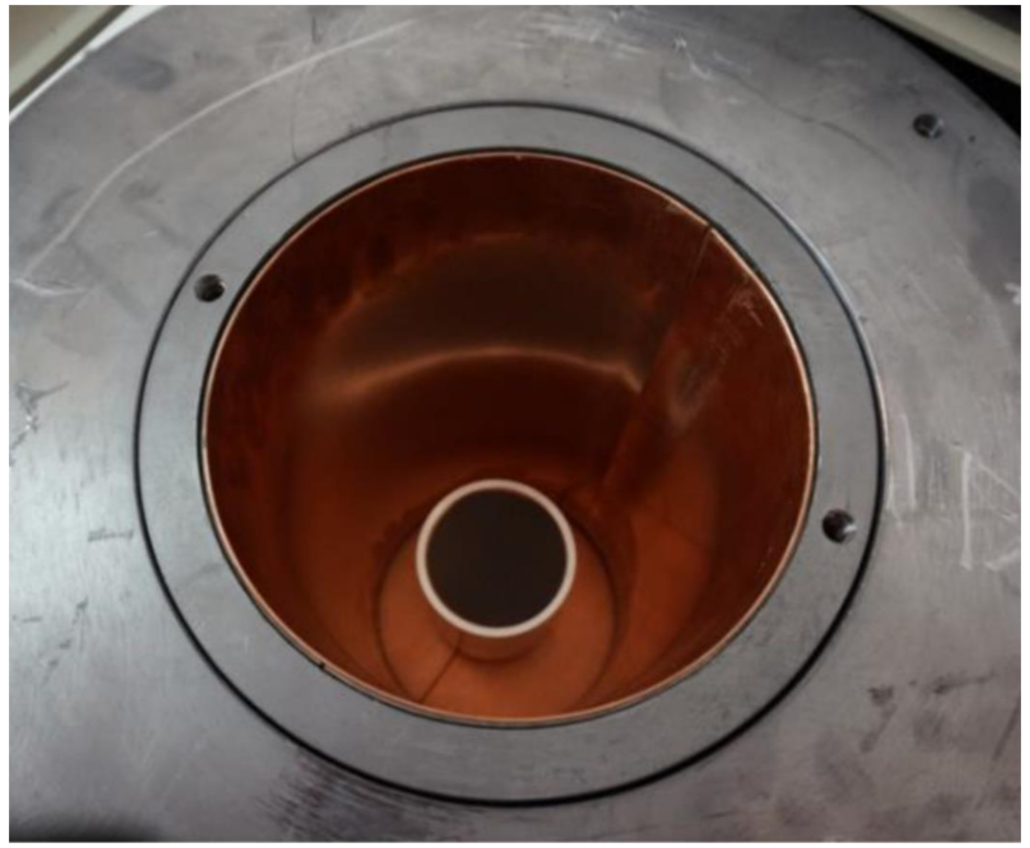
Conventional Gamma spectrometry

- Technique deployed at IMS stations and Radionuclide laboratories
- Spectra collected over measurement time



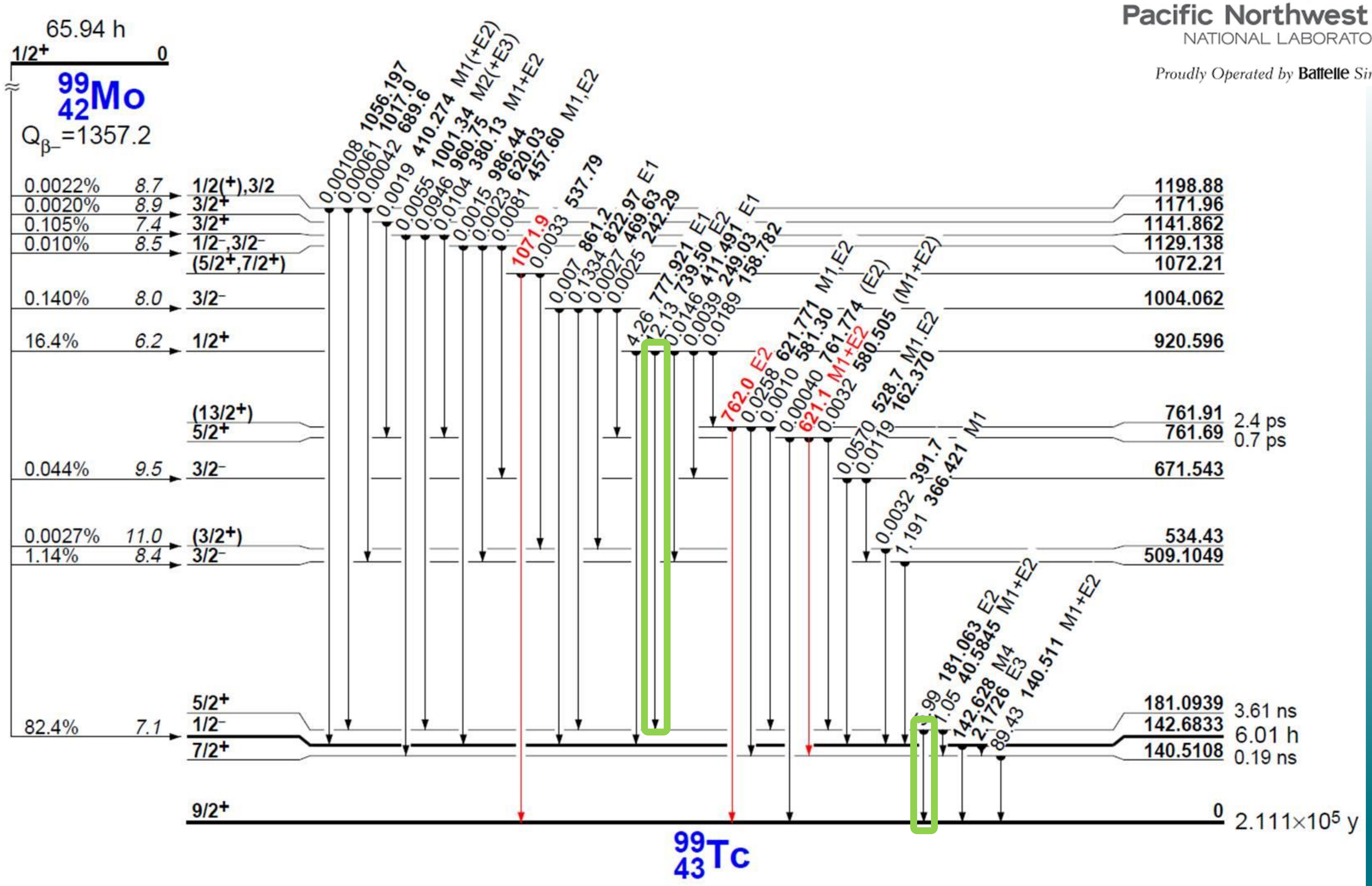


Options to improve sensitivity (Minimum Detectable Activity)

- Increase measurement time – not possible for IMS stations, RN labs already 7 days
- Increase sample size – not possible with current IMS stations
- Improving energy resolution of the detector – best available detectors used at RN labs
- Pre-concentration or radiochemical purification – outside of the treaty
- Reducing the background
  - Low background detector materials and shielding – done
  - Cosmic Veto shields / underground facilities – done
  - **Gamma-Gamma Coincidence**

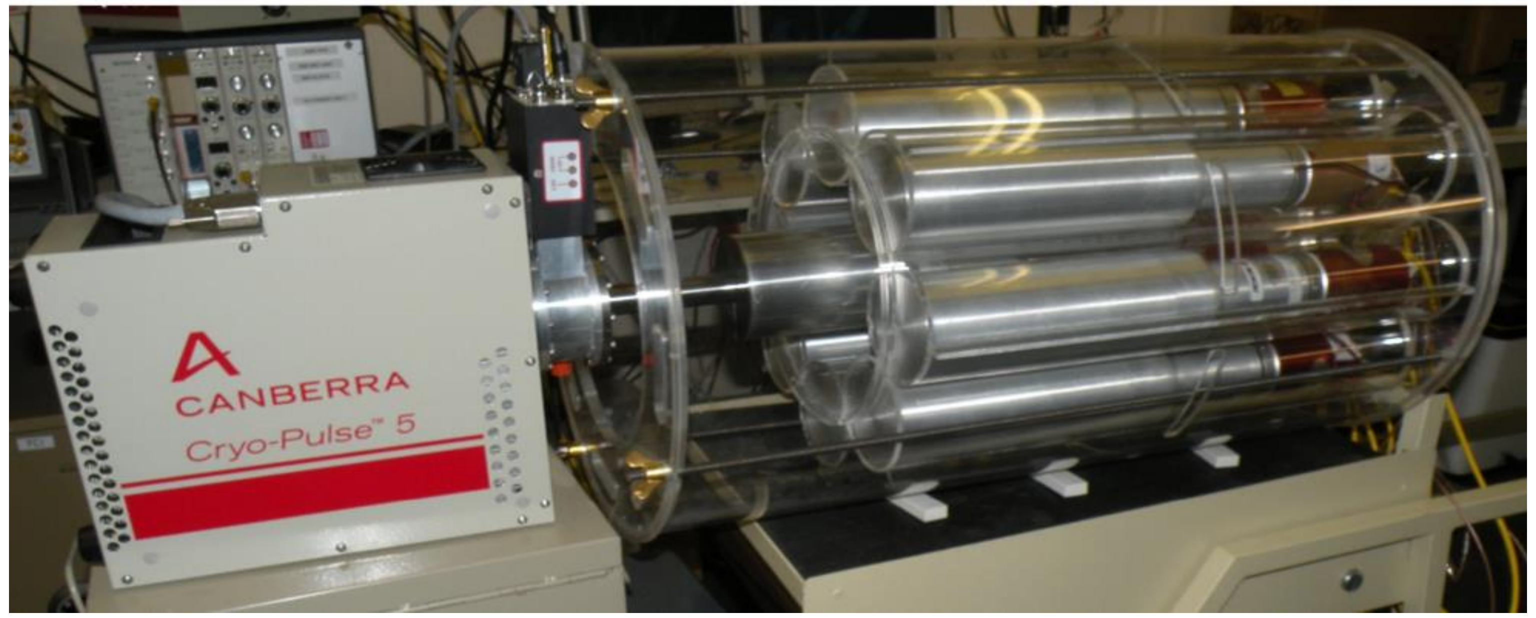
Gamma-Gamma Coincidence spectrometry

- Spectra constructed from events defined by two or more gammas detected within a specific window of time on multiple detectors
- List mode uses digital acquisition systems acquiring sequential list of events with timestamps – processed to recreate multiple spectra
- Allows the experiment to be 'replayed' event-by-event, and gated coincidence projections of peak regions
- Quantification and analysis of signals (singles & coincidence) using RIMMER

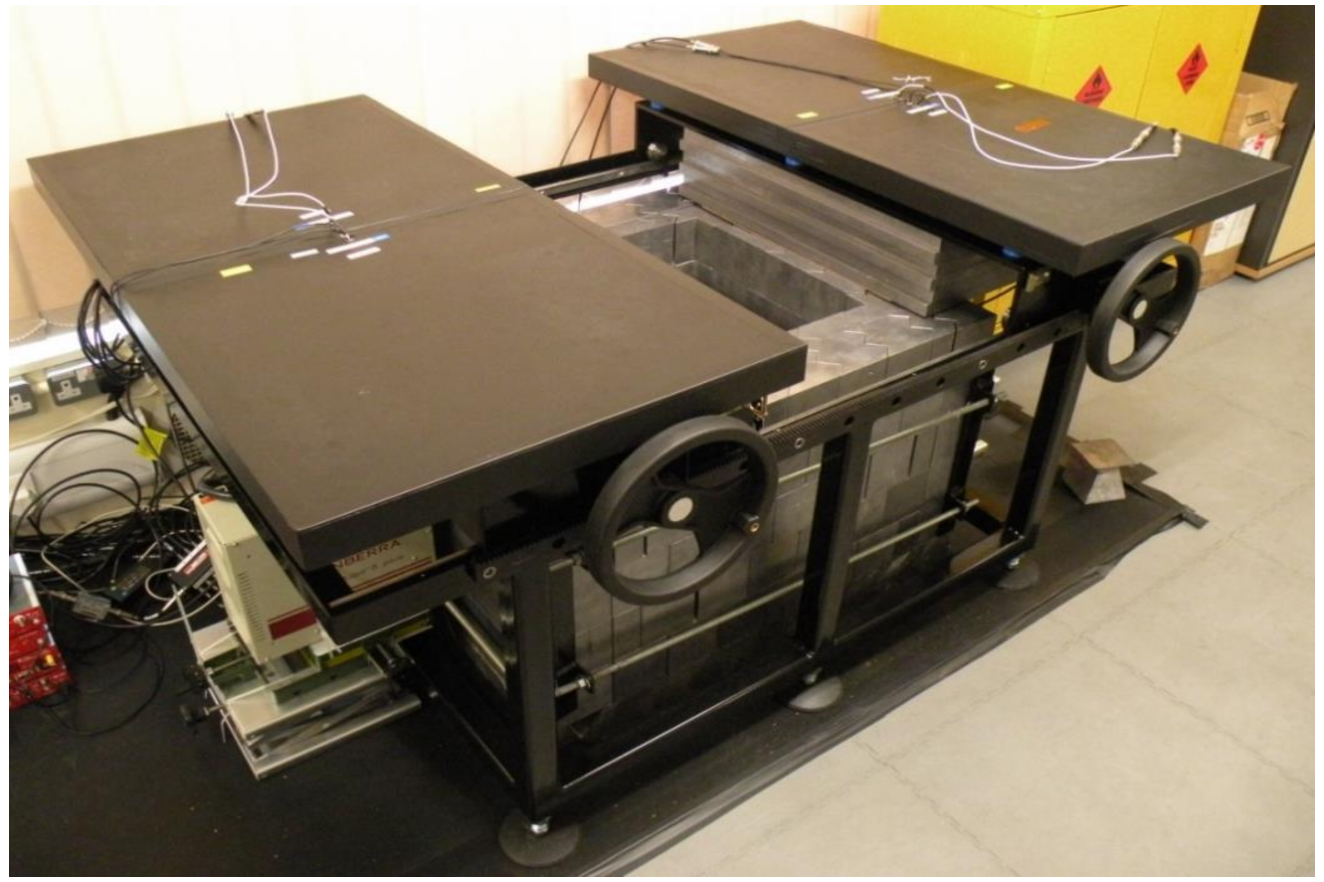


Examples of systems at AWE and PNNL

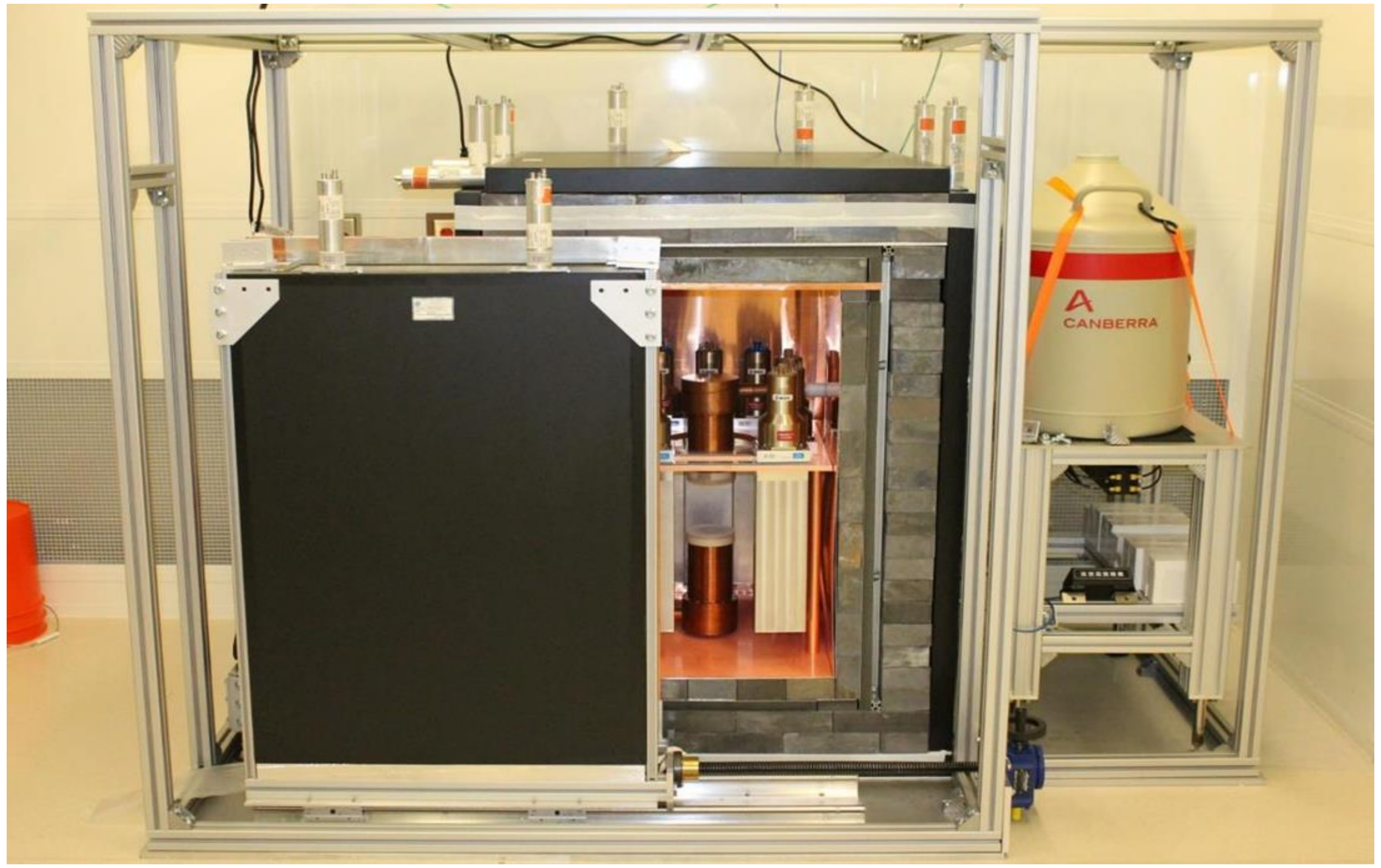
All systems developed based on Dual Broad Energy HPGe detectors



First prototype at AWE used to validate Geant4 modelling



Current prototype at AWE used to validate multi detector geometry configuration (minus Compton Suppression detectors)



Operational system located in PNNL shallow underground laboratory:

Advanced Radionuclide Gamma-spectrOmeter (ARGO)

Two Broad energy Germanium BEGe detectors surrounded by 12 Sodium iodide detectors

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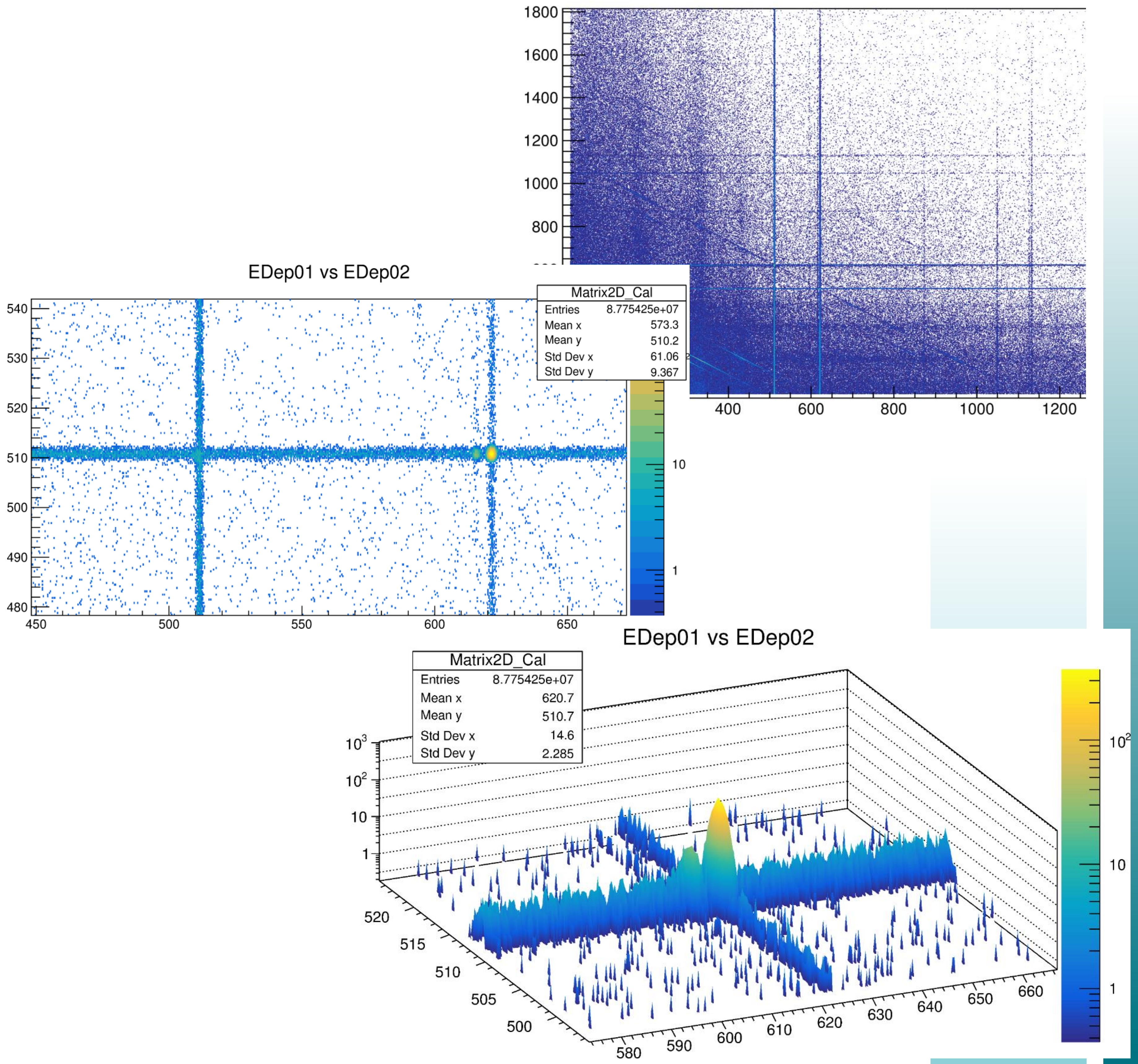
Validation of ARGO detector system and processing software

- CTBT relevant nuclides produced by irradiation of natural uranium using a Thermo Scientific neutron generator at PNNL

- Two day acquisition undertaken on ARGO – 44 minutes after end of irradiation

- List mode data was collected using a Canberra Lynx Digital signal Processors with custom developed acquisition software for 14 detectors

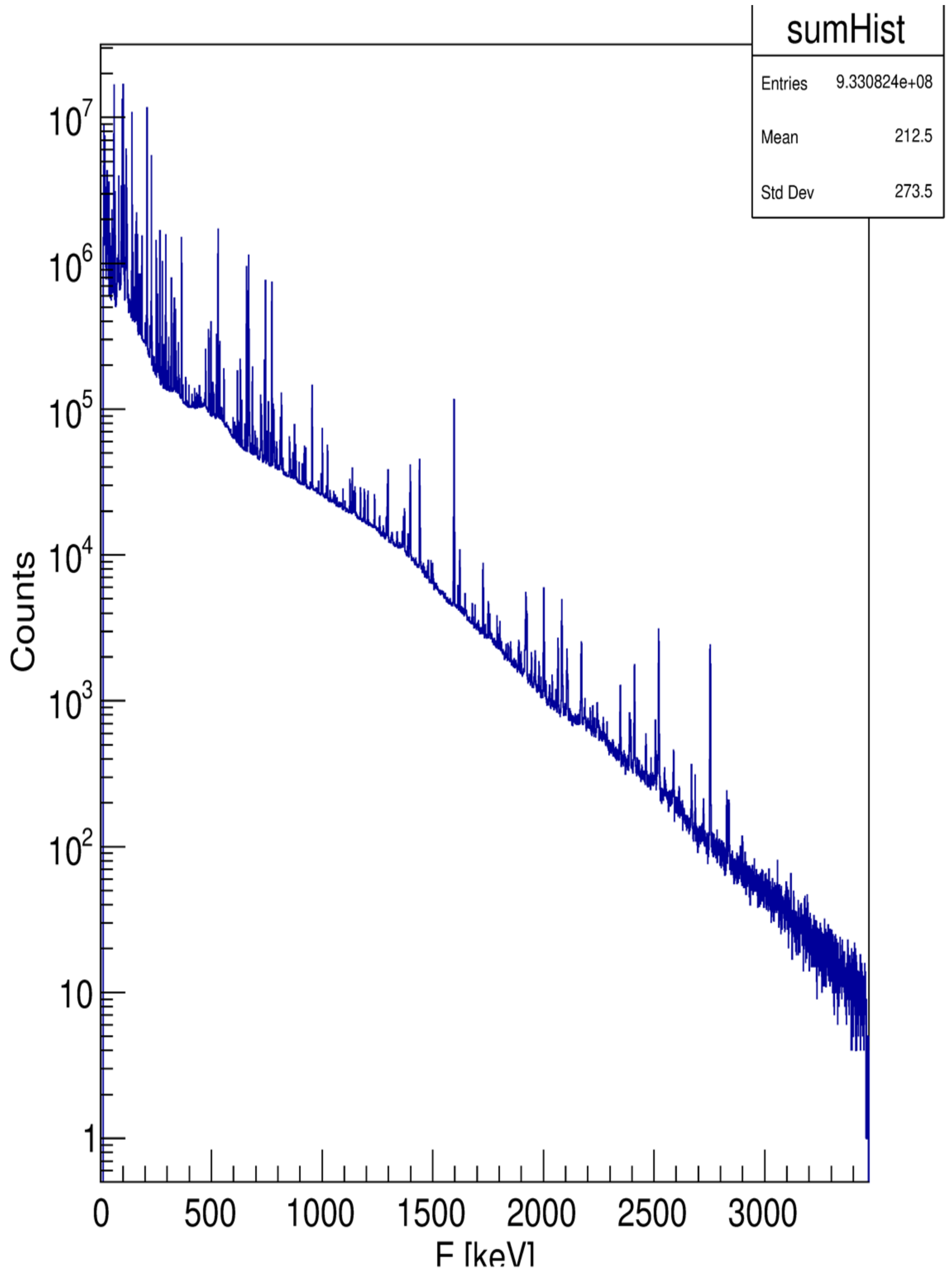
- Data was processed using a custom package (C++/ROOT based) sorts and builds gain matched calibrated singles histograms, coincidence histograms, anti-coincidence histograms, coincidence matrices, energy vs time matrices… etc



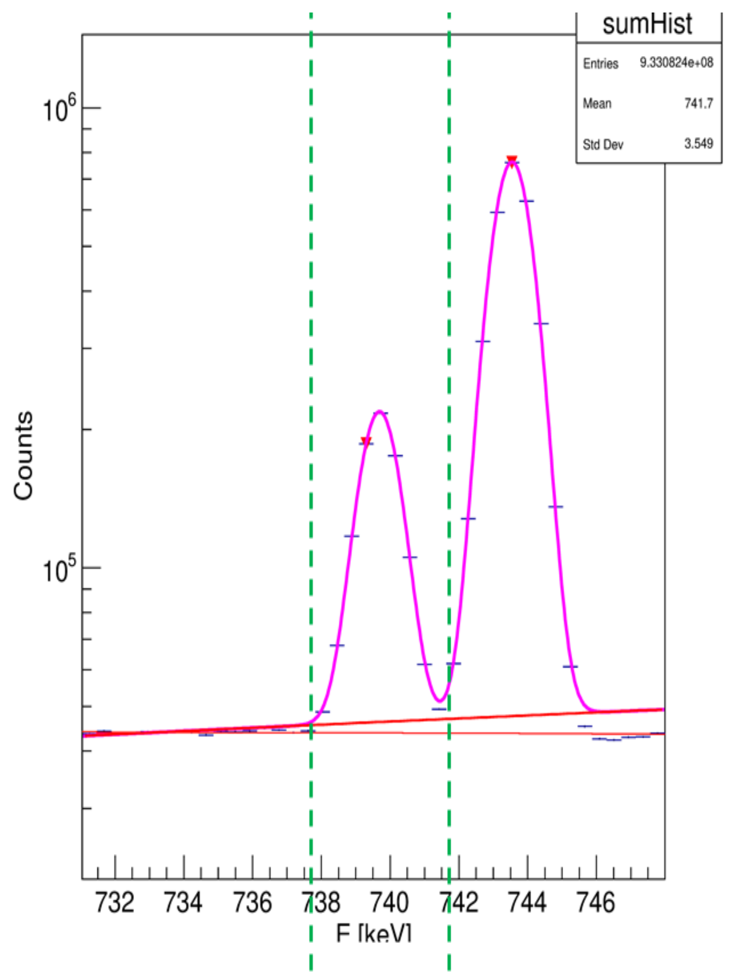
Validation example 1 – ^{99}Mo 181 / 739 keV Coincidence Gating

Validation of coincidence methods vs standard singles

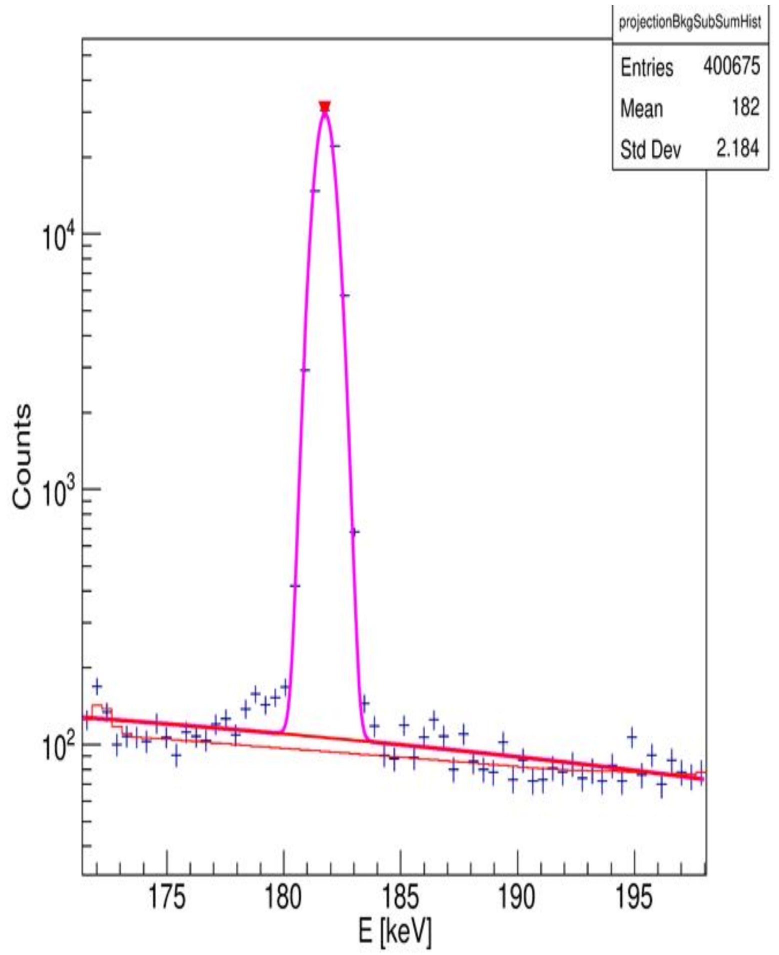
Sum spectra of HPGe1 and HPGe2



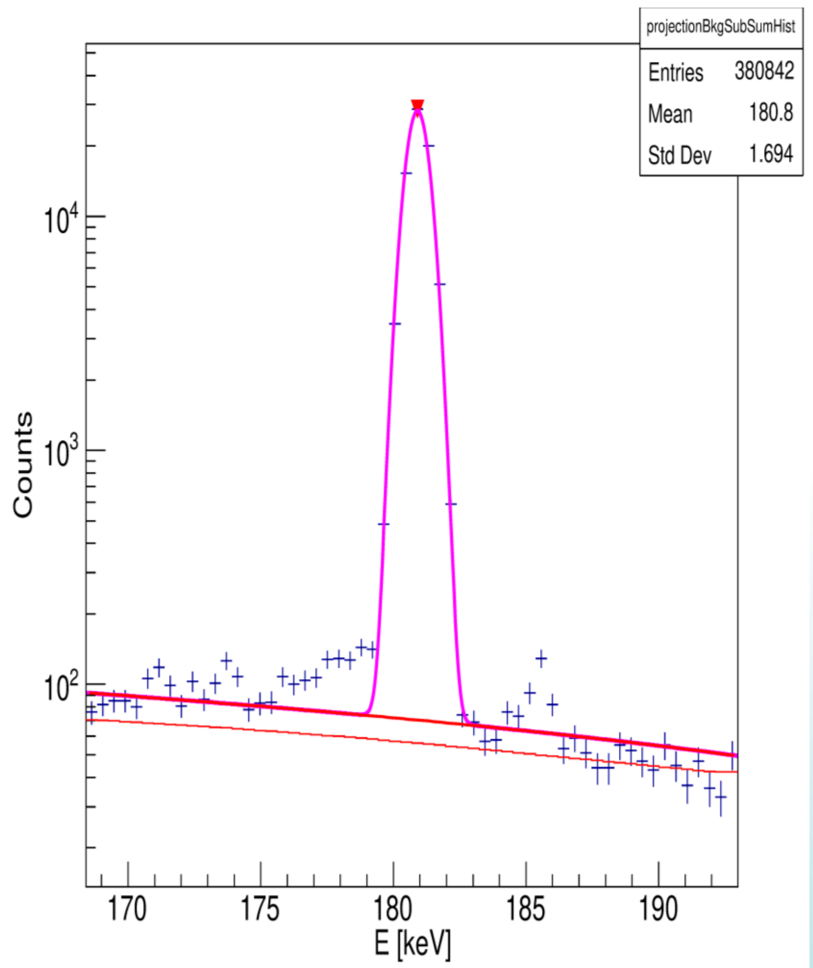
Coincidence gate
99Mo 739 keV



Extracted Spectra
181keV
(γ - γ)



Extracted Spectra
181keV
(Compton supressed γ - γ)



Mo-99	Bq	+/- (Bq)	% difference from singles
Traditional Singles (739keV)	710	45	
Coincidence (Gating on 739 extracting 181 keV)	702	105	-1.13
Coincidence (Gating on 739 extracting 181 keV) compton supressed	690	104	-2.82

Validation example 2 – ^{140}La and ^{134}Cs

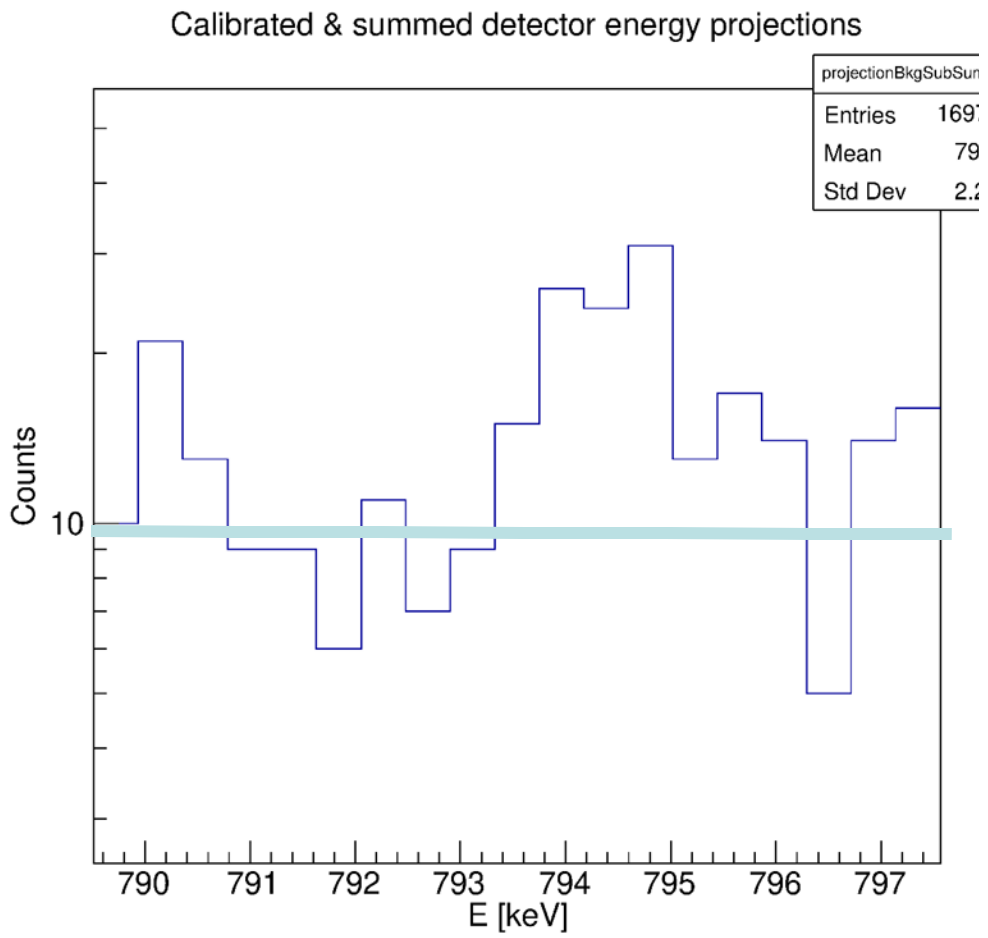
Its important to review nuclear data before choosing coincidence pairs for Compton suppressed γ - γ

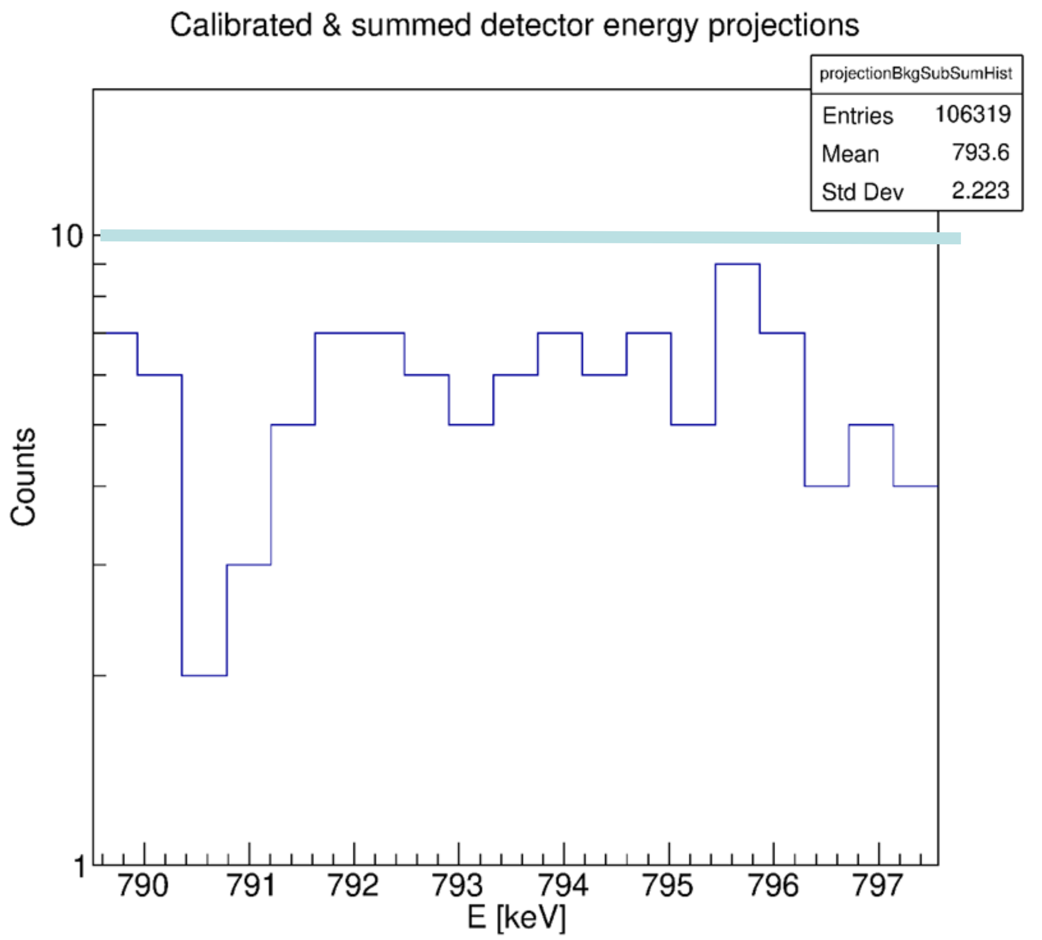
La-140	Bq	+/- (Bq)	% difference from singles
Traditional Singles (1596keV)	166	7	
Coincidence (Gating on 925 extracting 1596 keV)	160	24	-3.61
Coincidence (Gating on 925 extracting 1596 keV) compton supressed	156	23	-6.02

Cs-134 MDC	Bq	+/- (Bq)	% difference from singles
Traditional Singles	100		
Coincidence (Gating on 604 extracting 795 keV)	47		-53.00
Coincidence (Gating on 604 extracting 795 keV) compton supressed	33		-67.00

Gate	Extracted	Maximum gamma's in cacscade	Coincidence / coincidence (Compton suppressed)
925	1596	2	1.03
867	1596	2	1.02
432	487	3	1.67
815	1596	4	1.02
328	1596	5	2.24
329	487	5	1.69
487	1596	6	1.35

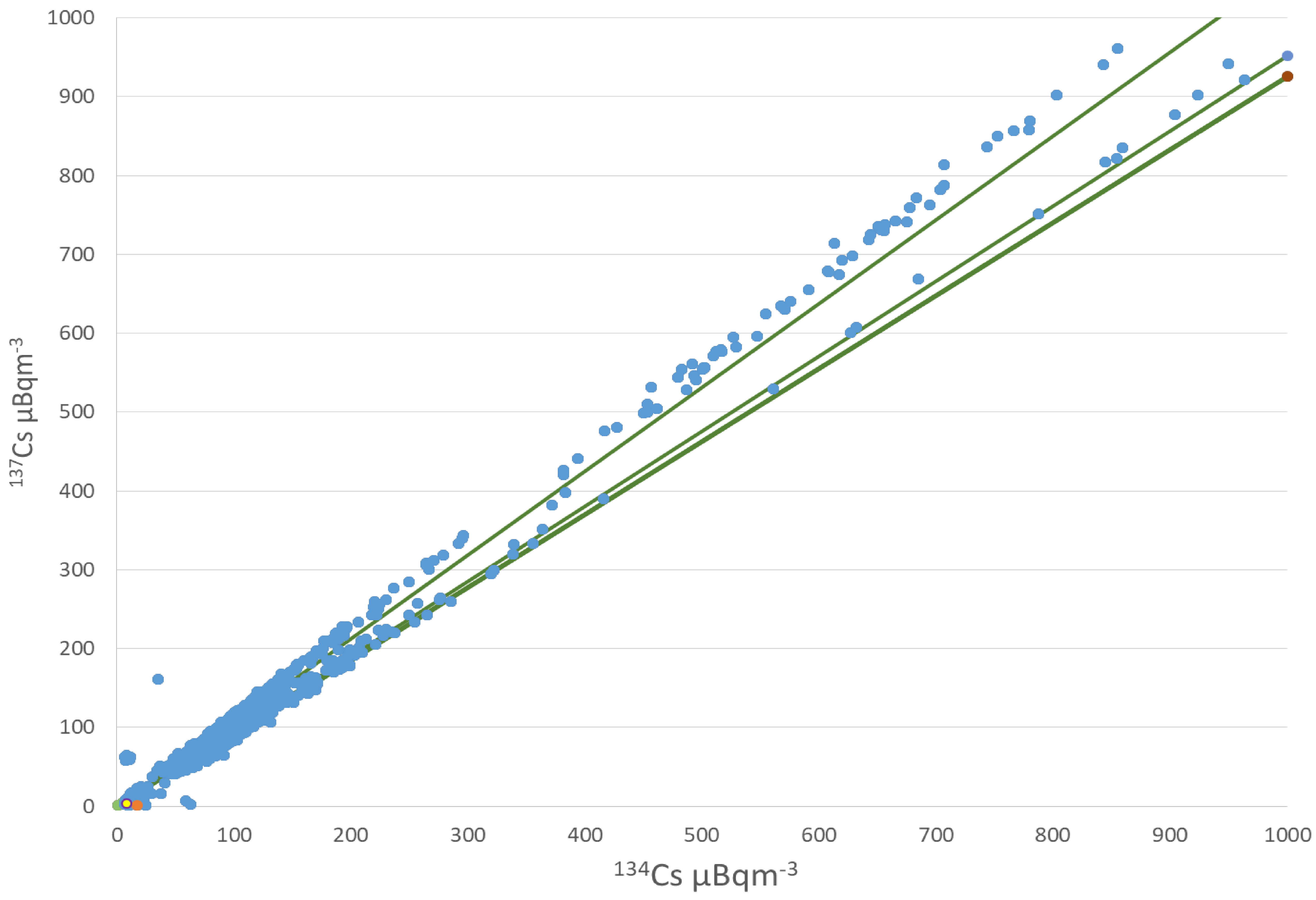
This signature would be expected to be a poor candidate, however the 815 keV and 1596 keV cascade occur 22% of the time and are only seen in coincidence with two additional emissions 0.015%





γ – γ Coincidence Examples: $^{134/137}$Cs ratio at JPP38

- The $^{134/137}$Cs ratio is well known from the releases from Fukushima Daiichi accident.
- Expected excess ^{137}Cs can be explained by resuspension of earlier Chernobyl / nuclear test debris
- Measurement with excess ^{134}Cs could be due to fresh releases when decay corrected back to Fukushima Daiichi accident.

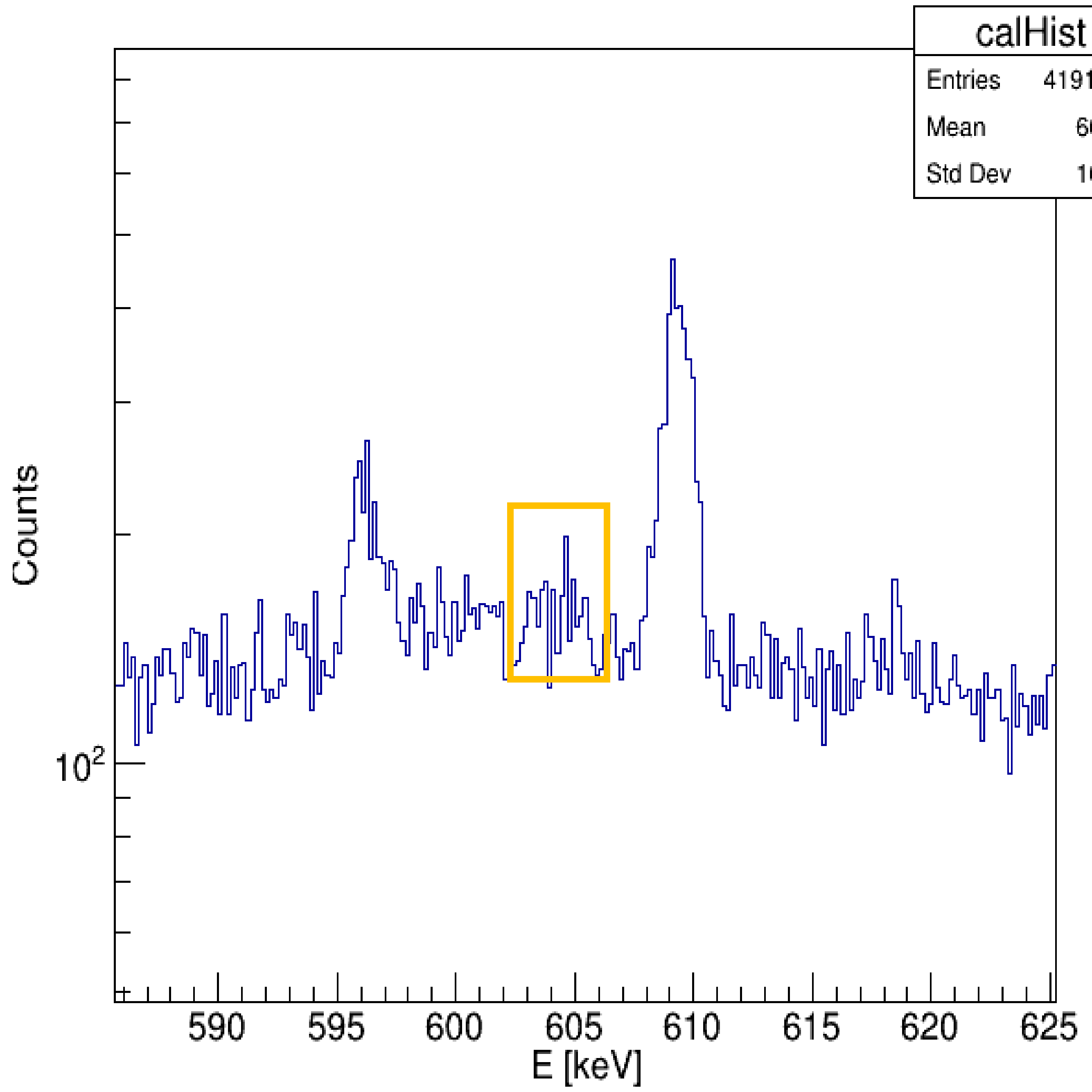


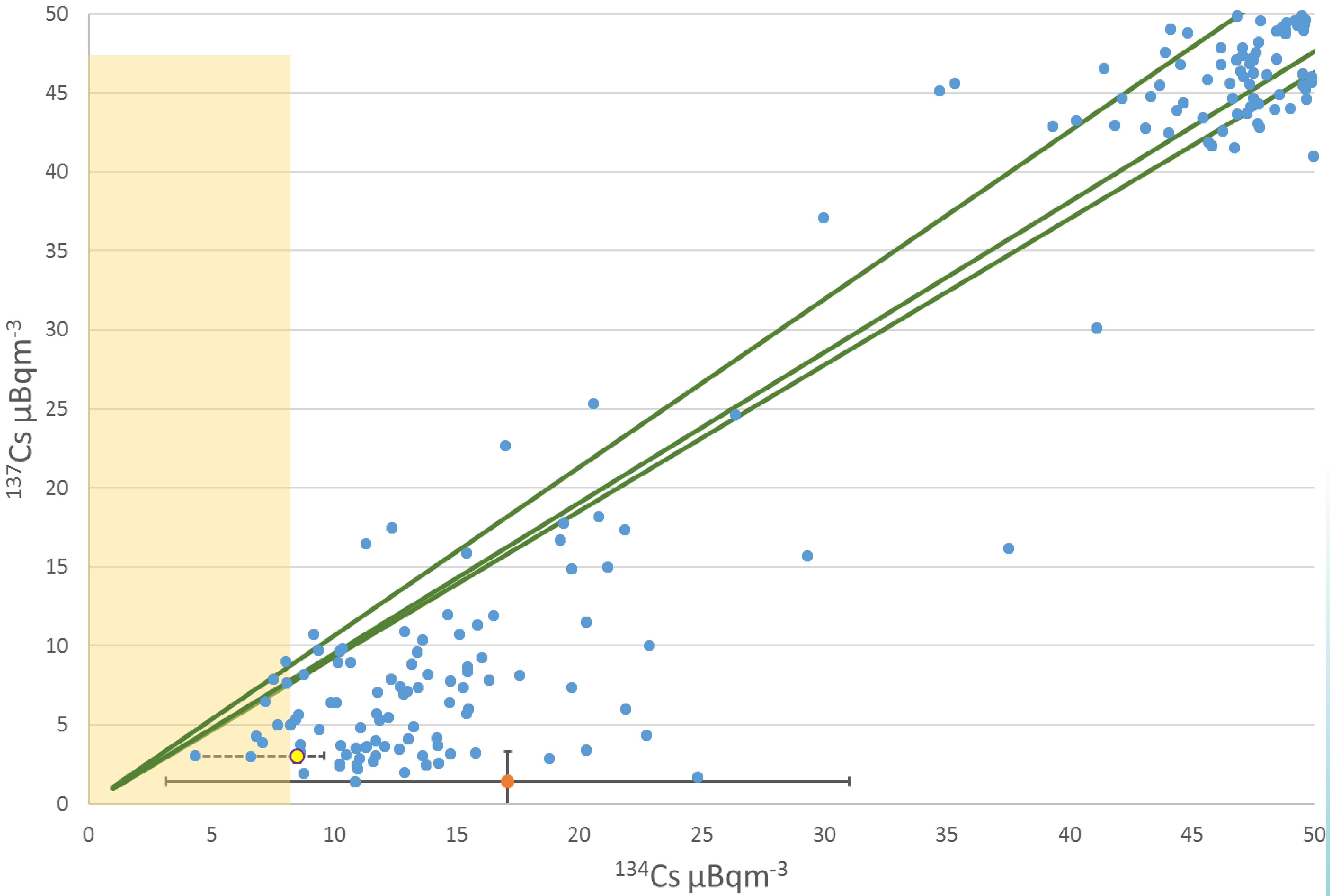
Green - Estimation of fuel compositions in Fukushima-Daiichi nuclear power plant. JAEA-Data/Code 2012-018, Japan Atomic Nishihara, K., Iwamoto, H. and Suyama, K. (2012)

Blue – IMS detections

γ – γ Coincidence Examples: $^{134/137}$Cs ratio at JPP38

Laboratory measurement at GBL15 close to MDA and still excess ^{134}Cs



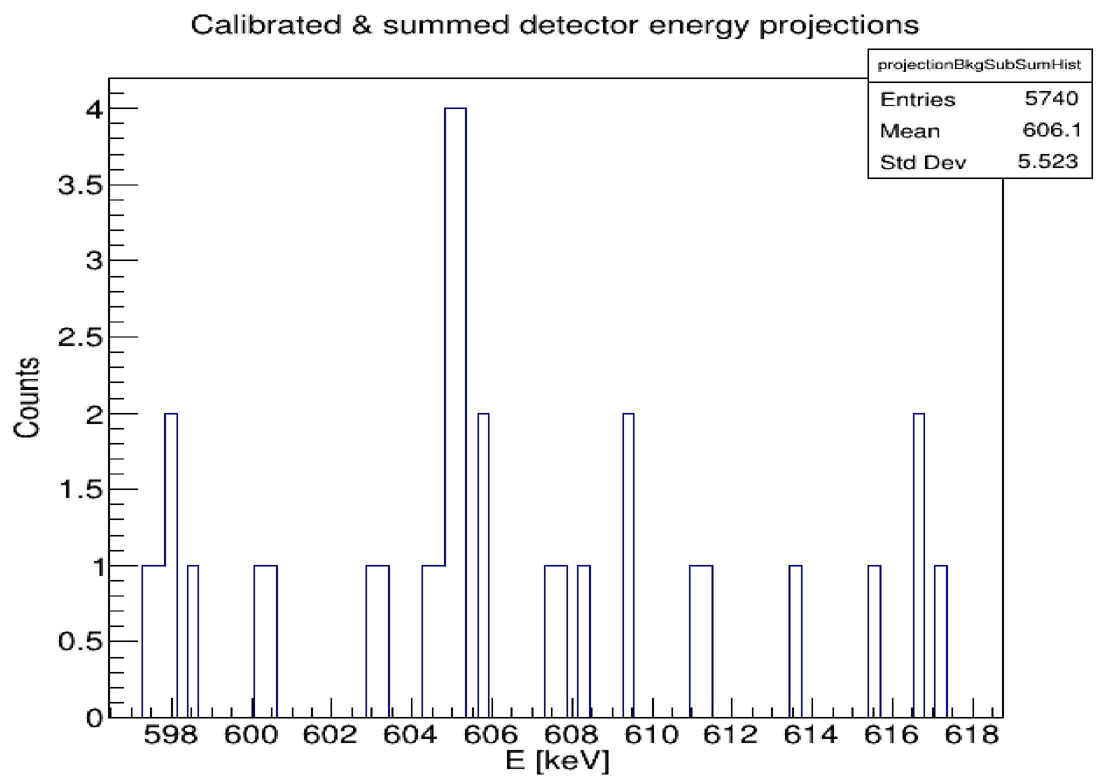


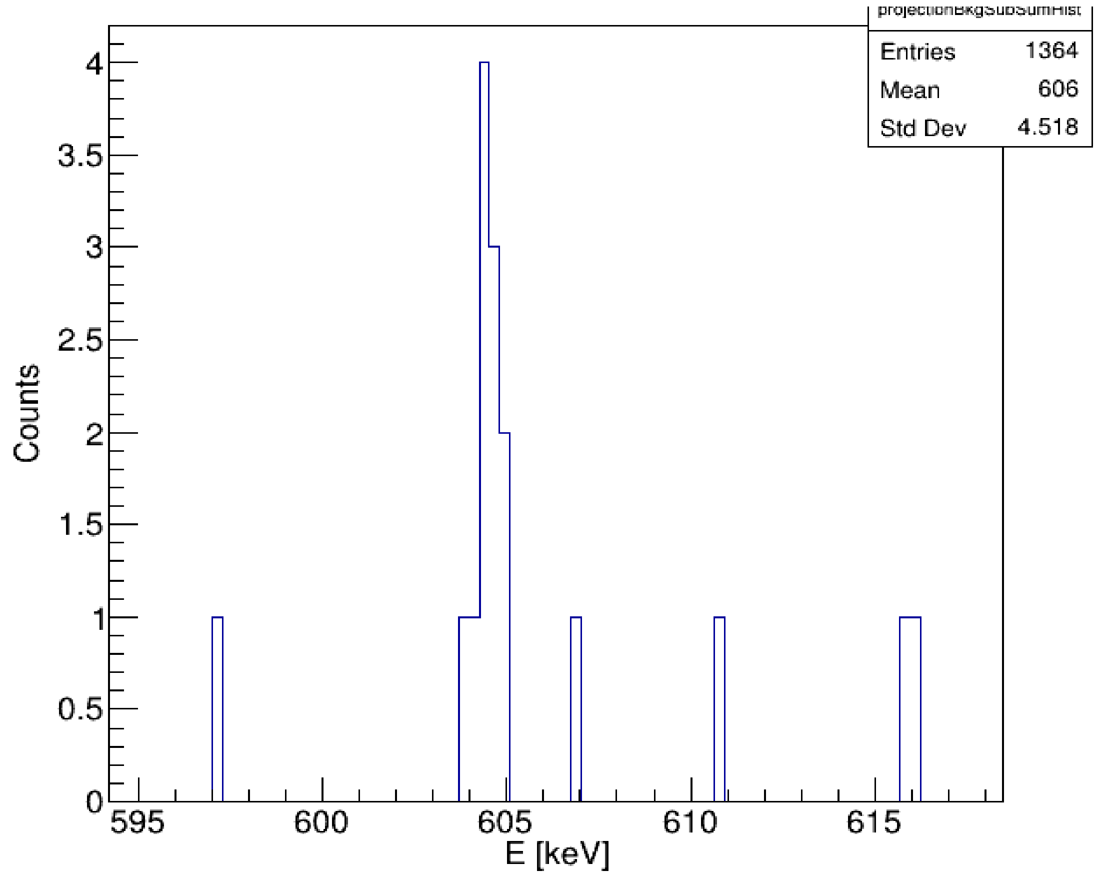
Green - Estimation of fuel compositions in Fukushima-Daiichi nuclear power plant. JAEA-Data/Code 2012-018, Japan Atomic Nishihara, K., Iwamoto, H. and Suyama, K. (2012)

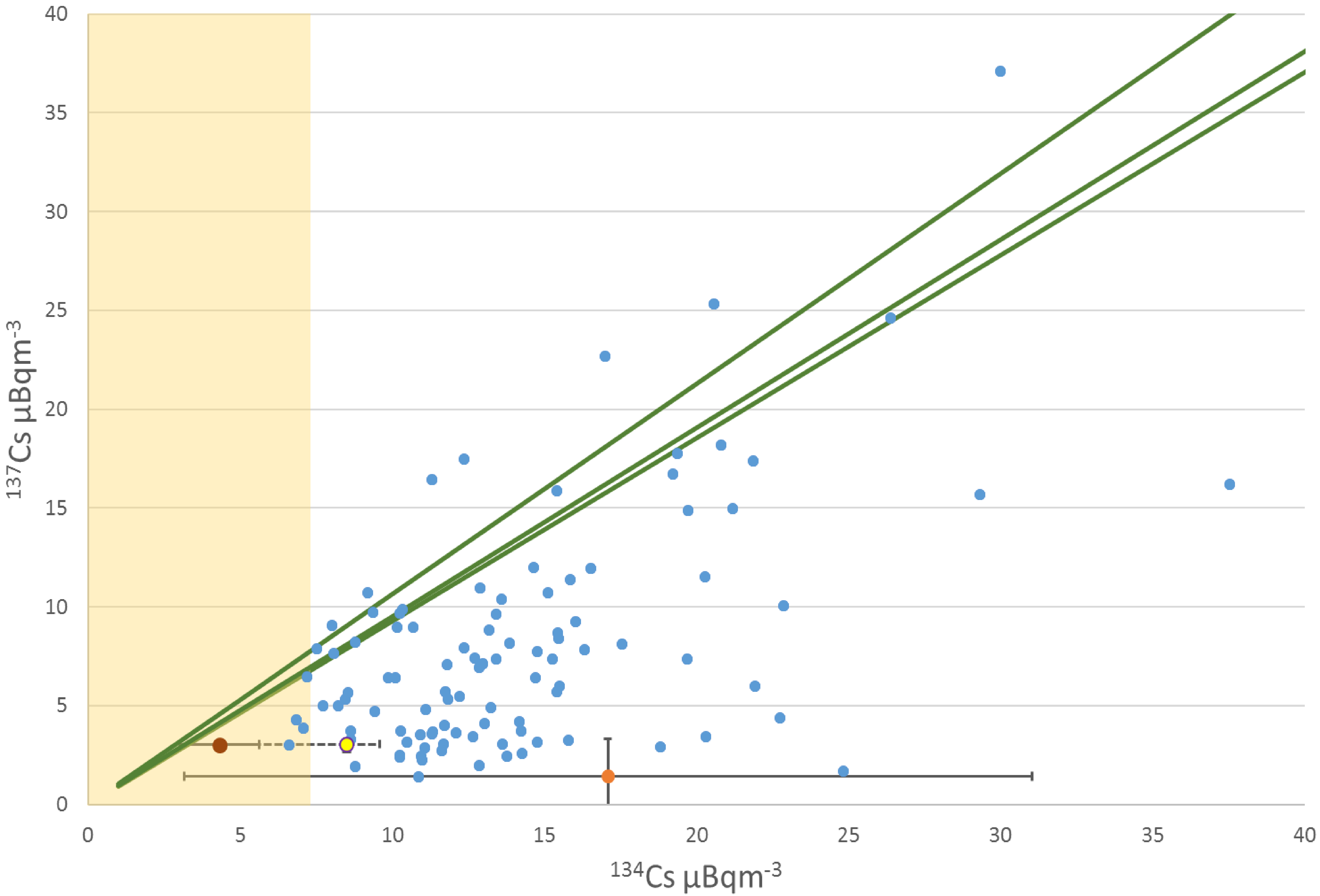
Blue / Orange – IMS detections

Yellow – Measurement at GBL15 using certified detector + Cosmic Veto

γ – γ Coincidence Examples: $^{134/137}$Cs ratio at JPP38







Measurement using Advanced Gamma system at GBL15 in standard coincidence and Compton suppressed coincidence modes are much closer to the theoretical decay corrected $^{134/137}$Cs ratio

γ – γ Coincidence Examples: $^{140}Ba/^{140}La$ in JPP37 (May 2010)

- A variety of opinions and hypothesis for the origin of these detections have been expressed
- What if this happened now and "special event analysis" was acceptable using Advanced gamma spectrometry at laboratories initiated by States Party / NDC requests
- Three scenarios were modelled and the concentration determined at each of the 80 stations (only $^{140}Ba/^{140}La$ were considered, not Xe detections) based on published parameters (ref 5)

Scenario 1:

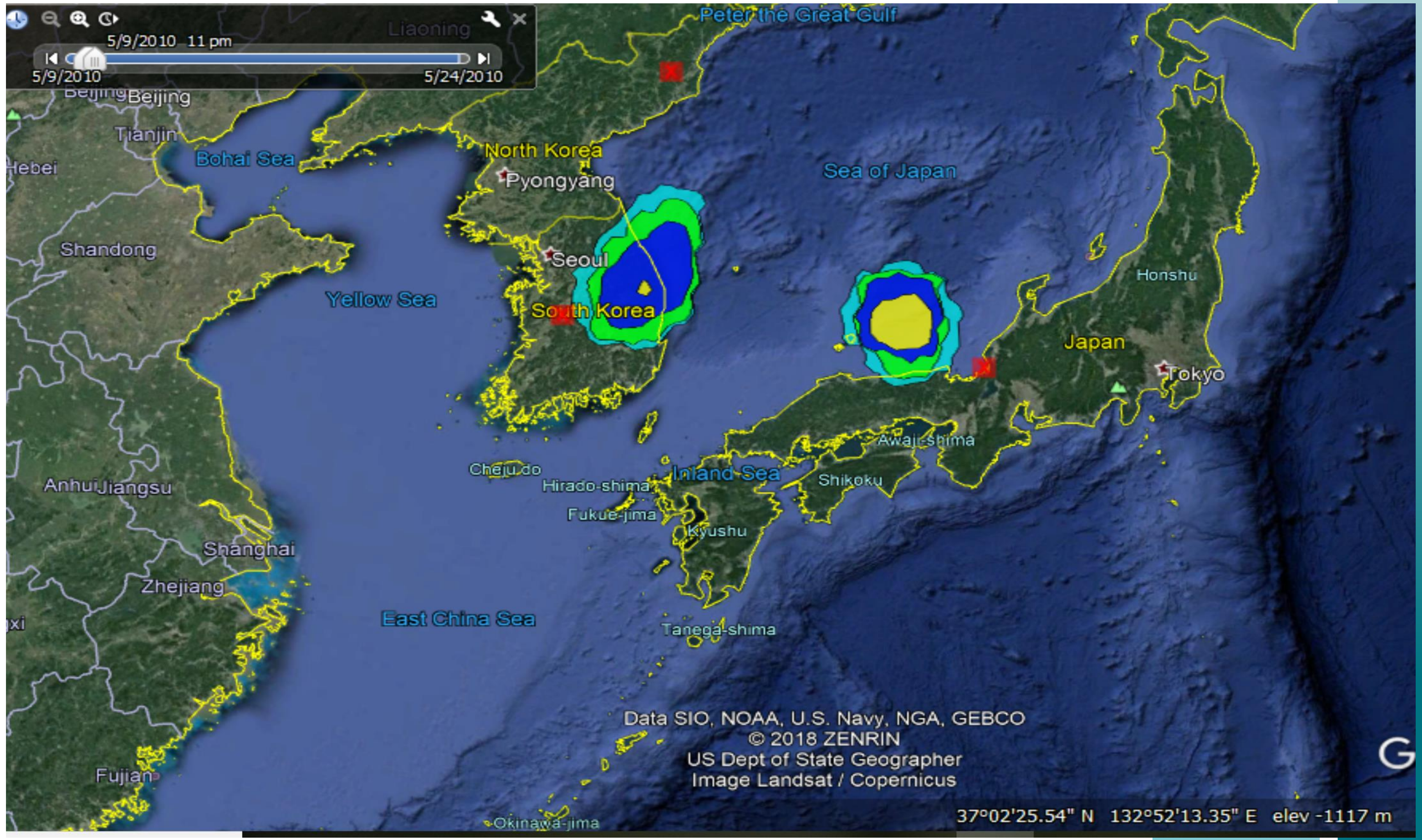
Release from North Korean test site beginning at 0600 UTC 11 May 2010

Scenario 2:

Release from Monju fast breeder reactor

Scenario 3:

Release from the 30 MW$_{(th)}$ research reactor HANARO, and associated radio-isotope production facility



γ – γ Coincidence Examples: ^{140}Ba/La in JPP37 (May 2010)

- Concentrations of ^{140}Ba were compared with detection limits of:
  - **IMS stations** – MDA at 2010 if operational or current MDA
  - **RN Laboratories** – MDA of labs used for JPP38 measurements in 2010
  - **Advanced γ spectrometry systems** (with and without Compton Suppression)

DATE	P37	P38	P44	P58	P74	P76	P79
16 May 2010	8.186E-05						
17 May 2010	2.266E-05						
18 May 2010	2.750E-05	6.763E-05		3.554E-07			
19 May 2010	2.811E-05	2.028E-05	2.056E-10	1.180E-05			
20 May 2010	5.081E-05	2.741E-06	1.349E-07	5.988E-07			
21 May 2010	4.379E-05	8.101E-08	4.293E-07		4.875E-08		
22 May 2010	5.240E-06				3.231E-08	2.832E-08	
23 May 2010	5.000E-06	2.189E-06			2.730E-07	1.239E-06	8.813E-07

Underground nuclear explosion from DPRK test site

- IMS station detections - **8**
- RN lab detections - **9**
- Advanced γ spectrometry - **18**
- **5** additional stations to aid in location analysis

γ – γ Coincidence Examples: ^{140}Ba/La in JPP37 (May 2010)

- Concentrations of ^{140}Ba were compared with detection limits of:
  - **IMS stations** – MDA at 2010 if operational or current MDA
  - **RN Laboratories** – MDA of labs used for JPP38 measurements in 2010
  - Advanced gamma-spectrometry systems (with and without Compton Suppression)

DATE	P37	P38	P44	P57	P58	P60	P74	P79	P20	P22
16 May 2010	8.186E-05								6.814E-08	
17 May 2010	2.266E-05	1.025E-06			1.894E-06				1.986E-04	
18 May 2010	2.750E-05	1.184E-03			1.544E-03				1.176E-03	
19 May 2010	2.811E-05	1.350E-04			5.408E-04				1.827E-06	
20 May 2010	5.081E-05				1.505E-05					
21 May 2010	4.379E-05	3.838E-06			9.141E-06					
22 May 2010	5.240E-06	3.128E-06			3.539E-06	9.476E-07		2.710E-07		
23 May 2010	5.000E-06	2.295E-05	8.536E-07		1.876E-06	1.506E-06		6.136E-08	5.484E-07	
24 May 2010	2.116E-07	6.209E-07	3.567E-09	4.110E-07	1.641E-06	2.527E-07	5.412E-08			5.243E-07

Release from Monju

- IMS station detections - **13**
- RN lab detections - **15**
- Advanced γ spectrometry - **33**
- **5** additional stations to aid in location analysis

γ – γ Coincidence Examples: ^{140}Ba/La in JPP37 (May 2010)

- Concentrations of ^{140}Ba were compared with detection limits of:
  - **IMS stations** – MDA at 2010 if operational or current MDA
  - **RN Laboratories** – MDA of labs used for JPP38 measurements in 2010
  - Advanced gamma-spectrometry systems (with and without Compton Suppression)

DATE	P37	P38	P14	P20	P44	P58	P70	P71	P74	P76
16 May 2010	8.186E-05	2.034E-05								
17 May 2010	2.266E-05									
18 May 2010	2.750E-05	3.843E-08				1.275E-06				
19 May 2010	2.811E-05	7.820E-06		1.288E-05				4.418E-05		
20 May 2010	5.081E-05	8.858E-05		7.638E-05		3.486E-05		2.935E-05		6.522E-07
21 May 2010	4.379E-05	5.519E-05		1.185E-07		5.473E-05		1.887E-06		2.766E-07
22 May 2010	5.240E-06	1.129E-06	6.600E-07		1.658E-07	9.916E-07	1.700E-07		6.302E-08	
23 May 2010	5.000E-06	3.578E-07	5.151E-07		1.286E-06	5.896E-07	1.721E-06		3.161E-08	4.629E-08
24 May 2010	2.116E-07	1.517E-09	1.587E-09		2.389E-08		2.932E-08		2.583E-09	2.415E-09

Release from research reactor HANARO

- IMS station detections - **14**
- RN lab detections - **16**
- Advanced γ spectrometry - **31**
- 4 additional stations to aid in location analysis

Conclusions (so far)

- A number of advanced detection systems developed / tested / operationalised
- Substantial effort invested in the understanding and analysis of high-multiplicity coincidence systems
- Software developed to operationally acquire / process / analyse large and complex datasets quickly and simply (currently used by 5+ laboratories)
- CTBTO laboratories have an important role in confirming IMS detections
- Advanced systems have the potential to offer far greater detection capability, potentially doubling the number of confirmed detections and trebling the number of detection locations per event
- Existing advanced γ spectrometry systems (co-located at Radionuclide Laboratories) have potential to support Special Studies and the URR development / production

References

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