Traceability for activity measurements in Nuclear Forensics

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Outline

- Background to traceability in radioactivity measurements, in particular regarding primary standards from NMIs.

- Example: (work in progress)
  - ^{243}Am standardisation

- Urban Vitrified Composite material

NPL Radioactivity Group

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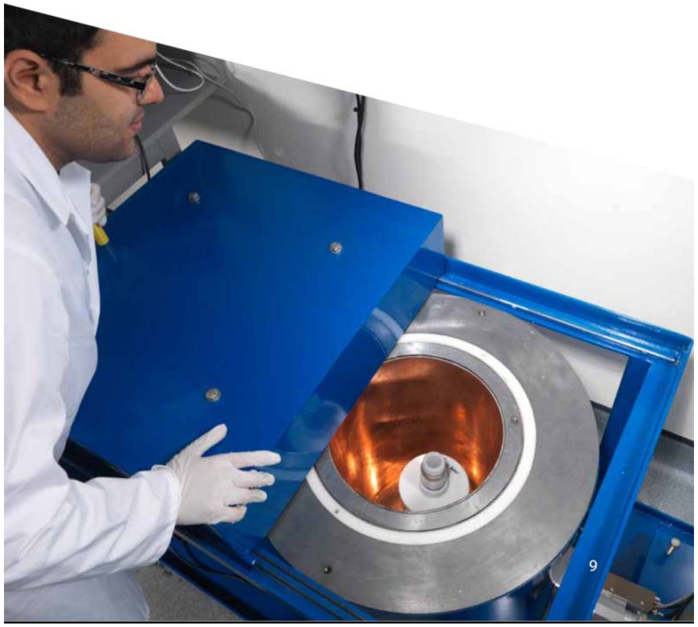
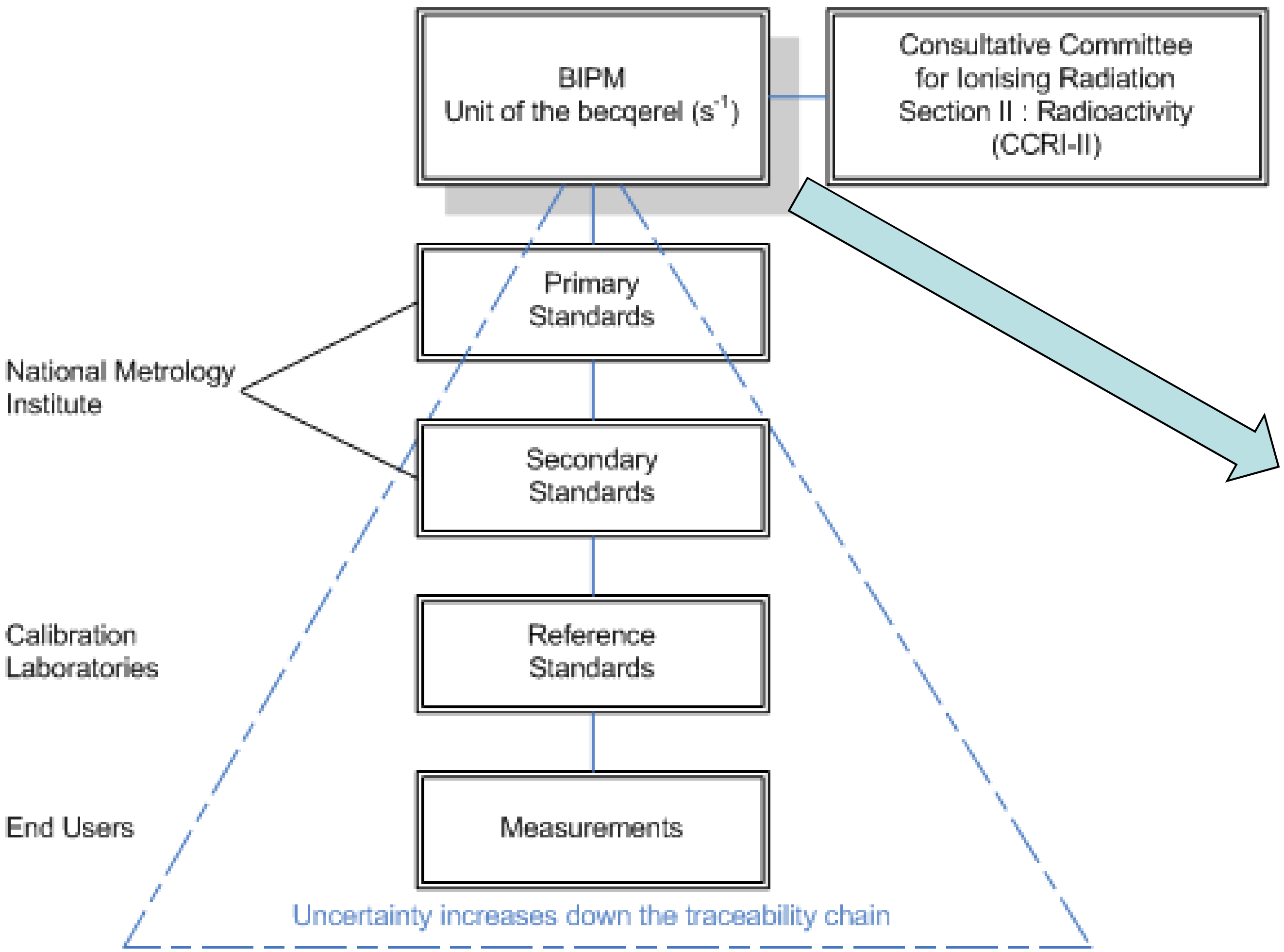
- NPL is the UK's National Metrology Institute (NMI)
- Maintain and develop UK Standards of Radioactivity
- 20 physicists, chemists and technicians
- Specialists in:
  - High accuracy radioactivity standards
  - Rare, high purity, radionuclides
  - Reference materials
  - Nuclear spectrometry
  - Radiochemistry
- For:
  - Nuclear industry
  - Nuclear medicine
  - Defence
  - Homeland Security and Nuclear Forensics



Traceability: Activity Measurements



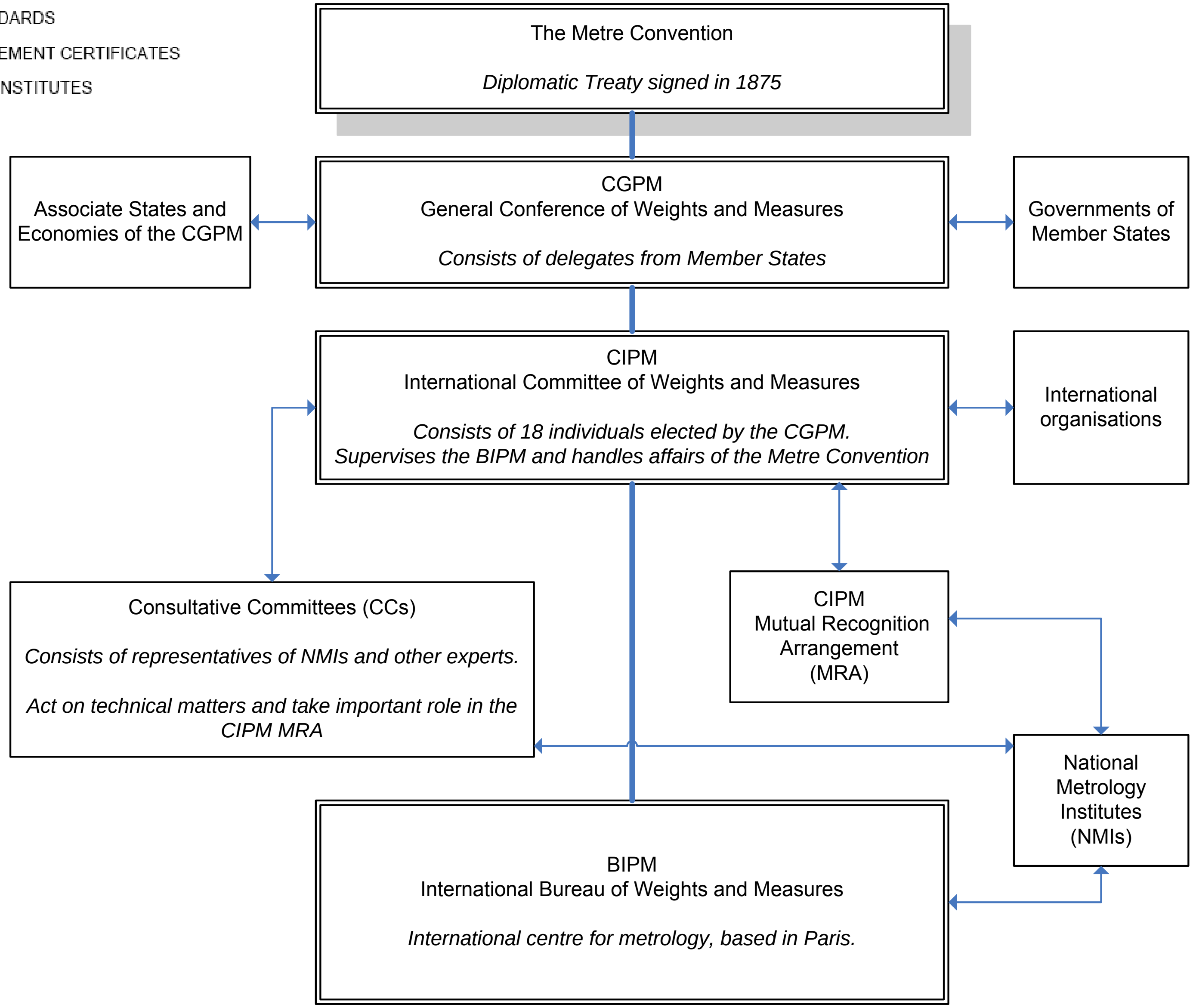
Mutual Recognition Arrangement

MUTUAL RECOGNITION

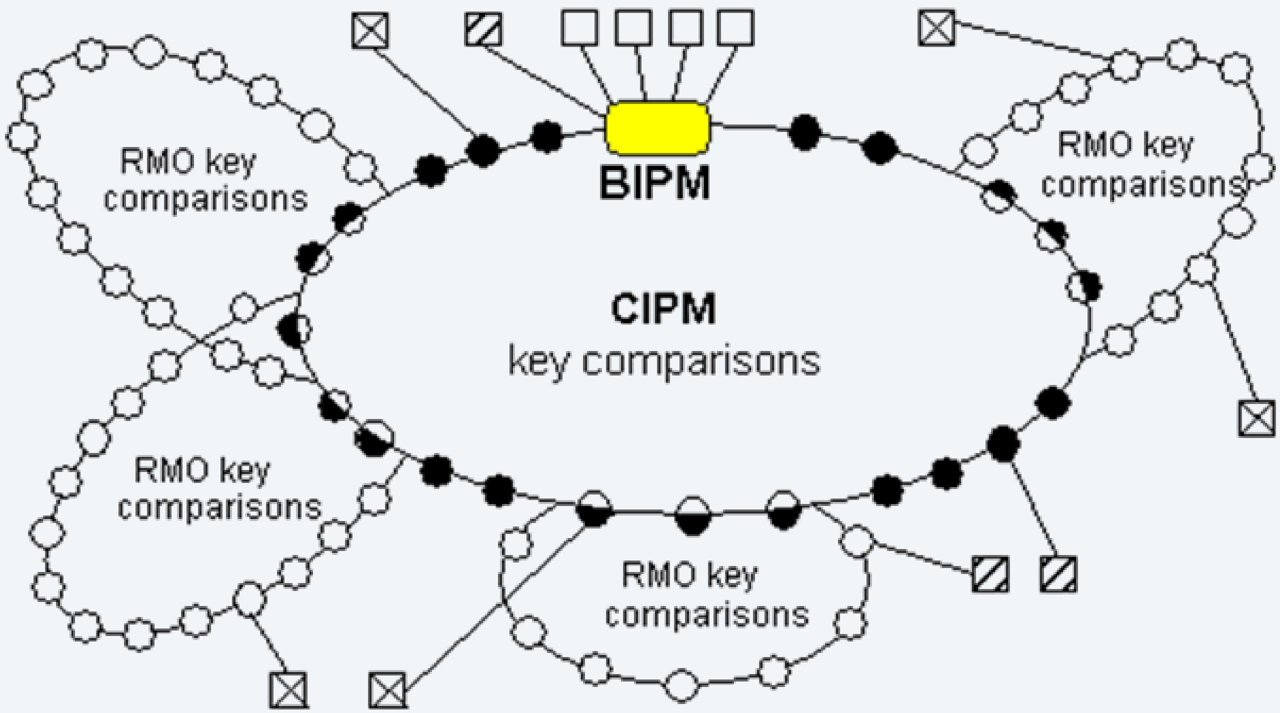
OF NATIONAL MEASUREMENT STANDARDS

AND OF CALIBRATION AND MEASUREMENT CERTIFICATES

ISSUED BY NATIONAL METROLOGY INSTITUTES

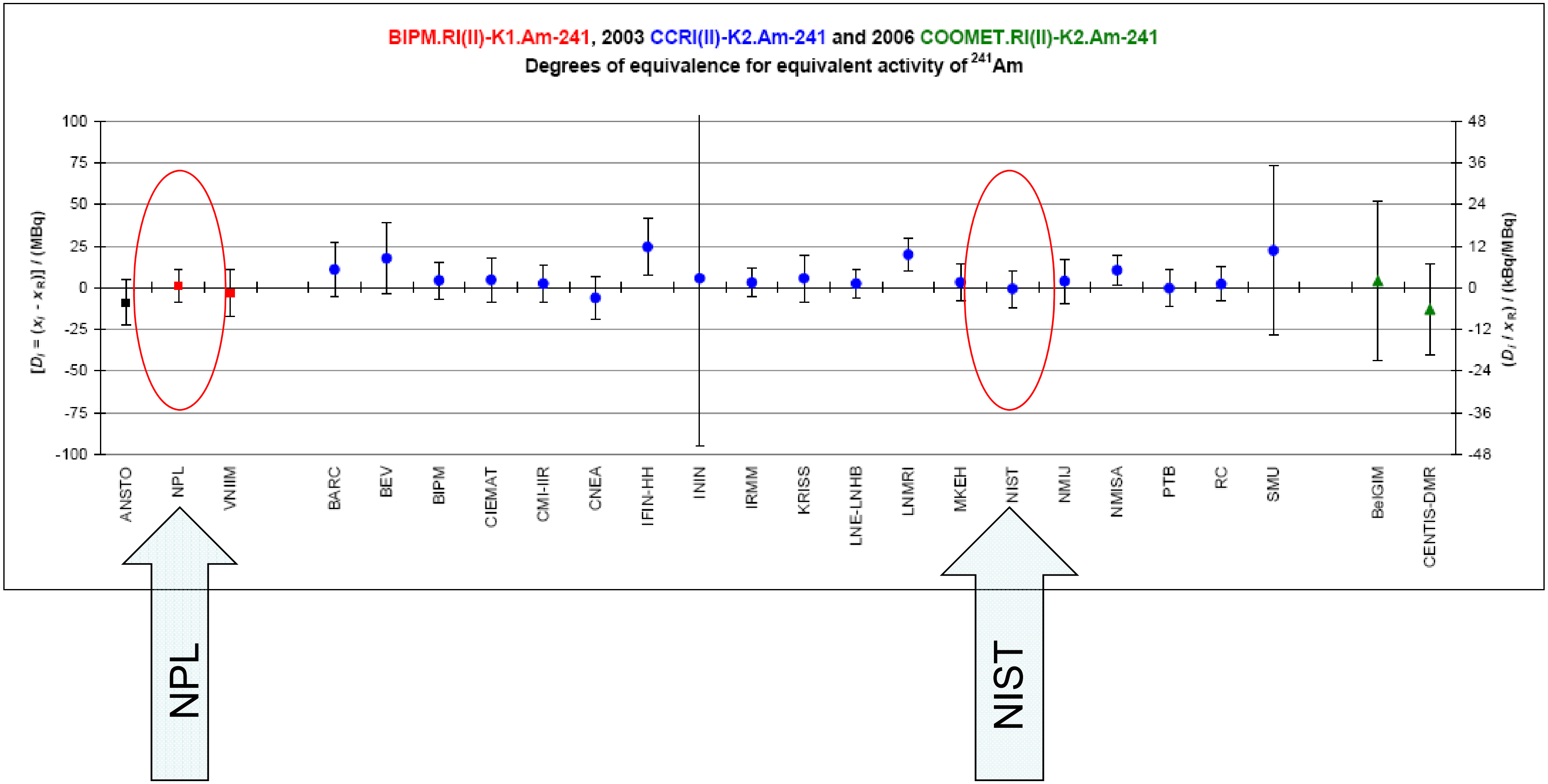


Scheme for Key Comparisons



- National metrology institute (NMI) participating in CIPM key comparisons
- NMI participating in CIPM key comparisons and in regional metrology organization (RMO) key comparisons
- NMI participating in RMO key comparisons
- NMI participating in ongoing BIPM key comparisons
- NMI participating in a bilateral key comparison
- International organization signatory to the MRA

Example : ^{241}Am



Many other comparisons

^{243}Am Standardisation

- WHY IS IT NEEDED ?
  - Yield tracer for radiochemical processes
    - α-spectrometry
    - Mass spectrometry
  - Chronometery
    - ^{243}Am – (^{239}Np) – ^{239}Pu
    - Measurement of ^{241}Pu – ^{241}Am
- Fingerprinting
  - Isotopic composition of americium

^{243}Am

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- ^{243}Am is the longest lived of Americium isotopes.
- Produced in the nuclear cycle by multiple neutron capture events on ^{239}Pu and ^{238}U. Not found in nature.
- On behalf of the US Reference Material Program, NIST (in collaboration with NPL) is seeking to prepare units of highly characterised ^{243}Am solutions.at 2 different activity concentrations.

^{243}Am

- Aim: high accuracy, high purity, ^{243}Am in solution
- Accuracy (Bq): 0.4 - 0.8%, (k = 2)
- Accuracy (mass): 0.75 - 1.10%, (k = 2)
- Purification
  - Removal of thorium, uranium, neptunium, plutonium – anion exchange in 7.2M HNO_3
  - Additional purification step to remove iron, zirconium, polonium and residual uranium, neptunium and plutonium
  - No attempt to remove curium and higher actinides

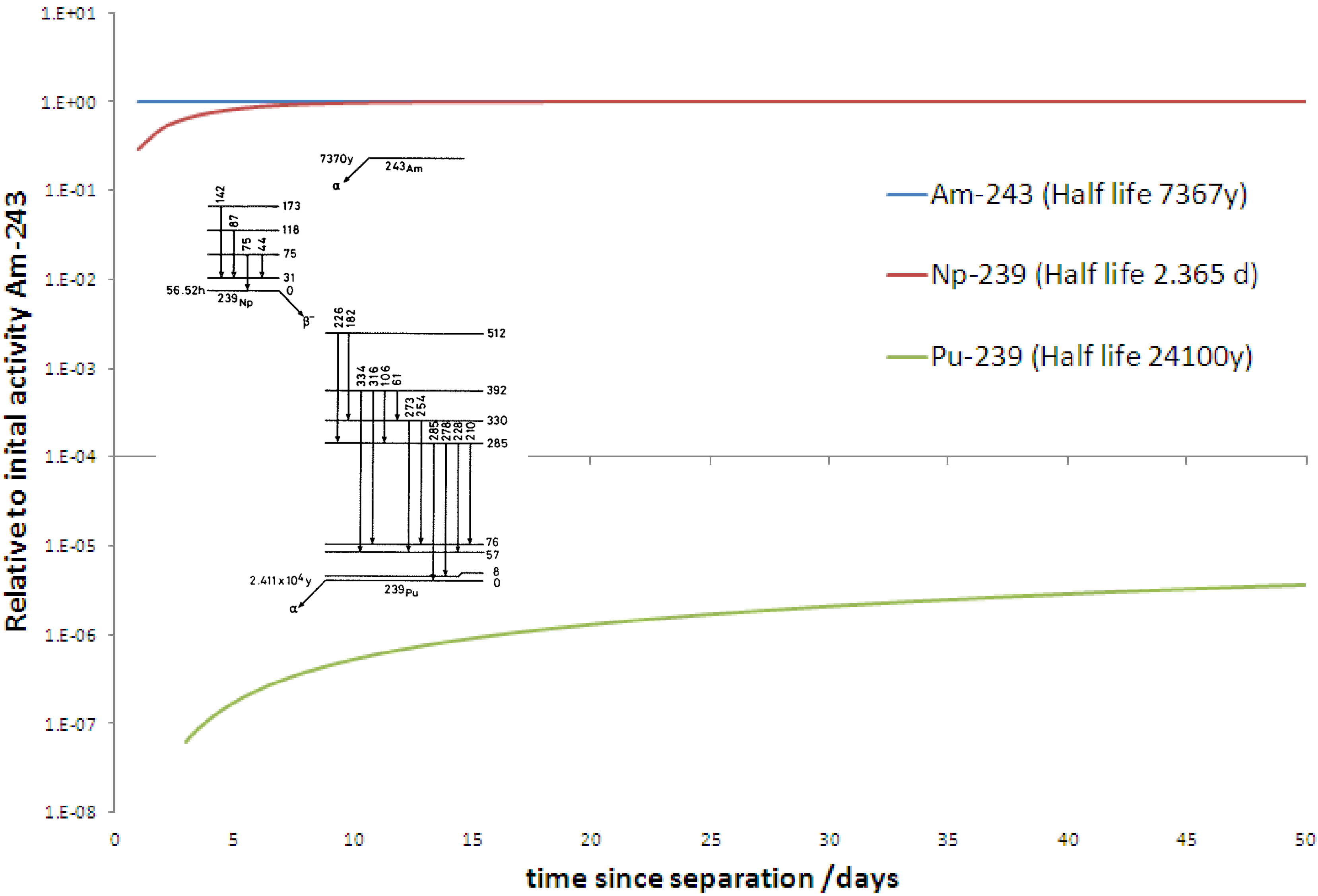
^{243}Am starting material

- Obtained by NPL in 1993 from Russian Federal Nuclear Facility : Azarmas-16)

- ^{243}Am	99.9985 %
- ^{241}Am	0.00148 (± 0.00014) %
- ^{242m}Am	0.000054 (± 0.000008) %
- $^{243/244}$Cm	0.0000034 (± 0.0000005)%

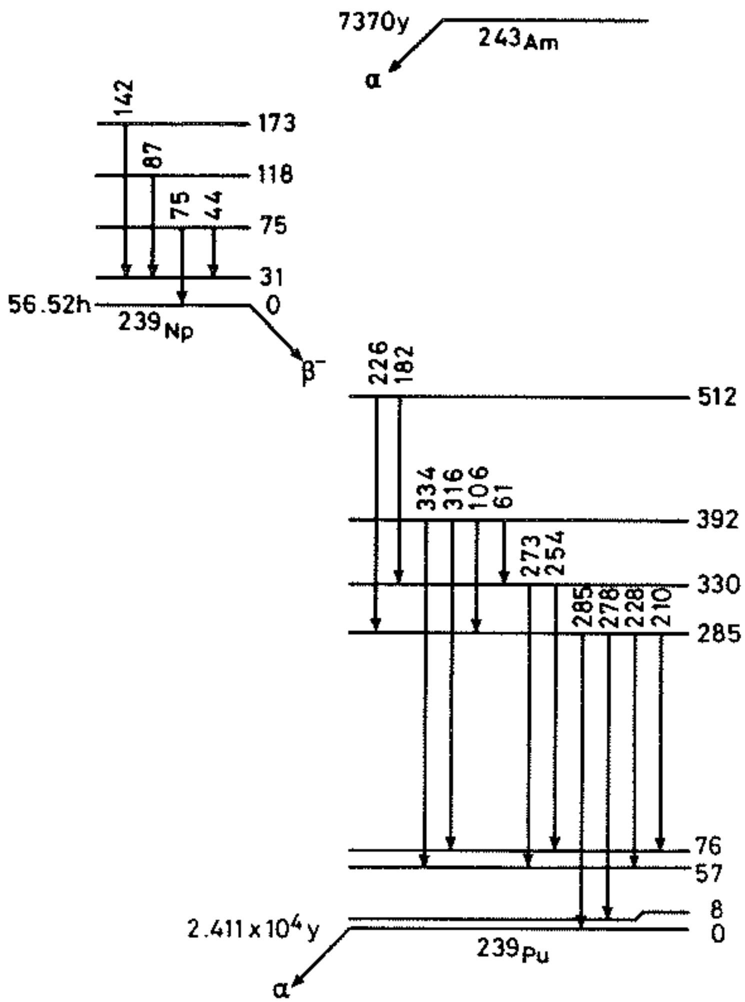
- Uncertainties (k = 1), including components for activity to mass conversion

Ingrowth of ^{239}Np and ^{239}Pu



^{243}Am : Measurement Strategy

Use a combination of various counters to monitor the efficiencies for various gamma transitions to determine absolute activity concentration:



- 4π (α+β)(LSC)–γ coincidence counting
- 4π (α+β)(HPPC)–γ coincidence counting
- 4π α(APPC)–γ coincidence counting

Combination of γ gates

α-spectrometry

Compare with ^{241}Am

^{241}Am standardised by similar techniques to high precision (approx 0.1% k =1)

Basics of 4πβ–γ coincidence method

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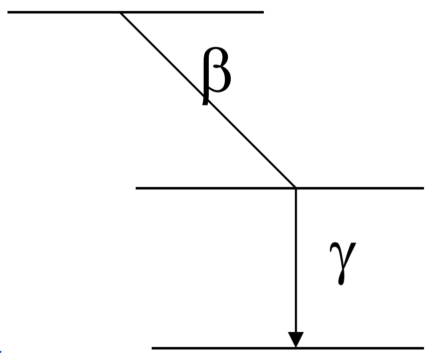
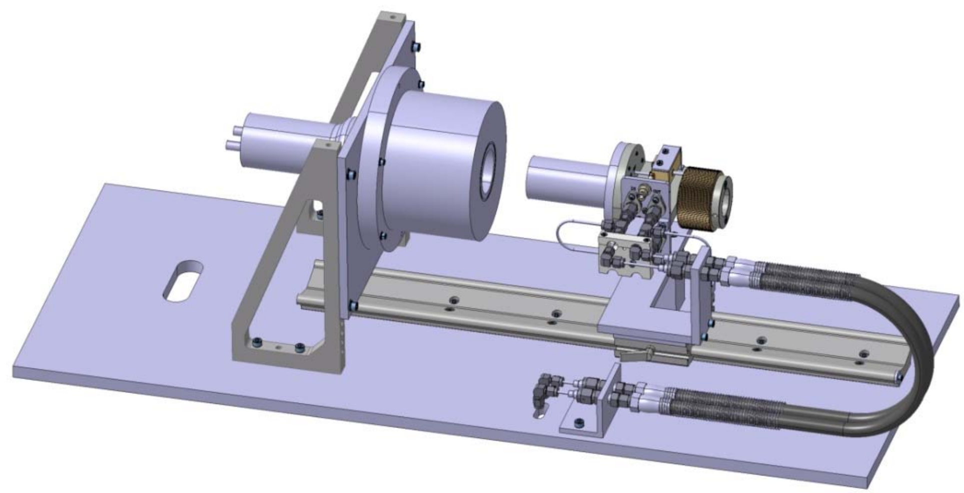
Count Rates



$N_\beta = N_o\,\varepsilon_\beta$

$N_\gamma = N_o\,\varepsilon_\gamma$

$N_c = N_o\,\varepsilon_\beta\,\varepsilon_\gamma$



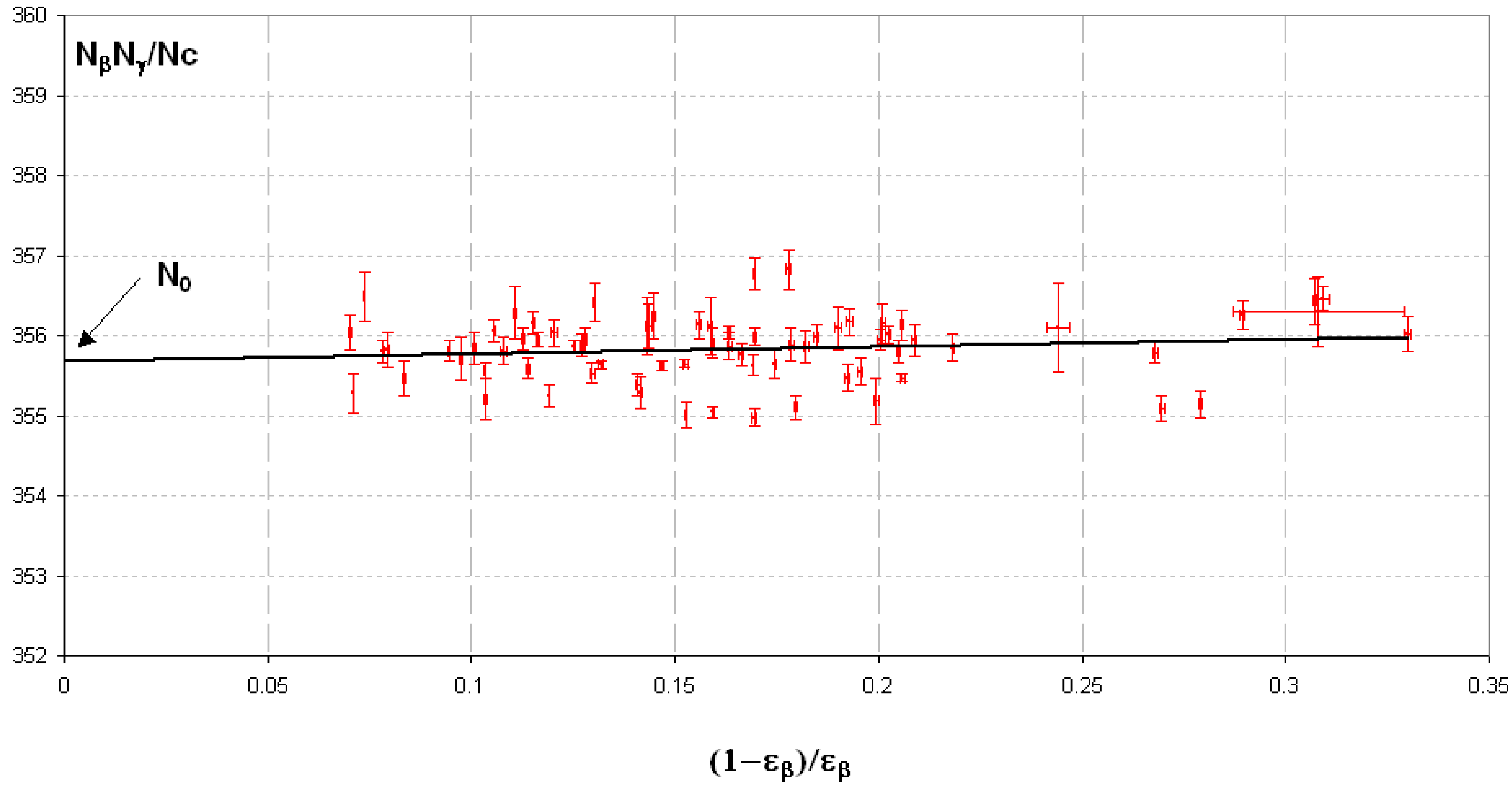
Efficiencies: $\varepsilon_\beta = N_c/N_\gamma$

$\varepsilon_\gamma = N_c/N_\beta$

$$Activity: N_o = \frac{N_\beta N_\gamma}{N_c}$$

Efficiency Extrapolation

$$\frac{N_\beta N_\gamma}{N_c} = No\left[1 + f\left(\frac{1 - \varepsilon_\beta}{\varepsilon\beta}\right)\right]$$



Simplified Coincidence Equations

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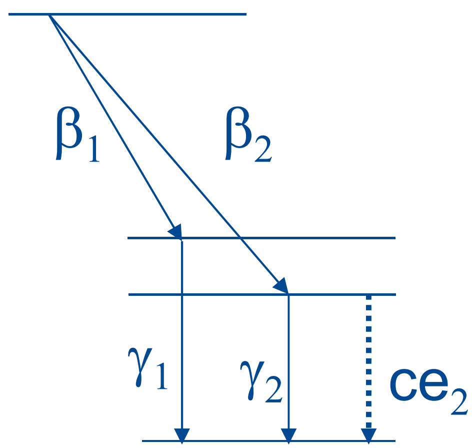
for multiple beta branches:



$$N_\beta = N_0 \sum_r a_r \left[\varepsilon_{\beta_r} + \left(1 - \varepsilon_{\beta_r}\right) \left[\left(\frac{1}{1+\alpha_r} \right) \varepsilon_{\beta\gamma_r} + \left(\frac{\alpha_r}{1+\alpha_r} \right) \varepsilon_{\beta ce_r} \right] \right]$$

$$N_\gamma = N_0 \sum_r a_r \left[\left(\frac{1}{1+\alpha_r} \right) \varepsilon_{\gamma_r} \right]$$

$$N_c = N_0 \sum_r a_r \left[\left(\frac{1}{1+\alpha_r} \right) \varepsilon_{\beta_r} \varepsilon_{\gamma_r} \right]$$

γ Gates : ^{243}Am

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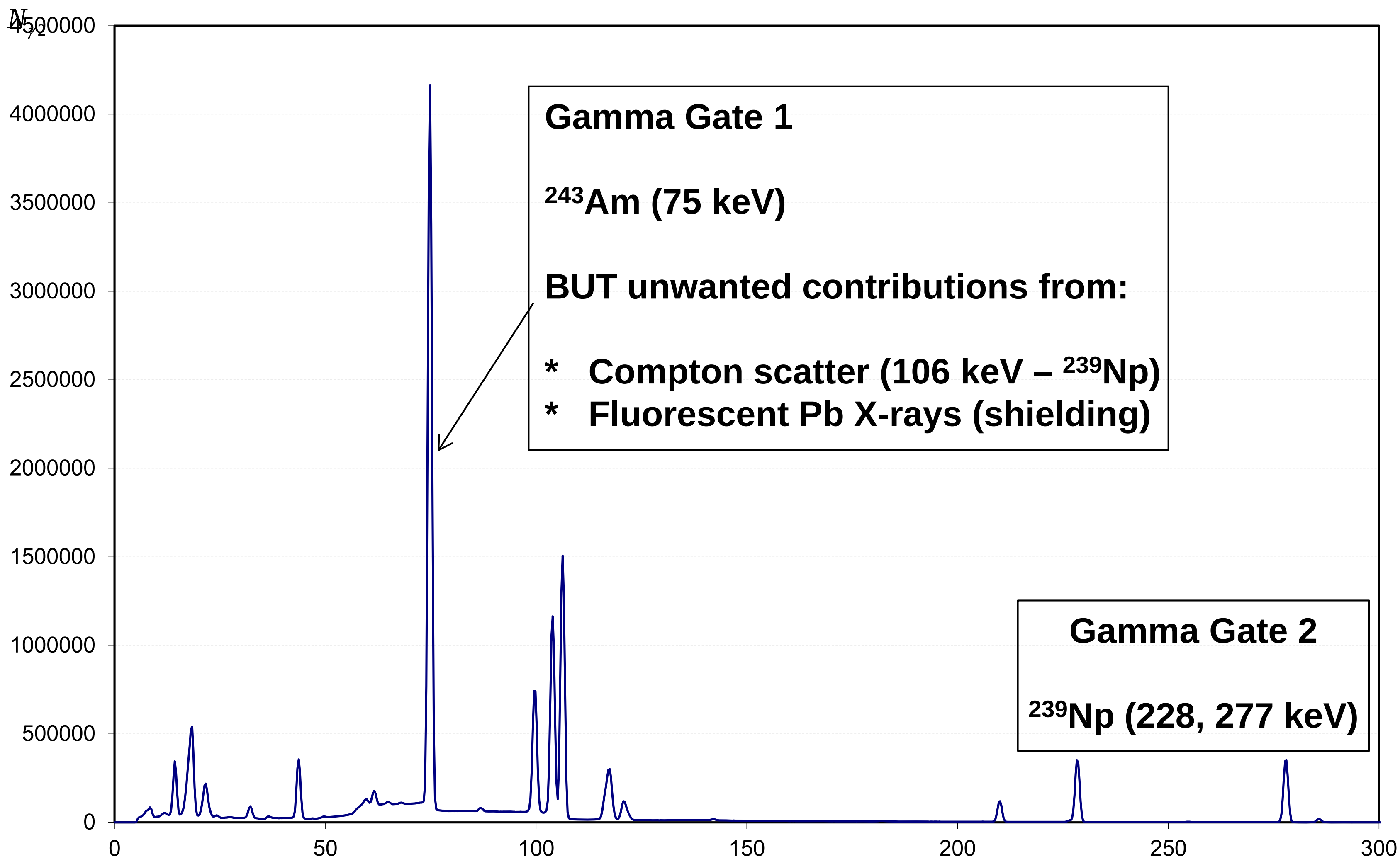
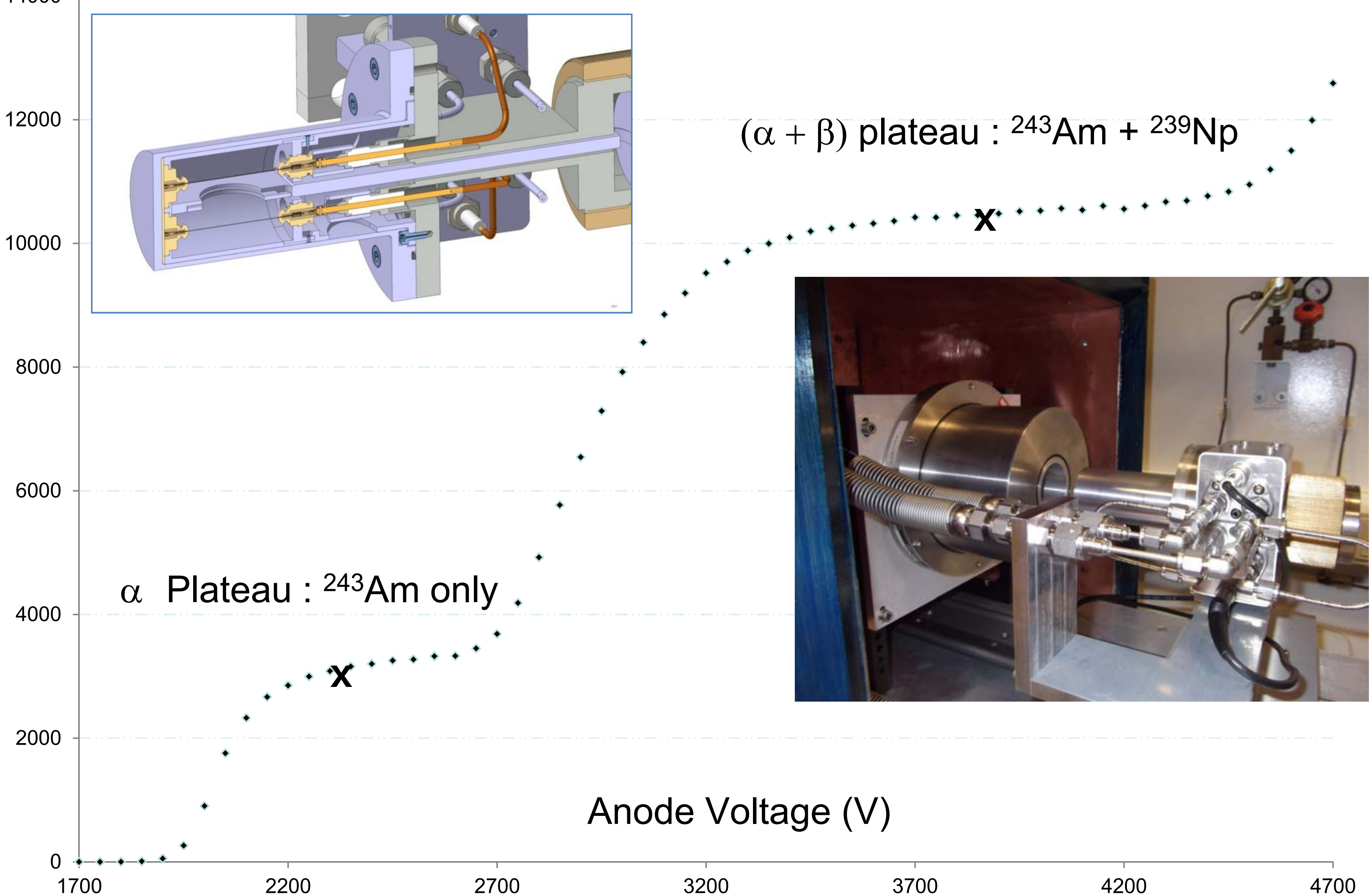


(High Pressure Proportional Counter - P10 gas - 5 bar)



On alpha plateau, γ gate on 75 keV

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$$
\begin{aligned}
N_{HPPC} &= N_0 \varepsilon_\alpha \\
N_{\gamma(75keV)} &= N_0 \left[\varepsilon_{\gamma(75keV)} + a\varepsilon_{\gamma(106keV\_Np239)} \right] \\
N_c &= N_0 \varepsilon_\alpha \varepsilon_{\gamma(75keV)}
\end{aligned}
$$

thus : bias in efficiency extrapolation:

$$
\frac{N_c}{N_{\gamma(75keV)}} = \varepsilon_\alpha \left[\frac{1}{1 + a\varepsilon_{\gamma(106keV\_Np239)}} \right]
$$

On α+β plateau, use multiple gates

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$$N_{HPPC} = N_0 \begin{bmatrix} \varepsilon_\alpha + (1-\varepsilon_\alpha)\varepsilon_{HPPC_{\gamma(Am-243)}} + \\ \varepsilon_\beta + (1-\varepsilon_\beta)\varepsilon_{HPPC_{\gamma(Np-239)}} \end{bmatrix}$$

$$N_{\gamma(75keV)} = N_0\left[\varepsilon_{\gamma(75keV)} + a\varepsilon_{\gamma(106keV\_Np239)}\right]$$

$$N_{c(75keV)} = N_0\left[\varepsilon_\alpha\varepsilon_{\gamma(75keV)} + \varepsilon_\beta a\varepsilon_{\gamma(106keV\_Np239)}\right]$$

$$\frac{N_{c(75keV)}}{N_{\gamma(75keV)}} = E_1$$

$$N_{\gamma(228,277keV)} = N_0\varepsilon_{\gamma(228,277keV)}$$

$$N_{c(228,277keV)} = N_0\varepsilon_\beta\varepsilon_{\gamma(228,277keV)}$$

$$\frac{N_{c(228,277keV)}}{N_{\gamma(228,277keV)}} = \varepsilon_\beta = E_2$$

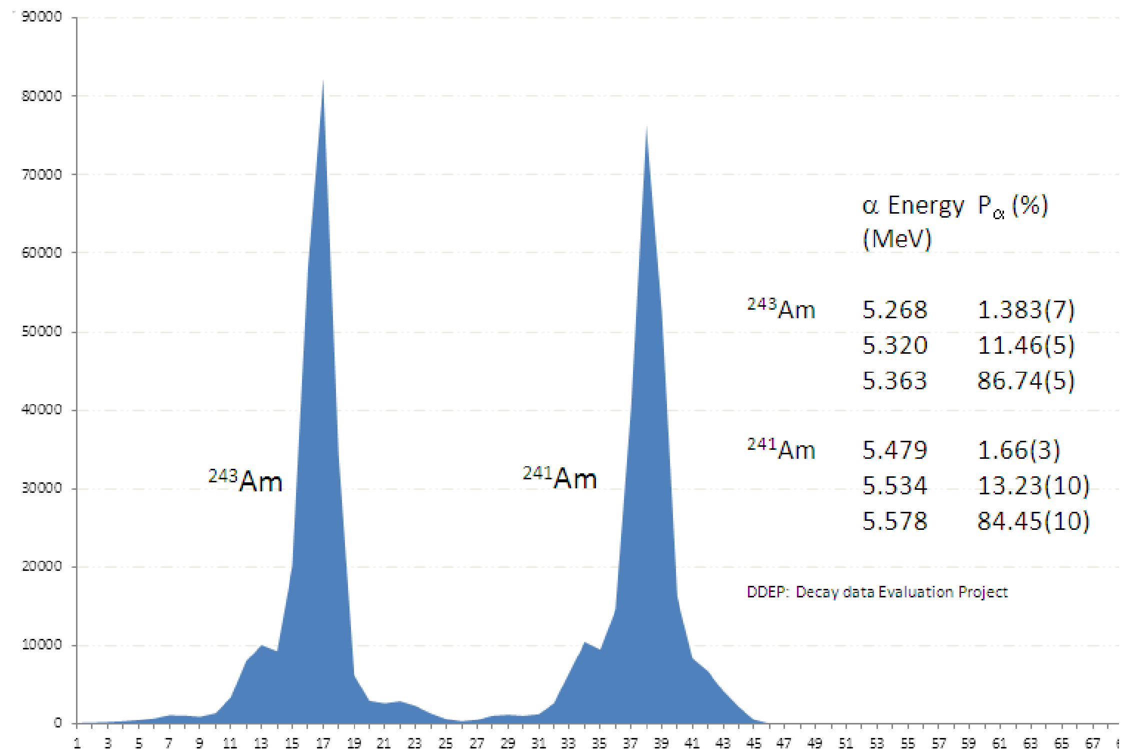
$$N_{HPPC} = N_0\left[2 - k_1(1-E_1) - k_2(1-E_2)\right]$$

4π(LS)-γ coincidence counting

Using two opposing PMTs, facing LS vial, and operating in coincidence as the “beta” channel.

Comparison with ^{241}Am standard

- α Spec
- ^{243}Am/^{241}Am readily resolved
- Expected to yield the lowest measurement uncertainty



Maintaining traceability

- Samples which are traceable to the ^{243}Am stock solution will be sent to the BIPM (Sevres, France) for assessment on the SIR.
- A pure solution of ^{239}Np will also be linked to the SIR.
- New measurements of nuclear data will be reported.

- New calibrations will be derived on NPL's secondary standard equipment :
  - Ionisation chambers
  - Gamma spectrometers
- Propose to compare with NIST SRM 4332D/E ?

Fresh, post Improvised Nuclear Device (IND) reference material

- Identified as a need at CIRMS workshop in 2009
  - Existing nuclear test debris is all over 20 years old
  - Above ground test material is all over 45 years old
  - Weathered
  - Matrix is rock: >300m deep, stable geology

Urban Vitrified Composite matrix (UVC)

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- Aim: to develop a material that mimics debris from detonation of an “improvised nuclear device” in a town.

- Approach chosen:
  - Controlled spiking of materials
  - Based on oxide fusion to generate a ‘glass’
  - Includes fission yield monitor

UVC Scope

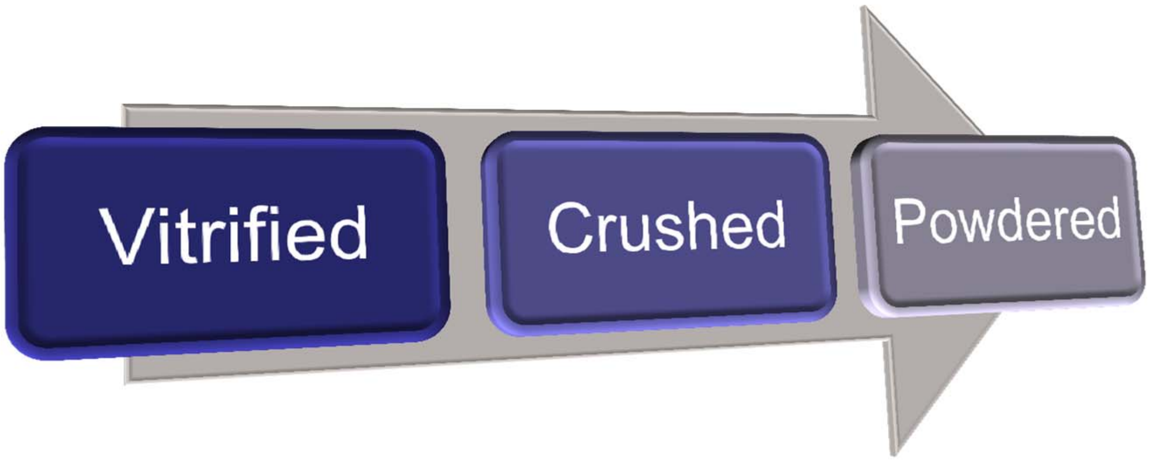
- Phase 1:
  - Production of 2.5 kg of blank UVC Matrix across 100 samples.
  - Fused aluminosilicate
- Phase 2:
  - UVC matrix doped with ^{235}U (enough for 10^{13} fissions after brief irradiation)
  - i.e: 250s in a thermal neutron flux of 5×10^{13} n/cm^2/s

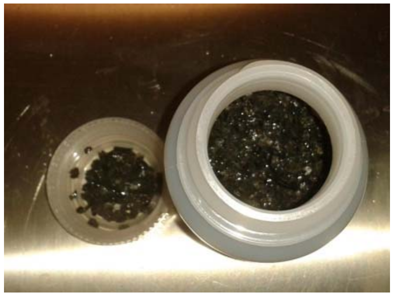
  - 22×10^{-6}g ^{235}U/g =1.8 Bq/g

Target composition of UVC matrix

Bulk matrix	Fused alumosilicate - contains >10% each of Aluminium, Silicon, Calcium, Fluorine and Oxygen				
Urban matrix ‘impurity’ elements (target values)					
Element	**ppm**	**Element**	**ppm**	**Element**	**ppm**
Si	186000	Cl	37	Nb	55
Al	7900	Sr	130	Ni	450
Fe	230000	Cr	850	Sn	36
Ca	220000	Zn	47	Ta	25
Mg	4800	F	260	V	150
S	3700	As	14	Zr	15
Na	700	B	3	N	2
K	1900	C	470	U	100
Ti	550	Co	37	W	10
P	200	Cu	220	Th	10
Mn	1200	Mo	190	O	660000

Scoping studies





Additions to the UVC project team

- Simon Jerome (NPL)
  - Pioneer of the project, with Ken Inn (NIST)
  - Simon has been very unwell and absent for several months. Expected back in 2013.
    - Replaced by:

- Professor Kym Jarvis (Imperial College, London)
  - Head of Environmental Research Centre's ICP unit, research interests include trace metal analysis, ICP-MS

- Professor Susan Parry, (Imperial College, London)
  - Professor of Radiochemistry, consultant to the IAEA, QA auditor, research interests include neutron activation analysis

- George Ham (UK Health Protection Agency)
  - Until recent retirement was in charge of the radioanalytical laboratory at HPA Chilton. 40 years experience in low-level radio analytical chemistry.

Next steps

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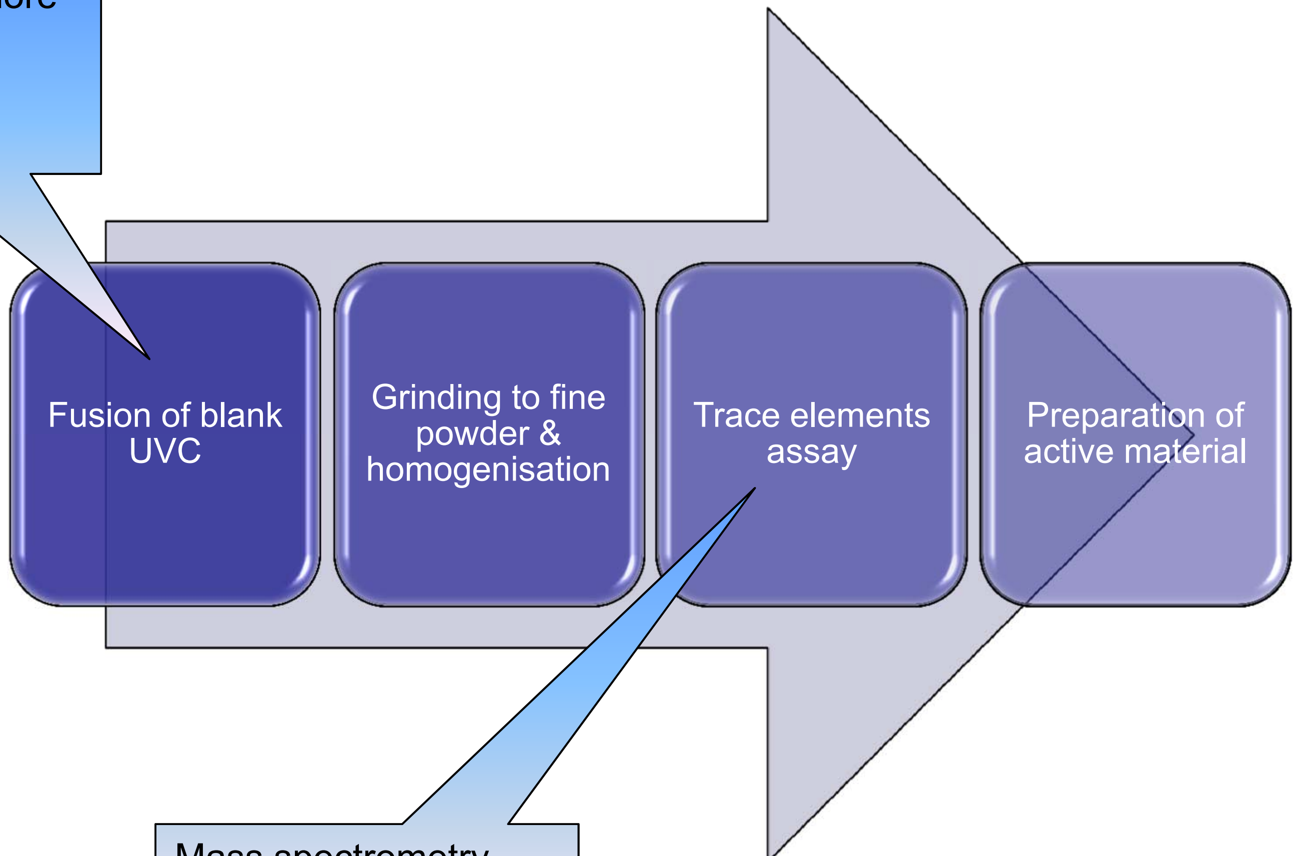
Glass mixture is more corrosive than expected.
Some delays …



Mass spectrometry
ICP-MS
Nuclear spectrometry