Rapid Trace Uranium Measurement by Neutron Activation Analysis

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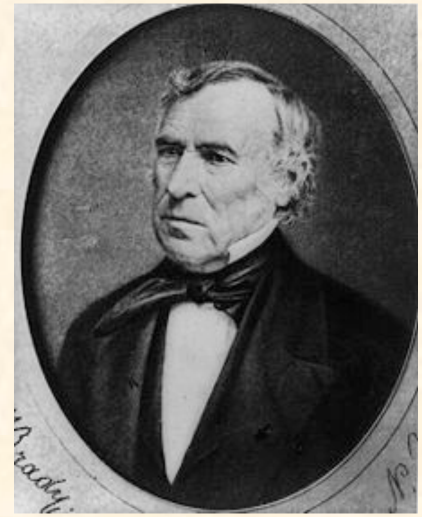
NAA for Forensics, ORNL Cases

- Bullet lead and gunshot residue - JFK
- Paint and ink – Counterfeit detection
- Plastic and glass – Taco Bell murders
- Metals and alloys – OK City bombing
- Geological materials – Mammoth Cave
- Special nuclear material - IAEA
- Attribution – RDD, etc.
- Biological materials – Hg in deer, Z.T., dino bone



Neutron Activation Analysis Features

- **Sensitive** – Many elements 10^{-12} g/g sensitivity
- **Accurate** – All error sources known, NIST primary method for certification of trace element concentration in RMs, Linear
- **Blank Free** – Usually perform analysis in different container than that used for irradiation
- **No Chemical "Matrix" Effects** – Nuclear techniques are not dependent on or sensitive to the electronic environment of the sample
- **Nondestructive** – Sample dissolution is usually not required
- **Versatile** – Techniques analyzing β, γ-ray, x-ray, and neutron radiation are employed on biological, geological, and engineered materials

Forensics Requirements

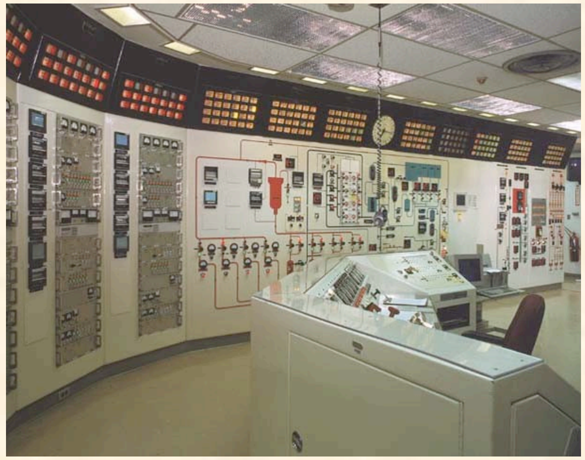
- Method must be accurate
- Sources of error must be known, quantified where possible
- Ideally, method described by respected standard (now a requirement)
- Easy to understand, explain
- Definitive

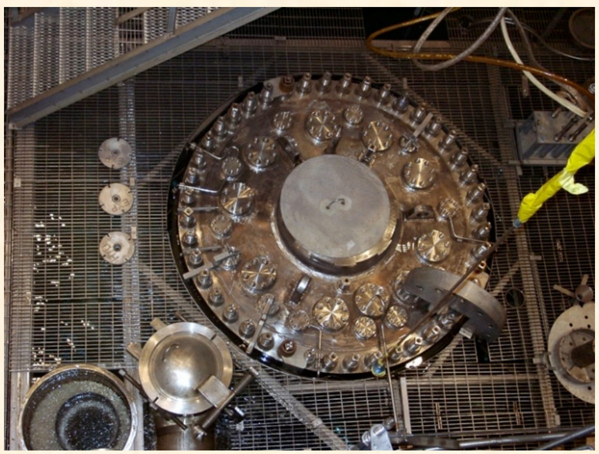
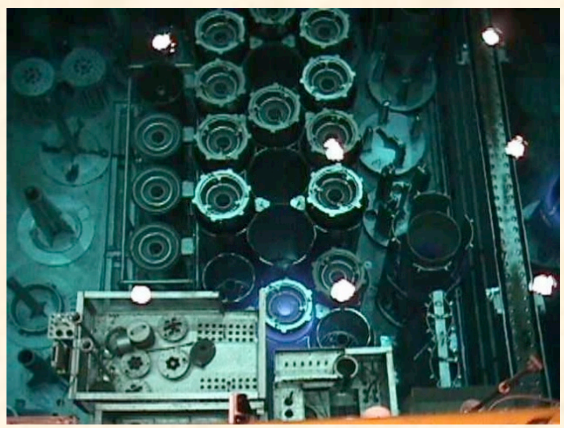
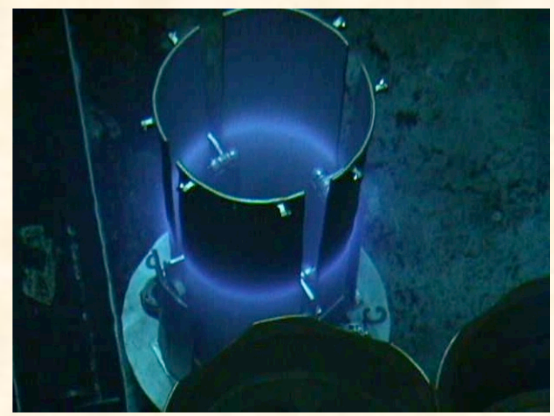
Why is rapid U measurement needed?

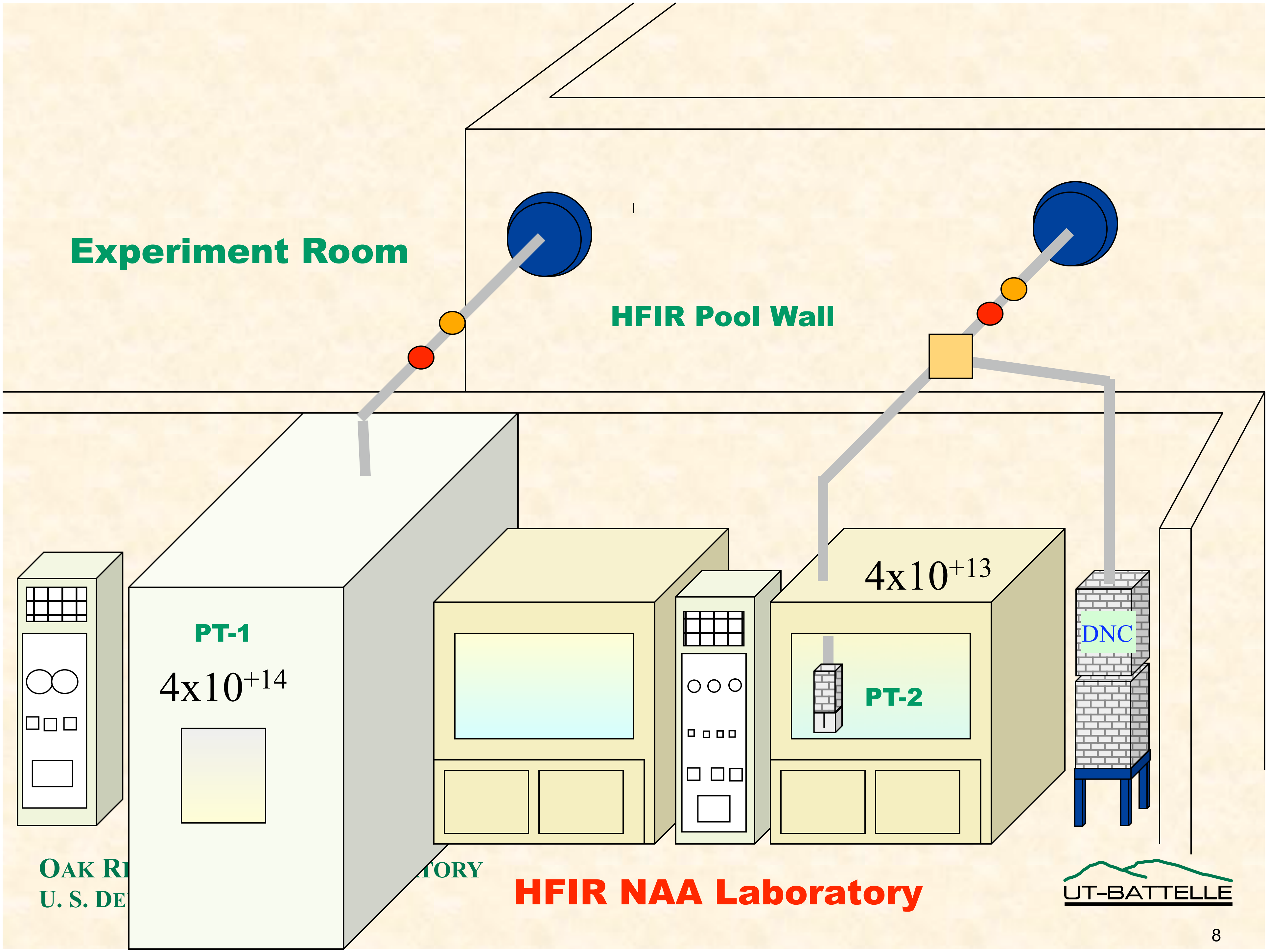
- Any nuclear event will quickly overwhelm the World's mass spectrometry labs
- Cleanroom protection
- Routine monitoring efforts generate large numbers of samples
- Wide-area applications using vegetation, swipes, or other substrates
- Fast response to LE needs

What's at stake?

High Flux Isotope Reactor


Experiment Room
HFIR Pool Wall
PT-1
$4x10^{+14}$
$4x10^{+13}$
DNC
PT-2
HFIR NAA Laboratory
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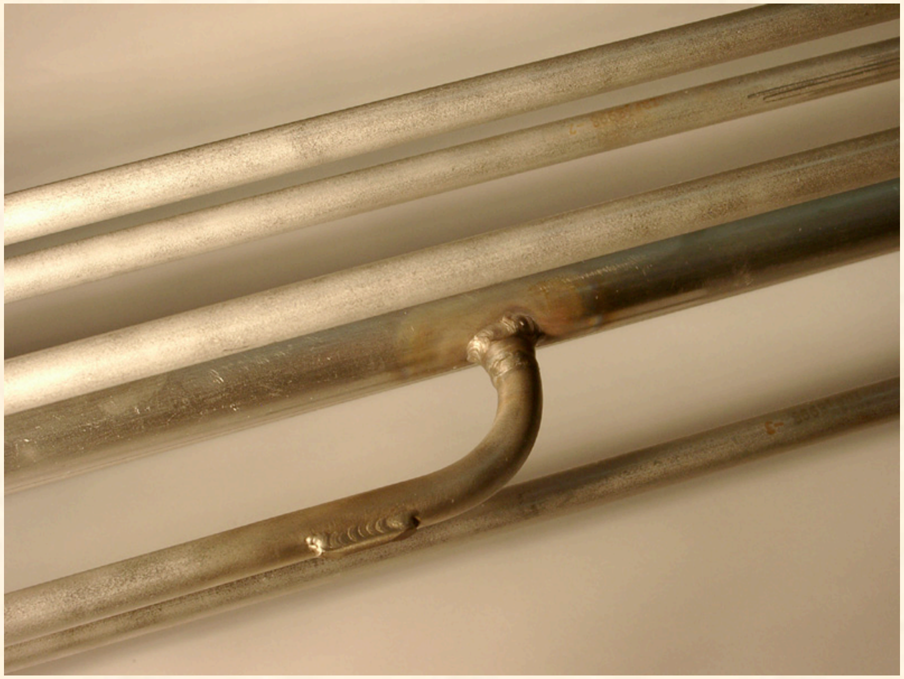
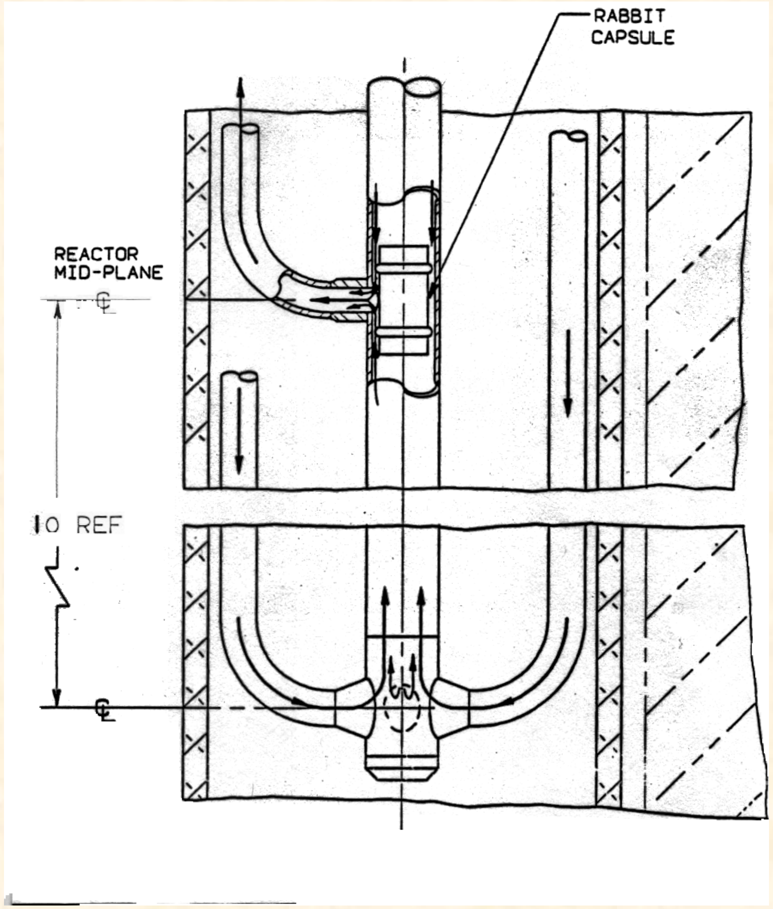
HFIR PT-2 Loading Station

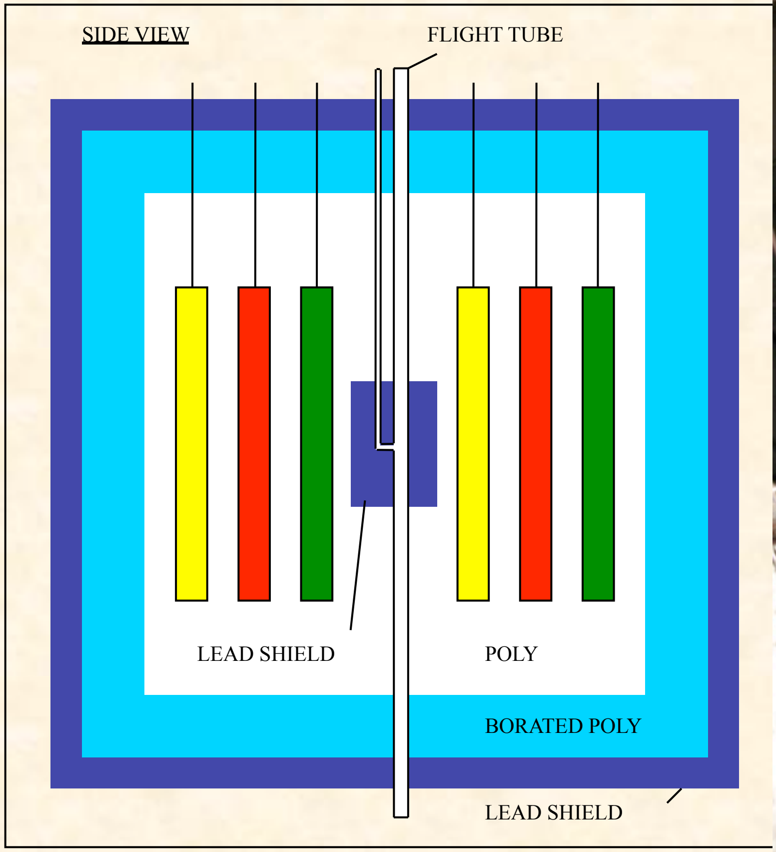
Pneumatic Transfer Facilities

Reactor Irradiation Position





Delayed Neutron Detector Array



Delayed Neutron Activation Analysis, DNAA

$$C(t) = \varepsilon \cdot \frac{m_f N_A \sigma_f \varphi}{M_f} \cdot \sum_i \frac{\nu_i}{\lambda_i}\left(1 - e^{-\lambda_i t_a}\right) e^{-\lambda_i t_c} \cdot \left(1 - e^{-\lambda_i t}\right)$$

m_f = mass of fissile nuclide

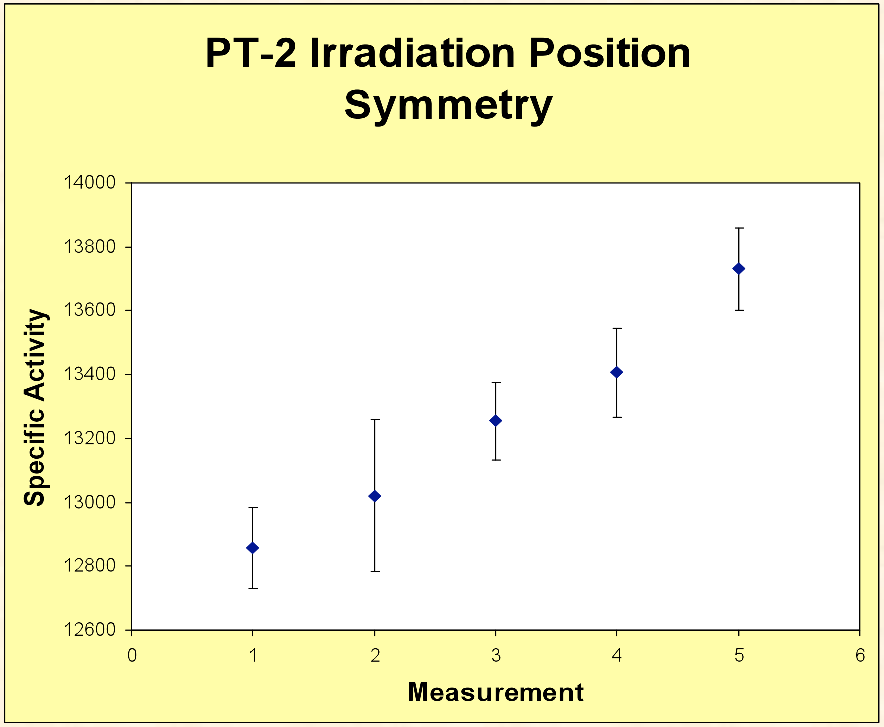
M_f = molar mass of fissile nuclide

φ = irradiation neutron fluence rate

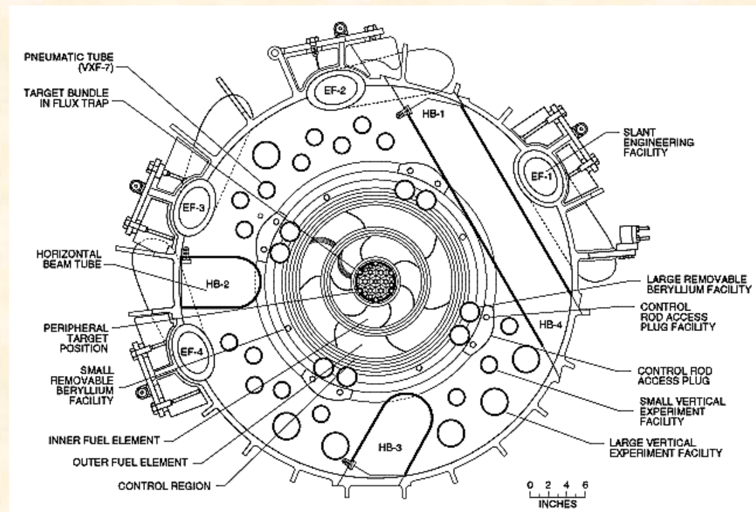
ν = average DN yield per fission per group

t_a t_c t = irradiation, decay, counting times

PT-2 Position Symmetry



- Radial gradient ~2-5%
- Insignificant axial gradient
- Samples do not rotate during irradiation
- Thermal 4.0 x 10^{13} n cm^{-2} s^{-1}
- Thermal/epithermal > 300



Cross section of reactor core at horizontal midplane

ORNL DNAA System Reproducibility and Linearity

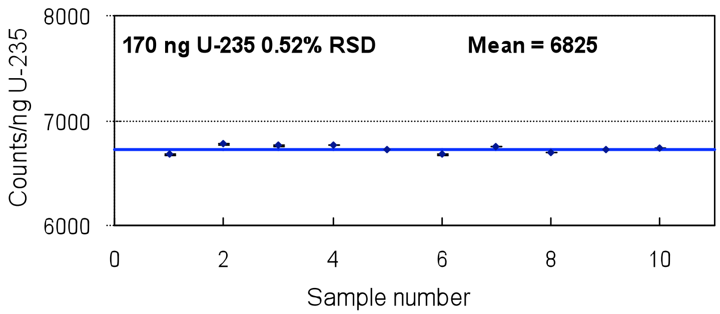


Figure 3(a) - Reproducibility Tests U-235

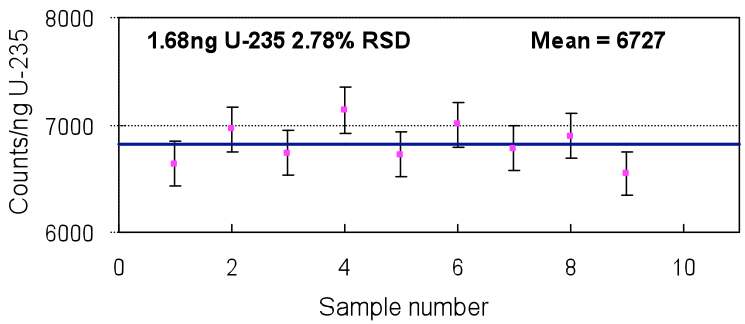


Figure 3(b) - Reproducibility Tests U-235

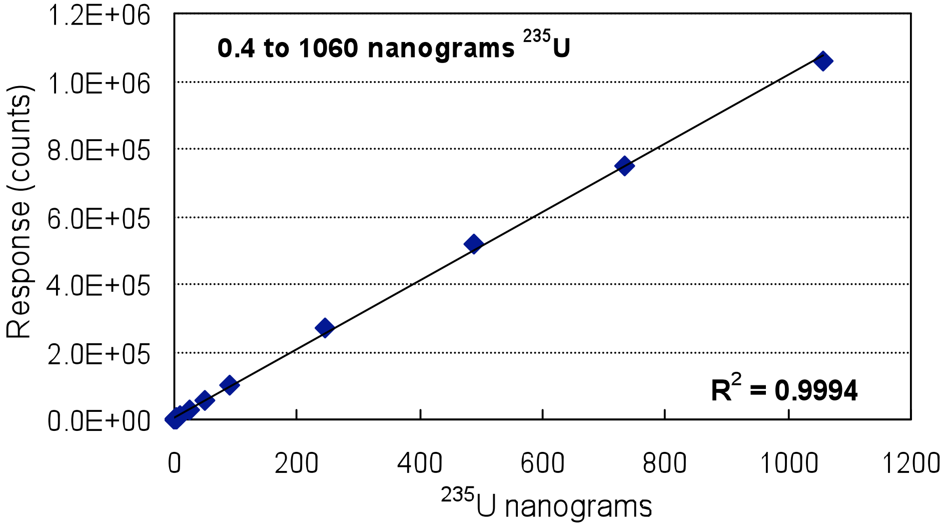
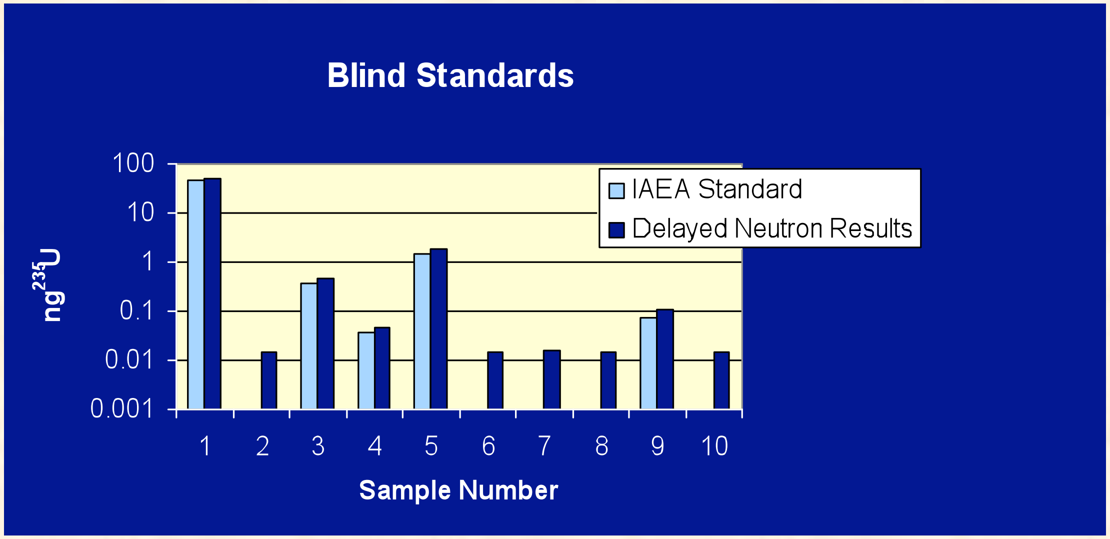


Figure 4. System linearity test

Uranium Method Performance

Material	True Total Uranium (μg/g)	Measured Uranium (μg/g)	Number of Observations	Percent Difference
2709 Soil	(3)	3.15 ± 0.08	7	5
1633a Fly Ash	10.2	10.3 ± 0.2	11	1.1
1575 Pine Needles	0.02	0.021 ± 0.002	14	6.5
4353 Soil	3.1	2.9 ± 0.2	3	6.1
3164 Soil	50	48.6 ± 0.02	316	2.8

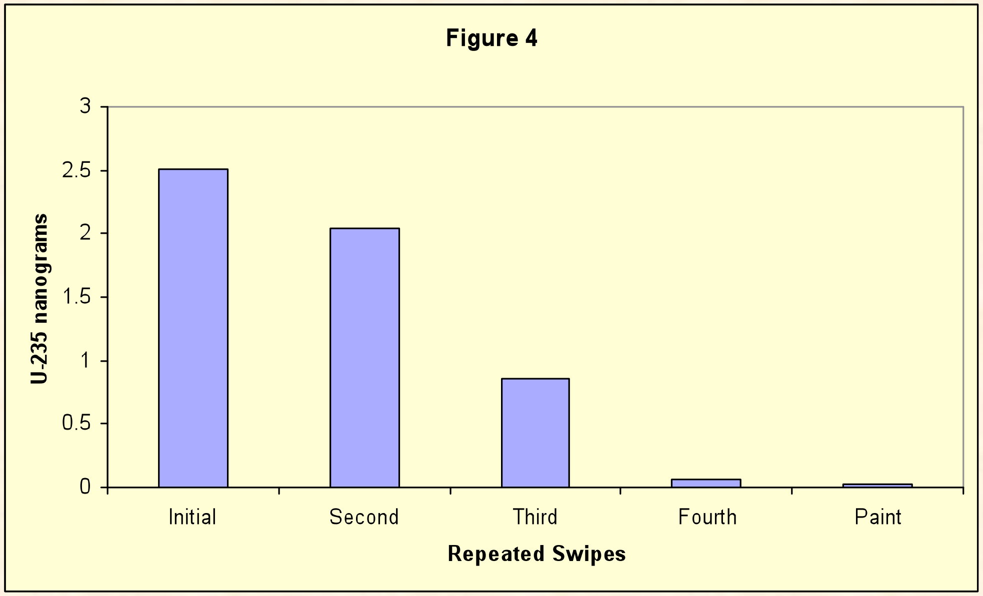


HFIR Uranium Survey

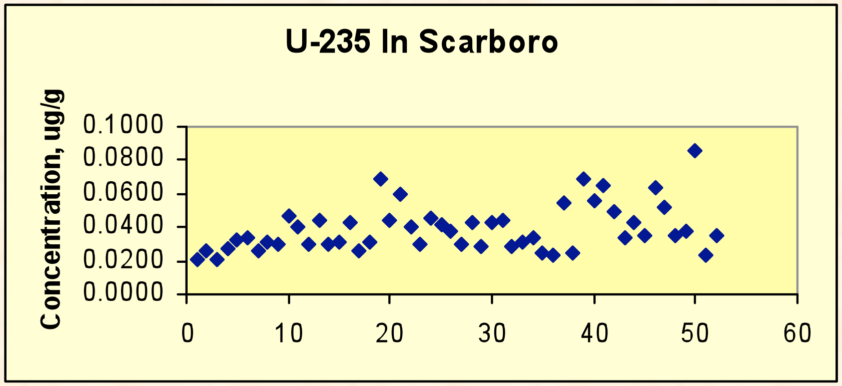
- 42 Ashless Filter Paper Swipes
- Decon Pad Floor 0.24 ± 0.07 ng
- Decon Sink 0.37 ± 0.07 ng
- FFWW 0.07 ± 0.05 ng

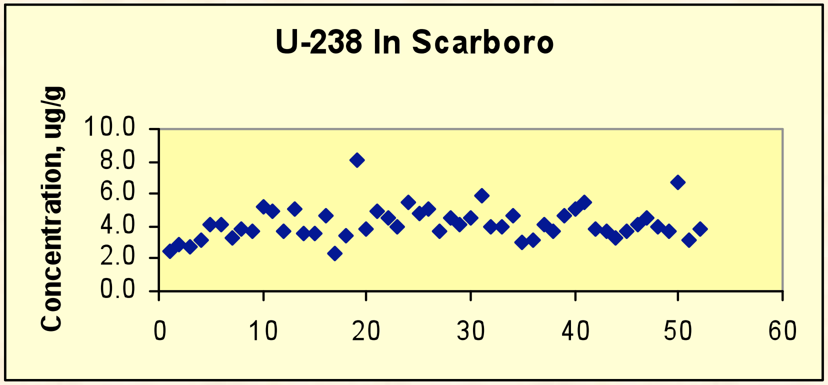
NAA Lab Survey

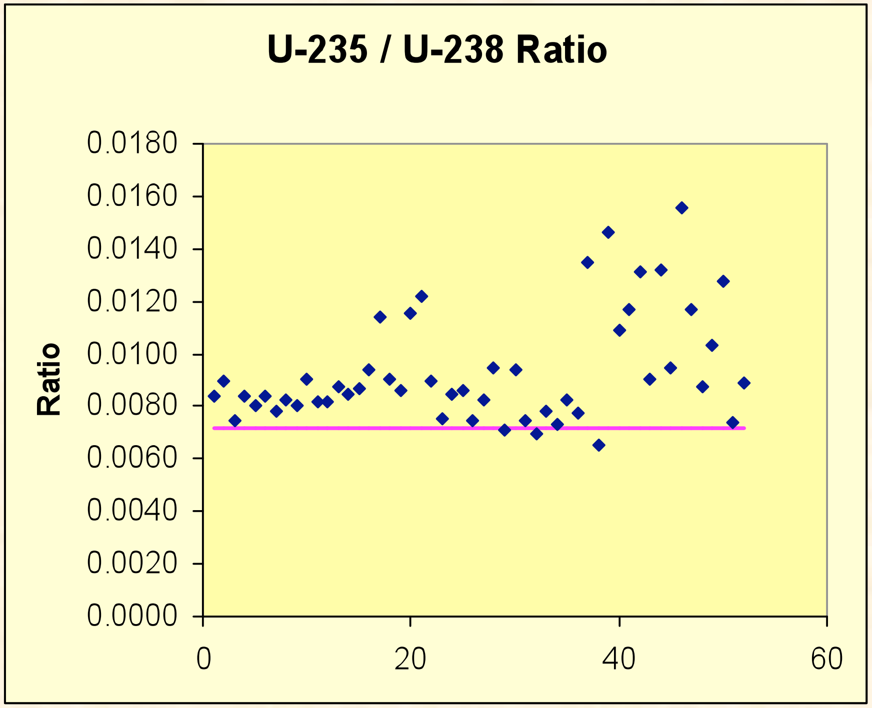
- **Hood-3 0.10 ± 0.06 ng**
- **Glovebox 0.30 ± 0.07 ng**
- **Paper Cutter 2.5 ± 0.07 ng**
- **U_3O_8 prepared in radiochemistry facility 1.30 ± 0.06 ng**



Uranium Contamination and Enrichment in Scarboro Community







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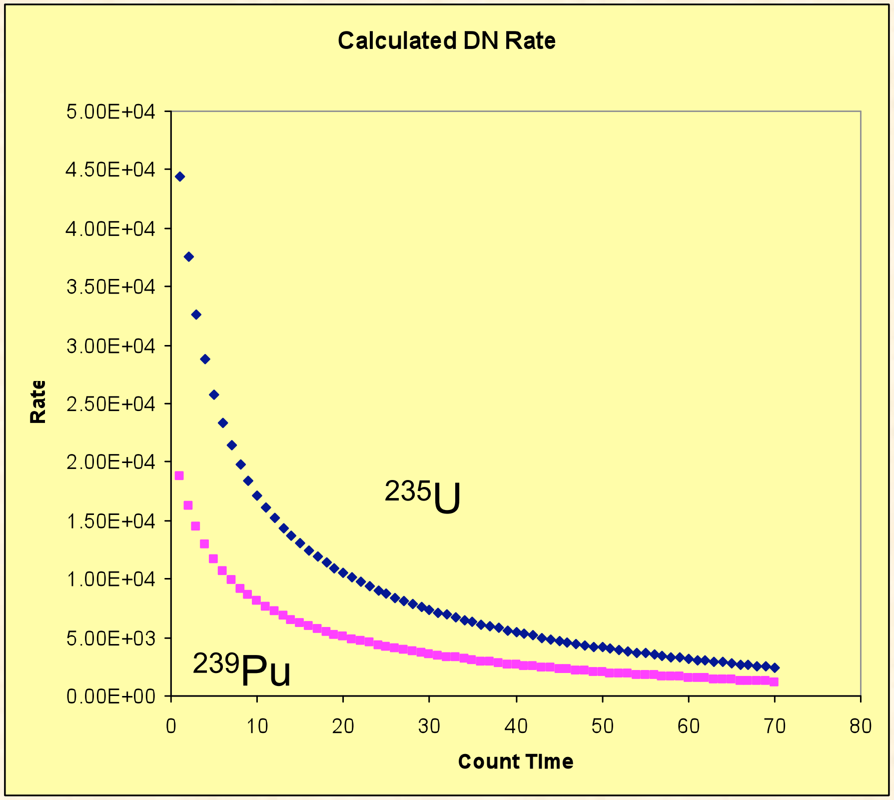
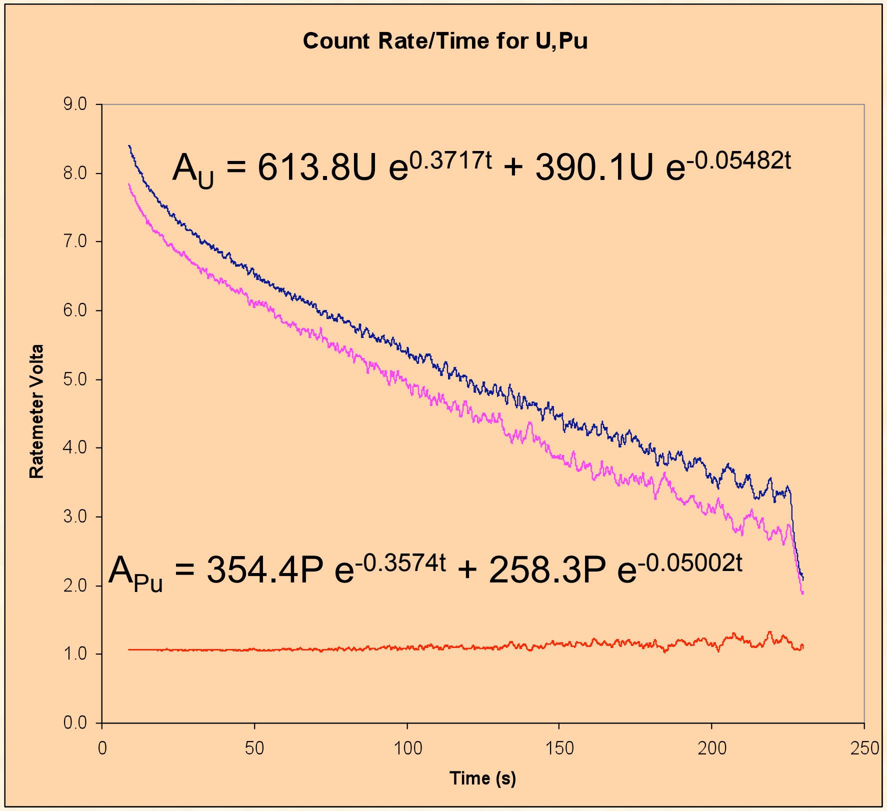
THE TENNESSEAN
High toxic levels
found in Scarboro
Toxic agents
found in area
of Oak Ridge
DOE survey
of Scarboro
scrutinized
Uranium is detected in
Oak Ridge soil samples

Simultaneous ^{235}U and ^{239}Pu Determination by DNAA

- SCHLECHTE, *et. al,* Radiochem. And Radioanal. Lett., 46(1-2), (1981), 103-114.
- HENKELMANN *et. al*, Nucl. Instr. Meth. In Phys. Res. B, 215 (2004), 246-251.





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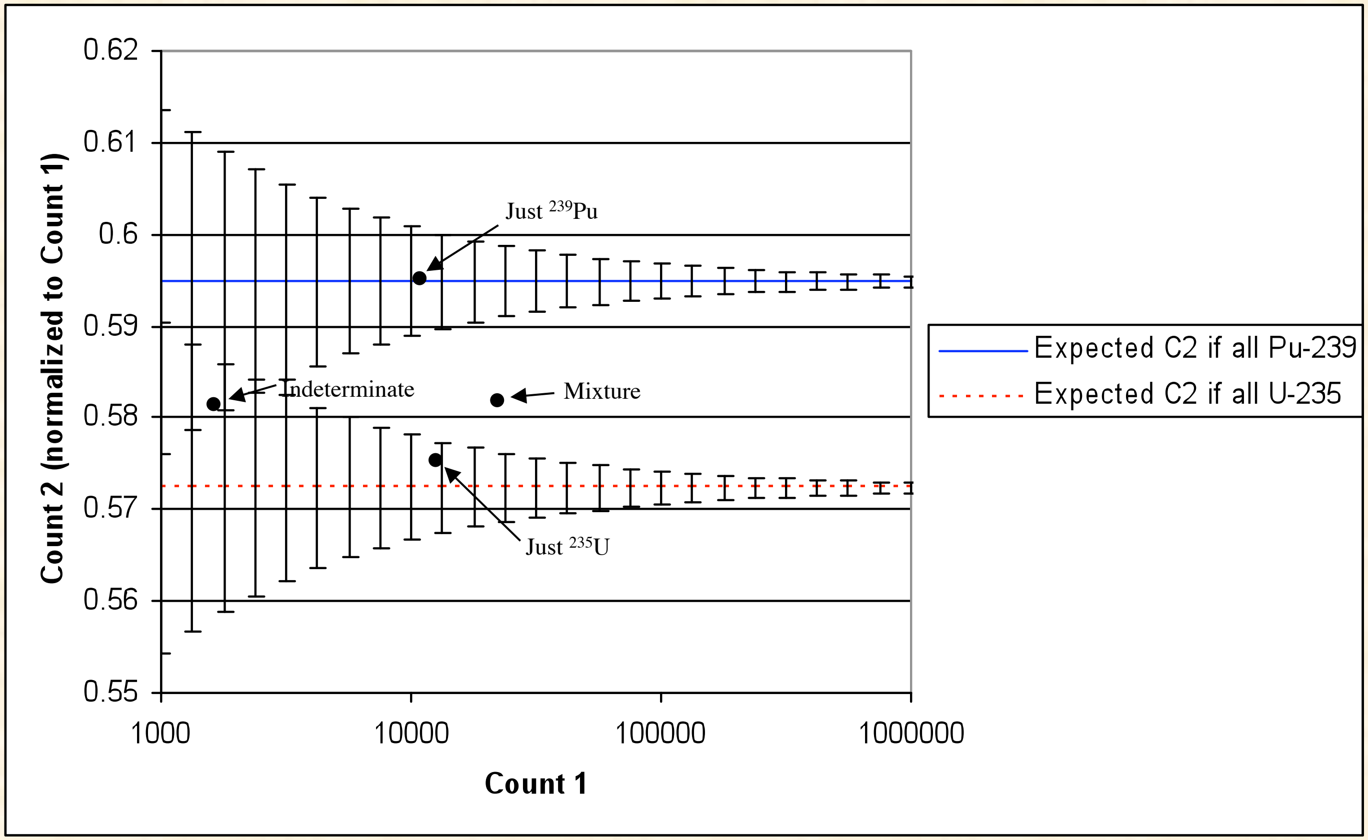
A Simple Comparison Method?



Conclusions

- DNAA is appropriately accurate, precise, and simple for Safeguards/forensics applications
- Swipes from IAEA since 2000 varied by over 5 orders of magnitude in ^{235}U
- DNAA facilitates uranium contamination surveys in near real time
- Great potential for environmental monitoring
- Throughput and cost appropriate for screening
- U 235/238 ratio by DNAA and INAA

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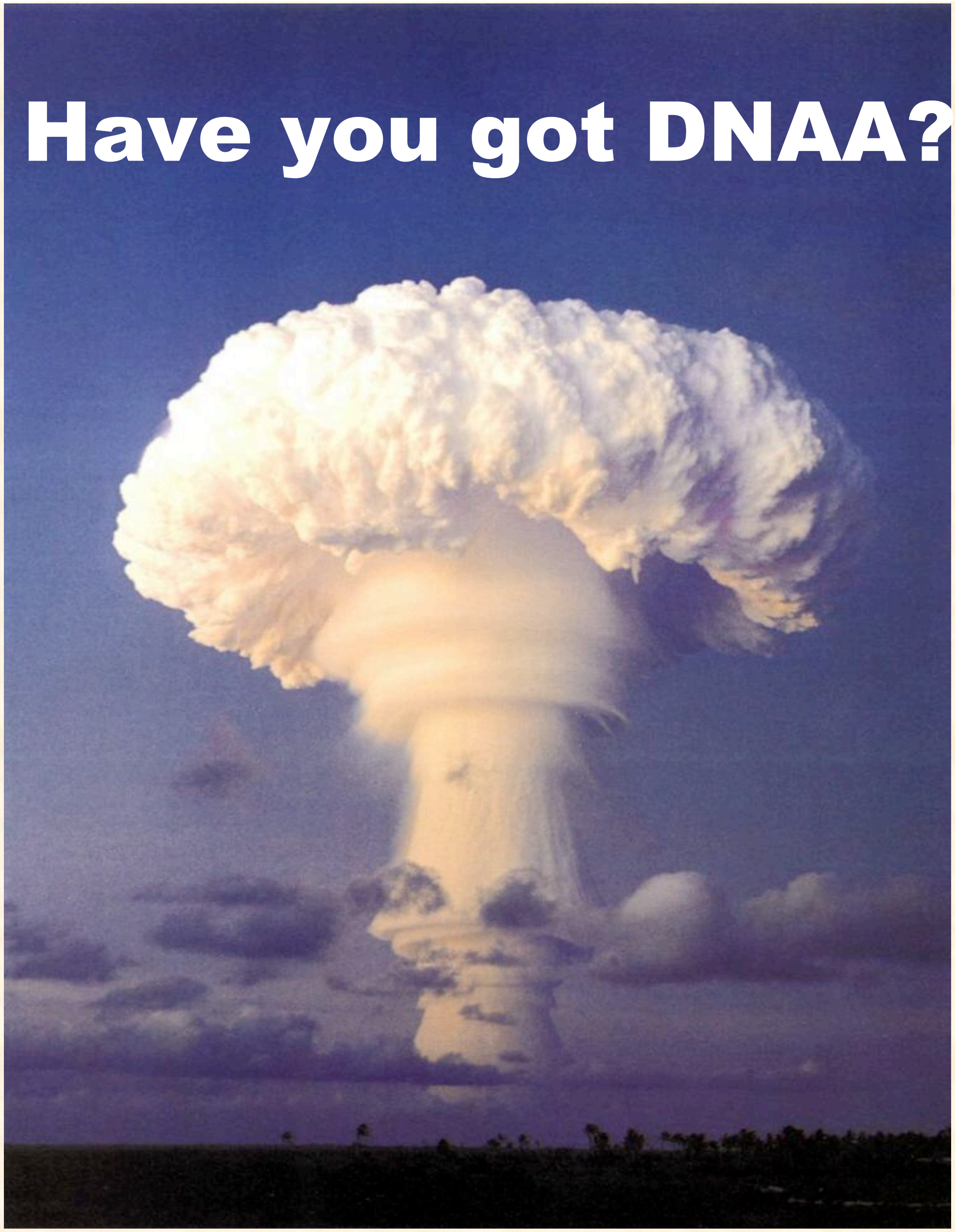
Have you got DNAA?