Lawrence Livermore National Laboratory

Resonance Ionization Mass Spectrometry (RIMS): New Technology for Nuclear Forensics

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This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344

LLNL-PRES-496534

Nuclear forensics: What do we mean?

- analysis of interdicted illicit nuclear or radioactive material and associated materials to provide evidence for nuclear attribution

- the **goal** is identification of forensic indicators in interdicted nuclear and radiological samples or the surrounding environment, **linking people** to **places**, **materials**, **events**

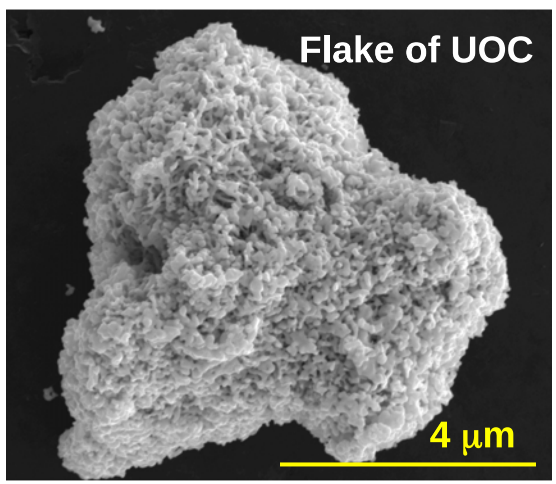
Helpful analytical data may include:

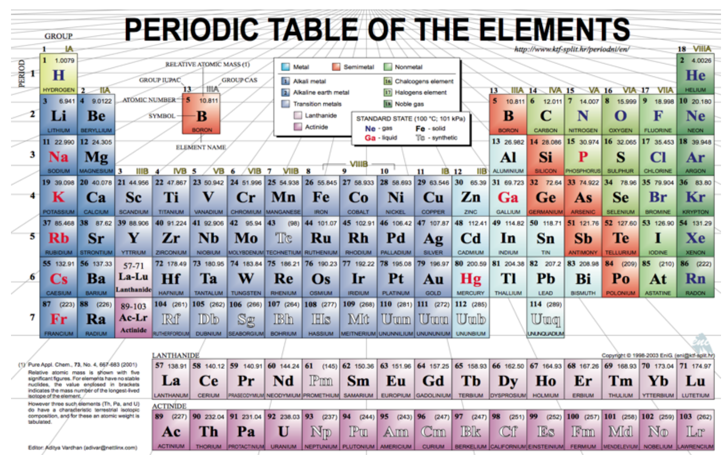
- Activity (elemental and isotopic info, safety)
- %U, Pu concentration (elemental data)
- Morphology, chemical form
- Trace elements (presence, concentration)
- Isotopic composition (^{235}U, ^{239}Pu, other actinides)
  - Natural decay products (sources, background)
  - Induced decay products (reactors, neutron sources)

Measuring nuclear materials: One size does not fit all

Analytical considerations

- Sample size?
- Spatial resolution?
  - Homogenous or heterogeneous samples?
- Analytical location (in the lab, in the field)?
- Desired precision/accuracy?
- Time frame?

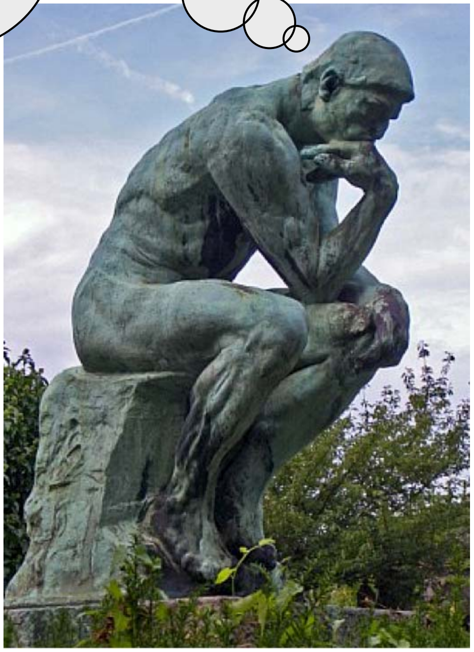




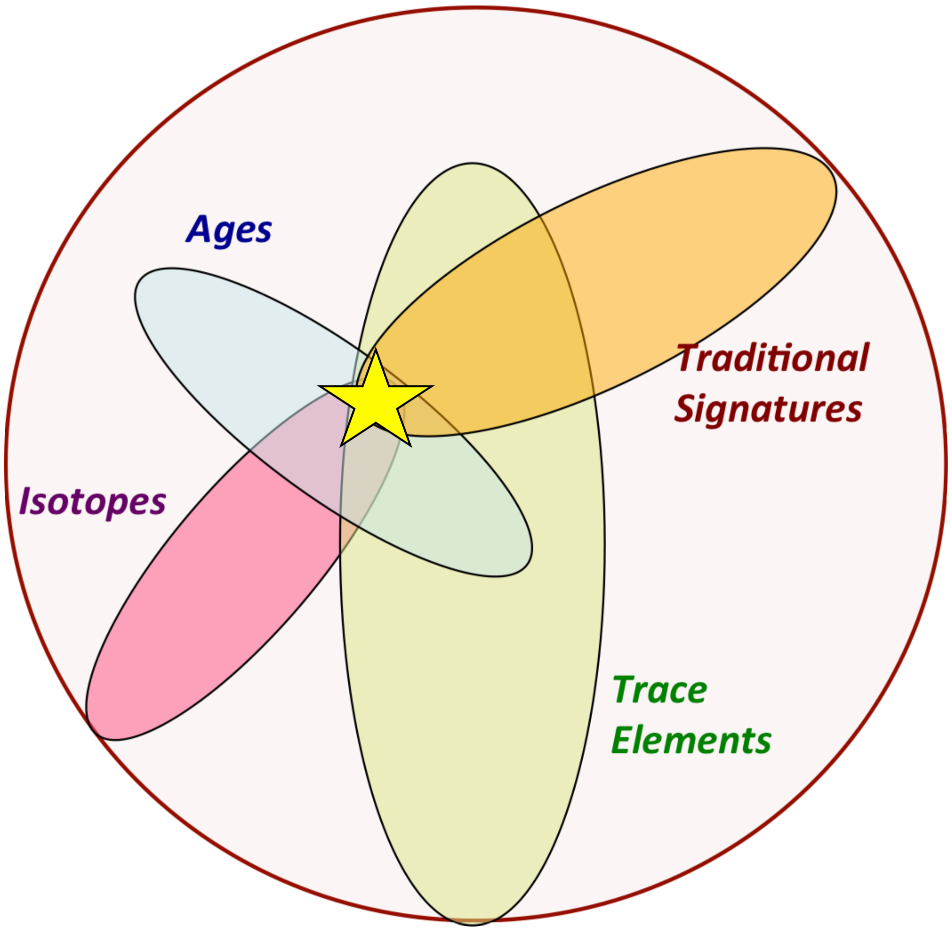
Chemical analyses are a primary tool for characterization of nuclear materials

- **Radiation detection / counting**
  - Alpha (α), gamma (γ), beta (β^-)
  - Selective isotopic information
- **Excitation/stimulation**
  - X-ray generation (fluorescence, energy dispersive spectrometry)
  - Atomic absorption and emission (atomic breakdown spectroscopy)
  - Complex samples and trace concentrations constitute a physical limitation
- **Ion formation (mass spectrometry)**
  - Any element and isotope
  - range of concentrations, spatial resolutions
  - Generally higher precision, but time consuming sample preparation

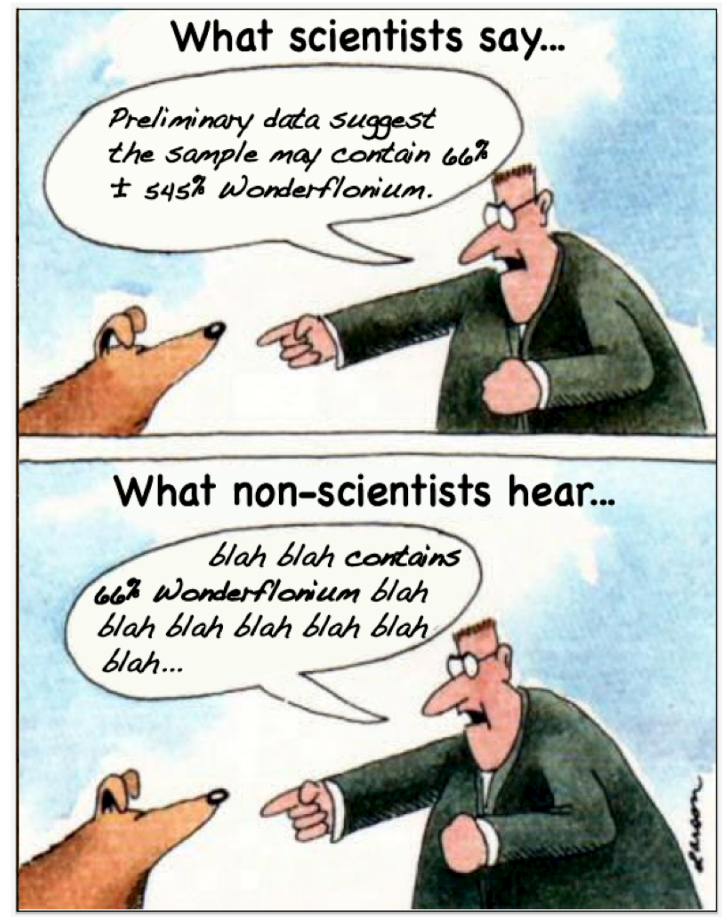
What method should I use? Which answers do I need?



There is no one path to an answer: Multiple signatures build attribution confidence



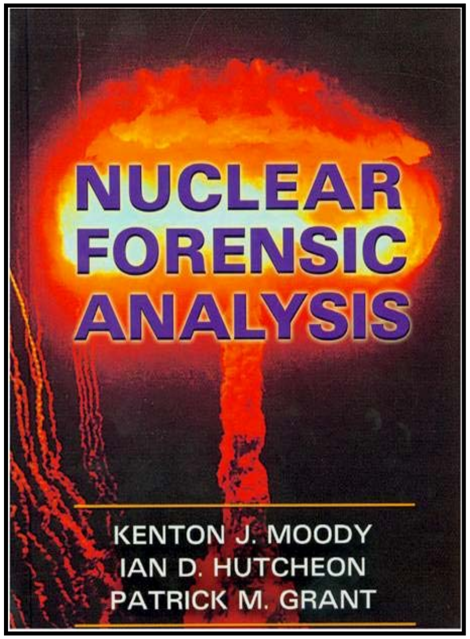
Accuracy matters!!!



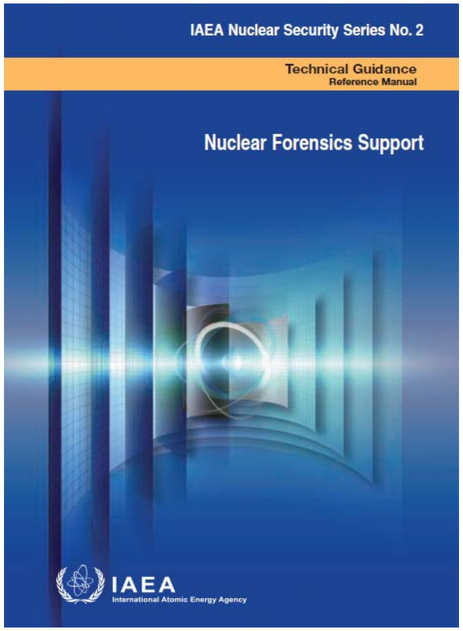
With apologies to Larsen

Moving capabilities forward: Nuclear Forensic Research

- More versatility for use?
- Improve answers (precision, accuracy)?
- Faster analyses?
- Indirect measurements?
- Smaller measurements?
- Fewer people needed?
- Reliable data processing?



*Reference text authored at LLNL, **2005***



*IAEA document by Kristo (LLNL), Smith (LLNL), Niemeyer (LLNL), and Dudder (PNNL), **2006***

U and Pu-isotopes contain critical clues about material origins, processes and intent

Actinide isotope measurements demand speed, precision and accuracy, rigorous conclusions

Resonance Ionization:
A Faster Path to a Better Answer

All Mass Spectrometers Work The Same Way:

- Make ions from the element(s) of interest
- Separate ions in space or time
- Detect the ions as separated masses

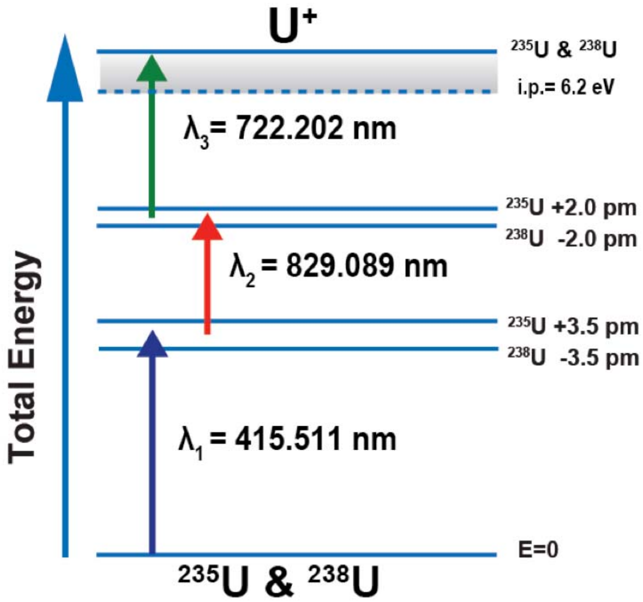
Resonance ionization uses intermediate electronic states specific to each element to form ions!

Lasers provide the energy

discrete photon energy

sufficient photon fluence

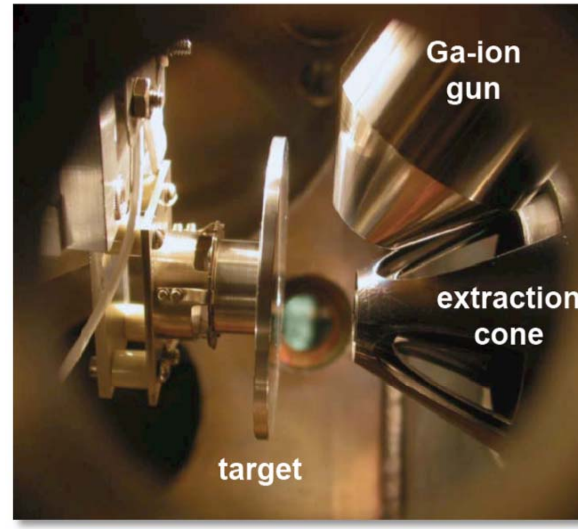
precision, timing, control

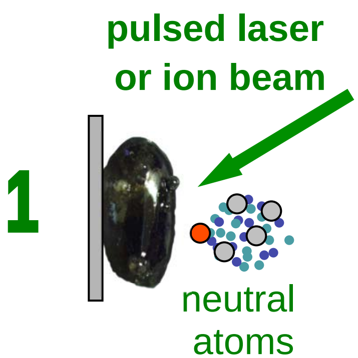


Almost any element can be ionized through this process!

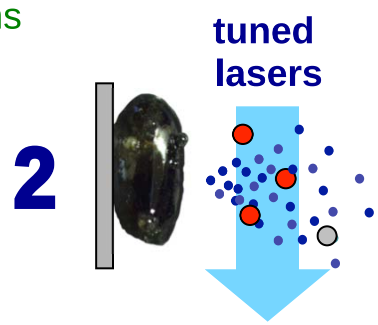
RIMS performs chemical separation in the instrument

Resonance ionization mass spectrometry measures actinide isotopes using selective laser ionization for chemical separation during analysis

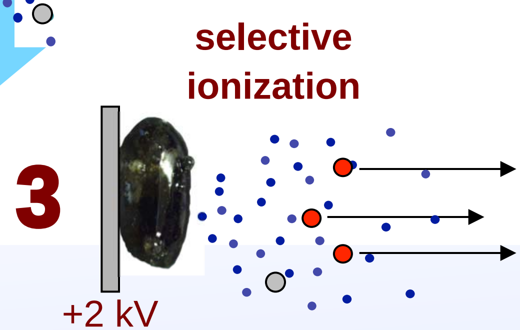




***Pulsed desorption event* removes atoms & molecules from solid materials**



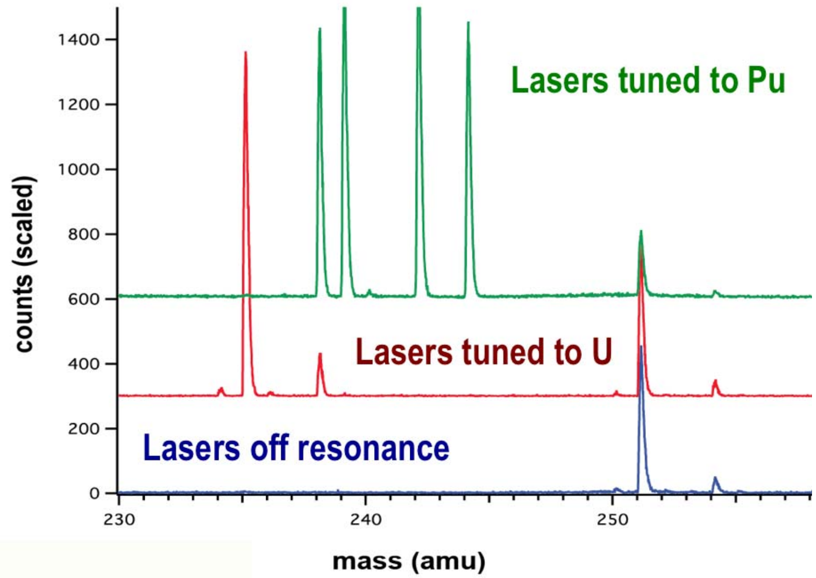
One element absorbs the *resonance and ionization lasers*, the rest are transparent

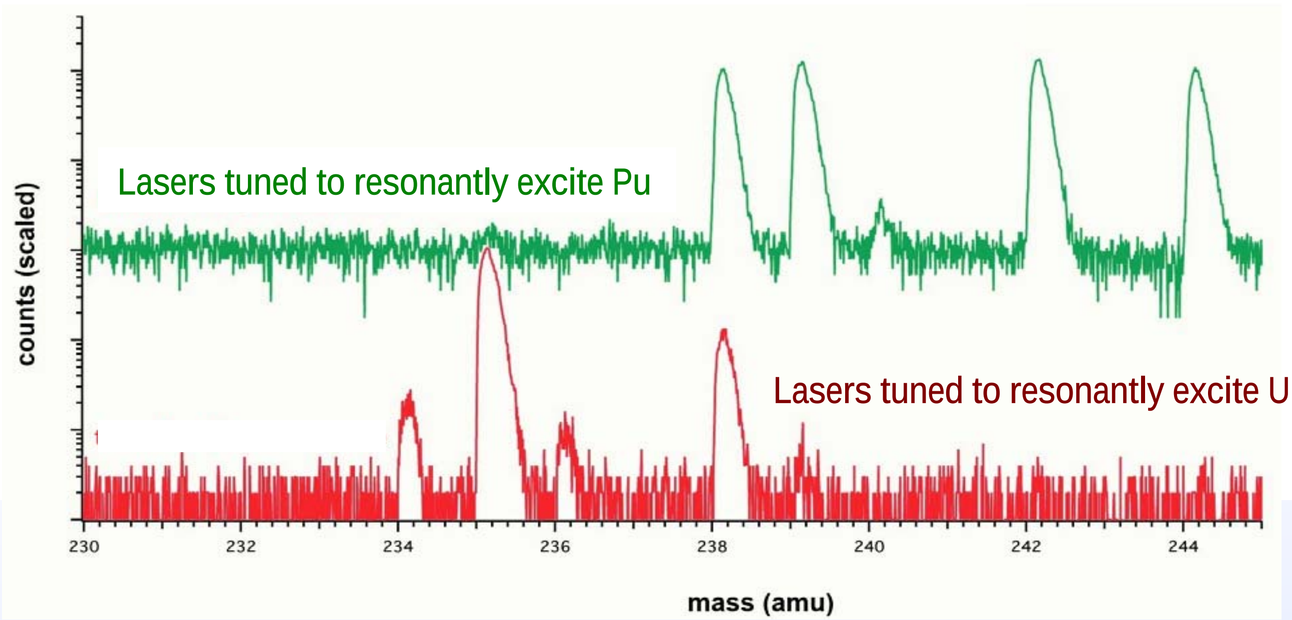


The photo-ions are accelerated to the *mass analyzer / detector*

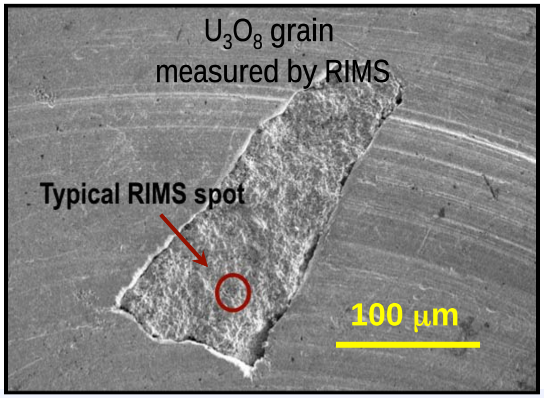
RIMS demonstrates discrimination between U and Pu from the same sample

- ^{238}U and ^{238}Pu would normally have to be separated chemically, prior to mass spectrometry
- **RIMS does this separation in the instrument**
- Tuned lasers ionize the desired element for analysis from a multi-actinide sample

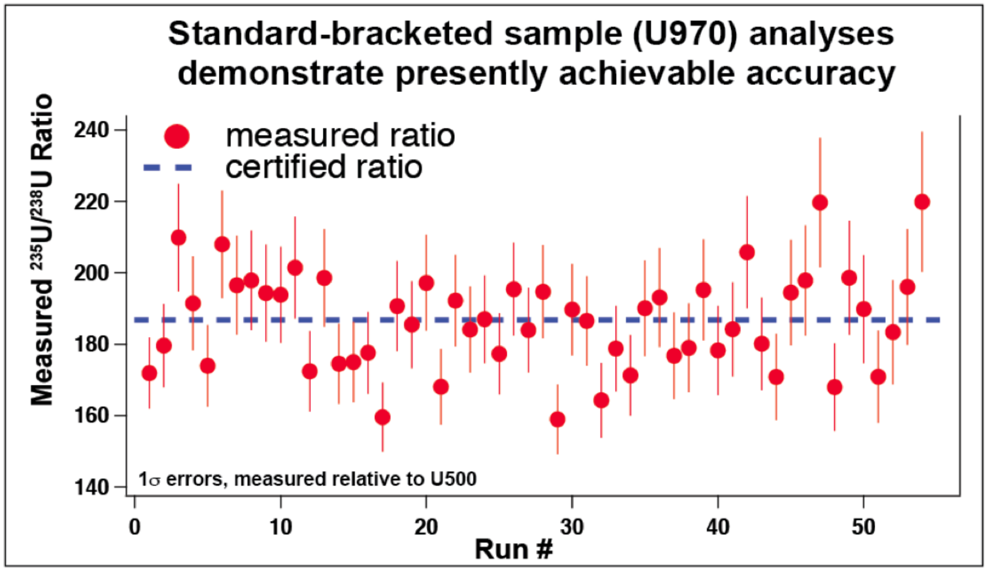




Secondary electron image



Actinide isotope ratios have been measured accurately in metal, oxide and glass samples



CRM970 (U_3O_8)

$^{235}U/^{238}U_{cert}$ = 186.77

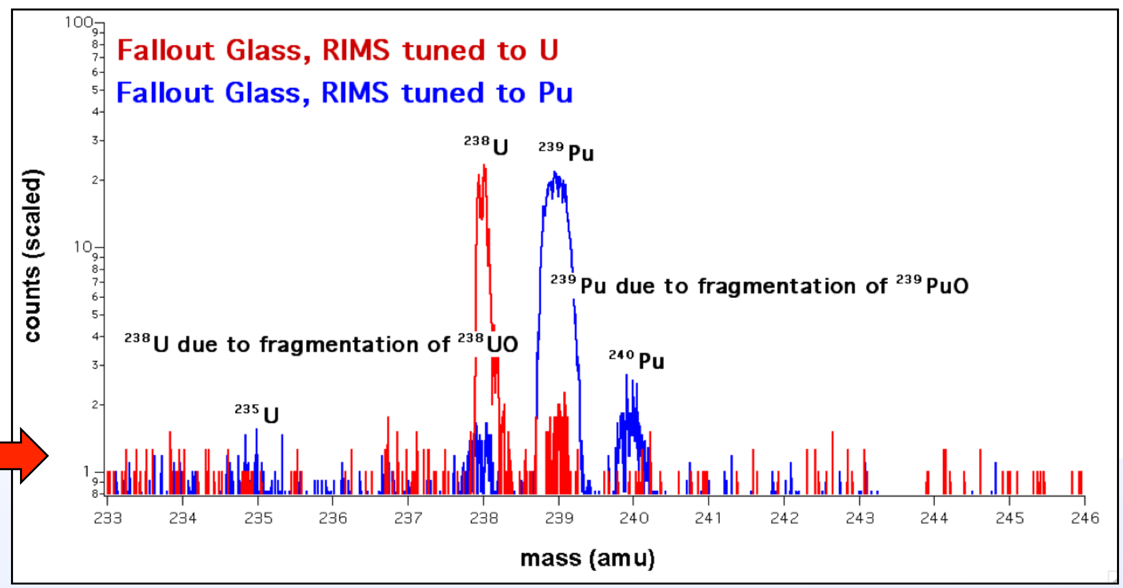
$^{235}U/^{238}U_{meas}$ = 185.16

(1σ) ± 1.76

χ^2 = 1.10

- We have measured isotope ratios over five orders of magnitude
- Completed complementary U and Pu analysis in ~2 hours or less

Preliminary results demonstrate detection of actinides in fallout debris samples with part-per-million concentrations of actinides



Sustaining and advancing nuclear forensic capabilities

Nuclear forensics is an emerging science

By necessity, it draws on a team of people to research and innovate, provide process knowledge and materials experience, and perform technical analyses

Research → **Application** ← Experience