

LA-UR-19-23368

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Title: Introduction to the use of isotope analysis in nuclear forensics

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Intended for: Undergraduate lecture at University Nevada, Las Vegas

Issued: 2019-04-15

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Introduction to the use of isotope analysis in nuclear forensics

Introduction to Radiochemistry - UNLV

Jeremy Inglis, LANL

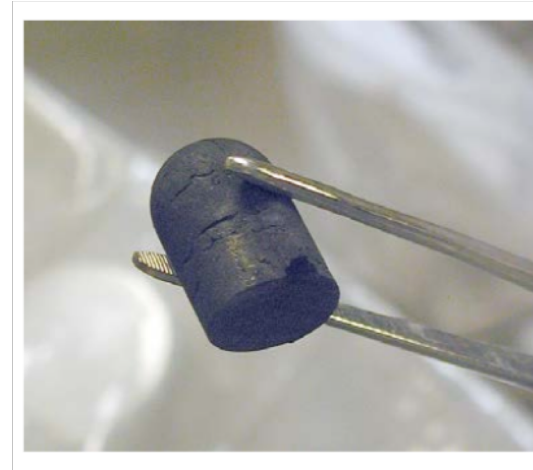


Agenda

- **Introduction to nuclear forensics**
- **Importance of isotope analysis**
- **Introduction to high precision isotope ratio mass spectrometry techniques**
- **Introduction to chemical separation procedures**
- **Examples of some of the measurement capabilities at C-NR**

Nuclear forensic Science

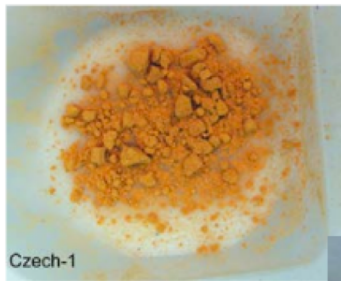
- **Nuclear Forensics - the collection and analysis of nuclear or radiological material to support investigations into the diversion, trafficking or illicit activities involving materials**
- **What is the material**
- **What was its intended use**
- **How was the material made**
- **When was the material produced**
- **Who is associated with the material**



Goal: link nuclear material in question to people, processes, event and/or locations

Nuclear materials

- There are nuclear materials involved at every step of the fuel cycle
 - Many different chemical forms
 - Different isotopic compositions



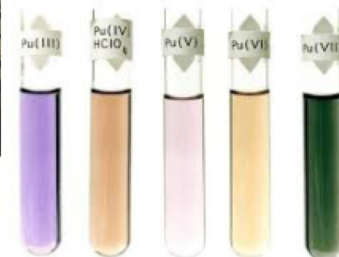
UOC

Fuel



Spent Fuel

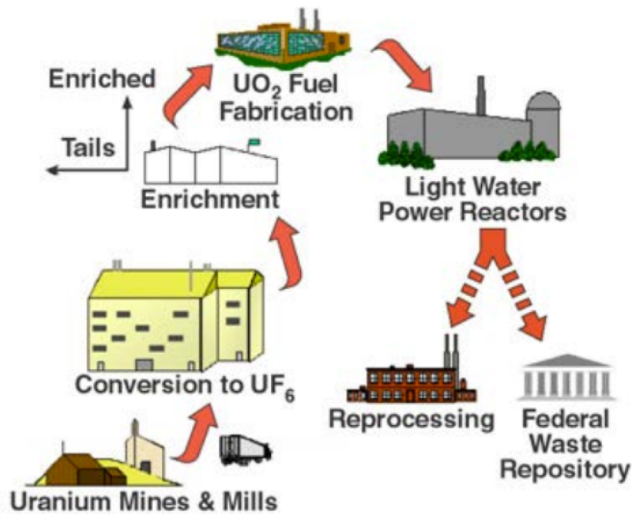
Finished Pu Metal



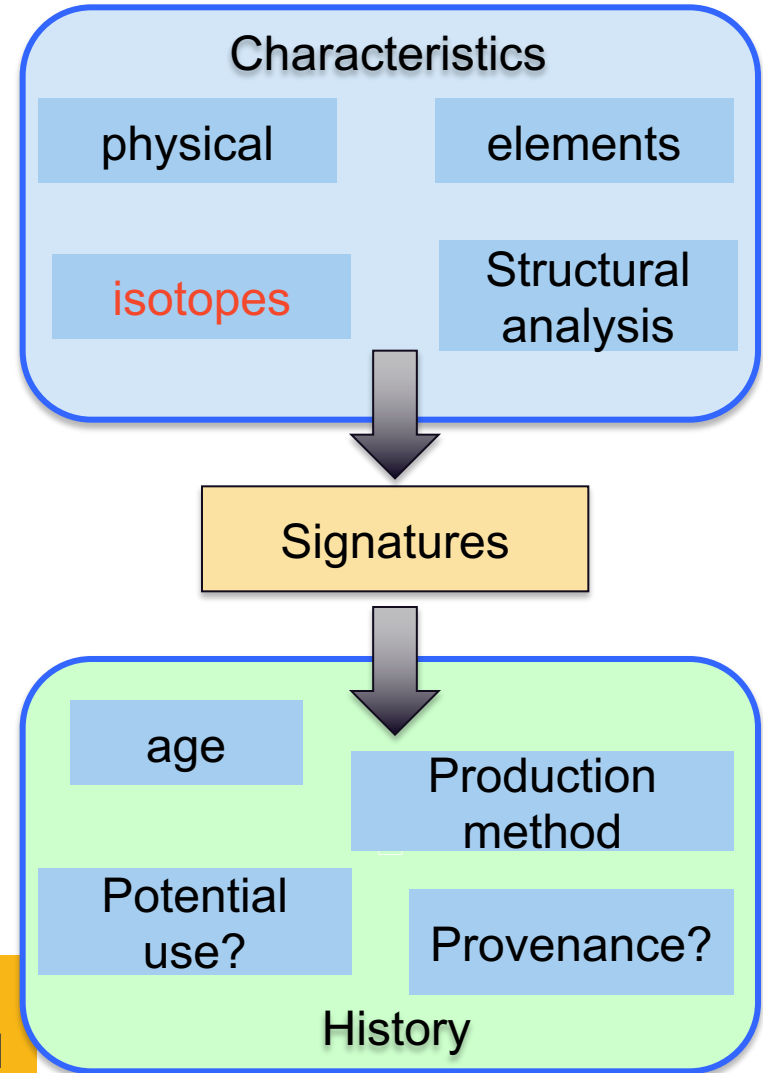
Reprocessed Pu

All of these materials (and many more) have the potential to be the subject of a forensic examination!

Nuclear Forensic - signatures



All nuclear materials have characteristics that are indicative of production history and method



Importance of isotope analysis in forensic characterization

Age of material

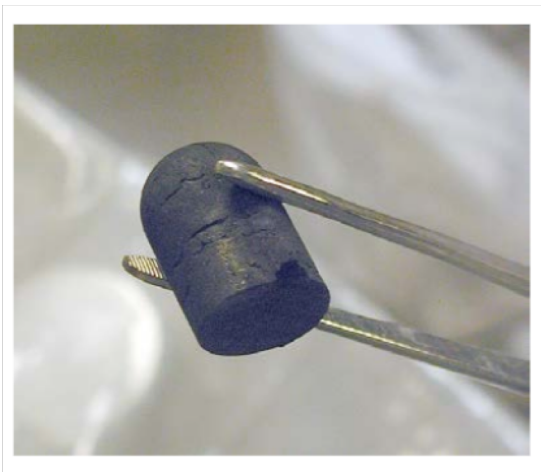
Reactor type

Level of Enrichment

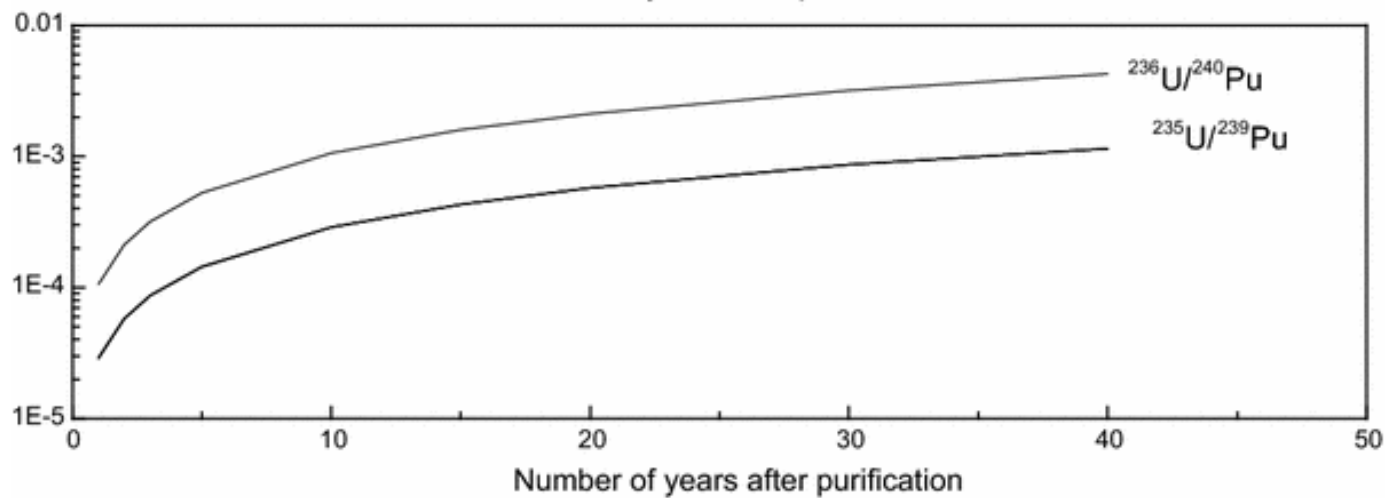
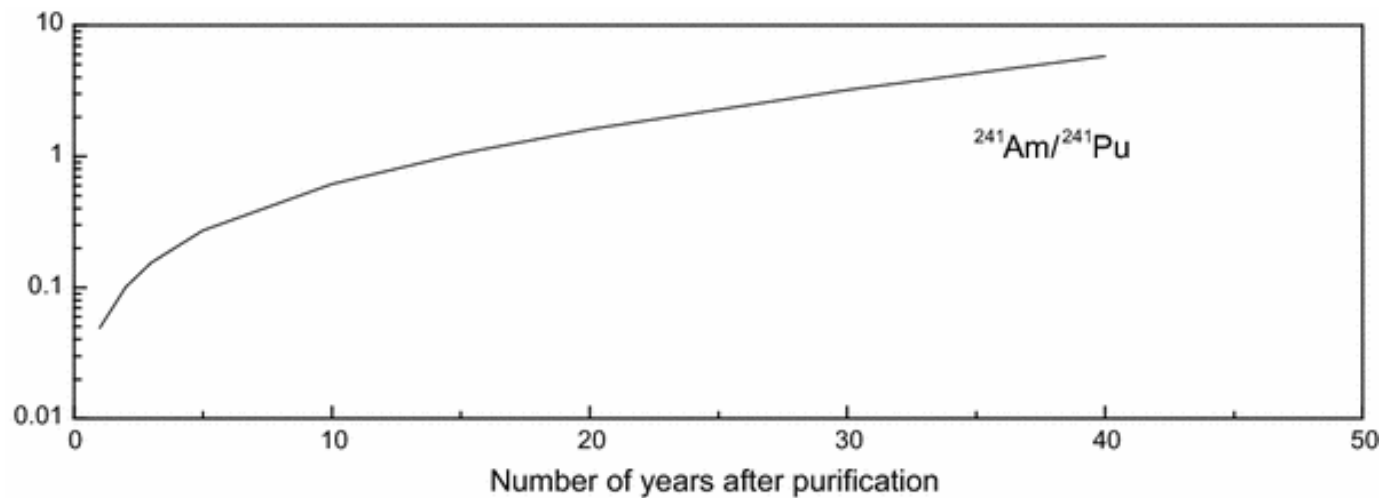
Reactor History?

Origin?

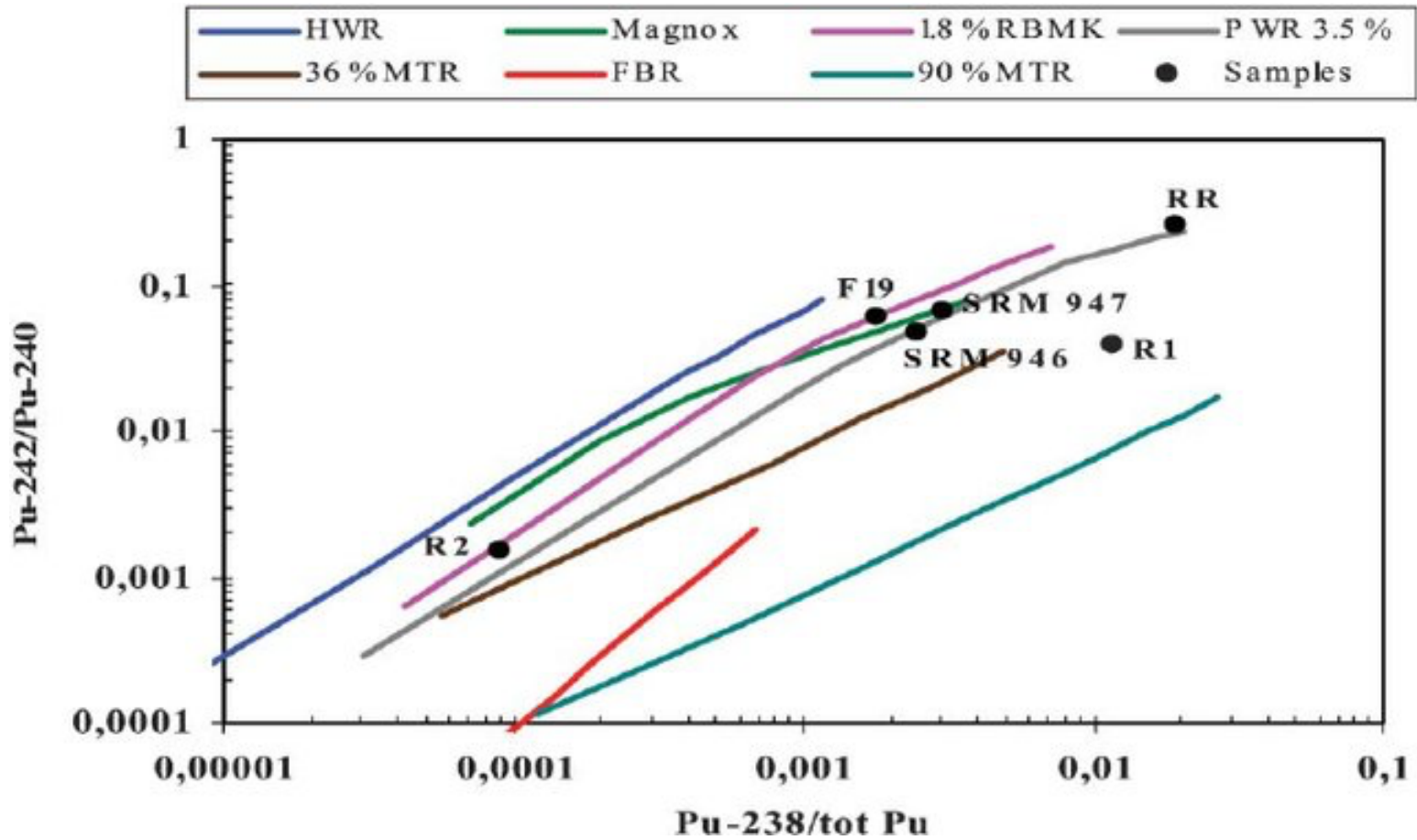
Isotope Analysis



Isotope ratios can provide age of sample



Isotope ratios can provide information on Reactor Type



After Mayer et al. 2005

Importance of isotope analysis in forensic characterization



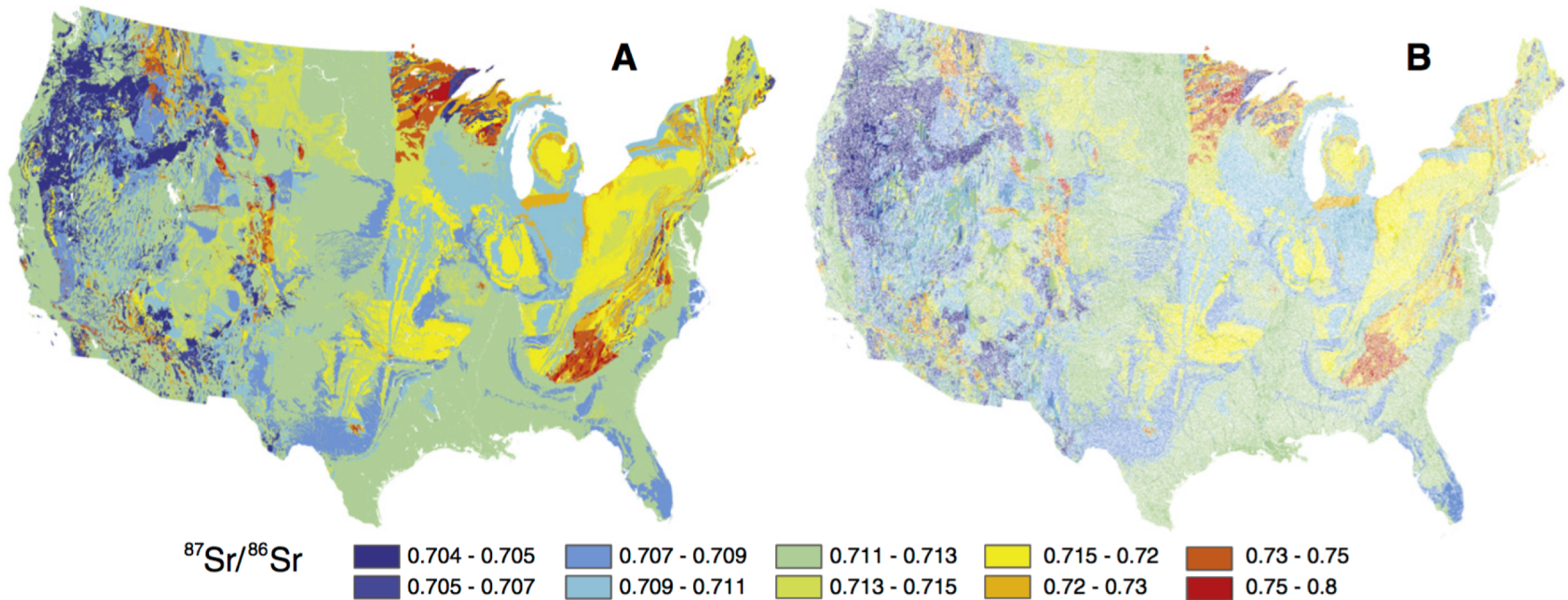
Mine
Type/location

Processing
history?

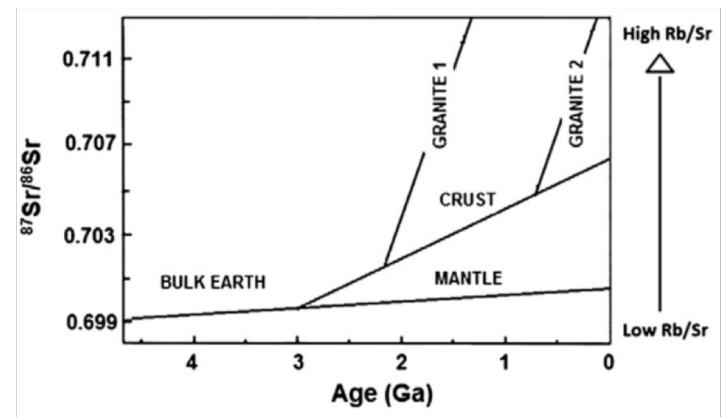
Milling
type/location?

**Isotope
Analysis**

Isotopes can provide Geo-location

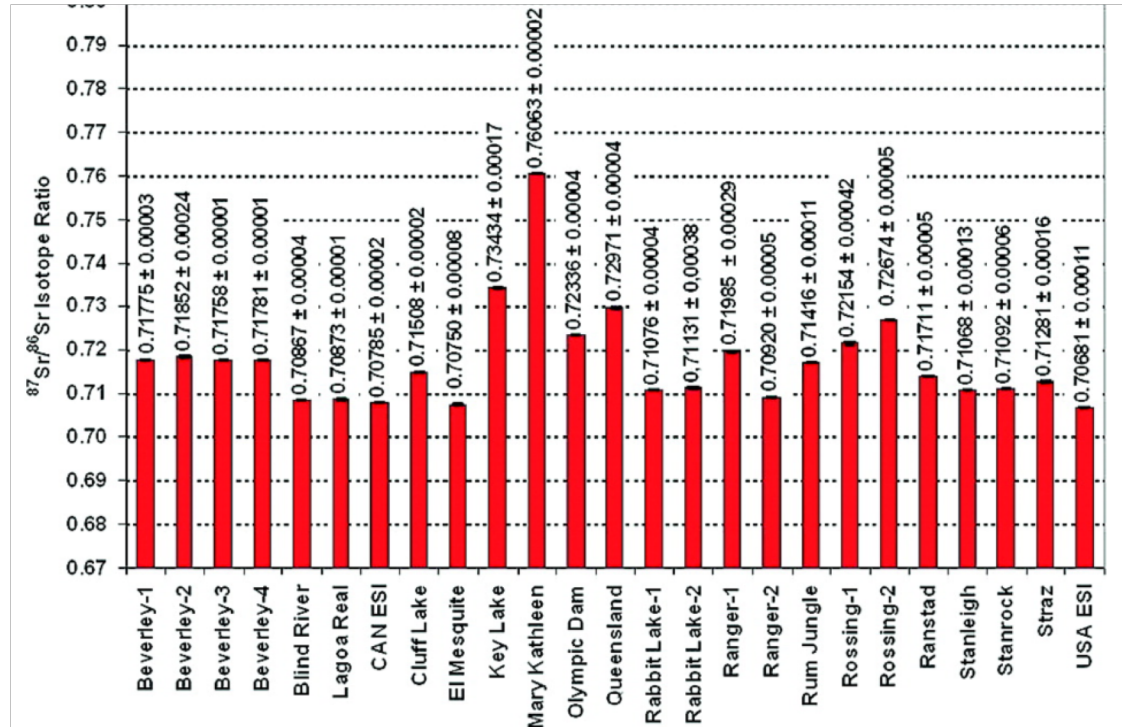


- The $^{87}\text{Sr}/^{86}\text{Sr}$ of a rock determined by
 - The initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio
 - The Rb/Sr ratio
 - The age



Strontium

- $^{87}\text{Sr}/^{86}\text{Sr}$ may provide provenance information
- Sr highly mobile element and can be inhomogeneous within an ore body
- Original value can be easily contaminated by other Sr during processing

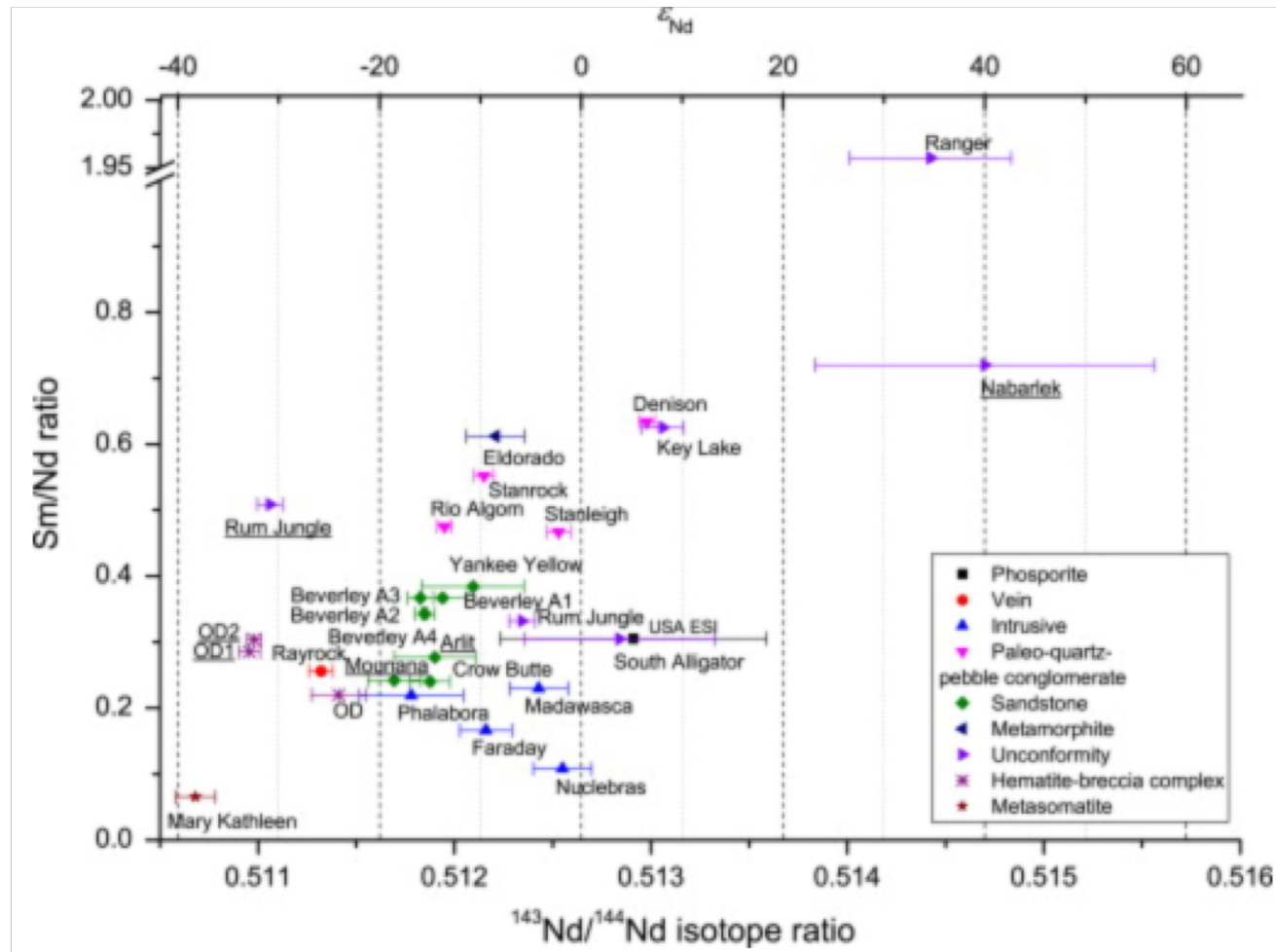


Varga et al. 2009

Neodymium

Has the potential to be used as an isotopic UOC fingerprint

High resolution $^{143}\text{Nd}/^{144}\text{Nd}$ measurements vital to differentiate ores

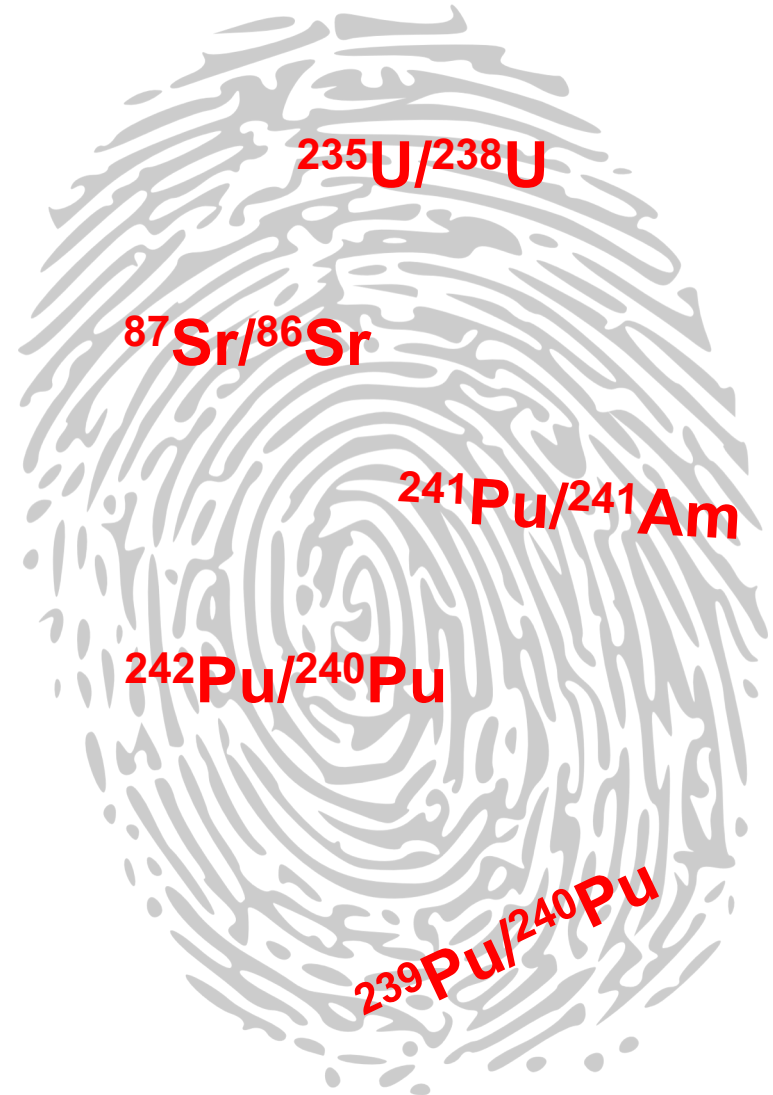


Krajko et al. 2014

Summary

The isotopes present in a sample represent chemical fingerprints, and can provide information on:

- The age of sample
- Geo-Location
- How it manufactured
- Its possible history and potential use



How do we make isotope measurements?

Multi-collector ICP-MS (MC-ICP-MS)

High precision, high accuracy
Isotope ratios (U, Th, Sr, Pb,
Fe, B...)



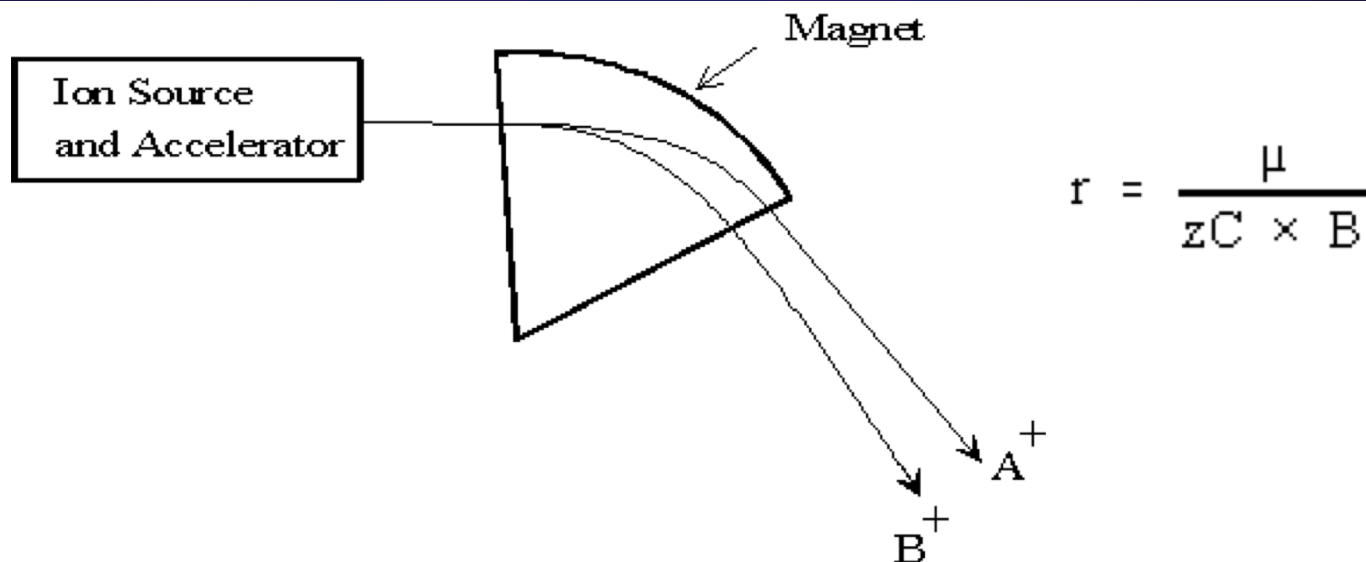
**Sector Field ICP-MS
(SF-ICP-MS)**
ppq – ppm element
conc. and some
isotope ratios



**Multi-collector
TIMS**
Pu, U, Am, Np,
Sr, Nd, others

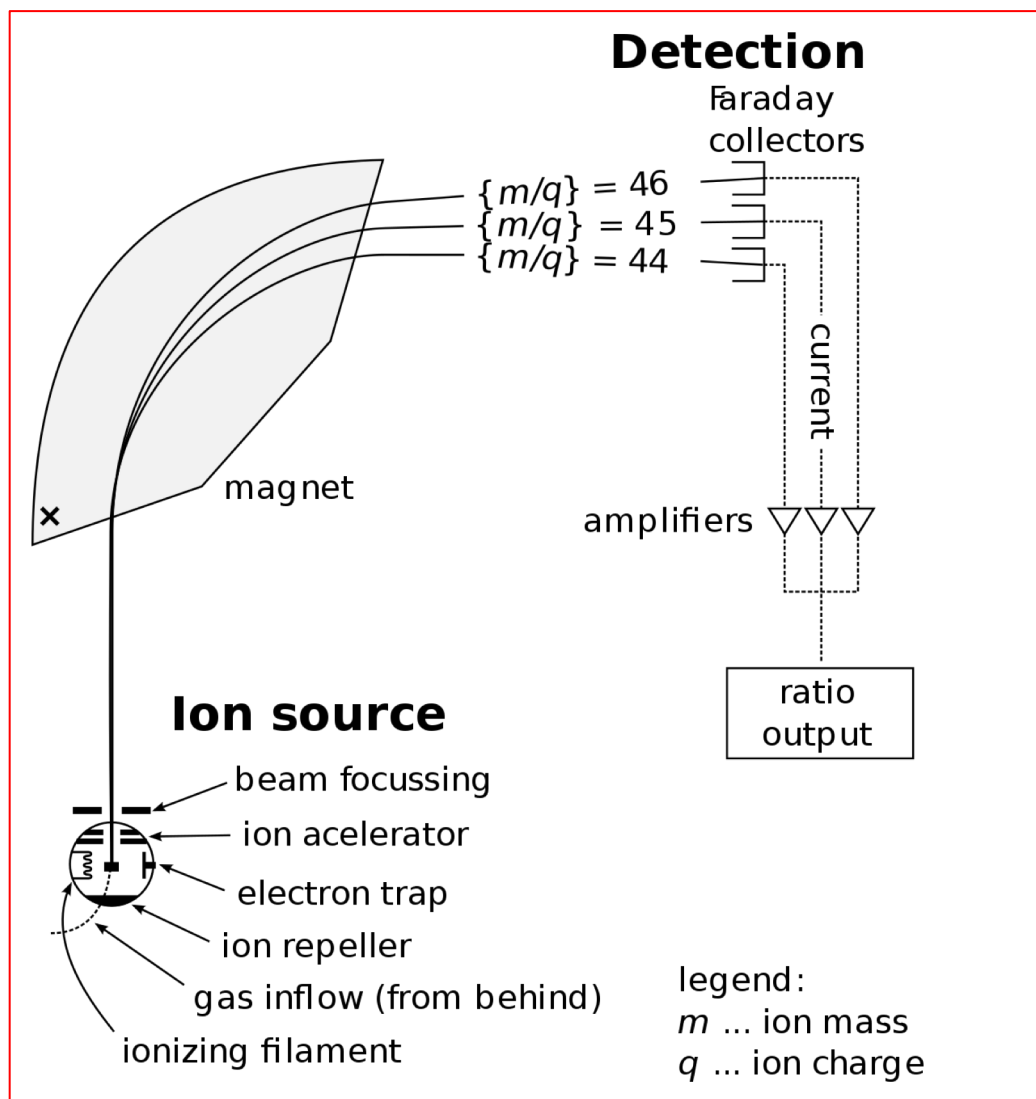


Principles of mass spectrometry – the magnetic sector



- Ions are accelerated from the source region into the magnetic sector by a 8 to 10 kV electric field.
- Since the ions are charged, as they move through the magnetic sector, the magnetic field bends the ion beam in an arc.
- The radius of this arc (r) depends upon the momentum of the ion (μ), the charge of the ion (C) and the magnetic field strength (B).

Principles of mass spectrometry



Ratio measurements

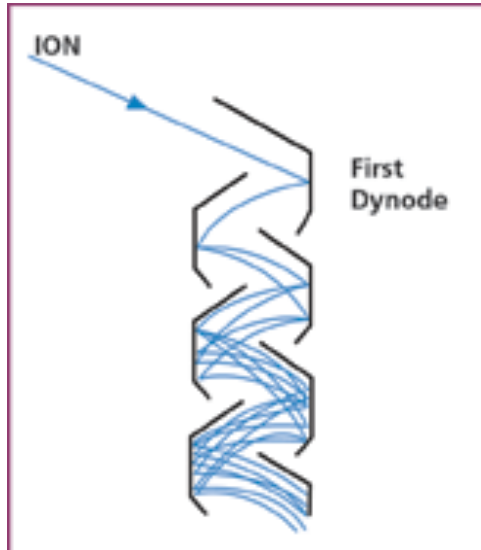
Modern mass spectrometers have movable cups that can be aligned with an individual ion beam.

$$V = C \times R$$

Typical resistor is 10^{11} Ohm

Ratios are calculated by observing voltage on each cups

Principles of mass spectrometry – detection of small beams



Tiny beams cannot be seen over the noise of Faraday cups

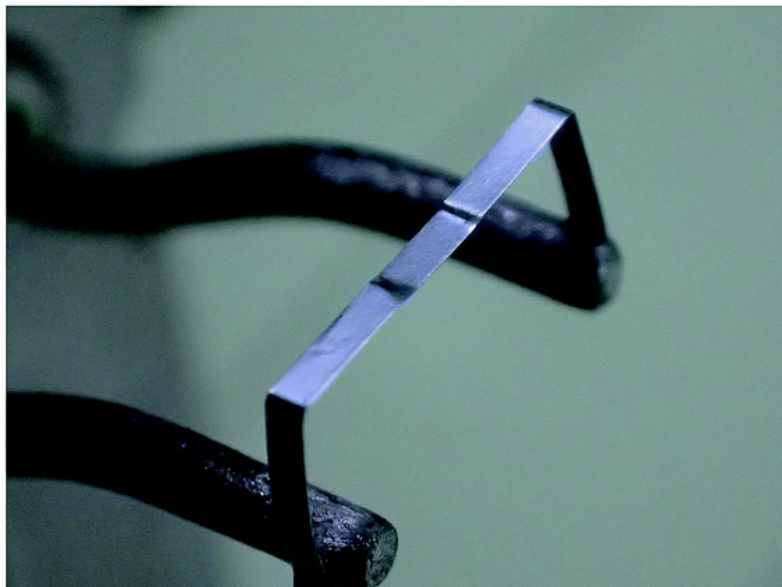
Requires a different detector, that works on the principle of secondary electron emission

When a charged or neutral particle, an ion or an electron, strikes a surface it can cause electrons associated with the outer layers of the surface to be released.

Cascade of electrons can be produced by placing a set of surfaces in series (dynodes)

Producing amplification from a single ion

Thermal ionization mass spectrometry



Sample is loaded onto a filament as a small drop of solution

Filament heated to evaporate and ionize the sample

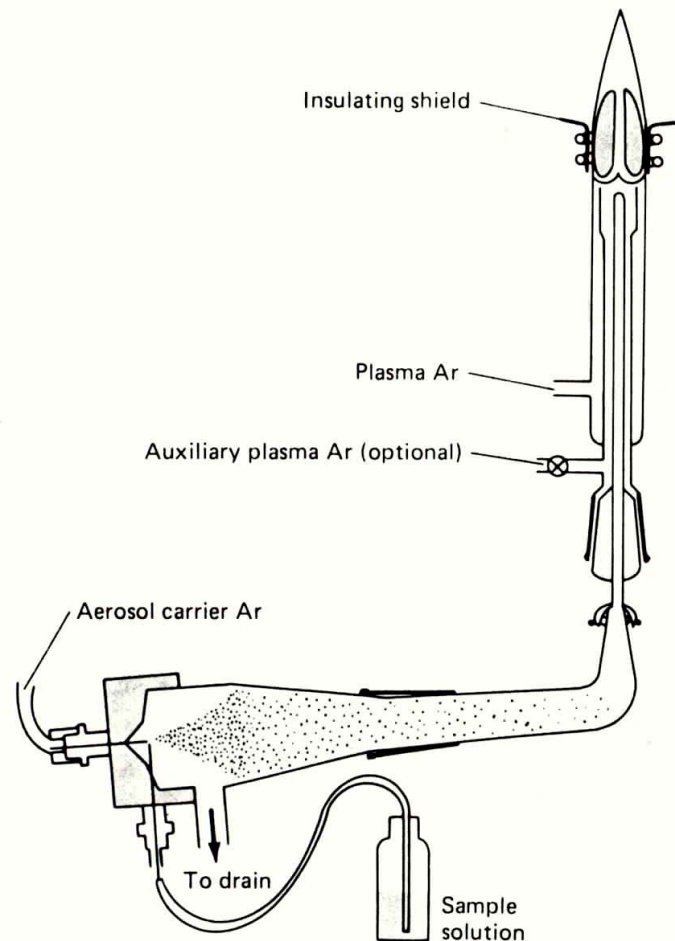


High precision analyses

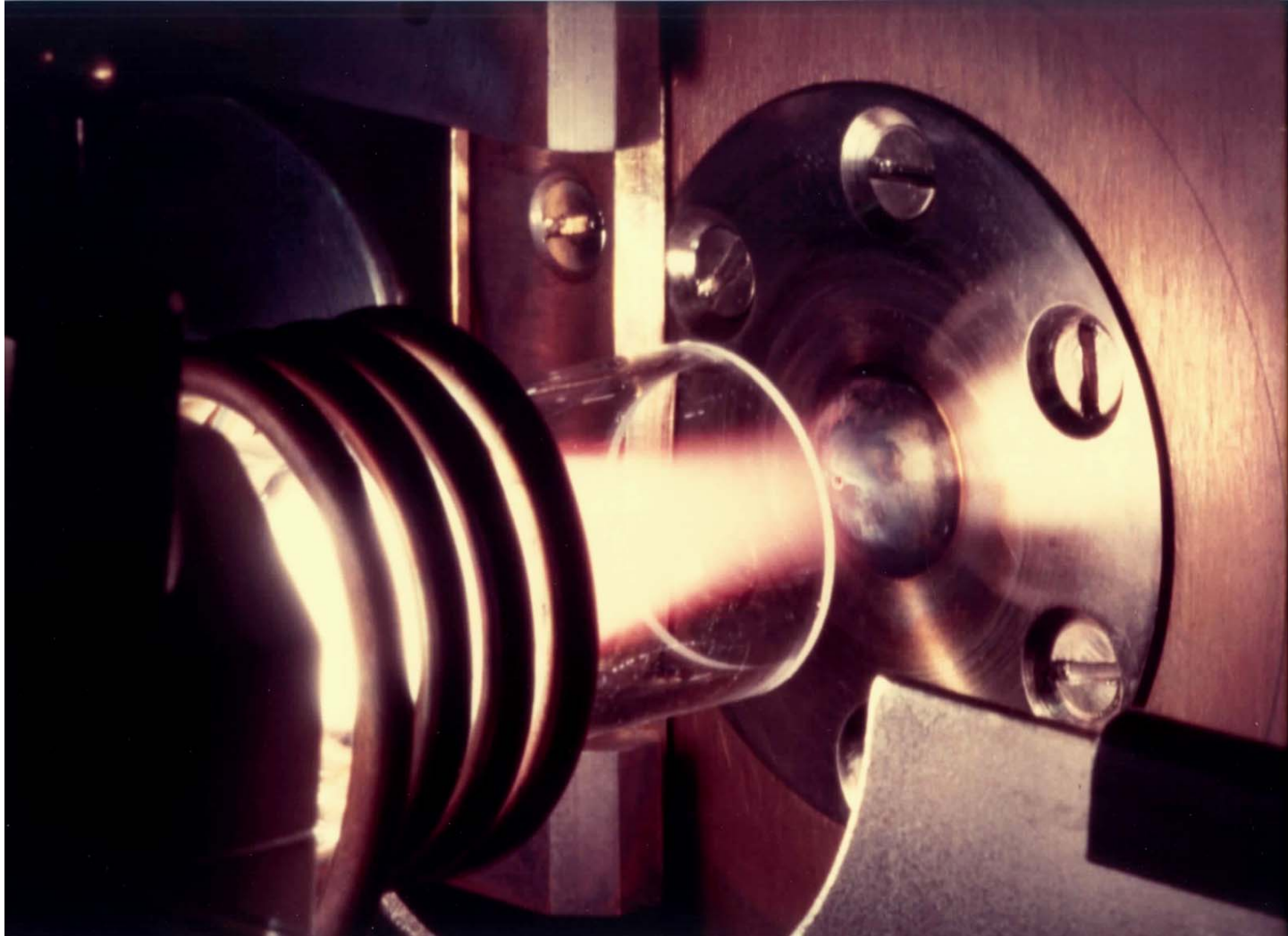
Detection of isotope ratios in very small amounts of material

Inductively Coupled Plasma

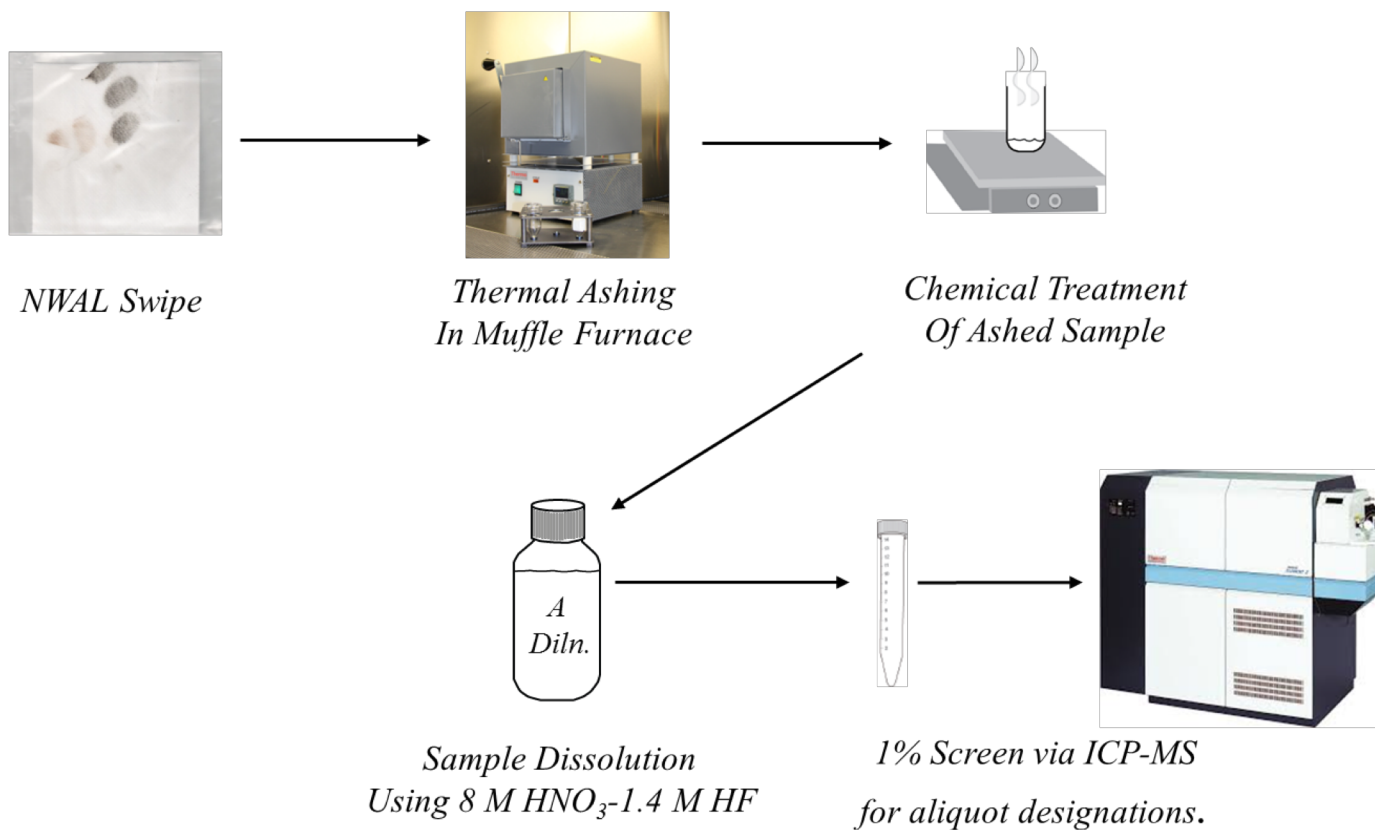
- A plasma is a hot, partially ionized gas.
- The ICP is an argon-based, radio frequency plasma.
- The input rf frequency is either 27 or 40 MHz at powers from 1 to 2 kW.
- The plasma is formed and contained in a three tube quartz torch.
- The temperature in the central analyte channel ranges from about 6000 to 8000K.
- At these temperatures most elements are largely atomized and ionized



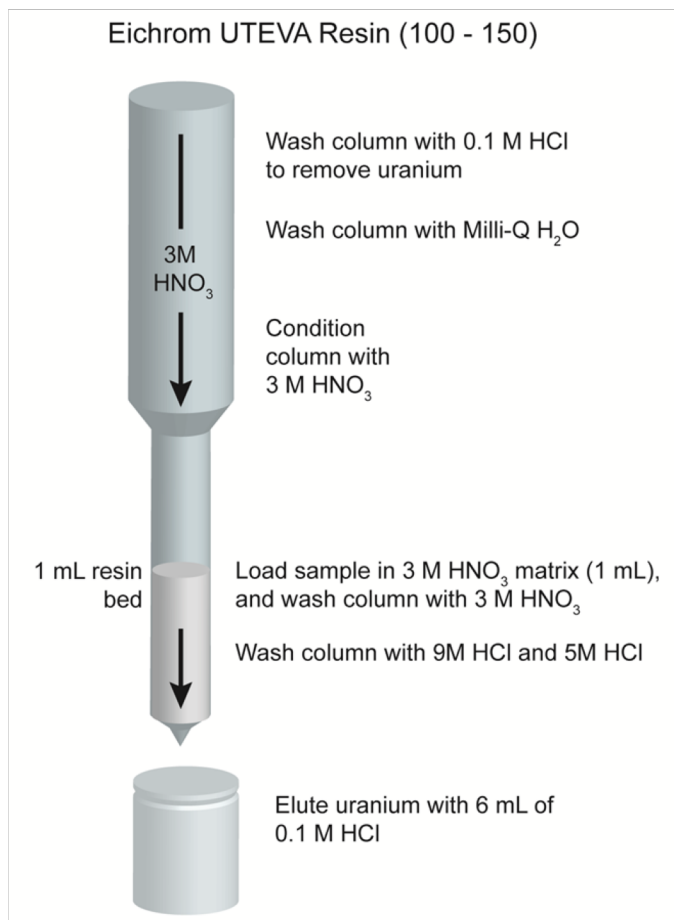
Inductively Coupled Plasma



Samples require separation chemistry prior to analysis



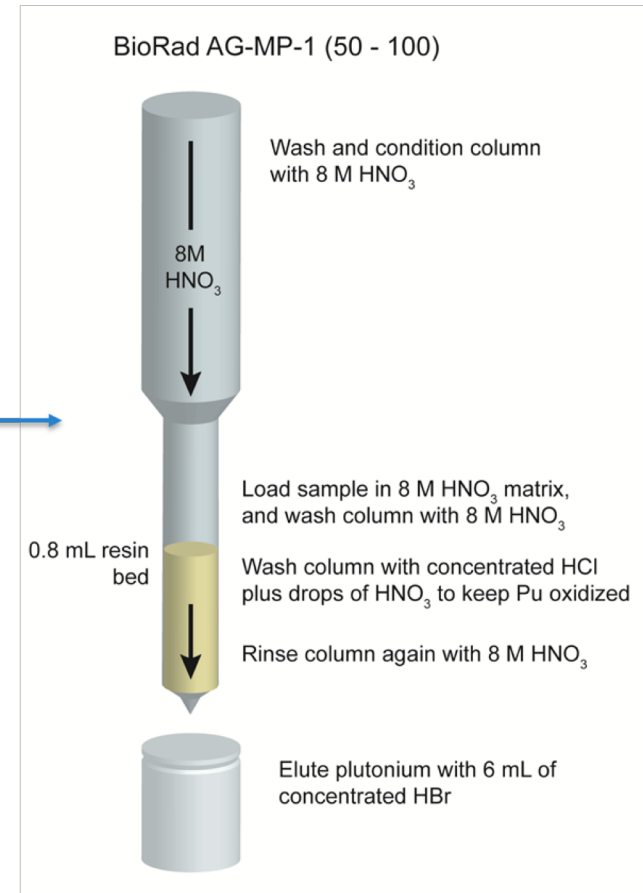
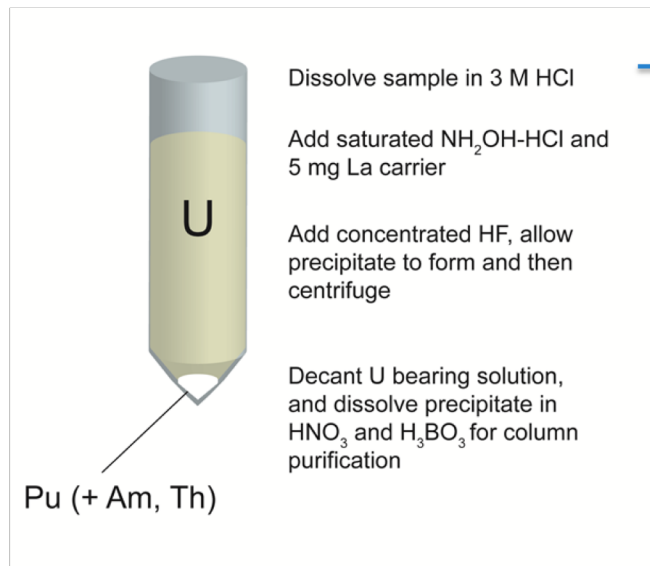
Uranium Purification



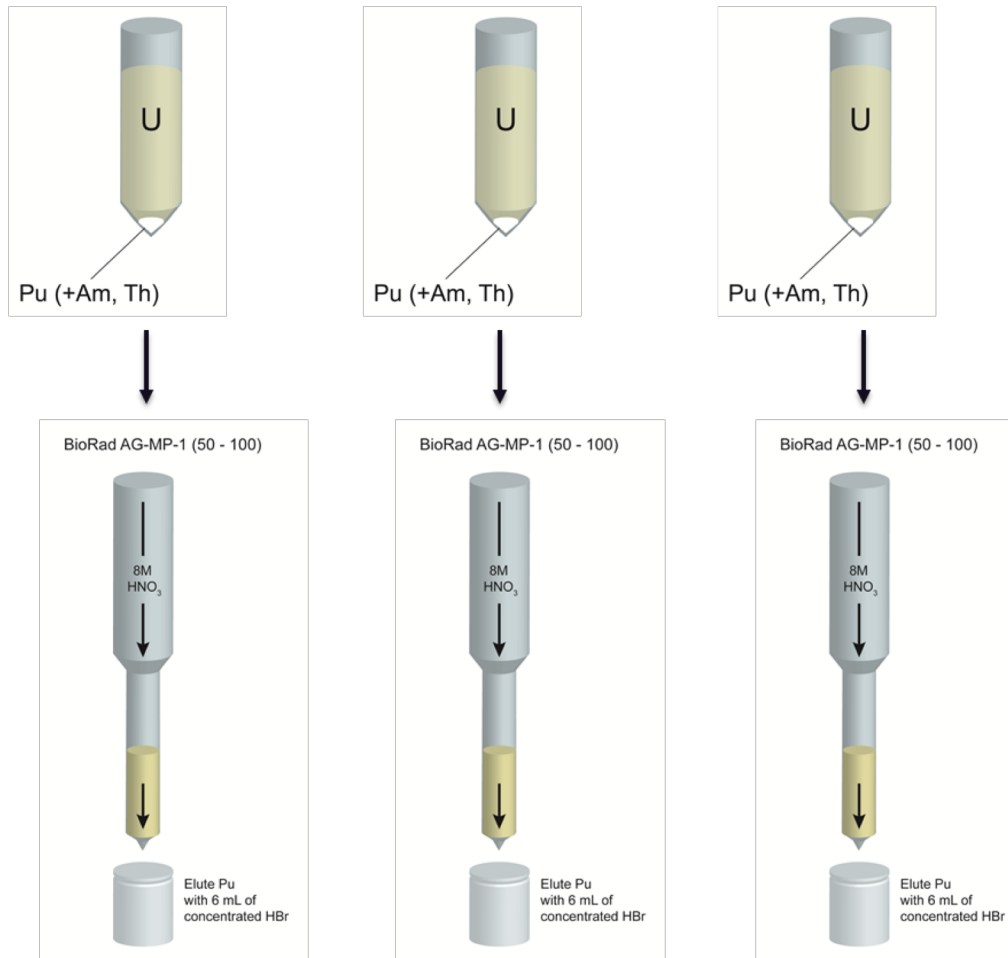
- After 1% screening of solutions, a subject matter expert will provide the aliquot sizes for traced and untraced uranium analyses
- Aliquots are weighed into Teflon vials, dried, and reconstituted in 3 M HNO₃
- An Eichrom UTEVA column is used for the chemical purification of uranium

Plutonium Purification

- Plutonium aliquots are weighed into Teflon vials, dried, reconstituted in 3 M HCl, and transferred to a Falcon tube
- A combination of a LaF_3 precipitation and an anion exchange column is used for Pu purification



Plutonium Purification



- For samples with high concentrations of U, multiple LaF₃ precipitations and anion exchange columns are used
- Typically, between one and three separations are used to purify trace Pu from nanogram to microgram quantities of U

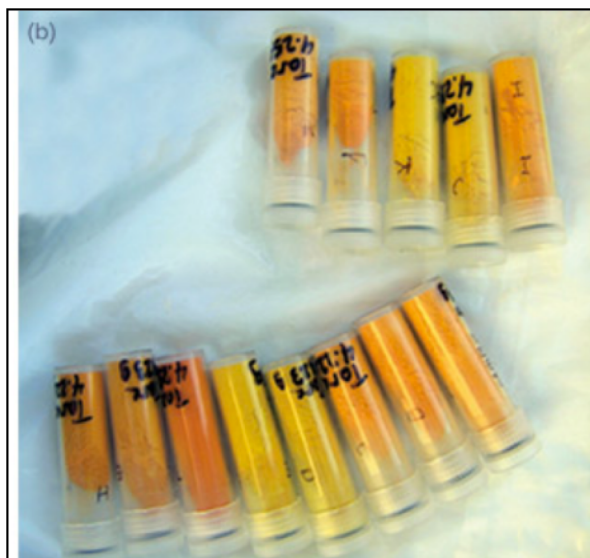
CN-R capabilities

RC-45 Clean Chemistry and Mass Spectrometry Facility



- Class 10,000 hallways and laboratories
- Class 100 work areas and exhausted laminar flow work cabinets
- Perchloric acid capability
- Ultra low-level sample handling
- Co-located clean radiochemistry and analysis capability
- Isotopic and elemental analysis

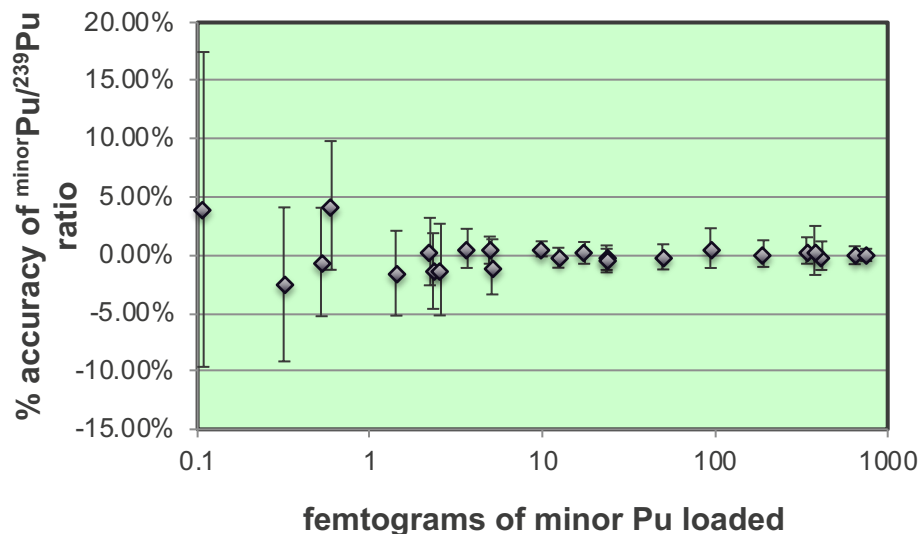
Mass spectrometry capabilities



Example 1 – measuring tiny amounts of Plutonium

Technique advantages

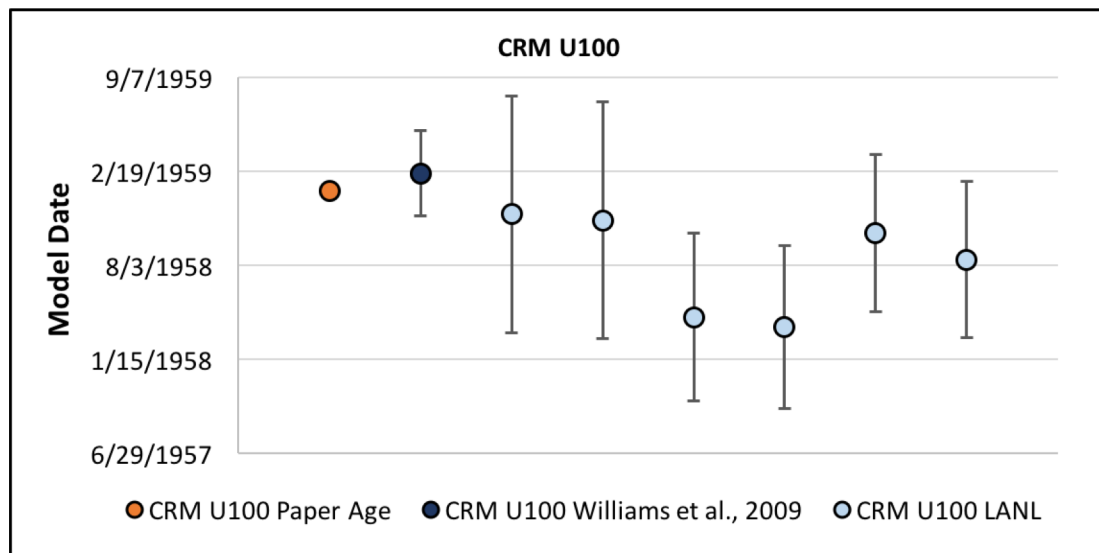
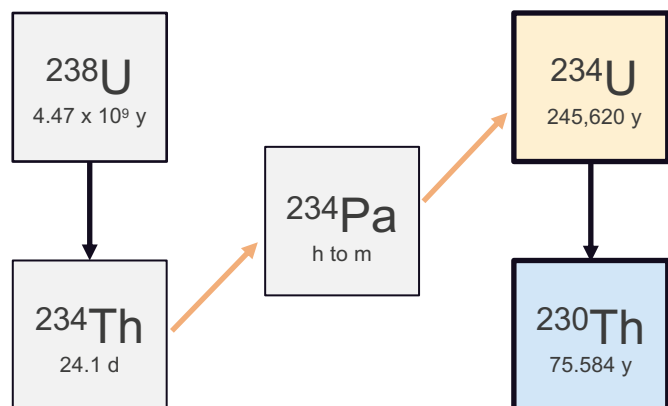
- Ultra-low noise detectors (<3 cpm), can detect single counts of an isotope
- Small load sizes (pg and below)
- High accuracy and external reproducibility of minor ratios
- Mass bias correction eliminated by total evaporation technique



- TIMS method combines an array of multiple ion counters with total evaporation
- Resin bead loading technique used to maintain >1% ionization efficiency
 - Two fold increase compared to direct load techniques
- femtogram detection limits – maintaining ratio accuracy
- Requires clean chemistry to ensure low “organics” background

Example 2 – U-Th Chronology of Uranium samples

^{230}Th - ^{234}U chronometry
commonly used for nuclear forensics



Thank you

