Analysis of Beryllium in Autopsy Tissues from Nuclear Weapons Site Worker

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Outline

- Beryllium in Autopsy Tissues – Nuclear Weapons Site Worker
- Comparison of Laboratory Results
- Methods of Analysis
- Summary

USTUR

- U. S. Transuranium and Uranium Registries (USTUR)
  - Studies uptake, translocation and biokinetics of actinides (U, Pu, and Am) in humans).
  - Determined beryllium concentration in human bone and soft tissue of occupationally exposed individual.
- Partner with SRNS, LLC (Savannah River Nuclear Solutions, LLC) Analytical Laboratories at Savannah River Site
- Multiple Laboratory Analyses on Digested Aliquots
  - Savannah River Nuclear Solutions, LLC IH Laboratory (SRS)
  - Laval University (LU)
  - Northern Arizona University (NAU)

Case Study: Nuclear Weapons Site Worker - #817

- Voluntary partial body donation to USTUR
- Rocky Flats engineer with primary exposure to Pu
  - ^{239}Pu concentration in tissues (Bq / kg wet)
    - *Lung: 38.0 ± 0.1*
    - *Femur (shaft): 2.2 ± 0.2*
- 37 year self-reported Occupational Exposure to Beryllium
  - 1963-1990
- Died in 2004 at age 76 from esophagus cancer (primary cause of death)

USTUR: Beryllium Preparation

- Digested Human Autopsy Tissue
  - Method: Whole lung tissue or femur tissue was ashed, digested with concentrated nitric acid, peroxide, and hydrofluoric acid
  - Diluted to volume with hydrochloric acid
  - Lung: 0.314 g tissue g^{-1} 6M HCl
  - Femur: 0.174 g tissue g^{-1} 6M HCl

SRS: Beryllium Preparation and Analysis

- SRS Digestion
  - An aliquot from USTUR of digested tissue (lung or femur) sample solution was concentrated with additional acid digestion by hydrofluoric and hydrochloric acid to remove trace nitric acid, with column separation to clean-up the matrix.
  - Sample analysis included matrix spike, laboratory control spike/laboratory control spike duplicate to demonstrate recovery during the digestion / concentration / clean-up stages.
- SRS Analysis by ICP-OES
  - Confirmation by SRS Q-ICP-MS

Intercomparison on Be Analysis

	Beryllium concentration, µg/kg, wet tissue	Method
Lung Tissue – SRS	0.224 µg Be/kg ± 0.028 (1σ)	ICP-OES
Lung Tissue – NAU	0.104 µg Be/kg ± 0.005	Q-ICP-MS
Lung Tissue – LU	0.072 µg Be/kg ± 0.004	Q-ICP-MS
Bone Tissue - LU	0.130 µg Be/kg ± 0.007	Q-ICP-MS

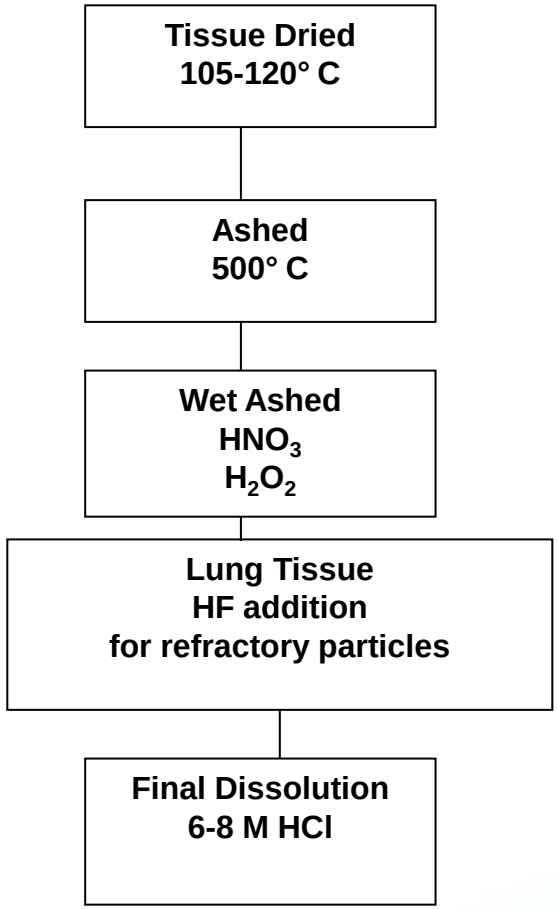
Results of SRS Analysis

- Lung Tissue
  - Acceptable Quality Control through the digestion/analysis
    - Lab Blank was ~ 5x less than the Reporting Limit (no cross-contamination)
    - LCS/LCSD were recovered at 106.4% / 109.16%
    - Matrix Spike was recovered at 102.84%
  - Concentrated sample ~7x
  - Replicate analyses agreed within 3.9%
  - ICP-OES wavelength agreement 2.6% (234.861nm and 313.107nm)
  - Results were confirmed by SRS Q-ICP-MS within 11.8%
- Bone Tissue
  - Diluted digested sample 10-fold to meet Quality Control criteria in matrix, reported less than reporting limit (< 26.1 µg Be/kg)

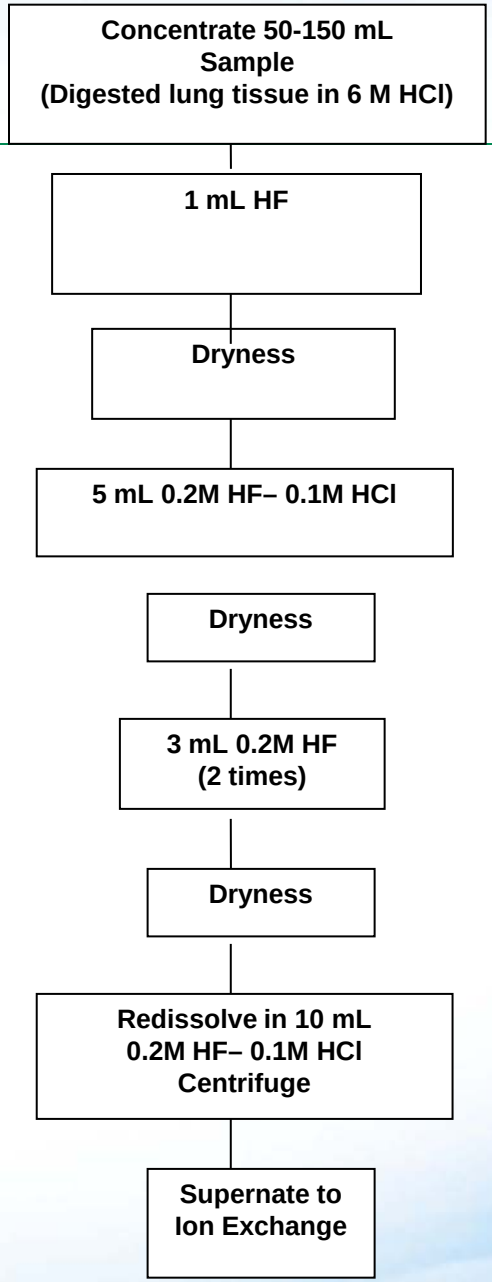
SRS Replicate Comparison

Lung tissue, USTUR digested solution	ICP-OES Reported, µg Be / L 234.861nm	ICP-OES Confirmed, µg Be / L 313.107nm	Q-ICP-MS Confirmed, µg Be / L	ICP-OES Report Limit, µg Be / L
Aliquot 1	0.076	0.078	0.067	0.067
Aliquot 2	0.079	0.080	0.080	0.073

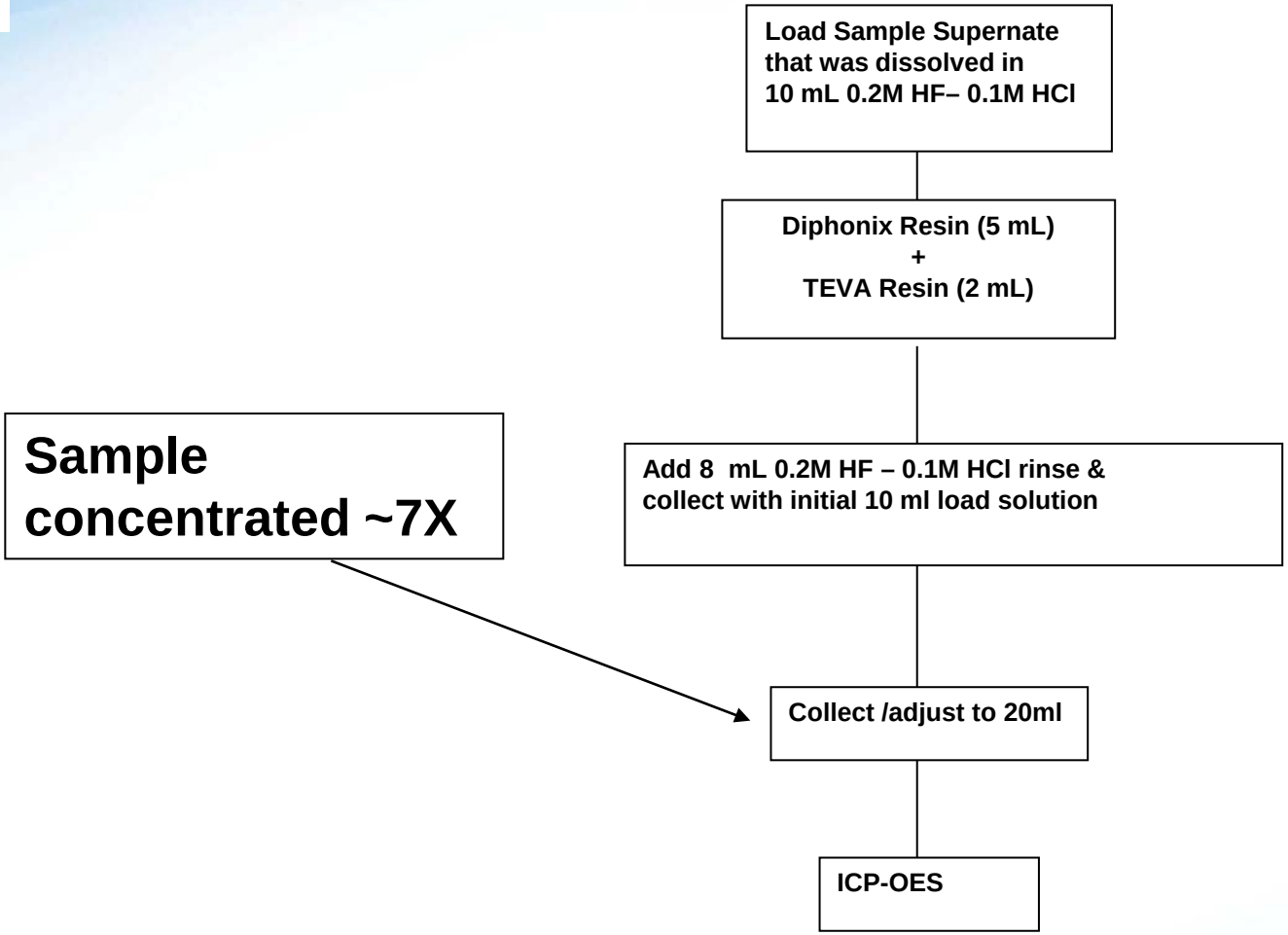
Digestion / Preparation
USTUR



Digestion / Preparation SRS

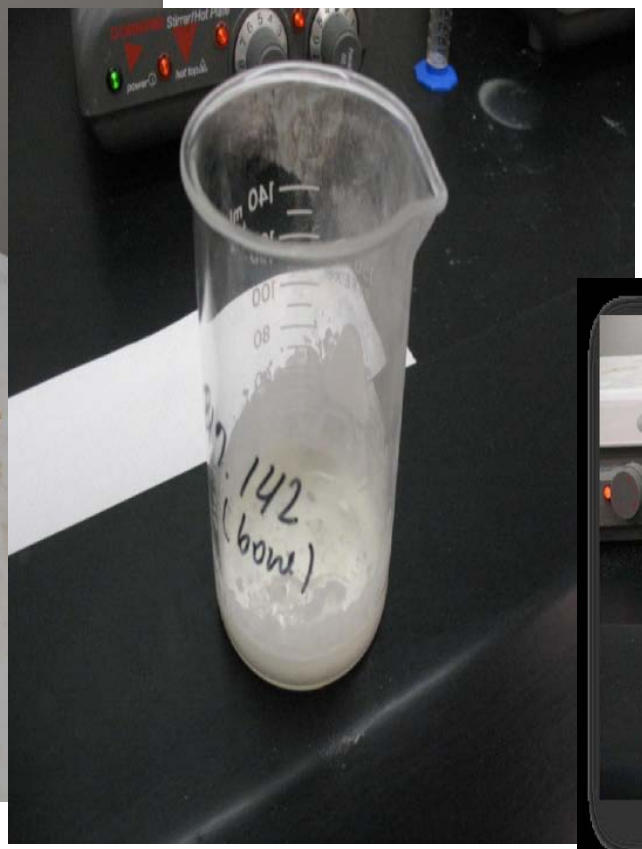


Column Separation - SRS

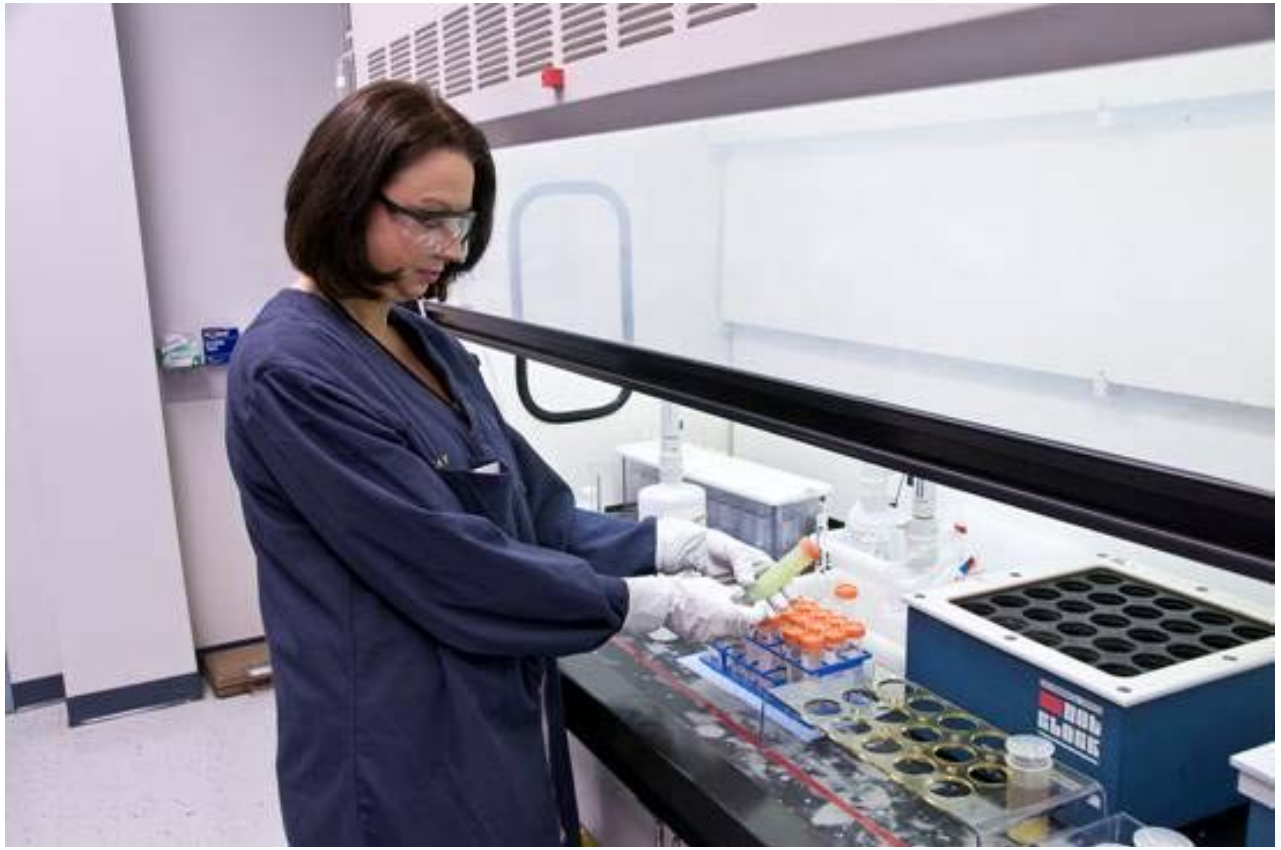


New Method for Removal of Spectral Interferences for Beryllium Assay using Inductively-Coupled Plasma Atomic Emission Spectrometry", S.L. Maxwell, M. Bernard, Talanta 76 (2008), 432-440

USTUR Sample Preparation

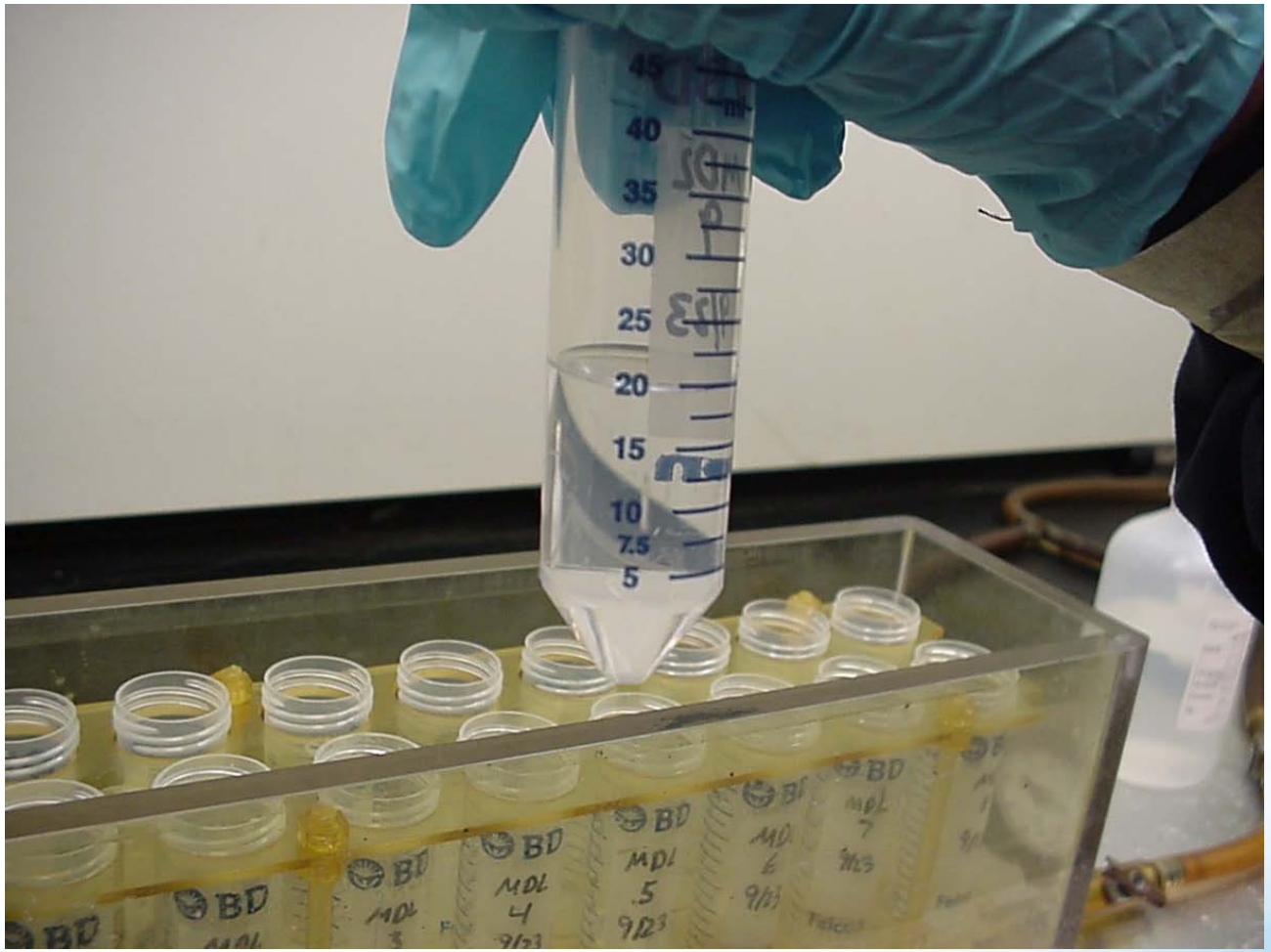


Hot Block Digestion - SRS

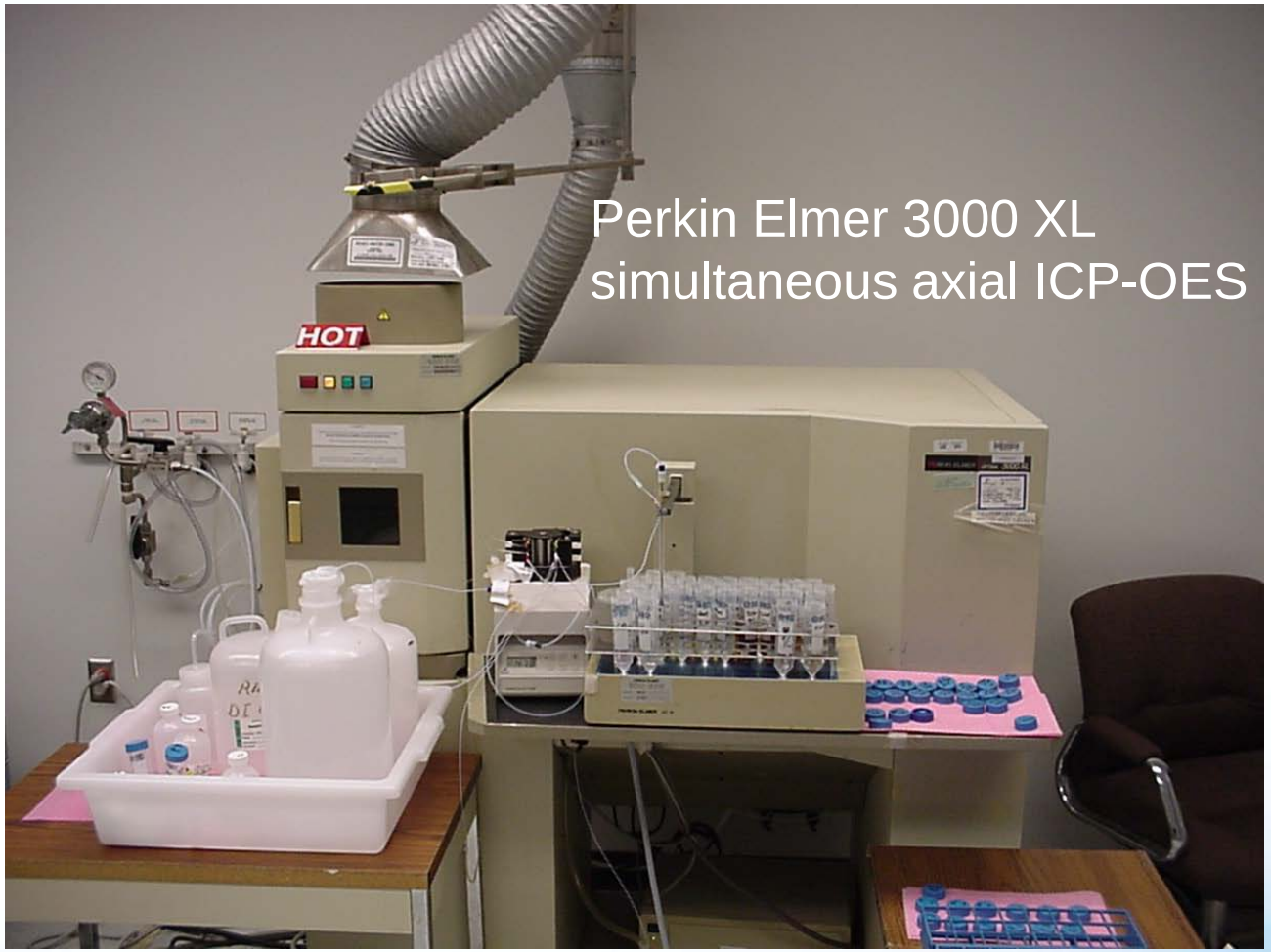


SRS Diphonix + TEVA resin Clean-up

SRS – Ready for analysis by ICP-OES



SRS – ICP-OES Analysis of Beryllium



Perkin Elmer 3000 XL
simultaneous axial ICP-OES

Analysis by Northern Arizona University: Q-ICP-MS

- Diluted USTUR-digested sample 20x
- Analysis
  - Thermo X2 Series ICP-MS
  - Apex Q introduction system for high sensitivity
  - Li-6 as Internal Standard

Analysis by Laval University – Q-ICP-MS

- Diluted digested sample 25x
- Analysis
  - Varian model 800 without Collision Reaction Interface
  - Sheath gas allows high salt matrix with limited effect on plasma, allowing analysis of bone tissue
    - Similar to Agilent's HMI
  - Rh as Internal Standard
    - Rh is close to Be in ionization potential
    - High Sensitivity instrument capable of analyzing in heavy matrix

Summary

- The analysis of beryllium in difficult matrix of human tissue can be done by different digestion and analysis techniques.
- Further testing is needed to resolve differences.
  - QC was in control for all techniques.
- Possibilities to investigate:
  - Homogeneity of sample, dissolution of all particulate
  - Ensure no beryllium cross-contamination of sample
  - Concentrate larger aliquot of sample for ICP-OES to obtain result approximately 2X Report Limit
  - Re-verify signal suppression correction

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