

The U.S. Transuranium and Uranium Registries: forty years' experience and new directions in the analysis of actinides in human tissues

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*“Learning from Plutonium
and Uranium Workers”*

This Presentation

- **USTUR: Who We Are and What We Do**
- **Human Tissue Analysis: Present and Future**
- **Mass Spectrometry for Actinide Analysis in Human Tissues**
- **Actinide Bio-Imaging in Human Tissues**



About the USTUR

- **Founded:** 1968 at the Hanford Occupational Health Foundation (HOHF), Richland, WA as the U.S. Transuranium Registry (USTR)
- **Mission:** to follow up occupationally exposed workers, from exposure through full lifespan, by studying the biokinetics (uptake, translocation and retention), and tissue dosimetry of the actinides (Pu, Am and U)
- **Registrants:** to date, **325 former workers** from various U.S. weapons or other sites with known history of exposure to actinides **voluntarily donated** their tissues (**289**) or entire bodies (**36**) for **scientific research**



Human Tissue Analysis

- **Sample Ashing/Acid Dissolution/Aliquot Selection**
- **Actinide Separation (ion-exchange, extraction chromatography)**
- **Electrodeposition (for alpha spectrometry)**
- **Alpha Spectrometry (AS) or Kinetic Phosphorescence Analysis (KPA)**
- **^{238}Pu , $^{239+240}\text{Pu}$, ^{241}Am , $^{234,235,238}\text{U}$ activities (AS) or total U mass (KPA)**
- **To date, >10,000 tissue samples are analyzed**



Can We Do Better?

- Sample Ashing/Dissolution: **Microwave Technology**
- Actinide Separation: **Vacuum-Assisted Extraction Chromatography**
- AS and KPA: **Mass Spectrometry**
- ^{238}Pu , $^{239+240}\text{Pu}$, ^{241}Am , $^{234,235,238}\text{U}$, ^{232}Th : $^{240}\text{Pu}/^{239}\text{Pu}$, ^{241}Pu , precise U isotopic measurements (DU), ^{236}U
- **Laser Ablation Mass Spectrometry: Study of Actinide Distribution in Tissues**



Inductively Coupled Plasma Mass Spectrometry (ICP-MS) for Actinides and Isotopic Ratio Measurements

Collaboration with Northern Arizona University



Dr. Michael E. Ketterer

**Professor, Department of Chemistry and
Biochemistry, Northern Arizona University
Flagstaff, AZ, USA**



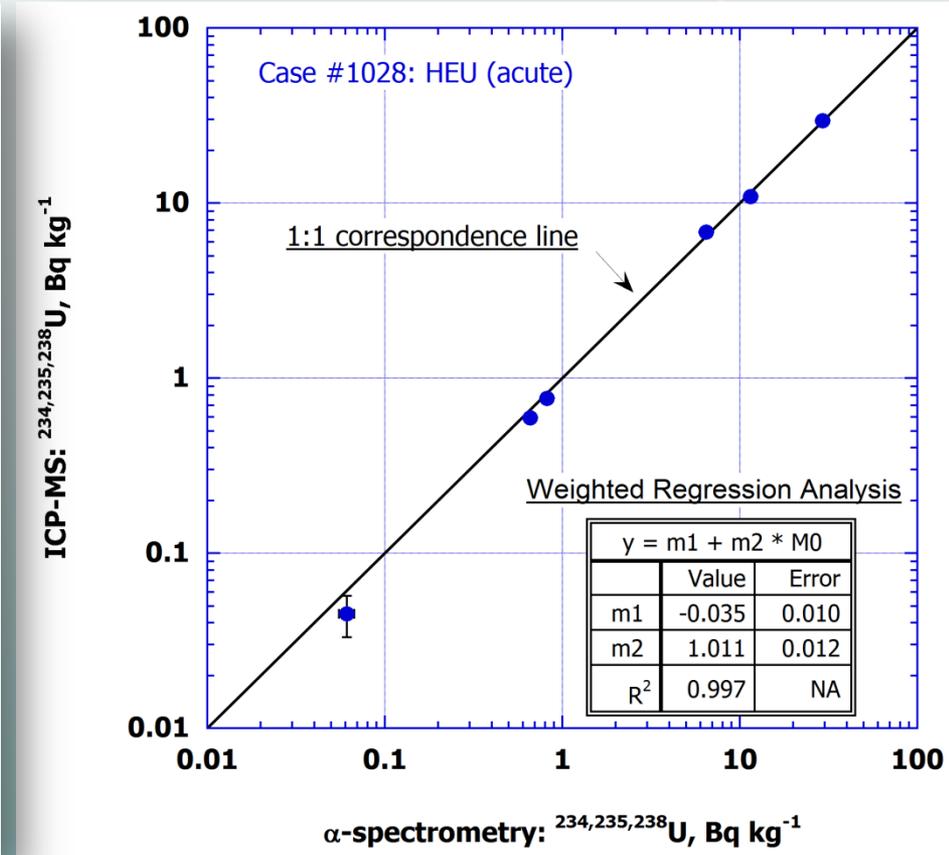
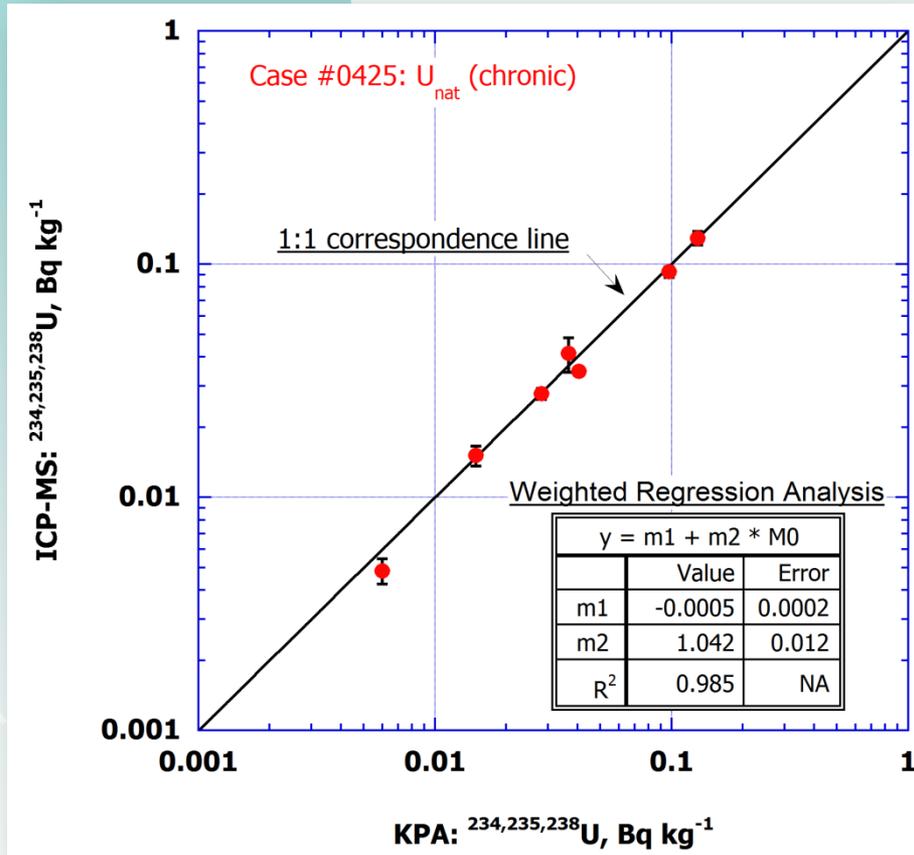
Samples: SF-ICP-MS vs. AS & KPA

- 0269: Hanford – 1956 Acute Inhalation – Soluble ^{239}Pu – Chelated
- 0425: Rocky Flats – 1965 Acute Inhalation (Fire) – Refractory ^{239}Pu
- 0720: Rocky Flats – 1965 Acute Inhalation (Fire) – Refractory ^{239}Pu
- 1028: Oak Ridge – 1954 Acute Inhalation – Highly Enriched Uranium

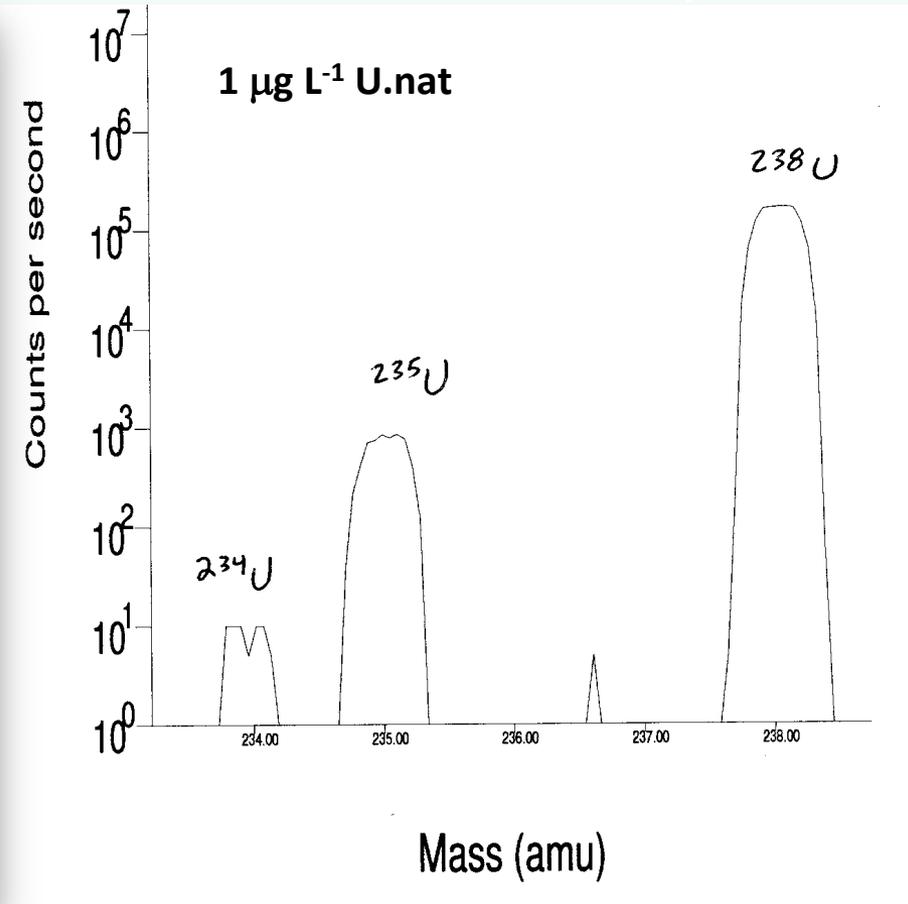
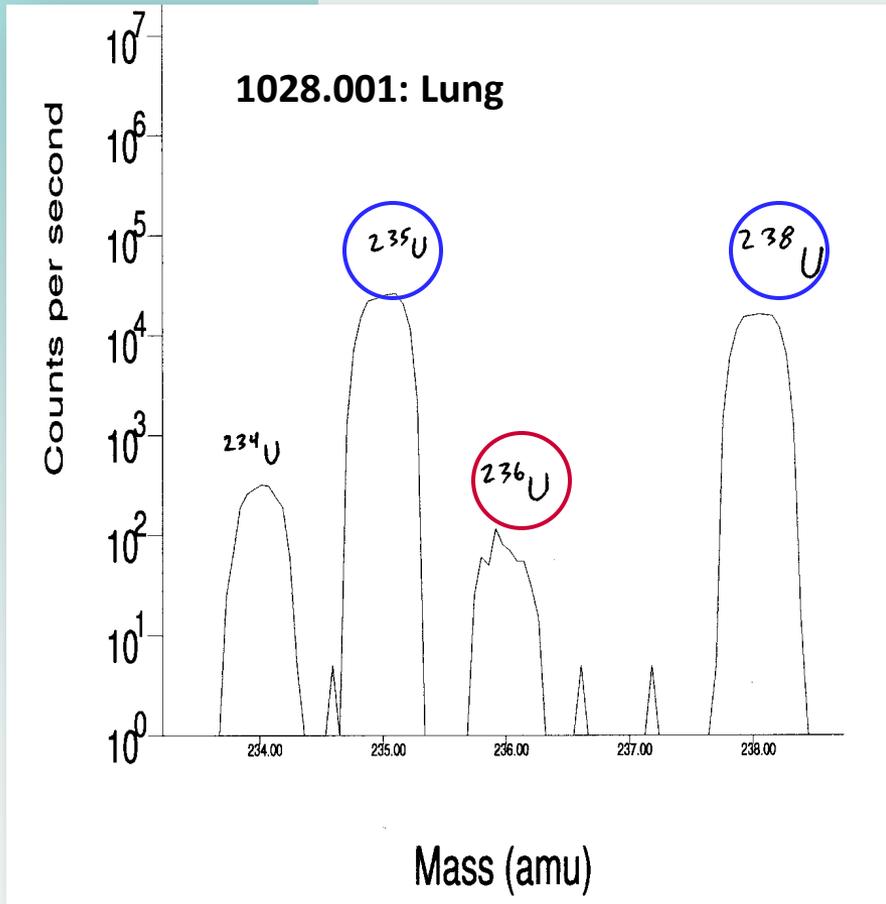
Case # (Age, y)	Follow-up (y)	# of Samples (Bone)	USTUR (AS, KPA)		
			U	Pu	Am
0269 (79)	38	4 (2)	x	o	o
0425 (83)	29	7 (3)	o	o	o
0720 (83)	40	2 (0)	x	o	o
1028 (86)	39	6 (3)	o	x	x



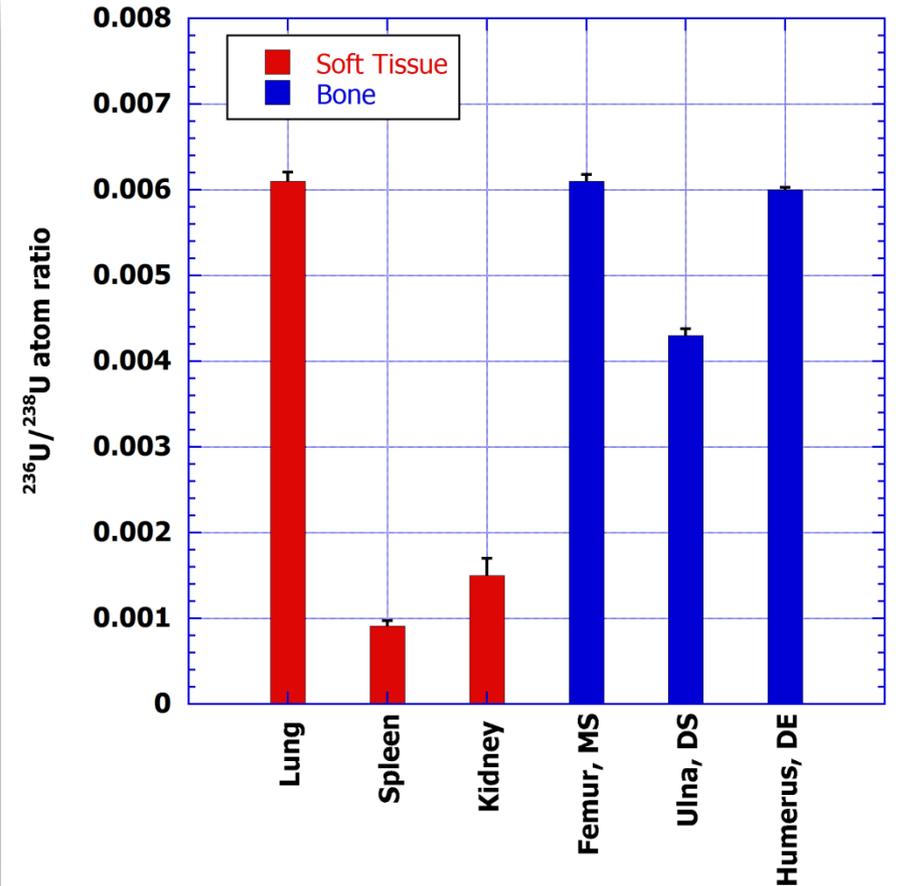
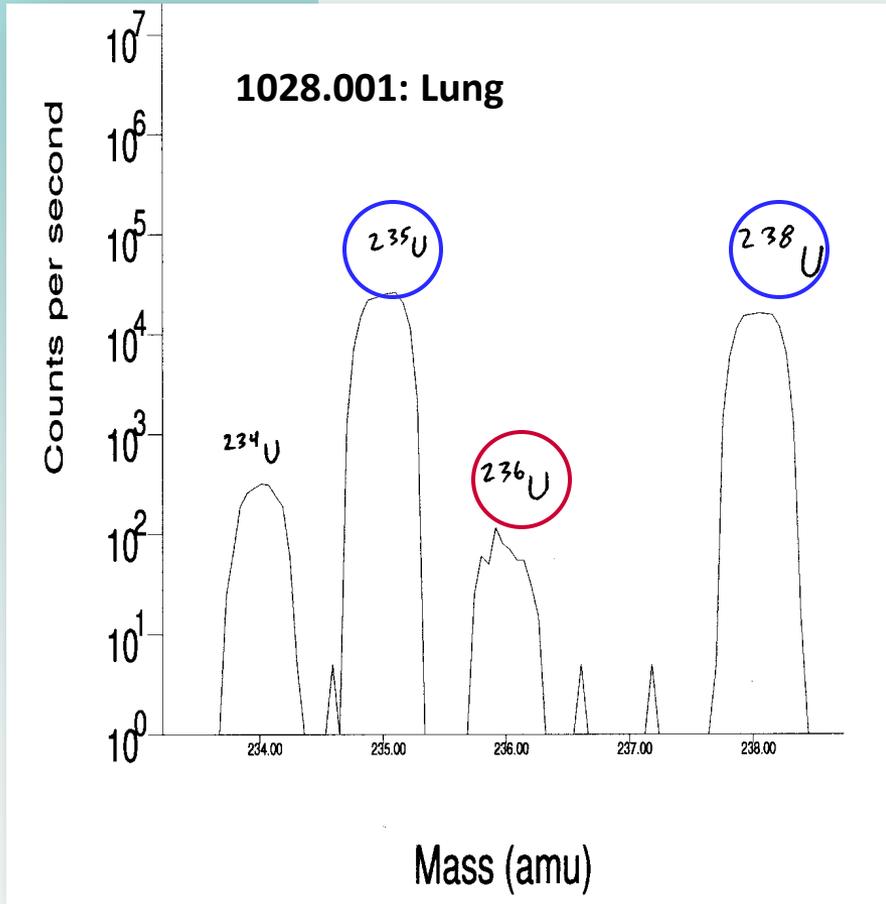
Uranium: SF-ICP-MS vs. KPA & AS



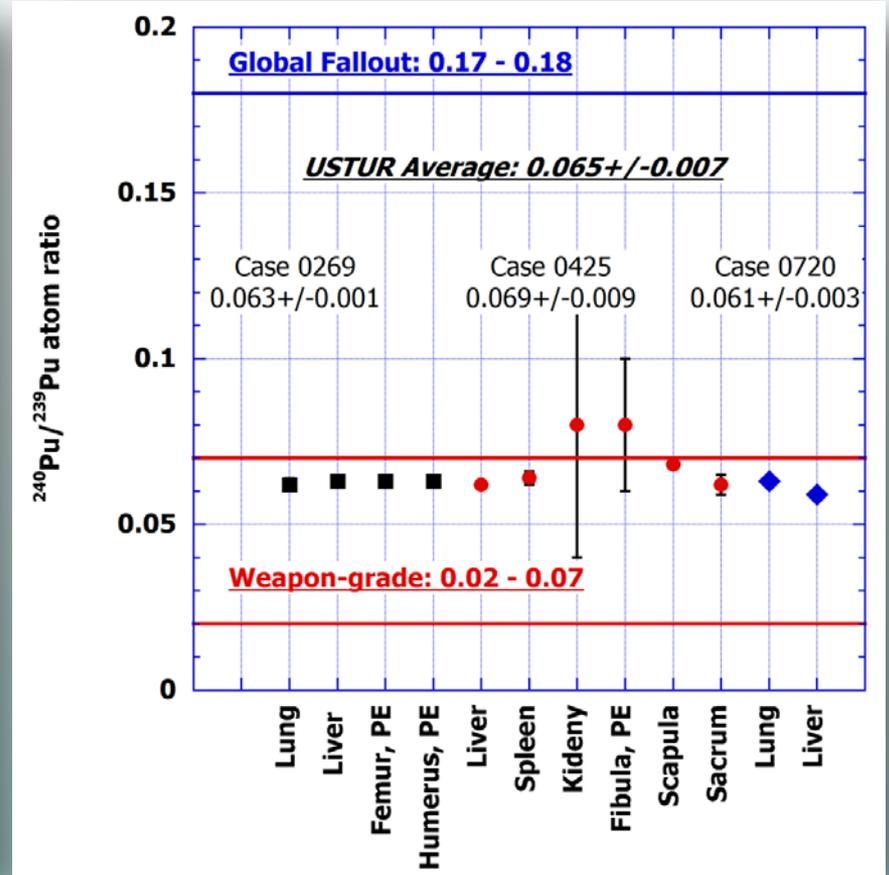
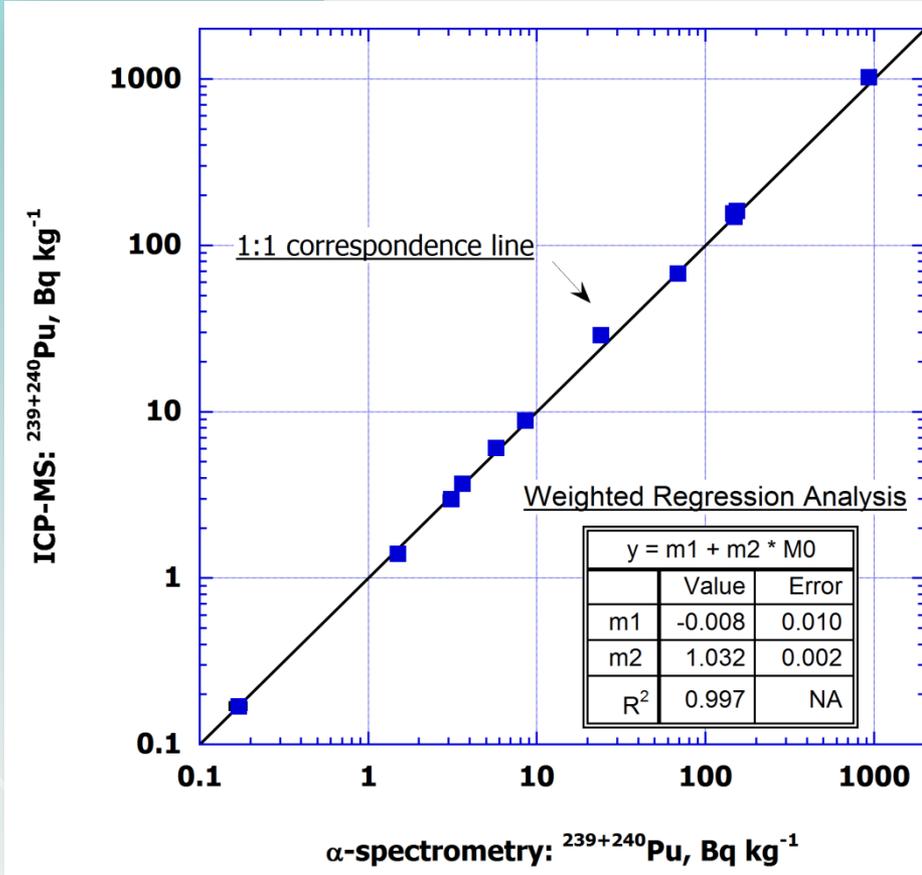
SF-ICP-MS: Anthropogenic ^{236}U



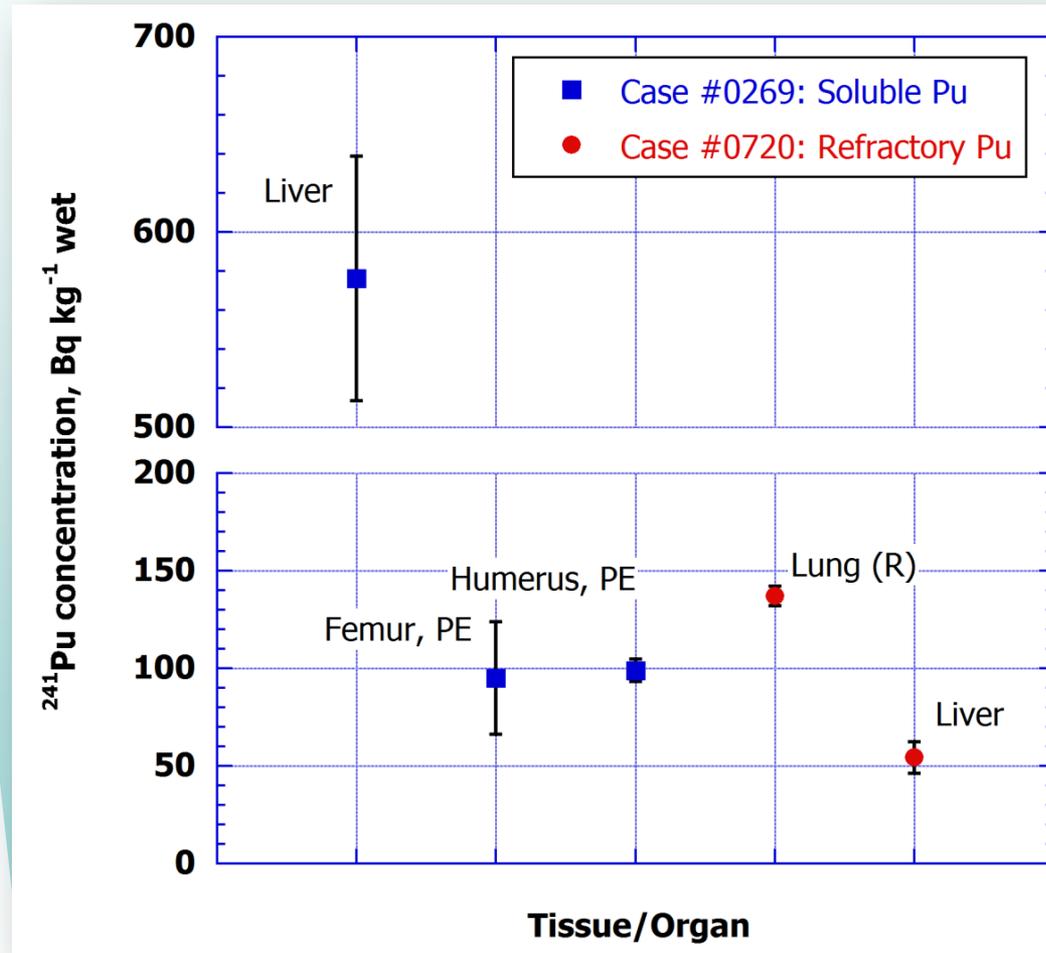
SF-ICP-MS: Anthropogenic ^{236}U



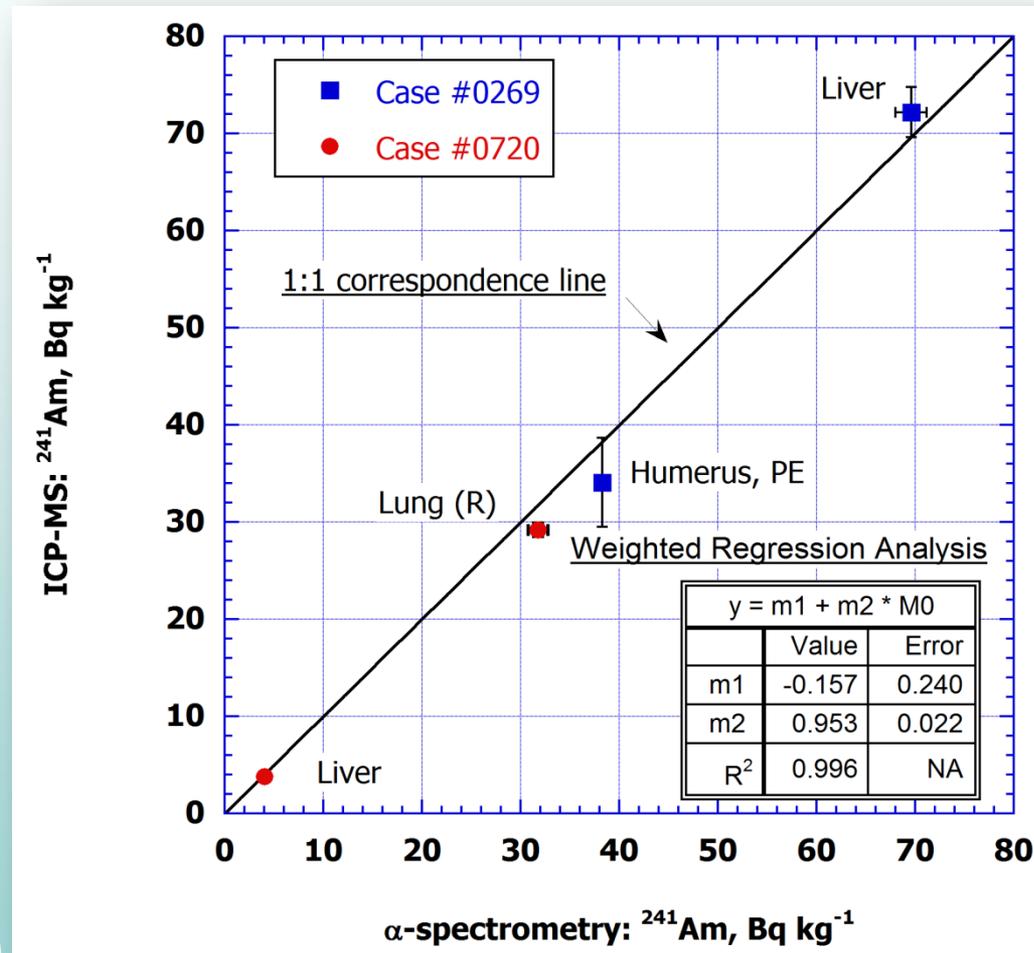
Plutonium: SF-ICP-MS vs. AS



SF-ICP-MS: ^{241}Pu (β -emitter)



Americium: SF-ICP-MS vs. AS



Laser-Ablation ICP-MS for Elemental Bio-Imaging of Actinides

*Collaboration with University of Technology
Sydney, Australia*



Dr. Philip Doble

**Director, Elemental Bio-imaging Facility
Department of Chemistry and Forensic
Science, University of Technology Sydney,
Australia**



Elemental Bio-Imaging (EBI)

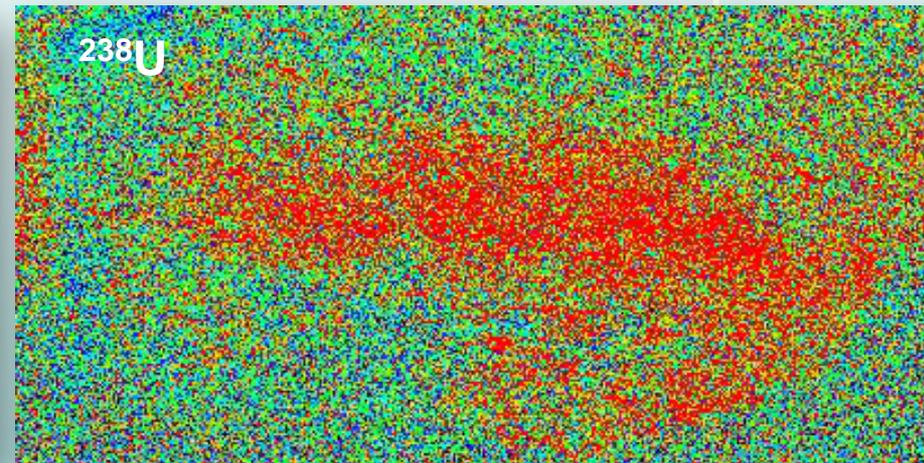
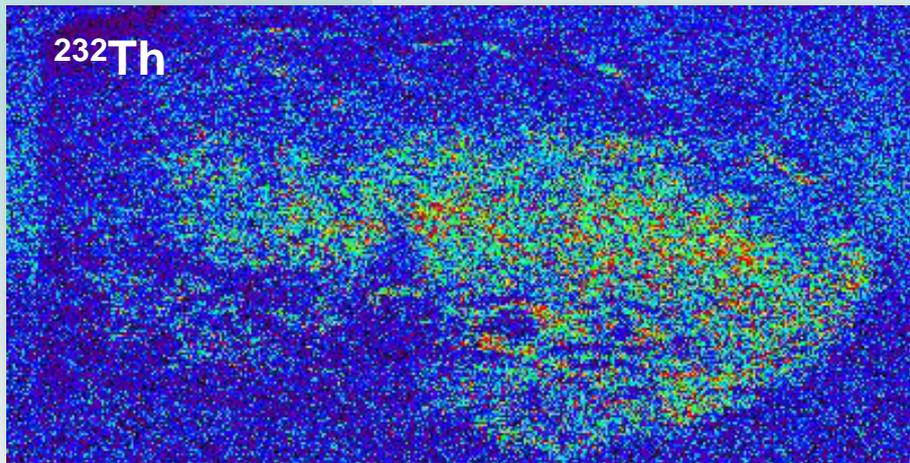
- **Imaging mass spectrometry using LA-ICP-MS**
- **Quantitative, isotope-specific determination and visualization of the spatial distribution of trace elements in thin tissue sections**
- **Technology transfer from analysis of geological samples**



Case 0846: Americium Exposure

- U.S. Radium Corp. – 1964-7 Chronic Inhalation – $^{241}\text{AmO}_2$
- Chelated – Intravenous Injection of Ca-DTPA
- Died at age 72 – Atherosclerotic Heart Disease

http://www.ustur.wsu.edu/Case_Studies/Narratives/0846_Narrative.php

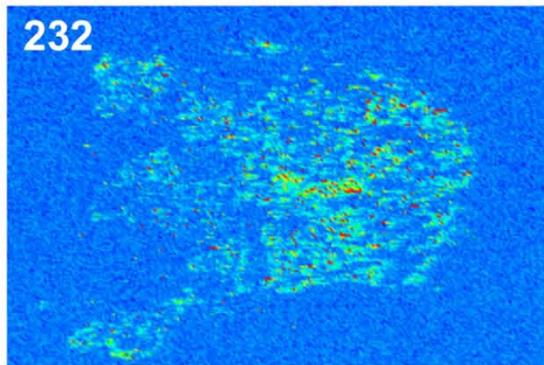


^{232}Th and ^{238}U concentration distributions in pulmonary lymph node from chronic environmental exposure

Case 1060: Uranium Exposure

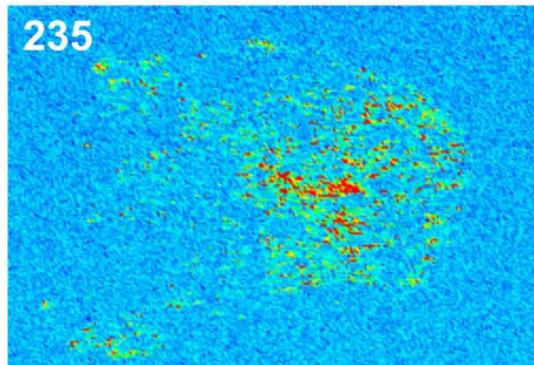
- Hanford – late 1940s Acute Inhalation – U_3O_8 (likely fume)
- Urinary excretion – **316 μg of U per day** (1st day after exposure)
- Died 2008 at age 83 – Cerebral Infarct

http://www.ustur.wsu.edu/Case_Studies/Narratives/1060_Narrative.php



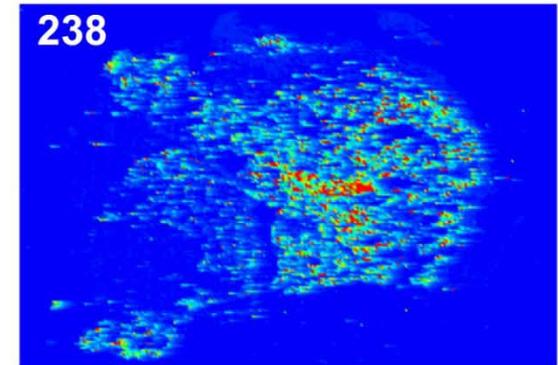
Concentration (ng/g)

0.000 53.3 107 160



Concentration (ng/g)

0.000 23.3 46.7 70.0



Concentration (ng/g)

0.000 2.83e+003 5.67e+003 8.50e+003

^{232}Th , ^{235}U and ^{238}U concentration distributions in parabrachial lymph node from occupational exposure

In Conclusion

- USTUR is a unique **scientific resource** for determining tissue retention and dosimetry for accidental occupational exposures to Pu, Am and U.
- ICP-MS provides significant new capabilities to the USTUR in the comprehensive analysis of actinide concentrations and isotopic compositions in human tissues.
- Elemental bio-imaging is a promising new avenue in USTUR research: capable of visualizing and quantifying actinide distributions in human tissues at both occupational and environmental levels of exposure.
- **Tissue samples (and supporting exposure data) are available for collaborating scientists to investigate the behavior of actinides and/or other elements in the human body.**



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Thank you for your attention!

