



U.S. Transuranium and Uranium Registries - A brief overview-

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***“Learning from Plutonium
and Uranium Workers”***

This Presentation

- History/Mission of the USTUR
- Tissue Analyses
- Terminal Dose Rates
- Elemental Bio-Imaging



National Plutonium Registry – The USAEC Vision



Pre-USTUR: History

- 1949: Hanford Autopsy Study
- 1968: founded National Plutonium Registry, at the Hanford Occupational Health Foundation
- 1970: name changed to the U.S. Transuranium Registry (USTR)
- 1978: founded the U.S. Uranium Registry (USUR)
- 1979: Stuart Gunn – first whole body donation
- 1983: Harold McCluskey ('Atomic Man') – dedicated issue of Health Physics Journal (October)



USTUR: History

- **1992: USTR and USUR merge into USTUR** - a 3-y DOE grant to Washington State University for the management and operation of the Registries
- 1992: USTUR expansion - creation of National Human Radiobiological Tissue Repository (NHRTR)
- 1996: the U.S. National Radiobiology Archives (NRA) transferred from Pacific Northwest National Laboratory (PNNL) to USTUR
- 2009: Laboratory Facilities moved to Richland, WA



USTUR: 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).

<http://www.ustur.wsu.edu/Mission/index.html>



USTUR: Registrants

...to date, **329 former workers** from various U.S. weapons or other sites with *known history of exposure* to actinides **voluntarily donated** their tissues (291) or entire bodies (38) for **scientific research.**

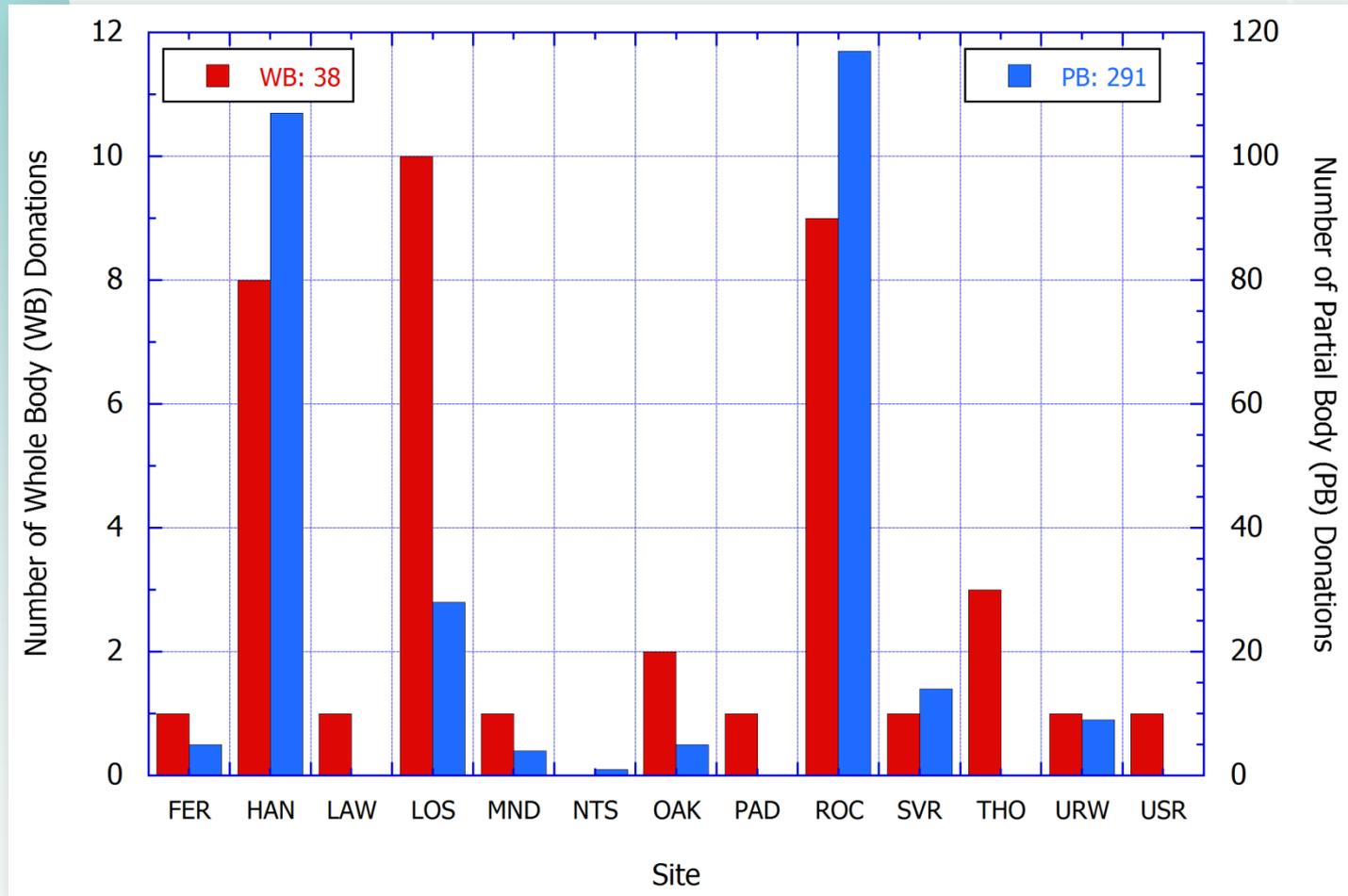


USTUR Registrant Statistics

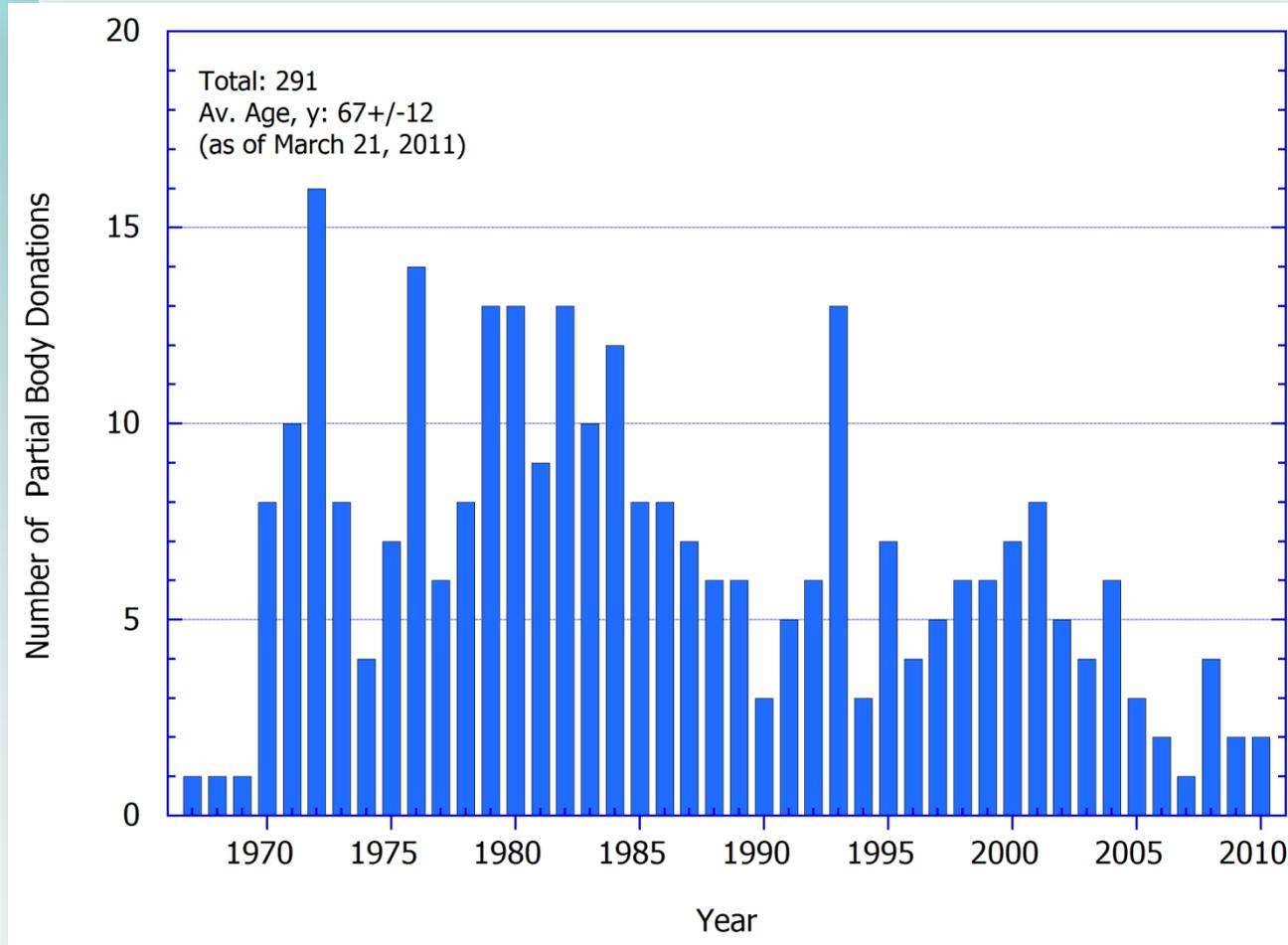
Total Active (Living) and Deceased Registrants[†] :	419
Living Registrants:	85
Potential Partial-body Donors:	65
Potential Whole-body Donors:	13
Special Studies:	7
Deceased Registrants:	334
Partial-body Donations:	291
Whole-body Donations:	38
Special Studies:	5
Inactive Registrants:	455
Total Number of Registrants:	874



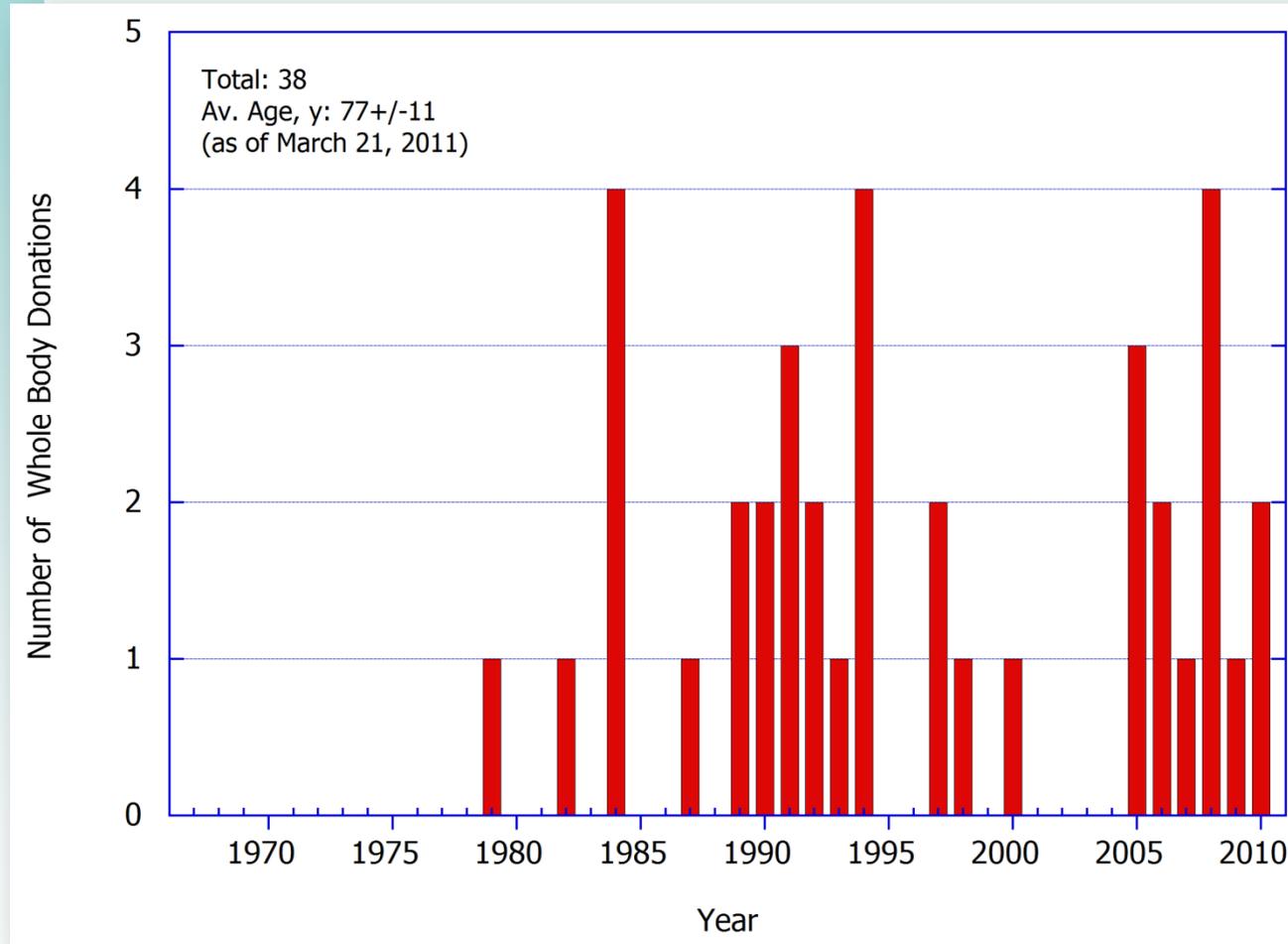
USTUR: Donations by Site



USTUR: Partial Body Donations



USTUR: Whole Body Donations



- Human Tissue Analyses -



Actinide Radionuclides

- $^{239+240}\text{Pu}$, ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu
- ^{241}Am
- ^{234}U , ^{235}U and ^{238}U , ^{236}U
- ^{232}Th



Analytical Measurement Techniques

- Alpha Spectrometry (AS): $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am , ^{234}U , ^{235}U , ^{238}U , ^{232}Th
- Autoradiography (AR): total α -Pu
- Kinetic Phosphorescence Analysis (KPA): total U
- Neutron Activation Analysis (NAA): ^{228}Th , ^{232}Th
- Inductively Coupled Plasma Mass Spectrometry (ICP-MS): ^{239}Pu , ^{240}Pu , $^{240}\text{Pu}/^{239}\text{Pu}$, (^{241}Pu , ^{241}Am), ^{234}U , ^{235}U , ^{238}U , ^{236}U



USTUR: Human Tissue Analysis

- 1) Tissue sample dry/wet ashing / dissolution
 - 2) Actinide separation from selected dissolved tissue sample aliquot
 - 3) Counting source preparation: electrodeposition
 - 4) Counting: α -spectrometry
- Inductively Coupled Plasma Mass Spectrometry

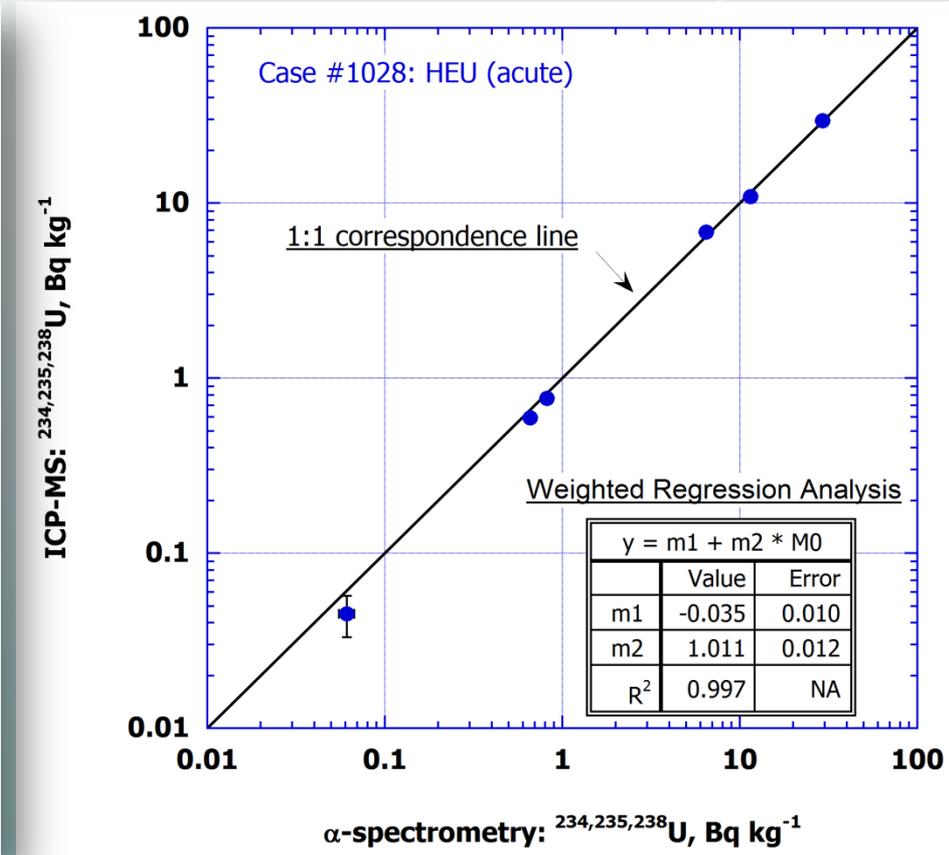
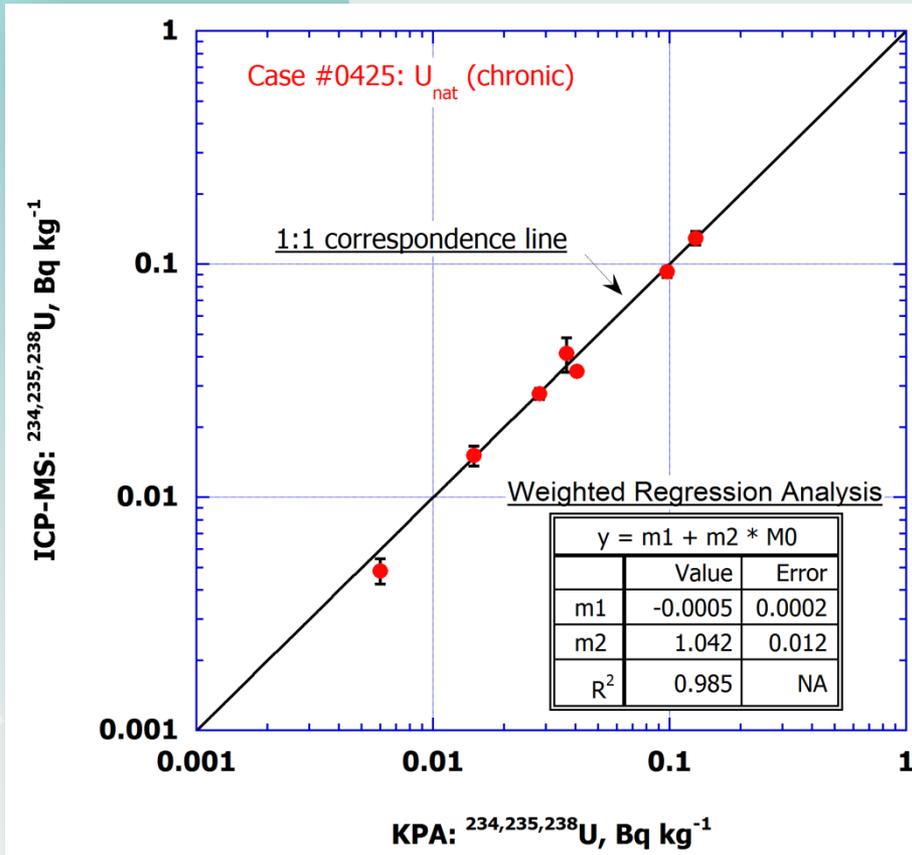


ICP-MS Benefits

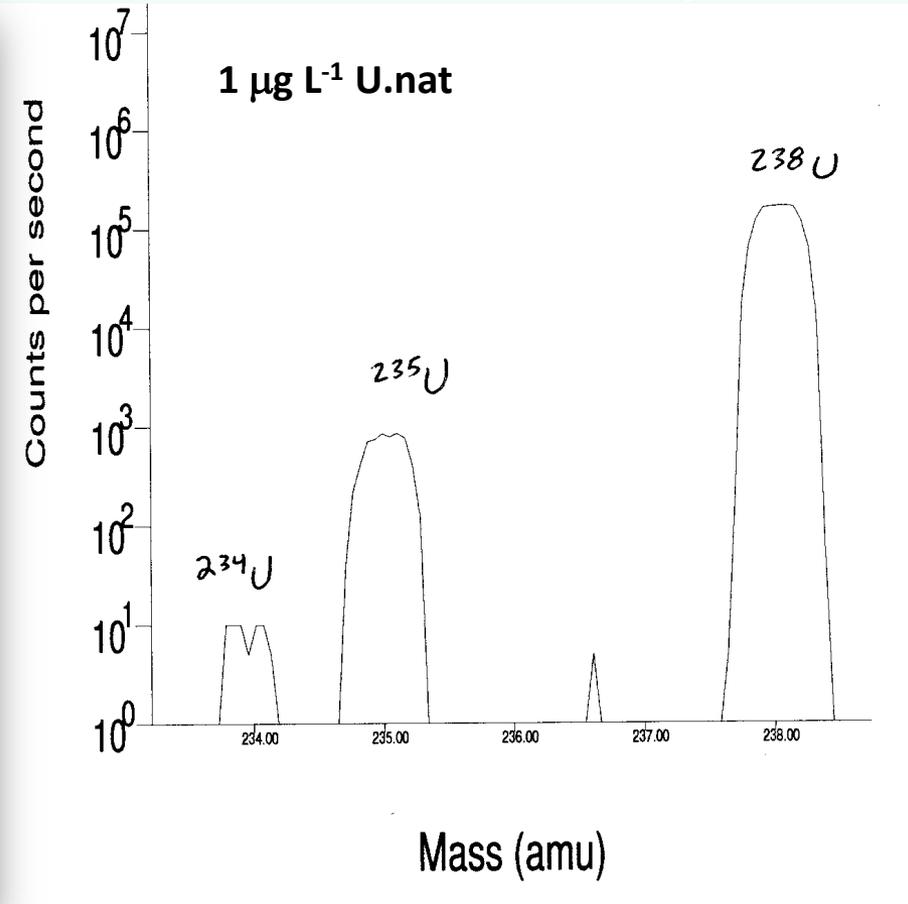
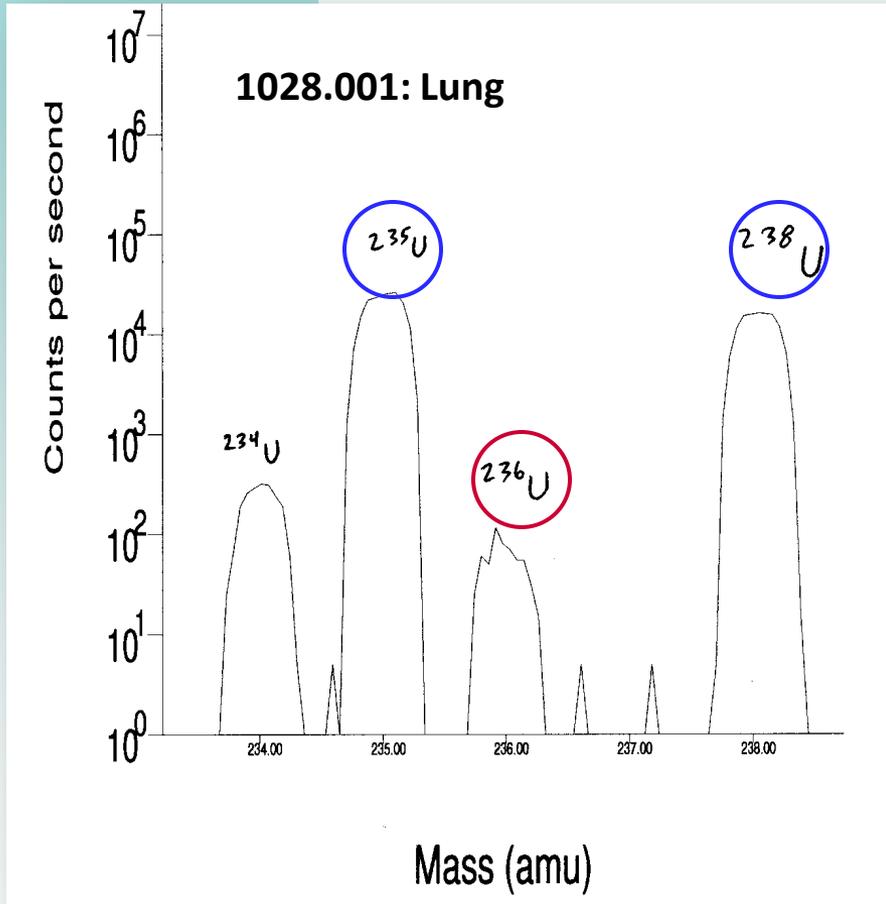
- **Sufficient sensitivity** to determine actinides at fg-levels (10^{-15} g)
- **Rapid, low cost** analysis
- **Isotopic (atom ratio) information** for source identification and confirmation
- **Accurate** determinations by isotope dilution method ($^{233,236}\text{U}$, $^{242,244}\text{Pu}$, ^{243}Am)



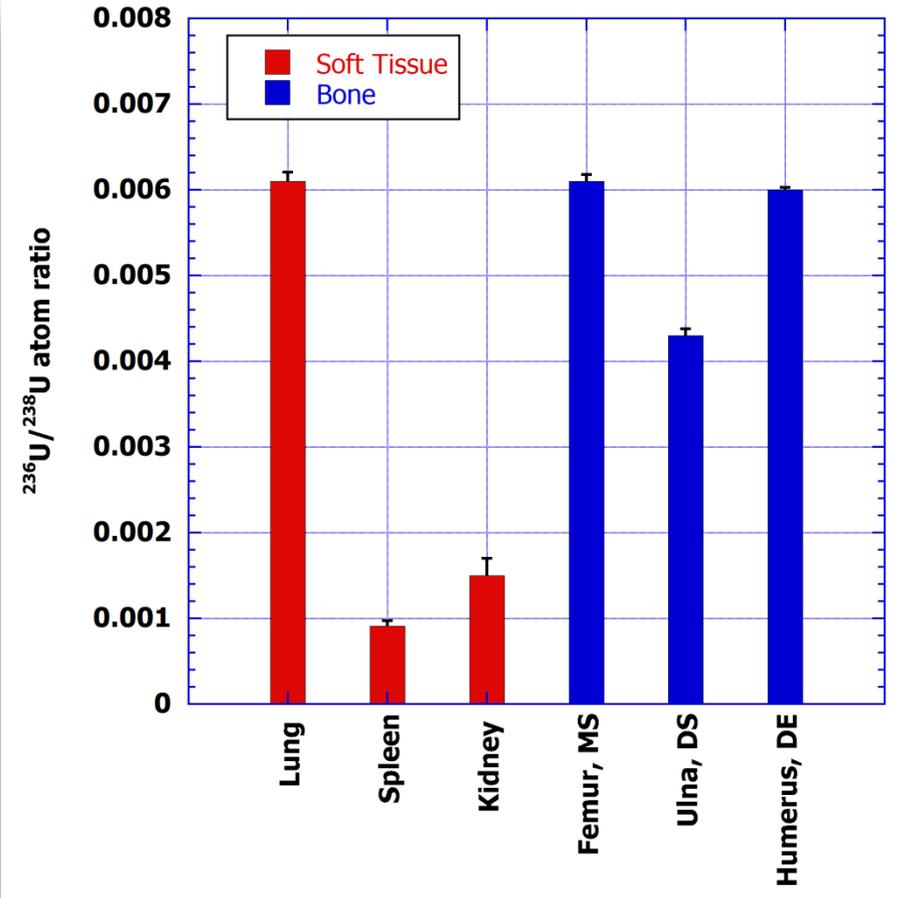
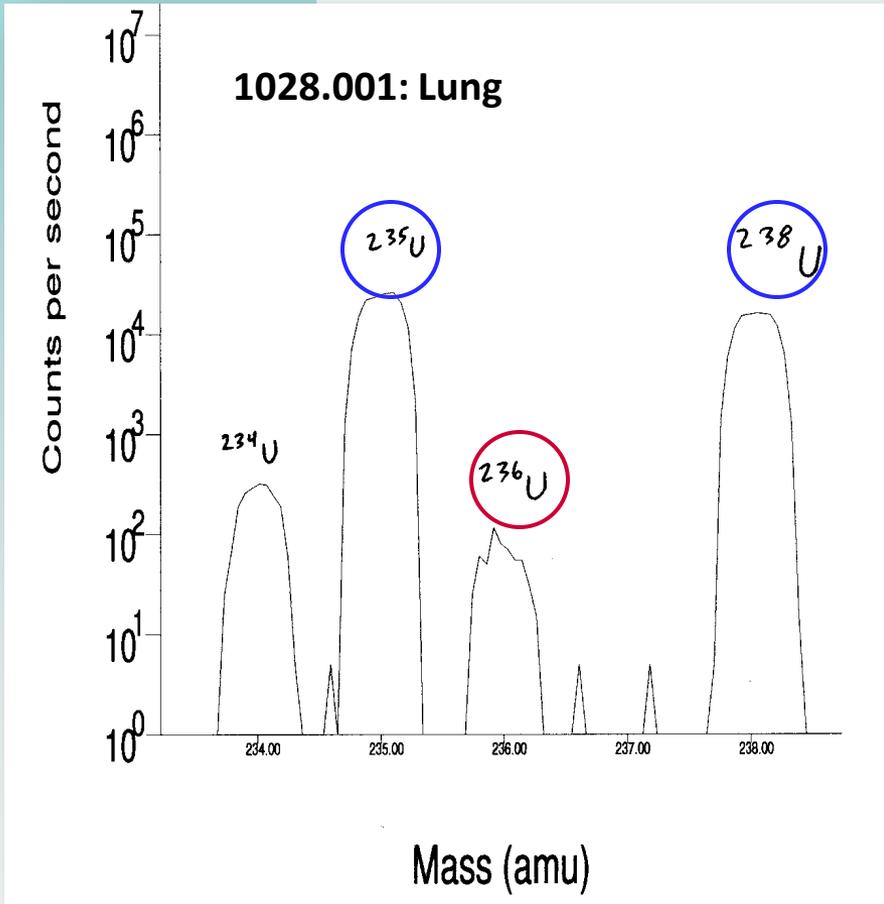
Uranium: SF-ICP-MS vs. KPA & AS



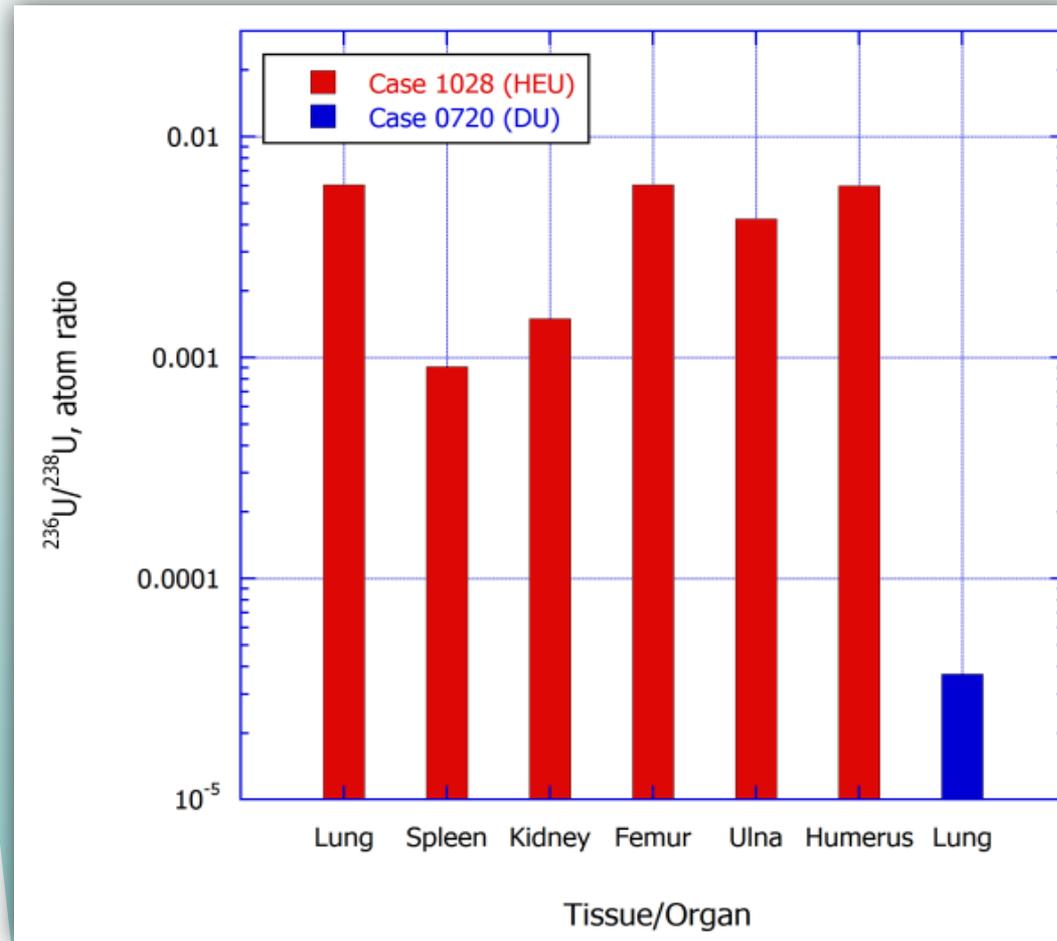
SF-ICP-MS: Anthropogenic ^{236}U



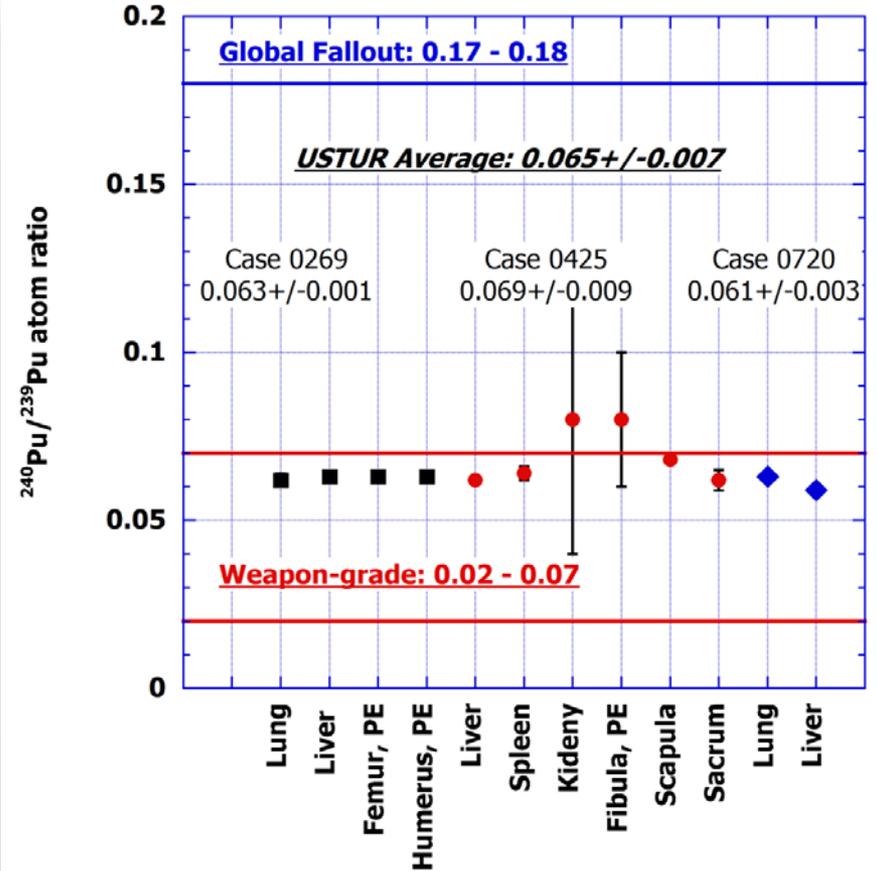
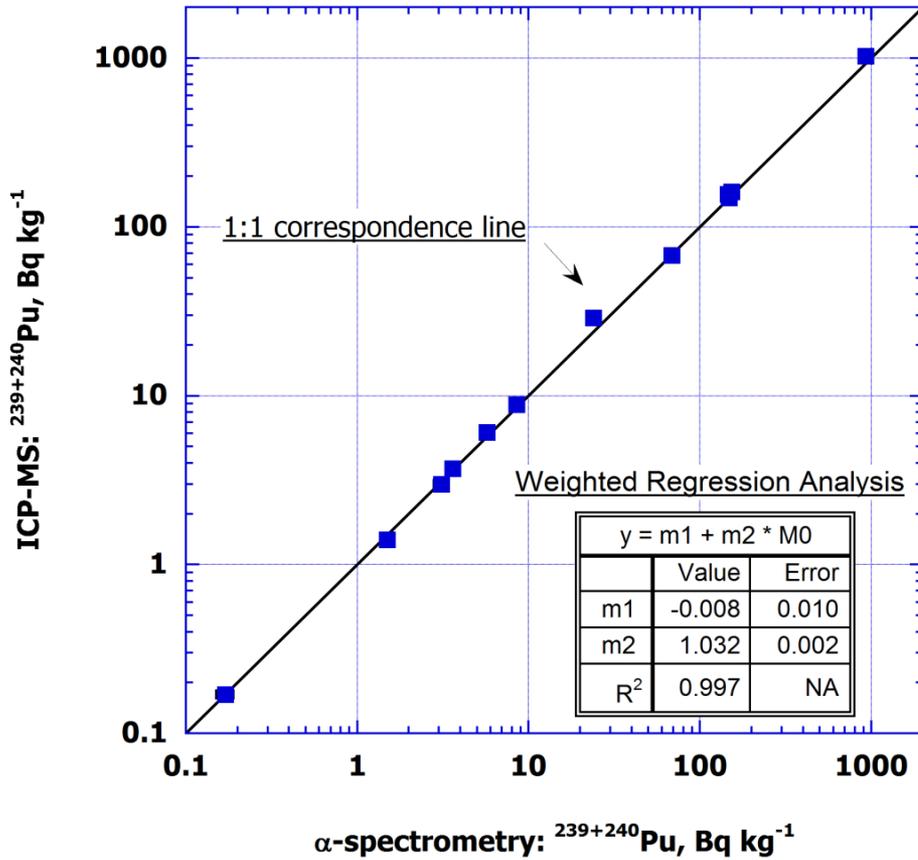
SF-ICP-MS: Anthropogenic ^{236}U



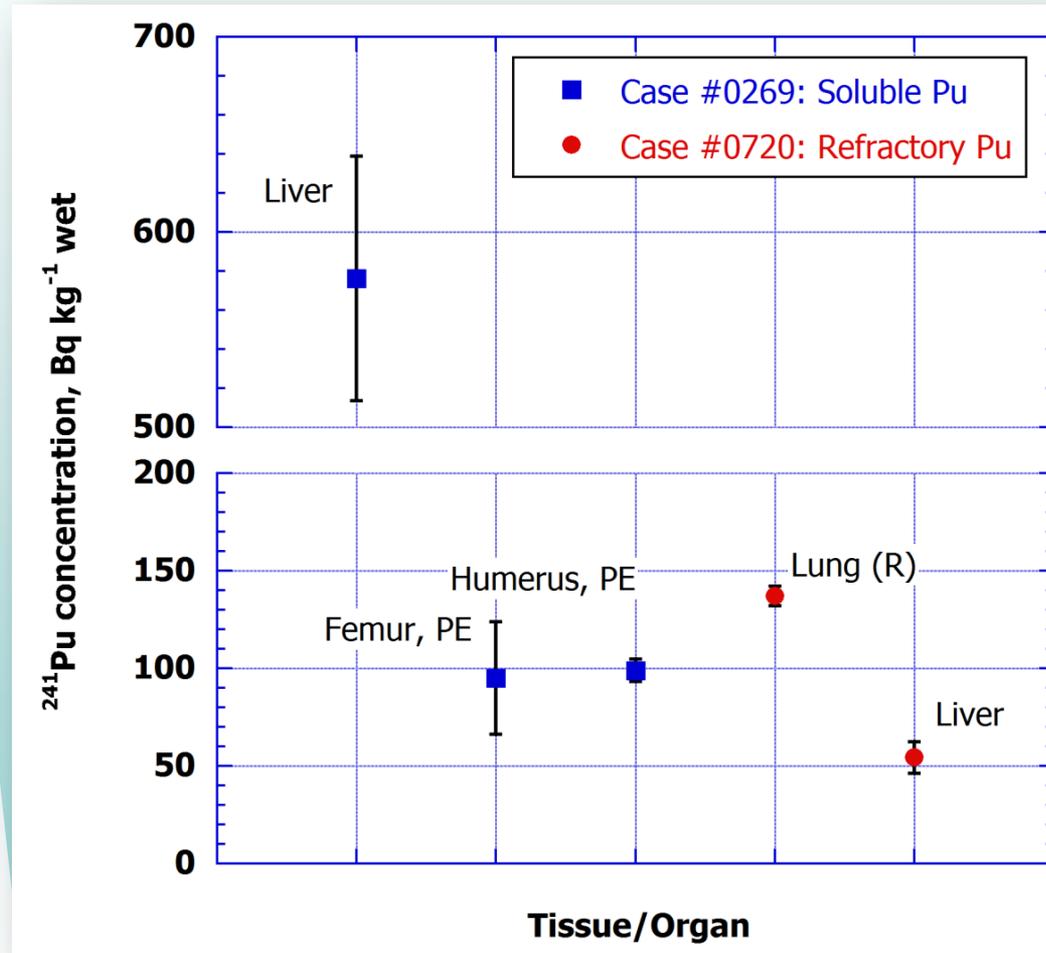
$^{236}\text{U}/^{238}\text{U}$ atom ratio signature of U exposure – HEU vs. DU



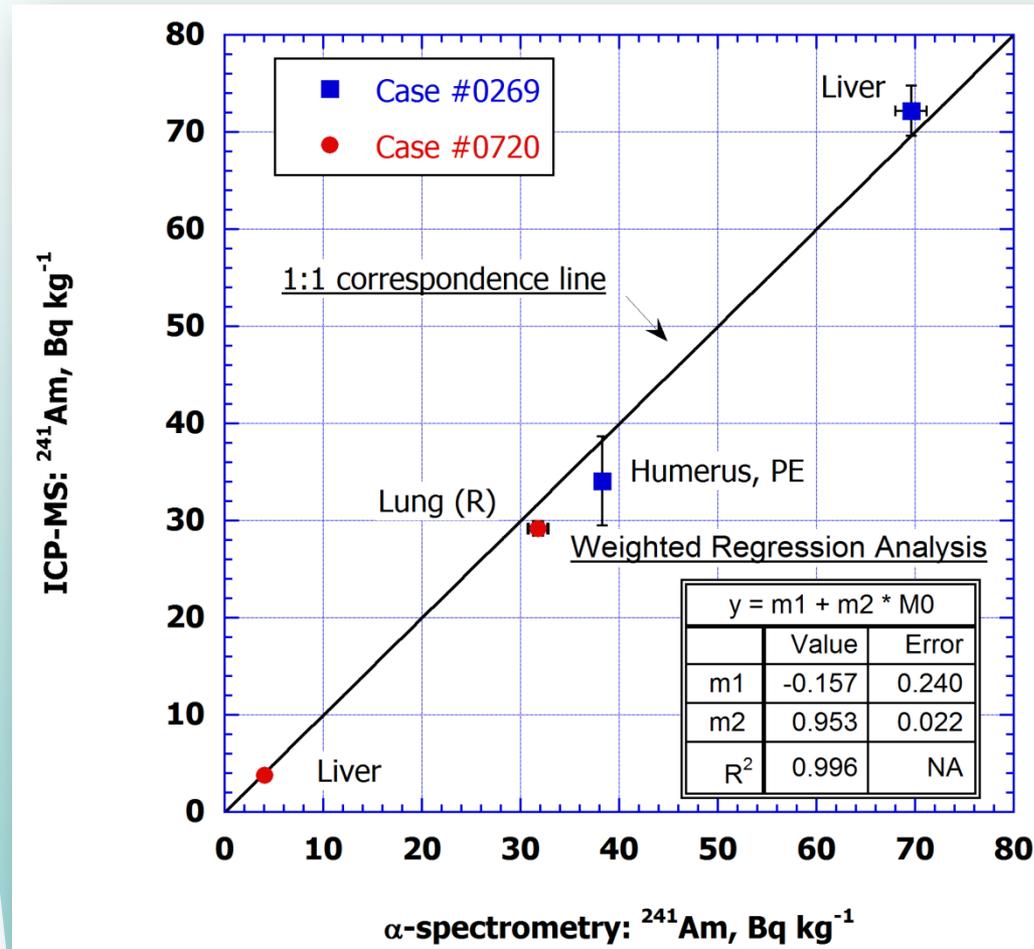
Plutonium: SF-ICP-MS vs. AS



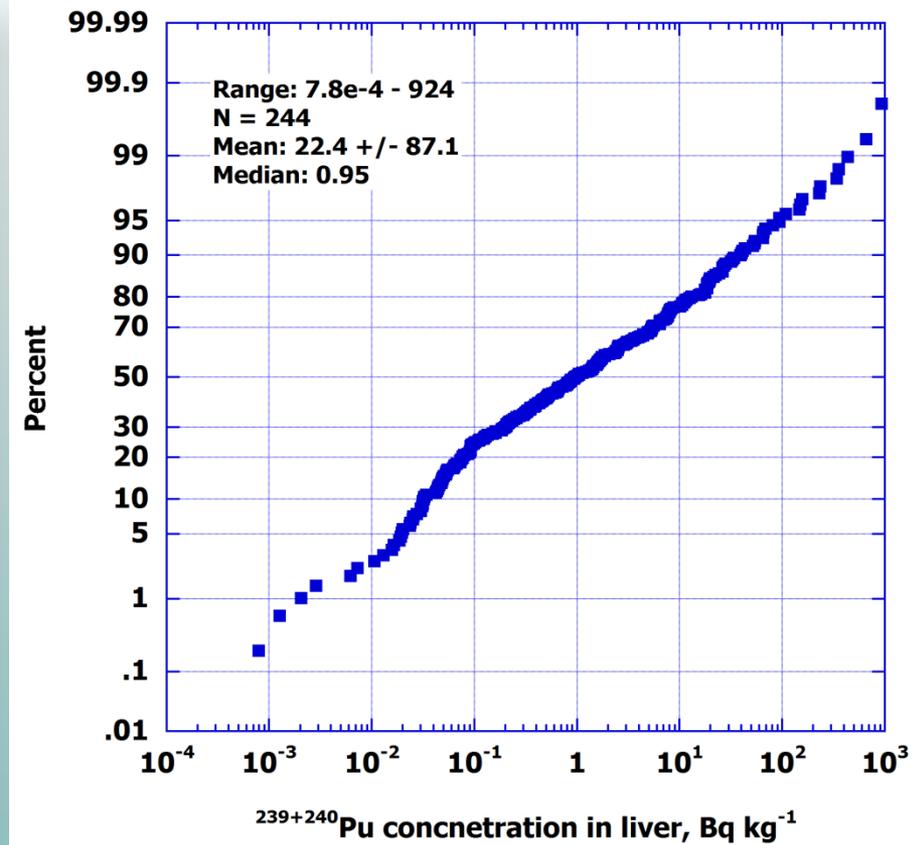
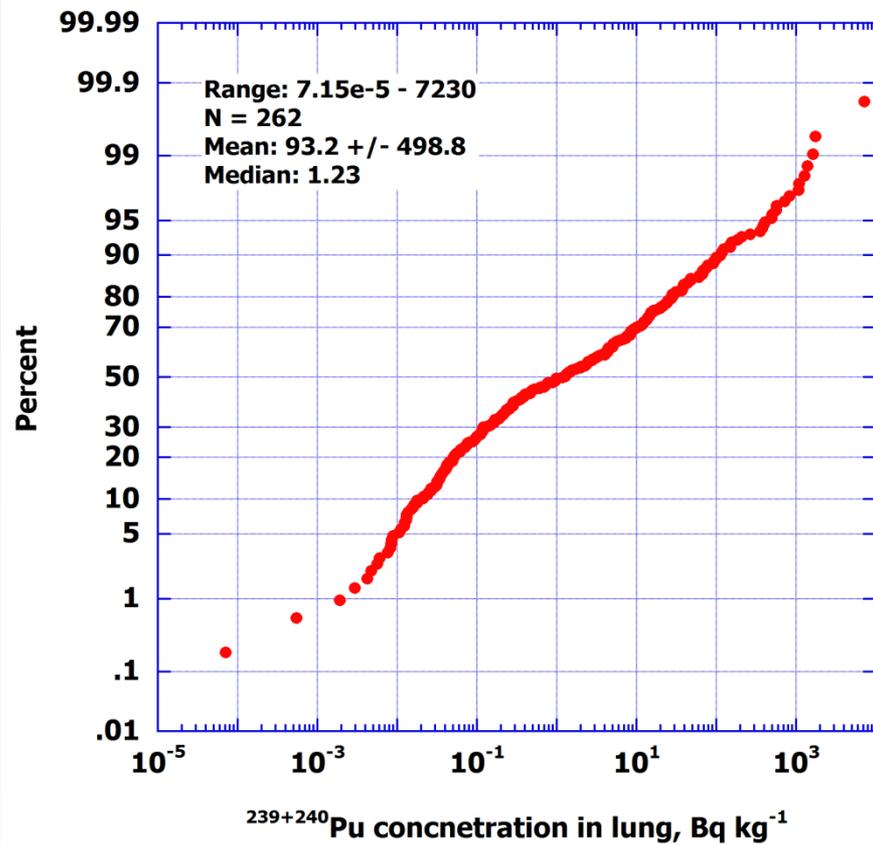
SF-ICP-MS: ^{241}Pu (β -emitter)



Americium: SF-ICP-MS vs. AS



$^{239+240}\text{Pu}$ Concentration in Lung and Liver



$^{239+240}\text{Pu}$: USTUR vs. General Public

Tissue	$^{239+240}\text{Pu}$ concentration, Bq kg ⁻¹ of wet tissue				Activity ratio, USTUR:GP	
	e.g. Case 0209		General public (GP)		USTUR:GP	
	Value	±SD	Average†	±SD	Value	±SD
<u>Soft Tissues:</u>						
Lung	1.34E+00	3.00E-02	3.75E-03	3.01E-03	356	287
Lymph Node	7.37E+01	2.23E+00	3.11E-03	1.36E-03	23,729	10,412
Liver	1.59E+00	3.47E-02	1.91E-02	3.89E-03	83.3	17.1
Kidney	1.61E-02	1.83E-03	1.48E-03	5.23E-04	10.9	4.1
Spleen	9.45E-02	7.59E-03	2.96E-03		31.9	2.6
<u>Bones:</u>						
Vertebrae	2.04E-01	3.59E-02	6.26E-03	2.72E-03	32.7	15.3
Rib	3.29E-01	5.55E-02	2.70E-03		122.0	20.5
Sternum	2.17E-01	1.54E-02	2.96E-03		73.2	5.2

† - from 4 studies in Japan, USA, Germany and Kazakhstan

Distribution of Terminal Lung and Liver Dose Rates in USTUR Registrants

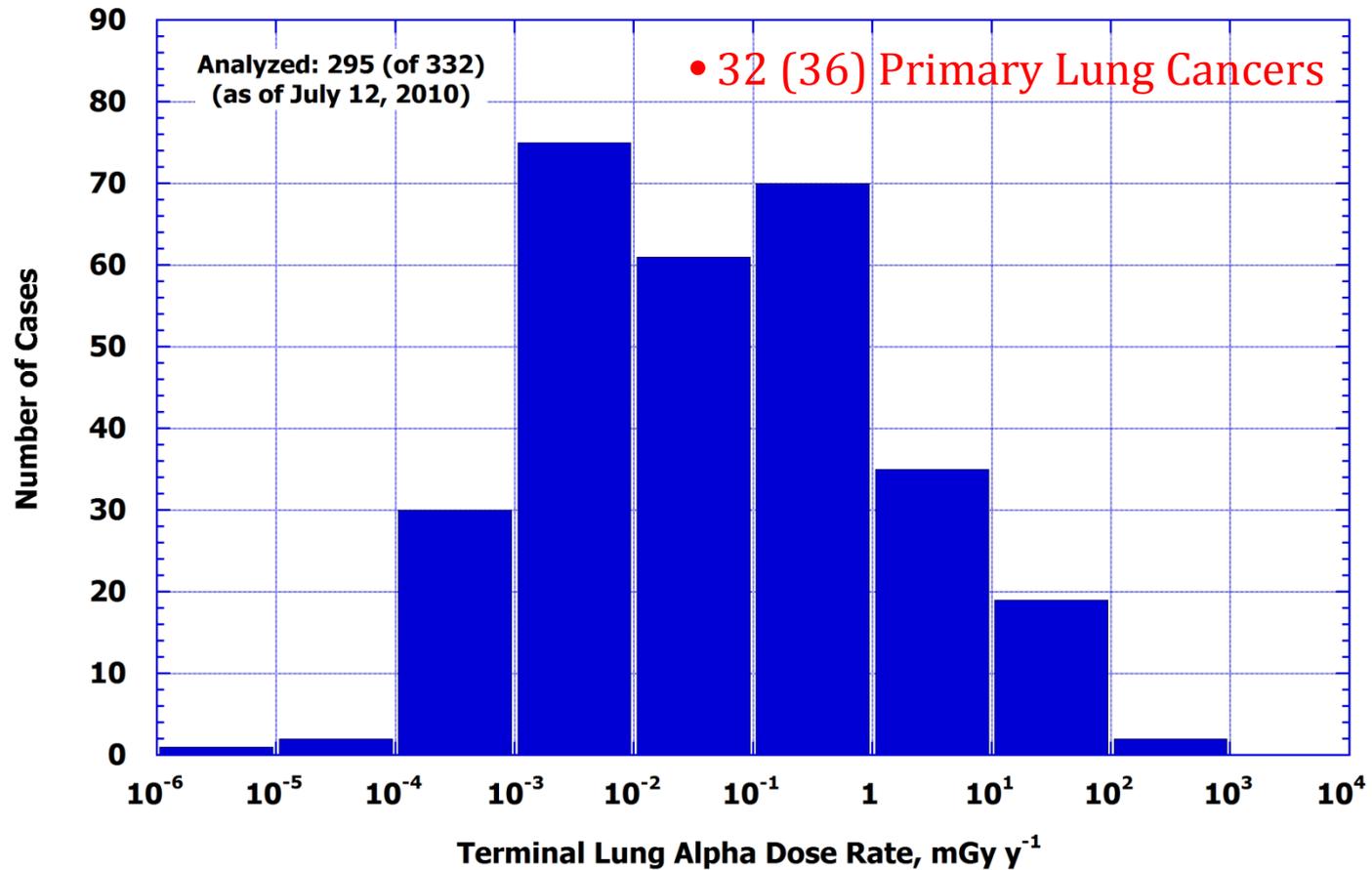
McCord, S.L., James, A.C., Tolmachev, S.Y.

To be presented at

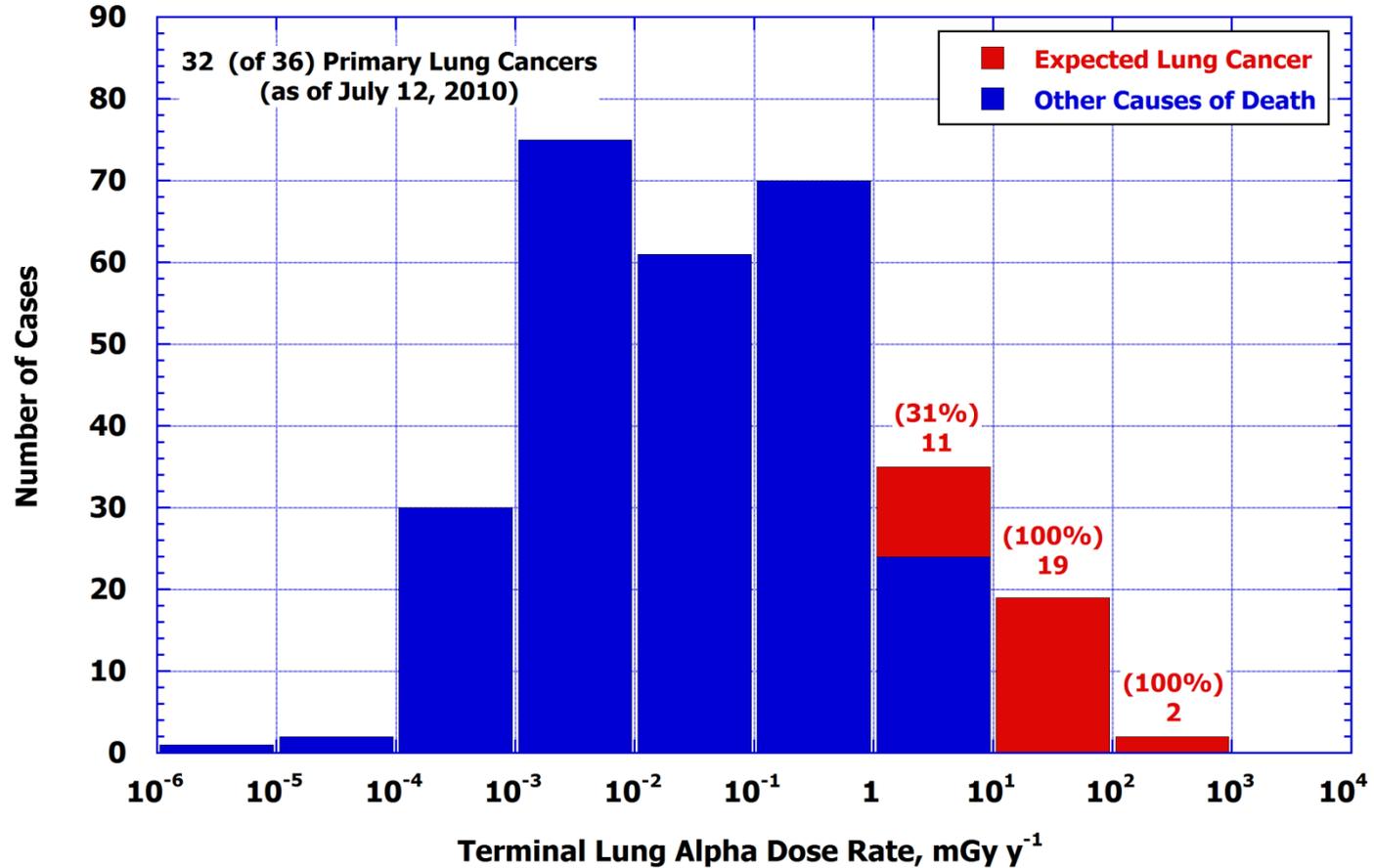
*Health Physics Society 56th Annual Meeting
26-30 June 2011, West Palm Beach, Florida*



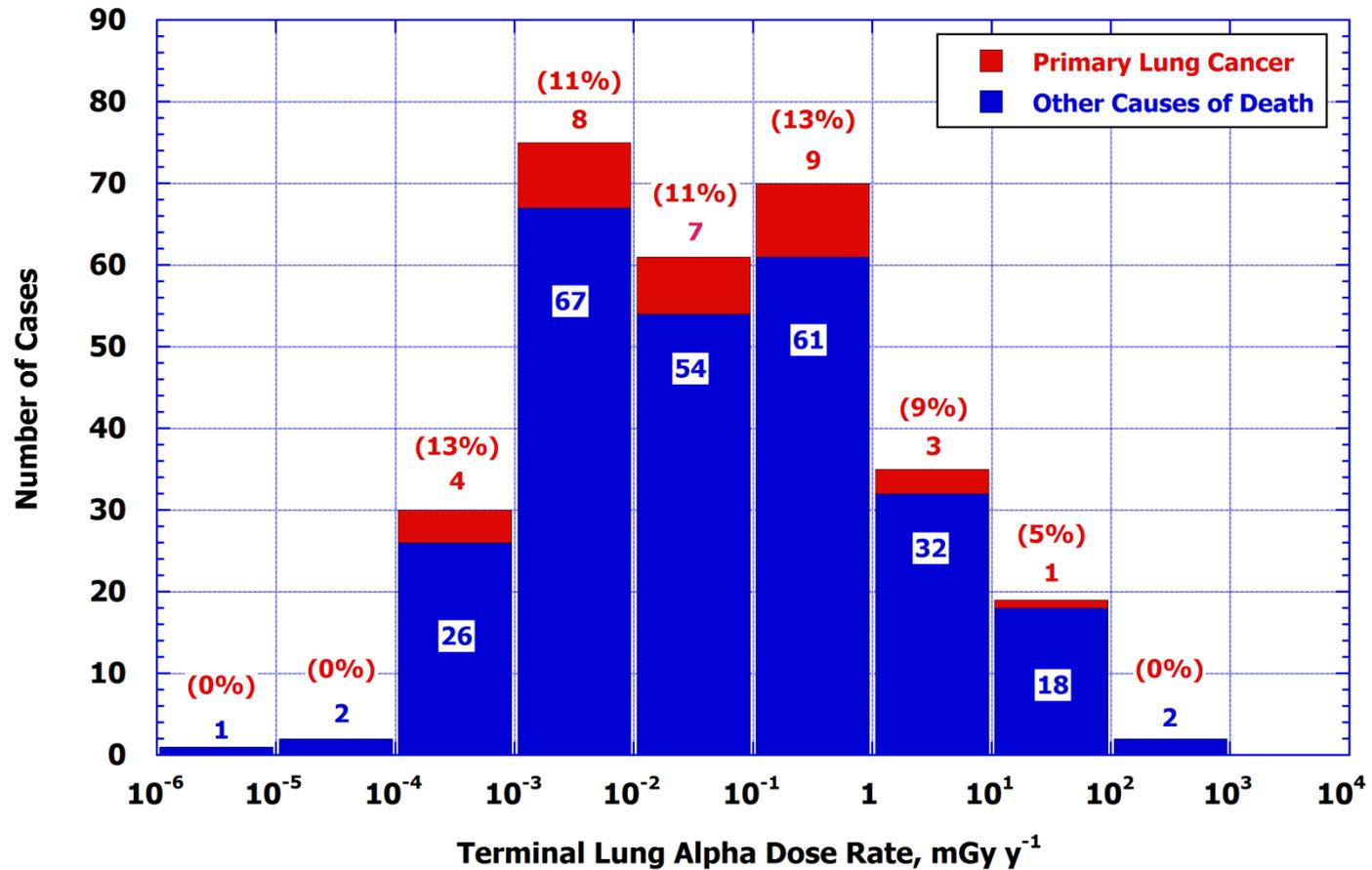
Terminal Lung Dose Rate



Lung Cancer Mortality: 'Speculations'



Lung Cancer Mortality: Reality



Message

- Other factors might (are) cause a cancer
- Do not blame radiation for everything!



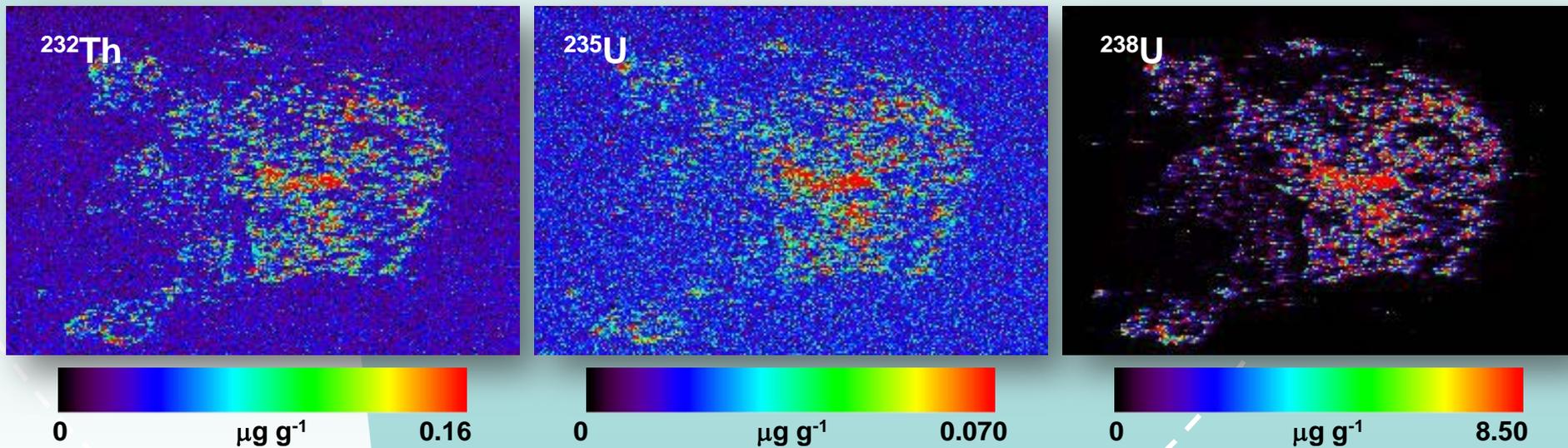
Elemental Bio-Imaging (EBI)

- Imaging mass spectrometry
- Quantitative, element(isotope)-specific determination and visualization of the spatial distribution of trace elements in thin tissue sections
- Technology transfer from analysis of geological samples



Case 1060: Uranium Exposure

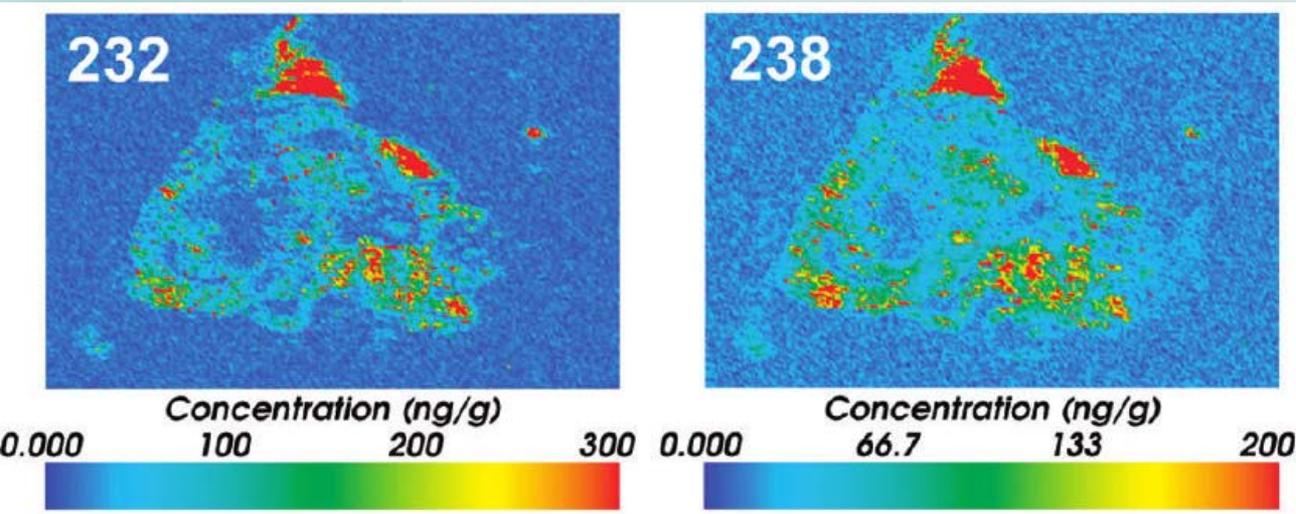
- Exposure – late 1940s Chronic/Acute Inhalation – U_3O_8 (likely fume)
- Urinary excretion – 316 μg of U per day
- Died 2008 at age 83 – Cerebral Infarct



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

Case 0407: Plutonium Exposure

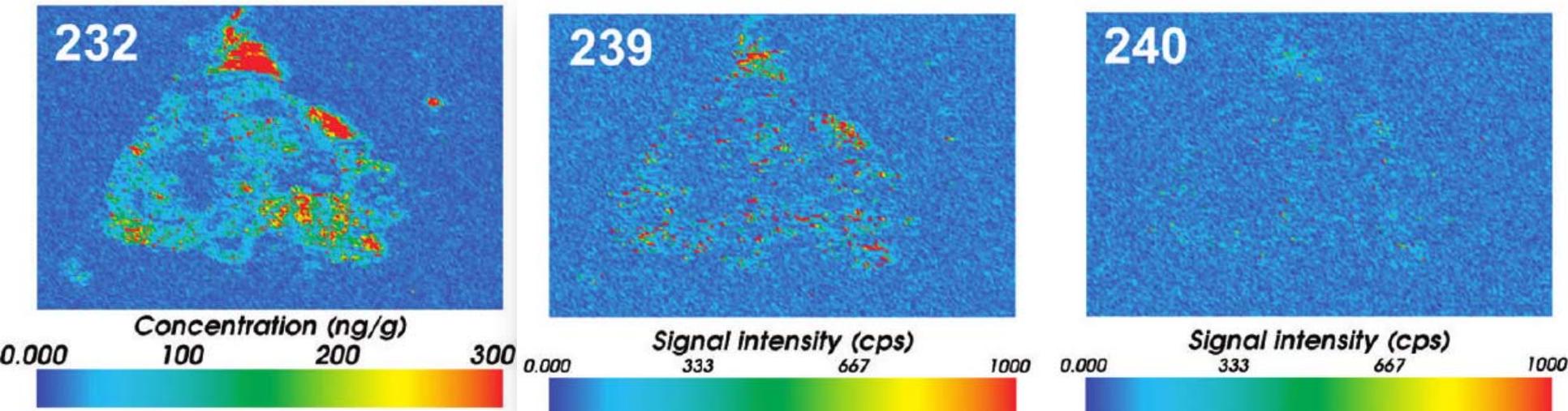
- Exposure – 1965 Acute (Fire) – ^{239}Pu (refractory)
- Chelated – Intravenous Injection of Ca-DTPA
- Died 2008 at age 90+ – Atherosclerotic Heart Disease



^{232}Th , ^{238}U , ^{239}Pu and ^{240}Pu distributions in paratracheal lymph node from chronic natural background (Th, U) and occupational exposures (Pu)

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^{232}Th , ^{238}U , ^{239}Pu and ^{240}Pu distributions in paratracheal lymph node from chronic natural background (Th, U) and occupational exposures (Pu)

Synchrotron Radiation micro X-ray fluorescence (SR- μ XRF) for Elemental Bio Imaging



USTUR – Ghent University

M.S. Project: Use of LA-ICP-MS for studying/mapping the distribution of actinides in human tissues



Ms. Eva Vergucht



Dr. Andrei Izmer



Prof. Dr. Frank Vanhaecke



Dr. Björn De Samber

Dr. Laszlo Vincze



LA-ICP-MS vs. SR- μ XRF

Analysis:

- **Destructive** vs. **non-destructive**

Determination:

- **isotopic** vs. **elemental** (*e.g.* ^{235}U , ^{238}U , ^{239}Pu , ^{240}Pu vs. U, Pu)

Achievable resolution:

- **5 μm** vs. **0.02 μm** (20 nm)

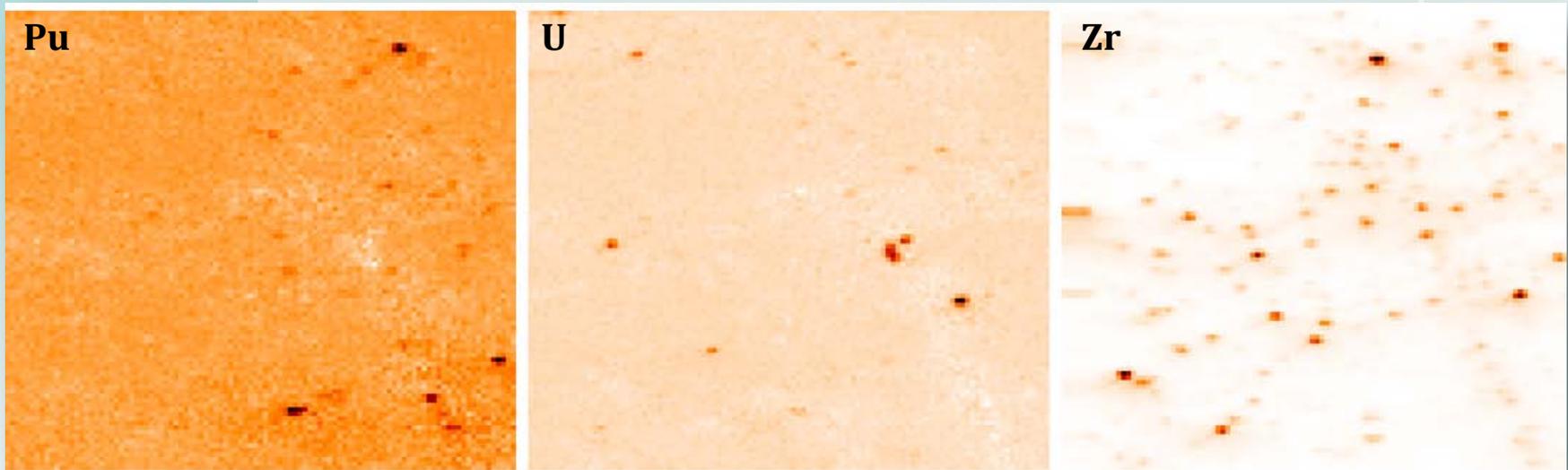
Installation:

- **any laboratory** vs. **synchrotron facility**



SR- μ XRF at DESY facility (Hamburg)

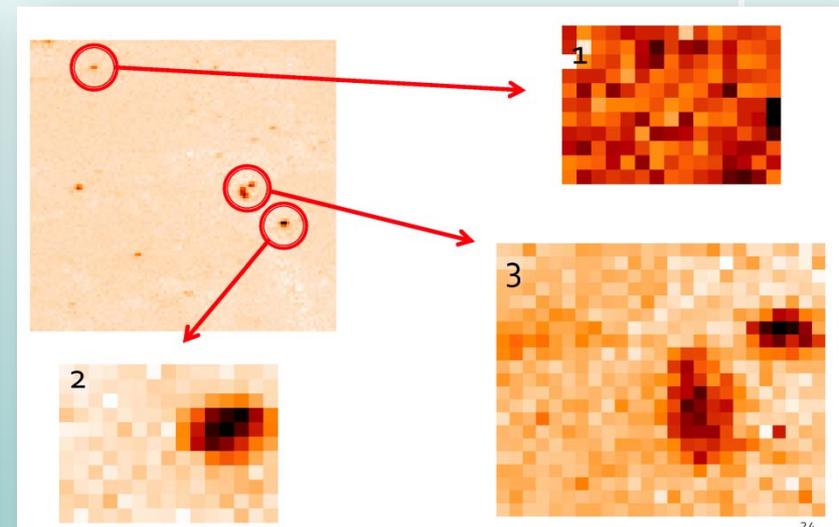
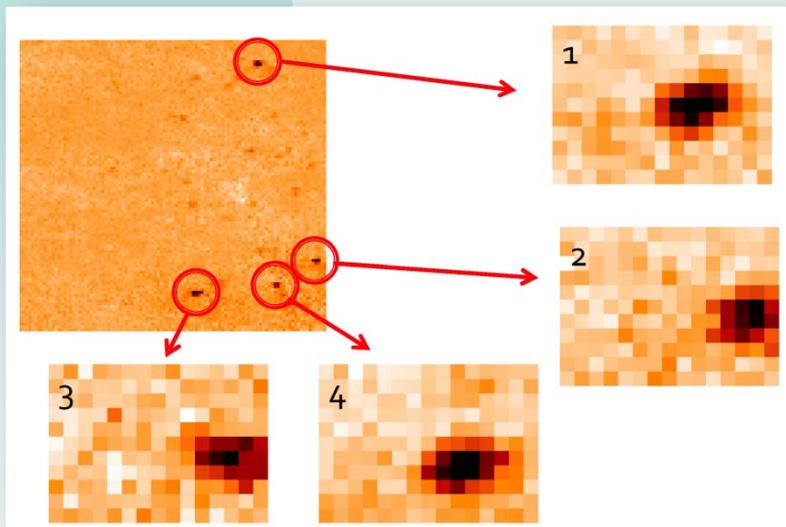
- Resolution: 18 μm



Elemental Pu, U and Zr distributions in paratracheal lymph node from USTUR Case 0407

SR- μ XRF: Work in Progress

- Application of nano-beam for particle characterization



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