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2015 Radiochemistry Progress Report

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*“Learning from Plutonium and
Uranium Workers”*



Outline

- Tissue Analysis Progress
(9/1/2014 – 8/1/2015)
- Equipment Upgrade and Facility Maintenance
- DQO/Quality Assurance Plan: US DOE Mixed Analyte Performance Evaluation Program (MAPEP)



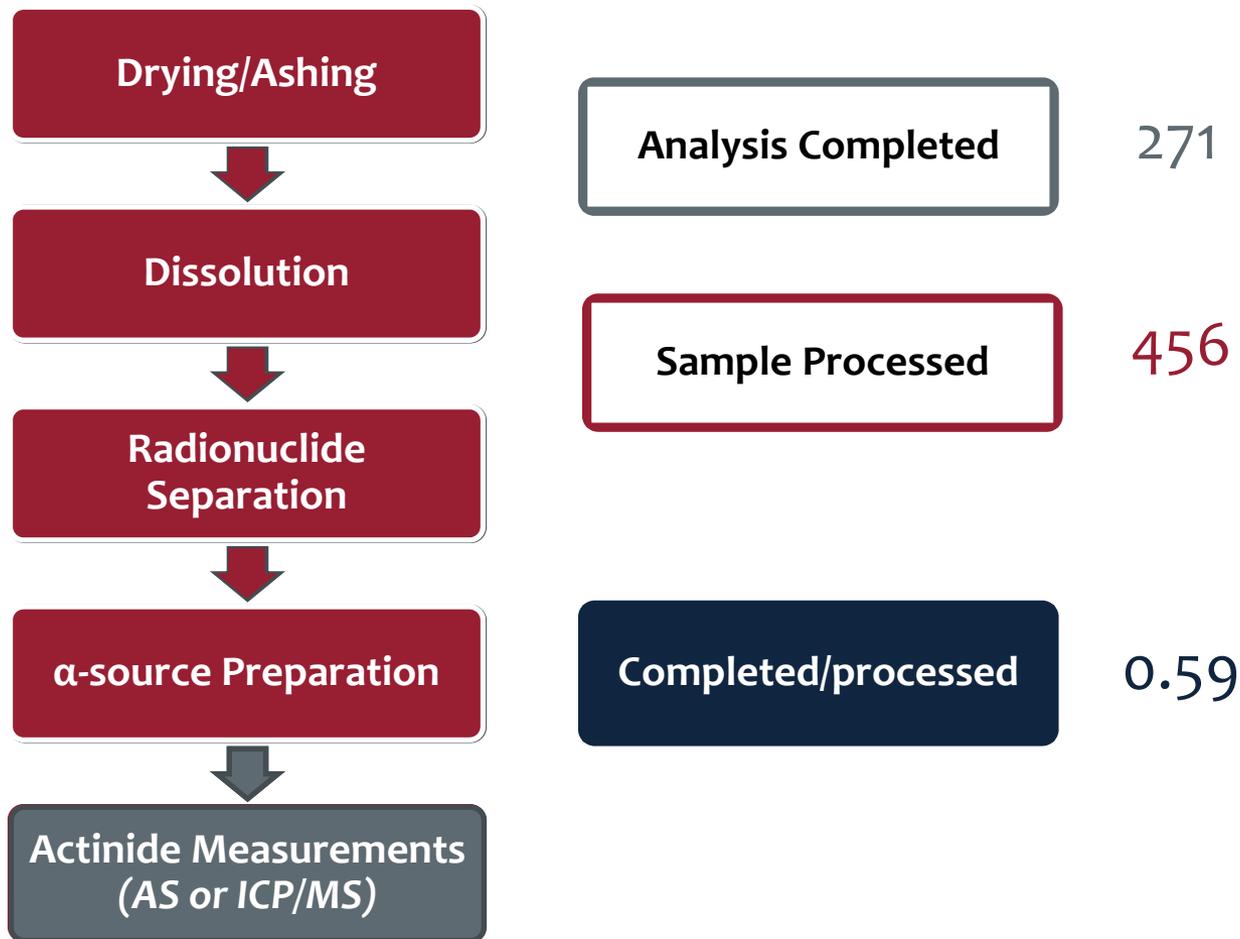


Tissue Analysis Progress





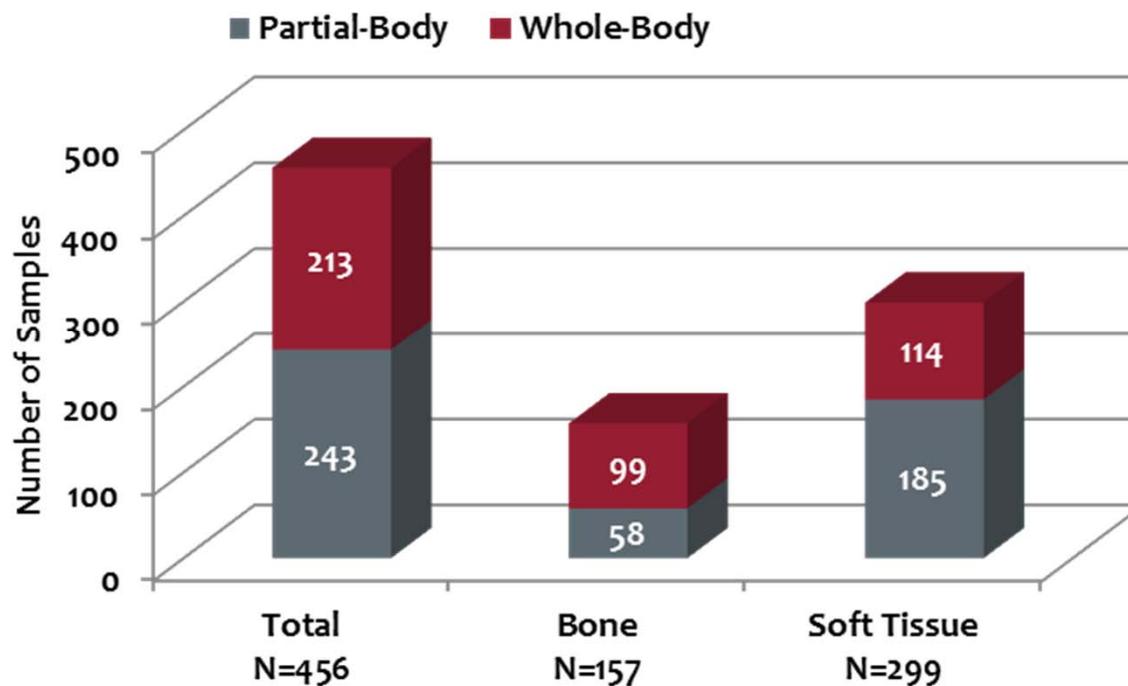
Tissue Analysis





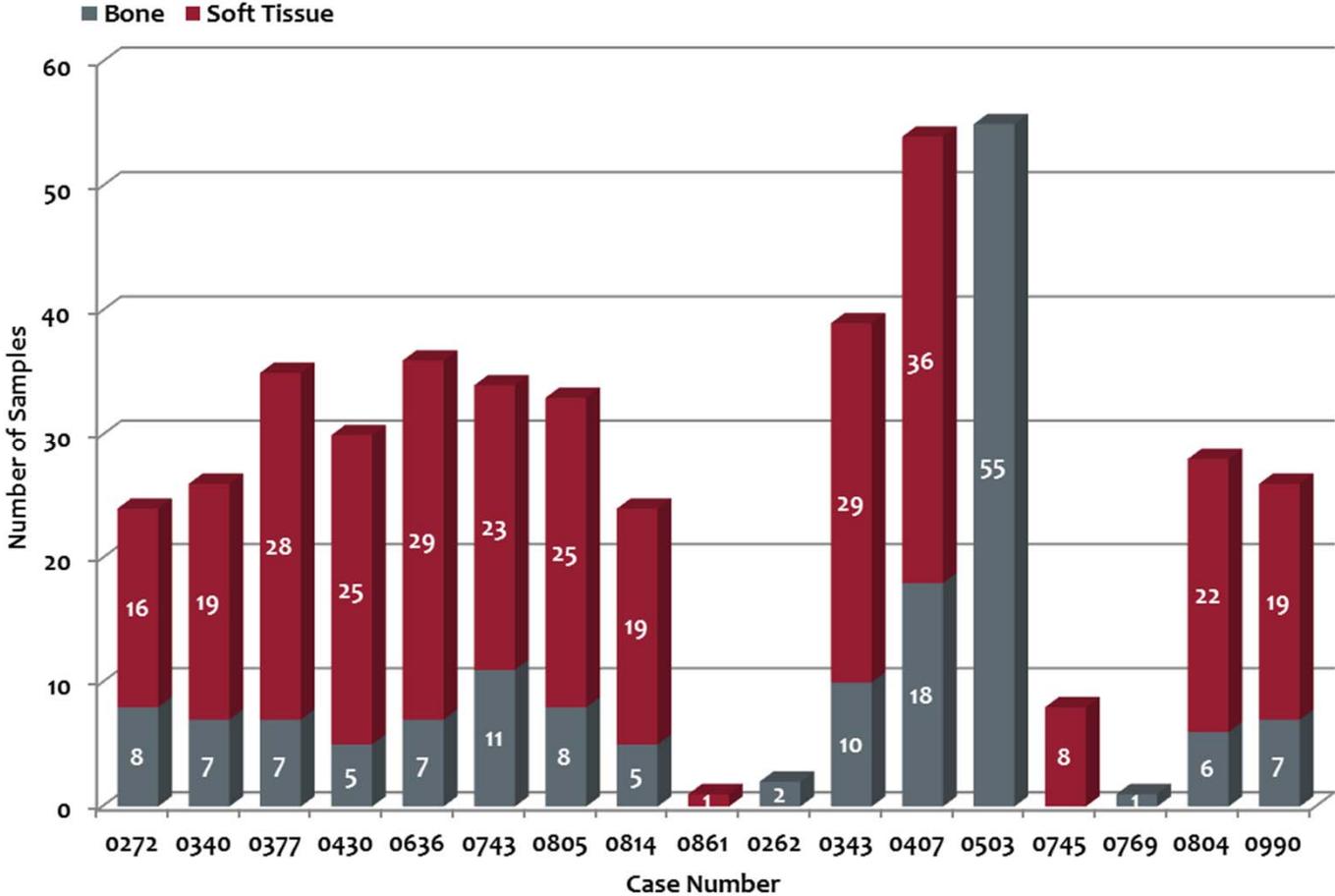
Radiochemical Analysis: *Tissue Type*

- Number of donations processed between 9/1/2014 – 8/1/2015 : 17
 - ✓ Whole-body: 8
 - ✓ Partial-body: 9



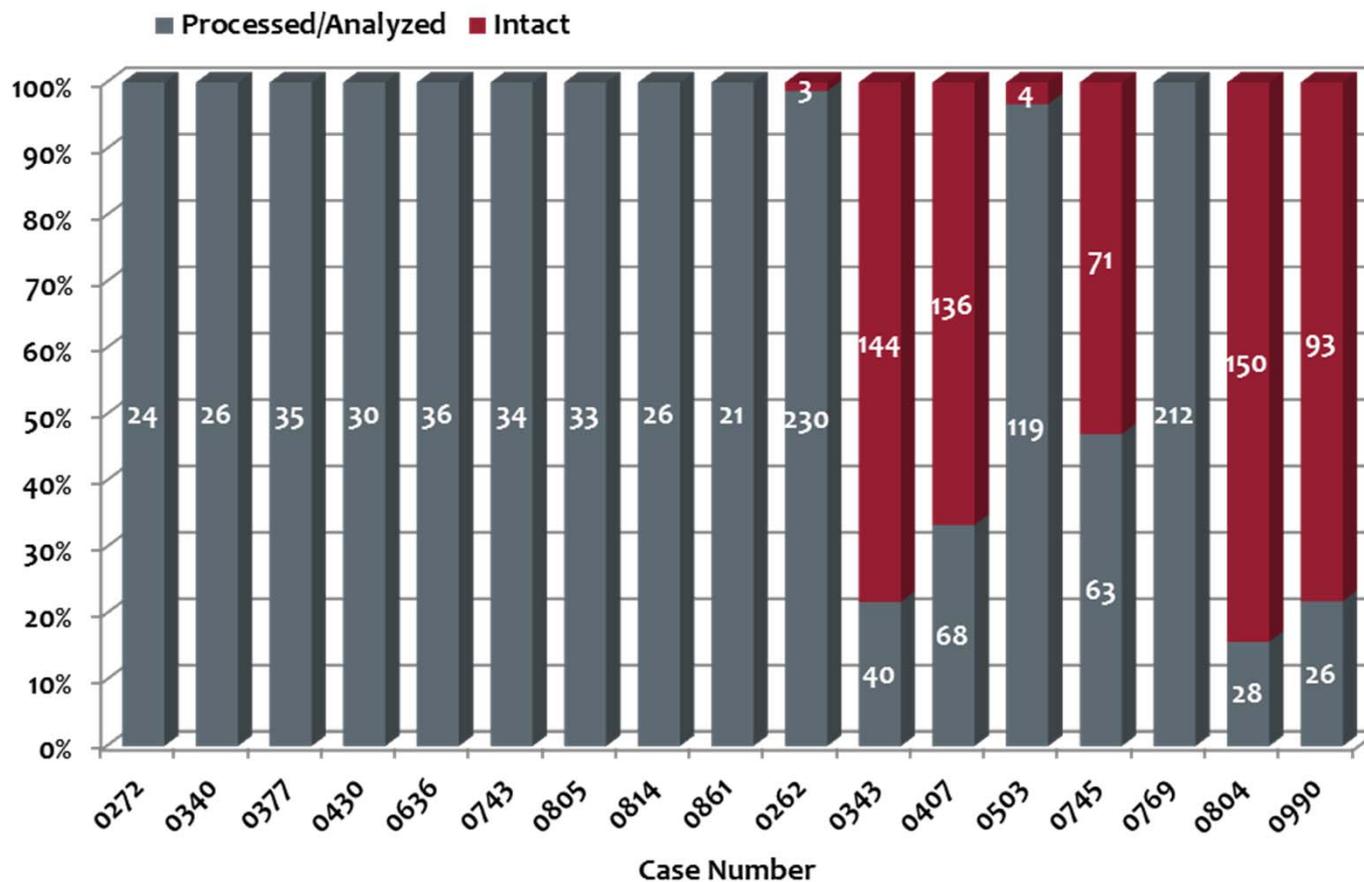


Donation Analysis Progress: *Tissue Type*





Donation Analysis Progress



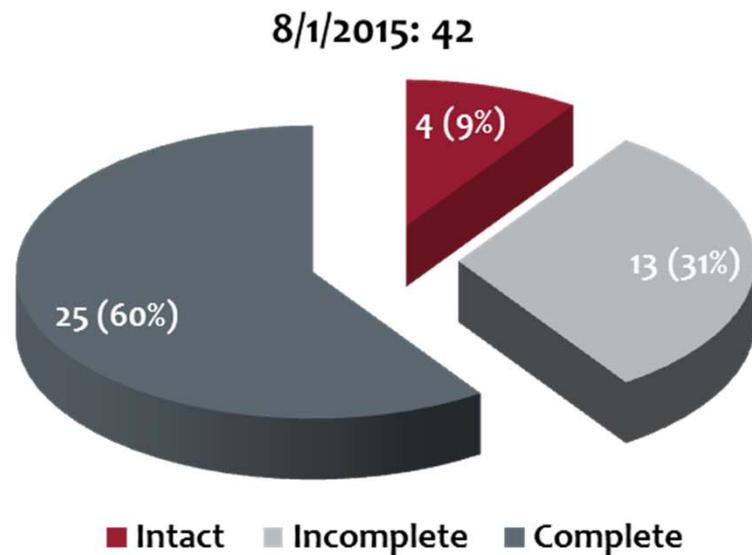
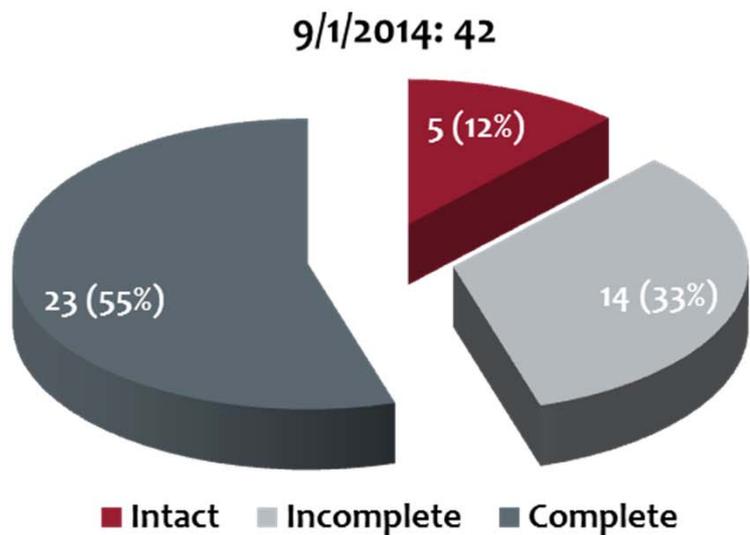


Donation Analysis Status Change: Whole-Body

- Number of cases: 3
 - ✓ Case 0503 (1994): incomplete → complete
 - ✓ Case 1007 (2005): incomplete → complete
 - ✓ Case 0990 (2005): intact → incomplete
- Analyzed method: Alpha Spectrometry (2) and ICP-MS (1)



Donation Analysis Status: Whole-Body





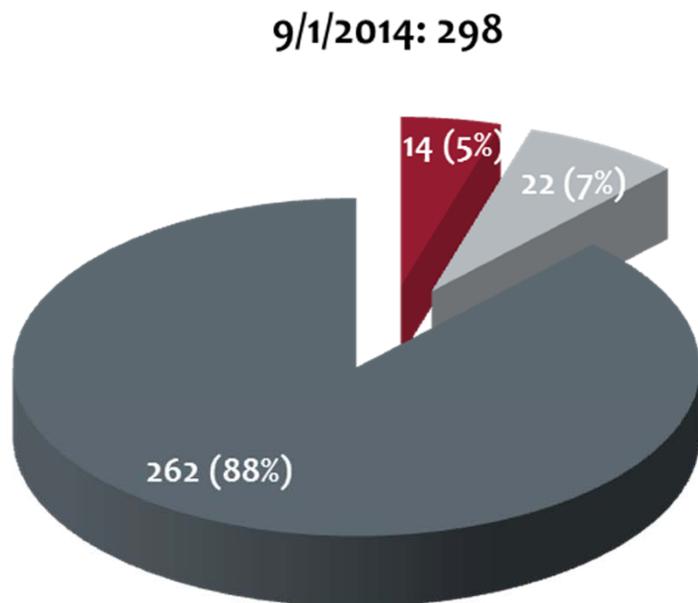
Donation Analysis Status Change: *Partial-Body*

- Number of cases: 9
 - ✓ Case 0430 (2005): intact → complete
 - ✓ Case 0340 (2008): intact → complete
 - ✓ Case 0636 (2012): intact → complete
 - ✓ Case 0814 (2013): incomplete → complete
 - ✓ Case 0272 (2013): intact → incomplete
 - ✓ Case 0743 (2013): intact → incomplete
 - ✓ Case 0805 (2014): intact → incomplete
 - ✓ Case 0377 (2015): intact → incomplete
 - ✓ Case 0837 (2003) complete → incomplete
- Analyzed by: Alpha Spectrometry

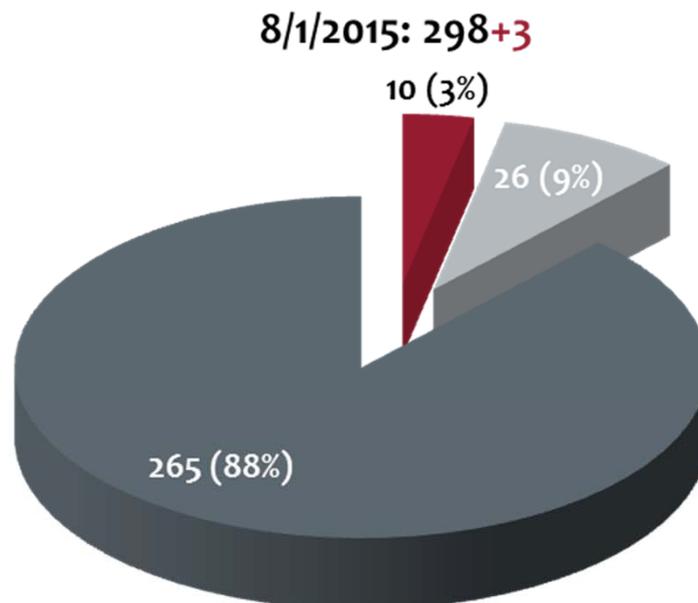




Donation Analysis Status: *Partial-Body*



■ Intact ■ Incomplete ■ Complete

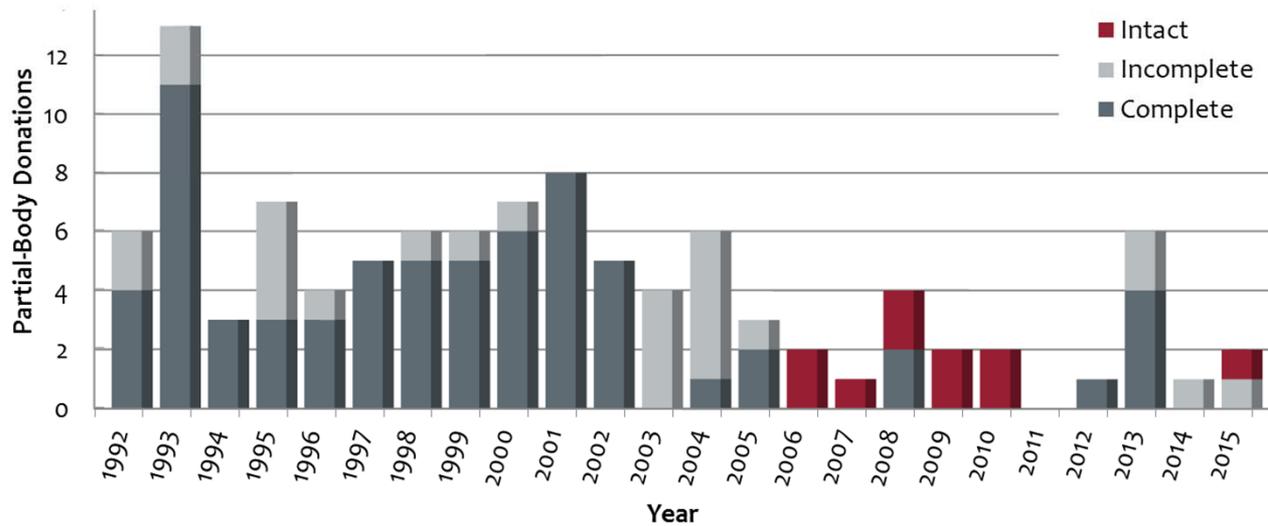
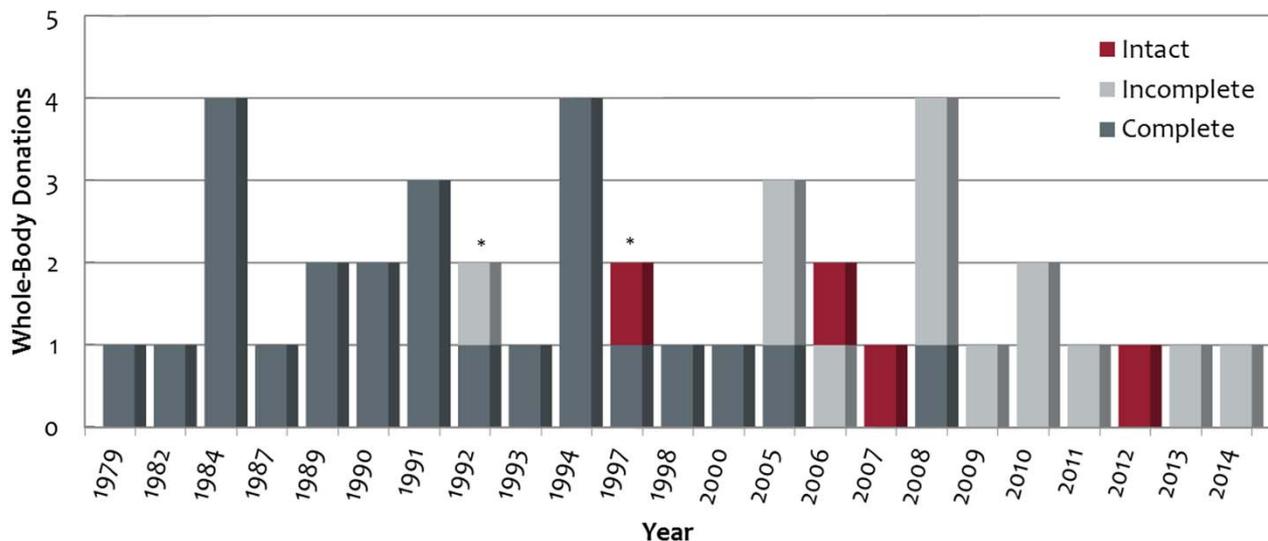


■ Intact ■ Incomplete ■ Complete





Donation Analysis Status Summary 2015





Equipment Upgrade and Facility Maintenance



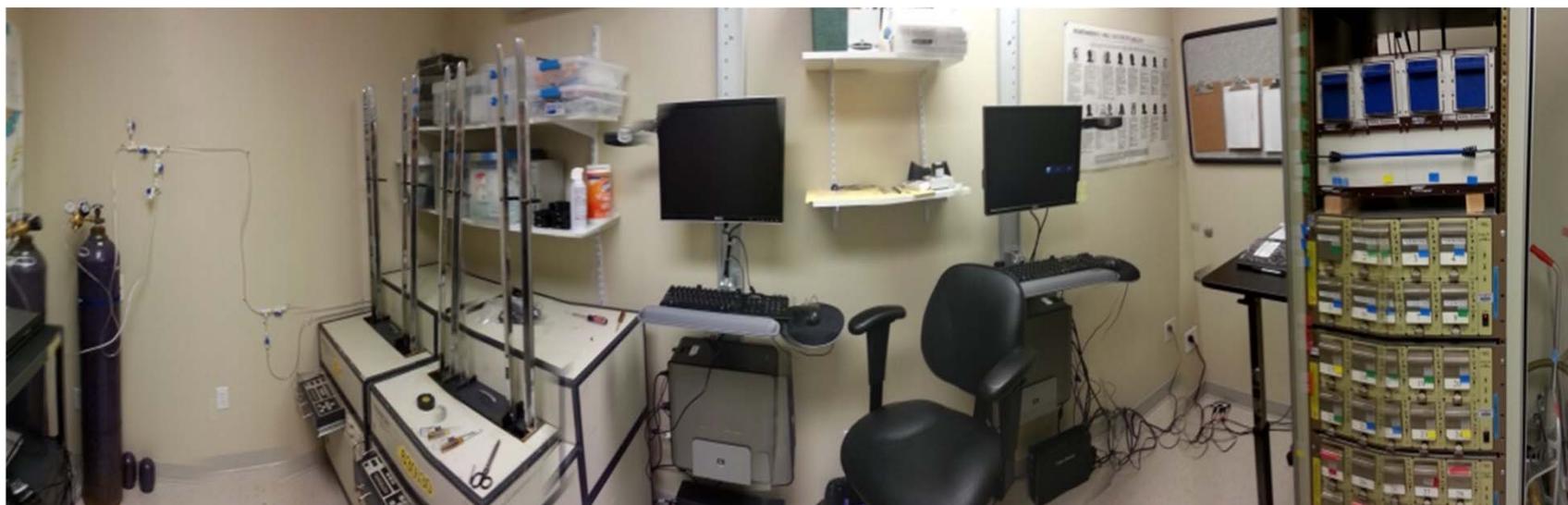


Counting Room

- Compact design utilizing ergonomic wall mounts
- Computer upgrade

**Tennelec LB-5100
Computer System**

**α -spectroscopy
Computer System**



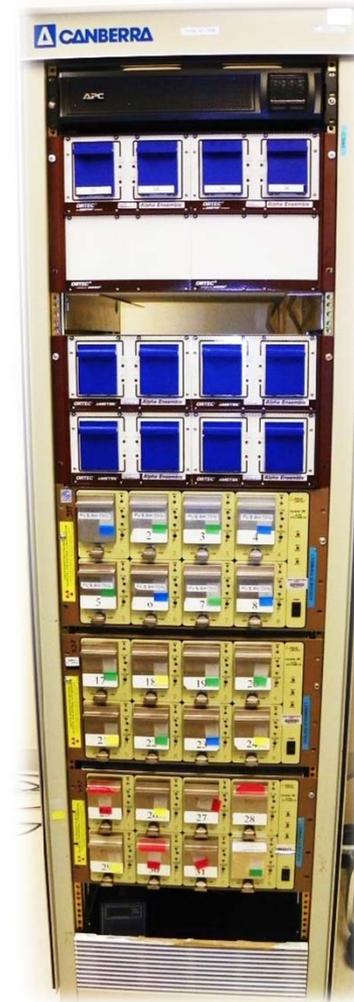


α -spectrometry System

- ALPHA-ENSEMBLE® 8-input benchtop spectrometer
- Low-background ENS-U450 ULTRA-AS detectors (8)
- Vacuum Pump
- Rack-mount kit
- Increased total capacity of the system – 36 detectors



9/1/2014



9/1/2015





UPS System

- Uninterruptible power supply (UPS) systems were purchased from APC
- Protects detectors and computer systems in the counting room from potential electrical surges and power outages.
 - ✓ degradation in detector performance and loss of data





New Electrical Circuit

- Installed two (2) 30Amp/120V circuits
- Required for APC UPS operation
- Special power outlet: L5-30





Sample Planchet Labeling

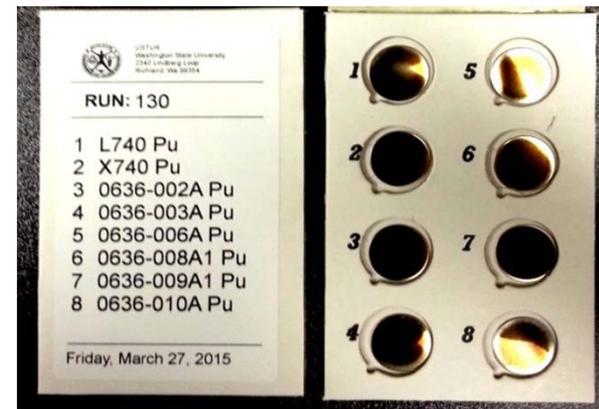
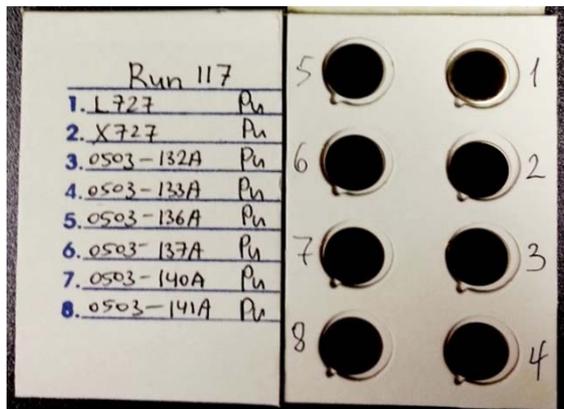
Before



After



Planchet engraving: (left) manual with stylus and (right) automatic with Gravograph M20



Planchet holders labeling: handwritten (left); printed and stamped position number (right).



Tennelec LB-5100 Gas-Flow Proportional Counters

- Computer system updated with new Eclipse software (version 3.3)
- GPIB-USB interface Installed
 - ✓ Faster communication between detectors and the computer
- Multiple GPIB cables replaced





Perkin Elmer BetaScout® LSC

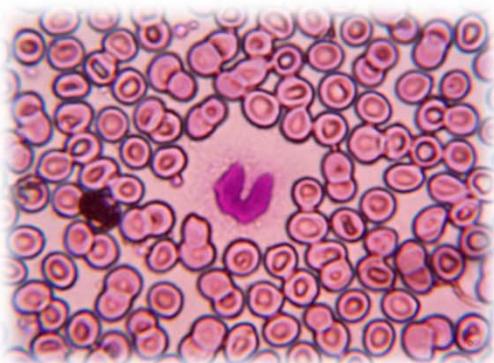
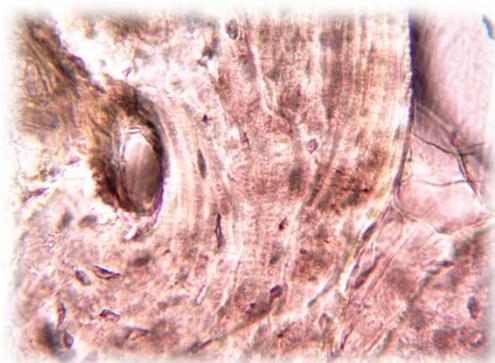
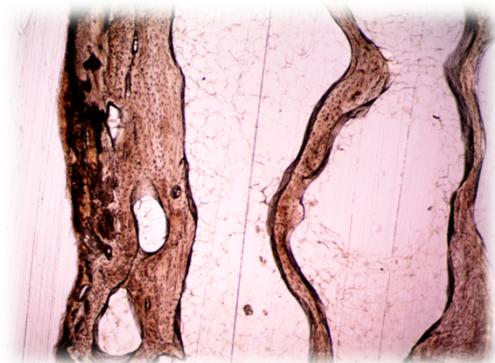
- Single-vial LSC
- Previously donated to USTUR
- Sent to Hidex (Lablogic: Brandon, FL) for service
- Firmware update, PMT check, test run
- Hidex control software and special cable
- Restored and fully functional





Microscope

- AmScope[®] T390 microscope
 - ✓ Photo port for digital image acquisition





Data Backup Server

- Network attached data storage (NAS) - 5 TBt
- Waterproof/fireproof
- Encrypted
- Instant disaster recovery
- Data remains local and all security details are 100% controlled by the user



ioSafe® NAS 1515+ (left) and USTUR office server (right)





US DOE MAPEP Study

RESL
Radiological and Environmental Sciences Laboratory



NIST Traceable *soil and water* *false positive testing*
real world samples *stable inorganic* **Radiochemical**
semi-volatile organics

M.A.P.E.P
Mixed Analyte Performance Evaluation Program

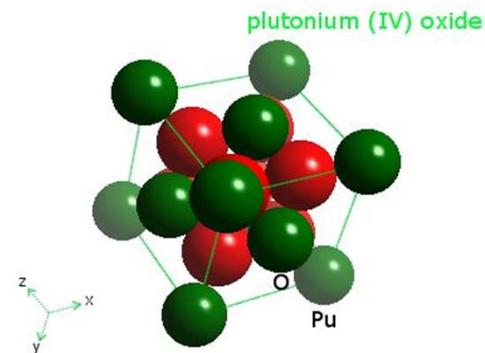
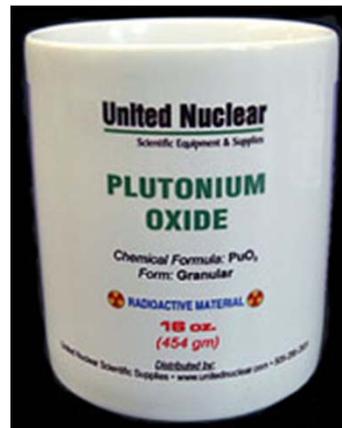




Motivation

Validation of USTUR analysis protocol: 'high-fired' plutonium oxide (PuO_2)

- MAPEP-11-MaS24: Radiological and Environmental Sciences Laboratory (RESL)
- Soil: ^{239}Pu in 'high-fired' form
- Certified: ^{239}Pu , ^{241}Am , ^{234}U , and ^{238}U





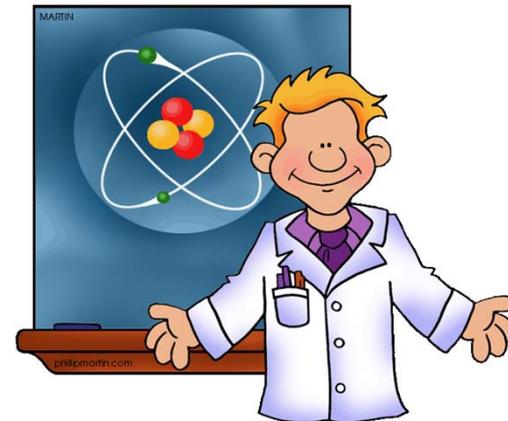
MAPEP Analysis Outline

Analysis: ^{239}Pu , ^{241}Am , and $^{234,238}\text{U}$

- USTUR Protocol:



- Sample size, g: 0.5x2, 1.0, 2.0
- Microwave Digestion
 - ✓ $\text{HNO}_3:\text{HCl}:\text{HF}$ – 9:4:3
 - ✓ $\text{HNO}_3:\text{HCl}$ – 9:4 (reverse *aqua regia*)
 - ✓ Temperature: 200 °C
 - ✓ Pressure: 40 bar
- TEVA-TRU-DGA: Pu/Am/U separation





MAPEP Results: Chemical Recovery

Sample Size, g	Chemical Recovery, %		
	^{243}Am	^{242}Pu	^{232}U
<i>Reagent Mixture $\text{HNO}_3 : \text{HCl} : \text{HF}$</i>			
1.0	34.2±1.8	98.2±3.0	102.7±2.4
0.5×2=1.0	33.4±7.4	101.6±3.7	96.7±3.9
1.0	27.3±1.4	101.6±2.8	92.0±4.7
2.0	42.3±3.5	101.8±3.6	101.6±4.2
Average:	34.3±6.2	100.8±1.7	98.3±4.9
<i>Reagent Mixture $\text{HNO}_3 : \text{HCl}$</i>			
1.0	23.0±4.0	101.6±8.3	91.0±7.1
~0.5 (residue)	98.4±1.5	101.7±4.0	93.9±7.0

- Soil matrix effect (Fe^{3+}): low $^{243}\text{Am}^{3+}$ tracer recovery





MAPEP Results: Measured vs Certified

Sample Size, g	Concentration, pCi g ⁻¹				Bias, %			
	²⁴¹ Am	²³⁹ Pu	²³⁴ U	²³⁸ U	²⁴¹ Am	²³⁹ Pu	²³⁴ U	²³⁸ U
<i>Reagent Mixture HNO₃ : HCl : HF</i>								
1.0	1.71±0.20	2.75±0.19	4.53±0.19	4.64±0.31	2.9	3.7	-4.8	-6.5
0.5×2=1.0	1.58±0.10	2.38±0.10	4.41±0.14	4.62±0.29	-4.7	-10.1	-7.3	-6.9
1.0	1.76±0.25	2.57±0.14	4.40±0.48	4.73±0.65	6.2	-3.2	-7.6	-4.6
2.0	1.50±0.20	2.48±0.09	4.32±0.17	4.47±0.17	-9.5	-6.4	-9.2	-9.8
Average:	1.64±0.12	2.54±0.16	4.41±0.09	4.61±0.11	-1.3	-4.0	-7.2	-7.0
<i>Reagent Mixture HNO₃ : HCl</i>								
1.0	0.37±0.07	0.14±0.01	1.33±0.14	1.39±0.18	-77.6	-94.9	-72.1	-71.9
~0.5 (residue)	1.05±0.11	2.42±0.10	3.09±0.18	3.28±0.13	-36.9	-8.6	-35.1	-33.8
<i>Reference Value</i>								
	1.66±0.02	2.65±0.04	4.76±0.14	4.96±0.15				

94.9% negative bias for ²³⁹Pu analysis w/o HF





MAPEP Summary

Complete “high-fired” PuO₂ digestion with HNO₃:HCl:HF

- ✓ USTUR measured vs. RESL Accepted values:

Concentration, pCi g ⁻¹			
²⁴¹ Am	²³⁹ Pu	²³⁴ U	²³⁸ U
USTUR			
1.64±0.12	2.54±0.16	4.41±0.09	4.61±0.11
RESL accepted range, min - max			
1.16 - 2.15	1.85 - 3.44	3.32 - 6.19	3.49 - 6.46

Incomplete digestion of PuO₂ with reverse *aqua regia* (HNO₃:HCl)

- ✓ 71.9 to 94.9% negative bias in Am/Pu/U results

Additional findings:

- ✓ No matrix effect on Pu and U chemistry: 92 – 102% tracer recovery
- ✓ Soil matrix effect on Am chemistry: <35% tracer recovery





Thank you for your Attention!

