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Beyond α-spectrometry for Actinide Determination in Human Tissues?

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U.S. Tansuranium & Uranium Registries (USTUR)

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USTUR: Human Tissue Analysis

- Tissue Digestion
- Actinide Separation
- Alpha Spectrometric Measurement

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ICP-MS: Alternative to α-spectrometry

- Inductively coupled plasma mass spectrometry (ICP-MS) is now a well established technique for ^{234,235,238}U and ^{239,240}Pu determination in environmental samples
- With recent ICP-MS instrument developments, ^{241}Am and ^{241}Pu determination in soil samples has been reported
- ICP-MS not previously applied for actinide determination in human tissue samples (low concentrations)
- USTUR routinely used α-spectrometry as "technique of choice" for ^{238}Pu and $^{239+240}$Pu and ^{241}Am in human tissues

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Human Tissue Samples for ICP-MS Trial

Case #	# of samples	α-spectrometry		
		$^{234,235,238}U$	$^{238, 239+240}Pu$	^{241}Am
0269	4	x	o	o
0425	8	o	o	o
0720	2	x	o	o
1028	6	o	x	x

Case # 0269, 0425, 0720 – Pu cases; #1028 – U case (HEU)

Case # 0425 (Pu) - analyzed for U (natural)

Total: 20 samples (9 bones and 11 soft tissues)

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NAU: ICP-MS instrument

- VG Axiom MC used in single collector mode
- ICP-MS Type: Sector Field
- Ultrasonic nebulizer
- Installed December 2001

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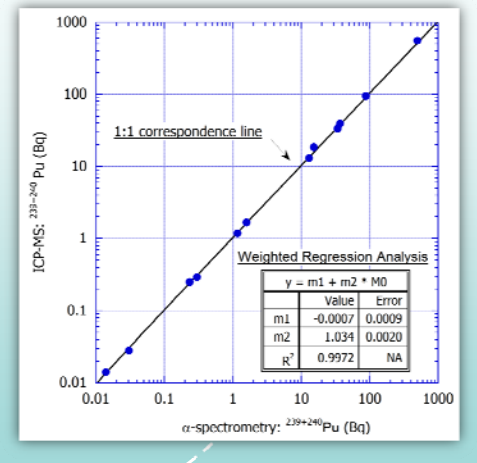
$^{239+240}$Pu: ICP-MS *vs* α-spectrometry

$^{239+240}$Pu

- **ICP-MS: ^{239}Pu, ^{240}Pu and ^{240}Pu/^{239}Pu ratio**
- **$^{239+240}$Pu was estimated for 13 samples (Pu cases)**
- **Gall bladder (0425.004) and all 6 samples from Uranium Case 1028: < ICP-MS LOD**

Wilcoxon matched-pairs test:

- ***p* = 0.0840**



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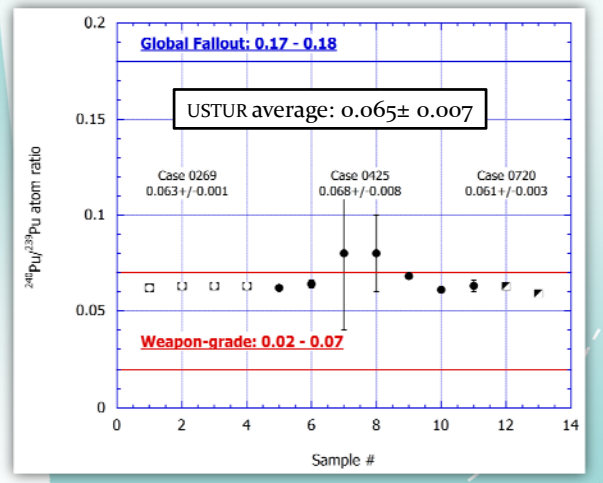
ICP-MS: ^{240}Pu/^{239}Pu atom ratio



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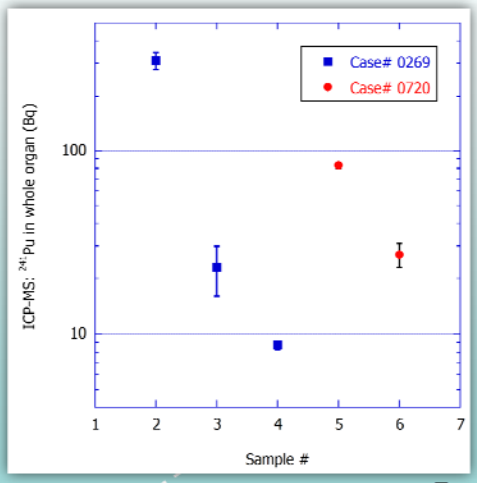
ICP-MS: Determination of ^{241}Pu

^{241}Pu

- $T_{1/2}$ = 14.1 y, β- emitter
- not detectable by α-spectrometry

^{241}Pu was detected in:

- liver (269.003)
- femur, PE (269.031)
- humerus, PE (269.052)
- lung (720.001)
- liver (720.004)



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^{241}Am: ICP-MS *vs* α-spectrometry

^{241}Am

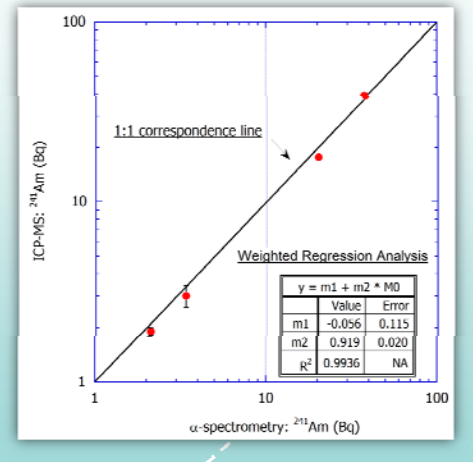
- $T_{1/2}$= 434.4 y, α-emitter
- never been measured by ICP-MS in human tissues

^{241}Am was detected in:

- liver (269.003)
- humerus, PE (269.052)
- lung (720.001)
- liver (720.004)

Wilcoxon matched-pairs test:

- *p* = 0.9263



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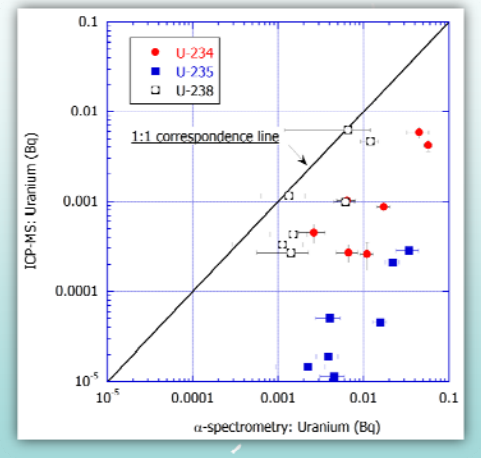
Uranium (nat.): ICP-MS *vs* α-spectrometry

Case #0425:

- **Pu case**
- **Not exposed to U_{anth}**
- **natural U, (low level)**

Wilcoxon matched-pairs test:

- ***p* = 0.0078 (^{234}U)**
- ***p* = 0.0078 (^{235}U)**
- ***p* = 0.0078 (^{238}U)**



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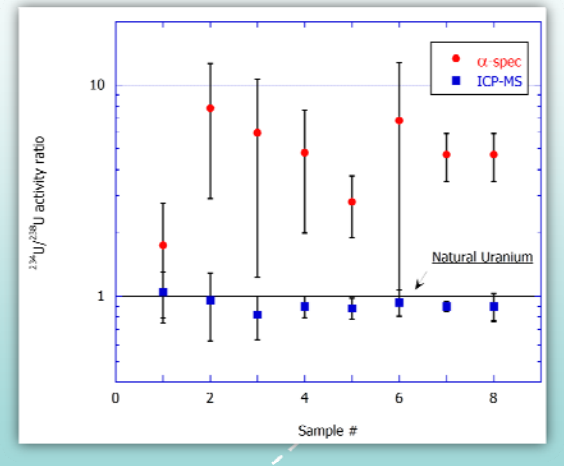
$^{234}U/^{238}U$: ICP-MS *vs* α-spectrometry

Case #0425:

- **Pu case**
- **Not exposed to U_{anth}**
- **natural U (low level)**

- **Activity ratio: $^{234}U/^{238}U = 1.0$**



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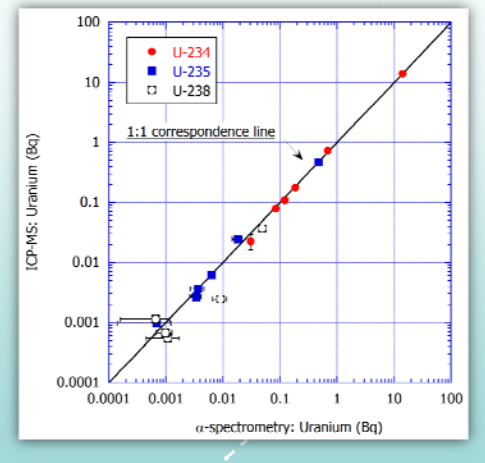
Uranium (anth.): ICP-MS *vs* α-spectrometry

Case #1028:

- U case
- exposed to U
- enriched U, high level

Wilcoxon matched-pairs test:

- $p = 0.9998$ (^{234}U)
- $p = 0.6875$ (^{235}U)
- $p = 0.3125$ (^{238}U)



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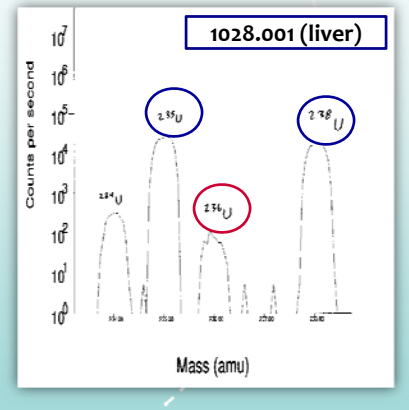
ICP-MS: Anthropogenic ^{236}U

Uranium (nat.) atom ratio:

- $^{235}U/^{238}U = 0.00725$
- $^{234}U/^{235}U = 0.00763$
- $^{236}U/^{238}U = 0.000$

Could never measure at USTUR



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Conclusions

✓This study confirmed the suitability of ICP-MS for determination of $^{239+240}$Pu, ^{240}Pu/^{239}Pu, ^{241}Am, and natural ^{234,235,238}U isotopes in bones and soft tissues of exposed individuals

✓ICP-MS ability to measure the ^{240}Pu/^{239}Pu isotopic ratio, ^{241}Pu and anthropogenic ^{236}U provides important new data - beyond α-spectrometry

✓Alpha spectrometry remains "technique of choice" for ^{238}Pu and low ^{241}Am determination

✓For the first time ^{241}Am was detected by ICP-MS in human tissue samples

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