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World Class. Face to Face.

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From Single Bone Analysis to Total Skeleton Content

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"Learning from Plutonium and Uranium Workers"

Goal

- Precise estimation of total actinide activity in skeleton

Approaches

- Analysis of the entire skeleton
- Analysis of selected/available bones

Pros and Cons

- Entire Skeleton Analysis:
  - *most precise estimation*
  - *very limited opportunities (USTUR)*
  - *large number of analyses*
- Analysis of Selected/Available Bones:
  - *typical scenario*
  - *how precise is the estimation?*

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

Registrant Statistics

Total Active (Living) and Deceased Registrants† :	416
Living Registrants:	81
Potential Partial-body Donors:	62
Potential Whole-body Donors:	12
Special Studies:	7
Deceased Registrants:	335
Partial-body Donations:	291
Whole-body Donations:	39
Special Studies:	5
Inactive Registrants:	458
Total Number of Registrants:	874

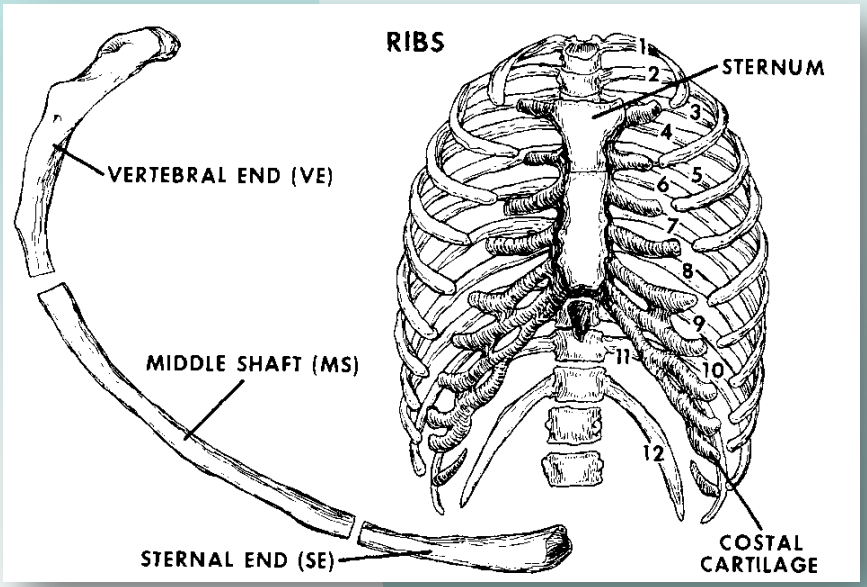
† - as of August 15, 2011

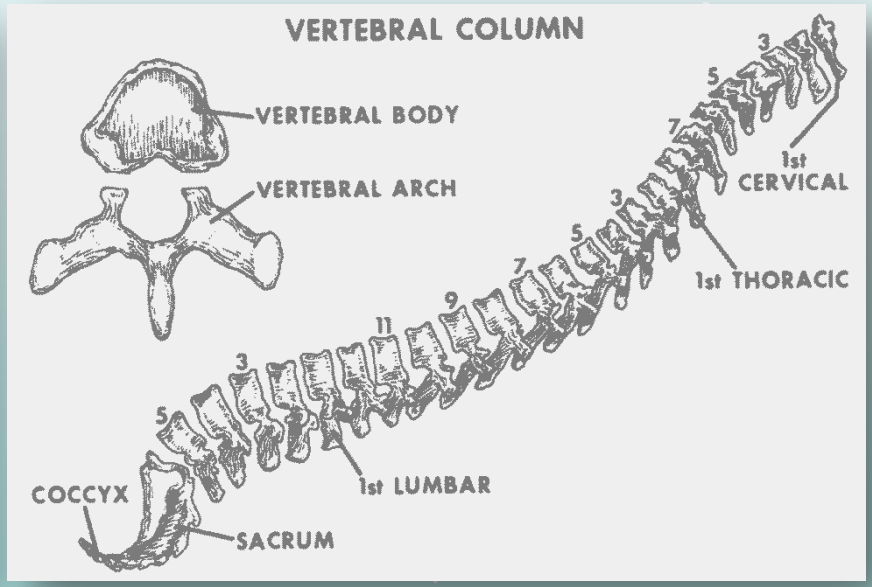
Human Tissue Analysis

- Autopsy/Dissection/Vacuum Packing/Inventory
- Tissue sample dry/wet ashing / Dissolution
- Actinide radiochemical separation
- Counting source preparation: electrodeposition
- Detection: α-spectrometry/ICP-MS

LANL Dissection Protocol

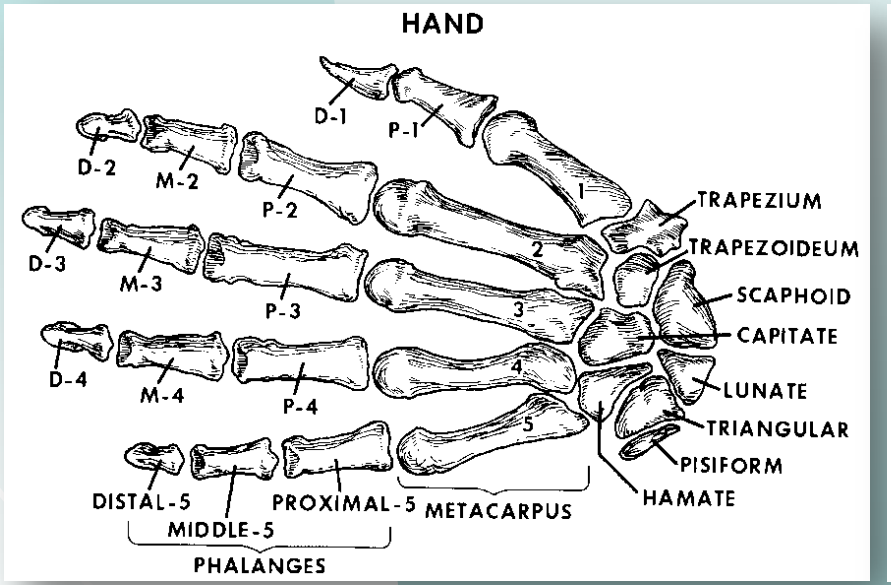
McInroy *et al* (1985, HPJ): *The U.S. Transuranium Registry Report of the ^{241}Am Content of a Whole Body. Part IV: Preparation and Analysis of the Tissues and Bones.*

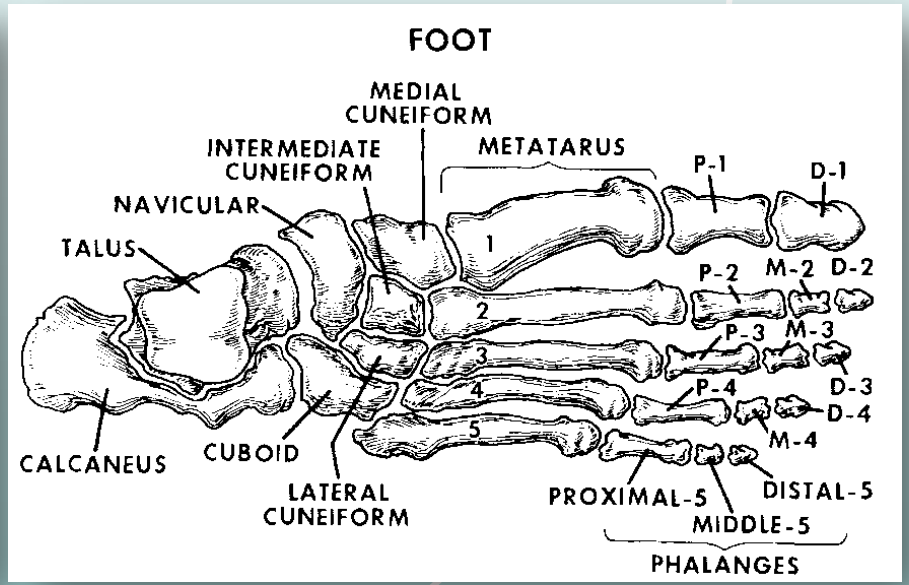




LANL Dissection Protocol (2)

McInroy *et al* (1985, HPJ): *The U.S. Transuranium Registry Report of the ^{241}Am Content of a Whole Body. Part IV: Preparation and Analysis of the Tissues and Bones.*





USTUR Dissection Protocol

Reduction in number of dissected samples:

- *whole ribs vs VE, MS, SE*
- *hand and wrist vs carpal bones + phalanges*
- *foot and ankle vs tarsal bones + phalanges*
- *fewer numbers of skin/fat/muscle samples*

Reduction in the number of analyzed samples

Whole Body: Number of Analyses

Case	Lab	Analysis†	Total	Bones	Ratio
0193‡	LANL	1982	423	307	73 %
0208	LANL	1984	307	170	55 %
0425	WSU	1994	120	87	73 %
0269	WSU	1994	106	84	79 %
0720	WSU	2005	127	84	66 %

† - Right side of the skeleton; odd ribs and vertebrae
‡ - Number of bones from the left side of the skeleton were analyzed

Whole- *vs* Partial- Body Analysis

Type	Skeleton	Number of Samples	
		Collected	Analyzed
Whole Body	Axial	110	62
	Limbs	70	50
Partial Body	Total	5 - 7†	5 - 7

† - ribs, clavicle, patella, sternum, vertebrae, femur (MS)

Total Skeletal Activity Calculation

Whole Body:

- Analysis: *right side of the skeleton*
- Calculation: *interpolation for even ribs and vertebrae*
- Total Activity: *activity in right side x 2*

Partial Body:

- Analysis: *rib(s), clavicle, patella, sternum, vertebra(e), femur (middle shaft)*
- Calculation: *weighted average skeletal concentration ICRP89 skeletal weight: -10.7 + 0.119H (cm)*
- Total Activity: *skeletal concentration × ICRP89 kg*

Can we do more from less?

Tasks

- 'Best bone' (group of bones) for the estimation of total actinide content in skeleton
- Precise estimation of skeleton content based on a limited number of bone analyses (partial-body donor, biopsy)
- Optimization of number of bone samples in whole-body analysis

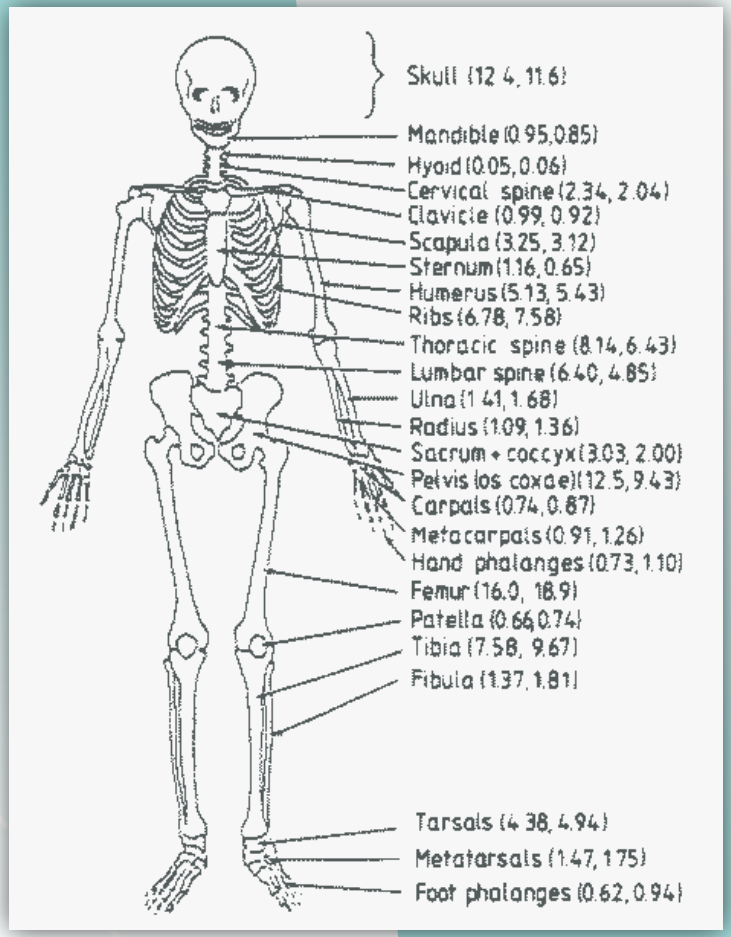
Actinide Distribution in Skeleton

- McInroy *et al* (1985, HPJ) *The U.S. Transuranium Registry Report of the ^{241}Am Content of a Whole Body. Part IV: Preparation and Analysis of the Tissues and Bones.*
- Lynch *et al* (1988, J. Radiol. Prot.) *Macrodistribution of plutonium and americium in four human skeletons.*
- Filipy *et al* (2003,HPJ) *Estimation of Actinide Skeletal Content in Humans Based on Bone Samples Collected At Autopsy.*

Methodology

- McInroy *et al* (1985, HPJ): *analysis of half of the skeleton. Single case analysis (first whole-body donation to the USTUR).*
- Lynch *et al* (1988, J. Radiol. Prot.) *defined a group of bones which best represents the total skeletal content. Analysis based on 4 whole-body cases.*
- Filipy *et al* (2003,HPJ): *least square regression analysis based on data from 8 whole-body cases.*

Distribution of ^{239}Pu in Skeleton



Lynch et al (1988, J. Radiol. Prot.) ***Macrodistribution of plutonium and americium in four human skeletons***

- Skull: 12.4 ± 1.1%
- Cervical Spine: 2.3 ± 0.2%
- Thoracic Spine: 8.1 ± 2.2%
- Lumbar Spine: 6.4 ± 2.0%
- Sacrum & Coccyx: 3.0 ± 0.7%
- Pelvis: 12.5 ± 1.2%
- Clavicles: 0.99 ± 0.14%
- Scapulae: 3.3 ± 0.3%
- Ribs: 6.8 ± 0.3%
- Sternum: 1.2 ± 0.2%
- Humeri: 5.1 ± 0.8%
- Femora: 16.0 ± 2.2%
- Tibiae: 7.6 ± 1.1%
- Patellae: 0.66 ± 0.09%

Proposed Methodology

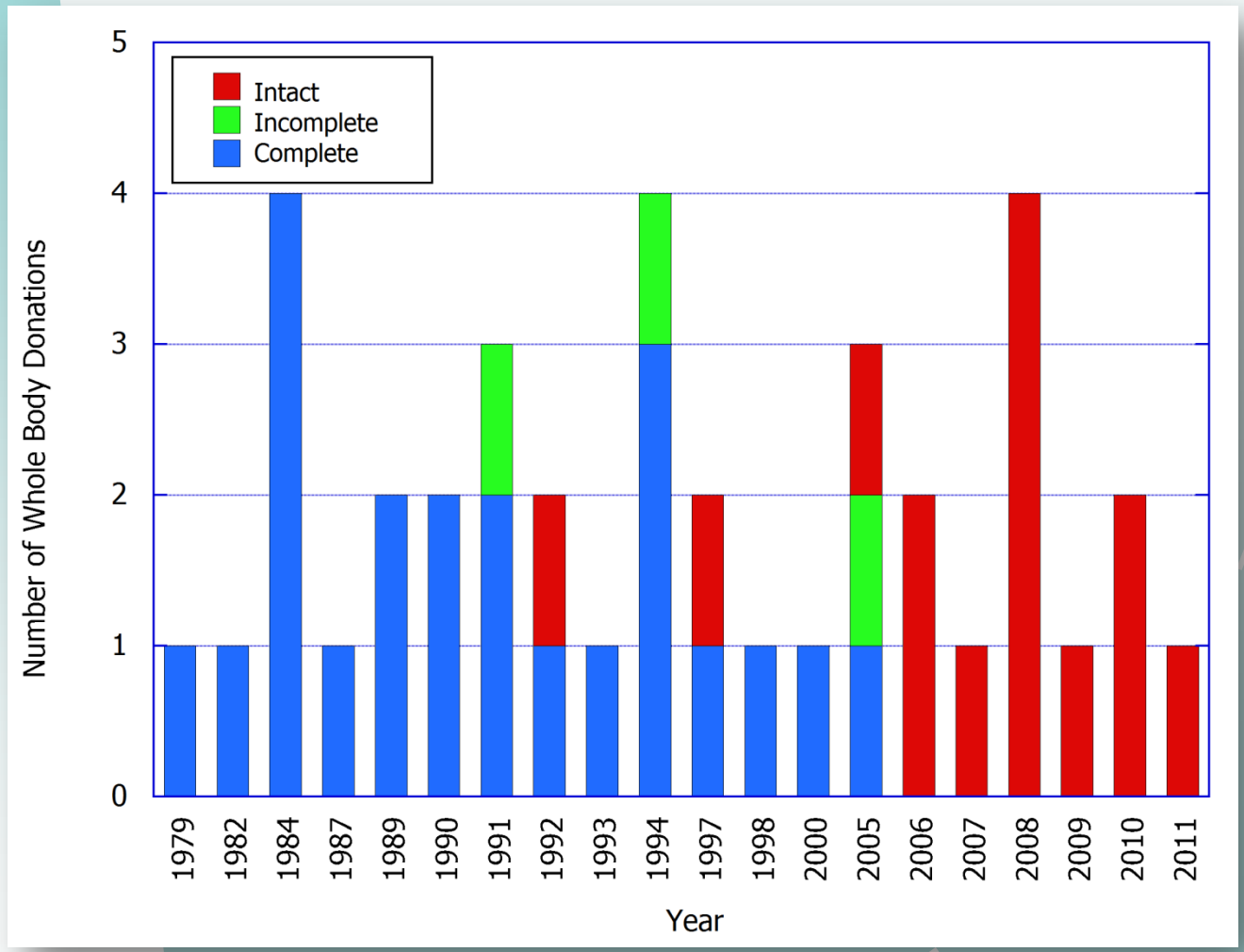
- $\text{Ratio} = \dfrac{\text{Individual Bone Concentration}}{\text{Total Skeletal Concentration}}$

- ‘Best bone’ $= \dfrac{C_{bone},\ \text{Bq kg}^{-1}}{C_{skeleton},\ \text{Bq kg}^{-1}} \rightarrow 1 \pm (\text{UNC} \rightarrow \text{min})$

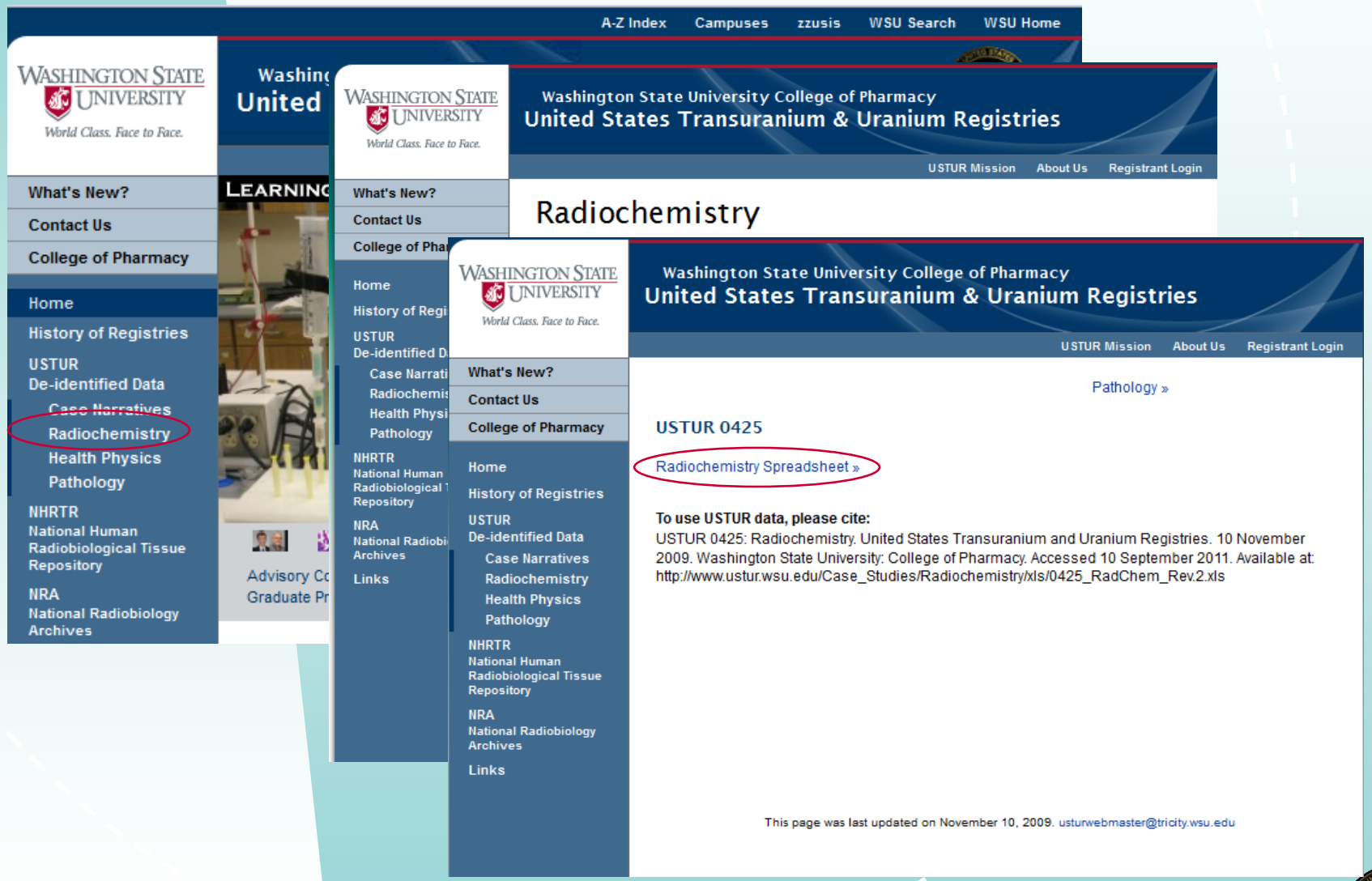
'To Do' List

- Conduct radiochemical analysis of available whole-body cases
- Calculate skeletal concentrations
- Populate set of $C_{bone}/C_{skeleton}$ for analyzed cases

Radiochemistry Status: *Whole- Body Cases*



Radiochemistry Results: *Where to Find?*



Radiochemistry Results: *What is Inside?*

USTUR Case 0425 - Derived Whole Body Pu/Am Contents

USTUR Case 0425 - Derived Whole Body Pu/Am Contents

Organ or Tissue	Wet Weight, g	Activity, Bq						Isotopic Activity Ratio, %				Activity Concentration in Wet Tissue, Bq kg^{-1}					
		$^{239+240}$Pu		^{238}Pu		^{241}Am		^{238}Pu:$^{239+240}$Pu		^{241}Am:$^{239+240}$Pu		$^{239+240}$Pu		^{238}Pu		^{241}Am	
		Meas.	s	Meas.	s	Meas.	s	Meas.	s	Meas.	s	Meas.	s	Meas.	s	Meas.	s
Calvarium:	486	1.53E+00	5.54E-02	2.06E-02	1.08E-02	4.99E-01	3.50E-02	1.35%	0.70%	32.6%	2.6%	3.15E+00	1.14E-01	4.24E-02	2.21E-02	1.03E+00	7.20E-02
Total Skull (R + L):	1093	2.72E+00	6.87E-02	2.70E-02	1.69E-02	9.40E-01	6.02E-02	0.99%	0.62%	34.5%	2.4%	2.49E+00	6.28E-02	2.47E-02	1.55E-02	8.60E-01	5.51E-02
Frontal 1	38.1	9.46E-02	6.60E-03	2.09E-04	1.24E-03	4.92E-02	5.13E-03	0.22%	1.31%	52.0%	6.5%	2.48E+00	1.73E-01	5.48E-03	3.24E-02	1.29E+00	1.35E-01
Frontal 2	14.4	3.71E-02	2.22E-03	6.59E-04	4.52E-04	1.72E-02	2.85E-03	1.78%	1.22%	46.4%	8.2%	2.57E+00	1.54E-01	4.58E-02	3.14E-02	1.19E+00	1.98E-01
Frontal 3	44.8	1.48E-01	8.08E-03	2.33E-03	1.35E-03	4.39E-02	4.57E-03	1.57%	0.91%	29.7%	3.5%	3.31E+00	1.80E-01	5.20E-02	3.00E-02	9.81E-01	1.02E-01
Parietal 1	64.9	1.96E-01	7.62E-03	3.15E-03	9.28E-04	6.40E-02	4.63E-03	1.61%	0.48%	32.6%	2.7%	3.02E+00	1.17E-01	4.85E-02	1.43E-02	9.86E-01	7.13E-02
Parietal 2	103.3	3.70E-01	2.50E-02	5.50E-03	5.04E-03	1.05E-01	1.58E-02	1.49%	1.37%	28.5%	4.7%	3.58E+00	2.42E-01	5.33E-02	4.88E-02	1.02E+00	1.53E-01
Temporal 1	38.8	1.06E-01	6.26E-03	2.92E-03	1.05E-03	3.73E-02	2.99E-03	2.76%	1.01%	35.2%	3.5%	2.73E+00	1.61E-01	7.53E-02	2.71E-02	9.61E-01	7.70E-02
Temporal 2	65.5	1.45E-01	6.72E-03	3.93E-03	1.33E-03	5.12E-02	4.70E-03	2.71%	0.93%	35.3%	3.6%	2.22E+00	1.03E-01	6.00E-02	2.04E-02	7.82E-01	7.17E-02
Temporal 3	16.2	2.52E-02	2.57E-03	-5.02E-04	7.32E-04	9.69E-03	2.19E-03			38.4%	9.5%	1.56E+00	1.58E-01	-3.10E-02	4.52E-02	5.98E-01	1.35E-01
Occipital	36.7	1.05E-01	6.14E-03	1.44E-03	1.08E-03	3.10E-02	3.06E-03	1.38%	1.03%	29.6%	3.4%	2.85E+00	1.67E-01	3.93E-02	2.93E-02	8.44E-01	8.33E-02
Maxilla (with teeth)	81.7	3.08E-02	8.74E-03	-7.19E-03	5.14E-03	2.50E-02	2.00E-02			81.1%	68.9%	3.77E-01	1.07E-01	-8.80E-02	6.29E-02	3.06E-01	2.45E-01
Mandible (with teeth):	145.8	2.23E-01	2.01E-02	1.51E-02	1.01E-02	1.00E-01	1.57E-02	6.75%	4.55%	44.9%	8.1%	1.53E+00	1.38E-01	1.03E-01	6.90E-02	6.88E-01	1.07E-01
Hyoid:								-	-	-	-						
Total Vertebrae:	2033.0	7.59E+00	7.71E-02	1.28E-01	1.32E-02	2.62E+00	3.02E-02	1.68%	0.17%	34.5%	0.5%	3.73E+00	3.79E-02	6.28E-02	6.47E-03	1.29E+00	1.49E-02
Whole Vertebrae																	
Cervical #1	38.9	2.27E-01	3.05E-02	9.91E-03	7.13E-03	7.15E-02	6.61E-03					5.85E+00	7.83E-01	2.55E-01	1.83E-01	1.84E+00	1.70E-01
Cervical #2	31.1	1.82E-01	1.22E-02	7.93E-03	2.85E-03	5.72E-02	2.64E-03	4.36%	1.59%	31.5%	2.6%	5.85E+00	3.92E-01	2.55E-01	9.17E-02	1.84E+00	8.50E-02
Cervical #3	23.2	1.06E-01	5.27E-03	5.16E-03	1.30E-03	3.63E-02	1.92E-03	4.88%	1.26%	34.3%	2.5%	4.56E+00	2.27E-01	2.23E-01	5.62E-02	1.56E+00	8.30E-02
Cervical #4	25.8	1.18E-01	7.69E-03	5.01E-03	1.74E-03	3.74E-02	2.58E-03										
Cervical #5	28.3	1.30E-01	5.52E-03	4.65E-03	1.09E-03	3.81E-02	1.60E-03	3.57%	0.85%	29.3%	1.7%	4.60E+00	1.95E-01	1.64E-01	3.86E-02	1.34E+00	5.66E-02
Cervical #6	30.5	1.42E-01	8.69E-03	5.32E-03	1.83E-03	4.73E-02	3.66E-03										
Cervical #7	32.6	1.54E-01	6.78E-03	6.06E-03	1.50E-03	5.73E-02	3.47E-03	3.94%	0.99%	37.2%	2.8%	4.72E+00	2.08E-01	1.86E-01	4.60E-02	1.76E+00	1.06E-01
Cervical #8	40.4	1.76E-01	1.04E-02	5.16E-03	1.96E-03	6.59E-02	4.74E-03										
Thoracic #1	48.1	1.96E-01	7.48E-03	4.06E-03	1.05E-03	7.20E-02	2.51E-03	2.07%	0.54%	36.7%	1.9%	4.08E+00	1.56E-01	8.44E-02	2.17E-02	1.50E+00	5.22E-02
Thoracic #2	44.7	1.97E-01	9.43E-03	3.28E-03	1.29E-03	6.81E-02	4.22E-03										
Thoracic #3	41.2	1.96E-01	6.06E-03	2.53E-03	7.99E-04	6.41E-02	3.17E-03	1.29%	0.41%	32.7%	1.9%	4.75E+00	1.47E-01	6.14E-02	1.94E-02	1.56E+00	7.70E-02
Thoracic #4	53.0	2.50E-01	1.06E-02	3.99E-03	1.52E-03	8.29E-02	5.61E-03										
Thoracic #5	64.7	2.95E-01	7.72E-03	6.01E-03	1.18E-03	1.01E-01	4.79E-03	2.04%	0.41%	34.1%	1.9%	4.55E+00	1.19E-01	9.29E-02	1.83E-02	1.55E+00	7.40E-02
Thoracic #6	57.9	2.51E-01	1.29E-02	5.53E-03	2.22E-03	8.77E-02	6.46E-03										
Thoracic #7	51.0	2.08E-01	9.75E-03	4.75E-03	1.79E-03	7.56E-02	3.97E-03	2.29%	0.87%	36.5%	2.6%	4.07E+00	1.91E-01	9.31E-02	3.51E-02	1.48E+00	7.79E-02
Thoracic #8	77.9	3.11E-01	1.64E-02	7.10E-03	2.74E-03	1.07E-01	7.43E-03										
Thoracic #9	104.7	3.96E-01	1.56E-02	7.67E-03	2.37E-03	1.29E-01	4.93E-03	1.94%	0.60%	32.6%	1.8%	3.78E+00	1.49E-01	7.32E-02	2.27E-02	1.23E+00	4.71E-02
Thoracic #10	111.2	4.07E-01	1.33E-02	4.81E-03	2.09E-03	1.38E-01	5.05E-03										
Thoracic #11	117.6	4.17E-01	1.99E-02	1.57E-03	2.98E-03	1.47E-01	6.35E-03	0.38%	0.72%	35.4%	2.3%	3.54E+00	1.69E-01	1.34E-02	2.54E-02	1.25E+00	5.40E-02
Thoracic #12	105.7	3.59E-01	1.65E-02			1.30E-01	5.57E-03										
Lumbar #1	201.0	5.38E-01	1.18E-02	5.21E-03	1.53E-03	2.38E-01	6.43E-03	0.97%	0.28%	44.3%	1.5%	2.67E+00	5.88E-02	2.59E-02	7.59E-03	1.19E+00	3.20E-02
Lumbar #2	196.0	6.54E-01	2.99E-02	6.66E-03	4.26E-03	2.36E-01	1.26E-02										
Lumbar #3	190.9	7.03E-01	2.25E-02	9.11E-03	3.25E-03	2.27E-01	9.08E-03	1.30%	0.46%	32.3%	1.7%	3.68E+00	1.18E-01	4.77E-02	1.70E-02	1.19E+00	4.76E-02
Lumbar #4	169.3	5.55E-01	2.87E-02	4.92E-03	4.54E-03	1.75E-01	1.27E-02										
Lumbar #5	147.7	4.19E-01	1.71E-02	1.28E-03	3.04E-03	1.30E-01	7.61E-03	0.31%	0.73%	31.0%	2.2%	2.84E+00	1.16E-01	8.66E-03	2.06E-02	8.79E-01	5.15E-02

WB_Summary_Pu_Am_ACJ | Pu_Am Sample Results | Data_NRC_AE_NAU_GEL | RadChem_Pu_Am_SYT

http://www.ustur.wsu.edu/Case_Studies/Radiochemistry/html/0425_RadChem.php

Work in Progress

Case	Age, y	Osteo-porotic	Primary Nuclide	Skeletal Conc., Bq wt.kg^{-1}	Cbone /Cskeleton				
					Ribs	Stern.	Vert.	Patella	Clavic.
0193	62		^{239}Pu	5.83	0.77	1.24	1.10	1.21	1.04
0208	69		^{239}Pu	10.9	0.74	1.29	0.62	1.24	1.00
0212	56	no	^{239}Pu	10.9	0.74	1.03	0.75	2.06	1.18
0213	68	no	^{239}Pu	20.2	0.86	1.18	1.06	1.42	0.87
0242	77		^{239}Pu	42.2	0.68	0.95	0.78	1.28	1.05
0259	55	no	^{238}Pu	10.53	0.64	0.65	0.63	1.66	0.81
0425	83	no	^{239}Pu	2.82	1.61	1.10	1.46	0.51	0.99
0679	72	yes	^{239}Pu	10.52	1.29	0.62	1.13	-----	1.04
0720	84	yes	^{239}Pu	8.49	0.70	0.71	1.16	0.91	1.22
0744	67	no	^{239}Pu	8.62	1.2	-----	0.98	1.27	1.00
0269	79	yes	^{239}Pu	119.1	0.91	1.01	1.01	1.16	1.05
0682	71	no	^{238}Pu	27.3	1.09	0.80	1.23	1.41	0.67

Work Summary

Case	Age, y	Osteo-porotic	Primary Nuclide	Cbone /Cskeleton					Variability by Case
				Ribs	Stern.	Vert.	Patella	Clavic.	
0193	62		^{239}Pu	0.77	1.24	1.10	1.21	1.04	1.07±0.19
0208	69		^{239}Pu	0.74	1.29	0.62	1.24	1.00	0.98±0.30
0212	56	no	^{239}Pu	0.74	1.03	0.75	2.06	1.18	1.15±0.54
0213	68	no	^{239}Pu	0.86	1.18	1.06	1.42	0.87	1.08±0.23
0242	77		^{239}Pu	0.68	0.95	0.78	1.28	1.05	0.95±0.23
0259	55	no	^{238}Pu	0.64	0.65	0.63	1.66	0.81	0.88±0.44
0425	83	no	^{239}Pu	1.61	1.10	1.46	0.51	0.99	1.13±0.43
0679	72	yes	^{239}Pu	1.29	0.62	1.13	-----	1.04	1.02±0.29
0720	84	yes	^{239}Pu	0.70	0.71	1.16	0.91	1.22	0.94±0.24
0744	67	no	^{239}Pu	1.2	-----	0.98	1.27	1.00	1.11±0.14
0269	79	yes	^{239}Pu	0.91	1.01	1.01	1.16	1.05	1.03±0.09
0682	71	no	^{238}Pu	1.09	0.80	1.23	1.41	0.67	1.04±0.30
12	70 ±10	all	Pu	0.94 ±0.30	0.96 ±0.24	0.99 ±0.25	1.28 ±0.39	0.99 ±0.15	Variability by Bone

Conclusion

- ‘Best bone’ = $\frac{C_{bone},\ Bq\ kg^{-1}}{C_{skeleton},\ Bq\ kg^{-1}}$ → 1 ± (UNC → min)

Clavicle is the one: 0.99 ± 0.15

Future Work

- Calculate $C_{bone}/C_{skeleton}$ using bone ash weights.
- Calculate $C_{bone}/C_{skeleton}$ for other bones typically collected at autopsy from partial-body donors: femur (whole, or middle shaft), scapula, calvarium.
- Populate $C_{bone}/C_{skeleton}$ for all individual bones.
- Apply advanced mathematical algorisms to define 'best group' of 3-5-7 bones.

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Thank you!