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Secular disequilibrium in materials: implications on background estimates for rare event detectors

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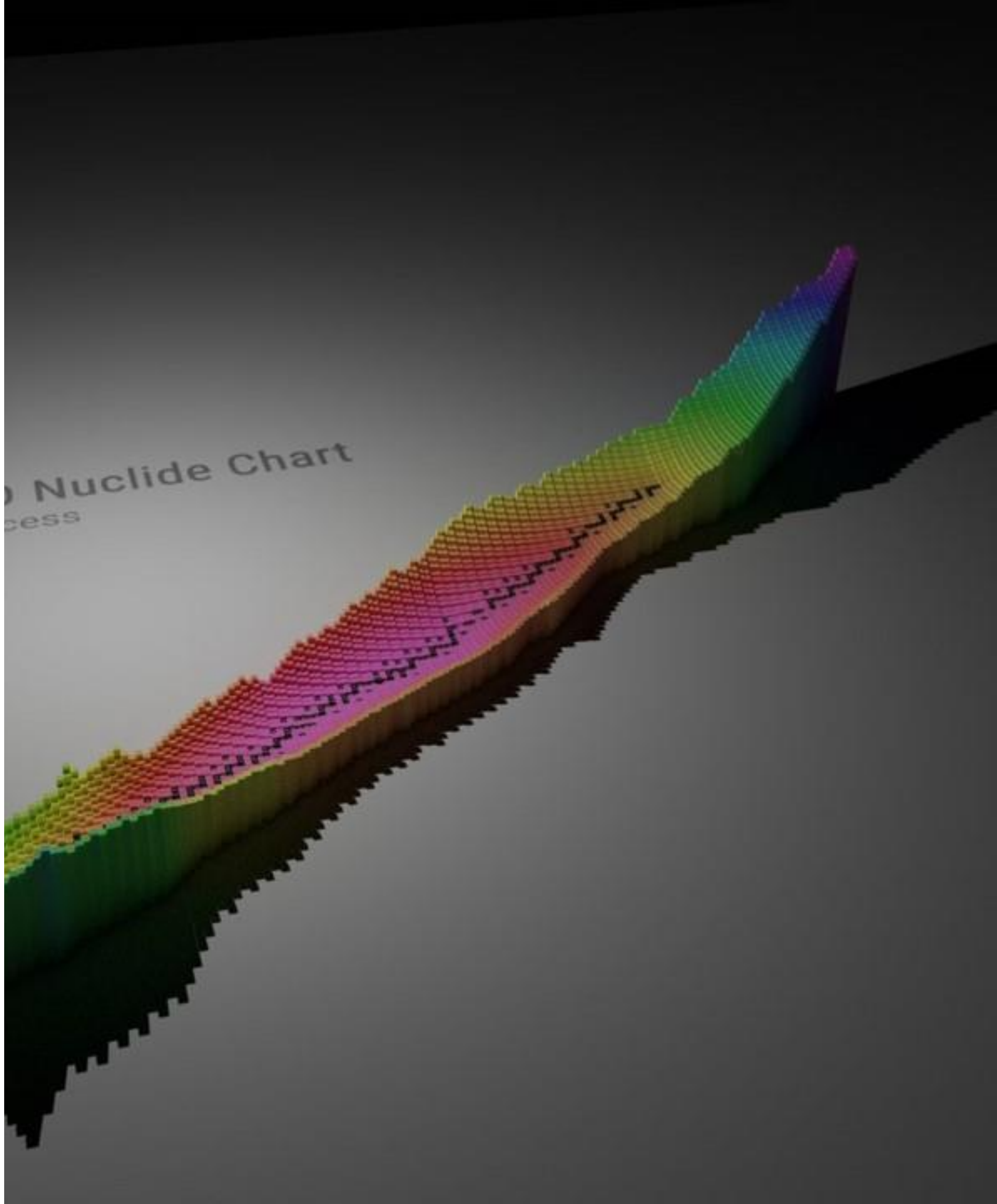
Kirby Hobbs

Isaac Arnquist
M. Laura Di Vacri
Tyler Schlieder



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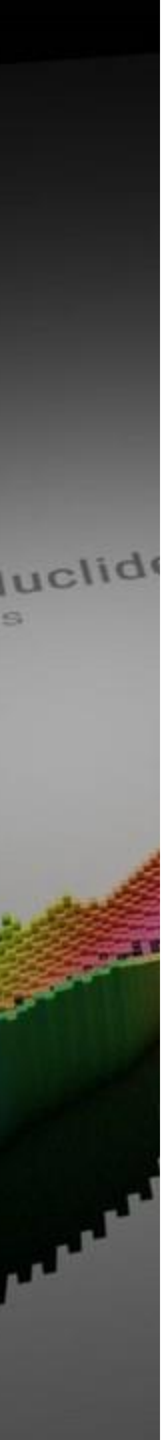
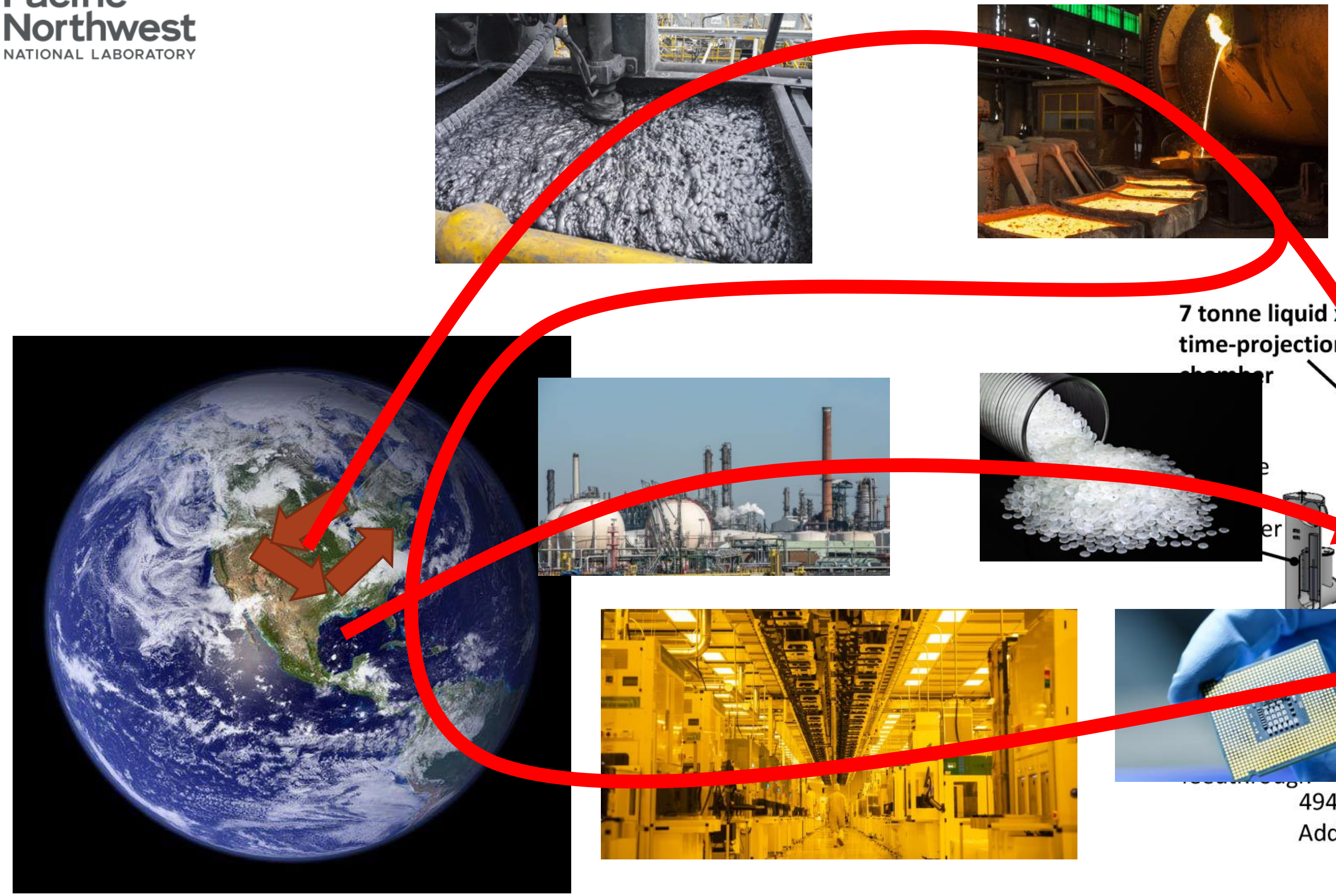
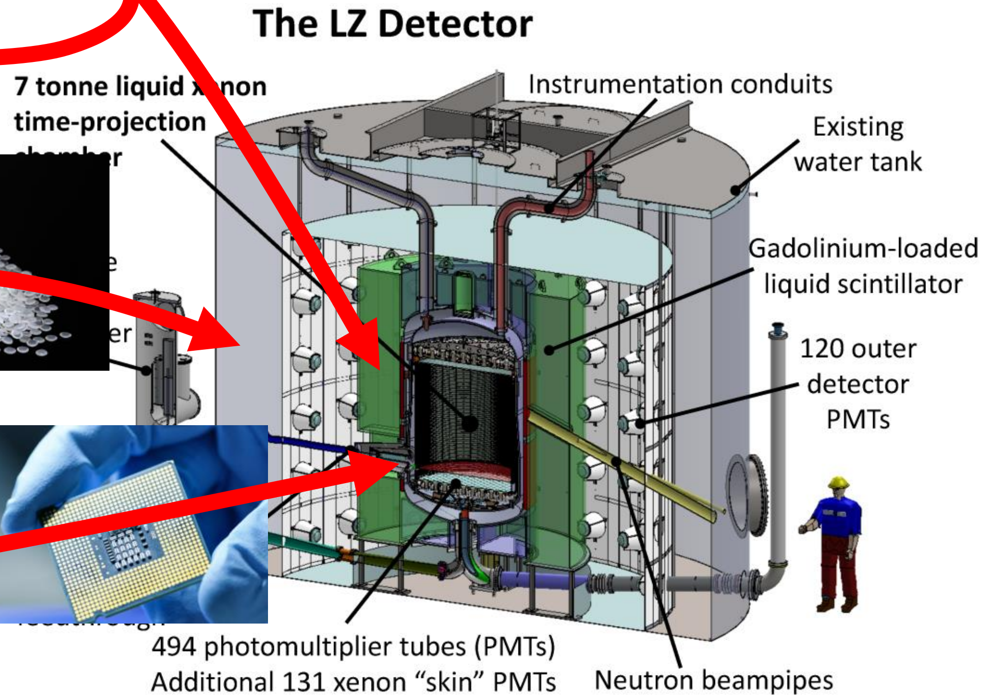
^{238}U and ^{232}Th and their progeny



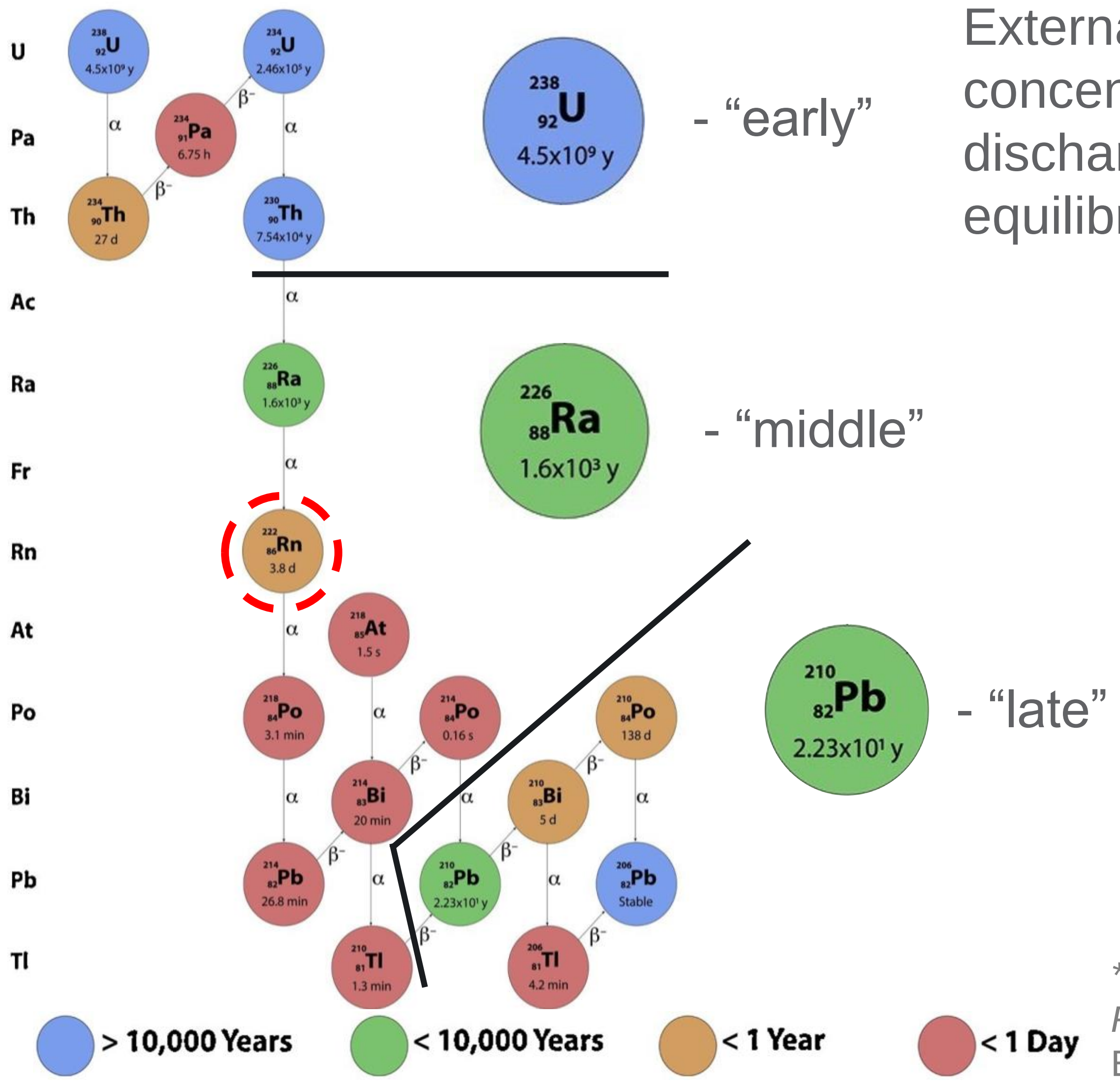


Sec equilibrium, *sans* radon

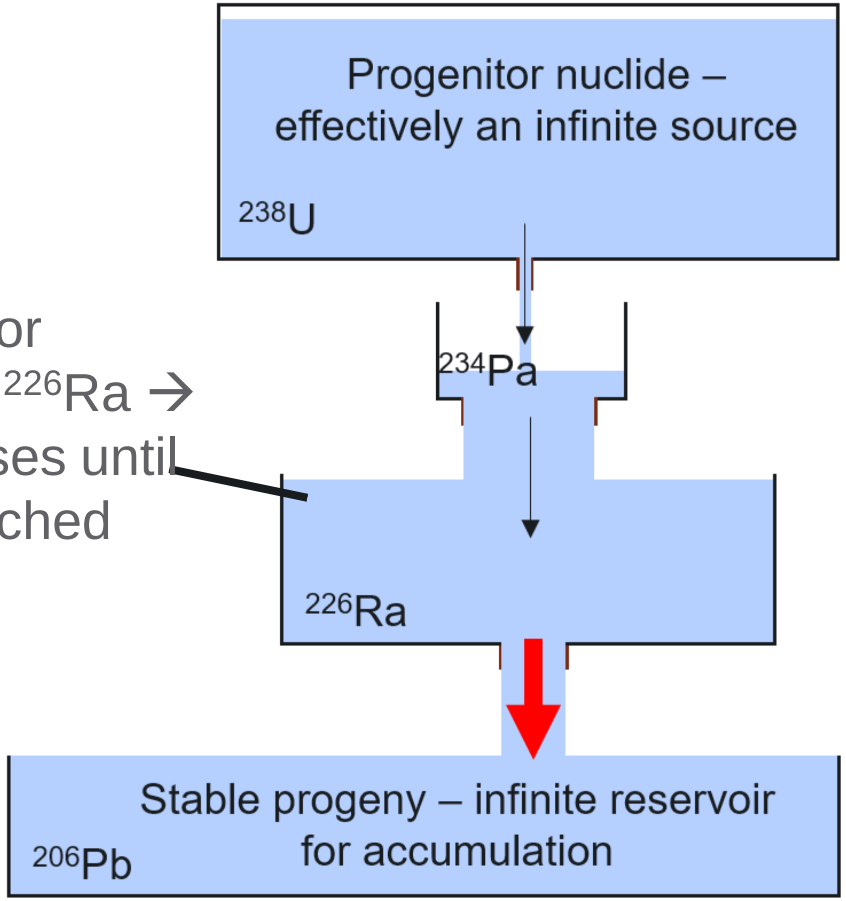


Sec equilibrium????

Secular Equilibrium



External source (or concentration) of ^{226}Ra → discharge increases until equilibrium is reached



- Secular equilibrium – discharge from each bucket is equal
- Short half-life = large hole
- Long half-life = small hole

L. Claiborne, C. Miller, (2012) Teaching Radioactive Decay and Radiometric Dating: An Analog Activity Based on Fluid Dynamics,* J. Geosci Educ. **60 (2), 114-122 (2012).

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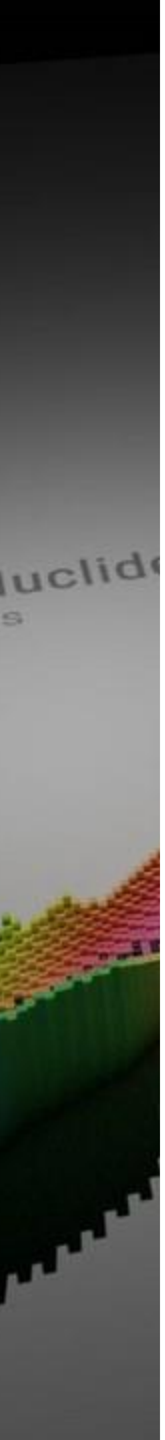
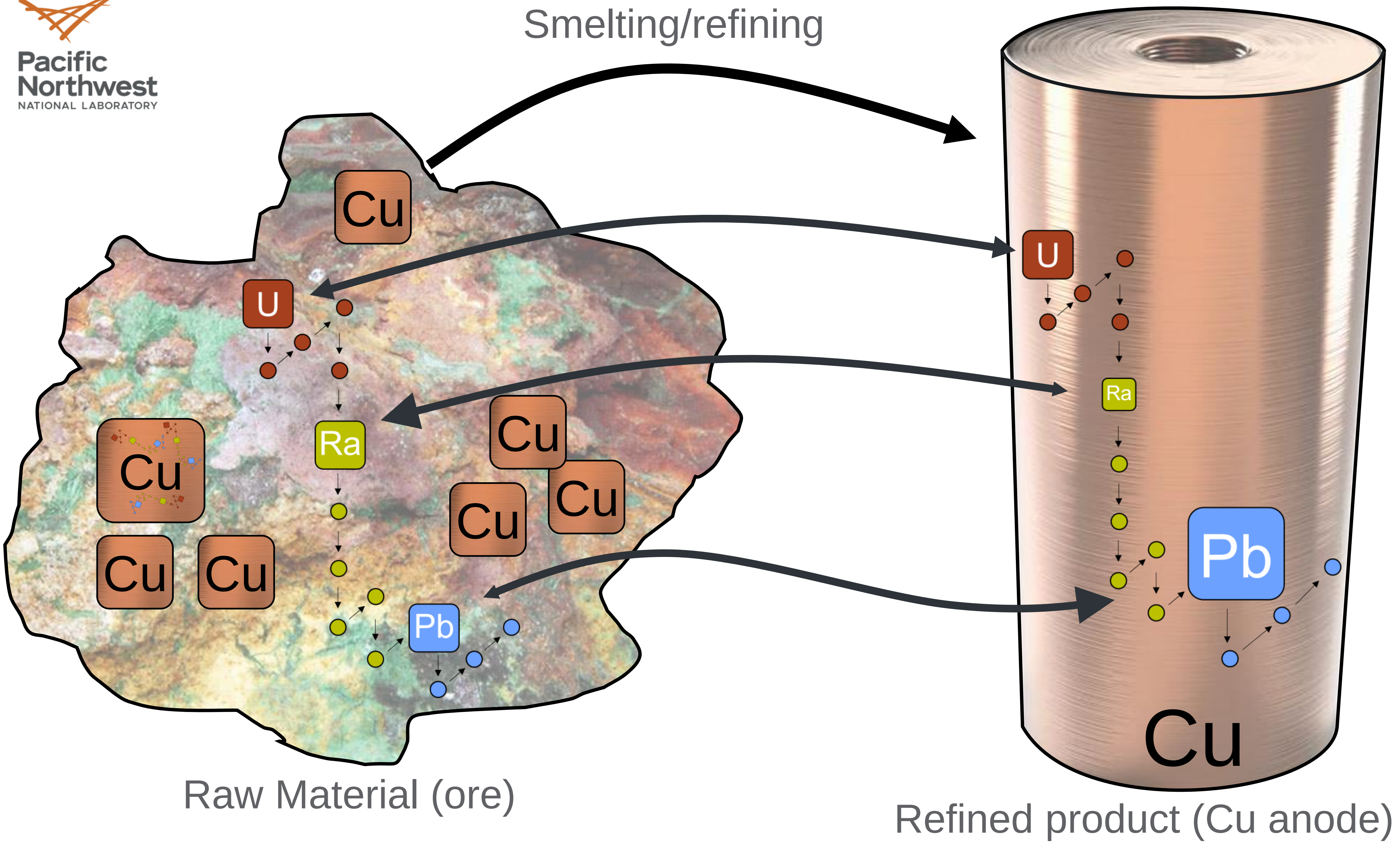


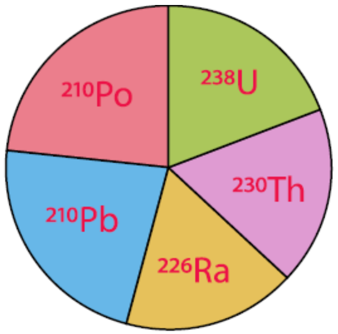


Motivation

Raw material/ore

Grinding
crushing
flotation

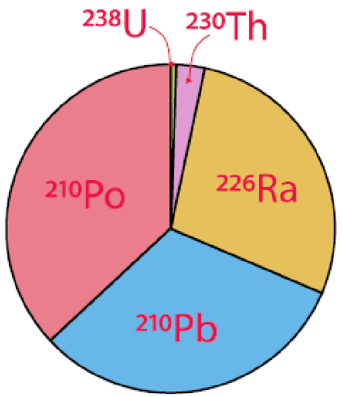
Cu-rich concentrate



% RN Bq/g

Sulfuric acid leach

Leached Cu-rich residue



% RN Bq/g

Smelting
refining

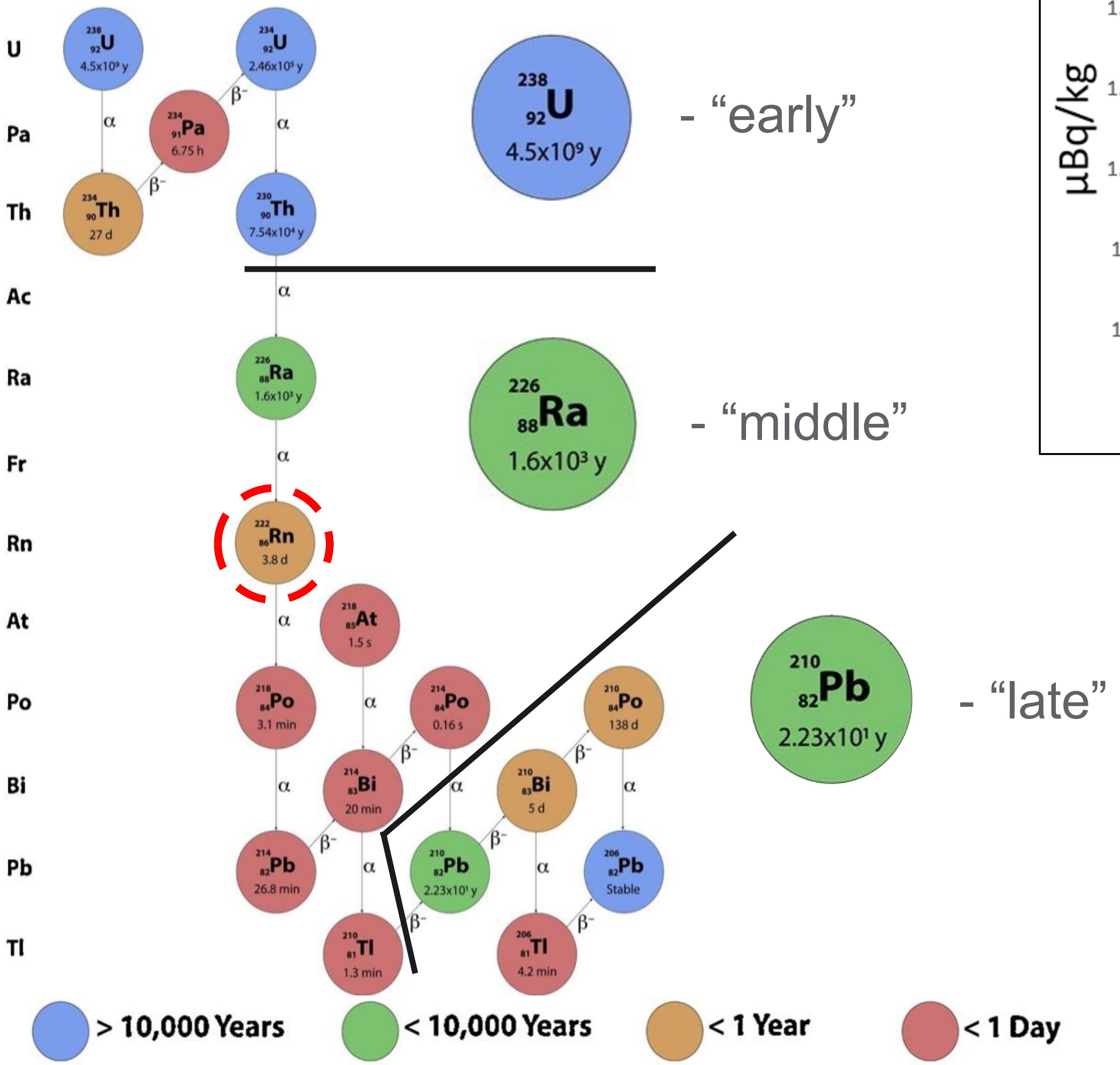
Cu metal

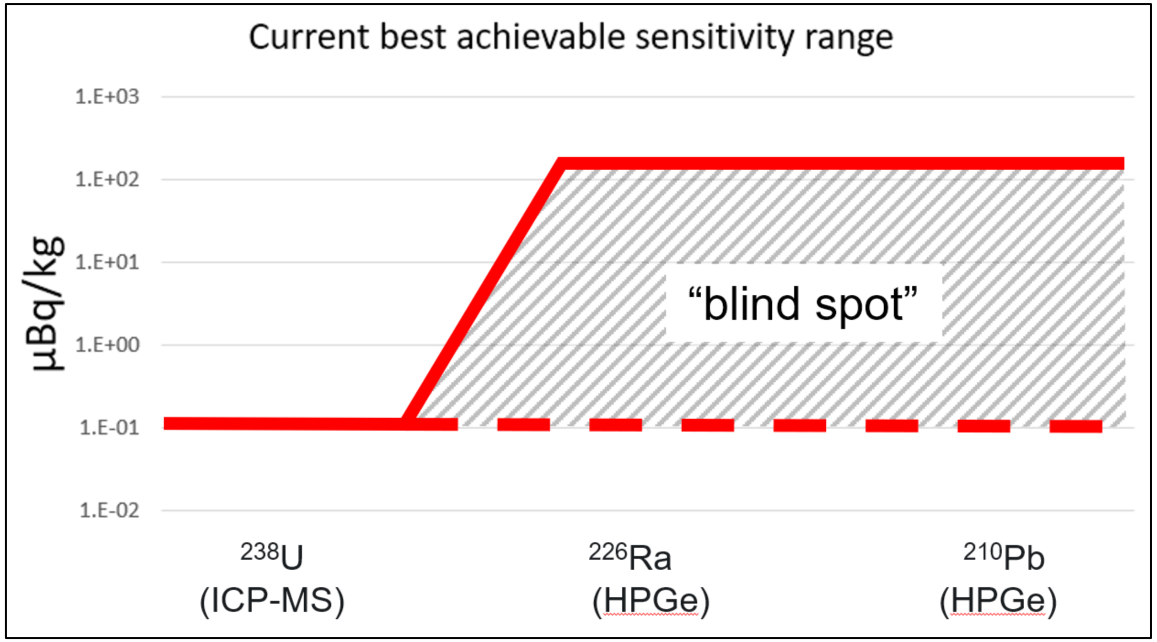
Secular disequilibrium has been observed in starting material for copper concentrates from the Olympic Dam mine (fourth largest copper mine)

- Would ***further*** processing “push” the ^{238}U decay chain back into equilibrium?
- Evolution over different processes (e.g., smelting, refinement, electrochemical deposition, etc.)

** Adapted from R. Ram et al. 2021.*

Challenges in Background Determinations



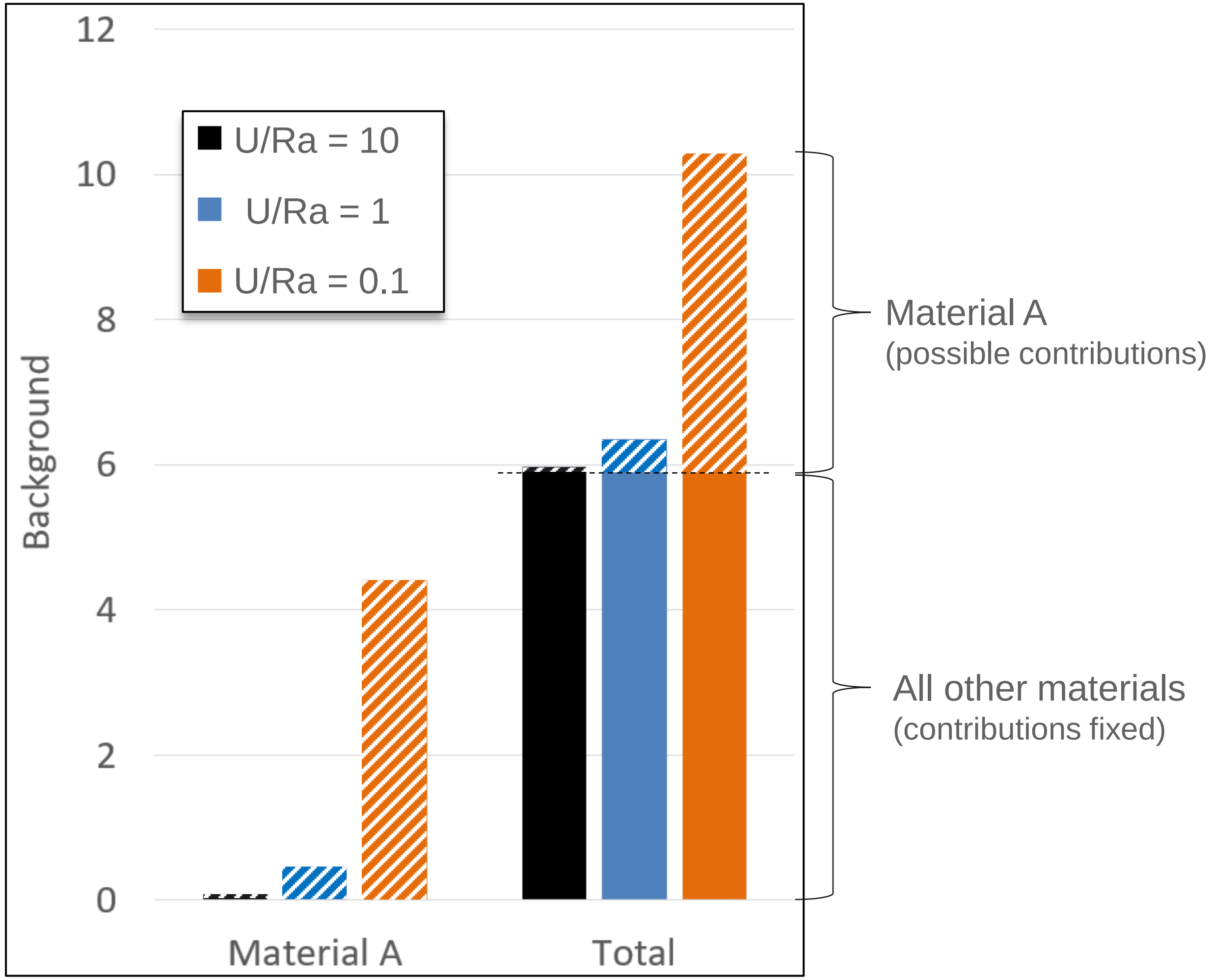


- ICP-MS and NAA assays measure ^{238}U at **nano**Bq/kg levels
- HPGe is not limited to the upper chain, but can reach sensitivities to about **micro**Bq/kg (large sample mass)
- Understanding backgrounds in ULB experiments is extremely important, secular disequilibria in materials can lead to inaccurate background predictions

Potential Impact on ULB Detectors

- Example of how secular equilibrium could affect background of a detector
  - Material A normally contributes 10% towards background when in equilibrium (U = Ra)
  - If U<Ra by 10x, Material A contributes upwards of 50% towards background
  - Small part + small fractionation = not a big deal
  - Small part + large fractionation = problem

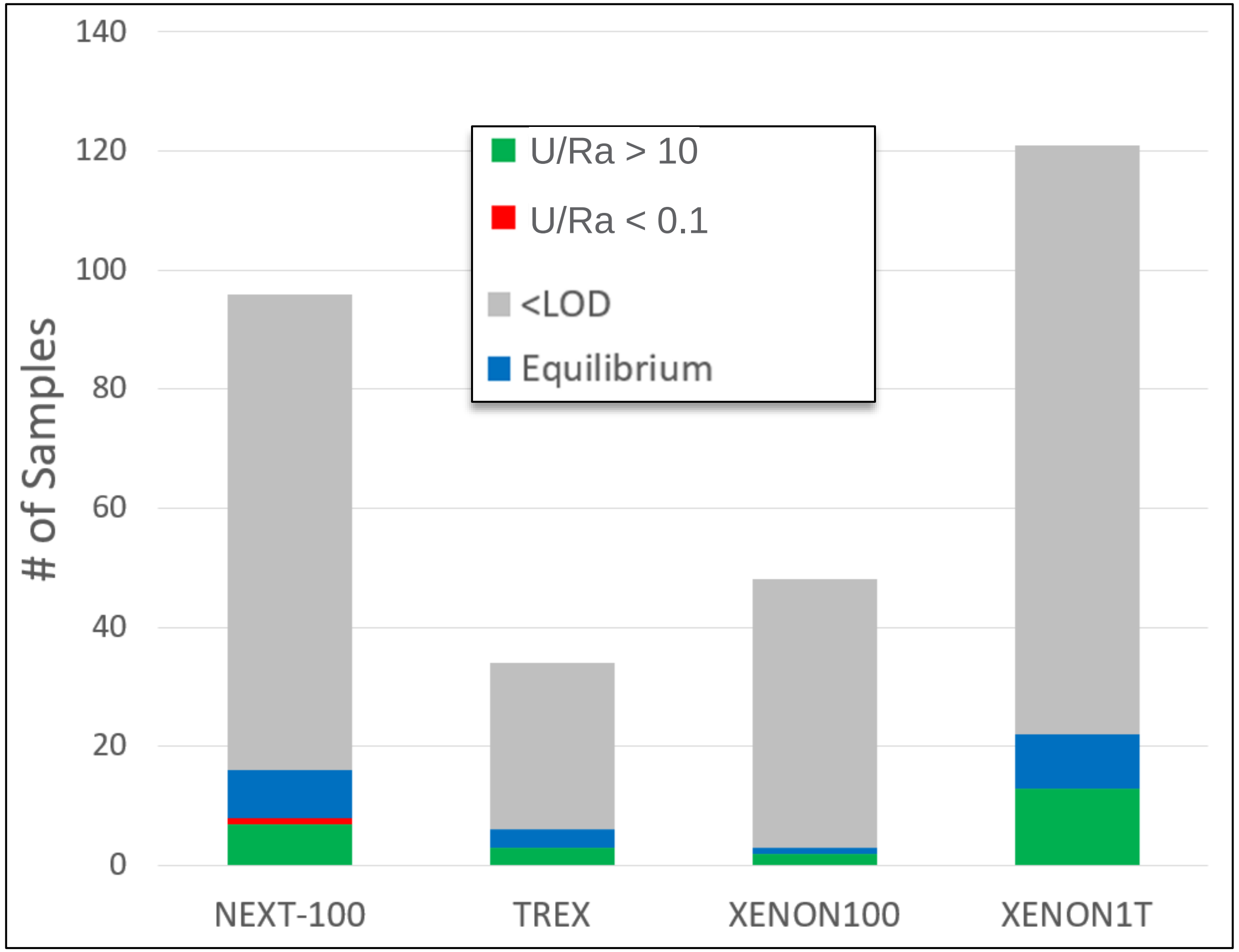
Detector X:



Secular Equilibrium in ULB Detectors

U/Ra fractionation:

- <LOD – secular equilibrium could not be determined due to instrumental detection limits or high measurement variability



Next generation detectors:

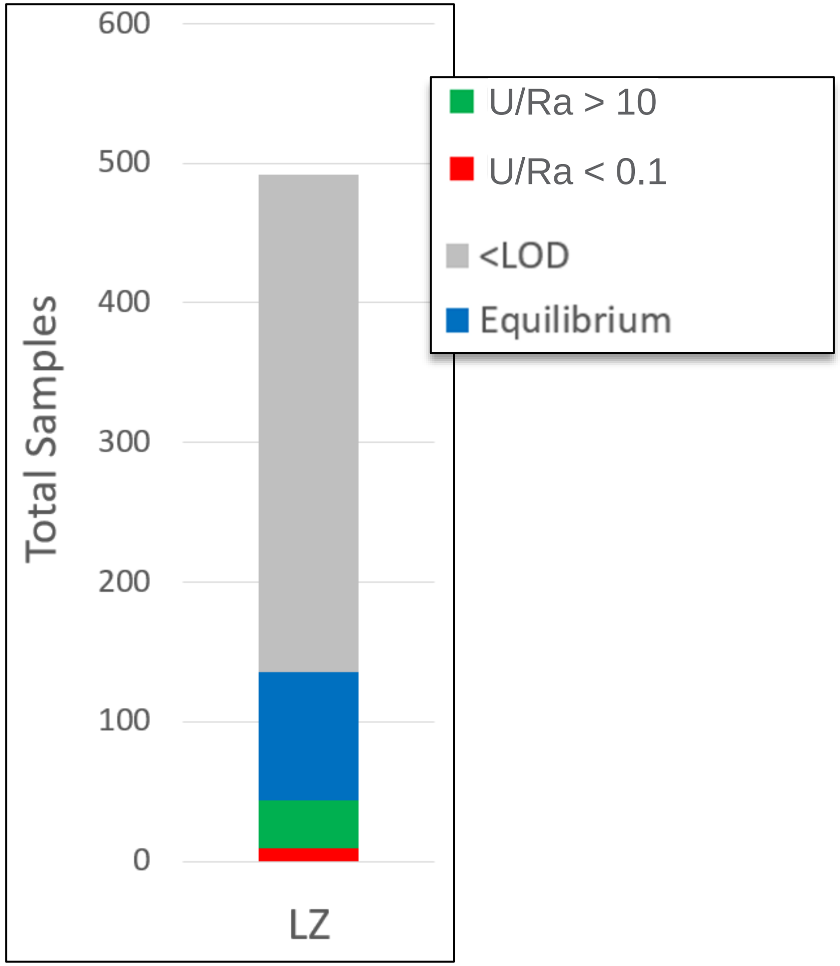
- increase in mass
- more stringent radiopurity requirements

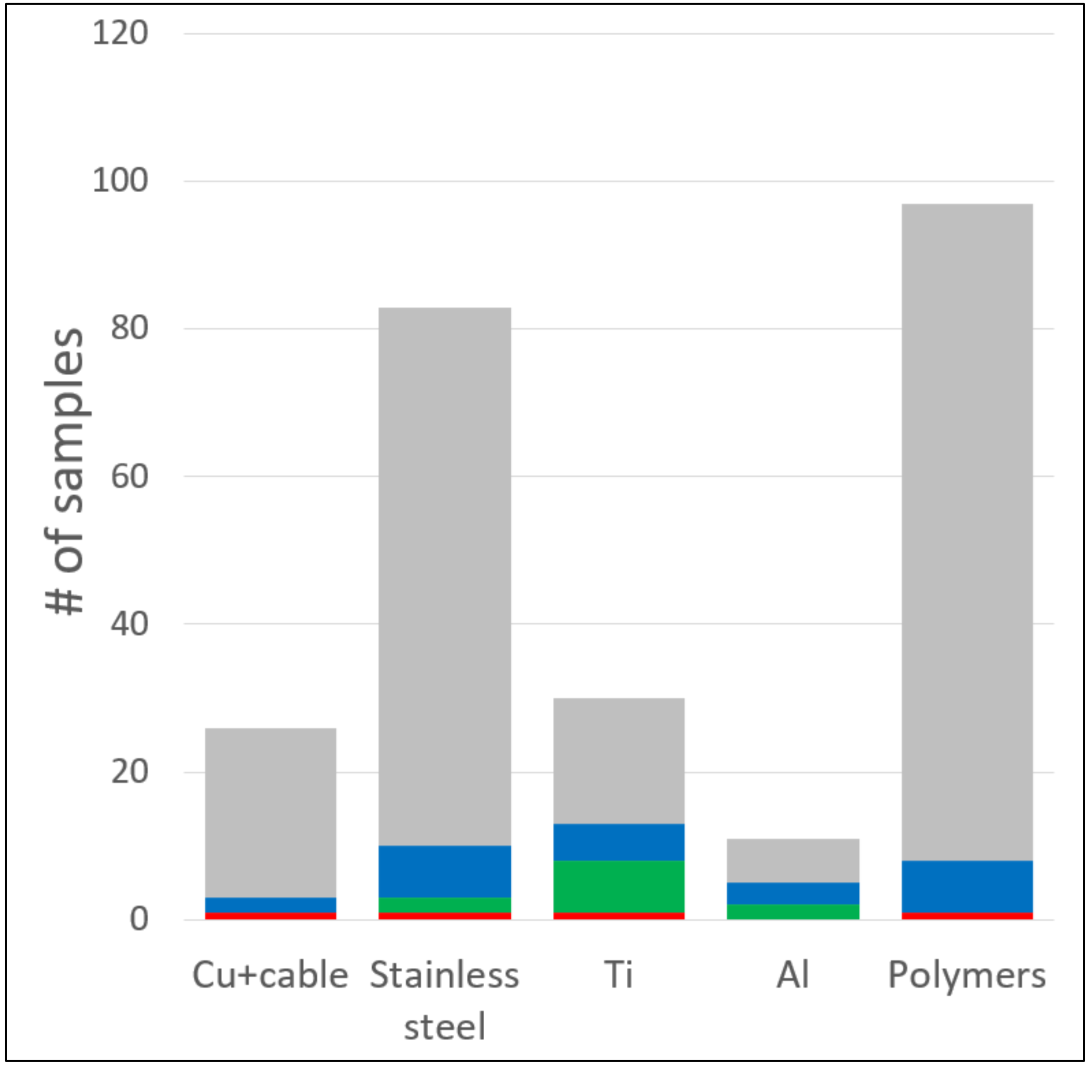
** Garnered from:*

V. Alvarez et al. 2013
E. Aprile et al. 2011
E. Aprile et al. 2017
J. Castel et al. 2019

LUX-Zeplin (LZ) Assay Data – U/Ra

U/Ra fractionation:

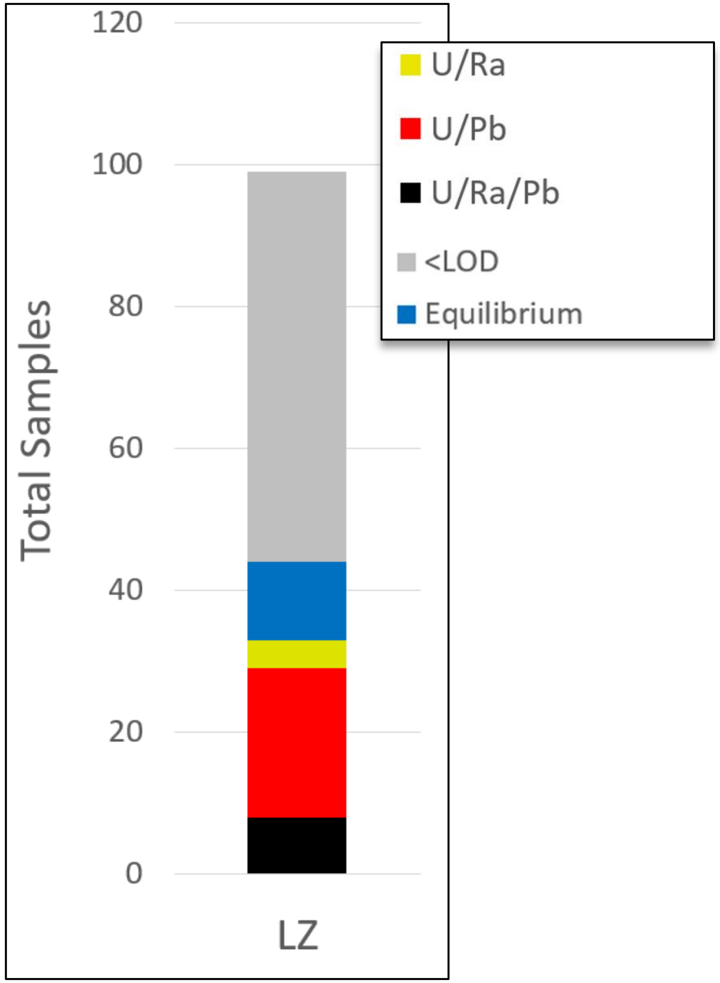




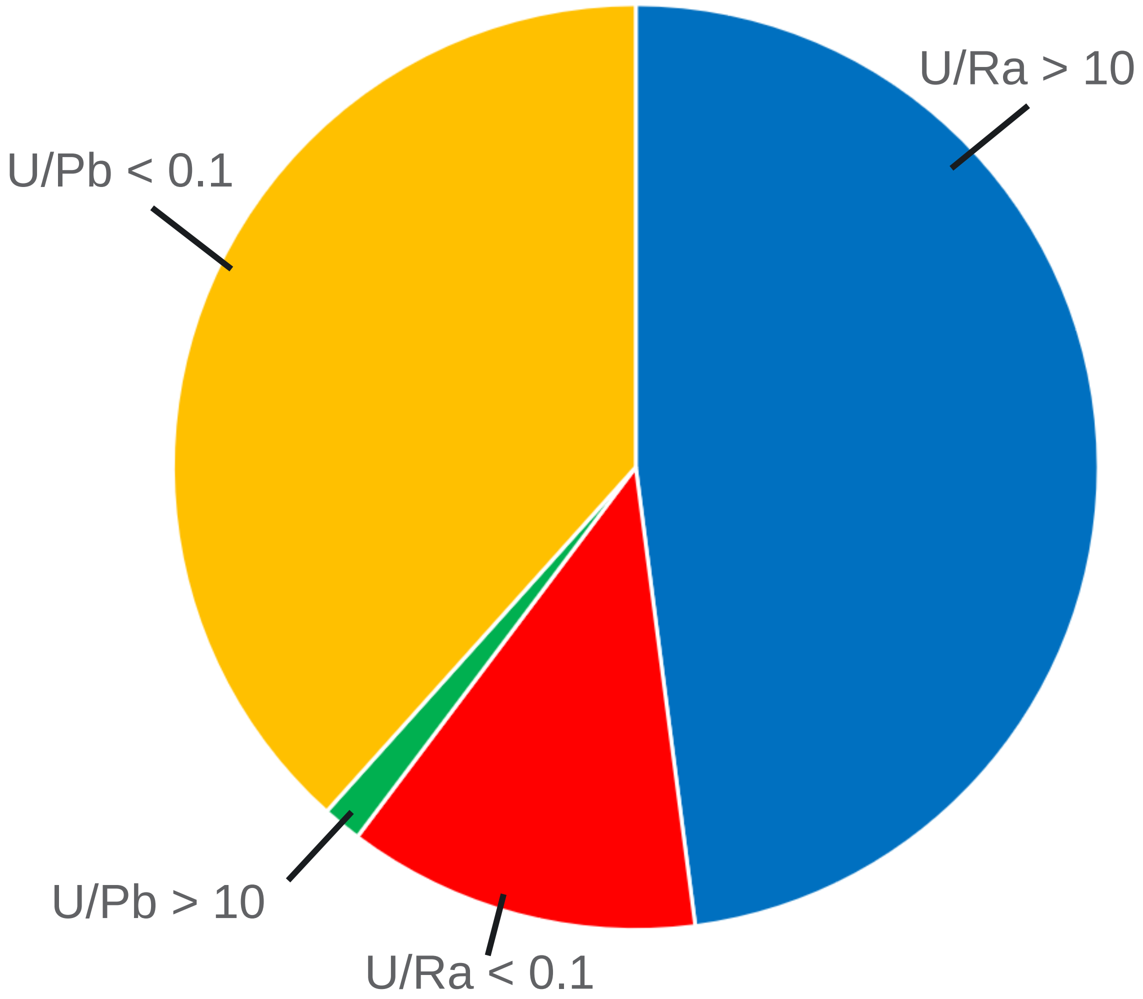
* *Garnered from* D.S. Akerib et al. 2020

LUX-Zeplin (LZ) Assay Data - ^{210}Pb

Samples with ^{210}Pb determinations:



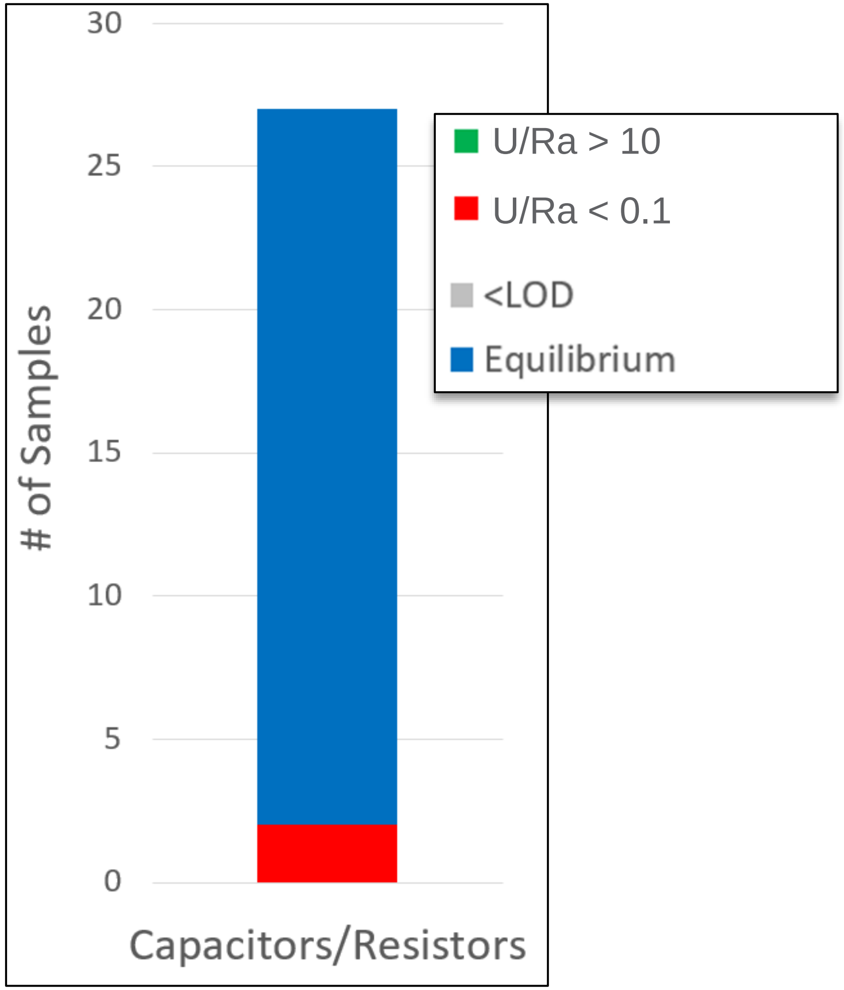
Type of disequilibrium:



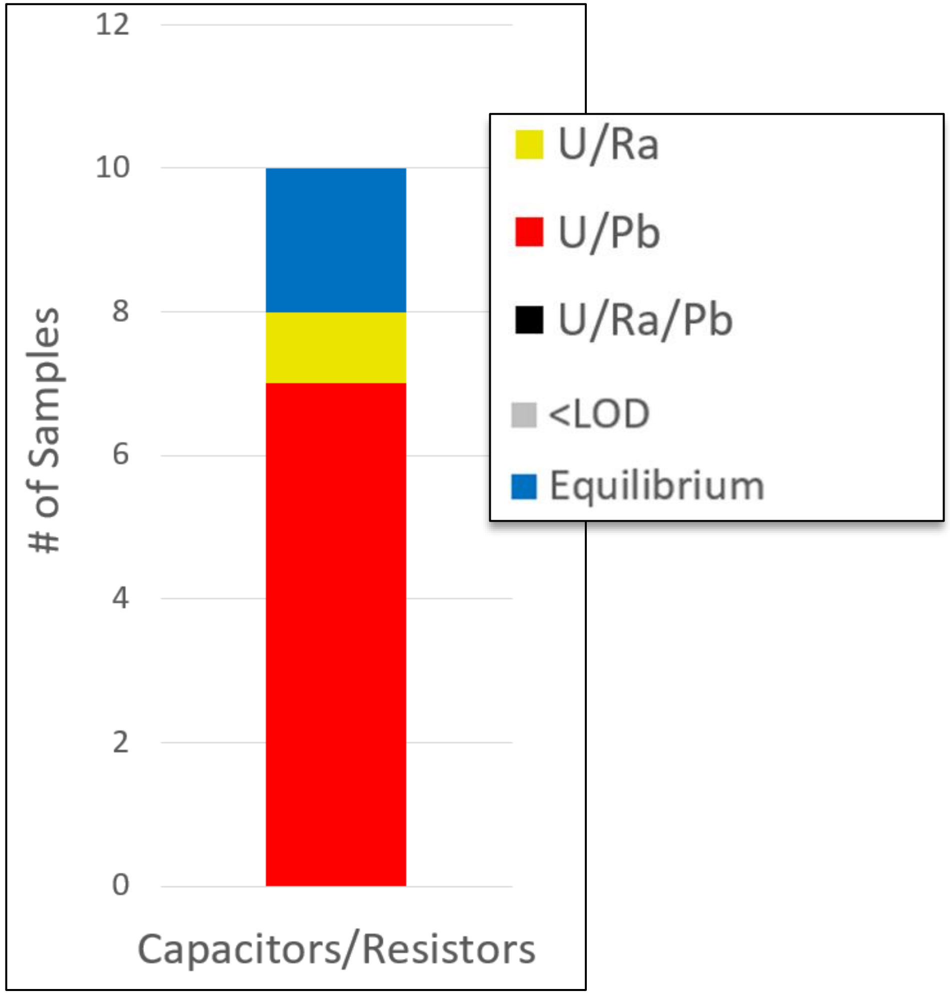
* *Garnered from* D.S. Akerib et al. 2020

LUX-Zeplin (LZ) Assay Data – Capacitors/Resistors

U/Ra fractionation:



Samples w/ ^{210}Pb determinations:



* *Garnered from* D.S. Akerib et al. 2020

Understanding the Problem – Better Assay

Use our automated separation method* to develop new analytical methods (measured using ICP-MS and other methods) with increased sensitivity in ^{238}U progeny assay

2DX prep*FAST* Station

Automated separation system

How much copper is required, ideally, to attain fractions of μBq/kg sensitivities?

↻ 1 g
for ^{238}U

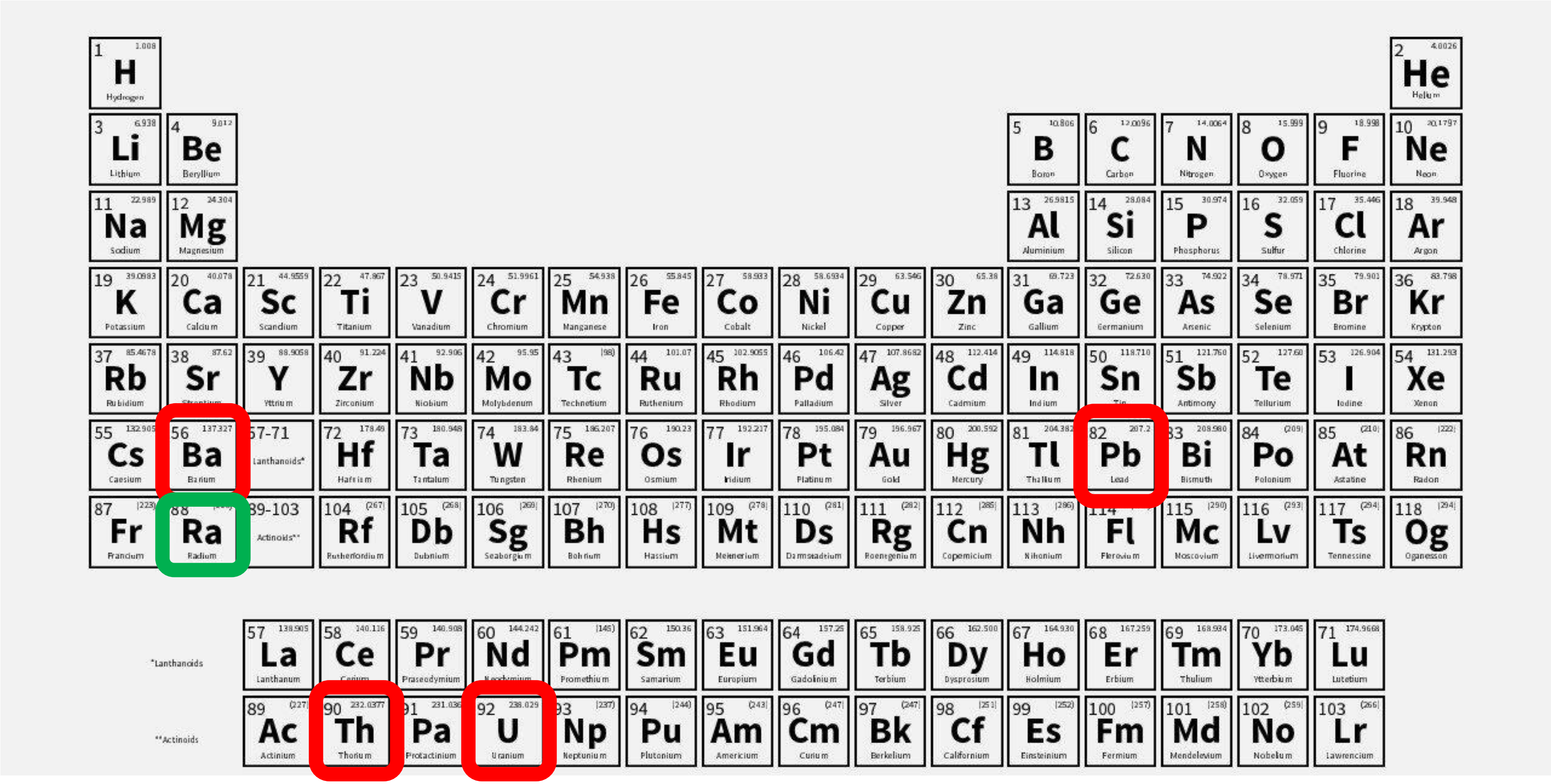
↻ 10000 g
for ^{226}Ra or ^{210}Pb

Increased sensitivity in decay chain progeny assay would allow assessment of secular disequilibria in HEP detector materials

* I.J. Arnquist et al. 2020

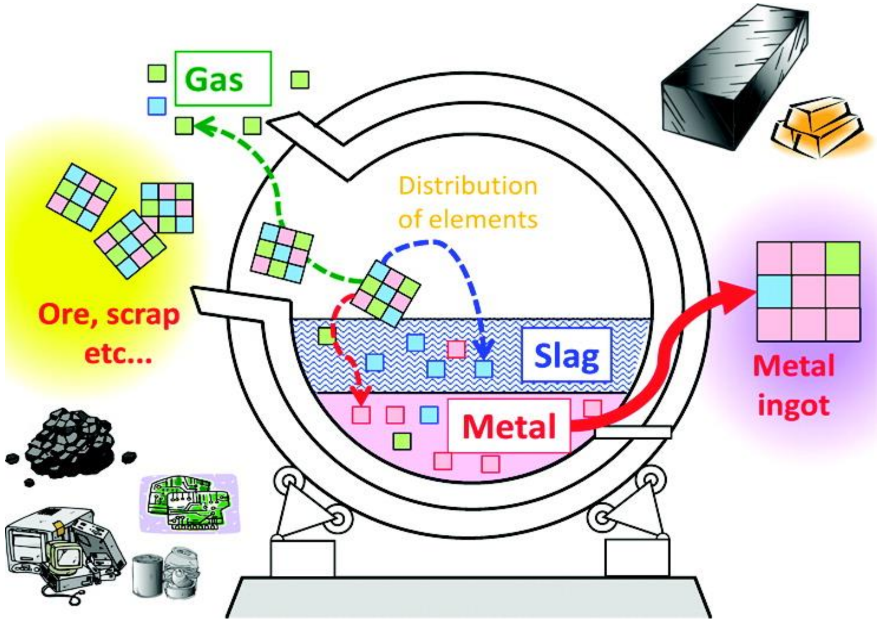
Approaches to Understand the Problem

- Spike samples with elements of interest (*e.g.*, U, Th, ^{stable}Pb, K, etc) or other surrogate isotope for short-lived radioisotopes of interest (*e.g.*, for Ra using Ba, which is nearly chemically identical) to track these elements through critical manufacturing steps (*e.g.*, smelting, plating, deposition *etc.*).



Leveraging secular disequilibrium in materials

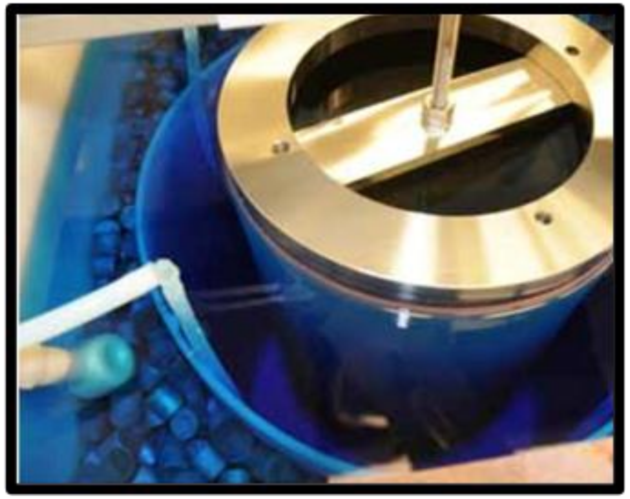
Investigate key production steps in electrochemical and metallurgical approaches to fractionate radioisotopes in materials



K. Nakajima et al., Thermodynamic Analysis for the Controllability of Elements in the Recycling Process of Metals. Environ. Sci. Technol. **45 (11)**, 4929-4936 (2011).

Metallurgical properties to leverage

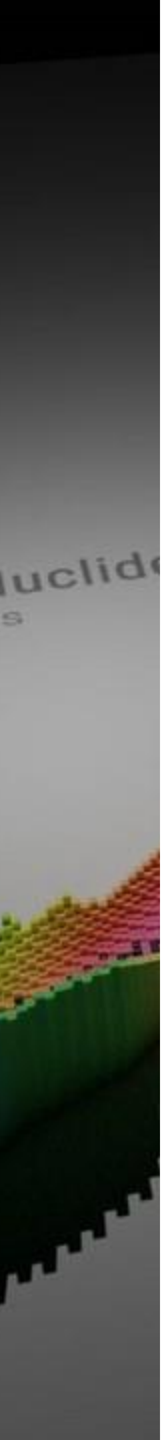
Element	T_m (°C)	T_b (°C)
Cu	1085	2571
Ti	1666	3289
U	1132	4172
Th	1750	4789
Ra	700	1737
Ba	729	1897
Pb	328	1750
K	64	765



Properties to leverage in electrochemistry

Element	Ionic Species	Standard Reduction Potential (V)	Ionic Radius (pm)
Cu	Cu^{2+}	+0.34	57-73
U	UO_2^{2+}	-1.44	~350
Th	Th^{4+}	-1.90	94-121
Ra	Ra^{2+}	-2.8	148-170
Ba	Ba^{2+}	-2.91	135-161
Pb	Pb^{2+}	-0.13	120-150
K	K^{+}	-2.93	137-164

Leverage chemical properties to develop methods to favorably fractionate the decay progeny in materials (^{238}U >> ^{226}Ra and ^{210}Pb)



Summary

- ICP-MS and NAA assays measure ^{238}U at nanoBq/kg levels. However, estimating backgrounds from ^{238}U can lead to over- or under-estimated background contributions if materials are not in secular equilibrium.
- HPGe assay does not suffer from this problem but is limited to about microBq/kg sensitivities.
  - Many experiments require materials (*e.g.,* copper, electronics components, *etc.*) in nanoBq/kg range → creates blindspots between ICP-MS/NAA and Ge
- This can be especially pertinent to materials (e.g., copper, polymers) used in high volume because, while very radiopure, they can make up the largest background contribution (*e.g.*, nEXO, SCDMS, *etc*.).
- Secular disequilibrium in the ^{238}U series decay chain has been measured in many different materials across multiple detectors
- Several approaches are available to investigate procedures that promote ***favorable*** secular disequilibria (^{210}Pb, ^{226}Ra << ^{238}U) and leverage these procedures to reduce backgrounds in ULB detectors

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Thank you

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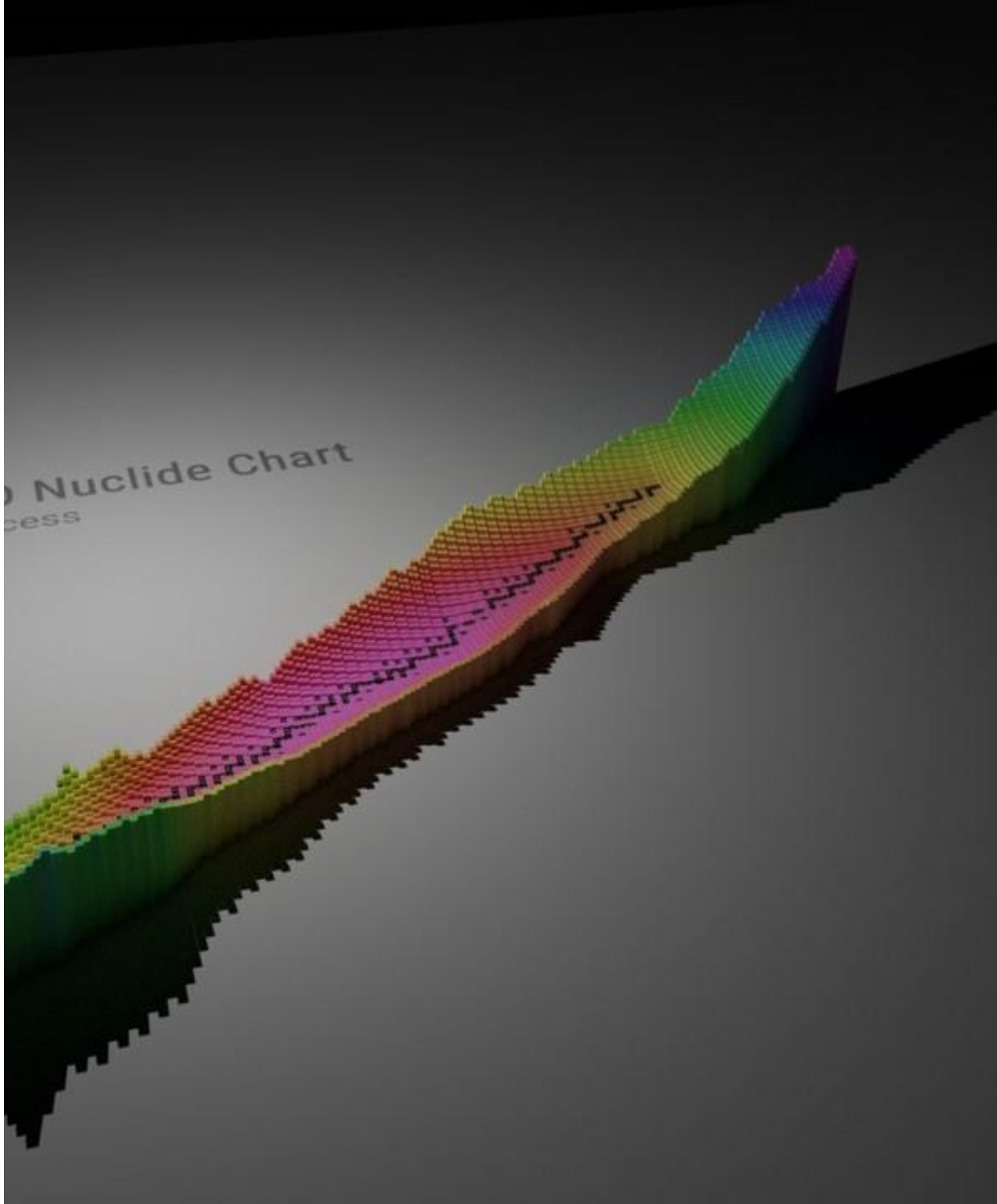

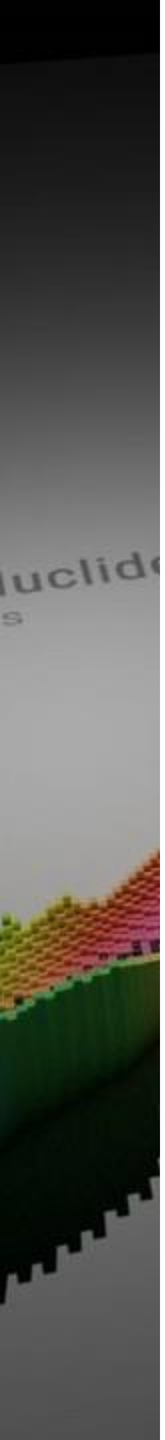

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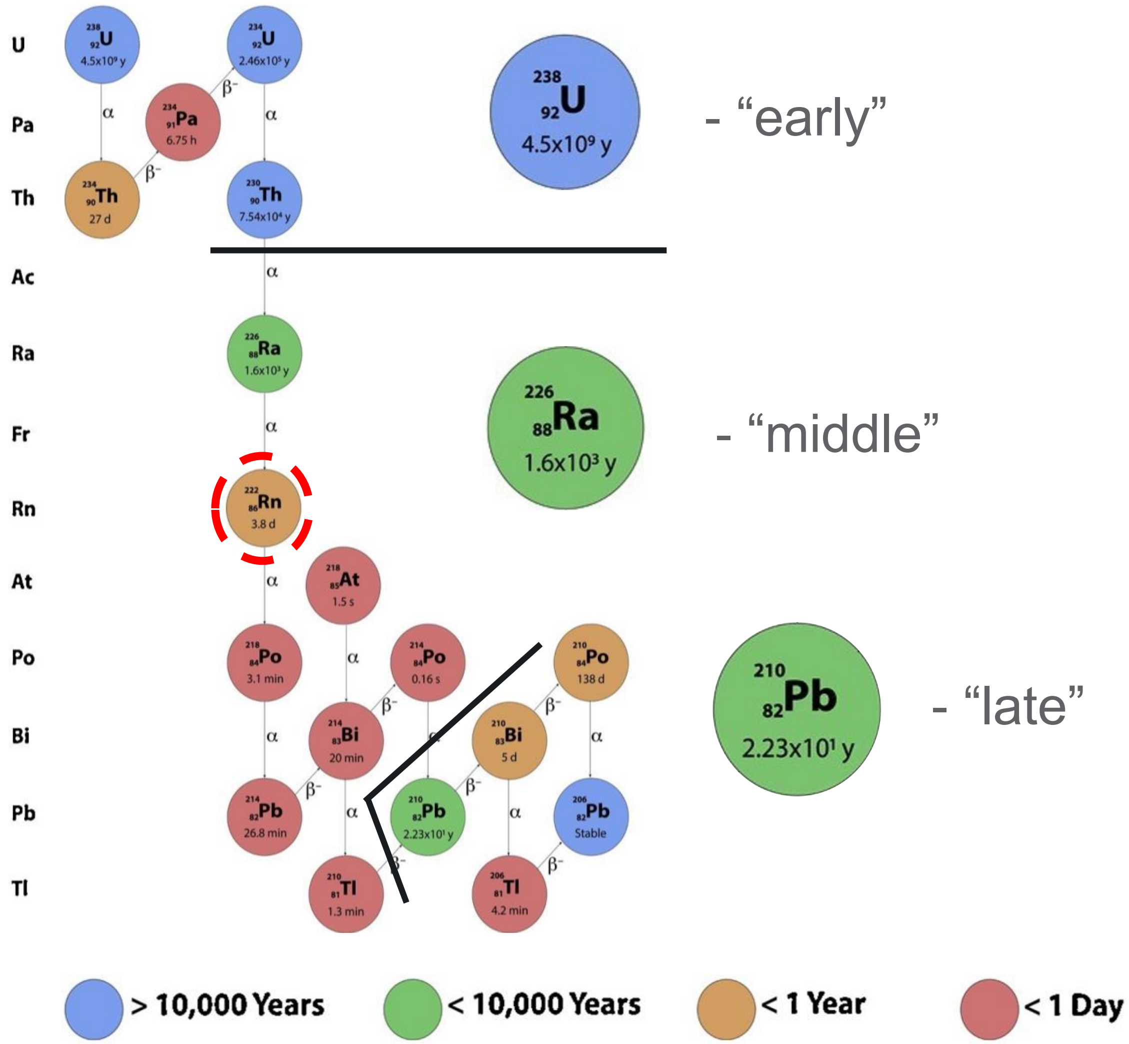
Decay Chains

^{238}U decay chain



^{232}Th decay chain