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Computational Age Dating of Special Nuclear Materials

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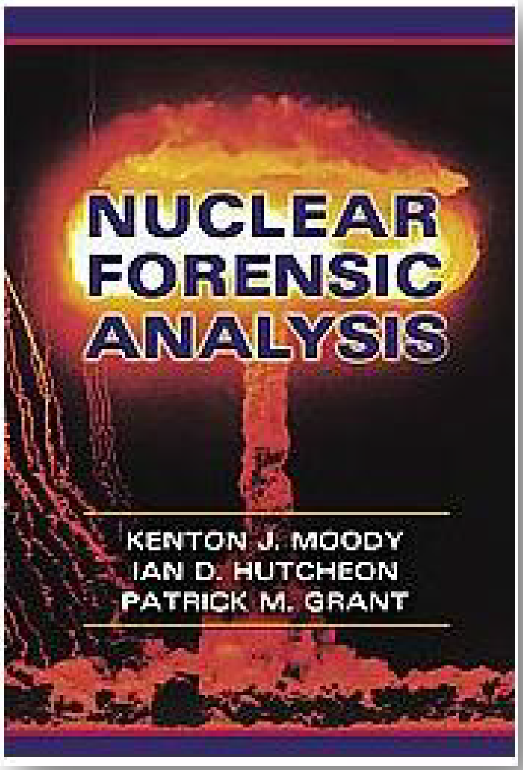
Outline

- Significance of SNM age dating for nuclear forensics
- Radioactive decay and age dating
- Bateman equations, solutions, and algorithms
- Current SNM age dating methods
- Constrained Progressive Reversal (CPR) method
- SNM age dating examples
- SNM mixing experiments
- Summary and conclusions

Significance of SNM Age Dating for Nuclear Forensics

- Scientific Applications
  - Chronometry
  - Material aging
  - Safeguarding
- Forensics Applications
  - Origin identification
  - Tampering / spoofing detection
  - Trafficking route detection
- Moody, Hutcheon, and Grant
  - Many uses and methods described
  - Ch. Chronometry, *Nuclear Forensics Analysis*, CRC Press, pp. 207–240



Radioactive Decay

- Radioactive Decay – A process by which an unstable (or radioactive) nuclide is transformed into a more stable nuclide
- Decay is accompanied by emissions of various forms of energy
- Radioactive decays have three main decay modes: Alpha, Beta, and Gamma
- One parent may have different decay children
- One child may have different parents
- For SNM age dating, we are mostly interested in four plutonium decay chains:
  - Pu-236 / 240 (4n or Thorium series)
  - Pu-237 / 241 (4n+1 or Neptunium series)
  - Pu-238 / 242 (4n+2 or Uranium series)
  - Pu-239 (4n+3 or Actinium series)
- All SNM age dating methods are based on the decay laws, or the Bateman equations that describe the full decay dynamics

Generalized Bateman Equations

- Radioactive decay is governed by a system of ordinary differential equations (SODE), known as Bateman equations

$$\frac{dN_i(t)}{dt} = -\lambda_i N_i + \sum_{j=1, i \neq j}^{m} \rho_{j,i} \lambda_j N_j$$

$$N_i(0) = N_{i,0} \qquad \text{for } i = 1,2,\ldots m$$

where

λ_i = decay constant for nuclide i

$\rho_{i,j}$ = branching factor from nuclide i to nuclide j

$N_i(t)$ = number of atoms of nuclide i in the system at time t

$N_i(0)$ = number of atoms of nuclide i in the system at time 0

or in matrix form

$$\begin{cases} \dfrac{d\mathbf{N}(t)}{dt} = \mathbf{A}\mathbf{N}(t) \\ \mathbf{N}(0) = \mathbf{N}_0 \end{cases}$$

$$\mathbf{A} = \begin{bmatrix} -\lambda_1 & \lambda_2 \rho_{2,1} & \cdots & \cdots & \lambda_m \rho_{m,1} \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ \lambda_1 \rho_{1,i} & \lambda_2 \rho_{2,i} & \cdots & \cdots & \lambda_m \rho_{m,i} \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ \lambda_1 \rho_{1,m} & \lambda_2 \rho_{2,m} & \cdots & \cdots & -\lambda_m \end{bmatrix}$$

where

$$\mathbf{N}(t) = \{N_i(t)\}_{i=1}^{m}$$

$$\mathbf{N}_0 = \{N_i(0)\}_{i=1}^{m}$$

Conventional SNM Age Dating Methods

- Mostly described in Moody, Hutcheon and Grant, *Nuclear Forensic Analysis*, CRC Press, Boca Raton, 2005), including:
  - Rate of depletion / in-growth
    - Can be used to track nuclides with known original concentration
    - Good for measuring 'pure' single-parent SNM
  - Ratio methods
    - Can be used to look at both depleted or accumulated nuclides
    - Ideal case: end product stable and detectable
    - Either original parent concentration or original child concentration must be assumed
  - Secular equilibrium methods
    - Looking at relationship between long-lived parent and short-lived child
    - Assuming the age of the material is much greater (usually 7X) than the half-life of the child product
    - Estimation uncertainty is usually 7X of the half-life of the child nuclides
- These methods need to track the decay dynamics of at most two nuclides
- We will introduce a Constrained Progressive Reversal method (CPR)

Constrained Progressive Reversal Method (I)

- The Constrained Progressive Reversal method is based on a linear transformation method for computing Bateman solutions (Yuan, 2010, *AJP*):

$$\mathbf{N}(t) = \mathbf{F}(t)\mathbf{N}(0)$$

where $\mathbf{F}(t) = e^{t\mathbf{V}} = \left\{ f_{i,i}(t) \right\}$ is a time dependent matrix and unique for each chain (4 chains of importance for SNM age dating).

For discrete decay calculation, we can use formulas:

$$\mathbf{N}(n\Delta t) = \mathbf{F}^n(\Delta t)\mathbf{N}(0)$$

$$\mathbf{N}(\text{day } n) = \mathbf{F}^n(1\text{ day})\mathbf{N}(\text{day } 0) \quad n = 1,2,\ldots$$

- These are handy formulas for computing and plotting. It is interesting to observe the following similarities:

Cases	Rate of Change	States of the Chain
Single Nuclide	$F(1) = e^{-\lambda}$ (scalar)	$N(t) = F^t(1)N(0)$
Multiple Nuclide	$\mathbf{F}(1) = e^{\mathbf{V}}$ (matrix)	$\mathbf{N}(t) = \mathbf{F}^t(1)\mathbf{N}(0)$

$\mathbf{F}(1) \Rightarrow$ Characteristic matrix (or rate matrix) of a decay chain

Constrained Progressive Reversal Method (II)

- The 'rate' matrix has some extremely nice features:

$$\mathbf{F}(1) \quad \text{Can be precisely calculated} \quad \text{(Yuan, 2010, JAP)}$$

$$\mathbf{F}(0) = \mathbf{I} \quad \text{Identity matrix}$$

$$\mathbf{F}(-1) = \mathbf{F}^{-1}(1) \quad \text{Simple matrix inverse}$$

- Using these nice features of the 'rate' matrix, we can easily calculate any future state of the chain:

$$\text{Future State of the Chain}: \mathbf{N}(n) = \mathbf{F}(1)\mathbf{N}(n-1) = \ldots = \mathbf{F}^{n}(1)\mathbf{N}(0)$$

- Also we can calculate any past state of the chain (as long as the physical sample exist):

$$\text{Past State of the Chain}: \mathbf{N}(-n) = \mathbf{F}(-1)\mathbf{N}(-(n-1)) = \ldots = \mathbf{F}^{n}(-1)\mathbf{N}(0)$$

- For manual calculations, these quantities are still challenging
- But with computers, it is a simple task, since computers are excellent for performing simple repeated tasks
- Now we have a pretty good method to propagate any SNM sample in any forward or backward direction, but what about age determination?

Constrained Progressive Reversal Method (III)

- Recall that the Bateman solution is unconstrained; it can be applied either in forward or backward direction:

$$\mathbf{N}(t) = e^{Vt}\mathbf{N}(0)$$

- In the forward direction, it is not of much concern
- In the backward or reversal direction, there are certain constraints that would prevent us from 'rolling' back the decays forever:
  - All components must be non-negative
  - All components must be no larger than correspondent maximum numbers of atoms allowed per unit mass of the sample
  - If the decay chains do not involve alpha decay, the total number of atoms in the system should be roughly a constant (atomic conservation)
- These constraints translate into these numerical conditions:

$$N_i(t) \geq 0$$

$$N_i(t) \leq N_i^{\max} \qquad N_i^{\max} = \text{number of atoms per unit mass for nuclide } i$$

$$\sum_{i=1}^{m} N_i(t) \equiv \sum_{i=1}^{m} N_i(0) \;\; \text{practically, it should be} \sum_{i=1}^{m} N_i(t) \approx \sum_{i=1}^{m} N_i(0)$$

$$\text{where} \quad t \geq -t_{age}, \quad t_{age} \text{ is the age of the SNM}$$

Constrained Progressive Reversal Method (IV)

- **Constrained Progressive Reversal (CPR) Method** for dating SNM:
  (1) Construct a set of constraints (such as those discussed earlier, but certainly you can add more constraints as you like):
  $$\{C_l(\mathbf{N})\} = \{C_l(\mathbf{N}), \quad l = 1,2,..L\}$$
  (2) Compute the 'rate' matrix (per day):
  $$\mathbf{F}(-1\,\text{day}) = \mathbf{F}(-1)$$
  (3) Check if any constraint is violated: $\{C_l(\mathbf{N}(0))\}$
  If yes, then the material was perhaps freshly made.
  (4) If none of the constraints is violated, then compute:
  $$\mathbf{N}(-1) = \mathbf{F}(-1)\mathbf{N}(0)$$
  and check constraints: $\{C_l(\mathbf{N}(-1))\}$
  (5) If any constraint is violated; then the material was perhaps made yesterday.
  (6) If none of the constraints is violated, then compute:
  $$\mathbf{N}(-2) = \mathbf{F}(-1)\mathbf{N}(-1), \ldots\ldots, \mathbf{N}(-\mathrm{n}) = \mathbf{F}(-1)\mathbf{N}(-(\mathrm{n}-1))$$
- This process may be repeated iteratively, but should stop after a finite number of steps.
- Best of all, computers can do this very easily!

Constrained Progressive Reversal Method (V)

- One may notice that the CPR method may produce multiple 'candidate' age estimates, or a set of age estimates:

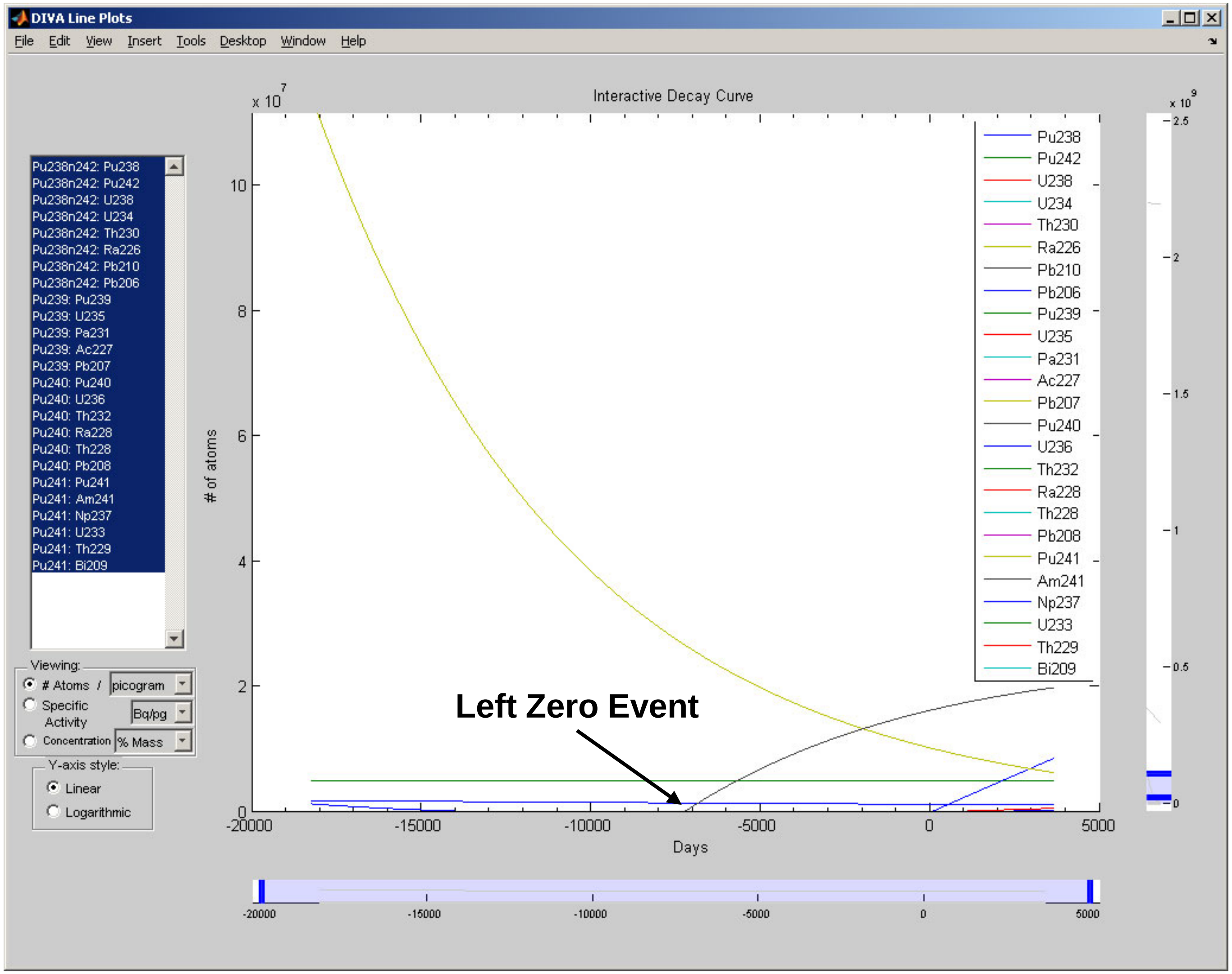
$$\Omega_{age} = \{t_1,\ldots,t_k\}$$

- If this set contains only one single estimate, then it is the age of the SNM.
- If this set contains multiple estimates, but they are within the range of error, then their average can be used as the age estimate. Of course, we can come up with the uncertainties of the estimate as well.
- If this set contains multiple estimates, and those estimates obviously differ significantly among themselves, then we have a reasonable doubt that the sample may have been contaminated, tampered with, or spoofed…

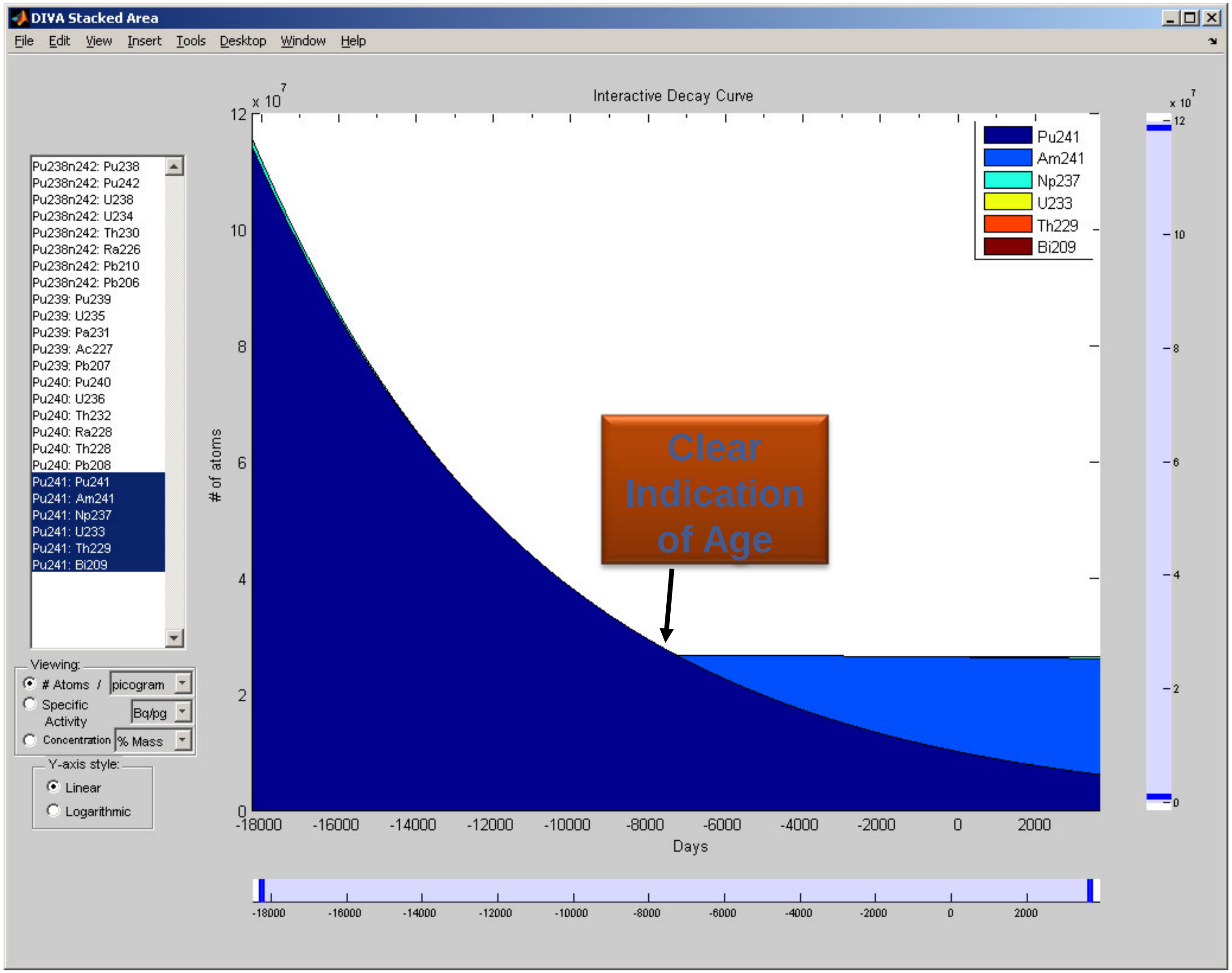
SNM CPR Age Dating Cases

#	Comments and Data Source	Nuclide	Quantity	Known Age (yrs)	CPR Age Est. (yrs)
#1	Measured in mass concentrations (S. Mukhopadhyay)	^{238}Pu	0.05%	19~20	19.9
		^{239}Pu	87.23%		
		^{240}Pu	12.09%		
		^{241}Pu	0.41%		
		^{242}Pu	0.20%		
		^{241}Am	6510 ppm		
#2	Measured in atomic abundances (R. Keegan)	^{241}Pu	4.69×10^{16}	18.97 ±1.0	18.91
		^{241}Am	6.90×10^{16}		
		^{233}Pa	4.13×10^{7}		
#3	Measured in atomic abundances (R. Keegan)	^{241}Pu	7.96×10^{17}	41.2 ±2.4	41.10
		^{241}Am	4.79×10^{18}		
		^{233}Pa	1.48×10^{10}		

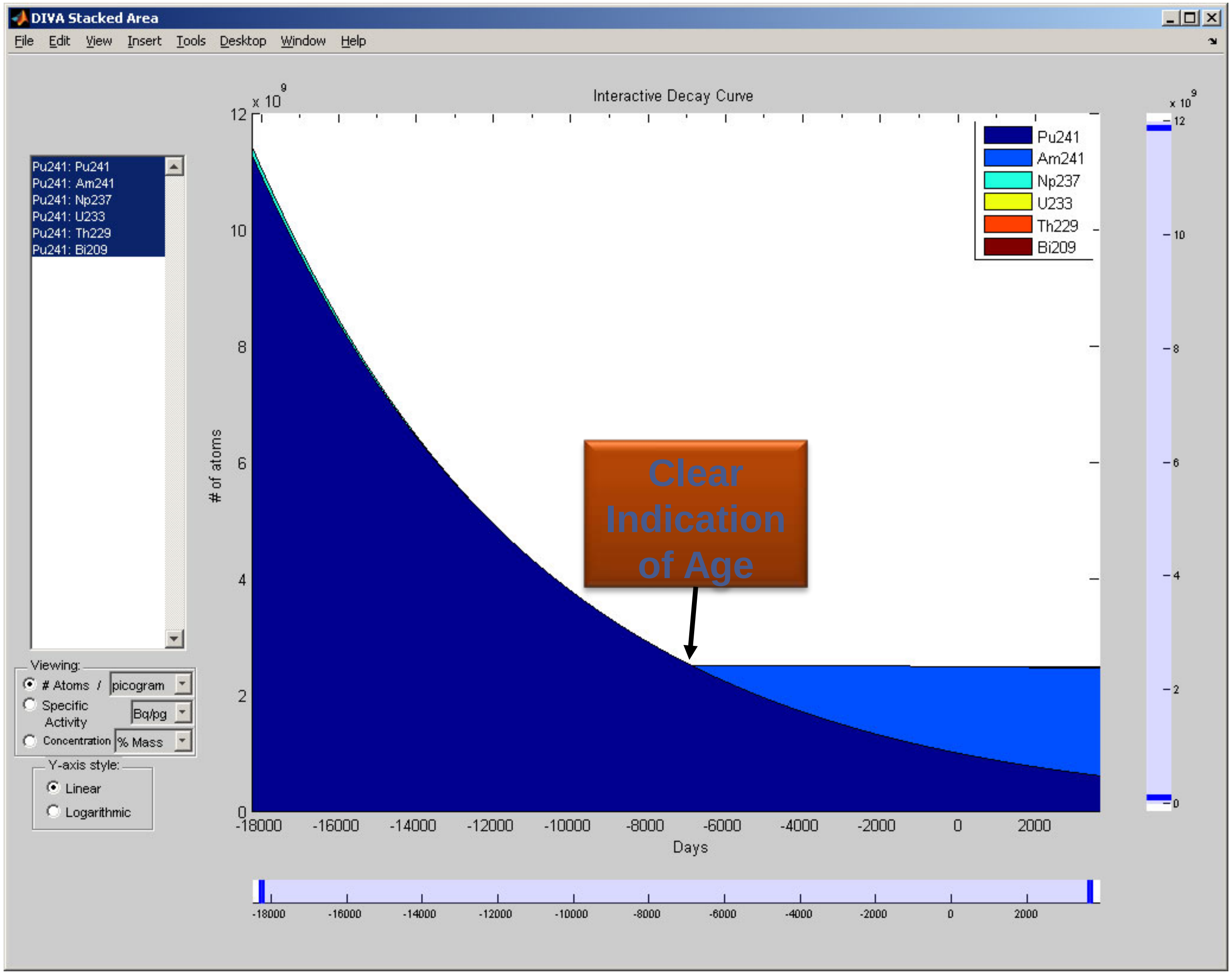
Sample 1: Line Plots



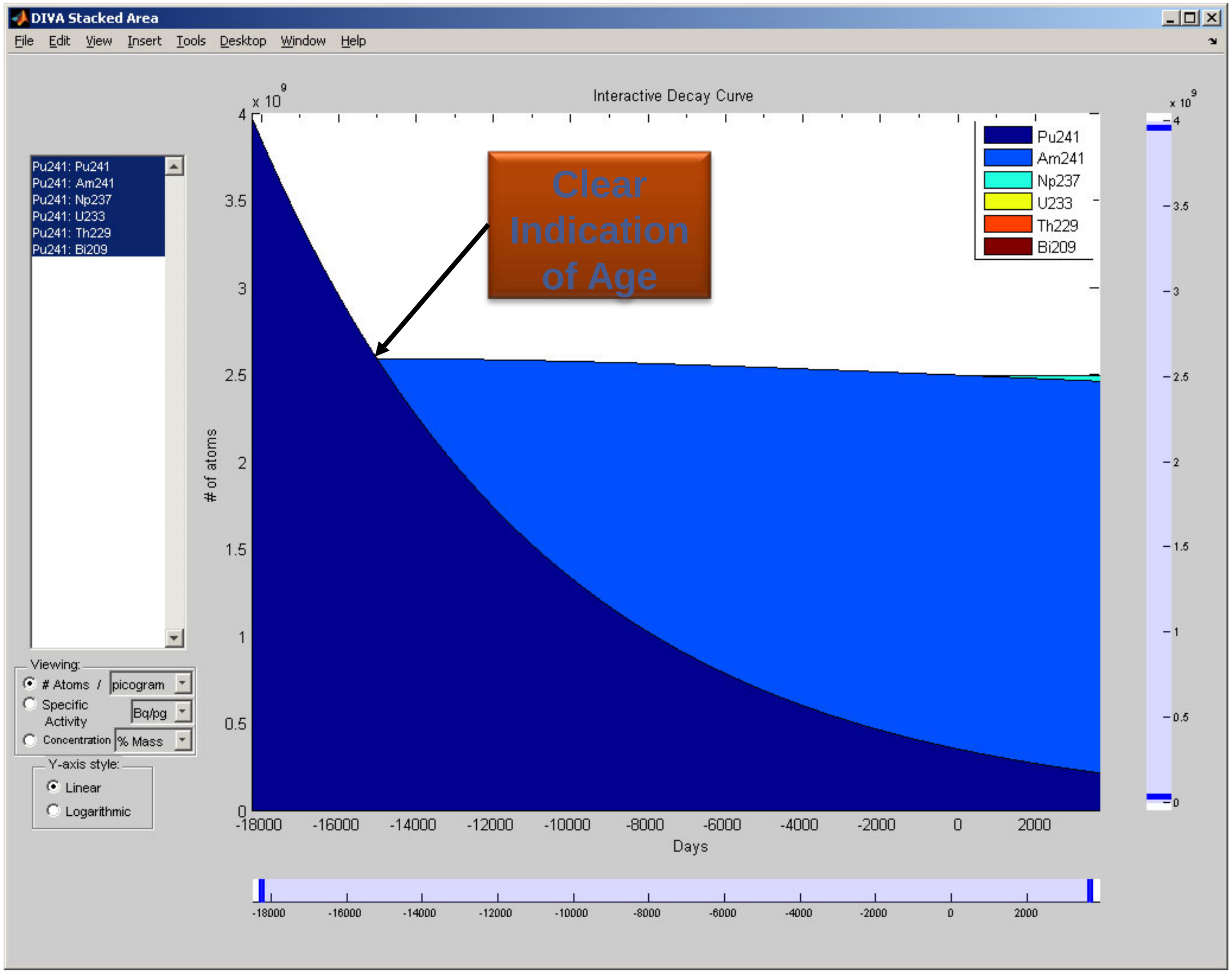
Sample 1: Stacked Area Plots (^{241}Pu Chain)



Sample 2: Stacked Area Plots



Sample 3: Stacked Area Plots

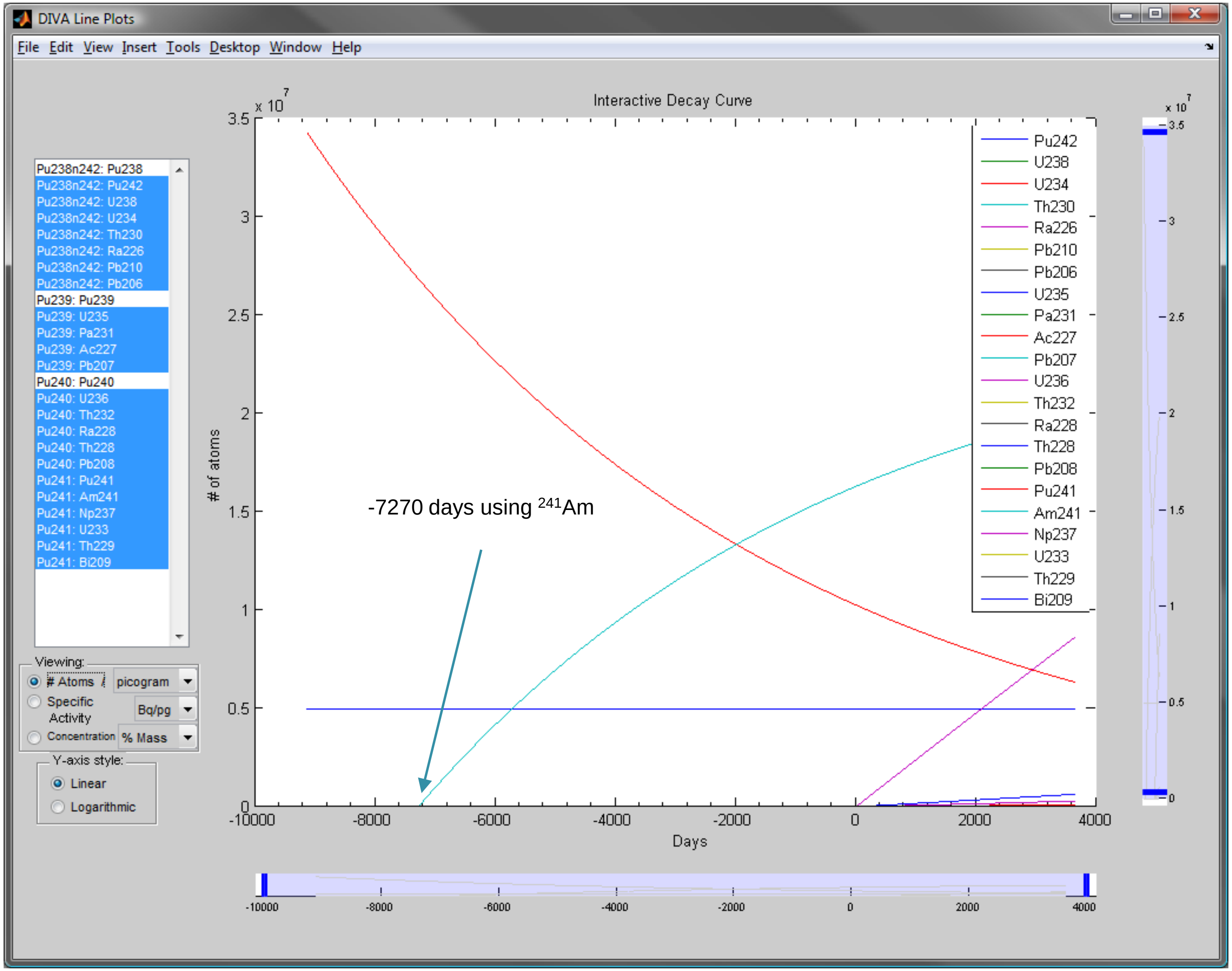


SNM CPR SNM Mixing Study

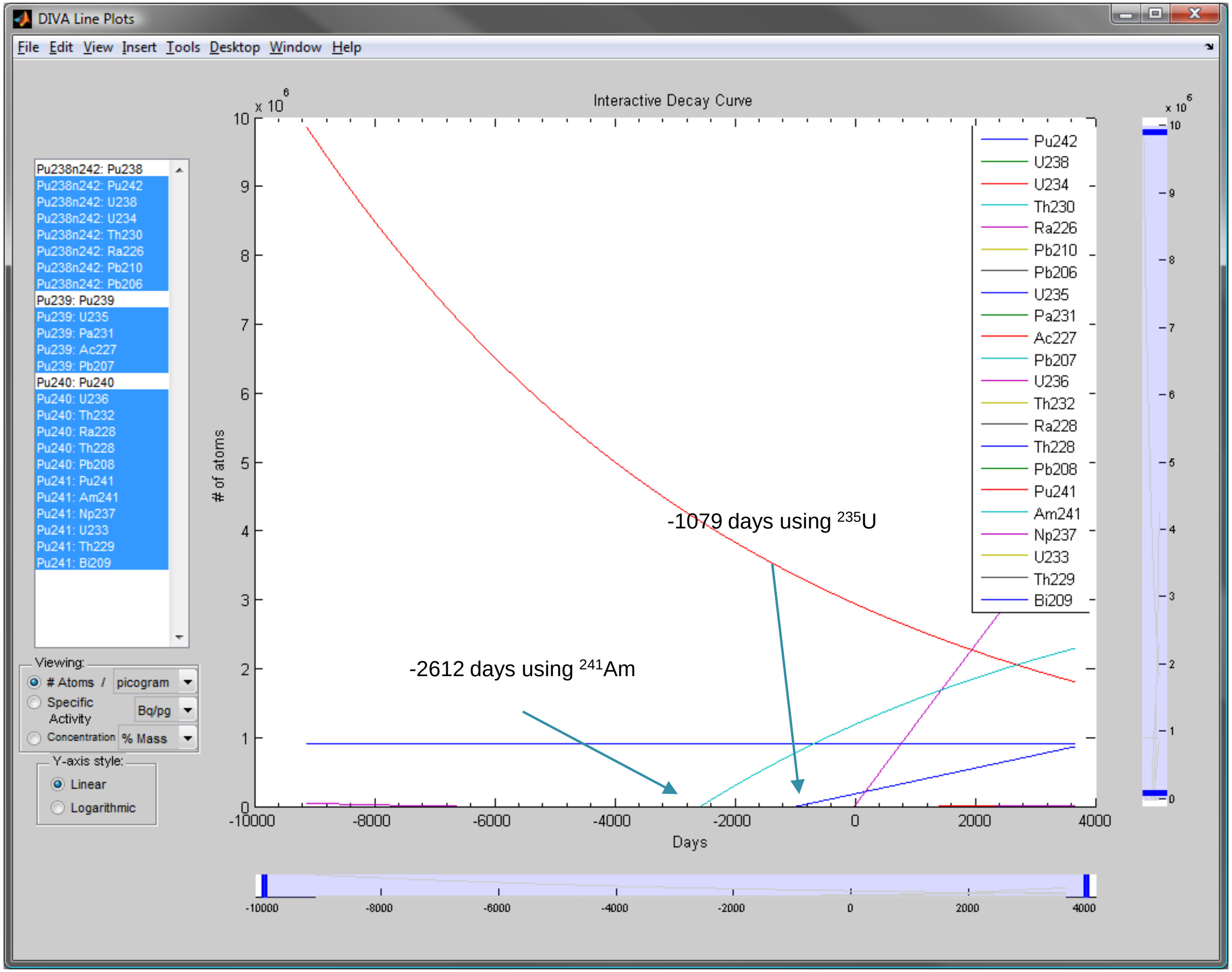
Two assays used for the study

- Assay from Dr. Sanjoy Mukhopadhyay (RSL)
  - Obtained by the team on 09/08/2010 through previous collaboration
  - The underlying sample is named SAMPLE_M
  - Assay date: 11/23/2008
  - Sample EOE dates: 19~20 years from 11/23/2008 (11/23/1988 ~ 11/23/1989)
  - CPR estimated age: 7270 days (19.9 years) from 11/23/2008
  - CPR estimated EOE date: 12/28/1988
- Assay from Dr. Lav Tandon (LANL)
  - Obtained by the team on 9/15/2011 through collaboration with LANL
  - The underlying sample is named SAMPLE_T
  - Assay date 05/20/2008
  - Sample EOE date: 1601 days from 05/20/2008 (~01/01/2004) (~4.38 years) – It may also be a mixed SNM, but this is unconfirmed
  - CPR estimated ages: 2612 days using ^{241}Am / 1079 days using ^{235}U from 5/20/2008
  - CPR estimated EOE dates: 03/26/2001 ~ 06/06/2005

SAMPLE_M Decay Dynamics



SAMPLE_T Decay Dynamics



Sample Mixing Schemes (Mixing date 9/29/2011)

Mixing Ratios	Stable Isotopes	SAMPLE_M	SAMPLE_T	Comments
MixS00M20T80	0%	20%	80%	Without non-radioactive component
MixS00M40T60	0%	40%	60%	
MixS00M50T50	0%	50%	50%	
MixS00M60T40	0%	60%	40%	
MixS00M80T20	0%	80%	20%	
MixS50M20T80	50%	10%	40%	With 50% non-radioactive component
MixS50M40T60	50%	20%	30%	
MixS50M50T50	50%	25%	25%	
MixS50M60T40	50%	30%	20%	
MixS50M80T20	50%	40%	10%	
MixS80M20T80	80%	4%	16%	With 80% non- radioactive component
MixS80M40T60	80%	8%	12%	
MixS80M50T50	80%	10%	10%	
MixS80M60T40	80%	12%	8%	
MixS80M80T20	80%	16%	4%	

Expectations of the Mixture Decay Dynamics

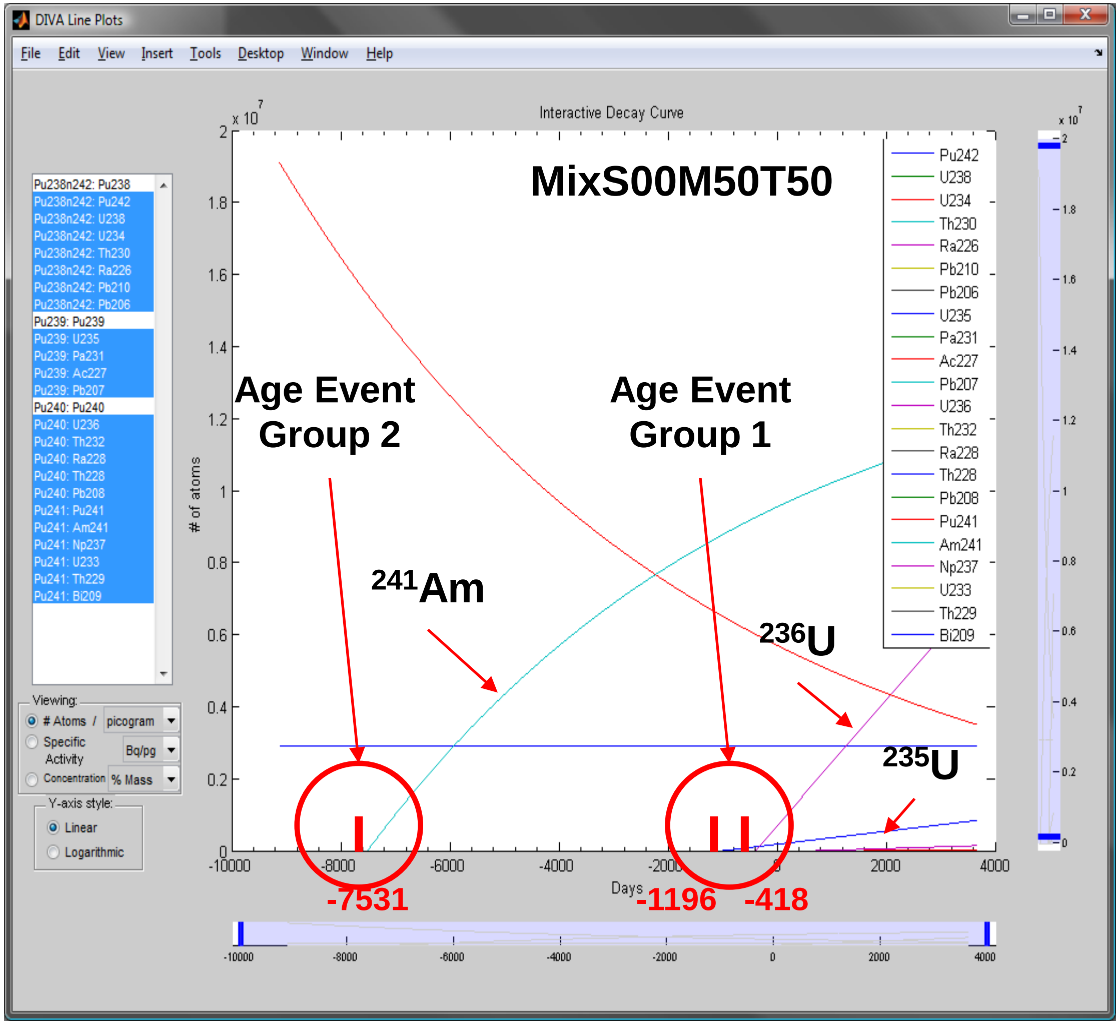
- Since (with respect to 09/29/2011):
  - SAMPLE_M
    - Actual age: 7981~8346 days (21.85~22.85 years)
    - CPR age = 7270 + 1040 = 8310 days (~22.75 years)
  - SAMPLE_T
    - Actual age: 1601 + 1227 = 2828 days (~7.74 years) – unconfirmed
    - CPR ages: 3938 days (~10.51 years) using ^{241}Am / 2306 days (~6.31 years) using ^{235}U
- The mixture decay dynamics should show these features:
  - Two distinctive age events or event groups
  - One event (or event group) should be close to –8000 days, primarily affected by the composition of SAMPLE_M
  - One event (or event group) should be close to –3000 days, primarily affected by the composition of SAMPLE_T

Decay Dynamics: Close Up with MixS00M50T50

Group	Chronometer	Age
#1	^{236}U	–418
	^{235}U	–1196
#2	^{241}Am	–7531

The ages of both Groups 1 and 2 are influenced by the ages of SAMPLE_T and SAMPLE_M, but with larger errors

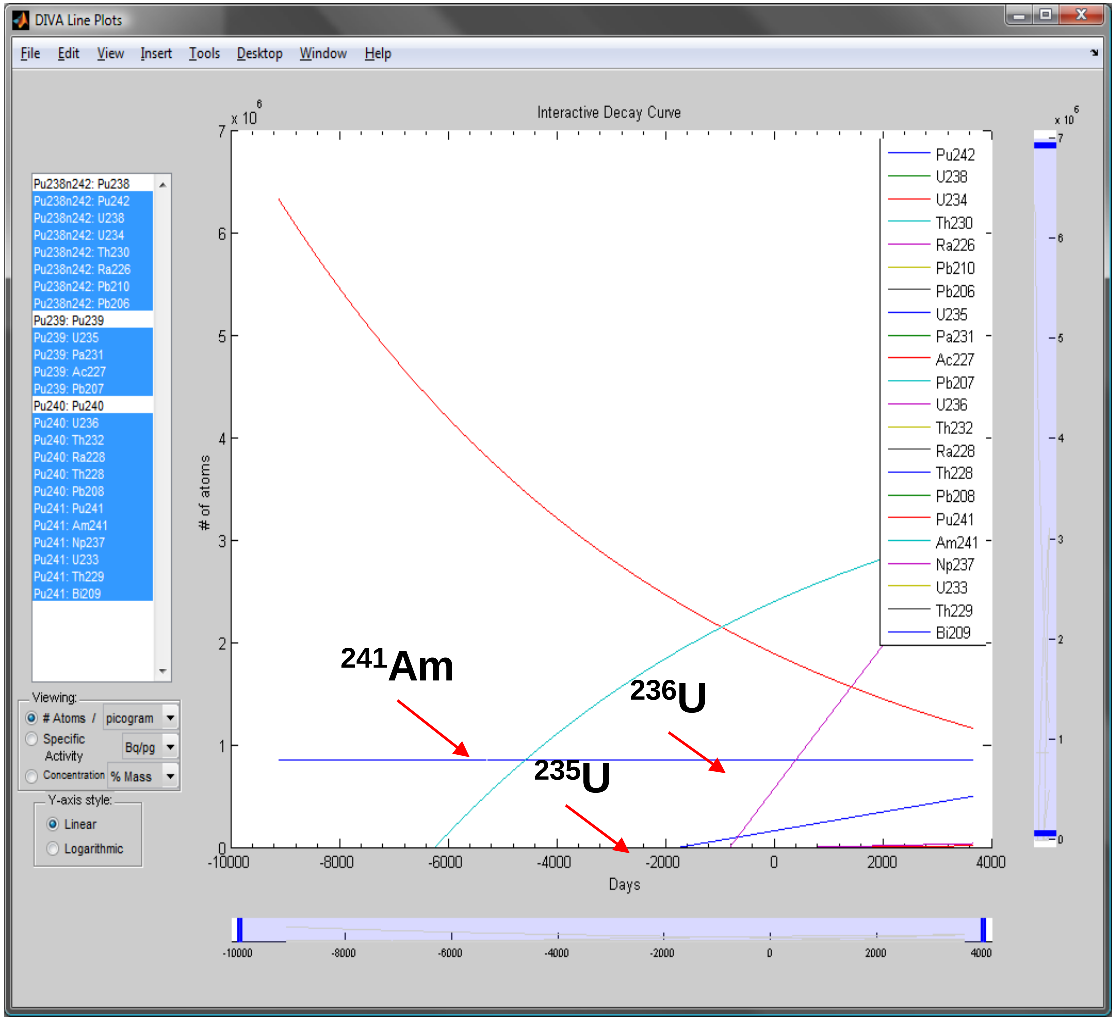
This illustrates the viability of using the CPR method for spoof detection (two EOE dates are indicated)



Decay Dynamics: Close Up with MixS50M20T80

Group	Chronometer	Age
#1	^{236}U	–840
	^{235}U	–1872
#2	^{241}Am	–6267

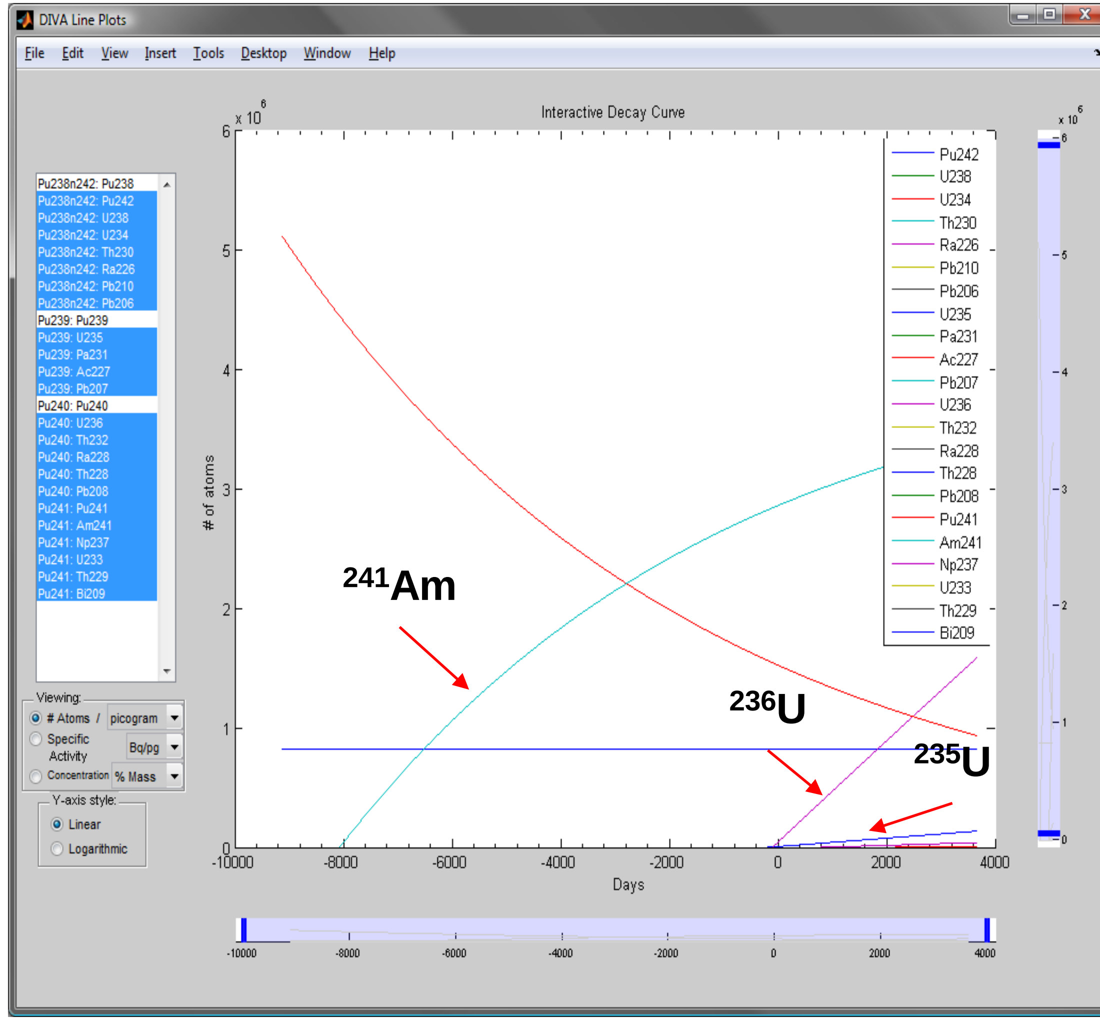
Group 1 ages are close to the age of SAMPLE_T, but the Group 2 age is far off the age of SAMPLE_M



Decay Dynamics: Close Up with MixS80M80T20

Group	Chronometer	Age
#1	^{236}U	–139
	^{235}U	–490
#2	^{241}Am	–8089

Group 2 age is close to age of SAMPLE_M, but Group 1 ages are far off the age of SAMPLE_T



Observations of the Mixture Decay Dynamics

- All dynamics generally show two groups of age estimates, regardless of mixing ratios. This is the key feature for spoof detection.
- Mixing ratios between SAMPLE_M and SAMPLE_T affect the age estimates of the two components.
- Amount of the stable constituents in the mixture appears having no effect on the age estimates.
- If there is a dominant component (≥80%) between SAMPLE_M and SAMPLE_T in the mixture, then one of the two age estimation groups should be very close to the true age of the dominant sample.
- The last observation may have an important forensics implication: If a SNM sample is only slightly contaminated, then among its possible multiple age estimates, one of them is (nearly) correct.

Summary and Conclusions

- We have presented an overview of the Constrained Progressive Reversal (CPR) method for computing decays, age dating, and spoof detecting. The CPR method is:
  - Capable of temporal profiling a SNM sample
  - Precise (compared with known decay code, such a ORIGEN)
  - Easy (for computer implementation and analysis)

- We have illustrated with real SNM data using CPR for age dating and spoof detection
  - If SNM is pure, may use CPR to derive its age
  - If SNM is mixed, CPR will indicate that it is mixed or spoofed

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