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Title:	Concordant Plutonium-241-Americium-241 Dating of Environmental Samples: Results from Forest Fire Ash ~~(Viewgraphs)~~
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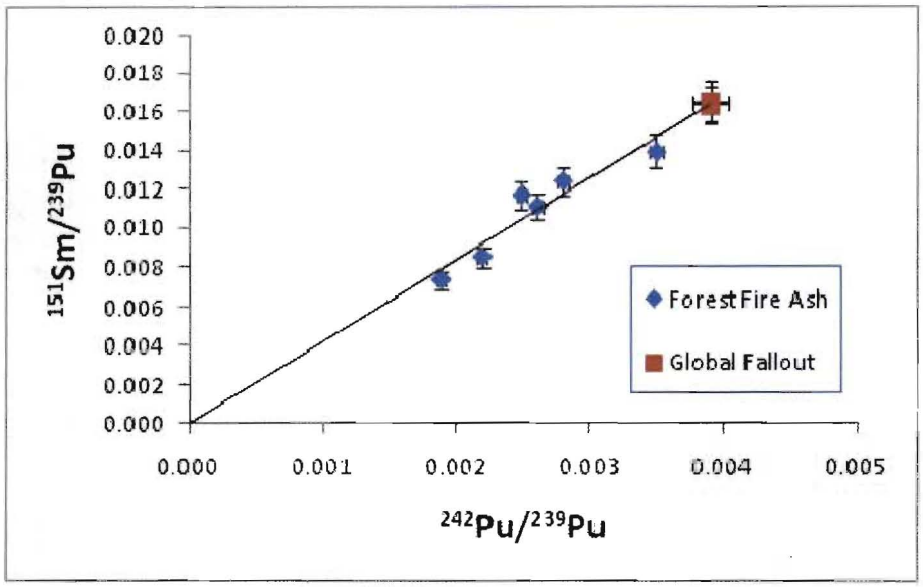
Form 836 (7/06)

Concordant ^{241}Pu-^{241}Am Dating of Environmental Samples: Results from Forest Fire Ash

Steven J. Goldstein, Warren J. Oldham, Michael T. Murrell, and Danny Katzman

We have measured the Pu, ^{237}Np, ^{241}Am, and ^{151}Sm isotopic systematics for a set of forest fire ash samples from various locations in the western U.S. including Montana, Wyoming, Idaho, and New Mexico. The goal of this study is to develop a concordant ^{241}Pu ($t_{1/2}$ = 14.4 y)-^{241}Am dating method for environmental collections. Environmental samples often contain mixtures of components including global fallout. There are a number of approaches for subtracting the global fallout component for such samples. One approach is to use ^{242}Pu/^{239}Pu as a normalizing isotope ratio in a three-isotope plot, where this ratio for the non-global fallout component can be estimated or assumed to be small. This study investigates a new, complementary method of normalization using the long-lived fission product, ^{151}Sm ($t_{1/2}$ = 90 y). We find that forest fire ash concentrates actinides and fission products with ~1E10 atoms ^{239}Pu/g and ~1E8 atoms ^{151}Sm/g, allowing us to measure these nuclides by mass spectrometric (MIC-TIMS) and radiometric (liquid scintillation counting) methods. The forest fire ash samples are characterized by a western U.S. regional isotopic signature representing varying mixtures of global fallout with a local component from atmospheric testing of nuclear weapons at the Nevada Test Site (NTS). Our results also show that ^{151}Sm is well correlated with the Pu nuclides in the forest fire ash, suggesting that these nuclides have similar geochemical behavior in the environment. Results of this correlation indicate that the ^{151}Sm/^{239}Pu atom ratio for global fallout is ~0.164, in agreement with an independent estimate of 0.165 based on 137Cs fission yields for atmospheric weapons tests at the NTS. ^{241}Pu-^{241}Am dating of the non-global fallout component in the forest fire ash samples yield ages in the late 1950's-early 1960's, consistent with a peak in NTS weapons testing at that time. The age results for this component are in agreement using both ^{242}Pu and ^{151}Sm normalizations, although the errors for the 151Sm correction are currently larger due to the greater uncertainty of their measurements. Additional efforts to develop a concordant ^{241}Pu-^{241}Am dating method for environmental collections are underway with emphasis on soil cores.


0.020
0.018
0.016
0.014
0.012
0.010
0.008
0.006
0.004
0.002
0.000
151Sm/239Pu
0.000
0.001
0.002
0.003
0.004
0.005
242Pu/239Pu
Forest Fire Ash
Global Fallout

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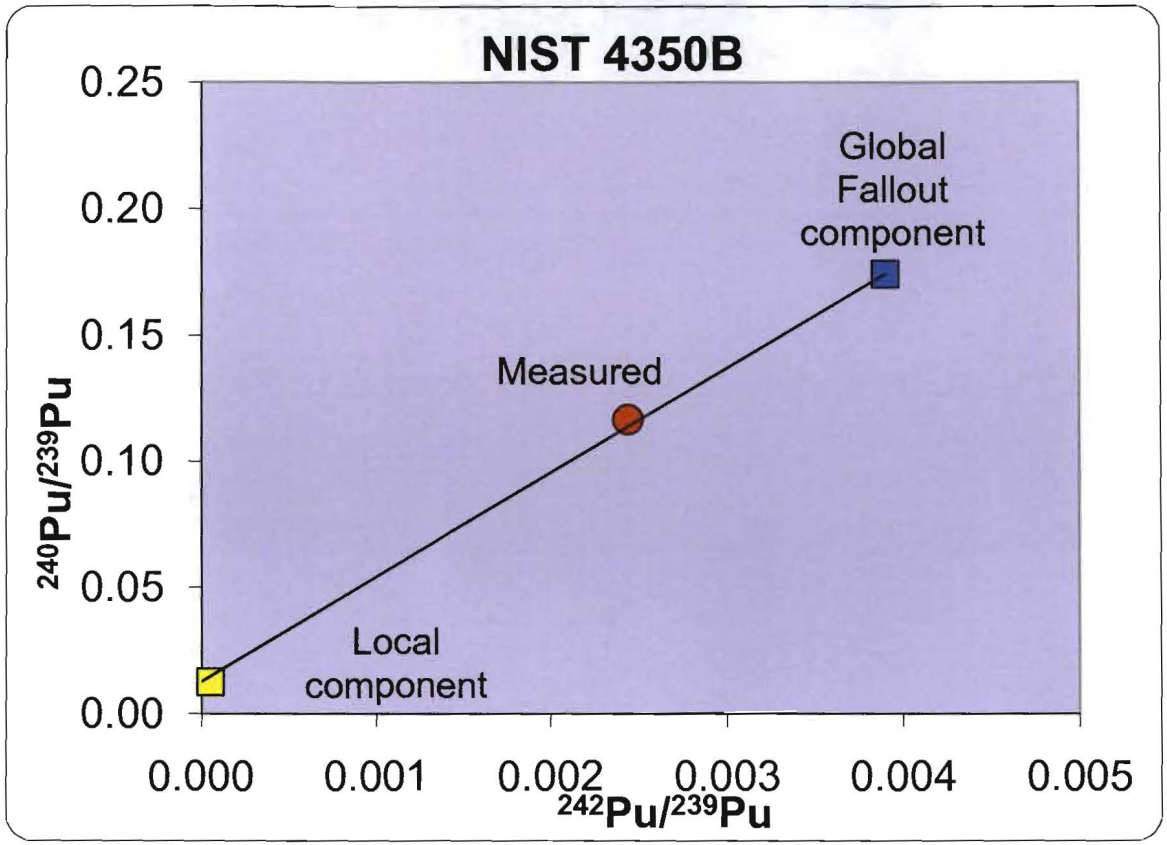
2010 Fall AGU Meeting – December 17, 2010

^{241}Pu-^{241}Am Age Dating of Environmental Samples

- ^{241}Pu ($t_{1/2}$ = 14.4 y) $\longrightarrow$ ^{241}Am ($t_{1/2}$ = 432.7 y)
- Environmental samples often contain mixtures of components
  - Global Fallout
    - Well mixed stratospheric component
    - Peak in middle 1950's-early 1960's
  - Regional Fallout
    - Mixture of global fallout with poorly mixed tropospheric component.
  - Local Component
    - Sample of interest to identify Pu isotopic signature of source and age-date.

An Illustrative Example: NIST SRM 4350B & $^{242}Pu/^{239}Pu$ Corrections

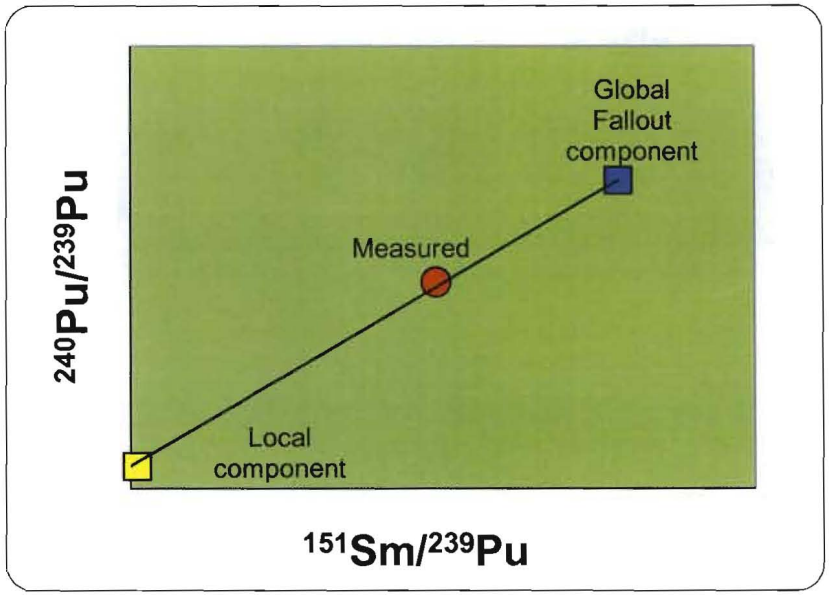


- NIST SRM 4350B: Columbia River sediment near Hanford, WA
- Typically use $^{242}Pu/^{239}Pu$ to correct for global fallout component, but need to know $^{242}Pu/^{239}Pu$ of local component.

$^{240}Pu/^{239}Pu$ Measured	+/- (%)	$^{240}Pu/^{239}Pu$ Local Component	+/- (%)	Local Component Age (cy)	+/- (y)
0.117	0.6	0.013	25	1945	11

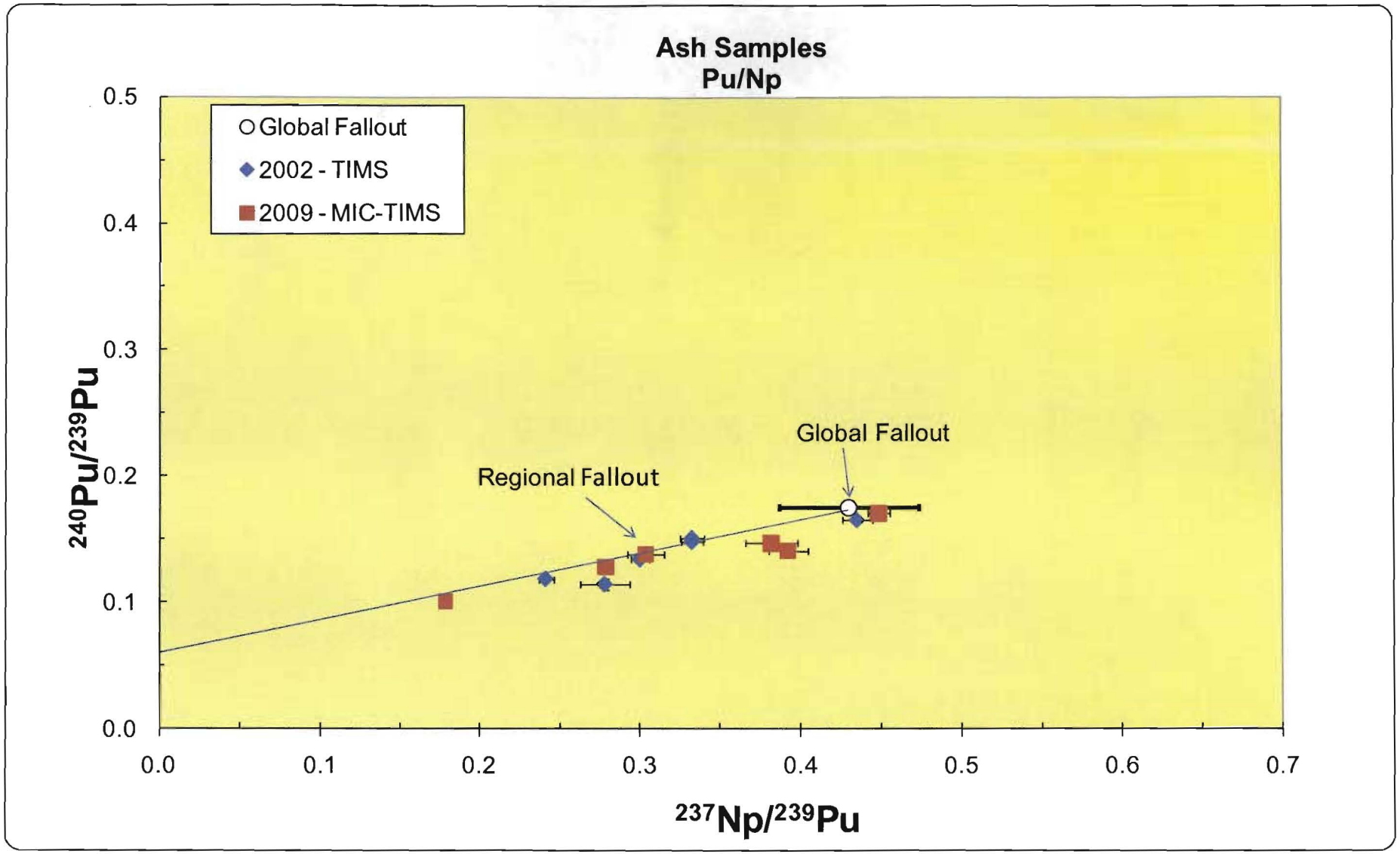
Hypothetical Mixing of Local and Global Components: ^{151}Sm Normalization

- Can we use other fallout nuclides for normalization which may behave geochemically similar to Pu such as ^{151}Sm?
  - ^{151}Sm ($t_{1/2}$ = 97 y): Long-lived lanthanide fission product produced by weapons testing.
  - Evaluate concordance of ages and Pu isotope ratios for local component from multiple approaches.
  - Based on concordance of two approaches, determine $^{242}Pu/^{239}Pu$ and $^{151}Sm/^{239}Pu$ of local component.

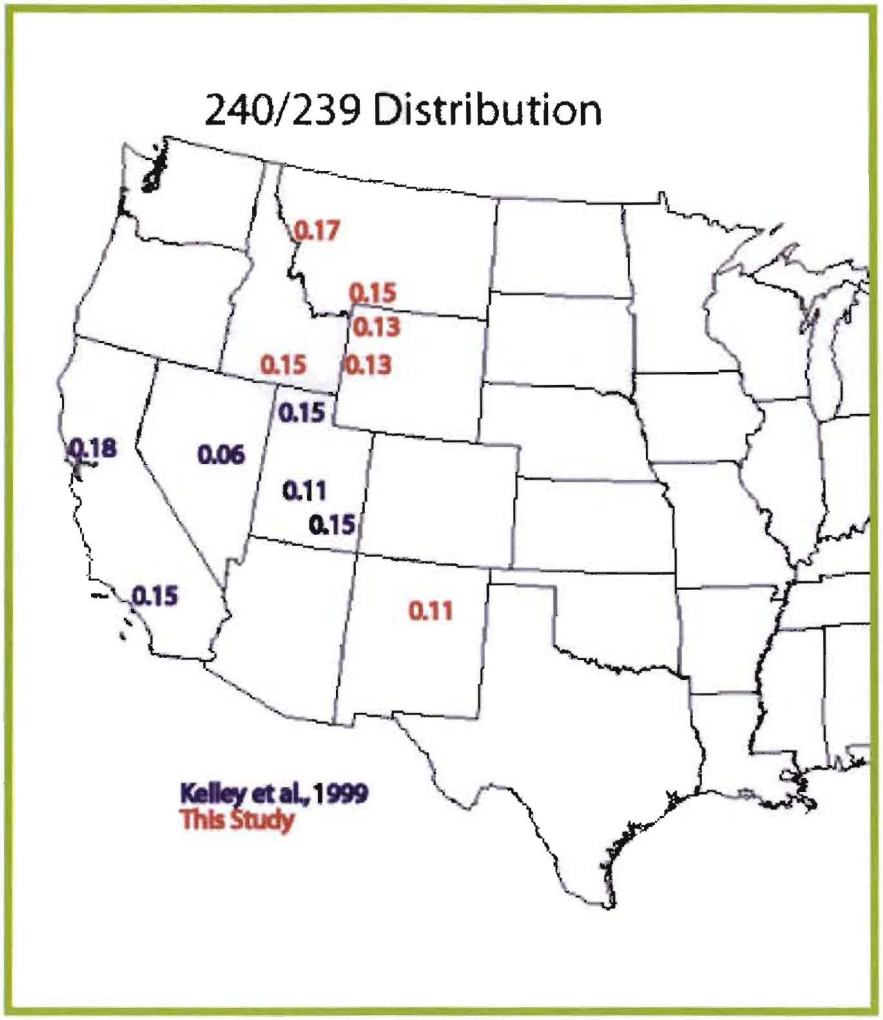


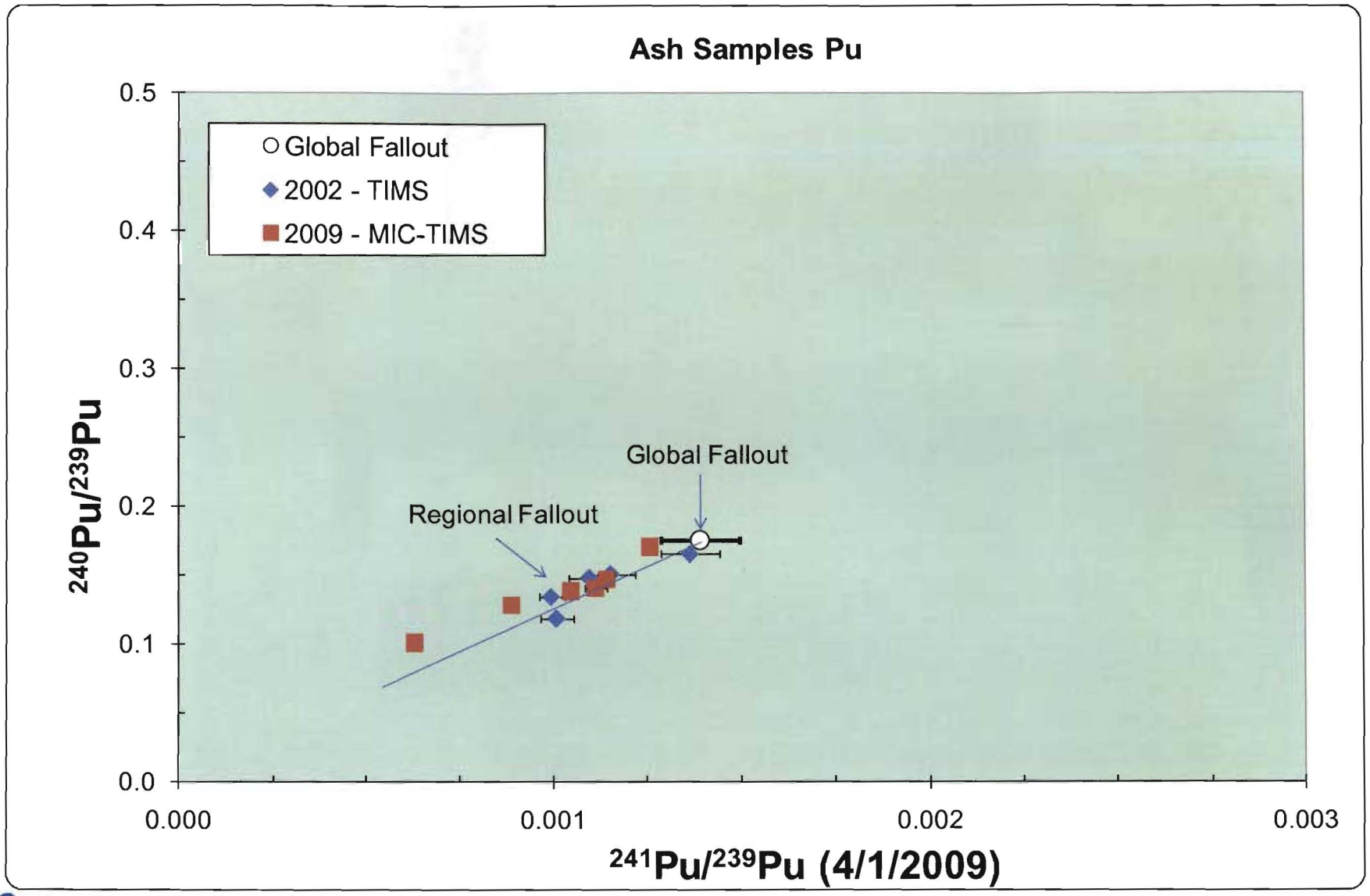
Forest Fire Ash

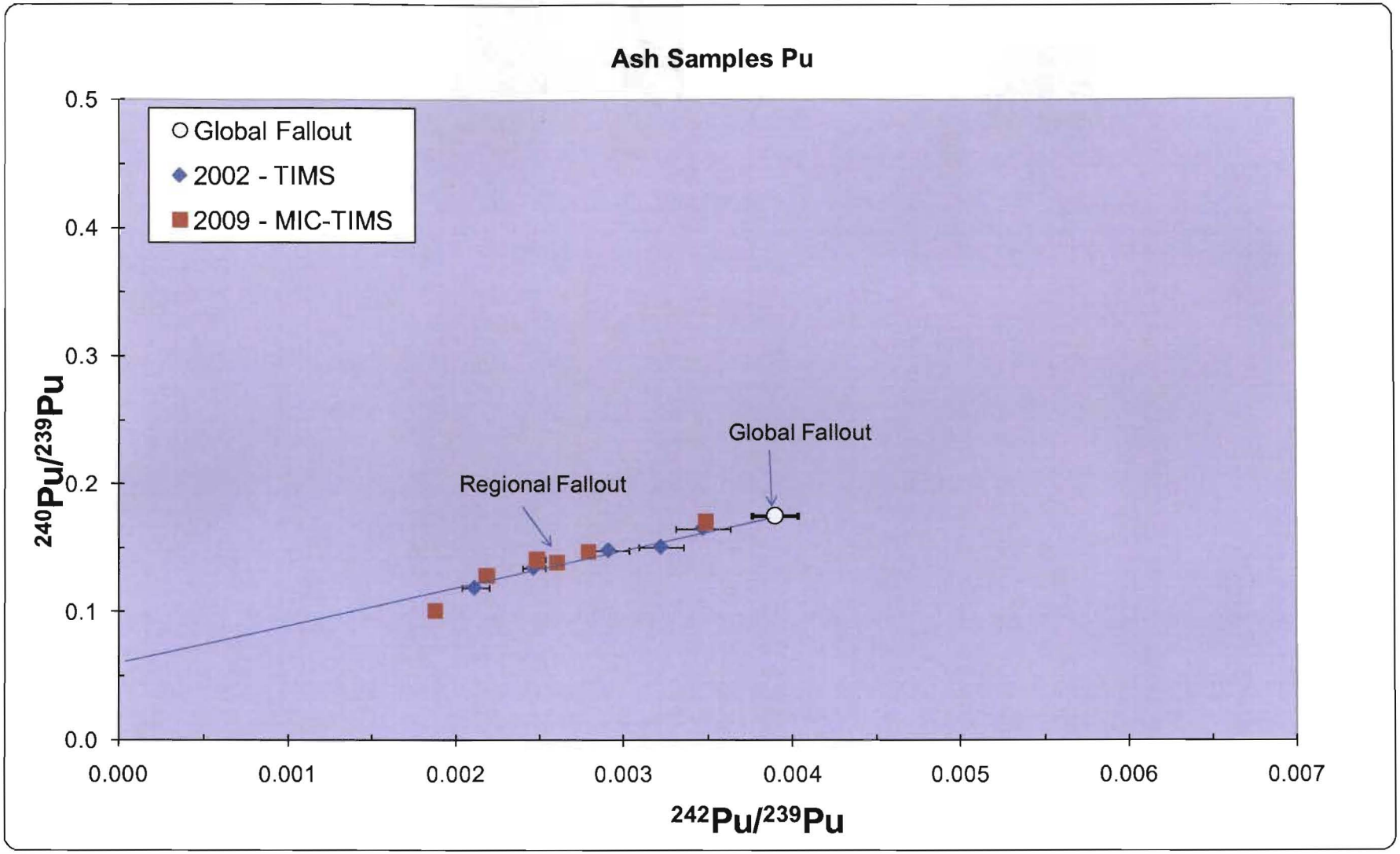
- Samples from 2000 Cerro Grande Fire near Los Alamos, New Mexico, along with other contemporary western U.S. fires in Idaho, Montana, and Wyoming.
- Concentrates actinides and fission products, so good way to determine regional and global $^{151}Sm/^{239}Pu$ systematics
  - Pu ~ 1E10 atoms/g, measureable by MIC-TIMS
  - Am ~ 1E8 atoms/g, measureable by MIC-TIMS or alpha spectrometry
  - ^{151}Sm ~ 1E8 atoms/g, measureable by liquid scintillation counting


Ash Samples
Pu/Np
Global Fallout
2002 - TIMS
2009 - MIC-TIMS
Regional Fallout
Global Fallout
240Pu/239Pu
237Np/239Pu
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

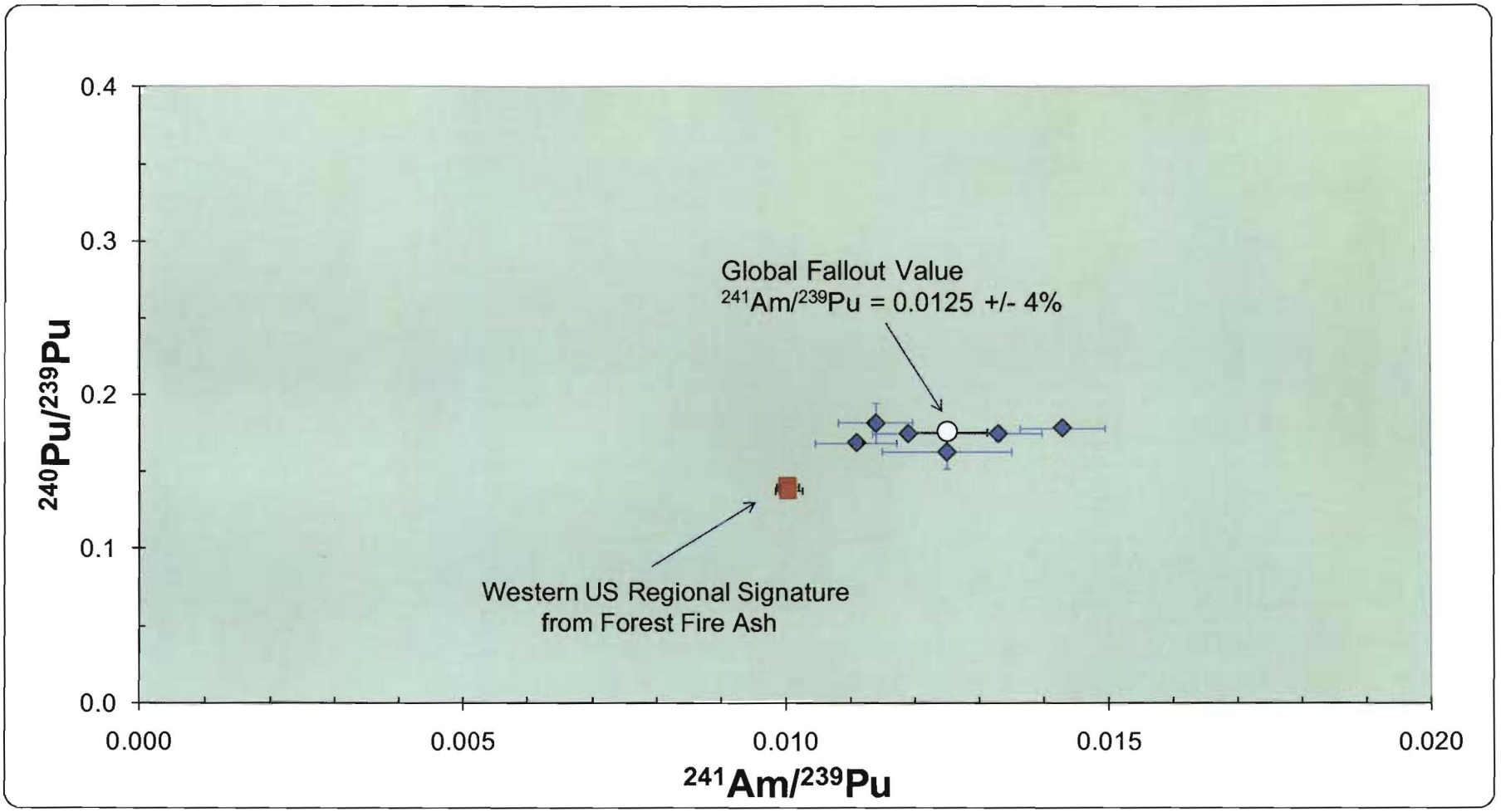
Forest Fire Ash: Spatial Distribution of Pu Signatures



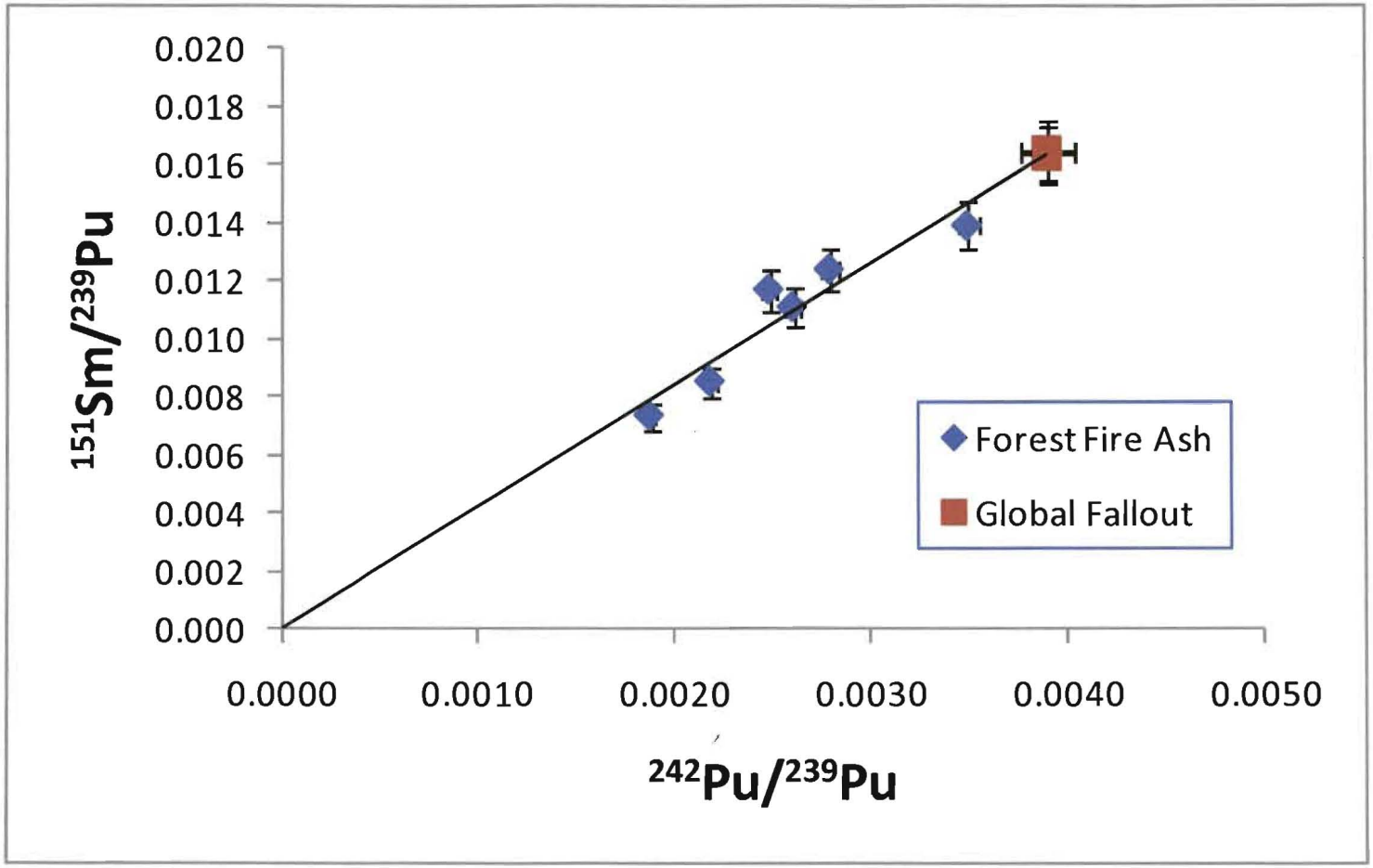

Ash Samples Pu
Global Fallout
2002 - TIMS
2009 - MIC-TIMS
Global Fallout
Regional Fallout
0.5
0.4
0.3
0.2
0.1
0.0
0.000
0.001
0.002
0.003
240Pu/239Pu
241Pu/239Pu (4/1/2009)


Ash Samples Pu
Global Fallout
2002 - TIMS
2009 - MIC-TIMS
Regional Fallout
Global Fallout
0.5
0.4
0.3
0.2
0.1
0.0
240Pu/239Pu
0.000
0.001
0.002
0.003
0.004
0.005
0.006
0.007
242Pu/239Pu

LANL Measurements of $^{241}Am/^{239}Pu$ in Global and Regional Fallout



$^{151}Sm/^{239}Pu$ in Global and Regional Fallout: First Measurements



Global Fallout – LANL Results

$^{237}Np/^{239}Pu$	+/- %	$^{240}Pu/^{239}Pu$	+/- %	$^{241}Pu/^{239}Pu$	+/- %	$^{242}Pu/^{239}Pu$	+/- %	$^{241}Am/^{239}Pu$	+/- %	$^{151}Sm/^{239}Pu$	+/- %
0.43	10	0.175	2.5	0.00127	7.5	0.00390	3.5	0.0125	4	0.0164	6

Table 1 - Atom Ratio Results for Global Fallout (corrected to 2/1/2010)

Americium – Age Dating of Non-Global Fallout Component in Forest Fire Ash

Sample ID	Age (^{242}Pu Corrected)	+/- (y)	Age (^{151}Sm Corrected)	+/- (y)
CABG-01-0091-04 Wyoming	1960	+ 6 - 4	1960	+ 28 -10
CABG-01-0064-04 Idaho	1954	+ 9 - 6	1953	+ 25 -9

Summary

- Forest fire ash samples have Pu-Np-Am-Sm isotopic systematics consistent with a regional western U.S. fallout signature.
- ^{151}Sm and Pu are correlated in forest fire ash samples, suggesting similar geochemical behavior in the environment.
- $^{151}Sm/^{239}Pu = 0.0164$ in global fallout presently.
- Concordant ages (middle 1950's-early 1960's) are obtained for two forest fire ash samples, corresponding to a peak in above ground weapons testing at that time.

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