UNIVERSITY OF NOTRE DAME

NONPROLIFERATION AND NUCLEAR FORENSICS: DETAILED, MULTIANALYTICAL INVESTIGATION OF TRINITITE POST-DETONATION MATERIALS

Antonio Simonetti
Dept. Civil & Environmental Engineering & Earth Sciences
University of Notre Dame

University of Notre Dame- TEAM

Dr. Jeremy Bellucci

Dr. Christine Wallace

Elizabeth Koeman

Dr. Peter C. Burns

Nuclear Forensics

- As "the technical means by which nuclear materials, whether intercepted intact or retrieved from ***post-explosion debris***, are characterized (as to composition, physical condition, age, provenance, history) and interpreted (as to provenance, industrial history, and implications for nuclear device design)." *Joint Working Group of the American Physical Society and the American Association for the Advancement of Science (2010)*

Questions addressed by nuclear forensic analysis & associated time frames

What the event a nuclear explosion? What was the yield?	Hours
Was U or Pu used? or both? Level of sophistication of device?	Hours-Days
Isotopic composition of fuel components? Provenance and history? Do isotope compositions of debris match any from known weapon tests?	Days
What was the most probable design of the device? Do these match any existing designs? Any other materials present that might suggest a particular source?	Several weeks

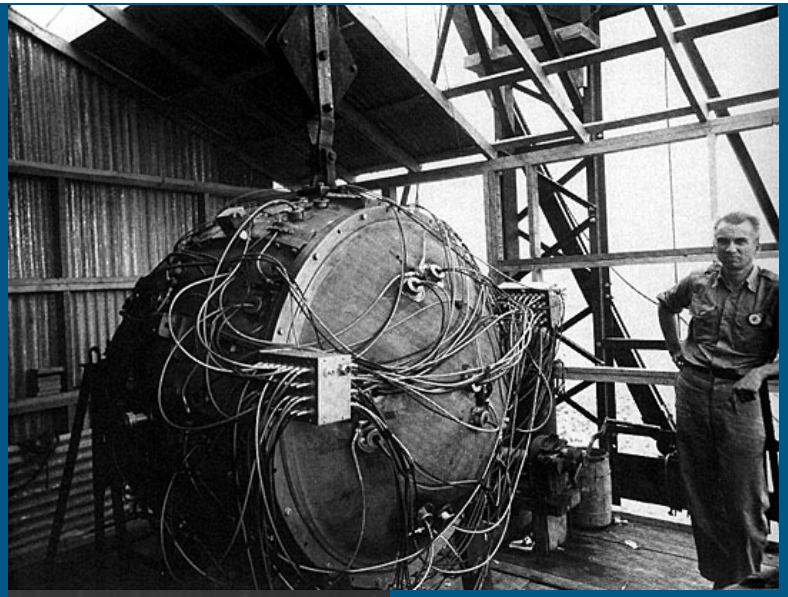
Forensic Analysis
Post-Detonation Materials (PDMs)
Historic Test Sites

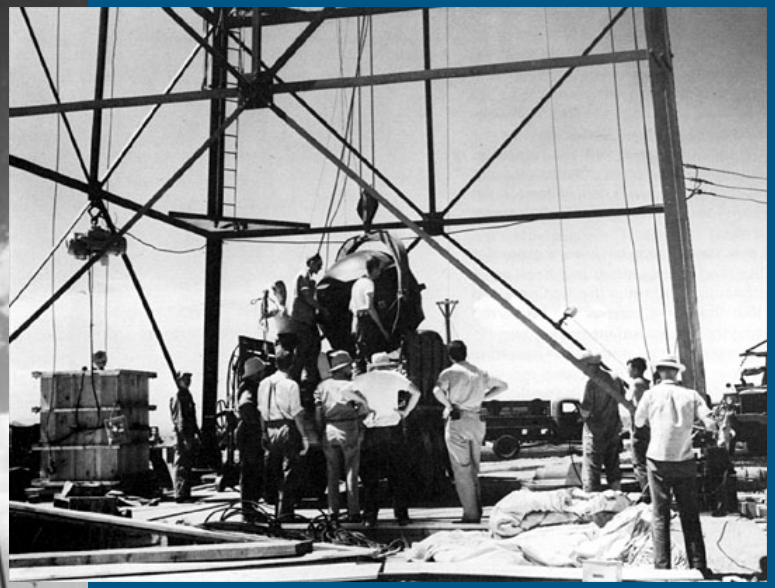
- Ideal for establishing and developing nuclear forensics protocols since the chemical and isotopic composition of weapons employed are well documented; PDMs provide a means to validate forensic results
- Once these new forensic techniques have been established, these can be applied to more recent and sophisticated nuclear detonations
- ***Source attribution*** is the ultimate goal of nuclear forensics!!
- **Trinity Test – Detonation of First Nuclear Device**

Why is the study of *post-detonation materials* from the Trinity test a good starting point?

- Design of trinity device was relatively "simple" and detonated in a remote location with "simple" geologic background (i.e., desert sand)

TRINITY TEST





http://www.trinityremembered.com/photos/index.html

- World's first detonation of a nuclear device
- Nicknamed "Gadget"
- July 16, 1945 at White Sands Missile Range, NM
- Detonated from a ~30 m high tower
- ^{239}Pu-implosion device
- Equiv. to 21 kilotons of TNT

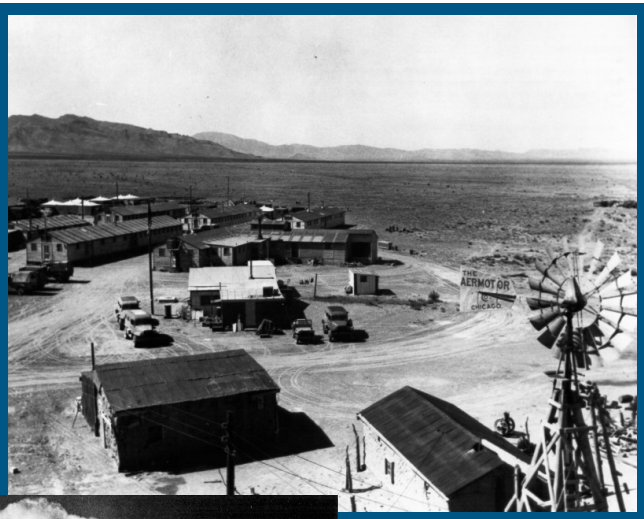
WHAT IS TRINITITE?

- The explosion resulted in the partial melting of the surrounding desert sand and incorporated components of the device and test site materials, which subsequently fused into blast-melt glass referred to as ***Trinitite***.

- Predominantly composed of silica-rich glass

- Contains remnant mineral grains from desert (arkosic sand): **Quartz**, **Feldspars**, **Micas**, **Calcite**, **Gypsum**, minor amounts of **ferromagnesian minerals**, **zircons**

- Can contain remnants of the device, tower, diagnostic equipment: mostly copper and iron

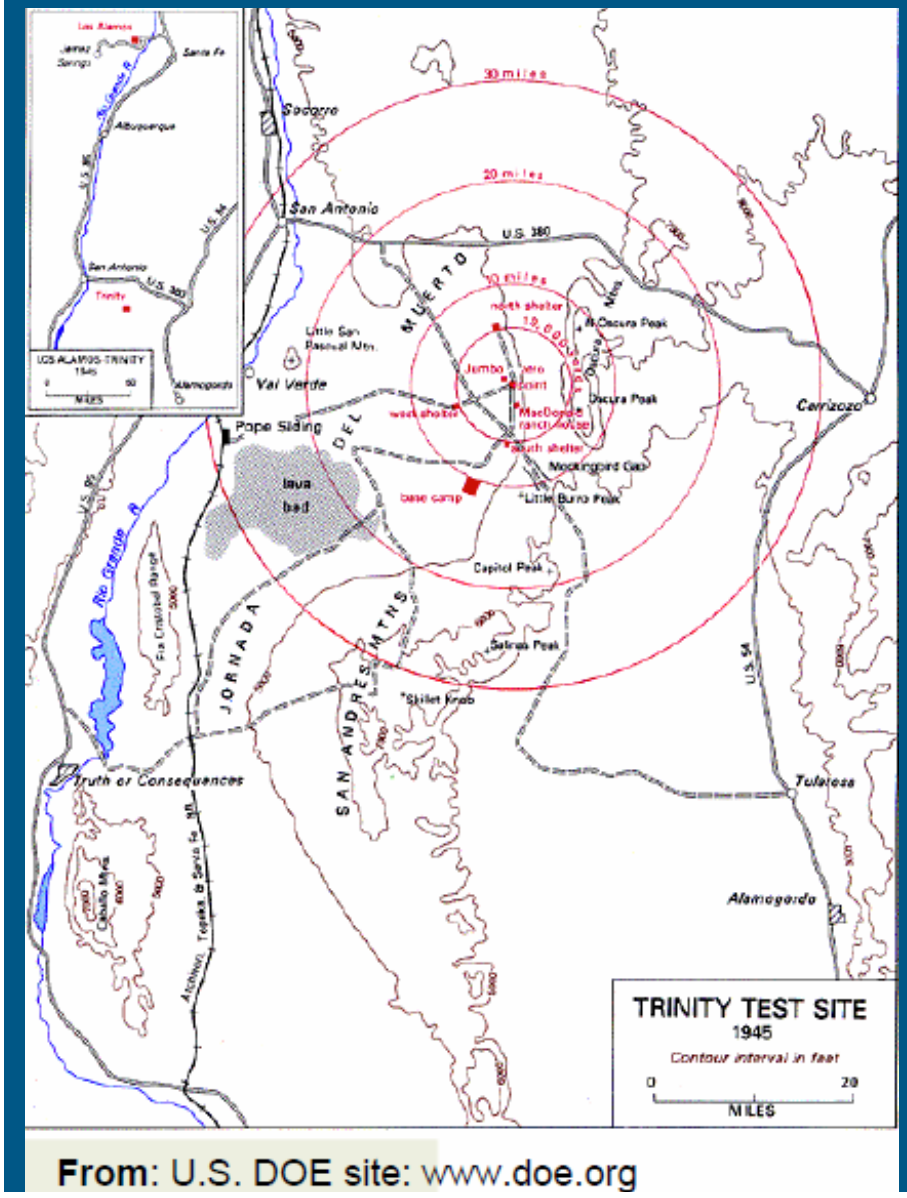


www.awesomestories.com

http://www.trinityremembered.com/photos/index.html


TRINITY SITE
The nuclear age began with the detonation of world's first atomic bomb at the Trinity Site on July 16, 1945. The site may have been named Trinity by J. Robert Oppenheimer, director of the Los Alamos Nuclear Physics Laboratory, who said at the blast, "Now, I am become Death, the destroyer of worlds," quoting from the Bhagavad Gita. The detonation of the bomb marked the culmination of the Manhattan Project.


SMILE
YOU ARE BEING FILMED
KEEP OUT
Enjoy
Coca-Cola
ATOM BOMB
TRINITITE
DOORBELL
WARNING
Area Patrolled by
DACHSHUND
Security



From: U.S. DOE site: www.doe.org

SITE SELECTION:

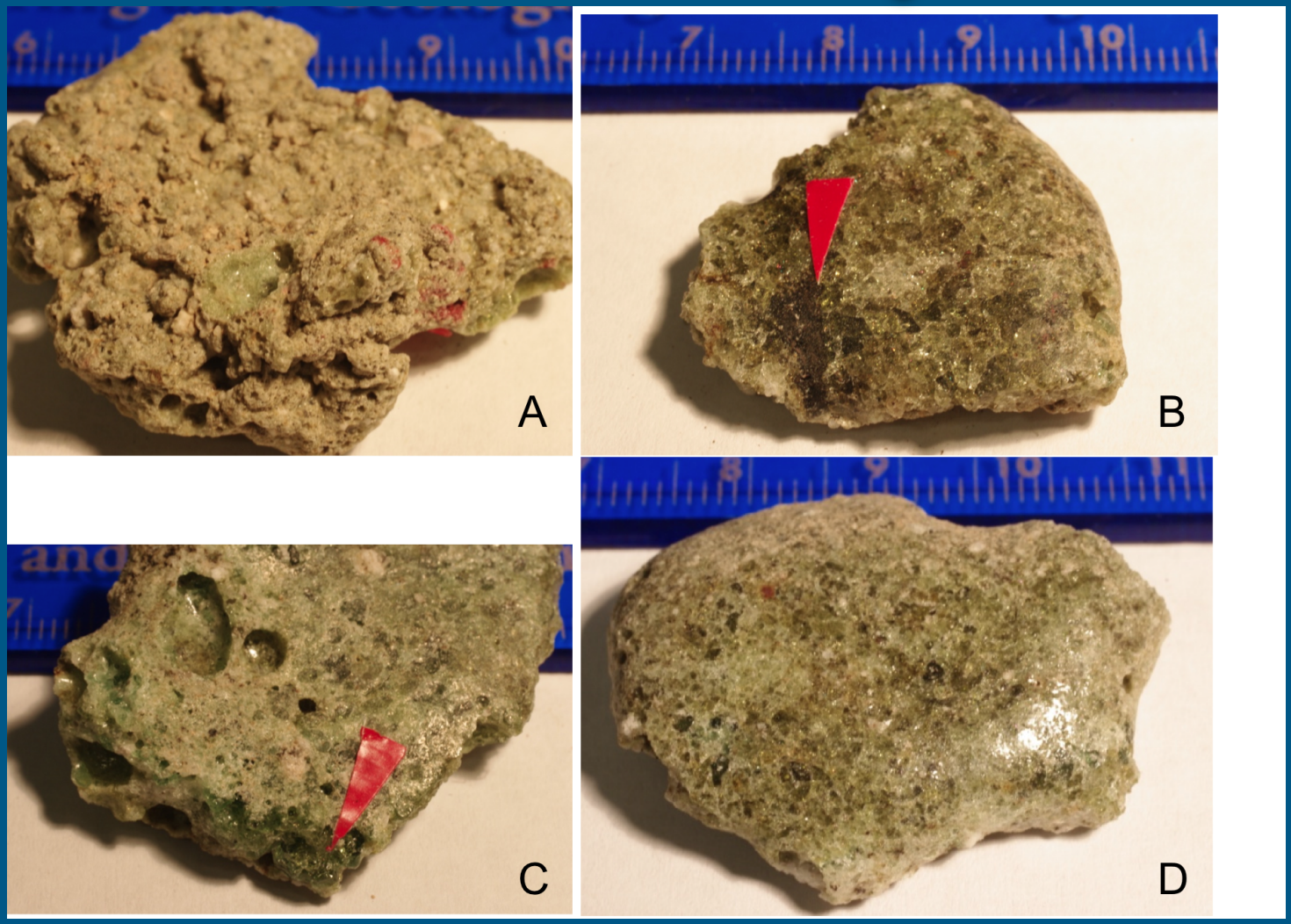
Flat area – minimize extraneous effect of the blast;

Good weather – necessary for good optical information;

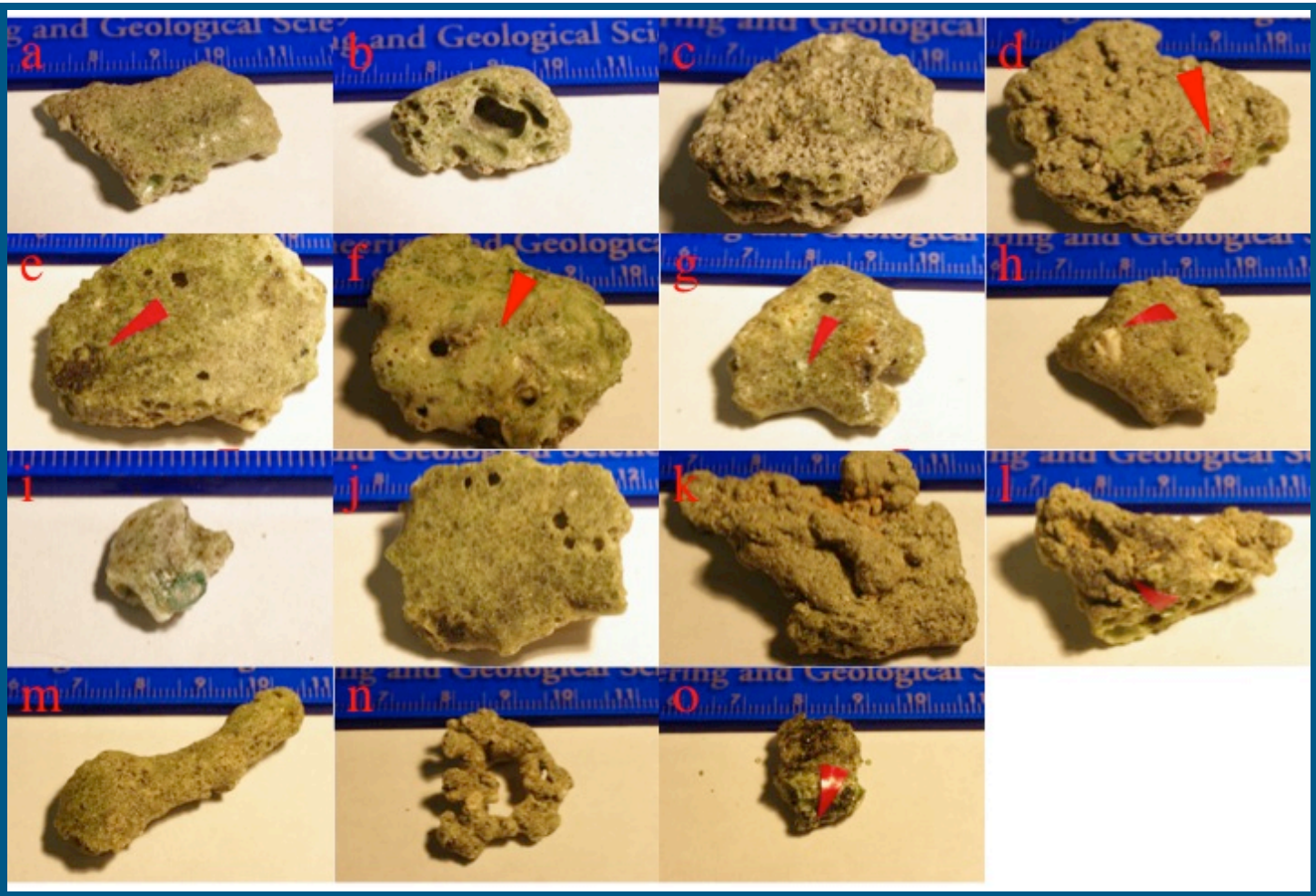
Minimum 20 km distance - from nearest settlement;

Proximity to Los Alamos – minimize transportation of personnel and materials;

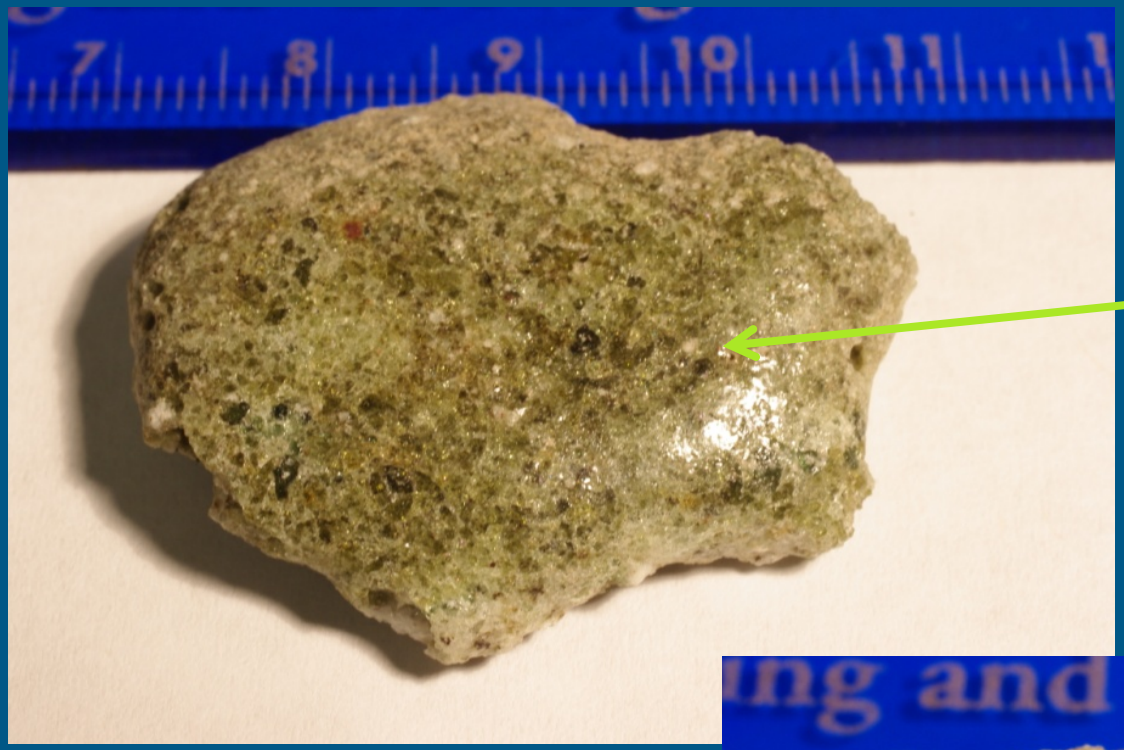
Trinitite Samples



A= Red inclusions; **B**= Black inclusions; **C**= “Coke Bottle”; **D**= “Regular”, green trinitite



- **A** = glass-like fused surface; **B** = large gas pockets on surface/perimeter **C** = rough texture of desert surface; **D** = **red inclusions**; **E** = black inclusions; **F** = light colored glass; **G** = blue inclusions; **H** = white inclusions; **I** = "Coke-bottle" inclusions; **J** = light/dark colored layered glass; **K** = protuberances/casts; **L** = metallic-like coating; **M** = elongated extrusions; **N** = lace-like structures; **O** = iron inclusions



Glass-like fused top surface

Bottom side shows rough texture of precursor desert sand

Vesicles

"Multi-scale Separation & Analysis of Heterogeneous Trinitite Phases"

GOALS and OBJECTIVES

- Conduct detailed chemical and isotopic characterization of trinitite samples at high spatial resolution, i.e. micron scale – development of 'forensic tools'
- The latter will be accomplished using a combination of micro-analytical techniques such as Electron Microprobe Analysis (EMPA), Scanning Electron Microscopy (SEM), Focused Ion Beam (FIB), Transmission Electron Microscopy (TEM), and Laser-Ablation-Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS).
- Development of scientific expertise in the area of nuclear forensics for graduate students and postdoctoral researcher involved.

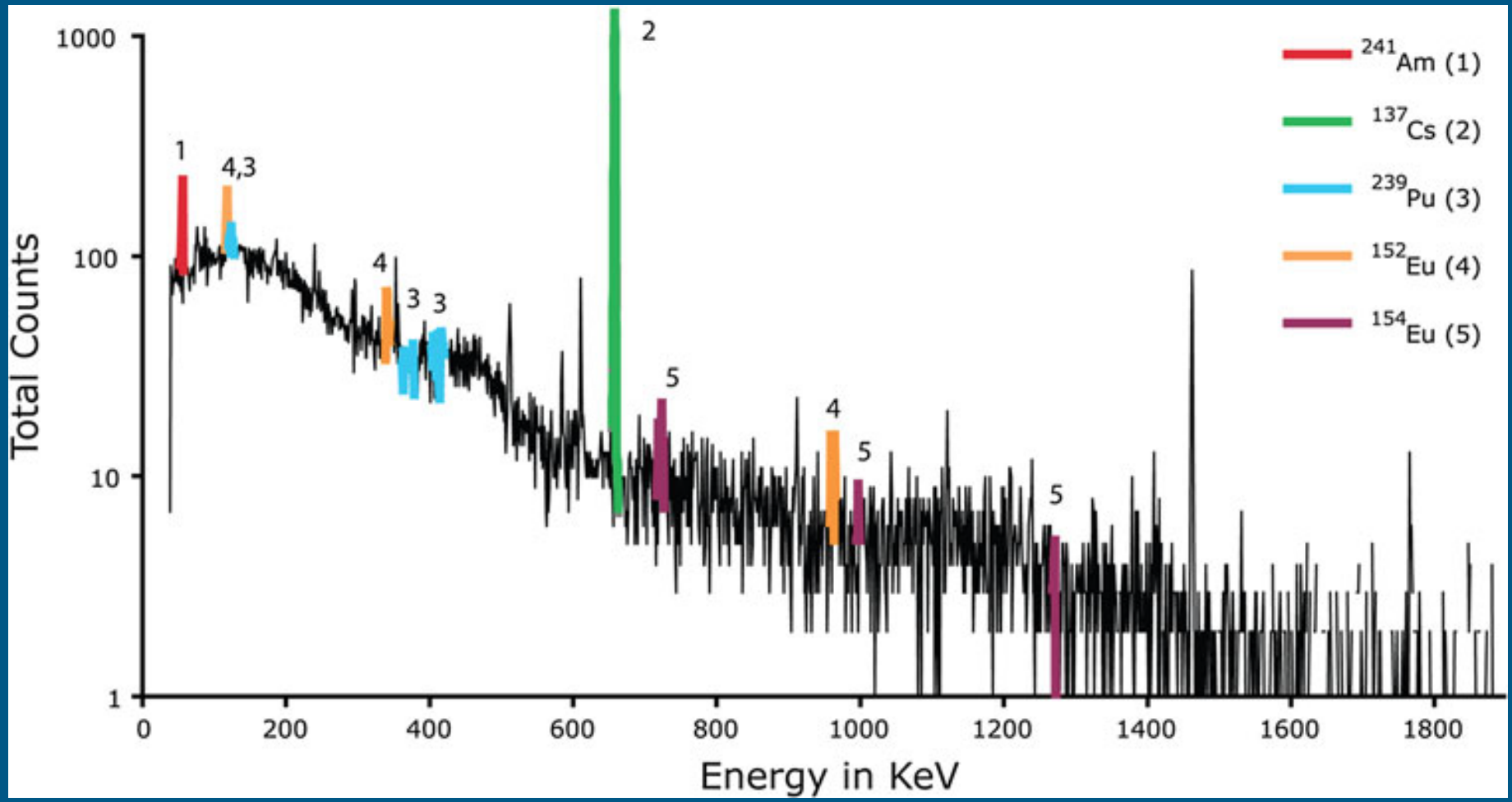
Analytical Methods

- **BULK SAMPLE:**
  - Opitcal microscopy
  - Gamma spectroscopy
  - Alpha track radiography
  - Beta autoradiography
  - Micro- XRF
  - Laser fluorination (stable oxygen isotope)
- **Micron-scale:**
  - SEM-EDS
  - EMP
  - FIB/ TEM
  - LA-(MC)-ICP-MS

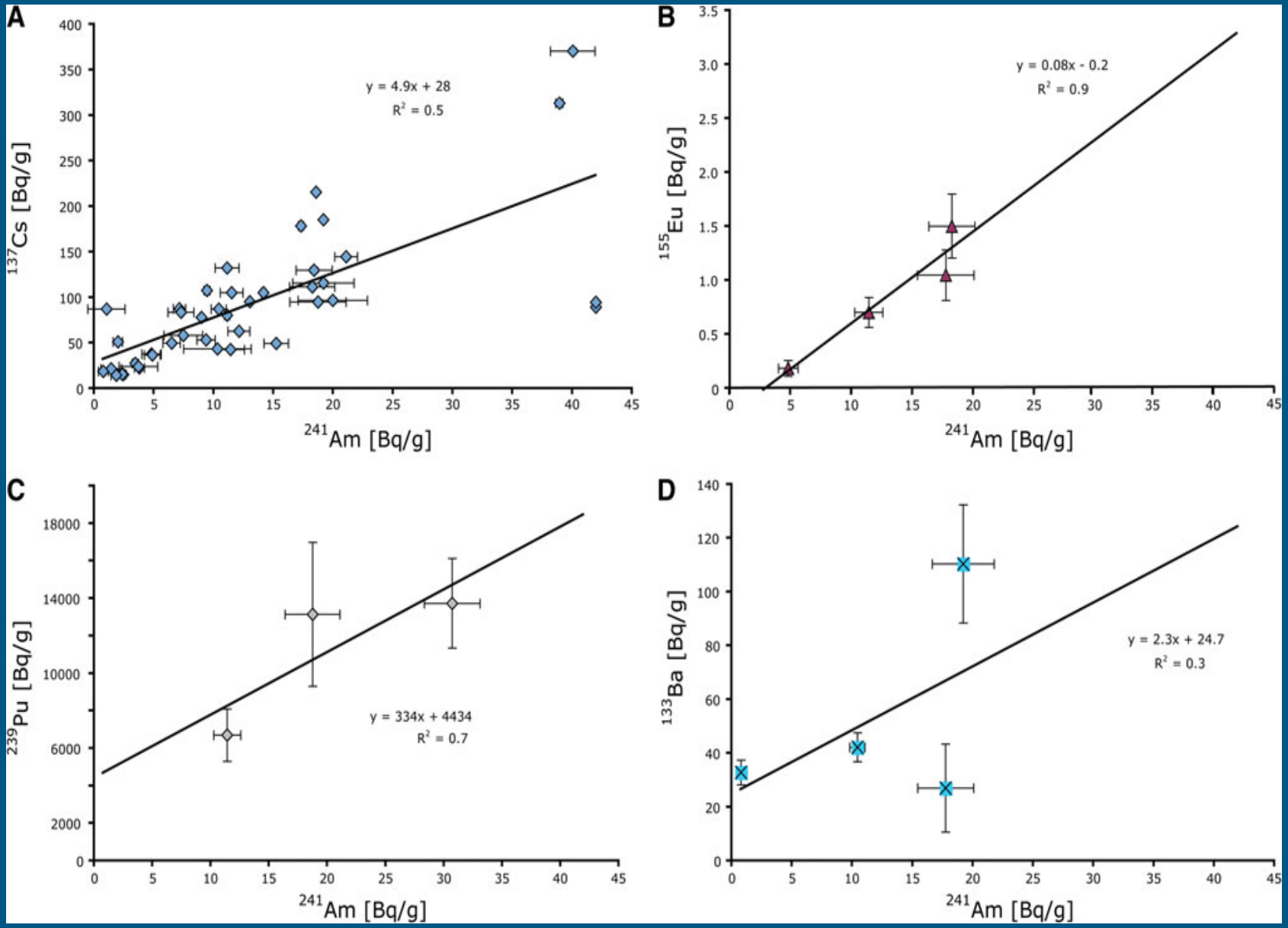
Origin of Radionuclides- Gamma Spectroscopy

Isotope	Half-life/ yr	Origin
^{137}Cs	30,0	fission product (beta decay of ^{137}Xe and ^{137}I and also independently)
^{60}Co	5,271	activation of ^{59}Co – from test tower steel and from soil
^{133}Ba	10,54	activation of ^{132}Ba – Ba part of explosive lense system ($Ba(NO_3)_2$ - Baratol)
^{152,154}Eu	13,33 / 8,8	activation of stable isotopes ^{151,153}Eu in soil by slow neutrons
^{241}Am	432,2	mostly by beta decay daughter of ^{241}Pu, produced mainly from ^{239}Pu during the explosion via double-neutron capture
^{239}Pu	24100	fuel

Bulk Analysis – Gamma Spectroscopy



Bellucci et al. (2013a, JNRC)



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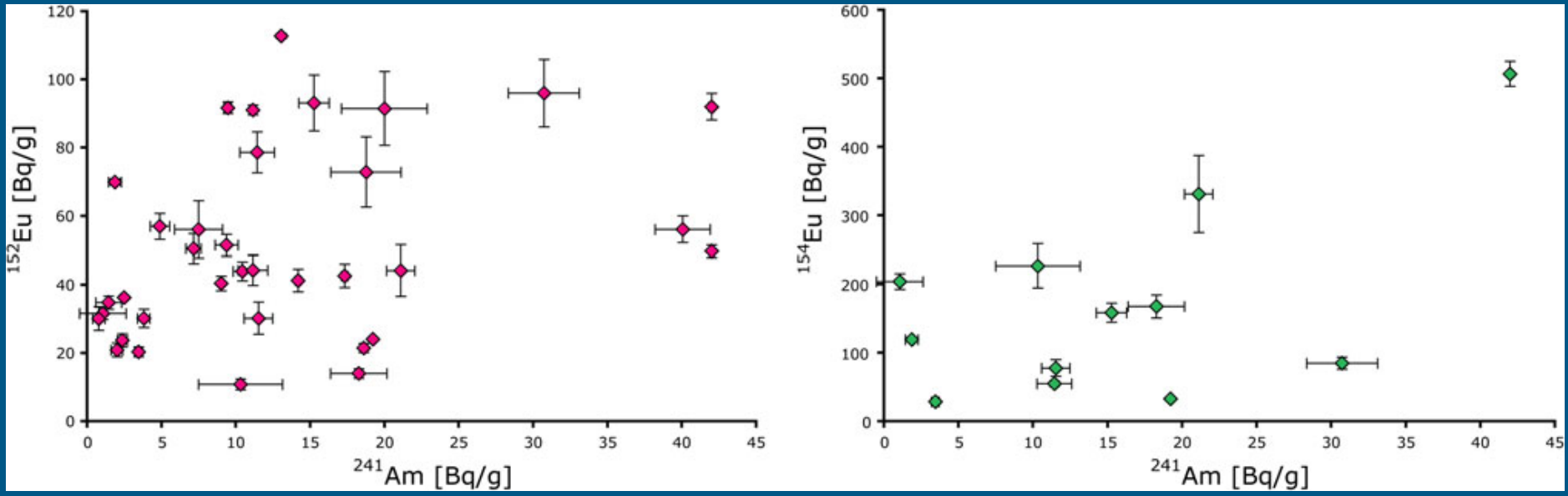

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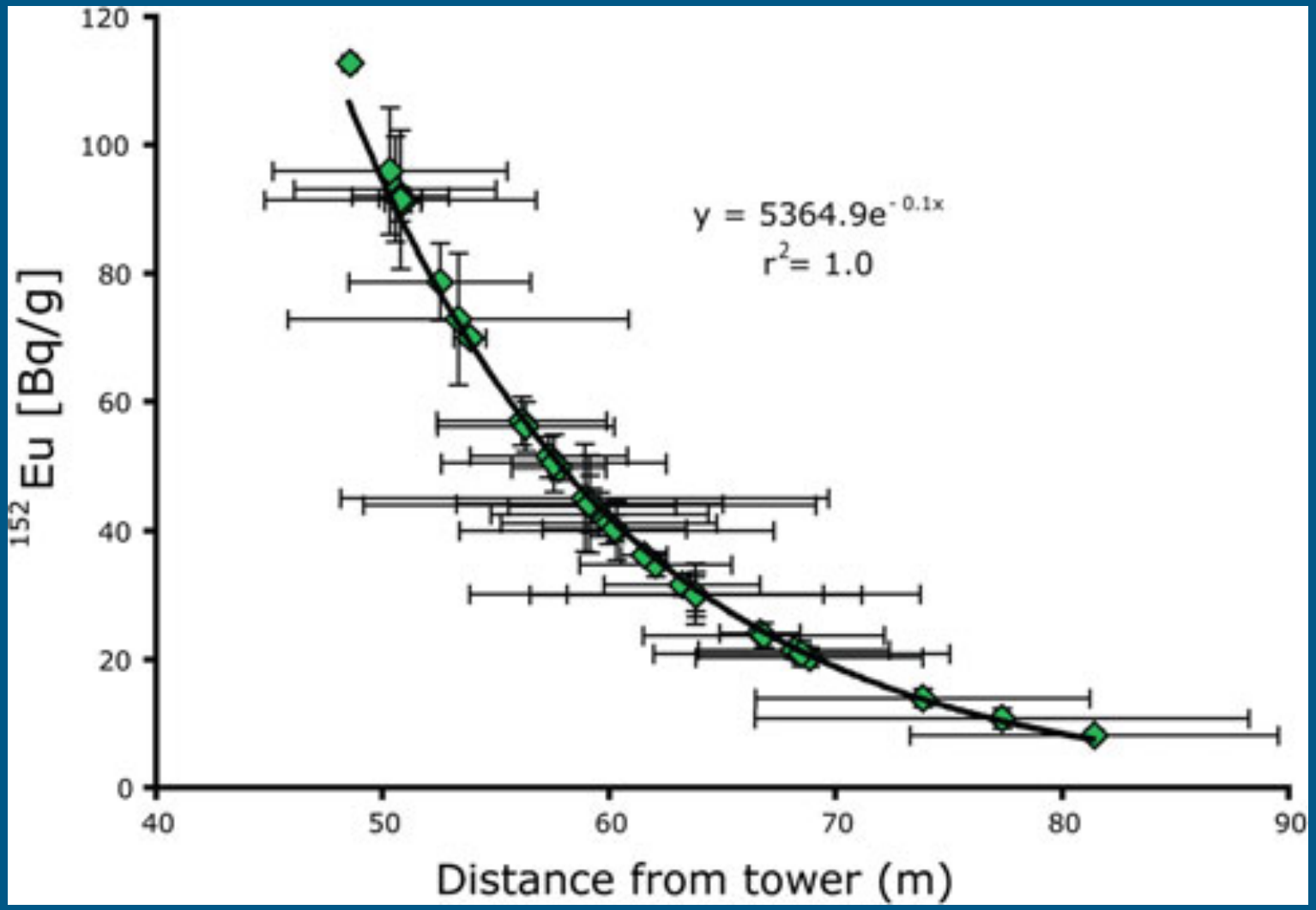
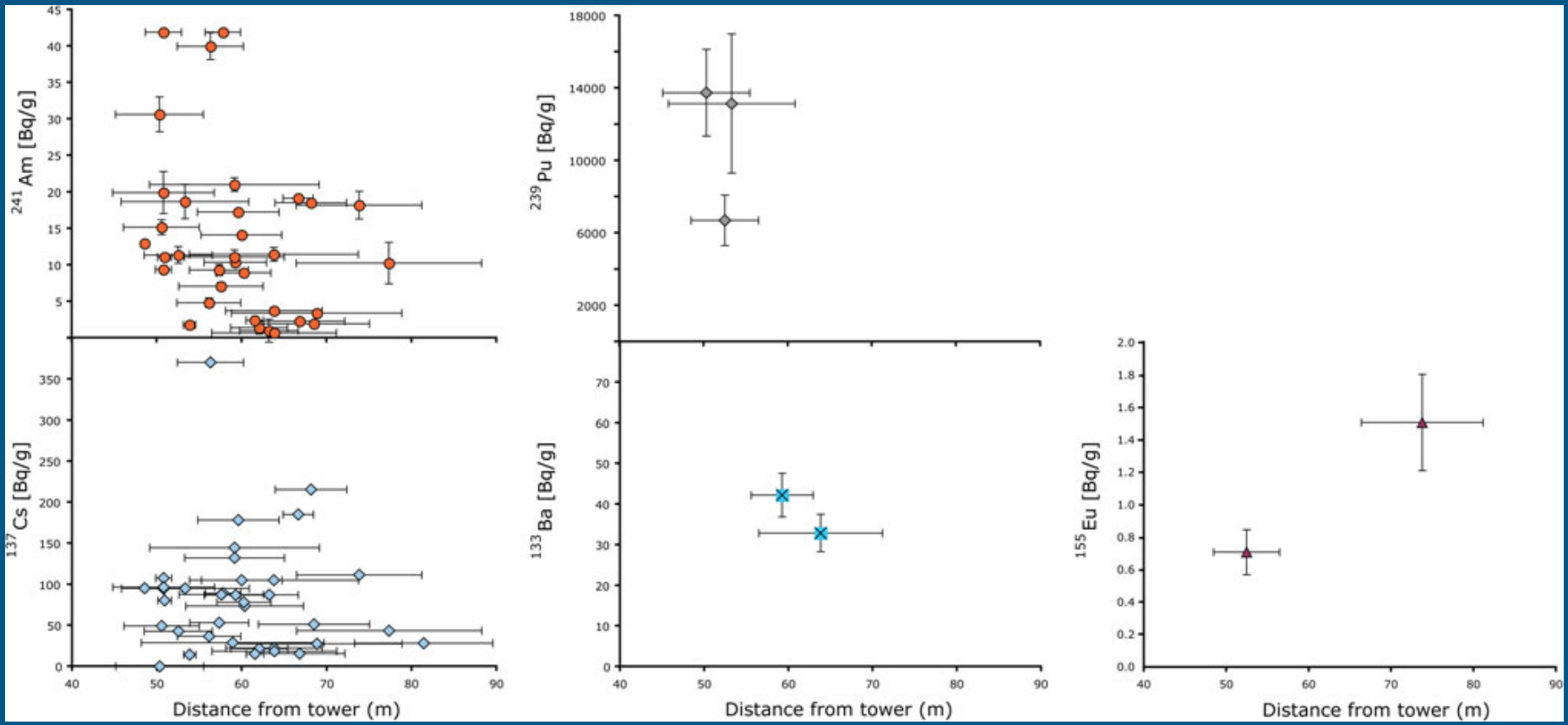


Bellucci et al. (2013a, JNRC)



Bellucci et al. (2013a, JNRC)

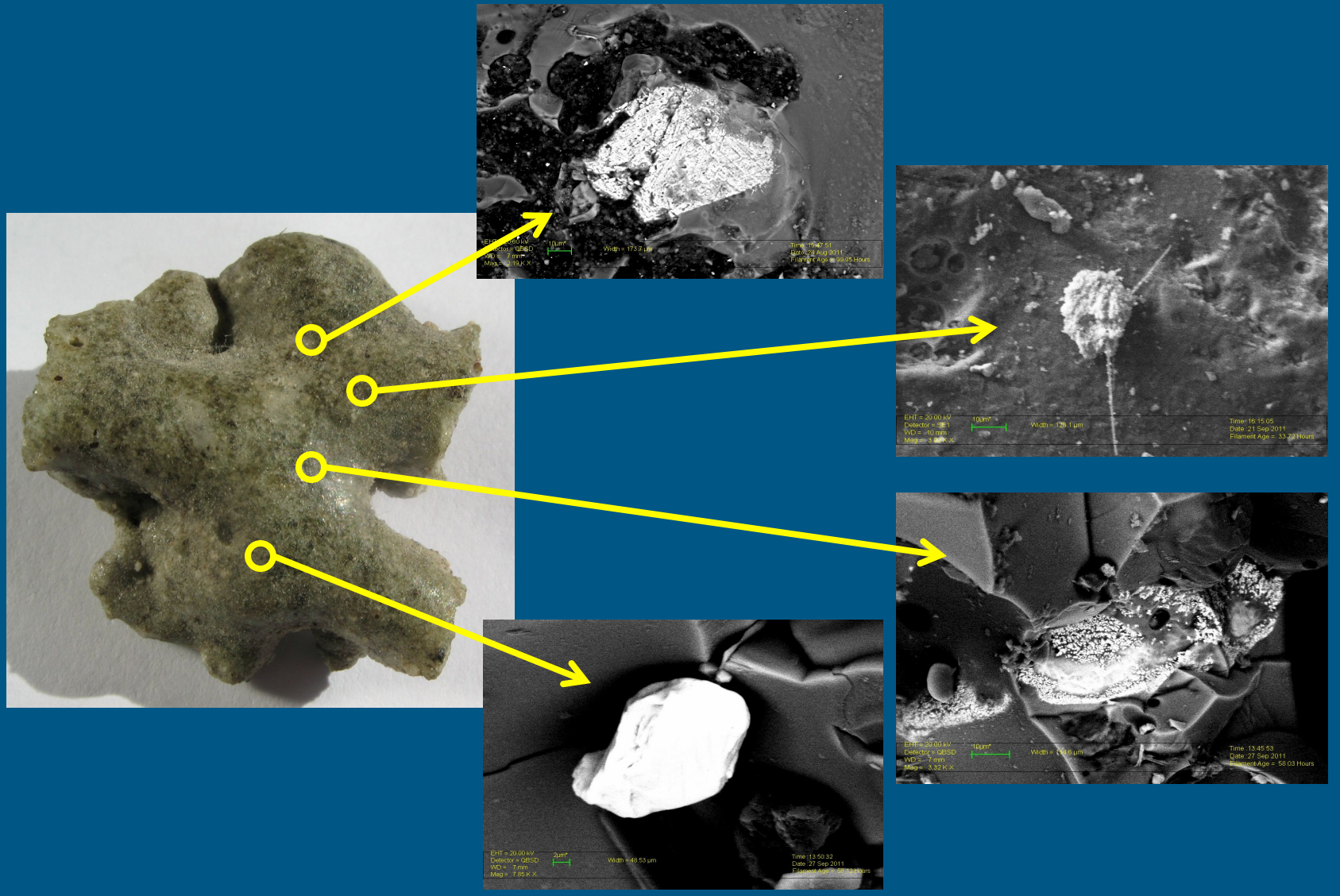
Gamma Spectroscopy – Conclusions

- Based on the relative activities of ^{137}Cs, ^{155}Eu, ^{239}Pu, and ^{241}Am, a similar behavior of these isotopes during the Trinity test is observed.
- The behavior of ^{133}Ba does not correlate with any of the bomb-derived isotopes, and therefore, its exact origin remains ambiguous.
- Based on the activity of ^{152}Eu, a spatial model calculation for the radioisotopes indicates their homogeneous distribution.

Bellucci et al. (2013a, JNRC)

Detailed, In-situ Microanalysis

SEM Investigation of Inclusions – Trinitite Surfaces



Bellucci & Simonetti (2012, *J. Radioanalytical Nuclear Chemistry*)

TRINITITE-HOSTED INCLUSIONS

Origin – desert sand

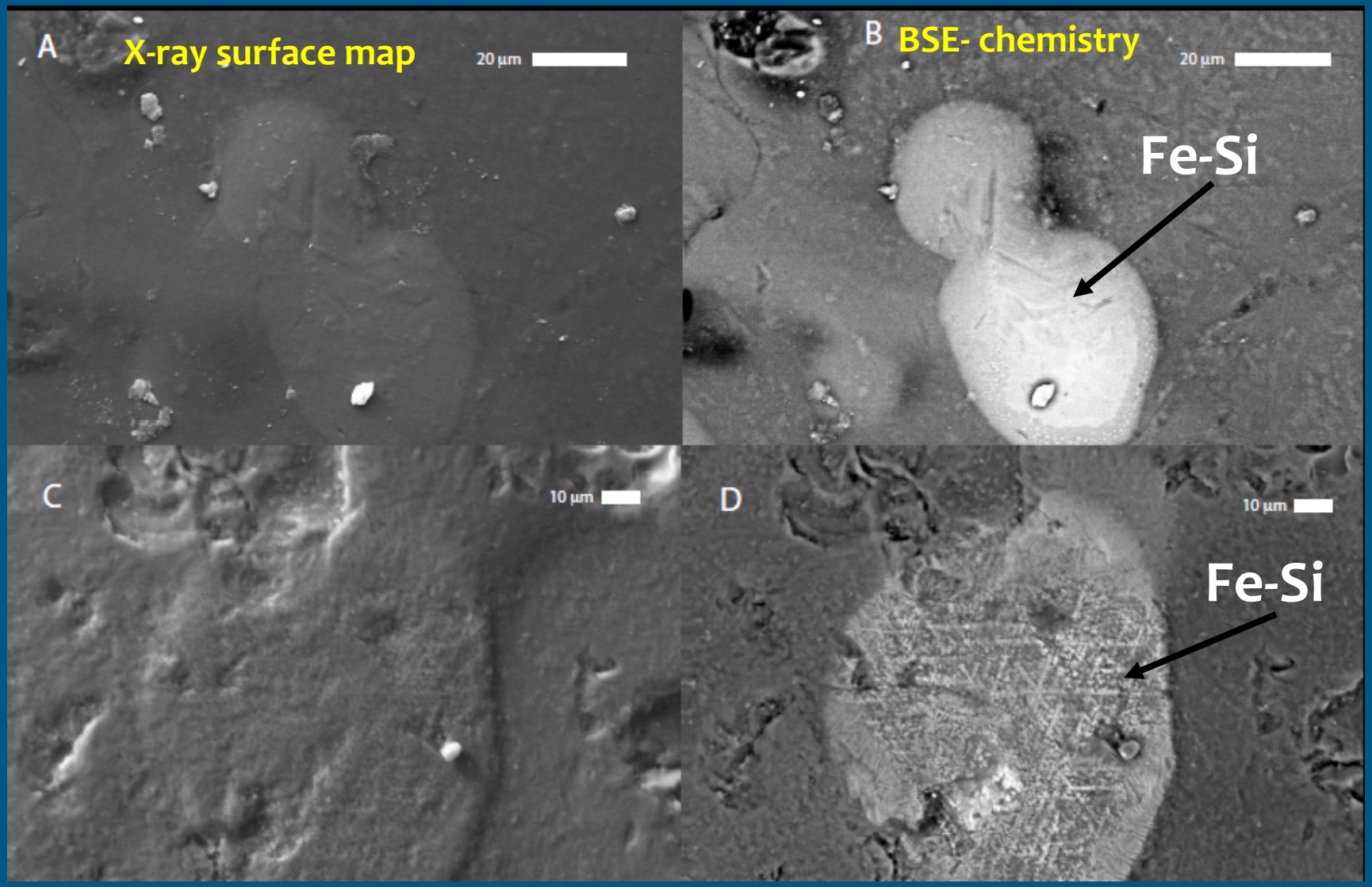
Trinitite samples and inclusions.

Sample	FeO	Iron-Silicate	Fe-Ti	Ti	PbO	W	Zircon	CuS	BaS	Qtz Glass	Plagioclase Glass	Hi-Ca-Glass
1	x	x									x	x
2		x			x	x	x				x	
3		x	x	x			x		x		x	
4A		x	x					x		x	x	
4B	x	x	x					x			x	
4C		x	x				x				x	
4D											x	x
4E		x									x	
4F	x	x								x	x	x
5A		x	x				x				x	x
5B		x					x				x	
5D		x	x				x			x	x	
5E	x	x	x				x				x	x
5F	x	x					x				x	

Origin – gadget device & materials, blast tower,

Bellucci & Simonetti (2012, *J. Radioanalytical Nuclear Chemistry*)

TRINITITE-HOSTED INCLUSIONS



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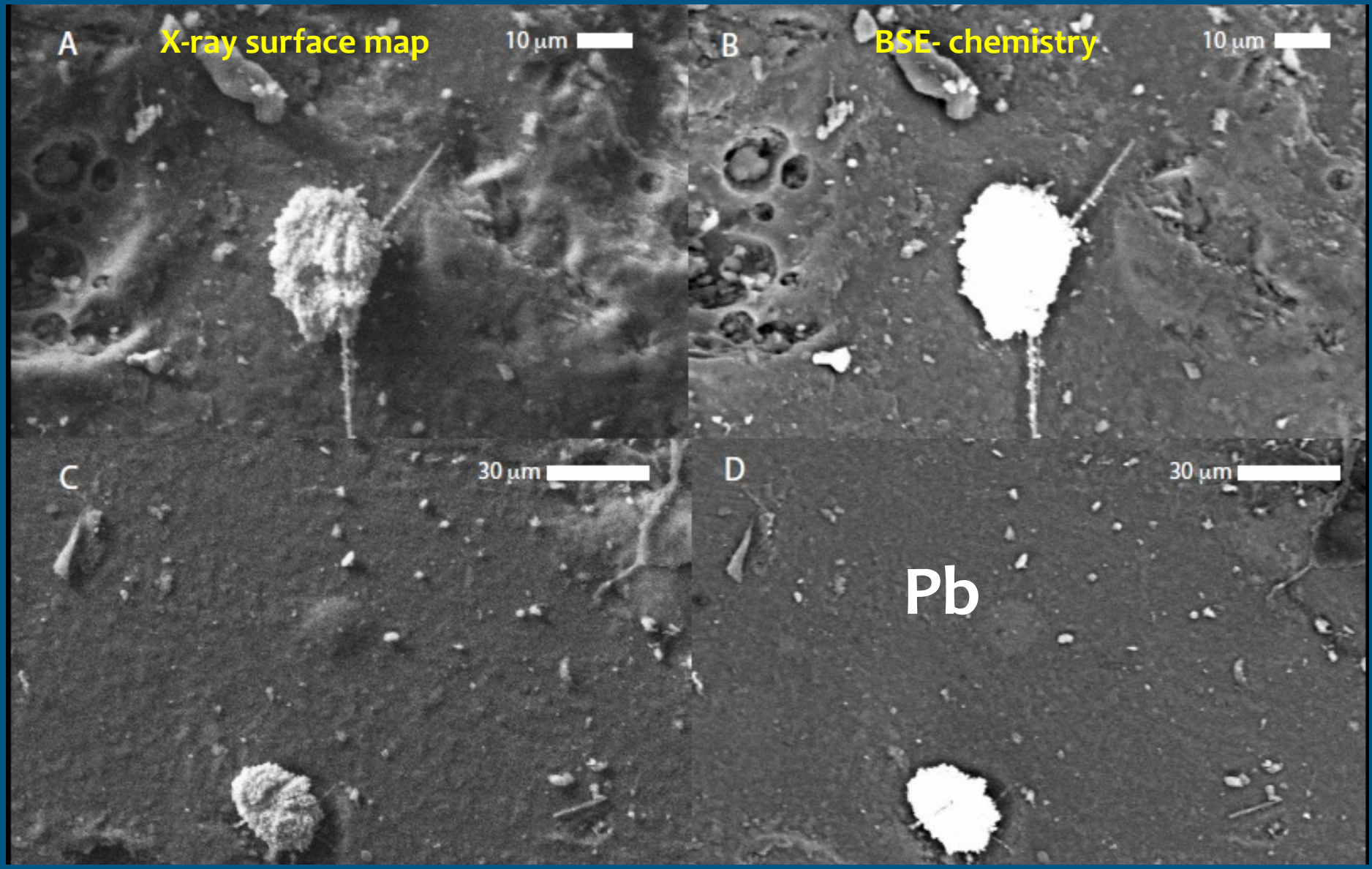

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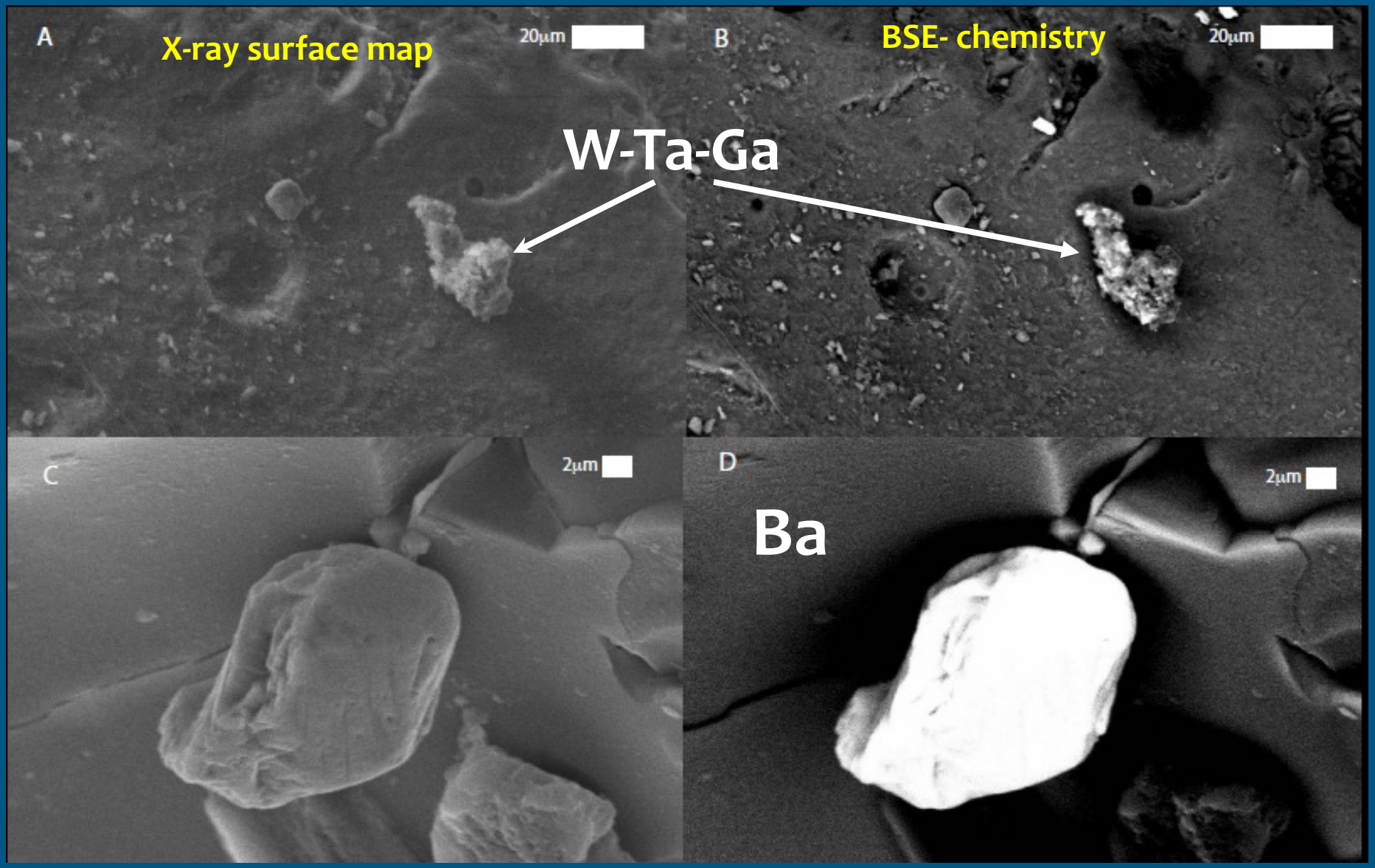

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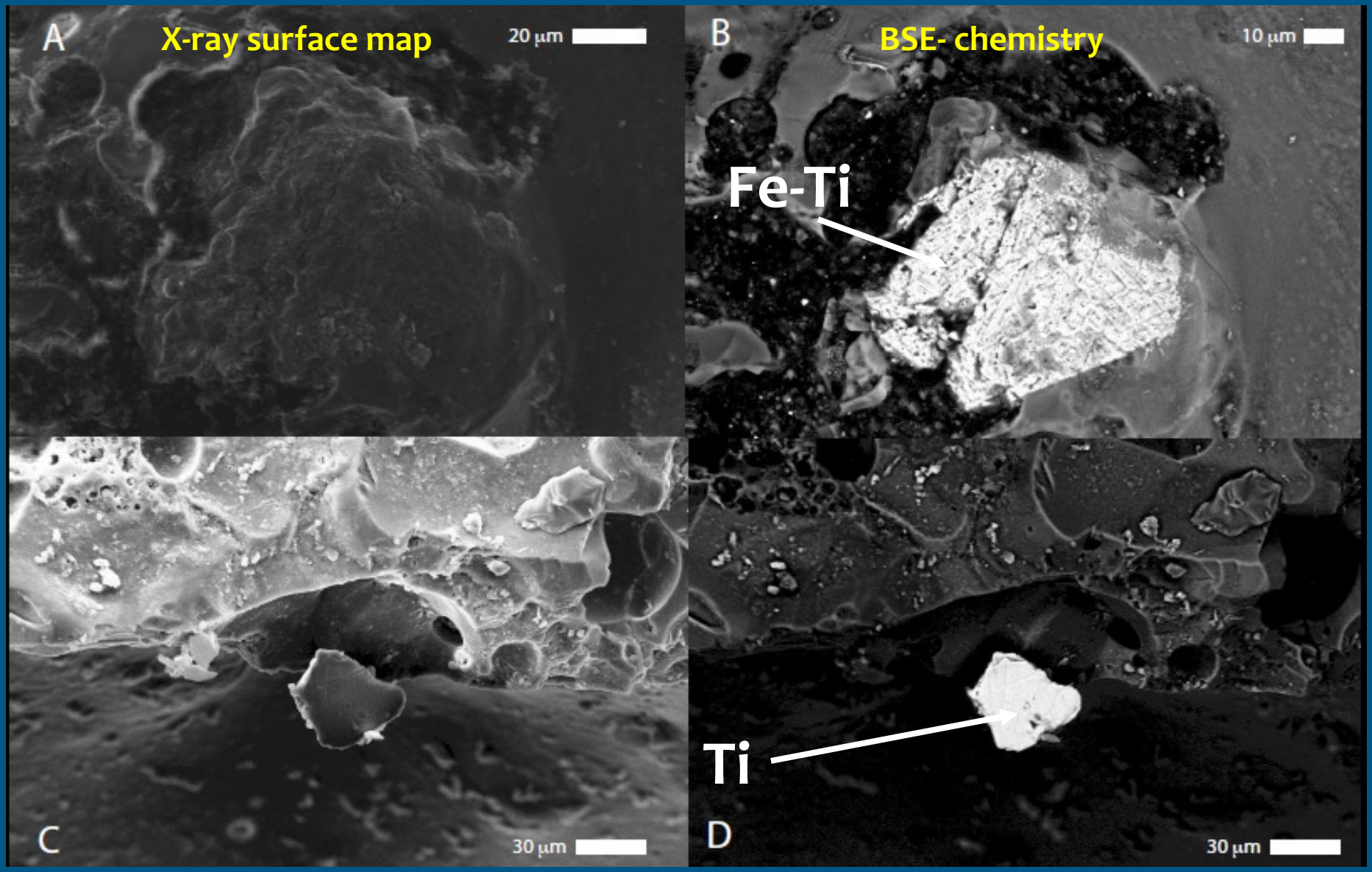


Bellucci & Simonetti (2012, *J. Radioanalytical Nuclear Chemistry*)

Blast modeling – Inclusion work
Two-stage process

Bellucci & Simonetti (2012, *J. Radioanalytical Nuclear Chemistry*)

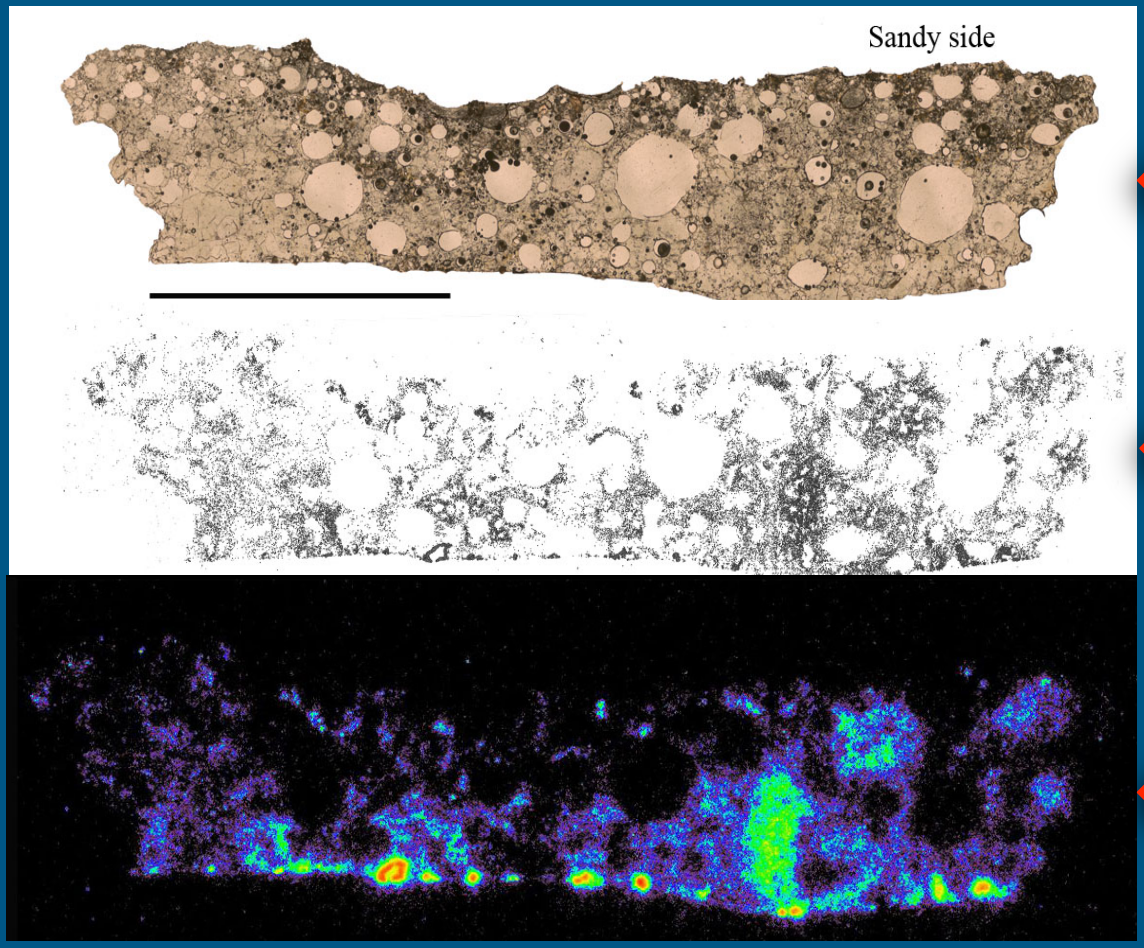
Conclusions – Trinitite-hosted inclusions

- **RELIEF:**
  – "Flat" or "Bell-shaped" inclusions formed simultaneously with main phase of blast melt
  - "Topographic" inclusions precipitated ("rained down") later

- **ORIGIN of ELEMENTS:**
  - Fe-Ti – blast tower
  - Pb, Ta, W – tamper of device
  - Ga – alloyed with Pu during enrichment process
  - Ba – device + natural

Bellucci & Simonetti (2012, *J. Radioanalytical Nuclear Chemistry*)

Distribution of Radioactive Elements- Micron scale

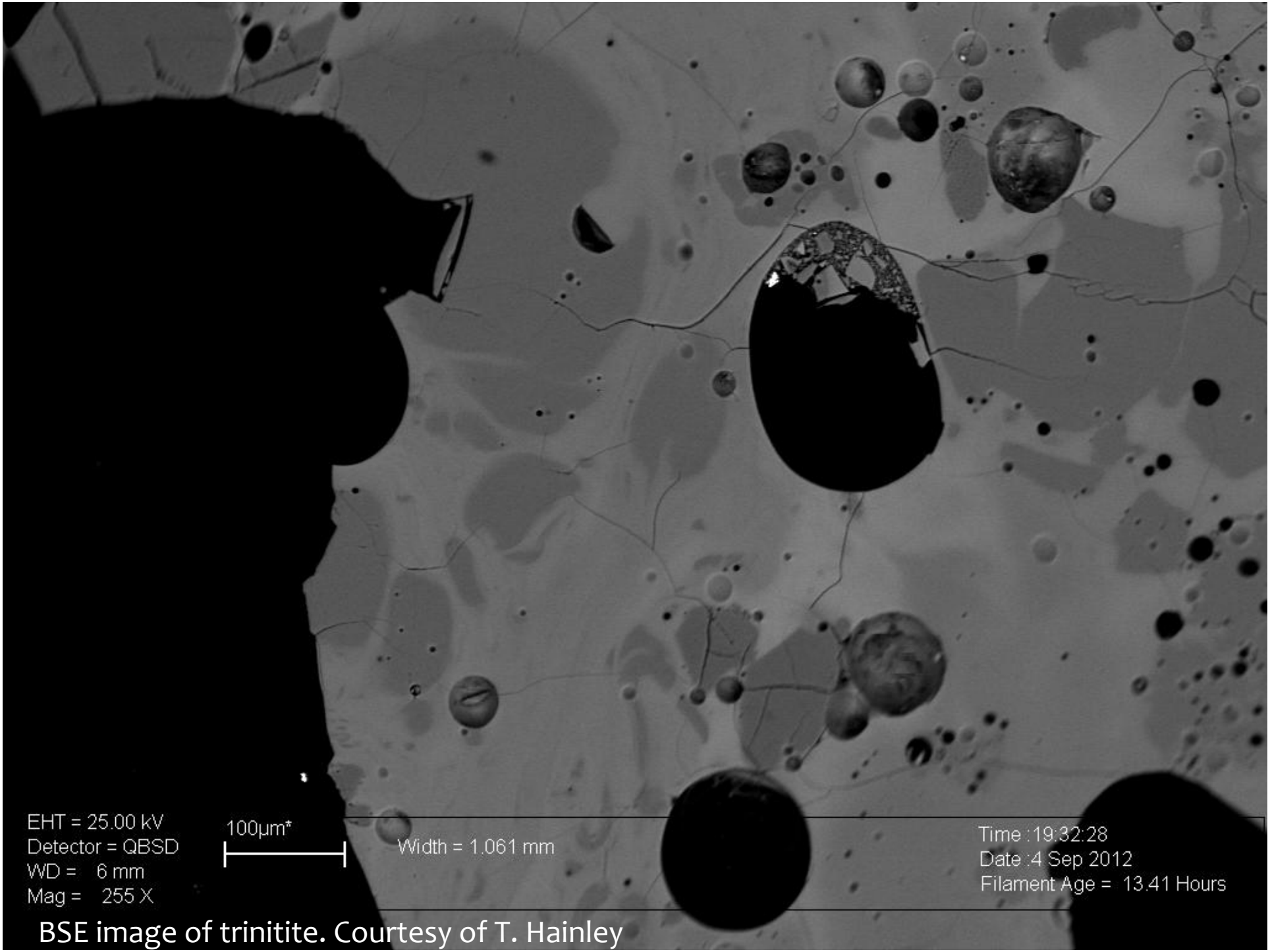


Optical map (PPL)

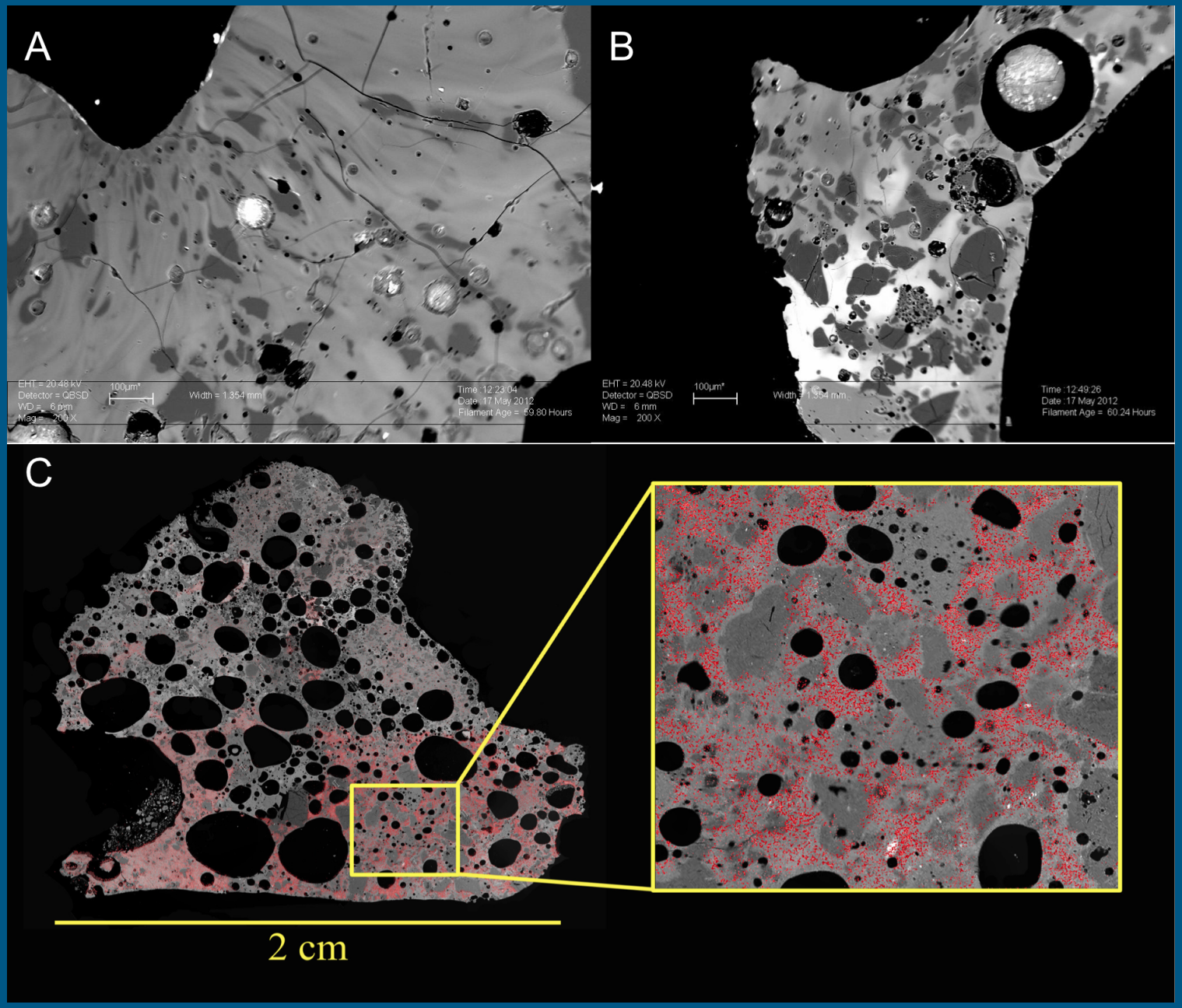
Alpha track radiography

Beta autoradiography

Wallace et al. (2013, JRNC)



BSE image of trinitite. Courtesy of T. Hainley



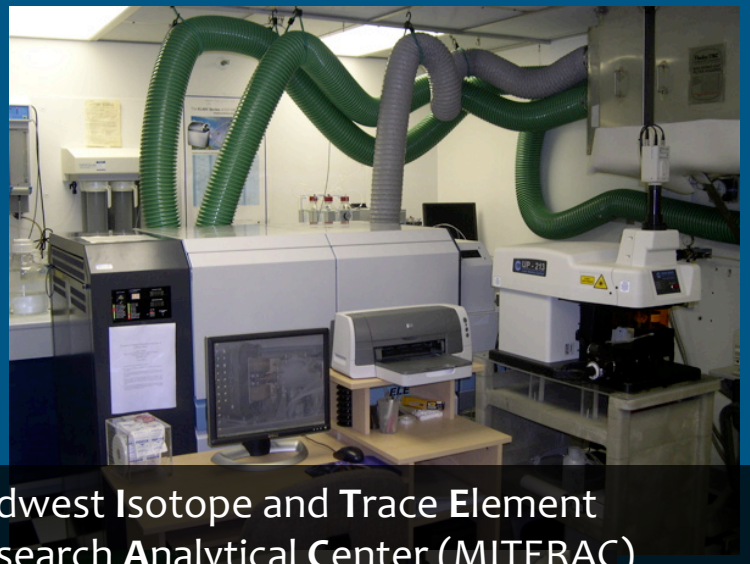
Wallace et al. (2013, JRNC)

Trace Element Analysis

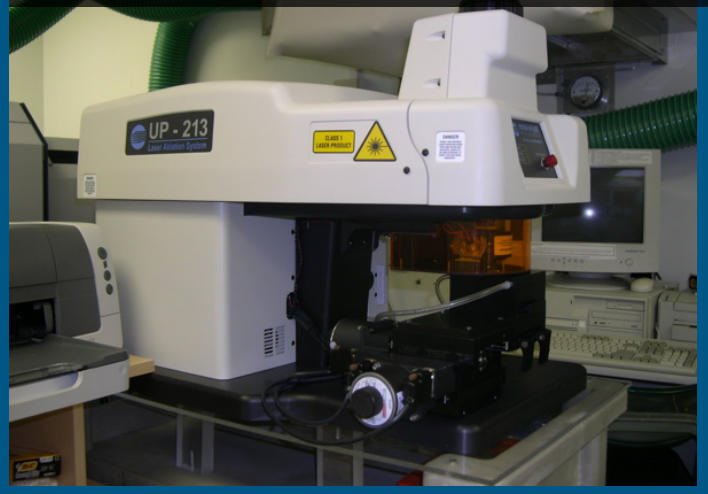
- Laser Ablation ICP-MS

- Parameters:
  - Standard: NIST 612
  - Spot Size: 55µm
  - Frequency: 5 Hz
  - Fluence: ~12 J/cm²

- New Wave Research UP-213 Nd:YAG laser & Element2 HR-ICP-MS



Midwest **I**sotope and **T**race **E**lement **R**esearch **A**nalytical **C**enter (MITERAC)



LA-ICP-MS Analyses - Radionuclides

Table 2 Isobaric mass interferences monitored and corrections applied

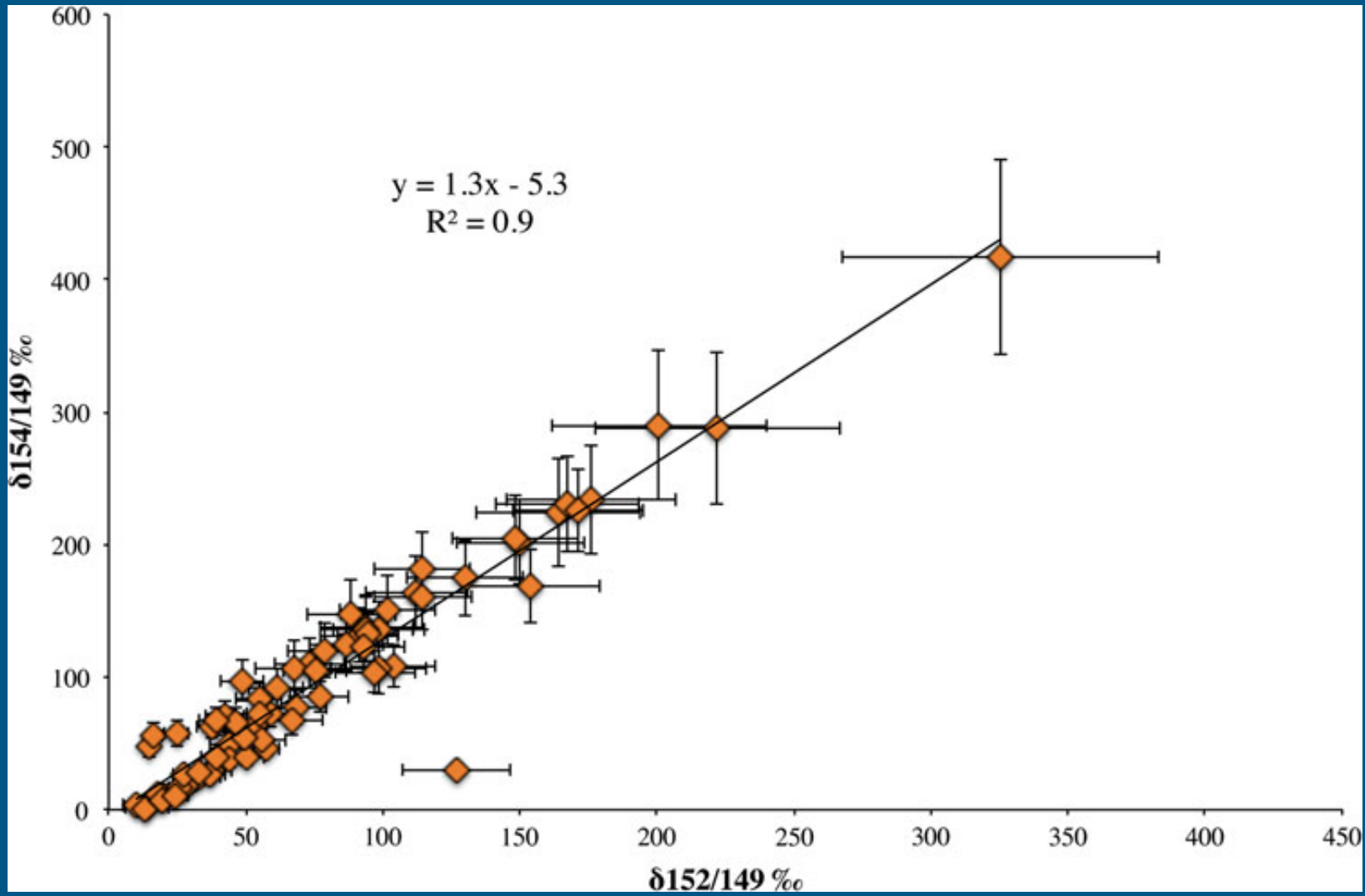
Mass	136	137	138	147	149	151	152	153	154	160	163
	Ba	Ba	Ba								
				Sm	Sm[b]	Eu	Sm	Eu	Sm		
							Gd		Gd	Gd	
							$^{136}Ba^{16}O$[a]		$^{138}Ba^{16}O$[a]	Dy	Dy

[a] Plasma oxide level (typically <1 %) was determined by comparison between total (measured) ion signal (cps) intensities for masses 152 and 154 and those calculated for 152Gd and 154Gd using natural atomic abundances of 160Gd and 163Dy based on laser ablation analyses of the NIST SRM 612 glass standard

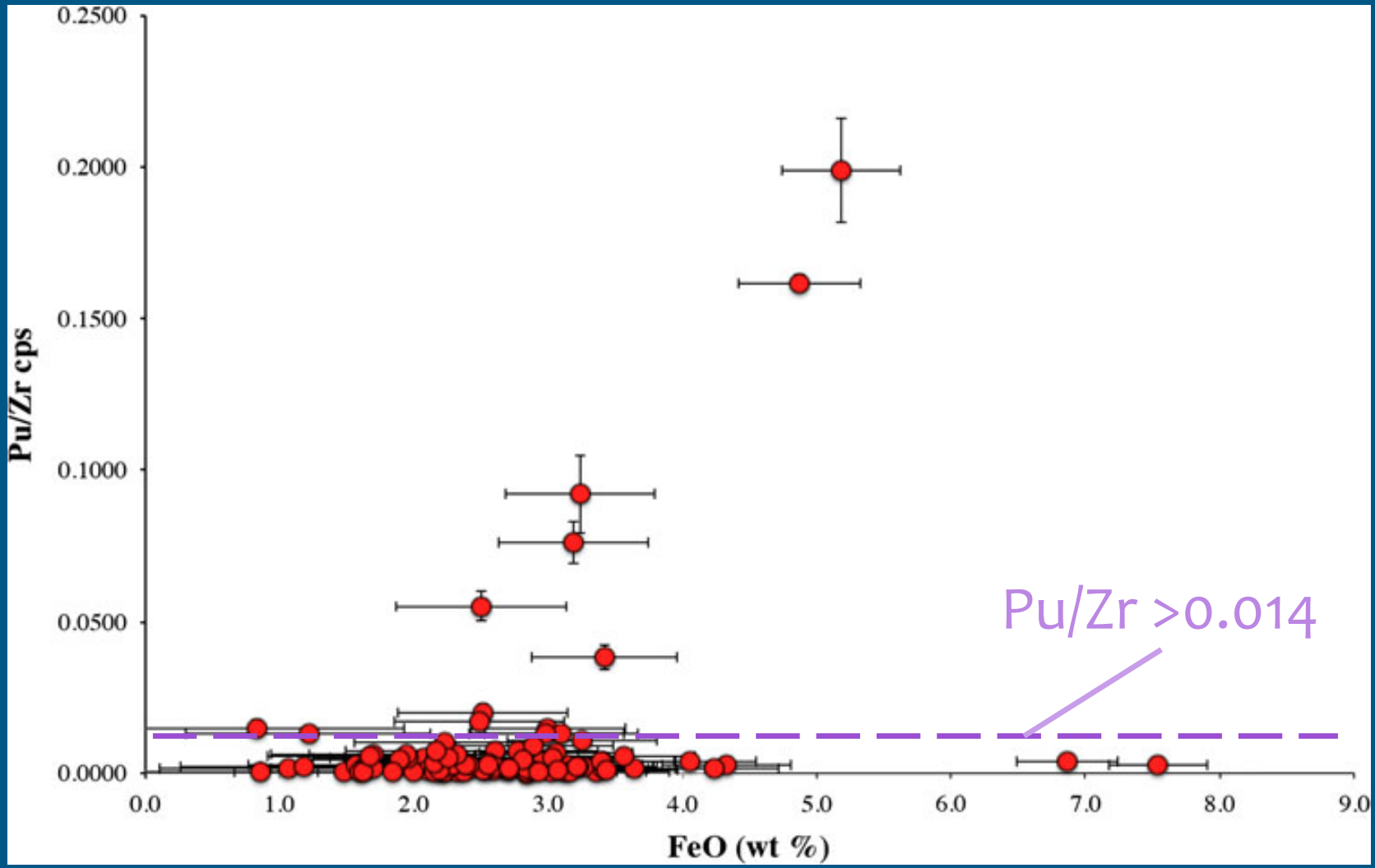
[b] Calculated ion signal intensities (cps) for 152Sm and 154Sm were based on natural abundance isotope ratios for Sm using the measured, interference-free ion signal for 149Sm. Excesses in absolute ion signal intensities (cps) calculated on masses 152 and 154 for laser ablation analyses of trinitite are attributed to the presence of activation products 152Eu and 154Eu, respectively

$$\delta^{a}X = \left({}^{a}R_{\text{sample}} - {}^{a}R_{\text{standard}} / {}^{a}R_{\text{standard}}\right) \times 1000.$$

Wallace et al. (2013, JRNC)



Wallace et al. (2013, JRNC)



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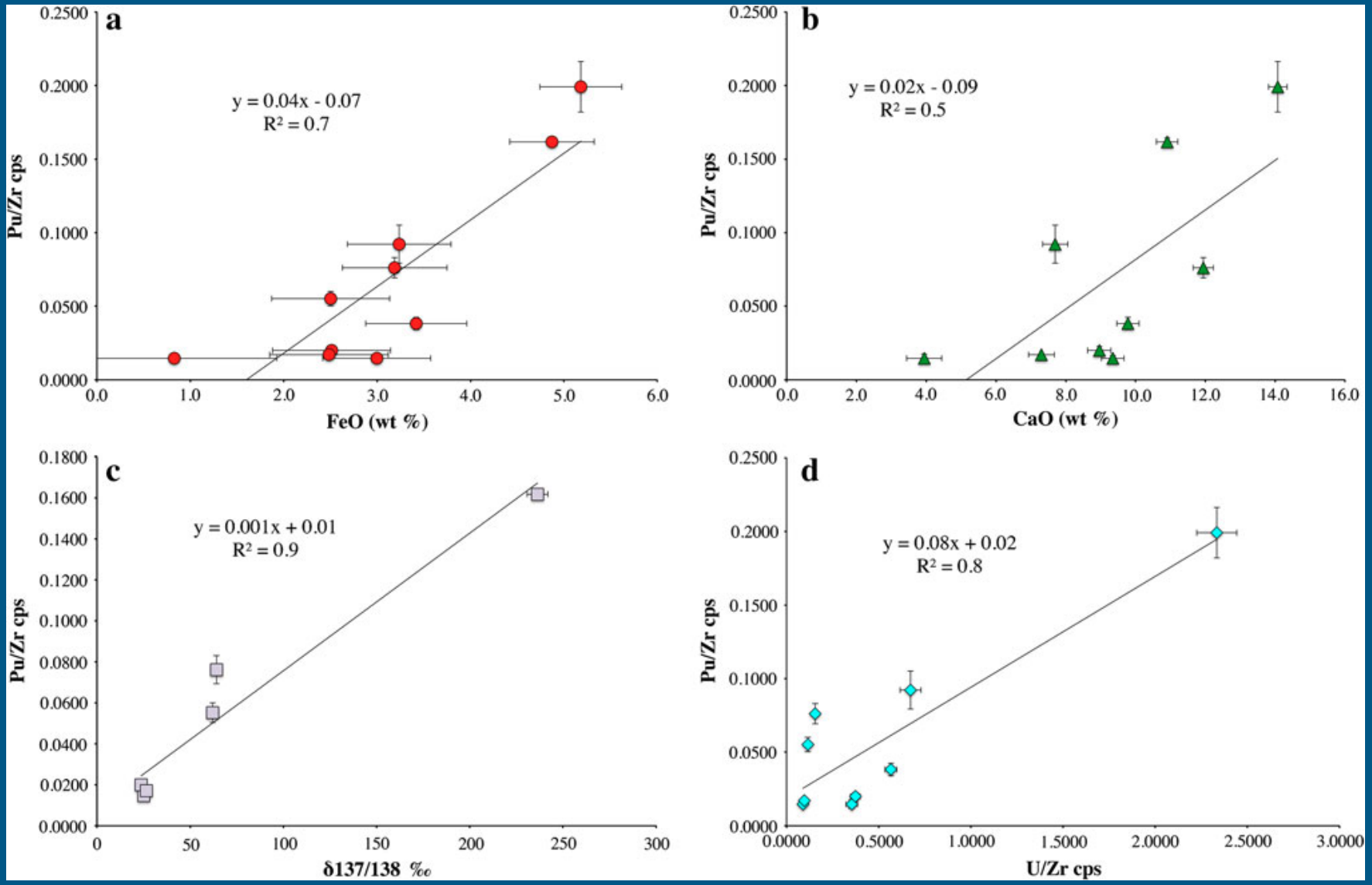

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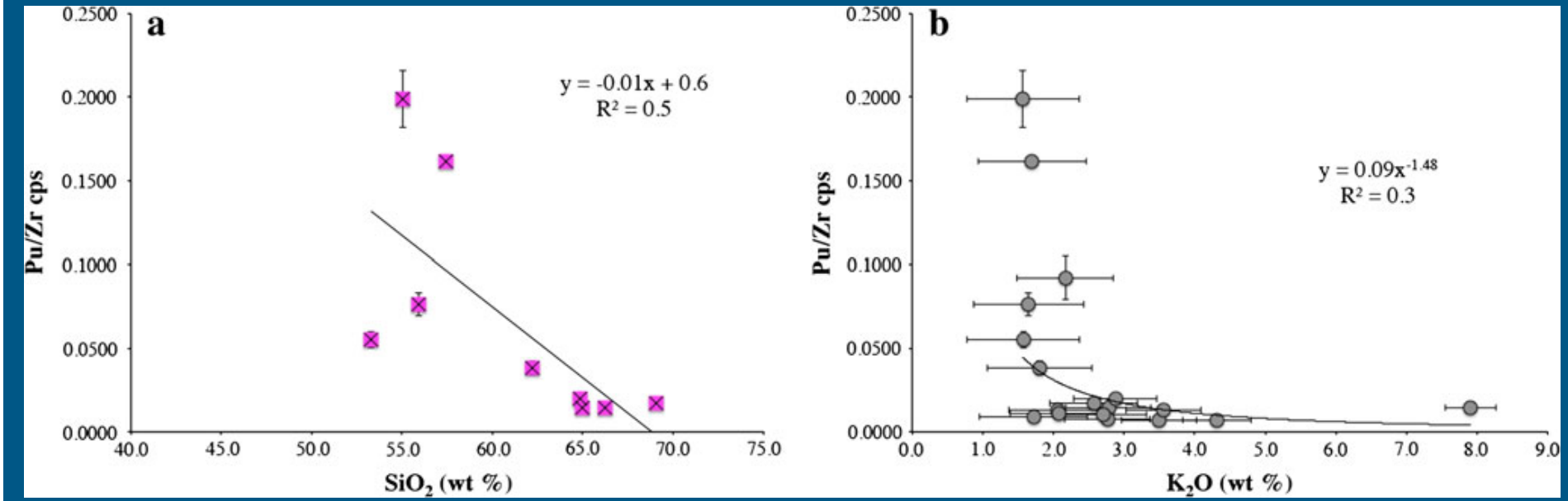
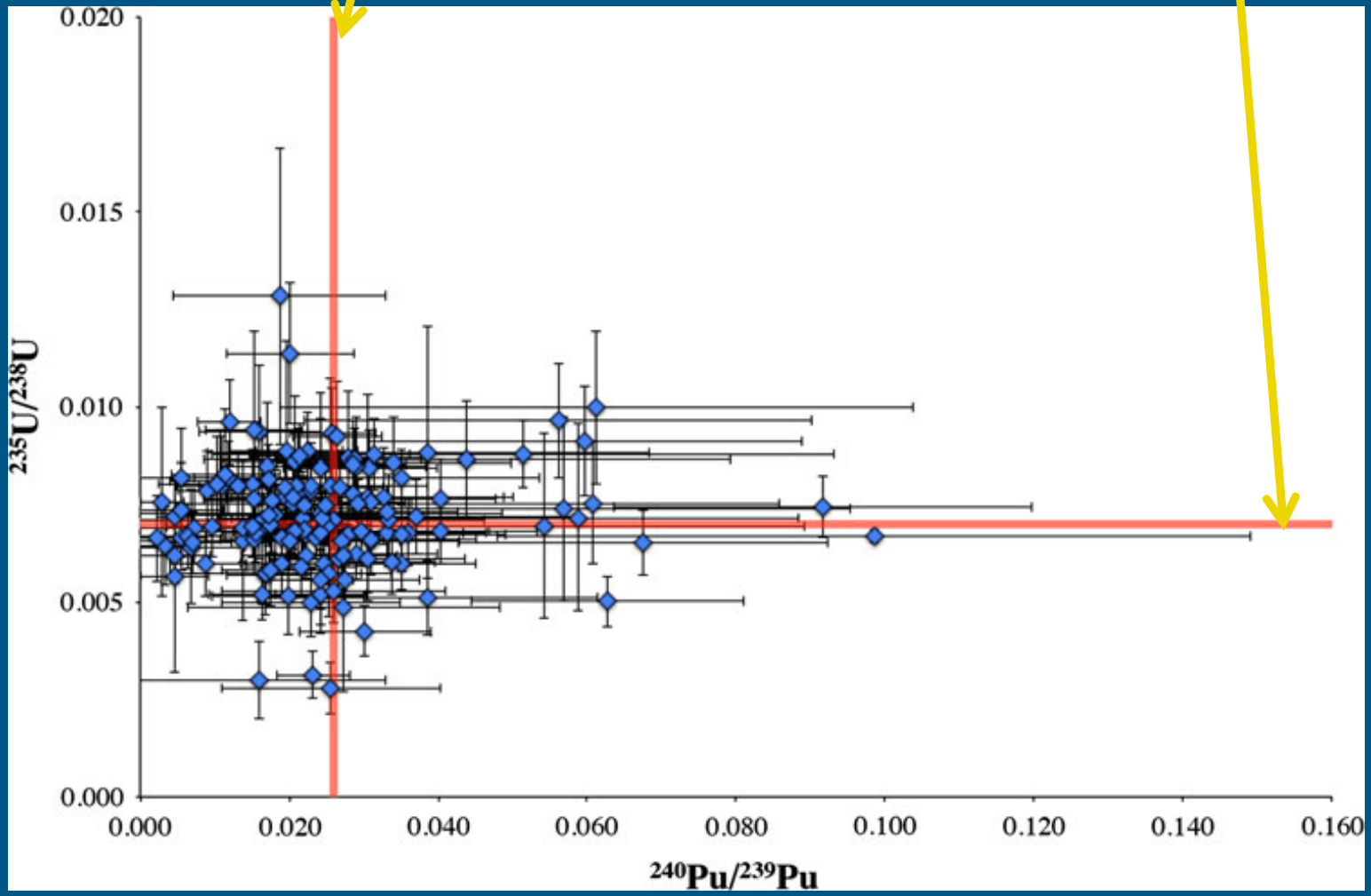


Wallace et al. (2013, JRNC)

$^{240}Pu/^{239}Pu = 0.0208 \pm 0.0012$

$^{235}U/^{238}U = 0.00718 \pm 0.00018$



Wallace et al. (2013, JRNC)

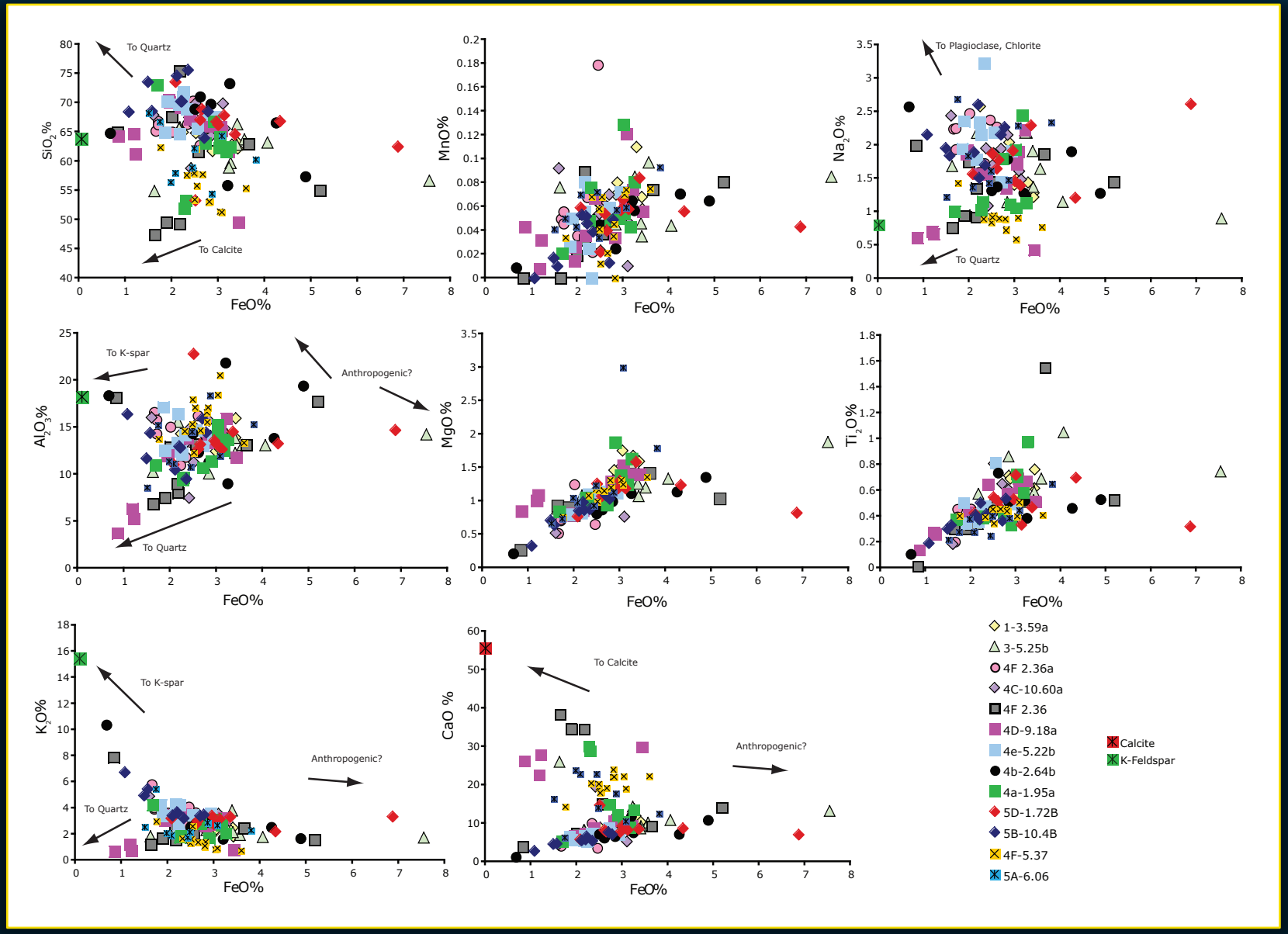
Distribution of Radioactive Elements – Conclusions

- Demonstrate for the first time that device-related radionuclides (e.g., U, Pu) are found primarily within the melt (glassy) component of trinitite.
- In areas characterized by higher Pu ion signals (i.e., abundances), these also contain elevated contents of U and fission products (e.g., ^{137}Cs), which confirm their association with the device.
- In contrast, crystalline (relatively intact, precursor) mineral phases, such as quartz, K-feldspar, are essentially devoid of radionuclides and other device-related components.

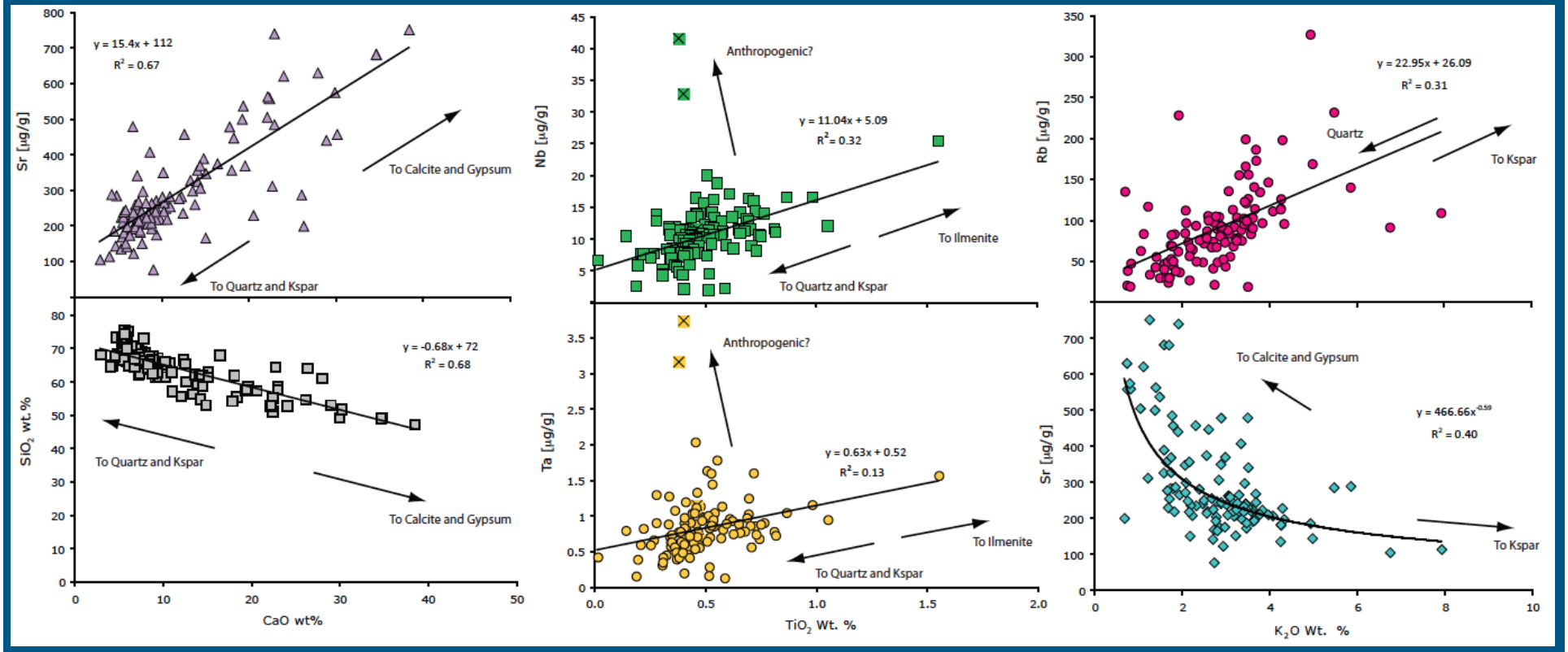
Wallace et al. (2013, JRNC)

Distribution of Major & Trace Elements

- Can the distribution of major and trace element abundances for Trinitite obtained in-situ by Electron Microprobe & LA-ICP-MS be used to decipher natural vs. anthropogenic (device-related) components?
- 13 Trinitite samples were investigated – total of 117 LA-ICP-MS analyses



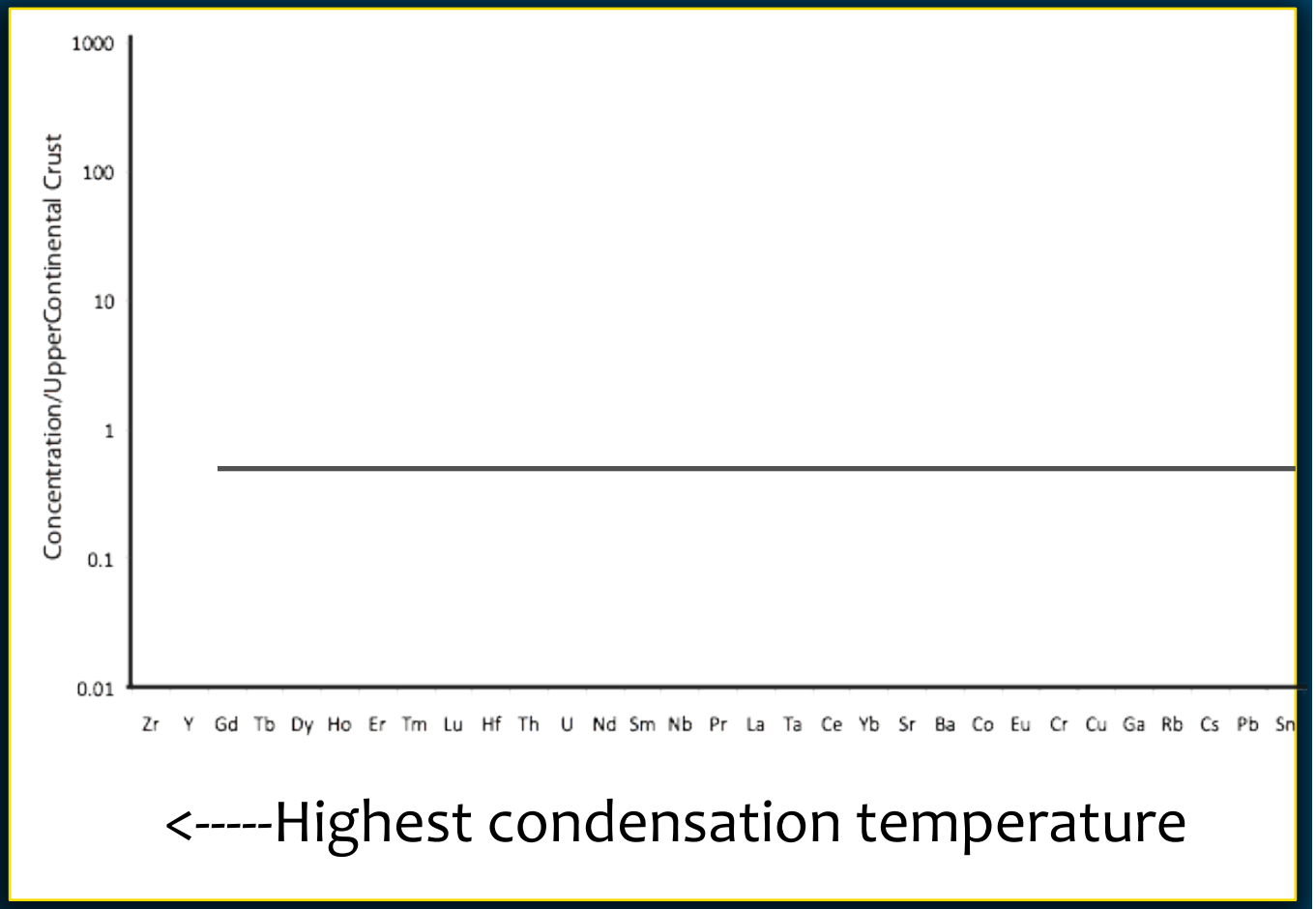
Bellucci et al. (in press, Chemical Geology)



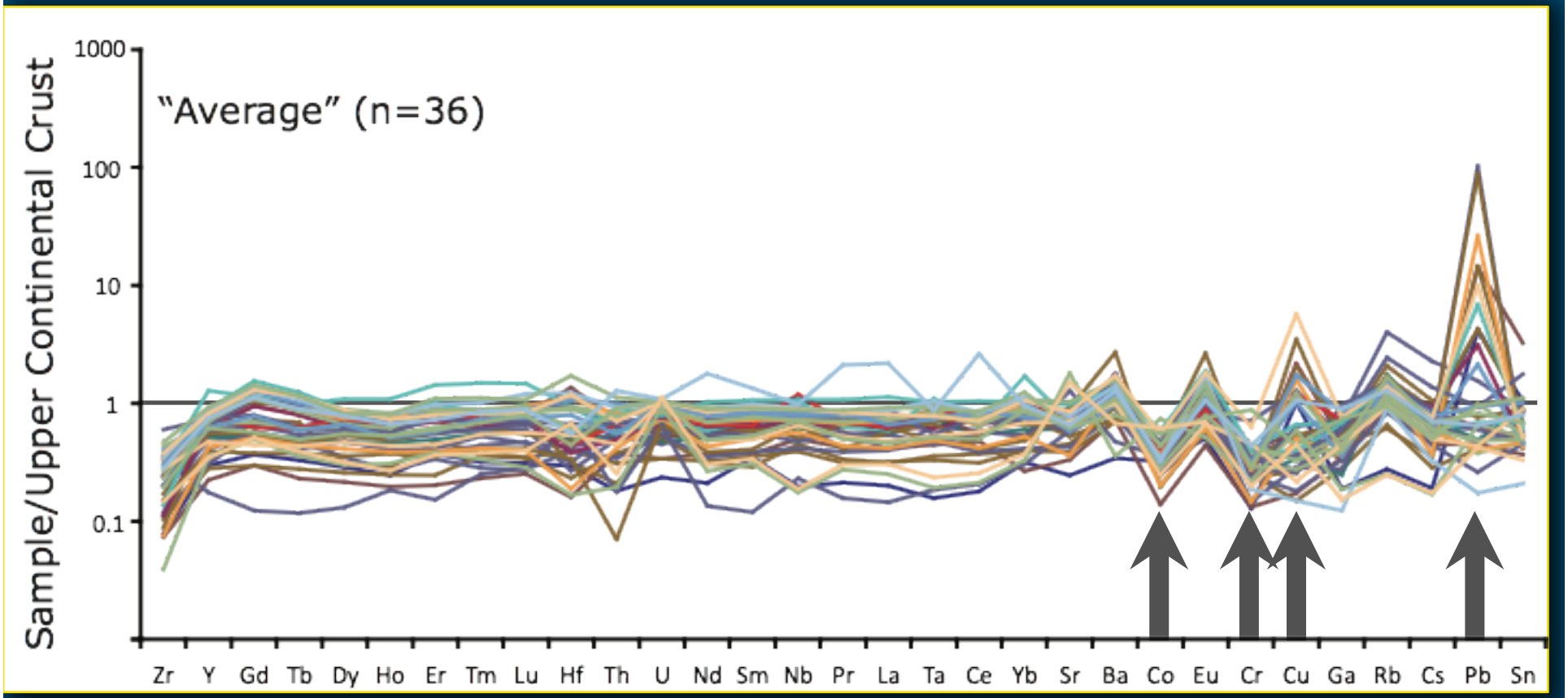
Bellucci et al. (in press, Chemical Geology)

Trace Elements Patterns

- Normalize to upper continental crust
- Looking for anomalous compositions
- Order by condensation temperature

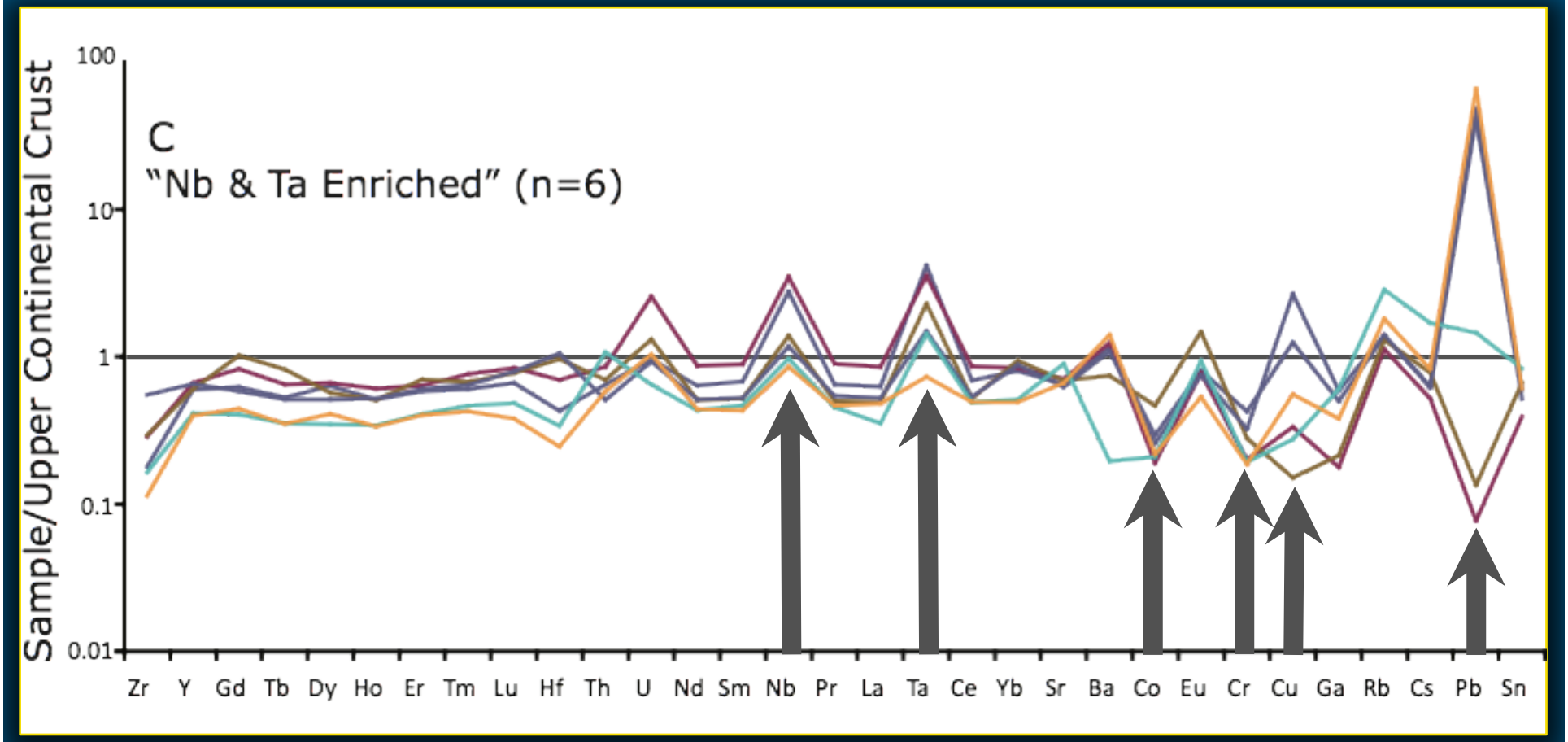


Samples with no clear mineral enrichments



Bellucci et al. (in press, Chemical Geology)

Nb and Ta: Enriched Anthropogenic



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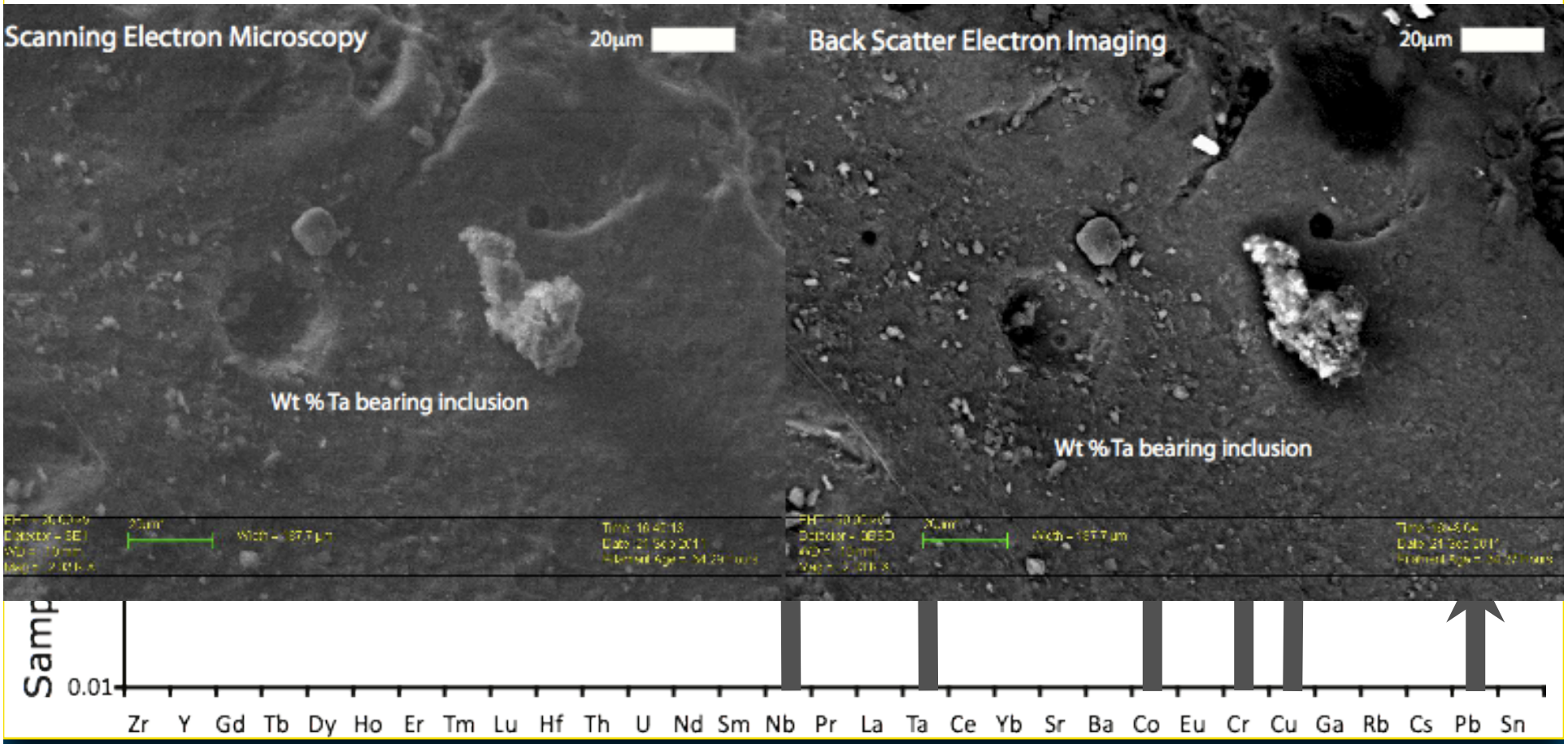
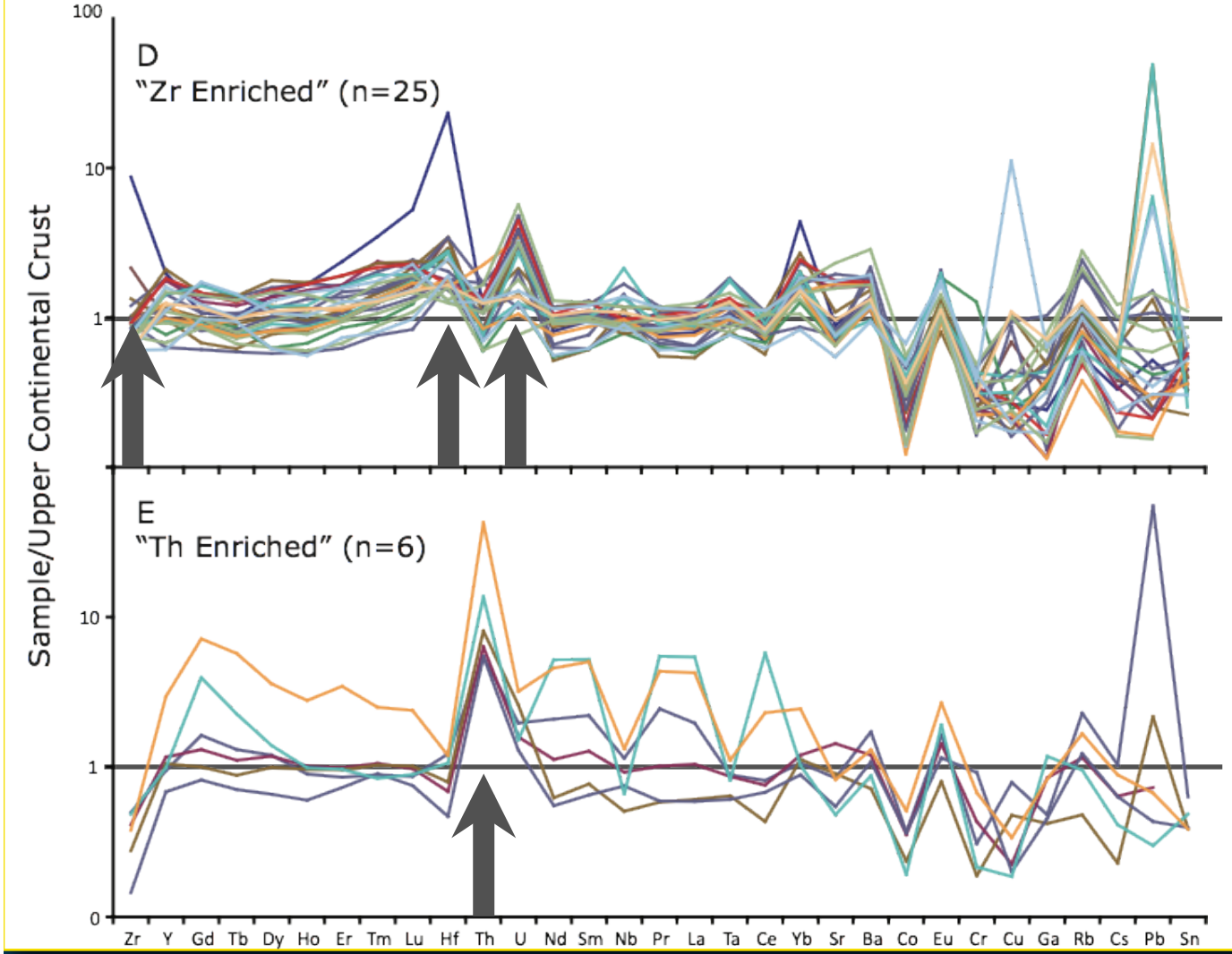
Nb and Ta: Enriched Anthropogenic



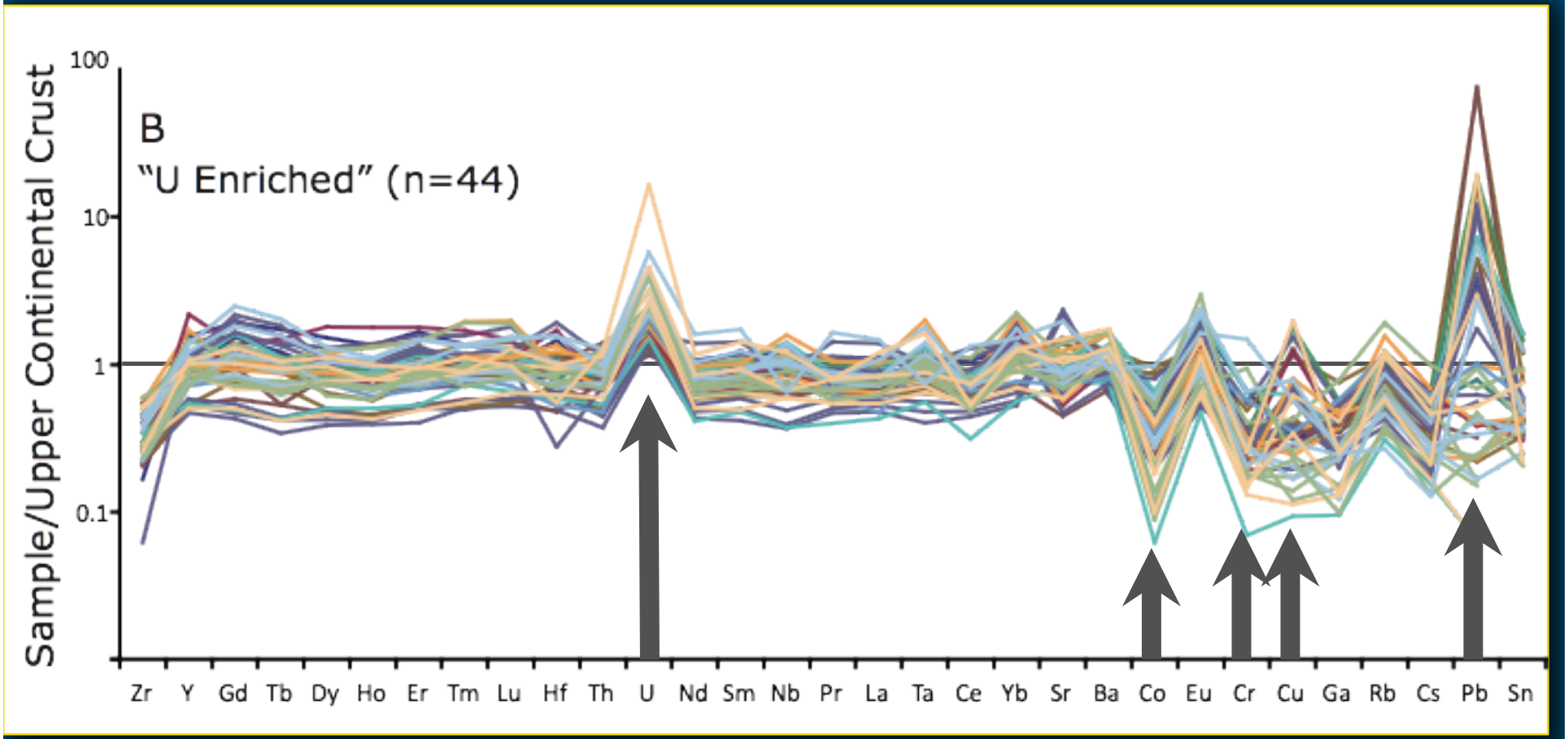
Bellucci et al. (in press, Chemical Geology)

Natural Enrichments



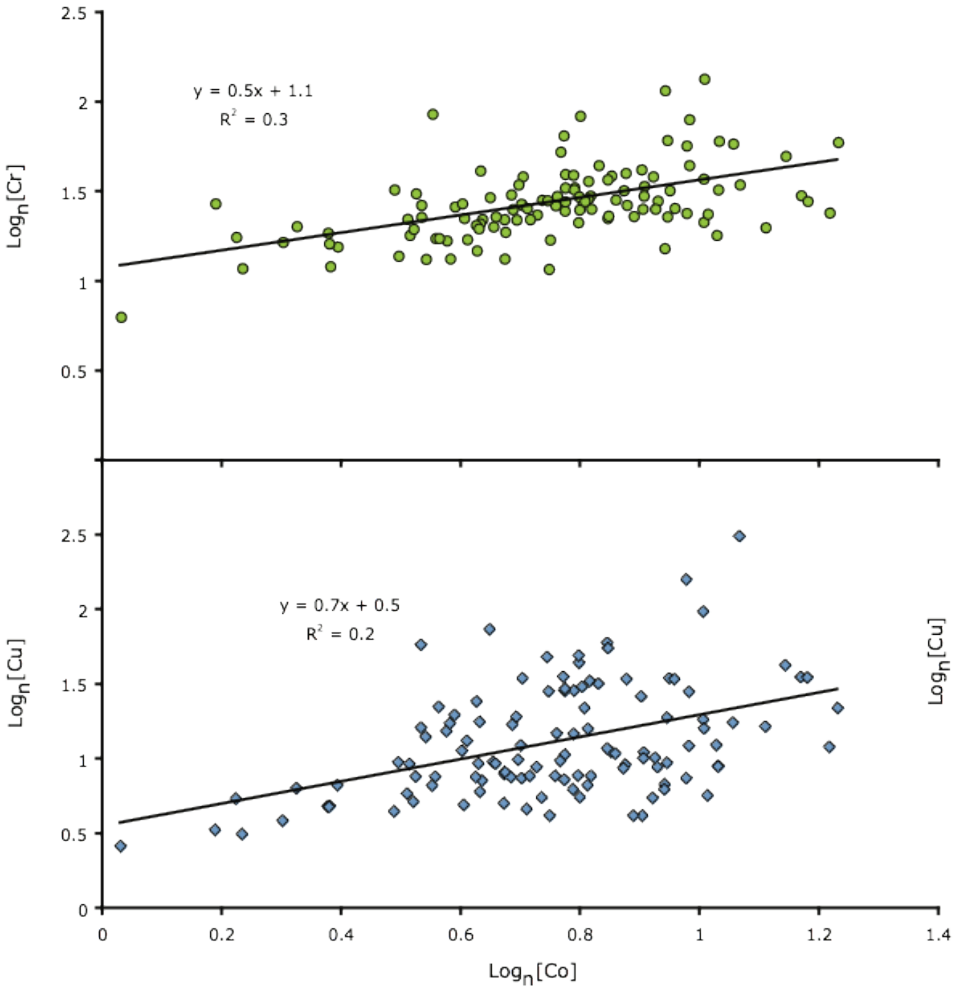
Bellucci et al. (in press, Chemical Geology)

U enrichment without geologic indicators

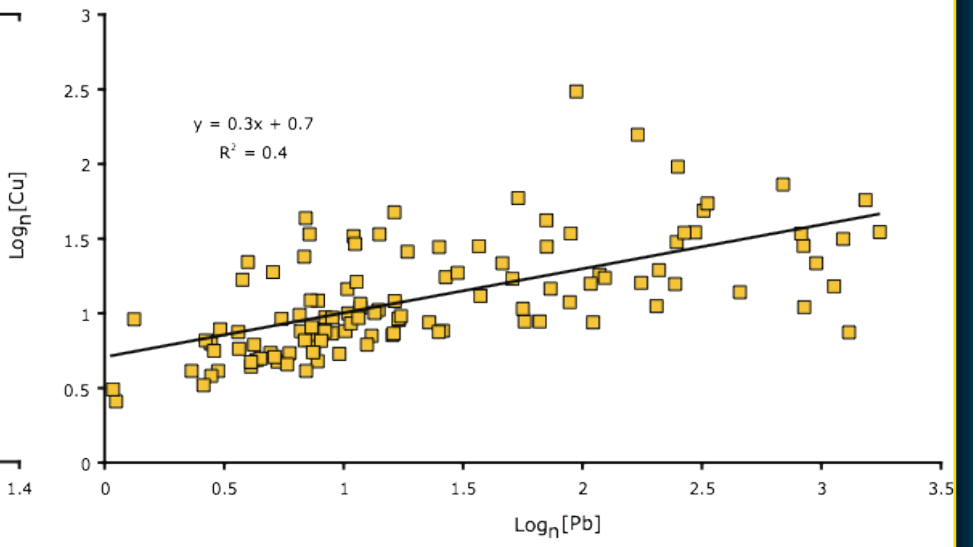


Bellucci et al. (in press, Chemical Geology)

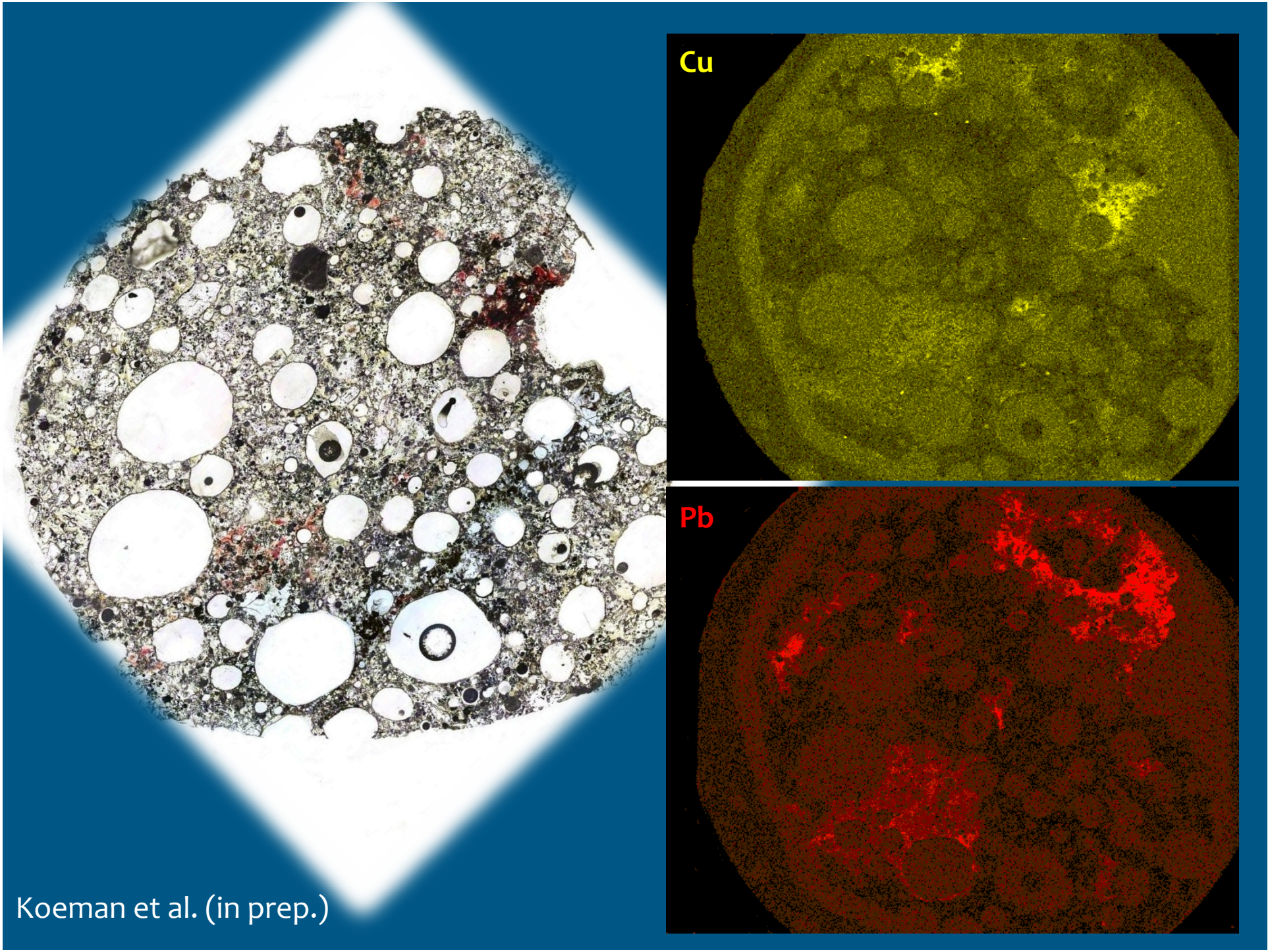
Metals define linear correlations



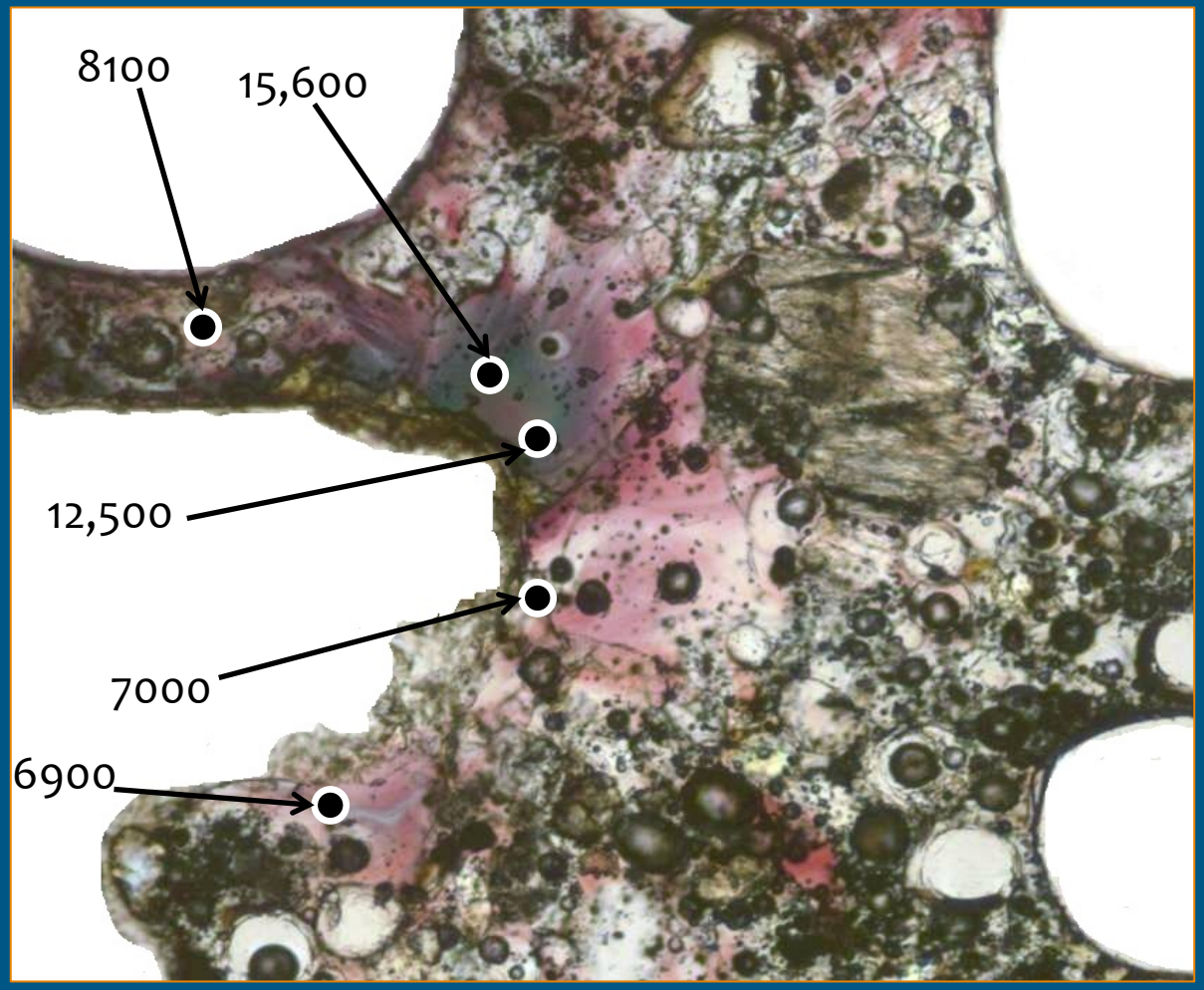
Indicate mixing between sand and likely materials from the bomb tower



Bellucci et al. (in press, Chemical Geology)


Cu
Pb
Koeman et al. (in prep.)

Lead Concentrations (ppm)



FOV 2.5mm

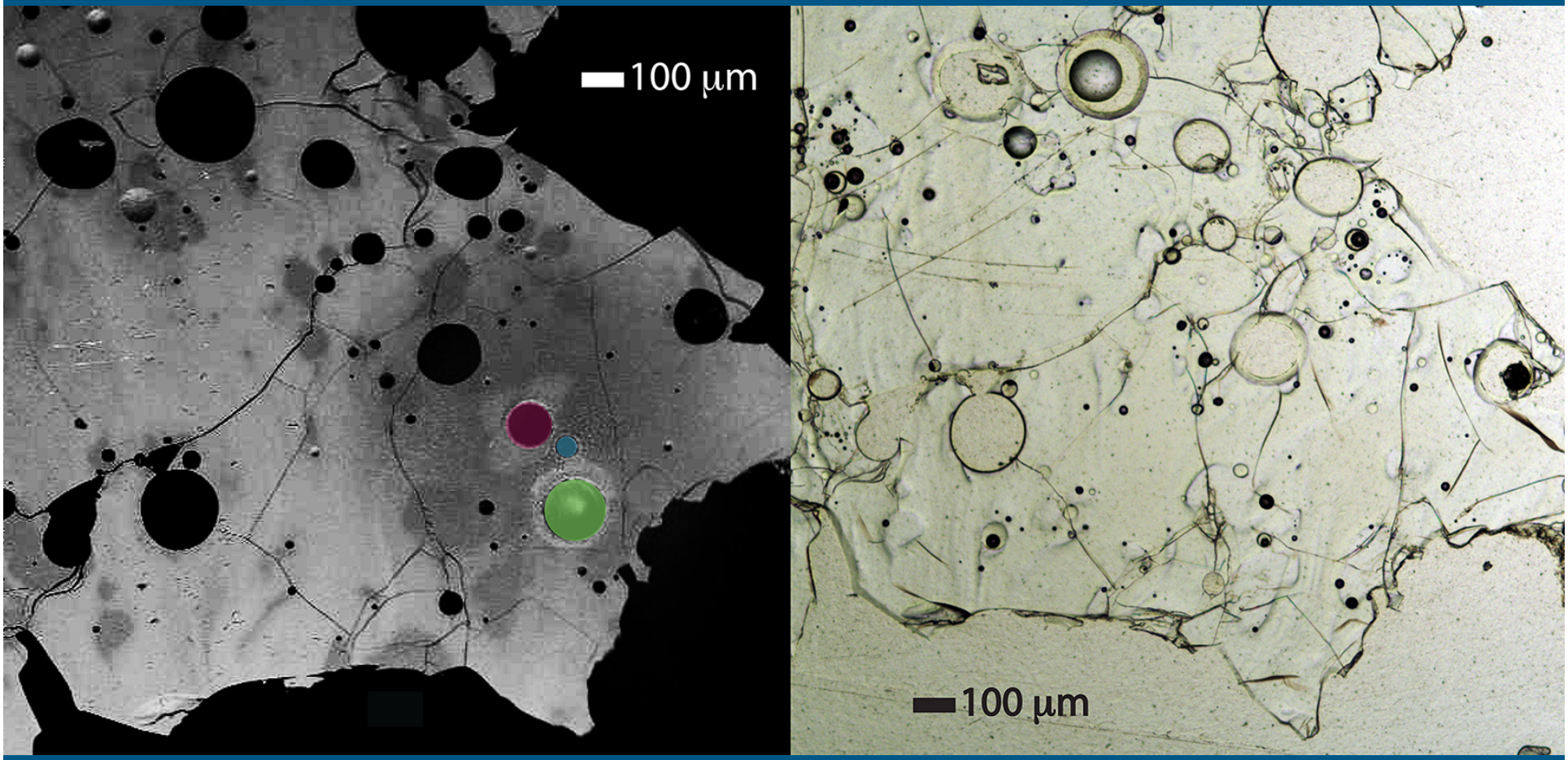
Origins of Trace Elements

- Most trace elements can be attributed to precursor minerals within desert sand:
  - Calcite/Gypsum: Sr
  - Barite: Ba
  - K-feldspar: Cs, Rb, Ga
  - Ilmenite: Nb, Ta
  - Apatite, Monazite, Zircon: U, Th, Y, Hf, REEs

  - Except for metals: Nb, Ta, Cu, Co, Cr, Pb
  - Some U is not from natural background

Bellucci et al. (in press, Chemical Geology)

U & Pb isotope compositions of Trinitite



Bellucci et al. (2013b, Analytical Chemistry)

Uranium isotopes

- ^{238}U
- ^{236}U: Produced by neutron capture of un-fissioned ^{235}U and decay of ^{240}Pu
- ^{235}U: Produced by decay of ^{239}Pu
- ^{234}U: Produced by decay of ^{238}Pu
- All U have half-lives that are long enough to make them appear stable since the time of detonation

Bellucci et al. (2013b, Analytical Chemistry)

Pu-Isotope Systematics

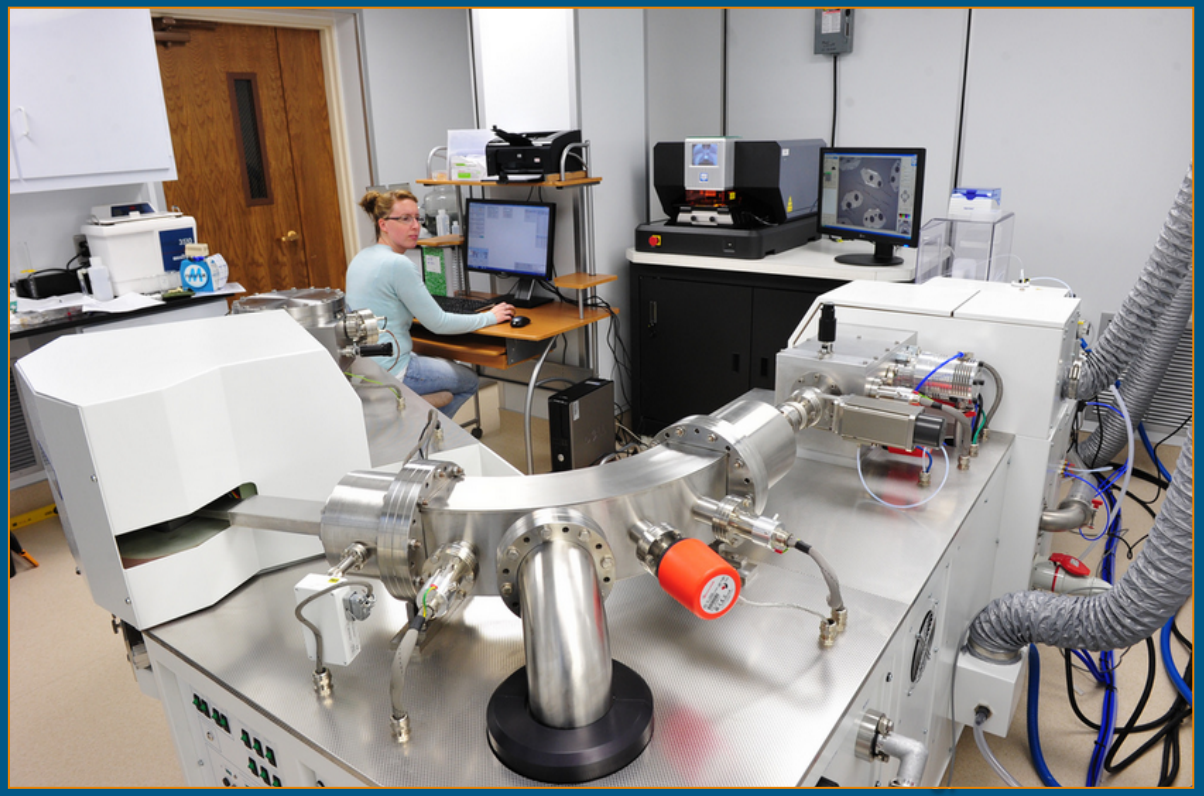
- $^{240}Pu \rightarrow ^{236}U$
  - Half life: 6,560 y
- $^{239}Pu \rightarrow ^{235}U$
  - Half life: 24,100 y
- $^{238}Pu \rightarrow ^{234}U$
  - Half life: 87.7 y

Bellucci et al. (2013b, Analytical Chemistry)

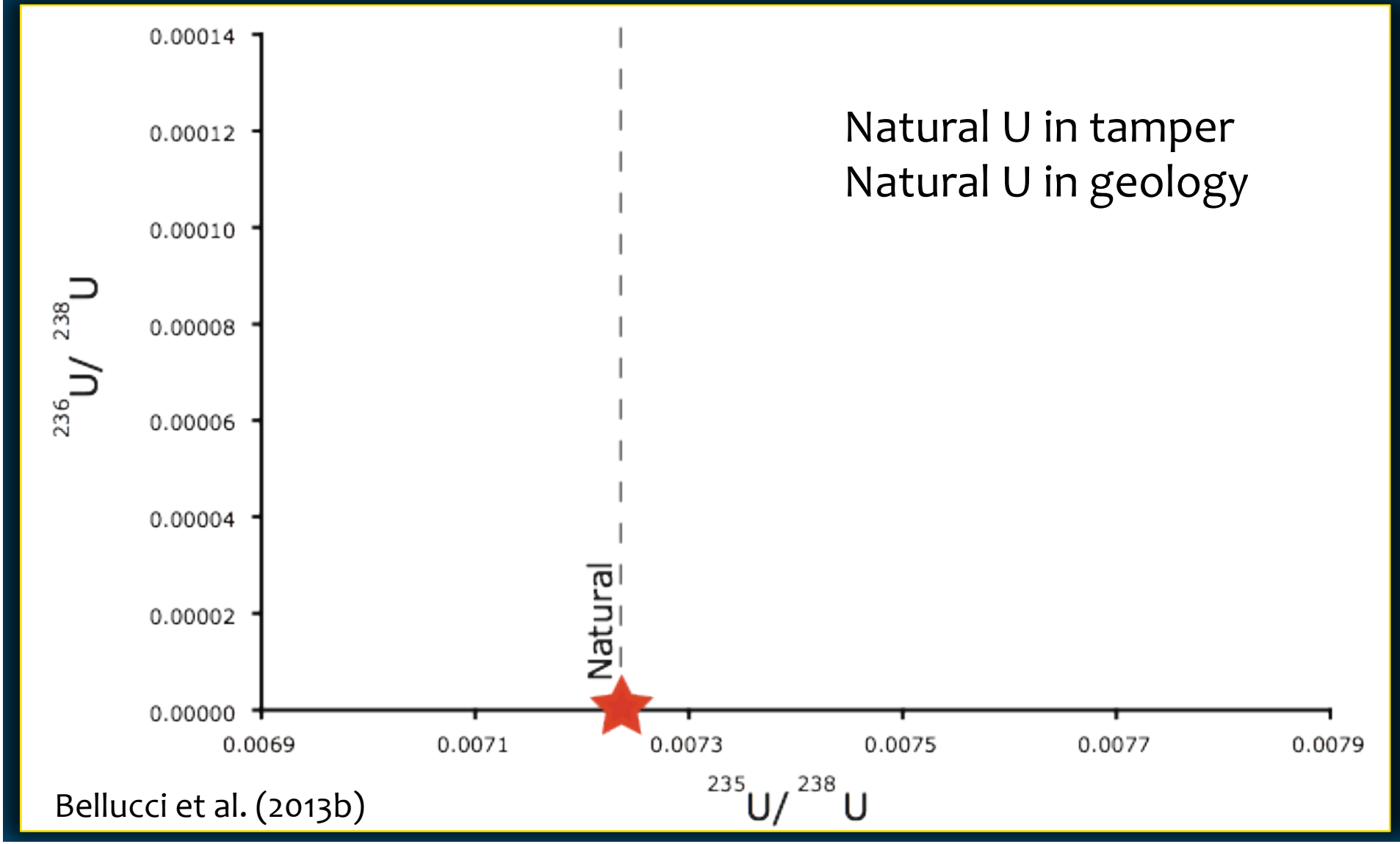
Isotope Analysis

Laser Ablation Multi Collector ICP-MS

- Nu Plasma II Parameters
- ESI New Wave 193 Excimer



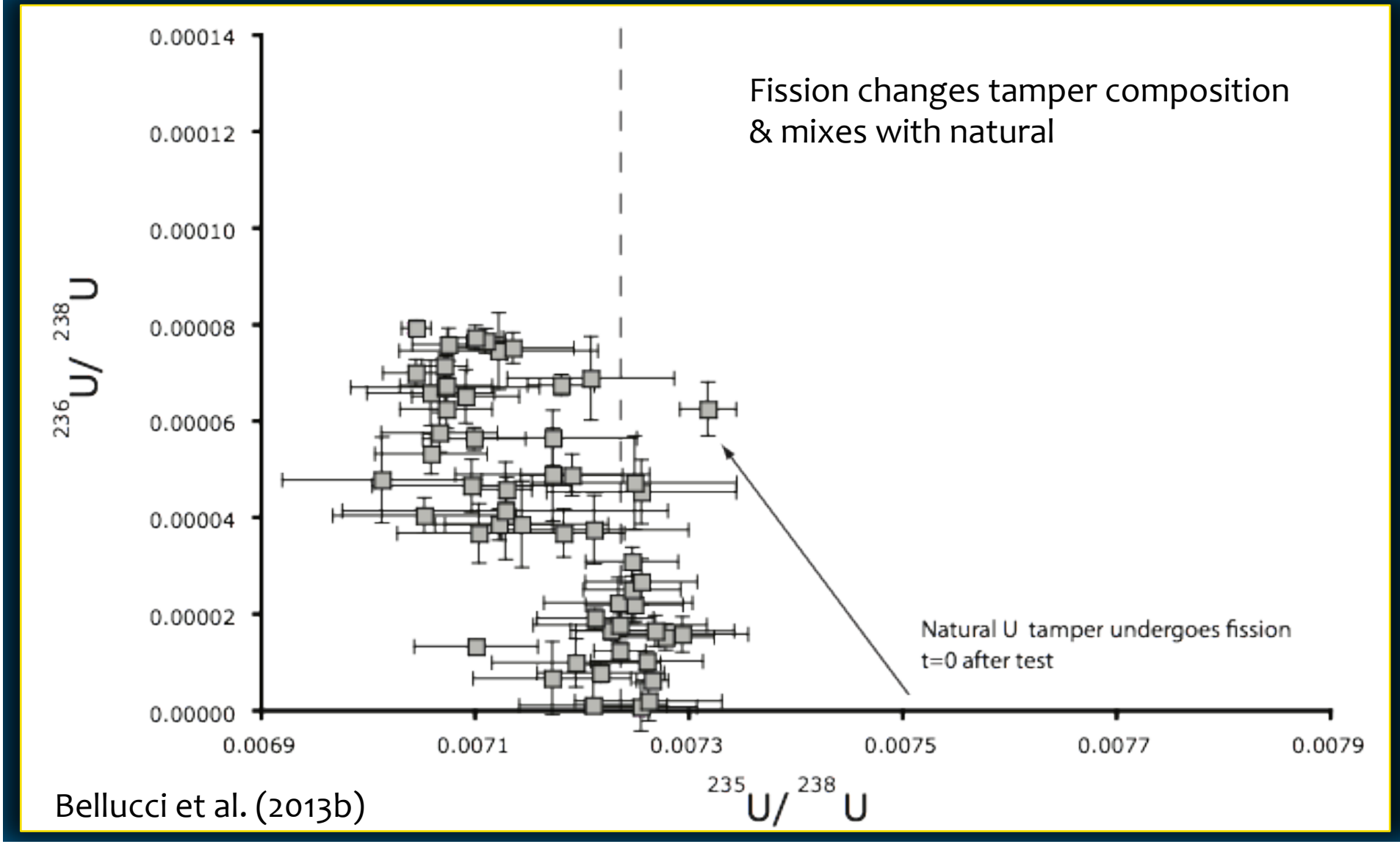
Uranium isotopes



Natural U in tamper
Natural U in geology

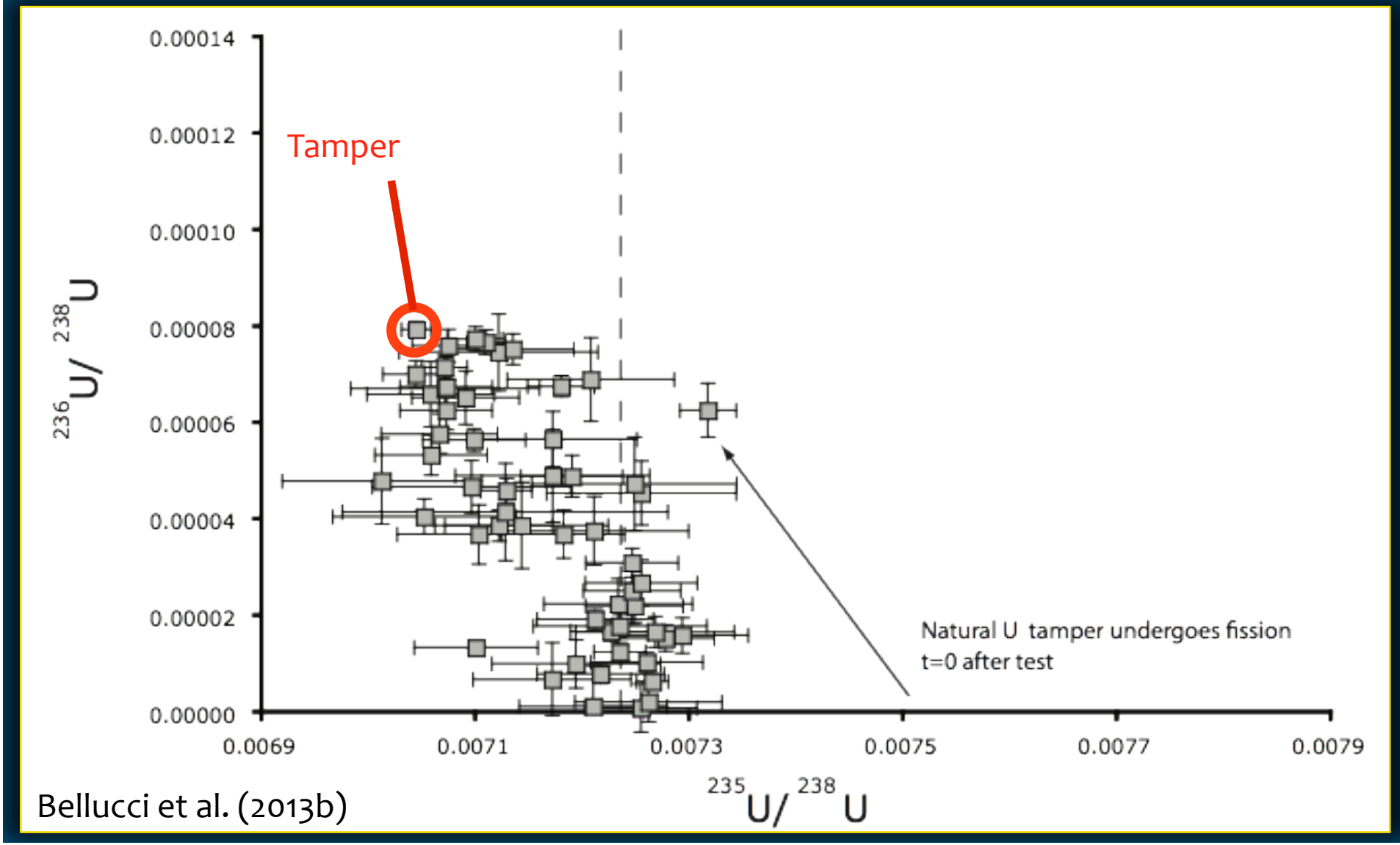
Bellucci et al. (2013b)

Uranium isotopes



Bellucci et al. (2013b)

Uranium Isotopes



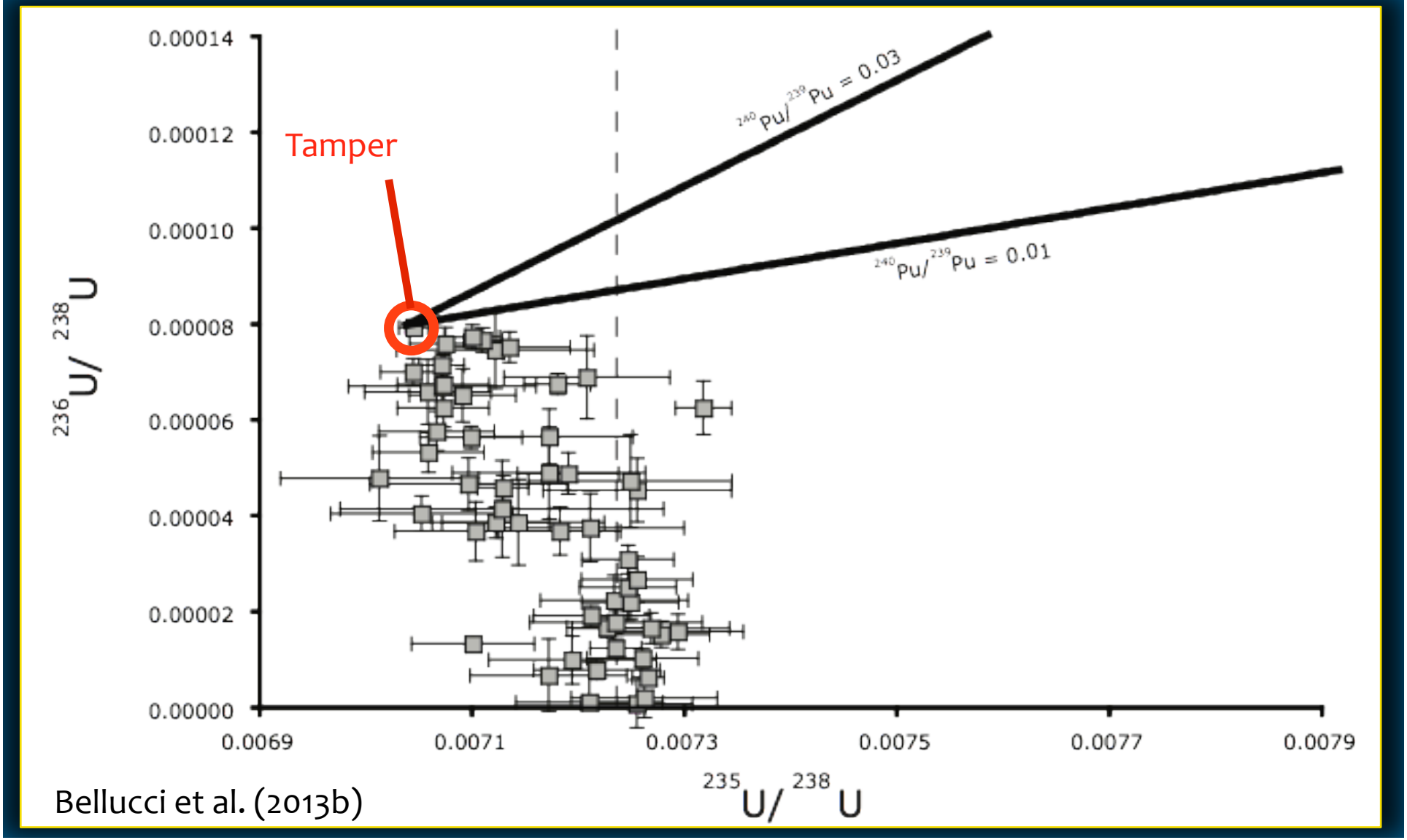
Bellucci et al. (2013b)

Influence of Pu

- Assuming an initial U isotopic composition
- Known half-lives
- One can mathematically predict the U isotopic composition resulting from the in-growth of Pu over ~68 years

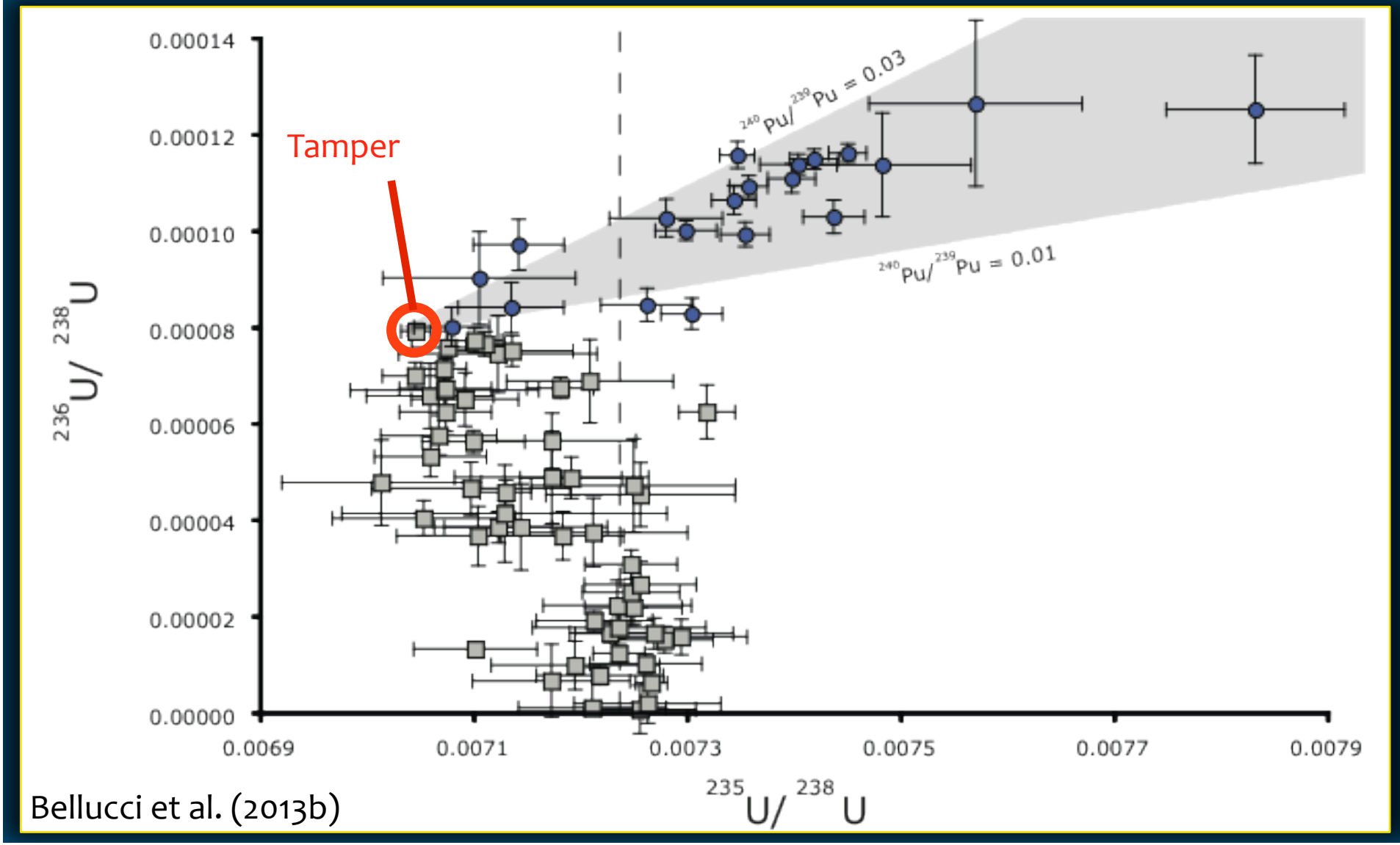
- $^{235}U/^{238}U\ (present) = {}^{235}U/^{238}U\ (initial) + {}^{239}Pu/^{238}U * (e^{\lambda 239Put} - 1)$
- $^{236}U/^{238}U\ (present) = {}^{236}U/^{238}U\ (initial) + {}^{239}Pu/^{238}U * {}^{240}Pu/^{239}Pu * (e^{\lambda 240Put} - 1)$
- $^{234}U/^{238}U\ (present) = {}^{234}U/^{238}U\ (initial) + {}^{239}Pu/^{238}U * {}^{238}Pu/^{239}Pu * (e^{\lambda 238Put} - 1)$

Pu model of "super grade Pu"

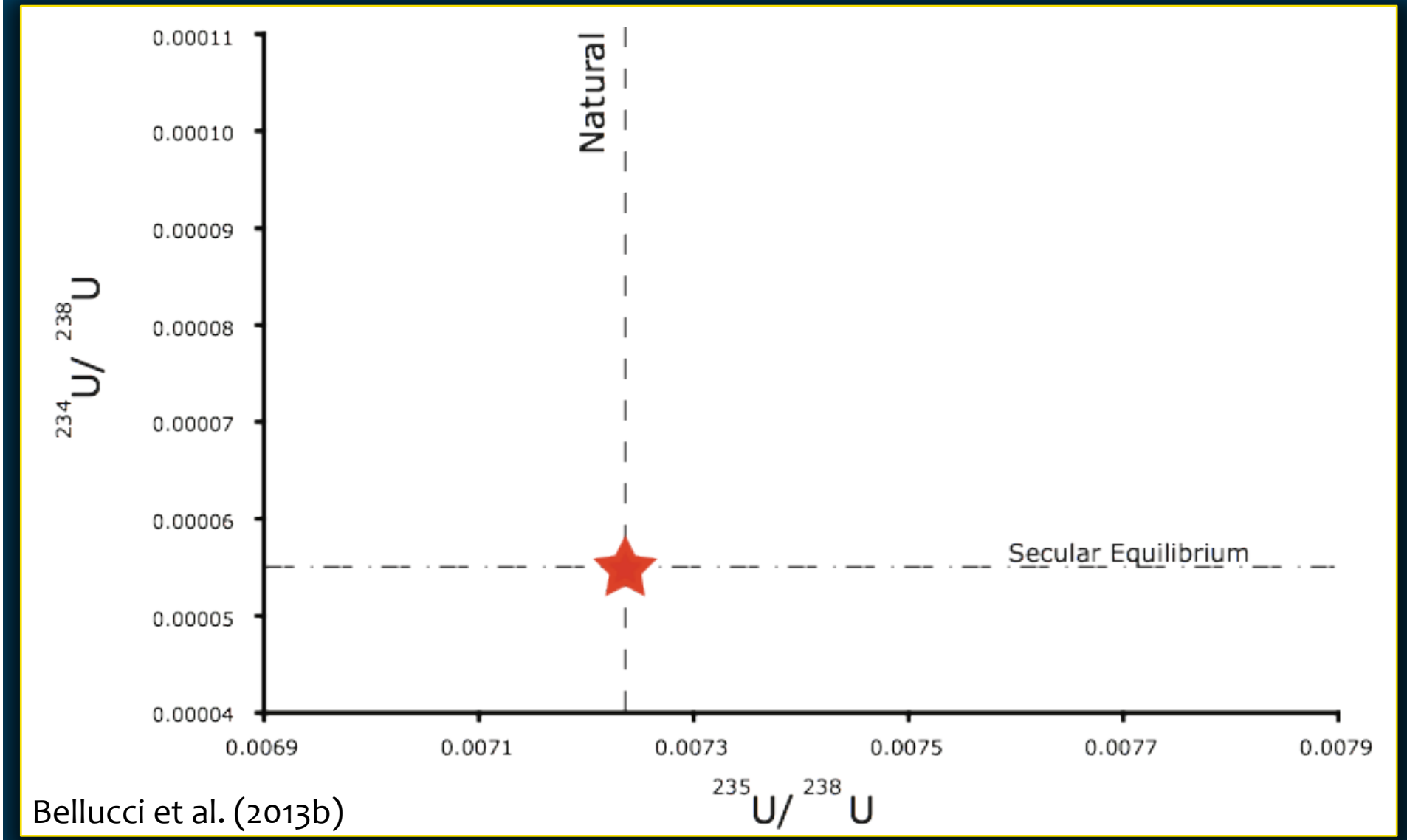


Bellucci et al. (2013b)

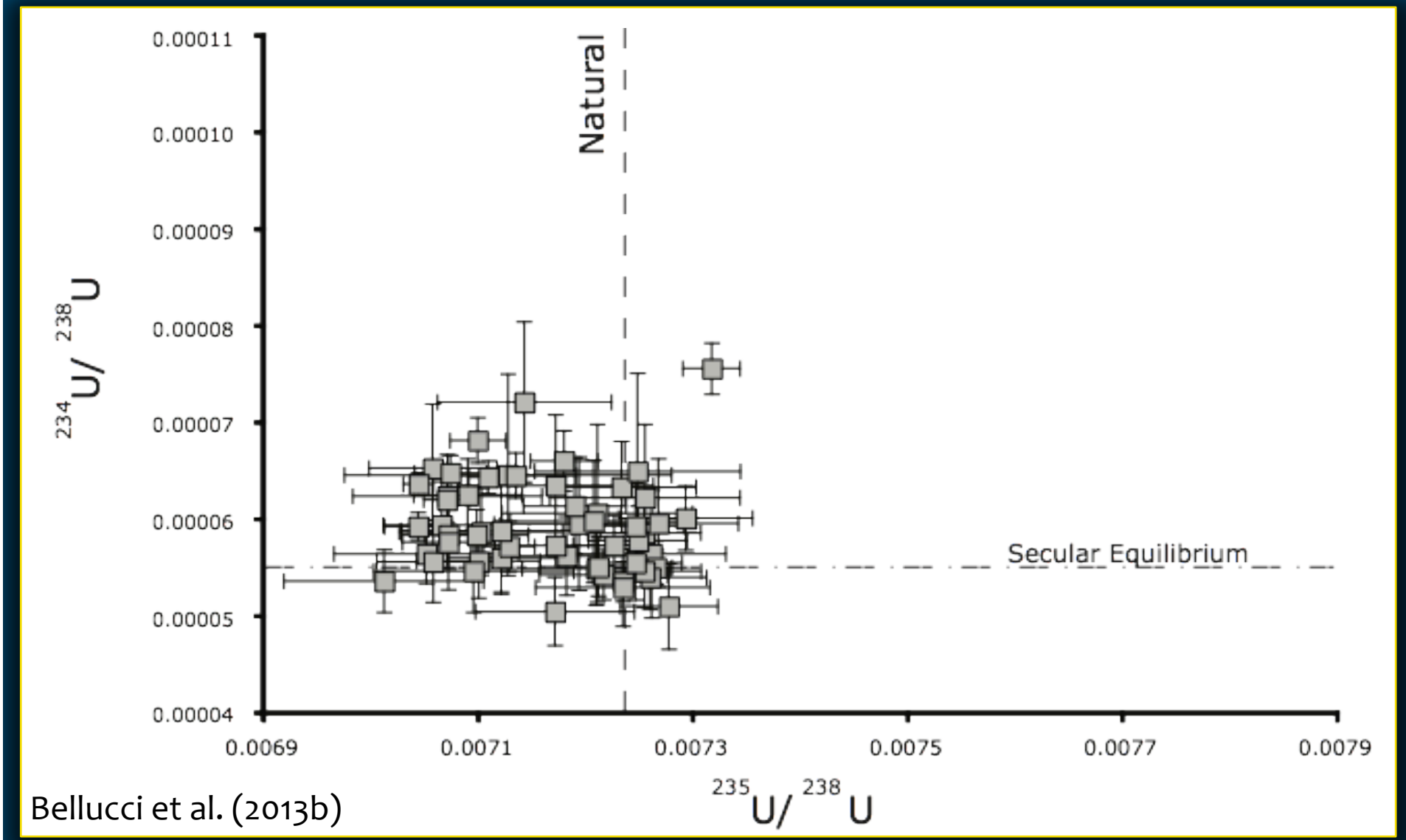
Spots with high Pu concentrations



Bellucci et al. (2013b)

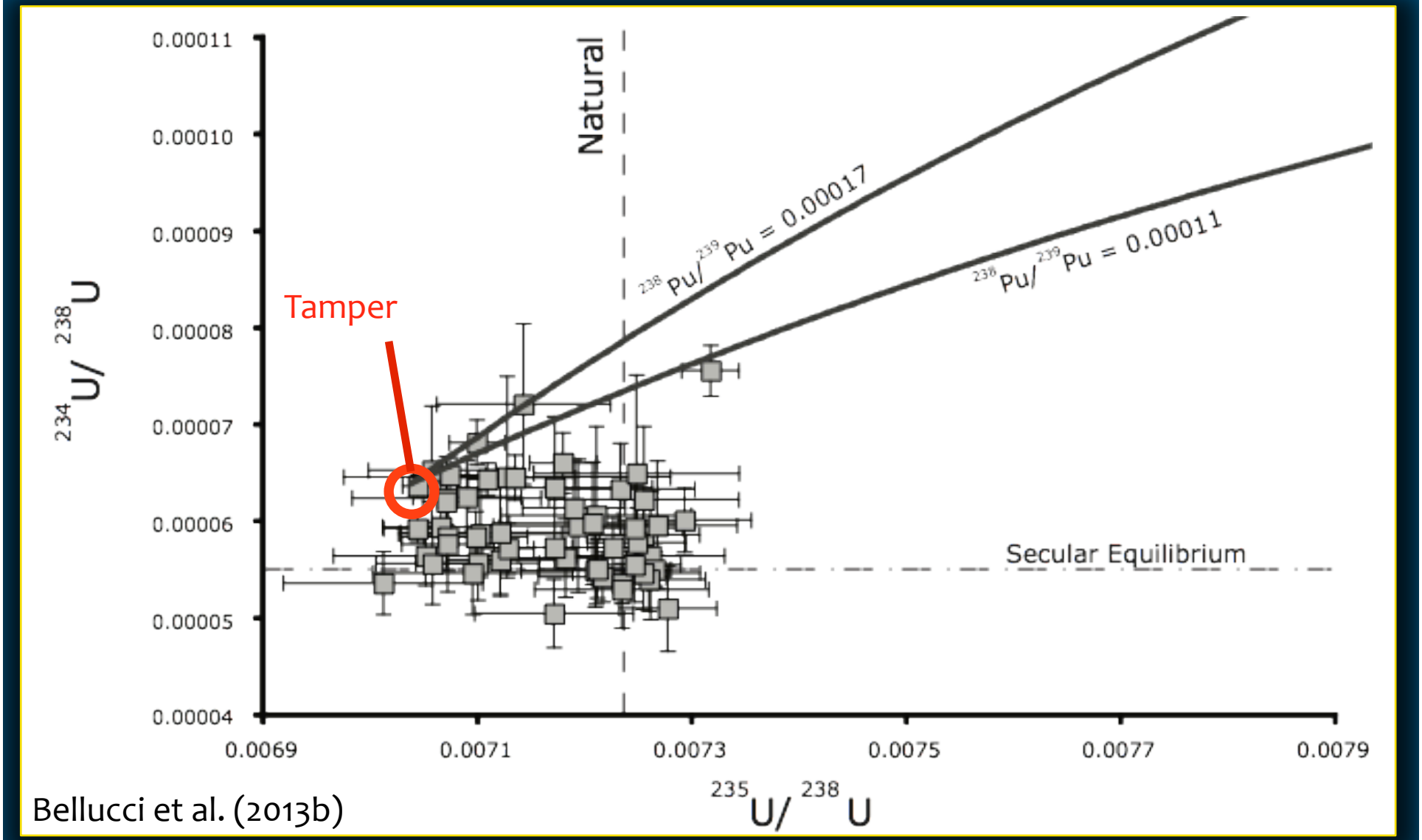


Bellucci et al. (2013b)



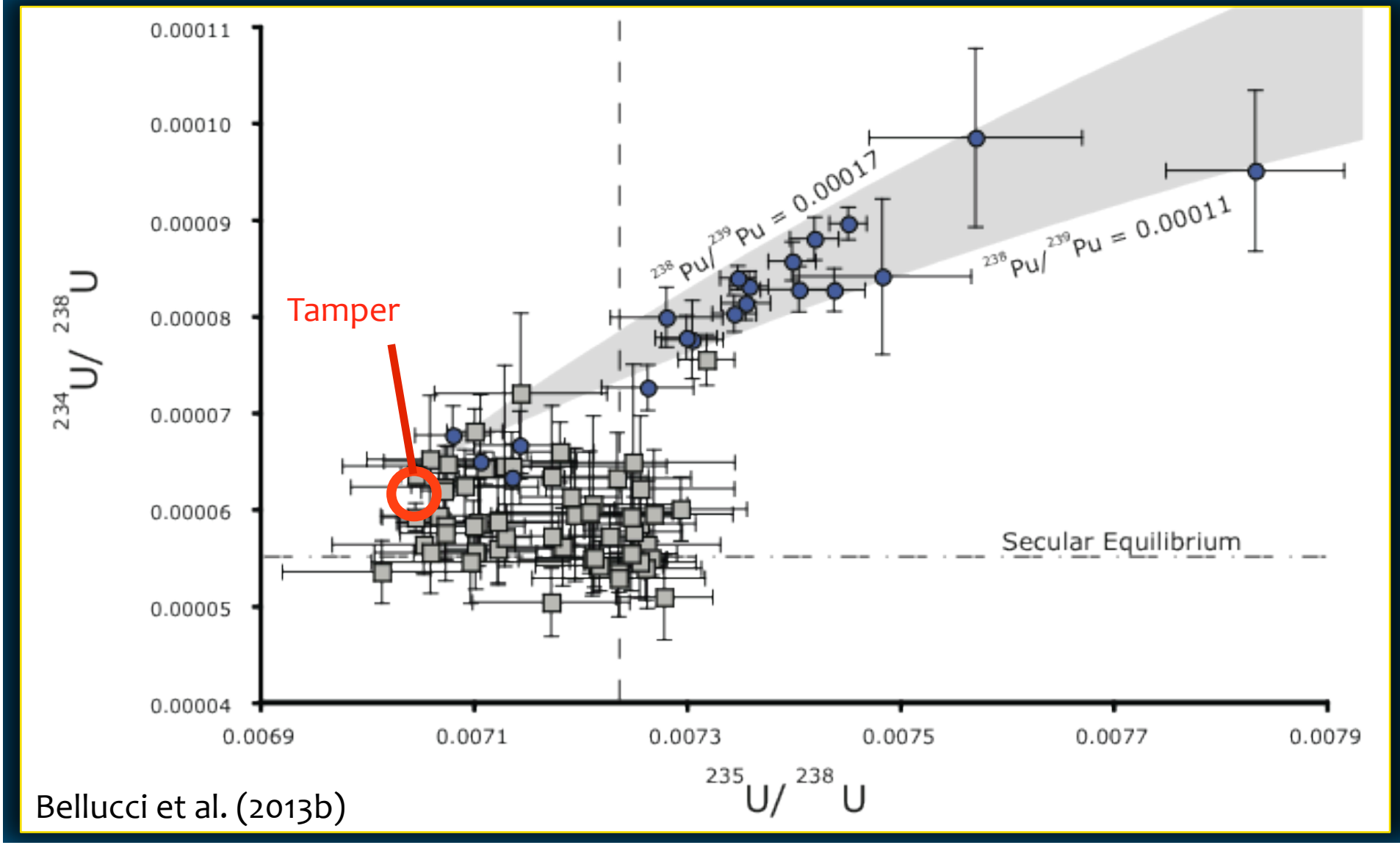
Bellucci et al. (2013b)

Pu-model



Bellucci et al. (2013b)

Spots with high Pu concentrations



Bellucci et al. (2013b)

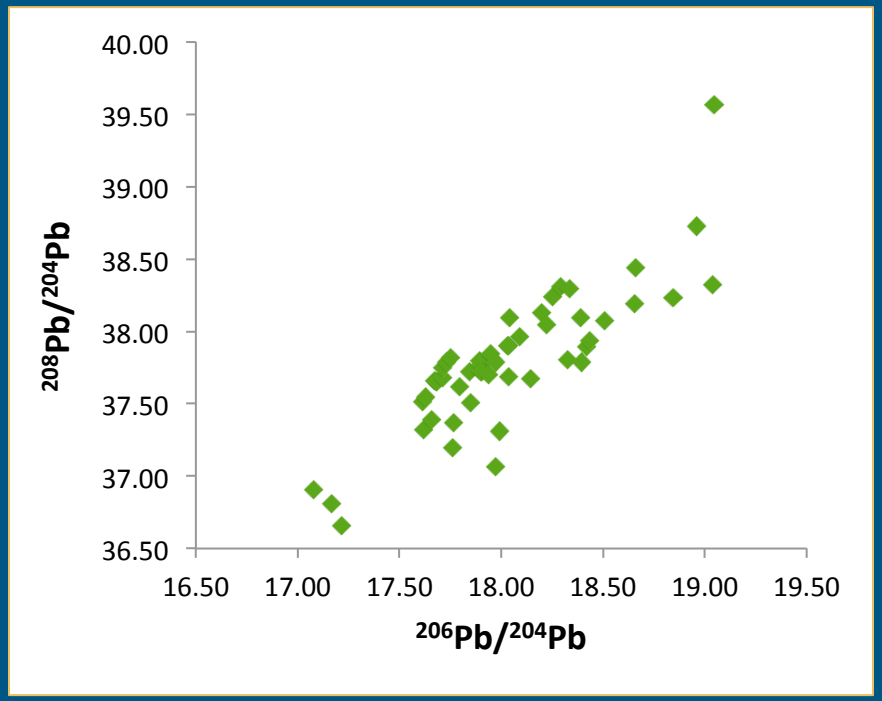
Conclusions

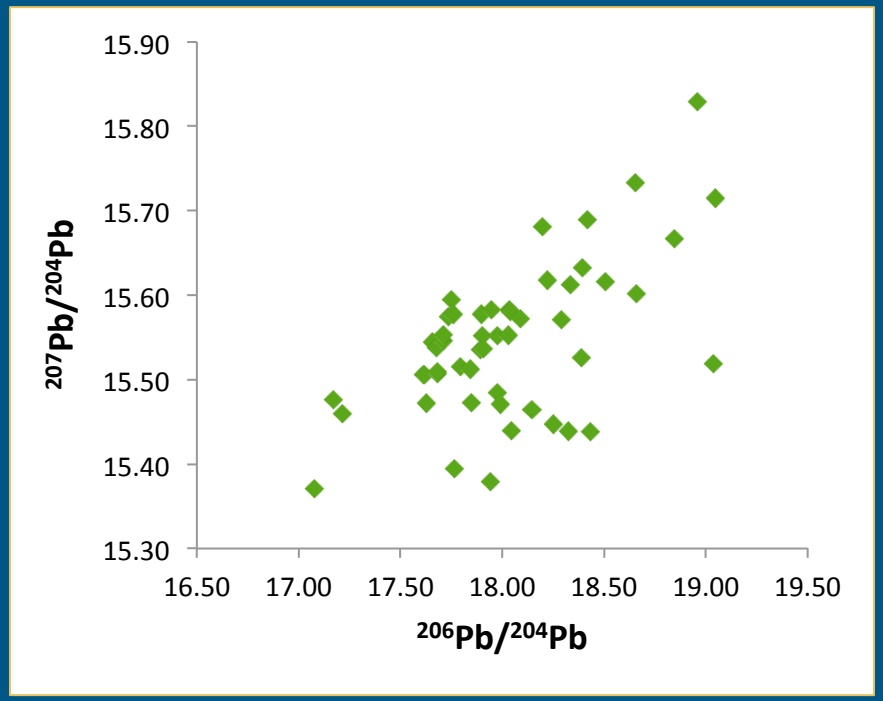
- Forensic investigation of PDMs is complex and requires a multi-analytical approach for accurate assessment of device's chemical & isotopic composition
- Traditional protocols involving "bulk" samples are time-consuming and will tend to "average out" the chemical and isotopic signals from device and matrix components
- Micro-analytical approach can provide both rapid and accurate forensic information – key attributes for source attribution purposes

On-going Research - Trinitite

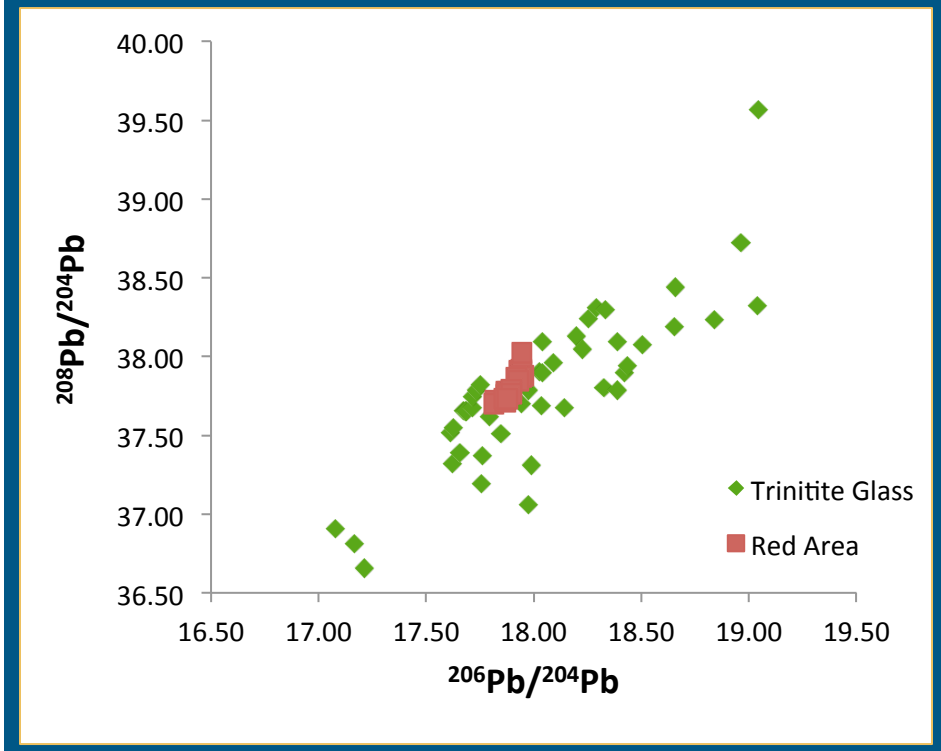
- Pu isotope analysis – LA-MC-ICP-MS (Dr. S. Mana)
- Oxygen isotope analysis – laser fluorination (E. Koeman et al., in press, *Analytical Chemistry*)
- Pb isotope analysis of "Red areas" – LA-MC-ICP-MS (E. Koeman & J. Bellucci)
- Li isotope analysis – MC-ICP-MS (T. Magna, Czech Geol. Survey)

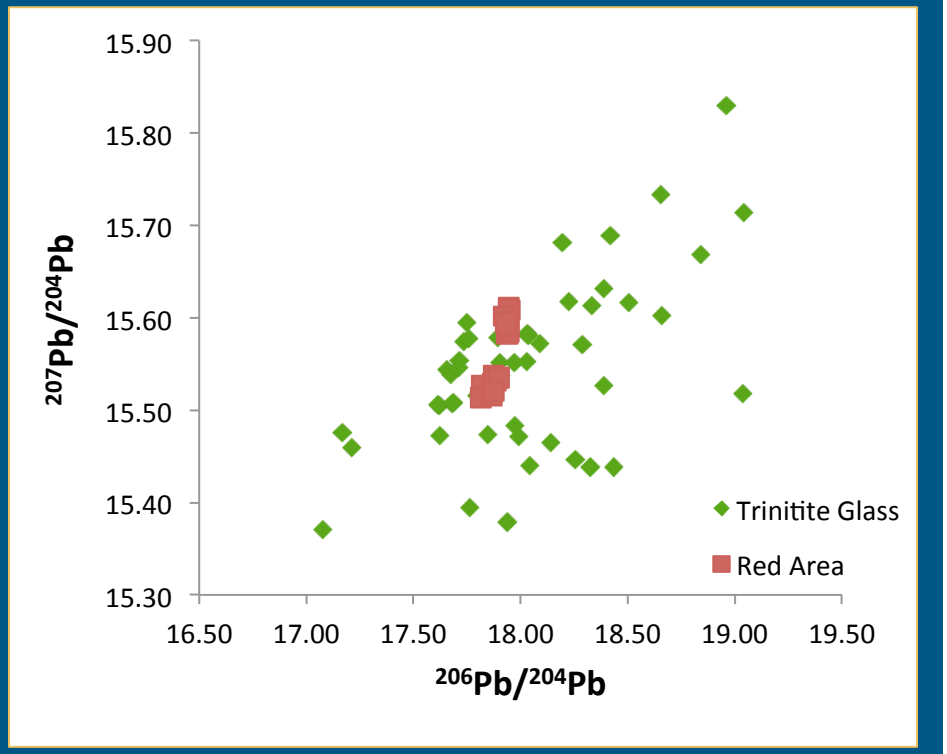
Lead Isotopes in Trinitite





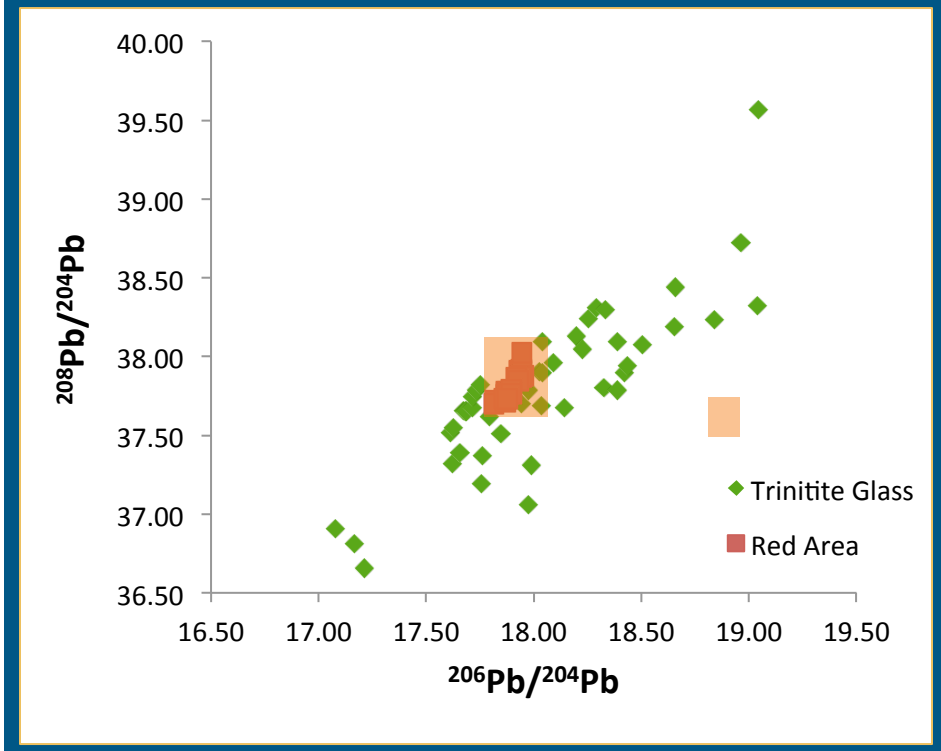
Pb Isotopes

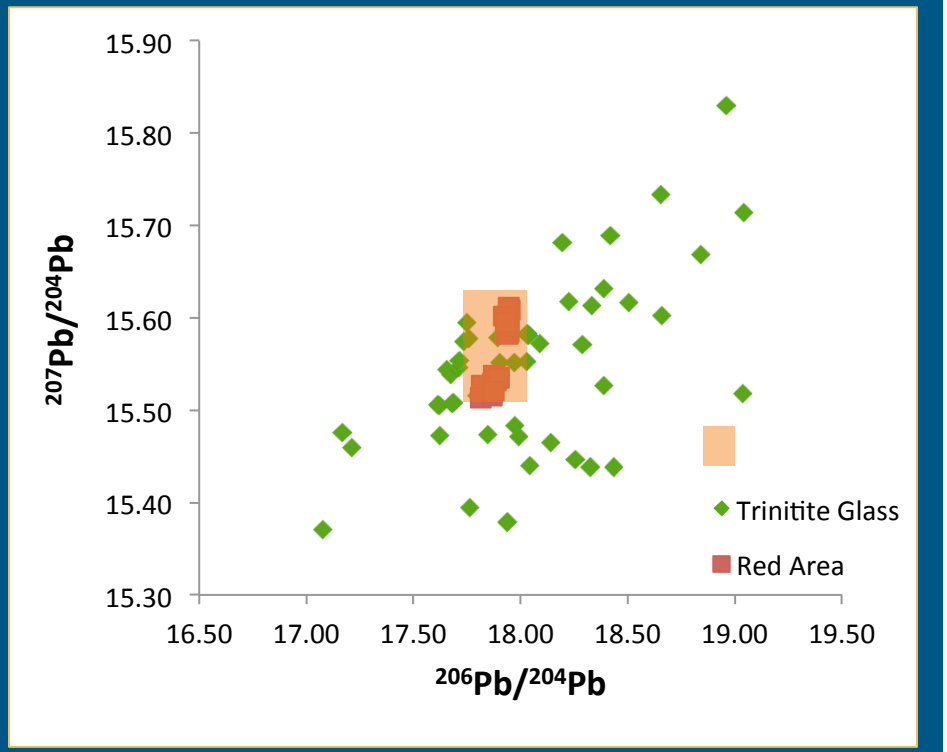


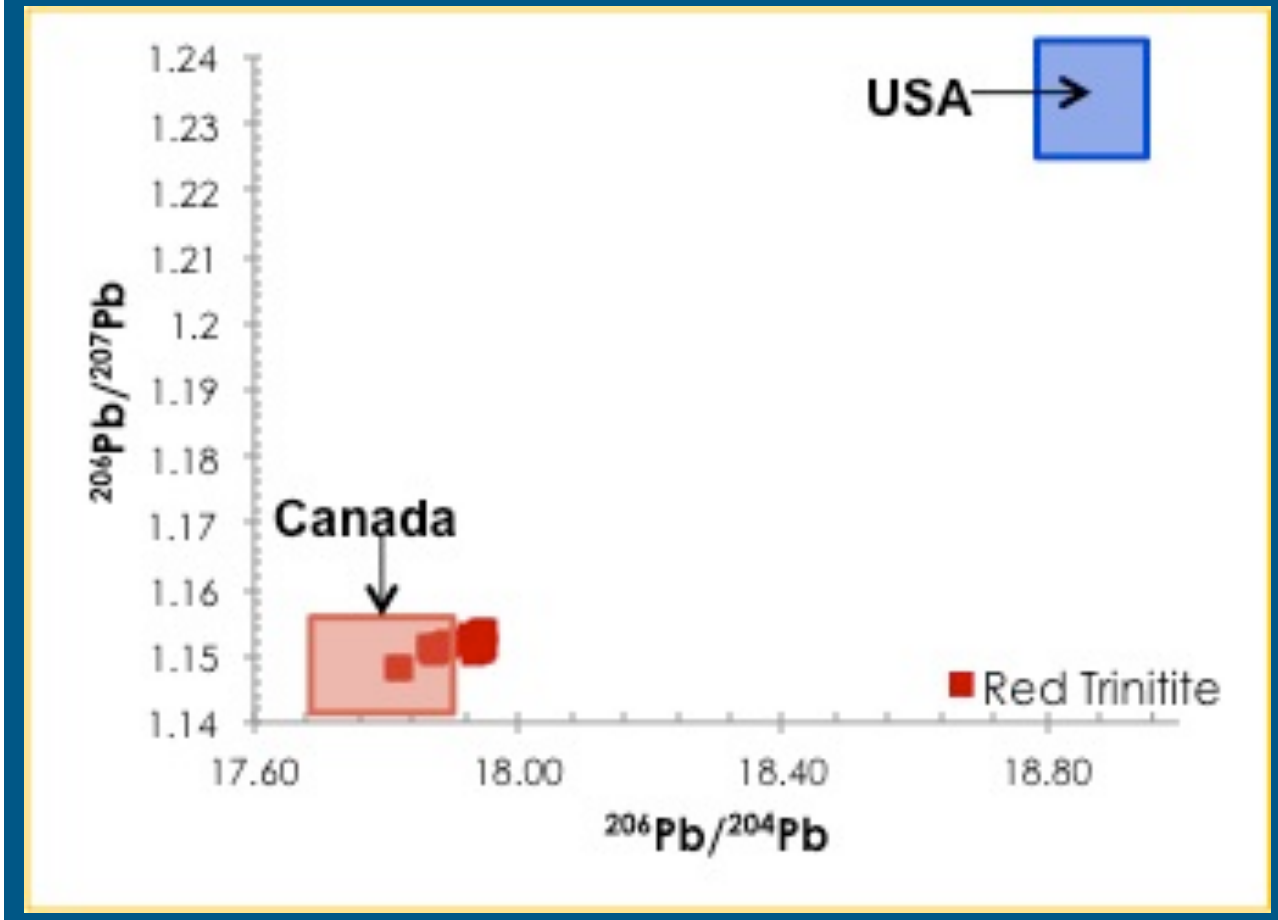


Koeman et al. (in prep.)

Estimated Device Composition







- Buchans Mine, NFLD (Canada)
  - Only mine active
  - Mined by American Smelting and Refining Co. from 1928-1984

Koeman et al. (in prep.)