Title: The Geochemistry of Nuclear Fallout

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The Geochemistry of Nuclear Fallout

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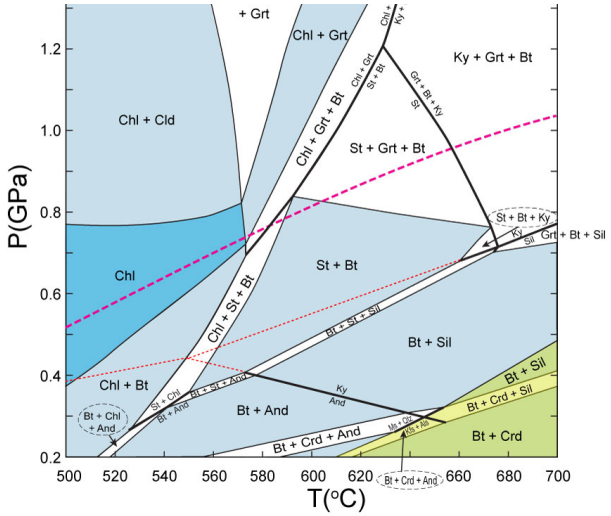
8 March 2018

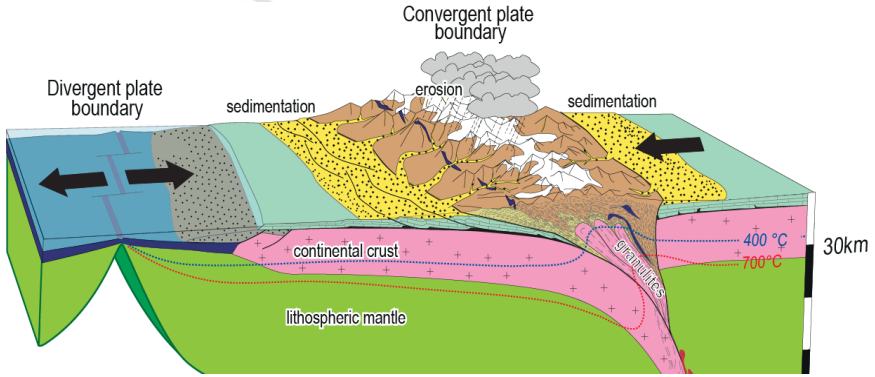
LA-UR-18-xxxx

A Geochemist's Skill Set

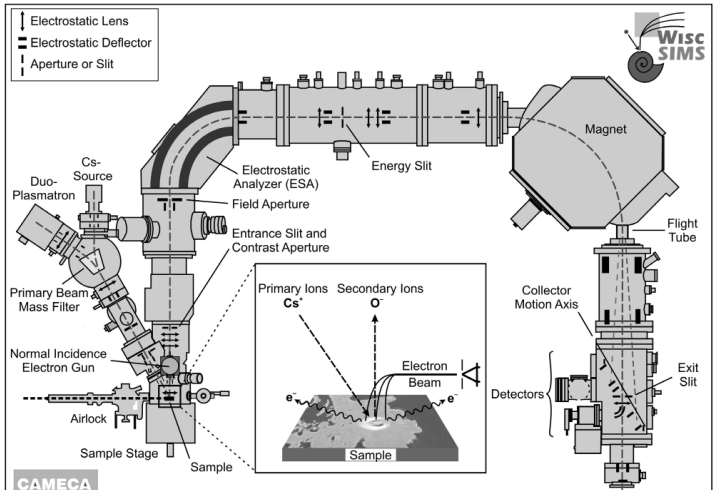
Los Alamos NATIONAL LABORATORY EST. 1943

1. Thermodynamics & Kinetics





2. Earth Materials Processes



3. Analytical Methods & Instrumentation

4. Field Work

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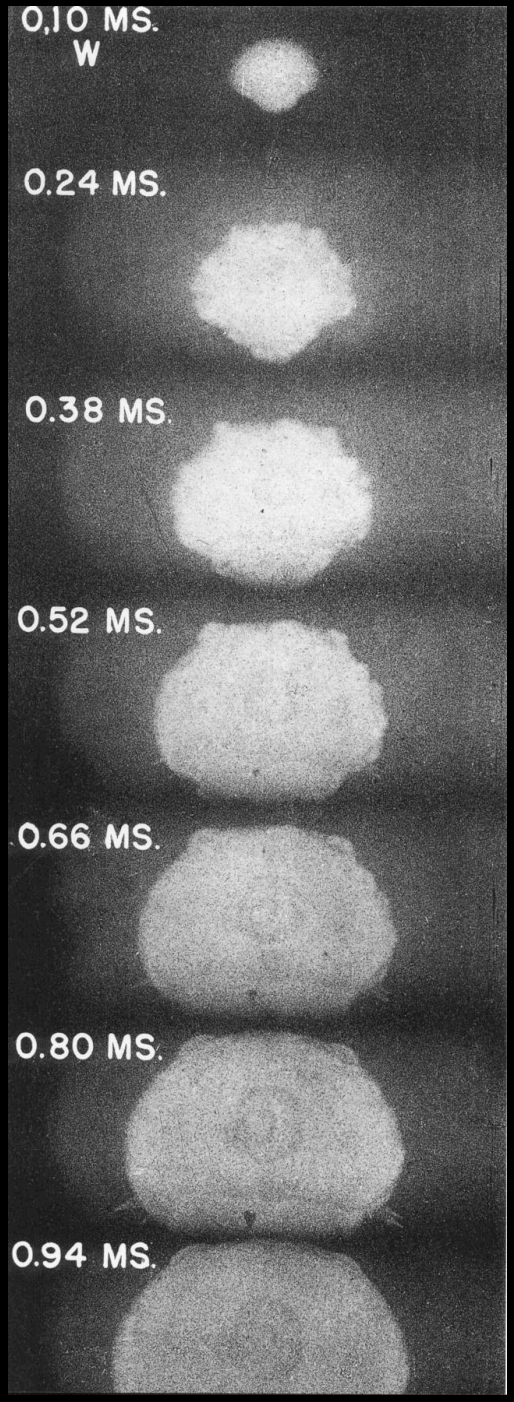
NNSA National Nuclear Security Administration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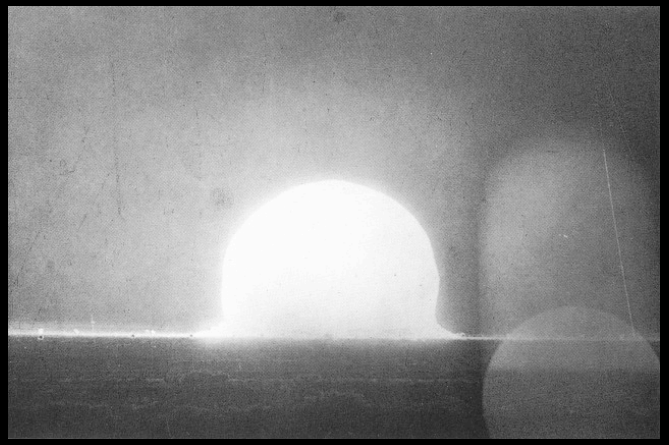

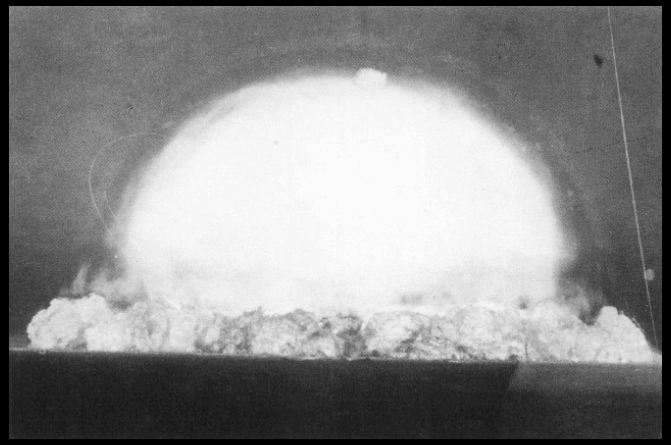

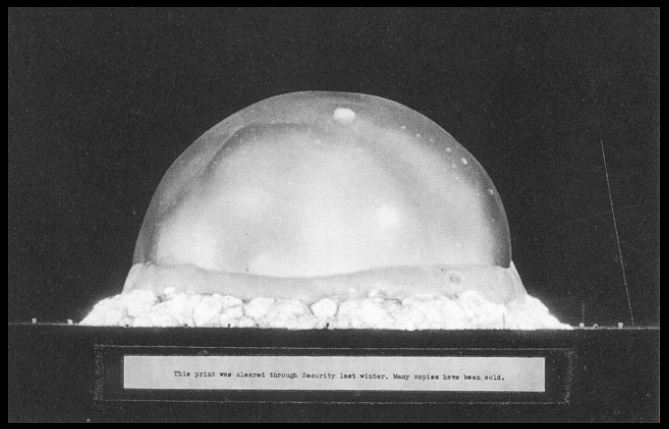
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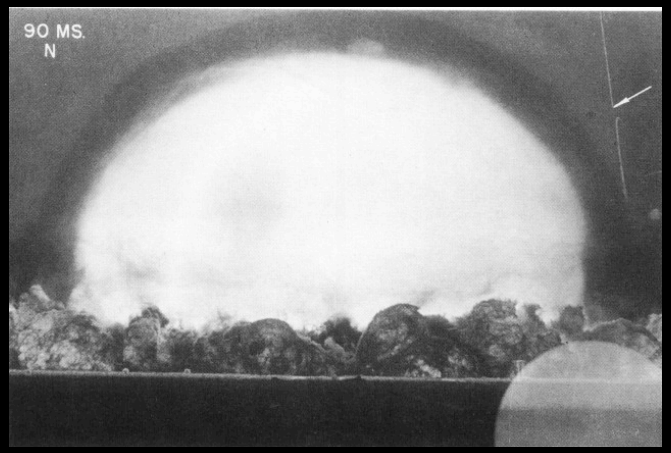
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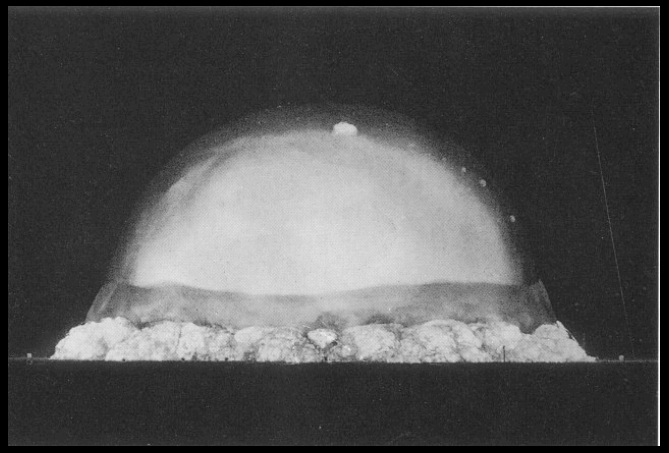
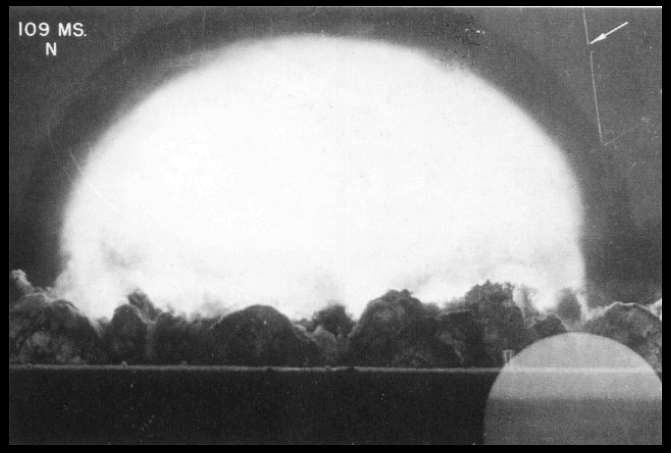



109 MS.
N

Trinitite

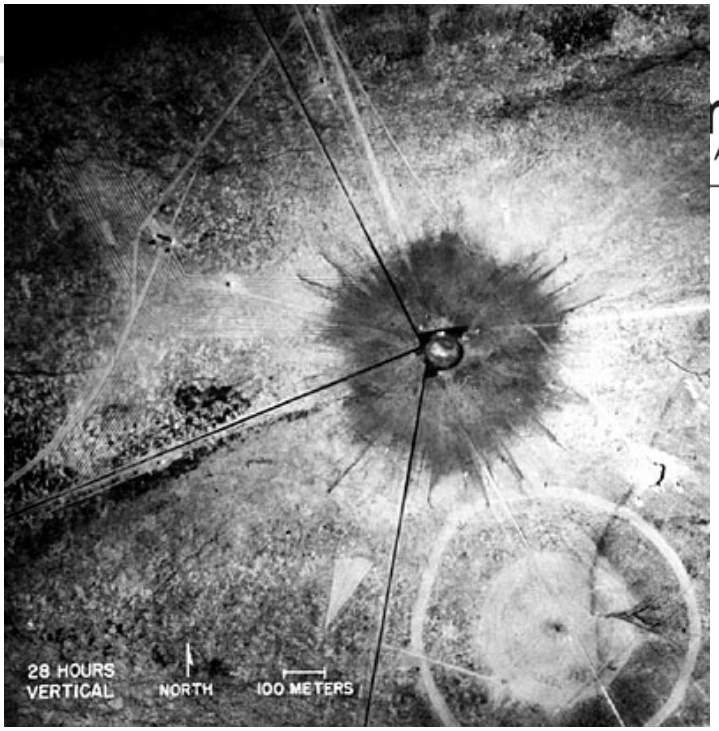
Eby et al. 2010

Trinitite: What is it Good For?

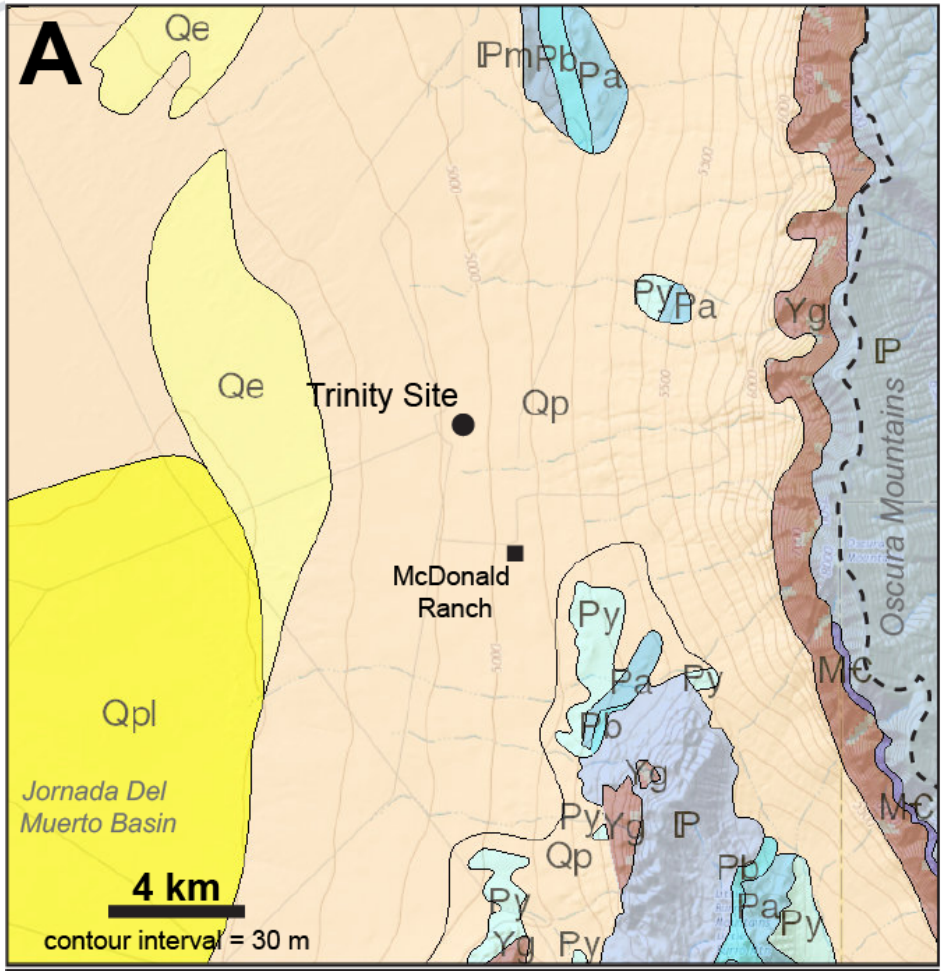
- Accessible example of nuclear explosion products
- (Cryptic) chemical and isotopic representative of an atomic bomb
- Ability to attribute a particular device to a particular source is a nuclear deterrent

Trinity Test

- ^{239}Pu fuel packed in natural U
- Detonated from a steel tower
- Arkosic sand ground material
  - Major minerals: quartz, microcline, anorthoclase, calcite
  - Minor minerals: gypsum, clay, hornblende, salts, zircon



Trinity Site Geology



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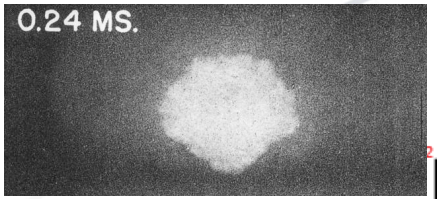
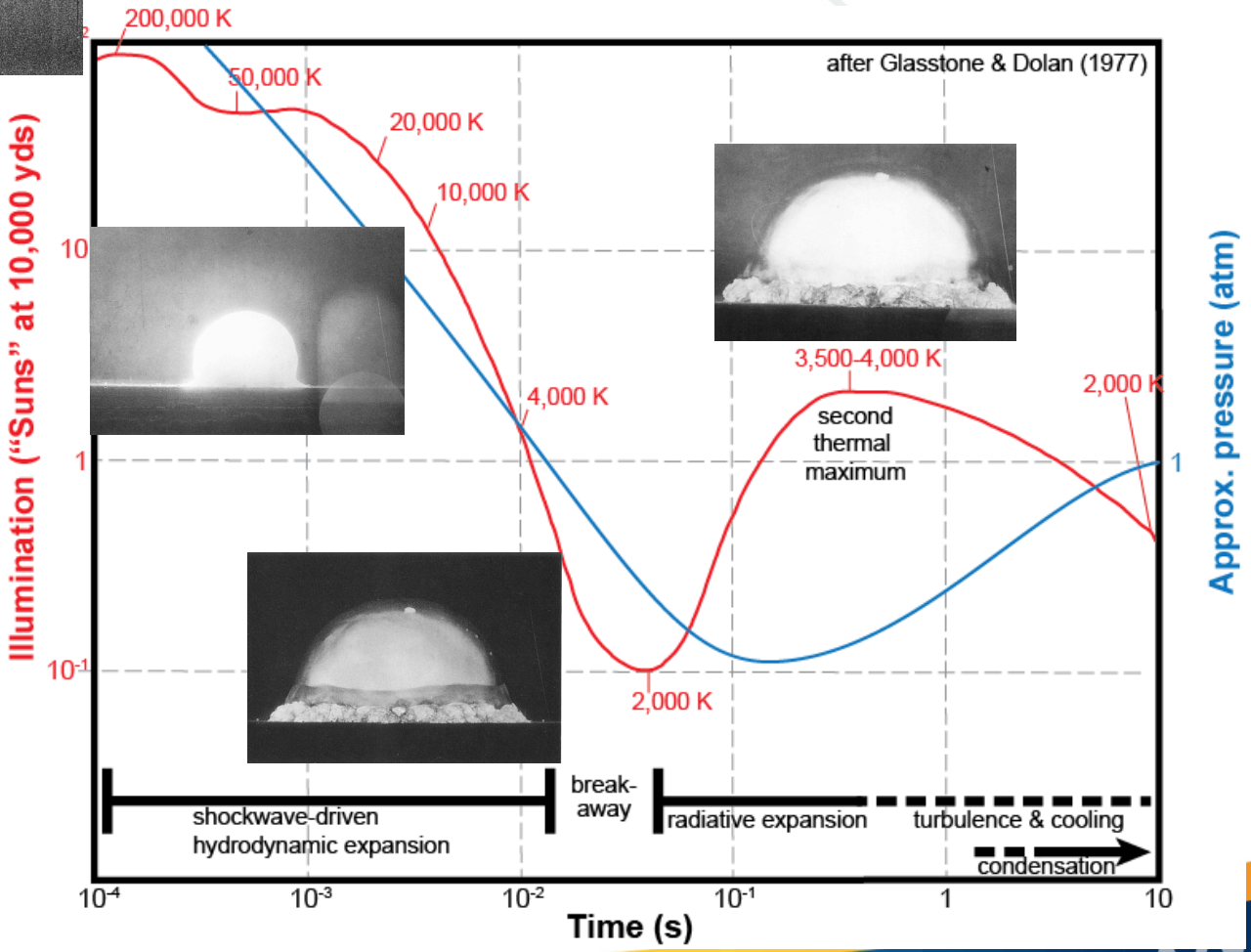
Nuclear Fireball Phenomenology





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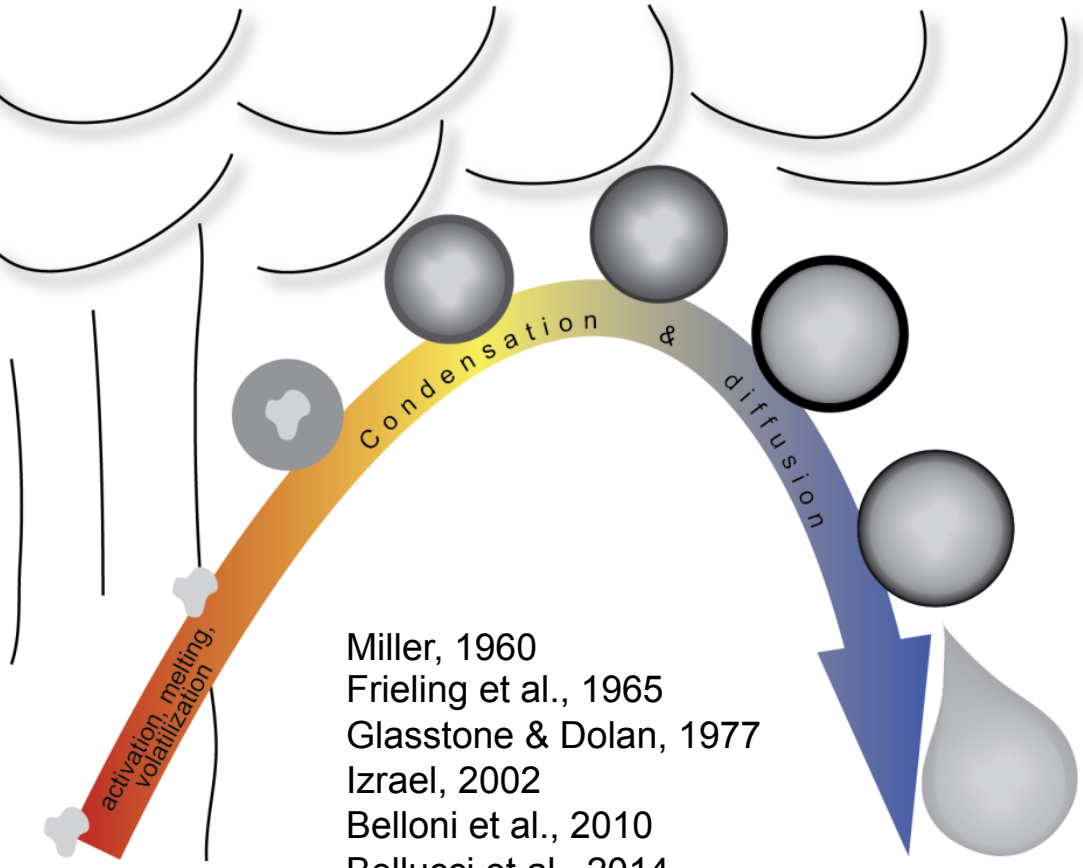
Existing Fallout Formation Model

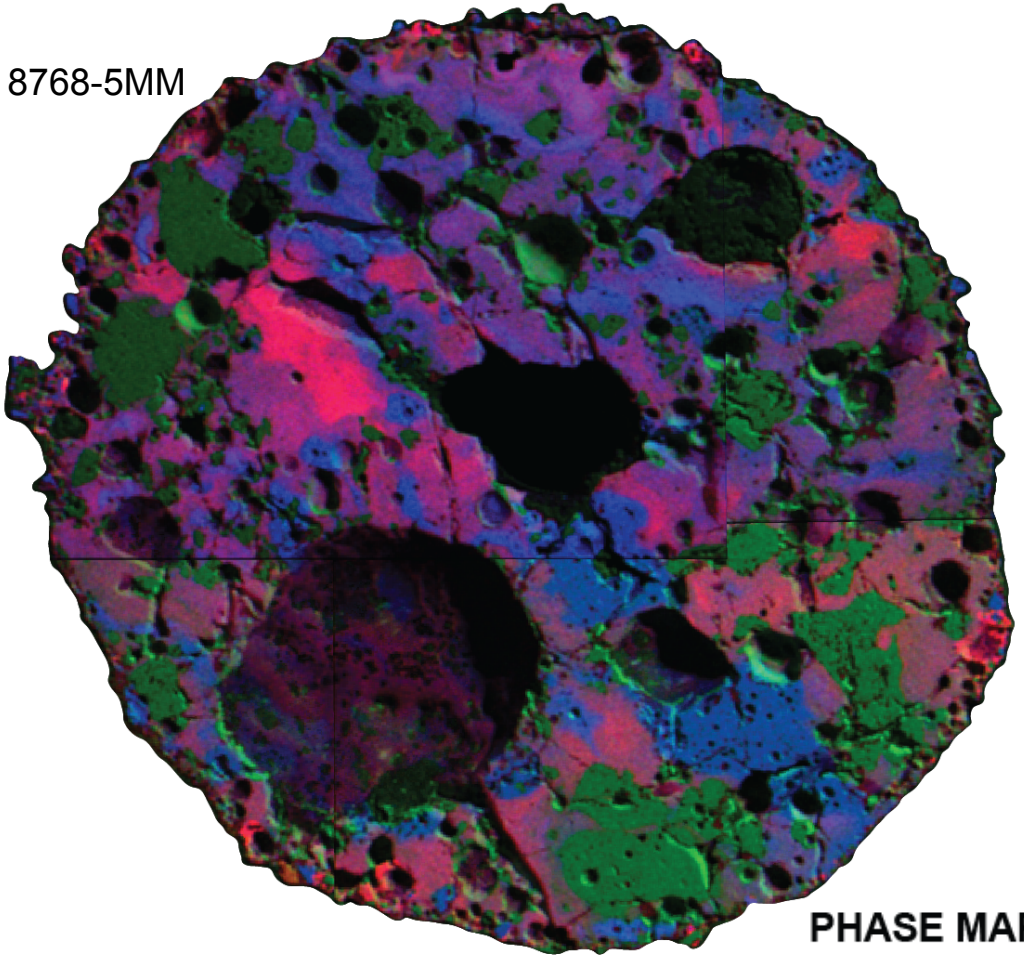
- Volatilization of device
- Partial melting and entrainment of ground material
- Condensation
  - Refractory bomb material
  - Ground material nuclei
- Homogenization by diffusion and/or melt mixing
- Cooling and fallout



Miller, 1960
Frieling et al., 1965
Glasstone & Dolan, 1977
Izrael, 2002
Belloni et al., 2010
Bellucci et al., 2014
Eby, 2015
Lewis et al., 2015
Weisz et al., 2017

Fallout Heterogeneity

8768-5MM



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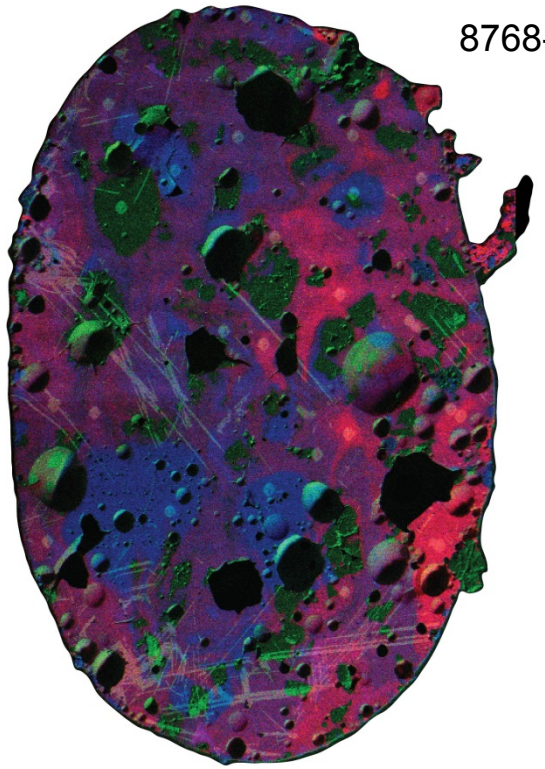

PHASE MAPS

Quartz/
Silica glass

Alkali-rich glass

CaMgFe glass

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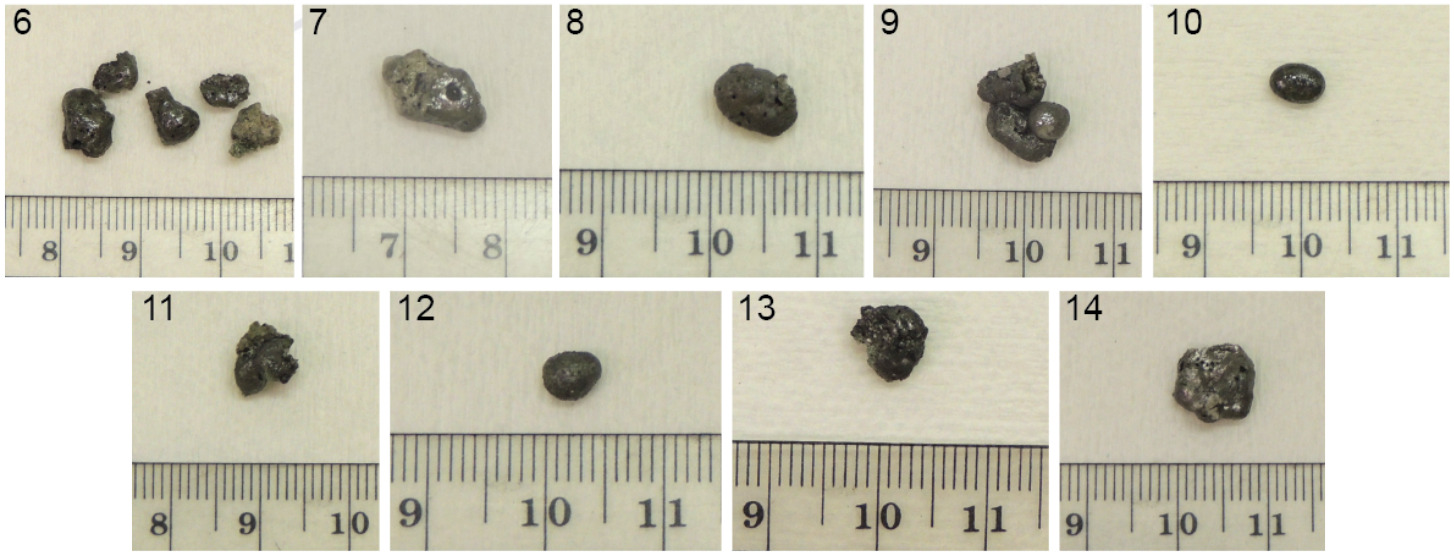
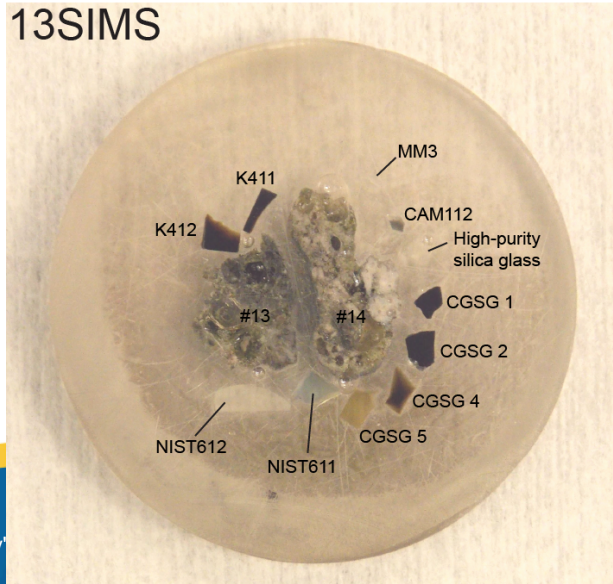
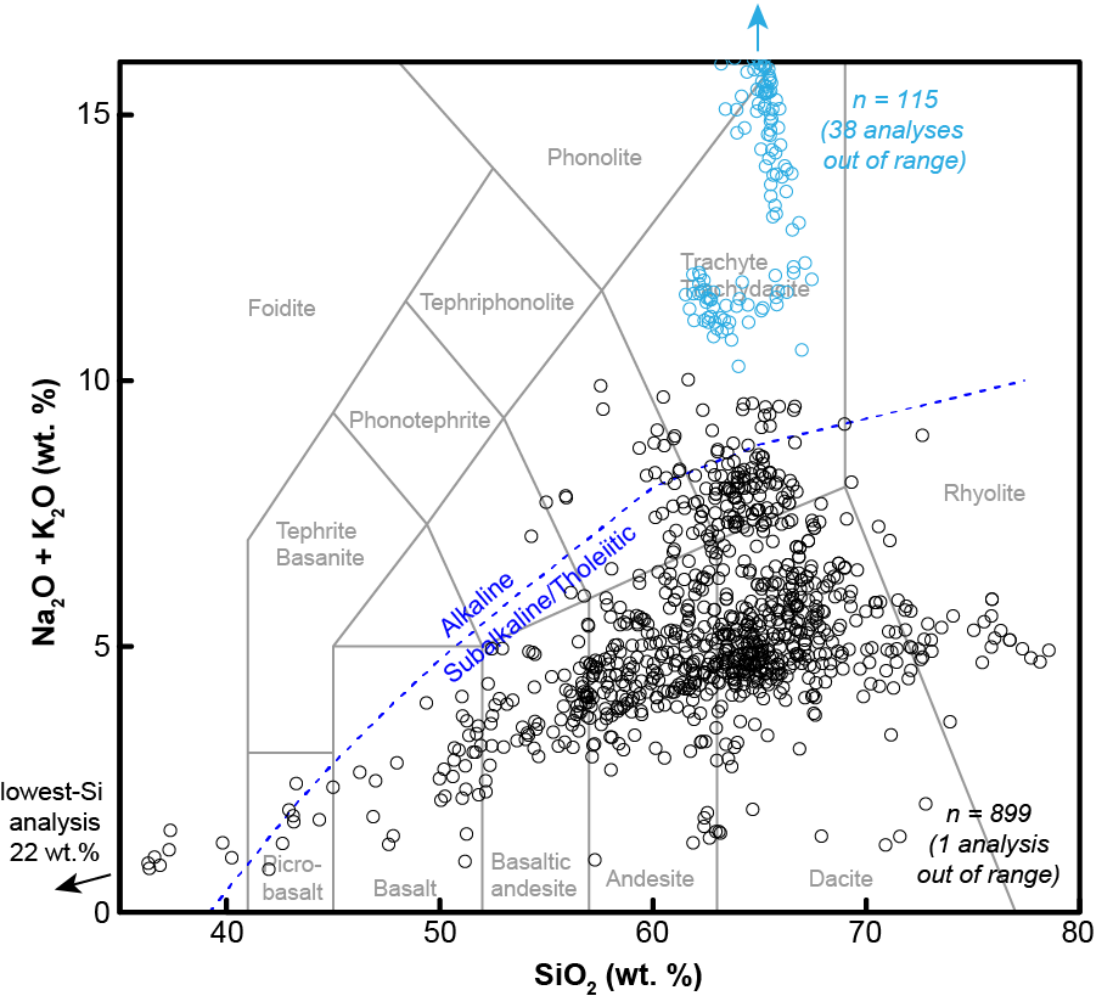
Trinity Bead Samples



- Element mapping (SEM)
- Major Element Analysis (EPMA)
- Activity mapping (autoradiography)

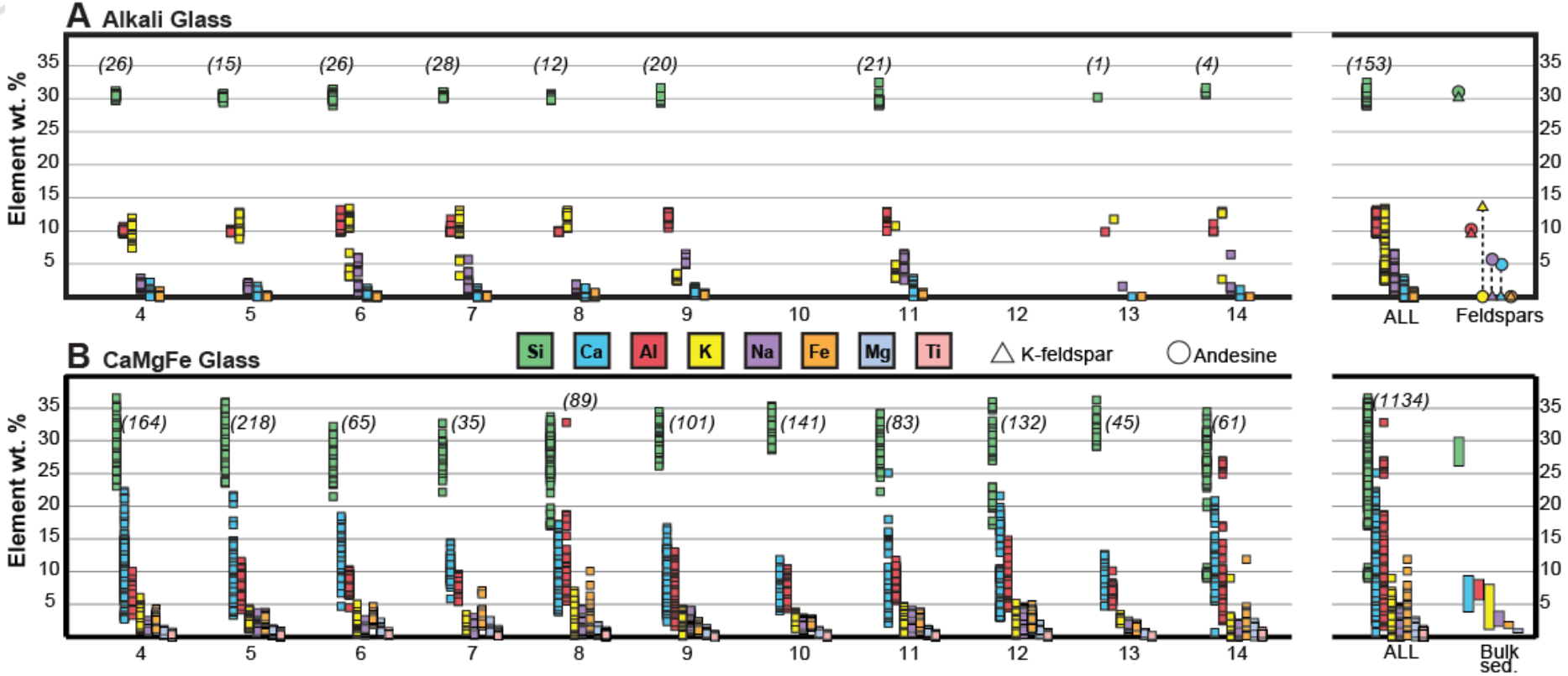


Major Element Geochemistry



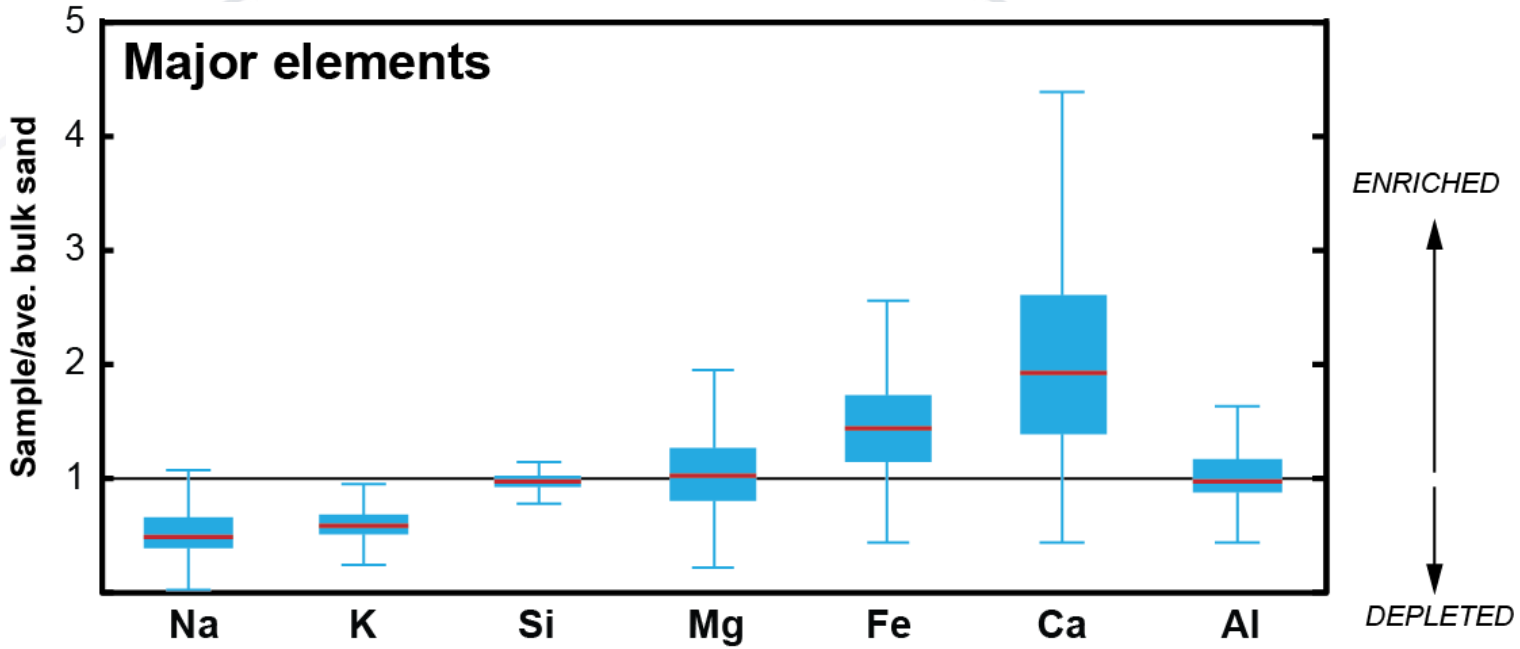
- Total alkali silica diagram
- Compositional diversity beyond known magmatic range

Major Element Geochemistry



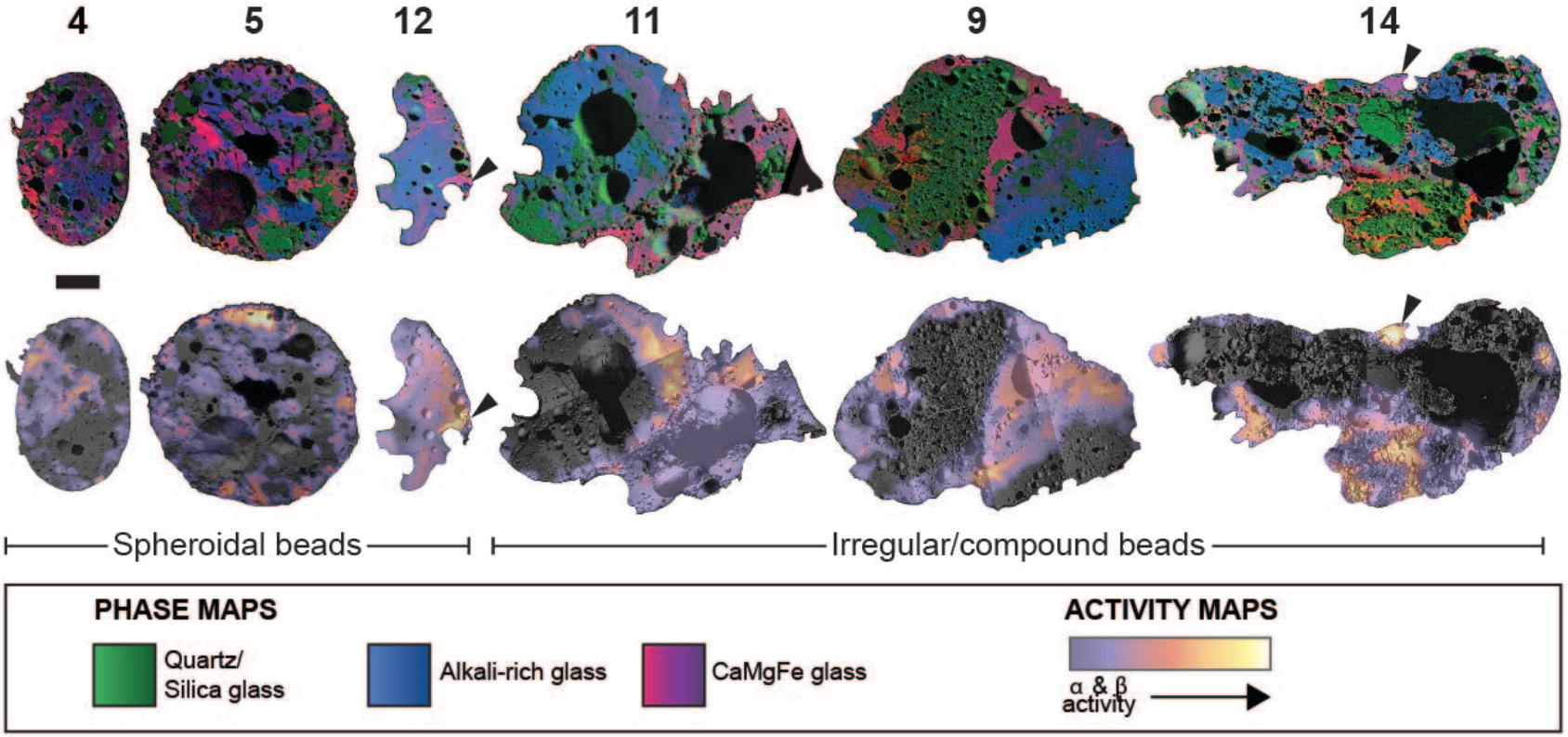
- Alkali glass = melted alkali feldspar
- Extreme compositional variability in CaMgFe glass

Major Element Geochemistry



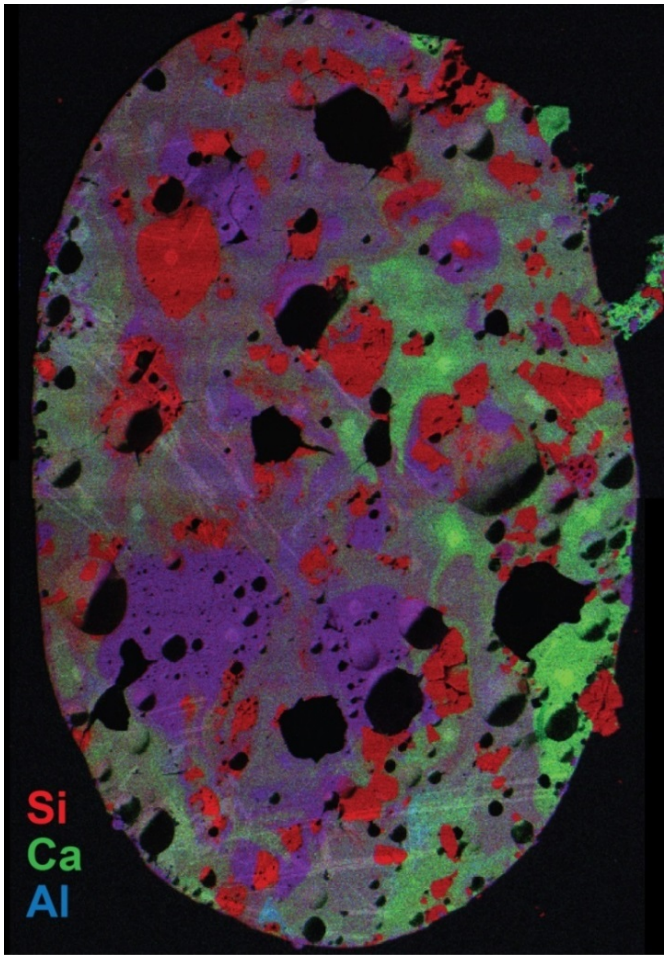
- Overall depletion of volatile elements
- Enrichment in intermediate-refractory elements

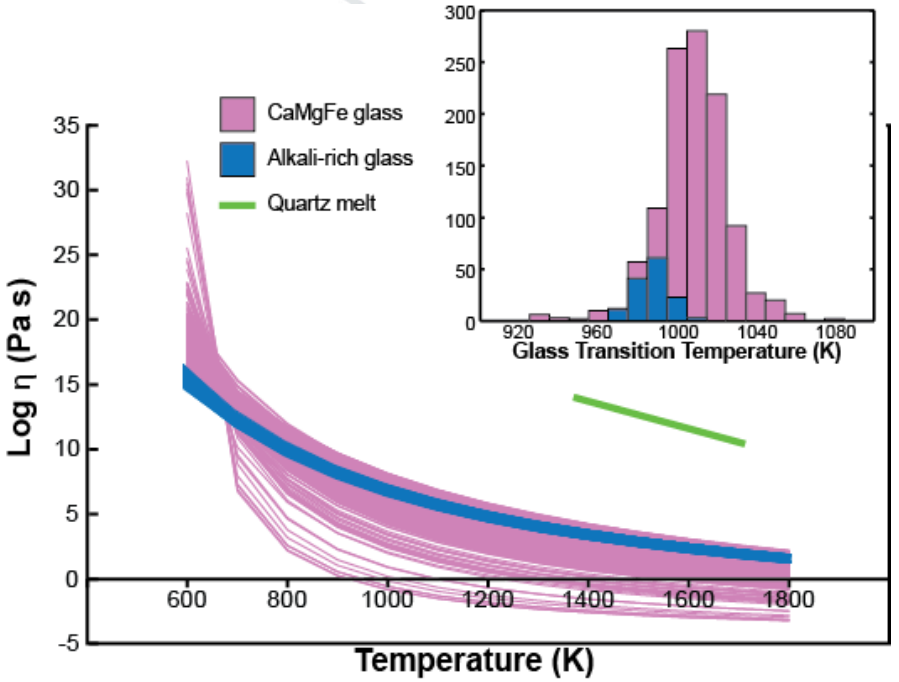
Where is the Radioactivity?



- Activity is strongly correlated with regions of CaMgFe glass, consistent with co-condensation of fissionogenic and nonfissionogenic elements

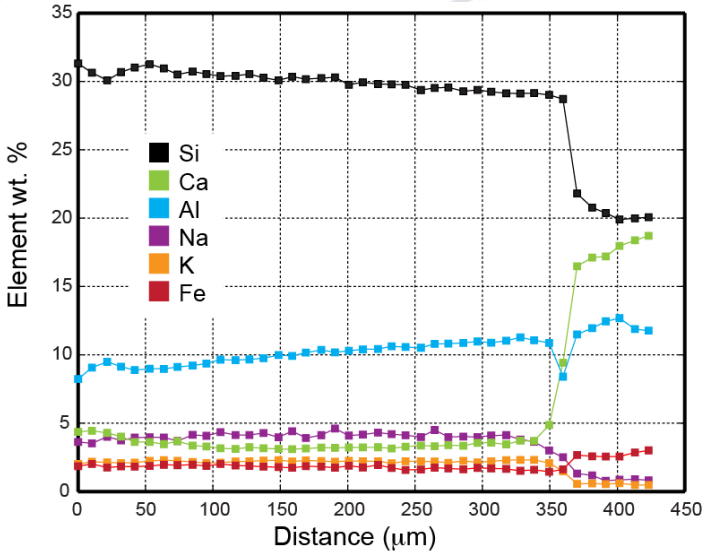
Viscosity & Melt Mixing

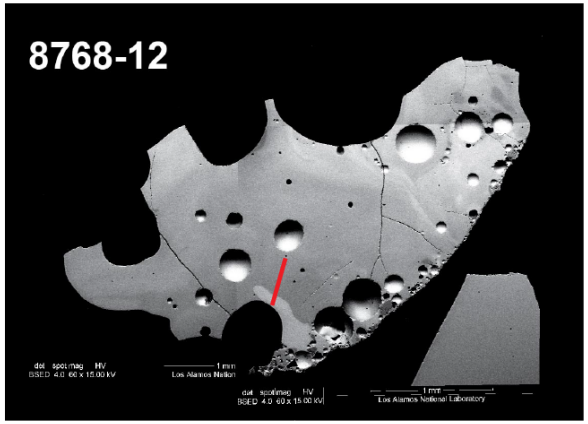


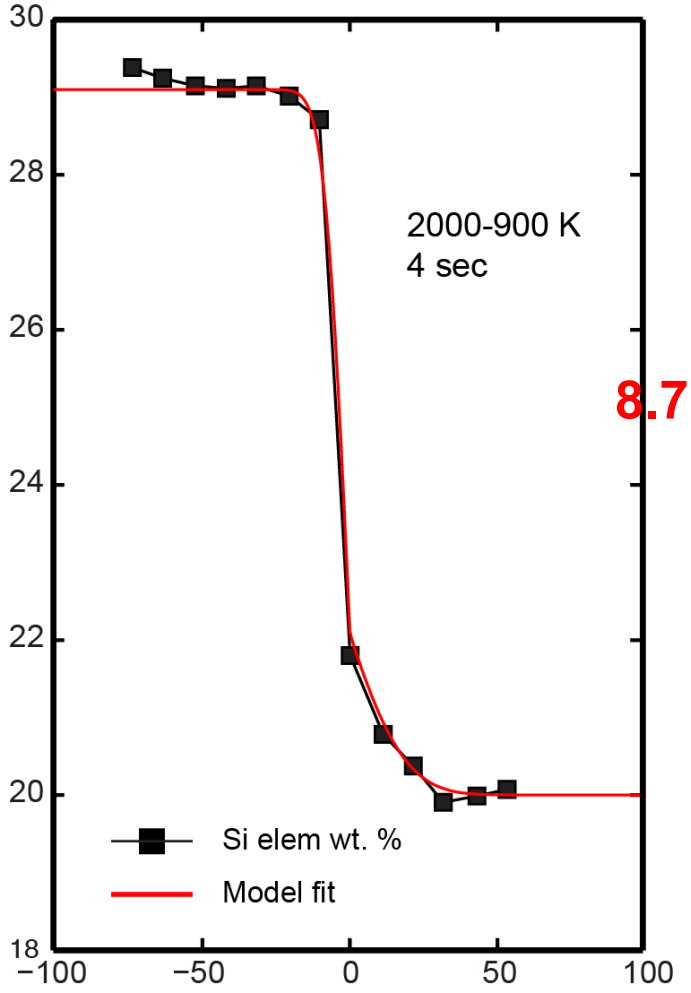


- Viscosity contrast likely limited physical mixing of CaMgFe, alkali, and silica melts

Diffusion & Timescales of Cooling







8.7 x 10^{15} °C/m.y.

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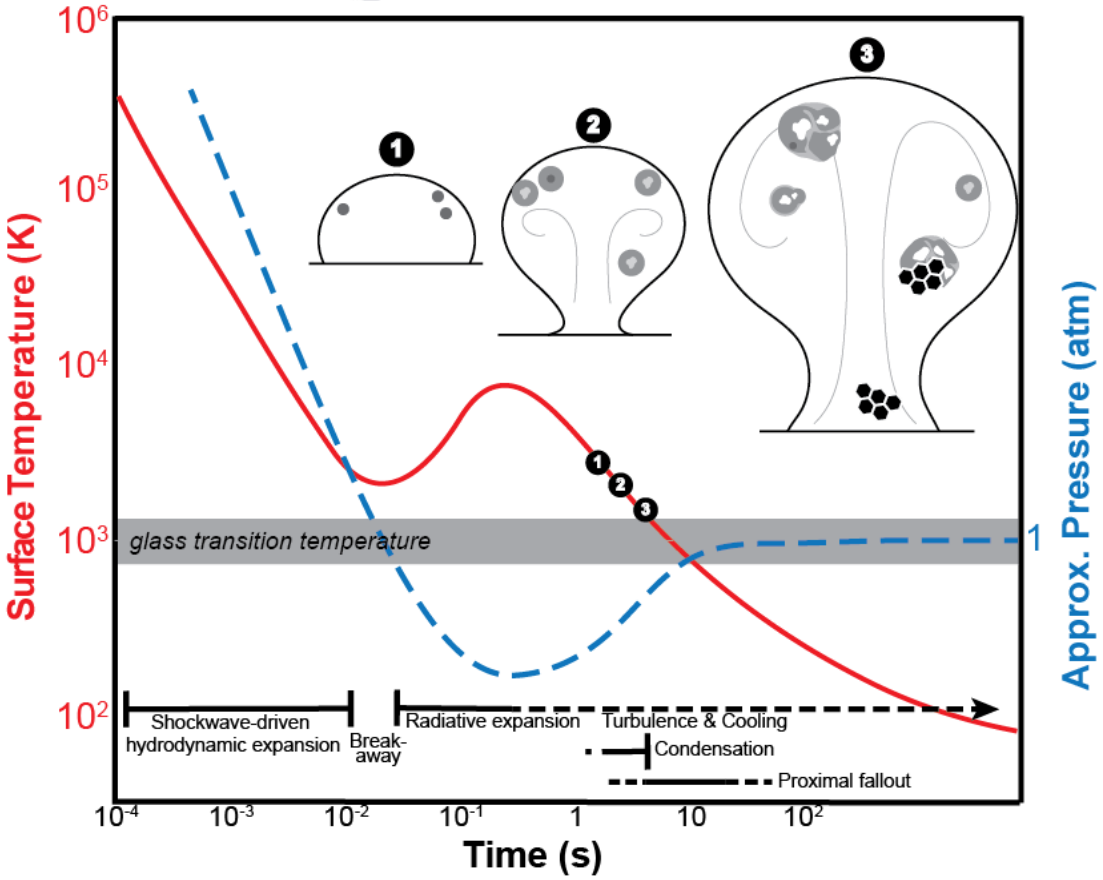
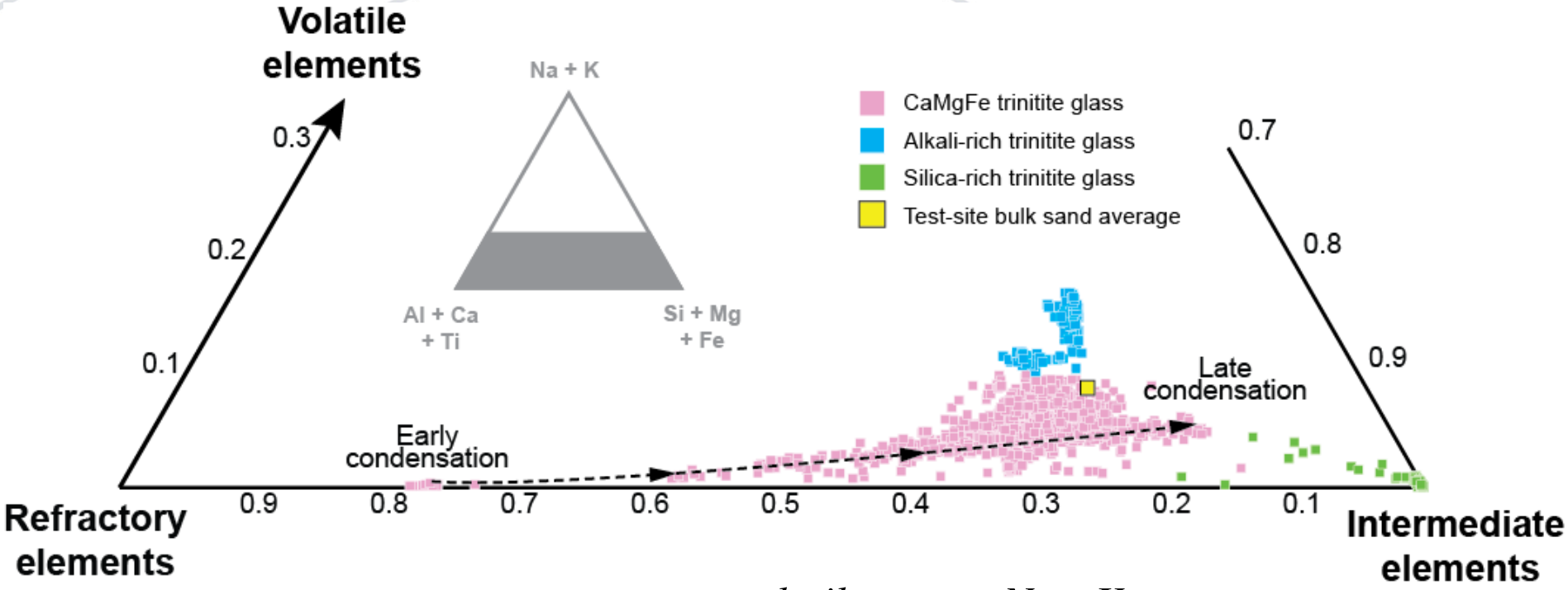
New Fallout Formation Model



- Vaporization of device, tower, sand
- Molten and solid sand entrainment
- Co-condensation of device and ground constituents
- Agglomeration
- Cooling and fallout

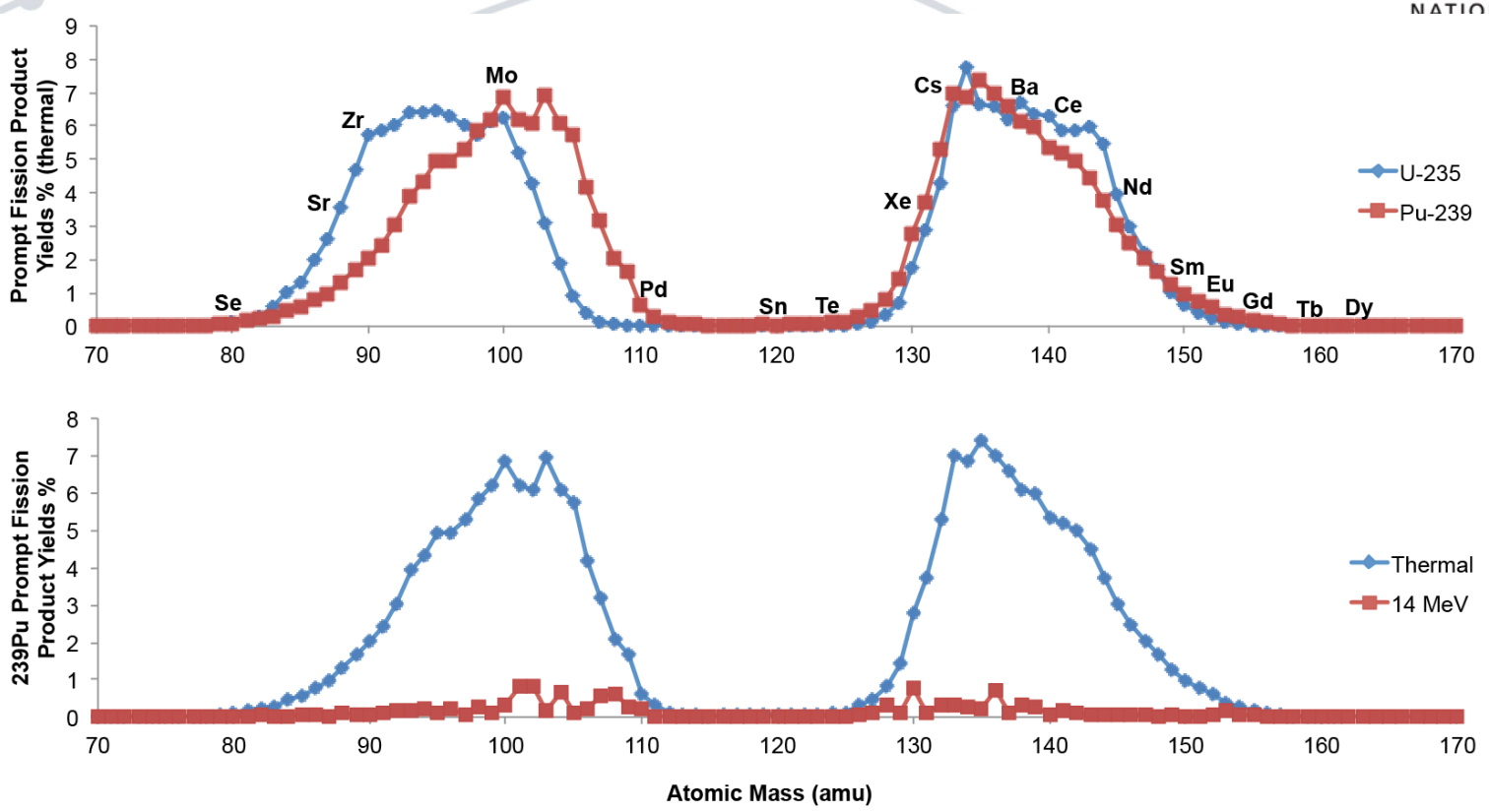
Condensation Chronology & Volatility Scale



$$Volatility\ Index = \frac{volatiles}{refractories} = \frac{Na + K}{Al + Ca + Ti}$$

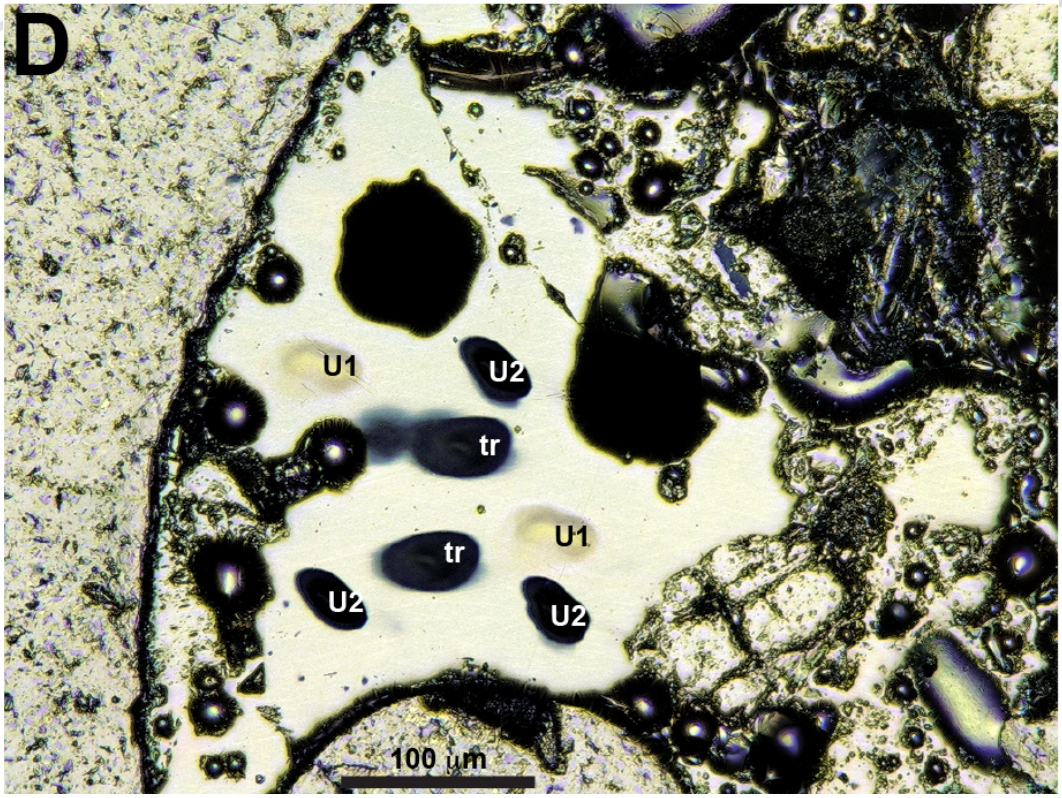
- Define a volatility scale based on the relative amounts of volatile and refractory major elements

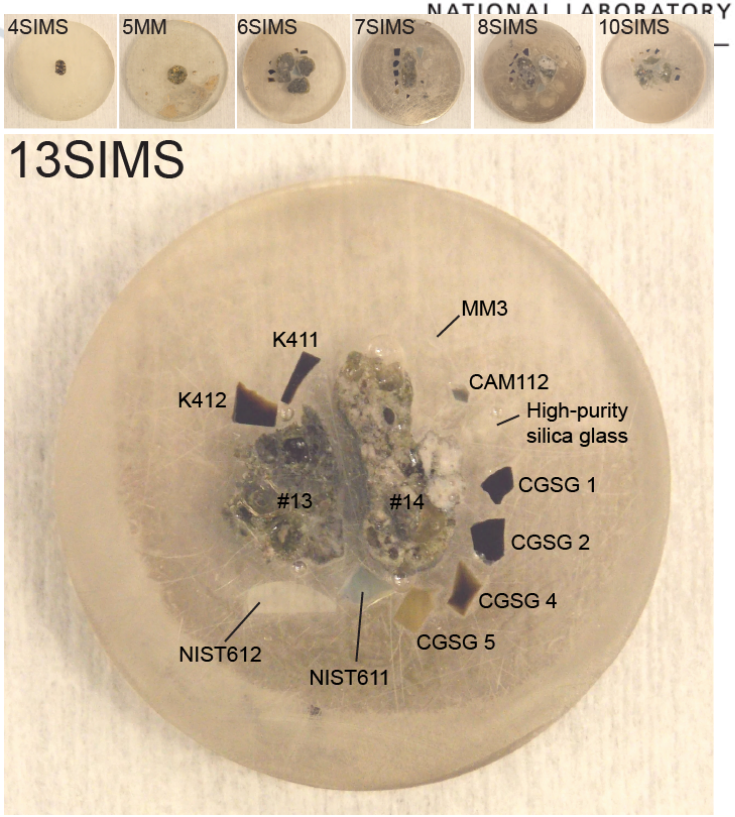
Fissionogenic Nuclides



- Yields of fissionogenic nuclides are related to the type of fuel used and the energy spectrum of neutrons produced

Trace Elements & Isotopes Methods

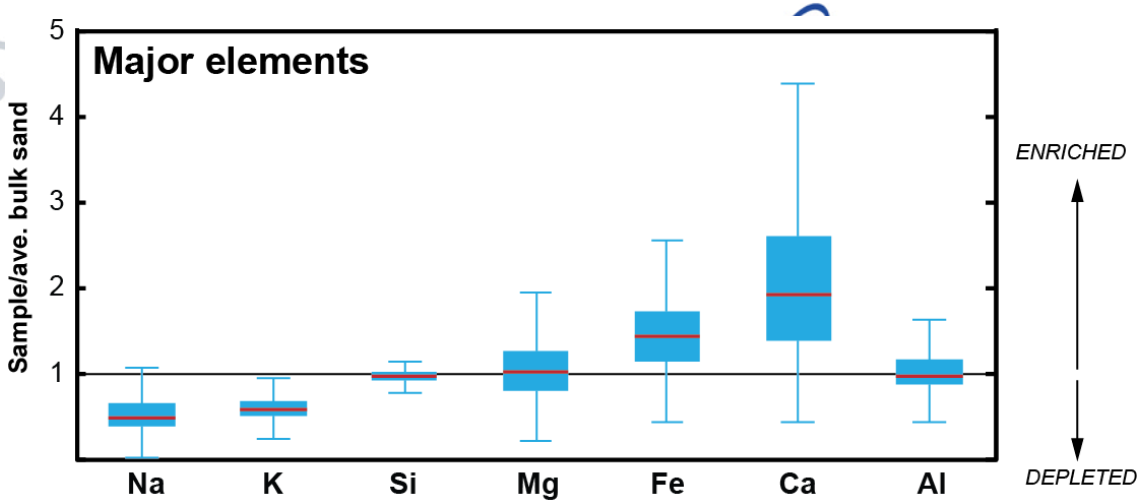


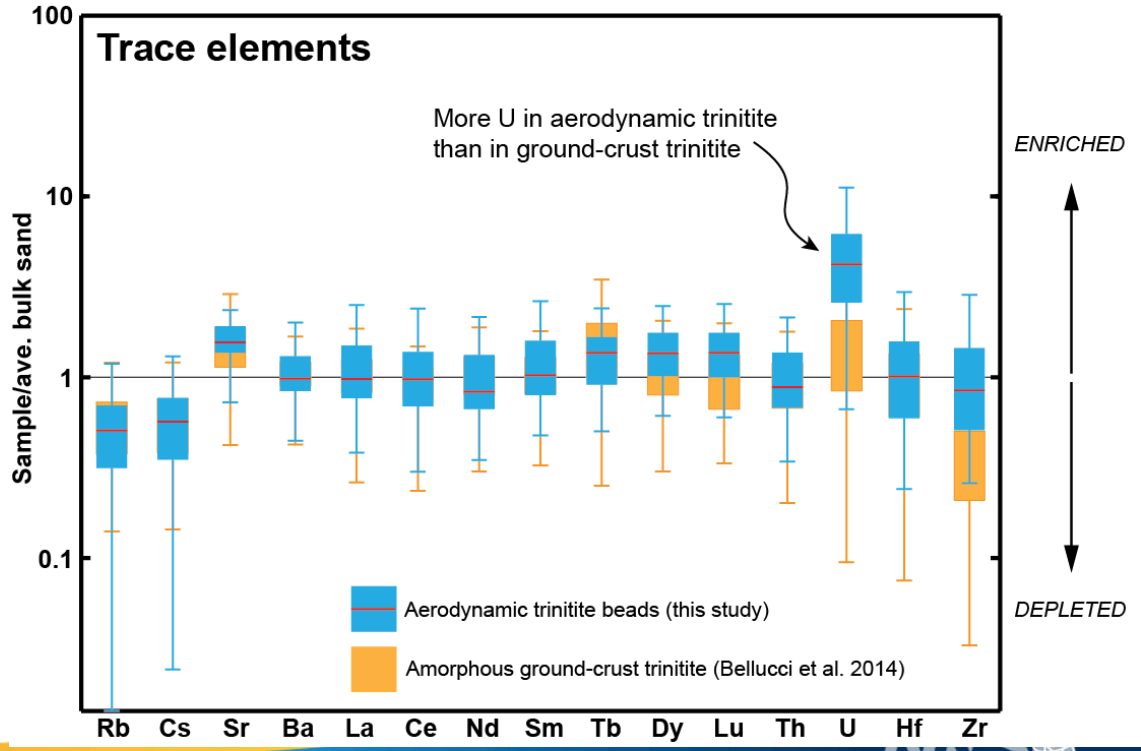


- Cameca IMS 6f SIMS for trace elements
- Cameca IMS 1280 SIMS for U-Pu isotopes

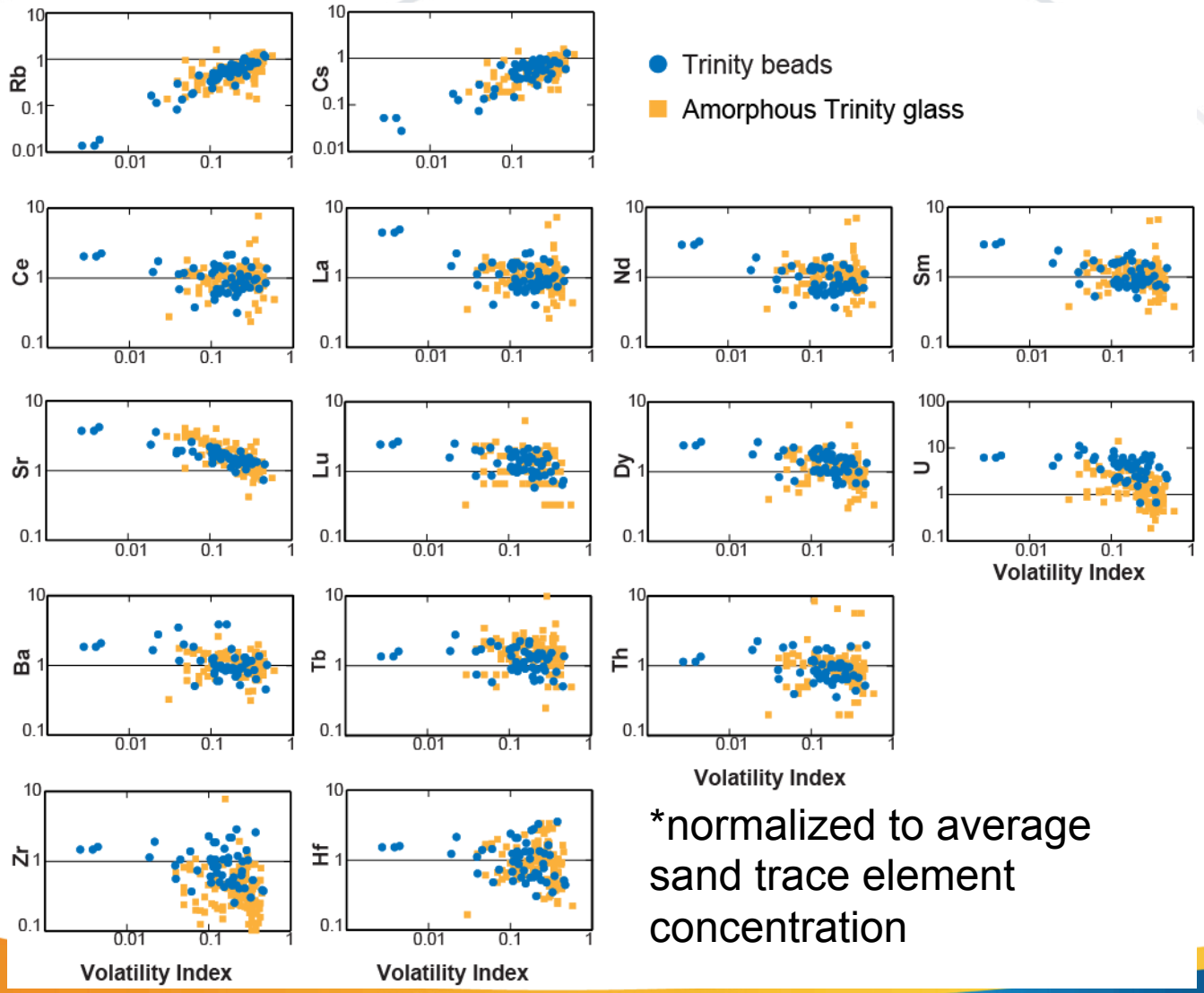
Trace Element Geochemistry

- Similar pattern to major elements
- Enrichment of U in beads relative to ground-crust trinitite





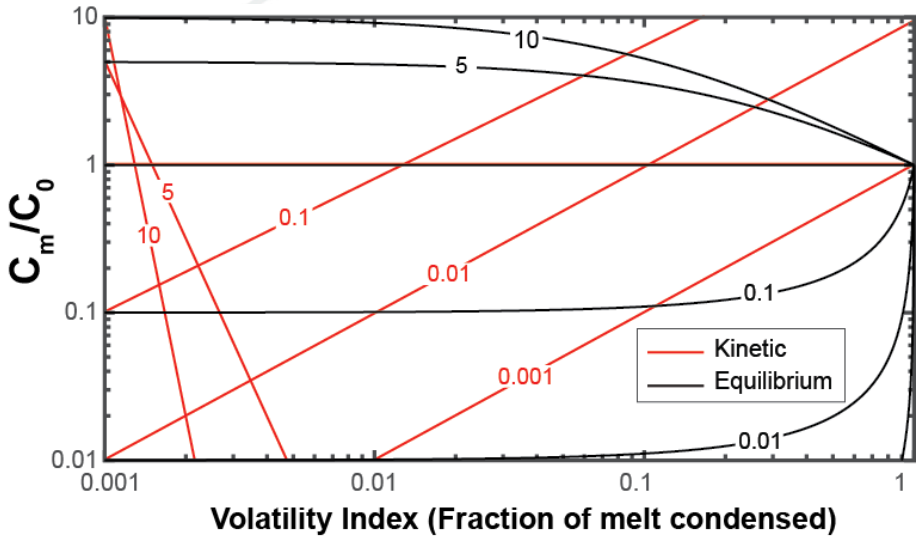
Trace Element Geochemistry

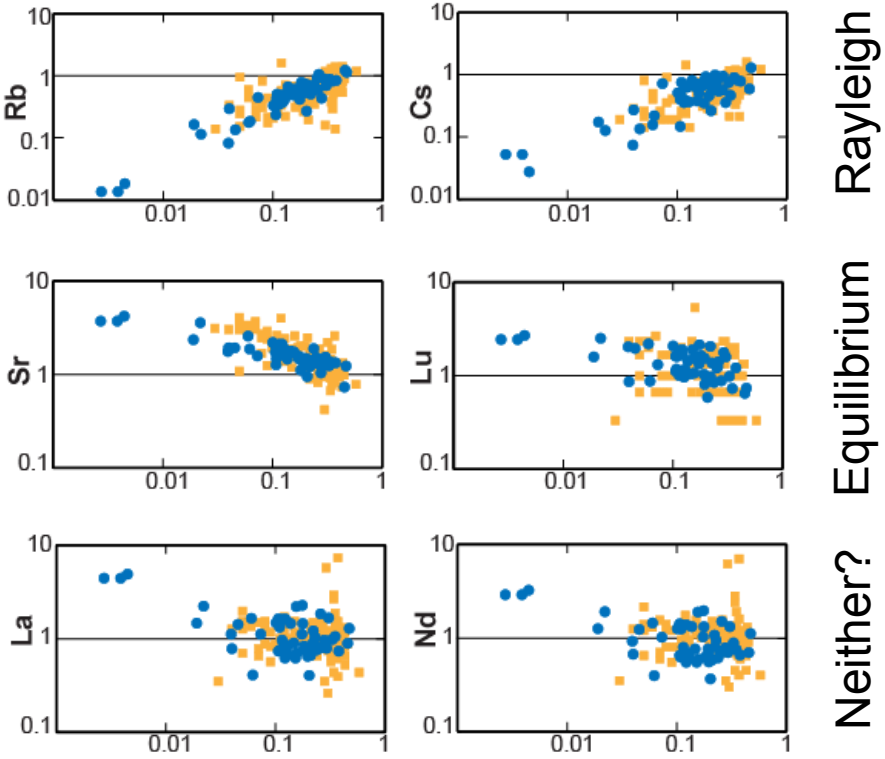


*normalized to average sand trace element concentration

- Coherent increasing or decreasing trends
- Greatest scatter in most refractory elements

Trace Element Geochemistry

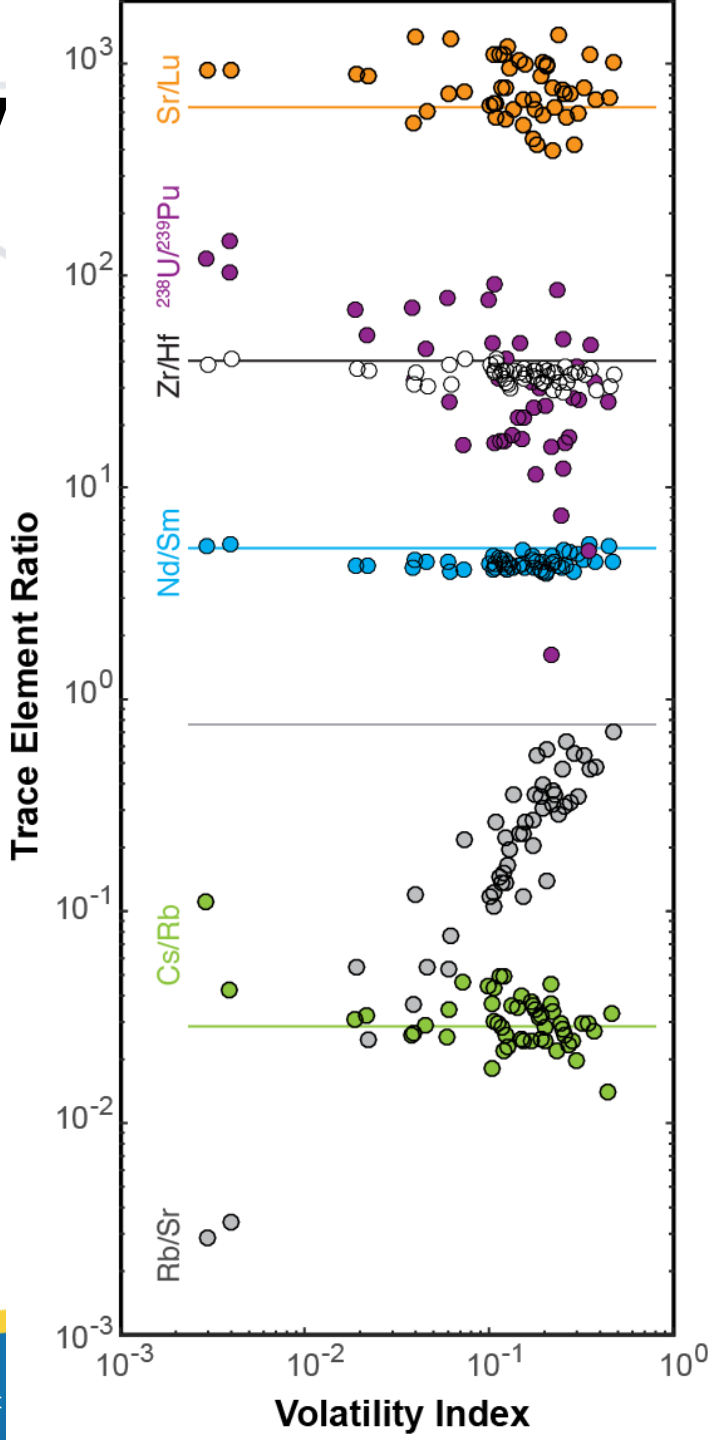




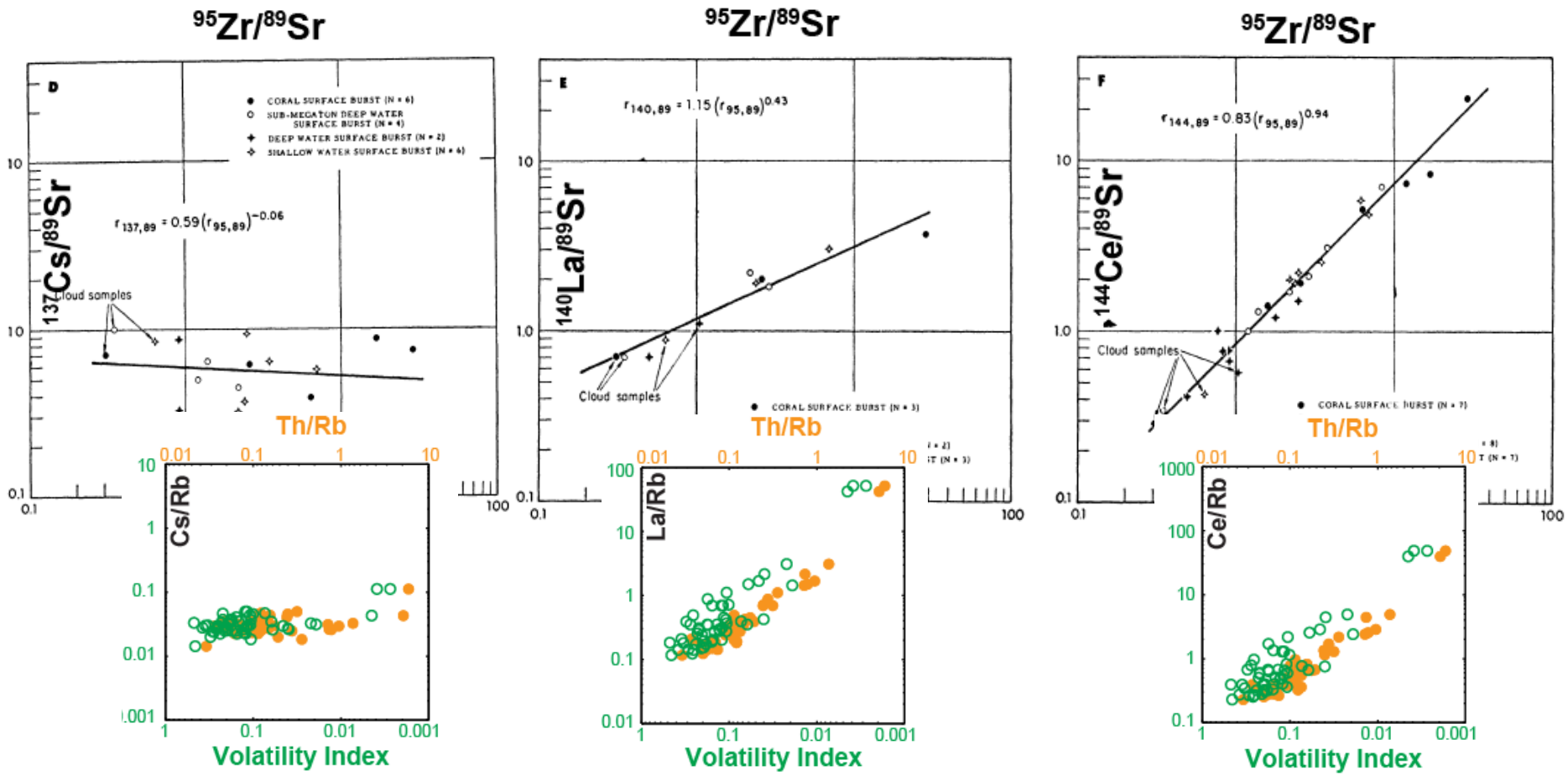
- Modeling suggests volatile elements experienced kinetic fractionation, whereas refractory elements experienced largely equilibrium fractionation

Trace Element Geochemistry

- Track element fractionation through ratios as a function of volatility index
- Strong element fractionations → sloped trends (e.g., Rb/Sr)
- Weak element fractionations → flat trends (e.g., Zr/Hf)



Comparison with Freiling (1961)

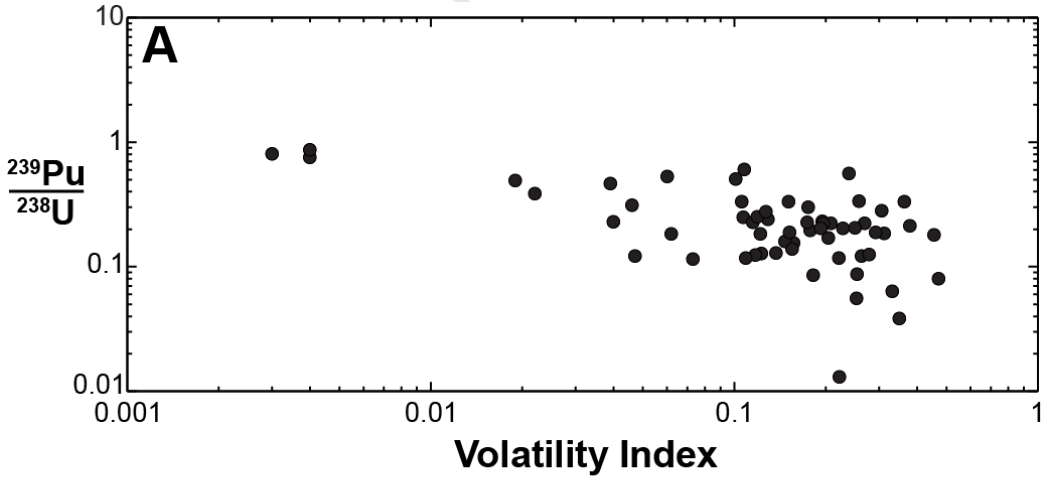


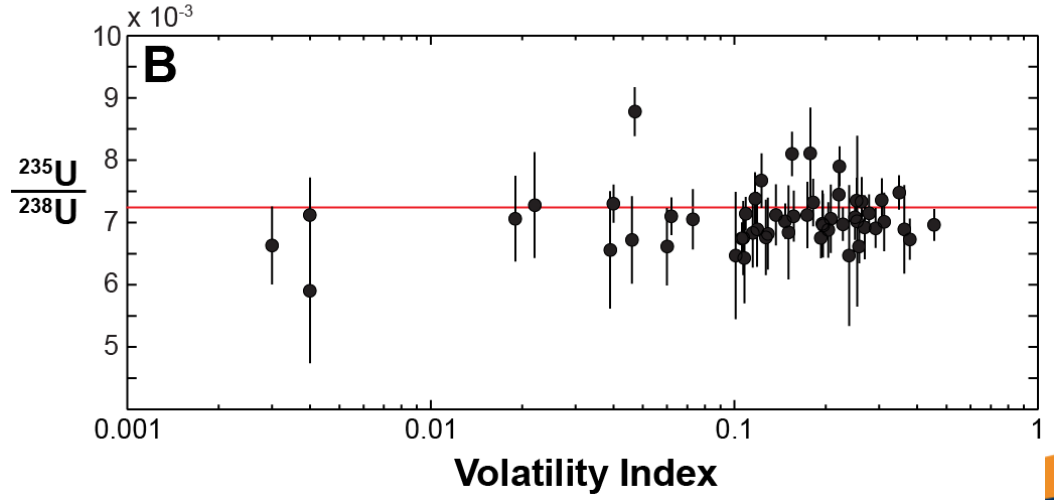
- Stable element-based volatility scales give similar results to fissionogenic isotope-based volatility scales

U-Pu Isotope Geochemistry

- Pu and U fractionated significantly in the Trinity fireball

- U isotopes did not fractionate in the Trinity fireball





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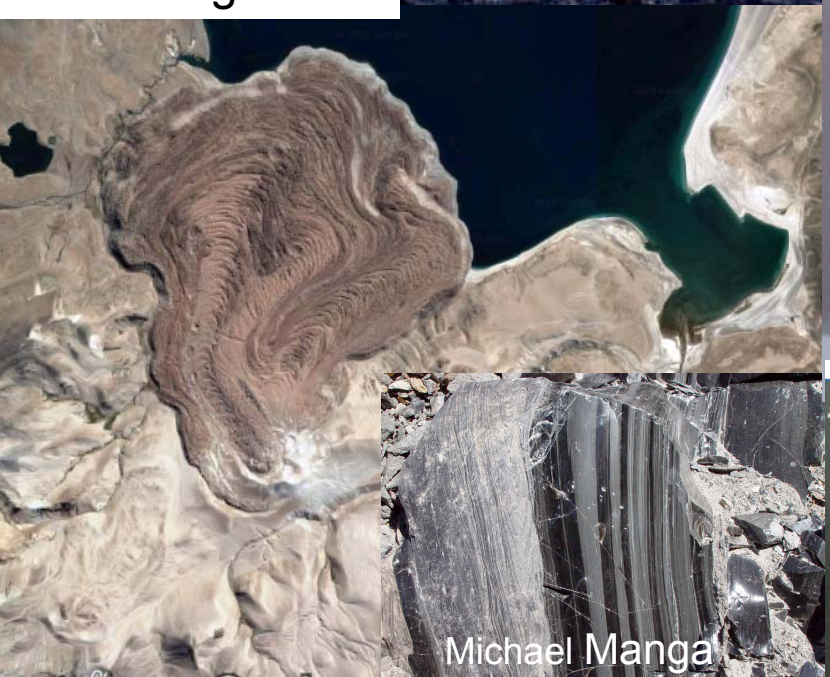
Nuclear Forensics Summary

- Test-site sand vaporized and co-condensed along with radioactive bomb material
- Glassy fallout preserves compositional variations of the nuclear fireball
- Stable elements are appropriate proxies for studying radionuclide fractionation during nuclear explosions
- Heavy isotopes were not fractionated during the Trinity explosion

Natural Glasses

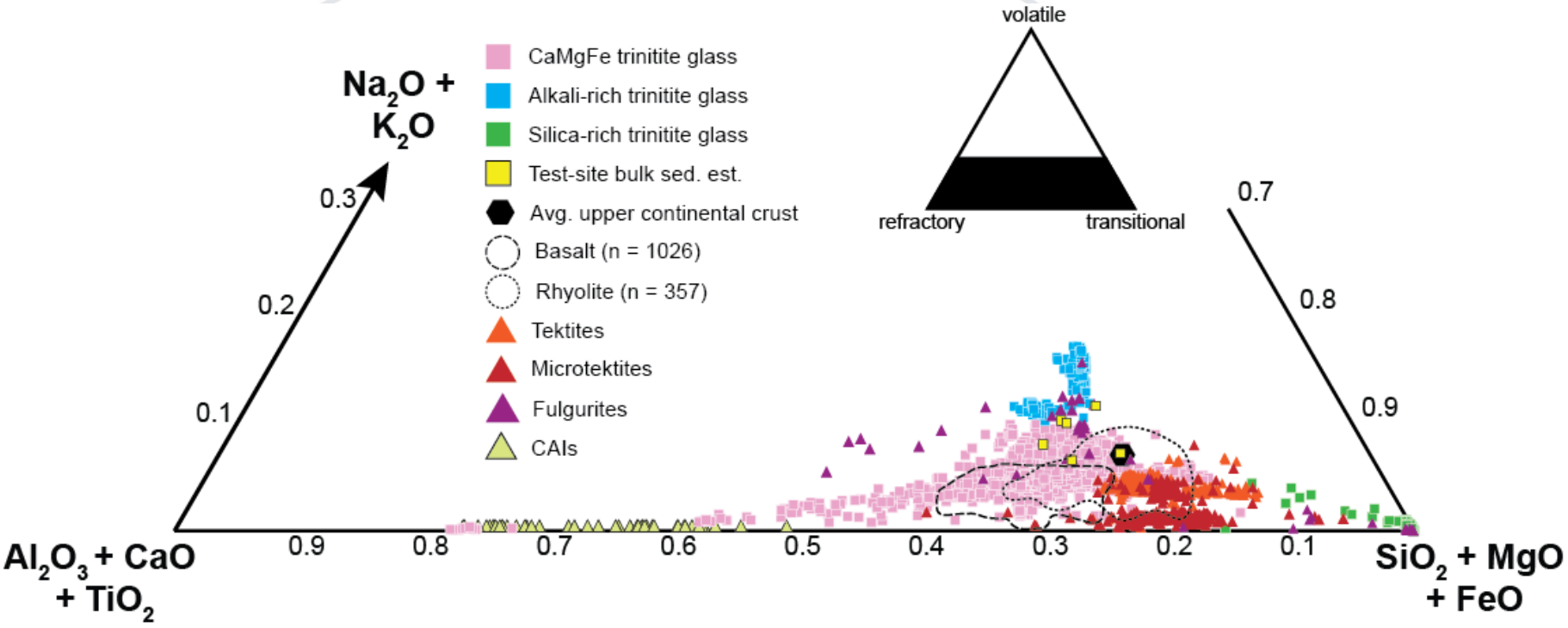
Volcanic glasses



Impactites

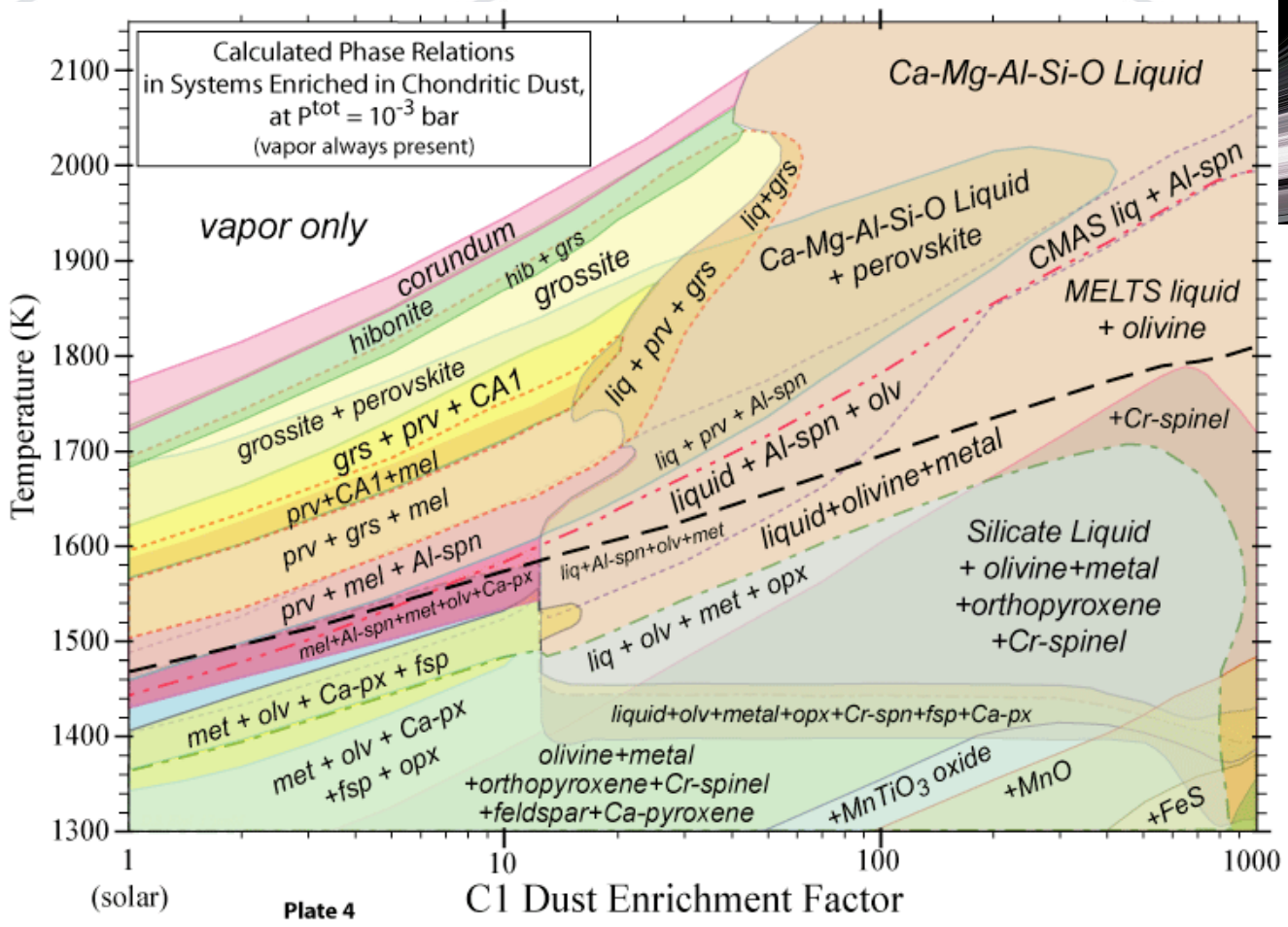
Fulgurites

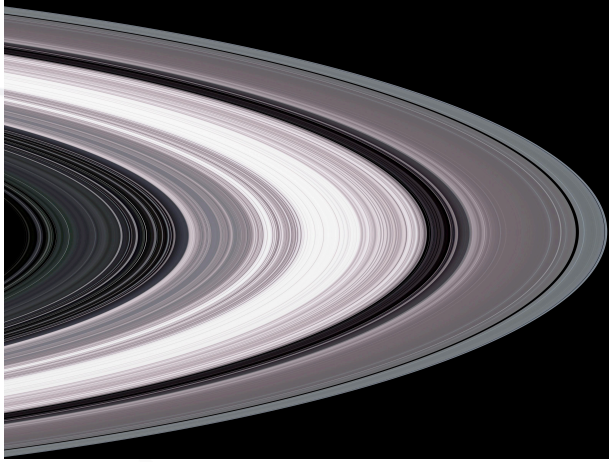
Natural Glasses



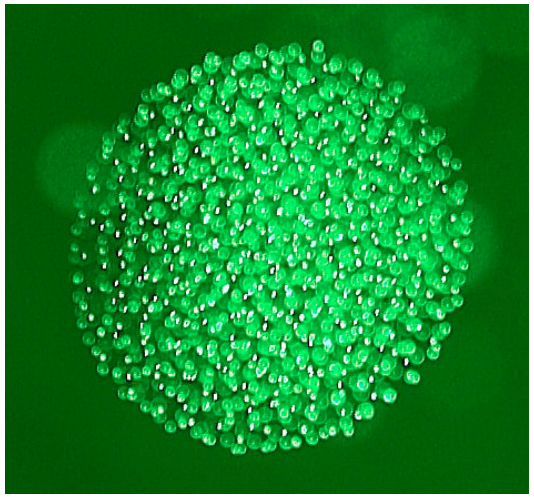
- Naturally formed glasses show a more limited compositional range than trinitite

Dusty Plasmas





NASA/JPL



Yukawa ball
Kading & Melzer 2006

Geochemistry Summary

- Naturally formed glasses show a more limited compositional range than trinitite

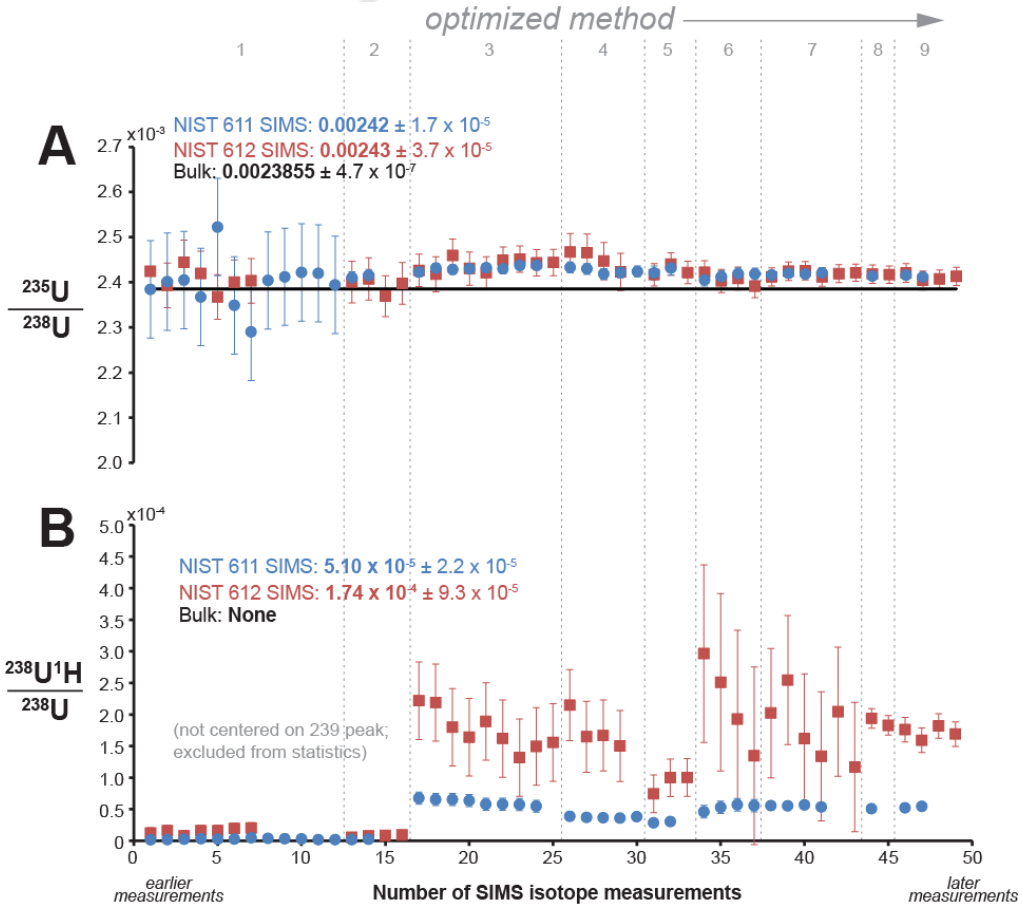
- Melting and vaporization behavior of natural glasses differed from those of trinitite, reflecting more heterogeneous energy distributions or greater physical fractionation

- Highly refractory fallout glass may be a process analogue for early solar system condensates like CAIs

Questions?

- This work was supported by:
  - A Los Alamos National Lab Seaborg-Strategic Outcomes Office Postdoctoral Fellowship to C. Bonamici
  - Office of Defense Nuclear Nonproliferation R&D Program (NA-22), through W. Kinman

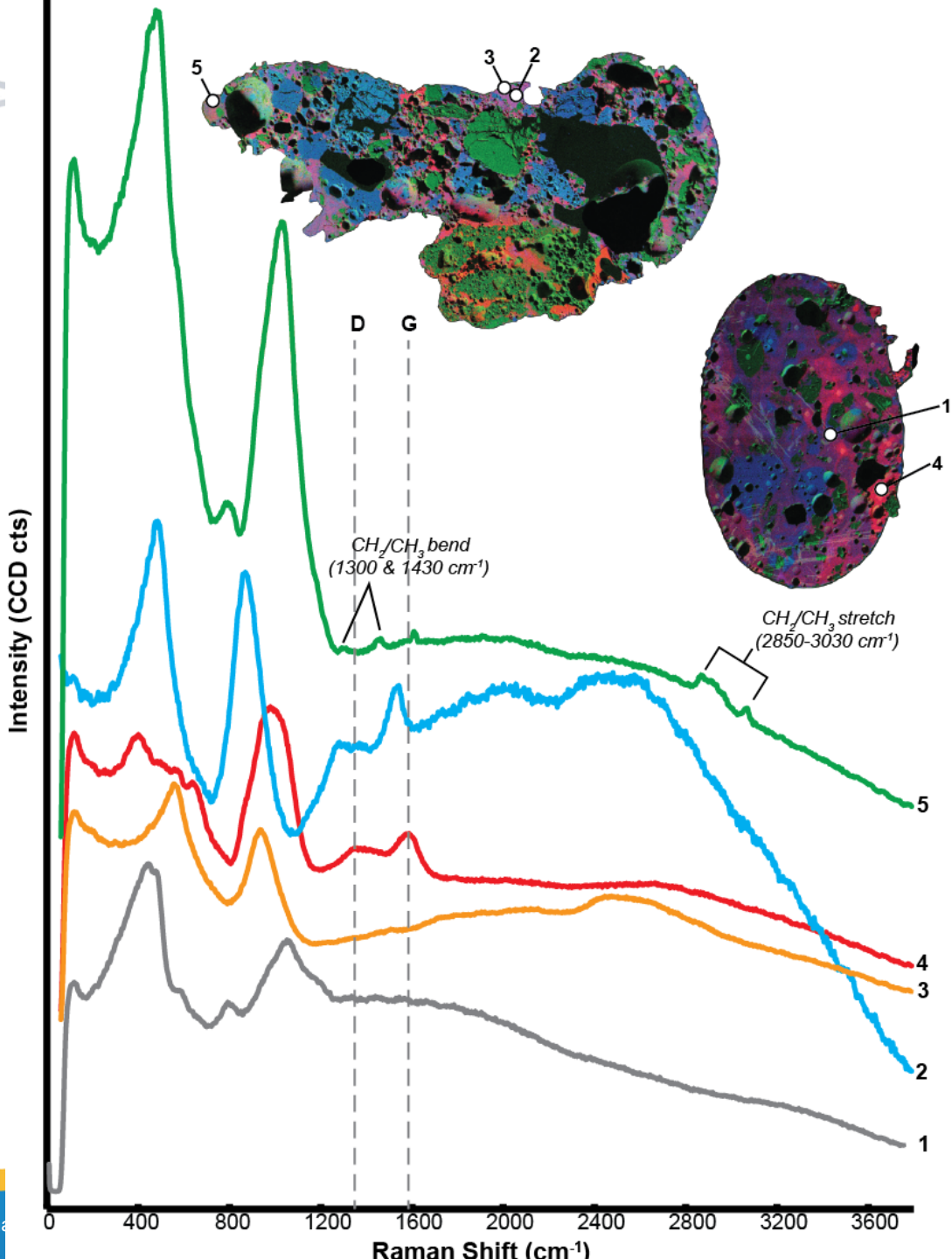
U-Pu Isotope Measurements



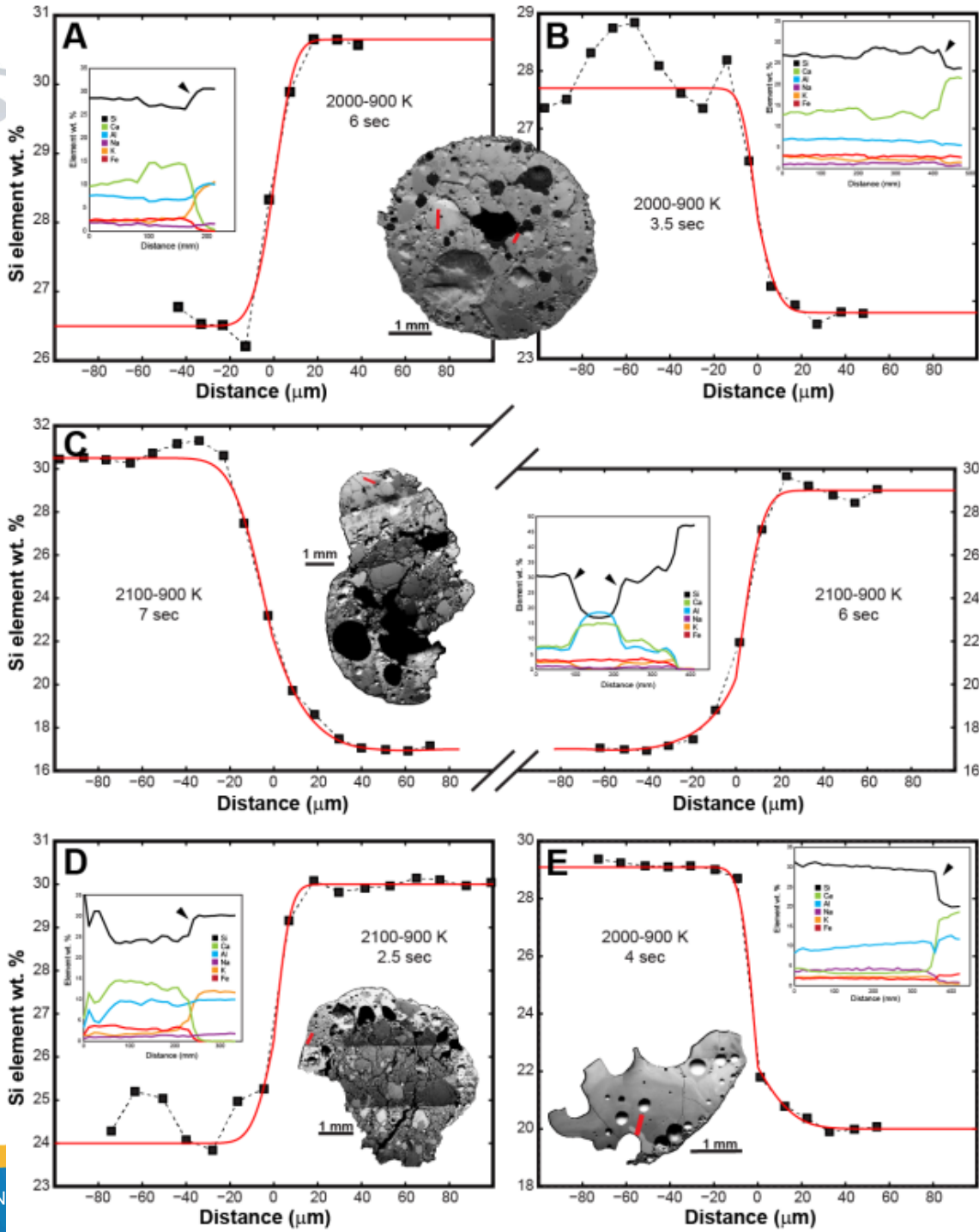
- Cameca IMS 1280
  - Static multicollector
  - ^{234}U, ^{235}U, ^{236}U, ^{238}U, 239
  - O^- primary beam
  - 420 cycles, 32 min
  - 2200 MRP
- NIST 611, NIST 612
- Hydride correction for $^{238}U^{1}H$ on ^{239}Pu
- Ingrowth correction for $^{239}Pu \rightarrow {}^{235}U$

Oxidation State in the Fireball

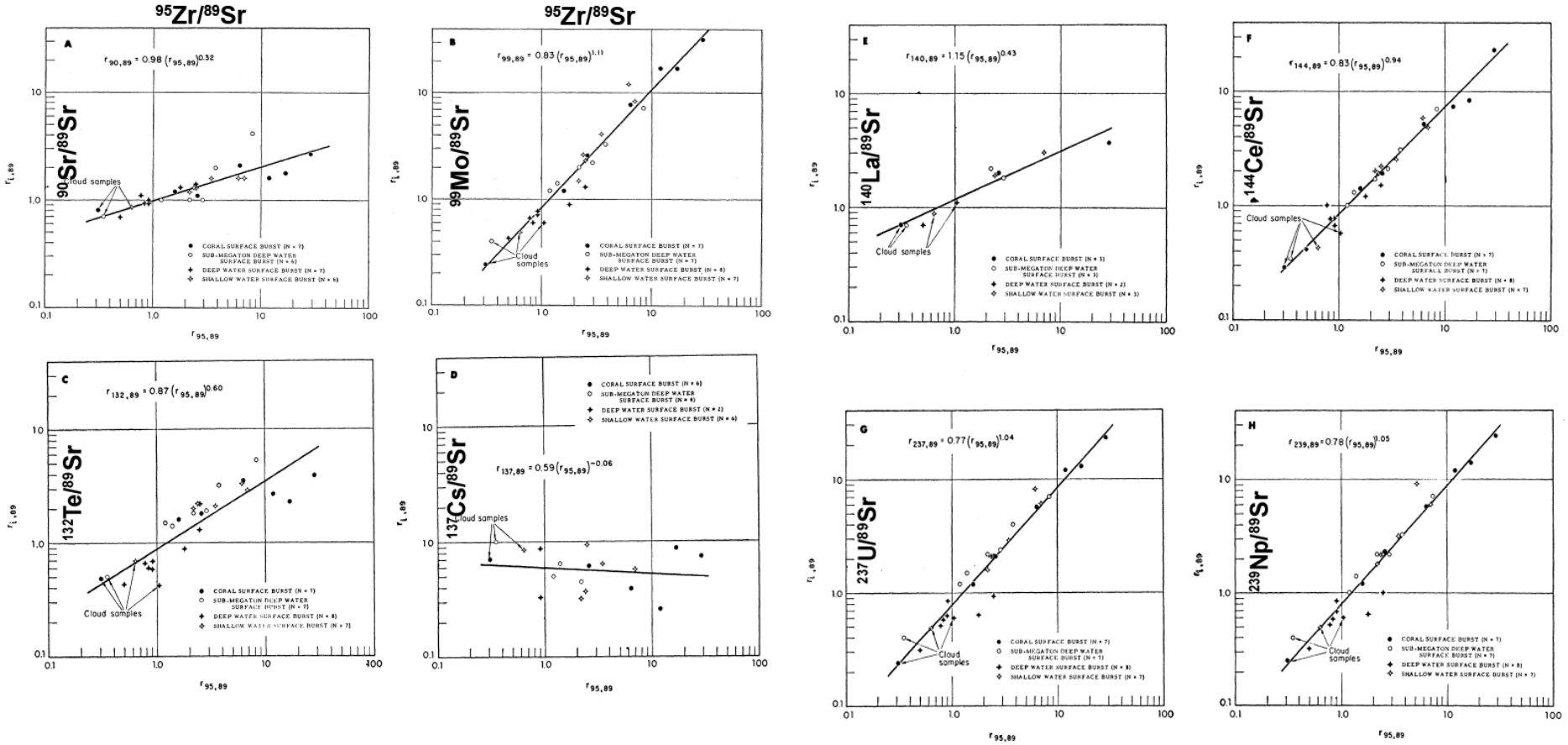
Laser Raman spectroscopy



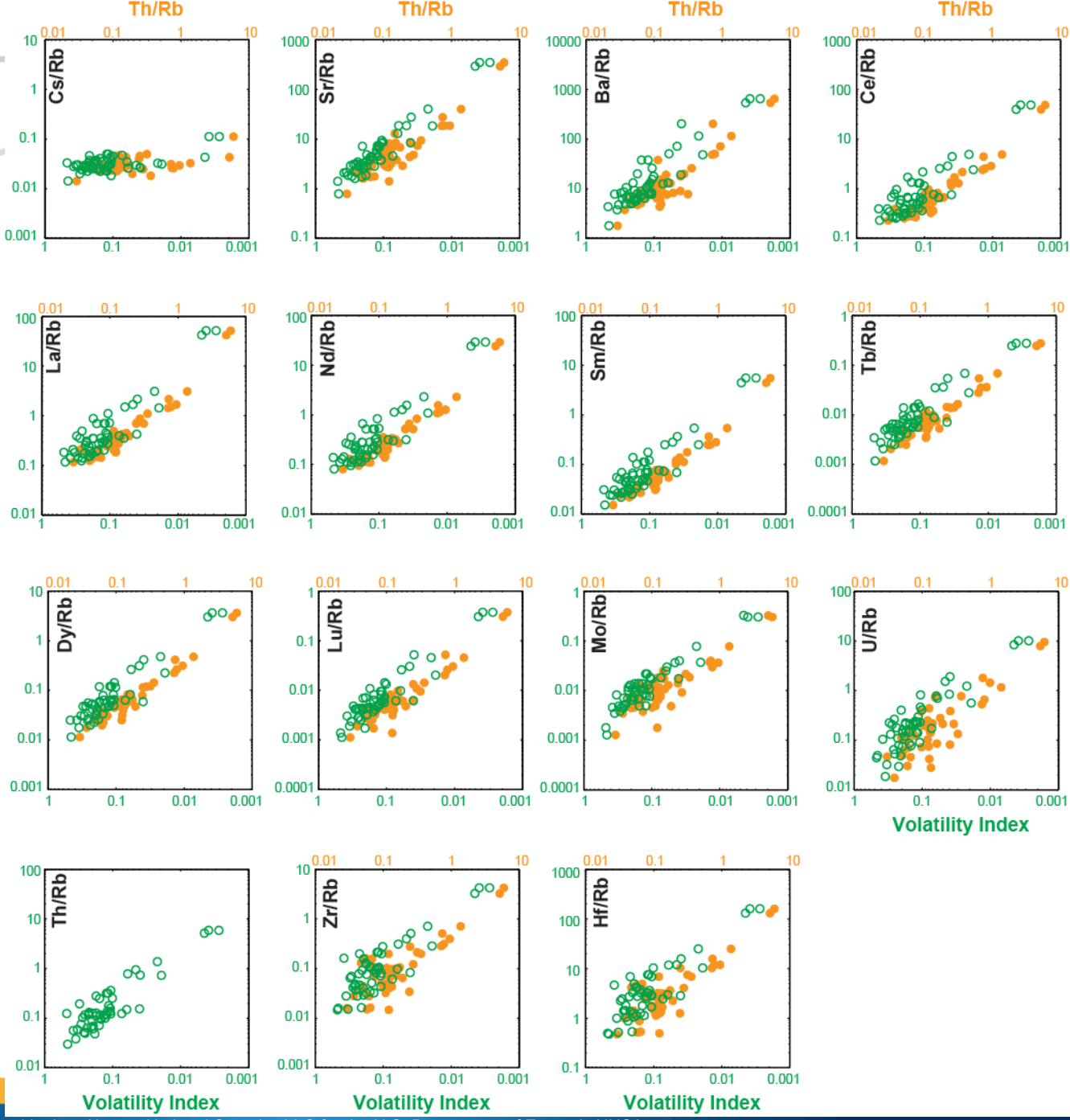
More Diffusion Modeling



Freiling 1961



UNCLASSIFIED


Th/Rb
Cs/Rb
Sr/Rb
Ba/Rb
Ce/Rb
La/Rb
Nd/Rb
Sm/Rb
Tb/Rb
Dy/Rb
Lu/Rb
Mo/Rb
U/Rb
Th/Rb
Zr/Rb
Hf/Rb
Volatility Index