U.S. Department of Energy
Office of Civilian Radioactive Waste Management

USGS Fracture Mineral Research - Fluid Inclusion Studies

Presented to:
Nuclear Waste Technical Review Board - Spring Meeting, 2001

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Date: May 9, 2001
Place: Washington, D.C.

YUCCA MOUNTAIN PROJECT

USGS Environmental Science Team

- This was a team effort involving:
  - Zell Peterman - Sr isotopes & thermal modeling
  - James Paces - geology & geochronology
  - Leonid Neymark - geochronology
  - Brian Marshall - Sr isotopes & thermal modeling
  - Edwin Roedder - fluid inclusions

History of USGS Secondary Mineral Studies

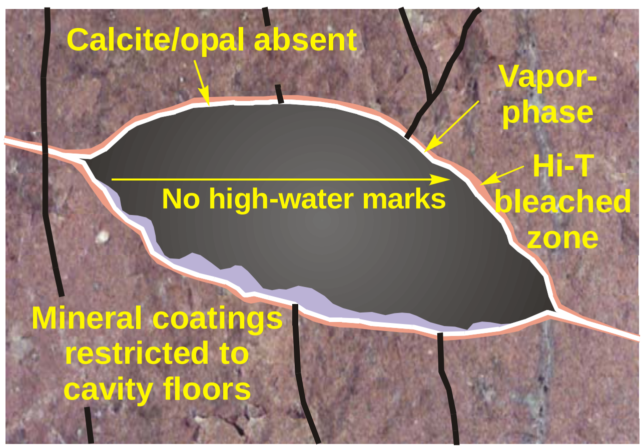
- Pre-1995: Only samples were from drill core. Data include mineralogy/petrology, stable isotopes, Sr isotopes, initial attempts at geochronology
- Since ESF construction in 1995, underground access has provided better samples and a better defined geologic context for more comprehensive petrographic, geochemical and geochronological studies of secondary minerals in the unsaturated zone (UZ)

Fracture surface

Lithophysal cavity floor

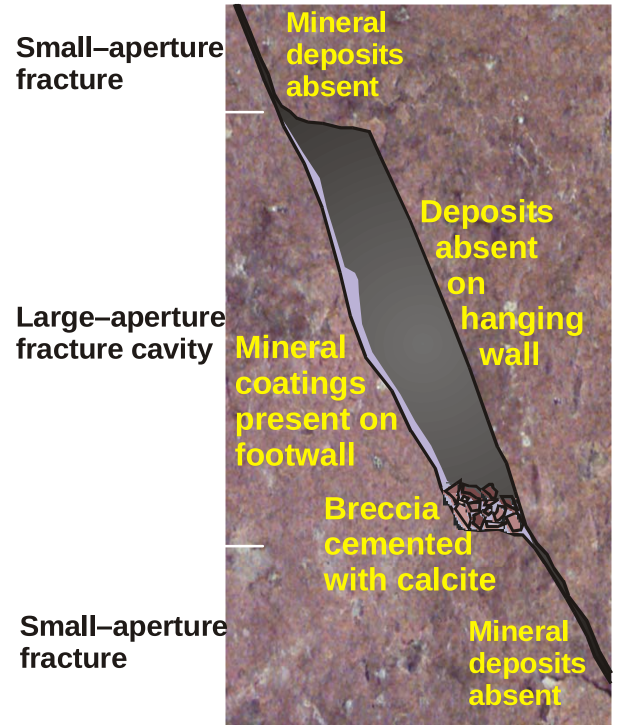
Location of Calcite — Opal Coatings Within Rock Cavities

- **Most deposits are present in ROCK VOIDS:**
  - **Fracture footwall surfaces**
  - **Floors of lithophysal cavities**
  - **Breccia cements**



Lithophysal Cavities

- Calcite and opal coatings
- Host tuff
- Void space



Fracture Cavities

Filled veins are present, but they are:
- **Usually less than several mm thick**
- **Volumetrically minor**

- **UZ coatings demonstrate gravity-driven flow of percolating waters**

(see supplemental slide 1 for more details)

Location of Calcite — Opal Coatings Within Rock Cavities

(Continued)

- <10% of fractures and cavities contain secondary calcite and silica minerals*
  - Percolating water followed connected pathways downward along fracture footwalls and cavity floors
- Geologic evidence indicates that cavities have remained hydrologically unsaturated since tuff emplacement
- In contrast, hydrothermal calcite deposited in fractures and cavities below the water table 10-12 Ma completely coated or filled those open spaces

*see supplemental slide 2 for more details

Depositional Model Based on Earlier Studies

$\delta^{13}C$, $\delta^{18}O$, $\delta^{87}Sr$, and REE analyses of calcite and U-series and U-Pb geochronology of silica minerals from the UZ and from calcretes in the overlying soils are consistent with the deposits having formed:

- In an unsaturated zone setting
- From downward percolating water of meteoric-infiltration origin
- Along focused flowpaths that bypassed many potential flowpaths and depositional sites
- Over a long depositional period, from at least 10 Ma, possibly since the tuffs cooled to <100°C

THESE DATA PRECLUDE FORMATION OF SECONDARY MINERALS FROM UPWELLING GROUND WATER.

(see supplemental slide 3 for more details)

Design of Fluid Inclusion Research

- In 1996, the State's scientists argued that elevated temperature (35-80°C) fluid inclusions (FI) in calcite were conclusive evidence of deposition from upwelling hydrothermal fluids
- NWTRB reviewed this work and recommended additional studies to assess the State's FI observations
- DOE requested a joint study with geoscientists from the State, the UNLV, and the USGS as participants, to:
  - Determine the spatial distribution of elevated temperature FIs
  - Measure the range of FI temperatures
  - Establish a temporal framework of FI formation
    - Relative: by defining paragenetic sequence
    - Absolute: by geochronometry of associated minerals
- All data collection to be from a shared set of participant-collected samples

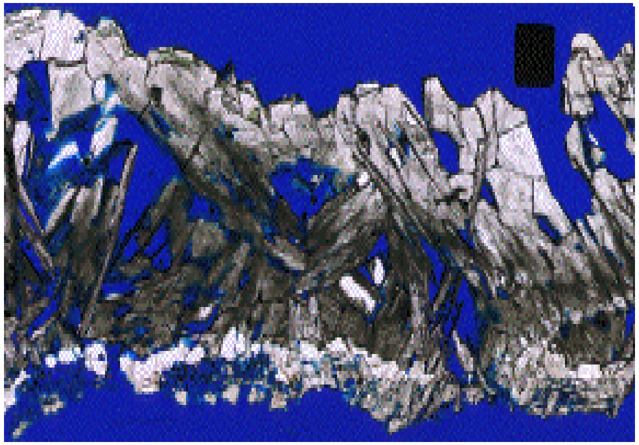
Secondary Mineral Paragenetic Sequence

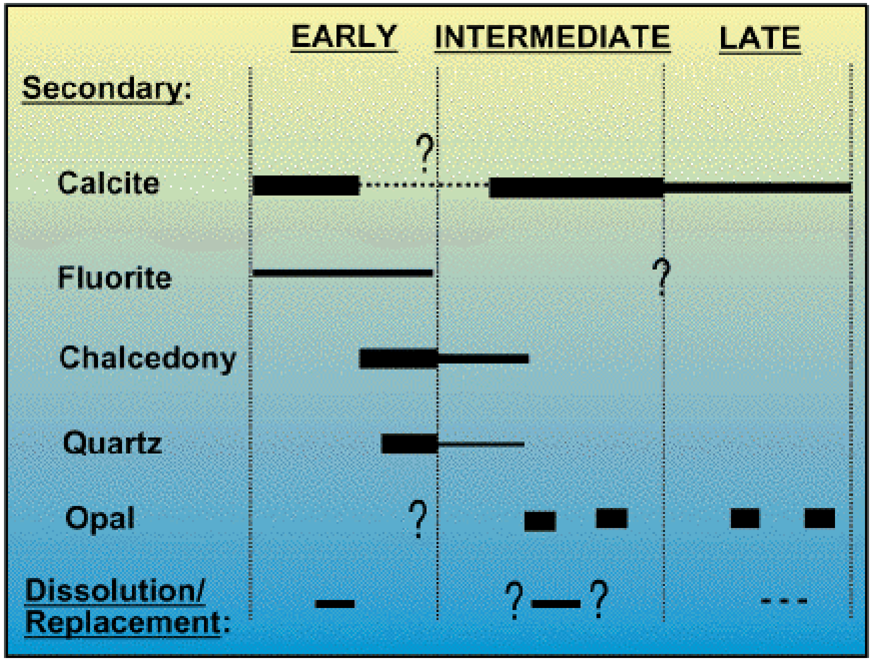
Mineral sequences can be divided into early, intermediate and late stages:

Early stage: calcite ± fluorite ± zeolites and commonly capped by chalcedony/quartz

Intermediate stage: calcite, commonly bladed and growth-zoned ± opal ± chalcedony/quartz (early part?) ± (fluorite?)

Late stage: calcite ± opal, mostly as overgrowths on earlier stage minerals, especially on the tips of calcite blades



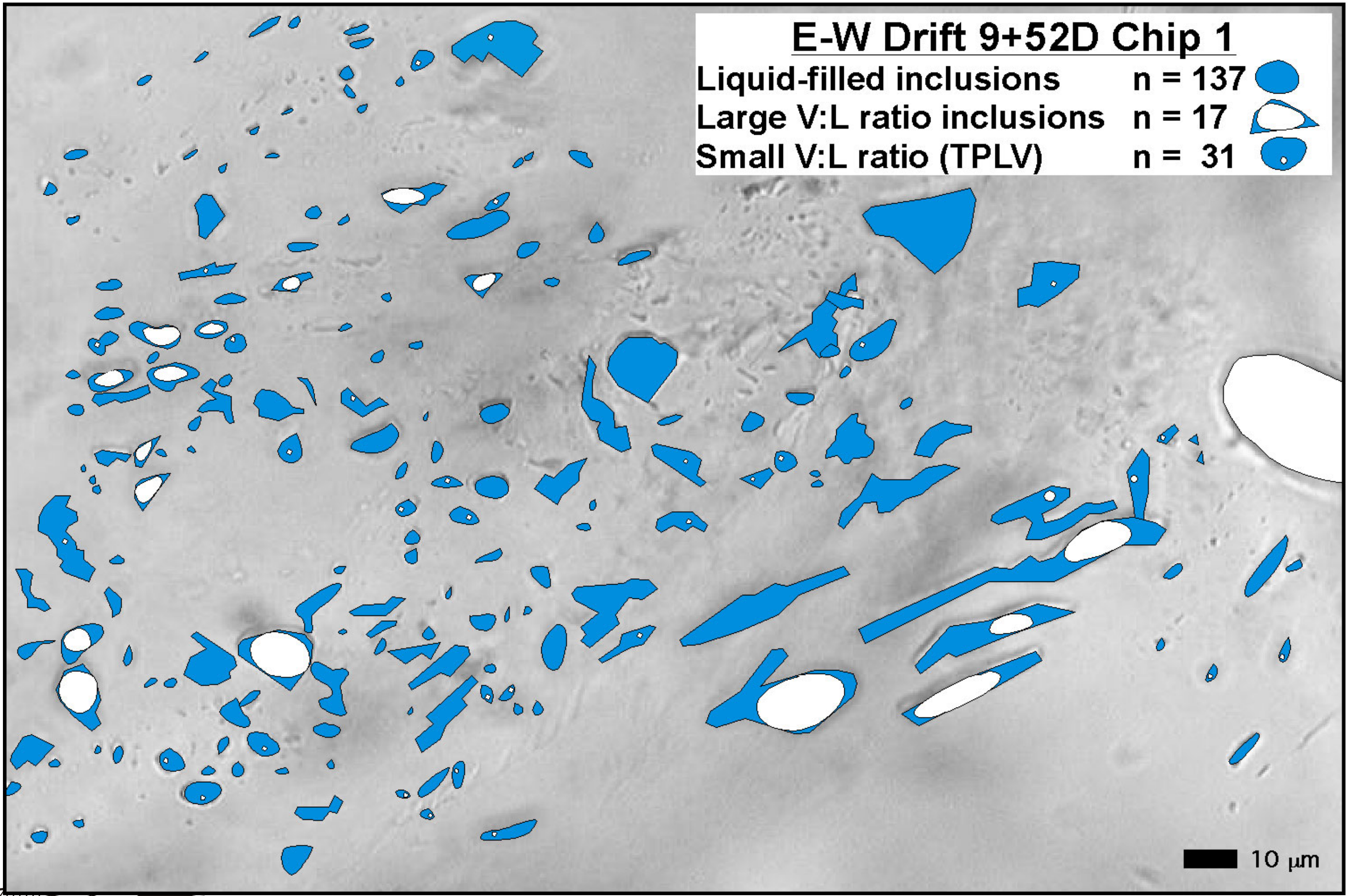


Fluid Inclusions

- Fluid inclusions of 3 types based on vapor (V) to liquid (L) ratios
  - All liquid (V:L = 0)
  - Highly variable vapor content (including all vapor) (V = 5-100%)
  - Small and consistent V:L (V ~ 1%)
    - suitable for temperature measurements
- Fluid inclusion assemblages (FIA) suitable for measuring temperatures occur in ~50% of sample localities
- But, with respect to the total number of FIAs observed, those including FIs suitable for temperature measurement are rare (probably considerably <1% of the total)
- FIAs with inclusions suitable for temperature estimates are found predominantly in early-stage calcite but locally in intermediate-stage calcite

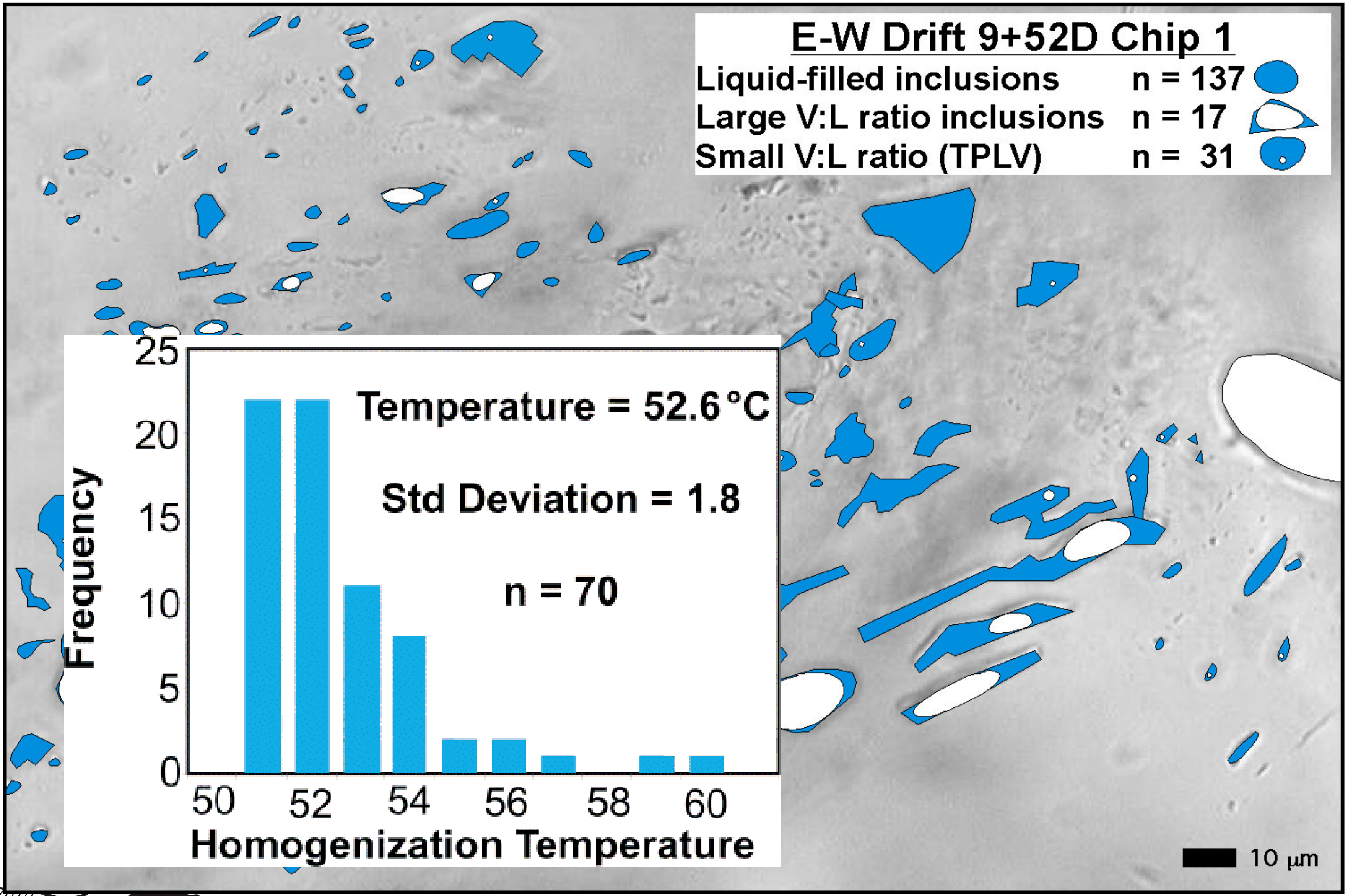
Fluid Inclusion Assemblage - I

FIAs containing all-L, vapor-rich, and small and consistent V:L ratio inclusions, as shown in this photomicrograph, are common.



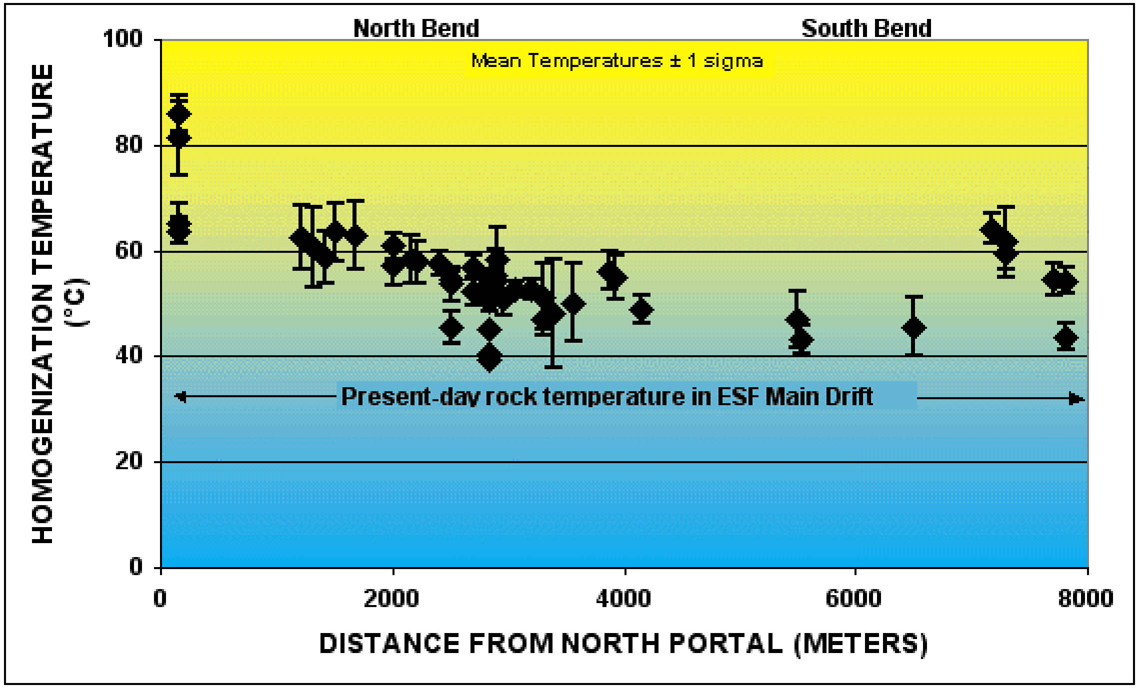
Fluid Inclusion Assemblage - II

The fluid inclusions with small and consistent V:L ratios commonly display very consistent homogenization temperatures within FIAs



Fluid Inclusion Homogenization Temperatures

Fluid inclusion homogenization temperatures in the ESF-ECRB generally range between 40 and 65°C with the higher temperatures from North and South Ramp localities.



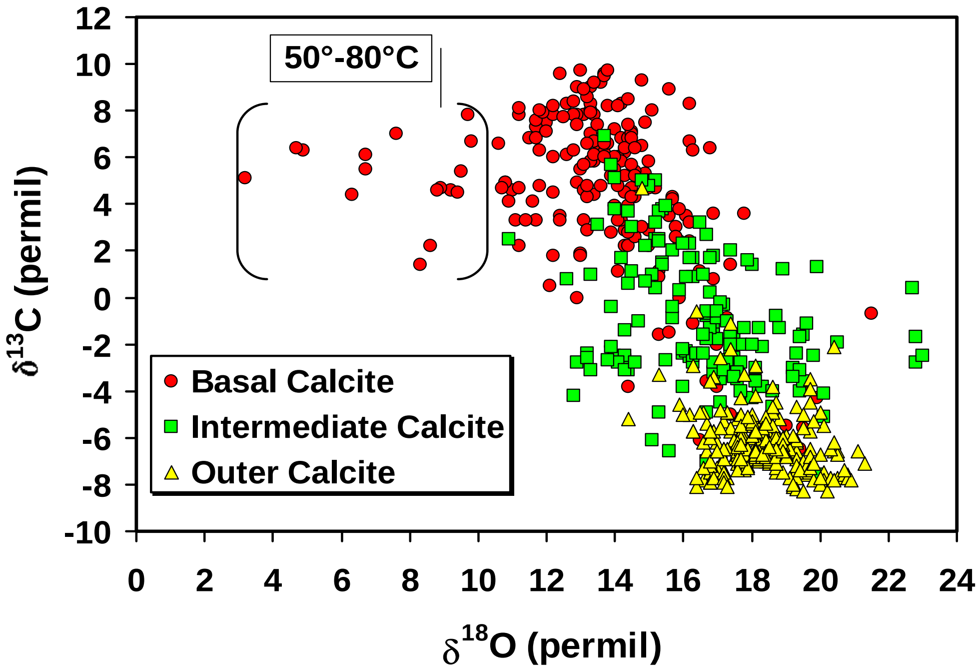
Oxygen and Carbon Isotopes in Calcite

The fractionation (distribution) of ^{18}O between a mineral and the depositing water is a function of temperature (i.e., the $\delta^{18}O$ difference decreases as temperature increases).

- If the water $\delta^{18}O$ is constant, the calcite $\delta^{18}O$ increases as temperature decreases
- Using the measured calcite $\delta^{18}O$ and estimating the water $\delta^{18}O$ allows the depositional temperature to be estimated

These estimates indicate:

- Elevated temperatures during deposition of the oldest parts of the paragenetic sequence
- A long-term increase of calcite $\delta^{18}O$ that is consistent with gradual cooling of the UZ rock mass



Temperature Estimates From Calcite $\delta^{18}O$

The $\delta^{18}O$ values of paleowaters that deposited calcite are broadly constrained.

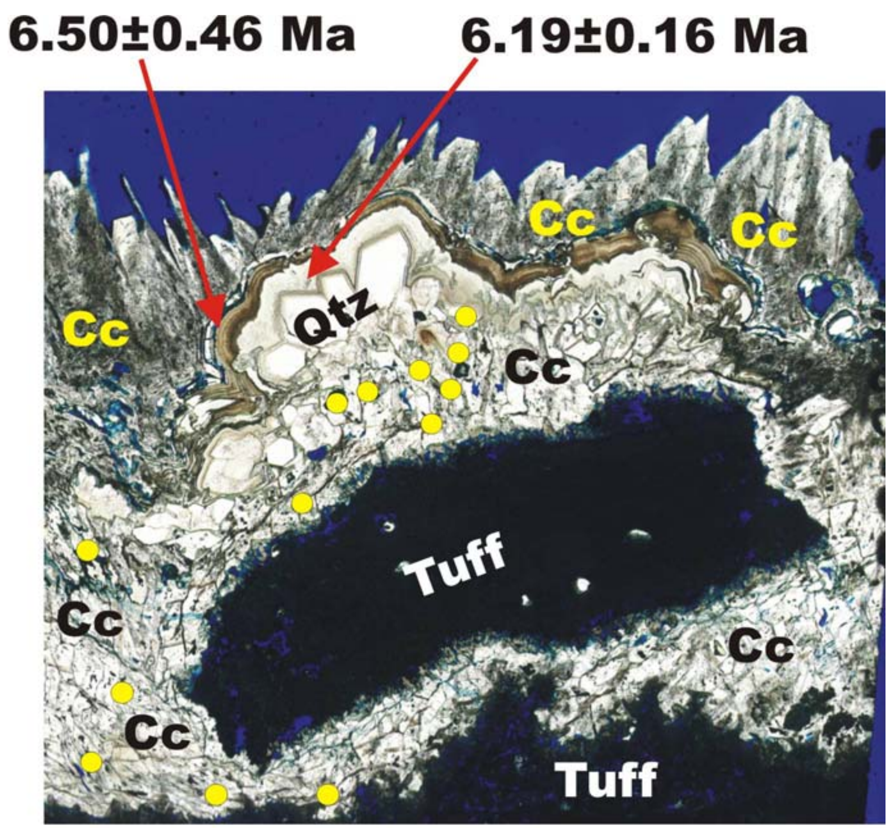
Assuming paleowater $\delta^{18}O$ values were comparable to recent recharge, the calculated formation temperatures of calcite range from 56 to ~100ºC:

- **These estimates are in good agreement with the fluid inclusion homogenization temperatures from the same sample sites.**
- **This agreement suggests that:**
  - **the $\delta^{18}O$ value of UZ fracture water has been relatively uniform**
  - **calcite $\delta^{18}O$ values provide a proxy for depositional temperature**

LOCATION	$\delta^{18}O_{CC}$ (permil)	$\delta^{18}O_{CC}$ TEMP.	FI TEMP.
ESF 1+62	3.2 – 6.3	77 - 100	64, 85
ESF 12+16	6.7	74	54 - 66
ESF 14+06	7.6, 9.8	53, 68	55, 64
ESF 72+94.5	8.6	60	57, 62
ESF 75+74.7	8.9	58	<60

Fluid Inclusion Geochronology

- Although calcite cannot be dated directly, age constraints are provided by U-Pb dating of intercalated opal or chalcedony.
- In this sample, calcite hosting elevated-temperature fluid inclusions is older than 6 Ma.
- U-Pb ages constraining elevated temperature calcite range from 1.9 to ~9 Ma but most are >5 Ma.
  - they are generally minimum ages (i.e. the dated mineral overlies the calcite)



ESF AI #5, 0+28.5E

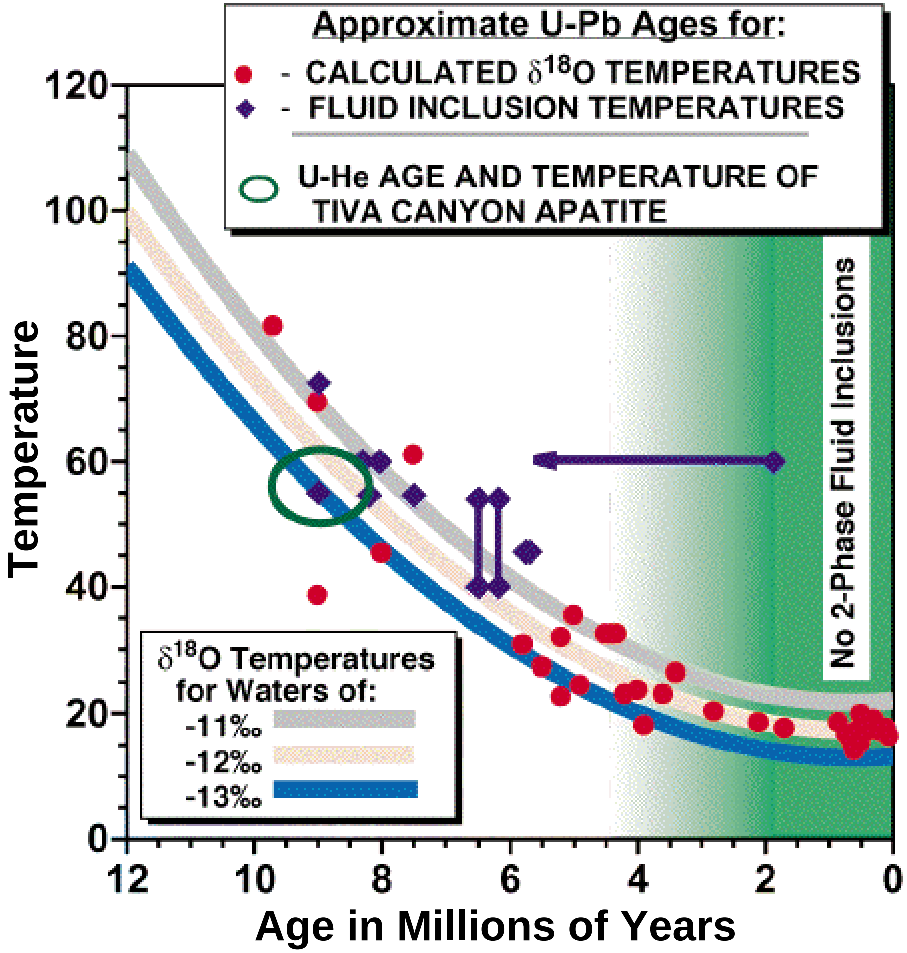
THE MINIMUM AGE OF ELEVATED TEMPERATURE CALCITE FORMATION IS >1.9 (1.1?)* Ma.

* see supplemental slide 4 for more details

Thermal History of the Yucca Mountain UZ

In addition to FIA temperatures constrained by U-Pb dates, a larger number of calcite $\delta^{18}O$ values and temperature estimates are also age-constrained*.

- **For samples hosted by the Topopah Spring Tuff, $\delta^{18}O$ temperatures were calculated assuming a water $\delta^{18}O$ of -11 to -13‰, similar to recent meteoric water percolating into the UZ.**
- **Also, apatite from the North Ramp has a U-He age of ~8.7 Ma with a closure temperature of ~56°C, in good agreement with the other age-constrained estimates of temperature.**

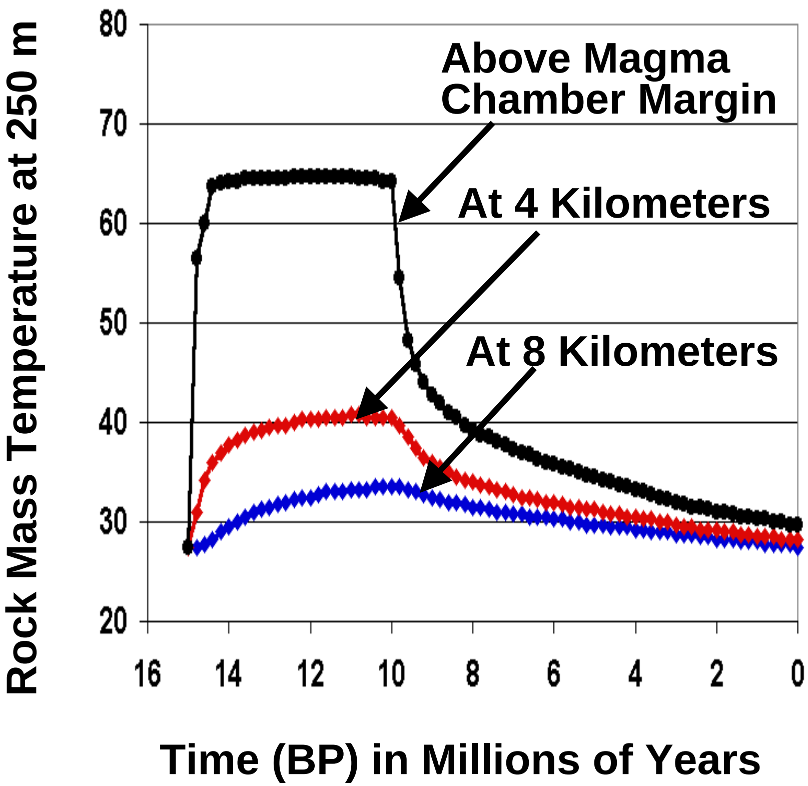


THESE AGE-CONSTRAINED TEMPERATURES INDICATE LONG-TERM, GRADUAL COOLING OF THE UZ ROCK MASS.

***(see supplemental slide 5 for more details)**

Preliminary Thermal History Models

Thermal modeling of the cooling history of the Timber Mountain Caldera predicts prolonged, gradual cooling of the UZ.



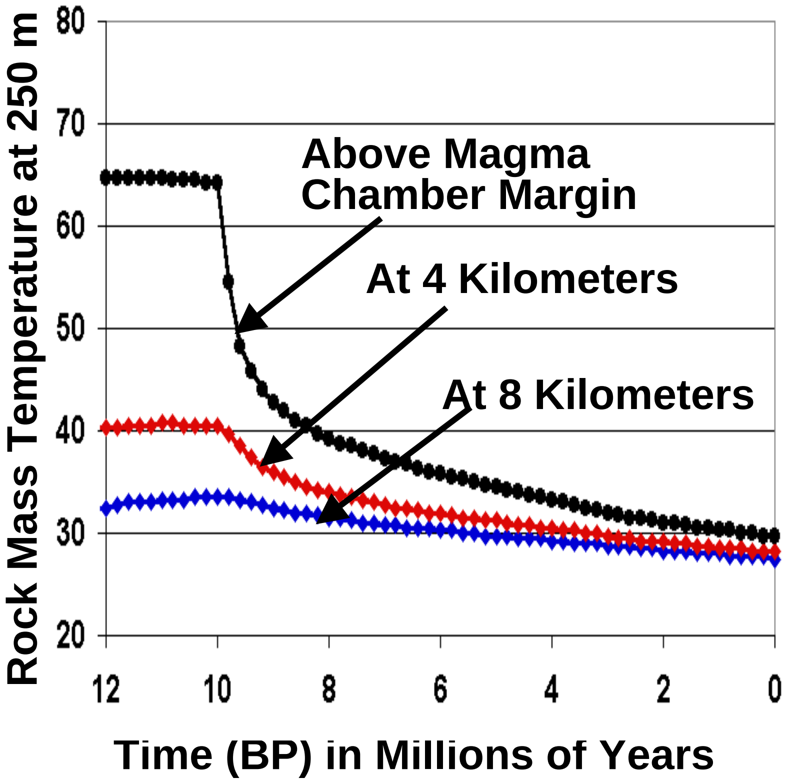
The cooling curves shown model temperature at 250 m depth immediately above the edge of the magma chamber and at 4 and 8 km distance and are based on the following assumptions:

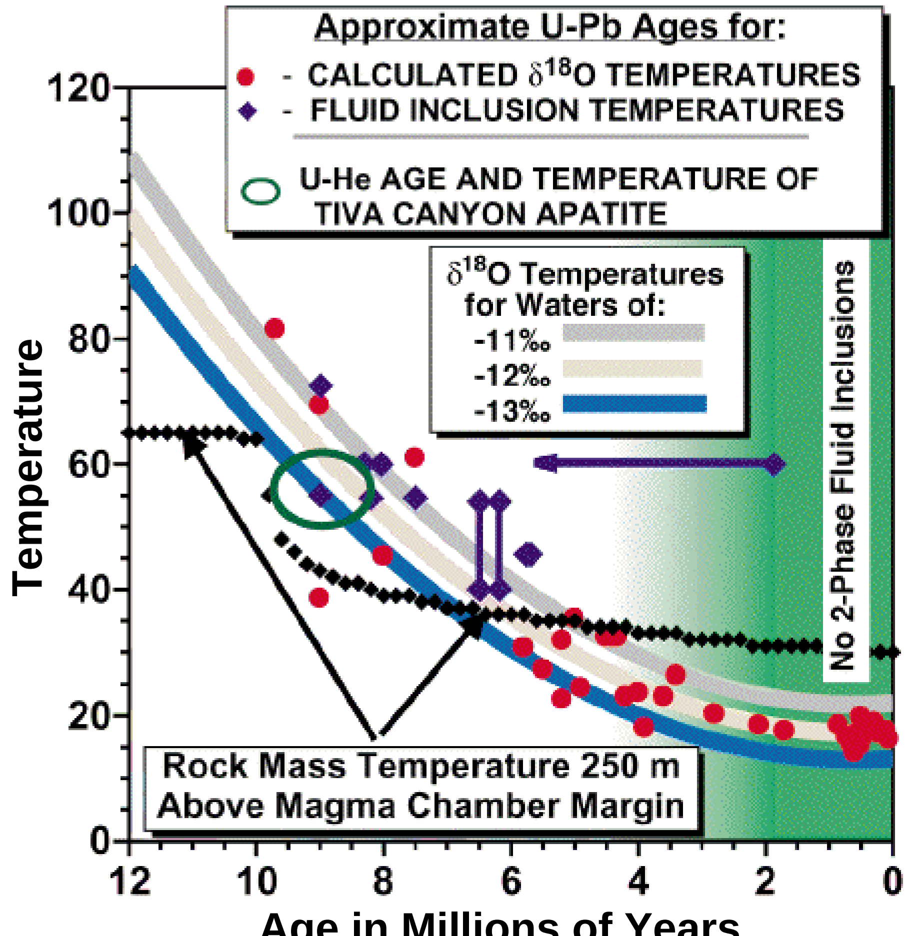
- **a disc-shaped, 30-km diameter, 5000 km^3 magma chamber centered 5 km below Timber Mountain**
- **A 900°C magma temperature from 15 to 11 Ma**
- **A 500-m-thick UZ at Yucca Mountain**
- **Intracaldera hydrothermal convection until 10 Ma**

(see supplemental slides 6 & 7 for more details)

Preliminary Thermal Models Compared to UZ Temperature Estimates

Preliminary thermal history modeling supports the prolonged cooling history of the UZ implied by the in situ temperature estimates.





USGS Conclusions

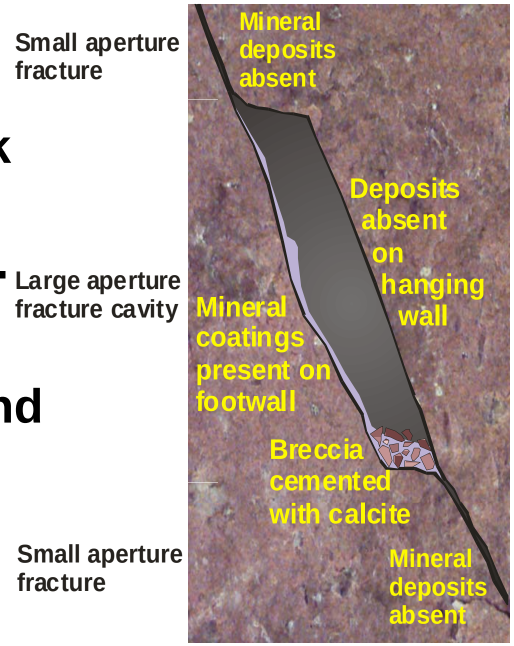
- Fluid inclusions and calcite $\delta^{18}O$ indicate elevated temperatures during the early and intermediate stages of calcite formation.
- These temperatures are consistent with the likely thermal history of the unsaturated zone tuffs as indicated by age constrained temperature data from fluid inclusions and $\delta^{18}O$ values and by thermal modeling.
- The fluid inclusion assemblages, which include inclusions with large and variable V:L ratios, are consistent with vadose zone formation.
  - Therefore, there is no evidence supporting flooding of the UZ.
  - Furthermore, the extremely sparse and heterogeneous distribution of the deposits is specifically inconsistent with flooding.
- The potential repository block has been at or near present-day temperatures for at least the past 2 m.y., likely the past 3 to 4 m.y.

Supplement 1

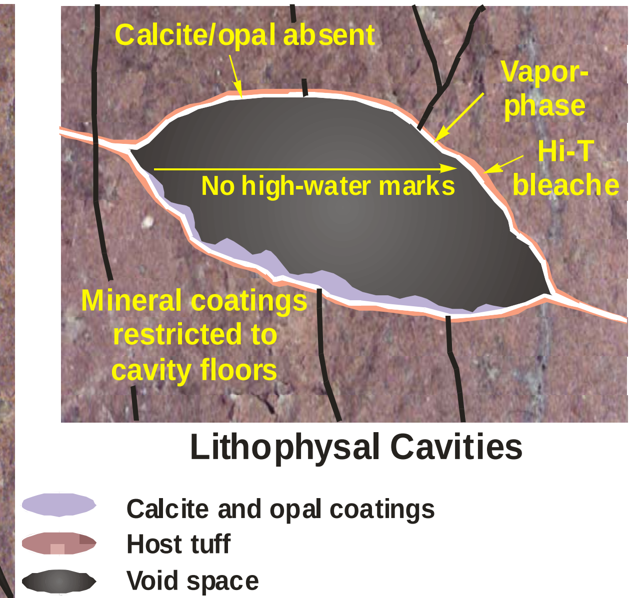
Locations of UZ mineral coatings

The physical locations of mineral coatings provide important information on hydrologic processes.

- **Coatings in the UZ are present in <u>unfilled</u> rock voids on fracture <u>foot-walls</u> and cavity <u>floors</u>.**
  - > **<u>no high water marks</u> on opposite walls and ceilings**
- **In contrast, SZ coatings show no gravitational preference and coat all cavity surfaces**



Fracture Cavities



Lithophysal Cavities

The <u>gravitational influence on UZ coatings</u> represents strong evidence that cavities have always remained hydrologically unsaturated, and <u>fracture water flows downward</u> on fracture footwalls and cavity floors.

Supplement 2

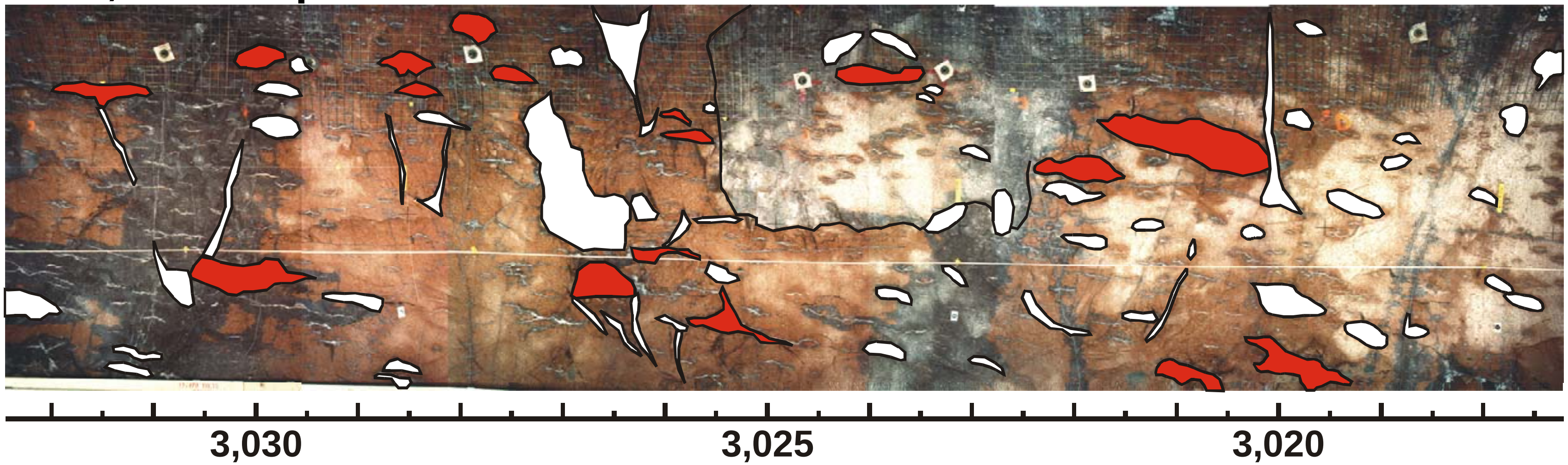
Distribution of UZ Mineral Coatings

- Only a small proportion of all fractures and rock voids contain calcite and opal coatings.
- In 30-m long surveys of the ESF tunnel walls, only a small percentage (0 to 40%, but typically <10%) of lithophysal cavities contain coatings.

Rock cavity — potential deposition site.

Rock cavity containing calcite-opal coating.

ESF; Left rib photomosaic



- Provides strong evidence that the <u>UZ was never inundated with water</u>, otherwise all rock voids would contain calcite and opal

Supplement 3

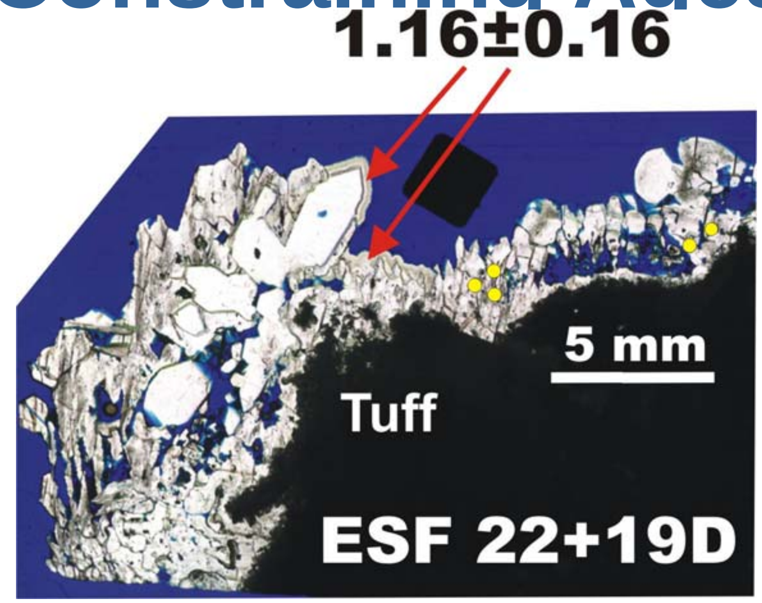
Geochemical Evidence for Descending Fracture Flow Deposition

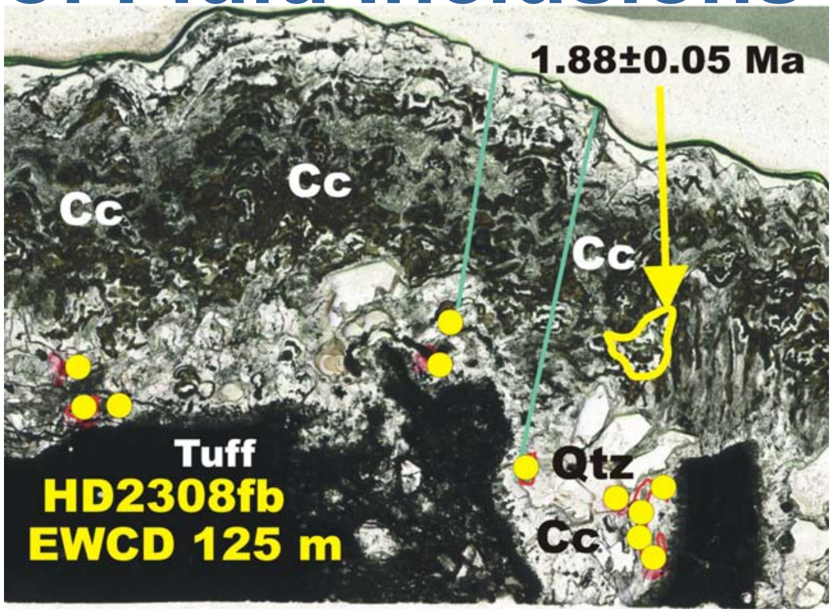
- $\delta^{13}C$ values of youngest UZ calcite overlap those of soil calcretes.
- $\delta^{18}O$ values of youngest UZ calcite are consistent with a meteoric water source warmed along the geothermal gradient during downward percolation.
- $\delta^{87}Sr$ values of youngest calcite overlap those of soil calcretes.
- REE concentrations of UZ calcite have pronounced negative Ce anomalies.
  - Not observed in SZ calcite (Vaniman and Chipera, 1996)
  - Small or nonexistent in regional ground water (Johannesson et al., 1997)
- $^{234}U/^{238}U$ ratios in shallow calcite are identical to soil carbonate and runoff
  - Observed ratios are much smaller than those in Tertiary volcanic or Paleozoic ground waters beneath Yucca Mountain.

Conclusion: Chemical and isotopic evidence is inconsistent with calcite/silica deposition from upwelling ground water.

Supplement 4

Constraining Ages of Fluid Inclusions





- **In these coatings ages of silica bounding TPFI-bearing calcite are as young as 1.2 – 1.9 Ma**
- **Dated silica is separated from TPFI-bearing calcite by Qtz**
- **How long was the age gap between the calcite and dated silica?**

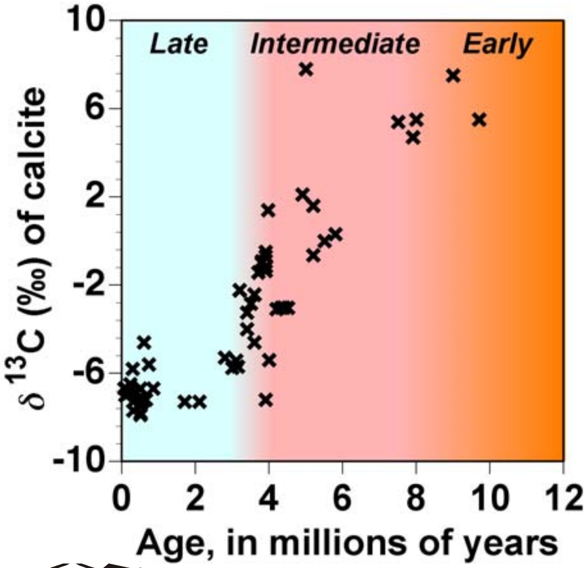
Supplement 5

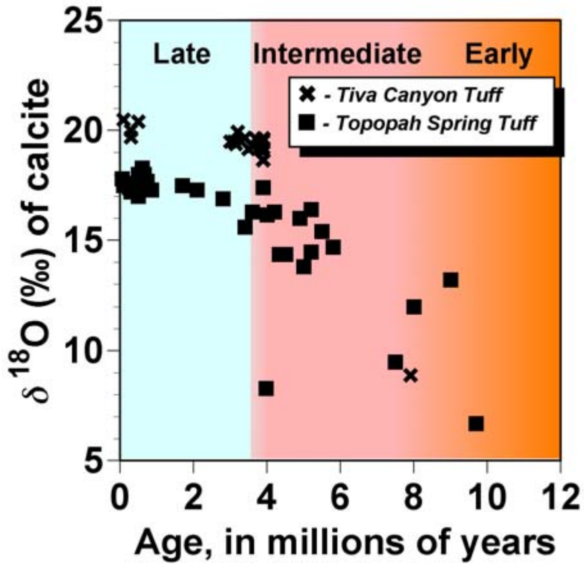
Age-Constrained δ13C and δ18O of Calcite

- Because the UZ appears to have been thermally stable during the Late Stage, the timing of the Intermediate to Late Stage transition is important.

- In addition to the temperature stabilization, calcite δ13C values suggest a concurrent shift in climate.

- U-Pb dating of UZ calcite indicates that the transition from Intermediate to Late stages probably began 3 to 4 Ma.





Supplement 6

Preliminary Thermal Modeling

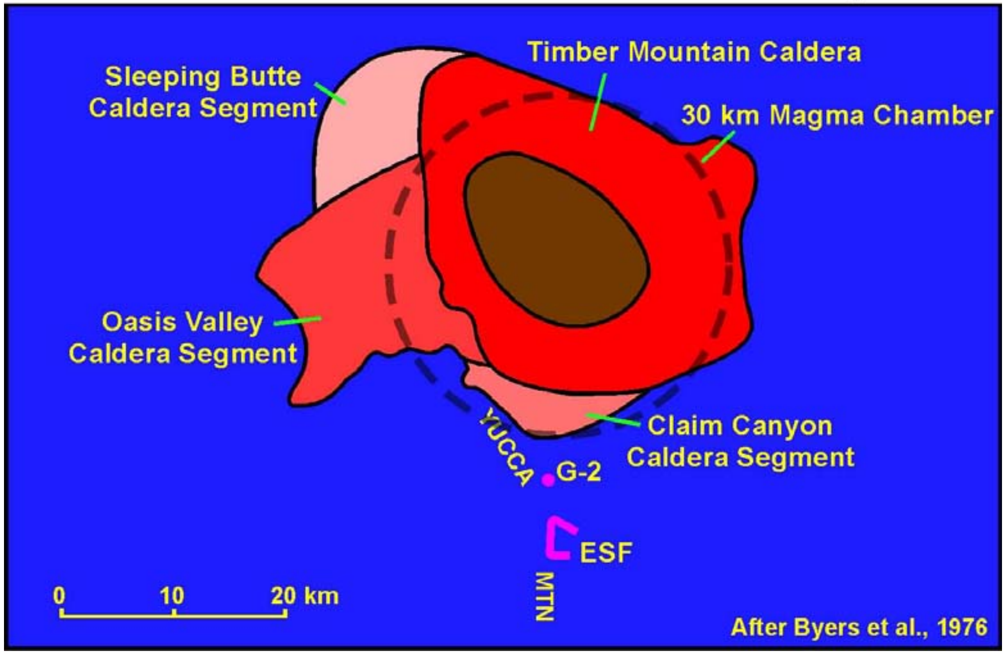
- **Primary considerations**
  - **Rock type distribution**
  - **Thermal conductivities & densities**
  - **Initial thermal gradient**
  - **Magma chamber volume & depth**

Future Considerations:

- **Distribution and timing of convection**
- **Erosional history**
- **Caldera subsidence with time**

Modeling based on Wohletz & Heiken (1992)

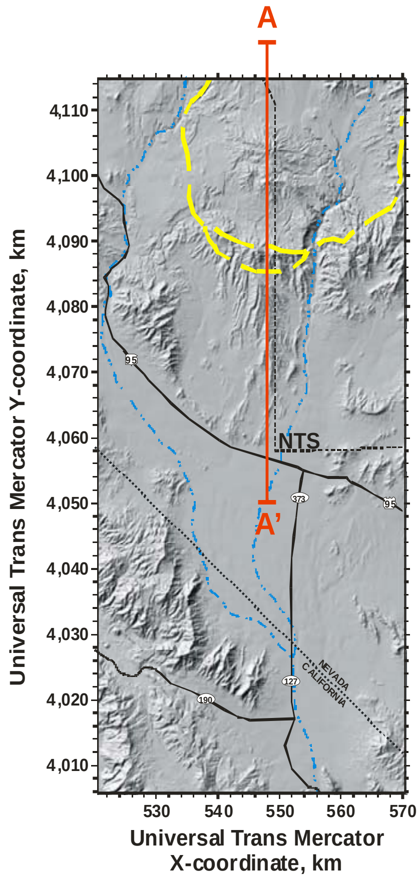
Timber Mountain Caldera Complex



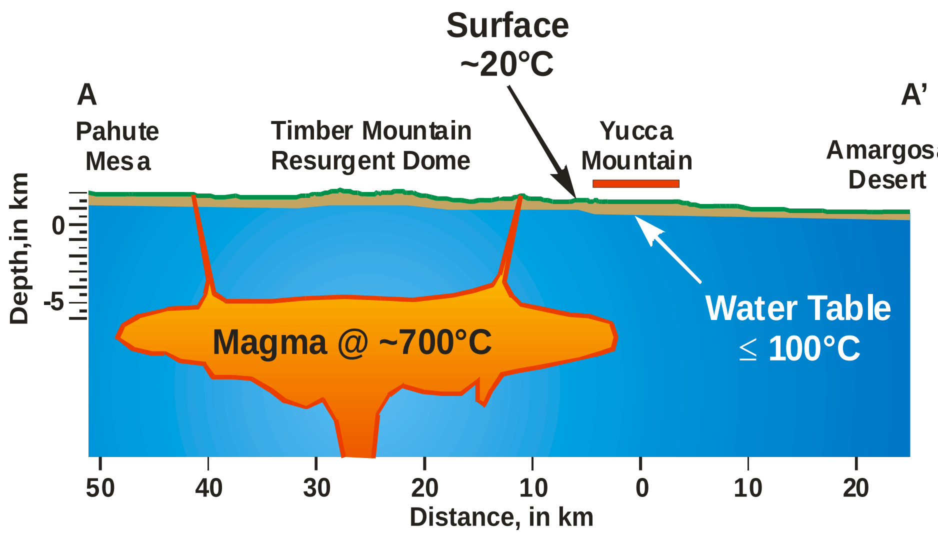
Supplement 7

Potential Heat Sources to the UZ Tuffs

- **Residual heat from tuff eruption:**
  - » Dissipates rapidly - 100's to 1,000's of years
  - » Responsible for fumarolic activity



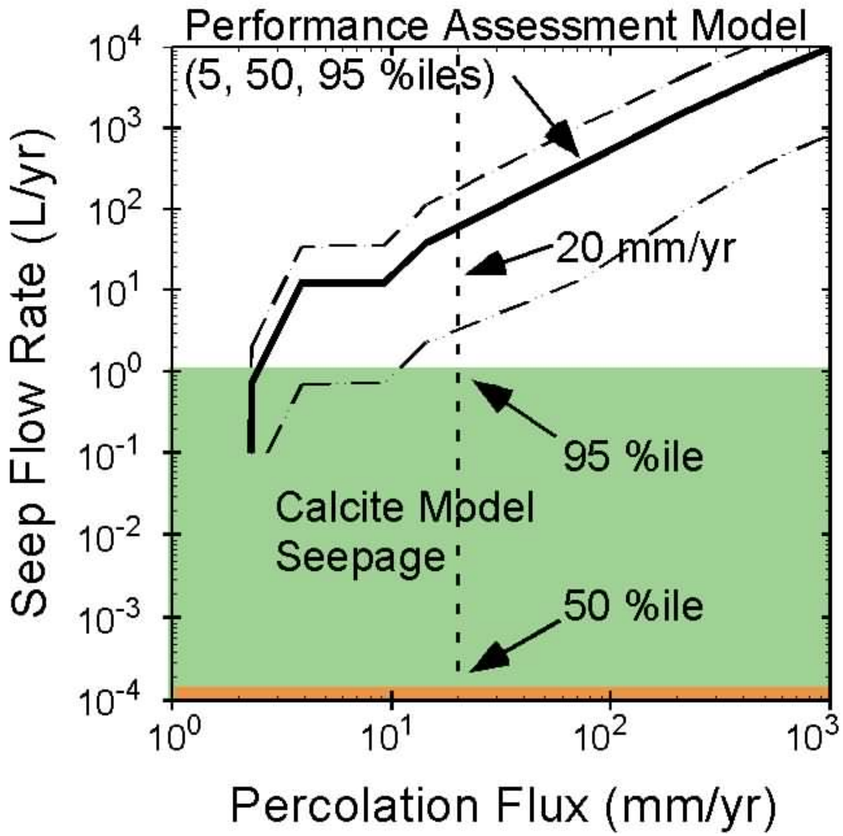
- **Slow cooling of buried magma chambers:**
  - » Repeated emplacement of large magma bodies
  - » Combined conduction & advection
  - » Requires millions of years to return to regional heat-flow levels



Supplement 8

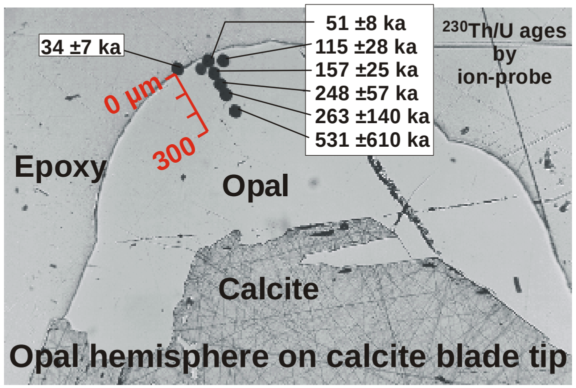
Calcite Abundance & Past Seepage

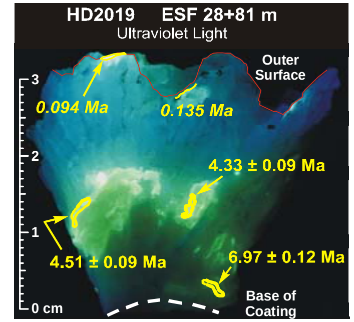
- Line surveys conducted in the ESF measure both frequency of calcite coatings and thicknesses.
- Estimates of water/calcite ratios based on geochemical models of pore water evolution and analog data allow past seepage volumes to be calculated.
- The amount of past water seepage calculated from calcite is much lower than estimates of seepage based on large-scale models of UZ flow.

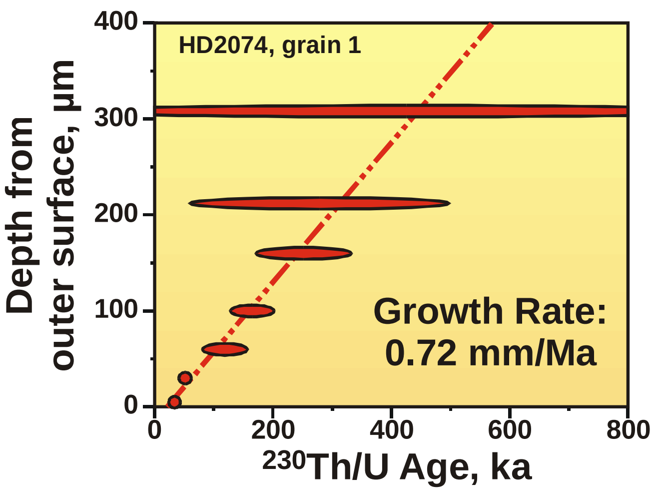


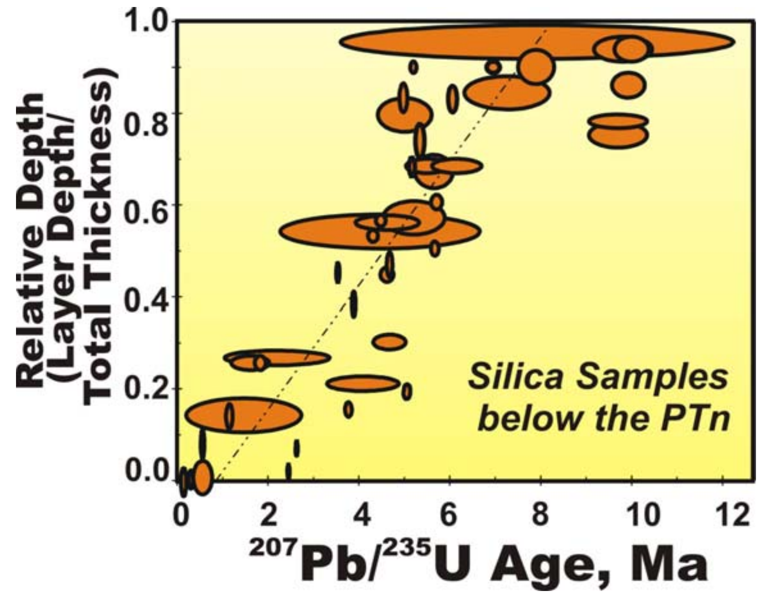
Secondary Mineral Deposit Growth Rates

- **Average depositional rates are very slow (mm/m.y) and, in deep UZ coatings, were remarkably consistent during the Tertiary to Pleistocene time span.**









- **Slow, uniform long-term average growth rates are consistent with continuous supply of downward percolating fracture water and also imply that fracture flow has been buffered from large variations in infiltration (i.e., climate change).**