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Mobilization of Uranium From the Nopal I Analog and Implications for Nuclear Waste Repository Performance

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Introduction

- Nopal I uranium ore body is a useful analog for understanding performance of the potential high-level waste repository at Yucca Mountain
- Among the evidence for recent radionuclide mobility are elevated uranium concentrations in fracture filling carbonates (crystalline calcite and caliche) and silicates
- U-Th isotopes used to constrain timing and stable isotopes used to constrain conditions of this mobilization

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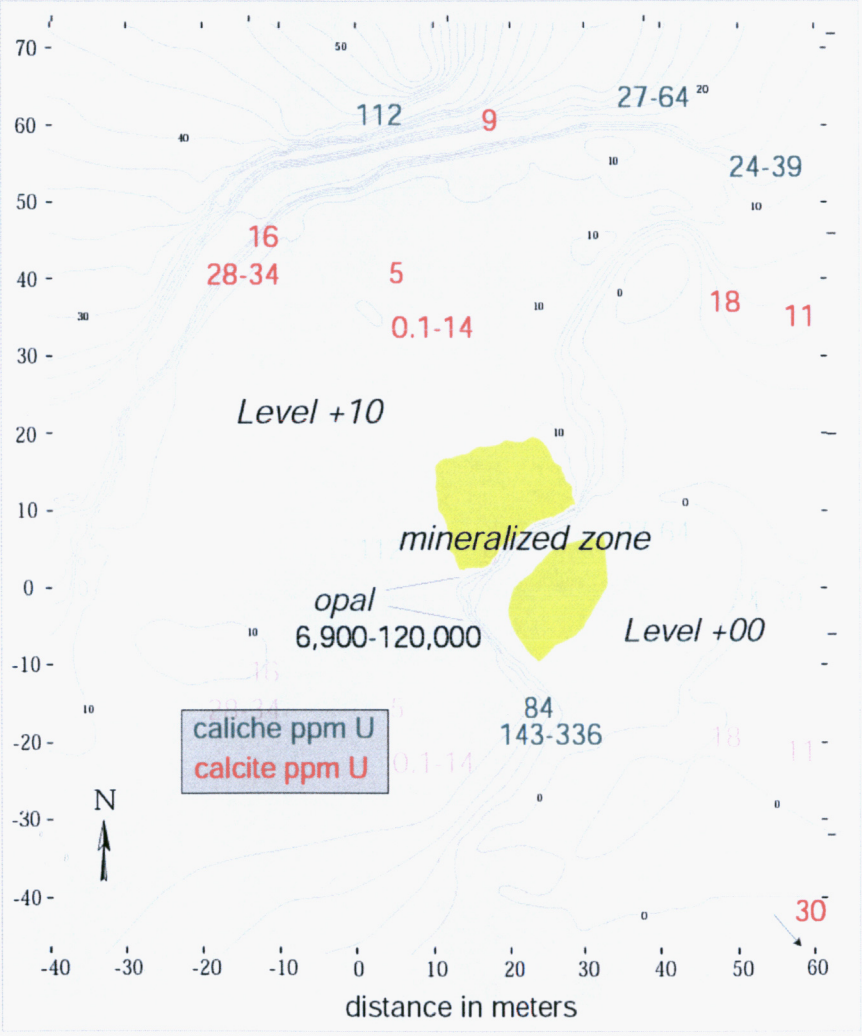
Uranium Concentrations



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Crystalline Calcite

- Fill tuff fractures and coat free surfaces typically with euhedral crystal forms up to 1 cm [0.4 in]
- Range from clear and colorless to opaque and dark reddish brown. Some have appreciable non-carbonate residual contents

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Caliche

- Along tuff fracture faces from 0 to 2 m [0 to 6 ft] below the pre-mining surface
- Layers typically range from 1 to 10 mm [0.04 to 0.4 in] thick in masses up to 15 cm [6 in] thick
- Nearly pure white to grayish beige
- XRD: calcite with and without quartz and sanidine

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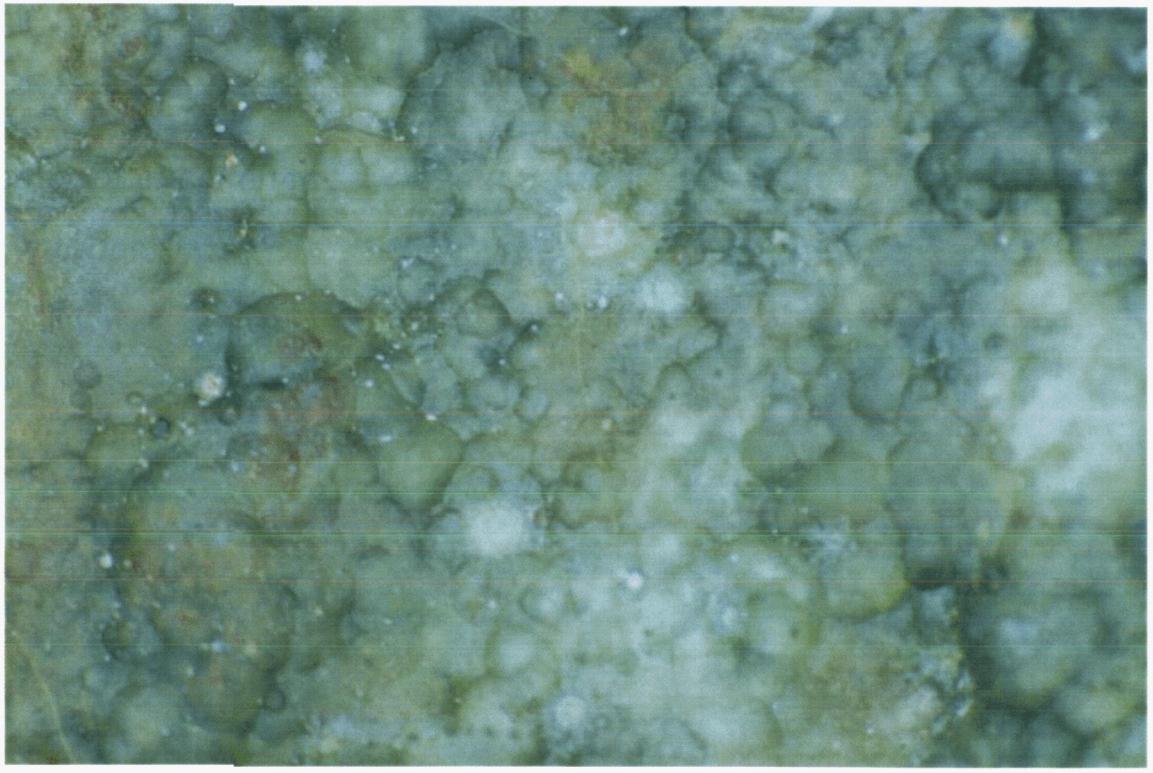
Opal



Field of view 13 mm [0.5 in] wide.

- Coats tuff or tuff breccia.
- May occur with uranophane that precedes and postdates opal deposition.
- In the presence of uranophane, the color may grade to deep yellow, suggestive of incorporation of microcrystals of a uranyl phase.

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Uranium Concentrations

Crystalline Calcite: 0.1 to 34 ppm

- Average 10 ppm.

Caliche: 24 to 335 ppm

- Carbonate contents are typically 60 to 90 %.
- 50 to 90 % of U is in leachate.
- Calculated U concentrations in leachable solid is 9-250 ppm; in residues, 50-400 ppm. Both components are U-rich.
- Caliches appear to be recorders of U migration from the ore body along near-surface fractures.

Opal:

- 6900 to 8800 ppm in clear greenish samples.
- Opal with strong yellow color has 12 percent U
  - included U minerals?

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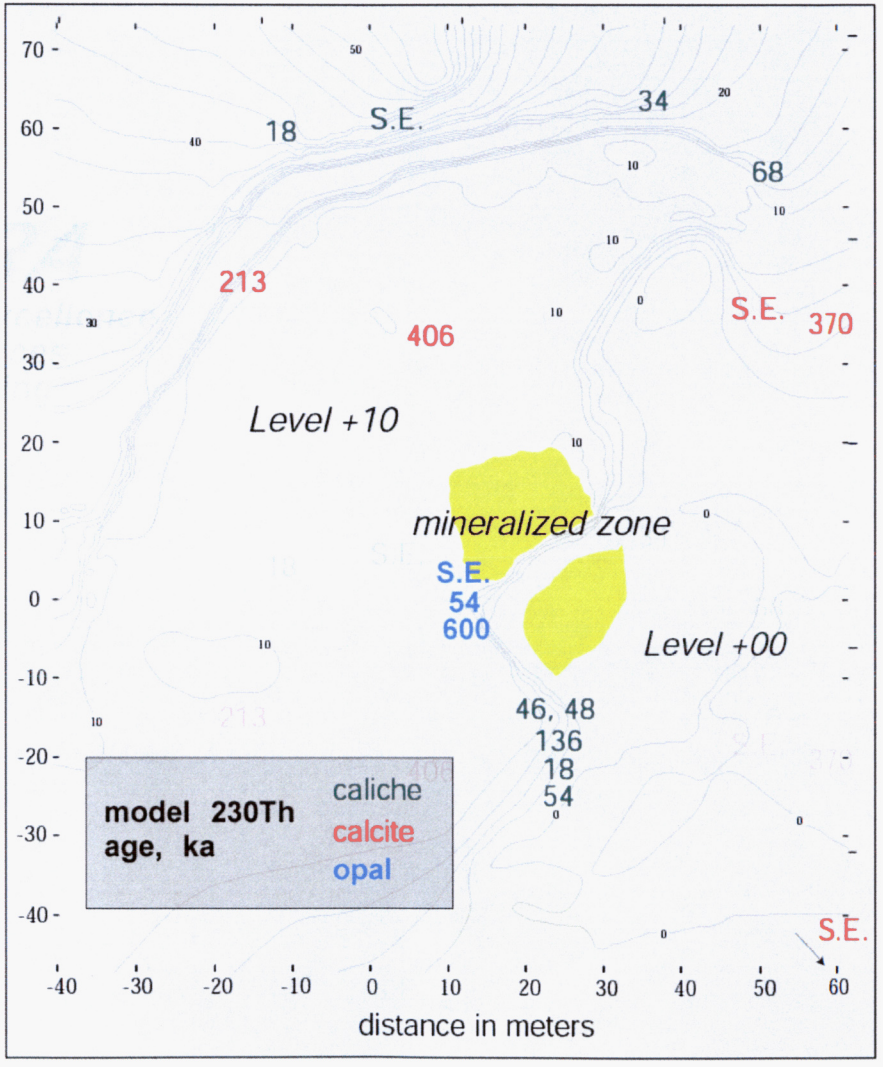
^{230}Th-^{234}U-^{238}U Dates



- Total rock dissolution
- Caliche: 18 to 136 ka, except one near secular equilibrium (Si-rich)
- Calcite: 210 to 410 ka or near secular equilibrium
- Opal: >600 ka, except one existing date at 54 ka

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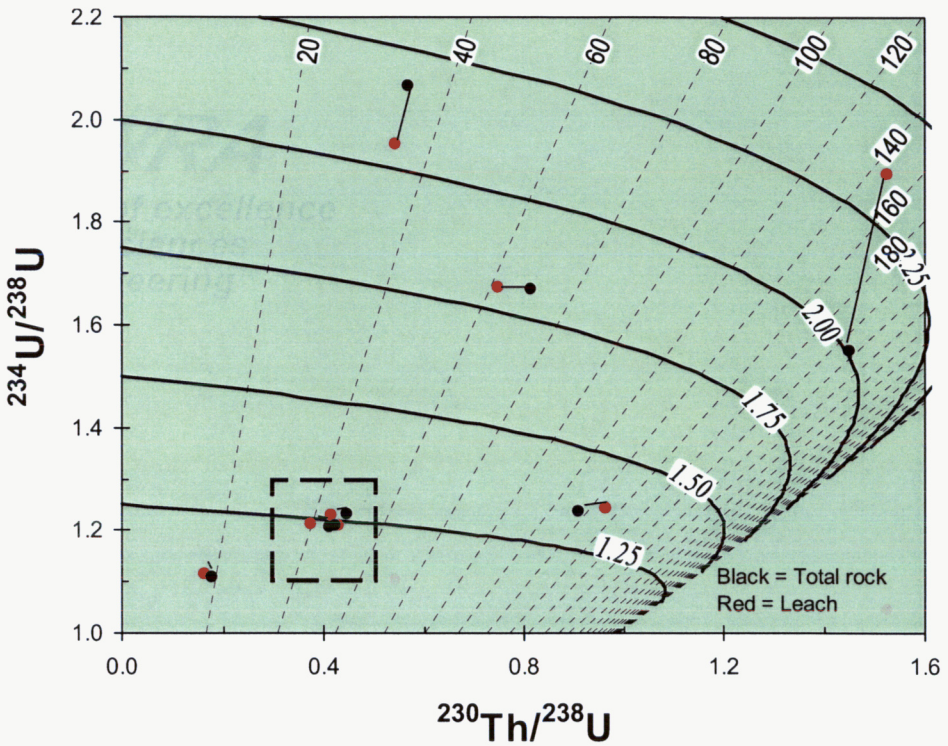
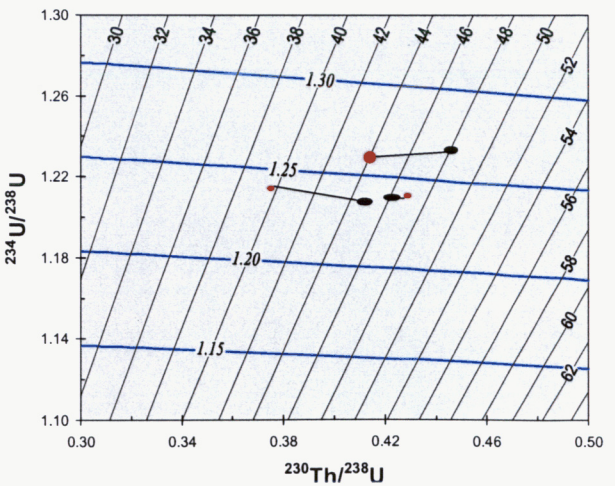
U-Th Carbonate Leachate Data



- Uranium in leachate = 49 to 92%.
- With one exception, leachate date is within 11% of total dissolution date. The exception is a calcite with 10-15% breccia inclusions.
- Leachate isotopic ratios are not consistently higher or lower than total dissolution ratios. Leachate total dissolution pairs do not provide reliable ages.



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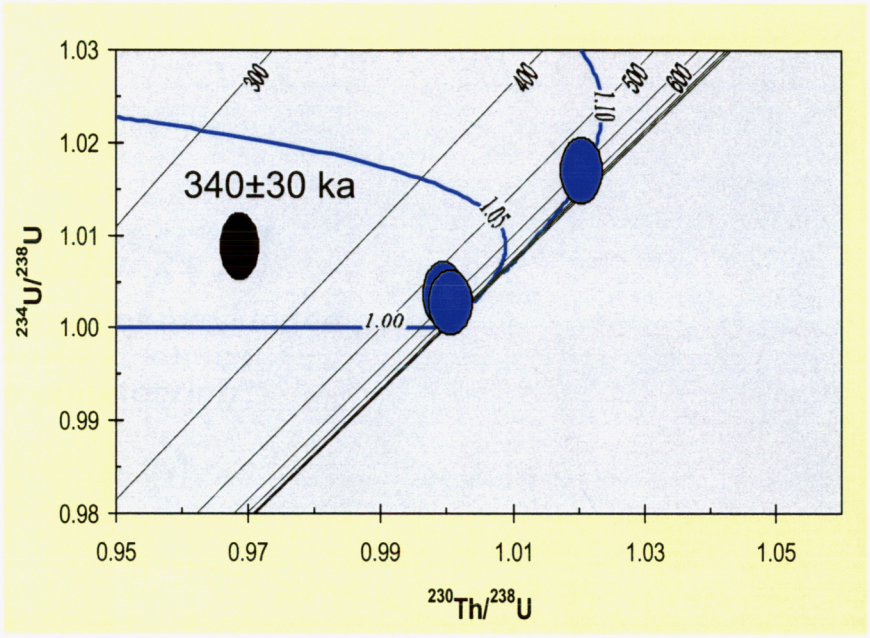
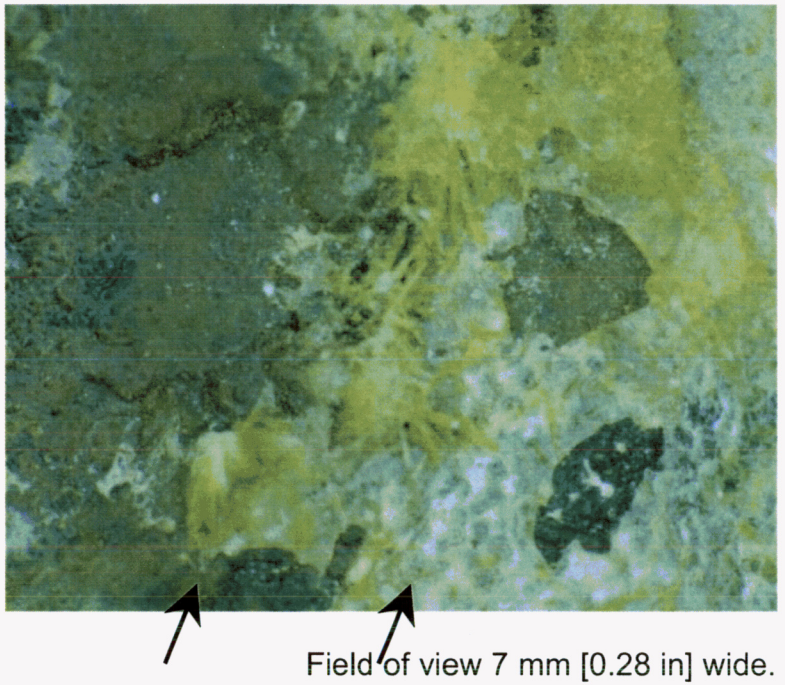
Opal and Uranophane



Blue = opal; black = uranophane



Field of view 7 mm [0.28 in] wide.

-Pair of opals from single sample both at secular equilibrium; two layers seen in photo.

-Uranophane from same sample; ^{230}Th age is consistent with textural evidence. Youngest U mineral age yet from Nopal I (340±30 ka).

-Younger opal (600±300 ka) from separate sample.

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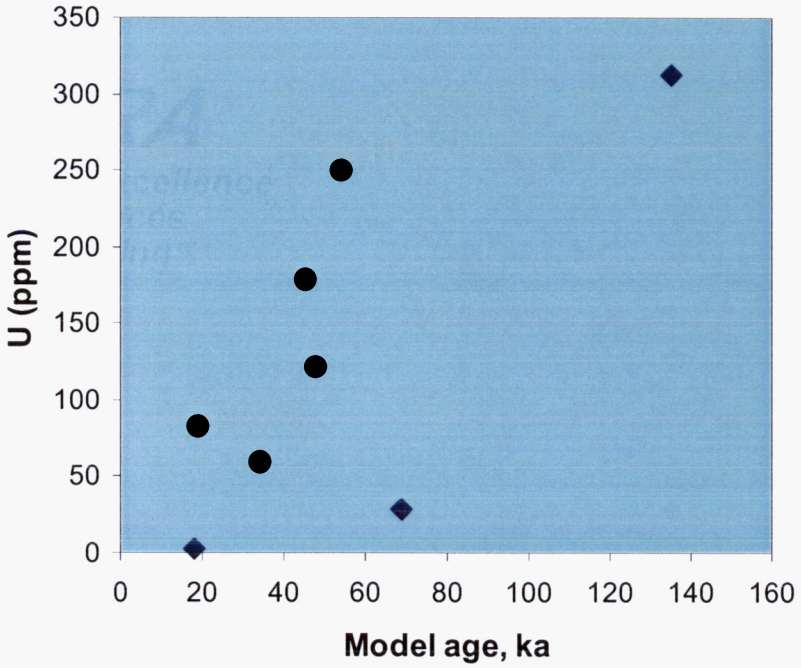
Timing of Uranium Mobilization



Caliche data

- Suggest of a period of enhanced mobility around 40 to 60 ka
- Samples from this apparent episode are from same area just south of ore body, but are from distinct layers and fractures
- Supported by existing 54 ka age for high-U opal

Other opals and uranophane record high U mobility at > 300 ka, consistent with existing Fe-rich fracture fill data

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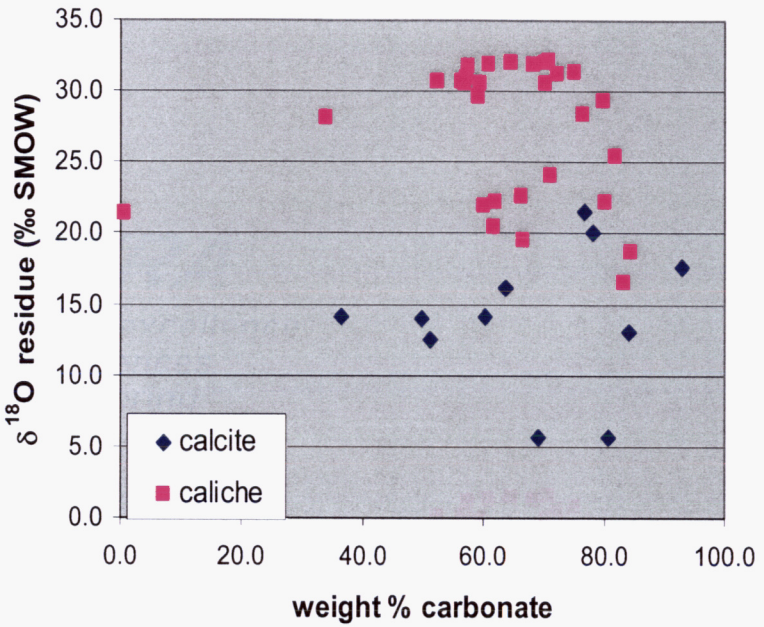
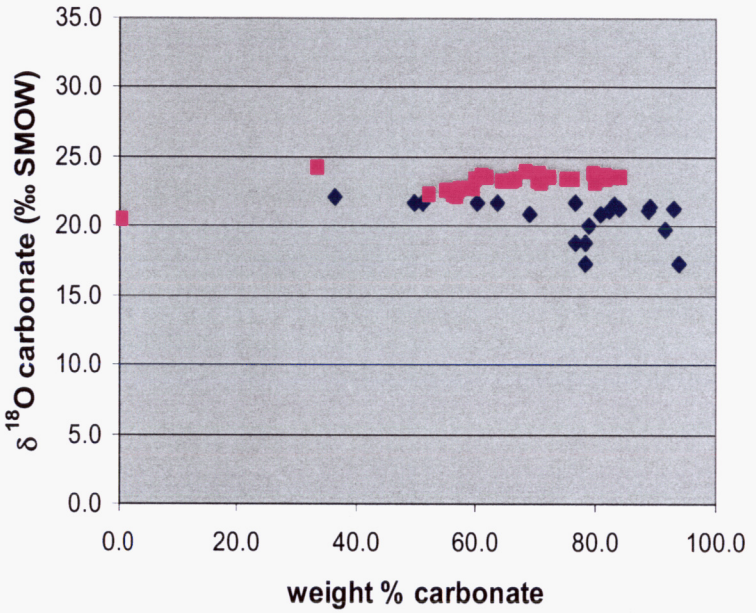
Oxygen Isotope Data





- $\delta^{18}O$ is much more uniform in the carbonate fractions, likely due to the influence on the residue values of non-authigenic silicates and oxides.
- Higher $\delta^{18}O$ in caliche residues suggest a higher proportion of authigenic silica in the residues than for calcites. Low-temperature fractionation between silica and carbonate is about 7-8 permil.
- Caliche carbonate values are consistent with low-temperature {15-20 °C [60-70 °F]} fractionation relative to water with the characteristics of regional meteoric water ($\delta^{18}O$ ~ -7 ‰ SMOW).
- Lower $\delta^{18}O$ in calcite carbonate could be due to higher formation temperatures (by 10 or more degrees).

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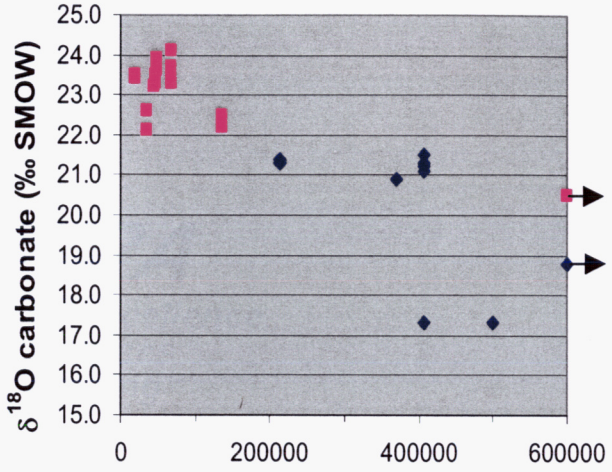
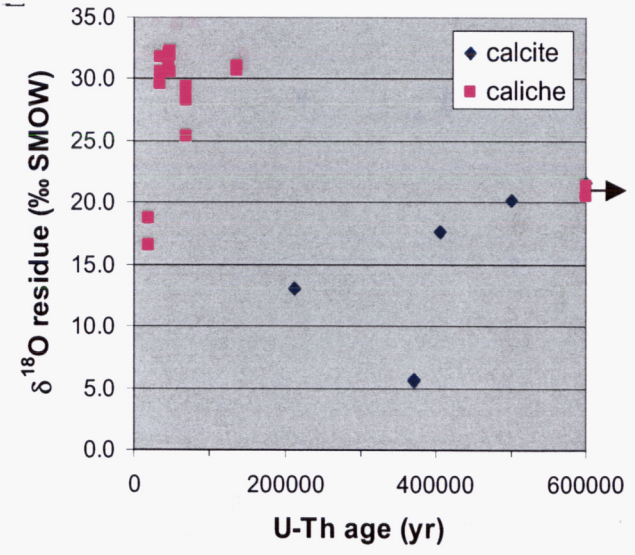
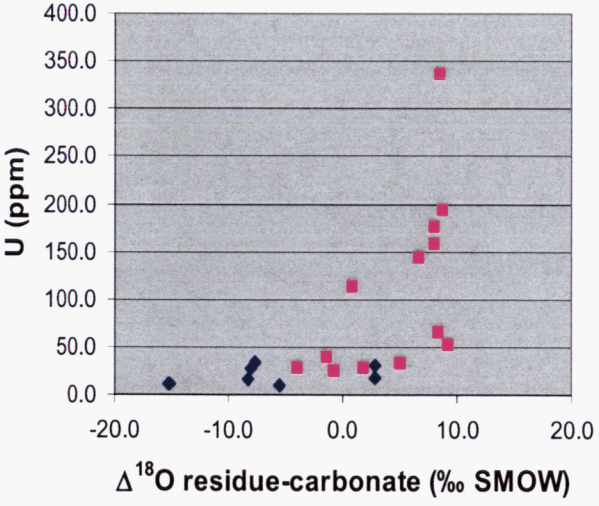
Oxygen Isotope Data





- Young caliches tend toward higher $\delta^{18}O$ (left) and higher $\Delta^{18}O$ residue-carbonate. (Youngest caliche had large amount of visible detritus.) Consistent with lower temperatures, but source variation and water-rock interaction effects are also possible.
- High-uranium caliches have high $\Delta^{18}O$ residue-carbonate (below), consistent with low-temperature formation of coexisting authigenic silica and carbonate.



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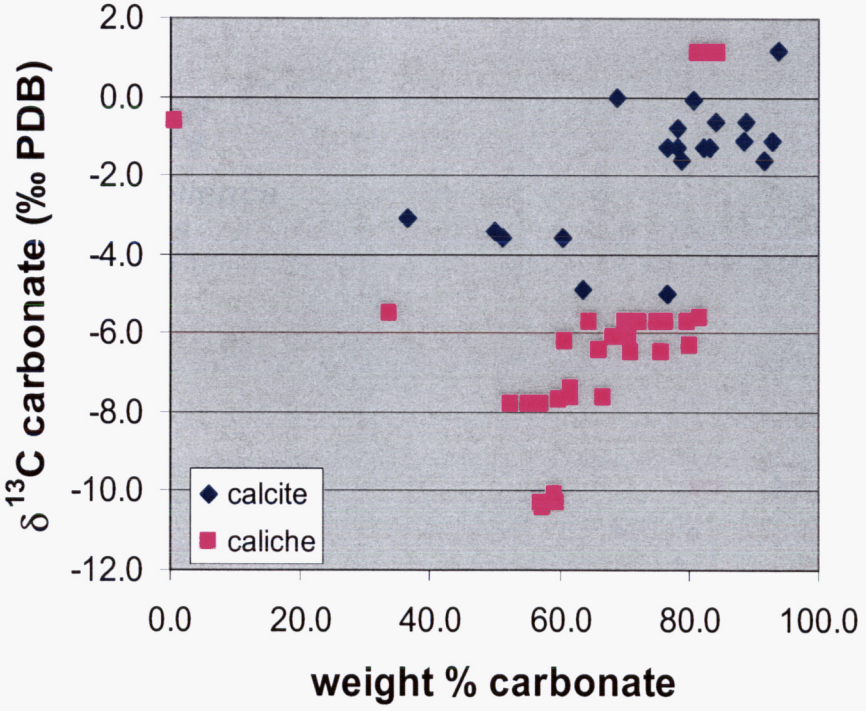
Carbon Isotope Data



- $\delta^{13}C$ is also distinct for caliches and calcites, overall.
- If aqueous carbon source $\delta^{13}C$ were constant, the lower caliche values would be due to lower formation temperatures than for calcites.
- However, carbon isotope fractionation is not as sensitive to temperature as oxygen, so a source effect is also likely.
  - More influence by vegetation on the more recent caliches?
  - Underlying marine limestone source for calcites?

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Summary

- Mixtures of authigenic carbonate and silicate in a high-uranium setting present challenges for precise ^{230}Th-^{234}U-^{238}U dating.
  - Leachate dates agree with total-rock dates within perhaps 10-20 %
- Evidence for some uranium mobility over the past few hundred thousand years.
  - Recorded in both authigenic carbonate and silica
  - Apparent period of enhanced mobility around 60 to 40 ka
  - Nopal I is not a closed system
- High-uranium opals and uranophane record older episodes of >300 ka.
  - Consistent with existing data on iron-rich fracture fills
  - Youngest Nopal U mineral age yet: uranophane at 340 ±30 ka.
- Stable isotope data consistent with aqueous deposition of younger U-rich caliche in a lower-temperature {15-20 °C [60-70 °F]}, near-surface environment.

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Acknowledgment

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