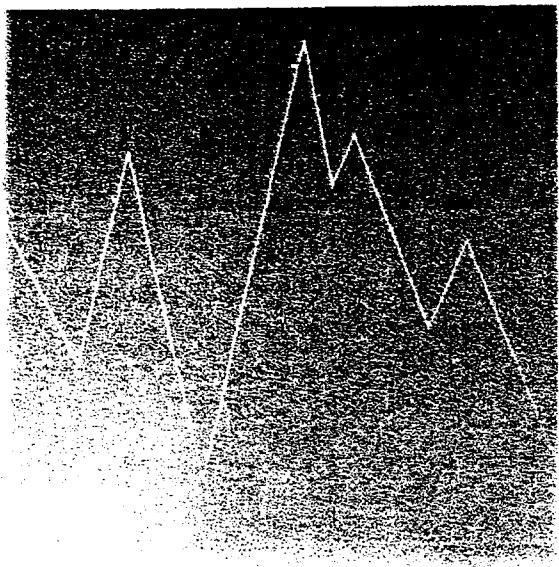


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Hydrologic Inferences from Strontium Isotopes in Port Water From the Unsaturated Zone at Yucca Mountain, Nevada

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Calcite is ubiquitous at Yucca Mountain, occurring in the soils and as fracture and cavity coatings within the volcanic tuff section. Strontium is a trace element in calcite, generally at the tens to hundreds of ppm level. Because calcite contains very little rubidium and the half-life of the ^{87}Rb parent is billions of years, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the calcite record the ratio in the water from which the calcite precipitated. Dissolution and reprecipitation does not alter these compositions so that, in the absence of other sources of strontium, one would expect the strontium ratios along a flow path to preserve variations inherited from strontium in the soil zone.

Strontium isotope compositions of calcites from various settings in the Yucca Mountain region have contributed to our understanding of the unsaturated zone (UZ), especially in distinguishing unsaturated zone calcite from saturated zone calcite. Different populations of calcite have been compared, either to group them together or distinguish them from each other in terms of their strontium isotope compositions. Ground water and perched water have also been analyzed; this paper presents strontium isotope data obtained on pore water.

Although pore water can be squeezed from the nonwelded tuffs at Yucca Mountain, the volumes recovered from reasonable lengths of drill core are small. We have used dry-drilled core samples that have dried during storage, as repositories of pore water salts that can be carefully leached with deionized water for analysis of strontium isotope ratios. Crushed core is sieved to obtain a coarse sand (30-60 mesh) fraction

that is then leached for less than an hour with deionized water to redissolve the pore-water salts. This water sample is then centrifuged and filtered; strontium is separated by standard techniques for analysis by thermal ionization mass spectrometry (TIMS). Strontium isotope ratios are reported as differences from modern seawater in parts per thousand.

The first samples tested were cuttings from borehole USW UZ-14. These samples were used primarily to determine the best experimental procedure. Most of these samples were leached with water, and then leached again with weak hydrochloric acid (HCl). From these results, we determined that leaching times of less than an hour were adequate to dissolve pore-water salts and that longer times may leach strontium from the rock or secondary minerals. The HCl leaches calcite with a strontium isotope composition similar to the pore water. The close comparison of ^{87}Sr from water-leached pore-water salts and that of water squeezed in adjacent core in this borehole indicates that the extraction method is valid. Strontium isotope compositions in the host volcanic rocks are distinct, indicating that pore water has not reached equilibrium with the tuffs.

Once we were satisfied with the experimental technique, we sampled whole core from USW SD-7, excluding core containing macroscopic calcite coatings. The pore water strontium data obtained from USW SD-7 are remarkable in their systematic variation with depth and their distinction from whole rock compositions. At the top of the core, the ^{87}Sr in the pore water is 3.6,

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matching the $\delta^{87}\text{Sr}$ found in surface coatings of calcite at the drill pad. Delta- ^{87}Sr increases with depth; this increase is especially evident within the nonwelded units of the Paintbrush Tuff (PTn) and is only slightly discernible in the underlying welded Topopah Spring Tuff (TSw).

Although the data preclude local equilibrium between pore-water and rock, the rock data can predict the strontium isotope composition of the pore water if recalculated as a down-hole cumulative value weighted according to strontium content of the rock samples and the associated depth interval. An additional weighting factor that takes into account the higher reactivity of the PTn provides a close match to observed pore-water strontium values throughout most of the TSw. Deviations of the pore water $\delta^{87}\text{Sr}$ from the predicted values are likely due to the presence of clays or zeolites which may contain a long-lived record of pore-water strontium compositions and could have been partially leached in the laboratory.

Our working model assumes that strontium is added to infiltrating water by dissolution of calcite in the soil zone. During times of increased surface vegetation, soil waters are more acidic and volcanic detritus in the soil zone can contribute radiogenic strontium into infiltrating water, thus increasing the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in the thick calcretes formed during these times. There are two separate populations of soil carbonate that can contribute strontium to infiltrating water; one is dominated by eolian carbonate with a $\delta^{87}\text{Sr} = 3.6$ and the other is dominated by calcretes in thick alluvial soils with $\delta^{87}\text{Sr} = 4.5$. Although the eolian signal, represented by soil A/B horizons and calcite coatings on bedrock surfaces, exists throughout the Yucca Mountain region, the strontium signal from calcretes may dominate the strontium contributed to infiltration where they are present. Water infiltrating into welded tuff (Tiva Canyon Tuff or Topopah Spring Tuff) tends to retain the strontium isotope composition of the overlying soil. However, the pore water data indicate that water infiltrating

into or percolating through nonwelded tuff (PTn) reacts readily and acquires a strontium isotope signature reflecting interaction with this unit. The strontium isotope composition of the volcanic rocks changes systematically over millions of years due to the decay of ^{87}Rb . As a result, pore waters interacting with these rocks would have $\delta^{87}\text{Sr}$ values that decrease linearly with age. Delta- ^{87}Sr of pore waters in the TSw unit are predicted to change from a modern value of 4.9 to about 0.4 at the time of TSw emplacement (12.7 Ma). The observation that $\delta^{87}\text{Sr}$ varies with microstratigraphic position within thick calcite coatings that occur in the TSw and the match between these ratios and the predicted values strongly suggests that the calcite coatings derive their strontium from the same source as the pore waters; in other words, both pore water and the fracture water leading to calcite deposition within the TSw derive strontium from water-rock interaction in the overlying section, dominantly in the nonwelded units. This model indicates that pore waters in the PTn are redistributed between pore and fracture water that subsequently percolates through the TSw.