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STRONTIUM ISOTOPE GEOCHEMISTRY OF SOIL AND PLAYA DEPOSITS NEAR YUCCA MOUNTAIN, NEVADA

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ABSTRACT

The isotopic composition of strontium contained in the carbonate fractions of soils provides an excellent tracer which can be used to test models for their origin. This paper reports data on surface coatings and cements, eolian sediments, playas and alluvial fan soils which help to constrain a model for formation of the extensive calcretes and fault fillings in the Yucca Mountain region. The playas contain carbonate with a wide range of strontium compositions; further work will be required to fully understand their possible contributions to the pedogenic carbonate system. Soils from an alluvial fan to the west of Yucca Mountain show that only small amounts of strontium are derived from weathering of silicate detritus. However, calcretes from a fan draining a carbonate terrane have strontium compositions dominated locally by the limestone strontium component. Although much evidence points to an eolian source for at least some of the strontium in the pedogenic carbonates near Yucca Mountain, an additional component or past variation of strontium composition in the eolian source is required to model the pedogenic carbonate system.

fault-filling calcite-silica veins is that these deposits should have characteristics in common with the extensive calcretes and other forms of pedogenic carbonate in the area. A complete understanding of the formation of the pedogenic carbonate bears directly on the question of the origin of the calcite-silica veins.

Previous studies⁴⁻⁶ of strontium isotopes contained in calcite from these pedogenic deposits have been instrumental in showing a common origin for most of the materials. In addition, the formulation of a model for the origin of the strontium (and, by inference, calcium) has been proposed⁷ but is not conclusive due to the incomplete characterization of strontium isotopes in possible source materials. In this paper, the model for the formation of pedogenic carbonate is re-examined in light of new strontium isotope data from various types of surficial carbonate deposits from the vicinity of Yucca Mountain. In addition, samples of calcrete from an alluvial fan draining an extensive Paleozoic limestone terrane provide data on the extent of local strontium contribution to these deposits.

I. INTRODUCTION

A large body of geological and geochemical data collected from various sites in the vicinity of Yucca Mountain show that pedogenic carbonate is ubiquitous within a few meters of the surface. In addition, calcite fracture coatings and small veins of calcite in the unsaturated zone have affinities to the pedogenic calcite although their characteristics require a more complex origin.^{1,2} Infillings of secondary carbonate, opaline silica, and other materials along fault zones such as the so-called "calcite-silica veins" exposed in trenches across the Bow Ridge fault on Exile Hill are also of pedogenic origin.³⁻⁵ A primary test of the hypothesis that downward-percolating surface water formed the extensive

II. MODEL

Models describing the formation of secondary calcium carbonate that cements loose, porous sediments in soils of arid and semi-arid regions usually require an allogenic component for the source of the large amounts of calcium carbonate.⁸ Because strontium is chemically similar to calcium and is a common minor or trace element in calcium-bearing minerals, its isotopic composition is an ideal tracer for the origin of calcium. Analyses of strontium in the pedogenic carbonates as well as various possible source materials provide important constraints on the origin of the alkaline earths in the pedogenic calcite.

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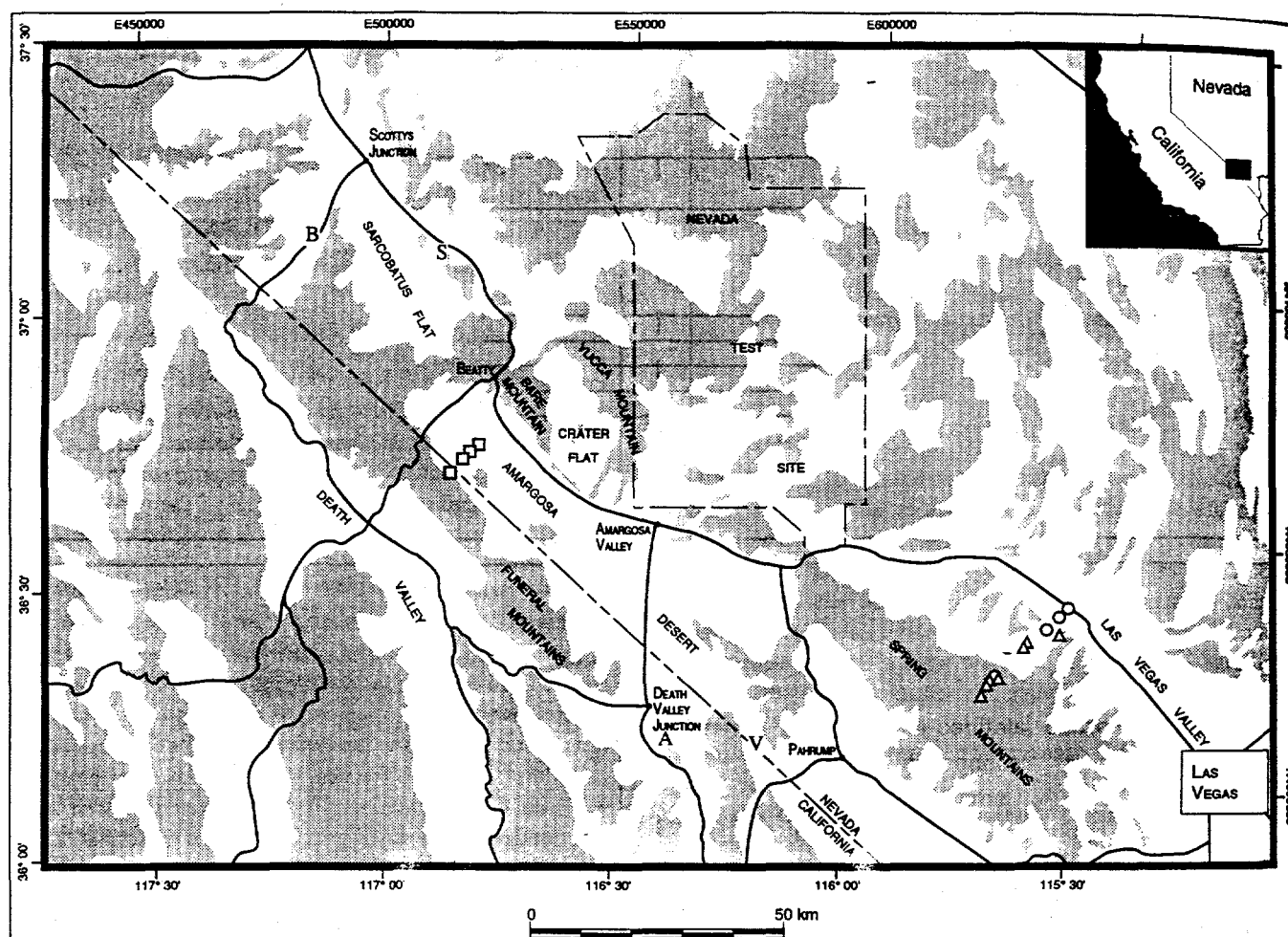


Figure 1. Map of the Yucca Mountain region showing localities discussed in the text; the upland areas are shaded. Bonnie Claire Lake, Sarcobatus Flat, Alkali Flat, and Stewart Valley playa samples are shown as B, S, A, and V, respectively. The soil samples from the alluvial fan emanating from the Funeral Mountains are shown as squares, and the Lee Canyon calcrete and bedrock samples are shown as circles and triangles, respectively.

Strontium isotopes in soil K horizons, as determined by analysis of the acid-soluble fractions of calcretes and rhizoliths have compositions characterized by $^{87}\text{Sr}/^{86}\text{Sr}$ values of 0.7117-0.7127. The lower values of $^{87}\text{Sr}/^{86}\text{Sr}$ are essentially identical to those of calcite in eolian sediments, surface coatings, and soil A/B horizons. Therefore, the model proposed for the pedogenic carbonate started with the eolian component as an end-member composition. However, there are two problems with this hypothesis: first, the samples taken as representative of eolian materials may not be good proxies for the actual dust flux either today or in the recent past and second, there is a paucity of other materials with higher $^{87}\text{Sr}/^{86}\text{Sr}$ required to explain the strontium composition of the pedogenic calcite. This latter problem was addressed by analyzing silicate residues from pedogenic carbonate samples and by making comparisons with the known $^{87}\text{Sr}/^{86}\text{Sr}$ of the extensive volcanic rocks in the Yucca Mountain area. However, the residues from the

pedogenic calcites are, in general, too depleted in radiogenic strontium to serve as the other end member. In addition, while volcanic rocks exposed at the surface have more radiogenic strontium, these rocks have low strontium concentrations, thus requiring a large amount of material to be weathered and leached in order to make a significant contribution to the strontium in the pedogenic carbonate. Therefore, the model presented previously may not be viable and further studies aimed at exploring possible variations in the eolian component are presented in this paper.

III. RESULTS

A. Surface Coatings and Cements

Surface coatings and cements of calcite occur in much of the Yucca Mountain region (Fig. 1) especially where surface water ponds or flows over bedrock. Because these

deposits are found on both calcium-rich (limestone) and calcium-poor (welded tuff) substrates, an eolian parent material is implied. Strontium isotope analyses of 11 surface coating-substrate pairs show that the surface coatings have a consistent $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7116 ± 0.0008 regardless of the substrate composition. The strontium-isotope compositions of the substrates vary from 0.708 to 0.716 (Fig. 2). This suggests that the source of the strontium in the coatings is homogeneous over a large area for the period represented by these deposits.

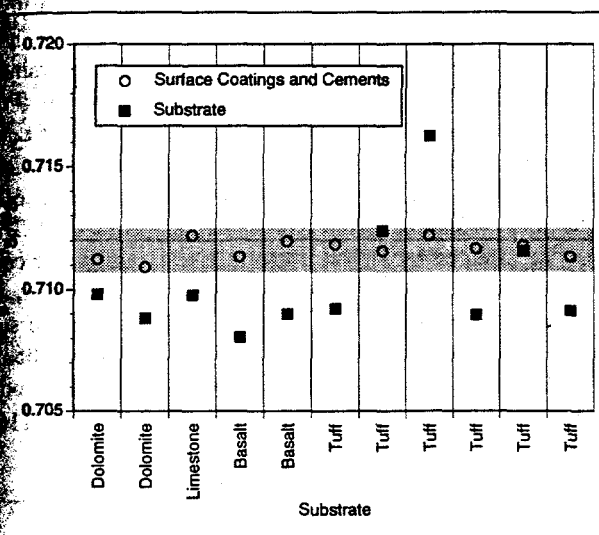


Figure 2. Strontium-isotope composition of surface coatings and substrate pairs. The substrate rock type is shown on the x-axis. The gray band emphasizes the similar strontium compositions of the surface coatings. Errors are within the size of the symbols.

B. Eolian Sediments

Collecting modern or ancient samples which are thought to be representative of the regional eolian dust flux has proven to be difficult. Few field settings have exposures of unambiguous eolian sediments, although silt-rich vesicular A horizons, hypothesized to be of eolian origin are ubiquitous in the region.⁸ Three samples which can be referred to as of eolian origin have been collected to date. Two samples were collected from dust accumulations in the ruins of a mill near Carrara on the west side of Bare Mountain (Fig. 1) and dune sand was sampled from adjacent to a mesquite bush near Big Dune in the Amargosa Desert. The latter sample is dominated by sand-size sediments whereas the former are dominated by silts. Both sites are unconsolidated deposits which have accumulated within the latest Holocene. About 25% of the silt-sized material from Carrara is leachable carbonate with $^{87}\text{Sr}/^{86}\text{Sr} = 0.7117$, similar to the surface-coating samples discussed above. However the sand-sized material from Big Dune is only 1% carbon-

ate with $^{87}\text{Sr}/^{86}\text{Sr} = 0.7123$. Strontium concentrations in the whole sample and in the residue indicate that 14% of the strontium in the Big Dune sample and 44% of the strontium in the Carrara sample reside in the carbonate fraction. The similarity of the strontium in the Carrara samples with that in the surface coating samples suggests that both of these materials derive their carbonate component from silt and finer eolian dust. The sand-sized material is probably a lesser contributor to the pedogenic carbonates because it contains much less carbonate.

C. Playas

Playas contain large accumulations of sediment that, when dry, could be a major source of eolian dust.⁸ Characterization of this eolian component is not a straightforward task because dust flux is variable in both time and space. In order to gain some insight into possible variations, samples of sediment deposited on playas were analyzed for strontium isotopes. The playa samples were collected from sites in the vicinity of Yucca Mountain including (from north to south) Bonnie Claire Lake, Sarcobatus Flat, Alkali Flat, and Stewart Valley (Fig. 1). The samples were collected with an auger and generally represent the top ≈ 10 cm of sediment, which is mostly silt and clay. The samples were collected from near the centers of the playas during the dry season.

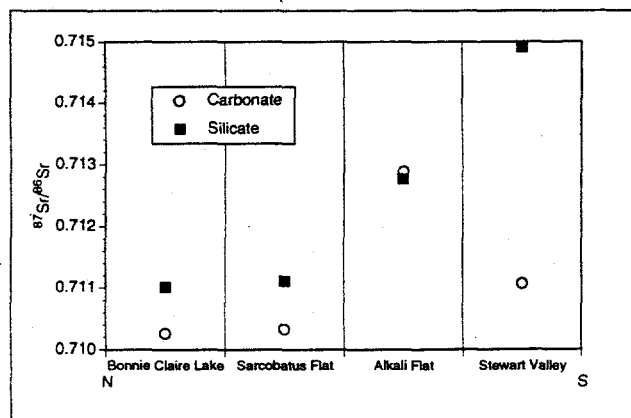


Figure 3. Strontium-isotope composition of four playa samples for which both the carbonate and silicate portions were analyzed. Errors are within the size of the symbols.

Elemental analyses (by X-ray fluorescence) of the playa samples were performed before and after leaching with acid to remove the carbonate component. Of the elements analyzed (K, Ca, Ti, Rb, Sr, Y, Zr, Nb, Ba, La, Ce), only strontium and calcium are severely affected by acid leaching. Strontium concentrations in the original samples are one to six times that in the acid-leached residues and calcium is reduced by factors of three to ten by acid leaching. Gravimetrically, 11-34% of the original samples was

dissolved by the acid. Both hydrochloric (1N) and acetic (10%) acids were used to leach the samples and $^{87}\text{Sr}/^{86}\text{Sr}$ values of the two leachates were identical in each of the four playa basins, although there are significant differences between basins. The strontium isotope systematics of these four samples show that the playas obtain carbonate-bound and soluble strontium from disparate sources, perhaps mostly from surrounding mountains. $^{87}\text{Sr}/^{86}\text{Sr}$ in the acid-soluble fractions ranges from 0.7103 to 0.7129 and that of the residues from 0.7110 to 0.7149 (Fig. 3). The playas probably contain a large eolian component although some strontium is likely derived from surface runoff and, in some cases, from seepage. The Alkali Flat sample contains 34% carbonate with $^{87}\text{Sr}/^{86}\text{Sr} = 0.7129$. If material represented by the Alkali Flat sample contributed substantially to the eolian flux in the past then the isotopic composition of the pedogenic carbonate in the vicinity of Yucca Mountain could be explained as a mixture of this source and the modern eolian flux. From a geographic perspective, the two samples to the north have strontium ratios lower than the eolian component at Yucca Mountain and the two samples to the south have both lower and higher strontium ratios. Closer to Yucca Mountain, two carbonate fractions leached from grab samples of Peters Playa have $^{87}\text{Sr}/^{86}\text{Sr} = 0.7125$ and 0.7142, showing a wide variation in composition from a small area. The lower ratio is from a sample collected from an active mine, and therefore may represent an exhumed horizon. It is clear from these few analyses of playas near Yucca Mountain that it may be desirable to obtain shallow core samples of some of the playas to determine a stratigraphy of the eolian flux. Also, the relative contributions to the playas from local runoff, groundwater discharge, and eolian dust need to be assessed. Whereas the playas may play a role in the formation of pedogenic carbonate (e.g., as a source of dust), their exact role may depend on climate and they, in themselves, are probably a complicated system.⁹

D. Alluvial Fan Soils

Previous analysis of carbonate in soils (A/B horizons) near Yucca Mountain⁶ indicated a homogeneous $^{87}\text{Sr}/^{86}\text{Sr}$ for both shallow (1-4 cm) and deep (≈ 20 cm) samples of 0.7117 ± 0.0002 , based on two samples from Trench 14 and one sample from Crater Flat. This strontium ratio is similar to that of surface coatings and indicates an eolian source for the strontium and calcium in these soil horizons. For this study, samples of the soil A-horizons on an alluvial fan draining the Funeral Mountains were collected. The Funeral Mountains are locally dominated by a late Precambrian terrane of quartzite, schist and marble that should have a fairly radiogenic strontium signature in contrast to the pedogenic carbonates. Four samples of the A horizon

were collected, immediately below the desert pavement, from soils developed on the alluvial fan at various distances from the mountain front. These samples range from 5 to 15% carbonate and were sampled to test the influence of surface runoff from a radiogenic strontium source on the soil carbonate system. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the carbonate and silicate components of these soils are plotted in Figure 4, which shows disparate behavior of the two systems. The silicate system clearly shows an increasing influence of strontium derived from radiogenic rocks of the Funeral Mountains terrane toward the mountain front. To a much lesser extent, the carbonate system also shows this influence, possibly due to leaching of feldspars in the drainage and upper fan areas and subsequent deposition of this strontium as carbonate in the soil. Because surface runoff rarely reaches the distal portions of the fan, the lower samples have strontium-isotope compositions closer to the other A/B horizon samples from Crater Flat and Trench 14. They may be affected by strontium from a less radiogenic component in the silicates at these elevations. However, the important point is that the carbonate system in these soils on the west side of the Amargosa Desert is still fairly homogeneous and when these data are combined with the other soil A/B horizons the overall average $^{87}\text{Sr}/^{86}\text{Sr}$ is 0.7116 ± 0.0007 .

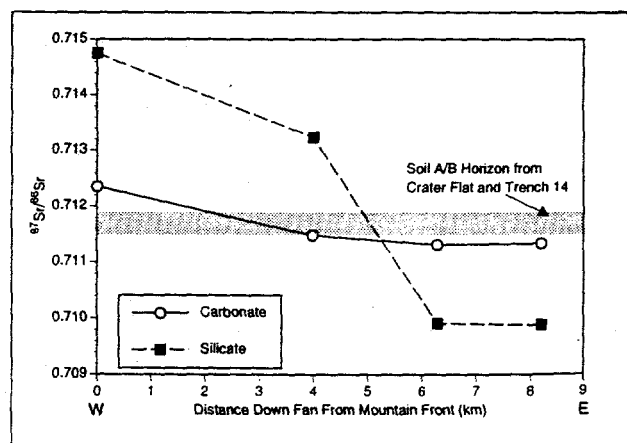


Figure 4. Strontium isotopic composition of four soil A horizon samples from an alluvial fan on the west side of the Amargosa Desert which drains the Funeral Mountains (Fig. 1). The range of $^{87}\text{Sr}/^{86}\text{Sr}$ in soil A/B horizons from Crater Flat and Trench 14 is shown for comparison. Errors are within the size of the symbols.

The strontium data on the pedogenic carbonate system components, comprising surface coatings, eolian sediments, playas, soil A/B horizons, and soil K horizons, is summarized in Figure 5. Note that 10 out of 11 surface coating samples show the same range of $^{87}\text{Sr}/^{86}\text{Sr}$ as the seven soil (A/B horizon) samples. The eolian sediments

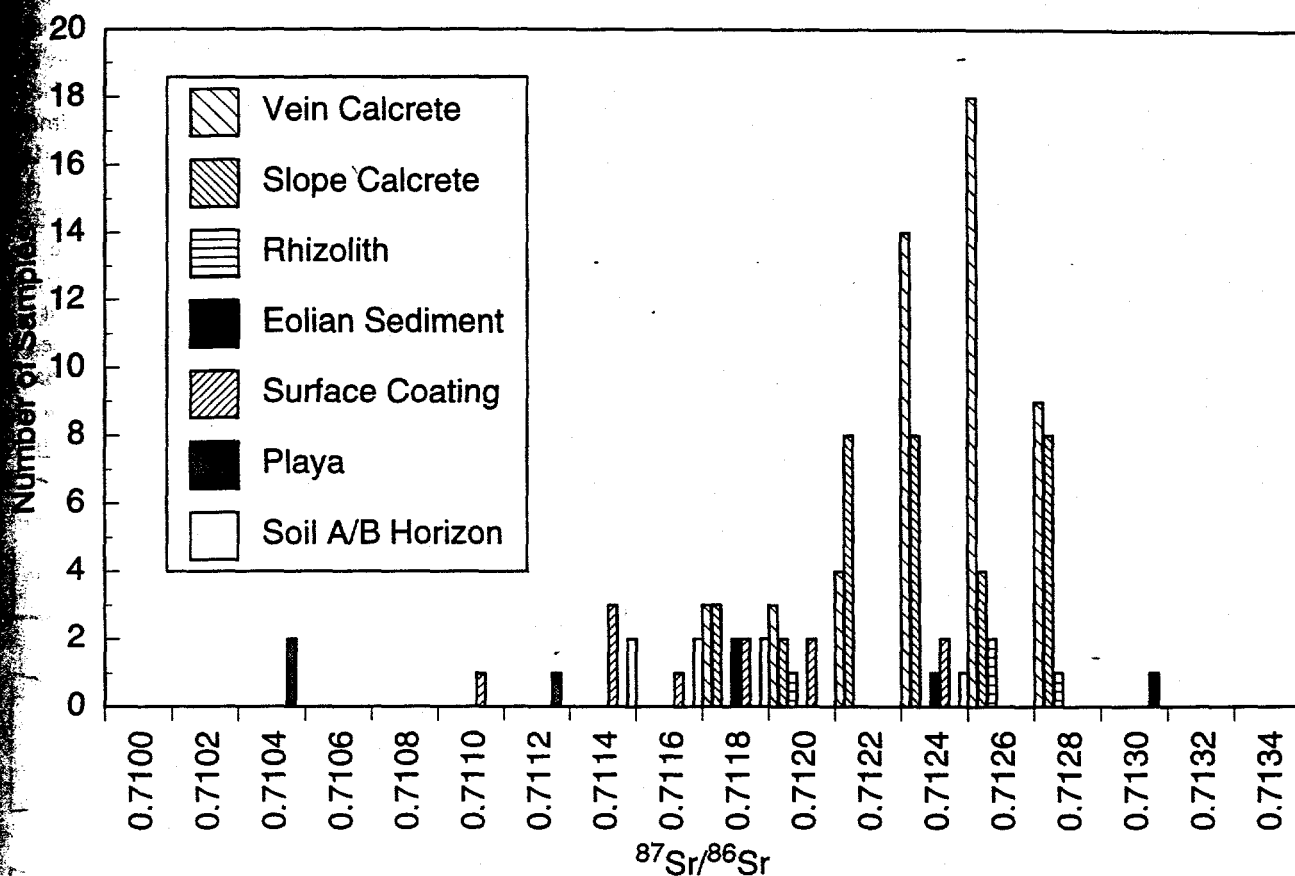


Figure 5. Histogram of strontium isotopic compositions for the various types of pedogenic deposits. Vein calcrete, slope calcrete and rhizoliths are all expressions of soil K horizons.

show a more restricted range of strontium ratios but lie entirely within the range of the soils and surface coatings. If the maximum $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in these three types of samples is 0.7124, then the extensive soil K horizons (calcretes) cannot be derived entirely from the eolian flux of carbonate as defined by this range of values for recent eolian deposits. However, the playa samples span the entire compositional range of the other materials, suggesting that the eolian component may vary spatially. Therefore, an eolian origin of K horizons which are 10's to 100's of thousands of years old¹⁰ cannot be ruled out, particularly in light of the possibility that the strontium-isotope composition of the playa sediments may have varied over time.

E. Lee Canyon Fan Calcrete

In another test of the strontium systematics in pedogenic carbonate, we analyzed four samples of thin (≈ 1 cm) calcretes exposed in gullies which dissect the alluvial fan emanating from the northeastern Spring Mountains at Lee Canyon.¹¹ The drainage for this fan is entirely within the Paleozoic limestone section which has already been

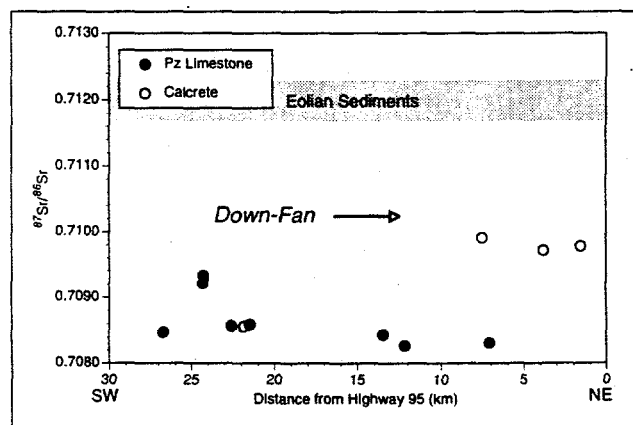


Figure 6. Strontium isotopic compositions of calcretes from the Lee Canyon fan (Fig. 1) as compared to limestone samples from the drainage of the fan. The gray band is the range of $^{87}\text{Sr}/^{86}\text{Sr}$ in three eolian sediment samples. Errors are within the size of the symbols.

characterized by strontium isotope analyses.⁶ The limestone outcrops in the upper reaches of Lee Canyon have $^{87}\text{Sr}/^{86}\text{Sr}$

ratios similar to those of unaltered lower Paleozoic limestones (0.7083 to 0.7093). The calcrete sample which occurs within sediments surrounded by limestone outcrops has a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7086, essentially identical with the ratios of the closest limestone samples (Fig. 6). The three samples collected from the lower part of the fan have distinctly higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (≈ 0.7098) which reflect the incorporation of the allogenic (eolian) component. The implication of these results is that surface runoff from an extensive limestone terrane carries a significant strontium load which can overwhelm the eolian flux into the pedogenic system at least locally, and affect calcretes for many kilometers down-gradient.

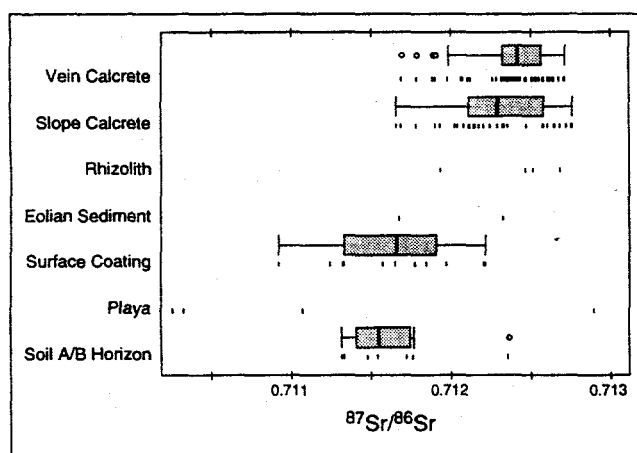


Figure 7. Dot and box plots of strontium isotopic compositions showing the distribution of values for each type of carbonate deposit. The boxes are only shown for sample types with a larger number of analyses. The boxes define the middle 50% of the data with a vertical line for the median, whiskers for the total range and circles for outliers.

IV. CONCLUSIONS

Through these additional strontium isotope analyses, a better understanding of the origins of secondary carbonates at Yucca Mountain is being elucidated. The various types of soil K horizon calcite (calcrete and rhizoliths) show a fairly tight grouping (Fig. 7). As discussed above, the eolian sediments and their direct derivatives (surface coatings and soil A/B horizons) also form a fairly tight cluster. The disparate $^{87}\text{Sr}/^{86}\text{Sr}$ values for these two broad groups is evidence that the strontium (and calcium) in the pedogenic K horizon carbonates cannot be derived entirely from a component represented by modern eolian carbonate dust. The eolian component may be a major contributor but it is not clear to what extent it varied with time or how ancient climatic regimes may have affected it.¹² If it is assumed that the modern eolian flux is the same as the ancient flux in terms

of strontium isotopes then an additional component is required to explain the strontium-isotope compositions of the K horizon carbonate. This component could come from the playas to the south (via eolian flux) or it could come from local runoff, which reacted with the silicate bedrock or detritus to obtain more radiogenic strontium. The latter hypothesis seems unlikely owing to the fact that there is little spatial variation in the strontium composition of the K horizon carbonate and, at least in some cases, the local detritus is less radiogenic than the pedogenic calcite. Also, the data from the Funeral Mountains alluvial fan is at least suggestive of little contribution from strontium leached out of silicates; other studies of similar deposits may reveal situations where a silicate component is more dominant.

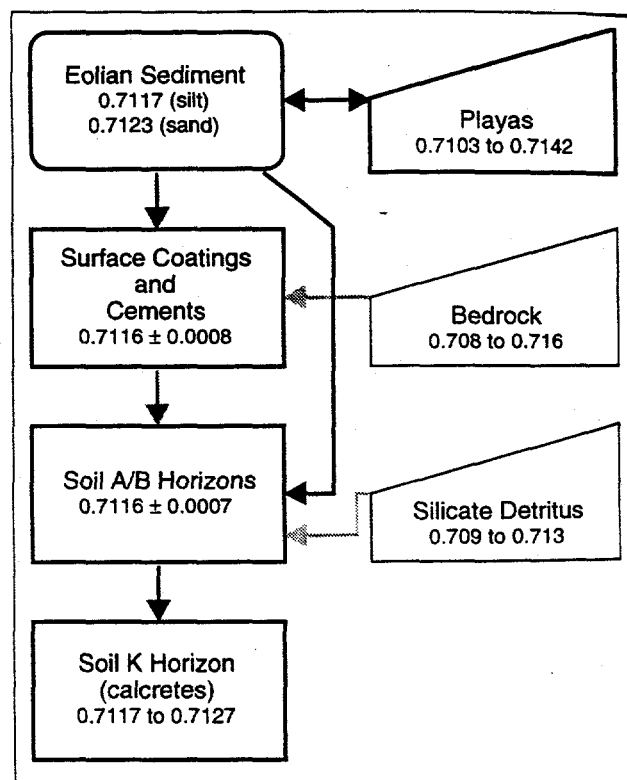


Figure 8. Box model for the strontium data on the pedogenic carbonate system. $^{87}\text{Sr}/^{86}\text{Sr}$ ranges are shown in each box; see text for discussion. The lighter gray arrows indicate lesser contributions for some components.

More work will be necessary before a conclusive model for the origin of the pedogenic carbonate can be demonstrated. The model currently hypothesized is best represented by the box diagram shown in Figure 8. Eolian sediment dominated by silt-size carbonate may be connected to playas as sinks or sources depending on climate. The eolian sediment directly forms surface coatings on bedrock and cements sediments in channels by solution and reprecipitation; it is also incorporated into soils and may

the notable K horizon calcretes over time. The surface may also contribute their carbonate to soils during events. Bedrock and even local silicate detritus contribute only minor amounts of cations to the secondary calcretes. Future studies on the playas and past climate may help to eliminate the uncertainties in the model.

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