

ISOTOPIC TRACERS OF GOLD DEPOSITION IN PALEOZOIC LIMESTONES,  
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## ABSTRACT

Strontium isotopic analyses of barren and mineralized Paleozoic carbonate rocks show that hydrothermal fluids added radiogenic strontium ( $^{87}\text{Sr}$ ) to the mineralized zones. At Bare Mountain, samples collected from mineralized areas have  $\delta^{87}\text{Sr}$  values (per mil deviation from primary marine values) ranging from +3.0 to +23.0 (mean of this log-normal distribution is +7.0), whereas unmineralized carbonate rocks have  $\delta^{87}\text{Sr}$  values of -0.6 to +2.9 (mean of  $+1.07 \pm 1.03$ ). In other ranges (Striped Hills, Spring Mountains, and ranges in the vicinity of Indian Springs Valley),  $\delta^{87}\text{Sr}$  values of the unmineralized carbonate rocks are even lower and virtually indistinguishable from primary marine values. This correlation of elevated  $\delta^{87}\text{Sr}$  values with mineralized zones provides a useful technique for assessing the mineral potential of the Paleozoic basement beneath Yucca Mountain, and may find broader use in mineral exploration in the Basin and Range province as a whole.

## INTRODUCTION

The objective of this study is to evaluate the usefulness of strontium isotopes as indicators of epithermal, heavy metal mineralization in Paleozoic and Late Proterozoic limestones of southern Nevada, especially in the vicinity of Yucca Mountain. The study is premised on the concept that ascending hydrothermal fluids would have transported radiogenic strontium (enriched in the radiogenic isotope  $^{87}\text{Sr}$ ) from Precambrian basement rocks into the host rocks, thus causing a detectable increase in the  $^{87}\text{Sr}/^{86}\text{Sr}$  value of the mineralized rocks. The approach was to collect and analyze mineralized and unmineralized carbonate rocks from the region around Yucca Mountain.

Yucca Mountain, located as it is in the resource-rich Basin and Range province of the western United States, is virtually surrounded by mineral deposits, many of which have been worked or are presently being exploited.<sup>1,2</sup> Any mineral deposits, if present at Yucca Mountain proper, could attract future exploration and development that might compromise the integrity of a nuclear repository. Therefore, careful assessment of the mineral resource potential of Yucca Mountain is an important part of site suitability studies.

Precious metal deposits in this region, notably gold, are commonly hosted either by Paleozoic carbonate rocks or by Tertiary volcanic rocks, both of which occur at Yucca Mountain.<sup>3,4</sup> The sediment-hosted (Paleozoic rocks), disseminated gold deposits are commonly referred to as Carlin-type deposits. Late Proterozoic and Paleozoic marine limestones were mineralized in the Cretaceous and Tertiary by hydrothermal solutions that deposited micron- to submicron-size gold particles.<sup>4,5,6</sup> Ore deposition was commonly structurally controlled,<sup>4</sup> and in some areas shallow-dipping thrust or detachment faults were important pathways for the mineralizing solutions.<sup>7</sup> These Late Proterozoic and Paleozoic shelf carbonate rocks crop out extensively in the mountain ranges of southern Nevada. Bare Mountain, immediately west of Yucca Mountain, contains strongly mineralized carbonate rocks (Fig. 1).<sup>8</sup> Other ranges such as the Striped Hills, the Specter Range, and the Spring Mountains are composed of the same stratigraphic sequences, but these rocks are unmineralized. The Paleozoic limestone terrain is overlain at Yucca Mountain by Miocene volcanic rocks, and assessment of the mineral resource potential of Yucca Mountain must include an evaluation of this carbonate-rock basement beneath the mountain. Because the Paleozoic carbonate is

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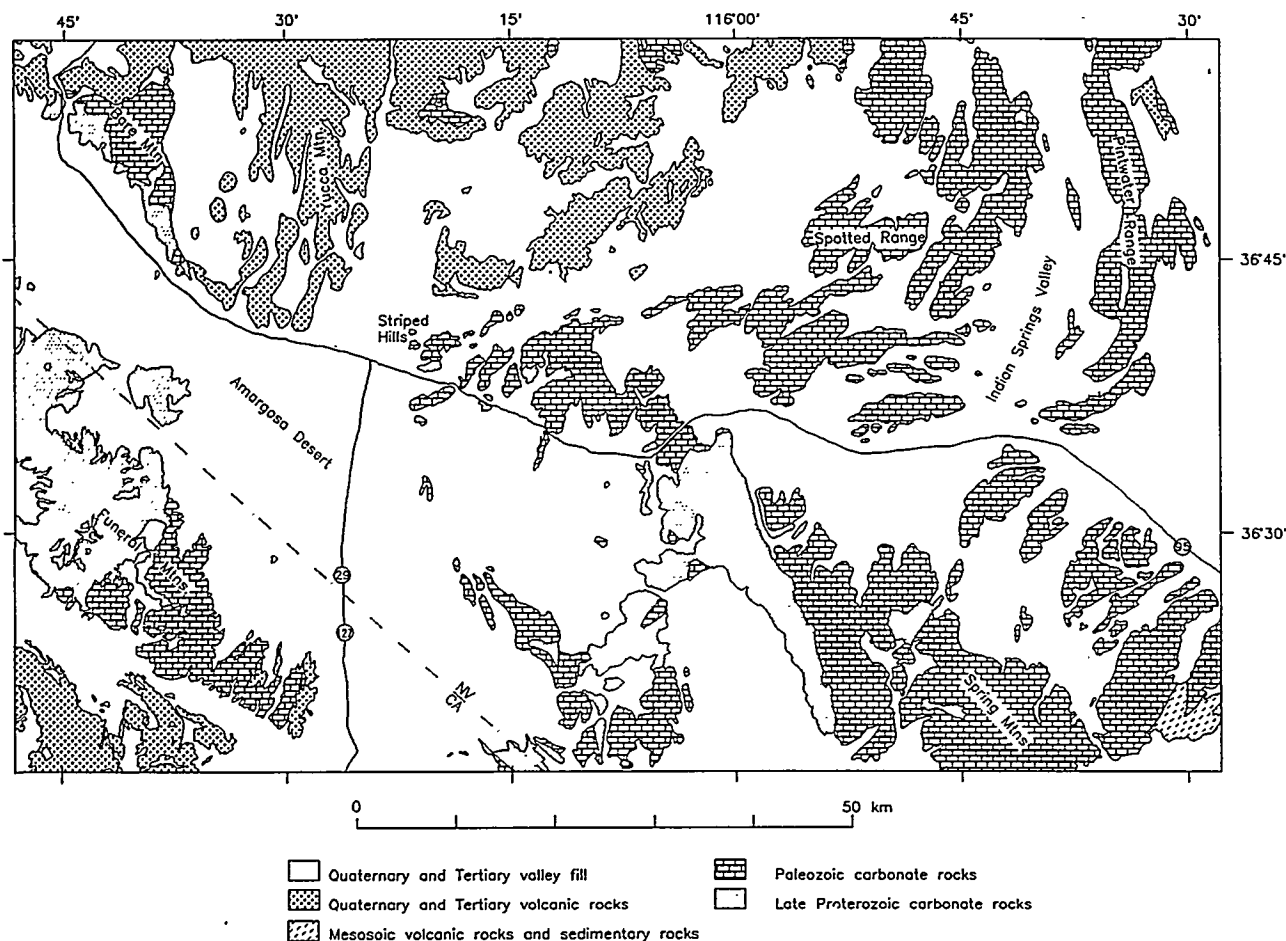


Figure 1 Regional geologic sketch map.

buried beneath the volcanic rocks, evaluation of its mineral potential must be made on the basis of core samples recovered in deep drilling. Physical signs of mineralized zones such as jasperoid or sulfide-rich zones, features commonly used as prospecting guides,<sup>4</sup> might not be encountered in widely spaced drill holes. Therefore, techniques that could be used to detect subtle, large-scale geochemical or isotopic anomalies associated with mineralized zones would be highly useful in evaluating the mineral potential of these rocks.

#### CONCEPTUAL MODEL

Romberger<sup>4</sup> summarized the characteristic features of a number of sediment- and volcanic-hosted disseminated gold deposits in the Great Basin and developed a conceptual genetic model which serves as the basis for the present investigation. He envisioned a hydrogeologic setting in which shallow, oxygenated ground water overlies a reducing ground water perhaps approaching chemical equilibrium with its host rocks. This system is perturbed by an increase in the geothermal gradient, and heated

plumes of the deep fluids move upward into the oxygenated zone where reactions facilitate the deposition of gold and other commonly associated elements such as mercury, arsenic, antimony, thallium, and barium. To this conceptual model, we add the postulate that the deeper fluids contain strontium with  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios significantly larger than those of Paleozoic marine carbonate rocks. As discussed below, addition of the exotic strontium to the rock strontium produces an elevated  $^{87}\text{Sr}/^{86}\text{Sr}$  in the mineralized zone. That the calcareous host rocks were open systems during mineralization, especially in the alkaline-earth elements, is well documented at the Carlin deposit.<sup>9</sup> Furthermore, previous strontium isotopic studies of carbonate-hosted mineral deposits have yielded encouraging results from the standpoint of detecting the effects of hydrothermal activity on  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios.<sup>10,11</sup>

Because of its long residence time, strontium is isotopically uniform throughout the earth's oceans at any particular point in geologic time. However,  $^{87}\text{Sr}/^{86}\text{Sr}$  values in the oceans have varied systematically with time throughout the Phanerozoic and Late Proterozoic in

response to global input from rivers and from hydrothermal exchange with oceanic basalt, especially near active rises.<sup>12,13,14</sup>

When marine limestones are deposited, they incorporate strontium with the  $^{87}\text{Sr}/^{86}\text{Sr}$  value of the sea water at that time. Thus, if the age of a limestone is known, its original  $^{87}\text{Sr}/^{86}\text{Sr}$  value can be estimated from the sea water strontium-isotope curve (Fig. 2). Because calcite and aragonite essentially exclude rubidium from their lattices ( $^{87}\text{Rb}$  is the radioactive parent of  $^{87}\text{Sr}$  with a half life = 48.8 m.y.), the original  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio will not change significantly with time provided the rock remains a closed system. If strontium of a different isotopic composition is added to, or exchanged with, the original strontium in the limestone, the primary  $^{87}\text{Sr}/^{86}\text{Sr}$  will be modified from its primary depositional value towards that of the extraneous or exotic strontium. Such modification of primary marine  $^{87}\text{Sr}/^{86}\text{Sr}$  values of carbonate rocks can result from the exchange of strontium with fluids during diagenesis or dolomitization, or from the introduction of exotic strontium by hydrothermal fluids that have interacted or equilibrated with Precambrian basement rocks. The first process would not likely result in significant changes in  $^{87}\text{Sr}/^{86}\text{Sr}$  values other than to integrate or smooth  $^{87}\text{Sr}/^{86}\text{Sr}$  values over a finite stratigraphic thickness. However, the introduction of strontium via hydrothermal fluids can substantially increase the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of the affected carbonate rocks. Thus from a mineral assessment standpoint, carbonate rocks with  $^{87}\text{Sr}/^{86}\text{Sr}$  values approximately consistent with their age and the marine curve (Fig. 2) would be considered unlikely to contain epigenetic mineralization. In contrast, anomalously large  $^{87}\text{Sr}/^{86}\text{Sr}$  values could indicate hydrothermal introduction of radiogenic strontium and therefore the potential for mineralization.

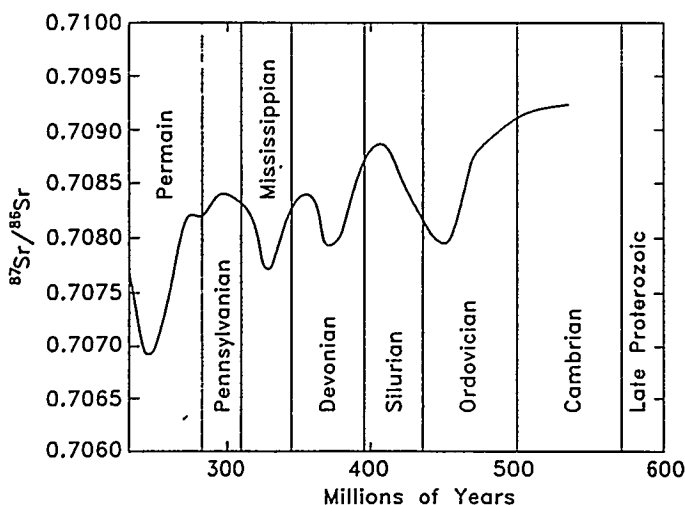


Figure 2 Temporal variation in the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of sea water during the Paleozoic. Adapted from Burke and others.<sup>13</sup>

To emphasize small but highly significant variations in  $^{87}\text{Sr}/^{86}\text{Sr}$  and to allow comparison of  $^{87}\text{Sr}/^{86}\text{Sr}$  of rocks of different ages, it is convenient to express this ratio as a per mil deviation from the value of sea water at the time the rocks were deposited. This value is defined as  $\delta^{87}\text{Sr}_i$ :

$$\delta^{87}\text{Sr}_i = \left\{ \left[ \frac{(^{87}\text{Sr}/^{86}\text{Sr})_i}{(^{87}\text{Sr}/^{86}\text{Sr})_{sw}} \right] - 1 \right\} \cdot 1000$$

where  $(^{87}\text{Sr}/^{86}\text{Sr})_i$  = present-day value of the carbonate rock and  $(^{87}\text{Sr}/^{86}\text{Sr})_{sw}$  = the sea-water value at the time of deposition of the rock.

All the data generated in this study are shown on Fig. 3 as  $\delta^{87}\text{Sr}_i$  values plotted at the ages of the rock units. Depending on location, virtually every unit has both 'normal' and 'elevated' values. In addition to  $\delta^{87}\text{Sr}_i$ ,  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios can also be expressed as per mil deviation from the value for modern sea water (0.70920), and this value is designated simply as  $\delta^{87}\text{Sr}$ .

#### APPROACH AND METHODS

To test the conceptual model outlined above, mineralized and unmineralized Late Proterozoic and Paleozoic limestones were collected from Bare Mountain, and suites of unmineralized samples were collected from the Striped Hills, Spring Mountains, and various ranges in the vicinity of Indian Springs Valley (Fig. 1). In addition, core samples of Silurian limestone from UE-25p#1,<sup>15</sup> the only drill hole at Yucca Mountain that penetrates Paleozoic basement rocks, were analyzed for comparison with the test data set.

In collecting outcrop samples, a potential contaminant is the surficial calcite of pedogenic origin which is ubiquitous as coatings on and fracture fillings in bedrock. This pedogenic calcite has a mean  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of  $0.71235 \pm 0.00027$  ( $\delta^{87}\text{Sr} = +4.4 \pm 0.4$ ).<sup>16</sup> In collecting samples for the present study, surficial calcite coatings and fracture fillings were avoided or trimmed from the selected specimens.

Subsamples of 10 to 20 grams were obtained from hand-specimen-sized samples in the laboratory using a small diamond core drill (0.5 inch diameter); these cores were pulverized to 200-mesh powder in a shatterbox. The samples were analyzed for selected elements, including rubidium and strontium by energy-dispersive, X-ray fluorescence spectrometry. Subgram aliquots of the samples were treated with 1N HCl to selectively dissolve the carbonate fraction, and strontium was purified on ion exchange columns.  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios were determined on either one of two thermal-ionization mass spectrometers—a six-inch radius of curvature NBS (National Bureau of Standards now National Institute of Science and Technolo-

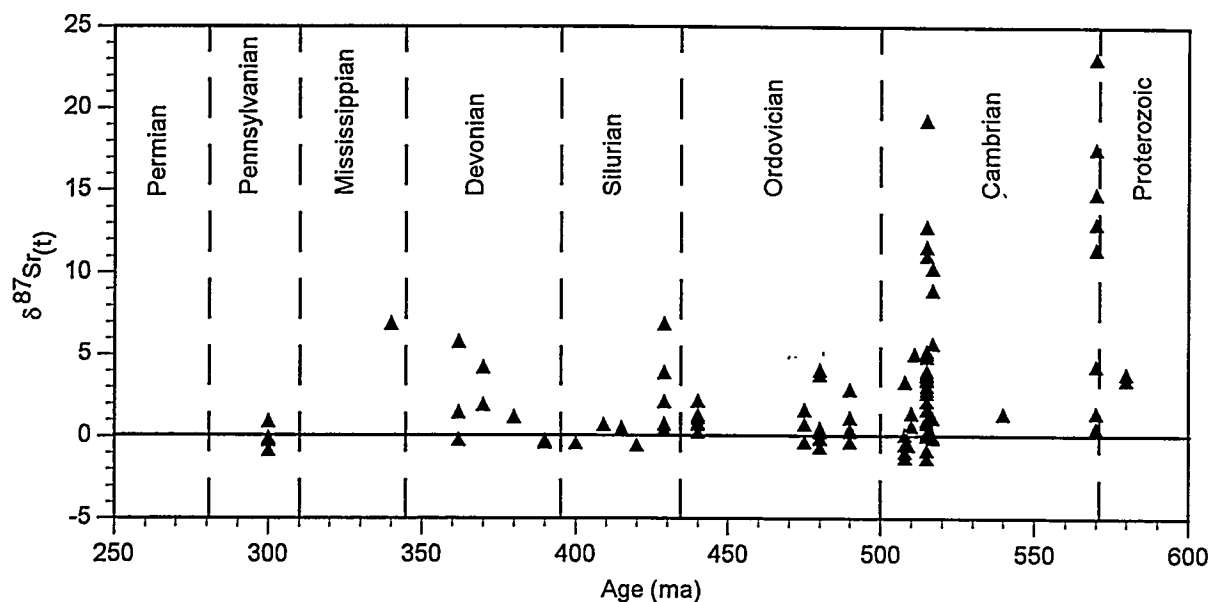


Figure 3  $\delta^{87}\text{Sr}_r$  values for samples as a function of their age (see text for definition of  $\delta^{87}\text{Sr}_r$ ).

gy) mass spectrometer or a Finnigan MAT 262 multisample, fully automated mass spectrometer operating in the static mode. The instruments were calibrated using the USGS standard EN-1, which is calcite from a modern *Tridacna* shell taken from Enewetok Lagoon in the western Pacific Ocean.  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios are adjusted to a value of 0.70920 for modern sea water. Analytical uncertainty in  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios from both instruments is better than  $\pm 0.01$  percent of the value at the 95-percent confidence level. This corresponds to an uncertainty of  $\pm 0.1$  or less in the  $\delta^{87}\text{Sr}_r$  values.

## RESULTS

$\delta^{87}\text{Sr}_r$  values for samples of Paleozoic carbonate rock are plotted on a histogram (Fig. 4). Collectively, the distribution of data points is highly skewed towards large  $\delta^{87}\text{Sr}_r$ . All of the  $\delta^{87}\text{Sr}_r$  values of 3.0 or larger are for samples from Bare Mountain. Data for the other ranges approximate normal distributions with central tendencies near zero. These data will be referred to as the 'Regional' and 'Bare Mountain' sets in the ensuing discussion.

### Regional Data

$\delta^{87}\text{Sr}_r$  values for Paleozoic limestones from the Striped Hills, the Spring Mountains, and mountain ranges in the vicinity of Indian Springs Valley display limited variability (Table 1). The means and limited dispersions ( $1\sigma$ ) for  $\delta^{87}\text{Sr}_r$  of  $+0.43 \pm 0.49$ ,  $-0.18 \pm 0.74$ , and  $+0.14 \pm 0.62$  for the Striped Hills, Spring Mountains, and Indian Springs Valley ranges, respectively, are consistent with primary marine values given the combined uncertainties in the

marine strontium-isotope curve for the Paleozoic and the ages of the rock units. The uncertainties in the marine strontium curve can be as much as  $+0.6\text{‰}$  and  $-0.2\text{‰}$ , from the curve shown in Fig. 2,<sup>13</sup> and where the rate of change in  $^{87}\text{Sr}/^{86}\text{Sr}$  is large, uncertainties in the age of the units contributes added uncertainty to the reference  $^{87}\text{Sr}/^{86}\text{Sr}$  value used to calculate  $\delta^{87}\text{Sr}_r$ . We conclude that the primary or near primary  $\delta^{87}\text{Sr}_r$  signatures of these rocks indicate that they have not been subjected to hydrothermal mineralization. This conclusion is consistent with the lack of indications of ore-bearing minerals in the sampled areas.

### Bare Mountain Data

In contrast to the primary or near primary marine  $\delta^{87}\text{Sr}_r$  values for carbonate rocks of the regional collection described above,  $\delta^{87}\text{Sr}_r$  values for Bare Mountain samples show a bimodal distribution with values as large as  $+23$  (Fig. 4). Sampling at Bare Mountain (Fig. 5) entailed collection from mineralized areas (mines, prospect pits, adits) and unmineralized areas. For the most part, the determined  $\delta^{87}\text{Sr}_r$  values conform to the original field assessment of the samples as being mineralized or unmineralized. However, a few samples thought to be unmineralized have elevated  $\delta^{87}\text{Sr}_r$  values. For example, the first suite of samples obtained from Bare Mountain, collected from Black Marble Hill (Fig. 5) for purposes other than the present study,<sup>16</sup> all have somewhat elevated  $\delta^{87}\text{Sr}_r$  values. Relatively recent mining claims are the only evidence known to us that rocks of Black Marble Hill may be mineralized.

Table 1. Means and standard deviations of  $\delta^{87}\text{Sr}_i$  values for Paleozoic carbonate rocks.

Area	No. of Samples	Mean $\delta^{87}\text{Sr}_i$	Sigma
Spring Mts.	16	-0.18	$\pm 0.74$
Indian Springs Valley	18	+0.14	$\pm 0.62$
Striped Hills	11	+0.43	$\pm 0.49$
Bare Mt. Unmineralized	29	+1.07	$\pm 1.03$
Bare Mt. Mineralized (normal dist.)	35	+8.58	$\pm 5.31$
Bare Mt. Mineralized (log normal dist.)	35	+6.96	+6.24 -3.29

For the following evaluation and discussion, we have separated the  $\delta^{87}\text{Sr}_i$  values from Bare Mountain into two populations based on the aforementioned classification of samples and on inspection of the bimodal distribution (Fig. 4). The population with  $\delta^{87}\text{Sr}_i$  values of less than 3.0 have a mean of  $+1.07 \pm 1.03$  (Table 1). The remaining 35 samples with  $\delta^{87}\text{Sr}_i$  of 3.0 to 23.0 have an arithmetic mean of  $+8.58 \pm 5.31$ . The highly skewed distribution is better approximated as log-normal with a mean of +6.96. The following discussion emphasizes the correspondence of elevated  $\delta^{87}\text{Sr}_i$  to known mineralized areas.

Ore at the Sterling Mine (Fig. 5) is controlled by a low angle fault that thrusts the Late Proterozoic Wood Canyon Formation over the Cambrian Bonanza King Formation.<sup>8,17</sup> Several samples of both units were collected, and all have significantly elevated  $\delta^{87}\text{Sr}_i$ . Within the Bonanza King, an ore shoot localized in a brecciated zone, and adjacent country rock, were sampled underground (Fig. 6). Samples at 22 ft, 9 ft, and 1 ft east of the contact of the ore zone have  $\delta^{87}\text{Sr}_i$  values of +4.8, +5.2, and +3.6, respectively. Two samples 4 ft and 60 ft into the ore shoot have much larger  $\delta^{87}\text{Sr}_i$  values of +11.5 and +19.3. All of these  $\delta^{87}\text{Sr}_i$  values are strongly elevated from primary marine values with the ore zone showing the greatest addition of radiogenic strontium.

Samples obtained from or near other known mineralized areas in Bare Mountain show variably elevated  $\delta^{87}\text{Sr}_i$  values (Fig. 5). Two samples from Tungsten Canyon have  $\delta^{87}\text{Sr}_i$  of +4.3 and +11.4. Three samples from the

vicinity of the Gold Ace Mine have values of +3.5, +3.8, and +11.4. Near the Telluride Mine (Fig. 5), three samples of apparently unmineralized Paleozoic limestone have primary  $\delta^{87}\text{Sr}_i$  values of -0.1, +0.2, and +0.6, whereas one sample of sparry calcite from a prospect pit has an elevated value of +3.7.

The association of large  $\delta^{87}\text{Sr}_i$  values with fault zones at the Sterling Mine, Secret Pass, Black Marble Hill, and the low angle normal fault near Beatty is consistent with the geologic observation of fault-controlled mineralization.<sup>7</sup> Near Beatty, a low angle normal fault (detachment) juxtaposes Miocene volcanic rocks (hanging wall) against Paleozoic and Late Proterozoic rocks.<sup>8</sup> Samples of the Papoose Lake Member of the Bonanza King Formation in the footwall have  $\delta^{87}\text{Sr}_i$  values decreasing from +10.3 at the fault to +8.9 and +5.7 a few tens of meters away from the fault. Three samples of the Wood Canyon in the footwall have  $\delta^{87}\text{Sr}_i$  values of +12.8, +18.9, and +19.8. At Secret Pass (Fig. 5), samples of the Goodwin Limestone and the Fluorspar Canyon Formation have  $\delta^{87}\text{Sr}_i$  values of -0.4 and +1.42, respectively. In contrast, carbonate from a two-foot thick fault zone separating Ordovician and Devonian rocks has a much larger  $\delta^{87}\text{Sr}_i$  of +3.7. Calcite from the shallow-dipping fault at Black Marble Hill has strongly elevated  $\delta^{87}\text{Sr}_i$  of +13.8.

#### UE-25p#1 Data

Drill hole UE-25p#1 at Yucca Mountain (Fig. 1) penetrates 3,954 ft of Miocene volcanic rocks and 1,841 ft of Paleozoic sedimentary rocks assigned to the Silurian Lone Mountain Dolomite and the Roberts Mountain Formation.<sup>15</sup> Two samples of the Lone Mountain Dolomite have  $\delta^{87}\text{Sr}_i$  values of +0.24 and +0.21, and a sample of the Roberts Mountain Formation has a value of +0.04. All of these are consistent with primary marine values for the Middle and Upper Silurian. In contrast, water from the carbonate aquifer in UE-25p#1 has a  $\delta^{87}\text{Sr}$  of +3.6. Assuming that the  $\delta^{87}\text{Sr}_i$  values of three analyzed core samples are isotopically representative of the aquifer, the significantly larger  $\delta^{87}\text{Sr}_i$  for the ground water indicates that it has not equilibrated its strontium with the carbonate rocks at the drill-hole site. Downward leakage from the Tertiary aquifer at this locality cannot explain the  $\delta^{87}\text{Sr}$  value of +3.6 because the hydraulic head in the Paleozoic rocks is 65 to 70 ft higher than the head in most of the overlying volcanic rocks. Furthermore, the strontium concentration in water from the Paleozoic rocks at UE-25p#1 is about eight times higher than that of water from the volcanic rocks.<sup>15</sup> Upgradient interaction with rocks having large  $\delta^{87}\text{Sr}$  appears to be a more likely explanation for the +3.6  $\delta^{87}\text{Sr}$  value for the ground water. Devonian limestones and the Mississippian-Devonian Eleana Forma-

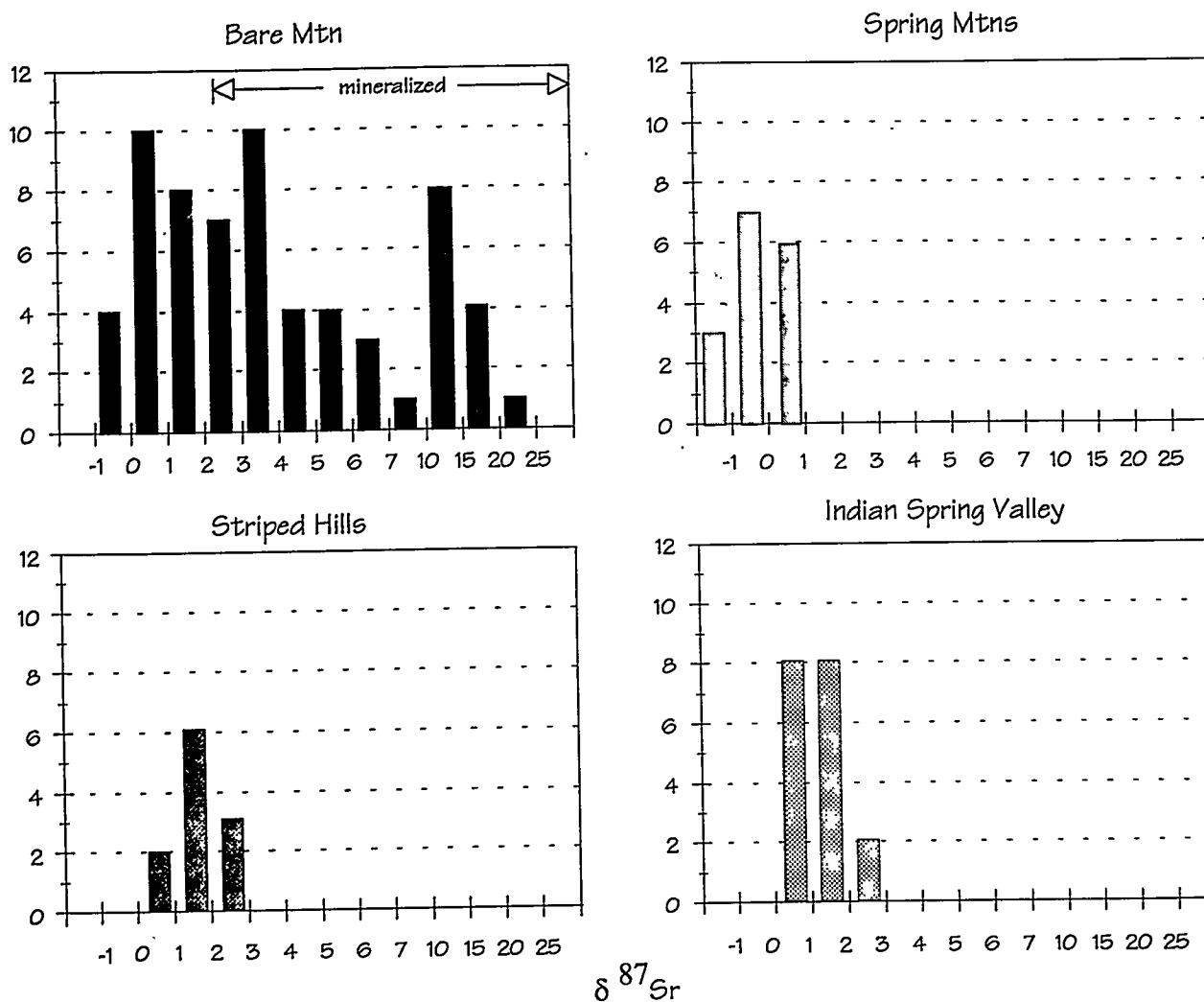


Figure 4 Histogram of  $\delta^{87}\text{Sr}_i$  values for mineralized and unmineralized Paleozoic carbonate rocks from Bare Mountain, Striped Hills, Spring Mountains, and ranges in the vicinity of Indian Springs Valley. Samples with  $\delta^{87}\text{Sr}_i$  values of 2.9 or less are unmineralized and those with values of 3.0 or larger are from mineralized areas.

tion (mainly argillite) occur upgradient to UE-25p#1.<sup>18</sup> Fourteen samples of the argillite from drill-hole UE-25a-3 have a mean  $\delta^{87}\text{Sr}$  of +9.8. Thus, the Eleana could contribute elevated  $\delta^{87}\text{Sr}$  values to ground water interacting with it. If it is assumed that the ground water interacted only with the Eleana and older limestones, its measured  $\delta^{87}\text{Sr}_i$  value of +3.6 could result from approximately one-third of the strontium in the groundwater being derived from the Eleana and two-thirds from the limestone units. Interaction with argillite of the Eleana Formation could also explain the rather large sodium content (150 mg/l) of the ground water.<sup>15</sup>

#### DISCUSSION AND CONCLUSIONS

Tertiary hydrothermal mineralization introduced strontium with large  $\delta^{87}\text{Sr}_i$  values into the host Paleozoic

and Late Proterozoic carbonate rocks. The source of this radiogenic strontium is the Precambrian basement, where the fluids resided prior to being thermally activated. Isotopic data and geologic observations regarding localization of mineralized zones indicate that faults were the pathways for the ascent of ore-forming fluids. Paleozoic marine carbonate rocks in areas that have not been subjected to mineralization retain  $\delta^{87}\text{Sr}_i$  values close to primary marine values. In contrast, mineralized carbonate rocks, as shown by samples from Bare Mountain, consistently have  $\delta^{87}\text{Sr}_i$  values of 3.0 or larger because of the addition of radiogenic strontium by hydrothermal solutions. Core samples of Silurian limestone units from the single penetration of Paleozoic basement beneath Yucca Mountain (drill hole UE-25p#1) have retained their primary  $\delta^{87}\text{Sr}_i$  values, and we conclude that the carbonate basement at this site has not been subjected to hydrothermal mineralizing solutions.

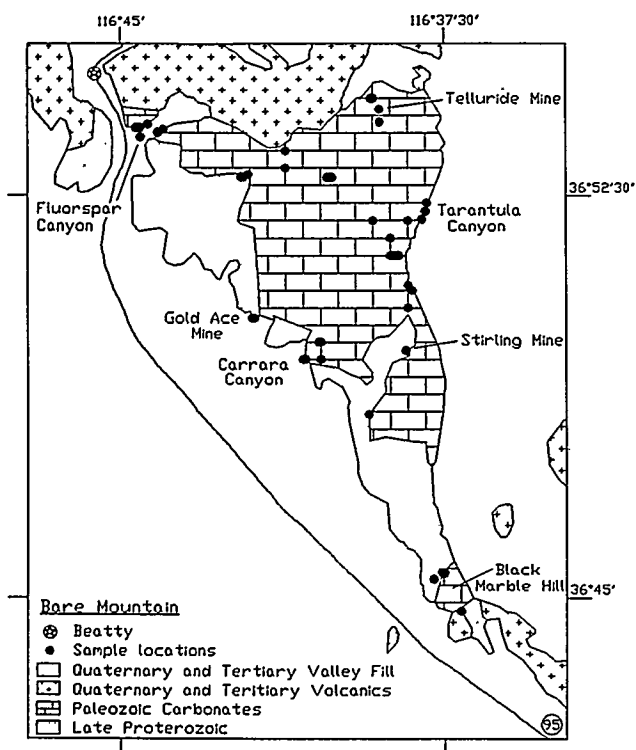


Figure 5 Geology of Bare Mountain showing sampled localities (dots).

The presence of  $^{87}\text{Sr}$ -enriched mineralized zones in carbonate rocks in the saturated zone could lead to the development of downgradient plumes of  $^{87}\text{Sr}$ -enriched ground water. Although ground water with elevated  $\delta^{87}\text{Sr}_i$  values could also result from interaction with Precambrian or younger clastic rocks (e.g. Eleana Argillite), the coupling of large  $\delta^{87}\text{Sr}_i$  values with elevated concentrations of pathfinder elements such as As, Tl, Sb, Ba, and Hg would be suggestive of a downgradient plume from a buried ore body.

Because of proximity, the observations relating mineralization and elevated  $\delta^{87}\text{Sr}_i$  values at Bare Mountain are assumed to be applicable to Yucca Mountain. Additional studies of other mineralized Paleozoic carbonate terrains will be necessary before the correlation can be extended beyond the Bare Mountain-Yucca Mountain area.

#### ACKNOWLEDGMENTS

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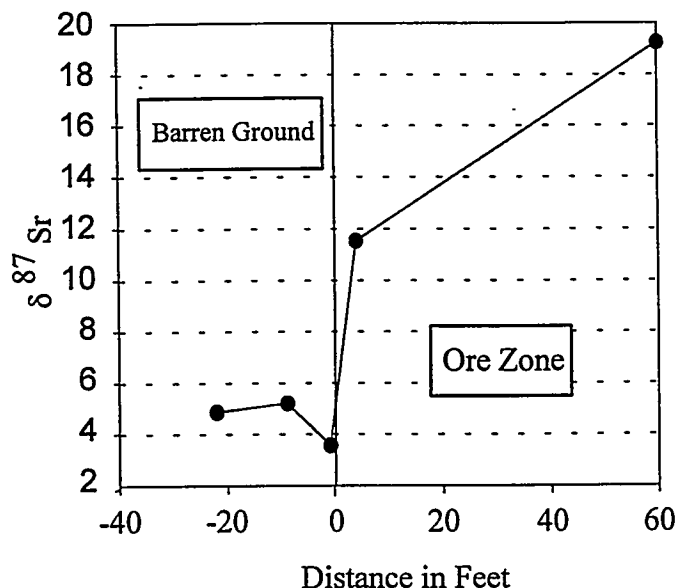


Figure 6 Variation of  $\delta^{87}\text{Sr}_i$  in Bonanza King Formation across the contact of an ore shoot in the Sterling Mine.

Hills. Richard Spengler (USGS) and Frances Singer (SAIC) provided critical assistance in logistics and sample collecting. James Cole (USGS) collected the samples from ranges in the vicinity of Indian Springs Valley. Bent Aquist and Greg Austin provided underground access at the Sterling Mine and allowed us to collect samples. Duane Craft and April Walker (both USGS) prepared the samples. Paul Sims and Frances Singer reviewed the manuscript. We are indebted to all of these people for their help and contributions.

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