Suitability of the Yucca Mountain Site to Accommodate a permanent Repository for High-Level Radioactive Waste and Spent Nuclear Fuel: an Independent Assessment

Part Two – Prognosis

Conceptual Model and Its Long-Term Implications

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Chapter 2.1. Stability of the Vadose Zone

2.1.1. Introduction

In addition to a small flux of infiltrating rainwater (downward), our conceptual model implies that the vadose zone is occasionally subjected to an upward flux of heated, gas-charged fluids. The horst-bounding faults, specifically the Paintbrush and Solitario Canyon faults, act as time-dependent, planar sources of heated, gaseous fluids. The upward flux may result from seismic pumping accompanying the characteristic earthquake (Richter magnitude ≥ 6.8). Seismically-induced flow would invigorate the co-existing Rayleigh-Bénard instabilities, otherwise dampened by the *in situ* stress field. Alternatively, the mature Rayleigh-Bénard instabilities could develop spontaneously (without the destabilizing influences of the characteristic earthquake).

The recurrence period for the characteristic earthquake at Yucca Mountain is about 10 Ka, so the seismic pumping period is correspondingly 10 Ka. However, for the mature Rayleigh-Bénard instabilities the return period could be significantly different. In general terms, one can infer that the distribution of the periods of mature instabilities is probably fractal, with long recurrence periods (say 10^5 or more years) for the large instabilities, but much shorter for the smaller ones.

Observations of the hydrologic effects of large earthquakes in the Great Basin, specifically the Hebgen Lake and Borah Peak events, suggest that large volumes of fluid (0.3-0.5 km^3) could be mobilized by seismic pumping (Hickman et al., 1994; Muir-Wood and King, 1994). Such volumes would be sufficient to partially inundate a vadose zone as thick as that at Yucca Mountain and even to discharge at the topographic surface. The inundation would be in the form of a "hydraulic mound", with the mound-forming fluid diffusing into permeable parts of the vadose zone, in the manner of a dampening plume as schematically shown on Figure 2-1.

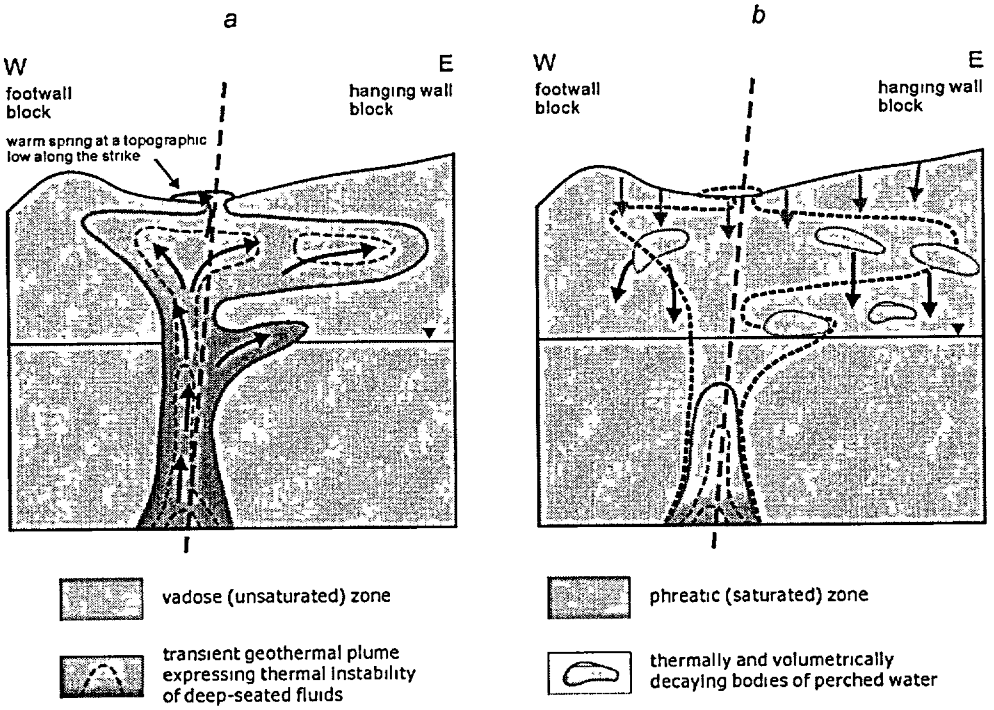


Figure 2-1. Schematic diagram illustrating changing hydrology of vadose zone at a time following the occurrence of characteristic earthquake.

a – Seismic pumping-induced and Rayleigh-Bernard instability assisted invasion of waters into the vadose zone along the fault-based conductivity channel. *b* – Decay of the mound, associated with formation of perched bodies of water, gravitation-driven filtration, and interaction with meteoric (rain) waters.

Characteristic features of the model: *1*. Upwellings of thermal fluids are restricted to deep-seated horst-bounding faults, such as Solitario Canyon and Paintbrush. *2*. Both upward and downward movements of geothermal fluids occur in the vadose zone. *3*. Asymmetry of the diffusing plume is a result of the near-surface enhancement of the conductivity structure, mostly affecting the hanging wall block. *4*. The local temperature gradient in the vadose zone expresses transient nature of the instability. *5*. At and around the fault-based conductivity channel cavities in the rock are completely lined with minerals, whereas away from this channel the mineral distribution in cavities may be an expression of the dissipation of perched bodies of water.

The formation and evolution of hydraulic mounds has been conceptually defined in relation to Rayleigh-Bénard convection. To test the conceptual model of hydraulic mounding, we now focus on the effects of mounding. Lateral gradients of temperature, mineral saturation, gas content, hydraulic head and other parameters would be set up in the vadose zone around a hydraulic mound. The potentials would be greatest within and adjacent to the fault-based conductivity channel. They would diminish as a function of the lateral distance away from this channel and disappear at a distance measured in terms of a few kilometers, or less. Diffusion is likely to be asymmetric. Two processes are relevant to the development of an asymmetry of mineralization. Our own investigations based on numerical modeling reveal that extensional faulting is accompanied by volumetric dilation of the footwall at depth and of the hanging wall at shallow levels. The horizontal stresses tend to be lower in the hanging wall. Therefore, more extensive fluid penetration into the hanging block is to be expected than into the footwall at shallow levels.

When the input of upwelling fluid ceases, the mound would breakdown into isolated and decaying bodies of perched water, as schematically shown on Figure 2-1. Over time, infiltrating rainwater would increasingly cool and dilute the perched fluids. The fluids would interact with the host rock. For these reasons, the temperature and composition of these perched fluids would vary spatially and temporally.

2.1.2. The fundamental discrepancy between the two conceptual models

By considering mass and energy transport as intrinsically non-linear, our conceptual model de facto implies episodic hydraulic mound formation and decay. The competing concept (U.S. DOE) treats the transport phenomena as being intrinsically steady and excludes any upward flux of energy or matter. Thus the DOE conceptual model precludes the establishment of hydraulic mounds induced by upwelling. The reliable resolution of this fundamental difference in the two conceptual models has a direct bearing on understanding the true balance between the attributes and the pitfalls of Yucca Mountain.

2.1.3. Resolution of the issue

The desired resolution would be greatly assisted if it were possible to identify a present-day analog of the hypothetical mound. In this regard, we wish to focus on a particularly interesting and potentially very important hydrologic feature, which has been recently discovered at the southern end of Yucca Mountain. This feature is located in the north central Amargosa Desert, approximately 20 km to the southwest of the proposed repository, as shown on Figure 2-2.

The hydrologic feature occurs beneath a series of Late Quaternary paleo-groundwater discharge sites, indicated by a spring-wetland (cienaga) environment, reconstructed by Forester et al. (1999) based on the fossil record (mollusks, ostracodes, diatoms). Some of the associated spring deposits were found by Paces et al. (1993) to carry ^{230}Th/U ages ranging from 45.7±4.9 Ka (2 samples), through 30±2.7 Ka (2 samples) to 17.4±2.9 Ka. It is thus evident that the discharge sites were repeatedly active in the late Quaternary. Included among these sites are the so-called Lathrop Wells Diatomite (LWD) and Crater Flat Wash (CFW) deposits, which occur at altitudes 803 and 797 m asl, respectively (Figure 2-2).

Figure 2-2. Vicinity of Yucca Mountain. Yellow circles identify locations of paleo-discharge sites Lathrop Wells Diatomite (LWD), Crater Flat Wash (CFW), and Crater Flat Deposits (CFD).

2.1.4. Configuration of the Potentiometric Surface

Before the Nye County Early Warning Drilling Project was implemented, it was generally believed that the water table was situated at a depth of about 100 m below the ground surface (an altitude of about 700 m above msl, as shown on Figure 1-43 in Part One). Water table fluctuations, which are expressed in the geologic record at this location, were and still are regarded as expressing past climatic changes (see for example Quade (1993), Paces et al. (1993), and Paces and Whelan (2001)). According to this viewpoint, fluctuations in the water table can be attributed to changes in the rates of meteoric precipitation and the resulting changes of aquifer recharge. Again we stress that this interpretation has been presumed *a priori* by other Yucca Mountain investigators. Although this interpretation is plausible, particularly in light of the limited database, it is not necessarily valid because other interpretations are possible as well.

The first opportunity to verify the presumption was recently provided by holes drilled under the auspices of the Nye County Early Warning Drilling Program (EWDP). The EWDP drill holes revealed that the actual values of the potentiometric heads are 786 m asl (water table at a depth of 17 m from the ground surface) in drill holes EWDP-1s and -1d) and 767 m asl (water table a depth of 30 m) in drill hole

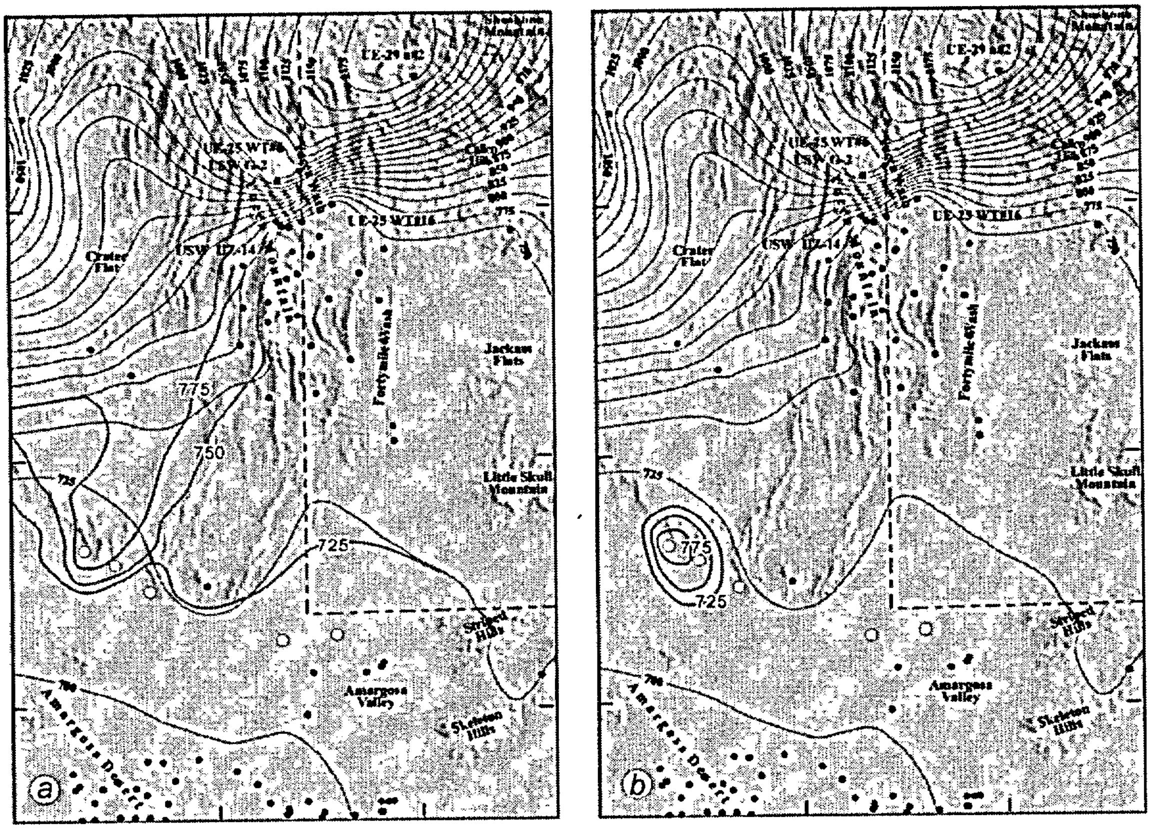


Figure 2-3. Two possible interpretations of the potentiometric surface based on the results of the Nye County Early Warning Drilling Program (NC EWDP) obtained in 1999. Gray isolines represent piezometric contours, as reported in the Yucca Mountain Viability Assessment report (U.D. DOE, 1998). Yellow circles show locations of the NC EWDP boreholes.

a – Revised configuration of potentiometric surface as constructed by Paces and Whelan (2001); *b* – Configuration, assuming that potentiometric anomaly expresses an "upward flux" state of the boundary conditions, at the base of the hydrologic system.

EWDP-9s. These values are 50 m to 100 m higher than those, which were expected by the U.S. DOE Yucca Mountain Project hydrologists, based on the earlier estimates as shown on Figure 1-43 (Part One).

There are two ways to reconcile the potentiometric "anomaly" with the rest of the potentiometric data. Both possibilities are illustrated on Figure 2-3.

Figure 2-3-*a* shows a potentiometric map based on the map originally constructed by Paces and Whelan (2001). The map implies a belief (unstated and unsubstantiated) that the saturated alluvium/colluvium in Crater Flat has (spatially) very variable hydraulic conductivity. Such a substantial variability could explain the major differences of hydraulic gradient along the flow path, both upstream and downstream of the potentiometric anomaly.

An alternate and equally admissible hypothesis is expressed by Figure 2-3-*b*. In drawing the contours shown in Figure 2-3-*b*, it has been assumed that the potential high expresses an upward flux of fluid from below (regarded as the (no-flow) base of the hydrologic system by the U.S. DOE investigators).

The two competing hypotheses have sharply differing conceptual implications. Resolution of the issue may be achieved by considering the *in situ* geothermal gradients along the flow path.

2.1.5. Geothermal gradients

Paces and Whelan (2001) noted that the temperature of the water in well EWDP-1s ranges between 27°C and 30°C. This temperature is substantially higher (ΔT = 9-12°C) than the mean annual surface temperature of 18°C (data recorded at the Gate 510 weather station which is located nearby in the southwestern part of the Nevada Test Site). The data imply an average value of the geothermal gradient from the ground surface to the water table as high as $dT/dz \approx$ 70°C/100m.

The Nye County EWDP webpage (www.nyecounty.com) lists values of the temperatures encountered in a number of the EWDP drill holes, at a varying but shallow depth (<80 m). These temperatures vary spatially between 35.3°C at a depth of 46.2 m in well EWDP-19 to 53.4°C at a depth of 12.5 m in well EWDP-12pa. Thus, the Nye County database confirms two critical features of the geothermal gradient (between 36°C/100m and 136°C/100m of depth). First, very high values can occur just below the ground surface. Second, there can exist very substantial lateral variation.

By contrast, the *in situ* temperature is only about 27°C at the water table in wells USW VH-1 and VH-2 (Sass et al., 1987), which are "up-gradient" of the EWDP wells. According to Robinson (1984), the water table is at a depth of 184.2 m in USW VH-1 and at 164 m in USW VH-2. Assuming 15°C for the mean ambient surface temperature yields, for the up-gradient portion of the flow path, an average value of the geothermal gradient of dT/dz = 6.5-7.3°C/100m. The sharp inconsistency between the average values of the shallow dT/dz along the flow path is illustrated in Figure 2-4.

Figure 2-4 shows that an important geothermal anomaly underlies the LWD and CFW discharge sites (approximately corresponding to the EWDP-9s borehole), in addition to these sites being underlain by the potentiometric anomaly. Here, the total (i.e., conductive and advective) flux of heat into the atmosphere is a factor of about ten greater than that in the up-gradient and down-gradient portions of the flow path. This can only be explained by a flux of energy from below. It cannot be explained by heterogeneity of hydraulic conductivity.

The conclusion that a localized upward flux of energy and matter (fluid) is occurring also indicates that the potentiometric map, constructed by the USGS scientists (Figure 2-3-*a*), is grossly misleading. This map, and the implicit assumption of an underlying no-flow boundary, has led Paces and Whelan (2001) to

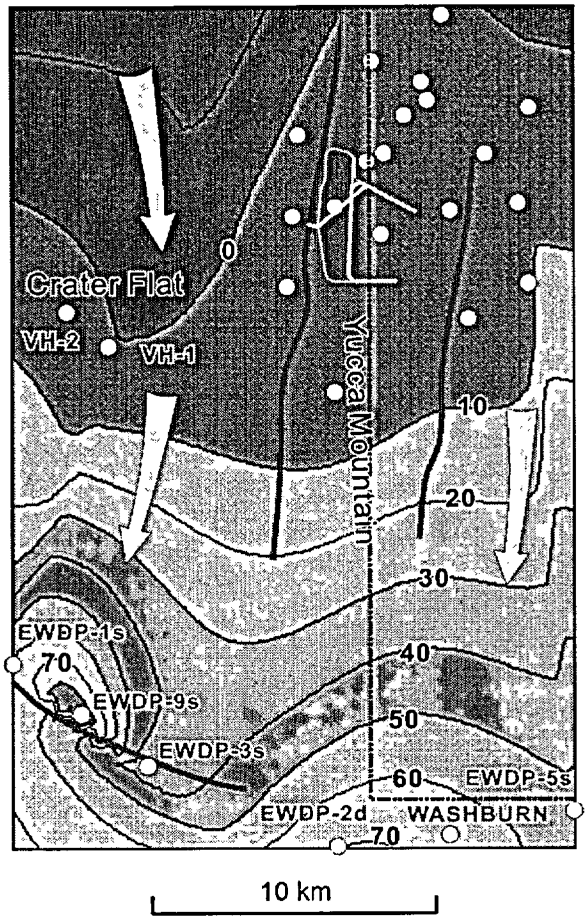


Figure 2-4. Values of geothermal gradient *dT/dz* (in °C per 100 m) calculated from the data of the *in-situ* temperature measurements in boreholes. Also shown are the footprint of the proposed repository and major faults. Arrows indicate general direction of regional water flow.

The map was constructed using Mathcad 6 PLUS software. Kinks on the right side of the map, as well as large area with negative values of dT/dz in the upper left corner are artifacts related to the absence of the data for these areas. Also, since no borehole data exist for the large area between Yucca Mountain and central Crater Flat, configuration of isograds in this area is also uncertain.

Borehole temperature data are from Sass et al. (1987) except for the Nye County EWDP and WAHSBURN boreholes. Data for these boreholes were obtained from the Nye County Nuclear Waste Repository Project Office website at www.nyecounty.com.

additional claims. Specifically, these authors stated, "*These data limit Pleistocene water-table fluctuations at the specified Amargosa Desert sites to between 17 and 30 m and eliminate the need to invoke large water-table fluctuations*".

The conclusion that the LWD and CFW discharge sites, and underlying potentiometric and geothermal anomaly, result from an upward flux of fluid is incontrovertible. The common occurrence of palaeo-springs is expressed as 2-3 m thick layers of diatomaceous earth, which forms at 20-30°C. These infer that the intensity and/or distribution of the upward flux has changed, or it has been changing, over the past 40 Ka However, the process responsible for the changes at this location remains to be identified. There are only two possibilities. These seem equally plausible without further considerations. The **first** possibility is that changes of the rate of aquifer recharge, induced by climatic change, control the rate of discharge. The **second** possibility is that tectonically-induced alterations of hydraulic conductivity and the (non-equilibrium) state of the underlying Rayleigh-Bénard instability control the discharge rate.

Because the respective conceptual implications are so profoundly different, it is essential to establish which one of the two possibilities is supported by the known facts. Two sets of pertinent facts are already available. One concerns the tectonically induced fluctuations of the piezometric head. The other concerns the stable and radiogenic isotope signatures of the present-day water and of the late Quaternary deposits from the LWD and CFW discharge sites.

2.1.6. Fluctuations of the Piezometric Head

2.1.6.1. Introduction

Attempting to describe hydraulic gradients in fractured rock by a single piezometric surface can only be, at best, an approximation. Pressure differences between individual fractures or clusters of fractures are characteristic of fractured aquifers and must be expected even in hydrologic subsystems, which are far closer to equilibrium (thermodynamic) than the Yucca Mountain geological system. It is emphasized, however, that while the dangers of assuming a linear pressure distribution with depth throughout the fractured rock section (i.e., variability of pressure in space) should not be ignored, here we focus on the implications of time-variability of the piezometric head.

2.1.6.2. Availability of piezometric head monitoring data

The USGS installed piezometric head monitoring systems in a number of the wells drilled in the interior of Yucca Mountain. Regrettably, we understand that no data are available either because all the monitoring systems failed or those data, which were collected, were lost or regarded as unreliable.

Records of the variation of the piezometric head in a handful of wells became available as a result of the Nye County Early Warning Drilling Program (EWDP). The EWDP wells, fitted with automatic monitoring systems, were generally completed at two depths. Two wells show substantial fluctuation of piezometric elevation: wells EWDP-1s and EWDP-3s. The fluctuations recorded by well EWPD-3s are "stepped" in nature and therefore it is suspected that they may be unreliable. They are not considered further.

A more or less continuous record of piezometric head variation in Well EWPD-1s is available on the Nye County website (www.nyecounty.com) for the period from May 1999 to April 2001. The well is sited on palaeospring deposits near the boundary between Crater Flat and Amargosa desert. It is therefore located down-gradient of Yucca Mountain. Well EWPD-1s penetrates a locally important active fault, the so-called Stewart fault, which is a part of the Walker Lane fault zone, the extensive structural feature shown on Figures 1-2 and 1-35 in Part One.

EWPD-1s was completed in early February 1999, with piezometers located in screened sections from 160-180 ft (probe #1 in the Tertiary tuffs) and 210-270 ft (probe #2 in the underlying Tertiary sedimentary rocks). The two sand sections in which the piezometers are placed are separated by only 14.4 ft of bentonite grout. No nearby drilling operations were conducted after completion during 1999.

The nearest wells drilled and completed after EWPD-1s were Wells EWDP-12PA and EWDP-12PB, half a mile south of EWPD-1s. These wells were drilled and completed between March 21st and April 4th, 2000. Well EWDP-12PC, drilled during the same period, was not completed until November 2000. (The next nearest well, EWPD-3DB, approximately 3.4 mi southeast of well EWPD-1s, was drilled (to 505 ft) and completed with conductor casing in December 2000.) Well locations and drilling and completion details can be accessed on the website.

2.1.6.3. Expectations

Before examining the piezometer records in EWDP-1s, it is helpful to review the expectations derived from our conceptual model and other observations. The model implies that reversible and irreversible changes in the aperture of fractures will occur, changing the storativity of these fractures. The changes may be induced, or they may be spontaneous. In a fractured rock mass, pressures are typically non-hydrostatic implying lack of, or restricted, hydraulic communication between separate clusters of fractures. Consequently, changes of hydraulic conductivity can also result in pressure change as monitored in a well.

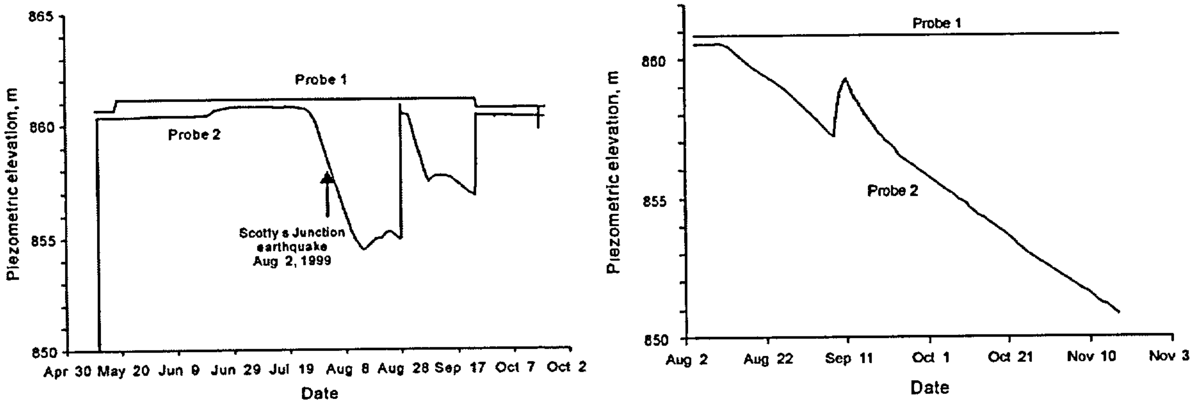
The higher the geological strain rate, the more favorable are the conditions to abrupt fluctuation of the piezometric head in the well. The back and forth transfers would be isothermal in the case of a wellbore in thermal equilibrium with the adjacent fractures. Piezometric level "spikes" may occur, expressing an interplay between the in situ stress and the pore pressure.

Such fluctuations will be observed only if the monitoring well is hydraulically connected to a “sensitive” location in the rock mass. Reversible changes of piezometric and potentiometric heads, which are sometimes observed in some areas before or after the occurrence of large earthquakes, suggest that the conductivity (and in some cases also the storage to a significant degree) of a minor proportion of the fracture population is particularly sensitive to changes of stress. One possible explanation is that small increases of the total stress, or increases of the pore pressure, open fractures by hydraulic re-opening of the cohesionless, previously fractured rock mass, at a singular point in the stress field (zero total stress implying pore pressure approximately zero). A singular point might arise in dilating low permeability rock where fractures have not formed and a hydraulically-isolated body of pore fluid (largely incompressible unless gassy) is subject to dilation and unable to draw in fluid through the intact rock at a rate faster than the dilation. Subsequent abrupt increase of the normal stress in the rock mass at that point, arising from migration or loss of the singular point, could create conditions favorable to hydraulic fracturing and consequent pressure dissipation. Buckling of fractured rock subject to sufficient stress difference is one process, which could lead to the dilation described above.

Subsequent sealing of the fracture by some mechanism such as mineralization may be a prerequisite for any further pressure fluctuations to occur by this mechanism at the same location in the rock mass as a previous fluctuation. Closed fractures without a sealing material would normally have a residual aperture available to permit pressure diffusion.

The expectation that certain wells are particularly sensitive to changes in a geological system is supported by observations elsewhere. Earthquake seismologists have striven for many years to identify earthquake precursors. One such potential precursor is a water level change. King et al. (2000) scrutinized the relation between well water level changes and distant earthquakes. They concluded that one well of the group studied, Well SN-3, was particularly sensitive to some remote earthquakes. They concluded that the most notable relationship was a water level drop of about 2 m during the 6 months before a M5.8 earthquake having an epicenter 50 km from the recording well. "*The high sensitivity of SN3 is probably due to its tapping a near-critical hydrologic system, namely, a permeable aquifer (weathered granite) connected to a permeable fracture zone on the higher-pressure side of the Tsukiyoshi fault, which has a weak, pliable and normally impermeable fault gouge. This near-critical condition allows SN3 to sense water leaks through the fissures, easily created and healed, in the gouge zone over a broad area of the fault plane. A near-critical condition such as this may possibly be a common characteristic of all earthquake-sensitive sites.*" This description is conceptually consistent with our model of fracture opening closing and changes of storativity.

The King et al. (2000) study demonstrates that some completions may be more sensitive than others. The study also shows that water level changes at one site may occur well before the occurrence of an earthquake at a remote site.



a. *b.*

Figure 2-5. *a* - Response of the piezometric elevation in the Nye County borehole NC-EWDP-1s to the Scotty's Junction, Nevada earthquake of August 2, 1999. *b* - Change in piezometric elevation in the Nye County borehole NC-EWPD-1s between August 2 and November 30, 2000.

Plotted using the data obtained from the Nye County Nuclear Waste Repository Project Office website at www.nyecounty.com.

2.1.6.4. The EWDP-1s piezometric head record

Little change of piezometric elevation was recorded in Probe #1 of well EWPD-1s. The general characteristics of the fluctuations recorded by Probe #2 may be summarized as follows:

1) Steady, with the exception of a rise of about 1.5 ft during a short period, from April 1999 to July 1999.
2) A largely linear decline of about 19 ft from July 1999 to August 1999.
3) Recovery, with both abrupt and progressive fluctuations, between July 1999 and September 1999.
4) Essentially constant from September 1999 to August 2000, with the exception of a rise of about 1ft in early November 1999.
5) A fall of almost 50 ft. from August 2000 to January 2001, including one transient period of recovery amounting to about 7 ft during an approximately 15-day period.
6) Irregular recovery to the end of the record in April 2001, at which time the piezometric elevation was about 30 ft below the initial level.

Some of this record is recorded in Figure 2-5.

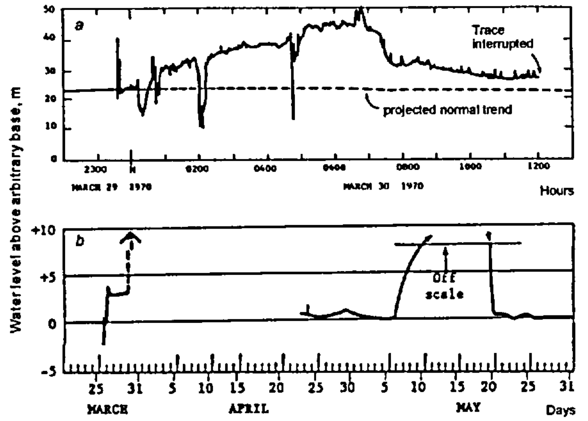


Figure 2-6. Hydrologic response of well UE-20p in Pahute Mesa, Nevada Test Site to the Handley nuclear detonation (March 26, 1970). From Dudley et al. (1971).

The monitoring period includes the August 2nd, 1999, Scotty's Junction earthquake. The epicenter of the Scotty's Junction earthquake is located within the Walker Lane fault system, approximately 60 km to the northwest from the water level monitoring site. The event had a Richter magnitude of about 5.5. The Probe #2 record implies that the Scotty's Junction earthquake significantly influenced water levels in the well, as shown on Figure 2-5 and as outlined above. Piezometric head changes occurred before the earthquake as well as afterwards. Temperature changes were insignificant.

2.1.6.5. Comments on the fluctuations of piezometric head in well EWDP-1s

It thus appears that the observed fluctuations of piezometric head underneath the LWD and CFW discharge sites are consistent with the proposed conceptual model and the conclusions of King et al. (2000). The observed fluctuations are remarkably consistent with those, which the model implies would be associated with a receding hydraulic mound, at early stages of tectonic cycles.

These fluctuations, the "anomalous" configuration of the potentiometric surface, and the observed heat flow all are elements which, according to the conceptual model, were present at Yucca Mountain in the past and which will be present in the future.

2.1.6.6. A second example of pore pressure fluctuation at the Nevada test site

An excellent example of pore pressure-rock stress interplay is shown on Figure 2-6. Figure shows water level fluctuations observed at the Nevada Test Site after the Handley nuclear detonation. The nuclear device was detonated on March 26, 1970 and had a reported yield of about 1 megaton, which is equivalent to an earthquake with a Richter magnitude of about 6.5. The water level monitoring well (UE-20p) was located at a distance of about 5 km from ground zero, so that the induced permanent strain (by growth of the detonation cavity) was small at the monitoring well location (of the order of 10^{-4}, or less). In spite of this small strain, the detonation induced significant fluctuations of the water levels, which must have been supported by very large volumes of groundwater. These fluctuations lasted for a period of at least two months, as shown on Figure 2-6.

2.1.6.7. The self-organized critical state of the Yucca Mountain geological system

An alternative perspective of the state of the Yucca Mountain rock mass, entirely consistent with the conceptual model, is provided by the concept of self-organized criticality (Bak and Tang, 1989; Bak et al., 1988). Bak and his co-authors investigated the dynamics of diffusion in non-linear systems using simple cellular automata. They showed that such a system will become stable (i.e., stationarity will obtain) precisely at the point when the network of minimally stable clusters has broken down to the level where a signal cannot be communicated across infinite distances (i.e. just below the percolation threshold or just before a globally minimally stable state is reached). At this point there is no length scale and no time scale and perturbations result in energy dissipation at all length scales. Once the system reaches this self-organized critical state, the system stays there.

The precise laws governing the interactions within a critically self-organized system are unimportant and a wide variety of non-linear dynamical systems have been found to exhibit characteristics of a self-organized critical state. Given that communication can occur at all length scales, in some cases small changes can be transmitted long distances.

The nature (not existence) of the critical state is largely defined by the properties of the boundaries. Therefore, changes at the boundaries change the nature of the physical state, which must evolve to maintain the state of self-organized criticality. Therefore, diffusive transfer of energy and matter (which are more readily apparent in some locations within the dynamical system than others) can reflect the evolving nature of the system boundaries. At times of greatest or most rapid change of boundary conditions, the greatest diffusive activity may be evident, as after the Handley event.

2.1.6.8. Implications of the fluctuations

The reversible, large fluctuations of piezometric and potentiometric levels, such as those that have been recorded in the UE-20p and NC EWDP-1s monitoring wells, are conceptually very important. They imply that, in some parts of the hydrologic subsystem of Yucca Mountain, the *in situ* stress field is more strongly heterogeneous and evolving faster than in other parts. In such parts, some of the fractures are dilated (i.e., open and opening), in response to the ongoing tectonic straining. The fractures apertures (a_t) can be described by the sum of the *in situ* stress induced component (a_s) and the residual component (a_r), where $a_t = a_s + a_r$. These apertures, and hydraulic storativity and conductivity, which are exponential functions of the aperture, are reversible in the long-term in that a_s can be lost by stress change and a_r by mineralization or sealing by gouge.

The conceptual model infers that both the spatial extent of dilated fractures and the degree of their dilation do not remain constant. Instead, they increase incessantly as the date of the characteristic

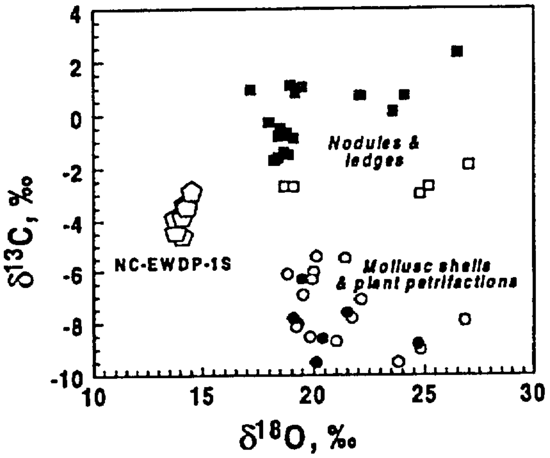


Figure 2-7. Isotopic composition of calcite from LWD deposits (small symbols; late Pleistocene, open circles and squares; middle Pleistocene, filled circles and squares, data from Paces et al., 1996) and calcite in equilibrium with water from EWDP-1s well at 25°C (large symbols). As presented in Paces and Whelan (2001).

earthquake approaches. At a time of the eventual occurrence of this earthquake, the degree of dilation around the ruptured fault will diminish. The corresponding piezometric and potentiometric levels will regain, at that time, their potential to rise, in accordance with the changing boundary conditions, and in this manner form a transient hydraulic mound.

Is it possible that the LWD and CFW discharge sites express the dawn of such a mound, but the present-day state of the underlying hydrologic system expresses early stages of its recession? The inferred *in situ* stress sensitivity and the conceptual model together imply that such may be the case. It follows that it is possible that the time-dependence of the upward flux at the LWD and CFW sites is controlled by the tectonics. This control can take the form of reversible alterations of the hydraulic conductivity structure (which are brought about by tectonically induced alterations of total stress at "sensitive" positive isotropic points or at "singular" points), of reversible alterations of the intensity of upward fluid and heat flux (induced by alterations of R_c-R_a), or both.

2.1.6.9. Concluding comments

Although not uniquely valid, the preceding hypothesis cannot be rejected. Detractors can always claim instrument malfunction as a basis to dismiss the EWDP-1s piezometric record. We emphasize that instrumentation faults have not been demonstrated or suggested by the Nye County workers. Given this, one must conclude that the "upwelling" conceptual model, involving hydraulic mounding, is at least as valid as the competing climate-control hypothesis. Regardless of which model individual readers might presently regard as having the greater scientific credibility, we must not forget that the competing hypotheses entail sharply contrasting consequences.

Given these piezometric head data, we pose one closing question. How is it possible to unreservedly accept the following conclusion of Paces and Whelan (2001): "*These data limit Pleistocene water-table fluctuations at the specified Amargosa Desert sites to between 17 and 30 m and eliminate the need to invoke large water-table fluctuations*"?

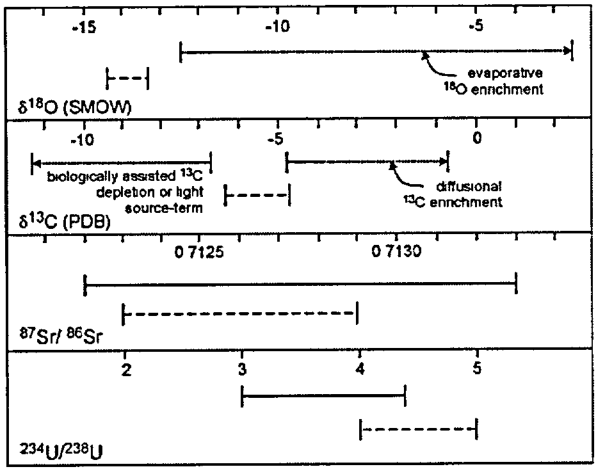


Figure 2-8. Variability of isotope compositions of fluids as a crude function of intensity of the upward fluid-heat flux. Based on data from Paces and Whelan (2001).

Solid lines – isotope properties of paleo fluids responsible for deposition of Pliocene carbonates at LWD site (intensity of upward fluid-heat flux is high); dashed lines – isotope properties of modern waters from EWPD 1s and 1d wells (intensity of upward fluid-heat flux is lower); and dotted arrows – alteration trends of a fluid while on the Earth's surface.

The $\delta^{13}C$ of paleo discharge fluids was estimated from $\delta^{13}C$ of carbonates by using fractionation factor of –1.75 ‰. The $\delta^{18}O$ of paleo fluids was estimated from $\delta^{18}O$ if carbonates using fractionation factor of 29.5 ‰ (assuming $T = 20°C$).

2.1.7. Isotope signatures

Paces and Whelan (2001) recently published data regarding the properties of stable ($\delta^{13}C$ and $\delta^{18}O$) and radiogenic ($^{87}Sr/^{86}Sr$ and $^{234}U/^{238}U$) isotopes in water sampled from wells EWDP-1s and EWDP-1d and for the spring deposits at the LWD site (see Figures 2-2 and 2-4). Together, these signatures can be used to infer their changes as a crude function of the changing upward flux at this site. The inferred changes could then be used to further enquire into the processes that control the changing intensity of this flux.

Water sampled from wells EWPD-1s and EWDP-9s carry values of $\delta^{18}O_{water}$ between -13.2 and -14.0 ‰ SMOW. Paces and Whelan (2001) also reported calculated values of $\delta^{13}C$ in a hypothetical calcite deposited in equilibrium with water from the EWPD-1s well (-3.0 to –4.5 ‰ PDB; Figure 2-7) from which the $\delta^{13}C_{water}$ can be assessed to range from –4.8 to –6.3 ‰ PDB (fractionation coefficient $\Delta^{13}C_{calcite\text{-}water} = -1.75$ ‰; Hoefs, 1987).

The values of $\delta^{18}O$ and $\delta^{13}C$ in Pliocene carbonate LWP deposits reported by Paces and Whelan (2001) can be used to evaluate the isotopic properties of the fluids that deposited these carbonates. Assuming equilibrium fractionation at $T = 20°C$ ($\Delta^{13}C_{calcite\text{-}water} = -1.75$ ‰ and $\Delta^{18}O_{calcite\text{-}water} = 29.5$ ‰), the $\delta^{18}O$ ranged from –12.5 to –2.5 ‰ SMOW. The $\delta^{13}C$ of carbonates form two distinct groups, possibly related to the different deposition processes. Carbonates with apparent links to the biologically assisted deposition (mollusc shells and plant petrifications) carry $\delta^{13}C$ substantially "lighter" than carbonates that do not have such affinity (see Figure 2-7). It is also possible that the "lightness" of $\delta^{13}C$ in some carbonates, at least in part, might be related to the light "source-term" CO_2 of the deep-seated water migrating with the ascending fluids. Correspondingly, the values of $\delta^{13}C$ of palaeowaters split in two groups: -0.8 to -5.0 and –7.8 and –11.3 ‰ PDB (Figure 2-8).

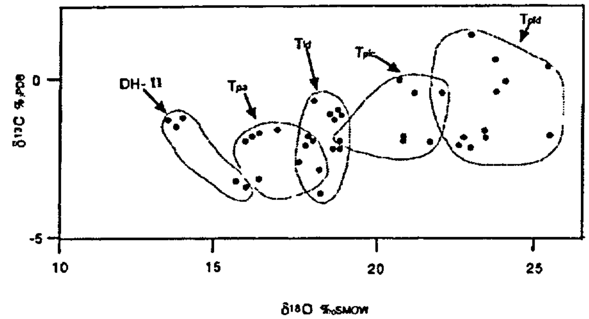


Figure 2-9. Isotopic character of carbon and oxygen incorporated in *per ascensum* carbonate deposits of the Amargosa Basin. Note that, as proximal facies (DH-II and Tpa) grade into distal facies (Tplc and Tpld) $\delta^{13}C$ and $\delta^{18}O$ values concurrently shift toward "heavier" region. Data from Hay et al. (1986) and Coplen et al. (1994).

DH-II – Devil's Hole crystalline calcite; Tpa – soft chalky limestone; Tld – dense nodular and fenestral limestone; Tplc – calcite disseminated in claystones; and Tpld – dolomite disseminated in claystones.

As soon as fluid saturated with CO_2 and $CaCO_3$ discharges at the ground surface, it begins to evolve. Specifically, the trend of enrichment in both $\delta^{18}O$ and $\delta^{13}C$ in waters and carbonate deposits of thermal springs, in response to outgassing and evaporation, is well documented (e.g., Gonfiantini et al., 1968, Chafetz and Lawrence, 1994). It is conceivable that an exceptionally dry and hot climate could produce similar evaporation-related isotopic enrichments in sienaga environments.

In the area of our study, the same trends of enrichment were documented in association with spring and marsh deposits in the Amargosa basin, some 15 km to the southeast of the LWD and CFW discharge sites. It may be observed in Figure 2-9 that, as the proximal lithofacies grade to become distal, the $\delta^{13}C$ value increases from about -3.0 to +2.0 ‰ PDB and the $\delta^{18}O$ value concurrently increases from about 15.5 to a maximum of 25.5 ‰ SMOW.

The data presented in Figure 2-8 suggest that the fluids from which carbonate deposits at the LWD site precipitated carried values of $\delta^{18}O$ somewhat "heavier" than that in modern waters. One question that needs to be addressed is: can this enrichment be expressing climate-driven fluctuations in the isotope properties of the discharging waters? Figure 2-10 shows the 500,000-year stable isotope record of the local climatic fluctuations, which has been reconstructed by Coplen et al. (1994) from Devil's Hole calcite deposits.

Figure 2-10 shows that the $\delta^{18}O$ and $\delta^{13}C$ time series induced by climatic changes are inversely correlated with each other. The figure shows that the $\delta^{18}O$ the pluvial (glacial) maxima are periods when $\delta^{18}O$ tend to have the lowest values. This trend is directly opposite to that which has been inferred for the LWD discharge site (see Figure 2-8). Here, the $\delta^{18}O$ is *higher* during the carbonate deposition period, which, if the system were driven by climatic fluctuation, would correspond to the timing of the most intense recharge of the aquifer.

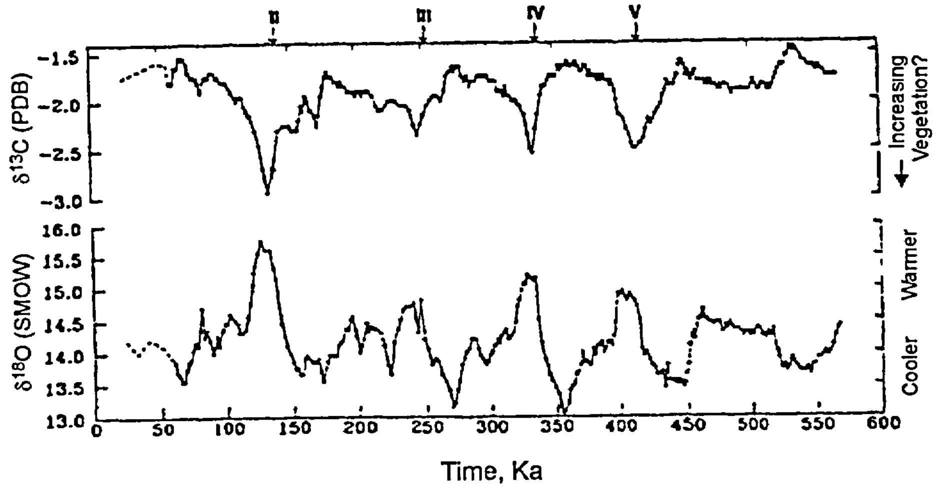


Figure 2-10. Variations in $\delta^{13}C$ and $\delta^{18}O$ in calcite deposits from Devil's Hole, Nevada (core DH-11) during middle and late Pleistocene. Roman numerals denote approximate timing of glacial terminations (glacial-interglacial transitions). Reproduced from Coplen et al. (1994).

This discrepancy contradicts the notion of changing climate being the factor controlling the flux of fluid at this site. In conjunction with an extreme variability of $\delta^{18}O$ values (10 ‰), which is quite atypical for low-temperature spring deposits, and the thermal character of waters forming the potentiometric anomaly underlying the LWD and CFW paleo discharge sites (see Figure 2-4), makes the proposal that climate controls the discharge at these sites untenable.

The temporal changes of the stable isotope signatures at the LWD and CFW discharge sites seem to have been accompanied by changes of the radiogenic isotope signatures, specifically the $^{87}Sr/^{86}Sr$ and $^{234}U/^{238}U$ ratios. Because Sr and U isotopes do not fractionate during ordinary chemical reactions, the $^{87}Sr/^{86}Sr$ and $^{234}U/^{238}U$ ratios from the discharge sites shown in Figure 2-8 can be regarded as a direct expression of the values of these ratios in the fluids at a time when the intensity of the upward flux was high. It thus appears that, as the intensity of the upward flux has been diminishing, the $^{87}Sr/^{86}Sr$ values have declined from a maximum between 0.7121 and 0.7133 (Pleistocene or late Quaternary waters) to the present-day minimum between 0.7123 and 0.7129. Concurrently, the $^{234}U/^{238}U$ values have increased from the Pliocene minimum between 3.0 and 4.4 to the present-day maximum of between 4.0 and 5.0, as shown on Figure 2-8.

The inferred temporal changes of the radiogenic isotope signatures are important because these changes may readily be rationalized in the light of the proposed conceptual model. The declining $^{87}Sr/^{86}Sr$ values indicate the diminishing Sr derivation from a reservoir, which is enriched in ^{87}Rb (parent for ^{87}Sr) and, at the same time, allows for prolonged accumulation of ^{87}Sr, via $^{87}Rb \rightarrow {}^{87}Sr$ β-decay. There is only one such primary reservoir in this area. It corresponds to the clastic complex (pre-Cambrian) of the basement, underneath the marine carbonates of an early Paleozoic age. Thus, the decline of $^{87}Sr/^{86}Sr$ values may be taken as expressing the diminishing depth of fluid circulation which would be expected to accompany the mound recession.

On the other hand, the increasing $^{234}U/^{238}U$ values indicate the increasing derivation of ^{234}U atoms from the α-decay damaged sites formerly occupied by the parent ^{238}U atoms. This increase is a clear sign of the opening of fractures in association with the increasingly non-equilibrium thermodynamic state. This allows fluids to enter the previously closed fractures and to remove (dissolve) the previously-formed "hot" ^{234}U atoms. In addition, the increasing fluid/rock ratio increases the efficacy of direct rock → fluid transfer of ^{234}U atoms, via the α-recoil mechanism. Thus, the increase of $^{234}U/^{238}U$ values may be taken as expressing the increasing dilation of shallow fractures, which would also be expected to accompany the mound recession.

2.1.8. Isotope Comparative Analyses

The hydrological mound discovered at the Stuart fault under the LWD and CFW paleo discharge sites may represent a modern analog of paleo mounds associated with other deep-seated faults at Yucca Mountain. One of the best-studied sites that may host traces of such activity is the Bow Ridge fault at Exile Hill on the east slope of Yucca Mountain. At early stages of the Yucca Mountain characterization, in 1980, a vein composed of micritic calcite and opal, as well as associated slope-parallel calcite-silica deposits was intersected by Trench 14.

The veins have been alternatively interpreted as pedogenic deposits (e.g., Quade and Cerling, 1990 and Stuckless, 1990) or as possible footprints of the ascent(s) of deep-seated fluids (Szymanski, 1992). The conclusions presented in the two former publications are based on the comparative analyses of isotopic properties of calcite from Trench 14 and other deposits with whose origin has been presumed to be known.

Initially, Quade and Cerling (1990) compared the Trench 14 calcite with modern pedogenic carbonate as part of the effort supported by the Nuclear Waste Projects Office of the State of Nevada. The following statement from Quade and Cerling (1990) summarizes their conclusions. "*Comparison of the stable carbon and oxygen isotopic compositions of the fracture carbonates with those of modern soil carbonates in the area shows that the fracture carbonates are pedogenic in origin and that they likely formed in the presence of vegetation and rainfall typical of a glacial climate.*" Later, the National Research Council of the National Academy of Sciences endorsed this conclusion (NAS/NRC, 1992). The subject comparison is reproduced on Figure 2-11-*a*.

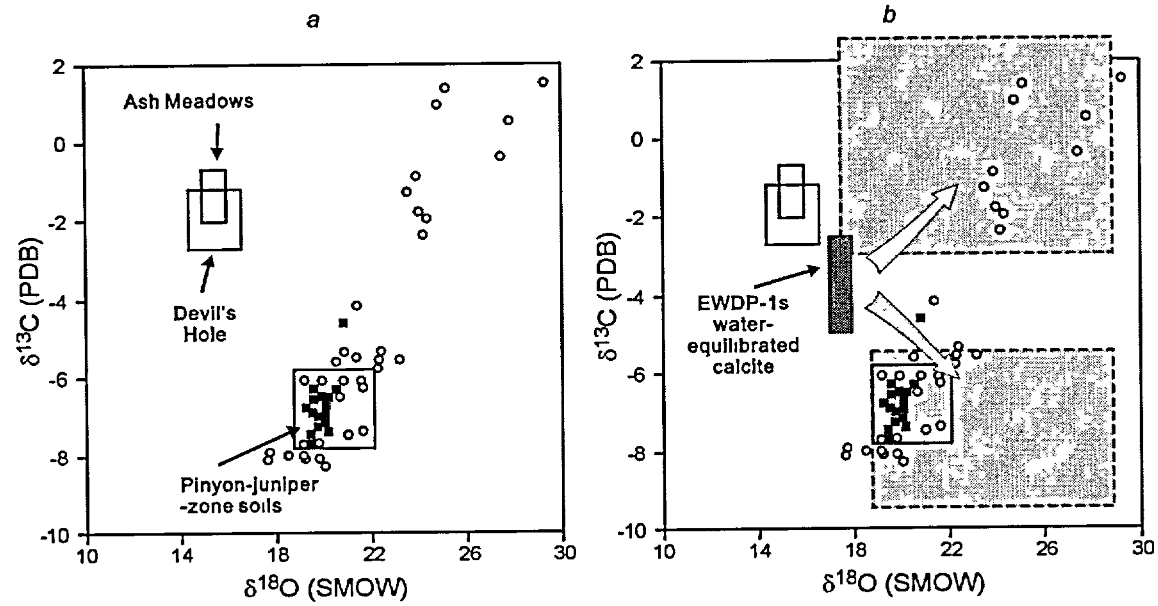


Figure 2-11. Comparison of isotopic properties ($\delta^{18}O$ and $\delta^{13}C$) of carbonates from the Yucca Mountain area.

a – Carbonates from Holocene soils (*open circles*), Trench 14 (*filled squares*) and springs at Devil's Hole and Ash Meadows. Reproduced from Quade and Cerling (1990). Original caption emphasizes that "*21 out of 22 analyses from Trench 14 fall within the isotopic field for Holocene-age soil carbonate found in the pinyon-juniper-sagebrush zone*", and that spring carbonates "*plot completely outside the observed range for carbonates in modern soils and Trench 14*". *b* – Hypothetical calcite formed in equilibrium with modern water from the Nye County EWDP-1s well and fields (*gray*) of isotopic properties of carbonates from Pleistocene spring discharge site LWD (data from Paces and Whelan, 2001). Note that EWDP-1s well was drilled at the LWD site (see Figures 2-2 and 2-4).

It is informative to examine the controversial deposits at Trench 14 and the modern pedogenic deposits in relation to other deposits at Yucca Mountain. The isotopic properties of waters of the Stuart fault potentiometric mound and of the groundwater discharge deposits associated with Pliocene activity of this mound offer an illuminating comparison. If it were presumed that the fluids from the modern-day hydrologic anomaly tapped by well EWDP-1s (see Figure 2-8) were discharged at the surface of Yucca Mountain at Trench 14, then the calcite precipitated therefrom would carry $\delta^{18}O$ values between 16.5 and 17.3 ‰ SMOW and $\delta^{13}C$ values between -2.5 and –5.0 ‰ PDB (assuming equilibrium isotope fractionation at 15°C).

Figure 2-11-*b* demonstrates that the Pleistocene deposits at LWD site carry $\delta^{18}O$ and $\delta^{13}C$ values, which are similar to those from the Trench 14 deposits and to modern pedogenic deposits in the pinyon-juniper-sagebrush vegetation zone. If the logic employed by Quade and Cerling (1990) were followed, one would conclude that the LWD deposits are of the pedogenic origin.

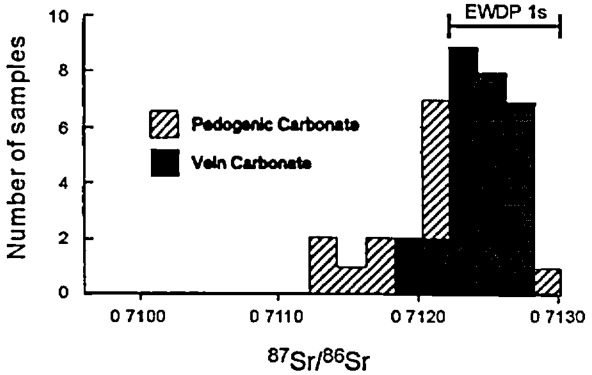


Figure 2-12. Isotopic character of strontium contained in samples of calcretes and associated veins at Trench 14, east slope of Yucca Mountain (from Stuckless, 1990). Line denoted EWDP 1s shows isotopic properties of water in modern hydrologic anomaly (as reported in Paces and Whelan, 2001). The phrase "Pedogenic carbonate" denotes calcretes.

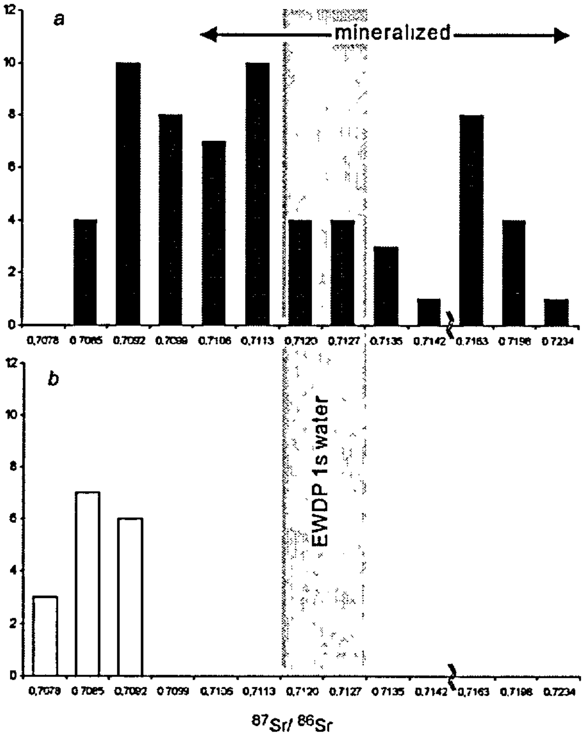


Figure 2-13. Histograms of ^{78}Sr/^{86}Sr for mineralized and non-mineralized Paleozoic carbonate rocks from Bare Mountain (*a*) and Spring Mountains (*b*). Reproduced (recalculated δ^{87}Sr values) from Peterman et al. (1986). Values of modern EWDP 1s water (as reported in Paces and Whelan, 2001) are shown for comparison. Note discontinuous scale after 0.7142.

Stuckless (1990) reported ^{87}Sr/^{86}Sr values from vein calcite and slope calcrete from Trench 14 and concluded that these values are incompatible with the upwelling water origin. Again, if it were presumed that the fluids from the Stuart fault mound were discharged at the surface of Yucca Mountain in Trench 14, then the deposits precipitated from them would carry ^{87}Sr/^{86}Sr values between 0.7121 and 0.7133. These values are identical, for all practical purposes to actual values from the Trench 14 deposits, as illustrated on Figure 2-12.

It is important to recognize that the ^{87}Sr/^{86}Sr values, shown on the figure, are also similar to the values that are associated with mineralized fault traces at Bare Mountain. These contain elevated (mineable) abundances of gold, fluorite and mercury. They also have elevated concentrations of arsenic, cadmium, lead, and zinc, as described in Tingley (1984).

The distributions of the ^{87}Sr/^{86}Sr ratio in this general area have been studied by Peterman et al. (1994). The results revealed important trends in this distribution, as shown on Figure 2-13.

The figure shows that the carbonate rocks, which lie along traces of the mineralized faults at Bare Mountain, are significantly enriched in the radiogenic isotope ^{87}Sr. The corresponding ^{87}Sr/^{86}Sr values range between 0.7106 and 0.7234. The adjacent but barren rocks, however, contain significantly less of the radiogenic isotope, so that the ^{87}Sr/^{86}Sr ratio is between 0.7084 and 0.7106. Moreover, the ^{87}Sr/^{86}Sr values decrease systematically to the east and to the southeast of Bare Mountain, becoming between 0.7078 and 0.7099 in the area of Spring Mountains. The latter values are indistinguishable, for all practical purposes, from the primary marine values, which range between 0.7080 and 0.7090, for the early Paleozoic carbonates, as given in Burke et al. (1982).

The high degree of strontium alteration of the fault traces at Bare Mountain implies that the transport of ^{87}Sr involved hydrothermal solutions ascending from the ^{87}Sr-enriched clastic complexes of the basement. Following Romberger (1986), Peterman et al. (1994) envisioned "... *a hydrologic setting in which shallow, oxygenated groundwater overlies a reducing groundwater 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*".

It is clear therefore that the mineralized faults served as pathways for the ascent of mineralized and ^{87}Sr-enriched solutions. Whilst considering the processes responsible for ^{87}Sr alteration at Bare Mountain we should not be seeking a solution in potentiometric head fluctuations, which could have been caused by past climatic changes. Instead, we should recognize the role, which the deforming, active faults play in long-term behavior of the unstable hydrologic subsystem. At Yucca Mountain, the intermittent ascent of a hydrothermal plume is intrinsic to the long-term behavior.

An increase in the geothermal gradient suggested by Peterman et al. (1994) is only one of several events, which may be involved in triggering the ascent of a hydrothermal plume. In the case of a quasi-stable Rayleigh-Bernard instability such as those, which occupy the horst-bounding fault zones at Yucca Mountain, any decrease of R_c-R_a is potentially capable of triggering the plume.

2.1.9. Consequences of the misinterpretations of the ^{87}Sr data

Without knowing the age, but considering the ^{87}Sr distributions shown on figures 2-12 and 2-13, how is it possible to accept the Stuckless et al. (1991) conclusion that the ^{87}Sr data "preclude" the upwelling origin for the Trench 14 deposits? Similarly, Quade and Cerling's (1990) conclusion, based on the comparison of stable isotopic properties, that the vein carbonates from Trench 14 are of "pedogenic" origin, does not withstand serious scrutiny. Yet the subsequent decision of U.S. DOE to proceed with the characterization of the site, investing 12 years of critically needed time on top of several billions of dollars, has been based to a large extent on these conclusions.

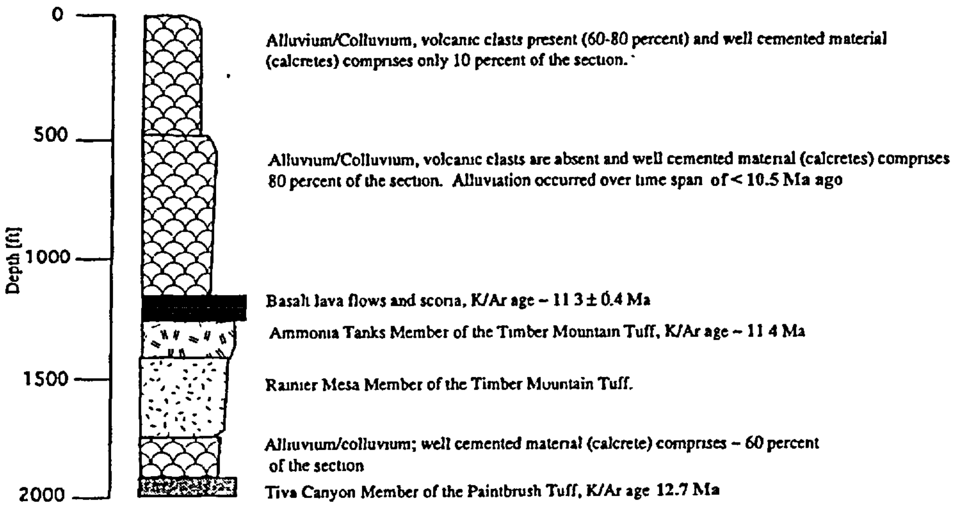


Figure 2-14. Lithologic log of the uppermost 700 m of the USW VH-2 core. Based on data of Carr and Parrish (1985).

2.1.10. Age of the Bare Mountain ^{87}Sr alteration

Given the isotopic data, at least in the case of Bare Mountain, one can be reasonably certain that the ^{87}Sr alteration is an expression of the ascent of hydrothermal plumes along faults in accordance with the conceptual model presented here. However, we have not yet determined the ages of the hydrodynamic instabilities expressed by the ^{87}Sr alteration. Because Bare Mountain is located at a lateral distance of only about 15 km to the west of Yucca Mountain, this age range is of critical importance.

Weiss et al. (1993) showed that the age of the heavy metal enrichment at Bare Mountain is between 11 and 14 Ma. Thus, it is possible that the ^{87}Sr enrichment is also of a late Tertiary age, and further that this enrichment, at least in part, has been produced by the Timber Mountain hydrothermal episode. Nonetheless, it is also possible that the spatial association with the heavy metal enrichment merely reflects the commonality of pathways of fluid ascent, so that the ^{87}Sr enrichment may have been introduced independently and at a different time.

2.1.11. The 12.7 Ma Sr, C, and O Isotope Record from Drill Hole USW VH-2

It can be expected that the answers to questions regarding the age of the ^{87}Sr alteration at Bare Mountain lie buried in the Crater Flat depression located immediately to the east of Bare Mountain. These deposits were formed during a time span, which in part includes the period of heavy metal enrichment of the Bare Mountain rocks. The ^{87}Sr-enriched faults occur at altitudes much higher than the floor of Crater Flat and the depression is infilled with colluvial and alluvial deposits laid down over the past 12.7 Ma.

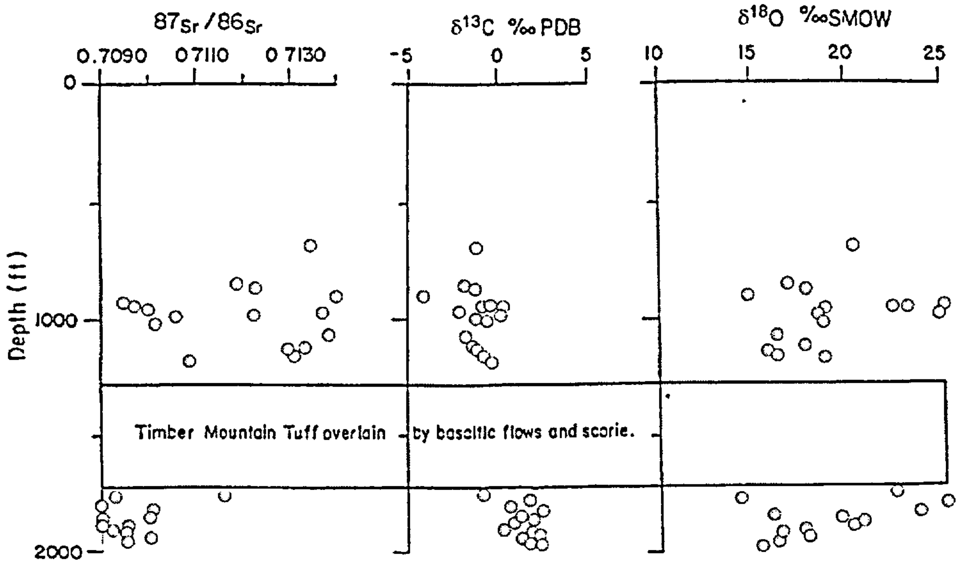


Figure 2-15. Isotopic character of strontium, carbon and oxygen incorporated in calcareous cements from the USW VH-2 borehole.

It is reasonable to assume that the depression-filling detritus was transported from the Bare Mountain upland into the subsiding depression by run-off. Our conceptual model infers that the run-off was partly composed of solutions discharging from the mineralized (heavy metal) faults. It is also reasonable to infer that the run-off could have been involved in cementing the accumulating detritus and, in this manner, record the isotopic composition of these solutions. With this possibility in mind, we have obtained a vertical isotope ($\delta^{13}C$, $\delta^{18}O$, and $^{87}Sr/^{86}Sr$) profile from drill hole USW VH-2, which records changes of the isotope indices as a function of stratigraphic time.

The USW VH-2 drill hole is situated in central part of the Crater Flat depression, half way between Bare Mountain and Yucca Mountain. This drill hole revealed that a 700-m thick section of detritus, derived and transported mainly from Bare Mountain, veneers the depression. The location of the drill hole is shown in Figure 2-4 and the stratigraphy and lithology of the section are summarized on Figure 2-14.

Figure 2-14 shows that the VH-2 section overlies the Tiva Canyon Member of the Paintbrush Tuff. Near the base of the section is the Timber Mountain Tuff, which is overlain by about 11 Ma old basalt flows and scoria. The section consists, above and below the volcanic sequence, of layers of $CaCO_3$-cemented detritus (i.e., calcrete) alternating with layers of loose detritus. The clast population consists of fragments of limestone-dolomite with occasional fragments of quartzite and phyllite. The cement consists predominantly of carbonate with variable amounts of clay, silt, and sand. The alternation of cemented

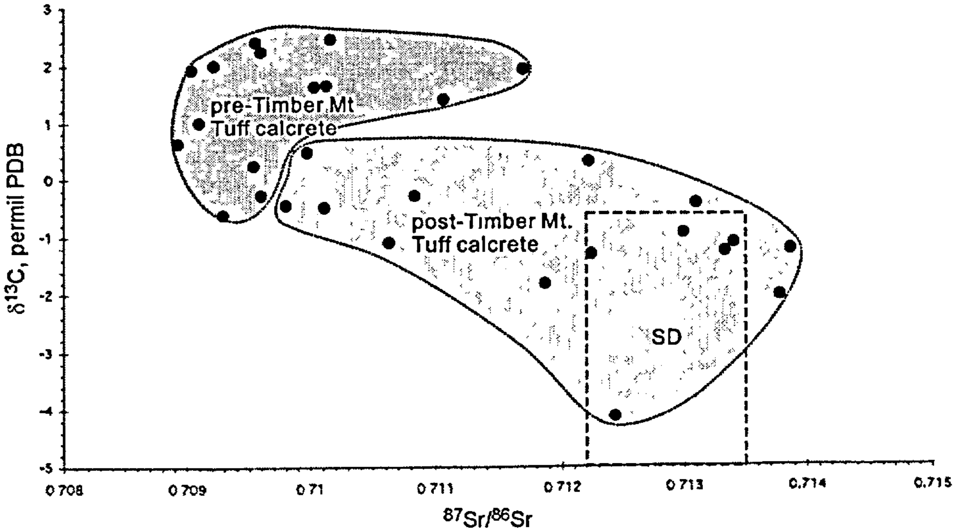


Figure 2-16. $^{87}Sr/^{86}Sr$ *vs.* $\delta^{13}C$ cross plot of calcretes from the USW VH-2 core. SD box – late Quaternary spring deposits from southern Crater Flat and from Devil's Hole. Based on data from Carr and Parish (1985), U.S. DOE (1993), Harmon (1993), Coplen et al. (1994), Marshall et al. (1990), Szymanski et al. (1996), and Paces and Whelan (2001).

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layers of calcrete with layers of loose detritus indicates that the cement has been introduced at the topographic surface and that the cementation continued intermittently.

The isotope record is summarized in the form of the distribution of the $^{87}Sr/^{86}Sr$, $\delta^{13}C$, and $\delta^{18}O$ values as a function of depth (Figure 2-15), which allows for crude estimates of the age, as shown on Figure 2-14. The isotopic character is also summarized in the form of a $^{87}Sr/^{86}Sr$ *vs.* $\delta^{13}C$ cross-plot, as shown on Figure 2-16. Both of the Figures 2-15 and 2-16 contrast the isotope compositions of the pre-Timber Mountain Tuff cements (i.e. ~11.5 Ma old) with those of the subsequent cements.

Figures 2-15 and 2-16 show that the VH-2 calcretes are developed in the form of two isotope facies, which were formed at different times. The older (11.6-12.7 Ma) calcretes occur at a depth ranging between 585 and 651 m (1919-2136 ft), below the Timber Mountain Tuff. The cementing carbonate is depleted in radiogenic strontium, and thus it is associated with $^{87}Sr/^{86}Sr$ values between 0.7089 and 0.7116. At the same time, this carbonate is enriched in "heavy" isotope of carbon ($\delta^{13}C$ values between -0.68 and 2.42 ‰ PDB).

The $^{87}Sr/^{86}Sr$ and $\delta^{13}C$ values both indicate that the cements have been derived from marine carbonates of an early Paleozoic age (Hoefs, 1987 and Burke et al., 1982). 0.7092 is the primary marine $^{87}Sr/^{86}Sr$ value for limestone of an early Paleozoic (Cambrian) age (Burke et al., 1982). The $^{87}Sr/^{86}Sr$ values of more than 0.7092 could be regarded as representing the fluids, which introduced the heavy metal mineralization. However, these values could as well be attributed to a down-slope contamination of the run-off. This contamination would be a result of the introduction of ^{87}Sr-enriched particles deflated from outcrops of the clastic-metamorphic basement. Introduction of ^{87}Sr-enriched strontium leached-out from the non-carbonate rocks (quartzite and phyllite) of the basement would produce similar results.

This hypothetical contamination is similar to the effects observed by Marshall and Mahan (1994) in modern (Holocene) calcretes emanating from the mouth of Lee Canyon in the northern Spring Mountains. The values of ^{87}Sr/^{86}Sr in the Lee Canyon calcretes were found to increase downslope from 0.7084 to 0.7100. Marshall and Mahan (1994) showed that this resulted from the addition of ^{87}Sr-enriched particles deflated from outcrops of the clastic-metamorphic basement.

Figures 2-15 and 2-16 show that, relative to the underlying facies, the post-Timber Mountain Tuff facies is enriched in radiogenic strontium and yields ^{87}Sr/^{86}Sr values between 0.7095 and 0.7137. This upper facies is depleted in the "heavy" isotope of carbon and has mostly negative δ^{13}C values (0.4 and -4.2 ‰ PDB). The elevated ^{87}Sr/^{86}Sr values indicate the involvement of Sr from the clastic basement. Some of these ^{87}Sr/^{86}Sr values are similar to those, which are associated with mineralized faults (Figure 2-13). The generally negative δ^{13}C values indicate the involvement of "light" carbon dioxide, which could have been provided by either a biogenic source or an igneous source, as given in Hoefs (1987).

The cessation of carbon derived from the locally abundant inorganic reservoir (marine carbonate) is particularly important. Presumably, the biogenic source was active both before and after eruption of the Timber Mountain Tuff. Why, then, is there a change? Is it possible that the plume activity, held responsible by Peterman et al. (1994) for the ^{87}Sr alteration at Bare Mountain, post-dates the Timber Mountain Tuff?

Our proposed conceptual model implies that plume activity is the cause of ^{87}Sr alteration. This model implies both the involvement of radiogenic strontium from the clastic-metamorphic basement and the involvement of "light" juvenile carbon dioxide.

Figure 2-16 shows that, in the case of the younger facies, the δ^{13}C and ^{87}Sr/^{86}Sr values are inversely related to each other, in that a decrease of one is accompanied by an increase of the other. The decrease in δ^{13}C can be attributed to the increased involvement of the "light" igneous CO_2. The increased values of ^{87}Sr can be explained by the greater post-Timber Mountain depth of fluid circulation, which promotes the acquisition of ^{87}Sr. The scattering of the ^{87}Sr may reflect dilution by rainwater (that is low in ^{87}Sr).

Figure 2-16 also shows that the ^{87}Sr/^{86}Sr and δ^{13}C values from the younger facies, are similar to those which are associated with local spring deposits of late Quaternary age (see Figure 2-8). The youngest are the Lathrop Wells diatomites (LWD), which according to Paces et al. (1993) have ^{230}Th/U ages ranging between 17.4±2.9 and 45.7±4.9 Ka. The others occur at and around the Devil's Hole cavern in Ash Meadows, with ^{230}Th/U ages between 550 Ka and essentially the present.

The overlap of the stable and radiogenic isotope signatures from the younger USW VH-2 facies and the late Quaternary spring deposits is potentially very significant. The correspondence implies that the acquisition of radiogenic strontium from the clastic-metamorphic basement and the upward transport of this strontium into the overlying rocks are relatively recent and ongoing phenomena. It also suggests that these phenomena are being assisted by carbon dioxide from a juvenile (igneous) source. Furthermore, the occurrence of ^{87}Sr-enriched rocks high above the present-day water table seems to be telling us that this transport involves intermittent upwelling of groundwater.

2.1.12. Concluding Remarks

The preceding analyses lead us to a conclusion that neither is there a basis nor is it reasonable to attribute the LWD and CFW potentiometric anomaly to lateral gradients of hydraulic conductivity. The data indicate that the anomaly is an expression of a local upward flux of groundwater. The established facts cannot support an interpretation that these conditions are being controlled by climatic fluctuations. This leaves tectonic control as the only viable remaining hypothesis.

The preceding conclusion should be regarded as a red flag of caution concerning the wisdom of permanently disposing of spent nuclear fuel in the interior of Yucca Mountain. Tectonic control of upward groundwater flux is not geographically restricted to the LWD and CFW sites. The isotope signatures of the present-day and former fluids at the confirmed paleo-discharge sites LWD and CFW are similar to those of the parent fluids for the Trench 14 calcite-silica deposits. This similarity implies that tectonically controlled upwelling of groundwater is intrinsic to the entire area wherein Yucca Mountain is situated.

Chapter 2.2. Origin of the Vadose Zone Interstitial Fluids

2.2.1. Introduction

The proposed conceptual model implies that the pore fluid in the siliceous tuffs comprising the vadose zone at Yucca Mountain is, in part, a remnant of hydrothermal fluid. The model implies that the pore fluid would have been introduced during one or more episodes of hydraulic mounding induced by past seismic and hydrothermal processes. This concept was initially developed and tested by Szymanski (1989 and 1992).

Any demonstration that there is a component of hydrothermal fluid in the vadose zone pore fluid would negate the basic premise of the conceptual hydrologic model upon which of the safety of Yucca Mountain is presently based. The issue of hydrothermal activity lies at the heart of the controversy. The origin of the pore fluid is another form of the questions, which surround the origin of the calcite-silica deposits at the ground surface and in the Exploratory Studies Facility.

2.2.2. Previous investigations

Questions regarding the origin of the pore fluids in the vadose zone tuffs at the Nevada Test Site are not new. They have lingered unresolved for the past four decades. Initially, Keller (1960 and 1962) has shown that there are large differences in bulk chemistry between the pore fluids and the fracture-based aquifer fluids. The differences were identified from electric resistivity logs, interpreted using laboratory measurements of the electric conductivity of tuff cores saturated with water with varying levels of salinity. It was concluded that the specific conductance of the pore fluids was a factor 25-30 higher than that of the fracture-based aquifer fluids. This conclusion, in turn, indicated that the concentration of total dissolved solids in the pore fluids could have been as high as 5,100 ppm. For the aquifer fluids, however, the corresponding concentration is typically less than 500 ppm.

A chemical compositional difference between pore fluid and fluid residing in fractures at the Nevada Test Site has been demonstrated. Fluids from Rainier Mesa, in the northeastern part of the Nevada Test Site, were examined by White et al. (1980) and later Russell (1987). These authors showed that, relative to the fracture-based fluids, the pore fluids carry significantly higher concentrations of dissolved chloride (Cl^-) and sulfate (SO_4^{2-}) anions. White et al. (1980) noted: "... *the mechanism responsible for differences in anion composition between interstitial and fracture waters is not obvious*".

Nonetheless, White et al. (1980) considered that the type and extent of ionic exchange reactions for the pore fluids must have been different from those that controlled the chemistry of the fracture-based fluids. The only way White et al. (1980) could explain the differing exchange reactions was to hypothesize that the anomalous fluids are, in part, relict fluids. The authors reasoned that fluids enriched in chloride and sulfate would have been trapped inside the interstitial space at a time when the tuffs were emplaced. They noted that even partial preservation of such fluids, over a time span as long as 12 Ma, would require that the interstitial space be effectively isolated from rainwater descending along fractures. Considering the relatively high matrix permeability of the tuffs, however, White et al. (1980) regarded this proposition with skepticism.

Szymanski (1992) reviewed the available data regarding the chemical and isotope compositions of the pore water at Yucca Mountain. He summarized the results by concluding, "... *the interstitial fluids contain two genetically distinct fluid phases. One of these phases is rainwater that, during episodic recharge events, is being introduced into the...pore space. Its presence is clearly manifested through both above-background tritium activity and 14C ages ranging between* 10^3 *and* $4.9 \cdot 10^3$ *years. The other fluid phase seems to represent an oxygen18- enriched and chemically evolved hydrothermal solution. Its presence is manifested through 1) elevated concentrations of the dissolved alkali earth elements, 2) abnormally high concentrations of the dissolved transition and noble metals, and 3) relative to LREE, HREE enrichment and overall high abundance of REE. ...being strongly held by capillary forces, the interstitial fluids remain immobile and, while exchanging vapor phase components, maintain their original concentrations of dissolved solids. In spite of their isotopically youthful meteoric appearance, these fluids also exhibit sharply contrasting to this appearance geochemical characteristics*".

Below we discuss evidence that allows us to infer that the pore fluid which is resides in the vadose zone tuffs at Yucca Mountain is not entirely composed of infiltrating rainwater.

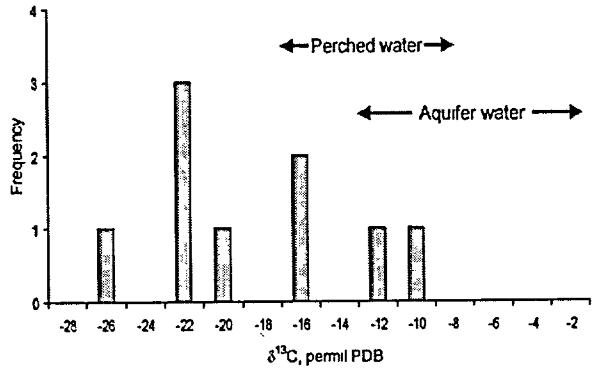

Figure 2-17. $\delta^{13}C$ values for the pore water residing in the non-welded tuffs of Yucca Mountain. Values, characteristic of perched waters and Tertiary volcanic aquifer waters are shown for comparison.

Waters for analysis were extracted by compression or centrifugation of cores from drill cores UE-25 UZ#4, UE-25UZ#5, and UE-25 UZ#16 and reported by Yang (1989) and Yang et al. (1996). Data for perched waters are from Yang et al. (1996), and for aquifer waters from McKinley (1985).

2.2.3. Stable Isotope Signatures

Carbon. Yang (1989) and Yang et al. (1996) reported $\delta^{13}C$ values for the pore water, samples of which were extracted by compression or centrifugation of cores from drill holes UE-25 UZ#4, UE-25UZ#5, and UE-25 UZ#16. These values range between a maximum of -10 and as low as -26.7 ‰ PDB, as shown on Figure 2-17.

The conspicuously "light" values shown on the figure are significantly lower than those expected to result from dissolution of soil CO_2-gas, which represents a mixture of the atmospheric CO_2 and the CO_2 respired by local plants. At an altitude of about 1400 m asl in Nevada, such gas has $\delta^{13}C$ values between -12 and -16 ‰ PDB (Amundson et al., 1988). At a temperature of about 15°C, the fractionation factor between gaseous and dissolved CO_2 is about 9 ‰ (Hoefs, 1987). Thus, dissolution of the soil CO_2 would produce fluids, which carry $\delta^{13}C$ value between -3 and -7 ‰ PDB.

The values shown on Figure 2-17 are also much lower than those for fluids, which are present today at Yucca Mountain or are known to have been there at some time in the past. These values are also much lower than those from the bodies of perched water, which according to Yang et al. (1996) have $\delta^{13}C$ between -9 and -17 ‰ PDB. They are also much lower than those from the aquifer fluids, which carry $\delta^{13}C$ between -2.3 and -13.1 ‰ PDB (Benson and McKinley, 1985).

The values shown on Figure 2-17 are much lower than those inferred to have been associated with parent fluids of the controversial calcite-silica deposits, which occur at the ground surface and in the Exploratory Studies Facility. These genetically homogeneous deposits have $\delta^{13}C$ values between -10 and about +9 ‰ PDB, as reported by Quade and Cerling (1990), Dublyansky (2001), and Whelan et al. (2001). The inferred $\delta^{13}C$ values for CO_2 dissolved in the parent fluids (as bicarbonate ion) therefore range between -11 and about +7 ‰ PDB.

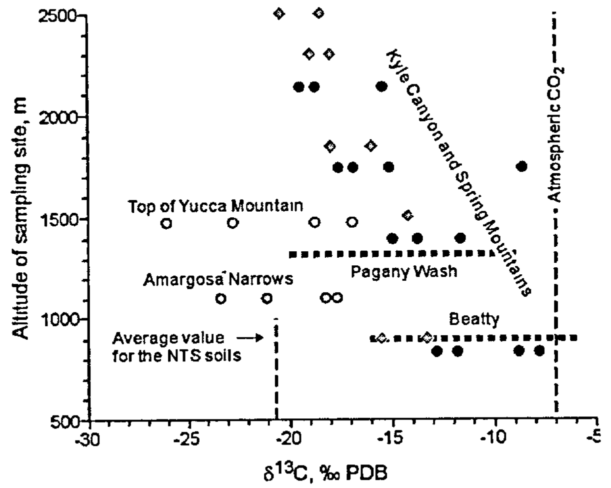

Figure 2-18. $\delta^{13}C$ in soil gas at different altitudes as observed at and in the vicinity of Yucca Mountain.

Data sources: Black filled circles – Kyle Canyon (Amundson et al., 1988); *gray diamonds* – Spring Mountains (Quade et al. 1998); *gray filled circles* – Amargosa Narrows (White and Chuma, 1987); *open circles* – top of Yucca Mountain (Thorstensen et al., 1989); *horizontal dashed lines* – U.S. Ecology site at Beatty and Pagany Wash, Yucca Mountain (McConnaughey et al. (1995). Average value for the Nevada Test Site soils (Boughton, 1986) and average atmospheric value are shown for comparison.

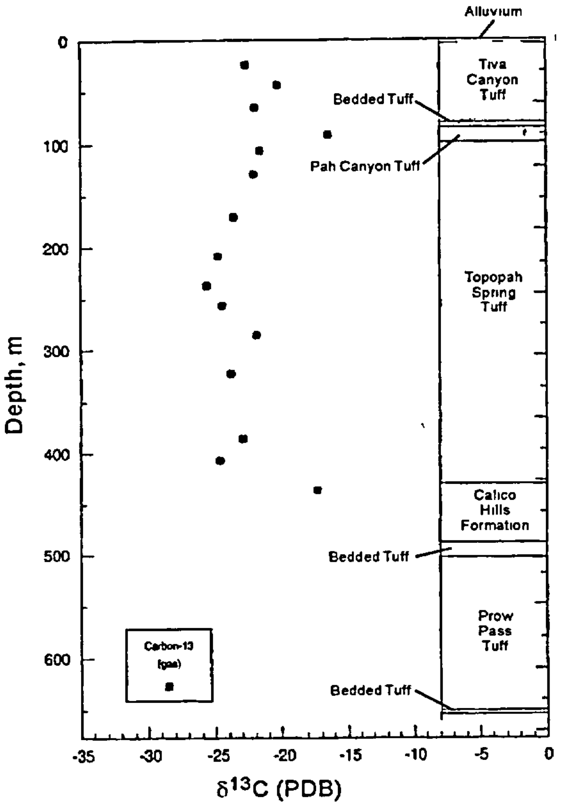

Figure 2-19. $\delta^{13}C$ (gas) values in borehole USW SD-12, Yucca Mountain area. From Yang et al. (1998).

It thus appears that the only way the pore fluid could have acquired its highly unusual $\delta^{13}C$ values is through equilibration with a gas containing CO_2 formed through oxidation of methane (CH_4). Somewhat similar gas seems to be circulating in the present-day vadose zone, as shown on Figure 2-18.

The figure shows that the vadose zone gas at Yucca Mountain has $\delta^{13}C$ values up to about -26 ‰ PDB. These values are consistently lower than the mixture of the atmospheric CO_2 and the CO_2 respired by plants at comparable altitudes. The $\delta^{13}C$ values measured in the gaseous CO_2 from boreholes in the unsaturated zone of Yucca Mountain are similarly light. Yang et al. (1998) reported values ranging between –17 and –25 ‰ PDB (Figure 2-19).

The enriched in light isotope of carbon (^{12}C) gas at Yucca Mountain seems to be expressing low intensity degassing of the aquifer fluids, which seem to contain CO_2 and CH_4.

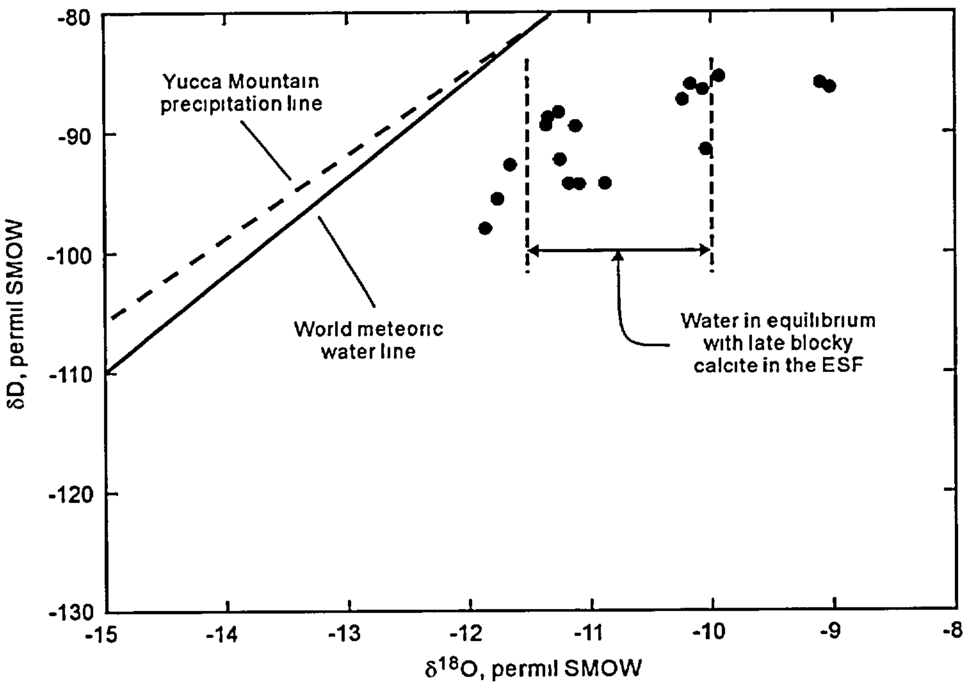

Figure 2-20. Isotopic composition of the Yucca Mountain vadose fluids relative to the World meteoric water line.

Pore waters for analyses were obtained by direct distillation and compression of rock cores from drill holes UE-25 UZ#4 and #5 (data from Yang 1989 and 1992). Vertical dashed lines indicate the range of values of $\delta^{18}O$ in a fluid, equilibrated with late blocky calcite from ESF (crystallized at 40-46°C; the range of $\delta^{18}O$ values was calculated from the $\delta^{18}O$ measured in calcite and fluid inclusion temperatures reported in Dublyansky, 2001).

It is important to note that some of the crystalline calcite, which occurs in the Exploratory Studies Facility, has an unusually "heavy" $\delta^{13}C$ values, reaching +9 to +10 ‰ PDB (Whelan et al., 2001; Dublyansky, 2001). At Yucca Mountain, all the ordinary sources of carbon dioxide (atmospheric, soil-organic, inorganic, igneous) would yield calcite with $\delta^{13}C$ values of no more that 2-4 ‰ PDB. The extreme enrichment in the "heavy" carbon requires some eccentric origin for the dissolved carbon species.

According to Ohmoto (1972) and Rye and Ohmoto (1974) the calcite with such a heavy carbon composition should precipitate from a fluid, which has acquired the dissolved carbon at a high temperature (~250°C) and in a low oxygen fugacity environment (log fO_2 of -37 or less). Under such conditions, the dissolved carbon would likely be partitioned into two chemical species – CO_2 and CH_4 – having sharply differing oxidation states (-4 and +4). Partition leads to substantial isotope fractionation with "light" (^{12}C) carbon preferentially concentrating in reduced species (methane) and "heavy" isotope (^{13}C) – in oxidized species (carbon dioxide). Only oxidized species would participate later in the precipitation of $CaCO_3$ and the later would inherit the "heavy" positive values of $\delta^{13}C$.

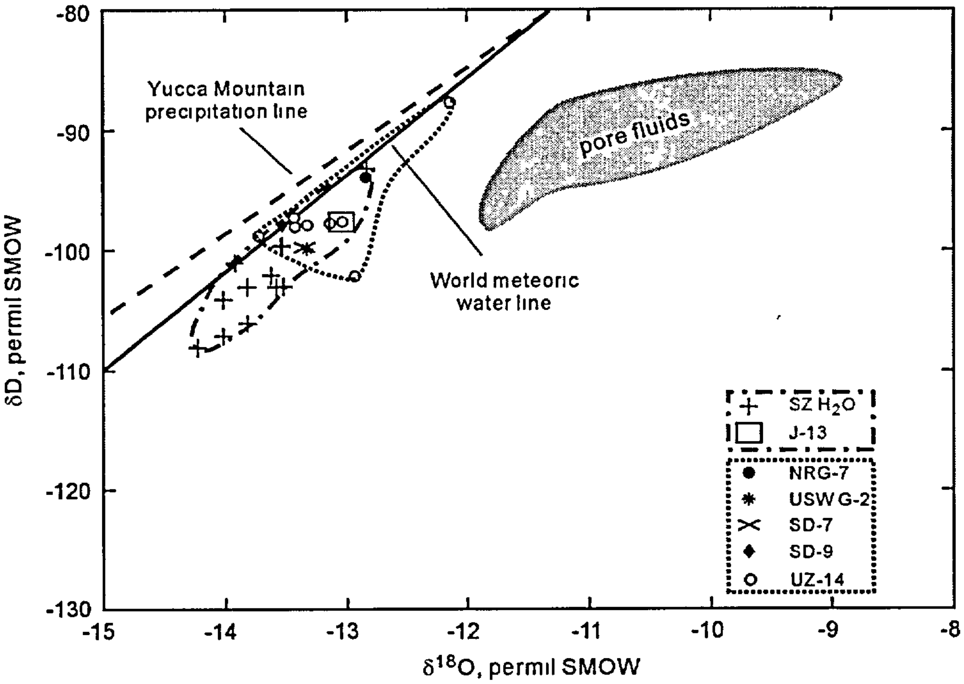

Figure 2-21. Isotopic compositions of the Yucca Mountain saturated zone and perched waters. Data for saturated zone waters (SZ H_2O and J-13) are from boreholes UE 25 b#1, c#3, p#1, a#2, USW G-4, H-1, -3, -4, -5, -6, VH-1, and J-13. A field or the Yucca Mountain vadose zone pore waters reported by Yang (1989; 1992; see Figure 2-20) is shown for comparison. Modified from Yang et al. (1996).

Oxygen. Yang (1989 and 1992) also reported $\delta^{18}O$ values for pore water, samples of which have been extracted by distillation and compression of rock cores from drill holes UE-25 UZ#4 and #5. These values are shown, relative to the meteoric water line, on Figure 2-20.

The figure shows that the $\delta^{18}O$ values range between -9.5 and about -12.5 ‰ SMOW and, relative to the meteoric line, exhibit a pronounced "oxygen isotope shift" of 0.5 to 2.5 ‰. Importantly, the range of the $\delta^{18}O$ values measured in pore waters is similar to that, inferred for the fluids that have deposited the latest crystalline calcite in fractures and cavities of the ESF (see Part Three, Chapters 3-5 and 3-6 for details). The range of the $\delta^{18}O$ values (shown in Figure 2-20) was calculated using $\delta^{18}O$ values measured in host calcite and the temperature, obtained from the fluid inclusions homogenization experiments.

Range for the isotope composition of aquifer fluids at Yucca Mountain is shown on Figure 2-21. The figure shows that the "oxygen isotope shift" is much smaller than that associated with the pore fluid. A mechanism, which is responsible for this difference, is not obvious, and there are two equally plausible explanations for it. The shift in pore fluid could be expressing the greater degree of the evaporative enrichment of the pore fluids at ambient temperature. On the other hand, it could as well be expressing the isotope enrichment brought about by the rock → fluid exchange at an elevated temperature, or by boiling.

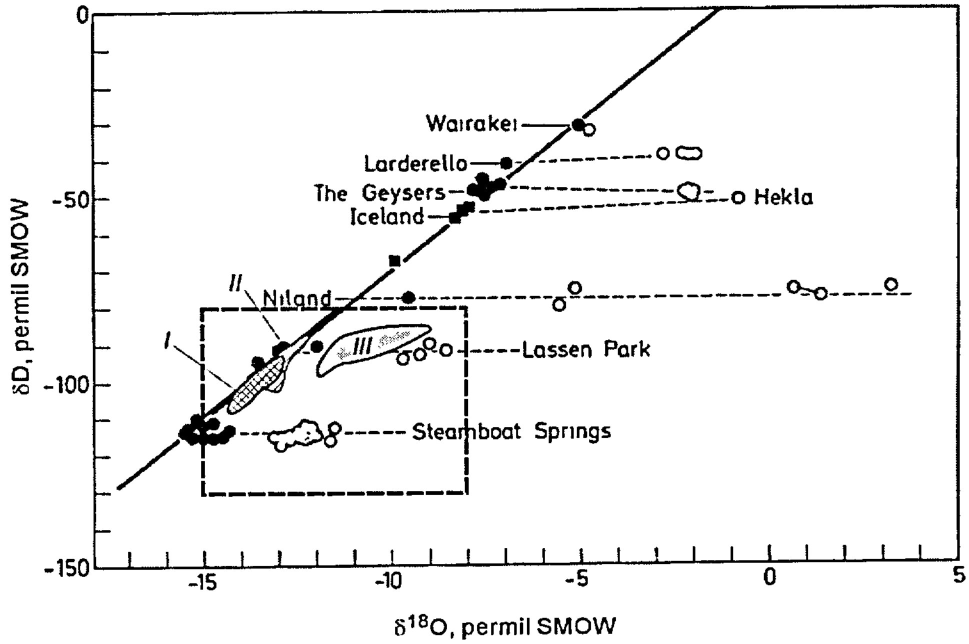

Figure 2-22. Observed isotopic variations in near-neutral, chloride type geothermal waters and in geothermal steam. Based on Craig (1963), reproduced from Hoefs (1987). Fields within rectangle correspond to isotope composition of the saturated-zone water (*I*), perched water (*II*), and the vadose-zone pore fluid (*III*) at Yucca Mountain (see Figures 2-21 and 2-22).

Squares – local meteoric waters or slightly heated near-surface groundwaters; *filled circles* – hot spring geothermal water; *circles with dots* – high temperature, high-pressure, geothermal steam.

Although not uniquely valid, the latter interpretation is clearly permissible. This is because the "oxygen isotope shift" observed in Yucca Mountain pore fluids is comparable to the ^{18}O enrichment, which is typically associated with geothermal waters and steams, as shown on Figure 2-22.

2.2.4. Major Element Chemistry

In addition to the thought-provoking stable isotope composition, the pore fluid chemistry seems to be incompatible with the shallow and alkali tuff environment. This incompatibility is clearly expressed by the concentrations of the major dissolved cations and anions, as well as by the content of total dissolved solids.

The major element chemistry has been determined by Yang et al. (1996) for samples of the pore fluid, which were extracted mainly from non-welded tuff cores representing upper and lower parts of the vadose zone (Figure 2-23).

Figure 2-23 shows that many samples of the pore fluids have surprisingly high concentrations of the dissolved alkaline earth elements (Ca and Mg), chlorine (Cl), and sulfate (SO_4). As a whole, these fluids appear to be spatially varying mixtures of two end-members. One end-member represents an alkali-

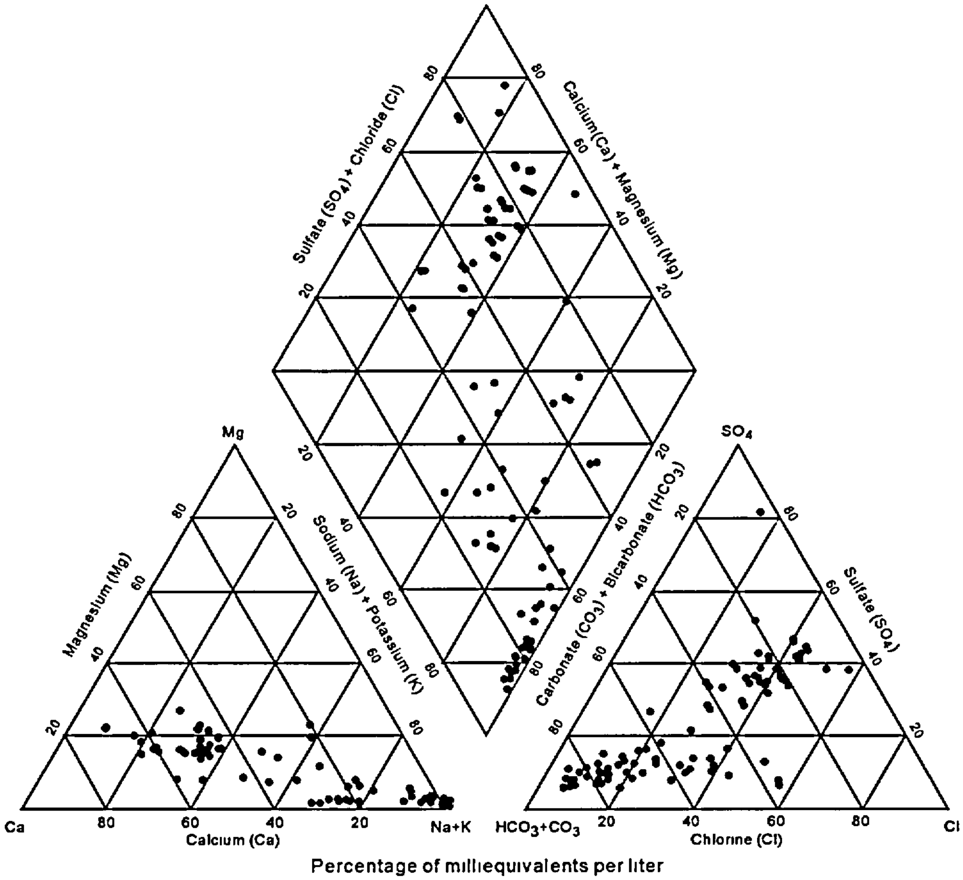

Figure 2-23. Piper diagram showing pore-water compositions. The content of total dissolved solid ranges from 300 to 2800 ppm. From Yang et al. (1996).

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bicarbonate-carbonate groundwater facies, whereas the other represents an alkaline earth-chloride-sulfate facies.

The amount of total dissolved solids varies between ~300 to 2800 ppm. This indicates that one of the end-members is fresh water but the other is brackish water, as defined in Gorrell (1958). Considering that the pore fluids occur at a depth of less than 500 m and entirely reside in the alkali tuffs, the presence of brackish alkaline earth-chloride-sulfate facies is anomalous and demands a rational explanation.

The first step in constructing an explanation is to compare the chemistry of the pore fluids with that of fluids that reside in the underlying volcanic aquifer (Figure 2-24).

The Piper diagram indicates that the aquifer contains fresh groundwater, which has an alkali-bicarbonate-carbonate chemical character, as expected for alkali bedrock. The only exception is a fluid sample from well UE-25 p#1, taken at a fault contact between the Tertiary tuffs and the early Paleozoic carbonates of the basement (Paintbrush fault). Here, the TDS is highest (812 ppm), the concentration of

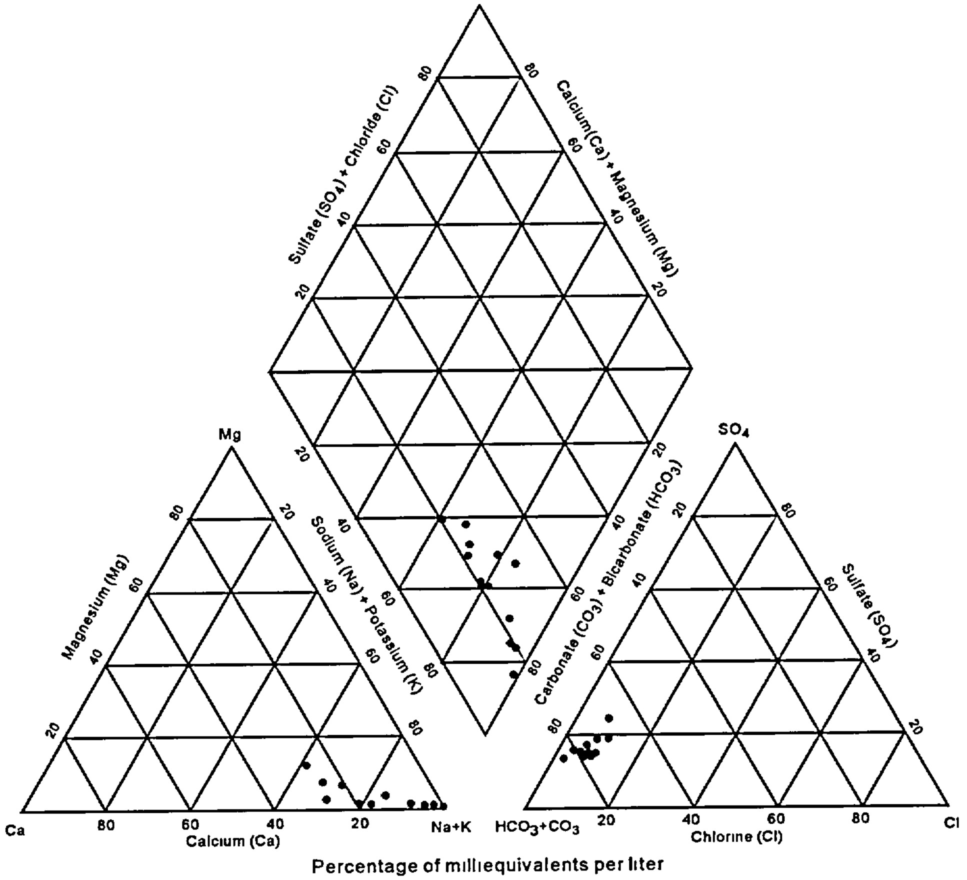

Figure 2-24. Piper diagram showing saturated-zone water compositions. The content of total dissolved solid ranges from 300 to 1000 ppm. From Yang et al. (1996).

alkaline earth elements (Ca+Mg = 139 ppm) is comparable to that of alkali elements (K+Na = 162 ppm), and the concentration of SO_4 and Cl anions is increased (SO_4+Cl = 188 ppm).

However, Carr at al. (1986) found that the UE-25 p#1 fluid is anomalous in terms of the temperature (56°C) and the piezometric head (20 m higher than that in the overlying tuffs). This fluid is associated with the ascent of hotter and more chemically evolved solutions along the Paintbrush fault, from the Paleozoic carbonates into the tuffs, as shown on Figure 1-30 (Part One).

According to Benson and McKinley (1985), the ^{14}C ages range between 3.8 and 18.5 Ka, for fluids residing in the tuffs, increasing to 30.3 Ka in the Paleozoic carbonates. The chemistry and the apparent ^{14}C ages together demonstrate that even a prolonged residence time, and the resulting chemical exchange reactions in the mixed tuff-carbonate environment, do not produce a fluid whose chemistry resembles that of the near-surface pore fluids.

It thus appears that there are valid reasons to interpret the origin of the pore fluids in a manner, which is embedded in the proposed conceptual model. These fluids may have been introduced into the interstitial space, in the past, by dissipation and diffusion of warm or hot fluids from a hydraulic mound into the vadose zone. Additional lines of evidence, the concentrations of trace elements and REE patterns in particular, are consistent with this interpretation.

2.2.5. Trace Element Abundance

A very revealing data set has been developed and reported by Smith (1991). This concerns the natural radionuclide and trace element hydrochemistry of the aquifer fluids (J-12 and J-13 wells), compared with that of the pore fluids. Samples of the latter were extracted from UE-25 UZ#4 cores by high-speed centrifugation. The results are reproduced in Tables 2-1 and 2-2.

Tables 2-1 and 2-2 show that, in contrast to the J-12 and J-13 aquifer fluid, the UE-25 UZ#4 samples (pore fluid) represent a mineral water enriched in many trace elements. The enrichment is illustrated, relative to the fresh J-12 and J-13 aquifer fluids, in Table 2-3.

Table 2-3 shows that, relative to the aquifer fluids (apparent ^{14}C age 9.1-9.9 Ka; Benson and McKinley, 1985), the pore fluids contain a significantly higher abundance of the dissolved alkaline earths elements (Ca, Mg, and Sr). The alkali metal abundances (Na, K, Rb, and Cs), however, are comparable. The other most obvious compositional differences involve concentrations of I, Cl, Mo, W, transition metals (Mn, Ni, Cu, and Zn), and noble metals (Au and Pt). Smith (1991) recognized the strong compositional discords and concluded that: "*...the compositional differences... indicate that the pore waters have evolved by significantly different processes.*"

Table 2-3 further shows that the major element chemistry of the pore water resembles that of fluids from the Paleozoic carbonate aquifer, as indicated by the eight- to ten-fold enrichment of calcium and magnesium. The enrichment in trace elements, including the rare earth elements (REE) and base and noble metals, indicates a hydrothermal source.

That the both fluids are genetically related is also indicated by the respective $^{87}Sr/^{86}Sr$ ratios. This similarity of isotopic character is shown on Figure 2-25.

Figure 2-25 shows that the pore water is associated with $^{87}Sr/^{86}Sr$ ratio between 0.7122 and 0.7126 (Figure 2-25-*a*). These values are substantially more radiogenic than those of the water from the Tertiary volcanic aquifer (Figure 2-25-*c*). The ESF calcite deposits have a much wider range of $^{87}Sr/^{86}Sr$ ratio, between 0.7097 and 0.7126. The data suggest, therefore, that the pore fluids and the parent fluids for some of the ESF calcite have acquired dissolved strontium at least partially from the same source.

Table 2-1

Chemical composition of groundwater, eastern part of Yucca Mountain (drill holes J-12 and J-13). From Smith (1991).

Sample	J-12	error	J-13	error	SLRS-1*	Cert. val
pH(field)	7.3	5				
pH(lab)	7.69					
F(ppm)	2 00					
Cl(ppm)	7.5					
NO_3(ppm)	8 2					
SO_4(ppm)	22 2					
HCO_3(ppm)	117					
Li	50	10	40	10	1.5	
B	130	10				
Na (ppm)	41 8	1.3				10 4
Mg (ppm)	2 45	0.15	2.4	0 2		5 99
Al	1 2	0.6	0.5			23 5
Si (ppm)	25 6	1.1	30	5		
Cl (ppm)	7 5	0 5				
K	5 5	0 7				1.3
Ca (ppm)	15	0 2	14	5	25	25.1
Sc	<1		<3		0 34	
Ti	0 08		0 1		0.7	
V	4	1	5	1	0.73	0 66
Cr	0.37	0 12	0 33	0 03	0.26	0 36
Mn	0.15	0 06	<0 3		1.19	1.77
Fe	3	1	3	1	27.5	31.5
Co	<0 01		<0 01		0.023	0.043
Ni	0 03	0 01	0 05	0 01	0.7	1.07
Cu	0 2	0 06	0 8	0 2	2 8	3 58
Zn	2.7	0 5	2 4	0 5	0.95	1.3
As	9	3	8	3	0 53	0 55
Rb	9 2	1 1	6 5	1	0 83	
Sr	35	1	33	2	133	136
Y	0 0025	0 0008	0 0025	0 0006	0 28	
Mo	5	0 5	5 3	0 1	0 62	0 78
Pd	<0 01		<0 01			
Cd	<0 05		<0 05		0 011	0 015
Sb	0 15	0 01	0 21	0 01	0 57	0 63
I	2		2		2 22	
Te	<0 01		<0 03		<0 01	
Cs	0 56	0 02	0 95	0 16	0 0032	
Ba	1.75	0.35	1/6 8		21 5	22 2
La	0 0004		0 0003		0 04	
Ce	<0 005		0 0004		0 059	
Pr	<0 001		<0 001		0 01	
Nd	<0 001		<0 001		0 037	
Sm	<0 001		<0 001		0 02	
Eu	<0 001		<0 001		0 0014	
Gd	<0 001		<0 001		0 006	
Tb	<0 001		<0 001		0 0007	
Dy	<0 001		<0 001		0 0035	
Ho	<0 001		<0 001		0 0009	
Er	<0 001		<0 001		0 003	
Tm	<0 001		<0 001		0 0003	
Yb	<0 001		<0 001		0 0028	
Lu	<0 001		<0 001		0 0004	
Hf	0 001		<0.002			

Table 2-1 (cont.)

Sample	J-12	error	J-13	error	SLRS-1*	Cert. val
Ta	<0.001		<0.001			
W	1.0	0.2	2.0	0.4	0.13	
Re	0.056	0.01	0.002/0.83		0.003	
Pt						
Au					0.0004	
Tl	0.01	0.004	0.008			
Pb	0.2	0.06	0.3	0.1	0.062	0.106
Bi	<0.001		<0.003		0.001	
Th	0.0001	0.00003	0.0004	0.0002	0.003	
U	0.55	0.13	0.4	0.14	0.25	0.28

Notes: Concentrations are in ppb unless noted; *SLRS-1 - Riverine Water Reference Material for Trace Metals (National Research Council, Canada).

Table 2-2

Chemical composition of the vadose zone interstitial fluids from borehole UZ#4, Yucca Mountain . From Smith (1991).

Sample	Pore water 6110926	error	USGS (reported)*	Pore water 6110923	Error	USGS (reported)*
pH			6.5			6.5
F(ppm)						
Cl(ppm)						
NO_3(ppm)						
SO_4(ppm)						190
HCO_3(ppm)						
Li	68			85	10	
B						
Na (ppm)			48			53
Mg (ppm)	27	5	21	25	1	23
Al				<5		
Si (ppm)			74			78
Cl (ppm)			160			210
K			5			14
Ca (ppm)	120	10	120	110	10	130
Sc	<3	1		0.4		
Ti				<3		
V						
Cr	<9	<3		0.4	0.1	
Mn	133	25	150	14	3	90
Fe	35	5	150	<20		<3
Co				<1		
Ni	40	10		<50	16	
Cu	40	10		10	1	
Zn	150	15		71	5	
As						
Rb	23	2		12	2	
Sr	1020	110		1180	110	
Y	0.3	0.1		0.21	0.05	
Mo	1880	160		1400	30	
Pd						
Cd				1.4	0.3	
Sb	1			0.7	0.2	

Table 2-2 (continued)

Sample	Pore water 6110926	error	USGS (reported)*	Pore water 6110923	Error	USGS (reported)*
I	50			40		
Te						
Cs	0.5	0.1		0.44	0.05	
Ba	27	3		18	1	
La	<0.1			0.007	0.007	
Ce	<0.1			0.048	0.02	
Pr	<0.04			<0.006		
Nd	<2			<0.05		
Sm	<0.03			<0.006		
Eu	<0.06			<0.006		
Gd	<0.2			<0.003		
Tb	<0.03			0.013	0.005	
Dy	0.09	0.03		0.032	0.006	
Ho	<0.1			0.022	0.006	
Er	0.14	0.04		0.12	0.02	
Tm	0.05	0.02		0.04	0.01	
Yb	0.5	0.2		0.5	0.1	
Lu	0.14	0.06		0.16	0.05	
Hf	0.01			0.3	0.1	
Ta	0.5			<0.2		
W	600	200		330	50	
Re	0.2	0.09		0.2	0.1	
Pt	0.2	0.1		0.2	0.05	
Au	0.12	0.05		0.2	0.1	
Tl	0.2			0.2		
Pb	0.25	0.1		1.7	0.3	
Bi	<0.03			<0.1		
Th	0.0024	0.0006		<0.01		
U	0.45	0.06		0.13	0.03	

Notes: Concentrations are in ppb unless noted; Samples of the intestitial fluids were obtained by employing the high-speed centrifugation method; * Personal communication, Al Yang (as reported in Smith, 1991)

It should also be noted that isotope properties of water from the modern potentiometric anomaly associated with the Stuart fault (see section 1.7) carry $^{87}Sr/^{86}Sr$ values virtually indistinguishable from those of the pore waters documented in Figure 2-25-*b*). In addition, the UE-25 p#1 fluid, sampled at a depth of 1.2 km, chemically resembles the pore fluids shown in Figure 2-25-*b* in terms of major element chemistry. The UE-25 p#1 fluid is associated with an $^{87}Sr/^{86}Sr$ ratio of about 0.7118 (Peterman et al., 1994). This isotope signature confirms the expectation derived from the conceptual model: the pore fluids and the ESF calcite both reflect the past large-scale amplification of a Paintbrush Fault convection cell [heat transfer-dominated dissipative structure].

Table 2-3

Enrichment of the vadose zone interstitial fluids from Yucca Mountain relative to the aquifer fluids (boreholes J-12 and J-13). Calculated from data reported by Smith (1991).

Element	Enrichment	Comments
Li	1.7	
Na	1 2	
Mg	10 7	
Cl	24 7	USGS
K	1.7	USGS
Ca	7 9	
Ti	33 3	Maximum
Cr	13 4	Maximum
Mn	490 0	
Fe	9 2	
Ni	1125 0	
Cu	50.0	
Zn	43 3	
Rb	2.2	
Sr	32.4	
Y	102.0	
Mo	318 4	
Cd	28 0	Minimum
Sb	4 7	
I	22 5	
Cs	0 6	
Ba	2 4	
La	20 0	Minimum
Ce	17 8	Minimum
Tb	13 0	Minimum
Dy	61 0	Minimum
Ho	22 0	Minimum
Er	130 0	Minimum
Tm	45 0	Minimum
Yb	500 0	Minimum
Lu	150 0	Minimum
Hf	103 3	Minimum
Ta	350 0	Minimum
W	310 0	
Pt	**	
Au	**	
Tl	22 2	
Pb	3 9	Estimate
Bi	32.5	Estimate
Th	9 6	
U	0 6	

Note: USGS – data of Yang, as reported by Smith (1991); ** - aquifer waters contained no measurable platinum and gold, whereas interstitial fluids contained 0.2 ppb of these metals.

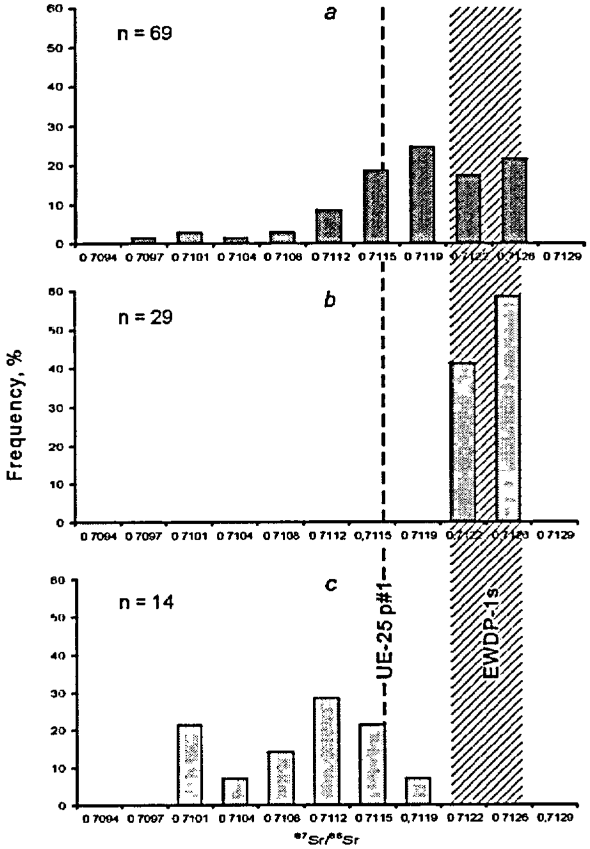

Figure 2-25. Histograms showing ranges of $^{87}Sr/^{86}Sr$ for calcite samples from ESF (*a*), pore waters from TSw tuff unit present at and below the ESF level (*b*), and regional Tertiary volcanic aquifer waters (*c*). Value of $^{87}Sr/^{86}Sr$ in the underlying carbonate aquifer (0.7118) and interval of values measured in water of the hydraulic mound discussed in previous section (well EWDP-1s; 0.7123 to 0.7128) are shown for comparison.

Isotopic composition of Sr in pore waters was determined by leaching of pore-water salts from three boreholes near ESF. Data presented on histograms *a* and *b* were recalculated from $\delta^{87}Sr$ values reported by Marshall and Futa (2001); histogram *c* shows the data reported by Marshal et al. (1991).

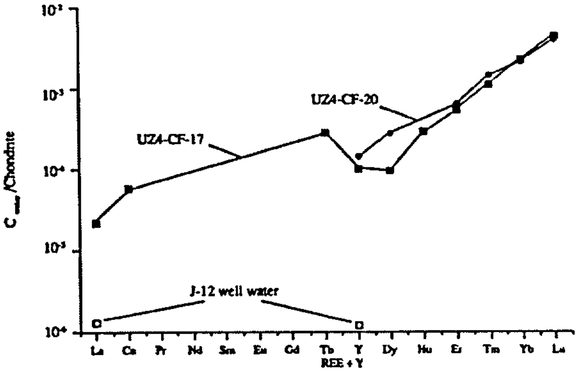

Figure 2-26. Comparison of the chondrite-normalized REE abundances of the aquifer fluids (UZ4-CF-17 and UZ4-CF-20) and interstitial fluids from the vadose zone (J-12). Modified from Smith (1991).

Note that pore fluids contain one- to two-order of magnitude more La and Y relative to aquifer fluids.

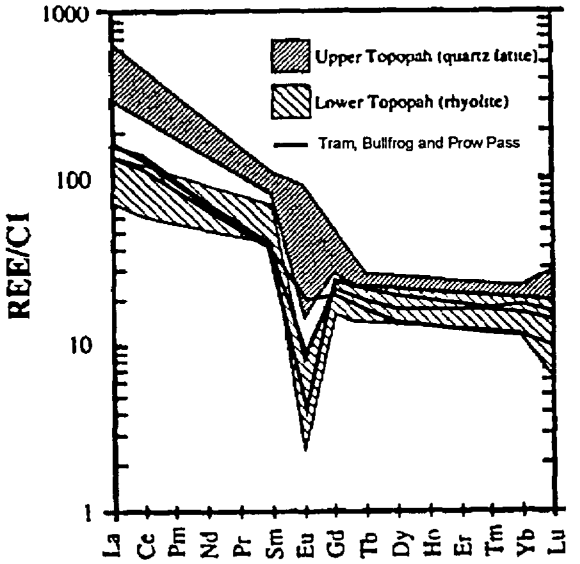

Figure 2-27. Ranges in chondrite-normalized REE patterns for upper and lower portions of the Topopah Spring Tuff (after Schuraytz et al., 1989 and Broxton et al., 1989 as reported in Denniston et al., 1997), and average values (of four samples each) of the Tram, Bullfrog and Crater Flat ignimbries (from Scott and Castellanos, 1984).

2.2.6. REE Enrichment

In addition to the overall enrichment in rare earth elements (REE) noted earlier, there is also an unusual enrichment of heavy rare earth elements (HREE) in the vadose-zone pore fluids. This enrichment, relative to light rare earth elements (LREE), is shown on Figure 2-26.

The chondrite-normalized REE abundance pattern for the pore fluids shown in Figure 2-26 is remarkable. It is the opposite of the pattern, which would develop if the pore fluids were representing ordinary rainwater infiltrating through the vadose zone.

Examining the chondrite-normalized REE pattern for the tuffs at Yucca Mountain is the best way of identifying this discord. This pattern is shown on Figure 2-27.

Figure 2-27 shows that, in contrast to the pore fluids, the host tuffs exhibit a pattern of relative enrichment of LREE. The sharply contrasting enrichment patterns are telling us that the pore fluid chemistry cannot possibly be attributed to chemical exchange reactions between infiltrating rainwater and the host tuff.

The HREE relative enrichment is rarely observed in a shallow environment. Nonetheless, it is a feature that sometimes has been noted in association with CO_2-enriched, highly chemically evolved and concentrated hydrothermal solutions. One such example of is shown on Figure 2-28.

An inclination of the smoothed REE curve, whether positive or negative, indicates that fractionation of the dissolved REE has occurred. This inclination results from ionic exchange reactions between fluid

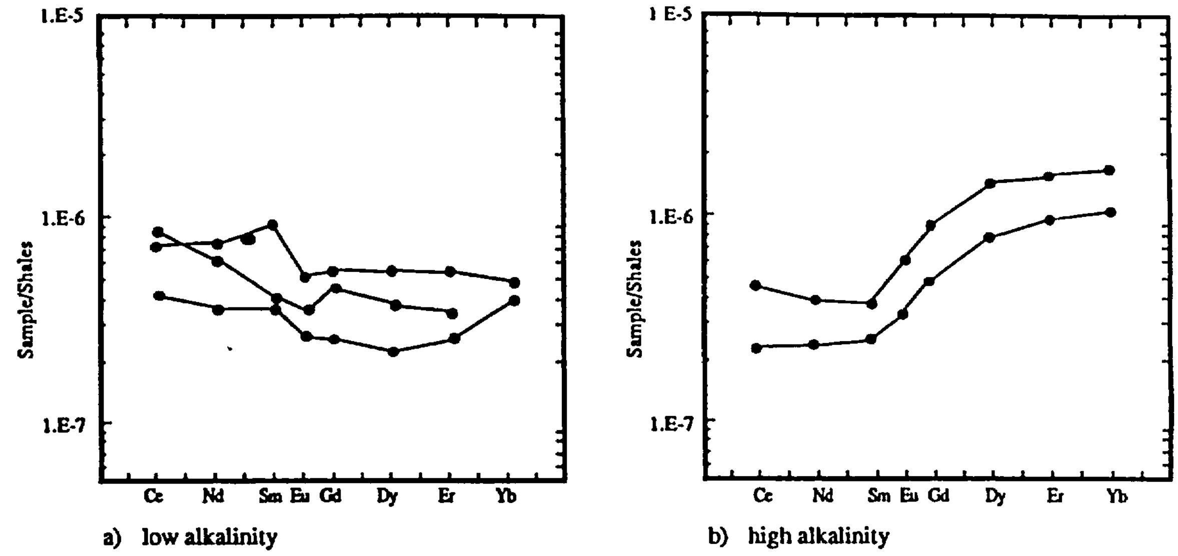

Figure 2-28. Shale-normalized abundance patterns as a function of fluid alkalinity. Hydrothermal fluids from Bulgaria. Modified from Michard and Albarede (1986).

and rock, and it reflects solubility of LREE complexing anions, relative to the HREE complexes. The differing solubilities of the respective complexes appear to express the CO_2 partial pressure and alkalinity of a solution. These jointly control the availability of REE complexing anions in this solution, such as mono- and bi-carbonate.

Empirical observations have led Michard and Albarede (1986) and Michard et al. (1987) to conclude that the abundance of complexing carbonate anions promotes the entrainment of HREE by hydrothermal solutions. Therefore, hydrothermal solutions are characteristically enriched in HREE, relatively depleted in LREE and carry the lowest value of the Ce/Yb ratio. The opposite seems to be true for solutions that carry low concentrations of the complexing anions.

2.2.7. Concluding Remarks

In summary, it is apparent that the infiltration of rainwater cannot explain the chemical and isotopic characteristics of pore fluids from the vadose zone at Yucca Mountain. The only reasonable explanation admits previous invasion(s) of the vadose zone by fluid of clearly hydrothermal origin. These fluids most likely have high alkalinity. They seem to have evolved in the Paleozoic carbonates and other pre-Cambrian rocks, which underlie the tuffs, probably in a high CO_2-partial pressure and low oxygen fugacity environment.

Chapter 2.3. Expected Geologic Expressions

2.3.1. The significance of the two main stages of thermal evolution of the crust

It is an inevitable conclusion of the inferred thermal evolution of the lithosphere that the hydrologic subsystem of Yucca Mountain and surroundings has been evolving during its entire life. This evolution is mainly in response to the changing character and dynamics of heat-transfer dominated dissipative processes. The most important factor, the changing character and intensity of the heat sources, which support hydrothermal processes in the mid-upper crust, has been considered in Chapter 1.3 (Part One).

The inferred changes in the components (advective and conductive) of the total input of heat into the crust (see Figure 1-22) distinguish two main stages of the evolution. Over the earlier stage, the total amount of heat contained in the crust was low relative to that of the subsequent stage. A temperature near and above the solidus (i.e., 600-850°C) was present only locally, within isolated magma chambers. Outside these chambers the temperature was lower than that which prevailed during the subsequent period.

A large but isolated body of magma acts as a "strong" and enduring heat source in support of hydrothermal processes. Heat losses are effectively compensated by convection of magma within the magma chamber. Therefore, a magma chamber is capable of supporting long lasting (of the order of 1 Ma, see for example White, 1974) hydrothermal circulation. Typically, such circulations produce pervasive alteration of the country rock as well as accumulation of economically viable ore deposits within it. Most if not all geologists, whose specialty is hydrothermal ore deposits, are familiar with such accumulations of hydrothermal minerals and their paragenetic assemblages.

During the subsequent stage, however, the advective component of the total input of heat into the crust and, initially, the total amount of heat contained therein were both declining, as shown on Figure 1-22-*c*. Consequently, the previously prominent "strong" heat sources (magma chambers) were increasingly becoming less frequent and intense, and eventually were eliminated completely. This does not mean that all hydrothermal process ceased to be active. The increasing intensity of the conductive component had, and still does, cause a steady rise in the total amount of heat within the crust.

The increasing heat capacity, in turn, results in an increase in the long-term average geothermal gradient. Eventually, the increasing geothermal gradient leads to the development of unstable fluid density configurations, so that the Rayleigh number (R_a) is high. Consequently, values of the amplification factor are potentially high and the growth rates are also high. If a suitable hydraulic conductivity channel is present (low values of the slenderness ratio and the heat transfer grouping), the potentially unstable fluid is free to circulate hydrothermally. If such channel is absent, so that the critical Rayleigh number (R_c) is prohibitively high, there is nothing to prevent the potentially unstable fluid from waiting for a more opportune moment. Eventually, such moment arises either spontaneously (increasing dT/dz and thus increasing R_a) or induced by tectonic action, which may take various forms. Some examples of such forms are seismogenic rupture of a fault, emplacement of a volcanic intrusion, creep of a fault and upward flow of fluid or gas.

Transport of heat within the crust was, and still is, mainly by conduction. As a result, heat sources supporting the late hydrothermal processes, were (and still are) "weak". These weak heat sources can be rapidly depleted and exhausted, as discussed in Murphy (1979). In contrast to "strong" heat sources, the hydrothermally enhanced losses of heat cannot be adequately compensated by slow conduction of heat from the surrounding rocks. Without adequate heating, circulation ceases. Cessation, however, is only transient. It is followed by a period during which heat conduction from the far field slowly restores the heat source.

Thus, it is expected that late hydrothermal processes would be manifested as relatively short-lived plumes, which would remain intermittently active for periods that are measured in terms of millions of years.

Because the intermittent, burst-like, activity is of a relatively short duration, of the order of 0.1 to 1.0 Ka, the resulting mineralization may be expected to be disappointing from a mineral resource perspective. Likewise, the resulting alteration may be expected to be meager (narrow and restricted to the wallrock of the conductive structures, such as faults, fractures, and breccia pipes). This alteration would mostly affect those rocks, which are most susceptible to it, such as a vitric tuff.

From a mineral resource perspective, there is no particular reason to pay attention to such alteration and authigenic mineral assemblages. It is not surprising therefore, that mineral resources-oriented geologists are generally not familiar with them. That these mineral assemblages are different from those formed as a result of circulation supported by magma chambers does not mean they are produced by infiltrating rainwater.

Whilst considering the mineralization of the interior of Yucca Mountain, it is essential to keep in mind that potentially these deposits could be expressing two different stages of evolution of the long-term behavior of the hydrologic subsystem. As emphasized, what sets the early stage apart from the late one is that the former has been motivated by a "strong" heat source, whereas a "weak" source or sources have motivated the latter. The geologic history and the proposed conceptual model jointly require that two thermodynamically distinct hydrothermal processes be imprinted upon Yucca Mountain. These processes must be expected to be associated with a number of sharply contrasting features: the dynamics (i.e., rates of work production in the form of fluid flow); the sources of the dissolved elements; the environments of mineral deposition; and, the spatio-temporal behavior. Thus, it must be expected that the two distinct hydrothermal processes have produced two chronologically and compositionally distinct assemblages of alteration and authigenic hydrothermal minerals. Otherwise the proposed conceptual model is false. Nonetheless, these assemblages are a collective expression of a body ascending within the sub-lithospheric mantle with correspondingly changing melting circumstances and inputs of energy and matter into the Earth's crust.

2.3.2. The Timber Mountain hydrothermal episode

The early heat-transfer dominated dissipative processes in this area are well known, including the early hydrothermal processes at Yucca Mountain. They are expressed in a variety of forms, including: the Timber Mountain Caldera; the fissure rhyolites such as the tuffs of Rainbow Mountain and Cutoff Road; the heavy metal mineralization at Bare Mountain; and, the adularia-sericite alteration. The associated K/Ar ages indicate that these processes were active between 11.0 Ma and at most 9.3 Ma ago, as noted in Noble at al. (1991) for example. This activity is referred to here as the Timber Mountain hydrothermal episode. (Somewhat more detailed review of the hydrothermal alteration episodes will be presented in Part Three, Chapter 3-8).

The early hydrothermal activity lasted sufficiently long to cause pervasive propylitic alteration of the vitric tuffs which at the time comprised Yucca Mountain. Alteration is in the form of propylitic alteration minerals such as zeolite and montmorillonite clays, whose radiometric ages are between about 10 Ma and 11 Ma. The vitrophyres of the Topopah Spring Member are still partially preserved, which indicates that the altering solutions were transported under low-level dynamics (low Rayleigh number, R_a). The corresponding values of the amplification factor and the growth rate were both low, so that the unstable aquifer fluids were confined to a depth below the Topopah Spring Member, seldom if ever reaching the ground surface. (Also, it can be inferred that the dynamics could only have remained low if the partial pressure of the dissolved CO_2 remained low for the entire duration of the Timber Mountain hydrothermal episode.)

It is likely that the immature state of development of the listric faults near the magma body, acting in concert with the body, restricted the early hydrothermal circulation to the uppermost crust. This crust at Yucca Mountain consists of the early Paleozoic marine carbonates underlying up to 3.0-3.5 km of late Miocene alkali tuffs. These overlying and then vitric tuffs must have been exceptionally susceptible to dissolution by the circulating fluids. Hence, it is likely that these tuffs were the main source of the dissolved major cations, suggesting that the circulating fluids have been enriched in sodium and potassium at the expense of calcium and magnesium. It may therefore be expected that the circulating fluids promoted zeolitic alteration of the tuffs, in the form of alkali zeolite such as erionite, phillipsite, and chabazite (Chipera and Bish, 1997). The duration of the thermal exposure of those tuffs, which underlie the Topopah Spring Member, is measured in terms of 100 Ka – 1 Ma or more. This prolonged exposure may be expected to have promoted the transformation of pre-existing (deuteric) smectite into orderly-interstratified clays such as allevardite, kalkberg, and illite (Howard, 1981 and Bish, 1989).

The underlying marine carbonates contain large concentrations of strontium and carbon. By contrast, the overlying alkali tuffs originally must have contained much lower concentrations of these elements. Hence, it is likely that the circulating fluids have acquired most of the dissolved Sr and C from the marine carbonates. Locally, these carbonates serve as a reservoir of inorganic carbon, which is typically enriched in the "heavy" isotope (^{13}C). At the same time, they serve as a reservoir of marine strontium, which is enriched in the non-radiogenic isotope ^{86}Sr. Thus, the circulating fluids may be expected to have been carrying C and Sr isotope signatures, which are similar to those of marine carbonates of an early Paleozoic age. Hoefs (1987) and Burke et al. (1982), respectively, give these values as $\delta^{13}C \sim 0$ ‰ PDB, for carbon, and as $^{87}Sr/^{86}Sr$ between 0.7080 and 0.7092, for strontium. The fluids so enriched may be expected to have produced calcitic veins and cements, which are enriched in ^{12}C but depleted in ^{87}Sr.

The inferred low partial pressure of the dissolved CO_2 implies that the fluids of the Rayleigh-Bénard convection cells acting during the Timber Mountain hydrothermal episode had low alkalinity. This chemistry, in turn, implies a low availability of the REE complexing anions, specifically mono- and bi-carbonate. As noted in section 2.6, empirical observations by Michard and Albarede (1986) and Michard et al. (1987) suggest that this low availability would promote the entrainment of LREE by hydrothermal solutions. Therefore, such solutions are characteristically enriched in LREE, but relatively depleted in HREE, and they carry high relative concentrations of Ce.

Based on the preceding inferences and empirical observations, it may therefore be expected that the early hydrothermal fluids at Yucca Mountain produced calcitic veins, which are enriched in LREE but depleted in HREE. In other words, such veins would be tagged by the chondrite normalized REE pattern, which is characteristic of the host tuffs (see Figure 2-27) and of low alkalinity hydrothermal solutions (see Figure 2-28).

2.3.3. Late Hydrothermal Processes

The proposed conceptual model implies that the Timber Mountain hydrothermal episode was superseded by hydrothermal processes which were (and still are) supported by "weak" heat sources. These processes would be spatially associated with the horst-bounding faults and, temporarily, with the characteristic earthquakes. By contrast to the earlier circulations, the subsequent ones would occur intermittently, from about 8-9 Ma ago up to essentially the present. They would assume the form of short-lived seismic pumping events, which sometimes would be followed by a large-scale amplification of the Rayleigh-Bernard instability, which at a time of the earthquake occurrence would be in a quasi-stable or dormant state.

The chemistry of the late solutions responsible for alteration can be interpreted to be very different to that of the fluids of the Timber Mountain episode. The late hydrothermal activity would involve solutions that were sufficiently warm and saline to cause selective zeolitic alteration. The zeolitic alteration would be restricted to the vicinity of fractures or pervasive in those ash-flow and ash-fall tuffs, which survived the earlier hydrothermal metamorphism and retained a vitric state. The resulting zeolitic minerals would carry K/Ar ages between 8-9 Ma and essentially the present.

However, by contrast to the earlier zeolites, the temporally equivalent montmorillonite clays with higher ordering of interstratification would not be expected to accompany the younger zeolitic minerals. This is because transformation of randomly interstratified smectite into such clays requires a temperature of at least 90-100°C and prolonged thermal exposure, which for the 90-120°C temperature range is of the order of 1 Ma. In addition, the transformation requires an adequate supply of potassium, and elevated concentrations of other cations (such as Ca, Mg, and Na) inhibit the transformation (for details see Hower and Altener, 1983 and Bish, 1989).

The proposed conceptual model implies that the late alteration aureole would be asymmetrically shaped, which would express enhanced penetration of the fluid into the hanging block and into the upper (i.e., closer to the surface) tuffs. This asymmetry would reflect asymmetry of the hydraulic conductivity structure, which is controlled by the *in situ* stress field, with the hanging block and the upper tuffs being more permeable than the footwall and the deeper tuffs are. The expected aureole therefore is thick and deep near the fault, but increasingly thinner and shallower with the increasing distance away from it.

In addition, the conceptual model implies that the late altering solutions were operating under high-level dynamics, so that the rates of work done in the form of fluid and gas flow were high, in particular at "dawn" of the instabilities. There are two reasons for this expectation. First, the instabilities would have occurred in slender conductivity channels around the horst-bounding faults, which requires a relatively high value of the Rayleigh number (R_a). Thus, the corresponding values of both the amplification factor and the growth rate would have been relatively high. Second, the model calls for the involvement of juvenile carbon dioxide under high partial pressure and the dissolution temperature. Thus, the model implies that CO_2-effervescence near the ground surface introduces positive feedback, potentially leading to a hydrothermal explosion.

Both the absence of magma bodies and the high mid-upper crustal strain rates would facilitate the intrusion of meteoric water into the deep interior (10-15 km) of the basement where major fault ruptures nucleate. This basement consists, at Yucca Mountain, of the early Paleozoic marine carbonates and the underlying clastic rocks of early Cambrian and pre-Cambrian age. Thus, relative to the overlying alkali tuffs, the basement as a whole is strongly enriched in alkaline earth elements, calcium and magnesium in particular. The resurgent and strongly CO_2-enriched fluids, equilibrated with the basement, would be strongly enriched in calcium and magnesium, at the expense of sodium and potassium.

The expected chemistry of the late altering solutions contrasts sharply with that of the Timber Mountain hydrothermal fluids. A contrasting chemistry may thus be expected of the respective alteration, in addition to different radiometric ages. The late fluids would be expected to have promoted alkaline earth zeolitization of the vitric and alkali tuffs, in contrast to the earlier alkali metal zeolitization. In addition, these late solutions would have promoted a spatially skewed development of alkaline earth metasomatism of these tuffs. Thus, the late zeolites may be expected to carry K/Ar ages, which are younger than about 8-9 Ma. The zeolites would be calcic, such as heulandite, stellerite, and laumontite (Chipera and Bish, 1997).

The underlying clastic rocks of the basement are abundant in ^{87}Rb, the parent for the radiogenic isotope of strontium (^{87}Sr). These very old rocks produce ^{87}Sr at relatively high rates. At the same time, they have been accumulating ^{87}Sr for a very long period, of the order of billions of years. These rocks, therefore, are typically enriched in radiogenic strontium and carry $^{87}Sr/^{86}Sr$ values between 0.712 and 0.736 or more (Faure, 1987).

The late solutions would have tapped this reservoir of strontium, in addition to being exposed to carbonic liquids and gases advecting from the mantle source. The juvenile CO_2 is characteristically depleted in the "heavy" isotope of carbon (^{13}C) and carries $\delta^{13}C$ values ranging from -5.0 to -8.0 ‰ PDB (Hoefs, 1987). The dissolution of this gas at a temperature of say 200-300°C produces solutions, which are strongly enriched in the "light" isotope of carbon (^{12}C) and carry $\delta^{13}C$ values as low as -12 ‰ PDB. Thus, by contrast to the earlier solutions, the late ones may be expected to have been strongly enriched in the "light" isotope of carbon (^{12}C), in addition to being enriched in the radiogenic isotope of strontium (^{87}Sr). These solutions would have facilitated the precipitation of veins and authigenic cements, which are depleted in ^{13}C but enriched in ^{87}Sr, in contrast to the earlier veins and cements.

2.3.3.1. Role of CO_2 in breccia formation

The proposed conceptual model implies that, at least at the onset of a hydro-tectonic instability, precipitation of the (late) veins and authigenic cements would be accompanied by rapid CO_2 effervescence. As noted earlier (see Figure 1-44), in an ascending and CO_2-dominated fluid, the effervescence commences at a depth, which depends on the CO_2 dissolution temperature and the degree of cooling. This threshold depth may be small, only a few hundred meters. It is likely therefore, that the ^{87}Sr-enriched and ^{13}C-depleted veins are present in the upper tuffs only, and the corresponding bottom depth denotes the onset of CO_2 effervescence. By positive feedback, this effervescence would accelerate the rate of mineral precipitation, particularly of calcite. It should thus be expected that precipitation varied as a function of depth, attaining a maximum at the ground surface and a minimum below.

The texture of hydrothermal calcite reflects the rate of its precipitation, and this texture may hence be regarded as an indicator of the rate of CO_2 effervescence (Archambeau and Price, 1991). The spatially varying rate would cause the abundant and rapidly formed surface deposits to be micritic or cryptocrystalline, and increasingly less abundant and deeper deposits to be macro-crystalline. The latter would then contain fluid inclusions, which would record the temperature, pressure, and chemistry of the parental and plume-forming solutions.

As noted earlier, the rate of CO_2 effervescence could be high enough to cause a hydrothermal explosion, in particular if the initial CO_2 dissolution temperature exceeded 200°C and the rate of cooling is such that the near-surface temperature drops near to ambient levels. Under these conditions, the CO_2 effervescence would commence at a very shallow depth, as shown on Figure 1-44. Here, the lithostatic pressure is very small, of the order of 10-20 bars. At such low stresses, the gas pressure can exceed the overburden load. Explosive fragmentation of the surface tuff would occur if the gas releases were sufficiently fast and voluminous. The compressibility of the gas would allow significant expansion of the gasified fluid without substantial pressure drop. Thus the gas effectively "energises" the fluid responsible for the brecciation. Much greater dilation is possible than can be achieved during brecciation by single-phase fluid. The outcome would be pipe-like bodies of hydrothermal eruption breccia (Nelson and Giles, 1985). The authigenic fraction of the breccia would be isotopically equivalent to the ^{87}Sr- and ^{12}C-enriched veins and cements.

Breccias are present only above the porous, ash-fall tuff. The porous tuff can act as a gas reservoir and the vitric tuff as a seal. This implies that either or both accumulation of a reservoir of compressed gas or pressure build up below a seal may be necessary for brecciation to occur. This observation does not, of course, preclude the role of gas exsolution during the fluid ascent. Exsolution of gas leads to acceleration of the rate of rise of the fluid by reduction of fluid density (positive feedback). This would result in very high rates of pressure rise in surface and near-surface rocks. An extremely rapid rise of pore pressure would be a necessary condition for pervasive fracture initiation and propagation (brecciation). Otherwise, one would expect simply the formation of a limited number of mode 1 fractures aligned normal to the least principal stress. Thus, the observations imply that near-surface seals, gas accumulation and rapid pressure rise combine to induce brecciation of surface and near-surface rocks.

2.3.3.2. Evolution of CO_2-charged aqueous fluids at the ground surface

As a CO_2-charged aqueous plume ascends, and then discharges at the ground surface, it de-gasses, cools, and evaporates. During this dissipation process, the lighter $^{12}C^{16}O_2$ and $H_2{}^{16}O$ molecules preferentially concentrate in the escaping gas and vapor phases. Thus, with increasing residence time at the ground surface, plume-forming solutions would become progressively enriched in the "heavy" isotopes of carbon (^{13}C) and oxygen (^{18}O) by means of an isotope enrichment diffusion mechanism. The evolving stable isotope compositions of the cooling and de-gassing solution would cause gradual changes in the stable isotope composition of the resulting deposits (Gonfiantini et al, 1968; Chafetz and Lawrence, 1994). Thus, surface deposits which have been precipitated from cooling and de-gassing solutions typically show trends of changes of the stable isotopes with changing lithofacies. As shown on Figure 2-11, Hay et al. (1984) identified such trends in spring deposits at Ash Meadows. These trends express the fact that the lowest or "lightest" δ^{18}O and δ^{13}C values are associated with the most proximal lithofacies. However, as these lithofacies become increasingly more distal, in time and space, they become increasingly more enriched in the "heavy" stable isotopes.

Thus, discharges of hydrothermal plumes at the surface of Yucca Mountain may be expected to exhibit trends of changing stable isotope character with changing lithofacies. The lithofacies, which are present at the surface, consist of four types of increasingly more distal (both in space and time) deposits. They include the most proximal authigenic breccia cements, then bedrock veins, followed by calcretes, and, finally, the most spatially and temporarily distal *senso stricto* "pedogenic" carbonates.

The interpretation of these trends represents an important distinction between the conceptual model proposed here and that of Quade and Cerling (1990). The most spatially and temporarily distal *senso stricto* "pedogenic" carbonates, as described above, are similar to the "modern soil carbonates" considered by Quade and Cerling (1990) in their studies of the Trench 14 deposits. Quade and Cerling (1990) assumed *a priori* that these pedogenic carbonates represent the most proximal lithofacies, and that the Trench 14 calcite-silica veins in the bedrock are the distal derivatives. Thus, the interpretation of the isotope-lithofacies trends which is based on the conceptual model presented here, in which the deposits are sourced by subsurface, upwelling fluids, is the complete opposite of the Quade and Cerling (1990) interpretation (which assumes the deposits are sourced by rainfall).

2.3.3.3. Calcite vein REE chemistry

The inferred high partial pressure of the dissolved CO_2 implies that the intermittent fluid circulations, which followed the Timber Mountain hydrothermal episode, were associated with high alkalinity. This chemistry, in turn, implies a high availability of REE complexing anions, specifically mono- and bi-carbonate. Based again on the empirical observations by Michard and Albarede (1986) and Michard et al. (1987), such solutions would be characteristically enriched in HREE, but relatively depleted in LREE, and carry low relative concentrations of Ce.

Based on the preceding inferences and empirical observations, it may therefore be expected that, relative to the earlier Timber Mountain veins, the late hydrothermal fluids would produce calcitic veins, which are enriched in HREE. These veins would therefore be tagged by a chondrite-normalized REE pattern having lesser or even opposite slope to that characteristic of the earlier Timber Mountain veins.

2.3.3.4. Summary of the geologic expressions of hydrothermal processes operating at Yucca Mountain

Table 2-4 summarizes the inferred contrasting expressions of the hydrothermal processes, which, as the proposed conceptual model implies, have been active at Yucca Mountain during its 13 Ma long life span. This table is a road map for focused deciphering of the geologic record, which has been written into the tuffs at Yucca Mountain.

Table 2-4.

Expected forms of expression of hydrothermal dissipation of heat over life span of Yucca Mountain.

Feature	Early circulation	Late circulation
Age	12-11 Ma age.	~ 9 Ma age up to essentially the present
Circulation character	Multipath at a small and nearly constant R_c-R_e difference.	Short-lived plumes at highly variable R_c-R_e difference.
Spatial extent	Between basal vitrophyre of the Topopah Spring Tuff and depth of several km.	Between the ground surface and base of the brittle zone.
Character of solutions (recorded in minerals)	Sodium-potassium, ^{13}C enriched and ^{87}Sr depleted.	Calcium-magnesium, ^{13}C depleted and ^{87}Sr enriched.
Character of country-rock alteration	Pervasive alkalic zeolitization and orderly interstratified montmorillonite minerals (allevardite, kalkberg, and illite).	Fracture-based alkaline earth zeolitization and metasomatism.
Style of country-rock mineralization	Gangue crystalline vein and replacement.	Gangue vein and authigenic cement, micritic at the ground surface but crystalline at depth.
Spatial extent of the mineralization	Ubiquitous below basal vitrophyre of the Topopah Spring Tuff, fracture-based intrusions into the overlying rock possible	Restricted to the ground surface and uppermost tuff.
REE pattern	Overall LREE enrichment.	Tendency toward HREE enrichment.
Auxiliary characteristics of the mineralization and alteration	Depthward increasing of the degree of thermal metamorphism	Hydrothermal eruption breccia, stable isotope gradients indicative of the concurrent cooling and degassing, age populations that are biased in favor of increasingly recent age.

Chapter 2.4. Isotopic and Chronological Implications

2.4.1. Introduction

It is implied that the ESF minerals (calcite, quartz, chalcedony, fluorite, apatite, heulandite, strontianite, and gypsum, some of which contain two-phase inclusions), represent work done during one or more hydrotectonic instabilities. The proposed conceptual model explains most if not all of the observed characteristics of these minerals. There is no alternative, coherent and contradiction-free explanation for the ESF minerals. (This will be discussed in more detail in Part Three.)

The model admits both ascending and descending flows in the vadose zone. It predicts a temporal and spatial evolution of the chemistry of the fluids and of the depositional environment. The model requires that the mineral-forming solutions be composed of two end-members giving rise to a spatiotemporally varying and diluting mixture. One end-member consists of reducing solutions that were accumulating inside the dilating hypocentral region of the Paintbrush fault zone before their introduction into the vadose zone. This fluid was transported by seismic pumping. The other end-member consists of more oxidizing and mainly fracture-based solutions, which were residing in the regional aquifer (aquifer fluids) (i.e., outside the conductive channel). The diluting solutions consist of infiltrating rainwater.

The sharply different environments to which the two end-members were exposed would result in contrasting stable and radiogenic isotope properties. The time- and space-varying mixing would produce composite solutions associated with spatiotemporally changing stable and radiogenic isotopic composition. Most likely, these varying properties would have been inherited by the precipitated minerals and be expressed as isotope gradients along the mineral growth directions. Based on this conceptual model, it may be inferred that such gradients are preserved in the ESF mineral aggregates, and, furthermore, that they are intrinsic to most, if not all, of these mineral aggregates. These gradients, and radiometric age gradients in particular, do not necessarily indicate exceedingly low rates (of the order of mm per Ma) of mineral growth, as Neymark et al. (1998) have suggested. Nor does it follow that they indicate prolonged cooling of Yucca Mountain (of the order of 5 Ma), after the Timber Mountain hydrothermal episode, as Whelan et al. (2001) have suggested. Instead, in our opinion, these gradients provide a powerful basis for validating the proposed conceptual model, thus resolving the lingering upwelling water controversy.

2.4.2. Isotopic characteristics of the parental solutions from which the ESF minerals were derived

In this section, we wish to focus attention on four isotopic characteristics of the parental solutions, which are particularly important from a genetic and chronological perspective. The first is the δ^{13}C-value in the solutions, the interpretation of which is required to infer the origin of the dissolved carbon dioxide. The second is the $^{207}Pb/^{204}Pb$ ratio in the solution, which is required for estimating the common lead correction and then for calculation of $^{207}Pb/^{235}U$ age for the corresponding host mineral. The third and fourth characteristics are the $^{234}U/^{238}U$ and $^{230}Th/^{238}U$ activity ratios of the solutions. These are required for calculation of $^{230}Th/^{234}U$ age of the corresponding host mineral and for inferring the origin of the parental solution.

2.4.2.1. Inferred isotope chemistry of fluids involved in seismic pumping

The model implies that the fluid, which is transported upward by seismic pumping, accumulates for a long time (~ 10 Ka) in a reducing environment in the basement at a depth of say 10-15 km. This fluid would therefore be very hot ($T > 300°C$) and would contain dissolved carbonic gases, such as CO_2 and CH_4, the concentration of which would increase with time. Because the fugacity of oxygen would be very low, the dissolved carbon would likely occur as two chemical species having sharply contrasting oxidation states. Isotopic fractionation between these species would lead to a situation whereby reduced species (methane; oxidation state +4, which would not be involved in the later precipitation of $CaCO_3$), would concentrate the "light" isotope (^{12}C). By contrast, the oxidized dissolved species (CO_2; oxidation state –4, which would later take part in precipitation of calcite), would be enriched in the "heavy" isotope (^{13}C). This mechanism was described in detail by Ohmoto (1972) and Rye and Ohmoto (1974).

It is likely that the fluid would be in a state of secular equilibrium at the time of the initial entrapment inside the dilating hypocentral region. This means that initially the activity of all daughter nuclides would be close to that of the parent. Initially, the $^{234}U/^{238}U$ activity and $^{207}Pb/^{204}Pb$ ratios would therefore be equal to those of the rock, or close to about 1 and about 15, respectively. Dilation at 10-15 km depth would cause the rock-fluid interfacial (contact) area to be large and redox potential to be low. Thus, it may be inferred that the solubility of uranium would be low during the accumulation period, but that of lead would be high. It may also be expected that those daughters of ^{238}U and ^{235}U, which are produced

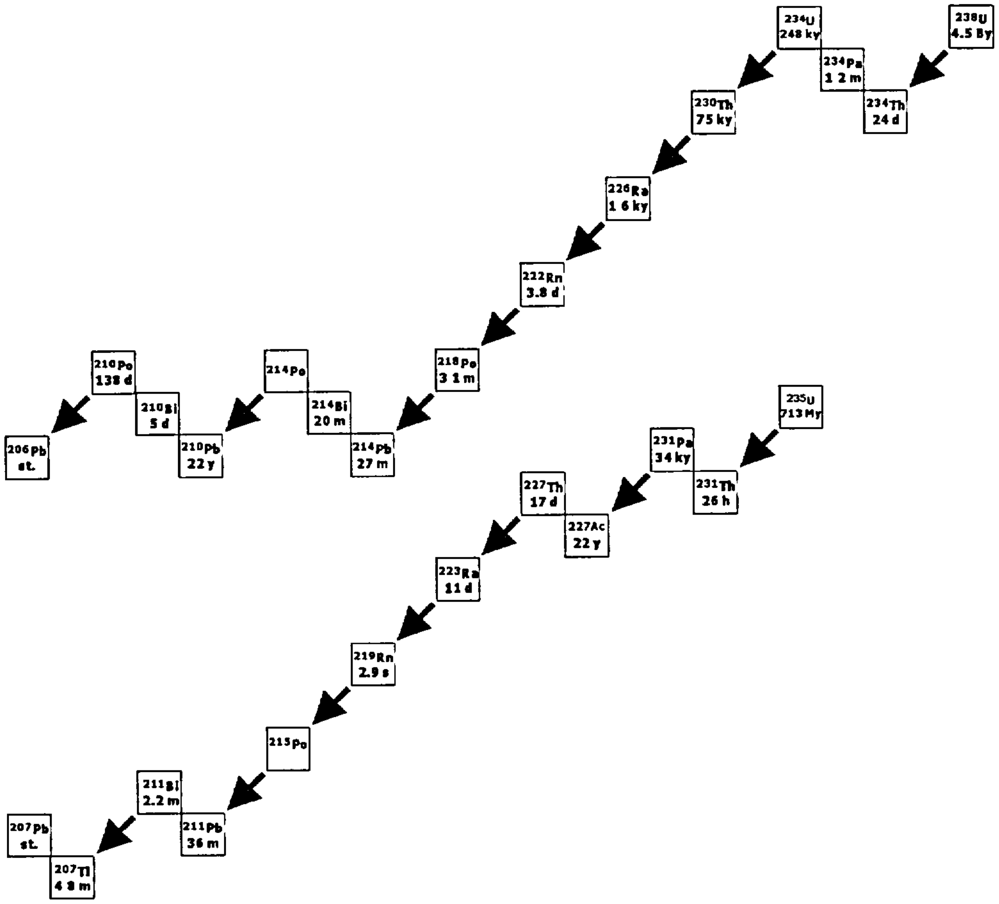

Figure 2-29. Decay chains for ^{238}U and ^{235}U. Arrows indicate α-decay.

with the emission of α-particles (see Figure 2-29), would be ejected into the fluid by means of the α-recoil mechanism. Likewise, it may be expected that the gaseous nuclides, ^{219}Rn and ^{222}Rn in particular, would diffuse into the accumulating fluid.

This open-system behavior would decrease the ^{238}U/(daughters of ^{238}U) and ^{235}U/(daughters of ^{235}U) ratios in the accumulating fluid, which then would tend to increase the ^{234}U/^{238}U and ^{230}Th/^{238}U activity ratios. However, the decrease of ^{238}U/(daughters of ^{238}U) ratio would only slightly affect the ^{234}U/^{238}U activity ratio. This is because the α-recoil of only one nuclide ($^{238}\text{U}\rightarrow{}^{234}\text{Th}$) would affect this ratio. Further, the solubility of U would be low, so that a meaningful increase in the ^{234}U concentration ($^{234}\text{Th}\rightarrow{}^{234}\text{U}$) would be hindered by low mobility of the excess uranium. Acceleration of the isotope exchange reaction between the fluid and the surrounding rock, by the elevated temperature, would further hinder significant increase of the ^{234}U/^{238}U activity ratio. This inferred tendency of the ^{234}U/^{238}U activity ratio to remain unchanged is supported by actual measurements of this ratio in samples of geothermal fluids and deep-seated brines. Typically, $^{234}U/^{238}U$ ratio for such fluids varies between 1 and about 3 (e.g., Osmond and Cowart, 1982, Zukin et al., 1987, Halloway et al., 1989).

The decrease of the ^{238}U/(daughters of ^{238}U) ratio would increase the magnitude of the $^{230}Th/^{238}U$ ratio to a greater extent. This is because the ^{238}U abundance would be low and, in this case, two α-recoils (i.e. ^{238}U and ^{234}U) would affect this ratio. Because the solubility of thorium is generally low, this effect would likely be small. Nonetheless, it would be meaningful because the greater-than-zero value of the $^{230}Th/^{238}U$ ratio would appreciably increase the apparent $^{230}Th/U$ age of a mineral, the calculation of which involves the assumption that the initial input of ^{230}Th is zero.

The decreasing ^{235}U/(daughters of ^{235}U) ratio, which the proposed conceptual model implies, produces another very important effect in the accumulating fluid. This effect involves the $^{207}Pb/^{204}Pb$ ratio, which enters $^{207}Pb/^{235}U$ age calculations of a mineral phase as a value for the common lead correction. An erroneously assumed value, in particular values, which are too low, would cause the calculated $^{207}Pb/^{235}U$ age to be older than the true age. The potential for assuming an erroneously low correction value arises because, in contrast to the solubility of U, the solubility of Pb is relatively high in a low-redox environment. As a consequence of this relatively high solubility, the ^{207}Pb daughter, which would be introduced into the accumulating fluid by decay of several short-lived daughters of ^{235}U (see Figure 2-29), could remain in this fluid as a dissolved species. These short-lived nuclides could diffuse into a dilatory opening as a gaseous phase (^{219}Rn)*, or they would be ejected into the fluid via the α-recoil mechanism (^{231}Th, ^{231}Pa, ^{227}Ac, ^{219}Ra, etc.). This inferred behavior of the ^{207}Pb daughter would progressively increase the $^{207}Pb/^{204}Pb$ ratio in the accumulating fluid from about 15 to some higher value. Although the $^{207}Pb/^{204}Pb$ increase would most likely be small, the ^{207}Pb abundance that is not supported by the ordinary lead (^{204}Pb) abundance could be meaningful in the sense of having a potential to distort (increase) $^{207}Pb/U$ age of the precipitated mineral. This is because the overall Pb solubility would be high, as a result of the oxygen fugacity being very low, so that small increases of the $^{207}Pb/^{204}Pb$ ratio (i.e. common lead correction for the fluid) could translate to fairly significant amount of the dissolved ^{207}Pb.

Thus, the proposed conceptual model implies that when the characteristic earthquake occurs, the hydrotectonically mobilized fluid would have a low $^{234}U/^{238}U$ activity ratio (ca. between 1 to 3) and a non-zero ^{230}Th abundance. This fluid would carry a decreased ^{235}U/(daughters of ^{235}U) ratio and a $^{207}Pb/^{204}Pb$ value, which is somewhat higher than that in the aquifer rocks and in the equilibrated with them oxidizing fluids. Thus, the ratios to be expected on the basis of the proposed conceptual model are meaningfully (from the geochronological perspective) different from those, which are implied by the Yucca Mountain

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* Numeric analysis of the diffusion of different Rn isotopes into the fluid is presented in Part Three, Chapter 3-7.

Project rainwater model for precipitation of the controversial mineral aggregates found in the ESF tunnel (see Part Three, Chapter 3-4 for description). The Yucca Mountain Project rainwater model implies the following ratios for the mineral forming fluids: ^{230}Th = 0, ^{207}Pb/^{204}Pb~15, and activity ratio ^{235}U/(daughters of ^{235}U)$\rightarrow$1 (i.e. approaching the secular equilibrium value).

2.4.2.2. Inferred isotope chemistry of aquifer fluids

The aquifer fluids owe their stable isotope and radiogenic isotope chemistry to exchange and mixing reactions occurring in a high redox potential (oxidizing) environment. This contrasts with the fluid associated with the seismically induced flow discussed in previous section. In the aquifer, the dissolved carbon species are mixtures of atmospheric carbon, biogenic carbon (acquired in soil horizon at a time of the recharge), inorganic carbon (dissolved from the Paleozoic carbonates), and carbon acquired through dissolution of advecting juvenile CO_2. The corresponding δ^{13}C value may be estimated based on analyses of samples of the aquifer waters.

The δ^{13}C values for shallow water in the Ash Meadows discharge area are between -3.8 and -7.3‰ PDB, and between -7.0 and -13.1‰ PDB for deep groundwater at Yucca Mountain (U.S. DOE, 1988). These values indicate that, prior to near surface CO_2-degassing, the aquifer fluid carries strongly negative δ^{13}C values, say around -10‰ PDB. Thus, it is evident that the aquifer fluid differs from the fluid which the model implies is subject to seismically induced flow and which would carry oxidized carbon species that are substantially equilibrated with the reduced species (most likely CH_4) and which, therefore, would be strongly enriched in the "heavy" isotope (^{13}C).

Because the redox potential is high (oxidizing), it is also reasonable to infer that the Pb solubility would be low. This low solubility would cause the ^{207}Pb/^{204}Pb ratio to remain about equal to that in the aquifer rocks, which is between 15.55 and 16.01 (Zartman and Kwak, 1993). Thus, the aquifer fluid could be associated with the common lead correction (^{207}Pb/^{204}Pb ratio), which is somewhat smaller than the one implied by the proposed conceptual model for fluid subject to seismic pumping.

Furthermore, it is likely that the aquifer fluid would carry the dissolved uranium in the form of the uranyl ion, which has +6 valence. In this oxidation state, the uranium solubility is relatively high (e.g., Zielinski and Rosholt, 1978). This high solubility allows the ^{234}U/^{238}U ratio to vary between 1 and 8 or more. This variability reflects the spatial and temporal variability of the input of ^{234}U from the aquifer rock into the aquifer fluid. In addition, the relatively high abundance of the dissolved uranium, coupled with the very low solubility of thorium, may be expected to keep the ^{230}Th abundance in the fluid very low, so that ^{230}Th/^{238}U $\rightarrow$ 0.

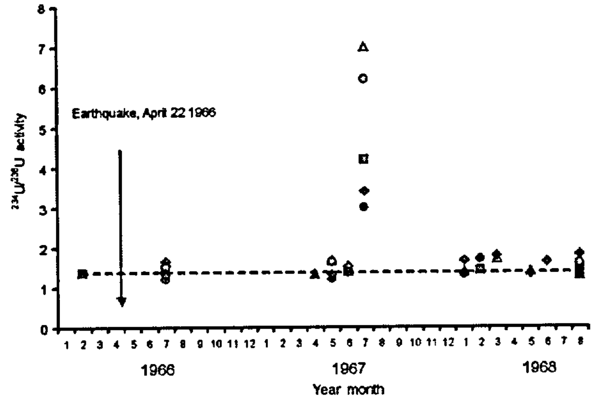


Figure 2-30. Variations of $^{234}\mathrm{U}/^{238}\mathrm{U}$ in waters of the Tashkent artesian basin that followed the Tashkent Earthquake, Uzbekistan (April 22, 1966). Data from 10 hydrogeological boreholes. Due to relatively slow water movement in the basin, the "response" to the earthquake was delayed by 15 months. The earthquake was also accompanied by increasing contents of He (more than 10-fold), U, and F dissolved in waters. By Zverev et al. (1980).

2.4.2.3. Observations on groundwater radiochemistry

Limited observations of radio-chemical effects of earthquakes, around ruptured faults elsewhere, suggest that some earthquakes be preceded and followed by increased values of the $^{234}\mathrm{U}/^{238}\mathrm{U}$ ratio in aquifer fluids. An excellent example of such an increase is shown in Figure 2-30. The $^{234}\mathrm{U}/^{238}\mathrm{U}$ activity ratio in waters of the Tashkent artesian basin (Uzbekistan) increased from a background value of ~1.4 to as much as 6-8 (in different boreholes) 15 months after the large earthquake that virtually destroyed the city of Tashkent on April 22, 1966. The increase was preceded and accompanied by increasing amounts of He (more than 10-fold), U, and F dissolved in waters (Zverev et al., 1980). Similarly, Dolidze at al. (1975) reported seismically induced increases in $^{234}\mathrm{U}/^{238}\mathrm{U}$ activity ratios in thermomineral waters of Georgia (Caucasus), with the highest recorded value being 14.

This increase in the $^{234}\mathrm{U}/^{238}\mathrm{U}$ activity ratio is commonly taken to indicate a transient increase in the supply of $^{234}\mathrm{U}$ nuclides, which is attributed to enhanced dilation of fractures prior to, during, and some time after the occurrence of larger earthquakes. An earthquake is believed to affect the radiochemistry of fluids in a two-fold manner.

On one hand, the co-seismic dilatancy of shallow rocks (not to be confused with the dilatancy involved in nucleation of large NDC earthquakes) increases the rock-fluid contact surface area. This has the effect of increasing the rate of direct input of $^{234}\mathrm{Th}$ nuclides into aquifer fluids via the α-recoil mechanism, which accompanies the $^{238}\mathrm{U}\rightarrow{}^{234}\mathrm{Th}$ radioactive decay. The transferred $^{234}\mathrm{Th}$ nuclides shortly thereafter decay into $^{234}\mathrm{U}$ nuclides, which have a significantly longer half-life (see Figure 2-29).

On the other, the dilation has the effect of exposing previously produced $^{234}\mathrm{U}$ "hot" nuclides, or so-called Szilard-Chalmers atoms, to leaching by aquifer fluids. By contrast to the $^{238}\mathrm{U}$ parent, the "hot" $^{234}\mathrm{U}$ nuclides are more vulnerable to leaching. This is because the $^{238}\mathrm{U}\rightarrow{}^{234}\mathrm{Th}\rightarrow{}^{234}\mathrm{U}$ decay (see Figure 2-29) involves α-particle emission, ejection of which has the effect of damaging the crystal lattice around the former sites of the $^{238}\mathrm{U}$ parents, making the "hot" nuclides more prone to the leaching.

Thus, the proposed conceptual model and the preceding remarks jointly imply that, in contrast to the ^{230}Th/^{238}U ratio, ^{234}U/^{238}U ratio in aquifer fluid would not remain the same over duration of a single evolutionary loop of the system. Instead, this ratio fluctuates, attaining a maximum (about 8-10) immediately before, during, and some time after the end of an evolutionary loop (i.e., time of characteristic earthquake) and a minimum (about 1-3) thereafter.

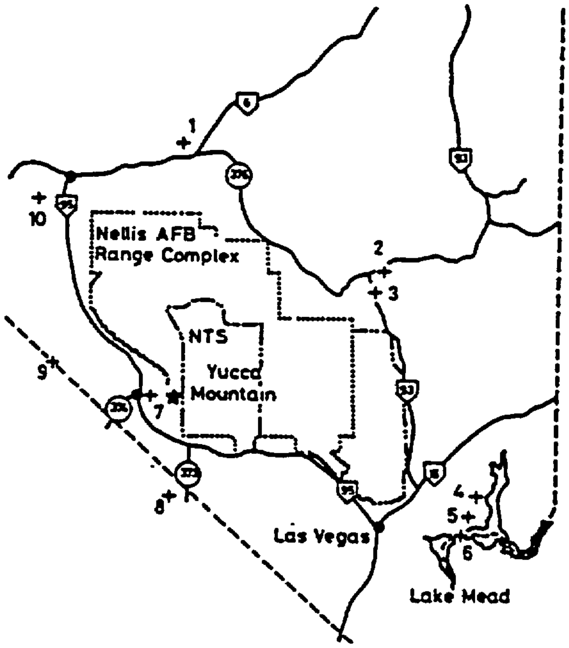


Figure 2-31. Spring samples. The numbers correspond to the sample numbers shown in Tables 2-5 and 2-6. From Holloway et al. (1989).

Thus, also in terms of the initial ^{234}U/^{238}U and ^{230}Th/^{238}U ratios, the aquifer fluid would be distinct from fluid that would be contained in the seismically induced flow. The former would carry ^{234}U/^{238}U ratio between 3 and 8 and ^{230}Th/^{238}U$\rightarrow$0, whereas the latter would carry ^{234}U/^{238}U less than ~3 and ^{230}Th/^{238}U value that is meaningfully more than zero. These differences are important for converting the measured (present-day) values of these ratios into the ^{230}Th/U ages of minerals. Erroneous assumptions of the initial ^{230}Th/^{238}U and ^{234}U/^{238}U values, too low in particular, would cause the calculated ^{230}Th/U age to be older than the true age.

It is particularly important to note that the inferred variability and interdependence of ^{230}Th/^{238}U and ^{234}U/^{238}U ratios in warm or hot fluids are both supported by measurements carried out on actual samples of such fluids. In this regard, Holloway et al. (1989) carried out radiochemical measurements of the concentrations of thorium and uranium isotopes in waters of hot and/or mineral springs near the Nevada Test Site. The sampling area is shown on Figure 2-31 and results of the analyses are presented in Tables 2-5 and 2-6.

Tables 2-5 and 2-6 show that low abundance of the dissolved ^{238}U (e.g., 90 fCi·l^{-1} for Warm Spring with T = 63°C) tends to be accompanied by relatively high abundance of the dissolved ^{230}Th. The ^{230}Th/^{238}U = 0.105 for this water. Concurrently, the corresponding relative abundance of the dissolved ^{234}U tends to be low, with the ^{234}U/^{238}U ~ 2.0. By contrast, the increased abundance of the dissolved ^{238}U (e.g., 1500 fCi·l^{-1} for Crystal Spring with T = 32 °C) tends to be accompanied by negligible abundance of the dissolved ^{230}Th, so that the ^{230}Th/^{238}U$\rightarrow$0. Concurrently, the abundance of the dissolved ^{234}U tends to be higher, so that the ^{234}U/^{238}U = 3.5.

Table 2-5

Thorium and uranium isotopes in natural waters in the vicinity of the Nevada Test Site, fCi·l^{-1}±2σ (from Holloway et al., 1989)

No	Sample	Temperature, °C	^{230}Th	^{232}Th	^{234}U	^{238}U
1	Warm Spring	63	9.5±1.5	4.3±1.0	185±27	90±17
2	Crystal Spring	32	<1	<1	5200±400	1500±100
3	Ash Spring	36	2.0±0 8	<1	2960±220	1150±90
4	Blue Point Spring	28	12±2	8±1	3030±150	980±50
5	Rogers Spring	28	9.5±2.4	9±2	3300±170	1040±50
6	Lake Mead, Drivers Cove	-	14±2	16±2	2700±530	1460±300
7	Bailey's Hot Spring	42	2±1	<1	11 100±800	3500±260
8	Badwater, Death Valley, California	-	35±6	60±9	1300±100	1020±85
9	Mineral Spring at Scotty's Castle	-	13±2	1.6±0.4	5300±1140	2720±600
10	Mineral Spring near Silver Peak, Nevada	21	33±9	23±7	15 900±1700	10 900±12(

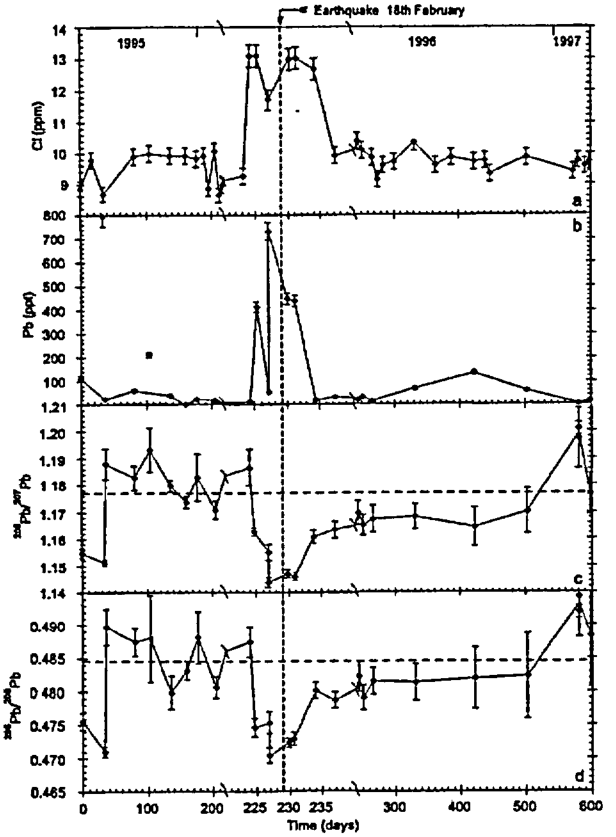
It is interesting to note that temporal changes in the lead radiochemistry of aquifer fluids, similar to those that the proposed conceptual model implies, were actually observed in association with larger earthquakes. On example of such observations is shown on Figure 2-32.

Figure 2-32 shows the effects of an earthquake with magnitude of 5.2, which occurred in the eastern part of the French Pyrenees on the 18 February 1996. The effects are depicted in the form of time series for abundance of the dissolved Pb as well as for the ^{206}Pb/^{207}Pb and ^{206}Pb/^{208}Pb ratios. In addition, the figure shows a cross-plot between ^{208}Pb/^{206}Pb and ^{207}Pb/^{206}Pb. Both the time series and the cross-plot

Table 2-6

Activity ratios of thorium and uranium isotopes in natural waters in the vicinity of the Nevada Test Site, (Ci·Ci^{-1})±2σ (from Holloway et al., 1989)

No.	Sample	^{230}Th/^{232}Th	^{234}U/^{232}Th	^{234}U/^{238}U
1	Warm Spring	2.2±0.6	43±12	2.1±0.5
2	Crystal Spring	-	-	3.5±0.3
3	Ash Spring	-	-	2.6±0.3
4	Blue Point Spring	1.5±0.3	379±53	3.1±0 2
5	Rogers Spring	1.1±0 4	367±84	3.2±0.2
6	Lake Mead, Drivers Cove	0.9±0 2	169±41	1.8±0.5
7	Bailey's Hot Spring	-	27800±14200	3 2±0.3
8	Badwater, Death Valley, California	0.6±0.1	21.7±3.7	1.3±0.1
9	Mineral Spring at Scotty's Castle	8.1±2.3	3313±1093	2.0±0.6
10	Mineral Spring near Silver Peak, Nevada	1.4±0 6	691±221	1.5±0.2



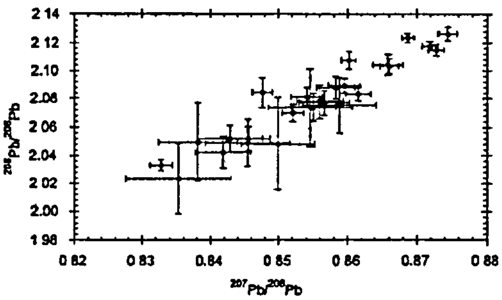


Figure 2-32. *Left:* Time series for the Alet spring (East Pyrenees, France) waters showing seismic-related geochemical anomalies: *a* – chloride content; *b* – lead content; *c* – $^{206}Pb/^{207}Pb$; and *d* – $^{206}Pb/^{208}Pb$. The horizontal dashed line represents the mean baseline values outside anomaly. *Above*: $^{208}Pb/^{206}Pb$ *vs.* $^{207}Pb/^{206}Pb$ cross-plot showing linear trend (r = 0.95) suggesting simple mixing process involving waters with distinct Pb isotope compositions. From Poitrasson et al. (1999).

imply that, as the time of the earthquake approaches and thereafter, the ^{207}Pb abundance significantly increased. This is precisely what the proposed conceptual model implies.

For example, if a hypothetical mineral were deposited from pre- or post-earthquake "baseline" water (horizontal dashed line in Figure 2-32), the common lead correction* for the $^{207}Pb/^{235}U$ dating would have been smaller than the correction that would have been appropriate for a mineral that was formed from the "earthquake-time" fluid (say, days 225-230 in Figure 2-32). Subsequently, if the "baseline" correction was used for calculating the $^{207}Pb/^{235}U$ age of the "earthquake" mineral, the resulting calculated age would appear substantially older than the true age.** Although the original authors (Poitrasson et al., 1999) attributed the ^{207}Pb contamination of the aquifer waters to an anthropogenic source, the effect is just the same as the one, which the model implies.

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* Correction accounting for the ^{207}Pb lead trapped by a mineral from the fluid during crystallization. This "allogenic" ^{207}Pb, which was not formed by *in situ* decay of ^{235}U contained in the mineral, must be extracted before calculating radiometric age of a mineral; otherwise the apparent age will be erroneously large.

** We want to emphasize that this hypothetical example is merely an illustration of the ways and means of how the varying ^{207}Pb contents in the mineral-forming fluid could affect the results of the radiometric dating. Obviously, due to very long half-life of ^{235}U, the $^{207}Pb/^{235}U$ method cannot be used for dating the modern waters or mineral precipitations.

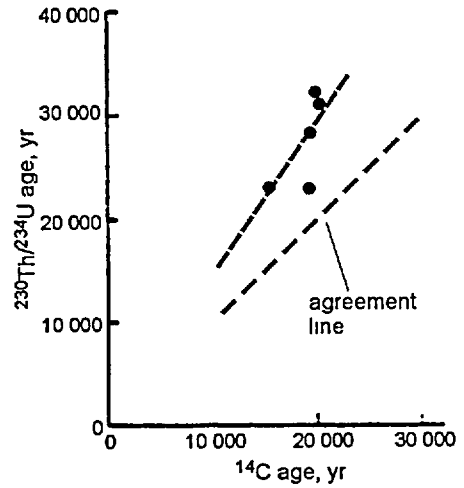

Figure 2-33. Comparison of ^{230}Th/U and ^{14}C ages for samples of carbonate pebble coatings from Eagle Mountain, California. From Schlezinger (1985).

Linear regression is: y = 1.468x + 61.8. Exact agreement would show slope of 1.0.

More and more examples of U daughter contamination at zero actual age are cited in the literature. These examples underline the limitations of U-Th and U-Pb dating of very young geologic materials. For example, dating of two growing speleothems, less than 100 years old, using both ^{230}Th/^{234}U and ^{231}Pa/^{235}U decay chains, yielded near-concordant, but incorrect, apparent ages of 2 and 3 Ka (Whitehead et al., 1999). It is evident in this case that both ^{230}Th and ^{231}Pa had been added to the speleothems, which is contrary to the fundamental assumption of ^{230}Th/U dating method. Another example of the contamination, in this case most likely by detrital ^{230}Th, is evident in results of studies of radiometric ages of coexisting samples of surface carbonates in the area of Eagle Mountain of California (Schlesinger, 1985). These studies included age dating of the same specimens by ^{14}C and ^{230}Th/U methods. The results are shown in Figure 2-33 and Table 2-7.

Figure 2-33 shows that the ^{230}Th/U ages are consistently biased, relative to the corresponding ^{14}C ages, in the direction of older age. The difference is as large as 50 %, so those carbonates with ^{13}C age of say 20 Ka yield a ^{230}Th/U age of 30 Ka. In addition, although the ^{14}C age remains relatively constant, the corresponding ^{230}Th/U ages increase, as shown on the figure, which suggests that varying amounts of ^{230}Th have been added over short period. The result is that a rapidly formed carbonate appears, in the light

Table 2-7

^{14}C and ^{234}U/^{230}Th ages and isotope properties from samples of the Eagle Mountain indurated calcic horizons (from Schlezinger, 1985)

Depth interval, cm	Bulk density, g cm^{-3}	$CaCO_3$, %	$\delta^{13}C$, ‰ PDB	$\delta^{18}O$, ‰ SMOW	^{14}C age, yr	^{230}Th/^{234}U age, yr
50-57	1.76	7.41±1.12	-6.3±0.7	-30.6±0 5	15 040±1500	23 00±3000
90-100	1.74	6.07±1.07	-6.6±0.4	-30.6±1.0	19 260±80	28 250±5000
125-135	1.70	12.04	-5.1±0.8	-32.2±0 8	19 090±90	23 000±2000
100-110	1.60	4.71	-5.9±0.3	-29.7±0.3	20 140±150	31 000±4000
135-155	1.86	3 99	-6.9±1.0	-30 0±0.8	19 770±520	32 000±4000

of ^{230}Th/U data, of having been formed over much longer period and at an earlier time.

The fundamental problem with the U-daughters-based dating is that the daughters are numerous, they are transported by multitude of mechanisms, their solubilities are poorly known in general, and there are only a few studies of their relative abundance in groundwater. For example, it has usually been assumed that the chemistry of Th and Pa are so similar that they are not likely to be deposited in excess of one another. However, this is increasingly known to be untrue. The Th/Pa ratios vary significantly in various ocean and continental waters, see for example Anderson et al. (1982 and 1983), Simpson et al. (1984), Shimmield et al. (1986), and Lao et al. (1993). Ivanovich and Alexander (1987) have found that in limestone groundwater ^{230}Th/^{232}Th ratio varies between 1 and 1000. It is generally considered that transport of Th would be by colloids (e.g., Langmuir and Herman, 1980). However, other researchers propose that carbonate complexes maybe responsible for Th mobility in natural waters (e.g., Dervin and Faucherre, 1973; Simpson et al., 1984; and Osthols et al., 1994). These complexes are believed to contribute to abnormal Th solubility in alkaline lakes (e.g., Anderson et al., 1982; and Simpson et al., 1984).

2.4.3. Significance of the U-Pb Radiochemistry as a tool for distinguishing between the conceptual models

In spite the obvious limitations, the Yucca Mountain radiochemistry database is potentially very useful, provided it is utilized with due caution and full awareness of its limitations. This is especially true in the case of U-series disequilibrium data from the ESF opal. In addition to having a chronological application (by allowing distinguishing opals that are less than 0.5 Ma old), these data cast serious doubts on the validity of a hypothesis, which explains the presence of opal among other minerals by deposition from percolating rainwaters (Paces et al., 1996; Neymark et al., 1997; Marshall et al. 1998).

Paces et al. (1996) have established that the specimens of uraniferous (U-abundance up to 250 ppm) opal have ^{234}U/^{238}U ratios between 1.0 and 9.5 (Figure 2-34).

Some of the samples, the data for which are shown on Figure 2-34, coat surfaces of high-angle fractures, so that if they were formed from infiltrating rainwater, the residence time for this water must have been very short. In addition, some of the samples represent the base, or the interior of mineral aggregates, which may be up to 10-20 mm thick. At the same time, these fracture-based opals yield ^{230}Th/U ages of about 300 Ka, as shown on Figure 2-35.

The observed variability of the ^{234}U/^{238}U ratio provides grounds for questioning the rainwater hypothesis. The variability in particular begs a question: how is it possible for the rapidly infiltrating fluid

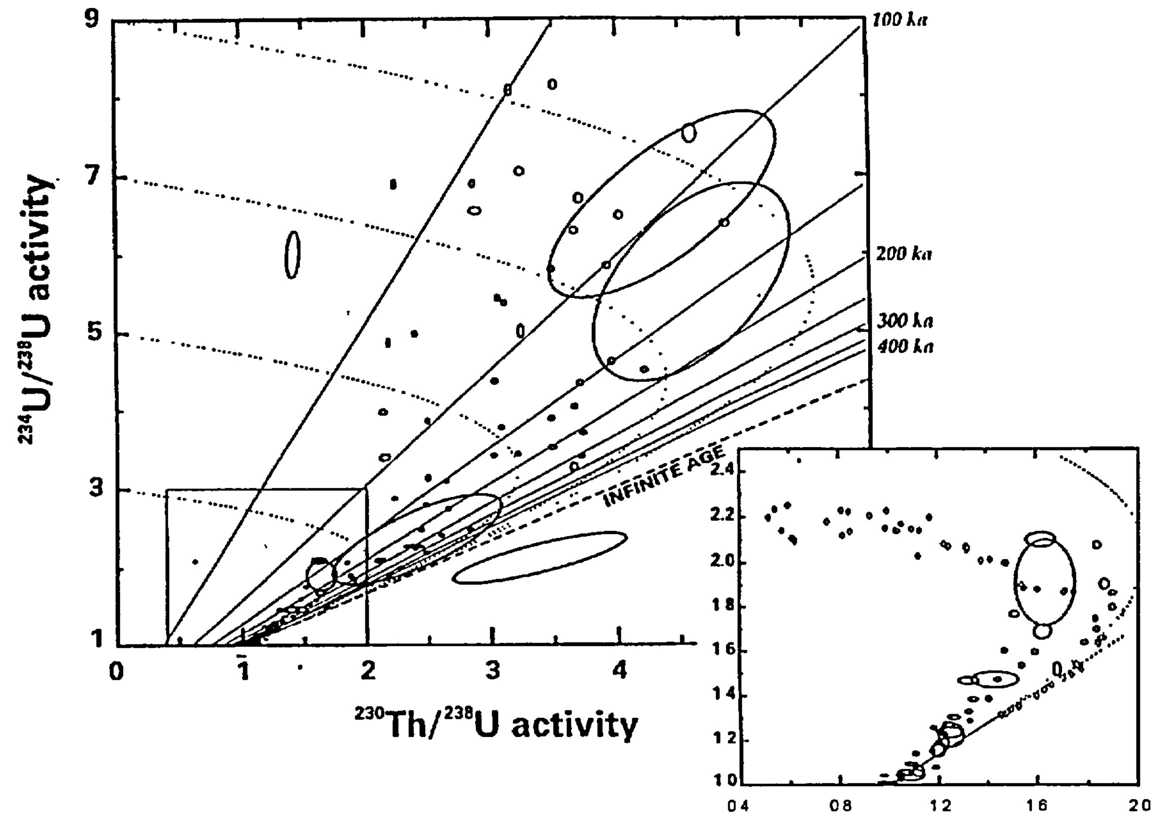

Figure 2-34. Uranium-series disequilibrium evolution diagam showing data for 97 analyses of ESF calcites and opals. Ellipses correspond to 2σ error limits. Dotted, curved lines represent loci of ^{234}U/^{238}U and ^{230}Th/^{234}U isotopic compositions that will develop through time in a material with initial ^{234}U/^{238}U values of 3, 5, 7, and 9 is that material behaves as closed system. Compositions of calcites from the saturated zone at Devils Hole (Ludwig et al. 1992; Szabo et al. 1994) representing deposition for the last 550 Ka are shown for comparison (open diamonds on inset). Reproduced from Paces et al. (1996).

to acquire, along a flow path of only 30-100m*, the high and variable values of ^{234}U/^{238}U activity ratios which are shown on Figure 2-34? This question is particularly appropriate in the light of U isotope data shown on Figure 2-36.

Figure 2-36 shows ^{234}U/^{238}U values plotted against concentration of the uranium dissolved in groundwater in the northern and eastern parts of the Nevada Test Site. Here, the piezometric head is less than the corresponding potentiometric head and the groundwater has a bicarbonate-sodium-potassium composition, so that one may be reasonably certain that the fluids represent downward flow through the tuffs. The cross-plot on Figure 2-36 provide strong evidence for the tendency of the ^{234}U/^{238}U ratio to remain stable. In spite of the factor of ten changes in the U-abundance, this ratio remains within a narrow range, between 2.7 and 5.0. In addition, there is a faint suggestion that the increasing U-abundance, which

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* The depth from ground surface to the sample collection sites.

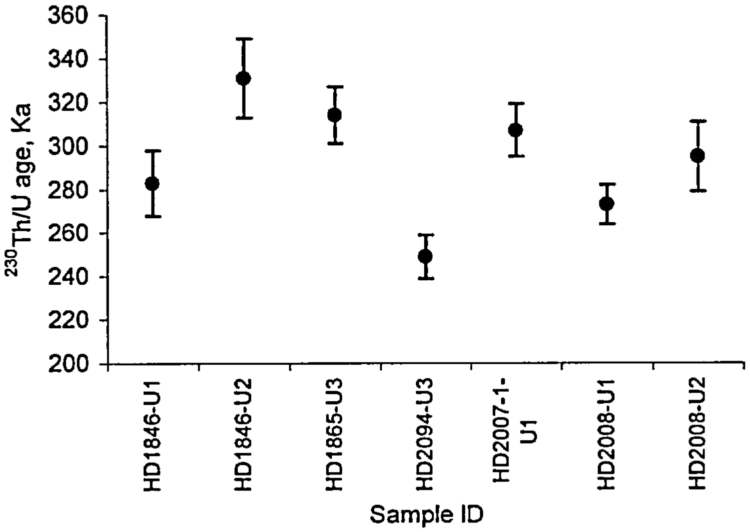

Figure 2-35. ^{230}Th/U disequilibrium ages for base (early) and internal layers of opal and chalcedony from the first 2000 m in the ESF. Description of samples: HD1846-U1 and –U2 - massive botryoidal silica; HD1865-U3 - chalcedony/opal: massive, milky silica; HD2094-U3 - opal: thin botryoidal film directly on fracture surface; HD2007-1-U1 - opal; interior layer; and HD2008-U1 and U2 - opal: older, large, individual opal hemisphere. Based on the data from Paces et al. (1996).

presumably expresses the increased residence time, has the effect of slightly decreasing the ^{234}U/^{238}U value. Thus, the question is why, at Yucca Mountain, this value has been changing so much over the past 500 Ka, as Figure 2-34 suggests?

In the context of the rainwater-source hypothesis, there is no rational answer to the preceding question. This demonstrates the falsehood of this hypothesis. Of course, one might attempt to rescue this hypothesis by proposing, more lightheartedly than seriously, that the variability reflects variability of the fluid residence time in fractures. The latter would express fluctuations of the infiltration flux in concert with the climatic fluctuations. If such were indeed the case then how can the tendency of the initial ^{234}U/^{238}U values at Devil's Hole to remain stable (see insert on Figure 2-34) be explained? The latter is believed to be a discharge area for the Yucca Mountain hydrologic subsystem and has been shown to preserve an almost 500 Ka-long climatic record in the carbon

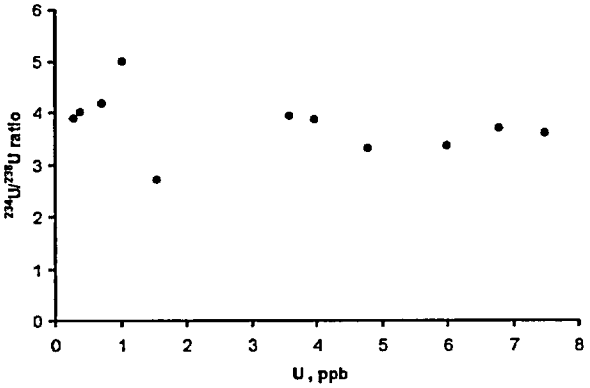

Figure 2-36. Correlation between the content of U and the ^{234}U/^{238}U ratio in groundwater in the volcanic aquifer recharge area at the Nevada Test Site. Data from Zelienski and Rosholt (1978).

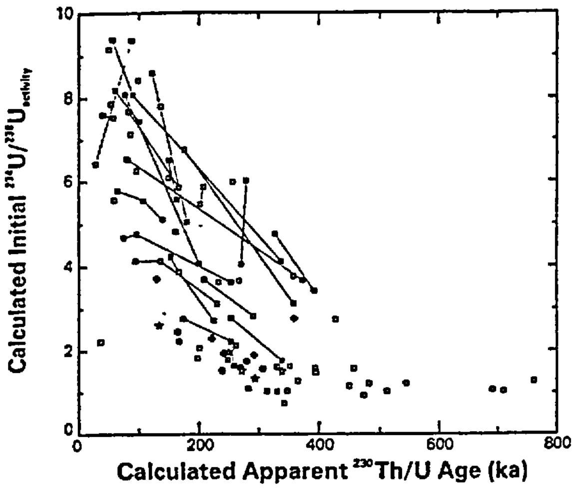

Figure 2-37. Calculated $^{234}U/^{238}U$ initial activity ratios in the ESF calcites and opals plotted against calculated apparent $^{230}Th/^{234}U$ ages. Multiple subsamples from the same occurrence are either connected by lines or shown with similar symbols other than squares. From Paces et al. (1996).

and oxygen isotopes of deposited calcite (Coplen et al., 1994; see also Figure 2-10). Additionally, how can a monotonic increase of the calculated initial $^{234}U/^{238}U$ values with a decrease of the calculated apparent ^{230}Th/U age be explained? (Figure 2-37).

A rational answer to the last question is suggested by Figure 2-8. This figure shows the increase of the $^{234}U/^{238}U$ activity ratio, as a function of the declining intensity of the upward flux, which is implied by the observations at the LWD and CFW discharge sites (see section 1.7). This figure illustrates the fact that, in a context of the proposed conceptual model, the temporal variability of $^{234}U/^{238}U$ ratio is not only the expected but also an essential expression of the underlying hydrotectonic processes. The observed variability reflects the evolution of the radiochemistry of the fluids, which have comprised the LWD-CFW hydraulic mound in the past, toward the present-day radiochemistry, in response to changing sources of the fluid and changing physico-chemical environment.

It thus becomes apparent that the proposed conceptual model provides a rational explanation for the U-series data on Figure 2-37, which shows that the calculated initial $^{234}U/^{238}U$ ratios increase systematically, as the calculated apparent ^{230}Th/U ages diminish. The implications are that the calculated apparent ^{230}Th/U ages, which are shown on this figure, do not reflect the corresponding actual ages. The result is that two different specimens with the same actual age appear, based on the ^{230}Th/U ages, as having been formed at different times, with both being older than the actual age. This illusion is schematically illustrated on Figure 2-38.

The most important isotope implications of the proposed conceptual model are summarized in the form of inferred changes of $\delta^{13}C$, $^{207}Pb/^{204}Pb$, $^{234}U/^{238}U$, and $^{230}Th/^{234}U$ values as a function of the corresponding changes in the local potentiometric head. These inferred changes are shown on Figure 2-39, as a function of time elapsed after the characteristic earthquake, or after the ending of a single evolutionary loop of the geologic/hydrologic system.

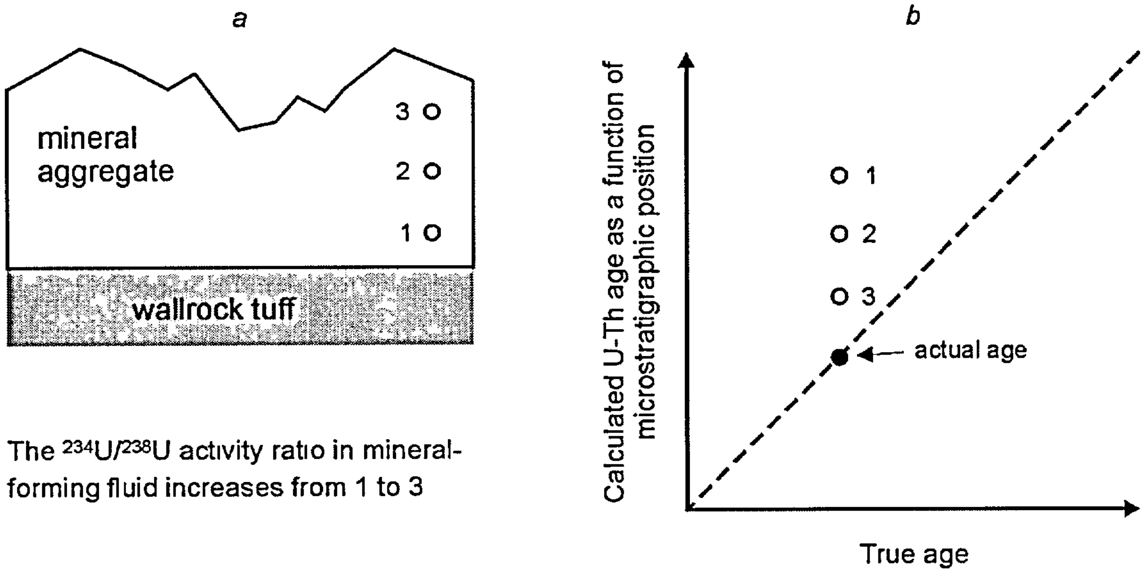

Figure 2-38. Schema, illustrating how calculated U-Th ages might appear to change systematically in accord with the microstratigraphic position of dated material (*a*) in a sample that was formed "instantaneously" (*b*).

Figure 2-39 shows that, as the seismically and thermally induced hydraulic mound forms and then recedes, the isotopic compositions of the fluids, which make up this mound, evolve in a predictable manner. The isotope evolution expresses the time-variability of the physico-chemical environment controlled by the volumetric ratio of the reducing fluid, which is associated with the seismically induced flow, to the oxidizing fluid, which is associated with flow induced by a Rayleigh-Bénard instability. This ratio would decrease from some large value at the onset to near zero at the recession.

As the inferred hydrotectonic mound matures and then recedes, the $\delta^{13}C$ values of CO_2 dissolved in the mound-forming fluid would decrease from very "heavy" to very "light", which reflects the diminishing fractionation between dissolved CO_2 and CH_4 as the fluid grades from strongly reducing to an oxidizing state. In addition, the decreasing $\delta^{13}C$ values would be expressing gradual restoration of the influence, which the biogenic and igneous sources exert in controlling the isotope properties of the dissolved CO_2.

Concurrently, the $^{234}U/^{238}U$ ratio would increase from about 1-3 up to about 8, which expresses the diminishing involvement of thermal solutions jointly with the dilation-induced mobilization of the "hot" ^{234}U atoms. In addition, the overall involvement of the reducing fluid, diminishing with time, would gradually decrease the $^{230}Th/^{238}U$ ratio to a very small value.

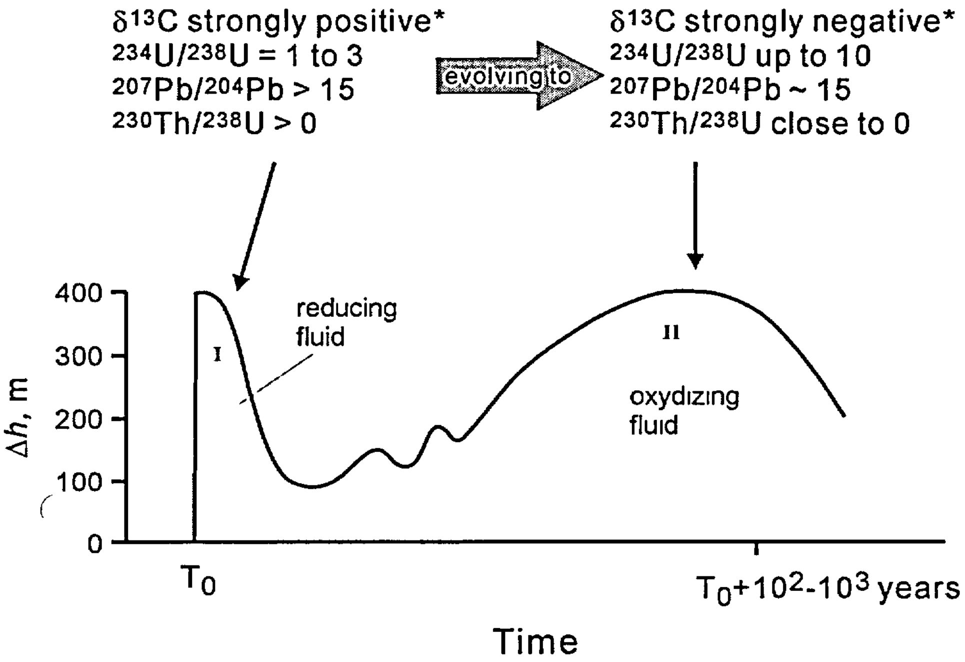

Figure 2-39. Inferred isotope ($\delta^{13}C$, $^{207}Pb/^{204}Pb$, $^{234}U/^{238}U$, and $^{230}Th/^{238}U$) evolution of the mound-forming fluid, which would act as a parent fluid for the ESF minerals.

Δh - variations of the local (within the mound) water table; I – water level rise induced by seismic pumping; II – water level rise induced by Rayleigh-Bernard instability; * - $\delta^{13}C$ refers to the CO_2 dissolved in the fluid and/or to the carbonate deposited from it (reduced carbon species in the fluid, e.g. methane, may have strongly negative values of $\delta^{13}C$).

The gradual increase of the $^{234}U/^{238}U$ ratio would be accompanied by gradual lowering of the $^{207}Pb/^{204}Pb$ ratio from relatively high values to about 15 and a gradual decrease of the Pb-abundance to a smaller value. Both of these declines express the diminution with time of the influence of the reducing fluid.

2.4.4. Concluding Remarks

Thus, it is evident that the proposed conceptual model has important implications concerning interpretation of the stable and radiogenic isotope properties of authigenic minerals at Yucca Mountain, including the ESF mineral aggregates. The following two points summarize these implications with reference to the $\delta^{13}C$, $^{234}U/^{238}U$, and $^{207}Pb/^{204}Pb$ isotope ratios from the ESF mineral aggregates.

On the one hand, the model implies that significant decreases in the $\delta^{13}C$ ratio, from very "heavy" values to later very "light" values, have been imparted to the ESF calcite over a very short time span, on the order of 100 to 1000 years. The decreases can be expected to have a fractal (scale-independent) expression. In other words, the amplitude of the $\delta^{13}C$ decrease, at the scale of a crystal, would be similar to that of the spatial but temporarily equivalent composite (i.e., at a scale of the ESF tunnel). Both of these decreases would tend to have a broadly monotonic character. They would be large enough to be distinct from the known climatic signals. Typically, time-series for the latter have a fluctuating (back and forth) character and amplitudes that are measured in terms of a few ‰, as shown on Figure 2-10. Thus, the utilization of the ESF $\delta^{13}C$ values for the purposes of reconstructing the palaeoclimate (e.g., Paces et al., 1996; Whelan and Moscati, 1998) seems to be a futile undertaking.

On the other hand, the conceptual model implies that the $^{230}Th/U$ and $^{207}Pb/^{235}U$ ages from the Yucca Mountain opals should be taken with more than just a grain of salt. This is because the dated opals (formed from colloidal solutions) would have trap the U, Pb and the U-daughters from fluid whose radiochemistry would be varying during a time span as short as say 100-1000 years. This means that the opals have acquired time-variant amounts of U, Th and Pb isotopes and, therefore, varying values of the initial $^{234}U/^{238}U$, $^{230}Th/^{238}U$, $^{207}Pb/^{204}Pb$, and ^{235}U/(daughters of ^{235}U) ratios. Thus, the conceptual model implies that the geochemical system of the Yucca Mountain opals is in violation of fundamental assumptions of $^{230}Th/U$ and $^{207}Pb/^{235}U$ dating methods*.

Scavenging from a fluid whose U-radiochemistry varies would have the effect of causing the higher opal to contain significantly less of the seemingly *in situ* produced ^{230}Th, relative to micro-stratigraphically lower opal. The actual abundance of *in situ* produced ^{230}Th in the lower opal, however, would be almost the same as that in the higher opal. The discrepancy, translated into differing apparent ages, represents an illusion, which is solely a result of assuming that both of the opals had the same values of the initial $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ ratios.

For the dating method, involving $^{207}Pb/^{235}U$ and $^{206}Pb/^{238}U$ systematics, additional complications arise due to the interference of radon isotopes (^{219}Rn and ^{222}Rn) that migrate via diffusion into the cavities in which minerals (calcite and silica minerals) grow and decay to respective stable lead isotopes (Pashenko and Dublyansky, 2002). This radiogenic lead, genetically related to uranium residing in the rock, surrounding cavity, become adsorbed on the colloidal silica and, upon coagulation and sedimentation of the latter become incorporated in opal. This mechanism leads to substantial distortion of the conventionally calculated U-Pb ages. The mechanism will be discussed in detail in Part Three, Chapter 3-7). The $^{207}Pb/^{235}U$ ages, calculated using "conventional" method (i.e., without taking account of the Rn-derived Pb isotopes) would lead the unaware observer to erroneous conclusions. One such conclusion

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* These dating methods require that correction for the presence of initial (non-*in situ* formed) daughter nuclides D_0 "*...is a constant whose value...either assumed or calculated from data for cogenetic samples of the same age ... It must be possible to assign a reliable value to D_0*" (Faure, 1986). The D_0 is definitely not a constant in the described system and the assignment of the reliable value to it is problematic.

would be that a mineral sequence, which in reality has been precipitated over a short period, has been precipitated over a much longer time span.

In the Yucca Mountain studies the ^{207}Pb-^{235}U ages have already lead to a number of conclusions that would be extraordinarily difficult to justify. For example, Neymark et al. (2000) concluded that the deposition rates for the minerals studied in the ESF ranged "*... between about 0 035 and 1.8 mm/m.y. ... These values are in general agreement with long-term rates of mineral deposition during the past 10 m.y.*" Such slow deposition rates do not look possible from the point of view of the theory of crystal growth. They would require the maintenance of ultra-low supersaturations over millions of years (this subject will be discussed in detail in Part Three, Chapter 3-7). Also, the joint consideration of the ^{207}Pb/^{235}U ages and fluid inclusion data resulted in the conclusion that the vadose zone at Yucca Mountain was conductively heated for a several millions of years (Whelan et al., 2001). Besides being unsupported by the extensive body of geologic and mineralogical evidence, this conclusion does not seem to be possible from a thermodynamic point of view (see discussion in Part Three, Chapter 3-8).

In the absence of reliable data, these impossibilities serve as "quality assurance" checks by which means the presence of *non in situ* produced ^{207}Pb or ^{230}Th may be ascertained.

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Suitability of the Yucca Mountain Site to Accommodate a permanent Repository for High-Level Radioactive Waste and Spent Nuclear Fuel: an Independent Assessment

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