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**DIALOGS BY JERRY SZYMANSKI REGARDING THE
YUCCA MOUNTAIN CONTROVERSY FROM
DECEMBER, 1990 TO MARCH, 1991**

VOLUME I

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SPECIAL REPORT Submitted to the
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Assembled by:

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**Dialogs by Jerry Szymanski Regarding the Yucca Mountain Controversy from
December, 1990 to March, 1991**

Volume I

REPORT ORGANIZATION

PART 1

"Stable Isotopic Evidence for a Pedogenic
Origin of Carbonates in Trench 14 near
Yucca Mountain, Nevada"

J. Quade and T.E. Cerling

PART 2

Letter to *Science*

J. S. Szymanski

PART 3

Letter to Editor of *Science*

T. E. Cerling and J. Quade

PART 4

Letter to Cerling

J. S. Szymanski

PART 5

News and Comment, *Science*

E. Marshall

PART 6

Letter to Szymanski

T. E. Cerling and J. Quade

PART 1

**"Stable Isotopic Evidence for a Pedogenic Origin of
Carbonates in Trench 14 near Yucca Mountain, Nevada"**

**Stable Isotopic Evidence for a Pedogenic Origin of
Carbonates in Trench 14 near Yucca Mountain, Nevada**

JAY QUADE AND THURE E. CERLING

ert soil atmosphere (3). Plants fractionate C along three differing metabolic pathways: C_3 , C_4 , and CAM. The C_3 plants, including conifers, mountain shrubs, and some grasses, make up virtually all of the biomass at higher elevations in the southern Great Basin. The $\delta^{13}C$ of these plants average -24 to -25 per mil at high-elevation sites, and coexisting soil carbonate has a $\delta^{13}C$ higher than in this biomass by 14 to 16 per mil (7). At lower elevations, C_4 and CAM (8) plants, as well as many C_3 shrubs and herbs, are present; here, C_4 plants are the most abundant, and they have an average $\delta^{13}C$ of -13 per mil. A larger proportion of atmospheric CO_2 , with an average $\delta^{13}C$ of -6 per mil, is present deep in soils at low elevations because of lower plant respiration rates. This C reservoir further increases the $\delta^{13}C$ of soil CO_2 —and therefore also of soil carbonate—that is derived from the mix of C_3 and C_4 plants. Holocene-age soil carbonates from each of the major vegetation zones therefore display a distinct $\delta^{13}C$ signature (Fig. 2). For example, the $\delta^{13}C$ of soil carbonates average -9 per mil in the ponderosa pine zone (>2400 m) and -7.4 ± 0.8 per mil (15 samples) in the pinyon-juniper-sage zone (1800 and 2300 m). Near sea level in Death Valley, the $\delta^{13}C$ of soil carbonates is -2 to $+2$ per mil, reflecting the contribution of C_3 creosote and C_4 desert holly shrubs, as well as some mixing of atmospheric CO_2 deep in the soil (3). The $\delta^{18}O$

Fig. 1. The fracture system filled with layered calcite and silica, exposed on the south wall, east end of Trench 14. The trench wall is about 3.5 m high.

Table 1. The ^{14}C dates (in ^{14}C years before present) by accelerator mass spectrometry on pedogenic carbonate from three (SM-2, 3, 4) soils in the Spring Mountains, 100 km south of Yucca Mountain (6). For each sample, we scraped 50 to 100 mg of material from clast undersides, taking care not to include any carbonate from the host clast or from older cements. Further details on soil sites, designated by these field numbers, is in (3).

Lab no.	Field no.	Age	Soil depth (cm)
AA-3697	SM-3(B)	1210 ± 50	15
AA-3698	SM-2A	1900 ± 60	Surface
AA-3699	SM-2B	3820 ± 55	10 to 15
AA-3700	SM-2C	1785 ± 65	30 to 40
AA-3701	SM-4	820 ± 50	50 to 60

composition of soil carbonate also decreases with elevation (Fig. 3), irrespective of parent material, primarily because the $\delta^{18}O$ composition of rainfall also decreases with increasing elevation. Evaporation or differing penetration of meteoric water seasonally also may influence the $\delta^{18}O$ of pedogenic carbonates (3).

We sampled five Holocene-age soils in the vicinity of Trench 14. The local vegetation is dominated by C_3 shrubs such as black bush (*Coleogyne ramosissima*) and Nevada joint fir (*Ephedra nevadensis*); creosote (*Larrea divaricata*), burrobrush (*Ambrosia dumosa*), and twin fruit (*Menodora spinescens*) are less abundant. Shadscale (*Atriplex confertifolia*), a C_4 shrub, is also common. All the herbs, and some of the grasses (mainly *Stipa* sp. and *Oryzopsis hymenoides*), are C_3 plants. Fluffgrass (*erioneuron pulchellum*), a C_4 grass, grows in the early summer. This mix of C_3 and C_4 plants, and moderate plant respiration rates, produces $\delta^{13}C$ values in soil carbonate of -4.2 to -7.1 per mil (14 samples) below a depth of 50 cm. The $\delta^{18}O$ (PDB) of the same carbonates nearly all fall between -7 and -10.6 per mil.

We analyzed 22 samples from most of the major veins exposed in Trench 14. The $\delta^{13}C$ of all but one of samples ranges from -6.3 to -7.7 per mil (average -7.0 ± 0.4 per mil, Fig. 2). The Trench 14 carbonates have low $\delta^{13}C$ values compared to those in nearby Holocene-age soil carbonate; these low values indicate that the Trench 14 carbonates did not form in equilibrium with the modern vegetation in the area. However, the C isotope data do overlap with values from modern soils about 750 m above the site, where cover is dominated by pinyon (*Pinus monophylla*), juniper (*Juniperus osteosperma*), and sagebrush (*Artemisia tridentata*). Samples from this vegetation zone yielded a $\delta^{13}C$ average of -7.4 ± 0.8 per mil. A single sample from Trench 14 yielded a result of -4.6 per mil, a value that is

Fig. 2. The $\delta^{13}C$ (PDB) of pedogenic carbonate in modern soils developed on volcanic parent materials along elevation transects in the southern Great Basin. The linear regression (shown) for 39 soil analyses is: elevation (in meters) = $(554.4 \pm 103.8) - (174.98 \pm 18.3)z$, where z is the $\delta^{13}C$ (PDB) of soil carbonate. The $\delta^{13}C$ compositions of all but one sample from Trench 14 are lower than that for carbonate in Holocene-age soils in the vicinity but overlap with the $\delta^{13}C$ range of soil carbonate ~ 750 m higher on the transects.

observed in modern soils nearby. The other 21 samples from Trench 14 display a narrow range of $\delta^{18}O$ (PDB), between -10.5 and -11.8 per mil (average of -11.3 ± 0.3 , Fig. 3). Again, this is close to the values for modern soil carbonate in the pinyon-juniper-sage zone (average -11.5 ± 1.4 per mil) and is unlike nearby modern soils at -9.8 ± 1.2 per mil (9).

Evidence from fossil pack-rat middens indicates that vegetation zones were displaced about 1,000 m downward during the last full-glacial period (16,000 to 19,000 years ago) (4). Juniper, sagebrush, and pinyon then dominated the vegetation at Yucca Mountain. Our evidence indicates that the Trench 14 carbonates, if formed in soils, would have precipitated in equilibrium with this plant assemblage during the glacial maxima. The $\delta^{18}O$ of ground water in the region during glacial times was 1.5 to 2.0 per mil less than that of modern ground water (10, 11); this is close to the 1.5-per-mil average difference we observe between carbonates in Trench 14 and nearby modern soils.

A further test of a soil versus spring origin comes in comparing the C isotopic composition of coexisting carbonate and occluded organic matter. The difference should be 14 to 16 per mil in soils with relatively high respiration rates, including those found under pinyon-juniper-sage cover (7). The Trench 14 carbonates contain 0.12 to 0.18% organic C (Table 2). The average difference between carbonates and occluded organic matter was found to be 14.6 ± 0.2 per mil (3 samples), which supports a pe-

water may also be in equilibrium with locally derived plant CO_2 , as HCO_3^- is in soil water.

Other evidence also supports a pedogenic origin for the Trench 14 carbonates. The morphology (2) and petrography of the carbonates and silica fillings are consistent with a soil origin, as is micromorphological, clay mineralogical, trace element (18), and isotope tracer (19) evidence. Oxygen isotopes from the silica cements indicate that the temperatures of formation were $\sim 15^\circ\text{C}$, consistent with that in a pedogenic environment (2).

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8. CAM plants, which include cactus and most yucca, are present at lower elevations but don't exceed 10% of the biomass at any site. They average ~ 18 per mil in $\delta^{13}\text{C}$.
9. We observed that the $\delta^{13}\text{C}$ (PDB) and the $\delta^{18}\text{O}$ (PDB) of carbonates appear to be constant below about 40-cm depth in modern soils at this general elevation (3). Above 40 cm, mixing with the atmospheric CO_2 causes progressive enrichment up to the soil surface. All the Trench 14 samples come from 0.9 to 3.4 m below the surface, and display no systematic trends with depth. All our modern soil carbonates that we used to fingerprint isotopically each of the vegetation zones come from 0.5 to 2.2 m depth.
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20. We thank M. Mifflin, I. Winograd, and L. Benson for their useful discussions and help, and R. J. Johnson for his logistical assistance. Analyses were performed at the University of Utah and Yale University. This work was supported primarily by the Nevada Nuclear Waste Project Office, and by National Science Foundation grant BNS-8703304.

25 June 1990; accepted 3 October 1990

PART 2

Letter to *Science*

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COMMENTS ON THE QUADES AND CERLING PAPER "STABLE ISOTOPIC EVIDENCE FOR A PEDOGENIC ORIGIN OF CARBONATES IN TRENCH 14 NEAR YUCCA MOUNTAIN, NEVADA," DECEMBER 1990 ISSUE OF SCIENCE

Please be advised that, after publication in December 1990, issue of Science of paper by Quade and Cerling entitled, "Stable Isotopic Evidence for a Pedogenic Origin of Carbonates in Trench 14 near Yucca Mountain, Nevada," I have been interviewed by a number of local newspaper reporters. When I was asked to comment on the scientific validity of the main conclusion, which is the per descensum pedogenic origin of the Yucca Mountain calcite-silica deposits, my response was blunt "nonsense." When I was asked about the reasons for this seemingly harsh, and most certainly not politely expressed viewpoint, my response was "these guys are a bit short on isotopes and a bit short on a file." Considering that the paper has been published in the highly respected scientific journal, I feel obligated to explain and justify my viewpoints; in a nut shell, this is the reason for this letter.

Analyses of isotopic data derived from samples representing the Yucca Mountain calcite-opaline silica deposit, performed for the purposes of determining the origin of these deposits, may be viewed, perhaps simplistically, as similar to searching for an identity of an individual that left his fingerprints at a crime scene. For such a search to be reliably successful, two essential requirements must be satisfied. First, the encountered fingerprints must meet certain minimum requirements with regard to the state of their preservation. Second, the available fingerprint index, or file, must contain a copy of fingerprints obtained from the individual that is being searched for.

For the Yucca Mountain deposits, a fairly detailed isotopic fingerprint may be constructed by considering the isotopic characters of uranium, strontium, carbon, and oxygen contained in these deposits. The necessary data are already available, and there is no compelling reason to exclude these data from considerations. The isotopic fingerprint that has been constructed solely on the basis of isotopic characters of stable carbon and oxygen is somewhat "fuzzy" and, for the purposes of identifying parent sources for both of the elements, requires a fair deal of interpretation and speculation. This is the message that I was trying to convey through my comment "these guys are a bit short on isotopes."

The isotopic file which, in my opinion, is absolutely essential for the performance of reliable interpretations of the intraformation residence time and history for the parent fluids for the Yucca Mountain deposits, based on

JAN 18 1991

-2-

the stable carbon and oxygen isotopic fingerprint, should be fairly complete. At a minimum, such a file should include the following elements: (a) the isotopic characters of carbon and oxygen contained in the local subsurface fluids, with both short and long intraformation residence time; (b) the isotopic characters of carbon and oxygen contained in the local subsurface fluids, with both simple and complex intraformation residence histories; (c) the isotopic characters of carbon and oxygen contained in deposits related to local deep-seated springs; (d) the isotopic characters of carbon and oxygen contained in deposits related to known (but not assumed) perched bodies of fresh-meteoric fluids; and (e) the isotopic characters of carbon and oxygen contained in veins associated with epithermal ore deposits, etc.

In arriving at their conclusion, Quade and Cerling considered only two elements of the isotopic file, namely: (a) the isotopic characters of carbon and oxygen contained in samples of the local cobble calcareous encrustations, so-called per descensum pedogenic deposits; and (b) the isotopic characters of carbon and oxygen contained in an incomplete suite of samples from the Ash Meadows spring deposits. Ignoring uncertainties that are associated with circumstances of formation of the calcareous encrustations, in my opinion, the employed deduction practice, at a minimum, may be faulted for not recognizing the principal of Aristotelian logic which states "if A is not always B then C, although included in B, does not have to be A." This is the message that I was trying to convey through my comment "these guys are a bit short on a file."

Enclosed please find both the carbon-oxygen isotopic fingerprint and the corresponding isotopic file, both that may be used in establishing identity of the parent fluids for the Yucca Mountain deposits. The oxygen-18 data considered by Quade and Cerling, if combined with similar data from the Yucca Mountain vadose zone, may be employed in performing very revealing reconstructions of the geothermal circumstances of formation of the local calcite-silica deposits. I have performed such reconstructions and have taken the liberty of including the reconstruction results in the enclosed package.

While reviewing the enclosed material, please note the following points: a) at Yucca Mountain, the calcite-silica veins, that contain the oxygen-18 and carbon-13 isotopic signals comparable to those from the surficial deposits, are known to occur down to a depth of at least 670m, Figures 5 and 9; b) the Yucca Mountain calcite-silica deposits exhibit the strong carbon-13 isotopic affinity with carbonate gangue veins from gold-bearing hydrothermal ore deposits, Figure 4; c) the Yucca Mountain calcite-silica deposits carry the same values of the $\delta^{13}\text{C}$ ratio as those expected for deposits produced from the local geothermal fluids, Figure 6; d) the Yucca Mountain calcite-silica deposits exhibit the oxygen-18 isotopic affinity with the local deep-seated spring deposits, Figure 8; e) the Yucca Mountain calcite-silica deposits carry the same values of the $\delta^{18}\text{O}$ ratio as those expected for deposits produced from the local geothermal fluids, Figure 12; f) the precipitation (crystallization) temperatures, for the Yucca Mountain surficial deposits, may

JAN 18 1991

-3-

have been in a range from 15 to ~ 53° Celsius, Figures 25 through 27; and g) values of the paleo-geothermal gradient, estimated based on the observed Yucca Mountain $\delta^{18}\text{O}/\text{dz}$ gradient, are a factor of 1.5 - 2.5 greater than those observed at the present time, Figure 41. All of the above observations, considered either individually or together, lead to an unequivocal conclusion that the Yucca Mountain calcite-silica deposits were formed via the per ascensum process, i.e., from upwelling geothermal fluids. An independent, but again convincing, verification and validation of this conclusion may be derived from considerations of uranium and strontium isotopic data. These considerations, as performed by me in a soon-to-be-released report, indicate that the isotopic characters of uranium and strontium contained in the Yucca Mountain calcite-silica deposits are similar to those dissolved in the local geothermal fluids. Because both the uranium isotopic data and the strontium isotopic data were not considered by Quade and Cerling, I did not include these data in the enclosed package.

Hopefully, based on the review of the enclosed material, you and other scientists associated with the Science journal may agree that labeling the conclusions reached by Quade and Cerling as "nonsense," although not a most polite way of expressing my viewpoint, is justified. The viewpoints expressed in this letter and in the enclosed attachments, are solely my own opinions. These viewpoints neither reflect the U.S. Department of Energy's official position nor opinions held by the majority of contractors associated with the Yucca Mountain Site Characterization Project Office. Should you or your associated scientists express an interest and desire, I stand ready to discuss the remaining isotopic and other data and facts that have a bearing on the matter of origin of the Yucca Mountain hydrogenic deposits.

In closing, the U.S. Department of Energy is responsible for a satisfactory performance of unprecedented task of developing a safe and socially acceptable high-level nuclear waste repository. Consequences that may result from an unanticipated and sharply adverse performance of the repository are of truly catastrophic proportions. While executing this difficult and very important task we must keep in mind our responsibilities to future generations and insist on a cool judgment and utmost sound science. Our decision must not and, if I can help, will not be based on questionable science and "wishful thinking." Should you have any questions regarding this letter or desire further clarifications, please do not hesitate to contact me.

Sincerely,

Jerry S. Szymanski

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P.S.

Analyses of the isotopic data from the Nevada Test Site, and from the Yucca Mountain calcite-opaline silica deposits, lead to a number of very profound conclusions. These conclusions bear upon a thermodynamic nature of the Nevada Test Site geodynamic system. Specifically, the local isotopic data offer a rare opportunity to demonstrate that this system exhibits:

(a) self-organization; (b) periodic and/or non-periodic motions;
(c) "structural instability," expressed as relatively recent "sink->source" and saddle->"sink" ("source") transformations. All of these three aspects of behavior of a dynamic system indicate that one is concerned with a non-monotonically evolving, non-equilibrium dissipative system, Nicolis and Prigogine, 1977.

In my considered opinion, however, the isotopic analyses are not essential for fairly reliable understanding of circumstances of formation of the Yucca Mountain calcretes, fault infillings, and subsurface veins. Allow me to share with you a statement made by the distinguished British scientist, Professor N. J. Price, who has examined both the field evidence and the pertinent in-situ experimental data. "Moreover, I maintain that to an experienced field geologists, with experience in studying veins, this conclusion (from time to time, the water table reaches the topographic surface), based on a study of the field evidence, is obvious" - a turn of phrase I could not but envy.

ATTACHMENTS

Part A - Isotopic Comparative Analyses

- Figure 1 Carbon-oxygen isotopic fingerprint. The Yucca Mountain surficial calcite-silica deposits.
- Figure 2 Carbon isotopic file - isotopic character of carbon contained in CaCO_3 deposits precipitated in surficial fluids that own their CO_2 content to biogenic activity.
- Figure 3 Carbon isotopic file - isotopic character of carbon contained in travertines, worldwide data.
- Figure 4 Carbon isotopic file - isotopic character of carbon contained in carbonate gangue minerals from a number of hydrothermal ore deposits.
- Figure 5 Carbon isotopic file - isotopic character of carbon contained in the Yucca Mountain subsurface veins.
- Figure 6 Carbon isotopic file - isotopic character of carbon contained in the local geothermal fluids; host rock is tuff "pile".
- Figure 7 Oxygen isotopic file - isotopic character of oxygen contained in travertines, worldwide data.
- Figure 8 Oxygen isotopic file - isotopic character of oxygen contained in the deep-seated spring deposits from the Ash Meadows Basin, Nevada.
- Figure 9 Oxygen isotopic file - isotopic character of oxygen contained in the Yucca Mountain calcite-silica subsurface veins.
- Figure 10 Oxygen isotopic fractionation data from travertine depositing springs of Central Italy and Yellowstone Park.
- Figure 11 Oxygen isotopic file - isotopic character of oxygen contained in the contemporary Yucca Mountain subsurface fluids.
- Figure 12 Oxygen isotopic file - isotopic character of oxygen contained in the contemporary Nevada Test Site geothermal fluids.
- Figure 13 Conclusions resulting from the oxygen -18 and carbon -13 comparative analyses.
- Figure 13a One of the principals of Aristotelian logic states: "if A is not always B then C, although included in B, does not have to be A".
- Figure 13b One of the principals of Aristotelian logic states: "if A is not always B then C, although included in B, does not have to be A".

Part B - Considerations of the Precipitation Temperatures for the Yucca Mountain Calcite-Silica Surficial Deposits.

- Figure 14 Interpretations of the precipitation temperatures, based on the oxygen -18 contents from samples of the Yucca Mountain surficial calcite-silica deposits - general remarks.
- Figure 15 Interpretations of the oxygen -18 content for the parent fluids of the Yucca Mountain surficial deposits.
- Figure 16 Time-series for the oxygen -18 content in fluids emerging from Cane Spring.
- Figure 16a The contemporary Cane Spring deposits - a modern analog for the Yucca Mountain surficial calcite-silica deposits.
- Figure 17 Time-series for the deuterium content in the Nevada Test Site subsurface fluids, based on the results of fluid inclusion studies of calcitic veins from Devil's Hole, Ash Meadows, and Furnace Creek area.
- Figure 18 Time-series for the oxygen -18 content in the parent fluids for the Devil's Hole (DH-2) vein.
- Figure 19 Potential errors that may pertain to estimates of the precipitation temperatures for the Yucca Mountain calcite-silica surficial deposits, based on the oxygen -18 contents from samples of these deposits.
- Figure 20 Illustration of uncertainties associated with interpretations of the precipitation temperature for the calcite-silica deposits, based solely on the oxygen -18 content of these deposits.
- Figure 21 1985 interpretation of the precipitation temperatures for the Yucca Mountain vadose zone veins.
- Figure 22 1990 interpretation of the precipitation temperatures for the Yucca Mountain vadose zone veins.
- Figure 23 Reconstructions of the precipitation temperatures for the Yucca Mountain surficial deposits, based on the oxygen -18 contents from samples of these deposits.
- Figure 24 Reconstruction A - the precipitation temperatures for the Yucca Mountain surficial deposits, based on the oxygen -18 contents from samples of these deposits.
- Figure 25 Reconstruction B - the precipitation temperatures for the Yucca Mountain surficial deposits, based on the oxygen -18 contents from samples of these deposits.
- Figure 26 Reconstruction C - the precipitation temperatures for the Yucca Mountain surficial deposits, based on the oxygen -18 contents from samples of these deposits.
- Figure 27 Reconstruction D - the precipitation temperatures for the Yucca Mountain surficial deposits, based on the oxygen -18 contents from samples of these deposits.
- Figure 28 Reconstruction E - the precipitation temperatures for the Yucca Mountain surficial deposits, based on the oxygen -18 contents from samples of these deposits.
- Figure 29 Summary - the precipitation temperature reconstructions. The Yucca Mountain calcite-silica deposits.

Part C - Considerations of the Yucca Mountain Paleo-Geothermal Gradient.

- Figure 30 Interpretations of the paleo-geothermal gradients, based on the observed $d\delta^{18}O/dz$ gradient - general remarks.
- Figure 30a Interpretations of the paleo-geothermal gradients, based on the observed $d\delta^{18}O/dz$ gradient - general remarks.
- Figure 31 Reliability assessment for the paleo-geothermal gradient reconstructions performed by Szabo and Kyser (1990).
- Figure 32 Analyses of the paleo-geothermal gradient, as reconstructed by Szabo and Kyser (1990).
- Figure 33 Implications resulting from the paleo-geothermal reconstructions made by Szabo and Kyser (1990).
- Figure 34 Comparison between the paleo-geothermal gradient and the contemporary geothermal gradient. Well UE-25a.
- Figure 35 Comparison between the paleo-geothermal gradient and the contemporary geothermal gradient. Well USW G-2.
- Figure 36 Comparison between the paleo-geothermal gradient and the contemporary geothermal gradient. Well USW G-3.
- Figure 37 Comparison between the paleo-geothermal gradient and the contemporary geothermal gradient. Well USW G-4.
- Figure 38 Alternate interpretations of the Yucca Mountain paleo-geothermal gradient - general remarks.
- Figure 39 Reconstruction A - the Yucca Mountain paleo-geothermal gradient, based on the oxygen -18 content of samples of the local calcite-silica deposits.
- Figure 40 Reconstruction B - the Yucca Mountain paleo-geothermal gradient, based on the oxygen -18 content of samples of the local calcite-silica deposits.
- Figure 41 Reconstruction C - the Yucca Mountain paleo-geothermal gradient, based on the oxygen -18 content of samples of the local calcite-silica deposits.
- Figure 42 Summary - the paleo-geothermal gradient reconstructions.
- Figure 43 Comparison between the contemporary geothermal gradient and the paleo-geothermal gradients, as reconstructed based on the $d\delta^{18}O/dz$ gradient from samples of the calcite-silica deposits and using various assumptions.
- Figure 44 Concluding remarks.

Part A - ISOTOPIC COMPARATIVE ANALYSES

Figures 1 through 13b

Note:

- The carbon-oxygen part of the isotopic fingerprint, for the Yucca Mountain surficial calcite-silica deposits, is a range of values for the $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ ratios;
- The observed range for the $\delta^{13}\text{C}$ ratio is from -3.0 to -7.5 per mil *PDB*; and
- The observed range for the $\delta^{18}\text{O}$ ratio is from 19.2 to 22.0 per mil *SMOW*.

Carbon-oxygen isotopic fingerprint. The Yucca Mountain surficial calcite-silica deposits. From Whelan and Stuckless, 1990.

- o For the Yucca Mountain area, a mean value for proportion of plants having the C-3 metabolic pathway is ~ 85 percent, Quade and Cerling, 1989.
- o A mean value for the $\delta^{13}C$ ratio for CO_2 produced by plants having the C-3 metabolic pathway is ~ -27 per mil PDB .
- o A mean value for the $\delta^{13}C$ ratio for CO_2 produced by plants having the C-4 metabolic pathway is ~ -12 per mil PDB .
- o A mean value for the $\delta^{13}C$ ratio for carbon dioxide produced by the local plants is ~ -24 per mil PDB - $(-27 \text{ per mil} \cdot 85 \text{ percent} + -12 \text{ per mil} \cdot 15 \text{ percent})/100$.
- o Dissolution of the locally produced biogenic CO_2 will yield fluids with values of the $\delta^{13}C$ ratio of about -16 per mil PDB ($10^3 \ln \alpha_{HCO_3^- - CO_2} \sim 8$ per mil PDB).
- o At temperatures less than $50^\circ C$, fluids carrying $\delta^{13}C \sim -16$ per mil PDB may yield $CaCO_3$ deposits with values of the $\delta^{13}C$ ratio of about -14 per mil PDB ($10^3 \ln \alpha_{CaCO_3 - HCO_3^-} \sim 2$ per mil PDB).

Note:

- a) Carbon isotopic fractionation data are from Hoefs, 1987; and
- b) Plant CO_2 data are from Deines, 1980.

Results of isotopic comparison: the observed Yucca Mountain range, from -3.0 to -7.5 per mil PDB , is too "heavy" to be the result of the local biogenic activity alone.

Carbon isotopic file - isotopic character of carbon contained in $CaCO_3$ deposits precipitated in surficial fluids that own their CO_2 content to biogenic activity.

Figure 2.

Note:

- a) In some regions, carbon contained in travertine deposits is largely derived from dissolution of marine limestones and, therefore, mean values for the $\delta^{13}C$ ratio tend to be positive, say +3.0 per mil PDB ; and
- b) In other regions, however, carbon contained in travertine deposits is isotopically "lighter" than that derived exclusively from marine limestones. A number of factors may be involved, including: i) participation of biogenically produced CO_2 ($\delta^{13}C$ values ranging from -35 to -10 per mil PDB); and ii) participation of CO_2 from deep, igneous sources ($\delta^{13}C \sim -7$ per mil PDB).

Results of isotopic comparison: the observed Yucca Mountain range, from -3.0 to -7.5 per mil PDB , is "lighter" than that from travertines produced as the result of marine limestone dissolution alone.

Carbon isotopic file - isotopic character of carbon contained in travertines, worldwide data. From Turi, 1986.

Figure 3.

Note:

a) The isotopic character of carbon contained in CO_2 derived from deep-sealed, igneous sources is described by a mean value of the $\delta^{13}C$ ration of about -7 per mil PDB ; and

b) Depending upon reaction temperature ($10^3 \ln \alpha_{HCO_3-CO_{(aq)}} = 9.483 \cdot 10^3 \cdot T^{-1} - 23.89$), dissolution of the igneous CO_2 may yield geothermal fluids with values of the $\delta^{13}C$ ratio ranging from $+1$ (temp. $\sim 20^\circ C$) to -14 per mil PDB (temp $\sim 300^\circ C$).

Results of isotopic comparison: the observed Yucca Mountain range, from -3.0 to -7.5 per mil PDB , is similar to that contained in carbonate gangue minerals from hydrothermal ore deposits.

Carbon isotopic character of carbon contained in carbonate gangue minerals from a number of hydrothermal ore deposits. From Hoefs, 1987.

Figure 4.

Note:

- Values of the CO_2 partial pressure for the Yucca Mountain subsurface fluids, as calculated and reported by Kerrisk (1987), range from $\log P_{\text{CO}_2} = -0.79$ to $\log P_{\text{CO}_2} = -2.86$ atm. The mean local value of $\log P_{\text{CO}_2}$ is well above the CO_2 partial pressure in the atmosphere ($\log P_{\text{CO}_2} \sim -3.5$ atm);
- The results of monitoring of the Yucca Mountain borehole UZ63 revealed that, during a 30-month long monitoring period, this borehole has exhaled ~ 1150 kg of carbon, Thorstenson et al., 1989; and
- Both of the above observations suggest that, at Yucca Mountain, the net CO_2 flux is from hydrosphere $\rightarrow$ vadose zone $\rightarrow$ atmosphere, but not vice versa. A common sole source of carbon, contained in both the surficial deposits and the subsurface veins, may not be the atmosphere - biosphere system.

Results of isotopic comparison: the observed Yucca Mountain range (surficial deposits), from -3.0 to -7.5 per mil PDB , is similar as that from samples of calcite silica veins from both the vadose zone and below the water table.

Carbon isotopic file - isotopic character of carbon contained in the Yucca Mountain subsurface veins. From Szabo and Kyser, 1990.

Figure 5.

Note:

- Phrase hydraulic "source" region is used to denote local areas that; i) exhibit positive values of the $d\phi/dz$ gradient $-\phi$ is hydraulic potential; ii) exhibit high values of the dt/dz gradient $-dt/dz \sim 35-55^\circ\text{Celsius per 1km of depth}$; and iii) contain fluids having relatively high concentrations of $\text{CL}^- + \text{SO}_4^{--}$ anions;
- At precipitation temperatures $\sim 50^\circ\text{Celsius}$, value of the isotopic fractionation factor $10^3 \ln \alpha_{\text{CaCO}_3 - \text{HCO}_3^-}$ is ~ -2 per mil P_{DB} ; and
- The -3.0 to -7.5 per mil P_{DB} range, from samples of the Yucca Mountain surficial deposits, indicates that the parent fluids for these deposits have carried values of the $\delta^{13}C$ ratio ranging from -5.0 to -9.5 per mil P_{DB} .

Results of isotopic comparison: the observed Yucca Mountain range, from -3.0 to -7.5 per mil P_{DB} , is similar to that expected for deposits produced by the local geothermal fluids.

Carbon isotopic file - isotopic character of carbon contained in the local geothermal fluids; host rock is tuff pile. Data from Claassen, 1985; Benson and McKinley, 1985; and White and Chuma, 1987.

Figure 6.

Note:

- A value of the $\delta^{18}O$ ratio, from a sample of a particular travertine deposit, reflects three main factors, namely: i) oxygen - 18 content for the parent fluid; ii) precipitation temperature; and iii) conditions of precipitation and the resulting value of the isotopic fractionation factor; and
- Various combinations of the controlling factors are possible and, consequently, the worldwide travertine deposits exhibit a very wide range of the $\delta^{18}O$ ratio, from +2 to about +32 per mil *SMOW*.

Results of isotopic comparison: the observed Yucca Mountain range, from 19.2 to 22.0 per mil *SMOW*, is within the corresponding range from the worldwide travertine deposits.

Oxygen isotopic file - isotopic character of oxygen contained in travertines, worldwide data. From Turi, 1986.

Figure 7.

Note:

- a) Both the $\delta^{13}\text{C}$ vs. lithology gradient and the $\delta^{18}\text{O}$ vs. lithology gradient may reasonably be attributed to an increasing residence time, at the topographic surface, of the deposits parent fluids;
- b) The increasing residence time for the parent fluid, at the topographic surface, is accompanied by: i) decreasing precipitation temperature; ii) increasing kinetic fractionation effects; and iii) increasing oxygen -18 and carbon -13 evaporative enrichment - each of these factors may account for the observed gradients;
- c) Tpa denotes soft - chalky limestone; Tld denotes dense nodular and fenestral limestone; Tpl denotes carbonate rocks and minerals disseminated in claystones;
- d) At the Ash Meadows topographic surface, the ambient temperatures are likely to be 5 to 10° Celsius higher than the corresponding temperatures at Yucca Mountain - the resulting differences in the equilibrium fractionation factor range from 1 to 2 per mil *SMOW*, the Yucca Mountain values being larger; and
- e) Differences in values of the $\delta^{13}\text{C}$ ratio, between the Ash Meadows deposits and the Yucca Mountain surficial deposits, are directly attributable to the corresponding differences in the parent fluids host rocks.

Results of isotopic comparison: the observed Yucca Mountain range, from 19.2 to 22 per mil *SMOW*, if adjusted for the differing equilibrium fractionation factors is compatible with the oxygen -18 content of the Ash Meadows spring carbonates.

Oxygen isotopic file - isotopic character of oxygen contained in the deep-seated spring deposits from the Ash Meadows Basin, Nevada. From Ilay et al., (1986).

Figure 8.

Note:

- The entire observed Yucca Mountain range of the $\delta^{18}O$ ratios for carbonate deposits, from 15.4 to 22.0 per mil $SMOW$, is very similar to that observed for the Ash Meadows Basin deposits, Figure 8; and
- The above similarities suggest that, the differences in oxygen -18 contents from samples of the respective surficial deposits are caused by the respective differences in either the precipitation temperatures or the isotopic fractionation factors, but not by major differences in the oxygen -18 contents of the respective parent fluids.

Results of isotopic comparison: the observed Yucca Mountain range, from 19.2 to 22 $SMOW$, if adjusted for the differing precipitation temperatures, is compatible with the observed oxygen -18 content of the Yucca Mountain subsurface veins.

Oxygen isotopic file - isotopic character of oxygen contained in the Yucca Mountain calcite-silica subsurface veins. Data from Szabo and Kyser (1990) and Whelan and Stuckless (1990).

Figure 9.

Note:

- During natural depositions of travertines, the conditions of equilibrium isotopic fractionation are seldom attained, mainly as a consequence of the kinetic or non-equilibrium processes;
- Depending upon conditions of precipitation, the combined or actual isotopic fractionation factor ($10^3 \ln \alpha_{\text{CaCO}_3 - \text{H}_2\text{O}}$) may be both larger and smaller than the equilibrium fractionation factor; and
- At a precipitation temperature of $t \sim 20^\circ \text{C}$, the combined isotopic fractionation factor may be assumed to range from 22 to 36 per mil SMOW .

Oxygen isotopic fractionation data from travertine depositing springs of Central Italy and Yellowstone Park. From Turi, 1986.

Figure 10.

Note:

- The observed range of values for the $\delta^{18}O$ ratio, from samples of the contemporary Yucca Mountain subsurface fluids, is from -14.2 to -12.7 per mil $SMOW$; samples are bulk water samples pumped out of large segments of exploratory wells - sampling "smoothing" may be involved, and the observed range may be smaller than the actually present one; and
- If the parent fluids, for the Yucca Mountain surficial deposits, have carried the same oxygen -18 contents then, the combined isotopic fractionation factor was in a range from 31.9 to 35.2 per mil $SMOW$, which is permissible by the empirical fractionation data from Figure 10.

Results of isotopic comparison: the observed Yucca Mountain range, from 19.2 to 22 per mil $SMOW$, is compatible with that expected for deposits produced by the contemporary Yucca Mountain subsurface fluids.

Oxygen isotopic file - isotopic character of oxygen contained in the contemporary Yucca Mountain subsurface fluids. Data from Benson and McKinley, 1985.

Figure 11.

Note:

- The observed range of values for the $\delta^{18}O$ ratio, from samples of the contemporary Nevada Test Site geothermal fluids, is from -14.8 to -12.3 $SMOW$; samples are bulk samples pumped out of large segments of exploratory wells - sampling "smoothing" may be involved, and the observed oxygen isotopic shift may be smaller than the actually present one; and
- If the parent fluids, for the Yucca Mountain surficial deposits, have carried the same oxygen 18 contents then, the combined isotopic fractionation factor was in a range from 31.5 to 36.8 per mil $SMOW$, which again is permissible by the empirical fractionation data from Figure 10.

Results of isotopic comparison: the observed Yucca Mountain range, from 19.2 to 22.0 per mil $SMOW$, is compatible with that expected for deposits produced by the contemporary Nevada Test Site geothermal fluids.

Oxygen isotopic file - isotopic character of oxygen contained in the contemporary Nevada Test Site geothermal fluids. Data from Claassen, 1985; Benson and McKinley, 1985; and White and Chuma, 1987.

Figure 12.

The above performed comparative analyses indicate that the $\delta^{18}O$ vs. $\delta^{13}C$ field, from samples of the Yucca Mountain caliche-silica deposits, is such that it is entirely reasonable to postulate that these deposits may have been formed from upwelling geothermal fluids (so-called *per ascensum* origin). Specifically, the observed field is compatible with both the corresponding field from the unquestionable *per ascensum* deposits and the expected field for deposits produced from the local geothermal fluids.

The noted by Quade and Cerling compatibility of the $\delta^{18}O$ vs. $\delta^{13}C$ fields, between the Yucca Mountain surficial deposits and from the local cobble encrustations, may be taken to have a two-fold meaning. On the one hand, it is conceivable that both the *per ascensum* process and the *per descensum* process [i.e., a) calcium and silica contained in the resulting deposits are provided by wind-blown dust; and b) the parent fluids for the resulting deposits are infiltrating meteoric fluids, directly from atmospheric precipitation] yield deposits that, for one reason or another, carry similar carbon and oxygen isotopic fingerprints. If this is the case indeed then, the $\delta^{18}O$ vs. $\delta^{13}C$ field should not be used as the origin discriminating factor. On the other hand, the Quade and Cerling premise, that the local calcareous cobble encrustations were formed via the *per descensum* process, may very well be wrong. Such encrustations could have been formed through a number of processes, including: a) the topographic surface evaporation of shallow, seasonal bodies of run-off fluids (so-called *sensu lato per descensum* process); b) the topographic surface evaporation of near-by, no longer active, deep-seated spring discharges; and c) some combination of a) and b). If this is the case indeed then, it is entirely improper to use $\delta^{18}O$ vs. $\delta^{13}C$ field, from the cobble encrustations, as a meaningful reference. (see Figures 13a and 13b for evidence that justifies this statement)

Clearly, in order to proceed with further resolutions of the origin dilemma, for the Yucca Mountain caliche-silica deposits, it is necessary to consider some additional factors. It is fortunate indeed that, in the case of Yucca Mountain, the already available oxygen - 18 data base is sufficiently broad to facilitate gaining some insights into geothermal circumstances of formation of the local caliche-silica deposits. It is here where a satisfactory resolution of the dilemma may lie. Let's consider two topics, namely a) the precipitation (crystallization) temperatures for the deposits and b) the depth vs. precipitation temperature gradient (so-called paleo-geothermal gradient).

LAT.	COUNTRY, REGION	RAINFALL SEASON	C _a VEGETATION	$\delta^{13}C$ OF CALCRETE				SOURCE
				-10	-5	0	%	
50°N	NETHERLANDS	a	LOW	●●●●				
44°N	FRANCE, PROVENCE	w	LOW		xx			1
41°N	ITALY, APULIA	w	LOW	xx	xx			2
40°N	USA, NEW JERSEY	a	LOW		+			2
38°N	GREECE	w	LOW		x			5
37°N	SPAIN, SOUTH	w	LOW	●●	●●	●●	●●	18
35°N	CYPRUS	w	LOW	●●	●●	●●	●●	2,3,4
34°N	MOROCCO, FEZ	w	LOW					2
33°N	LIBYA, NORTH	w	LOW		x	xx	xx	2
32°N	ISRAEL, JERUSALEM	w	LOW	▲	▲	▲▲		2
31°N	USA, TEXAS	a	MED.			▲	▲▲	6
28°N	TENERIFE	w	LOW		x	x	▲	7
25°N	INDIA, NW DESERT	a	HIGH		x			4
18°N	SENEGAL, DAKAR	a	HIGH			▲	▲	2
1°N	KENYA	a	HIGH	x	x			8
17°N	NAMIBIA, OMAMBO	a	HIGH			▲	x	2
23°S	NAMIBIA, CENTRAL	a	HIGH			●●●●	WAD + ▲▲▲▲	9
23°S	S. AFRICA, TRANSVAAL	a	HIGH				▲▲▲▲	9,10
26°S	AUSTRALIA, FINKE	a	HIGH			▲▲		10,11
27°S	S. AFRICA, KALAHARI	a	HIGH			●●	x	12
28°S	AUSTRALIA, BRISBANE	a	MED.			●●●●	+	10,11
30°S	S. AFRICA, N. CAPE	a	HIGH			+	▲▲	15
32°S	AUSTRALIA, TORRENS	a	MED.			●	▲	10,11
34°S	S. AFRICA, S.W. CAPE	w	LOW			▲	x	13
36°S	AUSTRALIA, VICTORIA	a	LOW			●	●	10,17
43°S	NEW ZEALAND, OTAGO	a	LOW			▲▲	▲	14
								16

Note:

- The worldwide data for calcretes are from Talma and Netterberg (1983);
- For the Nevada Test Site cobble encrustations, the $\delta^{13}C$ ranges from +4.1 to -9.11 ‰, Quade et al., (1989);
- This range overlaps with: i) the corresponding range from the worldwide "pedogenic" (?) calcretes, which is from +4 to -12 per mil ‰; ii) the corresponding range from the worldwide groundwater calcretes, which is from +1 to -10 per mil ‰; iii) the corresponding range from the worldwide travertines, which is from +12 to -24 per mil ‰, Figure 3; iv) the corresponding range from carbonate gangue from gold-bearing hydrothermal ore deposits, which is from +10 to -10 per mil ‰, Figure 4; v) the corresponding range from the Ash Meadows deep seated spring deposits, which is from +2.5 to -3.0 per mil ‰, Figure 8; and vi) the corresponding range from the Yucca Mountain calcite-silica deposits, which is from -3.0 to -8.5 per mil ‰, Figures 1 and 5; and
- One of the following statements must be true either i) the *per ascensum* process yields carbonate deposits that, in terms of the $\delta^{13}C$ ratios, are similar to those produced via the *per descensum* process; or ii) assumption of the *per descensum* origin for the "pedogenic" calcareous deposits is false.

One of the principals of the Aristotelian logic states: "if A is not always B then C, although included in B, does not have to be A".

LAT.	COUNTRY, REGION	$\delta^{18}O$ OF CALCRETE			SOURCE
		-10	-5	0	%
50° N	NETHERLANDS				1
44° N	FRANCE, PROVENCE		xx		2
41° N	ITALY, APULIA		xxxxxx		2
39° N	GREECE		x		18
37° N	SPAIN, SOUTH		xxxxxx	xxxxxx	2,3
35° N	CYPRUS		xxxxxx	xx	2
34° N	MOROCCO, FEZ				2
33° N	LIBYA, NORTH		xxxxxx	xx	2
28° N	INDIA, NW DESERT		xxxxxx	xxxxxx	2
1° N	KENYA		xx		2
17° S	NAMIBIA, OWAMBO				9
23° S	CENTRAL			xx	9,10
25° S	S. AFRICA, TRANSVAAL			xx	10
27° S	KALAHARI			xx	2,10
30° S	N. CAPE			xx	10
34° S	SW. CAPE			xx	10

Note:

- The worldwide data for calcretes are from Talma and Netterberg (1983);
- For the Nevada Test Site cobble encrustations, the $\delta^{18}O$ ranges from -0.4 to -12.6 ‰ PDB, Quade et al., (1989);
- This range overlaps with: i) the corresponding range from the worldwide "pedogenic" (?) calcretes, which is from +4 to -8.5 ‰ PDB; ii) the corresponding range from the worldwide groundwater calcretes (including "riverwater" calcretes), which is from -0.5 to -5 ‰ PDB; iii) the corresponding range from the worldwide travertines, which is from +1.5 to -28 ‰ PDB, Figure 7; iv) the corresponding range from the Ash Meadows deep seated spring deposits, which is from ~ -4.5 to ~ -15.5 ‰ PDB, Figure 8; and v) the corresponding range from the Yucca Mountain calcite-silica deposits, which is from ~ -8.5 to ~ -16.0 ‰ PDB, Figure 9; and
- One of the following two statements must be true either i) the *per ascensum* process yields carbonate deposits that, in terms of the $\delta^{18}O$ ratios, are similar to those produced via the *per descensum* process; or ii) assumption of the *per descensum* origin for the "pedogenic" calcareous deposits is false.

One of the principals of the Aristotelian logic states: "if A is not always B then C, although included in B, does not have to be A".

**Part B - CONSIDERATIONS OF THE PRECIPITATION TEMPERATURES FOR THE YUCCA MOUNTAIN
CALCITE-SILICA SURFICIAL DEPOSITS.**

Figures 14 through 29

- o A reliable interpretation of the precipitation temperatures, for the Yucca Mountain surficial calcite-silica deposits, constitutes an important aspect of resolving the dilemma that surrounds the origin of these deposits.
- o An unequivocal demonstration that, for the surficial deposits, the precipitation temperatures were ranging from 14 to say 20° Celsius (ambient temperature near the topographic surface) would indicate that both the *per descensum* and the *per ascensum* origins are plausible. An unequivocal demonstration that these temperatures were ranging from 15 to say 30° Celsius, however, would cause a rejection of the *per descensum* origin.
- o For the Yucca Mountain surficial deposits, reliable interpretations of the exact ($\pm 5^\circ$ Celsius) precipitation temperatures can not be made based solely on the oxygen -18 content of these deposits.
- o This is so because: i) a value of the $\delta^{18}O$ ratio, for the deposit's parent fluid, may not be reliably estimated with the required precision of ± 1.0 per mil *SMOW*; ii) a value for the combined isotopic fractionation factor may not be reliably estimated with the required precision of $\sim \pm 1.2$ per mil *SMOW*; and iii) assumption that, during a time span represented by these deposits [which is from $26 \pm 2 \times 10^3$ to more than 4×10^5 years B.P., Szabo et al. (1981) and Szabo and Kyser (1990)], both the isotopic composition of the parent fluids and the combined isotopic fractionation factor remained time-invariant cannot be justified.

Interpretations of the precipitation temperatures, based on the oxygen -18 contents from samples of the Yucca Mountain surficial calcite-silica deposits - general remarks.

Figure 14.

- o Figures 16 through 17 present the actual time-series for the isotopic compositions of the Nevada Test Site subsurface fluids that are, or were, involved in precipitations of CaCO_3 deposits. Both short and long-term spans are covered.
- o Examinations of these figures reveal that, at the Nevada Test Site, the oxygen -18 content of the subsurface fluids may not be assumed as time invariant. Both the monotonic variabilities and the oscillatory variabilities are evident. The oscillatory variabilities involve both the low frequency fluctuations and the high frequency fluctuations.
- o Relative to the contemporary subsurface fluids, the paleo-subsurface fluids could have been isotopically "heavier" by at least $\Delta\delta^{18}\text{O} \sim 5$ per mil *SMOW*.
- o A potential error in estimating the oxygen -18 content for the parent fluid, based on the corresponding content of the contemporary fluids, may easily exceed a value of $\Delta\delta^{18}\text{O} \sim 5$ per mil *SMOW*. The resulting potential error in estimating the specimen precipitation temperature, and using the equilibrium isotopic fractionation curve, is equal to $\Delta t \sim 22^\circ \text{Celsius}$, Figure 10.

Interpretations of the oxygen -18 content for the parent fluids of the Yucca Mountain surficial deposits.

Note:

- The time series indicates that, even during time-spans of few years, the oxygen -18 content in the local subsurface fluids does not remain time invariant;
- The observed monotonic oxygen -18 depletion is $\Delta\delta^{18}\text{O} \sim 1.2$ per mil SMOW in 6 years; and
- The observed $\delta^{18}\text{O}$ oscillatory variability has a maximum amplitude of $\Delta\delta^{18}\text{O} \sim 5$ per mil SMOW .

Time-series for the oxygen -18 content in fluids emerging from Cane Spring. From Lyles et al., 1990.

Figure 16.

- Note:
- Cane Spring is situated some 1400 ft above the so-called regional water table, around the spring orifice there is an ongoing contemporary deposition of CaCO_3 ;
 - Based on a variety of data, Szymanski (1989) has postulated that both the Cane Spring discharge and the underlying perched bodies of groundwater are deep-seated and together express a contemporary hydraulic mound, presence of which is the result of past hydro-tectonic activity;
 - The contemporary Cane Spring CaCO_3 deposits may be regarded as a modern analog for the Yucca Mountain caliche-silica deposits; and
 - The observed "oxygen isotopic shift" fully supports the previous interpretations regarding the hydrologic setting of the Cane Spring area.

The contemporary Cane Spring deposits - a modern analog for the Yucca Mountain surficial caliche-silica deposits.

Figure 16a.

Note:

- a) During the last 2.5×10^6 years, the deuterium content in the parent fluids for the local calcite veins have changed;
- b) The long-term monotonic depletion in deuterium is $\Delta\delta D \sim 40$ per mil $SMOW$ - the corresponding minimum value for the oxygen -18 depletion is $\Delta\delta^{18}O \sim 5$ per mil $SMOW$ in 2.5×10^6 years; and
- c) The short-period oscillatory variability has a maximum amplitude of $\Delta\delta D \sim 32$ per mil $SMOW$ - the corresponding minimum value for the short-period oxygen -18 variability is $\Delta\delta^{18}O \sim 4$ per mil $SMOW$.

Time-series for the deuterium content in the Nevada Test Site subsurface fluids, based on fluid inclusion studies of calcite veins from Devil's Hole, Ash Meadows, and Furnace Creek area. From Winograd et al., 1985.

Figure 17.

Note:

- Each dot represents a $\delta^{18}O$ analyzed sample;
- The average sampling interval is 1.27mm - the corresponding $\delta^{18}O$ ratio represents a mean value for a time span of 2300 ± 100 years; and
- During the time span from 4×10^4 to about 3.2×10^5 years B.P., which is equivalent to the time span represented by the Yucca Mountain subsurface veins, the mean value for the $\delta^{18}O$ ratio fluctuated with a maximum amplitude of about 2.7 per mil, *SMOW*.

Time-series for the oxygen - 18 content in the parent fluids for the Devil's Hole (DH-2) vein. From Winograd et al., (1988).

- o As shown in Figure 10, the empirical isotopic fractionation data indicate that, at the topographic surface, the deposition of CaCO_3 is often associated with a variety of kinetic processes. As a consequence of these processes, the combined, or actual, isotopic fractionation factor is different than the equilibrium isotopic fractionation factor.
- o The kinetic, or non-equilibrium, isotopic fractionation effects may be as large as $\Delta 10^3 \text{‰} \text{CaCO}_3 - \text{H}_2\text{O} \sim 6$ per mil *SMOW*. The corresponding potential error in estimating the precipitation temperature for a specimen that have undergone the non-equilibrium isotopic fractionation, using only the equilibrium fractionation relationship, may be as large as $\Delta t \sim 30^\circ \text{Celsius}$.
- o Combining both of the potential errors, it is prudent to restrict the reliability of the precipitation temperature estimates to a fairly wide range, most certainly wider than the desired range from 15 to 20 ° Celsius.

Potential errors that may pertain to estimates of the precipitation temperatures for the Yucca Mountain calcite-silica surficial deposits, based on the oxygen -18 contents from samples of these deposits.

- o Uncertainties that are associated with interpretations of the precipitation temperatures for the Yucca Mountain deposits, based solely on the oxygen -18 content of these deposits, may be best illustrated by pointing out the results of such interpretations as performed by Szabo and Kyser (1985) and Szabo and Kyser (1990).
- o A comparison of Figures 21 and 22 reveals that, the same investigators using the same oxygen -18 data came to two different conclusions. In accordance with the results of 1985 interpretations, samples yielding values of the $\delta^{18}O$ ratio ~ 20 per mil were precipitated at a temperature of about $\sim 25^\circ$ Celsius, Figure 20. Because some of the Yucca Mountain calcites and surficial fault infillings carry values of the $\delta^{18}O$ ranging from 19.2 to 20.0 per mil; it follows that some of these deposits were formed at temperatures ranging from 25 to 30° Celsius. These temperatures are far in excess of those that may reasonably be attributed to the *per descensum* process.
- o In accordance with the results of 1990 interpretations, however, samples that yield values of the $\delta^{18}O$ ratio ~ 20 per mil were precipitated at a temperature of about $\sim 14^\circ$ Celsius, Figure 21. This precipitation temperature may be regarded as consistent with both the *per descensum* process and the *per ascensum* process and, therefore, may not be used as the origin discriminating factor.

Illustration of uncertainties associated with interpretations of the precipitation temperature for the Yucca Mountain calcite-silica surficial deposits, based solely on the oxygen -18 contents from samples of these deposits.

Figure 20.

Note:

- Both the 1985 interpretation and the 1990 interpretation are based on the same oxygen - 18 data;
- Samples yielding values of the $\delta^{18}O$ ratio ~ 20 per mil *SMOW* were interpreted to have been formed at a temperature of $\sim 14^\circ$ Celsius; and
- The corresponding range of the precipitation temperatures, for the Yucca Mountain surficial deposits, is from 5 to about $\sim 18^\circ$ Celsius - considerations of the *per descensum* and the *per ascensum* mechanisms are permissible.

1990 interpretation of the precipitations temperatures for the Yucca Mountain vadose zone veins. From Szabo and Kyser, 1990.

- o Figures 24 through 28 present the results of five conceivable reconstructions of the precipitation temperatures for the Yucca Mountain surficial deposits. These reconstructions are based on the oxygen - 18 contents from samples of these deposits, as reported by Whelan and Stuckless (1990). As shown in Figure 1, samples of the Yucca Mountain surficial deposits yield values of the $\delta^{18}O$ ranging from 19.2 to 22.0 per mil *SMOW*.
- o Each reconstruction involves two steps. First, value of the combined isotopic fractionation factor ($10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^3 / T^2 - 2.82 + \beta$) was computed by subtracting the assumed value of the $\delta^{18}O$ ratio, for the deposits parent fluids, from the observed value of the $\delta^{18}O$ ratio, from samples of the deposits. Second, the precipitation temperatures were estimated using the empirical isotopic fractionation data from Turi (1986); if appropriate both the equilibrium isotopic fractionation effects and the non-equilibrium isotopic fractionation effects were considered.
- o For the parent fluids, three different isotopic compositions were considered. These are: i) reconstruction A - made assuming that the parent fluids have carried the oxygen -18 contents similar to those of the local contemporary atmosphere precipitation; ii) reconstructions B and C - made assuming that the parent fluids have carried the oxygen -18 contents similar to those of the local contemporary subsurface fluids; and iii) reconstructions D and E - made assuming that the parent fluids have carried the oxygen -18 contents heavier by $\Delta\delta^{18}O \sim 5$ per mil, than those of the local contemporary subsurface fluids.
- o Reconstruction D is considered as the most reliable and conservative interpretation.

Reconstructions of the precipitation temperatures for the Yucca Mountain surficial deposits, based on the oxygen -18 contents from samples of these deposits.

a) isotopic fractionation data, from Turi (1986).

b) weighted average values of the $\delta^{18}O$ ratio - atmospheric precipitation at the Nevada Test Site, from Ingraham et al., (1990).

Note:

- In performing this reconstruction, it has been assumed that: i) the parent fluids, for the Yucca Mountain surficial fluids, were isotopically identical to the contemporary atmospheric precipitation (at altitudes ranging from 1250 to 1400m, $\delta^{18}O$ values range from -11.7 to -11.3 per mil $SMOW$); and ii) the non-equilibrium isotopic fractionation effects were absent, such that the combined isotopic fractionation factor $10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^3 / T^2 - 2.82$;
- The minimum and maximum values for the combined fractionation factor ($10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^3 / T^2 - 2.82$) are 30.5 and 33.7, respectively;
- The corresponding range for the precipitation temperatures is from 0 to $\sim 14^\circ$ Celsius; and
- The measured contemporary *in-situ* temperatures, at and near the topographic surface, range from 15 to 21° Celsius, Sass et al., (1987) - the paleo and contemporary temperatures discrepancy indicates that one or both of the assumptions, employed to determine the paleo temperature, are false.

Reconstruction A - the precipitation temperatures for the Yucca Mountain surficial deposits, based on oxygen -18 contents from samples of these deposits.

Figure 24.

Note:

- a) In performing this reconstruction, it has been assumed that: i) during a time span represented by the surficial deposits, the isotopic compositions of the parent fluids were the same as those observed below the water table at the present time ($\delta^{18}O$ values ranging from -14.2 to -12.7 per mil $SMOW$, Figure 11); and ii) the non-equilibrium isotopic fractionation effects were absent, such that the combined isotopic fractionation factor $10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^6 / T^2 - 2.82$;
- b) The minimum and maximum values for the combined fractionation factor ($10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^6 / T^2 - 2.82$) are 31.9 and 36.2 per mil $SMOW$, respectively; and
- c) The corresponding range for the precipitation temperatures is from 8 to below 0° Celsius - this abnormally low range indicates that one or both of the employed assumptions are false.

Reconstruction B - the precipitation temperatures for the Yucatan Mountain surficial deposits, based on the oxygen-18 contents from samples of these deposits.

Note:

- In performing this reconstruction, it has been assumed that: i) during a time span represented by the surficial deposits, the isotopic compositions of the parent fluids were the same as those observed below the water table at the present time ($\delta^{18}O$ values ranging from -14.2 to -12.7 per mil $SMOW$, Figure 11); ii) the combined isotopic fractionation factor was larger than the equilibrium fractionation factor, such that $10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^6/T^2 - 2.82 + \beta$; and iii) the non-equilibrium isotopic fractionation is caused by a rapid escape of CO_2 from the parent solution; isotopically lighter CO_2 is eliminated preferentially, and $CaCO_3$ acquires its oxygen from both H_2O and CO_2 ;
- The minimum and maximum values for the combined isotopic fractionation factor ($10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^6/T^2 - 2.82 + \beta$) are 31.9 and 36.2, respectively; and
- The corresponding range for the precipitation temperatures is from 18 to 34° Celsius.

Reconstruction C - the precipitation temperatures for the Yucca Mountain surficial deposits, based on the oxygen 18 contents from samples of these deposits.

Figure 26.

Note:

- a) In performing this reconstruction, it has been assumed that: i) the parent fluids, for the Yucca Mountain surficial deposits, were isotopically heavier than the contemporary Yucca Mountain subsurface fluids, $\Delta\delta^{18}O \sim 5$ per mil *SMOW* (-9.2 to -7.7 per mil *SMOW*, Figure 18); and ii) the non-equilibrium isotopic fractionation effects were absent, such that the combined isotopic fractionation factor $10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^6/T^2 - 2.82$;
- b) The minimum and maximum values for the combined fractionation factor ($10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^6/T^2 - 2.82$) are 26.9 and 31.2 per mil, respectively; and
- c) The corresponding range for the precipitation temperatures is from 13 to $\sim 32^\circ$ Celsius.

Reconstruction D - the precipitation temperatures for the Yucca Mountain surficial deposits, based on the oxygen -18 contents from samples of these deposits.

Figure 27.

Note:

- In performing this reconstruction, it has been assumed that: i) the parent fluids, for the Yucca Mountain surficial deposits, were isotopically heavier than the contemporary Yucca Mountain subsurface fluids, $\Delta\delta^{18}O \sim 5$ per mil *SMOW* (-9.2 to -7.7 per mil *SMOW*, Figure 18); the combined isotopic fractionation factor was larger than the equilibrium fractionation factor, such that $10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^3 / T^2 - 2.82 + \beta$; and iii) the non-equilibrium isotopic fractionation is caused by a rapid escape of CO_2 from the parent solution; isotopically lighter CO_2 is eliminated preferentially, and $CaCO_3$ acquires its oxygen from both H_2O and CO_2 ;
- The minimum and maximum values for the combined isotopic fractionation factor ($10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^3 / T^2 - 2.82 + \beta$) are 26.9 and 31.2 per mil, respectively; and
- The corresponding range for the precipitation temperatures is from 38 to 53° Celsius.

Reconstruction E - the precipitation temperatures for the Yucca Mountain surficial deposits, based on the oxygen - 18 contents from samples of these deposits.

Figure 28.

- o Without having reliable information regarding both the isotopic compositions for the parent fluids and the isotopic fractionation conditions, it is not possible to specify a reliable, but at the same time narrow, range for the precipitation temperatures for the Yucca Mountain surficial deposits. Based on information presently at hand all that may be said, with some degree of certainty, is that these temperatures could have been anywhere within a range from 15 to as much as 53° Celsius, consistently with the *per ascensum* origin for the deposits.
- o The currently available data do not allow for ruling out the possibility that the precipitation temperatures, for all of the considered samples, were within the 15 - 20° Celsius ambient temperature range. As a consequence, based on the results of considerations of the precipitation temperature alone, the *per descensum* origin for the Yucca Mountain surficial deposits can not be rejected.
- o A more reliable and definitive picture of the geothermal circumstances of precipitation, for the Yucca Mountain calcite-silica deposits, may be derived from considerations of the paleo-geothermal gradients. This is so because such considerations may be performed on relativistic basis and therefore, do not require knowledge of the absolute isotopic composition for the deposits parent fluids.

Summary - the precipitation temperature reconstructions. The Yucca Mountain calcite-silica deposits.

Part C - CONSIDERATIONS OF THE YUCCA MOUNTAIN PALEO-GEOTHERMAL GRADIENT.

Figures 30 through 44

- o A reliable interpretation of the Yucca Mountain paleo-geothermal gradient, based on the observed $d\delta^{18}O/dz$ gradient from samples of the local calcite-silica deposits, constitutes an important aspect of resolving the dilemma that surrounds the origin of these deposits.
- o A reliable demonstration that, in the Yucca Mountain vadose zone, the geothermal gradients undergo temporal fluctuations, with amplitudes say $\Delta dt/dz \sim 10-15^\circ$ Celsius per 1 km of depth, would constitute a fairly definitive demonstration that: i) episodically, the Yucca Mountain vadose zone was being inundated with warm fluids from below the water table; and ii) all of the Yucca Mountain calcite-silica deposits were formed via the *per ascensum* process.
- o A reliable interpretation of the paleo-geothermal gradient is difficult to perform. This is so because such an interpretation requires two assumptions. These assumptions are: i) the oxygen - 18 contents of the parent fluids, for spatially and temporally different samples, were either the same or the $\delta^{18}O$ variability is both known and fixed in the spatio-temporal sense; and ii) the non-equilibrium isotopic fractionation effects are either absent or, relative to the equilibrium fractionation curve, maintain a known relationship.
- o It is not possible to exactly specify the oxygen - 18 content of the parent fluids for the Yucca Mountain calcite-silica deposits. This is so because: i) the contemporary Yucca Mountain subsurface fluids exhibit the $\delta^{18}O$ spatial variability of ~ 1.5 per mil *SMOW* (Figure 11) - this variability is known based on bulk fluid samples pumped out of large borehole segments, sampling "smoothing" may be involved, and the actual $\delta^{18}O$ spatial variability may be larger, say $\Delta\delta^{18}O \sim 3$ per mil *SMOW*; and ii) for the time-spans represented by the Yucca Mountain calcite-silica deposits, the locally known value for the $\delta^{18}O$ temporal variability ranges from 2.7 to about 4 per mil *SMOW* (Figures 17 and 18).

Interpretations of the paleo-geothermal gradients, based on the observed $d\delta^{18}O/dz$ gradient - general remarks.

- o We do not know much about the non-equilibrium fractionation effects, as such effects pertain to the Yucca Mountain calcite-silica deposits. In view of substantial errors that potentially may be involved, it is prudent to consider three fractionation scenarios.
- o The first scenario is that, the non-equilibrium isotopic fractionation effects are either absent or depth invariant. In this case, the paleo-geothermal gradient estimates are fairly reliable. The observed constant depth $\delta^{18}O$ variability, from samples of the deposits, may reasonably be regarded as reflecting the spatio-temporal $\delta^{18}O$ variability in the parent fluids for these deposits.
- o The second scenario is that, for some or all of the considered samples, the combined isotopic fractionation factor exceeds the equilibrium fractionation factor and the difference is depth variant. In this case, down to some depth, the observed constant depth $\delta^{18}O$ variability, from samples of the deposits, reflects i) the spatio-temporal $\delta^{18}O$ variability in the parent fluids, together with ii) the spatio-temporal variability of the combined isotopic fractionation factor. It is difficult to separate these two variabilities and, consequently, the paleo-geothermal gradient estimates may not be reliable. Because it is likely that the non-equilibrium fractionation effects diminish depthward, the paleo-geothermal gradient estimates, for deeper portions of the vadose zone, may be regarded as more reliable than those for shallower portions.
- o The third scenario is that, for some or all of the considered samples, the combined isotopic fractionation factor is smaller than the equilibrium fractionation factor and the difference is depth variant. Here again, it is difficult to properly interpret the observed constant depth $\delta^{18}O$ variability, from samples of the deposits. As a consequence, the reliability of the paleo-geothermal gradient estimates may be low, particularly for shallow parts of the vadose zone.

Interpretations of the paleo-geothermal gradients, based on the observed $d\delta^{18}O/dz$ gradient - general remarks.

Figure 30a.

- o Reconstructions of the Yucca Mountain paleo-geothermal gradient, performed by Szabo and Kyser (1990) and shown in Figure 22, were made using the oxygen-18 data from samples of the calcite-silica veins collected from cores extracted in boreholes UE-25a, USW G-2, and USW G-3. The data reported by Whelan and Stuckless (1990), and representing the surficial deposits and veins from borehole USW G-4, were not used.
- o The reconstructions were made employing four assumptions. These assumptions are: i) samples carrying values of the $\delta^{18}O$ ratio ~ 20 per mil smow were precipitated at a temperature $\sim 140^\circ C$; ii) spatially different samples were precipitated from parent fluids having the same oxygen-18 content; iii) during the time span represented by the subsurface veins, from $26 \pm 2 \times 10^3$ to more than 4×10^5 years B.P., the oxygen-18 content for the parent fluids remained the same; and iv) precipitation of the veins occurred as an equilibrium fractionation process, such that the actual fractionation factor was a sole and known function of the precipitation temperature.
- o The known oxygen-18 data from the Nevada Test Site (Figures 16 through 18) and the empirical fractionation data (Figure 10) indicate that none of the employed assumptions may be justified. Consequently, the reliability of the paleo-geothermal gradient reconstructions, as performed by Szabo and Kyser (1990), should be judged as low.

Reliability assessment for the paleo-geothermal gradient reconstructions performed by Szabo and Kyser (1990).

- o As shown in Figure 22, the reconstructed Yucca Mountain paleo-geothermal gradient exhibits a very conspicuous curvature. Down to a depth of $\sim 400\text{m}$, the depthward rate of the *in-situ* temperature increase is fairly low; value of the equivalent geothermal gradient is $dt/dz \sim 17^\circ\text{Celsius}$ per 1km of depth. At greater depths, however, the depthward rate of the *in-situ* temperature increase is sharply higher; value of the equivalent geothermal gradient is as large as $dt/dz \sim 50^\circ\text{Celsius}$ per 1km of depth.
- o The results of geothermal studies, performed by Sass et al., (1987), lead to a conclusion that the Yucca Mountain thermal conductivity structure is fairly homogeneous. Laboratory measurements revealed that a mean value for the local thermal conductivity is $\sim 1.7\text{ W m}^{-1}\text{K}^{-1}$, and deviations from this value are fairly small. *In-situ* measurements of downhole temperature, performed in boreholes UE-25a, USW G-2, and USW G-3 (Figures 34 through 36), indicate that the vadose zone geothermal gradients are fairly monotonic - a clear indication that also the *in-situ* thermal conductivity structure is homogeneous. Both of the above lines of evidence indicate that, the paleo-geothermal gradient curvature is abnormal.
- o This curvature indicates that, most likely, the paleo-geothermal gradient reconstruction is in error. To account for the curvature, two possibilities may be put forth. The first possibility is that, the parent fluids, for the spatially and temporally different veins, have carried different values of the $\delta^{18}\text{O}$ ratio. The actual time-series for either the oxygen -18 content of the deuterium content (Figures 17 and 18) indicate that this is a fairly reasonable possibility. The second possibility is that, the paleo-geothermal gradient curvature is telling us that, during formation of the veins, the non-equilibrium, or kinetic, processes were involved. The paleo-geothermal gradient curvature may be explained by assuming that, with a progressively smaller depth, the combined fractionation factor was progressively smaller than the equilibrium fractionation factor, Figure 10. This non-equilibrium fractionation effect is similar to that observed by Friedman (1970) during his studies of fractionation processes associated with the New Highland Terrace Spring discharge in the Mammoth Hot Spring area of Yellowstone Park, Wyoming. In very rapidly moving fluids, the CaCO_3 nucleation occurs prior to the CaCO_3 deposition. In other words, the CaCO_3 nuclei deposit above their nucleation depth where lower temperatures prevail. These nuclei record a lower than depositional value of the combined fractionation factor.

Analyses of the paleo-geothermal gradient reconstructed by Szabo and Kyser, 1990.

- o Putting aside the above reservations, it may be assumed that the paleo-geothermal gradient reconstruction, made by Szabo and Kyser (1990), is correct. What are the implications that result from this reconstruction?
- o Figures 34 through 37 present comparisons between the reconstructed paleo-geothermal gradient and the contemporary geothermal gradients, as recorded in the corresponding boreholes UE-25a, USW G-2, USW G-3, and USW G-4. Examination of these figures reveals that, in the lower parts of the vadose zone (~ 100 - 250 m above the contemporary water table), the reconstructed paleo-geothermal gradient is a factor of two higher than the contemporary geothermal gradient.
- o Radiometric ages of samples, that were used in the paleo-geothermal reconstructions, range from $26 \pm 2 \times 10^3$ to more than 4×10^5 years B.P., Szabo and Kyser (1990). The factor of two change in the geothermal gradient, occurring during such a short time span, demands a rational explanation. Here, however, there is only one possibility, namely: the basal parts of the Yucca Mountain vadose zone were being repeatedly invaded by warmer fluids from below the water table.
- o Because both the $\delta^{13}\text{C}$ ratios and the $\delta^{18}\text{O}$ ratios from samples of the surficial deposits are identical to those from samples of the deeper veins (Figures 5 and 8), there is little merit in insisting that the surficial deposits were formed via the *per descensum* process. Consequently, the Quade and Cerling conclusion "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" does not appear as a particularly sound one.

Implications resulting from the paleo-geothermal reconstructions made by Szabo and Kyser, 1990.

Comparison between the paleo-geothermal gradient and the contemporary geothermal gradient. Well UE-25a.

Comparison between the paleo-geothermal gradient and the contemporary geothermal gradient. Well USW G-2.

Comparison between the paleo-geothermal gradient and the contemporary geothermal gradient. Well USW G-3.

Comparison between the paleo-geothermal gradient and the contemporary geothermal gradient. Well USW G-4.

Figures 39 through 41 present alternate interpretations of the Yucca Mountain paleo-geothermal gradient, based on the observed $\delta^{18}O/dz$ gradient from samples of the local caliche-silica deposits. These interpretations were made using all of the available oxygen-18 data, as reported by Szabo and Kyser (1990) and Stuckless and Whelan (1990).

A common assumption employed in performing all three interpretations is that, for the surficial caliche-silica deposits, the precipitation temperature was $t \sim 20^\circ\text{C}$. The paleo-geothermal gradient interpretations are only marginally sensitive to the precipitation temperature assumption and, should this assumption be wrong, the resulting errors are insignificantly small.

Three conceivable isotopic fractionation scenarios were considered. These scenarios are: i) reconstruction A - made assuming that the combined isotopic fractionation factor is equal to the equilibrium fractionation factor ($\beta_{(u)} = 0$); ii) reconstruction B - made assuming that the combined isotopic fractionation factor is larger than the equilibrium fractionation factor ($\beta_{(u)} > 0$); and iii) reconstruction C - made assuming that the combined isotopic fractionation factor is smaller than the equilibrium fractionation factor ($\beta_{(u)} < 0$).

Reconstruction A is regarded as the most reliable and conservative interpretation.

Alternate interpretations of the Yucca Mountain paleo-geothermal gradient - general remarks.

Note:

- In performing this reconstruction, it has been assumed that: i) at the topographic surface, the precipitation temperature for calcretes and veins was $t \sim 20^\circ$ Celsius; ii) the constant depth variability of the oxygen -18 content, from samples of the calcite-silica deposits, reflects the spatio-temporal variability of the oxygen -18 content in the parent fluids, $\Delta\delta^{18}O \sim 3$ per mil; and iii) the non-equilibrium isotopic fractionation effects are absent and, therefore, $10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^6/T^2 - 2.82$ - where T is precipitation temperature in $^\circ$ Kelvin; and
- Value of the paleo-geothermal gradient is $dt/dz \sim 35^\circ$ Celsius per 1km increase in depth - the corresponding contemporary geothermal gradient ranges from 20 to 24 $^\circ$ Celsius per 1km increase in depth.

Reconstruction A - the Yucca Mountain paleo-geothermal, based on the oxygen -18 content of samples of the local calcite-silica deposits.

Note:

- In performing this reconstruction, it has been assumed that: i) at the topographic surface, the precipitation temperature for calcretes and veins was $t \sim 20^\circ$ Celsius; ii) the oxygen -18 content of the parent fluids, for spatio-temporally different samples, was the same; iii) the constant depth effects, such that combined $10^3 \ln \alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^6/T^2 - 2.82 + \beta(d)$; and iv) the non-equilibrium fractionation is caused by a rapid, and H_2O and CO_2 ; and
- Value of the paleo-geothermal gradient is $dt/dz \sim 33^\circ$ Celsius per 1km increase in depth - the corresponding contemporary geothermal gradient ranges from 20 to 24° Celsius per 1km increase in depth.

Reconstruction B - the Yucca Mountain paleo-geothermal gradient, based on the oxygen -18 content of samples of the local calcite-silica deposits.

Figure 40.

Note:

- a) In performing this reconstruction, it has been assumed that: i) at the topographic surface, the precipitation temperature for calcretes and veins was $t \sim 20^\circ \text{Celsius}$; ii) the oxygen -18 content of the parent fluids, for spatio-temporally different samples, was the same; iii) the constant depth variability of the oxygen -18 content, from samples of the calcite-silica deposits, is attributable to the non-equilibrium isotopic fractionation effects, such that combined $10^3 \ln \alpha_{\text{CaCO}_3-\text{H}_2\text{O}} = 2.78 \cdot 10^3 / T^2 - 2.82 - \beta_d$; and iv) the non-equilibrium fractionation is caused by a rapid upward movement of the parent fluids - the CaCO_3 nucleation occurs prior to its deposition and, consequently, the deposited CaCO_3 records, temperatures that are higher than those prevailing at the actual deposition sites; and
- b) Value of the paleo-geothermal gradient is $dt/dz \sim 58^\circ \text{Celsius per 1km increase in depth}$ - the corresponding contemporary geothermal gradient ranges from 20 to $24^\circ \text{Celsius per 1km increase in depth}$.

Reconstruction C - the Yucca Mountain paleo-geothermal gradient, based on the oxygen -18 content of samples of the local calcite-silica deposits.

- o Figure 43 presents a summary of reconstructions of the Yucca Mountain paleo-geothermal gradient. The contemporary geothermal gradient, as measured in Well USW G-3 by Sass et al., (1987), is used as a reference.
- o In all cases considered, the reconstructed paleo-geothermal gradient is significantly greater than the contemporary geothermal gradient, as measured in the corresponding wells. Values of the contemporary geothermal gradient are: i) Well UF-25a, $dt/dz \sim 22^\circ \text{ Celsius per 1 km of depth}$; ii) Well USW G-2, $dt/dz \sim 24^\circ \text{ Celsius per 1 km of depth}$; iii) Well USW G-3, $dt/dz \sim 22^\circ \text{ Celsius per 1 km of depth}$; and iv) Well USW G-4, $dt/dz \sim 20^\circ \text{ Celsius per 1 km of depth}$.

- o The reconstructed values of the paleo-geothermal gradient are: i) reconstruction by Szabo and Kyser (1990) - dt/dz ranging from 17, near the topographic surface, to $50^\circ \text{ Celsius per 1 km of depth}$, in deeper parts of the vadose zone; ii) reconstruction A - $dt/dz \sim 35^\circ \text{ Celsius per 1 km of depth}$; iii) reconstruction B - $dt/dz \sim 33^\circ \text{ Celsius per 1 km of depth}$; and iv) reconstruction C - $dt/dz \sim 58^\circ \text{ Celsius per 1 km of depth}$.

- o The interpreted temporal fluctuations of values of the geothermal gradient indicate that, the Yucca Mountain vadose zone was being episodically invaded by warm fluids from below the water table. Evidently, the resulting warming up of the vadose zone was accompanied, and recorded, by the episodic precipitation of the calcite-silica veins.

- o As far as the origin of the calcite-silica deposits is concerned, the available oxygen - 18 and carbon - 13 data are convincing and clear. Both the surficial deposits and the subsurface veins were produced via the *per ascensus* process.

Summary - the paleo-geothermal gradient reconstructions.

Figure 43.

- o In conclusion, the results of both the comparative isotopic analyses (Figures 1 through 13) and the paleo-geothermal analyses (Figures 14 through 43) indicate that the *per descensum* interpretations of the origin of the Yucca Mountain surficial deposits, as proposed by Quade and Cerling, lack a proper foundation. As a matter of fact, the very oxygen -18 and carbon -13 data, that were used in developing the *per descensum* interpretations, may be used to successfully discredit these interpretations.
- o An independent, but again quite convincing, demonstration that the Yucca Mountain calcite-silica deposits were formed via the *per ascensum* process may be constructed on the basis of uranium and strontium isotopic data. Considerations of these data, as performed in a soon to be released report by Szymanski, revealed that the isotopic characters of uranium and strontium contained in these deposits are identical to the isotopic characters of uranium and strontium dissolved in the local geothermal fluids.
- o To an experienced field geologist the *per ascensum* origin of the Yucca Mountain calcite-silica deposits, based on common sense considerations of abundant field evidence alone, is obvious. The gainut of isotopic data only confirms the *a priori* known, and reasonably secure, conclusion. Consequently, the *per descensum* interpretations, as proposed by Quade and Cerling, may hardly be regarded as an example of the most insightful and meticulous science. To the contrary, within the context of safety considerations of a high-level nuclear waste repository, these interpretations may rightfully be regarded as a good example of irresponsible science.

Concluding remarks.

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PART 3

Letter to Editor of Science

I-312122

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REC'D IN YMP

6 March, 1991

3/11/91

Christine Gilbert
Letters Editor: Science
1333 H Street
Washington, DC 20005

To the editor:

We have been asked to comment on a rather lengthy manuscript submitted to Science by Mr. J. Szymanski. His review covers a lot of ground and background literature. We decided to respond in some detail because we are concerned that the unwary reader might not carefully examine Szymanski's reasoning or his use of others' data, and be overawed by the many graphs and scientific jargon. We will only address those parts of his review that pertain to our work on the soils and spring deposits in the region.

First, the three and a half pages of text present Mr. Szymanski's opinion to which he is entitled, however expressed. However, it is clearly opinion and therefore need not be addressed. No science is evident. Following these three and a half pages of text are 44 appended figures. We will not address them all: much of it is devoted to a discussion of the Szabo and Kyser (1990) paper or to the work of Sass et al (1987).

Szymanski's review is so flawed that it is hard to know exactly where to begin. Perhaps the best place is with the conclusions: Figure 44.

"In conclusion, the results of both the comparative isotopic analyses (Figure 1 through 13) and the paleo-geothermal analyses (Figure 14 through 43) indicate that the *per descensum* interpretations of the origin of the Yucca Mountain surficial deposits, as proposed by Quade and Cerling, lack a proper foundation. As a matter of fact, the very oxygen-18 and carbon-13 data, that were used in developing the *per descensum* interpretations, may be used to successfully discredit these interpretations.

An independent, but again quite convincing, demonstration that the Yucca Mountain calcite-silica deposits were formed via the *per ascensum* process may be constructed on the basis of uranium and strontium isotopic data. Considerations of these data, as performed in a soon to be released report by Szymanski, revealed that the isotopic characters of uranium and strontium contained in these deposits are identical to the isotopic characteristics of uranium and strontium dissolved in the local geothermal fluids.

To an experienced field geologist the *per ascensum* origin of the Yucca Mountain calcite deposits, based on common sense considerations of abundant field evidence alone is obvious. The gamut of isotopic data only

confirms the *a priori* known, and reasonably secure, conclusion. Consequently, the *per descensum* interpretations, as proposed by Quade and Cerling, may hardly be regarded as an example of the most insightful and meticulous science. To the contrary, within the context of safety considerations of a high-level nuclear waste repository, these interpretations may rightfully be regarded as a good example of irresponsible science."

In the first paragraph, Szymanski states that the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ data from Yucca Mountain can be used to discredit (disprove?) our hypothesis that the calcite in the Trench 14 veins was derived from a "*per descensum* process", which we have called pedogenic processes. He feels that these studies lack a proper foundation. This can be addressed.

In the second paragraph, Szymanski states that uranium and strontium isotopic data confirm a geothermal origin of the Trench 14 deposits. He presents no data. However, the scientists who actually have done the work have come to conclusions very similar to ours for both the uranium isotopes (Muhs, et al., 1990, 1991; Stuckless, 1991) and the strontium isotopes (Marshall et al., 1990, 1991; Stuckless, 1991). Nothing in the second paragraph can be addressed since no argument is advanced.

The third paragraph has several interesting statements. The first is that it is obvious, from field evidence alone, that the deposits are of *per ascensum* (=geothermal?) origin. No field observations are presented, only a "testimonial". What are these "common sense considerations"? We, and others (e.g., Taylor, 1991), have noted the extreme fine-grained nature of the calcite, fine-grained silica, and the presence of pisolites, all of which are often found in soils in the western U.S. (Hay and Wiggins, 1980). Vein calcite associated with geothermal systems have larger grain size and often have abundant fluid inclusions and coarse grained quartz. Even lower temperature vein calcite, such as is found at Devil's Hole, is coarse grained, contains fluid inclusions, and shows banding (Winograd et al., 1988). On the basis of our field observations, we conclude that the carbonates at Trench 14 are more similar to soil carbonates than to the geothermal vein carbonates that we have observed. Other field geologists, including those with experience studying soils and studying geothermal deposits, with whom we have examined the Trench 14 carbonates are of similar opinion to ours (e.g., J. Whitney (USGS, Denver); R.O. Fournier (USGS, Menlo Park); J. Stuckless (USGS, Denver); M. Reheis (USGS, Denver).

The second interesting statement in the third paragraph is that the "gamut of isotopic data only confirms the *a priori* known . . . conclusion." *A priori* implies being known independantly of observation, existing in the mind prior to and independent of experience, or not based on prior study or examination (Random House Dictionary, 1987). We can only ask how Szymanski came to his conclusion without examining any evidence.

While we may disagree with Szymanski about whether or not our science is meticulous or responsible, we would like to point out that we have published our results in a peer-reviewed fashion where it is open to public scrutiny. We do not believe that newspaper and magazine articles or television interviews are the appropriate ways to disseminate scientific information or misinformation.

From the above, it is evident that the main issue to be discussed is whether the Trench 14 carbonates have an isotopic signature similar to soils or not. Trench 14 carbonates have an isotopic signature virtually identical to modern soil carbonates in the region; it is very different from groundwater vein carbonates of the region (Ash Meadows, Devil's Hole).

We begin with figures 1 and 2 which illustrate the danger of applying isotopic arguments without having read or understood the literature.

Figure 1 and 2.

Figure 1 presents the data of Whelan and Stuckless. Their isotope results are rather similar to those obtained by us. Figure 2 makes an interpretation of these results and purports to show that these could not be soil carbonates based on their $\delta^{13}\text{C}$ values. Figure 2, a text-figure with 7 sentences, has 6 major errors, 5 of which result in a cumulative error of more than 10 permil! In this discussion, Szymanski purports to show that the Trench 14

$\delta^{13}\text{C}$ values could not be soil carbonate because they are too positive by -7 to -11‰ (permil) compared to his expected value.

Error #1. Figure 2, sentence 1. The paper referred to is by Quade, Cerling, and Bowman (1989) not Quade and Cerling (1989). While not important to the argument, it illustrates the lack of care taken by Mr. Szymanski in the preparation of his argument.

Error #2. Figure 2, sentence 1. It is interesting that Mr. Szymanski quotes us as saying that the proportion of C_3 plants in the Yucca Mountain area is 85%. We say nothing of the sort. We would have interpreted our data to suggest that it would be most likely in the range of between about 50 and 75%. The midpoint of this value, 62.5%, would result in an error of about 3 permil in his arguments.

$\Sigma \text{ error} = 3\text{‰}$.

Error #3. Figure 2, sentence 2. Although the mean $\delta^{13}\text{C}$ value for modern C_3 plants is -27‰, such plants prior to about 150 years ago were about -26‰ because of the depletion of ^{13}C in the atmosphere due to the burning of coal and hydrocarbons (see Friedli et al., 1985).

$\Sigma \text{ error} = 4\text{‰}$.

Error #4. Figure 2, sentence 2. The $\delta^{13}\text{C}$ value of C_3 plants is enriched in desert ecosystems by 2 to 3‰. We pointed this out in our paper by referring to other published work and by presenting results from C_3 plants in the region. This effect has been repeatedly documented in the last ten years and cannot be ignored.

$\Sigma \text{ error} = 6\text{--}7\text{‰}$.

Error #5. Figure 2, sentence 4. Mr Szymanski has neglected the effect of diffusional enrichment of ^{13}C in soils. This effect is at least 4.4‰, and is more in desert ecosystems. It has been pointed out in many previous papers (Dorr and Münnich, 1980; Cerling 1984; Cerling et al., 1989; Quade et al., 1989; Quade and Cerling, 1990). Using a minimum of 5‰ enrichment this gives a cumulative error of:

$\Sigma \text{ error} = 11\text{--}12\text{‰}$.

Error #6. Figure 2, sentence 5 and 6. This is really two errors, but we will treat it as one. Mr. Szymanski states that the combined isotopic fractionation factor for $\text{CO}_2\text{--HCO}_3^-$ and $\text{HCO}_3^-\text{--CaCO}_3$ amounts to 10‰, assuming that they are constant below 50 °C. Isotopic fractionation factors are not constant with respect to temperature (Freidman and O'Neil, 1977). At 15 °C isotopic fractionation results about 10.75‰ enrichment in calcite relative to CO_2 . At lower temperatures the effect is even greater.

$\Sigma \text{ error} = 12\text{--}13\text{‰}$.

Thus, we find that Mr. Szymanski made a cumulative error on the order of 12 to 13‰. This reflects the fact that he apparently does not understand or has not read about the major developments in the isotope systematics of soils over the last ten years. Moreover, all his errors are in one direction, strongly suggesting that his reasoning is guided not by objectivity but by, as he states, "*a priori*" considerations.

The consequences of a 12 to 13‰ error are large and need little elaboration. In our paper, we noted that the samples from Trench 14 were actually more negative than local modern soil carbonates ("The Trench 14 carbonates have low $\delta^{13}\text{C}$ values compared to those in nearby Holocene age soil carbonate; these low values indicate that the Trench 14 carbonates did not form in equilibrium with the modern vegetation in the area", p. 1550). We are surprised that Mr. Szymanski did not notice the difference in magnitude between his expected results and our measured results for modern soil carbonates in the region.

Global comparisons of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values and the isotopic composition of soil carbonate, travertines, and gangue minerals. (Figures 3, 4, 7, 13a, and 13b)

Mr. Szymanski's rather elementary comparison of the isotopic results from Trench 14 to a global inventory of pedogenic carbonates (Figures 13a and 13b), travertines (Figures 3 and 7) and hydrothermal ore deposits (Figure 4) is irrelevant. This is an attempt to distract the reader from the close relationship of the values from the Trench 14 carbonates to those of soil carbonates in the region. The broad range of values for various carbonates in Figures 3, 4, 7, and 13 reflects the varied climates, latitudes, and temperatures of carbonate formation that is expected globally. We are well aware of this inventory; however, in our study we consciously focused on the range of climate and hydrologic conditions found in the region near Trench 14. And, as we detailed in our paper, we explained how we arrived at the constraints that can be placed on full-glacial climates. In our opinion these are modest and justified simplifications. To put it simply, we used a uniformitarianism approach that draws on the regional soil patterns and styles of spring discharge as its basis for comparison.

To repeat some observations we have already published on pedogenic carbonates in the region that readily distinguish them from carbonates formed from deep-seated spring sources:

1. There is a strong altitudinal gradient for $\delta^{13}\text{C}$ in pedogenic carbonates in southern Nevada (175 meters/[1‰ $\delta^{13}\text{C}$]) resulting from different types of vegetation growing at different altitudes (Quade et al., 1989; Figure 2, Quade and Cerling, 1990). No gradient would be expected for carbonates precipitated from deep seated springs.

2. There is a strong altitudinal gradient for $\delta^{18}\text{O}$ in pedogenic carbonates in southern Nevada (174 meters/[1‰ $\delta^{18}\text{O}$]) resulting higher elevation meteoric waters being depleted in ^{18}O compared to lower elevation waters (Figure 3, Quade and Cerling, 1990). No gradient would be expected for deep seated springs if the carbonates precipitated at the same temperature.

3. There is a strong correlation between $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ for soil carbonates in southern Nevada. All of the values of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ measured on modern soil carbonates in the region in our (Quade et al., 1989; Quade and Cerling, 1990) and others (Amundson et al., 1988, 1989) studies are different from all of the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values for spring deposits (Winograd et al., 1988; and Hay et al., 1986) in the region when plotted on a $\delta^{13}\text{C}$ versus $\delta^{18}\text{O}$ plot (figure 4, Quade and Cerling, 1990).

4. We have previously found that the difference between the $\delta^{13}\text{C}$ of soil organic matter and soil carbonate is expected to be between 14 and 17‰ based on theoretical grounds (Cerling 1984; Quade et al., 1989); we have observed this difference in modern soils and paleosols (Cerling et al., 1989).

The Trench 14 carbonates are similar to soil carbonates in each of above ways.

Figure 5.

Szymanski states that the $\text{P}(\text{CO}_2)$ for Yucca Mountain subsurface fluids are significantly higher than the atmosphere. This is not surprising: the $\text{P}(\text{CO}_2)$ for all soils is higher than the atmosphere and often reaches values as high as 10^{-2} (Holland, 1978; Richardson and McSween, 1989) The ultimate source of CO_2 in most groundwater systems is probably biogenic.

We cannot figure out what Figure 5 has to do with the caption to Figure 5.

Figure 6.

This is a diagram concerning "source" and "sink" regions. Szymanski has not shown how these are derived and therefore they cannot be addressed. We are quite puzzled as to what, if anything, they mean.

Figure 7.

See comment above on global comparisons

Figure 8 and 9. This figure and Mr. Szymanski's explanation is very intriguing and, we believe, is typical of his approach to science. Mr. Szymanski implies that the carbonates labeled Tpa, Tld, and Tpl in his Figure 8 represent "deep-seated spring deposits from the Ash Meadows Basin, Nevada." It is important to refer to the original paper. Hay et al., (1986) states that "Tpa consists chiefly of limestone and was deposited in spring-fed marshland and ponds. Tpl is chiefly Mg clays with considerable limestone and was deposited in playas and associated marshland" (p.1502). Szymanski has badly misquoted Hay et al. (1986) to advance his hypothesis. Note that he misquoted our work above (see discussion of Figure 2, Error #2). We find the frequent mis-quoting of others' work to be particularly annoying because it means that the reader must be aware that any statement Mr. Szymanski makes must be treated in the same way. We have no confidence in any statement made by Mr. Szymanski unless personally corroborated.

What are the consequences of this misrepresentation? It turns out that Szymanski's argument leans heavily on these results. In Figure 8 Szymanski states: "the observed Yucca Mountain range, from 19.2 to 22‰ SMOW, if adjusted for the differing equilibrium fractionation factors is compatible with the oxygen-18 content of the Ash Meadows spring carbonates". However, if you use the range of values in the original paper, then the range for spring carbonates in the Amargosa region (those labeled Tpa by Hay, et al., 1986, with a range of 15 to 18‰ SMOW) does not overlap that observed for any soil carbonates from southern Nevada that have been analyzed to date (Quade et al., 1989 Quade and Cerling, 1990; Amundson, et al., 1988, 1989). The overlap occurs only when Szymanski redefines the playa carbonates of Hay as "deep-seated spring carbonates".

Once again, Mr. Szymanski has badly misrepresented the evidence in order to further his own opinion. We suspect that, as Szymanski put it, "*a priori*" considerations are again at work. We are concerned that scientists who do not review Hay et al.'s (1986) work themselves may uncritically accept Szymanski's interpretations of Figures 8 and 9.

Figures 10, 11, and 12.

In these figures, Szymanski suggests that under certain conditions deep fluids beneath Yucca Mountain could precipitate carbonates in the observed range. We would agree, it is possible, but does not prove that it happened. Likewise, they could have formed from local precipitation during glacial periods. We are still unclear as to what Szymanski's "source" and "sink" terms are or what they mean.

Figure 13.

See comment above on global comparisons

The first 13 Figures were the ones most closely related to our work. In the next set of figures (14 to 29) Szymanski concludes that it is not possible to use estimated temperature of formation to constrain the origin of the Yucca Mountain deposits. We certainly agree, since it is based on a lot of assumptions. However, we do note that modern soil carbonates do form in the region and have $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values similar to, but slightly different than, the Trench 14 carbonates. We would have been interested if he had concluded that they couldn't exist! However, we should point out that Szymanski has assumed that the isotopic composition of average infiltrating water is the same as the isotopic composition of average meteoric water. This is certainly not true.

Figures 30 to 43 concern the work of Szabo and Kyser (1990) and Sass et al., (1987). We do not see what bearing they have on our paper.

In summary, we have examined Mr. Szymanski's document. We feel that he has made a number of major mistakes in some of his arguments and that he has misquoted

others' work to advance his hypothesis. We are not surprised that he arrived at his conclusions "*a priori*".

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Sincerely yours,

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PART 4

Letter to Cerling

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RESPONSE TO THE MARCH 6, 1991, LETTER BY CERLING AND QUADE

Thank you for your continued interest in the issue of the origin of the Yucca Mountain calcretes and bedrock calcite-opaline silica veins. As you are well aware, this thorny issue is of considerable importance in evaluations of suitability of the Yucca Mountain site to accommodate a high-level nuclear waste repository. The very substantial financial investment by the nuclear industry, the credibility of the U.S. Department of Energy (DOE), the continued commitment of the U.S. Government to the nuclear power option, and, of course, the safety of future generations are all at stake. We at DOE may satisfactorily discharge our responsibilities, but only, by employing objective and sound science and by insisting on cool and informed judgments. This of course, requires a debate, and if need be, the adversarial assessment of the critical and safety-related issues and concerns. In this context, I do sincerely welcome your viewpoints, as expressed in the March 6 letter. I regard this letter as a further encouragement for debate, and as an additional opportunity for critical assessments of the origin of the Yucca Mountain calcite-opaline silica deposits.

I have read the March 6 letter with a considerable interest; quite frankly, I found it interesting and quite revealing with respect to a number of points. It seems to me that, with reference to the discussions presented in the January 18, 1991, letter, you are raising three main objections. These objections pertain to: (1) estimate of the carbon isotopic composition for the soil CO₂ that may be expected to be derived, at Yucca Mountain, from the biogenic activity alone; (2) usage of the oxygen-18 data from Hay et al. (1986); and (3) my alleged a priori approach to resolving the origin issue for the Yucca Mountain calcretes and veins. In the first part of this letter, I will attempt to address all of these objections. The second part of this letter is concerned with four additional topics, which I regard as having a direct bearing on assessments of the scientific validity of the main conclusions proposed by Quade and Cerling. My specific reason for including the additional topics is two-fold. First, in addition to expressing the three main objections, the March 6 letter seems to be demanding further explanations; of course, I feel obligated to provide these explanations. Second, the reliance on both the isotopic composition vs. altitude gradient, from the local cobble encrustations, and the assumption of solely atmospheric-biospheric origin of the locally circulating carbon, as persistently advocated by you, need to be critically considered.

With the above introduction, let's get into the first part of this letter. As far as the first objection is concerned, I would like to raise five points. First, thank you for pointing out that: (1) "because of the depletion of ^{13}C in the atmosphere due to the burning of coal and hydrocarbons," the appropriate mean value of the $\delta^{13}\text{C}$ ratio, for carbon dioxide derived from biomass with the C-3 metabolic pathway, is -26 per mil PDB (instead of -27 per mil PDB , as used in the January 18 letter); and (2) the appropriate value for the $\text{CaCO}_3\text{-CO}_2$ fractionation factor is 10.75 per mil PDB (instead of ~10 per mil PDB , as used in the January 18 letter). I do accept both aspects of this objection and will correct the results of my analyses accordingly. Second, the ~85 percent estimate for proportion of plants with the C-3 metabolic pathway is based on the results of actual plant survey as reported by Quade et al. (1989). Figure 1 shows how I have arrived at this estimate. Perhaps, instead of using the phrase "Quade and Cerling (1989)," it would be more appropriate to say "based on the results of plant survey, as reported by Quade et al. (1989)." My misrepresentation, and I do apologize. I do strongly feel, however, that estimating the C-3 proportion based on the "soil CO_2 data," as advocated by you, is inappropriate and, potentially, introduces an element of circular logic. Third, the 5 per mil PDB correction for the diffusional carbon-13 enrichment in soil CO_2 , as suggested by you, may or may not be appropriate. In any event, this correction may be considered, but only for estimating the isotopic character or carbon contained in the soil CO_2 . Please make a note that, while constructing Figure 2 from the January 18 letter, my interest was narrowly restricted to the "biogenically derived CO_2 alone." Within this context, the suggested carbon-13 diffusional enrichment correction is clearly inappropriate. Fourth, for the Nevada Test Site region, if one is interested in estimating the isotopic character of carbon, that is contained in the CO_2 gas respired from the vadose zone, there is no compelling reason for relying on dubious assumptions and various carbon-13 "desert ecosystem" and "soil diffusional" enrichment corrections. The desired estimate may be derived, I should think fairly reliably, from the results of direct field observations and measurements. Please make a note, from the attached Figure 16, that the mean values for the $\delta^{13}\text{C}$ ratio, from samples of the shallow vadose zone CO_2 gas, are: (1) for the Amargosa Narrows area, $\delta^{13}\text{C}$ ~-20 per mil PDB ; (2) for the Yucca Mountain area, $\delta^{13}\text{C}$ ~-18.36 per mil PDB ; and (3) for the Nevada Test Site area as a whole, $\delta^{13}\text{C}$ ~-20.5 per mil PDB . Potentially, the respired CO_2 is a mixture of three end-members, namely: (a) CO_2 from the biosphere; (b) CO_2 from the atmosphere; and (c) CO_2 introduced as the result of degassing of the hydrosphere. In the second part of this letter, additional analyses are provided to make the point that, for the Nevada Test Site region, the latter source of carbon is of particular importance for adequate understanding of the isotopic character of the locally circulating carbon. You may agree that, the so-called subterranean source of carbon, is not considered in your work. In my opinion, this important omission introduces an element of circular logic into your entire work and, consequently, casts doubt on the proposed by you: (1) estimates of proportion of biomass with the C-3 metabolic pathway, based on "the soil CO_2 data"; (2) the "desert ecosystems" carbon-13 enrichment correction; and (3) the soil diffusional carbon-13 enrichment correction. Fifth, the appropriateness of your assertion that, while estimating the carbon isotopic

composition of the shallow per descensum fluids and assuming that such fluids acquire their carbon content solely from the biogenic source, I have made "a cumulative error on the order of 12 to 13 per mil" PDB may be best evaluated with reference to the direct field and laboratory measurements. Please make a note, from the attached Figure 17, that a reasonable value for the $\text{del } ^{13}\text{C}$ ratio, which may be associated with the Nevada Test Site shallow per descensum fluids, ranges from -9.59 to -16.40 per mil PDB ; a mean value is $\text{del } ^{13}\text{C} \sim -12$ per mil PDB . Correcting the -14 per mil PDB value, from Figure 2 of the January 18 letter, would yield values of the $\text{del } ^{13}\text{C}$ ratio ranging from -2 to -3 per mil PDB . This range is so far removed from the directly-observed values, during both the in-situ surveys and the laboratory soil-leaching experiments, that I see little choice but to reject the asserted error, as totally without merit. I do hope that, the attached Figure 18 provides adequate justification for this rejection.

With reference to the second objection, I must quite frankly admit to being completely ignorant of a substance that may underlie and justify it. Nevertheless, within the context of this objection, I would like to raise five points. First, the Amargosa Basin carbonate deposits that were investigated by Hay et al. (1986) [including: (1) Tpa calcite; (2) Tld calcite; (3) Tpl calcite; (4) Tpl dolomite, and regardless of whether distal (playas, marshland, ponds, etc.) or proximal] are deposits whose parent fluids were the deep-seated fluids emerging from nearby springs. I should think that, the direct field observations create a situation whereby no reasonable geologist may dispute this association--am I missing something? Perhaps, rather than using the phrase "deep-seated spring deposits," in Figure 8 from the January 18 letter, it would be more appropriate to use the phrase "deposits related to deep-seated springs" instead. My misrepresentation, and again I shall apologize for it. Second, as far as I can tell, in no place does the disputed Figure 8 contain either erroneous or misleading quotations from Hay et al. (1986). As a matter of fact, this figure is completely devoid of any quotations. The phrase "From Hay et al., (1986)" identifies source of the considered isotopic and lithologic data, and the note faithfully explains the lithologic notations, as employed by Hay et al., (1986). The attached Figure 2 demonstrates the latter point quite clearly. Third, you are mistaken and misleading the reader by declaring that: "Szymanski's argument leans heavily on these results," which are the isotopic data from Hay et al. (1986). As far as this point is concerned, the January 18 letter contains unmistakably clear language. The per ascensum interpretations, of the origin of the Yucca Mountain calcretes and veins, do "lean heavily," but: (1) on the common sense considerations of abundant and elementary field evidence; and (2) on the comparisons of the observed values of the contemporary geothermal gradients with the corresponding paleo-geothermal values, as reasonably interpreted based on the oxygen-18 contents of samples of the Yucca Mountain subsurface veins. The isotopic comparative analyses, of which the disputed Figure 8 is a part, were taken thus far as to conclude that: "the $\text{del } ^{18}\text{O}$ vs. $\text{del } ^{13}\text{C}$ field, from samples of the Yucca Mountain calcite-silica deposits, is such that it is entirely reasonable to postulate that these deposits may have been formed from upwelling geothermal fluids," Figure 13 from the January 18 letter. Fourth, one could "heavily lean" on the Hay et al. (1986) isotopic

data, however, to demonstrate that: (1) the per descensum interpretations of the origin of the Yucca Mountain calcretes and veins, as proposed by Quade and Cerling (1990), are of doubtful scientific validity; and (2) the parent fluids for the Yucca Mountain calcite-silica deposits [including: (a) the "mosaic" breccia cement; (b) the fault infillings; and (c) the calcretes] exhibit strong oxygen-18 affinity with the parent fluids for the Amargosa Basin per ascensum deposits. The attached Figures 3 through 8 illustrate how one could proceed while making both of such demonstrations. Figures 4 and 5 demonstrate that either: (1) the per ascensum process yields carbonates that, in terms of both the oxygen-18 content and the carbon-13 content, are similar to those produced via the per descensum process; or (2) the assumption of the descensum origin, for the referenced-by-you cobble encrustations, is FALSE. Please make a note, from Figures 3 and 6, that presence of the isotopic compositions vs. lithofacie gradients, for the Amargosa Basin deposits, and of the isotopic composition vs. morphological form gradients, for the Yucca Mountain deposits, strongly suggests (or indicates) the per ascensum origin for both of those deposits. Finally, after examining the attached Figure 8, you may agree that indeed, the parent fluids for the Yucca Mountain calcite-silica deposits exhibit the strong oxygen-18 affinity with the parent fluids for the distal and proximal Amargosa Basin deposits. Fifth, the carbon-oxygen isotopic comparative analyses, ones that you so emphatically reject as "irrelevant," may be of great value, in particular, for a satisfactory performance of the highly speculative isotopic inquiries into origins of various terrestrial gases, liquids, and solids. I do strongly advise you to employ such analyses in your future isotopic work. With regard to the carbon-oxygen isotopic analyses, as undertaken by Quade et al. (1988 and 1989) and Quade and Cerling (1990), the comparative considerations provide a badly needed tie to the reality, and one may derive a number of benefits from their utilization. Allow me to illustrate just two of such benefits. On the one hand, by comparing the attached Figures 2 and 9, you may agree that, the $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ field, from samples of the Amargosa Basin deposits, bears a striking resemblance to the corresponding field from samples of the carbonate gangue veins, associated with the Carlin and Cortez disseminated gold deposits of Nevada. For both of the considered data sets, the host rocks consist of marine limestones and, consequently, the oxygen-carbon comparative analyses may be regarded as particularly meaningful. Please, imagine that the parent fluids for the Carlin and Cortez carbonate veins, instead of leaving their sole mark in the host rock, would have discharged at the topographic surface and would be expressed in the form of cobble encrustations. As a matter of indisputable fact, we do know that, in the Great Basin, this process occurs; often it may be directly observed. What would be the $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ field for these hypothetical cobble encrustations? Well, allowing for some of the carbon-13 diffusional enrichment and for some drop in the precipitation temperature, one may reasonably conclude that such field would be somewhat analogous to that observed by you, around the Nevada Test Site, for the non-carbonate bedrock sites and for the low altitude carbonate sites. I do hope that, the attached Figures 10 and 11 illustrate this point with sufficient clarity. Is it not reasonable to suspect that some of the referenced-by-you cobble encrustations could have been formed from deep-seated fluids, while such fluids were residing at the topographic surface? Is it not

possible that your assumptions, regarding: (1) the solely climatic and atmospheric-biospheric control of the isotopic compositions of "pedogenic" carbonates; and (2) the per descensum origin of such carbonates, are erroneous? On the other hand, by comparing the attached Figures 6 and 9, you may also agree that the oxygen-18 contents, from samples of the Yucca Mountain calcite-silica deposits, are similar to those from the isotopically "heavier" carbonate gangue veins, associated with the Carlin and Cortez disseminated gold deposits. What sets both of the considered deposits apart, however, is the respective carbon-13 content. Considering that, for the Carlin and Cortez veins the host rock consists of marine limestones and, for the Yucca Mountain deposits, of the non-carbonate ignimbrites; the carbon isotopic differences are not surprising and may readily be justified. Using both the carbon-13 content of fluid inclusions entrapped in the carbonate gangue minerals associated with hydrothermal ore deposits (Figure 69) and the carbon-13 content of the isotopically "lightest" Yucca Mountain deposits (Figure 6), it may reasonably be expected that, if the Carlin and Cortez equivalent gangue veins would have precipitated in the Yucca Mountain ignimbrites, the carbon-13 content of these veins would be smaller by about 5 per mil PDB . Discharge the parent fluids for these hypothetical veins at the topographic surface, and allow for some of the carbon-13 diffusional enrichment and for some drop in the mineral precipitation temperature, and you will get the observed Yucca Mountain $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ field. Again, I do hope that, the attached Figure 11 makes this point with adequate clarity. Does it not appear to you that, as far as the Yucca Mountain calcite and opaline silica deposits are concerned, the per ascensum origin is a legitimate interpretation option? Does it not strike you that, the dubious per descensum concept of formation of "pedogenic" carbonates is only a possibility but not much else? Personally, I even doubt that. To summarize my response to your second objection, I again see little choice but, with the noted exception, to reject it as totally without substance and merit.

With reference to the third main objection, I would like to make two statements. On the one hand, it seems to me that, by suggesting "he arrived at his conclusion a priori," you are unfair and inadvertently, I am sure, misleading the reader. As far as the a priori conclusions are concerned, the January 18 letter contains unmistakably clear language. Figure 44 from this letter expresses my utmostly sincere viewpoint that: "To an experienced field geologist the per ascensum origin of the Yucca Mountain calcite-silica deposits, based on common sense considerations of abundant field evidence alone, is obvious. The gamut of isotopic data only confirms the a priori known, and reasonably secure, conclusion." I am certain you are aware that, both the carbon-13 and the oxygen-18 considerations, whether pertaining to a parent fluid or a therefrom precipitated mineral compound, fall into a category of "plausibility arguments." Such considerations seldom, if ever, yield uniquely constrained interpretations. For example, by intermixing a little of the biogenically derived CO_2 [for a start, produced by an adjustable mixture of plants with both the C-3 metabolic pathways and the C-4 (CAM) metabolic pathways] with a little of the atmospheric CO_2 and employing various "desert ecosystem" and "soil diffusional" carbon-13 enrichment corrections, one may duplicate the carbon isotopic compositions of most of the CO_2 genetic

species, that are known to exist in the terrestrial environment. Similarly, it is possible to explain, or duplicate, the oxygen-18 content of parent fluids, as interpreted for a particular accumulation of calcium carbonate, in a number of widely differing ways. This is so because an interpreter is commonly free to employ various combinations of assumptions that pertain to: (1) the oxygen-18 content for the pristine parent fluids; (2) the conditions of subsequent isotopic alterations of these fluids; and (3) the conditions of precipitated compound-parent fluid isotopic fractionations. Considering the substantial uncertainties that may be associated with each of these assumptions, and their combinations, it is neither surprising nor inappropriate for experienced field geologists, while attempting to resolve the origin dilemma for the Yucca Mountain opals and calcites, to rely primarily on the direct and quite informative field observations. This prudent preference, however, does not entitle you to declare that Szymanski has "arrived at this conclusion a priori." Please be advised that, favoring common sense considerations and well established principals, Szymanski has concluded a priori, but only to reject a priori the isotopic illusions and obscurations. On the other hand, however, two statements contained in the March 6 letter [specifically: (1) "no field observations are presented, only a testimonial"; and (2) "what are these common sense considerations?"] seems to be encouraging me, to take a sufficient time, and to share with you some of the direct field observations that justify my viewpoints regarding the obviously per ascensum origin of the Yucca Mountain calcite-silica deposits. Perhaps, to get familiar with the direct and abundant field evidence, it would be most appropriate for you to visit the Yucca Mountain area and examine it first hand. Whenever your schedule permits, I stand ready to escort you and further discuss this important topic. In the meantime, however, let me share with you seven fairly simple, yet reliable, lines of the direct field evidence. First, around Yucca Mountain, the observable rates of the sensu stricto per descensum accumulations, for both calcium carbonate and opaline silica, are vanishingly small, or nearly so. Such a state of affairs is indicated by the directly observable absence of: (1) a discernible calcite-opaline silica accumulations at the surface of the Lathrop Wells cinder cone; and (2) a discernible calcite-opaline silica cement bonding grains that comprise thin stringers of basaltic ash occurring, for example, in the interior of the Trench 14 vein. The most conservative age estimate, for both the cinder cone and the basaltic ash stringers, is $\sim 10^4$ years B.P. The vanishingly small rates for the hypothetical per descensum accumulations, as deduced here for the Yucca Mountain area, are in accord with viewpoints expressed by a number of the per descensum advocates. Machette (1985), for example, has stated that: "Carbonate accumulations rates in the Roswell-Carlsbad area of southeastern New Mexico, over the past 500,000 years, exceed $0.5\text{g/cm}^2/1000$ years and are the highest in the Southwest. At the other extreme, calcic soils in the Beaver, Utah, area have accumulated carbonate at rates of about $0.14\text{g/cm}^2/1000$ years; this rate is 25 to 75 percent of equivalent-age soils in New Mexico." Second, the Yucca Mountain fault infillings, up to a few inches wide, typically exhibit M-textures, in a sense that they consist of pure mineral phases, conspicuously lacking internal impurities such as detrital clays, silts, sands, pebbles, and small boulders. One is hard pressed to imagine either open or progressively opening bedrock

fissure which, while being filled-up with the vanishingly small rates by the per descensum deposits, remains isolated from the overlying soil cover or the detritus that is being transported, at the topographic surface, by wind action and run-off, including occasional flash floods. To account for the observed M-textures, it is necessary to assume the presence of a non-delinquent "cleaning agent" that removes the ever supplied wind- and water-born detritus. In sharp contrast to the descending fluids, the upwardly flowing per ascensum fluids would satisfactorily fulfill this necessary role. Third, the Yucca Mountain calcite deposits occur in the form of vertical veins emplaced through bodies of fairly loose eolian sand. Such veins may be observed in the interior of sand ramps developed on both the west and east sides of Busted Butte. You may agree that, even a stout, but reasonably inquisitive, proponent of the per descensum origin for these veins is immediately stuck on two unanswerable questions, namely: (1) how to form a vertical fissure, a few inches wide, in a fairly loose sand; and (2) how to keep such a fissure open, during a time span that is required for the slowly accumulating per descensum infiling. When dealing with both of these questions, however, the per ascensum advocate is blessed with the decisive advantage. In the context of his preferences, the questions become pale in comparison. He will point out that, a mineralized fluid contained in the ejected slug, from the underlying bedrock fault, seeps through walls of the propagating vertical fissure. The associated drop in the CO₂ partial pressure creates an opportunity for the dissolved calcium carbonate to come out of the ejected solution. Walls of the propagating fissure become stronger and, under the supporting influence of the fluid pressure, may remain open and accommodate the per ascensum calcium carbonate infiling. Fourth, the Yucca Mountain calcretes occur in the form of four textural varieties. These are: (1) the GS-textured calcretes [the allogenic clasts are in direct contact and the authigenic cement occurs in undilated pore space]; (2) the F-textured calcretes [the allogenic clasts "float" in the authigenic cement]; (3) the M-textured calcretes [the authigenic cement constitutes more than 90 percent of a specimen total volume]; and (4) the laminated M- and GS-textured calcretes [laminae of fine cemented sand, but grain supported, separate individual laminae of the detritus-free authigenic cement]. To account for the observed textural diversity, the per descensum advocate must rely on a dubious concept, that the enlargement of pore space may be caused by the authigenic cement crystallization pressures. If such advocate is reasonably inquisitive, however, soon he will become confronted with two unanswerable questions, namely: (1) why the calcretes with clearly comparable ages exhibit the observed textural diversity [presumably, the authigenic cement crystallization pressures exert their dilatory influence indiscriminately]; and (2) why the M-textured calcretes are laminated with the GS-calcretes [presumably the authigenic cement crystallization pressures act in all directions, but not selectively normal to the topographic surface]. When dealing with both of these questions, the per ascensum advocate is again blessed with the decisive advantage. Such advocate will immediately point out that: (1) the authigenic cement formation rates, for a number of reasons, vary in a spatio-temporal sense; (2) the authigenic cement precipitates both directly at the topographic surface and below the topographic surface [the ejected per ascensum fluids may flow either on the topographic surface or through pre-existing alluvial, colluvial, or eolian deposits]; and (3) the authigenic cement formation occurs

in association with ubiquitous introduction of the allogenic clasts, be it by wind action, run-off, or flash floods. Clearly, the spatio-temporally varying rates for both the authigenic cement precipitation and the allogenic clasts introduction are recorded through the textural diversity, as observed for the Yucca Mountain calcretes. A fast, and on the topographic surface direct, precipitation yields both the M-textured calcretes and the laminated M- and GS-textured calcretes. A slower, but still the topographic surface direct, precipitation leads to formation of the F-textured calcretes. Here, the authigenic cement precipitation rates are comparable to the allogenic clast introduction rates. Finally, the GS-textured calcretes are produced when the ejected per ascensum fluids flow through the pre-existing accumulations of various detritus. Fifth, typically the Yucca Mountain surficial infillings of the fault related fissures exhibit nearly vertically banded textures. As it may be observed in Trenches 8 and 14, for example, these infillings consist of veins which, in turn, are composed of bands of calcium carbonate alternating with bands of opal-CT. You may agree that, the observed mineral phase alternations may be taken as indicating that, either the parent fluids chemistry or the mineral phases precipitation conditions were alternating between those favoring precipitation of opal-CT and those favoring precipitation of micritic calcites. Alternatively, it may also be assumed that the opal-CT bands represent the epigenetic replacement of the earlier micritic calcites. In any event, one may reasonably conclude that either the parent fluids for the Yucca Mountain fault infillings or the conditions of precipitation of these infillings were fluctuating, either episodically or continuously. Whether episodic or continuous, the fluctuations involving both of the above factors are not the expected characteristics of the per descensum pedogenic processes. Here again, therefore, the per descensum proponents are confronted with the unanswerable question. For the per ascensum advocate, however, finding an answer to this question presents little challenge. Immediately he will point out that both the fluctuating chemical compositions of the parents fluids and the fluctuating conditions of the mineral phases precipitation are typical features of most geothermal systems. During certain periods of activity of such systems, for example, the parent fluids carry high values of the CO_2 partial pressure. The calcite precipitation is favored because of the rapid CO_2 escape from the parent solution. During some other periods of activity, however, the parent fluids may carry low values of the CO_2 partial pressure and, in this case, progressive cooling of the parent fluids dominates the mineral phase formation process. The opal precipitation may be favored, because of the SiO_2 prograde solubility with temperature. Sixth, at Yucca Mountain, the calcite-silica deposits are known to occur throughout the entire thickness of the local vadose zone; they also occur below the local water table. As it may be observed in rock cores extracted from the Yucca Mountain bedrock, the subsurface veins are both texturally and compositionally similar to the local surficial fault infillings. Both of the calcite-silica deposits yield comparable uranium-thorium ages and, as was pointed out in the January 18 letter, carry comparable concentrations of oxygen-18 and carbon-13. There is no rational reason, therefore, to even suspect that the subsurface veins may have been produced via depositional processes that are distinct from those involved in the precipitation of the surficial fault infillings and veins.

To account for the clearly evident affinity, the descensum proponents are forced to carry the "pedogenic" actions well below the soil horizon, and even below the water table. For the per ascensum advocate, however, to describe this proposition as ridiculous is to elevate its status by undue flattery. Seventh, some of the Yucca Mountain calcites and opals occur in the form of the hydro-clastic breccia dike cements and of the hydraulic fracturing residua. Both of these forms may be directly observed on the eastern flank of the so-called Harper Valley, situated just west of Busted Butte. After examining the attached Figures 12 and 13, you may agree that the referenced features do indeed represent the per ascensum injections veins and dikes. To sum up my response to the third main objection, it seems appropriate to cite words once employed by Dr. T. R. Harper to describe his impressions after a rather extensive geological reconnaissance of the Yucca Mountain area. "It shouts at you, it screams at you--this was produced by a forceful injection of fluids."

After having completed somewhat lengthy treatment of the main objections, as expressed in the March 6 letter, allow me for addressing more interesting and, as far as the Yucca Mountain calcretes and veins are concerned, more important topics. Four of such topics need to be considered. These are: (1) the carbon isotopic compatibility between the parent fluids for the Yucca Mountain calcretes and veins and the per descensum pedogenic fluids, as observed and interpreted for the Yucca Mountain area; (2) the diagnostic usefulness of the isotopic composition vs. altitude gradients, as observed by you during isotopic studies of the local cobble encrustations; (3) the uranium-strontium-carbon isotopic affinity between the parent fluids for the Yucca Mountain calcretes and veins and the Nevada Test Site geothermal fluids; and (4) the origin of carbon that (a) is contained in the carbon dioxide respired from the Yucca Mountain vadose zone and (b) is dissolved in the Nevada Test Site subsurface fluids. In my opinion, the above four topics, if considered together with the results of oxygen-18 geothermal reconstructions, provide firm basis to elevate the per ascensum interpretations of the origin of the Yucca Mountain calcretes and veins to the secure beyond a reasonable doubt status. The oxygen-18 geothermal reconstructions have received a rather complete treatment in the January 18 letter, and there is no need of repeating it here. I do sincerely hope that, after carefully reviewing the subsequent discussions, you may alter your viewpoints regarding my professional conduct and, most importantly, geological processes that are expressed through the disputed calcretes and veins.

With reference to the first topic, please examine the attached Figures 14 through 19. In these figures, four main points are emphasized. These are: (1) the parent fluids for the Yucca Mountain calcretes and surficial veins have carried values of the $\delta^{13}\text{C}$ ratio ranging from -4.75 to -9.25 per mil PDB , Figure 15; (2) the mean values of the $\delta^{13}\text{C}$ ratio, that may be expected for the Yucca Mountain per descensum pedogenic fluids and produced through dissolution of the locally observed carbon dioxide, range from -9.36 to -11.50 per mil PDB , Figure 16; (3) the observed values of the $\delta^{13}\text{C}$ ratio, from samples of the contemporary soil and bedrock per descensum fluids, range from -9.59 to -13.05 per mil PDB , Figure 17; and (4) the expected values for the

Nevada Test Site per descensum pedogenic fluids, based on the results of laboratory soil-leaching experiments, range from -9.6 to -16.4 per mil P_{DB} , Figure 17. As illustrated in Figure 18, I hope with sufficient clarity, both the observed and the expected values of the $\delta^{13}C$ ratio, for the Yucca Mountain per descensum soil and bedrock fluids, are consistently and significantly lower than the corresponding ratios, as interpreted, for the parent fluids of the Yucca Mountain calcretes and veins. You may agree that the conclusion "the $\delta^{13}C$ range from -4.75 to -9.45 per mil P_{DB} , interpreted for the parent fluids of the disputed deposits, is too "heavy" to permit genetic association with the local per descensum pedogenic fluids alone" is secure beyond a reasonable doubt. You may also agree that, my initial estimate of ~ -14 per mil P_{DB} for the per descensum pedogenic fluids that owe their CO_2 content to biogenic activity alone, if adjusted by the rightfully pointed out error of +1.75 per mil P_{DB} , is not as erroneous as the March 6 letter indicates.

With reference to the second topic, please examine the attached Figures 20 through 28. In these figures, four main points are also emphasized. These are: (1) the mean value for the carbon-13 vs. altitude gradient, from samples of the local cobble encrustations, is $d \delta^{13}C/dz \sim 4$ per mil P_{DB} per 1 km change in altitude of the cobble encrustation sampling site, Figure 22; (2) the mean value for the oxygen-18 vs. altitude gradient, from samples of the local cobble encrustations, is $d \delta^{18}O/dz \sim 2$ per mil $SMOW$ per 1 km change in altitude of the cobble encrustation sampling site, Figure 23; (3) the mean value for the carbon-13 vs. altitude gradient expected for both the cobble encrustations and the distal calcretes, precipitated from the Nevada Test Site region groundwaters, is also $d \delta^{13}C/dz \sim 4$ per mil P_{DB} per 1 km change in altitude of the deposit precipitation site, Figures 24 and 25; and (4) the mean value for the oxygen-18 vs. altitude gradient expected for both the cobble encrustations and the distal calcretes, precipitated from the Nevada Test Site region groundwaters, is also $d \delta^{18}O/dz \sim 2$ per mil $SMOW$ per 1 km change in altitude of the deposit precipitation site, Figure 26. After considering the above four points, you may agree that, the assertion from the March 6 letter, "no gradient would be expected for carbonates precipitated from deep seated springs" is, at best, totally without merit. You may also agree that, the conclusion "either (1) the per ascensum process yields carbonates that, in terms of the isotopic composition vs. altitude gradients, are similar to those produced via the per descensum process or (2) the assumption of the per descensum origin for the local cobble encrustation is FALSE" deserves the secure beyond a reasonable doubt status. Finally, you may further agree that, the viewpoint expressed in the January 18 letter, "the per descensum interpretations, as proposed by Quade and Cerling, may hardly be regarded as an example of the most insightful and meticulous science," is a rather mild and reserved characterization of the quality of your work.

With reference to the third topic, please examine the attached Figures 29 through 51. In these figures, two broad theses are emphasized. First, it is shown that, based on a variety of data (hydraulic, geothermal, and geochemical), the Nevada Test Site hydrosphere may be regarded as consisting of two contrasting thermodynamic configurations. These are: (1) the

so-called hydraulic "source" regions; and (2) the so-called hydraulic "sink" regions. [Both the "source" and "sink" terms have been borrowed from topological studies of non-linear equations of motion, see Stewart (1989) for example. They are used in the context of the so-called phase-space portraits of the equations of motion. The phrase "phase-space," in turn, is used to denote a mathematical space containing dynamical variables of a system. For such a space, orthogonal coordinate directions represent dynamical variables used to specify the instantaneous state of the system.] The respective locations for both of the Nevada Test Site configurations are depicted in Figure 30. A summary of the hydraulic, geothermal, and geochemical characteristics for both of the distinguished configurations is presented in Figures 31 through 35. Please make a note that, relative to the "sink" fluids and regions, the hydraulic "source" fluids and regions exhibit: (1) the positive values of the vertical hydraulic gradients (dh/dz), Figure 31; (2) roughly, a factor of two higher values of the geothermal gradient (dT/dz), Figures 31 and 32; (3) on the average, a factor of ten higher concentrations of the dissolved $Cl^-+SO_4^{--}$ anions, Figure 33 [with reference to the Chebotarev's groundwater evaluation sequence, the "source" fluids seem to be affiliated more with the evolutionary stage II, whereas the "sink" fluids represent the evolutionary stage I]; (4) the significantly higher values of the calcite saturation index, Figure 34; and (5) the noticeably higher values of the gypsum saturation index, Figure 35. Please also note, from the attached Figures 36 through 44, that, relative to the hydraulic "sink" based fluids, the hydraulic "source" fluids carry: (1) a factor of ten, higher concentrations of the dissolved uranium, Figures 36 and 37; (2) noticeably and consistently, lower values of the $^{234}U/^{238}U$ activity ratio, Figure 38; (3) significantly higher values of the $^{87}Sr/^{86}Sr$ ratio, Figure 39; (4) noticeably and significantly lower concentrations of carbon-14, Figure 40; (5) significantly higher concentrations of carbon-13, Figures 41 and 42; (6) noticeably higher magnitudes of the "oxygen isotopic shift," Figure 43; and (7) negative values of the "hydrogen isotopic shift," Figure 44. After considering the above lines of evidence, you may agree that there is no rational basis to regard the Nevada Test Site hydrosphere as even loosely fitting the traditional patterns of hydrologic thoughts. Specifically, the Nevada Test Site hydrosphere deviates from such patterns in a number of significant ways including: (1) the thermally driven fluid motions are present; (2) such motions appear to be involving uncommonly deep parts of the lithosphere; and (3) shallow subsurface fluids appear to be spatially varying mixtures of, at least, two distinct end-members. Clearly, if one is sincerely and seriously interested in the origin of the Yucca Mountain calcretes and veins, the particular thermodynamic character of the Nevada Test Site hydrosphere must be considered. You may agree that the faithful reliance on inapplicable hydrologic concepts is not a prudent way to proceed in that regard. Second, the interpreted isotopic signatures for the parent fluids of the Nevada Test Site calcite-opaline silica deposits, including those occurring in the Yucca Mountain area, display a close uranium-strontium-carbon isotopic affinity with the observed and corresponding isotopic signatures from the Nevada Test Site geothermal or "source" fluids. Please make a note that, this isotopic affinity is simultaneously justified by the following four observations: (1) the adjusted values of the $^{234}U/^{238}U$ ratio, for radiogenic

production of ^{230}Th , interpreted for the parent fluids of the Yucca Mountain calcretes and veins are similar to those from samples of the Nevada Test Site geothermal fluids, Figures 45 and 46; (2) the adjusted U-content vs. $^{234}\text{U}/^{238}\text{U}$ field, for both the radiogenic production of ^{230}Th and the solid-fluid U-content partition, from samples of the Yucca Mountain calcretes and veins, is similar to that observed from samples of the Nevada Test Site geothermal fluids, Figure 48; (3) the observed range of values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, from samples of the Yucca Mountain calcretes and veins, is identical to that observed from samples of the Nevada Test Site geothermal fluids, Figure 49; and (4) the interpreted range of values of the $\delta^{13}\text{C}$ ratio, for the parent fluids of the Yucca Mountain calcretes and veins, is identical to that observed from samples of the Nevada Test site geothermal fluids, Figure 50. Combining these four observations with the results of oxygen-18 geothermal reconstructions, as presented in the January 18 letter, and considering the entire content of this letter, leads to the establishment of a rather firm foundation for judging the origin of the Yucca Mountain calcretes and veins. You may agree that, short of questioning validity of the entire data base, there is no choice but to regard the per ascensum origin conclusion as secure beyond a reasonable doubt. You may further agree that the viewpoint expressed in the January 18 letter "within the context of safety considerations of a high-level nuclear waste repository these interpretations [i.e., ones proposed by Quade and Cerling] may rightfully be regarded as a good example of irresponsible science" is justified.

Before addressing the last and somewhat diverging topic, allow me to complete the considerations of the origin and significance of the Yucca Mountain calcite-silica deposits by making a few closing remarks. In this context, please be advised that I do regard the isotopic demonstrations made by the U.S. Geological Survey (USGS) scientists, specifically that the "sink" fluids (i.e., the contemporary shallow fluids of the Yucca Mountain area) are isotopically distinct from the "source" fluids (i.e., the parent fluids for the Yucca Mountain calcite-silica deposits), as a valuable observation. Wholeheartedly, I do accept this observation as valid. I do have, however, a fundamental disagreement with the USGS interpretations of the meaning of this observation. I will elaborate. The underlying assumption for the totality of syllogism, as constructed by the USGS scientists, in support of the per descensum origin of the Yucca Mountain calcite-silica deposits, is that the Nevada Test Site hydrosphere is a steady-state, non-evolving dynamic system. If considered in the enlarging time-space, the behavior of such a system may be adequately represented solely by the so-called stable point attractor, Stewart (1989). This attractor state is maintained for perpetuity. You may agree that, it does not make a bit of sense, scientific or otherwise, to investigate the thermodynamic character of a dynamic system by assuming its behavior. In this situation, we are engaged in a circular reasoning--our conclusion is our assumption. To avoid this clearly unsatisfactory shortcoming, the thermodynamic character of a system must be defined independently, based on the observed behavior of this system. In this context, a variety of measurements and observations may be used to demonstrate that, for the Nevada Test Site hydrosphere, the assumption of the ubiquitous stable point attractor state is not valid. There are several lines of

reasoning and evidence that, not only justify but also, demand this viewpoint. These are: (1) the substantial flux of mass and energy from the sub-lithospheric mantle into the very thin lithosphere, Szymanski (1989); (2) the time-dependence of the hydraulic and effective thermal conductivity structures, Szymanski (1989); (3) the resulting time-dependence of the R_a - R_c difference, Szymanski (1989); (4) the observed self-organization produced macroscopic order, as demonstrated in this letter; (5) the results of spectral analyses of the oxygen-18 time series from the DH-2 calcite vein, as performed by Crowley and Howard (1990); and (6) the results of periodic measurements of chemical and isotopic composition of the Nevada Test Site subsurface fluids, as performed, for example, by Claassen (1973) and Lyles et al. (1990). All of these points may reasonably be taken to suggest that the Nevada Test Site hydrosphere is behaving as a non-monotonically evolving, non-equilibrium dissipative system, Nicolis and Prigogine (1977). In the enlarging time-space, the behavior of such a system may simultaneously be represented by all four of the attractor states, namely: (1) the stable point attractor; (2) the stable limit cycle attractor; (3) the two-dimensional torous attractor; and (4) the three-dimensional torous attractor, Ruelle and Takens (1971); Swinney and Gallub (1986); and Stewart (1989). Furthermore, under the influence of even very small perturbations, and with the presence of a positive feed-back mechanism, the system possessing the torous attractor states may be presumed to be "structurally unstable," i.e., subject to "sink" $\leftrightarrow$ "source" and "saddle" $\leftrightarrow$ "sink" ("source") transformations, Stewart (1989). It is here where the extraordinary significance of the Yucca Mountain calcite-silica deposits lies--we have a unique opportunity to examine the behavior of the Nevada Test Site hydrosphere from the point of view of non-linear thermodynamics. In this context, by demonstrating that: (1) the Nevada Test Site hydrosphere consists of the two contrasting thermodynamic configurations; (2) in addition to the stable point and limit cycle attractors, the Nevada Test Site hydrosphere possesses also the unstable torous attractors; (3) the isotopic compositions of the parent fluids, for the Yucca Mountain calcite-silica deposits, are incompatible with the corresponding compositions of the local "sink" fluids; and (4) the isotopic compositions of the parent fluids, for the Yucca Mountain calcite-silica deposits, are similar to those of the local "source" fluids, we are making two important points. These are: (1) the Nevada Test Site hydrosphere, indeed, is a non-equilibrium dissipative system; and (2) this system contains a positive feedback mechanism. Being a dynamic system with the above two characteristics, the Nevada Test Site hydrosphere exhibits: (1) self-organization; (2) uniform, periodic, and non-periodic motions; and (3) the structural transformations. After considering the above remarks you may agree that for myself, as the DOE bureaucrat, being totally unimpressed by the isotopic arguments, as put forth by the USGS scientists in support of the per descensum origin of the Yucca Mountain calcite-silica deposits, is a very reasonable course of action indeed. You may further agree that the USGS scientists, rather than focusing on discrediting Szymanski and his rather conservative yet readily verifiable viewpoints, would be well advised to become engaged in resolving more pressing issues instead. One area that particularly deserves their attention is an inquiry into a nature of the positive feedback mechanism, which evidently operates in the Yucca Mountain

area. Here, there are, at least, five interrelated candidate processes. These are: (1) injection of basaltic melt from below the Moho discontinuity; (2) injection of silicic melt from above the Conrad discontinuity; (3) the CO₂ gas assisted hydrothermal eruption; (4) the CO₂ gas assisted seismic pumping; and (5) ordinary seismic pumping, in association with large scale faulting. Relative to the former four processes, the latter process is a fairly benign one. While preparing the 1989 report and, at the same time, attempting to sound sufficiently, but not overly, alarming I have exclusively focused my attention on this process. Recently, however, I have received a well deserved, for a change, criticism. "Szymanski may have seriously, if not grossly, underestimated the hazardous conditions at Yucca Mountain in his 1989 report."

I do sincerely hope that, after carefully considering totality of the proceeding discussions, you may acquire some appreciation for my position that, as far as the per descensum origin of the Yucca Mountain calcite-silica deposits is concerned, there is no single line of evidence one may sincerely put forth in support of it. The Quade et al. (1990) per descensum conclusions, as demonstrated herein, are devoid of any scientific basis. The per descensum isotopic arguments, as advocated by the USGS scientists, are solely based on the demonstratively erroneously assumed thermodynamic character of the Nevada Test Site hydrosphere. The textural arguments (i.e., "the extreme fine-grained nature of the calcite, fine-grained silica, and the presence of pisoliths"), as put forth by the USGS scientists in support of the per descensum interpretations, are demonstratively FALSE. A few years ago, I have personally escorted some of the USGS per descensum advocates to the Tecopa, California, Warm Springs area. Here, it may be directly observed that the ongoing warm spring discharge activity is accompanied by: (1) precipitation of the micritic calcites; (2) precipitation of amorphous and, therefore, understandably "fine-grained" silica; (3) formation of the pisoliths through biochemical algal-encrustation processes; and (4) localized development of the carbonate cobble encrustations. Likewise, the morphological arguments (specifically, absence of the discernible spring mounds), as advanced by Dr. R. O. Fournier of the USGS, do not appear as particularly meritorious ones. A fairly reasonable justification for this statement may be acquired through a visit to, for example, the 1954 Dixie Valley earthquake epicentral area. Here, it may be directly observed that, the hot (near boiling) discharge of fluids is accompanied neither by the development of spring mounds nor by the formation of spring aprons; the hydrogenic mineralization is almost totally absent. Similarly, the spring mound absences may be directly observed in association with a number of local spring discharges. Examples of such springs are: (1) the Sarcobatus Flat-Beatty springs; (2) the Tecopa Warm Springs; and (3) the Nevada Test Site Cane Spring. Clearly, while dealing with the Yucca Mountain calcite-silica deposits, we are really confronted with, I should think, a fairly easy, for the scientists to make, choice. Demonstratively, this choice is between: (1) the popular and convenient, but unsupported and unverifiable, dogma; and (2) the troublesome even alarming, but documented and verified, scientific conclusion. Understandably, after having carefully weighed both sides of the Yucca Mountain deposits dilemma, I am left solely with a conjecture about the reasons for the continuing, and

apparently broad and intensely held, objections to the evidently correct per ascensum resolutions of this dilemma. It seems to me that, within the context of such a conjecture, it may be helpful to cite a passage from the recent book entitled "The Truth About Chernobyl" by Grigori U. Medvedev. "Unfortunately, there are still plenty of credulous people who are quite willing to conform. After all, it is easier to believe what you are told than to subject it to sober scrutiny. To begin with, it is less trouble."

With reference to the fourth and last topic, please examine the attached Figures 52 through 71. In these figures, two additional and again broad theses are advocated. First, the CO₂ isotopic data are used to demonstrate that, for the Yucca Mountain area, there is no rational reason to believe that the vadose zone CO₂ originates solely from the atmosphere-biosphere source. Please make a note that, the available CO₂ and geothermal data allow for the following observations to be made: (1) for the Yucca Mountain vadose zone, the non-periodic convective circulations of gaseous phases are inevitable, Figures 54 through 57; (2) the isotopic character of carbon contained in the vadose zone CO₂ changes and becomes "heavier" with the increasing depth, Figure 58; (3) the vadose zone CO₂ exhibits the depthward diminishing concentrations of carbon-14, Figure 59; and (4) values of the CO₂ partial pressure fluctuate about the mean atmospheric value of $\log P_{CO_2} \sim -3.5$ atm, Figure 60. Taken together these four observations lead to the establishment of a conceptual understanding for both the spatio-temporal distribution of the CO₂ indexes (del ¹³C, PMC, and $\log P_{CO_2}$), as observed in the Yucca Mountain vadose zone, and the origin of CO₂ respired from the Yucca Mountain vadose zone. The essence of such understanding is depicted in Figure 61. In accordance with the proposed understanding, the non-periodic convective circulations of the vadose zone gaseous phases, result in a dynamic mixing of the CO₂ gases that originate simultaneously from two independent sources. One of the CO₂ sources is situated at and near the topographic surface, and corresponds to the atmosphere-biosphere system. The other CO₂ source, the so-called subterranean CO₂ source, is situated along the local water table. The CO₂ gas provided by the discontinuous subterranean source originates from degassing of the local upwelling, and supersaturated with respect to calcite, geothermal fluids. It was pointed out by Szymanski (1989) that, both the geothermal data summarized by Sass et al. (1987) and the geochemical data summarized by Kerrisk (1987) indicate that, such fluids are currently upwelling along two local faults, specifically the Solitario Canyon Fault and the Paintbrush Fault. Second, the carbon isotopic data are used to demonstrate that, for the Nevada Test Site hydrosphere, the dissolved carbon does not originate solely from the atmosphere-biosphere CO₂ source. Please make a note that, for the Nevada Test Site hydrosphere, the available data allow for the following six observations to be made: (1) the hydrosphere→ vadose zone→ atmosphere gradient is negative, but not vice versa, Figure 62; (2) the hydraulic "source" fluids are supersaturated with respect to calcite, whereas their "sink" counterparts are undersaturated, Figure 63; (3) relative to the "sink" fluids, the hydraulic "source" based fluids carry lower concentrations of carbon-14, Figure 64; (4) relative to the "sink" fluids, the hydraulic "source" based fluids carry significantly higher values of the del ¹³C ratio, Figures 65 and 66; (5) relative to the tuff "pile" based fluids,

the Paleozoic carbonates based fluids carry significantly higher values of the $\delta^{13}\text{C}$ ratio, Figures 65 and 66; and (6) the Paleozoic carbonates based fluids carry values of the $\delta^{13}\text{C}$ ratio that are conspicuously lower than the slightly positive values expected for fluids equilibrated with the marine limestones, Figure 65. Now, let's see what all of the six observations may be telling us. The observation that, the upwelling geothermal fluids typically yield positive values of the calcite saturation index, if coupled with the observed direction of the $\log P_{\text{CO}_2}$ gradient, may reasonably be taken to indicate that, at the Nevada Test Site, local degassing of CO_2 from the hydrosphere is taking place. The observation, that the hydraulic "source" fluids carry both the lower concentrations of carbon-14 and the higher values of the $\delta^{13}\text{C}$ ratio, may reasonably be taken to indicate that, for the Nevada Test Site, the atmosphere-biosphere CO_2 source is not a decisive factor in controlling the isotopic character of carbon dissolved in the ascending geothermal fluids. The atmosphere-biosphere CO_2 source, however, is a significant factor, but only for the descending "sink" fluids. The observation that, relative to the tuff "pile" based fluids, the Paleozoic carbonates based fluids carry higher values of the $\delta^{13}\text{C}$ ratio may reasonably be taken to indicate that some of carbon, dissolved in the Nevada Test Site subsurface fluids, is acquired through dissolution of the local marine limestones. The observation that, relative to the expected values of the $\delta^{13}\text{C}$ ratio for fluids equilibrated with marine limestones, the Paleozoic carbonates based fluids carry abnormally low values of the $\delta^{13}\text{C}$ ratio does not have an unequivocal interpretation. To account for this important observation, however, two different interpretations may be put forth. On the one hand, it may be assumed that fluids residing in the Paleozoic carbonates do carry significant concentrations of old and isotopically "light" carbon acquired from the atmosphere-biosphere CO_2 source. On the other hand, however, it is equally valid to likewise speculate that, at the Nevada Test Site, there is an additional source of CO_2 . This additional CO_2 source may be either the partially-incipiently molten sub-lithospheric, but very shallow, mantle, EOS (1990), or therefrom derived igneous bodies residing in the very thin (~25km) lithosphere. It may be important to note that the local presence of, at least, three such bodies may be interpreted based on the results of both the P-wave residuals study of teleseismic events, as reported by Monfort and Evans (1982), and the seismic reflection survey ran in Amargosa Desert, and reported by Brocker et al. (1989). Furthermore, the partially-incipiently molten state of the local sub-lithospheric mantle is indicated by both the direct field observations regarding rather conspicuous presence of the Plio-Quaternary volcanism [yielding Hy-to Ne-normative alkali basalts and hawaiites] and the results of seismic tomography studies of the sub-lithospheric mantle, Monfort and Evans (1982); Szymanski (1989); and EOS (1990). The attached Figures 67 through 70 indicate that, it is entirely feasible and proper to explain the abnormally low values of the $\delta^{13}\text{C}$ ratio, as observed for the Paleozoic carbonates based geothermal fluids, by postulating the partial involvement of the sub-lithospheric mantle indigenous CO_2 . After reviewing both of the above carbon isotopic considerations, you may agree that there is little merit in assuming that, at the Nevada Test Site, carbon, which (1) is contained in the CO_2 gas respired from the vadose zone and (2) is dissolved in the local hydrosphere, originates solely from the

atmosphere-biosphere source. You may also agree that this dubious assumption is a cornerstone for the per descensum considerations of the origin of both the local cobble encrustations and the Yucca Mountain calcretes and veins, as advocated by Quade et al. (1988 and 1989) and Quade and Cerling (1990). My dear doctors, without adequately understanding the origin of the CO₂ gas respired from the vadose zone, it is totally inappropriate to: (1) estimate the percentage of biomass with the C-3 metabolic pathway, based on the observed isotopic composition of the soil CO₂; (2) introduce the dubious concept of the "desert ecosystem" carbon-13 enrichment based on the observed, but seemingly abnormally "heavy," isotopic composition of the soil CO₂, and (3) interpret the observed discrepancies, between the observed carbon-13 content of the soil CO₂ and the expected carbon-13 content, as resulting from the "diffusional enrichment of carbon-13 in soils."

After having performed twice fairly involved analyses of your published work, I feel entitled to offer a two-fold tutorial sermon. On the one hand, we the scientists (you) and the bureaucrats (me) alike, being engaged and entrusted with execution and management of the project with potentially catastrophic biological consequences are obligated to: (1) think in terms of the "worst case scenario"; (2) examine all pertinent evidence, in particular if such evidence is readily accessible; and (3) keep an open mind, and even if only marginally justified, maintain multiple working hypotheses. Within the context of these remarks, I cannot help but to express my deep disappointment that you have: (1) rather than seriously consider the isotopic arguments presented in the January 18 letter, elected instead to dismiss these arguments as either "irrelevant" or not "closely related to our work"; and (2) rather than seriously consider the results of geothermal reconstructions offered in the January letter, decided instead to focus on dubious and rather silly isotopic corrections, and on falsely alleged "frequent misquoting of others." Both the isotopic arguments and the geothermal reconstructions have a clear and direct bearing on YOUR conclusions regarding both the origin of the Yucca Mountain calcretes and veins and the likelihood of occurrence, at the Yucca Mountain site, of potentially catastrophic upwelling of geothermal fluids. These conclusions have been published in the highly respected and influential scientific journal. As such, your demonstratively erroneous conclusions will, undoubtedly, be used to further the misguided U.S. Government commitment to the questionable Yucca Mountain site. The least you could do is to thoroughly examine, perhaps by your standards deficient, but honest and sincere effort made by the DOE bureaucrat who is directly charged with the responsibility of reviewing and understanding the results of your scientific work. On the other hand, while being students at the University of Warsaw, Poland, we were very firmly advised by members of the faculty that, if participating in a scientific debate, it is totally unacceptable to question the opponent's character and deliberately cast doubts on his competence and sincerity, rather than systematically exposing flaws in the logic and factual basis employed by him. I do strongly feel that, this firm advice is worth sharing with you and with your colleagues from the USGS. Please, let's keep debating scientific merits of the disputed issue, while refraining from personal characterizations. Short of you following this request, I will see no alternative, but to withdraw from further arguments.

In closing, I for one do not see a good reason to either alter or withdraw any of the main points made in the January 18 letter. To the contrary, within the context of additional analyses as presented herein, these points become further reinforced. Once more, I would like to express my appreciation for your continued interest in the dilemma presented by the Yucca Mountain calcretes and veins. Should you wish to discuss this dilemma further, or have any questions regarding this letter, please do not hesitate to contact me.

Sincerely,

Jerry S. Szymanski, Physical Scientist
Technical Analysis Branch
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U.S. Department of Energy

RSED:JSS-3557

Enclosures:

1. Ltr, 1/8/91, Livingston to
Distribution
2. Ltr, 2/5/91, Livingston to
Distribution
3. Ltr, 4/30/91, Livingston to
Distribution
4. List of Figures
5. Figures 1 through 71
6. References

P.S.

Incidentally, you may find opinions expressed by another DOE employee, Dr. Donald E. Livingston, to be of some relevance with regard to our dispute. These opinions pertain to the January 18 letter and to the cited by you per descensum interpretations performed by Dr. J. S. Stuckless of the USGS. In contrast to myself, Dr. Livingston is a geochemist with some 35 years of experience in various areas of isotopic geology. After careful considerations of both the direct field observations and the uranium-strontium-carbon-oxygen isotopic data, he has drafted the attached three memoranda.

You may also find a thought once expressed by Leo Tolstoy to be of some relevance with regard to our dispute. "I know that most men, including those at ease with problems of the greatest complexity, can seldom accept even the simplest and most obvious truth if it be such as would oblige them to admit the falsity of conclusions they reached perhaps with great difficulty, conclusions which they have delighted in explaining to colleagues, which they have proudly taught to others, and which they have woven, thread by thread, into the fabric of their lives."

cc w/encls:

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QA: N/A

[illegible]

TECHNICAL MEMORANDUM REGARDING SITE SUITABILITY OF YUCCA MOUNTAIN

In view of the above, I see an important opportunity for the U.S. Department of Energy (DOE) to take the initiative to confirm the hydrologic conditions that must be anticipated at Yucca Mountain for engineering design and performance assessment. I recommend that DOE negotiate with Dr. Brian Smith of the Lawrence Berkeley Laboratory and his assembled colleagues to conduct preliminary investigations of the Skull Mountain and Oasis Valley localities to assess their hydrologic circumstances.

If you have any questions, please contact me at 794-7944.

"ORIGINAL SIGNED BY"

Donald E. Livingston
Technical Analysis Branch
Regulatory & Site Evaluation Division
Yucca Mountain Site Characterization
Project Office

RSED:DEL-1545

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WBS 1.2.3.6.2
QA: N/A

FEB 05 1991

David C. Dobson, YMP, NV
J. Russell Dyer, YMP, NV

TRANSMITTAL OF DRAFT CRITIQUE OF "AN EVALUATION OF EVIDENCE TO THE ORIGIN OF VEIN DEPOSITS IN TRENCH 14, NEVADA TEST SITE, NEVADA, BY JOHN S. STUCKLESS

This memo transmits my draft critique (enclosure 2) of the subject document (enclosure 3) requested orally by David Dobson in November 1990. This version of the critique is somewhat expanded over the version informally submitted on December 26, 1990.

The subject document attempts to "preclude" the origin of vein materials at Trench 14 by upwelling groundwaters (page 1, paragraph 2, abstract; page 16, paragraph 1, summary) and to "show that the carbonate and opaline silica deposits exposed in Trench 14 must have formed by a pedogenic process" (page 1, paragraph 3, abstract). The implication is that the Trench 14 veins were formed by sensu stricto pedogenic processes; i.e., eolian, biogenic and meteoric processes which further implies the same genesis for all other carbonate materials and most other silica materials at Yucca Mountain. The article fails in any such demonstration.

From the title of the document it is certain that the subject of the paper is one of the most prominent, contested and timely issues of the Yucca Mountain Site Characterization Project (YMP). As such, the subject should be treated in the most comprehensive and scientific manner possible. Instead, this paper has dealt with the subject matter in a most parochial, myopic and subjective fashion. All related scientific information, from whatever source, should have been brought to bear on the subject.

This document fails to take into consideration major published reviews regarding the O-18 and C-13 content of travertine and caliche deposits throughout the world. It also fails to consider the vast amounts of available deuterium and O-18 data for meteoric precipitation and groundwater from the southern Nevada region. This document fails to consider morphological and geological evidence from unequivocal spring deposits and mineralized veins in Death Valley, California, at Bare Mountain and elsewhere. Even less excusable, this document fails to consider kinetic, nonequilibrium isotopic fractionation of low temperature irreversible geochemical processes.

Of particular importance are the results of the geochemical survey conducted by the Nevada Bureau of Mines and Geology, Castor, Feldman and Tingley (1989) for the YMP. That report was prepared to support the U.S. Department of Energy (DOE) application for the administrative land withdrawal of the Yucca Mountain addition. To Table 1 of that report (enclosure 1), I have added the maximum geochemical results of ore metals and pathfinder metals. Even though

Multiple Addressees

-2-

these results strongly suggest a lack of economic mineral deposits at Yucca Mountain, the maximum values clearly demonstrate that mineral bearing, epigenetic (post late miocene age), hydrothermal solutions have penetrated porous and factured portions of Yucca Mountain to the level of the present day surface.

A longer list of criticisms are documented in the present version of my critique which is enclosed. Also, much if not most of the data on which the article is based are not available in the Site and Engineering Properties Data Base, in open file reports nor publications.

I see two options that DOE and U.S. Geological Survey (USGS) can follow:

1. Request that the author withdraw this paper which may cause hard feelings within the USGS and DOE, but would forestall any public embarrassment for both organizations.
2. Not request withdrawal of the paper, but publicly disclaim any DOE or USGS position regarding conclusions reported in the paper after it has been presented and published.

Donald E. Livingston
Technical Analysis Branch
Regulatory & Site Evaluation Division

RSED:DEL-2046

Enclosures:

1. Geochemical Results from
Castor, Feldman and
Tingley (1989)
2. Critique
3. Subject Draft Report

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APR 10 1991

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NEW GEOLOGICAL FIELD OBSERVATIONS REGARDING SUITABILITY OF THE YUCCA MOUNTAIN SITE

Over that past few weeks, since my memorandums of January 8, 1991, and February 5, 1991, I have continued with weekend field reconnaissance trips to observe the geological evidence relating to geochemical data from Yucca Mountain. It is my professional responsibility to inform my management of my current and rapidly evolving concerns.

1. I have seen geomorphic, stratigraphic, mineralogical and structural evidence for Quaternary ground water discharge in the southern portion of Yucca Mountain.
2. I have seen structural, stratigraphic and geomorphic evidence of major Quaternary faulting in the southern portions of Yucca Mountain.
3. I have seen structural and stratigraphic evidence for injection breccias which seem to be related to mosaic breccias in Trench 14 that include zircons of possible Quaternary and older ages.
4. I have seen field evidence which indicates post-Topopah and post-Tiva Canyon intrusion of rhyolite and rhyolite tuff. The minimum age of intrusion is not known.

The above geological features indicate that Jerry Szymanski may have seriously, if not grossly, underestimated the hazardous conditions at Yucca Mountain in his 1989 report.

I recommend, as a verification of the above observations, that within the next two to three months the full complement of the geologically trained staff and management of the Regulatory and Site Evaluation Division and the Project Manager accompany me for at least a two-day field trip to observe these geological features. Each of the members of the staff and management should then prepare a one- to three-page report of observations for the Project Manager. Each report should include recommendations of a responsible course of action for the U.S. Department of Energy.

APR 10 1991

Multiple Addressees

-2-

In my 35 years of geological experience in the southwestern United States, I have never seen such an abundance of such diverse and clear geological evidence that collectively indicate such young and severe geological conditions. In the coming months I plan to continue this field reconnaissance and will keep you fully informed.

Respectfully,

Donald E. Livingston
Technical Analysis Branch
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RECOMMENDATION OF A PRUDENT COURSE OF ACTION FOR THE U.S. DEPARTMENT OF
ENERGY, YUCCA MOUNTAIN SITE CHARACTERIZATION PROJECT OFFICE (YMPO)

References: (1) Ltr, Livingston to Distribution, dtd 1/8/91
(2) Ltr, Livingston to Distribution, dtd 2/5/91
(3) Ltr, Livingston to Distribution, dtd 4/10/91

Over the past several months I have kept management fully informed of my growing concerns regarding the suitability of Yucca Mountain as a potential repository site and my concerns as to the nature of some of the studies being conducted. In addition, I have grave concerns of a different kind. Presently, I foresee in the offing the development of a raucous and public debate within the forum of the news media that will result in severely detrimental individual, institutional and political consequences for all involved. In my view, this ugly scenario can be forestalled by a modest and prudent course of action by the YMPO.

My views regarding the geochemical and isotopic characteristics of the site and my views of the character of some of the ongoing studies have been noted in my referenced letters (references 1 and 2). These views have been fully confirmed by field evidence (reference 3) and by the further analysis of the isotopic, geochemical and field data that is ongoing by Jerry Szymanski. This analysis, in part, has been presented to the members of the review panel from the National Academy of Sciences and, in part, will be distributed by means of Jerry Szymanski's letter to Thure E. Cerling and Jay Quade.

Again, I recommend that the YMPO initiate a modestly funded and prudently independent effort, to last about three years, that would investigate numerous indicated potentially hazardous conditions at the Yucca Mountain site. This effort should begin at the earliest opportunity. In my letter of December 4, 1990, I recommended funding for scientists of Lawrence Berkeley Laboratory and associates to conduct certain site suitability studies (enclosure). I stand by that recommendation. This effort can be efficiently conducted at a funding level of about \$500,000 per fiscal year for about three years. In addition, subsequent field observations in southern Yucca Mountain (reference 3) have indicated a dire need for more detailed field mapping of the bedrock exposures and Pliocene and Quaternary alluvial stratigraphy in this area in order to develop a more realistic and better documented tectonic model and geologic history for Yucca Mountain. A two-year effort at an estimated cost of \$200,000 per year should provide the required information. This additional recommendation should provide the necessary structural and geologic context required for the previously recommended studies.

Multiple Addressees

-2-

The three most prominent conditions that must be investigated at Yucca Mountain are:

1. the nature of calcite-silica veins and calcite-silica cemented alluvial and colluvial deposits,
2. the production and nature of carbon dioxide gas in the hydrosphere and the vadose zone, and
3. the age and nature of apparent post-Tiva Canyon member rhyolite intrusions at Yucca Mountain.

After duly pondering the management of this effort, by considering what I know of the background, responsibilities, and circumstances of each one the members of the staff and management of the Regulatory and Site Evaluation Division, I confidently conclude that I am best suited for this endeavor and I solicit such support from each of you as you consider these matters.

As manager of the effort, I would insist on a quarterly presentation and review of progress, status, and plans before YMPO management, including the Technical Project Officers of each participant organization so that planning could be conducted accordingly.

I am available to discuss these matters with each of you at your convenience.

Respectfully,

Donald E. Livingston
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Enclosure:
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LIST OF FIGURES

- Figure 1 Estimate of proportion of the C-3 biomass, based on the results of plant surveys.
- Figure 2 Explanation of the lithologic notations as employed by Hay et al. (1986).
- Figure 3 Isotopic characters of carbon and oxygen contained in samples of the Amargosa Basin *per ascensum* deposits.
- Figure 4 Carbon isotopic comparison, the Amargosa Basin *per ascensum* deposits and the local cobble encrustations.
- Figure 5 Oxygen isotopic comparison, the Amargosa Basin *per ascensum* deposits and the local cobble encrustations.
- Figure 6 The isotopic compositions vs. morphological form gradients, the Yucca Mountain calcite-silica deposits.
- Figure 7 Computation of the oxygen -18 normalization factors for the Yucca Mountain calcite-silica deposits.
- Figure 8 The normalized oxygen -18 comparison, the Amargosa Basin *per ascensum* deposits and the Yucca Mountain calcite-silica deposits.
- Figure 9 Oxygen isotopic file - isotopic character of oxygen contained in carbonate gangue veins from the Carlin and Cortez gold deposits.
- Figure 10 Isotopic comparative analyses - the Carlin and Cortez carbonate gangue minerals, the Amargosa Basin *per ascensum* deposits, and the reference cobble encrustations.
- Figure 11 Isotopic comparative analyses - the normalized Carlin and Cortez carbonate gangue minerals, the Yucca Mountain surficial and subsurface calcite-opaline silica deposits, and the reference cobble encrustations.
- Figure 12 Hydraulic fracture calcite residuum - eastern flank of the Harper Valley, southern part of Yucca Mountain.
- Figure 13 Hydro-clastic injection breccia dikes - eastern flank of the Harper Valley, southern part of the Yucca Mountain area.
- Figure 14 **Topic #1 - Evaluations of the carbon isotopic compatibility, between the interpreted isotopic composition of the parent fluids for the Yucca Mountain calcite-silica deposits and the observed and interpreted isotopic compositions of the Yucca Mountain *per descensum* pedogenic fluids.**
- Figure 15 Statement of the issue - topic #1.
- Figure 16 The interpreted range of values for the $\delta^{13}\text{C}$ ratio - the parent fluids for the Yucca Mountain calcretes and surficial veins.
- Figure 17 Isotopic character of carbon dissolved in the *per descensum* pedogenic fluids, interpreted based on the observed $\delta^{13}\text{C}$ ratios from samples of the Nevada Test Site vadose zone gases.

LIST OF FIGURES

- Figure 17 Isotopic character of carbon dissolved in the *per descensum* pedogenic fluids, based on direct measurements and on soil-leaching experiments.
- Figure 18 Summary - comparison of carbon isotopic compositions, the *per descensum* pedogenic fluids and the parent fluids for the Yucca Mountain calcretes and surficial veins.
- Figure 19 Conclusion - evaluations of the carbon isotopic compatibility, between the parent fluids for the Yucca Mountain calcretes and surficial veins and the local *per descensum* pedogenic fluids.
- Topic #2 - Evaluations of the diagnostic usefulness of the isotopic compositions vs altitude gradients from the local cobble encrustations.**
- Figure 20 Statement of the issue - topic #2.
- Figure 21 Carbon and oxygen isotopic data. Cobble encrustations - region around the Nevada Test Site.
- Figure 22 Isotopic character of carbon contained in the cobble encrustations as a function of altitude of land surface.
- Figure 23 Isotopic character of oxygen contained in the cobble encrustations as a function of altitude of land surface.
- Figure 24 Isotopic character of carbon dissolved in the Nevada Test Site subsurface fluids, as a function of altitude of land surface, and the expected $\delta^{13}\text{C}$ vs. altitude gradient and field for the *per ascensum* cobble encrustations and distal calcretes.
- Figure 25 Isotopic character of carbon dissolved in the Nevada Test Site calcium-rich subsurface fluids ("source" plus carbonate rock based fluids), as a function of altitude of land surface, and the expected values of the $\delta^{18}\text{O}$ vs altitude gradient and field for the *per ascensum* cobble encrustations and distal calcretes.
- Figure 26 Isotopic character of oxygen contained in the Nevada Test Site calcium-rich subsurface fluids ("source" plus carbonate rock based fluids), as a function of altitude of land surface, and the expected $\delta^{18}\text{O}$ vs altitude gradient and field for the *per ascensum* cobble encrustations and distal calcretes.
- Figure 27 Summary - comparative analyses of the isotopic compositions vs altitude gradients, the observed cobble encrustations and the expected *per ascensum* cobble encrustations and distal calcretes.
- Figure 28 Overall conclusions - comparative analyses of the isotopic compositions vs altitude gradients, the observed cobble encrustations and the expected *per ascensum* cobble encrustations and distal calcretes.
- Topic #3 - Evaluations of the uranium-strontium-carbon isotopic affinity, between the parent fluids for the Yucca Mountain calcite-silica deposits and the Nevada Test Site geothermal fluids.**
- Figure 29 Statement of the issue - topic #3.
- Figure 30 Inferred boundary conditions. The Nevada Test Site hydrosphere.
- Figure 31 Boundary conditions. Interpretation bases. The Nevada Test Site hydrosphere.

LIST OF FIGURES

- Figure 32a Downhole temperature profiles. Southern Indian Springs Valley "sink" region.
- Figure 32b Downhole temperature profiles. Rainier Mesa - Yucca Flat hydraulic "sink" regions.
- Figure 32c Downhole temperature profile. Borehole UE-25a3. North-western margin of the Skull Mountain hydraulic "source" region.
- Figure 32d Downhole temperature profiles. Boreholes J-11, TWF, and TW3. Skull Mountain hydraulic "source" region.
- Figure 32e Downhole temperature profiles. Pahute Mesa hydraulic "sink" and "source" regions.
- Figure 33a Groundwater chemistry. Hydraulic "sink" and dilated regions. The Nevada Test Site hydrosphere.
- Figure 33b Groundwater chemistry. Hydraulic "source" regions. The Nevada Test Site hydrosphere.
- Figure 34 Calcite saturation index. Water samples from the Nevada Test Site hydraulic "sink" and "source" regions.
- Figure 35 Gypsum saturation index. Water samples from the Nevada Test Site hydraulic "sink" and "source" regions.
- Figure 36 $^{234}\text{U}/^{238}\text{U}$ vs U-content field. Hydraulic "sink" and dilated regions. The Nevada Test Site hydrosphere.
- Figure 37 $^{234}\text{U}/^{238}\text{U}$ vs U-content field. Hydraulic "source" regions. The Nevada Test Site hydrosphere.
- Figure 38 Isotopic character of the dissolved uranium. Water samples from the hydraulic "source" and "sink" regions. The Nevada Test Site hydrosphere.
- Figure 39 Comparison of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. The Nevada Test Site "source" and "sink" regions.
- Figure 40 $\delta^{13}\text{C}$ vs PMC field. Water samples from the Nevada Test Site hydrosphere.
- Figure 41 Isotopic character of the dissolved carbon. Water samples from the Paleozoic carbonates based hydraulic "sink" and "source" regions.
- Figure 42 Isotopic character of the dissolved carbon. Water samples from the alluvium - tuff pile based hydraulic "source" and "sink" regions.
- Figure 43 Comparison of magnitudes of the oxygen isotopic shift. The Nevada Test Site "sink" and "source" regions.
- Figure 44a Relationship between the observed isotopic compositions, the meteoric water line, and altitude of the local land surface. Fluids from the hydraulic "sink" regions. The Nevada Test Site hydrosphere.
- Figure 44b Relationship between the observed isotopic compositions, the meteoric water line, and altitude of the local land surface. Fluids from the hydraulic "source" regions. The Nevada Test Site hydrosphere.

LIST OF FIGURES

- Figure 44c Comparison of isotopic compositions. "Sink" and "source" regions - the Nevada Test Site hydrosphere.
- Figure 45 $^{234}\text{U}/^{238}\text{U}$ and $^{230}\text{Th}/^{234}\text{U}$ activity ratios. The Nevada Test Site calcite-silica deposits.
- Figure 46 Comparison of the $^{234}\text{U}/^{238}\text{U}$ activity ratios. The Nevada Test Site calcite-silica deposits and the local geothermal fluids.
- Figure 47 Total U-content vs percentage of acid-soluble fraction. Calcite-silica deposits from the Nevada Test Site.
- Figure 48 Comparison of $^{234}\text{U}/^{238}\text{U}$ ratio vs U-content fields. The Nevada Test Site calcite-silica deposits and the Nevada Test Site subsurface fluids.
- Figure 49 Comparison of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. The Yucca Mountain calcite-silica deposits and the Nevada Test Site subsurface fluids.
- Figure 50 Comparison of the $\delta^{13}\text{C}$ ratios. The Yucca Mountain calcite-silica deposits and the Nevada Test Site subsurface fluids.
- Figure 51 Overall conclusions - evaluations of the uranium-strontium-carbon isotopic affinity, between the parent fluids for the Yucca Mountain calcite-silica deposits and the Nevada Test Site geothermal fluids.
- Topic #4 - An inquiry into the origin of carbon that is contained in the CO_2 gas respired from the Yucca Mountain vadose zone and is dissolved in the Nevada Test Site hydrosphere.**
- Figure 52 Statement of the issue - topic #4.
- Figure 53 General remarks - interpretation of the origin of CO_2 respired from the Yucca Mountain vadose zone.
- Figure 54 Introduction - interpretation of the results of studies of spatio-temporal distribution of the carbon indexes in the Yucca Mountain vadose zone.
- Figure 55 Location of the CO_2 study area, relative to the *in-situ* temperature at a depth of 350m. The Yucca Mountain vadose zone.
- Figure 56 Non-periodic convective flow.
- Figure 57 Interpretation key - origin of CO_2 respired from the Yucca Mountain vadose zone.
- Figure 58 Isotopic character of carbon contained in carbon dioxide from the vadose zone as a function of depth.
- Figure 59 The carbon -14 content in the carbon dioxide from the vadose zone as a function of depth.
- Figure 60 Log P_{CO_2} as a function of depth and time. The carbon dioxide from the Yucca Mountain vadose zone.

LIST OF FIGURES

- Figure 61 Conceptual understanding, the spatio-temporal distribution of the carbon indexes - the Yucca Mountain vadose zone.
- Figure 62 Histogram. Partial pressure of CO₂. The Nevada Test Site hydrosphere.
- Figure 63 Calcite saturation index. Water samples from the Nevada Test Site hydraulic "sink" and "source" regions.
- Figure 64 $\delta^{13}\text{C}$ vs PMC field. Water samples from the Nevada Test Site hydrosphere.
- Figure 65 Isotopic character of carbon. Water samples from the Paleozoic carbonates based hydraulic "sink" and "source" regions.
- Figure 66 Isotopic character of carbon. Water samples from the alluvium - tuff "pile" based hydraulic "source" and "sink" regions.
- Figure 67 Plausibility assessment - hypothesis that the abnormally low values of the $\delta^{13}\text{C}$ ratio, for the Paleozoic carbonates based "source" fluids, may be reflecting influx of the sub-lithospheric mantle indigenous CO₂.
- Figure 68 Further plausibility assessment - hypothesis that the abnormally low values of the $\delta^{13}\text{C}$ ratio, for the Paleozoic carbonates based "source" fluids, may be reflecting influx of the sub-lithospheric mantle indigenous CO₂.
- Figure 69 The isotopic character of carbon contained in the carbonate gangue minerals associated with the Carlin and Cortez gold deposits.
- Figure 70 The isotopic character of carbon contained in samples of the carbonate gangue minerals associated with various hydrothermal ore deposits.
- Figure 71 Overall conclusions - an inquiry into the origin of carbon that is contained in the CO₂ gas respired from the Yucca Mountain vadose zone and is dissolved in the Nevada Test Site hydrosphere.

the cobble encrustation collection site	SM-1	SM-2	SM-3	SM-4	SM-5	SM-6
site altitude [meters]	840	1550	1900	2270	2490	2740
percentage of C-3 plants - plant survey [%]	93-95	78	79	100	100	100
percentage of C-3 plants - soil CO ₂ data [%]	50	63	73	100	100	100

Note:

- a) the results of actual plant surveys indicate that, around the Nevada Test Site, a minimum value for the proportion of plants with the C-3 metabolic pathway is 78 percent;
- b) a reasonable estimate for the mean altitude of the Yucca Mountain area is ~ 1200 m;
- c) assuming that the proportions of the C-3 plants vary linearly with altitude (between 840 and 1550 m altitudes), a reasonable estimate for the Yucca Mountain C-3 proportion is ~ 85 percent; and
- d) there is no assurance that the soil CO₂ originates solely from the atmosphere - biosphere source and, consequently, estimates of proportions of plants with the C-3 metabolic pathway, based on the soil carbon dioxide data, are of doubtful validity and may lead to a circular reasoning.

Estimate of proportion of the C-3 biomass for the Yucca Mountain area, based on the results of plant surveys. Data from Quade et al. (1989).

- a) Composition of carbonate minerals in bedded claystones and carbonate rocks. Dolomite is represented by triangles and calcite by circles, which are closed for limestones of Tld. Tpa samples are of chalky limestones, most of which are soft; Tld samples are mostly of dense nodular and fenestral limestone; and Tpl samples include both carbonate rocks and carbonate minerals disseminated in claystones.

- b) Composition of calcite and dolomite in caliche breccia. Solid circles represent caliche breccia from East Playa, and open circles represent samples from the westernmost caliche breccia. Dolomite is from caliche breccia of East Playa.

Diagram showing the isotopic composition of carbonate minerals in Pliocene deposits of the Amargosa basin.

Explanation of the lithologic notations as employed by Hay et al. (1986). From Hay et al. (1986)

Note:

- the overlapping $\delta^{13}\text{C}$ vs $\delta^{18}\text{O}$ fields indicate that, all four of the distinguished lithofacies were precipitated from the common *per ascensum* parent fluids;
- the Tpa and Tld calcites represent proximal parts of the spring-fed depositional basin, while the Tpl calcites and dolomites represent the corresponding distal parts, as discussed by Hay et al. (1976);
- the observed isotopic composition vs lithofacie gradients may reasonably be taken as recording progressing alteration of: i) mineral precipitation environment [mainly the mineral precipitation temperature]; and simultaneously ii) the isotopic composition of the former pristine deep-seated fluids;
- the observed oxygen -18 content vs lithofacie gradient, from 15.5 to 26 per mil SMOW , may reasonably be taken to indicate that, for distal lithofacies, both the mineral precipitation temperature and the degree of oxygen -18 evaporative enrichment were respectively lower and higher than those for the proximal lithofacies;
- the observed carbon -13 content vs lithofacie gradient, from -3 to +2.5 per mil PDB , may reasonably be taken to indicate that, for the distal lithofacies, the diffusional carbon -13 enrichment (resulting from the CO_2 degassing) was higher than that for the proximal lithofacies;
- it is totally unreasonable to propose that, the isotopically "heavier" fluids [ones involved in precipitation of the distal lithofacies] were the *per descensum* pedogenic fluids;
- both the lithologic data and the isotopic data, as reported by Hay et al. (1986), provide direct and clear testimony that, after being discharged at the topographic surface, the pristine *per ascensum* fluids undergo, as expected, the progressive alteration of their initial stable isotopic contents;
- for dispositional sites with a fairly constant altitude, it may reasonably be presumed that the isotopic composition vs lithofacie gradients are the characteristic and diagnostic feature for the *per ascensum* deposits; and
- i) excluding the carbon-oxygen isotopic data, from samples of the Amargosa Basin distal lithofacies; ii) emphasizing, thereafter, the isotopic differences between the Amargosa Basin proximal lithofacies and the Yucca Mountain distal calcretes; and iii) ignoring the respective differences in the lithology of bedrock and in the resulting carbon -13 concentrations, as performed by Quade and Cerling (1990), may hardly be regarded as examples of "the most insightful and meticulous science".

Isotopic characters of oxygen and carbon contained in samples of the *per ascensum* Amargosa Basin deposits. Isotopic data are from Hay et al. (1986).

Explanation:

- - calcareous bedrock;
- - non-calcareous bedrock; and

horizontal bar represents the Amargosa Basin *per ascensum* deposits.

Note:

- a) the cobble encrustation carbon -13 data are from Quade et al. (1988) and the Amargosa Basin data are from Hay et al. (1986);
- b) samples of both the local cobble encrustations and the Amargosa Basin *per ascensum* deposits yield similar values of the $\delta^{13}\text{C}$ ratio; and
- c) either the *per ascensum* process yields carbonates that, in terms of the carbon -13 contents, are similar to those produced via the *per descensum* process or the assumption of the *per descensum* origin of the local cobble encrustations is FALSE.

Carbon isotopic comparison, the Amargosa Basin *per ascensum* deposits and the local cobble encrustations.

Explanation:

- - calcareous bedrock;
- - non-calcareous bedrock; and

horizontal bar represents the proximal and distal Amargosa Basin *per ascensum* deposits.

Note:

- a) the cobble encrustation oxygen -18 data are from Quade et al. (1988) and the Amargosa Basin data are from Hay et al. (1986);
- b) samples of both the local cobble encrustations and the Amargosa Basin distal *per ascensum* deposits yield similar values of the $\delta^{18}\text{O}$ ratio; and
- c) either the *per ascensum* process yields carbonates that, in terms of the oxygen -18 contents, are similar to those produced via the *per descensum* process or the assumption of the *per descensum* origin of the local cobble encrustations is FALSE.

Oxygen isotopic comparison, the Amargosa Basin *per ascensum* deposits and the local cobble encrustations.

Explanation:

- - sample represents the Trench 14 calcretes;
- - sample represents the Busted Butte calcretes;
- - sample represents the Trench 14 fault infilling;
- - sample represents the Busted Butte fault infilling; and
- + - sample represents Trench 14 and Busted Butte "mosaic" breccia.

The isotopic compositions vs. morphological form gradients, the Yucca Mountain calcite-silica deposits.

Note:

- a) the isotopic data are from Whelan and Stuckless (1990);
- b) the Yucca Mountain calcite-opaline silica deposits occur as three morphological varieties, namely: i) the "mosaic" breccia cement, ii) the fault infillings, and iii) the calcretes;
- c) the $\delta^{18}\text{O}$ vs $\delta^{13}\text{C}$ fields from the distinguished morphological forms are sufficiently similar to assume that all of these forms were precipitated in the parent fluids which, in the pristine state, have carried similar values of the $\delta^{13}\text{C}$ vs $\delta^{18}\text{O}$ ratio;
- d) it may reasonably be assumed that: i) the degree of departure from the pristine state, resulting from either the oxygen -18 evaporative enrichment or the carbon -13 diffusional enrichment, was the lowest for the parent fluids of the "mosaic" breccia cement, somewhat higher for the parent fluids of the fault infillings, and still higher for the parent fluids of the calcretes; and ii) the mineral precipitation temperatures were the highest for the "mosaic" breccia cement, somewhat lower for the fault infillings, and still lower for the calcretes;
- e) the observed isotopic compositions vs. morphological form gradients may reasonably be taken as indicating: i) for the oxygen -18 gradient, the combined influence of the mineral precipitation temperature and the oxygen-18 evaporative enrichment, and ii) for the carbon -13 gradient, the carbon -13 diffusional enrichment; and
- f) the observed isotopic compositions vs. morphological form gradients are consistent with the *per ascensum* origin of the Yucca Mountain calcite-silica deposits - similarly as is the case for the Amargosa Basin deposits, the Yucca Mountain distal calcretes are isotopically "heavier" than their proximal counterparts.

The isotopic compositions vs. morphological form gradients, the Yucca Mountain calcite-silica deposits. (continued)

Note:

- a) the normalized value of the $\delta^{18}\text{O}$ ratio, with reference to the Amargosa Basin deposits, was computed by subtracting the normalization factor from the observed Yucca Mountain values [the total normalization factor = the distal component plus the proximal component];
- b) relative to the Amargosa Basin, the mean altitude of the Yucca Mountain topographic surface is ~ 600 m higher;
- c) based on the attached Figure 26, the expected value of the $d\delta^{18}\text{O}/dz$ gradient, for the *per ascensum* cobble encrustations and distal calcretes precipitating at the local ambient temperatures, is ~ 2 per mil _{SMOW} per 1km decrease in altitude z ;
- d) the normalization factor, for the Yucca Mountain calcretes and the isotopically "heaviest" fault infillings, consists of the distal component and is equal to 2 per mil _{SMOW} $\times$ 0.6 = -1.2 per mil _{SMOW};
- e) it may be assumed that, for the Amargosa Basin and the Yucca Mountain area, the local ambient temperatures are 20 and 17 Celsius, respectively;
- f) using the geothermal data from Garside and Schilling (1979), it may be assumed that the precipitation temperature, for the Amargosa Basin proximal deposits, is ~ 32°Celsius [i.e., ~ 12°Celsius above the local ambient temperature];
- g) for the purposes of this normalized comparison, it may be assumed that, for the Yucca Mountain "mosaic" breccia cement, the precipitation temperatures were ~ 6°Celsius above the local ambient temperature;
- h) the proximal component of the normalized factor, for the Yucca Mountain "mosaic" breccia cement and for the isotopically "lightest" fault infillings, is -1.2 per mil _{SMOW} - it has been computed for $T = 23^\circ\text{Celsius}$, using the equilibrium fractionation equation $10^3 \ln \alpha_{\text{CaCO}_3-\text{H}_2\text{O}} = 2.78 \cdot 10^6 \cdot T^{-2} - 2.89$, from Turi (1986); and
- i) the total normalization factor, for the Yucca Mountain "mosaic" breccia cement and for the isotopically "lightest" fault infillings, is -2.4 per mil _{SMOW}.

Computation of the oxygen -18 normalization factors for the Yucca Mountain calcite-silica deposits.

Note:

- for the Yucca Mountain "distal" deposits, the normalization factor is $-1.2 \text{ per mil}_{\text{SMOW}}$;
- for the Yucca Mountain "proximal" deposits, the normalization factor is $-2.4 \text{ per mil}_{\text{SMOW}}$; and
- the Amargosa Basin isotopic data are from Hay et al. (1986), and the Yucca Mountain data are from Whelan and Stuckless (1990).

The normalized oxygen-18 comparison, the Amargosa Basin *per ascensum* deposits and the Yucca Mountain calcite-silica deposits.

Oxygen isotopic file - isotopic character of oxygen contained in carbonate gangue veins from the Carlin and Cortez gold deposits. From Rye, 1985.

Results of isotopic comparison: the observed Yucca Mountain range, from 19.2 to 22.0 per mil^{SMOW} is similar to some of the Carlin and Cortez carbonate gangue veins.

- a) calcite veins from the oxidized zone at Carlin, and all the veins at Cortez, have oxygen - isotopic compositions similar to that of the calcite component in the altered host rock; and
- b) other veins were derived from low-temperature groundwater.

Note:

- tie-lines connect coexisting calcite and dolomite
- ▲ - sample represents altered host rock and calcite veins in oxidized and unoxidized zones;
 - - sample represents calcite veins;
 - x - sample represents dolomite; and

Explanation:

a) the Carlin disseminated gold deposit of Nevada.

b) the Cortez disseminated gold deposit of Nevada.

Explanation:

- - sample represents the carbonate gangue minerals from the Carlin and Cortez gold deposits; Tpa, Tld, and Tpl denote lithofacies from the Amargosa Basin deposits;
- - sample represents the cobble encrustations, collected at altitudes less than 1550m and from the carbonate bedrock sites; and
- ⊙ - sample represents the cobble encrustations, collected at altitudes more than 1550m and from the Spring Mountain carbonate bedrock sites.

Isotopic comparative analyses - the Carlin and Cortez carbonate gangue minerals, the Amargosa Basin *per ascensum* deposits, and the reference cobble encrustations.

Note:

- a) the isotopic data, as shown, are from: Rye (1985), Hay et al. (1986), and Quade et al. (1988);
- b) the observed trend toward the increasing carbon -13 and oxygen -18 contents, for all of the considered lithofacies but excluding the cobble encrustations from the high altitude Spring Mountain area, may be accounted for by assuming that: i) all of the lithofacies were precipitated from fluids that, in the pristine state, have carried similar carbon - oxygen isotopic signatures; ii) the precipitation temperatures and the degree of evaporation are, respectively, the highest and the lowest for the vein - proximal lithofacies; iii) values of CO₂ partial pressure are the highest for the vein - proximal lithofacies; and iv) the progressive carbon -13 diffusional enrichment, the precipitation temperature controlled fractionation factors, and the progressive oxygen -18 evaporative enrichment, are responsible for the observed isotopic gradients;
- c) the observed two-fold clustering of the isotopic data, from the reference cobble encrustations, may be taken to indicate that two genetically different populations are involved;
- d) the cobble encrustations encountered in the low altitude Tytus Canyon area represent the *sensu stricto per ascensum* population - it was suggested, in the January 18 letter, that the cobble encrustations representing this population were precipitated from the highly evolved *per ascensum* fluids, discharged from nearby no longer active springs; and
- e) the cobble encrustations encountered in the high altitude Spring Mountain area represent the *sensu lato per descensum* population - it was suggested, in the January 18 letter, that the cobble encrustations representing this population were precipitated from seasonal run-off fluids, evaporating through the topographic surface. [Longwell et al. (1965), while discussing "gravel firmly cemented with calcium carbonate" from the Kyle and Lee Canyon areas, have used unmistakably clear language "the carbonate cement no doubt was dissolved either from the fragments or from the parent bedrock in the range." For both the Kyle Canyon cobble encrustations and the Lee Canyon cobble encrustations there is no rational reason to: i) call for the dubious *sensu stricto per descensum* concepts; and ii) insist for the solely atmospheric - biospheric source of carbon. Furthermore, to account for the observation made by Quade et al. (1988) that, relative to the non-carbonate bedrock sites the cobble encrustations from the carbonate bedrock sites contain isotopically "heavier" carbon, there is again no rational reason to propose that differing ecosystems are residing over the respective sites.]

Isotopic comparative analyses - the Carlin and Cortez carbonate gangue minerals, the Amargosa Basin *per ascensum* deposits, and the reference cobble encrustations. (continued)

Explanation:

- - sample represents the carbonate gangue minerals from the Carlin and Cortez gold deposits - value shown has been adjusted by -5 per mil PDB to account for the non-carbonate bedrock;
- - sample represents the cobble encrustations, collected at altitudes less than 1500m and from the non-carbonate bedrock sites;
- - sample represents the cobble encrustations, collected at altitudes more than 1500m and from the non-carbonate bedrock sites; and
- ⊙ - sample represents the calcite-silica veins from the Yucca Mountain vadose zone (depth ranging from 34 to 611m).

Isotopic comparative analyses - the normalized Carlin and Cortez carbonate gangue minerals, the Yucca Mountain surficial and subsurface calcite-opaline silica deposits and the reference cobble encrustations.

Note:

- a) the isotopic data, as shown, are from: Rye (1985), Szabo and Kyser (1985), Quade et al. (1988), and Whelan and Stuckless (1990);
- b) the isotopic compositions of the Carlin and Cortez carbonate gangue veins, as shown, have been normalized to account for the respective differences in lithology of the host rock - the assumed normalization factor is -5 per mil_{PDB}; and
- c) the observed trend toward the increasing carbon -13 and oxygen -18 contents, for all of the considered lithofacies, may be accounted for by assuming that: i) all of the lithofacies were precipitated from fluids that, in the pristine state, have carried similar carbon - oxygen isotopic signatures; ii) the precipitation temperatures and the degree of evaporation are, respectively, the highest and the lowest for the breccia cement - vein lithofacies; iii) values of the CO₂ partial pressure are the highest for the vein-breccia cement lithofacies; and iv) the progressive diffusional carbon -13 enrichment, the precipitation temperature controlled fractionation factors, and the progressive evaporative oxygen -18 enrichment, are responsible for the observed isotopic gradients.

Isotopic comparative analyses - the normalized Carlin and Cortez carbonate gangue minerals, the Yucca Mountain surficial and subsurface calcite-opaline silica deposits and the reference cobble encrustations. (continued)

Note:

- a) the host rock for the hydraulic fracture calcite residuum is the bedded tuff from the "caprock zone" of the Topopah Spring Member;
- b) presence of both the locally very intense oxidation and the jasperoidal opals indicates that the observed bedded tuff alteration is caused by hydrothermal solutions; and
- c) two different views of the same calcite residuum are shown.

Hydraulic fracture calcite residuum - eastern flank of the Harper Valley, southern part of the Yucca Mountain area.

- o Is the interpreted carbon isotopic composition of the parent fluids, for the Yucca Mountain calcretes and veins, compatible with the corresponding composition of the *per descensum* pedogenic fluids, as observed and interpreted for the Yucca Mountain area?

Statement of the issue - topic #1

Note:

- the observed range for the $\delta^{13}\text{C}$ ratio, from samples of the Yucca Mountain calcretes and surficial veins, is from -3.0 to -7.5 per mil_{PDB}, isotopic data are from Whelan and Stuckless, 1990;
- for precipitation temperature $T = 15^\circ\text{Celsius}$, the equilibrium fractionation factor $10^3 \ln \alpha_{\text{CaCO}_3 - \text{HCO}_3^-} = 1.75$ per mil_{PDB}; and
- the interpreted range for the $\delta^{13}\text{C}$ ratio, for the parent fluids of the Yucca Mountain calcretes and surficial veins, is from -4.75 to -9.25 per mil_{PDB}.

The interpreted range of values for the $\delta^{13}\text{C}$ ratio - the parent fluids for the Yucca Mountain calcretes and surficial veins.

- o A mean value of the $\delta^{13}\text{C}$ ratio for the CO_2 gas respired from the vadose zone, as observed at Yucca Mountain and Amargosa Narrows, is $\delta^{13}\text{C} = -18.36$ and -20.0 per mil_{PDB}, respectively, Yang et al., 1986, Thorstenson et al., 1989, and White and Chuma, 1987 - the Yucca Mountain value is based on samples that carry PMC > 100 percent and were collected at a depth ranging from 0 to 12.8m.
- o A mean value of the $\delta^{13}\text{C}$ ratio for soil gas, as observed at various locations throughout the Nevada Test Site, is $\delta^{13}\text{C} = -20.5$ per mil_{PDB}, Boughton, 1986.
- o The equilibrium isotopic fractionation factor $10^3 \ln \alpha_{\text{HCO}_3^- - \text{CO}_2(\text{g})} = 9.483 \cdot 10^3 \cdot T^{-1} - 23.89$, Turi (1986) - for 15°Celsius, the fractionation factor is ~ 9 per mil_{PDB}.
- o Dissolution of the CO_2 gas respired from the vadose zone, with the observed range from -18.36 to -20.5 per mil_{PDB}, may be expected to yield the *per descensum* pedogenic fluids with the $\delta^{13}\text{C}$ ratios ranging from -9.36 to -11.5 per mil_{PDB}.

Isotopic character of carbon dissolved in the *per descensum* pedogenic fluids, interpreted based on the observed $\delta^{13}\text{C}$ ratios from samples of the Nevada Test Site vadose zone gases.

- o A mean value for the $\delta^{13}\text{C}$ ratio from samples of shallow alluvium - based fluids, as observed around Amargosa Narrows, is $\delta^{13}\text{C} = -11.3$ per mil_{PDB} (range of the observed values is from -9.59 to -13.05 per mil_{PDB}), White and Chuma (1987).
- o A value of the $\delta^{13}\text{C}$ ratio from samples of shallow tuff "pile" - based fluids, as observed in the Yucca Mountain borehole UE-29 a#2 (depth to the water table is 29m, and PMC ranges from 60 to 62.3 percent), is $\delta^{13}\text{C} = -13.0$ per mil_{PDB}, Benson and McKinley, 1985.
- o A value of the $\delta^{13}\text{C}$ ratio for the Nevada Test Site soil water, as interpreted based on laboratory soil-leaching experiments, is $\delta^{13}\text{C} = -12.0$ per mil_{PDB} (the observed range for leached water is from -9.6 to -16.4 per mil_{PDB}), Spencer (1990).

Isotopic character of carbon dissolved in the *per descensum* pedogenic fluids, based on direct measurements and on soil-leaching experiments.

Summary - comparison of carbon isotopic compositions, the *per descensum* pedogenic fluids and the parent fluids for the Yucca Mountain calcretes and surficial veins.

The $\delta^{13}\text{C}$ range from -4.75 to -9.45 per mil_{PDB}, interpreted for the parent fluids of the Yucca Mountain calcretes and surficial veins, is too "heavy" to permit genetic association with the local *per descensum* pedogenic fluids alone.

Note:

- a) Quade and Cerling (1990) have proposed that, the Yucca Mountain calcite-silica deposits were precipitated during the glacial maxima, when the local climate was cooler than the contemporary climate;
- b) the postulated climate was analogous to that presently encountered at altitudes 750 - 790m higher, in the lower pinyon-juniper-sagebrush zone;
- c) it may reasonably be assumed that, during precipitation of the Yucca Mountain calcite-silica deposits, the local biomass could have contained higher proportion of plants with the C-3 metabolic pathway and, consequently, the biogenically derived CO_2 was isotopically "lighter" than the contemporary CO_2 ; and
- d) the depicted in Figure 18 carbon isotopic discrepancy, between the parent fluids for the Yucca Mountain deposits and the local *per descensum* pedogenic fluids, represents a minimum value.

Conclusion - evaluations of the carbon isotopic compatibility, between the parent fluids for the Yucca Mountain calcretes and surficial veins and the local *per descensum* pedogenic fluids.

What is the diagnostic usefulness of the isotopic compositions vs altitude gradients, as observed by Quade and Cerling (1990) for the local cobble encrustations?

Statement of the issue - topic #2.

Sample No.	Elevation (m)	$\delta^{13}\text{C}_{\text{PDB}}$	$\delta^{18}\text{O}_{\text{PDB}}$
SM-1a	840	-2.0	-9.0
SM-1b	840	-1.9	-7.8
SM-1c	840	-1.2	-7.8
SM-2b	1550	-3.1	-8.8
SM-2b	1550	-4.0	-11.2
SM-2c	1550	-5.2	-12.3
SM-3a	1950	-5.4	-11.2
SM-3b	1950	-5.0	-11.6
SM-3c	1950	-5.8	-10.4
SM-3(B)a	1900	-4.3	-11.0
SM-3(B)b	1900	-3.6	-9.4
SM-3(B)c	1900	-4.8	-9.2
SM-4a	2270	-8.1	-13.1
SM-4b	2270	-6.0	-8.2
SM-4c	2270	-5.8	-9.2
SM-4d	2270	-7.3	-11.4
SM-5a	2490	-8.62	-12.2
SM-5b	2490	-8.4	-12.2
SM-5b-rerun	2490	-8.4	-12.5
SM-5c	2490	-8.8	-11.5
SM-6a	2740	-9.11	-12.2
SM-6b-1	2740	-8.08	-11.8
SM-6b-2	2740	-8.7	-12.6
SM-6c	2740	-8.0	-11.7
TC-1a	300	3.4	-2.8
TC-1b	300	4.1	-2.6
TC-1c	300	2.5	-5.6
GM-1a	1830	-8.3	-11.0
GM-1a -1m	1830	-6.9	-11.6
GM-1c	1830	-6.5	-10.4
GM-1d	1830	-7.2	-11.0
GM-2a-90cm	1575	-6.3	-9.4
GM-2b	1575	-5.8	-8.8
GM-2c	1575	-5.5	-9.7
GM-3a	1160	-2.0	-6.7
GM-3b	1160	-0.9	-7.1
GM-3c	1160	-1.8	-7.0
GM-4a	900	-2.4	-6.8
GM-4b	900	-1.3	-7.5
PaM-1a	300	-1.7	-0.4
PaM-1b	300	-0.4	-3.5
PaM-1c	300	0.9	-6.2
PiVM-1a	1675	-6.1	-11.1
PiVM-1d	1675	-6.1	-11.9
PiVM-2a	2130	-8.1	-11.2
PiVM-2b	2130	-7.5	-10.0
PiVM-2c	2130	-7.7	-11.2

Carbon and oxygen isotopic data. Cobble encrustations - region around the Nevada Test Site. From Quade et al., 1988.

Note:

- carbon contained in the cobble encrustations developed over the calcareous bedrock is about 2 per mil_{PDB} "heavier";
- this observation may be taken to indicate that, partially, parent fluids for the cobble encrustations acquire their carbon-13 content from the underlying bedrock;
- the $d\delta^{13}\text{C}/dz$ gradient has been computed assuming that, for a fixed altitude and character of the underlying bedrock, the parent fluids have carried values of the $\delta^{13}\text{C}$ ratio varying by $\Delta\delta^{13}\text{C} \sim 3$ per mil_{PDB}; and
- the above assumption is reasonable, for either the *per descensum* pedogenic fluids or the *per ascensum* pedogenic fluids, and is justified by the results of actual measurements.

Isotopic character of carbon contained in the cobble encrustations as a function of altitude of land surface.

Explanation:

● - calcareous bedrock; and

○ - non-calcareous bedrock.

Note:

- $\delta^{18}\text{O}_{\text{SMOW}} = 1.03086 \cdot \delta^{18}\text{O}_{\text{PDB}} + 30.86$, Fritz and Fontes (1980);
- the $d\delta^{18}\text{O}/dz$ gradient has been computed assuming that, for a fixed altitude, the parent fluids have carried values of the $\delta^{18}\text{O}$ ratio varying by $\Delta\delta^{18}\text{O} \sim 4$ per mil_{SMOW}; and
- the above assumption is reasonable, for either the *per descensum* pedogenic fluids or the *per ascensum* pedogenic fluids, and is justified by the results of actual measurements.

Isotopic character of oxygen contained in the cobble encrustations as a function of altitude of land surface.

Explanation:

- - groundwater data (well and spring), from Hershey, 1989; and
 - - the Nevada Test Site groundwater data (well and springs), from Isherwood et al., 1982, Claassen, 1985, Kerrisk, 1987, White and Chuma, 1978, Spencer, 1990, Stuckless, 1990, and Lyles et al., 1990.
- tiefines represent the Whiterock Spring fluids and the Cane Spring fluids.

Isotopic character of carbon dissolved in the Nevada Test Site subsurface fluids, as a function of altitude of land surface, and the expected $\delta^{13}\text{C}$ vs altitude gradient and field for the *per ascensum* cobble encrustations and distal calcretes.

Note:

- a) for comparison with the local cobble encrustations, the $\delta^{13}\text{C}$ vs altitude field, shown between solid and dashed lines, should be shifted by the equilibrium fractionation factor $10^3 \ln \alpha_{\text{CaCO}_3 - \text{HCO}_3^-} \sim 2$ per mil_{PDB};
- b) relative to the observed $\delta^{13}\text{C}$ vs altitude field, from the local cobble encrustations, the expected $\delta^{13}\text{C}$ vs altitude field, for the *per ascensum* cobble encrustations and distal calcretes, is about a factor of two wider;
- c) the increased width of the expected $\delta^{13}\text{C}$ vs altitude field may be accounted for by recognizing that fluids which carry isotopically "light" carbon (situated between two dashed lines) are the calcium-poor fluids residing in the tuff "pile" based hydraulic "sink" regions - such fluids are not involved in precipitation of the local cobble encrustations; and
- d) the expected value for the $\delta^{13}\text{C}$ vs altitude gradient, from samples of the *per ascensum* cobble encrustations and distal calcretes, is $d\delta^{13}\text{C}/dz \sim 4$ per mil_{PDB} per 1 km decrease in altitude of carbonate precipitation site.

Isotopic character of carbon dissolved in the Nevada Test Site subsurface fluids, as a function of altitude of land surface, and the expected $\delta^{13}\text{C}$ vs altitude gradient and field for the *per ascensum* cobble encrustations and distal calcretes.

Explanation

- - groundwater data (well and spring) from the Spring Mountain area above altitude of 1600m, here, the host rock consists presumably exclusively of the Paleozoic carbonates, from Hershey, 1989; and
- - groundwater data from the Nevada Test Site hydraulic "source" regions - here, the resident fluids are supersaturated with respect to calcite and, potentially, may discharge at the topographic surface.

Note:

- a) selective usage of the carbon isotopic data is an attempt to exclude the calcium-poor fluids residing in the tuff "pile" based hydraulic "sink" regions; and
- b) both the expected $\delta^{13}\text{C}$ vs altitude gradient and the expected $\delta^{13}\text{C}$ vs altitude field, for the *per ascensum* cobble encrustations and distal calcretes, are similar to the corresponding gradient and field, as observed by Quade et al. (1988) for the local cobble encrustations.

Isotopic character of carbon dissolved in the Nevada Test Site calcium-rich subsurface fluids ("source" plus carbonate rock based fluids) as a function of altitude of land surface and the expected values of the $\delta^{18}\text{O}$ gradient and field for the *per ascensum* cobble encrustations and distal calcretes.

Explanation:

- - groundwater data (spring and well) from the Spring Mountain area above altitude of 1600m, from Hershey, 1989;
- ◐ - groundwater data (possibly "oxygen shifted") from the Spring Mountain area, from Hershey, 1989; and
- - groundwater data from the Nevada Test Site hydraulic "source" regions, from Isherwood et al., 1982, Claassen, 1965, Kerrisk, 1987, White and Chuma, 1987, Benson and McKinley, 1985, Stuckless et al., 1990, and Lyles et al., 1990.

Isotopic character of oxygen contained in the Nevada Test Site calcium-rich subsurface fluids ("source" plus carbonate rock based fluids), as a function of altitude of land surface, and the expected $\delta^{18}\text{O}$ vs altitude gradient and field for the *per ascensum* cobble encrustations and distal calcretes.

Note:

- a) for comparison with the local cobble encrustation, the $\delta^{18}\text{O}$ vs altitude field, shown between two solid lines, should be shifted by the equilibrium fractionation factor $10^3 \ln \alpha_{\text{CaCO}_3 - \text{H}_2\text{O}} \sim 32$ per mil_{SMOW};
- b) the expected value for the isotopic composition vs altitude gradient, from samples of the *per ascensum* cobble encrustations and distal calcretes, has been computed by summing up the values of three independent isotopic gradients;
- c) these gradients results from the altitude dependence of: i) the observed isotopic composition of the local subsurface fluids - $d\delta^{18}\text{O}/dz = + 0.7$ per mil_{SMOW} per 1km decrease in altitude of groundwater sampling site; ii) the observed mean ambient temperature of the topographic surface - $d\delta^{18}\text{O}/dz = -1.2$ per mil_{SMOW} per 1km decrease in altitude of carbonate precipitation site; iii) the interpreted oxygen -18 evaporative enrichment - $d\delta^{18}\text{O}/dz = + 2.5$ per mil_{SMOW} per 1km decrease in altitude of carbonate precipitation site;
- d) the value of $d\delta^{18}\text{O}/dz = -1.2$ per mil_{SMOW} per 1km, reflecting the altitude dependence of the mean ambient temperature of the topographic surface, has been computed using: i) the equilibrium fractionation equation $10^3 \ln \alpha_{\text{CaCO}_3 - \text{H}_2\text{O}} = 2.78 \cdot 10^6 \cdot T^{-2} - 2.89$, from Turi (1986); and ii) $dT/dz = 5^\circ\text{Celsius}$ per 1km decrease in altitude of the topographic surface - Major (1977) gives the local $dT/dz = 5.4^\circ\text{Celsius}$ per 1km;
- e) the value of $d\delta^{18}\text{O}/dz = + 0.7$ per mil_{SMOW} per 1km, reflecting the altitude dependence of the oxygen -18 content of the calcium-rich subsurface fluids, is given by two dashed lines;
- f) the value of $d\delta^{18}\text{O}/dz = +2.5$ per mil_{SMOW} per 1km, reflecting the altitude dependence of the oxygen -18 evaporative enrichment, is computed in Figure 26a; and
- g) both the expected $\delta^{18}\text{O}$ vs altitude gradient and the expected $\delta^{18}\text{O}$ vs altitude field; for the *per ascensum* cobble encrustations and distal calcretes, are similar to the corresponding gradient and field, as observed by Quade et al. (1988) for the local cobble encrustations.

Isotopic character of oxygen contained in the Nevada Test Site calcium-rich subsurface fluids ("source" plus carbonate rock based fluids), as a function of altitude of land surface, and the expected $\delta^{18}\text{O}$ vs altitude gradient and field for the *per ascensum* cobble encrustations and distal calcretes. (continued)

a) mean value of the isotopic composition vs altitude gradient - cobble encrustations from the Spring Mountain area, above altitude of 1800m.

b) the isotopic composition vs altitude gradient - subsurface fluids from the Spring Mountain area, above altitude of 1800m.

Note:

- isotopic data, as shown, are from Hershey (1989) and are computed from Quade et al. (1988);
- the observed isotopic composition vs altitude gradient, from samples of the *sensu lato per descensum* cobble encrustations from the Spring Mountain area, is the combined results of two altitude dependent factors, namely; i) the oxygen -18 evaporative enrichment; and ii) the oxygen -18 content of the local subsurface fluids; and
- the interpreted value for the oxygen -18 evaporative enrichment vs altitude gradient, from samples of the groundwater cobble encrustations, is $d\delta^{18}\text{O}/dz = +2.5 \text{ per mil}_{\text{SMOW}} \text{ per 1 km decrease in altitude of carbonate precipitation site.}$

Isotopic character of oxygen contained in the Nevada Test Site calcium-rich fluids ("source" plus carbonate rock based fluids), as a function of altitude of land surface, and the expected $\delta^{18}\text{O}$ vs altitude gradient and field for the *per ascensum* cobble encrustations and distal calcretes.

- o Values of the isotopic compositions vs altitude gradients, observed based on samples of the local cobble encrustations, are:
 - i) $d\delta^{13}\text{C}/dz \sim +4 \text{ per mil}_{\text{PDB}}$ per 1km decrease in altitude of the deposit sampling site; and
 - ii) $d\delta^{18}\text{O}/dz \sim +2 \text{ per mil}_{\text{SMOW}}$ per 1km decrease in altitude of the deposit sampling site.

- o For the *per ascensum* cobble encrustations and distal calcretes, the expected value of the carbon -13 content vs altitude gradient is $d\delta^{13}\text{C}/dz \sim +4 \text{ per mil}_{\text{PDB}}$ per 1km decrease in altitude of the deposit precipitation site - this expectation is based on the carbon-13 content vs altitude gradient, as observed from samples of the calcium-rich subsurface fluids.

- o For the *per ascensum* cobble encrustations and distal calcretes, the expected value of the oxygen -18 content vs altitude gradient is $d\delta^{18}\text{O}/dz \sim +2 \text{ per mil}_{\text{SMOW}}$ per 1km decrease in altitude of the deposit precipitation site - this expectation is based on:
 - i) the oxygen-18 content vs altitude gradient, as observed from samples of the calcium-rich subsurface fluids;
 - ii) the observed mean ambient temperature vs altitude gradient; and
 - iii) the interpreted oxygen -18 evaporative enrichment vs altitude gradient.

- o In addition to yielding the comparable values of the isotopic gradients, the calcium-rich subsurface fluids also carry values of the $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ ratio that are comparable to those interpreted for the parent fluids of both the Yucca Mountain calcretes and veins and the local reference cobble encrustations.

Summary - comparative analyses of the isotopic compositions vs altitude gradients, the observed cobble encrustations and the expected *per ascensum* cobble encrustations and distal calcretes.

- o Evidently, the observation made by Quade and Cerling (1990) that the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ ratios from samples of the local cobble encrustations change progressively with changing altitude of the sampling site, if considered together with the similar observation made based on the expected ratios for the *per ascensum* cobble encrustations and distal calcretes, has a two-fold meaning:
 - i) either the *per ascensum* process yields cobble encrustations and distal calcretes that, in terms of the isotopic compositions vs altitude gradients, are similar to those produced via the *per descendsum* process; or
 - ii) the assumption of the *per descendsum* origin for the local cobble encrustations, as proposed by Quade et al. (1988), Quade et al. (1989), and Quade and Cerling (1990), is FALSE.
- o The above conclusion, if considered together with the conclusion from Figure 19, indicates that the *per descendsum* interpretations of the origin of the Yucca Mountain calcretes and veins, as proposed by Quade and Cerling (1990), are devoid of any scientific bases.

Overall conclusions - comparative analyses of the isotopic compositions vs altitude gradients, the observed cobble encrustations and the expected *per ascensum* cobble encrustations and distal calcretes.

Is the *per ascensum* origin of the Yucca Mountain calcite-silica deposits supported by the isotopic characters of uranium, strontium, and carbon contained in these deposits?

Statement of the issue - topic #3.

Inferred boundary conditions. The Nevada Test Site hydrosphere.

Pahute Mesa Hydraulic "Source" Region

Vertical hydraulic gradient dh/dz - upward (maximum known value is $\Delta h = 174$ ft.).

Vertical geothermal gradient dT/dz ranging from 30 to about $60^\circ\text{C}/\text{km}$.

Lateral geothermal gradient dT/dx $5^\circ\text{C}/1$ mile.

Possible upper thermal boundary layers.

Water chemistry: $\text{Ca}^{++} + \text{Mg}^{++}$ from 1 to 40 ppm
 $\text{Na}^+ + \text{K}^+$ from 40 to 250 ppm
 $\text{Cl}^- + \text{SO}_4^{--}$ from 35 to 250 ppm
 HCO_3^- from 90 to 270 ppm.

Pahute Mesa Hydraulic "Sink" Region

Vertical hydraulic gradient dh/dz - downward (maximum known value is $\Delta h = 41$ ft.).

Vertical geothermal gradient $dT/dz \sim 30^\circ\text{C}/\text{km}$.

Water chemistry: $\text{Ca}^{++} + \text{Mg}^{++}$ from 4 to 60 ppm
 $\text{Na}^+ + \text{K}^+$ from 35 to 95 ppm
 $\text{Cl}^- + \text{SO}_4^{--}$ from 15 to 70 ppm
 HCO_3^- from 60 to 450 ppm.

Boundary conditions. Interpretation bases. The Nevada Test Site hydrosphere.

Rainier Mesa - Yucca Flat Hydraulic "Sink" Region.

Vertical hydraulic gradient dh/dz - downward (maximum known value is $\Delta h = 1560$ ft.).

Vertical geothermal gradient $dT/dz \simeq 20$ - 25°C .

Water chemistry: $\text{Ca}^{++} + \text{Mg}^{++}$ from 2 to 40 ppm
 $\text{Na}^+ + \text{K}^+$ from 22 to 65 ppm
 $\text{CL}^- + \text{SO}_4^{--}$ from 11 to 40 ppm
 HCO_3^- from 50 to 230 ppm.

Note: $\sigma_c \geq \frac{1}{3} \sigma_v$ indicating that dilation is either very shallow or non-existent.

Halfpint Hydraulic "Source" Region.

Vertical hydraulic gradient dh/dz - unknown.

Vertical geothermal gradient $dT/dz \simeq 45^\circ\text{C/km}$.

Water chemistry: $\text{Ca}^{++} + \text{Mg}^{++}$ from 55 to 400 ppm
 $\text{Na}^+ + \text{K}^+$ from 50 to 270 ppm
 $\text{CL}^- + \text{SO}_4^{--}$ from 85 to 1100 ppm
 HCO_3^- from 65 to 400 ppm.

Note: a) local water table is bent upward, maximum altitude is 4535 ft. amsl. (148 ft. above water table in upstream regions); and
 b) lateral hydraulic gradient 1300 ft/mile and 820 ft/mile.

Boundary conditions. Interpretation bases. The Nevada Test Site hydrosphere.

Skull Mountain Hydraulic "Source" Region.

Vertical hydraulic gradient dh/dz - uncertain (upward gradient is suggested by hydraulic head observations made during drilling of Well #73-68).

Vertical geothermal gradient $dT/dz = 45\text{-}50^\circ\text{C/km.}$)

Water chemistry: $\text{Ca}^{++} + \text{Mg}^{++}$ from 4 to 470 ppm
 $\text{Na}^+ + \text{K}^+$ from 47 to 450 ppm
 $\text{CL}^- + \text{SO}_4^{--}$ from 35 to 500 ppm
 HCO_3^- from 100 to 1000 ppm.

- Note: a) water table is bent upward, maximum altitude is 2982 ft. amsl. (about 600 ft. above water table in upstream regions);
 b) samples of perched water display chemical affinity with water from below the water table; and
 c) possible upper thermal boundary layer present in two borcholes.

Indian Springs Valley Hydraulic "Sink" Region.

Vertical hydraulic gradient dh/dz - unknown.

Vertical geothermal gradient $dT/dz < 20^\circ\text{C/km.}$

Water chemistry: $\text{Ca}^{++} + \text{Mg}^{++}$ from 16 to 75 ppm
 $\text{Na}^+ + \text{K}^+$ from 6 to 40 ppm
 $\text{CL}^- + \text{SO}_4^{--}$ from 8 to 70 ppm
 HCO_3^- from 180 to 300 ppm.

- Note: a) local water table is bent downward, minimum altitude is 2281 ft. amsl. (80 feet lower than water table downstream).

Boundary conditions. Interpretation bases. The Nevada Test Site hydrosphere.

Ash Meadows Hydraulic "Source" Region.

Vertical hydraulic gradient dh/dz - upward (maximum value of Δh - unknown).

Vertical geothermal gradient $dT/dz \sim 40^\circ\text{C}/\text{km}$.

Water chemistry: $\text{Ca}^{++} + \text{Mg}^{++}$ from 12 to 270 ppm
 $\text{Na}^+ + \text{K}^+$ from 20 to 300 ppm
 $\text{CL}^- + \text{SO}_4^{--}$ from 45 to 1300 ppm
 HCO_3^- from 120 to 380 ppm.

- Note: a) water table appears to be bent upward; downstream water table altitude 2200 ft. amsl., maximum altitude 2365 ft., upstream at altitude 2281 ft; and
b) fault-line spring discharge, temperature ranging from 23 to 34.5°C.

Bullfrog Hydraulic "Source" Region.

Vertical hydraulic gradient dh/dz - upward (maximum value of Δh - unknown).

Vertical geothermal gradient dT/dz - unknown.

Water chemistry: $\text{Ca}^{++} + \text{Mg}^{++}$ from 8 to 50 ppm
 $\text{Na}^+ + \text{K}^+$ from 100 to 350 ppm
 $\text{CL}^- + \text{SO}_4^{--}$ from 90 to 350 ppm
 HCO_3^- from 150 to 480 ppm.

- Note: a) perched waters - discharge temperature 21-26.5°C and chemical affinity with deeper fluids suggest that these waters represent mixtures of deep fluids with infiltrating meteoric fluids; and
b) water table may be bent upward; downstream - 2500 ft., maximum $\sim$ 4000 ft., upstream - 3800 ft.

Boundary conditions. Interpretation bases. The Nevada Test Site hydrosphere.

Yucca Mountain "Dilated" Region.

Vertical hydraulic gradient dh/dz - upward in deeper undilated regions; neutral immediately below the water table.

Vertical geothermal gradient $dT/dz = 40-60^\circ\text{C}/\text{km}$ in deeper undilated regions; $dT/dz = 20-30^\circ\text{C}/\text{km}$ immediately below the water table.

Water chemistry:	dilated zone	$\text{Ca}^{++} + \text{Mg}^{++}$ from 2 to 20 ppm $\text{Na}^+ + \text{K}^+$ from 40 to 120 ppm $\text{CL}^- + \text{SO}_4^-$ from 21 to 55 ppm HCO_3^- from 100 to 300 ppm.
	undilated zone	$\text{Ca}^{++} + \text{Mg}^{++}$ 140 ppm $\text{Na}^+ + \text{K}^+$ 162 ppm $\text{CL}^- + \text{SO}_4^-$ 188 ppm HCO_3^- 569 ppm.

Note: a) local values of $\sigma_e \rightarrow 0$, (indicating severe dilation) extend to a maximum depth of about ~ 1300 m; and
b) the Yucca Mountain area is a source - sink hybrid.

The geothermal, hydraulic, and geochemical data used in subdividing the Nevada Test Site hydrosphere, are from Walker (1962); Blankennagel and Weir (1973); Claassen (1973); Winograd and Thordenson (1975); White (1972); White et al. (1980); Garside and Schilling (1979); Sass and Lachenbruch (1982); Isherwood et al. (1982); Claassen (1985); and Benson and McKinley (1985).]

Boundary conditions. Interpretation bases. The Nevada Test Site hydrosphere.

Downhole temperature profiles. Southern Indian Springs Valley "sink" region. From Sass and Lachenbruch, 1982.

Downhole temperature profiles. Rainier Mesa - Yucca Flat hydraulic "sink" regions. From Sass and Lachenbruch, 1982.

Downhole temperature profile. Borehole UE-25a3. North-western margin of the Skull Mountain hydraulic "source" region. From Sass and Lachenbruch, 1982.

Downhole temperature profiles. Boreholes J-11, TWF, and TW3. Skull Mountain hydraulic "source" region. From Sass and Lachenbruch, 1982.

Downhole temperature profiles. Pahute Mesa hydraulic "sink" and "source" regions. Modified from Sass and Lachenbruch, 1982.

Groundwater chemistry. Hydraulic "sink" and dilated regions. The Nevada Test Site hydrosphere.

Note:

- a) fluids result from intermixing of at least two end-members;
- b) one end-member is represented by poorly evolved meteoric recharge; and
- c) the other member is represented by deep and chemically evolved fluids.

Groundwater chemistry. Hydraulic "source" regions. The Nevada Test Site hydrosphere.

Note:

- $\log(Q/K)$ data are from Kerrisk (1987);
- $N=52$ for hydraulic "source" regions; $N=64$ for hydraulic "sink" regions; and
- Q - the ion activity product, K - the equilibrium constant for the solubility reaction.

Calcite saturation index. Water samples from the Nevada Test Site hydraulic "sink" and "source" regions.

Note:

- a) $\log (Q/K)$ data are from Kerrisk;
- b) $N = 48$ for hydraulic "source" regions, $N = 61$ for hydraulic "sink" regions; and
- c) Q - the ion activity product, K - the equilibrium constant for the solubility reaction.

Gypsum saturation index. Water samples from the Nevada Test Site hydraulic "sink" and "source" regions.

$^{234}\text{U}/^{238}\text{U}$ vs U-content field. Hydraulic "sink" and dilated regions. The Nevada Test Site hydrosphere.

Explanation:

- - sample represents fluids from the tuff pile;
- - sample represents fluids from the Paleozoic carbonates; and
- - sample represents fluids from alluvium.

Note:

- a) the Climax Stock perched fluids were excluded from this plot.

$^{234}\text{U}/^{238}\text{U}$ vs U-content field. Hydraulic "source" regions. The Nevada Test Site hydrosphere.

Note:

a) $n = 27$ for hydraulic "source" regions; $n = 17$ for hydraulic "sink" regions.

Isotopic character of the dissolved uranium. Water samples from the hydraulic "source" and "sink" regions. The Nevada Test Site hydrosphere.

Note:

- total number of samples $n = 15$ for the hydraulic "sink" regions, and $n = 19$ for the hydraulic "source" regions; and
- isotopic data are from Stuckless (1990).

Comparison of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. The Nevada Test Site "source" and "sink" regions.

$\delta^{13}\text{C}$ vs. PMC field. Water samples from the Nevada Test Site hydrosphere.

Note:

a) N = 16 for hydraulic "source" regions; N = 7 for hydraulic "sink" regions.

Isotopic character of the dissolved carbon. Water samples from the Paleozoic carbonates based hydraulic "sink" and "source" regions.

Isotopic character of the dissolved carbon. Water samples from the alluvium - tuff pile based hydraulic "source" and "sink" regions.

Note:

- samples are bulk samples pumped out of large segments of exploratory wells - sampling "smoothing" may be involved, and the observed oxygen isotopic shift may be smaller than the actually present one:
- oxygen isotopic shift $\Delta\delta^{18}\text{O} = (\delta D_{\text{ob}} - 10)/8 - \delta^{18}\text{O}_{\text{ob}}$; and
- oxygen isotopic data are from Isherwood et al. (1982), Benson and McKinley (1985), Claassen (1985), White and Chuma (1987), Stuckless (1990), and Stuckless et al. (1990).

Comparison of magnitudes of the oxygen isotopic shift. The Nevada Test Site "sink" and "source" regions.

Note:

- Spring Mountain fluids are contemporary fluids; and
- Yucca Mountain and Rainier Mesa fluids from below the water table are older fluids - ^{14}C ages are comparable to those of fluids from the "source" regions.

Relationship between the observed isotopic compositions, the meteoric water line, and altitude of the local land surface. Fluids from the hydraulic "sink" regions. The Nevada Test Site hydrosphere.

Relationship between the observed isotopic compositions, the meteoric water line, and altitudes of the local land surface. Fluids from the hydraulic "source" regions. The Nevada Test Site hydrosphere.

Note:

- comparison is based on data from regions with comparable altitudes of the land surface; and
- comparison is based on data from fluid samples with comparable PMC.

Comparison of isotopic compositions. "Sink" and "source" regions - The Nevada Test Site hydrosphere.

$^{234}\text{U}/^{238}\text{U}$ and $^{230}\text{Th}/^{234}\text{U}$ activity ratios. The Nevada Test Site calcite-silica deposits.

Total U-content vs. percentage of acid-soluble fraction. Calcite-silica deposits from the Nevada Test Site.

Comparison of the $^{234}\text{U}/^{238}\text{U}$ ratio vs U-content fields. The Nevada Test Site calcite-silica deposits and the Nevada Test Site subsurface fluids.

Note:

- total number of samples $n = 15$ for the hydraulic "sink" regions, and $n = 19$ for the hydraulic "source" regions; and
- isotopic data are from Marshall et al. (1990), Stuckless (1990), Peterman (1990), and Stuckless et al. (1990).

Comparison of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. The Yucca Mountain calcite-silica deposits and the Nevada Test Site subsurface fluids.

Note: Isotopic data from Claassen, (1985), Benson and McKinley (1985), and White and Chuma (1987).

Results of isotopic comparison: the observed Yucca Mountain range, from -3.0 to -7.5 per mil PDB , is similar to that expected for deposits produced by local geothermal fluids.

Comparison of the $\delta^{13}\text{C}$ ratio. The Yucca Mountain calcite-silica deposit and the Nevada Test Site subsurface fluids.

- o The uranium-strontium-carbon isotopic affinity, between the parent fluids for the Yucca Mountain calcite-silica deposits and the Nevada Test Site geothermal fluids, indicate that these deposits were formed via the *per ascensum* process.
- o The above conclusion is identical to those drawn, independently, based on: i) the results of direct field observations, as discussed in this letter; ii) the results of carbon-oxygen comparative analyses, as discussed in this letter and in the January 18 letter; and iii) the results of oxygen -18 paleo-geothermal gradient reconstructions, as discussed in the January 18 letter.
- o The above two conclusions, if considered together with conclusions from Figure 19 and 28, DEMONSTRATE that the January 18 statement: " the *per descensum* interpretations, as proposed by Quade and Cerling, may hardly be regarded as an example of the most insightful and meticulous science. To the contrary, within the context of safety considerations of a high-level nuclear waste repository, these interpretations may rightfully be regarded as a good example of irresponsible science", is fully justified.

Overall conclusions - evaluations of the uranium-strontium-carbon isotopic affinity, between the parent fluids for the Yucca Mountain calcite-silica deposits and the Nevada Test Site geothermal fluids.

What is the origin of carbon that is contained in the CO₂ gas respired from the Yucca Mountain vadose zone and is dissolved in the Nevada Test Site hydrosphere.

Statement of the issue - topic #4.

- o Quade and Cerling (1990) have performed their carbon isotopic analyses of the cobble encrustations, and of the Yucca Mountain calcretes and surficial veins, assuming that all carbon contained in vadose zone gases and dissolved in subsurface fluids originates solely from the atmosphere-biosphere source.
- o This is a reasonable assumption but only for terrains that are devoid of: a) carbonate and other carbon bearing rocks; and b) Plio-Quaternary volcanism. At the Nevada Test Site, however, there is direct and clear evidence that both of these complicating circumstances are present.
- o In the absence of evidence to the contrary and recognizing that the Nevada Test Site upwelling hydraulic "source" fluids carry values of the calcite saturation index $\log (Q/K) > 0$, it is only prudent to expect that, in the Yucca Mountain vadose zone, the resident CO_2 may be a three end-members mixture. The potential end-members are: i) the atmospheric CO_2 - mean value of the $\delta^{13}\text{C}$ ratio is -7.0 per mil PDB , Keeling (1961); ii) the biogenically derived CO_2 - mean value of the $\delta^{13}\text{C}$ ratio is -24 per mil PDB , for ~ 85 percent biomass with the C-3 metabolic pathway; and iii) the subterranean CO_2 - for volcanic and geothermal areas, value of the $\delta^{13}\text{C}$ ratio ranges from 0 to -9 per mil PDB , Deines (1980).

General remarks - interpretation of the origin of CO_2 respired from the Yucca Mountain vadose zone.

- o Interpretations of the results of studies of spatio-temporal distribution of the carbon indexes ($\delta^{13}\text{C}$, PMC, and $\log P_{\text{CO}_2}$), from samples of the Yucca Mountain vadose zone CO_2 , must be made recognizing that, at the vadose zone base, there are very substantial lateral temperature gradients, $dT/dx \rightarrow 10^\circ\text{Celsius per km}$.
- o The lateral temperature gradients, with the observed magnitudes, virtually assure that, in the Yucca Mountain vadose zone, convective circulations of gaseous phases are taking place.
- o Fluctuations involving both the barometric pressure and the atmospheric temperature virtually assure that the convective circulations occur as the non-periodic flow, Lorentz (1963).
- o It may be expected, therefore, that $\sigma_{\phi_{xyz(t)}} \neq 0$ [for a given sampling site, $\sigma_{\phi_{xyz(t)}}$ denotes amplitude of fluctuations involving either the $\delta^{13}\text{C}$ ratio, the PMC index, or the $\log P_{\text{CO}_2}$ index].

Introduction - interpretation of the results of studies of spatio-temporal distribution of the carbon indexes in the Yucca Mountain vadose zone.

Location of the CO₂ study area. Relative to *in-situ* temperatures at a depth of 350 m. The Yucca Mountain - vadose zone. From Szymanski, 1989.

Note:

- $dx/dt = -\sigma x + \sigma y$, $dy/dt = Rx - y - xz$, and $dz/dt = -bz + xy$ - Lorenz's equations from Kelis-Borok (1990);
- functions x , y , z characterize the intensity of convection stream, horizontal and vertical temperature gradients, respectively;
- σ , b , and R are numerical parameters; and
- dx/dt controls values of the $\delta^{13}\text{C}$ ratio, the PMC index, and the partial pressure $\log P_{\text{CO}_2}$.

Non-periodic convective flow. Modified from Gleick (1987) and Kelis-Borok (1990).

a) CO_2 originates solely from the atmosphere-biosphere source.

b) CO_2 originates from both the atmosphere-biosphere source and the subterranean source.

Explanation:

- ϕ - denotes value of either the $|\delta^{13}\text{C}|$ ratio or the PMC index, $\bar{\phi}$ denotes mean value;
- $\bar{\phi}_{xy(z)}$ - denotes $\bar{\phi}$ vs depth gradient, for a given vertical reference profile; and
- $\sigma\phi_{xyz(l)}$ - denotes amplitude of ϕ fluctuations, for a given reference point.

Note:

- a) the subterranean source yields CO_2 that, relative to the atmosphere-biosphere derived CO_2 , contains smaller concentrations of carbon-14 and carbon-12.

Interpretation key - origin of CO_2 respired from the Yucca Mountain vadose zone.

Isotopic character of carbon contained in the carbon dioxide from the vadose zone as a function of depth.

The carbon-14 content in the carbon dioxide from the vadose zone as a function of depth.

Explanation:

- USW UZ-1 - time history, period from November 1983 to June 1985, see Yang et al. (1989) for details; and
- UZ6S and N-borehole data represent one-time measurements.

$\log P_{CO_2}$ as a function of depth and time. The carbon dioxide from the Yucca Mountain vadose zone

Conceptual understanding, the spatio-temporal distribution of the carbon indexes - the Yucca Mountain vadose zone.

Note:

- $N = 47$ samples for the hydrosphere, $N = 125$ for the vadose zone; and
- $\log P_{\text{CO}_2}$ data from the vadose zone and from the hydrosphere are from White and Chuma (1978), Yang et al. (1985), Thordarson et al. (1989), and Kerrisk (1987).

Histogram. Partial pressure of CO₂. The Nevada Test Site hydrosphere.

Note:

- $\log(Q/K)$ data are from Kerrisk (1987);
- $N = 52$ for hydraulic "source" regions; $N = 64$ for hydraulic "sink" regions; and
- Q - the ion activity product, K - the equilibrium constant for the solubility reaction.

Calcite saturation index. Water samples from the Nevada Test Site hydraulic "sink" and "source" regions.

Explanation:

- sample represents the tuff "pile" based hydraulic sink region;
- ◐ sample represents the tuff "pile" based hydraulic source region; and
- sample represents the Paleozoic carbonates based hydraulic source region.

$\delta^{13}\text{C}$ vs PMC field. Water samples from the Nevada Test Site hydrosphere.

Isotopic character of carbon. Water samples from the Paleozoic carbonates based hydraulic "sink" and "source" regions.

a) N=35 for hydraulic "source" regions; N=14 for hydraulic "sink" region.

Isotopic character of carbon. Water samples from the alluvium-tuff pile based hydraulic "source" and "sink" regions.

Note:

- the fractionation equation is from Turi (1986);
- mean value of the $\delta^{13}\text{C}$ ratio, for the sub-lithospheric mantle indigenous CO_2 , may be taken as ~ -6 per mil_{PDB}, Hoefs (1987);
- depending upon reaction temperature, dissolution of the sub-lithospheric mantle indigenous CO_2 may be expected to produce fluids with values of the $\delta^{13}\text{C}$ ratio ranging from $+1.45$ ($T = 20^\circ\text{Celsius}$) to -10 per mil_{PDB} ($T = 200^\circ\text{Celsius}$); and
- the expected range of the $\delta^{13}\text{C}$ ratio, for fluids produced through dissolution of the sub-lithospheric mantle indigenous CO_2 , is similar to that observed for the Nevada Test Site geothermal fluids.

Plausibility assessment - hypothesis that the abnormally low values of the $\delta^{13}\text{C}$ ratio, for the Paleozoic carbonates based "source" fluids, may be reflecting influx of the sub-lithospheric mantle indigenous CO_2 .

- o A fairly firm support for the hypothesis that, at the Nevada Test Site, some of the circulating carbon is derived from the local deep-seated (igneous) sources of carbon may be inferred from the results of carbon isotopic studies of samples of the carbonate gangue minerals associated with the hydrothermal ore deposits. Specifically, the results of such studies indicate that:
- i) the parent fluids for the hydrothermal ore deposits, typically, carry values of the $\delta^{13}\text{C}$ ratio ranging from -10 to about -2 per mil $_{\text{PDB}}$, Hoefs (1987);
 - ii) the hydrothermal ore forming fluids carry the dissolved carbon that is: a) isotopically "lighter" than carbon acquired during dissolution of marine limestones; and b) isotopically "heavier" than carbon produced by the biogenic sources;
 - iii) during early stages of the ore forming periods, which presumably correspond with peaks of igneous activity and periods with the most intense influx of the deep-seated CO_2 , the hydrothermal fluids carry relatively low values of the $\delta^{13}\text{C}$ ratio. [This very consistent observation may be taken to indicate that: a) dissolution of the deep-seated CO_2 is occurring at fairly high temperatures and b) the deep-seated carbon source predominates]; and
 - iv) during vanishing stages of the ore forming periods, which presumably correspond with termination of igneous activity and periods with the accordingly reduced influx of the deep-seated CO_2 , the hydrothermal fluids carry higher values of the $\delta^{13}\text{C}$ ratio [This observation may be taken to indicate that: dissolution of the deep-seated CO_2 is occurring at lower temperatures; and b) the marine limestone reservoir of carbon becomes important].

Further plausibility assessment - hypothesis that the abnormally low values of the $\delta^{13}\text{C}$ ratio, for the Paleozoic carbonates based "source" fluids, may be reflecting influx of the sub-lithospheric mantle indigenous CO_2 .

a) the Carlin disseminated gold deposit of Nevada

b) the Cortez disseminated gold deposit of Nevada

Note:

- for both deposits host rock is the Paleozoic limestone;
- samples of the carbonate gangue minerals, from the Carlin and Cortez deposits, yield values of the $\delta^{13}\text{C}$ ratio ranging from +1 to -2 per mil PDB and from 0 to -4 per mil PDB , respectively;
- White (1985) have reported that both of the gold accumulations were formed at a temperature ranging from 150 to 200°Celsius - the corresponding value of the equilibrium fractionation factor is $10^3 \ln \alpha_{\text{HCO}_3^- - \text{CO}_{2(g)}} = \sim -3.0$ per mil PDB , Hoefs (1987);
- the parent fluids for the Carlin and Cortez deposits have carried values of the $\delta^{13}\text{C}$ ratio ranging from -2 to -5 per mil PDB and from -3 to -7 per mil PDB , respectively; and
- observation that the marine limestone based, and hydrothermal ore forming, fluids have carried the abnormally low values of the $\delta^{13}\text{C}$ ratio may be taken to indicate the deep-seated (igneous) origin of the dissolved carbon.

The isotopic character of carbon contained in the carbonate gangue minerals associated with the Carlin and Cortez disseminated gold deposits of Nevada.

Explanation:

- - carbonate gangue minerals; and
 • - fluid inclusion

Note:

- relict fluids entrapped in fluid inclusions from samples of the carbonate gangue minerals, associated with various hydrothermal ore deposits, yield values of the $\delta^{13}\text{C}$ ratio ranging from -4 to about -10 per mil PDB ;
- for individual ore deposits, the different generations of the carbonate gangue minerals are arranged from early to late, as shown by arrows;
- the observed trend toward the isotopically "heavier" carbon (with decreasing age of the carbonate gangue minerals) may be taken to indicate that, as the volcanic activity and the resulting geothermal circulations decay, the contribution of CO_2 from the deep-seated igneous sources also decays; and
- observation that the hydrothermal ore forming fluids carry values of the $\delta^{13}\text{C}$ ratio ranging from -2 to about -10 per mil PDB may be taken to indicate the deep-seated (igneous) origin of the dissolved carbon, Hoefs (1987).

The isotopic character of carbon contained in samples of the carbonate gangue minerals associated with various hydrothermal ore deposits. From Hoefs (1987).

Overall conclusions - the result of isotopic considerations of carbon contained in the Yucca Mountain vadose zone and dissolved in fluids comprising the Nevada Test Site hydrosphere, allow for two important conclusions to be drawn.

Conclusion I - The carbon dioxide respired from the Yucca Mountain vadose zone is provided by two independent CO₂ sources. These are:
i) the atmosphere-biosphere; and ii) the subterranean fluids.

Conclusion II - At the Nevada Test Site, the circulating carbon appears to be provided by three independent sources of carbon. These are:
i) the atmosphere-biosphere; ii) the Paleozoic carbonates; and iii) the deep-seated igneous bodies.

Overall conclusions - an inquiry into the origin of carbon that is contained in the CO₂ gas respired from the Yucca Mountain vadose zone and is dissolved in the Nevada Test Site hydrosphere.

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PART 5

News and Comment, *Science*

The Geopolitics of Nuclear Waste

A scientist's contention that the United States is building its waste site in a region of unstable hydrology has unleashed a flood of controversy, but it has won few supporters

YUCCA MOUNTAIN, A BLEAK VOLCANIC RIDGE in the Nevada desert, emerged from obscurity in the 1980s when it was chosen as a potential graveyard for the intensely radioactive waste produced by U.S. nuclear plants. In 1987, Congress decided to strike all other candidates from the list and, as waste accumulates in utility storage ponds, this remote and unpopulated hillside has become an epicenter of controversy.

The program to build a waste repository at Yucca Mountain, run by the Department of Energy (DOE), has been criticized by virtually every Nevada politician. Now DOE scientists at the site have also been drawn into a bitter and paralyzing debate about risks to public health, focused at the moment on some arcane aspects of local geology. DOE has been told that it must find a place where deadly waste canisters can be buried without disturbance for 10,000 years.

It is hard to find anything that remains unchanged for that long, and the attempt to use science to "prove" that Yucca Mountain will remain dry and stable for 10 millennia is causing trouble.

The big item at the moment is the risk of flooding. The debate on this concern is likely to erupt again in the next few weeks as DOE releases the latest of several expert reviews of a disturbing theory developed by one of DOE's own staffers

in Las Vegas, an engineering geologist named Jerry Szymanski. DOE paid for this study after Szymanski criticized an earlier (and negative) review of his ideas as biased.

Szymanski thinks the proposed \$15-billion repository would sit on top of an intensely active structure that, if altered by an earthquake, would send a slug of groundwater up from deep in the mountain into the waste storage area. The canisters in it, if still hot, would produce steam and possibly break, leaking radionuclides to the outside world.

Szymanski's theory, and particularly his methods of promoting it, have upset a lot of people. It has already been slammed in two formal reviews and has virtually no support among geologists. Some are openly hostile.

For example, John Stuckless of the U.S. Geological Survey, coordinator of geochemical studies for Yucca Mountain, dismisses the Szymanski thesis, saying, "it reminds me of clinical ecology," a dubious medical specialty that links cancer to all kinds of manufactured products, including common plastic. "The guy's a moving target," adds Douglas Rumble, a geochemist at the Carnegie Geophysical Laboratory in Washington, D.C. He says Szymanski has never published any peer-reviewed articles and seems to change his arguments each time he's challenged.

But other researchers in DOE and the USGS praise Szymanski for coming up with original ideas about the geology of the "basin and range" in southwestern Nevada. Most, however, are highly skeptical that a flood could occur at Yucca Mountain's repository level, more than 300 meters above

the other way and "trying their level best to defend the indefensible." Asked for his opinion of the plan to put radioactive waste in Yucca Mountain, Price replied, "The only worse thing that I can think of would be tipping it down the volcanoes in Hawaii."

Carl Gertz, DOE's director of the Yucca Mountain project, says he intends to resolve the controversy by submitting it to a thorough, objective peer review. He has already spent more than \$1 million on peer review and is not yet close to solving his problem. The definitive study is likely to be performed by a 17-member panel at the National Academy of Sciences, chaired by C. Barry Raleigh of the University of Hawaii. It's due by the end of the year.

Meanwhile, other forces converging on Yucca Mountain will soon draw attention to the scientific arguments over the site. For one, state officials in Nevada, who have been using every obstacle at their command to block the project, are withholding the permits DOE needs to finish its exploratory research. The state also has filed suit in federal court to stop the program on constitutional grounds. Though the appeals court last year rejected its arguments unanimously, the final word may come down from the Supreme Court in the next month or two. Now the Bush Administration reportedly has included in its energy plan a section that would remove Nevada's authority to issue (or deny) environmental permits at Yucca Mountain, handing it over to a federal agency instead.

The scientific dispute has been simmering since 1987, when Szymanski, an expert on nuclear plant siting with a master's degree from the University of Warsaw, released a paper describing his theory. As he says, his ideas are "very novel" and "unorthodox." The tectonic and convective forces beneath Yucca Mountain, he claims, are so volatile that they could rearrange the geology of the site in a relatively short time, during the 10,000-year lifetime of the repository. This is a "certainty," in his view. He argues that there is good evidence that a catastrophic rise in the water table could occur and that, in fact, it has occurred in the past.

To grasp the situation, Szymanski argues, one must understand a set of interacting

Trench warfare. Do calcite deposits in trench 14 come from ancient groundwater or rainwater?

the present water table, deep in a parched desert.

A handful of researchers do take Szymanski seriously, however. Neville Price, for example, one of two people nominated by Szymanski to serve on a five-person independent panel reviewing his claims for DOE, gives his unreserved support. For 20 years Price taught structural geology at Imperial College, London, and is now an oil company consultant associated with University College, London. Price says that he and Szymanski's other panel nominee, seismologist Charles Archambeau of the University of Colorado, are "absolutely convinced" that Szymanski is right. The other three members of the panel, says Price, are leaning

thermal and hydrologic forces at work in the rocks under the desert. He maintains that Yucca Mountain now sits on what he calls a local "sink." The extensional (pulling apart) tress in its rocks is exceptionally high, the fractures are open and capable of absorbing large quantities of water, the water table is low, and the flux of heat from deep in the earth is suppressed. Adjacent to it, he says, are "source" regions, where all these conditions are essentially reversed: the stress is low, the water table is high, and convective heat moves upward more vigorously, producing higher temperatures near the surface. Szymanski contends that there is nothing permanent about the spatial location of a sink or source region; that, in fact, they move about the topography depending on conditions in the underlying rock. He also claims to be able to identify these regions by the distinctive "fingerprints" produced by the ratio of certain isotopes in local deposits.

Yucca Mountain, Szymanski says, though now a sink, has been a source in the past and will be a source again in the future. If it changes from one condition to the other within the next 10,000 years, he argues, the proposed repository site—now more than 300 meters above the present water table—will most certainly be swamped.

Szymanski put these ideas together as he was coordinating some geological studies for the Yucca Mountain program and wrote them up in a 322-page report. In December 1987, he sent copies to Gertz and a colleague working for the state of Nevada, who passed it along to then Governor Richard Bryan (Democrat). Bryan had run for office as an opponent of the repository and since then has been elected to the U.S. Senate on the same platform. When he got Szymanski's unreviewed paper, he called in the press and distributed it, using it as ammunition in his campaign against DOE.

Scientists at DOE and the Geological Survey, some of them with decades-long experience in the complex geology of the Southwest, were furious. They are still angry, and some of their outrage surfaced last month in *Science*, the journal of the American Geophysical Union. AGU president G. Brent Dalrymple of the U.S. Geological Survey blasted the press, Szymanski, and Szymanski's allies in a

front-page editorial. Popular accounts portray Szymanski as a lone, "little guy fighting the big, dumb bureaucracy," Dalrymple claimed. But, he wrote, in this case the loner "bypassed the conventional routes" for testing hypotheses and gaining scientific support. Instead, he "released his findings to the popular press and to those politicians eager to be rid of any waste disposal site." Dalrymple adds that "dozens of scientists have reviewed [Szymanski's] work and found little merit in it."

Many geologists who have read Szymanski's papers agree that the significance of his forecast has been exaggerated, even though his description of the forces in the basin and range makes sense. But the USGS experts argue that there is almost no evidence to support his claim that there will be a large, long-term shift in the water table. Mathematical models created at the Sandia

may be scarce, Archambeau, one of Szymanski's supporters, says he knows of two earthquakes that produced over a cubic kilometer of fluid—in Japan and near the Nevada-California border—within the past 25 years by "seismic pumping." This phenomenon is usually short-lived, but even a short gush such as this could damage the Yucca Mountain repository if it were large enough. However, Archambeau could not cite any well-established cases in which the water table had shifted hundreds of meters as a result of an earthquake—as it would have to do to fulfill Szymanski's prediction for long-term flooding of the repository. On the other hand, Archambeau claims that Death Valley, California, which is near Yucca Mountain, is full of high-elevation springs and may represent what Yucca Mountain would look like after a shift in the sink/source locations.

As proof that the water table was high at

Yucca Mountain in the past and could be high again, Szymanski has focused attention on a series of calcium carbonate and opal veins exposed in an exploratory pit, known as "trench 14," near the top of the mountain. Here, he has told many people, is clear evidence that groundwater had once covered most of the hillside, leaving behind some thick mineral deposits. Price also insists that these veins are an "unequivocal" sign that groundwater flowed through the area. "The structures of the

veins that I saw in trench 14 and elsewhere are like the veins I have seen in mountains in areas around the world.... One can explain them in terms of periodic upwelling of fluids."

Two researchers, Jay Quade and Thure Cerling of the University of Utah, have an alternative explanation for the deposits at a high elevation: rainwater. They recently published their evidence in *Science* (14 December 1990, p. 1549). By sampling deposits where the source of water is well known, they established a set of isotope values that they could use to identify carbonates created by deep groundwater, those created by trapped or "perched" water at a high elevation, and those created by rainwater. They then demonstrated that the carbonates in trench 14 match up with local perched or rainwater deposits and not with the groundwater deep below Yucca Mountain.

Archambeau is not persuaded. He sees no reason why trench 14 carbonates must match the signature of present-day groundwater beneath Yucca Mountain. He says that other

DOE

Hot rocks. A novel geological theory has set off an intense debate among scientists working for Carl Gertz, chief of DOE's waste repository program at Yucca Mountain, Nevada.

and Lawrence Livermore National Laboratories—according to George Barr of Sandia—show that under the most ex-

aggerated assumptions, the long-term changes will be no more than "tens of meters" up or down. Furthermore, says Isaac Winograd, an early student of this area known to some as the USGS "guru" on Yucca Mountain, the historic record indicates that no earthquake has shifted the water table by more than about 10 meters in this area of the Southwest. More important, Winograd says, even these fluid excursions don't last long, perhaps a year at most. "Szymanski tends to ignore geologic time," he says.

While the evidence for large water flows

groundwaters in the region have an isotopic signature like the trench 14 carbonates, and he suggests that in the past, when these carbonates were formed, Yucca Mountain may have been gushing with groundwater of a different kind—the kind that is typical of a Szymanski source zone.

Though Archambeau and Price have helped give Szymanski's theory credibility, it has yet to pass muster in formal peer reviews. In fact, it has flunked the two reviews completed so far. The first was conducted by a group of experts retained by the state of Nevada and the second by a panel of federal scientists chaired by William Dudley, Jr., of the USGS. Though only the DOE-USGS

review has been made public, *Science* has obtained, among others, a critique performed for the state by University of Nevada seismologist James Brune. In it, Brune comments that Szymanski's thesis seems "unlikely" and appears to be "scientifically unsound." Yet he writes that the possibility that it is correct "cannot be discounted."

Brune judged the discussion of earthquakes to be especially weak because it leaned on a theoretical model developed by a Bureau of Mines geophysicist named Brian Brady. In the early 1970s, Brady proposed his own method for predicting earthquakes and then, in an infamous case, went on to apply it. He forecast that a major quake would hit a town

in Peru on a specific day. It caused an international flap and a local panic, and when the day came, there was no quake. Brune says that an analysis performed by Clarence Allen of the California Institute of Technology found Brady's theory to be "completely untenable." The fact that Szymanski relied so much on it, Brune wrote, "casts a great deal of doubt on the validity of this part of the report."

Asked if he had read these comments and written any response, Szymanski told *Science* he had not bothered. He dismissed them as the work of contractors, whom he speaks of as "fleas." However, Brune was pleased to see that Szymanski's final draft—a sprawling 911 pages long—omits the Brady references.

Flap Erupts Over Dioxin Meeting

Everything about dioxin is so politically charged, even the science, that the organizers of a Banbury Center dioxin conference last fall expected their meeting would generate some controversy. But no one anticipated the furor that erupted in early February when participants learned that a public relations firm hired by the Chlorine Institute, which helped pay for the conference, was circulating a "consensus" summary of the meeting. The document, not surprisingly, supports the industry line on dioxin.

"I'm outraged," says Ellen Silbergeld, a toxicologist at the University of Maryland, who insists that the press materials—which none of the participants approved—misrepresent her views. "I agreed to participate based on my previously held high regard for Banbury and Cold Spring Harbor," she wrote in a letter to Banbury Center director Jan Witkowski. "I did not expect to be manipulated by industry or government spokespeople." Silbergeld sent copies of her letter to all the participants and to the press.

"It was basically mishandled," sighs Witkowski, who admits to a degree of naïveté in dealing with such a "highly politicized" issue as dioxin. He has just written a letter to all the meeting participants explaining that the Banbury Center in no way authorized the press materials that were sent out. "I am very sorry that what was intended to help promote the goals of the meeting has gone so awry," he wrote.

The intensity of Silbergeld's reaction may say more about the extreme sensitivities over dioxin than about the culpability of any of the parties involved, all of whom have taken some heat. The flap nevertheless illustrates what can go wrong when the roles and agendas of sponsors and participants aren't clearly spelled out.

What Silbergeld, who was formerly with the Environmental Defense Fund, finds most disturbing about the events is the "violation of process." And she has found an unexpected ally in Vernon Houk, director of the Center for Environmental Health and Injury Control at the Centers for Disease Control. Houk is in almost total disagreement with Silbergeld on the dangers of dioxin—he believes that they have been vastly overrated. Yet Houk told *Science* that he, too, is "disturbed" by the publicity campaign. "I don't think it is fair to

represent consensus when none was really sought," says Houk. Silbergeld's view is also supported by a number of the other participants, albeit with somewhat less emotion, who say they feel manipulated and misused.

When the Chlorine Institute, an industry trade group, approached Banbury director Jan Witkowski early last year about holding a dioxin meeting, he had no inkling it would turn out differently from the 15 or so other meetings the center runs each year, some of which are also sponsored by industry. And to the Chlorine Institute, the time was ripe for another look at dioxin's risks, given shifting sentiments among at least some scientists that dioxin may be less dangerous than previously believed. Chlorine Institute officials thus believed that a scientific meeting could be "beneficial to our interests, particularly our interest in the paper industry," explains Joe Walker, the institute's head of communications. Walker is referring to the enormous pressure the paper industry is now under to reduce the amount of dioxin that arises as a by-product in the bleaching process.

The Chlorine Institute lined up Robert Scheuplein of the Food and Drug Administration to run the meeting. He then asked Michael Gallo, a toxicologist at the Robert Wood Johnson Medical School in New Jersey, and Cornelius A. van der Heijden, a regulatory official in the Netherlands, to chair the meeting with him. From then on, the Chlorine Institute studiously kept itself out of the picture, say both Witkowski and the meeting organizers.

The three organizers picked all participants, with one exception: George Carlo, who was invited as an observer for the institute at its request. Carlo, an epidemiologist and lawyer who heads the Health and Environmental Sciences Group in Washington, D.C., is a regular consultant to the institute.

The meeting itself went surprisingly well; indeed, some agreement was reached by people from opposite sides of the dioxin debate on a number of issues. For example, there was nearly unanimous agreement that, in light of a new understanding of dioxin's molecular actions, the Environmental Protection Agency should take another look at how it assesses the risk of this chemical. Several people, most notably the meeting organizers, speculated

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JAN WITKOWSKI

The Dudley report, issued by DOE in July 1989, was just as harsh as Brune's analysis. The summary said that while some reviewers found Szymanski's ideas "constructive" and "stimulating," "most doubt that there is a sufficient technical basis to warrant pursuing author's hypotheses further." The authors singled out one problem in particular that others have complained about since—namely, that the thesis provides no numerical model that can be used to estimate the magnitude, frequency, or duration of the events Szymanski postulates.

Szymanski did not respond in detail to these criticisms, either, but said they are typical of the kind that come from people

who have been trained to think about geology and hydrology in static terms and are hostile to new ideas. "I do not want to be presumptuous," he said, "but prior to Charles Darwin, people had the idea that species remain unchanged; Darwin showed that they evolve. The situation is similar with hydrology."

Although Szymanski has taken a couple of heavy blows in the first rounds of peer review, the debate has not yet run its full course. Judging by the kind of arguments Price and Archambeau are using now—comparing the veins in trench 14 with even thicker (and relatively young) deposits elsewhere on Yucca Mountain—DOE and the

USGS may have to collect a lot more information on the quality, size, and location of carbonate deposits in the area before they will be able to present a complete case on the origin of the material in trench 14.

Meanwhile, the DOE staff continues to plug away at the massive "site characterization" effort at Yucca Mountain, costing about \$1 million a day. Gertz says the government is prepared to continue in this investigative mode, spending up to \$4 billion, until it is satisfied that it has laid to rest all reasonable concerns about safety. That is a huge financial investment. But at least it is paying for some intriguing geological research. ■ ELIOT MARSHALL

that such a reexamination would reveal that EPA overestimates how risky dioxin is—though others, like Silbergeld, rejected that idea (*Science*, 8 February, p. 624).

But the harmony was to prove short-lived. In retrospect, most participants with whom *Science* spoke say they failed to realize that the meeting was sponsored by industry, along with the EPA, even though the invitation clearly said so. And none of the participants *Science* spoke with, nor even meeting co-organizer Gallo, knew Carlo was there as the institute's observer. Silbergeld, Houk, and others say they have no problem with industry sponsorship of scientific meetings—as long as each one's role is clear.

At the meeting Carlo heard the message the Chlorine Institute hoped he would. The institute's intention, from the outset, was that "if the conference outcome was favorable we would take advantage of it and bring it to the attention of key people in the media," says Walker. They hired Edelman Medical Communications to do just that.

Witkowski saw no problem with the institute publicizing the results of the meeting—after all, they paid for half of it—as long as it was well done. But what he thought would be a straightforward press release turned into a fiasco of crossed signals and miscommunication. For example, Gallo, Scheuplein, and van der Heijden were delighted to cooperate—and indeed, each wrote his own one-page summary of the meeting for the public relations firm. But Gallo and Scheuplein now complain that they had assumed that Edelman was representing the Banbury Center, not the Chlorine Institute.

In addition, Witkowski's only condition in agreeing to the press release was that he have final approval. As agreed, the Edelman staff sent him the cover letter to the press, which he edited to remove any references to the Cold Spring Harbor Laboratory. But in December, Edelman sent out a press packet that included, along with the statements from the three chairmen alluded to in the letter, a background paper purportedly written by Carlo that none of the organizers or participants had seen. The paper asserted, among other things, that the Banbury meeting "reinforced the notion that dioxin is much less toxic to humans than originally believed."

The cover letter also differed from the version Witkowski had approved in two important respects. First, it referred to the meeting

No consensus. Ellen Silbergeld's objection to the press release won support from other participants.

as a "consensus conference." Says Witkowski: "Just those two words set the whole framework. It was not a consensus conference. That is what is causing all the trouble." What's more, the draft he approved carried a disclaimer, which was missing from the final version, saying that the three statements were the views of the chairmen and not the conference as a whole. Meeting organizers Gallo and Scheuplein also object to the letter—essentially to the two offending words—and insist that they never saw it.

When Silbergeld got hold of the press packet from a reporter at the end of January, she exploded, writing a letter to Witkowski that took all of the parties to task. Events then took a bizarre twist when Carlo, who received a copy of Silbergeld's letter, called Witkowski to say he had not written the paper after all and had no idea how his name ended up on it.

Carlo has since launched a massive telephone campaign—calling the participants and this reporter repeatedly—to clear his name. Carlo concedes that he did work with Edelman and the Chlorine Institute in developing the paper but asserts, nonetheless, that, "No one has the right to put someone's name on a document."

Nancy Turett, senior vice president at Edelman, admits to putting Carlo's name on the paper—she says because he was so extensively involved in drafting it. "The end product is very much a reflection of what Dr. Carlo thought should be in it," says Turett, who says she assumed that the Chlorine Institute had cleared the final document with Carlo. Institute officials say, meanwhile, that they thought Edelman had gotten Carlo's permission.

Turett also takes the blame for the revised letter, calling it a simple editorial mistake. "I thought there was a consensus. There is no big agenda. It is just a cover letter." She called *Science* back to add, however, that Edelman never distributes a single word without the client's approval.

All of which leaves Witkowski at the Banbury Center shaking his head in disbelief. He realizes, much to his dismay, that this flap threatens to "poison" the entire meeting and undermine the fragile—and unexpected—agreement the group forged on the molecular action of dioxin. "That is the kind of thing you would hope would happen at a scientific meeting," says one participant who asked to remain unnamed. "It was like a little flower just starting to bloom, then along came this mess. I feel very sad." ■ LESLIE ROBERTS

PART 6

Letter to Szymanski

20 June, 1991

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Yucca Mountain Site Characterization Project Office
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Dear Jerry:

We just received your letter dated 13 May, 1991. Thank you for your concise comments.

Sincerely yours,

Thure E. Cerling

Jay Quade

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