UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

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FROM: Paul T. Prestholt, Sr. On-Site Licensing Representative

DATE: March 29, 1991

SUBJECT: 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 find enclosed the above-referenced document.

PTP:nan
cc: Joe Holonich w/o enc.

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NH03

WBS 1.2.9
QA: N/A

JAN 18 1991

Editor, Science
1333 H Street N.W.
Washington, DC 20005

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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

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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 del^{13}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 del^{18}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 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 ddel^{18}O/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,

"ORIGINAL SIGNED BY"

Jerry Szymanski

Jerry S. Szymanski, Physical Scientist
U.S. Department of Energy
Nevada Operations Office

RSED:JSS-1769

JAN 18 1991

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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.

JAN 18 1991

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ATTACHMENTS

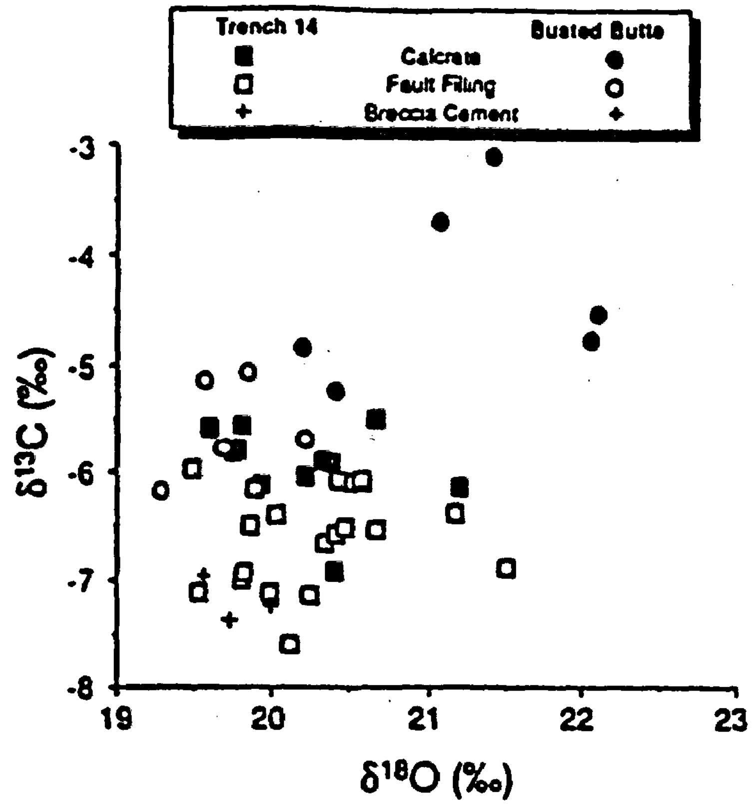
Part A - Isotopic Comparative Analyses

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

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

Part A - ISOTOPIC COMPARATIVE ANALYSES

Figures 1 through 13b



Note:

a) The carbon-oxygen part of the isotopic fingerprint, for the Yucca Mountain surficial calcite-silica deposits, is a range of values for the $\delta^{18}O$ and $\delta^{13}C$ ratios;

b) The observed range for the $\delta^{13}C$ ratio is from −3.0 to −7.5 per mil $_{PDB}$; and

c) The observed range for the $\delta^{18}O$ ratio is from 19.2 to 22.0 per mil $_{SMOW}$.

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

- 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.

- 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 $_{PDB}$.

- 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 $_{PDB}$.

- A mean value for the $\delta^{13}C$ ratio for carbon dioxide produced by the local plants is ~ 24 per mil $_{PDB}$ - (27 per mil · 85 percent + 12 per mil · 15 percent)/100.

- Dissolution of the locally produced biogenic CO_2 will yield fluids with values of the $\delta^{13}C$ ratio of about 16 per mil $_{PDB}$($10^3 \ln \alpha_{HCO_3^- - CO_2}$ ~8 per mil $_{PDB}$).

- At temperatures less than 50°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:

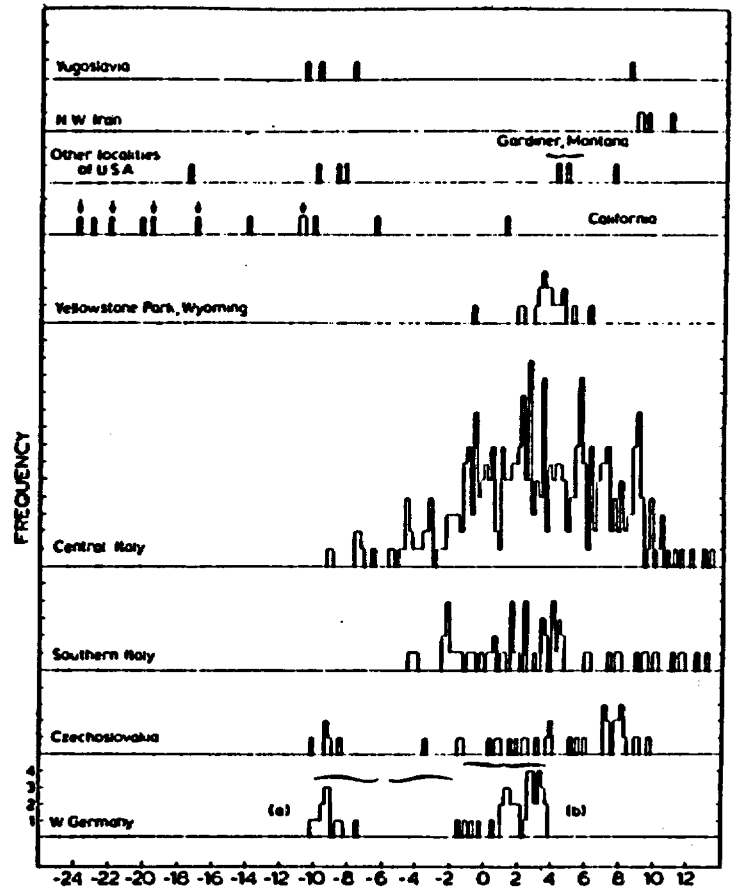
a) Carbon isotopic fractionation data are from Hoefs, 1987; and

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.



$\delta^{13}C_{PDB}$ (per mil) - travertines

Note:

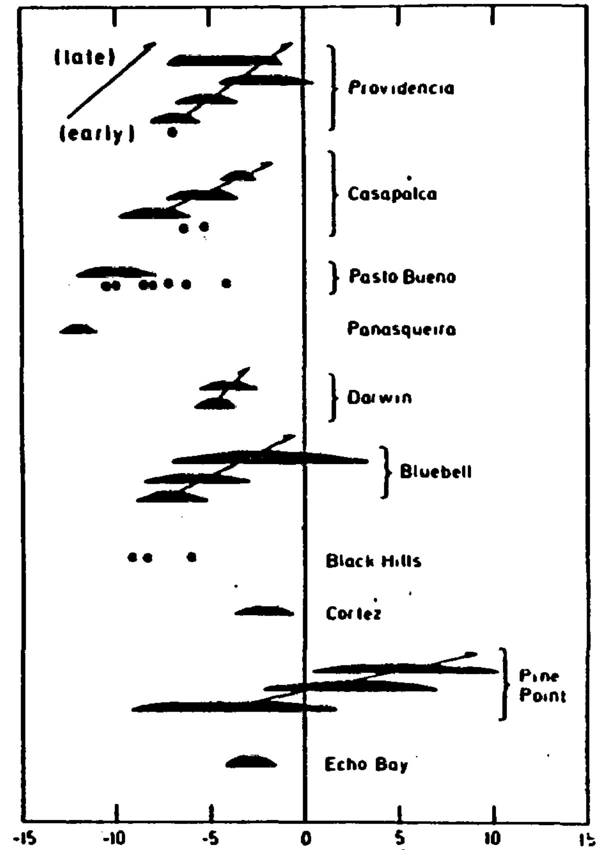
a) In some regions, carbon contained in travertine deposits is largely derived from dissolution of marine limestones and, therefore, mean values for the $\delta^{13}C$ ratio tend to be positive, say +3.0 per mil $_{PDB}$; and

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.



$\delta^{13}C$ (per mil)$_{PDB}$ - carbonate gangue from hydrothermal ore deposits

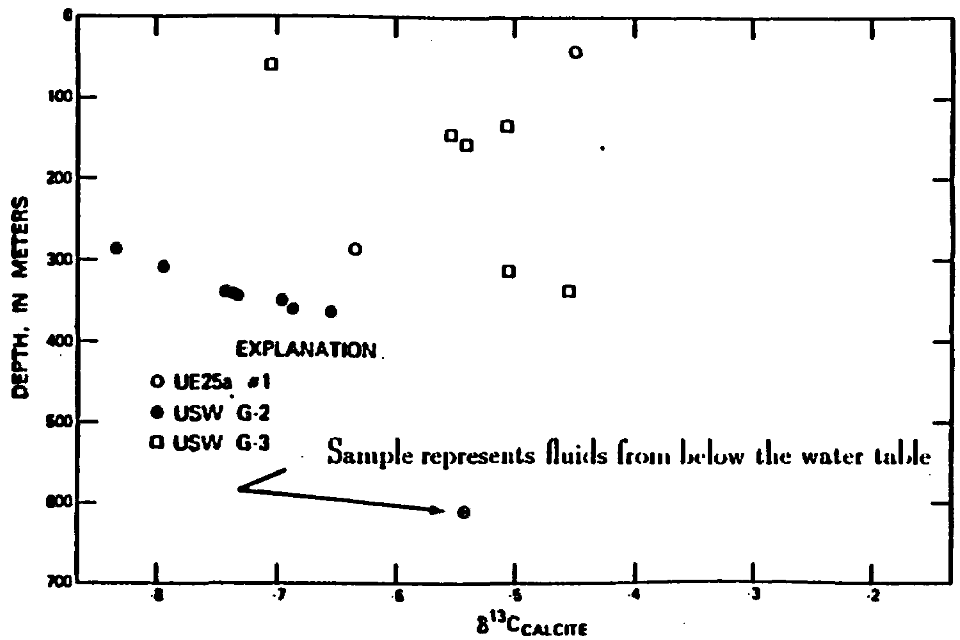
Note:

a) The isotopic character of carbon contained in CO_2 derived from deep-seated, 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 $\left(10^3 \ln \alpha_{HCO_3^- - CO_{2(a)}} = 9.483 \cdot 10^3 \cdot T^{-1} - 23.89\right)$, dissolution of the igneous CO_2 may yield geothermal fluids with values of the $\delta^{13}C$ ratio ranging from +1 (temp. ~ 20°Celsius) to -14 per mil $_{PDB}$ (temp ~ 300°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 file - isotopic character of carbon contained in carbonate gangue minerals from a number of hydrothermal ore deposits. From Hoefs, 1987.



Note:

a) Values of the CO_2 partial pressure for the Yucca Mountain subsurface fluids, as calculated and reported by Kerrisk (1987), range from $\log P_{CO_2} = -0.79$ to $\log P_{CO_2} = -2.86$ atm. The mean local value of $\log P_{CO_2}$ is well above the CO_2 partial pressure in the atmosphere ($\log P_{CO_2} \sim -3.5$ atm);

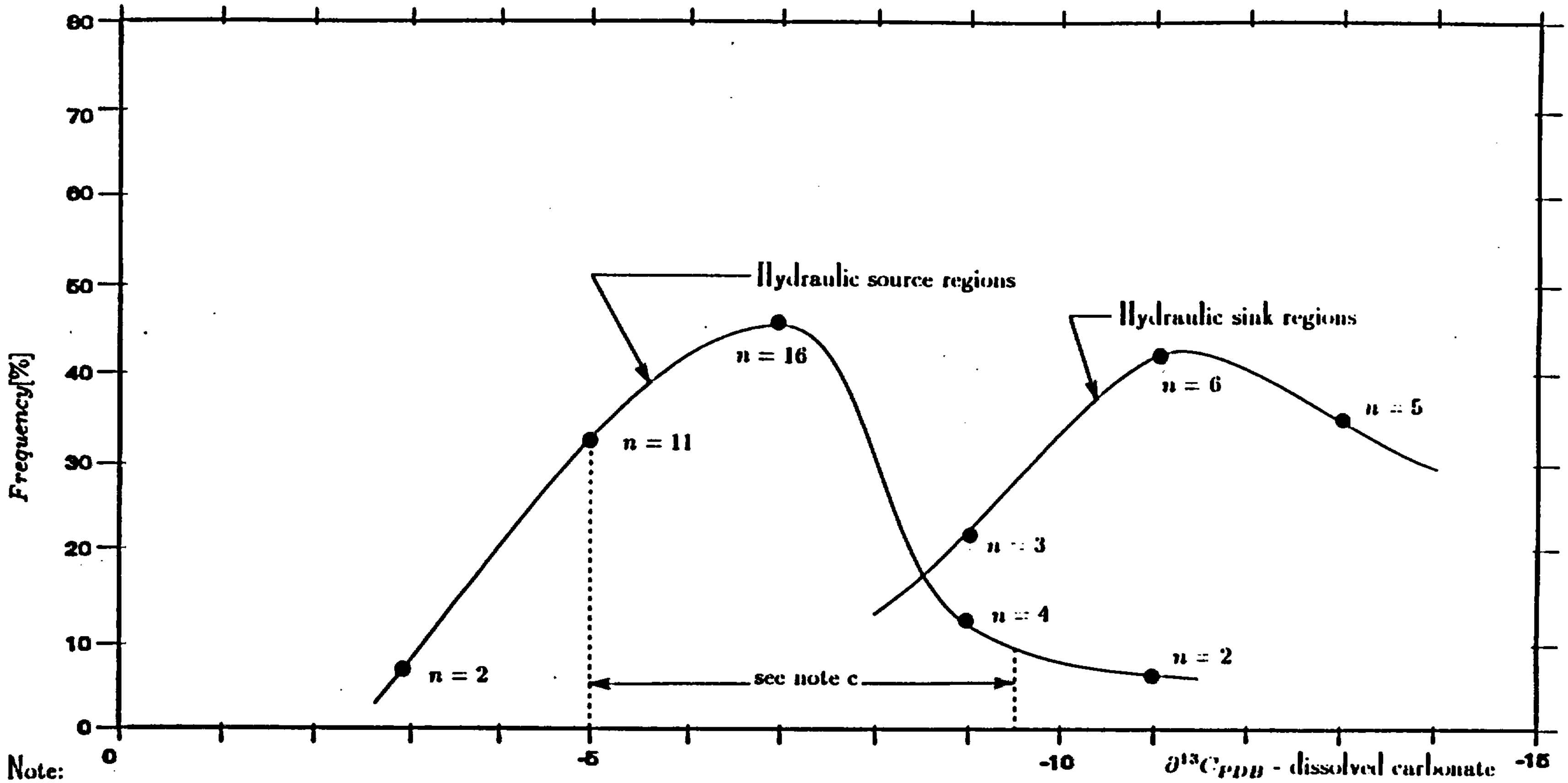
b) The results of monitoring of the Yucca Mountain borehole UZ63 revealed that, during a 30-month long monitoring period, this borehole has exhaled ~1150 kg of carbon, Thorstenson et al., 1989; and

c) Both of the above observations suggest that, at Yucca Mountain, the net CO_2 flux is from hydrosphere → vadose zone → 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:

a) Phrase hydraulic "source" region is used to denote local areas that; i) exhibit positive values of the $d\phi/dz$ gradient – ϕ is hydraulic potential; ii) exhibit high values of the dt/dz gradient - $dt/dz \sim$ 35-55°Celsius per 1km of depth; and iii) contain fluids having relatively high concentrations of $CL^{-}+SO_4^{--}$ anions;

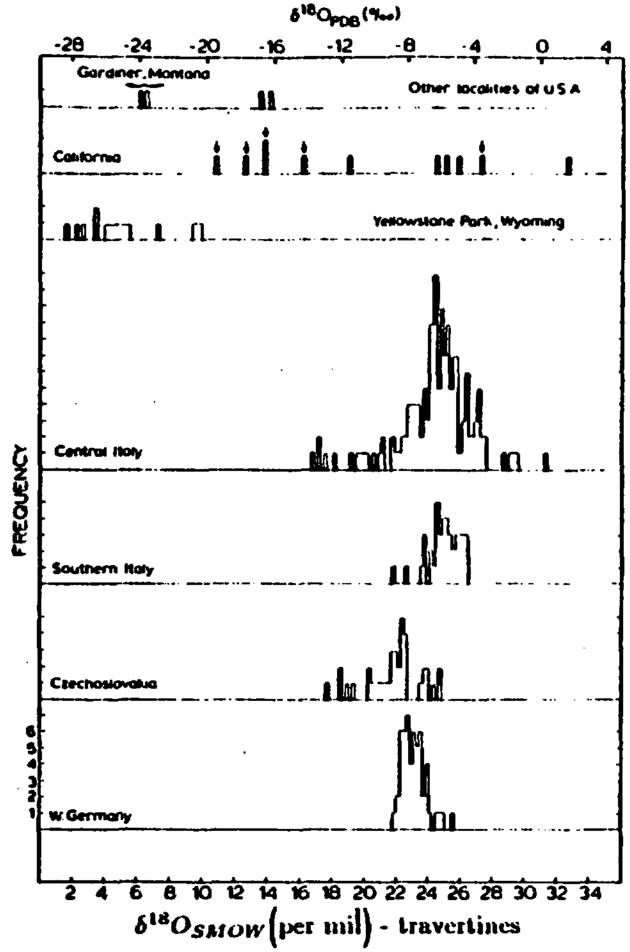
b) At precipitation temperatures ~50° Celsius, value of the isotopic fractionation factor $10^3 ln\alpha_{CaCO_3 - HCO_3}$ is ~ 2 per mil $_{PDB}$; and

c) The –3.0 to –7.5 per mil $_{PDB}$ 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 $_{PDB}$.

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 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:

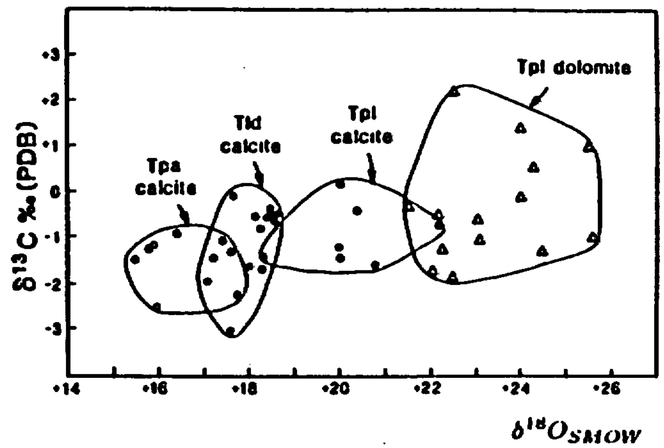
a) A value of the $\delta^{18}O$ ratio, from a sample of a particular travertine deposit, reflects three main factors, namely: i) oxygen 18 content for the parent fluid; ii) precipitation temperature; and iii) conditions of precipitation and the resulting value of the isotopic fractionation factor; and

b) 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 $\delta^{13}C$ vs. lithology gradient and the $\delta^{18}O$ 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;

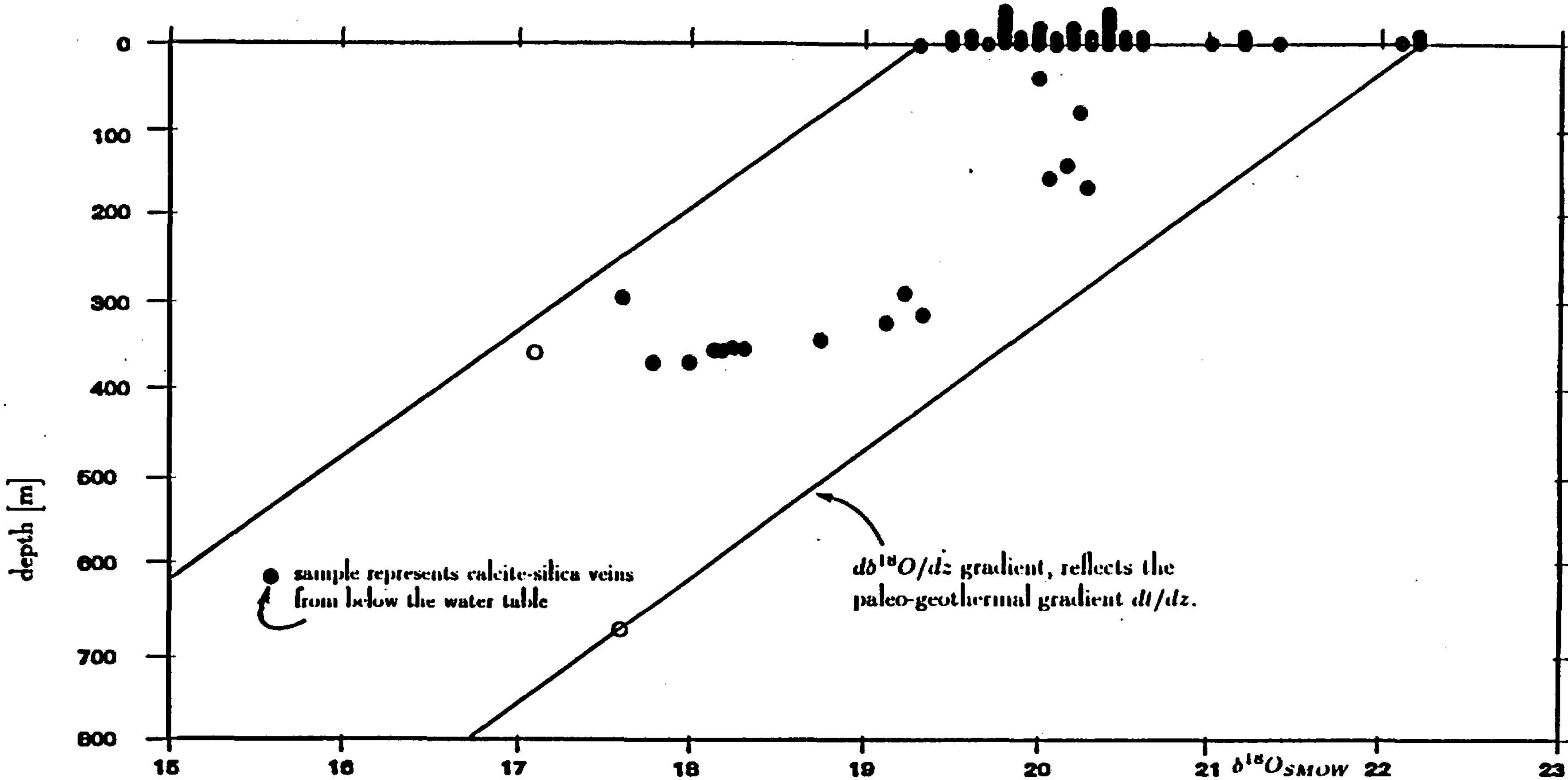
d) At the Ash Meadows topographic surface, the ambient temperatures are likely to be 5 to 10° Celsius higher than the corresponding temperatures at Yucca Mountain - the resulting differences in the equilibrium fractionation factor range from 1 to 2 per mil $_{SMOW}$, the Yucca Mountain values being larger; and

e) Differences in values of the $\delta^{13}C$ 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 Hay et al., (1986).

Figure 8



Note:

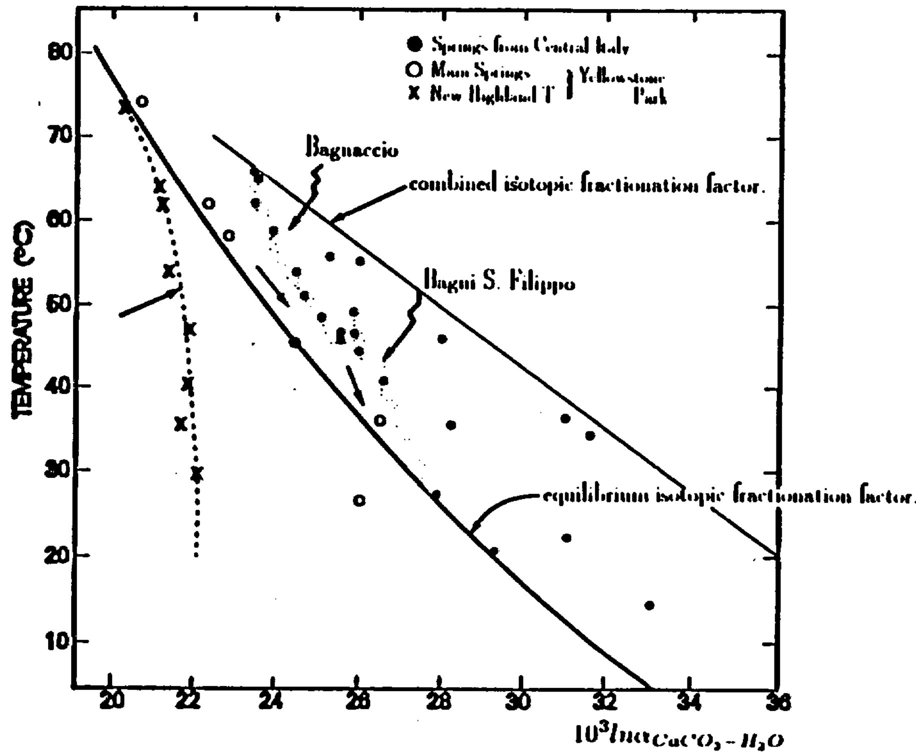
a) The entire observed Yucca Mountain range of the $\delta^{18}O$ ratios for carbonate deposits, from 15.4 to 22.0 per mil $_{SMOW}$, is very similar to that observed for the Ash Meadows Basin deposits, Figure 8; and

b) 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 percipitation 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:

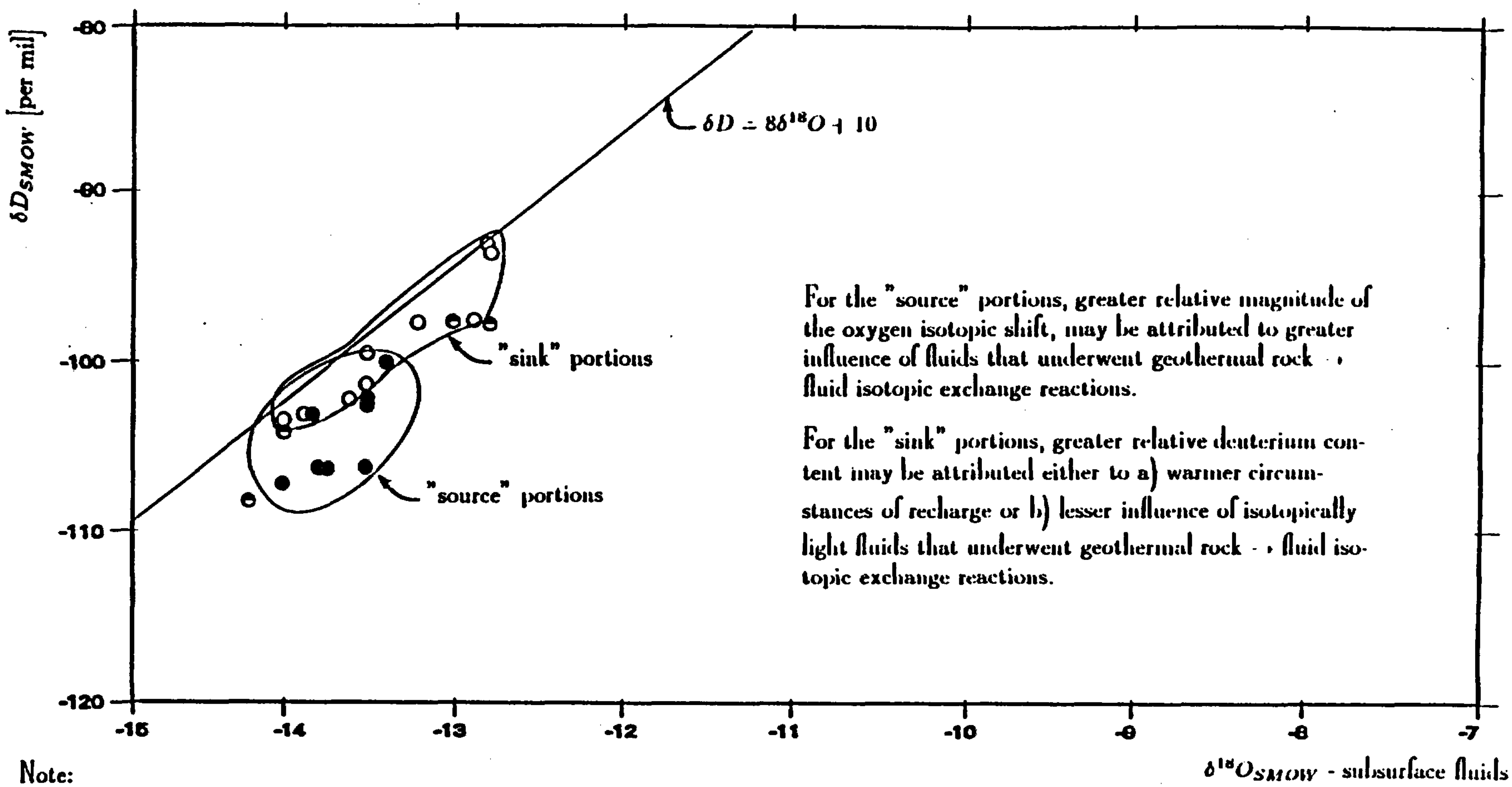
a) During natural depositions of travertines, the conditions of equilibrium isotopic fractionation are seldom attained, mainly as a consequence of the kinetic or non-equilibrium processes;

b) Depending upon conditions of precipitation, the combined or actual isotopic fractionation factor ($10^3 ln\alpha_{CaCO_3,\ H_2O}$) may be both larger and smaller than the equilibrium fractionation factor; and

c) At a precipitation temperature of t ~20° Celsius, 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:

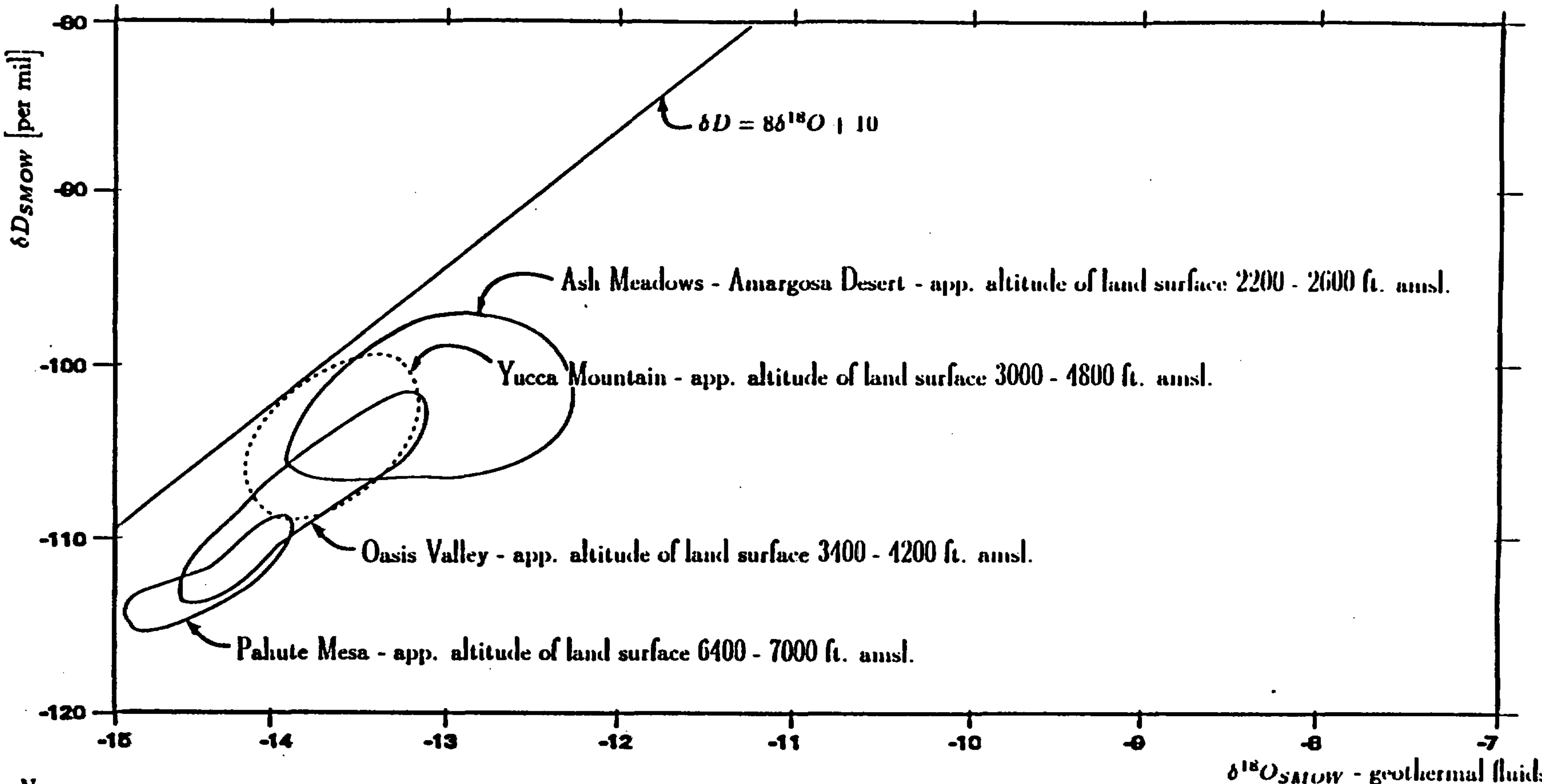
a) The observed range of values for the $\delta^{18}O$ ratio, from samples of the contemporary Yucca Mountain subsurface fluids, is from -14.2 to -12.7 per mil $_{SMOW}$; samples are bulk water samples pumped out of large segments of exploratory wells - sampling "smoothing" may be involved, and the observed range may be smaller than the actually present one; and

b) 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:

a) 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

b) 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.

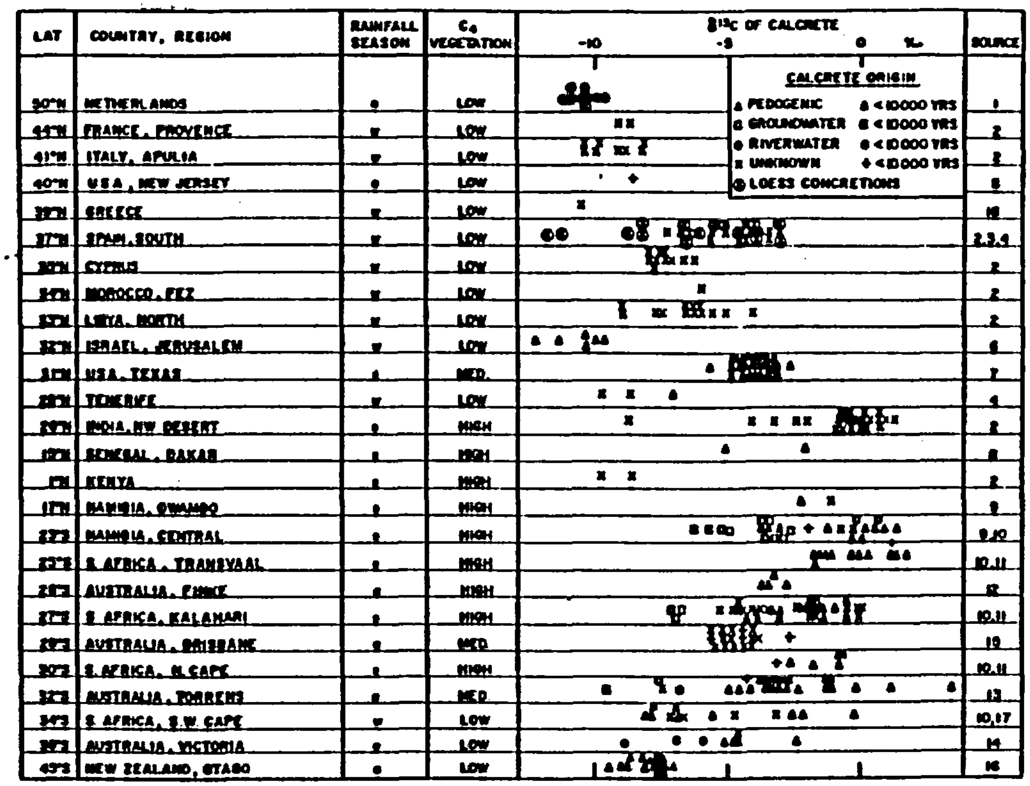
Oxygen isotopic file - isotopic character of oxygen contained in the contemporary Nevada Test Site geothermal fluids. Data from Claassen, 1985; Benson and McKinley, 1985; and White and Chuma, 1987.

Figure 12.

- The above performed comparitive analyses indicate that the $\delta^{18}O$ vs. $\delta^{13}C$ 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 (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 calcite-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 calcite-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).



Note:

a) The worldwide data for calcretes are from Talma and Netterberg (1983);

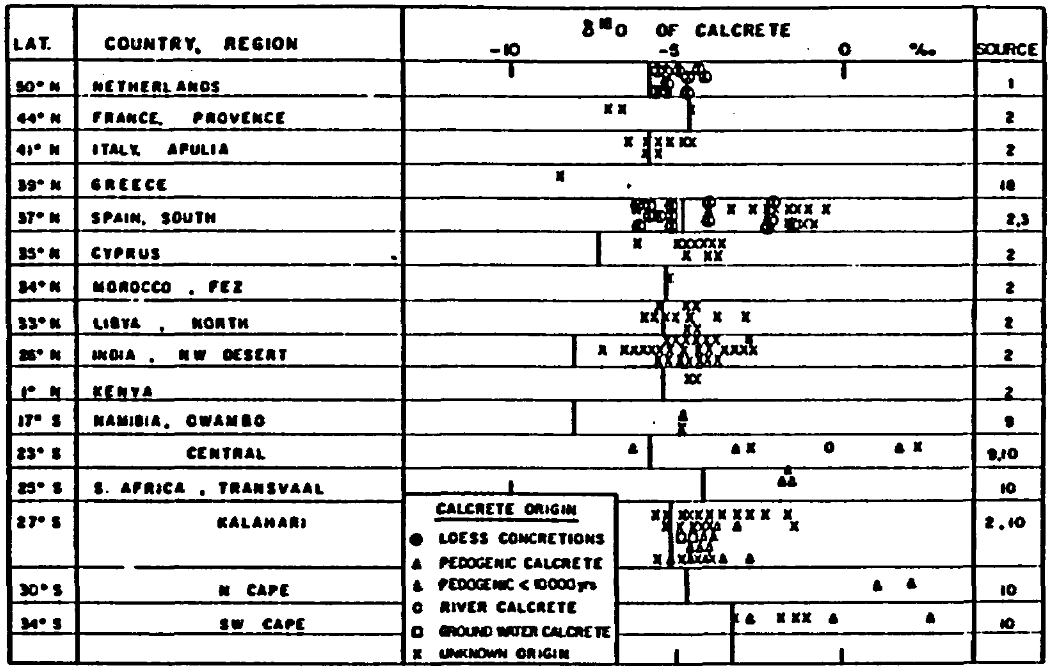
b) For the Nevada Test Site cobble encrustations, the $\delta^{13}C$ ranges from +4.1 to −9.11 *PDB*, Quade et al., (1989);

c) This range overlaps with: i) the corresponding range from the worldwide "pedogenic" (?) calcretes, which is from +4 to 12 per mil *PDB*; ii) the corresponding range from the worldwide groundwater calcretes, which is from +1 to −10 per mil *PDB*; iii) the corresponding range from the worldwide travertines, which is from +12 to −24 per mil *PDB*, Figure 3; iv) the corresponding range from carbonate gangue from gold-bearing hydrothermal ore deposits, which is from +10 to −10 per mil *PDB*, Figure 4; v) the corresponding range from the Ash Meadows dèep seated spring deposits, which is from +2.5 to −3.0 per mil *PDB*, Figure 8; and vi) the corresponding range from the Yucca Mountain calcite-silica deposits, which is from −3.0 to −8.5 per mil *PDB*, Figures 1 and 5; and

d) 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".

Figure 13a



Note:

a) The worldwide data for calcretes are from Talma and Netterberg (1983);

b) For the Nevada Test Site cobble encrustations, the $\delta^{18}O$ ranges from −0.4 to - 12.6 $_{PDB}$, Quade et al., (1989);

c) This range overlaps with: i) the corresponding range from the worldwide "pedogenic" (?) calcretes, which is from +4 to - 8.5 per mil $_{PDB}$; ii) the corresponding range from the worldwide groundwater calcretes (including "riverwater" calcretes), which is from 0.5 to −5 per mil $_{PDB}$; iii) the corresponding range from the worldwide travertines, which is from +1.5 to −28 per mil $_{PDB}$, Figure 7; iv) the corresponding range from the Ash Meadows deep seated spring deposits, which is from ~ −4.5 to ~ −15.5 per mil $_{PDB}$, Figure 8; and v) the corresponding range from the Yucca Mountain calcite- silica deposits, which is from ~ −8.5 to ~ −16.0 per mil $_{PDB}$, Figure 9; and

d) 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".

Figure 131.

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

Figures 14 through 29

- 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.

- 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.

- For the Yucca Mountain surficial deposits, reliable interpretations of the exact ($\pm$ 5° Celsius) precipitation temperatures can not be made based solely on the oxygen −18 content of these deposits.

- 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 $\pm$ 1.0 per mil $_{SMOW}$; ii) a value for the combined isotopic fractionation factor may not be reliably estimated with the required precision of ~ $\pm$ 1.2 per mil $_{SMOW}$; and iii) assumption that, during a time span represented by these deposits [which is from 26 $\pm$ 2 x 10^3 to more than 4 x 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.

- 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.

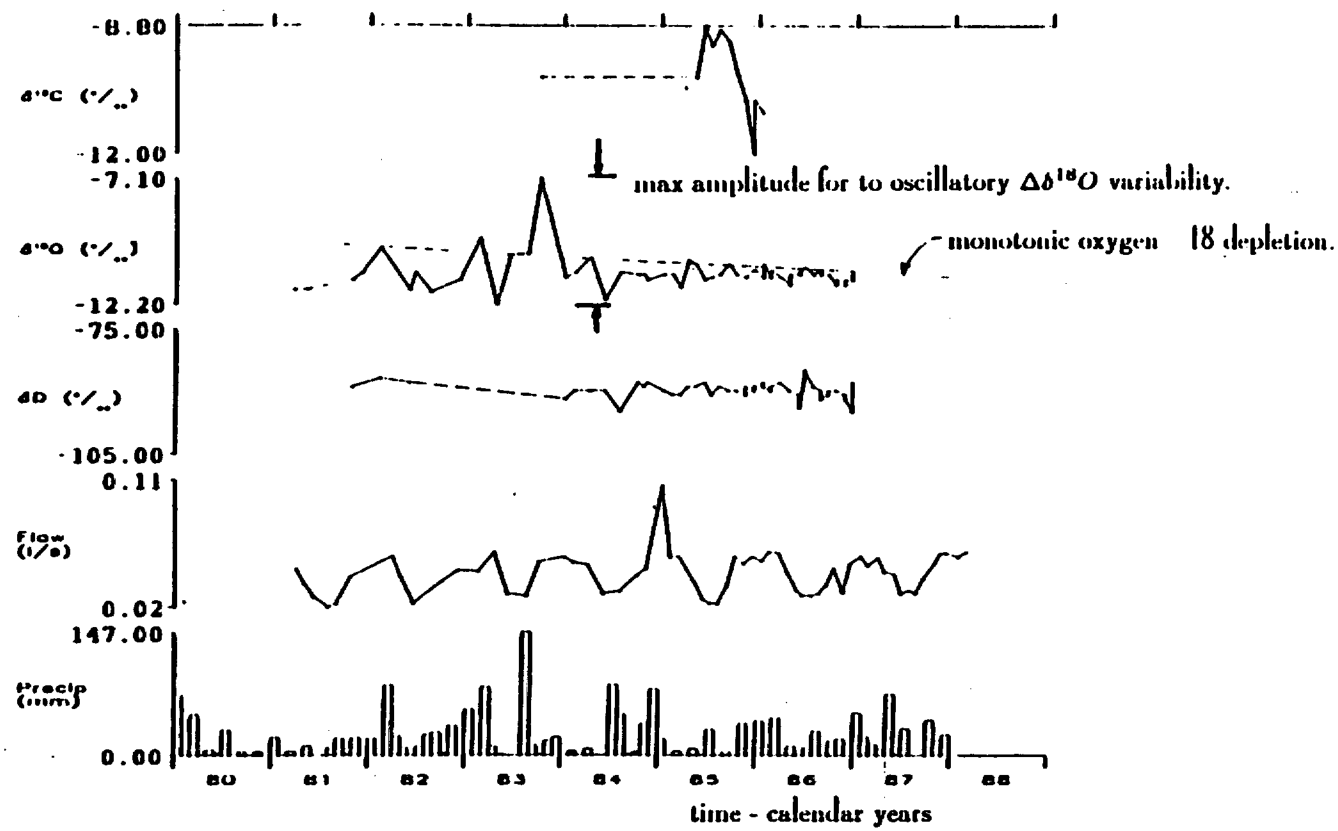
- Examinations of these figures reveal that, at the Nevada Test Site, the oxygen 18 content of the subsurface fluids may not be assumed as time invariant. Both the monotonic variabilities and the oscillatory variabilities are evident. The oscillatory variabilities involve both the low frequency fluctuations and the high frequency fluctuations.

- Relative to the contemporary subsurface fluids, the paleo-subsurface fluids could have been isotopically "heavier" by at least $\Delta\delta^{18}O \sim 5$ per mil $_{SMOW}$.

- 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 $\Delta\delta^{18}O \sim 5$ per mil $_{SMOW}$. The resulting potential error in estimating the specimen precipitation temperature, and using the equilibrium isotopic fractionation curve, is equal to $\Delta t \sim 22^{\circ}$ Celsius, Figure 10.

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

Figure 15.



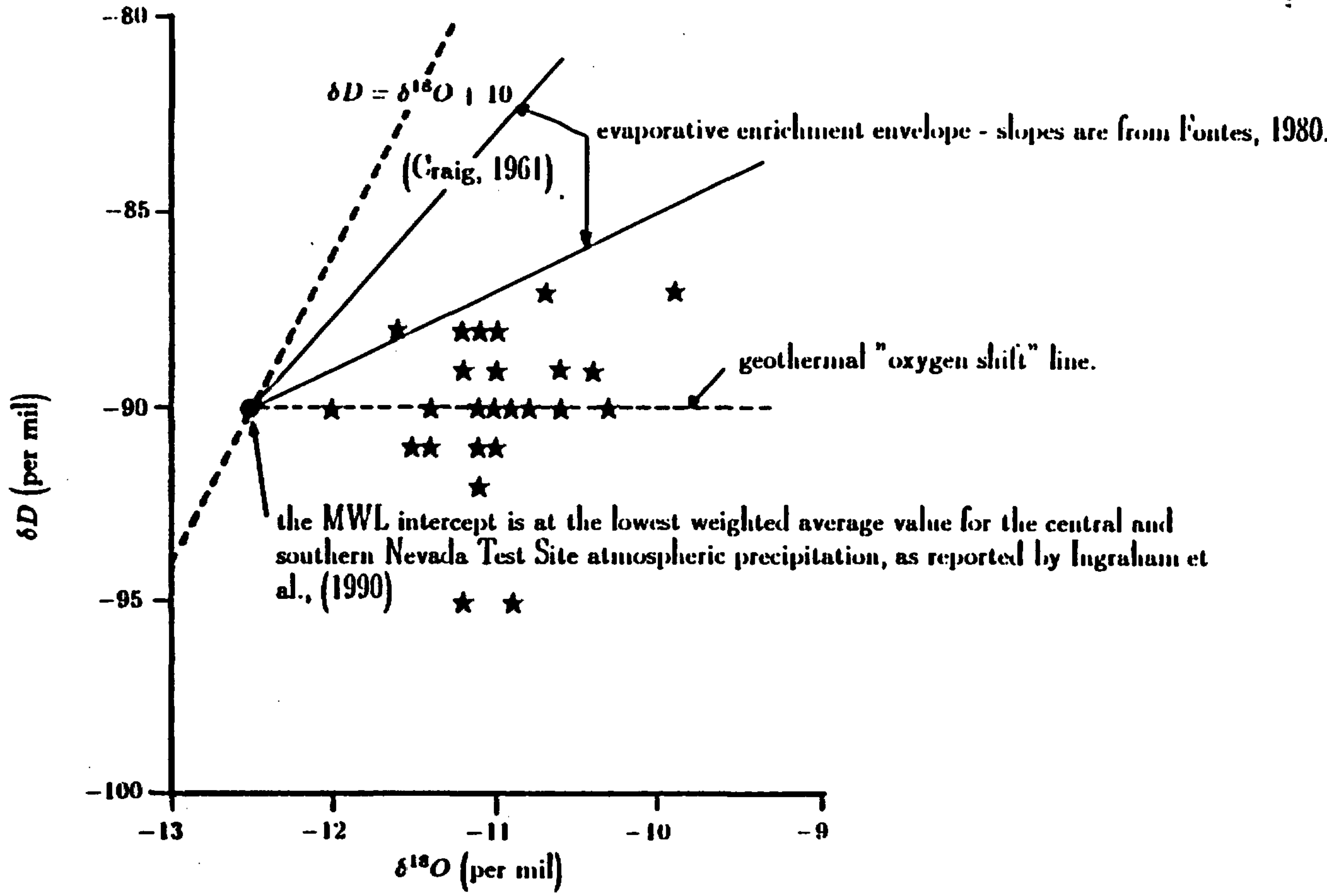
Note:

a) 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;

b) The observed monotonic oxygen −18 depletion is $\Delta\delta^{18}O \sim 1.2$ per mil $_{SMOW}$ in 6 years; and

c) The observed $\delta^{18}O$ oscillatory variability has a maximum amplitude of $\Delta\delta^{18}O \sim 5$ per mil $_{SMOW}$.

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



Note:

a) 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 $CaCO_3$;

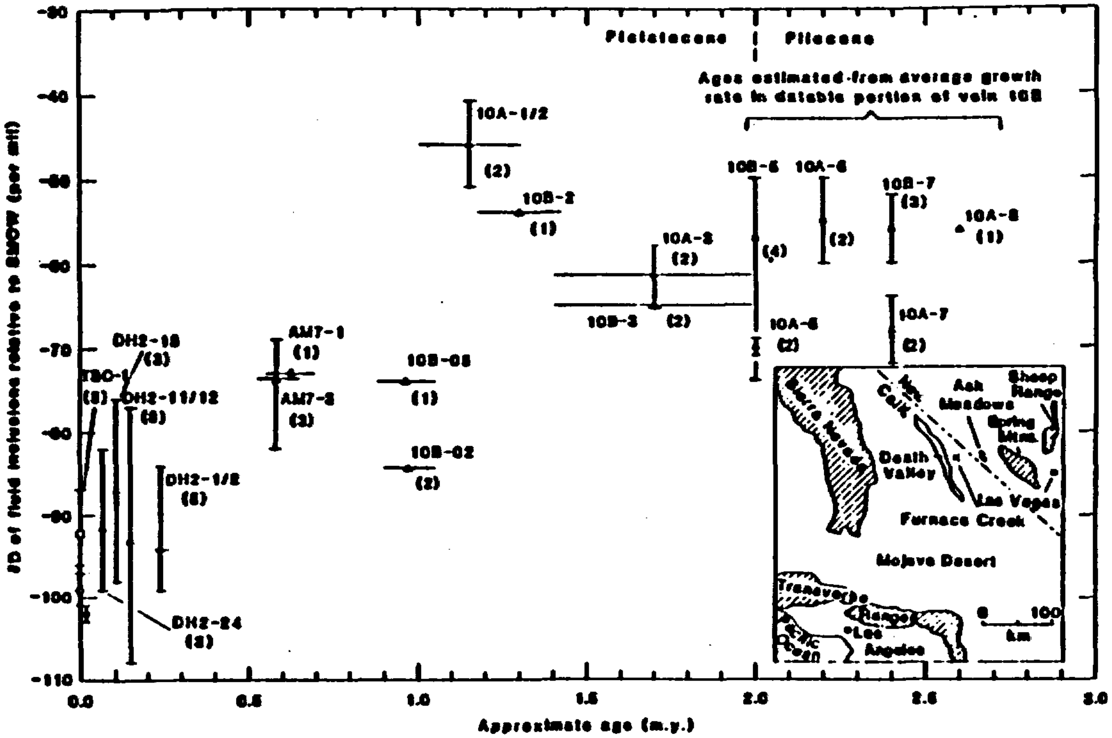
b) Based on a variety of data, Szymanski (1989) has postulated that both the Cane Spring discharge and the underlying perched bodies of groundwater are deep-seated and together express a contemporary hydraulic mound, presence of which is the result of past hydro-tectonic activity;

c) The contemporary Cane Spring $CaCO_3$ deposits may be regarded as a modern analog for the Yucca Mountain calcite-silica deposits; and

d) 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 calcite-silica deposits.

Figure 16a.



Note:

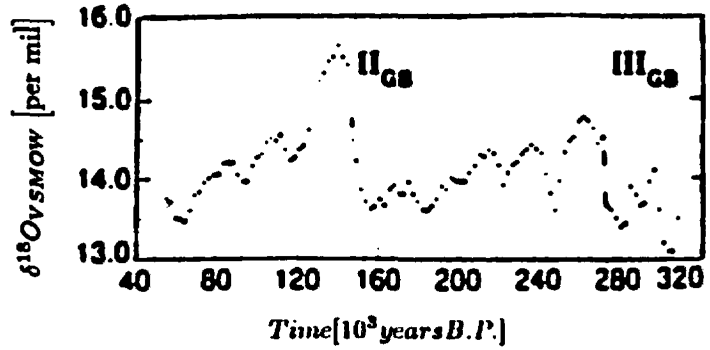
a) During the last 2.5 x 10^6 years, the deuterium content in the parent fluids for the local calcite veins have changed;

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 x 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:

a) Each dot represents a $\delta^{18}O$ analyzed sample;

b) 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

c) During the time span from 4 x 10^4 to about 3.2 x 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).

Figure 18

- 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.

- The kinetic, or non-equilibrium, isotopic fractionation effects may be as large as $\Delta 10^3 ln\alpha_{CaCO_3 - H_2O} \sim 6$ 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 $\Delta t \sim 30°$ Celsius.

- 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.

Figure 19.

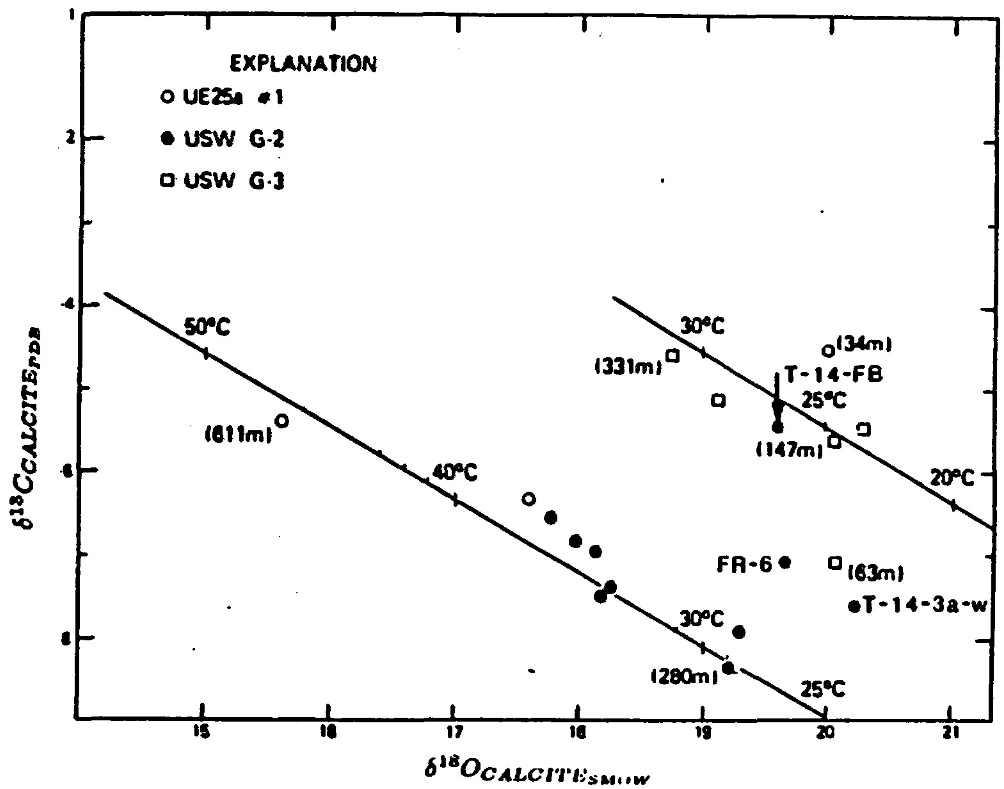
- Uncertainties that are associated with interpretations of the precipitation temperatures for the Yucca Mountain deposits, based solely on the oxygen −18 content of these deposits, may be best illustrated by pointing out the results of such interpretations as performed by Szabo and Kyser (1985) and Szabo and Kyser (1990).

- 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 ~25° 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.

- 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 ~14 ° 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:

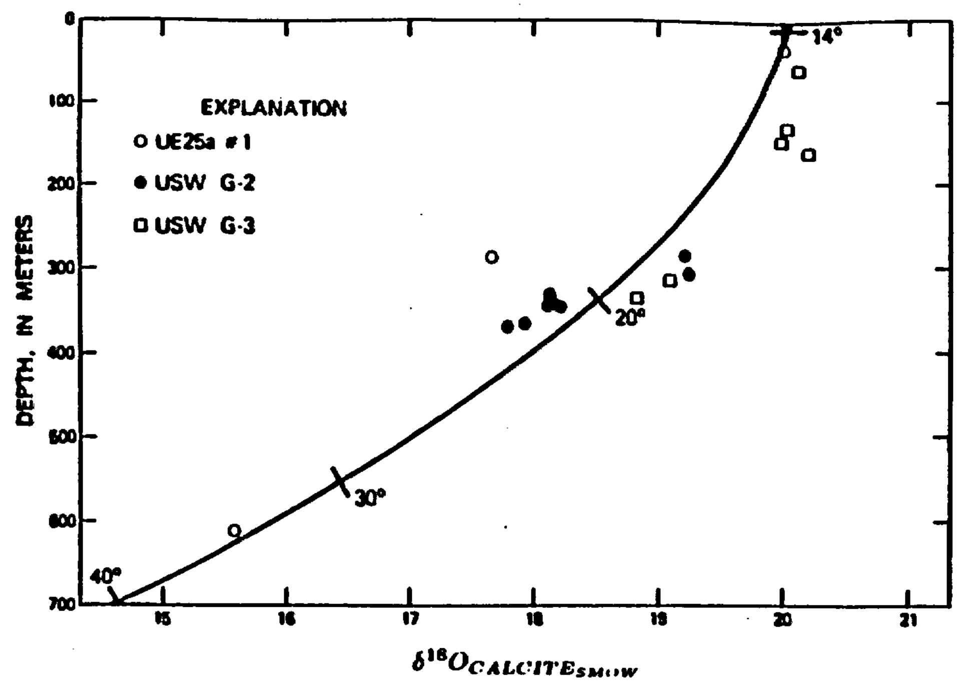
a) Samples yielding values of the $\delta^{18}O$ ratio ~20 per mil $_{SMOW}$ were interpreted to have been formed at a temperature ~25° Celsius;

b) The Yucca Mountain surficial deposits carry values of the $\delta^{18}O$ ratio ranging from 19.2 to 22.0 per mil $_{SMOW}$, Figure 1; and

c) The corresponding range of the precipitation temperatures, for the Yucca Mountain surficial deposits, is from 15 to ~ 28° Celsius - only considerations of the *per ascensum* mechanism are permissible.

1985 interpretation of the precipitation temperatures for the Yucca Mountain vadose zone veins. From Szabo and Kyser, 1985.

Figure 21



Note:

a) Both the 1985 interpretation and the 1990 interpretation are based on the same oxygen - 18 data;

b) Samples yielding values of the $\delta^{18}O$ ratio ~20 per mil *SMOW* were interpreted to have been formed at a temperature of ~14° Celsius; and

c) The corresponding range of the precipitation temperatures, for the Yucca Mountain surficial deposits, is from 5 to about ~ 18° 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.

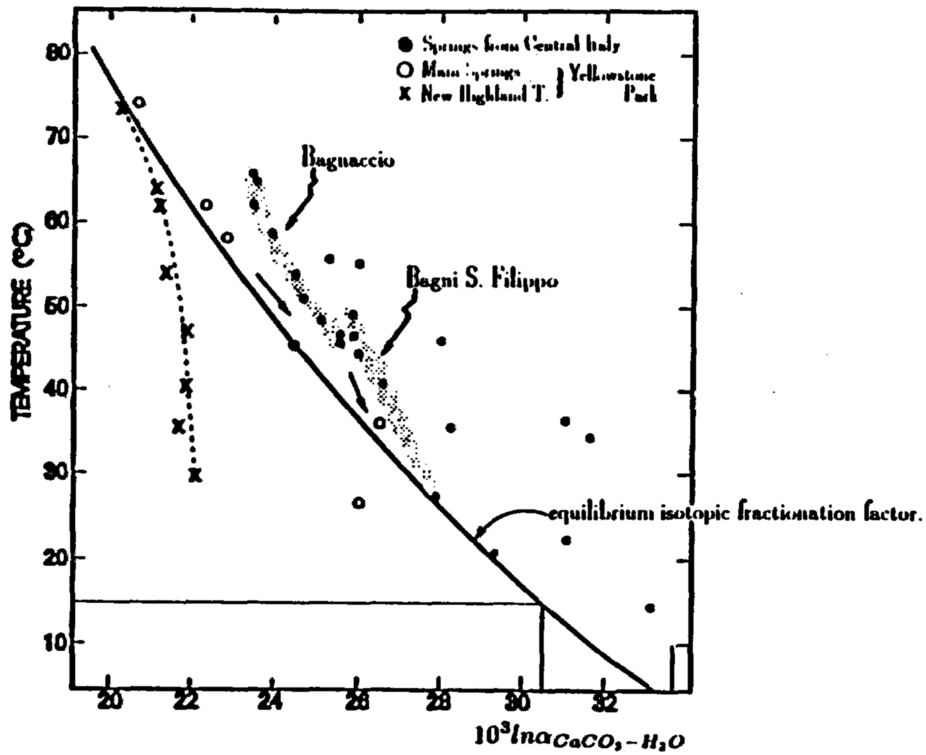
- Figures 24 through 28 present the results of five conceivable reconstructions of the precipitation temperatures for the Yucca Mountain surficial deposits. These reconstructions are based on the oxygen – 18 contents from samples of these deposits, as reported by Whelan and Stuckless (1990). As shown 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 $_{SMOW}$.

- Each reconstruction involves two steps. First, value of the combined isotopic fractionation factor ($10^3 ln\alpha_{CaCO_3-H_2O}$ $2.78 \cdot 10^6/T^2 - 2.82 + \beta$) 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.

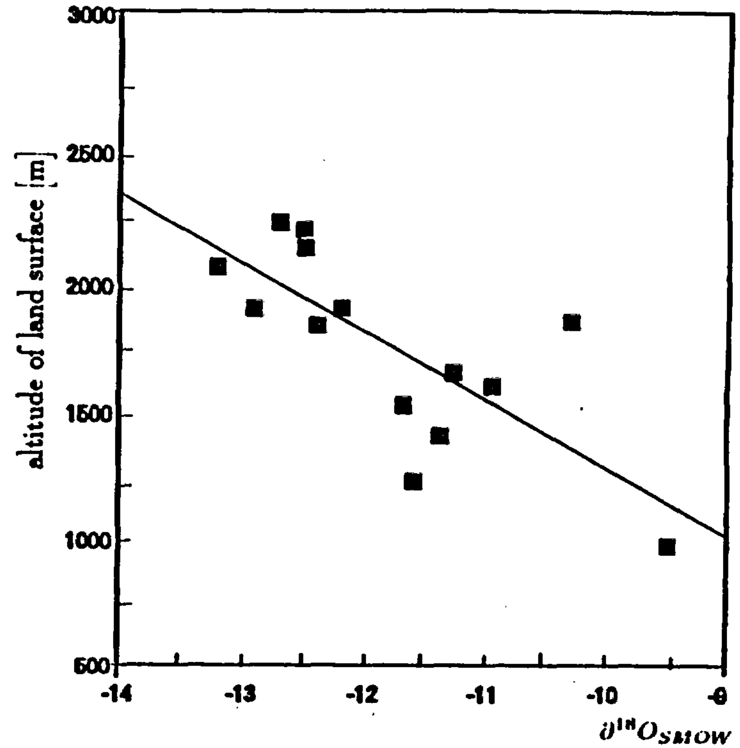
- For the parent fluids, three different isotopic compositions were considered. These are: i) reconstruction A - made assuming that the parent fluids have carried the oxygen –18 contents similar to those of the local contemporary atmosphere precipitation; ii) reconstructions B and C - made assuming that 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.

- 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:

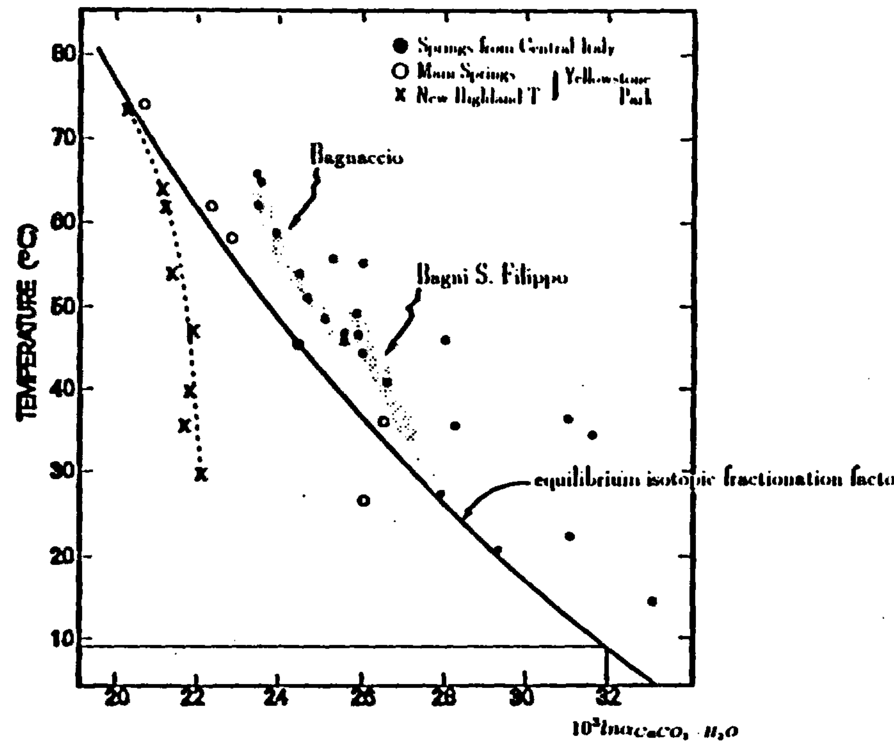
a) In performing this reconstruction, it has been assumed that: i) the parent fluids, for the Yucca Mountain surficial fluids, were isotopically identical to the contemporary atmospheric precipitation (at altitudes ranging from 1250 to 1400m, $\delta^{18}O$ values range from - 11.7 to - 11.3 per mil $_{SMOW}$); and ii) the non-equilibrium isotopic fractionation effects were absent, such that the combined isotopic fractionation factor $10^3 ln\alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^6/T^2 - 2.82$;

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 30.5 and 33.7, respectively;

c) The corresponding range for the precipitation temperatures is from 0 to ~ 14° Celsius; and

d) 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.



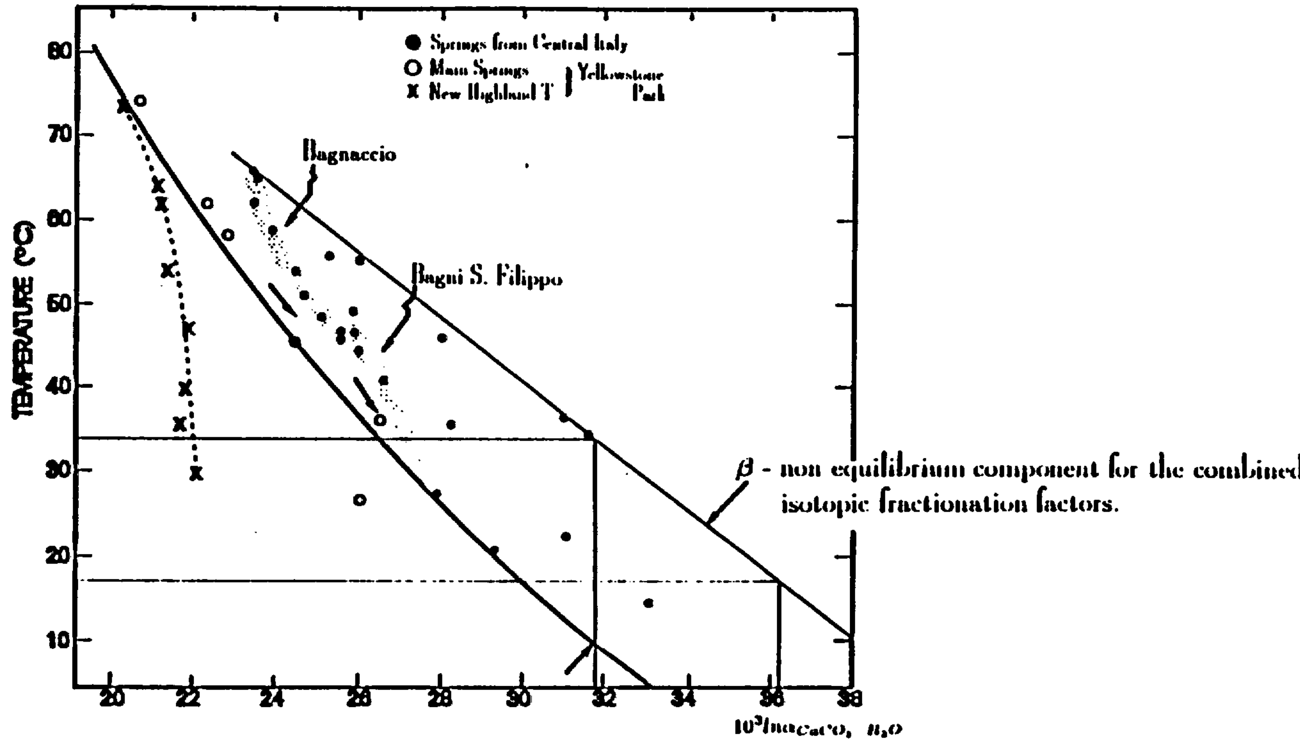
Note:

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 Yucca Mountain surficial deposits, based on the oxygen 18 contents from samples of these deposits.



Note:

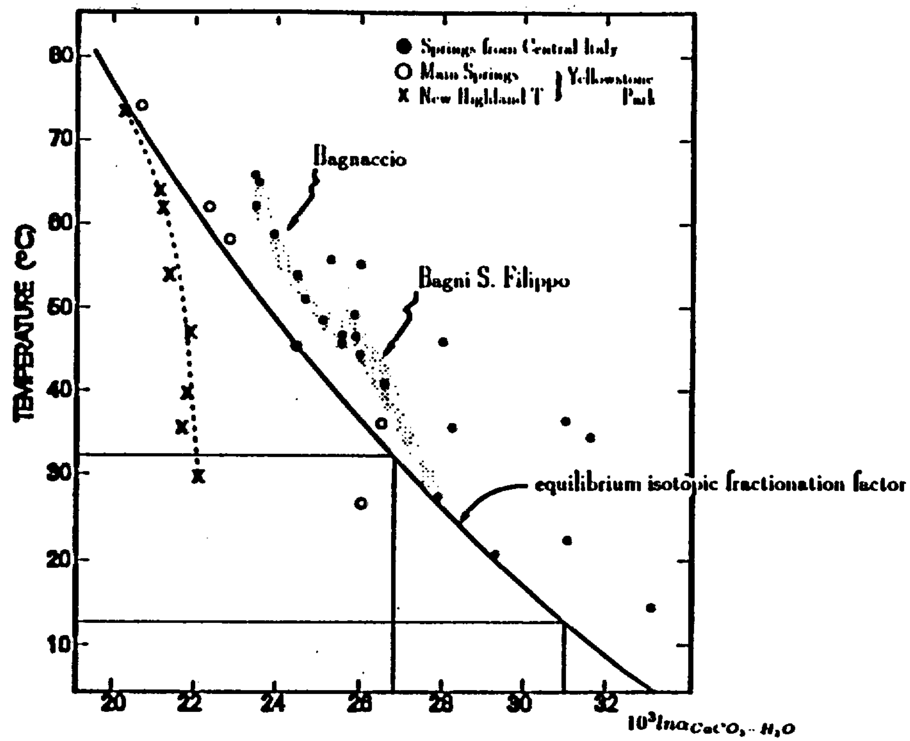
a) In performing this reconstruction, it has been assumed that: i) during a time span represented by the surficial deposits, the isotopic compositions of the parent fluids were the same as those observed below the water table at the present time ($\delta^{18}O$ values ranging from -14.2 to -12.7 per mil *SMOW*, Figure 11); ii) the combined isotopic fractionation factor was larger that 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 it's oxygen from both H_2O and CO_2;

b) 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

c) 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:

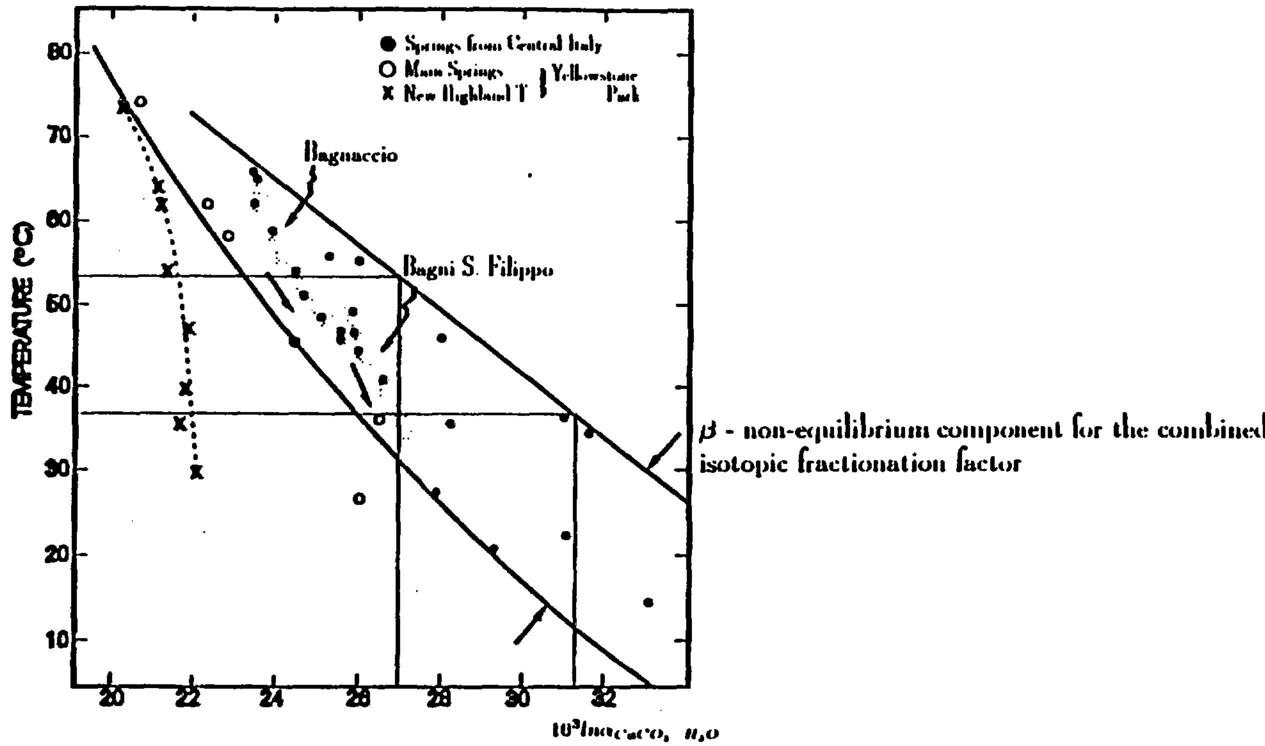
a) In performing this reconstruction, it has been assumed that: i) the parent fluids, for the Yucca Mountain surficial deposits, were isotopically heavier than the contemporary Yucca Mountain subsurface fluids, $\Delta\delta^{18}O \sim 5$ per mil $_{SMOW}$ (-9.2 to −7.7 per mil $_{SMOW}$, Figure 18); 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 ~ 32° 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:

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); 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 it's oxygen from both H_2O and CO_2;

b) 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 26.9 and 31.2 per mil, respectively; and

c) 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.

- 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.

- 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.

- 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.

Figure 29.

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

Figures 30 through 44

- 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.

- 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° Celsius per 1km of depth, would constitute a fairly definitive demonstration that: i) episodically, the Yucca Mountain vadose zone was being innudated 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.

- 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.

- 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 $\Delta\delta^{18} \sim 3$ 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.

Figure 30.

- 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.

- 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.

- 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.

- 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

- 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.

- 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 ~ 14° Celsius; ii) spatially different samples were precipitated from parent fluids having the same oxygen −18 contents; iii) during the time span represented by the subsurface veins, from 26 ± 2 x 10^3 to more than 4 x 10^6 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.

- 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).

Figure 31.

- As shown in Figure 22, the reconstructed Yucca Mountain paleo-geothermal gradient exhibits a very conspicuous curvature. Down to a depth of ~ 400m, the depthward rate of the *in-situ* temperature increase is fairly low; value of the equivalent geothermal gradient is dt/dz ~ 17° 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 ~ 50° Celsius per 1km of depth.

- 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 ~ 1.7 $Wm^{-1}k^{-1}$, 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 monotomic - 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.

- 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 $\delta^{18}O$ 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 are of Yellowstone Park, Wyoming. In very rapidly moving fluids, the $CaCO_3$ nucleation occurrs 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.

Figure 32.

- 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?**

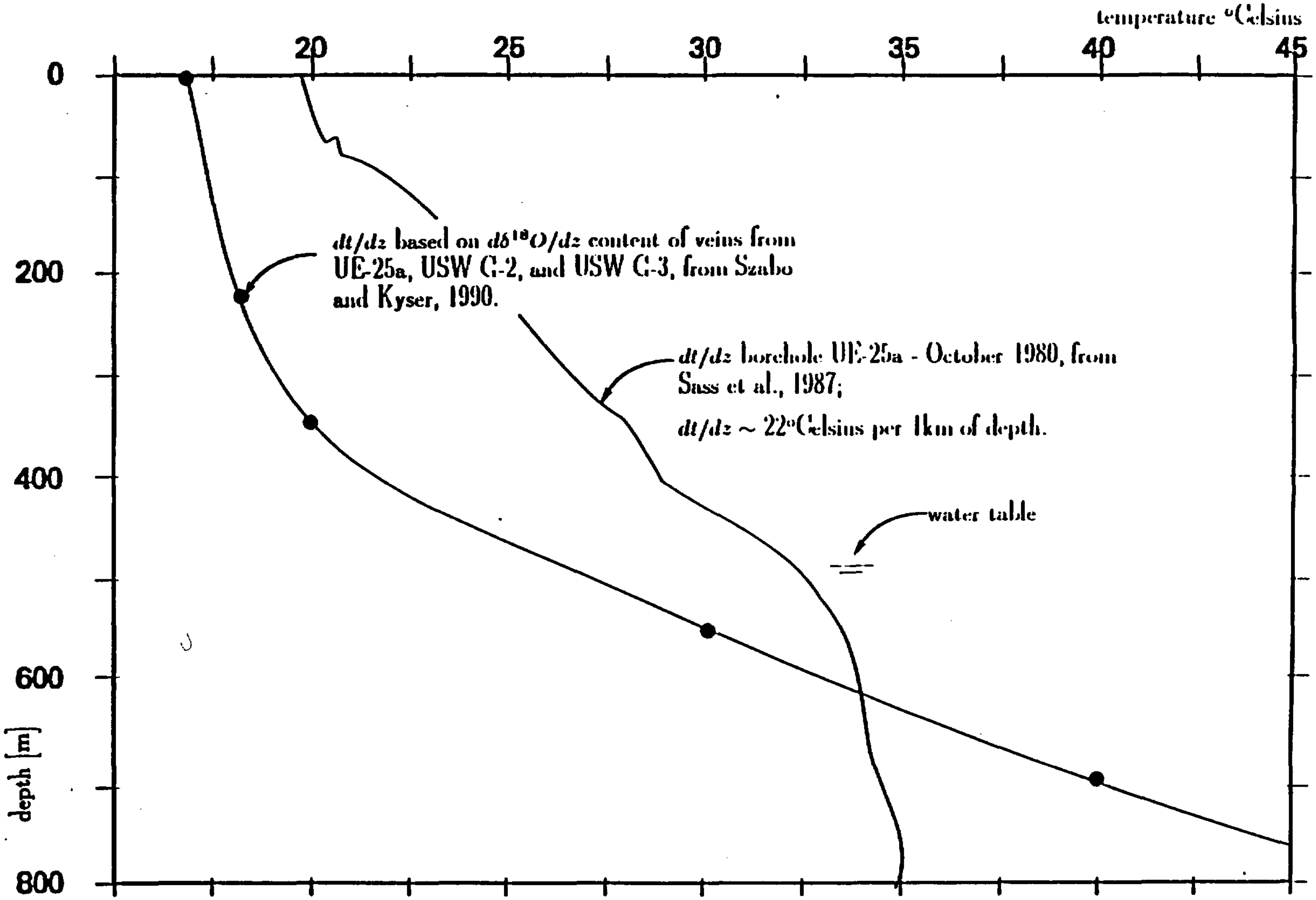
- Figures 34 through 37 present comparisons between the reconstructed paleo-geothermal gradient and the contemporary geothermal gradients, as recorded in the corresponding boreholes UE-25a, USW G-2, USW G-3, and USW G-4. Examination of these figures reveals that, in the lower parts of the vadose zone (~ 100 - 250 m above the contemporary water table), the reconstructed paleo-geothermal gradient is a factor of two higher than the contemporary geothermal gradient.

- 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.**

- Because both the $\delta^{13}C$ ratios and the $\delta^{18}O$ 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.

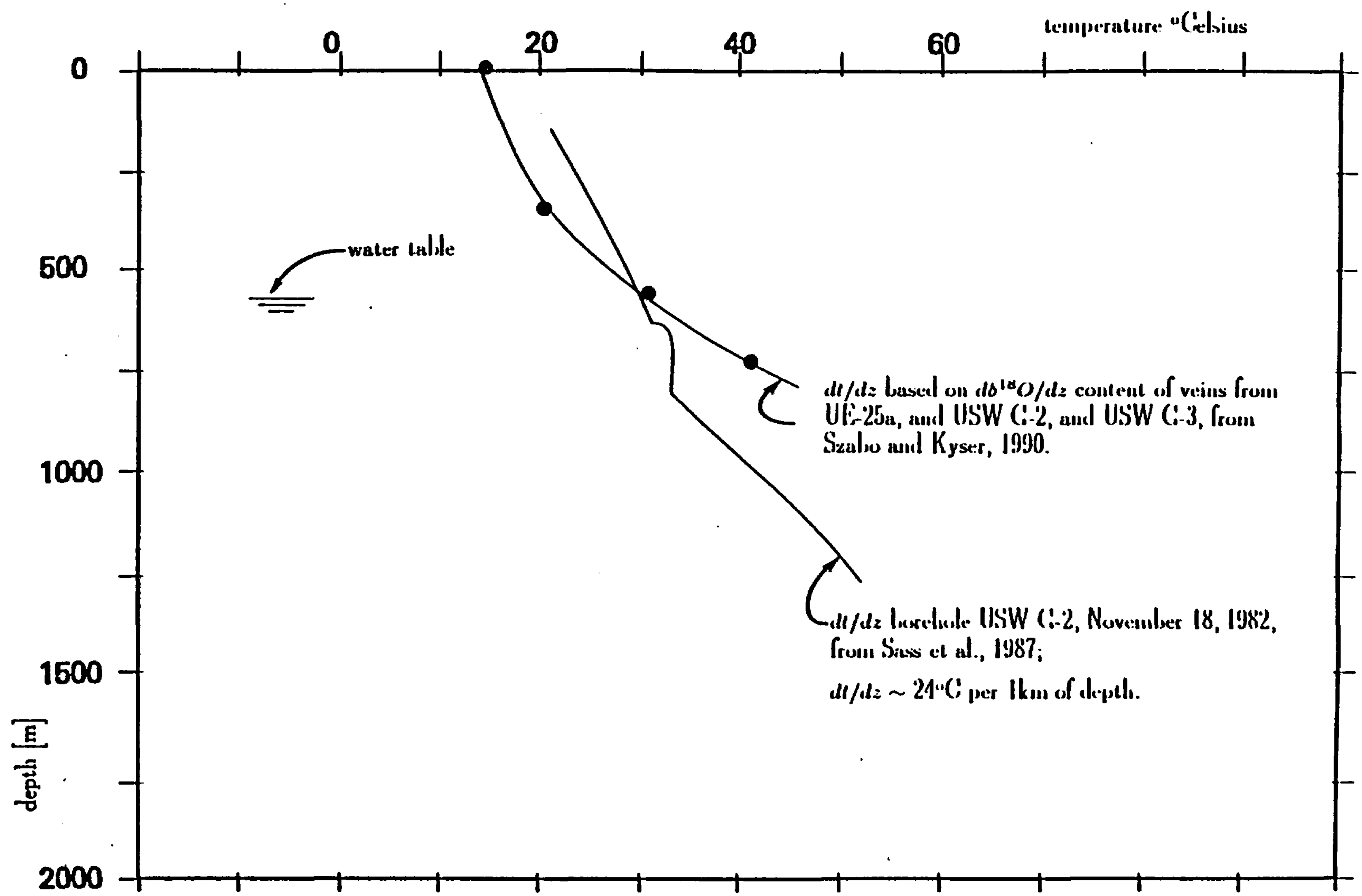
Implications resulting from the paleo-geothermal reconstructions made by Szabo and Kyser, 1990.

Figure 33.



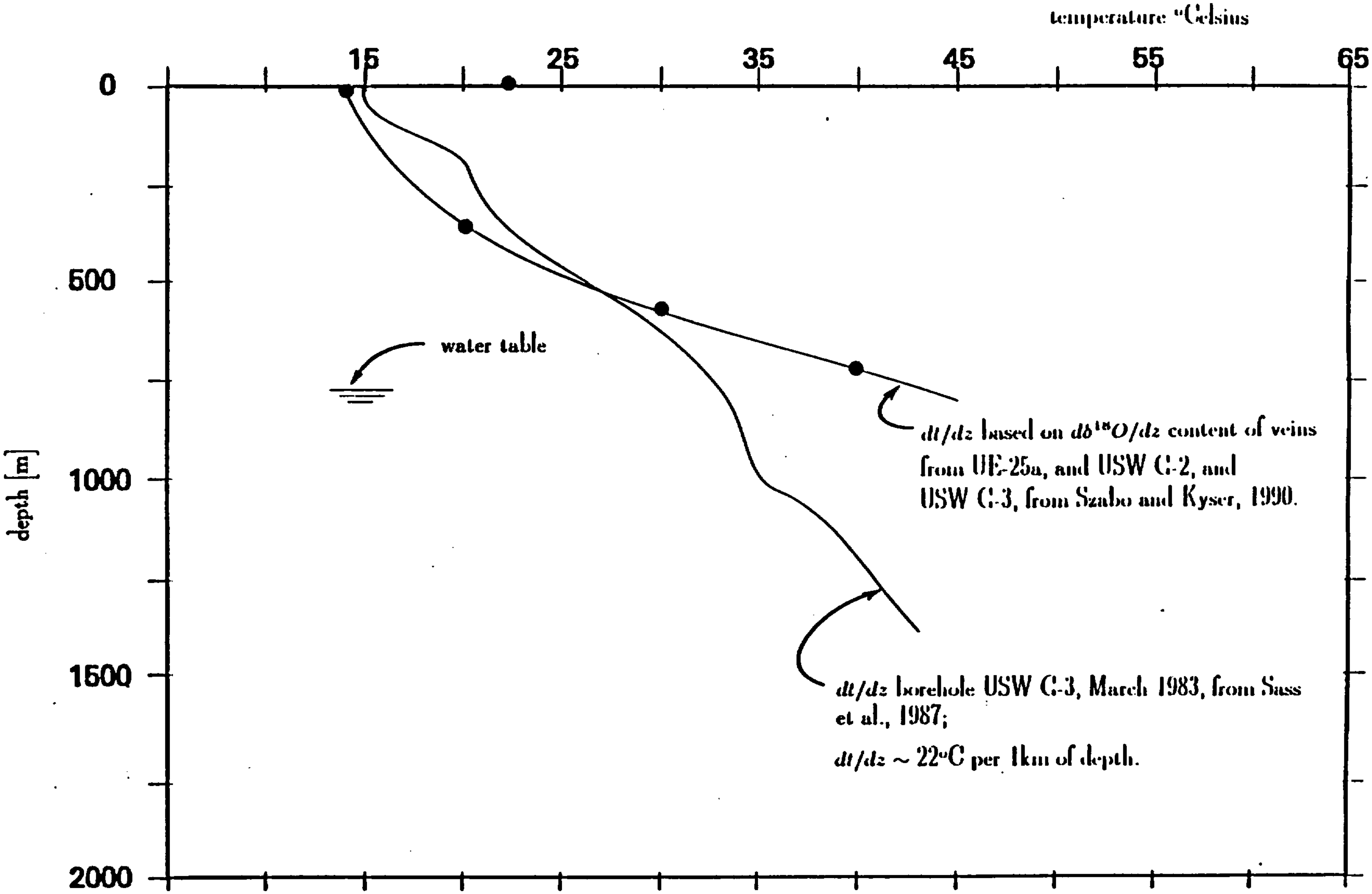
Comparison between the paleo-geothermal gradient and the contemporary geothermal gradient. Well UE-25a.

Figure 34.



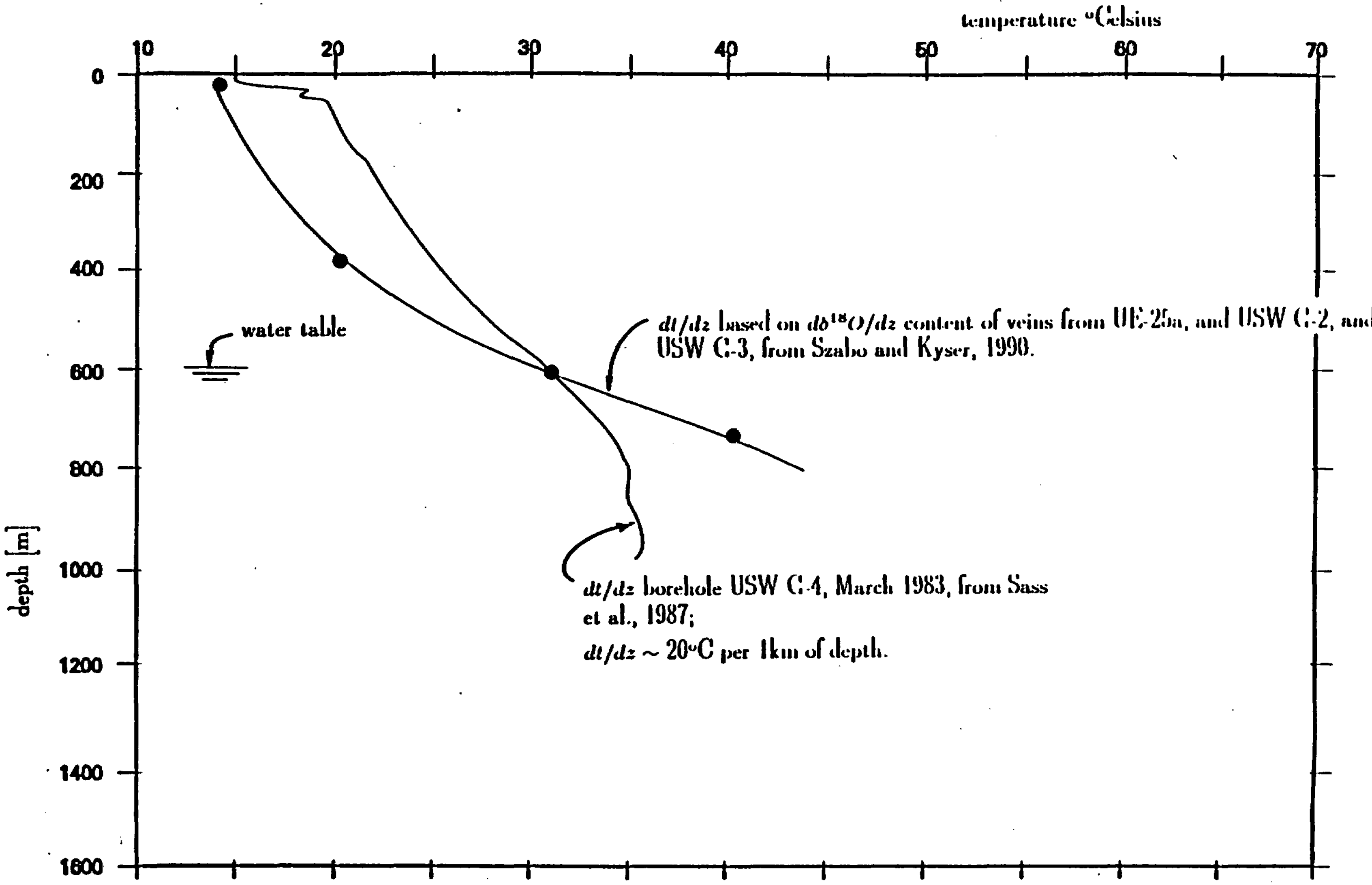
Comparison between the paleo-geothermal gradient and the contemporary geothermal gradient. Well USW G-2.

Figure 35.



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

Figure 36.



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

Figure 37.

- Figures 39 through 41 present alternate interpretations 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. These interpretations were made using all of the available oxygen 18 data, as reported by Szabo and Kyser (1990) and Stuckless and Whelan (1990).

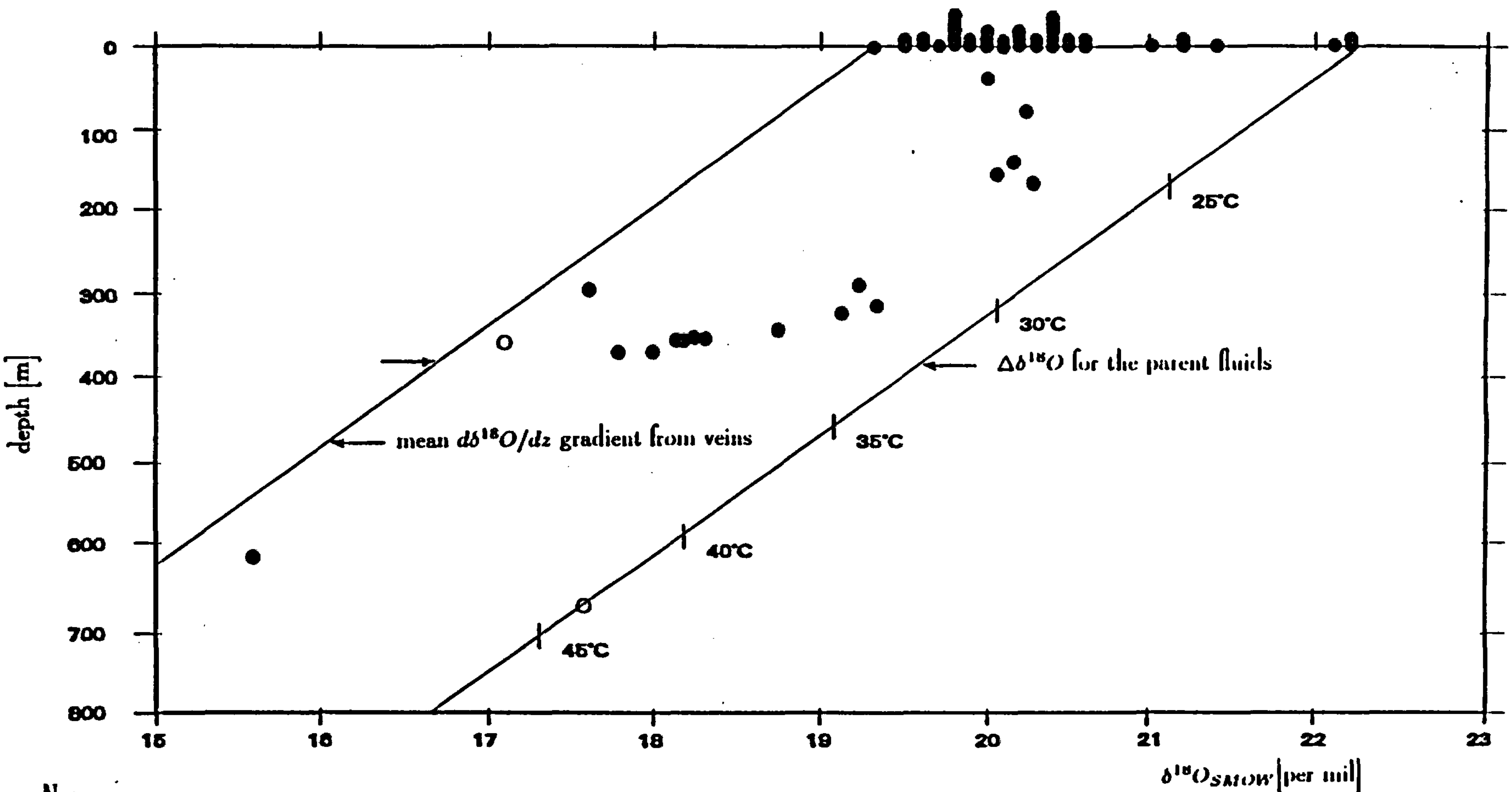
- A common assumption employed in performing all three interpretations is that, for the surficial calcite-silica deposits, the precipitation temperature was $t \sim 20°$ Celsius. The paleo-geothermal gradient interpretations are only marginally sensitive to the precipitation temperature assumption and, 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 ($\beta_{(d)} = 0$); ii) reconstruction B - made assuming that the combined isotopic fractionation factor is larger than the equilibrium fractionation factor ($\beta_{(d)} > 0$); and iii) reconstruction C - made assuming that the combined isotopic fractionation factor is smaller that the equilibrium fractionation factor ($\beta_{(d)} < 0$).

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

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

Figure 38.

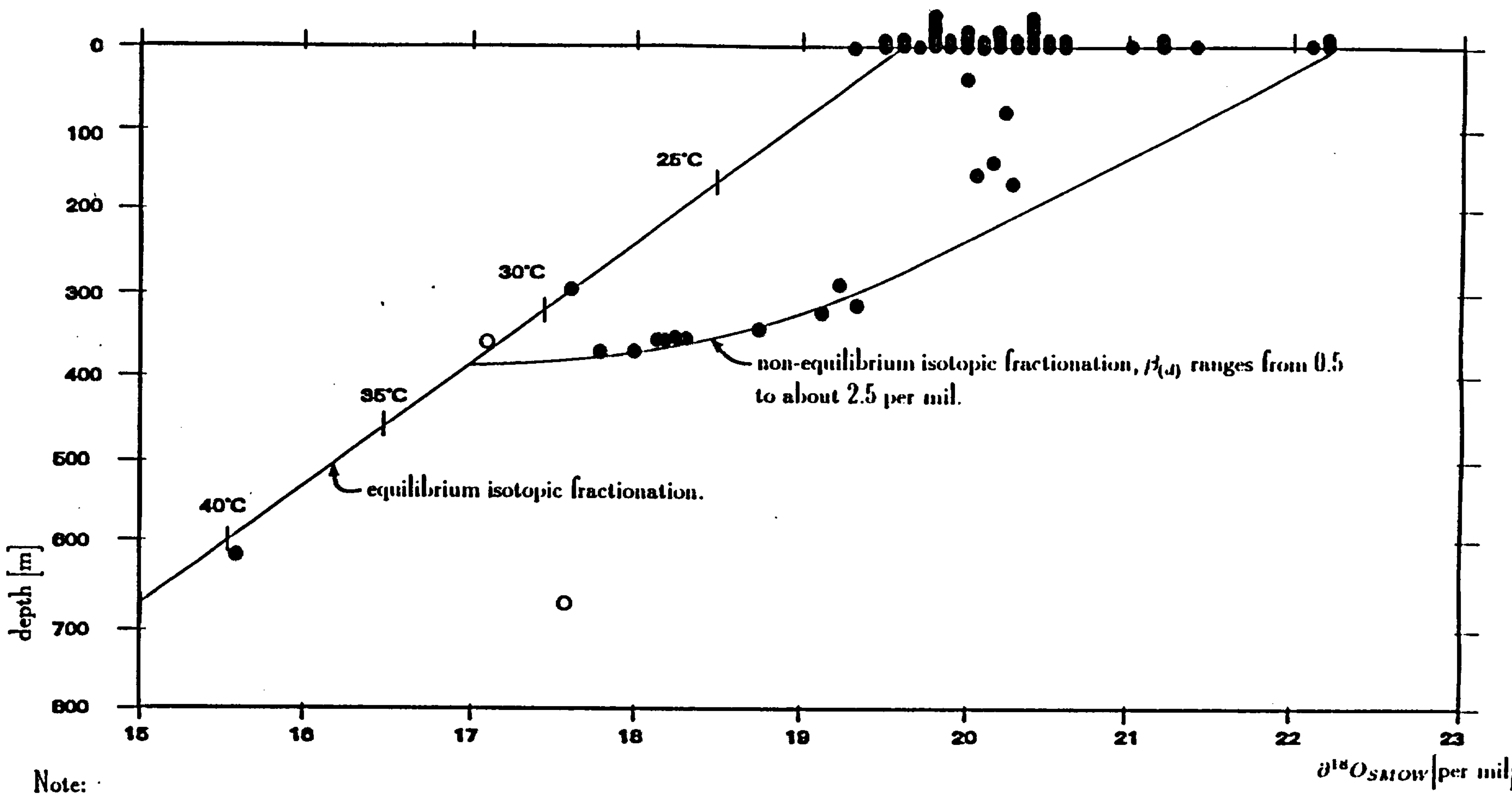


Note:

a) In performing this reconstruction, it has been assumed that: i) at the topographic surface, the precipitation temperature for calcretes and veins was $t \sim 20^o$ 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 ° Kelvin; and

b) Value of the paleo-geothermal gradient is $dt/dz \sim 35^o$ Celsius per 1km increase in depth - the corresponding contemporary geothermal gradient ranges from 20 to 24° 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.



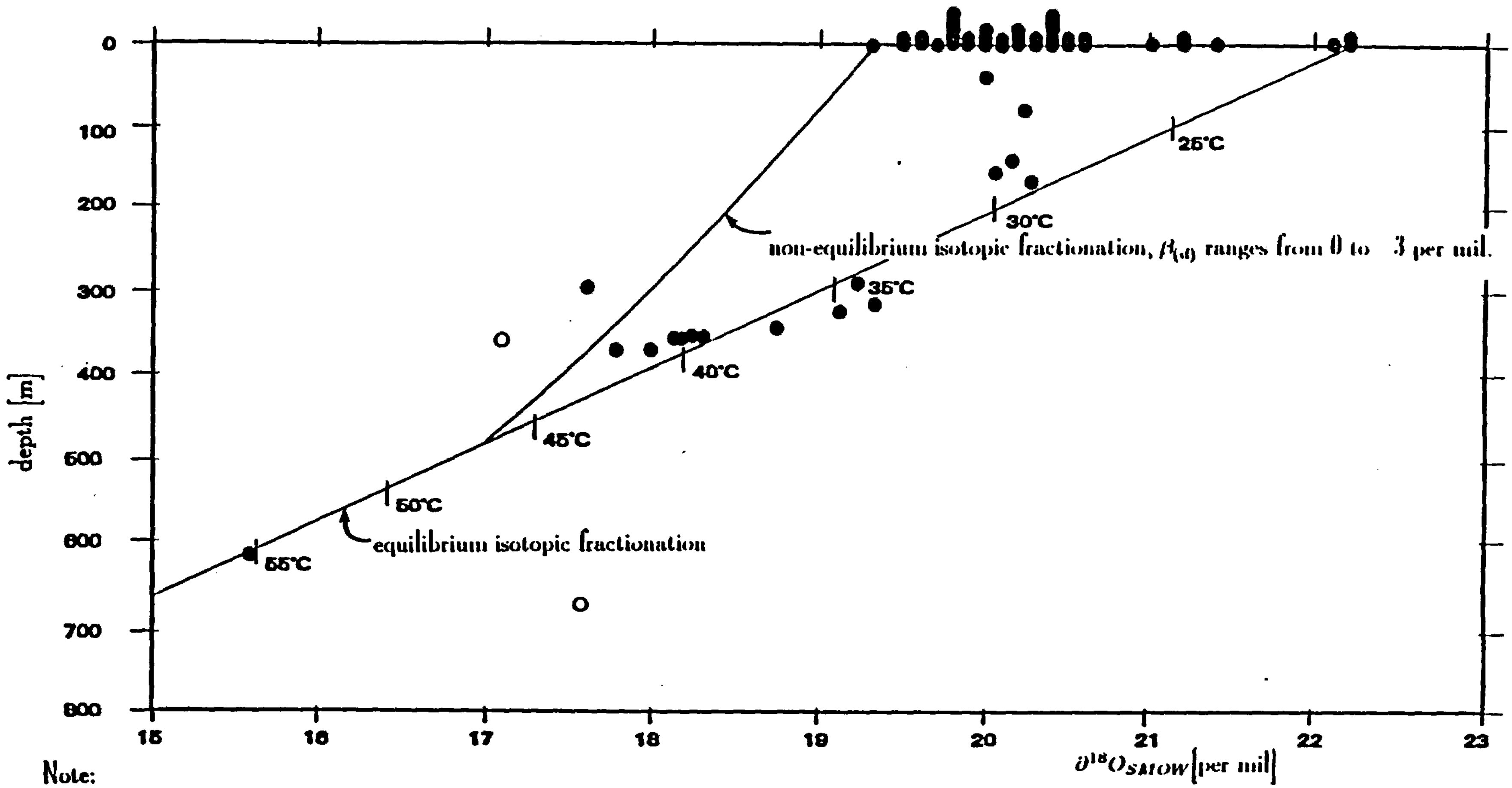
Note:

a) In performing this reconstruction, it has been assumed that: i) at the topographic surface, the precipitation temperature for calcretes and veins was t ~20° Celsius; ii) the oxygen −18 content of the parent fluids, for spatio-temporarly 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 $10^3 ln\alpha_{CaCO_3-H_2O} = 2.78 \cdot 10^6/T^2 - 2.82 + \beta_{(a)}$; and iv) the non-equilibrium fractionation is caused by a rapid, and depth variant escape of CO_2 from the parent solution - the CO_2 degassing rate decreases depthward, and $CaCO_3$ acquires its oxygen from both H_2O and CO_2; and

b) Value of the paleo-geothermal gradient is dt/dz ~33° 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 $t \sim 20^\circ$ 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 $10^3 ln\alpha_{CaCO_3 - H_2O} = 2.78 \cdot 10^6/T^2 - 2.82 - \beta_{(d)}$; and iv) the non-equilibrium fractionation is caused by a rapid upward movement of the parent fluids - the $CaCO_3$ nucleation occures 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 $dt/dz \sim 58^\circ$ Celsius per 1km increase in depth - the corresponding contemporary geothermal gradient ranges from 20 to 24° 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.

- 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.

- 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 UE-25a, $dt/dz \sim 22°$ Celsius per 1 km of depth; ii) Well USW G-2, $dt/dz \sim 24°$ Celsius per 1km of depth; iii) Well USW G-3, $dt/dz \sim 22°$ Celsius per 1km of depth; and iv) Well USW G-4, $dt/dz \sim 20°$ Celsius per 1km of depth.

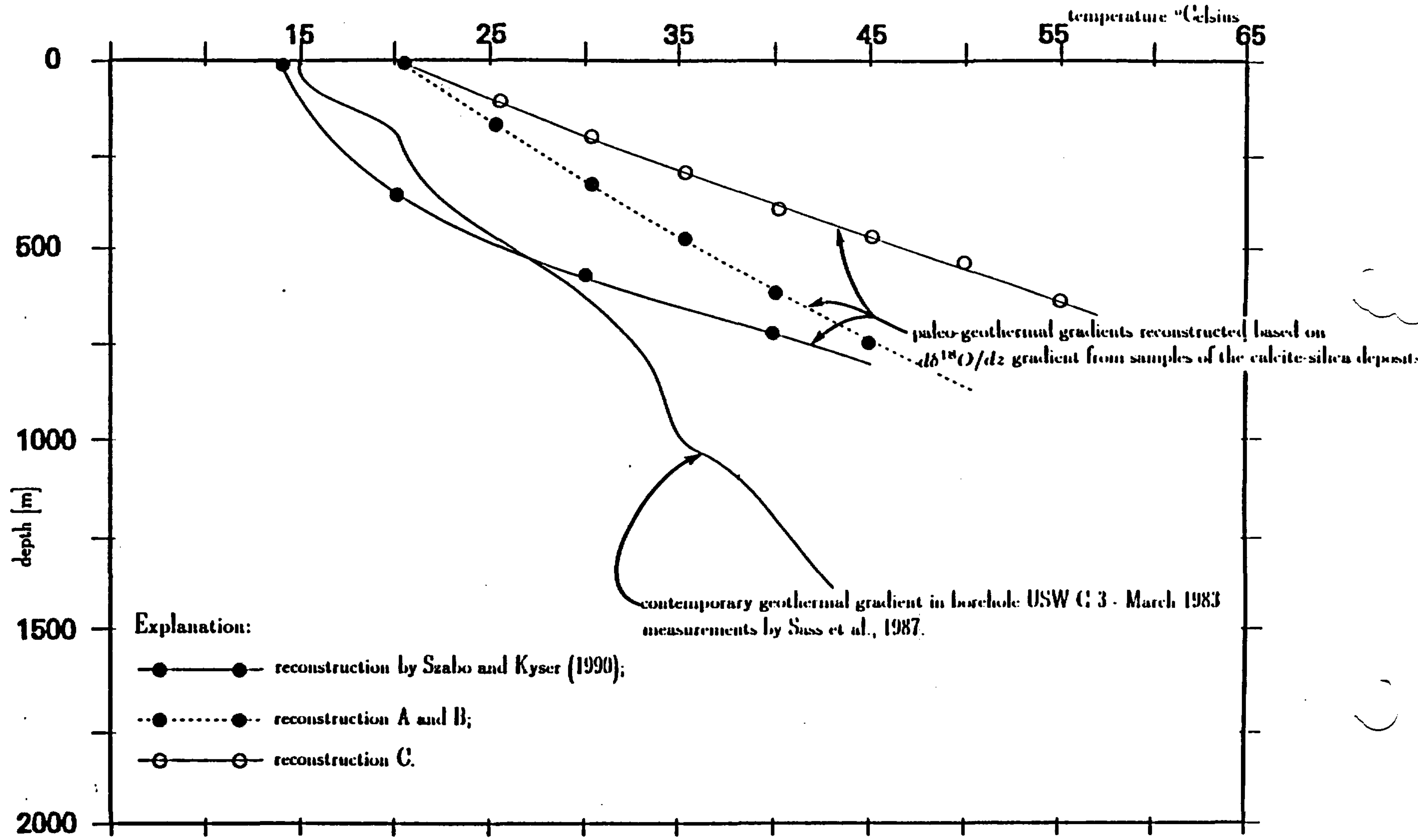
- The reconstructed values of the paleo-geothermal gradient are: i) reconstruction by Szabo and Kyser (1990) - dt/dz ranging from 17, near the topographic surface, to 50° Celsius per 1km of depth, in deeper parts of the vadose zone; ii) reconstruction A - $dt/dz \sim 35°$ Celsius per 1km of depth; iii)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

- 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.

- 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 ascensum* process.

Summary - the paleo-geothermal gradient reconstructions.

Figure 42.



Note:

a) Each of the considered cases yields the paleo- geothermal gradient that is substantially higher than the contemporary geothermal gradient; and

b) The geothermal gradients discrepancy indicates that precipitation of the calcite-silica deposits was accompanied by a significant warming up of the vadose zone and, therefore, occurred as the *per ascensum* process.

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.

- 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.

- 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.

- 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. 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.

Figure 44