UNITED STATES
NUCLEAR REGULATORY COMMISSION
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FROM: Paul T. Prestholt, Sr. On-Site Licensing Representative

DATE: May 17, 1991

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

Please find enclosed a copy of the above-referenced document.

PTP:nan
cc: Joseph Holonich, M/S 4 H 3 w/o encs.

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Rec'd with letter dtd 3/17/91

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QA: N/A

MAY 13 1991

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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 CO_2 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.

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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 del ^{13}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 $CaCO_3$-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 del ^{13}C ratio, from samples of the shallow vadose zone CO_2 gas, are: (1) for the Amargosa Narrows area, del ^{13}C ~-20 per mil $_{PDB}$; (2) for the Yucca Mountain area, del ^{13}C ~-18.36 per mil $_{PDB}$; and (3) for the Nevada Test Site area as a whole, del ^{13}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 del ^{13}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 del ^{13}C ~-12 per mil $_{PDB}$. Correcting the -14 per mil $_{PDB}$ value, from Figure 2 of the January 18 letter, would yield values of the del ^{13}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 del ^{18}O vs. del ^{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," 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 infilings; 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 del ^{13}C vs. del ^{18}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 del ^{13}C vs. del ^{18}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 del ^{13}C vs. del ^{18}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. <u>On the one hand</u>, 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 ~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 $0.5 g/cm^2/1000$ 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 $0.14 g/cm^2/1000$ years; this rate is 25 to 75 percent of equivalent-age soils in New Mexico." Second, the Yucca Mountain fault infilings, 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 CO_2 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 then 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 infilings of the fault related fissures exhibit nearly vertically banded textures. As it may be observed in Trenches 8 and 14, for example, these infilings 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 infilings or the conditions of precipitation of these infilings 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 infilings. 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 infilings 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 del ^{13}C ratio ranging from -4.75 to -9.25 per mil $_{PDB}$, Figure 15; (2) the mean values of the del ^{13}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 del ^{13}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 $_{PDB}$, Figure 17. As illustrated in Figure 18, I hope with sufficient clarity, both the observed and the expected values of the del ^{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 del ^{13}C range from -4.75 to -9.45 per mil $_{PDB}$, 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 $_{PDB}$ 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 $_{PDB}$, 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 del $^{13}C/dz$ ~4 per mil $_{PDB}$ per 1km 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 del $^{18}O/dz$ ~2 per mil $_{SMOW}$ per 1km 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 del $^{13}C/dz$ ~4 per mil $_{PDB}$ 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 del $^{18}O/dz$ ~2 per mil $_{SMOW}$ per 1km 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. <u>First</u>, 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. $^{234}U/^{238}U$ 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 $^{87}Sr/^{86}Sr$ 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 del ^{13}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 <u>irresponsible science</u>" 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 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" $\longleftrightarrow$ "source" and "saddle" $\longleftrightarrow$ "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 CO_2 gas assisted hydrothermal eruption; (4) the CO_2 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 CO_2 isotopic data are used to demonstrate that, for the Yucca Mountain area, there is no rational reason to believe that the vadose zone CO_2 originates solely from the atmosphere-biosphere source. Please make a note that, the available CO_2 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 CO_2 changes and becomes "heavier" with the increasing depth, Figure 58; (3) the vadose zone CO_2 exhibits the depthward diminishing concentrations of carbon-14, Figure 59; and (4) values of the CO_2 partial pressure fluctuate about the mean atmospheric value of log P_{CO2} ~-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 CO_2 indexes (del ^{13}C, PMC, and log P_{CO2}), as observed in the Yucca Mountain vadose zone, and the origin of CO_2 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 CO_2 gases that originate simultaneously from two independent sources. One of the CO_2 sources is situated at and near the topographic surface, and corresponds to the atmosphere-biosphere system. The other CO_2 source, the so-called subterranean CO_2 source, is situated along the local water table. The CO_2 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 CO_2 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 ^{13}C ratio, Figures 65 and 66; (5) relative to the tuff "pile" based fluids,

the Paleozoic carbonates based fluids carry significantly higher values of the del ^{13}C ratio, Figures 65 and 66; and (6) the Paleozoic carbonates based fluids carry values of the del ^{13}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_{CO2} 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 del ^{13}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 del ^{13}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 del ^{13}C ratio for fluids equilibrated with marine limestones, the Paleozoic carbonates based fluids carry abnormally low values of the del ^{13}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 $C0_2$ 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 $C0_2$. This additional $C0_2$ 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 del ^{13}C ratio, as observed for the Paleozoic carbonates based geothermal fluids, by postulating the partial involvement of the sub-lithospheric mantle indigenous $C0_2$. 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 $C0_2$ 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 CO_2 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 CO_2; (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 CO_2, and (3) interpret the observed discrepancies, between the observed carbon-13 content of the soil CO_2 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. <u>On the other hand</u>, 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

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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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91-1545

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WBS 1.2.5
QA: N/A

NNA.910207.0098

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TECHNICAL MEMORANDUM REGARDING SITE SUITABILITY OF YUCCA MOUNTAIN

This past week, January 2-4, 1991, I have thoroughly reviewed the letter and technical analysis that Jerry Szymanski is preparing to send to the editor of Science. The technical findings presented by Jerry Szymanski in this document are irrefutable. The technical analysis is sound and the data considered are sufficient to support his thesis. Furthermore, from my familiarity with the broader analysis and compilation of isotopic data regarding Yucca Mountain and southern Nevada at large, which is still in preparation by Jerry Szymanski, I am confident that any additional data, when properly considered, will continue to support these findings. In addition, I have again reviewed Section 4.5.2.3 "The Skull Mountain Hydrologic Anomaly" of Jerry Szymanski's 1989 report and conclude this locality is likely an active analogue of the paleo-conditions at Yucca Mountain as is the Oasis Valley locality near Beatty, Nevada, which was discussed by Jerry Szymanski in the 1987 version of his report.

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.

Donald E. Livingston

Donald E. Livingston
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Yucca Mountain Site Characterization
Project Office

RSED:DEL-1545

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YMP-5

ENCLOSURE 1

Department of Energy
Yucca Mountain Site Characterization
Project Office -
P. O. Box 98608
Las Vegas, NV 89193-8608

WBS 1.2.3.6.2
QA: N/A

FEB 05 1991

David C. Dobson, YMP, NV
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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

ENCLOSURE 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

Donald E. Livingston
Technical Analysis Branch
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RSED:DEL-2046

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

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APR 30 1991

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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 numerously 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.

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APR 30 1991

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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RSED:DEL-3483

Enclosure:
Ltr, 12/4/90, Livingston
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APR 30 1991

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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;

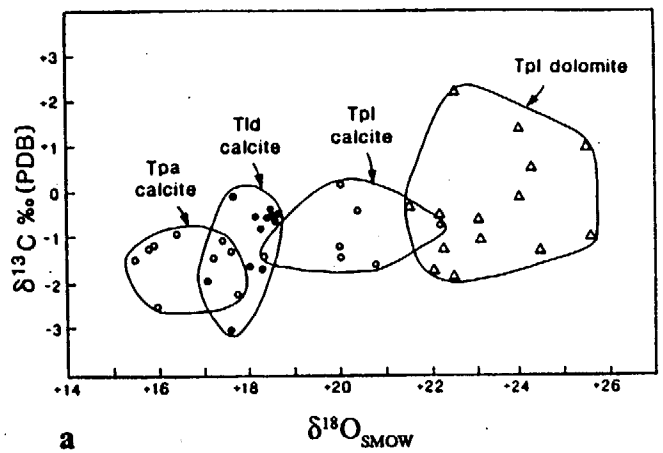
b) a reasonable estimate for the mean altitude of the Yucca Mountain area is ~ 1200 m;

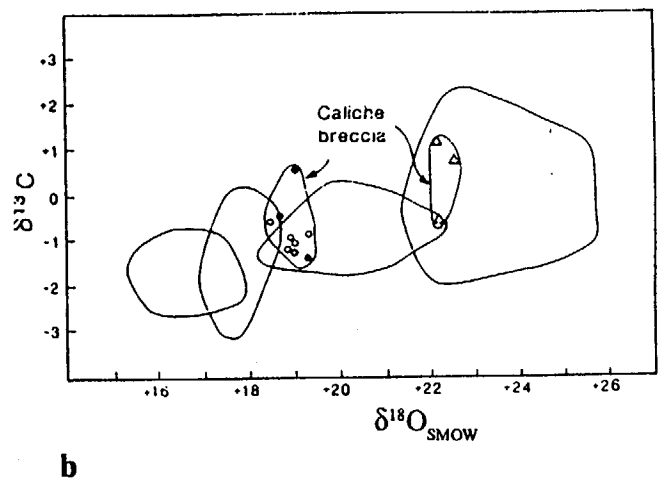
c) assuming that the proportions of the C-3 plants vary linearly with altitude (between 840 and 1550 m altitudes), a reasonable estimate for the Yucca Mountain C-3 proportion is ~ 85 percent; and

d) there is no assurance that the soil CO_2 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).

ENCLOSURE 5



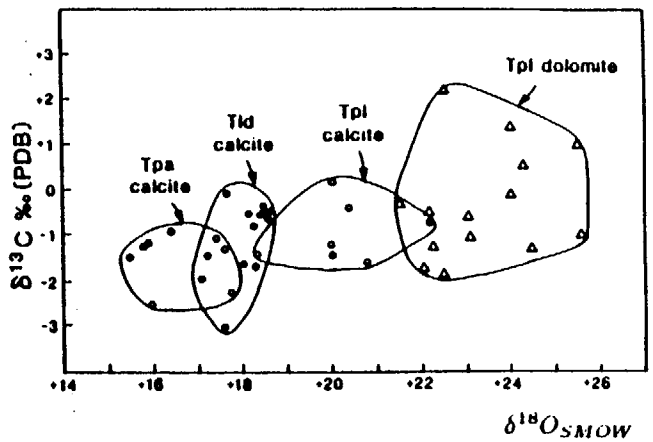


a) Composition of carbonate minerals in bedded claystones and carbonate rocks. Dolomite is represented by triangles and calcite by circles, which are closed for limestones of Tld. Tpa samples are of chalky limestones, most of which are soft; Tld samples are mostly of dense nodular and fenestral limestone;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

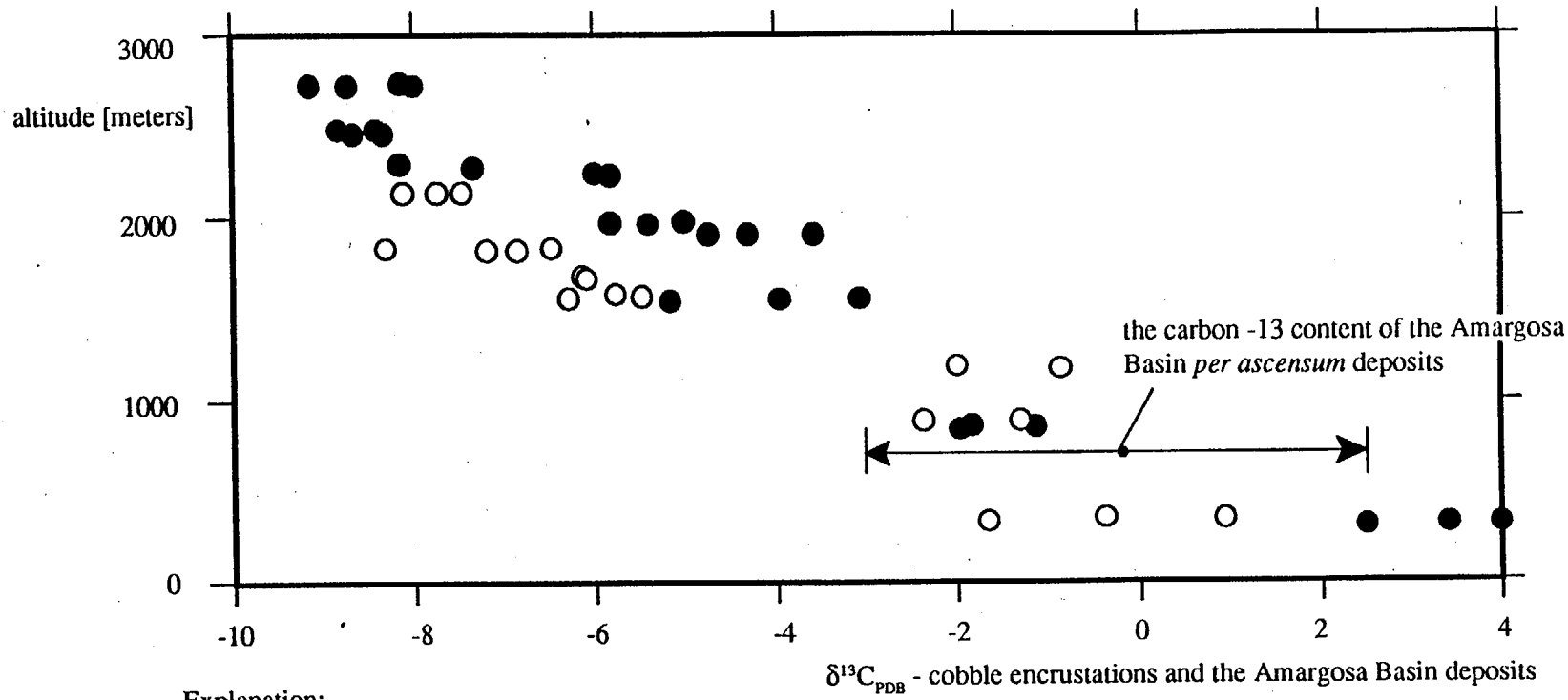
Explanation of the lithologic notations as employed by Hay et al. (1986). From Hay et al. (1986)



Note:

a) the overlapping $\delta^{13}C$ vs $\delta^{18}O$ fields indicate that, all four of the distinguished lithofacies were precipitated from the common *per ascensum* parent fluids;
b) 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);
c) 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;
d) 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;
e) 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;
f) 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;
g) 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 ascenusm* fluids undergo, as expected, the progressive alteration of their initial stable isotopic contents;
h) 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) 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;

O - non-calcareous bedrock; and

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

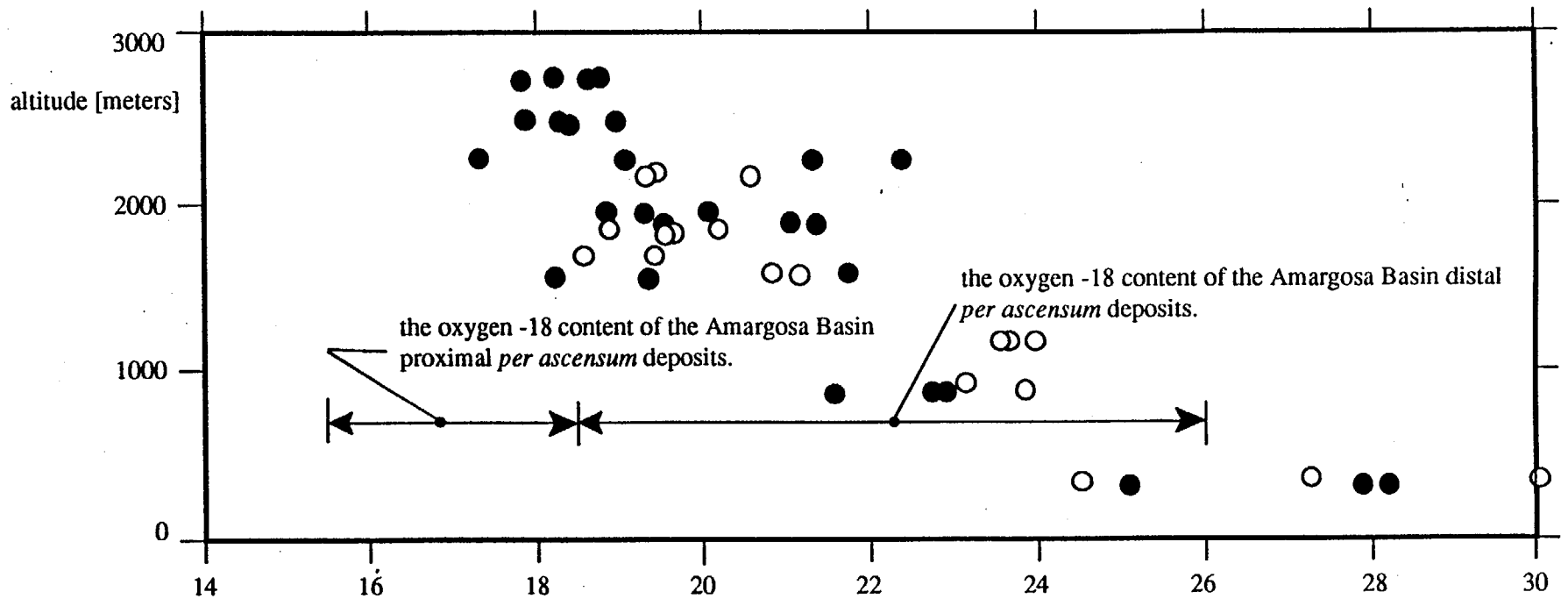
Note:

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 $\delta^{13}C$ 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:

$\delta^{18}O_{SMOW}$ - cobble encrustations and the Amargosa Basin *per ascensum* deposits.

● - calcareous bedrock;

○ - non-calcareous bedrock; and

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

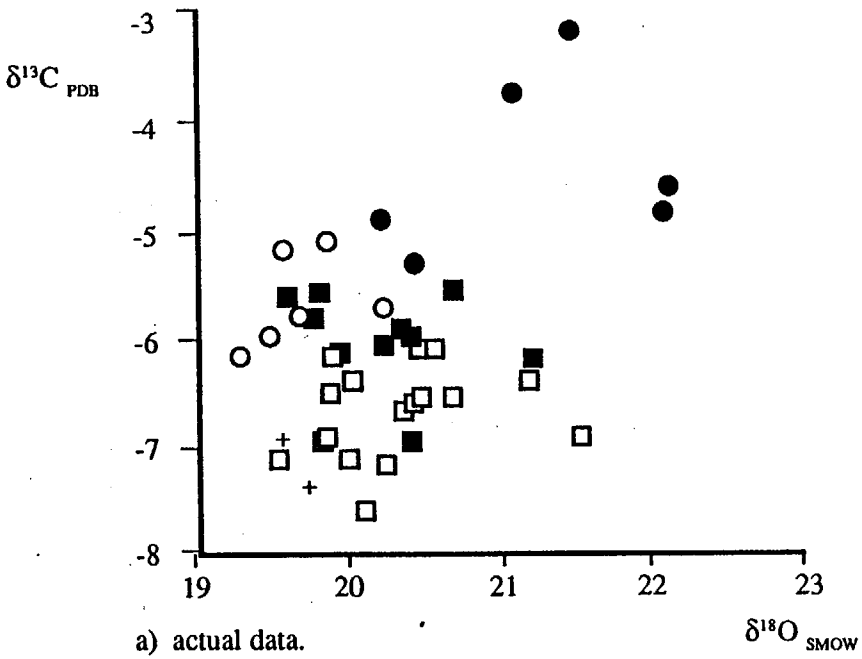
Note:

a) the cobble encrustation oxygen -18 data are from Quade et al. (1988) and the Amargosa Basin data are from Hay et al. (1986);

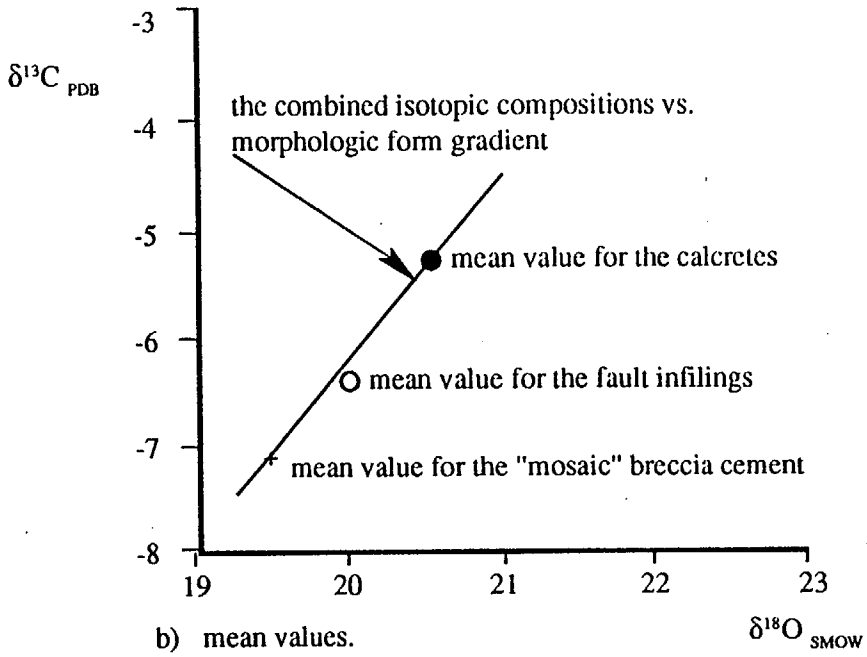
b) samples of both the local cobble encrustations and the Amargosa Basin distal *per ascensum* deposits yield similar values of the $\delta^{18}O$ ratio; and

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.



a) actual data.



b) mean values.

Explanation:

■ - sample represents the Trench 14 calcretes;

● - sample represents the Busted Butte calcretes;

□ - sample represents the Trench 14 fault infiling;

○ - sample represents the Busted Butte fault infiling; 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 infilings, and iii) the calcretes;

c) the $\delta^{18}O$ vs $\delta^{13}C$ 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 $\delta^{13}C$ vs $\delta^{18}O$ 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 infilings, 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 infilings, 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 consistant with the *per ascensum* origin of the Yucca Mountain calcite-silica deposits - similarily 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 $\delta^{18}O$ 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 $d\delta^{18}O/dz$ 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 infilings, consists of the distal component and is equal to 2 per mil $_{SMOW}$ x 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;

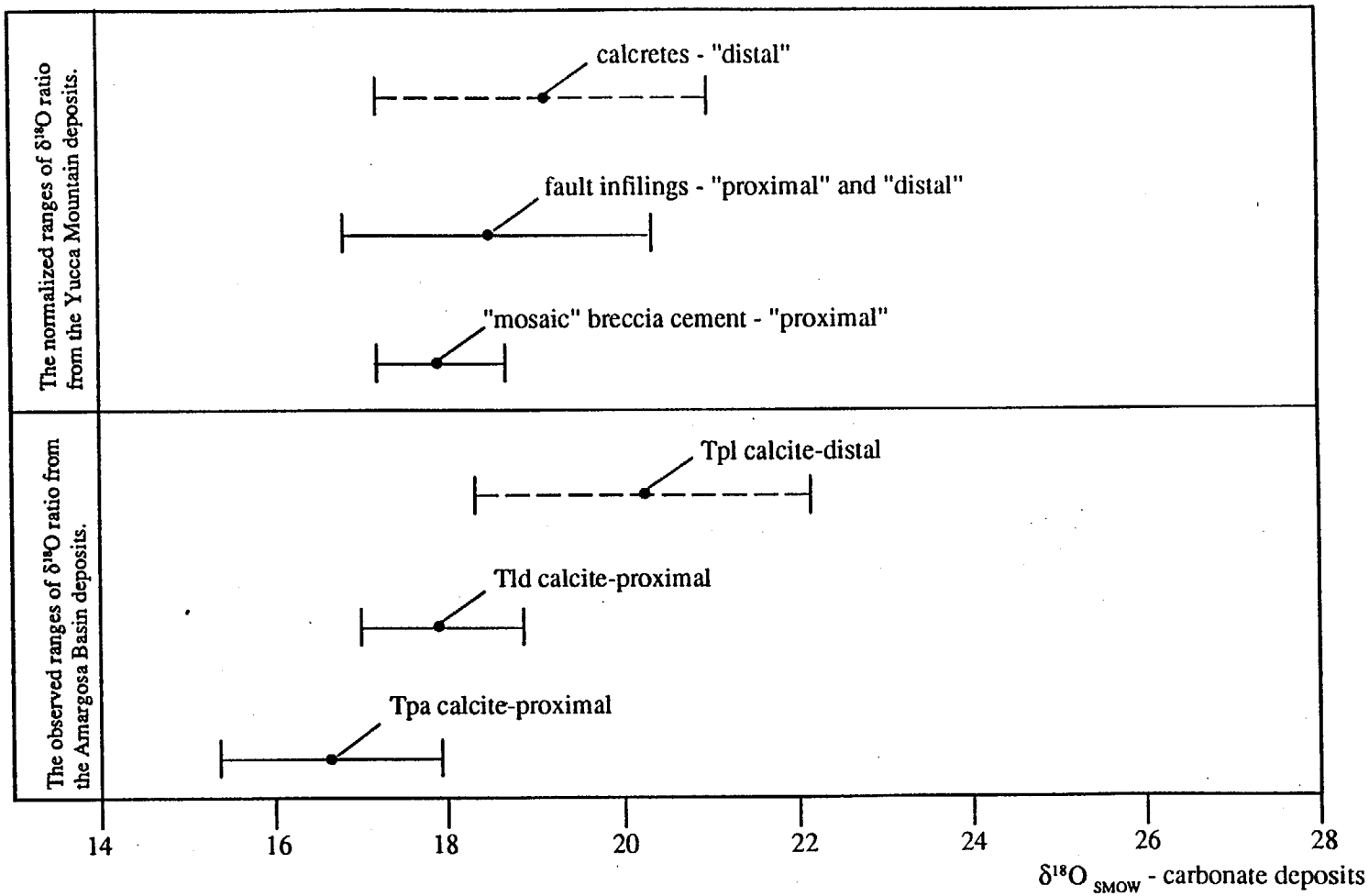
f) using the geothermal data from Garside and Schilling (1979), it may be assumed that the precipitation temperature, for the Amargosa Basin proximal deposits, is ~ 32°Celsius [i.e., ~ 12°Celsius above the local ambient temperature];

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 infilings, is -1.2 per mil $_{SMOW}$ - it has been computed for T = 23°Celsius, using the equilibrium fractionation equation $10^3 \ln\alpha_{CaCO_3\text{-}H_2O} = 2.78 \cdot 10^6 \cdot T^{-2} - 2.89$, from Turi (1986); and

i) the total normalization factor, for the Yucca Mountain "mosaic" breccia cement and for the isotopically "lightest" fault infilings, is -2.4 per mil $_{SMOW}$.

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



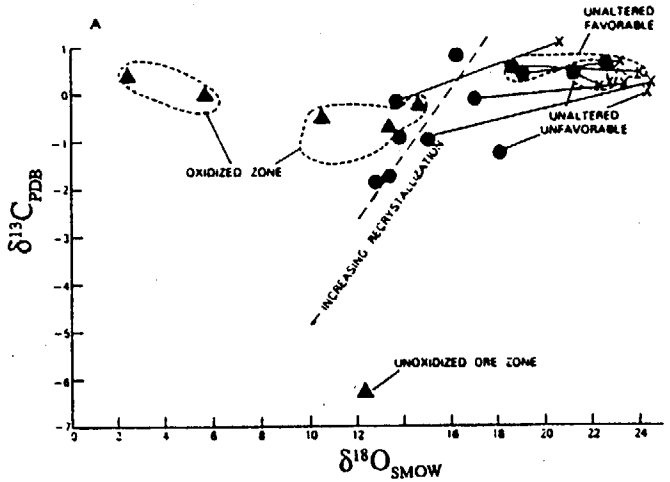
Note:

a) for the Yucca Mountain "distal" deposits, the normalization factor is -1.2 per mil $_{SMOW}$;

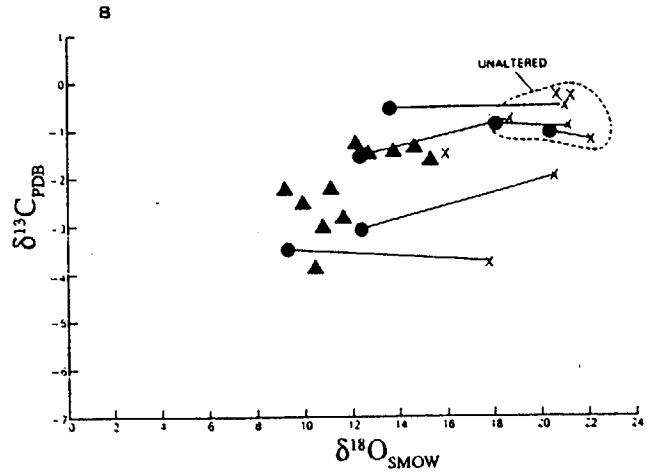
b) for the Yucca Mountain "proximal" deposits, the normalization factor is -2.4 per mil $_{SMOW}$; and

c) 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.



a) the Carlin disseminated gold deposit of Nevada.



b) the Cortez disseminated gold deposit of Nevada.

Explanation:

▲ - sample represents altered host rock and calcite veins in oxidized and unoxidized zones;

● - sample represents calcite veins;

x - sample represents dolomite; and

tielines connect coexisting calcite and dolomite

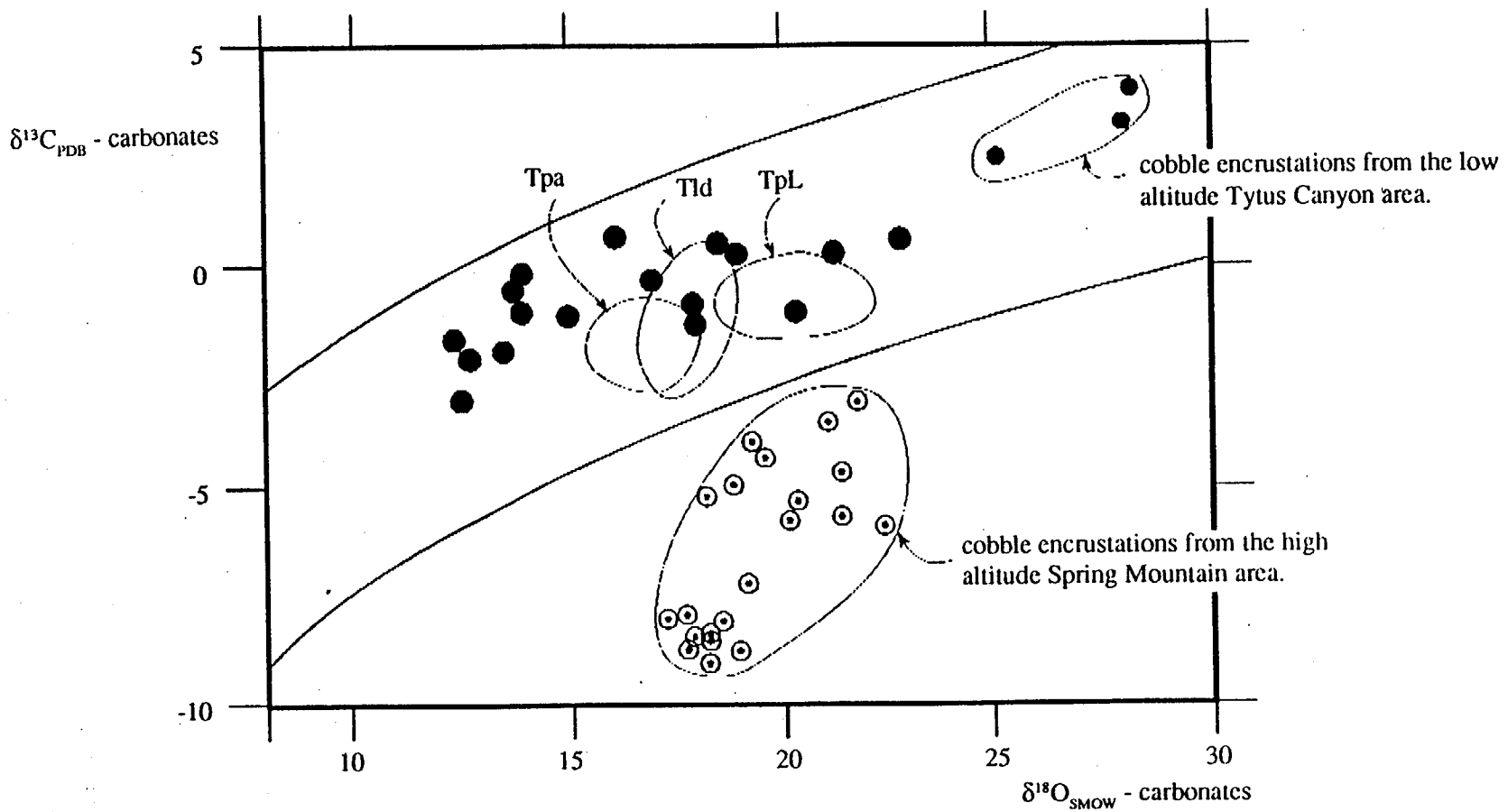
Note:

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.

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.

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



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.

Figure 10

Note:

a) the isotopic data, as shown, are from: Rye (1985), Hay et al. (1986), and Quade et al. (1988);

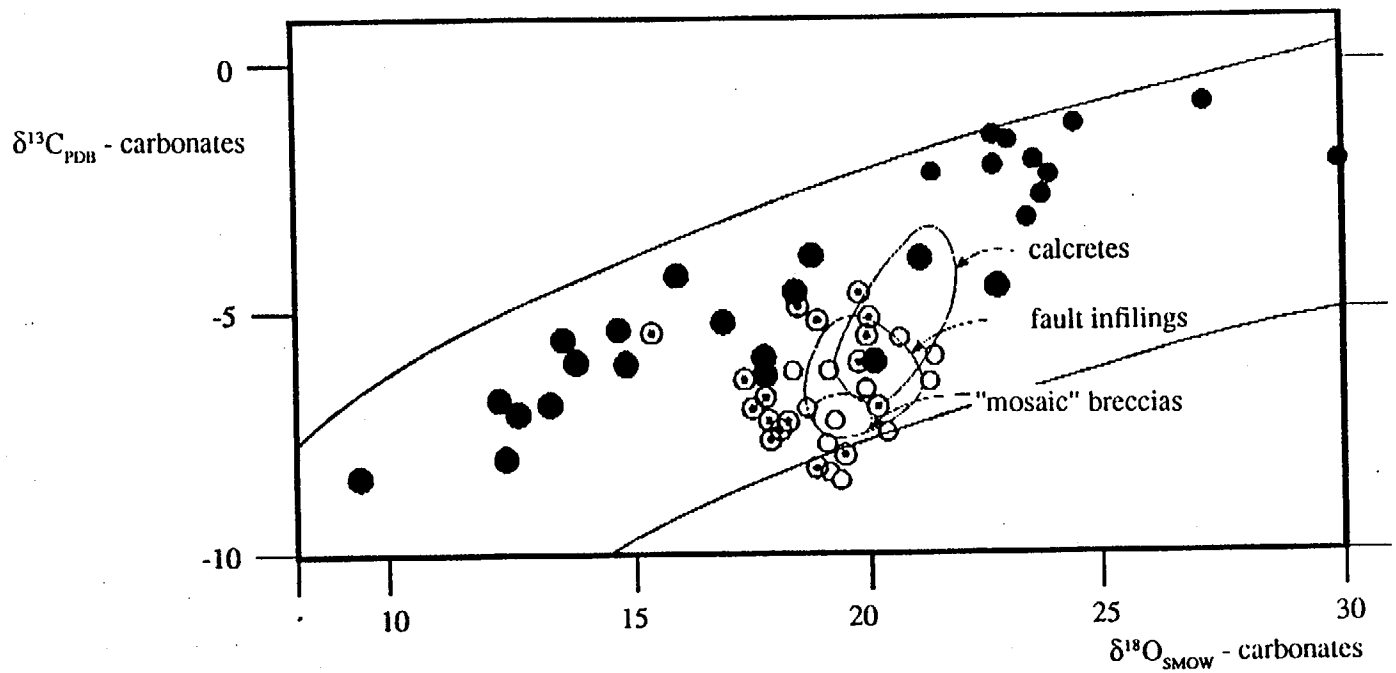
b) the observed trend toward the increasing carbon -13 and oxygen -18 contents, for all of the considered lithofacies but excluding the cobble encrustations from the high altitude Spring Mountain area, may be accounted for by assuming that: i) all of the lithofacies were precipitated from fluids that, 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 CO_2 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 atmosperic - 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.

Figure 11

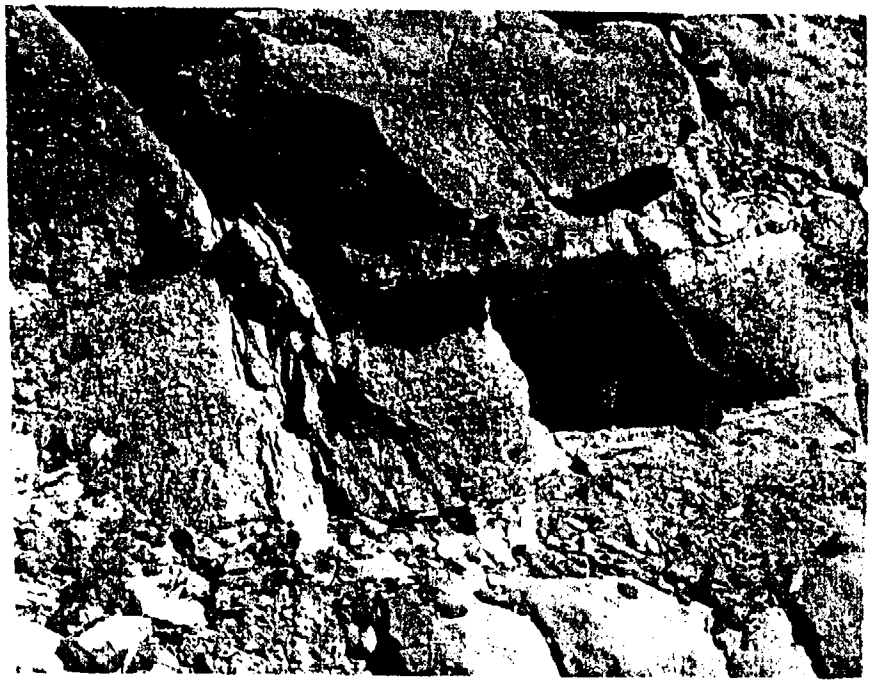
Note:

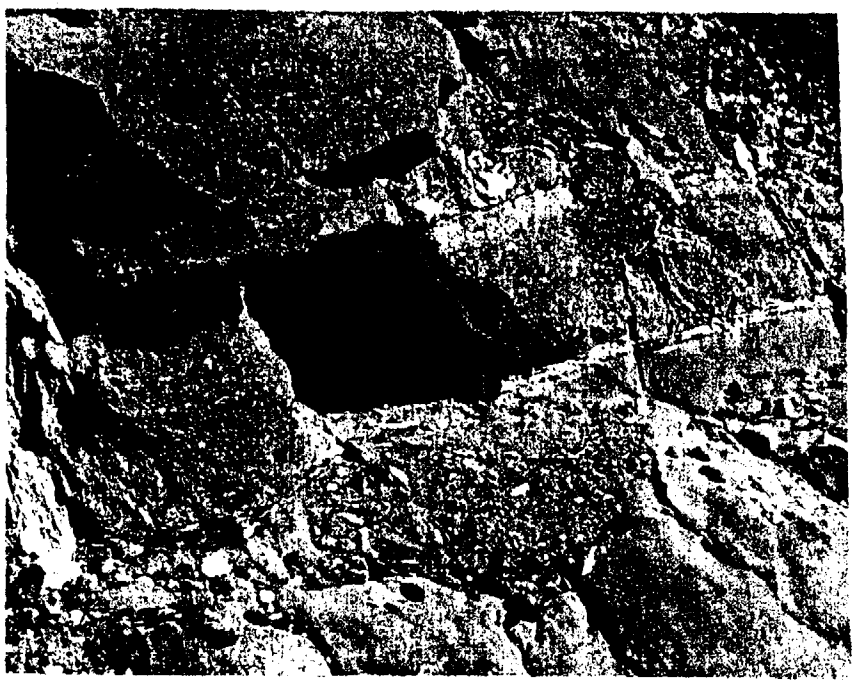
a) the isotopic data, as shown, are from: Rye (1985), Szabo and Kyser (1985), Quade et al. (1988), and Whelan and Stuckless (1990);

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 CO_2 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;

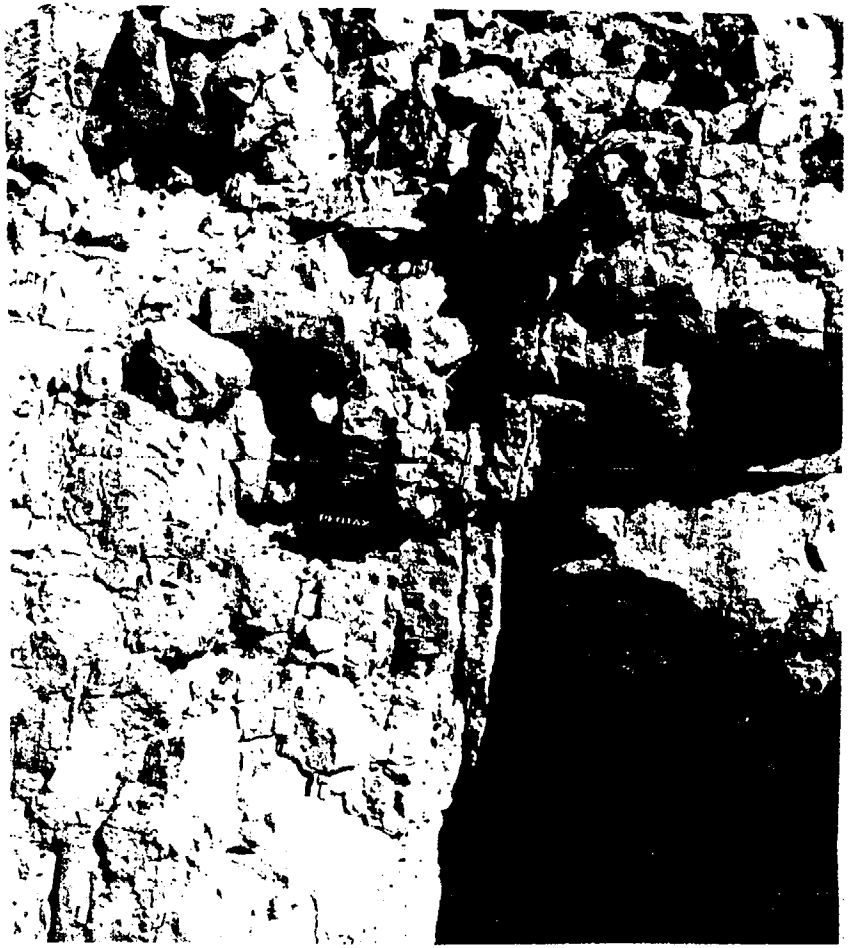
b) presence of both the locally very intense oxidation and the jasperoidal opals indicates that the observed bedded tuff alteration is caused by hydrothermal solutions; and

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.

Figure 12

a) opaline silica cement.



b) mixed calcite-opaline silica cement.

Note:

a) the host rock for the injection breccia dikes is the "columnar zone" of the Tiva Canyon Member; and

b) the breccia dikes contain clasts lithologically similar to the underlying Topopah Spring Member.

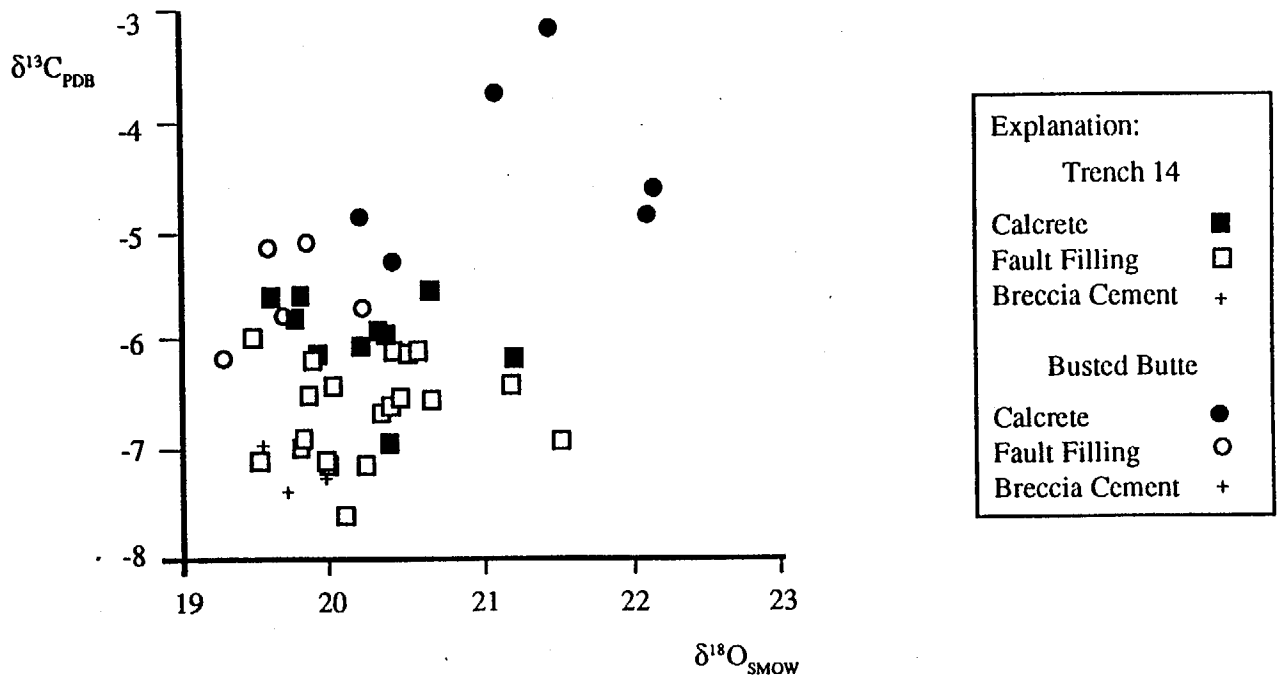
Hydro-clastic injection breccia dikes - eastern flank of the Harper Valley, southern part of the Yucca Mountain area.

Figure 13

- 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

Figure 14



Note:

a) the observed range for the $\delta^{13}C$ 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;

b) for precipitation temperature T = 15°Celsius, the equilibrium fractionation factor $10^3 \ln\alpha_{CaCO_3 - HCO_3^-} = 1.75$ per mil_{PDB}; and

c) the interpreted range for the $\delta^{13}C$ 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 $\delta^{13}C$ ratio - the parent fluids for the Yucca Mountain calcretes and surficial veins.

Figure 15

- A mean value of the $\delta^{13}C$ ratio for the CO_2 gas respired from the vadose zone, as observed at Yucca Mountain and Amargosa Narrows, is **$\delta^{13}C$ = -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.

- A mean value of the $\delta^{13}C$ ratio for soil gas, as observed at various locations throughout the Nevada Test Site, is **$\delta^{13}C$ = -20.5 per mil_{PDB}**, Boughton, 1986.

- The equilibrium isotopic fractionation factor $10^3 \ln\alpha_{HCO_3^- - CO_2(g)} = 9.483 \cdot 10^3 \cdot T^{-1} - 23.89$, Turi (1986) - for 15°Celsius, the fractionation factor is ~ 9 per mil_{PDB}.

- 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 $\delta^{13}C$ 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 $\delta^{13}C$ ratios from samples of the Nevada Test Site vadose zone gases.

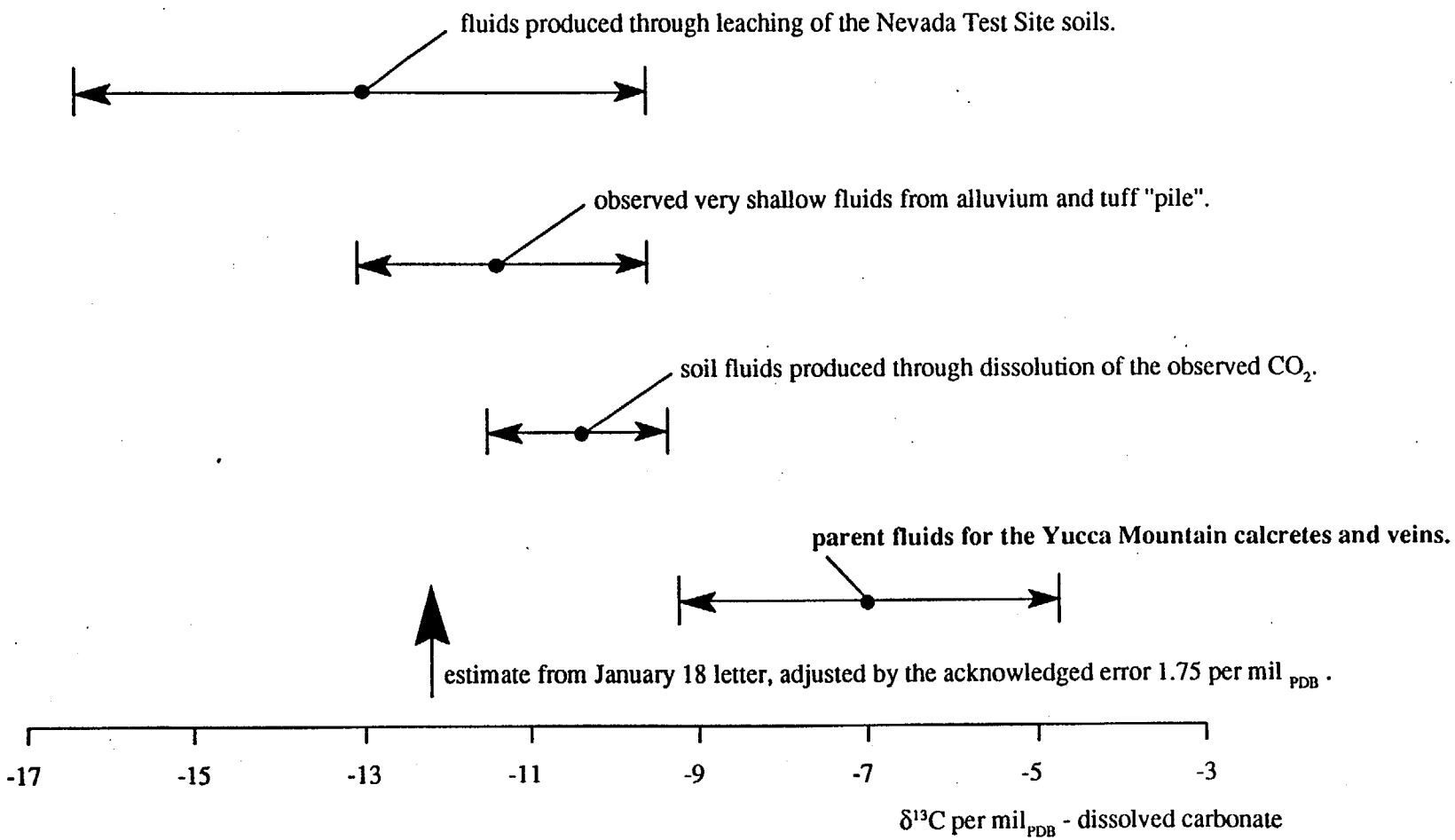
- A mean value for the $\delta^{13}C$ ratio from samples of shallow alluvium - based fluids, as observed around Amargosa Narrows, is $\delta^{13}C$ = **-11.3 per mil$_{PDB}$** (range of the observed values is **from -9.59 to -13.05 per mil$_{PDB}$**), White and Chuma (1987).

- A value of the $\delta^{13}C$ 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 $\delta^{13}C$ = **-13.0 per mil$_{PDB}$**, Benson and McKinley, 1985.

- A value of the $\delta^{13}C$ ratio for the Nevada Test Site soil water, as interpreted based on laboratory soil-leaching experiments, is $\delta^{13}C$ = **-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.

Figure 17



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

The $\delta^{13}C$ 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.

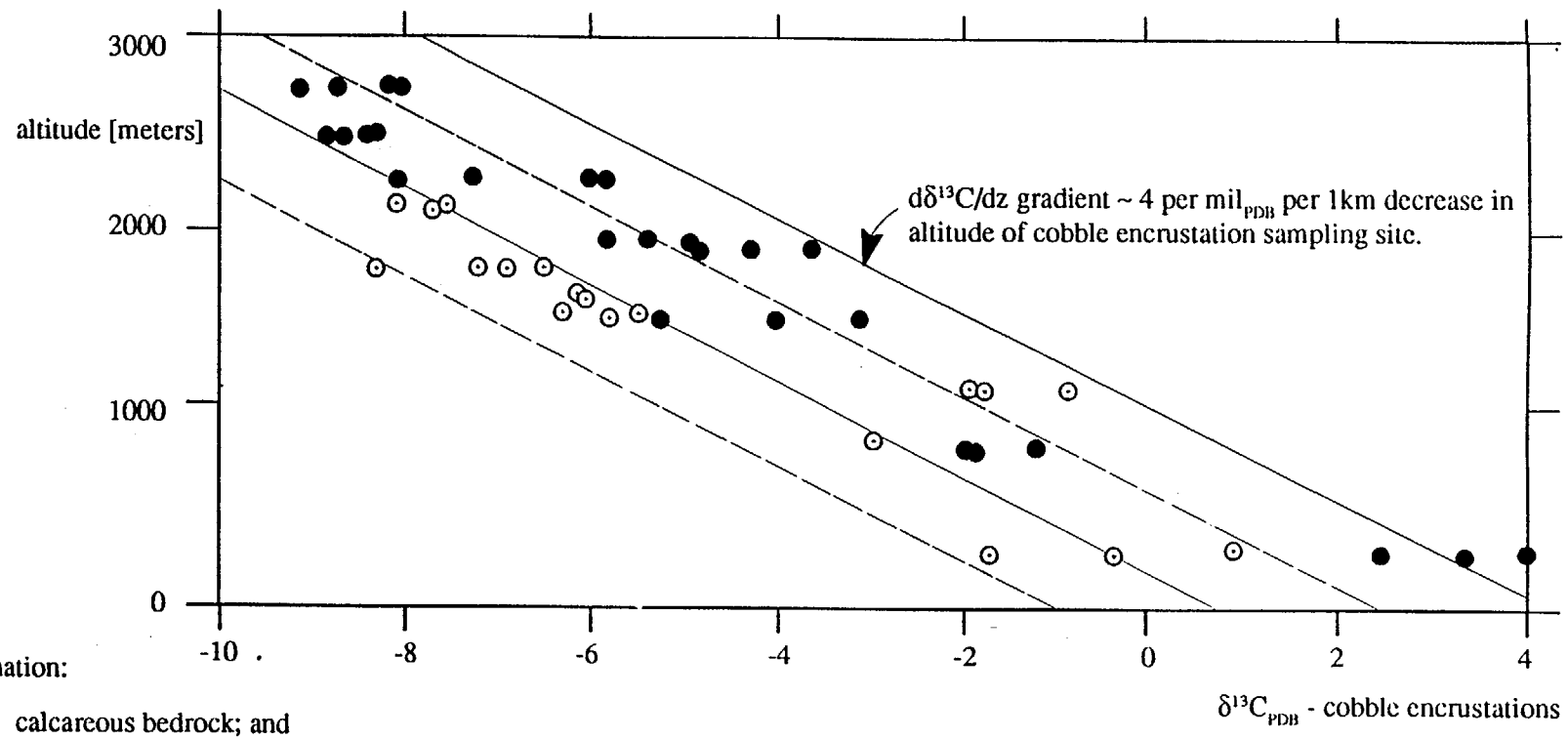
What is the diagnostic usefulness of the isotopic compositions vs altitude gradients, as observed by Quade and Cerling (1990) for the local cobble encrustations?

Statement of the issue - topic #2.

Figure 20

Sample No.	Elevation (m)	$\delta^{13}C_{PDB}$	$\delta^{18}O_{PDB}$
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.



Explanation:

● - calcareous bedrock; and

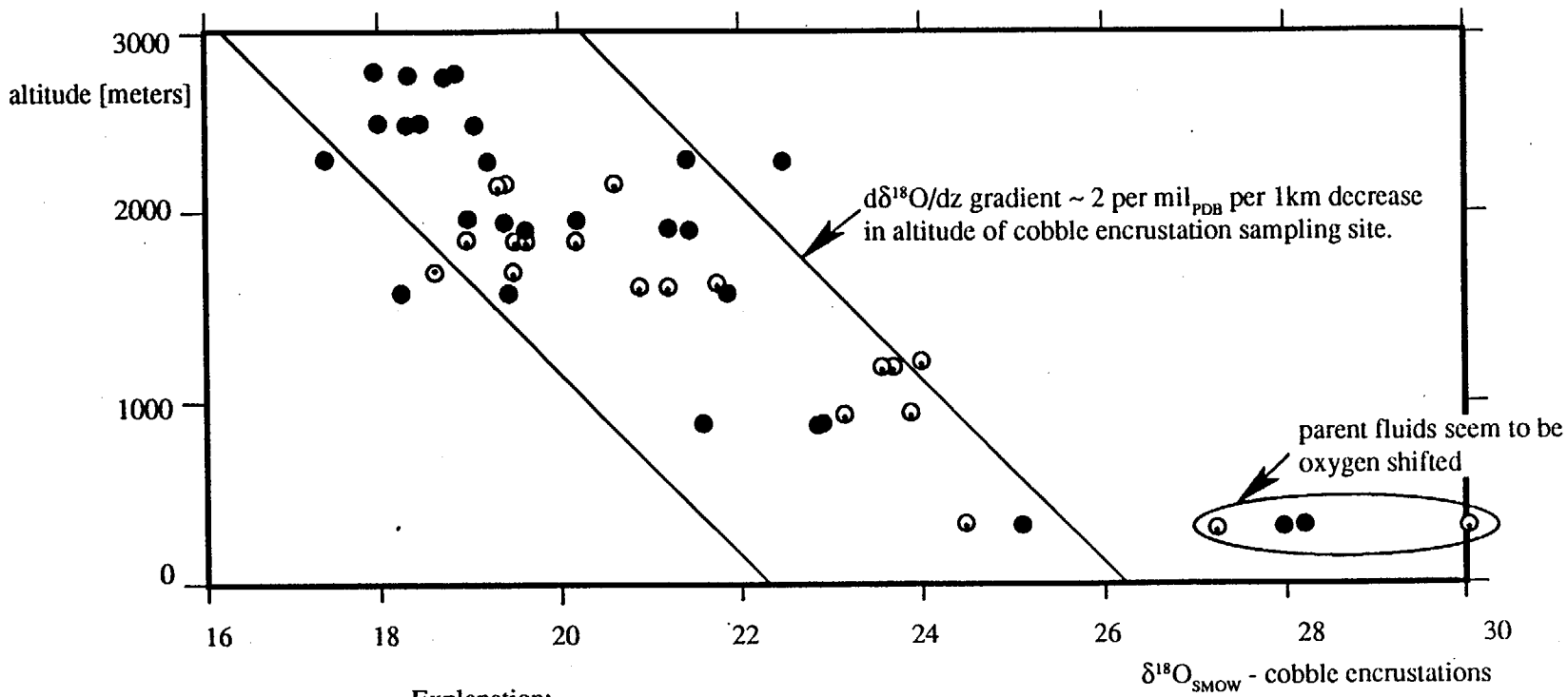
⊙ - non-calcareous bedrock.

Note:

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

Figure 22



Explanation:

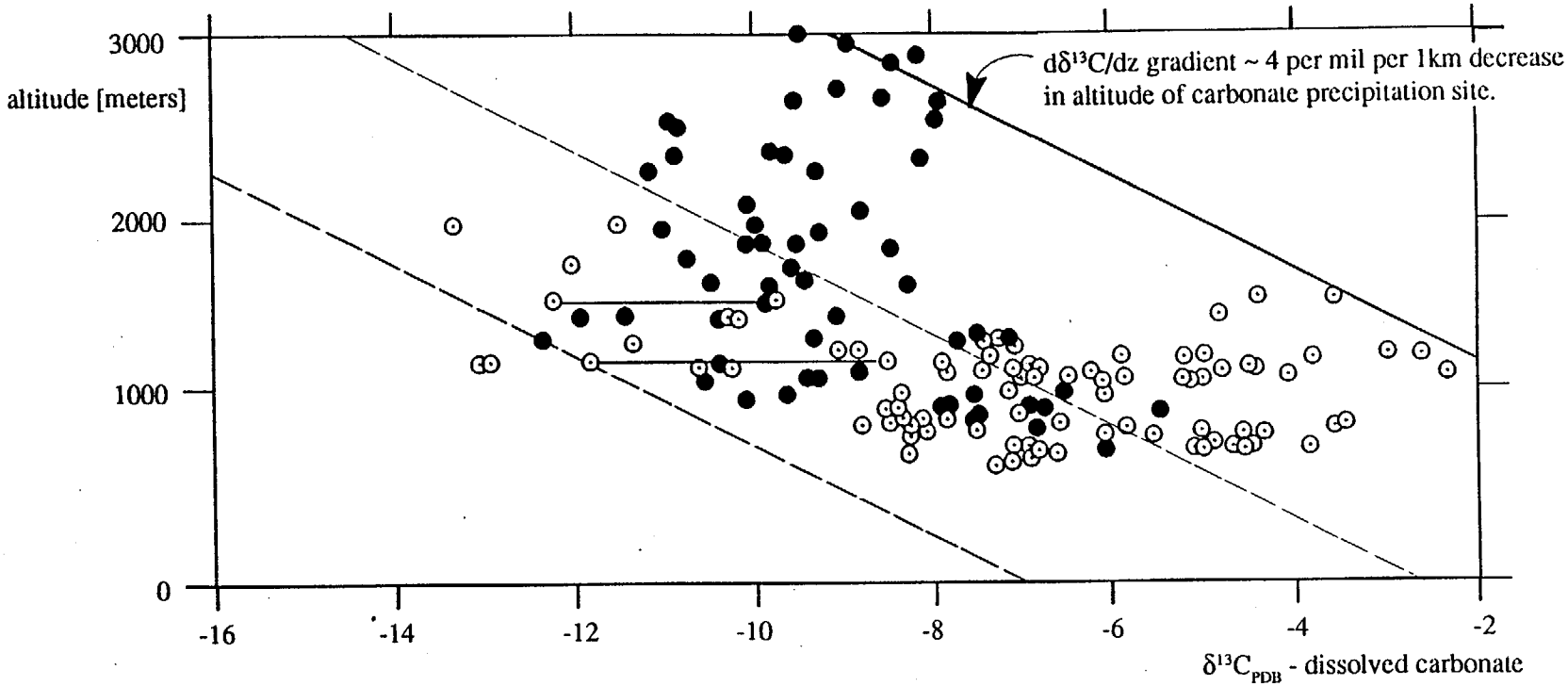
● - calcareous bedrock; and

⊙ - non-calcareous bedrock.

Note:

a) $\delta^{18}O_{SMOW} = 1.03086 \cdot \delta^{18}O_{PDB} + 30.86$, Fritz and Fontes (1980);
b) the $d\delta^{18}O/dz$ gradient has been computed assuming that, for a fixed altitude, the parent fluids have carried values of the $\delta^{18}O$ ratio varying by $\Delta\delta^{18}O \sim 4$ per mil_{SMOW}; and
c) 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.

tielines 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 $\delta^{13}C$ vs altitude gradient and field for the *per ascensum* cobble encrustations and distal calcretes.

Note:

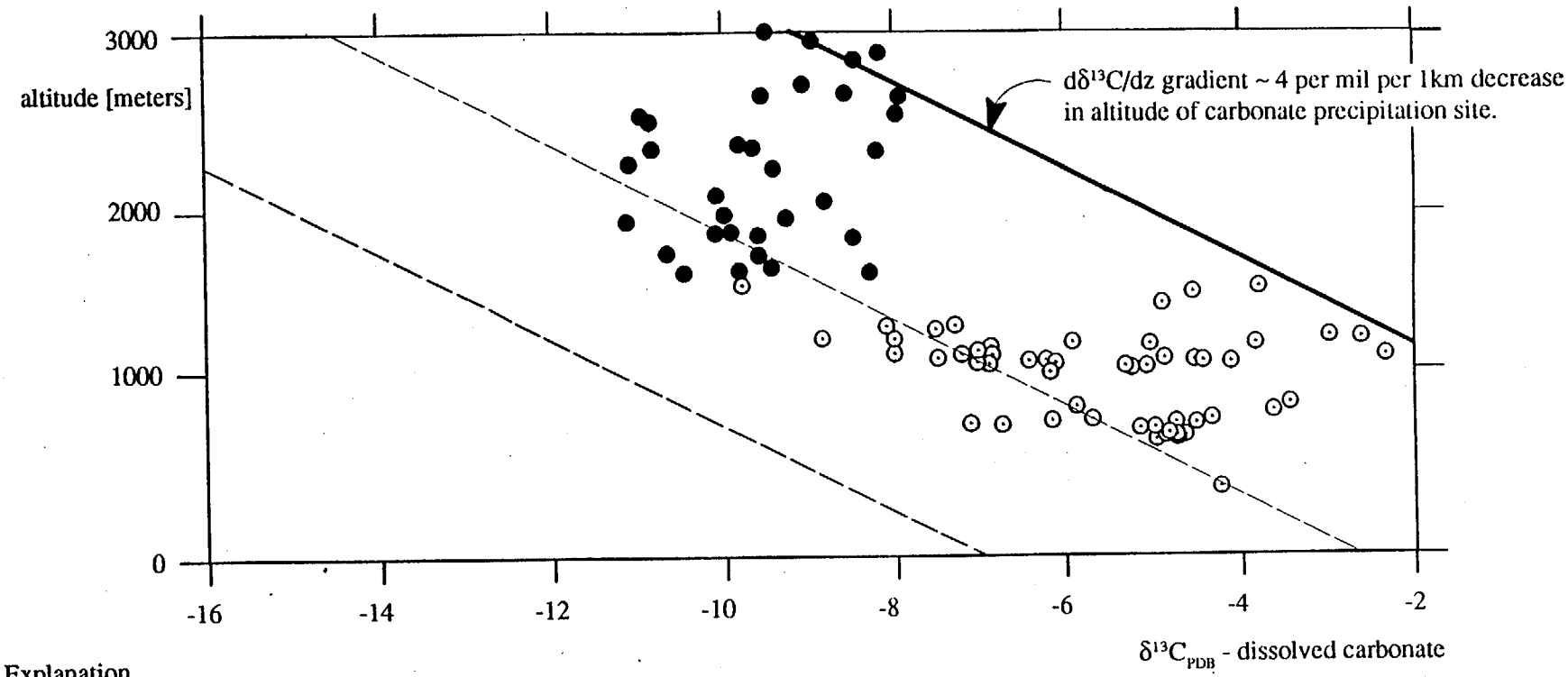
a) for comparison with the local cobble encrustations, the $\delta^{13}C$ vs altitude field, shown between solid and dashed lines, should be shifted by the equilibrium fractionation factor $10^3 \ln\alpha_{CaCO_3 - HCO_3^-} \sim 2$ per mil_{PDB};

b) relative to the observed $\delta^{13}C$ vs altitude field, from the local cobble encrustations, the expected $\delta^{13}C$ 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 $\delta^{13}C$ 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 $\delta^{13}C$ vs altitude gradient, from samples of the *per ascensum* cobble encrustations and distal calcretes, is $d\delta^{13}C/dz \sim 4$ per mil_{PDB} per 1km 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 $\delta^{13}C$ 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

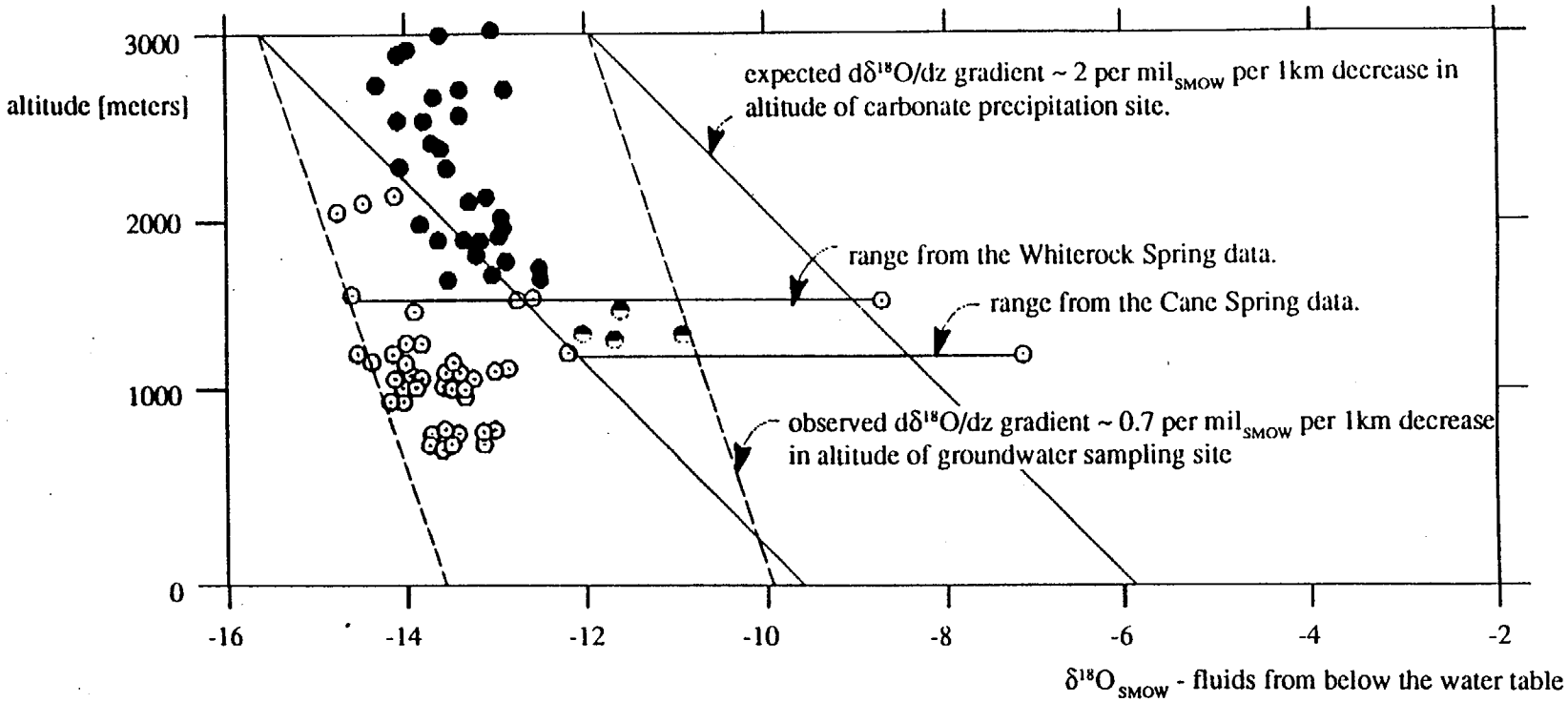
O - groundwater data from the Nevada Test Site hydraulic "source" regions - here, the resident fluids are supersaturated with respect to calcite and, potentially, may discharge at the topographic surface.

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 $\delta^{13}C$ vs altitude gradient and the expected $\delta^{13}C$ 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 $\delta^{18}O$ gradient and field for the *per ascensum* cobble encrustations and distal calcretes.

Figure 25



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
- O - 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 $\delta^{18}O$ vs altitude gradient and field for the *per ascensum* cobble encrustations and distal calcretes.

Note:

a) for comparison with the local cobble encrustation, the $\delta^{18}O$ vs altitude field, shown between two solid lines, should be shifted by the equilibrium fractionation factor $10^3 \ln\alpha_{CaCO_3 - H_2O} \sim 32$ 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 - $d\delta^{18}O/dz = +0.7$ per mil_{SMOW} per 1km decrease in altitude of groundwater sampling site; ii) the observed mean ambient temperature of the topographic surface - $d\delta^{18}O/dz = -1.2$ per mil_{SMOW} per 1km decrease in altitude of carbonate precipitation site; iii) the interpreted oxygen -18 evaporative enrichment - $d\delta^{18}O/dz = +2.5$ per mil_{SMOW} per 1km decrease in altitude of carbonate precipitation site;

d) the value of $d\delta^{18}O/dz = -1.2$ 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 $10^3 \ln\alpha_{CaCO_3 - H_2O} = 2.78 \bullet 10^6 \bullet T^{-2} - 2.89$, from Turi (1986); and

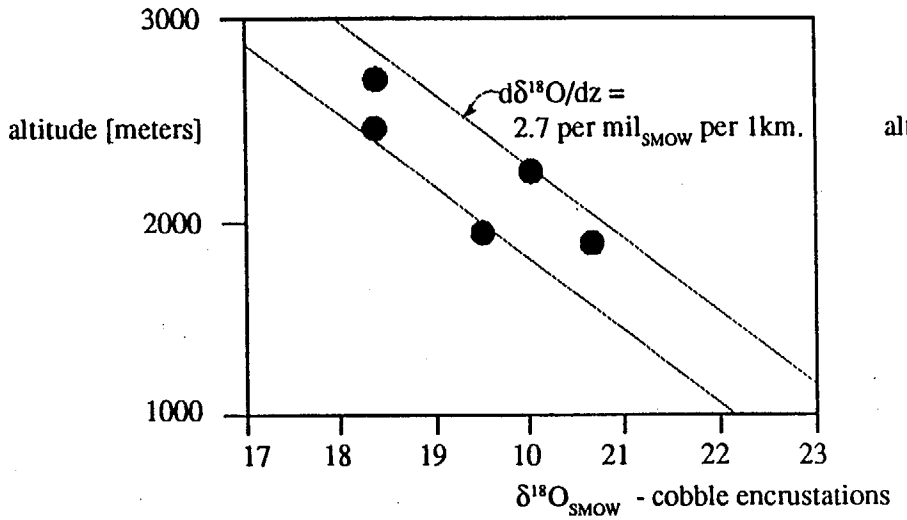
ii) $dT/dz = 5$°Celsius per 1km decrease in altitude of the topographic surface - Major (1977) gives the local $dT/dz = 5.4$°Celsius per 1km;

e) the value of $d\delta^{18}O/dz = +0.7$ 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;

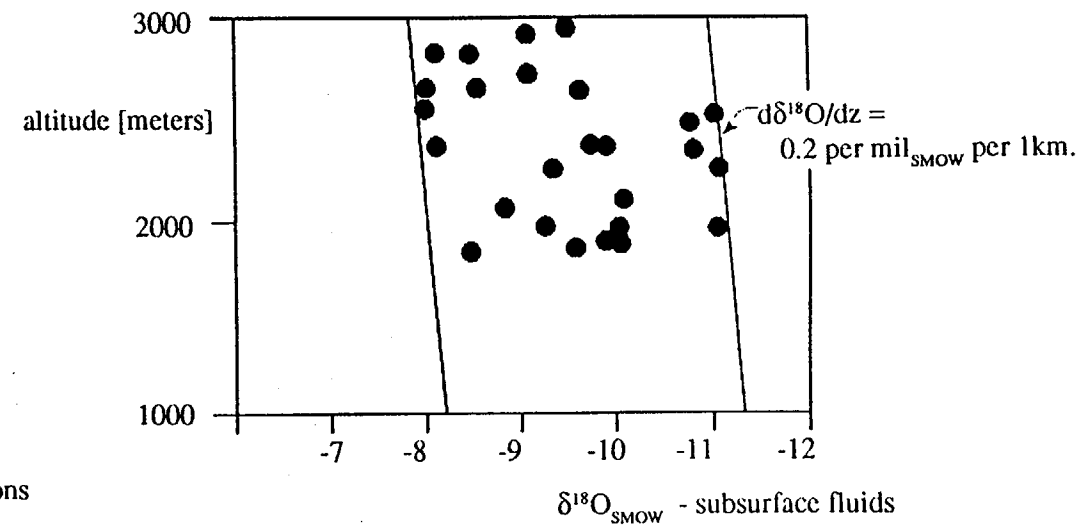
f) the value of $d\delta^{18}O/dz = +2.5$ per mil_{SMOW} per 1km, reflecting the altitude dependence of the oxygen -18 evaporative enrichment, is computed in Figure 26a; and

g) both the expected $\delta^{18}O$ vs altitude gradient and the expected $\delta^{18}O$ 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 $\delta^{18}O$ 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:

a) isotopic data, as shown, are from Hershey (1989) and are computed from Quade et al. (1988);

b) 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

c) the interpreted value for the oxygen -18 evaporative enrichment vs altitude gradient, from samples of the groundwater cobble encrustations, is $d\delta^{18}O/dz = +2.5$ per mil_{SMOW} per 1km 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 $\delta^{18}O$ vs altitude gradient and field for the *per ascensum* cobble encrustations and distal calcretes.

- Values of the isotopic compositions vs altitude gradients, observed based on samples of the local cobble encrustations, are:

  i) $d\delta^{13}C/dz \sim +4$ per mil_{PDB} per 1km decrease in altitude of the deposit sampling site; and
  ii) $d\delta^{18}O/dz \sim +2$ per mil_{SMOW} per 1km decrease in altitude of the deposit sampling site.

- For the *per ascensum* cobble encrustations and distal calcretes, the expected value of the carbon -13 content vs altitude gradient is $d\delta^{13}C/dz \sim +4$ per mil_{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.

- For the *per ascensum* cobble encrustations and distal calcretes, the expected value of the oxygen -18 content vs altitude gradient is $d\delta^{18}O/dz \sim +2$ per mil_{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.

- In addition to yielding the comparable values of the isotopic gradients, the calcium-rich subsurface fluids also carry values of the $\delta^{18}O$ and $\delta^{13}C$ 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.

- Evidently, the observation made by Quade and Cerling (1990) that the $\delta^{13}C$ and $\delta^{18}O$ 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 descensum* process; or

  ii) the assumption of the *per descensum* origin for the local cobble encrustations, as proposed by Quade et al. (1988), Quade et al. (1989), and Quade and Cerling (1990), is **FALSE.**

- The above conclusion, if considered together with the conclusion from Figure 19, indicates that the *per descensum* 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.

- Evidently, the observation made by Quade and Cerling (1990), that the $\delta^{13}C$ and $\delta^{18}O$ 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 descensum* process; or

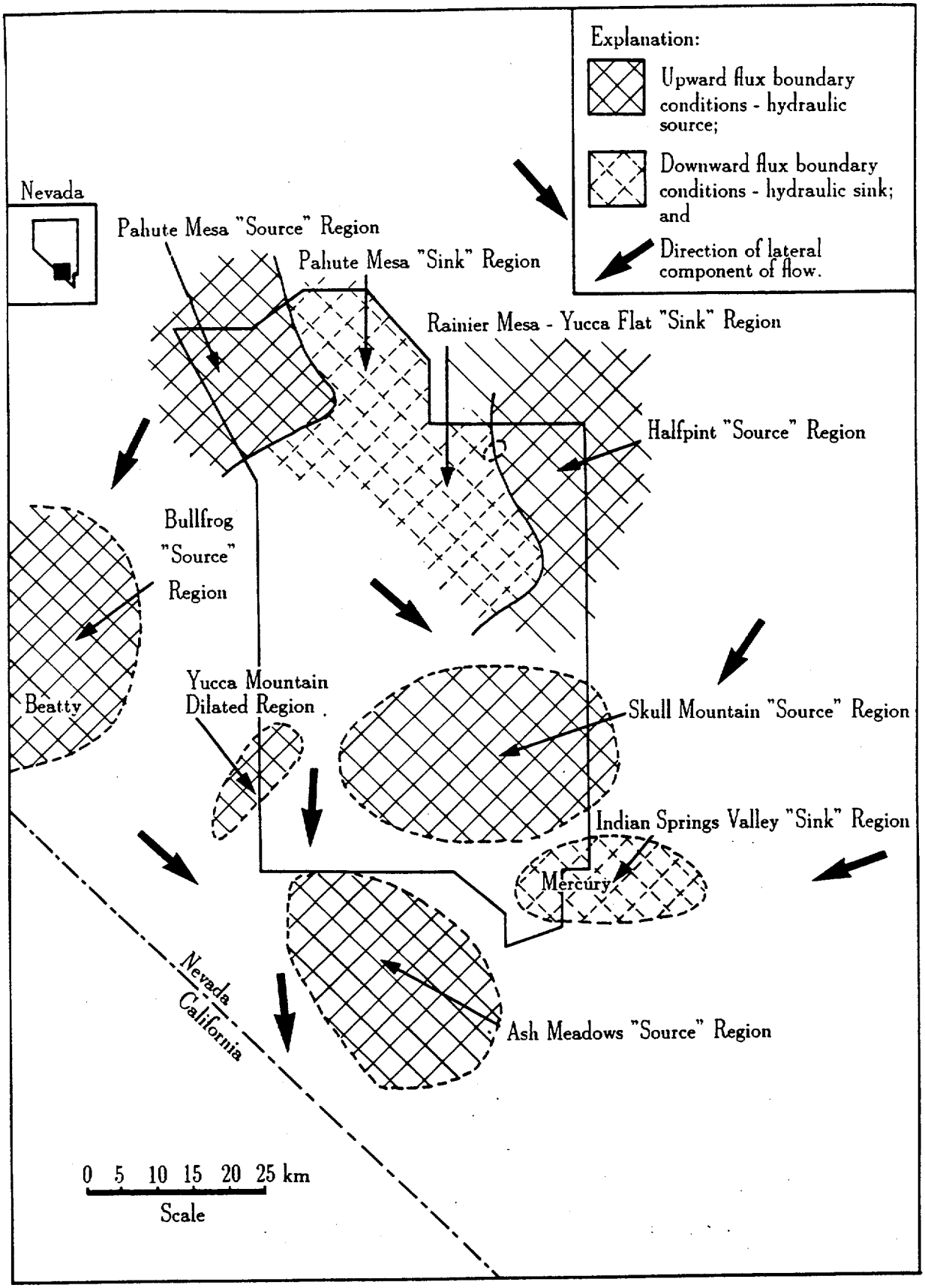
  ii) the assumption of the *per descensum* origin for the local cobble encrustations, as proposed by Quade et al. (1988); Quade et al. (1989); and Quade and Cerling (1990), is **FALSE.**

- The above conclusion, if considered together with the conclusion from Figure 19, indicates that the *per descensum* 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.

Figure 30

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°C/km.

Lateral geothermal gradient dT/dx 5°C/1 mile.

Possible upper thermal boundary layers.

Water chemistry:
$Ca^{++} + Mg^{++}$ from 1 to 40 ppm
$Na^{+} + K^{+}$ from 40 to 250 ppm
$CL^{-} + SO^{-}_{4}$ 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 ~ 30°C/km.

Water chemistry:
$Ca^{++} + Mg^{++}$ from 4 to 60 ppm
$Na^{+} + K^{+}$ from 35 to 95 ppm
$CL^{-} + SO_4^{-}$ from 15 to 70 ppm
HCO_3^{-} from 60 to 450 ppm.

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

Figure 31a

Rainier Mesa - Yucca Flat Hydraulic "Sink" Region.

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

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

Water chemistry:

$Ca^{++} + Mg^{++}$ from 2 to 40 ppm
$Na^{+} + K^{+}$ from 22 to 65 ppm
$CL^{-} + SO_4^{--}$ from 11 to 40 ppm
HCO_3^{-} from 50 to 230 ppm.

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

Halfpint Hydraulic "Source" Region.

Vertical hydraulic gradient dh/dz - unknown.

Vertical geothermal gradient $dT/dz \simeq 45°C/km$.

Water chemistry:

$Ca^{++} + Mg^{++}$ from 55 to 400 ppm
$Na^{+} + K^{+}$ from 50 to 270 ppm
$CL^{-} + SO^{4--}$ 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.

Figure 31b

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 = 45-50°C/km.}

Water chemistry: $Ca^{++} + Mg^{++}$ from 4 to 470 ppm
$Na^{+} + K^{+}$ from 47 to 450 ppm
$CL^{-} + SO_4^{-}$ 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 boreholes.

Indian Springs Valley Hydraulic "Sink" Region.

Vertical hydraulic gradient dh/dz - unknown.

Vertical geothermal gradient dT/dz < 20°C/km.

Water chemistry: $Ca^{++} + Mg^{++}$ from 16 to 75 ppm
$Na^{+} + K^{+}$ from 6 to 40 ppm
$CL^{-} + SO_4^{-}$ 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.

Figure 31c

Ash Meadows Hydraulic "Source" Region.

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

Vertical geothermal gradient dT/dz ~ 40°C/km.

Water chemistry: $Ca^{++} + Mg^{++}$ from 12 to 270 ppm
$Na^{+} + K^{+}$ from 20 to 300 ppm
$CL^{-} + SO_4^{--}$ 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 Δ h - unknown).

Vertical geothermal gradient dT/dz - unknown.

Water chemistry: $Ca^{++} + Mg^{++}$ from 8 to 50 ppm
$Na^{+} + K^{+}$ from 100 to 350 ppm
$CL^{-} + SO_4^{--}$ 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 ~ 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°C/km in deeper undilated regions; dT/dz = 20-30°C/km immediately below the water table.

Water chemistry:	dilated zone	$Ca^{++} + Mg^{++}$ from 2 to 20 ppm $Na^{+} + K^{+}$ from 40 to 120 ppm $CL^{-} + SO_4^{--}$ from 21 to 55 ppm HCO_3^{-} from 100 to 300 ppm.
	undilated zone	$Ca^{++} + Mg^{++}$ 140 ppm $Na^{+} + K^{+}$ 162 ppm $CL^{-} + SO_4^{--}$ 188 ppm HCO_3^{-} 569 ppm.

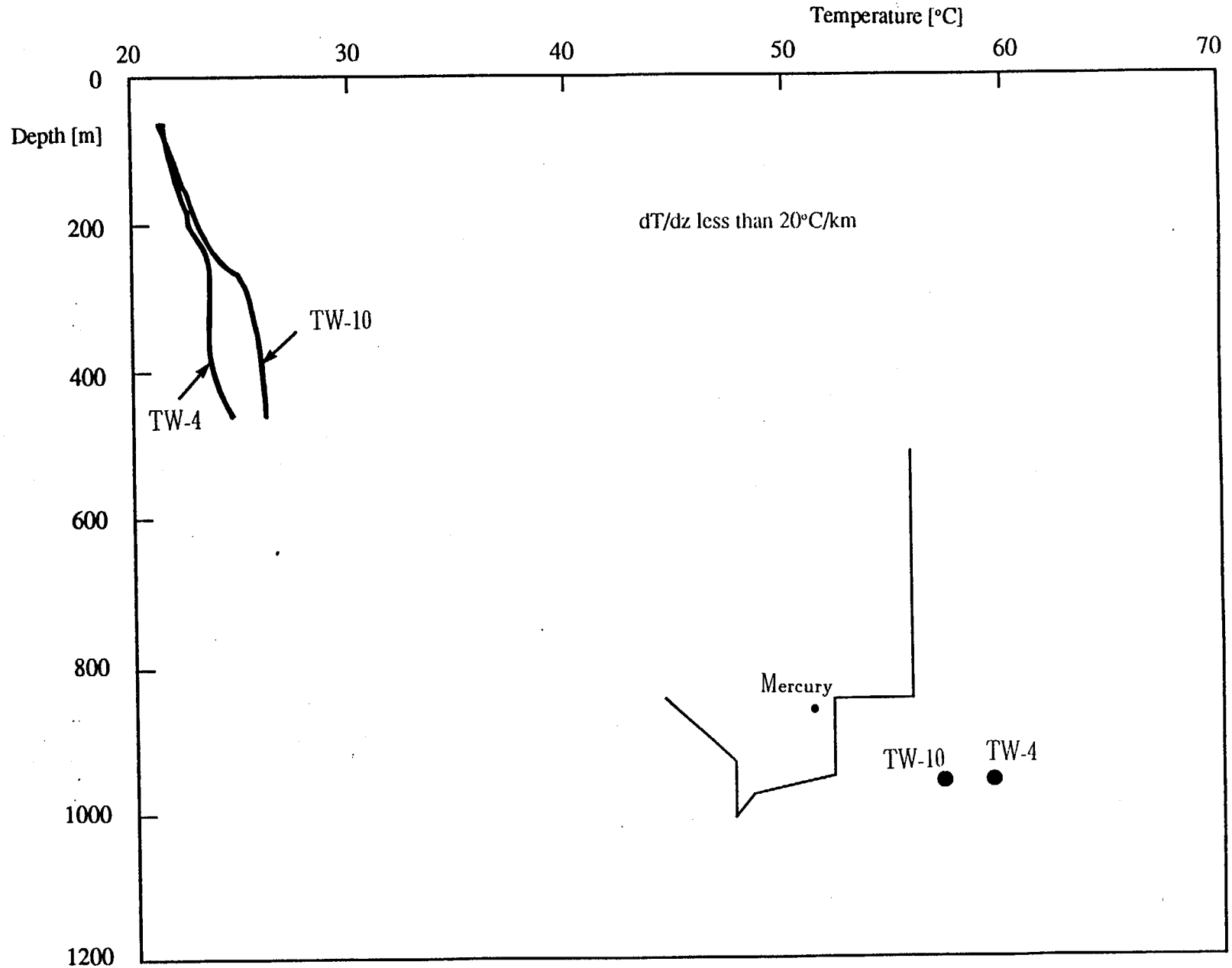
Note: a) local values of $\sigma_c \longrightarrow O$, (indicating severe dilation) extend to a maximum depth of about ~ 1300 m; and

b) the Yucca Mountain area is a source - sink hybrid.

The geothermal, hydraulic, and geochemical data used is 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).}

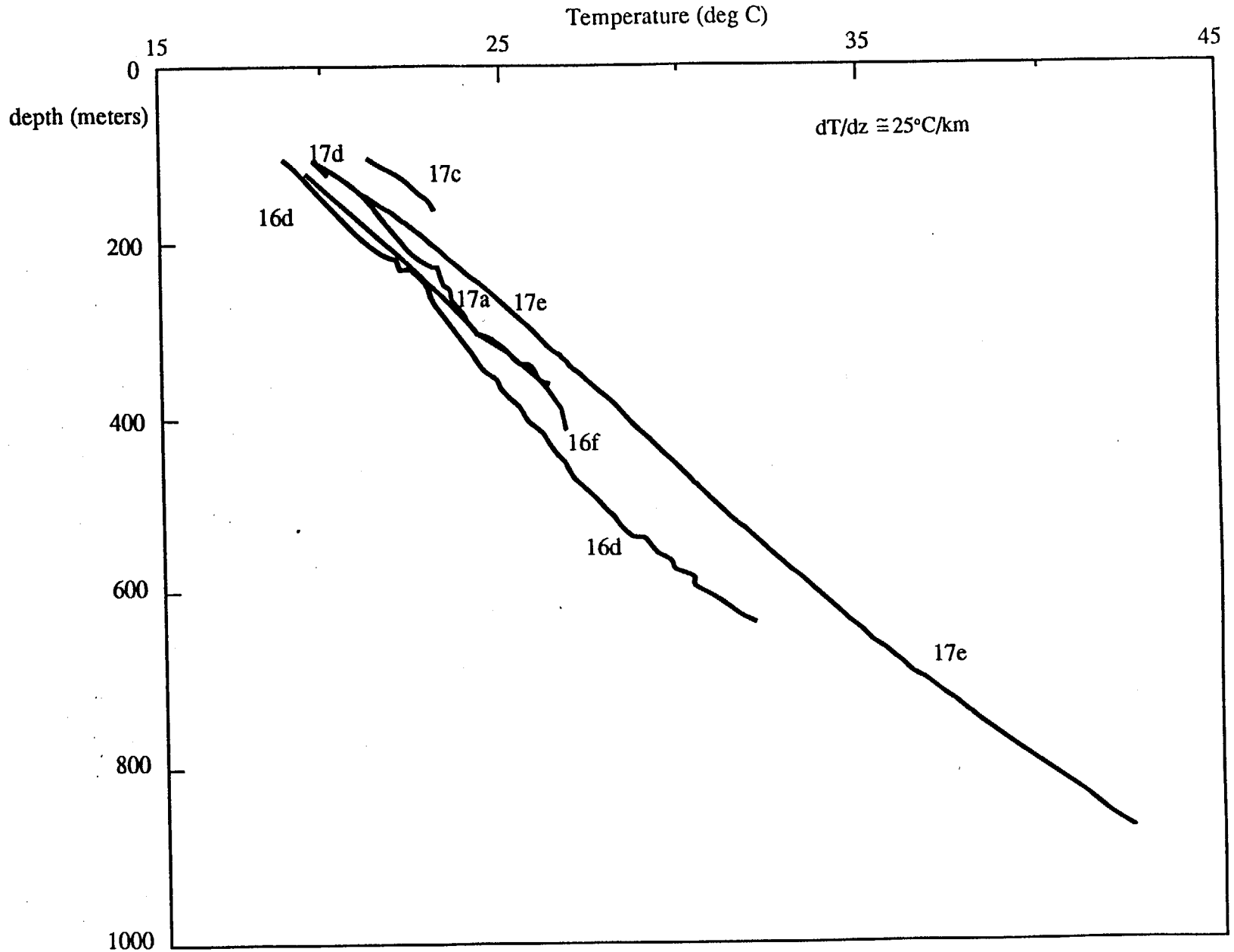
Boundary conditions. Interpretation bases. The Nevada Test Site hydrosphere.

Figure 31e



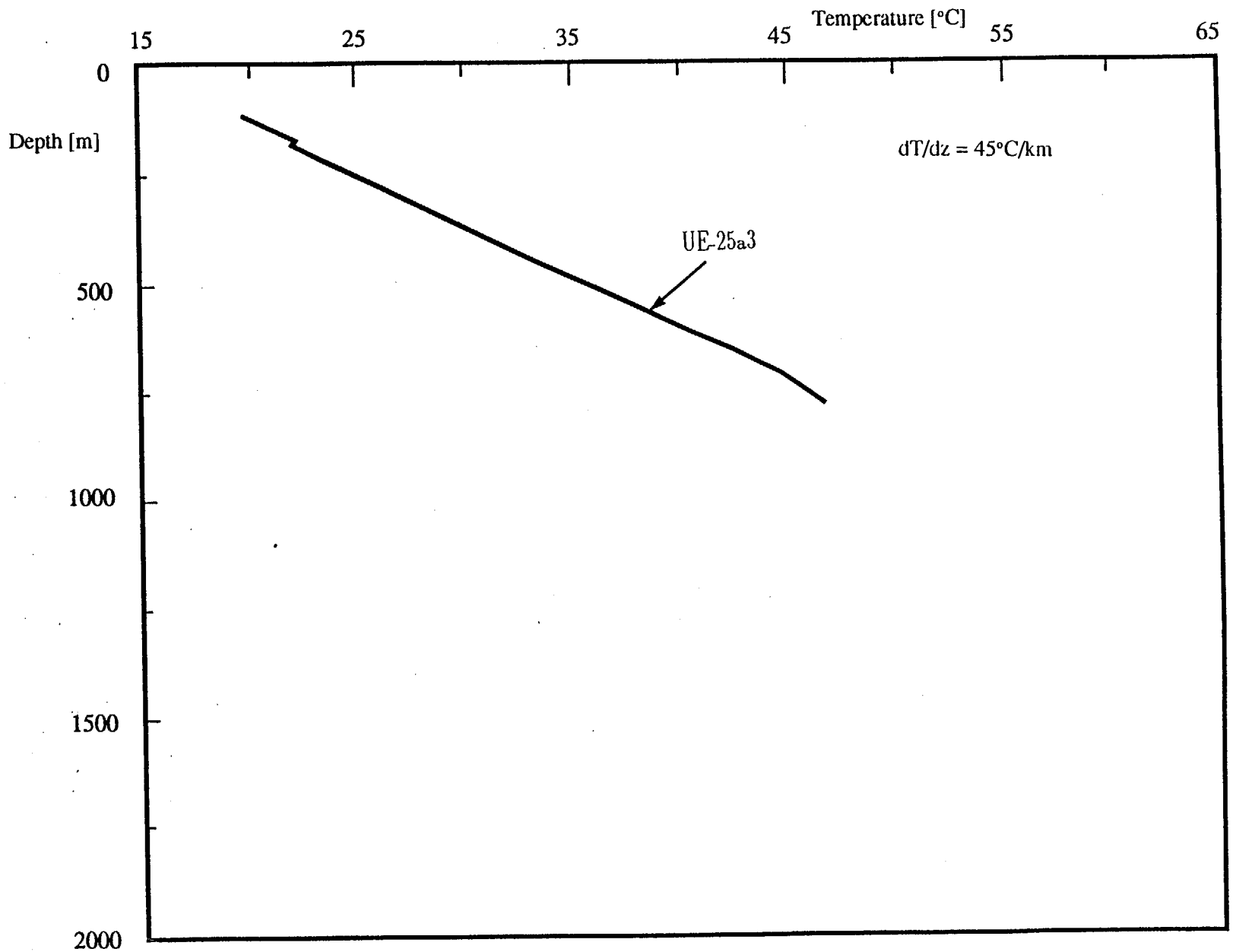
Downhole temperature profiles. Southern Indian Springs Valley "sink" region. From Sass and Lachenbruch, 1982.

Figure 32a



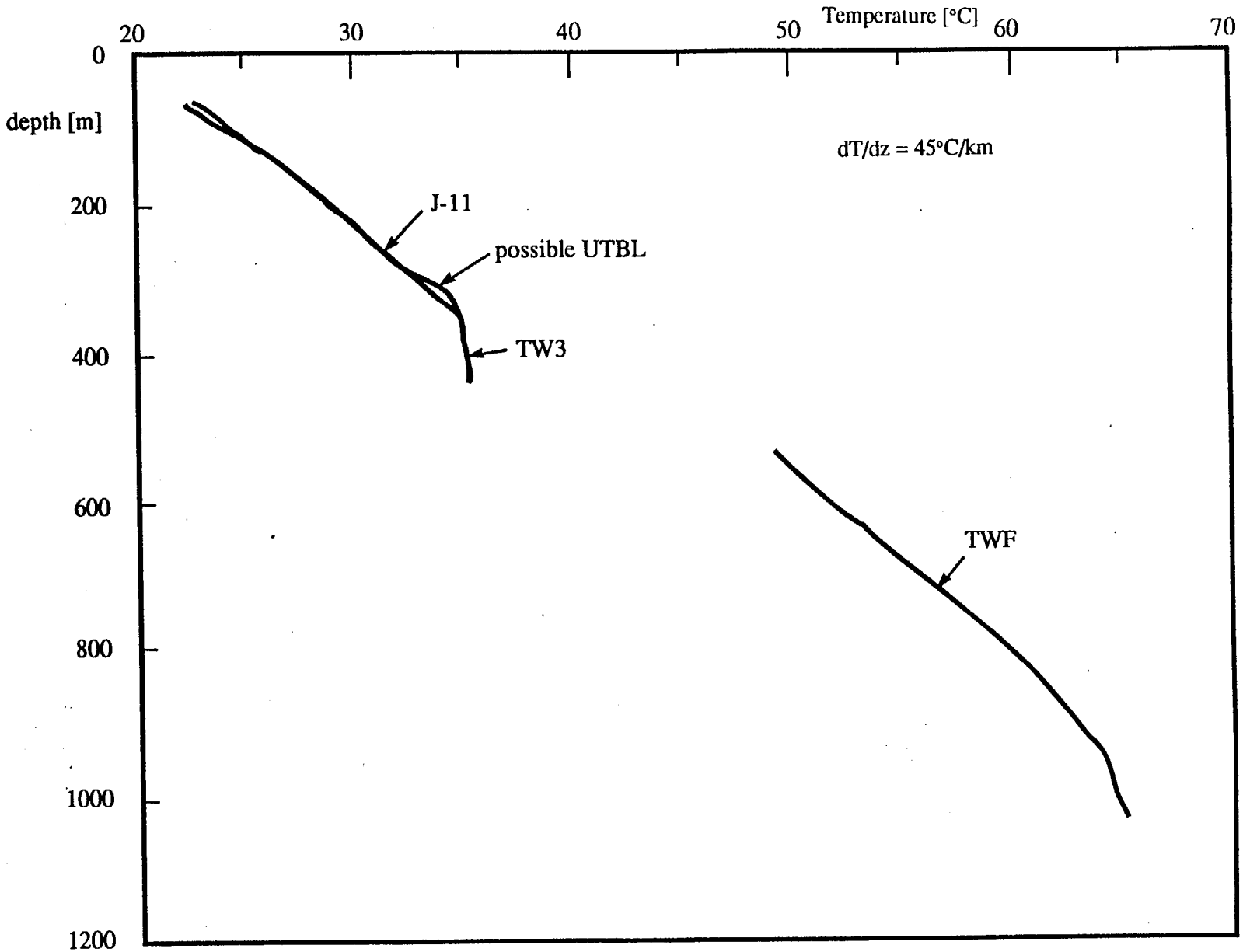
Downhole temperature profiles. Rainier Mesa - Yucca Flat hydraulic "sink" regions. From Sass and Lachenbruch, 1982.

Figure 32b



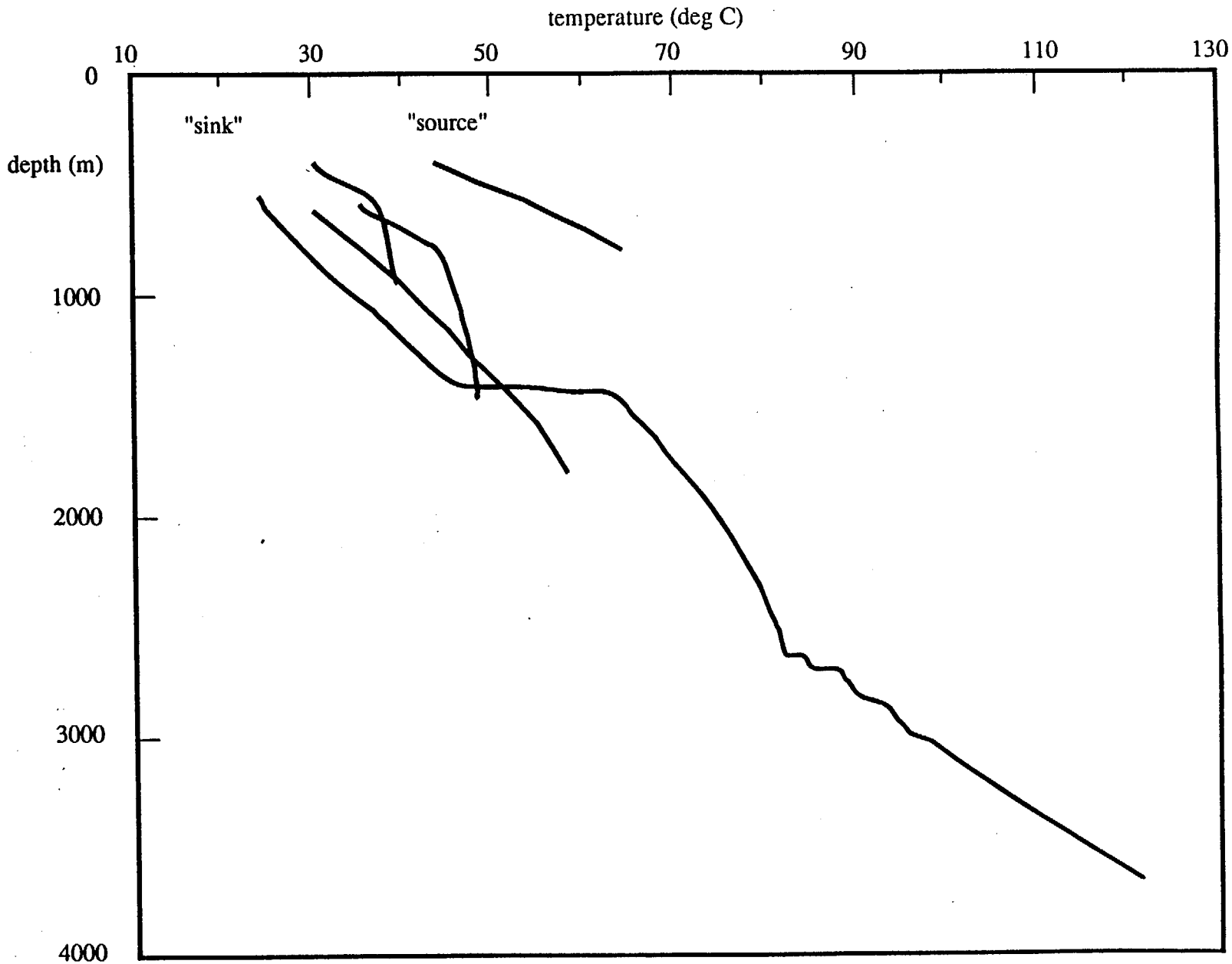
Downhole temperature profile. Borehole UE-25a3. North-western margin of the Skull Mountain hydraulic "source" region. From Sass and Lachenbruch, 1982.

Figure 32c



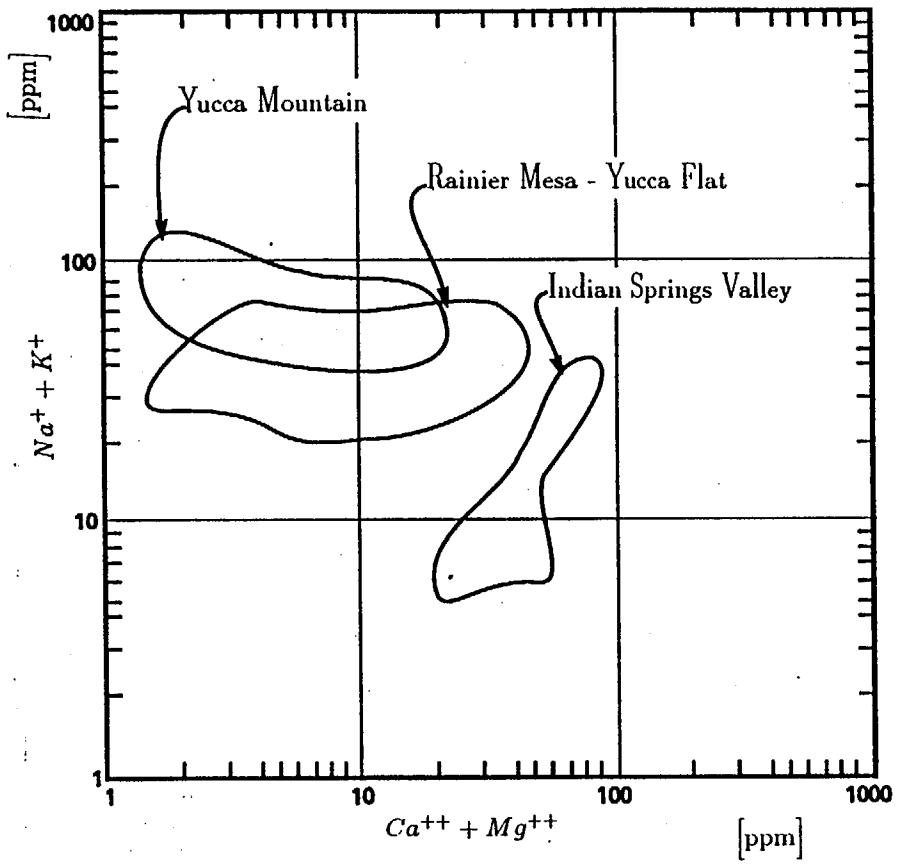
Downhole temperature profiles. Boreholes J-11, TWF, and TW3. Skull Mountain hydraulic "source" region. From Sass and Lachenbruch, 1982.

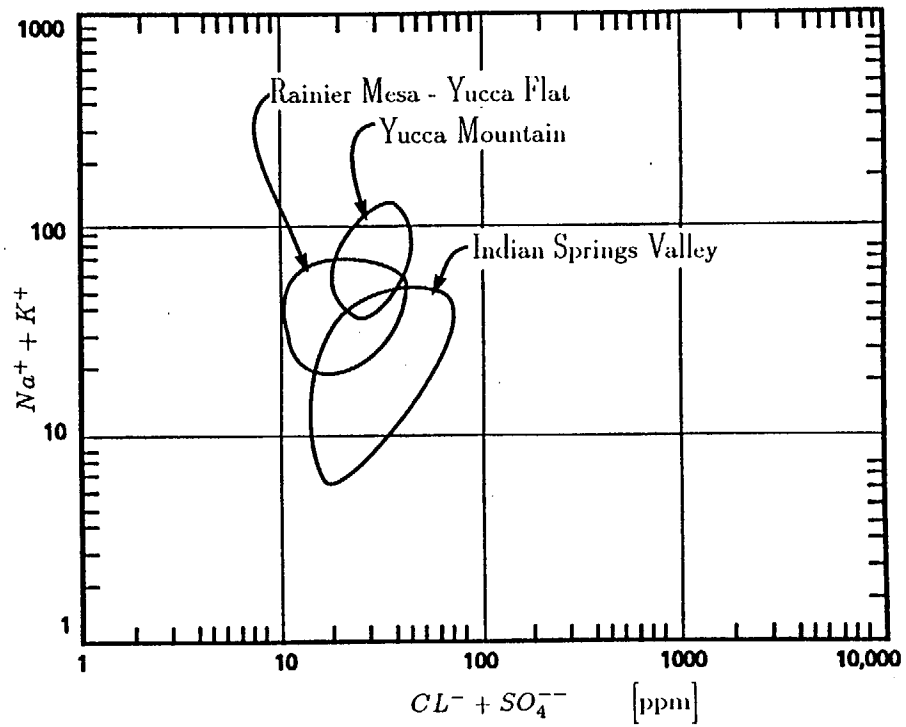
Figure 32d



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

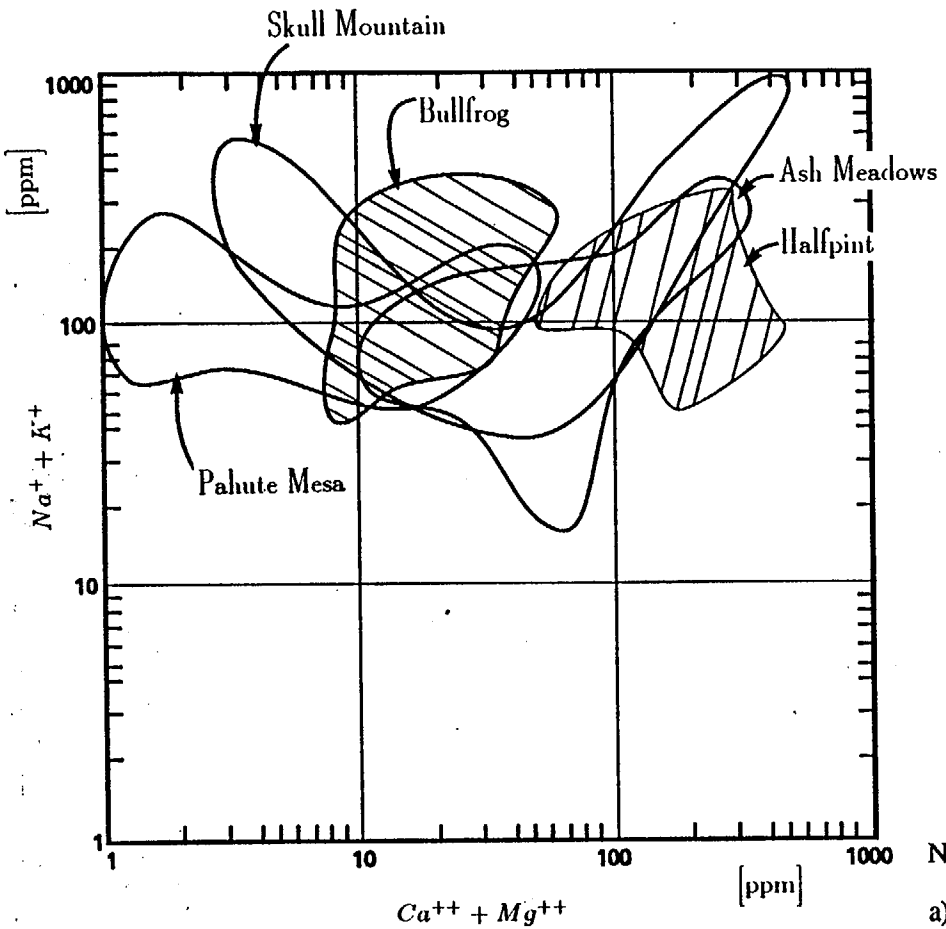
Figure 32c

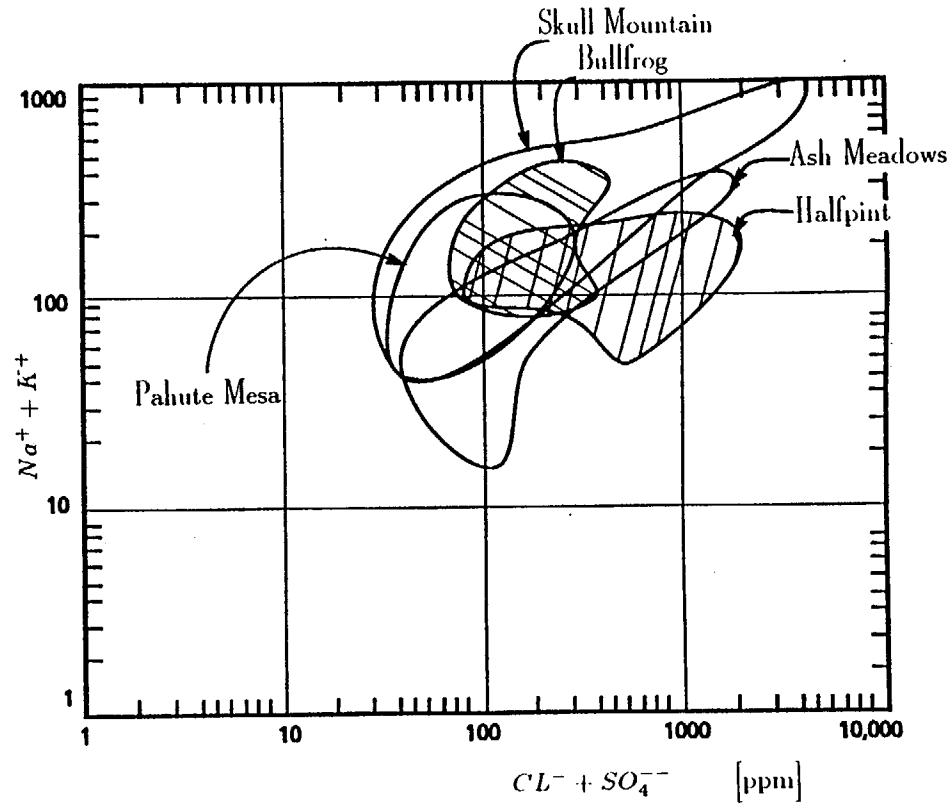




Groundwater chemistry. Hydraulic "sink" and dilated regions. The Nevada Test Site hydrosphere.

Figure 33a





Note:

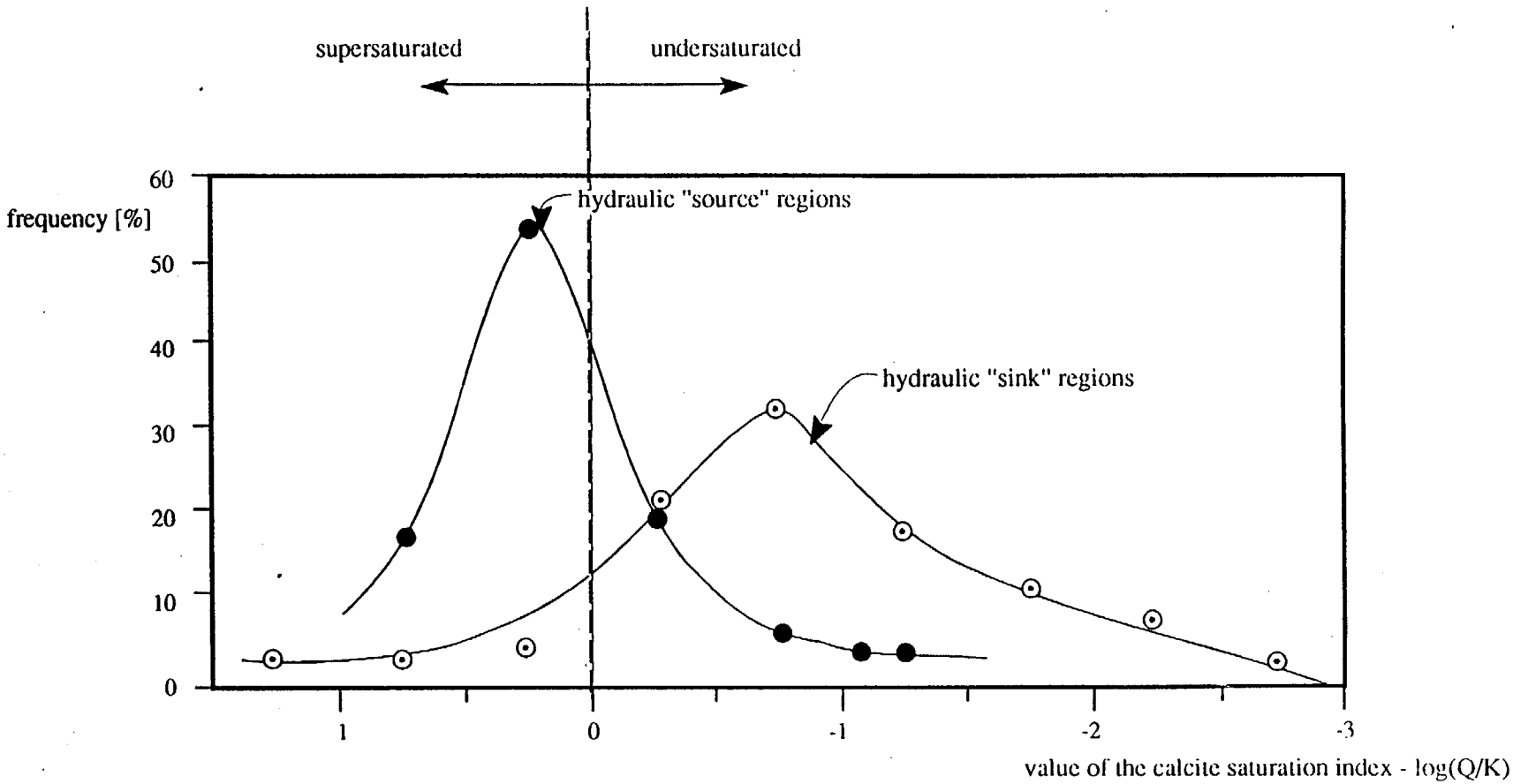
a) fluids result from intermixing of at least two end-members;

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.

Figure 33b



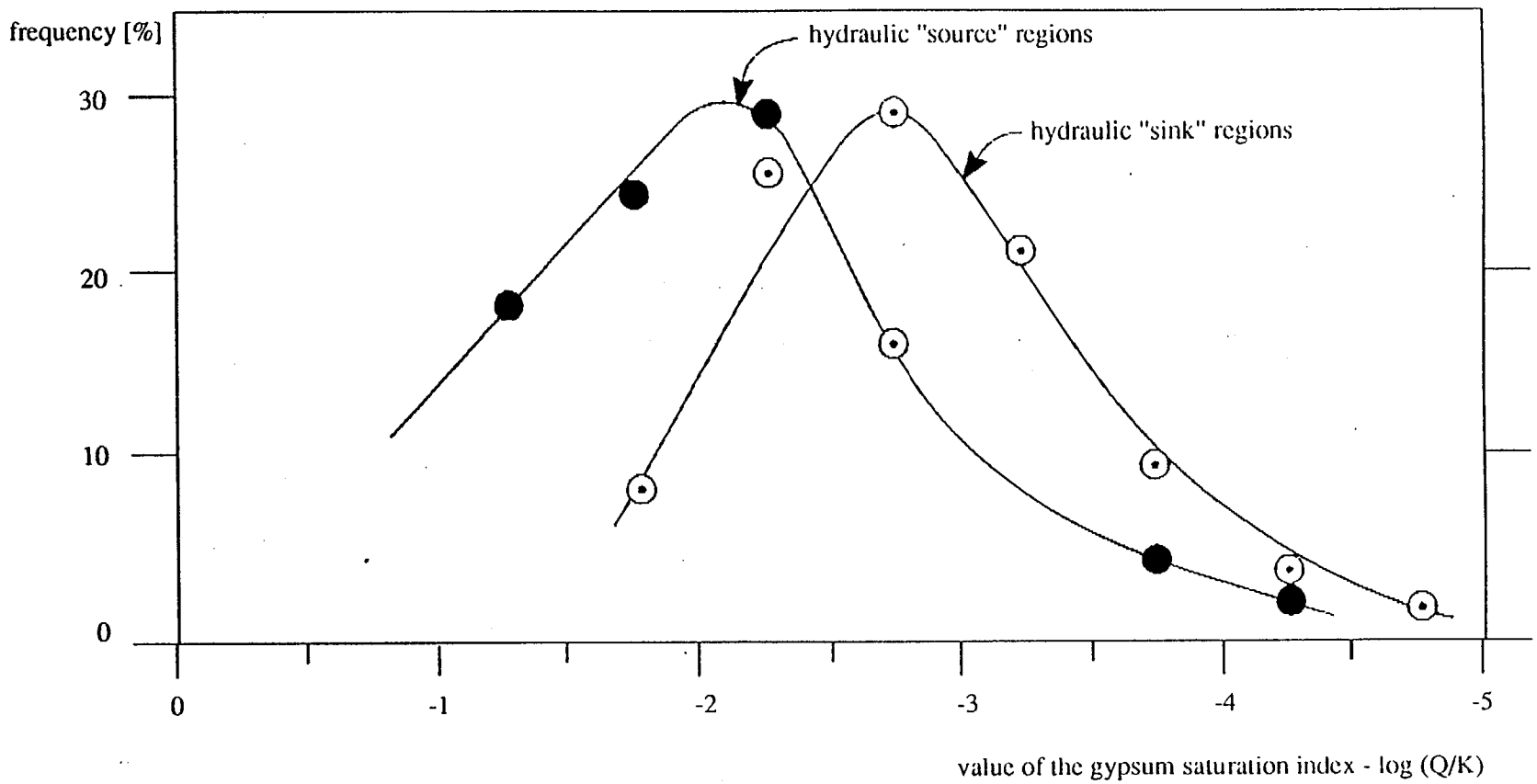
Note:

a) log (Q/K) data are from Kerrisk (1987);

b) N=52 for hydraulic "source" regions; N=64 for hydraulic "sink" regions; and

c) 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.



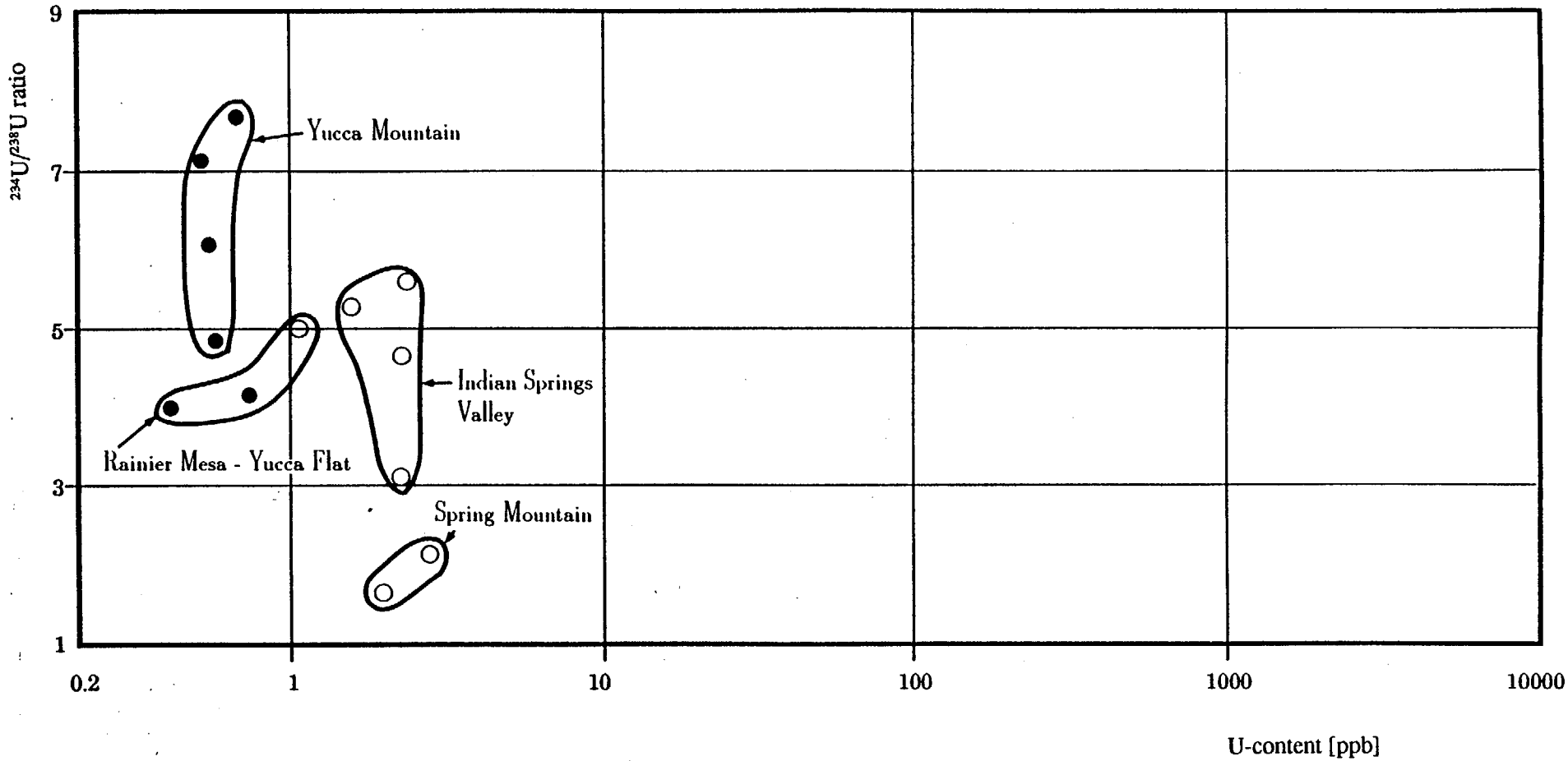
Note:

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.

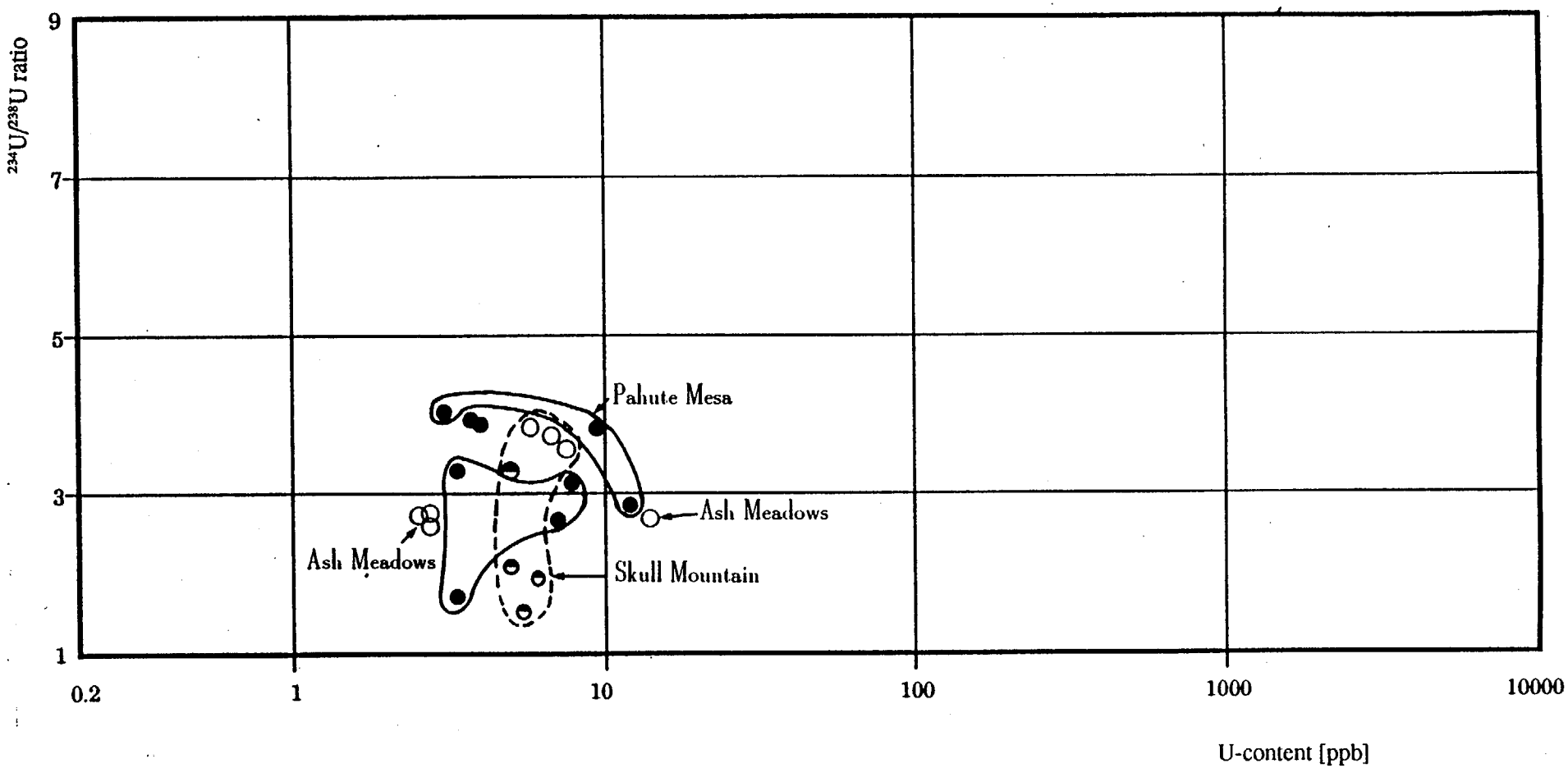


Explanation:

● - sample represents fluids from the tuff pile; and

○ - sample represents fluids from the Paleozoic carbonates.

$^{234}U/^{238}U$ 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

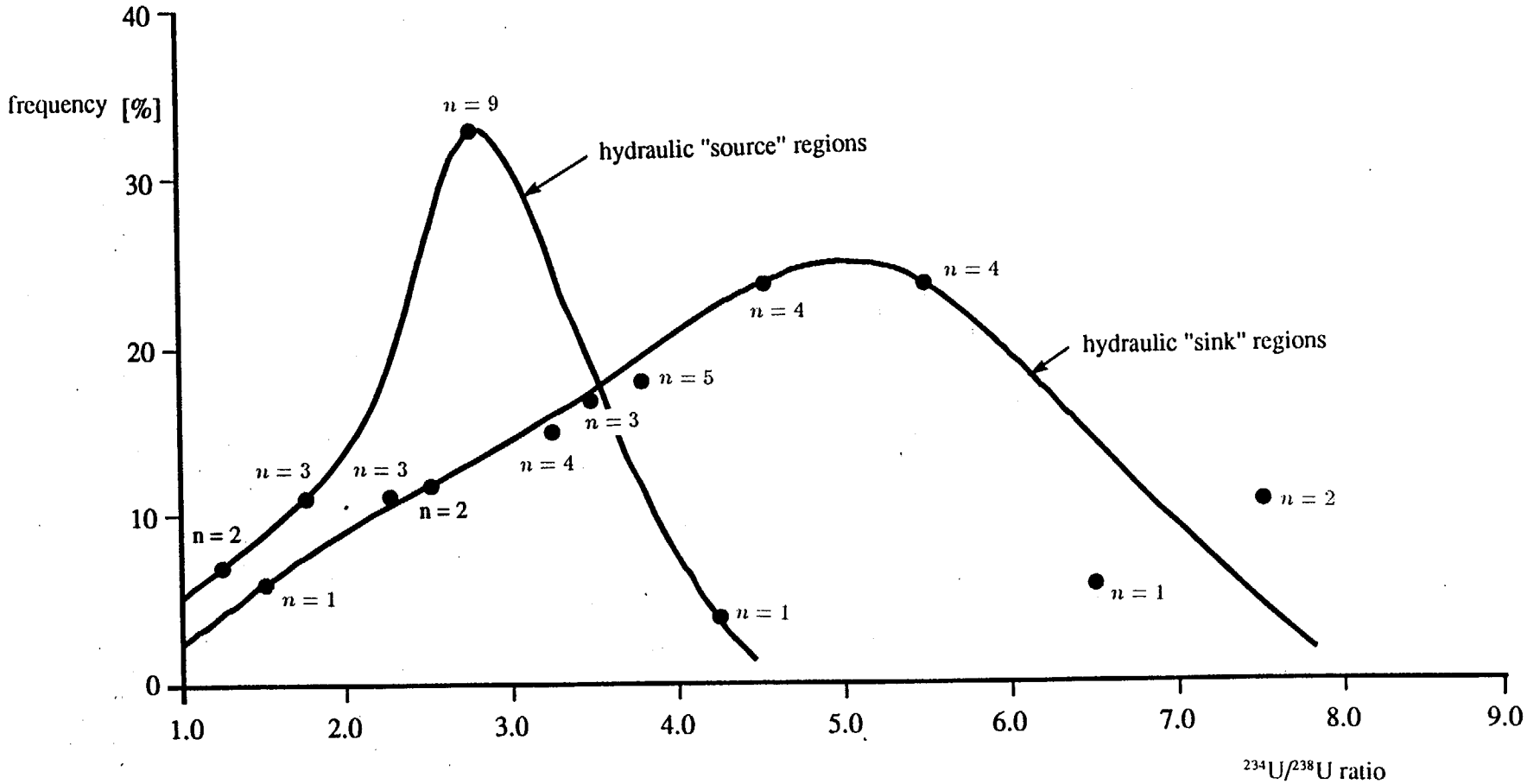
ⓞ - sample represents fluids from alluvium.

Note:

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

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

Figure 37

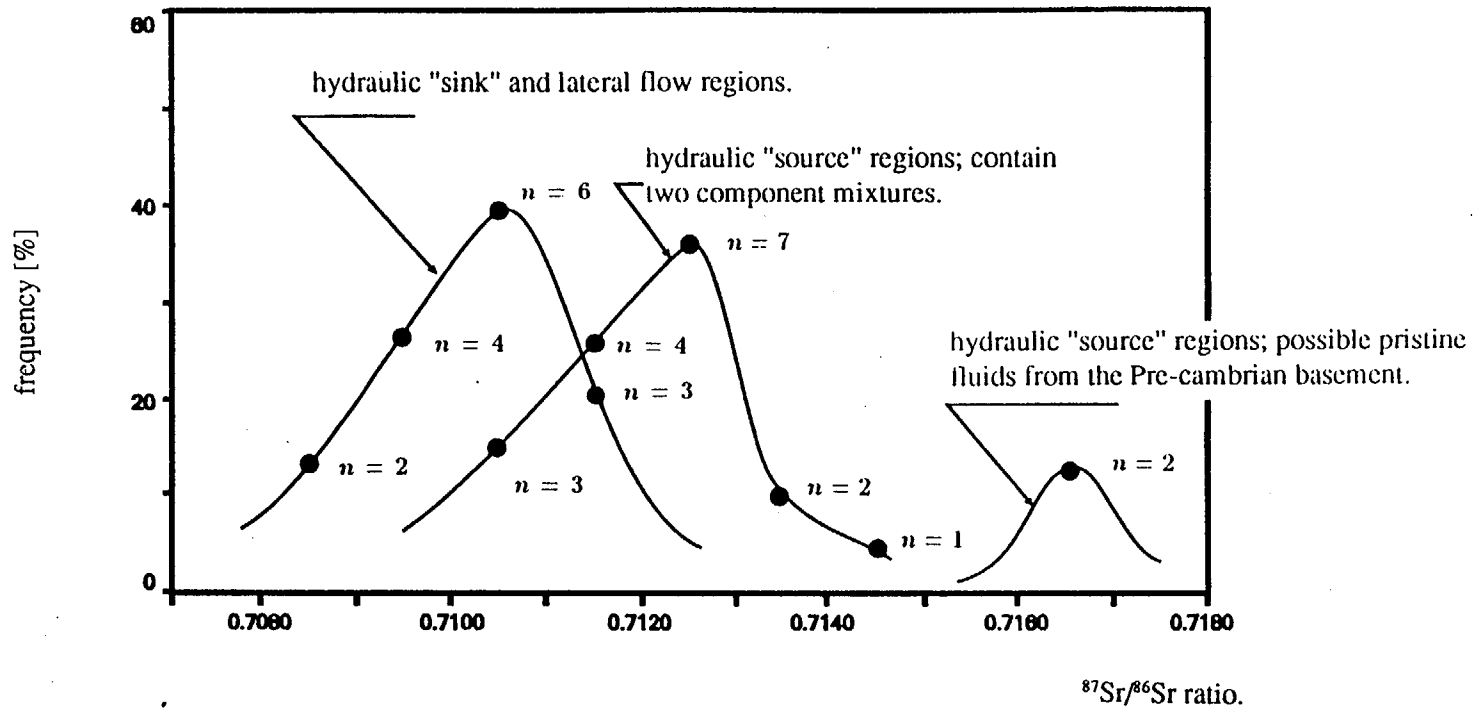


Note:

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.

Figure 39

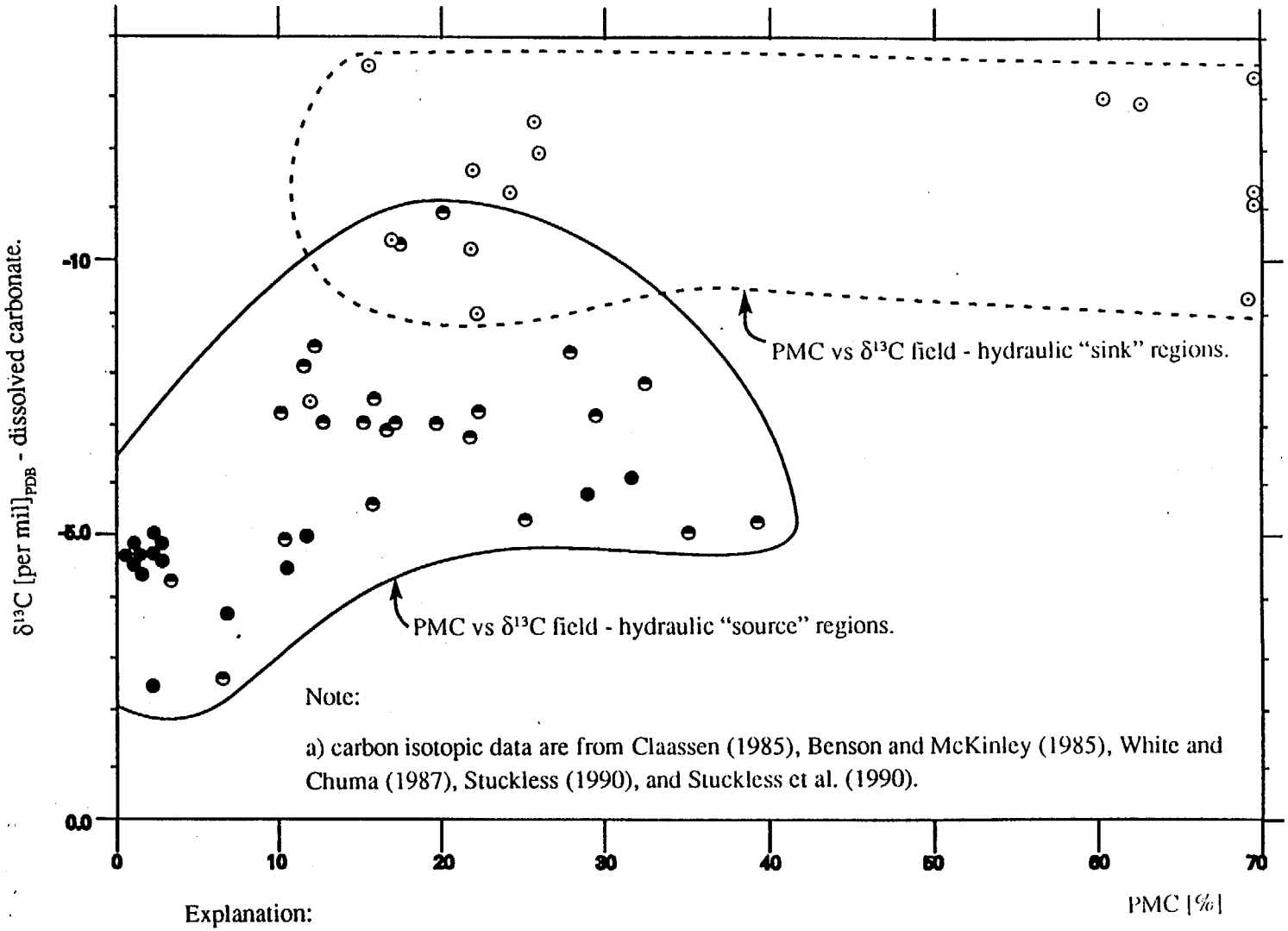


Note:

a) total number of samples n = 15 for the hydraulic "sink" regions, and n = 19 for the hydraulic "source" regions; and

b) isotopic data are from Stuckless (1990).

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

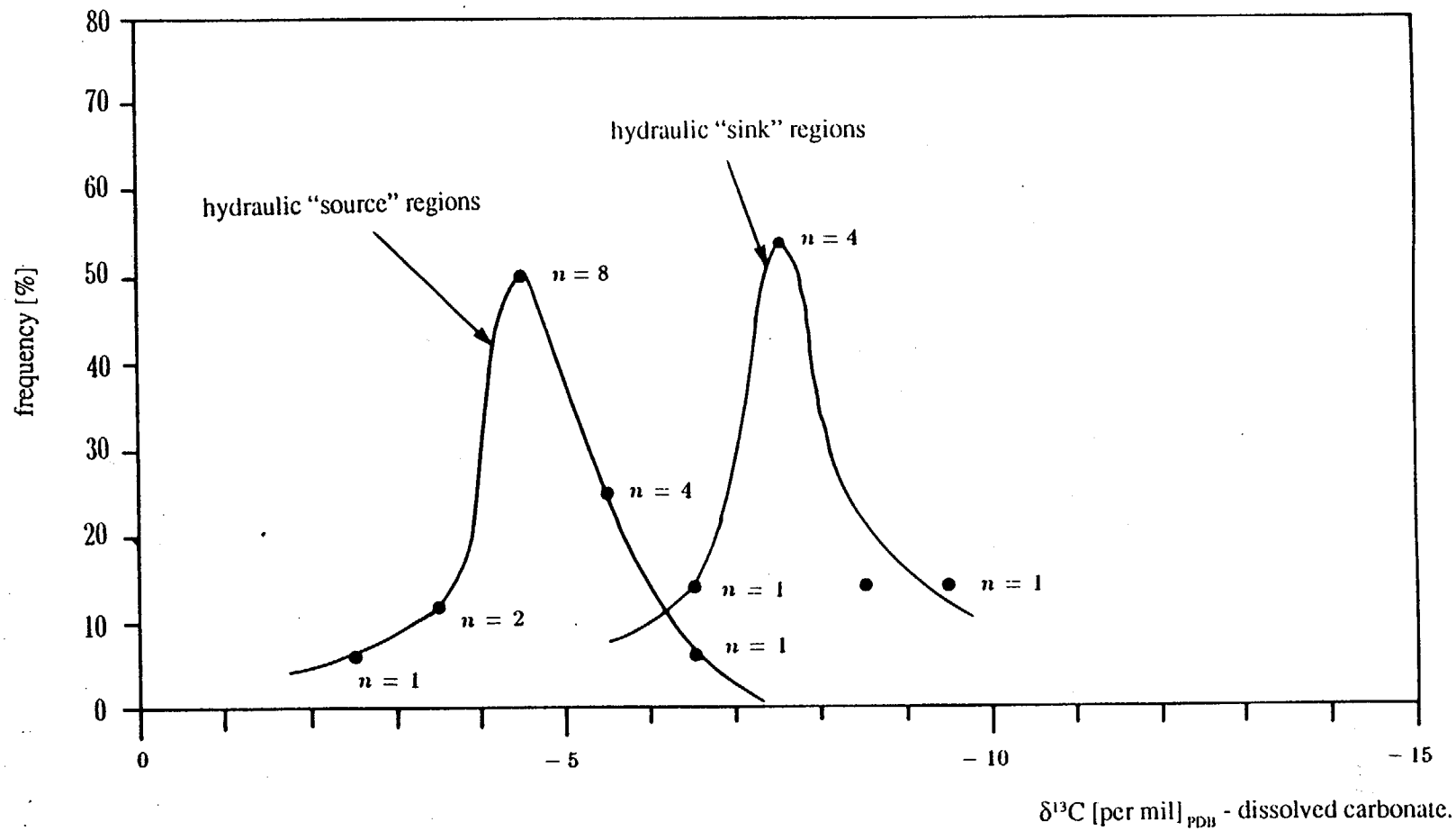


Explanation:

- ○ - 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.

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

Figure 40

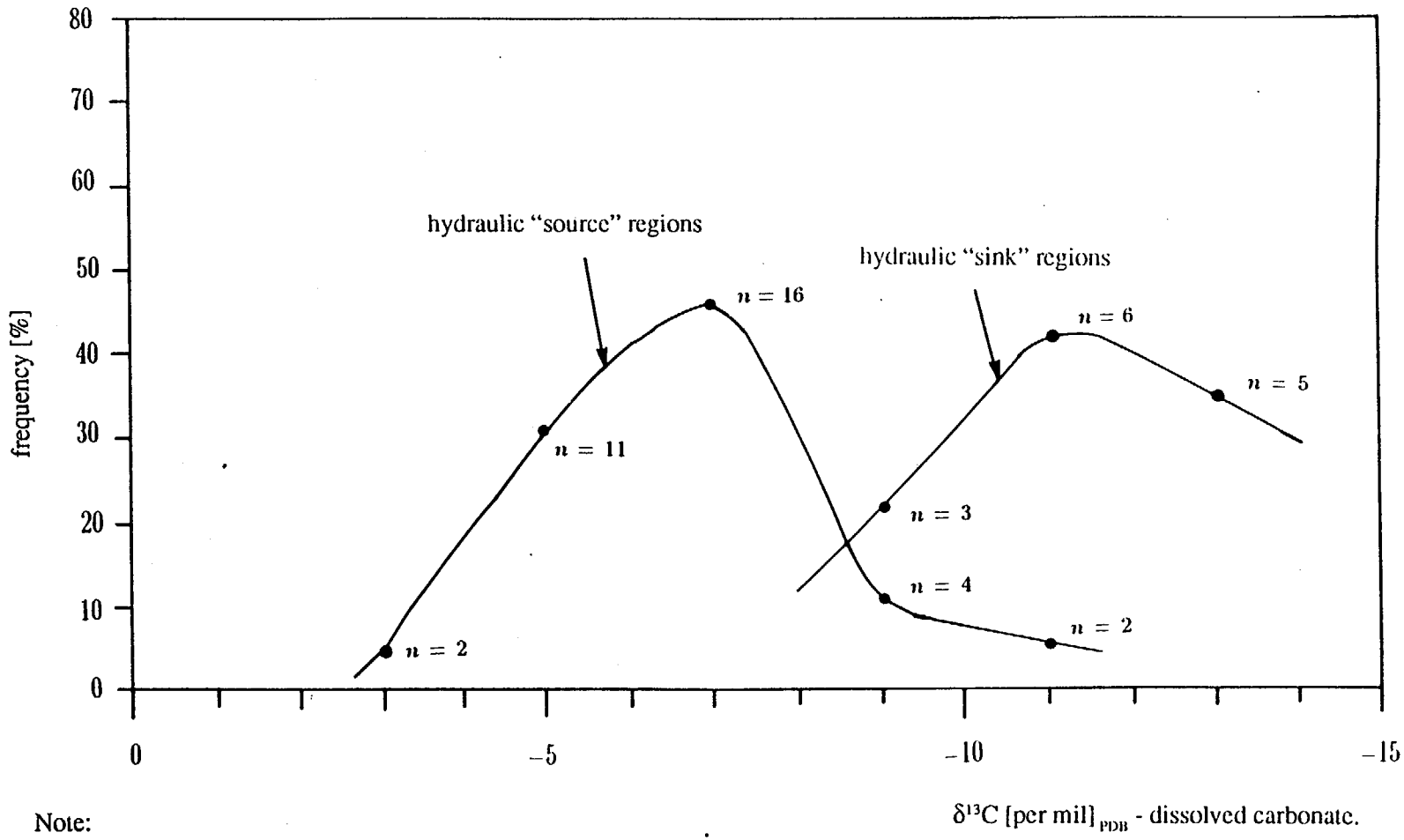


Note:

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.

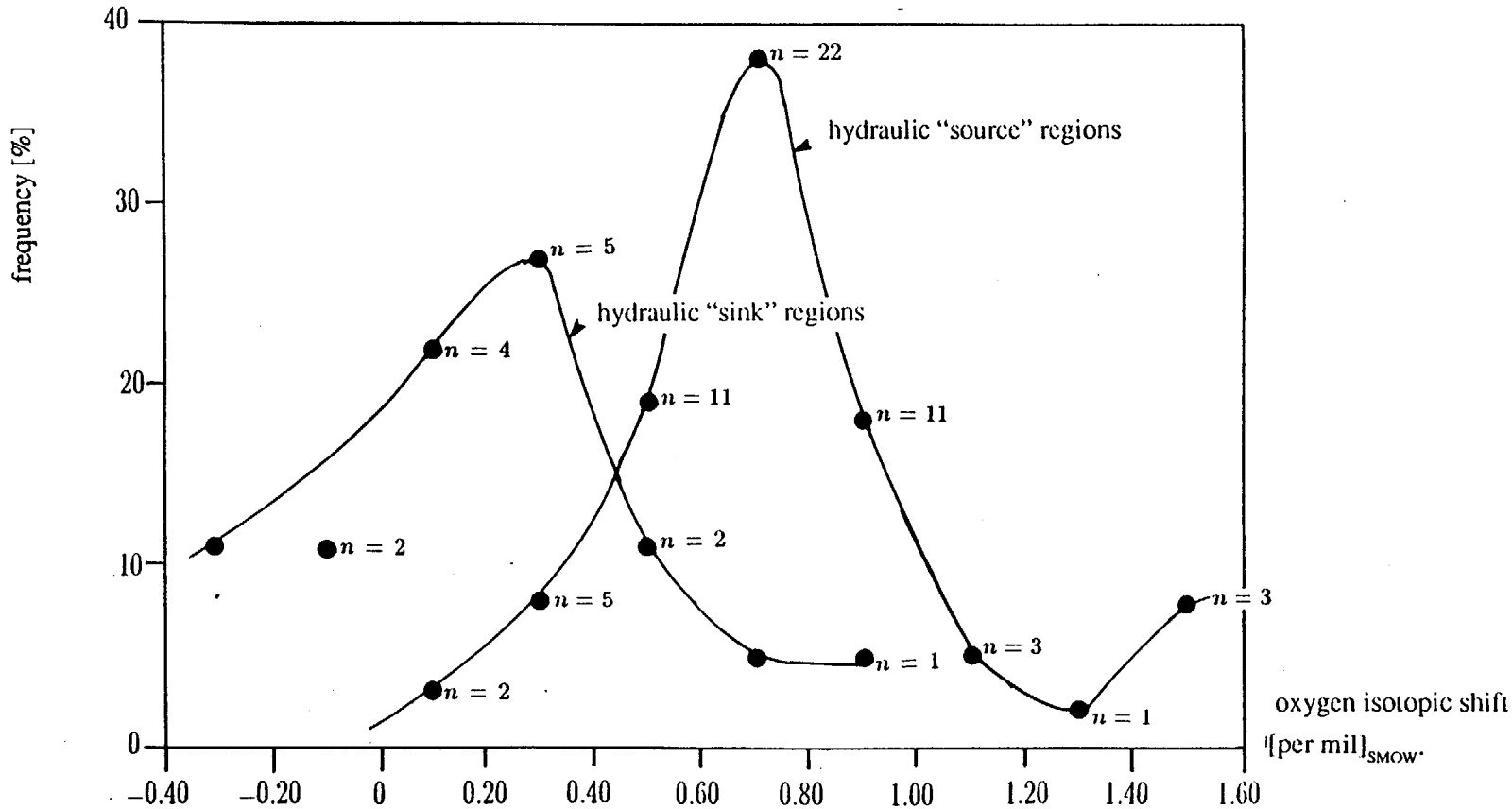
Figure 41



Note:

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

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



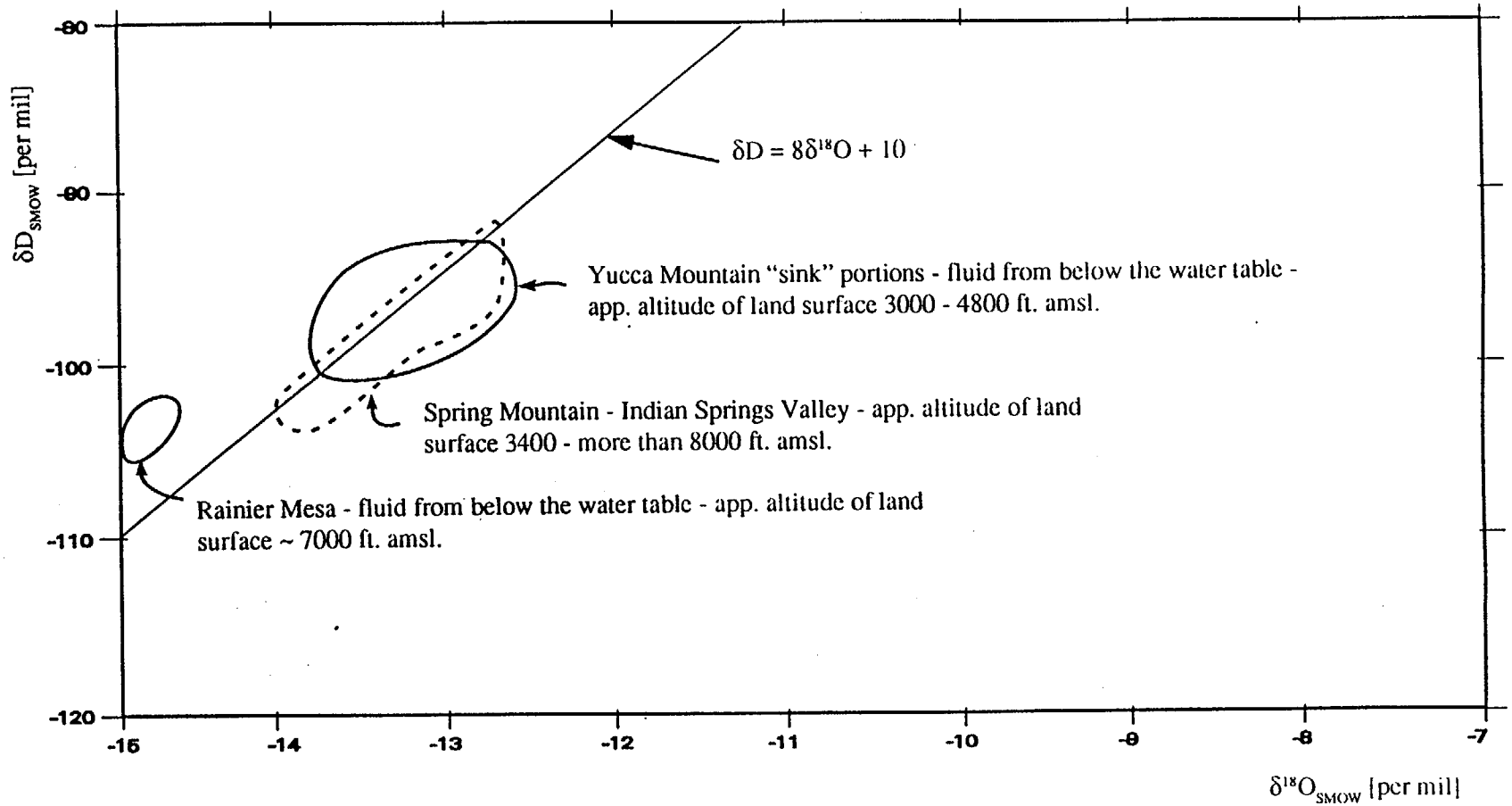
Note:

a) 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:

b) oxygen isotopic shift $\Delta\delta^{18}O = (\delta D_{ob} - 10)/8 - \delta^{18}O_{ob}$; and

c) 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.

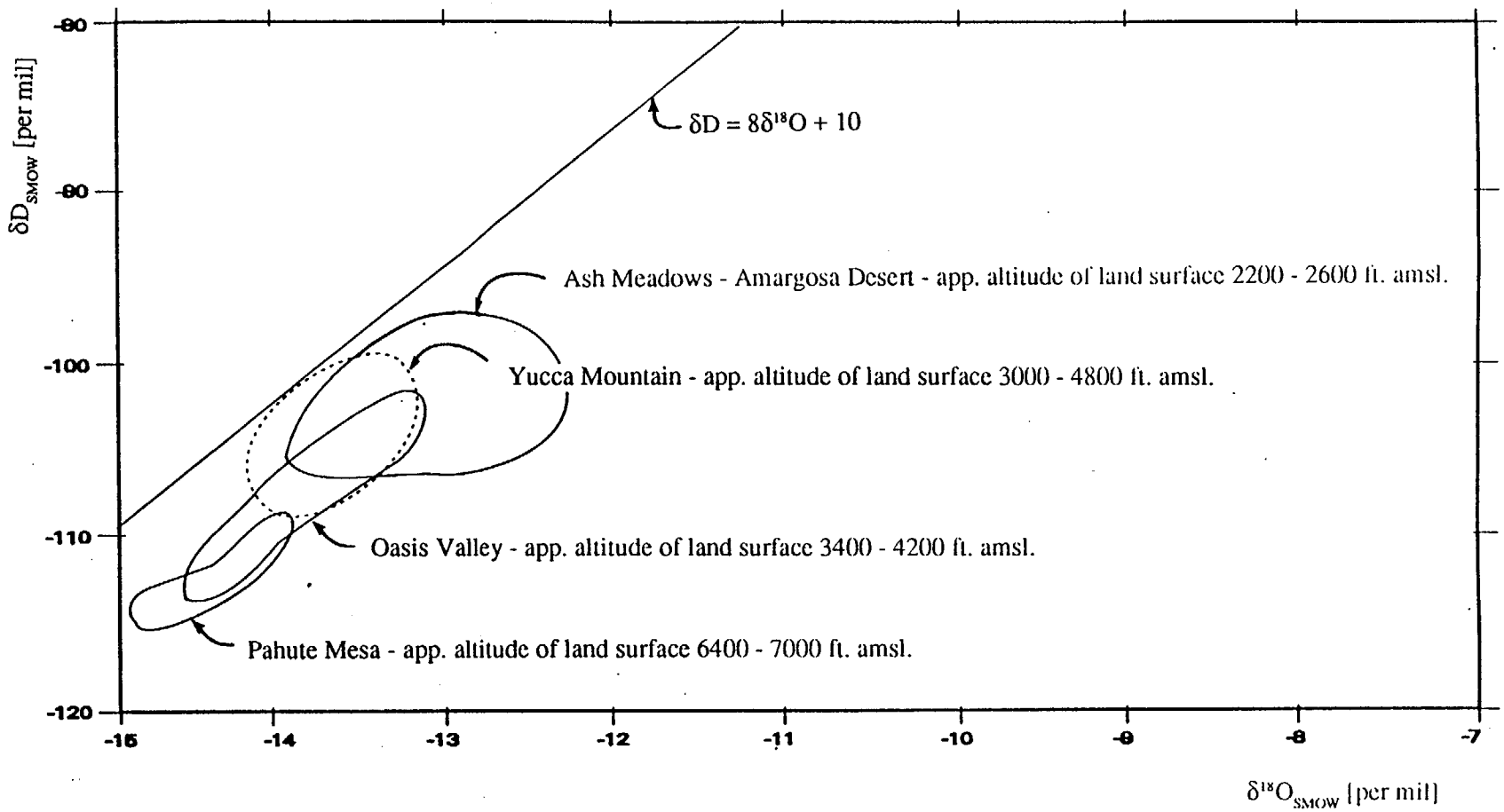


Note:

a) Spring Mountain fluids are contemporary fluids; and

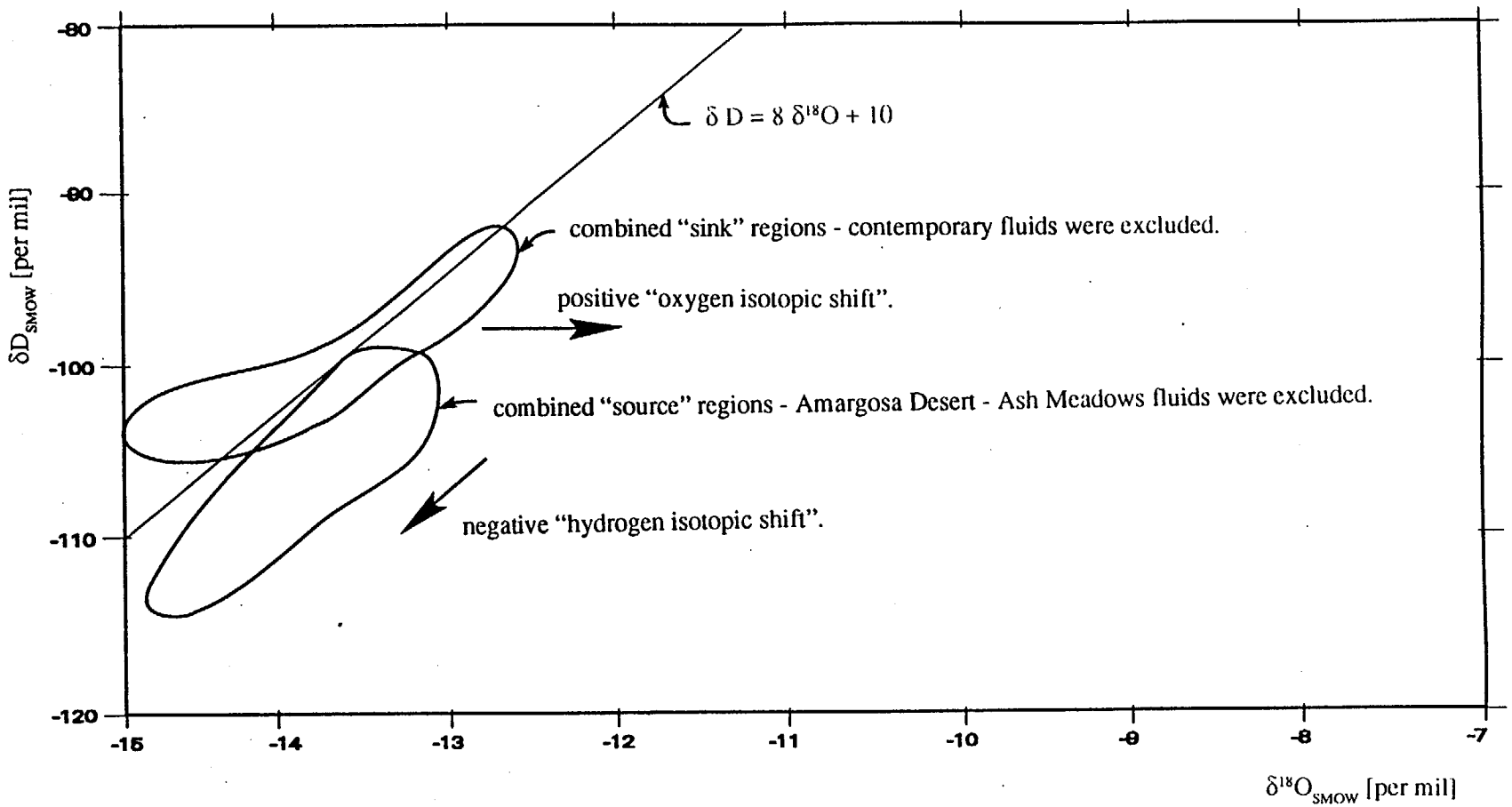
b) Yucca Mountain and Rainier Mesa fluids from below the water table are older fluids - ^{14}C ages are comparable to those of fluids from the "source" regions.

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.

Figure 4-4b



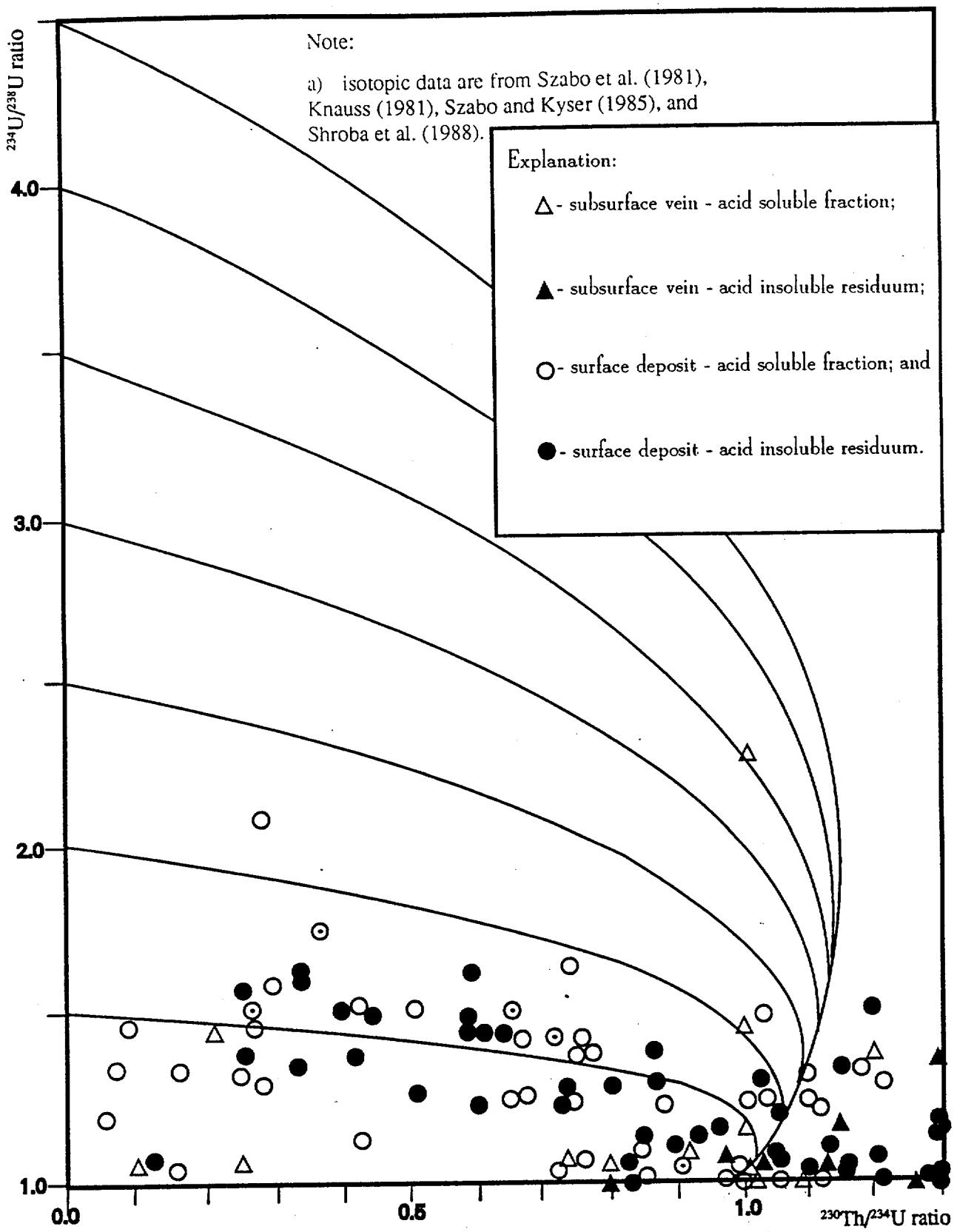
Note:

a) comparison is based on data from regions with comparable altitudes of the land surface; and

b) comparison is based on data from fluid samples with comparable PMC.

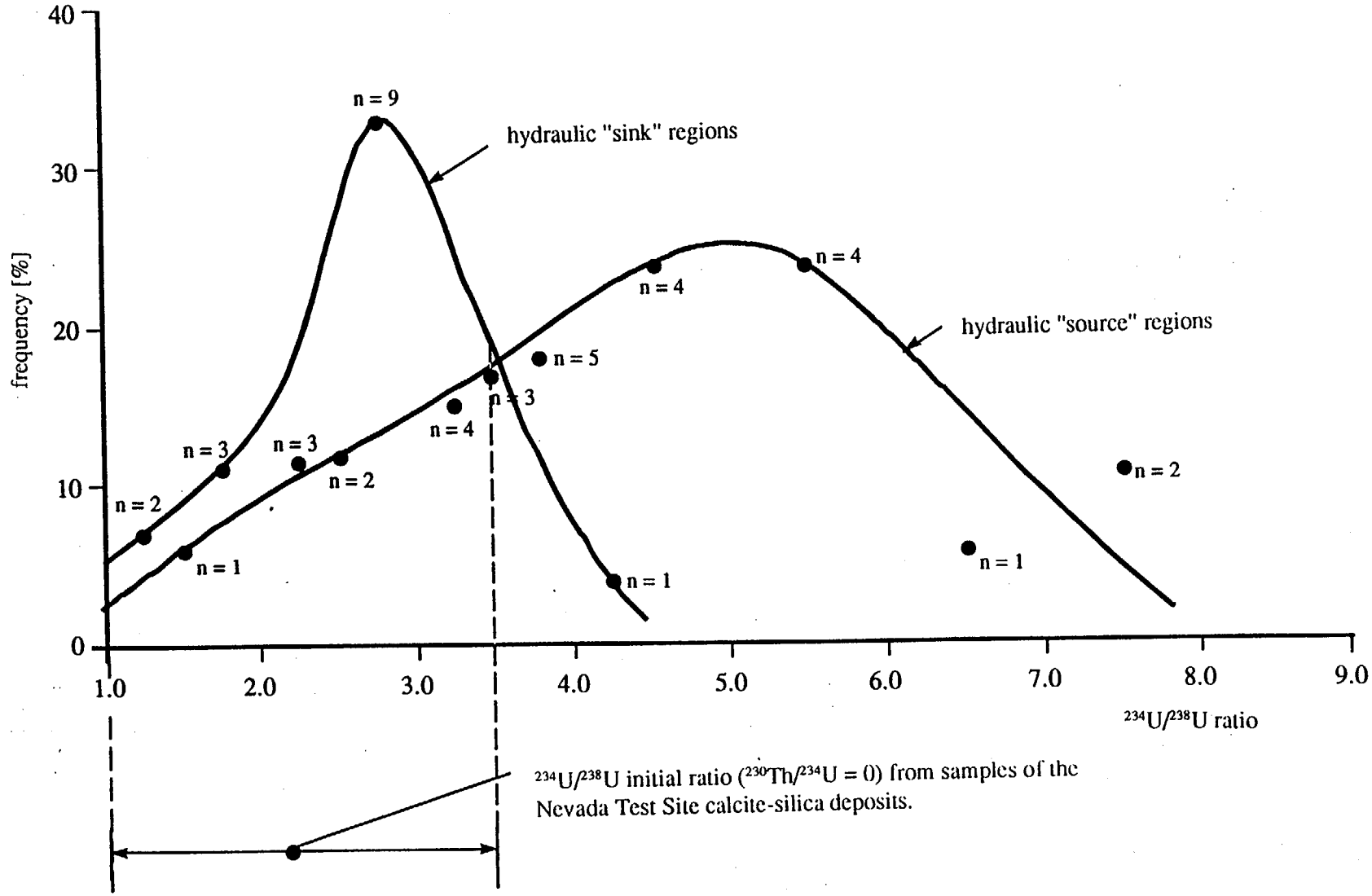
Comparison of isotopic compositions. "Sink" and "source" regions - The Nevada Test Site hydrosphere.

Figure 44c

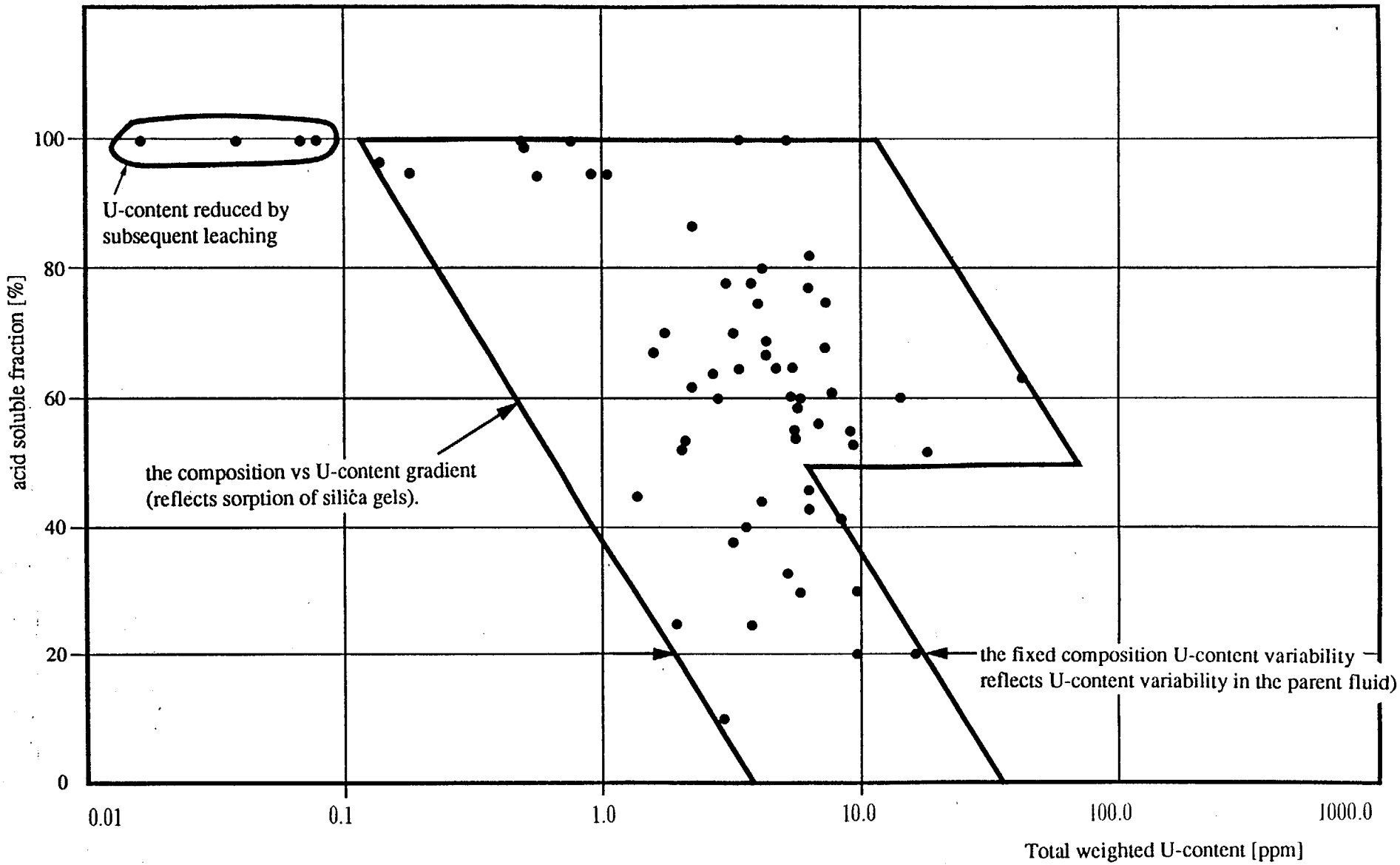


$^{234}U/^{238}U$ and $^{230}Th/^{234}U$ activity ratios. The Nevada Test Site calcite-silica deposits.

Figure 45

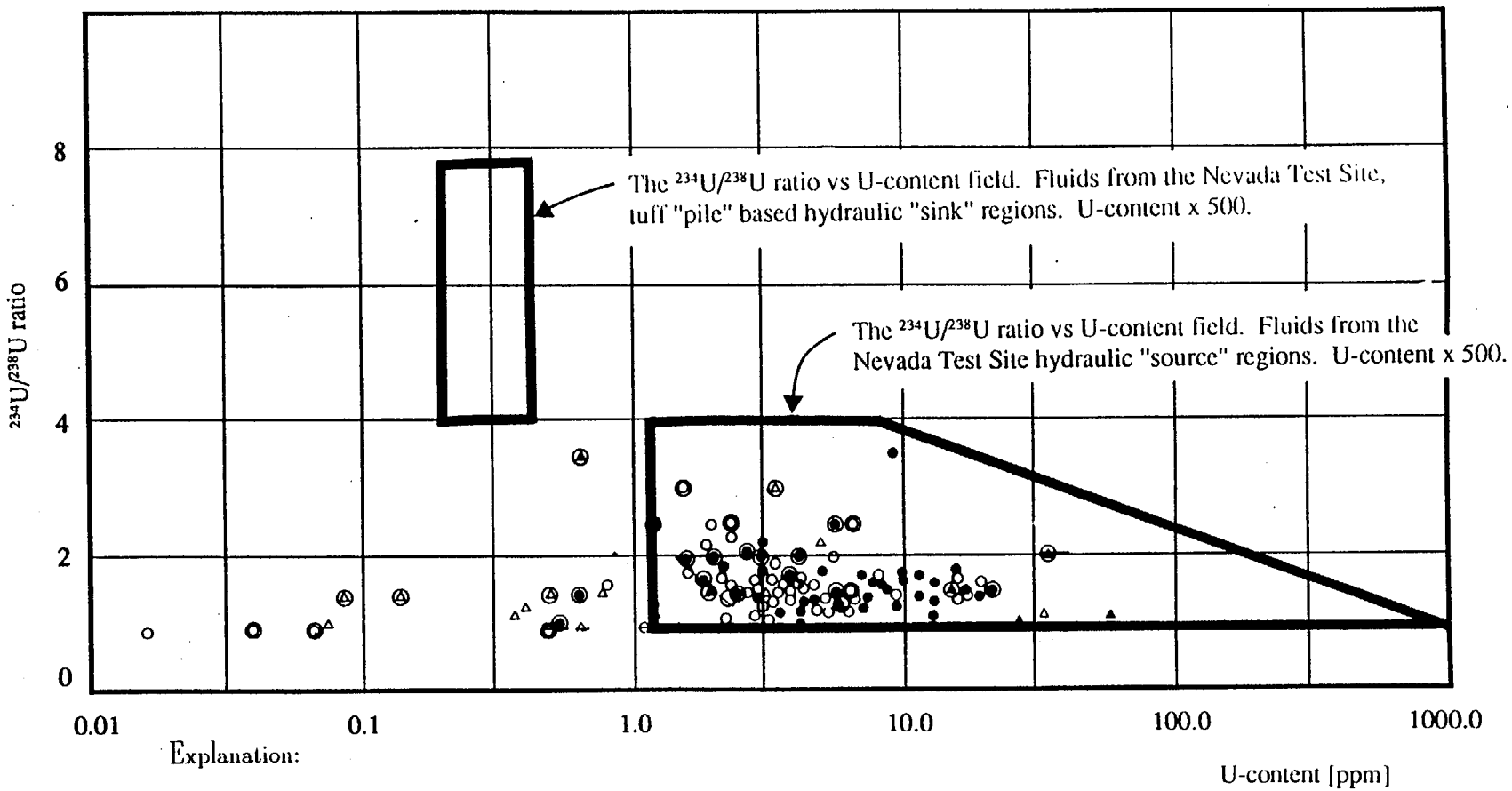


Comparison of the $^{234}U/^{238}U$ activity ratios. The Nevada Test Site calcite-silica deposits and the local geothermal fluids.



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

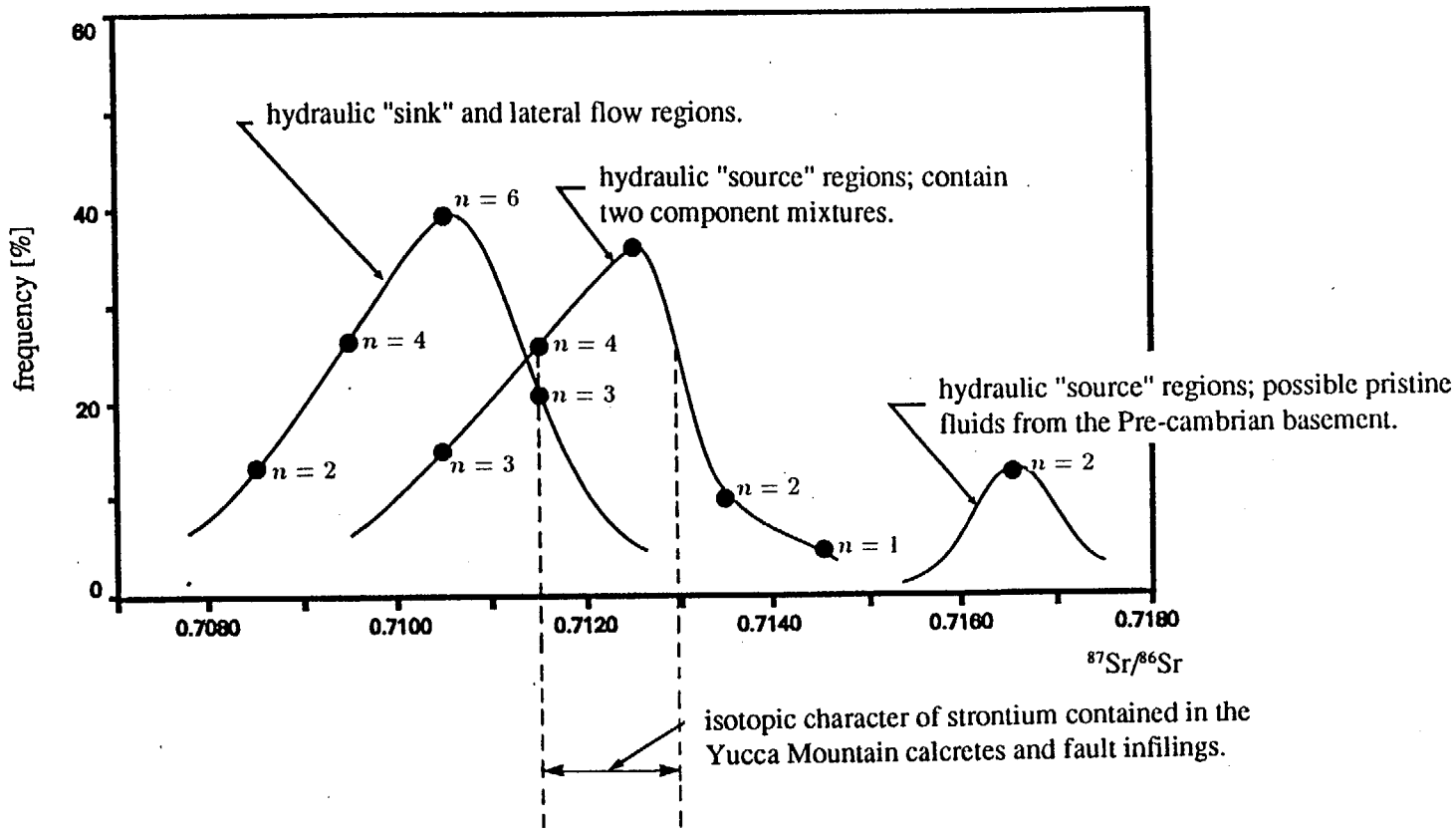
Figure 47



Explanation:

- △ - subsurface vein - acid soluble fraction;
- ▲ - subsurface vein - acid insoluble residuum;
- c - surficial deposit - acid soluble fraction;
- ● - surficial deposit - acid insoluble residuum;
- O - $^{234}U/^{238}U$ ratio plotted is a minimum value.

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

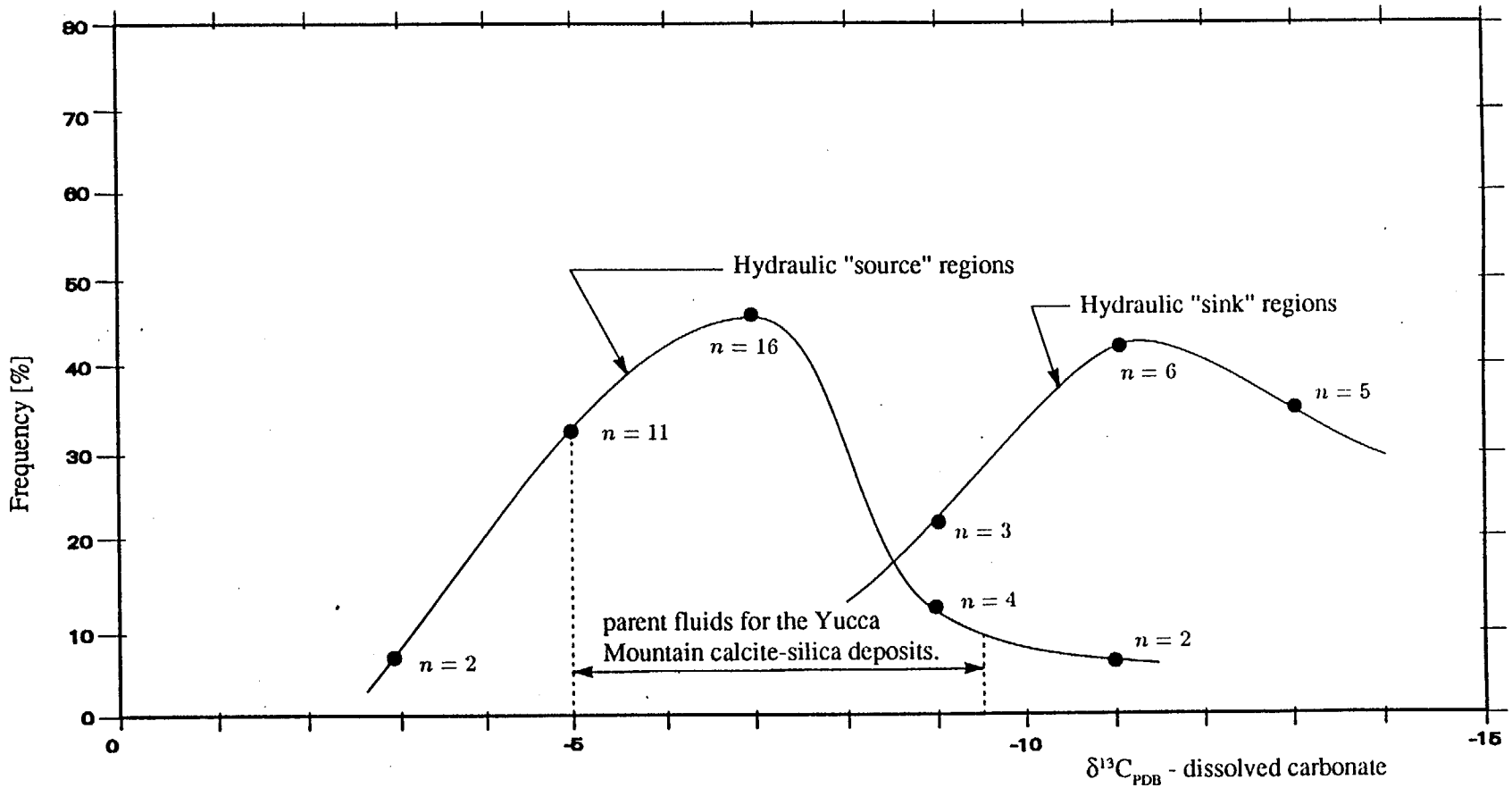


Note:

a) total number of samples n = 15 for the hydraulic "sink" regions, and n = 19 for the hydraulic "source" regions; and

b) isotopic data are from Marshall et al. (1990), Stuckless (1990), Peterman (1990), and Stuckless et al. (1990).

Comparison of the $^{87}Sr/^{86}Sr$ 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 $\delta^{13}C$ ratio. The Yucca Mountain calcite-silica deposit and the Nevada Test Site subsurface fluids.

- 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.

- 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.

- 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", if 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 CO_2 gas respired from the Yucca Mountain vadose zone and is dissolved in the Nevada Test Site hydrosphere.

Statement of the issue - topic #4.

Figure 52

- 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.

- 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.

- 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 $\delta^{13}C$ ratio is -7.0 per mil $_{PDB}$, Keeling (1961); ii) the biogenically derived CO_2 - mean value of the $\delta^{13}C$ 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 $\delta^{13}C$ 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.

Figure 53

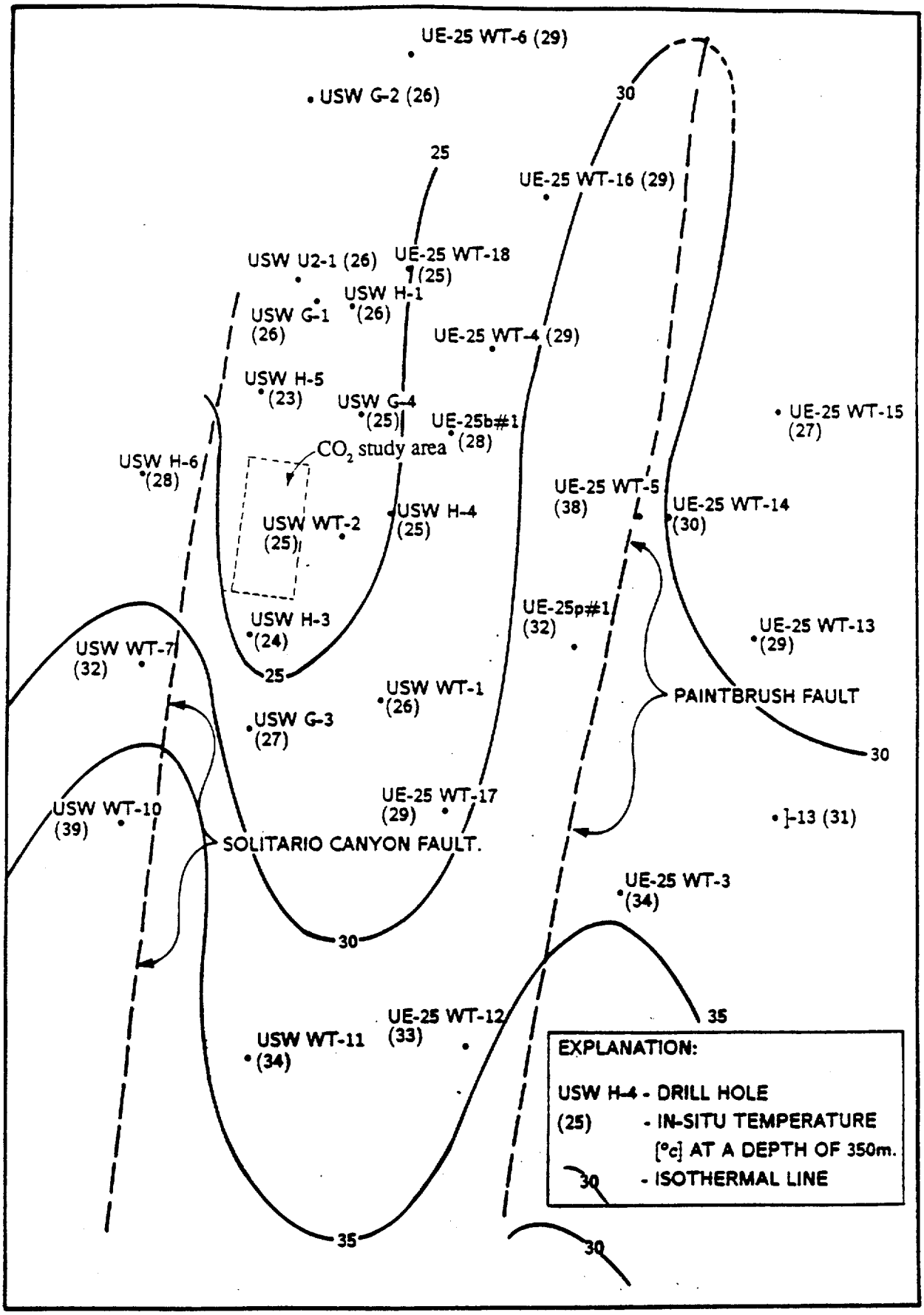
- Interpretations of the results of studies of spatio-temporal distribution of the carbon indexes ($\delta^{13}C$, PMC, and log P_{CO_2}), from samples of the Yucca Mountain vadose zone CO_2, must be made recognizing that, at the vadose zone base, there are very substantial lateral temperature gradients, $dT/dx \rightarrow 10°$Celsius per 1km.

- 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.

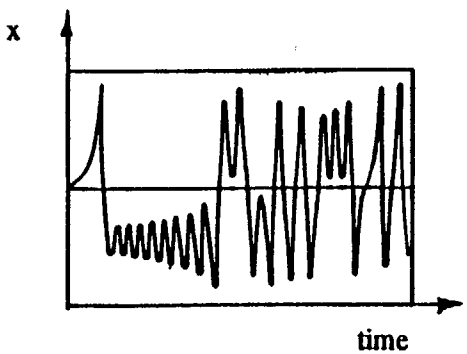
- Fluctuations involving both the barometric pressure and the atmospheric temperature virtually assure that the convective circulations occur as the non-periodic flow, Lorentz (1963).

- It may be expected, therefore, that $\sigma\phi_{xyz(t)} \neq O$ [for a given sampling site, $\sigma\phi_{xyz(t)}$ denotes amplitude of fluctuations involving either the $\delta^{13}C$ ratio, the PMC index, or the log P_{CO_2} index].

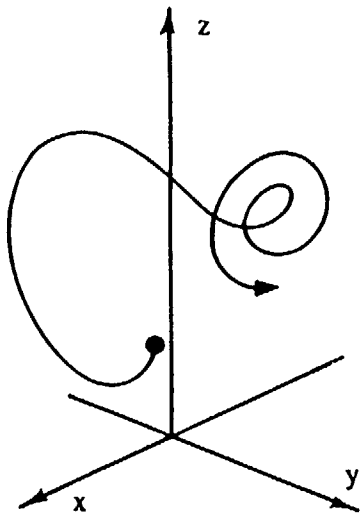
Introduction - interpretation of the results of studies of spatio-temporal distribution of the carbon indexes in the Yucca Mountain vadose zone.



Location of the CO_2 study area. Relative to *in-situ* temperatures at a depth of 350 m. The Yucca Mountain - vadose zone. From Szymanski, 1989.



a) time-series



b) phase-space representation

Note:

a) $dx/dt = -\sigma x + \sigma y$, $dy/dt = Rx - y - xz$, and $dz/dt = -bz + xy$ - Lorentz's equations from Kelis-Borok (1990);

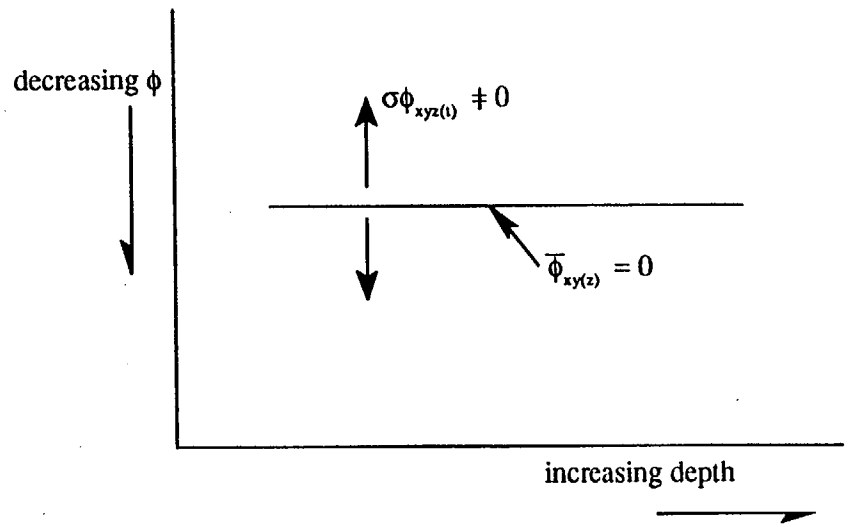
b) functions x, y, z characterize the intensity of convection stream, horizontal and vertical temperature gradients, respectively;

c) σ, b, and R are numerical parameters; and

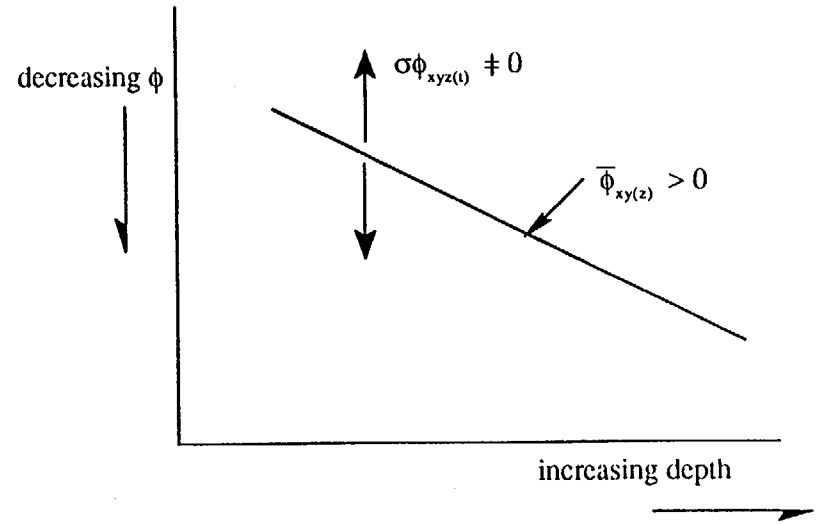
d) dx/dt controls values of the $\delta^{13}C$ ratio, the PMC index, and the partial pressure $\log P_{CO_2}$.

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

Figure 56



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



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

Explanation:

ϕ - denotes value of either the $|\delta^{13}C|$ ratio or the PMC index, $\overline{\phi}$ denotes mean value;

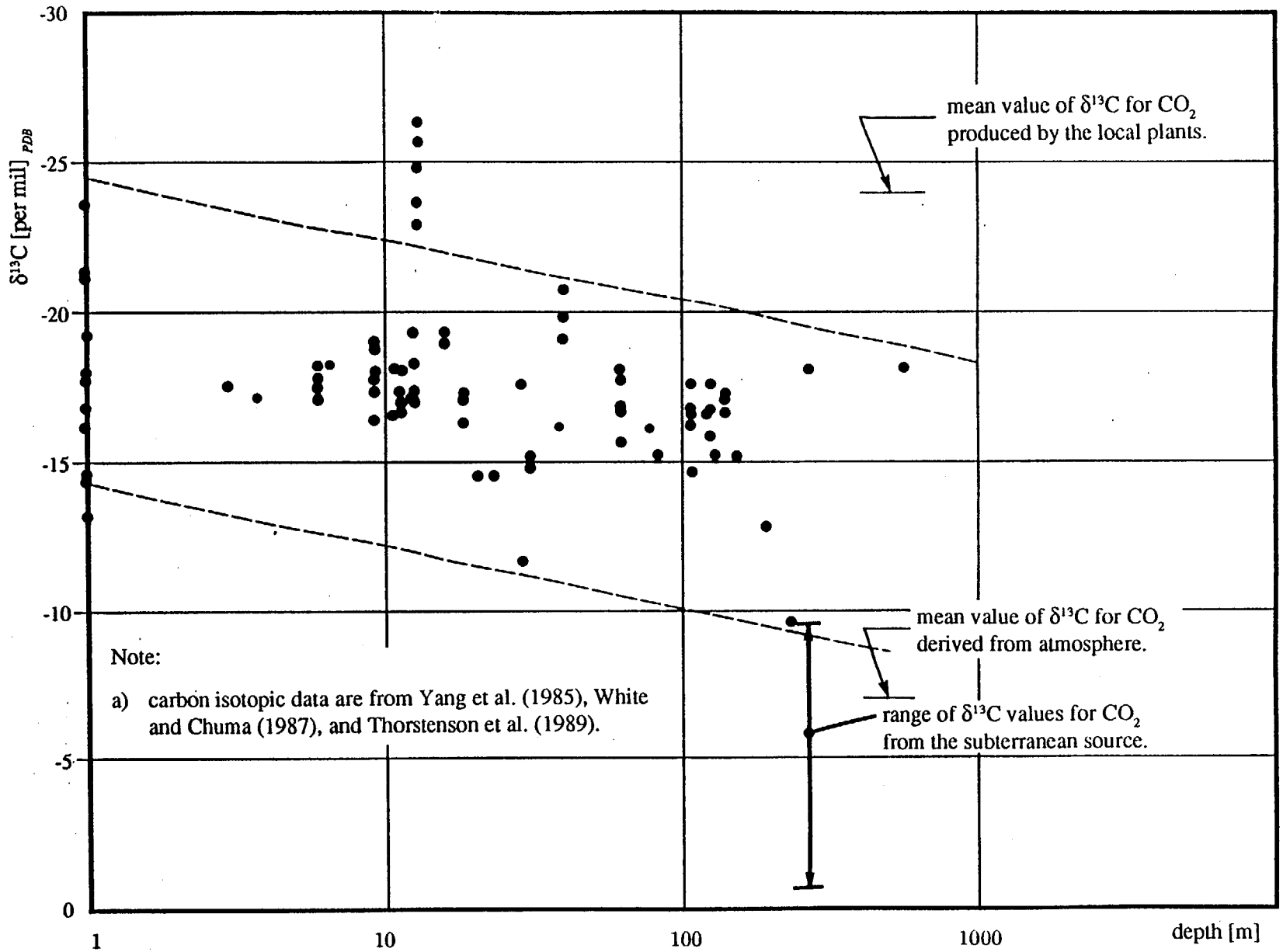
$\overline{\phi}_{xy(z)}$ - denotes $\overline{\phi}$ vs depth gradient, for a given vertical reference profile; and

$\sigma\phi_{xyz(t)}$ - denotes amplitude of ϕ fluctuations, for a given reference point.

Note:

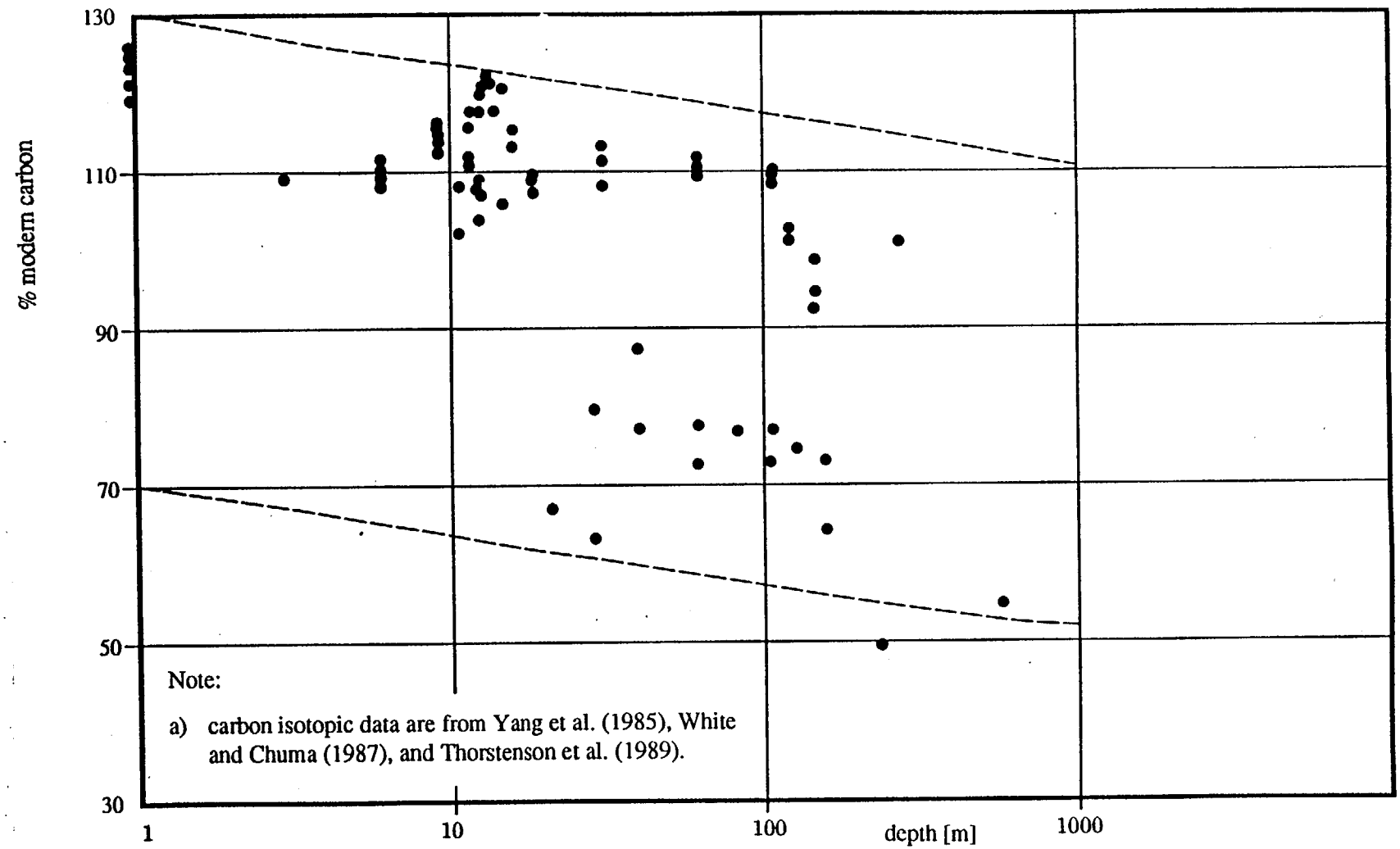
a) the subterranean source yields CO_2 that, relative to the atmosphere-biosphere derived CO_2, contains smaller concentrations of carbon-14 and carbon-12.

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



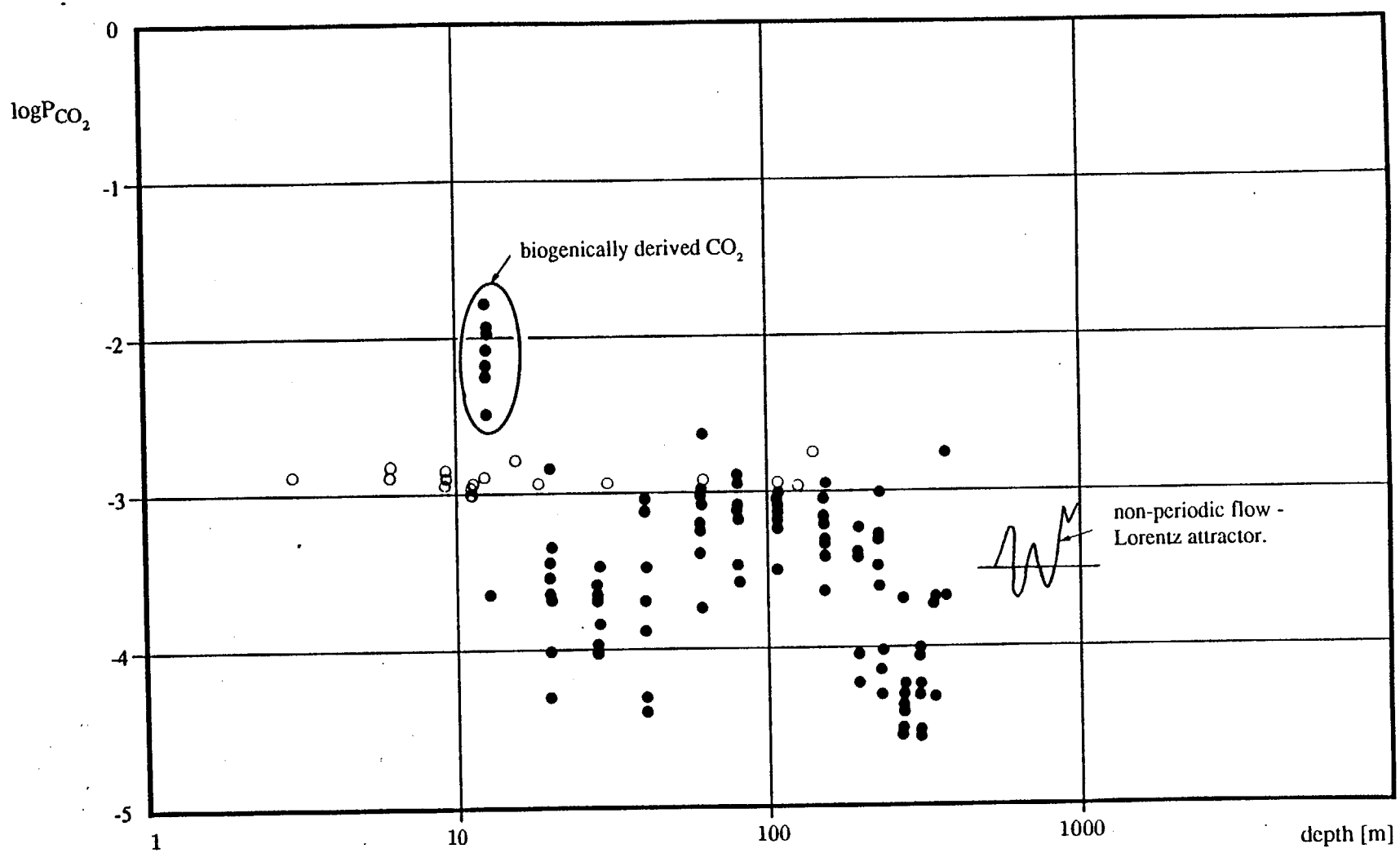
Isotopic character of carbon contained in the carbon dioxide from the vadose zone as a function of depth.

Figure 58



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

Figure 59

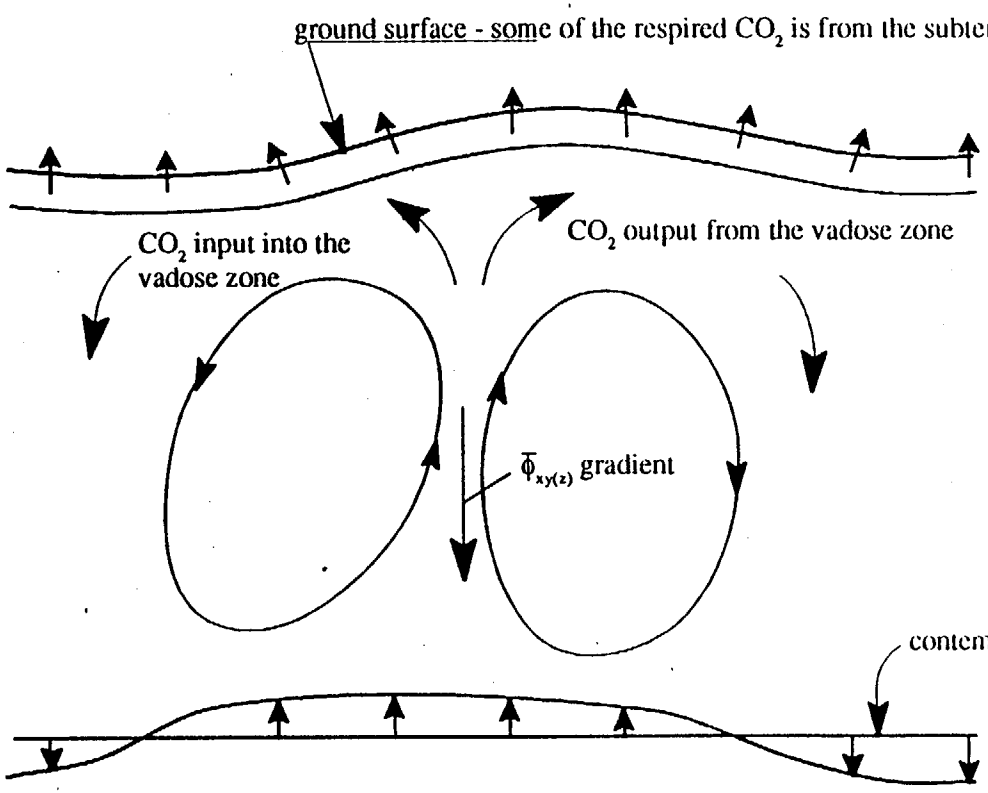


Explanation:

- ● 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.

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

Figure 60



The atmosphere-biosphere CO_2 source - value of the $\delta^{13}C$ ratio ranges from say -15 to -24 per mil $_{PDB}$, and PMC > 100 percent.

Non-periodic flow of gaseous phases - lacks spatio-temporal consistency.

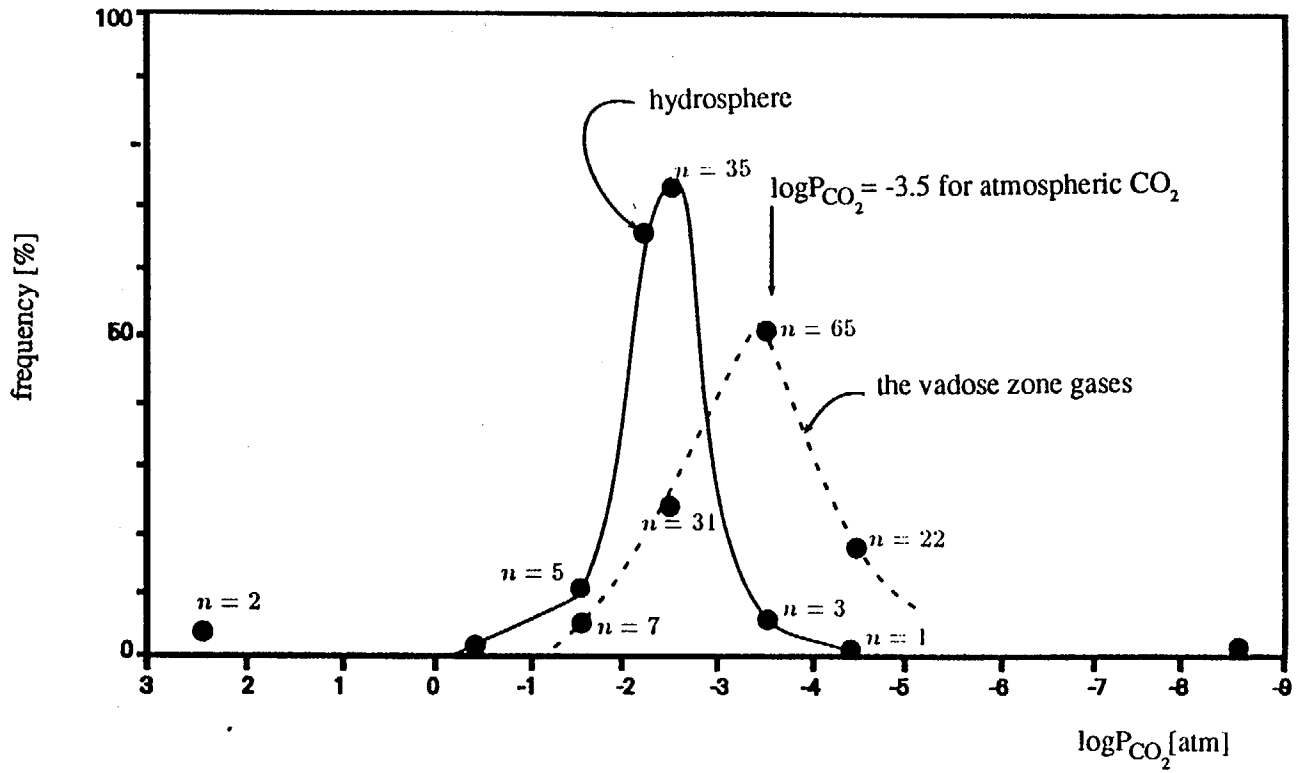
The subterranean CO_2 "source", in line with the CO_2 "sink" [for the subterranean CO_2 source - value of the $\delta^{13}C$ ratio may range from -1 to -9 per mil $_{PDB}$; and PMC $\longrightarrow$ 0 percent].

for the "source" region, dT/dx $\longrightarrow$ 10°C per 1km.

for the "sink" region, dT/dx $\longrightarrow$ 0°C per 1km.

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

Figure 61

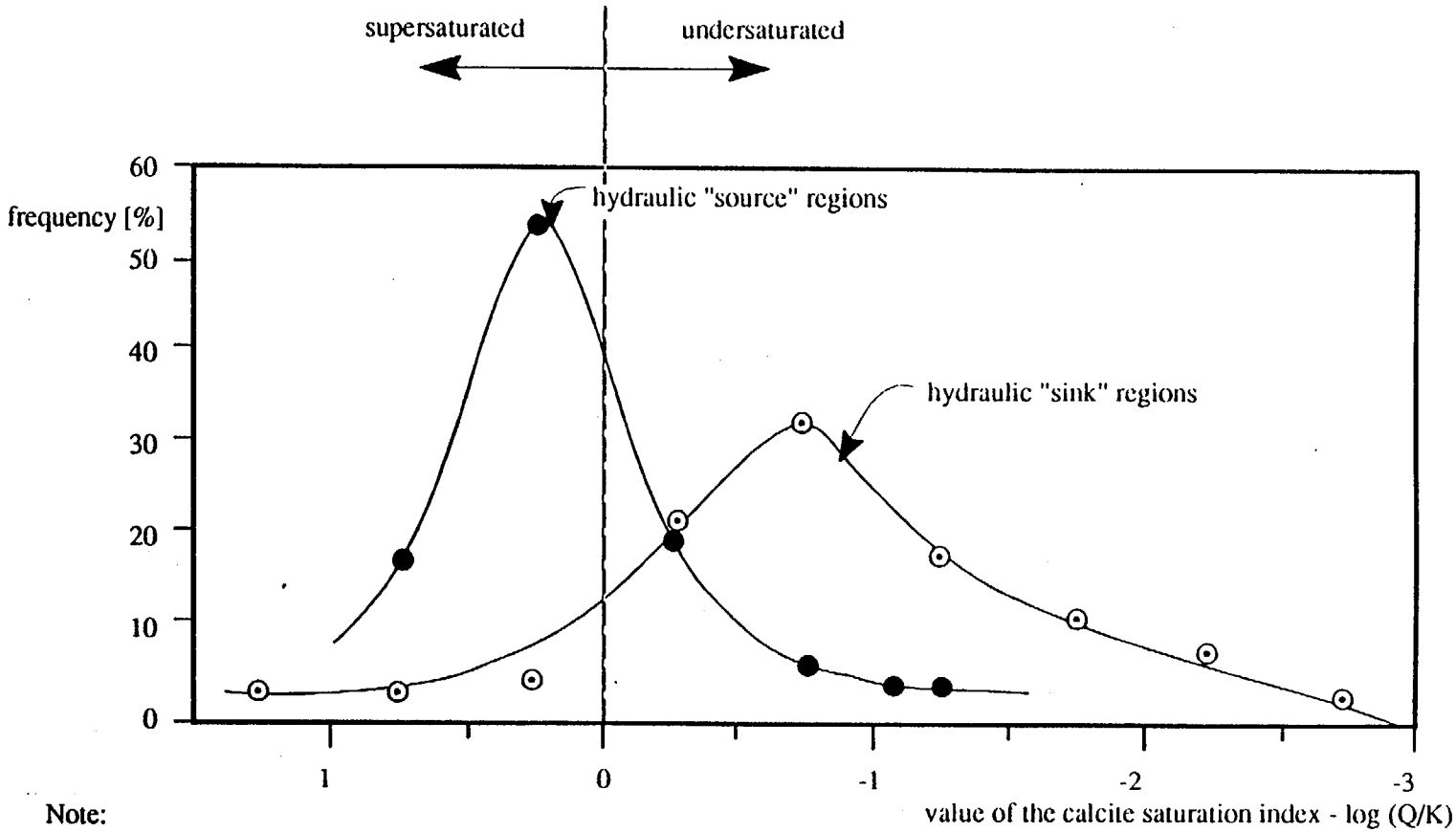


Note:

a) N = 47 samples for the hydrosphere, N = 125 for the vadose zone; and

b) $\log P_{CO_2}$ 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 $C0_2$. The Nevada Test Site hydrosphere.



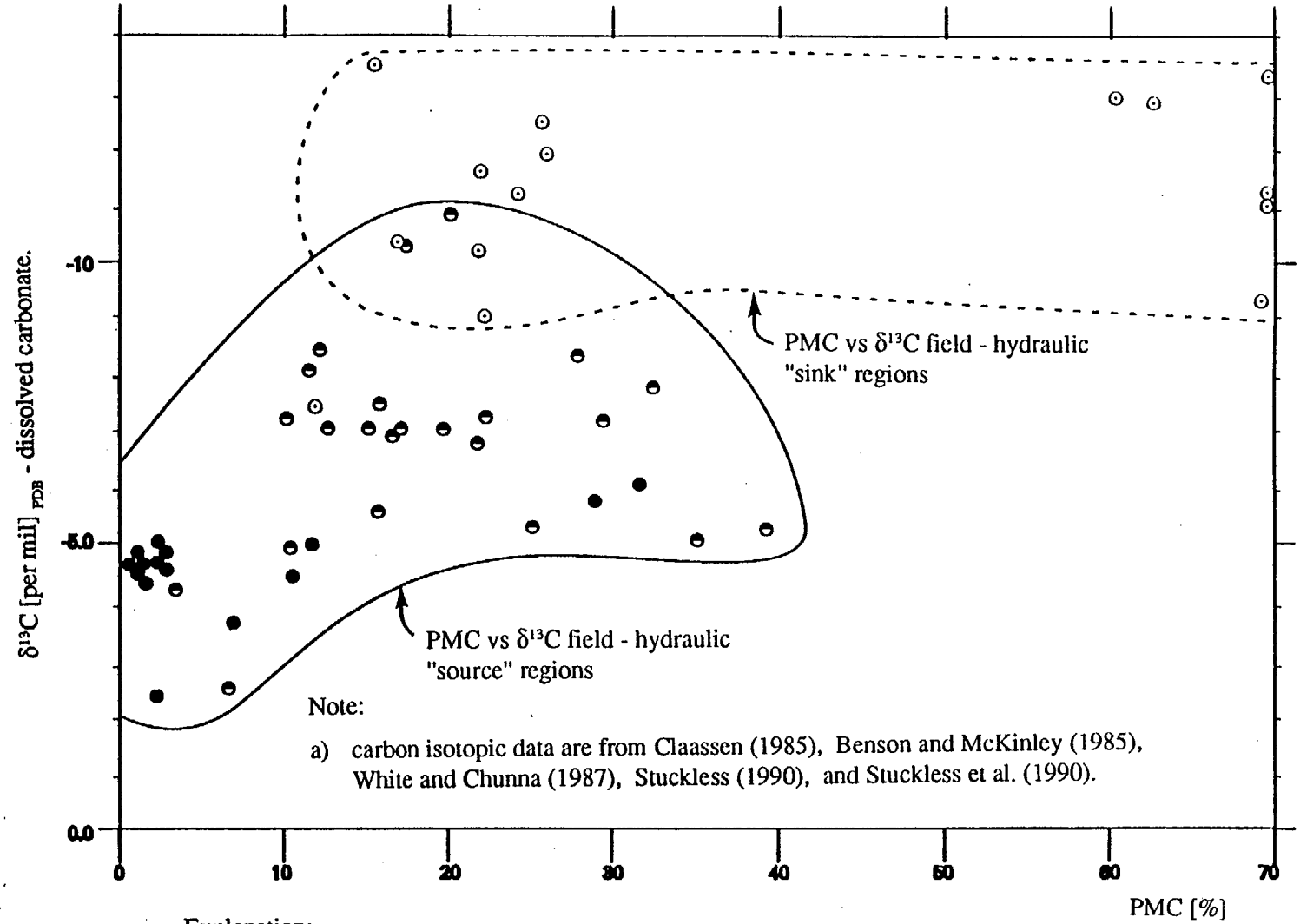
Note:

a) log (Q/K) data are from Kerrisk (1987);

b) N = 52 for hydraulic "source" regions; N = 64 for hydraulic "sink" regions; and

c) 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.

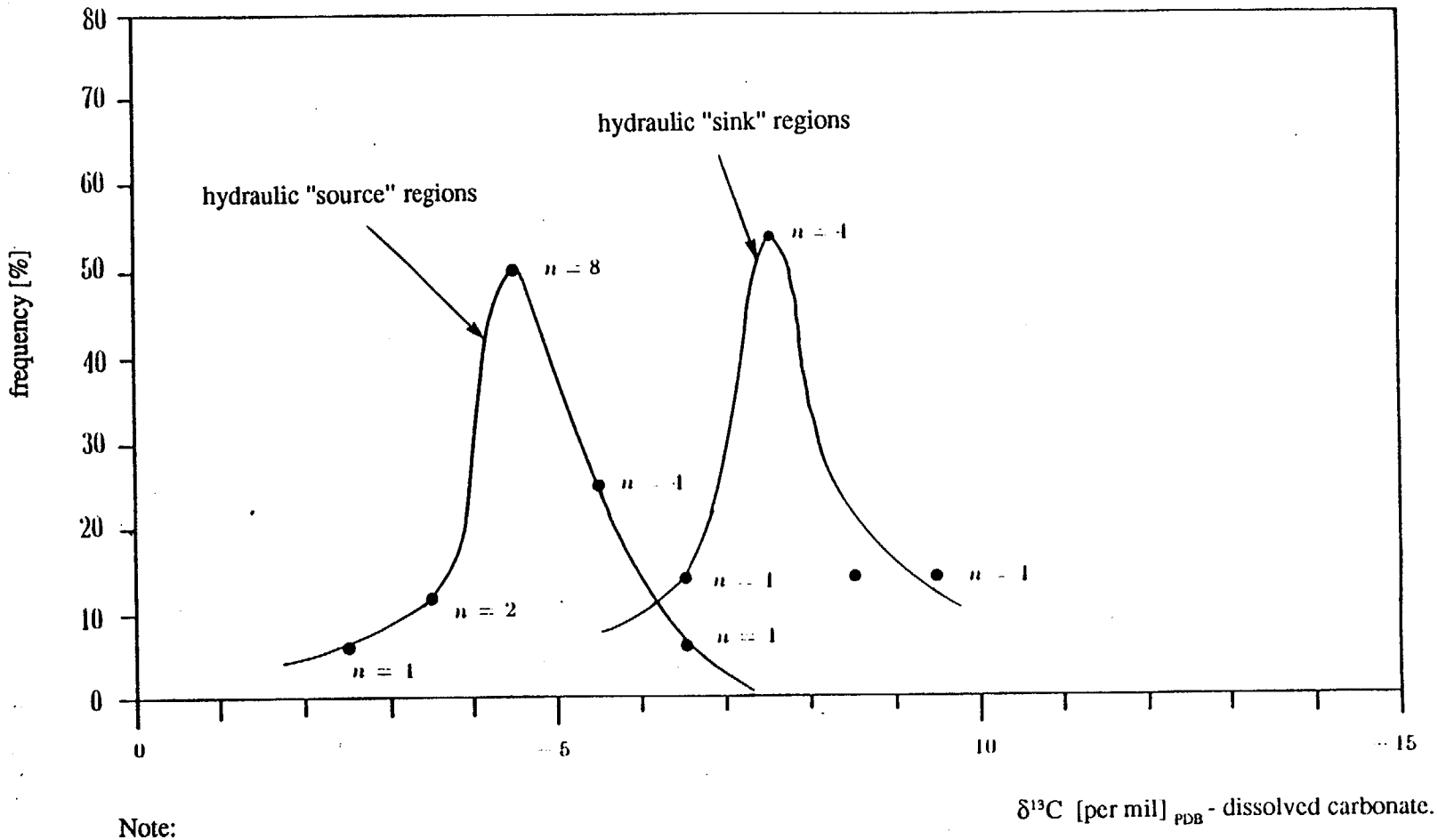


Explanation:

- ○ 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.

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

Figure 64

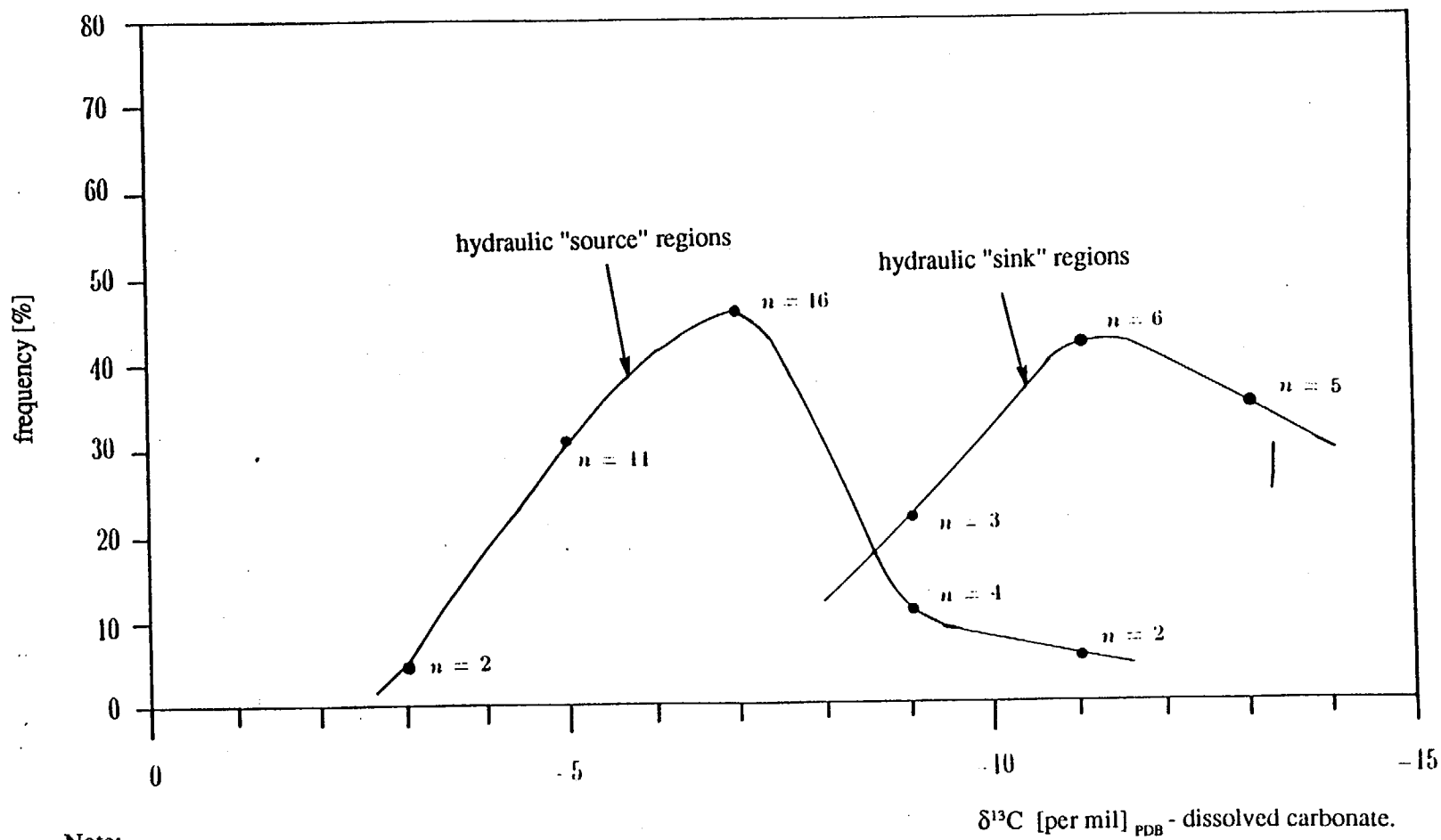


Note:

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

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

Figure 65

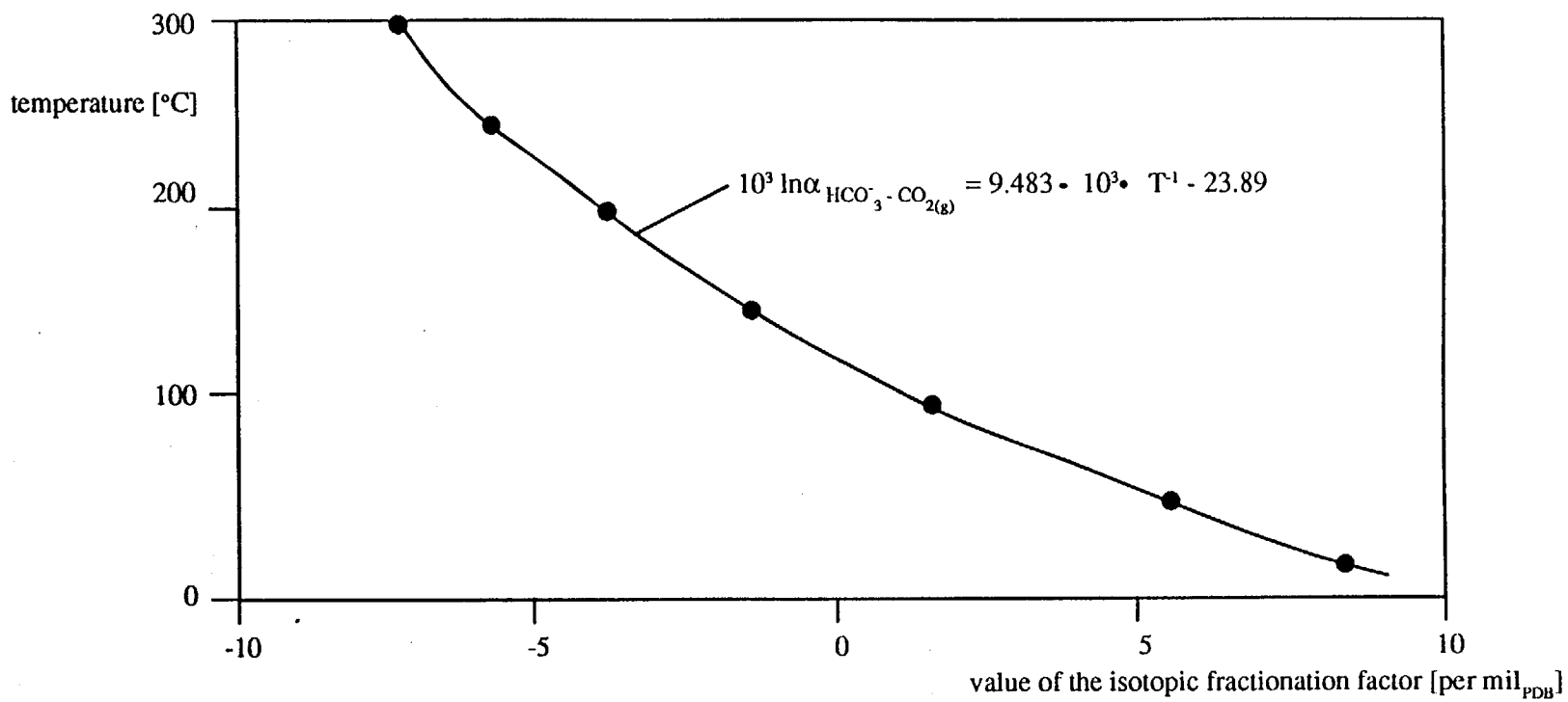


Note:

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.

Figure 66



Note:

a) the fractionation equation is from Turi (1986);

b) mean value of the $\delta^{13}C$ ratio, for the sub-lithospheric mantle indigenous CO_2, may be taken as ~-6 per mil $_{PDB}$, Hoefs (1987);

c) depending upon reaction temperature, dissolution of the sub-lithospheric mantle indigenous CO_2 may be expected to produce fluids with values of the $\delta^{13}C$ ratio ranging from + 1.45 (T = 20°Celsius) to -10 per mil $_{PDB}$ (T = 200°Celsius); and

d) the expected range of the $\delta^{13}C$ 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 $\delta^{13}C$ ratio, for the Paleozoic carbonates based "source" fluids, may be reflecting influx of the sub-lithospheric mantle indigenous CO_2.

Figure 67

- 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 $\delta^{13}C$ ratio ranging from -10 to about -2 per mil $_{PDB}$, Hoefs (1987);

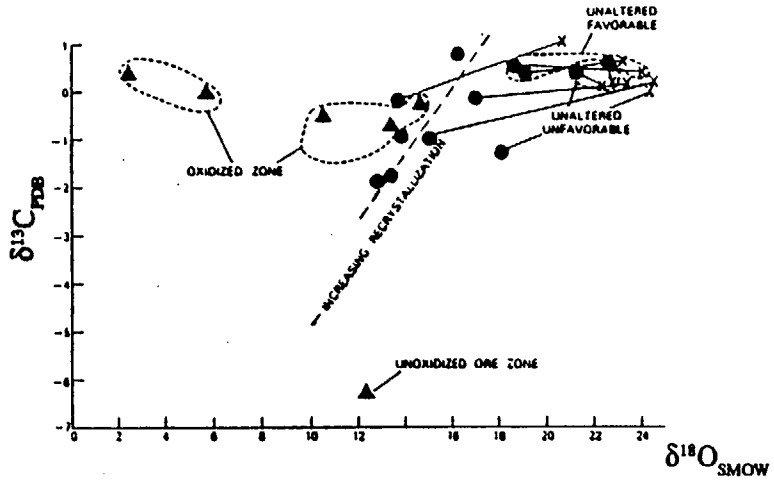
  ii) the hydrothermal ore forming fluids carry the dissolved carbon that is: a) isotopically "lighter" than carbon acquired during dissolution of marine limestones; and b) isotopically "heavier" than carbon produced by the biogenic sources;

  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 $\delta^{13}C$ ratio. [This very consistant 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

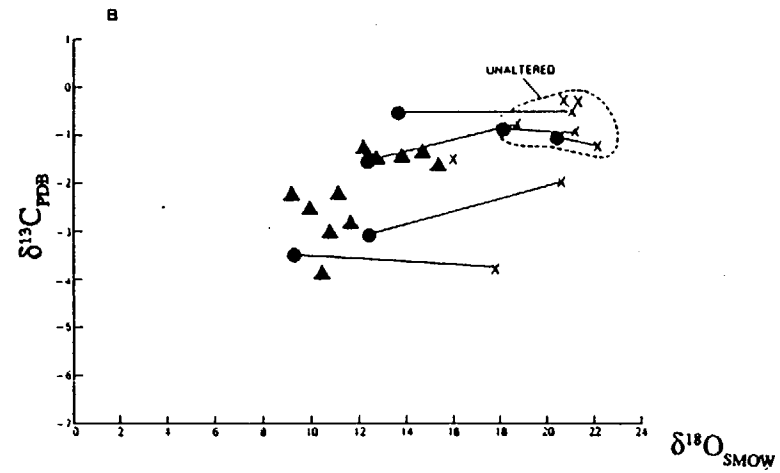
  iv) during vanishing stages of the ore forming periods, which presumably correspond with termination of igneous activity and periods with the accordingly reduced influx of the deep-seated CO_2, the hydrothermal fluids carry higher values of the $\delta^{13}C$ ratio [This observation may be taken to indicate 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 $\delta^{13}C$ ratio, for the Paleozoic carbonates based "source" fluids, may be reflecting influx of the sub-lithospheric mantle indigenous CO_2.

Figure 68



a) the Carlin disseminated gold deposit of Nevada



b) the Cortez disseminated gold deposit of Nevada

Note:

a) for both deposits host rock is the Paleozoic limestone;

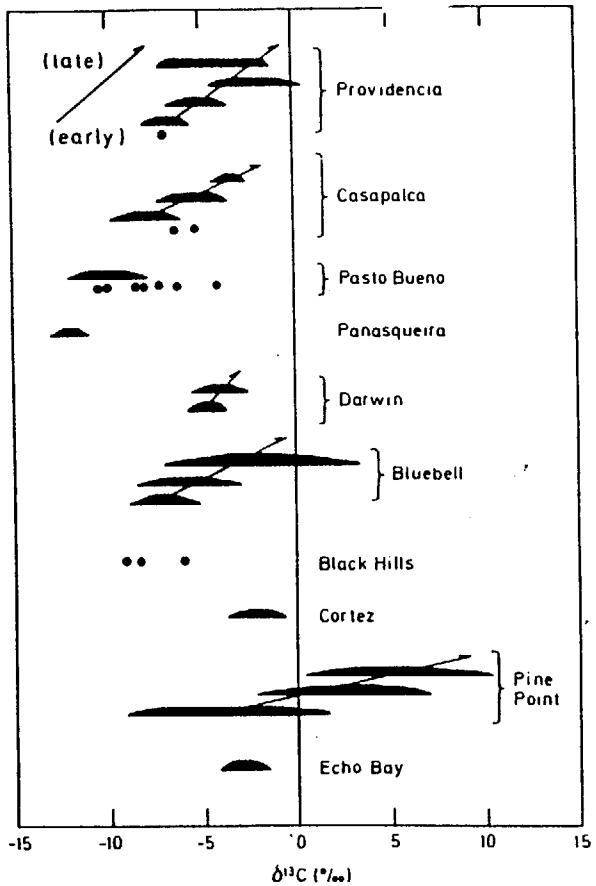
b) samples of the carbonate gangue minerals, from the Carlin and Cortez deposits, yield values of the $\delta^{13}C$ ratio ranging from +1 to -2 per mil $_{PDB}$ and from 0 to -4 per mil $_{PDB}$, respectively;

c) 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 $10^3 \ln\alpha_{HCO^-_3 - CO_{2(g)}} = \sim -3.0$ per mil $_{PDB}$, Hoefs (1987);

d) the parent fluids for the Carlin and Cortez deposits have carried values of the $\delta^{13}C$ ratio ranging from -2 to -5 per mil $_{PDB}$ and from -3 to -7 per mil $_{PDB}$, respectively; and

e) observation that the marine limestone based, and hydrothermal ore forming, fluids have carried the abnormally low values of the $\delta^{13}C$ 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:

a) relict fluids entrapped in fluid inclusions from samples of the carbonate gangue minerals, associated with various hydrothermal ore deposits, yield values of the $\delta^{13}C$ ratio ranging from -4 to about -10 per mil $_{PDB}$;

b) for individual ore deposits, the different generations of the carbonate gangue minerals are arranged from early to late, as shown by arrows;

c) 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

d) observation that the hydrothermal ore forming fluids carry values of the $\delta^{13}C$ 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 CO_2 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 CO_2 gas respired from the Yucca Mountain vadose zone and is dissolved in the Nevada Test Site hydrosphere.

Figure 71

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